

Experimental report

19/03/2024

Proposal: EASY-1133

Council: 10/2022

Title: Investigating Anion Ordering in Sulfide Argyrodite Solid Electrolytes

Research area: Materials

This proposal is a new proposal

Main proposer: Bartholomew PAYNE

Experimental team:

Local contacts: Emmanuelle SUARD

Samples: Li6PS5Cl

Instrument	Requested days	Allocated days	From	To
D2B	4	4	25/05/2023	26/05/2023

Abstract:

Solid-state batteries (Li-SSBs) can address the poor range, slow charging, and safety issues associated with current Li-ion batteries by utilizing lithium metal anodes. However, commercial Li-SSBs are plagued by the poor conductivities of current solid Li-ion electrolytes. We have utilised a novel spark plasma sintering (SPS) process to produce a commercially viable Li6PS5Cl solid electrolyte with significant improvement in ionic conductivity (increasing from 1 to 14 mS/cm). Using NMR relaxometry, we have evidence that the SPS process induces local structural change which contributes to the improved Li-ion conduction. We believe the local change is increased anion ordering, with some evidence of this coming from NMR spectroscopy and X-ray PDF measurements. However, due to the similar electron configuration of S²⁻ and Cl⁻ exact site occupancies cannot be obtained using X-rays or neutrons at D20. Therefore, we require high-res neutron PDF to calculate these occupancies and accurately determine the site disorder. We request a total of 2 hours of nPDF access, for the normal Li6PS5Cl material. These are the final experiments required before publication.

Rietveld refinement of a $\text{Li}_6\text{PS}_5\text{Cl}$ cold pressed sample was completed in TOPAS, Fig.1. The ratio of Cl on the 4a/4d Wyckoff sites was 49.2(4a) : 50.8(4d). The inverse is true for S^{2-} .

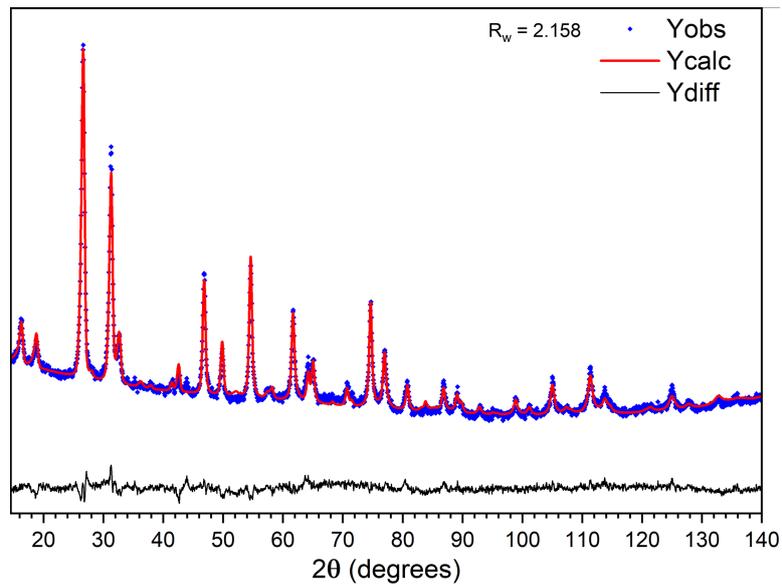


Figure 1: Rietveld refinement of neutron diffraction on cold pressed $\text{Li}_6\text{PS}_5\text{Cl}$ - collected on D2B

This data is being used in conjunction with neutron total scattering data (EASY-1128) in a body of work which is soon to be written up.