

演題: Organic/Inorganic Solid Electrolytes for Solid-State Lithium Batteries

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要 旨:

Recently, all solid-state lithium batteries (ASSLBs) have been paid worldwide attention due to their safety and high energy density features. In order to enable ASSLBs to operate at low temperature and high rate and exhibit high capacity, the solid electrolyte should have high ionic conductivity, electrochemical stability, mechanical properties and good interfacial contact with electrode materials. However, none of the solid electrolyte materials currently studied can meet the above requirements at the meantime: The advantages of inorganic solid electrolytes usually lie in ionic conductivity, electrochemical stability, and mechanical properties rather than interfacial contact, while organic solid electrolytes are the opposite. In this work, we have prepared PEO/LLZO solid electrolyte with complementary advantages and applied it to ASSLBs. Firstly, molecular dynamic simulations were performed to study the phase transition of LLZO from low ionic conductive tetragonal phase to highly ionic conductive cubic phase. Results showed that, by introducing a few lithium vacancies into LLZO lattice, the cubic phase can be efficiently stabilized at room temperature. Secondly, field assisted sintering technology and conventional solid-state method were used to prepare LLZO. By regulating lithium concentration and framework structure, Ta-doped LLZO with ionic conductivity as high as 1.1×10^{-3} Scm⁻¹ was obtained. Thirdly, the crystallization behavior, micromorphology, ion mobility, electrochemical stability and contact stability to lithium metal of PEO-LLZO solid electrolyte with different LLZO content were studied. With the change of LLZO content, the ionic conductivity of PEO-LLZO composite solid electrolyte has two peaks at 7.5% and 60%. Finally, two composite solid electrolytes were applied to ASSLBs (LiFePO₄/Li). All the ASSLBs can be safely operated over a wide range of current density (0.2~2C) and temperature (30~60°C) without incurring damage. ASSLBs with PEO-7.5%LLZO-SN solid electrolyte exhibit excellent cycling stability and prolonged life. At 1C and 60°C, the specific capacity of the 500th cycle is 108.8 mAh g^{-1} . Capacity retention rate is >80.0%.

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