Charge transport kinetics at a cathode-solid electrolyte interface studied by μ -cavity electrode

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All-solid-state batteries (ASSBs) are currently accepted as promising candidates for future energy storage devices with conventional layered cathode materials to realize higher energy densities and safe performance compared to flammable liquid electrolyte lithium-ion batteries. However, the interface between the cathode / solid electrolyte still remains a crucial issue that hinders the practical application of solid-state lithium batteries [1]. In typical solid-state batteries, the porous nature of the composite electrode and its porosity, electrode thickness, and uneven distribution of active materials complicate the estimating of the solid-solid interfacial kinetics. Hence, characterizing the interfacial properties of single cathode particles without affecting the above-mentioned parameters is required for fundamental understanding. Previously applied approaches (e.g., single particle measurement) for investigating the interfacial behavior of single active material particles with microelectrode are well adapted for characterization in liquid electrolyte systems and cannot evaluate active particles in combination with solid electrolytes in ASSBs [2]. Thus, the development of novel and sophisticated approaches is required.

The current study investigates the DOD-dependent interfacial charge transfer kinetics between cathode particles and solid electrolytes using a microelectrode with a cylindrical cavity trap. The NCM cathode particles from the prepared dry electrodes are immobilized in the cavity traps by compression on a glass substrate. Further, the as-prepared μ -cavity electrode was transferred to the solid electrolyte system based on the argyrodite-type (Li₆PS₅Cl) solid electrolyte. Several cycles were conducted for confirmation of the stability of electrochemical reactions. In the subsequent cycle, positive and negative pulsed currents were applied after each DOD state, and potential peak responses were measured. The obtained potential peak responses and applied pulsed current amplitudes were used to draw a Tafel plot and estimate the DOD-dependent charge transfer resistance between the cathode / solid electrolyte interface. In general, the charge transfer resistance is often specified in the literature to be equal to the interfacial impedance.

As a result, the study presents an alternative tool to investigate the interfacial charge transfer kinetics between cathode / solid electrolytes in the microscale. The illustration designed μ cavity electrode in the solid electrolyte system is given in Figure 1. A μ -cavity electrode is a versatile tool and not limited to secondary NCM particles, it is highly adopted for measuring the charge transfer properties of any particles in the microscale by providing the pressure that requires the solid electrolytes.



Figure 1. The schematic illustration of μ -cavity electrode in the solid electrolyte system.

The microcavity electrode is an alternative approach for single particle measurement and obtained results can exhibit meaningful clues of charge transfer resistance by Tafel plot and diffusion coefficient calculation for the fundamental understanding of interfacial behavior.

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References

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