

## Nuclear Waste Forms: The Road Not Taken

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During the past forty years, the materials science of nuclear waste forms has focused on the stability and long-term behavior on nuclear waste glasses and used nuclear fuels, mainly  $\text{UO}_2$ . During this same period of time, substantial quantities of Pu, now more than 2,000 metric tones, have accumulated, either still in the used nuclear fuel or chemically separated for weapons or energy applications. This "excess" plutonium, and associated "minor" actinides, such as Np, Am and Cm, offer a new, but seldom pursued, opportunity for the safe geologic disposal of transuranium elements. A variety of materials, with mineral analogues, including oxides, silicates and phosphates, have been investigated because of their high capacity to incorporate actinides, their chemical durability, and in some cases, their resistance to the radiation-induced transformation to the aperiodic state. There has been substantial interest in the use actinide-bearing minerals, such as isometric pyrochlore,  $\text{A}_2\text{B}_2\text{O}_7$  (A= rare earths, actinides; B = Ti, Zr, Sn, Hf), for the immobilization of actinides, particularly plutonium. Systematic studies of rare-earth pyrochlores have led to the discovery that certain compositions (B = Zr, Hf) are stable to very high doses of alpha-decay event damage. Three different processes have been observed: i) radiation-induced amorphization, ii) an order-disorder transformation and iii) phase decomposition. The radiation stability of these compositions is closely related to the structural distortions that occur for specific pyrochlore compositions and the effect of electronic structure on bonding. Based on this very fundamental understanding of the radiation response, durable, actinide waste forms can be designed for specific temperature and radiation dose conditions.