Microstructure analysis of MnO-Al_2O_3 doped UO_2 pellets

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1. Introduction

Korea Atomic Energy Research Institute (KAERI) has been developing the additives doped large grain UO_2 pellets for PCI remedy [1,2]. KAERI have designed several additives system, developed fabrication processes for additives doped UO_2 pellets, investigated pellets properties for the last several years. Recently, irradiation test for promising candidates in HANARO research reactor was accomplished and the discharged pellets are waiting for post irradiation examination (PIE). The discharged burnup is calculated to be 33MWd/kgU. The main concern of PIE will be the investigation of microstructural change after the irradiation and the fission products release behavior of irradiated pellets at an elevated temperature.

This paper will provide the results of microstructure analysis for non-irradiated additive doped UO_2 pellets. Pore and density distribution in a sample pellets are analyzed by using optical micrographs. SEM, EDS and WDS techniques are employed to characterize the additive particles remaining in a pellet and to examine the distribution of concentration of additive elements. The relationship between pore structure and distribution of additive elements are discussed in terms of evaporation of additive elements during the sintering process.

2. Experimental

Among the promising candidates, the MnO-Al_2O_3 doped UO_2 pellet was selected for microstructure investigation [1]. The density and averaged grain size of pellet were 10.72g/cm^3 and 37μm, respectively. Chemical analysis showed that there was a loss of doped elements after the sintering.

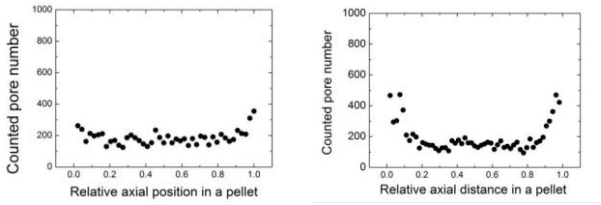
Pore and grain structure was analyzed by using optical microscope images. Sample pellets were longitudinally sectioned and polished. Two hundredfold (200×) of optical images were sequentially and equidistantly taken along the axial line of a pellet. The pore number, pore size and pore area in an individual image were calculated by using image analyzer.

After investigation of pore structure, the polished samples were thermally etched to measure grain size distribution in a pellet. The grain size of pellets was determined by linear intercept method.

The distribution of additive elements and composition of specific inclusions were measured by scanning electron microscopy (SEM), energy dispersive spectroscopy (EDS) and wavelength dispersive spectroscopy (WDS). Three locations of edge, middle, and center within each sample were selected for investigation. EDS and WDS mapping has been applied to quantify the number of undissolved additive particles in a pellet. Those mapping images were obtained from a polished section of test pellet to investigate the pore structure.

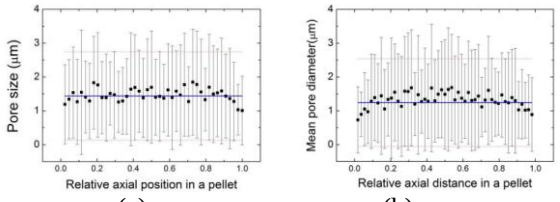
3. Results

3.1. Pore structure



(a) (b)

Fig. 1. Quantity variation of pores along the axial position in a pellet. (a) as-sintered, (b) resintered



(a) (b)

Fig. 2. Pore size along the axial position in a pellet. (a) as-sintered, (b) resintered

Fig. 1(a) shows the variation of pore quantity along the relative axial position of a pellet. Quantity of pores has a tendency to increase in the periphery region of pellet. Fig. 1(b) shows the change of pore numbers after the resintering test. Resintering test is a thermal annealing process conducted for 24h at 1700°C under H_2 atmosphere. While the pore quantity in the middle

and center region of a pellet slightly reduced, the number of pores in the periphery increased significantly after the resintering.

Figs. 2(a) and (b) show the locally averaged pore size variation along the pellet for the as-sintered and the resintered pellets. In contrast to variation of the quantity of pore (Fig. 1), pore size has a tendency to decrease in the periphery region.

3.2. Additive particle distribution

Fig. 3 shows EDS mapping images for Mn elements. Bright spots in the Mn mapping images imply that undissolved or precipitated particles of Mn are distributed in UO_2 matrix. The center region (①) of a pellet contains several Mn-rich particles. The number of Mn-rich particles reduced in the middle region (②). In the edge (③), Mn-rich particle was not detected within mapping images, indicating that limited number of Mn-rich particles exist in the edge.

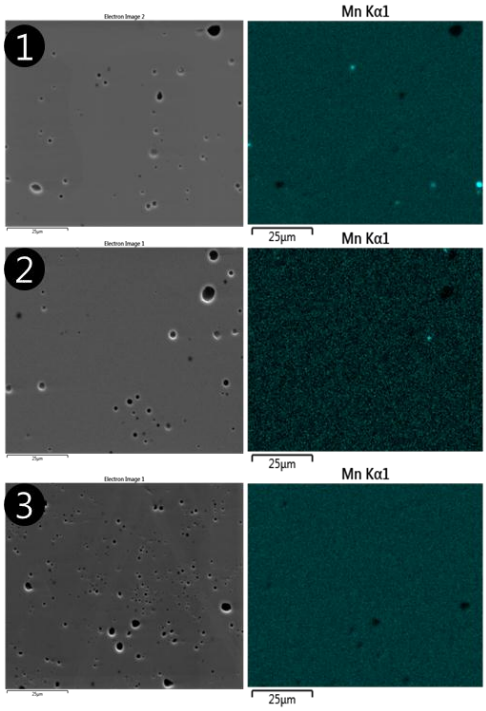


Fig. 3 SEM images, EDS mapping images from cross-section of pellet.

Nine points were randomly selected in the edge, middle and center part of pellet, respectively. The composition of Mn in UO_2 matrix at each point was measured by WDS. Fig. 4 shows measurement results of the weight ratio of Mn/U at respective point. The graph reveal that Mn content in UO_2 matrix decreased in the edge region. This result implies that the Mn dissolved in UO_2 matrix near pellet surface diffused out from the pellet.

Compared to the center and middle regions of the pellet, the edge can be characterized by increased quantity of small sized pores and reduced concentration of both Mn-rich particle and Mn element. This typical microstructure evolution might be explained as follow. Chemical analysis result showed that loss of Mn element occurred during the sintering. The Mn in the edge will preferentially diffused out from the pellet at the sintering stage. Consequently, the Mn elements in a particle in the edge will diffuse into the UO_2 matrix to reach an equilibrium solubility limit. Then the sites of particles where additive elements are evacuated will remain as a small pore.

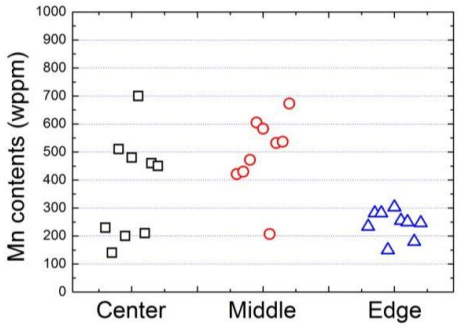


Fig. 4. The variation of Mn/U ratio according to the position in a pellet.

4. Conclusions

The microstructure of the MnO-Al_2O_3 doped UO_2 pellets are investigated by optical micrographs. SEM, EDS and WDS. Compared to the center and middle part of a pellet, the edge showed a different microstructure that is characterized by increased quantity of small sized pores and reduced concentration of both Mn-rich particle and Mn element. This typical microstructure evolution was explained in terms of the loss of Mn element during the sintering.

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