**Effect of sintering temperature on Li1.2Ti1.8Al0.2(PO4)3 solid electrolyte prepared via conventional techniques**

M.I. Kimpa1,2, M.Z.H. Mayzan1, J. A. Yabagi1,3, M.M. Nmaya1,3, M.A. Agam1\*

1) Materials Physics Laboratory (MPL), Faculty of Applied Science and Technology, Pagoh Higher Education Hub, Universiti Tun Hussein Onn Malaysia, 84600 Panchor, Johor, Malaysia

2) Department of Physics, School of Physical Science, Federal University of Technology Minna, Nigeria. P.M.B. 65, Minna, Niger State, Nigeria

3) Department of Physics, Faculty of Applied Sciences, Ibrahim Badamasi Babangida University Lapai, P.M.B. 10, Lapai, Niger State, Nigeria.

\*Corresponding e-mail: [arif@uthm.edu.my](mailto:arif@uthm.edu.my) Kimpa@futminna.edu.ng

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ABSTRACT – Li1.2Ti1.8Al0.2(PO4)3 (LATP) has been synthesized via conventional solid state techniques to study the influence of sintering temperature on lithium ion conductivity between 800 to 1000 °C. The optimum sintering parameter were identify through bulk density measurement. The size of the particles was characterized via particle size analyzer. The electrical property was measured by impedance spectroscopy. The highest ionic conductivity was observed for the samples sintered at 900 °C. The fabrication of Li1.2Ti1.8Al0.2(PO4)3 solid electrolyte is consider to be use as electrolyte materials in lithium ion rechargeable batteries.

# INTRODUCTION

Lithium ion batteries (LIBs) are favoured as energy source in various modern electronic devices such as laptop, tablet and mobile phone [1]. Presently, the rechargeable Li-ion battery has been expended to be used as power source for modern electric vehicles and storage device for renewable energy [2–4].

The parent composition of this material is related to NaZr2(PO4)3 (NZP) and belongs to sodium ion conducting solid electrolyte [5, 6] deduced from the LiTi2(PO4)3 (LTP) compound. This system is widely studied as solid electrolytes for migration of lithium ions as mobile ion situated at their lattices [7–9]. The structure of such compounds consists of PO4 tetrahedrons sharing corners with TiO6 octahedrons to form conduction channel for Li+ cations [6, 10]. Ionic conductivities is enhanced by partial substitution of aluminium content within the LiTi2(PO4)3 system which at present posses high ionic conductivities among inorganic lithium solid electrolytes [11, 12].

There are many reports in literature related to the substitution of aluminium in LTP system, in which the majority of the reports were based on various syntheses [13–15]. Currently, Li1+xTi2-xAl*x*(PO4)3 system at *x* = 0.0, 0.3 and 0.4 were the most studied composition, few works were reported on composite with *x* = 0.2 and the effect of bulk density on ionic conductivity was not explicitly fully reported.

This research work is carried out to study the effect of sintering temperature on the ionic conductivity of Li1.2Ti1.8Al0.2(PO4)3 composition.

1. **METHODOLOGY**

Li1.2Ti1.8Al0.2(PO4)3 materials were synthesized by conventional solid state techniques using raw materials Li2CO3 (99.99 % Alfa Aesa), TiO2 (99.99 % Alfa Aesar), Al2O3 (99.99 % Alfa Aesar) and NH4H2PO4 (98 % Alfa Aesar). The raw materials were initially ground in an agate mortar for 1 hour and calcined in alumina crucible at 450 and 950 °C for 1 h 30 mins and 2 h respectively. The calcined sample was further milled using planetary mono mill machine. The milled powders were then pressed into pellets of 13 mm diameter and 2-3 mm thickness using uniaxial hydraulic pressing machine and sintered at 800, 850, 900, 950 and 1000 °C for 8 hours with 5°C/min heating and cooling rates. Bulk density of the sintered pellet was determined using Archimedes principle (Mettler Toledo Density kit XS-64).

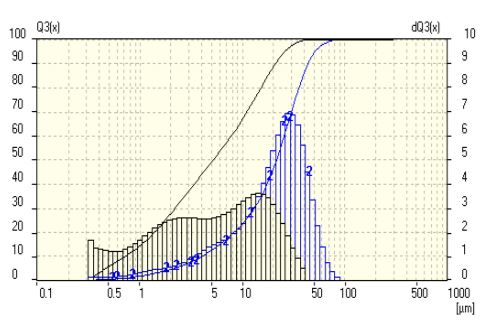
The synthesized material was characterized using particle size analyzer (Fritsch F500 analysette 22). The conductivity of the optimum data was performed using Agilent 4294A precision impedance analyzer in the frequency range 40 Hz to 2 MHz at room temperature.

1. **RESULTS AND DISCUSSION**

Figure 1 shows the bulk density of Li1.2Ti1.8Al0.2(PO4)3 composition sintered at various sintering temperature from 800 to 1000 °C. The bulk density data decrease from 800 °C sintering temperature to 850 °C. Further increase in temperature rise the bulk density to 2.83 g/cm3 at 900 °C and suddenly decrease the bulk density with increase in sintering temperature above 900 °C. The optimum sintering temperature is observed at 900 °C which indicate complete densification of grains. Figure 2 indicates the particle size of Li1.2Ti1.8Al0.2(PO4)3 powder sample based on volume and frequency distributions of the particles. The distribution is in form of bell shape with average particle size of 8.49 μm. Figure 3 shows the electrical conductivity measurement obtained from the impedance spectra and the fitted data. The impedance spectra consist of three region; the first region indicate the resistance of electrode *Rs*, the second region is the first semi circle known as bulk resistance and the third region is the second semi circle which contains the grain boundary resistance. These semi circles were identify by the fitted equivalent circuit shown as an inset in Figure 3. The semi circle is not clearly identify from the impedance plot due to low grain boundary which somehow could increase the ionic conductivity of the material. The conductivity value for Li1.2Ti1.8Al0.2(PO4)3 composition is calculated using *σ = t/RA* where *“σ”* is the dc conductivity, *“t”* is the sample thickness, *“R”* is the resistance value along *x*-axis of the impedance plot and *“A”* is the area of the sample.



Figure 1 Bulk density of Li1.2Ti1.8Al0.2(PO4)3 composition



Particle size (µm)

Volume Distribution (%)

Frequency distribution

Figure 2 Particle size distribution of Li1.2Ti1.8Al0.2(PO4)3 composition



Figure 3 Impedance spectra and its fitted data for Li1.2Ti1.8Al0.2(PO4)3 composition

# CONCLUSION

In this work, solid electrolytes Li1.2Ti1.8Al0.2(PO4)3 NASICON structure were successfully prepared via solid state reaction method. The bulk density of 2.83 g/cm3 was obtained at optimum sintering temperature of 900 °C with average particle size of 8.49 μm. This material has potential to be used as solid electrolyte in lithium ion rechargeable battery.

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