

<Technical Note>

RECYCLING PROCESS OF U_3O_8 POWDER IN $MnO-Al_2O_3$ DOPED LARGE GRAIN UO_2 PELLETS

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The effect of various process variables on the powder properties of recycled U_3O_8 from $MnO-Al_2O_3$ doped large grain UO_2 pellets and the effect of those recycled U_3O_8 powders on the sintered density and grain size of $MnO-Al_2O_3$ doped large grain UO_2 pellets have been investigated. The evolution of morphology, size, and BET surface area of the recycled U_3O_8 powders according to the respective variation of the thermo-mechanical treatment variables of oxidation temperature, powder milling, and sequential cyclic heat treatment of oxidation and then reduction was examined. The correlation between the BET surface area of recycled U_3O_8 powder and the sintered pellet properties of $MnO-Al_2O_3$ doped pellets showed that the pellet density and grain size of doped pellets were increased and then saturated by increasing the BET surface area of the recycled U_3O_8 powder. The density and grain size of the pellets were maximized when the BET surface area of the recycled U_3O_8 powder was in the vicinity of $3m^2/g$. Among the process variables applied in this study, the cyclic heat treatment followed by low temperature oxidation was a potential process combination to obtain the sinter-active U_3O_8 powder.

KEYWORDS : Doped Large Grain Pellet, Recycling Process, $MnO-Al_2O_3$, U_3O_8 Powder

1. INTRODUCTION

The recent development of fuel pellets for light water reactors is mainly focused on the large grain UO_2 pellets which could enhance pellet-cladding-interaction (PCI) behavior, because a large grain pellet is expected to enhance fuel plasticity at an elevated temperature of transient operation as well as the fission gas retention capability [1-8]. There are various process parameters of sintering to increase the grain size of UO_2 pellets. Among those, additives doping technology has been widely studied due to its effectiveness at increasing grain size and compatibility with an industrial pellet fabrication process [5-8]. MnO and Al_2O_3 doped UO_2 pellets are being developed as a potential candidate for PCI solution by KAERI [6]. Previous out-of-pile test results [6] showed that the grain size and softness of UO_2 pellets could be enhanced by doping Mn and Al in UO_2 . The average grain size of the UO_2 pellets could be enlarged by more than $50 \mu m$ by adding 1000 ppm of $MnO-Al_2O_3$ additives mixture. This grain size is about five times larger than that of conventional un-doped UO_2 pellets. A compressive creep test for this pellet at an elevated temperature to simulate operational transient conditions showed relatively fast visco-plastic deformation.

In a commercial UO_2 fuel pellet manufacturing process, defective UO_2 pellets or scraps are produced and they should be reused. A common recycling method is to oxidize them in air at about $450^\circ C$ to make recycled U_3O_8 powder, which is then added to UO_2 powder [9]. In this recycling process, however, addition of U_3O_8 to UO_2 leads to a drop in the pellet density, impeding grain growth and leading to the formation of undesired grape-like pore segregates because the recycled U_3O_8 powder has a larger particle size, small BET surface area, and thus poor sinter-ability compared to the raw UO_2 powder. This adverse effect of recycled U_3O_8 powder on UO_2 pellet properties may become more severe in large grain sized UO_2 pellets. Recycled U_3O_8 powder properties such as surface area and particle size strongly depend on the initial grain size of the UO_2 pellets because pulverization of UO_2 pellets to U_3O_8 powder initiates along the grain boundary during the oxidation. So, the particle size of recycled U_3O_8 obtained from Mn and Al doped large grain UO_2 pellets is expected to be much larger than that from conventional UO_2 pellets.

In this study, we have focused on the recycling process parameters for Mn-Al doped large grain pellets, which could provide U_3O_8 powder with a large BET surface area and small particle size. There have been several works

that dealt with the heat treatments of recycled U₃O₈ powder to improve its sintering properties [10-13]. It is known that a lowering oxidation temperature, mechanical ball-milling, and sequential cyclic heat treatment of oxidation and then reduction could enhance the recycled U₃O₈ powder properties. We investigate the effect of those process variables on the properties of U₃O₈ powder obtained from Mn-Al doped large grain UO₂ pellets. We further examine the effect of recycled U₃O₈ powders obtained by adopting various processes on the sintered pellet density and grain size of Mn-Al doped large grain UO₂ pellets. The optimal combination of process variables is also a part of the present work.

2. EXPERIMENTAL

2.1 Fabrication of U₃O₈ Powders to Investigate Morphology Evolution According to Process Variables.

The pure UO₂ pellets and 950ppm of Mn and 50ppm of Al doped UO₂ pellets were prepared by a conventional sintering process [6]. The green pellets were sintered at 1730°C for 4h in a dry hydrogen atmosphere. The Mn-Al doped UO₂ pellets will be denoted as reference UO₂ pellets. The average grain size and pellet density of reference UO₂ pellets were 52µm and 10.78g/cm³, respectively. U₃O₈ powders were prepared by heating those pure UO₂ pellets and reference UO₂ pellets in air for 24hrs at 325°C and 450°C, respectively. The UO₂ pellets were fully oxidized to U₃O₈ single phase after the heat treating as shown in the previous work [13]. For convenience, we will refer to the above two U₃O₈ powders obtained from the reference UO₂ pellets as ‘raw U₃O₈ powders’, hereafter. In order to reduce the particle size and thereby increase the BET surface area of raw U₃O₈ powders further, those powders were then either ball-milled or cyclic heat treated. In the ball-mill procedure, the raw U₃O₈ powders, zirconia balls, and ethanol were blended in a zirconia jar and then milled for 1, 3, or

5 hrs. In the case of cyclic heat treatment, the raw U₃O₈ powders were reduced at 700°C [14] and then re-oxidized at the same temperature of initial oxidation, repetitively. The cyclic heat treated U₃O₈ powders were obtained by repeating the cyclic heat treatment 1, 2, or 3 times. Table 1 lists the batch name and corresponding process variables of U₃O₈ powders prepared in this work. The BET surface areas of the U₃O₈ powders were measured with a BET surface area analyzer and powder morphology was examined by SEM.

2.2 Fabrication of Recycled U₃O₈ Powders Containing UO₂ Pellets.

Five and 10wt% of prepared U₃O₈ powders in table 1 were added to the MnO-Al₂O₃ additives doped ADU-UO₂ powders. The total amounts of Mn and Al in the powder mixture were respectively controlled to be 950ppm and 50 ppm in weight while considering the Mn-Al contents in recycled U₃O₈ powders. After mixing with a tumbling mixer, the powder mixture was pressed into green pellets at 300 MPa. The green pellets were sintered at 1730°C for 4h in H₂ gas atmosphere. The sintered density of the UO₂ pellets was measured by the water immersion method. The pellets were sectioned axially, ground and polished. The polished pellets were thermally etched at 1290°C in carbon dioxide gas in order to examine their grain boundaries. The grain structures were examined by an optical microscope and the grain size was determined by the linear intercept method.

3. RESULTS AND DISCUSSION

3.1 Effect of the Process Variables on the Powder Properties of the Recycled U₃O₈

Figure 1 shows SEM images of the U₃O₈ powders that were obtained by oxidizing the pure UO₂ pellets and Mn-

Table 1. Process Variables Applied in this Study and Corresponding Batch Names

		Process variables					
Name	Oxidation temperature (°C)						
	325			450			
	LT			HT			
Name	Ball-milling Time (hrs)						
	1hr	3hr	5hr	1hr	3hr	5hr	
	LTB-1	LTB-3	LTB-5	HTB-1	HTB-3	HTB-5	
Name	Repetition of Cyclic heat treatment (times)						
	1	2	3	1	2	3	
	LTC-1	LTC-2	LTC-3	HTC-1	HTC-2	HTC-3	

Al doped large grain UO_2 pellets at 325°C and 450°C, respectively. The initial grain size of the standard UO_2 pellet was 9 μ m and that of the doped pellets was 52mm. It can be seen that the powder morphology was changed according to the variation of oxidation temperature and initial grain size of the pellets.

The effect of oxidation temperature on the U_3O_8 powder morphology during the oxidation of standard UO_2 pellets has been reported in the previous work [13]. The popcorn-like shape shown in Fig. 1(b) is a typical one for conventionally recycled U_3O_8 powder. However, when the oxidation temperature was lowered to 325°C, the U_3O_8 powder became more porous and fault-plan-like shaped particles were frequently observed, as shown in Fig. 1(a). The observed morphology difference between Figs. 1(a) and (b) was interpreted in terms of an oxidation kinetics change [13]. At high temperatures, oxygen can deeply diffuse into the pellet along the grain boundaries to form an intermediate phase of U_3O_7/U_4O_9 . The U_3O_8 precipitated on those intermediate phases causes the spallation of grains [15-22]. However, when the oxidation temperature is lowered, the formation of an intermediate phase follows the discrete-layer mechanism because of a similar diffusion rate of oxygen between the grain boundary and along the lattice. Consequently, the U_3O_8 phase shape is also expected to have a sheath-like shape as in Fig. 1(a).

In the case of HT powder (Fig. 1(d)), the U_3O_8 particle size was increased very much due to the large initial grain size of the UO_2 pellets. In addition, plate-like parallel cracks developed on the powder surface instead of equi-angular cracks. Since the large grain pellet has a reduced number of exposed grain boundaries on the pellet surface, parallel propagation of oxidized layers from the surface might dominate the oxidation kinetics and plate-like particles were produced. Particle size difference between the U_3O_8 from standard UO_2 pellets and that from large grain UO_2

pellets was greatly reduced when the oxidation temperature was lowered to 325°C (LT), as shown in Figs. 1(a) and (c). Both powders also showed similar morphology.

According to the previous work [13], the BET surface area of U_3O_8 powder could be increased up to 2 times when the oxidation temperature is reduced from 450°C to 325°C. The measured BET surface areas of the LT and HT were 1.29m 2 /g and 0.85m 2 /g, respectively. Therefore, this result also reveals that low temperature oxidation can increase BET surface area and decrease particle size of recycled U_3O_8 , simultaneously.

In order to reduce particle size of recycled U_3O_8 powders further, two kinds of follow up processes have been applied. Mechanical ball-milling is a common method to reduce the particle size by applying mechanical force to powders. Cyclic heat treating is a method to utilize the mechanical stresses induced during the oxidation and reduction phase transition. This approach has been applied in the DUPIC pellet fabrication process [23].

Fig.2 shows the particle morphology evolution according to the milling time when the ball mill process was applied to the LT powder. The LT powders were crushed into small particles by the ball mill process. The BET surface area changes after ball milling are shown in Fig. 3(a). Even though the particle size was reduced by ball milling, milling of the oxidized powder seems to be ineffective to increase the BET surface area. The BET surface area of LTB powders did not change regardless of milling time. The BET surface areas of HTB powders even decreased after milling. Since the U_3O_8 particle was fragmented during the milling and there is almost no probability that fragmented particles physically re-bond during the milling, these results may suggest the possibility that fragmented U_3O_8 particles were agglomerated during the milling. However, the agglomerate size is speculated to be small because large sized agglomerates were not observed in Fig. 2.

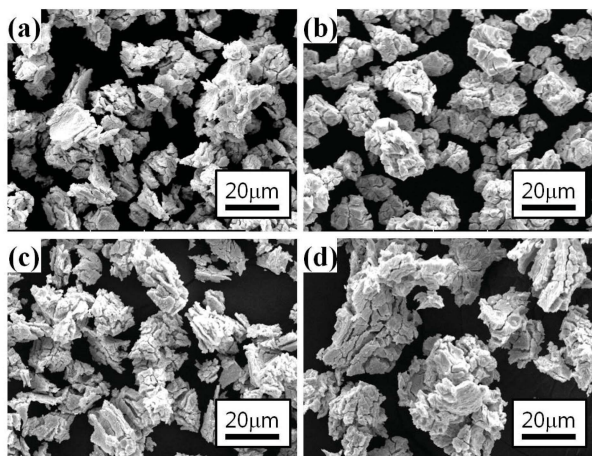


Fig. 1. SEM Powder Morphologies of U_3O_8 Powders Obtained by Oxidizing UO_2 Pellets at (a) 325°C and (b) 450°C and Mn-Al Doped UO_2 Pellets at (c) 325°C and (d) 450°C

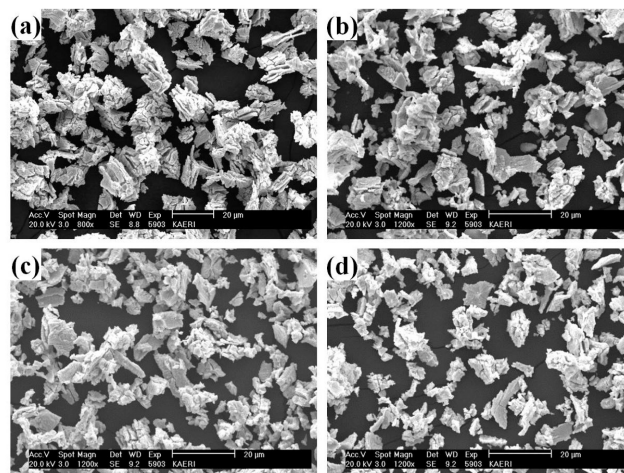


Fig. 2. SEM Powder Morphologies of LT Powders According to Milling Time : (a) None, (b) 1h, (c) 3h, (d) 5h

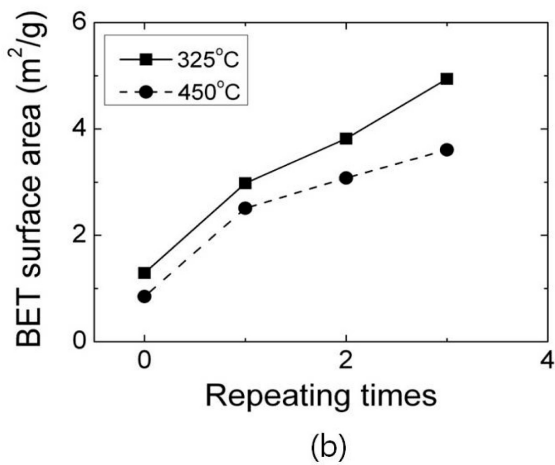
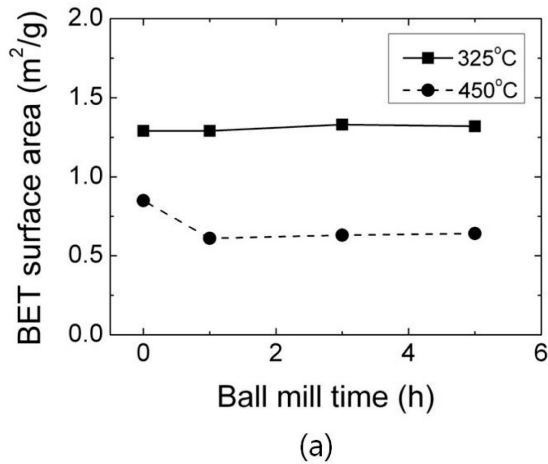


Fig. 3. BET Surface area Changes of the Oxidized U₃O₈ Powders According to (a) Ball Mill and (b) Cyclic Heat Treatment

Fig. 4 shows the particle morphology evolution of the U₃O₈ powder to which cyclic heat treatments were applied. During the cyclic heat treating of reducing and then oxidizing, the U₃O₈ powders were fragmented to very tiny particles due the thermal stresses. The BET surface area changes of U₃O₈ powders after the cyclic heat treatment are shown in Fig. 3(b). The graph shows that the BET surface area of LTC and HTC powders increased significantly. The BET surface area was increased with repetition and enlarged up to 3 times after the cyclic heat treating.

3.2 Effect of the Recycled U₃O₈ Powders on the Sintered Pellet Properties of UO₂

The effect of prepared U₃O₈ powders on UO₂ pellet properties has been tested. As shown in Table 1, fourteen kinds of U₃O₈ powders having different BET surface areas were prepared. Five and 10wt% of each U₃O₈ powder was added to Mn-Al doped UO₂ powders and sintered pellets

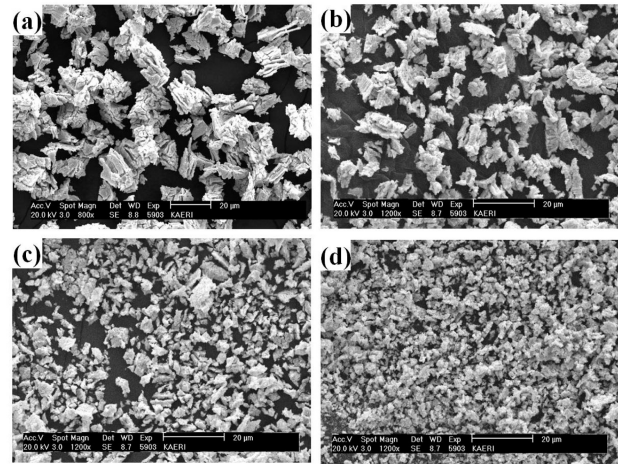


Fig. 4. SEM Powder Morphologies of LT Powders According to Cycle Number of Reducing and then Oxidizing : (a) None, (b) 1 Cycle, (c) 2 Cycles, (d) 3 Cycles

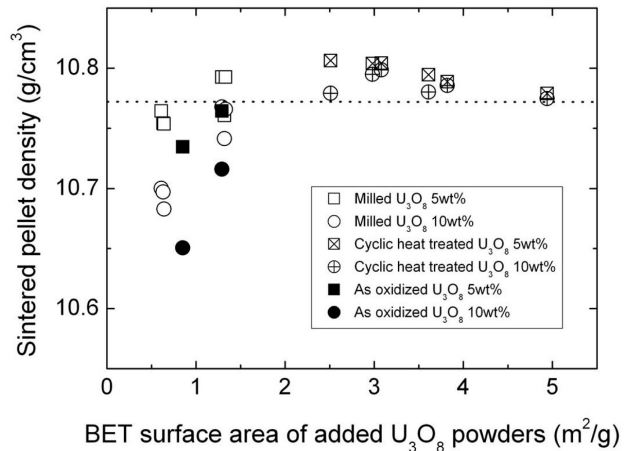


Fig. 5. The Correlation between Density of Recycled Pellets and the BET Surface area of Recycled U₃O₈ Powders

of those powder mixtures were fabricated through a conventional UO₂ pellet sintering process. Densities and grain sizes of those fabricated pellets were measured and compared to examine the effect of each recycled U₃O₈ powder on UO₂ pellet properties. Hereafter, the recycled U₃O₈ added doped UO₂ pellets will be referred to as the ‘recycled pellets’.

Fig. 5 shows the correlation between the sintered density of recycled pellets and the BET surface area of recycled U₃O₈ powders. The dotted line in the figure denotes the density of a reference pellet, which is a doped pellet without recycled U₃O₈. The density of the conventionally recycled pellet (HT containing pellet) is far below than that of the reference pellet. The density drop in 10 wt% of HT containing pellet is about 1% of the theoretical density

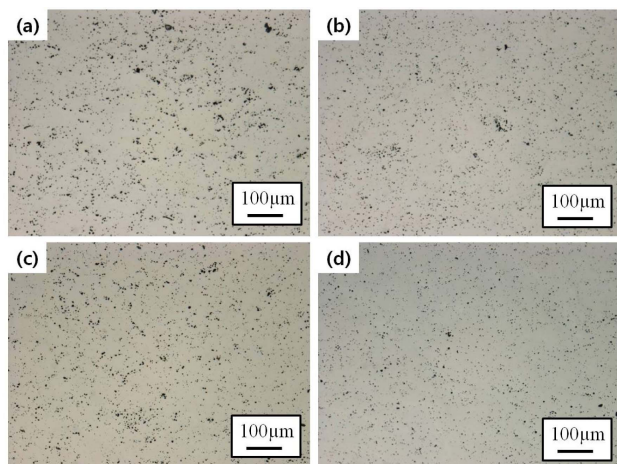


Fig. 6. Pore Structure of the Mn-Al Doped Pellets Containing 10wt% of Recycled U_3O_8 Powders : (a) HT, (b) LT, (c) LTB-5 (d) LTC-1

of UO_2 . This value is similar to that observed in the recycling process of standard UO_2 pellets. The density drop was slightly mitigated by substituting LT for HT powder, which has a higher BET surface area than that of HT. In previous works, it was noted that low-temperature oxidized U_3O_8 powder with a high BET surface area was effective in mitigating the density drop of the standard UO_2 pellets. Therefore, the observed density enhancement in LT containing UO_2 pellets is due to the enhancement of the BET surface area.

In the LTB and HTB containing recycled pellets, the pellet density was also increased in spite of the fact that the BET surface area of LTB and HTB was not enhanced. It is known that small particle size is also effective in increasing the sintered pellet density. Therefore the particle size reduction in LTB and HTB seems to be responsible for the observed density increase in these pellets.

The cyclic heat treated U_3O_8 powders of LTC and HTC were the most effective to mitigate the density drop in recycled pellets. Pellets of higher density than the reference pellet could be obtained by adding LTC and HTC powders. However, recycled U_3O_8 with a too high BET surface area was unfavorable because the pellet density was saturated or even slightly decreased when the BET surface area exceeds $3m^2/g$. So the density data shown in Fig. 5 reveals that there is an optimal range of BET surface area of recycled U_3O_8 to effectively mitigate the density drop. Fig. 6 shows the comparison of typical pore structures between 10wt% of HT, LT, LTB-5, and LTC-1 powder added recycled pellets. Large size pores and pore clusters were frequently observed in low density pellets, whereas tiny pores were homogeneously distributed in the high density pellets.

Fig. 7 shows the grain size variation in the recycled pellets according to the BET surface area of U_3O_8 . The

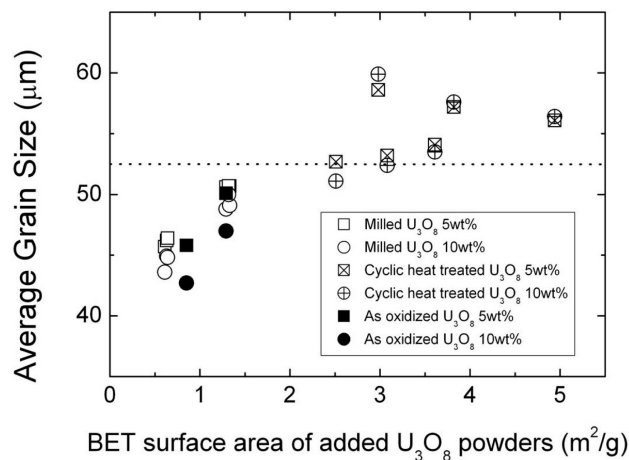


Fig. 7. The Grain Size Variation in the Recycled Pellets According to the BET Surface area of U_3O_8 Powders

dotted line in the figure indicates the average grain size of the reference UO_2 pellet. As with the sintered pellet density, the grain size of the recycled pellets was abruptly decreased when the conventional recycling process was applied. The grain size of the 10wt% of HT containing recycled pellet was decreased by 20%, as denoted by the filled-circle in Fig. 7. Since the main objective of doped pellet development is to increase the grain size of the pellets, significant declines in grain size during the recycling process should be avoided. The grain size reduction was mitigated by replacing the HT powder with LT powder. The grain size gap between the reference pellet and the LT containing pellet could be lessened by up to 10%.

As shown in Fig. 5, the ball-milled powders of LTB or HTB could mitigate the density drop in the recycled pellets. In contrast to density, however, Fig. 7 reveals that the grain size of doped UO_2 pellets was almost unchanged by adopting LTB or HTB. In the previous section, we mentioned the possibility of powder agglomeration in LTB and HTB. Indeed, the large size pores or pore clusters observed in Fig. 6(c) support the possibility of small agglomerations. The formation of large pores or pore clusters hinders the grain growth of a pellet. Therefore, the ball mill process was not an appropriate way to restore the grain size reduction of recycled UO_2 pellets.

The grain size of the doped pellets could be remarkably enhanced by LTC or HTC powders. Fig. 7 reveals that grain size even became larger than that of the reference pellet when the LTC or HTC powders were added. It was increased up to 20% larger than that of the reference pellet when 10wt% of LTC-1 was added to a UO_2 pellet. It is considered that recycled U_3O_8 powders act as an obstacle for densification and grain growth during the sintering of UO_2 pellets because the morphology of recycled U_3O_8 is quite different from that of raw UO_2 powder and normally

powder properties are degraded in recycled U_3O_8 powder. Our intention was to minimize the density and grain size drop in the recycled pellet by controlling the properties of U_3O_8 powders. So, the grain size enlargement observed in LTC and HTC containing UO_2 pellets surpasses our expectations and clearly means that the LTC and HTC promote densification and grain growth of Mn-Al doped UO_2 pellets.

At present, it is not clear how LTC or HTC enhance densification and grain growth of recycled UO_2 pellets. Nevertheless, we could consider the following possible reasons. It has been shown that crystalline particles having a larger particle size than raw powder could be a seed for abnormal grain growth. The particle size of the recycled U_3O_8 powder may be larger than that of raw UO_2 powder. The curvature difference between uniformly imbedded large crystalline grain originated from recycled U_3O_8 powder and surrounding fine grains originated from raw UO_2 powder could make the large grain grow by consuming the surrounding fine grains. Then a low number of grains can be selectively grown and those grains increase the maximum driving force for grain growth which is related with the size difference between the smallest and largest grains [11, 24, 25].

The pre-existing additives elements in LTC or HTC can be an alternative potential cause for grain size enlargement [24, 25]. The recycled U_3O_8 powder already contains additives because it was obtained by oxidizing the doped UO_2 pellets in which the additives are expected to be homogeneously distributed. The additives in the recycled U_3O_8 particle may operate earlier than additives in the UO_2 mixture because the additives in the UO_2 matrix should be dissolved into or along the UO_2 grain before it could

act as a grain growth promoter. Therefore, the crystalline UO_2 particles, originating from recycled U_3O_8 particles and containing additives, can grow fast at the early stage of sintering and may take a role of seeds for abnormal grain growth. However, further investigation should be made to explain the observed grain growth behaviors.

Another interesting thing in Fig. 7 is that the grain size difference between 5wt% and 10wt% of U_3O_8 added UO_2 pellets was diminished when LTC and HTC powders were used. In a commercial pellet fabrication process, the amount of recycled U_3O_8 added in UO_2 could be varied depending on the defective pellet generating rate. Then the pellet properties of density and grain size could be fluctuated according to the variables of U_3O_8 amount as shown in Fig. 5 and Fig. 7. In a quality control aspect, the pellet properties should be controlled consistently. So, other process parameters such as sintering temperature, sintering time, or pore former amount may need to be changed to compensate for the property changes caused by the difference of recycled U_3O_8 contents. Such variation of process parameters could be a big burden in commercial processes for mass production. Therefore, in commercial aspect, almost constant grain size and pellet density regardless of U_3O_8 content is a very beneficial advantage of the LTC and HTC containing pellet fabrication process because it can provide a wide flexibility in adjusting the process parameters. Fig. 8 shows the comparison of typical grain structures between 10wt% of HT, LT, LTB-5, and LTC-1 powder added UO_2 pellets. The number of pores and pore size is noticeably reduced and grain size is greatly enlarged in LTC-1 containing pellets.

Fig. 9 shows the overall correlation between pellet density and grain size for the sintered pellets observed in this study. The grain size of pellets proportionally increased

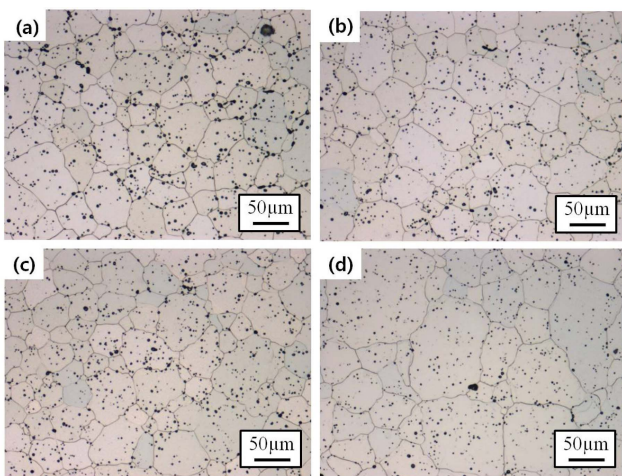


Fig. 8. Grain Structure of the Mn-Al Doped Pellets Containing 10wt% of Recycled U_3O_8 Powders : (a) HT, (b) LT, (c) LTB-5 (d) LTC-1

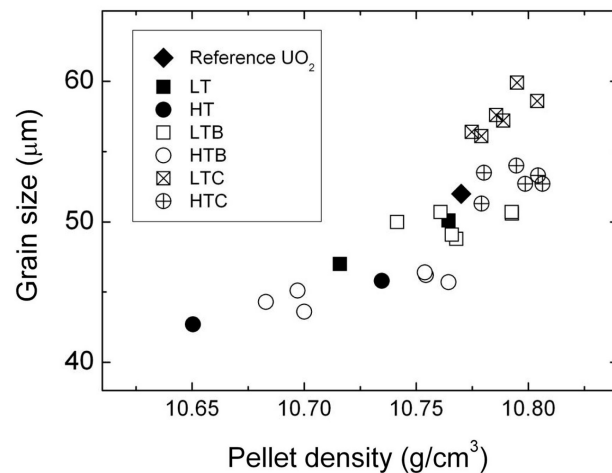


Fig. 9. The Overall Correlation between Pellet Density and Grain size for the Sintered Pellets

with increasing pellet density even in the high density region. This fact implies that porosity is an important parameter that affects the grain growth at the last stage of sintering. The cyclic heat treatment followed by low temperature oxidation was revealed as the most effective combination to enhance both grain size and density of pellets during the recycling process of Mn-Al doped large grain UO₂ pellets.

4. CONCLUSIONS

In the current work, the effect of recycled U₃O₈ powder properties on the property of MnO-Al₂O₃ doped large grain UO₂ pellets was investigated. The recycled U₃O₈ powder properties of morphology, the BET surface area, and size were controlled by applying low temperature oxidation, ball-milling, and/or cyclic heat treatment. It was found that the cyclic heat treating process could considerably increase the BET surface area of the recycled U₃O₈ powder. The correlation between the BET surface area of recycled U₃O₈ powder and the sintered pellet properties of MnO-Al₂O₃ doped pellets showed that the pellet density and grain size of doped pellets were increased and then saturated by increasing the BET surface area of the recycled U₃O₈ powder. The density and grain size of doped pellets were maximized when the BET surface area of recycled U₃O₈ powder was in the vicinity of 3m²/g. By addition of 10wt% of recycled U₃O₈ with a BET around 3m²/g, the grain size of the doped pellet was increased up to 20% larger than that of the reference doped pellet without recycled U₃O₈.

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