Study of the Mechanical Behaviour of Thin Film Solid Electrolytes Using Atomic Force Microscopy

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As the application of Li-ion batteries (LIBs) to power consumer electronics and electric vehicles increases, the safety, size, energy density and power density of the batteries become major concerns. Unlike conventional liquid electrolytes in LIBs, solid electrolytes (SE) are non-flammable and stable [1]. Miniaturized all-solid-state LIBs embedded in a 3D architecture have shown improvements in energy and power density by increasing the surface capacity [2]. However, the formation of microcracks and eventually mechanical breakdown due to the internal and extremal forces at the interface of SE/electrodes leads to reduced ionic conductivity and battery capacity. Furthermore, microcracks at a high current density can provide a pathway for Li dendrite growth and cause shortcuts within the cell [3].Therefore, understanding mechanical behaviour of SEs are necessary for improving battery performance [4]. To date detailed knowledge of the mechanisms behind the mechanical breakdown is limited. In this research, we studied the mechanical behaviour of two promising amorphous thin film SEs, Lithium Tantalate (LTO) and lithium phosphate (LPO) (Li conductivity of $2\text{x}10^{-8}$ mS.cm^{-1} and LPO is $3.3\text{x}10^{-8}$ mS.cm^{-1} at 26 °C, respectively) [5, 6].

LTO and LPO thin films were prepared using atomic layer deposition (ALD) (at 225 °C and 250 °C, respectively) which can fabricate uniform, crack and pinhole free ultra-thin films for 3D structures [5-7]. Thin film SE were deposited over a holey silicon nitride TEM grids (hole diameter of 2.5 µm) with a single layer of CVD graphene on top used to support the deposited LTO and LPO. The mechanical behavior of ALD prepared SEs films studied using an Atomic Force Microscopy (AFM) based indentation technique (Figure 1. a). A range of thicknesses of amorphous thin film SEs (e.g 5 nm, 15 nm, and 25 nm) were studied in order to investigate the influence of the thickness on the mechanical properties of the SE films. Studies of the elastic behavior reveal that by increasing the thickness of the film the mechanical effect of graphene supporting layer vanishes and intrinsic behaviour of the films dominates the mechanical responses to mechanical loadings [8]. However, LTO shows higher stiffness as compared to LPO films at all thicknesses. Moreover, fracture behavior of SE films was studied by indenting the films until failure (Figure 1 b and c). Both LTO and LPO SEs show brittle failure. LTO films were found to fail at higher forces (3.031 ±0.05 µN) as compared to LPO film (1.97 ±0.07 µN) with a similar thickness. These results suggest that the crack formation is delayed in LTO compare to LPO film and LTO has greater potential to suppress the crack formation at the SE/electrolyte interface that can improve the overall performance of the battery.

References

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Figures

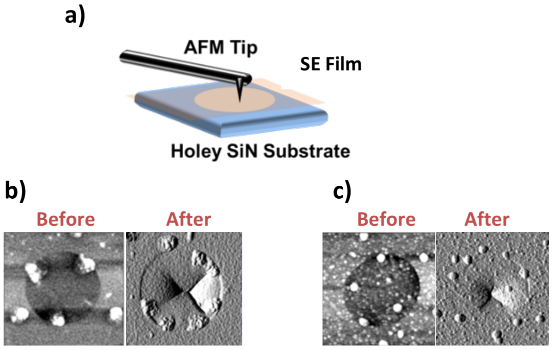

Figure 1. a) Schematic of AFM indentation experiment conducted on SE films, b and c) AFM image of 25 nm thick LTO and LPO films before and after AFM indentation to failure experiment, respectively.