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Ball-milling Effect on the Sinterability of the UO_2 ex-AUC Powder

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AUC 공정으로 변환된 UO_2 분말의 소결성에 미치는 Ball-milling 효과

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Abstract

In order to investigate the ball-milling effect on the property changes of UO_2 ex-AUC powder, the sinterability of ball-milled powder was studied in terms of the ball-milling time. Spherical shape was found to be kept for ball-milled UO_2 powder and the particle size showed a bimodal distribution, which seems to have a higher packing ratio compared with those having monomodal gaussian distribution. The increase of sintered density of the ball-milled UO_2 powder is assumed to be mainly affected by the packing ratio, which increases with longer ball-milling time. It is confirmed that the sinterability of UO_2 ex-AUC powder is improved by the ball-milling process.

요 약

AUC 공정으로 변환된 UO_2 분말의 ball-milling 효과를 관찰하기 위하여 ball-milling 시간에 따른 소결성을 연구하였다. Ball-milling된 분말은 구형화 형태를 보이며, 그의 입자 크기 분포는 bimodal 양상을 나타냈다. 분말크기 분포가 bimodal인 경우, 그의 packing ratio가 monomodal gaussian 분포에 비해 높다. Ball-milling된 분말의 소결밀도 증가는 packing ratio에 의한 영향이 컸으며, ball-milling 시간이 길어질수록 packing ratio는 증가 하였다. AUC 공정으로 변환된 UO_2 분말은 ball-milling을 함으로써, 그의 소결성이 향상됨을 확인하였다.

1. Introduction

For the fabrication of nuclear fuel pellets, the manufacturing processes, including powder treatment, compacting, sintering and centerless grinding process are usually selected and performed as continuous processes to make close control over the pellet properties.[1,2] The characteristics of as-received UO_2 powder and the conditions of various manufacturing processes, especially the powder treatment process,[3] strongly affect the pellet characteristics.

In order to make the mixed powder with enriched UO_2 or other oxide (Gd_2O_3, CeO_2, MgO etc), when following above steps, the intensive mechanical milling is normally utilized to improve the homogeneity of the mixed powder.[5,6] It is necessary to understand clearly the effect of the intensive mechanical milling effect on the UO_2 ex-AUC powder properties.

Therefore, the effect of powder treatment process on the UO_2 powder properties should be studied prior to the investigation of pellet characteristics changes by other manufacturing process factors.

The KAERI(Korea Atomic Energy Research Institute) has been manufacturing natural UO_2 powder by the AUC (Ammonium Uranyl Carbonate) conversion process. Generally the UO_2 ex-AUC powder has larger particle size and better flowability compared with the UO_2 powder by other conversion processes.

The purpose of present work is to investigate the physical property changes of the UO_2 ex-AUC powder from the ball-milling process, and to find out the effect of the property changes on the sinterability.

2. Experimental Procedure

A rotation speed of ball-milling cylindrical jar was first selected. Usually, the optimum condition can be attained when gravitational force is balanced with centrifugal force. The opitmal rotation speed for cylindrical jar has been estimated by Sundrica[7] as follows:

$$No = 32/d^{1/2},$$

where d is outer diameter of the jar in meters, and No is the optimal speed of rotation in rpm.

According to this equation, 80 rpm was chosen as an optimal speed and the powder sample was charged about 20% of inner volume of the jar.

To investigate the UO_2 ex-AUC powder property changes in terms of ball-milling time, 2, 4, 6, 8 and 24 hours of ball-mlling time were selected.

Compacting of green compact whose weight is about 20g was performed by double acting hydraulic press(France, Billaud co., die cavity diameter: 14.75mm) with the pressure of 4.83 ton/cm^2. Sintering was carried out at 1700°C in H_2 atmosphere for 4 hours.

Ball-milled UO_2 ex-AUC powders were examined in terms of specific surface area by BET, tapping density and bulk density by Scott volumetry. The particle size and its distribution were measured by laser particle size analysis system, and their morphologies were observed by SEM(Scanning Electron Microscope).

The sintering behaviours of different powder samples were investigated in terms of the linear shrinkage and shrinkage rate, i. e., sintering rate by dilatometric analysis.

3. Results and Discussion

A. Ball-Milling Effect

The morphologies of as-received UO_2 ex-AUC powder and after ball-milling are shown in Fig. 1. As-received powder shows round shape which has micropores within particles but the ball-milled powder after 24 hours of ball-milling time consists of large granular shape which is agglomerated from the individual fine particles.

The physical properties of the ball-milled UO_2 ex-AUC powder after 2, 4, 6, 8 and 24 hours of ball-milling are shown in Table 1. The average particle size of ball-milled powder decreased with the longer ball-milling time up to 8 hours, but it increased again after 24 hours of ball-milling. This phenomenon seems to be caused by the agglomeration of the ball-milled fine powders,as shown in Fig. 1-b. The specific surface area of ball-milled powder increased with ball-milling time up to 6 hours,whereas, in case of the powder after 6 hours of ball-milling, it hardly changed. This is mainly caused by the elimination of micropores within particles.

According to Gray and Beddow[8], the ratio of the tapping to the bulk density (it is called the Hausner ratio) will be slightly above unity for a spherical powder and for the best packing result. In this test, the Hausner ratio of ball-milled UO_2 ex-AUC powder is close to unity with the longer ball-milling time, as shown in Table 1. That means the UO_2 ex-AUC powder becomes more spherical shape and has a higher packing ratio with the longer ball-milling time.

Fig. 2 shows the particle size distribution of as-

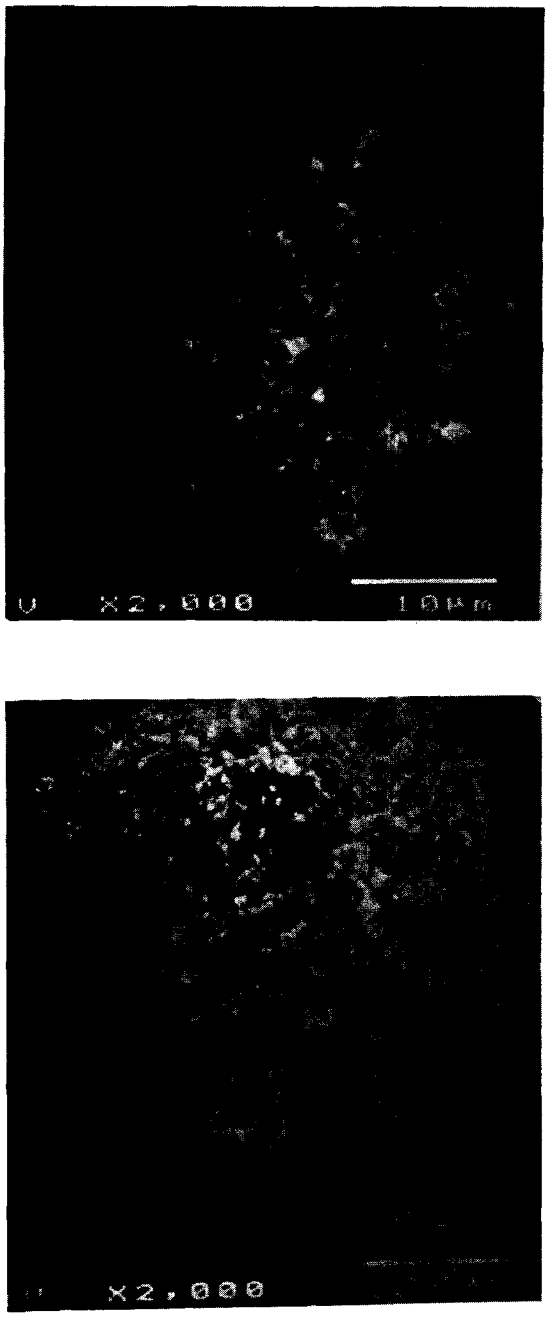

Fig. 1. Typical Morphologies of UO_2 ex-AUC Powder Before and After Ball-Milling

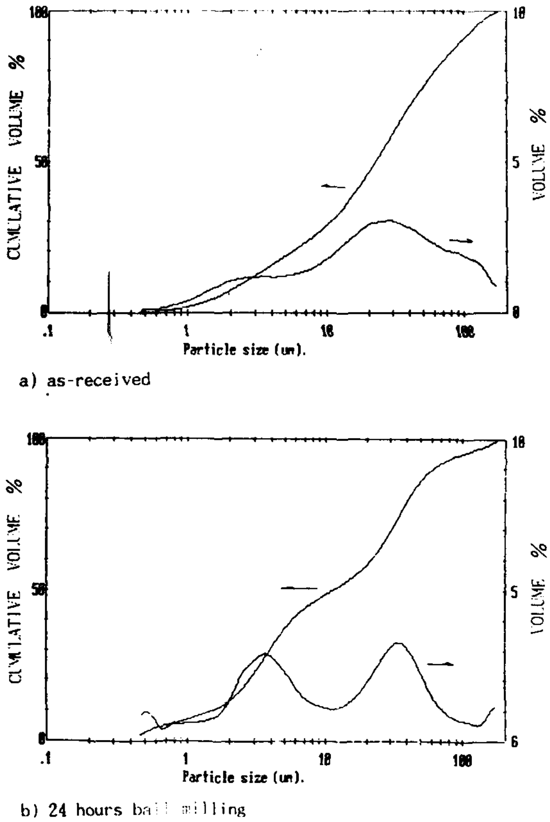

Fig. 2. Particle Size Distribution of UO_2 ex-AUC Powder Before and After Ball-Milling

received UO_2 ex-AUC powder and that of 24 hours ball-milled UO_2 ex-AUC powder. The particle size of as-received UO_2 ex-AUC powder showed a monomodal gaussian distribution,whereas that of 24 hours ball-milled UO_2 ex-AUC powder showed a bimodal type distribution. Accordingly, it can be assumed that a bimodal type distribution of partcle size has a higher packing ratio compared with a monomodal gaussian distribution.

B. The Characteristics of Green Compacts and Sintered Pellets

Fig. 3 shows the green and the sintered density as a function of ball-milling time. The green and sintered density increased with ball-milling time. Also, the sintered density increased with green density.

When we compare the Hausner ratio with the

Table 1. Physical Property Changes of UO_2 ex-AUC Powder with Ball-Milling Time

특 성	Tapping density (g/cc)	Bulk density (g/cc)	particle size (median, μm)	Hausner ratio (tap. den./ bulk den.)	specific surface area (m^2/g)
As received UO_2 ex-AUC	2.78	1.95	23.64	1.42	4.51
Ball-milling time					
2 hrs	2.78	2.21	19.88	1.26	5.01
4 hrs	2.74	2.15	18.56	1.27	5.06
6 hrs	2.79	2.35	10.81	1.18	5.48
8 hrs	2.80	2.43	7.16	1.15	5.32
24 hrs	2.73	2.43	11.19	1.12	5.42

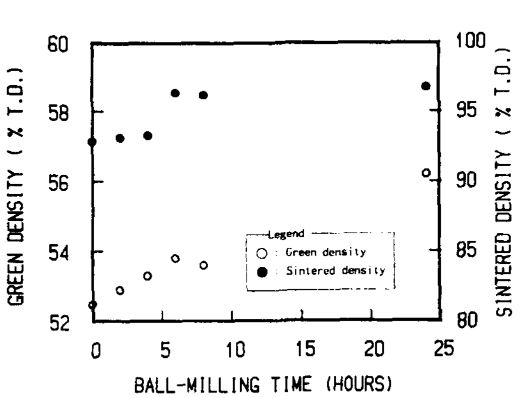

Fig. 3. The Green and Sintered Density as a Function of Ball-Milling Time

green and sintered density in relation to the ball-milling time(Table 1 and Fig. 3), the green and sintered density increase with the decrease of the Hausner ratio to the unity, i. e., the green and sintered density increase with packing ratio. Accordingly, it is confirmed that the sinterablity of ball-milled UO_2 ex-AUC powder can be improved rather by the packing ratio(the Hausner ratio) than by the specific surface area and the particle size.

The typical microstructures of sintered pellet of 2 and 24 hours ball-milled UO_2 ex-AUC powder are shown in Fig. 4 and 5. Although the grain sizes of sintered pellet were in the range of 3-5 μm regardless of ball-milling time, the number of its pores decreased and the size of its pores became smaller with ball-milling time.

Pore distribution of sintered pellet as a function of ball-milling time(2, 6 and 24 hours) is shown in Fig. 6. The pore distribution shifted to smaller size when ball-milling time became longer. From these results, it can be understood that the increase of sintered density by ball-milling process is caused by the pore elimination with the higher packing ratio.

C. Sintering Behaviour

The linear shrinkage and shrinkage rate of the green compacts in terms of ball-milling time are shown in Fig. 7 and 8. The shrinkage of green compacts started at 800°C, rapidly increased to 1100°C, and then the rate became slow, regardless of ball-milling time. But the amount of shrinkage of green compact increased with ball-milling time.

In Fig. 8, in case of 2 hours ball-milled UO_2 ex-AUC powder, the temperature of maximum sintering

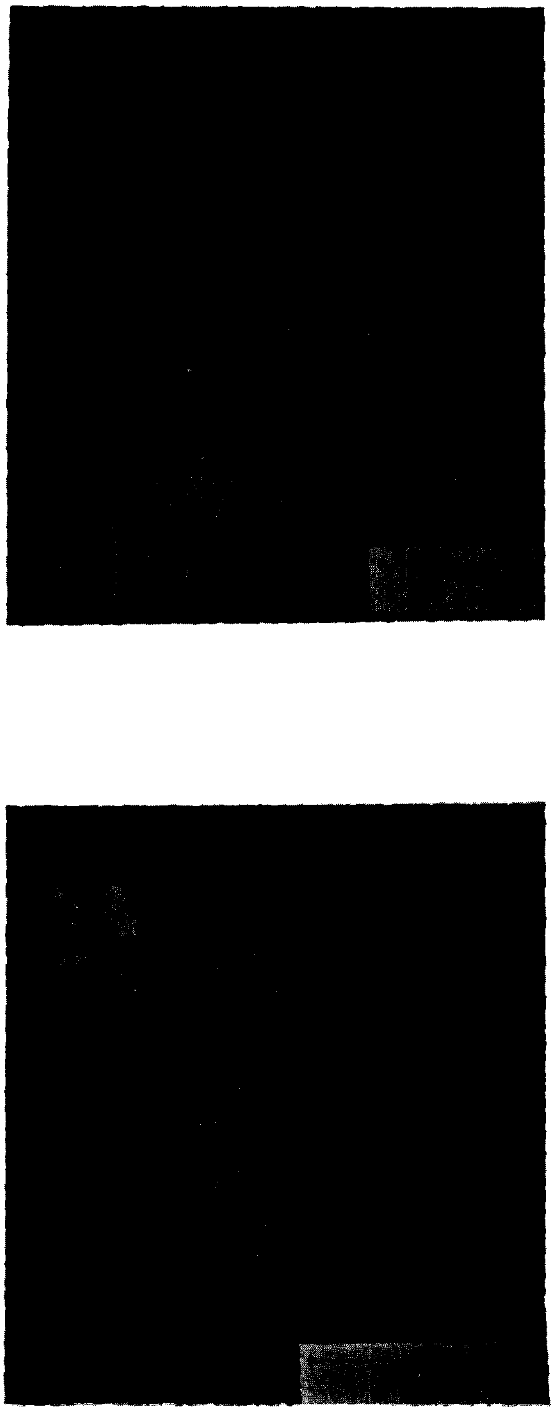

Fig. 4. Pore and Grain Microstructures of the UO_2 ex-AUC Sintered Pellet After 2 Hours Ball-Milling

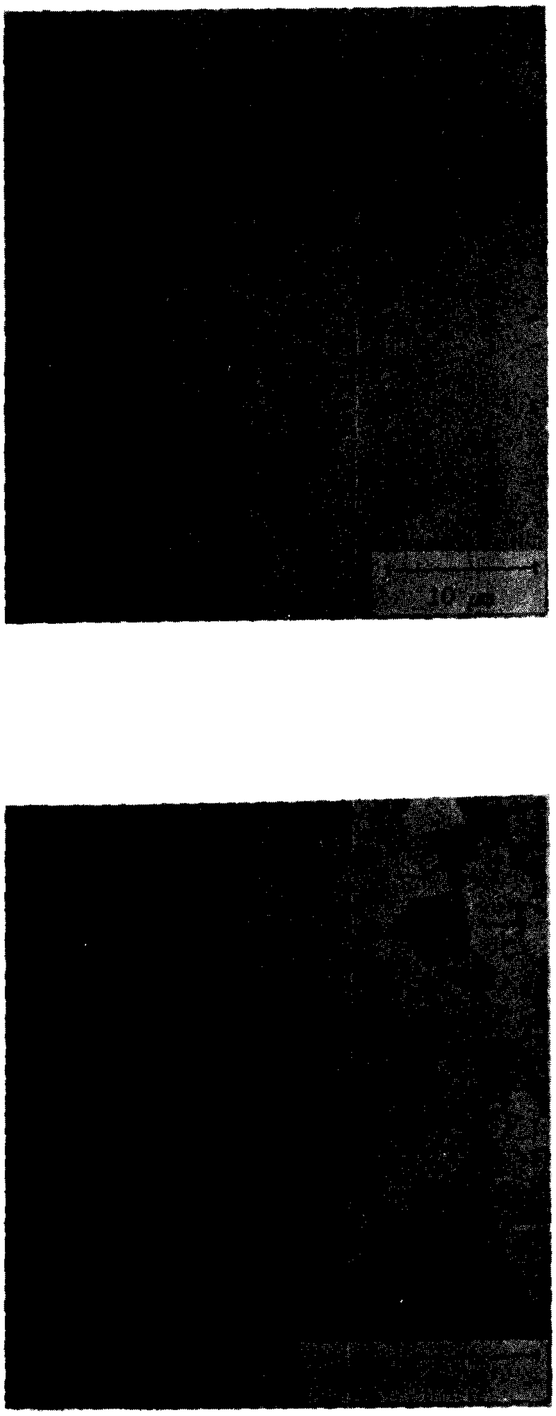

Fig. 5. Pore and Grain Microstructures of the UO_2 ex-AUC Sintered Pellet After 24 Hours Ball-Milling

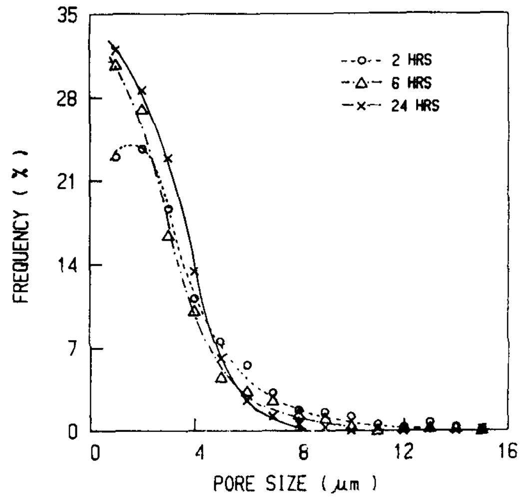

Fig. 6. Pore Size Distribution Changes of Sintered Pellet with Ball-Milling Time

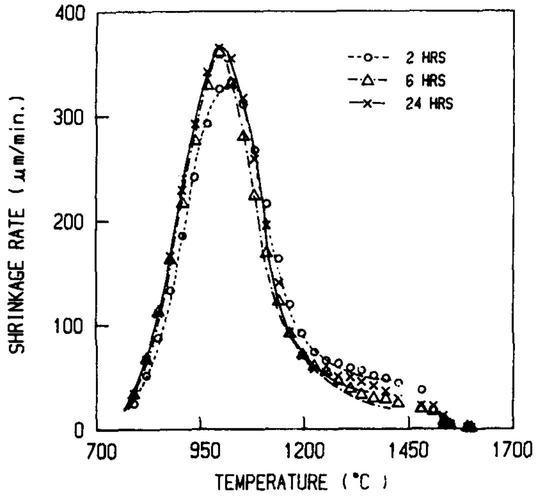

Fig. 8. Shrinkage Rate Changes with Ball-Milling Time

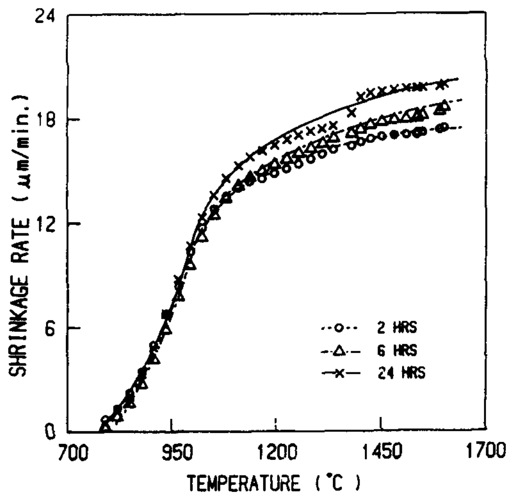

Fig. 7. Linear Shrinkage Changes with Ball-Milling Time

rate of green compact was about 1000°C, whereas it was lowered to about 980°C for the 6 and 24 hours ball-milled UO_2 ex-AUC powder. With the longer ball-milling time, the temperature of maximum sintering rate of green compact is lowered and its amount of shrinkage increases. It means the sinterability of the ball-milled UO_2 ex-AUC powder increases with ball-milling time.

This agrees well with the results of green compacts and sintered pellets. From the above results, the sinterability of UO_2 ex-AUC powder might be improved by ball-milling process.

4. Conclusions

1. The particle size of the ball-milled UO_2 ex-AUC powder showed a bimodal distribution,which seems to have a higher packing ratio compared with those having monomodal gaussian distribution.
2. The sintered density of ball-milled UO_2 ex-AUC powder increased with green density. This seems to be mainly affected by the packing ratio.
3. The sinterability of UO_2 ex-AUC powder might be improved by ball-milling process.

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