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Effects of Powder Property and Sintering Atmosphere on the Properties of Burnable Absorber Fuel : I. UO_2-Gd_2O_3 Fuel

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Abstract

UO_2-Gd_2O_3 fuel has been sintered to study the effect of powder property and sintering atmospheres on densification and microstructure. Three types of powders have been used; AUC-UO_2 powder and ADU-UO_2 powder were mixed with Gd_2O_3 powder, and co-milled AUC-UO_2 and Gd_2O_3 powder. UO_2-(2,5,10)wt% Gd_2O_3 pellets have been sintered at 1680℃ for 4 hours in the mixture of H_2 and CO_2 gases, of which oxygen potential has been controlled by the ratio of CO_2 to H_2 gas. Densities of UO_2-Gd_2O_3 fuel pellets are quite dependent on powder types, and UO_2-Gd_2O_3 fuel using co-milled UO_2 powder yields the highest density. A long range homogeneity of Gd is determined by powder mixing. As the oxygen potential of sintering atmosphere increases, the sintered densities of UO_2-Gd_2O_3 pellets decrease but grain size increases. In addition, $(U,Gd)O_2$ solid solution becomes more homogeneous. The UO_2-Gd_2O_3 fuel having adequate density and homogeneous microstructure can be fabricated by co-milling powder and by high oxygen potential.

1. Introduction

As a burnable absorber which suppresses initial excess reactivity at BOL (beginning of life) in LWRs, UO_2-Gd_2O_3 fuel has been widely used in the world. In our country UO_2-Gd_2O_3 fuel had been loaded together with KOFA fuel in all PWRs, and now it is being used in the Young Gwang nuclear unit 3 &4. UO2-Gd2O3 fuel has not been fabricated in Korea since the number of UO2-Gd2O3 rods has not been many. However, more burnable absorber are expected to be used in longer fuel cycle. Especially in the case of new burnable absorbers such as IFBA or UO_2-Er_2O_3 fuel their numbers reach 25% of the number of UO_2 fuel rads. So the fabrication of burnable absorber will be more and more important in the future.

Technology of the UO_2-Gd_2O_3 fuel was developed originally on the basis of UO_2 fuel technology. So both fabrication technologies are very similar; powder mixing, pressing and sintering. Currently, the Gd_2O_3 content of 8-10 wt% is not exceptional in PWRs. The UO_2-Gd_2O_3 fuel containing high Gd_2O_3 content is known to have low density and inhomogeneous microstructure, compared with normal UO_2 fuel. The fabrication of UO_2-Gd_2O_3 pellets has continuously been studied to solve those problems.

This work has been undertaken to study what variables are important in fabricating

UO_2-Gd_2O_3 fuel.

2. Experimental

Three types of powders were used. First, the UO_2 powder derived from AUC was mixed with Gd_2O_3 powder. Second, the UO_2 powder derived from ADU was mixed with Gd_2O_3 powder. Two-step mixing, so called mater mixing, was carried out in order to get a homogeneous powder mixture , in the first mixing step UO_2 powder was mixed with the same amount of Gd_2O_3 powder for 2 hours. In the second mixing step this preliminary mixture was diluted with extra UO_2 powder to meet final composition, and then mixed again. The third powder was prepared by co-milling AUC-UO_2 and Gd_2O_3 powders. Gd_2O_3 contents are 2, 5, and 10 wt%.

The powder mixtures were pressed into compacts, and they were sintered at 1680℃ for 4 hrs. Sintered pellets were characterized by density, grain size and Gd distribution. Sintered density was measured by water immersion method, and grain size by a liner intercept method. Gd distribution was analyzed by WDX attached to SEM.

Four sintering atmospheres were used in this work; pure hydrogen and mixtures of hydrogen and carbon dioxide gases. The ratios of CO_2 to H_2 gas in the mixtures are 0.05, 0.15, and 0.30. The atmosphere in a cooling-down stage was always pure hydrogen, independently of previous sintering atmospheres.

The mixture of hydrogen and carbon dioxide reacts in thermodynamic equilibrium to yield CO, H_2O and O_2. The equilibrium reaction is somwhat complicated, so the SOLGASMIX program [4] was used to calculate the oxygen potential of sintering atmospheres. The oxygen potential increases with increasing the ratio of CO_2 to H_2. The oxygen potential of H_2 atmosphere is below -400 kj/mole. So the oxygen potential of sintering atmosphere ranged from -400 to -300 kj/mole.

3. Results and discussions

The AUC-UO_2 powder has a spherical shape and smooth surface. Its particle size is about 20 ㎛. One particle is composed of very fine crystallites and a lot of micropores are formed between the crystallites. The ADU-UO2 powder has a particle size of about 1.0 ㎛, so the powder is much agglomerated. The size of agglomerate is very irregular. The co-milled powder has a pparticle sixe of about 2 ㎛, which is much smaller than before co-milling. The particle size of Gd2O3 powder is about 4 ㎛.

The homogeneity of powder mixing was analyzed in the powder compacts. In the powder compact derived from mixing AUC-UO_2 with Gd_2O_3 powders, Gd is located between spherical UO_2 particles and so UO_2 particles seem to be coated with Gd_2O_3 powder. And some large agglomerates of Gd_2O_3 powder are found. In the powder compact derived from mixing AUD-UO_2 with Gd_2O_3 powders, agglomerates of Gd_2O_3 powder are found. In addition, these agglomerates seem to be concentrated in some areas and to be depleted in other areas. In the powder compact derived from the co-milled powder, no large agglomerates of Gd_2O_3 powder are detected and so Gd concentration is quite homogeneous.

Fig. 1 shows the dependence of sintered density on Gd_2O_3 contents and powder types. Fig. 1(a) is in hydrogen atmosphere. Sintered densities of pure UO_2 fuel are little affected by powder types. But sintered densities of UO_2-Gd_2O_3 pellets are very much influenced by powder types. When mixing AUC-UO_2 with Gd_2O_3 , even the addition of 2 wt% Gd_2O_3 can decrease the sintered density down to 90 %, and then higher content of Gd_2O_3 has no significant effect. When the co-milled powders is used, the density decrease is small, and so the highest density is achieved.

Fig. 1(b) shows sintered densities in the 30CO_2-H_2 atmosphere. The density difference between powder types is still prominent, and the sintered density gradually decreases with Gd_2O_3 contents. Thus the dependence of sintered density on the powder types and Gd2O3 contents is simliar in high oxygen ptential.

The effect of oxygen potential on sintered density is shown in Fig. 2. The sintered density of UO_2-10wt%Gd_2O_3 fuel decreases with increasing the ratio of CO_2 to H_2. Three different powders show similar behaviors. The largest decrease in density is found in the fuel pellet derived from mixing AUC-UO_2 with Gd_2O_3 powders. The pore structure of this pellet indicates that many new pores are formed as the oxygen potential increases. So pore formation probably cause the decrease in sintered density.

Fig. 3(a) and 3(b) show line profiles of Gd concentration in hydrogen and in the 30CO_2-H_2 atmosphere. The profile of Gd concentration is more uniform in fig. 3(b) than in fig. 3(a). That suggests that small agglomerates of Gd_2O_3 dissolve easily in high oxygen potential. So it can be said that the homogeneity of Gd is enhaned by high oxygen potential .

The grain size of UO_2-Gd_2O_3 fuel is dependent on oxygen potential and powder types. Fig. 4 shows their relations. Grain size is larger in mixing method than in co-milling method. The grain size difference between powder types is large in low oxygen potential but the difference becomes small in higher oxygen potential. The grain size of UO_2-Gd_2O_3 fuel increases as the oxygen potential increases. It is generally agreed that grain growth is limited by pores, secondary phases, and microstructural inhomogeneity. If Gd concentration is not homogeneous, the interface between different Gd concentrations will act as a barrier to grain boundary movement. The high oxygen potential makes Gd concentration uniform, so naturally the high oxygen potential promotes grain growth.

When mixing ADU-UO_2 with Gd_2O_3 powder, grain structure is not homogeneous. Some areas have large grains and the other areas have very small grains. Such inhomogeneity is not much mitigated even in the highest oxygen potential. When we analyze Gd ditribution across this microstructureare, Gd concentration is high in the area of small grains but low in the area of large grains. As mentioned earlier, the powder compact made by mixing ADU-UO_2 with Gd_2O_3 powders has Gd-rich region and Gd-depleted region. So this inhomogeneity of sintered pellet seems to be derived from that of the powder compact. It is supposed that Gd does not diffuse a long distance during sintering. Thus it is thought that a long range homogeity of Gd can be attained by powder mixing rather than by sintering.

It has been found out that sintered density and grain size are significantly influenced by powder types and oxygen potential. These sintering variables should be properly chosen to fabricate high quality UO_2-Gd_2O_3 fuel. UO_2-5wt%Gd_2O_3 fuel using co-milled powder has the sintered density of 96% in hydrogen atmosphere. But grain size is too small to be accepted. As oxygen potential increases, sintered density decreases but grain size increases. So when the ratio of CO_2 to H_2 gas reach 0.3, grain size larger than 6 ㎛ is obtained with sintered density a little sacrificed. Consequently both density and grain size are within acceptable range.

4. Conclusions

Three types of powders ; 1) mixing of UO_2 ex-AUC with Gd_2O_3 powder, 2) mixing of UO_2 ex-ADU with Gd_2O_3 powder, 3) co-milling of UO_2 and Gd_2O_3 powders. and four sintering atmospheres have been investigated to study their effects on the properties of UO_2 -Gd_2O_3 fuel pellets. Following conclusions can be made:

(1) Powder types dominantly influences sintered density. Adequate density is obtained only by co-milling method, Other mixing methods yield the density below 94%. Powder types also determines a long range homogeneity of Gd.

(2) The oxygen potential of sintering atmosphere has a great effect on grain size. As the oxygen potential increases, the grain size increases but sintered density decreases. A short range homogeneity of Gd is enhanced by high oxygen potential.

(3) The optimization of sintering variables is necessary to fabricate high quality UO_2 -Gd_2O_3 fuel pellets.

References

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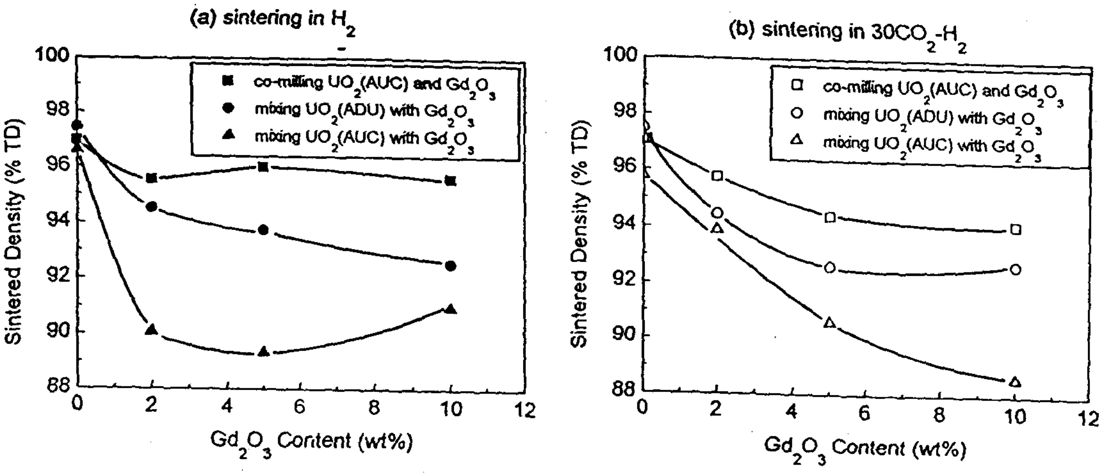

Fig.1. Sintered density of UO_2-Gd_2O_3 pellets as a function of Gd_2O_3 content.

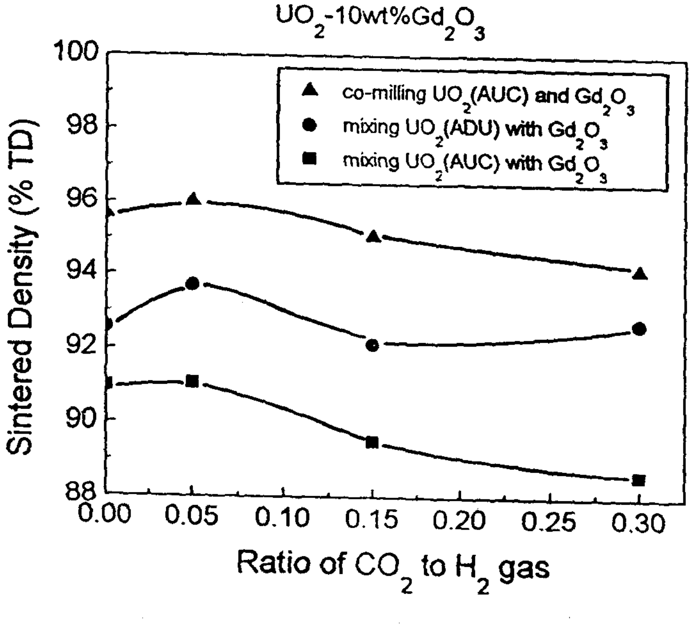

Fig.2. Dependence of sintered density of UO_2-10wt%Gd_2O_3 on oxygen potential.

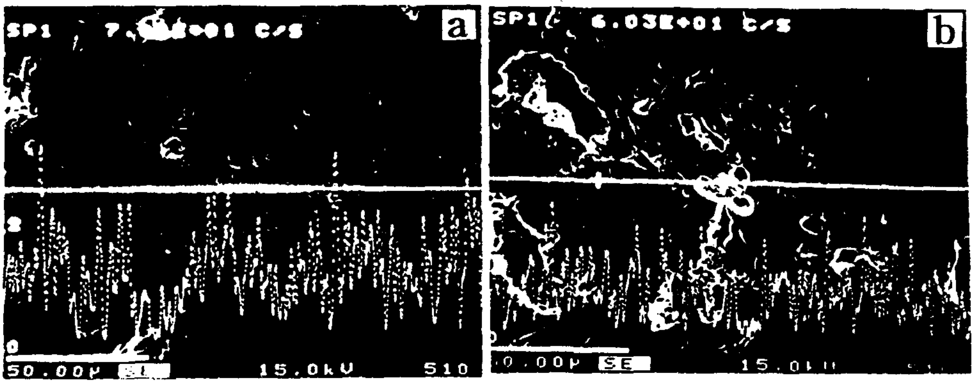

Fig.3. Profile of Gd concentration in UO_2-10wt% Gd_2O_3 fuel (AUC-UO_2 mixing) sintered in
(a) hydrogen, (b) CO_2/H_2=0.3

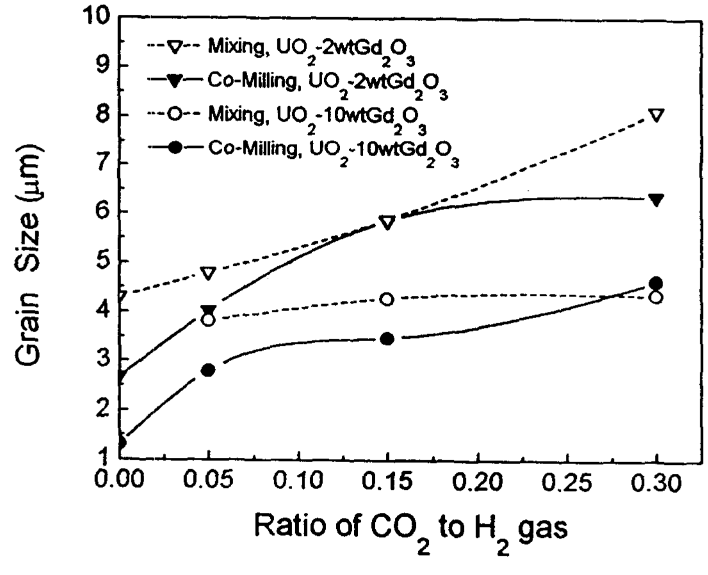

Fig.4. Dependence of grain size on oxygen potential and powder types