## 2. ENERGY STORAGE TECHNOLOGIES

Energy storage technologies, especially batteries, are critical enabling technologies for the development of advanced, fuel-efficient, light- and heavy-duty vehicles, which are critical components of the U.S. Department of Energy's (DOE's) Energy Strategic Goal: "to protect our national and economic security by promoting a diverse supply and delivery of reliable, affordable, and environmentally sound energy." The program's vision supports the development of durable and affordable advanced batteries covering the full range of vehicle applications, from start/stop to full-power hybrid electric, electric, and fuel cell vehicles. Much of this work will transfer to energy storage for heavy hybrid vehicles as well. Energy storage research aims to overcome specific technical barriers that have been identified by the automotive industry together with the Vehicle Technologies Program. These include cost, performance, life, and abuse tolerance. These barriers are being addressed collaboratively by the DOE's technical research teams and battery manufacturers.

In this merit review activity, each reviewer was asked to respond to a series of questions, involving multiple-choice responses, expository responses where text comments were requested, and numeric score responses (*on a scale of 1 to 4*). In the pages that follow, the reviewer responses to each question for each project will be summarized: the multiple choice and numeric score questions will be presented in graph form for each project, and the expository text responses will be summarized in paragraph form for each question. A table presenting the average numeric score for each question for each project is presented below.

Presentation Title	Principal Investigator and Organization	Page Number	Approach	Technical Accomplishments	Collaborations	Future Research	Weighted Average
PHEV Battery Cost Assessment	Brian Barnett (TIAX LLC)	2-5	3.33	3.33	2.67	3.17	3.23
A High-Performance PHEV Battery Pack	Mohamed Alamgir (Compact Power)	2-7	2.80	2.60	3.20	2.00	2.65
HEV and PHEV USABC Battery Development Projects	Richard Holman (A123Systems)	2-9	3.20	2.80	2.60	2.60	2.85
USABC PHEV Battery Development Project	Cyrus Ashtiani (Enerdel)	2-11	3.20	2.40	3.20	2.40	2.70
JCS PHEV System Development-USABC	Scott Engstrom (Johnson Controls-Saft)	2-13	3.00	2.40	3.20	2.60	2.68
Advanced Cathode Material Development for PHEV Lithium Ion Batteries	Jamie Gardner (3M)	2-15	3.50	3.50	2.00	3.25	3.28
USABC Battery Separator Development	Ron Smith (Celgard)	2-17	1.80	1.20	1.00	1.20	1.33
Multifunctional, Inorganic-Filled Separators for Large Format, Li- ion Batteries	Richard Pekala (Entek)	2-19	3.80	3.40	2.00	3.00	3.28
Hybrid Nano Carbon Fiber/Graphene Platelet-Based High-Capacity Anodes for Lithium Ion Batteries	Bor Jang (Angstron Materials)	2-21	3.40	3.20	2.80	3.40	3.23
New High-Energy Nanofiber Anode Materials	Xiangwu Zhang (NC State/NLE)	2-24	3.20	3.00	2.60	3.00	3.00
Stabilized Lithium Metal Powder, Enabling Material and Revolutionary Technology for High Energy Li-ion Batteries	Marina Yakovleva (FMC)	2-27	2.60	2.80	2.20	2.80	2.68
Protection of Li Anodes Using Dual Phase Electrolytes	Yuriy Mikhaylik (Sion Power)	2-30	2.80	3.00	3.00	2.60	2.90
Process for Low Cost Domestic Production of LIB Cathode Materials	Anthony Thurston (BASF)	2-33	2.40	2.40	2.40	2.80	2.45
Overview of Applied Battery Research	Gary Henriksen (Argonne National Laboratory)	2-36	3.00	3.17	3.50	3.17	3.17
Engineering of High Energy Cathode Material	Khalil Amine (Argonne National Laboratory)	2-39	3.33	3.33	3.20	3.17	3.30
New High Energy Gradient Concentration Cathode Material	Khalil Amine (Argonne National Laboratory)	2-42	3.00	3.00	2.50	3.17	2.96

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Presentation Title	Principal Investigator and Organization	Page Number	Approach	Technical Accomplishments	Collaborations	Future Research	Weighted Average
Design and Evaluation of Novel High Capacity Cathode Materials	Christopher Johnson (Argonne National Laboratory)	2-45	2.86	2.86	2.86	2.86	2.86
Search for High Energy Density Cathode Materials	llias Belharouak (Argonne National Laboratory)	2-48	3.00	2.57	2.57	3.00	2.73
Development of High-Capacity Cathode Materials with Integrated Structures	Sun-Ho Kang (Argonne National Laboratory)	2-50	3.00	3.00	3.00	2.86	2.98
Developing High Capacity, Long Life, and High Power Anodes	Khalil Amine (Argonne National Laboratory)	2-53	2.25	2.50	3.00	2.75	2.53
Lithium Metal Anodes	Jack Vaughey (Argonne National Laboratory)	2-55	2.75	2.75	2.75	2.75	2.75
Improved Methods for Making Intermetallic Anodes	Andrew Jansen (Argonne National Laboratory)	2-57	2.60	2.00	2.20	2.25	2.21
Novel Electrolytes and Additives	Dan Abraham (Argonne National Laboratory)	2-59	2.20	2.40	3.00	2.40	2.43
Electrolytes in Support of 5 V Li- ion Chemistries	Richard Jow (Army Research Laboratory)	2-61	3.20	2.80	2.60	3.00	2.90
Development of Advanced Electrolytes and Electrolyte Additives	Zhengcheng Zhang (Argonne National Laboratory)	2-63	2.75	3.00	2.75	2.75	2.88
Development of Novel Electrolytes for Use in High Energy Lithium-Ion Batteries with Wide Operating Temperature Range	Marshall Smart (Jet Propulsion Laboratory)	2-65	3.50	3.00	4.00	3.00	3.25
Novel Compounds for Enhancing Electrolyte Stability and Safety of Lithium-ion Cells	Kevin Gering (Idaho National Laboratory)	2-67	2.60	2.60	3.20	3.00	2.73
Screen Electrode Materials & Cell Chemistries and Streamlining Optimization of Electrode	Wenquan Lu (Argonne National Laboratory)	2-70	3.33	3.33	3.17	2.83	3.25
Materials Scale-up and Cell Performance Analysis	Vince Battaglia (Lawrence Berkeley National Laboratory)	2-73	3.00	2.83	3.50	2.83	2.96
Fabricate PHEV Type Cells for Testing & Diagnostics	Andrew Jansen (Argonne National Laboratory)	2-76	3.60	2.80	3.20	3.25	3.11
Electrochemistry Cell Model	Dennis Dees (Argonne National Laboratory)	2-79	3.50	3.50	2.75	3.33	3.39
Diagnostic Studies - Argonne	Dan Abraham (Argonne National Laboratory)	2-81	3.40	3.20	3.60	3.00	3.28
Electrochemistry Diagnostics at LBNL	Frank McLarnon (Lawrence Berkeley National Laboratory)	2-83	3.00	3.00	2.80	2.40	2.90
Diagnostic Studies to Improve Abuse Tolerance and Life of Li- ion Batteries	Xiao-Qing Yang (Brookhaven National Laboratory)	2-86	3.80	3.60	3.80	3.80	3.70
Develop and Evaluate Materials and Additives that Enhance Thermal and Overcharge Abuse	Khalil Amine (Argonne National Laboratory)	2-89	3.00	3.20	3.40	2.67	3.11
Abuse Tolerance Improvement	Peter Roth (Sandia National Laboratories)	2-92	3.50	3.17	3.00	3.33	3.25
Overcharge Protection	Thomas Richardson (Lawrence Berkeley National Laboratory)	2-95	3.67	3.17	2.83	3.00	3.23
High Energy Density Ultracapacitors	Patricia Smith (Naval Surface Warfare Center)	2-98	2.67	2.33	3.00	3.33	2.63
In Situ Characterization of Fatigue Behavior of Electrodes	Claus Daniel (Oak Ridge National Laboratory)	2-100	3.20	2.80	2.40	3.00	2.88

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Presentation Title	Principal Investigator and Organization	Page Number	Approach	Technical Accomplishments	Collaborations	Future Research	Weighted Average
Low Cost SiOx-Graphite and Olivine Materials	Karim Zaghib (Hydro- Quebec)	2-102	2.86	2.86	3.29	3.14	2.95
Cathodes	Michael Thackeray (Argonne National Laboratory)	2-105	3.38	3.25	3.25	3.13	3.27
The Synthesis and Characterization of Substituted Olivines and Layered Manganese Oxides	M. Stanley Whittingham (SUNY-Binghamton)	2-107	3.00	2.71	3.33	2.67	2.86
Stabilized Spinels and Polyanion Cathodes	Arumugam Manthiram (University of Texas at Austin)	2-109	3.00	2.83	2.50	2.83	2.83
Olivines and Substituted Layered Materials	Marca Doeff (Lawrence Berkeley National Laboratory)	2-111	2.86	2.86	3.57	2.86	2.95
Cell Analysis – High-Energy Density Cathodes and Anodes	Thomas Richardson (Lawrence Berkeley National Laboratory)	2-113	3.43	3.29	3.57	3.00	3.32
First Principles Calculations of Electrode Materials	Gerbrand Ceder (Massachusetts Institute of Technology)	2-115	3.50	3.50	3.00	3.25	3.41
First Principles Calculations and NMR Spectroscopy of Electrode Materials: NMR	Clare Grey (SUNY- Stony Brook)	2-117	3.20	3.40	3.40	3.00	3.30
Development of High Energy Cathode for Li-ion Batteries	Jason Zhang (Pacific Northwest National Laboratory)	2-119	3.17	3.33	2.83	3.00	3.19
Inexpensive, Nonfluorinated Anions for Lithium Salts and Ionic Liquids for Lithium Battery Electrolytes	Wesley Henderson (North Carolina State University)	2-121	2.71	2.57	2.86	2.71	2.66
Molecular Dynamics Simulation Studies of Electrolytes and Electrolyte/Electrode Interfaces	Grant Smith (University of Utah)	2-123	2.57	2.57	3.00	2.57	2.63
In Situ Characterizations of New Battery Materials and the Studies of High Energy Density Li-Air Batteries	Xiao-Qing Yang (Brookhaven National Laboratory)	2-125	3.40	3.33	3.83	3.00	3.37
Solid Electrolyte Batteries	John Goodenough (University of Texas at Austin)	2-127	2.86	2.71	1.57	2.71	2.61
Nano-scale Composite Hetero- structures: Novel High Capacity Reversible Anodes for Lithium- ion Batteries	Prashant Kumta (University of Pittsburgh)	2-129	3.40	3.20	3.00	3.20	3.23
Intermetallic Anodes	Michael Thackeray (Argonne National Laboratory)	2-131	3.00	2.86	2.71	3.00	2.89
Nanostructured Materials as Anodes	M. Stanley Whittingham (SUNY-Binghamton)	2-133	2.75	2.50	2.88	2.50	2.61
Nanostructured Metal Oxide Anodes	Anne Dillon (National Renewable Energy Laboratory)	2-135	2.86	3.00	3.67	3.00	3.05
Development of High Capacity Anode for Li-ion Batteries	Jason Zhang (Pacific Northwest National Laboratory)	2-137	3.20	3.60	3.20	3.00	3.38
Electrolytes - Advanced Electrolyte and Electrolyte Additives	Khalil Amine (Argonne National Laboratory)	2-139	3.00	2.80	3.00	3.00	2.90
Development of Electrolytes for Lithium-ion Batteries	Brett Lucht (University of Rhode Island)	2-141	3.00	3.17	3.33	2.83	3.10
Bifunctional Electrolytes for Lithium-ion Batteries	Daniel Scherson (Case Westerm Reserve University)	2-143	3.00	2.60	2.60	2.80	2.73

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Presentation Title	Principal Investigator and Organization	Page Number	Approach	Technical Accomplishments	Collaborations	Future Research	Weighted Average
Performance and Safety of Olivines and Layered Oxides	Guoying Chen (Lawrence Berkeley National Laboratory)	2-145	3.00	3.00	3.33	2.83	3.02
Positive and Negative Electrodes: Novel and Optimized Materials	Jordi Cabana (Lawrence Berkeley National Laboratory)	2-147	2.86	2.86	3.14	2.86	2.89
Electrode Fabrication and Failure Analysis	Vince Battaglia (Lawrence Berkeley National Laboratory)	2-149	3.20	3.00	3.20	3.00	3.08
Microscale Electrode Design Using Coupled Kinetic, Thermal and Mechanical Modeling	Ann Marie Sastry (University of Michigan)	2-151	3.00	3.17	2.50	2.83	3.00
Analysis and Simulation of Electrochemical Energy Systems	John Newman (University of California at Berkeley)	2-153	3.00	2.80	3.20	3.00	2.93
The Role of Surface Chemistry and Bulk Properties on the Cycling and Rate Capability of Lithium Positive Electrode Materials	Yang Shao-Horn (Massachusetts Institute of Technology)	2-155	3.20	3.40	3.00	3.20	3.28
Interfacial Processes - Diagnostics	Robert Kostecki (Lawrence Berkeley National Laboratory)	2-157	3.50	3.17	3.00	3.17	3.23
Model-Experimental Studies on Next-generation Li-ion Materials	Venkat Srinivasan (Lawrence Berkeley National Laboratory)	2-159	3.50	3.50	3.67	3.00	3.46
Investigations of Cathode Architecture using Graphite Fibers	Nancy Dudney (Oak Ridge National Laboratory)	2-161	2.80	3.00	3.00	2.80	2.93
Block Copolymer Separators for Lithium Batteries	Nitash Balsara (Lawrence Berkeley National Laboratory)	2-163	2.40	2.20	2.60	2.40	2.33
Interfacial Behavior of Electrolytes	John Kerr (Lawrence Berkeley National Laboratory)	2-165	2.60	2.40	2.60	2.40	2.48
Advanced Binder for Electrode Materials	Gao Liu (Lawrence Berkeley National Laboratory)	2-167	3.33	3.33	3.33	3.00	3.29
Atomistic Modeling of Electrode Materials	Kristin Persson (Lawrence Berkeley National Laboratory)	2-169	3.00	3.00	2.60	3.00	2.95
Coupled Kinetic, Thermal, and Mechanical Modeling of FIB Micro-machined Electrodes	Claus Daniel (Oak Ridge National Laboratory)	2-171	3.20	2.60	2.60	3.00	2.80
Long-Living Polymer Electrolytes	Christopher Janke (Oak Ridge National Laboratory)	2-173	2.20	1.80	1.20	2.40	1.90
In-Situ Electron Microscopy of Electrical Energy Storage Materials	Karren More (Oak Ridge National Laboratory)	2-175	3.20	2.80	2.40	2.80	2.85
Diagnostic Testing and Analysis Toward Understanding Aging Mechanisms and Related Path Dependence	Kevin Gering (Idaho National Laboratory)	2-177	3.50	3.25	3.25	3.50	3.34
OVERALL AVERAGE			3.04	2.92	2.93	2.88	2.94

NOTE: Italics denote poster presentations.

### PHEV Battery Cost Assessment: Brian Barnett (TIAX LLC)

### **REVIEWER SAMPLE SIZE**

This project had a total of 6 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Reviewers noted that the cost of the battery, particularly the dollar per kilowatt-hour (\$/kWh) figure, is a key element in the success of the DOE's program, as well as one of the most critical components to affect the success of automotive electrification. One reviewer praised the cost analysis is among the best he had seen in terms of a pure methodology, with its basis and findings essential to projecting market feasibility of PHEVs. The project was also characterized as a very timely topic with considerable value to researchers/developers engaged in this field.

Two reviewers particularly lauded the project's new approach to electric drive vehicle battery cost estimation, because the ability to accurately project the cost of different elements in the battery system provides the opportunity to concentrate activity in the areas that will bring the largest return, including accelerating the development in materials and other promising areas to meet cost objectives required for widespread adaptation of these technologies. A reviewer observed that this modeling can help accelerate development of most promising areas for cost reduction, therefore accelerating adoption of electric vehicles and accelerating the ability to meet DOE objectives of petroleum displacement.



One suggestion for fine-tuning the program was that because the costs are strongly affected by the production rate, two or more production rates should be included to quantify the effect of rate on total production cost of a battery.

# QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The reviewers complimented the project, with one noting that it demonstrated very extensive work and methodology, with wellthought-out assumptions. Another similarly noted the sound unbiased methodology with a broad range of production variables. Another cited that the approach to developing a cost model for batteries builds on the previous successful efforts to develop a model to forecast fuel cell costs. A third reviewer deemed the approach to be very solid for identifying the major trends to impact costs in a direct, "apples to apples" comparison, as long as the details of things such as process yields and system costs per material are adjusted in order to make this approach more robust.

However, one reviewer noted that given that the first two high-volume PHEV/EV vehicles coming to the market (the Chevrolet Volt and Nissan Leaf) use batteries with mixed metal oxide cathodes and laminated pouch construction, it would be very instructive to extend the current work to those systems to allow a reality check for these kinds of studies which are often based on a multitude of assumptions and hypotheticals. Another reviewer pointed out that the relative cost of a prismatic cell and prismatic-cell-based pack system versus a cylindrical cell and cylindrical-cell-based pack system is an important area which is not addressed. Also, the reviewer who cited the potential for "apples to apples" comparisons above said that cell manufacturing yield assumptions of 100% are not realistic. Another current drawback indicated by two reviewers is that there needs to be validation of the model, which means that the confidence level in the results would be low. For example, actual costs of cells compared to models should be integral part of this model validation.

## QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Reviewers recognized the project as being high-caliber work that is providing excellent results, although counterintuitive, within its current scope. One reviewer noted how the model makes it possible to quickly estimate the effect of a new material on the final cost of the battery; because cost is a strong driver in the program success, the model will serve to assist in making better decisions on the various program elements. Another reviewer said that it was a major accomplishment that the program has identified several key areas to substantially impact battery system costs and also helped identify the relative weight of factors.

Suggestions for improvement included questions about how the aggressive yield assumptions were made, and addressing the issue that the validation information is not clear. It was noted that within the scope of this project, there is less technology derivites but rather process-related optimization.

### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Several of the reviewers said that there was no specific evidence of collaboration or coordination with other institutions in the Tiax presentation, though two of the reviewers felt that the team has done a good job reaching out to collect information from the industry to utilize in their models. However, one reviewer suggested that TIAX could do much more by collaborating in the inverse fashion where they allow industry and academics to utilize their model to get the most value out of the work.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

Reviewers cited Tiax's steady progression to the end deliverables and that its plans to explore prismatic cell costs are appropriate.

Feedback as to what Tiax should pursue going forward included:

- studies should be Additional continued on new materials as the program develops new higher performance materials to replace the ones used in this study as part of an add-on or a new contract. Plan to revisit and update costs of specific materials and the entire system are key
- Plan to explore alternate cell format
- For prismatic designs, estimated cost differences between stacked and flat wound rigid case and flexible packaging variations should be examined.
- Relative estimated cost differences between a prismatic cell and prismatic-cell-based pack system versus a cylindrical cell and cylindrical-cell-based pack system should be examined.
- The country of origin for various material and hardware data should be classified and identified in some fashion.
- Future research should include the validation of the existing model before starting the next phase.

### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

One reviewer stated that the team has sufficient staff and resources to carry out the study. However, a second reviewer felt that it is not very clear the sources are available to provide models to the battery community to support models for stacked and flattened jelly roll prismatic and pouch cell this year or the next year. Three reviewers left no response to this section.

A reviewer advised that he would like to see this activity accelerated with the requirement that the actual model be made more available to the public, for example, with higher levels of funding, to give the best acceleration to the industry and DOE's goal of oil displacement.

## A High-Performance PHEV Battery Pack: Mohamed Alamgir (Compact Power)

### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The consensus among reviewers was that the project is well-aligned with DOE objectives, as it addresses key technical issues for PHEV battery packs such as cell cycle life and calendar life, which are critical in reliably displacing ICE power in automotive use. One reviewer observed how low temperature performance aggravates these issues and was a key limiter of cell performance, especially for power-sensitive applications such as the HEV and PHEV. The development of new cell/battery pack designs for PHEV applications is critical for advancing such vehicles, according to one reviewer, and another cited the outstanding development of the safety reinforced separator (SRS) for safety and cell stability.

One minor point of dissent was that the target PHEV10 application limits the potential for petroleum displacement.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED,

### FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?



Overall, reviewers seemed to feel that the approach taken was reasonable, with barriers well-identified and logical progression made, though two reviewers wanted more details, such as on the real improvements and differences existing between the various cell generations. Three reviewers specifically commented on the analysis of thermal management as being very accurate with an interesting innovation, noting the achievements in the areas of liquid cooling versus refrigerant-to-air cooling with OEM constraints of packaging. Two reviewers also noticed the SRS work, though one remark was that this is safety-related and does not directly address the stated goals of petroleum displacement. Some specific feedback for improvement includes: the data about cycle and calendar life lack unit scale to effectively understand the progress; and solvent permeation was not directly addressed or characterized.

## QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Though one reviewer said that progress was good, and another noted that the significant gains in the calendar life and shelf life of the battery pack should drive down application costs.

However, each of the four reviewers who responded to this question had feedback on what details were needed to better confirm improvements. Things that Compact Power should address include:

- Many presenters have used graphs without exact numbers. While it is not clear how these may impact confidentiality, this practice reduces the reviewers' understanding of the real process. However, the gap chart well quantified and clear, partially reducing the negative impact of the incomplete graphs.
- The refrigerant cooling is interesting, but some indications on the effective impacts (economical, technical, energy balance, complexity and reliability) would be useful.

- The refrigerated cooling thermal management (a refrigerant loop used to cool the air within the battery pack) appears to be an effective method to reduce/eliminate the need for high velocity air circulation (for air cooling), but, while this may maintain cell temperature during normal operation, it is unclear how helpful this would be to slow down or prevent thermal runaway during abuse conditions. How does this method of thermal management compare with liquid cooling during abuse.
- The safety reinforced separator (SRS) design in which an inorganic filler layer is applied to the surface of the polymer separator may not be optimal as indicated in the presentation by Entek (ES008). It is difficult to determine why the PLG2 design is "expected to meet the USABC target (cycle) life." Very limited data is provided for this design and no numbers are given in the redacted plot.
- One reviewer observed that PLG2 doesn't seem significantly better than PLG0 in terms of cycle life. PLG0 has a bad impedance rise but overall, the reviewer did not feel that PLG3 is that much better than PLG0.
- The data for cycle life and calendar life do appear promising (relative to PLG1), but there is considerable variability in the performance of the three cells.

### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Most reviewers noted the effective collaborations with the national labs for independent evaluations, particularly for a commercial outfit. One suggestion was that CPI could have involved academic interests for system modeling of heat transfer for the thermal management research.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The project was completed in March 2010, so this is not applicable. However, one reviewer indicated here that he was unimpressed by the pack design/temperature control work, which he felt should be funded by LG/CPI rather than by the DOE.

### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

Two reviewers said that there were no obvious resource issues, though the information provided in the presentation was not sufficient for an adequate analysis of resources. Three reviewers did not answer.

## HEV and PHEV USABC Battery Development Projects: Richard Holman (A123Systems)

### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The reviewers agreed that the project was clearly focused on the improvements of Li-ion batteries that support the DOE objectives. One reviewer observed that life and safety/abuse capability are key aspects for the application of PHEVs, a technology that will displace petroleum. Another stated that the large-format 20Ah cell design for LiFePO<sub>4</sub> is a significant step in providing higher energy density in a safe pack design for PHEV-40 applications.

### QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The reviewers felt that the standard product development processes were adhered to and that targets were within reach of production schedule, with a clear roadmap for meeting their deliverables. However, one reviewer felt that the approach was not explained in the description, although looking at project progress reveals the key steps to be improving cell design and BSF optimization. Another



reviewer said that it was not clear what the technology actually is in its project to develop prismatic cells using A123's technology from cylindrical cells, presumably based upon a high voltage nanophosphate material with a high voltage electrolyte (although this is only implied from the future work). One reviewer gave feedback that he would have preferred to see Arrhenius plots of some of their data to indicate changes in failure mechanisms (if any) with increasing temperature. He felt that all A123 showed was performance at various temperatures, whereas Arrhenius plots can be really helpful in understanding the effect of temperature, especially when paired with known or estimated activation energies for typical processes and failure modes.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

All reviewers had specific points of feedback to provide here. Two reviewers noted that the progress is impressive and steady on both activity lines, and two others noted the impossibility of gauging how well these cells have done at meeting/exceeding the USABC goals due to the presentations being qualitative rather than quantitative (gap analyses with color coding instead of quantitative data). Another reviewer said that the prismatic cells meet most of the PHEV10 and 40 performance targets, but do not achieve the system volume and cost goals. A third reviewer cited improvements in calendar life, cycle life, and power (though cycle life is not expected to meet the program goals), and a final reviewer commented on the nice safety performance.

Other specific points of feedback are:

- It is not clear why abuse testing is only referred to the EUCAR procedure and not to the USABC Abuse testing tests.
- The abuse tolerance at EU 4 is not outstanding. This reviewer acknowledged that the final abuse control is at OEM system level, and said that A123 should have expanded upon external short-circuit protection (which A123 does well) with integrated fuse. Potential improvements could be gained by advanced shutdown separators and/or CID/PTC devices

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- No information is provided regarding what the "doped nanophosphate" material actually is, although the presentation does indicate that this is a high-voltage cathode material.
- It is also not clear how far from the target the volume and cost metrics are. Redacted plots have been provided which give little information, but some short statements indicate that the cells have excellent capacity retention after thousands of cycles and have suitable abuse test characteristics.
- The SOC testing appeared to suffer from hardware issues (tightening of connections) which led to misleading data. The cells performed well for 80% SOC storage for 45°C and below, but not for the storage at 55°C.
- The cells cycle well, but the life is not expected to meet the program goals. For the HEV project with 32113 cells, the presentation indicates that most of the target goals will be met, except for the cost of the system.
- As color coding has been used, it is unclear how far from the cost goal the project is. No numbers are given in the redacted plots, so it is not possible to properly evaluate the cell performance.
- The A123 team don't seem to know what is their failure mechanism at 65°C, except that it's not iron solubility, which one reviewer did not find convincing.

### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Though collaborations were not specifically noted in the presentation, reviewers identified that the collaborations are reasonably limited to providing samples to independent national laboratories for testing and verifying project products.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

A reviewer wrote that the project does not propose any real future research to overcome eventual barriers but is clearly concentrated on production targets. However, another reviewer opined that the team should do more to get a better understanding of their failure modes at 65°C. A third reviewer observed that A123 said that next-generation separators (already developed) will be tested. Cycle life and storage testing will be conducted in PHEV cells. Scale-up of high voltage cathode material will be done with selected high voltage electrolytes. Cell characterization will be continued along with external evaluations of cell, modules and packs. No information is provided regarding how the project can meet the USABC volumetric and cost goals. The HEV project appears to be completed as sample cells have been delivered and a final report will be provided in June 2010.

### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

The reviewers' assessment was that A123 is well equipped to conduct all aspects of the projects, with its chemistry a viable option for HEVs and maybe PHEVs. Continued funding was recommended, even if a better evaluation would require the availability of quantitative data on the real number of products and complete planned activities.

# USABC PHEV Battery Development Project: Cyrus Ashtiani (Enerdel)

### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Comments on the titanate anode agreed that this can be a challenging alternative in low-cost systems if the project targets to have a highvoltage cathode and high-voltage electrolyte are successfully achieved. Also, the development of a titanate anode provides potential for improving both cycle life and safety within a PHEV pack. The focus on pushing to higher cell potentials begins to address one of the largest shortcomings of titanate vs. graphite/carbon anode materials. However, a dissenting reviewer stated that he did not believe the voltage penalty with Enerdel's anode can ever make this battery suitable for PHEV, and even an HEV would be a stretch.

Reviewers noted that developing a LNMO cathode from a surface coating could lead to significant energy density and lower production cost alternatives. Also, Enerdel's is a different approach to utilizing a high-voltage lithium battery cathode. By selecting an anode for which an SEI is not necessary for stable cycling, the remaining components (i.e., electrolyte composition) can be optimized to stabilize the cathode performance.



# QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

Most reviewers praised the approach, noting that the technical barriers are well identified with a clear approach towards handling the weak point of the LiTiO anode, the low working voltage of which must be counterbalanced by a large cathode voltage (provided by LNMO). Improved cathode materials and high voltage electrolyte formulations are likely necessary for this approach to be viable, but early demonstrations are promising.

One reviewer was more skeptical, however, stating that, in his view, many if not all of the electrolytes the Enerdel team are considering will have high viscosity and/or low conductivity that will effectively eliminate the high-rate performance of their battery, especially at low temperature. Without high rate, Enerdel does not really have much reason to use their LTO anode (except maybe safety), according to this reviewer.

## QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

The reviewers agreed that progress has been modest at best. One reviewer said that it can't meet several of the DOE goals, opining that the high potential is a serious handicap that Enerdel has not been able to overcome, and that there has been no demonstrated advantage to offset their lower energy.

A reviewer characterized the various approaches to develop new high-voltage electrolytes (sulfolane-based solvents, ionic liquids, fluorinated solvents and SEI-forming additives) as appearing to have been ineffective as all of these electrolytes performed worse in

cells than the high-purity conventional electrolyte formulation. It was suggested that the quality of the cathode material (material processing) played a significant role in the cell performance. The lifetime, weight, volume and price of the cells continue to be a challenge.

Specific criticisms included:

- There is no experimental evidence of the selection process of the electrolyte.
- The specific performances of the cathode and electrolyte developed are not clearly shown.
- The Enerdel team could have proven greater mass production capability.
- It is unclear if the same attention was devoted to creating high purity materials for the sulfolane-based solvents, ionic liquids, fluorinated solvents, and SEI forming additives.

### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

All reviewers noted the collaborations with the national laboratories for both materials and testing. One reviewer suggested that Enerdel could have involved an academic entity as well.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The project is completed, but work will continue in-house by Enerdel, including cell design optimization, cathode surface coatings, and advanced high-voltage electrolytes. Though one reviewer stated that the key steps for future improvements are justifiable with a focus on novel electrodes and advanced electrolyte, another disagreed, expressing the opinion that this system is much better suited for load leveling applications where power and very high cycle life are needed rather than energy, rather than for PHEV or HEV programs, and should consequently be funded by programs supporting those applications.

### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

Two reviewers felt the resources were adequate and funding of future research should be considered. Two reviewers did not comment, and one reviewer recommended killing work on this system for HEV and PHEV and instead funding it under a program aimed at supporting stationary power conditioning/load leveling.

## JCS PHEV System Development-USABC: Scott Engstrom (Johnson Controls-Saft)

### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Reviewers agreed that project is well in line with the DOE objectives because it is focused on PHEV design and optimization. In the project, prismatic cell lithium battery designs are being evaluated which utilize NMC as the cathode material and other lower cost materials that also have good energy for PHEV20 or 40 battery applications.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The consensus among the reviewers was that JCS was following appropriate cell development protocol with measurable output. JCS had utilized the same chemistry for cylindrical cells last year; the program scope was then changed to prismatic cells, leveraging existing Saft prismatic cell manufacturing capabilities. One reviewer felt that the approach is focused on some key aspects, but JCS needs to justify design choices better. However, another reviewer stated



that he liked the shift away from NCA to less expensive NMC that also has higher energy than LiFePO<sub>4</sub>, and also approved of the prismatic design for larger packs than cylindrical cells.

Specific places that could be improved included:

- Units would be nice
- The experimental results were more complete than in the presentations by other battery companies, but the planned work does not explain well how they really intend to significantly improve performance.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Reviewers noted that progress had thus far fallen short of DOE targets and would likely remain that way within the remaining time on the project. One reviewer noted JCS's significant gains in the cost arena, which are typically greater than similar projects, though short of USABC goal. Another observed that with the recent shift to NMC materials and prismatic cells, JCS understandably do not have much actual data to share yet. He expected JCS to be able to say much more next time, so that a rating of "fair" is more a reflection of where JCS is in its program rather than any reflection on the team's capabilities.

One reviewer did offer some specific critique, indicating that a plot was provided regarding packing efficiency in which it was noted that as the intra-cell spacing required for cooling increases, the packaging advantage of prismatic cells is reduced. It was not clear from this, however, whether or not the packaging advantage would be completely nullified. Redacted plots for cell performance were provided in which no values were given, which made it impossible to evaluate the performance thus far. It was verbally stated that the

current design is not yet close to the required USABC cost goal, but is approaching the goals for mass and volume. Very little specific information was provided.

### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Reviewers lauded JCS's collaborations on key components (for example, developmental separators from Entek and Celgard) and with national laboratories, though external evaluation at the national labs had yet to occur.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

Three reviewers said that the plans seemed vague, with a lack of detail on what barriers are really addressed, though one conceded that this is not unexpected for a commercial unit. Two reviewers did not comment.

### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

Two reviewers said that their general impressions were that the resources are reasonable, though one questioned whether the DOE needs to fund the building and testing of large demonstration packs. A second reviewer noted that this is one of the largest capacity hard-canned/vented prismatic cells under development and stated that funding for this novel approach should be increased.

## Advanced Cathode Material Development for PHEV Lithium Ion Batteries: Jamie Gardner (3M)

### **REVIEWER SAMPLE SIZE**

This project had a total of 4 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Reviewers unanimously agreed that this project supported the DOE objectives. Improved cathodes and anodes resulting in higher capacity and lower cost are crucial for advanced lithium battery development for EV and PHEV applications.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

One reviewer lauded 3M's good understanding not only of materials but also of cell chemistry and interactions with electrolyte, etc. Another cited that although the approach is not well detailed, it seems quite reasonable because it is supported by interesting results. For the cathode development, new compositions (related to NMC which is the benchmark) with less Co content (expensive and costly) are pursued.



Some criticisms included:

- No indications are given about the real technical barriers to be overcome.
- The concentration of work on NMC materials does not seem supported by a comparative analysis of competitive materials.
- The same applies to Si-based anode materials.
- 3M's cathode goals are modest because they are basically looking at relatively minor changes in materials. The main thing here is manufacturability of the materials at a lower cost.
- The anode program is more challenging, although 300 cycles is not enough of a cycle life. Si-based alloys are being explored. The key to these is stabilizing the cycling behavior and electrolyte optimization.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

The reviewers were positive about the progress being made. One reviewer said that the quantitative data presented confirm the technical progress with gap analyses that show both the results achieved so far and where things stand with respect to the targets. Another observed that they have met most of their goals on energy and costing. While not a huge jump in energy for the cathode, the progress shows that they have done a good job in working within the framework of what is basically the known space of cathodes using industrial mixture design, etc. The thermal stability data in this reviewer's view shows no significant improvement, but he felt that this was to be expected unless 3M sacrificed energy and went heavily in the Mn direction in the mixture space.

Another noted that the two cathode materials (Adv NMC 1 and 2) appear to meet the project goals of capacity, thermal stability and reduced cost. Cycle life analysis is still underway, but seems to be very promising (it is anticipated that over 2000 cycles will be obtained with this prototype material). The processing conditions produce materials with an excellent morphology in large batches. 18650 cells are being studied rather than coin cells. For the anode project, two different prototype Si-alloy materials are being tested.

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Melt spinning was used to produce the first material, while a proprietary manufacturing method is mentioned (low cost, high volume, quicker to scale) for the second material. Evidently, all is in order regarding the volumes of materials produced so there do not appear to be any issues with the scale of manufacturing. The materials also show promising results regarding their ability to meet the Year 1 targets for performance. Cycling data was not shown, but values are given for the cycle life (% fade) after 300 cycles (this did not include the initial 13% fade on the 1st cycle).

Two reviewers cited good anode results, particularly in view of the relatively early stages of such work. Overall, the anode materials provided a significant increase in volumetric and gravimetric capacity with reasonable cycling behavior.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Reviewers said they wanted to see more specific descriptions of any collaborations. One reviewer speculated that 3M has a large group and that confidentiality likely limits their ability to partner, so that lack of collaboration might be acceptable in this case. However, he would have liked to see plans to submit cell builds and materials to the national labs for testing and validation.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

Reviewers felt that the remaining steps were reasonable with attention to coating and final material selection for the cathode (one cathode material will be downselected for development), while for the anode material the proposed future work (optimization of coating and electrolyte formulation, cell design and abuse tolerance) is too generic. The same reviewer wanted 3M to be supplying samples to DOE for validation as indicated in the previous section.

### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

Reviewers agreed that 3M was well equipped and is working with other organizations as necessary for capabilities which they lack and external evaluations of their materials, so that resources are adequate. One reviewer specifically recommended continuing to fund this work through the DOE.

# USABC Battery Separator Development: Ron Smith (Celgard)

### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The majority of reviewers were dissatisfied with how the Celgard addressed – or did not address – the DOE objectives. One reviewer noted that though the work on separators is important – as yet another reviewer noted, the high-temperature melt integrity of separators is a critical property for the safety of lithium-ion batteries – the technical issues mentioned in the project seem marginal. Another suspected that Celgard is working in a vacuum, working on the correct subject but with a questionable execution.

Other reviewers registered even stronger objections, noting that Celgard's effort appears to be developing testing methodology that is closed or proprietary, and that this is a responsibility that should be left to Federal Agencies, the national labs, ASTM, SAE, or USABC and other consortia.

One reviewer was very unsatisfied with the completeness/level of information shared.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL



### BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

Reviewers generally expressed frustration with Celgard's proprietary methods. One reviewer did say that from the limited information provided, the approach appears to be very reasonable for the development of a test method to measure high temperature melt integrity (HTMI) and to use such a method to design safer separators for lithium batteries. Another noted that the work on standards is interesting, but it lacked details and justification of the differences with existing standards and procedures. The stability versus various electrolyte materials would be an added value for battery applications.

However, there was no description of Celgard's approach to its work on separator development. Another reviewer questioned whether Celgard should be attempting to define performance requirements outside of the scope of a potential customer. A third reviewer wrote that it doesn't seem appropriate to rely solely on Celgard's proprietary test methodology in a manner not subject to thorough review in order to set up a benchmark for future materials developed by Celgard. Finally, the last reviewer echoed this sentiment, pointing out that nothing in the Celgard talk addressed how they would actually make a better separator. The approach to coming up with a better test might have been useful, but for the fact that they are doing this alone and keeping it secret. For any new test to be useful, it has to be accepted and thus they would need to publicize the test and show why it is better than anything else out there. He indicated that Celgard will never get buy-in from others if they insist on doing their own thing, and said there's no (good) reason why they can't at least talk about test methods."

## QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL

### PROJECT AND DOE GOALS.

All reviewers agreed that there was insufficient information to be able to rate progress, so that this was not an acceptable review. The most strongly worded criticism stated that Celgard "did not report any accomplishments except that they have developed a test that they like, but refused to describe it or even say why it was better." There was no substantive information presented.

### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

The reviewers all said that they saw no evidence of any collaborations, and two suggested that they should look to collaborate with the national labs.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

No details were provided regarding future work for which a review can be made.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

Reviewers were at best neutral or did not answer. The more outspoken reviewers noted that though this work needs to be done, Celgard may not be the best choice of contractors to continue with this project without significant additional oversight.

The most strongly worded reaction was from a reviewer who stated that the lack of project details made it impossible to provide a good rating.

## Multifunctional, Inorganic-Filled Separators for Large Format, Li-ion Batteries: Richard Pekala (Entek)

### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Reviewers agreed that separator cost and stability are key issues for Li-ion battery development and applications. It is also very relevant and necessary, with the high temperature melt integrity of separators as a critical property for the safety of lithium batteries. The use of inorganic fillers to improve this property is widely viewed as one of the best and most practical methods of improving the melt integrity of membranes. One reviewer noted that this work mainly addresses safety by seeking to create stable films to high temperature (low shrinkage), while also providing a shut-down separator (other companies seem to have given up on doing both).

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

Reviewers recognized Entek's approach as being clearly focused and organized with clear steps to meet identified technical and economical objectives, and that they possess very good expertise in



# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Reviewers were impressed by Entek's progress in the few months since the project started in February 2010. One reviewer characterized the progress as interesting and well-described with some quantitative data well aligned with expectations. He also judged that the technical and economical barriers are likely to be overcome, with another reviewer observing that Entek is very close to getting a practical material, although it is not really addressing the cost issue, and instead is more focused on safety.

Another reviewer praised how a sample was provided to the reviewer and audience to demonstrate the quality of the developmental material. Embedding the inorganic filler inside the membrane rather than as a layer on the surface (the competing approach) results in separators with much improved properties (low MacMullin number, low shrinkage, and excellent wettability). The heat treatment step may be a vital part of the separator processing.

In terms of feedback, the reviewer noted that the puncture strength of the membranes is still not meeting the program goal. It is unclear why the Process B membrane (with 69% silica) has a much better puncture strength than the Process A membrane (with 67% alumina). One concern is the possible difficulty in drying these membranes prior to battery assembly as both fillers are known to be hydrophilic (which has caused considerable confusion in the polymer electrolyte with filler scientific literature). Questions regarding how the prototype samples perform in preliminary cell testing would have been helpful in gauging the utility of these separators.



#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

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Reviewers stated that partners were listed: Rhodia (which is supplying the inorganic filler) and Portland State University (for the electron microscopy work provided). However, no other collaborations or coordination with other institutions was indicated. It is not clear if these separators will be provided to DOE labs or other organizations for external evaluation. A reviewer advised that more integration and collaboration with electrodes and electrolytes projects are highly recommended. The stability of the investigated separators should be analyzed with the electrolyte and electrode materials studied in the program. Another reviewer recommended more co-operation with Sandia who are also looking at separator safety and developing new tests.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

Reviewers praised Entek's future plans as good. A reviewer noted that Entek seems to be moving forward aggressively and plans to get real samples into cell very soon. There were recommendations to improve collaborations and extend stability analysis to other materials sensitive to the program. Another reviewer noted that Entek's answers to questions show they are aware of and working on multiple issues not covered in the talk.

Another reviewer went into more detail in his assessment: For the future work, the concerns regarding the choice of filler will be addressed. New polymers and new process equipment may also be identified. Electrolyte compatibility and cell testing will be conducted. The project is listed as beginning in February 2010 so the progress thus far is very reasonable (it is assumed that the project has actually been on-going for a greater period of time).

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

All reviewers judged that Entek was well-equipped to conduct its project and using its resources efficiently. The reviewer who was most vocal in his disappointment about Celgard's presentation stated that Entek is "making great strides. I would like to see a separate effort on low cost materials."

ENERGY Energy Efficiency & Renewable Energy

Hybrid Nano Carbon Fiber/Graphene Platelet-Based High-Capacity Anodes for Lithium Ion Batteries: Bor Jang (Angstron Materials)

### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The reviewers all recognized that developing low-cost and highcapacity anode materials is an important step towards realizing higher energy density battery systems for electric vehicles. Three reviewers commented that the silicon (Si) alloys being investigated in this project were an attractive material for the goal of rangeextending capacity for PHEVs, which would in turn reduce the petroleum consumption, demand and (national) dependence. Also, a reviewer added that this work represents a U.S. technology base rather than a Japanese or Korean technology development.

Cautionary notes were that Si alloys have been plagued by problems related to volume expansion and particle fragmentation, and another reviewer added that "everyone" seems to be working on Si, while noting that this group does seem to have a number of approaches and one or more could prove workable.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL



### BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

Reviewers welcomed the team's approach, characterizing it as seeming effective, reasonable, and feasible to provide sufficiently high proportion of Si particles and thus high capacities exceeding 600 mAh/g. Another reviewer described the approach as well-presented, cost-effective, alternative, and well-defined with excellent variable considerations; he lauded that the approach is well-suited for manufacturing scale-up, in that both the nano graphene platelets (NGPs) and electro-spun carbon nano-fibers (CNFs) are low-cost nano materials and the Si deposition is being achieved by adopting the chemical vapor deposition system for mass production. A third reviewer conceded that though he was originally not excited about yet another way to form-C-coated silicon, the team at Angstron Materials also appears to have a rather unique method and also the background expertise to meet their goals.

Cautionary notes included:

- The high surface area is good for delivering high currents when demanded but has a problem in the first cycle loss when the anode is charged for the first time. With graphite having a surface area of about 3 m<sup>2</sup>/g, this amounts to about 5% of the total capacity of the cell going to form the SEI protective layer. While the SEI layer may have a different structure on silicon nanowires, at 9 m<sup>2</sup>/g, the first cycle loss will significantly reduce the total cell capacity.
- There is still the lingering issue of high irreversible capacity, which may negate the benefits of high reversible capacity, unless other lithium sources (than cathode) are explored.
- A reviewer was concerned about the large volume (low energy density in Wh/L) and large amount of electrolyte required for at least some of the team's approaches.
- The same reviewer also noted that costs don't seem too outrageous, but he was still not convinced that any of these manufacturing methods can really handle the huge scale and cost constraints of the auto business. In particular, he believed that while

electrospinning can be scaled up, its cost structure is simply too high for HEV/PHEV batteries. However, he noted that the team is looking at various methods, so it has options.

• The reviewer would also have preferred to see some more realistic and detailed cost estimates for a large-scale production system.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

The reviewers praised the project's progress in its first months, with a reviewer saying that the team had a very interesting approach, and another observing that the initial cycle life looks very good. One reviewer said that good progress has been achieved within one year both in terms of designs for suitable hybrid structures containing Si nano-particles during Li absorption and release and in developing low-cost materials and production methods. He also observed that the CNF or NGP geometry enables the supported coating to freely undergo strain relaxation in transverse directions, with NGPs providing a geometric confinement effect and a 2-D envelope maintaining good contact with Si particles. The resilience of such hybrid anode structures were demonstrated in half-cells, consistent with the DoE goals, according to this reviewer.

Another reviewer noted that the 650 mAh/g capacity for the silicon anode is about twice that of the graphite materials presently in use in Li-ion batteries, and that the team has designed a CVD process to produce a bed of amorphous silicon coated carbon nanowires and/or graphene sheets. The grain size is about 100 nanometers, a size which eases the strain when cycling as lithium enters/leaves the silicon alloy. The silicon loading can be controlled and the process is low-cost. The effectiveness of the anode has been demonstrated in half-cells. The team has developed the process for producing carbon nanowires, and the coated graphene sheets seemed to maintain capacity better on cycling.

A final reviewer raised a couple of concerns. He stated that the team needs to address costs more than they have done. Also, while there may be several ways to overcome initial irreversible capacity loss, they still need to demonstrate good coulombic efficiency after the first cycle or their cathode will run out of cyclable lithium with time.

### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Most reviewers felt that Angstron had reasonable collaborations, and with the right mix of partners.

One reviewer noted that K2 Energy is a forward-thinking group and a good match for Angstron. K2 presently has production in China as well as a new plant in Finland. Another reviewer observed that K2 Energy Solutions will assist in assessing these materials and in adopting them for various applications. A third reviewer stated that K2 is not what he would call a major player, however.

The first reviewer also characterized the Applied Sciences, noting that it will produce the vapor grown carbon nanofibers to go with Angstrom graphene sheets form a bed of carbon nanowires on graphene sheets. Another phrased this relationship as Applied Sciences will help in developing similar hybrid anode designs with vapor grown carbon fibers.

A reviewer said that though Angstron appears to have good links to other carbon sources, it still needs to do more in establishing collaborations. He felt that the biggest problem is that they appear not to have linkage with any of the DOE Labs, and said that Angstron should be providing samples to Argonne for characterization (and eventually to Battaglia at LBL), along with ANL. Cell builds at K2 should also include sampling to the DOE labs for an independent evaluation.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

Most reviewers were positive about the plans, with one saying it was an excellent plan to progress to production feasibility. Another said the proposed future research is effectively planned and the future plans are consistent with the overall goals and include developing low-cost mass production methods, continuing the evaluation of these materials in half-cells, and later in button cells and 18650 cells. A third reviewer described the plans themselves, noting that a larger lab-scale CVD system will be installed to produce greater quantities of experimental anode materials. The parameters of the Si coating processes will be optimized to make reproducible

anode materials for experimental purposes. Also, safe operating procedures will be developed for the silicon coating process along with the MSDS of Silane, and the detailed personnel training and protection requirements. The morphology of the silicon deposition, thickness, crystal structure and the weight percentage of Si coating will be characterized and the evaluation of Si-coated anode materials by the half-cell method will be conducted at Angstrom and K2 during FY 2010. A fourth reviewer said that Angstron will be pulling in their partners.

One reviewer was a bit more restrained, saying that he would have preferred to see some more realistic and detailed cost estimates for a large-scale production system. While this is not always easy to do, they should have enough information to at least make a stab at this. Also, Angstron (and others) need to address the coulombic inefficiency that can cause the cathode to run out of cycleable lithium during cycling. Even 99.9% anode coulombic efficiency is not good enough or 20% cathode capacity is lost in just 200 cycles. Finally, this reviewer would require them to provide samples of cells for independent testing by ANL/Sandia - both materials and cells.

### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

The reviewers felt that the resources were sufficient. One noted that the resources of about \$3.2 M with around 50% cost-share are consistent with the project scope in terms of developing new anode structures and scaling up the production methods. Another said that the budgeted amount should be ample.

A reviewer also agreed with the resource allocation, with some caveats. While he felt that Asian companies are ahead of the US in this area (cf. Panasonic's announcement of a 4Ah 18650 in the next year or two), the DOE needs to have someone in their own program to follow this approach and provide samples. He believes that Angstron is well-suited for this job; however, he would not recommend funding more than two efforts in this area. He would want to see a path to a viable low-cost manufacturing method before funding work beyond that time frame.

# New High-Energy Nanofiber Anode Materials: Xiangwu Zhang (NC State/NLE)

### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Reviewers generally felt that this project supported DOE's objectives, as the drive for new higher energy battery systems is essential for meeting the goal of establishing electric vehicles as a viable transportation method that does not use petroleum as the energy source, according to one reviewer. He added that the silicon anode materials significantly increase the capacity, 670 mAh/g, and therefore will result in higher capacity batteries, and therefore higher energy storage capability. Another reviewer observed that the energy density improvement is needed to reduce the weight and volume of the battery systems. The anode capacity improvement will help with the goal of increasing cell energy density.

A third reviewer echoed this characterization, stating that the objective is to develop high-energy anode materials for Li-ion cells, based on silicon nano-particles embedded in a carbon nanofiber. Lithium alloys, especially Si alloys, are promising to provide 2-3 times the capacity of graphite anodes, but their use is being deterred by problems related to volume expansion and particle fragmentation of Si upon alloying with Li. Such anodes, if successfully developed,



will contribute to an enhanced specific energy for Li-ion batteries to make them viable for PHEVs, which would in turn reduce the petroleum consumption, and pave the way towards petroleum replacement.

One reviewer offered some criticisms, however, stating that while they are aiming at high energy anodes and their approach does hold out the promise that they can maintain a Si-C conductive path during cycling, he did not believe that the team has anything significantly different from anyone else looking at C-Si anodes. He was also not convinced that spinning is commercially viable for the HEV or PHEV program, saying that just because it can be done on a large scale doesn't mean it is viable for the HEV program where costs are so critical.

# QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

All but one of the reviewers responded favorably to the approach. One reviewer said that the approach looks effective, reasonable and amenable to scale up, and the plan to demonstrate the performance of these materials is also sound. He described the approach as addressing the three main barriers of capacity, cycle life, and cost with such Si composite anode. The approach involves the development of Si/C nanofibers that were prepared by the electro-spinning and carbonization of Si/Polymer (e.g., PAN) precursor nanofibers. A second reviewer had a similar response, reflecting that the project was about electrospun carbon fiber support for Si, and indicating that the team seems to understand the issues and has an interesting approach. A third reviewer said the systematic development starting with spinned fibers and embedding silicone makes it very encouraging.

Another reviewer recognized that preliminary results give 650 mAh/g capacity, and said that the approach is interesting and novel. However, he had questions: the exact details of the processing to produce Si particles in and on the various fibers were not clear. He also asked why there was an effect of surfactant on the nature of the final product.

Other criticisms logged by the reviewers include:

- There is still the issue of high irreversible capacity of over 20%, which may negate the benefits of high reversible capacity, unless other lithium sources (than cathode) are explored.
- The approach seems OK, but this came across as a "me-too" program. The reviewer did not find the approach especially innovative in that there are many ways to embed Si on C (basically C-coat) Si. He did not see why this approach should really be any better and the volume density of their fiber approach would seem to be really poor. While this reviewer considered Wh/kg to be the main metric, Wh/L is still very important and the volume and added electrolyte needed to fill these porous structures seems likely to hurt both energy density and cost.

## QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

All reviewers approved of the progress made thus far. Comments included that the goal of twice the capacity of graphite was achieved with several different experimental configurations. The capacity was fairly constant during cycling. Another reviewer said that the progress achieved within one year is consistent with the project plans as well as with the DOE goals. Si/C nanofiber anodes, have been prepared using the electro-spinning technique, impregnating Si nanoparticles in carbon fibers made from PAN, and their performance demonstrated in coin cells, with a capacity of about 800 mAh/g, beyond the Year 1 Target of 650 mAh/g. Further, these materials have also been tested in 18650 cells, again demonstrating high stable capacity (of 600 mAh/g). A third reviewer said that good progress has been made in the first partial year, with improvements having been identified and applied (i.e. Si distribution).

Caveats included:

- A reviewer felt that the discussions should include more details and explanations of the experimental work.
- The fabrication of an 18650 cell was carried out to show the feasibility of the anode construction by American Lithium Energy. It was not clear why the cell voltage fell off at 150°C.
- Though the project has met the original goal, the cycle life may require additional development. Irreversible capacity may be limiting the energy improvement.

### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Reviewers noticed only two partners, one of which is a start-up focused on this work (Tec-Cell Inc., set up for transferring this spinning technology in the future, as one reviewer characterized it). The silicon-carbon/carbon composite anode - cobalt cathode cells were made by American Lithium Energy. A reviewer said the collaboration seems to be in good order as is the formation of a venture capital company to exploit the development. A reviewer speculated that possibly, other closer collaborations will be established with the DOE laboratories and other battery technologists in future.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The consensus among the reviewers was that the goals and milestones were reasonable, but there were some questions. A reviewer noted that the continued effort to establish the technology and improve the capacity to 1200 mAh/g and cycle life to 700 is absolutely needed. One reviewer said the future plans are consistent with the overall goals and include: i) optimizing the processing conditions and structure of Si/C nanofibers methods to achieve improvements in both capacity and cycle life, ii) continuing with their evaluation in coin cells and iii) fabricating 18650 cells with these Si/C nanofibers to demonstrate a capacity of > 600 mAh/g and cycle life of over 750 cycles with 70% depth of discharge. Another said that the effort to optimize the deposition process and optimize experimental parameters, such as flow rate, viscosity, surface tension and carbonization parameters is essential to success.

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A skeptical reviewer commented that the team didn't really explain how they were going to hit the next density goal. How is the performance going to be improved? A second reviewer said that while the goals and milestones seem quite reasonable. However, he was not sure how they expect to meet their goals. Also, the team and others need to address the coulombic inefficiency that can cause the cathode to run out of cycleable lithium during cycling. Even 99.9% anode coulombic efficiency is not good enough or you lose 20% cathode capacity in just 200 cycles.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

Reviewers were satisfied with the resources for this program. One explained that the resources of  $\sim$ \$2.6M with  $\sim$ 50% cost-share are consistent with the project scope in terms of developing new Si/C nanofibers using low-cost electro-spinning and demonstrating their performance in sealed cells. Another reviewer commented that the team seems able to do the work with what they have. Though this was not bad at all, he however questions the advisability of funding this work in terms of novelty. He was skeptical that even if they succeed, that material processing method can compete on either a cost or a volume basis with other, simpler approaches. Because it seems like everyone is looking at various ways to coat Si with C, this reviewer did not believe that the DOE needs to fund yet another such project.

**ENERGY** Energy Efficiency & Renewable Energy

Stabilized Lithium Metal Powder, Enabling Material and Revolutionary Technology for High Energy Li-ion Batteries: Marina Yakovleva (FMC)

### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The reviewers approved the project's support of DOE objectives, noting the potential for improved battery performance, including improving the energy density and reducing cell manufacturing cost. More specifically, a reviewer observed that the project addresses one way to help utilize high-capacity anodes that have either a high initial capacity loss and/or lower than desired coulombic efficiency. It also could enable non-lithium-containing cathodes (make cells with a charged cathode such as  $V_2O_5$  and discharged anode+SLMP).

A third reviewer said that the lithium metal preparation developed by FMC is unique and offers a good, safe method to incorporate the lithium into cell constructions. As the press for higher energy density continues, lithium metal offers the highest energy density over the Li-Ion system.

Finally, a reviewer described the project as follows: The high capacity Li-alloy anodes (both Si and Sn) have the disadvantage of high irreversible capacities, for which the lithium source comes



# QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The reviewers had a mix of reactions to the approach. A reviewer said that this potentially addresses a number of issues. Another reviewer said that FMC has worked hard to develop a safer means to incorporate lithium metal anodes in advanced battery systems. The "slump" materials offer a more efficient, safer means to incorporate lithium metal anodes into the cell construction. It can be used to pre-lithiate the graphite materials for use in Li-ion cell constructions, but the more interesting fact is that it offers a new opportunity for lithium metal in the new push for lithium-air and similar lithium metal and lithium alloy anode systems being developed.

Another reviewer said FMC's approach is to develop manufacturing methods for the SLMP. SLMP may be added to the anode to compensate for its irreversible capacity and even to lithiate the carbon anode such that it may be combined with non-lithiated cathodes. In either approach, the amount of SLM required to be added is considerably less, due to its high Li content,  $\sim$  98%. The approach involves the development of a process and prototype unit for the commercial production of dry stabilized lithium metal powder (SLMP) and the integration of SLMP Technology into the a Li-Ion system, e.g., MCMB/LiMn<sub>2</sub>O<sub>4</sub> system to demonstrate the



improvements both in the capacity as well as cycle life upon SLMP addition. A successful integration of the SLMP with the Li-ion chemistry is to yet to established, especially in the industrial production environment.

A couple of reviewers were more critical, however. One said that the use of stabilized lithium metal has not been successfully used in last 3-4 years and is not acceptable to most Li-ion battery developers. Another said that he is not sure why we need to have a pilot line facility at this stage. He recommended more lab work to demonstrate advantages and proof of concept of how you would actually implement this in a real battery production line needs to be done to assess practicality. He did not think a SLMP pilot line was needed to do this until and unless there are having trouble supplying samples. This reviewer still supported this work going forward, as it does open up new opportunities. Though he would much rather see lab work on using non-lithiated cathodes, he conceded that this needs to be done by others (such as LBNL/ANL).

## QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Progress achieved within the last two years was variously recognized as "excellent," and consistent with the project plans as well as with the DoE goals. One reviewer said that FMC offers help to the developers to efficiently incorporate lithium metal into cell constructions. It offers a much safer and efficient method over lithium foil to incorporate lithium into cell constructions. A second reviewer said that FMC has shown that the SLMP has potential, and a third characterized the team's activities as a prototype unit for the commercial production of dry stabilized lithium metal powder was designed, fabricated and tested and its process parameters have been optimized through design of experiments. Additionally, full pouch cell fabrication capability was developed. The benefits of the SLMP Technology have been successfully demonstrated in such pouch cells in the electrochemical system MCMB/ LiMn<sub>2</sub>O<sub>4</sub>.

A final reviewer said that the timing for installation was pretty good for an industrial process in light of safety issues when dealing with high-surface-area lithium metal. However, he felt that a pilot line is "a nice-to-have" feature, not an essential program goal, because he is not yet convinced that SLMP is even necessary yet. Cell work shows the idea basically works, but the improvement is limited to the irreversible capacity loss, which is not that high with the cells investigated. So the project is leading to incremental improvements only (although using hard carbons is certainly better than graphite as an evaluation tool). This reviewer would like to see this used with Si and other anodes with very high irreversible capacity loss ASAP.

### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Most reviewers felt that FMC needed more collaborations. One said they need a battery producer as a partner for this project. Another said that FMC says they are working with lots of people, but he feared it may not be an in-depth relationship in most cases, and he felt this company really needs to have a real partner working with them on this so that they can get feedback. He suggested the DOE try to pair them up with the people looking at high-energy anodes as it seems that both groups need each other's help to come up with a practical solution. A third reviewer noted that no partners were specified, but FMC claims to be collaborating with major research institutes and universities in the development of non-lithium providing cathodes and in enabling advanced anode materials, such as Si/Sn composites and hard carbons and in the development of the application technologies. It is also engaging in joint development agreements with major Li-ion battery manufacturers.

A final reviewer noted that FMC offers their expertise to all who chose to use lithium metal anodes. This includes not only the lithium metal but the easiest and safest method to incorporate lithium metal in production of the cell. As new people get involved in lithium metal, they will need the expertise in handling lithium metal safely. This is not a trivial thing, as the metal can be very reactive and most have had experience with lithium fires in one way or another in the R&D as well as production operations.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

Reviewers saw the future plans positively. One said that they had good plans and they seem to be working on the important areas as far as anodes go, though he was not so sure about non-lithiated cathodes. Good work but the reviewer would like to see it paired with DOE lab work on non-lithiated cathodes; they need help from the DOE labs on this. Another noted that the contract is based on the

use of lithium metal anodes in advanced battery development. FMC has the expertise and willingness to invest their efforts to assist in this important effort to develop new high-energy systems for vehicle propulsion.

A final reviewer said: The proposed future research is effectively planned and the future plans are consistent with the overall goals and include: i) Developing a process and design commercial unit to scale-up the production of SLMP dispersion in mineral oil (it is not clear how mineral oil dispersion is good for battery applications), ii) exploring the use of pilot scale alternative unit to produce dry SLMP powder directly from battery quality lithium metal and integrate SLMP Technology into the Li-ion cell using hard carbon/LiMn<sub>2</sub>O<sub>4</sub> system.

### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

The resources were seen as sufficient to meet the stated milestones. The resources of about \$6 million with around 50% cost-share over three years are consistent with the project scope in terms of developing commercial plant for the production of SLMP and integrating it into the Li-ion cells for improved capacity and life. One reviewer noted that the process is almost ready to go, so the team just needs a little help with scale-up and applications. As a result, he encouraged continued funding of this work, but would like to see it paired with DOE lab work on new non-lithiated cathodes.

## Protection of Li Anodes Using Dual Phase Electrolytes: Yuriy Mikhaylik (Sion Power)

### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Reviewers agreed that the project supports the DOE objectives. One reviewer noted the potential for improved battery performance, with another noting more specifically that the Li-S cell has the potential to be the maximum energy density. Li-S system has excellent energy density and offers a light weight, high-energy source for consideration as the next generation vehicle system. The metal lithium anode will provide higher energy storage capability than the Li-ion system. Other strengths of this system noted by a different reviewer are that in principle, Li-S could address a number of issues. The reasons are that the raw materials are relatively cheap, and Sion Power has demonstrated good rate, safety, and low temperature performance in the past.

A reviewer described his reasoning towards his approval of the project's alignment with DOE objectives as follows: the lithiumsulfur system offers high gravimetric and volumetric energy densities, due to the high specific capacity of the sulfur cathode. However, its advantages haven't been realized yet, due to the solubility of polysulfides from cathode in the electrolyte migrating to



the anode and affecting its cycle life. The objective of this project is to develop an electrolyte system with two immiscible electrolyte solutions for the anode and cathode compartments, each with adequate chemical/electrochemical stability and impermeability for the polysulfides towards the anode. The overall objective is to incorporate such an electrolyte system in large format cells and demonstrate the improvements in specific energy, cycle life and safety. These studies, if successful, will lead to widespread use of Li-S in PHEVs, which would in turn reduce the petroleum consumption, and pave the way towards petroleum replacement.

However, a caveat of Sion Power's Li-S system is that cycle life and energy are "not that great," according to a reviewer.

# QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

Reviewers expressed views that, in general, it was a good approach with good results so far, according to one reviewer. Another said it was a nice system and provides another option, and was interesting when viewed as a portfolio item. A third reviewer noted that the team has already surpassed the initial contract goals, and the key for initial success was the gel polymer developed to coat the lithium metal anode. The use of a two-phase cathode electrolyte solves several problems in the sulfur cathode formulations used in the rechargeable lithium metal anode.

Going forward, Sion Power should consider addressing the following: Sion Power needs to look at the impact of electrolyte on other performance/safety/cost aspects of cell. In particular, one reviewer pointed out that the lithium protecting layer using gel electrolyte is not robust enough to survive an automotive application. The cycle number should be equal to 5,000 cycles and the approach is targeted to 100 cycles. There is a big gap.

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The approach is to develop a two-component immiscible electrolyte system, specific to anode and cathode environments. The anode electrolyte will be compatible with lithium, immobilized in a gel and also works as a separator. More importantly, it doesn't dissolve polysulfides from the cathode. The catholyte, on the other hand, is tailored to improve the sulfur cathode performance with high polysulfide solubility. Even though the approach looks elegant and feasible from the preliminary studies, it is not clear how rapidly lithium can migrate across two immiscible phases to provide high rate capability. As well, it is not realistic to assume that the anode will be free of dendrites with the proposed gel electrolytes in ether solutions, based on the vast number of studies with Li metal rechargeable systems.

## QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Reviewers deemed the technical accomplishments and progress to be very good, with a couple of exceptions. One reviewer described it as reasonably good progress has been achieved within one year, consistent with the project plans as well as with the DoE goals. Improvements in both cycle life, beyond the modest target of 50 cycles, and in thermal safety have been demonstrated in small pouch cells with the proposed two-component electrolyte system.

Another reviewer said that the results demonstrate good promise for the system. The Wh/l and Wh/kg are essentially the same for the system. The two-phase cathode appears to be a good solution to the use of the sulfur cathode. The gel anode film gives good stability. Thermal runaway problems are under control and the demonstrated cycle life is twice that of the value targeted initially for this stage of development. A third reviewer echoed this, noting that Sion Power's team has exceeded durability and shown elimination of thermal runaway (up to 250°C).

Criticisms included that the DOE goal is 1,000+ cycle life, so that there is a big gap between the accomplishment and the goals. Another reviewer added that while the high-temperature safety data is very impressive, it's hard to understand how they can get above molten lithium without a problem. This was echoed in a different reviewer's statements as well, as he said the finding on the thermal safety beyond the melting point of lithium is unexpected and incomprehensible in a Li/metal gel polymer system. As for abuse tolerance, a reviewer said that he was still not sure about safety to crush Sion Power's cells either, but their nail penetration results are hopeful. Finally, a reviewer noted that though 350 Wh/kg sounds very good, although Wh/L is poor due to low density, making the battery pack quite large, which has cost and indirect weight implications.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

All reviewers recognized Sion's joint development agreement with BASF in Germany, where it has 14 scientists working on the project there. BASF has a working arrangement with Sion and works full time on the Li-S technology. A reviewer noted that the collaboration with BASF looks promising and adds strength to the team. However, another reviewer expressed that although the BASF chemical expertise will be very helpful in solving the polysulfide problem in the cell, he noted that there is no active development work discussion with them.

A couple of reviewers noted that there was no collaboration yet with any DOE laboratory, and one said he would like to see evidence of cooperation with Sandia on safety, especially for impact/crush work.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

It was recognized that Sion Power has generally stated a path forward, though a couple of reviewers had some salient questions. One reviewer said that the development of a larger format 2.5 Ah cell structure will provide the opportunity to confirm stability of the anode coating and the dual-phase electrolyte system, and the testing to confirm the potential to meet the USABC goals is also a good step. Another reviewer said the proposed future research is well planned and the future plans are consistent with the overall goals and include: i) optimizing the cell hardware including the gel polymer and electrolyte, ii) develop a model to simulate performance for automotive applications and iii) develop larger format prototype 2.5 Ah Li-S cells to demonstrate >350 Wh/kg specific energy and longer cycle life under USABC test conditions.

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Questions that some reviewers brought up about Sion Power's plan include that the lithium anode protection needs a robust Li-ion conducting barrier to last 1,000 cycles and the future research does not address the potential solution. Another reviewer said that he wanted to see a detailed cost estimate and volume/weight estimates, because he was wondering if Sion Power can ever meet the goals in any of these areas. One of the causes of low energy density is the relatively low percentage of actives in the cathode because they need a lot of solvent to solubilize the polysulfides, this reviewer opined. Work on improving the cathode actives should be pursued. Cycle life is poor as well.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

The program has sufficient resources to establish the feasibility of developing the system for use in vehicle propulsion. An increase in resources will be in order when the initial work produces a system with potential for longer cycle life. The resources of about \$2.8 million with about two-thirds cost-share over three years are consistent with the project scope in terms of developing Li-S cells with dual-phase electrolytes for improved capacity and life.

A reviewer who was a bit more mixed in his response said that the overall LOS still seems low, but the team is making progress and this is a good use of DOE funding, if only as part of a portfolio approach to give more options. However, the DOE needs to keep a close eye on this program to ensure that they can continue to address the cycle life issues. This reviewer would like to see a projected design for a complete battery pack to judge whether even if they were successful it would be a viable system. In other words, the reviewer was unsure that DOE would actually want their battery system even if Sion Power fixed their problems.

## Process for Low Cost Domestic Production of LIB Cathode Materials: Anthony Thurston (BASF)

### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Reviewers saw this project as quite relevant to the overall DOE goals, though dissenters had questions about degree of need and timing. A firmer supporter said that it is essential to develop domestic supplies of all critical materials used in advanced batteries for electric vehicles. This project takes advantage of the newly developed materials from Argonne National Laboratory to develop efficient production method. High-quality materials with an controlled particle size and purity are essential. Another reviewer shared this view, noting that it is important to develop low-cost production methods for cathode materials being developed under BATT and ABR and BASF is well positioned to undertake these studies. Another objective is to validate the cost and quality targets are met via coin cells, pouch cells and 18650 cells, with final incorporation into a battery pack for complete testing and extensive material characterization. These studies, if successful, will lead to the incorporation of high-energy cathodes in Li-Ion cells for enhancing the PHEVs, which would in turn reduce the petroleum consumption, and pave the way towards petroleum replacement.



A third reviewer added that possible battery cost reduction and production capability were the goals here, and another reviewer echoed this, saying the perception was that BASF will synthesize high energy composite layered cathode. The objective of the project is to reduce the cost of manufacturing cathode material which will help with reducing the cost of the battery systems.

A final reviewer had a mixed take, feeling that this project maybe addresses a supply chain need for a domestic (albeit foreign) manufacturer of cathode materials. His biggest issue is the large cost and why do this now, as surely this can wait until we know what materials we eventually want to make?

# QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

Reviewers noted that the project attempts to reduce the calcination time, which may reduce the cost of the current NMC cathode material. This will focus on improving one aspect of production and providing US production capacity. One reviewer built on this by saying that BASF already has expertise in the production of oxide materials. They are taking advantage of this expertise to develop the commercial production of the NMC cathode materials for advanced Li-ion batteries using advanced calcining operations in their Ohio facility.

Another reviewer phrased this sentiment in a slightly different fashion: The approach is to utilize BASF's production and R&D facilities in the US to develop low-cost production process for the cathode materials developed in DOE laboratories and to produce at a few ton levels. This requires the selection of proper starting materials and adopting suitable blending methods and calcining schedules. In addition, BASF will work with Farasis Energy, Inc. (Hayward, California) to evaluate these materials in 18650 cells and

with commercial partners such as automotive OEMs and Tier I suppliers to validate BASF's cathode materials and finally test a Li-ion battery pack containing BASF's cathode materials.

A dissenting reviewer characterized this work as scale-up using their existing equipment, which is just looking at a faster calcining method. He stated that the industry should be funding this on their own.

## QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Reviewers approved of the progress generally, with some caveats. A reviewer described that BASF has established a facility and is beginning to produce the NMC materials. The initial production gives good performance, close to the desired level. It would be expected as experience is gained in the production operation, the performance would be equivalent to the best materials from Japan and Korea. They have made arrangements to have production-type cells made by a third party to validate their internal tests.

Another reviewer said that reasonably good progress has been achieved within one year, consistent with the project plans as well as with the DOE goals. Improvements have been made in the synthesis of NMC-111 cathode with the process changes resulting in reduced processing time and increased potential production capacity, while maintaining product performance. The improvements in the pilot sample incorporated some of laboratory process improvements. Further experiments focused on optimizing the lithium stoichiometry, which helps reduce raw material costs and improves post-processing efficiency.

Questions included: What are their metrics? Any other improvements possible? Another reviewer noted that they have not started to synthesize the composite cathode material. A third noted that BASF seems to be making material faster and its performance is OK, but nothing very exciting or different. The cycle life of their best material does not look great.

### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

The collaborations with Farasis Energy and Argonne were noted by the reviewers. One reviewer said that is a bit early to accurately question on co-operation as it is just beginning. Farasis is capable of making commercial cells. It always takes a few trials in making cells to arrive at proper formulations for the best performance. Each cathode material has its own particle size, shape, surface, and handling characteristics. Another reviewer expanded on this, noting the collaboration with Farasis energy to assist in the assembly and testing of 18650 cells and packs from BASF-produced NCM cathode materials and to provide guidance for design modifications in order to meet customer requirements. There is also collaboration with ANL via technology transfer.

In a more critical vein, a reviewer noted that Farasis seems to be a very tiny outfit, and that there is no real discussion on working with ANL. A reviewer was wondering who the customers for this work are.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

Again, most reviewers approved of the proposed future research, though one reviewer said that he did not see what BASF was bringing to the program apart from manufacture/scale-up.

In the consensus view, one reviewer said that BASF is clearly focused on making the NMC materials and has the expertise to carry out the project to a successful conclusion. Another said that the proposed future research is well planned and the future plans are consistent with the overall goals and include: i) Completion of the Pilot Production Trials for NCM 111, NCM-A and NCM-B, ii) Validation of BASF Process Cost analysis for Production Customer evaluation and validation, iii) NCM production at Plant level and iv) Initiation of advanced cathode material lab phase in Pilot Trials for advanced cathode material. A third reviewer offered praise for the "good follow-on," but asked whether BASF have customers for production in 2011.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

Reviewers deemed the resources to be sufficient to carry out the proposed work. The resources of around \$5 million with approximately 50% cost-share over three years are consistent with the project scope in terms of developing low-cost production methods for the next generation cathode materials for their incorporation in Li-ion batteries. Two reviewers did not comment.

However, one reviewer objected, arguing that this is something that can wait and that industry should be funding on their own, and recommended killing this project from the DOE program. He asked, "Is this work really transferable to other cathodes? Without having a clear winner in the cathode area, it seems premature to do scale-up work."

## Overview of Applied Battery Research: Gary Henriksen (Argonne National Laboratory)

### **REVIEWER SAMPLE SIZE**

This project had a total of 6 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

All reviewers were agreed that the project was very relevant to the overall DOE goals. Each pointed out a key part of the program. One reviewer noted that ABR was aimed at expanding its portfolio and addressing long-term needs, which he deemed was a critical part of the program.

Developing high-energy and low-cost batteries was recognized as a key aim by one reviewer, who also advised not to forget long life, because life and cost are linked. More specifically, another said that high-energy couples are the only way to make substantial non-incremental improvements in cost (\$/kWh). Reducing battery \$/kWh will be biggest enable to replacing petroleum. Another reviewer echoed this understanding, describing the program as: The overall objective is to develop advanced Li-ion battery technologies and to assist industrial developers of high-energy/high-power Li-Ion batteries to meet the FreedomCAR long-term battery-level PHEV energy density (~200 Wh/kg) goal, while simultaneously meeting the cost, life, abuse tolerance, and low-temperature performance goals. These studies, if successful, will lead to the incorporation of high-



energy cathodes in Li-Ion cells for enhancing the PHEVs, which would in turn reduce the petroleum consumption, and pave the way towards petroleum replacement.

# QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

All supported the ABR program's approach, but there were some questions. On the positive side, a reviewer noted that the bulk of the tasks are well thought out. Another reviewer approved seeing tests done in 18650 cells. A third reviewer noted that the approach is based a systematic advancement of Li-ion battery technology, from developing new materials through their incorporation in prototype cells. Specific approaches in this advanced battery research include i) development of advanced high capacity/voltage electrode and electrolyte (high voltage) materials which will enhance the energy densities and lifetimes of the batteries and reduce their cost, as required in the 40-mile PHEVs. This reviewer deemed the approach to be sound with the required emphasis both on the science and engineering of these new materials. A fourth reviewer called the approach excellent for the intention of this project. Additional positive comments came from a reviewer who wrote that ANL's ability to integrate materials and approaches is key to reaching its goals. Its concept is very good and building ANL capabilities is also good.

On the other hand, reviewers also offered the following criticisms and suggestions: There are few items in the work plan which were quite a long shot and should have been transferred to the BATT program – as a case in point, the cathode work on silicates. The only deficiency in the approach is a lack of stipulated schedule for the incorporation of these advanced materials into the eventual products. One would like to see more rigid timelines for incorporating various generations of these new materials, especially with the automotive industry looking for such material advancements.
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One reviewer said he would have rated the approach outstanding except that throughout presentation it was mentioned that materials are sought in Asia (noticeably ignoring U.S. advanced materials manufacturers) and also industrial partners are not US parent companies (although they are called U.S. companies). There is no problem with ANL working with these companies, but there should be some way to involve companies that are established as U.S.-based companies. It seems like an obvious error in the approach to have tight relationships with foreign entities and no significant effort with existing U.S. companies in this area. The reviewer added that he did not understand why ANL almost exclusively looks for new materials outside of the U.S., and then when it has new materials, gets them scaled up or produced at pilot stage outside the U.S. Besides previous very visible examples, the latest case was the use of Korean Daejung EM to do (TM precursor) in ES0019. This reviewer understands keeping a global perspective, but the balance does not indicate the US DOE perspective.

The same reviewer also noted that several repeated themes in many presentations that should be considered or addressed: often project objectives were high-energy materials and often the presenters showed high-capacity materials. Capacity and energy are not the same, and in fact, in a number of occasions the theoretical energy densities would not even advance DOE goals, even if all materials worked perfectly. There is a need to establish baseline calculations and nomenclature around Wh/g and Wh/L to determine if programs are relevant to DOE goals.

A reviewer likes the targeted solicitations that are open to all and appreciate that the program is trying to reach out into the battery community. However, he felt that the DOE labs are overly represented in these programs and would like to see more new labs in the program - acknowledging that you have limited control over the proposals you get. For example, several PIs are working on both anodes and cathodes on this program, and the reviewer would prefer to see more people and institutions in the program. In other words, spread the funding out more.

The reviewer also said, "A bigger issue to me is that I feel strongly that the three- to four-year timeline of most proposals is far too long for seed money. Far too many of the proposals are complete projects. I would instead prefer to see more proposals that just target the killer issues and do proof-of-concept work only. Then if these are successful, the workers can apply for follow-up funding. This would force PIs to focus on the critical issues and not get sidetracked with engineering details. Almost as important, it would shut down unproductive programs early and permit funding of a wider portfolio. Thus, I'd suggest most projects have an 18-month to two-year timeline but with much more limited expectations. Obviously, the timeline has to be made on a case by case basis, but many proposals could be partially funded on the basis that they do the critical work we want, rather than funding an entire three- to four-year proposal.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

The reviewers were positive, though one did characterize the presentation as being more forward-looking than a report on results report, making it hard to say anything about the progress.

The positive commentary included a reviewer deeming that in general Argonne's labs have done an outstanding job in all fronts. I can take issue with specific topics here and there but that does not take away the high quality of work coming out of the labs. Another wrote that reasonably good progress has been achieved in the last year at various DOE and National laboratories and industrial partners. Several new materials have been developed in all cell components, i.e., anode, cathodes, electrolytes, additives as well as separators. In each category, various types of materials are being developed and assessed. Both modeling and materials diagnostic studies are underway to aid in the development of new materials. In addition, several studies are being carried out to demonstrate both the performance and safety of these advanced materials.

Some feedback that was more critical included: Few of the authors in the program have the unnecessary tendency of prematurely promoting certain results which the reviewer would not see pan out in following years. He said he very much respected the scientific aptitude of the team members, and it would be OK with him just to see the data. However, it bothers and confuses him when he sees results which were strongly hyped in previous years but don't show up at all in this year's review.

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Although this was about applied research, many of the programs project very long-term results. The reviewer said that he thought that more gains will be realized by having the researchers coordinate more of their efforts to meet tangible results and not just deliver excellent research papers.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Reviewers praised ABR's collaborations highly, calling them "excellent," "exemplary," and "very good." A reviewer noted that there are several fruitful collaborations among various partners in the ABR programs, i.e., among the DOE and national laboratories as well with the industrial developers.

Feedback included that a little more clarity on relationships in ABR's network would be helpful. Another reviewer said that he thought that some better partnering would help improve results. A third reviewer said that it was difficult to say where the collaborations are aligned to serve all US interests best, however. A final reviewer recognized that ABR had some outreach to other institutions, but he still would like to see more new faces in the program.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

Reviewers were positive, with one reviewer noting that ABR had plans for future projects that appear to support the barriers that are identified, though there were additional barriers to be addressed. Another deemed ABR to have a good portfolio looking at many key aspects. A reviewer described the proposed future research as to continue the material devolvement for stable and low cost electrode and electrolyte materials and fabricate cells with the most promising candidate materials, establish the performance and abuse tolerance of the current PHEV-type cells, and continue detailed modeling and diagnostic studies in support of materials development. These activities are directed towards meeting the overall DOE goals to develop high specific energy, safe and low cost batteries for automotive applications.

Specific suggestions included addressing additional barriers such as recycling. A reviewer said that one key would be what nextgeneration technology is adapted beyond the NCA baseline system; once that system is selected and logic given for its selection, it would be better to assess. Another reviewer asked whether there should be any topics related to research on batteries/battery systems for addressing the stationary market. A different reviewer noted that there needs to be a focus on life in a sealed battery (18650 or higher).

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

The reviewers were satisfied with the resources for the program, noting that they are sufficient to meet the stated milestones in the timeframe shown. One noted that the budget of about \$10-12 million per year over three years is well in tune with project needs and goals and the DOE objectives and are justified in the context multiple research efforts being pursued here. The emphasis is on basic research as this will place the United States in a leadership position in EV batteries globally and enable the US auto manufacturers to incorporate such batteries in electric vehicles.

However, one reviewer confessed to always being confused by the cell building facilities. Another reviewer expressed that he felt that additional resources may be needed to meet the additional recycling project suggested. Finally, a reviewer re-argued his point that though there is a need to keep funding this work, he would like to change the makeup of the projects and run many smaller ones as described above.

# Engineering of High Energy Cathode Material: Khalil Amine (Argonne National Laboratory)

## **REVIEWER SAMPLE SIZE**

This project had a total of 6 reviewers.

# QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The project was seen as being extremely relevant to goals for PHEVs and EVs, which do displace petroleum. Multiple reviewers agreed that the development of high-energy, long-life and abuse-tolerant Liion cathode material is absolutely critical to the commercial success of these batteries for automotive applications. In the words of another reviewer, the direction of program to produce disruptive cathode material at > 200 Ah/g and acceptable V is right on to enabling lower cost batteries, which are a key enabler to displacing oil.

A wordier characterization was as follows: In order to successfully meet the needs of PHEV and EV batteries, the specific energy densities of Li-ion batteries are to be significantly improved. This mandates an increase in the specific energies of the active materials, i.e., anode and cathodes. The development of high-energy composite layered mixed metal oxide cathodes, as is being pursued here, is thus highly relevant to the PHEV goals and the overall DOE objectives. These studies, if successful, will lead to the incorporation of highenergy cathodes in Li-Ion cells for enhancing the PHEVs, which



would in turn reduce the petroleum consumption, and pave the way towards petroleum replacement.

# QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

For the most part the reviewers approved of the approach, though there were some qualifications and criticisms. One reviewer said he thought the work is very solid, including that on morphology and composition to optimize rate and surface modification to improve rate and cycle life at high voltage. Another reviewer said that despite concerns about the stability of surface coating, he thought this approach is viable and should be pursued. He felt the group seems to have a very nice synthetic approach and the films look pretty homogenous.

A reviewer characterized the approach as appearing "sound and feasible," based on composite, solid solutions of  $xLi_2MnO_3 \cdot (1-x)LiNiO_2$ , whose compositions are being optimized here and the morphology suitably engineered to improve the rate capability. Various synthesis processes are being explored to obtain high packing density and different surface modifications are also being examined to enable high rate and long cycle life at high voltages (4.6V). However, the progress with this class of materials is rather slow, especially on the power densities and cycle life, possibly due to their inherently poor power densities, more so at low temperature, as reported form laboratories. Further, due to their sloping voltage profiles, the improvement in the specific energy is not as significant as in the specific capacity.

A couple of reviewers suggested some new research directions, as one wrote that he thought it would be appropriate to show work with groups developing an electrolyte that is stable at high voltage. He also said he thought it would be interesting to focus on storage and self-discharge when cells are stored at high voltage. A second reviewer suggested that he would like to see some work aimed

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getting a lithium ion conductive coating, either applied to the material or as part of changing the material's surface structure, though he was not sure this would be viable.

The criticisms included:

- While the surface-coated samples show initial good data, they oftentimes do not survive long cycle- or calendar-life tests. Hence he has general skepticism regarding the use of coating (AlF<sub>3</sub>) to alleviate the issue of life. We have seen such an approach fall apart after prolonged life tests, and so the reviewer said he would not use it as his first line of attack.
- Another reviewer felt this project has too many approaches to reach the intended goal. He said he thought that a more focused approach is needed to overcome the technical barriers, as not enough attention can be given to a high potential solution.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Reviewers noted the progress as being "good," "reasonably good," and "very impressive," though all had feedback and suggestions. A reviewer said that he hoped the authors do succeed in lowering the first cycle irreversible capacity as well as improving the rate capability using reproducible and inexpensive methods. Another reviewer noted that accomplishments are good working on addressing rate and stability, but that once performance is shown in larger cells, it will be more compelling. A third reviewer also identified the significant improvements in cycle life and stability, writing that though one can argue about the long-term stability of the coatings, he agreed with the PI that his work shows major improvements. The low impedance, better rate capability, and other features of these coated materials are also very welcome, which seem to be all around better rather than getting into the trade-off situation that is very common in the battery world. Still another reviewer commended the good progress, especially on capacity and density increase.

A reviewer characterized the team's work in this way: Reasonably good progress has been achieved in the last year, consistent with the project objectives. Some of the significant accomplishments include: i) Development of continuous co-precipitation process based on carbonate precursors, for spherical particle morphology with high packing densities of 2.1g/cc, ii) Understanding the effects of Li concentration on the material morphology and performance, especially with non-Cobalt doped formulations, ii) Improvement of power (even at 55°C) and life of Co-doped formulations with surface modification with e.g., AlF<sub>3</sub>.

Suggestions included:

A reviewer said that he would have loved additional clarifications as to the reasons for the irreproducibility of the non-Co samples as well as the poorer cycling performance of the Co-based samples. A second reviewer commented on this as well, observing that the Co doping is promising but variability is still high. What is the confidence level that this can be solved? What is the mechanism of Co impact on capacity variability?

Scale-up seems to be challenging and will need close collaboration with materials manufacturers.

Reviewers addressed questions of battery life, with one stating that calendar life is as important as cycle life (and maybe even more important) and needs to be addressed as well. Another reviewer added to his concern about life issues, as he noted that the program is stated as being 40% complete. However, significant improvements are needed to reach the stated DOE EV and PHEV goals, in particular the cycle, power, and calendar life. A few hundred demonstrated cycles in a small cell format versus a minimum of 5,000 in a large format cell is a long way from being 40%. If the DOE EV and PHEV goals are not the intended goals of this work, then there should be a more clearly defined set of goals for this work.

A reviewer said that he still had concerns about the stability in real cells of a cathode that is being charged to such a high voltage. He suspected that major electrolyte work may be necessary down the road to successfully implement the cathode material.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

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Reviewers all lauded the several successful collaborations from this team with teams from other DOE laboratories, industry partners, and even with overseas universities and companies (e.g., ECPRO, which assisted in the scale-up of the gradient concentration material, though one reviewer knew nothing about this company). One reviewer did suggest that it would be nice to see more collaboration with a group working on electrolyte, as high-voltage material will work only if a solution to electrolyte oxidation is found: perhaps a high-voltage electrolyte, SEI, etc.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

Reviewers indicated that the plans seem solid and look good, though they offered many suggestions. A reviewer characterized the proposed future research as to continue further optimization of these cathode materials in terms of composition, morphology (and tap density) and surface modifications in order to improve the specific energy, discharge rate capability and cycle life (at elevated temperatures), and to work with the industry partners to scale up these cathode materials for their incorporation and validation in 18650 cells. These activities are directed towards meeting the overall DOE goals to develop high specific energy, safe and low-cost batteries for automotive applications. Another reviewer said that he likes the approach to mixing particles of different sizes for better packing and higher density, which also can provide advantages in terms of using small particles for high rate and relying on the larger ones for energy.

One reviewer would recommend techniques other than coating to address the issue of life. However, another reviewer felt the work had too many approaches to the problem. The focus should be on life and scale-up, according to a different reviewer. The team needs to test its material in large cell format (18650 or higher). The team needs to get this material into real cells with a graphite anode as well. The team needs to look at long-term cycling and high temperature work as soon as possible to see if in fact these films can stand up to extended cycling. This work takes a long time so it's important to start it soon.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

The reviewers agreed that this is a highly worthwhile program which is at the least being sufficiently funded, and could possibly be accelerated with further funding, according to one reviewer. This reviewer deemed it one of best-matched programs to the DOE objectives, and relative to others in this area this should be accelerated with funding while others are reduced. A second reviewer echoed this sentiment, saying that with a little more focus on the highest potential solution(s), the work will reach the DOE goals in a more timely fashion. Another reviewer said the budget of \$300K per year is probably low from the perspective of this task alone, but seems reasonable and justified, since multiple efforts on this class of cathode materials are underway at DOE laboratories and being supported by ABR.

# New High Energy Gradient Concentration Cathode Material: Khalil Amine (Argonne National Laboratory)

## **REVIEWER SAMPLE SIZE**

This project had a total of 6 reviewers.

# QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The reviewers uniformly recognized the value of high-energy, longlife and abuse-tolerant Li-ion cathode materials towards achieving the commercial success of Li-ion batteries for PHEV and EV applications. In further detail, one reviewer characterized the work as follows: In order to increase the specific energies of Li-ion batteries to meet the 40-mile range of PHEVs, new high specific energy materials are required to be developed. The development of highenergy mixed metal oxide cathodes, with specific capacities in excess of 200 mAh/g, high rate capability, life characteristics and abuse tolerance is thus highly relevant to the PHEV goals and the overall DoE objectives. These studies, if successful, will lead to the incorporation of high-energy cathodes in Li-ion cells for enhancing the PHEVs, which would in turn reduce the petroleum consumption, and pave the way towards petroleum replacement.

# QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?



Reviewers approved of the researchers' approach, modulo some concerns and criticisms. A reviewer described the approach as based on achieving a desirable concentration gradient in the layered mixed metal oxide (NMC), such that the core of the particle is rich in nickel concentration, e.g.,  $\text{Li}[\text{Ni}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}]\text{O}_2$  for maximizing the specific capacity, while the surface is enriched with manganese, e.g.,  $\text{LiNi}_{0.5}\text{Mn}_{0.5}\text{O}_2$  for cyclic and thermal stability. Such gradient composition across the core and shell is achieved by successive coprecipitation in respective solutions. The approach looks interesting and feasible, as demonstrated here. Also, it integrates well with the state of art materials and their processing. Another reviewer built upon this, noting that the team's concept seems very sound and is a nice away to overcome the issue of cracks forming in two-phase/coated particles.

Concerns and criticisms included:

- A reviewer said that while this idea is certainly very elegant on paper, he was not sure how scalable, cost-effective the method is.
- The gradient core-shell approach type of material is very interesting. However, the reviewer wonders how easy it will be to control the consistency and quality of the product on large scale production, and believes this is a question to be addressed.
- The overall project has good clear focus, however there is a need to have clear, quantifiable goals and results, rather than just measures of "excellent" or "good."
- There is a need to evaluate safety in cells relative to individual materials.
- The project team should work with credible materials manufacturers, as there are many nuances in producing materials that are ready for implementation. It appears that BASF and ECPRO are not experienced.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL

## PROJECT AND DOE GOALS.

The progress was recognized to be good by most reviewers, though each had suggestions as well. One remained unconvinced, saying that he would like to see more data, because it was not clear that there was much change from the data that was presented last year.

A reviewer described the research as follows: Good progress has been achieved in the last year, consistent with the project objectives. Some of the significant accomplishments include: i) Development of a new gradient concentration cathode material with a high capacity of 209 mAh/g at 1 C rate when charged to 4.4V, ii) Verification of the gradient composition through analytical studies, iii) Demonstration of improved cycle life and abuse resistance relative to the core material (this was noted by another reviewer as well, who also expressed his hope that the results were repeatable), iv) Safety performance of gradient concentration material is excellent when compared to the bulk material and v) Further increase of Ni in the core and Mn in the shell. Another reviewer used the unorthodox word choice "very cute" to describe the results, especially the SEMs. He expanded upon this by saying that the pictures of the core and the shell are noteworthy as well as the data that showed the gradients for the respective elements.

Feedback included these comments:

- One reviewer found the abuse-tolerance data quite attractive but was not sure how comprehensive they are.
- End-of-Life abuse tests are very interesting to validate the beginning-of-life findings.
- A study of aging mechanism of this type of material would probably be very useful to understand how the core and the shell age.
- What is the impact of the shell thickness in performance? How easy is it to control?
- The work needs to get out of lab in order to be more credible.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

As with this PI's other project, reviewers acknowledged good collaborations with universities and other DOE laboratories in the material optimization and characterization and with industry partners on the incorporation of these materials in 18650 cells and similar prototype cells. However, some feedback was that because an electrolyte stable at high temperature will be key to the success of this type of material, the program should have more collaboration with electrolyte studies. Another reviewer felt that stronger manufacturing partners would be helpful for validating the economic viability of this approach and the benefits versus cost over standard approaches.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

A reviewer recognized that the steps outlined follow a progressive path to improving the performance and eventual production of the material as the deliverable of this project. The proposed future research is to continue further to scale up the synthetic process to obtain pure GCM in 100~500g batches, to optimize the composition and thickness of the outer layer of the gradient concentration to maximize the surface stability of the material, to carry out lifetime and thermal stability studies, and finally incorporate these materials in 18650 or similar prototype cells. These activities are thus directed towards meeting the overall DOE goals to develop high-specific-energy, safe and low-cost batteries for automotive applications.

However, reviewers were mixed in their feedback. One admitted that he was not fully convinced that this is the technique that would lead to the target material with the desired performance/cost metrics, but nonetheless, it would be interesting to use this technique on the composite cathode. Another asked why the team was not using a core material without Mn to increase the capacity even further. A final reviewer objected to the plan to scale-up the process, as it still seems to be in too early a stage of development to do that.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

It was felt that the resources were sufficient if the project keeps focused as outlined, with another reviewer judging that the budget of \$300K per year is adequate for the on-going as well as future studies. A different reviewer praised the good use of DOE funding, a novel approach, and the good results. He recommended that DOE keep funding at this level to continue progress, with the caveat that



he would not want to fund work at a chemical company to do scale-up because the technology not yet ready and this type of work should be borne by industry.

Design and Evaluation of Novel High Capacity Cathode Materials: Christopher Johnson (Argonne National Laboratory)

### **REVIEWER SAMPLE SIZE**

This project had a total of 7 reviewers.

## QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Most reviewers felt the overall DOE objectives of petroleum displacement were supported, with an exception. One reviewer described the work as follows: in order to increase the specific energies of Li-ion batteries to meet the 40-mile range of PHEVs, new high specific energy materials are required to be developed, which are inexpensive and thermally stable. The objective of this project is to evaluate low-cost vanadium-based compounds with high capacity as cathodes in conjunction with a lithium source in the form of Li-rich Fe oxides. These studies, if successful, will lead to the incorporation of high-energy cathodes in Li-Ion cells for enhancing the PHEVs, which would in turn reduce the petroleum consumption, and pave the way towards petroleum replacement. Another reviewer echoed this, stating that the cathode is one of the key cell components to developing a battery that can provide the energy needed to support the PHEV and EV energy and power needs. This work supports that effort.

In support of the project, it was noted that the PI's focus on

substantially increasing charge densities in LFO is appreciated. This reviewer recommended the use of energy densities instead of charge densities. Another reviewer praised the new approach to getting lithium into the cell and enabling the use of higher energy density cathodes.

However, a dissenting voice said that the energy density and cost profile of these materials are not appropriate, and that more basic research was necessary, but even so, the link to DOE objectives is not apparent. Finally, another reviewer admonished the team not to forget about life, which is a barrier.

# QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

Reviewers were split on this approach, described by one reviewer as based on blending  $Li_5FeO_4$ , which has a large amount of sacrificial lithium (5Li<sub>2</sub>O) functioning as the lithium source with a high capacity cathode,  $LiV_3O_8$ . This strategy is expected to allow the use of ultra-high capacity anodes such as Si-C nano-composites with large first-cycle irreversible capacity and improve the thermal stability of the cathode battery, due to the presence of Fe. Inexpensive and abundant Fe and V oxides will thus be utilized for a widespread use of Li-ion battery technology.

This reviewer added that one uncertainty of this approach is the poor cycle life of vanadium oxides, in general, which has been a deterrent factor in their use. Another reviewer echoed this sentiment, stating that approaches using Fe are definitely worth pursuing for cost, abundance, and environmental advantages, but he was not sure about V in that regard. He added that the problem with this approach is that it leaves a lot of inactive material in the positive electrode and so a large irreversible capacity, and that this seems to be a long-term research effort that still needs to go through many barriers.



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A third reviewer said there is a need to look at higher capacity cathodes, provide for safety, and address the cost concern as well, which he felt this work is attempting to do. There is a need to look at incorporating this material into a larger format cell. A fourth reviewer agreed, saying the approach gets high marks for being innovative and different. It could be an enabler for a whole new range of high-energy cathode materials, much in the same way that SLMP does. The emphasis on science, proof and understanding is very appropriate for this stage of development.

However, a dissenting voice said that he could not understand how this would move towards vehicle technology goals. Even if rate and fabrication constraints could be overcome, it is hard to imagine relevance based on very poor energy density and the commercial viability of vanadium.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Most reviewers felt there was good progress – but a few felt that when DOE goals were considered, this was less true. Good progress has been achieved relative to demonstrating the concept, according to one reviewer, but another reviewer felt that the results were only fair when judged with respect to the overall DOE goals. He noted that there were quite a lot of interesting results with the promise of high capacity that have been obtained, but these might not translate into high-energy systems because of low voltage. As a result, he would be tempted to place this type of research within the scope of a BATT program since these will need a lot of research/development time. A third reviewer noted the voltage range of  $LiV_3O_8$  to be very large, and the average voltage is low, which may make utilization in automotive applications difficult. Another reviewer applauded the good progress at proving that there are other viable choices for cathode material, however a concern was about the large irreversible capacity loss and the implications of needing a much larger battery to meet the goals. Although high capacities were shown, the energy density is nowhere near where this should be to be funded as anything other than blue sky research

Suggestions include the following:

- It was suggested that this may be a good system for high-capacity advanced anodes such as Si/C to compensate for their high irreversible capacities.
- A reviewer said they have demonstrated the concept and more importantly to me have gained understanding of the fundamentals involved through the XAS work. The voltage profiles are always very sloping for these V-based materials where they go through several oxidation state changes. However, that by itself is not a big deal and could even be turned to an advantage in terms of fuel-gauging the battery pack. The use of the Li<sub>5</sub>FeO<sub>4</sub> would seem less useful from a fuel-gauging perspective as it would only show up during deep discharge and typically you'd not want cells to cycle that deeply anyway.
- With two cathodes it would be helpful to be clearer when reporting data on mAh/g to state what the g are are they referring to the V cathode, both active, or the whole electrode?

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

The consensus was that there are good collaborations within the DOE laboratories in the material characterization and with industry partners in the evaluation of Fe-oxides for pre-lithiation, though a couple of reviewers wanted it to be clearer who the industry collaborators were. A suggestion was that the team may need to get more involved with cell makers as they move forward.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

Reviewers felt that the proposed future work looked clear and interesting, but two reviewers questioned whether it might better fit within the BATT program because of its long-term timeline. The proposed future research is to continue exploring new variable Fe/Co ratios for  $\text{Li}_x\text{Fe}_{1-y}\text{Co}_y\text{O}_4$  as well as pure  $\text{Li}_6\text{CoO}_4$ , and  $\text{Li}_6\text{MnO}_4$ , understand the conditions of release of  $\text{Li}_2\text{O}$  and evaluating new charged cathodes such as  $V_2O_5$ , MnO<sub>2</sub> and delithiated olivines MPO<sub>4</sub>(M=Fe, Mn, Co, Ni) in the blended cells. Also, the prelithiation precursor cathode system will be tested against high-capacity, high-energy anode. These studies are oriented towards the project goals of achieving low-cost and thermally stable high-capacity cathodes.

Feedback as to what other changes the team should consider include:

- Future research needs to look at life and the thermal stability of these materials.
- A reviewer would like to see more defined, quantifiable goals.
- The project really needs to address and get a better estimate of specific energy and energy density for a final cell. The reviewer is still very concerned about the low cell voltage of these V-based cathodes and also their density, which he believes may be quite low. One of the problems with a high-capacity, low-voltage approach to energy is that you need to pair it with a large anode to accept the capacity and energy/capacity estimates and data on just the cathode miss this effect.
- The same reviewer would like to see an energy density comparison of putting lithium in the cathode vs. the anode (SLMP) in combination with these non-lithium oxides. The PI is not expected to be an expert in SLMP or necessarily "attack" it, but a good faith comparison should be done, if not by this PI then by someone in the program using a consistent methodology (apples to apples as much as possible).

### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

Reviewers saw the resources as being sufficient, with one exception. The budget of \$300-400K per year is adequate for these studies, according to one, and another said that this is a new approach and needs continued support for a while at least. However, he would want to see a better estimate of possible energy density very quickly and certainly before the next review or funding decision.

The dissenting reviewer did not think this is a good use of funds for vehicle technology applications.

# Search for High Energy Density Cathode Materials: Ilias Belharouak (Argonne National Laboratory)

## **REVIEWER SAMPLE SIZE**

This project had a total of 7 reviewers.

# QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

As with the other cathode materials research projects, reviewers noted the need for the cathode as a key cell component to developing a battery that can provide the energy needed to support the PHEV and EV energy and power needs for use in an automotive application. In order to increase the specific energies of Li-ion batteries to meet the 40-mile range of PHEVs, new high specific energy materials are required to be developed. The objective of this project is to develop new cathode materials of the type, Li<sub>2</sub>MSiO<sub>4</sub> (M=Mn,Fe,Co) materials, with the ability intercalate multiple lithiums and thus provide high specific capacity. Another related objective of this project is to overcome the barrier of poor electronic conductivity of these materials. These studies, if successful, will lead to the incorporation of high-energy cathodes in Li-Ion cells for enhancing the PHEVs, which would in turn reduce the petroleum consumption, and pave the way towards petroleum replacement. One reviewer did level the criticism that he believed that the theoretical energy density of this system (based on theoretical capacity, voltage, and theoretical crystal density) is lower than needed to make any useful impact to vehicle technology goals.



# QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

Two reviewers were lukewarm to this approach, while others approved of it. The approach is based on exploring the materials isostructural to  $Li_3PO_4$  for their ability to intercalate two lithiums (with ~ 333 mAh/g). Since the as-prepared  $Li_2MnSiO_4$  is "almost" electrochemically inactive because of its large aggregates and low electronic conductivity, two different approaches are being adopted, i.e., to reduce the particle size to sub-micron level and to apply a suitable surface coating, e.g., carbon, to activate  $Li_2MnSiO_4$ . A reviewer said that the concept of trying increase capacity while still maintaining a high voltage is sound.

A reviewer said that through the strategy to look for multi-lithium-intercalating cathodes for an improved specific energy, the choice of  $Li_2MnSiO_4$  is rather poor due to its sub-par electronic conductivity and low capacity even after milling to sub-micron size and with surface coating. Another reviewer felt the work is mostly exploratory in nature since these are new systems. He acknowledged that silicate would be a highly attractive material in case it can be made to work, but that this is a daunting challenge and will need years of R&D work. He added that the capacities are so negligible that he wouldn't waste his time building a Li-ion cell. A third reviewer was also a naysayer, saying that upfront theoretically-based work utilizing theoretical densities and best-case scenarios for energy densities would have indicated this was not a promising area of research against the primary objectives of DOE.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Several reviewers made distinctions between the good progress made towards the project's goals and progress with respect to the DOE's goals, which some felt the project did not address well. One such view was expressed by the reviewer who stated that good

progress has been achieved relative to demonstrating the concept. Specifically, successfully synthesized, characterized and evaluated the targeted structures of  $Li_2MnSiO_4$  and integrated carbon nanotubes as conductive matrix during the synthesis. The project identified that the amorphization is responsible for the capacity fade of  $Li_2MnSiO_4$  upon lithium removal, and proved that iron incorporation into the cathode results in structure stabilization, and promise in terms of capacity retention. Another said that significant improvement of electrochemical activity has been made, but capacity and voltage remain low. A third reviewer conceded that while the results are nowhere near the DOE targets, significant understanding has been achieved of the behavior of silicate-type cathode materials, especially understanding of amorphization using a slew of analytical techniques as well as ways to alleviate this issue. Of course, the achieved capacities are still insignificant, and the reviewer hopes the results thus far give the authors sufficient clues as to the future direction of the work. Yet another reviewer complimented the studies on degradation mechanisms, although not yet complete. However, he was still very concerned about low discharge voltage and poor cycling, and said it needed major improvements for this to be any good.

One reviewer who registered the greatest concerns about the project said there was no clear difference in the data presented this year and what was presented last year. Some of the SEMs presented did show some of the accomplishments, but the relevance to the DOE goals was not clear.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Reviewers felt there are good collaborations within ANL in the material characterization. However, collaborations outside ANL (and DOE) are yet to be established, with one reviewer asking whether anyone in the industry is being viewed as a potential collaborator.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

Reviewers were generally mildly complimentary about the proposed work, with one reviewer saying it looks well thought out and two others saying the plan is OK. He added that a combination of mechanistic work and focused research on related materials might open up a new generation of stable/low-cost cathode materials. Another called the project as overall a very interesting approach. It is now needed to understand quickly the reason for the limitation of the capacity and find ways to lower the polarization between charge and discharge. Is the low capacity linked to a kinetics limitation?

Feedback and concerns included:

- It is now needed to understand quickly the reason for the limitation of the capacity and find ways to lower the polarization between charge and discharge. Is the low capacity linked to a kinetics limitation? As soon as this is understood, the life question should come next.
- The result goals need to be more clearly defined in quantifiable terms.
- One reviewer expressed that he was a little concerned about what he viewed as the low LOS for this material in view of all its problems. He would have liked to see plans to widen the search for materials, as currently it seems a bit too narrowly focused.
- Finally one researcher said that he thought the plan was OK if he thought the research was relevant, which he did not believe it was.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

Opinions were split, with three reviewers saying the budget of \$300K per year is adequate for these studies. One suggested that working with an industry partner would help. However, other reviewers were more skeptical. One admitted that based on other questions he does not think this should receive more funding. The other reviewer was more measured, recognizing that this is quite a long-term commitment. This reviewer was OK with that as long as they reevaluate their progress and know when to pull the plug on a group of materials and move on to something else, and that there was a need to be vigilant and avoid the tendency to "study a dead horse to death."

Development of High-Capacity Cathode Materials with Integrated Structures: Sun-Ho Kang (Argonne National Laboratory)

### **REVIEWER SAMPLE SIZE**

This project had a total of 7 reviewers.

## QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

As with all of the other high-capacity cathode materials projects, reviewers deemed the project to be highly relevant to DOE objectives. The specific energies of Li-ion batteries are to be increased to meet the 40-mile range of PHEVs, which mandates the development high-energy cathodes and anodes. The objective of this project is to develop low-cost and high-energy, and thermally-stable cathode materials with integrated structures with spinel components in the layered-layered nano-composite structure, based on manganese-rich formulations and demonstrate the performance enhancements relative to capacity, rate capability, cycling and thermal stability. These studies, if successful, will lead to the incorporation of high-energy cathodes in Li-Ion cells for enhancing the PHEVs, which would in turn reduce the petroleum consumption, and pave the way towards petroleum replacement.

Another reviewer echoed that the cathode is the key cell active material component to developing a battery that can provide the energy needed to support the PHEV and EV energy and power



needs. This project effort attempts to address some of the concerns associated with that component.

# QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The reviewers were generally supportive of this approach, though there were caveats. The approach is based on embedding spinel component in the 'layered-layered' composite structure. Spinel structure may be created in the composite structure by controlling lithium content (e.g., 0.5<X). A reviewer said that of the overall approach, the stabilization of the surface is the most important. This is the only way we will get "5V" materials actually working. The electrolyte will play a big role as well and that work needs to have collaboration with electrolyte research. A different reviewer commended the systematic approach, which he thought was sound for studying this system.

Concerns raised included:

- A reviewer questioned why the manufacturing partner for this is not US-based but rather Korea-based. He commented that this seems consistent with Argonne's approach, but he felt that it was something that is not necessary.
- Another reviewer observed that the approach appears to address the stated goals of the project, but he had a concern that the DOE-specific goals were not receiving adequate attention.
- It is a pretty empirical mixing of materials, according to a reviewer, who would have liked to see more electrochemistry such as use of differential capacity (like the Hawaii Group) to really understand their cathodes. They need to compare performance with a simple physical blending of the component materials. This is in their plans but really it should have done right away to see if there is any real difference before continuing detailed studies on these integrated materials.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

The responses of the reviewers were mixed in this category. Some reviewers were very complimentary, saying that these were excellent results in terms of a combination of data and analysis, with a very well-executed research plan. Another said that good progress has been achieved relative to demonstrating the concept, which confirms the high discharge capacities, and rate capability with the baseline chemistry ( $Li_{1.2}Ni_{0.25}Mn_{0.75}O_y$ ) at 2.0-4.95 V. During extended cycling however, the voltage profile indicates increasing polarization. Demonstrated high capacity of ~250 mAh/g and excellent rate capability (>200 mAh/g at 1C rate), when charged to 4.6 V, but only after a few formation cycles. Understood the effects of Li and Co contents on the performance and successfully adopted X-ray absorption spectroscopy and HR TEM to confirm the integrated layered-spinel structure. A third reviewer commented that the results are good and the next step approach makes sense. A fourth complimented the good analytical results in trying to get an understanding, especially the TEM work.

However, some questions and criticisms included the following:

- As a general question, we need to understand how these high-energy cathode materials compare, in terms of performance vs. life vs. safety vs. cost. What is the life of these products?
- Another reviewer felt that it is not clear, from the data presented, on the progress made toward improving the cycling performance of the system. It also seems that there is a strong need to set quantifiable goals, especially as this is an on-going program.
- Work on larger format cells is also needed.
- Results look decent, according to another reviewer, who said that however he was not sure that the material performance is significantly greater than the sum of their parts, except maybe for the rate performance. The project needs to demonstrate material stability, and the hanging profile with even just a few cycles does not sound very promising.

### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Collaborations were judged to range anywhere from excellent to not obvious. A reviewer noted that there are a few collaborations within ANL and outside in the material characterization. However, there are no collaborations with any industry partner to evaluate these materials in 18650 cells, which are recommended, in view of the promising nature of these materials. Another reviewer agreed with the need for an industry partner. The reviewer who did not see obvious collaborations stated that collaboration with groups working on electrolyte is key to success. One reviewer wanted a better explanation as to why a Korean partner was chosen to provide a precursor over a U.S. partner.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

Reviewers briefly noted the plans as being OK and solid, then quickly offered suggestions. The proposed future research is to continue these studies to i) identify the optimum chemistry in terms of Li-to-TM ratio and Co content, ii) develop close collaboration with industrial partner (good TM precursor), iii) investigate thermal safety characteristics, iv) further structural analysis with cycled and uncycled materials, v) explore physical blending of spinel and layered materials, and vi) finally assess in full cells with carbonaceous or advanced anode materials through collaborations.

Suggestions included:

- Please do not use the physical blend of spinel/layered materials, because that will hurt your calendar-life. Otherwise, you have a solid plan.
- A reviewer suggested more emphasis on high-voltage cathodes/sysetms.
- More life and safety data are needed.
- There is a need to set more definitive and quantifiable goals for future work, especially since this is an on-going program.

• A reviewer said there is a need to do the comparison with the simple blending ASAP. Also, look at cycled material to see if it stays an integrated material.

**QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?** Reviewers varied in response to this question. One reviewer said that because it is a cutting-edge material, he would recommend allocating more funds. However, more common was the view that the budget of \$300K per year is adequate for these studies. Suggestions included that working with an industry partner would help, and another reviewer expressed that there is a need to push for

a faster demonstration that the integrated material is significantly better than a simple blend before continuing to fund this work.

# Developing High Capacity, Long Life, and High Power Anodes: Khalil Amine (Argonne National Laboratory)

## **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

# QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Two reviewers were positive about the relevance of this program to DOE objectives, two did not answer, and one reviewer felt that it does not belong as part of a vehicle technologies research program. A favorable reviewer described the program as follows: the specific energies of Li-ion batteries are to be increased for the PHEVs, while the HEVs need improvements in power density. The objective of this project is two-fold, i.e., to develop high power anode for HEVs and high-energy and long-life anodes for PHEVs. Both these anode systems are based on non-carbonaceous anodes, more specifically titanium oxides. These studies, if successful, will lead to the incorporation of high-power anodes and high-energy anodes in Li-Ion cells for enhancing HEVs, and PHEVs, respectively, which would in turn reduce the petroleum consumption, and pave the way towards petroleum replacement.

However, the reviewer who disagreed said that most of the work is on high voltage anodes. The reviewer just did not believe that we can afford the voltage penalty of these materials as it reduces both power and energy and places a larger stress on the cathode (to make up for



the lower voltage both cathode and anode must run at a higher current, not a big deal for these anodes but it stresses the cathodes). Work like this has more applicability to load leveling and other stationary power needs and should not be funded under a Vehicle Technologies program.

# QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

Reviewers had some criticisms of this approach, even if on average they were tepidly positive. The approach for the high power anodes is based on  $Li_2MTi_6O_{14}$  (M=Ba,Sr,2Na) materials, which have lower-potentials (hence high cell voltages) and higher capacities compared to the traditional lithium titanate,  $Li_4Ti_5O_{12}$  and can provide long life and enhanced safety, due to the absence of SEI. The high-energy anodes being developed belong to titanium oxides with a high theoretical capacity of 335mAh/g based on  $Ti_4+/Ti_3+$  couple and new silicon-based composite system with a high packing density, low irreversible loss and long cycle life. This reviewer felt the proposed approaches appropriately address the barriers for HEV and PHEV batteries. Another reviewer agreed, but he also pointed out that there is also need a lower cost material.

Criticisms included:

- This work entails all current avenues of improving the anode. It thus lacks the focus of other similar projects.
- High-voltage anode for energy is more questionable. It will need to be coupled with high-voltage cathode, according to another reviewer.
- The reviewer who objected to the project in the previous category said that all the work on high-voltage anodes is irrelevant to Vehicle Technologies. The recent focus on Si is better.

#### QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL

## PROJECT AND DOE GOALS.

The reviewers described the results themselves, with only one characterizing it in any way (he deemed it "modest"). One reviewer noted that data on several novel materials were obtained, which really shed light on the titanate-type of anode materials. Another reviewer noted that capacity is still low, but rate capability looks great, but the voltage of the Sr material is not very different than titanate.

A third reviewer noted the modest progress that has been achieved relative to the standard LTO materials. Specific accomplishments include synthesis of  $MLi_2Ti_6O_{14}$  anode materials, using sol gel and demonstration of their performance, especially the  $SrLi_2Ti_6O_{14}$  material in half-cells and full cells. The results however do not compare well with those from the LTO anode. For the high-energy anode, nano-structured, high surface area and high packing density  $TiO_2$  Brookite has been synthesized from a thermal decomposition of oxalate and demonstrated to have good cyclability, though the capacities are, once again, lower than LTO. The silicon composite anode, prepared from ball milling showed good cycle life with a moderate capacity of 500 mAh/g, and with a high irreversible capacity.

The dissenting view was that most of the results were not especially worthwhile, although the decent packing of their Si anode was useful; not enough people in this field appreciate the cost and performance implications of low packing.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Only two reviewers commented on the collaborations. There are a few collaborations within ANL and outside in the material characterization, assessment, and with FMC for the SLMP. A different reviewer commented that he would like to see this PI or someone work with Professor Kumta on the Si anode work.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

Reactions to the proposed future research were mixed. A reviewer said that he did not think any of the titanate materials decribed here offer any significant advantages over Li-titanate to merit additional studies in full cells. Instead efforts should be directed at finding materials superior in capacity or slightly lower in voltage. Another reviewer described the proposed future research as to continue these studies on the high-power anode to evaluate  $SrLi_2Ti_6O_{14}$  in full cells, improve its power capability though new synthesis methods and through CNT coating, and study its interactions with the electrolyte. For the high energy-anode, SI composite anode will be evaluated in full cells and with FMC as a Li source to compensate for the irreversible capacity. However, this reviewer did not judge it.

A third reviewer offered the feedback that surface area needs to be decreased. A fourth was highly critical, saying that only the silicon anode work that has a decent potential is worthwhile in view. Higher capacity, high potential anodes leads to a major loss in power and energy, especially when one factors in the impact on the cathode that has to match the higher capacity and increase in delivery current. This is what they stated they plan to do in the presentation. Though the reviewer is glad that they are looking to evaluate the SLMP approach, he cautions about sinking too much effort into yet another carbon-coating method for Si, as there are already ways to do that and many more will be coming out over the next year or two.

### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

One reviewer said that the budget of \$600 k per year (\$300 k for the high power and \$300 K for the high-energy anode) is adequate for these studies.

The only other reviewer who answered this question wrote that he recommended discontinuing funding for the bulk of this program under the Vehicle Technology program. (He was very skeptical in his responses to other questions above). Low-voltage anodes may well be deserving under a stationary power funding mechanism. He instead recommended continuing to look at Si as long as they are really doing something new, i.e., not just another way to carbon coat silicon. This should not be that costly.

# Lithium Metal Anodes: Jack Vaughey (Argonne National Laboratory)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

## QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

All the reviewers approved of this project as supporting DOE objectives, but noted that it is a challenging project. One wrote that if one can find a way to have lithium metal anode work for long life and a safe system, this is great – but this is an extremely challenging task. Another reviewer echoed this notion, saying this is a very challenging project, but if successful it could enable a new class of higher-energy batteries for PHEV and EV applications. For EV applications Li-ion just does not have the energy and we need something like this to succeed to meet the longer-term EV goals.

A project description by another reviewer noted that the specific energies of Li-ion batteries need to be increased for meeting the range requirements of the PHEVs and the objective of this project is to develop metallic Li anodes in place of carbonaceous intercalation anode for enhancing the energy density of Li-Ion cells. These studies, if successful would lead to a successful Li-ion battery for PHEV, which in turn reduces the petroleum consumption, and pave the way towards petroleum replacement.



# QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The approach was deemed very good by a reviewer who commended the very good microscopy work. Another said he techniques used to characterize the Li surface are state-of-the-art. However, the approaches used to alter the Li surface are not new, and that the authors should take rather bold and innovative approaches to come up with breakthrough results and not follow approaches which have already been tested by many authors. This is a 40-year-old topic. The approach involves a study of Li morphology to understand the morphological evolution upon cycling and to develop and characterize different coating technologies, including polymeric and ceramic coating, that robust during cycling in Li cell environments. The approach is appropriate to the stated objectives. One reviewer pointed out that one of the main issue is dendrite growth and wondered how this approach would solve that issue.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Reviewers generally commended the results, noting different aspects. The analytical results are cutting edge and quite impressive, according to one reviewer, who then added that data obtained to stabilize the Li surface are just fair, since the approaches used have been rather well-known (polymeric coatings etc) and the authors did not succeed in obtaining results that are not expected (weakness of polymer coatings, or even silane coating). Another reviewer noted good progress has been achieved in understanding morphological changes on Li upon cycling, i.e., the formation of dendritic subsurface layer which grows during low-rate cycling, which is covered by a porous mid-layer that grows during mid-rate cycling. It also accounts for the volume expansion. The coating studies reveal that that controlling the ability of the electrolyte solvent to reach the lithium surface while maintaining ionic conductivity is a key synthetic variable Amongst the coatings examined, Zintl coatings are too brittle, polymeric coatings are unstable, and the silane coatings not adequately protective.

A third reviewer complimented the nice study on the impact of the current on the failure mechanism. Unfortunately, that's not very optimistic for automotive applications. He also noted that it seems that the coating is a difficult approach because of the inherent reactivity of the Li. What about the new electrolyte approach?

The fourth reviewer said that the finding about loss of solvent in the dendrite growth process is an important new insight. He also said that it was nice SEM work, although he thought many of these features have been seen in many, many previous published studies on lithium plating in organic electrolytes.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

A reviewer said there are a few collaborations but suggested a much wider collaboration due to the complexity of the system. Another reviewer said that the current collaborations were OK in terms of doing the work, but he felt they are missing opportunities. They should be working with Ohara who made coatings for PolyPlus's protected lithium anode. Also, the reviewer said he believed that John Kerr worked with someone on using polymer electrolytes and also got great images of lithium dendrites growing through these films. The reviewer felt that Kerr should also be collaborating on this work.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

Reviewers were mixed about the plan. One said that the plans seem OK, but he thought they need to get help from other collaborations as he outlined above. Another noted that this is a high pay-off research and the authors should take as unconventional an approach as possible to come up with novel ideas. He was not sure the proposed work is different enough to generate any new solution to this very important electrode work. A third reviewer said that he was not sure how working on a Li anode will really help the automotive industry (but acknowledge he may be wrong). Another question was whether we are really sure that we will find a solution to the more important issues of Li's safety concerns?

The proposed future research is to continue these studies to investigate the surface layer on Li with different coatings, cycling rate, and other additives, understand the effect of solvent on the protective nature of the silanes, evaluate the stability of ceramic lithium ion conductor in the cell environment and study the interactions between buffer layer and ceramic layer.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

Three reviewers did not answer this question, but of those who did, one reviewer said the budget of \$300 k per year is reasonable for these studies. The other reviewer recommends continuing to fund this work, acknowledging that it will be very slow and many never in fact succeed. He views this as a high impact/low LOS program, and believes that the DOE needs to fund quite a few of this type of project in order to really make a difference in the long run.

# Improved Methods for Making Intermetallic Anodes: Andrew Jansen (Argonne National Laboratory)

## **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

# QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Reviewers agreed that this was a relevant project that supported DOE objectives. One reviewer said that high-energy, low-voltage, and safe Li-ion negative electrodes are definitely needed for the success of batteries in the automotive industry. A second reviewer agreed, saying there is a critical need for a low-cost anode that meets the life and performance requirements for PHEV and EV applications, and that this work is geared toward identifying an anode that will/may meet those needs. Another reviewer built upon this, saying the specific energies of Li-ion batteries need to be increased for meeting the range requirements of the PHEVs and the objective of this project is to develop inter-metallic Li alloy anodes, especially Cu<sub>6</sub>Sn<sub>5</sub> in place of carbonaceous intercalation anode for enhancing the energy density of Li-Ion cells. These studies, if successful, would lead to a successful Li-ion battery for PHEV, which in turn reduces the petroleum consumption, and pave the way towards petroleum replacement.

# QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL



#### BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The reviewers generally approved of the approach, though many offered criticisms. One reviewer said that Jansen's approach is to limit electrode expansion of the alloy anode by working on binder or material synthesis. He felt the approach is good since the main challenge today is volume expansion. But in addition to that we need to work on limiting the volume expansion. Even if the electrode can accommodate the volume change, an electrode that expands by more than 100% is not practical. Another reviewer who was positive said that the approach to reach the stated goals of the project is sound, however, he was not clear on why a smaller size particle (< $0.5\mu$ m) was not requested from supplier, based on the identified 0.2-µm particle size in Huggin's model. Another reviewer liked the use of Huggins work but it seems that a theoretical understanding of how doping is likely to impact toughness, modulus etc. could be helpful - but he is not sure whether such a theory exists. However, he liked the paper estimate of what an actual battery would look like, and said that there is a need to do this more in other aspects of the program. The PI has a very good appreciation of the practical issues in getting from a material to a real battery.

Other reviewers were more tepid. One said the approaches are just fair and it is surprising that the authors spent so many resources on binders and electrolyte additives. These approaches are not going to solve the innate material issue. Another reviewer noted that the approach involves developing alternative methods of electrode fabrication with suitable binders and appropriate particle size for the intermetallic alloys, in particular  $Cu_6Sn_5$  that was developed earlier in the BATT program. This alloy provides high volumetric energy, but hasn't shown any compelling performance in the specific capacity to merit continued study /development. It will not truly address the technical barriers for intermetallic alloy anodes.

Another critic said that he would have liked a fresh look at new materials rather than an engineering-type study of ANL's CuSn anode, and said it seems a very limited approach to a very big area.

## QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL

## PROJECT AND DOE GOALS.

The reviewers were lukewarm about the results. One said that work related to characterizations and understanding of the strain/stress properties of the materials is certainly appreciated. However, the actual data for solving the key task of stabilizing the anodes are not exciting and kind of expected. Another noted that volume expension is still very high and difficult to handle in the cell. Another reviewer added that there appears to be a return (from last year) to the view that a binder that allows for extreme expansion, or alloys that prevent that expansion will be the solution. The appropriate particle size and process issues still remain concerns, as apparently they have not been fully identified. Reaching these decisions appears to be taking longer than originally planned. The Wildcat collaboration may help get the results faster.

More positively, reasonably good progress has been achieved in: i) understanding the mechanical properties of intermetallic alloys based on critical particle size, ii) identifying the metal suppliers, iii) developing coating processes relative to the conductive and resistive additive and in optimizing the binder.

Finally one reviewer allowed that while acknowledging that this is a very challenging high-impact/low LOS program, results to date are disappointing.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

One reviewer said there are only a few collaborations; it is probably worthwhile to collaborate with an industry partner to exploit the high volumetric energy density of these materials in 18650 cells. Another suggested that collaboration with a binder supplier should be initiated. A third thought that involving LBNL, for example, might open up the work to other anodes.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

One reviewer wrote that the proposed future research is to continue these studies with small particle size materials containing various metals in a combinatorial way (with Wildcat Discovery) and revisit the critical particle size vs mechanical properties topic as well as reexamine the influence of binders and electrolyte additives. It is worthwhile to compare the final energy density against graphite, as is being planned here. These materials will probably address the volumetric energy needs of anode, but fall short of the gravimetric requirements.

Several reviewers offered suggestions for going forward:

- The authors should move away from the current approaches of using binders and electrolyte additives to counteract this fundamental material issue. Please focus more on altering material chemistry, morphology etc.
- Cost will need be lower than graphite to be competitive.
- Needs to do a lot of work on volume expansion and/or volumetric energy to be competitive with carbon.
- What about thermal stability and abuse tolerance?
- A reviewer liked the use of Huggins work but it seems that a theoretical understanding of how doping is likely to impact toughness, modulus etc. could be helpful, but he is not sure whether such a theory exists. He suggests talking to someone like Gerd Ceder about this, a better fundamental understanding of how dopants could impact the physical properties could be a big help in directing the synthetic work.
- The reviewer would like the scope widened to cover other materials, and he thinks this program needs to take a step back and see what other materials they should look at.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

Only two reviewers answered this. One said the budget of \$300 k per year is appropriate for these studies. The other said that the effort is OK, but six years seems too long to fund this work without seeing more progress. Again, he would like to see work on something other than ANL's CuSn anode material.

# Novel Electrolytes and Additives: Dan Abraham (Argonne National Laboratory)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 6 reviewers.

## QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Reviewers deemed this to be very relevant and a critical area of research, with a couple of caveats. The specific energies of Li-ion batteries need to be increased for meeting the range requirements of the PHEVs. While there is a need to develop high-energy cathodes (and anodes), there is concurrent need to develop suitable electrolytes that afford safety and stability to the Li-ion cell, which is the objective of this task. These studies, if successful would lead to a successful Li-ion battery for PHEV, which in turn reduces the petroleum consumption, and pave the way towards petroleum replacement.

However, the caveats were that the stability needs to be higher than 50°C, and a different reviewer felt that though the program is trying to address cycle and calendar life issues, he does not believe the program is well focused to do this, as he outlines in a later section.

# QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?



Reviewers were generally critical of the approach. A positive reviewer commented that there were a number of attractive ideas being pursued to develop the next generation of electrolyte.

More commonly, the comments were critical. A different reviewer said that he didn't see any attempt in pushing the voltage to 5V. Even at 4.4 V the high fade rate in 4-5 cycles was pointing that we are not on the right path. Another reviewer stated that the reasons for the choice of glycol carbonate need to be better explained. It is not clear why that family of product has been chosen: thermal stability, cost, or what other reason? What are we trying to improve here? A third reviewer built upon this concern, saying that he has deep reservations about the approach to finding additives. What's the basis for trying to develop materials? I fear it may become a black hole that will suck up resources in a glorified fishing expedition among the 4 million+ organic compounds out there. If there was a basis for designing an additive I would feel more comfortable, but otherwise it would seem a never-ending, almost hopeless task.

A different reviewer noted that electrolyte is indeed a key component for the performance and safety of Li-ion cells. However, it not clear what advantages the GC-based electrolytes provide over the conventional EC-based systems.

A reviewer suggested that ionic liquids (IL) need to be looked at, but a couple of fundamental points need to be addressed in this area: low temperature performance and the problems of venting the cell in case of failure to essentially shut down the cell. This is an important safety feature in consumer cells and a high boiling point fluid may not be expelled well if the cell vents. Also ILs are hard to purify and Li-ion cells are exquisitely sensitive to impurities, so these need to be at least measured even if they can't be controlled very well.

### QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL

## PROJECT AND DOE GOALS.

Reviewers noted that much quality test data has been generated, but there are still concerns. One said the project has generated an immense amount of quality test data, some of which are novel and should lead to improved electrolyte system. Another reviewer agreed that some of the results looked OK, but usually this was only a partial data set. He was not sure that any of these materials are going to be useful overall. The use of differential capacity curves was nice, but the data do not suggest these materials are especially stable. My main concern is that I did not see any theory or experimental insight as to how these additives are supposed to work their magic.

A third reviewer characterized this as reasonably good progress accomplished in terms of: i) assessing the performance of graphitic anodes and metal oxides cathodes in GC (and its methyl ester version)-based electrolytes solutions, ii) identifying a couple for improved capacity retention and iii) examining a couple of ionic liquids based on pyrrolidinium, which performed well in half-cell, but not in full cells. Despite this progress, it is not clear to me what the GC-based electrolytes can offer. What is the rationale behind GC-based solutions except for being new?

Finally, a last reviewer said that he was not convinced about the explanation of GC oxidation that depletes the lithium reservoir. Based on the curve it seems to be more a reduction problem on the negative electrode than an oxidation on the positive electrode. What is the water content of this electrolyte? What about abuse tolerance? DSC?

**QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?** Reviewers commended the collaborations with universities and other laboratories in this effort.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

Reviewers had plenty of advice for the team. This included:

- The authors should carry out an exhaustive literature survey to minimize/eliminate redundant studies.
- Please pay attention to well-known functional groups such as acetates, ethers, etc., which have not proven to be stable enough for use in Li ion batteries.
- They need to include some thermal stability tests.
- I would suggest tearing down cells from the previous testing and looking in detail at the electrolyte and the electrodes to better indentify the failure mechanism (for example, where is the lithium lost). That can be done in collaboration with other labs offering diagnostic capabilities.
- The proposed future research is to i) continue studies with GC-cased electrolytes in terms of their electrochemical stability, SEI formation characteristics and electrochemical performance, ii) develop new electrolyte additives for stabilizing the electrode surfaces, and, iii) explore the use of ionic liquids. The proposed research is related to the overall project goals and the DoE objectives. The relevance of GC-based solutions is still unclear.
- A reviewer was not convinced that the glyercol additives are really that good. The team needs to develop an understanding of what you want additives to do and then design the additives (this means writing down proposed reactions, not just the desired results).

## QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

One reviewer stated that the budget of \$300 k per year is appropriate for these studies. However, another reviewer said that he has huge reservations about this work because of the lack of a clear understanding of how they expect these additives to work. Industry can and does do this much better and faster than DOE labs. Also, major issues with the Ionic Liquids need to be addressed. Consequently, this reviewer has to recommend that the DOE stops funding this work, at least until the issues raised can be addressed. Three reviewers did not answer this question.

# Electrolytes in Support of 5 V Li-ion Chemistries: Richard Jow (Army Research Laboratory)

### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

# QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Reviewers unanimously agreed that this project is highly relevant to the DOE's objectives. One reviewer described it as follows: Highvoltage cathode materials are being developed to increase the specific energy of Li-ion cells to meet the range of PHEVs. Such cathodes would need electrolytes that are stable at these high (5 V) voltages. The objective of this task is to develop high-voltage electrolytes that enable the dreamed-of 5-V Li-ion chemistry. These studies, if successful, would lead to a successful Li-ion battery for PHEV, which in turn reduces the petroleum consumption, and paves the way towards petroleum replacement. Two other reviewers added that the use of a higher voltage system should reduce the number of cells needed for a system and consequently reduce the cost. Since cost is a key to moving the PHEV and EV technology forward, this project supports the overall DOE objectives.

# QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?



Reviewers were positive about the approach, which involves developing new electrolytes based on i) unsymmetrical and unsaturated sulfones to be used a co-solvents or additives, ii) carbonate-based solutions with selective co-solvents and additives. A reviewer said that these approaches look quite reasonable, since sulfones are oxidatively stable at 5 V and the carbonate systems may be stabilized with proper additives that will form surface films on the cathode. The approaches address the technical barriers adequately. Another reviewer noted that the program seems to have the right approach by looking at key electrolyte families, isolating the issues associated with each, and determining the viability of eliminating those concerns. A third said that it is good to see high-voltage electrolyte studied, because this type of work is absolutely needed to make the high-voltage cathode application possible.

A concern is that the team will need to confirm compatibility with the other cell components. A last reviewer commented that this program is very challenging. Their approach of trying to use the sulfones is not bad, but they are known to be very extremely viscous, so low temperature would likely be very poor. The use of substitute materials for true 5V cathodes is good. He is also a little concerned about the "additive" approach unless there is a clear understanding of what the additives actually do - in detail. Basically, if one can design an additive to do something specific, then he is OK trying it out, otherwise this can turn into a fishing expedition and suck up valuable resources. But the group does seem to have some good ideas for what they want in an additive.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Reviewers commended the ARL additive results, but were mixed about other aspects of the project. One reviewer said that the ARL additive data look promising and provide very good clues as to the future directions for work. The current generation is definitely superior to conventional electrolyte systems, but he did not think they are stable enough yet to meet the demands of the 5V cell system

especially when data at high temperature have not yet been obtained. Another reviewer who commended the ARL results said that the team needs to look at the surface to see if there is any passivation film formed.

A third reviewer noted that good progress has been accomplished in terms of developing high anodic-stability electrolytes with sulfones. After evaluating several formulations ( $\sim 80$ ), it was concluded that the sulfone is unsuitable as a major solvent due to the issues connected with SEI on the anode, ion transport and electrode wetting. However, significant progress has been achieved in carbonate-based solutions, as demonstrated in cells with a spinel cathode. The progress achieved here is consistent with the DOE goals. A fourth reviewer also recognized that the project seems to have narrowed the test matrix by eliminating sulfones as an option. He mentioned the need to identify a collaborator for the next phase of work using the carbonate approach.

Concerns included the thought that it is not clear what the expectations/goals are for the program. Cost-reduction potential should be included in the goals. Also, low-temperature performance is a big problem with this chemistry, so LT performance improvement should be included as a concern to be addressed. Another criticism by a different reviewer was that the additives results look interesting, but he did not see any experimental understanding of how they worked, which bothers me.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

All reviewers noted the collaboration with ANL. One reviewer wondered where is the work with ANL high-voltage material, and another noted that he sees the same electrolyte being developed also at ANL (glycerol carbonate). One reviewer advised that in order to get at a real understanding of what these additives actually do, they may need more help from the other DOE labs. There is also a collaboration with a university and industry.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The proposed future research is to continue the i) Optimization of carbonate-based electrolytes, focusing on performances at elevated temperatures, ii) Studies on the characterization and diagnostics of the electrodes cycled with these electrolytes and iii) the design and development of new additives for improving performance of 5V cathodes.

One reviewer advised that while we do not know the nature of the ARLs, he thinks the work should continue in that regard because of the initial data. Another reviewer expanded upon this notion, saying he would focus on the ARL additives for now and especially in terms of understanding how they work. In fact, this understanding may prove more valuable than the additives themselves as it may direct future work in designing additives.

Other suggestions were:

- Characteristic and diagnostic studies are critical.
- The team needs to include calendar life tests at high temperature.
- There should be some planned work to establish low temperature performance.
- There should be a set of performance goals.
- There was very little cycle and calendar life work shown using existing additives.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

Reviewers supported this work, as one stated that he believes this is very important work that needs significant funding. Another reviewer said the budget of \$200K per year looks insufficient for these studies, but may be justified if leveraged from DOD. Two reviewers did not answer this question. A third reviewer also supported the research, saying he remains concerned about the approach to "magic" additives. However, he agreed that they need to follow up on their promising findings, but only as long as they address the fundamental issue of what they are actually doing. If they can provide additives that have a clear basis as to why one would expect them to work and how, this reviewer is fine with continuing to evaluate new additives. However, the work should be focused. Funding level is quite modest though and should be continued at the present level.

ENERGY Energy Efficiency & Renewable Energy

Development of Advanced Electrolytes and Electrolyte Additives: Zhengcheng Zhang (Argonne National Laboratory)

### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

## QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Reviewers were supportive of the relevance. High voltage cathodes materials are being developed to increase the specific energy of Liion cells to meet to the range of PHEVs. Such cathodes would need electrolytes that are stable at these high (5 V) voltages. The objective of this task is to develop high advanced electrolytes with high voltage stability, combined with high lithium ion conductivity, high thermal stability, non toxicity and non-flammability and also to identify electrolyte additive that provides a stable SEI on the electrodes for improved cycle life. These studies, if successful, would lead to a successful Li-ion battery for PHEV, which in turn reduces the petroleum consumption, and pave the way towards petroleum replacement.

One reviewer felt that ES024 and this program should be combined in some way.

# QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL



#### BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

One reviewer said that the approach with sulfone has the potential drawback of poor low-temperature performance. Ionic liquids will have the problem of stability. Another reviewer described the approach as involving developing new electrolytes based on sulfone as a primary solvent or co-solvent, and ionic liquids, and investigating their compatibility with different battery chemistries. In addition, various electrolyte additives including compounds containing oxalic group, ester group, vinyl group et al. will be examined for their "filming ability" on the cathode surface. The approaches are thus aimed at developing stable and safe electrolytes for Li-ion batteries to enable their widespread use in PHEVs.

A different reviewer noted the approach in each area looked quite good, although the known viscosity problems of sulfolanes are likely to be very difficult to overcome at even modestly low temperatures. No low temperature work on this seems to have been planned.

Other reviewers had these questions and comments:

- Are these compounds the same as ARL?
- Why are they using LiTFSI, as it is well known to oxidize Al. The project needs to find another salt.
- Generally, a reviewer was concerned about work on additives without a clear rationale for evaluating them. They seem to be basically picking up on leads from others, which is better than just "fishing," but have one's own rationale to design additives would be better.

## QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL

## PROJECT AND DOE GOALS.

Reviewers lauded the results thus far. One said that some of the additives look promising and this work should continue. Another agreed that there are quite a lot of very promising results. The LTFOP data at 55°C are impressive, and he hopes the authors can build on this success to develop the next generation of these additives/electrolytes.

A third reviewer said there is good progress accomplished in terms of developing high anodic-stability electrolytes with sulfones. The team has determined the wide electrochemical window and conductivities of trimethyl sulfone and ethyl methyl sulfone and demonstrated good cycling performance with spinel oxides in both TMS/ LiTFSI and EMS/LiTFSI electrolytes. They have also identified several new compounds with oxalic group as SEI formation additives and demonstrated their beneficial effect on both NMC cathode and MCMB anode at 55°C. They identified that succinic anhydride and maleic anhydride can form stable SEI earlier than EC, which might have several benefits. There is thus good progress towards the project and DOE goals.

Some advice and other feedback included the following:

- Someone mentioned that LiTFSI could work. Some corrosion and life studies are needed to prove it.
- The electrolyte work does not seem to be bearing much fruit. Wettability and viscosity issues with sulfolane were predictable based on its well-known properties. They may want to look a separator with surfactants such as Celgard 3501, if Celgard still makes it. The flammability comparison is flawed because of the different solvent ratios one can't tell if the difference is due to the replacement of EC with Sulfolane or the fact that the EC formulation simply had a higher content of the much more flammable EMC.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

The reviewers noted that there are a few collaborations within ANL and none yet outside ANL. It was suggested that they collaborate with industry partners as well.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The proposed future research is to continue investigating i) sulfone-based electrolytes to make them compatible with graphite anodes, ii) ionic liquids as new electrolyte solvents by screening existing electrolytes and develop new systems, and iii) electrolyte additives, in particular, succinic/maleic anhydride additives for thermal and morphological studies. These studies address overcoming the barrier outlined for this project, in the judgment of one reviewer. Another reviewer would propose evaluation of these materials at low temperatures also. A third reviewer was more critical, saying that he was not convinced that the sulfolane work justifies continuing and Ionic Liquids also have major issues with low temperature. Questions should also be asked about the acceptability of a highly non-volatile electrolyte as this would likely not effectively shut down the cell if it were to vent during abuse or malfunction. Only the additive work looks promising to me.

### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

Three of the reviewers did not answer this question. One reviewer said the budget of \$300 k per year looks reasonable for these studies.

The final reviewer commented that the electrolyte work is not looking very promising, and even if they succeeded with sulfolanes or Ionic Liquids, the reviewer was not sure they could be used in practical cells due to low temperature and venting issues. The reviewer would scale this back to just look at the additives unless they can show a clear path to a viable high voltage electrolyte. The reviewer did not believe they have great expertise in electrolytes; basically the reviewer just questioned whether they are the right group in the DOE to be doing the electrolyte work.

Development of Novel Electrolytes for Use in High Energy Lithium-Ion Batteries with Wide Operating Temperature Range: Marshall Smart (Jet Propulsion Laboratory)

Energy Efficiency &

**Renewable Energy** 

#### **REVIEWER SAMPLE SIZE**

U.S. DEPARTMENT OF

NERGY

This project had a total of 4 reviewers.

## QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The first reviewer saw that this project is a critical component for long-life cells. The second reviewer perceived that barriers addressed at the electrolyte level are in line with DOE objectives, and life is clearly one of the main limitations today. Another reviewer said that this project in particular attempts to address the low temperature performance of the lithium ion chemistry without negatively impacting the high temperature performance.

The fourth reviewer saw that obviously JPL has an even stronger interest in low temperature cells than DOE, but this program does support the DOE goals, especially if low temperature power is really a design criterion. However, this reviewer is not convinced that the - 30°C operation makes much sense, especially for initial market penetration and with the options to use heaters, engine power, etc.

# QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

One reviewer saw that many well-proven materials are being tested. The second reviewer felt that the approach is good because it combines fundamental work and studies in coil cells with evaluation in prototype cells. The third reviewer commented on the excellent approach to solving clearly defined objectives. Another reviewer liked their methodology a lot, including the use of harvested electrolyte and Quallion cells. The research generates a lot of actual test data that gives a good sense of confidence in the findings.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Several reviewers commented positively on the data. The first reviewer saw a great collection of data. The second reviewer noted that obviously a lot of work has been done with some nice results: overall, this is a nice work. Another reviewer felt the program met program goals of improving low temperature performance. The fourth reviewer saw good data and noted that some of the low temperature and ambient cycling results are quite impressive. The evaluation of additives is well thought out and showing some progress.

One reviewer was a little worried about the stability of these solvents at high temperatures. Another reviewer commented that it was a bit difficult to follow which of the results were the most promising. Also, some the results did not appear to be brand new. The third reviewer noted that high temperature performance did suffer. The fourth reviewer feared that these esters will not be able to provide good cycle or calendar life – JPL's own work shows problems at high temperature. This reviewer is not convinced that any amount of additives are going to fix this, and noted how Panasonic stopped using methyl butyrate in their cells a long time ago.



#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

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NERGY

Energy Efficiency &

**Renewable Energy** 

Reviewers had positive feedback about the collaboration. Specifically, the first reviewer was impressed by how everyone was really pulled together by the project team. The second reviewer felt that this seems to be a collaboration with lab and industrial partners. The third reviewer commented that many collaborators were used from various groups including different commercial battery suppliers: this is good, as it shows a wide range of interest from the industry in this program and the technology. The final reviewer noted that there was a "whole load" of impressive collaborators, and the team is also feeding work into modelers.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

One reviewer felt that the program identified good potential solutions and proposes to build on that work and involve a wide range of battery suppliers by incorporating the electrolytes into their commercial cells. A second reviewer felt that plans going forward are good. The reviewer had concerns about the likelihood of ensuring adequate stability with the esters via additives, so this reviewer suggests much more emphasis be given to cycle life and high temperature testing. For the DOE program these are critical features and cannot be traded to get better low temperature performance (there may be room to do that for some of NASA's applications).

Another reviewer saw a definite need to wait for high temperature stability before a judgment can be made about these electrolytes. A third review saw that extreme temperature stability of esters has always been an issue. If it is believed this is the direction to go, this needs to be the primary focus for future research.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

A reviewer said that annual funding seems low for this work, and this reviewer would like to see it doubled if and only if -30°C is really a "must have feature," which this reviewer seriously questions. Otherwise, this reviewer suggests keeping the funding level as it is.

**ENERGY** Energy Efficiency & Renewable Energy

Novel Compounds for Enhancing Electrolyte Stability and Safety of Lithium-ion Cells: Kevin Gering (Idaho National Laboratory)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

## QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The first reviewer commented that this project is relevant. A second reviewer also felt the project was relevant, and that working on improving abuse tolerance and cell life is the way to go. Another reviewer commented that both safety and longevity of Li-ion cells are inadequate for the current Li-ion cells, and this gets more challenging with the advent of high-voltage cathodes. The objective of this task is to develop new electrolytes based on novel solvents and on phosphazene additives. This would require an understanding on the effects of phosphazene additives on the other electrolyte components and on the SEI characteristics, as well as their tolerance to high and low voltages, which will be the topic of this study. These studies, if successful, would lead to a successful Li-ion battery for PHEV, which in turn reduces the petroleum consumption, and pave the way towards petroleum replacement.

The fourth reviewer commented that improved safety is key for this technology to be adopted into the vehicle market. A key goal of this program is to improve the electrolyte, which contributes to most of

the safety issues. The final reviewer felt that safety is a critical area and this program represents an attempt to make the electrolyte non-flammable.

# QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The first reviewer felt that the project seemed to be aware of previous work by Bridgestone and others, and the team has access to experts on these materials. The project is looking at the flammability and performance together, and the reviewer likes the approach in general. The second reviewer commented that the program has selected one group on which to concentrate activity and this narrows the focus. The reasons for the choice are clearly outlined and understood.

The third reviewer noted how the project utilizes the expertise at INL on the phosphazene chemistry, and how the approach involves synthesis and characterization tests such as transport properties, flammability, and so forth, with electrochemical testing (in Li-ion coin cells) of a series of phosphaznes. These will be blended in different proportions with the carbonate solutions. Candidate electrolytes will be tested in LiCoO<sub>2</sub>/graphite coin cells. One difficulty with this wide range of proportions for the phosphazenes is that it is unclear if the phosphazene is being utilized as an additive, co-solvent or primary solvent. The considerations are different for each application. The approaches are thus aimed at developing alternate and safe electrolytes for Li-ion batteries to enable their widespread use in PHEVs.

Another reviewer felt that the approach is good overall, but it is not very clear why phosphazene is the right chemistry, when it has "already failed." There is probably good justification of that, but this is just not very obvious from the presentation. The final reviewer



was not sure this approach will work at all. The flashpoint did not increase, the substance is viscous, and it might be needed to be added in significant quantities to bring about any gain in stability (but will certainly be at the expense of power).

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

A reviewer saw good progress has been accomplished in terms of evaluating various (about nine) phosphazene-based electrolytes in carbonate blends. Some of the effects of the phosphazenes are : i) to decrease the conductivity due to higher viscosity, ii) increase the SEI characteristics at the anode, probably causing higher interfacial impedance, and iii) providing moderate stability at the cathode and iv) overall causing considerable polarization over baseline. The CVs indicate considerable oxidative activity around 4.0 V, which suggest that they could only be used as additives, that too if the oxidation leads to a stable surface film on the cathode. In general, it is critical to determine the stability and compatibility of the phosphazenes at both the electrodes of Li-ion cells, before undertaking on an extensive study as this. The discharge capacity should be expressed as specific capacity to get an insight into the electrochemical compatibility.

Another reviewer felt that given the approach, the project has achieved some good characterization of the materials in question. This reviewer is very skeptical about any positive outcome of the project, as too many drawbacks are already coming to the surface. In fact, this reviewer feels that the original Bridgestone data were also not appealing to begin with from application point of view.

The third reviewer observed nice progress on developing additives with low viscosity, but conductivity is still affected. There is no change in flash point but the team needs to understand the true impact on abuse tolerance. This reviewer asked how much lithium is consumed during the first cycles. (What is the capacity loss?)

Another reviewer felt the work was good, but the overall results did not show any improvement in a key goal of the program-reduced flammability of the electrolyte. Relative to the stated benefit/strength of this additive, very little test data was presented to support the improved safety. It is good to know that cost is being considered as a concern for this process early on.

The fifth reviewer commented that the large amount of additive needed to reduce the flash point looks "pretty ugly" and was one of the problems with the original Bridgestone material. Good work, but the results to date don't look very encouraging.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

The first two reviewers saw very good collaboration, and the third reviewer commented that there are good collaborations with various DOE laboratories and with industry partners. Another reviewer felt that industry support/involvement is encouraging that the process has a great deal of promise.

The fifth reviewer saw good linkage with INL and SNL in the program. However, this reviewer adds, the team needs to talk with Sandia more about their battery test of Bridgestone's materials and devise meaningful tests going forward. Also, this reviewer suggests speaking to the US Navy, as they are very knowledgeable in this area and are currently doing aerosol flammability testing on all electrolytes used in their batteries.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first reviewer felt that future plans show that the issues identified will be addressed. One of these issues is determining the % level of phosphazene compounds that will improve flammability and what the resultant viscosity level will be. The second reviewer commented that if the authors can synthesize lower viscosity analogs, there might be some potential.

Another reviewer noted how the proposed future research is to continue investigating i) electrolyte systems having mostly phosphazene solvents, especially, non-cyclic phosphazenes with low viscosity, ii) selected phosphazenes as additives in volatile organic electrolytes for reduced flammability, and iii) the SEI characteristics (both on the cathode and anode) with the phosphazene

additives. Candidate solutions will be tested for abuse tolerance at SNL. This reviewer felt the proposed studies are consistent with the project and DOE goals.

The third reviewer commented that abuse tolerance is very important, as it is the main result expected from these compounds. To have good understanding of abuse tolerance, this needs to be done on large cells.

The final reviewer expressed some concerns about metrics for flammability. The PI needs to talk with Sandia more about Sandia's past battery test of Bridgestone's materials and devise meaningful tests going forward. This reviewer thought flash point is a pretty decent measure to start with, but it doesn't really address aerosol safety. The U.S. Navy is very knowledgeable in this area, and they are currently doing aerosol flammability testing on al electrolytes used in their batteries. This reviewer strongly suggested that the PI and Sandia get with the Navy to see what they can learn from each other. There is also an industry standard flame test measuring a burn length of a fabric-soaked cloth, although this reviewer does not like it. However, in that work (mostly in patents), a big benefit was seen from the LiPF<sub>6</sub> alone. Just a caution that going to a different salt could be a step backwards in their search for low flammability.

### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

The budget of \$400 k per year (last year) looks reasonable for these studies, according to one reviewer. According to the second reviewer, the funding level seems low, but this is a low LOS high reward program and maybe that's appropriate at this stage. If they get anywhere, then significant additional funding would be needed to capitalize on their work.

# Screen Electrode Materials & Cell Chemistries and Streamlining Optimization of Electrode: Wenquan Lu (Argonne National Laboratory)

## **REVIEWER SAMPLE SIZE**

This project had a total of 6 reviewers.

## QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The first reviewer commented that the stated objective of this project is to identify low-cost materials for the systems used in PHEV battery systems. The reduction of the cost of these systems is absolutely necessary for acceptance of this technology and consequently supports the DOE objective.

Another reviewer commented that with several advanced materials being developed and marketed by the vendors, it is essential to have their performance assessed for comparison in standard test vehicles and environments. The objective of this task is to identify and evaluate and understand the low-cost cell chemistries that can potentially meet the life, performance, abuse tolerance, and cost goals for PHEVs. Another objective is to understand the impact of the material physical properties and electrode formulation processes on the electrode performance. These studies, if successful, would lead to a successful Li-ion battery for PHEV, which in turn reduces the petroleum consumption, and paves the way towards petroleum replacement.



The third reviewer commented that evaluating the best materials available provides a valuable benchmarking feature to the program. Moreover, the fundamental work on particle conductivity and BCF electrodes provides greater understanding of how best to use and evaluate these materials.

According to the fourth reviewer, the stated goal of 100 Wh/kg (pack level) seems too low. Their understanding is that the pack level goal is 200 Wh/kg. The current technology for large format cells is already close to 100 Wh/kg pack and 150Wh/kg cell-level.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL

### BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

According to one reviewer, the project is working with the right materials and a good evaluation methodology. This reviewer liked the work on fundamental aspects of making an electrode. The second reviewer liked the creative approaches to measure particle conductivities. The team should focus on how we can use this to make high energy and/or high power cells. The reviewer recommended examining Robert Kostecki's LBNL work where he shows that regional isolation of active material results in electrode/cell premature fading of power and energy. How can we use this knowledge to make better electrodes?

The third reviewer summarized how the current approach is to carry out a detailed assessment of various cell components, i.e., specifically high energy density LiMnFePO<sub>4</sub>,  $Li_{1.05}(Ni_{4/9}Co_{1/9}Mn_{4/9})_{0.95}O_2$  from ANL, SMG graphite from Hitachi Chemical, and fluorinated electrolyte (FEC) from Daikin. The approach for the second objective is to study the distribution of the conductive additive and determining the effect of carbon coating and binder on the electronic conductivity. According to this reviewer, the approaches are thus aimed at evaluating the commercial materials and also understanding the electrode fabrication issues, as stated in the objectives.

Another reviewer felt that the approach allows for a broad level of investigation. There should be some constants identified at the cell and chemistry level to reduce the potential number of materials studied and to provide more useful information sooner. This reviewer thought it might be better to identify two or three chemistries and concentrate on the materials of two or three of the cell components.

The fourth reviewer was not completely clear as to the scope of this project. Are the team members evaluating materials or optimizing cell fabrication steps? In any case, the work is well organized and thought out.

For the fifth reviewer, the plot of capacity is valid for one type of material only. The working voltage will change that plot. That approach is what current battery suppliers are doing. Not sure about the added value. DOE should support work that lead to innovative solution. This reviewer is unsure if material screening needs to be a DOE funded activity. The second part of this work is much more valuable.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Several reviewers had positive feedback on the project's progress. The first reviewer saw that several important results with respect to characterization/optimization of the electrodes have been reported. These are very valuable to develop well-behaving cells for ANL and other labs. The second reviewer commented on the good work on various aspects of cell design and development with clear conclusions. It would be nice to tie up all the loose ends. E.G. insights on how to match electronic conductivity of electrode with the ionic conductivity to get optimum electrode ASI, or measuring particle conductivity and binder conductivity, etc. are all good. But now all the bits and pieces of information need to be tied together.

The second reviewer felt that good progress has been accomplished in terms of completing the evaluation of the selected materials, which showed interesting characteristics. Specifically, i) LiMnFePO<sub>4</sub> with high capacity of ~160mAh/g at 4 V with ~80% capacity retention at 2C rate and good cycling performance, ii) ANL's Li<sub>1.05</sub>(Ni<sub>4/9</sub>Co<sub>1/9</sub>Mn<sub>4/9</sub>)<sub>0.95</sub>O<sub>2</sub> with high specific capacity of 180mAh/g and low irreversible capacity loss of <10%, iii) the carbon coating in Hitachi's SMG improving reversible a capacity decreasing the irreversible capacity (SEI formation), and iv) the fluorinated electrolytes showing better oxidative stability and thermal stability with cathode. The second set of studies reveals that the contact resistance is a dominant contributor, and carbon coating on the NCM improves the thermal and conducting properties with expected improvements in the power characteristics. The results demonstrated good progress towards the goals of this project and DOE.

The third reviewer commented that the information provided on the materials tested was very good. The key performance aspects were identified. The results did give direction to suppliers on how their product can be improved. The final cell size used for endorsement and direction should be increased to a minimum of 4 Ah for power application and 10 for energy applications. Results can be significantly different when the cell size is scaled up.

Another reviewer saw great results of evaluations and new insight provided by the fundamental portion of the work. The 4-point conductivity measurements of single particles are very impressive. This reviewer wanted to repeat the audience's concerns that as the particles become smaller, the conductivity of the particle may reflect more and more surface conductivity rather than that of the bulk material - maybe the current doesn't even need to go through the bulk. 4-point methods overcome issues with making good contact to the particle - not surface vs. bulk conductivity, something this reviewer suggests thinking about.

The fifth reviewer felt it was a bit light on  $MnPO_4$ . What are the real conclusions? SMG carbon: irreversible loss due to surface modification or decrease of surface area? This reviewer felt the work is assessing the obvious here. Most of the battery suppliers probably know about this. The second part of that work shows more promising results.

### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Reviewers provided positive feedback on the collaborations. The first reviewer suggests more effort should be put into bringing industry partners from the battery/cell manufacturing arena, and the second reviewer stated that there are several contributors to this study from different organizations and a few collaborations within ANL as well as externally. The third reviewer felt the project was very "linked in" with potential suppliers. There is good cooperation with fundamental scientists at ANL and modelers.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first reviewer felt that these studies are quite logical and help mitigate the barriers. This reviewer summarized how the proposed future research is to continue search for new, commercial materials to help meet the performance, safety and cost goals for PHEVs. Specific studies include assessment of i) LiNiCoMnO<sub>2</sub> (Phoenix), ii) hard carbon (Kureha), iii) aqueous binder (SBR), and iv) fluorinated solvent (Daikin) and carbon black (Cabot Corp). Likewise, studies will continue on the effects of carbon coating on cathode performance, on the contact resistance and optimization of carbon additives through modeling. Multiple workgroups at ANL work on very similar items. It would be nice to have a sharper division of workplans for a better appreciation of all the work currently underway at ANL.

The second reviewer felt that this project is generally very good, and suggests that any time a kinetic or conductivity issue (electrolyte or particles) is encountered, the impact should be evaluated at low temperature to more clearly see the impact.

The third reviewer suggests shifting focus toward more energetic materials and focusing the effort on how to achieve 250Wh/Kg,  $\sim 500Wh/L$  type of cells. The reviewer recommended the team look a bit more forward to more ambitious targets. Another reviewer questions how we can make best use of Dr. Lu's expertise to help the industry. The coating approach is interesting. It is developed by several companies but it seems to be difficult to control. This reviewer thought the study need to focus on this. Binder free is good. This reviewer asks why not work on a binder that lowers effects on conductivity. The fifth reviewer felt that the project was in line with objective, but would still like to see a more focused approach. The team could possibly work with a supplier to scale up the cell size and confirm the system performance.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

The first reviewer commented that the first part is just material screening. Resources need to be focused on second part. Another reviewer felt that the budget of \$650 k per year looks slightly excessive for these studies ( $\sim$  \$400K would be appropriate). The third reviewer commented that combined funding level is quite high (\$750K), but they are doing a lot of work. This reviewer felt that funding still seems to be a bit excessive, and suggests DOE review and scale back their plans a little.
### Materials Scale-up and Cell Performance Analysis: Vince Battaglia (Lawrence Berkeley National Laboratory)

### **REVIEWER SAMPLE SIZE**

This project had a total of 6 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The first reviewer felt that the project offers an important validation tool for the BATT program. The second reviewer felt the project was very relevant for developing next generation materials. Another reviewer commented that this project allows for a none bias evaluation of materials from researchers, which may be used to support the development of systems to meet the DOE goals. The fourth reviewer answered that the work is relevant, but do not forget about life improvement as well.

The final reviewer noted that with several advanced materials being developed in the BATT program, it is essential to have their performance independently assessed against DOE/USABC performance targets in standard test vehicle and environment. The objective of this task is to identify and evaluate four new materials in this year. A successful verification will thus lead to their incorporation in prototype cells and/or redirect the research efforts under BATT. A successful development of Li-ion batteries for PHEV will reduce the petroleum consumption, and pave the way towards petroleum replacement.



### QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The first reviewer commented that the team has established and demonstrated a really good cell design for testing materials. This work is critical to moving the program forward, both from a validation point of view but also to ensure that we don't miss an opportunity because of poor cell-making by other PIs. The project is looking across the program and is very open. This reviewer thinks their ICL cutoff of 30% is a little harsh in view of possible fixes for this (SLMP, matched cathode, etc.). If they have already made the cells, why not continue to cycle or is this yet another result of insufficient testing capabilities. The second reviewer commented that this is a very interesting approach that links the work done in the labs to the real life.

The third reviewer wasn't sure comparing various materials based on one criterion (electrode loading as stated by the PI) will compare apples to apples. Even based on this one criterion, whether the loading density is measured in mAh/cc or mg active matl/cc or mWh/cc the team will get different electrodes and different results from cell testing. What about the composition of active material versus conductive carbon? binder? What if a material has carbon coating or has better conductivity than the other, would you use same composition to compare? In the end the cell should be optimized for the material; otherwise, just creating one leveling field will result in disparities in other areas.

According to the fourth reviewer, the approach is to get new materials from BATT PIs in 10g quantity, and test the materials in halfcells using electrode fabrication techniques developed in BATT program. The tests include capacity measurements and rate capability. Should the results be encouraging, full cells will be designed, fabricated and tested to identify performance attributes and limitations. Even though this approach looks good, it is not clear to this reviewer if this 'verification and validation' of the BATT materials is necessary. Most of this assessment would be done by the BATT investigator, and with promising materials this should be done with an industry partner.

The final reviewer felt the approach as outlined is good. However, the presentation did not follow the approach presented. Good evaluation work was presented (particle sizing, XPS and Ion Sputtering work, etc), but none of this work was identified as part of the approach. On another question, is there any coulombic or energy efficiency evaluation work done in step two of the approach?

### QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

One reviewer saw good progress being made in benchmarking various materials in the same lab. Another reviewer felt that moderate progress has been accomplished in terms of evaluating several BATT samples both for performance and understanding their morphology/surface properties. Specifically, the team completed the assessment of four different materials from BATT and communicated the results to the PIs. They determined that both the MIT and HQ LiFePO<sub>4</sub> materials have coatings and perform well. HQ's has 100 nm primary particles with a 5 nm carbon coating, while the MIT's sample has 30 nm primary particles with a 1-2 nm carbon coating from oxalate precursor residue. These studies should focus more on high-energy materials than LiFePO<sub>4</sub>, which is quite mature, to address the low energy density barrier of Li-Ion batteries.

The third reviewer felt that the work shows a great understanding of what it takes to optimize a cell construction and electrode fabrication to get the most out of a material to avoid missing opportunities - remember that missed opportunities are much worse than false positives as the latter will be corrected in follow up work. Missed opportunities may just get dropped and never reexamined.

The fourth reviewer saw good insights into composition of LFP from multiple sources (MIT, HQ, ANL, etc.) by XPS, and ion sputtering. LFP is a good material but cannot meet the needs of higher energy PHEV and EV applications. A single-minded focus on LFP will miss potentially better opportunities elsewhere.

Another reviewer commented that it was unfortunate that we did not see the very good results. The fifth reviewer was not sure how this program can be evaluated, since if there were no responses to the inquiry, there would be no work. This reviewer thinks there should be a minimum number of responses. There were eight responses and the work done was identified, but very little actual performance work was presented. Most of the data presented was descriptions of the particles and materials, which is more of a material characterization effort than performance testing. This reviewer can only imagine that the performance data could not be shared, and if so the program should be changed to reflect this 'no share' rule.

### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

The first reviewer saw excellent collaborations among many labs. The second reviewer felt that, as expected from the proposed work, there are several contributors and collaborators in this effort. The project will probably benefit more from some collaborations with an industrial partner in scaling up/verifying these materials.

The third reviewer was encouraged by work with MIT and others: this reviewer was looking forward to hear the results. It is worthwhile for DOE to make sure such collaborations are not used by the other party(ies) prematurely as a seal of approval of their work before substantive results are produced. Another reviewer concurred, noting that there appears to be good collaboration between the partners.

The fifth reviewer commented that the team was very open and working with other labs well. However, this reviewer thinks they could move from passively accepting materials to actively asking for them - including from suppliers outside the BATT program (coordinate with Lu's work). Another reviewer commented that we need to encourage more groups to participate in that collaboration.

## QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

One reviewer felt that there was a good plan, although they would like to see the team open up to evaluate materials outside the BATT PIs. The second reviewer remarked that if this can be accomplished before the end of the year, this is great. The most promising materials need to be evaluated in larger cells.

The third reviewer suggests for future presentations, please include more detailed plans for evaluating test conditions for various materials (test conditions, evaluation criteria, etc.) Another reviewer would like to shift focus to higher energy (voltage+capacity) chemistries. Even doped NCM material may not offer a big enough leap from commercial NCM. We need a quantum leap, not an incremental improvement. The fifth reviewer noted how the proposed future research is to continue the assessment of i) MIT high rate LiFePO<sub>4</sub> material, ii) ANL's high capacity NCM material, iii) H.Q. FePO<sub>4</sub> laminates and iv) low-cost, Al-substituted NCM from LBNL. These studies partly address the technical and cost barrier. Again, the relevance of this assessment between the developments under BATT incorporation by industry (if promising) is unclear. The final reviewer was not clear how these current materials performed. Only statements were made about the performance. This reviewer thought that further performance testing and validation should occur prior to the step of determining best automotive application. Performance results should be presented.

### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

The first reviewer felt that the budget of \$290k per year looks reasonable for these studies. Another reviewer commented that depending on how many labs are willing to participate and send their material, it could be good to provide more resources to Battaglia's team. The third reviewer commented that this project already can't keep up. This is critical work and the reviewer suspects test position limitations are slowing this down. This is a major strategic weakness that goes throughout the DOE programs (except maybe for JPL).

### Fabricate PHEV Type Cells for Testing & Diagnostics: Andrew Jansen (Argonne National Laboratory)

### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The first reviewer felt this was very relevant for having in-house capability to manufacture cells. The second reviewer commented that there is a need for capability to design/optimize and build larger cells for material evaluation is well understood. This reviewer supports continuing this activity.

The third reviewer commented, with several advanced materials being developed and proposed for the PHEVs, it is essential to have their performance assessed as large-format or sealed cells. The objective of this task is to obtain test cells for calendar and cycle life studies in pouch cell or rigid cell (e.g. 18650) formats from industrial battery vendors. A secondary objective is to develop the capability in-house to fabricate 18650 cells. A successful development of Liion batteries for PHEV will reduce the petroleum consumption, and pave the way towards petroleum replacement. According to the fourth reviewer, the evaluation of materials in more appropriate cell size formats is critical to identifying materials that will be used to build cells for use in PHEVs. This project does that and consequently supports the DOE objectives. The fifth reviewer



perceived that it is vital to get this effort going to make cells for the battery program.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The first reviewer recognized that it is critical for the DOE labs to have their own real cell-making ability to test their own material and also to get decent cells. This gives them a very powerful tool to advance their own material programs and also as a side benefit will give them a great learning opportunity in the making of real cells. The second reviewer noted that the approach is to establish subcontracts with battery developers to produce large-format or sealed cells (18650) for the high-energy applications (with thicker electrodes) based on screened materials from suppliers and from the ABR and BATT programs. In parallel, efforts are underway to fabricate pouch cells and/or 18650 cells in Argonne's new dry room facility with suitable electrode-fabrication equipment.

According to the third reviewer, the project was well though through and contained a methodical approach by the PI to design and build cells, but this reviewer does not fully agree with the statement that thin electrodes result-in higher impedance cells. The reviewer agrees with the point that thinner electrodes have higher ASI. If an electrode is used that is twice as thick but has 1/2 ASI, at the cell level the same impedance is seen because twice as much electrode can generally be used (hence twice as much electrode area) in the cell. Cell optimization needs to be thought through more carefully.

Another reviewer noted the clear concise approach for Argonne developing the expertise to fabricate cells, in-house for testing. One concern is whether Argonne will be cost competitive for the material developers to come to Argonne to fabricate these cells.

### QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL

### PROJECT AND DOE GOALS.

The first reviewer noted that there has been pretty good progress towards this long-awaited goal. Progress is impressive in setting up the facilities. The data on cell thickness and resistance is quite interesting. The second reviewer was encouraged to hear progress on building the dry room and installing electrode/cell fabrication equipment. However, how will this work be coordinated with the cell manufacturing and process development facility in Kentucky? The third reviewer saw excellent progress, and noted how the project obtained cells for initial evaluation as baseline performance barometer for cells produced by Argonne. The team has also procured equipment for cell fabrication.

Another reviewer perceived that moderate progress has been accomplished in terms of getting the prototype cells from industry partners and in setting up the cell fabrication facility at ANL. Specific accomplishments include: i) studied the effect of electrode thickness on performance, ii) identified electrode coating and cell fabrication vendors from high-energy electrodes and iii) initiated the design and installation of new dry room and iv) gathered information on the cell fabrication equipment. Surprisingly, there is not much done in terms of getting the cells made from the industry partners with any advanced materials. The progress is oriented towards meeting the project and DOE goals. The fifth reviewer thought the dry room layout looks good for what they have to do in the space available. The reviewer offered that this was a nice job and looks forward to seeing the results of cells made on this line. The team may need to at least think about adding a smaller version as a contingency if they find they have to separate one or more processes to avoid cross contamination (not sure DOE funding system really handles contingency funding very well). The reviewer hoped the new partners can make better baseline cells than before. This reviewer was not happy with the small scale of this work. This reviewer would have thought they could have made at least 300 baseline cells as most of the cost is involved in set-up/clean-up. The reviewer observed they can't test even the cells they do make: this is another issue.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Several reviewers generally saw evidence of collaboration. According to the first reviewer, as expected from the proposed work, there are several contributors and collaborators in this effort, including industry partners. The second reviewer saw multiple contributors. Another reviewer commented on how the project is working with better partners and seems to have a really good appreciation of the nuances of cell making and some of the pitfalls, learning from Sandia's and industry experience. The fourth reviewer thought the project would benefit from expanding relationship with other suppliers and OEM's.

## QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first reviewer commented that with this ability now in place, ANL should now be well-positioned to carry out controlled testing. The second reviewer felt that the proposed future research is to continue the evaluation of baseline 18650 cells for quality and performance, procure 18650 cells with high energy cathode materials from approved vendors, order cell making equipment for fabrication of cells in Argonne's new dry room facility and fabricate cells at ANL with advanced materials in support of the BATT and ABR. The planned effort is in tune with the project goals. Another reviewer sees the project moving into the next phase of activity and is on a good time schedule. The fourth reviewer believes that other than the cell testing issue, their plan looks fine. In this reviewer's opinion, the "elephant in the room" is really the test position limitations and this drags down the reviewer's assessment of the future plans. The fifth reviewer thought that more focus on cell design and optimization is needed.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

For one reviewer, the budget of \$400 k per year looks reasonable for these studies. Another reviewer commented that this is a critical program but is being hampered by cost-cutting everywhere. This reviewer commented:

1. The dry room is too small.

2. They should have ordered at least 300 baseline cells and much more cathode so they could make their own cells (using JCS and their own cathodes) for validation purposes.

3. This reviewer's single biggest concern is that even with a too-small baseline sample, they are limited by test positions at both national laboratories. While acknowledging that no one ever has enough test positions, this team is not even close; the problem will only get worse. This reviewer feels that this is false economy in that inadequate data will result that will slow down the entire program and give false directions. We have some of the smartest and most expensive researchers in the world being hampered by inadequate resources. This program needs a massive scale-up of testing positions and associated temperature chambers ASAP - it's already too late to meet the program needs. As the cycle life improves and the team starts looking at varied test protocols, this problem will only get worse. Basically, the better you get, the more test positions become limiting. When things don't work well, you don't have to do much testing.

## Electrochemistry Cell Model: Dennis Dees (Argonne National Laboratory)

### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The first reviewer felt the work was highly relevant. Another reviewer firmly believes that analytical models can shed light on and help in understanding the complex electrochemical phenomena. The effort should be supported. The third reviewer noted that identification of cell degradation mechanisms is key to the success of electric vehicle program. The fourth reviewer commented that modeling and fundamental work like this can be critical in understanding cell chemistry and directing future work.

### QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED,

### FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

According to one reviewer, the project had very effective approaches to tackle many key issues. For the second reviewer, this is very challenging work and the approach taken in trying to really have a fundamentals-based rather than empirical fitting model is excellent. The third reviewer felt that definitely more work needs to be done to



develop reliable models to link to experimental results and understand aging mechanisms.

### QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

The first reviewer commented that Dennis has been making significant contributions to our theoretical understanding of cell behavior. These are invaluable data and the reviewer is very pleased with the progress the author has been making. The second reviewer felt that good insights provided in graphite phase-transition during the lithium intercallation process. The third reviewer saw nice work on the modeling and understanding of the three stages of the graphite electrodes. Kinetics-limited mechanisms are very important to understand.

The fourth reviewer noted that the anode modeling work has shown great strides in a difficult area. Fitting models are fine when great precision is needed and the system is in a stable, mature state, but fundamental models like this work are the ones that can give true insight. (According to this reviewer, people often get too hung up on how well the model fits the data. In cases like this, it just has to be close enough so it can teach you something.)

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

One reviewer saw good collaboration with experimentalists and seems to fit in well with other models. This is a critical feature where a lot of modelers fall down. The reviewer was very happy to see the close collaboration with and understanding of "real" batteries by the PI in his work. This reviewer was still a little lost on all the modeling initiatives that the DOE labs have going and have been done in the past - do they really support each other and/or are they competing? For example, does his proposed capacity loss model fit in with the work that Gering is doing on life analysis and modeling? Some of Dees' slides address this, but it would be nice for someone

to show an overview of the various models and what they do next time (it is not necessarily Dees's job to do this of course). The second reviewer would love to see additional collaboration with other labs, especially universities. The third reviewer felt that collaboration between labs, universities (and maybe industry) should be encouraged. Why not a collaboration with LBNL?

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

A reviewer felt the project had an excellent plan of attack, and was very well focused. Another reviewer commented that an SEI model will be highly appreciated by the developers and scientists. We would also like to see how the electrode functions/degrades as a function of temperature, cycling regimes. Also, how about work on Li plating? Can the work be extended to include this issue also? The third reviewer would like to see modeling and simulation evolving to a point where they are used to predict the unknown rather than simply used to explain or map known phenomena. The reviewer understands the challenge, but thought the goals should be a lot loftier than the one set. For the fourth reviewer, the determination of the aging mechanism is critical to the process. A significant effort need to be put understanding these mechanisms.

### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

One reviewer commented was that the project is doing well with the funding and should continue at the current level. It is critical to ensure that Dees continues to get the support from the experimentalists he needs.

## Diagnostic Studies - Argonne: Dan Abraham (Argonne National Laboratory)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 6 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The first reviewer felt that the work is very relevant, and the second reviewer commented that the focus on life is good. The third reviewer thought the project addresses lifetime issues of Li-Ion cells, especially the benchmark chemistries. The team is also looking at some new materials to improve lifetime.

Another reviewer noted that the current and advanced materials being developed for PHEVs do not quite meet the requirements in terms of calendar life and cycle life. The objective of this project is to gain a fundamental understanding of material changes and processes responsible for system performance degradation. Such an understanding helps in the development of improved materials by overcoming the performance limitations. A widespread use of Li-ion batteries for PHEV will reduce the petroleum consumption, and pave the way towards petroleum replacement. The fifth reviewer thought the work would be more relevant if the focus is shifted to where the industry is moving. The industry is moving away from NCA-based oxides towards NMC oxides. Even better, looking towards the future of high voltage/high energy cathodes will make the effort payoff much more pronounced.



### QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The first reviewer commented on how the project is using a whole range of techniques to study and truly understand what's going on inside the cell, which the reviewer liked very much. The second reviewer believed the approach is good but it would be helpful if it was better described. Especially what is the intent of that study, what does the investigator want to get out of it? Can we expect an understanding of failure mechanism and direction on materials and cell design? The third reviewer thought the project is doing good work, but can produce more substantial results if it were not so scattered. Several positive structures were looked at without fully bringing any one material to a significant conclusion. The reviewer was not sure what the selection criteria for the material is/was, but it's important to select one which is most relevant, e.g. NMC or one of the newer HV cathodes which are needed for PHEV.

According to the third reviewer, the approach is to adopting various electrochemical-and physicochemical-diagnostic techniques, such as spectroscopy, microscopy, diffraction, and chemical analysis at different laboratories to understand the structures of the electrode materials and their rearrangements upon cycling, repeat similar studies with the cell components harvested from the cells to characterize the interface at the electrode/electrolyte interface through in-situ electrochemical studies and ex-situ analytical techniques and to study the effect of moisture in the electrolyte on the performance to propose reaction mechanisms. These studies are quite relevant to the stated objectives of understanding the life and performance - limiting processes. However, it will be more beneficial and relevant to develop in-situ diagnostic technique, since the interfacial changes are more dominant than the bulk changes in the electrodes.

### QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL

### PROJECT AND DOE GOALS.

The first reviewer was very impressed by the level of sophistication that has been brought to bear upon the diagnostic studies. A significant amount of very useful results has been generated from these studies, which are really helpful to understand Li ion battery chemistries. The second reviewer concurs, commenting that a lot of work has been done. The insights are good and tend to be more on the practical side of cell design and development. The third reviewer commented that a lot of work has been done, and hopefully more is to come.

According to the fourth reviewer, moderate progress has been accomplished in terms of understanding the structural aspects of Mnbased layered oxide materials of interest to PHEVs. In particular, it was shown that these composite oxides contain an intimate mixture of  $Li_2MnO_3$ -like and  $LiCoO_2$ -like areas with the Li atoms well-ordered both in, and normal to, the transition metal. Upon cycling, however, this ordering is lost after high-voltage cycling (> 4.5V). Similar diagnostics studies have been initiated on the PHEV baseline electrodes and cells. These studies are interesting, but the reviewer thought there is not much correlation with the performance loss or any feedback in the design of new materials. The progress is oriented towards meeting the project and DOE goals.

For the fifth reviewer, there were so many activities under this program that the reviewer found it very hard to assess progress from a 20 minute talk – which is a reflection on the review system, not the PI. Reviewing the slides helped and there seems to have been significant progress on a large number of areas, although several of the slides focused on activities completed and not enough on the actual gain in knowledge – basically some slides left the reviewer thinking "so what." Nevertheless, despite the large funding amount, the reviewer was very impressed with both the quality and especially the sheer volume of this work.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

For one reviewer, this is a true collaborative effort. For the second reviewer, there are several useful collaborators in this effort, mainly from universities and national laboratories. Another reviewer commented that this group seems to have really good connections to many partners, both universities and other national laboratories. The third reviewer commented that partnership with the industry is recommended.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first reviewer summarized how future plans include performance and performance degradation studies of electrochemical couples identified for PHEV cells, by continuing to (a) examine the SEI formation on graphite electrodes in various electrolyte systems, (b) correlate the surface film formation and electrochemical performance of positive electrode materials, and (c) correlate of electrolyte composition and electrolyte additives to cell performance and performance degradation (i.e., effect on calendar life). The planned effort is in tune with the project goals. As this reviewer mentioned before, new in-situ diagnostics to understand the interfacial changes will be more beneficial.

A reviewer thought that plans going forward were not well spelled out, but they seem to be well-focused and the reviewer is optimistic they will continue to do good work. For the third reviewer, it will be very useful to draw conclusions (not only just mention what has been done) as much as possible to aid the audience on a slide or two. With so much data in place, it is easy to lose track. The fourth reviewer recommend focusing on the most relevant materials (high energy cathodes/anodes) with the objective of addressing or shedding light on the most urgent problems (safety, life, ..) Another reviewer commented that the proposed future research will be better described by the other collaborators of that program.

### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

According to one reviewer, the budget of \$600 k per year looks reasonable for these studies. The second reviewer thought the funding level is quite high but the quality and especially the quantity of their work justifies continuing funding them at this level. The team is doing a nice job as far as the reviewer can tell.

### Electrochemistry Diagnostics at LBNL: Frank McLarnon (Lawrence Berkeley National Laboratory)

### **REVIEWER SAMPLE SIZE**

This project had a total of 6 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The first reviewer agreed the project is relevant. The second reviewer noted that diagnostics have provided invaluable insights into the inner workings of the material. The third reviewer commented that the project addresses lifetime issues of Li-Ion cells. The fourth reviewer noted that the current and advanced materials being developed for the PHEVs do not quite meet the requirements in terms of calendar life and cycle life. The objectives of this project are to understand factors that can enhance the stability of SEI layers from post-test characterization of components from ABRT test cells, ii) establish and investigate degradation mechanisms of PHEV cells, and iii) develop strategies to minimize irreversible cell capacity losses by developing surface treatment regimens to reduce side reactions. Widespread use of Li-ion batteries for PHEVs will reduce the petroleum consumption, and pave the way towards petroleum replacement.

### QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED,

#### FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

For the first reviewer, proper tools and methods were used and interpreted in a complementary manner. The second reviewer said the investigators are looking at diagnostic of the C anode to minimize irreversible capacity loss as well as the investigation of the failure mechanisms of PHEV cells.

The third reviewer said that the approach to functionalize the graphite edges was well thought out, even if it didn't work. Surface Raman showing degradation at both electrodes surfaces is also potentially very important. The reviewer thought the work focused too much on the Raman method and impedance alone. The reviewer would have liked to see other techniques used to get a more complete picture of what is going on in these electrodes. In particular, the reviewer thought they should get a better picture of the distribution of the effects they see on the bulk of the electrode - although the impedance data addresses this somewhat. Another reviewer commented that the analytical work is very useful. However, this reviewer is not so sure about the modification of the carbon surface work.

The final reviewer commented that the approaches include i) developing strategies to minimize irreversible capacity losses by a surface-modification of carbons, ii) carrying out diagnostic post-test diagnostic evaluation of components from ABRT test cells using spectroscopic, microscopic, X-ray, chromatographic, and related techniques, iii) understand factors that can enhance the stability of SEI layers and, iv) establish and investigate degradation mechanisms of PHEV cells, i.e., mainly through Raman spectroscopy and EIS techniques. Since the changes occurring at the electrode/electrolyte interfaces are dominant on the performance than the bulk characteristics, developing suitable non-electrochemical (more definitive) in-situ techniques will be more beneficial.



### QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL

### PROJECT AND DOE GOALS.

The first reviewer said that the earlier results obtained on carbon and metal-oxide electrodes are very useful. The second reviewer said there was excellent work on the graphitic anodes. The team needs to understand more about hard and soft carbons as they are more and more used in large format cells. Why do more amorphous carbons perform better in cycle life and even presumably in safety? What is the SEI layer differentiation between graphitic and amorphous carbons? The reviewer recommended that more work needs to be done with alternative anodes (LTO, Si-based...). Graphite is a good start but we should move on with newer materials.

The third reviewer commented the investigator gave a good view of how diagnostic techniques can help to understand failure mechanism of both positive and negative electrodes. Another reviewer felt moderate progress has been accomplished in terms of understanding the degradation mechanisms, mainly of the carbon anode. Based on the graphite anode structural degradation results, the team identified approaches to anode stabilization. Also, the team has identified candidate anode and cathode fade mechanisms in Gen-3 cells, which are similar as in gen-2 cells, i.e., contact resistances between primary particles and conductive carbon matrix, loss of available Li, and/or electrolyte starvation. Further, carbon disordering upon cycling, which is accelerated by complete delithiation from carbon, increases anode surface reactivity and causes SEI layer reformation, which shifts the cathode to a higher SOC and accelerates cathode degradation. Interesting that these findings are, it will be beneficial to see if similar failure occurs in a PHEV prototype cell with different designs, i.e., with different carbons (MCMB vs graphite) and electrode ratios.

The fourth reviewer thought the results shown were pretty good, but would have liked to see a better appreciation of how much of the anode is really being affected by this phenomenon. If the backside of the electrode near the carrier is basically OK, then it's hard to know how important the effect they find really is; maybe it would be less important for PHEV batteries as they are using thicker electrodes. Maybe the team should combine this with other techniques to try and get a better handle on how the effect they find varies with depth into the electrode.

### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

The first reviewer observed excellent collaboration with many labs. Another reviewer said to extend collaboration with industry/academia. The third reviewer wrote that there are several useful collaborations with the ABR partners from the DOE Laboratories. Collaboration with industry partners (with their hardware) will be helpful in developing a wide database on the failure mechanisms.

Another reviewer commented that we need to better understand how this work is connected to other groups work and what potential collaboration may look like. For example, the approach would very useful to understand surface chemistry and failure mechanisms of high voltage cathode and negative alloys. The fifth reviewer did not really see much evidence for cooperation aside from getting samples. The reviewer would have thought that bringing in other techniques in a combined effort would be more fruitful – the reviewer only saw the Raman and impedance studies done at LBNL.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first reviewer commented that the proposed future research is globally good. It would be interesting to apply this diagnostic approach to help the development of the most promising next generation materials (cathode, anode and separator). The second reviewer wrote that the analytical work should be continued at the same pace as before. The reviewer was not sure about the work to modify carbon: they thought some of this kind of work has been done before. The third reviewer said that the future studies are aimed at i) understanding the SEI layer formation and stabilizing the same, minimizing the irreversible capacity loss by changing the surface properties through pretreatments and ii) performing diagnostics tests of components from ABRT cells by examining electrode structures and surface films, establishing and investigating degradation mechanisms of PHEV cells and comparing them in ATD vs. ABRT cells. Again, developing in-situ techniques will be beneficial in understanding the interfacial changes. The fourth reviewer suggests going beyond graphite and old oxides (NCA). They are not part of the future. The final reviewer was not sure they saw or

heard a clear plan of where the team plans to go from here – little bit in terms of backing off on the fluorination reaction, but not much else.

### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

One reviewer remarked that the budget of \$600 k per year looks slightly excessive for the scope of this effort (based on last year's progress). For the second reviewer, the funding level is quite high: the reviewer felt this is excessive in view of the effort being expended. The largely negative results and lack of a clear technology approach to fix the problem also suggests to the reviewer that this project is overfunded and could at least be trimmed back.

**ENERGY** Energy Efficiency & Renewable Energy

Diagnostic Studies to Improve Abuse Tolerance and Life of Li-ion Batteries: Xiao-Qing Yang (Brookhaven National Laboratory)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The first reviewer commented that the project is very relevant to understand how batteries fail, so that we can develop better batteries. The second reviewer stated that the project provides good in-situ understanding of structural changes in active materials during charge/discharge and thermal instability, and the third reviewer stated that this project addresses the fundamental stability and safety of critical materials of Li-Ion batteries.

The fourth reviewer stated that the current and advanced materials being developed for PHEVs do not quite meet the requirements in terms of calendar life and cycle life. The objectives of this project are to undertake diagnostic studies utilizing new in situ diagnostic techniques with surface and bulk sensitivity to understand the thermal abuse tolerance as well as performance degradation (capacity and power fading) of Li-ion cells. Such understanding is crucial in mitigating these failures and designing advanced materials. Widespread use of Li-ion batteries for PHEVs will reduce the petroleum consumption, and pave the way towards petroleum replacement.



### QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The first reviewer thought the work had very good approaches. Other comments focused on the benefits of in situ analysis. A second commenter wrote that tools are well-suited to provide structural insights into materials in-situ. The third reviewer commented that insitu analysis during thermal heating is a very good approach as it allows actual observation of the mechanisms that lead to the failure and helps to develop a better strategy to prevent them.

A fourth commenter wrote the approach for understanding the abuse tolerance is based on the use of a combination of time resolved X-ray diffraction (XRD), in-situ soft and hard X-ray absorption (XAS), and in-situ transmission electron microscopy (TEM) techniques during heating of the electrode materials. For identifying the life-limiting process, the approach involves the use of in-situ XRD, soft and hard XAS studies of new electrode materials during charge-discharge cycling. These approaches are novel and sound and are expected to lead to good understanding understand of the advanced materials. A fifth commenter focused on how the team continues to leverage their surface/bulk XAS/XRF method by applying this to new materials and trying to address the coating approach to batteries. The ability to do in-situ work separates this group from many other groups in the world looking at these materials.

### QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL

### PROJECT AND DOE GOALS.

The first reviewer thought the team gained very valuable insights into the various mechanisms that cause fade in batteries. This technique is proving to be an invaluable arsenal in probing the structure/structural changes of the various battery materials. The presentation was "simply brilliant and lucid," according to this reviewer, who thoroughly enjoyed the talk.

The second reviewer commented on how this group continues to do world-class work and their results are always both impressive and trustworthy. They are showing insight into material stability that no one else is providing – both in the area of beam time and the TEM work. The third reviewer said there is a wealth of knowledge produced in understanding the role of various TM's in the oxides and how they actually work. Valuable insights are being developed into why layered, particularly nicklates are unstable and why NMC materials do better on safety. The fourth reviewer thought the presentation shows that Mn is very good to stabilize the structure, but because of stability of Mn the material has less capacity. This reviewer asked how to stabilize the structure without reducing capacity. Some nice results were developed about stabilization of LiNiCoAlO<sub>2</sub> structure with ZrO<sub>2</sub> coating.

The fifth reviewer summarized how good progress has been accomplished in terms of evaluating various materials for their structural changes during electrochemical cycling and thermal abuse. Specific accomplishments include: i) In-situ hard and soft X-ray absorption spectroscopy (XAS) study on charged  $\text{Li}_x \text{Ni}_{0.8} \text{Co}_{0.15} \text{Al}_{0.05} \text{O}_2$  (Gen2) and  $\text{Li}_x \text{Ni}_{1/3} \text{Co}_{1/3} \text{Mn}_{1/3} \text{O}_2$  (Gen3) cathode materials during heating, ii) Development of new in-situ diagnostic tool using high resolution TEM (HR-TEM) for thermal abuse studies, iii) In situ HR-TEM study of overcharged Gen2 cathode, iv) in-situ XRD studies of new Cr and F doped  $\text{LiMn}_{1-x} \text{Cr}_x \text{O}_{4-y} \text{F}_y$  spinel from ANL. Significant finds are: i) the thermal decomposition of Gen2 cathode occurs at the surface much earlier than in the bulk. The conversion of the layered structure the spinel and to the rock salt and its growth, occurs at the surface much earlier than in the bulk, which can be mitigates by a surface coating ii) Cr substitution in Cr and F doped cause structural changes in the spinel that account for the capacity loss and subsequent restoration, respectively. These studies demonstrate a definite progress to the stated objectives and are thus in tune with the overall goals.

### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

The first reviewer commented that the PI has involved "the entire world" in his collaboration, which is great. In comments, the second reviewer pointed out how the project has a good broad spectrum from DOE labs, industry, and academia. The third reviewer commented that there are several useful collaborations with the ABRT partners from the DOE laboratories as well as with industrial partners. The final reviewer remarked there is a long list of collaborators - very open. This is all the more impressive in light of their scheduling issues with beam time. The reviewer thought they must be a pleasure to work with.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first reviewer commented, "excellent!" The reviewer inquired, "can't probe SEI with this technique?" The second reviewer remarked this is one of the most exciting answers we are looking for. Does the coating really work? If yes, how does it work?

The third reviewer can't argue with the path that has generated a lot of good data so far. The reviewer thought it would be nice to plan a predictive tool for evaluating whether an active material will do well on safety, life, etc. Also, using these techniques to predict life or in general state-of-life of a material could be very helpful. The fourth reviewer really looks forward to seeing them continue to develop their in-situ TEM methodology and applying it to Li-Ion cells. This is especially important due to the surface reactivity of both electrodes and film formation at the anode and maybe cathode. In view of beam time issues, maybe the team should always ensure they have materials and plans ready to go so they don't miss beam time windows that apparently come and go. (The reviewer thought maybe the team does that already, but that wasn't the impression the reviewer got).

The final reviewer noted that the future studies are aimed at continuing the i) in-situ TEM studies on the thermal stability studies on the Gen and Gen 3 cathodes before and after surface modification ( $ZrO_2$ ,  $AlPO_4$ , and  $Al_2O_3$  etc) ii) In-situ XRD, TR-XRD, hard and soft XAS study of  $LiNi_xCo_yMn_zO_2$  (x + y + z = 1) to correlate the compositional effects with performance loss, and iii) Apply these

new techniques to various electrode materials to probe their structural changes at the surface and in the bulk simultaneously. The planned studies are helpful in mitigating the technical barriers of life and safety for Li-ion cells.

### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

The first reviewer commented that the budget of \$350 k per year looks quite reasonable for this effort. The second reviewer thought it might be more productive to add resources in this area that has produced so much already. The third reviewer thought they do very well with their current funding, but would like to see this group get more beam time and more funding - although the reviewer was not sure more beam time is a funding issue or just a priority call at Brookhaven Lab.

### Develop and Evaluate Materials and Additives that Enhance Thermal and Overcharge Abuse: Khalil Amine (Argonne National Laboratory)

### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The reviewer commented that this project is extremely important. Another reviewer had similar reactions, commenting that improving the safety is one of the most important characteristics in the development of PHEV and EV battery systems. This project investigates ways to improve the abuse tolerance of the Li-ion chemistry and consequently supports the DOE objectives. The third reviewer commented that it addresses safety and lifetime issues of Li-ion cells.

The fourth reviewer said that the current and advanced materials being developed for the PHEVs do not quite meet the requirements safety. The objectives of this project identify contributions from each of the cell components of different chemistries to the abuse characteristics and utilize this understanding to develop new abusetolerant materials and provide them to SNL for validation of safety benefits in 18650 cells. A widespread use of Li-ion batteries for PHEV will reduce the petroleum consumption, and pave the way towards petroleum replacement.



### QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The first reviewer thought the approaches were well thought out - a lot is going on at once, so they are covering a lot of ground in one program. The second reviewer noted that overcharge shuttles sound very attractive but the reviewer has yet to see a good, functional one.

For the third reviewer, a suggested change would be to concentrate on identifying and improving the key safety characteristics of one of the electrodes rather than looking at multiple areas of both. While identifying redox shuttles for overcharge protection may be good, from an automotive vehicle perspective, the most unlikely abuse situation is an overcharge event. The reviewer was not sure of the value of this work, unless there is another advantage developing this process, especially when done at the coin cell level.

The fourth reviewer commented that the approach targets improvements in all the cell components for increased safety, e.g., safer anodes and cathodes, additives for stable SEI, surface modification for cathode, safer electrolyte components (solvents and sat) and redox shuttles for overcharge protection. Materials are being secured from in-house researchers, partners and commercial sources and assessed for safety improvements, which are subsequently verified in 18650 cells. The approaches look reasonable and feasible and will lead to further understanding of safety issues of each component and later to safer cell components.

### QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL

### PROJECT AND DOE GOALS.

The first reviewer commented that the results shown were pretty good overall. Nice use of activation energies to characterize and try to understand processes. It was gratifying to see that the fundamental material safety characteristics in many cases do carry over to full cell studies (although SNL has certainly shown that this is not always the case.) The reviewer thought rates for the overcharge additives were a bit low. The team needs to look at higher charge rates and impact of low temperature where diffusion limitations may be more problematic on these additives as well. Another reviewer said that work done to elucidate what takes place on the carbon electrode as well as finding ways to alter them have been quite impressive at the author's lab. Data presented in another section on additives combined with these efforts make it a comprehensive effort to tackle this important issue. The third reviewer noted that good progress has been accomplished in terms of evaluating various cell components for safety. For example, i) three types of MCMB carbons with different surface modifications were evaluated at the component level (DSC) and later in 18650-cell format (ARC) at SNL, which show that the type of carbon impacts the safety of lithiated carbon. ii) Three electrolyte additives were identified for SEI stabilization on graphite, including LiDFOB. iii) Four salts were evaluated, with LiPF<sub>6</sub> showing the least thermal stability (230C vs 310C). iv) Several 18650 cells made with NCA and Al<sub>2</sub>O<sub>3</sub> coating were procured from industrial partners and being valuated for cycle life, abuse resistance and v) Three new redox shuttles corresponding 4.17 V, 4.2 V and 4.8V have been identified and demonstrated in coin cells. These studies demonstrate a definite progress to the stated objectives and are thus well in tune with the overall goals.

Another reviewer said that work presented was good, but results were not very clear, with respect to the objectives and the approach. Should present summarized results in line with objectives and approach. For example, what is the role of the anode and which anode provided the best safety in the chosen cell? If multiple areas are to be investigated, a test matrix with two cathodes and three anode types (or vice versa) would be more informative.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Reviewers commented on the collaborations. The first reviewer saw excellent linkages with external partners. The links to Asian companies are a very important advantage for this PI (obviously not something they cannot discuss openly in the briefing). Slides show results of working with Sandia, but PI's talk gave the impression that Sandia and ANL are not really talking as much as they need to. Just providing SNL and IDL with cells is not an accomplishment – it's what they did with them that matters. It seems to be a bit of a barrier, but the reviewer confessed that this is just an impression that may well be wrong. The reviewer suggests the SNL-ANL work at least be reviewed to ensure they are working together as well as they can since this linkage is critical for the program. The second reviewer thought that all the relevant labs have been included in the work. The third reviewer said that there are several useful collaborations with other DOE laboratories (SNL) and more importantly with the commercial materials suppliers and industrial partners. The final reviewer saw the good collaboration with suppliers, but there was no reference to the Fe phosphate provider.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

One reviewer suggests that the efficacy of coatings should be checked over the entire life span of the cells. Making statements from initial data could be misleading, and hence it is recommended that data should be acquired both at the beginning of life and at end of life. The second reviewer thought that the plan going forward was good. This reviewer's main concern is with them trying to do too many things at once. This reviewer would like to see a little bit more focus and bring fewer concepts through the development stages into 18650 size cells for SNL and others to test.

Another reviewer said that future plans are too varied and extensive to be accomplished and provide data that could be used to drive decisions. The fourth reviewer said that the future studies are aimed at continuing to i) explore electrolyte additive to reduce heat flow from SEI decomposition at low temperature, ii) investigate the safety of anode without SEI, iii) quantify the thermal effects of LiPF<sub>6</sub>, iv) investigate the role of none flammable electrolyte ionic liquids on the safety, v) investigate the effect of cathode composition, morphology and surface area on safety, vi) characterize of ANL's new redox shuttles, and continue exploring new shuttle structures. Efforts will be made to demonstrate the benefits with these cell components n 18650 cells made by an industrial partner. The planned

studies are helpful in mitigating the technical barriers of life and safety for Li-ion cells. One difficulty with this approach is that it is broadly diffused: it is more of evaluating what is out there vs. designing developing something new (except for the redox shuttles)

### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

Two reviewers thought the funding level was appropriate, with one reviewer remarking that the budget of \$440k per year looks quite reasonable for this effort, and the second reviewer stating that the funding level is appropriate. They are doing a lot with what they get. The third reviewer commented the resources for the proposed work is insufficient. The reviewer would propose a much more reduced and focused work plan, however, rather than increasing the resources to this project.

## Abuse Tolerance Improvement: Peter Roth (Sandia National Laboratories)

### **REVIEWER SAMPLE SIZE**

This project had a total of 6 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The first reviewer said that this is extremely important. Another reviewer said that of course developing intrinsically safe batteries is key to success for petroleum displacement. The third reviewer thought that improving the safety is one of the most important areas in the development of PHEV and EV battery systems. This project looks at materials and methods to improve the abuse tolerance of the Li-ion chemistry and consequently supports the DOE objectives. A fourth remarked that adequate safety is critical for successful implementation of DOE objectives (although when viewed against the huge number of existing cars that spontaneously burst into flames every year, the emphasis has to be on adequate, not perfect). A final reviewer wrote that Li-ion cells aren't tolerant to electrical or thermal abuse, which might lead to thermal runaway. The objectives of this program are to i) Identify degradation mechanisms of gas and heat-producing reactions in Li-ion cells, ii) develop advanced materials or combination of materials stable during abuse events, leading to enhanced safety, iii) demonstrate improved abuse tolerance in 18650 cells, and iv) develop techniques to understand and mitigate internal shorts. Improvement in safety is crucial to a



widespread use of Li-ion batteries for PHEV will reduce the petroleum consumption, and pave the way towards petroleum replacement.

### QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The first reviewer remarked there were well-tested approaches to identify abuse-tolerance of various cell systems. The second reviewer commented that it is absolutely critical to find a method to properly simulate internal short circuits. The third commenter said the approach is very good toward accomplishing the goal of evaluating new materials from other labs and suppliers. This may have been done earlier, but it would be good to confirm that SNL can produce a commercial grade cell by making an 18650 cell from the same materials as a commercially available cell and comparing the abuse test results. The same procedure should be considered for the coated cell work. This reviewer thought that a project that establishes non-destructive ways to evaluate the potential of a cell's abuse tolerance and confirming the results by actual testing would be a good project for this lab. The project would most likely involve the collaboration with another lab(s). A fourth said they are looking at important key elements of safety in various chemistries. They have built up an impressive capability over the years. Their work on using low temperature molten metals looks to be an interesting way to generate shorts, although details were sketchy.

The fourth reviewer said the approach focuses on cell level abuse testing through ARC, which has been the main forte of SNL's safety studies. The approach includes building 18650 cells using SNL in-house cell fabrication facilities, using new anode and cathode materials and electrolyte and electrolyte additives and evaluate their thermal and overcharge tolerance, specifically the gas and heat generation, as well electrolytes and electrolyte additives from other DOE laboratories. In addition, the mechanical and thermal integrity of the separators and the effects of internal shorts contributing to the safety will be studied. The abuse test facilities at SNL

are established and are being used by all the DOE laboratories as well as industry partners. The approach is thus quite well-designed, feasible and well-integrated with the efforts of ABR partners.

### QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

The first reviewer remarked the work is high quality and significant amount of test results were seen. The work is very impressive and, as always, very reliable data were shown. The second reviewer remarked that good progress has been accomplished in terms of evaluating several combinations of cell components for abuse tolerance. Specifically, i) Improved the peak heating rated in the prototyping test facility, ii) studied thermal stability of a variety of anodes and VC additives in Hitachi-built LiFePO<sub>4</sub>, iii) Developed a novel LiF/ABA anion receptor-based thermally-stable electrolyte, with ~ 100°C improvement in thermal stability and reduced gas generation, iv) established new coating capabilities for carbon anode and cathode electrodes, v) demonstrated the ability to initiate internal short circuits in coin cells using a low temperature alloy defect trigger, vi) established separator testing platform, viii) determined the amounts of gas generated with spinel, gen-2 and gen-3 cathodes, viii) determined the effects of fluorinated LiBOB (LiC<sub>2</sub>O<sub>4</sub>BF<sub>2</sub>) additives on the thermal reactivity of 18650 cells with LiMn<sub>2</sub>O<sub>4</sub> spinel and Gen3 cathodes. These studies demonstrate a definite progress to the stated objectives and are thus well in tune with the overall goals.

Another reviewer remarked that it was good to see that full size cells are being used for testing. The reviewer would like to see testing performed with larger cells, as response varies greatly as the cell size increases. It is not clear how the cell size effects can be captured accurately without going to larger format cells. Identifying a testing method that can be an accurate barometer for abuse tolerance determination with respect to cell size would be another good project for this lab.

The fourth reviewer asks if the failure mechanism on the overcharge test could be Li plating rather than incomplete shut down. For this kind of test, it would be good to have reproducibility tests. It may help to have a better understanding.

The fifth reviewer remarked progress seems good, although the methodology for internal shorts does not seem to have had much success this year (it's a difficult task, of course). The insight into gas generation is something that SNL is uniquely able to bring to the safety studies and their findings that heat output does not correlate with gas generation are very important. The reviewer thought the total amount of work they presented seemed a little low, but it's hard to judge from a 20 minute presentation. The reviewer suspected they do a lot of work for industry that they can't talk about, but we have to rate them on what was presented. Their cell capacity is very low for an 18650, even allowing for the fact that most cells for HEV applications have to be designed for high power, not high energy (maybe PHEV need to be a compromise construction). But with 2.8Ah 18650 cells being readily available, their input capacity just seems too low and needs to be increased.

The final reviewer commented on the good results in several areas. The reviewer was unsure about how the internal short mechanism is designed to work, but the reviewer understood the concept. The LiF/ABA electrolyte safety improvements are significant as well as  $AlF_3$  coating. The reviewer would like to see an assessment of other attributes of these new materials/modifications.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Reviewers generally saw good collaboration. One reviewer remarked there was great collaboration by this lab. The second reviewer observed a broad range of collaboration with other labs. Enlisting suppliers and other cell manufacturers might help. The LiF/ABA electrolyte should be tested in collaboration with others to understand what trade-offs are made for power, cold-cranking etc. as its conductivity seems too low. A third remarked that there are several useful collaborations with other DOE laboratories and more importantly with the commercial materials suppliers and industrial partners. A fourth reviewer thought collaboration with different labs and suppliers is good.

The final reviewer saw a good link to customers. This reviewer added that it was hard to say from the presentations, but he questioned the quality of the link with ANL. These two groups have to work very closely together to understand how safety aspects of materials affect actual cell safety. Maybe this is happening, in which case this point is moot, but the impression the reviewer got was that once ANL shipped samples to SNL, ANL was not that involved any more (a "throw it over the wall mentality"). The reviewer suggested at least reviewing this linkage. This reviewer believes Sandia needs to work much more closely with the separator companies and

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consumer standards organizations to help devise better test methods for resistance to internal shorting. While this reviewer believed they are involved in the auto safety standards committees, a lot of the science and understanding is still coming out of the consumer battery programs, fueled by the huge scale of the business and costly recalls (almost a billion dollars for Sony's and Panasonic's recalls alone). Also, the U.S. Navy is undertaking a major review of lithium batteries (primary and secondary) after several serious incidents. Sandia needs to be heavily involved in those efforts if they are not already. Sandia should be the "node" for all this safety stuff and this reviewer didn't think they are.

## QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first reviewer commented that the internal short-circuit simulation is very important. It is one of the main concerns of the whole industry today and it requires a lot of focus.

Other reviewers made comments about possible future activities. The second reviewer remarked that the future plans are in line with the proposed objectives. The reviewer would like to see SNL concentrate on identifying test methods that can be used to accurately predict the abuse response of various cell formats based on the results of one cell size (i.e. 18650). Another project could involve a similar approach with soft pouch cells. Another reviewer said that all cells containing coated materials should be examined also at end of life to evaluate the efficacy of the coating.

The fourth reviewer remarked overall that this is a good plan, but the reviewer would like to see more emphasis on finding a good way to create and test for internal shorts. This is critical for the industry and developers, even though it may not be as glamorous as developing new materials. SNL is by far the best lab to do the former, not necessarily the latter. The team needs to work on reaching out more to various organizations mentioned above.

The final reviewer remarked, the future studies involve continuing efforts to i) improve the thermal abuse tolerance of Li-ion cells though cell-level abuse tests with different materials (e.g., AlF<sub>3</sub>-coated NMC and Al<sub>2</sub>O<sub>3</sub> –coated Gen2, ii) improve overcharge abuse tolerance in full cells with new materials and additives, iii) demonstrate reduced electrolyte gas generation and improved thermal properties with LiF/ABA electrolytes, Fluorinated-LiBOB additives with different chemistries, iv) establish the electrode fabrication methods for new materials, and v) continue the internal short trigger tests, and support NREL's abuse model for Li-ion cells. The proposed studies address the technical barrier adequately.

### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

The first reviewer has visited this lab twice and, knowing that this is the premier safety-testing site in the U.S., would highly recommend additional funding, if possible, to make it the world's best lab in its category. The second reviewer thought that based on the stated goals, the resources are sufficient. However, given the level of interest in the larger format cells and the need for testing of the real cells the resources are limited. The third reviewer commented that this is a fairly costly program, but the work they do cannot be done cheaply. The reviewer recommended continuing at this funding level. A final reviewer remarked the budget of \$770 k per year looks marginally excessive (around \$550 K may be appropriate).

## Overcharge Protection: Thomas Richardson (Lawrence Berkeley National Laboratory)

### **REVIEWER SAMPLE SIZE**

This project had a total of 6 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Several comments emphasized the importance of this work. The first reviewer said this could be an important contribution to the stability/safety of the packs. The second reviewer remarked that intrinsic overcharge protection is a critical safety aspect of Li-ion batteries. A third commenter remarked Li-ion cells do not have builtin overcharge tolerance and need to be balanced through external electronics. Attempts to achieve built-in overcharge protection though redox shuttle haven't been quite successful. The objectives of this program are to develop a reliable, inexpensive overcharge protection system using an electroactive polymer for internal, selfactuating protection. Improvement in safety is crucial to a widespread use of Li-ion batteries for PHEV that will reduce the petroleum consumption, and pave the way towards petroleum replacement. A fourth remark was that this could be developed into an important safety feature (and less importantly a charge leveling device for maintaining good pack balance).

A final reviewer remarked there is a concern about the potential of a PHEV or EV system being overcharged while connected to the grid,

resulting in an abuse situation and decreased safety. This project attempts to reduce the potential for overcharging, which is in-line with the DOE objective of introducing this technology into the transportation market. The concern is that there is added cost for this at the cell level and the system cost in most cases will not be reduced, but more likely increased with the introduction of this agent into the system. The overcharge external hardware protection devices that are identified will most likely remain. The agent will become another protection device, rather than replacing existing ones-adding to the cost. Finally, the overcharge abuse condition, while one of the most undesirable in an automobile, it is also one of the least likely, because of the controls identified. It will be difficult to supplant something that is working.

### QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The first reviewer thought the approach to the stated objectives is right on line. The second reviewer remarked on the very interesting approach of electroactive polymers for cell balancing and overcharge protection. Another reviewer thought the project was a very well-focused work. The PI and his staff have clearly thought this through all the way to implementation, even though that will be a long way off course. Having a safety device that is sensitive to voltage rather than temperature opens up avenues for a new approach to overcharge protection.

The fourth reviewer has followed this field for a long time and still remains unconvinced that we will have any practical solution to this issue using this approach. The fifth reviewer thought this is an intriguing idea to impregnate the separator with an electroactive polymer that provides the bypass function during an overvoltage/overcharge. Questions to be addressed: What's the impact on round-trip coulombic and energy efficiency? What's the impact on cell self-discharge?



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Another reviewer noted the approach focuses on incorporating a shorting agent, which is a reversible, voltage-activated polymer impregnated in the separator between the current collectors but is an external component connected parallel to the cell. Studies are being carried out to optimize morphology and improve utilization of electro-active polymer composite with tunable redox voltage windows, and to investigate high-voltage polymers that are suitable for overcharge protection, and to explore alternative cell configurations to achieve maximum protection. The approach is indeed novel, but it is not clear how the positive and negative current collectors could be shorted even externally through such a device. Instead, it should function as a bypass with low enough resistance, but should return to high-resistance phase upon interrupting the charge. Also, the switching speed of the polymer from the insulator to the conductor state and vice versa might be slow, which may make this device ineffective.

### QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Many reviewers saw excellent progress. For instance, one commenter remarked the project was a highly qualified good approach based on the great progress made in the materials development front. These are very new classes of materials with new opportunities. This reviewer will keep an open mind and wait for these materials to be examined for their applications. A second reviewer remarked that very promising results were seen so far. Long life and robustness needs to be proven. It would be nice to show self discharge results to build some confidence on the concept. A third commenter saw excellent results, especially in view of the low funding effort. Their nanowire structures are very elegant and the conductivity results very good. This is important, because their conductivity goal is going to be fairly high if the safety device only occupies a small section of the cell volume, if they want to carry the high overcharge currents they are hoping to handle. The fourth commenter perceived that good progress has been accomplished in terms of demonstrating the concept with a few polymers with tunable redox voltages, external to the electrode/cell stack. Specifically, new electro-active polymers with different morphologies, using aligned and non-aligned polymer nanotubes, were evaluated. The sustainable current densities have been high and efficient configuration of the polymer is above the electrode stack, outside the electrochemical cell but within the cell housing. These accomplishments demonstrate a progress towards the project goals. The fifth reviewer said that it seems progress has been made at a slower pace since a few years ago; maybe the PI's need to elaborate on that more.

The sixth reviewer felt that the progress toward the goals is very good. However, based on the questions in Relevance, identifying another function for this agent would improve the potential of acceptance of this reversible soft-shorting agent at the commercial level. Has any testing been done to see how it performs under other abuse scenarios-in particular internal short circuit conditions, nail penetration, and crush? There appears to be a possible improvement in these areas using this agent and this should be explored.

### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

The first reviewer said that all the relevant labs are involved. The second commenter saw several useful collaborations with other DOE laboratories. Reviewers also commented on the relationship with LBNL and potential relationships with other labs. For instance, a reviewer encouraged the PI to find partners outside LBNL in other national labs, but more importantly in the supply chain and cell manufacturing. This is a great idea and deserves a faster path to potential implementation. Another reviewer thinks that SNL should be involved to verify the results and evaluate its performance in other abuse tests. The fifth reviewer felt that a lot of people at LBNL are utilized, but would want to see them working with or at least talking with cell builders about proof of concept cell designs. It is probably hard for them to get much collaboration outside of LBNL since the funding level is so low. The sixth reviewer saw a good collaboration network, however it would be interesting to collaborate with a group that can build prototype cells.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first reviewer wrote that it is a good plan going forward. However, while recognizing their points about the need for nanowires (good conductivity and access to electrolyte ions), the reviewer wanted to caution the PI about the risk of letting perfection getting in the way of "good enough." The reviewer is not convinced that we need a true nanowire assembly and think that this would make the

device much more expensive. Also, the reviewer would like to see the team partner with a cell builder - some are already interested in this area judging by the questions the PI made.

The second reviewer noted the future studies involve preparing composite separators with electroactive polymer nanofibers and evaluating their rate capability and cycle life. This reviewer suggest investigating other high-voltage electroactive polymers and optimizing their morphology for maximum protection and exploring other cell configurations for improved protection and lower cost. The proposed studies address the technical barriers adequately.

Other reviewers provided suggestions for future activities. According to the third reviewer, so many aspects of these materials need to be tested before we can say that they really work (cyclability, calendar-life, low-temperature performance, cost etc.) and we can say this class of materials is suitable or not. Another reviewer suggests the need to talk to separator companies and to plastic and gasket companies as well. The sixth reviewer wrote that proposed future plans concentrate on evaluation of only one aspect of abuse tolerance. The scope should be expanded to evaluate the effect of this agent on other more likely abuse scenarios.

### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

Some reviewers would like to see a funding increase, with one reviewer stating that it may be a good idea to "help" to scale-up that interesting concept. Another reviewer was really happy to see this concept resurrected from the BATT program. It is innovative and could provide an additional level of safety that is hard to do otherwise. For this reviewer, true safety comes from having redundant safety features - not just good cell designs. It could have implications even beyond the DOE's programs. Overall, this program needs a definite boost in funding and involvement of cell builders - would like to see this implemented in proof of concept cells ASAP. The current funding level is far too low for this project. They have shown good success and this work needs to be taken to the next level, which requires more time and money. Other reviewers felt the budget was sufficient, with one reviewer commenting that the budget of \$190K per year looks reasonable and adequate. Another reviewer agreed that resources for the work appear adequate for the future plans as stated. If work proposed in question 2 is addressed, more resources will be needed.

## High Energy Density Ultracapacitors: Patricia Smith (Naval Surface Warfare Center)

### **REVIEWER SAMPLE SIZE**

This project had a total of 4 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Several reviewers saw this as a relevant project, with one reviewer commenting that higher energy supercapacitors may be a good combination with Li-ion batteries. Another reviewer stated that Li-ion batteries do not offer high power densities, as required for HEV and PHEVs, and it would be beneficial to combine them with ultracapacitors, to extend the battery life, enable cold engine starts and reduce battery heating. The objective of the present study is to develop electrode/electrolyte materials that will enable an ultracapacitor to meet power assist and regenerative braking goal, i.e., 15-20 Wh/kg, 650 W/kg at cell level, operational temperature of 30 to 50°C, 750,000 -1,000,000 cycles, and survivability temperature range of -46 to 65°C. Such improvements in capacitors will enable a widespread use of Li-ion batteries for HEVs and PHEVs, which will reduce the petroleum consumption, and pave the way towards petroleum replacement.

While the third reviewer felt that this may be good work, with the recent improvements in high rate batteries, this reviewer does not feel work on capacitors or even hybrids is justified for the vehicle



### QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

Reviewers felt the approach is good, though one reviewer commented that the power target looks low, and believes it should be much higher. Another reviewer felt these approaches adequately address the technical barriers outlined here, and summarized that the approach involves i) identifying high capacity/capacitance electrode materials to increase Wh/kg, ii) developing stable electrolyte systems with wide electrochemical voltage window, temperature range, good cycle life, and iii) fabricate and evaluate prototype capacitors in order to assess energy density, cycle life, self-discharge and safety. The third reviewer remarked that lots of people are looking at this, and this reviewer is not sure that they really have any special edge over anyone else.

### QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

The first reviewer saw that good progress has been accomplished in terms of demonstrating specific energy ultra capacitors, specifically, i) Lithium ion asymmetric electrochemical capacitors show promise of significantly higher energy densities (>20 Wh/kg vs 5 Wh/kg) and low self-discharge (7% vs 17%) than conventional symmetric C/C capacitors, ii) Higher energy densities have been



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achieved with lithium ion capacitor prototypes utilizing carbon negative electrodes than with lithium titanate electrodes, iii) however, their low temperature performance is poor compared to conventional ultra-capacitors (activated carbon/activated carbon). The progress is consistent with the project goals. The second reviewer questioned, what is the explanation why KOH activated carbons have higher capacitance than steam activated ones: higher surface area, higher functional group? Was it the impact on life of those two activations methods? Specific power needs to be much higher than 3000W/kg to be able to associate supercapacitors with Li-ion batteries. The third reviewer stated that power density ratings are not much better than A123 and other batteries. LICs seem to have many of the safety/reliability issues of Li-ion cells - truly intermediate between conventional EDLCs and Li-Ion.

### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

According to one reviewer, there are several useful collaborations with material suppliers and prototype capacitor manufacturers. The second reviewer felt that the project needs better links to industry, although they said they are getting in with Maxell.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first reviewer felt that the proposed studies address the technical barriers adequately. This reviewer summarized that future plans are to i) continue carbon functional group analysis to seek their correlation, if any, with the electrochemical performance, ii) complete the assessment of lithium capacitors (LIC LiTO), and iii) investigate the voltage delay and perform three-electrode measurements for low temperature performance, to understand the SEI on graphite, develop electrolyte with wide electrochemical window and temperature range and ass the safety at the material as well as device level. The second reviewer felt that the proposed future research was okay, but this reviewer just doesn't see the benefit to the program even if they succeed.

### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

The first reviewer stated that the budget of \$250 k per year looks reasonable and adequate, particularly due to leverage from the Navy programs. The second reviewer recommended killing this project or shifting the bill to a stationary power program. This reviewer does not see relevance to vehicle technologies. Basically, even if they succeed, this reviewer did not see anyone using it in cars/trucks.

### In Situ Characterization of Fatigue Behavior of Electrodes: Claus Daniel (Oak Ridge National Laboratory)

### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Reviewers generally thought the project supports DOE objectives. The first reviewer stated that the project seeks to predict life performance for these PHEV and EV battery systems, and this goal supports the DOE objectives. The second reviewer states that we need good diagnostic tools to improve cell design. Cycle life is only one barrier, and this technique can help for calendar life as well. Another reviewer thought the project addresses one aspect of cycle life/lifetime, but is limited in scope. The third reviewer wrote that improvement in the cycle life will enable a widespread use of Li-ion batteries for PHEVs, which will reduce the petroleum consumption, and pave the way towards petroleum replacement. This reviewer also stated that Li-ion cell reactions involve intercalation processes, i.e., incorporation of lithium ions into lattices of carbon anodes and metal oxide cathodes, which results in an expansion of the lattice and the associated electrode degradation mechanically. The objective of this study is to develop in-situ tool to characterize mechanical degradation, such as crack initiation, crack growth, particle fracturing, particle loosening during cycling, gain fundamental understanding of accumulation of defects and resulting mechanical



degradation and thus correlate the mechanical degradation to capacity fade.

### QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

Reviewers addressed the innovative technique. For instance, the first reviewer called the technique novel, and commented that there is need to explore its full potentials. Another reviewer commented that this is a very innovative approach that, combined with other methods or techniques, can give outstanding result, and suggested not focusing on cycle life only. Calendar life is very important too. The third reviewer felt these approaches adequately address the technical barriers outlined here, and summarized that the approach involves utilizing acoustic emissions stemming from mechanical events to probe degradation during cycling and additional characterization techniques such as XRD, neutron diffraction, optical microscopy, and Raman spectroscopy are applied simultaneously in order to validate understanding. The fourth reviewer stated that cell failure reasons were identified and areas that need work identified as well.

The final reviewer stated that the project was an interesting build on work at Case Western (Sherson and a Japanese post doctorate student). According to the reviewer, the approach is worthwhile as a single method. However, the reviewer was not seeing the benefit of combining this with other techniques that others have already developed. Just do the sound work first and then see how useful it really is. The approach is novel, albeit somewhat resurrected. More to the point, it could give an additional window into what's happening to the electrode materials.

### QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL

### PROJECT AND DOE GOALS.

Reviewers focused on the innovative nature of the project. The first reviewer said that the work is very innovative and good, and this reviewer would like to see if this method can be applied to evaluation and prediction of a cell's abuse tolerance. The second reviewer commented that this is a very exciting technology that can have applications in the field as SOH. This could be a powerful tool. The third reviewer thought it was too early to expect much, but results look as though they could be useful.

Because it is a new technique, the fourth reviewer was unsure if it will be really effective in elucidating all the key underlying failure mechanisms However, there seems to be some potential out there to exploit this technique for better understanding of the electrode behaviors.

The fifth reviewer commented that progress is consistent with the project goals. This reviewer also wrote that moderate progress has been accomplished in terms of adopting and demonstrating acoustic emissions (AES) during cycling of carbon and silicon anodes, while also monitoring through XRD. Specific accomplishments include: i) Developed AES techniques using coin cells, which offer excellent signal transmission and cycling reproducibility, and ii) Added complimentary characterization methods (in-situ and ex-situ) in order to understand physical evidence of emission. Some of the useful scientific observations are: i) Emission frequency may allow for distinguishing the source of cracks, ii) Mud crack theory is not applicable to non-thin film electrodes; most cracking occurs during lithiation and cracks may initiate in the core of the particles, and iii) Brittle intercalation compounds may not need to be nano-sized to significantly reduce cracking.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Not a lot of collaboration was shown so far, but this reviewer was eager to see all the potential future collaborations. Another reviewer noted collaborations with ORNL, but external collaborations haven't been mentioned explicitly. The third reviewer was unsure if much collaboration is needed at this stage anyway. Another reviewer was not very clear about the collaborations.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

Reviewers provided suggestions for additional future research. One reviewer would like to see future work to be done on thicker electrodes using the same materials; and, the results of a comparison of predicted performance using a new commercial cell and the same commercial cell type, but at its end of life. The second reviewer suggests that tests should be carried out at the beginning of life and end of life for cycled as well as stored cells, especially at accelerated test conditions, to see if the technique works. The third reviewer suggests looking at the effect of rate and especially temperature on the sounds they detect. Materials could become more brittle at low temperature and also greater diffusion gradients that build up at high rate might affect the processes that cause the sounds they are detecting.

The final reviewer stated that the proposed studies address the technical barriers adequately. The reviewer also noted that the future studies include: i) Validation of scientific observation on the crack initiation, ii) development of in-situ combination characterization, iii) gain an understanding of relationship between particle size and mechanical degradation, iv) extending the studies to cathodes, and v) develop New quantitative "fatigue" theory models will be developed in order to understand degradation accumulation and failure.

### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

One reviewer stated that the budget of \$300 k per year looks reasonable and adequate for this effort. The second reviewer though the budget was okay for now, but this work should be limited to focusing only on the sound wave work and not let them expand the scope into other areas.

## Low Cost SiOx-Graphite and Olivine Materials: Karim Zaghib (Hydro-Quebec)

### **REVIEWER SAMPLE SIZE**

This project had a total of 7 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Reviewers commented on the relevance of a SiOx anode. The first reviewer commented that Si and Olivine is one of promising materials, and another reviewer commented that a successful development of an SiOx anode would lead to an improved capacity lithium ion battery, which would be very desirable for the vehicle program. Also, a low cost olivine cathode with improved voltage would improve the cost per unit energy of the lithium ion system to good effect. The third reviewer noted that investigation of alternative higher energy anode materials and doped Mn phosphate cathodes could provide for lower battery costs for automotive applications. The fourth reviewer suggests that manganese phosphate would be a useful cathode material, and that silicon oxide anodes would be helpful. Another reviewer perceived that the project addresses the key DOE objectives on EV and PHEV batteries of low energy and life. The final reviewer mentioned that the project is focused on developing/evaluation of the promising electrode materials and diagnostics tools.

#### QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO



### PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

Reviewers had positive feedback on the approach. The first reviewer said the approach is clear and reasonable with acceptable steps, and that the work on SEI is important and requires effort and good collaborations. Another reviewer thought the approach is good in advancing the methods of making SiOx, but this reviewer would like to see some full cell work to give a fair comparison of the results with the state of the art. Likewise, the olivines work would be enhanced by the manufacture of 18650 cells at this point to see what difficulties might be encountered. The third reviewer stated that a lot of work is being conducted and a great progress shown in developing SEM/TEM capabilities and 18650 testing facility. Very important work has started on the electrode engineering. It is not clear if all these activities are within the project objectives or outside of the project objectives. Elaborating more on the activities covered by this program will help. Another reviewer commented that the PI is obtaining useful fundamental data using several different methods. The final reviewer noted that the binder study for SiO was done by several people already.

### QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Several reviewers felt the progress is excellent. One reviewer commented that the progress is excellent and well-presented. The use of cycle life graphs with a few tens of cycles is not significant for the projected targets. It is suggested to give more acceptable results for the optimization and selection of the materials. The second reviewer commented that the PI is making progress and showing useful data. It appears that the blended cathodes have some promise as useful cathodes. According to the third reviewer, a lot of work is being conducted and a great progress shown in developing SEM/TEM capabilities and 18650 testing facility. Very important work has started on the electrode engineering. It is not clear if all these activities are within the project objectives or outside of the project

objectives. Elaborating more on the activities covered by this program will help. While the fourth reviewer thought progress is good, the comparison with this year's report and last year was somewhat disappointing in the level of accomplishment. In particular, the lack of data for anode efficiency may indicate that the anode is far from ready for development. The reviewer would like to see the SiOx anode used in a full cell to compare results with graphite directly. The cycle life data will also be much more meaningful. The olivine materials seem to be ready to be made into full size 18650 cells in order to make direct comparisons with other lithium ion systems. The fifth reviewer felt that although result details provided are too limited, dual material and dual layer activities seem quite promising.

The sixth reviewer commented that voltage cutoff for lithium extraction from charged SiOx was 2.5 V. It will result in too low cell voltage and is too far from practical. At voltage cutoff of around 0.5 V (a more practical value) the delivered capacity looks twice as low. Cycle life shown for SiOx is still far below from the approximately 1000 cycles typically achieved for commercial Li-ion systems. The role of SEI was not shown. Data on LiMnPO<sub>4</sub> mixed cathodes look promising. However, should be supported by improved safety data. Another reviewer commented that the advantage of multilayer electrodes needs to be proven.

The final commenter explained that this rating is based on evaluating the project against the objectives stated. Excellent work and progress must be noted for the work presented that is not stated under the objectives. Maybe the objectives have to be modified to better reflect the work being conducted? This reviewer makes the following queries:

Anode work:

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- The objective of the project is to replace the graphite anode with lower cost and higher energy anode material. Replacing 50% of graphite with SiOx addresses energy increase objective. How does it address cost?
- Was the recipe for the electrode preparation kept the same for the study comparing the effect of the binder?
- How can you explain such significant difference in the 1st cycle efficiency?
- How can you explain such low cycle efficiency for the PVDF system in the 2nd cycle?
- Are you planning to study your selected anode/graphite/binder system performance at low/high temperature and at the higher rates
- Is the 1:1 anode composition is the optimum composition for the cost/energy objective?

Cathode work:

- The approach to improve performance of the cathode material requires additional process steps (wet milling, re-heating). How do these steps affect cost?
- The recipe for the electrode preparation has 25% inactive materials. How does this affect the energy density of the final cell?
- LiMnPO<sub>4</sub> synthesized by the hydrothermal method has reduced PSD, but it also has 30% irreversible capacity. How do you plan to mitigate this?

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Reviewers generally saw good coordination. One reviewer felt there was a good network of really functional collaborations to meet project objectives. The second reviewer stated that all collaborators are strong. Their particular contribution was not shown well. Another reviewer saw excellent breadth of international collaboration, and the fourth reviewer stated that the PI is working well with his partners such as Sandia. The fifth reviewer stated that during the oral presentations, PIs have always complimented HQ on delivering samples, etc on time. The final reviewer commented that there are some collaborations but it is unclear how they work together.

## QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

A reviewer thought the plan for the next period is acceptable and reasonably structured, and the second reviewer commented that the PI's plans may lead to useful results. Their molten state plans may bear fruit.

Other reviewers made suggestions about avenues for future research. One reviewer stated that it is important to continue to develop the 18650 facility, but it would also be helpful to other BATT investigators to be able to take advantage of HQ coating, and calendering equipment. The BATT program has never had much strength in this area and HQ could serve as important addition. The fourth reviewer stated that test of new materials in the 18650 cells has to show how well the objectives (cycle life and high energy) have been met. Low materials cost evaluation should be shown in more details. The third reviewer suggests that further exploration of LTO/LFP-based system, limitations, and benefits of elimination of formation process should be included.

The fifth reviewer commented it would help if future research is reflected in the project objectives, and the final reviewer stated the future work for SiOx-graphite is not clear.

### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

Respondents commented that resources seem sufficient. One reviewer commented that there are many results and activities well covered by resources and commitment, and the second reviewer commented that there are a lot of data and resources seem sufficient. Another reviewer suggests that the PI should work with Richardson at LBNL to try to gain a more in-depth understanding of the structure of his new materials.

## Cathodes: Michael Thackeray (Argonne National Laboratory)

### **REVIEWER SAMPLE SIZE**

This project had a total of 8 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The reviewers saw that the program meets DOE objectives. One reviewer noted that the program to develop new, high capacity cathode materials is very consistent with the increased energy density goals of the DOE program, and another reviewer commented the work on novel or improved cathodes is essential for the DOE objectives. The third reviewer stated that high-energy cathode is one of the main issues to achieve higher energy density. The fourth reviewer saw that the program is well balanced/designed and focused on the objectives. Another reviewer commented that the layeredlayered cathode is one of the more promising new technologies currently in development, and TiO<sub>2</sub> could be a means toward increased life and safety in line with automotive targets. The sixth reviewer stated that the project provides potential for fundamental advancement in energy capability of automotive battery active materials at both fundamental and applied levels. This work provides useful exploratory investigations of novel methods for improvement of active material synthesis/treatment/processing methods. The seventh reviewer noted that the PI is attempting to develop new cathode materials.



### QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

Several reviewers commented on the autogenic reaction. The first reviewer thought the PI's approach may lead to useful results, and the autogenic reaction approach appears to be yielding useful results. The second reviewer commented that one of the barriers of previous work was the inability to charge and discharge the new cathode materials at higher rates. The approach to coat the surface of particles with an insoluble stable material is a good one to stabilize the interface with the electrolyte. Other approaches such as the use of autogenic reactions to produce carbon-coated  $TiO_2$  for the anode material are newer and subject to further investigation. Finally the use of molecular modeling of spinel surfaces has promise to improve the stability of these materials in high temperature cycling. The third reviewer commented that the approach is well balanced between experimental material research, process optimization and modeling, and the fourth reviewer thought the approach was generally good, but autogenic synthesis does not seem to lead to a big win, at least in the field of battery materials. The fifth reviewer saw a clear, defined approach. The sixth reviewer commented that the program is focused on solving technical barriers very well; at least "back of the envelope" cost estimations/comparisons will be greatly beneficial vs. baseline chemistries. To the seventh reviewer, it seems object was too wide (high power, high capacity and low cost for PHEV and EV)

### QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Reviewers generally saw a useful technology and good progress. The first reviewer thought progress on cathode material protective treatment was particularly good in terms of meeting objectives. Modeling was helpful for understanding, but it needs experimental data supporting simulation results. The second reviewer commented that the accomplishment of obtaining a stabilized surface for the

composite high voltage materials is substantial. This has been a key barrier to acceptance of the material class. There remain several other barriers, however, which must be addressed before final acceptance of the material class can be accepted as battery materials for the PHEV program. Among these are the high irreversible capacity shown on the first cycle, which detracts from the cell capacity, the low temperature performance of the system, which has not been sufficiently investigated, the rate capability of the system which has been improved, but is still not as good as competing materials such as NMC, NCA, and others, and the cycle life of the system, which still needs demonstration in full cells, preferably in 18650 cell size. The third reviewer commented the research on various cathodes is giving extremely interesting results with real possibility to be close to the solution of technical barriers. Longer cycle life testing is recommended to better verify stability with respect to DOE targets. The fourth reviewer commented that the PI's nickel phosphate material appears to be useful. The fifth reviewer saw a very good quality of results, and asks the following: Is there is a plan to address high irreversible capacity in cathode and thus, energy density of the full cell? Is there is a strategy to eliminate the need for the formation cycles? Autogenic reaction was very interesting tool to screen different chemistries, could it be difficult to control/reproduce process parameters? The sixth reviewer stated the surface coating showed the performance improvement.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

The first reviewer commented that the increased collaboration with Northwestern University indicates an improved level of collaboration. A wider distribution of "best bet" cathode materials to others in the BATT program for competitive evaluation would be useful. The second reviewer stated that the PI is working with his partners. The basic modeling work may help him understand better the material he is developing. The third reviewer thought saw very interesting work on simulation of atomic structure of spinel: might contribute to the improvement of the first generation of the EV batteries. The fourth reviewer can see the collaboration successfully improve the powder.

The fifth reviewer perceived the coordination with other institutions is not clearly specified even if in the previous year it was mentioned. The sixth reviewer stated that the project needed more clarity on what collaborators are supposed to do and actually did, and the seventh reviewer states that a high-level (at a minimum) description of specific technical focus of collaboration with external active material suppliers is desirable and should be included in the future.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

Generally, reviewers saw evidence of a well-defined plan. The first reviewer thought the plan for the next year is good and concentrates on key technical issues, and while the second reviewer also saw a well-defined plan, this reviewer commented that the project needs more clarity on  $TiO_2$  anode improvement, particularly on stability increasing of highly lithiated  $TiO_2$  anodes with autogenic synthesis. The third reviewer thought the PI's plan for future work is well-defined and will yield useful results.

The fourth reviewer also saw that future work is well-planned, and makes the following suggestions: might want to consider comparing doping vs. surface coating on cost and long cycle life; and need to do energy estimation for the  $TiO_2/Mn$ -cathodes systems. The fifth reviewer suggests that to achieve the 40 mile PHEV goal, the materials need to have high rate capability and safety as well as capacity.

The final reviewer would like to see the deficiencies noted above, namely low temperature performance, rate capability and cycle life, addressed in a systematic way so that better decisions on material acceptance can be made. The emphasis on surface studies to enhance the properties is well taken and should be pursued.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

Reviewers thought the resources seemed sufficient for the planned work. One reviewer mentioned that the resources are welldeveloped and represent a fully-developed program, the second reviewer stated that the resources are adequate to the work and the excellent result achieved, and a third commenter noted that the work is high-quality. ENERGY Energy Efficiency & Renewable Energy

The Synthesis and Characterization of Substituted Olivines and Layered Manganese Oxides: M. Stanley Whittingham (SUNY-Binghamton)

### **REVIEWER SAMPLE SIZE**

This project had a total of 7 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Reviewers perceive the project as supporting DOE objectives. The first reviewer stated that cathode material research is essential to support DOE objectives, and the second reviewer remarked the higher capacity cathode is one of the important milestones to achieve the PHEV goal. Another reviewer commented that the potential advancements are incremental relative to the current state-of-the-art. The fourth reviewer noted that the project focus is to develop new cathode materials which have high energy density as well as low weight and low material costs, using synthetic methods of low cost and to develop such synthetic methods. The final reviewer noted that the PI is producing new cathode materials.

### QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The first reviewer remarked that the PI's approach is appealing

because he is trying to develop materials that will intercalate more than one mole of lithium. Another reviewer commented that the approach is clearly centered on key barriers and with interesting proposed improvements.

The third commenter remarked that the NMC ratio is well studied and issues have been identified already. According to the fourth reviewer, the search for cathodes capable of cycling two lithium ions promises higher energy. However, extraction/insertion of two Li ions typically happened at too wide range of charge/discharge voltages (from  $\sim 5$  V to 1 V). This problem has to be addressed.

According to the fifth commenter, optimization of NMC composition for automotive or other applications is best left to viable materials suppliers at this stage of NMC implementation worldwide. The investigation of two electron materials is valuable.

The final reviewer remarked that all three approaches have the possibility of obtaining >200 Ah/kg in cathode capacity, which would meet DOE goals if successful. This reviewer summarized the approaches. 1) optimize the composition of NMC to minimize the cobalt proportion while maintaining capacity and rate capability, 2) investigate substitutions on phosphorus lattice of LFP to obtain better energy and rate performance and 3)investigate new materials that have the capability of intercalation more than one lithium ion per metal center.

### QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Reviewers saw evidence of good progress. The first reviewer remarked that the progress is good and well documented. There is no specific activity described on abuse tolerance of the selected materials. The second reviewer commented there is good progress on  $LiMO_2$  materials with ~200 Ah/kg and high rate. Two electrons materials (vanadium compounds) are at an infant stage. The third commenter wrote that the PI is making progress on developing novel, new cathode material.



The fourth reviewer stated that the 442 NMC material seems to be an optimum composition to maintain the rate capability of 333 composition and still reduce the level of Co. The goal of high capacity however, can only be met by charging to higher voltage levels which has caused capacity fading with cycling. The program should be extended on this family of materials by seeking methods to moderate the capacity fade when charging to 4.4 V or above. The final reviewer commented that people did not use high Ni content material because of the stability. PI should also focus on it, and should identify the pros and cons more clearly for each material.

### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Reviewers commented that collaborations are clear. A reviewer mentioned that the network of collaborations is clearly presented and justified, and the coordination seems to be very good. Another reviewer commented that the PI is working well with his partners.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first reviewer commented that the PI's plan to continue working on cathode materials with vanadium may yield useful material, and the second reviewer commented that the plan for each of the material class is reasonable. Some attention to stability tests and safety aspects is recommended. The third reviewer stated that oxides and olivines are already well developed at commercial scale. This reviewer asks if it would be better to concentrate on what is most promising and new. Two electron compounds? The fourth reviewer similarly queries if the focus should be on two electron materials. The fifth reviewer stated that the PI should identify the pros and cons more clearly for each material.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

The first reviewer stated that planned and required resources seem well justified by the work and the results presented, and the second reviewer commented that the PI's funding is sufficient. The third reviewer commented that they can do more failure mode analysis for each material.
# Stabilized Spinels and Polyanion Cathodes: Arumugam Manthiram (University of Texas at Austin)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 6 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Reviewers generally perceived the work to be vital. The first reviewer commented the research and development of high voltage cathode materials would significantly impact DOE objectives, and the second reviewer stated that new preparations of existing cathode materials and new materials for cathodes for lithium ion batteries are important aspects of the lithium ion battery program. The third reviewer commented that the PI is producing stabilized spinels with high voltages that may be useful, and the fourth reviewer noted that the cathode is the most expensive materials in battery materials.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The first reviewer remarked that the approach is well focused on the selected barriers. All the steps from materials research to material preparation and characterization are clear and justifiable. According to the second reviewer, the PI's approach is useful and novel, and the



third reviewer remarked that the cation's self-surface segregation improving cathode material stability looks interesting and promising.

The fourth reviewer stated that the approach varies for each material under investigation. The work with 4V spinel materials involves mainly cation substitution for manganese to reduce the solubility of manganese to as low a value as possible to address the cycle life problem with this material. The work with 5V spinel materials involves the substitution of cations in the nickel sites to try to stabilize the material and present a reduced surface to the electrolyte to minimize electrolyte decomposition. The work with polyatomic anions is more exploratory in nature and tries to develop ways to obtain higher than one electron per transition metal to increase capacity.

The fifth reviewer commented that the project uses a third element to stabilize Mn; spinel was studied many times and the difference from the previous study is unclear.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

One reviewer remarked that progress is good and well demonstrated, and a second reviewer saw that the PI's ability to produce high voltage material is useful. The third reviewer remarked that oxyfluoride transition metal oxides have been found to have slightly better stability than pure oxide materials. It remains to show that the other properties of these materials are as good as or better than the oxides and that they can be made economically. The performance of spinels remains in a trade off situation that the cycle life can be improved, but only at the expense of the capacity of the positive. This is a serious shortcoming since the capacity of the spinel needs to be improved, even though it is a low cost material. Interesting results on iron substituted 4.8V cathodes have shown that the iron is segregated at the surface of the crystallites, which helps to stabilize the material against electrolyte oxidation. Some progress has been made in the mixed metal olivines to obtain better energy from some of the compounds. Also, some progress has been made in the silicate systems, but they do not approach the goal of two electrons per transition metal.

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The fourth reviewer commented that silicate cathodes are at early stage of development. The team needs to show advantages over traditional Li-ion materials, and self-surface segregation during synthesis looks to be the most promising result. The final reviewer stated that it is very important to identify where the third element is in the molecular structure but it was not clear in the presentation.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

The first reviewer stated that collaborations have been useful in terms of analysis of materials and results. However, the reviewer would like to see some of the best materials supplied to the LBNL group for cell builds to compare the materials and formulations on an independent basis. The second reviewer commented that the collaboration network is acceptable even if key collaborations on specific materials could be improved.

According to the third reviewer, collaboration looks like analytical service support with exception of discussion with Professor John B. Goodenough, and the fourth reviewer remarked that the collaboration with Professor Goodenough has yielded useful results. Another reviewer stated that collaboration with viable industrial partners would be beneficial, and the final reviewer commented that collaboration seems only in-situ diffraction equipment.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first reviewer remarked that future work concentrates on most promising approaches, and the second reviewer commented that the PI's plan for future work is on target. The third reviewer would like to see a big push made on high voltage materials since this is the most promising program in BATT for this area. The use of  $TiO_2$  or lithium titanate spinel as negative material will not succeed unless a high voltage positive of high capacity is developed in tandem. The other work should continue as well to map out the field, but with lower priority. The fourth reviewer commented that the spinel study needs to be improved, and the reviewer does not think this direction can solve the issue significantly. The final reviewer remarked that future activities are well organized, but recommended better focus on the activities to verify and present better improved specific energy and stability (cycle and calendar life) of the selected materials.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

The first reviewer commented that the work seems well covered by the planned resources. The second reviewer mentioned that the PI's funding is too low, and the third reviewer remarked that the project needs more resources to see the fundamental issues for the spinel.

## Olivines and Substituted Layered Materials: Marca Doeff (Lawrence Berkeley National Laboratory)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 7 reviewers.

## QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Reviewers remarked that the focus on lowering cost reflected DOE objectives. The first reviewer remarked that the project to prepare low cost cathode materials for lithium ion batteries with higher performance and low toxicity is well within the DOE objectives to improve battery performance for hybrid electric vehicles. The second reviewer also remarked that low-cost materials are well functional to meet DOE objectives. According to the third reviewer, low cost processing and reduced cobalt could help these materials move toward the USBAC cost goals. A solution to the known performance problems with LMP could result in a cathode with all the advantages of LFP but 10-12% higher energy, although the authors seem to be using LMP only as a test case. The fourth reviewer remarked that the project provides potential for improvements in automotive battery basic electrode material costs and energy density. The fifth reviewer remarked that olivine is one of most promising materials, and the final reviewer commented that the PI is trying to make new materials for cathodes

#### QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO



# PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The first reviewer remarked that the project had a clear focus on technical barriers for the selected cathode materials. For the second reviewer, an important direction to lower cost is to reduce the amount of cobalt in NMC cathode materials without lowering the performance and cycle life. The approach to accomplish this is to replace some of the cobalt with main group elements that will not contribute to the capacity, but will improve rate capability in comparison to low cobalt materials without the additives. The preparation of LiMnPO<sub>4</sub>, a promising higher voltage olivine with potentially improved energy, has been developed using spray pyrolysis. This gives the possibility of a low cost synthesis and easy substitution of other elements such as magnesium, which is known to improve the properties of other olivines. The second reviewer mentioned that spray pyrolysis looks as promising method, and queries, what about mass production with this technique? Approach to substitute Co looks reasonable. The third reviewer remarked the PI's approach appears to be of limited utility, and the final reviewer stated that it was unclear what the issues are for less Co and how the PI wants to overcome the issues.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

The first reviewer found that the Al and Ti substitutions in low cobalt NMC (Ni and Mn at 0.4) were beneficial to rate capability because the lamellarity of the structure was improved over the unsubstituted material. Fe was found to have the opposite effect and also had bad effects on rate. The effect on electronic conductivity was mixed so the structure effect was believed to be dominant. For still lower Co (Ni and Mn at 0.45), the Al substitution could be carried out to 0.05 (reducing the Co still further) without harming capacity or rate. A decision should be made to focus on a more limited range of composition at this point and to better characterize the

material for rate, cycle life, irreversible capacity and cost. Spray pyrolysis results were not too encouraging because of low efficiency of discharge. This indicates a side reaction for the Mn phosphate materials which is not simply electrolyte reduction. The second reviewer commented that work is progressing well, and the improvements of specific capacity would be a significant added value. The work of spray pyrolysis is valuable and is adequately recognized. Some general information about economical aspects related to new materials and processes would be an added value. The third reviewer commented that barriers and objectives are well addressed, and posed the question of what the achieved (or potential) cost reduction due to Co substitution is.

The fourth reviewer commented that the PI's accomplishments are not encouraging, and the final reviewer stated the material synthesis was well analyzed but the material after degrade can be analyzed more.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Several reviewers commented that collaboration was clear. The first reviewer remarked that a long list of collaborators was provided. In this kind of structure/property investigation, it is encouraging to see maximum use of collaboration and methodology. The second reviewer saw a well-structured network with balanced key contributions, and the third reviewer remarked that there was a very good and clear description of collaborators roles. The third reviewer remarked that collaboration is clear.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first reviewer stated that the plan is good for ongoing project, and asks if Al and Ti are the only possible options for Co substitution. According to the second reviewer, the work on low-Co NMC should be expanded to prepare more material, particularly for Al substitution for more thorough electrochemical evaluation. This could be an important result, but the effects need to be sorted out better for both Al and Ti substitution. The spray pyrolysis may have beneficial effects on mixed Fe/Mn phosphates such as found by Manthiram and should be considered. The third reviewer remarked that the future plan takes into account the results achieved and previous years' reviews. There is not yet enough focus on stability and cycle life, which was intended to be implemented with collaborations (Vince Battaglia). The fourth reviewer remarked that the PI should consider changing directions concerning material development, and the final reviewer suggests the PI should focus on the failure mode more.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

Two reviewers found funding sufficient. The first reviewer commented that funding seems fine for the amount of work and the challenging targets. There is only one doubt for this reviewer, related to the statement in the second slide that the funding "supports one postdoc and one student." The second reviewer stated that funding seems sufficient. The third reviewer commented that the PI's funding should be reduced to cover the in-situ XRD only.

## Cell Analysis – High-Energy Density Cathodes and Anodes: Thomas Richardson (Lawrence Berkeley National Laboratory)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 7 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Reviewers saw that the work supports overall DOE objectives. The first reviewer remarked that work on cell analysis, cathodes and anodes are all valuable to the lithium ion battery program, and the second reviewer commented that key performances of Li battery electrodes are addressed and clearly support DOE objectives. The third reviewer remarked that the PI is developing useful experimental methods and anode materials. The fourth reviewer commented that high energy density and cathode is the most important milestone for the PHEV goal.

The final reviewer stated that high energy anodes represent a potential step change improvement in cell energy (Wh/L). The prelithiation looks promising, but the practical aspects of using prelithiated material must be addressed. Modeling/Li mapping work could also elucidate where to focus for improvements in formulation, electrode design, etc.

#### QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO



# PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The first reviewer saw a good approach in terms of analytical techniques applied for new and known materials exploration, and the second reviewer remarked that the technical approach is acceptable even if mostly qualitative: no clear quantitative targets are defined. The technical barriers related to kinetics and structure are reasonable. The third reviewer noted that the PI has provided charge distributions on electrodes that will be very useful. The fourth reviewer commented that the approach to see the inhomogeneous current distribution through the electrode area is very interesting.

According to the final reviewer, the approach to better understand the reaction distribution in the cathode uses synchrotron radiation from the LBNL advanced light source with an in-situ cell technique. The approach to anode alloy work is to develop a method of prelithiating the alloy to eliminate the irreversible capacity normally found which will improve cycle life and cell balance. The approach to cathodes is to explore nonolivine phosphates of transition metals to seek high capacity materials. These approaches are all directed at current barriers in present materials and in understanding of cathode operation.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Reviewers had positive remarks about the experimental methods. For the first reviewer, particularly impressive was experimental visualization of charge distribution at microns scale in the real electrodes. The second reviewer remarked that the PI has developed experimental methods that should be used by others. The third reviewer commented that the results and the applied investigation techniques are impressive and valuable. The progress is evident and needs only better investigation in lab cells.

The fourth reviewer commented that the synchrotron work has yielded outstanding results in analyzing the distribution of reactions under high charge regimes. It would also be of interest to examine the distribution under conditions of high discharge rate after slow charge to a uniform highly charged cathode. The work on anodes is also novel and may be quite useful in developing high capacity anodes with good cycle life and high first cycle efficiency. The cathode work did not yield successful results to date, but is worthy of continued work.

The fifth reviewer remarked that the mapping was good, work was okay on the anode, but the cathode work has not had much positive progress. The final reviewer commented that the data indicate the failure mode for cell analysis, and if PI can suggest the improvement, it would be better.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

The reviewers saw evidence of strong collaboration. According to the first reviewer, the collaborations are outstanding in both utilizing facilities for analytical work and in relating to models developed by Srinivasan. The second reviewer remarked that the collaborations are existing and well justified, and the third reviewer commented that the collaborators were strong with clear described contribution. The fourth reviewer felt the PI's collaboration with V. Srinivasan at LBNL is outstanding and useful, and the final reviewer felt the collaboration was clear.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

Some reviewers perceived that the future work will be worthwhile. The first reviewer remarked that the PI's plans for future work will bear fruit. The second reviewer thought the future work is well organized with recommendations for more close-to-product work on both electrodes to confirm specific characteristics in lab cells. Cycle life and abuse tolerance should be more considered or at least analyzed, because they are project addressed barriers.

According to the third reviewer, the future work will include more cathode analysis which is likely to be quite fruitful in the opinion of the reviewer. Also, anode prelithiation should be continued with best bet preparations shown by other workers. The cathode future work was not fully described.

For the fourth reviewer, the future plan is too general without specifics even for exploratory project, and the fifth reviewer sensed that the future work is a little too general for cathodes. The final reviewer suggests an increased focus on extension of study to electrodes harvested from commercial cells.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

Two reviewers remarked that resources are sufficient. One reviewer remarked that the PI's budget should be increased based on his excellent progress in developing useful experimental techniques.

## First Principles Calculations of Electrode Materials: Gerbrand Ceder (Massachusetts Institute of Technology)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 4 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The first reviewer commented that theoretical activities are complementary to the research and development of new materials able to support DOE objectives. The second reviewer remarked that the project is using first principles methods in an attempt to identify materials that could offer step improvements over the state-of-theart. The conventional approach ("fishing") results in very slow advances in the field. The third reviewer commented that the rate capability decides the usable energy for the material. The final reviewer mentioned that the PI is working on developing materials and theory to help develop materials for cathodes.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

One reviewer mentioned that the PI's approach is bearing fruit, while the second reviewer commented that first principles are really instrumental in assisting material research work with the capacity of



fast materials screening and selection. The approach is well focused in key technical barriers with fundamental studies. The third reviewer remarked that voltage and thermal stability is very important to improve the usable energy of the materials.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

A reviewer saw excellent results with clear descriptions. The second reviewer remarked that the PI's accomplishments in material development are valuable if they can be proven, reproduced by others. Their modeling efforts are useful. The third reviewer commented that it was interesting to see the results of the voltage and thermal stability. It was explained well why LFP has a bad rate capability in case of large particles.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Reviewers saw clear collaboration. The first reviewer saw well organized collaborations with key partners, and the second reviewer commented that the PI is collaborating with others in useful ways, especially having an exchange LBNL at MIT.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first reviewer felt that the PI's plans for future work are appropriate, and commented that he may want to provide materials to others for validation of his results. The second reviewer agreed that the plan is fine, and commented that of course, the attention and the focus must be on the new materials announced and not yet disclosed. The third reviewer commented that the future plan seemed little bit too general.

Energy Efficiency & Renewable Energy

**QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?** Responding reviewers commented that the funding levels are appropriate. First Principles Calculations and NMR Spectroscopy of Electrode Materials: NMR: Clare Grey (SUNY-Stony Brook)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Reviewers felt that this project definitely supports DOE objectives. The first reviewer remarked that this is essential research to support materials development for DOE programs. The second reviewer remarked that this is one of the few fruitful attempts at experimentally demonstrating what is happening in alloy anode materials and how it could affect performance. Extension of this work (including to other materials) could guide future work in this area. The third reviewer remarked that NMR is a good method to analyze the materials, and the final reviewer commented that the PI is using NMR to help develop and characterize new materials.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

Reviewers generally remarked that the approach is useful. The first reviewer said that the use of different characterization techniques is a



# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Reviewers saw evidence of progress in the work. The first reviewer remarked excellent progress with complete analysis of results. The second reviewer saw good insight into understanding of Si lithiation through simulations. This reviewer also commented that the NMR study on Li dendrites look interesting. However, it is not clear how it distinguished dendrites from mossy Li. The third reviewer thought the PI is making progress through her NMR work, but no useful new materials have been obtained. The fourth reviewer suggested that if PI can show some support data with different analytical technology for Li dendrite, it would be better.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

The first reviewer saw well established collaborations on key aspects of the work, and the second reviewer also saw very strong collaborators. The third reviewer remarked that the collaboration is clear. The final reviewer thought the PI's listed collaborators are sufficient. However, it is not clear how Ceder's modeling work is being used in her work.



# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

Several reviewers thought future plans are appropriate. The first reviewer saw a good plan for exploratory project. The second reviewer thought the plan for anode is very good while for the cathode it may be recommended to look also to the materials that are more interesting in BATT program. According to the third reviewer, the PI's plans for next year are appropriate. It may be useful for her to limit the scope of materials investigated. The final reviewer would like to see Li dendrite studied more.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

One reviewer remarked that the PI's funding is higher than needed to support her efforts. Another reviewer commented that resources seem sufficient.

## Development of High Energy Cathode for Li-ion Batteries: Jason Zhang (Pacific Northwest National Laboratory)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 6 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Several reviewers remarked that the work is relevant. The first reviewer remarked that high-energy cathodes are necessary to reach DOE objectives, and the second reviewer stated that alternative and new cathodes may accelerate the achievement of DOE objectives. The third reviewer remarked that the project is "out of the box thinking" towards supporting DOE objectives, and the fourth reviewer stated that the high energy cathode is most important milestone for PHEV goal. The final reviewer summarized that the PI is trying to develop new cathode materials.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The first reviewer remarked that the PI's approach to using organic chemistry to produce high capacity cathodes is useful. According to the second reviewer, the approach for the high energy cathode is clear. The PI should focus on the failure mode analysis also. The

third reviewer remarked that the approach is clearly centered on key technical barriers of the materials investigated. The fourth reviewer noted that multiple routes are used to address the barriers, which is good, and also suggests using energy vs. capacity to support selection of the materials under investigation. At minimum, cost estimation vs. baseline chemistries should be provided. The fifth reviewer has a concern that organic cathode materials usually have very low densities and that the PI should consider this property along with the specific energy in reporting results. This reviewer also summarized the work thusly: 1. New synthetic methods for LiMnPO<sub>4</sub> and Li<sub>2</sub>CoPO<sub>4</sub>F; 2. Characterization of materials; and 3. Synthesis of organic cathode materials

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Several reviewers perceived that results are good despite the project being preliminary. The first reviewer saw excellent results, and commented that the stability tests are still preliminary but interesting: more cycles are needed. The second reviewer commented that the PI has produced interesting new materials.

The third reviewer commented that this is a new program, so the results are not expected to be substantial yet. The preferred orientation of LiMnPO<sub>4</sub> may have some interesting effects depending on the orientation. Since olivines are 1D conductors, the long dimension should not be the conducting axis since this could increase the polarization and decrease the capacity. Cycling data should also be emphasized since this is a shortcoming of this material. It is not clear to the reviewer what the excess lithium in LiMnPO<sub>4</sub> compositions means, whether a new phase is formed and what its electrochemical activity is. The work on pyrophosphate is interesting, but it is not clear what the electrochemical effect is.

The fourth reviewer stated that the material analysis after synthesis was done well, and that the PI should focus on the failure mode analysis also. The final reviewer said the team should focus on rate capability and columbic efficiency.



#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

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The first reviewer saw acceptable collaborations for the work planned. Other reviewers made suggestions about future collaboration. According to the second reviewer, it would be useful to expand the collaborations of a new program such as this. Those described are certainly useful. The third reviewer suggests that the PI should develop collaboration with a group that could help him by using math modeling of the materials he is developing. Perhaps Ceder at MIT might be able to help. The final reviewer thought that the collaboration is clear but not much.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first reviewer saw a good plan for the prosecution of work, and recommends more focus on the investigation of life and stability of best selected cathodes. The second reviewer remarked that results for the work performed are well understood and considered in future work. The third reviewer commented that the PI's plans for the future are appropriate, especially his proposed vanadium cathode development work.

The fourth reviewer suggests that the workers should quickly assess the Wh/liter of the organic cathodes before investing a lot of work in this area. The nonstoichiometric LiMnPO<sub>4</sub> compounds are interesting, but need further structural work to better define them and interpret the electrochemistry. According to the fifth reviewer, PI should focus on the failure mode analysis also.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

Reviewers thought resources are sufficient. One reviewer remarked that resources are fine, but this reviewer is not clear if the project will continue after September 2010, because future plan refers to 2011 also. Another reviewer commented that the quality of work is good.

ENERGY Energy Efficiency & Renewable Energy

Inexpensive, Nonfluorinated Anions for Lithium Salts and Ionic Liquids for Lithium Battery Electrolytes: Wesley Henderson (North Carolina State University)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 7 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Reviewers saw the project as relevant. The first reviewer remarked that low cost, better performing electrolyte salts would be very important for the DOE goals, and the second reviewer concurred that new electrolyte materials are essential to support DOE objectives. The third reviewer remarked that the electrolyte is important to improve the life of battery. The fourth reviewer summarized that the PI is working on developing new electrolytes.

According to the fifth reviewer, electrolyte and electrolyte salt are an appreciable component of the total cost of Li-ion cells, and work is still needed to truly meet the demands of the automotive market in their entirety. That said, the work here seems to lack clear goals as to what is "success."

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?



The first reviewer remarked that the project had well-clarified barriers and approach, and the second reviewer commented that lithium salt typically is the most expensive part of electrolyte. It is particularly important to be introducing new, low cost salts. Ionic liquids are attractive due to their safe behavior at elevated temperatures. Reduction of their cost with new anions is important as well. The third reviewer stated that the two types of salts described in approaches have never been thoroughly investigated and seem to have a good chance of revealing some new structures which may be more stable that LiBOB. The comparison materials are very appropriate. The fourth reviewer thought the PI's approach is appropriate in his approach to find new electrolytes, and the fifth reviewer stated that the material property characterization is very good basic research for university. According to the sixth reviewer, the project could benefit from more focused and targeted approach (fewer materials to be investigated, greater intuition in choosing path).

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

The reviewers saw the results of the work as useful. For instance, the first reviewer remarked that the results are interesting and timely, even if some more relation to the project objectives must be given. It is not easy to estimate the effective progress. The second reviewer remarked that LiBOB solubility observations are useful, and the third reviewer commented that replacement of fluorine based salts with boron and cyanide containing anions looks interesting and promising. It is necessary to address potential cyanide generation at abuse conditions. Phase behavior of ionic liquids with lithium salts should be studied in more details for better electrolyte formulation optimization. According to the fourth reviewer, the material characterization seems done very well. We want to see the relationship with the electrochemical performance.

The fifth reviewer remarked that this is a new program, so results are not expected to be as substantial. The recognition of solid solvate formation is important as it often limits solubility of a salt. The low solubility for most of the prepared materials indicates a problem with the approach. Certainly, the inclusion of aliphatic substituents goes in the wrong direction as it probably causes localization of the negative charge on the wrong atoms (O bonded to B instead of carbonyl oxygen). The investigator should develop some new concepts to increase delocalization of the negative charge. The final reviewer thought that the PI has made some new materials, but it is not clear that the materials will be used in cells.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

The first reviewer remarked that the PI has developed good collaborations, which is to be encouraged, and the second reviewer commented that there is a well-structured network of collaborations. The third reviewer saw a useful scope of international collaborators, and added that collaboration with viable industrial partner would be useful. The fourth reviewer thought the PI's interaction with ARL is potentially useful.

Another reviewer remarked that collaboration with lab/company producing experimental or commercial batteries and providing new electrolytes evaluation in the batteries will be helpful, and the final reviewer suggests the project can collaborate more for electrochemical study.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first reviewer commented the future work is well aligned with the achieved results, even if there is the recommendation to keep in mind some of the project objectives (low cost materials and processes, which require dedicated work to be more evidently showed). The second reviewer remarked it will be nice to see new electrolytes tested in the real rechargeable battery to prove their advantages. The third reviewer thought the PI's plans for the future are appropriate, but he should compare some of his new electrolytes to existing electrolytes.

The fourth reviewer suggests referencing questions under results, and recommends that IL work should continue to seek new structures. The fifth reviewer remarked that scope should be narrower and more focused, and the final reviewer wants to see the characterization continuously.

**QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?** Reviewers remarked that funding is adequate. **ENERGY** Energy Efficiency & Renewable Energy

Molecular Dynamics Simulation Studies of Electrolytes and Electrolyte/Electrode Interfaces: Grant Smith (University of Utah)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 7 reviewers.

#### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The first reviewer commented that the project is theoretical work to support DOE objectives, and the second reviewer remarked this effort is mainly directed at better understanding the interfaces and interactions among phases in lithium ion batteries which should contribute to improving lithium ion batteries.

The third reviewer thought that while there is interesting learning here, it was not clear exactly how this work impacts the auto electrification goals. Another reviewer commented that the modeling seems more important for further material development. The final reviewer summarized that the PI is trying to understand better the formation of the SEI layer through modeling.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The first reviewer commented that the approach includes modeling the electrolyte phase, the SEI, the electrode-electrolyte interface and lithium intercalation/deintercalation. These are the key issues in lithium ion batteries. The second reviewer commented that molecular simulations appear to be a powerful tool for deeper understanding of battery related materials/electrodes performance. The approach needs more direct correlation with experimental data: at least, correlation was not well shown in the presentation. In response to the question, the third reviewer said this was a limited but key study to understand technical barriers in electrolyte effects. Another reviewer couldn't get how the breakdowns in the current performance of the working models would be overcome. The fifth reviewer remarked that project focus is too broad. The sixth reviewer stated that Smith's approach seems to be biased toward specifying the structure of the SEI a prior as opposed to not doing so. The final reviewer noted that SEI is very difficult to investigate so the modeling may be the good method.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

The first reviewer saw good results with clear progress. According to the second reviewer, the project shows good insight into materials performance for a wide (maybe too wide) range of conditions and areas. Again the data should be correlated with experiment at least semi-quantitatively. It will be nice to estimate area specific impedance for electrodes interface (ohm\*cm<sup>2</sup>) and its activation energy. Too many items are under consideration, so analysis for every item can be quite shallow for the given time and resources.

The third reviewer commented that detailed QC computations show that the formation of the SEI is likely to be more complicated than previously thought. The opening of a PC or EC ring after formation of the anion radical at the negative electrode is the critical step



and it seems to occur in a different way. The predicted longer lifetime of the anion radical is a key finding and effects the way subsequent reactions occur.

Another reviewer remarked that the PI has not achieved a better understanding of the formation of the SEI as planned, and the final reviewer wants to see not only the current mechanism analysis but also new electrolyte system based on the modeling data. Also the SEI composition that is the goal for this project was not clear.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Some reviewers saw good collaboration, with the first reviewer remarking that the broad collaboration the PI has is very important for this project. To have an impact, the experimentalists need to have an appreciation of the results to direct further experiments. The second reviewer saw an acceptable collaboration network, and the third reviewer thought that collaborations are strong, but the presentation needs more clarity on collaborators' contributions.

The fourth reviewer suggests the PI should be collaborating with other theoreticians (G. Ceder or P. Balbuenia, for example). The final reviewer remarked that the collaboration was not clear.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first reviewer remarked that future work plan looks promising, and the second reviewer concurred that the future work is well aligned to the achieved results. The third reviewer saw a good plan, with exception of whole-cell (anode/electrolyte/cathode) simulations. It is not clear what will be the whole cell simulations output (cycle life, energy density?).

The fourth reviewer suggests addressing shortcomings prior to moving to other systems. Another reviewer suggests that greater focus and more targeted effort is needed. The sixth reviewer suggests that the PI needs to focus on the SEI as opposed to attempting to model the entire cell, and the final reviewer stated that we want to see also the experimental data to compare with the modeling.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

One reviewer commented that funding is too high for the amount of useful results. Another reviewer remarked that resources seem insufficient.

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In Situ Characterizations of New Battery Materials and the Studies of High Energy Density Li-Air Batteries: Xiao-Qing Yang (Brookhaven National Laboratory)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 6 reviewers.

#### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The first reviewer said structural studies are important to understanding the operation of lithium-ion battery materials, while another said the lithium-air technology is one of the candidates for the next-generation battery. A third reviewer said the fundamental characterization work for most advanced materials in support of battery research and then DOE objectives. The final reviewer commented that Yang is trying to develop useful experimental techniques for studying potential material for cathodes. He is also working on lithium-air cells.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

Comments were mixed in this section. The first reviewer said Yang's approach is to use XRD to characterize electrode materials. Another said they suggested various X-ray analytical techniques directly

In Situ Characterizations of New Battery Materials and the Studies of High Energy Density LI-Air Batteries Energy Storage Xiao-Qing Yang (Brookhaven National Laboratory) 5.00 4.00 3.00 2.00 1.00 0.00 Approach Tech Accomplishments Collaboration Future Research Weighted Average Sufficient Excessive (33%) Yes (100%) Relevant to DOE objectives Sufficiency of Resources

address cathode materials performance issues. A third reviewer said the structural studies involve x-ray light sources at BNL, which gives unique capability to this group. In-situ work has proved valuable. TEM and SAED methods have also been developed to expand the structural determinations. New solvent types have been developed to affect the electrochemistry. A fourth commenter said a clear and complete approach is well focused on key technical barriers for assisting the overall BATT subprogram research on new materials for conventional Li and for Li/air systems. The complete characterization activities were finalized to also support internal material research.

Another reviewer said the degree of effort on lithium-air is not useful. The third reviewer said the objective of this test is very wide: this reviewer was not sure why the cathode characterization and lithium-air study were done in the same program.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

The first reviewer said the materials are characterized very well during charge and discharge, while another observed an impressive amount and quality of results and information giving significant inputs for further research and development. To-be-followed work is that on lithium-air and GDE. A third reviewer said studies have shown an unexpected two-phase region between the two plateaus of LiFeMnPO<sub>4</sub> materials. Studies of ANL materials are ongoing. The use of small pore carbons gives premature polarization of oxygen electrodes due to precipitation of lithium oxides and blocking of pores. Larger pore sizes and treated carbon to discourage immediate precipitation are recommended. This reviewer added that new anion receptor solvents have been developed, which are capable of forming a better SEI than previous solvents.

Another reviewer said that very useful knowledge has been generated on the behavior of various cathode materials. Boron compounds as SEI modifiers look promising. However, this reviewer added, their application for lithium-air to dissolve lithium oxides through complexation will require too high an equivalent weight of additives and dramatically reduce specific energy. The final reviewer commented that Yang has not developed exciting new information about materials or lithium/air cells.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Comments were generally positive in this section. The first reviewer said the group continues with very wide collaborations, while another noted very strong collaborators and well-defined collaboration plan. A third reviewer said the degree and breadth of international collaboration is excellent. Another reviewer said Yang is working with others to help improve his results, while a fifth commenter said the collaboration was clear and well coordinated. The final reviewer said the project is extremely dependent on a well-organized network of collaborations, which are involved in many ways: sample suppliers, cooperation on novel material and systems.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

Comments in this section were mixed. The first reviewer said the future plan seems well-established, while another said plans for further work are consistent with past results and should give meaningful experimentation. A third reviewer said the prosecution of work is logically related to the results achieved. This reviewer added that even if it does not seem necessary, increased effort on lithium-air is strongly recommended.

A fourth reviewer said good plan for cathode materials study, but added that the advantages of proposed boron materials/additives for lithium-air system are not clear. Another commenter said lithium-air should be a minor focus of the activity. The final reviewer commented that Yang's proposed work should be changed to exclude lithium-air so that he can spend more time on his other proposed tasks.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

One reviewer said the resources seem adequate and related to the large effort, while another added that the resources seem sufficient. The final commenter said Yang's funding is higher than needed, especially if the lithium/air work is dropped.

# Solid Electrolyte Batteries: John Goodenough (University of Texas at Austin)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 7 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Comments were generally positive in this section. The first reviewer said solid electrolytes in combination with liquid electrolytes have opened new opportunities of high energy density batteries. Another commenter thought that advanced research for BATT subprogram in line with DOE objectives, while a third stated that Goodenough is trying to develop higher capacity cathodes and a better solid state separator. A fourth reviewer stated the solid state battery is one of the next generation batteries, while a final reviewer said yes, the project is supportive of DOE objectives but with huge barriers.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

Comments were mixed in this section. The first reviewer said there was clear identification of technical barriers with interesting solutions to be verified. Another said the approach is to develop new solid electrolytes that will be stable in water to allow high energy



liquid cathodes to be employed such as redox couples. A third reviewer said the proposed design does not solve the Li dendrites problem known for the Li/liquid electrolyte interface. Another said Goodenough has developed a very successful approach to meeting his goals in the past. It is not clear that he will be successful this time. The final reviewer said the approach for this study is not clear.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

One reviewer said Goodenough has not yet reached his goal of discovering a better solid electrolyte separator, while another doesn't see the sufficient test results and effort from this slide. A third reviewer said the results are mostly preparatory, showing good potential for improvements. However, the solid electrolyte identified is extremely interesting. Another reviewer commented that the work has proceeded to the stage of allowing a sealed cell to be made and optimized. A new finding is that the previously employed solid electrolyte (nasicon) is not stable with acid solutions presently used with lithium air technology. Thus, the system will not be stable and unusable long-term in present status for rechargeable batteries. The authors have also found that water and air cathodes are quite inefficient in comparison to transition metal redox couples of ions in solution. The final reviewer said it is not clear how milestone "Optimize components of the cell (Apr. 10)" has been met. This reviewer added that there are no cycling data and confirmation of materials stability.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Commenters generally agreed in this section. The first reviewer said they have not yet identified any partners or collaborators for the work, and another added that no collaboration is envisaged in this phase. A third reviewer said there are no partners despite very challenging objectives. A fourth reviewer said no evidence of external collaboration, while another could not see any collaboration.

The final reviewer commented that Goodenough is not collaborating with others; however, in his case, he has proven in the past to be extremely capable without collaboration.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first reviewer said Goodenough is planning to continue to search for a better solid electrolyte, which is good since he may discover such. Another reviewer said the PI proposes developing a new solid electrolyte that will not have solubility in the aqueous electrolytes that will carry the redox couples. This will necessitate revisiting the sealing on the anode compartment. A third reviewer said the focus on the solid electrolyte is reasonable, even if it is not much developed and clarified. Another commented that the future work is poorly written (figures instead of text). This reviewer added that it is not clear what will be done towards objectives and what will be actual author's contribution. The final reviewer said the future work is not clear.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

The first reviewer said no specific risk is seen. A second commented that Goodenough's funding is sufficient, while the final commenter disagreed, stating that the resources seem insufficient.

Nano-scale Composite Hetero-structures: Novel High Capacity Reversible Anodes for Lithium-ion Batteries: Prashant Kumta (University of Pittsburgh)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

#### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The first reviewer said Kumta is trying to develop a better anode material, while another said novel anode research is necessary for DOE objectives. A third reviewer said a high capacity anode is important to achieve PHEV goals. The final reviewer said high capacity/alloy anodes potentially represent a large step improvement in the energy density (volumetric) of Li ion cells if the significant life issues, voltage/hysteresis problems, volume expansion complications (pack design), and safety can be sufficiently demonstrated.

### QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

One reviewer said the focus is high on well identified barriers. A second commenter said there was an impressive range of proposed synthetic techniques to improve anode material performance, while



another said Kumta's approach is attractive due to the high surface area per unit volume approach. The final reviewer said Si material is not novel, and added that it is not clear how CNT can improve the cons of Si materials.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

One reviewer commented that Kumta has developed a useful material if it can be shown to cycle well. He needs to continue cycling his best material. It appears that the material does not have sufficient cycle life. He should compare his results to those obtained by others (Zhang at NC State, e.g.). Another reviewer said Si is a well-known material and it is important how Si can be used for a long time. PI should focus more on the life study. This reviewer added that 20 cycles is not enough. Another reviewer said relevant progress was made on novel anodes, and added that the stability should be verified well beyond 30 cycles. The final reviewer commented good progress on gravimetric and volumetric anode specific capacity. It is not clear what level of active material loading or capacity (mAh/cm<sup>2</sup>) can be reached with magnetron spattering. Is it practical for mass production? Capacity loss of ~0.1% /cycle offers only ~ 200 cycles to 80%. It is too far from 5,000 cycles.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

The first reviewer said the collaboration is clear, while another commented about the mixed collaborations with industry and national labs that were well described and motivated. A third reviewer observed strong collaborators, with a good description of their contribution. The final reviewer said Kumta should publish more with his collaborators.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

One reviewer said this is quite an ambitious plan, and added that the good thing is that materials will be tested in the real cells. A second reviewer said the next period plan is well organized and aimed at solving defined barriers. The use of BATT cathodes is strongly recommended to increase comparability and support research progress of the entire subprogram. Another reviewer said Kumta's proposed work is acceptable. The final commenter said the plan is clear but added that Si is not a novel material.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

The first reviewer said Kumta's funding is sufficient, while another said the resources seem sufficient. A third reviewer said the project is adequately supported. The final reviewer said that the budget/timeline slide is in contradiction with future plans. It looks that the project was 100% completed in 2009. This reviewer asked, what is the funding for 2011?

# Intermetallic Anodes: Michael Thackeray (Argonne National Laboratory)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 7 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The first reviewer said high-energy anodes are necessary to develop batteries for DOE goals, while another said a high capacity anode is important for PHEV goals. A third reviewer commented on the novel anodes that are relevant to DOE objectives. Another commenter said Thackeray is trying to develop better anodes for lithium ion cells. The final reviewer said new syntheses and electrode's design are keys for the commercial success of the Sn/Si based anodes.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

Comments were mixed in this section. The first reviewer said the approach is well focused on key barriers of metallic anodes, while another said the approach is very clear. A third reviewer said the approach is to develop anode materials with a few hundred millivolts EMF positive to Li. The purpose is to reduce the SEI formation and lithium plating problems. The emphasis is on tin and copper tin



alloys on copper foam. This reviewer added that the specific energies are not much higher than graphite, however, and this will make it difficult to establish a benefit of the technology in the reviewer's opinion. Other work on autogenic reactions to form tin embedded in carbon may have a better chance at achieving high energy, however.

Another commenter said the approach is good in addressing cycling performance of the novel anode material, but does not address the irreversible capacity issue: this is a very important consideration for energy density of the cell. A fourth reviewer said electrodeposition is reasonable approach with high flexibility. In itself it is a very old and well known approach. The presenter needs to provide more clarity on what is new/unique with this approach for this particular application. The final reviewer commented that Thackeray's approach does not appear to be new or novel.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Comments were mixed in this section. The first reviewer said excellent progress was made on materials and preparation processes. Another commented that the test results are well analyzed, but wanted to see the comparison with the regular substrate. A third reviewer said that, while this is a new program, it follows from a previous program which developed the copper foam carrier. While the cycling appears to be good for certain mixtures, the electrode capacity is on the order of 200 mAh/g, which is substantially less than graphite. The autogenic method has produced higher specific capacity (up to 800 mAh/g), but the cycling data is preliminary.

Another reviewer said the higher anode capacity was achieved only for first few cycles. The foam Cu matrix looks helpful; however, can far overweight active materials. Is it taken into account? A fifth reviewer said Thackeray's success to date with electrodepostion of anode materials is disappointing. The final reviewer noted a very good overview of the work performed earlier, and very good initial results for the work performed under the current project objectives, but rate of progress seems low based on the results presented. This

reviewer is just curious about the possibility of using the team's 3-D Cu-foam for depositing lithium and evaluating such Li anode for the rechargeable applications. Are the autogenic reactions reproducible in terms of physical properties of the reacted products?

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Comments were mixed in this section. The first reviewer observed a good choice of partners, while another said some collaborations are mentioned but not evident in the presentation. A third reviewer said the role of collaborators is not shown, while another said the collaboration is not clear. One reviewer said that collaboration with viable industrial partner would be beneficial. The final reviewer said Thackeray may want to collaborate with Martin at University of Florida (for example) to obtain insight into production of high surface area electrodeposited material.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first reviewer felt there was very good understanding of the challenges and opportunities, while another commented that the future work is well defined. A third reviewer said the future work was good for exploratory project. Another commented that future plans will focus on the continued work with copper foams and electrodeposited alloys. The reviewer would like to see a greater emphasis placed on higher specific capacity alloys studied. The other work will continue on autogenic reaction preparations, which offer greater promise. Another commenter felt that the next period plan is not very specific. Stability of developed anode materials needs more attention. A final reviewer said Thackeray's plan should be modified to include help from others on the electrodeposition project, or it should be dropped.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

Comments were mixed in this section. One reviewer said the resources seem adequate. A second commented that Thackeray's budget is excessive for this project based on the success to date. A third reviewer said the team might want to have more people to increase the rate of progress, and added that the team has a lot of potential and interesting ideas. The final reviewer said the resources seem sufficient.

## Nanostructured Materials as Anodes: M. Stanley Whittingham (SUNY-Binghamton)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 8 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Comments were generally positive in this section. The first reviewer said the search for low-cost, high-capacity anode materials is in the objectives of DOE for lithium ion batteries. Another added that anode materials must be improved to better support DOE objectives. A third reviewer said Whittingham is trying to find better anodes for lithium ion cells. Another commented the project is focused on developing materials with high gravimetric and high volumetric energy densities. The final reviewer said a high capacity anode is important for PHEV goals.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

One reviewer said there is a good identification of technical barriers, while another stated the approach is well explained and allows for exploration of other than Si/Sn-based electrode materials. A third reviewer said the approach including amorphous, nanostructured,

and composite materials for anodes is not brand new but still the most successful so far. It will be nice to show in some details how different is it from others in this project. A fourth reviewer said a lot of effort was expended on tin cobalt alloys, which has now been abandoned because of cost and material availability concerns. Also, it was noted that an effort on Al alloys has been abandoned because of poor efficiency and fade characteristics. This reviewer added that the present program will focus on pure nanophase tin and silicon alloys, and the reviewer agrees with this approach. Another said that Whittingham's approach could be focused more. The final reviewer said it is obvious that the PI should focus on the life for metal anode like Sn.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Comments were mixed in this section. The first reviewer said there were clear indications and excellent results aimed to drive selection and future work and choices. Another said good and clear progress was shown on Sn-containing materials. Well presented go/no-go criteria and data were seen. What is the expected cost reduction? A third reviewer said the results with Sn show the possibility of high efficiency cycling (at least with lower charge rates) and low cost. The right combination of properties still needs to be developed. Experiments resulted in the elimination of Sn-Co alloys and Al alloys from consideration as noted above. Si alloys formed by ball milling will probably need carbon protection to achieve the needed near 100% efficiency, however.

A fourth commenter said Whittingham's technical accomplishments are mostly negative, while another stated that the result is only charge-discharge performance. The reviewer thinks the PI needs more failure mode analysis to improve further. The final reviewer commented that the project end date is 12/31/10 and percent complete is "continuing" with more funding requested for FY11 suggesting low rate of progress vs. objectives stated for FY10. This reviewer noted interesting results for Li insertion/de-insertion rates for nano-amorphous tin. What was the electrode thickness (active material loading per cm<sup>2</sup>)? What is the density of these



materials (g/cm<sup>3</sup>)? Also, this reviewer wanted to confirm that data presented on slides 8 and 9 is for Sn-Co-C. It will help to better appreciate results if the current density is translated into the C-rates or electrode thickness is referenced. What is the approach to mitigate the irreversible capacity losses? In general, the better the cycleability of the nano materials, the higher the surface area, the lower first cycle efficiency.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Comments were mixed in this section. The first reviewer noted well established and reasonable collaborations, while another said the team had clearly shown collaborators' roles, but added the presentation needs more information on what they delivered. A third reviewer said Whittingham is working with others, and another said good partner selection. Another said the collaboration is not clear. The final reviewer would like to see more collaboration with others in the silicon field to take advantage of carbon-coating methods.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

One reviewer said the general focus is good, but would like to see more specifics. For example, this reviewer added, what approach is planned for nanoparticle protection to still meet cost/performance requirements? A second reviewer said adequate planning is observed based on results. The work on stability of anode materials needs more focus. Another noted most of the planned work is with tin, with little emphasis on Si. This reviewer would like to see a more equal balance with the two materials. Another reviewer said the plan is not clearly identified, while a fifth reviewer said the plan needs more specifics on protective layer: nature, method to create. The final reviewer said that Whittingham should consider ending this project.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

One reviewer said the resources seem unclear, and another said it is difficult to question on the resources as the objectives are very broad. The final commenter said Whittingham's funding is excessive based on the results obtained.

## Nanostructured Metal Oxide Anodes: Anne Dillon (National Renewable Energy Laboratory)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 7 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Comments were generally positive in this section. The first reviewer said high-capacity anodes are of interest for improved lithium-ion batteries, while another added novel anodes are functional to DOE objectives. A third reviewer commented on potential for improvements in both energy density and safety, which will accelerate implementation. A fourth reviewer said this work investigates alternative higher energy electrode materials, while another said Dillon is trying to find better anode materials for lithium cells. The final commenter said this kind of old material should be re-investigated with the combination of newer technology.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

Comments were mixed in this section. The first reviewer said that the atomic layer deposition approach, as well as hydrothermal process, looks very effective to meet objectives. Another reviewer

said the team has well-defined the issue for the materials and is trying to apply new technology such as CNT. This seems to be a very good approach. Another said the approach is to use metal oxide nanoparticles to investigate displacement reactions involving many electrons for rechargeable anode materials. For MoO<sub>3</sub>, hot wire CVD was used to prepare the nanoparticles and Atomic Layer Deposition (ALD) was used to coat the nanoparticles to achieve reversible reactions. This reviewer added that  $Fe_3O_4$  nanoparticles were also investigated using a hydrothermal preparation and single wall nanotubes to construct binder free electrodes.

A fourth reviewer said technical barriers were identified but not clearly focused in the described approach. Another said excellent work was done towards novel higher energy materials / electrode structures. However, there seems to be little to no focus on the practical limitations in some areas of the approach (cost/practicality of ALD, etc.). The final reviewer said Dillon does not appear to be making progress on this project.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

The first reviewer said relevant results well documented on both anodes. Iron-based anode is very promising. Cycle life is a key point and needs longer testing. Another commented excellent and well-presented progress. Specific capacity is great. Do these anode materials provide enough voltage differences with cathode to be practical? What was the average cell discharge voltage? Surface loading of surface capacity of electrodes was not shown. This reviewer added that, to get practical, it should be of 2 -3 mAh/cm<sup>2</sup>. Another reviewer said there are more issues that remain, but added that we can see significant improvement for the life. The fifth reviewer said ALD coatings applied to the full electrode containing MoO<sub>3</sub> materials allowed cycling at higher rates than seen before while maintaining reversibility. The ALD coating seems to insure continued contact between the active material and the conductive phase. The full cell using MoO<sub>3</sub> anode with an Argonne National Laboratory cathode gave high capacity, but the voltage was



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comparatively low. The specific energy of the cell was not reported, however. The use of SWNT with  $Fe_3O_4$  gave very high capacity anode materials, but again the voltage is somewhat positive for an anode material and the hysteresis between charge and discharge is high. The final reviewer commented that Dillon has not produced useful material.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

The first reviewer said the level of collaboration is very good and seems quite well coordinated. Another said there is good collaboration and it is clear, while a third reviewer said well-structured network of collaborations. The final commenter said there was a well-presented collaboration scheme with clear collaborator roles and deliverables.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

Comments were mixed in this section. One reviewer said the future work is defined well, while another had no problems with future directions. A third reviewer said examination of practical trade-offs (cost of ALD, cost reduction? of binder-free) for actual implementation should have a more significant focus. Another commented that the work is heading toward a final test cell design and a decision to go or not with displacement types of anode materials. Some of the techniques developed in the program could be usefully employed with alloy structures and other anode types. A fifth reviewer said the plan is acceptable but requires more details. The final reviewer said Dillon is working with others, but this line of research does not appear to be useful.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

Comments were mixed in this section. The first reviewer said the resources are sufficient. Another said the resources should be increased to extend material research with more cathodes. The final commenter said that Dillon's funding is excessive for this project, and added that it should be terminated.

## Development of High Capacity Anode for Li-ion Batteries: Jason Zhang (Pacific Northwest National Laboratory)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Comments were generally positive in this section. The first reviewer said novel anodes are in support of DOE objectives, while another commented that Zhang is trying to develop a new anode material for lithium ion cells. A third reviewer said the project is focused on the search for the replacement materials for the matured Li-ion chemistry and the means of manufacture using low-cost production methods. The final reviewer said the high-capacity anode is important for PHEV goals.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED,

#### FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

One reviewer said the approach is clear with well-identified barriers. Another stated that Zhang's approach is appropriate and novel. A third reviewer said clear understanding of the current technology limitations, and the project is well designed and integrated with other efforts. The final reviewer said using graphene sheet is new but



Si+CVD was done by other groups also. This reviewer doesn't think this is new technology.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

One reviewer said excellent results were shown with interesting materials. A second stated that Zhang has made progress in developing new anode materials using Si and carbon. His success today is encouraging. However, he should try his material in a full cell with a cathode as soon as possible. A third reviewer said excellent work and progress are demonstrated. This reviewer's only suggestion is to compare cost of the production methods to graphite cost production, not to thin film deposition methods. In the presentation materials, this reviewer noted, capacity is shown to be close to 6000 mAh/g, above Si theoretical capacity. How can the team explain this phenomenon? Also, the first cycle efficiency is very low – does the team have a strategy to mitigate it? This reviewer suggests reporting specific capacity based on the composite material, not just Si, and also suggests translating the current density into the C-rates or provide thickness/loading of the composite material: difficult to compare to other results. The final reviewer said the data seems interesting but the reference data that the team chose is also not good.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

The first reviewer noted good coordination of a group of collaborations. Another said the collaboration is clear. A third said there was good outreach to allow people outside of the BATT/ABR to participate. A final reviewer said Zhang is collaborating with others. However, he should work with someone who can help him make a full cell.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

One reviewer said the plan is consistent with the excellent results already achieved. It is recommended to analyze cost aspects and safety. A second reviewer said this is the right direction for the Si/Sn work, and added that it is very important to continue working on the binder P, important for the success of the commercialization of the Si-based technology. This reviewer would like to see more data in the future. Is the future work on SLMP as an anode targeting rechargeable or primary applications? A fourth reviewer said that Zhang's proposed work is fine except that he needs to add building full cells. Another suggested that the future work can be improved more towards to Si life issues. The final reviewer said Li-metal investigation should either be totally eliminated or should be a very minor aspect of future work.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

The first commenter said adequate resources were available. A second reviewer said Zhang's funding is sufficient, while another said the resources seem sufficient. The final reviewer said very good quality data and good progress demonstrated.

## Electrolytes - Advanced Electrolyte and Electrolyte Additives: Khalil Amine (Argonne National Laboratory)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

## QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Comments were generally positive in this section. The first reviewer said Amine is trying to develop better electrolytes, while another added electrolyte work is necessary to improve battery performances and support DOE objectives. The final reviewer commented that the electrolyte additive is a very critical key for the current lithium ion technology to improve the life and abuse tolerance.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

One reviewer said Amine's screening approach has merit, and another added that screening the additive by different reduction potentials is a good approach. A third reviewer observed a reasonable approach with some qualitative statement, but added that more specification of technical barriers would be preferable. The final reviewer said the proposed quantum chemical screening approach looks reasonable. This reviewer added that what is unique and innovative in this particular application is not clear.



# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

The first reviewer said Amine's discovery that LiDFOB has lower impedance and providing an explanation for same is very useful. A second reviewer noted a limited presentation with interesting results, while another stated that this is a long term program and this is just the first year, so not much progress is seen. The final reviewer said it looks like a very preliminary, early stage of the project. Does the number of 100 screened candidates really matter? What is the connection between material structure and quality of SEI formed?

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

The first reviewer said Amine is working closely with others, while another said the collaboration is clear. A third commented on the adequate collaborations. Another reviewer said there was a good collaboration plan and hopes it will result in experimental data feedback. The final reviewer said collaboration with a viable industrial partner is needed.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first reviewer said the future work is defined well, and another said the future work is reasonable. A third reviewer said Amine's plans for next year are appropriate and it would be helpful to have a better understanding of the SEI formation as planned. The final

reviewer commented that the team needs better focus on objective: predict functional additives that form a stable Solid Electrolyte Interface.

**QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?** One reviewer said Amine's funding for this project is appropriate, while the other commenter said the resources seem sufficient.

## Development of Electrolytes for Lithium-ion Batteries: Brett Lucht (University of Rhode Island)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 6 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Comments were generally positive in this section. One reviewer said electrolyte work is necessary to improve battery performances and support DOE objectives, while another commented that the project is attempting to address some of the key limitations with current electrolyte technology. A third reviewer said Lucht is trying to develop a better salt for use in high voltage cathode lithium ion cells, while the final reviewer added that new salt development is important to improve life.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED,

#### FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The first reviewer said Lucht's approach is appropriate, while another said the approach is well focused on barriers. A third commenter noted this new salt was considered for a long time after LiBOB, but not a lot of work was done. So this is an interesting program. The final reviewer said new salt introduction is a relatively



rare event for batteries and always gives more opportunities. What is the cost of new salt compared with traditional LiPF<sub>6</sub>?

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Comments were generally positive in this section. The first reviewer said there were interesting results on a novel electrolyte composition. The excellent accelerated testing work should be suggested for extension to other BATT projects. A second reviewer observed a well-presented and detailed description on a novel salt. Aging and calendar life issue are well addressed. Difficult to question on additive X. Another commenter said Lucht's success in finding an additive to form a protective layer on the cathode to prevent electrolyte oxidation is interesting. The final reviewer said the result is excellent, and added that it is much better than LiBOB.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

One reviewer said there was a well-balanced network of collaborations, while another commented on the good description of collaborator roles. A third reviewer said Lucht is collaborating with the appropriate people, and another added that the collaboration is very clear. The final reviewer said that collaboration with a globally viable consumer electronics Li-ion cell manufacturer should be pursued.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

One reviewer said the future plans are defined well, while another said the future work is well aligned with achieved results. This second reviewer added that focus on stability, accelerated aging and coating effects is recommended to be maintained. A third

reviewer said the plan includes investigation of cycling behavior of  $LiPF_4(C_2O_4)$  with Propylene Carbonate. Is PC the solvent beneficial for cathode? The final commenter said Lucht's plans for the future to improve further his electrolyte additives may be useful.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

One reviewer said Lucht's funding is sufficient, and another agreed that the resources are sufficient. The other remaining commenter said that the extension of accelerated testing to a large number of materials developed in BATT would require more resources.

## Bifunctional Electrolytes for Lithium-ion Batteries: Daniel Scherson (Case Westerm Reserve University)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The first reviewer said electrolyte work with safety features is necessary to improve battery performances and support DOE objectives. Another commented that electrolyte development is important to improve the abuse tolerance. The final reviewer said Scherson is trying to develop flame retardant additives for electrolyte use in lithium ion cells.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

One reviewer said well-identified barriers were shown, while another said Scherson's approach to finding and testing these flame-retardant ions is appropriate. A third reviewer commented that phosphorous and boron moieties have been known as flame retardants. This reviewer asked, is the salt most effective way to introduce them into the electrolyte? The final commenter said the approach for this program is clear, but needs to confirm the other performance like life.



# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

The first reviewer said Scherson's success in finding a candidate material for consideration for use as a flame retardant material is encouraging. His new spectroelectrochemical cell will be useful in this project and others. A second reviewer said they can work more with other groups to confirm the performance and abuse tolerance of the actual battery with this new electrolyte. Another noted complete characterization of the developed materials, with no evidence of the searched effect to improve abuse tolerance. Results of in-cells testing not clear. The final reviewer said development looks to be at an early stage, and added that more convincing data are needed on improved electrolyte/cell stability. It looks that salt's purity can be the issue. How is it addressed?

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

One reviewer said there were reasonable partners and their roles. The second said there were good collaborations but the involvement of other BATT participants to verify the effect in different Li systems and materials. Another commenter stated that Scherson's collaboration with industry is useful. This reviewer added that he may also want to interact with ANL, e.g., to test his new salt in lithium ion cells being tested there. The final reviewer said they can work more to do the electrochemical test.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first commenter said the future plan is defined well. Another said the planned future work is fine even if not clear, because there are undefined changes towards no presented completely new materials. A third reviewer said all bullets in the slide with the exception of the last one need more specifics. The final reviewer commented that Scherson's plans for the future are appropriate, but added that they seem to be limited in scope.

#### **QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?** One reviewer said Scherson's funding is sufficient, while the other commenter said they should have more electrochemical data to

confirm the product.
Performance and Safety of Olivines and Layered Oxides: Guoying Chen (Lawrence Berkeley National Laboratory)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 6 reviewers.

## QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Comments were generally positive in this section. The first reviewer said novel materials for Li batteries are relevant to DOE objectives, while another noted that Chen is trying to find useful cathode materials. A third reviewer said there is definitely a need for high energy density, safe and cheap cathode materials. The final reviewer commented that this kind of basic research is very good for national lab work.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

Comments were generally positive in this section. The first reviewer said that the approach is good with clear specifications on technical barriers of the selected materials. Another said this was a welldefined, classical academic approach correlating structure and performance. A third reviewer commented Chen's approach is appropriate. Another said the approach to improving the rate



capability is traditional method such as Mg doping. This reviewer added that Mg doping sometimes works to improve the life. The final reviewer said very good use was made of the LBNL characterization equipment. Question: the team's approach to improve thermal stability of LMP is through substitution of Mn with Mg up to 0.5 moles. How will this approach affect the capacity of the material?

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

The first reviewer said good progress was made but more focus on the addressed objectives is necessary, and added that the key properties for Li cells should be more outlined. Another commenter said it was good from the point of knowledge gaining. More guidelines need to be generated on layered oxides improvement/stability. A third reviewer said Chen's technical accomplishments are acceptable, but it appears that LiMnPO<sub>4</sub> may not be a useful cathode material. A fourth commenter said milestones should better reflect the objectives; difficult to judge the degree of progress of this project. Overall, the work is very interesting and useful. This reviewer added that the team might want to consider focusing on fewer topics or add resources. The final reviewer thinks the PI will do more electrochemical study for NMC333. This reviewer added that we can see the effectiveness of Mg doping.

### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Comments were generally positive in this section. The first reviewer noted well-organized collaborations, and another said that all collaborators are strong, and well organized. A third reviewer stated that Chen's collaboration is appropriate and she is publishing with her collaborators. Another commented on the good selection of partners, while the final reviewer said the collaboration is clear.

One reviewer said Chen's proposed future work is appropriate, while a second said that future work is defined well. A third reviewer observed a fairly good plan. A fourth reviewer said that future work is well outlined and will require significant efforts. The final reviewer said the plan lists activities but needs to be more focused on one of the objectives. Provide guidelines to design and develop electrode materials with improved energy density, rate capability, and safety, especially with regard to thermal stability.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

One reviewer characterized resources as adequate, and a second reviewer agreed the resources are sufficient. Another said Chen's funding is appropriate. The final reviewer commented that the project is 30% complete, while the end date is September 30th, 2010.

ENERGY Energy Efficiency & Renewable Energy

Positive and Negative Electrodes: Novel and Optimized Materials: Jordi Cabana (Lawrence Berkeley National Laboratory)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 7 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Comments were generally positive in this section. The first reviewer said that high voltage cathodes and high capacity conversion reaction materials are useful in trying to achieve DOE goals. Another said new material development is important to achieve PHEV goal, while a third reviewer said novel materials for Li batteries are functional to DOE objectives. A fourth commenter said Cabana is trying to find better materials for lithium ion cells. The final reviewer said the program is focused on the development of the high voltage spinels and investigates anode materials based on the conversion reactions for higher energy density batteries.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

One reviewer said this was a good approach with well-focused technical barriers. Another commented Cabana's approach is



appropriate. A third reviewer said there was a good selection of the characterization tools to understand the targeted correlations. This reviewer added that the team might consider partnering with companies specializing in high throughput sample production to have more effective use of the PI's expertise in characterization of materials. Another commented that the approach is fine but if the PI can work on more failure mechanisms, it would be better.

A fifth reviewer said the high voltage LNMO material is studied with structural methods which are linked to electrochemical properties. The conversion reaction of NiO and Cu-containing oxides and mixed metal oxides are studied with an emphasis on structural details of reactants and products. The final reviewer said the three bullets in the Approach slide offer certain advantages addressing the barriers, but each can be considered as a separate project. This reviewer asked, what are the connections between them in this one project?

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Comments were mixed in this section. The first reviewer commented that this is a new program which involved setting up a new lab in the process. The structural work looks quite strong and the prospects for learning about reaction mechanisms are good. A second commenter said Cabana's finding that non nano scale particles perform better than nano scale particles is interesting. Another reviewer said interesting results even if better focus on project targets should be advisable. A fourth reviewer added that the work looks like analytical characterization, not development. This reviewer added that other items (NiO conversion and Cu-M-O) are at very rudimentary stage.

Another reviewer suggested that the PI should do more failure mode analysis for LMNO materials so the PI can improve the synthesis method even further. The final reviewer said significant progress against the objectives and milestones is demonstrated. This reviewer noted the important finding about particle's morphology on high rate performance, and added that it would be beneficial to do safety assessment as well vs. nanostructures. Conversion reactions; would be helpful to have a table of theoretical capacities vs. practical capacities for the purpose of assessing the progress.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Comments were generally positive in this section. The first reviewer said the collaboration is clear, while a second commenter said Cabana's work with others is helpful. A third reviewer indicated a reasonable network of collaborations, while another said collaborators are strong with well-defined functions. Another commenter said good use was made of the NMR expertise. The final reviewer stated that it is useful to have substantial collaboration on a new program, and this one seems to fulfill the need.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first reviewer said the plan looks comprehensive and is attacking important problems, while another said the future work is well related to the achieved results. A third reviewer said Cabana's planned future work is appropriate. Another said the future work outline is clear and supports the initial findings, while a fifth reviewer agreed the future work is clear. The final reviewer commented that the plan needs more quantified clarity about what is expected to be achieved in terms of the performance for high voltage spinel phases.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

One reviewer said Cabana's funding is sufficient, and another agreed that the resources are sufficient. The final commenter simply stated good progress was demonstrated.

# Electrode Fabrication and Failure Analysis: Vince Battaglia (Lawrence Berkeley National Laboratory)

### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

## QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

One reviewer said cell standards for materials characterizations for Li batteries is relevant to DOE objectives, while another commented that Battaglia is developing electrode fabrication techniques for lithium ion cells.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

Comments in this section were mixed. One reviewer said Battaglia's approach is appropriate and useful. A second reviewer said the basic idea to have a common cell design and preparation for most BATT materials testing is the best approach to solve and verify technical barriers. Another reviewer said this was a very clear and well structured approach. However, failure modes may not be limited to those described in the approach. The final reviewer didn't believe this is national lab work, and commented that this is more engineering.



# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

The first reviewer said Battaglia's results for his formation studies are very useful, especially since cell manufacturers do not reveal such information. Another commented on good progress with a clear service to the BATT subprogram. This reviewer added that it is useful to have a standard cell design (not necessarily the best one) for comparing materials and preparation processes. Another commented that the correlation of cycle life with anodes mechanical properties is impressive. Is it possible to generate quantified criteria for electrodes mechanical properties? Data/mechanisms on cathode dissolution are not clear. This reviewer added that high voltage and ordinary electrolyte showed differences but the mechanism is not shown. Is it simply different cathode cation solubility? The final reviewer said the result needs to be analyzed with more scientific method. This final reviewer asked, how does the mixing order affect the performance?

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

The first reviewer said that the networking is one of the scopes of the project and is well organized. A second reviewer noted the good collaboration slide with clear roles and contributions. Another commenter said Battaglia is working with several collaborators from different areas of expertise and interest. It would be useful if he could work with cell manufacturers who are doing cell formation. A final reviewer noted some collaboration relating to supplying materials.

One reviewer said Battaglia's plans to work on stress effects will probably yield important information to assist in designing cells for maximum life time. Another said the plan related to the work already done and obviously connected to previous results. A third reviewer said "Work with Modeling group to figure out where stress is most important" sounds good for generation of quantitative criteria. The final reviewer suggested reconsidering the program.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

The first reviewer said the resource's adequacy is strongly related to the effective collaborations and the number of samples available from BATT participants. Another commented that Battaglia's funding is at an appropriate level. It may be useful to consider providing additional funds for him to use to interact more closely with a cell manufacturer. The final reviewer could not judge if the resources are sufficient for this program.

ENERGY Energy Efficiency & Renewable Energy

Microscale Electrode Design Using Coupled Kinetic, Thermal and Mechanical Modeling: Ann Marie Sastry (University of Michigan)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 6 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The first reviewer observed this was a modeling program to aid in electrode design to optimize battery performance, while a second commenter said modeling is useful to accelerate the development of Li technologies, which is required for meeting DOE objectives. A third reviewer commented that this kind of basic research is good for a national lab or university project. Another commenter said Sastry is trying to use modeling to help develop better electrodes for lithium ion cells. The final reviewer commented that the project is questionable, unless it enables 5V spinel, and even then, other materials are more promising.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

One reviewer said Sastry's approach is a valuable one because her results help cell manufacturers to understand better how their cells

work. Another reviewer said the 3D microscopic electrode model is a very unique technique. A third commenter said both 3D and 1D modeling are employed to obtain the maximum information about electrode structures, including conductivity factors. Another reviewer commented on the good integration of different aspects in the modeling work. A final reviewer said multiscale modeling looks to be the most effective way to understand whole battery performance. Modeling SEI as a key player for stability is a necessary thing to do. Thermo-electrochemistry is part of approach. The role of "thermo" is not clearly shown in the presentation.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

The first reviewer said Sastry's technical accomplishments are significant, and added that her findings for the diffusion coefficients will be useful to cell developers. Another reviewer noted the complete set of combined results with various applications in modeling work. A third commenter stated that the many variables in an electrode design result in various trade-offs in performance. This reviewer added that the results are quite reasonable in view of empirical electrode design studies and should lead to some optimal design criteria. Another reviewer said SEI layer experimental characterization looks important for further simulations. The tradeoff between ionic and electronic conductivities and its impact on porous electrode behavior and cell energy was well studied before. This reviewer asked, what did this development add to it? The final reviewer commented that the progress of this program is not clear in this presentation this year.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

One reviewer observed adequate collaborations, while another noted that Sastry is collaborating with several groups. This reviewer added that she should be encouraged to work closely with GM personnel working on projects of common interest. A third commenter



said the slide did not show too much on collaboration and collaborators' roles. The final reviewer can see some company's name, but the collaboration is not clear.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first reviewer said the plan is well related to the previous results, while another added that the plan is well focused on objectives. This reviewer asked if there are any plans to study nucleation mechanism and SEI forming new governing equations. A third reviewer said Sastry's plan to develop a model for the SEI formation is a good one, and added that it would be very useful to have a definitive explanation of how the SEI layers form on the anode and cathode. Another commented that the effects of SEI formation will be brought into the modeling considerations as it is known to have substantial effects on electrode conductivity. The final reviewer stated that the future work is too general.

**QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?** The final reviewer said there are no budget-related slides, and cannot answer the question on resources. ENERGY Energy Efficiency & Renewable Energy

Analysis and Simulation of Electrochemical Energy Systems: John Newman (University of California at Berkeley)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

One reviewer said modeling programs are directed to improve performance of high energy lithium batteries, while another said the study of degradation mechanisms is helpful in supporting DOE objectives. A third reviewer said Newman is attempting to explain processes that occur in lithium ion cells. The final reviewer commented that SEI and Li dendrites need to be understood to improve the battery life.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The first reviewer said there were well-identified technical barriers. A second commenter said Newman's approach is well established and useful, while another said it is an interesting approach to use redox shuttle for SEI study. The final reviewer said that rotating disk experiments are run to better understand shuttle mechanisms for



overcharge protection. A new program was instituted to understand the effects of shape change of lithium metal electrodes.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

One reviewer said significant progress was made in mechanism comprehension, and added that the same analysis could be extended to other materials. Another commenter said Newman's explanation of how lithium plating can occur in cells is very useful. A third reviewer said that ferrocene-ferrocinium couple was shown to be a highly reversible shuttle reaction and used as a model system. It was shown that development of passivating films on the negative electrode cause a shift in the kinetics of the shuttle reaction. The model of lithium metal shape change shows that there could be a problem with this phenomenon in a lithium electrode. A detailed model will be constructed. A final reviewer said the results of ferrocenium reduction were not explained very well in this presentation yet.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

One reviewer said the collaboration level is outstanding, as models are applied to actual system studies in various laboratories. Another commenter said Newman's interaction with others is useful to his work. The third reviewer said some collaborations are in place, while another reviewer could see some collaboration. The final reviewer said inclusion of collaboration with external industrial partner is excellent, but shape change in Li metal anodes does not seem like a pertinent area in which to involve an industrial partner. This reviewer added, don't stop collaboration, but change to more pertinent subjects if possible.

One reviewer said Newman's proposed work is appropriate, while another stated that the future work is defined well. A third reviewer said future plans are adequate to the work already done, and said more focus should be placed on alternative materials where to apply the same modeling approach. The final commenter said the modeling in these areas is quite difficult, but progress does seem to be likely. A better model of shuttle behavior could be quite useful in developing a practical material. The shape change problem will be studied with a detailed model.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

The first reviewer said Newman's funding is sufficient for this project, while a second reviewer commented that the resources are sufficient. The final reviewer said the presentation does not fully clarify the amount of work done in relation to the significant resources (budget?) used.

The Role of Surface Chemistry and Bulk Properties on the Cycling and Rate Capability of Lithium Positive Electrode Materials: Yang Shao-Horn (Massachusetts Institute of Technology)

Energy Efficiency &

**Renewable Energy** 

#### **REVIEWER SAMPLE SIZE**

U.S. DEPARTMENT OF

NERGY

This project had a total of 5 reviewers.

## QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The first reviewer said structure studies emphasize the understanding of electrode problems with high capacity electrodes. They also lead to new materials. Another commented that the study of degradation mechanisms is important to favor the use of Li batteries, and a third reviewer said the cycle and rate is important performance for PHEV application. A fourth commenter said that Shao-Horn is using surface chemistry techniques to help understand better processes that occur on electrode materials for lithium ion cells. The final commenter said the current Li-ion commercially used chemistry is not only cathode-limited in terms of cathode-to-anode ratio, it is truly cathode-limited in terms of performance. Fundamental study of the surface chemistry of the cathode materials is very important to the further improvements.

# QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL



#### BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

One reviewer said the approach is well-addressing key aspects, while another commented that Shao-Horn's approach is appropriate and useful. A third reviewer said the team is combining surface and bulk studies at the atomic level to try to understand the changes that take place in cathode materials and catalysts during operation. This seems to be a unique program in the BATT portfolio. Another reviewer said XPS is not so common to analyze Li-ion battery material. A final reviewer said the importance of the surface coatings was mostly studied in regard to the material safety performance. It has been also shown that doping materials has the same effect without the need to introduce an extra step in the manufacturing process. Limited data exist that show doping material with Mg and Ti improves both safety and performance (cycling at higher voltages and rate capability). It will be important to compare coating vs. doping. Also, could the formation of the metal fluorides be a "scavenging" of the acidic species? If a high quality, dry electrolyte is used, can the formation of the fluorides be observed?

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

The first reviewer said there was a good systematic approach and high quality work. Another commented that Shao-Horn's findings concerning annealing electrode materials are interesting and may lead to changes in the processes used to develop active materials. A third reviewer said good progress was made, but it should better demonstrated with some results on the improved life and stability. Another reviewer suggested that if the PI can provide the data other than XPS to support the results, it would be better. The final reviewer commented that the group has discovered that surface fluoride compounds are critical to good cycling behavior. To some degree, this can occur on bare surfaces if the salt is  $\text{LiPF}_6$  (perhaps with a small amount of HF), but it can also be effected by surface coatings. In addition to enhancing cycle life, the charging efficiency is also improved. Test case materials include LCO, LNMO and

LMNO with excess Li. This reviewer added that additional work on catalysts for bifunctional oxygen electrodes has yielded outstanding efficiency in a very difficult area.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

One reviewer said the collaboration with Argonne has been very productive, while another agreed Shao-Horn's collaboration with Argonne personnel appears to be useful. A third reviewer said the collaboration is clear. The fourth reviewer noted only one collaboration of value, but the applications of the method to other materials are recommended. The final reviewer suggests the team look into the industry collaborations.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

One reviewer said there was clear understanding of challenges and proper tool selection, while another said the future work is defined well. A third reviewer commented that the plan to continue work on LMNO is a good one. This reviewer hopes that the work with ANL will continue, as the Argonne material needs better definition. A fourth reviewer said the plan is acceptable even if more materials would be preferable. The final reviewer said Shao-Horn's proposed future work is reasonable. However, it is not clear that the annealing and quenching work will be useful without guidance from a cell manufacturer.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

The first reviewer said Shao-Horn's funding is sufficient, and another agreed that the resources are sufficient. The final reviewer commented that the progress is very good.

# Interfacial Processes - Diagnostics: Robert Kostecki (Lawrence Berkeley National Laboratory)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 6 reviewers.

## QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The first reviewer said Kostecki is trying to develop methods for better understanding of the active material interface processes that occur in a lithium ion cell. Another reviewer said studies of important reaction mechanisms use spectroscopic techniques that are well developed by the group. A third commenter said that the study of mechanisms and diagnostics of Li batteries are functional to BATT subprogram to investigate new materials for DOE objectives. Another reviewer said this project provides valuable insight into fundamental understanding of several potential future higher energy or higher voltage Li-ion electrode active materials and identifies significant fundamental issues with these materials which must be addressed for further progress. The final reviewer commented that this kind of basic research is good for national labs.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

Comments were generally positive in this section. The first reviewer said there was a well-focused approach to assist materials and cell development, while another noted the excellent breadth of techniques employed to investigate and characterize interfacial phenomena. A third reviewer said the approach was outstanding from point of applying various and most sophisticated analytical techniques. Another commenter said the approach is very clear, while a fifth reviewer said Kostecki's approach is adequate, and added that he may want to discuss his data analysis technique for this DS data. The final reviewer said in-situ methods have been developed to study the diffusion of lithium in Al and Sn for alloy reactions. The surface characteristics of LiMnPO<sub>4</sub>, an important high energy cathode for lithium ion batteries, were studied by spectroscopic methods.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

One reviewer said excellent progress with clear indications on future improvements. Another reviewer said that observations and definitive conclusions regarding LiMnPO<sub>4</sub> stability in typical Li-ion electrolyte systems are of great value. A third commenter said that the SEI on Sn electrodes was found to be poorly formed for most electrolytes. The use of additives to improve this film was indicated. The oxidation of LiMnPO<sub>4</sub> on charge results in unstable surface species which cause degradation of the surface. This may be an important mechanism of degradation of these electrodes.

A fourth reviewer said the result is fine, and added that we want to have some improvement plan based on the phenomena that the PI observed. Another commented that Kostecki has presented interesting technical accomplishments. However, this reviewer added that it is not clear why he studied Li+ transport through Al. The final reviewer said the work was good for understanding of failure mechanisms. Recommendations for improvement, however, sound too generic and did not help. For instance, suggesting better additives or electrolyte to form stronger SEI without suggesting their chemical structure or nature did not help at all.



#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Comments in this section were mixed. The first reviewer said there were clear and well justified collaborations, while another said all collaborators are very strong. A third reviewer commented that Kostecki should work more closely with others around the country who are using similar techniques. The final reviewer can see a little collaboration with outside of team.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first reviewer said future work is defined well, while another said future work was well related to the ongoing activities and, eventually, even better related to BATT progress. A third reviewer commented that they will study surface properties of other intermetallic electrodes and also other high voltage cathode materials. Another reviewer said Kostecki has reasonable plans for next year except for the DS experiments for electrode materials. The proposed work will probably not yield the desired diffusion coefficients for diffusion of Li+ in the active materials. The final commenter said the plan is excellent for understanding of failure mechanisms, diagnostics and analytical techniques development. However, this reviewer added, without better materials developed and their structures proposed, it is not clear how the project plan addressed and can meet the objective. Evaluate and improve the capacity and cycle life of intermetallic anodes and high voltage cathodes.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

The first reviewer said Kostecki's funding is sufficient, and another agreed the resources are sufficient.

# Model-Experimental Studies on Next-generation Li-ion Materials: Venkat Srinivasan (Lawrence Berkeley National Laboratory)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 6 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Comments were generally positive in this section. One reviewer said modeling activities are complementary to the research and development of new materials able to support DOE objectives. Another commented this was excellent, practically based work, both from an understanding point as well as a materials design/guidance standpoint. A third reviewer thinks using the modeling for failure mode analysis is very useful. Another commenter stated that the modeling and experimental studies are carried out on next generation lithium ion electrode materials. The final reviewer said Srinivasan is using modeling to analyze data to understand better the phenomena that occur in the electrodes in lithium ion cells.

# QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

Comments were generally positive in this section. The first reviewer

said the approach is well organized and addressing key barriers, while another commented that Srinivasan's approach is very useful since he is using detailed modeling to understand his experimental data. A third commenter thinks this method is a typical modeling study and very effective. The final reviewer said the approach on cathode materials is to try to understand various limiting properties on the reaction kinetics, e.g. particle size, electrode thickness, etc. This reviewer added that the approach on anode materials is to try to understand the effect of voltage hysteresis and other properties such as mechanical stress on Si cyclability.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

One reviewer commented that Srinivasan's results associated with this silicon anode work are very helpful, while another said excellent progress was made in the study of the mechanisms by means of models. Stress test equipment is also interesting for general work on Li materials. A third reviewer said ultra fast LiFePO<sub>4</sub> electrodes are better understood as a result of modeling. Si anodes are limited due to stress factors and are aggravated at high rates of charge/discharge. This reviewer added that binder failure is an important phenomenon that should lead to careful binder selection. The final reviewer said the scope is wide but LFP results and binder-related failure analysis is very interesting. This reviewer wants to see the other materials results.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

The first reviewer noted the clear and well-coordinated network in BATT, while another said Srinivasan is working with others in a meaningful way. The final commenter could see a lot of collaboration.



The first reviewer said the plan follows the achieved results, while another commented the future work is defined well. Another reviewer said Srinivasan's proposed work is attractive, and added that understanding the stress effects in the proposed silicon anodes will be a useful result from his future work. Another suggested examining high rate cycling effects, examining a full cell Si/NMC cell and comparing to graphite negative cell, and extending stress analysis to Si electrodes to look for failure modes.

**QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?** One commenter said Srinivasan's funding is appropriate for this project, and another said the resources are sufficient.

# Investigations of Cathode Architecture using Graphite Fibers: Nancy Dudney (Oak Ridge National Laboratory)

### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

One reviewer said novel materials can improve performances and reduce cost to accelerate the introduction of Li batteries. Another commented that Dudney is trying to develop a novel approach to preparing cathodes for lithium-ion cells. The final reviewer said carbon fiber can be used for basic material research.

# QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The first reviewer said the focus on the improvements in electrode non active materials is well motivated. The second reviewer said using a conductive carbon skeleton without blocking binder is attractive approach, but added that introduction of active material into skeleton is not an easy task and is difficult to scale up. Another commenter said Dudney's approach is appropriate for this project. However, this reviewer added the energy density appears to remain a problem. The final reviewer said this approach may be good to



establish the basic material research method, but this reviewer doesn't see any benefit to using this much carbon fiber in the electrode. Practically we cannot use this much conductor in the electrode.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

One reviewer said significant progress has been made on novel carbon fiber support, while another commented that Dudney's novel cathode with HQ material is encouraging, but the low volumetric energy density is a concern. A third reviewer would recommend that the PI compares the power density results including conductor weight and volume. The final reviewer said full utilization of active material was achieved. However, it appears that the slurry technique applied for introducing active material into skeleton is not effective and has limitations in terms of volumetric energy density. Images of fibers filled with LiFePO<sub>4</sub> suggest also that due to high void volume the electrodes will be overweight with electrolyte leading to low battery specific energy.

### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

The first reviewer said good collaborations in place, while another said collaborators are strong. Materials and test provided are adequate. A third reviewer commented that the collaboration is clear, while another added that Dudney is collaborating with HQ and others as needed. The final reviewer said collaboration with a viable industrial partner would be beneficial and should be aggressively pursued.

One reviewer said there was an acceptable plan for future work. A second reviewer said Dudney's plans for finishing this project are appropriate. Another commenter stated there was a good plan to finalize project. To show the advantages of method explored, it may require a very different technique to fill carbon skeleton porosity with active material. The final reviewer said that just establishing the test method is fine but this solution doesn't seem practical.

**QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?** One reviewer said Dudney's funding is sufficient, and another agreed the resources are sufficient.

# Block Copolymer Separators for Lithium Batteries: Nitash Balsara (Lawrence Berkeley National Laboratory)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

## QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

One reviewer said new separators are also necessary to improve Li applications, while another commented that Balsara is trying to develop dry separators (block copolymers) for lithium ion cells. A third reviewer said the solid electrolyte is the next generation cells. The final reviewer said there is no evidence that success of this program will alleviate other basic limitations of Li-metal anode-based systems for long cycle-life automotive applications.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The first commenter highlighted a clear focus on key specific barriers of separators. Another said this technology has a lot of barrier to be overcome: focusing on just material property is fine at this stage. A third reviewer said creating both active solid electrolyte separators and passive porous separators by block copolymer selfassembly sounds attractive. More details on nature of polymers will help for better understanding.



Another reviewer said Balsara's approach seems limited in scope. This reviewer added that it may be possible to make better time dependent measurements that would provide more useful information about lithium ion diffusion through block copolymers. It is probable that the materials he is studying will not be used in cells due to the very low diffusion coefficients he is report for Li+ ions. The final reviewer said insufficient technical content was provided to make a judgment.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

One reviewer said very good results, though another stated the gap between the target and the results needs to be clear. A third reviewer said the data presented look random and eclectic. It is difficult to conclude how they relate to battery performance. A fourth reviewer said insufficient technical content was provided to make a useful judgment. The final reviewer said Balsara's accomplishments seem to be a little thin. It is clear from his results that it is unlikely that block copolymers will provide the rate needed for lithium ion cells in vehicles.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Comments were mixed in this section. One reviewer noted the strong collaborators with well-defined roles, while another said the collaboration is clear. Another reviewer observed acceptable collaborations. A fourth commenter said the apparent lack of involvement with viable industrial development partner at this stage is unfortunate. The final reviewer said Balsara's collaboration is weak relative to using his materials in cells for use in vehicles.

Comments were mixed in this section. One reviewer said the future work is defined well, while another stated the prosecution of the work is essentially related to the completion of characterization activities. Another said the plan for further evaluation in full cells is appropriate but is far overdue. A fourth commenter said Balsara's plans are reasonable, but added that it is unlikely that these materials will prove to be useful.

The final reviewer noted the PI's statement, "Complete measurement of diffusion coefficient and transference numbers of dry block copolymer electrolytes. Evaluate same in full cells." This reviewer said this statement needs clarity on what kind of cells will be tested. What are the targeted conductivities and at what conditions?

### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

The first reviewer said the resources are sufficient, while the other commenter said Balsara's funding is too large relative to the success of his program. He should be encouraged to consider some other separator material.

# Interfacial Behavior of Electrolytes: John Kerr (Lawrence Berkeley National Laboratory)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

## QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

Comments were generally positive in this section. The first reviewer said these activities are complementary to the research and development of new materials able to support DOE objectives. Another reviewer commented Kerr is trying to use polymers to improve the performance of lithium ion cells, while the final reviewer said the polymer is one of the key technologies for the next generation.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

Comments in this section were mixed. The first reviewer said this is a clear and acceptable approach. Another said it is a good idea that polymers are used as binder also to reduce the polarization. Usually the interface between particle and polymer is the issue. A third reviewer stated the approach is good and includes a development of wide spectra of single Li ion conducting polymer electrolytes serving



separately a variety of negative and positive electrodes. However, this reviewer added, adjustment and modification of polymers for planned electrochemical systems and particular electrodes may require resources and time far exceeding the existing project. The final reviewer said Kerr's approach seems to be of limited utility.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Comments in this section were mixed. The first reviewer said there were good results, and another said the characterization of material was done well. Probably PI can get more help for electrochemical study. The engineering work is sometimes important for a polymer study. A third reviewer said progress was slow because of too wide a range of materials and systems taken for investigation, and added that maybe it makes sense concentrating on something delivering more definitive knowledge and performance improvement. The final reviewer commented that Kerr's technical accomplishments are not stated clearly.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Comments in this section were mixed. The first reviewer said there was a well-organized network of collaborations, while another said the collaboration is done well. A third reviewer said all collaborators are strong. However, their number looks too high to be well coordinated and focusing on objectives. The final reviewer said Kerr is collaborating with others, but it is not clear that he is interacting with the right people who could help this reviewer direct his efforts.

Comments in this section were mixed. The first reviewer said future work related to the achieved results, while another said the future plan is defined well. A third reviewer noted too many goals for future work (3 slides) that are not necessarily focusing on objectives, while another commented that focus on a few areas of investigation is needed with specific limited number of targets. The final commenter stated that Kerr's plans to work on polyelectrolytes will probably not yield useful results.

## QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

One reviewer said the resources are sufficient. A second reviewer said, taking into account the group presented and number of collaborators, the resources are sufficient but can be regrouped to meet the objectives on time. A final reviewer commented that Kerr's funding is excessive relative to his useful results.

# Advanced Binder for Electrode Materials: Gao Liu (Lawrence Berkeley National Laboratory)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 6 reviewers.

## QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The first reviewer highlighted new materials for improving Li cells, while another said Liu is trying to find a new binder material for lithium ion cells. A third reviewer said developing binders specifically for the use with Si-based anodes is necessary for the success of commercialization of the Si technology. The final reviewer said that new material development is important for the next generation.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The first reviewer said Liu's approach is reasonable, while another commented that the conductive polymer for Si is an interesting idea. A third reviewer said the project is clearly focused on the effect of binder. This should remain the main activity of the project. Another commenter said the conductive binder approach for large volume changing materials looks innovative. Certain binder mechanical



properties and chemical stability should be considered as well. The final reviewer said no information for the binder itself is provided, making it difficult to understand the approach in this regard.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

The first reviewer noted the excellent progress with complete experimental results, while another said Liu's results with the FMC material is interesting and may lead to useful material. Another commenter said the retained capacity looks impressive. Irreversible capacity loss and approaches to fix it did not promise a fast solution. This reviewer asked, is loss a function of binder amount? A fourth reviewer commented that steady progress towards the goals is demonstrated. This reviewer is interested in an explanation for the higher-than-theoretical capacity data. The final reviewer said the conductive polymer should work to improve the life. This reviewer wanted to see the comparison between regular polymer and conductive polymer, and also wanted to see the effectiveness of conductive polymer.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Comments were generally positive in this section. The first reviewer said structured collaborations were shown, while another commented the collaboration is clear. A third reviewer said strong collaborators with well-defined roles were used, while another added that the collaboration with viable external industrial partners is excellent. Another reviewer said Liu's collaboration with others is acceptable. This final reviewer commented excellent choice of partners to include industry and academia.

The first reviewer said the future work is defined well, while another said Liu's proposed plans are reasonable. A third reviewer commented on the good plan with significantly extended life cycling. Another reviewer said that the proposed plan seems like it can overcome remaining performance problems. The final reviewer would like to see a more focused approach to developing binders, and added that this is very necessary research.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

The first reviewer said Liu's funding is sufficient, and another agreed that the resources are sufficient. The final commenter noted steady progress, and good quality work.

# Atomistic Modeling of Electrode Materials: Kristin Persson (Lawrence Berkeley National Laboratory)

### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The first reviewer said modeling activities are complementary to the research and development of new materials able to support DOE objectives. Another said that Persson is trying to use atomistic modeling to understand the processes that occur in lithium ion cells. This final reviewer said this kind of basic research is good for national lab work.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

One reviewer said Persson's approach is appropriate for this project, while another said that the approach to use atomistic modeling is clear, together with some related technical barriers of Li batteries. The third reviewer said the proposed atomistic modeling with calculated phase diagrams appears to be quite effective for batteries materials development. The final reviewer said people add the third element, mixed oxide or surface coated material, and those are



already commercially available. This reviewer asks if the PI can work on these materials for modeling.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

One reviewer said real progress has been made with possibility to improve materials and conductivity properties. Another observed the good accomplishments as a first step demonstrating that approach works. Particularly, Li graphite diffusion data are impressive. This reviewer added that the database development for accelerated materials design looks promising. Another reviewer said Persson's results are interesting, but it is not clear that she is providing useful information. This final reviewer asked if these modeling results can be fed back to the actual material synthesis.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

Comments in this section were mixed. One reviewer said good collaborations with well defined collaborators' roles. Another reviewer said external (outside of VT) collaboration would be useful, while another reviewer can see a little collaboration. This final commenter said Persson should spend more time with other atomistic modelers such as Balbuenia at Texas A&M University.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first reviewer felt there was an acceptable plan, while another said the future work is defined well. A third reviewer said future plans are good, but total effort should be more focused with reduced number of investigation areas. A fourth commenter said surface-effects studies are important for nano-sized materials, and added that typically all such materials are covered with SEI in the real

battery system. Any plans to include SEI surface effects? The final reviewer said Persson's plans for next year are reasonable except for the Cu work, which should be reconsidered since it is unlikely to produce useful materials.

**QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?** The first reviewer said Persson's funding is sufficient for this project, while the other commenter agreed that the resources are sufficient. Coupled Kinetic, Thermal, and Mechanical Modeling of FIB Micro-machined Electrodes: Claus Daniel (Oak Ridge National Laboratory)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The first reviewer stated Daniel is trying to understand the effect of stress in lithium ion cells. Another said that theoretical activities complemented by experimental work are complementary to the research and development of new materials able to support DOE objectives. A third commenter said the project explores a novel approach to examining stress/strain behavior in Li-ion battery electrode materials, which may provide the potential for related optimization of usage conditions and increased cycle life in automotive battery systems. The final reviewer said this kind of basic research is good for national labs.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

One reviewer said the work is clearly focused, while another said this seems to be a good method for fundamental understanding. A

third reviewer said Daniel's approach is interesting and may lead to useful information. However, he should carry out more traditional experiments for comparison to his results. The final reviewer said the approach can deliver the most fundamental and direct understanding of stress-strain and state of charge relationships. It can be very costly expending such experiments on various cathode and anode materials and creating data base. This reviewer also asked about the influence of SEI generated on the sample surface. Can SEI affect mechanical response? SEI is a function of electrolyte nature, and it can add more complexity to data interpretation.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

The first reviewer felt there were good results with the possibility to be generally applied. Another reviewer commented that the project is at early stage, and added "nice images." A third reviewer said Daniel's accomplishments to date appear to be limited to establishing small-scale electrode materials and an apparent qualitative comparison to Sastry's simulation. A final reviewer said there are not too many results yet because this is a long-term project.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

One reviewer said Daniel's collaboration with others has been successful. A second reviewer noted only one collaboration, while another added that maybe one partner is sufficient for such an academic experimentation. The final reviewer said there was limited collaboration that could be extended to other BATT participants, contributing with materials and data for the full model.



The first reviewer said Daniel's plans for the future are appropriate, while another said the future work is defined well. A third reviewer said the plan addresses the objectives. The final reviewer said future work is well related to results achieved. More involvement of other BATT participants is recommended to enlarge materials analysis.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

The first reviewer said Daniel's funding is sufficient for this project, and another agreed that the resources are sufficient. The final reviewer is not an expert in the equipment used. However, data demonstrated and normalized to budget suggest that it is a very expensive endeavor.

# Long-Living Polymer Electrolytes: Christopher Janke (Oak Ridge National Laboratory)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

## QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

One reviewer said work on a polymer electrolyte may increase application possibilities for Li batteries, while another said the polymer electrolyte is key technology for next generation. A third reviewer said Janke's work appears to be directed toward preparing a better separator for lithium metal cells. The final reviewer said there is no evidence that success of this program will alleviate other basic limitations of Li-metal anode-based systems for long cycle-life automotive applications.

# QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED,

#### FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

The first reviewer said this is advanced material, so the PI is focusing on the material property and general polymer issue such as Li dendrite and interface. A second commenter said Janke's approach seems to be limited in scope, while another said the interesting approach not clearly explained. A fourth reviewer said no



description of the actual technical approach is provided, as any details are evidently proprietary. The final reviewer said the approach sounds innovative with possible big practical value. Is this hardening process applicable for mass production?

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

The first reviewer said there were good results for a just-started project, while another said this project was just started, and not much progress is seen yet. A third reviewer stated Janke's accomplishments are not significant to date. Another commented said not too much data shown so far – only hardness data and no connection with conductivity or anode performance. The final reviewer said no evidence was provided to indicate that the hardened PEO films are viable (no ionic conductivity, interfacial resistance, or other electrochemical data provided), and no evidence was provided to indicate that the hardened to indicate that the hardening process is viable.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

The comments in this section were generally in agreement, with reviewers independently commenting that no collaboration envisaged, no collaborators shown, and no evidence of collaboration provided. A fourth reviewer said Janke is apparently not working with others, and the final reviewer commented that this project was just started and the collaboration is not seen yet.

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first reviewer said the future plan is defined well, while another felt that Janke's plans are not explained clearly. The third reviewer said the plan is fine, with recommendation to analyze the effect of ceramic additives in the PEO electrolyte. Furthermore, life

cycling test should be useful to confirm the effect of modified PEO on the performances of complete cells. The final reviewer said there are only iterations on formulations, treatment, etc. This reviewer asked, is the project objective to stop dendrites?

**QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?** One reviewer said Janke's funding is excessive relative to his accomplishments, while the other commenter cannot judge it at this moment.

# In-Situ Electron Microscopy of Electrical Energy Storage Materials: Karren More (Oak Ridge National Laboratory)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

## QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

The first reviewer stated that in-situ technology is very useful, while another commented this work is functional to new material and Li system development. A third reviewer said More is trying to understand the SEI formation using small scale electrodes with insitu SEM. The final reviewer said advances in understanding of SEI formation processes and other processes at the electrode surface may aid automotive Li-ion battery development.

## QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

One commenter stated that the equipment will help in overcoming technical barriers. A second reviewer said More's approach is appropriate and may be extremely useful. Another thinks the approach is that the PI wants to establish a unique in-situ method for the basic research. A fourth commenter said in-situ electron microscopy characterization of electrodes and SEI can deliver



valuable information, and added that it is a good approach for academic ongoing activity. The final reviewer said that, although there may be significant challenges in sample preparation, the ability to examine negative electrode carbon materials other than HOPG will be critical.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

Comments were mixed in this section. One reviewer said More's accomplishments to date are exceptional, especially the observation of an apparent thin SEI layer on carbon. A second reviewer said the project is at an early stage and it needs more time to demonstrate the feasibility of the process, while another reviewer added that this project just started: the reviewer doesn't see the progress yet. The final reviewer commented that progress is still low and the collaboration process must be accelerated to extend the use of the proposed techniques.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

The first reviewer said collaboration is part of the project activities. A second reviewer commented that the project is just starting, but future evidence of collaboration outside of ORNL will be desirable. Another said this project has just started, but the collaboration is planned. A fourth reviewer commented that most of the work was done by ORNL. This reviewer added that they are expecting to demonstrate better collaboration in future. The final reviewer said More should be working with atomistic scale modelers to help design her experiments and analysis of her data.

One reviewer said the future work is defined well, while another said the plan addresses objectives of this ongoing academic research. A third reviewer said More should add work with theoreticians. The final reviewer said the plan is described enough, and not enough clear actions are taken to address the possible collaborations to extend the use of the microbattery.

**QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?** One reviewer said More's funding is adequate for this project, while the other commenter cannot judge it at this moment. **ENERGY** Energy Efficiency & Renewable Energy

Diagnostic Testing and Analysis Toward Understanding Aging Mechanisms and Related Path Dependence: Kevin Gering (Idaho National Laboratory)

#### **REVIEWER SAMPLE SIZE**

This project had a total of 5 reviewers.

### QUESTION 1: DOES THIS PROJECT SUPPORT THE OVERALL DOE OBJECTIVE OF PETROLEUM DISPLACEMENT? WHY OR WHY NOT?

One reviewer said the work was very relevant, while another said this was a great attempt to develop a realistic method for estimating calendar life. The final reviewer said Li-ion batteries are yet to meet the life requirements of PHEVs and their degradation is strongly related to their usage. A quantitative understanding of this connection between degradation and use is still desirable. The objective of this study is to develop a platform of diagnostic testing (DT) that is geared toward specific issues in vehicular applications, and employ it to examine mechanistic contributions to cell aging. In addition, advanced modeling tools to complement DT will be developed and also an optimized operational protocol will be identified to minimize the generalized aging process (chemistryspecific). More specifically, the objective is to bridge the gap between ideal laboratory test conditions and PHEV field conditions. Improvement in the cycle life will enable widespread use of Li-ion batteries for PHEVs, which will reduce the petroleum consumption, and pave the way towards petroleum replacement.



# QUESTION 2: WHAT IS YOUR ASSESSMENT OF THE APPROACH TO PERFORMING THE WORK? TO WHAT DEGREE ARE TECHNICAL BARRIERS ADDRESSED? IS THE PROJECT WELL-DESIGNED, FEASIBLE, AND INTEGRATED WITH OTHER EFFORTS?

Comments were generally positive in this section. The first reviewer said using commercially available systems to carry out the studies is an excellent idea. A second reviewer said it is very important to have work done on developing testing conditions to simulate real field conditions. We have too much gap today between laboratory tests and real life. These gaps may lead to over- or under-engineering the battery.

A third reviewer said the approach is to bridge the gap between the laboratory results and field observations by isolating the predominant aging factors, such as the nature and frequency of duty cycles, and the frequency and severity of thermal cycles. Such factors will be studied through DT in controlled and repeatable laboratory conditions to facilitate mechanistic evaluation of aging processes and path dependence thereof. Also, additional modeling tools will be developed and employed to promote diagnostic analysis over multiple domains, i.e., aging mechanisms as well as key performance issues. These approaches adequately address the technical barriers outlined here.

The final reviewer "loves the use of Sanyo cells," as this takes out the high level of variability associated with lab cells that would likely torpedo this effort (too much "noise" from cell to cell variation in life). The methodology seems very robust and addresses some really important variables and especially how they interact in a relatively small set of batteries. This reviewer's only concern is that this work needs to continue until a good portion of the cells actually reach end of life to capture any changes in degradation mechanisms as the cells age. However, the reviewer realizes that test position issues may preclude this. Work by Hawaii on understanding data and use of diff. capacity is also outstanding (this reviewer saw their paper in Vancouver ECS). The reviewer thinks

this work is really going to get a handle on a very thorny issue and provide an indispensable tool going forward when applied to HEV/PHEV batteries.

# QUESTION 3: CHARACTERIZE YOUR UNDERSTANDING OF THE TECHNICAL ACCOMPLISHMENTS AND PROGRESS TOWARD OVERALL PROJECT AND DOE GOALS.

One reviewer said this is a very timely topic and the progress made has been quite good, and recommended a focus on both. Another reviewer said it is too early to say yet, but results look very good. Programs looking at lifetime are inherently difficult to rate early in their progress; one cannot expect much at this stage and so such programs must be judged more on their approach than their 'progress'.

The final reviewer said moderate progress has been accomplished in terms of developing the Diagnostic Testing (DT) and combining with suitable models. Specific accomplishments include: i) Initiated path dependence studies with Sanyo Y 18650 cells (16) which are on-going and ii) Developed key computational methods and benchmarked on Gen2 and other Li-ion cell performance and aging data, e.g., capacity loss, cell conductance loss, performance over multiple domains, incremental capacity analysis, and equivalent circuit analysis. The early results obtained thus far are useful for assessing beginning-of-life trends and initial estimates of parameters for aging models. Finally, INL and HNEI have developed key computational tools used to model, diagnose, and predict performance and aging of electrochemical cell. The progress is consistent with the project goals.

#### QUESTION 4: WHAT IS YOUR ASSESSMENT OF THE LEVEL OF COLLABORATION AND COORDINATION WITH OTHER INSTITUTIONS?

The first reviewer said this showed excellent team work. Another commented that there are good collaborations within DOE, but also with external researchers. In particular, collaboration with the Hawaii Natural Energy Institute (HNEI) provides a synergistic basis due to the complementary histories of INL and HNEI in battery testing, research, and modeling. The final reviewer highlighted the good work with Hawaii. This reviewer still is not sure how this fits or doesn't fit with some of the modeling work on lifetime done at other DOE labs in the past. This reviewer asked, are they competing and/or complementary efforts?

# QUESTION 5: HAS THE PROJECT EFFECTIVELY PLANNED ITS FUTURE WORK IN A LOGICAL MANNER BY INCORPORATING APPROPRIATE DECISION POINTS, CONSIDERING BARRIERS TO THE REALIZATION OF THE PROPOSED TECHNOLOGY, AND, WHEN SENSIBLE, MITIGATING RISK BY PROVIDING ALTERNATE DEVELOPMENT PATHWAYS?

The first reviewer said the approach of pure calendar vs. aggressive and moderate cycle life is really good. The second reviewer said this was a great plan, although this work should be carried on for a lot longer than 18 months – want to get to the point where most cells "fail" or reach end of life. The third reviewer would recommend a sharp focus on developing models on failure/degradation mechanisms (how do you coordinate with Dennis Dees?). This reviewer asked, how about storing cells without any pulses?

The final reviewer commented that the future studies will include: i) monitoring the aging trends for the path dependence studies, ii) mechanistic analyses and modeling of mature data sets applying INL and HNEI modeling tools, iii) demonstration of INL diagnostic/predictive modeling capabilities through software integrating key modules regarding performance over life, iv) quantifying the impact of thermal cycling on Sanyo Y cell aging, v) extension to other duty-cycles (e.g., FUDS, DST), and other Li-ion cell chemistries. These studies will duly address the technical barriers outlined in the project.

#### QUESTION 6: HOW SUFFICIENT ARE THE RESOURCES FOR THE PROJECT TO ACHIEVE THE STATED MILESTONES IN A TIMELY FASHION?

The first reviewer said this is very important work that needs resources. Another commented that the budget of \$460 k per year (last year) looks reasonable and adequate for this effort. The final reviewer said again this effort is test position limited. This reviewer is also concerned that it is not going to run long enough in part because of this and the general "impatience" of a program with an annual review basis.