The Conductivity Enhancement of 1.5Li$_2$O-P$_2$O$_5$ Solid Electrolytes by Montmorillonite Addition

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Abstract: Most solid electrolyte materials have not shown enough conductivity to be used as an electrolyte for a battery in electronic devices. The mixture of 1.5 Li$_2$O and P$_2$O$_5$ has been reported to show a good conductivity higher than that of Li$_3$PO$_4$, which is thought to be due to phase mixtures that are formed during manufacturing process. Montmorillonite (MMT) was used to explore the effect of phase mixture on conductivity of new 1.5Li$_2$O-P$_2$O$_5$-MMT solid electrolyte composite, which was prepared through conventional solid-state reaction procedures. This study was conducted, how the addition of MMT affects process of forming 1.5Li$_2$O-P$_2$O$_5$-MMT compound, and whether it influences electrical properties and permittivity of compound. Morphology, hygroscopicity, and electrochemical characteristics of this material were analyzed in this study. The shape of glassy-like flakes was reduced in micrographs, and granular lumps were getting larger as MMT was added. Addition also tended to reduce hygroscopicity, as indicated by a reduced rate of porous absorption. Whole Nyquist plot consisted of only one imperfect semicircular arc, indicating only one relaxation process occurred in materials. Capacitance of all arcs indicated main contribution of response was from bulk material. Slope of dielectric loss of samples indicated that conduction in the samples was mainly dominated by dc conduction. MMT clays acted as a medium that absorbed liquid phase in solid-state reaction, increasing formation of dominant phase, which determined total conductivity of compound. Conductivity was higher than that of Li$_3$PO$_4$, where the sample of 20 wt% MMT addition was most polarizable and most dielectric compound.

Keywords: Lithium Phosphate, Li$_2$O-P$_2$O$_5$, solid electrolyte, Montmorillonite, conductivity, Electrochemical Impedance Spectroscopy

1. Introduction

Batteries play a very important role, both as energy storage devices, and as energy suppliers, in electrical and electronic devices. However, research on batteries has not yet found Lithium-ion battery specifications that meet the application requirements, namely high energy density, long service life, good safety factors, and wide usage temperature range (Koniak & Czerepicki, 2017). Researchers broadly see the aspects in the battery that make it possible to improve its performance, including electrolyte material, stable electrode material, high energy density, additives, binders, current collector, and efficient packaging (Oleg et al., 2022). Among these aspects, electrolytes play an important role in the development of battery technology.

Traditional batteries, such as lithium-ion batteries in mobile phones, generally depend on the flow of charge in a liquid electrolyte made of lithium salt solution in organic solvents. This will cause an irreversible decrease in the capacity, as a result of the formation of a stable layer between the electrodes and electrolytes, which is often called the Solid Electrolyte Interphase (SEI). In addition, the use of liquid electrolytes also inhibits the increase in the battery life cycle, limits the temperature range of use, makes packaging difficult, and causes leakage, and safety problems in the batterie (Lin et al., 2020). Therefore, the utilization of solid inorganic electrolytes with high thermal stability is an interesting choice to replace liquid organic electrolytes, to solve problems related to loss of capacity, lifetime, and safety issues (Sahu et al., 2014; Purwamargapratala et al., 2020). Because it has the same function as liquid electrolytes, some conditions absolutely must be met by solid electrolyte materials, including having high room temperature conductivity, negligible electronic conductivity with high ionic transfer value, and considerable electrochemical stability (Kaur et al., 2021). One important requirement for obtaining high ionic conduction is that the microstructure of solid ionic material has disorders, which can be attempted by raising the temperature to increase the number of intrinsic defects, or by adding impurities to create vacancies or defects in the structure (Hou et al., 2018). In general, solid electrolytes have advantages in terms of ease of design to make greater battery density, do not cause leakage, and have better resistance to collisions and vibrations (Guo et al., 2022). Solid electrolytes are also conductors which only deliver one type of charge so during operation are very beneficial to reduce the overpotential of the cells (Quartarone and Mustarelli, 2011; Ohno et al., 2021). In addition, solid electrolytes show better electrochemical stability and good compatibility with higher potential cathodes for increasing their energy density (Xiaxin et al., 2016). Moreover, some solid electrolytes have a
conductivity value equivalent to liquid electrolytes, with negligible electronic conductivity values (Kartini et al., 2014).

Phosphate oxide-based electrolytes attract a lot of attention to be applied as solid electrolytes, ceramic glass, amorphous semiconductors, and optoelectronic devices (Kartini et al., 2014). This conductor material is relatively easy to prepare, resistant to heat and vibration, has a large coefficient of heat expansion, and the choice of composition is varied (Das et al., 2008). However, its use has become very limited due to its poor chemical resistance. Therefore, the development of phosphate-based conductors that are modified into alloys or composites, is an interesting thing to do further.

The Li$_2$O-P$_2$O$_5$ system has three polymorphs that can be prepared through the reaction of melt-quench solids, as shown in the Naka no phase diagram (Masaki et al., 2022), namely Lithium Orthophosphate Li$_3$P$_2$O$_7$, Lithium Pyrophosphate Li$_2$P$_2$O$_7$, and Lithium Metaphosphate LiP$_2$O$_5$. However, these phases show low enough conductivity to be applied as a solid electrolyte to the battery. Research on the development of phosphate oxide-based solid electrolytes in the Li$_2$O-P$_2$O$_5$ system has been widely reported in the form of scientific articles. All these studies are carried out in a variety of compositions for improvements using characterization techniques, to investigate various aspects of solid electrolyte composition, including microstructure, physical properties, thermal properties, and electrochemical properties, to find ideal solid electrolytes stable with good performance to be applied in a cell (Jodi et al., 2017).

The approaches to increase the conductivity are mostly done by adding one or more other metal oxides, such as aluminium, titanium, etc., into new alloys, or modifying them into composites, with the addition of carbonates or sulphates (Raguenet et al., 2012; Xie et al., 2022; Purwamargapratala et al., 2019). All approaches have one thing in common: creating defects or disorders within the material framework. That is because irregularities or defects up to a certain concentration in the material provide more space for ions to move and polarize so it is expected to be able to increase its conductivity. The addition of clay montmorillonite to Li$_3$P$_2$O$_7$ compounds to form composites increased the conductivity of the compound (Jodi et al., 2016; Purwamargapratala et al., 2022; Takahashi et al., 2013).

The conductivity of the Li$_2$O-P$_2$O$_5$ compound also was improved by modifying the composition from stoichiometry (Muhammad et al., 2020). The deviation from the stable phase composition makes some materials do not react with other precursors but it reacts with the gas in the environment. This can cause oxidation or reduction reactions, which result in defects or impurity phases and increase the conductivity.

In this study, montmorillonite (MMT) was used as an additive to the 1.5Li$_2$O-P$_2$O$_5$ compound which is a composition that deviates from the stable phase of Li$_2$O-P$_2$O$_5$. MMT is a soft phyllosilicate microcrystalline mineral that can be found in almost all corners of the world and can be extracted from bentonite. MMT has a multi-layered structure in which two tetrahedral silicate layers and an octahedral Aluminate layer form flat plate sheets in a sandwich configuration, in which between the plate sheets are cavities that can be filled with cations, as well as water molecules. This structure is expected to be able to store or receive ions from the outside, or in other words, be a pathway for ion movement. Free space and weak bonds between atoms in the MMT structure are expected to make it easier for atoms to vibrate, that matter can be proven by looking at the value of the dielectric characteristic of the new composite. How far the addition of MMT to the Li$_2$O-P$_2$O$_5$ system will influence the dielectric performance of the system and its conductivity, further research is needed. This study was conducted determine the effect of adding MMT affects the process of forming 1.5Li$_2$O-P$_2$O$_5$-MMT compound, and whether it influences the electrical properties and permittivity of the compound.

2. Experimental method

2.1. Materials

Li$_2$CO$_3$ (Lithium Carbonate, Alfa Caesar, 99%), and NH$_4$H$_2$PO$_4$ (Ammonium dihydrogen Phosphate, Merck, 98%) were the precursors used to synthesize the Li$_i$O-P$_2$O$_5$ compound. The clay used in this study was Montmorillonite K10 (MMT K10, Sigma Aldrich). Supporting materials that were widely used are silver paste as a current collector in EIS measurements, liquid Nitrogen as a quenching medium, Stainless Steel dyes for pellet powder samples, and the furnace for the heating and sintering process. Digital scales, magnetic stirrers, ceramic mortar, glass and ceramic cups, bakers and plates, spatulas, etc., were the laboratory tools used during preparation and synthesis.

2.2. Sample preparation

The precursors were weighted with a digital scale based on a predetermined composition of 1.5Li$_2$O-P$_2$O$_5$, while the MMT clay was added in certain compositions by wt%. All were mixed using a magnetic stirrer for 2 hours and then heated gradually to a temperature of 650 °C, to remove components that are not needed in the reaction. For the solid-state reaction of 1.5Li$_2$O-P$_2$O$_5$, there is a phase change of around 630 °C from β-Li$_2$O$_5$P$_2$O$_7$ to α-Li$_2$O$_5$P$_2$O$_7$+ liquid phases (Masaki Shimoda, et al., 2022). The mixture was held for two hours at the reaction temperature and then rapidly quenched in liquid nitrogen. After being ground using a mortar for 1 hour, the mixture was dried at 80 °C for 4 hours. A part of the powder samples was formed into cylindrical pellets with a diameter of 15 mm, using press machines with a pressure of 6000 psi. Each sample is coded as LMKxx which means Lithium Phosphate MMT (1.5Li$_2$O-P$_2$O$_5$-MMT) composite with a total MMT content of xx wt%.

2.3. SEM characterization

SEM equipment integrated with EDS from JEOL, JSM 6510LA was used to take the micrograph photos of the samples for morphological characterization and elemental analysis. The powder samples were affixed to a double tape and coated with gold used for this characterization. The resulting photo from the SEM is generated from electrons being fired at the surface of the sample. The result of the interaction between electrons and atoms on the surface of the sample in the form of Back Scattering electrons, Secondary electrons, or X-rays, is captured by the detector and converted into an image that can be seen on the monitor.

The EDS (Energy Dispersive X-Ray Spectroscopy) testing was then carried out to see the elemental composition present in certain spots, which will be used as a test value to determine the composition of the compounds in the samples.

2.4. Hygroscopicity characterization

Hygroscopicity characterization quantitatively was carried out by weighing method (Muhammad et al., 2020). Although it depends on the accuracy and reliability of weight balance, this technique has advantages such as accessible and easy preparation, suitable for more than one form of sample, and does not require a special shape or form of sample. The pellet sample was weighed and placed on a glass slide and put in an enclosed chamber with a specified relative humidity of 95% and
left exposed to the air in the chamber. After a predetermined amount of time, the sample is then weighed again to measure the change in sample weight before and after being exposed to the air. The level of hygroscopicity is then calculated by considering the change in the sample weight and the surface area of the sample exposed to the air.

2.5 Impedance Spectroscopy characterization

HIOKI 3532-50 LCR meter (Electrochemical Impedance Spectrometer) was used to characterize the electrical properties of the samples. A pellet sample was held in sandwich geometry by silver paste electrodes. The impedance of the sample was measured within the 42 Hz to 5 MHz frequency range in ambient temperature. The output data, namely impedance value $Z$ and phase angle $\theta$, is then plotted in the form of a Nyquist plot as the relationship between the real part of the impedance and the imaginary part of the impedance. The value of solution resistance $R$ is obtained from the intersection of the impedance loop with the x-axis, the relaxation frequency $\omega$ is from the frequency at the peak of the circular arc, while the capacitance value is calculated from the resistance value and the relaxation frequency using the equation 1.

$$\tau = RC = \frac{1}{\omega R}$$  \hspace{1cm} (1)

Conductivity data were calculated from the impedance considering the sample constant, using the formula (Eq 2),

$$\sigma = \frac{1}{A} \frac{d}{Z R_p}$$  \hspace{1cm} (2)

where $R_p$, $d$, and $A$ are the sample polarization resistance, sample thickness (the distance between the electrodes), and the interface area between the sample and electrode. Plotted conductivity data as a frequency function was then fitted to the formula of Jonschers universal power law (Eq 3) to get the value of dc conductivity

$$\sigma(\omega) = \sigma_{dc} + \omega \sigma_i$$  \hspace{1cm} (3)

where $\sigma(\omega)$ is total conductivity, $\sigma_{dc}$ is the direct current conductivity of the sample, and $\omega \sigma_i$ is the pure dispersive component of ac conductivity. The equation for the complex permittivity is given as (Eq 4),

$$\epsilon^*(\omega) = \epsilon' + i \epsilon'' = \frac{Z_1'}{(Z_1')^2 + (Z_2')^2} + i\frac{Z_2'}{(Z_1')^2 + (Z_2')^2}$$  \hspace{1cm} (4)

where $\epsilon^*(\omega)$ is complex permittivity, while $Z$ is complex impedance. The ‘ and “ signs indicate the real part and the imaginary part of the impedance ($Z$) and permittivity ($\epsilon$).

3. Results and discussion

Figure 1 shows the SEM micrographs of 1.5Li$_2$O-P$_2$O$_5$-MMT composites produced from a secondary electron beam. The LMK00 surface has a glassy surface covered in flakes that may have come from quenched melt LiPO$_3$ glass. The surface shape of the flake was still seen on the surface of the LMK10 sample with 10% by weight MMT content. As the MMT content is added, the shape of the flake is reduced and appears to be an elongated dense lump (like a pillar) at 20% by weight MMT content. The lump size is getting clearer and bigger on larger MMT content. This may be related to the ability of MMT to absorb liquid during the reaction process.

In the synthesis of the 1.5Li$_2$O-P$_2$O$_5$ compound through the melt-quench solid reaction, a minor LiPO$_3$ phase is produced, as the solid reaction process will follow the phase diagram provided by AYu. (Ayu et al., 2016). At temperatures below 600°C, the reaction will produce a mixed phase of Li$_2$PO$_3$ + LiPO$_3$. As the temperature rises, the LiPO$_3$ phase turns to liquid, and at above 630°C, only the monoclinic Li$_2$PO$_3$ phase remains.

In the reaction without MMT, the liquid phase which has not reacted completely returns to glassy LiPO$_3$ flakes when the mixture is quenched. However, when MMT particles are added to the reaction, the liquid phase may be mixed (or absorbed) into the MTT cavity, forming a solid granular-like pillar. Because this liquid phase is evenly dispersed, as the temperature increases, evaporation of this phase is faster and more evenly distributed, producing large solid large pillars.

Elemental analysis of LMK00 using EDS provides an EDS diagram pattern which is dominated by Phosphorus and Oxygen, that interprets the Li$_2$O-P$_2$O$_5$ phases. The composition of Phosphorus and Oxygen are respectively 23.38 and 76.62 at%, implying that the alloy is dominated by the Li$_2$O-P$_5$O$_7$ phase, which is in accordance with the result analysis conducted by Jodi (Jodi et al., 2021). For LMK composites with MMT content, the EDS diagram shows the Aluminium and Silicon peaks in addition to the Phosphorus and Oxygen, which imply the presence of an aluminium-silicate layer of MMT. The composition of elements on LMK compounds from the EDS scan is summarized in Table 1 which shows that the Aluminium and Silicon compositions increased with increasing MMT content.

Some phases of the Li$_2$O-P$_2$O$_5$ compound exhibit high hygroscopic properties such as the LiPO$_3$ phase. Figure 2a shows the change in mass of the LMK samples when placed in an environment with high humidity of 95%, for 24 hours and 144 hours. The first 24 hours showed that the LMK10 sample indicates a higher level of mass change due to water vapor absorption compared to other samples. This is probably because in the LMK10 sample there are still many LiPO$_3$ glassy phases as shown by SEM results, so the vapor absorption occurs by two phases together, namely LiPO$_3$ and MMT. However, after 144 hours, along with the addition of MTT content, there was a tendency to decrease the change in sample mass. It stated that the addition of MTT content can reduce the rate of absorption of water vapor, which means reducing the hygroscopicity level as explained by the average rate of absorption of water vapor as shown in Figure 2b.

![Fig 1. SEM micrographs of LMK samples. (a) 1.5Li$_2$O-P$_2$O$_5$ with no MMT addition, (b) 10 wt%, (c) 20 wt%, and (d) 30 wt% MMT addition.](image-url)
Table 1
The composition of elements on LMK samples.

<table>
<thead>
<tr>
<th>Atomic %</th>
<th>LMK00</th>
<th>LMK10</th>
<th>LMK20</th>
<th>LMK30</th>
</tr>
</thead>
<tbody>
<tr>
<td>Oxygen-O</td>
<td>76.62</td>
<td>73.37</td>
<td>71.28</td>
<td>66.03</td>
</tr>
<tr>
<td>Phosphorus-P</td>
<td>23.38</td>
<td>21.85</td>
<td>19.15</td>
<td>20.03</td>
</tr>
<tr>
<td>Aluminium-Al</td>
<td>---</td>
<td>0.62</td>
<td>0.72</td>
<td>1.77</td>
</tr>
<tr>
<td>Silicon-Si</td>
<td>---</td>
<td>4.16</td>
<td>8.85</td>
<td>12.17</td>
</tr>
</tbody>
</table>

Figure 2. Water vapor absorption characteristic of LMK samples. a). the change in mass of samples in high humidity environment, and b). the average rate of water vapor absorption.

Table 2
Impedance parameters calculated from Nyquist impedance plot.

<table>
<thead>
<tr>
<th>Impedance parameter</th>
<th>LMK00</th>
<th>LMK10</th>
<th>LMK20</th>
<th>LMK30</th>
</tr>
</thead>
<tbody>
<tr>
<td>Resistance $R_b$ (Ω)</td>
<td>4.91x10^{13}</td>
<td>2.01x10^{13}</td>
<td>1.94x10^{13}</td>
<td>7.51x10^{13}</td>
</tr>
<tr>
<td>Relaxation frequency $\omega$ (Hz)</td>
<td>1.21x10^{7}</td>
<td>2.47x10^{7}</td>
<td>3.14x10^{7}</td>
<td>1.21x10^{7}</td>
</tr>
<tr>
<td>Relaxation time $\tau$ (s)</td>
<td>8.26x10^{4}</td>
<td>4.04x10^{4}</td>
<td>3.18x10^{4}</td>
<td>8.26x10^{4}</td>
</tr>
<tr>
<td>Capacitance $C_{r}$ (F)</td>
<td>1.68x10^{-11}</td>
<td>2.01x10^{-11}</td>
<td>1.64x10^{-11}</td>
<td>1.10x10^{-11}</td>
</tr>
</tbody>
</table>

Figure 3. shows the Nyquist impedance plot of LMK composite samples measured at room temperature, which depicts the response of the material to the electric field applied to the sample. The whole curve in Figure 3 can be said to consist of only an imperfect semi-circular arc, which indicates that in all samples only one relaxation process occurs. The intersection of the impedance loop with the x-axis indicates the resistive properties of the material or solution resistance, which shows that the LMK30 has the highest value, while the LMK20 has the smallest resistance value. Similarly, the sample with 30 w% MMT content has the highest value of the capacitive impedance. The real part (resistive) and the imaginary part (capacitive) impedance values for each sample are summarized in Table 2, which are calculated from the experimental data plot that is fitted to the circle arc equation using data analysis and Graph Plotting Software. The capacitance value is calculated using equation (1) considering the relaxation frequency and the resistance. The capacitance of all arcs is in the order of pF (picofarad), which indicates that the main contribution of the response that occurs in the sample is the response of the bulk material (Taher et al., 2016).

Figure 4 shows the impedance behaviour as a function of frequency. The resistive impedance of the LMK sample decreases with increasing frequency and is followed by a steep decrease to a certain impedance value. At higher frequencies, the resistive impedance values are likely to be coincident at low values as an indication of the charge release process (Jodi et al., 2017).
The capacitive impedance behaviour of LMK samples is characterized by the appearance of peaks at higher frequencies. This shows the existence of a relaxation process in the sample (Subohi et al., 2016). The LMK30 has a larger capacitive impedance compared to other LMK samples, while the lowest capacitive impedance is owned by the LMK20. The peak impedance frequency (relaxation frequency) value shifted towards a greater frequency as the capacitive impedance value decreased, indicating the relaxation time is getting shorter. It indicates that the LMK20 compound has a faster time to reach a new equilibrium in response to an applied electric field compared to other compositions (Thomas et al., 2017). The addition of MMT content to the compound is estimated to add immovable ion content in the dielectric sample of the Li2O-P2O5 compound and facilitate the relaxation process. Because in the dielectric material, in general, the relaxation process occurs due to the presence of an immovable charge at low temperatures and due to the presence of defects or vacancies at high temperatures (Sen et al., 2008).

The conductivity curves of LMK composite alloys as a function of measurement frequency are shown in Figure 5. The conductivity of each frequency point on this curve is calculated from the measured impedance value. The complex conductivity curves of all samples have two parts of an area, namely the area where the conductivity forms a near-plateau region and the area where the conductivity value increases with increasing frequency. The near-plateau region marks dc conductivity where conductivity is independent of the frequency, while the second one is ac conductivity which is frequency dependent (Jayswal et al., 2013). The ac conductivity indicates the material’s hopping ion conduction mechanism (Sassi et al., 2015), and has characteristics of power law in terms of angular frequency ($A\omega^s$) where $s$ is the degree of interaction between mobile ions and the lattices around them ($0 \leq s \leq 1$), and $A$ is a constant which determines the strength of polarizability (Dhahri et al., 2018). The conductivity values are obtained by fitting the measurement curves in Figure 6 to equation (3). The fitted conductivity values are described as the dotted line curve in Figure 5 and summarized in Table 3.

The conductivity of the compound increases with the addition of MMT and reaches a maximum in the LMK20. Further addition of MMT content decreases the conductivity value to smaller order than before the addition. The LMK conductivity with MMT content up to 20wt% is in order of $10^{-4}$ S/cm, two orders higher than that of the 2Li2O-P2O5 compound, and 4-5 orders higher than the conductivity of the Li3PO4 compound (Jodi et al., 2016). The exponential power value of all alloys is in the range of value $s = 0.63 \sim 0.78$. This value approaches the exponent value for the conductivity of glass material which contains a high alkaline content of $s=0.6$, which is measured in the kHz ~ MHz frequency range at room temperature (Jodi et al., 2017).

That is clearly seen that the addition of MMT is not linearly correlated with changes in the impedance and conductivity of the material. The morphology of the sample shows that the MMT acts as a medium that absorbs the liquid in the reaction, thereby accelerating the formation of the desired phase. Jodi et al stated that the addition of MMT to a certain level reduces the formation of the LiPO4 and increases the formation of the LiP2O7 which has high conductivity and is the dominant phase determining the total conductivity (Heri Jodi et al., 2021). However, more detailed research is needed to determine the direct correlation of changes in MMT volume to changes in Li4P2O7 conductivity. Further addition of MMT makes its own conductivity affect the total conductivity to be lower than the conductivity of the dominant phase.
10-4 S/cm. LMK20 composites become the most polarizable and reaches a minimum in the LMK content. The bulk resistance shows a decrease along with the addition of MMT content. The impedance response of all compounds is Aluminium and Silicon elements along with the addition of Oxygen and Phosphorus elements in all LMK compounds, where there is an increase in the content of MMT content. The impedance response of all compounds is dominated by the grain response, indicated by a semi-circle on the impedance curve, and picofarad order of capacitance value.

4. Conclusion
The addition of MMT content to the 1.5Li2O-P2O5 compound formed a new 1.5Li2O-P2O5-MMT (LMK) electrolyte composite. MMT content in LMK composites influences the morphology of the compound which changes from a structure covered in glassy shapes to pillar-like lumps. It is believed that MMT acts as a medium that absorbs the liquid phase in the solid-state reaction and forms pillar-like lumps. EDS analysis shows the predominance of Oxygen and Phosphorus elements in all LMK compounds, where there is an increase in the content of MMT content. The impedance response of all compounds is dominated by the grain response, indicated by a semi-circle on the impedance curve, and picofarad order of capacitance value.

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