Recycling process of Mn-Al doped large grain UO_2 pellets

Ik Hui Nam, Jae Ho Yang, Young Woo Rhee, Dong-Joo Kim, Jong hun Kim, Keon Sik Kim, Kun Woo Song*

Korea Atomic Energy Research Institute,
(150-1 Dukjing-Dong), 1045 Daedeokdaero, Yuseong-gu, Daejeon-si 305-353, Korea
nam1313@kaeri.re.kr

1. Introduction

To reduce the fuel cycle costs and the total mass of spent light water reactor (LWR) fuels, it is necessary to extend the fuel discharged burn-up [1]. Research on fuel pellets focuses on increasing the pellet density and grain size to increase the uranium contents and the high burn-up safety margins for LWRs.

KAERI are developing the large grain UO_2 pellet for the same purpose. Small amount of additives doping technology are used to increase the grain size and the high temperature deformation of UO_2 pellets. Various promising additive candidates had been developed during the last 3 years and the $MnO-Al_2O_3$ doped UO_2 fuel pellet is one of the most promising candidates.

In a commercial UO_2 fuel pellet manufacturing process, defective UO_2 pellets or scraps are produced and those should be reused. A common recycling method for defective UO_2 pellets or scraps is that they are oxidized in air at about 450°C to make U_3O_8 powder and then added to UO_2 powder. In the oxidation of a UO_2 pellet, the oxygen propagates along the grain boundary. The U_3O_8 formation on the grain boundary causes a spallation of the grains. So, size and shape of U_3O_8 powder deeply depend on the initial grain size of UO_2 pellets. In the case of Mn-Al doped large grain pellets, the average grain size is about 45μm and about 5 times larger than a typical un-doped UO_2 pellet which has grain size of about 8~10μm. That big difference in grain size is expected to cause a big difference in recycled U3O8 powder morphology.

Addition of U_3O_8 to UO_2 leads to a drop in the pellet density, impeding a grain growth and the formation of graph- like pore segregates. Such degradation of the UO_2 pellet properties by adding the recycled U_3O_8 powder depend on the U_3O_8 powder properties. So, it is necessary to understand the property and its effect on the pellet of the recycled U_3O_8.

This paper shows a preliminary result about the recycled U_3O_8 powder which was obtained by oxidation of Mn-Al doped large grain pellets.

2. Experimental

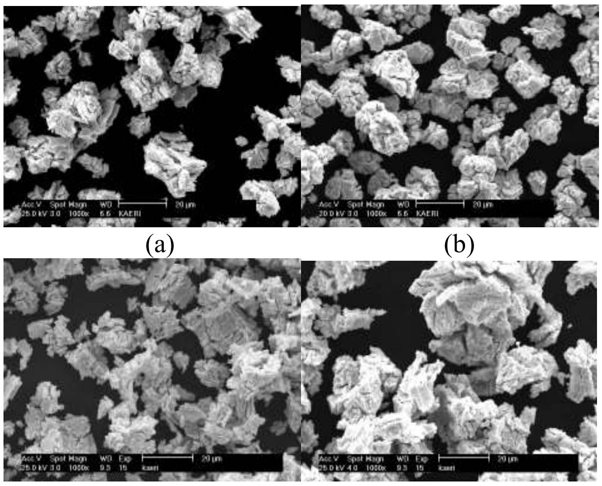
The 1000ppm of Mn-Al doped UO_2 fuel pellets were prepared by a conventional sintering process. Sintered pellets were oxidized to U_3O_8 powders at different temperatures of 330 and 450 °C in air, respectively. The U_3O_8 powders morphology was examined by SEM.

3~10wt% of those U_3O_8 powders and $MnO-Al_2O_3$ additives were added to the UO_2 powder. Total amount of Mn-Al in the powder mixture was 1000ppm in weight. Powder mixtures were mixed with a tumbling mixer. The powder mixture was pressed into green pellets at 300 MPa. The green pellets were sintered at 1730 °C for 4 h in flowing H_2 gas.

The sintered density of the UO_2 pellets was measured by a water immersion method. The pellets were sectioned axially, ground and polished. The polished pellets were thermally etched at 1250 °C in carbon dioxide gas in order to examine their grain boundaries. The pore and grain structure were examined by an optical microscope and grain size was determined by a linear intercept method.

3. Results

3.1. Recycled U_3O_8 powders morphology.



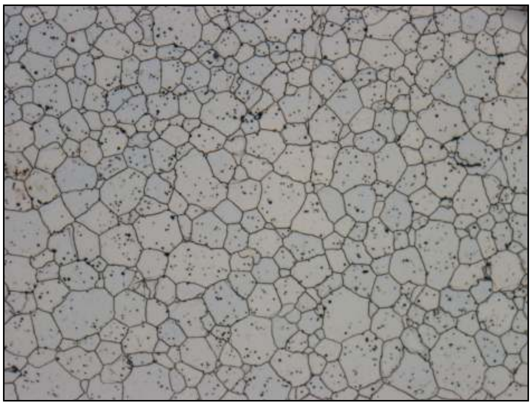
(a) (b) (c) (d)

Fig. 1 SEM powder morphologies of recycled U_3O_8 powders obtained by oxidizing the conventional UO_2 pellets (a), (b) and 1000ppm Mn-Al doped large grain pellets (c), (d). The oxidation temperatures were 330℃ (a), (c) and 450℃ (b), (d), respectively.

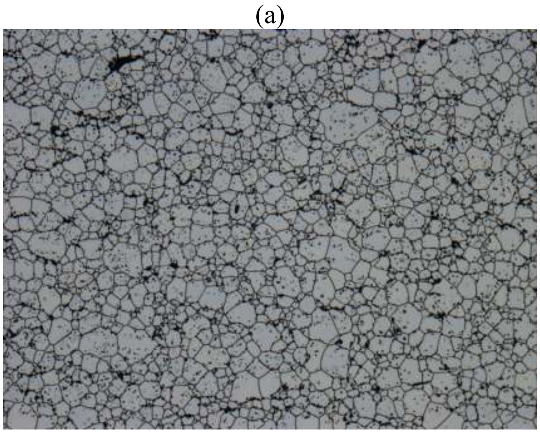
Fig. 1 shows the SEM micrographs of U_3O_8 powder which was obtained by oxidizing the conventional and large grain UO_2 pellets at different temperatures. In this figure, we can find that the U_3O_8 particle size from the large grain pellets is quite larger than that from conventional UO_2 pellets. As mentioned above, this size difference is due to the grain size difference between the pellets before oxidation. Generally, It is known that the sinter-ability of powder decrease with increasing the particle size. So, the pellet density and grain size might be more decreased when we reuse the U_3O_8 powder of (c) or (d) instead (a) or (b) in the fabrication of UO_2 pellet.

3.2. Effect of recycled U_3O_8 powders on the Mn-Al doped UO_2 pellets.

5wt% of large grain U_3O_8 powder which is correspond to the powder of Fig. 1(d) was added to Mn-Al doped UO_2 powder and mixture was sintered into pellets. Fig.2 shows the grain structure of 1000ppm of Mn-Al doped UO_2 pellets without or with the addition of large grain U_3O_8 powder, respectively. Fig. 2(b) clearly reveals the density and grain size reduction by addition of large grain U_3O_8 powder. The pellet density was measured to be 98 and 95.8%TD, respectively. In conventional process, about 0.2%TD of density was decreased by addition of 1wt% U_3O_8 powder. In this case, 0.5% of density drop was observed by the addition of 1wt% U_3O_8.



(a)



(b)

Fig. 2. Grain structures of sintered pellets.
(a) Mn-Al doped UO2 pellet, (b) Mn-Al and 5wt% of U_3O_8 doped UO2 pellet.

4. Conclusions

The powder size of a recycled U_3O_8 from Mn-Al doped large grain UO_2 pellet was very larger than that from conventional UO_2 pellet. These large size recycled U_3O_8 powder accelerate the density drop and grain size decrease of UO_2 pellet when it reuse in the fuel fabrication process. . So, it is necessary to develop an advanced recycling process of large size U_3O_8 powder in order to enhance the sinterability, thereby to enhance the nuclear fuel pellet properties.

Acknowledgements

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[1] Ph. Dehaudt et al. , Procceding of the IAEA Technical Committed Meeting, "Advanced in fuel pellet technology for improved performance at high-burnup", Tohyo, October 28-November 1, 27 (1996)