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Divalent Ni²⁺ cation conduction in NASICON-type solid

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Abstract

Divalent Ni²⁺ cation-conducting solid electrolytes, $(Ni_xHf_{1-x})_{4/(4-2x)}Nb(PO_4)_3$, were successfully developed by introducing Ni²⁺ cations into a HfNb(PO_4)_3 solid exhibiting a three-dimensional NASICON-type structure. Although the HfNb(PO_4)_3 solid has been reported to be purely a Hf⁴⁺ ion conductor, the conducting species was changed to the divalent Ni²⁺ cation upon Ni²⁺ doping, due to the facile migration of lower-valence ions in the rigid crystal lattice. In addition, by selecting the well-ordered NASICON-type structure as the mother crystal, the $(Ni_xHf_{1-x})_{4/(4-2x)}Nb(PO_4)_3$ solid electrolytes exhibited lower activation energies and higher Ni²⁺ cation conductivities than a previously reported NiZr₄(PO₄)₆ solid with the β -Fe₂(SO₄)₃-type structure. Among the $(Ni_xHf_{1-x})_{4/(4-2x)}Nb(PO_4)_3$ solids, $(Ni_{0.06}Hf_{0.94})_{4/3.88}Nb(PO_4)_3$ (x = 0.06) showed the highest Ni²⁺ ion conductivity, of 2.27×10⁻⁴ S·cm⁻¹ at 600 °C, which is ca. 23 times higher than that of the NiZr₄(PO₄)₆ solid.

Keywords: Solid electrolyte, Divalent, NASICON-type structure, Electroceramics, Functional

1. Introduction

Solid electrolytes, in which ions migrate macroscopically in solids as charge carriers, are promising candidate materials for components of electrochemical devices such as batteries, fuel cells, chemical sensors, and others, due to their favorable thermal and mechanical properties which realize high performance and long-term stability. Specifically, solid electrolytes whose conducting species are Li^+ , H^+ , or O^{2^-} ions are extensively studied for their potential applications in electrochemical energy storage devices [1-4].

In the field of solid electrolytes, it is generally accepted that ion migration is strongly influenced by the valence state of the conducting ion species, where conducting ion species with higher valence number exhibit stronger electrostatic interactions with surrounding anions; thus, the migration of high-valence ions is regarded to be poor in solids [5]. Based on this theory, many researchers have focused on monovalent ions such as Li^+ , H^+ , and Na^+ , which interact with the surrounding counter anions in weaker electrostatic interactions.

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