

Understanding Processing Induced Defects and Decomposition in PuO₂, PuF₄ and Uranium Alloys

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We summarize here the key roles of X-ray diffraction (XRD) in gaining insights into the processing of three nuclear materials. The objective of one study is to optimize processing of a uranium metal alloy for use in nuclear reactors. The second study involves the influence of thermal annealing on the crystallinity of an approximately 50-year-old sample of PuF₄. The third study investigates changes in PuO₂ powder as a function of synthesis and processing conditions. The common thread between each of these studies is the understanding of defects and decomposition in nuclear materials using powder XRD.

Nuclear material safeguards controls are driving U.S. research reactors and radioisotope production facilities to start using low enriched uranium fuel (<20% ²³⁵U) in place of high enriched fuels (≥20% ²³⁵U). To achieve this lower enrichment target, the uranium density must increase to sustain criticality. Replacing uranium oxide fuels with uranium metal fuels is a way to accomplish this goal. However, the ordinary form of uranium metal that is stable under ambient conditions, α-U, swells and cracks in a reactor. Several uranium metal alloys stabilize the high-temperature phase of uranium (γ-U) down to room temperature and also perform well in nuclear power reactors are not straightforward to produce. The work presented here illustrates the role of XRD in elucidating the results of a time, temperature and transformation study of uranium alloys.

One of the last remaining samples of plutonium tetrafluoride that was produced at the Hanford Site's Plutonium Finishing Plant was recovered and analyzed at the Pacific Northwest National Laboratory. The plutonium was stored in a hermetically sealed container since 1971. Our XRD studies showed that the sample had undergone metamictization through self-induced radiolysis during its storage time. The work presented is an investigation into the metamictization and the effects that thermal annealing has on its reversal through recrystallization.

A processing platform has been set up at the Pacific Northwest National Laboratory to study plutonium oxide (PuO₂) processing chemistry. This platform allows examination of the effects of processing conditions on the final PuO₂ products. This work illustrates the changes in XRD line profiles occurring as a function of processing conditions.