Project ES071: Design and Scalable Assembly of High Density Low Tortuosity Electrodes

Linsen Li, Jonathan Sander, Anvesh Gurijala, Randall Erb, and **Yet-Ming Chiang**
Massachusetts Institute of Technology, Cambridge, MA and Northeastern University, Boston, MA.

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 100µm 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. Previously under BATT support our group showed that the introduction of dual-scale porosity (Fig. 1) into sintered $LiCoO_2$ cathodes where electronic conductivity is not limiting permits ~3x higher area capacity (mAh/cm^2) than in conventional electrodes at up to 2C rates. However, high-temperature sintering is energy-intensive and time-consuming. Therefore it is not suitable for large-scale production. In addition, sintering process hinders incorporation of conductive carbon materials into the electrode, which makes it difficult to prepare electrodes from active materials limited by electronic conductivity, such as NCA, NMC, and LNMO. To address these challenges, it is highly desired to develop a low-cost, scalable, room-temperature process to prepare high-capacity low-tortuosity electrodes.

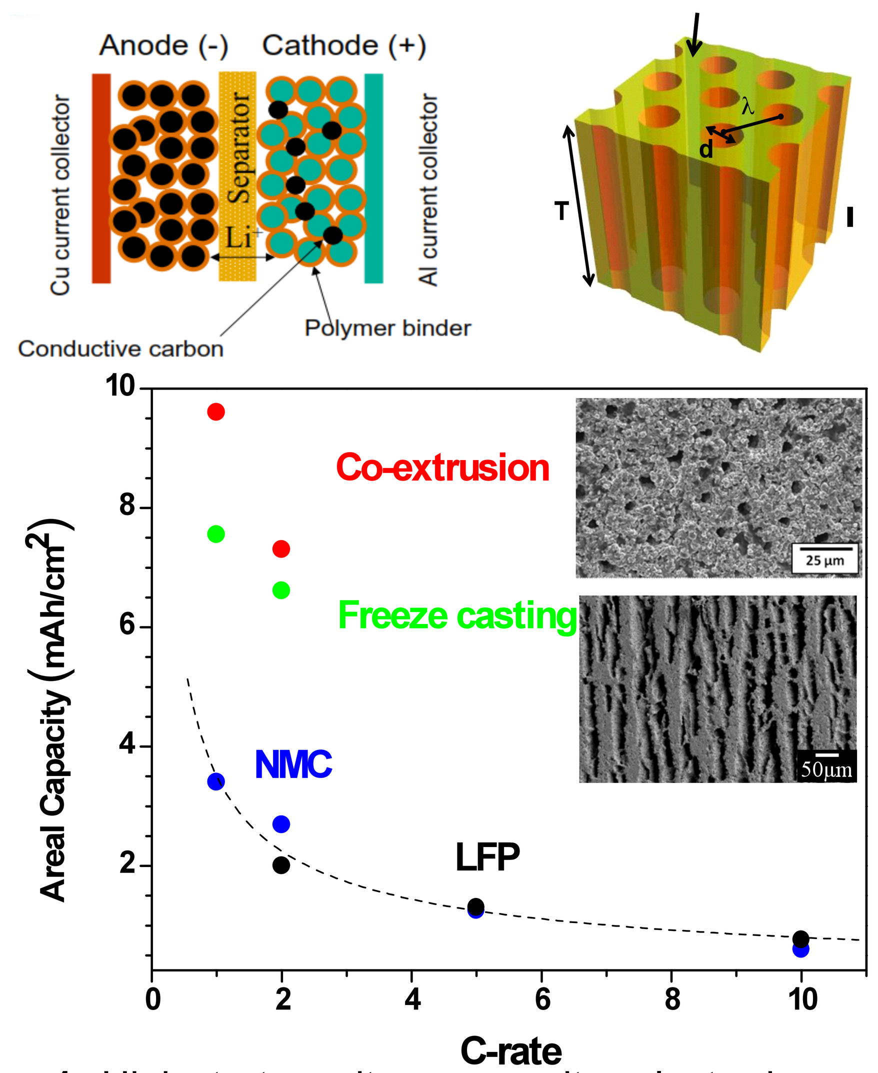


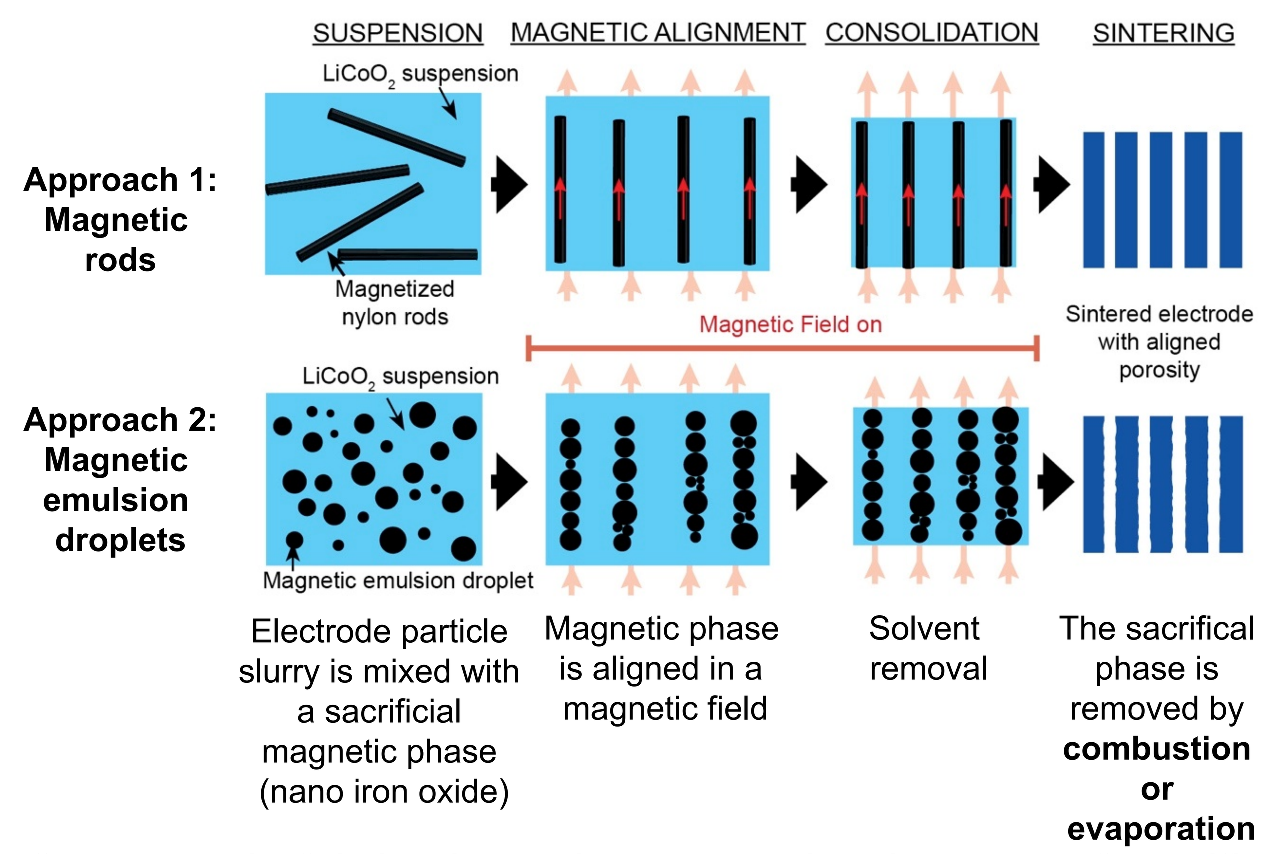
Fig. 1 High tortuosity composite electrode vs. dense thicker low tortuosity electrode with oriented pores that yields higher area capacity as demonstrated in $LiCoO_2$ [1].

Objectives

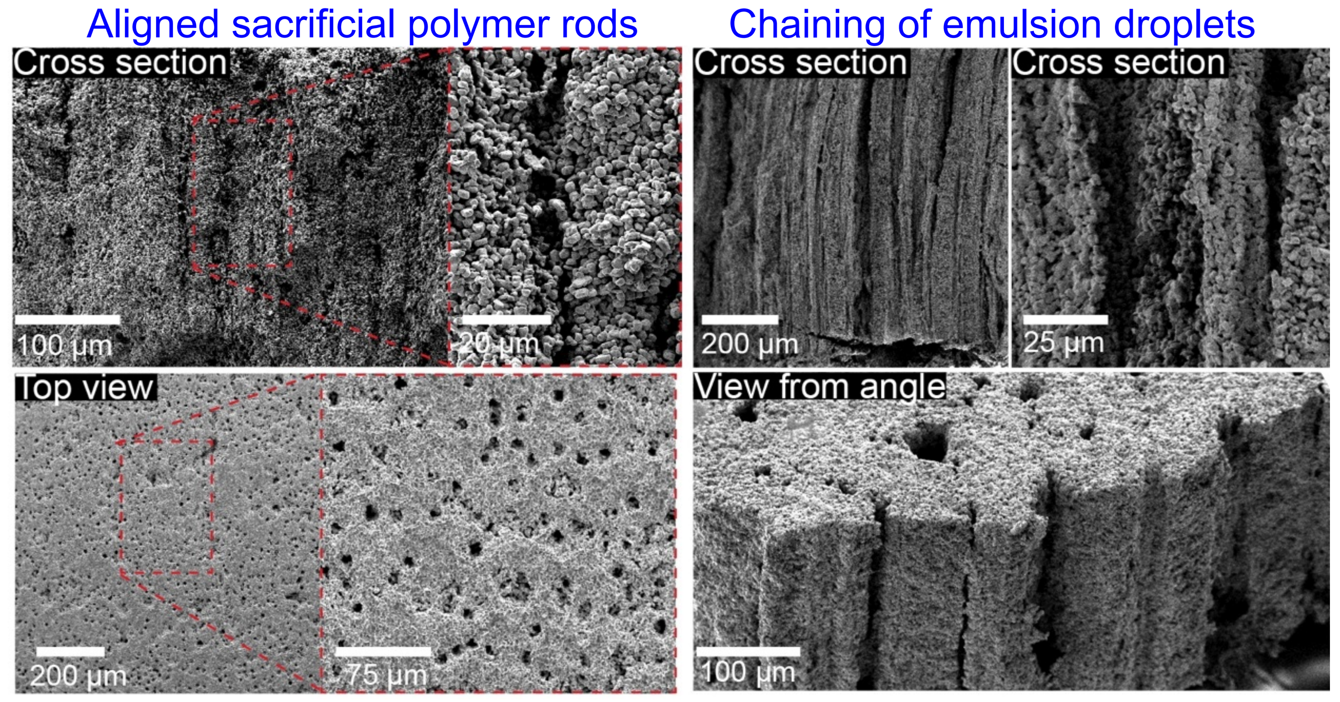
In 2015-2016: we focus and expand work on magnetic alignement approach; fabricate thicker, denser cathodes and anodes having at least 10 mAh/cm^2 area capacity that passes an accepted EV drive cycle test, towards overall project objective of enabling higher energy density and lower-cost EV cells and packs.

Results and Discussion

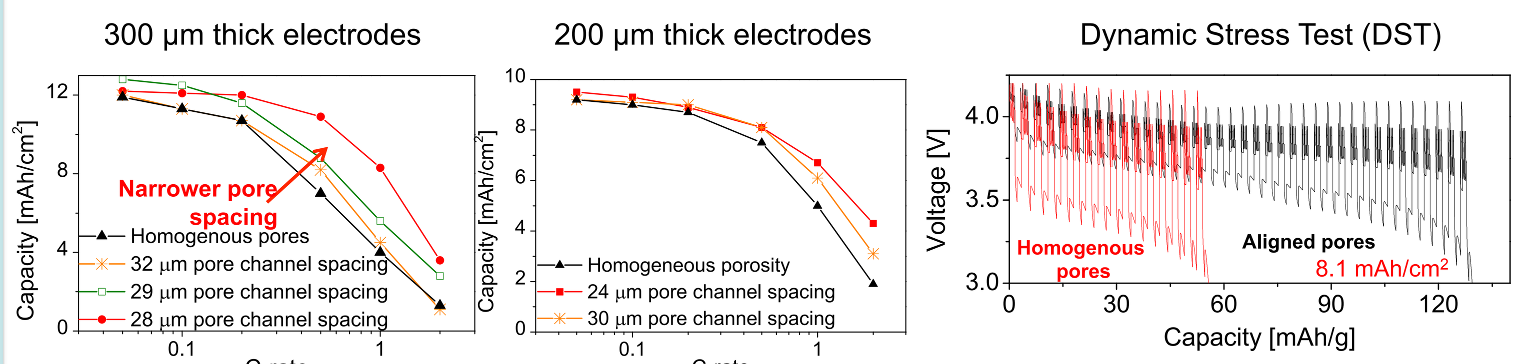
Magnetic Alignment Approaches (**Sintered Electrodes**) [2]:



$LiCoO_2$ 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))



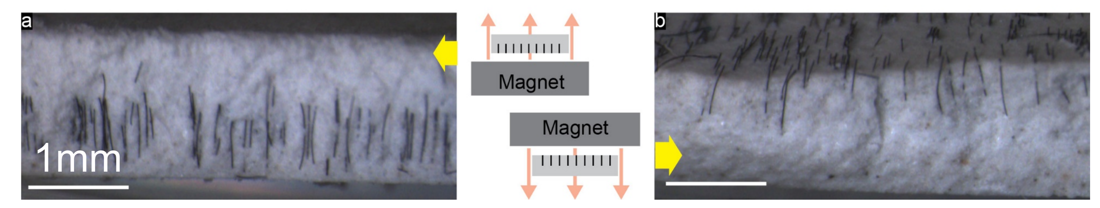
Summary

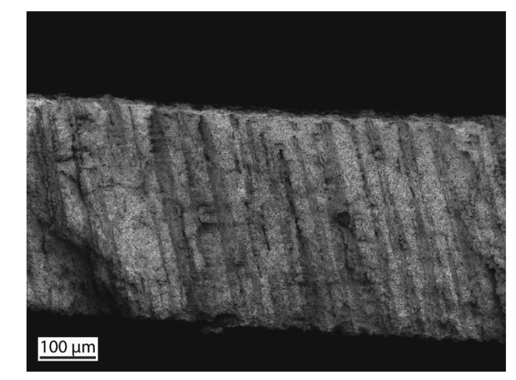
- Magnetic alignement approach can introduce low-tortuosity pores preferentially oriented in the primary transport direction of battery electrodes
- Usable capacity of sintered $LiCoO_2$ electrodes increases by up to a factor of 3 at 2C continuous discharge rates.
- Electrodes with aligned pore channels deliver areal capacities above 8 mAh/cm^2 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 Magentic Emulsion Approach:

Nylon-rod derived electrodes VS **Magnetic-emulsion based electrodes**

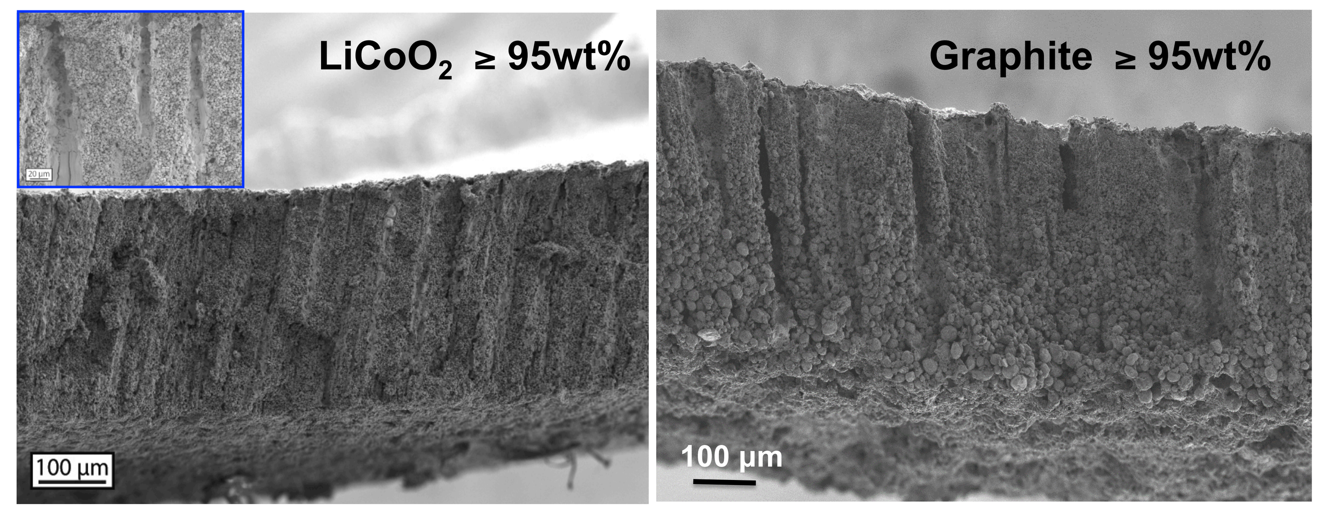




- Emulsion approach vs Nylon rod: greater flexibility, i.e. through pores, no need to precut nylon rods to match thickness
- **More importantly, it opens up the path to rapid fabrication of low-tortuosity electrodes at room-temperature (without high-temperatre sintering)**

Magnetic-emulsion based electrodes (without Sintering)

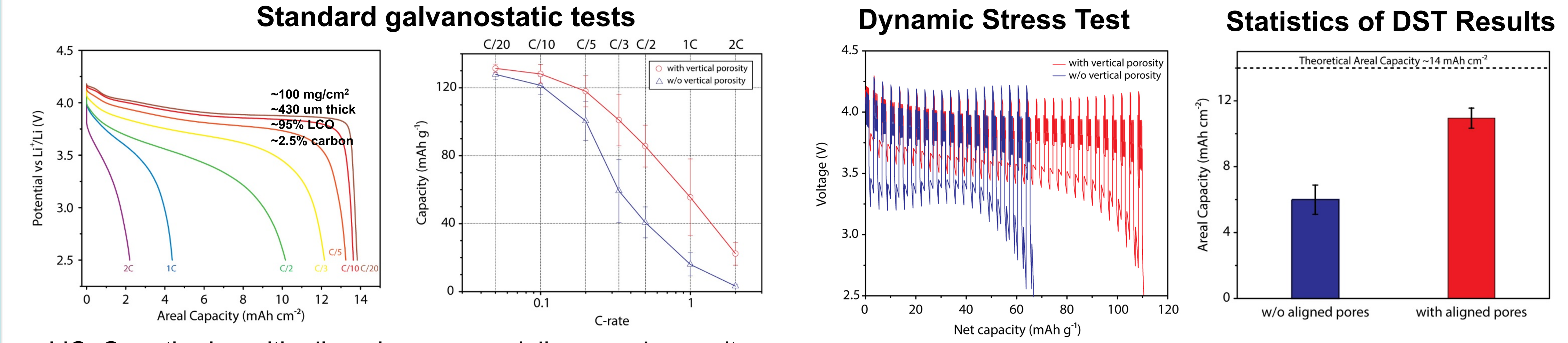
$LiCoO_2$/carbon black/binder Cathode | **MCMB Graphite/binder Anode**



- Both $LiCoO_2$ cathode and MCMB graphite anode can be prepared through the magnetic-emulsion approach (areal capacity > 10 mAh/cm^2). The key is that there must be a good surfactant to stabilize the oil-in-water (ferrofluid-in-water based slurry) emulsion

Electrochemical testing:

Standard galvanostatic tests | **Dynamic Stress Test** | **Statistics of DST Results**



- $LiCoO_2$ cathodes with aligned pores can deliver areal capacity > 10 mAh/cm^2 even at C/2 rate (theoretical capacity ~14 mAh/cm^2)
- $LiCoO_2$ cathodes with aligned pores clearly outperform those without aligned pores (4 vs 4, ~430-450 µm in thickness, ~42% porosity, ~14 mAh/cm^2 theoretical capacity)
- $LiCoO_2$ cathodes with aligned pores can deliver areal capacity 11.2 mAh/cm^2, 110 mAh/g vs $LiCoO_2$ cathodes w/o aligned pores 6.5 mAh/cm^2, 70 mAh/g under DST test (Maximum discharge rate 2C, maximum charge rate 1C, peak power > 700 W/kg)

Summary and Outlook

- We have developed a new approach to prepare high-capacity low tortuosity electrdoes 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 expecially useful for those with high capacity/energy density but limited by electronic conductivity, such as NCA, NMC, and high-voltage LNMO.
- LCO electrodes with aligned porosity outperform those without aligned porosity under both standard galvanostatic and DST tests. MCMB graphite anodes and full cells (LCO-graphite) will be tested.

References

[1] C.K. Erdonmez, C.-J. Bae, J.W. Halloran & Y.-M. Chiang, Adv. Mater., **25** (2013) 1254.
[2] J.S. Sander, R.M. Erb, L. Li, A. Gurijala, Y.-M. Chiang, Nature Energy 2016, in revision
[3] L. Li, R.M. Erb, Y.-M. Chiang, 2016, in preparation

Acknowledgements

This work was supported by the Assistant Secretary for Energy Efficiency and Renewable Energy, Office of Vehicle Technologies of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231, Subcontract No. 6920999 under the Batteries for Advanced Transportation Technologies (BATT) Program.