INTERDIFFUSION STUDIES ON THE UO_2-GD_2O_3 SYSTEM

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Homogeneous mixtures of gadolinium oxide with uranium dioxide nuclear fuel are used as burnable poison in modern nuclear power reactors. The incorporation of gadolinium into the nuclear fuel is important because of radioactivity compensation and power distribution adjustment. It's difficult to produce sintered pellets with the required minimum density from UO_2-Gd_2O_3 dry mechanical powder blending because of the occurrence of a blockage during sintering process. This mechanism occurs because of the low diffusivity of phases rich in gadolinium, which actuate as a diffusion barrier. This investigation aims at studying the interdiffusion process for the $(U,Gd)O_2$ system by determining the gadolinium profile concentration in a couple of sintered UO_2/Gd_2O_3 The shape of the concentration profile was qualitatively determined by EDS analysis.

The chemical evolution between pure UO_2 towards Gd_2O_3 phase was verified through WDS quantitative analysis. From the interpenetration curves, the interdiffusion coefficient was determined through graphic integration applying the Matano-Boltzmann method. (FIG.1) shows UO_2/Gd_2O_3 interface. Uranium and gadolinium concentrations were determined along the reference line, and indicate interpenetration of approximately 16 µm after sintering for 3 hours at 1650°C. The gadolinium penetration into the UO_2 phase is sensibly higher than uranium penetration into Gd_2O_3 phase. (FIG.2) presents experimental results of quantitative gadolinium concentration analysis performed in points spaced 0.5 µm along a normal line to the interface UO_2/Gd_2O_3 line. The interdiffusion coefficient in the UO_2-Gd_2O_3 system was calculated as a function of gadolinium molar fraction. (FIG.3) indicates both the sudden decrease in the interdiffusion coefficient for gadolinium concentrations above 50 mol% and the increase in the interdiffusion coefficient value for gadolinium concentrations of about 80 mol%. These results agree with results presented in (FIG.4), which shows the densities after sintering UO_2-Gd_2O_3 fuel pellets, as a function of gadolinium concentration. UO_2-Gd_2O_3 sinterability decreases drastically starting from the composition $(U_{0.5}Gd_{0.5})O_2$ due to a sudden decrease of the system interdiffusion coefficient. Experimental evidences indicated the existence of phases in the $(U,Gd)O_2$ system with a different structure from the UO_2 fluorite type. These new phases occur for Gd molar fractions higher than 0.5 and coincide with the decrease of both the density after sintering and the interdiffusion coefficient. The diffusion barrier mechanism seems to explain the sintering behavior of UO_2-Gd_2O_3 fuel. This work aims at investigating the blocking mechanism during UO_2-Gd_2O_3 fuel sintering fabricated by dry mechanical blending method.

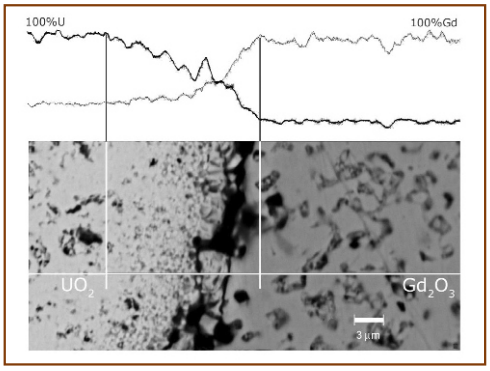

FIGURE 1 - Scanning electron micrograph with UO2/Gd2O3 interface.

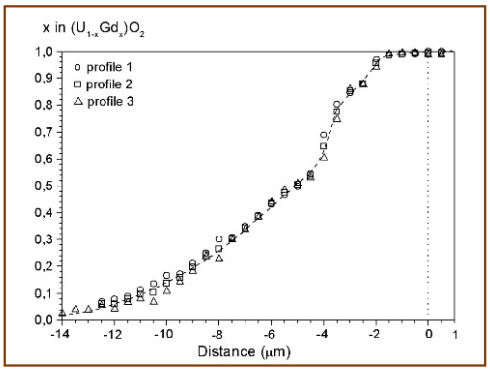

FIGURE 2 - Concentration profile across UO2/Gd2O3 interface.

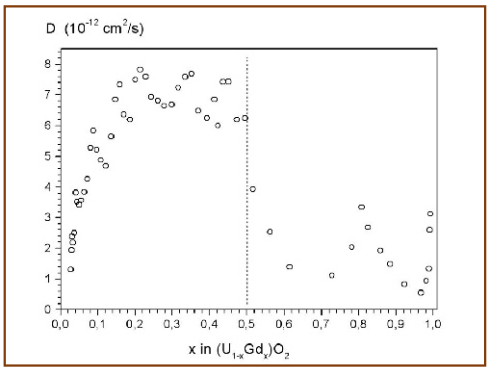

FIGURE 3 - Interdiffusion coefficient as a function of gadolinium molar fraction.

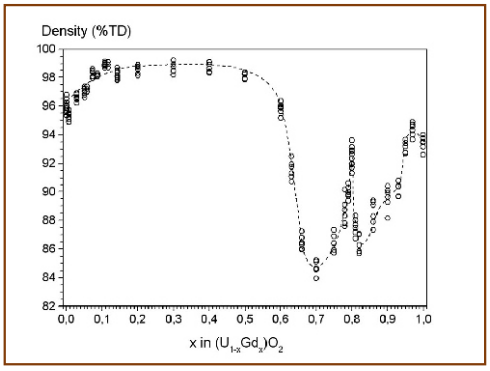

FIGURE 4 - Effect of the gadolinium concentration on (U,Gd)O2 pellets sintered density.