

Evolution of $\text{UO}_2\text{-Gd}_2\text{O}_3$ inter-diffusion layer at the sintering temperature of UO_2 pellet

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1. Introduction

Mixed oxide pellets of $\text{UO}_2\text{-Gd}_2\text{O}_3$ are typical burnable absorbers used as nuclear fuel for LWR. The role of Gd is to regulate the reactivity of reactor core by absorbing excess neutrons in the early stage of fuel operation. In particular, whereas natural Gd has a large neutron absorption cross-sectional, secondary isotopes show low cross-section, which benefit can minimize the reactivity penalty at end-of-cycle.

Recently, the efforts are underway in nuclear industry to increase uranium enrichment higher than 5% to achieve long-fuel cycle length. In addition, the soluble Boron-free core is a major design requirement for advanced light water reactors such as SMR. Accordingly, the total amount of Gd introduced into nuclear fuel and/or core is expected to increase further.

There are several technical issues in increasing the amount of Gd in $\text{UO}_2\text{-Gd}_2\text{O}_3$ nuclear fuel system. Due to the deleterious effect of Gd_2O_3 on UO_2 sintering behavior, a strictly controlled manufacturing process is required to obtain homogeneous and high density pellets. Regarding fuel performances, thermo-physical properties of UO_2 fuel pellet such as fuel melting temperature, thermal conductivity and mechanical strength are largely decreased by the addition of Gd. In order to resolve the manufacturing and performance issues of the homogeneous $\text{UO}_2\text{-Gd}_2\text{O}_3$ system, composite fuel designs, in which spherical, disk or cylindrical types of Gd_2O_3 are heterogeneously distributed in UO_2 matrix, have been proposed. The $\text{UO}_2\text{-Gd}_2\text{O}_3$ system is known to form interaction layers at an elevated temperature. Therefore, to evaluate the fuel performance of the composite fuel, it would be essential to know the interaction behavior at the interface during the sintering and the properties of these reaction layers [1-4].

This paper presents the interface morphology between UO_2 and Gd_2O_3 after the sintering at 1730°C. Formation of interaction layers and inter-diffusion profiles are investigated and compared with literature.

2. Experimental

Three kinds of Gd_2O_3 disc were prepared by heat-treating the Gd_2O_3 green samples at temperatures of 1200°C, 1300°C and 1350°C, respectively. Those Gd_2O_3 disc embedded UO_2 green pellets were prepared using uniaxial press. The sintered pellets were

fabricated by sintering the green pellets at 1730°C for 6h under dry H_2 gas atmosphere.

The sintered pellets were longitudinally sectioned and polished to investigate the $\text{UO}_2\text{-Gd}_2\text{O}_3$ interface. The interface morphology between UO_2 and Gd_2O_3 after the sintering was investigated using SEM and EDS.

3. Results

Fig. 1 shows the $\text{UO}_2\text{-Gd}_2\text{O}_3$ interface structure observed by back scattered electron microscopy. The strip-shaped dark region is the vertical cross section of Gd_2O_3 disk and the bright region is UO_2 matrix.

The micrograph reveals the appearance of voids and microcracks at the edge of disk, which are extending to the pellet surface, as well as the interconnected porous zones around the disk. The density of Gd_2O_3 in the green pellet is much higher than that of UO_2 matrix because the Gd_2O_3 was pre-heat-treated before sintering. The prematurely dense region of Gd_2O_3 can hinder the densification of UO_2 matrix. The differential densification between two constituents will generate voids and mechanical stresses which are the cause of the cracks. Moreover, the differential thermal expansions during cooling can also contribute to the formation of microcracks in the matrix.

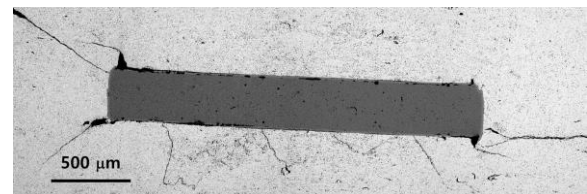


Fig. 1. Back scattered SEM image of the polished surface of sample pellet.

Figs. 2(a)-(c) show the enlarged back scattered SEM images obtained at the interfaces of sintered pellets. It is obvious that the interaction layer was most widely developed in a pellet containing the Gd_2O_3 disc pre-heated at 1200°C. In contrast, despite the same sintering conditions, the width of interaction layer of the pellets including the Gd_2O_3 disk pre-heated at 1300°C and 1350°C was similarly reduced. The density of pre-heated Gd_2O_3 disk was increased with increasing annealing temperature and was in the range between 71~74%TD. Since the initial density difference between the Gd_2O_3 discs was small and the reaction width is not linearly proportional to the initial density, it seems

unlikely that the initial density has affected the diffusion behavior greatly.

Another possibility would be a difference in a crystal structure. It is known that the Gd_2O_3 transforms from cubic to monoclinic structure at $1250^\circ C$. It is expected that the residual monoclinic phase in the Gd_2O_3 disk pre-heated at $1300^\circ C$ and $1350^\circ C$ may delay the chemical interaction with UO_2 matrix. Further study is necessary to understand this behavior.

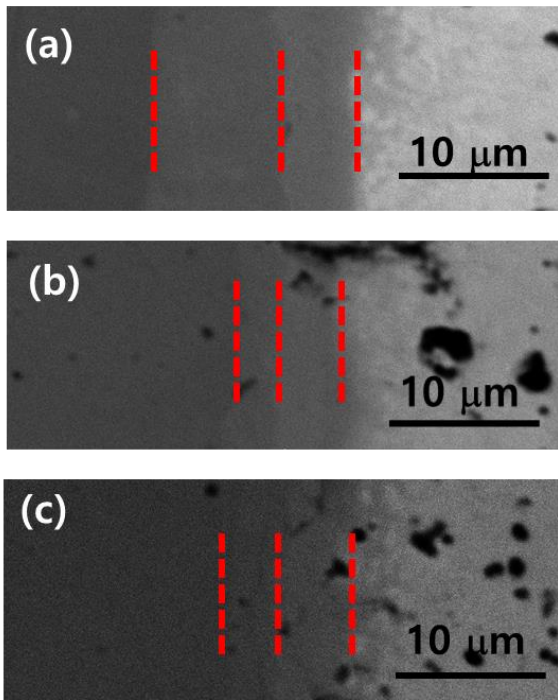


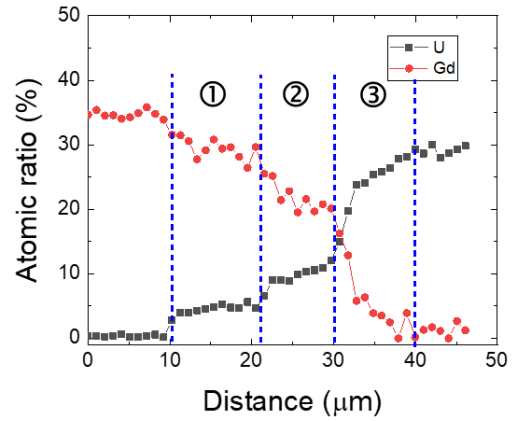
Fig. 2. Back scattered SEM image of the polished surface of sample pellet. The Gd_2O_3 disk was preheated at (a) $1200^\circ C$, (b) $1300^\circ C$, and (c) $1350^\circ C$, respectively.

Fig. 3 shows the EDS elemental line scanning data obtained at interface region. The cation concentration profiles indicated interaction layer of approximately $30\sim 40\ \mu m$ after the sintering for 6h at $1730^\circ C$. Three different inter-diffusion layers have been identified. Adjacent to the Gd_2O_3 , two kinds of compound-like reaction layers are formed. The Gd/U ratios of two phases were about 6 and 2, respectively. According to the phase diagram of $UO_2-Gd_2O_3$, the first phase is probably UGd_6O_{11} . The second phase, which observed in this experiment for the first time, is believed to have a composition of UGd_2O_5 . The third phase, consisting mainly U-rich phases surrounded by Gd-rich phase, corresponds to the solid solution, the concentration of Gd decreasing steadily down to zero.

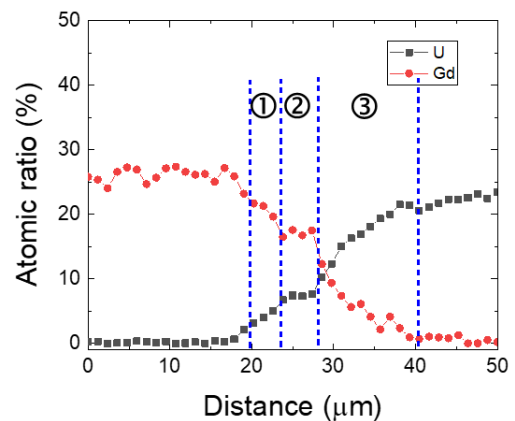
3. Conclusions

The inter-diffusion behavior between UO_2 and Gd_2O_3 at the sintering environment of UO_2 pellet has been investigated. The new compound, which is expected to

have a composition of UGd_2O_5 was observed for the first time. The experimental results also suggested the possibility that residual monoclinic or cubic phases of Gd_2O_3 may alternate the inter-diffusion between UO_2 and Gd_2O_3 .



(a)



(b)

Fig. 3. Cation concentration profiles across the interface The Gd_2O_3 disk was preheated at (a) $1200^\circ C$ and (b) $1300^\circ C$, respectively.

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