Effect of Sintering Atmosphere on the Scrap Recovery of UO2+5wt%CeO2 Pellets

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1. Introduction

Nuclear fuel pellet involves complex phenomena during irradiation in a reactor such as the densification and the swelling. These phenomena depend largely on sintered density and pore size distribution of the pellet [1]. Pellets manufactured by addition of M₃O₈ phase scrap powder can control porosity and density for reducing densification or to make larger grain size for reducing fission gas release [2, 3]. Harada[4] indicated that the densification and grain growth of UO_{2+x} were enhanced by increasing the oxygen partial pressure. In this present work, the effect of M₃O₈ addition on the sintering behavior of UO₂+5wt%CeO₂ is investigated by its addition in the range of 0-50 wt% and by oxidizing step. And these pellets were observed of changes in sintered density and microstructure.

2. Experimental

2.1 Powder Treatments

UO2 powder used in this work was BNFL IDR UO2, which has an O/U ratio of 2.11 and an average particle size of 2.2 µm. CeO₂ powder, as a surrogate material of PuO2, has an average particle size of 6.66 μm and a specific surface area of 9.46 m²/g. M₃O₈ powder was prepared by heating scrap MO2 pellets in air at 500 for 5 h. UO2 and 5wt%CeO2 mixture is obtained by admixing weighed amounts of the UO2 and CeO₂ powders in a Turbula mixer for 1h, and then milling for 80 min by using a planetary mill (FRITSCH pulverisette 6). M₃O₈ scrap powder was admixed to UO2+5wt%CeO2 matrix powder up to 50wt% by either mixing or milling. In the milling method, scrap powder and matrix powder were mixed and then milled altogether. In the mixing, the weighed amount of scrap powder was added to the matrix powder, which was milled by using the planetary mill and then mixed together by using the Turbula mixer for 1h.

2.2 Sintering

The powder mixture was pressed with a compaction pressure of 300 MPa, by using a double-acting hydraulic press. The diameter and length of the compacts were 10mm and 8mm, and the compacts have green density of $6.1\Box 6.7$ g/cm³. In the conventional single-stage process, the compacts were heated at heating rate of $3\Box/\text{min}$ and then held at $1700\Box$ for 4 h in a reducing atmosphere of H_2 . The three-stage process used in this work was carried out in H_2 atmosphere to setting temperatures ($800\text{-}1400\Box$) and then changed H_2

atmosphere to CO₂ gas for 2 h during heating up. The density of the pellets was measured by the liquid immersion method. The average grain size on the polished and chemically etched surface was determined using the linear intercept method.

2. Results

Properties of MO_2 powder treated with milling and mixing methods is shown in Fig. 1. Surface area of MO_2 powder treated with milling and mixing methods is decreased with increasing M_3O_8 addition amount. Particle size is about 0.4 um without scrap.

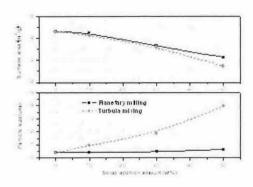


Fig. 1 Properties of MO₂ powder treated with milling method and mixing method

The density variations of MO₂ pellets sintered at 1700°C for 4 hours with different atmosphere control and admixing methods are shown in Fig. 2.

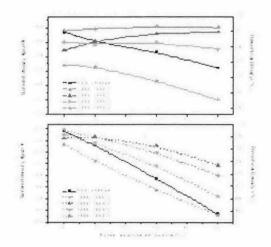


Fig. 2 Density variation of MO₂ pellets sintered with different sintering atmospheres at 1700 for 4 hours with a) milling method and b) mixing method.

As shown in Fig. 2(a), the density of sintered pellets sintered with milling method was regularly decreased with increasing the amount of scrap addition in the case of oxidizing step between 1400 Gnd 1600 Gnd non-oxidizing step. But the sintered density of pellets in the case of oxidizing step below 1400 was almost not changed with increasing the amount of scrap. As shown in Fig. 2(b), the density of pellets sintered with mixing methods was regularly decreased with increasing the amount of scrap addition. The density differences of pellets in the identical condition were greatly increased with increasing scrap amount.

	MO ₂	MO2+50wt% M3O3
Without step		
Oxidizing step (1200-1400°C)	_	

Fig. 3 Microstructures of MO_2 and $MO_2+50wt\%$ M_3O_8 pellets sintered at $1500\Box$ for 4 h in the H_2 atmosphere.

The microstructures of sintered MO₂ pellets with oxidizing step and admixing methods are shown in Fig. 3. The grain size for the pellets without scrap was about two times larger than that of the pellets added with 50wt% scrap. The grain size of MO₂ pellets sintered with oxidizing step between 1200 °Cand 1400 °Cwere larger than in other sintering conditions used in this work.

3. Conclusion

The surface area of MO_2 powder treated with milling and mixing methods is decreased with increasing M_3O_8 addition amount. The density of pellets sintered with milling method was little changed and that of mixing method was regularly decreased with increasing the amount of scrap addition. The grain size of MO_2 pellets sintered with oxidizing step between 1200° Cand 1400° C were larger than in other sintering conditions. Pore and grain structure can be controlled by the addition method of scrap powder and by the control of oxidizing step.

ACKNOWLEDGEMENT

This study was performed under the auspices of the Korea Ministry of Science and Technology.

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