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Kinetic and microstructural study of a model MOX material

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In the current French nuclear fuel cycle, the plutonium extracted from UO_2 spent fuels is used as a fissile material in MOX (mixed-oxide) fuels. These fuels are manufactured by sintering UO_2 and PuO_2 powder mixtures. This work focuses on the study of the sintering behaviour of such materials through dilatometry. The samples were obtained using a freeze granulation process, which makes it possible to obtain spherical granules with a relatively homogeneous plutonium distribution (both in the as-obtained granules and sintered pellets) [1].

Sintering was performed under a controlled atmosphere of $\text{Ar} + 4\% \text{H}_2 + 1200 \text{ ppm H}_2\text{O}$. Several heat treatments were applied with varying heating rates as well as dwell temperatures and durations. Sintered pellets microstructural characterizations were performed on polished samples and after thermal etching for grain boundary revelation by optical microscopy and scanning electron microscope. Using the grain sizes and densities (obtained by hydrostatic weighing) for the different heat cycles, a sintering map was established. In addition, the diffusion mechanisms occurring during sintering were identified [2]. Densification is thus controlled by volume diffusion and grain growth by the grain boundaries.

The dilatometric data allowed the sintering activation energy to be calculated using two methods: MSC (Master Sintering Curve) and CRH (Constant Rate of Heating). This value of sintering activation energy depends on the atmosphere and above all on the physicochemical characteristics of the material. The obtained value, around 625 kJ/mol, corresponds to what is found in the literature [3] for plutonium volume self-diffusion in $\text{U}_{0.55}\text{Pu}_{0.45}\text{O}_2$ for an oxygen potential of -374 kJ/mol at 1700°C.

Based on these results, a model of the material behaviour during sintering (both during heating and dwell) was established which exhibits a good agreement with the experimental data. This model makes it possible to predict the sintering behaviour of this material for any given thermal treatment (under a given investigated atmosphere) and therefore to determine the thermal cycle to be applied in order to obtain the desired post-sintering density [4] and/or grain size.

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