

Oxidation of Silicon Nitride in Wet Air and Effect of Lutetium Disilicate Coating

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Oxidation behavior of silicon nitride (Si₃N₄) was investigated in flowing air (2.45 cm/s) containing 10%-50% H₂O at a total pressure of 1.8-10 atm at 1300°-1500°C for 100 h. The oxidation of Si₃N₄ progressed with volatilization of the SiO₂ scale; it was more enhanced at a high partial pressure of H₂O rather than at high temperature. The total pressure had little effect on the oxidation. In order to avoid the oxidation, Si₃N₄ substrate was coated with lutetium disilicate (Lu₂Si₂O₇) layer through the intermediate SiO₂-rich phase. While the coating layer well suppressed the oxidation in case of small amount of water vapor, it was not sufficiently effective to suppress the oxidation when the water vapor was rich. SiO₂ volatilization was observed between the layer and substrate. The flexural strength of the coated Si₃N₄ at room temperature was somewhat increased after the oxidation in wet air, while that of the uncoated one was almost unchanged. This increase was attributable to crack healing of the substrate during the oxidation.

I. Introduction

However, one problem of Si_3N_4 is its low oxidation resistance at above 1000°C.^{7,8} The oxidation behavior in dry air obeys a parabolic law, in which oxygen or cation diffusion is rate controlling.⁸ The weight gain is due to the generated SiO₂ scale, which prevents Si₃N₄ from the further oxidation. In contrast, the oxidation behavior in water vapor obeys a paralinear law, in which linear weight loss due to volatilization of the generated SiO₂ scale progresses after the parabolic weight gain enhanced by water vapor oxidation.^{9–11} Therefore, Si₃N₄ continuously recedes because of the simultaneous oxidation and volatilization. Many studies have been conducted to clarify the oxidation mechanisms of Si₃N₄ with volatilization of SiO₂ in water vapor.⁸ However, almost all the previous studies in the literatures were conducted below 1400°C at a small total pressure; the oxidation behavior under more severe conditions (higher temperatures and higher pressures) has not been completely understood.

The oxidation also causes deterioration of mechanical properties of Si_3N_4 .¹² When the oxidation of Si_3N_4 progresses, the cracked SiO_2 scale is generated on the surface because of the large mismatch of thermal expansion coefficients. In addition, in the case of Si_3N_4 with additives, the region beneath the scale is damaged because of the migration of the cationic ions in the grain boundary regions from the interior to the surface. These behaviors lead to the deterioration of the mechanical properties.¹³

The objective of the present research is to investigate the oxidation behavior of Si_3N_4 with Lu_2O_3 -SiO₂ additive in the flowing air containing 10%-50% H₂O at a total pressure of 1.8–10 atm and 1300°-1500°C. The influence of temperature, H₂O partial pressure, and total pressure on the oxidation was examined. Si_3N_4 was also coated with lutetium disilicate ($Lu_2Si_2O_7$) in order to suppress the oxidation. $Lu_2Si_2O_7$ has high melting point and thermal expansion coefficient close to that of Si_3N_4 .^{14,15} Therefore, it can be a candidate for material of environmental barrier coating (EBC). Si_3N_4 coated with $Lu_2Si_2O_7$ was also exposed to wet air and the effect of EBC was examined.

II. Experimental Procedure

(1) Materials

The raw materials used for the present research were α -rich Si₃N₄ (SN-E10, UBE Industries Ltd., Tokyo, Japan; >99.5% purity), Lu₂O₃ (Shin-Etsu Chemical Co. Ltd., Tokyo, Japan; 99.9% purity), and SiO₂ (quartz, Kojundo Chemical Laboratory Co. Ltd., Saitama, Japan; 99.9% purity, 0.8 µm particle size) powders. Si₃N₄ was mixed with 8 wt% of Lu₂O₃ and 2 wt% of SiO₂ in ethanol for 48 h by using ball mill, and cold isostatic pressed (CIPed) at 392 MPa before sintering. It was sintered in 1 MPa-N₂ atmosphere at 2000°C for 4 h in graphite resistance furnace (FVPHP-R-10, FRET-40, Fujidempa Kogyo Co. Ltd., Saitama, Japan). The specimens with dimensions of 3 mm × 4 mm × 37 mm were cut and ground (R_a :<0.2 µm), and their edges were chambered at 45°.

The coated specimens were prepared by the reaction sintering method after dip-coating, which was described in our previous reports.^{15,16} The slurry for dip-coating was composed of Lu_2O_3 and SiO₂ powder with dispersant, binder, deformer, and distilled water. It was ball milled for 48 h. The specimens dipped in the slurry were CIPed at 392 MPa, and reaction sintering was conducted in air at 1550°C for 20 h. The coated specimens were prepared by repeating this treatment three times. (The composition of SiO₂/Lu₂O₃ in the slurry was 1.0 (molar ratio) for the first treatment, and that was 2.0 for second and third ones.)

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Fig.1. Displacement of the specimens and measurement place at the oxidation tests in wet air.

(2) Evaluations

In comparison with the oxidation in wet air, Si_3N_4 specimens were exposed to a static dry air at 1300°, 1400°, and 1500°C for 100 h. Total pressure was 1 atm. The specimens were set on the triangular poles of Si_3N_4 during the oxidation test.

The uncoated and coated Si₃N₄ specimens were exposed in the oxidation test system (MS-1201, Motoyama Co. Ltd., Osaka, Japan) using Al₂O₃ tube (CP, Nikkato Corp., Osaka, Japan; 99% purity with 0.2% Na₂O) and Al₂O₃ sample holder (SSA-S, Nikkato Corp.; 99.6% purity with 0.04% Na₂O). Before the oxidation test, the tube and holder were aged at 1500°C for more than 500 h to decrease the effect of impurity. The oxidation tests were conducted at 1300°, 1400°, and 1500°C in the flowing air (2.45 cm/s; STP) containing 10%, 30%, and 50% H₂O at a total pressure of 1.8, 4.2, and 10 atm. Exposure time was 100 h for each test. The heating and cooling rate was $5^{\circ}C/$ min. The flowing gas and pressure were introduced when the target temperature was attained, and were kept during 100 h testing period. The displacement of the specimens and measurement place were shown in Fig. 1 schematically. Each specimen was set on two Lu₂Si₂O₇ pellets (1 mm \times 3 mm \times 4 mm) during the oxidation test in order to avoid the direct diffusion of impurities such as Na from Al_2O_3 to specimens. The air containing H₂O was flowed to the direction perpendicular to the face (4 mm \times 37 mm) of the specimens, and the dummy Si₃N₄ specimen was set just before the air reached the target specimens. Dummy specimen was used for fixing the oxidation conditions of all the target specimens during each test. The oxidation of Si₃N₄ was measured at the center of specimens from the generated SiO₂ scale thickness and the change of total thickness of specimens to the direction parallel to the flow. The SiO₂ scale thickness was obtained from scanning electron microscopy (SEM; JSM-5600 and JSM-6330F, JEOL Ltd., Tokyo, Japan) observation, and the total thickness change was obtained from measuring the total thickness before and after the oxidation test by micrometer. Three specimens were used for each measurement.

Table I.Results of Oxidation Tests of Silicon Nitride (Si_3N_4) in a Static Dry Air ($P_{total} = 1.0$ atm)

Temperature (°C)	Total pressure (atm)	Time (h)	SiO ₂ scale thickness $(x/side, \mu m)$	Calculated total oxidation $(t, t = 0.5406x/side, \mu m)$
1500	1.0	100	$\begin{array}{c} 3.31 \pm 0.81 \\ 2.06 \pm 0.63 \\ 0.81 \pm 0.81 \end{array}$	1.79
1400	1.0	100		1.11
1300	1.0	100		0.435

The calculated total oxidation (*t*) was obtained from the SiO₂ scale thickness by the Eq. (1). It was assumed that 1 mole of Si₃N₄ (3.36 g/cm³) is changed to 3 moles of SiO₂ (cristobalite; 2.334 g/cm³ of density) without volatilization.

Before and after the oxidation tests, the four-point bending tests (inner and outer spans were 10 and 30 mm, respectively) were conducted at room temperature at a cross-head speed of 0.5 mm/min (Sintech 10/GL, MTS Systems Corp., Eden Prairie, MN). Three specimens were used for each measurement. The microstructures and compositions were examined by SEM, and energy dispersive spectroscopy (EDS; JED-2140, JEOL Ltd.). Crystalline phases were identified by X-ray diffractometry (XRD; RINT 2000, Rigaku Corp., Tokyo, Japan), using Cu $K\alpha$ radiation at 40 kV and 100 mA.

III. Results and Discussion

(1) Oxidation Behavior of Si_3N_4

The density of the uncoated specimen was $3.35-3.37 \text{ g/cm}^3$, being close to the theoretical density. Table I summarizes the results of oxidation tests in dry air. The calculated total oxidation (*t*) (in this paper, total oxidation means the thickness of Si₃N₄ consumed during oxidation) was obtained from the SiO₂ scale thickness (*x*) by:

$$t = \frac{1}{3} \frac{M_{\text{Si}_3\text{N}_4}}{M_{\text{SiO}_2}} \frac{\rho_{\text{SiO}_2}}{\rho_{\text{Si}_3\text{N}_4}} x = 0.5406x$$
(1)

where $M_{\rm Si_3N_4}$ and $\rho_{\rm Si_3N_4}$ are the molecular weight and density of the consumed Si₃N₄, respectively. $M_{\rm SiO_2}$ and $\rho_{\rm SiO_2}$ are the corresponding ones of the generated SiO₂ scale. It was assumed that 1 mole of Si₃N₄ ($M_{\rm Si_3N_4} = 140.28$, $\rho_{\rm Si_3N_4} = 3.36$ g/cm³) was changed to 3 moles of SiO₂ (cristobalite; $M_{\rm SiO_2} = 60.08$, $\rho_{\rm SiO_2} = 2.334$ g/cm³) through the reaction Eq. (2) without volatilization of SiO₂.

$$Si_3N_4(s) + 3O_2(g) = 3SiO_2(s) + 2N_2(g)$$
 (2)

Table II summarizes the results of oxidation tests in wet air. The measured total oxidation (t') was obtained by subtracting the SiO₂ scale thickness (x) from the thickness change (x') of the specimens (t' = |x'-x|). The calculated total oxidation (t) was

Table II. Results of Oxidation Tests of Silicon Nitride (Si₃N₄) in Wet Air

Temperature (°C)	Total pressure (atm)	Partial H ₂ O pressure (atm)	Time (h)	SiO ₂ scale thickness $(x/side, \mu m)$	Thickness change $(x'/side, \mu m)$	Measured total oxidation (t' , $t' = x'-x /side$, µm)	Calculated total oxidation (t , $t = 0.5406x/side$, μ m)
1500	1.8	0.54 (30%)	100	4.91 ± 0.27	+1.17	3.74 ± 0.27	2.65 ± 0.15
1400	1.8	0.54 (30%)	100	3.22 ± 0.54	+0.33	2.88 ± 0.54	1.74 ± 0.29
1300	1.8	0.54 (30%)	100	1.05 ± 1.05	+0.75	0.30 ± 1.05	0.57 ± 0.57
1400	1.8	0.18 (10%)	100	2.14 ± 0.18	+0.58	1.56 ± 0.18	1.16 ± 0.10
1400	1.8	0.90 (50%)	100	2.15 ± 0.54	-1.17	3.31 ± 0.54	1.16 ± 0.29
1400	4.2	0.42 (10%)	100	4.38 ± 0.45	+1.83	2.54 ± 0.45	2.37 ± 0.24
1400	4.2	1.26 (30%)	100	5.18 ± 0.89	+0.50	4.68 ± 0.89	2.80 ± 0.48
1400	4.2	2.10 (50%)	100	9.65 ± 0.90	+1.00	8.65 ± 0.90	5.21 ± 0.48
1400	10	3.00 (30%)	100	10.72 ± 1.08	+4.81	5.90 ± 1.08	5.79 ± 0.58

The measured total oxidation (t') was obtained by subtracting the SiO₂ scale thickness from the thickness change of the specimen. The calculated total oxidation (t) was obtained by the Eq. (1). It was assumed that 1 mole of Si₃N₄ (3.36 g/cm³) is changed to 3 moles of SiO₂ (cristobalite; 2.334 g/cm³ of density) without volatilization.



Fig. 2. Temperature dependence on the measured total oxidation (*t*) of silicon nitride (Si_3N_4) in the flowing air (2.45 cm/s) containing 30% H₂O (0.54 atm) at a total pressure of 1.8 atm for 100 h, in comparison with the oxidation in a static air at a total pressure of 1 atm.

obtained by the Eq. (1), assuming that 1 mole of Si_3N_4 was changed to 3 moles of SiO_2 (cristobalite) through the reaction Eqs. (2) and (3) without volatilization of SiO_2 .

$$Si_3N_4(s) + 6H_2O(g) = 3SiO_2(s) + 2N_2(g) + 6H_2(g)$$
 (3)

In Table II, the measured total oxidation was more than the calculated one in almost all the cases. This indicates that the SiO₂ phase was volatilized probably as Si(OH)₄ by the Eq. (4), which was identified in the reaction of SiO₂ with H₂O by mass spectrometry by Opila *et al.*¹⁷

$$\operatorname{SiO}_2(s) + 2\operatorname{H}_2\operatorname{O}(g) = \operatorname{Si}(\operatorname{OH})_4(g) \tag{4}$$

Figure 2 shows the temperature dependence of the measured total oxidation of Si_3N_4 in 30% H₂O (0.54 atm) at a total pressure of 1.8 atm for 100 h, in comparison with the oxidation in a static dry air at a total pressure of 1 atm. At 1400° and 1500°C, the oxidation was enhanced in wet air in comparison with that in dry air. The enhanced oxidation of Si_3N_4 in wet air has been also observed by other researchers.⁸ The solubility of H₂O in SiO₂ is much higher than that of O₂, although the diffusivity of H₂O considering both solubility and diffusivity is much higher than that of O₂. The resultant permeability of H₂O considering both solubility and diffusivity is much higher than that of O₂. The oxidation reaction of Si_3N_4 was more sensitive to temperature in wet air than in dry air. Figure 3 shows the H₂O partial pressure dependence of the measured to-



Fig. 3. H₂O partial pressure (P_{H_2O}) dependence on the measured total oxidation (*t*) of silicon nitride (Si₃N₄) at 1400°C in the flowing air (2.45 cm/s) at a total pressure of 1.8 atm (**■**), 4.2 atm (**▲**), and 10 atm (**♦**) for 100 h. The total oxidation in a static dry air at 1400°C at a total pressure of 1 atm for 100 h was also shown (\bigcirc).

tal oxidation of Si_3N_4 at 1400°C at a total pressure of 1.8, 4.2, and 10 atm for 100 h. The dependence in dry air was also shown as a white circle. In Fig. 3, the measured total oxidation increased with increasing the H₂O partial pressure. The total pressure had little effect on the oxidation, although the data in the higher total pressures had a scattering.

Figure 4 shows the XRD patterns of Si_3N_4 before and after the oxidation in wet air at 1400°C for 100 h. As the H₂O partial pressure increased, more Lu₂Si₂O₇ and SiO₂ (cristobalite) phases were crystallized. SiO₂ phase was formed by oxidation of Si₃N₄, and Lu₂Si₂O₇ phase was formed by reaction between the SiO₂ phase and remaining Lu-related elements in the grain boundary regions. XRD patterns of Lu₂Si₂O₇ phase showed stronger preferred orientation of (*hk*0) plane, as the H₂O partial pressure increased. It indicates that Lu₂Si₂O₇ was easy to be crystallized anisotropically during the oxidation in wet air.

Figure 5 shows SEM micrographs of Si₃N₄ after the oxidation at 1300°-1500°C in 30% H₂O (0.54 atm) at a total pressure of 1.8 atm for 100 h. Several cracks were observed in specimens at each temperature, because SiO₂ phase was crystallized to form a cristobalite phase with larger thermal expansion coefficient than that of Si₃N₄ to produce thermal shrinkage in the cooling process (the phase transition of cristobalite phase from β to α also occurred to produce further thermal shrinkage). There was no distinct difference of the morphologies among the specimens at different temperatures. Figure 6 shows SEM micrographs of the surface of Si_3N_4 after the oxidation at 1400°C in 10%–50% H_2O (0.18–3 atm) at a total pressure of 1.8–10 atm for 100 h. The surface morphology was drastically changed by the oxidation conditions at a constant temperature (1400 $^{\circ}$ C). As the H₂O partial pressure increased, the amount of Lu₂Si₂O₇ particles (white colored) increased. This was also observed by XRD analyses. In addition, more pores were generated probably through the volatilization of SiO₂ scale (Eq. (4)) and/or the exhaust of N₂ gas (by Eqs. (2) and (3)). At $P_{\text{H}_2\text{O}} = 2.1$ atm ($P_{\text{total}} = 4.2$ atm (50%)), the surface was quite rough with plenty of pores. However, the number of the pores decreased as the total pressure increased at a constant water vapor content. At $P_{\text{total}} = 4.2 \text{ atm } (P_{\text{H}_2\text{O}} = 1.26 \text{ atm } (30\%)), \text{ fewer pores were observed than at } P_{\text{total}} = 1.8 \text{ atm } (P_{\text{H}_2\text{O}} = 0.9 \text{ atm } (50\%)). \text{ In }$ particular, at $P_{\text{total}} = 10 \text{ atm} (P_{\text{H}_2\text{O}} = 3.00 \text{ atm} (30\%))$, few pores were observed on the dense SiO_2 scale in comparison with at $P_{\text{total}} = 4.2 \text{ atm } (P_{\text{H}_2\text{O}} = 2.1 \text{ atm } (50\%))$. In fact, the measured total oxidation was almost the same with the calculated total one at $P_{\text{total}} = 10$ atm ($P_{\text{H}_2\text{O}} = 3.00$ atm (30%)) (Table II), indicating that almost no volatilization of SiO₂ scale occurred. It seemed that increasing total pressure suppressed the volatilization of SiO₂ phase, although it had little effect on the total oxidation at these oxidation test conditions. It is possible



Fig. 4. X-ray diffraction patterns of silicon nitride (Si₃N₄) before (1) and after (2–4) the oxidation at 1400°C for 100 h. ((2) *P*_{total} = 1.8 atm, *P*_{H₂O} = 0.18 atm (10%), (3) *P*_{total} = 4.2 atm, *P*_{H₂O} = 2.1 atm (50%), (4) *P*_{total} = 10 atm, *P*_{H₂O} = 3 atm (30%)). ■, β-Si₃N₄ (33-1160); ●, lute-tium disilicate (Lu₂Si₂O₇) (34-0509); ♥, SiO₂ (39-1425).



Fig. 5. Scanning electron micrographs of silicon nitride (Si₃N₄) after the oxidation at $1300^{\circ}-1500^{\circ}$ C in the flowing air (2.45 cm/s) containing 30% H₂O (0.54 atm) at a total pressure of 1.8 atm for 100 h.

to observe that increasing total pressure would decrease the total oxidation for longer oxidation test periods than 100 h (SiO₂ scale would prevent the further oxidation of Si₃N₄ by minimizing the volatilization of SiO₂).

Oxidation of Si₃N₄ was estimated to be about 4 μ m at 1400°C in 10% H₂O at a total pressure of 10 atm for 100 h from Fig. 3.

However, the gas velocity in the present research was very small (2.45 cm/s; STP) in comparison with that in the gas turbine conditions. The volatilization rate of SiO₂ was deduced to be proportional to square root of gas velocity.^{9,17} Assuming that the oxidation rate of Si₃N₄ was the same as the volatilization rate of SiO₂ and that the oxidation continued linearly with time,



Fig.6. Scanning electron micrographs of the surface of silicon nitride (Si_3N_4) after the oxidation at 1400°C in the flowing air (2.45 cm/s) containing 10%–50% H₂O (0.18–3 atm) at a total pressure of 1.8–10 atm for 100 h.



Fig. 7. Scanning electron micrographs of silicon nitride (Si₃N₄) coated with lutetium disilicate (Lu₂Si₂O₇) before (a, b) and after (c, d) the oxidation at 1400°C in the flowing air (2.45 cm/s) for 100 h ((c) $P_{total} = 1.8$ atm, $P_{H_2O} = 0.18$ atm (10%), (d) $P_{total} = 4.2$ atm, $P_{H_2O} = 2.1$ atm (50%)).

the total recession of Si_3N_4 would be roughly 2.6 cm at a gas velocity of 100 m/s for 10000 h (at 1400°C in 10% H₂O at a total pressure of 10 atm). The combustion gases for gas turbine also contain some amount of impurities such as sulfur, sodium, and potassium, which cause hot corrosion of Si_3N_4 .^{8,19} Therefore, it is quite necessary to fabricate EBC on the surface of Si_3N_4 for the use of gas turbine components.

(2) Effect of the $Lu_2Si_2O_7$ Coating

In order to avoid the oxidation of Si₃N₄ in wet air, it was coated with $Lu_2Si_2O_7$ layer. $Lu_2Si_2O_7$ has a high melting point and its thermal expansion coefficient is very close to that of Si_3N_4 .^{14,15} Therefore, it can be a candidate for material of EBC. Si₃N₄ coated with Lu₂Si₂O₇ through the intermediate SiO₂-rich phase (containing crisotobalite) was fabricated by reaction sintering after dip-coating. Figures 7(a) and (b) show the SEM micrographs of the coated Si_3N_4 . The Lu₂Si₂O₇ layer on Si_3N_4 substrate was about 15 μm in thickness. In our previous research where this coated Si₃N₄ was exposed to long-term cycling oxidation test in dry air at 1500°C for up to 1000 h, the total weight gain decreased by up to 51% (indicating that oxidation resistance improved by about 100%).¹⁶ In the present research, the oxidation resistance was examined in more severe conditions. Figure 7(c) shows the micrographs after the oxidation at 1400°C at $P_{\text{total}} = 1.8$ atm and $P_{\text{H}_2\text{O}} = 0.18$ atm (10%) for 100 h. There was no distinct evidence of oxidation, indicating that $Lu_2Si_2O_7$ layer protected the Si₃N₄ from the oxidation in wet air at this condition. However, at more severe conditions (containing larger amount of water vapor), Lu₂Si₂O₇ layer could not completely protect Si₃N₄ from oxidation. After the oxidation at 1400°C at



Fig.8. Flexural strength of the uncoated silicon nitride (Si_3N_4) and Si_3N_4 coated with lutetium disilicate $(Lu_2Si_2O_7)$ at room temperature before and after oxidation tests at $1300^\circ-1500^\circ$ C in the flowing air (2.45 cm/s) containing 10%-50% H₂O (0.18–3.0 atm) at a total pressure of 1.8–10 atm for 100 h.

 $P_{\text{total}} = 4.2$ atm and $P_{\text{H}_2\text{O}} = 2.1$ atm (50%) for 100 h, the more SiO2-rich (containing cristobalite) phase was generated between the coating layer and substrate. This was because the coating layer was not completely dense with several pores, which permitted oxygen and water to approach the Si₃N₄ substrate. Several pores were also observed in the SiO₂-rich phase. They formed probably through the volatilization of SiO₂ and/or the exhaust of oxidation product gas such as N2. Therefore, it is necessary to further develop coating process in order to fabricate completely dense coating layer without pores or SiO_2 phase for higher oxidation resistance in wet air.

Figure 8 shows flexural strengths of the uncoated Si₃N₄ and Si₃N₄ coated with Lu₂Si₂O₇ at room temperature before and after oxidation at 1300°-1500°C in 10%-50% H₂O (0.18-3.0 atm) at a total pressure of 1.8-10 atm for 100 h. Before the oxidation, the strength of the coated one $(493.8 \pm 43.9 \text{ MPa})$ was a little higher than that of the uncoated one $(467.4 \pm 116.3 \text{ MPa})$. It was attributed mainly to healing effects of surface and inside defects during the sintering process of the coating layer.²⁰ After the oxidation, the strength of the coated Si₃N₄ was increased a little in most cases (the average strength of all the coated specimens after oxidation was 531.0 ± 63.4 MPa), while that of the uncoated one was almost unchanged (487.8 ± 54.8 MPa). The small improvement at the coated Si₃N₄ was attributed probably to the further healing during the oxidation, which was conducted minimizing the formation of the defects by the coating layer.

IV. Conclusions

Oxidation behavior of Si₃N₄ was investigated in the flowing air (2.45 cm/s) containing 10%-50% H₂O at a total pressure of 1.8-10 atm at 1300°-1500°C. The oxidation of Si₃N₄ was enhanced by increasing H₂O partial pressure and by increasing temperature. Influence by H₂O partial pressure was larger than that by temperature. The total pressure had little effect on the oxidation, and suppressed the volatilization of SiO₂ scale at these oxidation test conditions. The oxidation was accelerated with volatilization of the oxidation scale in wet air. The surface morphology also drastically changed by the oxidation conditions.

In order to prevent the oxidation, Lu₂Si₂O₇ layer was coated on Si₃N₄ substrate through the intermediate SiO₂-rich phase by reaction sintering after dip-coating. Lu₂Si₂O₇ layer was about 15 µm in thickness. It was relatively dense with several pores, and strongly jointed to the substrate. Although this layer prevented the substrate from the oxidation in small amount of water vapor, it did not completely prevent the oxidation in case of the large amount. The more SiO₂-rich phase was generated between the coating layer and substrate. The traces of SiO₂ volatilization between the layer and substrate and exhaust of the oxidation product gas were observed. After the exposure to the air containing water vapor, the flexural strength of the coated Si₃N₄ was increased, while that of the uncoated one was almost unchanged. It was attributable probably to the further healing

of the substrate during the oxidation. It is necessary to further develop coating process in order to fabricate completely dense coating layer without pores or SiO₂ phase for higher oxidation resistance in wet air.

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