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Combustion synthesis of network silicon nitride porous ceramics

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Abstract

A porous silicon nitride ceramics, composed of a network interconnected β-Si_3N_4 rods, was prepared by using the combustion synthesis method. Si powder with more than 20 wt.% diluent α-Si_3N_4 was fully nitrided under high nitrogen pressure. The interconnected open porosity of the obtained combustion products was as high as 64.40%. In the combustion products, rod-like β-Si_3N_4 grains were formed to build up the network silicon nitride porous ceramics.

Keywords: B. Porosity; D. Si_3N_4; Combustion synthesis

1. Introduction

Combustion synthesis, or self-propagating high temperature synthesis (SHS) is a technique for producing ceramics, intermetallic, and composite materials [1–3]. It is a process, which utilizes the heat generated by an exothermic reaction to sustain itself in the form of a combustion wave after external ignition. The SHS technique has aroused material researchers' interest because of its advantages such as low processing cost, great energy efficiency, high purity of the products, high production rate as well as environmental friendliness.

Recently, porous ceramics are becoming more and more important to our modern technological world because they find applications in a variety of industrial fields ranging from heterogeneous catalyst carrier, membranes used for isolating bacteria in bioreactors and environment filters for hot fuel gases and diesel engine emissions [4]. In particular, porous Si_3N_4 ceramics [5,6] show superior mechanical properties such as high strength, good thermal shock, and high strain and damage tolerance. Considering the advantageous nature of the SHS technique with regard to the environmental friendliness, we successfully prepared highly porous Si_3N_4 ceramics using this technique.

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2. Experimental

Si (98.92 wt.%, 6.10 μm), Y_2O_3 (99.95 wt.%, 0.52 μm) and α-Si_3N_4 (α = 93 wt.%, 0.52 μm) as a diluent were used as starting materials for the fabrication of porous Si_3N_4 ceramics. The samples were prepared according to the composition shown in Table 1. The mixtures were ball-milled for 24 h using Si_3N_4 balls. Absolute ethanol was used as the milling media. The resultant slurry was dried to obtain an agglomerate-free powder mixture with a rotary evaporator in a drying box at 80 °C for at least 5 h to ensure that the powders were completely free of alcohol. The dried powders were then sieved to 60-mesh.

The experiments were performed in a cold isostatic pressure vessel under 5 MPa nitrogen pressure. The samples, packed in a porous graphite crucible, were buried under a layer of igniting titanium powders placed on the top of them. The ignition was carried out using a tungsten coil. The combustion wave temperature was measured with a W/Re thermocouple protected by a layer of BN, which was inserted into the center of the powders.

To examine the relative density together with the amount of closed and open pores, Archimedes' method was employed. X-ray diffractometry (XRD) analysis was conducted to examine the phases in the obtained porous silicon nitride ceramics. The morphologies of combustion products were studied by using scanning electron microscopy (SEM). The free silicon amount in the combustion products was determined by chemical analysis.

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3. Results and discussions

Table 1 shows the XRD analysis and chemical analysis results for all the combustion products. In the products of samples B and C, there is only β-Si_3N_4 phase with trace amount of free Si; while in the sample A, there are 12 wt.% free Si and β-Si_3N_4 phase. This result suggests that to fully nitride Si powders under high nitrogen pressure, as much as >20 wt.% α-Si_3N_4 diluent is needed.

The open porosity was 62–64% for all samples (Table 2). There was virtually no closed porosity in all of the samples.

Fig. 1 shows SEM photographs of the combustion products. Rod-shaped β-Si_3N_4 grains, which formed a three dimensional network structure, were observed in the three samples. The randomly connected fibrous grains in three dimensions made the synthesized products with high open porosity. Some pores and hollows can be seen clearly from the microstructure.

Table 1
Phase composition of combustion products for different starting powder (nitrogen pressure: 5 MPa)

Sample	Phase composition of starting powders (wt%)			Phase composition of combustion products (wt.%)		
	Si	α-Si_3N_4	Y_2O_3	α-Si_3N_4	β-Si_3N_4	Free Si
A	85.5	9.5	5	Trace	88	12
B	71.3	23.7	5	0	100	Trace
C	47.5	47.5	5	0	100	Trace

Table 2
Properties of the obtained silicon nitride porous ceramics

Sample	Relative density (T.D%)	Open porosity (%)	Closed porosity (%)
A	35.31	64.40	0.29
B	35.93	64.01	0.06
C	37.81	62.07	0.12

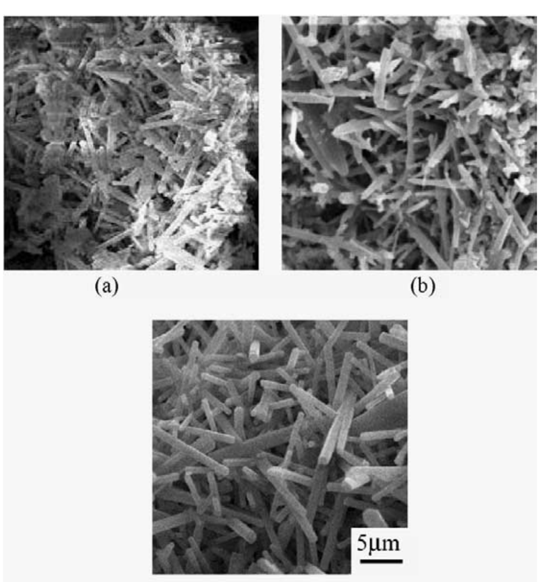

Fig. 1. SEM photographs of combustion products: (a) sample A; (b) sample B; (C) sample C.

It is well known that the Y_2O_3 acts as a sintering aid for silicon nitride to form a liquid phase and to accelerate the α to β phase transformation and grain growth via the dissolution- reprecipitaton [7–9]. In the present experiment, it is proposed that the Y_2O_3 had the same role as that in the sintering process of silicon nitride. We note that every one of the combustion wave temperatures of the three samples was over 2000 °C, which is considerably higher than both the melting point of Si (1410 °C) [10] and the eutectic point of Y_2O_3–SiO_2–Si_3N_4 system (1550 °C) [11,12]. Therefore, a Y–Si–O–N liquid could be formed that could be beneficial for the α to β phase transformation and the β-Si_3N_4 grain growth. Moreover, it is reported [13] that the liquid Si can act as suitable solvent for Si_3N_4 and assists in the α to β phase transformation. So, the combustion synthesized rod-shaped grains, which build up the network silicon nitride porous ceramics, result from the synergy role of the liquid Si and Y–Si–O–N liquid phase.

4. Conclusions

Porous silicon nitride ceramic with interconnected β-Si_3N_4 rod-shaped grains and high porosity was synthesized by using combustion synthesis method. The resultant porous silicon nitride had high porosity over 60%. By discussing the formation mechanism of rod-shaped grains, it can be concluded that rod-like grains result from the synergy role of liquid Si and Y–Si—O–N liquid phase produced by the high temperature.

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