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Influence of starting material composition and carbon content on the preparation of Mg-α SiAlON powders by carbothermal reduction-nitridation

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Abstract

This paper investigated the synthesis of Mg–α SiAlON from Talc ($Mg_3(Si_2O_5)_2(OH)_2$) and halloysite clay ($Al_2Si_2O_5(OH)_4$). Talc, halloysite and carbon black as a reducing agent were used as the starting materials, and 3 wt.% α-Si_3N_4 was added as the seed in all cases. The mixtures were heated in flowing N_2 (gas) to synthesize Mg–α SiAlON powders. The chosen molecular ratios of talc to halloysite were 1.0, 1.5, 2.0 and 2.5. The influences of various reaction parameters such as the carbon content, temperature and holding time at the top temperature were studied. The results showed that the synthesized powders were composed of α-SiAlON, β-SiAlON, β-SiC, 15R-AlN polytypoid and AlN phases; the phases and the particle morphology greatly depended on starting material composition and synthesis parameters. A larger amount of talc was needed to compensate for Mg evaporation loss in the starting composition. Higher carbon content seemed to retard the reaction rate, resulting in coarse particle size with an irregular grain shape. The highest content of Mg–α SiAlON, 90 wt.%, was achieved at 1480 °C for 4 h at talc to halloysite ratios of 1.5 and 2.0. © 2002 Elsevier Science Ltd. All rights reserved.

Keywords: Carbothermal reduction-nitridation; Clay; Powder preparation; SiAlON; Talc

1. Introduction

SiAlON ceramics are expected to have high hardness, high strength even at elevated temperatures, good corrosion resistance and outstanding thermal shock resistance owing to their lower thermal expansion coefficients compared to other ceramics. There are two main types of SiAlON ceramics used for engineering applications. One, β-SiAlON (β′), is derived from a β-Si_3N_4 structure developed as the first member in the SiAlON family. It has commonly been described by the formula of $Si_{6-z}Al_zO_zN_{8-z}$ ($0<z\leqslant 4.2$). The other is α-SiAlON (α’), which is generally characterized as $M^{\nu+}{}_xSi_{12-m-n}Al_{m+n}O_nN_{16-n}$, where M is a metal cation and x is related to its valence ν according to the relationship of $x=m/\nu$. The structure of α-SiAlON is based on α-Si_3N_4 where Si^{4+} is partially replaced by Al^{3+}. At the same time, charge compensation is achieved by the accommodation of other ions, such as Li^{3+}, Ca^{2+}, Y^{3+}, Mg^{2+} or other rare-earth ions getting into its unit cell.[1–3]

Typically, the preparation of SiAlON ceramics involves the use of high-purity silicon nitride and various additives. For example, β-SiAlON is prepared from compositions in the Si_3N_4–AlN–Al_2O_3 system.[4] On the other hand, α-SiAlON can be obtained from compositions in the Si_3N_4–AlN–Al_2O_3–M_xO_y system, where M is referred to as a metal ion entering into the α-SiAlON unit to stabilize the structure. Although SiAlON ceramics have already been applied practically, their high temperature characteristics are still unsatisfactory and cost reduction is an important subject that has been left behind yet. Therefore the search for alternative synthesis routes or new SiAlON materials is still being pursued intensively.

In 1979, Lee and Cutler[5] first successfully synthesized β-SiAlON from kaolinite ($Al_2Si_2O_5(OH)_4$) by the carbothermal reduction-nitridation (CRN) method.[5] Since then, a wide range of other inexpensive mineral

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materials have been tried in a similar way of synthesizing β-SiAlON, such as halloysite,[6–9] montmorillonite-polyacrylonitrile,[10–12] and bentonites.[13,14] The results show that fine β-SiAlON powders was obtained from most of these raw materials. The composition of β-SiAlON i.e. the *z* value in its formula, was confirmed to depend on the amount of Si loss through the intermediate gas phase of SiO(g) around 3.2 under various conditions.[15] Some by-products, for example SiC,[8–11,16–20] AlN,[5,6,9–11,21] AlN polytypoid[5,9,15,18–19,21–22] and Al_2O_3,[19–21] were also found in the final products, which makes controlling reaction parameters to eliminate such impurities extremely important.

Compared with the numerous publications on β-SiAlON, the synthesis of α-SiAlON by CRN can hardly be found in the literature. Mitomo et al.[23] reported that the single-phase Ca–α SiAlON powder was accelerated at 1450 °C by additional heating at 1550 °C for 1 h in the $CaO–SiO_2–Al_2O_3$ system with an excess of 20% carbon. Y–α SiAlON, which was mentioned in the same paper, first formed in the $Y_2O_3–SiO_2–Al_2O_3$ system at 1300 to 1450 °C and was then homogenized by subsequent heat-treatment at 1600 °C for 1 h. In their work, relatively expensive starting materials such as tetraethyl-ortho-silicate, aluminium-isopropoxide, calcium-ethoxide and yttrium nitrate were used. A similar conclusion was obtained by Van Rutten et al.[24] by using SiO_2, Al_2O_3 and $CaSiO_3$ or $CaCO_3$ as source materials with the theoretical stoichiometric amount of carbon. Nearly pure Ca–α SiAlON of 95 wt.% was successfully synthesized at 1500 °C for 65 h at 0.5 l/min nitrogen-flow rate.

Recently, Ekstrom et al.[25] reported that Y–α SiAlON powders were synthesized from halloysite with pure Y_2O_3 doped with SiO_2 at 1475 °C for 8 h, or doped with Si but pre-reacted at 1350 °C for 4 h and then fired at 1475 °C for an additional 8 h. The amount of carbon of a 10% excess corresponding to complete CRN reaction was prepared for the starting composition. The synthesized powders contained a mixture of crystalline α-, β-SiAlON and Y-compound.

Although Mg, like Li, Ca, Y, and some rare earth elements, has been long recognized to be a main stabilizing element for α-SiAlON as mentioned before, until now there were few papers about Mg–α SiAlON except for that formed by Mg–Ca dual cations.[26–29] A single phase Mg–α SiAlON ceramic is difficult to synthesize because its formability is much lower than that of the other ions.[30,31]

Recently, we used halloysite ($Al_2Si_2O_5(OH)_4$) and talc (($Mg_3(Si_2O_5)_2(OH)_2$) instead of higher purity powders as the starting components to fabricate Mg–α SiAlON powders.[32] Talc is a hydrous magnesium silicate with a high ratio of MgO/SiO_2, and is abundant and inexpensive. The Mg cation in talc mineral was finally incorporated into the SiAlON structure, and Mg–α SiAlON was formed during the CRN process.

Because two minimal materials were used, the chemical reactions during the heating process became much more complex, and the presence of phases, particle size and morphology of the synthesized powders turned out to be more sensitive to the composition of starting materials and other synthesis parameters. Since both Si and Mg are lost during the CRN process at high temperatures occur, it is difficult to calculate the values of *m* and *n* in the Mg–α SiAlON formula from the starting compositions.

The aim of this study is, therefore, to further investigate the optimal conditions for converting these raw materials to Mg–α SiAlON powders during the CRN process.

2. Experimental

New Zealand halloysite clay (Kyoritsu Yogyo Materials Co. Ltd.), talc (Kyoritsu Yogyo Materials Co. Ltd.), and Carbon Black (650B, Mitsubishi Chemical Co.) with a particle size of 0.018 μm and BET surface areas of below 200 m^2/g, were used as the starting materials. Table 1 lists the characteristics of the talc and the halloysite clay. In order to accelerate the CRN reactions and increase the conversion of α-SiAlON in the final products, we also added a 3 wt.% α-Si_3N_4[32] powder (E10, UBE Co.) as seed in all cases. The molecular ratios of talc to halloysite selected were 1.0:1.0, 1.5:10, 2.0:1.0 and 2.5:1.0. The powder mixtures were milled in ethanol using sintered silicon nitride balls for 24 h. After drying, the batch mixture was ground and passed though a sieve with 300 μm holes. The well-mixed granules were then heated in a graphite crucible placed horizontally in an alumina tube furnace and fired at 1450–1500 °C for 2–16 h. A stream of highly pure nitrogen gas (99.9995% N_2, $O_2 < 0.5$ ppm, $CO < 1.0$ ppm, $CO_2 < 1.0$ ppm, $CH_4 < 1.0$ ppm) flowed through the tube at a rate of 700 ml/min at normal pressure. The

Table 1
Chemical analysis of the talc and halloysite clay

Component	Talc (wt.%)	Halloysite (wt.%)
MgO	32.02	<0.04
Al_2O_3	0.01	34.41
SiO_2	60.63	51.89
Fe_2O_3	–	0.3
TiO_2	0.01	0.09
CaO	0.97	<0.01
K_2O	0.01	0.05
Na_2O	0.01	<0.09
P_2O_5	–	0.11
C	–	0.3
Ig. loss	6.18	12.34
Total	99.86	99.63

Table 2
Phases present of the products synthesized by the CRN process from different starting composition

C/Σ O (mol)	Talc (wt.%)	Halloysite (wt.%)	C (wt.%)	The final phases
0.80	54.0	24.6	21.4	α′(s), β′(w), $MgAl_2O_4$(w), 15R(w),SiC(w)
0.96	51.7	23.5	24.8	α′(s), β′(w), 15R(w), SiC(w)
1.0	50.1	22.8	27.1	α′(s), β′(w), 15R(w), SiC(w)
1.5	45.4	20.7	33.9	α′(m), SiC(m), β′(w), 15R(w)
2.0	40.8	18.6	40.6	SiC(m), α′(m), β′(w), 15R(w)

s, strong; m, medium; w, weak.

heating rates were chosen at 11.5 °C/min, and the natural cooling rate was approximately 20 °C/min. The residual carbon was removed by burning the resultant powders at 700 °C for 2 h in air and its content was measured from the weight loss after firing. The crystalline phases of products were analyzed by X-ray diffraction (XRD, Rigaku, RAD-2R), and the α-SiAlON and β-SiAlON contents were estimated from peak heights of the α-(102), α-(210), β-(101) and β-(210) reflections according to the following formula proposed by Gazzar et al.[33]

$$\alpha(\%) = I_{102(\alpha)}+I_{210(\alpha)}/\ I_{102(\alpha)}+I_{210(\alpha)}+I_{101(\beta)}+I_{210(\beta)} \qquad (1)$$

This equation applies for pure α- and β-Si_3N_4 and is also assumed to be valid for SiAlON materials because the structures are very similar.

The morphology of samples was characterized by scanning electron microscopy (SEM, Jeol JSM-5200).

3. Results and discussion

3.1. *Effect of molecular ratios of carbon*

Carbon is a main reduction factor during the CRN process. The amount and the surface area of carbon influence the reaction mechanism and reaction time, and play determining roles in the phases and morphology of particles of the final products. The carbon molecular ratio of 1.0 is supposed as a stoichiometric amount of carbon at which the starting materials could be completely converted. The composition with carbon molecules of 0.8, 0.96, 1.0, 1.5 and 2.0 at a talc to halloysite ratio of 1.5, and the phases of synthesized powders at 1480 °C for 4 h are summarized in Table 2. The SEM morphologies of particles are shown in Fig. 1 for various carbon contents.

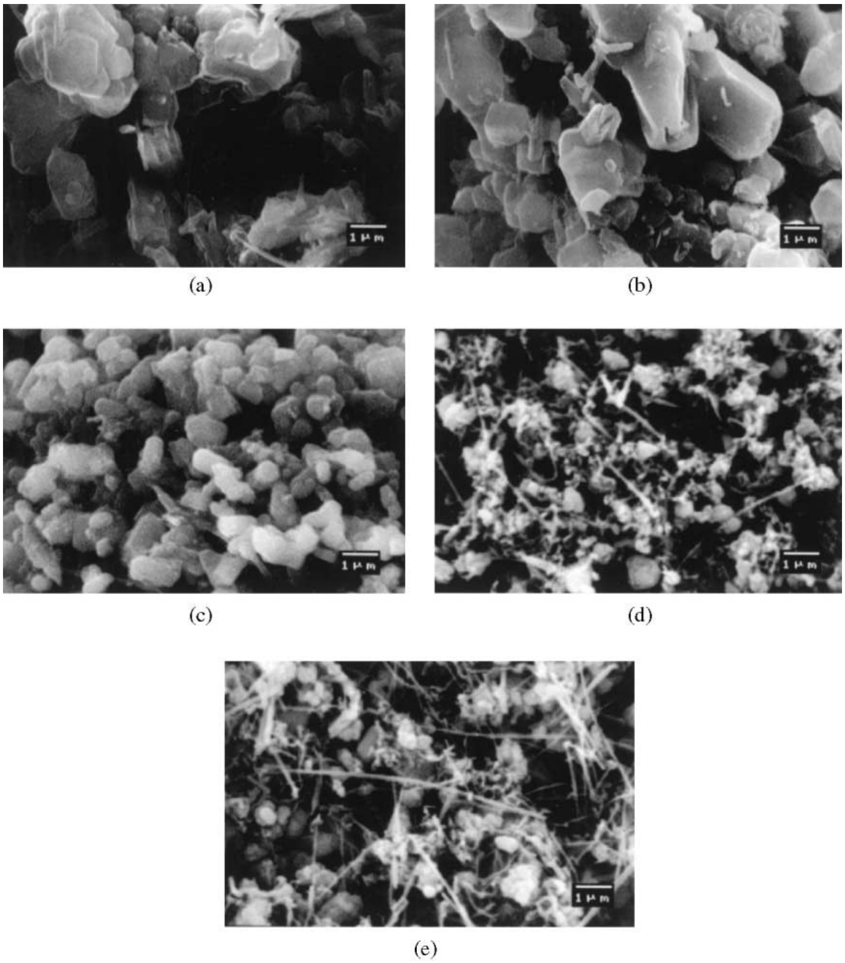
As shown in Table 2, the carbon content greatly influenced the reaction rate. At a stoichiometric ratio of 0.8, the full conversion of the CRN process was not possible. Spinel ($MgAl_2O_4$) was detected instead of Al_2O_3 or X-phase, and this is usually found due to a shortage of carbon.[34] It was suggested that reduced Mg reacted with Al_2O_3 by diffusion to form spinel. This coincided well with the results by Kokmeijer et al.[18] Sugahara[12] also reported that $MgAl_2O_4$ existed when starting materials were composed of Mg-compounds. When the stoichiometric ratio exceeded 0.96, full conversion by the CRN process may occur. An optimal carbon content was a molecular ratio of 1.0. The products consisted mainly of α-SiAlON and β-SiAlON phases, small amounts of 15R (15R-AlN polytypoid) and β-SiC were also identified.

Since the carbon particles (0.018 μm) are much smaller than the talc particles (5 μm) and halloysite particles (2 μm), the talc and halloysite particles should have been covered by carbon particles after 24 h of ball-mixing. The higher the carbon content, the thicker the layer it formed, and it became difficult for nitrogen gas to penetrate such a thick carbon layer to the inner parts of the talc and halloysite particles to promote the reaction at higher carbon ratios. This hindered the conversion of Mg–α SiAlON. At the some time, the excess of carbon content enhanced the partial pressure of CO which favored the formation of SiC whiskers according to reaction Eq. (2).[35] This could account for SiC whisker formation, in which a solid-vapor reaction could not produce SiAlON phases. Thus, a poor yield in synthesized powders was found in those samples with higher carbon ratios of 1.5 and 2.0. When the reaction time was extended to 16 h, SiC decreased and the α-SiAlON content increased to 88 wt.%. As shown in Fig. 2, the particles had irregular grain shapes and a wide size distribution. Therefore, we concluded that higher molecular carbon ratios decrease the reaction rate of the Mg–α SiAlON formation.

$$SiO(g) + 2C(s) \rightarrow SiC(whisker) + CO(g) \qquad (2)$$

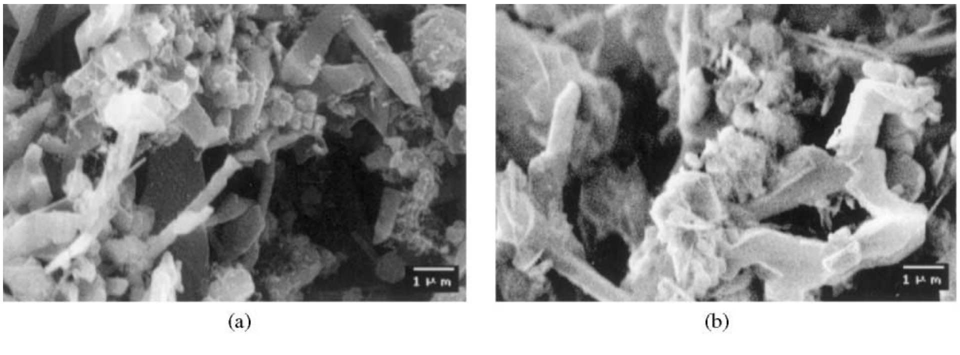
3.2. *Effect of the molecular ratios of talc to halloysite*

The previous investigation revealed that synthesized Mg–α SiAlON had a formula of $Mg_{0.4}Si_{10.6}Al_{1.4}O_{0.6}N_{15.4}$ from a starting composition of talc to halloysite molecular



(a) (b) (c) (d) (e)

Fig. 1. SEM micrographs of the products obtained at carbon molecular ratios of (a) 0.8, (b) 0.96, (c) 1.0, (d) 1.5 and (e) 2.0; 1480 °C for 4 h in N_2 flow; molecular ratio of talc to halloysite of 1.5.



(a) (b)

Fig. 2. SEM micrographs of the products obtained at carbon with molecular ratios of (a) 1.5 and (b) 2.0; 1480 °C for 16 h in N_2 flow; molecular ratio of talc to halloysite of 1.5.

ratio of 1.5; i.e. the x value is 0.4 and the ratio of Mg to Si is 0.04, which is much lower than that of the staring materials (Mg/Si = 0.56). We suggest that some of the Mg vaporizes during the reaction in a flowing N_2 atmosphere and becomes Mg-containing glass.[32] In other starting compositions chosen for this research as molecular ratios of talc to halloysite, 1.0 (Mg/Si = 0.50), 2.0 (Mg/Si = 0.6) and 2.5 (Mg/Si = 0.63), Mg contents all greatly exceeded the necessary value in the final synthesized samples. The chemical compositions of starting materials using talc and halloysite are listed in Table 3.

The CRN process involves a complex sequence of reactions, and the appearance and disappearance of the various phases during reaction with carbon in a nitrogen atmosphere. The two minerals decomposed at an early stage of the process, and then yielded β-SiC and

Table 3
Compositional ratios of MgO, Al_2O_3 and SiO_2 calculated from starting materials of talc and halloysite

Talc/halloysite (mol)	MgO	Al_2O_3 (mol)	SiO_2	MgO	Al_2O_3 (wt.%)	SiO_2
1.0	3.0	1	6	20.6	17.5	61.9
1.5	4.5	1	8	23.6	13.4	63.0
2.0	6.0	1	10	25.5	10.8	63.7
2.5	7.5	1	12	26.7	9.1	64.2

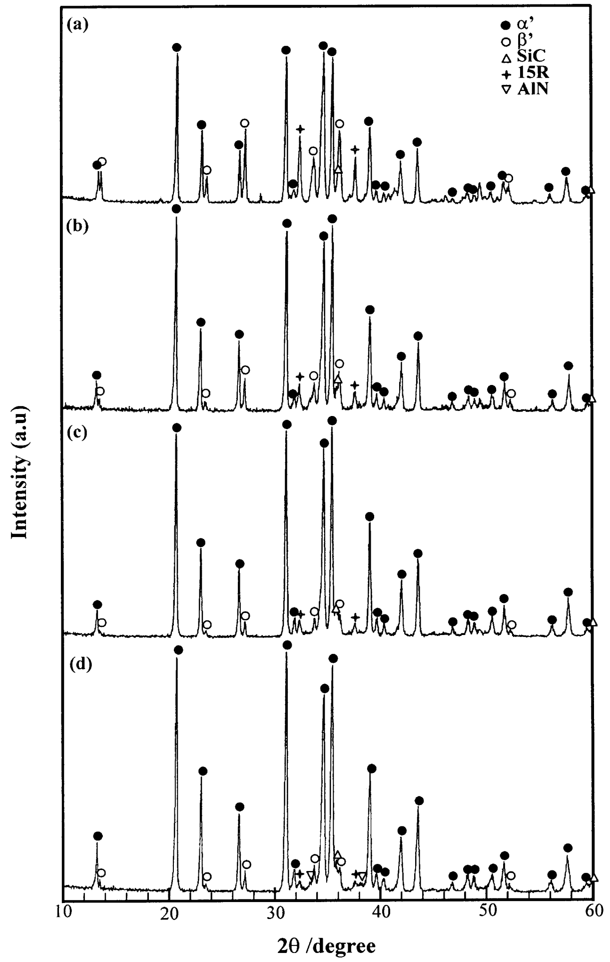


Fig. 3. XRD patterns of the synthesized products fired at 1480 °C for 4 h in N_2 flow; molecular ratios of talc to halloysite of (a) 1.0, (b) 1.5, (c) 2.0 and (d) 2.5.

$MgSiN_2$ as intermediate phases through a sequence of reactions and were finally transformed into Mg–α SiAlON. The XRD patterns of the synthesized powders from talc and halloysite at different molecular ratios fired at 1480 °C for 4 h are given in Fig. 3.

The reaction products were α-SiAlON, β-SiAlON, β-SiC, and 15R as main phases and lesser amount of AlN phase. Precise XRD measurement revealed that the α(210) peak of synthesized powders moved toward the lower 2*θ* angle, which means that the cells of the α-SiAlON phase became larger. This suggests that the Mg^{+2} cation was introduced into α lattices and formation of Mg–α SiAlON was confirmed. This was also verified by EDS analysis.[32]

The yield of α-SiAlON phase was less at a talc to halloysite ratio of 1.0. The Mg content was considered insufficient to form Mg–α SiAlON since some Mg in talc had evaporated due to decomposition of the intermediation $MgSiN_2$ during the reaction at higher temperatures in a flowing N_2 atmosphere. When the ratio was over 1.5, synthesized Mg–α SiAlON could be enhanced to 90 wt.%, whereas 15R phase decreased. In a composition with a ratio of 2.5, the Mg–α SiAlON content did not increase further owing to the volatilization at high temperatures.

The presence of 15R was explained by an excess of carbon.[5] Higgins et al.[36] stated that impurities such as calcium and magnesium resulted in over nitriding and the formation of 15R on prolonged heating. Another explanation is β-SiAlON might decompose in the presence of AlN, leading to 15R formation.[15,37] The experimental details reported by Yoshimatsu et al.[38] showed that 15R appeared in the range $0.8 \leqslant C_a \leqslant 1.0$ (C_a: the ratio of carbon according to stoichiometric reaction). In our research, 15R appeared at lower temperatures prior to β-SiAlON formation and tended to decrease with the increase of the talc to halloysite molecular ratio. Since 15R phase is near the AlN corner of the quaternary of the Si–Al–O–N phase diagram,[39] some 15R appeared with starting mineral containing more alumina. A similar observation was reported by Mazzoni et al.[40]

3.3. Effect of firing temperature and holding time on the reaction

The CRN reaction of clay commonly takes place at temperatures of 1400–1450 °C in flowing nitrogen gas and requires a few hours to complete the whole process.[25] The reaction products are very sensitive to firing temperature and holding time which are the two main factors determining thermodynamic activity.

The two minerals thermally break at an early stage before the CRN reaction, resulting in $MgSiO_3$, SiO_2 and $Al_6Si_2O_{13}$ (mullite). In this study, we investigated temperature effects on synthesis at five stages, ranging from 1350 to 1500 °C. Fig. 4 shows the XRD profiles of the reaction products with a talc to halloysite molecular ratio of 1.5. The products were detected after 4 h holding at 1350, 1400, 1450, 1480 and 1500 °C. The crystalline phases observed were α-SiAlON, β-SiAlON, β-SiC, 15R and a small amount of AlN. It seems that elevating temperature to 1450 °C can accelerate the CRN reactions and improve the formation of α-SiAlON. The AlN value also increased with an increase of reaction temperature, whereas the opposite effect was observed for β-SiC and 15R. A higher content of α-SiAlON was present for 4 h holding between 1480 and 1500 °C. The amount of β-SiAlON also increased slowly in this range of temperature. The samples were expected to contain a liquid phase. This liquid phase became saturated after absorbing the nitrogen component, causing the precipitation of β-SiAlON, which might be transformed from α-SiAlON at temperatures between 1400 and 1550 °C. In Fig. 5 the data based on XRD intensities of the product using the same mixture fired at 1480 °C for 0–6 h are semi-schematically plotted.

As shown in Fig. 5 the intermediate phase of Si_2N_2O, β-SiC and $MgSiN_2$ played certain roles in the whole CRN process. The Si_2N_2O decreased with holding time and disappeared completely after 15 min. The formation of Si_2N_2O intermediate phase might be described by reaction (3):

$$2SiO_2+3C+N_2 \rightarrow Si_2N_2O+3CO \quad (3)$$

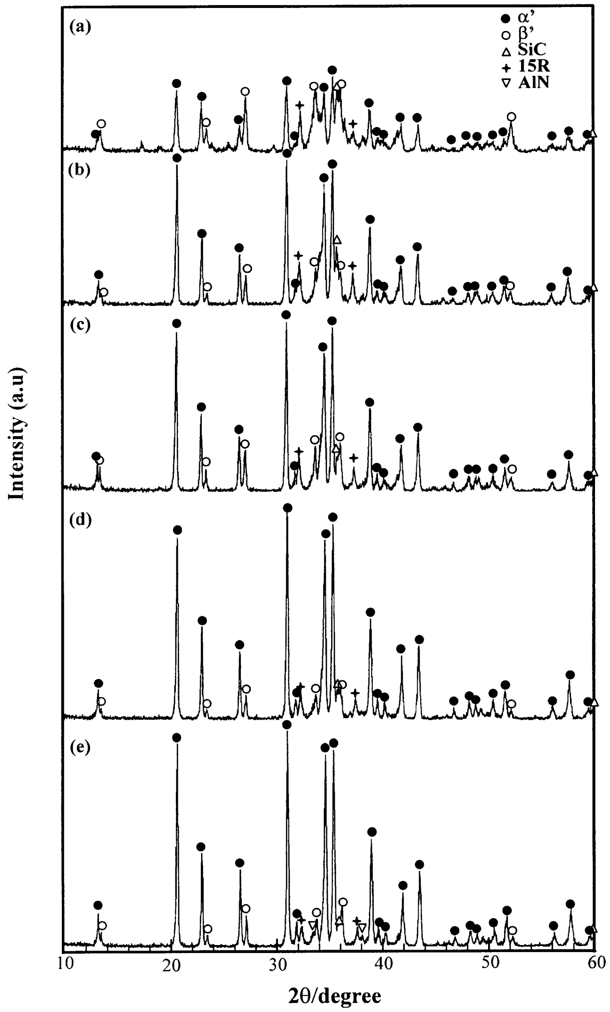


Fig. 4. XRD patterns of the synthesized products fired at (a) 1350, (b) 1400, (c) 1450, (d) 1480 and (e) 1500 °C for 4 h in N_2 flow; molecular ratio of talc to halloysite of 1.5.

A similar result was reported in which Si_2N_2O phase did not form in the absence of liquid and the amount of Si_2N_2O gradually decreases above a certain temperature.[41]

Si_2N_2O subsequently reacted with C to form β-SiC in a later step, which was facilitated by oxygen pressure over 1460 °C[41] in the Si_2N_2O+C mixture. The amount of oxides decreased with the extension of reaction time.

Meanwhile, most of the SiC particles possibly formed directly according to reaction (4):

$$SiO_2+2C \rightarrow SiC+2CO \quad (4)$$

α-Si_3N_4 formed by CRN is observed at 1500–1550 °C, and the formation of SiC is promoted at higher temperatures.[42] The addition of α-Si_3N_4 as a seed for nitridation converted the starting silica into α-Si_3N_4 at lower temperatures.[43] However, the presence of impurity elements such as iron[5,41] altered the phase equilibria by changing the thermodynamic activity, resulting in the formation of liquid phases at lower temperatures and the production of SiC at 1350–1500 °C. In our study using minerals as starting materials, β-SiC usually appeared at the beginning of the CRN process, a little earlier than α-SiAlON phase formed. It increased continuously at 1480 °C up to a maximum after about 30 min, then decreased rapidly. In contrast, α-SiAlON phases increased at the same time.

$MgSiN_2$, which is somewhat like β-SiC, continued to form after a soaking time of 0–15 min at 1480 °C, and then decreased. Finally, it vanished after 2 h and might also be an important intermediate compound to produce SiAlON in this system. $MgSiN_2$ decomposes into Si_3N_4, Mg (vapor) and N_2 (vapor) according to reaction (5).[32,44]

$$MgSiN_2 \rightarrow Si_3N_4+Mg+N_2 \quad (5)$$

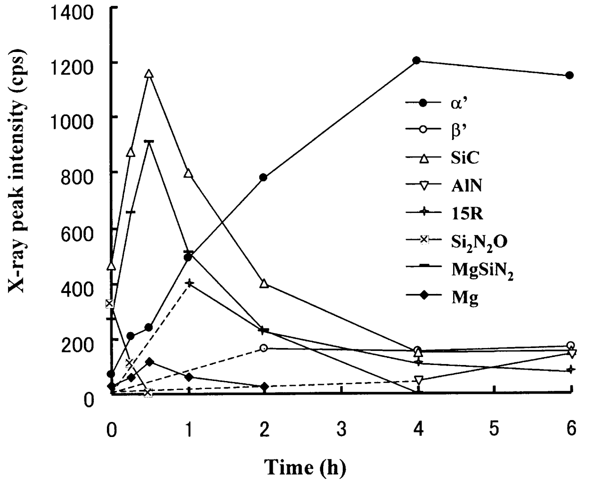


Fig. 5. Phases present of the products fired at 1480 °C for different times in N_2 flow; molecular ratio of talc to halloysite of 1.5.

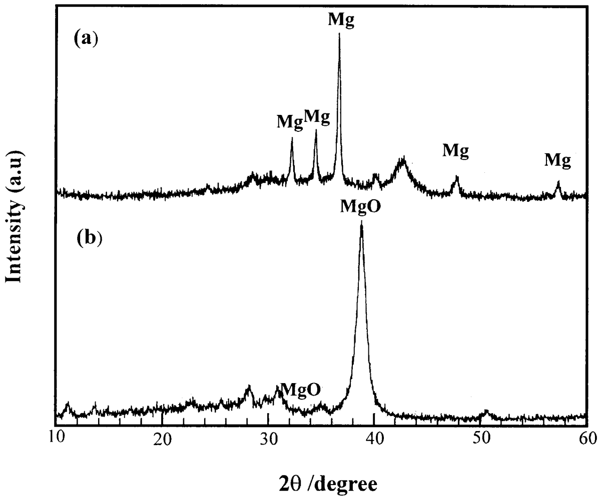


Fig. 6. XRD patterns of deposited matters nearby the gas outlet; 1480 °C, 4 h, N_2 flow; molecular ratio of talc to halloysite of 1.5; (a) detected matters, (b) after annealing the deposited matters of 500 °C for 2 h.

No traces of α-Si_3N_4 or β-Si_3N_4 were detected in this system. They might have been transformed into SiAlON phases (α', β') in which Mg, and Al-compound exist. As shown in Fig. 6, the wool-like deposited material was X-ray amorphous on the inner wall of the alumina tube nearby the gas outlet, but a few weak peaks were identified as Mg. After this volatile was annealed at 500 °C for 2 h in the atmosphere, MgO was detected. Mg loss was also confirmed to occur even at 1300[12] to 1427 °C[45] during the CRN process. According to the reaction (6) by Sugahara et al. and reaction (7) by Mazzoni et al.,

$$MgO + C \rightarrow Mg + CO \qquad (6)$$

$$MgSiO_3 + SiC \rightarrow Mg(g) + 2SiO(g) + CO \qquad (7)$$

α-SiAlON increased up to its maximum content. β-SiAlON was detected after α-SiAlON appeared, and increased with soaking time. As shown in Fig. 5, in the early stage at 1480 °C, the β-SiAlON peak was very weak. Identification of β-SiAlON by XRD was rather difficult because its d(201) diffraction line overlapped with the one peak of β-SiC at $d = 0.251$ nm.

In our research, 15R appeared at lower temperatures prior to β-SiAlON and decreased gradually.

Lee et al.[5] also reported that reaction (8) could take place when the temperature exceeded 1450 °C using kaolinite as starting material:

$$Al_2O_3{\cdot}2SiO_2 + 9C + N_2 \rightarrow 2SiC + 2AlN + 7CO \qquad (8)$$

Or according to reaction (9):

$$Al_2O_3 + 3C + N_2 \rightarrow 2AlN + 3CO \qquad (9)$$

However, we did not observe any trace of Al_2O_3 during the whole process. AlN was probably formed by previous SiO evaporation in the lack of silica.[33] Some 15R phase may transform to AlN with increasing temperatures and soaking time. Therefore, AlN was only observed at higher firing temperatures and longer holding times. This result is similar to the investigation of Mackenzie et al.[6]

4. Conclusions

The synthesis of Mg–α SiAlON powders by CRN using talc and halloysite is greatly affected by starting material composition and reaction parameters. From the above discussion, we can conclude that the appropriate ratio of talc to halloysite is 1.5 and 2.0; where an extra amount of magnesium is necessary to compensate for the evaporation of Mg and to reduce β-SiAlON, AlN and 15R phases and a glass phase in the final compositions. The formed $MgSiN_2$ disappeared at longer time. The key roles of $MgSiN_2$ and β-SiC intermediation were also confirmed. The CRN reactions occurred mainly over 1450 °C. Higher carbon content prolonged the SiAlON phase's conversion; the particles had an irregular grain shape and a wide distribution. An optimal carbon content at a molecular ratio of 1.0 could yield a larger content of Mg–α′ SiAlON formation with fine particles. The formation of β-SiAlON and AlN was increased by increasing the reaction temperature and holding time, whereas the α-SiAlON, β-SiC and 15R phases decreased. The optimum conditions for the synthesis of Mg-α SiAlON are 1480 °C for 4 h at talc to halloysite ratio of 1.5 and 2.0. The highest synthesized amount of Mg-α SiAlON was 90 wt.%, and the amounts of β-SiAlON, β-SiC, 15R and AlN were much smaller.

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