Experimental report

22/07/2024

Proposal: EASY-1184 **Council:** 4/2023

Title: Structure determination of a new SrAl2SiO6 polymorph synthesised by glass crystallisation

Research area: Chemistry

This proposal is a new proposal

Main proposer: MICHAEL PITCHER

Experimental team:

Local contacts: Stanislav SAVVIN

Samples: SrAl2SiO6

Instrument	Requested days	Allocated days	From	To
D2B	3	3	23/11/2023	24/11/2023

Abstract:

We have recently explored the SrO-Al2O3-SiO2 phase diagram using a computationally-guided glass-crystallisation strategy to identify new metastable compounds (manuscript currently in review at ChemComm, featuring data from D2B/EASY-745). During this work, we have isolated a new polymorph of the compound SrAl2SiO6. Using laboratory PXRD and selected area electron diffraction, we have indexed this phase to an orthorhombic unit cell of dimensions 9.72 x 16.84 x 18.22A, with C2221 as the most probable space group. This is a large unit cell (~3000 A^3) and we have only lab-quality PXRD data. Nevertheless, by charge-flipping and simulated annealing with rigid bodies we have established approximate positions for the Sr and Al/Si atoms. The oxygen positions (rigid body orientations) remain highly imprecise. We would therefore like to collect a room temperature neutron diffraction pattern to locate the oxide positions unabiguously and complete the structure solution of this phase.

Experimental Report : EASY-1184 (D2B)

The objective of this experiment was to solve the crystal structure of a new metastable oxide with nominal composition $SrAl_2SiO_6$, using *ab initio* methods (simulated annealing and/or charge flipping). The sample was of reasonably large volume (~3g of material). Data were collected initially at 1.6 Å at ambient temperature, and then at 2.4 Å (also at ambient temperature) to provide better resolution (demanded by the large unit cell volume and the low symmetry of the unit cell).

In the structure solution step, an iterative process was followed, with the 2.4 Å D2B data used together with high resolution PXRD data (from the CRISTAL beamline at SOLEIL, collected in December 2023) to generate a structural model that satisfied both scattering techniques. The final Rietveld refined structure was modelled in a hexagonal cell with unit cell volume of 1490 $Å^3$ and 23 independent atomic positions. Later, complementary analysis was carried out using STEM-HAADF methods and MAS-NMR to provide a full picture of the long-range and local structures in this material.

The final structure and composition of the material are not reported here, due to an ongoing patenting process.

Expected outputs:

1 patent (submission expected August 2024)

1 PhD thesis chapter (Euan Duncan, CEMHTI/Université d'Orléans, defence scheduled December 2024)

1 journal article (submission expected December 2024)

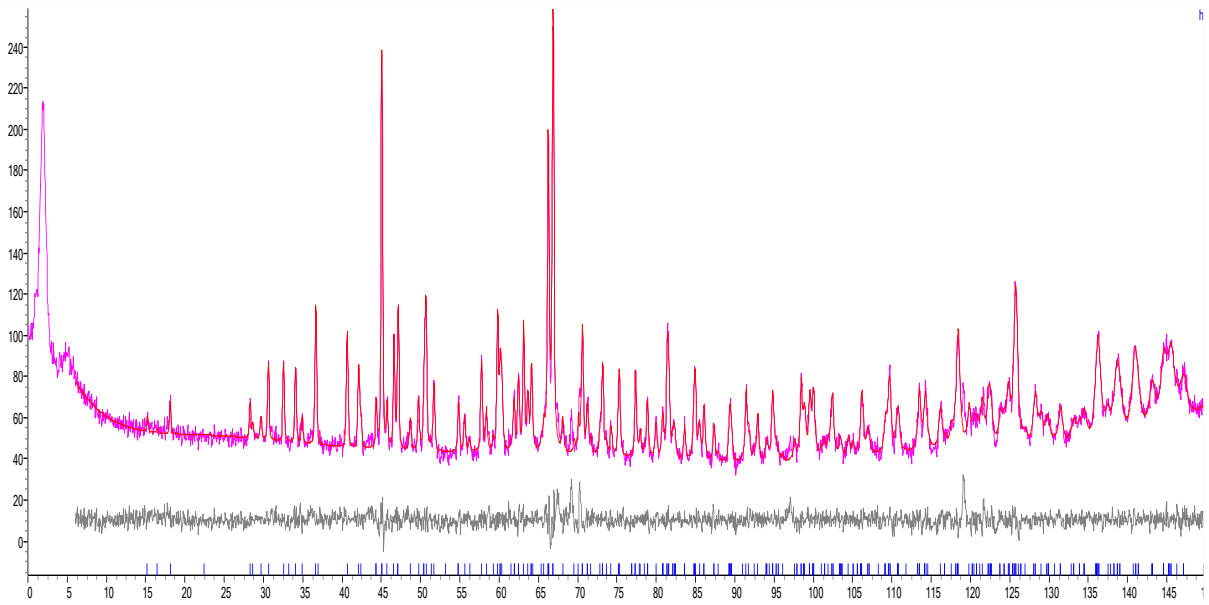

Figure: Diffractogram collected at 2.4 Å incident wavelength, with Le Bail fit corresponding to the final unit cell.