Alternative Hybrid Core Material For Vacuum Insulation Panels (Silica-Fly Ash-Glass Fiber)

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Abstract: Vacuum insulation panels, one of the most promising insulation materials consisting of an evacuated core material, an air tight envelope and in special cases an absorbent known as getter. However, despite its outstanding properties, it faces some challenges such as relatively high cost and quite a short service life which can be attributed to the core material used. In this paper, Hybrid core materials (HCM) consisting of various percentages of fly ash, fumed silica and glass fiber were used as a core material for vacuum insulation panels, and the composition ratio vs thermal conductivity were investigated to ascertain the optimum composition ratio that showed the lowest thermal conductivity and best insulation properties. This was to produce VIPs at a relatively cheaper cost. The optimum ratio of the HCM that showed the best insulation properties including lower thermal conductivity is that of 65% fly ash (FA), 30% fumed silica (FS) and 5% glass fiber (GF). The HCM produced exhibited similar qualities as that of silica powder core VIPs. Even though produced at a relatively lower cost, the insulation properties were not compromised. Furthermore, the thermal conductivity of each of the VIPs from the HCMs prepared were measured after undergoing a temperature stress of 60 °C for 6 months.

Index Terms: Core material, Fly ash, Fumed silica, Microstructure, Thermal conductivity, VIP

1 INTRODUCTION

CHINA and many other developed countries have realized the need to channel efforts to promote green buildings, thus making good use of energy efficiency products. This is as a result of the high energy consumed by buildings [1] and [2] estimated to be about 20–40% of the total energy used worldwide [3]. In China, about 65% of energy consumed by buildings is spent on space heating and cooling [4] . Traditional insulation materials in the form of organic and inorganic have therefore gained popularity in energy consumption reduction in buildings due to their low thermal conductivity [5]. They come in the form of powders, fibers and foams. Expanded polystyrene (EPS), extruded polystyrene (XPS), and polyurethane (PU) foams are examples of the organic insulation materials used while silica, glass wool, rock wool all form part of the inorganic insulation materials [5] and [6]. However, their poor resistance to fire poses a big threat to the building industry hence paving way for the development of other alternative insulation materials with excellent insulation properties and better fire resistance. In China, two fire incidents took place in Beijing and Shanghai as a result of the use of organic insulation material in the year 2010 [5]. The process made in the search of alternative insulation brought about the application of vacuum technology which was developed from the principle with which Dewar flask operates. In this case, a Vacuum Insulation Panel (VIP). Vacuum insulation panels have seen significant application in the transportation industry, storage facilities and appliances and most importantly building construction [7]. Vacuum insulation panels comprise of an evacuated porous core material with pore size of about 10-100nm which is enclosed in a thin air tight barrier laminate popularly known as the envelope and in special cases, a getter material is embedded in it. Depending on the kind of core material being evacuated, pressures of about 0.2-3 mbar can be acquired in order to achieve a thermal conductivity of 0.004 W/mK and low [7] and [8].

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A lower thermal conductivity is attainable at a high vacuum level, in that, heat transfer is minimized within the porous core material thereby creating large thermal resistance [7]. VIPs like any other system has some defects which makes its usage a bit challenging. To name but a few of these defects are its expensive nature and instability in their insulation properties (thermal conductivity) over a period of time [2], [3] and [9]. The core material of VIPs contribute significantly to these defects. The type of core material used can affect the cost of the VIP and it is also responsible for the vacuum level [3] and [10].

2.0 CORE MATERIALS

Many of the conventional or traditional insulation materials have been used as core materials but aerogels were seen to be the best. However, due to its expensive nature its application is limited and has therefore paved way for the use of silica powders since they have similar properties as silica aerogels [11]. For instance, silica powders show less sensitivity to increase in gas pressures even at 1000 Pa and they have low thermal conductivity as well [3], [5] and [11]. Silica powder which is the next most appropriate alternative to silica aerogels shows a significant increase in cost when used as core material for VIP even though it's quite less expensive compared to silica aerogels [7]. Core materials in order to best fit for use as insulation materials need to fulfil certain requirements. The pore size and porosity are of much importance thus, having an open pore structure and very small pore size and diameter. Thermal conductivity (λ) is affected by the gas pressure (P_{gas}) of the core material whereas the typical pressure ($P_{1/2}$) depends on the pore size [12] and [13]. Represented mathematically as

$$\lambda = \frac{\lambda_0 + \lambda_{gas}}{1 + P_{1/2}/P_{gas}} \quad \text{..... Eqn. 1}$$

In the development of an alternative and effective core material at a relatively low cost, hybrid core materials (HCM) consisting of fibers and multi powders showed a positive impact on insulation [3] and [7]. The fibers played a role in the HCM by providing mechanical support and to enhance easy compression of the powders while the powders mainly reduce or eliminate the various forms of thermal conduction and radiation [3] and [14]. In this experiment, the HCM consists of Fly ash (FA), Fumed silica (FS), and Glass Fibers (GF).

However, fly ash (FA) is the main raw material used to partially substitute FS, hence gains much attention.

2.1 FLY ASH POWDER (FA)

2.1.1 Production

In China, large amount of fly ash is produced as a result of using coal in the generation of electricity and recorded about 50.2% of the world wide coal consumption [15]. Fly ash a particulate matter, is one of the by-product generated from thermal plants as a result of coal combustion [16] and [18]. There's so much motivation these present times to look into Fly ash since it presents itself as a perfect candidate for the building and construction industry [17]. Pulverized coal is fed into the boilers combustion chamber where ignition of coal takes place and generates heat to produce a molten mineral. The flue gases produced in the process are cooled off thereby causing the molten residue to solidify and form ash. Ashes produced from the combustion of coal come in the form of coarse and fine particles whereby the coarse particles are known as bottom ash or slag representing about 5–15 wt.% of the total ash produced. Meanwhile, fine ash also known as fly ash forming about 85–95 wt.% of the total ash generated remain suspended in the flue gases and obtained by the use of particulate emission control devices such as electrostatic precipitators, bag houses and mechanical cyclones. [16], [17] and [19]. Fly ash can also be referred to as Coal Ash, Pulverized Flue Ash, and Pozzolona [20].

2.1.2 Physical and Chemical properties of fly ash Powder

Generally, fly ash is a fine grained powdery particulate matter with size in the range of 0.5 µm – 100 µm, spherical in shape and possesses a light tan color [19] and [20]. It has a specific surface area of 308 m^2/kg and specific gravity of 2.4. Basically, two types of fly ash exist. They are class C and class F types and the difference is based on their silica, alumina and iron content. The primary components of fly ash are silica (SiO_2), alumina (Al_2O_3) and oxides of iron (Fe_2O_3) despite the existence of other minerals in minute quantities. Class F fly ash has its silica, alumina and iron content adding up to not more than 70% while that of Class C adds up to not more than 50% [17], [19], [20] and [21]

$$Class\ F = SiO_2 + Al_2O_3 + Fe_2O_3 \geq 70\ \% \quad \text{..... Eqn. 2}$$

$$Class\ C = SiO_2 + Al_2O_3 + Fe_2O_3 \geq 50\ \% \quad \text{..... Eqn. 3}$$

The image below shows a sample of class F fly ash

Fig 1: *Image of class F fly ash*

3.0 MATERIALS AND METHODS

3.1 Materials

Fly ash, fumed silica and glass fibers raw materials used were commercial grade provided by Yichen New Energy Co.Ltd. (Qingdao P.R China). Five Different ratios of the HCMs were prepared. The total mass of each of the HCM was 250 g. While the wt. % of fly ash and fumed silica were varied, glass fiber content was kept constant. The HCMs obtained are as follows: 70% FA: 25% FS: 5% GF, 65% FA: 30% FS: 5% GF, 60% FA: 35% FS: 5% GF, 50% FA: 45% FS: 5% GF, 45% FA: 50% FS: 5% GF Table 1. Shows the various samples of HCMs prepared for the experiment

Table 1: Samples of HCMs prepared

	Composition wt. (%)		
Sample ID	FA	FS	GF
Sample A	70	25	5
Sample B	65	30	5
Sample C	60	35	5
Sample D	50	45	5
Sample E	45	50	5

3.2 Preparation of VIP Samples

Fumed silica particle size was within the range of 6-42 nm while that of fly ash was less than 100 µm. Glass fiber strands were also 2-6µm long and 3-6 mm wide. Also, the envelope used in this study comprised of an outer glass fiber cloth (100 g), PE (100g), PA (15g), PET (12g) and Al (7g). The contents of the HCMs were introduced into a hopper and mixed thoroughly in a mixing drum. They were dried at a temperature of 100 °C for about 120 minutes in order to get rid of any form of moisture. The dried HCMs were transferred into moulds and pressed into boards of desired shape and size at a compression or initial pressure of 1.2 MPa. Subsequently, the moulds/core boards were wrapped air tight in a thin inner filmed bag and inserted into the gas barrier envelope which is sealed at one end. It is then placed in the vacuum sealing chamber. The chamber is then evacuated to an inner pressure of 1000 Pa and the open end of the envelope is heat sealed. This process was repeated for all the HCMs resulting in the acquisition of 5 VIPs. A square shaped VIP with dimensions 250mm*250mm was produced.

3.3 Microstructure and Pore size distribution

The surface morphologies of all the HCM samples were scrutinized using the scanning electron microscope (SEM-PW_ PHENOM_XL) before they were molded into boards. The HCMs were dried at a temperature of 100°C for approximately 4 hours under vacuum in order to obtain accurate results during the pore size distribution analysis by the use of a mercury intrusion porosimeter (MIP, AutoPore IV 9510, USA).

3.4 Thermal conductivity and Effect of thermal Stress

The thermal conductivities of the VIPs produced were

measured using the heat flow meter Netzsch HFM 436 while a high temperature furnace DHG-9036A was used in the determination of the effect of thermal stress on each sample.

4 DISCUSSION AND RESULTS

4.1 Microstructural Characterization

X-Ray Diffraction (XRD) and X-Ray Fluorescence (XRF) analysis depicted the fly ash used was that of class F based on Eqn. (2) and (3). The table 2 shows the percent by weight of each of the component of the fly ash.

Table 2: *Composition of class F fly ash and their various wt. %*

Chemical Compound	Percent Composition
SiO_2	54.90
Al_2O_3	25.80
Fe_2O_3	6.90
CaO	8.70
MgO	1.80
SO_3	0.60
Others	1.5

Fig 2 illustrates SEM images of fly ash, fumed silica, glass fiber and the prepared HCM A, B, C and D respectively. Fig. 2 (A) shows particles that are seen to be glassy, and spherical in nature. Fly ash particles exhibit irregular shapes with varying sizes within the range of microns (coarse) and nanometers (fine). The dominant size of the fly ash particle is about 60 µm and has bulk density of about 540 kg/m^3 translating into porosity (φ) of about 70-85%. Fig. 2(B) as shown represents the SEM image of fumed silica. Fumed silica particles were seen to be more closely linked in a chainlike form and with random pore size structures. Particle sizes were also uniformly distributed and in few nanometers of 6-42 nm. It's seen to be very porous with porosity of about 90-91% and bulk density of about 130kg/m^3 Fig. 2(C) shows the SEM image of glass fiber. As shown, glass fibers had a length of about 2-6µm and diameter of 3-6 mm. The surface of the fibers were also glassy and smooth in nature. Fig. 2(D) depicts the HCM prepared from a mixture of fumed silica, fly ash and glass fibers. It is a made up of multi scale particles including fibers, thus having different pores and particle size. Also, its porosity ranged between 80-86%. The pores are not so conspicuous due to the multi scale particle sizes. Because the pore size of A (FA) is relatively larger than B (FS), the small pores in B are able to occupy or fill the larger pores in A in the formation of D (HCM). The fibers are evenly distributed giving the skeleton a form of structural strength.

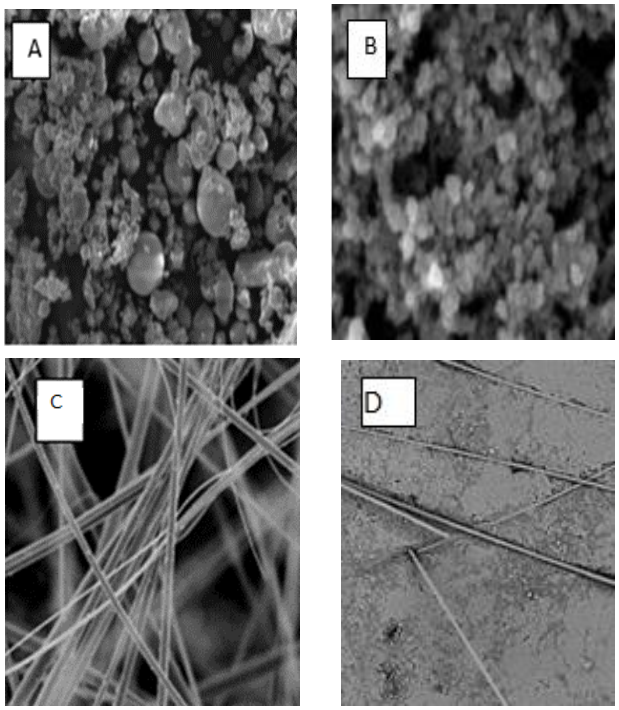


Fig 2: *SEM images of* ***(A)*** *Fly ash* ***(B)*** *Fumed silica* ***(C)*** *Glass fiber* ***(D)*** *HCM.*

The figures below show the various HCMs prepared

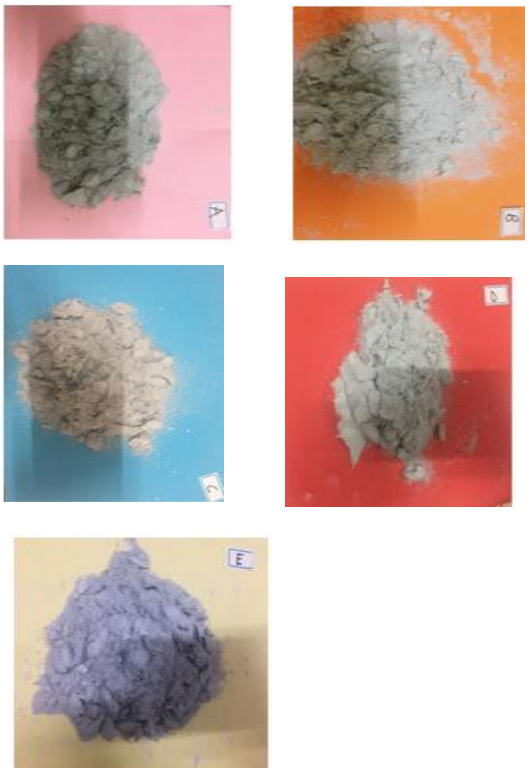


Figure 3: *Images of various sample of HCM*

The table below shows the various samples of HCMs and their respective porosity obtained by Mercury Intrusion Porosimetry (MIP)

Table 3: *Porosity of various HCMs prepared*

Sample ID	Ratio (%) FA:FS:GF	Porosity (%)
A	70:25:5	87
B	65:30:5	90
C	60:35:5	86
D	50:45:5	78
E	45:50:5	80

The pore size and porosity of the core material, the medium through which heat is transferred determines its internal pressure. However, thermal conductivity of the material depends on its internal pressure. Porosity was obtained from the bulk volume and pore size while the bulk volume and pore volume were obtained using the Mercury Intrusion Porosimetry (MIP). The powders were developed into core boards for vacuum insulation panels from the HCMs by compression using a Universal Testing Machine at a pressure of 1.2MPa. Dimensions obtained were 250*250 mm and their thermal conductivities under pressures of 1000 Pa were measured using the heat flow meter Netzsch HFM 436. Vacuum insulation panels namely A1, A2, A3, A4 and A5 with HCM compositions as 70% FA: 25% FS: 5% GF, 65% FA: 30% FS: 5% GF, 60% FA: 35% FS: 5% GF, 50% FA: 45% FS: 5% GF, 45% FA: 50% FS: 5% GF respectively were produced. The table below shows the VIP samples and their core composition

Table 4: *VIPs and their HCM composition*

Sample ID	HCM composition Ratio (%) FA:FS:GF
A1	70:25:5
A2	65:30:5
A3	60:35:5
A4	50:45:5
A5	45:50:5

Presented below is a sample of the hybrid core (fly ash, fumed silica and glass fiber) VIP with the glass fiber cloth envelope and a cut section showing the core material

Fig 4: (a) *VIP with hybrid core material of fly ash, fumed silica and glass fiber* ***(b)*** *Cut section of the VIP showing its core material*

4.2 Thermal conductivities of the vacuum insulation panels

It can be deduced from the graph below that, sample A2 comprising 65FA: 30FS: 5GF showed the lowest thermal conductivity (0.007W/m.K). Investigations proved that, there was a thorough mixture of fumed silica and the fly ash particles. However, the large pores in the fly ash were filled completely or partially by that of fumed silica, hence reducing the pore size of the HCM. The glass fiber strands were also seen to be uniformly distributed. The average pore size of the sample A2 core material reduced giving it high porosity. At higher pressures, fumed silica helps in the reduction of gaseous conductivity of fly ash. However, comparing A2 and A1, a slight increase in fly ash and decrease in fumed silica as seen in A1 caused a rise in thermal conductivity. A1 and A3 showed the same thermal conductivity. Even though their porosities were 87 and 86 respectively, it could happen that some of the large pores of fly ash were left unfilled by fumed silica particles. A4 and A5 showed the highest thermal conductivity of 0.019 and 0.018 respectively. It could be attributed to the relatively large pore size of fly ash left unfilled completely or partially by fumed silica. Even though the overall thermal conductivity of the HCM VIP, is higher than pure fumed silica core VIPs which is about 0.004 W/m.K, the difference is minimum and it is reasonable to achieve cost reduction when fly ash is partially replaced with fumed silica.

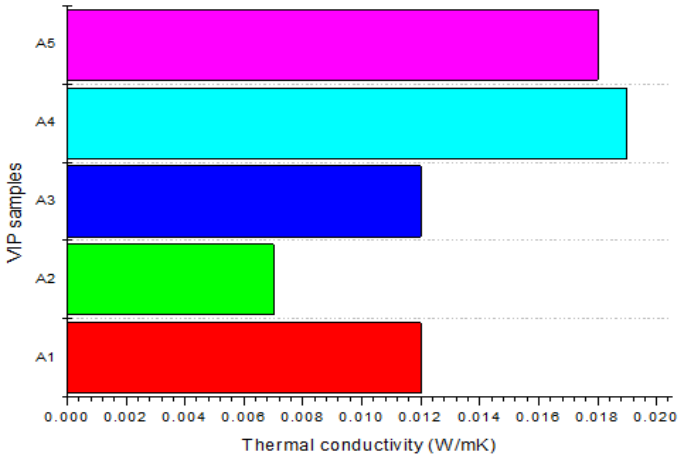


Fig *5: graph showing the thermal conductivity of HCM samples*

4.3 Effect of thermal stress for service life determination

Thermal conductivity of vacuum insulation panels increases over time thereby reducing its insulation performance due to the entrance of gases and water through the envelope. Internal pressure also increases as a result [2] , [22]. In this experiment, thermal stress was applied on the various core boards produced from each of the HCMs. The main parameters used in reference to thermal conductivity are humidity and temperature but this work focused on only thermal stress. The five samples of HCM VIPs namely, A1, A2, A3, A4 and A5 prepared went through thermal stress for 6 months. Temperature is said to be one factor of VIP deterioration [23]. VIPs were exposed to stress under a temperature of 60°C in a high temperature furnace (DHG-9036A). After about 30 days, the samples were brought

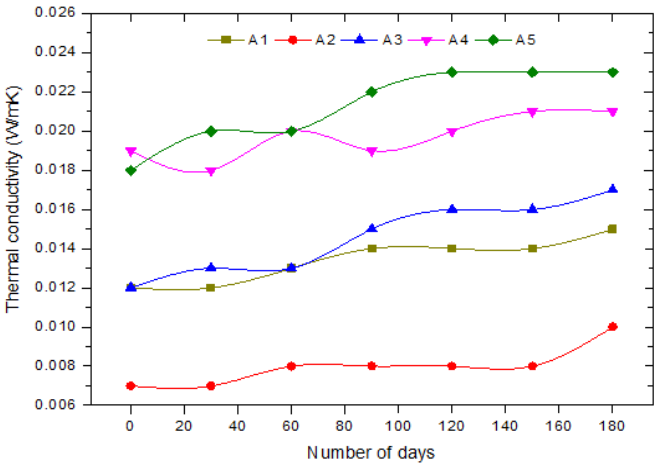
out of the furnace and their thermal conductivity and dimensions measured.

Fig 6: (A) *Left: High temperature furnace (Open) with prepared VIP* ***(B)*** *Right: High temperature furnace (Closed) with prepared VIP.*

The figures below show the images of the heat flow meter (Netzsch HFM 436) used in measuring the thermal conductivities

Fig. 7 (A) *Left: Heat flow meter (Netzsch HFM 436) (Closed) with prepared VIP* ***(B)*** *Right: Heat flow meter (Netzsch HFM 436) (Opened) with prepared VIP sample*

The results obtained after the application of thermal stress on



the VIPs are represented in the graph below.

Fig 8: *A graph showing thermal conductivities of the HCM VIP samples after application of thermal stress*

Increase in thermal conductivity of the VIPs can be attributed to increase in pressure and moisture content of the core material. In this graph, change in thermal conductivity is attributed to increase in pressure and moisture content in the core material which can result when VIPs are subjected to extreme high temperatures. However, the change in thermal conductivity over time can be used to estimate the service life of VIPs. Moisture in VIPs cause an increase in the pore gas pressure and solid conduction by changing the contact between particles of the core material thereby increasing the thermal conductivity [22]. From the graph, it can be seen that there was a slight increase in thermal conductivity of all the VIP samples after their initial thermal conductivity. For sample A2, it can be seen that, the thermal conductivity increased slightly after the 30th day and remained constant from the 60th day until the 150th day. That notwithstanding, the increase was gradual. All the VIPs showed some significant rise in thermal conductivity. Further tests can be performed to determine the accelerated aging in the presence of high temperatures and relative humidity. A4 showed an anomaly. Its thermal conductivity kept rising and decreasing and remained stable on the 150th and 180th day. A5 even though started with an initial thermal conductivity lower than that of A4, it turned out to have the highest thermal conductivity after the 180th day. The microstructure of the pores are also responsible for the change over the time period.

5.0 CONCLUSION

The amount of each constituent of the hybrid core material influences the thermal conductivity of the VIP. Hybrid core materials constituting fly ash, fumed silica and glass fiber were analyzed in this work for use as VIP core materials so as to acquire VIPs at a relatively low cost. Fly ash readily available in China, having similar properties as fumed silica and obtained at a cheaper was the major raw material. The microstructure and other properties that influence thermal conductivity of this HCM were also examined. Pore size and thermal conductivity play keen roles in optimizing the mass % of each of the constituents of the HCM in order to produce an effective core material for VIPs. Deductions made were that, fly ash had pore sizes of < 100 µm and porosity between 80 and 86% with thermal conductivity of 0.02 W/m.K at a pressure of 1000 Pa which doesn't render it best as an insulation core material. However, upon addition of fumed silica and glass fiber, the thermal conductivity improved fairly. The small particles of fumed silica filled the large pores of the fly ash either completely or partially thereby reducing its overall pore size resulting in a low thermal conductivity. The thermal conductivity improved from 0.02 W/m.K to 0.007 W/m.K at a pressure of 1000 Pa which was quite impressive. Based on the results obtained after performing several experiments on the samples, HCM sample B corresponding to VIP sample A2 (65% fly ash (FA), 30% fumed silica (FS) and 5% glass fiber (GF)) possessed the highest porosity of 90% and showed the lowest thermal conductivity of 0.007 W/m.K as well. Effective core material for VIPs must have a high porosity, lower thermal conductivity and small pore size. Low cost materials are needed in order to help reduce the cost of VIPs. These materials will partially or completely displace expensive fumed silica. In this research work, Fly ash is to partially replace fumed silica. The cost of fly ash is as 10 times less expensive compared to fumed silica. A ton of fly ash costs US $15 while a ton of fumed silica is about US $500-900. Several researchers have worked on low cost materials as cores for VIPs but none has worked on fly ash, fumed silica and

glass fiber composite.

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