

Disorder By Design: Long- and Short-Range Pyrochlore Ordering

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Carbon-neutral energy generation is being developed in order to combat climate change. Two technologies of current interest, which are related to renewable and nuclear energy respectively, are next-generation oxygen-ion conductors for fuel cells and materials suitable for long-term storage and disposal of radioactive nuclear wastes [1-2].

Pyrochlores of the structure $A_2B_2O_7$ have found immense applications in each of the above areas. Ionic conductors for fuel cells require flexibility and movement in their anionic sublattice, whereas the storage of radioactive nuclear wastes needs a robust lattice from which ions cannot escape. This is a seemingly contradiction in requirements. It is believed that the oxygen vacancies present in the pyrochlore structure allow for the development of short-range disorder, whilst keeping the long-range order intact [3].

The pyrochlore structure can be viewed as a superstructure of the defect-fluorite structure. The defect-fluorite structure (A_2BO_5) consists of a random distribution of cations and oxygen vacancies. However, by choosing A and B with a particular ionic radii ratio, the ordered pyrochlore superstructure may form under ambient conditions. This ordering of oxygen vacancies may be analysed using neutron powder diffraction and used to reason the enhanced properties and applications of pyrochlores [4].

The current work aims to characterise oxygen-vacancy disorder in defect pyrochlores so to enable the rational design of defect pyrochlores that are optimised for specific applications. We have done this by looking at ‘stuffed’ pyrochlores of the form $A_2(B_{2-x}A_x)O_{7-x/2}$ where the smaller B-type cation, in this case Ti^{4+} , is progressively replaced by a larger A-type cation (Tm^{3+}). We wish to determine whether controlling the disorder in the cation sublattice will allow us to tailor-make stuffed pyrochlores targeting specific applications across ionic conductivity, magnetism, photocatalysis and the storage of long-term radioactive waste.

Series of stuffed pyrochlores have been synthesised using conventional solid-state methods and their long-range average structures characterised by Rietveld refinement against combined neutron and synchrotron X-ray diffraction data. The local short-range order has been characterised by Raman spectroscopy and XANES. Other measurements have also been performed regarding their applications, demonstrating a vast improvement in their ionic conductivity at high temperatures.

These results will be presented, along with a judgement as to whether inducing certain types of disorder within the pyrochlore structure can lead to them being purposely engineered for specific applications.

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Level of Expertise

Student

Speakers Gender

Male

Do you wish to take part in the poster slam

Yes

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