Process Development and Scale-up of Advanced Cathode Materials

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Overview

- **Timeline**
  - Project start date: Oct 2010
  - Project end date: Sept 2014
- **Barriers**
  - Barriers addressed
    - Cost – Reduce cost to manufacturer materials
    - Performance – Optimize processing for maximum performance
- **Budget**
  - Funding received in FY11: $850K + Internal funds
  - Funding received in FY12: $1.5M
- **Partners**
  - Argonne's EES Applied R&D Group
  - Argonne's EES Materials Screening Group
  - We are open to work with any ABR partner

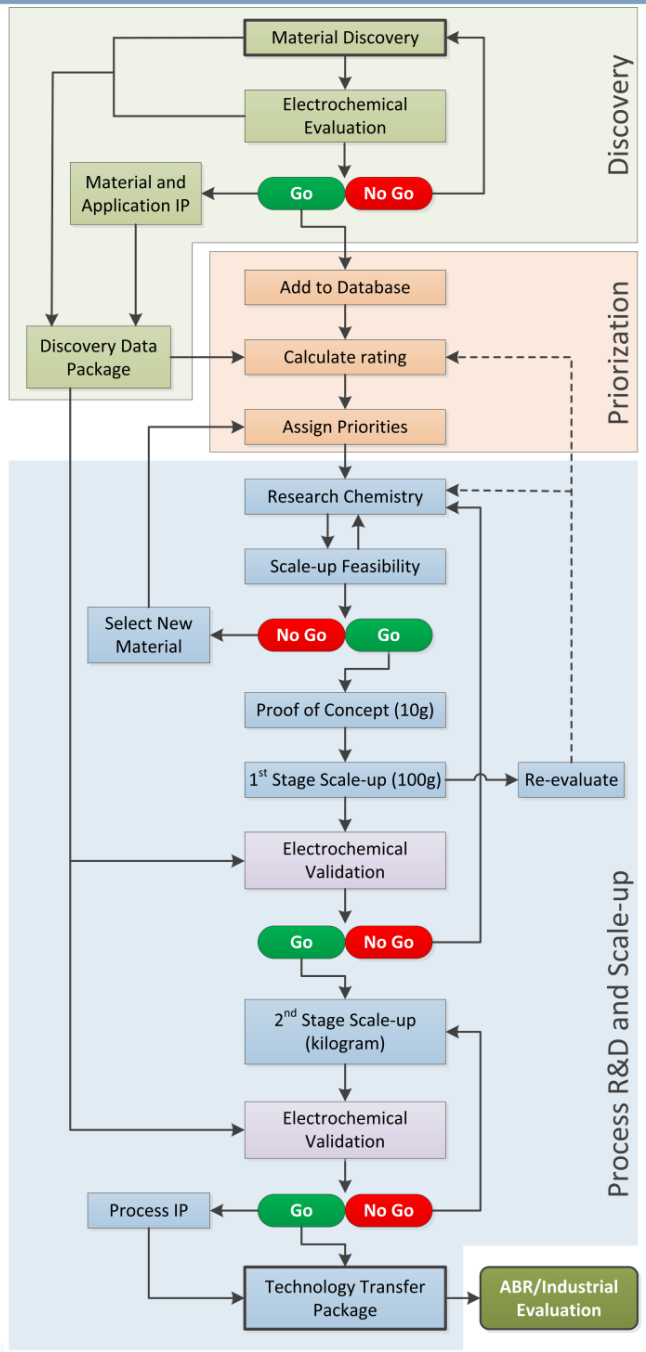
Objectives and Relevance of this Program

- **The objective of this program is to provide a systematic engineering research approach to:**
  - Identify and resolve constraints to development of cost-effective process technology for the scale-up of advanced battery cathode materials.
  - To provide sufficient quantities of these materials produced under rigorous quality control specifications for evaluation by the market.

- **The relevance of this program to the DOE Vehicle Technologies Program is:**
  - This program is a key missing link between discovery of advanced battery materials, market evaluation of these materials and high-volume manufacturing or transportation applications.
  - This program will produce quantities of materials for prototype cells to enable quick-turnaround validation screening of new materials chemistries throughout the R&D process.
  - This program will provide the basis for meeting broader industrial needs to reduce the risk associated with developing and maintaining a domestic commercially viable battery manufacturing capability.

Approach

- Identification and prioritization of candidate materials
- Develop experimental work planning and control (EWPC) and Quality Control (QC) procedures
- Determine scale-up feasibility
- Process modeling and production cost estimation (limited now)
- Proof-of-concept/small-scale verification of process
- First-stage scale-up/product validation (100 g scale synthesis)
- Second-stage scale-up/process verification (kilogram scale synthesis)
- Create Technology Transfer Package
- Mate the materials available for industrial evaluation
- Project reporting



Approach - Milestones

Milestones

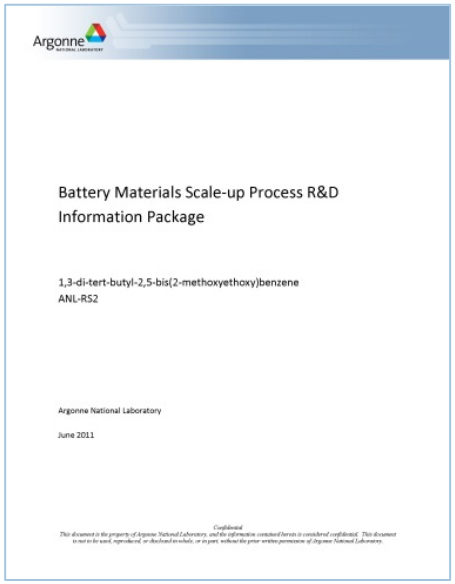
- FY11 – Purchase and install equipment, establish interim process scale-up and analytical labs - COMPLETED

- FY12 – 1-2 cathode materials to be scaled
  - $Li_{1.14}Mn_{0.57}Ni_{0.29}O_2$ – IN PROGRESS
  - 2nd material to be determined

- FY13 – 2-4 cathode materials to be scaled

Approach – Deliverables

- For each cathode material selected we will:
  - Develop a scalable manufacturing process.
  - Develop analytical methods and quality control procedures.
  - Prepare a “technology transfer information package” which will include:
    - Summary of the original process used by discovery researchers to synthesize the material.
    - Summary of the scalable (revised) process suitable for large scale manufacturing.
    - Detailed procedure of the revised process for material synthesis.
    - Analytical data/Certificate of Analysis for the material.
    - The material impurity profile.
    - Electrochemical performance test data.
    - Preliminary estimates of production costs.
    - Material safety data sheet for the material.
  - Make kilogram quantities of the material available for industrial evaluation.
    - The material will be fully characterized chemically and electrochemically.


Argonne

Battery Materials Scale-up Process R&D
Information Package

1,3-di-tert-butyl-2,5-bis(2-methoxyethoxy)benzene
ANL-RS2

Argonne National Laboratory

June 2011

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Technical Accomplishments and Progress
Set-up Interim Materials Synthesis Lab

- Cathode materials synthesis lab was established
  - Wet processing area
    - 4L and 20L TSK transparent CST reactors
    - 20L Tae Hyun stainless steel CST reactor
    - GL Filtration filter washer dryer
    - Fujisaki spray dryer
  - Dry processing area (powders hood)
    - Powrex vertical mixer
    - TURBULA® Shaker-Mixer
    - Retsch shaker sieve, crusher, mill
    - NGK calcination furnace
    - Nissinn air classifier
- Characterization lab was upgraded
  - Coin cell fabrication and cycling was added
    - VAC glovebox
    - Maccor cycler

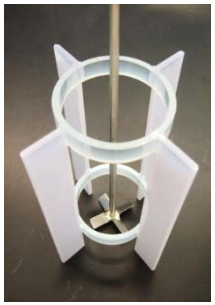
Technical Accomplishments and Progress First-Stage Optimization for $Li_{1.14}Mn_{0.57}Ni_{0.29}O_2$

- **4L Co-precipitation – 100g scale**
  - Modification of baffle, impeller and RPM
    - Optimal particle size
    - Minimize particle size distribution
- **Washing & filtering**
  - Minimize impurities & wastewater
- **Drying**
  - Minimize moisture content & drying time
- **Sieving**
  - Remove large particles & minimize moisture absorption
- **Mixing**
  - Uniform distribution of precursor & Li_2CO_3
  - Optimal Li mole ratio for cathode material
- **Calcination**
  - Maximize capacity & cyclability
- **Classification**
  - Optimal particle size

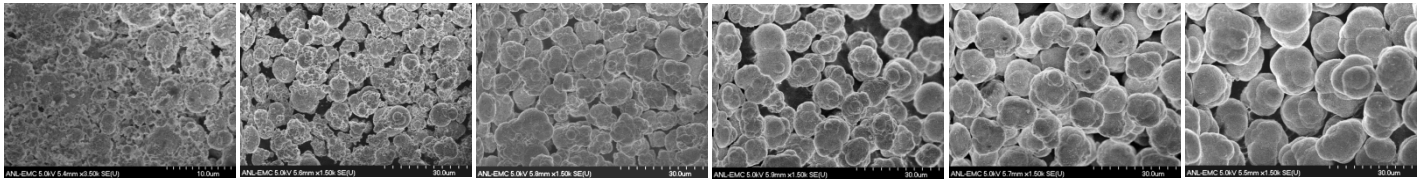
Technical Accomplishments and Progress
Examples of Process Optimization

Reactor Geometry Optimization - Reactor dynamics were changed with modified insert, mixer blade and RPM to improve particle shape and decrease particle size.

- 4L CST transparent reactor (#2011_08_16)

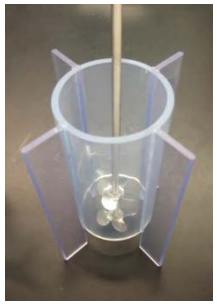


Operation
Normal baffle
Square cross
1000rpm
49.6°C
pH 7.87

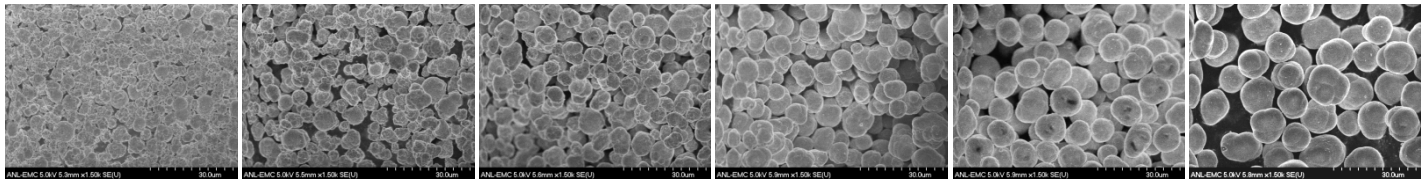


	After 2hr	After 3hr	After 4hr	After 5hr	After 6hr	After 7hr
D10/D50/D90	0.21 / 7.77 / 12.33	0.25 / 9.99 / 15.06	1.47 / 11.68 / 16.88	9.07 / 13.44 / 19.25	11.01/ 15.34 / 21.47	12.11 / 17.12 / 24.30

- 4L CST transparent reactor (#2011_09_21)



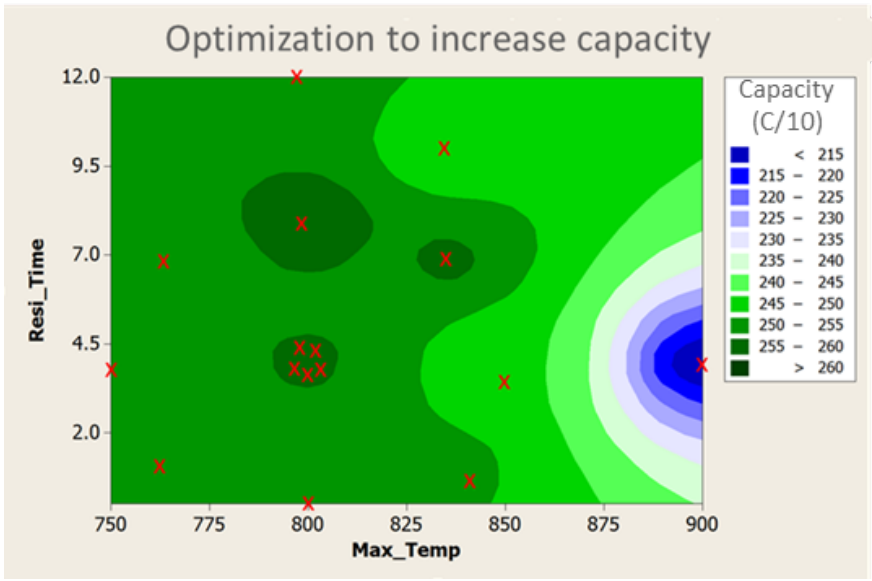
Operation
Draft tube
2 Propeller
2000rpm
50.3°C
pH 7.88

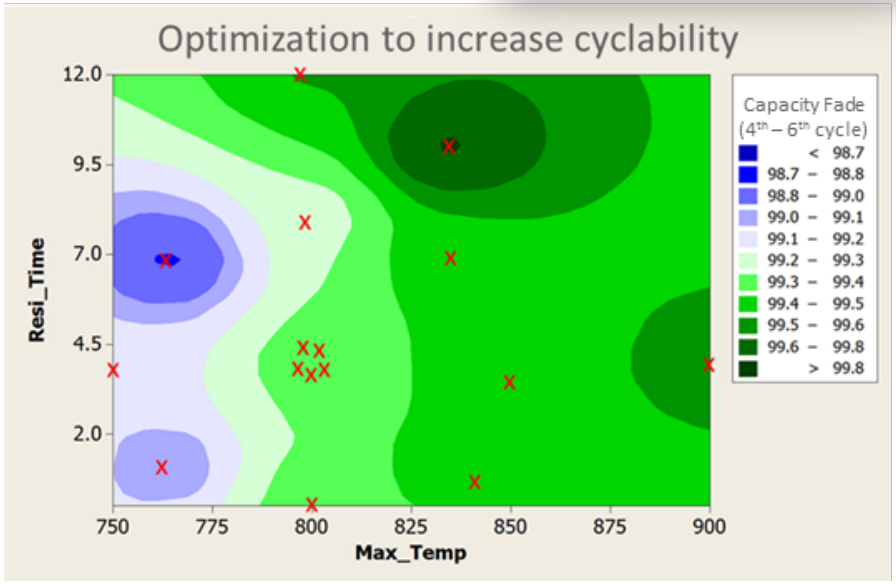


	After 2hr	After 3hr	After 4hr	After 5hr	After 6hr	After 7hr
D10/D50/D90	1.28 / 6.46 / 10.00	1.43 / 7.35 / 11.08	0.29 / 8.08 / 11.99	0.22 / 9.11 / 13.32	0.24 / 10.66 / 15.56	8.94 / 12.87 / 17.96

Technical Accomplishments and Progress
Examples of Process Optimization

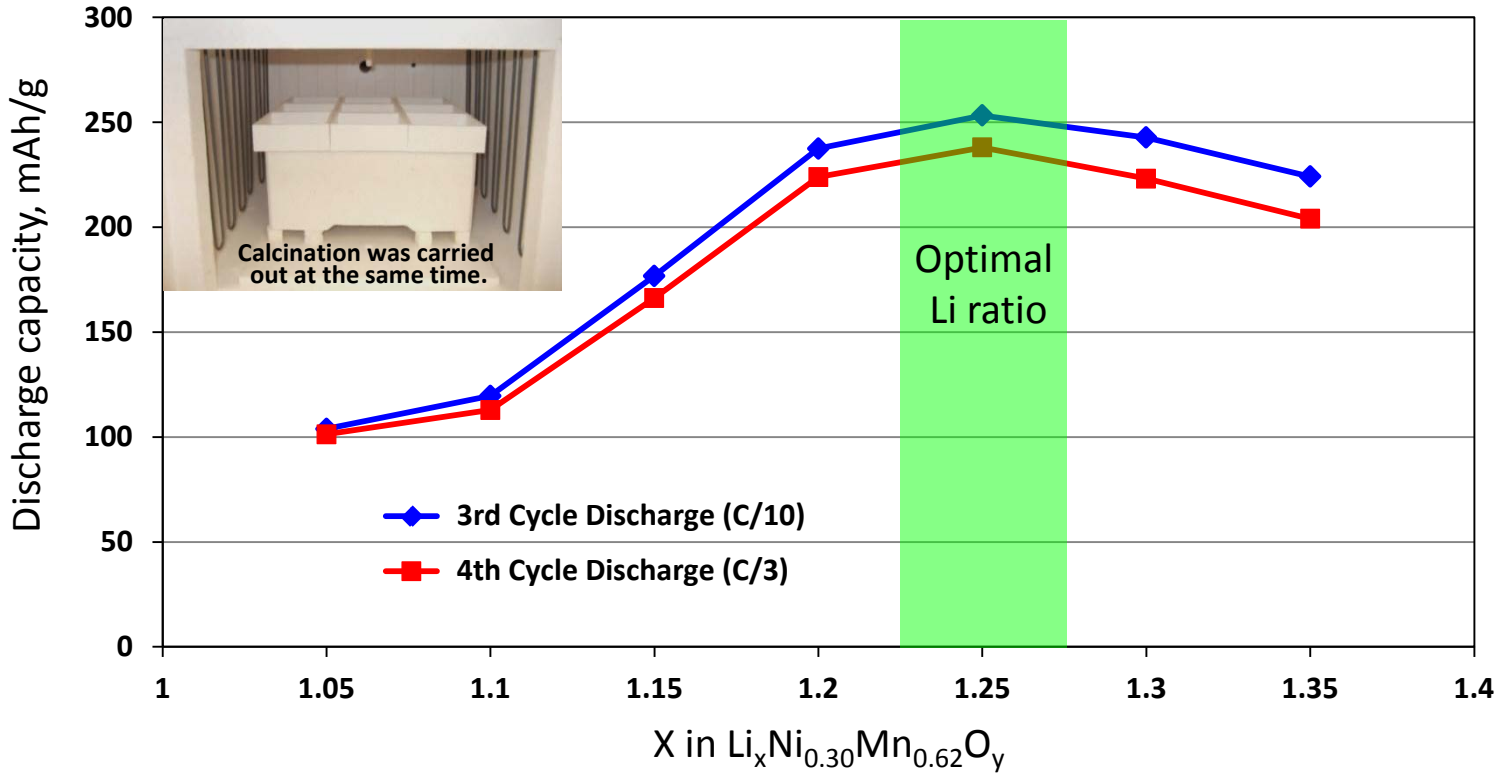
Calcination Parameter Optimization - 16 calcinations were run to determine optimal capacity and cyclability parameters using statistical analysis software Minitab.™





Technical Accomplishments and Progress
Lithium Content Optimization

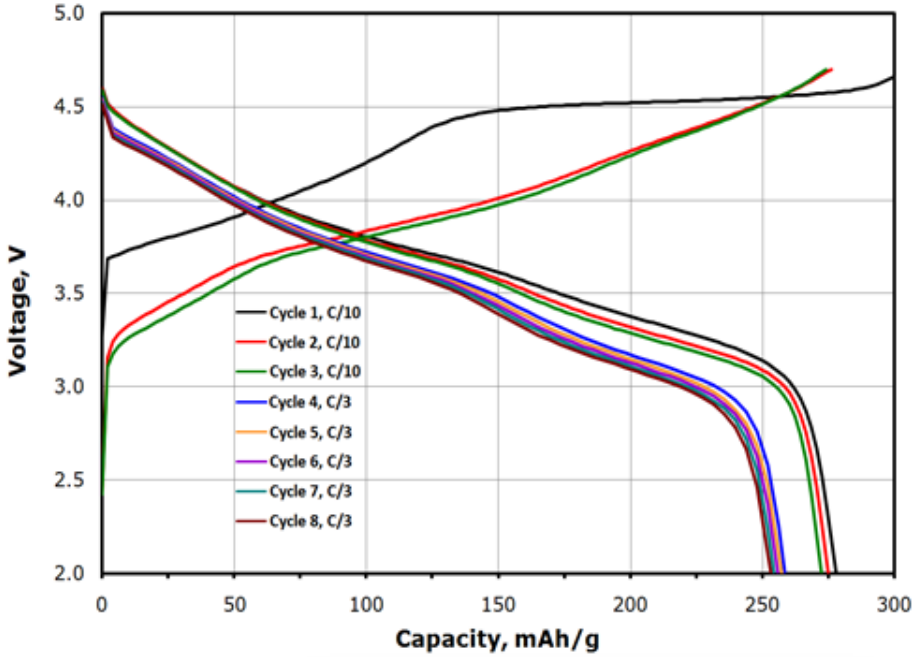
- Same precursor (ES #2011_09_21) was used for the 7 samples.
- Moisture content of dried precursor was considered for Li_2CO_3 addition.
- Optimized calcination condition (835°C and 10hr) was applied.

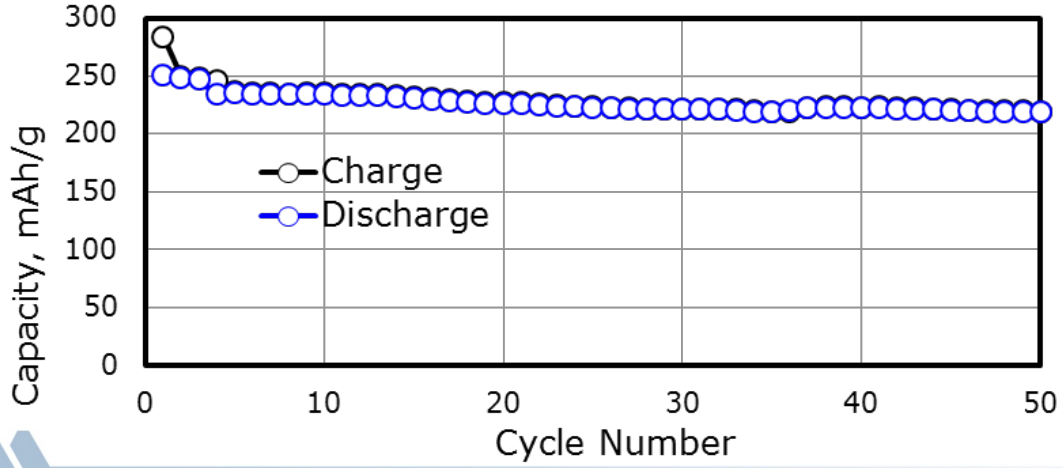


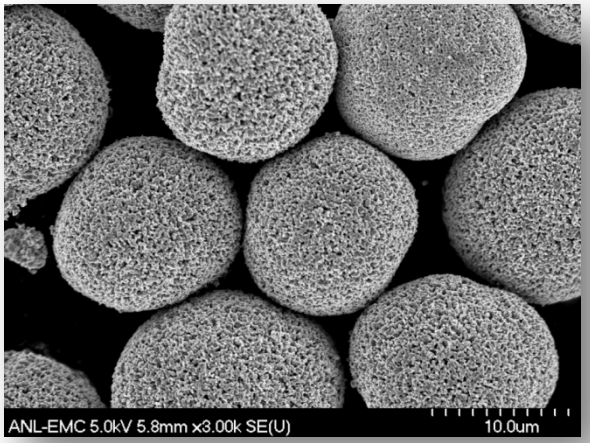
- ✓ X is a calculated value based on dried precursor.
- ✓ Addition of 1.25 Li ratio to dried precursor shows the best capacity.

Technical Accomplishments and Progress Electrochemical Validation

- Coin cell results from first-stage scale-up
  - Active material: $Li_{1.20}Ni_{0.30}Mn_{0.61}O_{2.1}$ 400g stock (ES #2011_08_16)
  - Slurry composition: 84% active material, 4% acetylene black, 4% SFG-6, 8% PVdF
  - Anode: Li metal, Electrolyte: GEN2, Separator: Celgard® 2325 MM

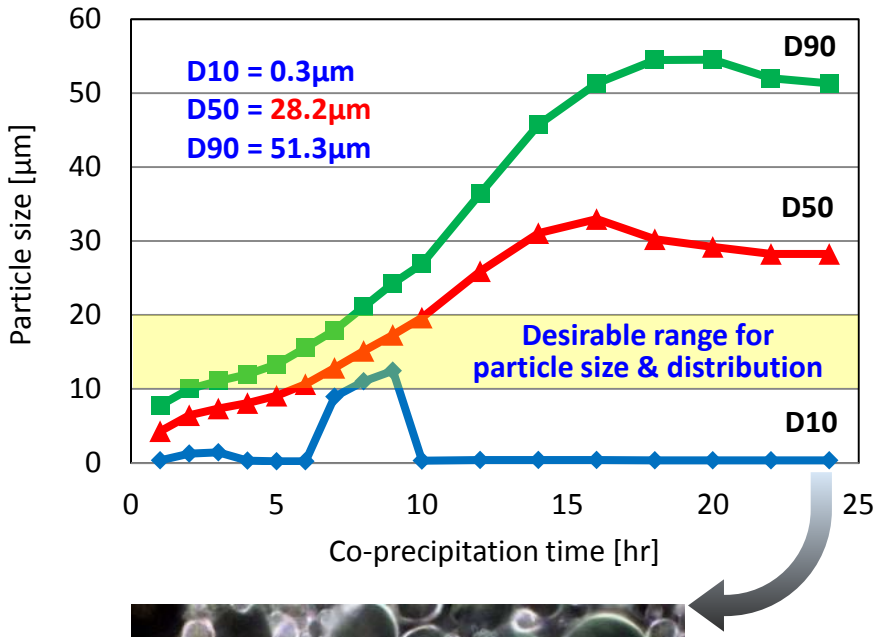




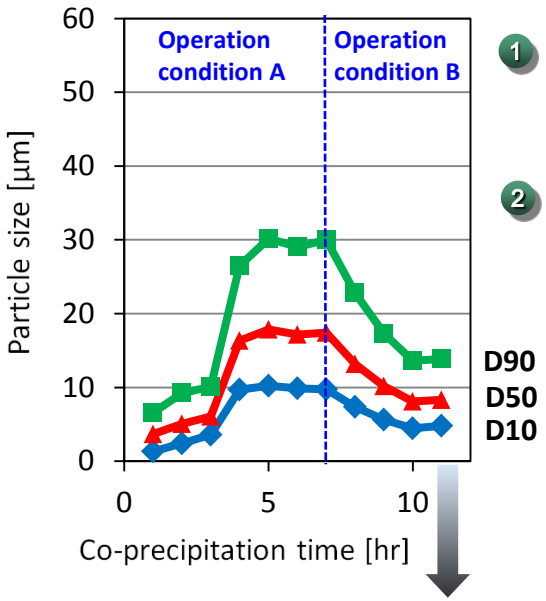


Technical Accomplishments and Progress
Precursor Particle Growth Problem

- 24hr long-run test using 4L CSTR (#2011_09_21)
  without process improvement

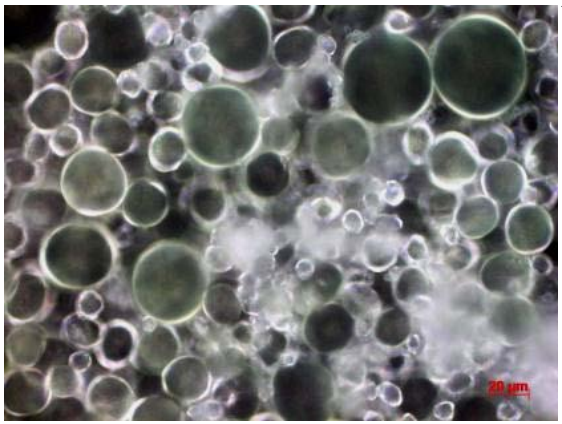


- 3rd long-run test using 20L CSTR (#2012_02_22)
  with process improvement



1. Particle size & distribution can be easily controlled by changing operation condition.
2. Unsteady start-up time was reduced greatly.
   --> Save raw materials
   (decreased off-spec.)

D10 = 4.8μm
D50 = *8.3μm*
D90 = 13.9μm



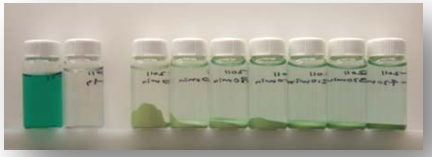
Microscope (X50,000) after 24hr

- Tiny particles were removed
  (increased tap density)
- Huge particles were removed
  (positive effect on calendaring and improved cathode property)

Microscope (X50,000) after 11hr

Technical Accomplishments and Progress Summary of Scale-up of $Li_{1.14}Mn_{0.57}Ni_{0.29}O_2$

- QC and EWPC procedures developed and approved
- Feasibility determination/proof of concept assessment was completed
- First-stage scale-up was conducted on a 4L reactor (200g per hour scale)
  - Optimization of each synthesis step was completed
- Precursor particle growth issue was confirmed
  - Solution has been identified and tested
- Second-stage scale-up was conducted on a 20L reactor (1 kg per hour scale)
  - Optimization on the 20L scale is in progress
  - 1 kg sample has been provided to the Materials Screening group



Collaborations

- Argonne's Electrochemical Energy Storage, Applied R&D group
  - Cathode chemistry was initial material scaled
- Argonne's Electrochemical Energy Storage, Material Screening group
  - Conducted electrochemical validation of scaled materials
- **We invite all ABR program participants to submit new candidates to our advanced cathode materials process R&D program.**

Activities for Next Fiscal Year

- Ongoing – Update cathode materials to scale spreadsheet with materials from other labs in the ABR program
  - Rank and prioritize the materials

- FY 12 – Complete scale-up of $Li_{1.14}Mn_{0.57}Ni_{0.29}O_2$ and 2nd cathode material
  - Provide material samples for electrochemical validation
    - Electrochemical validation done by Argonne's material screening program
  - Provide material samples for industrial evaluation
  - Relocate interim labs to the Materials Engineering Research Facility

- FY 13 – Scale 2-4 cathode materials from bench to kilogram scale
  - Provide material samples for electrochemical validation
    - Electrochemical validation done by Argonne's material screening program
  - Provide material samples for industrial evaluation

Activities for Next Fiscal Year
The Materials Engineering Research Facility (MERF)

- 10,000 sq. ft. facility consists of "Group-H occupancy" pilot labs and high bay spaces.
- All interim labs will be relocated to the MERF, starting in April.
- 50% DOE – 50% DoD funded

Analytical Lab

Pilot Labs (benchtop)

Pilot Labs (walk-in)

High Bay Spaces

Summary

- The procedures used to make small, research sample of materials are not suitable for large scale production. Manufacturing processes for the newly discovered advanced materials must be scalable to facilitate the transition from basic research to commercial application.
- This program has been developed to provide a systematic approach to process R&D and scale-up, and to provide sufficient quantities of advanced cathode materials for industrial evaluation.
- Argonne's process R&D program enables industry to carry out large-scale testing of new cathode materials.
- Integration of materials discovery with process R&D will expedite the time needed to commercial deployment.
- Optimization of the first stage scale-up of $Li_{1.14}Mn_{0.57}Ni_{0.29}O_2$ was successfully completed. Second stage of scale-up is in progress, material was generated at the kilogram scale.
- Samples of the material were provided to various entities for industrial evaluation.

Acknowledgements and Contributors

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