Pre-Treatment and Bulk Separation of Used Fuels with Carbonate-Peroxide Solutions

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ABSTRACT:

The objective of this project is to provide key data for the further development of carbonate-peroxide chemistries as a dissolution and pre-processing method for used uranium-based fuels. High assay, low enriched uranium (HALEU) fuels are expected to reach higher burnup levels than current light water reactor fuels and to be good candidates for reprocessing because of the uranium enrichment remaining at their end-of-life. However, the higher burnup levels will lead to significant increases in radiolysis damage to organic reprocessing solutions, reducing separation efficiencies and producing additional radioactive mixed wastes. Additionally, two components of advanced nuclear fuel cycles that are under current consideration by the DOE and that will have significant effects on the reprocessing of future nuclear fuels are the voloxidation of used fuels (to remove volatile radionuclides for improved management) and the dissolution of used nuclear fuels by means other than HNO_3 (to avoid corrosion issues and toxic NO_x gas production).

It has long been known that carbonate solutions, combined with an oxidizer such as hydrogen peroxide, are effective at dissolving uranium oxides and used nuclear fuels without the hazards associated with HNO_3. Carbonate-peroxide solutions can also facilitate the bulk separation of some fission products from solution, creating a pre-treatment step prior to traditional solvent extraction purification. Ammonium carbonate and hydrogen peroxide have been shown to provide dissolution of uranium oxides while also allowing recycling of the ammonium carbonate. However, there is some concern about incomplete dissolution of uranium species in ammonium-based carbonate-peroxide solutions. This is coupled with the lack of stability of $(NH_4)_2CO_3$, which tends to decompose into $NH_3(g)$ and $CO_2(g)$ at relatively low temperatures, causing undesired, premature reprecipitation of uranium species.

In this work, we propose to develop: (1) a comprehensive understanding of the individual solubility and speciation behavior of epsilon-UO_3 (the form of uranium in used fuel after undergoing advanced voloxidation) and prominent fission products, as well as the system as a whole (i.e., uranium and fission products together), under varying $(NH_4)_2CO_3$, chemical stabilizer, and H_2O_2 conditions; and (2) detailed investigations into the role of the preferred chemical stabilizer and chemical reductants in the dissolution and chemical stability of the system, as the mechanistic action(s) of these important solution additives are not well understood.