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Ultrasound-assisted conversion of UO_2 into U(VI) peroxides in aqueous solution saturated with Ar/O_2

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In the French nuclear industry, uranium is used as nuclear fuel in the oxide form U(IV)O_2 . After being used in the reactor, spent nuclear fuel is very damaging for organisms and must be safely contained for thousands of years. During storage, the potential intrusion of groundwaters through containment failures could result in the production of radiolytic species such as H_2O_2 that can contribute in the corrosion of UO_2 -based matrices and promote the dissolution and release of radionuclides into the surroundings. Sonochemistry provides an original route to slowly accumulate H_2O_2 in solution through the acoustic cavitation phenomenon. [1-3]

After the formation of well-characterized UO_2 powder with different refractory character using the oxalate route, behavior's powder was studied as a function of the sonochemical parameters (Acoustic frequency, solution composition, gas). This revealed the possible complete conversion of U(IV)O_2 into (meta-)studtite [$(\text{UO}_2(\text{O}_2)(\text{H}_2\text{O})_2-x\text{H}_2\text{O})$] with optimized conditions. [4]

The evolution of the reaction media was followed by ICP/AES, UV-Visible absorption spectroscopy while the solids taken during the experiments were characterized by SEM, XRD and AFM.

The formation of studtite crystal structures was observed on the surface of UO_2 platelets with a preserved morphology of the initial oxide (Figure 1.b). This behavior suggest a complex formation mechanism of uranyl peroxide. Interestingly, specific sonochemical conditions allowed the observation of an unexpected structure (Figure 1.c).

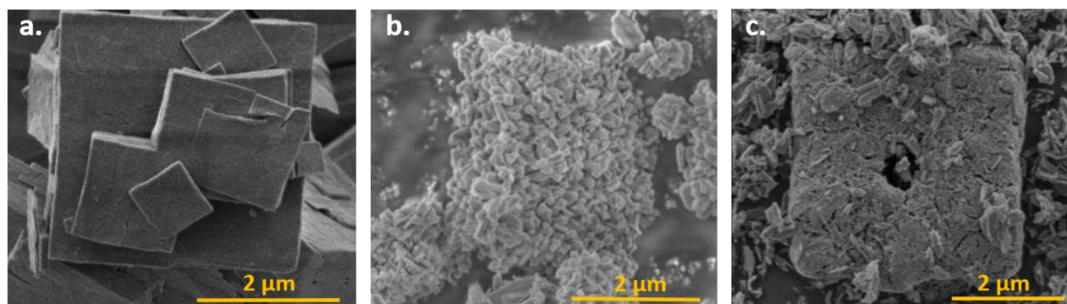


Figure 1: a) SEM image of UO_2 platelets synthesized from oxalate route b) and c) SEM image of the uranyl peroxide observed after sonication of UO_2 in H_2SO_4 $5 \cdot 10^{-4}$ M at 346 kHz ($20 \text{ W} \cdot \text{mL}^{-1}$, 20°C , Ar/O_2 , and 6h)

The evolution of H_2O_2 accumulation rate under these conditions suggests that H_2O_2 contributes in oxidizing U(IV)O_2 into U(VI) and complexing the latter in solution. This work describes an original alternative to study the behaviour of actinides in solution in the presence of H_2O_2 .

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