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Synthesis of nanocrystalline oxide fuel

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High-performance ceramics with nanosized microstructures and functionalized porosities provide the technical base for a large variety of applications in many technologies. This type of microstructure of interest in the nuclear field is due to its appearance at the periphery of Light Water Reactor (LWR) UO₂ fuel at high burn-up. As shown in the past, the fuel spontaneously transforms to a closed porous nanocrystalline structure during irradiation [1,2]. The mechanical properties of this newly formed material are far superior to the original due to the nanostructure.

The aim of these studies is the development of a fuel consisting of nc-UO₂, which, besides the advantages of enhanced plasticity and faster creep characteristic of the nc-state, diminish the pellet-cladding-interaction (PCI)-stresses and cladding failure risks. In addition, the potentiality has to maintain closed porosity under irradiation to safely retain fission gases. The latter is supported by observations of the in-pile behaviour of the nanoscale High-Burnup Structure (HBS) formed in LWR-fuels after prolonged irradiation [3], and by experimental simulation studies with nc-ZrO₂ [4].

First studies at the JRC-ITU were performed to determine the critical ranges of U-concentration and acidity (pH) for nanophase UO_{2+x} precipitation from U(IV)-sulphate solutions [5]. It was shown that the precipitation of UO_{2+x} nanoparticles aggregates occurred at very low U-concentrations (<10⁻⁶ M), but the formation of complex U-sulphate compounds overwhelmed the formation of oxide nanoparticles at higher U-concentrations. Thus, the sulphate route for large-yield production of nanoscale UO_{2+x} material [6] was excluded.

An alternative route also based on the precipitation of colloidal U(IV)-oxide from slightly oversaturated (near equilibrium), electrolytically reduced U(IV)-solutions, has been explored using U-chloride solutions. Positive results yielding 3 to 5 nm UO_{2+x} crystals with the typical fcc structure (see Fig.1) were obtained for the whole range of U-concentrations, especially for the industrial relevant range [U] ≈ 10⁻² M (pH ~ 1.8 to 2.8).

High-temperature X-ray diffraction investigations of the material indicate that the crystal size remains <10 nm for temperatures below 700° C, followed by a sharp increase, culminating with sizes around 80 nm at 1200° C (see Fig. 2,3). Furthermore, an important shift in the lattice parameter has been observed as a function of temperature (Fig. 2), indicating a lattice expansion that extends beyond that caused by thermal effects. The origin of this abnormal (size-dependent) lattice expansion could be the reduction of the material during thermal treatment or the relaxation of binding-forces at the crystal surface. Detailed investigations are in course to elucidate these trends.

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