



EFFECT OF REACTION CONDITIONS ON THE SYNTHESIS OF ULTRAFINE AIN POWDER WITH GLUCOSE AS CARBON SOURCE

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Using aluminum hydroxide $(Al(OH)_3)$ and aluminum nitrate $(Al(NO_3)_3, 9H_2O)$ as aluminum sources and highly active glucose $(C_6H_{12}O_6)$ as a carbon source, we synthesized ultrafine AlN powder through carbothermal reduction-nitridation. Then, we explored the effects of aluminum sources, reaction temperature, holding time and other reaction conditions on the phase composition and micromorphology of the synthesized AlN powder. Result showed that AlN powder and different proportions of Al_2O_3 -AlN composite powder can be prepared by controlling reaction temperature or holding time. Under the conditions of this experiment, $Al(OH)_3$ more beneficial as an aluminum source for carbothermal reduction-nitridation than $Al(NO_3)_3, 9H_2O$. The optimum reaction conditions for the synthesis of single-phase AlN powder with $Al(OH)_3$ as aluminum source were holding time of 3 h and temperature of 1450°C. The powder sample synthesized under this reaction condition is mainly composed of flaky, short-rod, and irregularly shaped fine particles with sizes ranging from 50 nm to 100 nm.

INTRODUCTION

Owing to the development of microelectronic technologies, especially high-frequency and high-power integrated circuits, integrated circuit substrates have been increasingly used in several industries. Integrated circuit substrates must exhibit high thermal conductivity for the timely distribution of heat generated from substrate materials for high-power applications and prevent the burnout of the integrated block at high temperatures [1]. Aluminum oxide (Al₂O₃) exhibits excellent physical and chemical properties, can be produced from a wide source of raw materials, has a comparatively cheap price, and mature processing and manufacturing technology [2]. Thus, it is one of the most commonly used substrate materials in integrated circuits [3]. However, its thermal conductivity is relatively low and thus its application remains limited. Aluminum nitride (AlN) is a new type of advanced ceramic material that possesses a series of excellent properties, including high thermal conductivity (theoretical and actual values of 320 and 260 W/m·K, respectively, which are 10-15 times of that of alumina ceramic), reliable electrical insulation, low dielectric loss and dielectric constant, and thermal expansion coefficient near to that of silicon phase [4-5]. AlN has been widely applied in substrate, electronic film, integrated circuit substrate, microelectronic, and protective armor materials and electronic optical devices. Therefore, AlN has highly

broad application prospects in the electronics industry [6-7]. Furthermore, it is an ideal packaging material for a new generation of highly integrated semiconductor substrates and electronic devices [8-9].

AlN ceramics with excellent comprehensive performance can be produced by synthesizing AlN powder with high purity, small particles, high sintering activity, and stable performance. Present methods for AlN powder synthesis include carbothermal reduction nitridation (CRN) [10-12], aluminum powder direct nitriding [13], self-propagating high temperature synthesis [14], and chemical vapor deposition [15]. By contrast, CRN is a simple process with a wide range of raw materials and low production cost. Moreover, CRN yields high-purity AlN powder with excellent particle size and sintering performance. Therefore, CRN has become the most important industrial production method for AlN powder. In conventional CRN, the commonly used Al₂O₃ and carbon raw materials are difficult to homogeneously combine. Moreover, the Al₂O₃ raw material has low reactivity; therefore, CRN requires high nitriding temperature and long reaction time, thus increasing the particle size of the synthesized AlN powder [16-17]. In recent years, considerable effort has been expended to improve CRN by reducing nitriding temperature. Qi et al. prepared hexagonal bipyramids via CRN using alumina and carbon as raw materials and with Fe₂O₃, SiO₂, and Na₂O as mineralizers under alternate gas pressures; they found that the growth of AlN hexagonal bipyramids is dominated by the vapor-solid mechanism [18]. Wang et al. investigated the effects of various additives on conversion fraction, weight loss, phase compositions, and surface morphology of spherical AlN granules during carbothermal synthesis [19]. However, these methods above mentioned called for high requirement of starting materials and the morphology of AlN powder was strongly dependent on the morphology of the aluminium source types used. Several studies have shown that different aluminum sources, raw material ratio, and reaction temperatures are the key factors that affect the quality of AlN powder [20-21]. Although many researchers have used different starting materials and mixing methods, the homogeneous mixing and good bonding between aluminum and carbon are desirable to reduce the nitridation temperature and improve the properties of AlN powders. Unfortunately, reports on the effects of raw materials, reaction temperature, holding time, and other reaction conditions on the properties of AlN powders synthesized by carbothermal reduction nitridation are few. In our previous study [22], we investigated the effects of different aluminum sources and reaction temperature on the phase composition and micromorphology of AlN powder synthesized from different aluminum sources (aluminum hydroxide, aluminum nitrate, and α -aluminum oxide) and highly active nanocarbon black and discussed the synthesis reaction mechanism of AlN powder. Glucose is cheap and easy to obtain, and we can easily form fine carbon particles in the later carbonization and decomposition process. With a large specific surface area, glucose conducive to the synthesis of ultrafine powders. Therefore, based on previous studies, we select glucose as a carbon source to comparatively study the effects of aluminum sources, reaction temperature and holding time, and other reaction conditions on reaction weight loss rate, phase composition, and micromorphology of AlN composite powder.

EXPERIMENTAL

Processing

The process diagram for the synthesis of AlN ultrafine powder is shown in Figure 1. A theoretical weight loss rate of 63.64 % for a single glucose molecule was calculated during synthesis reaction at high temperatures by using the reaction formula of single-carbon and H₂O gaseous substances generated from the carbonization decomposition of glucose ($C_6H_{12}O_6 \cdot H_2O$). Thus, we were able to calculate the amount of carbon in the reaction process of carbothermal reduction nitridation on the basis of glucose levels. First, we used an electronic analytical balance to weigh Al(OH)₃ and glucose according to the required proportion. Then, the weighted materials were placed in an agate jar. A certain amount of absolute ethyl alcohol (solid-liquid ratio of 1:2) was added as a liquid medium. A zirconium oxide porcelain ball was used as a grinding body for 1 h of ball milling in a planetary fast ball mill. The slurry was dried for 24 h in an electric drying oven with forced convection of 100°C. Then, the dried powder material was placed into a tube atmosphere furnace after it was subjected to a screening treatment. The powder was vacuumed for 20 min under the protection of flowing N₂ (purity \geq 99.99 %) gas. The temperature was increased at a rate of 10°C·min⁻¹ to 500°C and held for 20 min. Carbothermal reduction nitridation reactions were induced by increasing the temperature to 1400°C, 1450°C, and 1500°C at a rate of 10°C·min⁻¹ and holding for 2, 3, and 4 h. At the end of the holding time, the temperature was reduced to approximately 250°C at a cooling rate of 10°C·min⁻¹. The furnace was cooled naturally and the carbon-containing AlN powder was obtained. Subsequently, the carbon-containing AlN powder was decarbonized in a box-type resistance furnace. The temperature was set to 650°C and the holding time was 4 h. The AlN powder was obtained after natural cooling to room temperature. To compare the effects of different types of aluminum sources on the synthesis performance of AlN powder, we used Al(NO₃)₃·9H₂O as the aluminum source instead of Al(OH)₃ to prepare AlN powder based on the above experimental process.



Figure 1. Preparation process of AlN powder.

Characterization

We used a Y-2000 x-ray diffractometer to analyze the phase composition of the powder sample at different reaction temperatures and holding times. We used an Avater360 infrared spectrometer to analyze the phase structure of the powder sample. A Quanta FEG 250 scanning electron microscope was used for the analysis of the microstructure of the powder sample.

RESULTS AND DISCUSSION

Phase composition

Figure 2 shows XRD patterns of the powder samples synthesized at different temperatures (1400°C, 1450°C, and 1500°C) and different holding times (2, 3, and 4 h) with Al(OH)₃ as an aluminum source. The reaction

temperature and holding time significantly affect the phase composition of the synthesized powder samples. When the reaction temperature is 1400°C and the holding time is 2 h, strong Al₂O₃ and AlN characteristic diffraction peaks occur in the synthesized powder sample, and no Al(OH)₃ characteristic diffraction peak occurs in the starting material. This result indicates that Al(OH)₃ in the precursor is completely decomposed and generated to Al₂O₃ under these reaction conditions and that the newly generated partial Al₂O₃ and single carbon produced by the decomposition of glucose initiated the carbothermal reduction nitridation reaction to produce AlN powder at 1400°C and holding time of 3 h. The AlN characteristic diffraction peak in the XRD pattern is further enhanced. Meanwhile, the characteristic diffraction peak of Al₂O₃ is significantly weakened, indicating that a large amount of AlN powder are generated in the system in this reaction condition. When the holding time is extended to 4 h at 1400°C, no considerable change is observed in the XRD pattern of the synthesized powder sample. Meanwhile, weak Al₂O₃ diffraction peaks remain in the synthetic product. The AlN diffraction peak shape becomes increasingly sharp, indicating that the crystallinity of the synthesized powder samples is improved. When the reaction temperature is increased to 1450°C and the holding time is 2 h, weak Al₂O₃ diffraction peaks and strong AlN diffraction peaks occur in the synthesized product. When the holding time is extended to 3 h at 1450°C, the XRD pattern indicates that the diffraction peak of the AlN crystal phase mainly exists in the system, and the diffraction peak of Al₂O₃ crystal phase disappears. This result indicates that the carbothermal reduction nitridation reaction is completed among the mixed materials in the system under these conditions. No significant change is observed in the XRD pattern of the powder sample synthesized at 1450°C and holding



Figure 2. XRD patterns of the powder samples synthesized from precursor mixtures with $Al(OH)_3$ as aluminum source under different reaction conditions.

aracteristic of 2 h under this experimental condition. Figure 3 shows the XRD patterns of the powder samples synthesized with $Al(NO_3)_3 \cdot 9H_2O$ as an aluminum source at different reaction temperatures (1400, 1450 and 1500°C) and different holding times (2, 3, and 4 h). At a reaction temperature of 1400°C and the holding time of 2 h, strong characteristic diffraction peaks of Al_2O_3 and AlN occur in the XRD patterns. This result indicates that a large amount of Al_2O_3 and AlN powder was generated in the system under these reaction conditions. Compared with Figure 2, with $Al(NO_3)_3 \cdot 9H_2O$ as the aluminum source, under the reaction conditions of

powder was generated in the system under these reaction conditions. Compared with Figure 2, with Al(NO₃)₃·9H₂O as the aluminum source, under the reaction conditions of 1450°C and 3 h of holding time or 1500°C and 2 h of holding time, strong AlN characteristic diffraction peaks and weak Al₂O₃ characteristic diffraction peaks occur in the XRD patterns of the synthesized product. When Al(NO₃)₃·9H₂O is used as an aluminum source, singlephase AlN powder can be obtained at 1450°C and 4 h of holding time.

time of 4 h. The main crystal phase remains AlN. When

the reaction temperature is 1500°C and the holding time

is 2 h, only the AlN characteristic diffraction peak is

observed in the XRD patterns of the synthesized powder

sample. Thus, a single-phase AlN powder is generated in the system under these conditions. The above analysis

shows the different proportions of Al₂O₃-AlN composite

powder or single AlN powder can be synthesized by

controlling the reaction temperature or holding time

according to the requirements for the use of synthetic

materials. Single-phase AlN powder can be obtained at

1450°C and holding time of 3 h or at 1500°C and holding

According to the results of the comparison between Figures 2 and 3, compared with that with $Al(NO_3)_3 \cdot 9H_2O$ as aluminum source, carbothermal reduction-nitridation reaction of the precursor mixture with $Al(OH)_3$ as aluminum source is conducted more easily because



Figure 3. XRD patterns of the powder samples synthesized from precursor mixtures with $Al(NO_3)3.9H_2O$ as aluminum source under different reaction conditions.

Al(OH)₃ will undergo decomposition high temperature and generate γ -Al₂O₃ crystal phase. In phase transition, the lattice is distorted and even forms a broken bond. These effects result in defects and volume changes in the material in the system. Volume changes cause cracks in the material, thereby increasing the diffusion coefficient and mass transfer rate and greatly enhancing reaction activity [23]. The phase transition of $Al(NO_3)_3 \cdot 9H_2O$ at high temperatures shows a similar effect, although the effect is slightly worse than that from Al(OH)₃ possibly because Al(NO₃)₃·9H₂O uses crystal water, which is continuously precipitated from the system at high temperatures. Furthermore, -OH is changed into -O-, which subsequently binds to the Al atom. The resulting bonds increase the α -Al₂O₃ crystal phase in the system. However, α -Al₂O₃ with stable crystal form and relatively poor activity is not conducive to the reaction of carbothermal reduction and nitriding in the system [24-25]. Therefore, the comprehensive analysis shows that Al(OH)₃ is a more suitable aluminum source than $Al(NO_3)_3 \cdot 9H_2O$ in this experimental system.

To further analyze the composition of the synthesized powder sample, we use the KBr mixed compression method to prepare samples. We use commercially available single-phase AlN, commercially available single-phase Al₂O₃, and the powder samples obtained at 1400°C for 4 h from the precursor mixture with Al(OH)₃, and Al(NO₃)₃·9H₂O as aluminum sources (marked as L3 and S3 respectively) for FTIR analysis. The result is shown in Figure 4. The (AlN)₂ infrared characteristic adsorption peaks occur near 660, 780, and 1300 cm⁻¹ in Figures 4a, c, and d, respectively. Some weak peaks in 500 - 1500 cm⁻¹ are caused by the Al-N bonds in trace Al/N binary byproducts (Al_xN_y) [26], thereby indicating that AlN exists in the sample reacting for 4 h at 1400°C with Al(OH)₃ and Al(NO₃)₃·9H₂O as aluminum sources. In Figures 4b, c, and d, an Al₂O₃ infrared adsorption peak appears in the position near 1750 cm⁻¹, thereby indicating



Figure 4. FTIR patterns of commercially available single-phase AlN, commercially available single-phase Al_2O_3 , and the powder samples obtained at 1400°C for 4 h from the precursor mixture with Al(OH)₃, and Al(NO₃)₃·9H₂O as aluminum sources.

that Al_2O_3 occurs in the samples synthesized at 1400°C at a holding time of 4 h with $Al(OH)_3$ and $Al(NO_3)_3$ ·9H₂O as precursor mixtures. This finding is consistent with the previous XRD analysis results. Moreover, the infrared spectrum indicates that a small amount of water and N_2 is adsorbed on the surface of the synthesized powder sample. The adsorption band near 1630 cm⁻¹ corresponds to N_2 and the peak near 3450 cm⁻¹ is O-H vibration peak in water.

Synthesis reaction weight loss

We selected glucose as a carbon source and $Al(OH)_3$ or $Al(NO_3)_3 \cdot 9H_2O$ as an aluminum source. The processes involved in the synthesis of AlN powder by carbothermal reduction and nitriding reaction generate CO gas, H_2O gaseous matter, and partial C_xH_y gaseous substance. These gaseous substances result in mass loss in the sample. Therefore, masses of these substances can be used in calculation of mass weight loss rate, which is used to determine the synthesis reaction of AlN powder. Tables 1 and 2 show the mass weight loss rates in the synthesis process with $Al(OH)_3$ and $Al(NO_3)_3 \cdot 9H_2O$ as aluminum sources, respectively.

In Table 1, an increasing trend is observed in the mass weight loss rate of the synthesized powder sample at increased reaction temperature or holding time when Al(OH)₃ is used as an aluminum source. At a reaction temperature of 1400°C and holding time of 2 h, the powder sample has a high mass weight loss rate (76.76%). This result is attributed to escape H₂O gaseous

Table 1. Mass loss rate of the powder sample in the synthesis with $Al(OH)_3$ as aluminum source.

| NO. | Reaction temperature (°C) | Holding time (h) | Mass loss rate (%) |
|-----|------------------------------|---------------------|-----------------------|
| L1 | 1400 | 2 | 76.76 |
| L2 | 1400 | 3 | 83.21 |
| L3 | 1400 | 4 | 83.52 |
| L4 | 1450 | 2 | 80.60 |
| L5 | 1450 | 3 | 83.75 |
| L6 | 1450 | 4 | 84.54 |
| L7 | 1500 | 2 | 84.80 |

Table 2. Mass loss rate of the powder sample in the synthesis with $Al(NO_3)_3$.9H₂O as aluminum source.

| NO. | Reaction temperature (°C) | Holding time (h) | Mass loss rate (%) |
|-----|------------------------------|---------------------|-----------------------|
| S1 | 1400 | 2 | 79.53 |
| S2 | 1400 | 3 | 81.66 |
| S3 | 1400 | 4 | 82.04 |
| S4 | 1450 | 2 | 82.73 |
| S5 | 1450 | 3 | 83.39 |
| S6 | 1450 | 4 | 84.75 |
| S7 | 1500 | 2 | 83.74 |
| | | | |

matter after the decomposition of glucose and aluminum hydroxide in the precursor mixture under this reaction condition and to the decomposition of glucose in the raw material and production of single carbon and H₂O gaseous matter. The decomposition of glucose produces CO, CO₂, acetic acid, acetone and other gaseous products that subsequently escape with air flow, thereby resulting in significant mass loss [27]. The presence of the TG-DTA curve of single glucose materials indicates that [28] the mass loss of glucose can reach 81.16 % (17.52 % higher than theoretical mass weight loss) when glucose is decomposed at high temperatures. This finding further verifies the above analysis results. When the reaction temperature is 1400°C and the holding time is increased to 3 h, the weight loss rate of the synthesis reaction of the powder sample increases to 83.21 %. When the reaction temperature is 1400°C and the holding time is 4 h, the weight loss rate of the synthesis reaction of the powder sample is increased by only 0.31 %. Meanwhile, at a constant holding time but increased reaction temperature, the weight loss rate of synthesis reaction of the powder sample shows a gradual increasing trend. For example, when the reaction time is 2 h, the weight loss rate of synthesis reaction at 1400°C is 76.76. Furthermore, when the reaction temperature is increased to 1450°C and 1500°C, the weight loss rates of synthesis reaction are increased to 80.60 % and 84.80 %. Table 2 indicates that the change in mass weight loss of synthesis reaction with Al(NO₃)₃•9H₂O as aluminum source shows a similar trend to that with Al(OH)₃ as an aluminum source. The above analysis shows that under these experimental conditions, the increase in the reaction temperature or the extension of the holding time is conducive to the synthesis of the powder sample. This finding is similar to the XRD analysis results.

Microstructure and EDS

Figure 5 shows SEM photos of the synthesized powder samples at different reaction conditions with Al(OH)₃ as the aluminum source. In Figures 5a-c, the powder samples generated at a reaction temperature of 1400°C are mainly composed of flaky, short-rod, and irregularly shaped particles, which form a large sheet or lump aggregate crystal structure. The powder samples obtained at 1400°C and holding of 2 h are mainly composed of many flaky 100 - 300 nm particles and some nearly spherical 50 - 100 nm particles. They form a wrapped-type aggregate structure (Figure 5a). At increased holding time, the amount of flaky particles synthesized in the powder samples gradually decreases, whereas short-rod particles and spherical particles gradually increase. When the reaction temperature is 1400°C and the holding time is 4 h, the powder samples synthesized in the system form a variety of microstructures comprising flaky, short rod, and spherical particles. They mainly aggregate in flaky shapes (Figutre 5c). By combining with the previous XRD analysis, we find that the powder samples shown in Figures 5a-c are mainly AlN-Al₂O₃ composition powder synthesized in the system. In contrast to the powder sample synthesized at 1400°C, the powder sample synthesized at 1450°C has a slightly changed microstructure (Figures 5d-f). When the reaction temperature is 1450°C and the holding time is 2 h, the powder sample is composed of 1-2 μ m lamellar aggregates (accumulations). The aggregates are mainly composed of flaky, short-rod, or approximately spherical particles with sizes of 50 - 100 nm (Figure 5d). When the holding time is increased from 3 h and 4 h, the micromorphologies of the synthesized powder samples have no significant change. However, the sizes of these



a) 1400°C + 2 h

Figure 5. SEM photos of the synthesized powder samples at different reaction conditions with $Al(OH)_3$ as the aluminum source. *(Continue on next page)*



b) 1400°C + 3 h



c) 1400°C + 4 h



d) 1450°C + 2 h

Figure 5. SEM photos of the synthesized powder samples at different reaction conditions with $Al(OH)_3$ as the aluminum source. *(Continue on next page)*

Effect of reaction conditions on the synthesis of ultrafine aln powder with glucose as carbon source



e) 1450°C + 3 h



f) 1450°C + 4 h



g) 1500°C + 2 h

Figure 5. SEM photos of the synthesized powder samples at different reaction conditions with Al(OH)₃ as the aluminum source.

small particles increases, and the structures of the aggregates change and form a wrapped-type structure. By combining with the previous XRD analysis, we find that the powder samples in the wrapped-type structure shown in Figures 5e-f are mainly AlN particles. The micromorphology of the powder sample synthesized at 1500°C and 2 h of holding time (mainly AlN particles) is basically the same as that synthesized at 1400°C and 4 h of holding time (Figure 5g). The particles in Figure 5e are finer and even more uniform with respect to size than those in Figures 5f and g and are mostly 50 - 100 nm in size. The above analysis result shows that the best reaction conditions for the synthesis of single-phase AlN powder are temperature of 1450°C and holding time of 2 h. This is consistent with the previous XRD analysis results.

Figures 6 and 7 show the SEM photos and EDS analysis results of the powder samples obtained through

a 3 h reaction that used Al(OH)₃ as aluminum source at 1400°C and 1450°C, respectively. The EDS spectrum in Figure 6 shows that Al, N, O, C, and other elements occur in the synthesized powder samples and that the contents of Al, N, O, and C elements in the samples are 45.17 %, 12.42 %, 31.47 %, and 10.32 %, respectively. The presence of a few carbon elements may be ascribed to the residual carbon in the reaction products. As shown in Figure 7, the EDS spectrum shows that the contents of Al, N, and O elements in the synthesized powder sample are 68.26 %, 20.97 %, and 2.51 %, respectively, thereby indicating that mainly Al and N exist in the synthesized product. The above analysis shows that EDS analysis results are consistent with the previous XRD analysis results.

Figure 8 shows SEM photos of the powder samples synthesized with $Al(NO_3)_3$ $\cdot 9H_2O$ as aluminum source under different reaction conditions. Figures 8a-c indi-



Figure 6. SEM photos (a) and EDS analysis result (b) of the powder sample at 1400° C and 3 h of reaction with Al(OH)₃ as aluminum source.



Figure 7. SEM photos (a) and EDS analysis result (b) the powder sample at 1450°C and 3 h of reaction with Al(OH)₃ as aluminum source.

Effect of reaction conditions on the synthesis of ultrafine aln powder with glucose as carbon source



a) 1400°C + 2 h



b) 1400°C + 3 h



c) 1400°C + 4 h

Figure 8. SEM photos of the synthesized powder samples at different reaction conditions with $Al(NO_3)_3 \cdot 9H_2O$ as the aluminum source. (*Continue on next page*)



d) 1450°C + 2 h



e) 1450°C + 3 h



f) 1450°C + 4 h

Figure 8. SEM photos of the synthesized powder samples at different reaction conditions with $Al(NO_3)_3 \cdot 9H_2O$ as the aluminum source. (*Continue on next page*)



g) 1500°C + 2 h

Figure 8. SEM photos of the synthesized powder samples at different reaction conditions with $Al(NO_3)_3 \cdot 9H_2O$ as the aluminum source.

cate that most of the synthesized powder samples are microstructures of approximately spherical particles (approximately 50 - 100 nm), and a certain degree of agglomeration occurs, thereby forming a certain shape of the collective structure. Figures 8d-f indicate that the micromorphologies of the powder samples synthesized at 1450°C and different holding times are the same as those at 1400°C. At the same reaction temperature, given the extension in holding time, approximately spherical particles of the synthesized powder sample are slightly increased in the size. Moreover, the particles in the powder sample obtained at 1450°C and 4 h of holding time are slightly sintered. Figure 8g shows that the powder sample prepared at 1500°C and 2 h of holding time is mainly composed of many uniformly dispersed approximately spherical particles (approximately 50 - 100 nm) and

a few flaky particles (approximately 100-300 nm).

Figure 9 shows SEM photos and EDS analysis results of the powder sample synthesized with $Al(NO_3)_3 \cdot 9H_2O$ as an aluminum source at 1450°C and 3 h of reaction. As shown the figure, the contents of Al, N, and O elements in the powder samples are 60.31 %, 24.57 %, and 10.57 %, respectively. When compared with Figure 7, the content of O in Figure 9 is considerably high because of the strong AlN and weak Al_2O_3 characteristic diffraction peaks in the powder sample synthesized at 1450°C and 3 h reaction when $Al(NO_3)_3 \cdot 9H_2O$ is used as an aluminum source. This result indicates that a large amount of AlN powder and a small amounts of Al_2O_3 powder are present in the synthesized product. The powder sample synthesized through a 3 h reaction with $Al(OH)_3$ aluminum



Figure 9. SEM photos and EDS analysis result of the powder sample at 1450°C and 3 h of reaction with $Al(NO_3)_3$ ·9H₂O as aluminum source.

source at 1450°C has strong AlN characteristic diffraction peaks, and no Al_2O_3 characteristic diffraction peak is found. Thus, the precursor mixture basically synthesizes single-phase AlN powder under this reaction condition. Therefore, EDS spectrum shows that the proportion of oxygen content is extremely small. The above analysis shows that EDS analysis results are consistent with the previous XRD analysis results.

CONCLUSIONS

- The different proportions of Al₂O₃-AlN composite powder or single AlN powder can be synthesized by controlling the reaction temperature or holding time according to the requirements for the use of synthetic materials. Under the conditions of this experiment, Al(OH)₃ more beneficial as an aluminum source for carbothermal reduction-nitridation than Al(NO₃)₃·9H₂O. Single-phase AlN powder can be obtained at 1450°C and holding time of 3 h or at 1500°C and holding of 2 h using Al(OH)₃ as aluminum source. When Al(NO₃)₃·9H₂O is used as an aluminum source, single-phase AlN powder can be obtained at 1450°C and 4 h of holding time.
- When Al(OH)₃ or Al(NO₃)₃·9H₂O is used as an aluminum source, a high mass weight loss occur in the reaction of the synthesized powder sample, and mass weight loss increased at elevated reaction temperature or extended holding time. This result is attributed to escape H₂O gaseous matter after the decomposition of glucose and aluminum hydroxide in the precursor mixture under this reaction condition and to the decomposition of glucose in the raw material and production of single carbon and H₂O gaseous matter. The decomposition of glucose produces CO, CO₂, acetic acid, acetone and other gaseous products that subsequently escape with air flow, thereby resulting in significant mass loss.
- Different types of aluminum sources, reaction temperature, and holding time significantly affect the synthesized morphologies of the powder samples. When Al(OH)₃ is used as an aluminum source, the singlephase AlN powder sample synthesized at 1450°C and holding of 3 h is mainly composed of many flaky, short-rod, and irregularly shaped particles. Its structure is fine and uniform overall. Furthermore, the particle size is mostly in the range of 50 - 100 nm. When Al(NO₃)₃·9H₂O is used, the single-phase AlN powder sample prepared at 1450°C and holding of 4 h is mainly composed of nearly spherical 100 - 300 nm particles, and some particles are slightly sintered. EDS analysis results are consistent with the previous XRD and SEM analysis results.

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