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# Synthesis of monolithic alumina-silica hollow microspheres and their heat-shielding performance for adiabatic materials

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ABSTRACT

$Al_2O_3$ ceramic foams-based composites were firstly synthesized to be used as the thermal insulation material which has excellent mechanical properties of the substrate material and better thermal properties of hollow microspheres. In this research, by doping TEOS, the monolithic hollow microspheres were prepared via a novel and effective synthesis route using propylene oxide as the gelation initiator to induce the gelation of aluminum chloride hexahydrate solution. The influence of TEOS on the morphology and high-temperature stability of the monolithic hollow microspheres was clarified in detail. Based on the optimized additive amount of TEOS, $Al_2O_3$ ceramic foams were introduced as the substrate material of alumina-silica hollow microspheres to fabricate the final $Al_2O_3$ ceramic foams-based composites. Benefited from this special structure, the $Al_2O_3$ ceramic foams-based composites displayed excellent mechanical properties and thermal properties. The samples changed less in appearance and did not show significant shrinkage after heat-treatment at 1200 °C. The density, bending strength and thermal conductivity of the $Al_2O_3$ ceramic foams-based composite were 0.32 g/cm$^3$, 1.8 MPa and 0.12 W/m K, respectively.

## 1. Introduction

The fabrication of hollow microsphere has received considerable attention because of their well-defined structure, large specific surface area and low density. These characteristics endow this kind of material various potential applications, such as catalyst support [1], controlled release of drugs [2], adsorption [3], electromagnetic [4,5], as well as in lightweight adiabatic materials [6].

It is well known that raw materials in alumina and alumina-silica system are widely used in refractory materials which depend on their characteristics such as high melting point and thermal stability. However, the synthesis of well-defined alumina and alumina-silica hollow microspheres used as lightweight adiabatic materials has rarely been studied. Therefore, it is desirable to explore feasible and easily repeatable methods for the synthesis of alumina and alumina-silica hollow microspheres which can be used as lightweight adiabatic materials.

In addition, $Al_2O_3$ ceramic foams have attracted intense interest associated mainly with their specific structure and properties, such as unique three-dimensional skeleton structure, high surface area, low density, high thermal stability and good resistance to chemical attack [7,8].

In the previous work [9], we have developed a new sol-gel synthetic route accompanied with chemically induced self-transformation to prepare monodispersed and uniform γ-AlOOH hollow microspheres. Compared with the traditional sol-gel process which requires a long time for gel process and complex external environment, this new sol-gel process is simple and timesaving. During the approach, the gel could be obtained in several seconds and the hollow microspheres were fabricated after a chemically induced self-transformation process at 60 °C. The advantages of this straightforward method for the preparation of hollow microspheres is efficient, environmentally friendly and ease manipulation. Nevertheless, the hollow microspheres prepared by the method were mainly monodispersed rather than monolithic, which limited their applications in lightweight adiabatic materials, adsorption and sensors. As a result, it is quite necessary to explore feasible and easily repeatable methods for the synthesis of monolithic hollow microspheres with certain mechanical strength.

Herein, a novel method was used to prepare monolithic hollow microspheres by doping TEOS. Propylene oxide was used as the gelation initiator to induce the gelation of aluminum chloride hexahydrate solution. With the doping of TEOS, the monolithic hollow microspheres have certain mechanical strength and high-temperature stability. In order to improve the mechanical strength as load-bearing materials in lightweight adiabatic materials, $Al_2O_3$ ceramic foams were introduced as the substrate material of alumina-silica hollow microspheres to

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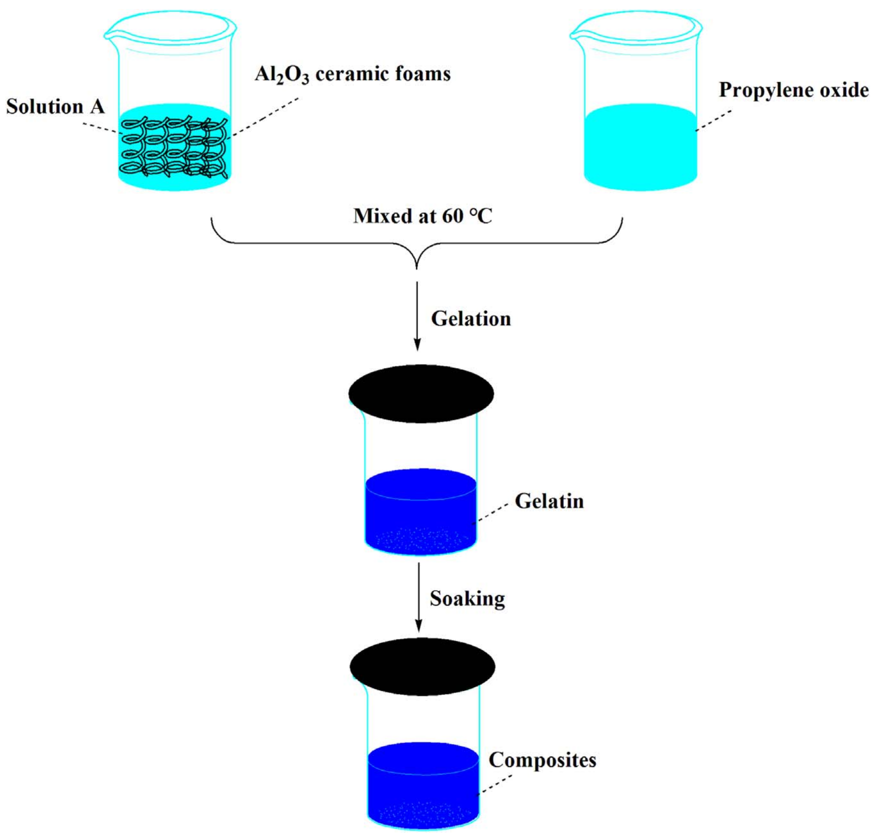


**Fig. 1.** Schematic illustration of the formation of $Al_2O_3$ ceramic foams-based composites.

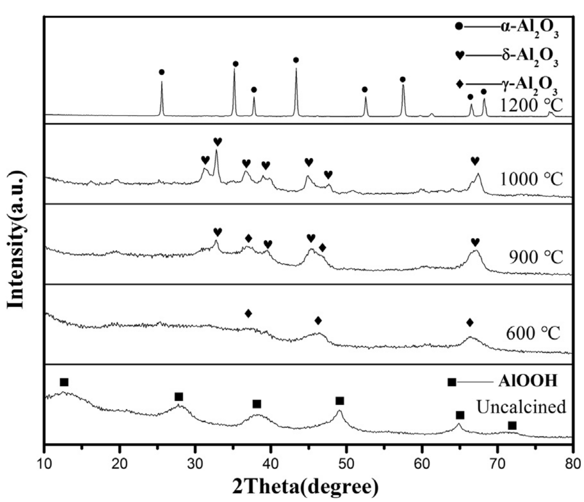


**Fig. 2.** XRD patterns of alumina hollow microspheres calcined at different temperatures.

fabricate the $Al_2O_3$ ceramic foams-based composites which have excellent mechanical properties of the substrate material and better thermal properties of hollow microspheres.

## 2. Experimental

### *2.1. Materials*

All the analytical grade chemicals including aluminum chloride hexahydrate, tetraethylorthosilicate (TEOS), propylene oxide and anhydrous ethanol (purchased from Sinopharm Chemical Reagent Co., Ltd.) were used without further purification. Distilled water was used in all experiments. $Al_2O_3$ ceramic foams were purchased from Jiangxi Runyu Technology Co., Ltd.

### *2.2. Synthesis of monolithic hollow microspheres*

Aluminum chloride hexahydrate (20 mmol, 4.83 g) and TEOS (0 g, 0.25 g, 0.5 g, 0.75 g and 1 g marked as samples a, b, c, d and e, respectively) were first dissolved in the mixture solvent of distilled water and anhydrous ethanol (4 ml of distilled water, 6 ml of anhydrous ethanol) to form a clear solution, and then the resultant homogeneous solution was sealed and maintained at 60 °C. Finally, propylene oxide (0.1 mol, 7 ml) was rapidly added into the mixture solution. Then the resultant solution was sealed and maintained at 60 °C for gelation. After the gelation reaction, the obtained samples were soaked in anhydrous ethanol at 60 °C for 48 h. In the soaking process, the anhydrous ethanol would be replaced every 12 h.

For exploring the thermal insulation property and mechanical properties of the resultant monolithic aluminum, the monolithic dried gel was heat treated at various temperatures in air with a heating rate of 1 °C/min.

### *2.3. Synthesis of $Al_2O_3$ ceramic foams-based composite*

The main synthetic schematic was shown in Fig. 1, Aluminum chloride hexahydrate (20 mmol, 4.83 g) and TEOS (0.5 g) were first dissolved in the mixture solvent of distilled water and anhydrous ethanol (4 ml of distilled water, 6 ml of anhydrous ethanol) to form a