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P-31 Nuclear Magnetic Resonance as a Probe of Plutonium Incorporation and Radiation Damage in Phosphates

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Plutonium doped YPO₄ was prepared to test the extent of solid solution and the effect of radiation damage over several years. Y_{0.92} 238Pu_{0.08}PO₄ showed the presence of a Pu substituted xenotime phase and a 238PuP₂O₇ phase by X-ray diffraction (XRD). The 31P magic-angle spinning nuclear magnetic resonance (MASNMR) spectrum showed peaks for PuP₂O₇ and peaks assigned to phosphorus coupled to Pu³⁺ substituted randomly into Y³⁺ sites. A time series of 31P MASNMR and XRD measurements observed the loss of the reflections due to the 238PuP₂O₇ phase in the XRD pattern of the Y_{0.92} 238Pu_{0.08}PO₄ sample within 4 weeks. The MASNMR spectra obtained at longer times in both the Y_{0.92} 238Pu_{0.08}PO₄ and Y_{0.96} 238Pu_{0.04}PO₄ samples showed an additional broadened and shifted 31P resonance assigned to amorphised xenotime with a chemical shift consistent with Q₀ orthophosphate. Thus, the amorphised local structure is assigned to a predominantly Q₀ phosphate arrangement subject to local densification.

Summary

This paper is about understanding the solid solubility of Pu in crystalline phosphates and the effect of its alpha decay on the local structure of the crystal. We use novel actinide enabled magic angle spinning nuclear magnetic resonance to elucidate the nature of bonding in the amorphised phase.

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