



# Silicon nitride foams from emulsions sintered by rapid intense thermal radiation

Elisângela Guzi de Moraes<sup>a,\*</sup>, Duan Li<sup>b,c</sup>, Paolo Colombo<sup>a,d</sup>, Zhijian Shen<sup>b,c</sup>

<sup>a</sup> Dipartimento di Ingegneria Industriale, University of Padova, Via Marzolo 9, 35131 Padova, Italy

<sup>b</sup> Department of Materials and Environmental Chemistry, Arrhenius Laboratory, Stockholm University, S-106 91 Stockholm, Sweden

<sup>c</sup> Berzelii Center EXSELENT on Porous Materials, Stockholm University, S-106 91 Stockholm, Sweden

<sup>d</sup> Department of Materials Science and Engineering, Pennsylvania State University, University Park, PA 16801, United States

Available online 23 February 2015

## Abstract

Open cell Si<sub>3</sub>N<sub>4</sub> foams were fabricated by an emulsification process and subsequent sintering. Emulsification took place in an aqueous environment by using octane as alkane phase and a surfactant as stabilizer. Effects of different octane concentrations (50 vol% and 70 vol%) and stirring velocities (700 rpm and 1000 rpm) on porosity and pore size distribution were investigated. The shaped foams were pressureless sintered at 1600 °C inside a modified SPS set-up. Si<sub>3</sub>N<sub>4</sub> foams decorated with SiC nanowires and with an average cell size of 8–41 μm and total porosity of 80–86 vol% were obtained. The compressive strength ranged from 2.0 MPa up to 9.9 MPa. Both the foaming and sintering processes contributed to the development of different microstructures.

© 2015 Elsevier Ltd. All rights reserved.

**Keywords:** Foams; Silicon nitride; Emulsification; Spark plasma sintering; Sintering by intense thermal radiation

## 1. Introduction

Ceramics with a cellular structure and containing designed interconnected porosity above 60 vol% find applications particularly where the transport of fluids is required, e.g. molten metal and exhaust particulate filters at high temperature, gas-burner systems, catalyst support and in energy-related industries [1–6]. These porous components exhibit a special combination of properties such as light weight, high temperature stability and permeability to fluids that cannot normally be reached by conventional dense materials or materials with a non-designed porosity [7].

Silicon nitride (Si<sub>3</sub>N<sub>4</sub>) is one of the most widely used ceramics in many engineering applications due to its outstanding thermo-mechanical properties, such as flexural strength and Young's modulus around 900 MPa and 310 GPa, respectively, fracture toughness between 3 and 12 MPa m<sup>1/2</sup> [8], and strain-to-failure around 3 × 10<sup>-3</sup> [9,10]. Its excellent thermal shock

resistance can be attributed to the combination of a low thermal expansion coefficient, medium elastic constant, and moderate thermal conductivity [8,11,12]. However, the high cost of production (sintering is generally assisted by pressure), due to the highly covalent bonding between silicon and nitrogen atoms and very slow solid-state diffusion [13], limits significantly the use of silicon nitride-based ceramics to specialized, high value applications. The additions of sintering additives, which are usually metal oxides that form a low-melting-point eutectic liquid with the oxide surface layers of the silicon nitride powder, improve sintering activity considerably and promote high densities without the use of pressure during sintering [14,15]. The development of more economical processes for the production of silicon nitride component in structural applications, with proper process control and uniformity in the properties of the final product, is a challenge from a technological point of view [16].

Various processing methods for the production of highly porous Si<sub>3</sub>N<sub>4</sub>, such as partial sintering [17,18], reaction sintering [19,20], the use of sacrificial templates using starch consolidation [21], direct foaming [22], gelcasting [23] and preceramic polymers [24], have been proposed. Recently, emulsions have been used as efficient intermediates in the production of porous

\* Corresponding author. Tel.: +39 049 8275023; fax: +39 049 8275505.

E-mail address: [elisangela.guzidemoraes@studenti.unipd.it](mailto:elisangela.guzidemoraes@studenti.unipd.it) (E.G. de Moraes).

materials via direct foaming processes [2,25,26]. They consist of two immiscible fluids, one being dispersed in the other, and are thermodynamically unstable due to their large oil–water interfacial area and thus high overall free energy. Therefore, surface active agents (e.g. surfactants or proteins) are used to reduce the free energy of the system [27,28]. The objective of the present work is to develop cellular silicon nitride ceramics with tailored porosity by using an emulsification process that enables the fabrication of components possessing a well interconnected (open) cell morphology and small, uniform cell sizes [29] and improved mechanical properties by a novel processing approach (rapid pressureless sintering by intense thermal radiation).

## 2. Experimental procedure

Silicon nitride powder (grain size  $d_{50} = 0.6 \mu\text{m}$ , purity  $\sim 96 \text{ wt}\%$ , main impurity Fe, oxygen content  $7.4 \pm 0.08 \text{ wt}\%$ ,  $\alpha$  phase  $\sim 91.5\%$ , Yantai Tomley Hi-tech Ind.&Tra. Co., Ltd, Yantai, Shandong, China) was used as raw material in this study. The equilibrium  $\text{pH}_{\text{iep}} 8.27 \pm 0.01$  of the powder was measured after 24 h aging in water, following the procedure reported in Ref. [30]. High purity  $\text{Y}_2\text{O}_3$  ( $d_{50} = 50 \text{ nm}$ , Inframat Advanced Materials L.L.C., Manchester, NH, USA), and  $\text{MgO}$  ( $d_{50} = 4.6 \mu\text{m}$ , Bitossi Ceramiche S.R.L., Montelupo Fiorentino, Firenze, Italy), were used as sintering additives; the label 5YM was used to denote specimens containing 5 wt%  $\text{Y}_2\text{O}_3$  and 5 wt%  $\text{MgO}$ , and the amount of sintering aids was inferred from the literature [13]. The powder mixture was wet-milled in ethanol for 4 h, using silicon nitride cylinders and planetary velocity of 300 rpm. The slurry was dried and sieved through a 300  $\mu\text{m}$  screen. The powder mixture was treated in air at 600 °C for 2 h, in order to form a silica layer on the particle surfaces to improve their aqueous dispersibility and, consequently decrease the viscosity of the suspension [31]. 1 wt% polyacrylic acid (PAA, Sigma–Aldrich Sweden AB, Stockholm, Sweden) was used as dispersing agent to stabilize the suspensions. 0.22 vol% polysorbate (Tween 80, VWR International, Bedfordshire, UK) was used as nonionic surfactant and consequently foam stabilizer. Water-based  $\text{Si}_3\text{N}_4$  slurries with 35 vol% of solids and containing PAA were prepared by ball milling for 2 h at 200 rpm. Afterwards, the emulsification process took place by addition of 50–70 vol% alkane phase (octane,  $\text{C}_8\text{H}_{18}$ , Sigma–Aldrich Sweden AB, Stockholm, Sweden) and subsequently the suspension was stirred at 700 or 1000 rpm for 3 min. Then, the emulsions were poured in a Teflon mold and dried at ambient air for approximately 24 h.

Rapid pressureless sintering was conducted in a modified SPS set-up (Dr. Sinter 2050, Sumitomo Coal Mining Co., Tokyo, Japan) under vacuum. The green foams were loaded in a covered cylindrical graphite crucible with an inner diameter of 50 mm and outer diameter of 70 mm. The samples were protected by a  $\text{Si}_3\text{N}_4$  powder bed and isolated by graphite felts. Two sintering regimes, namely PLSPS-1 and PLSPS-2, were set as follows: the temperature was automatically raised to 600 °C over a period of 5 min, and from there onwards it was monitored and regulated by an optical pyrometer focused on the wall centrally inside

the crucible through a hole of  $\sim 5 \text{ mm}$  in diameter. For PLSPS-1, in the first step the samples were heated to 1500 °C with a heating rate of 50 °C  $\text{min}^{-1}$  and maintained for 10 min, then heated to 1600 °C at 50 °C  $\text{min}^{-1}$  and held for 3 min. Same steps and temperatures were used for PLSPS-2, differing only for the heating rate and dwell time, which were instead 100 °C  $\text{min}^{-1}$  and 5 min for the first step, respectively. More details about this rapid sintering procedure can be found elsewhere (see Ref. [32]).

The bulk density and the total porosity were calculated from the weight-to-volume ratio of the samples, while the open porosity was determined using a mercury intrusion porosimetry (Micromeritics AutoPore III 9410, Norcross, GA, USA). The surface tension and the contact angle of mercury were set to 0.485  $\text{N m}^{-1}$  and 130°, respectively. The crystalline phases were determined on powdered samples by X-ray diffraction (XRD, X'pert PRO MPD diffractometer, PANalytical, Almelo, Netherlands), using  $\text{CuK}\alpha$  radiation  $\lambda = 1.542 \text{ \AA}$ , at 40 kV and 40 mA. The  $2\theta$  range was varied from 10° to 90° with a step size of 0.05° and a step time of 2 s. The data were analyzed utilizing the ICSD database, and the weight fractions of the  $\alpha$ - and  $\beta$ - $\text{Si}_3\text{N}_4$  crystalline phases were evaluated by the method described in Ref. [33]. The microstructure of the  $\text{Si}_3\text{N}_4$  based foams was characterized by a field emission scanning electron microscope (FE-SEM, JSM-7000F, JEOL, Tokyo, Japan) and a Schottky-type field emission transmission electron microscope (TEM, JEM-2100F, JEOL, Tokyo, Japan) operated at 200 kV, equipped with an energy-dispersive X-ray spectroscopy (EDX) detector. Electron diffraction (ED) data were collected using a  $\text{LaB}_6$  based transmission electron microscope (TEM, JEM-2100, JEOL Ltd, Tokyo, Japan) at 200 kV. For TEM study, the sample was crushed into powder and dispersed in ethanol. The average cell and cell window sizes were obtained from SEM images using the linear intercept method according to ASTM E112-12 (diagonal opposite directions),

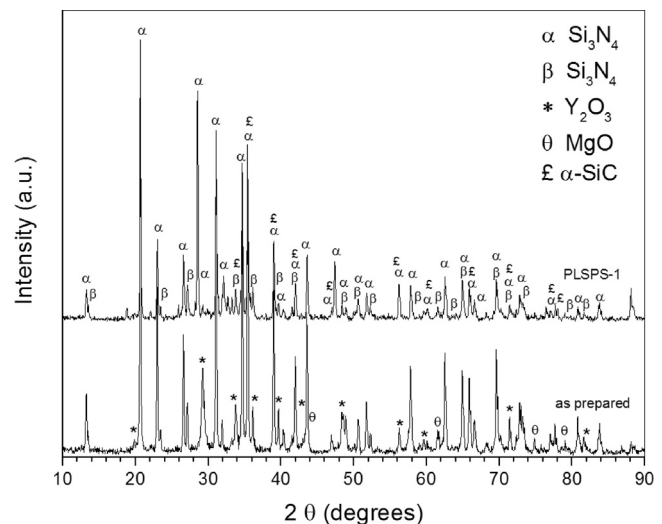


Fig. 1. XRD data for the as-prepared and as-sintered  $\text{Si}_3\text{N}_4$  foams (PLSPS-1 sintering regime) containing 5 wt%  $\text{Y}_2\text{O}_3$  and 5 wt%  $\text{MgO}$  (ICSD  $\alpha$ - $\text{Si}_3\text{N}_4$  #041-0360,  $\beta$ - $\text{Si}_3\text{N}_4$  #033-1160,  $\alpha$ - $\text{SiC}$  #049-1428).

Download English Version:

<https://daneshyari.com/en/article/1473703>

Download Persian Version:

<https://daneshyari.com/article/1473703>

[Daneshyari.com](https://daneshyari.com)