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ScienceDirect
Progress in Natural Science: Materials International 20(2010) 81−86
Progress in Natural Science: Materials International
www.elsevier.com/locate/pnsc

Synthesis and sintering of beryllium oxide nanoparticles

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Received 23 August 2010; accepted 23 October 2010

Abstract: BeO nanoparticles were prepared by polyacrylamide gel route and the sintering properties of synthesized powder as a function of sintering temperature and time were investigated. Thermal behaviors of the polyacrylamide gel and beryllium salt were studied by thermogravimetry analysis (TGA). The calcination temperature (690°C) of the beryllium sulfate covered in polyacrylamide xerogel was almost 150 °C lower than that obtained by traditional methods. The nanocrystallites of the BeO particles were estimated by X-ray diffraction (XRD) according to Debye–Scherrer equation. Transmission electron microscopy (TEM) investigation revealed that the average particle size of the BeO nanoparticles prepared by calcining at 800 °C for 2 h, ranges from 15 nm to 25 nm. The sintering temperature was down to 1 600 °C, which was around 200 °C lower than conventional sintering temperature of the compact sample with common powders. The densification rate was fast at the first 3 h. The thermal conductivity of the sample sintered at 1 600 °C for 6 h was 186.6 W/(m·K).

Key words: BeO; nanoparticles; polyacrylamide gel; sintering; thermal conductivity

1 Introduction

Beryllium oxide (BeO) ceramics possesses high thermal conductivity, high melting point, high intensity, high insulation nature, high chemical and thermal stability, low dielectric constant, low dielectric loss and good technology applicability, so it is widely applied in vacuum electronics technology, nuclear technology, and microelectronics and photoelectron technology[1−2]. To get advanced BeO ceramics, beryllia powders with high performance are necessary. In recent years, nanoparticles with excellent physical and chemical properties have become an interesting field to researchers[3]. However, there are few reports about BeO nanoparticles.

Polyacrylamide gel route is a new improved method of synthetic powder fabrication, in which a solution of the respective cations is soaked. In this method, a steric entrapment of stoichiometric cation solution occurs in nanocavities formed inside the gel, that is, an homogeneous micro solution with cations in the desired stoichiometry. In 1989, this process was firstly described by Douy and Odier[4] for preparing three different ultrafine powders: $YBa_2Cu_3O_{7-x}$, $2SiO_2$-$3Al_2O_3$ and $LaAlO_3$. And then, more and more researchers were interested gradually in this method and many different oxide ultrafine powders or nanoparticles, metallic and oxide compounds, such as ZnO[5], Al_2O_3[6], BiO_2[7], $CaAl_{12}O_{19}$[8], $La_{0.8}Sr_{0.2}MnO_3$[8], $BaZrO_3$[8] and ultrafine powders for solid oxide fuel cells[9] (SOFCs) $Zr_{0.84}Y_{0.16}O_{1.92}$, $Ce_{0.8}Gd_{0.2}O_{1.9}$ and $La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.2}O_{2.85}$ were prepared. This polymer gel method has many advantages: 1) relatively lower calcination temperature than traditional preparation technique; 2) simple and convenient process; and 3) lower cost and time-saving. In the present work, we report the polyacrylamide gel route for the preparation of BeO nanopowders. As for sintering properties of the nanoscale BeO powder synthesized by the acrylamide polymerization technique, there is lack of reports in the literatures. So its sintering properties as a function of sintering temperature and time are also investigated.

2 Experimental

2.1 Raw materials

The characteristics of the used materials are given in Table 1. All of the reagents are AR grade, except that the distilled water produced in our laboratory is chemically pure (CP) grade.

2.2 Experimental procedure and sample preparation

Figure 1 shows the flow chart of the preparation of BeO powders by the polyacrylamide gel route. First,

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Table 1 Characteristics of raw materials

Raw materials	Function	Molecular formula	Supplier
Beryllium sulfate tetrahydrate	Initial salt	$BeSO_4{\cdot}4H_2O$	Shuikoushan Nonferrous Metals Co. Ltd., China
Acrylamide (AM)	Monofunctional monomer	$C_2H_3CONH_2$	Shanghai Chemical Reagent Co., Ltd., China
N,N'-methylene bis acrylamide (MBAM)	Difunctional monomer	$(C_2H_3CONH_2)_2CH_2$	Shanghai Chemical Reagent Co., Ltd., China
Ammonium persulfate (APS)	Initiator	$(NH_4)_2S_2O_8$	Changsha Organic Chemical Reagent Co., Ltd., China
Water	Solvent	H_2O	self-produced

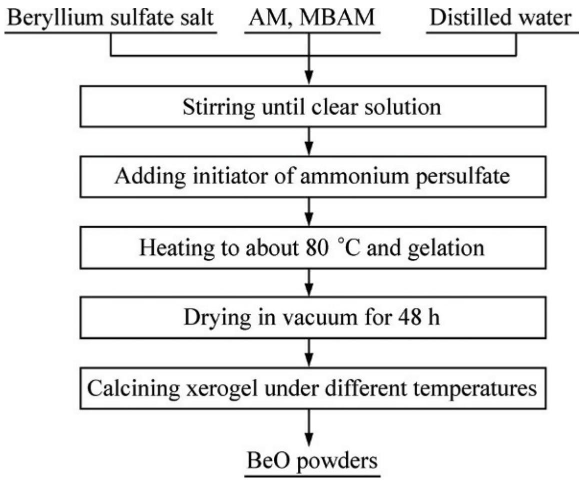

Fig.1 Flow chart of preparation of BeO powders by polyacrylamide gel route

$BeSO_4{\cdot}4H_2O$ was dissolved in the distilled water and the concentration of beryllium sulfate aqueous solution is 1.5 mol/L. Acrylamide (AM) and N,N'-methylene bis acrylamide (MBAM) monomers with a molar ratio of 20 as the polymerization agents were added in the prepared solution. The mixture was stirred up to transparent solution. 0.5 mL ammonium persulfate (APS) solution (ASP: 10 %, mass fraction) as initiator was added, and then the temperature of the solution was increased slowly to 60 °C in water bath. The free-radical crosslinking copolymerization of AM and MBAM was initiated by the initiator APS and the heat activation effect. The mixture was turned to transparent hydrogel gradually. In order to make sure that the organic monomers reacted absolutely, the condition was held for 1 h. The gel was dried at 80 °C for 48 h in a vacuum drier.

The xerogel thus formed was homogenized in a ceramic mortar and submitted to subsequent thermal treatment. The xerogel was heated in a laboratory furnace and calcined in air at different temperatures (700−1 000 °C) for 2 h to obtain white powder. The thermal treatment was applied at a heating rate of 5 °C/min up to the desired holding temperature, followed by the same cooling rate.

The synthesized powders were pressed uni-axially into cylindrical samples under 200 MPa for 1 min. The compacts were then sintered in an air box furnace at different temperatures (800−1 680 °C) for 2 h and at 1 400 and 1 600 °C for different time (2−10 h) (heating rate 10 °C /min), respectively.

2.3 Characterization techniques

To define the precise calcinations temperature, thermogravimetric analysis for the xerogel and beryllium sulfate salts using TGA (NETZSCH STA 449C) was conducted at a heating rate of 5 °C/min from room temperature to 1 000 °C, and an air flow of 30 cm^3/min. XRD data collection on the synthesized powders was carried out for phase identification and crystallite size determination using a Japan Rigaku D/Max2550VB+ diffractometer with Cu $K_{\alpha 1}$(λ=1.54Å) radiation in the range of 2θ=30°−90° at a 2θ step of 0.08° for 0.2 s per point. The crystallite size, size distribution, structure and morphology of the beryllia powders were obtained by transmission electron microscopy (Philips CM-300).

The bulk density of BeO sintered body was measured based on Archimedes' law and compared with the theoretical density to obtain the relative density. The thermal conductivity (TC) was measured by the laser flash method with thermal diffusivity analysis. The tested temperature was 25 °C. The specimen size was d 10 mm×4 mm, and both sides of the specimen were polished. The scanning electron microscopy (SEM, Sirion−2000, Philips) was used to observe the fracture surfaces of the specimens, which were carbon coated to achieve an electrically conductive surface so as to avoid charging.

3 Results and discussion

3.1 Thermal decomposition of precursors

TG curves of $BeSO_4{\cdot}4H_2O$ and sulfate based xerogel are presented in Fig.2. As can be seen from curve (a) in Fig.2, the decomposition of the beryllium salt was