

## **Alpha Radiolysis Studies for Uranium-233 Oxides and Oxyfluorides**

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Recently, the radiolysis of sorbed water and other impurities contained in actinide oxides has been the focus of a number of studies that centered on the establishment of criteria for the safe storage and transport of the oxides. These criteria are designed to prevent the production of large pressures or products (e.g.,  $H_2$  and  $F_2$ ) that could be deleterious to storage containers and that might result in the release of radioactivity. The development of a storage standard for  $^{233}U$  (ref. 1) resulted in the identification of several needed radiolysis studies. Uranium-233 contains a contaminant isotope,  $^{232}U$ , which has a daughter isotope,  $^{208}Tl$ , that emits a 2.6-MeV gamma ray. Hence, these materials have a relatively high alpha activity as well as a large gamma radiation field, both of which complicates their handling.

Experimental studies have been conducted on the gamma radiolysis of sorbed water on uranium oxides and on fluoride impurities in these oxides (primarily on uranium oxyfluorides).<sup>2-4</sup> These experiments have demonstrated that with respect to gamma radiation these materials can be safely stored. To complete the understanding of radiolysis in a  $^{233}U$  oxide system, a similar set of experiments has been conducted for alpha radiolysis. In all of these experiments,  $^{238}U$  was used as a surrogate for the  $^{233}U/^{232}U$ , with radiation supplied from a separate source. A surrogate alpha emitter,  $^{244}Cm$ , was used in place of the  $^{233}U/^{232}U$ . The use of the surrogate offers the advantages of (1) a higher specific alpha activity, which provides for an accelerated experiment and (2) avoiding the use of  $^{233}U$ , which results in a significant personnel dose avoidance and more easily handled materials. The surrogate uranium oxides were prepared by doping natural uranyl nitrate hexahydrate with the desired amount of curium nitrate. The uranium and curium were then coprecipitated as a hydroxide, and this precipitate was subsequently heated to the appropriate temperature to obtain the desired oxide (about 350EC for  $UO_3$  and about 650EC for  $U_3O_8$ ). Samples of the doped  $UO_3$  and  $U_3O_8$  were placed in a container, loaded with a known moisture content, and then sealed. The pressure of the container was continuously monitored. Samples of uranyl fluoride were prepared in a similar manner to study the effects of alpha radiation on fluorides. Gas samples have been periodically withdrawn from the containers, and sample monitoring has continued for more than 1 year. This paper reports on the results to date of the experiments and their relationship to the safe, long-term storage of  $^{233}U$  oxides. These results, combined with those from the earlier gamma radiolysis experiments, form the basis for resolving technical issues regarding the safe, long-term storage of these oxides.

### References

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