Geometric Characteristics of Lithium Ion Battery Electrodes with Different Packing Densities.

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The microstructure of electrodes plays a critical role in determining the performance of lithium ion batteries (LIBs), because the microstructure can affect the transport and electrochemical processes within electrodes (1-3). Increasing the volume fraction of active materials in the electrode will increase the energy density. However, the electrodes' structural properties could also be changed significantly and the critical physical and electrochemical processes in LIBs will be affected. Therefore, the performance of a LIB can be optimized for a specific designing condition by operating electrode microstructures. For instance, Hellweg suggested a spatially varying porous electrode model to improve lithium ion transport in electrolyte phase at high charge/discharge rates (4). He showed that the power density of the graded porosity electrode was higher than a homogeneous porosity electrode without energy loss. In this study, we investigate the realistic geometric characteristics of electrode microstructures under different packing densities and the effect of packing density on the performance of LIBs. Moreover, a spatially varying porous electrode will be studied to increase the electrode energy density without losing rate capability.

To investigate geometric characteristics of porous microstructures, cathode electrodes were fabricated from a 94:3:3 (weight %) mixture of LiCoO₂ (average particle radius = 5 μ m), PVDF, and super-P carbon black. To change the packing density, initial thickness of the electrodes was set in a range of 40 \sim 80 μ m. Then all electrodes were pressed down to 40 µm by using a rolling press machine. A synchrotron X-ray nano-computed tomography instrument (nano-CT) at the Advanced Phothon Source of Argonne National Lab was employed to obtain morphological data of the electrodes, with a spatial resolution of 60 nm. The morphology data sets were quantitatively analyzed to characterize their geometric properties. Fig. 1 shows the porosity (ε) , specific surface area (A_s , μ m-1), tortuosity (τ), and pore size distribution of 4 different electrode microstructures. The pore size distribution of the un-pressed electrode ($\varepsilon =$ 0.56, black color) demonstrates nonuniformly dispersed active material. The highest packing density electrode ($\varepsilon =$ 0.36, red color) shows the highest tortuosity. The charge/discharge experiments were also conducted for these 4 different electrodes. The geometric properties and cell testing results will be analyzed and reported.

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Fig. 1 Geometric characteristics (porosity ε , specific surface area A_s , tortuosity τ , pore size distribution) of x-ray generated porous electrode microstructure with different packing densities.

References:

1. R. E. Garcia and Y. M. Chiang, *Journal of the Electrochemical Society*, **154**, A856 (2007).

2. I. V. Thorat, D. E. Stephenson, N. A. Zacharias, K. Zaghib, J. N. Harb and D. R. Wheeler, *J. Power Sources*, **188**, 592 (2009).

 R. Thiedmann, O. Stenzel, A. Spettl, P. R. Shearing, S. J. Harris, N. P. Brandon and V. Schmidt, *Computational Materials Science*, **50**, 3365 (2011).
B. Hellweg, Microstructural Modeling of Lithium Battery Electrodes, in *Materials Science and Engineering*, Massachusetts Institute of Technology (2000).