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Abstract

$Li_{1+y}Ti_{2-x-y}Ge_xAl_y(PO_4)_3$ NASICON-Type Electrolytes with Enhanced Conductivity for Solid-State Lithium-Ion Batteries †

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Abstract: The use of lithium-ion batteries allows for a reliable and efficient storage of electricity. Commercial batteries use flammable liquid organic electrolytes, which have a low thermal and electrochemical stability. Replacing liquid electrolytes with solid ones would solve these problems. NASICON-structured electrolytes, in particular LATP ($Li_{1+y}Ti_{2-y}Al_y(PO_4)_3$) and LAGP ($Li_{1+y}Ge_{2-3y}Al_y(PO_4)_3$), are among the most promising electrolytes for all-solid-state batteries. The partial replacement of titanium ions with germanium ions can lead to materials that combine the high lithium-ion conductivity of LATP with the high chemical stability of LAGP. The aim of this work was to synthesize and study the ionic mobility of $Li_{1+y}Ti_{2-x-y}Ge_xAl_y(PO_4)_3$ (x = 0–2, y = 0–0.3) with the NASICON structure. $Li_{1+y}Ti_{2-x-y}Ge_xAl_y(PO_4)_3$ (x = 0–2, y = 0–0.3) electrolytes were synthesized with the solid-state method and investigated using X-ray diffraction and scanning electron microscopy, impedance spectroscopy, and NMR spectroscopy. The processes occurring during the solid-state synthesis of $Li_{1+y}Ti_{2-x-y}Ge_xAl_y(PO_4)_3$ were studied. An increase in conductivity from 10^{-7} S/cm to $4.6 \cdot 10^{-6}$ S/cm at 25 °C was found when 10% of titanium ions were replaced with germanium. The additional introduction of aluminum resulted in an increase in lithium conductivity of up to $1.4 \cdot 10^{-4}$ S/cm (25 °C). Since grain boundaries were of decisive importance for the overall ionic conductivity of the NASICON-structured phosphates, the influence of the precursor mechanical treatment on the microstructure and ionic conductivity of the prepared materials was studied. The use of the mechanical treatment led to a significant increase in grain size (reducing the grain boundaries and their resistance) and an increase in ionic conductivity (up to $6.4 \cdot 10^{-4}$ S/cm at 25 °C). The obtained materials could be considered promising solid electrolytes for all-solid-state lithium batteries with high safety and stability.

Keywords: ion conductivity; solid electrolyte; lithium-ion battery; NASICON

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