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Ultrasound-assisted Conversion of ThO₂ into a Th peroxy sulfate

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✓ oral □ poster

Nanostructured materials can be defined as materials with a microstructure the characteristic length scale of which is on the order of a few (typically 1-10) nanometers.[1] Nanostructuring of materials is an important subject of study because of their interesting physical and chemical properties which can be optimized according to their size, morphology and structure. The nanoscale particles that compose these materials have a large surface-to-volume ratio and provide a greater amount of active sites compared to other larger materials. Therefore, nanostructured materials may find applications in catalysis, biomedical, synthesis of luminescent materials, etc. Nevertheless, nanostructuring of actinide oxides and their subsequent reactivity are poorly reported in the literature.[2] Recent studies showed that nanostructuring of plutonium oxide appears to play a key role in the sonochemical formation of Pu colloids in aqueous solutions.[3]

In this work, the sonochemical behavior of nanostructured thorium oxides is studied. First, different nanostructured ThO₂ were prepared by oxalic route or in basic conditions in the presence of a polymer before being characterized. The sonochemical reactivity of these oxides was then studied in dilute aqueous solutions at low frequency ultrasound (20 kHz) under Ar/20% O₂ bubbling. The effects of cavitation on solid materials were studied with Raman spectroscopy, AFM, SEM and HR-TEM, while the liquid medium was characterized by UV-vis and ICP-AES spectroscopies. Despite its very refractive behavior, a significant amount of ThO₂ was found to dissolve in sonicated 0.5 M H₂SO₄ solution. In more dilute acidic conditions, a lower dissolution of ThO₂ was observed. In these media, the sonolysis of nanostructured ThO₂ allowed its conversion into a new crystalline phase. The formation of this phase can be attributed to the physical and chemical effects driven by the acoustic cavitation which allows: (i) the oxide particle fragmentation and size reduction, and (ii) the sonochemical generation of H₂O₂. This compound was then prepared in homogeneous solution in the absence of ultrasound. UV-Vis, Raman, FTIR spectroscopies coupled to synchrotron X-Ray Diffraction and EXAFS spectroscopy evidenced the formation of a thorium peroxy sulfate (Fig. 1), poorly referenced in the literature and for which a structure has never been reported.

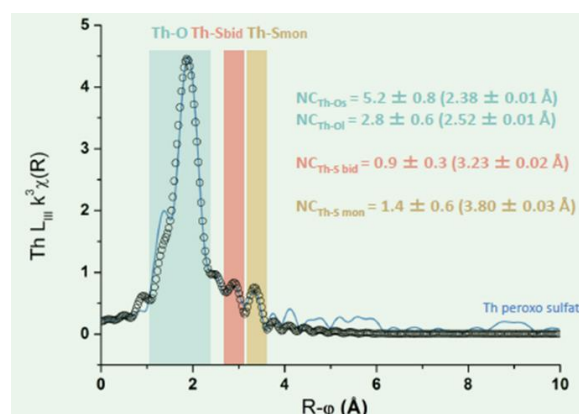


Fig. 1: Fourier Transform of experimental EXAFS spectrum obtained for the Th peroxy sulfate.

[1] H. Gleiter, *Acta Materialia*, 48, 1-29, 2000

[2] V. Tyrpekl, J.-F. Vigier, D. Manara et al., *Journal of Nuclear Materials*, 460, 200-208, 2015

[3] E. Dalodiere, M. Virot, V. Morosini et al., *Scientific Reports*, 7:43514, 1-10, 2017