Advanced Powder Technology xxx (2015) xxx–xxx

Contents lists available at ScienceDirect

Advanced Powder Technology

journal homepage: www.elsevier.com/locate/apt

Original Research Paper

Preparation and formation mechanism of aluminum nitride ceramic particles from large aluminum powder by self-propagating high temperature synthesis

Liang Qiao *, Shuwen Chen, Jingwu Zheng, Liqiang Jiang, Shenglei Che

College of Materials Science and Engineering, Zhejiang University of Technology, Hangzhou 310014, China

ARTICLE INFO

Article history:
Received 9 October 2014
Received in revised form 28 January 2015
Accepted 15 February 2015
Available online xxxx

Keywords:
Aluminum nitride
Self-propagating high temperature synthesis
Mechanism of particle formation
Sintering

ABSTRACT

Aluminum nitride ceramic powders were prepared by self-propagating high temperature synthesis using the large spherical aluminum powders as reactants. The sintering behavior of the as-received powder was characterized using the conventional liquid sintering method. X-ray diffraction and scanning electron microscope were used to detect the phase compositions and observe the morphologies of the products. The results show that the large aluminum particles evolve into the aggregates consisting of many small AlN particles with spherical shape and stable size. The small spherical particles after deagglomeration by ball milling can be densely sintered at 1830 °C for 2 h. The mechanism that the Al liquid formed at the high temperature repeatedly flows and reacts with the coming nitrogen gas was proposed to explain the evolution from the large aluminum particle into the aggregate with small particles. Some further experiments were also carried out to demonstrate this mechanism.

1. Introduction

Aluminum nitride is considered to be a promising substrate and package material for high power integrated circuits because of its high thermal conductivity, low dielectric constant and thermal expansion coefficient close to that of silicon [1–3]. However, the cost of the raw powder is high to limit its applications in a wide range. The self-propagating high-temperature synthesis (SHS) is attractive due to the quick nitridation reaction and cost-effective alternative for the AlN production [4–8]. Many factors such as the Al contents, the additives, and the pressure of nitrogen gas, which affect the transformation from Al to AlN in the SHS method, have been studied in the previous literatures [9–12]. However, the mechanism of the formation and growth of AlN particle in the process is not clear up to now. In this paper, the AlN particles were prepared by SHS method and the sintering behavior was characterized. In addition, the possible formation mechanism was proposed and discussed based on the microstructure observation.

2. Experimental method and material

The combustion experiments were carried out in the SHS autoclave with the volume of 60 L. The commercial spherical aluminum powders (d_{50} = 28.76 μm, GJB1738-93) and the aluminum nitride powder (d_{50} = 1.47 μm, Toyo aluminum J powder) were used as raw reactants and moderator respectively. The mass ratio of Al and AlN is 2:3. NH_4Cl with 1 wt% content was added into the mixture of Al and AlN to absorb a part of heat in the combustion. Then the mixture was ball-milled for 12 h at the rate of 30 rev/min using the ZrO_2 balls with the size of 10 mm in diameter. Subsequently, they were ignited using a tungsten filament and reacted under the nitrogen pressure of 8 MPa. The as-received products were broken and ball-milled for 48 h for deagglomeration. Then the as-received AlN particles were used as moderators in the next cycle of the same process. After more than five cycles to eliminate the effect of the starting moderator, the final AlN powder was achieved. To study the sintering properties of the AlN powders, commercial Y_2O_3 was used as sintering aid with the amounts of 3 wt%, 5 wt%, 7.5 wt% and 10 wt%, respectively. After ball milled using ethanol as the mixing medium and dried in vacuum oven, the mixed powders of Y_2O_3 and AlN were granulated with polyvinylbutyral (PVB), and then pressed into Φ23 mm × 2.5 mm pellets. The green bodies were sintered in the graphite furnace at 1800–1890 °C for 2 h in a flowing nitrogen atmosphere after de-waxed at 600 °C in the air.

The size distribution of the as-received AlN powder was measured by laser particle size analyzer (Microtrac SDC). The phase compositions of the SHS product were identified by X-ray diffraction (XRD, PANalytical X'Pert PRO) with Cu Ka. The densities of

* Corresponding author. Tel./fax: +86 571 88320142.
E-mail address: lqiao@zjut.edu.cn (L. Qiao).

http://dx.doi.org/10.1016/j.apt.2015.02.007
0921-8831/

the sintered pellets were measured by Archimedes displacement method, respectively. SEM (Hitachi SU1510) was used to observe the particle morphologies of the powders and the sintered pellets.

3. Results

The starting mixture becomes the whole block with a little strength like white bread after combustion. Fig. 1 shows the phase compositions of the surface and middle parts of the product. On the surface, there is a grey layer containing Al and AlN phases (in Fig. 1(a)), which indicates that the aluminum powder on the surface of the starting mixture is hard to be reacted completely though the nitrogen gas is sufficient there. Differently, the middle part under the grey layer displays the homogenous white product with the single AlN phase (in Fig. 1(b)). Because the grey layer containing Al on the surface is very thin, the AlN powder can be achieved after a simple separation.

Fig. 2 shows the typical morphologies of the AlN products. It can be seen that many AlN particles with small sizes lower than 10 μm stack closely into large aggregates, which sizes are similar with that of the original Al particles as shown in Fig. 2(a). This implies that the aggregates are evolved from the original aluminum particles. The profiles of the aggregates with a hole left in the center can also be observed as seen in Fig. 2(b). Though some irregular morphologies can be found due to the complex combustion environment as reported by some literatures, the aggregates, which consist of small spherical particles, are the main feature of the product particles.

Fig. 3 shows the size distribution of the AlN powder after deagglomeration by ball milling. It can be seen that most particles have the size lower than 8 μm, which is consistent with the size of the small AlN particles on the aggregates as shown in Fig. 2. The effective milling must be provided to further crush the small particles for the fine size lower than 2 μm (d_{50}).

Fig. 4 shows the relation between the densities and sintering temperatures of AlN ceramics with different additions of Y_2O_3 using the SHS powder. In general, the theoretical density of AlN is 3.26 g/cm^3. However, the sintered body of AlN has always a larger value around 3.24–3.4 g/cm^3 due to the existence of Y–Al–O compounds when different Y_2O_3 contents are used as the sintering additives. Therefore, the measured densities were used in Fig. 4 rather than the related one of theoretical density. It can be observed that the densities of sintered samples increase as the sintering temperatures and the Y_2O_3 contents increase. The densities increase significantly when the sintering temperatures are in the range of 1800–1830 °C. They begin to become stable when the temperature reaches 1830 °C, which indicates the powders have been densely sintered at this temperature.

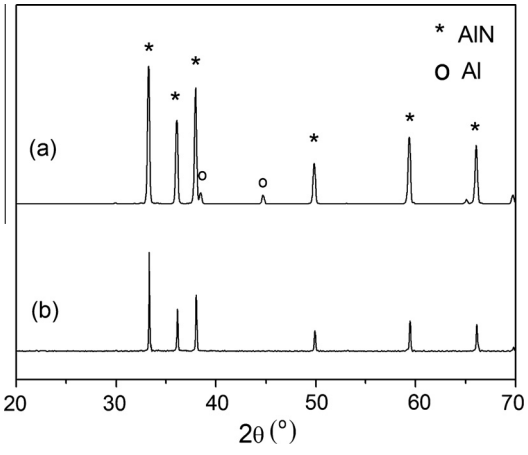

Fig. 1. XRD patterns of the surface (a) and middle part (b) of the SHS product.

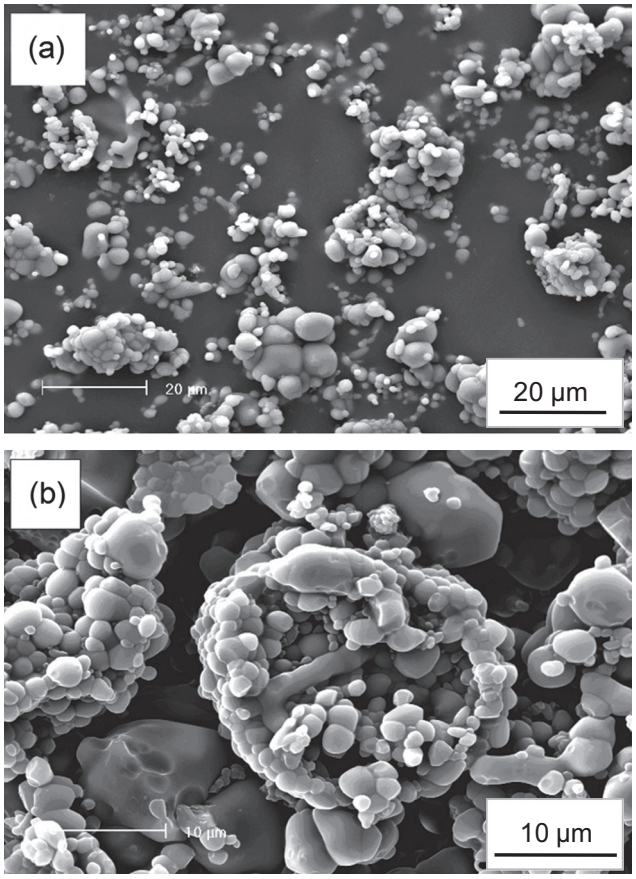

Fig. 2. Morphologies of AlN products. (a) aggregates with small particles and (b) aggregates with the hole in the center.

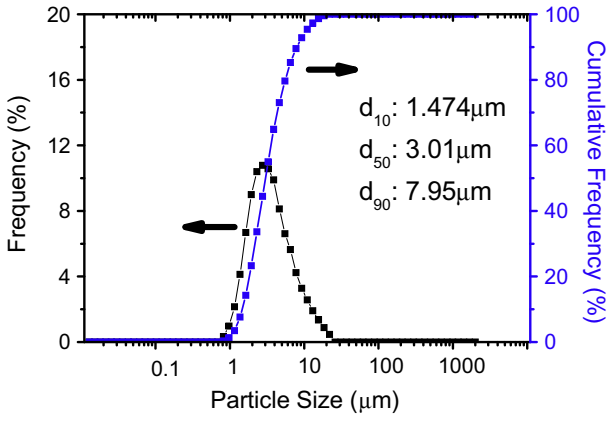

Fig. 3. Size distribution of the AlN powder after deagglomeration by ball milling.

Figs. 5 and 6 show the BSE images of AlN green bodies before sintering and ceramic bodies after sintered at 1830 °C for 2 h with different Y_2O_3 contents, respectively. It can be seen that there are many pores in the AlN green bodies with the Y_2O_3 particles scattering in them before sintering. However, after sintering, the pores are unobvious with the grains packing closely, which agrees with the increasing densities as shown in Fig. 4. This implies that the densification process becomes slow at 1830–1890 °C. It can also be found that the secondary phases of Y–Al–O compounds detected by XRD well distribute in intergranular and triangle grain boundaries as shown in Fig. 6(c) and (d). The sufficient liquid layers

Please cite this article in press as: L. Qiao et al., Preparation and formation mechanism of aluminum nitride ceramic particles from large aluminum powder by self-propagating high temperature synthesis, Advanced Powder Technology (2015), http://dx.doi.org/10.1016/j.apt.2015.02.007