

**Grant Number**: EP/R024006/1 and the university of Birmingham

**Project title**: Water based synthesis of highly conductive GaxLi7-3xLa3Hf2O12 garnets with comparable critical current density to analogous GaxLi7-3xLa3Zr2O12 systems.

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### Abstract

Next generation lithium ion batteries are envisaged as those which feature an all solid-state architecture. This will enable the higher energy density storage required to meet the demands of modern society, especially for the growing electric vehicle market. Solid state batteries have, however, proved troublesome to implement commercially due to the lack of a suitable solid-state electrolyte, which needs to be highly conductive, have a low interfacial resistance and a suitably wide electrochemical stability window. Garnet materials are potential contenders for these batteries, demonstrating many of the desired properties, although there remain challenges to overcome. Here we report a facile synthesis of Li7La3Hf2O12 and Ga/AlxLi7-3xLa3Hf2O12 garnets, with the synthesis of Ga0.2Li6.4La3Hf2O12 requiring only dissolution of precursors in water and heating to 700°C. Ga0.2Li6.4La3Hf2O12, was shown to display a high room temperature conductivity (0.373 mS cm-1 at 28 °C). Moreover, in Li|garnet|Li cells, we observed a comparable critical current density compared to Ga0.2Lai6.4La3Zr2O12, despite a lower density and higher area specific resistance compared to literature values, suggesting Hf systems may be further engineered to deliver additional improvements for use in future solid state batteries.

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| **File name** | **Description** |
| Data files |
| Al-LLHO mop up -Folder and .txt files | Impedance spectroscopy measurements at difference temperatures  |
| Ga-LLHO folder and .txt files | Impedance spectroscopy measurements at difference temperatures |
| LLHO\_v2\_Li7\_Diss – folder and .txt files | Impedance spectroscopy measurements at difference temperatures |
| C.V.  | Ga0.2Li6.4La3Hf2O12 Cyclic voltammetry data |
| Nd3\_LHO.raw | Li7Nd3Hf2O12 X ray diffraction data |
| LLHOPr0.5Hf1.5.raw - LLHOPr0.5Hf1.5\_step 6 | Li7La3Hf1.5Pr0.5O12 X ray diffraction data and attempted purification |
| LLHO\_Via\_Water\_10%\_xs\_Li\_700c, LLHO\_Norm\_700c\_700c\_10% and LLHO\_RT\_Start.raw | Li7La3Hf2O12 X ray diffraction data |
| LLHO\_Al0.28\_Li6.4\_700c\_Step\_1 to step 5.raw | Al0.28Li6.16La3Hf2O12 X ray diffraction data and purification steps – first file should read LLHO\_Al0.28\_Li6.16\_700c\_Step\_1.raw |
| Gd\_LHO\_PINK.RAW | Li7Gd3Hf2O12 X ray diffraction data |
| Ga\_LLHO\_STEP\_2.RAW and Ga\_LLHO\_New\_batch.RAW | Ga0.2Li6.4La3Hf2O12 X ray diffraction data |
| Ga\_LLHO\_CE\_0.2.RAW to Ga\_LLHO\_CE\_0.2\_step\_5.raw | Ga0.2Li6.4La3Hf1.8Ce0.2O12 X ray diffraction data and attempted purification  |
| Eu\_LHO\_PINK.RAW |  Li7Eu3Hf2O12 X ray diffraction data |
| Er\_LHO.raw |  Li7Er3Hf2O12 X ray diffraction data |

**Publications**: Water based synthesis of highly conductive GaxLi7-3xLa3Hf2O12 garnets with comparable critical current density to analogous GaxLi7-3xLa3Zr2O12 systems.