

# Optimization of the contents of hollow glass microsphere and sodium hexametaphosphate for glass fiber vacuum insulation panel

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**Abstract.** In this paper, various additive amounts of hollow glass microspheres (HGMs) and sodium hexametaphosphate (SHMP) powders were blended with flame attenuated glass wool (FAGW) to form hybrid core materials (HCMs) through the wet method. Among them, the SHMP was dissolved in the glass fiber suspension and coated on the surface of glass fibers while the HGMs were insoluble in the glass fiber suspension and filled in the fiber-fiber pores. The average pore diameter of the FAGW/HGM HCMs was 8-11  $\mu\text{m}$  which was near the same as that of flame attenuated glass fiber mats (FAGMs, i.e., 10.5  $\mu\text{m}$ ). The tensile strength of the SHMP coated FAGMs was enhanced from 160 N/m to 370 N/m when SHMP content increased from 0 wt.% to 0.2 wt.%. By contrast, the tensile strength of the FAGW/HGM HCMs decreased from 160 N/m to 40 N/m when HGM content increased from 0 wt.% to 50 wt.%. Both the FAGW/HGM HCMs and SHMP coated FAGMs were vacuumed completely to form vacuum insulation panels (VIPs). The results showed that both the addition of SHMP and HGM led a slight increase in the thermal conductivity of the corresponding VIPs. To obtain a high-quality VIP, the optimal SHMP content and HGM content in glass fiber suspension was 0.12-0.2 wt.% and 0 wt.%.

## 1. Introduction

Thermal insulation is an indispensable part of current strategies to lower the total energy consumption worldwide. Conventional insulation materials, like expanded polystyrene, glass wool, and polyurethane, feature high thermal conductivities between 20 to 40 mW/(m·K) and poor thermal insulation performance [1]. Thick layers (tens of cm) of these conventional insulation materials are necessary to achieve a low overall thermal conductivity for their applications in structures and buildings [2].

Vacuum insulation panels (VIPs) are super insulation materials with a low thermal conductivity of < 8 mW/(m·K). In recent years, VIPs have been widely applied in buildings sectors, refrigerators and aeronautics and astronautics [3-5]. Core material is the skeleton of a VIP and greatly influences the



thermal insulating performance of the VIP. Core materials with a low thermal conductivity, low outgassing rate, and high mechanical properties are preferred in the high-quality VIPs. In Asia countries, glass fibers are commonly used as core materials due to their excellent thermal insulation properties, low density and superior chemical and thermal stability ( $> 1000\text{ }^{\circ}\text{C}$ ). The resulting VIPs are known as glass fiber VIPs, i.e., GF-VIPs [3]. Compared with fumed silica VIPs and foam based VIPs, GF-VIPs possess a lower initial thermal conductivity; and thus, they are more suitable for the applications in the household appliances like refrigerators, freezers, and vending machines.

In general, polymer binders are not added in the glassfiber core materials (GF-CMs) due to their high outgassing rate. But, the mechanical properties of GF-CMs that do not contain binders were very low because of the weak interlocking force among glass fibers. Thus, in order to get a better GF-CM with enhanced overall properties, extensive works have been carried out. Li et al. [6] proposed a novel core material structure of for VIPs, i.e. fiber pore packing with different-size powders structure, and stated that it is the development direction of future VIP core materials. Di et al. [7] used the glass fiber chopped strand and flame attenuated glass wool (FAGW) as the core materials for VIPs and found that the service life of GF-VIPs with getters was above 15 years. Kim et al. [8] studied the relation between the density of glass wool and external pressing load and found that GF-VIPs were superior when the pressing load was less than 25 kPa. Regarding enhancing the tensile strength of GF-CMs, Li et al. [9, 10] optimized the number of beating revolutions for centrifuged glass wool mat (CGM), flame attenuated glass wool mat (FAGM), and hybrid glass wool mat (HGM).

In current technology, there are two different fabricating techniques for GF-CMs: (1) wet method (like the papermaking technique); and (2) dry method (like the hot pressing technique and centrifugal-spinneret-blow technique). The essential of wet method is to form wet glass fiber mats through the removal of the solvent in glass fiber suspension by the vacuum filtration. In the current wet method, the solvent for glass fiber suspension is recycled white water (tap water with some small fibers). The addition of chemical additives in the white water is beneficial to the bonding strength among the GF and fiber dispersion in the GF suspension. However, there are few kinds of literature on this aspect. The knowledge thereof is of vital importance to obtain a high-strength GF-CM and low-thermal-conductivity GF-VIPs.

In this paper, various amounts of sodium hexametaphosphate (SHMP) and hollow glass microspheres (HGMs) was added in the white water to form wet-laid mats, i.e. the SHMP coated FAGMs and FAGW/HGM HCMs. A total of 20 pieces of the SHMP coated FAGMs and FAGW/HGM HCMs were horizontally put together to get the GF-CMs. Microstructural morphologies, pore diameter distribution and tensile strength of the GF-CMs were investigated. The aim of this paper was to investigate the SHMP and HGM addition on the tensile strength of the GF-CMs and thermal conductivity of the associated GF-VIPs.

## 2. Experimental

All the raw materials, including FAGW, HGMs, and SHMP, were provided by Suzhou V.I.P. New Material Co., Ltd. (Taicang, P.R. China). The SHMP content in the glass fiber (FAGW) suspension was 0 wt.%, 0.04 wt.%, 0.12 wt.%, and 0.2 wt.%, respectively. A total of 0 g, 2.02 g, and 4.71 g of HGMs, 4.71 g of FAGW, and 1500 ml of the above white water were stirred in a cylindrical tank for 30 min and then dispersed in the GBJ-A fiber dissociation device, respectively. The rotation frequency and the beating revolution of impeller were  $48.3 \pm 1.65\text{ s}^{-1}$  and 5000 r, respectively. Each associated FAGW suspension was dispersed by mechanical crushing and abrasion on fibers through contacting with edge and faces of rapidly moving metal blades in white water. Each dispersed FAGW suspension was then transported to a papermaking machine (ZCX-200A) and stirred by a blender for 5 min. After blending, white water was drained out from the suspensions by the vacuum pump through an 80 mesh copper net while FAGW and HGMs were aligned at the surface of the copper net and then formed a wet mat at the surface of the copper net. Finally, the wet mat was dried at  $150\text{ }^{\circ}\text{C}$  for 60min.

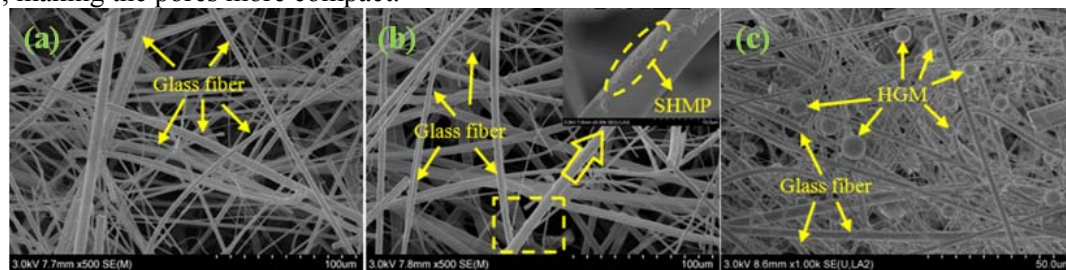
Surface morphologies of the as-prepared mats were observed by the field emission scanning electron microscopy (FE-SEM, Hitachi S-4800). The pore diameter distribution of the mats was

determined by the mercury intrusion porosimeter (MIP, AutoPore IV 9510). Tensile strength tests of the rectangular SHMP coated FAGMs and FAGW/HGM HCMs with a length of 100 mm and width of 15 mm were carried out by the electronic diaphragm tension machine (XH-018A), according to the JB/T 7630.1-2008 standard where the clamping space and velocity of the grips were 50 mm and 100 mm/min, respectively. Thermal conductivities of the as-prepared VIPs were evaluated by the heat flow meter (Netzsch HFM 436).

### 3. Results and discussion

#### 3.1. Microstructure

Figure 1 shows the SEM micrographs of the FAGM, SHMP coated FAGM and FAGW/HGM HCM. As shown in Figure 1(a) and (b), the glass fibers were slightly curled, randomly oriented and possessed a great aspect ratio. The mean diameter of glass fibers was 3.5  $\mu\text{m}$ . The glass fibers built the pores with various diameters ranging from 1  $\mu\text{m}$  to 100  $\mu\text{m}$ . As for the SHMP coated FAGM, there was a thin SHMP coating on the surface of glass fibers. But the SHMP coating was not continuous; SHMP coating was attached at only a part of glass fibers. As shown in Figure 1(c), the FAGW/HGM HCMs were densely packed with HGMs and glass fibers. The HGMs were filled in the fiber-fiber pores, making the pores more compact.



**Figure 1.** SEM micrographs: (a) FAGM; (b) SHMP coated FAGM; and (c) FAGW/HGM HCM.

#### 3.2. Pore structure of core material

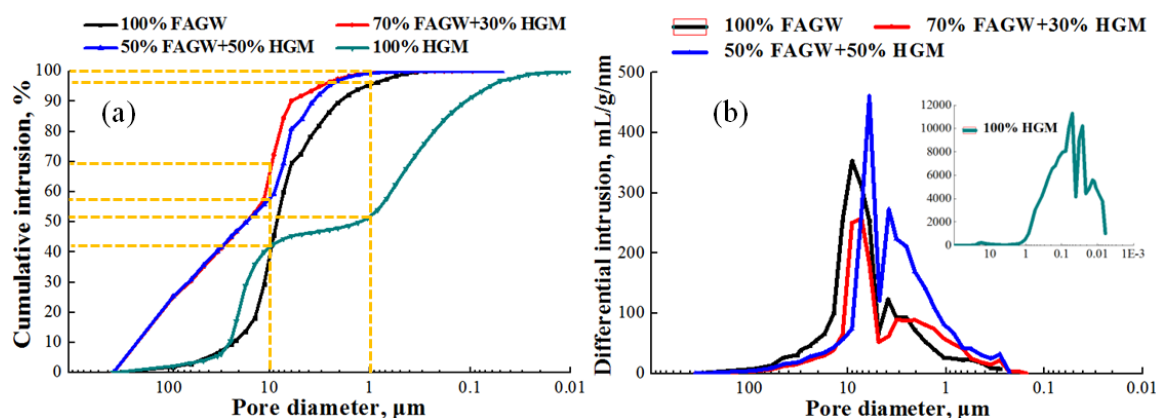
Table 1 shows the pore parameters of various core materials. Each core material possessed a high porosity of 84-91%. Also, the average pore diameter of Sample 1-3 was 8-11  $\mu\text{m}$  while that of Sample 4 was only 0.3  $\mu\text{m}$ .

**Table 1.** Pore parameters of various core materials

	Composition, %			Total intrusion volume V, mL/g	Total pore area A, m <sup>2</sup> /g	Average pore diameter (4V/A), $\mu\text{m}$	Porosity, %
	FAGW	HGM	SHMP				
Sample 1	100	0	0	6.9	2.6	10.5	86.7
Sample 2	70	30	0	4.8	2.3	8.3	85.8
Sample 3	50	50	0	3.9	1.5	10.6	84.5
Sample 4	0	100	0	8.2	116.2	0.3	90.8

Figure 2 shows the distribution of cumulative intrusion and differential intrusion in the core material calculated by MIP technique. As shown in Figure 2(a), around 41% (Sample 1), 70% (Sample 2), 57% (Sample 3), and 41% (Sample 4) of pore volume was occupied by pores with diameter  $\geq 10$   $\mu\text{m}$  while 54% (Sample 1), 30% (Sample 2), 43% (Sample 3), 10% (Sample 4) of pore volume was taken over by pores with diameters of 1-10  $\mu\text{m}$ . As shown in Figure 2(b), the pore diameter distribution of Sample 1-3 was similar with the highest peak of  $\approx 9$   $\mu\text{m}$  and two short peaks at different pore diameters. By contrast, the highest peak of Sample 4 was around at 0.07  $\mu\text{m}$  and the remaining

two peaks were at the range of 0.01-0.05  $\mu\text{m}$ . Thus, the presence of HGMs in the FAGW made the pore distribution in the core material moving into a multi-peak pattern.

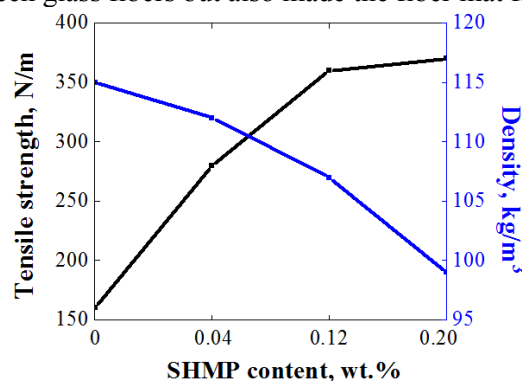


**Figure 2.** Distribution of cumulative intrusion and differential intrusion in the core material calculated by MIP technique.

### 3.3. Tensile strength of core material

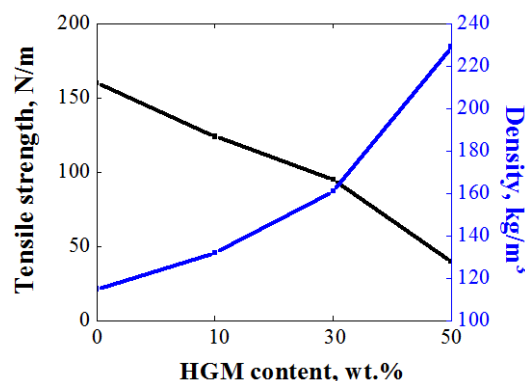
SHMP is a mixture of polymeric metaphosphates and a hexamer of composition  $(\text{NaPO}_3)_6$ . SHMP is usually used as the deflocculant in wet method for nonwoven mats. Also, it can be a dispersing agent that helps to break down the fiber aggregations.

Figure 3 shows the effect of SHMP addition on tensile strength and density of core material. The tensile strength significantly enhanced from 160 N/m to 370 N/m while the density slightly decreased from 115  $\text{kg/m}^3$  to 99  $\text{kg/m}^3$  when the SHMP addition increased from 0 wt.% to 0.2 wt.%. It was noted that the tensile strength at SHMP content of 0.12 wt.% and 0.2 wt.% was near the same, one was 360 N/m while the other was 370 N/m. Hence, the addition of SHMP not only had a great effect on enhancing the bonding between glass fibers but also made the fiber mat fluffier.



**Figure 3.** Effect of SHMP addition on tensile strength and density of core material.

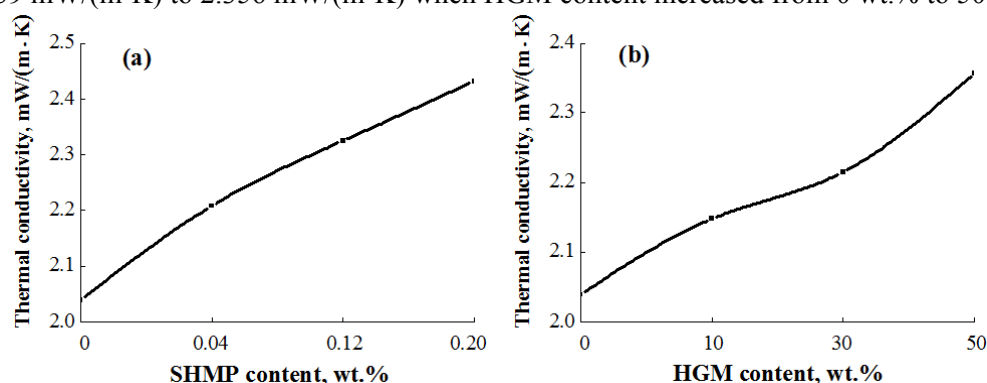
HGMs were microscopic glass spheres and acted as lightweight fillers in the composite core materials. In this paper, the diameter of HGMs was 15-135  $\mu\text{m}$ . Figure 4 shows the effect of HGM content on tensile strength and density of core material. The tensile strength of the core material significantly decreased with the increase of HGMs content while the density increased rapidly with HGMs content. The maximum density arrived at 229  $\text{kg/m}^3$  when the HGMs content was 50 wt.%.



**Figure 4.** Effect of HGM content on tensile strength and density of core material.

### 3.4. Thermal conductivity of VIP

Figure 5 shows the thermal conductivity of VIPs. Compared with VIPs with pure FAGMs, the VIPs possessed a higher thermal conductivity. Both the addition of SHMP and HGM increased the thermal conductivity of the VIPs. As shown in Figure 5 (a), the thermal conductivity of VIPs roughly linearly increased from 2.039 mW/(m·K) to 2.432 mW/(m·K) when SHMP content increased from 0 wt.% to 0.2 wt.%. As shown in Figure 5 (b), the thermal conductivity of VIPs roughly monotonously increased from 2.039 mW/(m·K) to 2.356 mW/(m·K) when HGM content increased from 0 wt.% to 50 wt.%.



**Figure 5.** Thermal conductivity of VIPs: (a) core material: SHMP coated FAGMs; (b) core material: FAGW/HGM HCMs.

## 4. Conclusion

Adding the SHMP in the glass fiber suspension was very beneficial to the enhancement of tensile strength of the FAGMs while adding the HGMs in the glass fiber suspension increased the density and decreased the tensile strength of the GF-CMs and increased the thermal conductivity of the corresponding GF-VIPs. The optimal SHMP content and HGM content in the glass fiber suspension were 0.12-0.2 wt.% and 0 wt.%.

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