Synthesis of Magnesium Silicon Nitride by the Nitridation of Powders in the Magnesium–Silicon System

Hiroshi UCHIDA, Kiyoshi ITATANI, Mamoru AIZAWA, F. Scott HOWELL and Akira KISHIOKA

Department of Chemistry, Faculty of Science and Engineering, Sophia University, 7-1, Kioi-cho, Chiyoda-ku, Tokyo 102

マグネシウム–ケイ素系粉体の窒化による窒化ケイ素マグネシウムの合成

内田 寛・板谷清司・相澤 守・F. Scott Howell・岸岡 昭

上智大学理工学部化学科, 102 東京都千代田区紀尾井町 7-1

Magnesium silicon nitride (MgSiN_2) was synthesized by the nitridation of powders in the magnesium (Mg)–silicon (Si) system. Although an Mg and Si powder with an Mg/Si ratio of 1.0 (=stoichiometric ratio of MgSiN_2) was heated up to 1400°C in nitrogen (N_2), a single phase of MgSiN_2 could not be obtained, due to evaporation of Mg during heating. On the basis of the information that Mg_2Si formed as an intermediate phase during the synthesis of MgSiN_2 , an Mg_2Si powder was heated at a temperature between 600 and 1600°C for 1h in N_2 . Although MgSiN_2 and other phases such as Si and amorphous Mg_3N_2 were present in the temperature range of 900°C to 1300°C , the powder heated at 1400°C consisted of only MgSiN_2 phase. The chemical composition of the powder heated at 1400°C for 1h was almost stoichiometric; the content of the oxygen was as low as $0.76\text{ mol}\%$. The powder consisted of agglomerates with sizes of $\sim 5\text{ }\mu\text{m}$; these agglomerates were furthermore composed of primary particles with an average size of $0.54\text{ }\mu\text{m}$. The oxidation resistant temperature of MgSiN_2 was estimated to be $\sim 830^\circ\text{C}$. The MgSiN_2 powder had good resistivity to hydration.

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1. Introduction

Magnesium silicon nitride (MgSiN_2) has orthorhombic crystal structure similar to aluminum nitride (AlN) with a high theoretical thermal conductivity of $319\text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$.¹⁾ Thus MgSiN_2 is expected to be available as electronic substrates/packages, if the thermal conductivity and mechanical strength of MgSiN_2 are comparable to those of AlN .²⁾ Groen et al.³⁾ and Hintzen et al.,⁴⁾ however, reported that the thermal conductivity of MgSiN_2 ceramics is as low as $15\text{--}17\text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, regardless of the mechanical properties being sufficient for the practical uses. The poor thermal conductivity of MgSiN_2 ceramics is ascribed to the presence of oxygen in the crystal lattice of MgSiN_2 .^{3,4)}

Some further research is, therefore, needed to examine the conditions for the synthesis of MgSiN_2 with low oxygen content. The MgSiN_2 has previously been synthesized by the reaction of magnesium nitride (Mg_3N_2) with silicon nitride (Si_3N_4)^{3)–5)} and by the nitridation of magnesium silicide (Mg_2Si).⁵⁾ Nevertheless, no detailed information on the synthesis conditions of MgSiN_2 to obtain a pure compound has been available yet.

The present authors previously examined the synthesis conditions of Mg_3N_2 through the low-pressure chemical vapor deposition, i.e., the nitridation of the vaporized Mg.⁶⁾ Based on this information, the authors furthermore examined (i) the synthesis conditions of MgSiN_2 by the nitridation of the powders in the Mg–Si system and (ii) some properties of the resulting powder.

2. Experimental procedure

2.1 Starting materials and synthesis of MgSiN_2

Two kinds of the starting materials, i.e., (i) Mg powder (Wako Pure Chem.; purity, 99.9%) and (ii) Si powder (Nilako; purity, 99.999%), were used in this experiment; the particle sizes of Mg powder determined using a scanning electron microscope (SEM) were in the range of 300 to $500\text{ }\mu\text{m}$, whereas those of Si powder were in the range of 10 to $100\text{ }\mu\text{m}$. After mixing these powders with the desired

Mg/Si ratios, about 0.3 g of the powder mixture was pressed at 50 MPa to form the compact with a diameter of 10 mm and a thickness of $\sim 2\text{ mm}$. The compact was heated up to the desired temperature at the rate of $10^\circ\text{C}\cdot\text{min}^{-1}$ in a nitrogen (N_2) atmosphere and was kept for 1h. The heat-treated compact was pulverized using an agate mortar and pestle.

2.2 Evaluation of the powder properties

The crystalline phases of the powder were identified using an X-ray powder diffractometer (XRD; Model RAD-IIA, Rigaku, Tokyo; 40 kV , 25 mA) with Ni-filtered Cu $K\alpha$ radiation. The Mg and Si contents were determined using an energy-dispersive X-ray microanalyser (EDX; Model EMAX-5770, Horiba, Kyoto); the oxygen and nitrogen contents were determined using an N/O determinator (Model TC-136, Leco, St. Joseph, MI, USA). The binding energy of Si was examined using an X-ray photoelectron spectroscopy (XPS; Model SSX-100, SSI, Mountain View, CA, USA). The differential thermal analysis (DTA) and thermogravimetry (TG) were conducted using a DTA-TG apparatus (Model 8076D1, Rigaku, Tokyo) at the heating rate of $10^\circ\text{C}\cdot\text{min}^{-1}$ in the N_2 atmosphere.

The particle shapes of the powders were observed using a high-resolution transmission electron microscope (HR-TEM; Model H-9000, Hitachi; accelerating voltage, 300 kV) and using scanning electron microscope (SEM, Model S-4500, Hitachi; accelerating voltage, 20 kV). The specific surface area was measured by a BET technique, using N_2 gas as an adsorption gas.

Assuming the primary particles to be spherical, we calculated the average primary particle size (G_{BET}) as follows:

$$G_{\text{BET}} = \frac{6}{\rho s} \quad (1)$$

where ρ is the true density and s is the specific surface area of the powder. The true density of the powder was measured picnometrically at 25.0°C . The crystallite size

(G_{XRD}) of MgSiN_2 was calculated by X-ray diffractometry on the basis of Scherrer's formula:

$$G_{\text{XRD}} = \frac{K\lambda}{\beta \cos \theta} \quad (2)$$

where K is a constant ($=0.9$) and λ is the wavelength ($=0.15405$ nm) of $\text{Cu K}\alpha_1$, β is a half-width of a characteristic X-ray reflection (322) and θ is a Bragg angle.

The water-proof test of the powder was conducted as follows: 1.0 g of the powder was immersed in 200 cm³ of boiling water for 24h; the water-immersed powder was dried in an oven at 110°C. The changes in sample weight were calculated on the basis of the precise measurements of the sample weights before and after immersing the powder in the boiling water.

3. Results and discussion

3.1 Synthesis of MgSiN_2

3.1.1 Reaction among Mg, Si and N_2

The reaction among Mg, Si and N_2 was examined to search for the optimum conditions for the synthesis of MgSiN_2 . The mixing ratio of Mg to Si was adjusted to be 1.0 (=stoichiometric ratio). Details of the synthesis of MgSiN_2 by this technique are described below.

First, the reaction process among Mg, Si and N_2 during the heating was examined using DTA-TG and XRD. Figure 1 shows the DTA-TG curves of the Mg and Si powders with the Mg/Si ratio of 1.0 in N_2 atmosphere. The DTA-TG curve contained the exothermic effects at 550°C and 770°C. The TG curve showed that the sample weight increased from 550°C up to 1000°C and became almost constant in the range of 1000°C to 1100°C; the sample weight decreased slightly with a further increase in temperature up to 1400°C.

On the basis of the above DTA-TG data, the crystalline phases of the heat-treated powders were examined using the XRD. Typical X-ray diffraction patterns of the heat-treated powders are shown in Fig. 2, together with the pattern of the as-mixed powder. The crystalline phases of the powder at room temperature were Mg and Si, whereas those of the powder heated at 700°C were Si, Mg_2Si ,⁷⁾ Mg_3N_2 ⁸⁾ and MgO (trace). Although MgSiN_2 ⁹⁾ was detected above 900°C, Si and MgO were still present in the powder. The powder heated at 1400°C contained not only MgSiN_2 but also $\alpha\text{-Si}_3\text{N}_4$ and MgO; the X-ray intensities of MgSiN_2 were higher than those of the powder heated at 900°C.

Since Si, Mg, Mg_2Si and Mg_3N_2 were detected from the powder heated at 700°C, the first exothermic effect at 550°C

Fig. 1. DTA-TG curve of Mg and Si powder with the Mg/Si ratio of 1.0 in N_2 atmosphere at the heating rate of $10^\circ\text{C}\cdot\text{min}^{-1}$.

Fig. 2. Typical X-ray diffraction patterns of the Mg and Si powder with the Mg/Si ratio of 1.0 in N_2 atmosphere heated up to the desired temperature at the rate of $10^\circ\text{C}\cdot\text{min}^{-1}$.

● Mg, ■ Si, ○ Mg_3N_2 , ⊙ Mg_2Si ,
○ MgO, ○ MgSiN_2 , □ $\alpha\text{-Si}_3\text{N}_4$.

is attributed to (i) the solid-state reaction between Mg and Si (Eq. (3)) and (ii) the solid-gas reaction between Mg and N_2 (Eq. (4))

Although MgO was present in the powders heated at and above 700°C, the formation of such oxide may chiefly be derived from the oxygen and/or water (H_2O) vapor adsorbed on the surfaces of starting Mg particles, because Mg particles are more sensitive to the oxygen and H_2O vapor than Si particles.¹⁰⁾

Since MgSiN_2 was detected at 900°C, the second exothermic effect at 770°C is attributed to the formation of MgSiN_2 as follows:

Although no distinct weight increase occurred in the range of 800°C to 1100°C, this phenomenon may be explained in terms of the balance of the weight increase and decrease, i.e., (i) the weight increase due to the participation of N_2 in the formation of MgSiN_2 and (ii) the weight decrease due to the evaporation of Mg (melting point: 650°C).¹⁰⁾ The slight decrease in sample weight with temperature from 1100°C to 1400°C is attributed to the evaporation and decomposition of amorphous Mg_3N_2 and/or the evaporation of the small amount of the unreacted Mg above the boiling point (1107°C).¹⁰⁾ Moreover, although $\alpha\text{-Si}_3\text{N}_4$ was detected at 1400°C, this phenomenon is not ascribed to the decomposition of MgSiN_2 but to the nitridation of the residual Si, because the X-ray intensities of MgSiN_2 were enhanced with temperature:

Since the evaporation of Mg during the heating makes the formation of the single MgSiN_2 phase difficult, we furthermore examined the nitridation behavior of Mg and Si powder with the Mg/Si ratio of 2.0; the Mg content was two times as high as the stoichiometric value. When the Mg and Si powder was heated up to 1400°C, the crystalline

phases were MgSiN_2 , $\alpha\text{-Si}_3\text{N}_4$ and MgO (trace). Regardless of an increase of Mg content in the starting powder, the single MgSiN_2 phase could not be obtained. Here the vapor pressure (P) of Mg in the range of 651°C to 1127°C can be expressed as follows:¹¹⁾

$$\log 760 P = -\frac{7610}{T} - 1.020 \log T + 11.61 \quad (7)$$

where T indicates the absolute temperature. The vapor pressures at 650°C (melting point) and 1107°C (boiling point) are found from calculation to be 2.9×10^2 Pa and 1.0×10^5 Pa, respectively; the vapor pressure at 1107°C is about 350 times as high as that at 650°C. Such rapid evaporation of Mg above the boiling point (1107°C) makes the synthesis of stoichiometric MgSiN_2 difficult.

As the present data indicate, we could not find the optimum conditions for the formation of MgSiN_2 through the reaction among Mg, Si and N_2 ; the reason may be that Mg are easily evaporated during heating.

3.1.2 Reaction of Mg_2Si with N_2

We found that Mg_2Si forms as an intermediate phase during the synthesis of MgSiN_2 . Based on this information, we furthermore examined the optimum conditions for the synthesis of MgSiN_2 by the nitridation of Mg_2Si . Here Mg_2Si was synthesized by the solid-state and solid-liquid reactions between Mg and Si; about 0.3 g of the powder mixture with Mg/Si ratio of 2.0 was pressed at 50 MPa and then heated at 700°C for 1h in Ar atmosphere.

After the pulverization of the heat-treated compact, the properties of the resulting powder (Mg_2Si) were recorded as follows: (i) the color of the powder was pale-gray, (ii) the chemical composition determined using the EDX was: Mg 66.50 mol%, Si 32.96 mol% and was almost in accord with the stoichiometric composition; (iii) the oxygen content determined using the N/O determinator was 0.64 mol%; (iv) the specific surface area and primary particle size were $6.8 \text{ m}^2 \cdot \text{g}^{-1}$ and $0.44 \mu\text{m}$, respectively. The preparation of the Mg_2Si powder with the stoichiometric composition may be achieved, partly because the vapor pressure of Mg calculated on the basis of Eq. (7) is still as low as 7.2×10^2 Pa at 700°C, and partly because the reaction products in the Mg-Si system, which are formed at the interfaces between Mg and Si particles during the heating up to 700°C, prevent Mg from vaporizing.

Figure 3 shows the DTA-TG curves of Mg_2Si powder in N_2 atmosphere. The DTA curve showed that the exothermic effects started at 800°C and 820°C, whereas the endothermic effect appeared at 1110°C. The TG curve showed that the sample weight increased remarkably with

Fig. 3. DTA-TG curve of the Mg_2Si powder in the N_2 atmosphere at the heating rate of $10^\circ\text{C} \cdot \text{min}^{-1}$.

temperature from 670°C up to 820°C; the sample weight was reduced in the range of 820°C to 1320°C and then remained unchanged on further heating up to 1400°C.

Mg_2Si powder was heated at a temperature between 600°C and 1600°C for 1h in N_2 atmosphere. The crystalline phases of the heat-treated powders were examined using XRD. Typical X-ray diffraction patterns are shown in Fig. 4; each sample powder was heated at the desired temperature for 1h. The crystalline phases at 750°C were Mg_2Si , Si and Mg_3N_2 . When the temperature was raised up to 900°C, the crystalline phases were Mg_2Si , MgSiN_2 and Si. Only MgSiN_2 was found at 1400°C. On further heating up to 1600°C, however, $\alpha\text{-Si}_3\text{N}_4$ and $\beta\text{-Si}_3\text{N}_4$ were present.

The phase changes during the nitridation of Mg_2Si were checked on the basis of the characteristic reflection intensities of the reaction products. Results are shown in Fig. 5. The starting Mg_2Si decreased with temperature and disappeared at 1000°C. Si and Mg_3N_2 appeared at 600°C. Although Mg_3N_2 disappeared at 800°C, Si was still present in the powder up to 1300°C. The MgSiN_2 formed above 800°C and increased with temperature up to 1400°C; however, it disappeared on further heating. Moreover, $\alpha\text{-Si}_3\text{N}_4$ and $\beta\text{-Si}_3\text{N}_4$ formed above 1400°C; they increased with a further increase in temperature.

Since Mg_3N_2 and Si were detected at 600°C, Mg_2Si may react with N_2 to form Mg_3N_2 and Si:

As described before, MgO forms when Mg and Si react with N_2 at and above 700°C. Although no MgO forms when Mg_2Si reacts with N_2 , this phenomenon may be ascribed to the low oxygen content (0.64 mol%) in the Mg_2Si powder. The exothermic effects at 800–820°C, accompanying the weight increases, are attributed to the formation of MgSiN_2 :

As Eq. (9) indicates, the reaction among Mg_3N_2 (crystalline phase), Si and N_2 appears to produce MgSiN_2 and amor-

Fig. 4. Typical X-ray diffraction patterns of the Mg_2Si powders in N_2 atmosphere heated up to the desired temperature at the rate of $10^\circ\text{C} \cdot \text{min}^{-1}$.

Note that the sample was heated at each temperature for 1h.

○ Mg_2Si , ○ MgSiN_2 , ● Mg_3N_2 , ■ Si, ■ $\alpha\text{-Si}_3\text{N}_4$, ■ $\beta\text{-Si}_3\text{N}_4$.

Fig. 5. Phase changes due to the reaction between Mg_2Si and N_2 with temperature at the heating rate of $10^\circ\text{C}\cdot\text{min}^{-1}$. Note that the sample was heated at each temperature for 1h.

○ Mg_2Si ($2\theta=40.1^\circ$), ○ MgSiN_2 ($2\theta=36.0^\circ$),
● Mg_3N_2 ($2\theta=51.9^\circ$), ■ Si ($2\theta=28.4^\circ$),
■ $\alpha\text{-Si}_3\text{N}_4$ ($2\theta=28.4^\circ$), ■ $\beta\text{-Si}_3\text{N}_4$ ($2\theta=30.9^\circ$).

phous materials in the Mg–N system or nominal composition of Mg_3N_2 . Although we predicted that the porous particles might form after an exothermic effect at $800\text{--}820^\circ\text{C}$, SEM observation showed that the surfaces of the particles were smooth.

The appreciable weight loss at temperatures exceeding 820°C up to 1320°C , accompanying the endothermic effect, corresponds to the evaporation and decomposition of Mg_3N_2 (decomposition temperature: $\sim 1300^\circ\text{C}$):¹²⁾

Only MgSiN_2 is present in the sample heated at 1400°C . Since MgSiN_2 disappeared to form α - and β - Si_3N_4 above 1400°C , MgSiN_2 is decomposed to form Si_3N_4 , Mg (vapor) and N_2 (vapor) as follows:

Although α - and β - Si_3N_4 were detected from the powder, β - Si_3N_4 may be more stable than α - Si_3N_4 .¹³⁾

The above results indicate that the heat-resistant temperature of MgSiN_2 appears to be $\sim 1400^\circ\text{C}$ and that MgSiN_2 decomposes into α - Si_3N_4 and β - Si_3N_4 at temperatures exceeding 1400°C .

3.2 Evaluation of some properties of the resulting powder

Since the excellent crystalline MgSiN_2 was obtained by nitriding the Mg_2Si at 1400°C for 1h, some properties of this powder are examined in this section.

3.2.1 Crystallographic evaluation

Table 1 shows the typical chemical composition of this powder. The Mg and Si contents were almost in accord with the stoichiometric contents. The content of oxygen

Table 1. Quantitative Analysis of the Synthesized Compounds

	Mg	Si	N	O
	mol%	mol%	mol%	mol%
Present study (1400°C , 1 h)	25.20	25.55	48.49	0.76
Groen et al. ³⁾	25.49	24.63	45.12	4.76
Hintzen et al. ⁴⁾	— ^{a)}	— ^{a)}	44.28	5.93
Theoretical	25.00	25.00	50.00	—

a) Mg and Si contents were not listed in the paper.

was 0.76 mol%; it was lower than the content reported by Groen et al. (4.76 mol%)³⁾ and that by Hintzen et al. (5.93 mol%)⁴⁾. Moreover, the lattice parameters of MgSiN_2 determined by X-ray diffractometry were $a=0.5276\text{ nm}$, $b=0.6453\text{ nm}$ and $c=0.4984\text{ nm}$; the lattice volume was $169.68 \times 10^{-3}\text{ nm}^3$. These values are almost in accord with those reported previously,^{3,4,14)} which indicates that most of the oxygen may not be solid-soluted into the MgSiN_2 lattice. The theoretical density calculated on the basis of the lattice parameters and lattice volume is estimated to be $3.148\text{ g}\cdot\text{cm}^{-3}$.

We furthermore examined the binding energy of Si(2p) by XPS to check for the presence of oxygen; the data were compared with those reported by the previous researchers.^{15,16)} Typical results are shown in Fig. 6. The Si–Si binding energy appeared at 99.4 eV ; the Si–N binding energy appeared at 102.8 eV ; the Si–O binding energy appeared at 102.8 eV .

The detection of Si–O binding by XPS indicates that the silicon oxide (SiO_2) and/or silicon oxynitrides ($\text{Si}_2\text{N}_2\text{O}$) may be present on and near the surfaces of MgSiN_2 particles.³⁾ According to the EDX analysis, 0.76 mol% of oxygen are included in the powder. Taking the XPS and EDX results into account, most of the oxygen seem to be present on and near the surfaces of the particles.

3.2.2 Powder properties

The color of the MgSiN_2 powder was white. The specific surface area of this powder was $3.6\text{ m}^2\cdot\text{g}^{-1}$, while the true density was $3.102\text{ g}\cdot\text{cm}^{-3}$. The primary particle size calculated on the basis of the specific surface area was $0.54\text{ }\mu\text{m}$, whereas the crystallite size (322) of MgSiN_2 calculated on the basis of X-ray diffractometry was 42 nm ($0.042\text{ }\mu\text{m}$).

A typical TEM micrograph of this powder is shown in Fig. 7, together with the electron diffraction pattern. The powder was composed of the particles with the sizes of $\sim 0.5\text{ }\mu\text{m}$. The electron diffraction pattern had the spots, reflecting the orthorhombic structure of MgSiN_2 .

The color of the resulting powder is white as reported by Groen et al.³⁾ The true density ($3.102\text{ g}\cdot\text{cm}^{-3}$) is almost in accord with the theoretical density ($3.148\text{ g}\cdot\text{cm}^{-3}$). Moreover, the particle sizes ($\sim 0.5\text{ }\mu\text{m}$) observed by HR-TEM agree well with the primary particle size ($0.54\text{ }\mu\text{m}$) calculated from the specific surface area. These particles are, therefore, regarded as primary particles. The primary particles appear to be polycrystals, because the primary particle size ($0.54\text{ }\mu\text{m}$) is over ten times as large as the crystallite size ($0.042\text{ }\mu\text{m}$).

3.2.3 Resistivity to the oxidation and hydration

The application of MgSiN_2 to the engineering materials

Fig. 6. XPS Si(2p) spectrum of the MgSiN_2 powder obtained by nitriding the Mg_2Si at 1400°C for 1h.

Fig. 7. HR-TEM micrograph and electron diffraction pattern of the MgSiN_2 powder obtained by nitriding the Mg_2Si at 1400°C for 1h.

and/or high-thermal conducting materials requires an ability to resist oxidation and/or hydration. Thus the resistivity to the oxidation and hydration was examined in this section.

First, DTA was conducted to examine the resistivity to the oxidation of MgSiN_2 . Results are shown in Fig. 8. DTA curve showed that the exothermic effects started at 710°C and 1270°C . Although little changes in sample weight were observed below 830°C , the sample weight increased with a further increase in temperature.

On the basis of the above DTA-TG results, the crystalline phases of the heat-treated powders were examined using the XRD. Typical X-ray diffraction patterns of the heat-treated powders are shown in Fig. 9, together with the pattern at room temperature. Although only MgSiN_2 was present at room temperature, MgO and Mg_2SiO_4 (trace) formed at 1040°C , together with MgSiN_2 . When the temperature was raised up to 1210°C , Mg_2SiO_4 , MgO and MgSiN_2 (trace) were present. On further heating up to 1400°C , the crystalline phases were Mg_2SiO_4 and MgSiO_3 (protoenstatite).

Since MgSiN_2 , MgO and Mg_2SiO_4 were detected at 1040°C , the first exothermic effect which started at 710°C may be attributed to (i) the thermal decomposition of MgSiN_2 and (ii) the formation of MgSiO_3 (protoenstatite):

Regardless of the gradual exothermic effect, however, little weight changes occurred in the range of 710°C to 830°C , which suggests that the thin MgO and SiO_2 (amorphous) layers formed on the surfaces of the particles at the initial stage prevent from a further oxidation inside the particles.

Since Mg_2SiO_4 and MgSiO_3 (protoenstatite) were detected at 1400°C , the solid state reaction in the range of 1270°C to 1400°C may be expressed as follows:

Contrary to the prediction of no weight increases, according to Eq. (14), the sample weight increases in this temperature range. The exothermic effect which started at 1270°C appears to occur due to the rapid oxidation of the residual MgSiN_2 , initiating from the crack formation of the oxide films on the particles.

It is concluded that MgSiN_2 may be stable up to $\sim 830^\circ\text{C}$, because no appreciable weight changes were observed below this temperature.

Fig. 8. DTA-TG curves of MgSiN_2 powder in air at the heating rate of $10^\circ\text{C}\cdot\text{min}^{-1}$.

Fig. 9. Typical X-ray diffraction patterns of the MgSiN_2 powders heated up to the desired temperature in air at the heating rate of $10^\circ\text{C}\cdot\text{min}^{-1}$.

○ MgSiN_2 , ● MgO , ▲ Mg_2SiO_4 ,
△ MgSiO_3 (protoenstatite).

The water-proof test was conducted by immersing the powder in boiling water. The sample weight increased up to 21.06 mass% after the water-proof test. Although the XRD data were omitted in this paper, no other crystalline phases were observed after the water-proof test; however, the reflection intensities of MgSiN_2 were slightly lowered after the water-proof test.

An increase in sample weight indicates that the magnesium and silicon hydroxides may be formed on and near the surfaces of the particles. Regardless of the formation of such hydroxides, however, it is noteworthy that the crystal structure of MgSiN_2 is still maintained even after the "severe" water-proof test.

4. Conclusion

The synthesis conditions of magnesium silicon nitride (MgSiN_2) were examined by the nitridation of the powders in the Mg-Si system. The results obtained can be summarized as follows:

(1) The mixed Mg and Si powders with Mg/Si ratio of 1.0 (stoichiometric ratio of MgSiN_2) was heated up to 1400°C in nitrogen (N_2) atmosphere. The single MgSiN_2 phase could not be obtained by this technique, because some Mg were evaporated during heating.

(2) The Mg_2Si powder was heated at a temperature between 600°C and 1600°C for 1h in N_2 atmosphere. The single MgSiN_2 phase was obtained by nitriding the Mg_2Si at 1400°C for 1h. The chemical composition of the resulting powder was almost in accord with the stoichiometric composition; the content of the oxygen was as low as 0.76 mol%.

(3) The MgSiN_2 powder was composed of agglomerates with the sizes of $\sim 5\mu\text{m}$ in which the primary particles with the sizes of $\sim 0.5\mu\text{m}$ were adhered to one another. The oxidation-resistant temperature of the MgSiN_2 powder was estimated to be $\sim 830^\circ\text{C}$. The MgSiN_2 powder has good resistivity to hydration.

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