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Abstract

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

Ekaterina Kurzina * and Irina Stenina

Institute of general and inorganic chemistry of Russian Academy of Science; irina_stenina@mail.ru
* Correspondence: katya.kurzina@gmail.com

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Abstract: The use of lithium-ion batteries allows to reliable and efficient storage of electricity.Commercial batteries use flammable liquid organic electrolytes, which have low thermal andelectrochemical stability. Replacing liquid electrolytes with solid ones will 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. Partialreplacement of titanium ions by germanium ions can lead to materials that combine the highlithium-ion conductivity of LATP with the high chemical stability of LAGP. The aim of thiswork 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 by 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-statesynthesis of $Li_{1+y}Ti_{2-x-y}Ge_xAl_y(PO_4)_3$ have been studied. An increase in conductivity from 10-7 S/cm to 4.6.10-6 S/cm at 25 °C was found when 10% titanium ions were replaced bygermanium. Additional introduction of aluminum results in increase in lithium conductivity up to 1.4.10-4 S/cm (25 °C). Since grain boundaries are of decisive importance for the overall ionicconductivity of the NASICON-structured phosphates, the influence of precursor mechanicaltreatment on the microstructure and ionic conductivity of the prepared materials was studied.The use of mechanical treatment leads to a significant increase in grain size (reducing the grainboundaries and its resistance) and an increase in ionic conductivity (up to 6.4.10-4 S/cm at 25 °C).The obtained materials can be considered as promising solid electrolytes for all-solid-statelithium batteries with high safety and stability.

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

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