2019 DOE Vehicle Technologies Office Annual Merit Review

An Integrated Flame Spray Process for Low Cost Production of Battery Materials

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Project ID#: bat269

Overview

Timeline

- Start: January 1, 2016
- End: December 31, 2019

Budget

- Total project funding
  - DOE share: $2,215,556
  - Contractor share: $310,694
- Funding received in FY 2018
  - $270,058.60
- Funding for FY 2019
  - N/A

Barriers addressed

- Low cost battery target of $125/kWh
- Energy density target of $250 Wh/kg
- Cost-effective battery materials manufacturing processes

Partners

- Project lead
  - University of Missouri
- Collaborator
  - EaglePicher Technologies

Relevance

Overall objective

The overall objective of this project is to develop an advanced manufacturing technology for battery materials production at low cost and in a green chemical process using glycerol as solvent to replace water.

Specific objectives

- To reduce material cost by at least 25% to $34/kg or less as compared to a baseline $45/kg;
- To achieve a lab scale production rate of 3 kg/day and a pilot scale production rate of 4 metric tons per year of cathode powders.
- To demonstrate battery cells with 250 Wh/kg energy density.

Objectives for the Review Period

1. To control flame spray process
2. To optimize powder morphology and size
3. To achieve a material with high specific capacity

Resources

At University of Missouri

- Custom-designed and fabricated flame spray reactor for the work.
- Full dedicated lab (~1500 ft^2) with walk-in hood for the experimental setup of the flame spray work.
- A battery lab (~900 ft^2) equipped with Unilab Mbraun glovebox and Innovation Technology glovebox, Arbin battery cycler, and coin cell fabrication equipment.
- Wet chemistry lab with fume hood for any chemistry work and battery materials handling.
- Several potentiostats/galvanostats for electrochemical property measurements.
- On campus facilities include SEM, TEM, XRD, ICP-MS, and other equipment.

At EaglePicher Technologies

- Complete battery fabrication facilities
- Dry rooms
- Complete battery testing equipment
- Battery R&D facilities

Milestones

FY18/FY19 Milestones and Work Status

Milestone	Descriptions	Status
Go/No Go*	Demonstrate a production capacity of 3 kg/day with the high throughput spray technology.	Completed
Tech	Select an in situ or semi-in situ coating process.	Completed
Tech	Demonstrate control of active material powder sizes to around 10 microns on average.	Completed
Tech	Cell design including matching anode and suitable electrolytes.	On-going
Tech	Demonstrate five (5) cells of 250Wh/kg energy density.	On-going

FY19/FY20 Milestones and Work Status

Milestone	Descriptions	Status
Tech	Cost reduction of at least 25% to $34/kg or less for active material NMC demonstrated.	On-going
Tech	Materials processing method using either a flow process or a stationary process will be selected.	To start
Tech	Pilot production line construction and material production.	To start

* The Go/No Go target will be aimed again in the next period when producing powders with a different chemistry.

Approach

The **overall approach** is to develop a new flame spray process to make active cathode powders with new chemistry. Glycerol, as a cheap industrial byproduct, is used as solvent to replace water and as a fuel to process transition metal oxide powders to reduce energy consumption. The process also uses natural gas through combustion to provide energy to anneal the powders during the synthesis to lower material cost.

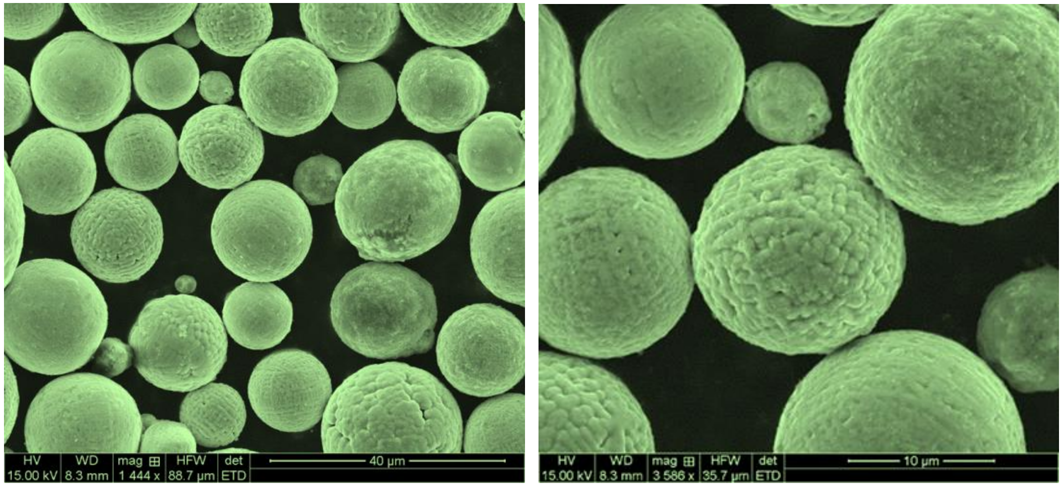
Approaches to technical barriers:

- ❑ Modifications of the flame spray reactor has been made to resolve the lithium loss issue (completed);
- ❑ Morphology and size are controlled through flow and temperature distribution in the reactor (completed);
- ❑ High capacity powders have been made with NCA chemistry (on-going).
- ❑ Process economic analysis for achieving low cost target (on-going).

Technical Accomplishments and Progress (1)

1. Flame spray reactor control

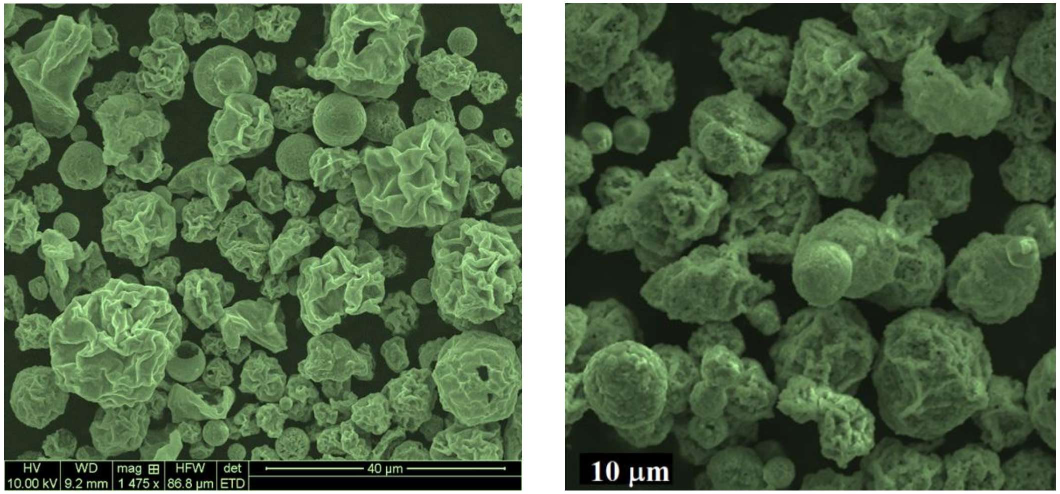
Improvements in the flame reactor have been made in this period in which the reactor can be controlled in temperatures to achieve certain desired powder morphology and properties.



SEM images of NCM(333) powder with spherical morphology.

2. Powder morphology

Powder morphology has been under control. The NCM powders produced can have a spherical particle shape and wrinkled surface structures. Their sizes can be controlled in a range of a few to a few tens of microns.



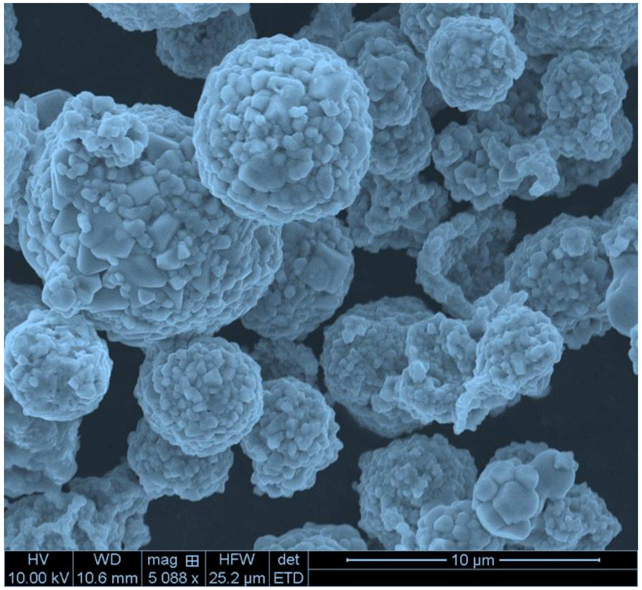
SEM images of NCM(333) powder with wrinkled surface.

Technical Accomplishments and Progress (2)

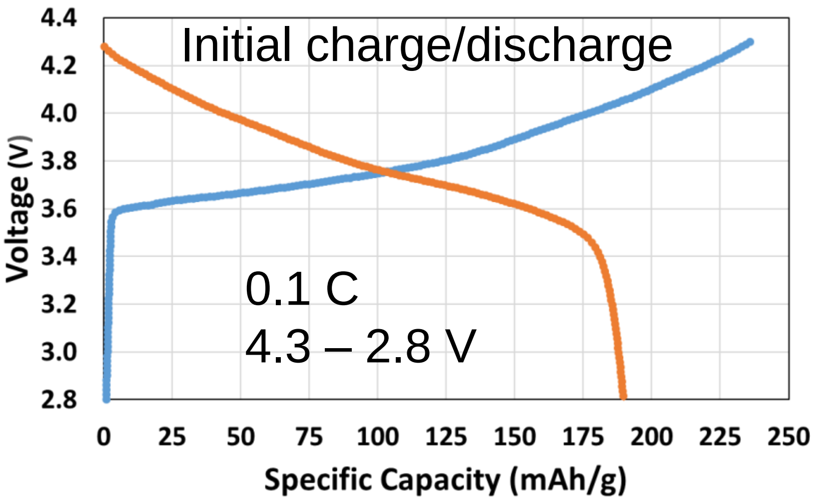
3. High capacity powders

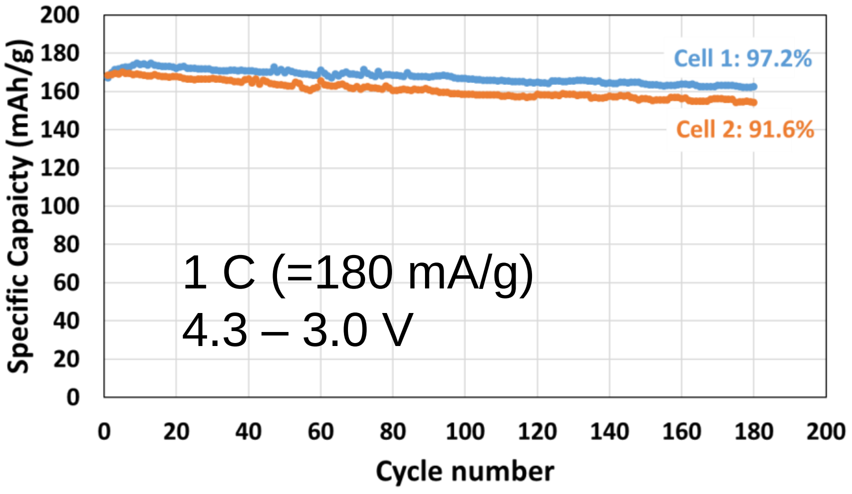
To achieve high capacity, $Li(Ni_{0.8}Co_{0.15}Al_{0.05})O_2$ (NCA) chemistry was used and NCA powders were made in the flame spray reactor. The powder morphology is shown on the right.

The NCA material was tested in coin cell. It was found that an initial discharge capacity of 190 mAh/g can be achieved at 0.1 C in 4.3-2.8V. Furthermore, at 1C the NCA materials show a good retention rate of over 90% after 180 cycles.



NCA powder

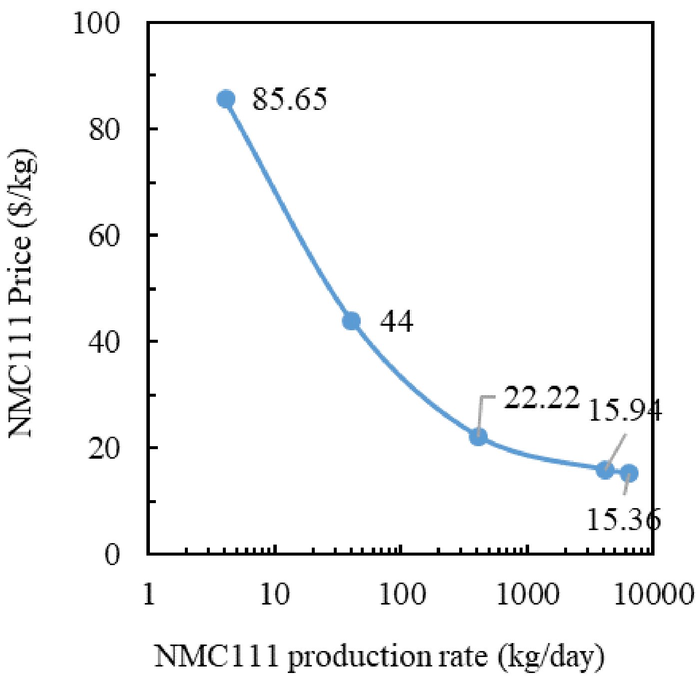


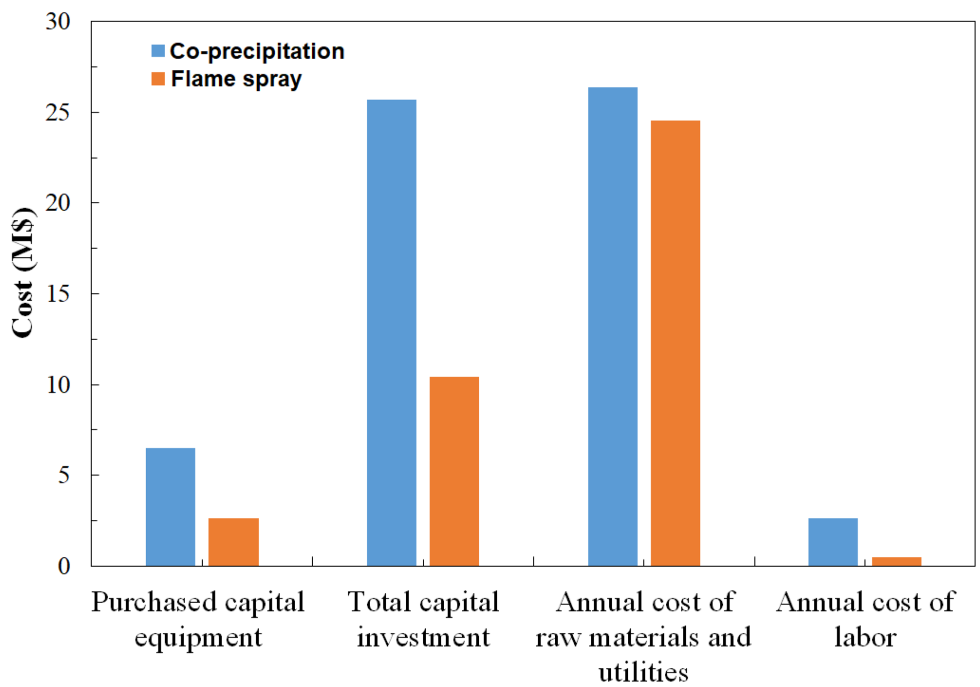


Technical Accomplishments and Progress (3)

4. Process Economic Analysis

To achieve lost target, the flame spray process was analyzed on its economical benefit in cost reduction. The analysis is based on a discounted cash flow method. The cathode material NMC111 was used as an example in the analysis and results were compared to that of the co-precipitation process*. It was shown that the flame spray process is a much lower-cost process than co-precipitation with 33% cost savings in the NMC material.





*S. Ahmed, P.A. Nelson, K.G. Gallagher, N. Susarla, D.W. Dees, Cost and energy demand of producing nickel manganese cobalt cathode material for lithium ion batteries, Journal of Power Sources 342 (2017) 733.

Responses to Reviewers' Comments (1)

Some major concerns of the reviewers are addressed below.

<u>Reviewer 3 in Q1</u>: According to the reviewer, flame spray is historically an inconsistent process with considerable waste. This, combined with issue of Li vapor pressure, raises concerns regarding process viability at scale-up for manufacturing.

- ❖ **Response**: There are different flame spray processes. Our flame spray process is different from others; it does not generate much waste which is an advantage in itself. In addition, the Li loss issue has been resolved.

<u>Reviewer 2 in Q2</u>: The reviewer stated that the reported successes have been few. Also, the team reports need to re-evaluate go/no-go milestones involving past work.

- ❖ **Response**: In developing the new flame spray process, we have accomplished much. While the unexpected Li loss delayed development in materials production, the problem has been resolved as presented above. In this period, NCA powders have been made with very good performance.

Responses to Reviewers' Comments (2)

Reviewer 1 in Q4: The reviewer mentioned that the team is simply continuing its effort based on the original work plan without extensive modification from lessons learned.

- ❖ **Response**: The team has been improving the process with what it learned from experience. In particular, dramatic modifications were done to the flame reactor to successfully reduce the Li loss.

Reviewer 3 in Q4: Referring to Slide 12, the reviewer suggested not spending time and money on surface coatings until the bare NMC can perform as well as commercial powders. The coating just complicates interpretation of what is causing poor performance. Currently commercial NMC cells can cycle hundreds of times at the voltages used here. To remove the maximum voltage (Vmax) from the fade mechanism, the reviewer urged that these cells be tested at a more reasonable 4.3V and demonstrate acceptable cycle life.

- ❖ **Response**: We agree with the reviewer suggestion. While we have done more work on the NMC, we had decided to make NCA powders in order to achieve the 250 Wh/kg target. The results on NCA presented above show very good cycle life even without surface coating.

Collaboration with Other Institutions

- EaglePicher Technologies (EPT) is a collaborator in the project. EPT as a battery manufacturer and developer has vast capabilities in battery research. The role of EPT in this project is to design and make battery cells to achieve the targeted energy density,

- Collaborations with Argonne has been forged, with a goal that ANL will benchmark our materials. ANL has conducted some initial work on our materials.

- Future collaborations with academic institutions, research labs and industries are to be made.

Remaining Challenges and Barriers

- To achieve the energy density target (250 Wh/kg), the NCA chemistry was used. The NCA powders were made and test results showed that they have a desired performance. Expected challenges could arise in cell design and fabrications using the NCA powders, but we expect to overcome them.

- Other Ni-rich chemistries will be explored in the flame spray process to ensure achievement of the target.

Proposed Future Research

- To continue producing NCA powders and improve their properties (FY19). This is to achieve desirable specific capacity.
- To produce other powders with new chemistries (e.g., Li-rich) (FY19). This will help achieve high energy density cells.
- To design and fabricate battery cells to achieve the targeted energy density (FY19). This is for cell demonstration.
- To design and integrate other processes (e.g., surface coating) (FY19/20). This is for cost reduction and battery performance improvement.
- To scale up the flame spray process (FY19/20). This is to demonstrate the feasibility of the production process.

Any proposed future work is subject to change based on funding levels.

Summary

In this performance period,

1. The flame spray reactor has been modified to resolve lithium loss and control powder morphology.

2. NCM powders can be controlled with desired shapes and sizes.

3. High capacity NCA powder has been made and tested, which showed promising specific capacity and retention rate.

4. Economical analysis of the flame spray process shows that it is a lower cost process as compared to the co-precipitation process.