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Nuclear Wasteforms Studied with Positron Annihilation Lifetime Spectroscopy (PALS)

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A nuclear wasteform serves to contain radionuclides and enable safe disposal of nuclear waste over long timeframes. In ceramic wasteforms, radionuclides are locked into specific atomic sites within the crystal structure through strong inorganic bonds, effectively preventing their release. Fluoride-pyrochlores are being explored for the immobilisation of actinides from Generation IV molten salt nuclear reactors. However, the mechanisms by which actinides are incorporated into the fluoride-pyrochlore matrix remain poorly understood. Key factors include the actinide valence state and whether the atoms occupy substitutional or interstitial sites within the structure.

When positrons are introduced into well-crystallized ionic solids, they thermalize and annihilate with electrons, typically within 0.1–0.2 ns. However, the presence of cation vacancies or other structural defects creates regions of low electron density where positrons become trapped and/or form positronium, resulting in extended lifetimes of up to 1 ns or more. Consequently, positron annihilation lifetime spectroscopy (PALS) serves as a highly sensitive technique for detecting cation vacancy mechanisms involved in actinide incorporation into ceramic wasteforms.

In this study, a desk-top PALS system with a ^{22}Na source was employed to investigate the potential involvement of a cation vacancy mechanism for the incorporation of actinides in fluoride-pyrochlore structures. Cerium (Ce^{4+}) was used as an actinide surrogate to simulate U^{4+} , Pu^{4+} and Th^{4+} . Ce^{4+} was targeted for substitution at the Ca^{2+} site in the series $(\text{NaCa}_{1-2x}\text{Ce}_x)\text{Nb}_2\text{O}_6\text{F}$, with x values of 0, 0.1, 0.2, and 0.25. Prior to analysing the fluoride-pyrochlore samples, the validity of the PALS technique was confirmed using a model system in which Nb^{5+} was incorporated into rutile (TiO_2) via a charge-compensating vacancy mechanism.

The experimental and data analysis techniques will be outlined, and PALS results for both the rutile and pyrochlore structures will be presented and discussed.

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