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#5-254 Experimental Study and Modeling of Cation Diffusion during MOX Sintering: Influence of Manufacturing Parameters

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The only way to optimize the use of plutonium and minor actinides is to close the fuel cycle. This involves recovering uranium and plutonium from spent fuel to fabricate new fuels, such as MOX. The production of (U,Pu)O2±x oxide must comply with the specifications in terms of precise plutonium content, controlled oxygen deviation from stoichiometry, and homogeneity of the distribution of plutonium. This homogenization helps prevent undesirable effects in the fuel during irradiation, such as generation of high burn-up zones as well as formation of insoluble phases during recycling. In this context, the main objective of this study is to examine the diffusion of cations depending on the fuel fabrication conditions during sintering. The experimental method used in this research work involves placing two diffusion half-couples in contact: one with composition UO_2 and the other with composition $U0.77Pu0.23O_2$. These samples are then subjected to heat treatment in a furnace under different oxygen potentials, at various temperatures, and for different dwell times. The calculation of diffusion coefficients in the bulk and at grain boundaries for the different conditions was performed using microprobe X-Ray WDS characterization maps. EBSD characterizations were also conducted to assess the effect of grain orientation on diffusion. In the second part of this work, we chose to cut thin films from the samples prepared in the afore-mentioned experiments in order to better visualize diffusion of Pu by heating the slices and observing the evolution of their concentration in the bulk and at grain boundaries using transmission electron microscopy (TEM). This experimentation on the MOX material allows, for the first time, the acquisition of more precise and consistent data on Pu diffusion. Our results show that Pu diffusion is much faster at grain boundaries than in the bulk in the samples under reducing conditions. Alongside this experimental work, we developed a numerical simulation model for diffusion at grain boundaries, coupled with bulk diffusion. This model was brought into play to simulate the experimental profiles, allowing us to adjust the diffusion coefficients in a much more advantageous way than using traditional methods alone. Moreover, the simulation in this work also uses the data obtained from the experimental section for the different conditions to simulate the time required to achieve complete homogenization of Pu in the pellets for each of these conditions. This helps determine the optimal conditions for fuel homogenization, thereby contributing to increasing the amount of recycled spent fuel.

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