

Variation in the surface morphology of polycrystalline UO_2 powder induced by helium precipitation

Hiroyuki Serizawa^{a,*}, Junji Matsunaga^b, Noriko Shirasu^a, Kuniyoshi Nakajima^a, Shinji Kashibe^b, Yoshiyuki Kaji^a

^a Nuclear Science and Engineering Directorate, Japan Atomic Energy Agency (JAEA), Tokai Research and Development Center, (Nuclear Science Research Institute), 2-4 Shirane Shirakata, Tokai-mura, Naka-gun, Ibaraki-ken 319-1195, Japan

^b Research Dept., Fuel Group, Nippon Nuclear Fuel Development Co., Ltd., 2163 Narita-cho, Oarai-machi, Higashi-ibaraki-gun, Ibaraki-ken 311-1313, Japan

ARTICLE INFO

Article history:

Received 1 July 2013

Received in revised form 30 July 2013

Accepted 31 July 2013

Available online 22 August 2013

Keywords:

Oxide

Surfaces

Helium

Blister

Image crystal

ABSTRACT

This report addresses the precipitation of helium in polycrystalline UO_2 , which deforms the morphology of the grains and their surfaces. The formation of intragranular gas bubbles by the diffused helium can adversely affect the sintering process of ceramic-type nuclear fuel pellets. Helium was injected into pulverized UO_2 particles at 1473 K by hot isostatic pressing (HIP). The specific surface area measured by volumetric gas adsorption instrument implied that small pores should exist on the as-helium-treated sample surface. Field-emission scanning electron microscopy observations showed that numerous shallow basins (approximately 500 nm in radius) with hexagonal fringe were formed on the surface. The basin resembles a ruptured blister whose lid has been forced open. SEM observations showed a uniform polygonal-shaped section of the gas bubble on the fracture surface; this implies that precipitated helium forms a negative crystal in the grain. These interesting results suggest the possibility that the rupture of the negative crystal formed in the vicinity of the surface is related to the formation of the basin with a hexagonal fringe.

© 2013 The Ceramic Society of Japan and the Korean Ceramic Society. Production and hosting by Elsevier B.V. All rights reserved.

1. Introduction

The management of radioactive waste accumulated through the use of nuclear power plants is a matter of public concern because of the long-term hazard it poses with respect to the environment. The transmutation of minor actinides (MA: Np, Am, Cm) into short-lived elements has been discussed as an option to reduce the high radiotoxicity of these wastes in the future nuclear fuel cycle. Numerous concepts related to fuel and reactor types have been devised, including a combination of MA-MOX fuel with a fast breeder reactor (FBR) [1]. The addition of MA elements to MOX will change the physicochemical properties of the matrix, i.e., specific heat, thermal conductivity, creep rate, elastic moduli,

etc., which are generally relevant parameters in the design of new fuel. However, to develop MA-MOX fuel, we must also consider a new problem in the fuel manufacturing process that arises from helium produced by the α -decay of MA. The precipitated helium in raw powder diffuses from the grain interiors to the surface during sintering to form a gas bubble in a region of grain boundaries.

The solubility and diffusivity of helium in UO_2 are higher and more rapid compared with those of the fission product (FP) gas elements [2,3]. Because of their large atomic radii, xenon and krypton are essentially insoluble, whereas helium dissolves in UO_2 up to concentrations as high as $6.7 \times 10^{-4} \text{ cm}^3 \text{ (STP)/(g atm)}$ at 1473 K [2]. First-principle modeling conducted by Yun implies that the dominant diffusion path of helium is to hop through a single vacancy [4], whereas xenon is likely to diffuse through the processes of vacancy clustering and de-clustering [5]. According to Ronchi and Hiernaut, the diffusion coefficient of helium in UO_2 is almost four times greater than that of the FP gas element [3]. Similar results are presented in another report [6]. Our recent work also shows that the diffusion coefficient of helium is much greater than that of the FP gas element [7], as suggested by Ronchi and Hiernaut [3]. Because the solubility and diffusion mechanism are closely related to the precipitation behavior of the noble gas element, deformation of the matrix texture by the growth of the helium gas bubble should be quite different from that induced by the formation of a xenon or krypton gas bubble [8].

* Corresponding author. Tel.: +81 29 282 6380; fax: +81 29 282 6122.

E-mail address: serizawa.hiroyuki@jaea.go.jp (H. Serizawa).

Peer review under responsibility of The Ceramic Society of Japan and the Korean Ceramic Society.



A series of the studies conducted by Sattonnay et al. clarified that the growth conditions of gas bubbles in UO_2 vary strongly depending on the implanted species; the precipitation of helium or xenon occurs above a threshold temperature, T_c [9]. According to them, xenon implanted to UO_2 nucleates at a lower temperature compared to helium implanted under the same conditions. They hypothesized that the difference in nucleation temperature is likely due to the differences in both severity of radiation damage and diffusion mechanism involved in each case. Their hypothesis is noteworthy with respect to the behavior of helium in nuclear fuel, because the release of helium produced by α -decay might form an intra- and inter-granular bubble and thereby induce swelling of the pellet. However, whether helium in UO_2 under irradiation behaves in this manner is unclear because the temperature region in which the authors performed their experiments was considerably low. To clarify the helium behavior in nuclear fuel loaded in a nuclear plant, experiments performed in the higher-temperature region are necessary. Recently, E. Maugeri et al. examined the behavior of helium release from UO_2 [10]. They showed that the release of helium occurs primarily at approximately 850 K (helium depletion of the surface) and in the temperature region beyond 1200 K (main release from the grains). Notably, the temperature range of helium release from grain coincides with the sintering temperature range of oxide fuel. Indeed, significant sintering of pressed fuel powder is known to occur in this temperature range [11–16].

Although helium is a noble gas element similar to the FP gas elements (xenon and krypton), the behavior of helium in fuel is peculiar. So far, the behavior of helium in plasma facing materials has been intensively studied with respect to microstructural changes [17–22]. The behavior of helium in UO_2 has become a topic of investigation because of the development of high-burn-up MOX fuel for LWR. This development is because MOX fuel contains a large amount of α -emitters and helium is accumulated in the fuel pellet. Accordingly, we initiated a series of studies to clarify the effect of the helium accumulated in raw powder during the sintering process of MA-MOX fuel for FBRs. In the present study, we focused on the variation in the surface morphology by the precipitation of dissolved helium in polycrystalline UO_2 . The grains were examined by BET, field-emission scanning electron microscopy (FE-SEM), and a scanning ion microscopy (SIM). Small defects similar to the trace of a blister were detected on the surface of S5. We focused on the hexagonal fringe that surrounds the basins, since the polyhedral shape is similar to the cross section of the polyhedral cavity formed in polycrystalline UO_2 . Such kind of cavity is called negative crystal. Recently, our investigation clarified that precipitated helium forms the negative crystal in UO_2 [23]. The shape of the negative crystal formed in the polycrystalline sample is usually truncated octahedron of which cross section is hexagonal. The similarity of the shape is interesting, and it is reasonable to consider that there should be some causal dependences between the blister formation and the negative crystal formation. In this manuscript, we discuss a series of phenomena paying attention on this point.

2. Experimental

In this study, a sintered UO_2 pellet (density: 10.68 g cm^{-3}) was ground into small particles to accelerate the infusion of helium by increasing the specific surface area of the sample. Appropriately-sized particles with diameters of 300–500 μm were separated by passing them through screens.

The scheme of the experimental setup to dissolve helium is shown in Fig. 1. The UO_2 sample (5.0g) was placed in a resistance furnace capable of operation under a high pressure of helium (Nikkiso Co. Ltd., Japan). The nominal purity of helium gas was

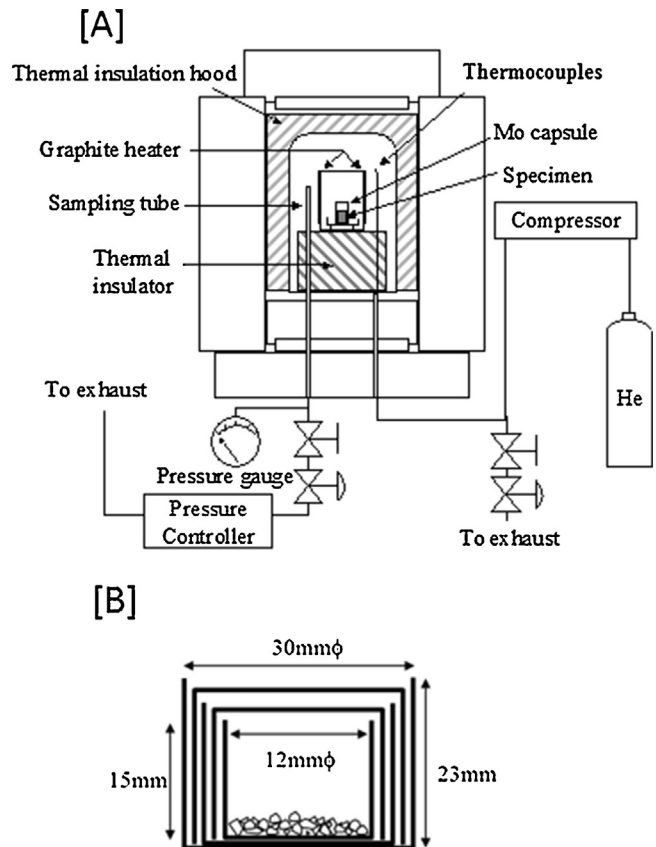


Fig. 1. Schematic of the experimental apparatus to dissolve helium in UO_2 : (A) the HIP setup and (B) the molybdenum capsule containing a UO_2 sample.

99.99995%. Table 1 summarizes the absorption conditions for each sample. As an example, the temperature–pressure record during the helium treatment of sample S2 is shown in Fig. 2. Both the heating and the cooling rates were approximately 20 K/min. The sample chamber was pressurized before the maximum temperature was attained and leaked after the temperature was decreased to restrain the release of dissolved helium. The stoichiometry of the sample was confirmed by thermogravimetric analysis prior to the absorption experiment.

The specific surface areas of samples S1, S2, S3, and S4 were measured by the BET method using krypton as an adsorptive gas. The apparatus employed in the present study was a BELSORP-max-32-N volumetric gas adsorption instrument (BEL Japan Inc.), which is designed to measure wide-range adsorption isotherms for surface area and pore size distribution analysis. The specific surface area was estimated from the krypton isotherm using the BET equation [24]. The pore size distribution of the sample was calculated from the krypton adsorption isotherm using the Dollimore–Heal method [25].

The surfaces of samples S1 and S5 were examined by a field emission-scanning electron microscope (SU-70, Hitachi

Table 1
Helium absorption conditions for each sample.

Sample	Temperature (K)	Pressure (MPa)	Heating time (h)
S1	–	–	–
S2	1473	30	25
S3	1473	61	25
S4	1473	91	25
S5	1473	91	100

Download English Version:

<https://daneshyari.com/en/article/1473415>

Download Persian Version:

<https://daneshyari.com/article/1473415>

[Daneshyari.com](https://daneshyari.com)