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Temperature-dependent changes at the interface between UO_2 and Zr

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1. Introduction

Studies on the interaction between uranium dioxide and zirconium are intensively performed to obtain information on the interface between a Zircaloy cladding and uranium dioxide pellets in a fuel rod [1-5]. Specially, the chemical properties of uranium dioxide at high temperature are changed with zirconium through their interaction [6]. In other words, the uranium dioxide is melted below its melting point with zirconium. To understand this phenomenon, we conducted temperature-dependent experiments, in which the interfacial changes of the UO_2-Zr samples are observed at various temperatures using a scanning electron microscope (SEM) equipped with an energy-dispersive X-ray spectroscope (EDX).

2. Experimental details

UO_2 and Zr pellets having a diameter of 6.35 mm were manufactured using uranium dioxide powder and zirconium powder (Aldrich), respectively.

The heating of the UO_2 and Zr samples was conducted in a LECO EF-400 electrode impulse furnace at high temperature for 2 min using a high-temperature graphite crucible 782-720 in an Ar atmosphere. The temperature was evaluated with a two-color infrared pyrometer (Raytek, MR1SCCF) above 1000 ºC and an infrared pyrometer (IMPAC Infrared GmbH, Infratherm IS 8 plus) below 1000 ºC.

A Zr pellet was located on a UO_2 pellet, and they were coincidently annealed in a high-temperature graphite crucible (LECO 782-720) in an Ar atmosphere. After the heating, the bottom of the Zr sample and the top of the UO_2 pellet were measured to examine the interface of the UO_2 and Zr pellets. To minimize the exposure to air, the heated samples were rapidly transported to a vacuum chamber.

An SEM experiment was conducted using a JEOL JSM-6610LV with an Oxford Instruments EDS.

3. Results

Figure 1 shows SEM images of the UO_2 pellets after annealing with Zr pellets at a variety of temperatures. In Figure 1a, the grain size of a UO_2 sample at 875 ºC with a Zr pellet is similar with that of a raw UO_2 pellet at room temperature. After annealing with the Zr samples at 1890 and 2040 ºC, the grain size of the UO_2 pellets increases, as shown in Figure 1b-c. In particular, we found a large number of pores at the boundaries of the UO_2 grains after heating with a Zr pellet at 2230 ºC.

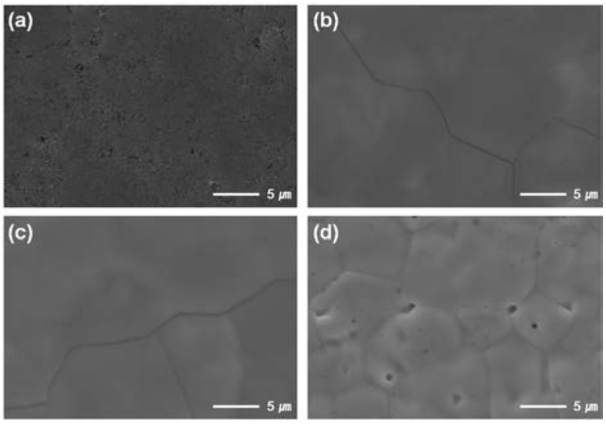

Fig. 1. SEM images (×4,000) of the UO_2 pellets after heating with the Zr pellets at (a) 875, (b) 1890, (c) 2040, and (d) 2230 ºC.

Figure 2 shows SEM images of the Zr pellets after annealing with UO_2 pellets at same temperatures shown in Figure 1. The SEM images in Figure 2a-b are analogous to those obtained from a raw Zr pellet at room temperature. Moreover, many small products with a size of approximate 1 μm are shown in Figure 2b. The grain size of the Zr pellets increases after the heating with UO_2 at 1890 and 2230 ºC, in which the morphologies of Zr pellets seem to be melted.

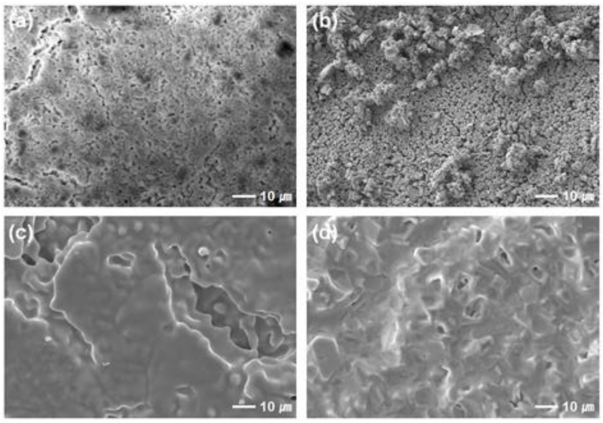

Fig. 2. SEM images (×1,000) of the Zr pellets after the annealing with the UO_2 pellets at (a) 875, (b) 1270, (c) 1890, and (d) 2230 ºC.

Finally, we carried out an experiment to investigate the molten product of a UO_2-Zr sample when they are fully melted. Through an SEM measurement with EDS elemental mapping, the distribution of uranium and zirconium elements in the molten product is inspected, as shown in Figure 3. We found that the molten mixture has a heterogeneous elemental topography. There are two different kinds of areas. First, the dark color of uranium and bright protrusions of zirconium exist together. We suggest that this region is caused by α-Zr(O) and ZrO_2 features. Second, an area consisting of evenly dispersed uranium and zirconium elements is shown. We propose that this area results from the (U,Zr)O_2 configuration.

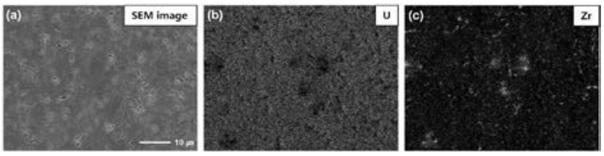

Fig. 3. (a) SEM image and EDS results of (b) uranium and (c) zirconium elements acquired from identical regions of the molten product. The brighter the part is, the richer the element is in (b) and (c).

4. Conclusions

We studied the UO_2-Zr system after heating at high temperature for 2 min using an SEM installed with an EDX. Based on an analysis of the SEM images, we can see that the size of the grains increases when the heating is applied to a UO_2-Zr sample. Furthermore, we propose that the molten product of the UO_2-Zr sample is composed of two distinct regions comprised of a heterogeneous morphology.

We anticipate that observations of an interface at various temperatures will contribute to an improvement in our knowledge in a system of UO_2 pellets and Zircaloy cladding in a fuel rod.

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REFERENCES

[1] P. Hofmann, C. Politis, The kinetics of the uranium dioxide-Zircaloy reactions at high temperature, Journal of Nuclear Materials, Vol.87, p. 375, 1979.
[2] D. R. Olander, The UO_2-Zircaloy chemical interaction, Journal of Nuclear Materials, Vol.115, p. 271, 1983.
[3] M. Miyake, M. Katsura, S. Yamanaka, A study of the reaction between UO_2 and Zr, Journal of Nuclear Materials, Vol.154, p. 123, 1988.
[4] P. Piluso, G. Trillon, C. Journeau, The UO_2-ZrO_2 system at high temperature (T > 2000 K): importance of the meta-stable phases under severe accident conditions, Journal of Nuclear Materials, Vol.344, p. 259, 2005.
[5] K.-T. Kim, UO_2/Zry-4 chemical interaction layers for intact and leak PWR fuel rods, Journal of Nuclear Materials, Vol.404, p. 128, 2010.
[6] P. Hofmann, Current knowledge on core degradation phenomena, a review, Journal of Nuclear Materials, Vol.270, p. 194, 1999.