Conductivity and electrochemical stability of Li\(^+\) substituted high-entropy Li\(_x\)(Mg\(_{0.2}\)Co\(_{0.2}\)Ni\(_{0.2}\)Cu\(_{0.2}\)Zn\(_{0.2}\))\(\text{O}\) ceramics

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Abstract

In this work, high-entropy Li\(_x\)(Mg\(_{0.2}\)Co\(_{0.2}\)Ni\(_{0.2}\)Cu\(_{0.2}\)Zn\(_{0.2}\))\(\text{O}\) (where \(x = 0, 0.1, 0.2, 0.3, 0.4\)) ceramics with rock-salt structure were synthesized via simple sol-gel method and sintered at 1000 °C. The crystal structure, cross-section micromorphology, conductivity, electronic conductivity and electrochemical stability of the sintered Li\(_x\)(Mg\(_{0.2}\)Co\(_{0.2}\)Ni\(_{0.2}\)Cu\(_{0.2}\)Zn\(_{0.2}\))\(\text{O}\) ceramics were investigated by X-ray diffraction, SEM, AC-impedance, DC-polarization and cyclic voltammetry, respectively. The Li\(_x\)(Mg\(_{0.2}\)Co\(_{0.2}\)Ni\(_{0.2}\)Cu\(_{0.2}\)Zn\(_{0.2}\))\(\text{O}\) ceramics present single rock-salt structure and no impurities were detected. The Li\(_{0.3}\)(Mg\(_{0.2}\)Co\(_{0.2}\)Ni\(_{0.2}\)Cu\(_{0.2}\)Zn\(_{0.2}\))\(\text{O}\) (where \(x = 0.3\)) sample has the highest conductivity of 1.46 × 10\(^{-5}\) S/cm at 30 °C, with an activation energy of 0.334 eV. Conductivity of the Li\(_x\)(Mg\(_{0.2}\)Co\(_{0.2}\)Ni\(_{0.2}\)Cu\(_{0.2}\)Zn\(_{0.2}\))\(\text{O}\) ceramics is enhanced by about four orders of magnitude with the partial substitution of (Mg\(_{0.2}\)Co\(_{0.2}\)Ni\(_{0.2}\)Cu\(_{0.2}\)Zn\(_{0.2}\))\(\text{O}\) by lithium. The electronic conductivity of the Li\(_{0.3}\)(Mg\(_{0.2}\)Co\(_{0.2}\)Ni\(_{0.2}\)Cu\(_{0.2}\)Zn\(_{0.2}\))\(\text{O}\) ceramics is much lower than its ionic conductivity, which suggests that Li\(_{0.3}\)(Mg\(_{0.2}\)Co\(_{0.2}\)Ni\(_{0.2}\)Cu\(_{0.2}\)Zn\(_{0.2}\))\(\text{O}\) can be treated as ionic conductor. The Li\(_{0.3}\)(Mg\(_{0.2}\)Co\(_{0.2}\)Ni\(_{0.2}\)Cu\(_{0.2}\)Zn\(_{0.2}\))\(\text{O}\) sample is electrochemically stable between 1.23 and 4.7 V.

Keywords: high-entropy ceramics, sol-gel synthesis, solid electrolyte, rock-salt, ionic conductivity

I. Introduction

Traditional Li-ion batteries suffer from safety problems such as burning and explosion due to the leakage risk of liquid organic electrolyte. [1] Therefore, all-solid-state Li batteries (ASSLB) with high safety property have gained more and more interests. [2] Solid electrolyte plays the key part of ASSLB and currently cannot meet the application requirements of ASSLB. Development of new solid electrolytes has been a hot topic in recent years. [3].

Based on high entropy ceramics, varied novel solid electrolytes could be designed and synthesized. The composition of high entropy ceramics can be very different and adjusted and the structure has high designability. High-entropy ceramics have been widely studied for application in thermal protection, electro-catalysis, energy storage/conversion and electronic devices [4–6]. Thus, Wang et al. [7,8] reported on the application of high entropy oxide Li\(_x\)(Co\(_{0.2}\)Cu\(_{0.2}\)Mg\(_{0.2}\)Ni\(_{0.2}\)Zn\(_{0.2}\))\(\text{O}\) for a novel cathode material in Li-ion batteries. High entropy oxide (Mg\(_{0.2}\)Co\(_{0.2}\)Ni\(_{0.2}\)Cu\(_{0.2}\)Zn\(_{0.2}\))\(\text{O}\) was a promising chemical anchor [9] to restrain shuttle effect of dissolution of lithium polysulphides for Li-S batteries. High entropy ceramics and its derivatives exhibit good reversible capacity and cycling stability, which is promising for application in Li-ion batteries. [10–13].

For solid electrolyte, Berardan et al. [14] reported a series of high entropy ceramic-based solid electrolytes with promising properties. For example, (Mg\(_{0.2}\)Co\(_{0.2}\)Ni\(_{0.2}\)Cu\(_{0.2}\)Zn\(_{0.2}\))\(\text{O}\) exhibits a Na\(^+\) conductivity of 6.00 × 10\(^{-6}\) S/cm at room temperature whereas Na\(_{0.15}\)(Mg\(_{0.2}\)Ni\(_{0.2}\)Cu\(_{0.2}\)Zn\(_{0.2}\))\(\text{O}\) exhibits a Na\(^+\) conductivity of 6.00 × 10\(^{-6}\) S/cm at room temperature.

In this study, a series of Li-ion conducting solid electrolytes with the general formula of Li\(_x\)(Mg\(_{0.2}\)Co\(_{0.2}\)Ni\(_{0.2}\)Cu\(_{0.2}\)Zn\(_{0.2}\))\(\text{O}\) (where \(x = 0, 0.1, 0.2, 0.3, 0.4\)) were designed based on (Mg\(_{0.2}\)Co\(_{0.2}\)Ni\(_{0.2}\)Cu\(_{0.2}\)Zn\(_{0.2}\))\(\text{O}\) high-entropy ceramics. The Li\(_{0.3}\)(Mg\(_{0.2}\)Co\(_{0.2}\)Ni\(_{0.2}\)Cu\(_{0.2}\)Zn\(_{0.2}\))\(\text{O}\) ceramics were
synthesized via a simple sol-gel method, calcined and sintered at 1000°C. Crystalline structure, microstructure, conductivity and electrochemical stability of Li$_x$(Mg$_{0.2}$Co$_{0.2}$Ni$_{0.2}$Cu$_{0.2}$Zn$_{0.2}$)$_{1-x}$O ceramics (where $x = 0, 0.1, 0.2, 0.3$ and 0.4) were synthesized by using sol-gel method. C$_6$H$_8$NiO$_4$·4H$_2$O (AR, GUOYAO reagent Ltd.), C$_4$H$_6$CoO$_4$·4H$_2$O (AR, Kelong reagent Ltd.), C$_6$H$_8$MgO$_4$·4H$_2$O (AR, Maklin reagent Ltd.), C$_4$H$_6$CuO$_4$·4H$_2$O (AR, Yongda reagent Ltd.), C$_6$H$_8$ZnO$_4$·2H$_2$O (AR, Yongda reagent Ltd.) and C$_6$H$_8$O$_2$·H$_2$O (AR, GUOYAO reagent Ltd.) were used as starting materials. All reagents were weighted in stoichiometric amounts and dissolved in deionized water. In a typical sol-gel procedure, 0.02 mol C$_6$H$_8$NiO$_4$·4H$_2$O (AR, Maklin reagent Ltd.) were used as starting materials. All reagents were weighted in stoichiometric amounts and dissolved in deionized water. 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Figure 2. SEM images of \( \text{Li}_x(\text{Mg}_{0.2}\text{Co}_{0.2}\text{Ni}_{0.1}\text{Cu}_{0.3}\text{Zn}_{0.2})_3\) ceramic pellets for \( x = 0.01, 0.02, 0.03 \text{ and } 0.04 \)

Figure 3. SEM images of \( \text{Li}_0.3(\text{Mg}_{0.2}\text{Co}_{0.2}\text{Ni}_{0.1}\text{Cu}_{0.3}\text{Zn}_{0.2})_3\) ceramics sintered at different temperatures

Table 2. Lattice parameter and density of samples

<table>
<thead>
<tr>
<th>Samples</th>
<th>Lattice parameters [Å]</th>
<th>Theoretical density [g/cm³]</th>
<th>Measured density [g/cm³]</th>
<th>Relative density [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>( (\text{Mg}<em>{0.2}\text{Co}</em>{0.2}\text{Ni}<em>{0.1}\text{Cu}</em>{0.3}\text{Zn}_{0.2})_3 )O</td>
<td>4.2068</td>
<td>6.26616</td>
<td>3.83</td>
<td>61</td>
</tr>
<tr>
<td>( \text{Li}<em>{0.1}(\text{Mg}</em>{0.2}\text{Co}<em>{0.2}\text{Ni}</em>{0.1}\text{Cu}<em>{0.3}\text{Zn}</em>{0.2})_3)O</td>
<td>4.2061</td>
<td>6.08222</td>
<td>3.89</td>
<td>63</td>
</tr>
<tr>
<td>( \text{Li}<em>{0.2}(\text{Mg}</em>{0.2}\text{Co}<em>{0.2}\text{Ni}</em>{0.1}\text{Cu}<em>{0.3}\text{Zn}</em>{0.2})_3)O</td>
<td>4.20836</td>
<td>5.89508</td>
<td>3.91</td>
<td>66</td>
</tr>
<tr>
<td>( \text{Li}<em>{0.3}(\text{Mg}</em>{0.2}\text{Co}<em>{0.2}\text{Ni}</em>{0.1}\text{Cu}<em>{0.3}\text{Zn}</em>{0.2})_3)O</td>
<td>4.20889</td>
<td>5.71337</td>
<td>4.08</td>
<td>71</td>
</tr>
<tr>
<td>( \text{Li}<em>{0.4}(\text{Mg}</em>{0.2}\text{Co}<em>{0.2}\text{Ni}</em>{0.1}\text{Cu}<em>{0.3}\text{Zn}</em>{0.2})_3)O</td>
<td>4.21637</td>
<td>5.50448</td>
<td>4.11</td>
<td>75</td>
</tr>
</tbody>
</table>

Table 3. \( AS_{mix} \) values of high-entropy ceramic samples

<table>
<thead>
<tr>
<th>Samples</th>
<th>( \sigma_{30°C} ) [S/cm]</th>
<th>( E_a ) [eV]</th>
</tr>
</thead>
<tbody>
<tr>
<td>( (\text{Mg}<em>{0.2}\text{Co}</em>{0.2}\text{Ni}<em>{0.1}\text{Cu}</em>{0.3}\text{Zn}_{0.2})_3)O</td>
<td>7.44 \times 10^{-9}</td>
<td>4.60 \times 10^{-6}</td>
</tr>
<tr>
<td>( \text{Li}<em>{0.1}(\text{Mg}</em>{0.2}\text{Co}<em>{0.2}\text{Ni}</em>{0.1}\text{Cu}<em>{0.3}\text{Zn}</em>{0.2})_3)O</td>
<td>6.12 \times 10^{-5}</td>
<td>0.392</td>
</tr>
<tr>
<td>( \text{Li}<em>{0.2}(\text{Mg}</em>{0.2}\text{Co}<em>{0.2}\text{Ni}</em>{0.1}\text{Cu}<em>{0.3}\text{Zn}</em>{0.2})_3)O</td>
<td>1.46 \times 10^{-5}</td>
<td>0.334</td>
</tr>
<tr>
<td>( \text{Li}<em>{0.3}(\text{Mg}</em>{0.2}\text{Co}<em>{0.2}\text{Ni}</em>{0.1}\text{Cu}<em>{0.3}\text{Zn}</em>{0.2})_3)O</td>
<td>7.82 \times 10^{-8}</td>
<td>0.342</td>
</tr>
</tbody>
</table>

Conductivity was calculated by the following equation:

\[
\sigma = \frac{d}{R \cdot S}
\]  

(2)

All samples correspond to rock-salt structure and no diffraction peaks of impurity phase were detected. With the increase of lithium content, diffraction peaks of the \( (\text{Mg}_{0.2}\text{Co}_{0.2}\text{Ni}_{0.1}\text{Cu}_{0.3}\text{Zn}_{0.2})_3\)O ceramics shifted to lower angles. This phenomenon is attributed to the bigger ionic radius of Li\(^{+}\) (0.076) than Cu\(^{2+}\) (0.073), Mg\(^{2+}\) (0.072), Co\(^{2+}\) (0.065), Ni\(^{2+}\) (0.069) and Zn\(^{2+}\) (0.074). This result suggests that Cu\(^{2+}\) (0.073), Mg\(^{2+}\) (0.072), Co\(^{2+}\) (0.065), Ni\(^{2+}\) (0.069) and Zn\(^{2+}\) (0.074) were partly substituted by Li\(^{+}\) successfully. The lattice parameters of all samples were calculated and listed in Table 2.

Cross-section morphologies of the fractured high-entropy ceramic pellets were shown in Fig 2. Loose connection between grains was observed in SEM images. The grain size distribution of the samples is not uniform. Furthermore, grain size of the \( (\text{Mg}_{0.2}\text{Co}_{0.2}\text{Ni}_{0.1}\text{Cu}_{0.3}\text{Zn}_{0.2})_3\)O ceramics increases slightly with the increase of lithium content. Porosity was observed in SEM images for all samples (Fig. 3) and corresponding densities of the sintered samples are listed in Table 2. As calculated, relative densities of the \( (\text{Mg}_{0.2}\text{Co}_{0.2}\text{Ni}_{0.1}\text{Cu}_{0.3}\text{Zn}_{0.2})_3\)O ceramic pellets are only about 70 %TD. The low density could hinder the transport of lithium ions greatly. According to the experience, relative density of oxiderceramic type solid electrolyte should reach at least 90% to achieve rapid ion conduction (ionic conductivity of \( 10^{-7} \) S/cm or higher) at room temperature.

Figure 4a shows the Nyquist plots of \( (\text{Mg}_{0.2}\text{Co}_{0.2}\text{Ni}_{0.1}\text{Cu}_{0.3}\text{Zn}_{0.2})_3\)O ceramics. Conductivity was calculated by the following equation:

\[
\sigma = \frac{d}{R \cdot S}
\]  

(2)
Figure 4. Nyquist plots of \((\text{Mg}_{0.2}\text{Co}_{0.2}\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.2})\text{O}\) (a) and \(\text{Li}_x(\text{Mg}_{0.2}\text{Co}_{0.2}\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.2})_{1-0.5x}\text{O}\) (b) at 30 °C

Figure 5. The Arrhenius plots of high-entropy ceramics

Figure 5 shows the temperature dependence of total conductivity \((\sigma)\) vs. \(1000/T\) of the high-entropy ceramics. The activation energy of the samples was calculated by the following equation:

\[
\sigma \cdot T = \sigma_0 \exp \left( - \frac{E_a}{k_B \cdot T} \right)
\]

In this equation, \(\sigma\) is the total conductivity, \(T\) is the absolute temperature, \(\sigma_0\) is the pre-exponential factor, \(E_a\) is the activation energy and \(k_B\) is the Boltzmann constant. \(E_a\) of each ceramic sample was calculated and listed in Table 3. \(E_a\) of the sample \(\text{Li}_{0.3}(\text{Mg}_{0.2}\text{Co}_{0.2}\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.2})_{0.85}\text{O}\) is 0.334 eV, which is comparable to the activation energy of traditional solid electrolytes such as LLTO (Li\(_{0.33}\text{La}_{0.56}\text{TiO}_3\)) and LLZO (Li\(_7\text{La}_{3}\text{Zr}_2\text{O}_{12}\)) [16,17].

Figure 6. DC polarization curve of \(\text{Li}_{0.3}(\text{Mg}_{0.2}\text{Co}_{0.2}\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.2})_{0.85}\text{O}\)

Figure 6 shows the DC-polarization curve of the \(\text{Li}_{0.3}(\text{Mg}_{0.2}\text{Co}_{0.2}\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.2})_{0.85}\text{O}\) sample. For solid electrolyte, it is important to distinguish between ionic and electronic conductivity. Total conductivity (ionic and electronic conductivities) of the samples was obtained by AC-impedance. Electronic conductivity of the samples could be determined by DC polarization. Due to the usage of Ag blocking electrodes, the steady current in Fig. 5 was attributed to electronic conduction only. The electronic conductivity of the \(\text{Li}_{0.3}(\text{Mg}_{0.2}\text{Co}_{0.2}\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.2})_{0.85}\text{O}\) sample was calculated by the following equation:

\[
\sigma_{\text{ele}} = \frac{I \cdot d}{U' \cdot S}
\]

The electronic conductivity of the \(\text{Li}_{0.3}(\text{Mg}_{0.2}\text{Co}_{0.2}\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.2})_{0.85}\text{O}\) sample is \(1.97 \times 10^{-9}\) S/cm, which is four orders of magnitude lower than its total conductivity and its \(\sigma_{\text{ele}}/\sigma_{\text{ion}}\) ratio was extremely low. Therefore, the contribution of electronic conduction to total conductivity is negligible in the \(\text{Li}_{0.3}(\text{Mg}_{0.2}\text{Co}_{0.2}\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.2})_{0.85}\text{O}\) ceramics. So, the results of DC polarization confirmed that this high-entropy ceramics can be treated as a Li\(^+\) conducting solid electrolyte.

Figure 7 shows CV curves of the \(\text{Li}_{0.3}(\text{Mg}_{0.2}\text{Co}_{0.2}\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.2})_{0.85}\text{O}\) as work electrode and metallic Li as counter electrode. The CV curves of the \(\text{Li}_{0.3}(\text{Mg}_{0.2}\text{Co}_{0.2}\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.2})_{0.85}\text{O}\) has no redox peak between 1.23 and 4.7 V vs. Li\(^+/\)Li. This voltage range (3.47 V) is wider than the operating voltage range of \(\text{LiNi}_0.5\text{Mn}_1.5\text{O}_4/\text{Li}_4\text{Ti}_5\text{O}_{12}\) (3.3 V), \(\text{LiFePO}_4/\text{Li}_4\text{Ti}_5\text{O}_{12}\) (2.5 V) [18] and other \(\text{Li}_4\text{Ti}_5\text{O}_{12}\)-based Li-ion batteries.
In this work, a series of rock-salt structured high-entropy ceramics, Li$_x$(Mg$_{0.2}$Co$_{0.2}$Ni$_{0.2}$Cu$_{0.2}$Zn$_{0.2}$)$_{0.85}$O (x = 0, 0.1, 0.2, 0.3, 0.4) were synthesized via a simple sol-gel method. The Li-doped high-entropy ceramics (x = 0, 0.1, 0.2, 0.3, 0.4) presented single rock-salt phase without any impurities. The sample with x = 0.3 (Li$_{0.3}$(Mg$_{0.2}$Co$_{0.2}$Ni$_{0.2}$Cu$_{0.2}$Zn$_{0.2}$)$_{0.85}$O) exhibited a high conductivity of 1.46 × 10$^{-5}$ S/cm at 30°C, with the activation energy of 0.334 eV. Electronic conductivity of Li$_{0.3}$(Mg$_{0.2}$Co$_{0.2}$Ni$_{0.2}$Cu$_{0.2}$Zn$_{0.2}$)$_{0.85}$O is 1.97 × 10$^{-9}$ S/cm, which is four orders of magnitude lower than its total conductivity. The result of cyclic voltammetry experiment suggests that Li$_{0.3}$(Mg$_{0.2}$Co$_{0.2}$Ni$_{0.2}$Cu$_{0.2}$Zn$_{0.2}$)$_{0.85}$O is stable against metal lithium in the voltage range of 1.23 to 4.7 V.

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