



DoNuTS Technical Meeting

Time: 1600 Wednesday, 11 February 2009

Place: NE Conference Room, 1106 Etcheverry

Speaker: Charles Yeamans, UCB Nuclear Engineering

Subject: Synthesis of Uranium Fluorides from Uranium Dioxide with Ammonium Bifluoride and Ammonolysis of Uranium Fluorides to Uranium Nitrides

Actinide nitrides, in particular uranium mononitride (UN), are being considered as fuel types for advanced reactor systems. Here we demonstrate a low-temperature synthesis route on uranium that could be developed into a commercial fabrication process for UN and mixed actinide nitride fuels. UN was successfully synthesized from UO_2 by first reacting with NH_4HF_2 in a ball mill at 20°C to form tetravalent ammonium uranium fluorides. The fluorination reaction progresses through $\beta\text{-NH}_4\text{UF}_5$, $\delta\text{-(NH}_4)_2\text{UF}_6$, and $\gamma\text{-(NH}_4)_2\text{UF}_6$ intermediate phases before finally forming $(\text{NH}_4)_4\text{UF}_8$, and is also shown to occur between solid uranium dioxide at the surface of the particles and ammonium fluoride vapor. Then the reaction of $(\text{NH}_4)_4\text{UF}_8$ with ammonia gas at 800°C forms an $\alpha\text{-U}_2\text{N}_3/\text{UN}_2$ solid solution product with a composition of $\text{UN}_{1.83}$. The x-ray powder diffraction pattern is the fcc pattern commonly referenced as that of UN_2 and this intermediate product contained 1 wt % UO_2 as an impurity. The final product, UN, was obtained by decomposing the intermediate uranium nitride at 1150°C under argon to produce UN with a 9 wt % UO_2 impurity. Surface area increased by a factor of ten during ammonolysis, consistent with the action of a hydriding agent.