

The Influence of Sintering Atmosphere on the Reduction Behaviour of Refractory Bricks and the Basic Properties of UO_2 Pellet

Seung Jae Lee, Kyu Tae Kim and Bum Jin Chung*

Dept. of PWR Fuel Development, Korea Nuclear Fuel Company, Ltd.
150 Deogjin-dong, Yusong-gu, Taejeon, 305-353, Korea
*Nuclear R&D Division, Ministry of Science and Technology, Gwacheon,
Kyunggi 427-760, Korea
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The UO_2 pellets are usually sintered under hydrogen gas atmosphere. Hydrogen gas may cause unexpected early failure of the refractory bricks in the sintering furnace. In this work, nitrogen was mixed with hydrogen to investigate the effect of nitrogen gas on a failure mechanism of the refractory bricks and on the microstructure of the UO_2 pellet. The hydrogen-nitrogen mixed gas experiments show that the larger nitrogen the mixed gas contains, the less the refractory materials are reduced by hydrogen. The weight loss measurements at 1400°C for fire clay and chamotte refractories containing high content of SiO_2 indicate that the weight loss rate for the mixed gas is about half of that for the hydrogen gas. Based on the thermochemical analyses, it is proposed that the weight loss is caused by hydrogen-induced reduction of free SiO_2 and/or SiO_2 bonded to Al_2O_3 in the fire clay and chamotte refractories. However, the retardation of the hydrogen-induced SiO_2 reduction rate under the mixed gas atmosphere may be due to the reduction of the surface reaction rate between hydrogen gas and refractory materials in proportion to volume fraction of nitrogen gas in the mixed gas. On the other hand, the mixed gas experiments show that the test data for UO_2 pellet still meet the related specification values, even if there exists a slight difference in the pellet microstructural parameters between the cases of the mixed gas and the hydrogen gas.

Key words : Sintering, Refractory Bricks, UO_2 Pellet

I. Introduction

UO_2 powder made from the uranium reversion process is pressed into a cylindrical form and then sintered in a sintering furnace. A schematic diagram of the sintering furnace and the thermal distribution of hot

zone are shown in Fig. 1. This figure shows that the operating temperature is in the range of 900 to 1750°C . Fig. 2¹⁾ shows $\text{H}_2\text{O}/\text{H}_2$ ratios in equilibrium with UO_{2-x} , UO_2 and UO_{2+x} .^{2,3)} To produce stoichiometric UO_2 pellets in the temperature range of 900 to 1750°C , the $\text{H}_2\text{O}/\text{H}_2$ ratio should be in the range of 10^{-2} – 10^{-4} . With the use of

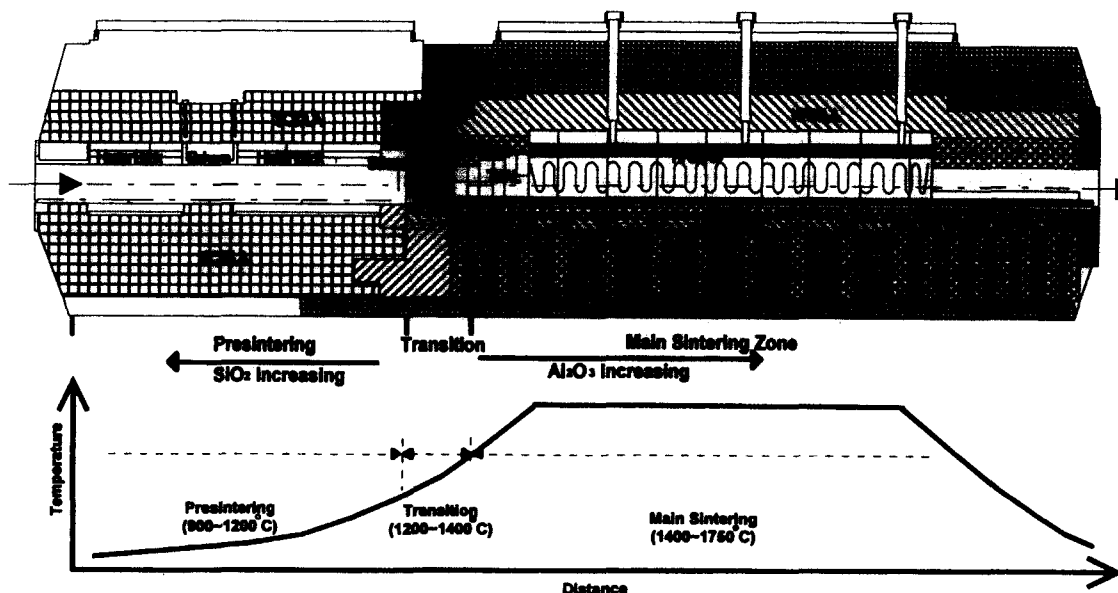


Fig. 1. A schematic diagram of the UO_2 sintering furnace and sintering temperature profile.

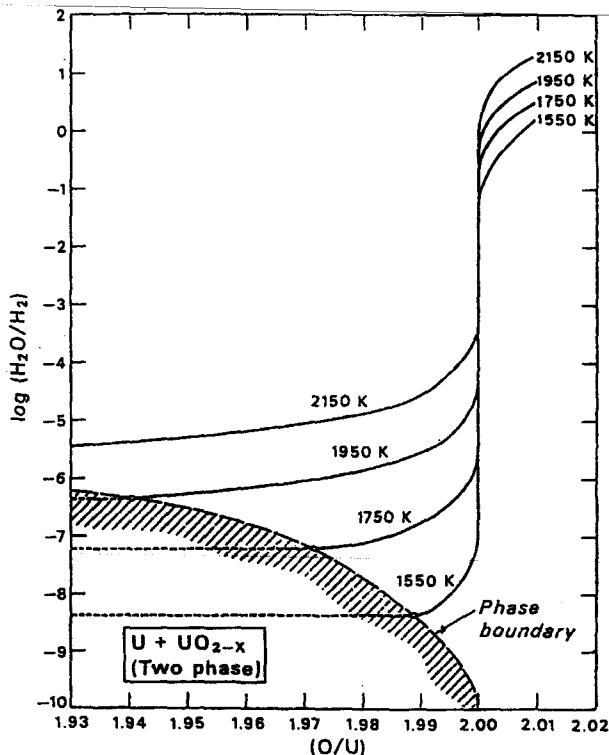


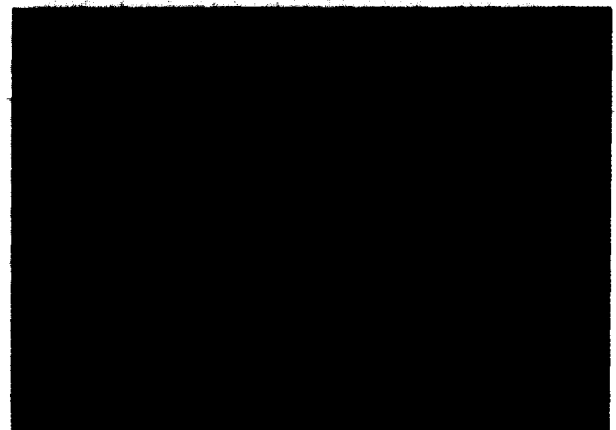
Fig. 2. H_2O/H_2 ratios in equilibrium with UO_{2-x} , UO_2 and UO_{2+x} .¹⁾

this H_2O/H_2 ratio, the oxygen partial pressure in the main sintering zone is calculated to be in the range of 10^{-11} – 10^{-16} atm. It is recently found that the hydrogen gas may cause unexpected early failure of the refractory bricks in the presintering and transition sintering zones which are operated in the temperature range of 900–1400°C (Fig. 3). These failures may be caused by hydrogen interaction with refractory materials, resulting in gas phases such as SiO and H_2O .^{4,8)} As seen in Fig. 3, it is natural that the fire clay and chamotte refractories in the presintering and transition sintering zones should fail due to the SiO_2 reduction since those refractories contain high contents of SiO_2 , while the pure alumina bricks in the main sintering zone should not fail. On the other hand, it is reported that Ni-Cr heating elements in the presintering and transition sintering zones and Mo heating elements in the main sintering zone break frequently.⁹⁾ The failure of Ni-Cr heating elements may be caused by the reaction of Ni-Cr and SiO gas, whereas the failure of Mo heating elements by the reaction of Mo and H_2O gas.⁹⁾ The SiO gas and H_2O gas may be produced during the SiO_2 reduction by hydrogen gas, as mentioned above.

It is well known that the use of hydrogen gas mixed with inert gas may retard the hydrogen-induced SiO_2 reduction. In this study, the effect of nitrogen gas on the reduction behaviour of SiO_2 containing refractory bricks was investigated at the same condition of UO_2 sintering. Additionally, the effect of nitrogen gas on the micro-



(a) Presintering zone



(b) Transition sintering zone

Fig. 3. Photographs of broken refractory bricks.

structure of the UO_2 pellets was also analyzed.

II. Experimental Procedure

1. Hydrogen-induced Reduction Tests for Refractory Bricks

The fire clay and chamotte refractory bricks (Table 1) were selected as test specimens to investigate the effect of the H_2-N_2 mixed gas on the reduction rate of the refractory bricks in the presintering and transition sintering zones. The pure alumina bricks in the main sintering zone were also selected to check the possibility of the reaction with hydrogen, even if thermochemical analyses indicate that the pure alumina will not interact with hydrogen gas and/or nitrogen gas at the operating temperature considered. Note that the locations of the test specimens in the sintering furnace are shown in Fig. 1. As shown in Table 1, four specimens of SC37LU, SC 37LA, SCHAM and SC40AA contain more than 50% of SiO_2 , while the other specimens have very low SiO_2 content.⁹⁾ Table 2 summarizes test conditions and items to be analyzed. As given in this table, two kinds of sintering atmospheres such as hydrogen gas (100% H_2) and 50% H_2 -50% N_2 mixed gas were used at the temper-

Table 1. Test Specimens for Hydrogen-induced Reduction Tests

Specimens	Location in sintering furnace*	Materials	SiO ₂ (%)
KR99AC	S	High alumina	≤0.2
KR98AA	S	High alumina	1.0
TKAL	T	High alumina	1.0
KR81LA	T+S	High alumina	1.5
KR84LB	P+T	Alumina	11.83
SC37LU	P	Alumina-silica (Andalusite/Sillimanite)	50-60
SC37LA	P	Alumina-silica (Andalusite/Sillimanite)	54.63
SCHAM	P	Silica-alumina (Chamotte)	55-70
SC40AA	P (Heater brick)	Silica-alumina (Fire clay)	57

(*): P (presintering zone), T (transition sintering zone), S (main sintering zone)

atures of 1200 and 1400°C for 5, 10 or 20 h, respectively. The test temperatures were taken into account since the presintering and transition sintering zones are operated in the temperature range of 900–1400°C. Weights of the test specimens were measured before and after tests to calculate the reduction rate of the test specimens.

2. Microstructural Tests for UO₂ Pellets

UO₂ powders are blended usually with U₃O₈ powders as a pore former to control sintered density of UO₂. The physical properties of UO₂ and U₃O₈ powders are given in Table 3. The blended powders were compacted to green pellets with density of 5.6±0.05 g/cm³. The green pellets were sintered under the test conditions given in

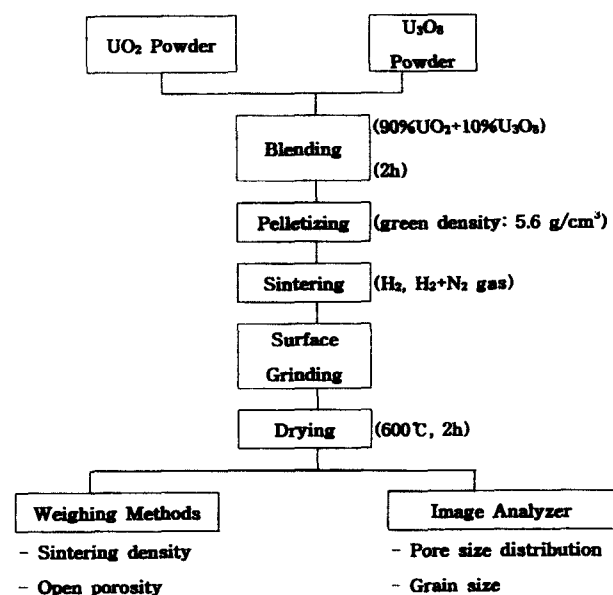
**Fig. 4.** A flow chart of the experimental procedure for microstructural analysis of the UO₂ pellet.

Table 4. The target density of UO₂ pellets was in the range of 10.41–10.57 g/cm³. As shown in this table, three kinds of sintering atmospheres such as hydrogen gas (100%H₂), 70%H₂-30%N₂ and 50%H₂-50%N₂ mixed gases were used at the temperatures of 1600, 1700 and 1750°C for 5 h, respectively. Microstructural parameters of UO₂ pellets such as density, grain size, pore size distribution and open porosity were measured to investigate the effect of the mixed gas on the microstructure of UO₂ pellets. The experimental procedures of the specimens are schematically drawn in Fig. 4.

Table 2. Test Conditions for Hydrogen-induced Reduction of Refractory Bricks

	Sintering conditions			Analysis items	Specimen size (mm)
	Temperature (°C)	Holding time (hr)	Atmosphere		
Nine refractory test specimens	1200 1400	5 10 20	100%H ₂ 50%H ₂ -50%N ₂	• Weight • Density • Visual exam.	30×20×20

Table 3. Characterization of UO₂ and U₃O₈ Powder

	U-content (%)	O/U ratio	Moisture content (ppm)	BET surface (m ² /g)	Apparent density (g/cm ³)	Flowability (sec/50 g)	Particle size* (%)	F-content (ppm)
UO ₂	87.460	2.106	1370	5.57	2.20	1.36	99.85	41
U ₃ O ₈	84.790	2.654	620	0.53	-	-	-	-

(*): The amount of UO₂ powder passed through 200 μm-Sieve.

Table 4. Test Conditions for the Microstructural Analysis of UO₂ Pellet

	Sintering conditions			Analysis items	Specimen size (mm)
	Temperature (°C)	Holding time (h)	Atmosphere		
UO ₂ pellet	1600 1700 1750	5	100%H ₂ 70%H ₂ -30%N ₂ 50%H ₂ -50%N ₂	• Density • Grain size • Open porosity • Pore size distribution	O. D.=8.0 Length=10.0

Table 5. Weight Losses of Test Specimens Under Various Sintering Conditions

Test specimen		Weight loss (%)								
		TKAL	SCHAM	SC37LA	SC37LU	KR81LA	KR84LB	SC40AA (HB)	KR98AA	KR99AC
100% H ₂	1200°C-5 h	0.0791	0.2969	0.8688	0.5363	0.0903	0.0406	0.2662	-	-
	1200°C-10 h	0.1235	0.3161	0.9912	0.5690	0.1154	0.0472	0.2809	-	-
	1200°C-20 h	0.1476	0.3651	1.0675	0.6143	0.1233	0.0836	0.2926	-	-
	1400°C-10 h	0.2837	0.4608	1.5755	0.8178	0.3067	0.4915	-	0.4009	0.1317
50%H ₂ - 50%N ₂ Mix	1200°C-5 h	0.0632	0.2902	0.8288	0.4934	0.0259	0.0397	0.2554	-	-
	1200°C-10 h	0.0980	0.3121	0.9627	0.5362	0.0664	0.0459	0.2579	-	-
	1200°C-20 h	0.1384	0.3327	1.0571	0.5853	0.1005	0.0702	0.2613	-	-
	1400°C-10 h	0.2539	0.2429	1.2136	0.2424	0.1231	0.0732	-	0.2969	0.0811

(-): not tested.

III. Results and Discussion

1. Reduction Test Results of Refractory bricks

The weight loss data for nine test specimens are given in Table 5. The weight loss data obtained under the test condition of 1400°C and 10h were plotted in Fig. 5 and those under the test conditions of 1200°C and 10 h also in Fig. 6. From these figures, it is found that the higher content of SiO₂ the test specimens contain, the larger weight loss they undergo. Note that four specimens of SC37LU, SC37LA, SCHAM and SC40AA contain SiO₂ more than 50%, while the other specimens very low SiO₂ content. In addition, the weight loss goes up with increasing test temperature and/or holding time, as expected. The amounts of weight loss at 1200°C for the 50%H₂-50%N₂ mixed gas are a little less than that for the hydrogen gas, while the amounts of weight loss at 1400°C for the 50%H₂-50%N₂ mixed gas are much less than that for the hydrogen gas. It should be noted that at the test temperature of 1400°C, the weight loss of

SC37LU, KR81LA and KR84LB for the 50%H₂-50%N₂ mixed gas is less than half of their weight loss for the hydrogen gas.

2. Thermochemical Prediction for Corrosion Behaviors of Refractory bricks

The interactions of Al₂O₃(s) with hydrogen gas which generate gas phases such as AlH, AlOH, AlOOH are not taken into account since these reactions will not occur in the test temperature range considered in this work. The interactions of SiO₂(s) with hydrogen gas which generate gas phases such as SiH₄ and SiH may not occur because the Gibbs free energies of formation for SiH₄(g) and SiH(g) are much greater than those of SiO(g) and Si(s). On the other hand, the interactions of the refractory bricks with nitrogen gas may be also neglected since nitrogen gas is generally known to behave like inert gas under the hydrogen-nitrogen mixed gas atmosphere.

In this work, three kinds of reactions are considered to

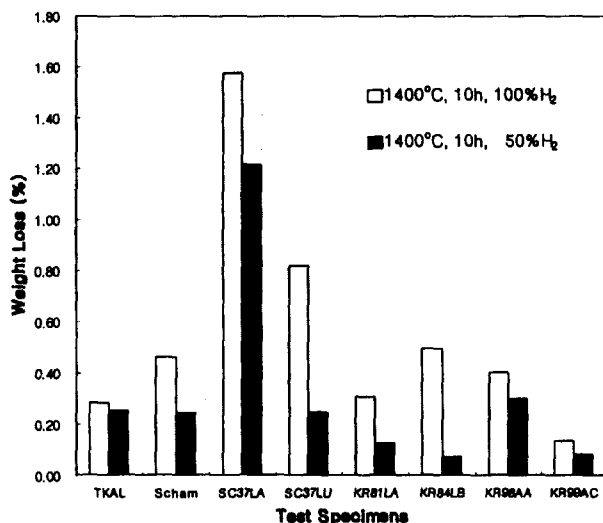


Fig. 5. Comparison of weight losses of the refractory bricks between 100% H₂ gas and 50% H₂-50% N₂ mixed gas (1400°C, 10 h).

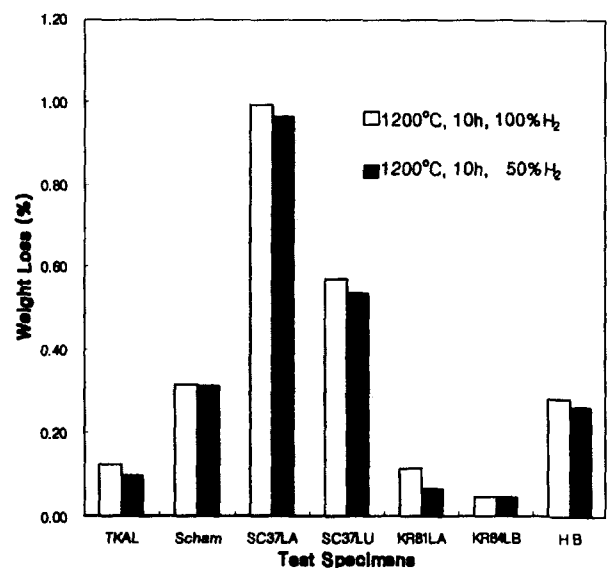


Fig. 6. Comparison of weight losses of the refractory bricks between 100% H₂ gas and 50% H₂-50% N₂ mixed gas (1200°C, 10 h). (HB: Heater Brick; SC40AA)

propose a possible mechanism(s) for the hydrogen-induced reduction of the refractory bricks in the presintering and transition sintering zones.

- Case 1 : Possible Reactions of $Al_2O_3 \cdot SiO_2(s)$ with $H_2(g)$

$$Al_2O_3 \cdot SiO_2(s) + H_2(g) = Al_2O_3(s) + SiO(g) + H_2O(g) \quad (1-1)$$

$$Al_2O_3 \cdot SiO_2(s) + 2H_2(g) = Al_2O_3(s) + Si(s) + 2H_2O(g) \quad (1-2)$$

$$2(Al_2O_3 \cdot SiO_2(s)) + 3H_2(g) = 2Al_2O_3(s) + SiO(g) + Si(s) + 3H_2O(g) \quad (1-3)$$
- Case 2 : Possible Reactions of $3Al_2O_3 \cdot 2SiO_2(s)$ with $H_2(g)$

$$3Al_2O_3 \cdot 2SiO_2(s) + 2H_2(g) = 3Al_2O_3(s) + 2SiO(g) + 2H_2O(g) \quad (2-1)$$

$$3Al_2O_3 \cdot 2SiO_2(s) + 4H_2(g) = 3Al_2O_3(s) + 2Si(s) + 4H_2O(g) \quad (2-2)$$

$$3Al_2O_3 \cdot 2SiO_2(s) + 3H_2(g) = 3Al_2O_3(s) + SiO(g) + Si(s) + 3H_2O(g) \quad (2-3)$$
- Case 3 : Possible Reactions of $SiO_2(s)$ with $H_2(g)$

$$SiO_2(s) + H_2(g) = SiO(g) + H_2O(g) \quad (3-1)$$

$$SiO_2(s) + 2H_2(g) = Si(s) + 2H_2O(g) \quad (3-2)$$

The thermochemical analyses were performed with the use of the Gibbs free energy of formation for each component given in the JANAF table. The Gibbs free energies of formation for the reactions described above were plotted in Fig. 7. From this figure, it can be said that the reactions considered in cases 1 and 2 are not possible due to high positive Gibbs free energies, while the reactions considered in case 3 are possible due to negative Gibbs free energies. However, the reaction (3-1) is more likely to occur than the reaction (3-2). It should be noted that cases 1 and 2 deal with the interactions of hydrogen with stabilized SiO_2 in andalusite ($Al_2O_3 \cdot SiO_2$) and mullite ($3Al_2O_3 \cdot 2SiO_2$), whereas case 3 deals with the interactions of hydrogen with free SiO_2 and/or SiO_2 bonded to Al_2O_3 in silica bricks such as fire clay and chamotte. According to the reaction (3-1), the reactions of hydrogen gas with free SiO_2 and/or SiO_2 bonded to Al_2O_3 may cause evaporation of free SiO_2 and/or SiO_2 bonded to Al_2O_3 , resulting in the weight loss of the test specimens. However, the reaction rate of hydrogen with free SiO_2 may be different from that with SiO_2 bonded to Al_2O_3 .

Unfortunately this cannot be clarified since the relative contents of free SiO_2 and SiO_2 bonded to Al_2O_3 for each test specimens are not known.

As explained in section III. 1, the addition of nitrogen gas to hydrogen gas results in lower hydrogen-induced SiO_2 reduction rate. Considering that nitrogen gas behaves like inert gas under the hydrogen-nitrogen mixed gas atmosphere, nitrogen gas is not able to retard the chemical reaction rate between hydrogen and SiO_2 by changing the Gibbs free energy of formation. The nitrogen-induced retarding effect, therefore, may be caused by the reduction of the surface reaction rate between hydrogen gas and refractory bricks in proportion to nitrogen volume fraction in the mixed gas. If the rate controlling step is the chemical reaction at the reaction surface, the hydrogen-induced SiO_2 reduction rate may be decreased in proportion to nitrogen volume fraction in the mixed gas. In other words, the decrease of the weight loss rate is nearly proportional to nitrogen volume fraction in the mixed gas. As given in Table 5, the weight loss data of SCHAM and KR81LA at 1400°C for the 50% H_2 -50% N_2 mixed gas is about half of their weight loss for the hydrogen gas. This may indicate that the rate controlling step for SCHAM and KR81LA at 1400°C is the chemical interaction at the reaction surface. However, other weight loss data given in Table 5 indicate that the differences between the mixed gas and the hydrogen gas are less than 10%. This indicates that other mechanism (s) for the weight loss may prevail, for example, $SiO_2(s)$ diffusion in the refractory brick matrix, diffusion of $SiO(g)$ and/or $H_2O(g)$ in the mixed gas atmosphere, convection of $SiO(g)$ and/or $H_2O(g)$ in the mixed gas atmosphere, etc.

3. Microstructure of UO_2 Pellets

Density data of the UO_2 pellets sintered for 5 h at 1750°C are given in Fig. 8. Density data for other test conditions are not given since they show the same trend.

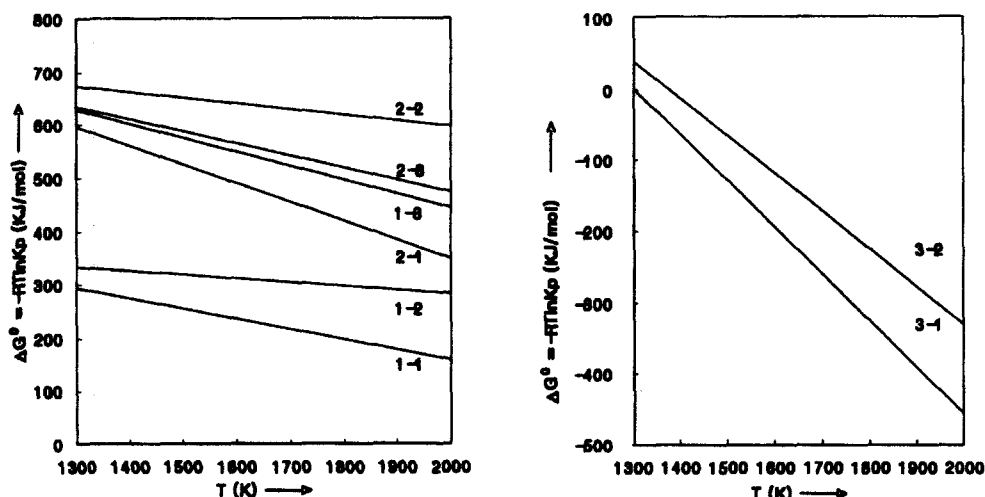


Fig. 7. Gibbs free energy of formation for the chemical reactions considered in this work.

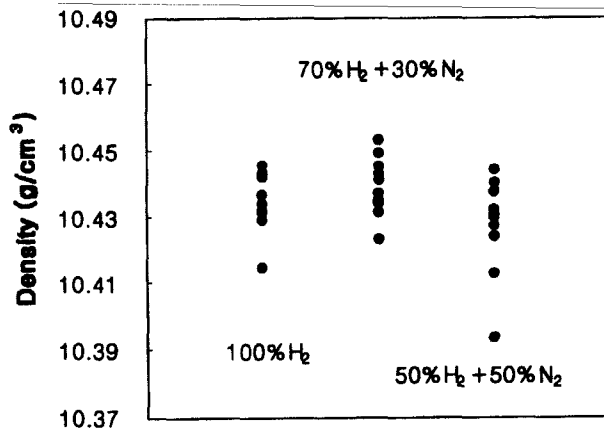


Fig. 8. Density variations of the UO₂ pellets sintered at 1750°C for 5 h in various sintering atmospheres.

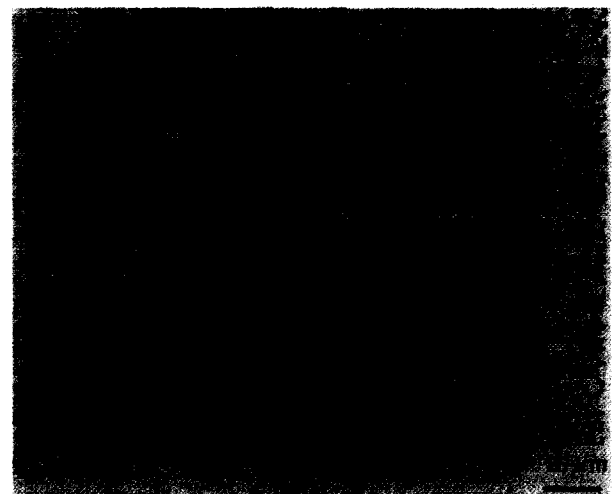
It can be seen that the mixed gas produces larger UO₂ density scattering than the hydrogen gas. However, the mean density is almost the same, regardless of sintered gas composition, and the upper/lower density data still meet the target density of 10.41 to 10.57 g/cm³. Open porosity data are given in Table 6. The mixed gas generates smaller open porosity than the hydrogen gas. Since the smaller open porosity are the better for UO₂ fuel performance, the addition of nitrogen may have a positive effect on open porosity. Pore sizes and distributions are summarized in Table 6. The pore size distributions appear to be monomodal and the differences in the average pore size are negligible, regardless of sintered gas composition. Grain sizes are also given in Table 6. The mixed gas generates less grain growth than the hydrogen gas but the grain sizes still meet the specification lower limit of 5 μm (Fig. 9).

4. Thermochemical Prediction for Microstructural Behaviors of UO₂ Pellets

To produce stoichiometric UO₂ pellets, the H₂O/H₂ ratio should be controlled in a certain range. Fig. 2 shows that in order to have stoichiometric UO₂, the H₂O/H₂ ratio at



(a) H₂



(b) 50 % N₂

Fig. 9. Microstructures of the UO₂ pellets sintered at 1700 °C for 5 h under (a) 100% H₂ and (b) 50% H₂-50% N₂.

1950 K should be in the range of 10^{-0.5}~10⁻⁵, while the H₂O/H₂ ratio at 1550 K in the range of 10⁻¹~10⁻⁷. In other words, hypostoichiometric UO_{2-x} may be produced if

Table 6. Average Open Porosity, Pore Size (distribution) and Grain Size of the UO₂ Pellets Sintered Under Various Sintering Conditions

Sintering gas composition	Sintering condition	Properties of the sintered UO ₂ pellets		
		Average open porosity (%)	Average pore diameter (μm) [#]	Average grain size (μm)
100% H ₂	1600°C-10 h	3.401	1.9	5.3
	1700°C-5 h	2.662	1.9	7.7
	1750°C-5 h	1.841	1.6	7.9
70% H ₂ +30% N ₂	1600°C-10 h	-	-	-
	1700°C-5 h	-	-	-
	1750°C-5 h	1.617	1.6	6.3
50% H ₂ +50% N ₂	1600°C-10 h	2.479	1.8	5.5
	1700°C-5 h	2.494	1.8	6.3
	1750°C-5 h	1.843	1.7	6.5

(#): all test specimens show the monomodal type in pore size distribution, (-): not tested.

the H_2O/H_2 ratio at 1950 K is less than 10^{-5} , whereas hyperstoichiometric UO_{2+x} may be produced if the H_2O/H_2 ratio at 1950 K is more than $10^{-0.5}$. The operating temperature in the sintering furnace is in the range of 1173~2023 K. This indicates that the H_2O/H_2 ratio should be in the range of 10^{-2} ~ 10^{-4} to produce stoichiometric UO_2 pellets in the sintering furnace. The oxygen partial pressures corresponding to these H_2O/H_2 ratios are in the range of 10^{-19} ~ 10^{-23} atm at 1173 K (presintering zone) and 10^{-11} ~ 10^{-15} atm at 2023 K (main sintering zone). However, the addition of nitrogen gas to hydrogen gas may change the oxygen partial pressure if the reaction of $N_2(g)+3H_2(g) \rightarrow 2NH_3(g)$ occurs and/or if nitrogen has a considerable effect on the amount of $H_2O(g)$ generated during the hydrogen-induced SiO_2 reduction. The thermochemical analysis indicates that the reaction of $N_2(g)+3H_2(g) \rightarrow 2NH_3(g)$ is not probable since ΔG_r for this reaction is much larger than ΔG_r for the reaction of $2H_2(g)+O_2(g)=2H_2O(g)$. From the weight loss data for the refractory bricks, however, it is thought that oxygen partial pressure for the mixed gas may decrease since the mixed gas retards the hydrogen-induced SiO_2 reduction rate, resulting in less $H_2O(g)$ evaporation. If the mixed gas contains a little lower oxygen partial pressure than the hydrogen gas, the hypostoichiometric UO_{2-x} pellets can be produced and other microstructural changes occur due to oxygen diffusivity change.

As described in section III.3, the mixed gas generates a slight difference in UO_2 density scattering, open porosity and grain growth rate, compared with the hydrogen gas. This may be caused by a little lower oxygen partial pressure in the mixed gas or by the decrease of the surface reaction rate between hydrogen gas and UO_2 pellets which results from the reaction surface covered by nitrogen in proportion to the nitrogen volume fraction in the mixed gas. However, it should be noted that the UO_2 pellet for the mixed gas still meets the UO_2 pellet-related specification values, even if there exist a slight difference in the UO_2 pellet microstructural parameters between the mixed gas and the hydrogen gas.

IV. Conclusions

Nitrogen gas was mixed with hydrogen gas to investigate the effect of nitrogen gas on the operating life of the refractory bricks in the sintering furnace and on the microstructural parameters of the UO_2 pellets.

It can be generally said that the larger nitrogen the mixed gas contains, the less the refractory bricks are reduced by hydrogen. The weight loss measurements at 1400°C for the fire clay and chamotte refractories containing high content of SiO_2 indicate that the weight loss rate for the mixed gas is about half of that for the

hydrogen gas.

Thermochemical analyses indicate that the weight loss may be caused by hydrogen-induced reduction of free SiO_2 and/or SiO_2 bonded to Al_2O_3 in the fire clay and chamotte refractories. However, the decrease of the hydrogen-induced SiO_2 reduction rate for the mixed gas atmosphere may be due to the decrease of the surface reaction rate between hydrogen gas and SiO_2 in proportion to volume fraction of nitrogen gas in the mixed gas.

The mixed gas experiments show that the UO_2 pellet sintered for the mixed gas still meets the related specification values (density: 10.30~10.58 g/cm³, grain size: $\geq 5 \mu m$, open porosity: information only), even if there exists a slight difference in the UO_2 pellet microstructural parameters between the mixed gas and the hydrogen gas. The mixed gas generates larger UO_2 density scattering, smaller open porosity and less grain growth rate, compared with the hydrogen gas.

Thermochemical analyses indicate that the effect of nitrogen gas on the UO_2 pellet microstructure may be caused by a little lower oxygen partial pressure in the mixed gas. On the other hand, it may be caused by the reduction of the reaction surface between hydrogen gas and UO_2 pellets in proportion to the nitrogen volume fraction in the mixed gas. The additional theoretical and experimental analysis will be performed to check these causes in the future.

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