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

J. Margate [1], M. Virost [1], T. Dumas [2], C. Jégou [3], M. Cot-Auriol [1], T. Chave [1]
S.I. Nikitenko [1]

[1] ICSM, Univ Montpellier, CEA, CNRS, ENSCM, Marcoule, France

[2] CEA, DES, ISEC, DMRC, Univ Montpellier, Marcoule, France

[3] CEA, DES, ISEC, DPME, Univ Montpellier, Marcoule, France

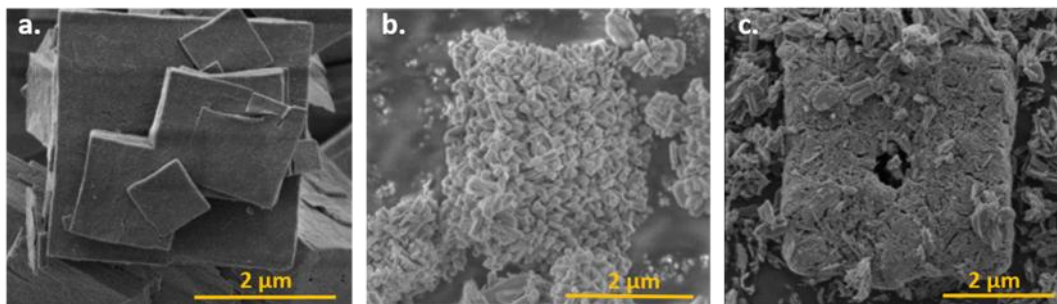
matthieu.virost@cea.fr

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]

This work focuses on the sonolysis of uranium dioxide in pure water and slightly acidic media under oxygenated atmosphere. After the preparation of well-characterized uranium dioxide 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. Detailed investigations on the remaining solutions (UV-Visible absorption spectroscopy and ICP-OES) and solid residues (SEM, XRD, and FTIR techniques) allowed us to attribute this behavior to the sonochemical generation of hydrogen peroxide and physical effect from acoustic cavitation phenomenon.

The formation of crystalline studtite structures was observed on the surface of uranium dioxide platelets with a preservation of the morphology suggesting a complex formation mechanism other than classical dissolution/precipitation. Interestingly, under specific sonochemical conditions, center-holed platelets are observed. A mechanism based on a sonocapillary effect providing the circulation of the reactants within the oxide was proposed leading to a publication and will be discussed during the conference. [5].

Figure 1: a) SEM image of UO_2 platelets synthesized from the 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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