Grain-scale nanoindentation on mixed uranium-plutonium oxides: a coupled experimental and simulation approach

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Abstract

Uranium-plutonium mixed oxides ((U,Pu)O) are currently the reference fuels for future French Gen-IV Sodium-cooled Fast Nuclear Reactors due to their thermomechanical performance and extensive operating experience. These fuels are manufactured as dense ceramic pellets (_˜95% of theoretical density), with a plutonium molar fraction ranging from 15% to 35% and an oxygen-to-metal ratio (O/M) from 1.96 to 1.99. Under irradiation, (U,Pu)O undergoes significant microstructural and compositional changes due to the high thermal gradient (_˜1500 K over a 3 mm radius) and the accumulation of fission products.

The resulting heterogeneity leads to local variations in mechanical properties such as Young's modulus and Poisson's ratio. These properties are essential inputs for multi-scale fuel performance codes such as GERMINAL (CEA) or TRANSURANUS (JRC), which simulate the fuel behaviour during irradiation (1, 2). Despite their importance, experimental data available on (U,Pu)O remain scarce in the literature, limiting the validation of molecular dynamics results (3). Furthermore, the elastic properties of single crystals are of paramount interest as they are fundamental for establishing reliable correlations between composition, microstructure, and mechanical behaviour. This work aims to address this lack through a combined experimental and simulation approach.

The experimental strategy relies on nanoindentation measurements to investigate the grain-scale mechanical properties on fully characterised (U,Pu)O samples. Due to the radiotoxicity of plutonium, all experiments, including sample preparation and handling, were conducted in glove boxes within the ATALANTE nuclear facility of CEA Marcoule centre. Our nuclearised nanoindentation setup enables local measurements ($< \mu$-scale), providing indentation modulus and hardness data. For each parameter studied (Pu content, oxygen stoichiometry, alpha self irradiation, grain orientation, density), multiple indentations were performed to ensure sufficient quality data for statistical analysis, while invalid measurements near porosities or grain boundaries were discarded. Complementary techniques, including Electron BackScattered Diffraction for grain orientation, Raman spectroscopy and Electron Probe MicroAnalysis for local Pu content and oxygen stoichiometry determination, and optical

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microscopy for porosity analysis, further helped analysing local composition and microstructural influences.

To extract elastic properties from raw force-displacement data, the approach developed by Oliver and Pharr (4), although widely used, does not account for anisotropy or heterogeneities. To interpret these measurements and understand their variability due to crystal anisotropy and surrounding microstructural effects, a finite element digital twin was developed. The latter includes the tip blunting and local microstructural parameters, such as surrounding grains or porosities. The mechanical behaviour of oxide ceramics at the grain scale was modelled using recently developed constitutive laws in a crystal plasticity framework (5).

Measurements on samples with varying plutonium content (0-100%) provide novel data, including PuO elastic constants for grains of known crystallographic orientation. These results fill gaps in the literature, especially at high Pu contents and give some insight into the effect of elastic anisotropy in nanoindentation measurements. Additionally, sensitivity and parametric studies conducted using the numerical model provide valuable guidance for estimating measurement uncertainties, allowing for more accurate interpretation of experimental results and increased confidence in the derived elastic constants values.

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