

## Effect of a Li<sub>2</sub>O Additive on the Sintering Behavior of UO<sub>2</sub> in the H<sub>2</sub> and CO<sub>2</sub> Atmospheres

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### ABSTRACT

The variation of the sintered density and grain size of UO<sub>2</sub> as a function of the Li<sub>2</sub>O amount and sintering atmosphere was observed. Li<sub>2</sub>O enhanced the grain growth of the UO<sub>2</sub> pellet in H<sub>2</sub>, but rather hindered it in CO<sub>2</sub> atmosphere. Grain size of the UO<sub>2</sub> and UO<sub>2</sub>-0.1 wt%Li<sub>2</sub>O pellets was, respectively, 8 μm and 100 μm at 1680°C in the H<sub>2</sub> atmosphere, and that of each pellet was, respectively, 24 μm and 17 μm at the same temperature in the CO<sub>2</sub> atmosphere. As-received Li<sub>2</sub>O powder, which had been composed of Li<sub>2</sub>O and LiOH, was converted to the Li<sub>2</sub>CO<sub>3</sub> phase after heating to 800°C in CO<sub>2</sub>. On the other hand, the Li<sub>2</sub>O and LiOH phases remained unchanged in H<sub>2</sub> atmosphere. In the H<sub>2</sub>, the as-received Li<sub>2</sub>O powder began to evaporate at about 1050°C and then about 20 wt% residue was left at 1500°C. But, most of the Li elements evaporated at 1500°C in the CO<sub>2</sub> atmosphere.

**Key words :** UO<sub>2</sub>, Li<sub>2</sub>O, Sintering, Evaporation, Grain size

### 1. Introduction

A recent trend in nuclear power generation is to improve the utilization of fuel. Research and development programs are currently being carried out in this respect for improving its economy and safety, namely; a high and extended burn-up. Fission gas release is a major concern in a high burn-up fuel and many studies have been conducted in order to increase the grain size in the pellet which is expected to prolong the diffusion path of the fission gas to the grain boundaries.<sup>1-3)</sup> Several researchers have studied the effects of additives on the grain growth of UO<sub>2</sub> and shown that Nb<sub>2</sub>O<sub>5</sub>,<sup>4,7)</sup> TiO<sub>2</sub>,<sup>5,8-11)</sup> and Ta<sub>2</sub>O<sub>5</sub><sup>12)</sup> increased the grain size of the UO<sub>2</sub>. However, in the case of the Nb<sub>2</sub>O<sub>5</sub>-doped UO<sub>2</sub> pellet, the fission gas release rate rather reportedly increased as compared with the undoped UO<sub>2</sub> because of the Nb residue, thereby the effect of the large grain size was compensated.<sup>13)</sup> In this study, Li<sub>2</sub>O, known to have a low melting point and a high evaporation rate, has been selected and tested to see whether how many Li residues remained in the pellet after sintering and there is any effect on the sintered density and grain growth of the UO<sub>2</sub>.

### 2. Experimental Procedure

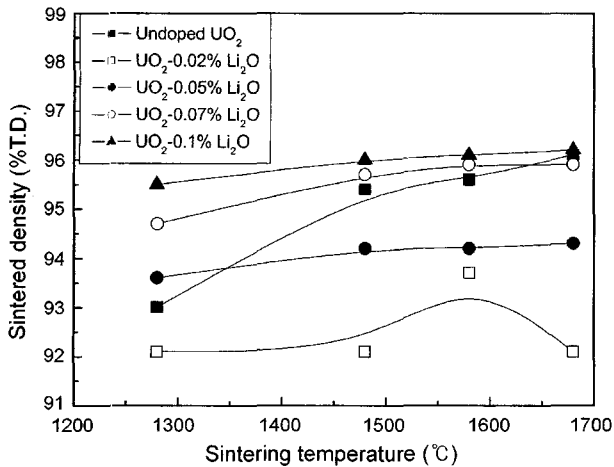
UO<sub>2</sub> powder used in this study was ex-AUC UO<sub>2</sub> manufac-

tured at KAERI (Korea Atomic Energy Research Institute). Its average particle size, specific surface area and O/U ratio was 25 μm, 4.8 m<sup>2</sup>/g and 2.2, respectively. Li<sub>2</sub>O powder with a purity of 97% was purchased from the Aldrich company. UO<sub>2</sub> powder was mixed with weighted amounts of Li<sub>2</sub>O powder, at concentrations varying between 0.02 and 0.1 wt% Li<sub>2</sub>O. Mixing of these powders was carried out by Turbula mixer for 1 h. The prepared powder mixture samples were compacted with a compaction pressure of 3 ton/cm<sup>2</sup> and the green pellet specimens were sintered at 1280, 1480, 1580, and 1680°C in H<sub>2</sub> for 4 h and at 1100°C and 1680°C in CO<sub>2</sub> for 4 h. The flow rate of H<sub>2</sub> and CO<sub>2</sub> gas was 250 SCCM. The pellets sintered in CO<sub>2</sub> were reduced at 1100°C for 1 h in a H<sub>2</sub> atmosphere to adjust the O/U ratio to 2.0. The densification behavior of the UO<sub>2</sub> and UO<sub>2</sub>-0.1 wt%Li<sub>2</sub>O was observed by a dilatometer (NETZSCH, DIL402C) at the temperature of 1650°C with a heating rate of 5°C/min in a H<sub>2</sub> atmosphere. The evaporation behavior of Li<sub>2</sub>O powder was observed by a thermogravimeter (ATI Cahn, TG171) at a temperature of 1500°C with a heating rate of 5°C in the H<sub>2</sub> and CO<sub>2</sub> atmospheres. For the evaporation experiments, a Mo crucible and a Pt crucible were used in the H<sub>2</sub> and CO<sub>2</sub> atmospheres, respectively. Phase analysis was conducted with scattering angles between 20° and 70° by an X-ray diffractometer (Mac Science; MXP3A-HF, CuKα at 30 mA and 40 kV). The sintered density and average grain size were measured by the water immersion method and the linear intercept method, respectively. The Li elements in UO<sub>2</sub> pellets were measured by a atomic absorption spectrometer (Perkin Elmer 311).

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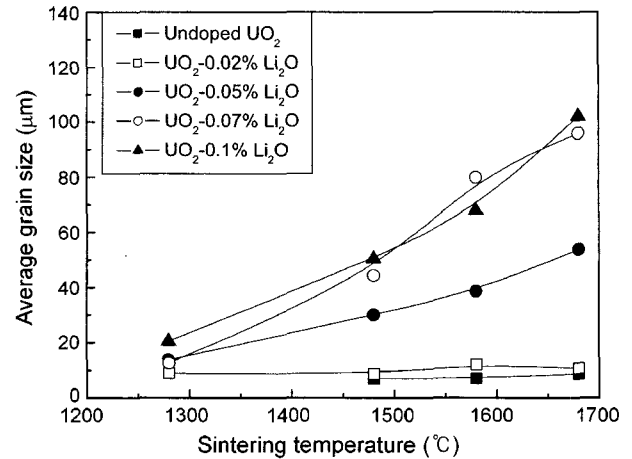


**Fig. 1.** The variation in the sintered density of the undoped  $\text{UO}_2$  and  $\text{Li}_2\text{O}$ -doped  $\text{UO}_2$  pellets sintered at 1280, 1480, 1580, and 1680°C for 4 h in  $\text{H}_2$ .

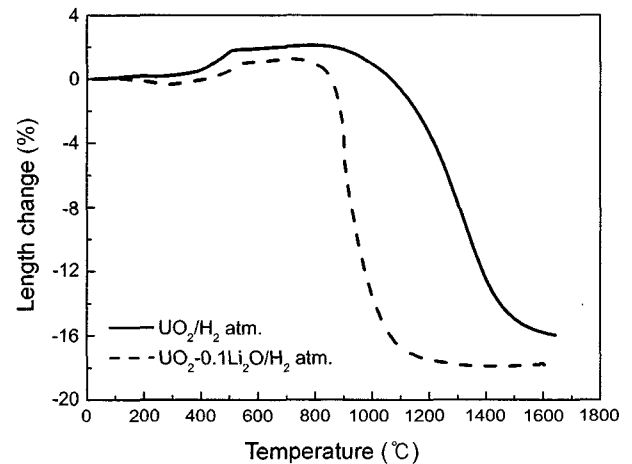
### 3. Results and Discussion

The variation in the sintered density of the undoped  $\text{UO}_2$  and  $\text{Li}_2\text{O}$ -doped  $\text{UO}_2$  pellets sintered at 1280, 1480, 1580, and 1680°C for 4 h in  $\text{H}_2$  is shown in Fig. 1. The variation in the sintered density with the sintering temperatures was different between the undoped  $\text{UO}_2$  and  $\text{Li}_2\text{O}$ -doped  $\text{UO}_2$ . At 1280°C, the sintered density of the undoped  $\text{UO}_2$  pellet was 93%T.D. and that of the 0.05 wt% $\text{Li}_2\text{O}$ -doped  $\text{UO}_2$  and 0.1 wt% $\text{Li}_2\text{O}$ -doped  $\text{UO}_2$  pellets was increased to 93.6%T.D. and 95.5%T.D., respectively. However, the  $\text{UO}_2$ -0.02 wt%  $\text{Li}_2\text{O}$  pellets have a lower sintered density than the undoped  $\text{UO}_2$  pellets at all the sintering temperatures studied in this experiment. Sintered density of the  $\text{UO}_2$  pellets doped with an amount of more than 0.05 wt% $\text{Li}_2\text{O}$  was not increased any more above 1480°C, that is, the 0.05 wt% $\text{Li}_2\text{O}$ -doped  $\text{UO}_2$  pellet has the same sintered density (94.2%T.D.) at both 1480°C and 1680°C and in the case of the 0.1 wt% $\text{Li}_2\text{O}$ -doped  $\text{UO}_2$  pellet, the sintered density is 96%T.D. and 96.2%T.D., respectively, at these temperatures. On the other hand, the sintered density of the undoped  $\text{UO}_2$  pellet increased from 95.4%T.D. to 96.1%T.D. as the sintering temperature increased from 1480°C to 1680°C.

Fig. 2 shows the variation of the grain size of the undoped  $\text{UO}_2$  and  $\text{Li}_2\text{O}$ -doped  $\text{UO}_2$  pellets sintered at 1280, 1480, 1580, and 1680°C for 4 h in  $\text{H}_2$ . The grain growth proceeded as the  $\text{Li}_2\text{O}$  amount increased to 0.05 – 0.1 wt%, that is, the average grain size of the undoped  $\text{UO}_2$  pellet sintered at 1480°C and 1680°C is about 5  $\mu\text{m}$  and 8  $\mu\text{m}$ , respectively, and that of the 0.1 wt% $\text{Li}_2\text{O}$ -doped  $\text{UO}_2$  increased up to about 50  $\mu\text{m}$  and 100  $\mu\text{m}$  at these temperatures, respectively. However, the small amount of  $\text{Li}_2\text{O}$ , for example in  $\text{UO}_2$ -0.02 wt% $\text{Li}_2\text{O}$ , did not increase the grain size of  $\text{UO}_2$ , which was probably due to the density decrease in this pellet. It is confirmed by these results that the  $\text{Li}_2\text{O}$  addition more than 0.02 wt% is capable of enhancing to a great extent the uranium diffusion.



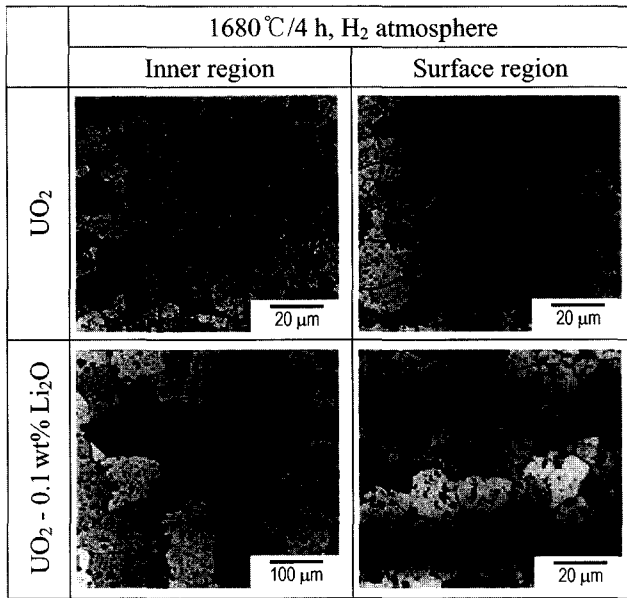
**Fig. 2.** The variation in the average grain size of the undoped  $\text{UO}_2$  and  $\text{Li}_2\text{O}$ -doped  $\text{UO}_2$  pellets sintered at 1280, 1480, 1580, and 1680°C for 4 h in  $\text{H}_2$ .



**Fig. 3.** The dilatometric result of the  $\text{UO}_2$  and  $\text{UO}_2$ -0.1 wt%  $\text{Li}_2\text{O}$  pellets sintered at 1650°C in  $\text{H}_2$ .

Fig. 3 shows the dilatometric results of the undoped  $\text{UO}_2$  and  $\text{UO}_2$ -0.1 wt% $\text{Li}_2\text{O}$  pellets heated up to 1650°C in a  $\text{H}_2$  atmosphere. Undoped  $\text{UO}_2$  starts its densification at about 900°C and has a maximum shrinkage rate at 1300°C. The densification of  $\text{UO}_2$ -0.1 wt% $\text{Li}_2\text{O}$  started at the temperature of about 800°C and was completed at about 1300°C and the maximum shrinkage rate was observed at about 900°C. Therefore, in the  $\text{UO}_2$ -0.1 wt% $\text{Li}_2\text{O}$  pellet, the reason for the sintered density being so little between 1480°C and 1680°C is because its densification was almost completed at about 1300°C.

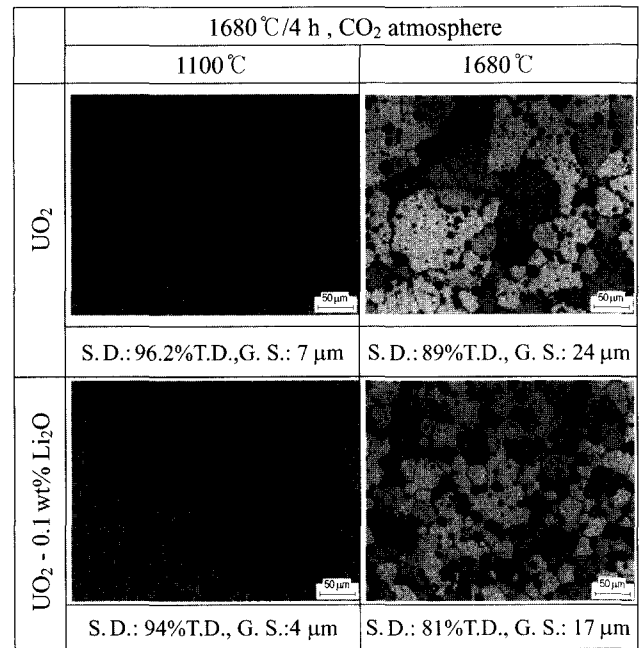
Fig. 4 shows the microstructures of the undoped  $\text{UO}_2$  and  $\text{UO}_2$ -0.1 wt%  $\text{Li}_2\text{O}$  pellets sintered at 1680°C in a  $\text{H}_2$  atmosphere. Undoped  $\text{UO}_2$  pellet has an average grain size of 8  $\mu\text{m}$  and the pores are homogeneously distributed on the grain boundaries and in the grains and the difference in the grain size between the inner and surface regions of the pellet was negligible. However, as shown in this figure, there is a large difference in the grain size between the inner and



**Fig. 4.** Microstructures of the surface and inner region of the UO<sub>2</sub> and 0.1 wt%Li<sub>2</sub>O-doped UO<sub>2</sub> pellets sintered at 1680°C in the H<sub>2</sub> atmosphere.

surface regions in the 0.1 wt%Li<sub>2</sub>O-doped UO<sub>2</sub> pellet. This difference of the grains size between the inner and surface regions is commonly observed in the Li<sub>2</sub>O-doped UO<sub>2</sub> pellet in this study and this difference is larger as the Li<sub>2</sub>O amount increases. For example, the microstructure of the 0.1 wt%Li<sub>2</sub>O-doped UO<sub>2</sub> can be divided into approximately three regions; that is, the surface, transition and coarse grain regions along the direction from the surface towards the center of the pellet. Surface region with the width of 30~50 μm from the surface has an undoped UO<sub>2</sub>-like grain structure with an average grain size of 8 μm but, in the transition region, grains between 20 μm and 70 μm are distributed with a width of a few hundred from the surface region and finally the coarse grain region is observed in the inner region, in which the average grain size is about 100 μm. Grain size distribution of each zone in the Li<sub>2</sub>O-doped UO<sub>2</sub> pellets is different with the sintering temperatures and Li<sub>2</sub>O amount. The grain size data shown in Fig. 3 are those measured in the coarse grain region of each Li<sub>2</sub>O-doped UO<sub>2</sub> pellet. The width of coarse grain region in UO<sub>2</sub>-0.1 wt%Li<sub>2</sub>O pellet become narrow as the sintering temperatures and Li<sub>2</sub>O amount decrease. The width of coarse grain region in the UO<sub>2</sub>-0.1 wt%Li<sub>2</sub>O pellet was narrowest, about the width of a few hundred μm, at 1280°C and relatively large after 1480°C. From these results, it is confirmed that Li<sub>2</sub>O remarkably enhanced the grain growth of UO<sub>2</sub> around 1480°C by liquid phase sintering.

Fig. 5 shows the microstructures of the UO<sub>2</sub> and UO<sub>2</sub>-0.1 wt%Li<sub>2</sub>O pellets sintered at 1100°C and 1680°C for 4 h in a CO<sub>2</sub> atmosphere. Sintered density of both the UO<sub>2</sub> and UO<sub>2</sub>-0.1 wt%Li<sub>2</sub>O pellets decreased as the sintering temperature increased from 1100°C to 1680°C, that is, from 96.2% T.D. to 89% T.D. in the UO<sub>2</sub> and from 94% T.D. to 81% T.D. in

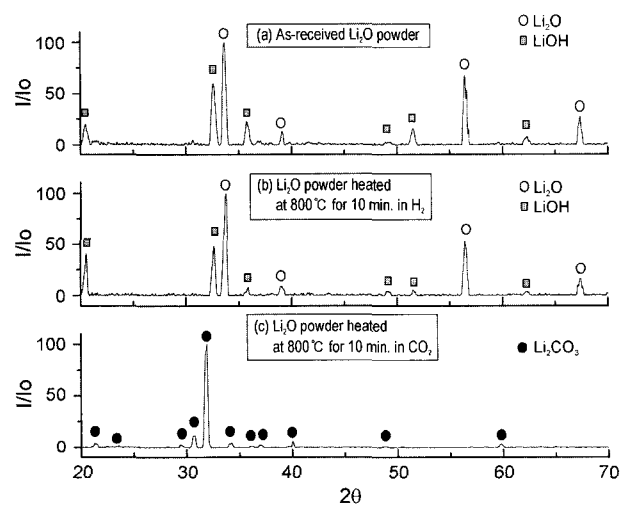


S. D.: sintered density, G. S.: grain size

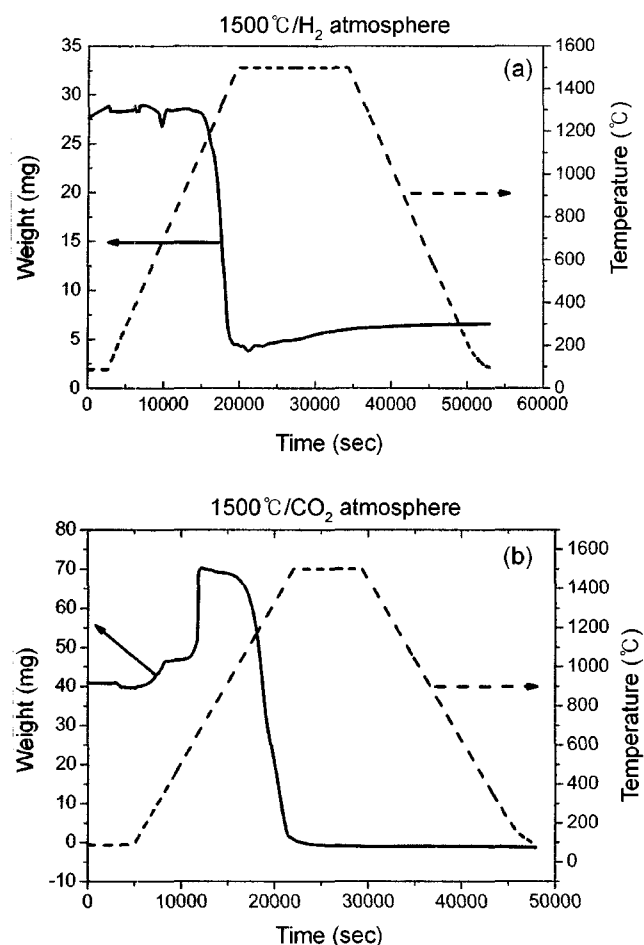
**Fig. 5.** The microstructures of the UO<sub>2</sub> and UO<sub>2</sub>-0.1 wt%Li<sub>2</sub>O pellets sintered at 1100°C and 1680°C in the CO<sub>2</sub> atmosphere.

the UO<sub>2</sub>-0.1 wt%Li<sub>2</sub>O, respectively. The UO<sub>2</sub> pellet sintered at 1100°C has bimodal grain structures typical for a AUC-UO<sub>2</sub> pellet sintered in a CO<sub>2</sub> atmosphere. Its average grain size is about 7 μm and there are fine grain clusters of 2 – 3 μm at many places. When the temperature increased up to 1680°C, the bimodal grain size distribution of the UO<sub>2</sub> pellet disappeared and its grain size increased up to about 24 μm. The grain size of the UO<sub>2</sub>-0.1 wt%Li<sub>2</sub>O pellets at 1100°C and 1680°C was 4 μm and 17 μm, respectively. As contrasted with the results in the H<sub>2</sub> atmosphere, Li<sub>2</sub>O rather impeded the grain growth of the UO<sub>2</sub> in the CO<sub>2</sub> atmosphere.

Fig. 6 shows the XRD patterns of three kinds of Li<sub>2</sub>O pow-



**Fig. 6.** XRD phase analysis of the Li<sub>2</sub>O powders.



**Fig. 7.** Thermogravimetry of  $\text{Li}_2\text{O}$  powder during heating up to  $1500^\circ\text{C}$  in  $\text{H}_2$  and  $\text{CO}_2$  atmospheres. Heating rate is  $5^\circ\text{C}/\text{min}$ : (a)  $\text{H}_2$  atmosphere and (b)  $\text{CO}_2$  atmosphere.

ders. As-received  $\text{Li}_2\text{O}$  powder purchased from the Aldrich company actually consisted of the  $\text{Li}_2\text{O}$  and  $\text{LiOH}$  phases by an XRD analysis at room temperature, as shown in Fig. 6(a). The  $\text{Li}_2\text{O}$  powder, heated in the  $\text{H}_2$  atmosphere up to  $800^\circ\text{C}$  for 10 min, was also composed of  $\text{Li}_2\text{O}$  and  $\text{LiOH}$  indicating that no response had occurred in this atmosphere. All the phases were, however, converted to the  $\text{Li}_2\text{CO}_3$  single phase as shown in Fig. 6(c) when the as-received  $\text{Li}_2\text{O}$  powder was heated in  $\text{CO}_2$  up to the same temperature and time.

Fig. 7 shows the thermogravimetry of  $\text{Li}_2\text{O}$  powder which was heated up to  $1500^\circ\text{C}$  in  $\text{H}_2$  and  $\text{CO}_2$  atmospheres. In the  $\text{H}_2$  atmosphere, the as-received  $\text{Li}_2\text{O}$  powder started its evaporation at about  $1050^\circ\text{C}$  and continued to evaporate up to  $1500^\circ\text{C}$  and finally, about 80 wt% of the as-received  $\text{Li}_2\text{O}$  powder was evaporated. Kudo *et al.*<sup>14)</sup> also mentioned that the evaporation rate of the lithium oxide such as  $\text{LiO}$ ,  $\text{Li}_2\text{O}$ , and  $\text{Li}_3\text{O}$  was relatively high between  $1000^\circ\text{C}$  and  $1200^\circ\text{C}$ . On the other hand, the  $\text{Li}_2\text{O}$  powder showed a different evaporation rates in the  $\text{CO}_2$  atmosphere. The weight of the  $\text{Li}_2\text{O}$  powder started to increase as the temperature approached  $700^\circ\text{C}$ . This is probably related to the conversion from the  $\text{LiOH}$  and  $\text{Li}_2\text{O}$  phases to the  $\text{Li}_2\text{CO}_3$  phase

during heating, as shown in Fig. 6. The powder started to evaporate slowly between  $700^\circ\text{C}$  and  $1000^\circ\text{C}$  and then rapidly above  $1000^\circ\text{C}$ . In the end, all of the  $\text{Li}_2\text{O}$  powder was evaporated at around  $1450^\circ\text{C}$  in the  $\text{CO}_2$  atmosphere.

Undoped  $\text{UO}_2$  pellet with the grain size of  $8\ \mu\text{m}$  can be conventionally fabricated by sintering at  $1700^\circ\text{C}$  in  $\text{H}_2$  atmosphere. Like other powder materials,  $\text{UO}_2$  powder is also bonded by solid state diffusion or liquid phase sintering.  $\text{TiO}_2$ ,<sup>5,8-11)</sup>  $\text{Nb}_2\text{O}_5$ ,<sup>4-7)</sup> and  $\text{Ta}_2\text{O}_5$ <sup>12)</sup> have been used as sintering additives to enhance the sinterability of  $\text{UO}_2$  and these additives resulted in large-grained  $\text{UO}_2$  by solid state sintering. For instance,  $\text{Ta}_2\text{O}_5$  has been reported to greatly increase its grain size  $1700^\circ\text{C}$  in  $\text{H}_2$  atmosphere. Kim *et al.*<sup>12)</sup> have explained this enhanced grain growth by assuming that  $\text{Ta}^{+5}$  ions substituting for a  $\text{U}^{+4}$  ions in the lattice imparts an effective positive charge to the lattice and thereby uranium vacancies will be formed in order to maintain electrical neutrality and the increase in the concentration of uranium vacancies leads to the increase in the uranium diffusion coefficient.

$\text{Li}_2\text{O}$  powder become melted below  $900^\circ\text{C}$  and therefore  $\text{UO}_2$ - $\text{Li}_2\text{O}$  shows different sintering behavior from the  $\text{UO}_2$ - $\text{Ta}_2\text{O}_5$  phase. The melting point of the  $\text{LiOH}$  and  $\text{Li}_2\text{O}$  phase is known to be  $471^\circ\text{C}$  and  $1570^\circ\text{C}$ , respectively<sup>15)</sup> and it is considered that the  $\text{LiOH}$  and  $\text{Li}_2\text{O}$  phases enhanced the sintering behavior of  $\text{UO}_2$ , mainly by liquid phase sintering. So, at the sintering temperature of  $1280^\circ\text{C}$ , the sintered density of  $\text{Li}_2\text{O}$ -doped  $\text{UO}_2$  pellets was higher than that of undoped  $\text{UO}_2$ , except the  $\text{UO}_2$ -0.02 wt% $\text{Li}_2\text{O}$  pellet. However, the change of sintered density with temperatures in undoped  $\text{UO}_2$  was larger than that in  $\text{Li}_2\text{O}$ -doped  $\text{UO}_2$ . That is because  $\text{UO}_2$  has maximum shrinkage about  $1300^\circ\text{C}$ . In the end, the density difference between undoped  $\text{UO}_2$  and  $\text{UO}_2$ -0.1 wt% $\text{Li}_2\text{O}$  was negligible at  $1680^\circ\text{C}$ .

$\text{Li}_2\text{O}$  amount is related to the volume fraction of liquid and the liquid volume fraction has a significant influence on the sintering rate and the microstructure in  $\text{UO}_2$ . That is to say, the densification and grain growth depend on the amount of liquid phase<sup>16)</sup> because liquid connectivity depend on the volume fraction of liquid and the diffusion of solid phase is through the liquid phase. Usually, liquid phase initially gathers into the center region of the sintered pellet during sintering and then spreads out toward the surface region due to the capillary force. At the low volume fraction of liquid ( $\text{Li}_2\text{O}$ ),  $\text{Li}_2\text{O}$  sites could be a pore sites if the liquid phase gathered in center region can not homogeneously redistribute toward the surface. It is considered that low volume fraction of liquid resulted in low sintered density in  $\text{UO}_2$ -0.02 wt% $\text{Li}_2\text{O}$ .

The interesting thing in  $\text{Li}_2\text{O}$ -doped  $\text{UO}_2$  is its grain size distribution. For example,  $\text{UO}_2$ -0.1 wt% $\text{Li}_2\text{O}$  sintered in  $\text{H}_2$  atmosphere has small grain size in the surface region and large one in the center. This phenomenon is assumed to happen by the evaporation of  $\text{Li}_2\text{O}$  powder and shrinkage behavior of  $\text{UO}_2$ .  $\text{Li}_2\text{O}$  starts to evaporate at around  $1050^\circ\text{C}$  as shown in Fig. 7(a) and at around this temperature the

shrinkage rate of UO<sub>2</sub> is low as shown in Fig. 3. Therefore, when the Li<sub>2</sub>O-doped UO<sub>2</sub> compact is sintered in H<sub>2</sub> atmosphere, UO<sub>2</sub> will have open structure in the surface at around 1050°C and, in this case, the Li<sub>2</sub>O phase distributed in the pellet surface region may be easily evaporated and therefore may not affect the grain growth of the UO<sub>2</sub>. On the other hand, the Li<sub>2</sub>O located in the inner region of the pellet will not be easily evaporated and can enhance the grain growth of UO<sub>2</sub>.

Lay and Carter<sup>17)</sup> have studied the role of O/U ratio on sintering of UO<sub>2</sub> and reported that the uranium diffusion coefficient of UO<sub>2</sub> in oxidizing atmosphere having an O/U ratio of 2.02 is 10<sup>8</sup> times greater than in H<sub>2</sub> atmosphere having an O/U ratio of 2.00. So, undoped UO<sub>2</sub> pellet with about 7 μm, in CO<sub>2</sub> atmosphere, can be fabricated even at 1100°C as shown in Fig. 5.

Dopants such as Ta<sub>2</sub>O<sub>5</sub> did not play a good role in CO<sub>2</sub> atmosphere even if they increased the uranium diffusivity in H<sub>2</sub> atmosphere. Ta<sub>2</sub>O<sub>5</sub> increased the grain size of UO<sub>2</sub> in reducing atmosphere but it did not increase that of UO<sub>2</sub> in oxidizing atmosphere because UO<sub>2</sub> has already oxygen interstitials in the atmosphere.<sup>12)</sup>

Like in the Ta<sub>2</sub>O<sub>5</sub>-doped UO<sub>2</sub>, the grain size of UO<sub>2</sub>-0.1 wt%Li<sub>2</sub>O was also not increased in the CO<sub>2</sub> atmosphere. This is because the Li<sub>2</sub>O and LiOH phases were converted to the Li<sub>2</sub>CO<sub>3</sub> phase during the heating in CO<sub>2</sub> as shown in Fig. 6. Grain growth remarkably occurred around 1480°C by liquid phase sintering in Li<sub>2</sub>O-doped UO<sub>2</sub> as mentioned before. The boiling point of Li<sub>2</sub>CO<sub>3</sub> phase is 1300°C and thus the phase will be a gas phase around 1300°C. In this case, the Li<sub>2</sub>O powder can be a pore former and therefore it is expected to rather hinder the grain growth of the UO<sub>2</sub>. As shown in Fig. 5, a lot of pores are principally located on grain boundaries. Unlike in the H<sub>2</sub> atmosphere, UO<sub>2</sub>-0.1 wt%Li<sub>2</sub>O pellet has, in CO<sub>2</sub> atmosphere, about the same grain size between surface and inner region. This is probably because Li<sub>2</sub>O and LiOH phases converted to Li<sub>2</sub>CO<sub>3</sub> in CO<sub>2</sub>.

The Li residues in the UO<sub>2</sub>-0.1 wt%Li<sub>2</sub>O pellets sintered at 1600°C for 4 h in the H<sub>2</sub> or CO<sub>2</sub> atmosphere were measured. Li amount in UO<sub>2</sub>-0.1 wt%Li<sub>2</sub>O is about 232 ppm. Li residues of the UO<sub>2</sub>-0.1 wt%Li<sub>2</sub>O pellet in H<sub>2</sub> and CO<sub>2</sub> atmospheres were about 40 ppm and 100 ppm, respectively. According to Kutty *et al.*,<sup>18)</sup> the onset of shrinkage occurs at around 700°C and maximum shrinkage rate was observed between 900°C and 1000°C. So, the reason for more Li residues in CO<sub>2</sub> atmosphere is considered that it was probably difficult for Li<sub>2</sub>CO<sub>3</sub> gas phase to be evaporated out of the pellet during the sintering.

#### 4. Conclusions

The variation of the sintered density and the grain size as a function of the Li<sub>2</sub>O amount and sintering atmosphere in UO<sub>2</sub> was observed.

1. In the H<sub>2</sub> atmosphere, the Li<sub>2</sub>O enhanced the grain

growth of the UO<sub>2</sub> pellet and the Li<sub>2</sub>O-doped UO<sub>2</sub> pellets had a grain size distribution similar to that of the undoped UO<sub>2</sub> around the pellet surface and a larger grain size in the inner part.

2. The undoped UO<sub>2</sub> started densification at about 900°C and continued to densify until 1650°C while the densification of the UO<sub>2</sub>-0.1 wt%Li<sub>2</sub>O started at about 800°C and was completed at around 1300°C.

3. In the CO<sub>2</sub> atmosphere, the Li<sub>2</sub>O rather impeded the grain growth of the UO<sub>2</sub> pellet largely because the Li<sub>2</sub>O and LiOH phases were converted to Li<sub>2</sub>CO<sub>3</sub> with a lower boiling point during the heating in this atmosphere.

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