

Oxidation of Scrap Duplex Burnable Absorber Pellets

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1. Introduction

Duplex burnable absorber pellet has two distinct portions with different compositions. One is the annular outer portion composed of an enriched $\text{UO}_2\text{-2\%Er}_2\text{O}_3$ and the other is the cylindrical inner portion composed of a natural or depleted $\text{UO}_2\text{-12\%Gd}_2\text{O}_3$. A duplex structure consisting of different materials is advantageous over a structure using either a sintered $(\text{U,Gd})\text{O}_2$ or $(\text{U,Er})\text{O}_2$ material as a sintered burnable absorber pellet, because the duplex structure exhibits an improved nuclear performance in a nuclear reactor [1].

Generally, about 10 % of pellet can not satisfy the specifications during a nuclear fuel pellet fabrication. In a commercial UO_2 or burnable absorber pellet fabrication process, those scrap pellets are usually recycled as a U_3O_8 or M_3O_8 ($\text{M}=\text{U-Gd}$ or U-Er) powder form though an oxidation at 350-550 °C in air.

However, in the case of a duplex burnable absorber pellet, a simple oxidation technique could not be used for a scrap recycling, because it is not helpful to mix the enriched uranium of the annular outer portion and the natural or depleted uranium of the cylindrical inner portion. Also it is undesirable to mix gadolinium and erbium.

Thus, it is necessary to develop a new technique for recycling a scrap duplex burnable absorber pellet. In this study, the oxidation behavior of a scrap $\text{UO}_2\text{-12\%Gd}_2\text{O}_3/\text{UO}_2\text{-2\%Er}_2\text{O}_3$ duplex pellet has been investigated and a promising method for dividing the recycled scrap powders has been proposed.

2. Experimental

A mixture of 2% by weight of Er_2O_3 powder and UO_2 powder was charged into a tubular mixer and then mixed for 1 hour to prepare a mixed powder, $\text{UO}_2\text{-2wt\%Er}_2\text{O}_3$, for the annular outer portion of a duplex nuclear fuel. A Gd_2O_3 powder was subjected to ball milling using zirconia balls for 12 hours to enhance the solid state reaction. Then, the powder thus prepared was mixed with UO_2 powder in a tubular mixer for 1 hour, and then pulverized in a pestle and mortar for 10 minutes to prepare a mixed powder, $\text{UO}_2\text{-12wt\%Gd}_2\text{O}_3$, for the cylindrical inner portion.

The mixed powder of $\text{UO}_2\text{-2wt\%Er}_2\text{O}_3$ was charged into the annular outer portion and the $\text{UO}_2\text{-12wt\%Gd}_2\text{O}_3$ was charged into the cylindrical inner portion, which was then subjected to pressing to produce a duplex compact following the same procedure in a previous study. [2]

The compact was heated to 1730 °C at a rate of 5K/min., and then maintained under a reducing gas atmosphere at 1730 °C for 4 hours in a mixed gas $\text{H}_2\text{-3\%CO}_2$ to fabricate a sintered duplex pellet for the scrap recycling experiments.

Samples for the oxidation test were prepared with the above powders and compacted in a simple cylindrical mold. The compacts were sintered at the same condition as the duplex.

Sintered simple pellets were cut to 1.0-1.2 mm in thickness and 7-8 mm in diameter from a sintered pellet and polished both sides for the oxidation test. The isothermal oxidation tests were conducted at various temperatures in flowing air at a rate of 50 cc/min by using a thermo-gravimetric analyzer (TGA, Shimadzu TGA-51). Samples were quickly heated up to the oxidation temperature in Ar with a heating rate of 30 K/min.

Oxidation of the pellet was conducted at several temperatures in a box furnace and the morphologies of the oxidized powders were investigated.

3. Results

Figure 1 shows the sintered $\text{UO}_2\text{-12\%Gd}_2\text{O}_3/\text{UO}_2\text{-2\%Er}_2\text{O}_3$ duplex pellet. Apparently, no cracks are found at the surface of the annular and the cylindrical portions.

Before the scrap duplex pellet oxidation, the oxidation of the simple pellets was conducted, where the simple pellet refers to the pellet with the composition of the inner or the outer region of the duplex pellet. Weight gains during the isothermal oxidation of the simple $\text{UO}_2\text{-2wt\%Er}_2\text{O}_3$ and $\text{UO}_2\text{-12wt\%Gd}_2\text{O}_3$ pellets at various temperatures are shown in Fig. 2 as a function of the oxidation time.

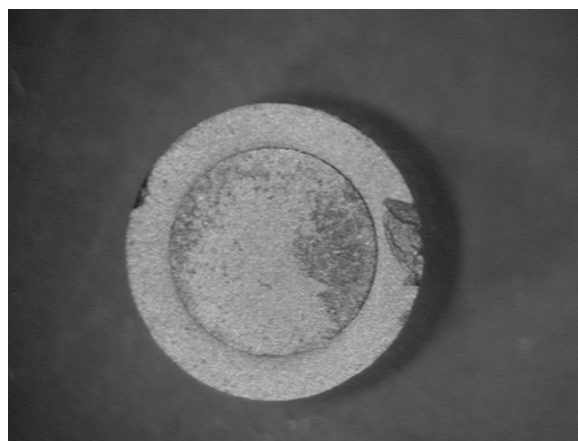


Fig. 1. Sintered Duplex Burnable Absorber Pellet.

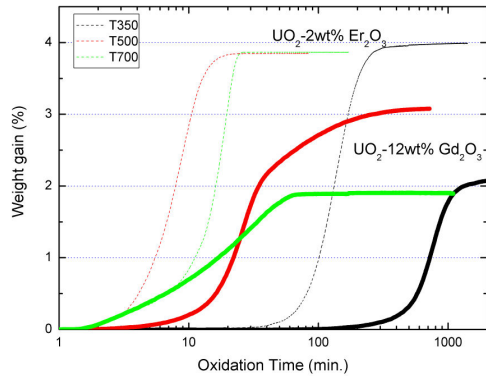


Fig. 2. Weight gain versus time for sintered $\text{UO}_2\text{-2wt\%Er}_2\text{O}_3$ and $\text{UO}_2\text{-12wt\%Gd}_2\text{O}_3$ pellets at 350, 500 and 700 °C.

A fully developed sigmoidal curve is observed at all temperatures for the $\text{UO}_2\text{-2wt\%Er}_2\text{O}_3$ pellets. It appears that the $\text{UO}_2\text{-2wt\%Er}_2\text{O}_3$ pellets are fully oxidized to the M_3O_8 phase. The most rapid oxidation occurs at 500 °C. In the case of the $\text{UO}_2\text{-12wt\%Gd}_2\text{O}_3$ pellets, the weight gain seemed to be restricted to certain values. It appeared not to be fully oxidized to the M_3O_8 phase. Thomas et al. [3] reported that this is attributed to the higher stability of M_4O_9 phase than that of M_3O_8 . The stability of the M_4O_9 phase is known to be depended on the concentration of Gd [4].

These oxidation characteristics could result in different morphologies of the oxidized powders. Figure 3 shows the morphologies of the M_3O_8 powder obtained by a oxidation of sintered $\text{UO}_2\text{-2wt\%Er}_2\text{O}_3$ and $\text{UO}_2\text{-12wt\%Gd}_2\text{O}_3$ pellets at various temperatures. $\text{UO}_2\text{-2wt\%Er}_2\text{O}_3$ pellets are oxidized to fine powders at all temperatures; however, the oxidized powders of the $\text{UO}_2\text{-12wt\%Gd}_2\text{O}_3$ pellets seemed to be coarse. In the case of the oxidation at 700 °C, the shape of the sintered pellet remained after an oxidation. We tried to use this oxidation characteristic to divide the inner and the outer portion of the duplex pellet. We successfully managed to divide the both portions.

4. Conclusion

The oxidation behavior of simple pellets which had the compositions of the inner and the outer portion of a duplex burnable absorber pellet was investigated. $\text{UO}_2\text{-2wt\%Er}_2\text{O}_3$ pellets showed a similar oxidation behavior to the UO_2 pellets. However, in the case of the $\text{UO}_2\text{-12wt\%Gd}_2\text{O}_3$ pellets, the weight gain did not proceed after certain values. $\text{UO}_2\text{-12wt\%Gd}_2\text{O}_3$ pellets could not be fully oxidized to the M_3O_8 phase because of the higher stability of M_4O_9 . Different morphologies of the oxidized powders resulted from the unique oxidation characteristics of the high Gd content pellet. The inner and the outer portions of the duplex pellet could be

divided by using the above oxidation characteristics, successfully.



Fig. 3. Morphologies of the M_3O_8 powder obtained by oxidation of sintered $\text{UO}_2\text{-2wt\%Er}_2\text{O}_3$ and $\text{UO}_2\text{-12wt\%Gd}_2\text{O}_3$ pellets at various temperatures.

Acknowledgement

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