Study on the interface between UO_2 and Zr at the moment of melting

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1. Introduction

Researches on the interaction between uranium oxide and zirconium are interesting for the improved knowledge of chemical situation at the interface of UO_2 pellets and Zircaloy cladding in a fuel rod [1-7]. In particular, at high temperature the chemical property of uranium oxide is changed with zirconium due to their interaction, leading to the melting of uranium oxide at a lower temperature than its melting point [8]. Through the observation of the interface at the moment of melting, we can infer the melting process of uranium oxide with zirconium. To perform this study, we fabricated a sample composed of UO_2 and Zr in a pellet. We found an appropriate heating temperature and time (about 2000 ºC for 2 minutes) to examine the interface between the unmelted UO_2 and the dissolved UO_2 with Zr in which UO_2 is incompletely melted. The investigation is conducted using a Scanning Electron Microscopy (SEM) equipped with an Energy-dispersive X-ray spectroscopy (EDX).

2. Experimental details

A UO_2-Zr pellet (ϕ : 6.35 mm) was fabricated, in which a half of the pellet was made of UO_2 powder and the other consisted of Zr powder (Aldrich). The heating of the sample was performed in a LECO EF-400 electrode impulse furnace at approximately 2000 ºC for 2 minutes using a high temperature graphite crucible 782-720 in an Ar atmosphere. The temperature of the sample was measured with a Raytek MR1SCCF pyrometer. SEM experiments were executed using JEOL JSM-6610LV installed with Oxford Instruments X-Max EDX.

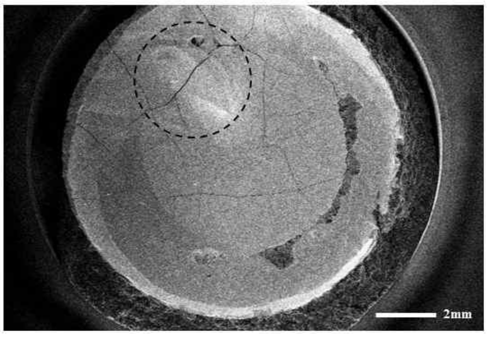

Fig. 1. SEM image after heating of a UO_2-Zr sample at 2000 ºC for 2 minutes. The unmelted UO_2 is displayed in the dotted circle.

3. Results

An SEM image (Figure 1) after the heating of a UO_2-Zr sample at 2000 ºC for 2 minutes is exhibited, in which the undissolved UO_2 is definitely shown with the molten mixture on the graphite crucible. Even if the melting point of UO_2 is known to be 2850 ºC [8], the component of UO_2 in the UO_2-Zr sample begins to melt with Zr at 2000 ºC in our experiment. Moreover, through the analysis of the SEM images, we have found that the melting of UO_2 sequentially occurs at the interface between UO_2 and Zr.

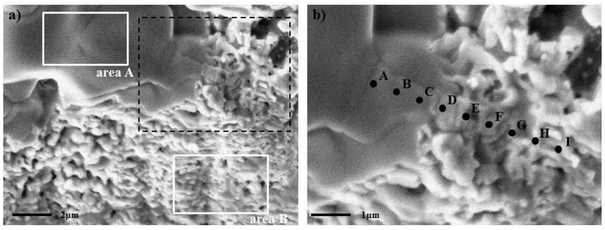

Fig. 2. (a) SEM image obtained from the interface between UO_2 and Zr after the annealing of the UO_2-Zr pellet at 2000 ºC for 2 minutes. (b) Magnified SEM image at a dotted square shown in (a). Nine points marked as A – I describe the positions of EDX measurement.

Table I: EDX results at points A – I shown in Figure 2b.

	Uranium (atomic%)	Zirconium (atomic%)	Oxygen (atomic%)
A	32.6	0	67.4
B	34.0	0	66.0
C	29.2	3.8	67.0
D	25.3	3.3	71.4
E	40.0	7.0	53.0
F	25.4	3.7	70.9
G	23.4	4.5	72.1
H	26.8	4.9	68.3
I	27.8	5.6	66.6

Figure 2a displays the SEM image acquired from the interface of UO_2 and Zr at the initial state of UO_2 melting. Through EDX inspections at areas A and B in Figure 2a, the relative atomic percentage of uranium, zirconium, and oxygen was measured as 29, 0, and 71 as well as 20, 11, 69, respectively. This means that areas A and B were composed of the unmelted UO_2 and the molten product, respectively. To investigate the systematic elemental change at the interface, we have conducted EDX measurements at nine points (represented by A – I in Figure 2b). The relative atomic ratio is presented in Table 1. On the basis of these EDX data, we could obtain the following information. First, the element of zirconium initially appears at point C, and its relative ratio successively increases from spots C through I. Second, the relative ratio of oxygen is largely unchanged except for point E, indicating that the stoichiometry of oxygen is similarly maintained at regions of the unmelted UO_2 and the molten mixture. Third, at the point E the portions of uranium and zirconium increase whereas that of oxygen decreases. These results deviate from the tendency of relative element ratio presented in Table 1. Since previous study has revealed that there are the regions of oxygen-stabilized α-Zr(O) phase and metallic (U,Zr) alloy on the interface between uranium oxide and zirconium after the heating at high temperature [8], we have guessed that the point E corresponds to these regions, resulting in the reduction of the relative oxygen percentage.

3. Conclusions

We studied a UO_2-Zr system after the heating at 2000 ºC for 2 minutes using SEM installed with EDX. Based on the analysis of SEM images and EDX data, we suggest that 1) UO_2 is melted at a lower temperature than its melting temperature, 2) the melting of UO_2 sequentially arises at the interface between UO_2 and Zr, 3) the element of zirconium initially appears at the interface (point C in Figure 2b), and 4) the stoichiometry of oxygen is similarly maintained at locations of the unmelted UO_2 and the molten product except for point E.

We predict that the research into the observation of interface at the initial state of melting will contribute to an improvement of knowledge in a system of UO_2 pellets and Zircaloy cladding.

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