The Effect of the O/U Ratio on the Sintered Density of the UO₂ Pellet

S. H. Na_a, K. H. Kang_a, Y. H. Kim_a, C. J. Park_a, K. C. Song_a and M. J. Yoob

^a Korea Atomic Energy Research Institute, 150 Deokjin-dong, Yuseong-gu, Daejeon 305-353, Korea, shna@kaeri.re.kr ^bKOREA Nuclear Fuel Company, Ltd., 493 Deokjin-dong, Yuseong-gu, Daejeon 305-353, Korea

1. Introduction

The sintered density of the UO₂ pellet is an important factor to assure a stable nuclear reactor control. There are some methods to control the sintered density of the UO₂ pellet, that is, a sintering temperature and its time, a green density, an addition of pore-former or U_3O_8 , etc. In general, it is well known that the sintered density of UO₂ pellet increases as the sintering temperature and its time and the green density increases. However the addition of a pore-former or U_3O_8 decreases the sintered density of the UO₂ pellet, due to the leave various sizes of pore in the UO₂ matrix during sintering.

In this work, the effect of the O/U ratio on the sintered density of the UO_2 pellet are investigated.

2. Methods and Results

To have various O/U ratio of the natural ex-ADU UO_2 powder, the powder was milled by a DM(Dynamic Mill) with varying milling time. Fig. 1 shows the DM and the O/U ratio of the milled powder as a function of milling time. The DM rotates 25 rpm with zirconia balls(dia. ; 8mm, internal volume ; 70vol.%) and 50g of the UO_2 powder inside. As shown in Fig. 1, the O/U ratio of the powder increases as the milling time increases, and above 2 hours of milling time, the O/U ratio of the powder sharply increases. But up to 2 hours the O/U ratio of the powder shows little increase and satisfies with about 2.27 of the O/U ratio.



Fig. 1 Milling time vs. O/U ratio of the powder

The milled powder is compacted to make green pellets with a compaction pressure of 300 MPa. Then the green

pellets are sintered at the temperature of 1750 °C, H_2 atmosphere with two sintering times (6 and 12 hrs). The sintered density of the UO₂ pellet is measured by an immersion method.

Fig. 2 shows the sintered density as a function of the O/U ratio of the UO_2 powder. As shown in Fig. 2, as the O/U ratio of the powder increases, the different degree of decrease of sintered density appears 3 steps, according to the O/U ratio of the powder. At the first step (< 2.268), the sintered density decreases linearly, at the second step(2.268~2.272), the sintered density decreases sharply, and at the final step(> 2.272), the sintered density decreases linearly again. Similar phenomenon occurred at the sintering condition of 12 hours. With increasing of the sintering time, the sintered density increases up to 2.268 of the O/U ratio, but above 2.272 the sintered density shows no difference. It is considered that these appearances (sharp decrease and no difference) are attributed to the formation of the U_4O_9 in the milled powder.



Fig. 2 Sintered density vs. O/U ratio of the powder

The excess oxygen, which is produced at the transformation of the UO_2 from the U_4O_9 during sintering in the H₂ atmosphere, leaves various sizes of pores in the matrix. In general, small size of the pore in the matrix disappears and the large pore remains or enlarges with increasing a sintering time. Therefore, in spite of the increase of the sintering time, the larger pore, due to the transformation of the UO_2 from a new phase U_4O_9 of milled powder during sintering, formed and did not eliminate.

Fig. 3 shows the XRD(X-Ray Diffraction) pattern with varying the O/U ratio of the milled powder. As shown in

Fig.3, the O/U ratio of the milled powder shows remarkably the phase of U_4O_9 in large part and the UO_2 in small part.



Fig. 3 XRD pattern of various O/U ratio of powder

3. Conclusion

- The sintered density of the UO_2 pellet shows 3 decrease steps according to the O/U ratio of the UO_2 powder.
- Below 2.27 of the O/U ratio, the sintered density decreases as the O/U ratio increases, but the sintered density increases as the sintering time increases.
- In the range of about 2.27(± 0.02) of the O/U ratio, the sintered density sharply decreased. It is considered that the transformation of the UO₂ from the U₄O₉ during sintering leaves large pores.
- Above 2.27 of the O/U ratio, in spite of increase of the sintering time, the sintered density of the UO_2 pellet increase little. It is considered that the larger pores formed in the UO_2 matrix during sintering did not eliminated.

Acknowledgement

This work has been carried out under the Nuclear R & D program by the Ministry of Science and Technology (MOST), Korea.