2. Battery R&D

The Vehicle Technologies Office (VTO) supports research, development, deployment, and demonstration (RDD&D) of new, efficient, and clean mobility options that are affordable for all Americans. The office's investments leverage the unique capabilities and world-class expertise of the national laboratory system to develop new innovations in vehicle technologies, including: advanced battery technologies; advanced materials for lighter-weight vehicle structures and better powertrains; energy-efficient mobility technologies and systems (including automated and connected vehicles as well as innovations in connected infrastructure for significant systems-level energy efficiency improvement); combustion engines to reduce greenhouse gas (GHG) emissions; and technology deployment and integration at the local and state level. In coordination with the other offices across the Office of Energy Efficiency and Renewable Energy (EERE) and the U.S. Department of Energy (DOE), the Vehicle Technologies Office advances technologies that assure affordable, reliable mobility solutions for people and goods across all economic and social groups; enable and support competitiveness for industry and the economy/workforce; and address local air quality and use of water, land, and domestic resources.

The VTO Battery R&D subprogram supports the decarbonization of transportation across all modes, serves to increase American advancement/manufacturing of battery technology, and creates good paying jobs with the free and fair chance to join a union and bargain collectively. The subprogram supports research with partners in academia, national laboratories, and industry covered under the Energy Storage Grand Challenge key priority and distinct crosscuts. The Energy Storage Grand Challenge encompasses R&D across energy storage including the discovery of alternative lithium battery materials, processing for raw materials, development of advanced battery cells, discovery of innovative cell manufacturing techniques, and battery recycling. The Critical Minerals crosscut aims to reduce or eliminate cobalt and nickel in lithium battery cathode materials, develop substitutes for graphite such as silicon composite anodes and lithium metal anodes, and develop advanced recycling and processing through scale up of bench-scale recycling processes and innovative separation processes seedlings. The Advanced Manufacturing crosscut is focused on coordination with the Advanced Manufacturing Office for joint projects scaling up solid state battery materials and lithium metal electrode processing technologies addressing critical materials for batteries.

The Battery R&D activity supports early-stage R&D of high-energy and high-power battery materials, cells, and battery development that can enable industry to significantly reduce the cost, weight, volume, and charge time of PEV batteries. This activity is organized into sub-activities: advanced battery materials research, advanced battery cell R&D, and battery recycling R&D. Advanced battery materials research is coordinated with the Critical Minerals Initiative and includes: early-stage research of new lithium-ion cathode, anode, and electrolyte materials (currently accounting for 50-70 percent of PEV battery cost) and the development of "beyond lithium-ion" technologies, such as lithium metal anodes, solid-state electrolytes, and sulfur-based cathodes, that have the potential to significantly reduce weight, volume, and cost by three times, with a target of $60/kWh. Advanced battery cell R&D includes: early-stage R&D of new battery cell technology that contains new materials and electrodes that can reduce the overall battery cost, weight, and volume while improving energy, life, safety, and fast charging; and high-fidelity battery performance, life, fast charging, and safety testing of innovative battery technologies including recycled material and cells. Battery recycling R&D includes the development of innovative battery materials recycling and reuse technologies, and the Lithium-Ion Battery Recycling Prize, both to assure sustainability and domestic supplies of key battery materials and minerals.

Project Feedback

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 1.0 to 4.0)*. 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.

Table 2-1 – Project Feedback

Presentation ID	Presentation Title	Principal Investigator (Organization)	Page Number	Approach	Technical Accomplishments	Collaborations	Future Research	Weighted Average
bat359	Status and Challenges of Electrode and Electrolyte Materials for High Energy Cells	Stanley Whittingham (Binghamton University)	2-7	3.67	3.50	3.67	3.50	3.56
bat360	Cathodes Beyond Lithium Nickel Manganese Cobalt Oxide (NMC) 811	Arumugam Manthiram (University of Texas at Austin)	2-12	3.83	3.75	3.25	3.33	3.66
bat361	Understanding and Improving Lithium Anode Stability	Yi Cui (Stanford University / SLAC National Accelerator Laboratory)	2-18	3.50	3.38	3.50	3.50	3.44
bat362	High Capacity S Cathode Materials	Prashant Kumta (University of Pittsburgh)	2-23	3.50	3.30	3.50	3.50	3.40
bat364	Synergistic Effects of Electrode and Electrolyte Materials for High Energy Lithium Cells	Jihui Yang (University of Washington)	2-29	3.20	3.30	3.60	3.30	3.31

bat365	Stabilizing Lithium Metal Anodes by Interfacial Layer and New Electrolytes	Zhenan Bao (Stanford University / SLAC National Accelerator Laboratory)	2-34	3.30	3.20	3.40	3.10	3.24
bat366	Manufacturing and Validation of Lithium Pouch Cells	Mei Cai (General Motors Company)	2-40	3.50	3.38	3.88	3.50	3.48
bat367	Multiscale Characterization Studies of Lithium Metal Batteries	Peter Khalifah (Brookhaven National Laboratory)	2-44	3.67	3.75	3.50	3.58	3.68
bat368	Full Cell Diagnostics and Validation to Achieving High Cycle Life	Eric Dufek (Idaho National Laboratory)	2-49	3.38	3.50	3.50	3.50	3.47
bat369	High Energy Rechargeable Lithium-Metal Cells, Design, Fabrication and Testing	Jie Xiao (Pacific Northwest National Laboratory)	2-52	3.88	4.00	3.75	3.63	3.89
bat496	Silicon Consortium Project: Advanced Characterization of Silicon Electrodes	Robert Kostecki (Lawrence Berkeley National Laboratory)	2-56	3.38	3.13	3.38	3.25	3.23
bat497	Silicon Consortium Project: Electrochemistry of Silicon Electrodes	Christopher Johnson (Argonne National Laboratory)	2-60	3.13	2.88	3.38	3.00	3.02
bat498	Silicon Consortium Project: Next-Generation Materials for Silicon Anodes	Nathan Neale (NREL)	2-64	3.63	3.50	3.63	3.63	3.56

bat499	Silicon Consortium Project: Mechanical Properties of Silicon Anodes	Katherine Harrison (Sandia National Laboratories)	2-68	3.25	3.25	3.50	3.50	3.31
bat500	Silicon Consortium Project: Science of Manufacturing for Silicon Anodes	Gabriel Veith (Oak Ridge National Laboratory)	2-72	3.70	3.50	3.70	3.50	3.58
bat501	Integrated Modeling and Machine Learning of Solid-Electrolyte Interface Reactions of the Silicon Anode	Kristin Persson (Lawrence Berkeley National Laboratory)	2-76	3.63	3.38	3.63	3.50	3.48
bat523	Development of Long Life Lithium and sulfurized polyacrylonitrile (SPAN) Cells	Ping Liu (University of California-San Diego)	2-79	3.80	3.50	3.60	3.40	3.58
bat524	Advanced Electrolytes for Lithium Metal Batteries	Chunsheng Wang (University of Maryland)	2-85	3.50	3.63	3.63	3.50	3.58
bat525	Fluorinated Solvent-Based Electrolytes for Low Temperature Lithium-ion Battery	John Zhang (Argonne National Laboratory)	2-89	3.25	3.25	3.25	3.25	3.25
bat526	Ethylene Carbonate-lean Electrolytes for Low-Temperature, Safe Lithium-ion Batteries	Bryan McCloskey (Lawrence Berkeley National Laboratory)	2-92	3.33	3.17	3.00	3.00	3.17
bat527	Synthesis, Screening and Characterization of Novel Low-Temperature Electrolyte for Lithium-ion Batteries	Xiao-Qing Yang (Brookhaven National Laboratory)	2-95	3.17	3.17	3.17	3.17	3.17

bat528	Structurally and Electrochemically Stabilized Silicon-rich Anodes for Electric Vehicle Applications	Murali Ramasubramanian (Enovix)	2-98	3.60	3.50	3.30	3.20	3.46
bat529	Rationally Designed Lithium-Ion Batteries Towards Displacing Internal Combustion Engines	Rick Costantino (Group 14 Technologies)	2-102	3.40	3.60	3.30	3.00	3.44
bat530	Ultra-Low Volume Change Silicon-Dominant Nanocomposite Anodes for Long Calendar Life and Cycle Lif	John Tannaci (Silanano)	2-107	3.00	3.00	2.90	2.60	2.94
bat531	Solid State Lithium-ion Batteries Using Silicon Composite Anodes	Pu Zhang (Solid Power Battery)	2-112	3.50	3.38	3.13	3.00	3.33
bat532	Electrolytes with Lithium-ion Batteries with Micro-sized Silicon Anodes	Chunsheng Wang (University of Maryland)	2-116	3.25	3.50	3.88	3.38	3.47
bat533	Fluorinated Local High Concentration Electrolytes Enabling High Energy Density Silicon Anodes	Amy Marschilok (Stony Brook University)	2-122	3.50	3.25	3.38	3.38	3.34
bat534	Devising mechanically compliant and chemically stable synthetic solid-electrolyte interphases on silicon	Pierre Yao (University of Delaware)	2-127	3.00	2.75	2.88	3.25	2.89

bat553	Understanding solid electrolyte interphase (SEI) reactions in Lithium metal and Lithium-Sulfur batteries	Perla Balbuena (Texas A&M University)	2-132	3.25	3.38	3.00	3.25	3.28
bat554	Fabricate and Test Solid-State Ceramic Electrolytes and Electrolyte/Cathode Laminates †	Mike Tucker (LBNL)	2-136	3.17	2.83	3.17	3.00	2.98
Overall Average				**3.45**	**3.38**	**3.42**	**3.31**	**3.39**

† Denotes poster presentation.

Presentation Number: bat359
Presentation Title: Status and Challenges of Electrode and Electrolyte Materials for High Energy Cells
Principal Investigator: Stanley Whittingham, Binghamton University

Presenter

Stanley Whittingham, Binghamton University

Reviewer Sample Size

A total of six reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 83% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 17% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

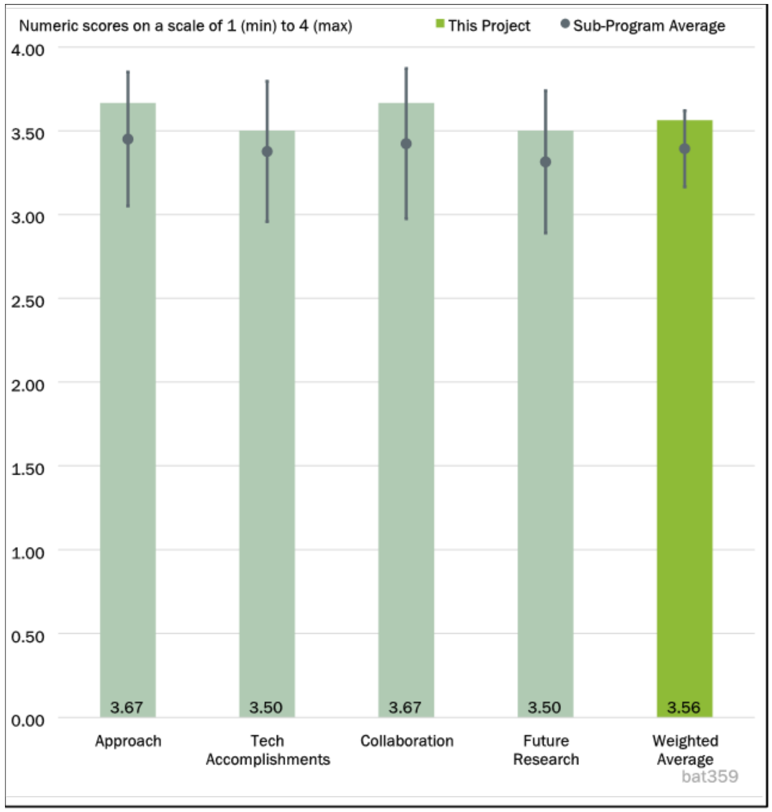

Figure 2-1 - Presentation Number: bat359 Presentation Title: Status and Challenges of Electrode and Electrolyte Materials for High Energy Cells Principal Investigator: Stanley Whittingham, Binghamton University

Question 1: *Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?*

Reviewer 1

The reviewer interprets the aim of the project as the following: Can we increase the energy density of Li/NMC cells by overcharging the cathode while still maintaining acceptable cycle life and safety? To answer this question, this project studies the thermal reactivity of an NMC811 cathode in the presence of different liquid electrolytes. The work presented is a great start towards answering this question.

Reviewer 2

The reviewer commented the principal investigator (PI) and team are working to address major technical barriers associated with nickel (Ni)-rich NMC both at the materials and cell component level. Both in situ calorimetry and ex-situ DSC provides critical on the exothermicity of electrolyte-electrode reaction at higher voltages and the effect of additives and coatings.

Reviewer 3

The reviewer said this project is targeted at developing high-energy cathode materials with a specific capacity of greater than 220 mAh/g for Li metal batteries. The team is aiming to improve the cycling and thermal stability of Ni-rich cathodes through a combination of surface coating and lithiation, which sounds promising.

Reviewer 4

The reviewer commented the team addressed the barrier of improving energy density by first looking into each individual component and their interfaces, starting from leveraging the state-of-the-art battery material candidates. Their thermal stability and reactivity are investigated by thermal analysis and operando studies.

Reviewer 5

The reviewer said that though the technical barriers are addressed by the author, the following questions should be considered. Does the niobium (Nb) coating work in LiFSi only, and what happens with $LiPF_6$?. The reviewer noted that it is good to show and compare the improvement due to Nb coating for LiFSi and $LiPF_6$.

Reviewer 6

The reviewer said the team has thoroughly addressed the technical barriers, and the approaches are reasonable. One question is for the thermal studies The reviewer noted it looks as though most of the experiments will be conducted with a coin cell. The heat mass could lead to much noise. The reviewer said the team might also consider the experiments with pouch cells, which could generate more reliable data. In addition, the team might also test the thermal behaviors at different stage of battery life.

*Question 2: **Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.***

Reviewer 1

The reviewer said the Nb coating is certainly interesting and it shows improvement for the retention capacity. It is also important to show what is the cycling capacity in terms of mA/hr for the improved retention capacity.

Reviewer 2

The reviewer said the thermal benchmarking of bare cathodes versus coated cathodes provides important information about the role of coatings, reactivity with cell components, and parasitic reaction that generated heat. Safety evaluation for high Ni-content NMC cathodes is critical. The reviewer noted that Nb-containing NMC show better performance, and the team has developed a method to incorporate Nb at the lithiation stage, thus avoiding an extra synthesis step.

The reviewer pointed out that more mechanistic studies are needed to understand the role of Nb in improving the performance of Ni-rich NMC. Is Nb mainly on the surface of the cathode in some oxide form or in the bulk cathode particle. Is the coating uniform or random morphology? The reviewer suggested that the chemical formula of electrolytes and additives should be elaborated and described in detail to analyze and interpret the DSC studies.

Reviewer 3

The reviewer said the team has enabled operation of a Ni-rich cathode with an ultrahigh Ni content of 90% for 200 cycles without obvious fading via Nb coating/substitution. The results look very impressive. It could potentially lead to a much-improved energy density and cycle life of Li metal batteries for Battery500.

Reviewer 4

The reviewer said the team investigated several pairs of electrolytes/cathodes for the thermal analysis, and the team found the Nb coating showed improved cycling stability of high Ni NMC. Will the team check the thermal stability of the Nb-coated high Ni NMC with electrolytes and see potential improvement? Will the team link the thermal analysis with cycling stability data in the future studies?

Reviewer 5

The reviewer said this project demonstrated the experimental capability to measure the thermal reactivity of a NMC811 cathode in a variety of different electrolytes, and provided a few points for consideration:

A commercially relevant cathode active material (CAM) + electrolyte baseline should be established. Many cell suppliers use a proprietary CAM coating and electrolyte formulation. If the goal is a systems approach, then the CAM/electrolyte system should include a CAM coating.

The reviewer said increasing voltage cutoff will exacerbate transition metal dissolution and increase crosstalk with the Li-metal anode, which may reduce cycle life. Are there plans to consider this issue in addition to CAM/electrolyte reactivity? The reviewer understands that some of the electrolyte formulations are proprietary. For the formulations that are known, what fundamental mechanisms are thought to influence the results? M47 and ED2 cause significantly higher heat generation, for example. Why is that?

Reviewer 6

The reviewer said it looks the baseline selection needs to be improved. For example, the team has shown the cycle stability of NMC9055 can be improved by doping Nb; however, the baseline performance is worse than the commercial product.

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer said the PI and team are fully integrated and demonstrated great teamwork.

Reviewer 2

The reviewer remarked the team has done an excellent job to collaborate with other institutions, and it is well organized.

Reviewer 3

The reviewer commented the team collaborates well with the Battery500 consortium team.

Reviewer 4

The reviewer said the team has very good collaborations among different institutes, which is reflected from highly productive publications.

Reviewer 5

The reviewer pointed out that this project utilizes advanced, sometimes proprietary, liquid electrolytes from a variety of sources so the collaborative nature of this project is on point. The reviewer said CAM from other DOE projects should be evaluated in addition to the commercial Targray baseline. For example, the NATM from BAT360 should be evaluated.

Reviewer 6

The reviewer said collaboration within the project team is good, but that no industry contributions are specified. This project seems to collaborate with national labs and other external entities, but it is yet to be done. The reviewer cited as examples LATP coating (Yang) on separator in LiFSi can be combined with Nb coating and tested, and the addition of $LiPO_2F_2$ (Peter) works in $LiPF_6$. Can it work with LiFSi? The reviewer said if Nb coating works for $LiPF_6$ then the addition of $LiPO_2F_2$ (Peter) can also be tested.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer remarked proposed future research can achieve its target to some extent. It might be a challenge to reach 500 WHr/Kg and it depends on the approach for full cell considering the improvements with other team members.

Reviewer 2

The reviewer said the team has raised the most critical issues. It would be great if the tests can be done in the pouch cell to for better validation, besides the coin cells.

Reviewer 3

The reviewer remarked the future work of the team will support Battery500 Keystone 2 and 3, develop cathode reactivity strategic plan and provide better cathode materials for the Battery500 projects.

Reviewer 4

The reviewer said the proposed research has covered most of the barriers of Ni-rich cathodes at the particle, material, and electrode level. The reviewer noted that air stability of the Ni-rich cathode, particularly when the Ni content is more than 90%, should be taken care of.

Reviewer 5

The reviewer remarked the proposed future work makes sense; however, it is not clear how the data presented here will be applied. The reviewer asked how will the data help set cutoff voltage, and will the data be used in continuum cell modeling efforts?

Reviewer 6

The reviewer found that the proposed future work is a little broad—needs more specificity to exact technical challenges that need to be addressed. Right now, it is at a high level.

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

The reviewer said yes, it supports overall VTO program objectives.

Reviewer 2

The reviewer remarked overall, the project advanced VTO goals for enabling advanced Li-metal batteries both in terms of safety and performance.

Reviewer 3

He reviewer said yes, it supports the overall VTO objectives.

Reviewer 4

The reviewer remarked the project definitely supports the VTO objectives in developing 500 Wh/kg batteries for automotive application.

Reviewer 5

The reviewer commented project objectives align well with the VTO objectives in improving Li-ion battery performance with no sacrifice of scalability.

Reviewer 6

The reviewer pointed out that understanding the thermal reactivity of Li-metal batteries is of critical importance.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer commented the team led by Prof. Whittingham is the pioneer in cathode development.

Reviewer 2

The reviewer said proposed project milestones are achieved in a timely fashion.

Reviewer 3

The reviewer said resources are adequate.

Reviewer 4

The reviewer remarked yes, the collaboration among national labs, universities, and electric vehicle (EV) manufactures is very critical to cover the fundamental understanding and practical implementation.

Reviewer 5

The reviewer does not see any resource problems the team may encounter in pursuing the proposed work.

Reviewer 6

The reviewer said project resources are sufficient.

Presentation Number: bat360
Presentation Title: Cathodes Beyond Lithium Nickel Manganese Cobalt Oxide (NMC) 811
Principal Investigator: Arumugam Manthiram, University of Texas at Austin

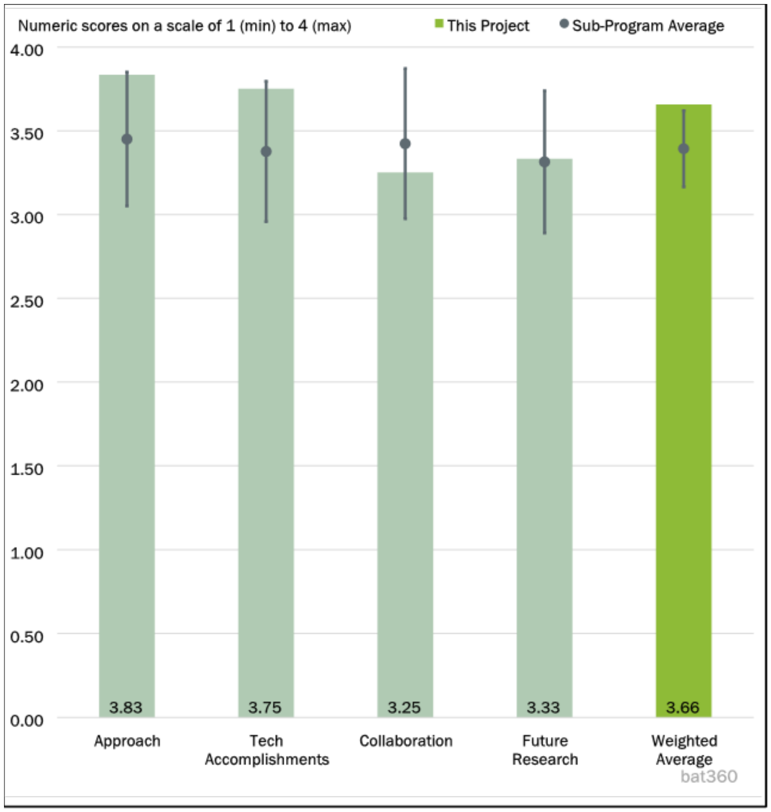

Figure 2-2 - Presentation Number: bat360 Presentation Title: Cathodes Beyond Lithium Nickel Manganese Cobalt Oxide (NMC) 811 Principal Investigator: Arumugam Manthiram, University of Texas at Austin

Presenter

Manthiram, University of Texas at Austin

Reviewer Sample Size

A total of six reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 100% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 0% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

Question 1: *Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?*

Reviewer 1

The reviewer commented as usual, Dr. Manthiram performs world-class research and generates very relevant and insightful findings. His titanium (Ti)- doped Ni rich cathode shows excellent performance and associated diagnostics are well performed and explained.

Reviewer 2

The reviewer said technical barriers are well addressed and designed, and the timeline reasonably planned. It clearly addresses less cobalt (Co) or no-Co cathodes with a good approach.

Reviewer 3

The reviewer commented there is significant technical progress made in this project that cannot be done without a successful approach and experimental plan that address the performance and safety barriers. The PI and team benchmarked the performance of Ti and aluminum (Al) doped Ni-rich cathodes (NATM) with respect to NMC and Ni-Co. The reviewer noted that NATM showed better performance in terms of capacity retention. The cycled cathodes were characterized using scanning electron microscope (SEM), time-of-flight secondary ion mass spectrometry (ToF-SIMs), DSC, and high-resolution transmission electron microscopy (HRTEM). The team tested different elelctrolytes with NMC and Ni-Co cathodes.

Reviewer 4

The reviewer said that the cathode is a critical bottleneck for improving energy density, and this project is very important and well-designed. The reviewer said the project includes approach fundamental understanding of the failure mechanism of high Ni cathodes, identifying the important factors which are responsible for the capacity fading, and controlling those factors to retain the capacity. The approach is rational and effective.

Reviewer 5

The reviewer detailed that this project seeks to understand the synthesis of high Ni cathode active material (CAM) without Co or Mn. They present a compelling NATM) formulation and seek to understand the effect that each dopant has on synthesis and performance. The reviewer noted that the Li metal work seems unrelated to the project Milestones, but it is great work nonetheless.

Reviewer 6

high-resolution transmission electron microscopy

The reviewer said the team addresses energy density barriers by advanced synthesis of high Ni, beyond NMC cathodes; incorporating a high salt concentration electrolyte; and a mechanism understanding from in whole cell characterizations. The approach from material synthesis, cell integration, to characterizations is well designed to achieve the goal of the project.

Question 2: *Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.*

Reviewer 1

The reviewer remarked the team reported new compositions such as NC, NM, NATM, and NA . It seems NA seems to be a good optimized candidate out of all in terms of capacity (220) and retention (82%). The reviewer provided the following comments the team should consider.

The author also showed the exothermic peak for all except NA. Is the exothermic peak higher or lower and how much is it suppressed? The author also mentioned localized high concentration electrolyte (LHCE) reduces carbon dioxide (CO_2) generation dramatically with minimal effect on oxygen generation for LiFSi and not for $LiPF_6$-based localized saturated electrolyte (LSE). Then, what will be the approach for Ni-rich electrodes (NA and others) when cells exhibit better cycle life in $LiPF_6$-based LSE than in LP57 and LHCE?

Reviewer 2

The reviewer provided the following remarks.

- NATM cathodes showed a high temperature threshold and minimal heat release compared to other Ni-rich compositions. Ti and Al bonded to surface O atoms provided better thermal stability.
- STEM and TOF-SIMs results showed minimal cracking and better interfacial stability for NATM cathodes compared to other Ni-rich counterparts. The reviewer noted better cathode-electrolyte interface (CEI) stability.
- Regarding the pure Ni-rich cathode; the reviewer said $LiNiO_2$ showed much better stability and cycle life in LSE compared to LHCE and the baseline LP57.
- Differential Electrochemical mass spectroscopy (DEMS) results showed much lower CO_2 evolution when using LHCE for Ni-Co cathodes. The reviewer noted a favorable electrode-electrolyte interaction, and would be interested in similar experiments for LATM cathodes.
- Dual salt and LHCE electrolytes showed closed pack Li-morphology during plating at a respectable current density of 1 mA/cm^2 and forms mainly amorphous Li rather than crystalline. The reviewer

asked if the team has tested greater than 1 mA/cm^2 current density. What are the coulombic efficiency (CE) values reported for 1 mA/cm^2 plating and stripping?

Reviewer 3

The reviewer said very good progress, no issues.

Reviewer 4

The team synthesized a new cathode material NATM, which showed excellent battery performance and was explained by improved crack mitigation, reduced surface reactivity, and better thermal stability. The team also studied the outgassing and lithium plating behaviors of different cathodes and electrolytes. It was mentioned that all the cathode materials tested were uncoated. It is intriguing to know whether applying coating can further improve the cell performance. Is this in the future project plan?

Reviewer 5

Regarding NATM, the reviewer congratulates the University of Texas at Austin team on the development of their NATM CAM. The reviewer agrees with the authors that it is crucial to understand capacity fade mechanisms, and encourage the authors to study the mechanism for secondary particle cracking more closely. Cracking occurs when tensile stresses are generated on the surface of a secondary particle due to a decrease in volume of the particle surface. This may occur when the surface of the secondary particle is de-lithiated before the core of the secondary particle. The reviewer suggested the team look to silicon (Si) anodes for an analogy. The battery community knows that NCM materials shrink 3%-8 vol% upon de-lithiation, which is far less than Si, but still substantial. The reviewer said most of this volume change occurs during the H2 to H3 phase transition. For NMC333, it has been shown that Ti substitution will minimize the overall volume change from 4% to 3% (K.C. Kam et al., J. of the Electrochem. Soc., (2012), 159 A1383). The broadening of the H2 to H3 phase transition for NATM hints that such a mechanical explanation is likely as previously reported for NCA (G. W. Nam et al., ACS Energy Letters, (2019) 4, 2995-3001). The reviewer noted that reduced surface reactivity may also be a factor as the authors suggest. If the volume of the surface rock-salt phase is less than that of layered NATM then surface reactivity may explain the author’s results as well. Elucidation of this question may yield a fundamental design principle to avoid secondary particle cracking.

Regarding outgassing, the reviewer said the authors demonstrated DEMS capability. The team should evaluate O_2 evolution of NATM using this method. Regarding Li-metal, the reviewer said the authors studied Li plating morphology and structure with different electrolytes. The reviewer said follow-up work should consider the effect of different CAMs to better understand crosstalk. How does TM dissolution, or lack thereof. affect Li-metal electrode cycling efficiency?

Reviewer 6

The reviewer said the team made significant progress during the past year. The team has developed a Co- and Mn-free high-Ni cathode and demonstrated good cycle and thermal stability. However, the loading of the cathode materials is relatively low (approximately 2mAh), which is far off from the targeted energy density of Battery500. The reviewer said when the loading is increased to more than 3.5 mAh, the cell performance could be different. The team should try higher loading, so it can provide direct performance data to the Battery500 team for better integrating with other teams’ efforts.

The reviewer said the team claims amorphous Li can lead to improved cycle efficiency, and dual salt electrolyte and LHCE will reduce the formation of crystalline Li. The team might need more evidence and explanation.

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer said the team was actively collaborating with other institutes, including PNNL for cell evaluation, Stanford for fluorinated electrolytes, University of Washington for coating, Brookhaven National Laboratory and the Advanced Photon Source (APS) for synchrotron X-ray scattering.

Reviewer 2

The reviewer said the authors demonstrated excellent collaboration and coordination with other organizations. The thermal reactivity of NATM should be evaluated by BAT359.

Reviewer 3

The reviewer noted there is always an issue with scaling up cathode material production so the CAM can be used by other members of the Battery500 team. This reviewer had not noticed many of Dr. Manthiram's materials being used in cell builds at Pacific Northwest National Laboratory (PNNL) although the reviewer may have missed that.

Reviewer 4

The reviewer said collaboration within the project team is good, but no industry contributions are specified. This project seems to collaborate with national labs and other external entities, but new collaborative experiments are yet to be done with the team.

Reviewer 5

The collaboration across the Battery500 team is excellent, but the reviewer was not clear about the specifics of this project as far as collaborations. The reviewer would like to know if LSE and LHCE electrolytes were provided by partners.

Reviewer 6

The reviewer noted the team has pretty good collaboration across the Battery500 teams, which are mainly universities and national labs. Can the team also have more collaboration with industries for further evaluation based on industry requirements for EV applications, such as through USABC program. This would significantly accelerate the commercialization of those interesting cathode materials.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer said the PI has a great path forward for the second year on all major technical barriers with desired performance metrics.

Reviewer 2

The reviewer said the team has clearly listed the future work to be conducted in the project. The accomplishment of the tasks will help achieve the overall project target in further improving energy density and cycling stability.

Reviewer 3

As mentioned, Dr. Manthiram's work is world class. The reviewer believed DOE's work on Ni-rich CAMs might be ready for de-emphasis. The reviewer explained that DOE has committed to pursuing earth abundant battery materials and this PI might be able to greatly aid in the development of sulfur (S), silicate, manganese (Mn)-rich layered, Mn phosphate, or other earth abundant CAMs.

Reviewer 4

The reviewer said the proposed project target 500 Whr/Kg may be possible with some of these cathodes. LATP coating (Yang) on separator in LiFSi, possible Nb coating (Dr. Stanley Whittingham) could be considered to improve the performance as a team work.

Reviewer 5

The reviewer said the author's proposed future work is reasonable, and encouraged DEMS and Li-metal morphology studies in conjunction with NATM.

Reviewer 6

The reviewer commented the future directions are rational. The team identified the most critical factors such as doping, but pushing everything to the limit could be high risk before solving the capacity fading for the current design. The reviewer also pointed out the safety of the new cathode materials needs to be addressed as well.

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

This project is highly relevant because improving the stability of CAMs is critical to achieving Battery500 goals.

Reviewer 2

The reviewer said yes, it supports overall VTO program objectives.

Reviewer 3

The reviewer remarked the project is critical and has close relevance with the VTO Battery program for high energy and low-cost battery technologies, as the new materials have more than 220 mAh/g capacity and are Co free.

Reviewer 4

The reviewer said the project supports the development of Ni-rich and low-Co cathodes and screens the relevant electrolyte systems for performance and safety. Overall, this project support development of Li-metal cells with high energy density and deep cycles.

Reviewer 5

The reviewer remarked the project is relevant to VTO objectives in batteries.

Reviewer 6

The reviewer said very relevant but, as mentioned above, it may be time to move towards earth abundant CAMs.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer saw no issues.

Reviewer 2

The reviewer said the proposed project milestones are achieved in a timely fashion.

Reviewer 3

The reviewer said the team is adequately funded to work on the proposed deliverables.

Reviewer 4

The reviewer cannot see a resource limitation encountered by the team.

Reviewer 5

The reviewer said resources of this project are sufficient.

Reviewer 6

The reviewer remarked yes; the Battery500 Consortium got the best research teams in the United States. Different teams have different expertise and strengths. The collaboration among the team would be a tremendous resource for the success.

Presentation Number: bat361
Presentation Title: Understanding and Improving Lithium Anode Stability
Principal Investigator: Yi Cui, Stanford University/SLAC National Accelerator University

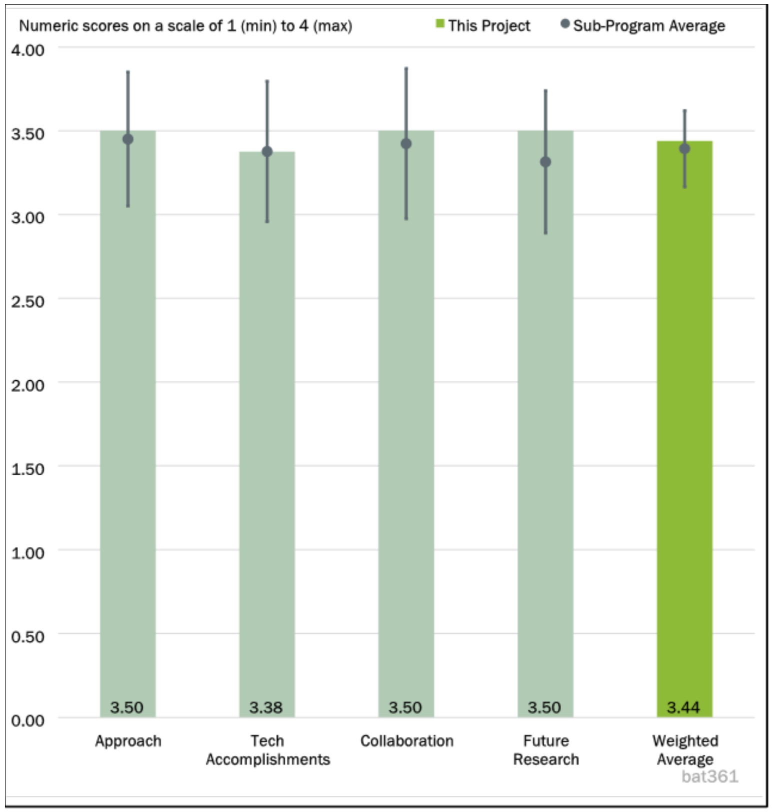

Figure 2-3 - Presentation Number: bat361 Presentation Title: Understanding and Improving Lithium Anode Stability Principal Investigator: Yi Cui, Stanford University/SLAC National Accelerator University

Presenter

Yi Cui, Stanford University/SLAC National Accelerator University

Reviewer Sample Size

A total of four reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 75% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 25% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

Question 1: *Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?*

Reviewer 1

The reviewer said the team has strong records in transmission electron microscopy (TEM) nanostructure characterization and has added operando X-ray diffraction (XRD) and transmission X-ray microscopy (TXM) to their tools. The team is attacking the most critical problems in the Li-metal battery with a 3D host composite.

Reviewer 2

The reviewer remarked as usual, the team is very creative in finding ways to address challenging issues. Here the team aims to reactivate dead Li-metal particles isolated from the anode by utilizing their dynamic polarization to the electric field and the existing voltage drop between the cathode and anode. The reviewer said the approach is novel, elegant, and consistent with the goal of Battery500 program. The reviewer believed the team deserves credit for their outside of box thinking and the thought-provoking approaches they often bring to the community.

Reviewer 3

The reviewer said the project is well designed. It aims to highlight how detached Li particles from the anode surface can move around inside the electrolyte upon applied potential across electrode. In fact, the team uncovered that the dead Li particles are not really dead materials, rather they can dance around. This phenomenon becomes important at high current rate batteries.

Reviewer 4

The reviewer remarked the project is effective in developing a robust Li-metal anode, which enables high cell specific energies of 500 Wh/kg. It does contribute to overcoming the barriers in the development of EV batteries that can meet DOE/USABC goals both in cost and performance by enhancing the specific energy and reducing the cost of EV batteries. This is part of a larger activity to develop various cell components for the 500 Wh/kg cells. The reviewer said the approach of developing a 3D Li anode architecture, new electrolytes that form stable solid-electrolyte interface (SEI), and new polymer coatings to further protect the Li anode is aimed at overcoming the technical barriers towards high specific energy and long life from the Li anode to be used in advanced high energy batteries. This year's effort has focused on understanding the formation of dead Li and retrieving part of the dead Li from the anode.

The reviewer cited as two weaknesses it would be more useful to emphasize the development of a Li anode (in conjunction with a new electrolyte) that will have reduced corrosion and minimize the occurrence of dead Li or dendrites, either with a 3D architecture or robust surface film, than to recover from the dendrite-induced effects; and the effort here seems to be a bit diffused briefly touching upon various aspects that may be interesting from academic perspective. But the Battery 500 program is more focused on advancing technology to fulfil the needs of future EVs. The reviewer said it would be greatly helpful if, and almost crucial, that a durable Li metal anode has emerged at the completion of this project to benefit the Battery 500 program

Question 2: *Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.*

Reviewer 1

The reviewer remarked the team has designed an experimental in situ set up to resemble the issue occurring inside battery system and provided sufficient evidence to support their hypothesis. The experimental results were further verified by simulation data.

Reviewer 2

The reviewer said excellent progress has been made in identifying various failure modes in the use of Li-metal anodes, using rational materials design and advanced characterization, which have been well summarized in several good publications. The project touched upon a new class of fluorinated electrolytes forming a stable SEI and new polymer coatings on the anode in previous years. The reviewer said this year's effort has focused on understanding the formation of dead Li and partly retrieving the dead Li using high charge/discharges currents. The method has been explained mechanistically in terms of field-induced migration of dendrites (dead Li) to get it connected to the anode and be reactivated. This is quite interesting and useful and will need to be validated in pouch cells.

The reviewer cited as a weakness the mechanism proposed here explains only one type of dead Li, i.e., located above the anode and formed from electronically disconnected dendrites. However, based on the reactivity and passivation characteristics of Li, it is conceivable that some of dead Li may be inside the anode, i.e., Li covered by thick SEI and buried inside the anode. In that scenario, the retrieval of dead Li may still be possible with high current discharge by the Li underneath the dead Li pushing the latter to the surface. Second, another possibility is the thermal effect from the high currents burning the dead dendrites. Finally, applying high charge/discharge currents may have adverse effects on the subsequent performance of cell. It is important to demonstrate the benefits of the proposed method in pouch cells for further validation to recover dead Li.

Reviewer 3

In the presentation, the team mainly introduced their work on the isolated Li-metal, published in 2021. This seems not directly related to the two milestones listed in Slide 4, which are studying Li nucleation mechanism using atomic force microscopy (AFM) and initiating the Li host with Cu substrate.

Reviewer 4

The reviewer remarked overall, good progress has been made on this project. The team is very productive in distributing their research results, with an impressive body of publications already appearing in high impact journals.

This reviewer's main concerns are related to the effectiveness of the approach in realistic cells. Several aspects are not clear and perhaps this can be worked into the next year's work. As there is no control on dead Li shape and orientation, the percentage of dead Li that can be activated is limited. The team demonstrated recovery of Li in Cu–Li cells with more than 100% CE, which suggests full activation of dead Li. The reviewer said considering the effectiveness depends on shape and orientation of the dead Li particles, it is unclear to this reviewer how it might be possible to get more than 100% CE.

More understanding in working mechanisms is needed. Do Li particles really migrate to the anode or do they just experience shape changes (one end gets fatter during charge and the other during discharge)? Does the shape of dead Li particles change over cycling? The reviewer remarked there was no experimental observation or evidence on actual particle migration and reattachment to the anode.

The reviewer said for sure the dead Li particles are covered with SEI products. What role does the SEI layer play and how do the different components in SEI affect the polarization, considering some are more isolating than others? In that sense, the nature of the electrolyte would be critical and it should be investigated in the study.

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer remarked collaborations across the team in the Battery500 program and SLAC/Stanford University are outstanding and well executed.

Reviewer 2

The reviewer said while other groups did not actively participate in the design and testing of this work, the outcome is critically important for other groups working under this program.

Reviewer 3

The reviewer said the team has collaborated with researchers from universities and national labs. In the future, the team should indicate the affiliations of these researchers and also their contributions.

Reviewer 4

The reviewer noted that there are several on-going collaborations with the DOE Battery500 team members, i.e., Drs. Jun Liu, Jie Xiao, Jason Zhang, Wu Xu, Stanley Whittingham and especially several faculty members both from SLAC and Stanford University.

The reviewer cited as weaknesses that it is not clear what the specific activities of collaborations are with the Battery500 team. A more active collaboration to share the materials (3D anodes, electrolytes, Li coatings) from this project with the team or analyzing the anodes from the cycled cells from the team will be useful. Further, collaboration with any battery company manufacturer will be beneficial for a rapid validation of the method/materials.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer said the team is aiming at the most critical problems in the Li-metal battery and the approaches are adequate.

Reviewer 2

The reviewer remarked that the proposed future study is well planned and involves further development of 3D Li metal anodes with stable interfacial modification and minimal volume change both globally and locally; electrolytes to form stable SEI assisted by cryoEM study; and polymer coating layer/electrolyte to facilitate the formation of stable SEI. Overall, the future plans are well aligned with the needs of Battery 500.

Reviewer 3

The reviewer said it has been discussed how to minimize the observed dancing effect of Li particles inside the electrolyte by modifying the electrolyte compositions. It is anticipated that the electrolyte composition can affect the conductivity of electrolyte and consequently the applied potential gradient to particles, and type of SEI layers on the anode surface, which could affect the detaching process of Li particles.

Reviewer 4

The reviewer said overall, the proposed future work is logical towards addressing the barriers in Li-metal based batteries. In this reviewer's opinion, the biggest challenges facing Li-metal cells is not necessarily CE or even cycle life. It is the safety issues relating to Li dendrite formation and propagation. Future research should tackle the safety issues, not only in this project but also the Battery500 program in general.

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

The reviewer said the project, as part of the Battery500 program, will contribute greatly to accelerating vehicle electrification in the United States.

Reviewer 2

The reviewer commented the project supports the overall DOE objectives by identifying the life-limiting mechanisms of a Li anode for Battery500 chemistries, including the formation of dead Li, and mitigating these limitations through a combination of theoretical analysis, advanced characterization, and experimental verification. The R&D here is being carried out well by a capable research team with access to advanced analytical techniques with the primary objective of developing a robust Li anode that can be coupled with a high energy cathode (high Ni NMC or S cathode). Overall, the reviewer found this project is quite relevant to the DOE VTO Battery program's objectives and goals.

Reviewer 3

The reviewer said an effective use of Li-metal anode is critical in achieving the Battery500 goals. Current technology faces significant challenges in terms of low CE and poor cycling life of a Li-metal anode. By developing approaches to utilize dead Li, it can improve CE as well as safety, both of which are very relevant to the overall DOE objectives.

Reviewer 4

The reviewer noted the outcome of this project is important for the battery community if the team can find a better way to prevent or minimize dancing of the dead Li particles inside the electrolyte.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer remarked the resources for the overall Battery500 project are commensurate with the scope and are adequate to achieve the targeted milestones. Budget details for this specific project have not been provided.

Reviewer 2

The reviewer said the 5-year budget looks adequate for the targeted R&D activities of Battery500. Maybe it will be more informative to specify the budget for each subproject.

Reviewer 3

The reviewer remarked it is unclear how much funding this project receives so it is difficult to judge whether or not enough resources are available. But the overall Battery500 program has sufficient resources.

Reviewer 4

The reviewer said resources provided by VTO is outstanding.

Presentation Number: bat362
Presentation Title: High Capacity S Cathode Materials
Principal Investigator: Prashant Kumta, University of Pittsburgh

Presenter

Prashant Kumta, University of Pittsburgh

Reviewer Sample Size

A total of five reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 100% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 0% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

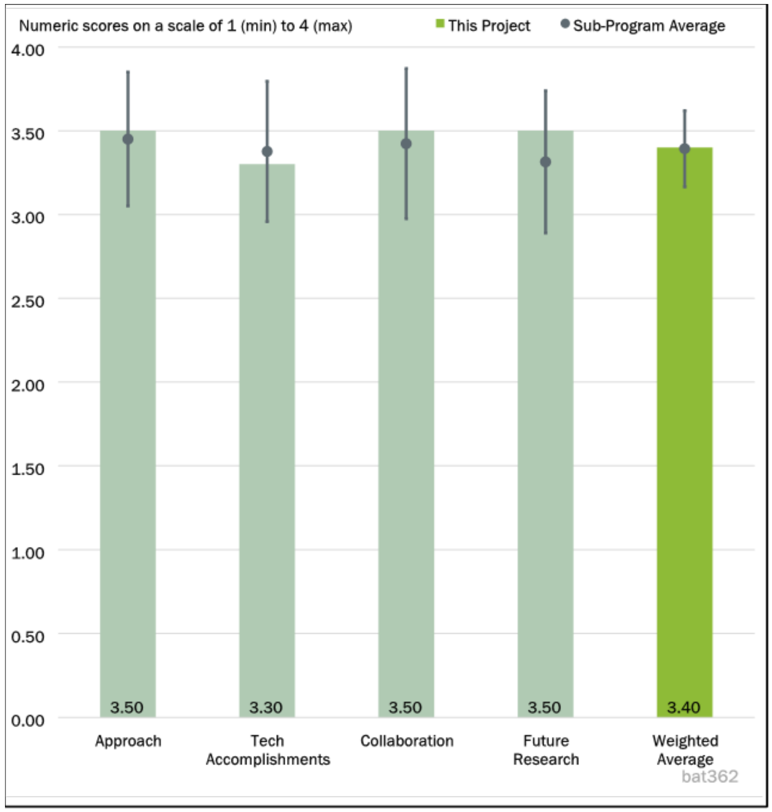

Figure 2-4 - Presentation Number: bat362 Presentation Title: High Capacity S Cathode Materials Principal Investigator: Prashant Kumta, University of Pittsburgh

Question 1: *Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?*

Reviewer 1

The reviewer said Dr Kumta is using state of the art approaches to developing and testing S cathodes.

Reviewer 2

The reviewer said the project addresses the two key technical barriers for future EV batteries, which are low specific energy and high cost; improvements in these two categories are the goals and objectives of the Battery 500 program, to which the current project belongs. Lithium-sulfur (Li-S) chemistry is in principle well suited to meet for these performance targets, but it is deterred by few technical hurdles, the most important one being the polysulfide shuttle. This project is developing new S-capturing architectures, i.e., metal organic frameworks (MOFs) and porous and mesoporous ordered ceramics (POCs), in place of conventional hierarchical porous carbons. The reviewer noted that with these architectures, it may be possible to control the chemical affinity of S with different metal/ceramic frameworks. The initial results are reasonably promising though with low cathode capacity (only 400-500 mAh/g at the cathode level at C/3 (?)). In addition, the team is developing coated separators with polysulfide trapping additives (PTA). Functional electrocatalysts are being identified for enhancing the redox kinetics of lower sulfides. These cathode improvements are being combined with a Li alloy developed earlier by the PI. Cathodes with high S loadings are being developed to improve the cell specific energy, which is still well below the target. Overall, the approach looks reasonable and effective and contributes to overcoming the barriers of Li-S chemistry.

The reviewer cited as weaknesses even though the MOFs and POCs allow high S loadings, both the S utilization and the cathode (not S) specific capacity realized so far are considerably low to give any enhancement in specific energy. It is not clear to this reviewer how high specific energies of greater than 400 Wh/kg are possible with these architectures (i.e., if the required area-specific capacity mAh/cm^2 will be achievable). The reviewer also noted that a comparison with the porous carbon-S composites is needed to verify/demonstrate the benefits with the MOFs and POCs over the carbon hosts, and that Slide 6 does not apply to Li-S; need a chart that shows the targeted cathode capacity, E/S and negative electrode to positive electrode capacity ratio (N/P).

Reviewer 3

The reviewer noted that a LiS rechargeable battery is a very challenging chemistry to realize. However, there is a huge potential for this chemistry to be used for not only EVs, but also stationary energy storage and aviation energy storage. In particular, the S cathode materials desolation in the electrolyte during electrochemical process is a critical technical barrier to be solved. The reviewer said the proposed work is focused on preventing/managing polysulfides dissolution, the major technical barrier for Li-S battery chemistry. The project developed porous organometallic framework materials (POFM) and mesoporous ordered ceramic (MOC) structures with high surface area(100-1,000m^2g-1), control porositym and porous channels in size ranges of 0.2nm–100nm, and with channels available for infiltration of S and encapsulation of doped Li_2S within and around the POFM and MOC architectures. The reviewer pointed out this high surface area and high porosity approach to manage polysulfide dissolution has demonstrate initial success supported by earlier literature and research works. The project is well designed and timeline to achieve the program goal is reasonable.

Reviewer 4

The reviewer said the PI aimed to increase the rechargeable capacity of a S cathode. By synthesizing composite including organic composite S materials, the team demonstrated a capacity over 700 mAh/g S, and high S loading. Numerical calculation was applied for the better understanding of mechanism. The reviewer remarked the project is well allied with overall Batter500 objectives and the PI indeed engaged collaboration with other participant PIs in the project.

Reviewer 5

The reviewer said that though the technical barriers are addressed by the author, the following comments should be considered.

- How will the PTA inhomogeneity be controlled?
- S (low density) should be considered while achieving 500 Whr/Kg in full cell.
- How feasible is it to use MOC and POMC for practical use, what is the stability in the electrolytes used, how much of S is trapped (in terms of percentage), and could the sulfur shuttling should be completely stopped?

Question 2: *Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.*

Reviewer 1

The reviewer noted that this project just started in 2021. In collaboration with GM, the PI has selected a universal S baseline chemistry to evaluate and benchmark the performance of the POFM and MOC confined materials. The PI's lab has developed the POFM type 1 materials, and its initial performance has been evaluated and compared with the baseline chemistry. The reviewer said performance of POFM type 1 is similar to the baseline performance. Considering the project is in its initial stage of 13% in progress, the progress made by the PI team has been excellent.

Reviewer 2

The reviewer said progress has been in alliance with the project plan. The team made and tested the composite materials and polymeric electrolyte membranes. In the next AMR, we should expect the PI to demonstrate the integrated cell consisting of all components, e.g., composite electrode, polymeric membrane, and improved Li anode.

Reviewer 3

The reviewer remarked that progress is somewhat limited as this is a new project in the full Battery500 program. This reviewer would like to see the S projects (not just this one) ensure that their loading and porosity will result in a cell that is equal to today's Li ion in Wh/L. S cells struggle to match Li-ion in this critical metric. In addition, Dr. Kumta should continue to strive to reduce the electrolyte volume in his cells. The reviewer said that earlier modeling work identified 3ml/Ah as the highest electrolyte volume that could be used, and this project is currently at 8. In addition, there is always a concern when using scaffolding structures in the cathode, which introduce dead weight and volume.

Partially to address those "dead weight" concerns, this reviewer would encourage all S projects to report mAh/g (electrode) not mAh/g (sulfur). The latter is almost universally used in the battery community and it can be very misleading.

Reviewer 4

The reviewer said excellent progress has been made in the demonstration of laboratory-scale synthesis of modified complex framework material (CFM)-S hybrid composite and type 1 POFMs with high areal capacity of 3.5 mAh/cm^2 and decent cycle life of 80 cycles. Preliminary studies with the S cathodes using the framework materials (POFM) demonstrate the feasibility and show that it possible to incorporate high S loadings of 2-5 mg/cm^2. There is a possibility that these POFMs and similar MOCs can be optimized further with desired electronic conductivity and polysulfide trapping additives. The reviewer said separators coated with polysulfide trapping additives showed some improvement in cycle life, albeit with reduced capacity. The team is identifying new functional electrocatalysts (FECs) for enhancing the kinetics of lower sulfides. It will be interesting to see how well all these cathode and separator improvements and the Li alloy developed earlier perform synergistically in Li-S pouch cells.

The reviewer cited as weaknesses even though the initial results with these MOFC/POFMs look promising, they are far from what is needed to demonstrate high specific energy of Li-S, beyond the current Li-ion batteries. This requires high S loadings combined with better S confinements or PS trapping and improved S utilization and above all, low E/S. Not much is being done on the latter. The reviewer also noted that the GM baseline coin cell data does not look as encouraging, it may not be the most current data, and that the fluoroethylene carbonate (FEC) are being designed for Li_2S2-Li_2S reaction, which occur later (deep) in the discharge or early in charge. How about the rest of reactions involving higher polysulfides?

Reviewer 5

The reviewer said that technical progress is good compared to the project plan, and the stability of MOC and POMC must be examined while trapping S. The PTA coated on Celgard showed a stable capacity of 500 mAh/g for 80 cycles while the specific capacity of 700 mAh/g is shown without PTA coating. Is it because for the full cell in the second case or any other parameter to be considered?

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer noted how this is a Battery500 project, with broad collaboration with national labs, universities, and industry. In particular, the PI is working with GM on S battery baseline characterization and benchmarking, collaboration on materials characterization using state of the art synchrotron and electron microscopy facilities at national labs, and materials and phase evolution characterization using in situ XRD with Malvern Analytical. The reviewer noted that the PI's institution also has access to the NECST Laboratory–Energy Innovation Center in Pittsburgh for the development of modified coin cell testing and carbon nanoarchitectures, and development of pouch cell testing.

Reviewer 2

The reviewer said Battery 500 has been a well-managed team of PIs. The PI of this project was well integrated in the Batter500 teams. The PI collaborated with other PIs within the program and outside the program. His efforts contributed to overall objective of the large project.

Reviewer 3

The reviewer noted several ongoing collaborations with the DOE Battery500 team members, i.e., Brookhaven National Laboratory; Idaho National Laboratory; and SLAC/Stanford University on materials characterization using synchrotron and electron microscopy facilities. There is collaboration with Malvern Panalytical for Materials and phase evolution characterization using in-situ XRD. Blomgren Consulting Services Ltd. is another useful collaborator for electrolytes, additives, system performance. The reviewer noted internal collaborations within the University of Pittsburgh, i.e., with Dr. D. Krishnan Achary for solid-state nuclear magnetic resonance (MAS-NMR) characterization and Nanomaterials for Energy Conversion Storage Technology (NECST) Laboratory–Energy Innovation Center, for the development of modified coin cell testing and carbon nanoarchitectures and development of pouch cell testing. The reviewer noted that more active collaboration with GM and possibly an industrial partner (Li-S company) will be beneficial.

Reviewer 4

The reviewer cited no issues but there is little collaboration as this project is just getting started as a Battery500 project.

Reviewer 5

The reviewer said collaboration within the project team is good, but no industry contributions are specified. This project seems to collaborate with national labs and other external entities. But it is yet to be done.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer said the proposed future study involving identification of POFM with optimized porosity and porous channels to incorporate high S loadings greater than 8mg/cm^2; type 2 and type 3 POFMs with cationic and anionic centers to trap polysulfides; mixed electronic/ionic conducting MOCs with functional electrocatalysts; and anode stabilizing agents to withstand PS attack are appropriate, effective, and will contribute to overcoming the barriers of Li-S chemistry and move towards Battery500 performance goals.

Reviewer 2

The reviewer noted that the project is centered on using POFMs ad MOCs to confine/manage polysulfides dissolution during the electrochemical operation of the S cathode electrode. The future work is focused on fully developing both the concept and materials choices to evaluate the effectiveness of polysulfides management by this class of materials. The reviewer noted that future work will also investigate the impact of the materials on the battery performance such as loading and low temperature performance.

Reviewer 3

The reviewer said S cathodes have the possibility to reach 500 Whkg and it depends on the approach for full cell considering the improvements with other team members.

Reviewer 4

The reviewer referenced prior comments on projected Wh/l and electrolyte volume

Reviewer 5

The reviewer commented future research was based on the current achievements. The PI proposed to optimize the physical properties of POFM and identity POFMs with centers of t rapping polysulfides. The reviewer said the PI should better understand the chemical nature of such entrapment.

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

The reviewer said very relevant, S is Earth-abundant and very inexpensive.

Reviewer 2

The reviewer said the project supports the overall DOE objectives by addressing the performance deterrents of Li-S cells, which otherwise have the ability to meet Battery500 goals both in terms of specific energy and cost, more than any other technology being developed under Battery500. The reviewer said advancing this technology will be beneficial in several commercial as well as U.S. Department of Defense applications, beyond EVs (dual use). The R&D strategy adopted here is sufficiently novel, and albeit with a high risk has also high pay-off. The reviewer found that overall, this project is quite relevant to the DOE VTO Battery program's objectives and goals.

Reviewer 3

The reviewer commented this project is highly relevant to the battery storage subprogram, as the success of this project leads to significant reduction of both battery cost and the usage of critical materials.

Reviewer 4

The reviewer said yes. It supports overall VTO program objectives.

Reviewer 5

The reviewer remarked Li-S battery has been considered as one of the most promising chemistries to replace Li-ion batteries in transportations. The research on S cathodes is relevant to VTO objectives.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer said resources for the overall Battery500 project are commensurate with the scope and adequate to achieve the targeted milestones. Budget details for this specific project have not been provided.

Reviewer 2

The reviewer saw no issues with resources.

Reviewer 3

The reviewer remarked resource and project timelines are well aligned.

Reviewer 4

The reviewer said proposed project milestones are achieved in a timely fashion.

Reviewer 5

The reviewer remarked the PI and team of Battery500 have adequate resources for the proposed research.

Presentation Number: bat364
Presentation Title: Synergistic Effects of Electrode and Electrolyte Materials for High Energy Lithium Cells
Principal Investigator: Jihui Yang, University of Washington

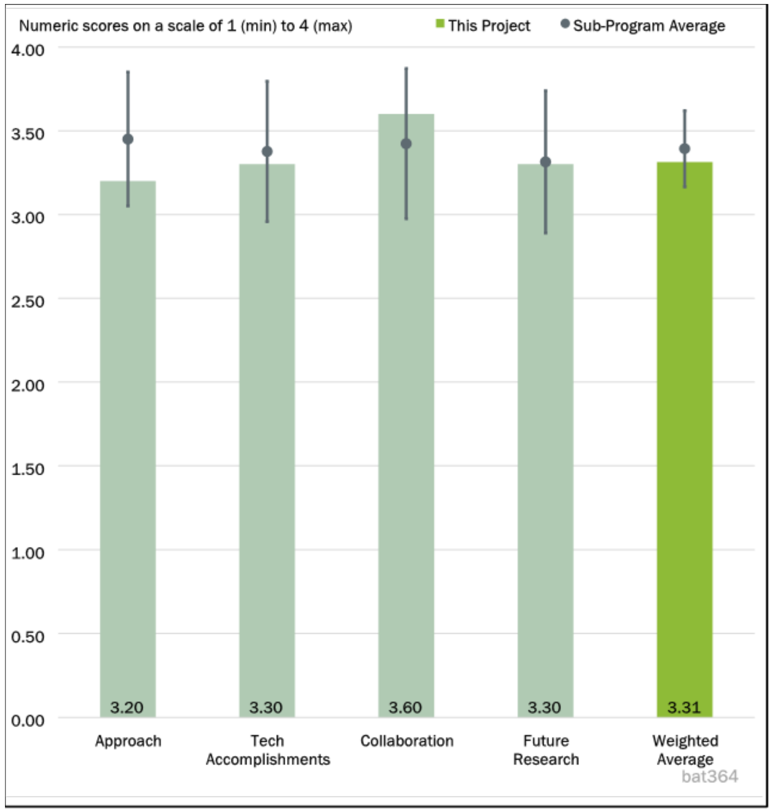

Figure 2-5 - Presentation Number: bat364 Presentation Title: Synergistic Effects of Electrode and Electrolyte Materials for High Energy Lithium Cells Principal Investigator: Jihui Yang, University of Washington

Presenter

Jihui Yang, University of Washington

Reviewer Sample Size

A total of five reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 100% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 0% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

Question 1: Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?

Reviewer 1

The reviewer remarked the PI and team are addressing key technical barriers that limit performance and stability of Li-metal cells. Enabling uniform Li-deposition and stripping using a three-dimensional (3-D) architectured current collector host, choice of electrolytes, and coatings. The reviewer said the combination of these improves the CE and capacity retention.

Reviewer 2

The reviewer found team did find the right problems to work on, including the anode structure, surface coating, and electrolytes; however, most approaches proposed have been reported in the literature. The team might need to come up with novel solution.

The reviewer said the 3D electrode architecture can reduce the local current density, but might compromise the volumetric energy density which is critical for EV applications. On the other hand, higher specific surface area could lead to more side reactions, leading to quicker capacity fading which the loading anode is well controlled.

Reviewer 3

The reviewer said that though the technical barriers are addressed by the author, the following comments should be considered.

Previously, the authors reported better results with Al-NMC811. The author should compare Al-NMC811 with LATP coating and LATP coating on a separator. The authors should address this result.

NMC811 in LiFSi seems to degrade too fast. The reviewer commented how does the LATP coating on a separator play a role when the NMC811 surface is in contact with a good amount of LiFSi. Also, the reviewer remarked address the mechanism in the case of Al-NMC811 with LATP coating and compare.

Reviewer 4

The reviewer said the University of Washington team is developing battery component integration techniques to improve the whole cell performance from coin cell to pouch cell. Even though the single component of batteries demonstrates excellent performance, without proper selection and integration of all cell components, the full battery performance can be underestimated.

Reviewer 5

The reviewer remarked the authors aim to improve the performance of Li metal electrodes via a 3D architecture and coatings on either the separator or electrode. The reviewer pointed out that the liquid electrolyte formulation and stack pressure are also critical factors to consider.

Question 2: Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.

Reviewer 1

The reviewer noted that the team has made significant progress, including carbon host for a 3D electrode, the electrolyte for anodeless battery, and coating for separator. The team could show more details on the progress such as the cycle life for carbon host and copper host, which are critical for the community to see the technology can really improve the capacity retention or not.

Reviewer 2

The reviewer found that the authors showed improvement in capacity for the LATP coating. It is important to compare Al-NMC811 with LATP coating and LATP coating on separator and explain the mechanism.

Reviewer 3

The reviewer noted that the team tried a different carbon host and choice of electrolytes to determine the higher CE. Choice of electrolytes include carbonates, ethers, and LHCE. The team conducted similar experiments on copper host. The reviewer would like to understand how these results will affect the full cell performance in presence of a Ni-rich cathode. Are the carbon host presented part of the 3D architectured current collector? If so, more information needs to provided how they are integrated with a metal current collector or stand alone.

Reviewer 4

The reviewer remarked the team has studied coulombic efficiencies of cells made with various anode chemistry and structures as well as electrolytes, and found that the F5DEE electrolyte showed the highest CE and lowest overpotential. The reviewer said it is interesting to know that no matter what carbon host was used, the cell can all achieve greater than 99% CE with the use of LHCE electrolyte. In summary, the team stated that 3D CC is beneficial for the uniform deposition of Li-metal, although the reviewer cannot find the direct evidence from the team in the slides. What is the team’s next plan for 3D structured Li host? Is 3D anode design still an essential need giving the high CE achieved from the use of proper electrolytes?

Reviewer 5

Regarding host materials, the reviewer said the authors systematically evaluated Li-metal electrode CE as a function of host material and electrolyte. Cross sectional SEM shows dense Li deposits and greater than 99% CE when LHCE is used. The reviewer said that though not presented in the AMR slide deck, the paper associated with the carbon host work (Yao Liu et al., ACS Energy Letters, 2021, 6, 1550 - 1559) yields an interesting result. For NMC622/hard carbon full cells, "The capacity retention ratios were maintained at 80%, 57%, and 53% for N:P ratios of 1:1, 1:2, and 1:4, respectively, after 200 cycles." The reviewer noted that this result aligns with the findings of BAT369. Further work should investigate the mechanism for why N/P ratio = 1:1 yields the best capacity retention. Projections should be calculated to understand how the use of host materials affects cell energy density and cost.

Regarding separator coating, the reviewer said the authors present an impressive improvement in capacity retention by using an LATP-coated separator. The baseline Li/NMC811 cell presented here should be compared to those of other Battery500 projects. The authors should also investigate the mechanism for this improvement. The reviewer noted that by citing C.-Z. Zhao et al., Science Advances, 4, 11, 2018, the authors suggest that the LATP solid state electrolyte functions as an ion redistributor to smooth out Li-metal deposition. The reviewer is not convinced that the LATP coating participates in ionic transport. The authors should study the charge transfer impedance of the liquid electrolyte/LATP interface by employing the use LATP pellets. HF scavenging could be another possible mechanism in which case alumina could be used as a much cheaper separator coating material.

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer said the team has excellent collaboration with different institutions in the Battery500 Consortium. The expertise among the collaborators is complimentary, which is critical for the project to be successful.

Reviewer 2

The reviewer said the PI and team have demonstrated outstanding collaboration with other Battery500 teams. The reviewer noted that PNNL and Stanford provided electrolytes for the study, and the University of Texas (UT) Austin provided Ni-rich cathodes. Overall, a highly integrated project and deliverables.

Reviewer 3

The reviewer remarked the authors demonstrated excellent collaboration and coordination with other organizations.

Reviewer 4

The reviewer said collaboration within the project team is good, but no industry contributions are specified. This project seems to collaborate with national labs and other external entities. But it is yet to be done. The reviewer said it might be good to work with Nb coated NMC811 (Stan) combining LATP coating.

Reviewer 5

The reviewer commented the team is in close collaboration with PNNL on separator coating, UT-Austin and Binghamton University on Ni-rich NMC synthesis and characterization, Stanford on new electrolyte formulation, and INL on pouch cell test.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer said proposed future work makes sense.

Reviewer 2

The reviewer remarked the team has clearly defined the future work based on the current findings. For the 3D architecture anode design, relevant length scales might be considered to avoid the reduction of energy density due to unnecessary increase of anode thickness.

Reviewer 3

The reviewer said it is proposed that Li host behavior without depletion of Li-metal at discharge state will be investigated. It might be possible to reduce depletion of Li-metal and may not be possible without depletion.

Reviewer 4

The reviewer said that more specific details need to be provided beyond a high level. For example, what kind of new electrolytes and host (beyond what is presented here). The reviewer said specific targets such as achievable current densities and cycle number need to be projected to measure success against existing base line results. For example- "Identify proper experimental parameters- E/S ratio". What would be the new E/S ratio the team is targeting to have. The reviewer said that Phase-1 of Battery500 has already provided some base line performance and the goal should be compare and move beyond what had been accomplished.

Reviewer 5

The reviewer said the proposed future research is too general. It should show some specific directions which address the barriers for the project.

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

The reviewer said yes, it is relevant to the target for the Battery500 Consortium.

Reviewer 2

The reviewer said yes, it supports overall VTO program objectives.

Reviewer 3

The reviewer remarked the project is part of the Battery500 goal of achieving a cell level target of 500 Wh/Kg for Li-metal and various approaches to eliminate excess inactive materials and improve cell performance by using next generation electrolytes and coating strategies. One of the goals is to deliver the results in a pouch cell format ready for industry to evaluate and follow up.

Reviewer 4

The reviewer commented the project supports the VTO battery objectives in high energy density and long cycle life by developing better interfaces and anode architectures.

Reviewer 5

The reviewer noted that this project dovetails with work on advanced electrolyte formulations.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer said the team has strong collaborations with other Battery500 teams and sufficient resources in conducting the battery performance evaluation task and interface modification task.

Reviewer 2

The reviewer said the resources of this project are sufficient.

Reviewer 3

The reviewer remarked the proposed project milestones are achieved in a timely fashion.

Reviewer 4

The reviewer noted that this is a consortium project—all PIs and their labs together deliver the milestones through collaboration.

Reviewer 5

The reviewer commented yes, the Battery500 Consortium has the best research teams in the United States and can provide sufficient resources to the success of this project.

Presentation Number: bat365
Presentation Title: Stabilizing Lithium Metal Anodes by Interfacial Layer and New Electrolytes
Principal Investigator: Zhenan Bao, Stanford University/SLAC National Accelerator University

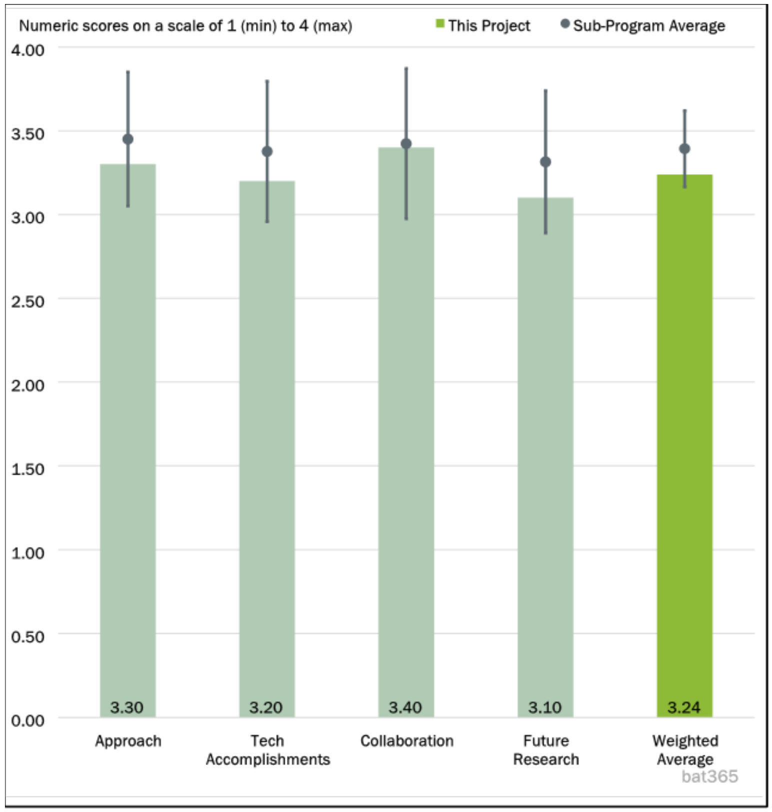

Figure 2-6 - Presentation Number: bat365 Presentation Title: Stabilizing Lithium Metal Anodes by Interfacial Layer and New Electrolytes Principal Investigator: Zhenan Bao, Stanford University/SLAC National Accelerator University

Presenter

Zhenan Bao, Stanford University/SLAC National Accelerator University

Reviewer Sample Size

A total of five reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 80% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 20% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

Question 1: *Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?*

Reviewer 1

The reviewer detailed that the proposed work concerns high CE electrolytes and coatings to stabilize a Li-metal anode. The work encompasses synthesis, cell assembly and theory. The reviewer is particularly impressed by the effort to use theory to design the optimal solvation structure for the applications. The proposed molecular structures on Slide 6 are interesting as they include both oxygen and fluorine atoms to address the trade-offs between ion transport and electrochemical stability. The reviewer said the PIs have an excellent track record of publications in high profile journals. The project is well-designed and well planned.

Reviewer 2

The reviewer said very good approaches being evaluated for enabling Li metal.

Reviewer 3

The reviewer said the PIs present results showing an anti-correlation between Li-ion solvation in an electrolyte and Li-metal stripping/plating reversibility. To do so, the team systematically changed the electrolyte solvating power through solvent/non-solvent mixtures, and selective fluorination of small molecular ethers. The reviewer noted the team’s electrochemical analysis clearly indicates the important relationship between poorly solvating electrolytes and improved Li-metal reversibility and plating uniformity.

Reviewer 4

The reviewer said it appears that the team has an excellent division of labor, and is using expertise and personnel time appropriately across the different sites. It appears that the milestones have mostly been satisfied on time and the team has achieved progress toward more stable batteries with higher energy density.

Reviewer 5

The reviewer noted that according to Slide 6, the PIs on this task have four different approaches for dealing with the CE. The reviewer remarked when the team talks about CE, it is hard to tell if the team is talking about the cell's CE or the rate of side reactions on the Li. Approach 1, make a new conductive electrolyte that is stable with carbon-fluorine (C-F) and C-O chemistry (the reviewer does not believe this will stand up to Li's reduction potential.) The reviewer said that Approach 2 is to develop a new co-solvent that is stable to Li (easier said than done). Approach 3, use a localized high concentration electrolyte, which is an electrolyte with a reductively stable salt like LiFSI, another solvent that is miscible with the main solvent but has a low solubility for the salt (this means that the overall salt solubility will decrease but the local solvation of the salt will be primarily with the main solvent). The reviewer was not sure why this will work if it has already been shown that the main solvent is not stable. (Is LiFSI 100% stable against Li?). Approach 4 is an inorganic-SEI and electromechanical stabilization stiffened electric double layer. Overall, it is good to come up with an electrolyte that is overall more stable to reduction than is the standard electrolyte but to fix the lithium passivation problem will require more, like a fully passivating film made either in situ or ex situ. No real discussion of the SEI that will be formed if any of these electrolytes is not stable, except maybe the last Approach.

Question 2: Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.

Reviewer 1

The reviewer said technical accomplishments include: a systematic study on Li-ion solvating power of dioxolane mixtures with various co-solvents and their impact on Li plating, the development of partially fluorinated ethers and a study on their role in influencing Li metal plating reversibility, and an optimal electrolyte that promotes 'quick activation' of high CE Li metal cycling. Given the level of funding, this is good progress for the Stanford Battery500 team.

Reviewer 2

The reviewer said the PIs have studied mixtures of solvents (nonfluorinated) to study the interrelationship between solvation structure, studied by NMR, cycling behavior, studied in electrochemical cells, and electrodeposited Li morphology by SEM (Slide 7). The reviewer noted that the main conclusion is lower solvation of Li^+ results in lower overpotentials and more planar deposition. This knowledge is important for enabling Li-metal anodes. The reviewer was impressed by the systematic study of ionic conductivity as a function of chain length shown in Slide 9. Perhaps the PIs may want to study the effect of chain length on other ion transport properties, relevant to battery operation. The reviewer pointed out the effort to understand CE as a function of molecular composition in Slide 12 is also noteworthy. The PIs have made excellent technical progress. Some indication of how theory is being incorporated into molecular design would have been good to see.

Reviewer 3

The reviewer said very good progress reported, and the reviewer would encourage the PIs to adhere to standards discussed elsewhere, such as a maximum electrolyte volume of 3ml/Ah. The reviewer believed this team is using 8, which makes performance look much better than it would at 3 but also hurts cell Wh/kg due to the extra non active material weight.

Reviewer 4

The reviewer said the new electrolyte materials developed and presented are enhancing CE and longevity of batteries at the pouch cell level, which is exciting. Below are specific technical questions related to the PI's presentation of results at the AMR. (These are repeated below in the Question 8 section.)

The reviewer said regarding the idea of electrolyte formulations with varying "solvation strength" for Li+: The PI mentioned that changes to the electrolyte solvents can cause the anion to solvate Li^+ more or less strongly. Can one also think of this as the solvent solvating anions better than it solvates Li^+? Furthermore, C-F bonds in solvent molecules are quadrupolar and assumedly should associate better with C-F bonds in FSI than C-H bonds in other species. The reviewer remarked might you use another related concept that is the use of momentarily associated ion-ion clusters that are locally complex structurally? From the answer to the related question during the talk, it sounds like this team is thinking about some these perspectives.

Regarding solvation and ^7Li NMR chemical shift trends: What does this team see with anion shifts (19F) and/or solvent shifts (1H or 19F)? Note also that FSI and PF6 have only one 19F shift, but you also have 13C for FSI and 31P for PF6.

The reviewer remarked would it be instructive to compare these DME-based electrolytes with the perfluoropolyether (PFPE) systems studied several years ago (Balsara and DeSimone)?

Reviewer 5

The reviewer said the first TA is in regards to mixing different solvents with DOL (considered a good solvent for limited dendrite growth.) They explain that NMR can determine if the solvation of Li increases or decreases with the addition of solvents to DOL. DOL is a poor solvent in that the over potential for Li deposition increases with cycling. The reviewer said the presenters believe that reduced solvation reduces the over potential for Li deposition and provide some data to that effect. DOL plus hexane in equal parts shows the lowest Li solvation of the samples tested, the lowest overpotential for Li deposition and the best, long term, steady cycleability. The reviewer said the team believes part of this is because the deposited Li is more two dimensional when deposited from DOL+Hex.

The next TA is results based on an H-cell. The reviewer suggests that the team put a baseline electrolyte in one half (DOL?) and the standard plus a diluent in the other half of the H-cell and measured the voltage of the cell as a measure of whether the Li-ions are more or less solvated. This voltage is a measure of both the thermodynamic difference and the transport properties through the barrier between the two cells. The reviewer said the team also show a plot of CE versus the voltage in their H-cell—the reviewer was unclear how the team got the CE data. Without knowing where the CE data came from, it is hard to tell what this means. The reviewer noted the team shows conductivity of a solvent with three blocks—the one in the middle has lots of C-F bonds and the ones on the outside are polyethylene oxide (PEO) type strands. The team show that the more PEO type, the better the conductivity but the lower the oxidative stability. The reviewer suggests it is not clear why the oxidative stability is of note for protecting Li.

The reviewer remarked that the team then shows that the best overall performing cells are with a solvent that is a short piece of PEO with fluorinated end groups and say this is the best combination of conductivity, CE, Li, CE, overpotential, and oxidative stability. The reviewer was unsure how the team is measuring the lithium CE.

The next slide shows the results of lithium stripping and plating. For 1 mAh/cm^2 of cycled Li the team gets an efficiency as high as 99.9% but for the 5 mAh/cm^2 cell, the team gets 99.5%. If the cell is to cycle 1000 times, 0.005 times a thousand is 5 which means they will lose 5 times the amount of the minimum amount of Li needed before they run out of Li in the cell. For a 5 mAh/cm^2 cell, that is 5 microns/mAh x 25 = 125 microns of Li - way too much. The reviewer noted that the team also shows full cell cycling data that falls roughly into two groups: one group is around 150 cycles and the other between 200 and 250 cycles. This would not be considered excellent. The reviewer said these projects could use more interaction among the participants. It comes off as 4 projects under one banner.

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer said Battery500 is a strong team (PNNL, Stanford, Binghamton, etc.), with clear synergies between team members. Overall, the coordination between team members appears excellent and quite collaborative.

Reviewer 2

The reviewer said that Slides 5 and 13 (which are similar) show the connection between the team and other members of the Battery500 and U.S.-German teams. Evidence for these collaborations is provided clearly in the publication list on Slide 20.

Reviewer 3

The reviewer said good interaction with other team members.

Reviewer 4

The reviewer remarked that it appears that the collaborations among the team members at multiple universities (Stanford, UT-Austin, University of California-San Diego (UCSD), University of Washington, and SUNY Binghamton) and national laboratories (PNNL, SLAC) are fruitful and solidly operating. The reviewer might expect more attempt to understand (and present) how the electrolyte stability and conductivity are related, and how this relates to ion clustering or local chemical environment. This is a challenging long-term project, and so developing understanding should perhaps feed into success more efficiently.

Reviewer 5

The reviewer said Slide 13 indicates that SLAC/Stanford can work in isolation on developing electrolytes and can reach out for other parts of the battery when needed. This task appears as four separate projects with no meaningful collaboration.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer said the proposed work slide (Slide 14) is an accurate description of the program, where it stands, and what the PIs are trying to control with the ultimate goal of enabling high energy density Li-metal batteries.

Reviewer 2

The reviewer remarked good future plans.

Reviewer 3

The reviewer said proposed future research addresses the need for enhanced mitigation of electrochemical breakdown of battery components during use, notably electrolytes. From the presentation, the specifics of new directions are not exceedingly clear.

The reviewer provided specific technical questions related to the PI's presentation of results at the AMR, which might be useful for future research. Regarding the idea of electrolyte formulations with varying "solvation strength" for Li+: The PI mentioned that changes to the electrolyte solvents can cause the anion to solvate Li+ more or less strongly. The reviewer asked can one also think of this as the solvent solvating anions better than it solvates Li+? Furthermore, C-F bonds in solvent molecules are quadrupolar and assumedly should associate better with C-F bonds in FSI than C-H bonds in other species. Might the PI use another related concept that is the use of momentarily associated ion-ion clusters that are locally complex structurally? From the answer to the related question during the talk, it sounds like this team is thinking about some these perspectives.

Regarding solvation and 7 Li NMR chemical shift trends: What does this team see with anion shifts (19F) and/or solvent shifts (1H or 19F)? Note also that FSI and PF6 have only one 19F shift, but you also have 13C for FSI and 31P for PF6.

The reviewer asked would it be instructive to compare these DME-based electrolytes with the perfluoropolyether (PFPE) systems studied several years ago (Balsara and DeSimone)?

Reviewer 4

The proposed future research seems reasonable, although somewhat vague—the future work slide provides rather high-level research directions with no clear specific plans, although the reviewer is sure the plan entails an extension of the current year's technical accomplishments.

Reviewer 5

The reviewer said future work needs to be carefully reconsidered. The first thing the team conveys is the need to understand the balance between stability and solvation. But the investigators already know that oxygen in their solvent promotes Li solvation and fluorine in their solvent promotes electrochemical stability. The team could use more collaboration to think through this complicated problem.

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

This project strongly supports VTO objectives, as understood by this reviewer. This project is producing new electrolyte and interfacial layer materials that are improving the efficiency and longevity of Li-based batteries.

Reviewer 2

The reviewer pointed out that stabilizing and enabling Li-metal is critical.

Reviewer 3

The reviewer said the project is relevant to two subprograms—batteries and materials. The PIs present a systematic approach for resolving the conflicting needs of electrochemical stability and rapid ion transport.

Reviewer 4

The reviewer remarked the project supports the VTO subprogram objectives because getting to pure Li anodes leads to a major advance in energy density. This, as seen, is an extremely challenging objective, especially with a liquid electrolyte.

Reviewer 5

The reviewer said yes; if enabled, reversible Li-metal electrodes could provide an important step forward in Li battery energy density. This work aims to understand how to control Li metal deposition in liquid electrolytes.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer said resources are sufficient to meet milestones in a timely fashion. The PIs have an excellent track-record of publishing their findings in the literature.

Reviewer 2

The reviewer remarked the team has access to world-class facilities, tools, and laboratory support personnel. The funding level and collaborative team appears to be quite sufficient to accomplish the stated goals. It is difficult to say $75 million from DOE is appropriate for this entire team, because this reviewer was tasked with only assessing one PI (Bao) in this huge multi-PI and multi-institution effort.

Reviewer 3

The reviewer commented the level of funding is sufficient given the size of the team and the problems facing Li metal electrodes.

Reviewer 4

The reviewer saw no issues. But, as with all other Battery500 projects, it is not possible to answer this question as no PI reported the funding to their specific project. This is not a comment directed at this project, but rather at all Battery500 projects.

Reviewer 5

The reviewer remarked the PIs do not list the amount of resources used in this project. The reviewer noted that instead of listing generally how much each of the projects in this task cost, which the team could have done on the second slide under budget, the team listed the entire budget of the program. Therefore, this reviewer must assume that $75 million is being spent on this effort, which is way too much.

Presentation Number: bat366
Presentation Title: Manufacturing and Validation of Lithium Pouch Cells
Principal Investigator: Mei Cai, General Motors

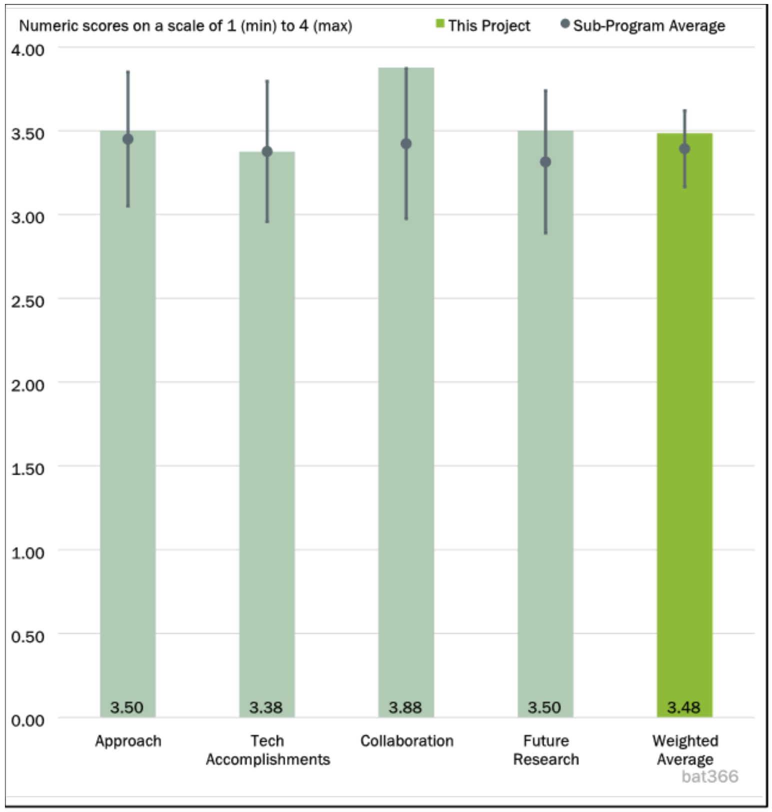

Figure 2-7 - Presentation Number: bat366 Presentation Title: Manufacturing and Validation of Lithium Pouch Cells Principal Investigator: Mei Cai, General Motors

Presenter

Mei Cai, General Motors

Reviewer Sample Size

A total of four reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 100% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 0% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

Question 1: Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?

Reviewer 1

The reviewer said the team has built and commissioned facilities for continuous fabrication of Li-S, and made and tested the pouch cells. Dual function electrolyte has been designed and tested for the high performance of Li-S batteries. The reviewer noted that although the stability is still low, the team should be able to overcome the technical barriers by incorporating the technical advances from the collaborating team members in next stage.

Reviewer 2

The reviewer said the team proposed well-known approaches for their Li-S cell, polymer electrolyte to protect Li-metal and liquid electrolyte for S cathode, with coated separator to trap polysulfide.

Reviewer 3

The reviewer remarked the project is well planned and designed to meet the challenge of doubling the energy density of current commercial EV Li-ion cells. The technical achievements reported indicated that the timeline is reasonably well-planned.

Reviewer 4

The reviewer said that the team demonstrated Li-S pouch cell performance using dual electrolyte approach—solid electrolyte coating on Li-metal and liquid for S. The reviewer noted a dedicated coating line for S electrode fabrication and scale-up—single and double side coating of C/S electrodes. The team developed a coating on separators for trapping polysulfides

Question 2: Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.

Reviewer 1

The reviewer pointed out that the dual-phase electrolyte proves to work for the stability of the anode. Implementing the polysulfide trapping layer enhanced the cyclability of Li-S batteries. The team accomplished the continuous fabrication of Li-S cathode process with normal and high S loading with reasonable cycling stability of 100 times at 0.1C.

Reviewer 2

The reviewer said the team has made significant progress and is on track of developing a high energy Li-S pouch cell. In the past, many researchers have been working on developing Li metal and Li/S cell technologies but have to give up because they cannot meet the challenge of improving the energy density and cycle life of the Li metal anode system. The GM team has made Excellent progress in increasing the energy density and is now working toward improving the cycling of Li metal anode system.

Reviewer 3

The reviewer said the team demonstrated good cycle life in both medium (4mAh/cm^2) and high loading (6mAh/cm^2), albeit very excessive Li (500um) and high E/S ratio of 8. The reviewer was not clear how the CE can be greater than 1 when the discharge capacity is less than the charge capacity (Slide 11). The team, and any other Li-S performers, should report the self-discharge of their cells at 100% state of charge at RT to track progress on polysulfide mitigation. The reviewer said that CE alone is not specific enough to track the polysulfide issue because CE is affected by Li-metal cycling.

Reviewer 4

The reviewer remarked the polymer coated Li-metal shows stable plating and striping compared to uncoated ones at the state current density. The reviewer would like to know if 1000 hrs. is arbitrary or based on some baseline performance metrics.

The reviewer noted that the metal oxide/C coating on separators traps polysuflides and improves performance but still there is a loss of Li-inventory. More mechanistic investigation is needed to conclude that coating separator improves sulfur utilization. The reviewer also noted that reported performance for Li-S cells for two S loadings with described protocol is an excellent progress. What is justification is there for 70% porosity? Is it optimal? How does it compromise with the overall energy density?

Question 3: Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?

Reviewer 1

The reviewer remarked there is outstanding collaboration and coordination across the Battery500 teams. Each Battery500 team has been using their own expertise to develop solution for a specific challenge that help the GM team integrate the developed solutions into the high-energy Li-metal anode pouch cell.

Reviewer 2

The reviewer said the team works in close collaboration with other team members by incorporating the technical advances from cathode, anode, electrolyte as well as fabrication involving the UCSD, University of Pittsburgh, Penn State University, PNNL/University of Washington, and General Motors.

Reviewer 3

The reviewer said the team is very collaborative with other Battery500 partners with each team/institution having specific goal provided for S electrode, electrolyte, or developing diagnostic tools.

Reviewer 4

The reviewer remarked GM partnered with four national labs and several well-known universities working on battery technologies.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer commented the team will continue to optimize the Li-S fabrication by collaborating with other team members, and fabricate the large pouch cell of 5Ah. With the facilities available, the reviewer expects more exciting results.

Reviewer 2

The reviewer said the proposed future research is outstanding and clearly defined to mitigate and develop solutions for the current challenges of preventing Li dendrite formation and polysulfides cross contamination. The proposed future research program has a great potential of reaching the goal of a stable Li cell with energy density of 500 Wh/kg.

Reviewer 3

The reviewer remarked scaling up Li-metal coating and electrode fabrication is an important step forward. Team needs to provide go/no-go metrics. The reviewer pointed out that carbon/sulfur electrodes have been tried before—what is unique about the S electrode architecture that has the right loading and performance targets.

Reviewer 4

The reviewer said future work should focus on demonstrating cycle life with significantly reduced excess Li and reduced E/S ratio, and also self-discharge rate comparable to transition metal oxide cathodes.

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

The reviewer commented the project is extremely relevant for the United States to reach the goal of carbon-free clean energy. It is practically impossible to reach that goal without the success of this project.

Reviewer 2

The reviewer said the project is relevant to materials development, batteries production, and also EVs.

Reviewer 3

The reviewer said proposed work met VTO program objectives.

Reviewer 4

The reviewer said relevance is adequate. Having an active industry partner helps to support deliverables and commercialization.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer said with the fabrication facilities available, the allocated $15 million for running the tests will be sufficient for manpower, consumables, and characterization tests.

Reviewer 2

The reviewer said resources are adequate.

Reviewer 3

The reviewer said the long-term commitment and funding resources that DOE provides to this project are the key enabler for the Battery500 team to be successful. The resources are sufficient for the project to achieve the stated milestones on time.

Reviewer 4

The reviewer said sufficiently large funding for the large team.

Presentation Number: bat367
Presentation Title: Multiscale Characterization Studies of Lithium Metal Batteries
Principal Investigator: Peter Khalifah, Brookhaven National Laboratory

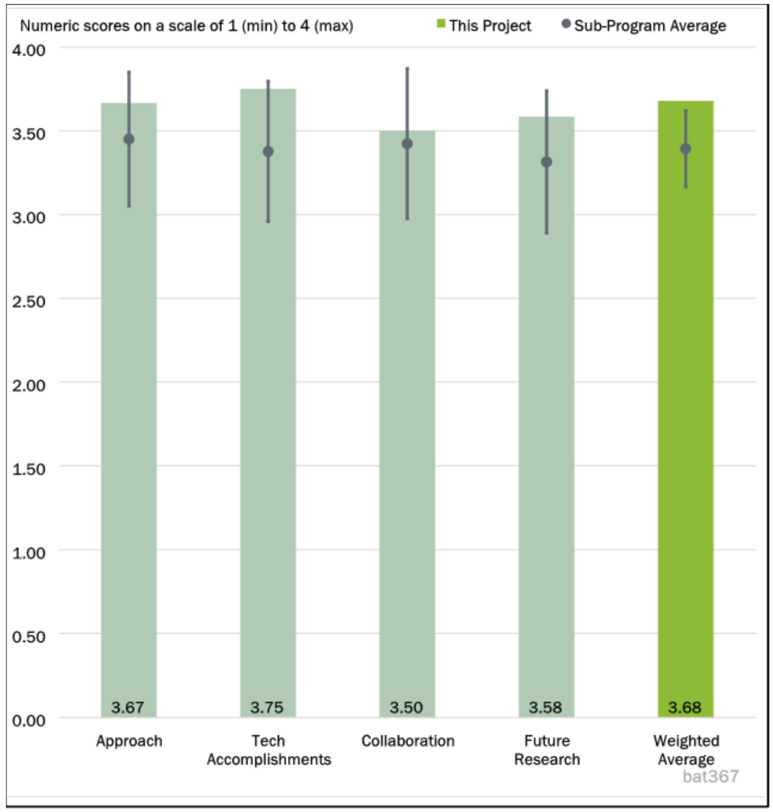

Figure 2-8 - Presentation Number: bat367 Presentation Title: Multiscale Characterization Studies of Lithium Metal Batteries Principal Investigator: Peter Khalifah, Brookhaven National Laboratory

Presenter

Peter Khalifah, BNL

Reviewer Sample Size

A total of six reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 100% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 0% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

Question 1: Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?

Reviewer 1

The reviewer said the project is well designed with very effective approaches. The combination of synchrotron techniques and the density functional theory (DFT) calculation worked well in understanding and solving problems in Li-metal batteries. The reviewer noted the team has very strong records in applying synchrotron structure/morphology characterization in battery study.

Reviewer 2

The reviewer remarked synchrotron measurements are being integrated with modeling. Going from single atoms to full pouch cell—these are important and are not always done.

Reviewer 3

The reviewer noted the team utilized X-ray synchrotron XRD to study Li-deposition on copper in a pouch cell (anode free), and used spectroscopic tools to investigate the role of additives towards improving the performance of Li-NMC 811 cells.

Reviewer 4

The reviewer remarked the team has developed a very unique capability to quantify the dead Li with good spatial and temporal resolution, which can provide a critical fundamental understanding of the failure mechanism of Li metal electrolyte and guidance for how to optimize the electrode and electrolyte design.

Reviewer 5

The reviewer noted that though the technical barriers are addressed by the author, the following comments should be considered.

The reviewer said the authors must show the baseline electrolyte. It seems $LiPF_6$. The authors claim adding $LiPO_2F_2$ greatly improves cycling of Li/NMC811 cells at high voltage. It is good. However, Stan showed Nb coating on NMC811 provided 220 mAh/g in LiFSi. How will the team collaborate when they use two different electrolytes?

Reviewer 6

The reviewer commented the team uses synchrotron facilities to run operando or ex situ characterizations on pouch cells, targeting understanding the Li deposition/stripping behaviors, sulfurized polyacrylonitrile (SPAN) intermediates, and the effects of electrolyte additives. The approach also has a portion of modeling and calculation efforts stated in the project.

Question 2: Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.

Reviewer 1

The reviewer said the team has established the excellent capability to characterize the Li plating and stripping, and understand the structural and bonding changes in SPAN. Most importantly, the team was able to get deep understanding of the enhanced the performance of electrolyte additives with the novel approaches.

Reviewer 2

The reviewer said the team has made impressive progress on the project, including the TXM studies on CEI interphase protection, and X-ray pair distribution function (PDF) studies on SPAN cathode. The team has recently made excellent progress in the characterization of Li metal plating stripping and the mechanism studies in $LiPO_2F_2$ addition.

Reviewer 3

The reviewer said direct quantification of Li-metal in a cycling pouch cell is a difficult experiment. The team succeeded in getting very useful information about the Li-metal distribution with a novel experimental design and the high intensity of synchrotron X-ray beams. This will allow more thorough studies on the Li-metal deposit/depletion mechanism in electrochemical cycles. The reviewer noted the Li-metal scattering is rather weak even with the high-intensity X-rays, which may cause difficulties in detecting Li signals at the beginning of the Li deposit, where the thin Li layer has not formed a crystalline form. The reviewer said the team may consider combining the TXM technique to investigate the oxidation state of the NMC cathodes to compare the spatial map of Ni2+/Ni3+ distribution and the Li-metal distribution.

Ex-situ PDF analysis on SPAN at different state of charge provides interesting insights into the cathode and the new UCSD electrolyte. Adding the proper amount of lithium difluorophosphate in the electrolyte greatly enhanced the cycling performance of the high-nickel cathode, which represents a great breakthrough in high nickel cathode Li-metal battery. The team reports their results in trying to understand the origin of the cycling enhancement with DFT calculation and spectroscopic studies.

Reviewer 4

The reviewer noted that the addition of $LiPO_2F_2$ greatly improves cycling of Li/NMC811 cells at high voltage. It is important to show the baseline electrolyte and compare $LiPF_6$ and LiFSi.

Reviewer 5

Regarding the spatial resolution of Li-metal, the reviewer noted it is related to pre-existing structure/chemistry--related to local NMC, but where does the heterogeneity come from? Heterogeneity comes during discharge, and it is Li-metal heterogeneity that causes heterogeneity in cathode. The reviewer noted that operando measurements of Li-metal a big advance

Reviewer 6

The reviewer noted that one of the conclusions from Li-plating and stripping experiment from synchrotron XRD suggest strong inhomoegeneity observed during discharge. One comment stated, "Conventional wisdom says Li-deposition is the challenge," and the reviewer likes to know how broad is the statement. Li-plating process can be highly non-uniform as well. The reviewer cited good progress that has been towards applying X-ray spectroscopy for studying the TM dissolution with and without $LiPO_2F_2$ additive, and the X-ray tomography aided by AL-ML approaches used to quantify degree of cracking in 811 cathodes.

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer commented the team has been extensively collaborating with other teams in the Battery500 Consortium, providing the critical information for other teams to further optimize the materials they are developing. The synergistic effect among the teams is reflected by the fruitful publications and presentations.

Reviewer 2

The reviewer remarked the team works in close collaboration with other Battery500 partners. SPAN samples provided by UCSD, electrolyte additive and SEI (Army Research Lab). High-Ni NMC provided by UT-Austin.

Reviewer 3

The reviewer said the team is in close collaboration with almost all Battery500 teams.

Reviewer 4

The reviewer said the team has collaborators from universities and other national labs. All contributions from the collaborating teams are clearly listed. The team may consider develop collaborations with industrial partners.

Reviewer 5

The reviewer said many collaborators with both universities and labs, and no industry collaborators

Reviewer 6

The reviewer said the collaboration within the project team is good, but no industry contributions are specified. This project seems to collaborate with national labs and other external entities. But it is yet to be done. The reviewer said it might be good to show the performance in LiFSi so that it can be combined with Nb coated NMC811 (Stan) and LATP coated separator (Yang).

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer remarked unique capability to answer questions that have long been outstanding.

Reviewer 2

The reviewer said the team is on the right track in conducting the project and the proposed future work is continuing on what the team is doing well now but aiming at new problems.

Reviewer 3

The reviewer said the proposed future research can achieve its target to some extent. It might be good to focus on one MNC811 or S. It is important to discuss the changes on Li electrodes during charge/discharge.

Reviewer 4

The proposed future research directions are rational and cover the most critical problems which need to be tackled for future progress.

Reviewer 5

The reviewer commented the proposed future work is well based on previous studies and is a good step forward in achieving optimized battery performance in various battery systems. The characterization technique is feasible in understanding the proposed scientific problems. It will be nice to see more integration of the characterization with modeling effort in future work.

Reviewer 6

The reviewer said more specific goals and details needs to be provided as part of future research needs. The high level science goals are excellent but how do they connect to specific objectives and milestone for Battery500?

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

The reviewer remarked the team is working on increasing the energy density of Li-metal batteries using advanced techniques, which is directly supporting the VTO's objective in vehicle electrification.

Reviewer 2

The reviewer said the techniques are aimed precisely at solving the problems of Battery500.

Reviewer 3

The reviewer said yes, it supports overall VTO program objectives.

Reviewer 4

The reviewer commented this project is focused on using advanced tools and characterization for studying bulk and interfaces involving Li-metal and high voltage cathode. This work supports development of high energy density Li-ion.

Reviewer 5

The reviewer remarked the team developed and deployed characterizations, especially synchrotron methods, to assist the optimization of cathode materials and improvement of cell designs. It is supportive of DOE's battery objectives in achieving high energy density and long cycle life batteries.

Reviewer 6

The reviewer pointed out that understanding the failure mechanism is the prerequisite for extended battery life with improved energy density.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer said resources are sufficient.

Reviewer 2

The reviewer said the project (second phase) is just starting. The funding level ($75 million for 5 years) is proper.

Reviewer 3

The reviewer said the team has what it needs.

Reviewer 4

The reviewer said proposed project milestones are achieved in a timely fashion.

Reviewer 5

The reviewer said ongoing collaboration listed in the presentation is a good proof that the resources of the project are sufficient, not only from the expertise from the teams, but also from the spectrum of work scope.

Reviewer 6

The reviewer commented the work requires constant access to the user facilities and close collaboration with partner teams. The current resources seem to be sufficient to conduct the proposed work.

Presentation Number: bat368
Presentation Title: Full Cell Diagnostics and Validation to Achieving High Cycle Life
Principal Investigator: Eric Dufek, Idaho National Laboratory

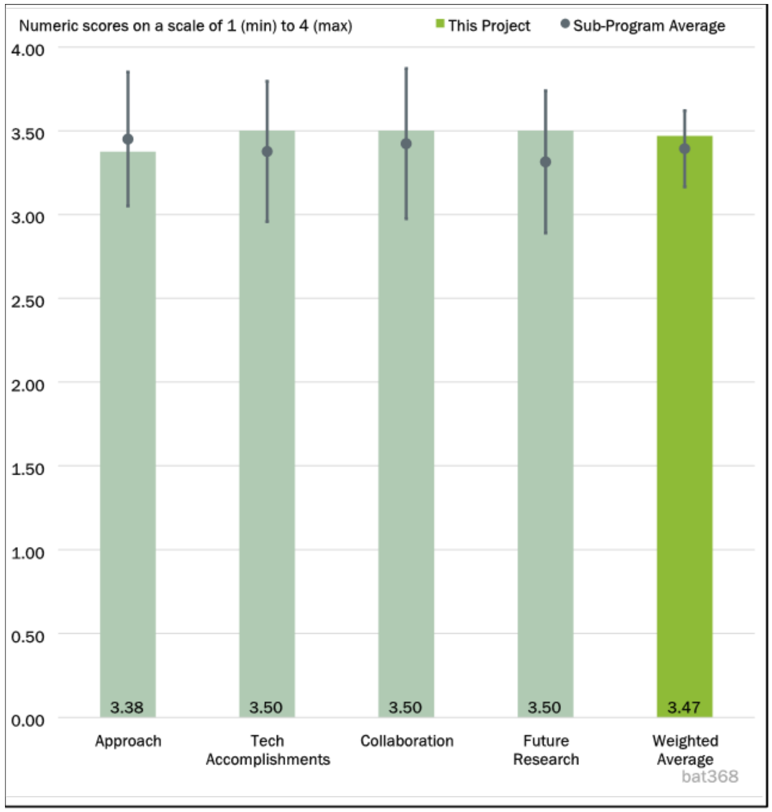

Figure 2-9 - Presentation Number: bat368 Presentation Title: Full Cell Diagnostics and Validation to Achieving High Cycle Life Principal Investigator: Eric Dufek, Idaho National Laboratory

Presenter

Eric Dufek, INL

Reviewer Sample Size

A total of four reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 100% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 0% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

Question 1: *Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?*

Reviewer 1

The reviewer remarked the team has conducted experiments and modeling for the performance of Li-ion batteries as well as Li-S batteries, and published five journal papers one in Nature Energy and one in Advanced Energy Materials.

Reviewer 2

The reviewer noted the team's approach is to understand impact of electrolyte on SEI and use the understanding to improve cycle life of Li-ion and Li-S cells.

Reviewer 3

The reviewer said the team conducted research at a pouch level by adjusting pressure and processing temperature. This is an essential complement to the battery research in materials level.

Reviewer 4

The reviewer noted good approaches, but did not see anything novel.

Question 2: *Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.*

Reviewer 1

The two milestones completed in fiscal year (FY) 2022 are well presented and reflect actual progress in developing the Li-S battery. The applied pressure changes the porosity and tortuosity of the cathode and has clear effects on the cycling stability. Real-time pressure monitoring can be used as a tool to predict cell failure.

The reviewer asked the team how could the pressure be applied to the cell in the battery application, either to realize better performance or monitor the condition, without significantly increasing the cost? The reviewer also noted the team investigated the temperature effects in the formation cycle on the cycling performance of Li/NMC pouch cells. The results show that the room temperature (25°C) is the best condition.

Reviewer 2

The reviewer said the team developed good understanding on the impact of external pressure for Li/S cells and the impact of LHCE on SEI of Li metal rechargeable cells.

Reviewer 3

The reviewer noted that two milestones have been completed successfully, with another two are on track. The team studied the pressure effect on batteries, and investigated porosity and totuosity effects. The team explored the effect of localized high concentration electrolyte, along with the solvation driven morphology. The team published high quality of journal papers.

Reviewer 4

The reviewer said very modest pressures give higher Li density, as expected. Pressure reduces cracking in cathode, surprising. The reviewer noted that looking at separator constriction is new and important. Results are mainly empirical with little effort at fundamental understanding, perhaps appropriate for a development project.

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The team collaborates with all battery 500 PIs, students, and postdocs. It is better to indicate the contributions or roles of the collaborators to the specific project (Bat 368).

Reviewer 2

The reviewer noted that the research team works with universities and national labs. The technical approaches have been shared with other team members within the consortium for their batteries assessment.

Reviewer 3

The reviewer said Battery500 has very good collaboration throughout.

Reviewer 4

The reviewer noted that INL is part of the Battery500 team.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer said the team is going to refine the established methods, and also expand them to pouch cells, for Li/NMC and also Li-S.

Reviewer 2

The reviewer remarked focus on calendar life is highly important.

Reviewer 3

The reviewer remarked it is better to be more specific about future research plans, and asked will the team continue the pressure studies or some other methods are to be developed and adopted for evaluating the impact on battery cells of different applying conditions?

Reviewer 4

The reviewer said the team needs to provide insights on fixed pressure versus fixed gap on cycle life of Li metal cells.

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

The reviewer remarked research at a cell level is important in boosting the performance of the Li-S and Li-metal batteries.

Reviewer 2

The reviewer remarked the research is critical for advancing to practical cells.

Reviewer 3

The reviewer remarked the research is relevant to materials and batteries analysis. The results will also be relevant for electrification as they are valuable for batteries study.

Reviewer 4

The reviewer said the work met the Battery500 program objectives.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer said resources are adequate.

Reviewer 2

The reviewer said funding is sufficient.

Reviewer 3

The reviewer was not clear how many resources are utilized for the specific project (bat368). The overall funding level to the Battery500 is adequate.

Reviewer 4

The reviewer pointed out there are $15 million to distribute for the next stage. The refinement and expansion of the methods to other materials/batteries (pouch cell) will not cost too much for consumables. The reviewer noted manpower cost should be similar to that of last year.

Presentation Number: bat369
Presentation Title: High Energy Rechargeable Lithium-Metal Cells, Design, Fabrication and Testing
Principal Investigator: Jie Xiao, Pacific Northwest National Laboratory

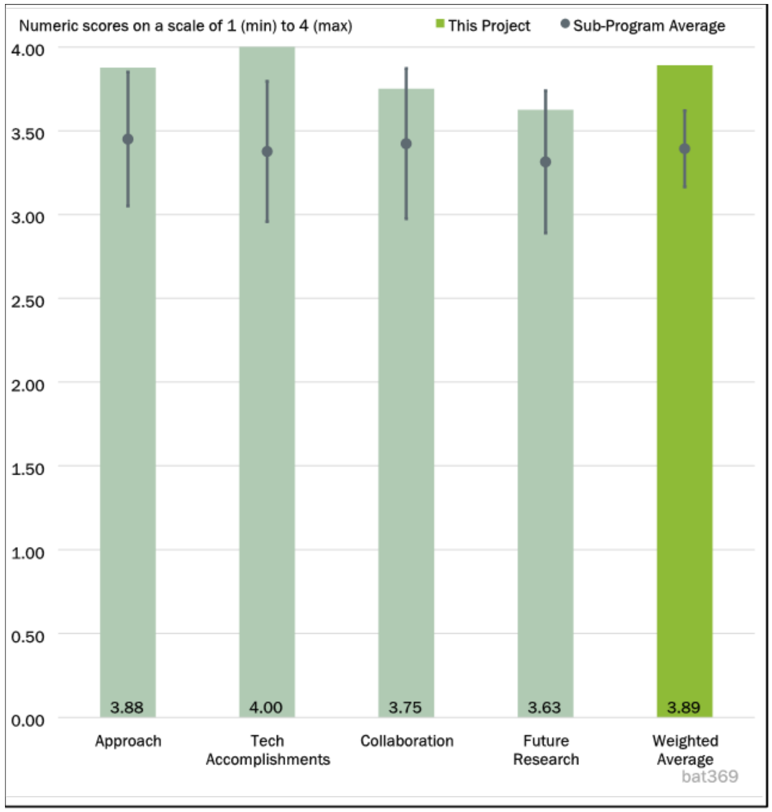

Figure 2-10 - Presentation Number: bat369 Presentation Title: High Energy Rechargeable Lithium-Metal Cells, Design, Fabrication and Testing Principal Investigator: Jie Xiao, Pacific Northwest National Laboratory

Presenter

Jie Xiao, PNNL

Reviewer Sample Size

A total of four reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 100% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 0% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

Question 1: Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?

Reviewer 1

The reviewer noted that PNNL as the leading team of the Battery500 consortium, gives a report on the overall approach and achievements of the project. The aim of the project is to develop high-energy Li-metal pouch cells up to 500 Wh/kg. The reviewer thought one of the important advantages of the approach is combining and focusing the strengths of nation-wide universities, national laboratories, and industrial partners to solve a very critical problem in batteries.

Reviewer 2

The reviewer noted that the team fabricated pouch cells with 350 Wh/kg capacity (2 Ah) using Li/NMC622 and achieved 600 stable cycles. The mechanism for the long stability and also the failure of the battery has been explored. The team published one paper in Nature Energy. Thin Li anode (20 micron) is found to be more effective than the thick one (greater than 50 microns). The reviewer said even higher capacity pouch cell at 400 Wh/kg (2.5 Ah) has been made and found to cycle greater than 200 times (on going).

Reviewer 3

The reviewer remarked developing design principles is extremely valuable. So is understanding dynamic interactions among cell components.

Reviewer 4

The reviewer commented the project approach is well designed and focused on achieving both cycling and high specific energy. The journal publications by the team that lay out their approach regarding high-energy pouch cells are important and impactful, and clearly move the project beyond the typical coin cell/single layer cell work of academic groups. The reviewer noted there is a strong quantitative road map already in place, which is very important for the approach.

The only concern this reviewer had with the approach is using a low charging rate of C/10. While this could work for some delivery vehicles, it is not expected to work for passenger vehicles, and the cycling stability optimization approach for a 1C or 2C charge rate is almost certainly different than a C/10 rate. The reviewer encouraged this effort to collect some data on 1C charge rate, to provide the community a sense for how the cells would do at that rate.

Question 2: *Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.*

Reviewer 1

The reviewer noted the research team has achieved more than the objectives originally planned. The other objectives are on track with excellent results demonstrated.

Reviewer 2

The reviewer said the consortium has made exciting progress in the well-planned project. The team has realized 350 Wh/kg pouch cell at 600 stable cycles in 2021 and is working on a 400 Wh/kg cell, which already reached 200 cycles.

The team presents an interesting study on the effect of Li-metal thickness on the cycling performance of Li-metal batteries. The reviewer said the dry/wet SEI explanation is plausible but not well supported by experiment and/or theory. One of the shortcomings of a thin (20 um) Li film is that it reduces the length of vertical channels in the Li anode and therefore increases the local current density.

Reviewer 3

The reviewer said results from the cell building efforts appear outstanding. Getting high-energy, multi-layer, Ah class pouch cells with Li-metal to cycle well is very difficult, and the results speak for themselves. There is a clear roadmap and testing pipeline, and the milestones are being reached. The reviewer said the team is clearly quantifying the influence of key variables (e.g., electrolyte and Li amounts) on cycling performance.

The reviewer offered as one small suggestion, the “dry” versus “wet” SEI hypothesis would benefit from greater empirical evidence. While the sketches are plausible, as the team likely knows there are other possible explanations, and additional work could be done to strengthen the explanation presented.

Reviewer 4

The reviewer said Phase 1 was amazingly successful, but way too much time was spent talking about Phase 1. As a result, the talk was much too long, substantially reducing chances for questions by judges. Explanation for why thicker Li gives poorer results is very insightful. The reviewer is eagerly awaiting experimental confirmation.

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer said the strength of the Battery500 consortium is in the collaboration crossover of the nationwide institutes with expertise in all different aspects of battery research and development.

Reviewer 2

The reviewer noted that Battery500 is a large team, with many institutions, and individuals with a range of priorities, which is very difficult to manage. However, it appears the team is functioning well together, and the connections among the efforts, including the cell building activity, are clear and appropriate. The reviewer noted that the cell building team has an important leadership role in the project because it is the ultimate metric to assess progress, and the leadership in this activity appears strong.

Reviewer 3

The reviewer noted the team works closely with industrial partners (eight), universities (nine) and national labs (three) in fabricating the pouch cell using NMC/Li for high energy density batteries.

Reviewer 4

The reviewer remarked Battery500 has very good collaboration throughout.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer remarked future research is well defined with the objective of realizing 500 Wh/Kg Li-metal cell. Because the second phase of the project just started, this reviewer is optimistic about the final success of the project. The tasks and contributions of each collaborating institutes are clearly indicated.

Reviewer 2

The reviewer said the team has done an excellent job articulating a roadmap for future work to get to 500 Wh/kg, and the proposed work fits into that roadmap well. The future targets are challenging, but at least for the coming year it seems likely they will be achieved.

Reviewer 3

The reviewer said the team is moving towards 500 Wh/kg cell in its future plan. This is ambitious and also achievable based on its current accomplished technical achievements.

Reviewer 4

The reviewer said improved utilization of metal is a very important insight that will impact how people think.

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

The reviewer said yes, the project supports the overall VTO objective very well.

Reviewer 2

The reviewer said the project is relevant to batteries, materials synthesis, and analysis, also eventually impact on electrification (EVs).

Reviewer 3

The reviewer said this is aimed at the heart of Battery500.

Reviewer 4

The reviewer remarked the project is clearly relevant. As mentioned above, this reviewer's main concern is the use of a C/10 charge rate, and how this large effort and optimization is done for this charge rate, while at least for passenger applications a rate of 1C or 2C (or higher!) is more relevant. Just collecting and presenting some data at higher charge rates would be very helpful for the community.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer noted that this is a large program, and that is appropriate, given the range of activities and the need to produce pouch cell results.

Reviewer 2

The reviewer said the funding level ($75 million for 5 years starting 10/1/2021) is adequate for the project.

Reviewer 3

The reviewer said resources are sufficient.

Reviewer 4

The reviewer noted there are $15 million for the consortium for the next stage. The reviewer believed industrial partners can also make some contributions.

Presentation Number: bat496
Presentation Title: Silicon Consortium Project: Advanced Characterization of Silicon Electrodes
Principal Investigator: Robert Kostecki, Lawrence Berkeley National Laboratory

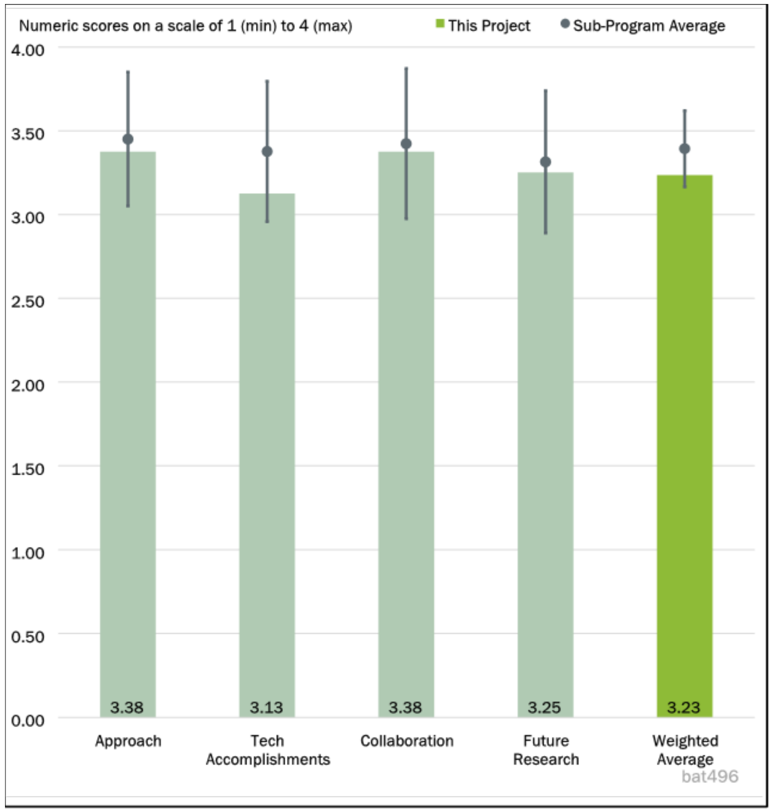

Figure 2-11 - Presentation Number: bat496 Presentation Title: Silicon Consortium Project: Advanced Characterization of Silicon Electrodes Principal Investigator: Robert Kostecki, Lawrence Berkeley National Laboratory

Presenter

Robert Kostecki, LBNL

Reviewer Sample Size

A total of four reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 100% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 0% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

Question 1: *Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?*

Reviewer 1

The reviewer said that yes, the project is well designed and the timeline is reasonably planned.

Reviewer 2

The reviewer mentioned that the approach taken by the team to study Si surface is very essential to the use of Si in Li ion batteries. The reviewer added that the approach is put together well to cover a wide range of options focusing on this problem from different directions.

Reviewer 3

The reviewer said that the project has a set of well-defined objectives and the tasks have been structured around those objectives.

Reviewer 4

The reviewer stated that the approach is quite comprehensive, utilizing a wide range of characterization techniques such as nano-Fourier-transform infrared spectroscopy (FTIR), neutron reflectometry, cryo-TEM/ electron energy loss spectroscopy, NMR, scanning spreading resistance microscopy, SEM- energy-dispersive X-ray spectroscopy, and Raman. The reviewer added that however, it is unclear whether the measurements were performed on the same Si baseline material. The reviewer added that it is also unclear whether some of these techniques, such as nano-FTIR and neutron reflectivity, could be applied to Si-composite electrodes which are non-planar. The reviewer declared that the influence of the polymeric binders on the composition and structure of the SEI and calendar life of the Si-composite electrodes should be investigated.

Question 2: *Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.*

Reviewer 1

The reviewer said that yes, it is as planned.

Reviewer 2

The reviewer suggested that it would be helpful to clearly delineate technical accomplishments made during this budget period as several results presented were published prior to the current budget period.

Reviewer 3

The reviewer claimed that there has been a great deal of effort placed to the development of analytical techniques, and that there has been terrific progress in this area. The reviewer noted that in parallel the techniques have generated a range of hypotheses for potentially observable phenomena, but little effort has been placed to date to pursue those observations. The reviewer concluded that this should be part of the future work and the reviewer looks forward to seeing this. The reviewer said that it will not only enhance this particular project’s accomplishments but support several of the other Silicon Consortia projects.

Reviewer 4

The reviewer stated that the activities done by the teams capture a variety of characterization techniques, but that the results are not progressing towards a common answer. The reviewer added that the solution from the activity is creating more problems that seem too complex to solve than progress towards a robust reliable solution.

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer claimed that there is outstanding collaboration between the various teams in the program.

Reviewer 2

The reviewer stated that the collaboration is excellent, but that it would be better to have some collaboration with industrial companies.

Reviewer 3

The reviewer noted that the collaboration is largely within the National Labs. The reviewer suggested that it may be helpful to extend the collaboration to companies that can make the best performing Si-electrodes in terms of calendar and cycle life. The reviewer added that it may also be helpful to extend the collaboration to universities that have some unique capabilities, such as mechanical characterization, and that it would be helpful to group the large number of collaborators to teams and highlight their individual contributions. The reviewer observed that it may be an oversight that one of the collaborators (Muhammad Ihsan Ul Haq) was listed twice on the "Collaboration and Coordination with other Institutions" page.

Reviewer 4

The reviewer said that it is clear that the methods developed here are being shared with the other Consortia members and that there is progress, as a result of this work. It appears to the reviewer that most of the other labs and teams of recipients of this project's work and don't necessarily directly contribute to the developments in this project. The reviewer suggested that the Principal Investigator should incorporate references to joint work on the individual slides.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer claimed that the project clearly defined a purpose for future work, and that it would be very interesting to learn more about the efficacy of the artificial SEI, i.e., Langumir-Blodgett surface film.

Reviewer 2

The reviewer stated that one of the items under Future Plan, "Explore and study range of silicon and silicon-carbon model systems materials...," should include studying the best performing Si-containing electrodes made by companies, especially those with DOE VTO funding.

Reviewer 3

The reviewer commented that proposed future work opens up several new paths towards an answer but that the program needs to focus on a solution that is reasonable for today's application while balancing the compromises needed to achieve the solution.

Reviewer 4

The reviewer explained that the score was a little lower than it could be, mainly because of the breadth of work that can come out of this project. The reviewer suggest that the PI may have to end up prioritizing the areas of focus. The reviewer added that the future work is otherwise highly relevant, when evaluated against the project's and consortium's goals.

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

The reviewer commented that Silicon is a promising anode candidate for high energy batteries, such as for EV.

Reviewer 2

The reviewer stated that it is highly relevant to the Battery subprogram.

Reviewer 3

The reviewer claimed that the project is very much relevant as Si-based anode will open the battery industry to achieve higher energy utilization as a complete product.

Reviewer 4

The reviewer noted that the analytical methods developed here do not need to apply strictly to silicon anodes, and for this reason this project is highly relevant.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer stated that the resources are amazing for characterizations.

Reviewer 2

The reviewer claimed that the team has excellent resources for advanced characterization of silicon electrodes.

Reviewer 3

The reviewer declared that resources allocated to the project are sufficient to achieve its stated goals.

Reviewer 4

The reviewer observed that the resources on this project are assumed to be those responsible in the development and validation of the test techniques, and then otherwise shared with joint projects. The reviewer concluded that, to that end, the project appears to be nicely balanced.

Presentation Number: bat497
Presentation Title: Silicon Consortium Project: Electrochemistry of Silicon Electrodes
Principal Investigator: Christopher Johnson, Argonne National Laboratory

Presenter

Christopher Johnson, ANL

Reviewer Sample Size

A total of four reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 100% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 0% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

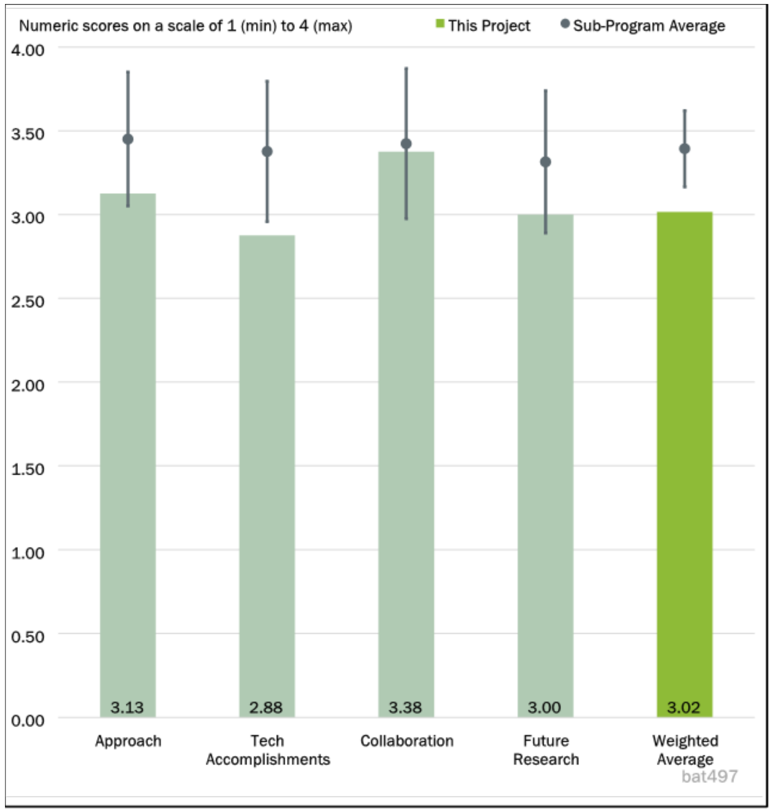

Figure 2-12 - Presentation Number: bat497 Presentation Title: Silicon Consortium Project: Electrochemistry of Silicon Electrodes Principal Investigator: Christopher Johnson, Argonne National Laboratory

Question 1: *Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?*

Reviewer 1

The reviewer commented that yes, the project is well designed, and the timeline is reasonably planned.

Reviewer 2

The reviewer expressed that the Voltage Hold method does not seem to be a robust quantitative method for evaluating the calendar life of full cells containing silicon in the negative electrode. The reviewer added that even under ideal conditions, it only provides qualitative information related to the calendar life. The reviewer suggested that the team should quickly decide whether this method should be dropped from the project and focus on developing a more quantitative calendar life evaluation method.

Reviewer 3

The reviewer remarked that the stated goal of identifying a technique to screen materials using a short-term test for the calendar life problem is very interesting to explore, but that the data shows that this process is more complex and the test is not going to be really short term. The reviewer noted that more understanding into the mechanism of failure in the voltage hold test will give better pathway towards defining a more realistic pass/fail criteria.

Reviewer 4

The reviewer declared that the majority of the effort has been based around voltage hold (Vhold) method development, validation of the method and validation of materials selection via the method. The reviewer added that the project is now moving to the other methods selected for electrochemical analysis. The reviewer observed that the timing appears relatively open-ended, and the reviewer is not convinced the timeline is being considered as relevant to the project.

Question 2: *Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.*

Reviewer 1

The reviewer expressed that it is as planned.

Reviewer 2

The reviewer stated that the Vhold approach has been well developed as a qualitative method, and that it can be extended to other work groups. The reviewer added that there were some other studies shown and these appear as incomplete at this point in time. The reviewer expressed that the scanning electrochemical microscopy method is an example of this and that it will be nice to see this development get fleshed out in the coming year. The reviewer articulated that the aged cells analysis was not presented in a manner as complete as expected so that the conclusions shown were acceptable, but that further discussion after the presentation with some of the investigators resolved the reviewer's concerns.

Reviewer 3

It seemed to the reviewer that the team spent much of its effort in demonstrating the ineffectiveness of the Voltage Hold method for evaluating the calendar life, though it could have been predicted since the calendar life should be evaluated under the open circuit condition, i.e., without an externally applied driving force, e.g., the applied potential.

Reviewer 4

The reviewer claimed that the validation process for developing this technique will take time, and that accelerating these processes causes new reactions that are creating a lot of noise in the study. The reviewer recommended that the focus of the work needs to be narrowed to study how the acceleration is affecting the anode and how it can be related to the mechanism in long term tests.

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer observed that there is outstanding collaboration between the various teams in the program.

Reviewer 2

The reviewer noted excellent collaboration and suggested that it may be better to have some industrial collaboration.

Reviewer 3

The reviewer declared that the collaboration is mainly among several National Labs.

Reviewer 4

The reviewer would like to see a little more engagement on evaluating different Si materials and electrode structures coming from the other labs. The reviewer clarified that this would really reinforce the electrochemical methods validation. The reviewer suspected that this will be the case as the scanning electrical mobility spectrometer method becomes employed by the other projects.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer noted that calendar life is critical and gassing on anode side is an enemy. The reviewer suggested that more work needs to be done on gassing mechanism analysis which may be very helpful to find effective additives for Si-based anode if there are any.

Reviewer 2

The reviewer declared that the team did not disclose much detail other than stating, under Proposed Future Research, "Develop electrochemical impedance methods on pouch cells to electrochemical impedance methods on pouch cells to measure Si equilibrium kinetics and its affect on calendar life." It was unclear to the reviewer how high frequency electrochemical impedance spectroscopy (EIS) could be helpful to evaluate the very slow (hundreds of hours) Si equilibrium kinetics.

Reviewer 3

The reviewer expressed that future work needs to focus on the relationship between the short-term test and long-term test.

Reviewer 4

The reviewer claimed that the PI indicated that more involvement with electrochemical modeling is needed, and this should be included the future work. The reviewer otherwise agreed with the proposed future work.

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

The reviewer stated that silicon is a promising anode candidate for high energy batteries, such as for EV.

Reviewer 2

The reviewer noted that the project supports the overall VTO objectives on EV batteries.

Reviewer 3

The reviewer claimed that the project is very much relevant as Si-based anode will open the battery industry to achieve higher energy utilization as a complete product. The reviewer added that developing a short-term test will reduce product development time and save cost for the battery industry.

Reviewer 4

The reviewer declared that the methods employed are very useful in directing materials selection and electrode characterization. The reviewer said that they can be used across the consortium and on other programs.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer mentioned that the team has excellent experimental and modeling capabilities.

Reviewer 2

The reviewer claimed that the resources are sufficient.

Reviewer 3

The reviewer observed that the resources allocated to the project are sufficient to achieve its stated goals.

Reviewer 4

The reviewer saw no issues with respect to the resources employed in this work.

Presentation Number: bat498
Presentation Title: Silicon Consortium Project: Next-Generation Materials for Silicon Anodes
Principal Investigator: Nathan Neale, National Renewable Energy Laboratory

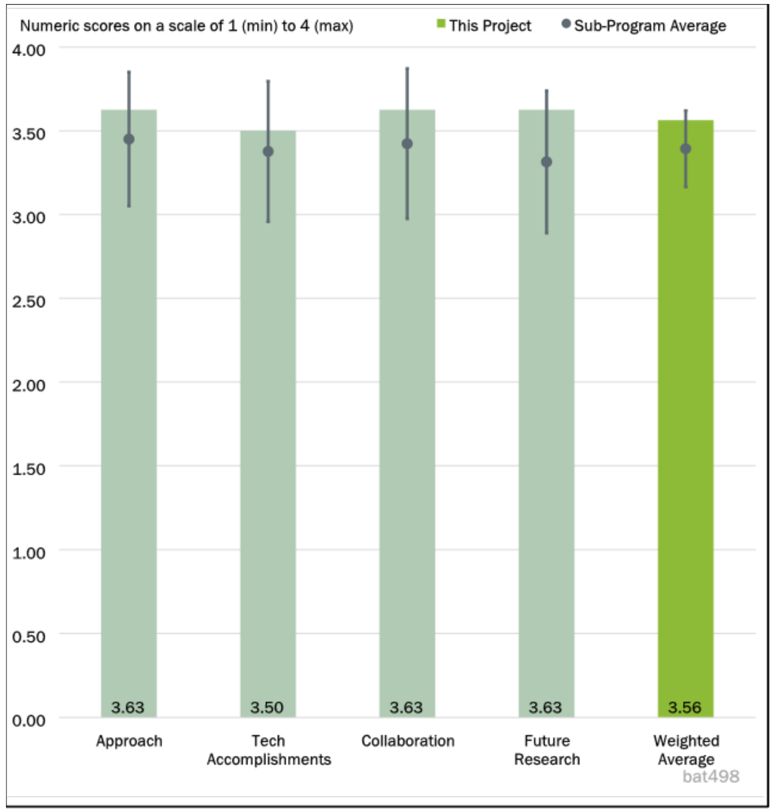

Figure 2-13 - Presentation Number: bat498 Presentation Title: Silicon Consortium Project: Next-Generation Materials for Silicon Anodes Principal Investigator: Nathan Neale, National Renewable Energy Laboratory

Presenter

Nathan Neale, NREL

Reviewer Sample Size

A total of four reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 100% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 0% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

Question 1: *Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?*

Reviewer 1

The reviewer noted that the project is well designed and the timeline is reasonable.

Reviewer 2

The reviewer commented that the approach to study different methods of Si manufacturing and how these techniques affect material property and thereby cycling/calendar aging is remarkable. The reviewer declared that the team's focus on working on techniques that have high yield and relatively low cost makes this project more relevant.

Reviewer 3

The reviewer observed that the work was diverse, covering a wide set of candidate materials. The reviewer added that the project has addressed several of the barriers and is now prioritizing the work.

Reviewer 4

The reviewer claimed that although Splat Quenching and Laser Quenching are among the known rapid quenching methods of forming amorphous alloys from the respective molten alloys, solid state reactions (e.g., annealing at a temperature which allows inter-diffusion but not crystallization) and mechanical alloying (e.g., ball milling) have emerged since the 1980s as alternative ways of making some amorphous alloys, including silicon-nickel (Si-Ni) and other Si-containing amorphous alloys. The reviewer suggested that the team may look up publications since the 1980s to evaluate whether these methods of forming amorphous alloys are more cost effective and scalable than the splat and laser quenching methods for making Si-containing amorphous alloys for Li-ion battery applications.

Question 2: *Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.*

Reviewer 1

The reviewer expressed that the success in understanding how each technique affects material surface is evident in the results presented. The reviewer concluded that it is encouraging to see that more than two techniques have yielded promising results.

Reviewer 2

The reviewer said it is as planned.

Reviewer 3

It was unclear to the reviewer why PEO works well for Si nanoparticles made by Plasma-enhanced chemical vapor deposition (PECVD), but PEO does not work for Si nanoparticles made by the milling process. The reviewer explained that milling single crystal ingots to form Si nanoparticles seems to be an expensive endeavor since it is already expensive to form these single crystal boules. The reviewer recommended that the team should perhaps mill metallurgical grade Si polycrystals to form low-cost Si nano-particles for Li-ion battery applications.

Reviewer 4

The reviewer stated that the initial approach was to find methods to produce meaningful amounts of suitable Si. The reviewer claimed that this has been achieved and the project has moved to electrode engineering. The reviewer added that it is known that the industry has made considerable progress in the development of high Si content anode materials and electrodes, and feedback from their lessons learned into progress here would be very valuable, if it is possible.

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer observed that there is outstanding collaboration between the various teams in the program.

Reviewer 2

The reviewer noted great collaborations and recommended that it would be better to have some collaboration with industry.

Reviewer 3

The reviewer suggested that the team may consider expanding collaborations to include experts on making Si-containing amorphous alloys by solid-state reaction or ball milling.

Reviewer 4

The reviewer remarked that the PI clearly recognizes the need for collaboration within the consortium and makes good use of it. The reviewer noted that it would be good if additional collaboration from industry would be possible, but this might dilute the efforts in meeting project scope. The reviewer clarified that it would be specifically great if a comparison on electrode fabrication could be made between the lab material and some commercially available high-Si content materials.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer remarked that studying electrolyte interaction is critical as each of these techniques creates new and different interactions. The reviewer added that developing 3D electrodes and strategically designed surfaces creates more excitement towards the future work. The reviewer concluded that the approach is very well thought off and executed by the team.

Reviewer 2

The reviewer noted the well prioritized tasks and good use of the work products from the other Consortium projects.

Reviewer 3

It seemed to the reviewer that the utilization of the Si is low (1000m Ah/g capacity vs.76%–80% Si in anode), which needs to be addressed.

Reviewer 4

The reviewer recommended that the team should figure out why PEO works well for Si nanoparticles made by PECVD, but does not work well for Si nanoparticles made by the milling process.

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

The reviewer claimed that Si is a promising anode candidate for high energy batteries, such as for EV.

Reviewer 2

The reviewer declared that the project is highly relevant to the Battery subprogram objectives.

Reviewer 3

The reviewer expressed that the project is very much relevant as Si-based anode will open the battery industry to achieve higher energy utilization as a complete product.

Reviewer 4

The reviewer stated that this project is central to achieving the Consortium goals, and can expand and contract effectively with Consortium scope prioritization and scope adjustments as it is sufficiently agile.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer stated that the resources are sufficient.

Reviewer 2

The reviewer noted that the resources allocated to the project are sufficient to achieve its stated goals.

Reviewer 3

The reviewer agreed with the PI's statement indicating that the project is most successful with the appropriate level of collaborative support.

Reviewer 4

The reviewer said that although the team has sufficient resources, it may consider collaborations with experts on amorphous alloys and mechanical characterizations.

Presentation Number: bat499
Presentation Title: Silicon Consortium Project: Mechanical Properties of Silicon Anodes
Principal Investigator: Katherine Harrison, Sandia National Laboratories

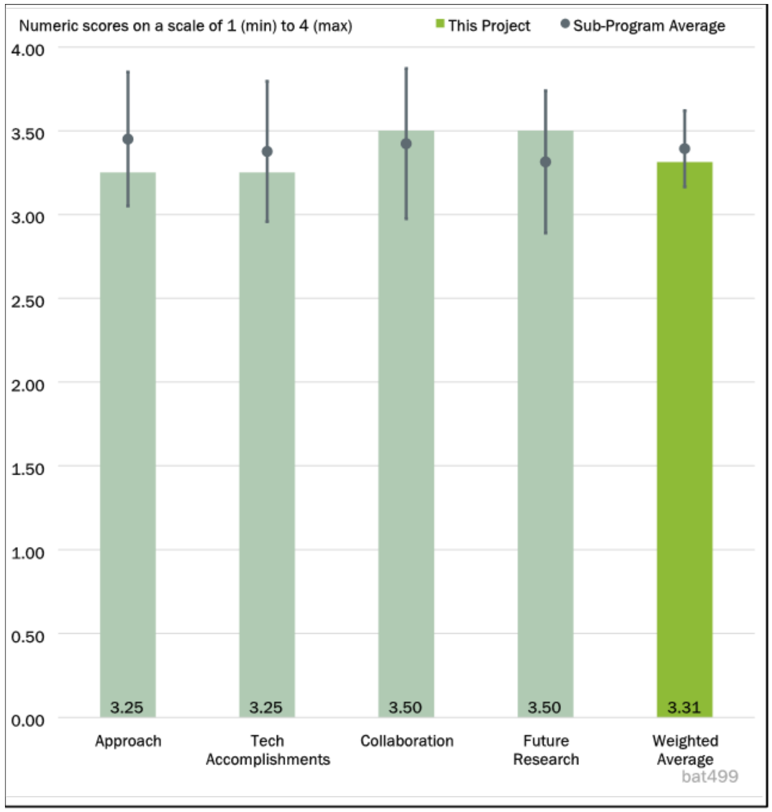

Figure 2-14 - Presentation Number: bat499 Presentation Title: Silicon Consortium Project: Mechanical Properties of Silicon Anodes Principal Investigator: Katherine Harrison, Sandia National Laboratories

Presenter

Katherine Harrison, SNL

Reviewer Sample Size

A total of four reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 100% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 0% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

Question 1: *Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?*

Reviewer 1

The reviewer stated that the project is well designed and the timeline is reasonable.

Reviewer 2

The reviewer claimed that the approach to study the mechanical property of Si anodes is critical towards commercialization.

Reviewer 3

The reviewer expressed that the approach taken by the mechanical characterization thrust is comprehensive. The reviewer observed that some of the characterization methods, however, seem to be more useful than others. The reviewer clarified that for example, microcalorimetry results were inconsistent with the electrochemical test results, probably because microcalorimetry was not done under realistic cell operating conditions.

Reviewer 4

The reviewer commented that the structure of the work is appropriate for the project objectives. The reviewer added that the work is also well prioritized. What was not as clear to the reviewer was how this work overlaps with the electrode structure development work in the adjacent projects. The reviewer clarified that this is important to note since this project does not just address mechanical characterization of the electrode but also steers electrode design.

Question 2: *Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.*

Reviewer 1

The reviewer observed that the team is focusing on the techniques that work and has promising results which has streamlined the project and helped narrow the task. The reviewer stated that the mechanical perturbation is an interesting approach.

Reviewer 2

The reviewer said it is as planned.

Reviewer 3

The reviewer declared that the results highlight the importance of mechanical degradation. The reviewer added that the more detailed characterization of the mechanical properties, at various stages of calendar aging and cycle number, would be necessary to understand degradation mechanisms.

Reviewer 4

The reviewer remarked that progress has been good, but that it is also clear that additional electrode structural optimization is needed. The reviewer noted that this includes the areas of other binder candidates, which will play an increasing role in the higher loading electrodes.

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer noted that there is outstanding collaboration between the various teams in the program.

Reviewer 2

The reviewer noted the excellent collaborations and said that it would be better if the team included industrial collaboration as well.

Reviewer 3

The reviewer mentioned that collaborations are mainly among the National Labs, and that it may be helpful to expand collaborations to include experts on characterization of mechanical behavior of Si-electrodes to help solve mechanical damages especially in high loading Si cells.

Reviewer 4

The reviewer stated that the team collaborates well with other project teams. The reviewer added that it might be helpful to consult with industrial materials suppliers for additional experience in materials selection and evaluations. The reviewer concluded that this is potentially most beneficial in the binder system, but could also extend to the selection of current collector materials (e.g.: copper/nickel composites) and conductive additives.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer stated that future work is clearly defined, and that large format cells (i.e., 3-5Ah) are recommended for study on gas generation and stability. The reviewer clarified that in coin cells, it is not possible to observe gas generation.

Reviewer 2

The reviewer said that hopefully, more insights can be gained to help understand the degradation mechanisms caused by the many parameters identified by the team, such as, Si material, electrode manufacturing conditions, electrochemical aging/cycling protocols, pouch versus coin cells, and size and loading of the electrodes.

Reviewer 3

The reviewer noted that the team's focus on understanding the influence of mechanical effect in the life of Si anode is very well reflected in the future work.

Reviewer 4

The reviewer observed a very good choice of future thrust areas, and added that this could be helped by collaboration on optimizing the non-active materials components.

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

The reviewer remarked that the success in this project is essential in moving the consortium ahead. The reviewer added that it will also provide the platform to evaluate alternative Si-containing active materials. The reviewer exclaimed that the team did good work.

Reviewer 2

The reviewer claimed that Si is a very promising anode candidate for high energy batteries, such as for EVs.

Reviewer 3

The reviewer affirmed that the project supports the VTO battery objectives.

Reviewer 4

The reviewer declared that the project is very much relevant as Si based anode will open the battery industry to achieve higher energy utilization as a complete product.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer claimed that the resources are sufficient.

Reviewer 2

The reviewer noted that the resources allocated to the project are sufficient to achieve its stated goals.

Reviewer 3

The reviewer observed no issues and good collaboration with adjacent teams.

Reviewer 4

The reviewer suggested that it may be helpful to expand external collaborations to strengthen mechanical characterization. The reviewer added that this may help determine which of the many hypothetical parameters are important in affecting the calendar and cycle life of Si-containing electrodes.

Presentation Number: bat500
Presentation Title: Silicon Consortium Project: Science of Manufacturing for Silicon Anodes
Principal Investigator: Gabriel Veith, Oak Ridge National Laboratory

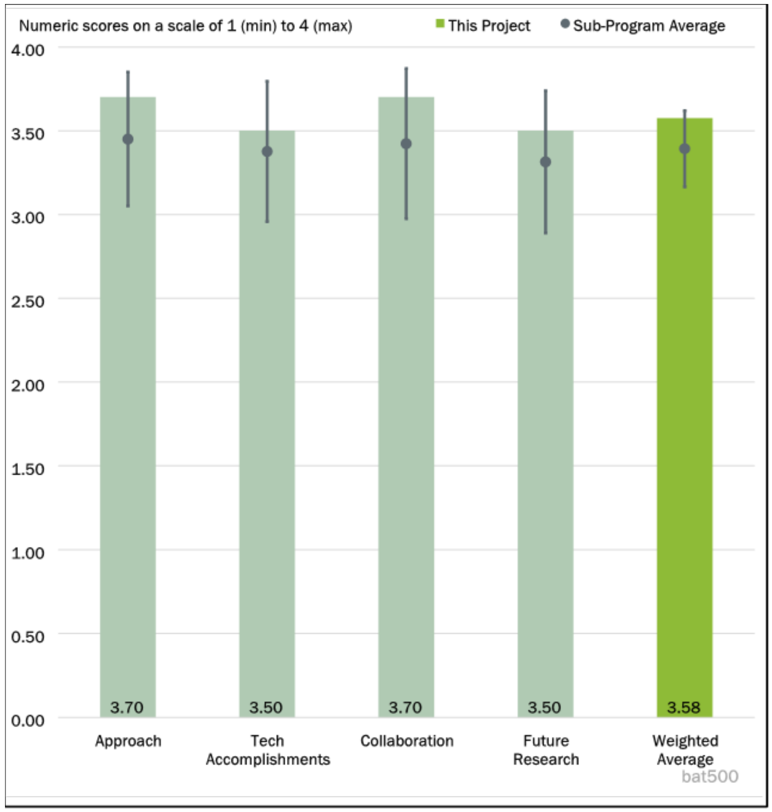

Figure 2-15 - Presentation Number: bat500 Presentation Title: Silicon Consortium Project: Science of Manufacturing for Silicon Anodes Principal Investigator: Gabriel Veith, Oak Ridge National Laboratory

Presenter

Gabriel Veith, ORNL

Reviewer Sample Size

A total of five reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 80% of reviewers felt that the resources were sufficient, 20% of reviewers felt that the resources were insufficient, 0% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

Question 1: *Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?*

Reviewer 1

The reviewer noted that the focus on reproducibility and process control is especially critical to the development of high-capacity thick Si electrodes for automotive applications.

Reviewer 2

The reviewer remarked that the project is well-designed and the timeline is reasonable.

Reviewer 3

The reviewer declared that the team’s approach to understanding electrode manufacturing issues is excellent. The reviewer added that the methods used to eliminate noise in development and testing are very well thought off and executed.

Reviewer 4

The reviewer commented that the work has addressed several important issues related to Si-based anode materials such as low initial coulombic efficiency, low loading electrode, poor cycle life, etc. The reviewer added that the approaches taken seemed reasonable and effective.

Reviewer 5

The reviewer observed that although the 200-600g per batch rate may be sufficient for producing enough Si electrode materials for the project team members, it is unclear whether manufacturing science fundamentals and knowledge gained at this batch level could be scaled to the eventual industrial-scale production of Si-containing electrodes for EV applications. The reviewer suggested that the team may develop and test scaling relationships for manufacturing Si electrodes over a wide range of batch sizes. The reviewer added that since evaporation is a line-of-sight deposition process, pre-lithiation of thick Si-composite electrodes by the evaporation method may be problematic since Li can only be deposited on the surface of the electrode, but not throughout the depth of the electrode.

Question 2: *Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.*

Reviewer 1

The reviewer remarked that both the calendar and cycle life performance are still under target. The reviewer recommended more understanding on the degradation mechanisms.

Reviewer 2

The reviewer observed that it is as planned.

Reviewer 3

The reviewer claimed that the generating of materials reproducibly using a scalable process is an important milestone achievement. The reviewer suggested that more work may be needed to develop the binder, where the use of bitumen may be problematic without tighter property specifications. The reviewer added that understanding electrode surface phenomena, including the surface wettability, may present significant opportunities with regard to the electrode performance, particularly at high load. The reviewer noted that this may be an area where a focused study of trace additives may be useful.

Reviewer 4

The reviewer said that milling single crystal ingots to form Si nanoparticles seems to be an expensive route since it is already expensive to form these single crystal boules. The reviewer recommended that the team should perhaps mill low-cost Si polycrystals to form Si nano-particles for Li-ion battery applications.

Reviewer 5

The reviewer expressed that the results show good progress into understanding the failure mechanism in high energy anodes. The reviewer added that scalability is achieved but the performance optimization is the critical next step. The reviewer noted that the team approach to understanding the mechanism in each different manufacturing process is critical in understanding the effect of electrode manufacturing in anode performance.

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer observed that there is outstanding collaboration between the various teams in the program.

Reviewer 2

The reviewer mentioned that the collaboration between several national labs and universities seems to be efficient.

Reviewer 3

The reviewer commented that the collaboration across EERE and universities is outstanding, but that more industry engagement is desirable for technology transfer.

Reviewer 4

The reviewer stated that there is great collaboration among national labs, but that it would be very beneficial to have some industrial collaborations.

Reviewer 5

The reviewer remarked that the collaborations are mainly within several National Labs. The reviewer suggested that expanding collaborations to include experts on mechanical characterization may be helpful to the overall project goals, such as high loading Si-electrodes.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer claimed that future research will be helpful to the broader research community on a fundamental level. The reviewer added that steady progress toward the targets for electrode cycle life and capacity indicates a high probability of success.

Reviewer 2

The reviewer remarked that future work planned in this project is critical towards this process being commercialized.

Reviewer 3

The reviewer mentioned that processing (mixing/coating) is very critical for good electrode performance, especially for thick electrodes, and it should be the main focus. The cell density of 400Wh/kg for Silicon cells seems too high for the reviewer and needs to be justified.

Reviewer 4

The reviewer observed that overcoming mechanical degradation may be challenging.

Reviewer 5

The reviewer declared that a more detailed plan should be described.

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

The reviewer noted that this work is of fundamental relevance to the battery program, as Si electrodes will be an important development in commercialization of automotive cells.

Reviewer 2

The reviewer remarked that Si is a very promising anode candidate for high energy batteries, such as for EVs.

Reviewer 3

The reviewer expressed that the project supports the VTO battery objectives.

Reviewer 4

The reviewer observed that the project is very much relevant as Si based anode will open the battery industry to achieve higher energy utilization as a complete product.

Reviewer 5

The reviewer claimed that the program does support the VTO subprogram objects, and that it also addressed the issues in a promising way.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer claimed the resources are sufficient.

Reviewer 2

The reviewer commented that the resources allocated to the project are sufficient to achieve its stated goals.

Reviewer 3

The reviewer made no comments.

Reviewer 4

The reviewer declared that although the team has sufficient resources, it may consider collaborations with experts on mechanical property characterizations.

Reviewer 5

The reviewer stated that while progress toward the project goals is outstanding, more resources are needed in this area for a broader impact on manufacturability of advanced electrode materials.

Presentation Number: bat501
Presentation Title: Integrated Modeling and Machine Learning of Solid-Electrolyte Interface Reactions of the Silicon Anode
Principal Investigator: Kristin Persson, Lawrence Berkeley National Laboratory

Presenter

Kristin Persson, LBNL

Reviewer Sample Size

A total of four reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 100% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 0% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

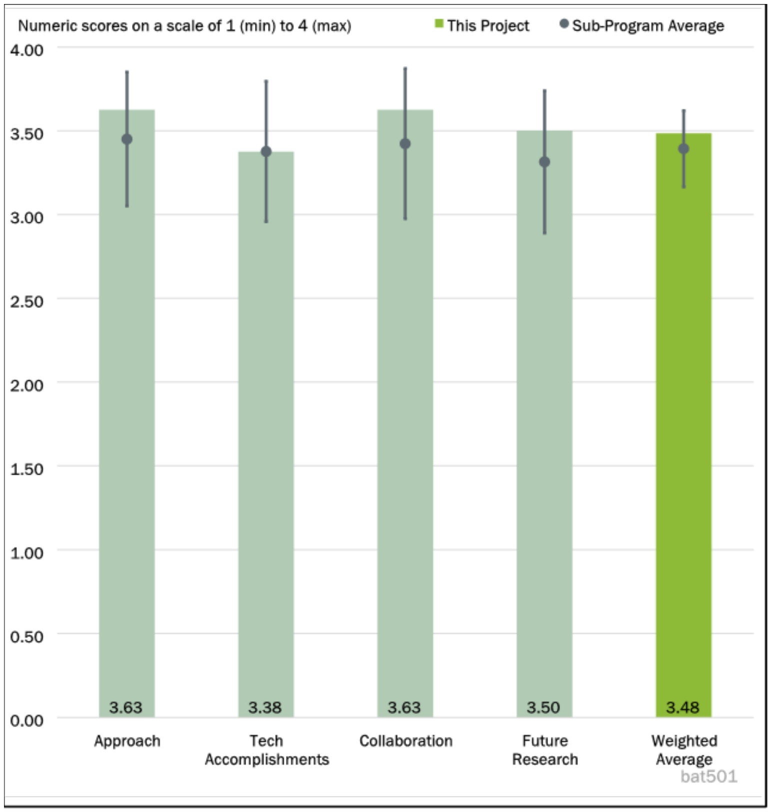

Figure 2-16 - Presentation Number: bat501 Presentation Title: Integrated Modeling and Machine Learning of Solid-Electrolyte Interface Reactions of the Silicon Anode Principal Investigator: Kristin Persson, Lawrence Berkeley National Laboratory

Question 1: *Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?*

Reviewer 1

The reviewer noted that the project is well designed and the timeline is reasonable.

Reviewer 2

The reviewer mentioned that the approach seems to be comprehensive, bridging multiple length and time scales to simulate SEI dynamics.

Reviewer 3

The reviewer claimed that the atomistic simulation to understand the mechanism of SEI formation on Si anode is critical to develop the material for commercial application.

Reviewer 4

The reviewer remarked that the modeling work provides a fundamental understanding about the SEI formation which is one of the most important degradation factors for Si anodes.

Question 2: *Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.*

Reviewer 1

The reviewer stated that the accomplishments demonstrated by the team show the various approaches taken by the team to understand the mechanism.

Reviewer 2

The reviewer said it is as planned.

Reviewer 3

The reviewer noted that the team is able to model SEI growth without any fitting parameters, capture depth-dependent SEI species, and validate voltage-hold experiments. The reviewer hoped that specific predictions from the multiscale modeling effect will soon help guide the overall project to achieve the required calendar and cycle life of Si-electrodes.

Reviewer 4

The reviewer expressed that some interesting findings were revealed through the modeling work. The reviewer added that the correlation between the failure mechanism and findings is still required.

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer noted the excellent collaborations.

Reviewer 2

The reviewer remarked that the National Labs and university collaborations seem effective.

Reviewer 3

The reviewer observed outstanding collaboration between the various teams in the program.

Reviewer 4

The reviewer commented that great team work has been demonstrated to put all the understanding together.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer commented that if great team work continues, clearer understanding about Si SEI is more likely to be achieved.

Reviewer 2

The reviewer noted that future work is clearly defined, and that the team is very capable on characterization. The reviewer asked if the team planned to validate the reactions/mechanisms found in simulation by experiments.

Reviewer 3

The reviewer stated that in addition to helping understand what is known about the SEI, the team may aim at making predictions based on the validated models to help develop more stable SEIs.

Reviewer 4

The reviewer mentioned that more future work is needed in building the relationship between the calendar aging mechanism and the science behind the failure modes.

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

The reviewer remarked that Si is a very promising anode candidate for high energy batteries, such as for EVs.

Reviewer 2

The reviewer noted that the project supports the VTO battery objectives.

Reviewer 3

The reviewer expressed that the project is very much relevant as Si-based anode will open the battery industry to achieve higher energy utilization as a complete product.

Reviewer 4

The reviewer said yes, the project supports the VTO subprogram objectives.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer stated that the resources are sufficient.

Reviewer 2

The reviewer said that the team seems to have sufficient resources.

Reviewer 3

The reviewer remarked that the resources allocated to the project are sufficient to achieve its stated goals.

Reviewer 4

The reviewer made no comments.

Presentation Number: bat523
Presentation Title: Development of Long Life Lithium and sulfurized polyacrylonitrile (SPAN) Cells
Principal Investigator: Ping Liu, University of California-San Diego

Presenter

Ping Liu, University of California-San Diego

Reviewer Sample Size

A total of five reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 80% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 20% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

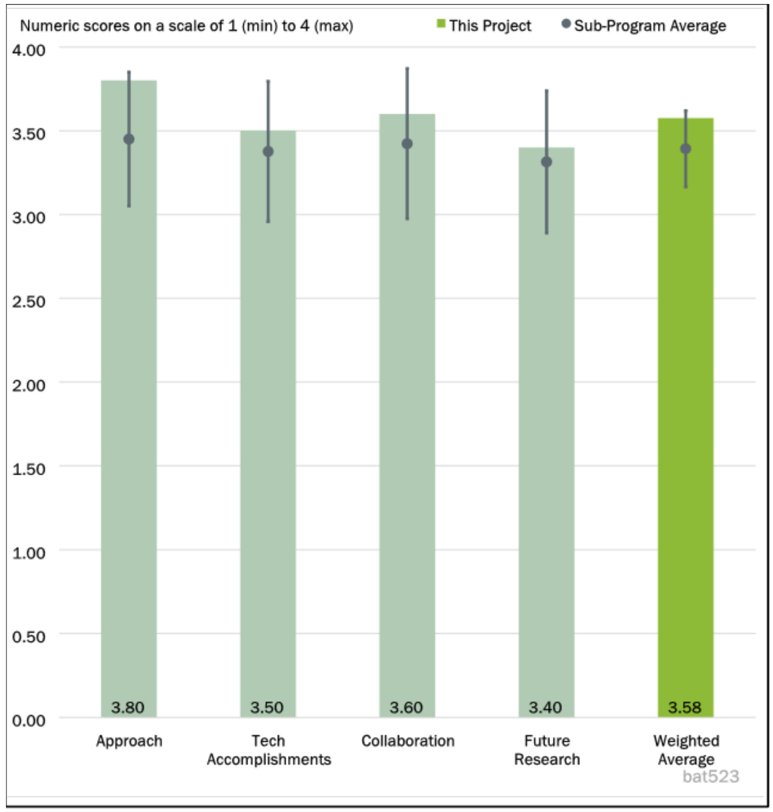

Figure 2-17 - Presentation Number: bat523 Presentation Title: Development of Long Life Lithium and sulfurized polyacrylonitrile (SPAN) Cells Principal Investigator: Ping Liu, University of California-San Diego

Question 1: *Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?*

Reviewer 1

The reviewer expressed that Dr. Liu and team perform world class research.

Reviewer 2

The reviewer stated that the roadmap of this research showed that the SPAN cathode is promising but further improvement in capacity is needed. The reviewer added that the project is, so far, well designed when it comes to the fundamental understanding of the research and the timeline is reasonably planned.

Reviewer 3

The reviewer stated that the project addresses the two key technical barriers of current EV batteries, which are low specific energy and high cost; improvements in these two categories are the objectives of the Battery500 program, to which the current project belongs. The reviewer added that lithium-sulfur chemistry is in principle well suited to meet these performance targets but is deterred by the enormous challenge from the polysulfide intermediates that dissolve in most of the organic electrolytes. The reviewer noted that while another project is aiming to mitigate the polysulfide challenge, this project is developing a stable, low-cost alternative to S-C composites based on sulfurized polymer, especially sulfurized-polyacrylonitrile (SPAN). The reviewer remarked that this material has been well studied in literature and has been shown to cycle well but with specific capacity (1.5 mAh/cm^2). The reviewer expressed that accordingly, the targeted specific energy is lowered to 300 Wh/kg. The reviewer concluded that there is noticeable compromise in performance vs. the Battery500 target. The reviewer said that specific energy demonstrated in Phase I is below 300 Wh/kg (265 Wh/kg), but good cycle life was demonstrated with a new electrolyte. The reviewer commented that this year's effort has focused on the first cycle irreversible transformation for the SPAN cathode with much lower discharge voltage. The reviewer affirmed that overall, the approach looks reasonable but only partly contributes to overcoming the barriers of Li-S chemistry, unless the capacity is improved with new polymers. The reviewer listed two weaknesses as being: i) Even though the cycle life is improved significantly compared to conventional Li-S cells, the specific capacity of the cathode and hence the specific energy of the cells are significantly lower. The specific energy realized in 2 Ah cells was only 265 Wh/kg, just about the same as the current Li-ion cells. ii) It is difficult to improve the capacity with new sulfurized polymers; this project should have been the topic of study, probably more than the first cycle irreversible transformation of SPAN.

Reviewer 4

The reviewer noted that LiS rechargeable battery is a very challenging chemistry to realize. The reviewer said that however, there is a huge commercial potential for this chemistry to be used not only for EV, but also for stationary energy storage and aviation energy storage. The reviewer explained that in particular, the sulfur cathode materials dissolution in the electrolyte during electrochemical process is a critical technical barrier to be solved. The reviewer added that the proposed work is focused on preventing/managing polysulfides dissolution, the major technical barrier for LiS battery chemistry. The reviewer expressed that the project develops LiSPAN composite structures to manage polysulfides dissolution during the electrochemical process. The reviewer observed that the project objectives include developing next generation high-energy, low-cost batteries for electric vehicles, designing, fabricating and validating high energy pouch cells up to 500 Wh/kg, scaling up pouch cell capacity to 5-10 Ah, demonstrating long cycle life of up to 1,000 deep charge-discharge cycles, and achieving total control of battery chemistries for robust, scalable and commercially viable cells based on the LiSPAN LiS technologies. The reviewer claimed that SPAN is proposed as a stable, low-cost alternative to S-C composites, and that although Roadmap shows SPAN's promise but further improvement in capacity is needed. The reviewer stated that as of today, developing synthetic approaches for SPAN and alternate polymers is being explored by the PI's team. The reviewer declared that the SPAN composite approach to manage polysulfide dissolution has demonstrated initial success supported by earlier literature and research works. The reviewer concluded that the project is well designed and the timeline to achieve the program goal is reasonable.

Reviewer 5

The reviewer said that the PIs tried to understand the mechanism of the redox reaction of SPAN, which is an active area of research for the polymeric sulfur cathode. The reviewer claimed that one of the barriers of Li-S battery is the so-called shuttle effect which resulted from the high dissolution of polysulfide ions. The reviewer added that the polymeric sulfur materials like SPAN can restrain the sulfide on the polymer backbone. The reviewer expressed that by means of both electrochemical cycling and advanced analytical tools, the PIs proposed a mechanism of SPAN redox reaction. The reviewer recommended that the PIs should focus on if the proposed mechanism can explain the higher discharge capacity than the theoretical capacity based on sulfur, i.e., something else along with sulfur contributing to charges.

Question 2: Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.

Reviewer 1

The reviewer remarked that excellent progress has been made in the demonstration of laboratory scale synthesis of SPAN material and its performance of 265 Wh/kg in 2 Ah pouch with new electrolyte. The reviewer was not sure if its cycle life is as long as shown in the top-left figure on the slide. The reviewer added that during this review period, the first cycle life irreversible transformation was addressed using several techniques. The reviewer stated that the results indicate that the cathode (SPAN) loses both hydrogen and sulfur from non-aromatic cleavages contributing capacity loss and the C=S bonds are irreversibly transformed but contribute to reversible capacity subsequently. The reviewer remarked that interestingly, SPAN becomes more conducting with improved conjugation, which explains the increase in the discharge voltage. The reviewer claimed that these studies may help design new sulfurized polymers with higher capacities. It was not clear to the reviewer how Li anode performed as well for that many cycles. the reviewer asked if it is a protected Li anode. The reviewer questioned how much excess Li was used here (N/P). The reviewer recommended that there should have been some effort to identify new higher-capacity sulfurized polymers. The reviewer asked if the sulfur content can be increased at all. The reviewer noted that some design study is needed to verify if high specific energies beyond Li-ion batteries are possible with the SPAN-like cathodes having low capacity, lower cell voltage and poor conductivity (requiring higher proportion of conductive diluent).

Reviewer 2

The reviewer observed very good progress in reducing the ICL of these promising SPAN materials. The reviewer added that the 2Ah pouch cell at 265Wh/kg is very promising. The reviewer noted that although we expect Li/S cells to be much higher, the goal for commercialization is likely going to be “as good or better” as the cost advantage should be very high.

Reviewer 3

The reviewer noted that this project started in late 2021, and that the PI’s team has successfully identified first cycle mechanism loss of hydrogen and S by the formation of H_2S, limited sulfur separation in the materials, the presence of C-S bond in non-aromatic structures, and irreversible loss of C-S and formation of S_2- during the first cycle. The reviewer remarked that considering the project is in its initial stage of 13% in progress, the progresses made by the PI’s team have been excellent.

Reviewer 4

The reviewer expressed that the PIs reported a 1 Ah pouch cell based on SPAN cathode, and that the PIs reported that the difference of the first cycle of a SPAN cathode was due to the formation of H_2S from the gap S. The reviewer added that limited S separation after the first cycle was also reported. The reviewer declared that the major accomplishment was the new electrolyte which can improve the performance of SPAN. The reviewer recommended that the PIs check the S distribution after multi-cycles, and take into consideration if the H_2S formation was from gap S or just the residual elemental S remaining during the synthesis.

Reviewer 5

The reviewer commented that the research group performed a comprehensive study to understand the first cycle irreversible transformation for the SPAN cathode. The reviewer listed that they found a loss of hydrogen and sulfur in the form of H_2S; the SPAN cathode becoming more conducting with improved conjugation; and the C=S bond irreversibly transformed but subsequently contributing to reversible capacity. The reviewer added that the understandings achieved are expected to help the follow-on studies to further improve the SPAN as a cathode material for Li-S batteries. The reviewer suggested that the group, however, should investigate the reproducibility of these materials and cells.

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer observed no issues.

Reviewer 2

The reviewer claimed that this is a Battery500 project with broad collaboration with national labs, universities, and industry. The reviewer added that in particular, the PI is working with INL on SPA synthesis transferring, PNNL and University of Maryland, Pennsylvania State, and University Pittsburgh on supplying standard SPAN electrode, Stanford University on evaluating electrolyte, Brookhaven National Laboratory (BNL) on the mechanism study, and Texas A&M on computational study.

Reviewer 3

The reviewer expressed that as part of the Battery500 Consortium, the PIs were well integrated into the team of PIs which include those from national labs and industries.

Reviewer 4

The reviewer commented that the research has been deployed in close collaboration with national labs toward the fundamental understandings, but not with industries yet. The reviewer recommended that industry collaboration is needed in the future.

Reviewer 5

The reviewer stated that there are several ongoing collaborations with the DOE Battery500 team members, which are well detailed. The reviewer clarified that, for example, they collaborated with Idaho National Laboratory for SPAN synthesis scale up, Pacific Northwest National Laboratory and University of Maryland, Pennsylvania State University and University of Pittsburgh for the SPAN electrodes, Stanford University for evaluating electrolytes, BNL for mechanistic studies using in-situ XRD and PDF, and Texas A&M University for computational study of SPAN structures. The reviewer recommended that more active collaboration with GM and possibly an industrial partner (Li-S company) will be beneficial to demonstrate the SPAN comprehensively in more representative prototype cells.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer claimed that even though the preliminary results are encouraging from a cycle life perspective, there is a serious question on how to raise S loading without affecting the reversibility (cycle life) and how to ensure good cycle life with a Li anode at high current densities required against dense cathodes. The reviewer added that the proposed future study is aimed to address these questions. The reviewer explained that specifically, the proposed study will focus on how to tune chemical synthesis conditions, raise reversible S content by compositing SPAN and S and identify compatible electrolytes especially for Li/SPAN cells with lean electrolyte.

Reviewer 2

The reviewer believed this team is considering a SPAN/S composite cathode. The reviewer observed that the issue is that the two function best with different electrolyte classes. The reviewer did not see cell Wh/kg as necessarily a showstopper here and it seemed like SPAN might be good enough. The reviewer recommended that if it is not, an investigation into re-engineering the SPAN material to enhance the S content might be valuable.

Reviewer 3

The reviewer declared that the project is centered at using SPAN composites to confine/manage polysulfides dissolution during the electrochemical operation of the sulfur cathode electrode. The reviewer noted that future work is focused on fully understanding the mechanism of the SPAN materials during the electrochemical process, and developing SPAN materials composition to evaluate the effectiveness of polysulfides management by this class of composite materials. The reviewer articulated that future work also investigates the impact of the materials to the battery performance such as loading and impacts to lithium metal electrode.

Reviewer 4

The reviewer stated that the PIs clearly defined the future work, which was based on the current achievements and in alliance with the overall Battery500 goals. The reviewer added that in the literature, it has been widely reported that there is an optimal S loading in a SPAN, raising S loading beyond the value will have a negative impact on the SPAN cathode. The reviewer asked the team to address how raising S loading can increase capacity while maintaining reversibility.

Reviewer 5

The reviewer stated that yes, the purpose of future work has been clearly defined. The reviewer added that the detailed approaches are not provided. The reviewer asked what approaches will be taken to reduce the capacity loss, and what structures of the SPAN electrode will be developed.

Question 5: Relevance: Does the project support the overall VTO subprogram objectives?

Reviewer 1

The reviewer stated that the project supports the overall DOE objectives by overcoming the deterrents of low specific energy and high cost of current Li-ion cells, with a new system involving Li anode and SPAN cathode. The reviewer added that even though the benefits are not significant to approach 500 Wh/kg, there has been a marginal increase in specific energy, which can be further improved with new S-polymer. The reviewer observed that the stated goals of demonstrating high specific energy (goal: 500 Wh/kg) and long cycle life (1000 cycles) in pouch cells of 5-10 Ah are consistent with the DOE goals. The reviewer concluded that, overall, this project is quite relevant to the DOE VTO battery program objectives and goals.

Reviewer 2

The reviewer declared that it is highly relevant given the abundance and affordability of sulfur, and that this is critical research for enablement of wide scale EV adoption.

Reviewer 3

The reviewer expressed that this project is highly relevant to the battery storage subprogram, as the success of this project leads to significant reduction of both battery cost and the use of critical materials.

Reviewer 4

The reviewer remarked that Li-S batteries are considered as one of the most promising chemistries to replace Li-ion batteries in transportation. The reviewer added that investigating polymeric sulfur cathode supports the overall VTO objectives.

Reviewer 5

The reviewer said that yes, the project supports the overall VTO objective in battery analysis, energy density enhancement, and materials development.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer remarked that resources for the overall Battery500 project are commensurate with the scope adequate to achieve the targeted milestones. The reviewer added that the budget details for this specific project haven't been provided.

Reviewer 2

The reviewer observed no issues.

Reviewer 3

The reviewer declared that the resources and project timeline are well aligned.

Reviewer 4

The reviewer stated that Battery500 includes national labs and multiple research universities, and that it can provide more than adequate resources to the proposed research.

Reviewer 5

The reviewer expressed that the research group has necessary facilities and equipment/instrumentation at UCSD. and that the external collaborators listed are supposed to provide necessary support.

Presentation Number: bat524
Presentation Title: Advanced Electrolytes for Lithium Metal Batteries
Principal Investigator: Chunsheng Wang, University of Maryland

Presenter

Chunsheng Wang, University of Maryland

Reviewer Sample Size

A total of four reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 50% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 50% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

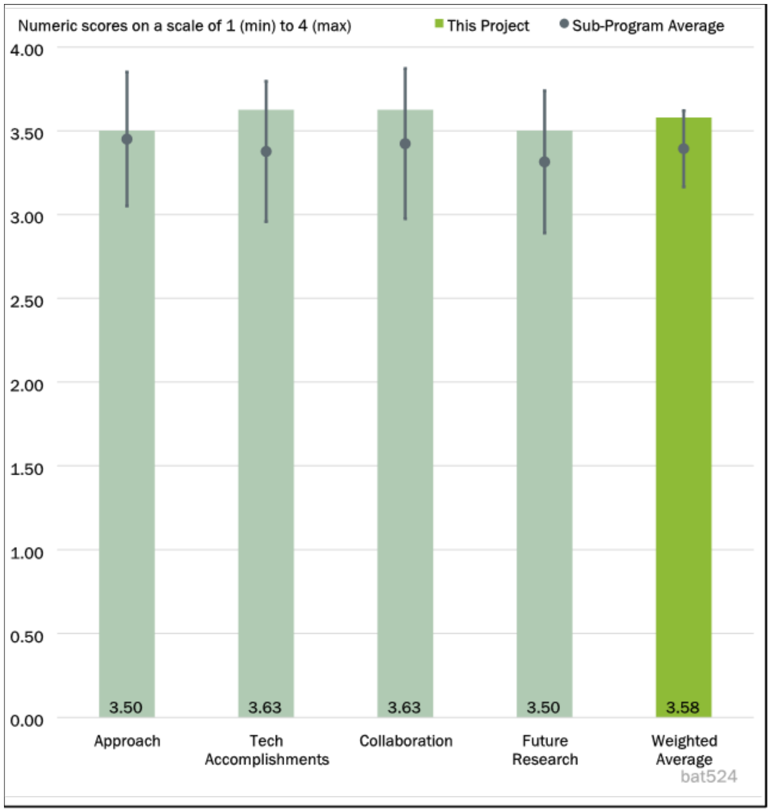

Figure 2-18 - Presentation Number: bat524 Presentation Title: Advanced Electrolytes for Lithium Metal Batteries Principal Investigator: Chunsheng Wang, University of Maryland

Question 1: Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?

Reviewer 1

The reviewer claimed that the University of Maryland team concentrates on novel electrolyte development and has achieved noticeable progress.

Reviewer 2

The reviewer declared that the effort to use lithium-fluoride (LiF) as an SEI on Li does not consider the impact of grain boundaries in the LiF, in which case the properties of pure LiF are not as important. The reviewer added that using LiF for CEI for mechanical reasons looks like a better idea. The reviewer expressed that using an ionic liquid can solve a lot of problems, but it's not clear that this is a useful approach, both because of cost and because of viscosity problems. The reviewer remarked that fluorinated carbonates have already been widely studied.

Reviewer 3

The reviewer stated that this project is well designed, the timeline is reasonable, and they have already made great progress in fluorinated electrolyte and solvent-free ionic liquid electrolyte. The reviewer noted that there have already been three publications involving this work, the reviewer assumed that much of it was already done prior to the funding of this project.

Reviewer 4

The reviewer articulated that this project proposes to stabilize the interface of Li metal and high-energy cathodes such as NMC and Sulfur through modification of electrolyte structures and formulations. The reviewer remarked that electrolytes are for sure a critical component in forming different SEI/CEI. The reviewer affirmed that the team has studied the formation mechanism of SEI/CEI well, and developed several classes of electrolytes, achieving excellent results.

Question 2: *Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.*

Reviewer 1

The reviewer claimed that the University of Maryland team accomplished two milestones in Fiscal Year 2022 (FY 2022). The reviewer added that they developed ionic liquid additives that form a LiF-rich SEI that enables greater than 85% capacity retention after 200 cycles for an NMC811 Li cell, and a solvent-free ionic liquid electrolyte demonstrating 80% capacity retention at the 330 cycle for anode-free NMC811 Cu cells. The reviewer remarked that this result is quite impressive as compared to the performance of anode-free NMC811 cells with other electrolytes from the Journal of The Electrochemical Society, 2021 168 120508.

Reviewer 2

The reviewer articulated that fluorinated solvents create an improvement, but that few details are given on how to understand the results. The reviewer added that CE is 99.9% with ionic liquid, but capacity falls with time. The reviewer stated that there was no explanation for why the capacity is falling. The reviewer observed that very flat Li metal surfaces are a major achievement, but that there was no real discussion of how to make ionic liquids practical.

Reviewer 3

The reviewer noted that this work addresses some key issues with lithium metal batteries including solvent-free electrolyte and lithium metal cathode as well as its surrounding issues including low temperature conductivity and dendritic formation.

Reviewer 4

The reviewer commented that the team has developed two generations of electrolytes including fluorinated carbonate electrolytes for Li/S batteries, and solvent-free ionic liquid electrolytes for Li metal and NMC cathode. The reviewer added that both these two electrolytes have led to significantly improved cycle stability not only in coin cells with high loading, but also in practical pouch cells.

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer remarked that the team has effective collaborations with universities, national labs, and industrial companies, and that the contribution of each collaborator is clearly indicated.

Reviewer 2

The reviewer observed that the collaboration within Battery500 is excellent.

Reviewer 3

The reviewer noted great collaboration between national labs, universities, battery suppliers and original equipment manufacturers (OEMs).

Reviewer 4

The reviewer stated that the team at the University of Maryland has collaborated well with other Battery500 consortium teams in the testing of cathodes/Li metal anode using their electrolytes.

*Question 4: **Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?***

Reviewer 1

The reviewer claimed that future research is clearly defined with target performance specified.

Reviewer 2

The reviewer was unclear that ionic liquids is a useful path forward, but remarked that studying why the PI observes such flat surfaces is very important.

Reviewer 3

The reviewer noted a lack in specific detail on operating temperature range. The reviewer added that the cycle should target 1000 cycles.

Reviewer 4

The reviewer stated that one of the critical problems for ionic liquid electrolytes is their high viscosity. The reviewer added that the team has proposed to use fluoro-antisolvent to reduce the viscosity and further enhance the cycling and calendaring performance under lean electrolytes. The reviewer declared that one of the critical barriers would be also fast-charging capability, and that it should be addressed particularly in high-viscosity electrolytes and thick electrodes.

*Question 5: **Relevance: Does the project support the overall VTO subprogram objectives?***

Reviewer 1

The reviewer expressed that this project is an important part of Battery500 and contributes greatly to the VTO objective on vehicle electrification.

Reviewer 2

The reviewer observed that the goals are very appropriate.

Reviewer 3

The reviewer remarked that this supports the VTO objective of increasing battery energy density, improving cycle life, as well as increasing safety/reducing toxicity with no solvent electrolyte.

Reviewer 4

The reviewer commented that electrolytes developed by the University of Maryland team would for sure play a critical role in enabling 500Wh/kg and 1000 cycle batteries that VTO is targeting.

Question 6: Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?

Reviewer 1

The reviewer claimed that the team has sufficient resources for their research project.

Reviewer 2

The reviewer declared that it was sufficient.

Reviewer 3

The reviewer expressed that there are significant resources dedicated to this project from both a monetary stand point as well as personnel.

Reviewer 4

The reviewer commented that the team led by Professor Wang is a world-leading group in electrolyte development for various batteries systems.

Presentation Number: bat525
Presentation Title: Fluorinated Solvent-Based Electrolytes for Low Temperature Lithium-ion Battery
Principal Investigator: John Zhang, Argonne National Laboratory

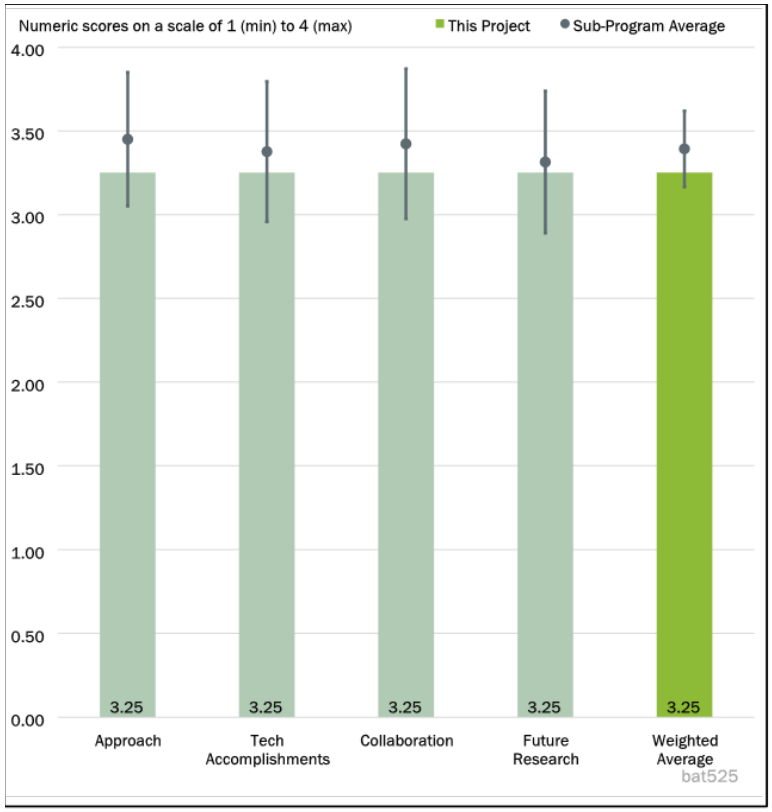

Figure 2-19 - Presentation Number: bat525 Presentation Title: Fluorinated Solvent-Based Electrolytes for Low Temperature Lithium-ion Battery Principal Investigator: John Zhang, Argonne National Laboratory

Presenter

John Zhang, ANL

Reviewer Sample Size

A total of two reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 100% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 0% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

Question 1: *Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?*

Reviewer 1

The reviewer stated that the presentation quality is high, and it was well presented during AMR. The reviewer added that there is good rationale and excellent lab techniques. The reviewer observed that overall there was solid progress in obtaining lab results, but some data and explanations on cause and effect are still lacking on the modeling side.

Reviewer 2

The reviewer remarked that guided by modeling [DFT and MD), they designed and developed well-known fluorinated electrolytes for low-temperature performance.

Question 2: *Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.*

Reviewer 1

The reviewer stated that low-temperature (LT) improvements of best systems discovered by this work meet program objectives in relation to the second-generation electrolyte, but that they are still modest and require further work to improve the lifecycle stability. The reviewer asked if the fluorinated compounds are prohibitively expensive. The reviewer mentioned that the proportion of FEC is still high (10%) compared to typical loadings seen elsewhere (less than 5%). The reviewer added that increased contact-ion pairs (CIP) in electrolytes with fluorinated solvents is problematic both for electrolyte conductivity and for breaking the energy of CIP ion association. The reviewer noted that this is a trade-off with potentially lower de-solvation energies that the fluorinated esters may provide. The reviewer remarked that there is no mention of the underlying cause behind why the CIP species are more prevalent under these conditions. The reviewer explained that this is likely due to lower permittivity of the fluorinated solvents. The reviewer concluded that the lowered de-solvation energies are claimed, but that no actual comparisons were given, especially over the number of ligands that would comprise a fully solvate lithium ion. The reviewer questioned if the solvent residence times have been validated against data, such as NMR bandwidth studies.

Reviewer 2

The reviewer claimed that they developed fluorinated ester for good LT electrolyte. The reviewer added that they developed good understanding of the Li+ solvation and impact of SEI and charge transfer on LT performance. The reviewer observed that their improved LT performance was in a cell with low loading of 1.7mAh/cm^2.

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer claimed that specific collaboration tasks with industry (Navitas, NOHMs) were not clear.

Reviewer 2

The reviewer observed that they collaborated with industry, Army Research Laboratory, and national labs.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer remarked the good set of goals that hopefully will be achieved by close of project this September.

Reviewer 2

The reviewer suggested that they need to carry out the proposed future research in a cell with more practical loading of 3 mAh/cm^2 or higher.

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

The reviewer stated that there is good alignment with VTO objectives.

Reviewer 2

The reviewer claimed that the effort met VTO's program objectives.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer remarked that the resources are adequate given the teaming and research plan.

Reviewer 2

The reviewer observed that funding is sufficient.

Presentation Number: bat526
Presentation Title: Ethylene Carbonate-lean Electrolytes for Low Temperature, Safe Lithium-ion Batteries
Principal Investigator: Bryan McCloskey, Lawrence Berkeley National Laboratory

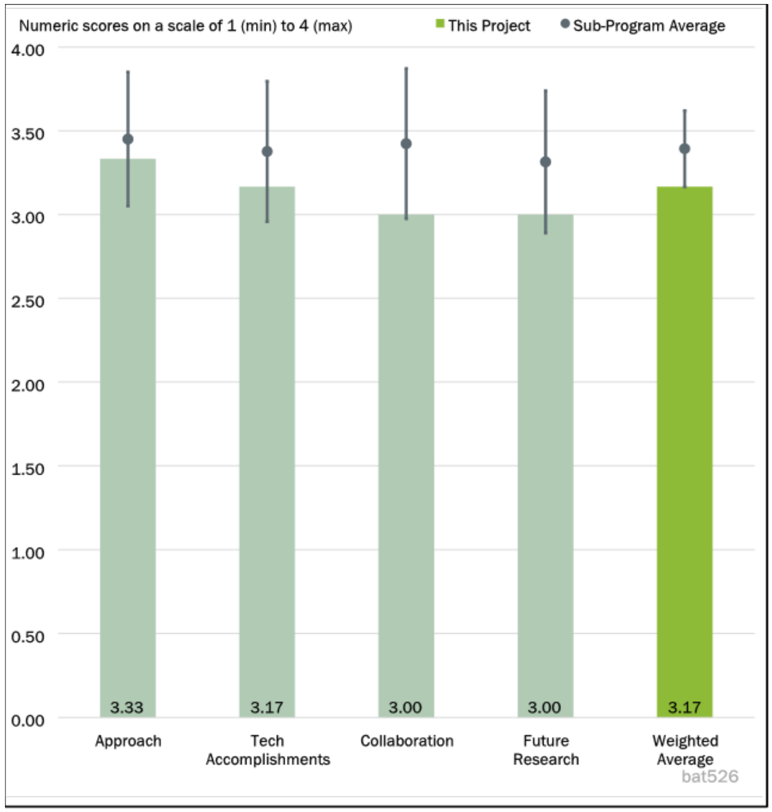

Figure 2-20 - Presentation Number: bat526 Presentation Title: Ethylene Carbonate-lean Electrolytes for Low Temperature, Safe Lithium-ion Batteries Principal Investigator: Bryan McCloskey, Lawrence Berkeley National Laboratory

Presenter

Bryan McCloskey, LBNL

Reviewer Sample Size

A total of three reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 100% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 0% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

Question 1: *Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?*

Reviewer 1

The reviewer observed that cold temperature performance is a critical area for progress, and the approach using co-solvents shows some initial promise. The reviewer added that coordinating this effort to a greater extent with research on Si electrodes is needed.

Reviewer 2

The reviewer stated that this was a clear presentation, and was easy to follow. The reviewer thanked the presenter. The reviewer added that the team has a good lab approach that helps isolate LT terms of interest.

Reviewer 3

The reviewer commented that they adopted a well-known approach for good LT electrolyte by replacing the high melting point solvent, ethylene carbonate (EC).

Question 2: *Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.*

Reviewer 1

The reviewer noted that the effects at cold temperature following cyclic aging would be more informative in understanding the potential for commercial applications, as the SEI layer effect is a critical element of this research.

Reviewer 2

The reviewer appreciated the electrode-wise evaluations. The reviewer added that the γ-butyrolactone (GBL) work seems overemphasized, as there are other routes to replace EC with stable viscosity-reducing compounds that will maintain acceptable free ion populations. The reviewer noted that on Slide 11, this is a newer technique with somewhat controversial or puzzling outcomes such as zero or negative t^+ values. The reviewer claimed that this team should provide a rationale for why the trends and magnitudes are as they appear, and why this differs from traditional electrochemical methods, probably relating to a paper or two from Balsara. The reviewer asked the team to also include those citations. The reviewer affirmed that the team correctly asserts that R_{CT} dominates cycling performance at low temperature, and that this is shown to be due more on the anode side. The reviewer claimed that they stop short at providing a mechanistic explanation for this. The reviewer commented that the A1 additive provides modest improvement to get just above 70% capacity at −20°C, C/3 rate. The reviewer also suggested that the team should consider other common-sense co-solvents to reduce viscosity, such as low molecular weight esters and fluorinated variants. The reviewer declared that even small amounts (10%–20%) can have a noteworthy improvement in capacity delivery at LT. The reviewer concluded that this project taken as a whole, has a high quality of work, but recommended that more electrolyte systems could have been considered, screened and tested to arrive at a richer field of options.

Reviewer 3

The reviewer stated that they were unsuccessful in replacing EC with GBL but developed good understanding of the impact of charge transfer resistance on LT performance. The reviewer added that they developed an additive that resulted in better LT performance in a NMC622/graphite cell than the baseline second generation electrolyte. The reviewer noted that the performance difference in Slide 14 might not be statistically significant.

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer commented that greater collaboration with materials development in EERE would strengthen the relevance of this activity.

Reviewer 2

The reviewer noted that the collaboration appears to be primarily within Lawrence Berkeley National Laboratory (LBNL), with support on materials from Argonne National Laboratory (ANL). The reviewer observed that there was no mention of collaboration with industry. The reviewer suggested that they should consider expanding collaboration either with other DOE labs, universities and/or with industry.

Reviewer 3

The reviewer observed that they collaborated with LBNL and Cell Analysis, Modeling, and Prototyping Facility (CAMP) at ANL.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer claimed that the project is nearly complete, and will likely meet the project deliverables. The reviewer stated that extending this work to future electrode materials would be interesting.

Reviewer 2

The reviewer noted that the suggested future work has reasonable targets, but they are few and quite limited in scope.

Reviewer 3

The reviewer suggested that in addition to the proposed future work, they should also characterize the impact of the A1 additive on high-temperature performance.

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

The reviewer observed that this work is of fundamental relevance to the battery program, as improved cold temperature performance is needed for nationwide adoption of EVs.

Reviewer 2

The reviewer noted that there is good alignment with VTO objectives.

Reviewer 3

The reviewer claimed that the work met the VTO program objectives.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer claimed that looking at the project as it nears completion, the resources appear to have been adequate for a successful study of low-temperature electrolytes.

Reviewer 2

The reviewer observed that the resources are adequate given the teaming and research plan.

Reviewer 3

The reviewer stated that funding is sufficient.

Presentation Number: bat527
Presentation Title: Synthesis, Screening and Characterization of Novel Low Temperature Electrolyte for Lithium-ion Batteries
Principal Investigator: Xiao-Qing Yang, Brookhaven National Laboratory

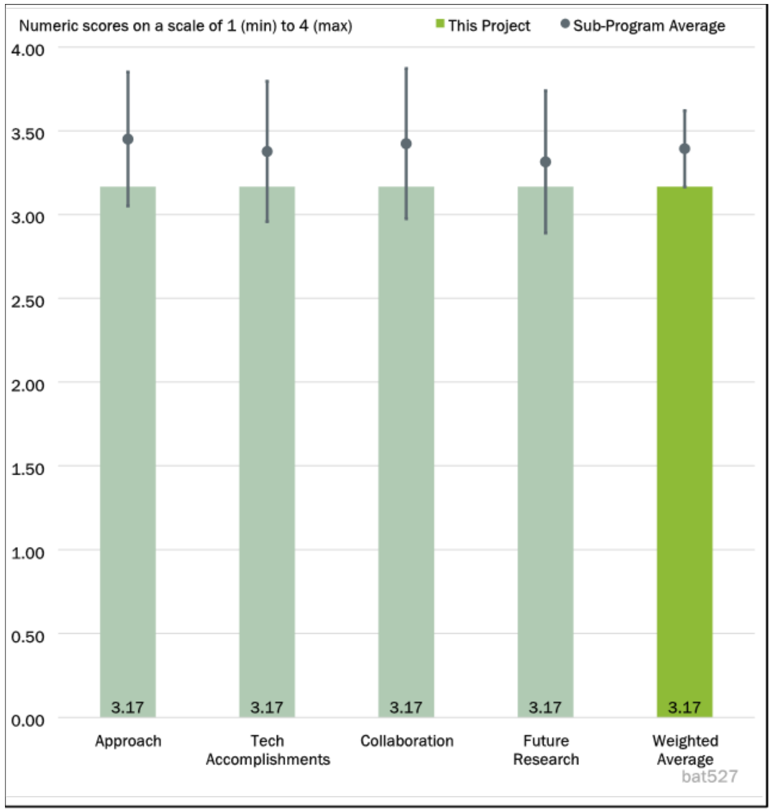

Figure 2-21 - Presentation Number: bat527 Presentation Title: Synthesis, Screening and Characterization of Novel Low Temperature Electrolyte for Lithium-ion Batteries Principal Investigator: Xiao-Qing Yang, Brookhaven National Laboratory

Presenter

Xiao-Qing Yang, BNL

Reviewer Sample Size

A total of three reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 100% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 0% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

Question 1: Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?

Reviewer 1

The reviewer noted that the overall thrust is going in a positive direction. The reviewer observed that the presentation is very wordy and not well organized for review. The reviewer concluded that the contributions from the team members seem disjointed. The reviewer added that the electrolyte design strategy with LHCEs is understood; however, the extent of CIP and SSIP formation is concerning in that very low fractions of "free" lithium ions are projected (less than 0.07) over the temperature range of -60 to 60°C (Slide 16). The reviewer remarked that the team seems particularly keen in publishing and securing patents.

Reviewer 2

The reviewer claimed that this project is nearly complete with a wide scope of electrolytes tested at low temperatures and cathode materials.

Reviewer 3

The reviewer declared that guided by modeling (MD), they relied on a combinatory approach of salt, solvent and additives to achieve good LT performance.

Question 2: *Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.*

Reviewer 1

The reviewer stated that cycle life is still of concern if it can be brought up to 1000 cycles. The reviewer added that commercial grade cell testing would also be of value.

Reviewer 2

The reviewer remarked that they demonstrated good performance at both RT and LT, using LHCE with different diluents, and in Li/SPAN or NMC811 cells. The reviewer added that most of the data was based on relative low loading of 2.5 mAh/cm^2. The reviewer observed that instead of the plethora of data, they need to focus on demonstrating overall good performance (LT and HT) in one cell chemistry.

Reviewer 3

The reviewer asked what F1, F2, F3…are. The reviewer added that not knowing what these are does not help with evaluation of the technical objectives. The reviewer asked if the fluorinated compounds are prohibitively expensive. The reviewer noted that achievable capacity at LT is obtained at 0.2C cycling rate (Slides 13,14) and 0.1C (Slide 13). The reviewer stated that however, the performance requirement is that the achievable capacity (particularly at -20 C) be determined at a C/3 rate. The reviewer noted that in relation to responses to last year's reviewer comments, one reviewer mentioned the thermodynamics of electrolyte phase behavior at lower temperatures, wherein there can be electrode surface-driven interactions. The reviewer remarked that the response was "The work done this year followed this suggestion and carried out the thermodynamic studies of electrolyte at low temperatures." The reviewer mentioned that nothing is found in the presentation regarding the thermodynamics of electrolyte phase behavior. The reviewer observed that on the Summary slide it states "Improved low temperature performance was obtained when esters (methyl acetate, MA and methyl propionate, MP) are added as co-solvents in the carbonate-based electrolyte formulations." The reviewer noted that these LT results are nowhere to be found in the presentation materials, except for scant mention on Slide 10. The reviewer concluded that there was a mixed message on anodes used in the work with graphite mentioned in some places while lithium metal is also mentioned (Slides 8,9). The reviewer asked what the ultimate focus to meet the LT goals of this project was.

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer claimed that the team had good representation from PIs in each organization, and that there is no mention of industrial collaborators.

Reviewer 2

The reviewer noted the collaboration between several national labs and universities, but stated that they are missing industry partners.

Reviewer 3

The reviewer remarked that BNL collaborated with universities and other national labs.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer noted a good set of goals that hopefully will be achieved by close of project this September.

Reviewer 2

The reviewer observed that the work is mostly done with the exception of publications.

Reviewer 3

The reviewer remarked that the project will be complete by the end of FY 2022.

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

The reviewer claimed that there is good alignment with VTO objectives.

Reviewer 2

The reviewer noted the low temperature battery electrolyte that also has good high temperature performance.

Reviewer 3

The reviewer remarked that the work met VTO program objectives.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer remarked that the resources are adequate given the teaming and research plan.

Reviewer 2

The reviewer expressed that they achieved a great amount of work in a three-year time frame that the project is awarded for.

Reviewer 3

The reviewer affirmed that the funding is sufficient.

Presentation Number: bat528
Presentation Title: Structurally and Electrochemically Stabilized Silicon-rich Anodes for Electric Vehicle Applications
Principal Investigator: Murali Ramasubramanian, Enovix

Presenter

Murali Ramasubramanian, Enovix

Reviewer Sample Size

A total of five reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 80% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 20% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

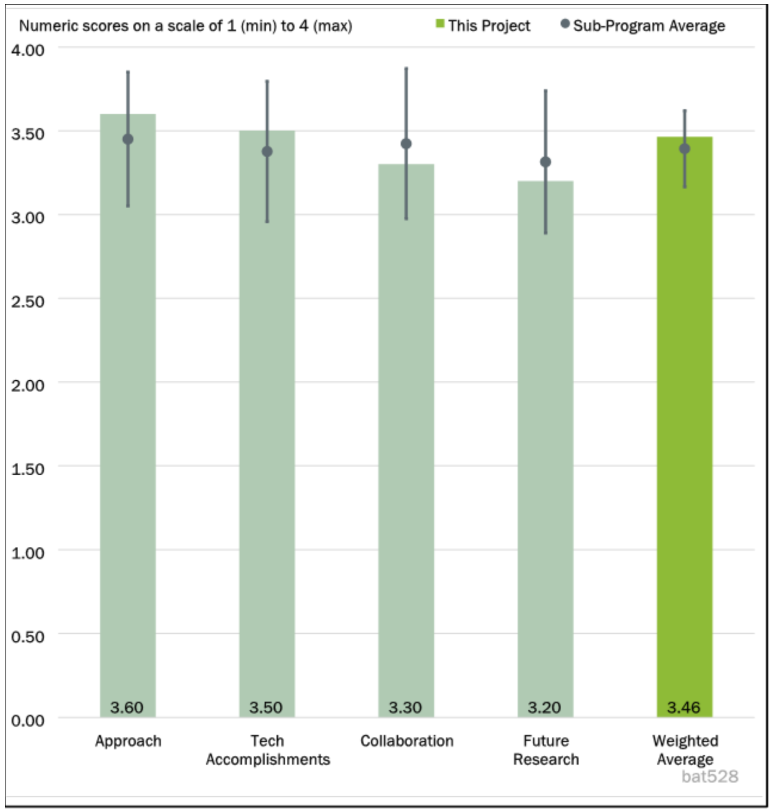

Figure 2-22 - Presentation Number: bat528 Presentation Title: Structurally and Electrochemically Stabilized Silicon-rich Anodes for Electric Vehicle Applications Principal Investigator: Murali Ramasubramanian, Enovix

Question 1: Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?

Reviewer 1

The reviewer noted that the Enovix cell design is unique, and that the approach of reorienting the electrodes to more easily apply pressure and allow prelithiation is promising, interesting, and very innovative. The reviewer added that demonstrating that the design can be scaled-up with higher energy density, and that it can be fabricated inexpensively, are the keys to overall success in Year 2.

Reviewer 2

The reviewer commented that the cycle and calendar life of Si anode are the two most prominent problems and the technology seems to provide a good solution.

Reviewer 3

The reviewer noted the very well-designed project, and stated that a lot was accomplished with the time given. The reviewer looked forward to the following year's results. The reviewer remarked that the approach is clearly laid out and well-explained.

Reviewer 4

The reviewer commented that the Enovix architecture and cell design will be illustrated by cycling performance and calendar year in the first year and the scaling up ability (energy density at EV battery sizes) will be confirmed in the second year. The reviewer concluded that the project is well designed with good plans.

Reviewer 5

The reviewer expressed that Enovix proposed oriented electrodes to the small area sides of the cell to contain the pressure from the large Si volume change. The reviewer stated that while it is an innovative technique to mitigate swelling, it is not clear if their technique can be mass produced, especially when pre-lithiation is still needed to reduce first cycle irreversibility loss.

Question 2: *Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.*

Reviewer 1

The reviewer expressed that the project has demonstrated initial performance (cycle life, calendar life) which is promising and trending to meet targets. The reviewer added that the energy density, however, remains far from the target even when projected into a larger cell size. The reviewer also remarked that the amount of prelithiation is likely critical to the life results. The reviewer claimed that since this amount was not disclosed, it makes it hard to confirm the results are realistic and can be scaled. The reviewer articulated that a more fundamental understanding of the degradation mechanism is necessary and should be pursued.

Reviewer 2

The reviewer commented that great progress has been made in the calendar life performance, but that the electrode loading or packing efficiency still needs to be improved.

Reviewer 3

The reviewer noted the amazing accomplishment, and hoped everything works out.

Reviewer 4

The reviewer declared that the cycling performance is even better than that of the project plan. The reviewer added that the calendar life is also very promising based on the results until now. The reviewer remarked that the model Si porosity and SEI evolution need to be further developed to provide guidance.

Reviewer 5

The reviewer observed that the Enovix cell demonstrated greater than 1000 cycles but in a 200 Wh/kg cell. The reviewer added that the team also demonstrated greater than 9 months of capacity retention at 50C prolonged storage at 100% state of charge (SoC) in the 200 Wh/kg cell. The reviewer claimed that the Enovix cell with Si anode also demonstrated less leakage current than graphite-based cells at 4.2 volt (V), indicative of long calendar life.

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer noted that the project team seems to be communicating and collaborating effectively.

Reviewer 2

The reviewer observed that Enovix partnered with the National Renewable Energy Laboratory (NREL) and Mitsubishi Chemical.

Reviewer 3

The reviewer suggested that more mechanism understanding is needed, and that their national lab partner can be helpful for characterization.

Reviewer 4

The reviewer observed that there is a great partnership with NREL and Mitsubishi Chemical Corporation, but that they could possibly partner with OEMs.

Reviewer 5

The reviewer commented that the calendar life estimation and modeling are made by NREL. The reviewer added that they also have collaboration with Mitsubishi Chemical Corporation on the electrolyte designation. The reviewer suggested that more collaboration may be needed on the characterization to demonstrate the mechanism behind it.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer claimed that the unique cell architecture is the enabler for the Si anode in Enovix's design. The reviewer stated that it is not fully clear how this cell can be effectively scaled up and still meet 350 Wh/kg. The reviewer observed that detailing the design details, and electrodes particularly, will significantly increase the energy density and would be helpful. The reviewer expressed that more disclosure and analysis of prelithiation amount is also important. The reviewer concluded that the project needs to demonstrate that this cell architecture can be scaled up to EV relevant size at EV compatible costs.

Reviewer 2

The reviewer stated that improvement in energy density is required.

Reviewer 3

The reviewer looked forward to the following year's results.

Reviewer 4

The reviewer remarked that the future work is well illustrated. The reviewer observed that even though they are demonstrating a cell-size scalability by building greater than 2 Ah cells with high energy goals, it is highly possible that they can achieve the targets based on their results.

Reviewer 5

The reviewer stated that in addition to demonstrating more than 1000 cycles, they should report RT calendar life data projected based on their 45^0C data, if necessary, in cells with more than 350 Wh/kg at higher than

4.3V charge cut-off. The reviewer recommended that they also need to report thickness change at 1000 cycles and at the completion of calendar life in cells with more than 350 Wh/kg in the same cells.

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

The reviewer mentioned that this project is clearly innovative and interesting. The reviewer stated that it is difficult to balance disclosing too much to lose competitive advantage and too little to lose the ability to convey a convincing plan. The reviewer added that right now, however, the approach to ultimately reach the targets in energy density and cost is fuzzy. The reviewer recommended that more details should be provided to ensure the project can be successful and will further the DOE's goals.

Reviewer 2

The reviewer said yes.

Reviewer 3

The reviewer commented that the silicon anode will greatly improve existing battery performance.

Reviewer 4

The reviewer observed that it supports the batteries subprogram objectives.

Reviewer 5

The reviewer noted that the work met the VTO program objectives.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer expressed that a lot of work is being done for the budget.

Reviewer 2

The reviewer stated that the project seems well resourced.

Reviewer 3

The reviewer articulated that the resources including high level cell building, modeling, and electrolyte developing should be sufficient for this project.

Reviewer 4

The reviewer claimed that funding is sufficient.

Reviewer 5

The reviewer made no comments.

Presentation Number: bat529
Presentation Title: Rationally Designed Lithium-Ion Batteries Towards Displacing Internal Combustion Engines
Principal Investigator: Rick Costantino, Group 14 Technologies

Presenter

Rick Costantino, Group 14 Technologies

Reviewer Sample Size

A total of five reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 100% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 0% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

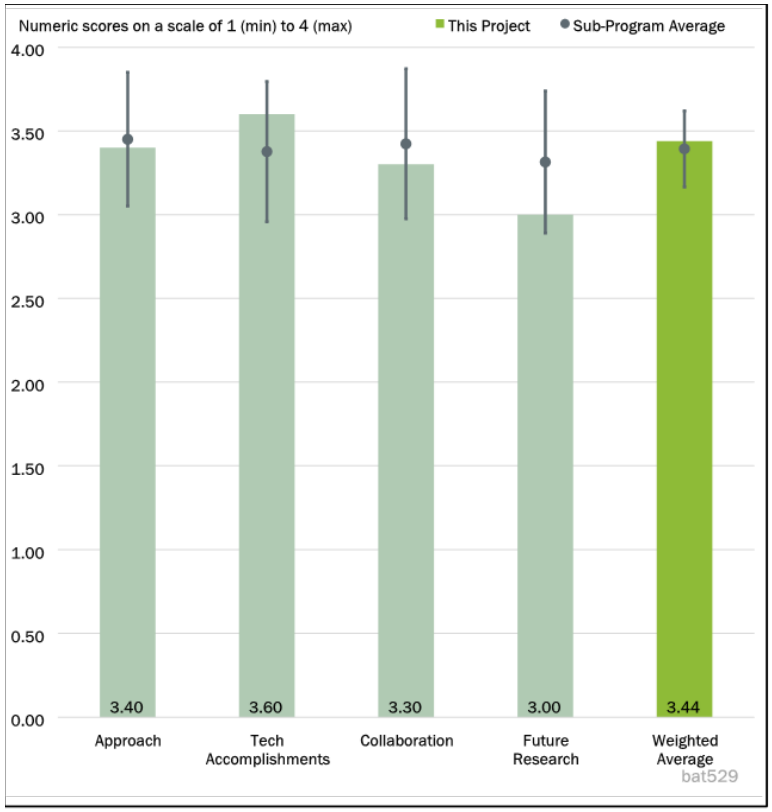

Figure 2-23 - Presentation Number: bat529 Presentation Title: Rationally Designed Lithium-Ion Batteries Towards Displacing Internal Combustion Engines Principal Investigator: Rick Costantino, Group 14 Technologies

Question 1: *Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?*

Reviewer 1

The reviewer stated that the first 2 years have shown impressive progress toward developing durable Si electrodes.

Reviewer 2

The reviewer expressed that embedded Si inside a carbon cage seems a pretty effective way to protect the Si particles losing connection from the conductive network which greatly improved the cycle life of Si.

Reviewer 3

The reviewer articulated that the project is well designed. The reviewer noted that the first year has goals to deliver a cell with more than 340 Wh/Kg, more than 300 cycles and more than 3-year calendar life; the second year has goals to deliver the same energy density with more than 600 cycles and more than 5-year calendar life; and the last year has goals to deliver more than 1000 cycles and more than 10-year calendar life. The reviewer observed that the plan is well designed step by step, which makes it very reasonable.

Reviewer 4

The reviewer declared that the team proposed carbon scaffold and designed porosity to mitigate the large volume change of high Si percentage anode using nano Si. Group 14's approach is very similar to the earlier Sila approach.

Reviewer 5

The reviewer claimed that the approach, based on Group 14's unique material, is well sequenced, the timeline looks reasonable, and the performance metrics obtainable. The reviewer added that the material itself, even though limited fundamental information was provided (reporting the particle void volume and showing additional photos would be helpful), addresses the known shortfalls in Si-anode technology. The reviewer remarked that no information was provided on cost, however, which will be a key for eventual market adoption.

Question 2: *Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.*

Reviewer 1

The reviewer observed that great progress has been made on the cycle life of Si materials.

Reviewer 2

The reviewer stated that advanced characterization of electrode materials will be important going forward, including deeper investigation of surface characteristics, particularly at high load. The reviewer added that the analysis will enable another feedback loop into the process/material improvements.

Reviewer 3

The reviewer claimed that the initial results are encouraging, demonstrating that the material functions reasonably well. The reviewer posed the following questions:

- Does processing differ from typical Li-ion electrodes? Solvent types? Compatible with existing equipment?
- What is the impedance growth? This is a known issue with Si containing anodes and should be shown.
- What is coulombic efficiency?
- Do you understand the degradation mechanism? Understanding why the capacity fades (and likely impedance grows) is likely necessary to significantly increase the life as required for the Phase II and III metrics.
- More details on the "interim" builds would be useful for the community. What specifically was done to improve the cell's performance? Was it all anode conductivity enhancements?
- The reduction in gassing with OS3® additive is encouraging but do you have a gassing target? How much gassing can be allowed?

Reviewer 4

The reviewer expressed that the target on energy density and cycle life have been achieved. The reviewer added that the calendar life is still to be determined, but calendar life data over 9 months looks promising.

Reviewer 5

The reviewer remarked that they demonstrated 350 cycles to 80% capacity retention but it was not clear if the cycle life data was in the reported 340 Wh/kg cell. The reviewer suggested that they should also report thickness change at 350 cycles. The reviewer observed that they reported excellent 45^0C storage for over 9 months at 100% SoC but it was not clear if the excellent 45^0C data was done in the reported 340 Wh/kg cell. The reviewer recommended that they should also report thickness change after 45^0C prolonged storage. The reviewer clarified that they projected more than 800 cycles using modified interim anode design but did not report the energy density or thickness change.

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer stated that the material analysis may be an opportunity to expand collaboration with DOE labs.

Reviewer 2

The reviewer claimed that different teams appear to be working well together with adequate communication and planning.

Reviewer 3

The reviewer remarked that great teams are combined in this program and each party has demonstrated significant contribution.

Reviewer 4

The reviewer articulated that Group 14 partnered with well-known industry leaders of components such as electrolyte, binder, and cell maker to demonstrate good performance of their Si anode.

Reviewer 5

The reviewer expressed that they do have many collaborations with other industry and national laboratories, but detailed contributions of them are not so clear. The reviewer added that considering the time needed to do the calendar life test, they may need to collaborate with others on the modeling and prediction of calendar life.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer stated that the description of future work is overly general for a reviewer to offer useful comment, and that progress to date shows success. The reviewer was hopeful that further concentrated effort will yield more positive results.

Reviewer 2

The reviewer expressed that the barriers are clearly identified and the correct focuses are clarified.

Reviewer 3

The reviewer commented that they proposed to further optimize the component of the lithium-silicon and test down-selected combinations to achieve the final project goals. The reviewer added that the target of energy density and cycle life is highly achievable based on their results, but the target on calendar life still requires many efforts.

Reviewer 4

The reviewer remarked that in addition to demonstrating more than 1000 cycles, they should report RT calendar life data projected based on their 45^0C data, if necessary, in cells with greater than 350 Wh/kg at more than 4.3V charge cut-off. The reviewer added that they also need to report thickness change at 1000 cycles and at the completion of calendar life in cells with more than 350 Wh/kg in the same cells.

Reviewer 5

The reviewer claimed that it is not clear from the slides what work will be done to further improve the performance. The reviewer explained that the bullets are too broad and non-specific, and that without additional details it is difficult/impossible to determine the likelihood of success.

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

The reviewer claimed that the project appears to be a significant contribution to commercialization of Si electrodes for automotive applications, of prime relevance to the Battery project area.

Reviewer 2

The reviewer explained that the advanced Si-anodes are clearly relevant and, if successful, can significantly impact the trajectory of EV vehicles in the future.

Reviewer 3

The reviewer said yes.

Reviewer 4

The reviewer claimed that the project supports the batteries as well as the materials objectives.

Reviewer 5

The reviewer expressed that the work met the VTO program objectives.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer noted that the resources appear appropriate for progress toward final build of a battery meeting the technical requirements.

Reviewer 2

The reviewer remarked that the resources appear well matched with the project's results so far.

Reviewer 3

The reviewer observed that they have many collaborations with world leaders in different areas including binder, conductive additive, and electrolytes. The reviewer added that they have support from national labs like PNNL. The reviewer concluded that the resources are very sufficient.

Reviewer 4

The reviewer stated that the funding is sufficient.

Reviewer 5

The reviewer made no comments.

Presentation Number: bat530
Presentation Title: Ultra-Low Volume Change Silicon-Dominant Nanocomposite Anodes for Long Calendar Life and Cycle Life
Principal Investigator: John Tannaci, Silanano

Presenter

John Tannaci, Silanano

Reviewer Sample Size

A total of five reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 100% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 0% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

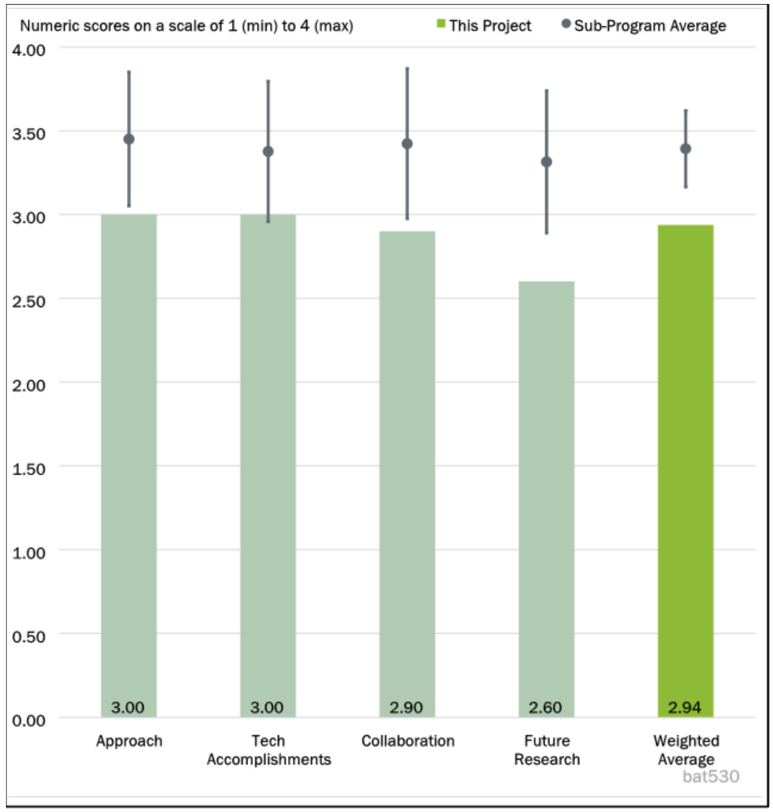

Figure 2-24 - Presentation Number: bat530 Presentation Title: Ultra-Low Volume Change Silicon-Dominant Nanocomposite Anodes for Long Calendar Life and Cycle Life Principal Investigator: John Tannaci, Silanano

Question 1: Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?

Reviewer 1

The reviewer claimed that by investigating the process-structure-property relationship and designing electrolyte, the energy density and cycling performance target will be achieved in the small format cells in the first year, and then applied to large size 1 Ah cells in the second year. The reviewer added that in the third year, the performance will be further improved to achieve high cycling performance and calendar life. The reviewer concluded that the project is well designed and has a reasonable timeline.

Reviewer 2

The reviewer expressed that Sila developed the carbon scaffold, with designed porosity, and nano-Si to mitigate the swelling issue of Si anode.

Reviewer 3

The reviewer explained that Sila’s technology is promising based on initial results. The reviewer stated that the optimization of the structure has provided improved performance, but that it is difficult to determine the appropriateness of the work plan given the vague descriptions of the technology itself. The reviewer asked what the structure is, what the basics of the chemistry are, what the likely degradation mechanisms will be, what the amount of anode utilized is, and what mechanism will be used to avoid volume change during cycling.

Reviewer 4

The reviewer noted that too little technical information was provided and it is not clear how various issues were addressed.

Reviewer 5

It was not clear to the reviewer how Sila particles reduce volume changes in silicon dominated anodes. The reviewer asked if the chart on Slide 6 is supposed to be a comparison of a lithiated Sila anode and an unlithiated anode to show 250-300% increase in volume or if it just shows that Sila particles have more surface area. The reviewer expressed that Sila particles show good improvements in anode capacity as well as improved cycle life, but more tests need to be performed. The reviewer couldn't tell if only two cells were tested. The reviewer added that it was unclear what kind of cells these are and which cathodes were being used. The reviewer thought more explanation was needed on the methodologies, materials use, and testing processes. The reviewer understood that this is industry work, that not everything can be openly said, but there can still be more background on what experiments took place and how they were performed, on how many samples were considered, and so forth.

Question 2: *Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.*

Reviewer 1

The reviewer articulated that Sila demonstrated high energy density over250 Wh/Kg, more than 750 Wh/L and over 750 cycles with 80% capacity retention by small format cells. The reviewer added that in the meantime, the calendar life is as long as over 2.5 years at 25°C in 0.2 Ah+ cells. The reviewer noted that 1Ah cells are still on track, but they already made good progress on the target.

Reviewer 2

The reviewer mentioned that the technical progress is on schedule according to the technical plan. The reviewer remarked that Phase I targets are being met, but that impedance growth should be added as a target and reported. The reviewer posed the following questions:

- Do you understand the degradation mechanism?
- What is the coulombic efficiency to the first cycle and during cycling?
- Energy density is significantly lower than the ultimate target. What is limiting it? How will this limit be overcome? Are the new particles with higher volumetric energy density enough? Is the performance and life of the enhanced particles impacted?

Reviewer 3

The reviewer claimed that some promising cycle life was demonstrated, but no details were provided in terms of cell format, electrode loading, etc.

Reviewer 4

The reviewer remarked that it seems like the project is making good progress towards its goals. The reviewer added that the work with PNNL and Army Research Lab (ARL) should produce interesting results. The reviewer expressed that the PI needs to explain with more clarity and precise language in which area the technical progress has been made and how it has been made.

Reviewer 5

The reviewer observed that they demonstrated over 1000 cycles in an LCO/Si cell with medium loading of 3 mAh/cm^2. The reviewer added that they also demonstrated 4 months of 45°C storage with higher loading of 4mAh/cm^2 but in a different cell using NMC/Si chemistry. The reviewer suggested that the team needs to report thickness change after cycling or high temperature storage.

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer stated that the groups are working well together and effectively contributing as a single team.

Reviewer 2

The reviewer noted that Sila collaborated with PNNL and ARL.

Reviewer 3

The reviewer declared that a number of OEMs are working to improve anode capacity using silicon, and Sila has said they will be working with OEM partners to make automotive cells using automotive grade active cathode, which is good progress. The reviewer recommended that they should also sort out the industry level tests that are necessary to reach automotive specifications for batteries.

Reviewer 4

The reviewer expressed that Sila collaborated with PNNL to develop the electrolyte and collaborated with ARL to model and screen the electrolyte and to investigate the SEI formation mechanism. The reviewer added that the results of the modeling part was not provided in this review. The reviewer recommended that more collaboration on the electrolyte designation may be helpful for them to achieve a better cycling performance and calendar life.

Reviewer 5

The reviewer claimed that it is not quite clear what the collaborators have contributed to the program.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer mentioned that while initial results are encouraging, there are no concrete details provided to improve to Phase II targets. The reviewer suggested that more specific plans and actions should be reported to ensure confidence in the program.

Reviewer 2

The reviewer claimed that the barriers were obvious but was not sure of the developer's approach to close the gap.

Reviewer 3

The reviewer stated that there is not enough information on future work and that it is unclear what they will do to determine if they will achieve their future target. The reviewer added that the proposed work includes surface chemistry, slurry optimization, and coating density, but this is run of the mill work for any materials processing. The reviewer questioned what was getting done and asked how the team will actually achieve the future target.

Reviewer 4

The reviewer expressed that Sila will further improve their anode by surface chemistry, slurry optimization and coating density. The reviewer added that in the meantime, Sila will work with PNNL and ARL to further design the electrolyte and SEI for better performance. The reviewer noted that it might not be easy to improve their cell from over 250 Wh/Kg to 350 Wh/Kg, but that it was still possible for them to achieve it after more than one year.

Reviewer 5

The reviewer commented that in addition to demonstrating over 1000 cycles, they should report RT calendar life data projected based on their 45^{o}C data, if necessary, in cells with over 350 Wh/kg at over 4.3V charge cut-off. The reviewer stated that they also need to report thickness change at 1000 cycles and at the completion of calendar life in cells with over 350 Wh/kg in the same cells.

*Question 5: **Relevance: Does the project support the overall VTO subprogram objectives?***

Reviewer 1

The reviewer noted that the advanced Si composites are well aligned with DOE's targets for EV improvements.

Reviewer 2

The reviewer made no comments.

Reviewer 3

The reviewer stated that the increase in silicon content will increase capacity, but it has to be demonstrable on a large-scale format battery that it will have the cycle life as graphite anodes.

Reviewer 4

The reviewer claimed that Sila designed silicon-dominant nanocomposite materials which will help the development of next-generation Li-ion batteries, and that it will definitely support the batteries objectives.

Reviewer 5

The reviewer remarked that Sila's work met VTO's program objectives.

Question 6: Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?

Reviewer 1

The reviewer commented that Sila is good at silicon nano-composite anode and EV cell building, and they have support on electrolyte from both PNNL and ARL. The reviewer added that it will make it easier for them to achieve the milestone in a timely manner.

Reviewer 2

The reviewer remarked that funding is sufficient.

Reviewer 3

The reviewer declared that it is unclear what resources are available to Sila, and that the project team should make it clear what resources they have and require to achieve their targets.

Reviewer 4

The reviewer stated that the resources appear to match outputs and that it is difficult to judge without more details on the future work scope/efforts.

Reviewer 5

The reviewer claimed that if the program will continue, much more technical details and gap analysis need to be provided. The reviewer added that the presentation was poorly prepared and that it is not clear at all where their technology is and how big the gap is to the target.

Presentation Number: bat531
Presentation Title: Solid State Lithium-ion Batteries Using Silicon Composite Anodes
Principal Investigator: Pu Zhang, Solid Power Battery

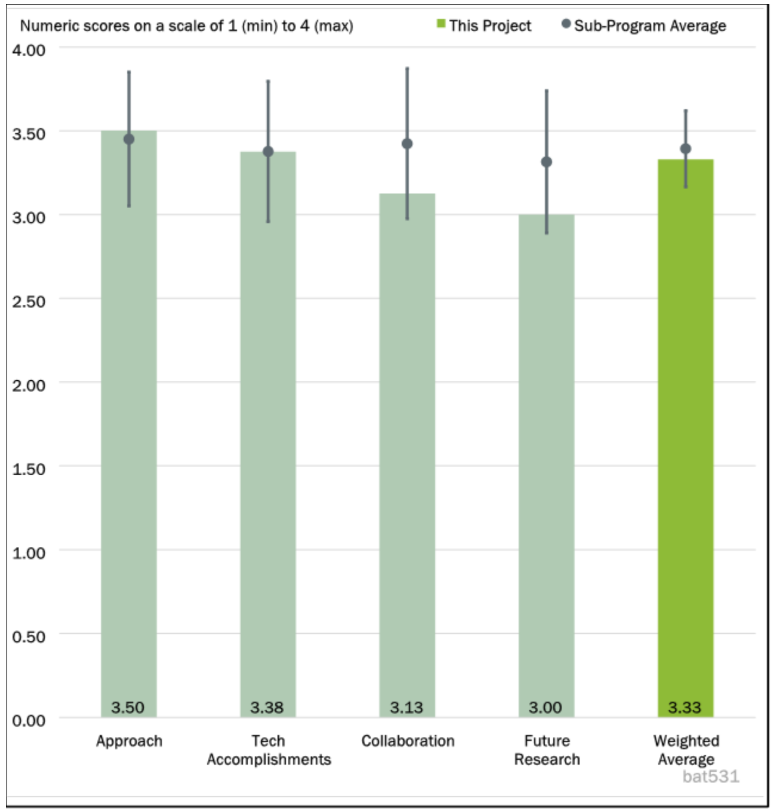

Figure 2-25 - Presentation Number: bat531 Presentation Title: Solid State Lithium-ion Batteries Using Silicon Composite Anodes Principal Investigator: Pu Zhang, Solid Power Battery

Presenter

Pu Zhang, Solid Power Battery

Reviewer Sample Size

A total of four reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 100% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 0% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

Question 1: Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?

Reviewer 1

The reviewer declared that the combination of the solid electrolyte and Si seems to be a good approach to address Si anode degradation in cycle and calendar life.

Reviewer 2

The reviewer remarked that the project is well designed from December 2020 to December 2022. The reviewer added that the approach of every quarter is very detailed including equipment secured, cathode selected, single cell stack, and roll-to-roll coated, which make the plan very reasonable.

Reviewer 3

The reviewer observed that Solid Power leveraged their unique solid-state electrolyte to achieve good performance in a Li-ion cell based on Si anode.

Reviewer 4

The reviewer commented that the approach relies upon sulfide material forming a stable SEI with Si composites. The reviewer claimed that this is a promising approach because sulfide materials have shown some stability to lithiated Si. The reviewer expressed that solid state, however, introduces additional issues with Si in terms of ability to handle the volume changes during lithiation. The reviewer asked if the ionic structure of the electrode survives well enough to enable long life cycling. The reviewer mentioned that understanding if this issue is critical to evaluating the approach. The reviewer observed that so far, initial cycling results look encouraging but more emphasis on degradation mechanism understanding may be necessary to meet the project's goals.

Question 2: *Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.*

Reviewer 1

The reviewer noted that good improvement in capacity and cycle life was demonstrated. The reviewer added that the 1500 mAh/g specific capacity for Si anode delivered pretty decent cycle life performance.

Reviewer 2

The reviewer commented that Solid Power achieved Si composite anode with an electrode level capacity of 1500 mAh/g. The reviewer added that a stable cathode was developed to match the Si anode, which enabled a pouch cell with a good cycling performance of up to 500 cycles. The reviewer remarked that the target was well achieved.

Reviewer 3

The reviewer stated that the project has demonstrated that sulfide cells with Si anodes function reasonably well. The reviewer remarked that the ability to process the materials into high quality cells is impressive. The reviewer made the following comments and suggestions:

- Impedance growth needs to be measured and tracked. This is directly relevant to the stability of the ionic pathways in the anode.
- Calendar life needs to be measured and reported.
- More focus on understanding degradation mechanism will better guide future work.
- What is the structure of the Si composite? Before and after SEM images would be informative.

Reviewer 4

The reviewer claimed that they achieved over 500 cycles at 45^{o}C using NMC cells with Si dominant anode, albeit the loading was only 3mAh/cm^2, insufficient to meet the 350 Wh/kg goal.

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer claimed that it is unclear what Argonne's role is. The reviewer suggested directing them toward understanding the degradation mechanism.

Reviewer 2

The reviewer made no comments.

Reviewer 3

The reviewer stated that Solid Power collaborated with ANL to do materials characterization and cell failure analysis, which will be helpful for them to learn the mechanism behind and further improve the battery performance. The reviewer suggested that it would be better to show the results from the collaboration in the review meeting.

Reviewer 4

The reviewer noted that Solid Power collaborated with ANL.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer stated that Solid Power will further improve the stability of the composite anode to achieve a better cycle life and make thinner separators to get a higher energy density. The reviewer added that considering there is still much space to further reduce the thickness of the separator, it is highly possible for them to achieve the targets.

Reviewer 2

The reviewer made no comments.

Reviewer 3

The reviewer expressed that future work should focus on loading 4 mAh/cm^2 and performance at RT or below. The reviewer added that they also need to focus on calendar life and develop an understanding on the calendar life degradation mechanism using their SSE, to validate the calendar life degradation mechanism proposed by the Si Consortium. The reviewer recommended that in addition to demonstrating over 1000 cycles, they should report RT calendar life data projected based on their 45ºC data, if necessary, in cells with over 350 Wh/kg at greater than 4.3V charge cut-off. The reviewer also suggested that they need to report thickness change at 1000 cycles and at the completion of calendar life in cells with over 350 Wh/kg in the same cells.

Reviewer 4

The reviewer observed that the proposed future research section is just a restatement of the project's goals. The reviewer explained that it does not provide any details on the flow or plan of work for the project, and that more details are needed. The reviewer added that understanding the degradation mechanism is key to effectively planning the future work to ensure successful completion of the project.

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

The reviewer claimed that this program combines solid state and Si anodes and that both are highly relevant to the DOE's targets.

Reviewer 2

The reviewer said yes.

Reviewer 3

The reviewer remarked that Solid Power make solid electrolyte enabling high performance Si anode, the roll-to-roll process enabling a scalable cell manufacturing, which support next-generation safe Li-ion batteries.

Reviewer 4

The reviewer observed that this effort met the VTO's program objectives.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer claimed that additional work is needed to understand degradation mechanisms. The reviewer added that overall, however, the program might be well resourced but that resources may just need to be redirected.

Reviewer 2

The reviewer made no comments.

Reviewer 3

The reviewer stated that Solid Power have good capability to build large solid state pouch cells, and that the collaboration with ANL and DOE VTO will make the milestones achievable.

Reviewer 4

The reviewer expressed that the funding is sufficient.

Presentation Number: bat532
Presentation Title: Electrolytes with Lithium-ion Batteries with Micro-sized Silicon Anodes
Principal Investigator: Chunsheng Wang, University of Maryland

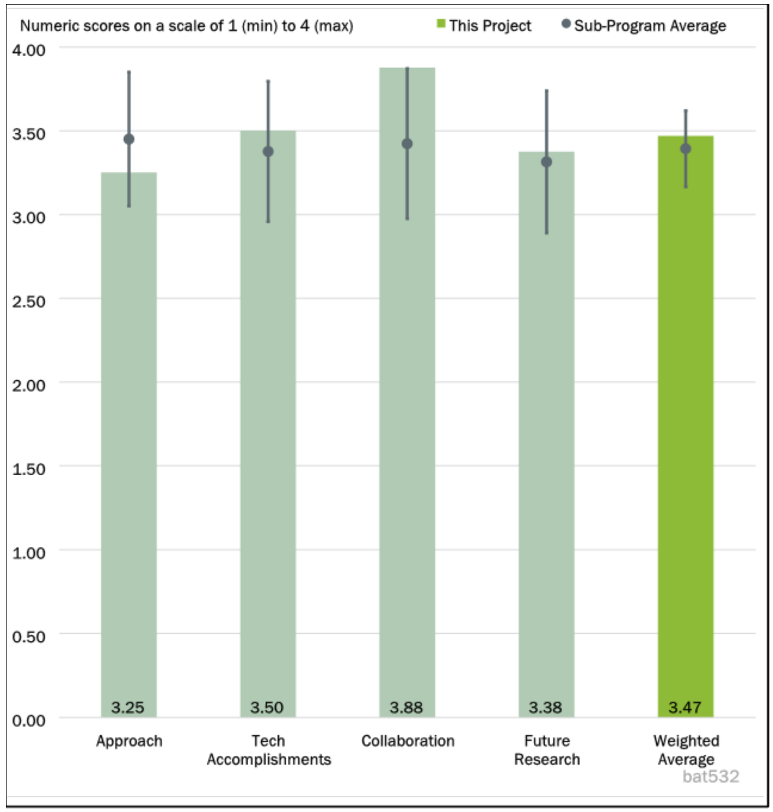

Figure 2-26 - Presentation Number: bat532 Presentation Title: Electrolytes with Lithium-ion Batteries with Micro-sized Silicon Anodes Principal Investigator: Chunsheng Wang, University of Maryland

Presenter

Chunsheng Wang, University of Maryland

Reviewer Sample Size

A total of four reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 100% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 0% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

Question 1: *Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?*

Reviewer 1

The reviewer mentioned that the project designs new electrolyte formulations that significantly improve the performance of silicon microparticles. The reviewer observed that uniquely, the project also breaks down the calendar life into measurable quantities, and that it is well-designed and addresses the technical barriers of silicon anode.

Reviewer 2

The reviewer suggested a quicker down selection of materials and electrolytes. The reviewer stated that a large amount of good work was completed, but more focused effort will lead to more meaningful accomplishments.

Reviewer 3

The reviewer claimed that the technical barrier for designing Electrolytes for Lithium-ion Batteries with Micro-sized Silicon Anodes is the following: finding electrolytes that will form a coherent shell around the entire micro-sized particle, while maintaining contact to the particle, not cracking, and not allowing fresh electrolyte to the particle surface. The reviewer explained that this team's approach is to try three different highly fluorinated electrolytes in an attempt to form an intact SEI film of mostly LiF around the micron-sized Si particles. The reviewer articulated that in doing so, the researcher will minimize the surface area of Si exposed to the electrolyte in all stages of formation and use. The reviewer noted that they make certain assumptions but do not actually test the assumptions. The reviewer stated that they try fluorinated electrolytes and get better results but this does not mean that their proposed mechanism is correct.

Reviewer 4

The reviewer expressed that there is a lot to unpack in this report and not a lot of details about materials and experiments. The reviewer observed that the units shown in most graphs, such as mAh/cm^2, make it difficult to estimate the gravimetric capacity or energy. The reviewer added that the basic premise is that LiF is weakly bonded to the Si, so able to form a dense, robust SEI skin that can slide to accommodate the stress from Li alloying. The reviewer claimed that assuming the liquid electrolyte does not fill the microcracks formed within the Si, the cracks can neatly close and heal when the Si is delithiated. The reviewer added that the polymer outer component of SEI is believed key to keeping the liquid out. The reviewer remarked that the approach to refining the liquid electrolytes is not evident, except that F is good. The reviewer stated that the rationale that moves the study from the THF to sulfolane to ionic liquids is not obvious, or perhaps, the approach is designed to be multiprong to quickly identify a success. The reviewer declared that while the program is set to address the Si electrode refinement as well as the electrolyte, except for comparison of cycling micro and nano Si particles, the core effort is towards the electrolyte formulation. The reviewer stated that the properties of the Si anode are not clear. The reviewer commented that while this is a new program under FOA, there has been significant earlier work that serves as the foundation, and that published work demonstrated improvements in cycle performance of 2M $LiPF_6$ in solvent mixtures such as pure THF and THF with methyl groups etc. The reviewer added that earlier work also revealed improvement from fluorinating the solvents, presumably still THF-based. The reviewer noted that almost all the results presented in the slides are electrochemical tests, with emphasis on obtaining the CE (coulombic efficiency), leakage current decay, and SEI resistance from partial fits of EIS. The reviewer added that 3D X-ray tomography and SEM are used to check for cracking in the Si anode, but otherwise reveal little about the SEI, and that there is no characterization of the SEI itself.

Question 2: Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.

Reviewer 1

The reviewer claimed that through three generations of electrolyte design, the project achieved superior performance compared to existing literature.

Reviewer 2

The reviewer encouraged the addition of actual multi-temperature calendar life testing. The reviewer added that it would be helpful to summarize together, for the same material and electrolyte composition, both the cycle and calendar life.

Reviewer 3

The reviewer mentioned that technical accomplishments include showing that starting from an all fluorinate electrolyte and replacing some of the solvent with sulfolane reduced the conductivity by 20% but improves the oxidative stability to above 5.3 V. The reviewer added that in a Si/Li cell, the team achieved 99.6% CE, in an lithium-iron phosphate (LFP)/Si cell, they achieved 99.9% CE, and in an NCA/SI cell they achieved 99.3% CE. The reviewer noted that the technical accomplishments also included showing that using an ionic liquid as the electrolyte, they could remove the solvent, achieve oxidative stability to 4.5 V and decent conductivity. The reviewer remarked that a cell of Si/Li achieved a CE of 99.9% and a cell of NCM811/Si achieved a CE of

99.9 %, although the capacity of the cell in mAh/cm^2 was not provided. The reviewer claimed that the project team also demonstrated that in a high-capacity Si/Li cell (4mAh/cm^2) they could achieve good capacity retention and a CE of 99.85% when cycled at C/10, although the capacity retention declined significantly at C/5. The reviewer added that in a full cell of NMC811/Si of a capacity of 4 mAh/cm^2 the project team achieved a capacity retention of 85% after 100 cycles at C/10. The reviewer added that microscopy analysis showed no cracking of the micro-sized Si particles after 300 cycles. The reviewer commented that in half cells they performed a test protocol that included cycling and a 180-hour voltage hold, so they could measure CE from the cycling and rate of side reactions from the voltage hold. The reviewer claimed that the project team found that the cell with macro-Si and the mix THF electrolyte lost the least amount of capacity during the hold and had a similar sustained current at the end of 180 hours as graphite with a typical electrolyte. The reviewer articulated that these two cells also had the best coulombic efficiency. The reviewer noted that the cell with micro-Si also performed better than nano Si with the same electrolyte. The reviewer added that the cell with ionic liquid had the worst loss of capacity during the hold but had the lowest rate of side reactions by the end of the 180 hours. The reviewer claimed that the worst performing cell was the micro-Si with the typical electrolyte for graphite cells. The reviewer added that the EIS of the Si half cells shows that the impedance rise with the mix THF electrolyte is less than the impedance rise of the typical graphite electrolyte. The reviewer stated that overall, the PI started out with the hypothesis that larger Si would cycle better if he could encapsulate the particles with a LiF based SEI. The reviewer noted that the project team then tested the larger Si in different electrolytes with higher fluorine content and showed better performance than the less fluorinated electrolyte and smaller Si. The reviewer remarked that the project team also achieved decent full cell cycling. It was unclear to the reviewer as to whether the project team is forming an intact SEI around the micro particles or that the electrolyte itself is not as reactive. The reviewer added that the project team's micro particles may be working well because the team is cycling at a slow rate. The reviewer concluded that the project team improved the performance of the Si electrode and full cell performance, which matters.

Reviewer 4

The reviewer stated that in the current program, accomplishment number 1, sulfolane was added to the fluorinated solvent (presumably THFs), which extends the voltage window above 5V. The reviewer explained that this provides for reasonable (although noisy) cycling to 4.3V of Si//NCA cell. The reviewer added that this was not emphasized, but appears promising. The reviewer added that the sulfolane-based electrolyte was called out for future research. The reviewer claimed that accomplishment number 2, is electrolyte formed with ionic liquid solvent providing good cycling of 811 versus Li anode cells, and Si versus Li half cells results as well. The reviewer stated that this provides for separating the CE of each electrode. The reviewer added that the full cell 811//Si cycled for over 100 cycles. The reviewer highlighted that after 300 cycles, there was no noticeable cracks in the micron-sized Si particles. The reviewer expressed that accomplishment number 3 addresses the calendar life with 180-hour high voltage aging. The reviewer detailed that for this, the work cycled back to the earliest electrolytes with mixed THF. The reviewer explained that the logic here confused the reviewer. The reviewer asked why the leakage current and EIS were measured and fitted, rather than simply comparing the energy and capacity of cycles before versus after the extended high voltage hold (in this case 180 hours). The reviewer rationalized that it was perhaps to identify aging over shorter time periods. The reviewer notes that the researchers observe the transient decrease in the current at the constant voltage hold, and the rise in the SEI resistance as extracted from the fits of results. The reviewer said that no attempt was made to quantify the uncertainty and the reviewer was skeptical of the accuracy implied by three or four significant figures. The reviewer that key for this analysis reveals that side-by-side, the micro-sized Si have better calendar life than do the nano-Si particles.

Question 3: Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?

Reviewer 1

The reviewer observed that the roles and responsibilities are clearly defined.

Reviewer 2

The reviewer noted that the recipient works with Brookhaven National lab to characterize the Si electrode after cycling, which is important to the success of the project.

Reviewer 3

The reviewer noted that this is a team of 4 different institutions that each have a particular role to play and that there is pretty good coordination in that respect.

Reviewer 4

The reviewer remarked that vital collaborators are Kang Xu and Oleg Boridin at Army Research Lab, providing fundamental and theoretical guidance for electrolyte formulations based on long experience. The reviewer noted that Saft is important for electrode coating and engagement as an industry partner. The reviewer observed that Stony Brook University provides synchrotron study of electrodes. The reviewer suggested that perhaps additional collaborations can provide needed ex- and in-situ characterizations of the SEI formation and aging.

Question 4: Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?

Reviewer 1

The reviewer stated that the project team wants to build a mathematical model that explains why large particles and F-rich SEI's leads to better performance. The reviewer added that the project team wants to optimize the formulation of its electrolyte with sulfolane for full cells. The reviewer observed that the project team wants to find a way to reduce the viscosity of their ionic liquid electrolyte. The reviewer commented that all of this is fine, but the project team never measured the calendar life of a full system. The reviewer added that before optimizing any electrolyte or modeling a system it would be nice to know if the project team is even close to a target of ten-year calendar life.

Reviewer 2

The reviewer stated that it would be useful to invest some time to evaluate the SEI formed on the Si in order to test the basic premise of this work. The reviewer added that the basic premise is that LiF is weakly bonded to the Si, so able to form a dense, robust SEI skin that can slide to accommodate the stress from Li alloying. The reviewer claimed that assuming the liquid electrolyte does not fill the microcracks formed within the Si, the cracks can heal when the Si is delithiated. The reviewer expressed that the polymer outer component of SEI is believed key to keep the liquid out. The reviewer suggested that instead of characterizing the SEI and its growth/dissolution with spectroscopic and physical analysis, the future work proposes modeling the degradation and relies on calendar life as measured by leakage current and EIS. The reviewer noted that this approach differs from other researchers. The reviewer declared that other future work will continue investigations and improvement of both the sulfolane and ionic liquid electrolytes. The reviewer concluded that there are likely opportunities to improve both of these electrolyte systems.

Reviewer 3

The reviewer recommended adding the actual calendar life testing to provide validation to the modeling exercise.

Reviewer 4

The reviewer noted that the presenter ran out of time, so the future work was not clearly described and no questions and answers were conducted. The reviewer added that based on the achievements, it is likely that the project will continue to deliver good results.

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

The reviewer stated that the project supports the VTO subprogram objectives in that it seeks to improve the performance of Si in cells which, if successful, should lead to higher energy density cells.

Reviewer 2

The reviewer expressed that this project supports the overall DOE objectives by extending the calendar life of silicon anode.

Reviewer 3

The reviewer claimed that the study of advanced electrolytes for high voltage cells utilizing Si anode is highly relevant for success of the VTO program. The reviewer added that it would be helpful if the investigation provided results in the same units as the specs written for the program, namely Wh/kg and Si content of the anode.

Reviewer 4

The reviewer remarked that the effort does support the goals of the VTO subprogram, and that the emphasis on micro-scale Si is especially relevant. The reviewer added that it is unclear if there are other significant barriers, such as cost, to the electrolyte systems under consideration. The reviewer commented that it would be good to at least acknowledge their potential impact.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer stated that VTO is suggested to increase funding for productive projects like this.

Reviewer 2

The reviewer claimed that the resources are sufficient in that it is $1 million spread over 2 years. The reviewer added that the University of Maryland only has to pay for some work at ARL, and that they get free work from Saft and Brookhaven.

Reviewer 3

The reviewer stated that they are sufficient for a new university lead experimental program.

Reviewer 4

The reviewer remarked that the effort appears well resourced.

Presentation Number: bat533
Presentation Title: Fluorinated Local High Concentration Electrolytes Enabling High Energy Density Silicon Anodes
Principal Investigator: Amy Marschilok, Stony Brook University

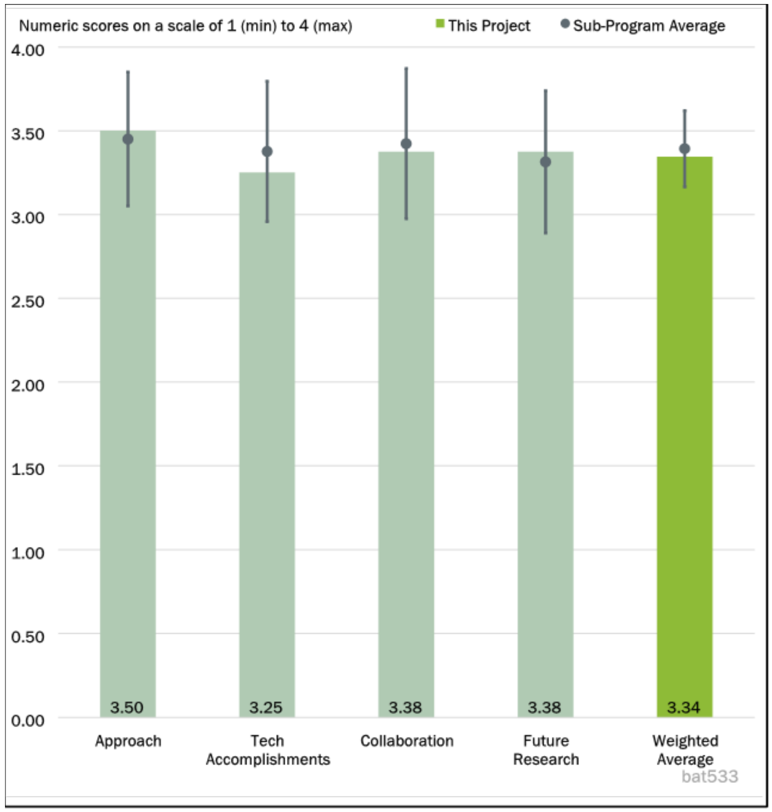

Figure 2-27 - Presentation Number: bat533 Presentation Title: Fluorinated Local High Concentration Electrolytes Enabling High Energy Density Silicon Anodes Principal Investigator: Amy Marschilok, Stony Brook University

Presenter

Amy Marschilok, Stony Brook University

Reviewer Sample Size

A total of four reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 100% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 0% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

Question 1: *Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?*

Reviewer 1

The reviewer commented that the technical barrier is developing an electrolyte that will allow for 10 years of calendar life. The reviewer stated that the approach is to design an electrolyte where there is fluorine in the anion and in the solvent. The reviewer mentioned that the project team will use a fluorinated localized high concentration electrolyte in this effort, and hope this will work. The reviewer declared that there have been indications that SEI films with high fluorine content passivate surfaces better than SEI's without fluorine. The reviewer claimed that the timeline is to try two sets of electrolytes and measure their progress.

Reviewer 2

The reviewer stated that this is a complex study with many factors that influence the success of a new electrolyte. The reviewer claimed that while the program is directed towards Si anodes, full cell tests dictate that performance must enhance both the composite anode and high Ni (622) cathode. The reviewer mentioned that earlier studies by groups investigating similar LHCE used the lower voltage 532 and 333 cathodes. The reviewer added that while this study is supporting the effort to stabilize the Si anode, the cathode chosen itself presents significant challenges as it has a higher Ni content (622) than earlier studies with LHCE local high concentrated electrolyte contain non-solvating diluent. The reviewer remarked that the initial approach used a control of $LiPF_6$ EC/ dimethyl carbonate (DMC), but this is well known to be ineffective in Si cells. The reviewer observed that the capacity loss is following a very different rate dependence for this control. The reviewer stated that the research team wisely switched to comparison of their novel electrolytes to the 1M electrolytes with FEC addition. The reviewer concluded that this is a better "control" or reference for the current fluorinated localized high-concentration electrolyte (FLHCE) study because the work is more likely to reveal the effects of diluent and fluoridated cosolvents.

Reviewer 3

The reviewer remarked that the approach is well thought out and systematic

Reviewer 4

The reviewer commented that the project evaluated several electrolyte solvent molecules to address the technical barriers of Si anode, and that it will be more effective when directed by rational design.

Question 2: Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.

Reviewer 1

The reviewer expressed that at this meeting the project team presented progress on their first-generation electrolyte, which consists of four variations of LiFSI, FEC, FEMC, and TTE (all materials are fluorinated). The reviewer added that when compared to EC:EMC, they all have higher viscosity and lower conductivity. The reviewer mentioned that amongst themselves, the order in conductivity and viscosity is the same order as the concentration of LiFSI - high LFSI correlates to high conductivity and viscosity. The reviewer claimed that the contact angle of these electrolytes shows better wetting on the separator than the typical Li-ion electrolyte shows. The reviewer explained that these FLCHEs show least voltage stability but are good enough for most Li-ion cathodes and the LiFSI does not show aluminum corrosion as it does in EC:EMC. The reviewer remarked that the capacity fade of the full cells made with the FLHCE all show about the same rate. The reviewer claimed that there are differences in initial capacity that can be attributed to differences in impedance. The reviewer observed that with regard to impedance, the control electrolyte starts with less impedance than the FLHCEs, but this rapidly changes with cycling with the control growing much worse. The reviewer stated that the PI also introduced the second-generation electrolyte. The reviewer clarified that this contains similar solvents as the first generation but with the replacement of TTE with BTFE and TFEPE. The reviewer added that the next set of electrolytes have a wider range of viscosity than the first generation (in general, slightly higher), and about the same contact angle as the control (less wetting than the first generation). The reviewer concluded that in general, the first-generation electrolytes (FLHCEs) were better than the control (EC:EMC), and that there is no clear indication that the second generation will be better that the first.

Reviewer 2

The reviewer remarked that it is still early as this is a new Si anode FOA program. The reviewer noted that significant progress was demonstrated in exploring new liquid electrolytes. The reviewer added that while following the lead from other research teams, notably Wu and Zhang at PNNL, these compositions are new and Gen 1 compositions already demonstrate a slight performance exceeding that of standard electrolytes with the FEC additive. The reviewer remarked that much is being studied and reported about the electrolytes, including viscosity, conductivity, and separator wetting, as well as the electrochemical stability and performance in full Si/NMC cell and various reference cells with graphite and LFP electrodes. The reviewer expressed that it is exciting to see isothermal microcalorimetry used for this study. The reviewer stated that a wise comparison was made at the 15th cycle control, so that remaining capacity is comparable. The reviewer wondered if comparing X-ray photoelectron spectroscopy and EIS for comparable capacity, 80%, would be formative than at a specific cycle number particularly for controls near zero capacity. The reviewer had questions for the PI, but time ran out. The reviewer asked the following: How much liquid was used in each cell? Can volume of mixing when diluent be used to learn about ion association and solvation? Why not start with maximum 4M salt solution? The reviewer added that the table on Slide 6 of Gen 1 electrolytes indicates that salt concentration is significantly higher for number four than for number two, yet text suggests that the two are comparable and number two is highlighted.

Reviewer 3

The reviewer said that the work completed so far is systematic and consistent, and looks forward to data from year 2 and 3 goals which will tie together answers to the key questions.

Reviewer 4

The reviewer claimed that performance improvement is shown, but the absolute performance is still not as good as the best literature values. The reviewer explained that for example, the percentage of Si is only 30% (the minimum of DOE target), and cycle life is only 100 cycles (much lower than DOE target).

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer stated that there is a strong collaboration with BNL but nothing more. The reviewer added that the project team should find a way to work with the Si consortium and share electrodes and electrolytes.

Reviewer 2

The reviewer noted that collaborations are limited to BNL and Stony Brook University, which is sufficient at this stage in new FOA. The reviewer added that the source of cell test electrodes is not clear. The reviewer explained that if made at Stony Brook University, it may be useful to coordinate with CAMP or industrial electrode fabrication. The reviewer stated that collaborations to view the SEI and CEI evolution with voltage and cycle age may be achieved most efficiently with aid of another program.

Reviewer 3

The reviewer noted that it was very clear who is doing what and what partner skills and resources are being brought to the table.

Reviewer 4

The reviewer stated that the recipient works with Brookhaven National Lab. The reviewer added that the project would be more productive if one other university collaborator with extensive and complementary expertise on Si anode is involved.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer remarked that the future work involves characterizing the Gen 2 electrolyte to the same extent the Gen 1 was. The reviewer stated that the project team also wants to perform more isocalorimetry and that their goal is to demonstrate 20% capacity improvement over EC:EMC at cycle 100, but the EC:EMC only maintains 10% of its original capacity at 100 cycles, so this is not a great accomplishment. The reviewer declared that there is no guarantee that Gen 2 will be any better than Gen 1. The reviewer observed that the project team also expects to demonstrate a 50% capacity retention improvement at 500 cycles, which is essentially a 50% capacity retention in total. The reviewer recommended that there needs to be more of an analysis between Gen 1 and Gen 2 and what else is going on with Si to move to a better electrolyte. The reviewer said that it is possible that a particular formulation will hit a home run, but it is not apparent in any of the presentations at this AMR.

Reviewer 2

The reviewer declared that yes indeed, future research is well defined and likely to be successful. The reviewer added that it is important to reveal scientific understanding as well as achieving capacity retention. The reviewer noted that future work will emphasize alternative diluents, both different molecules and different concentrations, and that this is the most important and interesting part of study. The reviewer added that the PI plans to assess CEI formation and variation with each deep cycle. The reviewer observed that the premise is that the thin inorganic SEI at the Si interface (LiF + Li oxide is more stable than a composite SEI with organics. The reviewer explained that whether it is expected to rapidly grow and shrink with Si cycling, or rapidly passivate any microcrack formation, or both, is not clear from the presentation. The reviewer suggested that the observation of the SEI (and CEI) variation during a full cycle, or at least at full charge and full discharge states, would be enlightening, but also potentially rather complicated study for Si + C composite. The reviewer stated that to achieve best practical cycling performance, additional effort will be productive following the PI's intuition to with salt compositions and additives. The reviewer added that study of calendar life will be initiated in future work and that it will use the prescribed method of applying an extended hold at high V and noting small leakage current. The reviewer concluded that while this is a simple means to isolate side reactions that occur at higher V, demonstrating its relevance for the Si SEI formation may be ambiguous and the PI may have freedom to adjust such a test.

Reviewer 3

The reviewer encouraged calendar life tests to include actual, multi-temperature calendar life tests, in addition to voltage hold tests.

Reviewer 4

The reviewer commented that the goal of improving the performance was described, and that it would be more convincing if hypothesis is clearly laid out.

Question 5: Relevance: Does the project support the overall VTO subprogram objectives?

Reviewer 1

The reviewer said that this work supports the overall VTO subprogram objectives to increase energy density by finding ways to get experimental electrode materials to perform better.

Reviewer 2

The reviewer stated that this program is highly relevant to achieving required cycle life, calendar life, voltage stability needed to achieve the higher energy density of Si anodes as substitution for graphite anode in Li-ion batteries. The reviewer added that the best electrolyte formulation considering cost, safety in addition to cycling performance warrants additional study under investigation in this program.

Reviewer 3

The reviewer expressed that this is highly relevant to the study and advancement of Si anodes; however, some acknowledgment of other barriers, such as cost, environmental impact, etc. would be helpful.

Reviewer 4

The reviewer claimed that this project supports the overall DOE objectives by extending the calendar life of silicon anode.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer stated that the resources are sufficient to support the work proposed.

Reviewer 2

The reviewer expressed that they are sufficient for new university led experimental programs.

Reviewer 3

The reviewer claimed that effort appears well resourced and funded.

Reviewer 4

The reviewer said that some aspects of the project (e.g., combining electrolyte with engineered silicon anode materials) would benefit from working with another university.

Presentation Number: bat534
Presentation Title: Devising mechanically compliant and chemically stable synthetic solid-electrolyte interphases on silicon
Principal Investigator: Pierre Yao, University of Delaware

Presenter

Pierre Yao, University of Delaware

Reviewer Sample Size

A total of four reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 75% of reviewers felt that the resources were sufficient, 25% of reviewers felt that the resources were insufficient, 0% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

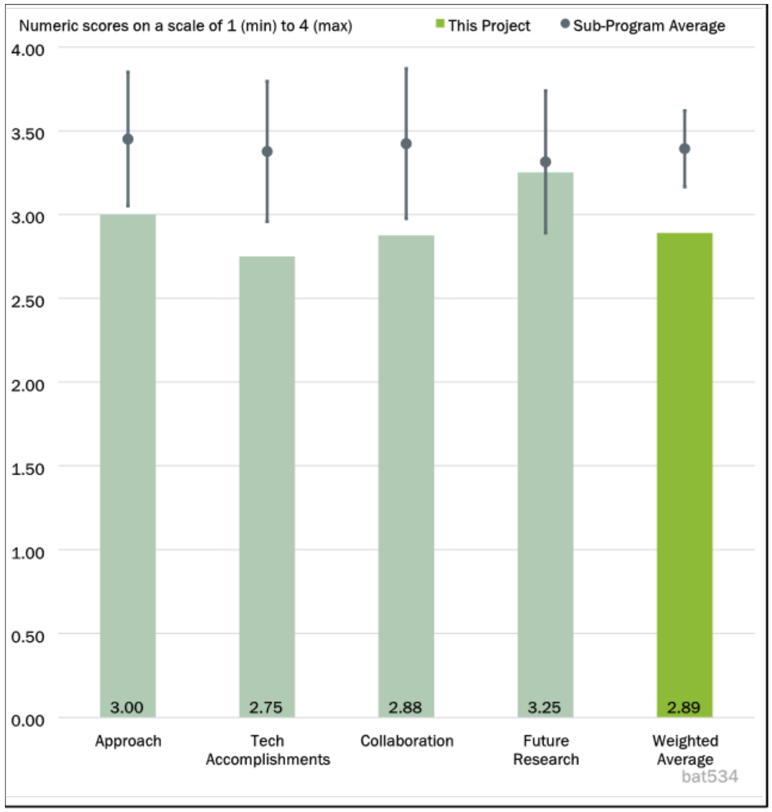

Figure 2-28 - Presentation Number: bat534 Presentation Title: Devising mechanically compliant and chemically stable synthetic solid-electrolyte interphases on silicon Principal Investigator: Pierre Yao, University of Delaware

Question 1: Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?

Reviewer 1

The reviewer stated that the investigators want to apply a polymeric/elastomeric artificial SEI on the surface of Si to passivate the surface, and that they want to apply it either through electropolymerization or electrodeposition. The reviewer added that they are going to test a number of polymers for solvent stability and surface conformity, and expect to identify at least one conformal coating by the time of this review. The reviewer claimed that they then expect to produce a number of protected Si electrodes and test them in full cells against an NMC cathode. The reviewer concluded that their plan is straight forward and reasonable.

Reviewer 2

The reviewer observed that the approach is simple and attractive: Synthesize and coat Si anode ex-situ to allow for greater choice and control of the artificial SEI than can be achieved in the actual battery. The reviewer noted that with an electrodeposition solution coating process, there is an opportunity to conformally coat even porous Si electrodes. The reviewer added that early results for this new program showed promise but are very preliminary. The reviewer stated that the approach used both: electropolymerization EP, where monomers are polymerized on surface; and electrophoretic deposition (EPD), from solutions already containing polymers of interest. The reviewer observed that as these methods are not self-limiting, the synthetic SEI thickness can be adjusted with the deposition time. The reviewer articulated that Si electrode samples were fabricated as thin film, 40 nm thick, on copper foil which provided a convenient electrode for the synthetic SEI deposition and allowed for good characterization of the surface coating by FTIR and AFM methods. The reviewer said that subsequently the SEI-coated Si film was tested in coin cell versus Li with standard LiPF6-EC+DMC electrolyte. The reviewer suggested that perhaps a better electrolyte would include the FEC additive. The reviewer added that it appeared that there was some contamination or other non-uniformity of the Si thin film that influenced the conformity of the polymer coatings. The reviewer remarked that cast particle electrodes (presumably from CAMP) with nanoparticles of Si with carbon and polyvinylidene difluoride (PVDF) binder (PAA was not stable in the EPD solution) were also used for Si anodes for coating and cycling. The reviewer concluded that after coating and cycling, the composite electrode delaminated from the metal foil, so this was not a good test of the chitosan SEI.

Reviewer 3

The reviewer liked the approach of separating the SEI formation outside the formation of the anode itself.

Reviewer 4

The reviewer expressed that the idea of artificial SEI has been extensively tested in literature, and that the approach in this project is not an advance to what has been tested, and the hypothesis is not well-described.

*Question 2: **Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.***

Reviewer 1

The reviewer expressed that to evaluate an electropolymerization approach, the project team produced a model surface through deposition of a thin film of Si onto copper foil. The reviewer added that the project team decided to try THF as the monomer as Li-TFSI salt dissolves in THF. The reviewer explained that they developed a beaker cell for depositing polymers onto the SI film and a number of instruments to characterize the film once deposited, including ATR-FTIR spectroscopy, AFM nano IR spectroscopy, and Thermo-Fischer. The reviewer explained that they tried electropolymerization of styrene to polystyrene and found that it was not uniformly deposited on the Si film. The reviewer mentioned that they tried electropolymerization of polythiophene and found it uniformly deposited onto stainless steel but not silicon. The reviewer described that they tried electrophoretic deposition of chitosan and found that they could uniformly deposit it on a film of Si. The reviewer claimed that they showed they could get more lithium into a Si thin film when chitosan was deposited on its surface, and that the impedance rise of the Si surface was less after cycling when coated with chitosan. The reviewer added that they then moved to a composite Si electrode with 70% Si (Paraclete), 15% carbon additive and 15% binder and found improved cycling as the amount of electrophoretic deposition of chitosan increased. The reviewer concluded that these accomplishments indicate steady progress.

Reviewer 2

The reviewer noted that using homemade Si film, there were problems achieving a conformal coating of either polystyrene or polythiophene, and that better EP films were achieved on stainless steel substrates. The reviewer added that characterizations of the Si film properties, such as crystallinity and surface contamination, were not presented, and that likely the Si is amorphous. The reviewer observed that the best coating to date was EPD of chitosan, about 2um thick after 1 minute at 2V EPD onto the 40nm Si film coated copper. The reviewer said that this chitosan SEI coating did not block the Li cycling of the Si any more than for the natural SEI formed at the bare Si film, and that both were highly resistive. The reviewer detailed that the observation that the chitosan coating approximately tripled the first cycle capacity loss compared to the uncoated Si anode is concerning as prelithiation will likely be needed and should be investigated at an early opportunity. The reviewer mentioned that chitosan coating was also achieved on nano-silicon 70% composite electrodes, and that this chitosan coated Si particle electrode showed rapid capacity degradation after only 4 cycles in Li half-cell assembled in coin cell. The reviewer added that thicker chitosan coatings did not improve this as much as one might expect, and the whole anode coating spalled from the foil current collector. The reviewer commented that alternative fabrication is needed for the Si particulate anode or alternatively, porous Si anodes are called for and noted in the future research directions. The reviewer stated that the first go/no-go milestone was to demonstrate 50% strain and that it would be good to see this result and the testing method. The reviewer claimed that this is an important advantage of the elastomer coating over the inorganic SEI formation, and that the elastomer properties should be measured before and after any prelithiation process.

Reviewer 3

The reviewer suggested that a radically different approach like this clearly has more barriers to overcome. The reviewer posed a few key questions:

- Requiring a PVDF binder is a major issue that is incompatible with the thrust of research and commercial application.
- It is not clear what the deposition uniformity is on a porous electrode.
- Does the chitosan material have the necessary mechanical properties to withstand repeated expansion and contraction?

Reviewer 4

The reviewer noted that the initial idea was not successful and that the performance is far from the target. The reviewer questioned the timely completion of the project.

Question 3: ***Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?***

Reviewer 1

The reviewer said that their source for electrodes is Argonne, and that they also consult with Argonne about Si in general.

Reviewer 2

The reviewer declared that collaboration with CAMP and ANL for electrodes and guidance on cell fabrication is useful, but given the different conditions required to form the SEI by EPD and EP, the anodes require significant modification, such as for the binder and porosity. The reviewer mentioned that other project teams may be able to provide alternative or more suitable anodes.

Reviewer 3

The reviewer noted that this is a multi-disciplinary effort that may be aided by collaboration from other entities. The reviewer asked if there is sufficient knowledge and skill within the team to develop an electrophoretic deposition process, or if there is opportunity to enhance collaboration and accomplishments by bringing in talent from other areas where electrophoretic deposition is routine done, such as paints and corrosion coatings.

Reviewer 4

The reviewer noted that the collaboration with Argonne National Lab seems to be ineffective. The reviewer explained that the team had a hard time to reach the baseline performance and appears to lack technical expertise on Si anode.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer commented that in the future they will try their process on electrodes from Argonne based on P84 binder, which has shown some success in the Si Consortium Project. The reviewer mentioned that they hope to pretest in coin cells and that depending on the results, they intend to test other coatings applied by electrophoretic deposition. The reviewer concluded that it is a straight forward and clear plan.

Reviewer 2

The reviewer suggested that future research will continue on the same pathway, as a number of issues need to be resolved to determine if this approach can offer success to form a stabilized Si anode with a synthetic SEI coating. The reviewer was not overly optimistic that this will prove successful, as most research activities have determined that the inorganic SEI coatings provide better protection to stabilize the Si anode. The reviewer noted that elastomeric coating make sense if they passivate the surface and do not grow to a thick and resistive SEI.

Reviewer 3

The reviewer encouraged incorporation of some of the above comments, and added that it would be good to obtain impact of chitosan material and thickness on first cycle efficiency of the system.

Reviewer 4

The reviewer declared that some of the future plans are good (e.g., replacing PVDF with advanced binder), but there is still concern on the capability of the team to achieve comparable baseline performance as in the community.

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

The reviewer stated that the program is very relevant to VTO objectives as it seeks new SEI films for Si, and that this will be needed to get Si to work as an anode in Li-ion cells.

Reviewer 2

The reviewer declared that while a long way from coming close to the goals of the Si battery program, this program offers an alternative approach to depositing the SEI coating. The reviewer noted that others have similarly tried to prepare electrodes (Li, graphite, Si) with ex-situ or ‘synthetic’ SEI coatings materials and methods, but had limited success. The reviewer said that it is a challenging undertaking; nevertheless, this approach is definitely worth revisiting.

Reviewer 3

The reviewer observed that this type of radically different approach to solving difficult problems is highly appreciated and encouraged.

Reviewer 4

The reviewer noted that while having a lot of difficulties in execution, the goal of this project supports the overall DOE objectives by extending the calendar life of silicon anode.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer said that the PI appears to be making good progress with the present resources.

Reviewer 2

The reviewer noted that resources were sufficient for new university led experimental program.

Reviewer 3

The reviewer asked to please see comment for question 6.

Reviewer 4

The reviewer commented that the team would benefit from having another university partner, to tap into expertise on Si anode without using too much budget.

Presentation Number: bat553
Presentation Title: Understanding solid electrolyte interphase (SEI) reactions in Lithium metal and Lithium-Sulfur batteries
Principal Investigator: Perla Balbuena, Texas A&M University

Presenter

Perla Balbuena, Texas A&M University

Reviewer Sample Size

A total of four reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 100% of reviewers felt that the resources were sufficient, 0% of reviewers felt that the resources were insufficient, 0% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

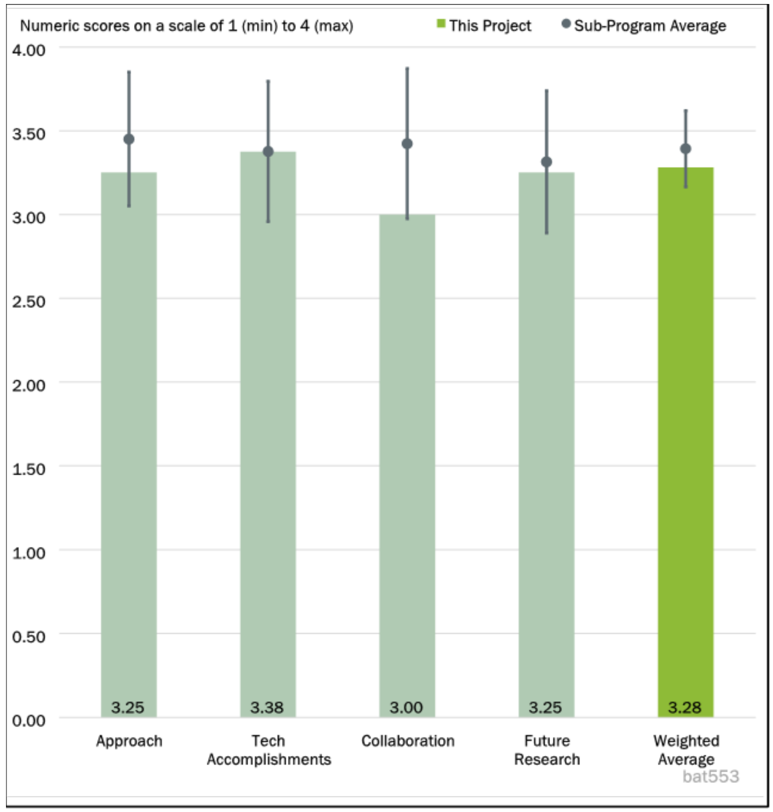

Figure 2-29 - Presentation Number: bat553 Presentation Title: Understanding solid electrolyte interphase (SEI) reactions in Lithium metal and Lithium-Sulfur batteries Principal Investigator: Perla Balbuena, Texas A&M University

Question 1: Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?

Reviewer 1

The reviewer stated that this work is oriented towards fundamental understanding of the chemistry, charge transfer, and other processes at the SEI layer. The reviewer claimed that such fundamental understanding may be important for achieving the ultimate Battery500 goals, and is generally helpful to help understand limitations on system performance, why a system degrades, etc. The reviewer added that one area for improvement in the approach would be to have specific experimental observations to be explained and orient at least some of the modeling work around capturing those observations. The reviewer explained that this could be a charge transfer resistance, a capacitance, a composition or SEI thickness, etc. The reviewer concluded that the work does a nice job of presenting quantitative results, but the connection to experiment is not as clear.

Reviewer 2

The reviewer noted that MD simulation is an important part of the battery research project, especially, if its results can be verified by experiment and used to guide cell developments.

Reviewer 3

The reviewer claimed that the approach to the work is appropriate and the research for the planned milestones is well designed. The researcher articulated that to understand the reactions near and in the solid electrolyte interface (SEI), the researchers have carried out ab initio molecular dynamics and theoretical modeling to identify and characterize the key reaction steps at the atomic level. The reviewer added that they are also conducting simulations to unravel relevant effects in the SEI region, including the salt concentration, solvent chemistry, SEI morphology, ion transport and external pressure. The reviewer claimed that in the technical aspect, they have adopted the "Blue Moon" ensemble to correct the calculated free energy from the constrained molecular dynamics simulations. The reviewer stated that the results from these studies can help to understand and possibly guide experiments.

Reviewer 4

The reviewer observed that the project is well designed and has been reasonably planned.

Question 2: *Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.*

Reviewer 1

The reviewer observed that there are a significant number of new results, and with a good level of detail, and in different areas, including for example the work on pressure effects on reactions. The reviewer claimed that this group is working with the materials from the Battery500 consortium, and is being productive in looking at key issues associated with SEI processes.

Reviewer 2

The reviewer stated that the team has conducted extensive simulation works on interfacial resistance, SEI morphology, ion transportation, and external pressure effects. The reviewer didn't see any attempts to compare the results to experiments or to explain existing experimental results.

Reviewer 3

The reviewer noted that the current progress is fit, and that the researchers have completed the milestone of Q1. The reviewer added that the key steps and the barriers at the interface region have been identified and characterized using constrained ab initio molecular dynamics simulations and density functional theory calculations. The reviewer declared that they have also made substantial progress to reach the planned targets for Q2-Q4.

Reviewer 4

The reviewer declared that the work done is an early phase of the project, and that the PI should emphasize more on what are products of SEI. The reviewer stated that the pressure effect on reactions at porous interphasial structures characterized are not clear and asked how the ionic conductivity across the interface is.

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer said that it is clear this team is modeling materials of interest to Battery500, but it is hard to tell if there are significant direct collaborations with other modeling groups in Battery500, or experimental groups.

The reviewer recommended that some more direct touch points with experimental groups could help focus this work.

Reviewer 2

The reviewer stated that the team has listed collaborations with experimental teams from universities and national labs, and hopes to see works combining the simulation with the experiment reported in their next annual review.

Reviewer 3

The reviewer observed that in the current progress report, the comparison between the theoretical results and the experimental ones is lacking. The reviewer explained that since more experimentally related factors are under study, the researchers could show more modeling-experiment combined results in the future.

Reviewer 4

The reviewer made no comment on this.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer said that the proposed future research would benefit from using at least a couple of SMART milestones. The reviewer noted that, as written, there are goals such as “study”, “investigate”, “characterize” but no specific and quantitative goals that could help a reviewer assess the progress in the work.

Reviewer 2

The reviewer commented that the proposed c-AIMD works are interesting and are concentrated in SEI structures. The reviewer recommended that the simulation work be combined with the experiment, or even better, to provide clues to guide better cell design.

Reviewer 3

The reviewer stated that the project has clearly identified the undergoing and the future work, and that the currently planned research steps are reasonable and can serve well for the targets.

Reviewer 4

The reviewer noted that the project has clearly defined a purpose for future work.

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

The reviewer stated that the work is relevant to Battery500, which is clearly relevant to the VTO goals.

Reviewer 2

The reviewer said the simulation is an important part of the battery research project and can contribute in general to the VTO objectives.

Reviewer 3

The reviewer commented that the studies can support the VTO subprogram objectives in the areas including Batteries and Analysis.

Reviewer 4

The reviewer noted that the project does support the VTO Battery500 program.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer said that Battery500 resources are appropriate for such a large effort with different types of work (from fundamental to cell building).

Reviewer 2

The reviewer stated that the team has sufficient resource for their project.

Reviewer 3

The reviewer noted that according to the current progress made, the resources are sufficient for the project. The reviewer added that the researchers are simulating large systems and they are planning to use classical force fields generated by ab initio density functional calculations to complete the tasks. The reviewer said that these calculations are affordable with the current computing resources.

Reviewer 4

The reviewer commented that the resources of the project are sufficient to achieve the project milestones.

Presentation Number: bat554
Presentation Title: Fabricate and Test Solid-State Ceramic Electrolytes and Electrolyte/Cathode Laminates
Principal Investigator: Mike Tucker, Lawrence Berkeley National Laboratory

Presenter

Mike Tucker, LBNL

Reviewer Sample Size

A total of three reviewers evaluated this project.

Project Relevance and Resources

100% of reviewers felt that the project was relevant to current DOE objectives, 0% of reviewers felt that the project was not relevant, and 0% of reviewers did not indicate an answer. 67% of reviewers felt that the resources were sufficient, 33% of reviewers felt that the resources were insufficient, 0% of reviewers felt that the resources were excessive, and 0% of reviewers did not indicate an answer.

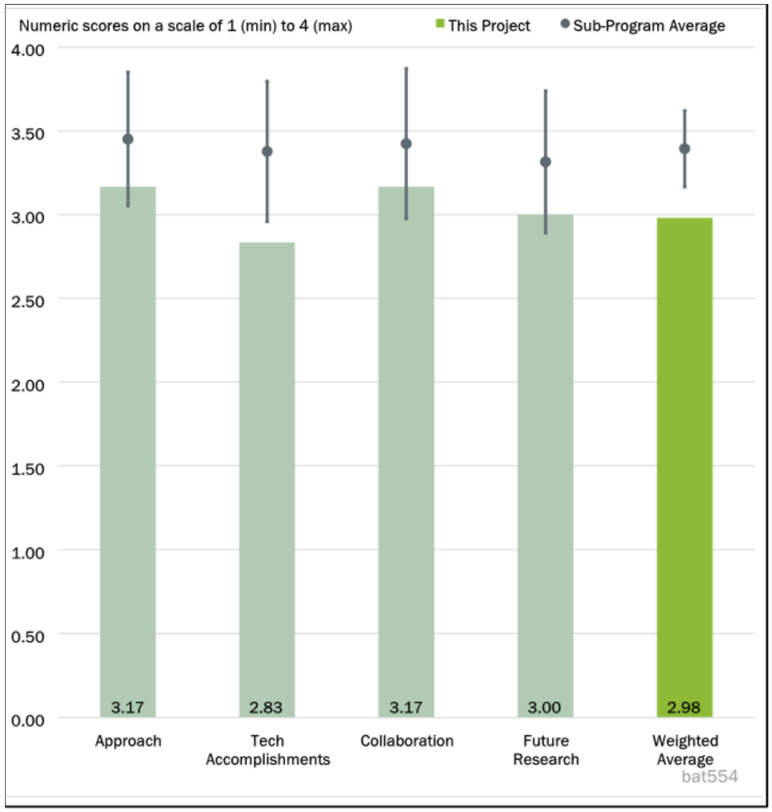

Figure 2-30 - Presentation Number: bat554 Presentation Title: Fabricate and Test Solid-State Ceramic Electrolytes and Electrolyte/Cathode Laminates Principal Investigator: Mike Tucker, Lawrence Berkeley National Laboratory

Question 1: Approach to Performing the Work: Is the project well designed, and is the timeline reasonably planned?

Reviewer 1

The reviewer stated that this is an interesting approach, similar to what Eric Wachsman at the University of Maryland has worked on. The reviewer added that the goal of making a thin electrolyte and thick cathode is important and clearly a focus in the work, and that the approach leverages the experience of the PI with ceramics processing.

Reviewer 2

The reviewer remarked that the focus of this project is to develop scalable methods to form thin sheets of lithium lanthanum zirconium oxide (LLZO), interfaced with a porous scaffold that will be infiltrated with cathode active material. The reviewer added that the advantage of this approach, compared to pre-mixing cathode particles with LLZO before sintering, is that the sintering temperatures required are too high, and result in reactivity with the cathodes. The reviewer claimed that a disadvantage/challenge of this approach is the difficulty in achieving infiltration of the porous scaffold, which has not yet been achieved. The reviewer said that other novel aspect of this work is the incorporation of MgO particles to control LLZO grain size, and the introduction of an organic cathode that should be easier to infiltrate. The reviewer expressed that the project is well designed with a reasonable timeline and that the remaining technical barriers that have not yet been fully addressed include demonstrating the effectiveness of the infiltration of the cathode, measuring CCD, and improving energy density and rate capability to technologically-relevant values. The reviewer mentioned another barrier that remains to be addressed is improving the fundamental understanding of why the observed capacity fade occurs, which is not fully understood.

Reviewer 3

The reviewer expressed that the overall project technical barriers were not presented. The reviewer believed the poster template format did not include the overall project timeline; however, the technical barriers are somewhat addressed. The reviewer added that the dendrite formation did not appear to be addressed. The reviewer commented that the LLZO scaffold content was studied by means engineering porosity; however, achieving high (over 70%) porosity may be challenging while maintaining adequate mechanical integrity to prevent disintegration during operation.

Question 2: *Technical Accomplishments and Progress: Comments on the technical progress that has been made compared to project plan.*

Reviewer 1

The reviewer stated that the goal of making and demonstrating a full cell in 3.5 years with $1M budget is a really challenging one. The reviewer added that the team appears to have made some progress, and does show some cell cycling results, but it looks like a liquid catholyte was used for some tests (which wouldn't meet VTO goals), and there are some missing details for other cell tests (e.g., the one with an all-solid-state cell, what is the loading?). The reviewer claimed that as the project is ending, it will be important for the authors to publish on the challenges they faced, and "lessons learned" from what they attempted in this project.

Reviewer 2

The reviewer noted that the progress on ceramic manufacturing, including the dense and porous microstructures is on track and successful. The reviewer remarked that a noteworthy accomplishment is the control of grain size through introduction of MgO. The reviewer added that full cell development, especially with regards to achieving high energy density and good rate capability, appears to be a bit behind schedule, but is the focus of the remaining two quarters.

Reviewer 3

The reviewer said that based on the project timeline for FY 2022, most of the objectives were satisfactorily met. The reviewer clarified that for example, the FY 2022 Q1 and Q2 plans were to fabricate LLZO scaffolds and infiltrate them with a liquid cathode, and it was achieved. The reviewer added that the Q3 and Q4 milestones were not due at the time the slides were submitted.

Question 3: *Collaboration and Coordination Across Project Team: Are there specific contributions made by industry, national laboratories, or other external entities? Are there areas where more collaboration is needed?*

Reviewer 1

The reviewer stated that Collaboration with two companies is mentioned, although the significance of that work is hard to assess. The reviewer added that given the limited size of the budget for this project, having it mostly happen within LBNL is appropriate.

Reviewer 2

The reviewer noted that the team has identified multiple collaborators, including a company that is supplying sub-micron sized NMC powders that should be more compatible with infiltration into the porous structure.

Reviewer 3

The reviewer declared that the project team appears to be effectively collaborating and Ragan is providing shear compaction expertise.

Question 4: *Proposed Future Research: Has the project clearly defined a purpose for future work? To what extent will future work likely achieve its targets?*

Reviewer 1

The reviewer commented that the proposed work is focused on making cells and testing them. The reviewer added that presumably the authors had a goal of doing more of this during the project, which ends soon. The reviewer declared that is future work is desired here, additional development work is likely needed before committing resources to full cell evaluation is appropriate, and gave an example of working on making a solid-state cathode with the LLZO scaffold.

Reviewer 2

The reviewer noted that the next steps are to achieve full infiltration of the cathode, achieve high ionic and electronic conductivity of the cathode (the current organic cathode suffers from low electronic conductivity), and achieve the final targeted dimensions (membrane thickness and high porosity cathode scaffold, with desirable mechanical properties. The reviewer expressed that it will be challenging to achieve all of these metrics, but the focus on the cathode is a good one, and it is likely that meaningful progress will be made.

Reviewer 3

The reviewer stated that the planned future works is reasonable; however, since the cycling of the Li anode was not described, it is not possible to assess the likelihood of full cell cycling success.

Question 5: *Relevance: Does the project support the overall VTO subprogram objectives?*

Reviewer 1

The reviewer stated that the goal is to make a high-energy, solid-state cell, and this is relevant to VTO.

Reviewer 2

The reviewer commented that this is a valuable project to the VTO portfolio, providing process knowledge for solid-state batteries. It was unclear to the reviewer how likely it will be to achieve high energy density and rate capabilities (in line with DOE targets) within the timeframe of the remaining project.

Reviewer 3

The reviewer was not able to provide a yes/no answer as there was insufficient information. The reviewer explained that if the cell performance characteristics were disclosed, it would help provide a more definitive answer. The reviewer noted that ceramic electrolytes are heavier and more expensive than liquid electrolytes; thus, replacing liquid with ceramic electrolytes, especially in anode and cathode, will make it difficult to achieve Wh/kg metrics relevant to VTO. This is the reviewer’s assessment based on the information provided in the poster.

Question 6: *Resources: Are the resources sufficient for the project to achieve the stated milestones in a timely fashion?*

Reviewer 1

The reviewer suggested that the resources are sufficient to achieve the project milestones.

Reviewer 2

The reviewer claimed that the resources appear to be sufficient.

Reviewer 3

The reviewer stated that this project would have made more progress with a higher budget. The reviewer added that going from a cell architecture concept to a prototype with $1M is a real challenge, and more resources would likely have led to more progress.

Acronyms and Abbreviations

3-D	Three-dimensional
Ah	Ampere-hour
Al	Aluminum
ANL	Argonne National Laboratory
APS	Advanced Photon Source
ARL	U.S. Army Research Laboratory
BNL	Brookhaven National Laboratory
BTFE	bis(2,2,2-trifluoroethyl) ether
CAM	Cathode active material
CAMP	Cell Analysis, Modeling, and Prototyping Facility
CE	Coulombic efficiency
CEI	Cathode-electrolyte interface
C-F	Carbon-fluorine bond
CFM	Complex framework materials
CIP	Contact ion pairs
cm	Centimeter
Co	Cobalt
CO_2	Carbon dioxide
COVID-19	Coronavirus disease 2019
Cu	Copper
DEMS	Differential Electrochemical mass spectroscopy
DFT	Density functional theory
DMC	Dimethyl carbonate
DOE	U.S. Department of Energy
EC	Ethylene carbonate
EERE	Office of Energy Efficiency and Renewable Energy
EIS	Electrochemical impedance spectroscopy
EPD	Electrophoretic deposition
EV	Electric vehicle
FEC	Fluoroethylene carbonate
FEC	Functional electrocatalysts

FEMC	Methyl 2,2,2-trifluoroethyl carbonate
FLHCE	Fluorinated localized high-concentration electrolyte
FY	Fiscal year
g	gram
GBA	γ-butyrolactone
GHG	Greenhouse gas
GM	General Motors
HRTEM	High-resolution transmission electron microscopy
INL	*Idaho National Laboratory*
Kg	Kilogram
kWh	Kilowatt-hour
LBNL	Lawrence Berkeley National Laboratory
LFP	Lithium iron phosphate
LHCE	Localized high-concentration electrolyte
Li	Lithium
LiF	Lithium fluoride
LiFSI	Lithium bis(fluorosulfonyl)imide
$LiPF_6$	Lithium hexafluorophosphate
Li-S	Lithium-sulfur
Li-TFSI	Lithium bis(trifluoromethanesulfonyl)imide
LLZO	Lithium lanthanum zirconium oxide
LSE	Localized saturated electrolyte
LT	Low-temperature
MA	Methyl acetate
mAh	Milliamp-hour
Mn	Manganese
MOC	Mesoporous ordered ceramic
MOF	Metal-organic framework
MP	Methyl propionate
N/P	Negative electrode to positive electrode capacity ratio
NA	$LiNi_{0.95}Al_{0.05}O_2$
NATM	Co- and Mn-Free $LiNi_{0.93}Al_{0.05}Ti_{0.01}Mg_{0.01}O_2$

Nb	Niobium
NC	$LiNi_{0.94}Co_{0.06}O_2$
NCA	Nickel cobalt aluminum oxide
NECST	Nanomaterials for Energy Conversion Storage Technology
Ni	Nickel
NM	$LiNi_{0.95}Mn_{0.05}O_2$
NMC	Nickel manganese cobalt oxide
NMR	nuclear magnetic resonance
OEM	Original equipment manufacturer
ORNL	Oak Ridge National Laboratory
PDF	Pair-distribution function
PECVD	Plasma-enhanced chemical vapor deposition
PEO	Polyethylene oxide
PFPE	Perfluoropolyether
PI	Principal Investigator
PNNL	Pacific Northwest National Laboratory
POCs	Porous and mesoporous ordered ceramics
POFM	Porous organometallic framework materials
PTA	Polysulfide trapping additives
PVDF	Polyvinylidene difluoride
R&D	Research and development
RDD&D	Research, development, deployment, and demonstration
S	Sulfur
SEI	Solid-electrolyte interface
SEM	Scanning electron microscopy
Si	Silicon
SIMS	Secondary Ion Mass Spectrometry
SLAC	Stanford Linear Accelerator Center
SNL	Sandia National Laboratories
SoC	State of charge
SPAN	Sulfurized polyacrylonitrile
TEM	Transmission electron microscopy

TFEPE	1,1,2,2-tetrafluoroethyl n-propyl ether
Ti	Titanium
ToF	Time-of-Flight
TXM	Transmission X-ray microscopy
U.S.	United States
UCSD	University of California-San Diego
USABC	United States Advanced Battery Consortium
UT	University of Texas
V	Volt
Vhold	Voltage hold
VTO	Vehicle Technologies Office
Wh	Watt-hour
XRD	X-ray diffraction

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