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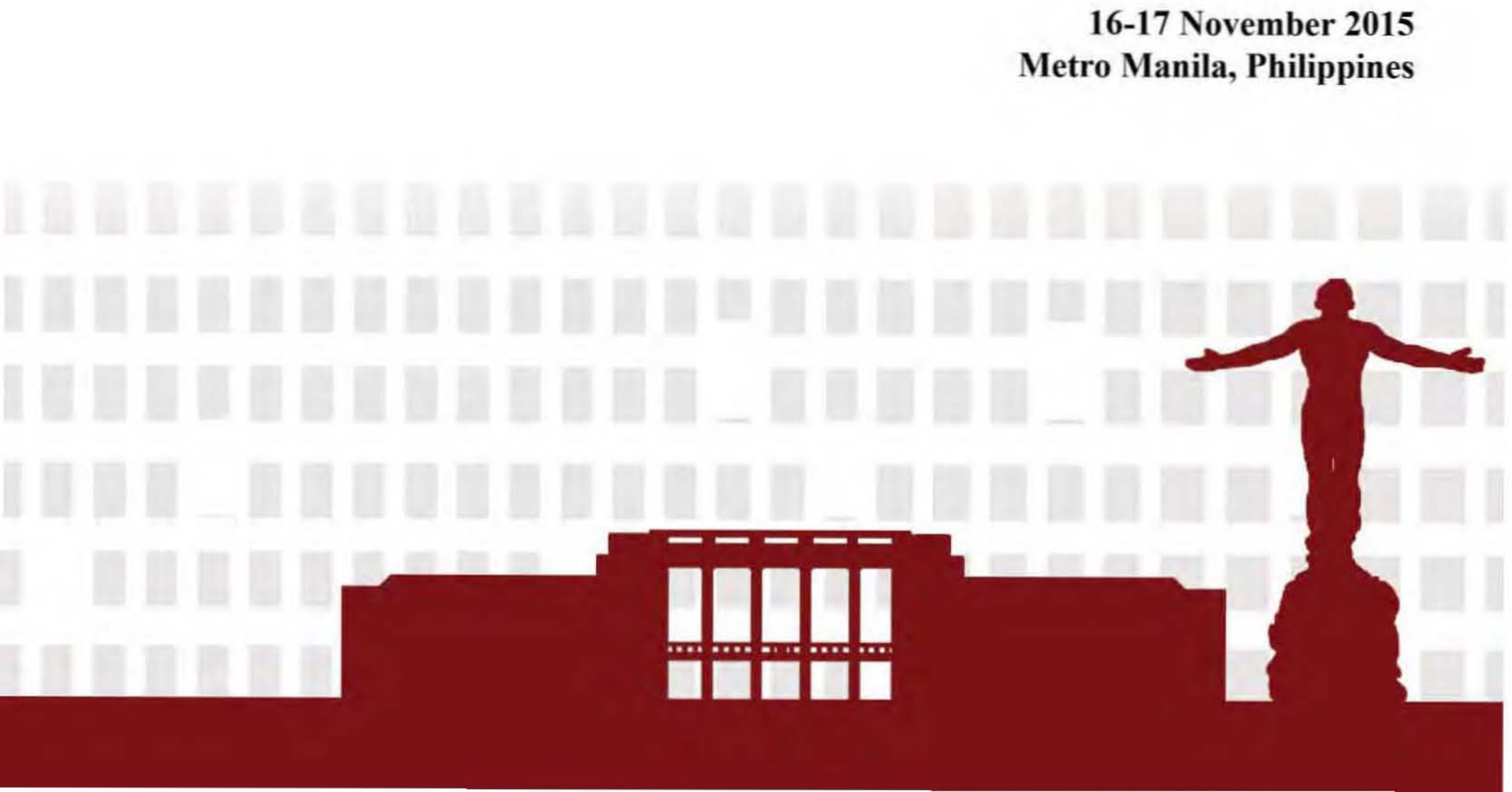

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PREPARATION OF Ga-DOPED $Li_7La_3Zr_2O_{12}$ SOLID ELECTROLYTE FOR LITHIUM-ION BATTERY APPLICATION

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ABSTRACT

Nowadays, most commercially available Li-ion batteries utilize combustible organic solvent-based electrolyte. This hinders its larger-scale applications such as in electric vehicles and electrical power storage systems primarily due to safety concerns. Among other possible alternatives, $Li_7La_3Zr_2O_{12}$ (LLZ) gained increasing attention on account of its good chemical stability with Li anode and high ionic conductivity. Conventional solid-state synthesis of $Li_{7-3x}Ga_xLa_3Zr_2O_{12}$ with mole fractions x= 0.00, 0.10, 0.20 and 0.30 at varying sintering time and temperature were performed in an attempt to improve the microstructure and electrochemical properties. X-ray Diffraction (XRD) results have shown that upon Ga-doping the cubic-phased LLZ was stabilized. Without Ga, a tetragonal-phased LLZ was formed at similar synthesis condition. Scanning Electron Microscopy (SEM) images have shown that Ga acts as a sintering aid. Upon comparison with pure LLZ which was synthesized in similar manner, Ga-doped LLZ exhibits reduced visible pores and better contact among particles. Increasing the amount of Ga dopant and addition of Li source promote densification and grain agglomeration.

Keywords: $Li_7La_3Zr_2O_{12}$, Ga-doped, Solid electrolyte, Lithium-ion battery

Introduction

Nowadays, most commercially available Li-ion batteries utilize combustible organic solvent-based electrolyte. This hinders its larger-scale applications such as in electric vehicles and electrical power storage systems primarily due to safety concerns. In this light, the development of a safer and more stable alternative such as solid-state electrolytes is then explored. Among other materials, $Li_7La_3Zr_2O_{12}$ (LLZ) has gained increasing attention due to its chemical stability with Li anode and relatively high ionic conductivity. However, LLZ only attains high conductivity when it is stabilized at cubic phase [1]. Solid-state syntheses of $Li_{7-3x}Ga_xLa_3Zr_2O_{12}$ of varying Ga molar concentrations were performed in an attempt to systematically improve the microstructure and electrochemical properties of the LLZ material.

Methodology

$Li_{7-3x}Ga_xLa_3Zr_2O_{12}$ with mole fractions of Ga x = 0.00, 0.10, 0.20 and 0.30 are synthesized via solid-state reaction. The precursor materials are ground with ethanol, and then oven-dried at 180 °C. Calcination was done at 900°C for 2 h. After cooling, the synthesized powder is then reground and pressed into pellets. Samples are then sintered at 1000°C for 15 h. The samples were fully characterized using XRD, FT-IR, SEM-EDS, and EIS.

Results and Discussion

XRD results as shown in Fig. 1 a-d reveal that upon Ga-doping, cubic-phased LLZ were stabilized. Without Ga dopant, a tetragonal-phased LLZ was formed at similar synthesis condition. Upon addition of Ga at x=0.10, a cubic-phased LLZ with an impurity, $La_2Zr_2O_7$ was present. At x=0.20 and 0.30, pure cubic-phased LLZ were formed.

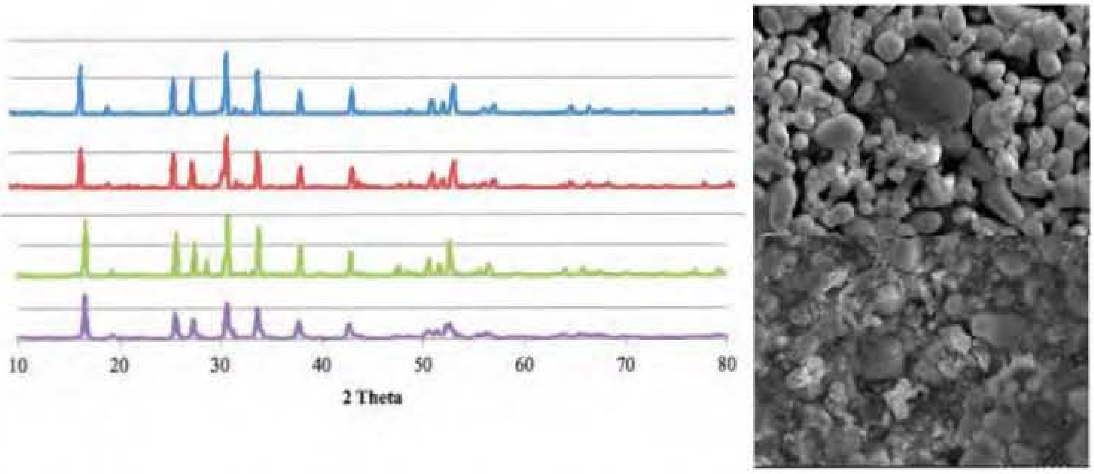

Figure 1. XRD patterns of (a) Pure LLZ (b) 0.1 Ga-LLZ (c) 0.2 Ga-LLZ and (d) 0.3 Ga-LLZ samples sintered at 1000°C, 15 h; SEM images of (e) Pure LLZ and (f) 0.3 Ga-LLZ sintered at 1000°C, 15 h

SEM images in Fig. 1 e-f have shown that Ga-doped LLZ exhibits reduced visible pores and better contact among particles, as compared to pure LLZ. Furthermore, EDS revealed a uniform elemental distribution of in the synthesized material. A pure cubic-phased and more densed LLZ material is expected to have reduced grain boundary resistance and therefore, improved ionic conductivity. At x=0.20, the Li-ion conductivity measured through EIS reached 1.59×10^{-6} S/cm, and increased to 1.34×10^{-5} S/cm upon addition of 10% excess Li source.

Conclusion

In this study, cubic-phased Ga-doped LLZ with Li-ion conductivity up to 1.34×10^{-5} S/cm were successfully synthesized via solid-state reaction, at relatively low sintering temperature of 1000°C and sintering time of 15h.

Acknowledgments

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Reference

[1] C. Bernuy-Lopez, et.al, "ChemInform Abstract: Atmosphere Controlled Processing of Ga-Substituted Garnets for High Li-Ion Conductivity Ceramics.," *ChemInform*, vol. 45, no. 33, p. no–no, Aug. 2014.