# Experimental and modelling studies of nuclear materials

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Advanced nuclear fuel cycles offer considerable promise for improvements in safety, performance, actinide management, and provide opportunities for associated methods of energy production (e.g., hydrogen based systems). In general, the proposed reactor systems require new materials capable of performing under the extreme conditions imposed by temperature, radiation fields, and corrosive media. Here we present a comparison of the results obtained from minerals with laboratory observations using ion irradiation methods. The results generally set out the groups of potential actinide host phases in terms of those with intrinsic radiation tolerance due to recovery of damage on picosecond time scales (e.g., fluorite), those with favorable kinetics for longer term damage recovery (e.g., monazite), and many others with unfavorable kinetics. We have also conducted atomistic modelling studies of some of these minerals and other materials of interest. Together with information from the literature, the results are briefly summarized in terms of structure, bonding, and the energetics of defect formation and recovery. We briefly report on the effects of decay of Tc to Ru in rutile.

**Figure 1:** Atomistic simulations using empirical potentials showing the total Frenkel defect formation energies from the lanthanide, titanium, and oxygen atoms in the orthorhombic $Ln_2TiO_5$ compounds.

# The role of Th-U minerals in assessing the performance of nuclear waste forms

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A number of alternative crystalline nuclear waste forms have been proposed for deployment in geological repositories. Materials range from general purpose polyphase waste forms for high level wastes to highly specialized waste forms designed specifically for actinides and certain fission products. Here we review the information available from natural systems relevant to the performance of candidate waste form phases. Observed mineral transformations include (approximately) constant volume chemical changes and breakdown to new phase assemblages, with or without loss of Th and U. Together with laboratory experiments, the geochemical data indicate that zirconolite is a promising host phase for actinides and certain fission products. Pyrochlore has a slightly higher dissolution rate in the laboratory and is generally more susceptible to geochemical alteration. Monazite also has low dissolution rates in laboratory experiments, favorable geochemical behavior, and minimal physical property changes due to retention of crystallinity over geological time scales. Although zircon has been studied extensively, the 17-18 % volume expansion due to alpha decay continues to be a major issue for application as an actinide host phase in nuclear waste forms.

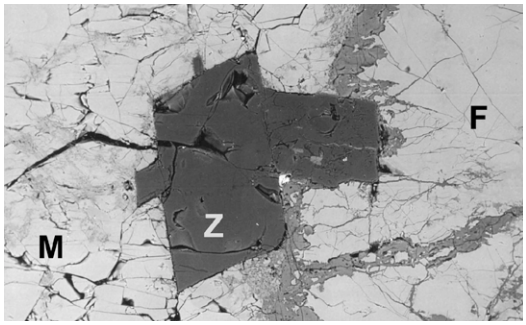


**Figure 1:** Monazite (M), zircon (Z), and fergusonite (F) from the Rutherford #2 pegmatite (t = 289 Ma), Amelia, Virginia. These $ABO_4$ minerals have reacted differently in the presence of late stage granitic pegmatite fluids. Altered areas are darker gray and most prominent in fergusonite. Image width ~ 1 mm.

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