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Plutonium Futures – The Science 2022

Sonochemical conversion of UO_2 into U(VI) peroxides reveals unexpected morphologies

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Due to the chemical toxicity and level of radioactivity, predicting the behavior of radionuclides in the geosphere or under deep underground storage becomes a subject of major concern. Regarding this problem, it is essential to understand the nature of the species that can potentially form when ground and surface waters enter in contact with actinide-based materials. Hydrogen peroxide (H_2O_2), which is one of the major species formed as a result of water radiolysis, is known to contribute in the corrosion of UO_2 -based matrices through the formation of peroxide precipitates known as studtite and metastudtite. These species can further promote the dissolution and release of radionuclides into the surroundings.[1-3]

Sonochemistry, which deals with the effect of powered ultrasound on chemical reactions, is known to share some similarities with radiolysis particularly through the in-situ generation of hydrogen peroxide resulting from water molecule splitting. Studies dealing with sonochemistry of actinides have been poorly referenced in the literature. Nevertheless, it was reported that a careful selection of the sonochemical parameters allows the dramatic increase of hydrogen peroxide production in mild conditions with the typical observation of additional physical effects (erosion of solids, mass transfer, fragmentation...).[4]

This work focuses the sonolysis of UO_2 in pure water and slightly acidic media under oxygenated atmosphere. After the preparation of well-characterized UO_2 platelets using the oxalic route, their complete conversion into (meta-)studtite $[(\text{UO}_2(\text{O}_2)(\text{H}_2\text{O})_2) \cdot x\text{H}_2\text{O}]$ is observed under ultrasound.[5] The deep investigations on the remaining solutions (UV-Vis abs. spectroscopy and ICP-OES) and solid residues (SEM, XRD and FTIR techniques) allow attributing this behaviour to the sonochemical generation of H_2O_2 . The formation of crystalline studtite structures is observed on the surface of UO_2 platelets with a preservation of the morphology (Figure 1.b) suggesting a complex formation mechanism other than classical dissolution/reprecipitation. Interestingly, under specific sonochemical conditions, centre-holed platelets are observed (Figure 1.c).

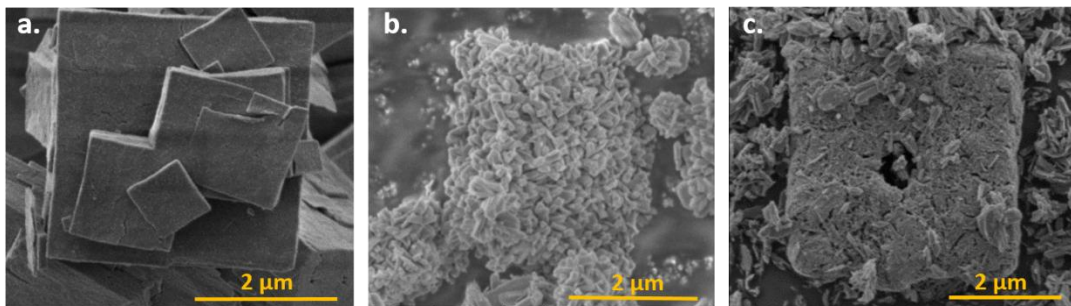


Figure 1: a) SEM image of UO_2 platelets synthesized from oxalate route b) and c) SEM image of the uranyl peroxide observed after sonication of UO_2 in H_2SO_4 $5 \cdot 10^{-4}$ M at 346 kHz ($20 \text{ W} \cdot \text{mL}^{-1}$, 20°C , Ar/O_2 , and 6h)

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