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Microstructure and properties of lightweight fibrous porous mullite ceramics prepared by vacuum squeeze moulding technique

Rubing Zhang [a,*], Changshou Ye [a], Xiaobo Hou [a], Shaohua Li [a], Baolin Wang [b]

[a] School of Civil Engineering, Beijing Jiaotong University, 100044 Beijing, China
[b] Institute for Infrastructure Engineering, Western Sydney University, Penrith, NSW 2751, Australia

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ABSTRACT

A novel fibrous porous mullite network with a quasi-layered microstructure was produced by a simple vacuum squeeze moulding technique. The effects of organic binder content, inorganic binder and adsorbent on the microstructure and the room-temperature thermal and mechanical properties of fibrous porous mullite ceramics were systematically investigated. An anisotropy microstructure without agglomeration and layering was achieved. The fibrous porous mullite ceramics reported in this study exhibited low density (0.40 g/cm^3), low thermal conductivity (~0.095 W/(m K)), and high compressive strength (~2.1 MPa in the *x*/*y* direction). This study reports an optimal processing method for the production of fibrous porous ceramics, which have the potential for use as high-temperature thermal insulation material.

1. Introduction

Mullite ($3Al_2O_3$–$2SiO_2$) is a material of particular interest because of its unique set of properties, such as high chemical and thermal stability, low thermal conductivity, low thermal expansion coefficient, and high mechanical strength, which makes it a promising candidate for use in structural ceramics at high temperatures under oxidizing and corrosive environment [1,2]. Over the past decade, mullite fibers have been used as reinforcement phase for enhancing the strength of oxide fiber-reinforced ceramic matrix composites, which combine the advantages of mullite monolithic ceramics and fiber [3–6]. However, because of the relatively high density and thermal conductivity both mullite ceramics and mullite fiber reinforced oxide ceramic matrix composite have limited applications in the field of lightweight high-temperature thermal insulation.

Porous ceramics have potential applications as structural and functional components by tailored microstructures due to their unique properties, such as light weight, large surface area, low thermal conductivity, good strain and damage tolerances, and good thermal shock resistance [7–11]. The porous mullite ceramics can be obtained from the commonly used fabrication methods such as gel-casting [12], direct consolidation [13], direct foaming [14], and TBA-based freeze casting [15]. Zhang et al. [12] fabricated porous mullite ceramics with a relatively low density (0.84–1.64 g/cm^3), low thermal conductivity (0.16–0.22 W/m K), and high compressive strength (6.21–14.70 MPa) by gel-casting process using fly ash cenospheres as a pore-forming agent. Porous mullite materials with porosity ranging from ~35 to 57 vol% has been prepared by a direct consolidation method based on the swelling properties of starch in water. The thermal conductivity of materials highly depends upon porosity, for instance, with porosity ⩾45%, thermal conductivity is very low at room temperature (1.11–2.41 W/m K) and almost constant with temperature [13]. Zhang et al. [14] reported porous mullite ceramics with low thermal conductivity (as low as 0.09 W/m K), high porosity (73–86 vol%) and compressive strength (1–22 MPa) by foaming and starch consolidation, which contained a multimodal microstructure with large spherical pores and small pores on internal walls. Wang et al. [15] prepared tubular porous mullite supports with macroporous structures (2.1–7.2 μm), high porosity (56.9–76.8%), and high bending strength (23.3–48.8 MPa) by TBA-based freezing casting method. However, these porous mullite ceramics where powder was used as raw material, exhibit a brittle fracture; hence, they cannot be suitable for high temperature thermal insulation in aerospace vehicle applications.

Recently, porous fibrous ceramics with bird-nest-type structures present a dissipative damage tolerant behavior, ultra-low density and thermal conductivity; which have attracted attention of materials scientists and aerospace engineers [16,17]. The ultra-low thermal conductivity and high tolerance to damage can be

* Corresponding author.
E-mail addresses: zrb86411680@126.com, rbzhang@bjtu.edu.cn (R. Zhang).

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attributed to the fibrous structure of the material as fibers are only bonded at crossing points by binder. Many efforts have been made to develop fibrous porous ceramics by vacuum moulding method. It is believed to be ideal processes to fabricate porous fibrous ceramics with a 3D skeleton structure because it can realize the near-net shape forming of complex shape by low cost and simple equipment [18]. Dong et al. [16] fabricated porous fibrous mullite ceramics with high porosity (74.2–78.3%), low thermal conductivity (0.231–0.248 W/m K), relatively high compressive strength (1.3–3.2 MPa) and high rebound-resilience (90–98%) by vacuum moulding method. In their work, the mixture of ($SiO_2+B_2O_3$) sols melt into continuous phase, which consequently act as a high-temperature binder at the crossing points of the mullite fibers. A silica binder was chosen to hold the zirconia fibers together by Sun et al. [17]. The formed fibrous zirconia ceramics prepared by vacuum moulding had a low linear shrinkage at high temperatures (less than 2%), ultra-high porosities (72–89%), ultra-low thermal conductivity (0.056–0.16 W/(m K)), and relatively high compressive strength (0.6–13.3 MPa). In this work, the route to fabricate fibrous porous mullite ceramics has been improved using vacuum squeeze moulding instead of vacuum moulding. The difference between them is whether or not the wet fiber network is pressed during vacuum moulding. Compared to porous fibrous ceramics with a 3D skeleton structure prepared by vacuum moulding, novel quasi-layered structured fiber skeletons prepared by vacuum squeeze moulding have higher strength.

Inorganic binder not only affects the sintering temperature of porous fibrous ceramics but also modifies the microstructure of fiber skeletons, which in turn affects the high temperature stability of porous ceramics. The aim of this work is to thoroughly evaluate the effect of organic and inorganic binder on the microstructure and mechanical properties of fibrous porous mullite ceramics. The inorganic binder selected for this study is SiC and B_4C. The advantage is that in an oxidation environment, B_4C allow the transient liquid phase to sinter at low temperature around 600 °C or lower. Moreover, SiC can offer the compounds of higher melting points as the liquid B_2O_3 was eliminated, and it avoids any possible adverse effect of the additives on the mechanical properties at high temperatures of the final products. In this work, the effects of processing parameters, including binder content and adsorbent, on the microstructure of the resulting ceramic and the room-temperature thermal and mechanical properties were systematically investigated. In addition, comparative studies have been performed to investigate synthesis mechanism and microstructural development. This paper may provide an insight on the synthesis of fibrous porous ceramics fiber skeletons with ultra-low thermal conductivity and high strength for use as high-temperature thermal insulation material.

2. Experimental procedures

Mullite fibers used as the skeleton structure of fibrous porous mullite ceramics were purchased from Zhejiang Hongda Crystal Fiber Co., Ltd., China. The diameter and length of the used mullite fibers was ~6 μm and ~500 μm, respectively. The fabrication flow chart of the mullite fibrous ceramics is shown in Fig. 1. As shown in Fig. 1a, slurries were prepared by mixing distilled water with mullite fibers, 5 wt% polyethyleneimine (PEI) dispersant, 0.5–2.0 wt% polyacrylamide (PAM) adsorbent (Sigma-Aldrich Trading Co., Ltd., Shanghai, China), equivalent SiC and B_4C inorganic binder (5–20 wt% of the fibers), and 0–15 wt% starch organic binder. Suspensions with different PAM, organic and inorganic binder contents were produced to explore their effects on the microstructure and properties of fibrous porous mullite ceramics. Then the well-stirred slurry was poured into a buchner funnel and filtrated by a vacuum pump (Fig. 1). During the filtration, the water of the wet fiber network was simultaneously extruded out by the squeeze head. In order to obtain a quasi-layered structure, an initial pressure of 160 kPa was applied on the head. Finally, the wet mullite fiber network was dried in an oven at 80–100 °C for 24–28 h, and the dried felt was sintered at 1400 °C for 1 h, with a heating rate of 3 °C/min. After calcination, the organic binder burned out and the inorganic binder bonded the fibers at the crossing points (Fig. 1).

Open porosities and densities of the sintered samples were determined by the water-immersion technique using the Archimedes method. The room temperature thermal conductivity of the samples in the z direction was measured by the guarded heat flow test method (DRE-III, Xiangtan Xiangyi Instrument Co., Ltd., Xiangtan, China). In accordance with GB/T 8489-2006 standard [19], the room-temperature compressive strength of the cylindrical samples with Φ15 mm × 20 mm was measured by a testing machine (Zwick Z050, Zwick, Ulm, Germany) with a crosshead speed of 1.0 mm/min. The samples were machined with the compressive surface perpendicular to the pressing direction. More than five samples of each measurement were selected to obtain the average value. Microstructure of the sintered samples was observed by scanning electron microscope (FEI Quanta 200, FEI Company, Hillsboro, USA) with the acc. voltage of 10 kV, the current of 40 μA and the working distance of 10 cm.

3. Results and discussion

The chemical composition and the content of the binder significantly affect the mechanical properties of fibrous porous ceramics. In this study, starch and SiC–B_4C powder mixture were used as organic and inorganic binder, respectively, to fix the fibers

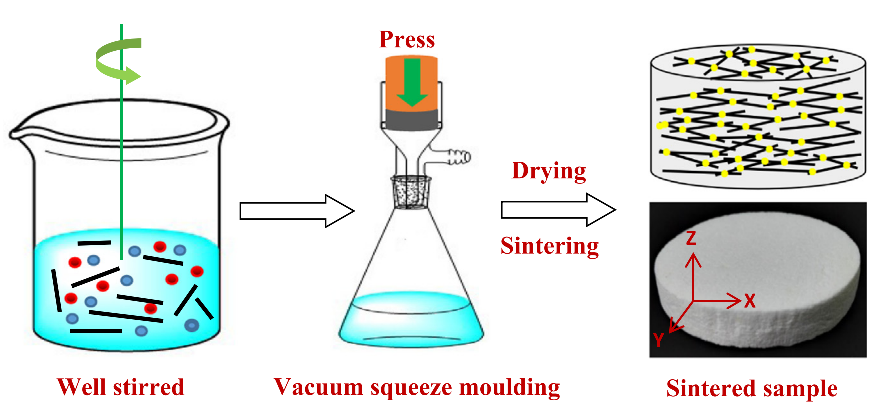

Fig. 1. Fabrication flow chart of fibrous porous mullite ceramics.