Pacific Northwest NATIONAL LABORATORY

October 11, 2021

Na-ion batteries: Development and Scaling-up of Advanced Cathode Materials

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#304 Sodium Batteries
DOE-OE Peer Review
Oct 26-28, 2021

U.S. DEPARTMENT OF ENERGY BATTELLE

PNNL is operated by Battelle for the U.S. Department of Energy

Project team

- **PNNL Contributors**
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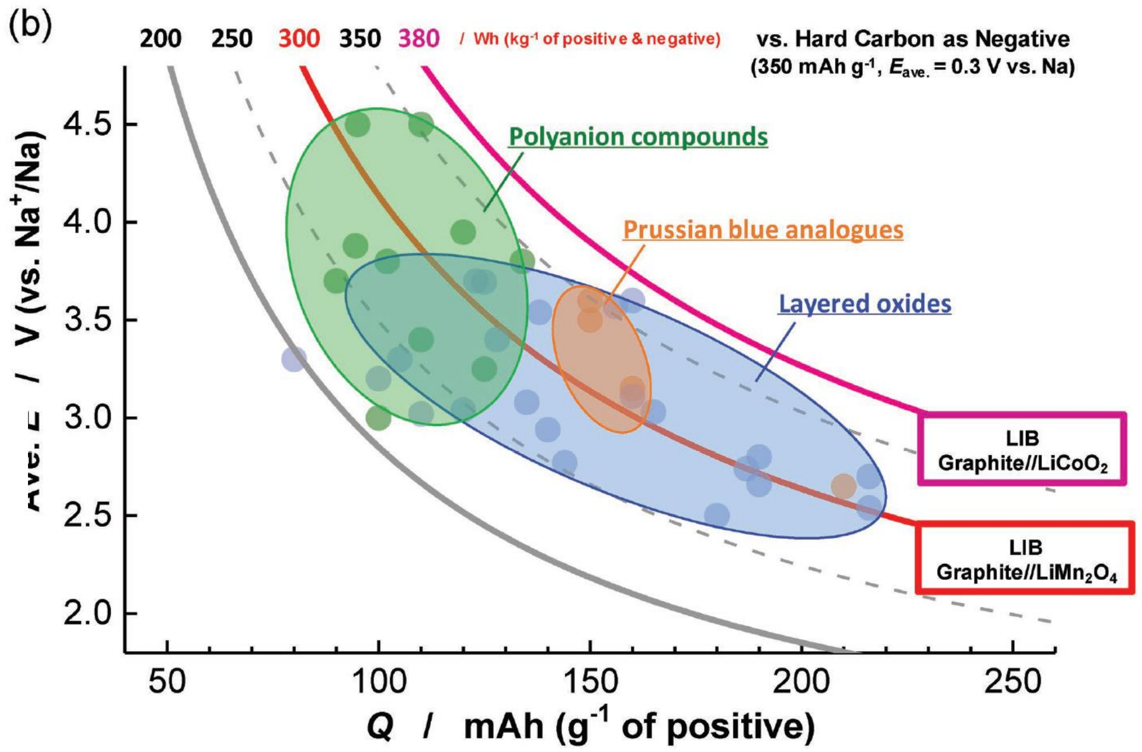
  - Namhyeong Kim
  - Hee-Jung Chang
  - Hyungkyu Han
  - Bhuvaneswari Sivakumar
  - Matthew Fayette
  - David Reed
  - Vincent L. Sprenkle

- **External collaborators**
  - Dr. Guiliang Xu (Argonne National Lab)
  - Prof. Donghai Wang (Penn State University)
  - Prof. Xin Li (Harvard University)
  - Dr. Jianlin Wang (Oak Ridge National Lab)
  - Dr. Wanli Yang (Lawrence Berkeley National Lab)
  - Prof. Yanyan Hu (Florida State University)
  - Prof. Feng Lin (Virginia Tech University)

Project overview



- ❑ Na source is much cheaper than Li
- ❑ SIBs are comparable to some LIBs.

Stationary Energy Storage

E-bikes

Electric Vehicles

- ❑ Low-cost materials for sodium-ion batteries
- ❑ Materials scaleup and pouch cell assembly
- ❑ Mechanism study

S. Komaba et al., Adv. Energy Mater., 2018, 8, 17, 1703415   Statista.com, Interatec

Project objectives

- Develop Co-free layered cathode materials with reduced amount of Ni.
- Understanding of the material structure.
- Large cell performance evaluation.

Project milestones in FY21

- **Milestone 1.**

Develop at least one Co-free cathode material that can deliver > 120 mAh/g specific capacity and >80% retention over 100 cycles.
Due Date:03/31/2021

- **Milestone 2.**

Demonstrate >50 mAh single layer pouch cell using Co-free cathode and hard carbon anode
Due Date:06/30/2021

- **Milestone 3.**

Publishing 2 high impact journal articles on advanced Na-ion battery materials

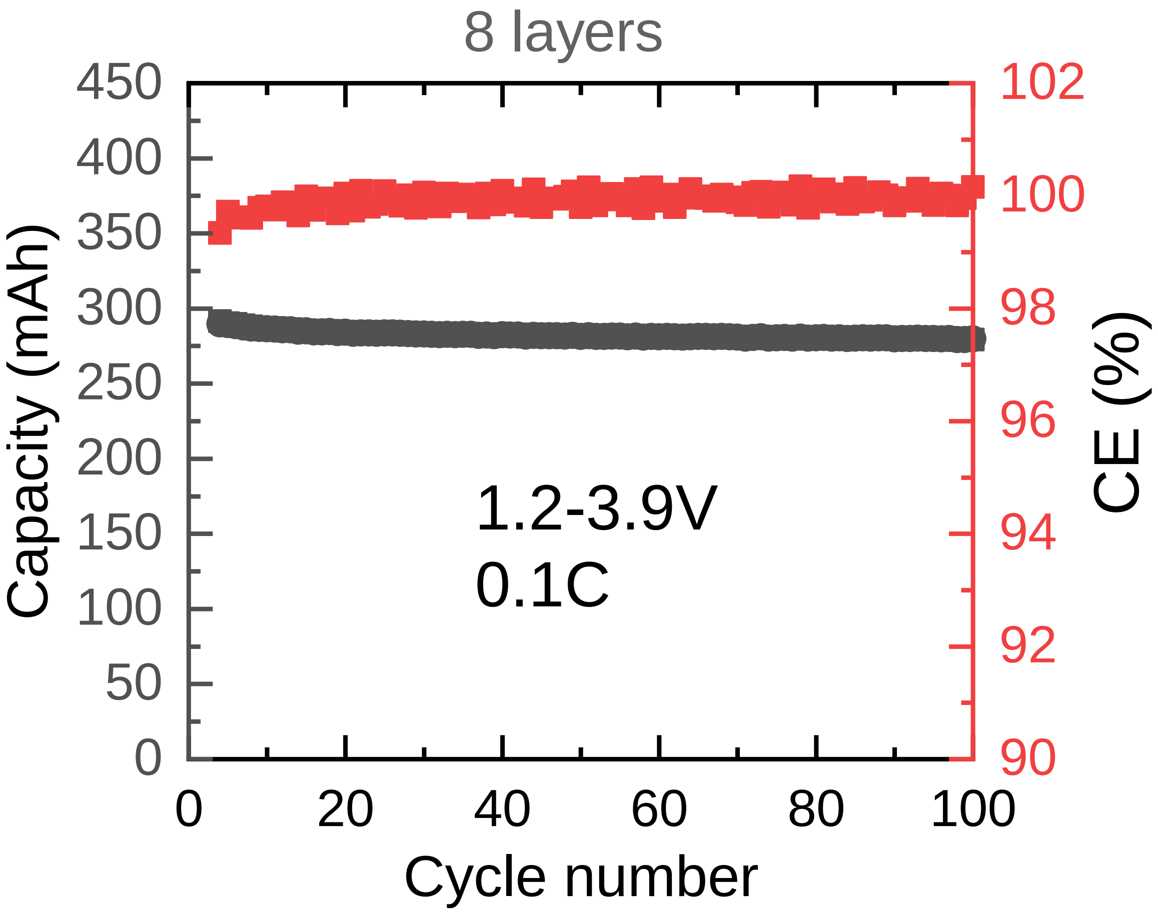
Project achievements in FY21

- **Research highlights**
  - Gen 2 (Ni-rich NMC cathode) material synthesis scaled up to 1 kg/batch level, multi-layer pouch cell assembly was scaled up to ~ 1Ah.
  - Gen 3 (Ni-low, Co-free cathode) material synthesis scaled up to 50g/batch, single-layer pouch cell assembly achieved 50 mAh.
  - Discovered new mechanisms for promoting cathode performance.
- **Publications**
  - Nano Energy, 2021, 89, 106371
  - Angew. Chem. Int. Ed., 2021, 60, 8258
  - Nanotechnology, 2021, 32, 42251
  - ACS Energy Lett., 2021, 6, 547 (co-first author)
  - filed one invention disclosure
- **New collaborations established**
  - Dr. Wanli Yang at Advanced Light Sources, Lawrence Berkeley National Laboratory
  - Dr. Jianlin Li at Oak Ridge National Laboratory
- **Professional activities**
  - Co-organized the ACS Spring Meeting Battery Symposium (March 2021)
  - Will give one oral presentation at the ECS 2021 fall meeting.

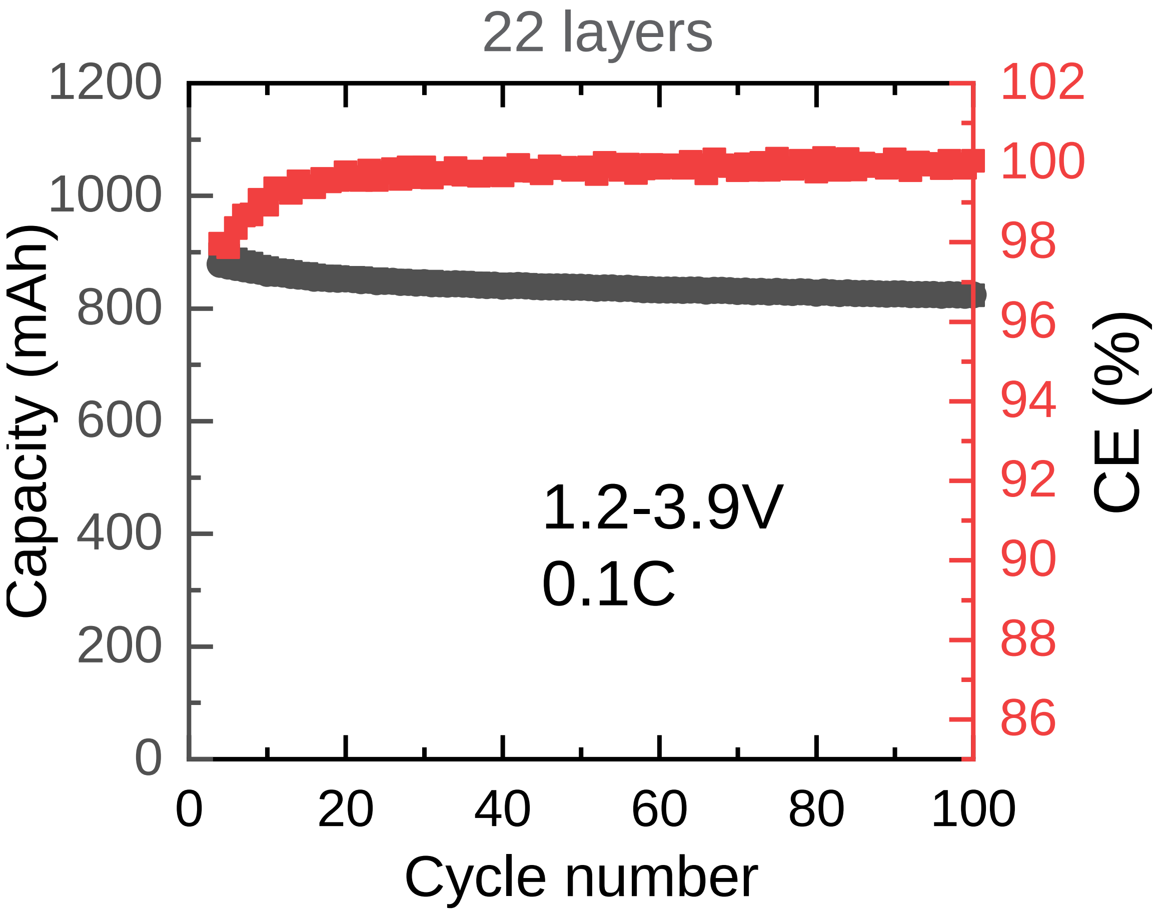
Project achievements in FY21 (1)

Gen 2 Ni-rich NMC: O3-$NaNi_{0.68}Mn_{0.22}Co_{0.10}O_2$ synthesis scaled up to 1 kg level. Multilayer pouch cells assembled.

Electrode coated by ORNL
Cell fabricated using the ABF at PNNL



290 mAh capacity
93.5% retention after 100 cycles



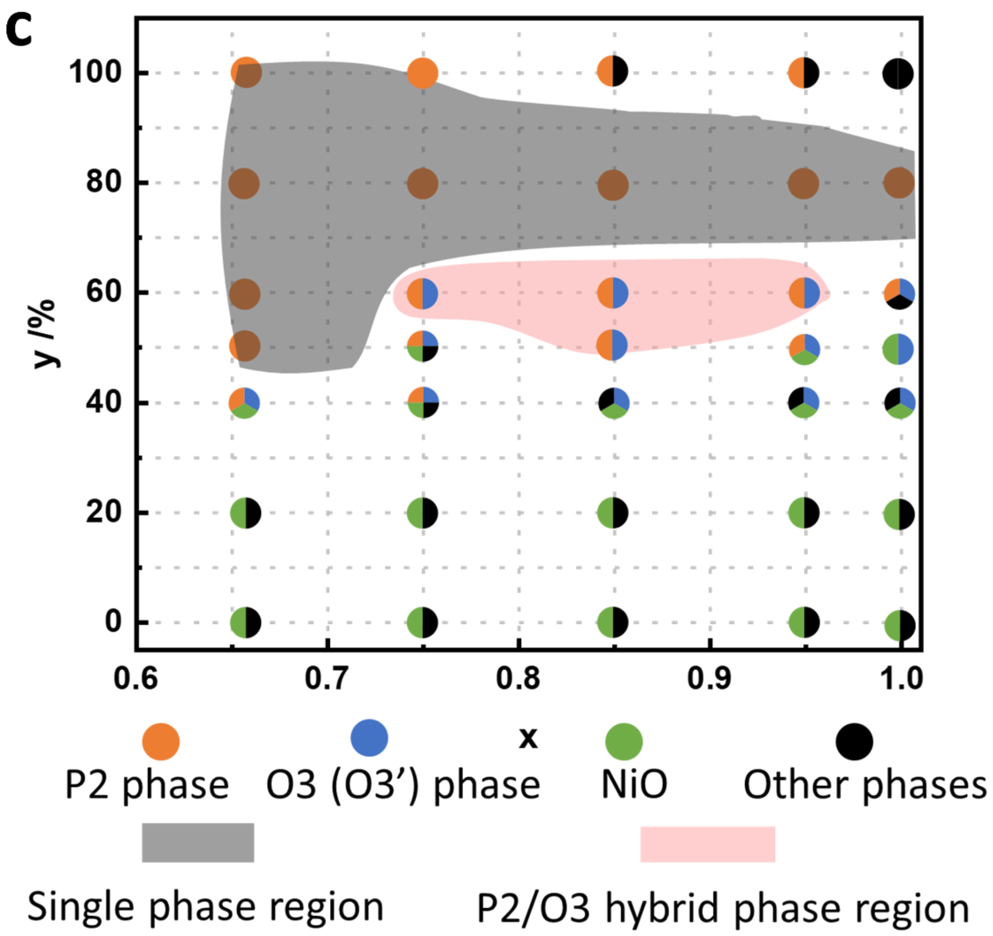
900 mAh capacity
93.7% retention after 100 cycles

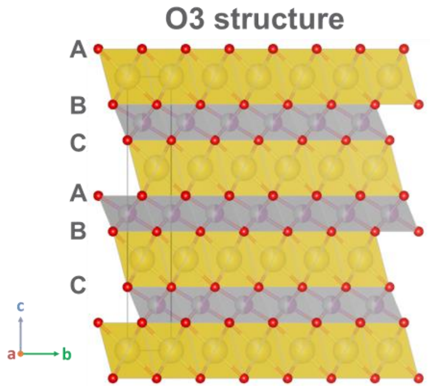
PNNL unpublished work

(retention calculated based on capacity after 3 formation cycles)

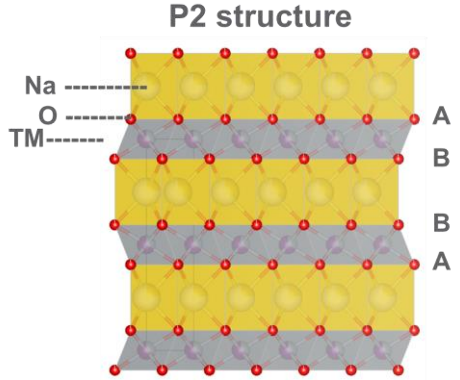
Project achievements in FY21 (2)

Phase diagram of $Na_xMn_yNi_{1-y}O_2$ – Roadmap to low Ni content layered cathode





Pros:
High Na stoichiometry
Cons:
Adverse phase transition
High Na diffusion barrier



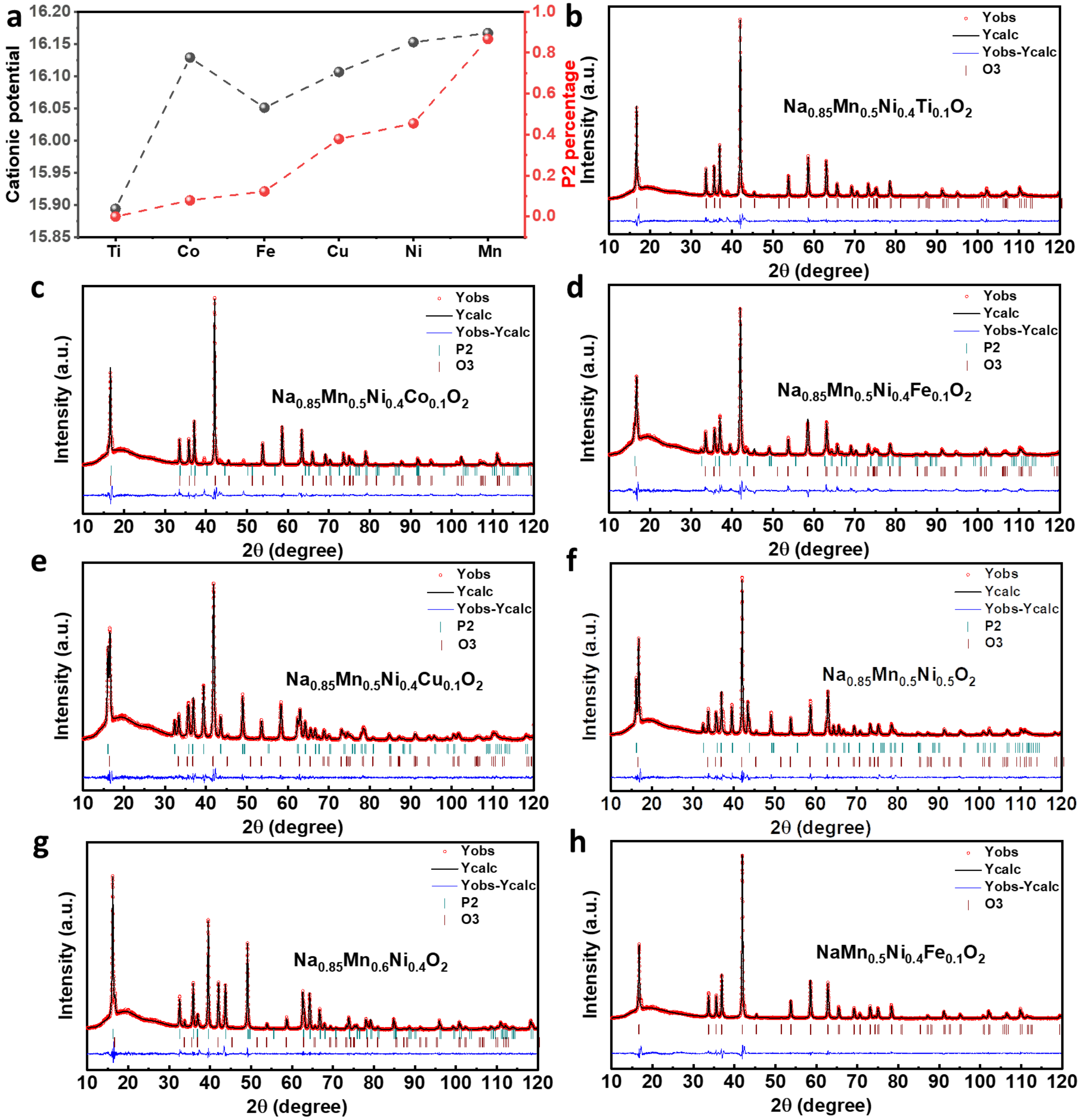
Pros:
Low Na diffusion barrier
Cons:
Adverse phase transition
Low Na stoichiometry

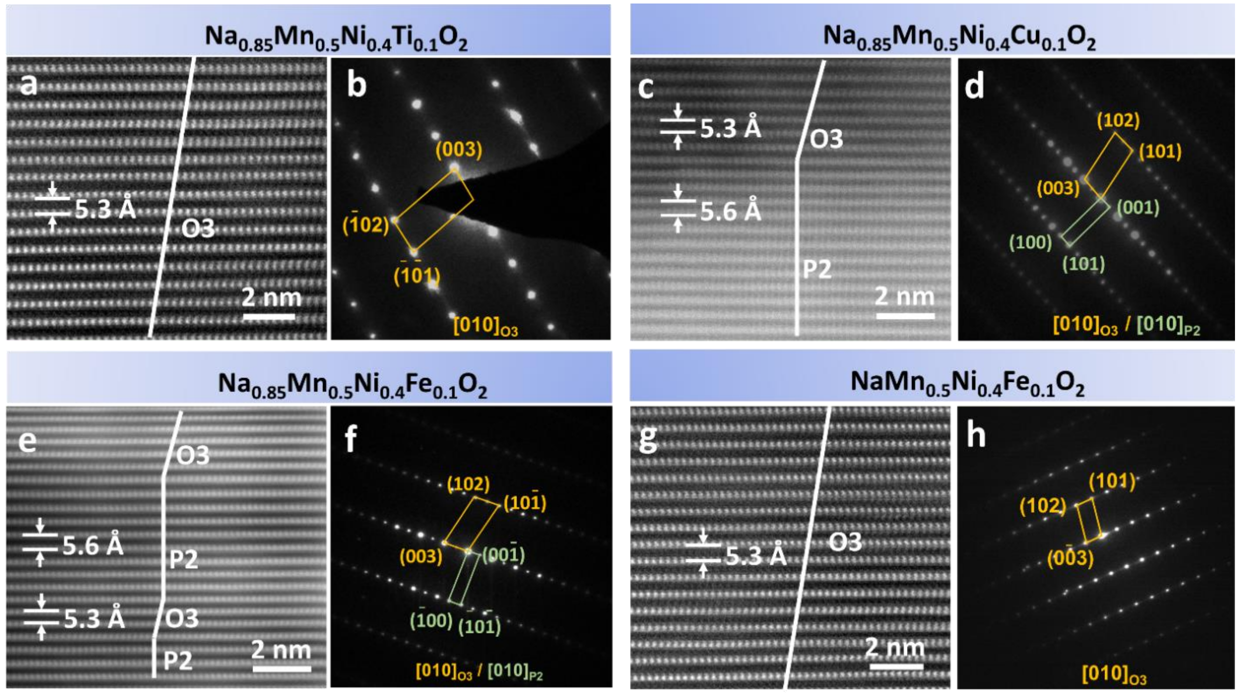
- Synthesis phase diagram of $Na_xMn_yNi_{1-y}O_2$ was constructed.

Nano Energy, 2021

Project achievements in FY21 (2)

Phase construction with alien element doping: $Na_{0.85}Mn_{0.5}Ni_{0.4}M_{0.1}O_2$

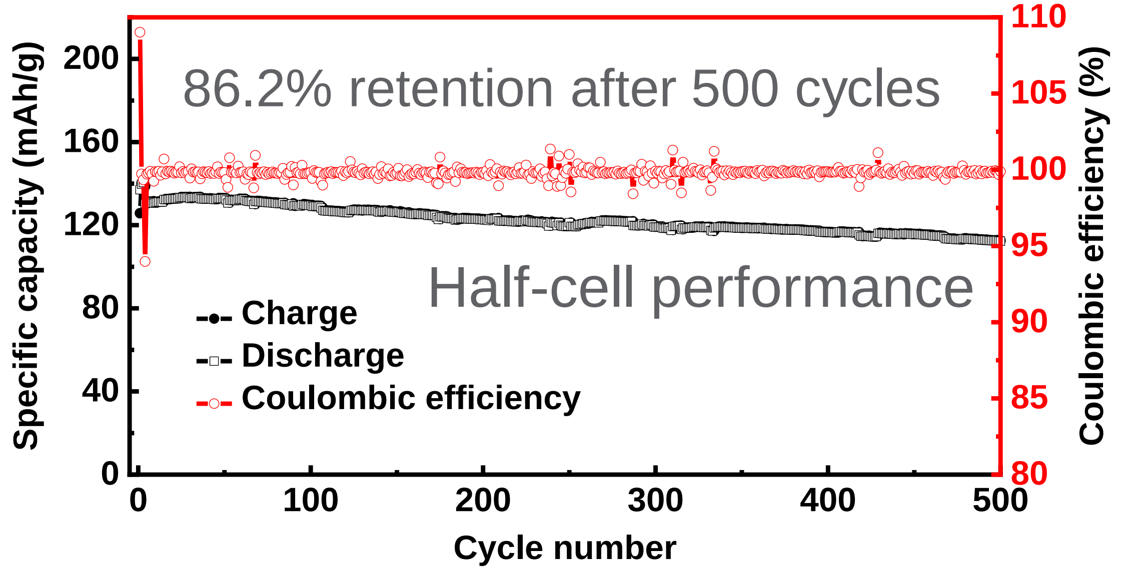


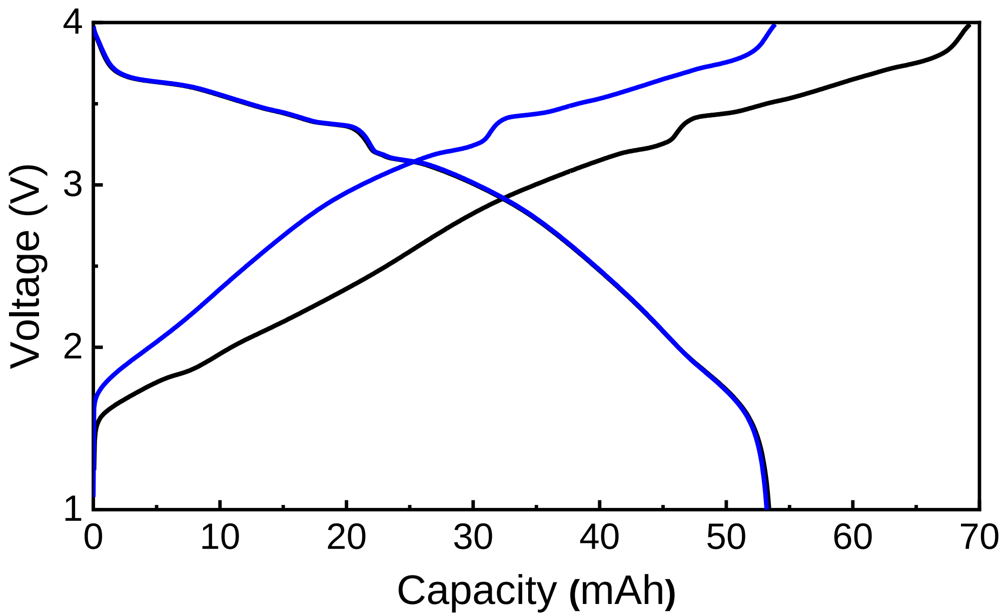


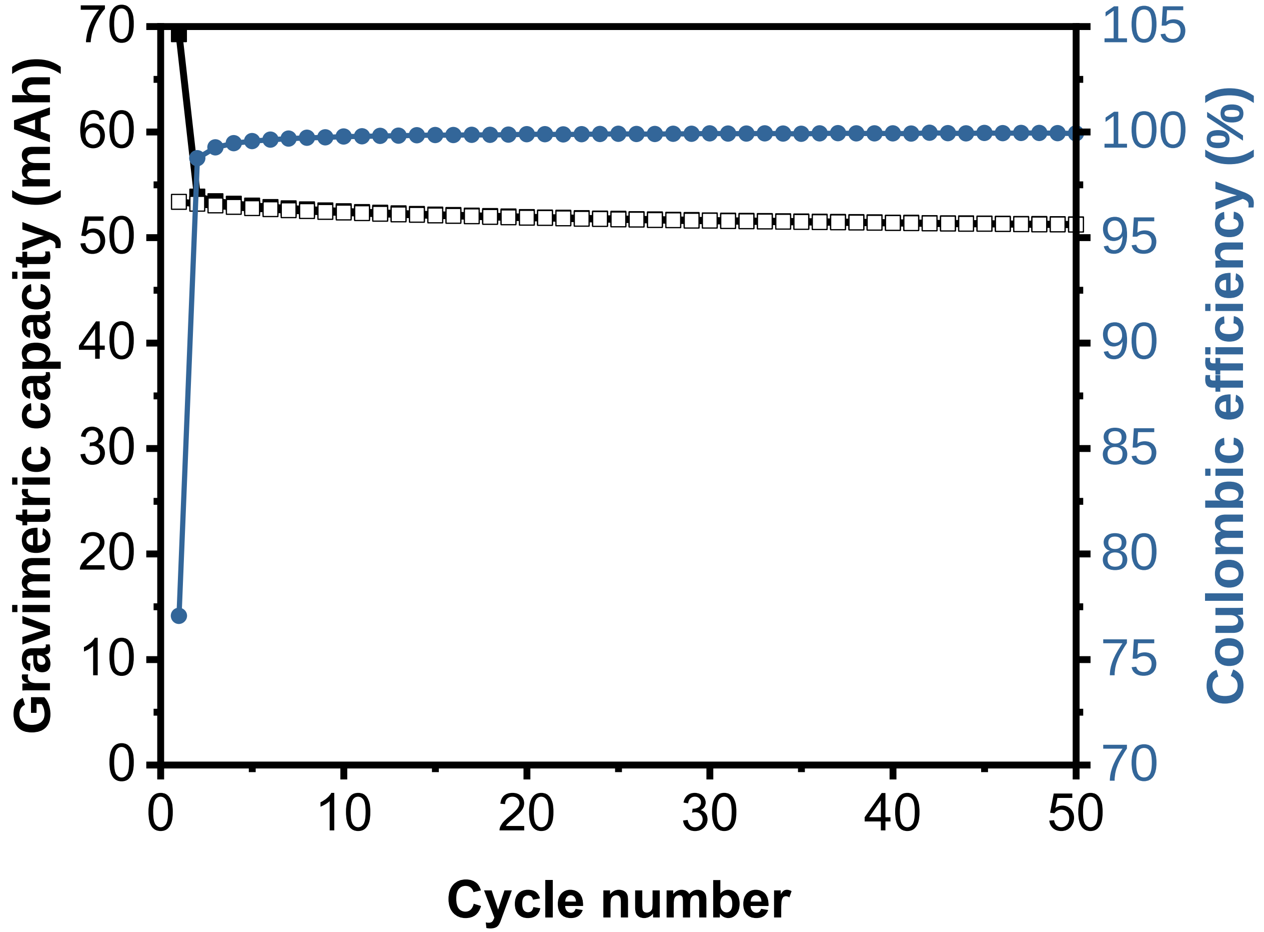
- ❑ Materials with different ratios of P2/O3 structures were synthesized via element doping.
- ❑ The P2/O3 structures co-exist in the same particle as revealed by high-resolution TEM.

Project achievements in FY21 (2)

$Na_{0.85}Mn_{0.5}Ni_{0.4}Fe_{0.1}O_2$-HC single-layer pouch cell assembly





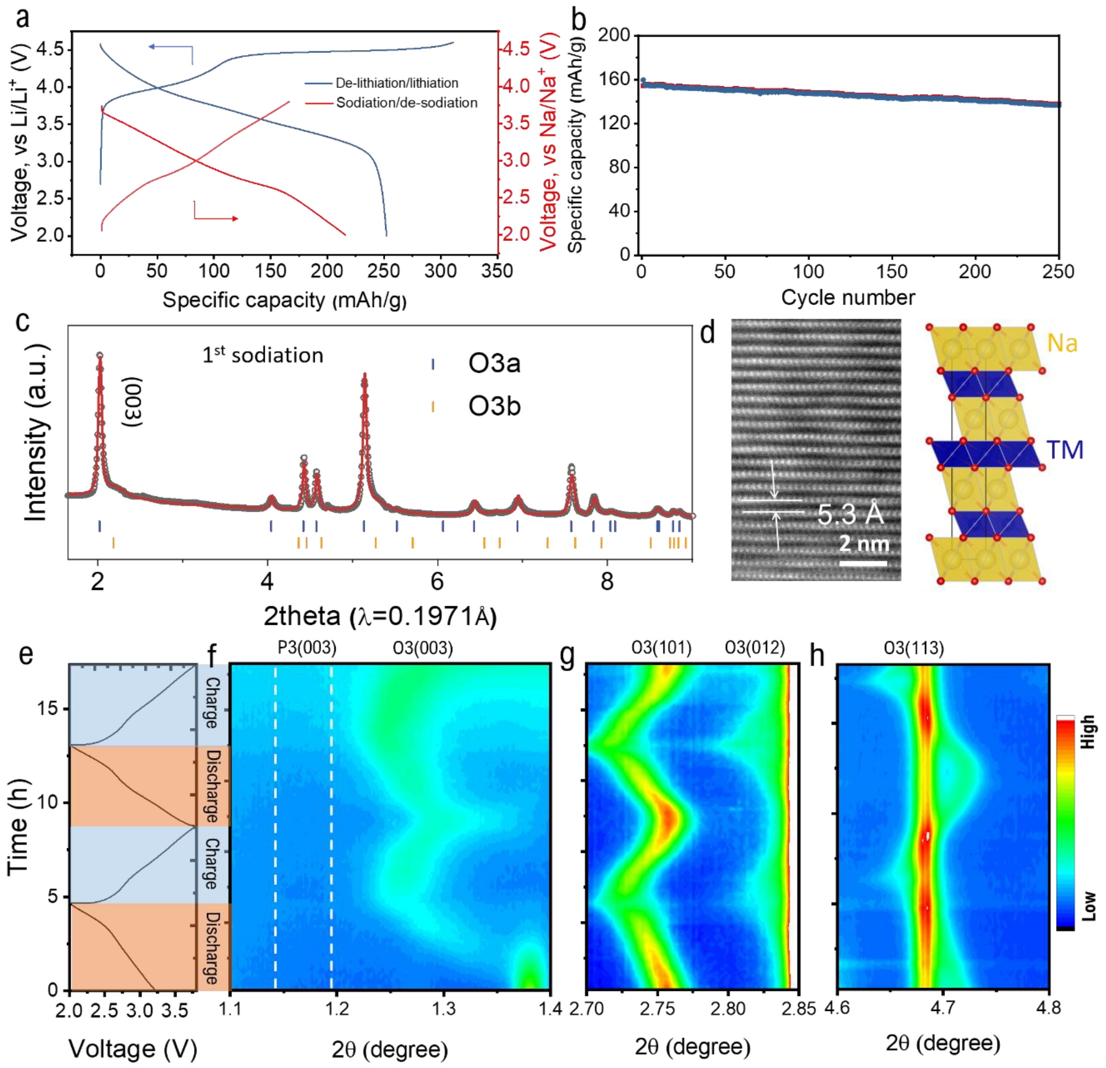


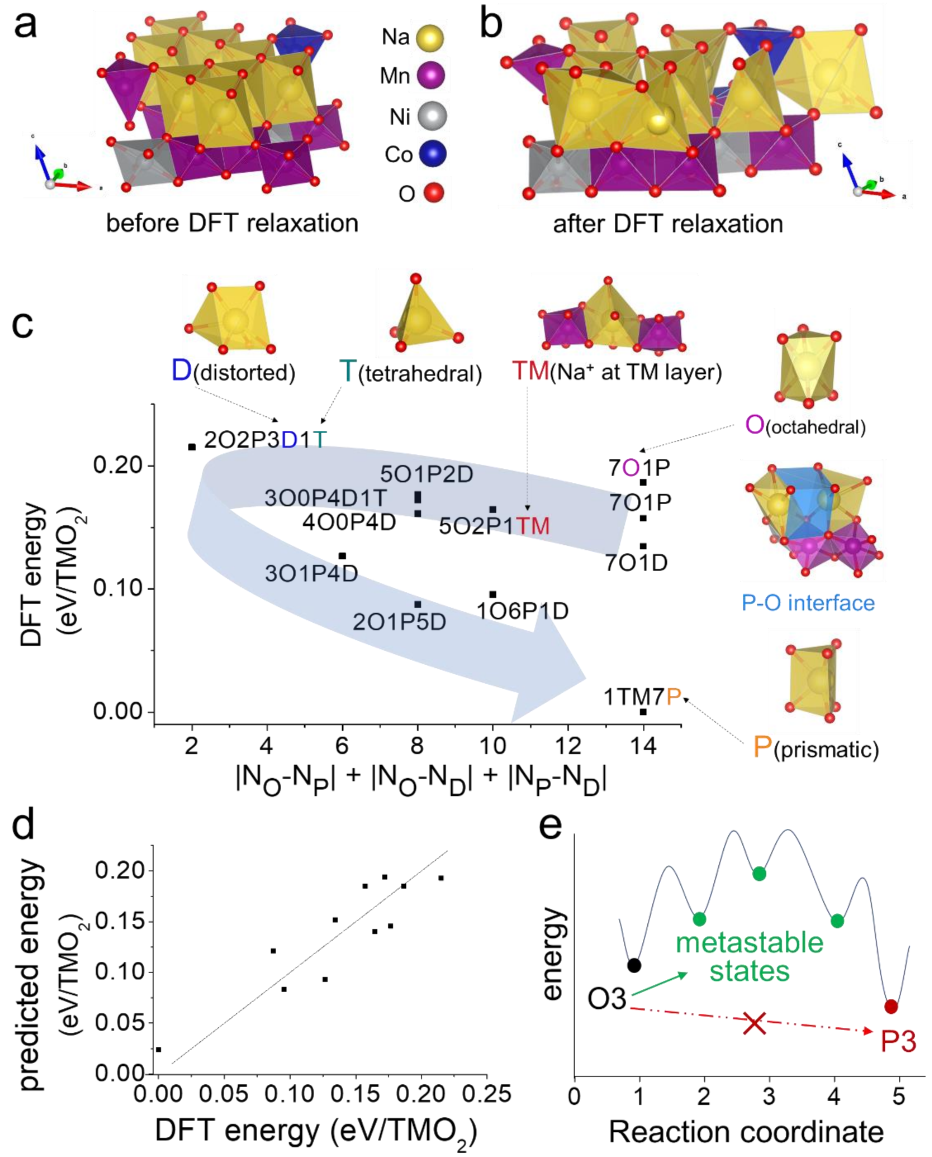
- ❑ Half cell shows 130 mAh/g capacity and 86.2% retention after 500 cycled.
- ❑ Pouch cell reaches 53 mAh capacity and 96% retention after 50 cycles. (Milestone reached).

Project achievements in FY21 (3)

New mechanisms aimed at promoting performance proposed

Realizing high-performance O3-structured Mn-rich layered cathode *via* oxygen and transition metal vacancies (Angew. Chem. 2021)





- ❑ 160 mAh/g capacity reached, higher than similar materials.
- ❑ Solid-solution structural evolution during cycling.
- ❑ Intermediate phases responsible for the solid-solution reaction by DFT calculation.

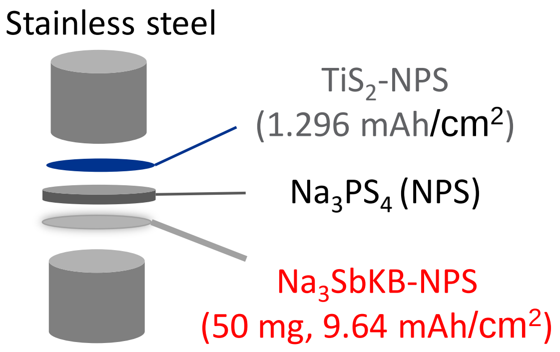
Project achievements in FY21 (4)

All-solid-state sodium-sulfur batteries Na-alloy anode and sulfur cathode

Safety **Cost**

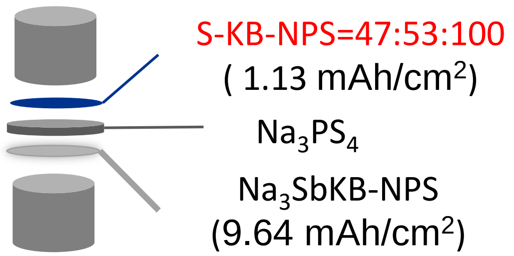
Prof. Donghai Wang

- Stable cycling of Na_3**Sb**KB-Na_3PS_4/TiS_2 solid-state battery cells

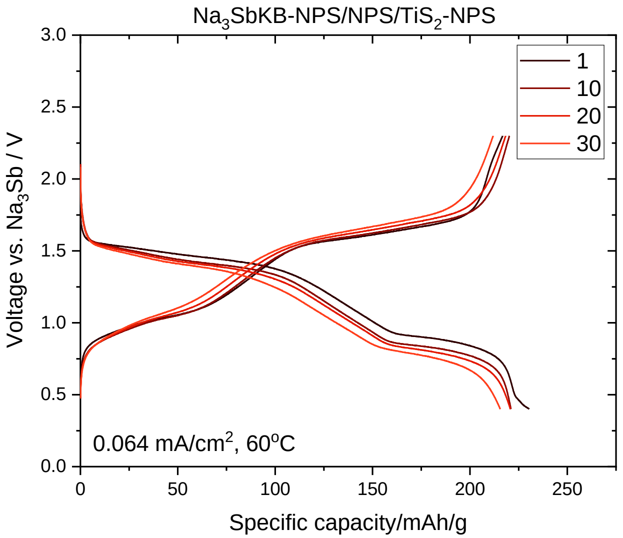


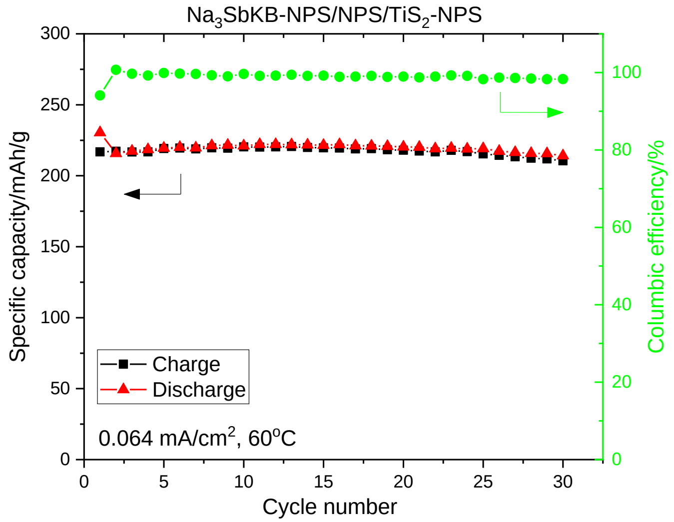
- ❑ Na-Sb alloy anode works well.
- ❑ Stable cycling with over 210 mAh/g capacity.

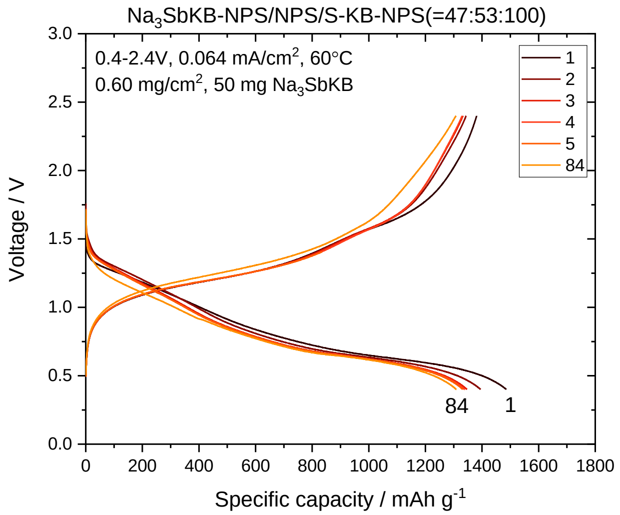
- Electrochemical performance of all-solid-state Na alloy-S batteries

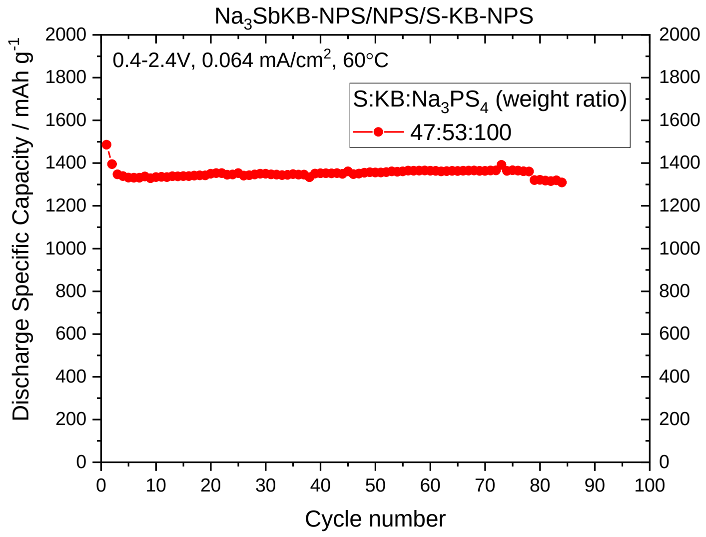


- ❑ Low-cost sulfur-based cathode
- ❑ Low temperature operation (60 ^{o}C)
- ❑ Stable cycling of Na-Sb alloy-Sulfur Batteries









Proposed work for FY22

- Scaling up P2/O3 hybrid materials to kg level and assemble multilayer pouch cells.
- Further reducing the amount of Ni while maintaining or increasing the capacity.
- Fundamental cathode material performance promoting method and degradation mechanism investigations.

Acknowledgements and project contacts

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Thanks **Dr. Imre Gyuk**, Energy Storage Program Manager

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Thank you!