Glass-Ceramic Solid Electrolytes for All-Solid-State Rechargeable Batteries

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All-solid-state rechargeable lithium or sodium batteries attract much attention because of their high safety, long cycle life, versatile geometry and high energy density [1]. Development of superior inorganic solid electrolytes is important for realize those rechargeable batteries. Very recently, LGPS-type crystalline $Li_{9.54}Si_{1.74}P_{1.44}S_{11.7}Cl_{0.3}$ is reported to show a quite high lithium-ion conductivity of 2.5 x 10^{-2} S cm^{-1} at 25^{o}C [2] and this sheds light on the possibility of high-rate capability in all-solid-state batteries.

Mechanical properties are also of great significance to solid electrolytes for forming favorable solid-solid interfaces in all-solid-state batteries. We have prepared sulfide and oxide glass-based electrolytes with both high conductivity and good deformability. Glass electrolytes are useful as a precursor for precipitating metastable crystalline phases, which are difficult to prepare by a conventional solid phase reaction. Several metastable phases such as $Li_7P_3S_{11}$ and cubic-Na_3PS_4 are crystallized from glassy state and the prepared glass-ceramic electrolytes exhibit higher conductivities than their mother glasses [3,4]. These sulfide glass-based electrolytes also have favorable ductility for forming good solid-solid interfaces with electrode active materials [5]. Relative density of the Na_3PS_4 glass pellet prepared just by cold-pressing at 360 MPa reaches 94%, which is higher than 90% of the Li_3PS_4 glass. The oxide glass electrolytes in the system Li_3BO_3-Li_2SO_4-Li_2CO_3 also have a similar relative density to those sulfide systems [6] and an all-solid-state cell with the oxide electrolytes operate as a lithium secondary battery at 100^{o}C.

Amorphous materials are also useful as electrode active materials. For instance, amorphous transition metal sulfides such as TiS_3 functioned as an active material with high capacity. Because amorphous TiS_3 (*a*-TiS_3) has high ductility and electrical conductivity, an all-solid-state lithium cell using the *a*-TiS_3 active material in a positive electrode layer without any conductive additives (solid electrolytes and carbons) exhibits good charge-discharge cyclability [7]. Amorphous oxide electrodes in the system $LiCoO_2$-Li_2SO_4 prepared via mechanochemistry also have good deformability and act as a mixed conductor [8]. Amorphous electrolyte and electrode materials with high conductivity and good ductility are useful for forming favorable solid-solid interfaces, leading to improving electrochemical performance of all-solid-state rechargeable batteries.

References:

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