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Disorder By Design: Energy, Pyrochlores and the Art of 'Stuffing'

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In accordance with the Paris Climate Agreement, Australia will reduce its carbon emissions to 28% of its 2005 levels by 2030, with 24% of Australia's electricity production to come from renewable sources. Two technologies being developed in Australia to assist with this transition include next-generation oxygen ion conductors and long-term storage for radioactive waste. The former is an attractive candidate, as it does not generate carbon dioxide in hydrogen gas fuel cells, and the latter is particularly important in closing the nuclear fuel cycle as Australia is a major exporter of uranium. However, significant technical issues have arisen in the development of these technologies, such as their lack of efficiency and short equipment lifespans.

Pyrochlores of the structure $A_2B_2O_7$ have found immense applications in each of the above areas: oxygen ion conductors and for radioactive waste storage. However, this appears to be an apparent contradiction in requirements, with one requiring flexibility and movement in its anionic sublattice and the latter needing a robust lattice from which ions cannot escape. It is believed that the oxygen vacancies present in the pyrochlore structure allows for short-range disorder, whilst keeping the long-range order consistent.

In this work, we are concerned with looking at the oxygen-vacancy disorder, and 'tailoring' it to improve the applications of pyrochlores. We have done this by looking at 'stuffed' pyrochlores of the form $A_2(A_{0.67-x}B_{1.33+x})O_{6.67+x/2}$. Increasing the amount of the larger A-type cation that normally occupies the eight-coordinate sites results in some of them occupying the six-coordinate B-site, in a process known as stuffing. It is envisaged that this increase in disorder in the cation sublattice will allow the possibility of engineering these for specific applications.

The poster will focus on two parts: the synthesis and characterisation of the stuffed pyrochlores, and their physical applications.

Two series of eleven stuffed pyrochlores, namely $Yb_2(Yb_{0.67-x}Ti_{1.33+x})O_{6.67+x/2}$ and $Tm_2(Tm_{0.67-x}Ti_{1.33+x})O_{6.67+x/2}$ with x = 0-0.67 have been synthesised using conventional solid-state methods and their long-range average structure characterised by Rietveld Refinement of conventional X-ray diffraction. The local short-range order has been characterised by Raman and infra-red spectroscopy.

Further characterisation was undertaken using soft x-rays and x-ray powder diffraction at the Australian Synchrotron. It is also planned to determine the displacement of oxygen ions using the ECHIDNA Diffractometer at ANSTO. Since synthesising the two series of stuffed pyrochlores, various measurements have also been undertaken regarding their photocatalytic, associated band gaps, ionic conductivity and magnetic properties, yielding promising results.

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-built for their applications.

Speakers Gender

Male

Travel Funding

No

Level of Expertise

Student

Do yo wish to take part in the poster slam

Yes

Primary author(s) : MULLENS, Bryce (University of Sydney)

Co-author(s) : KENNEDY, Brendan (The University of Sydney); ZHANG, Zhaoming (ANSTO)

Presenter(s) : MULLENS, Bryce (University of Sydney)

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