

THEME 1: THE DYNAMIC SOLID

Session 1.8: Minerals and glasses as wasteforms

CONVENED BY:

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Nature has been hiding trace components in minerals and glasses since solids first formed. What can we learn from natural materials? And what can we learn from synthetic minerals and glasses? This session collects contributions on all topics relating to the storage of waste in mineral and glass form.

1.8.11

Structural phase transitions and solid solubilities of $(\text{Nd,La})_2(\text{Zr,Ti})_2\text{O}_7$ phases

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Pyrochlores ($\text{A}_2\text{B}_2\text{O}_6\text{X}$, where $\text{X}=\text{O}, \text{OH}, \text{F}$), are highly refractory minerals, which have been proposed as excellent matrices for the immobilization of Pu and other actinides in High Level Radioactive Waste (HLW).

In order to predict the behaviour of a structure upon incorporation of HLW, it is crucial to understand the extent of solid solution across a range of compositions. It is also necessary to explore the extent of the pyrochlore phase as a function of B site composition (Zr/Ti). Nd is a good analogue for Pu in pyrochlore systems, due to the similar ionic radii of the two species.

With this in mind, synthetic ceramic materials across the two series: $\text{La}_{(2-x)}\text{Nd}_x\text{Zr}_2\text{O}_7$ and $\text{Nd}_2\text{Zr}_{(2-x)}\text{Ti}_x\text{O}_7$ ($x=0, 0.4, 0.8 \dots 2.0$) were produced by solid-state mixed oxide methods. Composition driven changes in the crystal structure were investigated by Electron Microscopy, Electron Microprobe Analysis and Neutron Powder Diffraction. Rietveld analysis was performed on diffraction data to assess changes in the symmetry and unit cell parameter. Partial solid solubility of titanate in zirconate is observed. The titanate end-member displays the previously-noted monoclinic structure, and compositions towards the titante end of the composition range are exsolved into a pyrochlore-titanate two phase mixture. Additionally, the pyrochlore compositions show the possibility of structural disorder towards the disordered fluorite structure.

These materials also have additional potential for application as fast ion conductors, and as such, measurements of the dielectric spectroscopy are reported as a function of composition.

