Contributions of Additives to a Densification and Grain Growth of UO_2+5w%CeO_2

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1. Introduction

Several kinds of additives have been used in the powder processes for a ceramic fuel fabrication to accelerate the sinterability of a compact and/or to modify the microstructure of a sintered pellet [1-3]. The properties of fuel pellet, such as plasticity, fission gas retaining ability, thermal conductivity, can be improved by the addition of an additive to UO_2 based ceramic fuel [4]. The contributions of an additive to the densification and grain growth of a fuel pellet are different from the formation of a solid solution and defect structure in a UO_2 matrix [5].

Ten kinds of oxides were used as additives in this study. Each oxide was added to a UO_2+5w%CeO_2 mixed oxide. The contribution of each additive to the sinterability of the mixed oxide was evaluated under two different sintering atmospheres.

2. Experimental

The main raw powders used in this study were IDR-UO_2(Integrated Dry Route UO_2 powder, BNFL) and CeO_2(Aldrich, 99.9%, rare earth impurities<1000ppm). Ten kinds of oxide, Li_2O, $LiAlO_2$, Cr_2O_3, ZrO2, Ta_2O_5, MoO_3, Al_2O_3, Y_2O_3, TiO_2 and Nb_2O_5 were used as additives.

Each additive was admixed to UO_2+5w%CeO_2 with the amount of 0.02 to 1.0 w% and milled the mixed oxide using an attrition mill. Each milled powder was pressed into a compact with a pressure of 300MPa, then sintered at 1973K for 4h under $92N_2+8H_2$ or $84N_2+8H_2+8CO_2$ atmospheres.

Sintered density and grain size of the pellets were evaluated for various additives and sintering atmospheres.

3. Results and Discussion

Cations of an additive oxide enter into the UO_2 lattice substitutionally or interstitially during a sintering that forms defect structures. The effects of additives on the sintering process of UO_2+5w%CeO_2 are different from the formation mechanism of a solid solution, cation valence and oxygen potential in the sintering process.

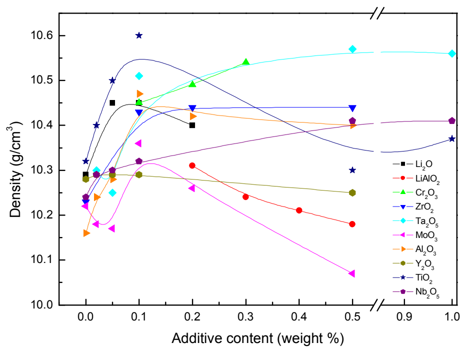

Fig. 1. Effect of an additive on the density of sintered in reducing atmosphere.

Fig. 1 shows the density change of the (U, Ce)O_2 pellets with additive content. Most additives except for MoO_3 and Y_2O_3 contribute to the densification of (U, Ce)O_2 in less than 0.1w% of an additive content. The density increased with the additive content up to 1.0w% only for Ta_2O_5, Nb_2O_5 but the other additives did not contribute to the densification above 0.1w%.

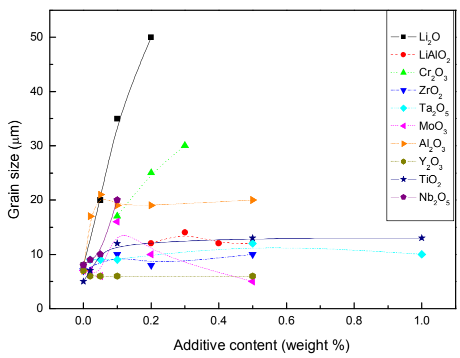

Fig. 2. Effect of additive on the grain growth of the (U, Ce)O_2 pellets.

Fig. 2 shows the additive effect on the grain growth of the (U, Ce)O_2 pellets for a sintering in reducing atmosphere. Among the additives, Li_2O

accelerated the grain growth most, and next in the order of Cr_2O_3, Nb_2O_5 and Al_2O_3. Fig. 2 shows Ta_2O_5 did not contribute to the grain growth but was very effective on the densification as shown in Fig. 1. TiO_2, $LiAlO_2$, Y_2O_3 and ZrO_2 also were not effective for the grain growth of the (U, Ce)O_2 pellets in a reducing atmosphere.

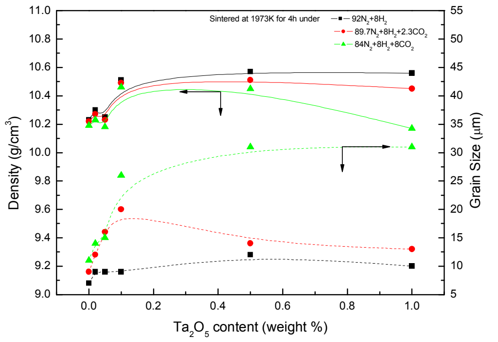

Fig. 3. Shift of Ta_2O_5 effect on the densification and grain growth of (U, Ce)O_2 pellets with adding CO_2 gas to sintering atmosphere.

When a CO_2 gas was added to the sintering atmosphere, the effect of Ta_2O_5 on the densification and grain growth of the pellets was changed as shown in Fig. 3. Ta_2O_5 accelerated the grain growth with an increasing oxygen potential in the sintering atmosphere.

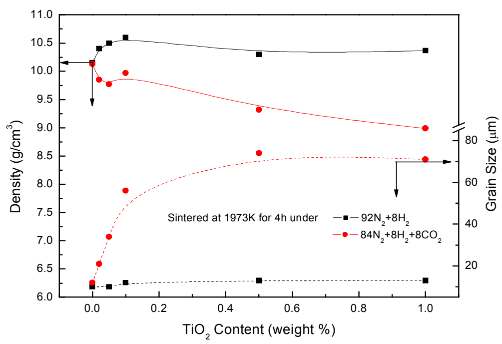

Fig. 4. Effect of TiO_2 on the density and grain size of (U, Ce)O_2 pellet under a different sintering atmosphere.

Under a reducing atmosphere, some Ce^{4+} will be reduced to Ce^{3+}. If Ta^{5+} and Ce^{3+} cations enter into the UO_2 lattice substitutionally, both $Ta^{\cdot}$ and Ce' could combine and form a defect cluster, ($Ta^{\cdot}$ Ce'), for a charge compensation. When adding CO_2 to the sintering atmosphere, the defect cluster could not exist because Ce existed in the lattice as a neutral Ce_U^x. Fig. 3 shows the contribution of Ta_2O_5 on the densification and the grain growth could be affected by an oxygen potential in the sintering atmosphere.

The effect of TiO_2 was also changed by adding CO_2 gas, as shown in Fig. 4. In the case of Ta_2O_5 and TiO_2, oxygen potential in the sintering atmosphere more prominently contributed to the density and grain size of the pellet rather than their content.

4. Conclusion

The contribution of oxide additives to the densification and grain growth of (U, Ce)O_2 was studied under different sintering atmospheres. In the reducing atmosphere, Ta_2O_5 affected the density the most and Li_2O the grain growth of the pellet.

In the case of Ta_2O_5 and TiO_2, the oxygen potential was more effective on the grain growth of the pellet than an additive content.

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