



Mechanical behavior of Li-ion-conducting crystalline oxide-based solid electrolytes: a brief review

Jeff Wolfenstine¹ · Jan L. Allen¹ · Jeff Sakamoto² · Donald J. Siegel² · Heeman Choe³

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Abstract

Li-ion-conducting solid electrolytes are receiving considerable attention for use in advanced batteries. These electrolytes would enable use of a Li metal anode, allowing for batteries with higher energy densities and enhanced safety compared to current Li-ion systems. One important aspect of these electrolytes that has been overlooked is their mechanical properties. Mechanical properties will play a large role in the processing, assembly, and operation of battery cells. Hence, this paper reviews the elastic, plastic, and fracture properties of crystalline oxide-based Li-ion solid electrolytes for three different crystal structures: $\text{Li}_{0.19}\text{Al}_{0.27}\text{La}_2\text{Zr}_2\text{O}_{12}$ (garnet) [LLZO], $\text{Li}_{0.13}\text{La}_{0.87}\text{TiO}_3$ (perovskite) [LLTO], and $\text{Li}_{1.3}\text{Al}_{0.3}\text{Ti}_{1.7}(\text{PO}_4)_3$ (NaSICON) [LATP]. The experimental Young's modulus value for (1) LLTO is ~200 GPa, (2) LLZO is ~150 GPa, and (3) for LATP ~115 GPa. The experimental values are in good agreement with density functional theory predictions. The fracture toughness value for all three of LLTO, LLZO, and LATP is approximately $1 \text{ MPa m}^{-1/2}$. This low value is expected since, they all exhibit at least some degree of covalent bonding, which limits dislocation mobility leading to brittle behavior.

Keywords Solid electrolyte · Li-conductor · Young's modulus · Hardness · Fracture toughness

Introduction

Many new battery chemistries based on the use of a metallic Li anode are under consideration in order to increase the energy density over the presently used Li-ion batteries. These include aqueous Li-air and all solid-state batteries. These batteries require a solid Li-ion-conducting electrolyte. The major requirements for the solid Li-ion-conducting electrolyte are high Li-ion conductivity, low electronic conductivity, and chemical/electrochemical stability with the Li anode/cathodes [1–5]. In many applications, the solid electrolyte may also be required to exhibit adequate mechanical properties. These include the elastic, plastic, and fracture properties of the solid electrolyte. A scenario where elastic properties are important was described by Monroe and Newman [5]. Using

the linear elasticity theory, it was shown that to prevent the initiation of dendrites during charging at the Li electrode/solid electrolyte interface, the shear modulus of the Li-ion-conducting solid electrolyte must be greater than twice the shear modulus of Li. Regarding fracture properties, it has been recently suggested that the critical current (maximum current above which Li dendrite formation occurs) during charging is dependent on the fracture stress (higher fracture stress, higher critical current) of the Li-ion-conducting solid electrolyte [6]. At present, information on the mechanical properties of solid Li-ion conductors is limited. Thus, it is the purpose of this paper to present a short summary of the recent work on the elastic, plastic, and fracture properties of crystalline oxide-based Li-ion solid electrolytes for three different crystal structures (garnet, perovskite, and NaSICON).

Discussion

Elastic properties (Young's modulus)

Young's modulus, E , is a measure of the stiffness of a solid material. It is an elastic property which depends on the bonding and crystal structure of the solid. It is independent of microstructure (e.g., grain size).

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