Abstract
This work aims to create new electrode architectures via scalable fabrication methods that enable higher active materials density and reduced inactive content compared to today’s technology, while satisfying the duty cycles of vehicle applications. The specific approach uses magnetic alignment to produce low-tortuosity pore structures in dense, low- or zero-additive electrodes.

Introduction
Today’s Li-ion cells have relatively poor materials utilization, a major cause of which is the fact that electrodes with thickness above 100 µm do not have adequately fast kinetics due to multiple contributions to the resistance. An alternative electrode architecture is needed to increase active materials and reduce the use of inactive components.

Objectives
In 2015-2016: we focus and expand work on magnetic alignment approach: fabricate thicker, denser cathodes and anodes having at least 10 mAh/cm² area capacity that passes an accepted EV drive cycle test, towards overall project objective of enabling higher energy density and lower-cost EV cells and packs.

Results and Discussion

Magnetic Alignment Approaches (Sintered Electrodes) [2]:

- **Approach 1:** Magnetic rods
  - Suspension
  - Magnetic Alignment
  - Consolidation
  - Sintering

- **Approach 2:** Magnetic-emulsion droplets
  - Electrode particle slurry is mixed with a sacrificial magnetic phase (nano iron oxide)
  - Magnetic phase is aligned in a magnetic field
  - Solvent removal
  - The sacrificial phase is removed by combustion or evaporation

LiCoO² electrode fabrication using magnetic alignment of sacrificial pore former

Electrochemical testing: From standard galvanostatic tests towards EV drive cycles (e.g., Dynamic Stress Test (DST))

- 300 µm thick electrodes
- 200 µm thick electrodes
- Dynamic Stress Test (DST)

Summary
- Magnetic alignment approach can introduce low-tortuosity pores preferentially oriented in the primary transport direction of battery electrodes.
- Usable capacity of sintered LiCoO² electrodes increases by up to a factor of 3 at 2C continuous discharge rates.
- Electrodes with aligned pore channels deliver areal capacities above 8 mAh/cm² under DST tests, more than twice that of the highest areal capacity conventional Li-ion electrodes.

Magnetic Alignment Approaches (w/o sintering, w/ carbon/binder additives) [3]:
- Advantage of Magnetic Emulsion Approach:
  - Nylon-rod derived electrodes
  - EMulsion approach vs Nylon rod: greater flexibility, i.e. through pores, no need to precut nylon rods to match thickness
  - More importantly, it opens up the path to rapid fabrication of low-tortuosity electrodes at room-temperature (without high-temperature sintering)

Electrochemical testing:

- LiCoO² cathodes with aligned pores can deliver areal capacity > 10 mAh/cm² even at C/2 rate (theoretical capacity ~14 mAh/cm²)
- LiCoO² cathodes with aligned pores clearly outperform those without aligned pores (4 vs 4.430-450 μm in thickness, ~92% porosity, ~14 mAh/cm² theoretical capacity)
- Both LiCoO² cathode and MCMB graphite anode can be prepared through the magnetic-emulsion approach (areal capacity > 10 mAh/cm²).

**Summary and Outlook**
- We have developed a new approach to prepare high-capacity low-tortuosity electrodes at room-temperature through magnetic alignment, without the need of high-temperature sintering. This approach allows convenient incorporation of conductive carbon and binders and therefore can be generally used for any cathode or anode materials. This is especially useful for those with high capacity/energy density but limited by electronic conductivity, such as NCA, NMC, and LMO. To address these challenges, it is highly desired to develop a low-cost, scalable, room-temperature process to prepare high-capacity low-tortuosity electrodes.

**References**

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**Fig. 1** High tortuosity composite electrode vs. dense thicker low tortuosity electrode with oriented pores that yields higher area capacity as demonstrated in LiCoO². [1]