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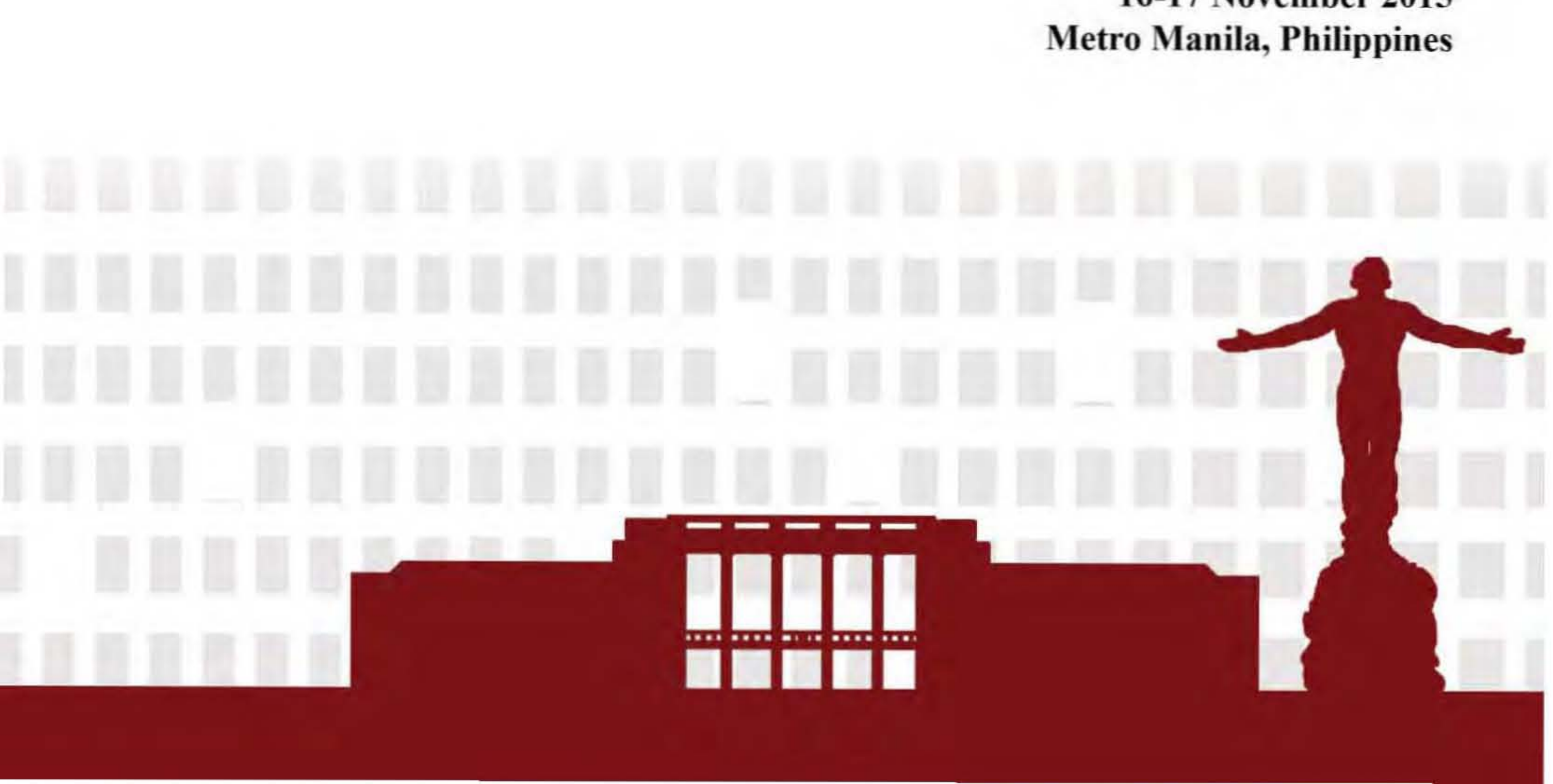

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PROCESSING AND PROPERTIES OF $Li_7la_3zr_2o_{12}$ Li-Ion CONDUCTING SOLID ELECTROLYTE FOR LITHIUM-ION BATTERIES

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ABSTRACT

$Li_7La_3Zr_2O_{12}$ (*LLZ*) solid electrolytes were synthesized via conventional solid state reaction using different temperatures. Characterization was carried out for crystal structure (XRD), ionic conductivity (EIS) and surface morphology (SEM). XRD results show that sintering at higher temperature favors the formation of cubic LLZ and increases the stability of LLZ pellets. Moreover, the study identified 1150°C as an optimum sintering temperature that obtained a relatively high ionic conductivity 8.15 x 10^{-5} Scm^{-1} at room temperature. The increase in ionic conductivity is also attributed to its high relative density value of 92.88%. The study suggests that low ionic conductivity of LLZ is caused mainly by poor crystal structure, low relative density, and the impurities present in LLZ pellet.

Keywords: Ionic Conductivity, $Li_7La_3Zr_2O_{12}$, Lithium-ion Battery, Solid Electrolyte

Introduction

Solid electrolytes have long been a demand for lithium-ion batteries to replace organic liquid electrolytes due to flammability and stability issues. The challenge lies on the discovery of solid electrolytes that possess both high ionic conductivity and good chemical stability against other metals which can improve the high energy density requirement of lithium-ion batteries suitable for electric vehicles and energy storage applications. One good example is $Li_7La_3Zr_2O_{12}$ (*LLZ*) which is proven to possess both properties. [1] This study aims to characterize LLZ solid electrolytes synthesized at varying temperatures.

Methodology

LLZ was synthesized via conventional solid-state reaction. Stoichiometric amounts of Li_2CO_3 (Techno Pharmchem, 99.5%; dried at 180°C for 6 h; 10 wt% was added to compensate for Li losses), La_2O_3 (Techno Pharmchem, 99%; dried at 900°C for 2h) and ZrO_2 (Emfutur, 97.2%; dried at 180°C for 6 h) were ground using agate mortar and then calcined at 900°C for 2h. The powder mixture were pelletized and sintered at 1000°C, 1150°C and 1200°C for 36 h using alumina crucibles. In addition, sintering at 1150°C for 15 h was also carried out to investigate the effect of sintering duration on the relative density of LLZ pellet. The sintered LLZ pellets were characterized for its structure (XRD), ionic conductivity (EIS) and surface morphology (SEM).

Results and Discussions

Figure 1 shows the effect of sintering temperature on the structure of LLZ pellets. Tetragonal LLZ is formed from 1000°C to 1150°C while cubic LLZ is formed at 1200°C. In addition, increasing the temperature stabilizes the tetragonal LLZ as depicted by the narrowing and reduction of splitting of peaks.

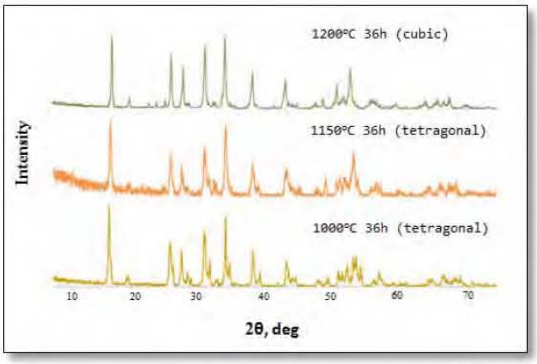

Figure 1. Stacked XRD of LLZ pellets sintered at 1000°C, 1150°C and 1200°C for 36 h

Although 1200°C produced a cubic LLZ, the presence of impurity lowers the ionic conductivity of which 2.34 x10^{-7}S cm^{-1}, 5.25 x 10^{-5} S cm^{-1}, and 2.30 x 10^{-6} S cm^{-1}at 1000°C, 1150°C and 1200°C, respectively. Results show that LLZ sintered at 1150°C has the highest conductivity due to its more defined tetragonal structure and absence of impurities. It can be seen from Figure 2 SEM images the agglomeration of grains when sintered at 1150°C for 36 h thereby reducing pores and producing a denser pellet. The calculated relative densities are (a) 91.64% and (b) 92.88% using Archimedes principle.

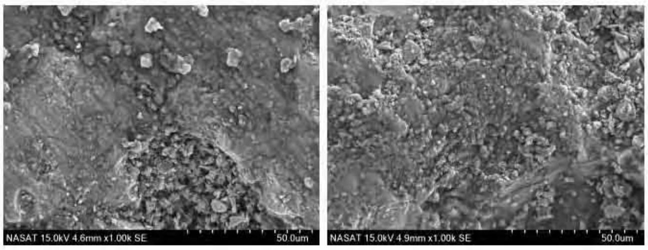

Figure 2. SEM images of LLZ sintered at 1150°C for (a) 15 h and (b) 36 h

Conclusion

The present study suggests that low ionic conductivity of LLZ solid electrolyte is caused mainly by poor crystal structure, low relative density, and the impurities present in LLZ pellet.

Acknowledgement

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Reference

[1] Murugan, R., Thangadurai, V., & Weppner, W. (2007). Fast lithium ion conduction in garnet-type Li(7)La(3)Zr(2)O(12). *Angewandte Chemie (International Ed. in English)*, *46*(41), 7778–7781. doi:10.1002/anie.200701144