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Open-cell reaction bonded silicon nitride foams: Fabrication and characterization

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Abstract

In this study, a newly designed fabrication procedure was utilized to produce silicon nitride foams. The main goal of the present study was to obtain Si_3N_4 foams with high levels of porosity and pore interconnectivity via an economical fabrication procedure including sacrificial template technique, gel-casting and reaction bonding processes. The fabrication procedure was studied and optimized in terms of suspension preparation and rheology, gel-casting parameters, and reaction bonding conditions. The produced foams have a precisely controlled level of porosity which can be varied up to 87 vol%. BET analysis showed that the surface area of the foam is of the order of 2.01 m^2/g. The pore interconnectivity of the foam was investigated via polyester resin infiltration. Based on XRD and SEM analysis, the dominant nitriding reactions are the gas-phase reactions which lead to α-Si_3N_4 in the form of whiskers.

Keywords: Silicon nitride; Reaction bonding; Foam; Gel-casting; Pore interconnectivity

1. Introduction

Traditionally, ceramic materials have been made as dense as possible to reduce the possibility of sudden failure and fracture of this brittle group of materials. Recently, cellular ceramics have been found to be very interesting due to their unique and remarkable characteristics.[1,2] Although these materials exhibit special features and functionalities, they are not being used to their full potential. Designing new ways to fabricate homogenous ceramic foams with tailored properties including pore structure, pore size, and pore interconnectivity will make them suitable for many new and high-tech applications such as bioimplants, diesel particulate filters (DPF), separation membranes, and for the fabrication of interpenetrating composites.[2–5] There are two properties in the foam structure which have not received sufficient attention. The first one is pore interconnectivity which is necessary in all the above mentioned applications where fluid transport is necessary. The second property is the porosity level of the foam which affects many foam characteristics such as surface area and permeability.

Among all the ceramic materials, Si_3N_4 has outstanding properties and potential including high strength and toughness, high temperature resistance, high thermo-chemical corrosion resistance, good wear resistance, and good thermal shock resistance which make this ceramic material a great candidate for many engineering applications.[6] Several methods have been used to fabricate Si_3N_4 foams including partial sintering of a Si_3N_4 compact,[7] impregnation of polyurethane sponge,[8,9] freeze drying,[10,11] sacrificial template technique,[6,12] and direct foaming method[13] but all the produced foams suffer from low levels of porosity and also lack pore interconnectivity. There are also some other fabrication issues and difficulties in working with silicon nitride powders including the high cost of the starting material, the high sintering temperatures (around 1900 °C), the large amounts (up to 10 wt%) of sintering additives required due to a high degree of covalency in the Si–N bond, and the large linear shrinkage (15–20%) which makes it essential to perform time-consuming and costly post-sintering machining steps to get the required sample dimensions.[14] In terms of wet processing techniques which are capable of producing homogeneous ceramics, working with Si_3N_4 suspensions is also undesirable due to the rheological problems of the suspensions including inconsistency in Si_3N_4 slurries required for the industrial production[15,16] and high shear thickening behavior of these slurries.[17]

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Table 1
Comparing the advantages and disadvantages of RBSN and SSN.

Reaction bonded Si_3N_4		Sintered Si_3N_4	
Advantages	Disadvantages	Advantages	Disadvantages
Low cost Si powder [25]	Very long procedure for dense and large bodies [26]	High mechanical strength [27]	15–20% linear shrinkage during sintering [14]
Almost no dimensional changes during nitriding; increasing the process reliability and reduces post machining [24]	Residual Si in the dense nitrided sample	Good thermal shock resistance [6]	Very high sintering temperature
Low nitriding temperature	10–20% residual porosity in the dense body [24]	High toughness	15–20% linear shrinkage during sintering; large dimensional changes and costly post-sintering machining steps [14]
Low production cost [25]			Large amounts of sintering additives

In this study a new fabrication procedure was designed in order to produce a Si_3N_4 foam with an open-cell structure and a controlled level of porosity but without the issues of working with Si_3N_4 powder. To the best of the authors' knowledge there is no classified study on reaction bonded silicon nitride (RBSN) foams per se and there are also no reports of the fabricated Si_3N_4 foam with open-cell structures and pore interconnectivity. There are only a few investigations in the area of RBSN foams in which the utilization of the dry-processing techniques such as uniaxial pressing or extrusion has led to inhomogeneous closed-cell structures with low levels of porosity.[18–20] The fabrication procedure designed in this study relies on the combination of the sacrificial template technique, gel-casting, and reaction bonding processes which leads to homogeneous RBSN foams with interconnected porosity. The sacrificial template method was selected in this study as the foam fabrication technique since it is capable of tailoring foam properties including pore size, pore morphology, pore size distribution, total porosity, and pore interconnectivity.[1] The fabrication procedure was also based on wet colloidal processing to improve homogeneity of the foam and reduce the microstructural defects which typically form in dry processing techniques due to poor particle packing and distribution. The combination of gel-casting as a near-net shape, in situ, consolidation technique and the reaction bonding process in this study offers advantages including a short processing time, high wet and dry strengths, possibility of producing high-quality homogeneous bodies with complex shapes, the possibility of using different mold materials, machinability, higher reliability, less flaws and defects in the final products, the capability of producing large samples and an easy and economical fabrication procedure.[21–24] Tables 1 and 2 compare the advantages and disadvantages of RBSN, sintered silicon nitride (SSN), and gel-casting technique. The tables show that the newly designed foam fabrication procedure eliminates all the fabrication disadvantages of SSN and RBSN ceramics while combining the advantages of RBSN and gel-casting techniques.

The main goal of this investigation was to fabricate an RBSN foam with an open-cell structure and a controlled level of porosity. Silicon-PMMA suspensions were made and characterized in terms of stability and rheology to optimize the suspension behavior and consequently the foam properties. The gel-casting step was also optimized in terms of the monomer content, monomer to cross-linker ratio, and polymer pyrolysis. Furthermore, this investigation examines the nitriding conditions and the dominating nitriding mechanisms. Finally, the RBSN foams were also studied in terms of foam properties including the porosity, pore interconnectivity, and microstructure.

2. Materials and methods

2.1. Foam fabrication

Porous RBSN foams have been fabricated from Si powder (ABCR, 99.995%, −8 μm) and PMMA beads (Microbeads, 10–40 μm) as the pore former via the gel-casting technique. Acrylamide (AM, $C_2H_3CONH_2$, Sigma-Aldrich) was used as the monomer and N,N′-methylenbisacrylamide (MBAM, $(C_2H_3CONH_2)_2CH_2$ Sigma-Aldrich) was utilized as the crosslinker. Three different dispersants were used in this study including Darvan® 821A (R.T. Vanderbilt Minerals LLC, poly(acrylic acid) ammonium salt, PAA-NH_4), DS001 (Polymer Innovations Inc.), and Dolapix PC 75 (Zschimmer & Schwarz GmbH & Co KG). Poly(acrylamide) (PAM, Acros Organics, M.W. = 5,000,000–6,000,000) and DF002 (Polymer Innovations Inc.) were the binder and the antifoaming agent, respectively. Ammonium persulfate (APS, $(NH_4)_2S_2O_8$, Sigma-Aldrich) and N,N,N′,N′-tetramethylethylenediamine (TEMED,

Table 2
Advantages and disadvantages of the gel-casting technique.

Gel-casting	
Advantages	Disadvantages
Near net-shape fabrication technique even for complex shapes [23]	Reactive components [23]
Highly homogeneous and uniform bodies with high green strengths [15]	Irritating monomers with the capacity to sensitize exposed workers [23]
High machinability	
Low levels of organic additives [28]	