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Study on Preparation of Homogenized Highly Dispersible AlON Powder

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Abstract: The quality of Aluminum oxynitride (AlON) powder is the main factor affecting the properties of ceramics. To accurately control the nitrogen AlON content and improve the dispersion degree of powder, AlON powder was prepared from γ-Al_2O_3 and activated carbon powder by carbothermic reduction and nitridation. The uniformity of distribution of raw materials and the dimension and microstructure of AlON powder was controlled by changing the time and rotation rate of the ball milling process. The influences of ball milling process parameters on the microstructure and distribution uniformity of Al_2O_3/C mixed powder, microstructure, and particle size of AlON powder were studied. The results show that powder agglomeration was effectively reduced, and the uniformity of raw material distribution was increased by controlling the time and rotation rate of the ball milling process. The Al_2O_3/C mixed powder has good particle size and uniform distribution with roller ball milling for 24 h. After holding the powder at 1700 ℃ for 60 min, the homogenized AlON powder was obtained, and the residual carbon content was about 0.04 wt%. When the time of planetary ball milling at 250 rpm was 16 h, highly dispersible AlON powder was obtained.

1. Introduction

Yamaguchi and Yanagida first proposed that there might be a spinel structure in the Al_2O_3-AlN binary system [1]. Spinel AlON can not only be used to prepare transparent armor but also as communication and luminescent materials [2].

The structure of AlON includes spinel and wurtzite [3]. Wurtzite structure belongs to a hexagonal crystal system, and the transmittance of AlON transparent ceramics is anisotropic. Light rays will be birefringent when passing through the grain boundary, which reduces the transmittance of AlON transparent ceramics. The spinel structure belongs to the cubic crystal system, and its optical properties are isotropic without birefringence. Therefore, spinel AlON shows excellent optical properties. When the structure of AlON is spinel, the content of AlN is 16.7 - 40 mol% [4]. With the increase of AlN content, the vacancy concentration of AlON decreases the constant lattice increases, and the refractive index of AlON increases [5]. The difference in nitrogen content in adjacent AlON grains can cause light scattering, so the control of nitrogen content in AlON is of great significance.

The properties of powders have a great influence on the properties of AlON transparent ceramics. Solid-state reaction [6], carbothermic reduction and nitridation [7], and aluminothermic reduction [8] were the main preparation methods of AlON powder preparation. The solid-state reaction requires High purity ultra-fine Al_2O_3 and AlN powder as raw materials, which was kept at 1800 ℃ for a long time and had a high cost. A large amount of AlON powder can be prepared quickly by the aluminothermic reduction, but the controllability of the reaction is poor, and the reaction products usually contain Al_2O_3. Carbothermic reduction and nitridation is a common method for preparing AlON powders. Because of its advantages of low raw material cost, high purity of reaction products, and strong controllability. The synthesis temperature of the three AlON synthesis methods was above 1700 ℃. The heat preservation at high temperatures will lead to the growth of AlON grains and reduce the sintering activity of the powder, which is detrimental to the process of AlON ceramics sintering. In this paper, AlON powder was prepared using Al_2O_3 and activated carbon as raw materials using the carbothermal reduction nitriding method. High dispersion AlON powder was obtained from planetary ball milling treatment.

2. Experiments

2.1 Experimental materials

γ-Al_2O_3 (> 99.99%, Beijing Deke Daojin Science and Technology Co., Ltd., Beijing, China) and carbon (Sinopharm Chemical Reagent Co. Ltd, Shanghai, China) were used as raw materials in AlON powders synthesis.

2.2 AlON synthesis

The mass ratio of Al_2O_3: C was set to 94.2: 5.8. The raw materials were mixed by different ball milling processes, with anhydrous ethanol as the grinding medium and Al_2O_3 ball as the grinding ball. The ball milling process parameters were shown in Table 1. The obtained slurry was placed in the oven, and dried at 80 ℃ for 12 h. The mixed powder was placed in a vacuum graphite furnace, held for 1 h at 1550 ℃ and 1700 ℃, respectively, in the atmosphere of flowing nitrogen, and then passed through a 200-mesh screen. The synthesized AlON powder was refined by planetary milling. The technological parameters of planetary ball milling were shown in Table 2.

Table 1. Related parameters of ball milling

No.	ratio of ball to powder	type of ball milling	rotation rate (rpm)	time (hour)
H12	5	roller	60	12
H24	5	roller	60	24
P1-1	7	planetary	200	6
P1-2	7	planetary	200	12
P2-1	7	planetary	250	6
P2-2	7	planetary	250	12

Table 2. Related parameters of ball milling

No.	Al_2O_3 ball mass(g)			Ultrasonic time (min)	rotation rate (rpm)
	3mm	5mm	10mm		
1	0	350	150	0	300
2	250	150	100	0	250
3	250	150	100	0	300
4	250	150	100	10	300

2.3 Characterization

The microstructure was detected by scanning electron microscopy (SEM, JSM-F100, JEOL, Japan) and the element distribution was analyzed by X-ray energy spectrum analysis (OCTANE elect EDS, USA). The particle size was detected by a laser particle sizer (Bettersize-2000, Liaoning, China). The Phase analysis was seen by X-ray diffractometry (XRD, D/max 2500, Rigaku, Japan). The content of carbon was tested by a High-frequency infrared carbon and sulfur analyzer (LECO, CS844, USA).

3. Results and discussion

3.1 Effect of ball milling process on morphology and uniformity of raw materials

FESEM micrographs of Al_2O_3/C mixed powder by different ball milling and EDS mapping of the same area were shown in Fig 1. In Fig.1 H12, the Al_2O_3/C mixed powder aggregated after roller ball milling for 12 h, and rod-like carbon particles with a length of more than 10 μm appeared in the agglomerated particle. As shown in Fig. 1 H24, the agglomeration phenomenon of the powder was significantly inhibited, and the powder particles were fine and dispersed with ball milling for 24 h. It can be seen from the EDS mapping that the raw materials were evenly distributed, and no evident enrichment occurred. The mixed powder was ground for 6 h (200 rpm, 300 rpm) by planetary ball milling, as shown in Fig. 1, P1-1, and P2-1. It can be observed that the mixed powder was finely dispersed, but large-sized spherical and rod-like particles appeared in all of them. When the grinding time extended to 12 h (200 rpm, 300 rpm), SEM and EDS were shown in Fig. 1, P1-2, and P2-2. When the rotation rate was 200 rpm, the mixed powder was finely dispersed, and the raw material was not enriched. When the rotational speed increased to 300 rpm, the fine Al_2O_3 powder agglomeration was evident. Large-size activated carbon particles will lead to excessive AlN generated, resulting in high nitrogen content of AlON particles. When the Al_2O_3 particles aggregated, the activated carbon in the aggregated particles was less, resulting in the low nitrogen content of AlON particles and the uneven nitrogen content of AlON. As shown in Fig. 1 H24 and P1-2, the size of aggregate particles of the mixed powder obtained by roller ball milling for 24 h and spherical milling for 12 h (300 rpm) was small, and the size of H24 particle was minor than P1-2, so H24 was chosen as the mixing method of raw material.

The Al_2O_3/C mixed powder was held for 1 h at 1550 ℃ and 1700 ℃, respectively, and repeated the experiment three times. The XRD diffraction pattern and residual carbon content of the reaction product were shown in Fig. 2. As shown in Fig. 2, the reaction products were all single-phase AlON, and their carbon content was 0.038 wt%, 0.044 wt%, and 0.042 wt%, respectively. The effective activated carbon of 5.76 wt%, so the nitrogen content of AlON was similar.

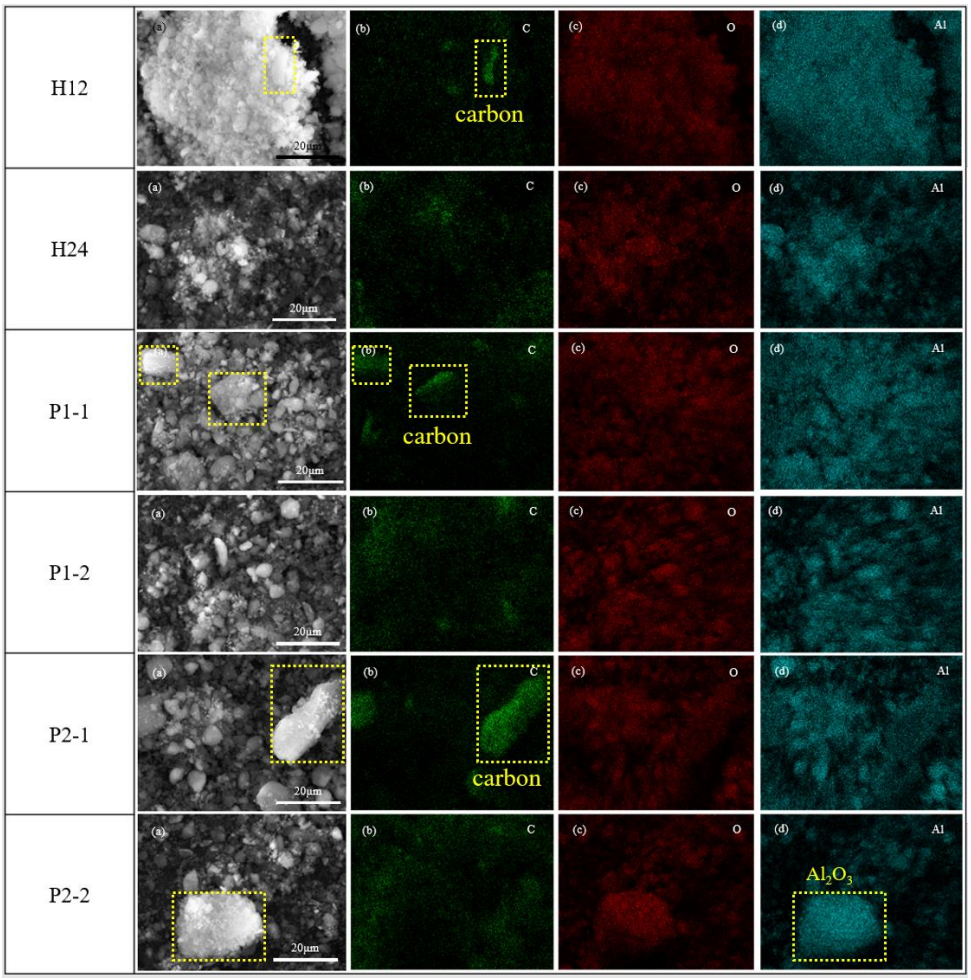

Fig. 1. FESEM micrographs of powders by different ball milling and EDS mapping of the same area: (a) FESEM micrographs of powders; (b) carbon is green; (c) oxygen is red; (d) aluminum is blue

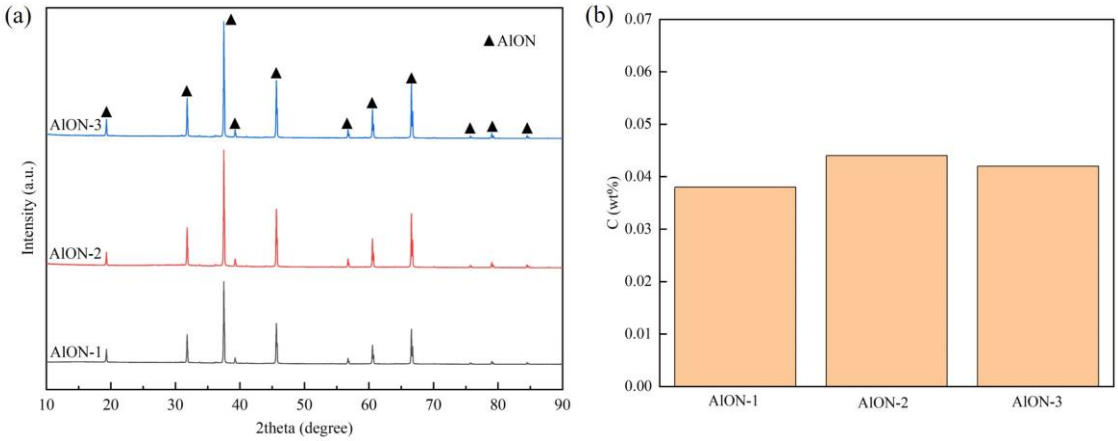

Fig. 2. (a) XRD diffraction pattern of the reaction product; (b) the carbon content of the reaction products

3.2 Effect of planetary ball milling on particle size and morphology of AlON

When the synthesis temperature of AlON powder was 1700 ℃, the powder was easy to gather and grow at this temperature (D_{50}=23.45 μm). . The particle size of the powder was too large, which was not

conducive to forming raw material and ceramic sintering. The microstructure of the AlON powder held at 1700 ℃ for 1 h was shown in Fig. 3. The particles were spherical and ellipsoidal, and there were connections between particles.

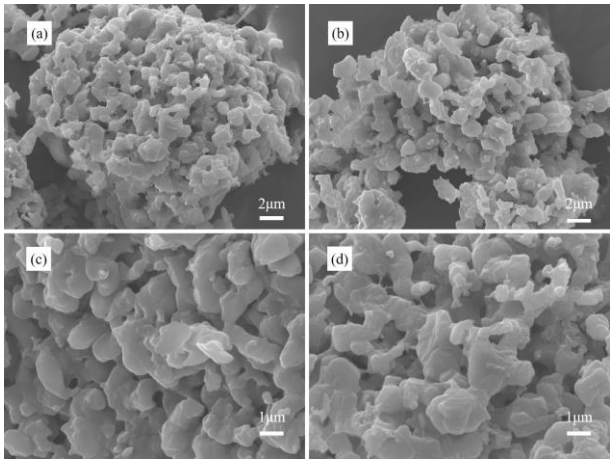

Fig. 3. Microstructure of the AlON powder prepared with holding at 1700 ℃ for 1 h

The effect of grind balls ratio and ball milling time on particle size were shown in Fig.4. As was shown in Fig. 4 NO.1, when the ball milling time was up to 12 h, the particle size of the powder decreased. The powder started to aggregate, and the particle size increased with the ball milling time increased for 12 h. The particle size was the smallest (D_{50} = 1.954 μm) with ball milling for 12 h., the tiny particles aggregated again with the ball milling time of more than 12 h. The powder's particle size increased with the ball milling time extension. It was shown in Fig. 4 NO.2 and NO.3 that the increase of small size grinding ball can effectively reduce the particle size. Meanwhile, the powder agglomeration was inhibited in the ball milling process. The number of small-size ball mills increased, the contact area between the powder and the grind ball increased, and the impact of the powder increased, so the powder agglomeration was inhibited. After running at 250 rpm and 300 rpm for 16 h, the sizes of AlON powders were 0.724 μm and 0.642 μm, respectively, with a slight difference in particle size. It can be seen from the comparison between No.3 and No.4 in Fig. 4 that ultrasonic treatment has no significant influence on small-size particles. At this time, the AlON particles were small and dispersed, and the agglomeration was less, so there was no apparent change in particle size after the ultrasound.

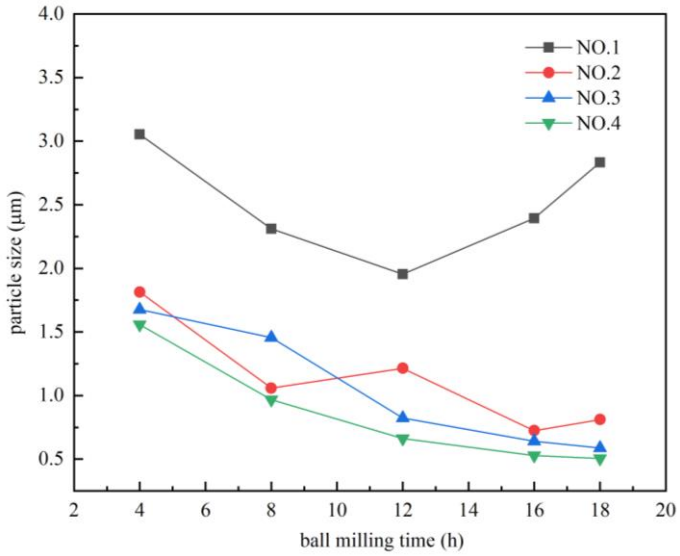

Fig. 4. The effect of grind balls ratio and ball milling time on particle size

The microstructure of AlON powder after 16 h in planetary ball milling was shown in Fig. 5. The aggregated AlON powder was dispersed, and the particles were spherical and ellipsoidal. When the rotation rate was 250 rpm, there were almost no particles larger than 1 μm. When the rotation rate increased to 300 rpm, there were particles larger than 1 μm. In conclusion, long-time ball milling and high rotation speed were not conducive to obtaining high-dispersion AlON powder.

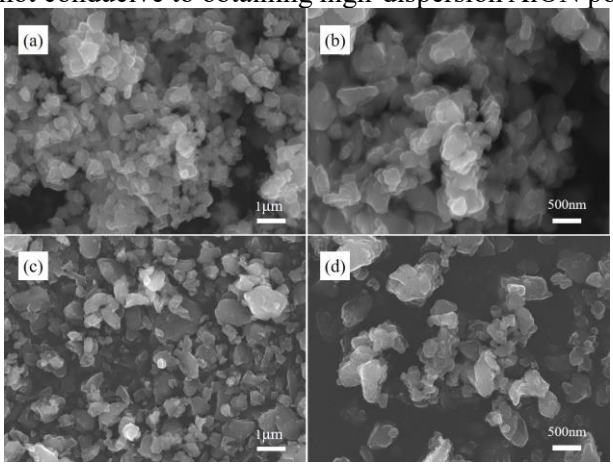

Fig. 5. Microstructure of AlON powder after planetary grinding at different rotation rates

4. Conclusion

Because of the problems of difficult composition control, and large particle size in the process of AlON powder preparation, the mixed raw materials, and AlON powder were treated by ball milling. γ-Al_2O_3 and activated carbon powder were used as raw materials and anhydrous ethanol as a medium. The influence of ball milling parameters on the distribution uniformity and microstructure of raw materials was studied in this paper. The results showed that prolongation of ball milling time can effectively inhibit powder agglomeration. The fine dispersed mixed powder was obtained after 24 h roller ball milling and 12 h planetary milling (200 rpm), and AlON powder with uniform nitrogen content was obtained by holding at 1700 ℃ for 1 h. The AlON powder was refined by planetary ball milling and ultrasonic dispersion. AlON powder was easy to agglomerate in ball milling, and the agglomerate phenomenon can be effectively inhibited by adding small-size grinding balls. The effect of ultrasonic dispersion on finely dispersed particles was not significant. Highly dispersible AlON powder with 0.724 μm was obtained by planetary grinding at 250 rpm for 16 h. In this paper, AlON powders with high dispersion and homogenization were synthesized, which provided clear guidance for the content control and morphology optimization of AlON powders.

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