Materials Science and Engineering A 496 (2008) 269–272

Contents lists available at ScienceDirect

Materials Science and Engineering A

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

Structure and properties of AlN ceramics prepared with spark plasma sintering of ultra-fine powders

Xueli Du[a,b,*], Mingli Qin[c], Abdur Rauf[d], Zhihao Yuan[a], Baohe Yang[b], Xuanhui Qu[c]

[a] Nanomaterials & Nanotechnology Research Center, School of Materials Science and Engineering, Tianjin University of Technology, Tianjin 300191, People's Republic of China
[b] Tianjin Key Lab. of Film & Communicate Devices, Tianjin University of Technology, Tianjin 300191, People's Republic of China
[c] School of Materials Science and Engineering, University of Science and Technology of Beijing, Beijing 100083, People's Republic of China
[d] National Center of Nanotechnology, Pakistan Institute of Engineering & Applied Science, Islamabad 45650, Pakistan

ARTICLE INFO

Article history:
Received 11 October 2007
Received in revised form 9 May 2008
Accepted 12 May 2008

Keywords:
AlN ceramic
Ultra-fine AlN powders
Spark plasma sintering (SPS)
Fracture toughness
Hardness

ABSTRACT

The AlN powders with grain size about 100 nm were synthesized by low-temperature carbon thermal reduction method. The synthesized powders without any additives could be completely densified through spark plasma sintering (SPS) technique above 1470 °C in vacuum under the pressure of 40 MPa. The properties of AlN ceramics sintered at different temperature and pressure were investigated. The results indicated that the density of SPS sintered AlN ceramics decreased with the reduction of sintering pressure and temperature. The grain size of the sintered samples reduced from about 1 μm to 200 nm as the sintering pressure and temperature decreased from 40 MPa and 1600 °C to 30 MPa and 1500 °C. No inter-granular phase was found in the microstructure of sintered samples, which resulted in a strong grain bond and consequently improved the mechanical properties especially the fracture toughness. Moreover, the effects of the grain size and porosity on the mechanical properties of the sintered samples were also discussed.
© 2008 Elsevier B.V. All rights reserved.

1. Introduction

Aluminum nitride (AlN) has interesting properties such as high thermal conductivity, high volume resistance, and moderate dielectric properties. Its thermal expansion coefficient is close to that of silicon and it is one of the mechanically strong and thermally stable ceramics. These excellent attributes make AlN a useful material for widespread applications [1,2]. However, the applications of AlN have been constrained because of several inherent impediments. It is difficult to synthesize pure AlN powders due to chemical instability [3]. Furthermore, owning to high covalent bonding and low self-diffusion coefficients of the constituent elements, the sintering of AlN ceramics is difficult too. Generally, AlN ceramics with complete densification and high thermal conductivity can be obtained at temperature higher than 1800 °C with sintering additives [4,5]. That not only increases the production cost of AlN ceramics but also promotes significant grain growth, which results in deterioration of mechanical properties. Consequently the research of AlN ceramics is oriented toward achieving complete densification at low sintering temperature [6].

Spark plasma sintering (SPS) is a newly developed technique that enables ceramic powders to be fully sintered at relatively low temperature in a very short time [7]. In the SPS process, a pulsed direct current is applied to the sintered powders and the activation of powder particles is thought to be achieved as the application of electrical discharges [8].

The sintering behavior of AlN ceramics is strongly dependent on the size of starting powders. Nanometer AlN particles have shown potential sintering activity for low-temperature sintering [9–12]. The so-called low-temperature (∼1500 °C) carbon thermal reduction is an effective method to synthesize ultra-fine AlN powders [13,14]. The general synthesizing temperature of AlN powders through carbon thermal reduction method is 1600–1800 °C [15,16]. In this paper, 100 nm AlN powders were synthesized through low-temperature carbon thermal reduction method and then sintered with SPS technique. The properties of the synthesized powders and sintered samples were investigated.

2. Experimental

Analytically pure aluminum nitrate ($Al(NO_3)_3{\cdot}9H_2O$), glucose ($C_6H_{12}O_6{\cdot}H_2O$) and urea ($CO(NH_2)_2$), as starting materials, were dissolved in distilled water together. With continuous heating on electric stove the mixed solution was boiled, concentrated and combusted, then producing the powders of Al_2O_3 and C. The mixed powders were thermally reduced to AlN at 1550 °C in the N_2 atmo-

* Corresponding author at: Tianjin University of Technology, Tianjin 300191, People's Republic of China.
Fax: +86 22 2367 9345.
E-mail address: d7288@126.com (X. Du).

0921-5093/$ – see front matter © 2008 Elsevier B.V. All rights reserved.
doi:10.1016/j.msea.2008.05.027

sphere and followed decarburization at 700 °C in the air. The details of the synthesizing procedure have been reported [14].

The synthesized AlN powders without any sintering additives were sintered by SPS (SPS-1050, Sumitomo Coal Mining Co. Ltd., Japan) with a graphite die of 10 mm in diameter. The samples designated as SPS1600 and SPS1500 were heated to the corresponding temperatures of 1600 °C and 1500 °C in 15 min from room temperature and held for 4 min under an applied pressure of 40 MPa and 30 MPa in vacuum, respectively. Commercially AlN powders synthesized by self-propagating high-temperature synthesis with mean grain size 3 μm were doped with 5 wt.% Y_2O_3 and sintered by the same procedure as SPS1600, produced the sample of SPS1600Y. During sintering the temperature was measured by means of a tungsten rhenium alloy thermo-couple.

High frequency combustion infrared absorption (YB/T178.6-2000) was used to analyze carbon content and inert gas pulse heating infrared thermal conductivity (QB-Q-02-97) was used to analyze oxygen and nitrogen content of the synthesized powders. Laser particle size analyzer (LMS-30) was employed to measure particle size distribution. X-ray diffraction (XRD, Rigaku, D/max-RB12, Cu Kα, λ = 0.15406 nm) was used to identify the phase composition. The morphology of the synthesized AlN powders and fracture surfaces of sintered samples were observed by field emission scanning electron microscopy (SEM, SUPRA-55, ZEISS). Density of the sintered sample was tested by Archimedes displacement method. Micro-hardness instrument (HVS-50) was used to measure the Vickers hardness of the sintered samples and fracture toughness was calculated from indentation parameters.

3. Results and discussions

3.1. Properties of synthesized AlN powders

The properties of synthesized AlN powders are shown in Table 1. The powder's mean size is 100 nm and oxygen content is 1.69 wt.%, which is a bit larger than the commonly reported value [14]. This resulted from the oxygen absorption during combustion and decarburization processes, which were carried out in air. However, the oxygen quantity is so small that could not be detected by the XRD analysis. The use of closed reaction chamber and decarburization in hydrogen atmosphere may reduce the oxygen content. Microstructure of the powders is shown in Fig. 1. It is found that the particles of synthesized AlN powders are isotropic and the particle size is coincided with the result in Table 1. Fig. 2 shows XRD pattern of the synthesized powders. AlN is identified as the only crystalline phase.

3.2. Sintering characteristics

Fig. 3 shows the linear shrinkage of the sample SPS1600 during SPS processes under the pressure of 40 MPa in vacuum. L_0 is the initial height of the sample and L is the height of the sample during SPS sintering. The curve clearly indicates that a remarkable shrinkage occurred at the temperature higher than 1100 °C and reached the maximum shrinkage at about 1470 °C. In other words, the synthesized AlN powders without any sintering additives can be fully sintered above 1470 °C by SPS technique under the pressure of 40 MPa. It is interesting to mention that the sintering temperature in this experiment is much lower than that of published literature (>1600 °C, with additives) [1,6].

Table 1
Properties of AlN powders synthesized by low-temperature carbon thermal reduction

Mean particle size (μm)	Specific surface area ($m^2 g^{-1}$)	Chemical composition (wt.%)		
		N	O	C
0.10	17.4	32.56	1.69	0.17

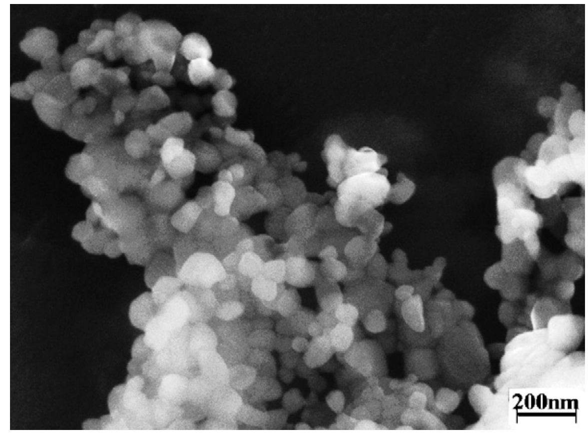

Fig. 1. SEM of the AlN powders thermally reduced at 1550 °C.

3.3. Properties of sintered samples

Fig. 4 and Table 2 illustrate SEM photographs of fracture surfaces and properties of SPS1600, SPS1500 and SPS1600Y, respectively. It can be seen that SPS1600 and SPS1500 with density of 3.28 g cm^{-3} and 3.20 g cm^{-3} are composed of well developed isotropic AlN grains. There is so many visible pores in the microstructure of SPS1600Y that the density of it is only 3.04 g cm^{-3}. SPS1600Y is almost not sintered although it was doped with 5 wt.% Y_2O_3 as sintering additives. This result reveals that the synthesized ultra-fine AlN powders possessed excellent sintering behavior. The indentation test of SPS1600Y was not fulfilled owing to its large porosity.

On comparing with the theoretical density of pure AlN (3.26 g cm^{-3}), the density of SPS1600 (3.28 g cm^{-3}) is slightly higher, which can be attributed to the minor quantity of Al_2O_3 as evident from the oxygen content (Table 1). From Fig. 4(b), some pores are visible, which resulted in the low density of SPS1500. This result implies that the density of SPS sintered AlN ceramics decreased with the reduction of sintering pressure and temperature. The particle sizes of SPS1500 and SPS1600 are about 200 nm and 1 μm, respectively, as estimated from their SEM

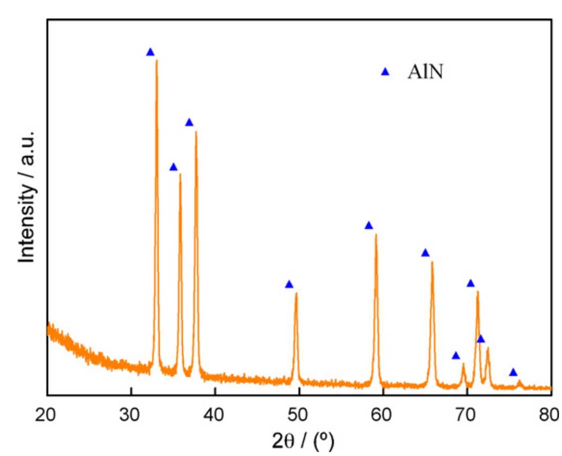

Fig. 2. XRD of the AlN powders thermally reduced at 1550 °C.

Download English Version:

https://daneshyari.com/en/article/1581841

Download Persian Version:

https://daneshyari.com/article/1581841

Daneshyari.com