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ULTRASOUND-ASSISTED REDUCTIVE DISSOLUTION OF CeO₂ and PuO₂ IN THE PRESENCE OF Ti PARTICLES

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Although UO₂ fuel can be easily dissolved in nitric media, the reprocessing of (U,Pu)O₂ (MOX fuel) may involve the formation of solid residues (“dissolution fines”) despite the more drastic chemical conditions used for its dissolution. PuO₂ can be dissolved in the presence of fluoride F⁻ (complexing dissolution) or silver Ag²⁺ (oxidative dissolution) ions in concentrated nitric media.¹ The existence of a lower oxidation state for plutonium, Pu(III), offers the opportunity to perform the reductive dissolution of PuO₂.¹ This option avoids potential corrosion problems for equipment, and allows softer chemical conditions during treatment. However, investigations related to this topic in nitric acid solutions are scarce and poorly documented in the literature. In addition, dissolution rates are usually quite low and often require solvents or reactants inappropriate for nuclear industry.

In this work, the reductive dissolution of CeO₂ and PuO₂ was investigated under ultrasound irradiation. Preliminary studies were dedicated to CeO₂ (S_{BET}= 54 m².g⁻¹) as an inactive surrogate for PuO₂ because of its comparable redox and crystallographic properties. Sonochemical dissolutions (20 kHz, 0.7 W.mL⁻¹, Ar, 33°C) were carried out in 0.5 M HNO₃ / 0.1 M [N₂H₅][NO₃] / 2 M HCOOH in the presence of Ti particles as a source of in-situ generated reductive species (Ti^{III}). Using these conditions, ceria dissolution is significantly enhanced and is attributed to a depassivation phenomenon of Ti particles driven by ultrasound irradiation. Low frequency ultrasound can indeed facilitate the depassivation of metallic surfaces and dissolution of oxides due to grain fracturation, surface erosion, mass transfer increase, and decrease of diffusion layers.² The injection of fluorides (0.01 M) during sonolysis allows increasing depassivation of Ti which lead to the complete dissolution of the oxide. This work was then transferred to PuO₂ after setting a sonochemical reactor in a glove box (Atalante). The dissolution of PuO₂ (S_{BET}= 5 m².g⁻¹) is also highly enhanced under ultrasound irradiation with however lower rates than what observed for CeO₂. The total dissolution of PuO₂ can be achieved in few minutes with a sufficient amount of fluorides or a larger surface area for PuO₂.

References

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