The effects of PE additive on the performance of polystyrene vacuum insulation panels

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ABSTRACT

The effects of adding polyethylene (PE) in polystyrene (PS) foaming material on the cell structure and the heat transfer of vacuum insulation panels (VIPs) are examined in this study. Several parameters are proposed to describe the foam structure, namely, the broken cell ratio, the average cell size and the solid volume fraction. Adding 2% PE was effective in altering the cell structure and reducing the heat transfer, while adding 5% PE did not improve the performance further. The lowest thermal conductivity found in this study is 4.4 mW m⁻¹ K⁻¹, which is among the best published performances of VIP.

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1. Introduction

Vacuum insulation panel (VIP) features extremely low thermal conductivity and is suitable for numerous energy conservation applications, such as refrigerator insulation. It is constituted of porous material enclosed in evacuated non-permeable package that is normally made of metal foil envelope. Evacuating the package to a vacuum effectively eliminates the heat transfer by gas convection and conduction. Combining the vacuum with the low thermal conductivity of the porous material, which acts as the VIP's structural support, can greatly reduce overall heat transfer. Commercially available VIPs have currently reached an effective thermal conductivity that is two to six times lower than ordinary foam insulation. The porous cells in the materials must be largely open, that is, broken and connected forming a network so that all the gases can be effectively evacuated. Further reduction in the heat transfer relies on the balance between solid conductive and radiative heat transfer, as we have proposed in an earlier study[1]. Nevertheless, controlling the material structures to obtain optimum performance VIPs is currently still a challenge.

Many previous studies have attempted to determine the heat transfer of solid conduction [2–5], gaseous conduction [5] and thermal radiation [6–18] in porous medium. Most of the studies assumed all closed-cell or all open-cell structures in their analyses. Our earlier work [1] has attempted to characterize the geometrical parameters in VIPs with cell structures in-between all closed-cell and all open-cell, which means that part of the cells are closed and contain gases. It was found that broken cell ratio and cell size have been the deciding factors in reducing thermal transfer. The present study examines the possibility of increasing broken cell ratio by adding polyethylene (PE) into polystyrene in manufacturing the porous materials of VIP. Polyethylene has a higher solidification temperature and becomes hardened when the temperature is still above the melting point of polystyrene. Solidified PE particles could exert shear forces on surrounding molten PS to augment the breaking of closed cells during their expansion in the foaming process. Furthermore, the possibility of modulating cell sizes through PE additives is also examined in detail. Small cell size implies greater solid conduction routes, while large cell size leads to enhanced radiation transport. It is believed there is an optimal cell size that can render the lowest total heat transfer [1]. Adding PE provides a possibility of modulating the cell size and reducing the total heat transfer.

A total of 42 samples with different PE contents, namely, 0 wt%, 2 wt% and 5 wt%, are fabricated in this study. Their heat transfer rates are measured and analyzed. The results will be helpful in manufacturing VIP with improved performance.

2. Experiments

2.1. Sample fabrication

The samples were prepared by the following procedure. A mixture of polystyrene, polyethylene, carbon black, and calcium stearate were put into a batch die of 400 mm diameter and subjected to a 40-ton press. After mixing with the molten mixture, foaming was performed by introducing CO₂ and R-134a into the die to form
a supercritical fluid. The high pressure gas in the die was released after 6 h, forming a plain board measuring 250 mm long × 250 mm wide × 6–26 mm thick. After about an hour of heating, the material was enclosed by a metal foil envelope, which was sealed after the enclosed air was evacuated to 10⁻⁴ torr. Experiments were designed to vary the cell geometry of the samples by modulating die temperature and gas pressure. Heaters controlled the die temperature, and maintained a fixed temperature ranging from 398 K to 408 K with a stability of ±0.5 K. The minimum optical thickness of all the samples in this study is 45, thereby prevailing an optically thick medium that can be treated as a diffusion process. The radiant transfer is simply [20],

\[ q_r = -k_r \nabla T = -\left(\frac{16\sigma_T^3}{3\sigma_e}\right) \nabla T \]

where the equivalent thermal conductivity is defined as

\[ k_r = -\left(\frac{16\sigma_T^3}{3\sigma_e}\right) \]

where \( T_m \) is the arithmetic mean of the boundary temperatures. The Rosseland mean extinction coefficient (\( \sigma_e \)) is defined as

![Diagram](image_url)

**Fig. 1.** The schematic process of dual pressure control system for modulating the forming pressure [1].
The local energy flux ($q_t$) in VIPs is composed of the transfer by combined gas conduction and solid conduction ($q_{s,g}$), and by radiation ($q_r$),

$$q_t = (q_{s,g} + q_r) = (-k_{s,g} + (16\sigma T_m^4)/(3\sigma_e))\nabla T$$

Then, the concept of equivalent thermal conductivity applies,

$$k_t = k_{s,g} + k_t$$

where $k_t$ is the equivalent total thermal conductivity, $k_{s,g}$ is the equivalent thermal conductivity of combined solid and gas, and $k_t$ is the fraction of equivalent thermal conductivity induced by thermal radiation. An EKO model HC-072 conductivity meter was used in this study to measure $k_t$ and keep the temperature difference on both sides of the sample at 0.1 K during the measurements. The equivalent thermal conductivities of all the samples were measured at a hot side temperature of 30 °C and a cold side temperature of 0 °C. The equivalent thermal conductivity is calculated by

$$k_t = (E \cdot L)/(S \cdot \Delta T)$$

where $E$ is the output of the heat-flow meters, $L$ is the thickness of the sample, $S$ is the sensitive of heat-flow meter, and $\Delta T$ is the temperature difference between the hot and the cold plate. The equivalent thermal conductivity uncertainty of the data of sample L3 is estimated by

$$\langle \delta k/k_t \rangle = [(k_t/q_t)^2\delta q_t^2 + (k_t/S)^2\delta S^2 + (k_t/\Delta T)^2\delta \Delta T^2]^{0.5}/k_t$$

$$= [(6.6/23.34)^2(0.02)^2 + (6.6/0.00646)^2(0.00005)^2 + (6.6/22.8)^2(0.1)^2]^{0.5}/6.6$$

$$= (0.05896/6.6) = 0.0089$$

Thus, conductivity measurement uncertainty was controlled to within 0.89%, as estimated by the method of Wu et al. [12].

This study uses a Perkin-Elmer Spectrum 2000 Fourier Transform Infrared Spectrometer to measure the spectral transmittance of each sample. A thinly sliced foam specimen was subjected to normal incident irradiation in the wavelength range of 2.5–25 μm for the measurement. The moisture and volatile organic gas contents of specimens were first removed by an oven. The spectral transmittance is defined as

$$\tau = \int_{0}^{\infty} \frac{1}{\sigma_e} \sigma_b d\lambda$$

where $\sigma_b$ is the spectral emissive power, and $\sigma_e$ is the total emissive power of a blackbody. By neglecting the emission terms and in-scattering terms of a cold homogeneous medium under the influence of a relatively strong but unidirectional beam of radiant energy, the radiation intensity is governed by Beer’s law,

$$\frac{di_r}{ds} = -\sigma_e i_r(s)$$

where $\sigma_e = (\sigma_{se} + \sigma_{sc})$ is the spectral extinction coefficient. The transmittance is defined as

$$\tau = \frac{i_r(s)}{i_r(0)} = \exp(-\tau_{se}s)$$

The local energy flux ($q_t$) in VIPs is composed of the transfer by combined gas conduction and solid conduction ($q_{s,g}$), and by radiation ($q_r$),

$$q_t = (q_{s,g} + q_r) = (-k_{s,g} + (16\sigma T_m^4)/(3\sigma_e))\nabla T$$

Then, the concept of equivalent thermal conductivity applies,

$$k_t = k_{s,g} + k_t$$

Note that $k_t$ and $k_{s,g}$ reveal the contribution by radiation and combined solid and gas, respectively. To further distinguish the contribution by solid and by gas, this study employs a broken cell.
ratio, $\phi$, representing the ratio of broken cell volume to the total cell volume

$$\phi = \frac{V_b}{V_{tb}} = \frac{V_t - (m/\rho_{s-g})}{V_t - (m/\rho_t)}$$

where $V_b$ or the broken cell volume is the vacuum volume inside the VIP (which actually contains air in extremely low pressure), $V_{tb}$ is the volume of all the cells, $V_t$ is the apparent volume (total volume), $m$ is the weight of the sample, $\rho_{s-g} = m/(V_{tb} + V_t)$ is the density of the combined solid and gas in the unbroken cells, $V_{s-g}$ is the volume of combined solid and gas in the unbroken cell, $V_{tb}$ is the volume of gas in the unbroken cell, and $V_t$ is the volume of solid. The apparent density, or foam density, $\rho_t = m/V_t$, was measured using the ASTM D-1622 method. Note that this approach disregards the weight of the extremely low-pressure gas in the vacuum. Subtracting the broken cell volume from the total volume produces $V_{s-g}$. The former was measured by an AccuPyc 1330 Pycnometer with an accuracy of 0.03%. The term $\rho_b$ is the density of the solid, taken as the density of the raw polystyrene, which is 991.96 kg m$^{-3}$.

The solid volume fraction, $f_s$, is the ratio of solid volume to the total volume and is readily obtained by dividing the foam density of the sample by the polystyrene density.

$$f_s = V_s/V_t = 1 - \left(1 - f_{s-g}/\phi\right)$$

(11)

3. Results and discussion

Table 1 summarizes the measurement results of the samples without PE additive. The samples fall into two distinct groups with different solid volume fraction. The first group, referred to as PE0L, has a lower solid volume fraction, and includes PE0L1 to PE0L6 with $0.0413 < f_s < 0.0494$. The second group, referred to as PE0H, has a higher solid volume fraction and includes PE0H1 to PE0H8 with $0.065 < f_s < 0.0706$. Similar results of samples with 2.0 wt% and 5.0 wt% PE additive are also listed in Table 1, respectively. Similar to the samples without PE additive in Table 1, each PE additive contains two distinct groups with different solid volume fraction. The groups with higher solid volume fraction are designated as PE2H and PE5H, and the groups with lower solid volume fraction are designated as PE2L and PE5L for the 2% and 5% PE samples, respectively. Note that all solid volume fractions in the 42 investigated samples are extremely low (less than 0.07), indicating a good foaming process. Nevertheless, the distinction between high and low solid volume fractions in each table is sharp and allows us to investigate the effects of solid volume fraction.

Figs. 3 and 4 show examples of spectral transmittance and spectral extinction coefficient, respectively. Note that the spectra do not reveal any CO$_2$ absorption, which could occur at 2.7 $\mu$m.
of CO₂ and H₂O trapped in the unbroken cells is insignificant in terms of influencing radiation heat transfer. This is reasonable since most of the cells in the samples are broken and evacuated.

Fig. 5 plots the broken cell ratio versus the cell size. Each group shows an almost linear dependence of cell size on open cell ratio. Higher solid volume fraction is typically associated with larger cell size for a given PE additive weight percentage. The trend can be explained by the fact that a higher solid volume allows the cells to expand further before breaking. In the meantime, in order to obtain a higher broken cell ratio, more of the unbroken cells must be expanded further until they are broken, which also increases the average cell size. Different slopes of the relationship between broken cell ratio and cell size for different PE additive weight percentages in Fig. 5 are attributed to the effects of PE on the strength of cell membranes. These effects are also responsible for the larger cell sizes of PE2 and PE5 when compared to PE0. PE0H is an exception because its solid volume fraction is too high, which leads to large cell size as explained earlier. PE’s high melting temperature makes them more likely to solidify than PS during the cooling process in foaming and create membrane shear stress when the cells are growing, which helps to raise the broken cell ratio. If the PE additive is too much, however, the cell membrane strength could be augmented too much and the cells would grow larger without becoming broken. It will become evident in the following discussion that 2% PE is appropriate in terms of balancing cell size and broken cell ratio, while 5% PE leads to larger cell size and lower broken cell ratio. To summarize, cell size is influenced by three parameters, the broken cell ratio, the solid volume fraction and the PE additive. Among which, the PE additive is the easiest one to control and is an effective way to modify cell morphology.

Figs. 6 and 7 plot the Rosseland mean extinction coefficient against variations in cell size and broken cell ratio, respectively. The extinction coefficient in VIP consists of two parts, the absorption part, \( \sigma_a \), and the scattering part, \( \sigma_s \), that is, \( \sigma_e = \sigma_a + \sigma_s \). The former represents the absorption effect of solid material and depends largely on the solid volume fraction. The latter is affected by the morphology of the porous foam structure, which is characterized by the average cell size and the broken cell ratio. Smaller cell size implies a shorter mean free path and a larger scattering coefficient for thermal radiation. The mean extinction coefficient therefore increases as the cell size decreases, as evident in Fig. 6. For a given PE additive percentage, Fig. 7, the group with higher solid volume fraction exhibits only a slight increase in extinction coefficient compared with the lower solid volume fraction group.
although the average solid volume fractions of the two groups differ significantly. This can be explained by the fact that the solid volume fraction of all the samples are so small that the extinction is dominated by scattering and the solid absorption contribution is relatively insignificant. Extinction coefficients generally decrease as the broken cell ratio increases, as shown in Fig. 7, due to reduced scattering by closed cell membrane. Although radiation extinction is a complex process influenced by cell morphology, the broken cell ratio proposed in this study is a suitable parameter to correlate the extinction coefficient for a given PE additive percentage. Adding 2% PE is effective in increasing the extinction coefficient, due mainly to the alteration of cell morphology. Increasing the PE additive to 5% does not increase the extinction further. On the contrary, the extinction at the same broken cell ratio drops to a lower amount than the case without PE additive. This can be explained partly by the fact that the cell size has grown too large in 5% PE samples. The trend in Figs. 6 and 7 should be examined carefully, as the cell size, the solid volume fraction and the broken cell ratio all appear to influence the extinction coefficient. Nevertheless, the apparent higher extinction coefficient for higher solid volume fraction shown in Fig. 6 can be explained by the lower broken cell ratio associated with higher solid volume fraction, as evident in Fig. 5.

Fig. 7. Rosseland mean extinction coefficient varied with broken cell ratio with/without PE additives on high and low solid volume fraction.

Fig. 8 shows the equivalent thermal conductivities of samples without PE additive, including the total thermal conductivity, $k_t$, the thermal conductivity by solid/gas conduction, $k_{sg}$, and the thermal conductivity by radiation, $k_r$. The total thermal conductivity of the lower solid volume fraction group, PE0L, is generally lower than that of the higher solid volume fraction group, PE0H. This difference is mainly caused by a change in solid/gas conduction, which accounts for more than 80% of the heat transfer in the samples. Also, solid/gas conduction increases as the cell sizes decrease, which is associated with lower broken cell ratio and creates more conduction transport routes in the material. On the other hand, radiation decreases as the cell size decreases. Note that the decrease in radiation (increase in extinction coefficient) is attributable to the change in broken cell ratio, as explained earlier. Consequently, there is a best cell size (best broken cell ratio), which leads to the lowest total thermal conductivity after combining $k_{sg}$ and $k_r$ for each group of samples. In Fig. 8, the lowest total thermal conductivity is around 6.5 mW m$^{-1}$ K$^{-1}$, which occurs in the PE0L group at a broken cell ratio of approximately 0.95 corresponding to a cell size of about 100 μm. Figs. 9 and 10 shows the equivalent thermal conductivities of PE2 and PE5 groups, respectively, with trends similar to that in Fig. 7. The best cell size of

Fig. 9. The relation between equivalent thermal conductivity and cell sizes for PS core material with 2% PE additive.

Fig. 10. The relation between equivalent thermal conductivity and cell sizes for PS core material with 5% PE additive.
the 2% PE group, Fig. 9, falls at around 170 μm, resulting in a total thermal conductivity of 4.4 mW m⁻¹ K⁻¹, which is the lowest in all the samples investigated in this study. Increasing the PE additive to 5% does not reduce the total thermal conductivity further. Both solid/gas conduction and radiation are enhanced in the 5% PE groups when compared to 2% PE group. The enhanced radiation could be explained by the alteration in cell morphology and the enhanced solid/gas conduction is explained by the lower broken cell ratios of the 5% PE groups, as discussed earlier.

4. Conclusions

This study analyzes heat transfer in practical VIP, that is, VIP with a broken cell ratio higher than 90%. The structure of these non-black-body VIP foams consists of struts, closed cells and open cell residue membranes. PE additive is used as a way to alter the foam structure and the heat transfer. Two parameters, namely, the broken cell ratio and the average cell size, are proposed to characterize the structure. The experimental samples are further grouped based on their solid volume fraction to reveal the influence of the solid material on heat transfer. Some conclusions derived from the experimental findings may help improve VIP performance, as summarized below.

1. Under a specific solid volume fraction, a best cell size (best broken cell ratio) leads to the lowest total thermal conductivity.
2. Radiation heat transfer, as manifested by the mean extinction coefficient, is influenced predominantly by broken cell ratio. The effects of solid volume fraction upon radiation are relatively insignificant in the samples investigated in this study. PE2 samples have smaller cell size and therefore higher extinction than PE5 samples.
3. An appropriate amount of PE additive has proven to be effective in tuning the cell structure and improving the VIP performance. The best PE content among the three additive percentages investigated in this study was 2%.
4. Solid volume could affect the absorption coefficient in radiation transfer, but the effects are not obvious because the solid volume fraction is extremely low in this study, and the extinction coefficient is dominated by scattering. However, the solid volume fraction has a crucial effect on solid conduction, which is the dominant heat transfer mechanism in VIP. A rule of thumb to improve VIP permeance can be derived from the findings in this study. Firstly, the solid volume fraction must be kept low to diminish the solid conduction. Secondly, the cell size and broken cell ratio must be carefully controlled to an optimum value to produce the lowest total thermal conductivity. A high broken cell ratio may cause high radiation transfer, and does not necessarily imply low total thermal conductivity. In contrast to conventional closed-cell foam, where a small cell size reduces the heat transfer of trapped gas, the best cell size in practical VIP with high broken cell ratio ranges from 100 to 300 μm. The lowest thermal conductivity obtained in this study reached 4.4 mW m⁻¹ K⁻¹, and was among the best when previously compared to published VIP performance results.

References