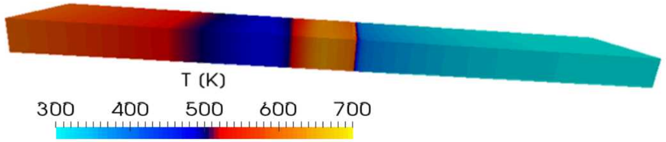
SAND2019-11428C

Heat Release from Thermal Decomposition of Layered Metal Oxide Cathodes in Lithium-Ion Batteries



PRESENTED BY

Randy Shurtz, John Hewson

2019 DOE Office of Electricity Peer Review
September 25, 2019

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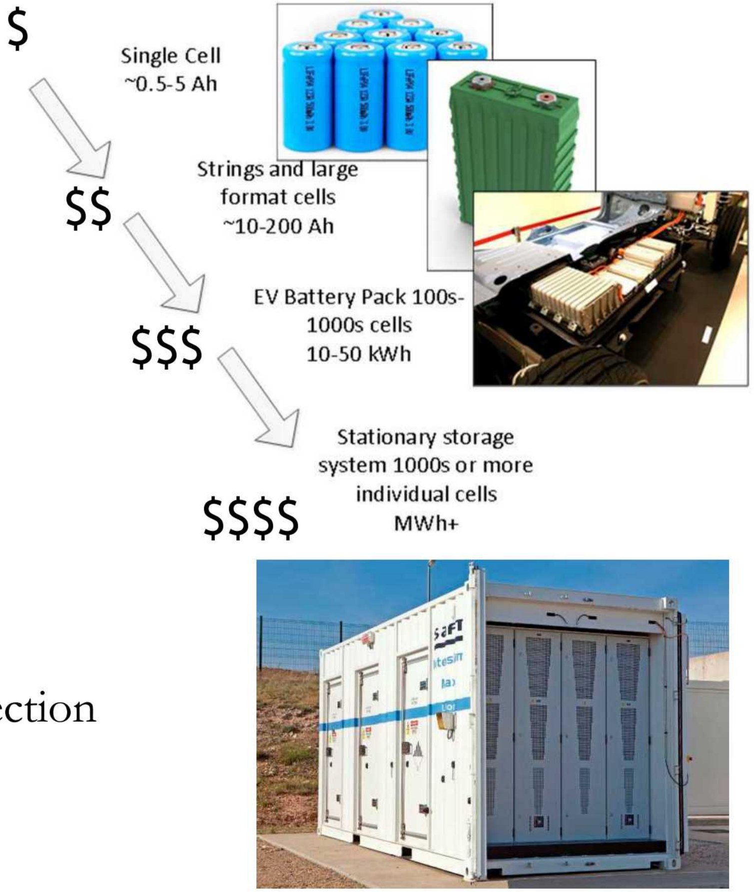
OVERVIEW of Thermal Runaway Modeling

SIGNIFICANCE:

- Heat source terms in legacy thermal runaway models have limitations
  - Outdated with respect to current battery materials
  - Designed for low-temperature onset rather than high-temperature propagation
- Models should be designed to keep pace with deployment of new materials
  - Transition from empirical approaches to materials-centric approaches
  - Gain ability to forecast safety characteristics in the early stages of materials selection

ALIGNMENT WITH CORE MISSION OF DOE OE:

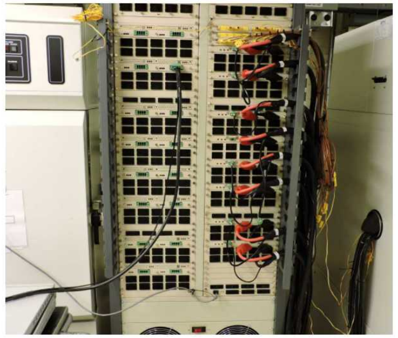
- Validated safety and reliability is one of the critical challenges identified in 2013 Grid Energy Storage Strategic Plan



www.cnn.com www.nissan.com
www.samsung.com www.saft.com
www.internationalbattery.com

PROJECT TEAM

Sandia Battery Test Facilities

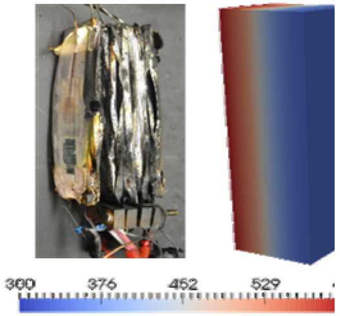


Yuliya Preger
Summer Ferreira
Armando Fresquez
Heather Barkholtz (former SNL)

Sandia Battery Abuse Lab

- Loraine Torres-Castro
Joshua Lamb
June Stanley
- Chris Grosso
- Lucas Gray
- Jill Langendorf
- Eric Deichmann

Sandia Fire Sciences

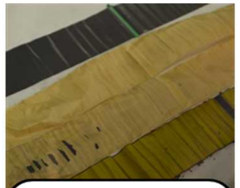


John Hewson
Randy Shurtz
Andrew Kurzawski

Center for Integrated Nanotechnologies

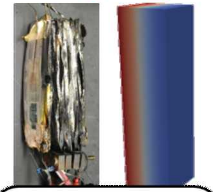
Sergei Ivanov

Baseline electrochemical performance analysis

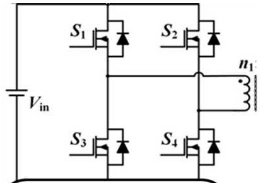


Materials characterization and thermal stability testing

Whole-cell abuse response analysis

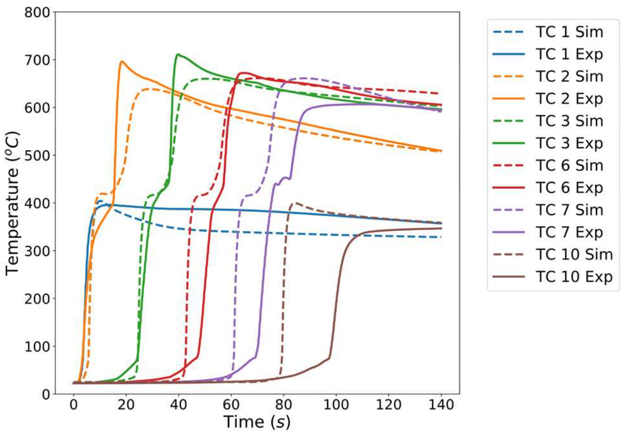


Modeling of thermal propagation



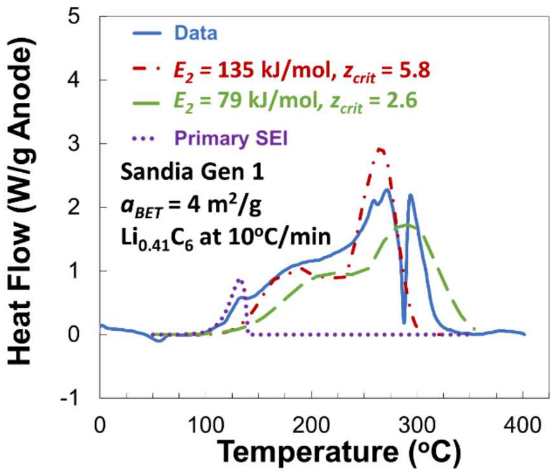
Design of new power electronics for battery safety

Thermal Runaway Modeling OBJECTIVES



<u>Predict thermal runaway behavior in large systems (multi-cell)</u>

- Discussed by John Hewson and Andrew Kurzawski in the Safety Session
- "Predicting and mitigating cascading failure in stacks of lithium-ion cells"



<u>Develop improved heat-source models for thermal runaway</u>

- Include proper dependence on material properties, temperature, state of charge
- Extend to additional electrode materials of commercial interest

<u>Promote effective methods and collaboration in thermal runaway studies</u>

- Publish perspectives and new models
- Set up thermal runaway collaboration workshops (task for full project team)

PROJECT METRICS AND MILESTONES

Milestone

1. Link safety modeling to materials science
2. Develop new models for decomposition of battery materials
3. Continue to evaluate recent models

Milestone #	Current Status
1, 2, 3	◦ Published 2 articles on anode decomposition models
1, 2	◦ Published a perspective article that promotes better utilization of calorimetry measurements for modeling ◦ Initiated battery workshop to promote coordination
1, 2	◦ Developed method to calculate total heat release from the most widely used class of cathode materials ◦ Identified data appropriate for calibrating decomposition rates from LCO, NMC, and NCA (follow-up papers)

Thermal Runaway Modeling CHALLENGES and OPPORTUNITIES

Calorimetry studies often report insufficient information for model development

- Material properties like surface area not reported
- Single scan rates for DSC do not permit derivation of activation energies
- Sample mass and state of charge not reported on consistent basis
- Species measurements often sparse or absent altogether

Opportunities include work, creativity, and outreach

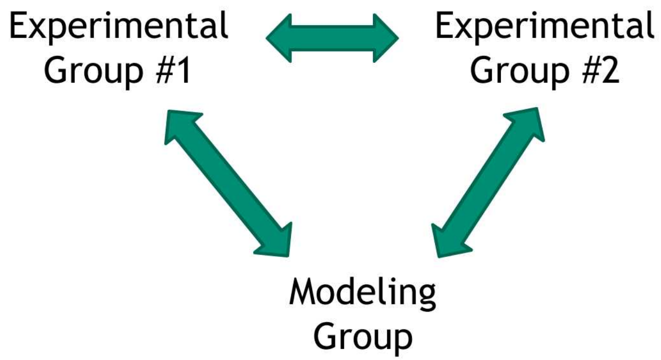
- Identify data appropriate for model development through careful searching (work)
- Novel perspectives or comparisons may compensate for apparent deficiencies in data (creativity)
- Share perspective on better ways to conduct and communicate research (outreach)

PROJECT RESULTS

Published invited perspective article in the Journal of the Electrochemical Society

- Identified methods for measurements, analysis, and reporting that:
  - Allow quantitative comparisons between diverse calorimetry measurements
  - Facilitate development of thermal runaway models
  - Promote mechanistic understanding of thermal runaway

Enhanced Flow of Data and Insights → Enhanced Development of New Insights

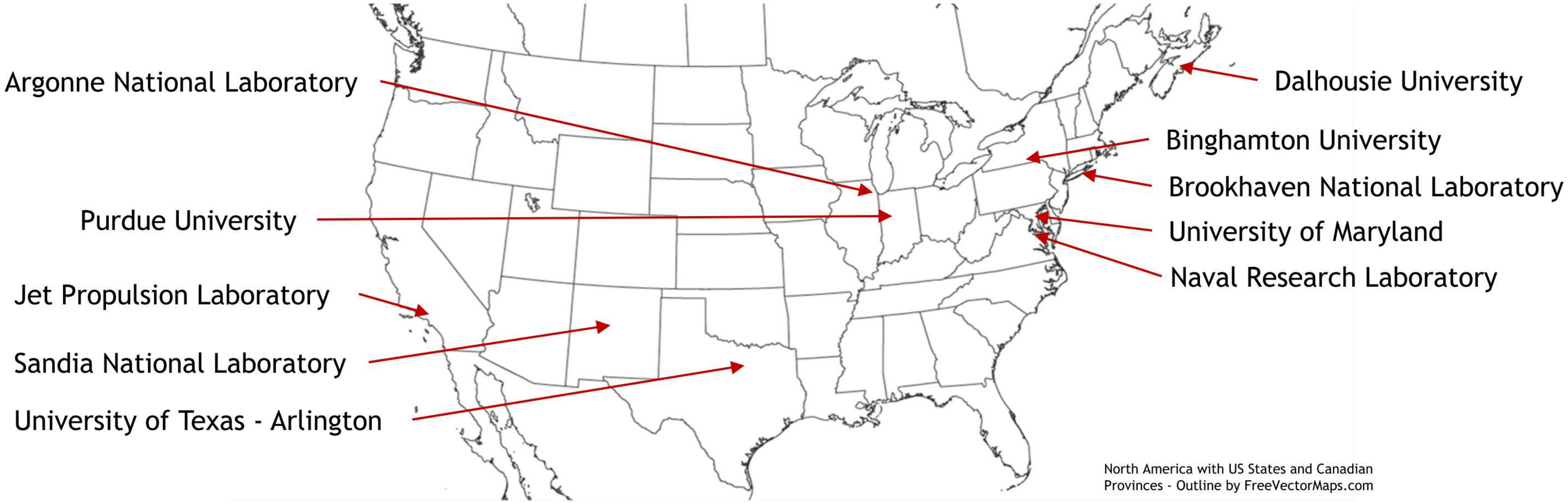


- Perspective article highlighted in ECS News on July 16, 2019
  - Title: "From Calorimetry Measurements to Furthering Mechanistic Understanding and Control of Thermal Abuse in Lithium-Ion Cells"
  - Reference: R. C. Shurtz, Y. Preger, L. Torres-Castro, J. Lamb, J. C. Hewson and S. Ferreira, *J. Electrochem. Soc.*, **166**, A2498 (2019). DOI 10.1149/2.0341912jes

PROJECT RESULTS

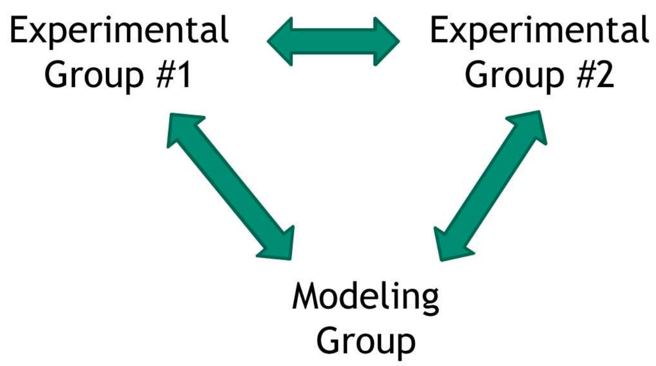
Initiated lithium-ion battery calorimetry workshop series to promote progress and collaboration

- Follows pattern of a successful workshop series for combustion measurements and model development
- Participants at initial kickoff meeting included 6 Sandians (safety team) and 9 additional researchers
  - Adjacent to the 235th ECS meeting in Dallas, Texas (May 2019)
- Used content from perspective article to highlight the benefits of improved communication and coordination
- 8 groups from outside Sandia shared feedback about directions to take the workshop series and how to collaborate
- Schedule planning meeting for May 2020 (ECS @ Montreal), first full-scale workshop for June 2021 (ECS @ Chicago)



North America with US States and Canadian Provinces - Outline by FreeVectorMaps.com

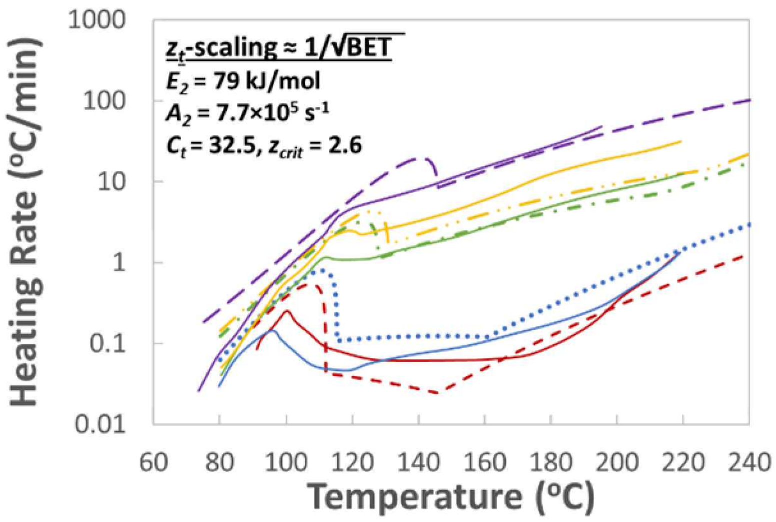
Enhance Flow of Data and Insights

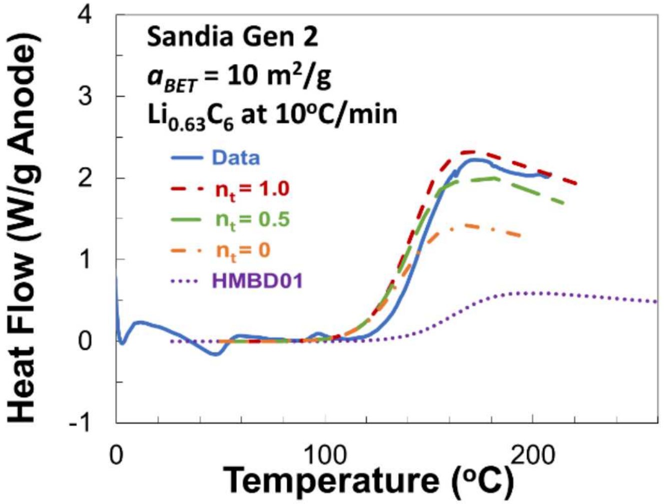


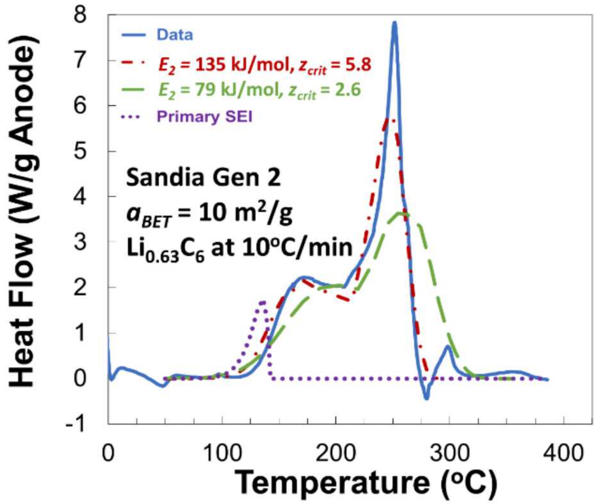
PROJECT RESULTS

Developed new model for heat generation from lithiated graphite anodes in electrolyte

- Improves predictions of maximum cell temperatures and cascading failure rates
  - Total heat release from reaction thermodynamics rather than empirical
  - Includes large exotherm occurring after onset of thermal runaway in full cells
  - Accounts for effects of graphite surface area and limited electrolyte on heat generation rates
  - Successfully predicts a wide variety of published calorimetry measurements
- Published in the Journal of the Electrochemical Society as two open-access papers
  - R. C. Shurtz, J. D. Engerer and J. C. Hewson, *J. Electrochem. Soc.*, **165**, A3878 (2018). DOI 10.1149/2.0171814jes
  - R. C. Shurtz, J. D. Engerer and J. C. Hewson, *J. Electrochem. Soc.*, **165**, A3891 (2018). DOI 10.1149/2.0541816jes

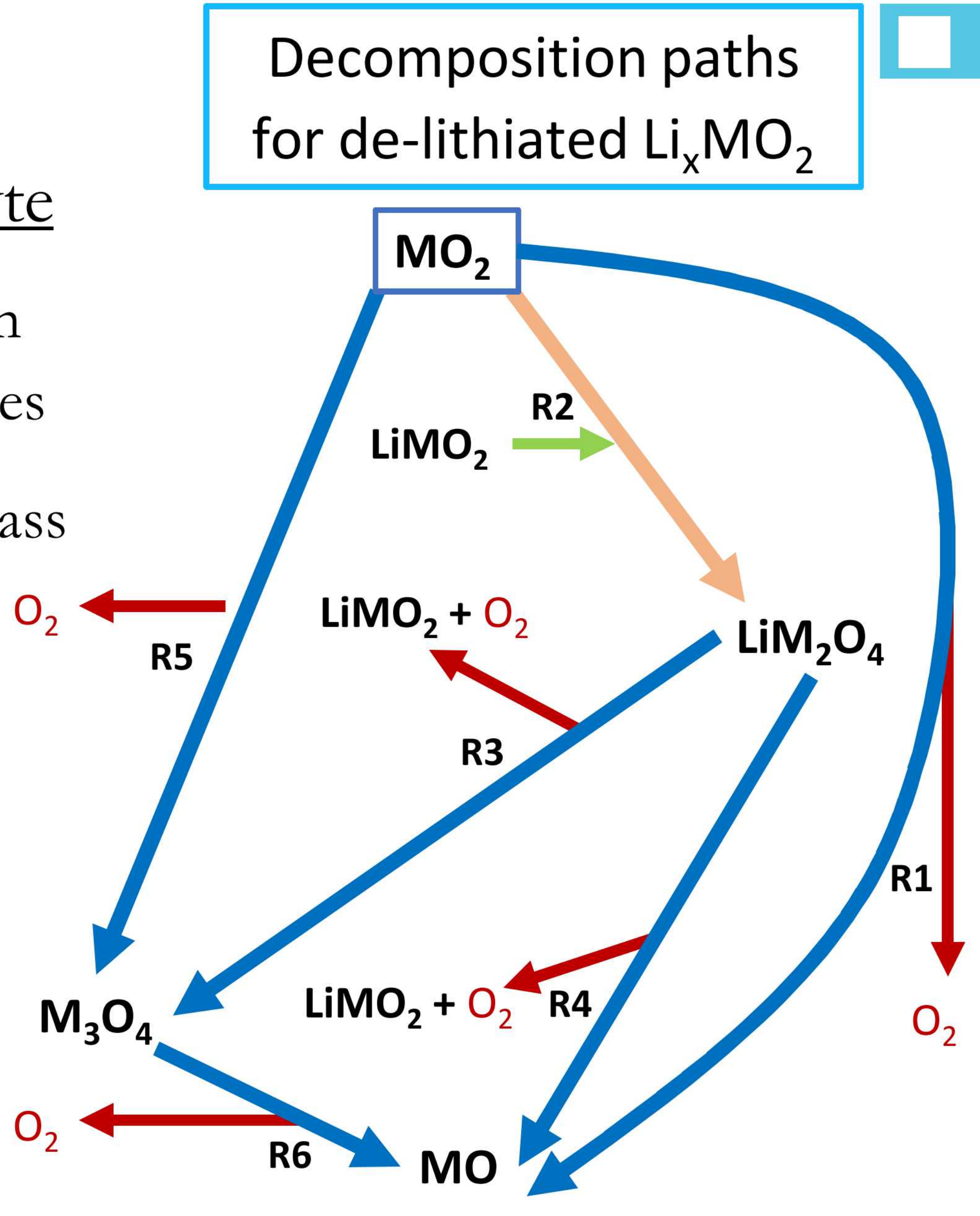






Layered Metal Oxide Decomposition in Electrolyte

- Compiled a database of 36 enthalpies of formation for cathode materials from over 42 literature sources
- Up-front predictions of heat release for a whole class of Li_xMO_2 cathode materials with electrolytes
  - Real or proposed compositions predicted rapidly
- Manuscript of journal article nearing completion
- Web-based calculator to be developed



M = Ni, Co, Mn, Al as well as mixtures (NMC, NCA, etc.)

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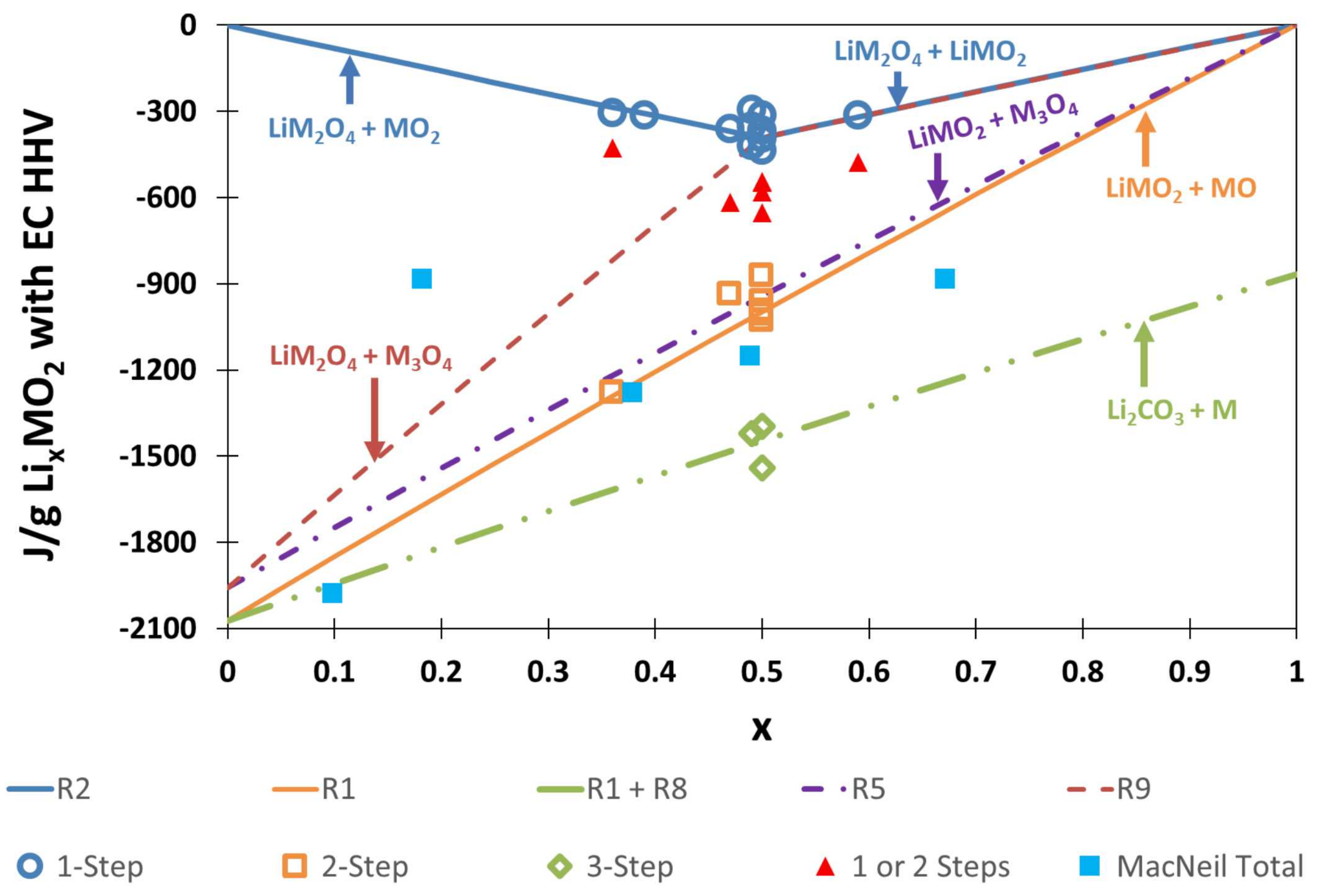
Comparison to Li_xMO_2 Cathode Calorimetry with Electrolyte

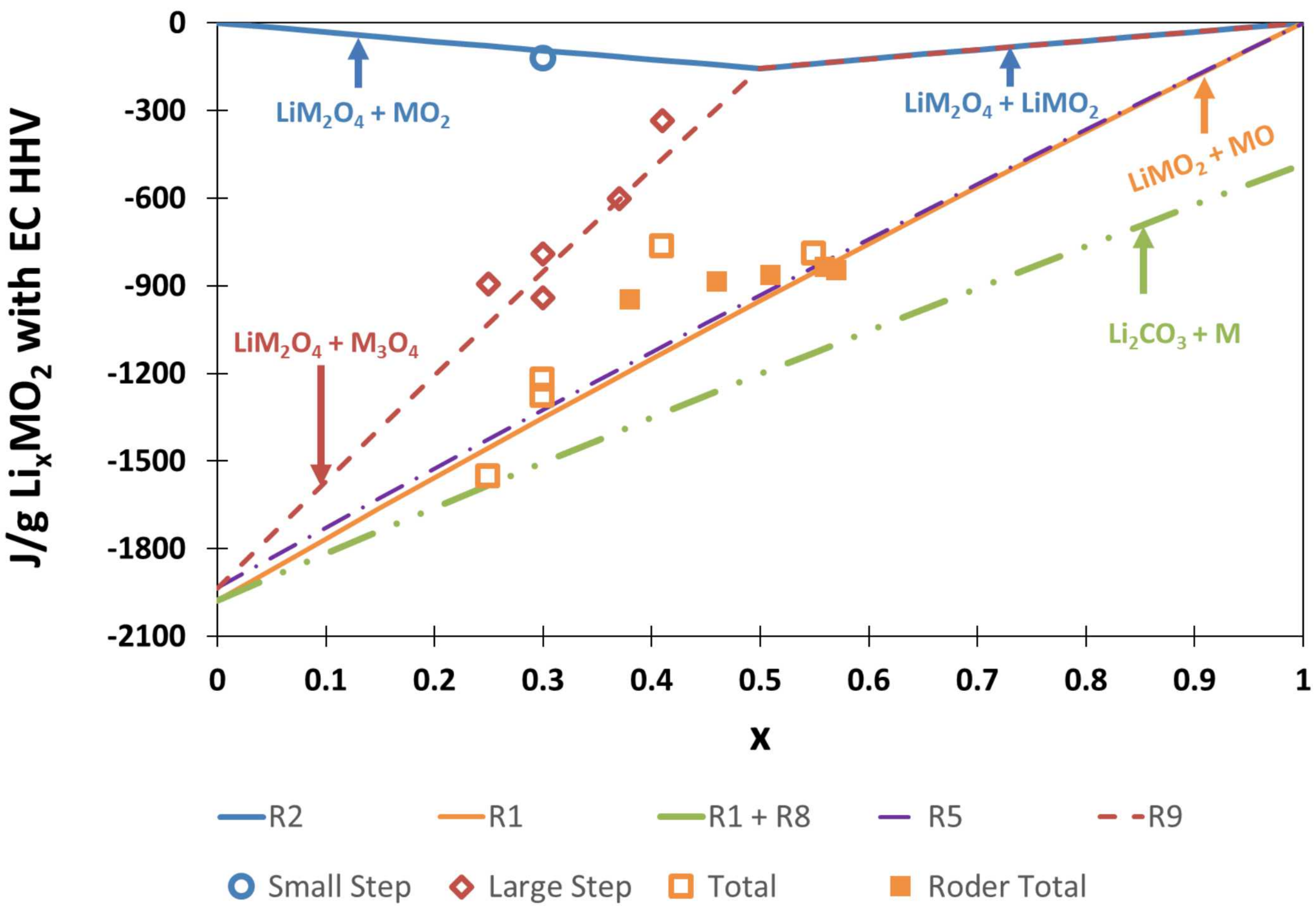
Li_xCoO_2 (LCO) measurements consistent with thermodynamic predictions

- 1st such comprehensive comparison on any Li_xMO_2 materials
- 60 total calorimetry measurements compiled from 24 articles for LCO, NMC, and NCA
  - Each data point shown required careful evaluation and processing for comparisons
- Clearly demonstrates and explains variability observed with state of charge (SOC)
  - SOC Proportional to 1-x



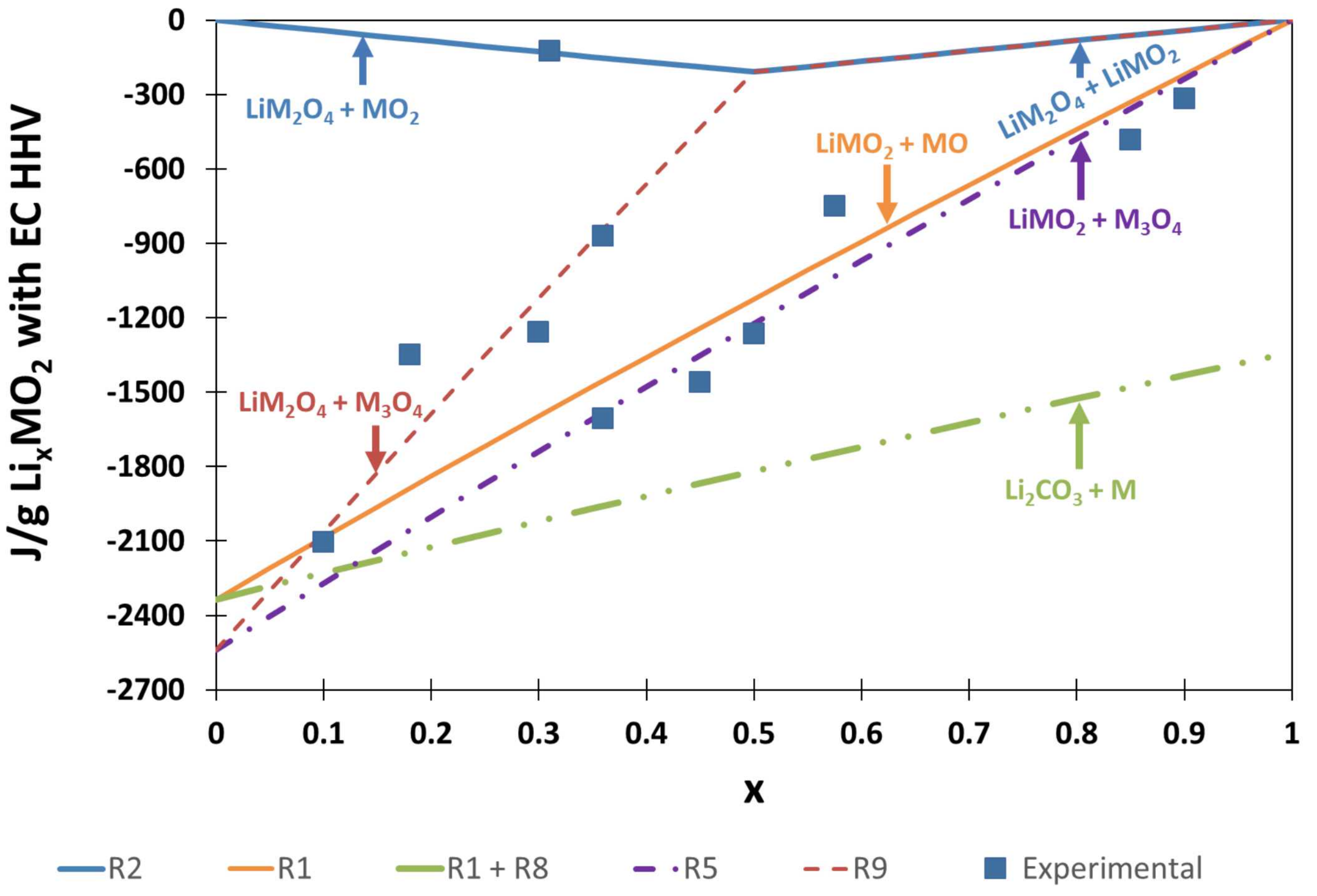
Comparison to Li_xMO_2 Cathode Calorimetry with Electrolyte

$Li_xNi_{0.33}Co_{0.33}Mn_{0.33}O_2$ (NMC 1:1:1) measurements also consistent with predictions



Comparison to Li_xMO_2 Cathode Calorimetry with Electrolyte

$Li_xNi_{0.80}Co_{0.15}Al_{0.05}O_2$ (NCA 80:15:5) measurements also consistent with predictions



60 Calorimetry Measurements from 24 Articles Extracted, Evaluated, and Processed

Literature for LCO Comparisons

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Criteria and Processing for ARC and DSC measurements

- Thermal decomposition of Li_xMO_2 with electrolyte or solvent in pressure-tight containers
- Total and/or stepwise heat release reported per unit mass of Li_xMO_2
- Sufficient information to designate residual degree of lithiation (SOC is proportional to 1-x)
- Sufficient information to correct mass to a binder-free basis

Literature for NMC Comparisons

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- E. P. Roth and D. H. Doughty, *J. Power Sources*, **128**, 308 (2004).
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LOOKING FORWARD

Lithium-Ion Battery Calorimetry Workshops (with full safety team)

- Set up website for sharing and modeling thermal runaway data
- Schedule first workshop, continue recruiting participants

Cathode Decomposition Modeling

- Publish Li_xMO_2 thermodynamics paper
- Set up web-based calculator to estimate total heat release for arbitrary Li_xMO_2 cathodes
- Develop and publish new models for thermal runaway in Li_xMO_2 cathodes

Integrate New Heat Source Models into Cascading Failure Simulations

FY2019 Publication Summary

- R. C. Shurtz, Y. Preger, L. Torres-Castro, J. Lamb, J. C. Hewson and S. Ferreira, "From Calorimetry Measurements to Furthering Mechanistic Understanding and Control of Thermal Abuse in Lithium-Ion Cells" *J. Electrochem. Soc.*, **166**, A2498 (2019).
  - DOI 10.1149/2.0341912jes
- R. C. Shurtz, J. D. Engerer and J. C. Hewson, "Predicting high-temperature decomposition of lithiated graphite: I. Review of phenomena and a comprehensive model" *J. Electrochem. Soc.*, **165**, A3878 (2018).
  - DOI 10.1149/2.0171814jes
- R. C. Shurtz, J. D. Engerer and J. C. Hewson, "Predicting high-temperature decomposition of lithiated graphite: II. Passivation layer evolution and the role of surface area" *J. Electrochem. Soc.*, **165**, A3891 (2018).
  - DOI 10.1149/2.0541816jes
- In Progress:
  - "Heats of Reaction for Thermal Decomposition of Layered Metal Oxides in Electrolyte" by R. Shurtz and J. Hewson

PROJECT CONTACTS

THANK YOU

- Funded by the U.S. Department of Energy, Office of Electricity, Energy Storage program. Dr. Imre Gyuk, Program Director.
- Sandia National Laboratories is a multi-mission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC., a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA-0003525.

For questions about this presentation, contact Randy Shurtz: rshurtz@sandia.gov

For further details, see the following poster:

- Heat Release from Thermal Decomposition of Layered Metal Oxide Cathodes in Lithium-Ion Batteries
  - Randy Shurtz, John Hewson