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**Project title**: Evaluation of Ga0.2Li6.4Nd3Zr2O12 garnets: exploiting dopant instability to create a mixed conductive interface to reduce interfacial resistance for all solid state batteries.

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### Abstract

The next major leap in energy storage is thought to arise from a practical implementation of all solid-state batteries, which remain largely confined to the small scale due to issues in manufacturing and mechanical stability. Lithium batteries are amongst the most sought after, for the high expected energy density and improved safety characteristics, however the challenge of finding a suitable solid-state electrolyte remains. Lithium rich garnets are prime contenders as electrolytes, owing to their high ionic conductivity (> 0.1 mS cm-1), wide electrochemical window (0 – 6 V) and stability with Li metal. However, the high Young’s modulus of these materials, poor wetting of Li metal and rapid formation of Li2CO3 passivating layers tends to give a detrimentally large resistance at the solid-solid interface, limiting their application in solid state batteries. Most studies have focused on La based systems, with very little work on other lanthanides. Here we report a study of the Nd based garnet Ga0.2Li6.4Nd3Zr2O12, illustrating substantial differences in the interfacial behaviour. This garnet shows very low interfacial resistance attributed to dopant exsolution which, when combined with moderate heating (175°C, 1h) with Li metal, we suggest forms Ga-Li eutectics which significantly reduces the resistance at the Li/Garnet interface to as low as 67 Ω cm2 (much lower than equivalent La based systems). The material also shows intrinsically high density (93%) and good conductivity (≥ 0.2 mS cm-1) via conventional furnaces in air. It is suggested these garnets are particularly well suited to provide a mixed conductive interface (in combination with other garnets) which could enable future solid-state batteries.

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| **File name** | **Description** |
| Data files |
| Impedance folder – contains impedance spectroscopy data for of Li5+xNd3Nb2-xZrxO12 (0 ≤ x ≥ 2) and Li6.4, M0.2Nd3Zr2O12 (M = Al/Ga). | Impedance spectroscopy measurements at difference temperatures  |
| Impedance\_2 folder | Impedance spectroscopy measurements at difference temperatures |
| X-ray diffraction Folder | X-ray diffraction patterns for of Li5+xNd3Nb2-xZrxO12 (0 ≤ x ≥ 2) and Li6.4, M0.2Nd3Zr2O12 (M = Al/Ga). |
| Cell cycling | Symmetry cell cycling for Ga0.2Nd3Zr2O12 and cyclic voltammetry data |

**Publications**: Evaluation of Ga0.2Li6.4Nd3Zr2O12 garnets: exploiting dopant instability to create a mixed conductive interface to reduce interfacial resistance for all solid state batteries.