A02-0384 - *(Digital Presentation)* Activating the Ion Transmission at the Cathode-Electrolyte Interface in All-Solid-State Batteries

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Abstract

All-solid-state batteries (ASSBs) have garnered increasing attention due to the enhanced safety, featuring nonflammable solid electrolytes as well as the potential to achieve high energy density. [1] The advancement of the ASSBs is expected to provide, arguably, the most straightforward path towards practical, high-energy, and rechargeable batteries based on metallic anodes.[1]

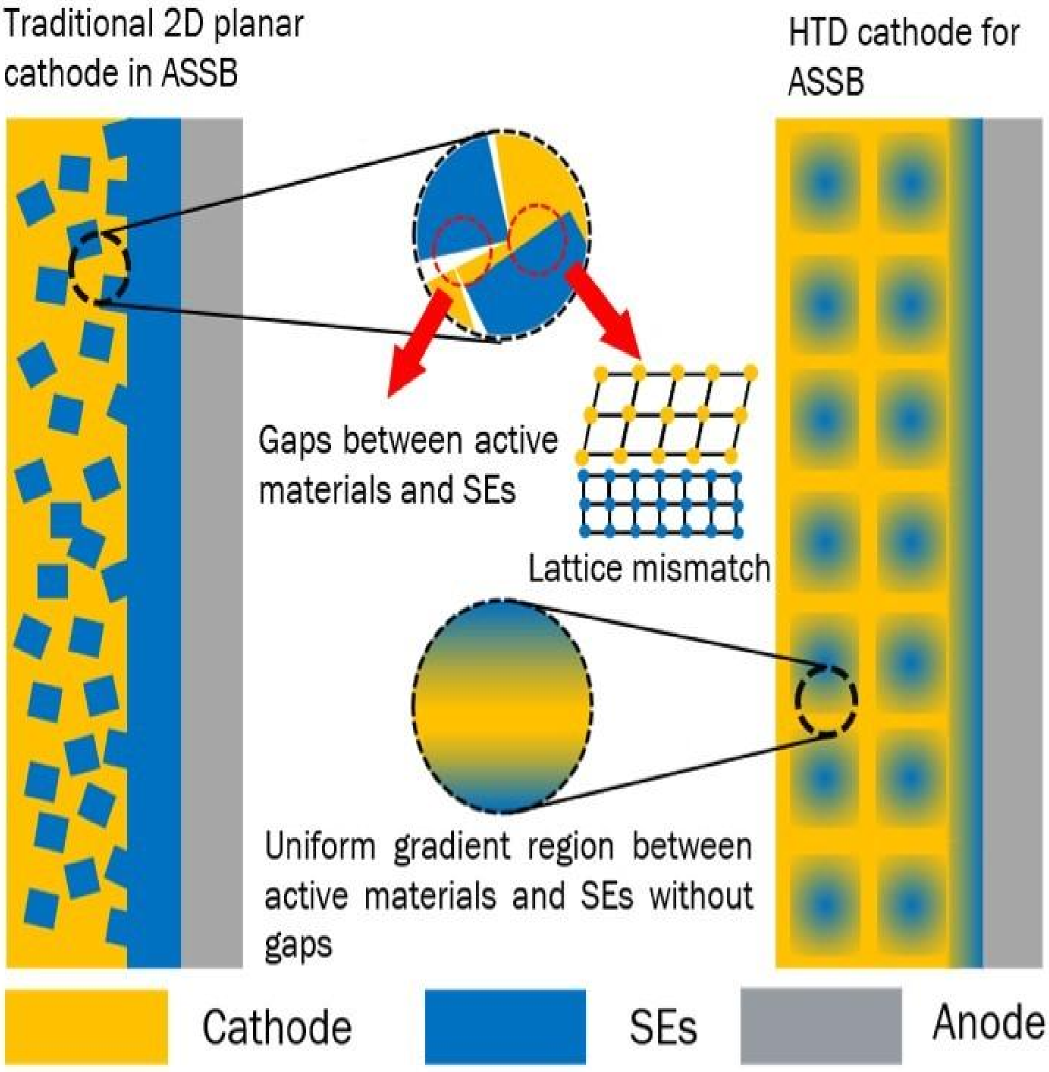
However, the sluggish ion transmission at the cathode-electrolyte (solid/solid) interface would result in the high resistant at the contact and limit the practical implementation of these all solid-state materials in real world batteries.[2] Several methods were suggested to enhance the kinetic condition of the ion migration between the cathode and the solid electrolyte (SE).[3] A composite strategy that mixes active materials and SEs for the cathode is a general way to decrease the ion transmission barrier at the cathode-electrolyte interface.[3] The active material concentration in the cathode is reduced as much as the SE portion increases by which the energy density of the ASSB is restricted. In addition, the mixing approach generally accompanies lattice mismatches between the cathode active materials and the SE, thus providing only limited improvements, which is imputed by random contacts between the cathode active materials and the SE during the mixing process. Implementing high-pressure for the electrode and electrolyte of ASSB in the assembling process has been verified is a but effective way to boost the ion transmission ability between the cathode active materials and the SE by decreasing the grain boundary impedance. Whereas the short-circuit of the battery would be induced by the mechanical deformation of the electrolyte under high pressure. [4]

Herein, we demonstrate a novel way to address the ion transmission problem at the cathode-electrolyte interface in ASSBs. Starting from the cathode configuration, the finite element method (FEM) was employed to evaluate the current concentration and the distribution of the space charge layer at the cathode-electrolyte interface. Hierarchical three-dimensional (HTD) structures are found to have a higher Li^+ transfer number (t_{Li+}), fewer free anions, and the weaker space-charge layer at the cathode-electrolyte interface in the resulting FEM simulation. To take advantage of the HTD structure, stereolithography is adopted as a manufacturing technique and single-crystalline Ni-rich (SCN) materials are selected as the active materials. Next, the manufactured HTD cathode is sintered at 600 °C in an N_2 atmosphere for the carbonization of the resin, which induces sufficient electronic conductivity for the cathode. Then, the gel-like $Li_{1.4}Al_{0.4}Ti_{1.6}(PO_4)_3$ (LATP) precursor is synthesized and filled into the voids of the HTD structure cathode sufficiently.

And the filled HTD structure cathodes are sintered at 900 °C to achieve the crystallization of the LATP gel. Scanning transmission electron microscopy (STEM) is used to unveil the morphology of the cathode-electrolyte interface between the sintered HTD cathode and the in-situ generated electrolyte (LATP). A transient phase has been found generated at the interface and matched with both lattices of the SCN and the SE, accelerating the transmission of the Li-ions, which is further verified by density functional theory calculations. In addition, Electron Energy Loss Spectroscopy demonstrates the preserved interface between HTD cathode and SEs. Atomic force microscopy is employed to measure the potential image of the cross-sectional interface by the peak force tapping mode. The average potential of modified samples is lower than the sample that mix SCN and SEs simply in the 2D planar structure, which confirms a weakened space charge layer by the enhanced contact capability as well as the ion transmission ability. To see if the demonstrated method is universally applicable, $LiNi_{0.8}Co_{0.1}Mn_{0.1}O_2$ (NCM811) is selected as the cathode active material and manufactured in the same way as the SCN. The HTD cathode based on NCM811 exhibits higher electrochemical performance compared with the reference sample based on the 2D planar mixing-type cathode. We believe such a demonstrated universal strategy provides a new guideline to engineer the cathode/electrolyte interface by revolutionizing electrode structures that can be applicable to all-solid-state batteries.

Figure 1. Schematic of comparing of traditional 2D planar cathode and HTD cathode in ASSB

1. Tikekar, M. D., *et al.*, *Nature Energy* (2016) **1** (9), 16114
2. Banerjee, A., *et al.*, *Chem Rev* (2020) **120** (14), 6878
3. Chen, R., *et al.*, *Chem Rev* (2020) **120** (14), 6820
4. Cheng, X., *et al.*, *Advanced Energy Materials* (2018) **8** (7)



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