



# Fabrication and characterization of low-cost and green vacuum insulation panels with fumed silica/rice husk ash hybrid core material



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## ABSTRACT

Novel fiber/powder hybrid core materials (HCMs) containing various combination ratios of fumed silica (FS), rice husk ash (RHA), carbon black, titanium dioxide and polyester chopped strand were prepared by dry powder mixing method. The HCMs and their corresponding vacuum insulation panel (VIP) samples were thoroughly characterized to get insight information about their microstructures, structural stability and thermal conductivities. The results revealed that the HCMs possessed a hierarchical meso-/macro-structure with a high porosity of 80.5–87.9% and a fine pore size of 150.9–210.3 nm. Both compression and rebound ratios of the VIPs initially increased (up to ~59%) with the increasing RHA content and thereafter decreased till 39% and 31% respectively. The initial total thermal conductivities of the VIPs were ~5.5 mW/(m K), which increased with the increasing RHA content as well. The proposed VIPs possessed not only a low total thermal conductivity (at 100–100,000 Pa) in between that of glassfiber-VIPs and opacified FS-VIPs but also a much longer service life than glassfiber-VIPs. The addition of 36 wt.% RHA resulted in a 32% reduction in the cost of HCMs, while still maintaining its super insulation capability. The optimum RHA content that led to a low-cost and highly thermal efficient VIP was 26–36 wt.%.

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## 1. Introduction

Energy used for thermal management of buildings accounts for approximately 20–40% of the global energy which overcomes the industry and transportation sectors in EU and US [1–3]. Since this energy is mainly produced by fossil fuels, it may produce a large mass of polluted air containing CO<sub>2</sub> and SO<sub>2</sub> and become a great contributing factor in environmental degradation and global warming [3–6]. In order to reduce the carbon emissions and consumption of fossil fuels, there is a need to improve the high energy-conservation technology and extend their usage in building sectors [7–10].

Vacuum insulation panels (VIPs) are regarded as “super and green insulation materials” that are free from ozone depleting chemicals throughout their production, application, and recycling processes, thus possessing dual features of energy-saving and environmental friendly.

In recent years, VIPs are getting more and more popularity in building industry because of their extremely low thermal conductivity and ultra-thin thickness that allows the buildings to conserve energy and have more habitable space as well [11]. However, the present VIPs have two main drawbacks, one is high cost and the other is unstable thermal conductivity over their useful lifetime [12]. It is known that the nature of VIP core material not only greatly affects the solid ( $\lambda_{\text{solid}}$ ), gas ( $\lambda_{\text{gas}}$ ), and radiative thermal conductivities ( $\lambda_{\text{rad}}$ ), of the VIP product, but also its stability and durability in the long run [13–15]. VIP core materials with small pore diameter ( $D$ ) and low outgassing rate are preferred for a high-quality VIP (with a low and stable  $\lambda_{\text{tot}}$ ) for its applications in buildings. Hence, micro-fiber cores, like glassfiber mat, and organic polymer foam cores, like polyurethane foam, are considered to be inappropriate for applications in building sector because of their large  $D$  and high outgassing rate [16–18]. On the other hand, conventional nanoporous core materials, like fumed silica (FS) and silica aerogel (SA), exhibit a low  $\lambda_{\text{tot}}$  around 4 mW/(m K) even at high pressures ( $\approx 1000$  Pa), but only a few of them were used in buildings due to their high cost [19].

The high cost of a VIP is mainly due to the core materials with costly fillers [20]. In recent years, significant attempts have been made to

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develop hybrid core materials (HCMs) with multi-component fiber/powders and they have performed well in thermal insulations [9]. In this sophisticated and heterogeneous system, the fibers are used to reinforce the mechanical strength of the HCMs, while the powders are added to reduce  $\lambda_{\text{rad}}$  (i.e., served as opacifiers) and/or isolate thermal conduction (i.e., served as heat-insulating fillers). Both the composition and microstructure of the HCMs are designed carefully to achieve a uniform and nanoporous structure through blending various additive amounts of coarse and fine particles. Many researchers have investigated the thermo-physical properties of diverse alternative cheap fillers such as expanded perlite [20,21], hollow glass microspheres [22], house dust [21], fly ash [21], xonotlite [23], pumice [24,25] and zeolite [24,25], as the VIP core material, without a sacrifice of their service performance. These fillers were given full consideration as the second phase alternative materials for FS or SA based VIPs, but the achieved  $\lambda_{\text{tot}}$  and cost are still high for their widespread uses.

Rice husk is a degradation-resistant agricultural waste that is abundantly available in rice producing countries. It can create serious disposal problems in the land and surrounding areas of dump, if handled improperly [26]. However, under controlled combustion condition, it can be converted into a fine insulator, named rice husk ash (RHA) [27]. The extensive application of RHA as the heat-insulating fillers in building industry may transform “this-waste” into wealth. However, only a few studies have been reported on its use for this purpose and their practical performance has yet to be explored. A comprehensive understanding of the influence of microstructure,  $D$  distribution, compression behaviors of the RHA added core material on the capacity and share to each part in  $\lambda_{\text{tot}}$  and its thermal transport mechanisms under vacuum and atmospheric pressure has become an urgency for which the existing literature is highly deficient.

Therefore, in this paper, a series of novel HCM samples were prepared with 8 wt.% of PCS fibers, 1 wt.% of titanium dioxide (TD) powders and 5 wt.% of CB powders, accounting for 14 wt.% of the samples, and FS and RHA of altered ratios, constituting the remaining 86 wt.% of the samples. The microstructure,  $D$  distribution, BET specific surface area ( $S$ ), and  $\lambda_{\text{tot}}$  of various HCMs at various pressures and the compression ratio ( $\omega$ ) and rebound ratio ( $\eta$ ) of the resultant different VIPs were thoroughly investigated to examine the influence of the additive amount of RHA on the pore structure, compression behavior, thermal insulation properties, and cost of the HCMs and VIPs. RHA content was optimized as well to keep the cost of VIPs minimum and environment friendly along with its high thermal insulation performance in building sectors.

## 2. Materials and methods

### 2.1. Materials

Raw materials, including FS, RHA, PCS fibers, TD and CB powders, and envelope materials, were all provided by YinXing Electric Co., Ltd. (Chuzhou, P.R. China). Table 1 listed the composition of various HCM samples produced and used in the VIPs. Particle size of the FS primary particles was 7–40 nm. Mean diameter and length of the PCS fibers

was 12  $\mu\text{m}$  and 6 mm, respectively. The overall thickness of the envelope materials used in this study was 95  $\mu\text{m}$  while their structure and component was polyamide (PA) film/polyethylene terephthalate (PET) film/aluminum (Al) film/polyethylene (PE) film. This envelope ensured a low oxygen transmission rate of  $\leq 0.05 \text{ cm}^3/(\text{m}^2 \cdot \text{day})$  and a low water vapor transmission rate of  $\leq 0.01 \text{ g}/(\text{m}^2 \cdot \text{day})$  during VIP processing.

### 2.2. Preparation of HCMs and VIPs

The HCMs were prepared by the dry powder mixing method. The total mass of the fillers in each HCM sample was 250 g. Fig. 1 shows the flow chart of the preparation process of HCMs and VIPs. First, a given amount of the fillers were put in a closed mixer chamber and then mechanically dispersed by a spiral rotational impeller. Later, the uniformly dispersed fiber/powder mixture was sealed in a polyester fiber non-woven bag. Afterwards, it was pressed into a plat HCM. Afterwards, the plat HCM was dried at 120  $^{\circ}\text{C}$  for 90 min and then vacuumed and sealed in an envelope using a vacuum machine (see details in our previous published research [28]). Following the same procedures, the 6 groups of VIP samples with dimensions of 290 mm  $\times$  290 mm were fabricated.

### 2.3. Microstructure, $S$ and $D$ distribution analysis

Surface morphologies of all types of HCM samples were examined under the field emission scanning electron microscope (FE-SEM, Hitachi S-4800, Japan) and transmission electron microscope (TEM, JEM-200CX, Japan). The formulated fiber/powder raw mixtures were dried at 200  $^{\circ}\text{C}$  under vacuum for 3 h before their pore structure analysis.  $\text{N}_2$  sorption isotherms,  $S$  and  $D$  distribution of the formulated fiber/powder raw mixtures in micro- and meso-pore range were produced by specific surface area analyzer (BUILDER, SSA-4300, China).  $D$  distribution in meso- and macro-pore range of the formulated fiber/powder raw mixtures was analyzed by the mercury intrusion porosimeter (MIP, AutoPore IV 9510, USA).

### 2.4. Fourier transform infrared (FTIR) spectroscopy

For FTIR spectroscopy, 200 mg of dry potassium bromide (KBr) powder was mixed with 2 mg each of the formulated fiber/powder raw mixtures by grinding, to get a homogenized blend. Later, each individual blend was compressed to form a transparent and thin tablet with a thickness of 1 mm and diameter of 13 mm. Finally, FTIR spectra of each tablet were obtained by the FTIR spectrometer (Nicolet 6700, USA) at a resolution of 0.1  $\text{cm}^{-1}$  over a spectral range of 400–4000  $\text{cm}^{-1}$ .

According to the Rossland equation,  $\lambda_{\text{rad}}$  of the optically thick HCM specimen was calculated by [29,30]:

$$\lambda_{\text{rad}} = \frac{16n^2 \cdot \sigma \cdot T_m^3}{3E_R(T_m)} \quad (1)$$

where  $n$  was the effective refraction index ( $n \approx 1$  for low-density silica),  $\sigma$  was the Stefan-Boltzmann constant ( $\sigma = 5.67 \times 10^{-8} \text{ W}/(\text{m}^2 \text{ K}^4)$ ),  $T_m$  (K) was the average temperature within the specimen ( $T_m^3 = (T_1 + T_2)(T_1^2 + T_2^2)/4$ , where  $T_1$  and  $T_2$  was the temperatures of the VIP surfaces [31]),  $E_R$  was the Rossland mean extinction coefficient ( $E_R(T_m) = e(T_m) \cdot \rho = 1/l_{\text{ph}}$ , where  $l_{\text{ph}}$  was the mean free path of the thermal photons,  $\rho$  was the sample density,  $e(T_m)$  was the mass specific extinction [31]).

**Table 1**

Composition of various HCM samples produced and used in the VIPs.

	Composition, wt.%				
	RHA	FS	PCS	TD	CB
Sample 1	0	86	8	1	5
Sample 2	6	80			
Sample 3	16	70			
Sample 4	26	60			
Sample 5	36	50			
Sample 6	46	40			

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