

Low pressure sintering of sialon using different sintering additives

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The sintering behavior and the mechanical and oxidation properties of Sialon were investigated by varying the composition, amount, and cationic radii of sintering additives composed of $\text{AlN-Er}_2\text{O}_3\text{-M}_2\text{O}_3$ ($\text{M} = \text{Y}$ or Sc). The optimum additive composition for the pressureless sintering of Sialon was determined to be $(0.6 \text{ AlN}:0.4 [0.3 \text{ Er}_2\text{O}_3:0.7 \text{ M}_2\text{O}_3])$ and the optimum content was 10–15 mass%. The maximum relative densities of Sialon using $\text{AlN-Er}_2\text{O}_3\text{-Y}_2\text{O}_3$ and $\text{AlN-Er}_2\text{O}_3\text{-Sc}_2\text{O}_3$ additives (termed SAEY and SAES) were 94.4 and 97.9% at 0.1 MPa N_2 , respectively. The pressureless sintered SAES sample showed fairly high strength (506 MPa) using the optimized additive composition. In both systems, the oxidation resistance was improved by decreasing the total amount of sintering additives.

Key-words: Sialon, Pressureless sintering, Rare-earth oxides, Strength, Oxidation

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1. Introduction

Silicon nitride is reputed to have good mechanical properties.^{1,2)} Sintering additives such as Al_2O_3 and Y_2O_3 have been added to silicon nitride to facilitate densification, which deteriorates the compound's high temperature properties due to the low softening temperature of the resultant grain boundary phases.³⁾

One of the representative methods of liquid phase sintering is to use AlN or Al_2O_3 as sintering additives for Si_3N_4 . In this method, the sintering additives promote the formation of a liquid phase, but only a small amount of the glass phase remains at the end because the additives are partly incorporated into Si_3N_4 grains to form Sialon.⁴⁾

High temperature stability and oxidation resistance of Sialon have been generally reported to be inferior to those of Si_3N_4 .³⁾ In order to improve the high temperature properties and oxidation resistance of Sialon, rare earth oxides have been added as sintering additives.⁵⁾ However, the high viscosity of the resultant liquid phases suppresses densification of Sialon and high pressures and temperatures have been required to obtain dense samples. In this respect, $\text{AlN-(R}_2\text{O}_3\text{-M}_2\text{O}_3)$ additives (R_2O_3 : rare earth oxide, M_2O_3 : metal oxide) present a possibility to improve the sintering behavior and oxidation resistance of Sialon.⁵⁾ Huang et al. reported enhanced sinterability (relative density of 99%) and high temperature strength (1150 MPa at 1200°C) in a $\text{Si}_3\text{N}_4\text{-AlN-L}_2\text{O}_3\text{-Y}_2\text{O}_3$ system using pressureless sintering.⁶⁾

Sialon sintered with Er_2O_3 has excellent high temperature strength and oxidation resistance.⁷⁾ However, densification of the Sialon was reported to be difficult due to the highly refractory grain boundary phase.⁷⁾ Application of a second additive may promote the densification by decreasing the eutectic temperature of the grain boundary phase. However,

studies of $\text{Si}_3\text{N}_4\text{-AlN-Er}_2\text{O}_3\text{-M}_2\text{O}_3$ (M : metal) systems are scarce.

In the present study, the sintering behavior and the mechanical and oxidation properties of Sialon ($\text{Si}_3\text{N}_4\text{-AlN-(Er}_2\text{O}_3\text{-M}_2\text{O}_3)$, M : Y or Sc) were investigated using various mixing ratios and amounts of sintering additives. The cations of the SAEY (Si_3N_4 , AlN , Er_2O_3 , Y_2O_3) system have similar radii (Er^{3+} : 0.1 nm, Y^{3+} : 0.101 nm) whereas those of the SAES (Si_3N_4 , AlN , Er_2O_3 , Sc_2O_3) system are different (Sc^{3+} : 0.0732 nm).⁸⁾⁻¹⁰⁾

2. Experimental procedure

$\alpha\text{-Si}_3\text{N}_4$ (SN-E10, UBE), AlN (Grade C, HCST), Y_2O_3 (99.99%, Ventron), Sc_2O_3 (99.9%, Reacton), and Er_2O_3 (99.9%, STREM) were used as starting materials.

After weighing, the raw powders were intensively mixed for 2 h using a planetary mill with isopropanol and SiC balls. The slurry was dried using a vacuum rotary evaporator at 60°C and the mixed powder was then sieved (125 μm) to minimize agglomeration. Finally, the powder was compacted into bars (80 × 14 × 16 mm) using a cold isostatic press (CIP, 240 MPa, 3 min).

To determine the optimal composition and additive amount for the densification and properties of the SAEY and SAES systems, three series of sintering experiments were performed at 1,800°C for 3 h under 0.5 MPa N_2 . In the first series, the $\text{Er}_2\text{O}_3\text{:M}_2\text{O}_3$ ratio ($\text{M} = \text{Y}, \text{Sc}$) was varied (0.9:0.1, 0.7:0.3, 0.5:0.5, 0.3:0.7, and 0.1:0.9) while the $\text{AlN:(Er}_2\text{O}_3 + \text{M}_2\text{O}_3)$ ratio was fixed at 0.6:0.4. The total amount of additives was 20 wt%. In the second series, the $\text{AlN:(Er}_2\text{O}_3 + \text{M}_2\text{O}_3)$ ratio was varied (0.9:0.1, 0.7:0.3, 0.6:0.4, 0.4:0.6, and 0.2:0.8) with a fixed additive content (20 mass%) and fixed $\text{Er}_2\text{O}_3\text{:M}_2\text{O}_3$ ratio based on the results of the first series. In the third series, the amount of sintering additive (5, 10, 15, 20, 25, and 30 mass%) was varied while using fixed $\text{Er}_2\text{O}_3\text{:M}_2\text{O}_3$ and $\text{AlN:(Er}_2\text{O}_3 + \text{M}_2\text{O}_3)$ ratios based on results of the first and the second series. The optimum composition was determined by measuring the sintered density of the samples using Archimedes' method.

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Based on the results, pressureless sintering (0.1 MPa N_2) was carried out at 1750°C and 1800°C for 3 h using the optimized additive composition. In all cases, a powder bed ($Si_3N_4:BN=6:4$) was used to enhance the densification during sintering.¹¹⁾

To observe the formation of secondary phases, the samples were analyzed by x-ray diffraction (XRD, D5000, Siemens) with $CuK\alpha$ radiation. The effect of the cationic radius of the sintering additives on the lattice parameter of α -Sialon was measured using 2θ values of the (201) and (102) reflection of Sialon.¹²⁾

The microstructure of the samples was observed using scanning electron microscopy (SEM, JEOL, JSM-6300F, Japan) after plasma etching (RF plasma barrel etcher, Bio-Rad).

For the measurement of mechanical properties, the sintered samples were cut into $50 \times 3 \times 4$ mm sized bars according to the ASTM standard and polished to a $3 \mu m$ finish.¹³⁾ The four point bending strength was measured using a Zwick 1476 universal testing machine (5 specimens per condition, upper span of 7 mm, lower span of 20 mm). Young's modulus, hardness, and fracture toughness of the samples were measured using an ultrasonic tester (5072PR, Panametrics) and a Vickers indenter (AVK-A, Akashi, loading condition: 10 kg, 15 s).

The surface of samples was polished to a $1 \mu m$ finish to measure the weight gain after oxidation in air for 72 h at 1000°C, 1200°C, and 1400°C using a box furnace.

3. Results and discussion

3.1 Sintering behavior

Table 1 shows the relative densities of the sintered samples in this investigation. In the first series, the relative density decreased with the amount of Er_2O_3 in both the systems due to the high eutectic temperature of the Er_2O_3 - SiO_2 system (1680°C) except for a slight improvement of the SAES A1 system.⁹⁾

Huang et al. reported that the sinterability of Si_3N_4 was improved by the application of a La_2O_3 and Y_2O_3 mixture with AlN.¹⁴⁾ La_2O_3 and Y_2O_3 react to form compounds (La_3YO_6 , $LaYO_3$) and the eutectic temperature of the La_2O_3 - Y_2O_3 system is much lower than the melting temperature of each component (1570°C vs. 2250°C).¹⁵⁾ On the other hand, Er_2O_3 was reported to form a complete solid solution with Sc_2O_3 .¹⁶⁾ The differences between the La_2O_3 - Y_2O_3 and Er_2O_3 - Sc_2O_3 systems is believed to induce the discrepancy between the results of Huang et al. and the present research.

The Y_2O_3 - Si_3N_4 system maintains its strength at high temperatures and shows high oxidation resistance, but the application of the system has been limited partly due to deterioration during long-term oxidation at 1000°C.¹⁷⁾ The deterioration has been attributed to volume expansion of secondary phases such as $Y_2Si_3O_3N_4$. One of the methods to solve this problem is the addition of rare earth oxides.¹⁸⁾ For this reason, the optimum composition in the first series was chosen as $Er_2O_3:M_2O_3=0.3:0.7$ despite the slight decrease of the sintered density compared to the samples containing lower amounts of Er_2O_3 .

In the second series, the densities of the sintered samples were measured while varying the AlN: ($Er_2O_3 + M_2O_3$) ratio. In this series, the $Er_2O_3:M_2O_3$ ratio was fixed at 0.3:0.7 based on the results of the first series. In both systems, the densification was improved with increasing AlN content up to 60

Table 1. Relative Densities (RD) of the SAEY and SAES System Samples after Sintering at 1,800°C for 3 h under 0.5 MPa N_2

Series	SAEY	Mixing ratio by weight	RD (%)	SAES	Mixing ratio by weight	RD (%)
1 st Series	A0	0.0 : 1.0 = $Er_2O_3 : Y_2O_3$	96.0	A0	0.0 : 1.0 = $Er_2O_3 : Sc_2O_3$	98.3
	A1	0.1 : 0.9 = $Er_2O_3 : Y_2O_3$	93.6	A1	0.1 : 0.9 = $Er_2O_3 : Sc_2O_3$	98.6
	A2	0.3 : 0.7 = $Er_2O_3 : Y_2O_3$	93.6	A2	0.3 : 0.7 = $Er_2O_3 : Sc_2O_3$	97.4
	A3	0.5 : 0.5 = $Er_2O_3 : Y_2O_3$	85.5	A3	0.5 : 0.5 = $Er_2O_3 : Sc_2O_3$	93.6
	A4	0.7 : 0.3 = $Er_2O_3 : Y_2O_3$	75.4	A4	0.7 : 0.3 = $Er_2O_3 : Sc_2O_3$	90.8
	A5	0.9 : 0.1 = $Er_2O_3 : Y_2O_3$	69.8	A5	0.9 : 0.1 = $Er_2O_3 : Sc_2O_3$	75.1
2 nd Series	A6	1.0 : 0.0 = $Er_2O_3 : Y_2O_3$	71.3	A6	1.0 : 0.0 = $Er_2O_3 : Sc_2O_3$	71.3
	B1	0.8 : 0.2 = $EY : AlN$	87.1	B1	0.8 : 0.2 = $ES : AlN$	89.0
	B2	0.6 : 0.4 = $EY : AlN$	90.2	B2	0.6 : 0.4 = $ES : AlN$	92.3
	B3	0.4 : 0.6 = $EY : AlN$	93.6	B3	0.4 : 0.6 = $ES : AlN$	97.4
	B4	0.2 : 0.8 = $EY : AlN$	83.1	B4	0.2 : 0.8 = $ES : AlN$	86.5
3 rd Series	B5	0.1 : 0.9 = $EY : AlN$	75.8	B5	0.1 : 0.9 = $ES : AlN$	86.1
	C1	5 mass% AEY	87.2	C1	5 mass% AES	99.1
	C2	10 mass% AEY	93.9	C2	10 mass% AES	98.4
	C3	15 mass% AEY	94.5	C3	15 mass% AES	97.8
	C4	20 mass% AEY	93.6	C4	20 mass% AES	97.4
	C5	25 mass% AEY	94.1	C5	25 mass% AES	96.7
	C6	30 mass% AEY	93.4	C6	30 mass% AES	96.1

mass%. Both Al_2O_3 - Y_2O_3 and Al_2O_3 - Sc_2O_3 have eutectic points near this composition (60 mass% AlN), which explains the high relative density of Sialon with the B3 composition in Table 1.^{19),20)} Accordingly, the optimum composition in the second series was determined as AlN: ($Er_2O_3 + M_2O_3$) = 0.6:0.4 (B3 in Table 1).

In the third series, the relative density of the SAEY system was nearly constant when the amount of additives was at least 10 mass%, but that of the SAES samples decreased gradually with an increase of the total amount of additives.

The deterioration of Si_3N_4 at high temperatures has been attributed to the softening of an amorphous grain boundary phase. Therefore, many studies have been performed to minimize the residual amorphous phase after sintering.²¹⁾ Decreasing the amount of sintering additive is one possible solution to reduce the amorphous phase.²²⁾ Consequently, the optimum compositions of the SAEY and SAES systems in that respect are C2 (10 mass%) and C3 (15 mass%), respectively, as shown in Table 1.

The samples having these optimum compositions were pressureless sintered at 1750°C or 1800°C under 0.1 MPa N_2 for 3 h (**Table 2**). Fused Si was formed at the surface of the C2 samples sintered at 1800°C. At 0.1 MPa N_2 , the decomposition of silicon nitride ($Si_3N_4 \rightarrow 3Si(l) + N_2$) has been reported to occur above 1841°C,²³⁾ while the phenomenon was observed at 1800°C in the present investigation. This is due to the lower decomposition temperature of Sialon compared to Si_3N_4 . Mitomo and Fukunaga reported the decomposition reaction of Y-Sialon under 0.1 MPa N_2 as follows:²⁴⁾

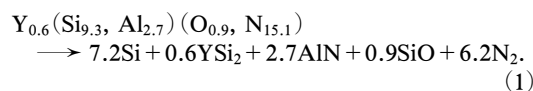


Table 2. Relative Densities (RD) of the SAEY and SAES System Samples Prepared under Different Sintering Conditions using Optimized Compositions (C2 and C3 in Table 1)

Sintering Condition	SAEY	RD (%)	SAES	RD (%)
1750°C, 0.1 MPa N ₂ gas, 3 h	C2	93.6	C2	97.9
1800°C, 0.1 MPa N ₂ gas, 3 h	C3	94.4	C3	96.8
1800°C, 0.5 MPa N ₂ gas, 3 h	C2	93.9	C2	98.4
1800°C, 0.5 MPa N ₂ gas, 3 h	C3	94.5	C3	97.8

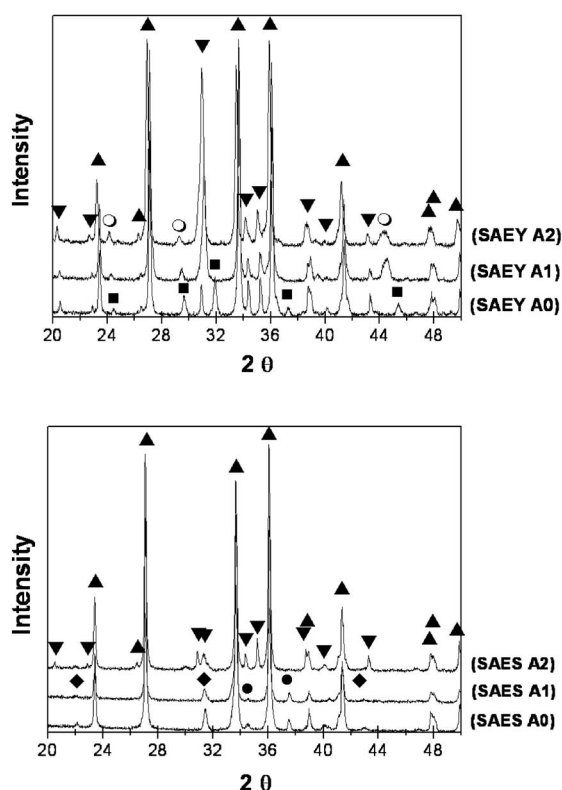


Fig. 1. XRD patterns of the samples after sintering at 1800°C for 3 h under 0.5 MPa N₂ (▼: α -Sialon, ▲: β -Sialon, ■: Y₂Si₃O₅N₄, ○: Y₄SiAlON, ◆: SiO₂, ●: Al₃SiO₂N₃).

They observed that reaction (1) began at 1700°C and became intense at 1750°C and above. The present results indicate that Sialon containing Er₂O₃ and Sc₂O₃ decomposes rather distinctly at 1800°C under 0.1 MPa N₂ pressure.

The high temperature properties of Si₃N₄ deteriorate above 1400°C if Si is present in the sample.²⁵⁾ Accordingly, the C2 composition was pressureless sintered at 1750°C. Fused Si was not formed under this condition and the relative density was comparable to that of samples sintered at 1800°C under 0.5 MPa of N₂ pressure (Table 2). By the optimization of the additive conditions, Sialon containing refractory rare earth oxide additives could be densified under 0.1 MPa of N₂ pressure without suffering from decomposition.

3.2 Microstructure analysis

After sintering, the secondary phases of the SAEY and SAES systems were analyzed by XRD (Fig. 1). β -Sialon was

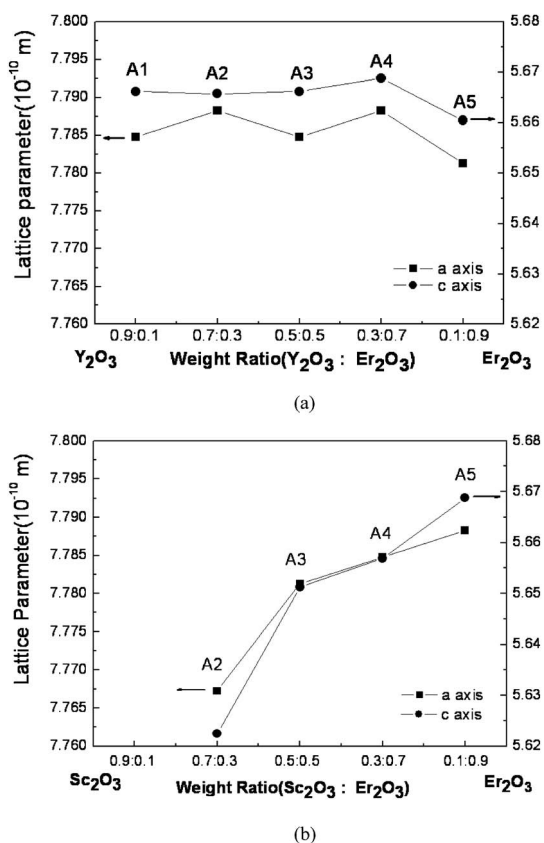


Fig. 2. Lattice parameters of α -Sialon in the (a) SAEY and (b) SAES system samples after sintering at 1800°C for 3 h under 0.5 MPa N₂.

the main phase in all cases and α -Sialon was also observed under some conditions. Watari et al. reported that the phase transformation from α - to β -Sialon was enhanced when using Y₂O₃ instead of Er₂O₃ because the liquidus temperature and viscosity of Er₂O₃-containing phases are higher than those containing Y₂O₃.²⁶⁾ However, the intensities of reflection for α -Sialon were not strongly affected by the Er₂O₃:Y₂O₃ ratio in the present investigation (Fig. 1). On the other hand, α -Sialon was observed in the SAES system when the amount of Er₂O₃ was at least 30 mass%. The addition of refractory Er₂O₃ is believed to increase the viscosity of the grain boundary phase of the SAES system. As a result, the relative density of the system and the amount of β -Sialon decreased with increases of the amount of Er₂O₃ in the SAES system.

Figure 2 shows the variation of the lattice parameters of α -Sialon with increasing Er₂O₃:M₂O₃ ratio. The lattice parameters of the SAES system increased proportionally to the Er₂O₃ content. In contrast to the SAES system, the lattice parameters in the SAEY system did not vary significantly. The ionic radii of Y³⁺ and Er³⁺ are very similar (Er³⁺: 0.1 nm, Y³⁺: 0.101 nm) and, accordingly, the change of the Er³⁺:Y³⁺ ratio did not influence the lattice parameters of α -Sialon.^{7),8)} On the other hand, the lattice parameters of the SAES system were strongly affected by the mixing ratio due to the different ionic radii of Er³⁺ and Sc³⁺ (Sc³⁺: 0.0732 nm).⁹⁾ The lattice parameters of β -Sialon were reported to be not affected by the mixing ratio, because the

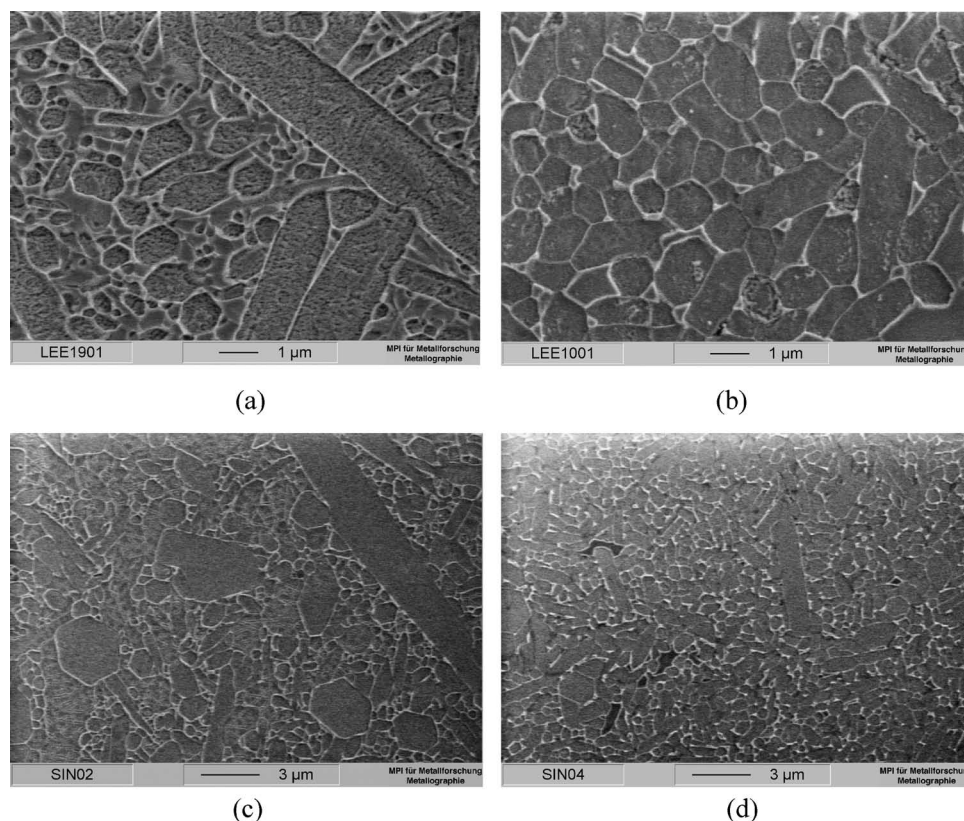


Fig. 3. Microstructures of the samples after sintering at 1800°C for 3 h under 0.5 MPa N₂: (a) SAEY A1, (b) SAES A1, (c) SAEY A3 and (d) SAES A3.

additives are not incorporated in the grain.²⁷⁾

Figure 3 shows the microstructures of the SAEY and SAES materials. Elongated β -Sialon grains were observed in the SAEY samples and the microstructure was not strongly affected by the Er₂O₃:Y₂O₃ ratio (Fig. 3(a), (c)). In contrast, the formation of elongated β -Sialon grains was suppressed with increasing amounts of Sc₂O₃ in the SAES samples (Fig. 3(b), (d)). Both Y₂O₃ and Er₂O₃ have been reported to promote the elongation of the Si₃N₄ and Sialon grains.^{28),29)} Thus, the morphology of the Sialon grains in Fig. 3 was not strongly affected by the mixing ratio of the two oxide additives. Elongated grains were also observed in Si₃N₄ sintered with Sc₂O₃,²⁸⁾ but the present results indicate that among Er₂O₃ and Sc₂O₃, the former induces more distinct elongation of the Sialon grains.

The amount of liquid phase also affected the microstructure of Sialon (**Fig. 4**). Lee et al. reported that the increase of the quantity of sintering additives promoted the formation of the β -phase and increased its aspect ratio, which enhanced the strength and fracture toughness of Si₃N₄.³⁰⁾ In the present investigation, enhancement of grain growth did not occur significantly in the SAES system with additive amounts up to 15 mass%. On the other hand, strong grain growth occurred when the additive content was above this value (Fig. 4(d)).

3.3 Mechanical properties

The strength of the SAEY samples was not tested due to their low relative density. **Fig. 5** shows the strength of the SAES samples with various Er₂O₃:Sc₂O₃ ratios and additive

amounts. The relative density and strength are directly related to each other except for the sample A1 (Fig. 5(a)). The A1 sample was rather dense (relative density, RD: 98.6%), while its strength was lower than that of the rather porous SAES A3 sample (RD: 93.6%). This result suggests that the microstructure (Fig. 3(b), (d)) and/or composition of the glass phase influence the strength of the SAES samples.

Although the amount of liquid phase increased rapidly and grain growth became distinct when the additive content was above 15 mass% (Fig. 4), such changes did not have a strong influence on the SAES sample strengths (Fig. 5(b)). Since the composition of the additives was the same in the C1-C5 SAES samples, the result also indicates that the low strength of the SAES A1 sample described above is more strongly attributed to the composition of the glass phase rather than to the microstructural change.

Table 3 shows some mechanical properties of the SEAS samples sintered under 0.1 or 0.5 MPa N₂. The Young's modulus, hardness, and strength of the pressureless sintered samples were slightly lower than that of the over-pressure sintered samples, presumably due to their lower relative densities. The fracture toughnesses of the samples were 5.2–6.2 MPa·m^{1/2}. The pressureless sintered samples had higher fracture toughness values than those sintered under 0.5 MPa N₂.

Recently, Zhou et al. reported the pressureless sintering of Sialon at 1750–1900°C with Al₂O₃ and Y₂O₃ additives.²⁹⁾ In their report, the fracture toughness and hardness of the samples sintered at 1850°C were 6.2 MPa·m^{1/2} and 17.6 GPa, respectively. However, a Sialon with similar mechanical

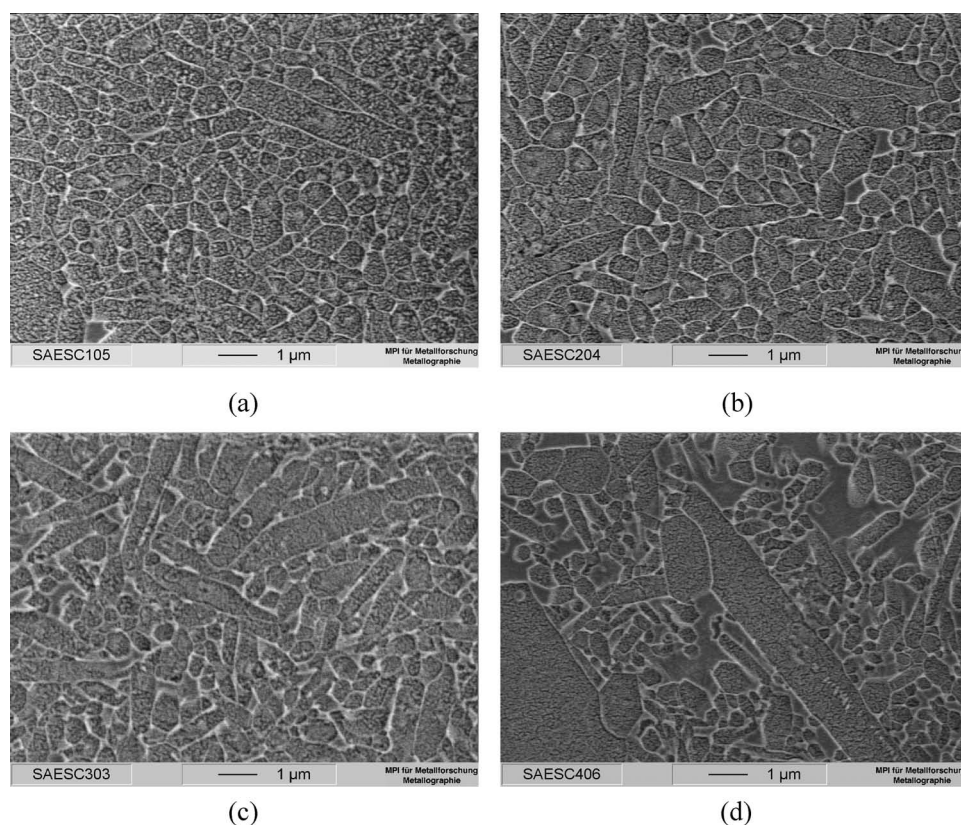


Fig. 4. Microstructures of the SAES system samples sintered at 1800°C for 3 h under 0.5 MPa N₂ with (a) 5, (b) 10, (c) 15 and (d) 20 mass% additives (composition C1–C4 in Table 1).

properties could be obtained by pressureless sintering at 1750°C in the present investigation after the optimization of the additive conditions.

3.4 Oxidation behavior

Figure 6 shows the weight gain of samples after oxidation in air at different temperatures. Weight gain by oxidation was not clearly observed in the SAEY samples up to 1200°C in spite of their high porosity. At 1400°C, however, their weight increased rapidly. They have open porosity due to their low relative density. Once oxidation begins, the weight gain of porous samples is expected to be faster than that of dense bodies due to the high internal surface area. Raj et al. reported that porous silicon nitride suffered from rapid weight gain during oxidation until the pores were closed due to volume expansion during the oxidation of Si₃N₄.³¹⁾

The SAES materials gained weight at 1200°C (Fig. 6(a)). The SAES A0 sample suffered from rather rapid oxidation at this temperature, while moderate oxidation was observed in the SAES A2 system even at 1400°C. Er₂O₃ clearly improved the oxidation resistance of the SAES materials.

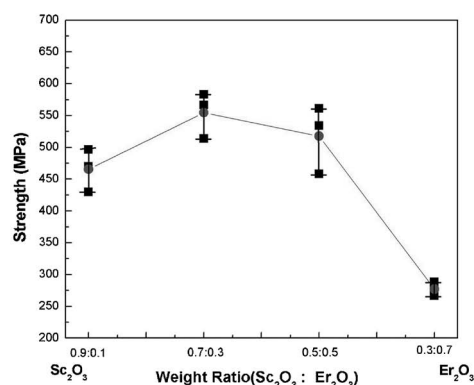
The amount of sintering additive also affected the oxidation behavior of the Sialon samples tested in the present investigation (Fig. 6(b)). The weight gain by oxidation became distinct with the increase of the additive content. The oxidation behavior of the SAEY materials was more susceptible to the amount of liquid phase than that of the SAES system at 1400°C.

Cristobalite (SiO₂) was the only phase observed in the silicate layer formed on the SAEY A1 sample after oxidation.

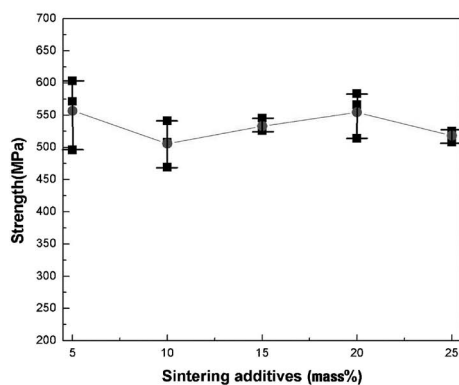
In contrast, β -Sialon and Sc₂SiO₇ as well as cristobalite were detected in the silicate layer of the SAES A1 sample (**Fig. 7**). A Sialon phase was not detected in the SAEY samples because the silicate layer was very thick. Intensive bubble formation was observed in the silicate layer of the SAEY materials.

The SAEY samples were heavily damaged after oxidation at 1400°C. In contrast, the silicate layer formed on the SAES system was thin and dense. Microcracks were formed beneath the silicate layer parallel to the surface in the oxidized SAES and SAEY samples (**Fig. 8**). The thickness of the damaged area was about 100 μm. Similar damage was also observed by Lee et al. after the oxidation of Sialon using MgO, Al₂O₃, and Y₂O₃ as sintering additives.³²⁾ O'Meara and Sjöberg reported that the oxidation reaction was not restricted to the silicate/Si₃N₄ interface, but occurred simultaneously in the sub-scale region, which constitutes a very large oxidation reaction zone.³³⁾ They also reported that pores were detected both in the scale and in the sub-scale regions, which is consistent with results of the present investigation.

The oxidation resistance of the pressureless sintered Sialon was improved by the addition of Er₂O₃. Among the tested samples, the SAES C2 showed the best oxidation resistance. However, although the weight gain of the SAES C2 sample by oxidation was not significant at 1400°C, the sample suffered from damage at the sub-scale region. The result indicates that the mechanical properties of the samples may decrease at 1400°C in air. Further investigation is required to determine the maximum temperature for the application of the Sialon prepared in the present research.



(a)



(b)

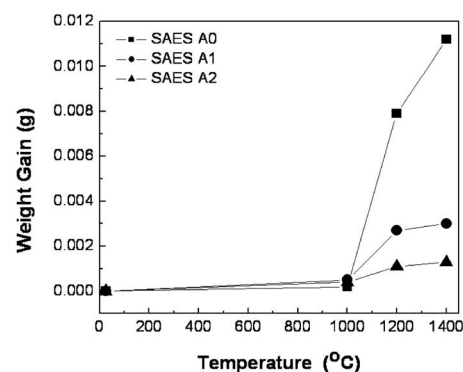
Fig. 5. Flexural strengths of the SAES system samples with variations of the (a) Sc₂O₃:Er₂O₃ ratio (A1–A4 in Table 1) and the (b) amount of sintering additives (C1–C5 in Table 1).

Table 3. Flexural Strengths of the SAES System Samples Prepared using Different Sintering Conditions and Optimized Compositions

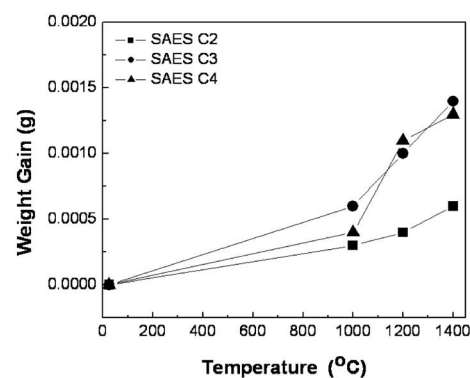
Sintering Condition	Composition	Young's modulus (GPa)	Hardness (GPa)	Strength (MPa)	Fracture toughness (MPa·m ^{1/2})
1750°C, 0.1 MPa N ₂ gas, 3 h	C2	310	15.6	468.8	6.1
1800°C, 0.1 MPa N ₂ gas, 3 h	C3	299	15.4	517.1	6.2
1800°C, 0.5 MPa N ₂ gas, 3 h	C2	310.8	16.0	505.8	5.7
1800°C, 0.5 MPa N ₂ gas, 3 h	C3	313	16.6	532.9	5.2

4. Conclusions

Sintering, and the mechanical and oxidation properties of Sialon were investigated while varying the composition, amount, and ionic radii of sintering additives. By optimizing the additive composition, Sialon with a fair relative density (97.9%) and strength (517 MPa) was obtained using pressureless sintering. The lattice parameters of α -Sialon in the SAES system were strongly affected by the mixing ratio of Er₂O₃ and Sc₂O₃, while such behavior was not observed in the SAEY system due to the similarity of the cationic radii of Er³⁺ and Y³⁺. The strength of the SAES system was affected by the Er₂O₃:Sc₂O₃ ratio, but was not strongly influenced by the additive content. In both systems, the oxidation resis-



(a)



(b)

Fig. 6. Weight gains of the SAES system samples with variations of the (a) composition and the (b) amount of the additives after oxidation at different temperatures for 72 h in air.

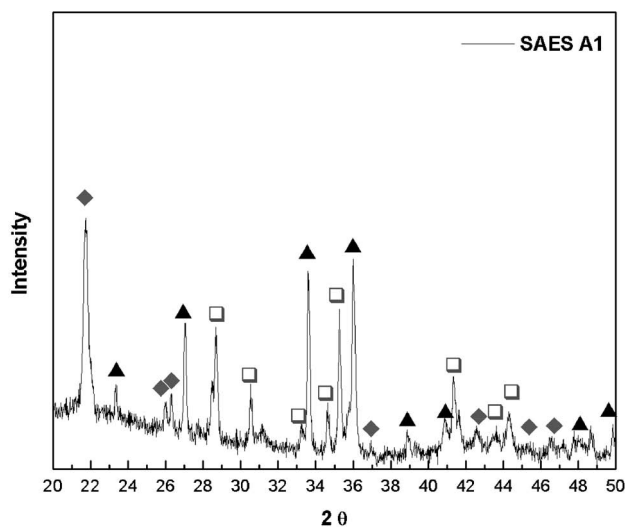


Fig. 7. XRD patterns of the SAES A1 sample after oxidation at 1400°C for 72 h in air. (◆: SiO₂ (cristobalite), ▲: β -Sialon, □: Sc₂SiO₇).

tance was improved with increasing the mixing ratio of Er₂O₃ in spite of a decrease in the relative density. The weight gain by oxidation was reduced when the amount of sintering additives was decreased.

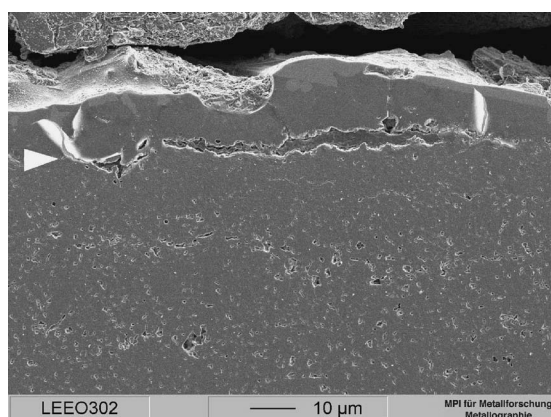


Fig. 8. Cross-sectional morphology of the SAES A1 system samples after oxidation at 1400°C for 72 h in air.

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References

- 1) R. Raj, *J. Am. Ceram. Soc.*, **76**, 2147–2174 (1993).
- 2) H. Gu, *J. Am. Ceram. Soc.*, **85**, 33–37 (2002).
- 3) H. J. Choi, J. G. Lee and Y. W. Kim, *J. Eur. Ceram. Soc.*, **19**, 2757–2762 (1999).
- 4) T. Y. Tien, "Phase Diagrams in Advanced Ceramics," Ed. By A. M. Alper, Academic Press, San Diego (1995) pp. 127–156.
- 5) W. S. Cho and K. Hayashi, *J. Ceram. Soc. Japan*, **100**, 936–940 (1992).
- 6) Z. K. Huang, T. Y. Tien and T. S. Yen, *J. Am. Ceram. Soc.*, **69**, C241–C242 (1986).
- 7) M. K. Cinibulk, G. Thomas and S. N. Johnosn, *J. Am. Ceram. Soc.*, **75**, pp. 2050–2055 (1992).
- 8) Y. I. Lee and Y. W. Kim, *J. Mater. Sci.*, **36**, 699–702 (2001).
- 9) F. F. Lange, *J. Am. Ceram. Soc.*, **62**, 428–430 (1979).
- 10) H. Y. Choi, Y. W. Kim and J. G. Lee, *J. Kor. Ceram. Soc.*, **31**, 777–783 (1994).
- 11) S. H. Lee, G. Rixecker, F. Aldinger, S. C. Choi and K. H. Auh, *J. Mater. Res.*, **17**, 465–472 (2002).
- 12) C. P. Gazzara and D. R. Messier, *Am. Ceram. Soc. Bull.*, **56**, 777–780 (1977).
- 13) ASTM C1161–90, 1991, Standard test method for flexural strength of advanced ceramics at ambient temperature.
- 14) Z. K. Huang, A. Rosenflanz and I. W. Chen, *J. Am. Ceram. Soc.*, **80**, 1256–1262 (1997).
- 15) J. Cassedanne and H. Forestier, *G. R. Acid. Sci.*, **253**, 2594–2596 (1961).
- 16) S. J. Schneider, R. S. Roth and J. L. Warig, *J. Research Natl. Bur. Standards*, **65A**, 370–373 (1961).
- 17) J. K. Patel and P. D. Thompson, *Br. Ceram. Trans. J.*, **87**, 70–73 (1988).
- 18) D. M. Mieskowski and W. A. Sanders, *J. Am. Ceram. Soc.*, **68**, C160–C163 (1985).
- 19) "Phase Diagrams for Ceramists–1969 supplement," Ed. by E. M. Levin, C. R. Robbins and H. F. McMurdie, The American Ceramic Society, Columbus (1969) Fig. 2343–2344.
- 20) "Phase Diagrams for Ceramists Vol. 6," Ed. by R. S. Roth, J. R. Dennis and H. F. McMurdie, The American Ceramic Society, Columbus (1987) Fig. 6435.
- 21) S. M. Wiederhorn, B. A. Fields and B. J. Hockey, *Mater. Sci. Eng.*, **A176**, 51–60 (1994).
- 22) Y. Murakami, K. Akiyama, H. Yamamoto and H. Sakata, *J. Ceram. Soc. Japan*, **106**, 35–40 (1998).
- 23) J. Peng, "Thermochemistry and constitution of precursor-derived Si–(B)–C–N ceramics," Ph. D. Thesis, University of Stuttgart, Stuttgart (2002) pp. 42–43.
- 24) M. Mitomo and O. Fukunaga, *J. Ceram. Soc. Japan.*, **89**, 631–633 (1981).
- 25) B. T. Lee, J. H. Yoo and H. D. Kim, *Mater. Sci. Eng.*, **A333**, 306–313 (2002).
- 26) K. Watari, M. Yasuoka, C. Valecillos and S. Kanzaki, *J. Eur. Ceram. Soc.*, **15**, 173–184 (1995).
- 27) V. A. Izhevskiy, L. A. Genova, J. C. Bressiani and F. Aldinger, *J. Eur. Ceram. Soc.*, **20**, 1975–1985 (2000).
- 28) H. J. Choi, J. G. Lee and Y. W. Kim, *J. Mater. Sci.*, **32**, 1937–1942 (1997).
- 29) C. R. Zhou, Z. B. Yu and V. D. Krstic, *J. Eur. Ceram. Soc.*, **27**, 437–443 (2007).
- 30) Y. I. Lee, Y. W. Kim, H. J. Choi and J. G. Lee, *J. Mater. Sci.*, **36**, 699–702 (2001).
- 31) R. Raj, L. An, S. Shah, R. Riedel, C. Fasel and H. J. Kleebe, *J. Am. Ceram. Soc.*, **84**, 1803–1805 (2001).
- 32) S. H. Lee, G. Rixecker, F. Aldinger, S. C. Choi and K. H. Auh, *J. Mater. Sci. Lett.*, **20**, 2145–2148 (2001).
- 33) C. O. Orneare and J. Sjöberg, *J. Am. Ceram. Soc.*, **80**, 1491–1500 (1997).