Dry Pressed, High Areal Loading Electrode Architectures Enabled by Holey Graphene

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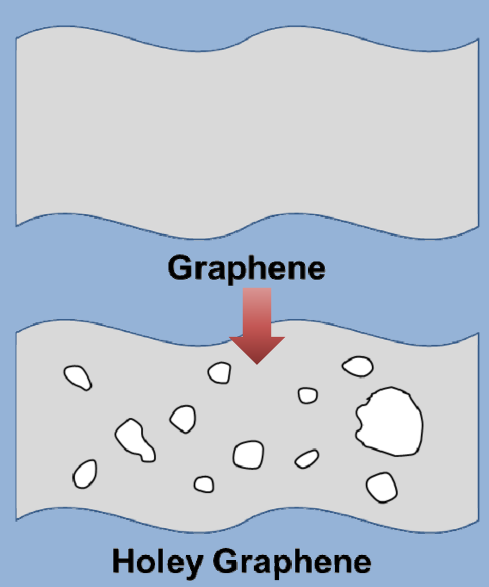
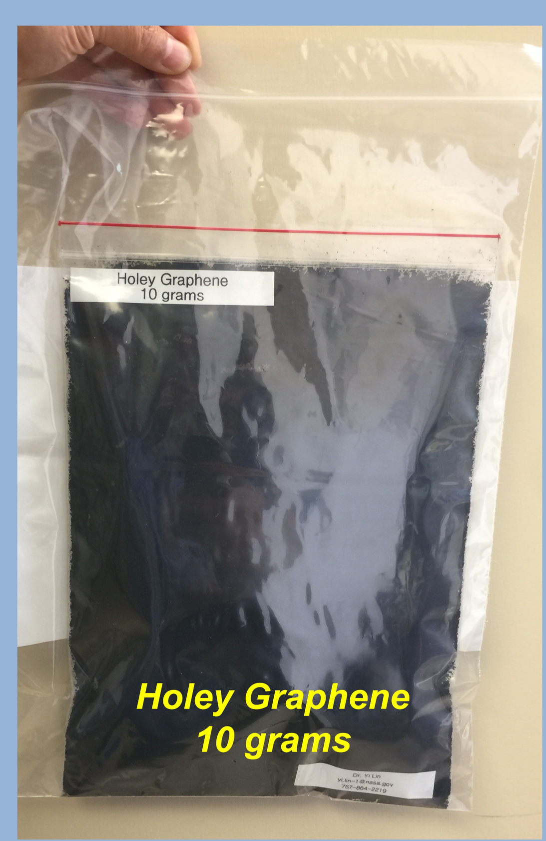
Graphene is known to be an advanced carbon additive in novel battery electrodes with good electrical conductivity, high surface area, and mechanical robustness to maximize the electrochemical function of the active materials. Holey graphene, a structural derivative of graphene with an array of through-the-thickness holes across the lateral surface of the nanosheet, allows more facile cross-plane ion and gas transport than intact graphene, making it an ideal electrode material for electrochemical energy storage. A unique property of holey graphene is that it can be compression molded into robust articles or architectures under solvent-free conditions without the need for parasitic binders. This unique dry compressibility of holey graphene has enabled facile dry fabrication of high mass loading electrodes with both high density and high porosity and controlled architectures, which have found use in a variety of energy storage applications including, but not limited to, supercapacitors and lithium-oxygen (Li-O_2), lithium-sulfur (Li-S), lithium-selenium (Li-Se), and various Li ion battery systems.

Scalable Synthesis

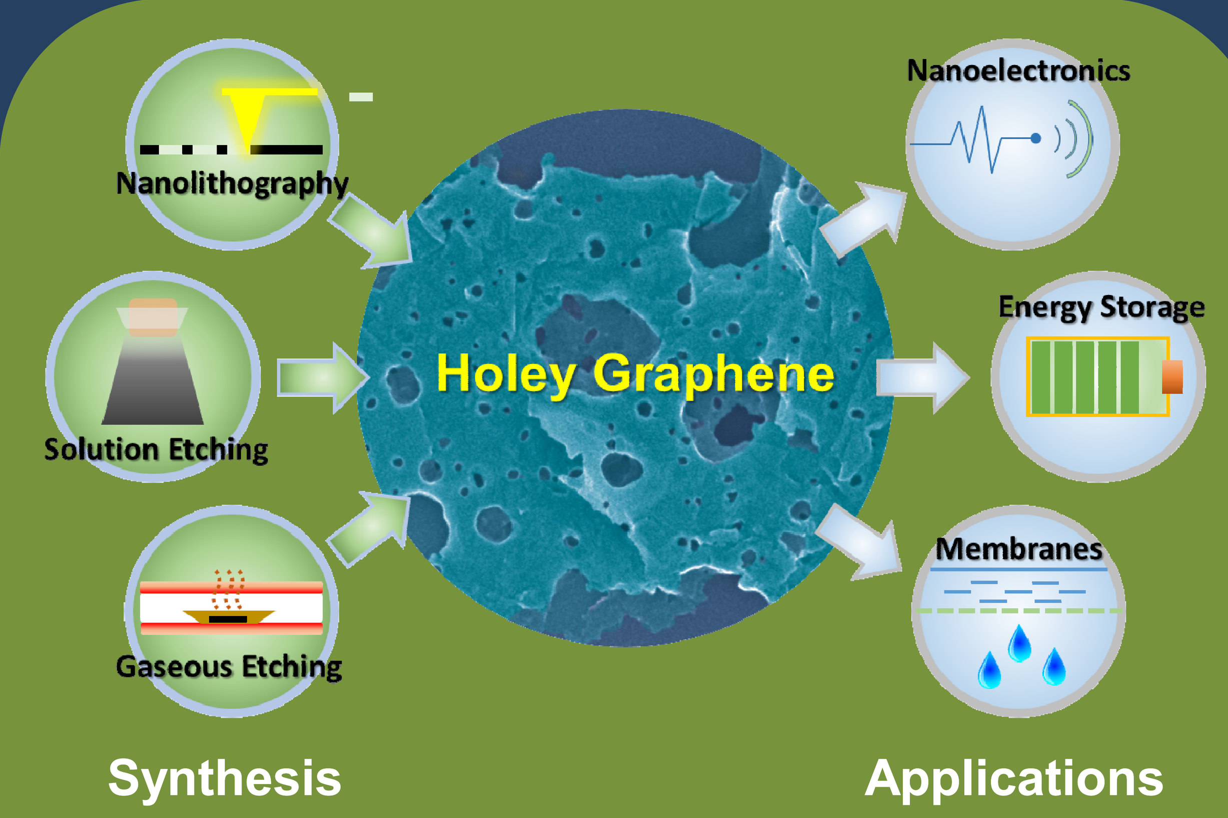
Catalysis-Free Air Oxidation





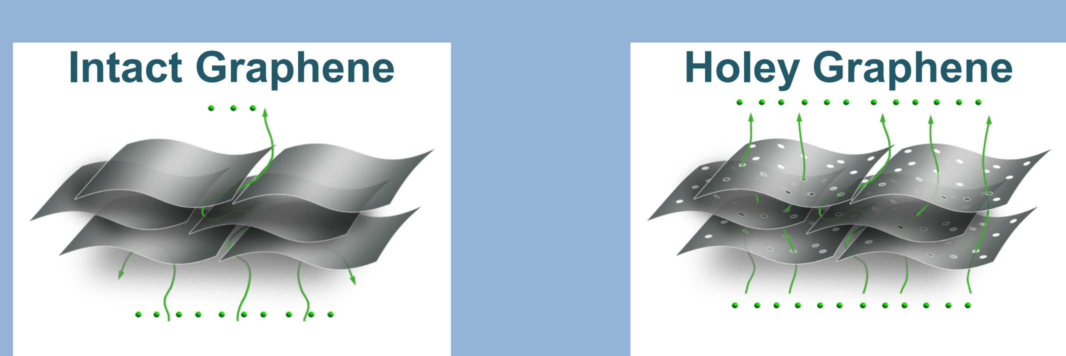
- ✓ Open furnace
- ✓ 400 – 500 °C
- ✓ 1 – 10 h

ACS Nano **2014**, *8*, 8255;
Adv. Func. Mater. **2015**, *25*, 2920.



Mater. Res. Lett. **2017**, *5*, 209.

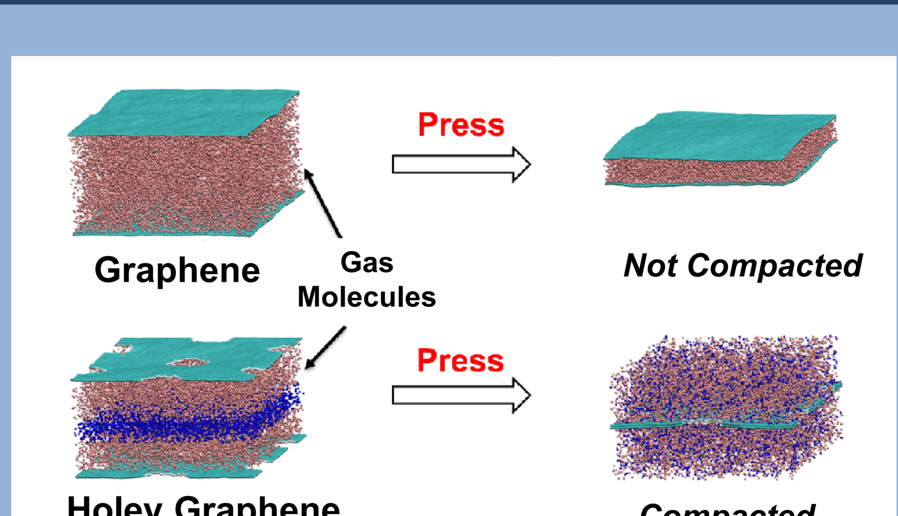
An Ideal Electrode Material



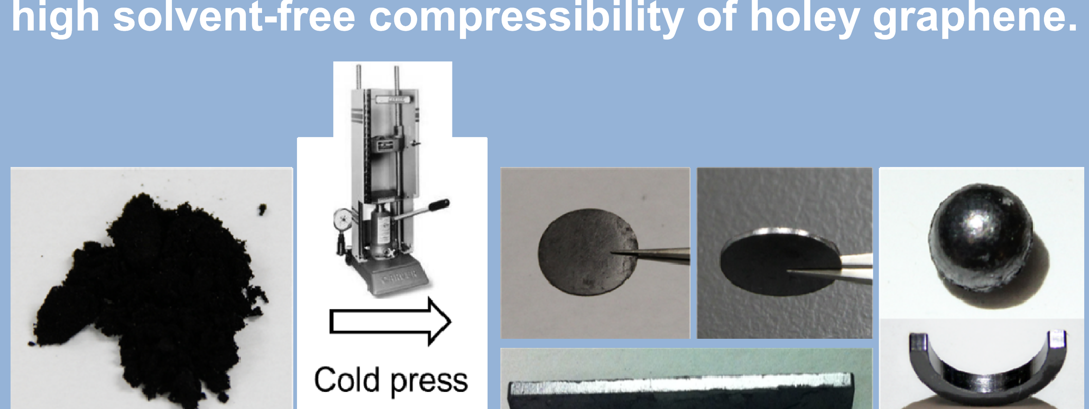
- ❑ In-plane porosity: Improve ion transport path
- ❑ Accessible surface area: Improve gravimetric capacitance
- ❑ Volume reduction: Mitigate need to create large pores/spacing
- ❑ Hole-edge functionalities: Improve mesoscale wetting
- ❑ Electrical Conductivity: Retain graphitic crystallinity

ACS Nano **2014**, *8*, 8255.

Unique Dry Compressibility



- ❑ The presence of nanometer holes on graphene sheet allows the facile escape of air molecules, enabling the high solvent-free compressibility of holey graphene.

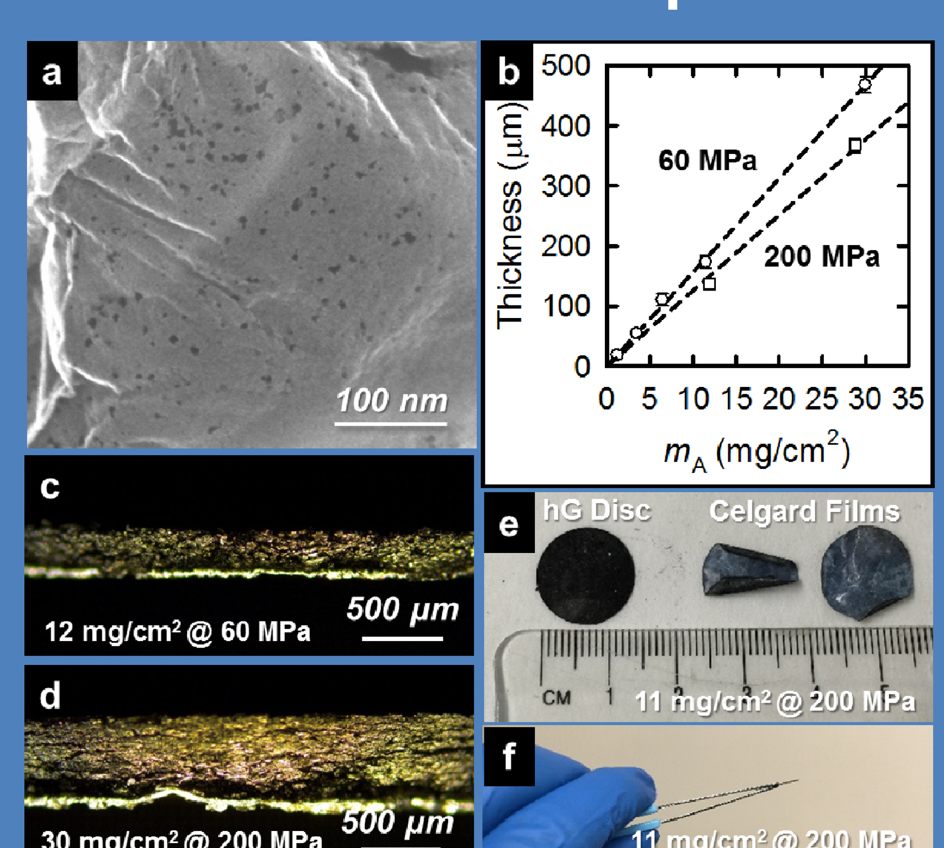


- ❑ Dense graphene architectures (density as high as 1.4 g/cm^3) can be facilely prepared within minutes.

ACS Nano **2017**, *11*, 3189.

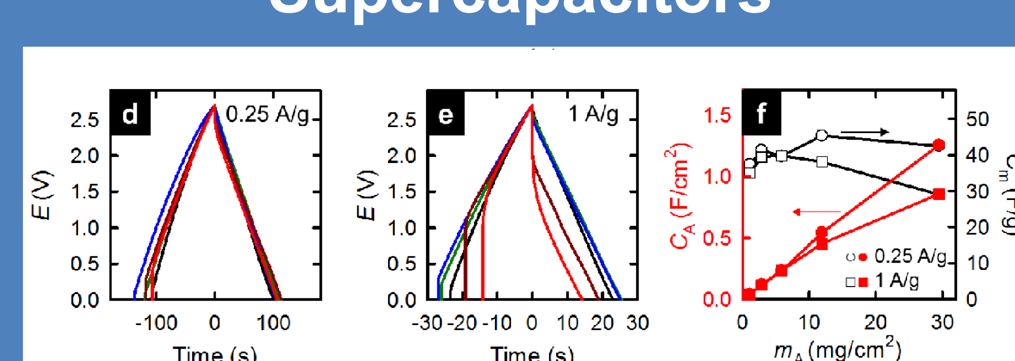
High Mass Loading Neat Carbon Electrodes

Facile Electrode Preparation



- ✓ Ultrahigh areal loading (up to 30 mg/cm^2)
- ✓ Solvent-free
- ✓ Binder-free
- ✓ Ambient temperature
- ✓ Completion within minutes

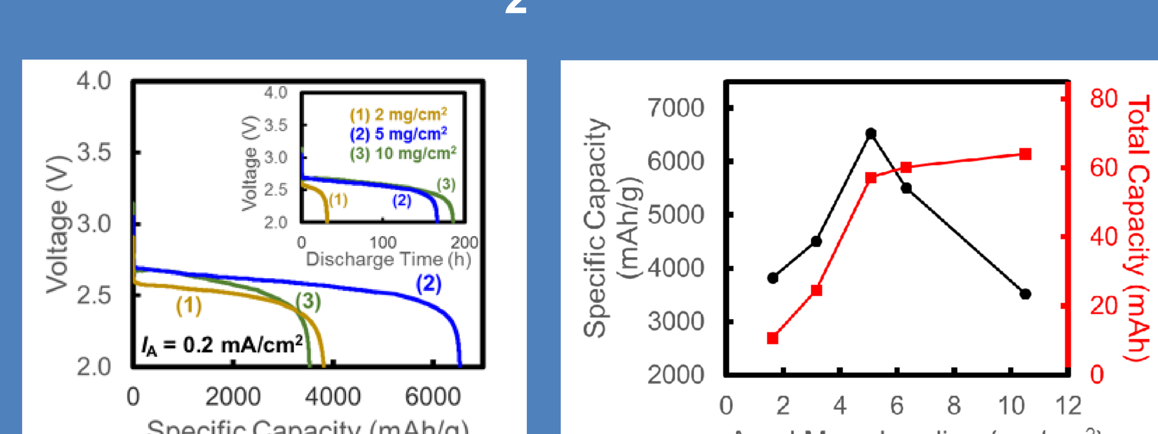
Supercapacitors



- ❑ Little variation of C_m with increase of mass loading.
- ❑ Near-linear increase of C_A with mass loading.

ACS Appl. Mater. Interfaces **2016**, *8*, 29478.

Li-O_2 Batteries

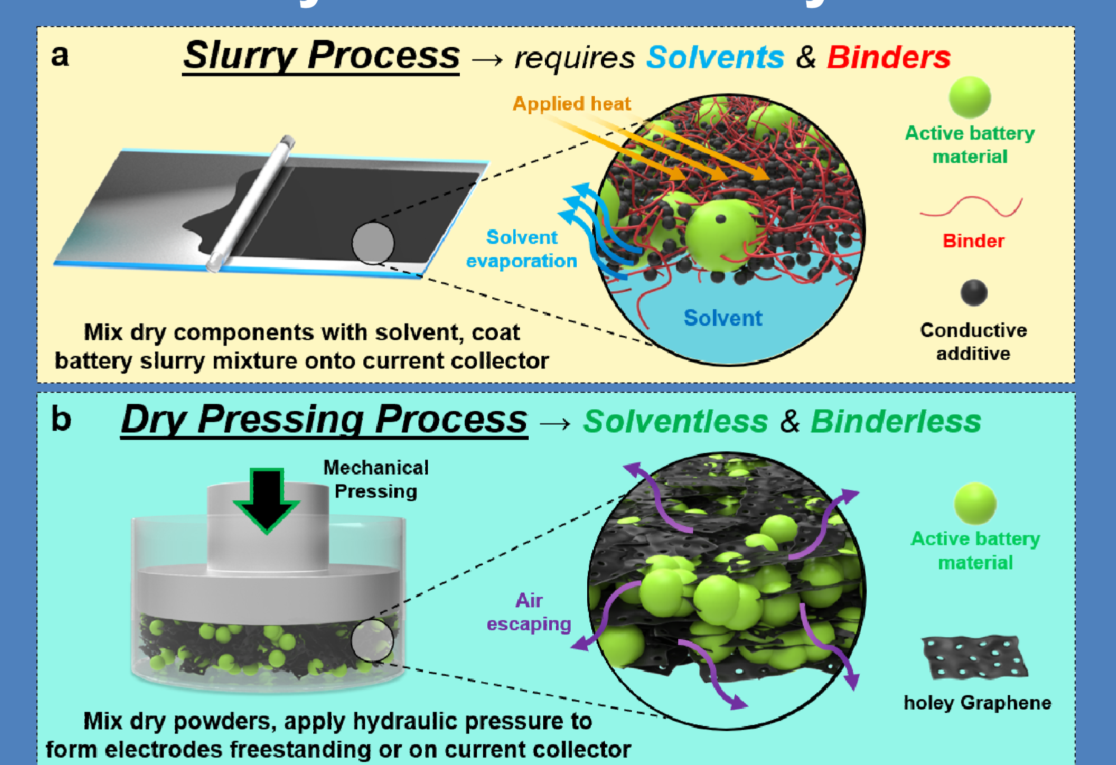


- ❑ Little variation of C_m with increase of mass loading.
- ❑ Near-linear increase of C_A with mass loading.

Nano Lett. **2017**, *17*, 3252.

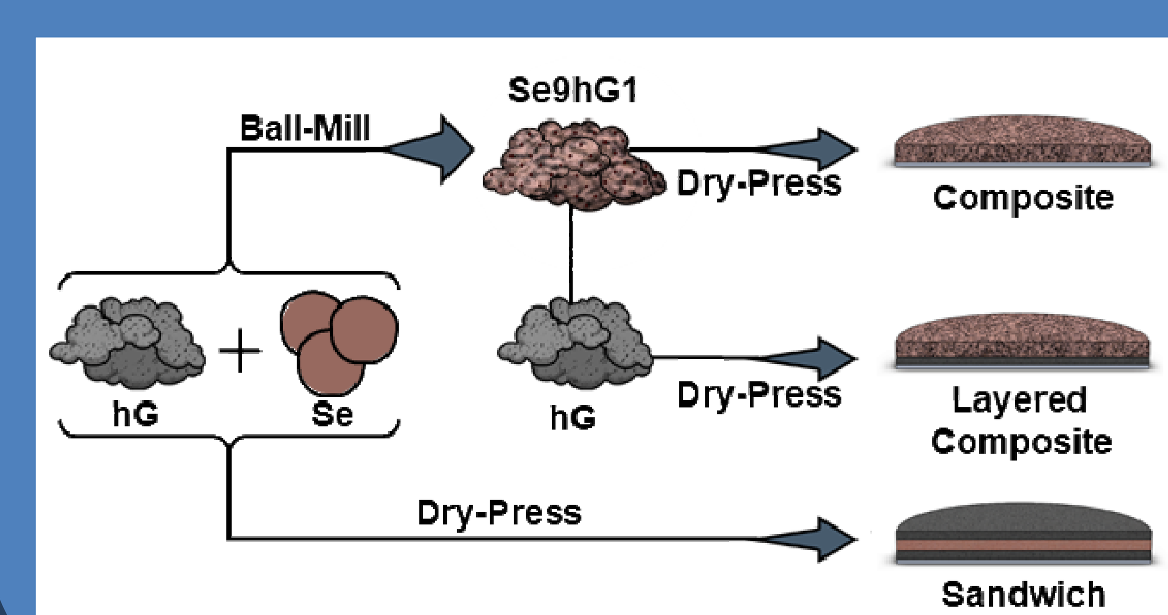
Facile, Solvent-Free Preparation of Composite Electrodes

Dry Press vs. Slurry



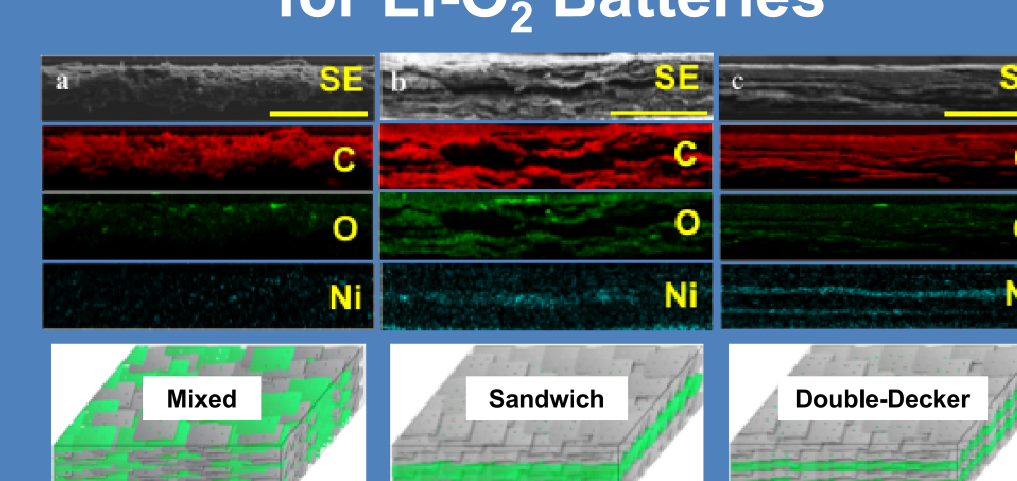
ACS Appl. Energy Mater. **2019**, *2*, 2990.

Electrode Architecture Variations



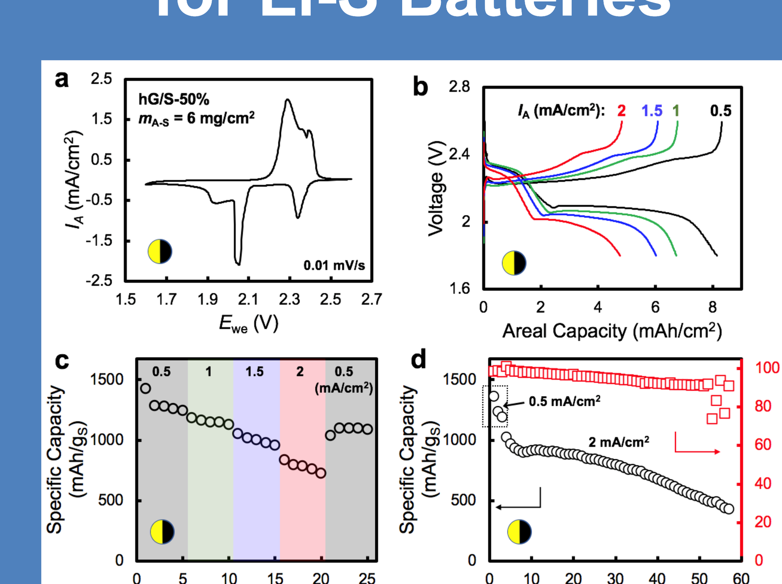
High Mass Loading Composite Electrodes

Composite Catalytic Air Cathodes for Li-O_2 Batteries



Nano Energy **2017**, *31*, 386.

Composite S Cathodes for Li-S Batteries



Batteries & Supercaps **2019**, *2*, 774.

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