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Fabrication and properties of thermal insulating material using hollow glass microspheres bonded by aluminum-chrome-phosphate and tetraethyl orthosilicate

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Abstract

Thermal insulation material made by hollow glass microspheres (HGM) with different content of aluminum-chrome-phosphate solution (ACP) and tetraethyl orthosilicate (TEOS) as binders was formed, dried and sintered at 250 °C, 450 °C or 650 °C for 2 h. Properties such as density, compressive strength, thermal conductivity and microstructure of the specimens were determined. It is found that TEOS improved the distribution of ACP and increased the compressive strength of the specimens. HGM bonded by appropriate amount of ACP and TEOS achieved preferable value of density, compressive strength and thermal conductivity which were significant for thermal insulation materials. The compressive strength of specimens sintered at 450 °C and 650 °C was higher than that of the specimens sintered at 250 °C.

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1. Introduction

With the growing awareness of Global Warming and fossil fuel depletion, thermal insulation materials are playing more and more important roles. Expansion vermiculite and expansion perlite are wildly used in lightweight thermal insulation materials. However, the mechanical strength of these porous materials is usually quite low because of their hollow structure. The compressive strength of expansion vermiculite materials is usually lower than 1 MPa. Influence of expanded vermiculite on physical properties and thermal conductivity of clay bricks was studied by Mucahit Sutcu [1]. It was found that although the thermal conductivity of the porous sample with 10% vermiculite addition showed 30% reduction compared to the reference brick, the compressive strength decreased 55%, and the addition of expanded vermiculite also increased the water absorption. The thermal insulation property of material with porous structure declined a lot with water being absorbed. To

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sum up, the strength and water absorption made it impossible for them to be used in many fields. Compared to them, the new developed silica aerogel can be used in fields such as automobiles, space vehicles and solar ponds because of the low thermal conductivity and good hydrophobicity. Parmenter and Milstein fabricated fiber reinforced aerogels with compressive strength as high as 1.6 MPa [2]. However, the mechanical strength restricted the application of the silica aerogel.

Hollow glass microsphere (HGM) consists of outer stiff glass and inner inert gas, which results in some unique properties, such as light weight, low thermal conductivity, and chemical stability [3]. Based on these properties, HGM has been used in many fields, especially as filler for organic system.

Researches using HGM as filler to enhance the properties of the organic matrix such as polypropylene [4–7], polyurethane [8], epoxy resin [9–11] and silicone rubber [12] have been done. The thermal insulation and mechanical properties of the composites improve a lot when HGM is added. Besides, these micro-particles do not generate stress concentration on the

interface between the fillers and the matrix owing to their smooth spherical surface. The results of HGM-filled epoxymatrix composites prepared by Yung et al. [11] showed that the density and the thermal properties of composites monotonically varied with HGM content. Compared with neat epoxy, the coefficient of thermal expansion and thermal conductivity of 51.3 vol% HGM-filled composites decreased by 54.3% and 13.3%. HGM filled silicone rubber foams prepared by Gao et al. [12] achieved high foaming extent, low density and low thermal conductivity which were significant for thermal insulation materials. Besides, the mechanical properties such as hardness and tensile strength of the HGM filled materials were much higher than that of pure silicone rubber foams.

However, few researches have been done focusing on using hollow glass microspheres to make thermal insulation materials directly. Concentrated aqueous aluminum phosphate solution is an important inorganic binder which can consolidate to metal or ceramic to form a strong amorphous aluminum phosphate network. It is widely used because phosphatebonded materials have considerable strength, temperature stability, and abrasion resistance. However, incompletely cured specimens were found to be unstable with respect to rehydration in air [13]. This strongly restricts the use of aluminum phosphate as a low temperature binder. The addition of chromium promoted the dehydrolytic condensation and the formation of an amorphous structure [14]. With Cr³⁺ added, the composites achieved excellent rehydration resistance with a low thermal conductivity by curing at the low temperature. What is more, research showed that the addition of Cr³⁺ could strengthen amorphous network, which improved the thermal shock resistance [15]. As a result, aluminum-chrome-phosphate was chosen as the binder. The low-viscosity binder solution which wetted the particles initially formed a thin coating on the particles and then concentrated in the contact regions [16]. This model can take full advantages of the strength of the binder itself while maintaining the low thermal conductivity of the HGM.

In this paper, materials with lightweight, high-strength and low thermal conductivity were fabricated by bonding HGM with ACP and TEOS. The materials were sintered at a relative low temperature. The effects of sintering temperature and the content of ACP and TEOS on density, morphology, compressive strength and thermal properties of the materials were studied. The purpose of this study is to find out suitable ways preparing thermal insulation materials by HGM.

2. Experiment

2.1. Materials

The hollow glass microspheres used in the study were made of soda-lime–borosilicate glass and supplied by 3 M Shanghai Ltd. with a trade name of S60HS. Table 1 lists the properties of microspheres. Fig. 1 shows the SEM image of HGM as received. Aluminum–chrome–phosphate solution was purchased from Tianjin with a density of 1.40–1.55 g/cm³, the mole ratio

Table 1 The physical properties of HGM.

Target crush strength (90% survival, MPa)	True density (g/cm ³)	Particle size (µm, by volume)			
		Distribution			Effective top
		10th%	50th%	90th%	size (95%)
124	0.60	11	30	50	60

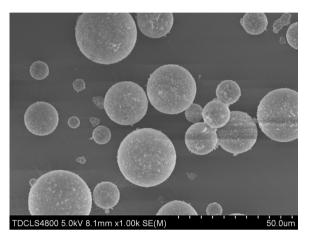


Fig. 1. 3M S60HS HGM.

Table 2
Recipe of the materials with different TEOS content.

Materials	Weight ratio (wt%)		
HGM ACP	100 30		
TEOS	0,5,10,15,20,25,30,35		

Table 3
Recipe of the materials with different content of ACP solution.

Materials	Weight ratio (wt%)		
HGM ACP	100 15,20,25,30,35,40,45,50		
TEOS	15		

of Al:Cr:P=1:1:3 ($P_2O_5 \ge 30$ wt%, Al $_2O_3 \ge 6.6$ wt% and $Cr_2O_3 \ge 11$ wt%) and pH=2. TEOS was purchased from Jiangtian chemical industry Co., Ltd. with a density of 0.929–0.936 g/cm³ and the solid content no less than 28%.

2.2. Process of experiments

The surface of the HGM was treated with 0.5 mol/L NaOH solution at 90 °C for 10 min, with the ratio of 10 g HGM to 100 mL NaOH solution. HGM, ACP and TEOS were weighed with a designated ratio (listed in Tables 2 and 3). The mixture was compression molded into specimens with a diameter of 10 mm for mechanical strength tests as well as 20 mm for thermal tests. The wet green bodies were set at room temperature for 12 h and dried at 90 °C for 1 h. Then,

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