Neutron and resonant x-ray diffraction studies of zirconolite 2M

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Zirconolite (nominally $CaZrTi_2O_7$) is a constituent phase of potential waste forms for the safe immobilisation of actinide wastes. Structural studies of such materials provide important information about cation ordering, lattice parameters, and strain effects, and provide input into the modeling of alpha decay damage and the development of future wasteform designs. A suite of zirconolites based on the replacement of Ti with Nb and Fe has been studied using high resolution neutron diffraction and resonant x-ray diffraction to determine the degree of disorder across the available cation sites. Resonant x-ray diffraction is a unique method which allows the location of certain cations to be determined accurately by taking advantage of the change in scattering power close to an absorption edge (e.g., Nb-K and Zr-K). Using standard x-ray diffraction alone this is not possible and there is little scattering difference between Nb and Zr.

Previous results on these materials have shown that the majority of the $CaZrTi_{2-2x}Nb_xFe_xO_7$ solid-solution series adopt the 2M polytype structure. Raman spectroscopy and measured lattice parameters have shown that the exchange of Ti with Nb+Fe has a non-linear effect on the unit cell dimensions while retaining the 2M polytype. Mössbauer spectroscopy has shown that the Fe preferentially fills the Ti split (Ti2) site. The new results provide a more complete picture of the cation order-disorder problem and are generally consistent with the behaviour of lattice parameters across the series.