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Thermal conductivity of polyurethane foams

Jhy-Wen Wu, Wen-Fa Sung, Hsin-Sen Chu*

Department of Mechanical Engineering, National Chiao Tung University, Hsinchu, Taiwan 30010, Republic of China

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Abstract

The thermal conductivity of polyurethane (PU) foams at gas pressure between 760 torr and 0.014 torr is investigated theoretically and experimentally. Six different cell sizes ranging from 150 to 350 μm of PU foam insulation are used as the samples. Experimental results are also obtained for the spectral extinction coefficient using a Fourier transform infrared spectrometer. The thermal conductivity of PU foams for different cell sizes at 760 torr varies from 33.3 to 34.5 mW/m K, and reduces to 6.82–9.15 mW/m K at a gas pressure of 0.014 torr; the effective thermal conductivity decreases when the cell size becomes smaller. At gas pressure 0.014 torr, radiative heat transfer accounts for approximately 20% of the total heat transfer through PU foams, while solid conduction accounts for the other 80%. © 1999 Elsevier Science Ltd. All rights reserved.

Nomenclature

$e_{b\lambda}$	hemispherical spectral emissive power [W/(m$^2\cdot\mu$m)]
e_b	hemispherical total spectral emissive power [W/(m^2)]
f_v	solid volume fraction
k_c	solid conductivity [W/m K]
k_r	radiative conductivity [W/m K]
k_{eff}	thermal conductivity [W/m K]
L	thickness [m]
N	conduction-to-radiation parameter, $k_c\bar{\sigma}_e/4\sigma T(x)^3$
q^c	conductive heat flux [W/m^2]
q^r	radiative heat flux [W/m^2]
$q_{r\lambda}$	spectral radiative heat flux [W/m^2]
T	temperature [K]
$T_{1,2}$	temperature of hot wall and cold wall [K]
x	coordinate.

Greek symbols

ε	emissivity
σ	Stefan–Boltzmann constant, 5.667×10^{-8} [W (m$^2\cdot$K^4)]
σ_e	extinction coefficient [m^{-1}]
$\bar{\sigma}_e$	Rosseland mean extinction coefficient [m^{-1}].

Subscripts

a	absorb
b	blackbody
c	conduction
e	extinction
eff	effective
w	wall
m	mean
r	radiative
s	scatter
1, 2	cold wall and hot wall.

Superscripts

c	conduction
r	radiative.

1. Introduction

Because of worsening world-wide energy problems thermal insulation is indeed one of the major concerns in the development of heat-transfer technology. Instead of air, foams polymeric foams are the most efficient thermal insulation systems because they contain CFC gas, which is trapped in the closed-cell structures of the foams during production. As the thermal conductivity of air is approximately three times higher than that of CFC gas, and at least 50% of the heat is transferred by conduction

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* Corresponding author. Tel.: +886 3571212155115; fax: +886 35727930; e-mail: hschu@cc.nctu.edu.tw

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through the gas, it is advantageous to have a low-conductivity gas, such as (R-11), inside the foam. Increasing concern over the threat of ozone depletion and global warming, led to CFCs being phased out in 1996 (2010 for developing countries). Without these low thermal conductivity blowing agents, the performance of foam insulation will decrease. Alternative blowing agents, though less detrimental to the environment, result in higher thermal conductivity foam insulation. These factors provide strong incentives to develop more efficient insulation materials with reduced environmental impacts, and with performances that exceed those of CFC technologies. As a result, methods for developing advanced techniques for thermal insulation have become an important issue.

The main functions of thermal insulation are conservation of energy, control of temperature, and control of heat transfer. Among the numerical and experimental studies on this topic: Ostrogorsky et al. [1] discussed the degradation of foam thermal properties due to the diffusion of the air into the foam and developed an analytical model to predict the effective diffusion coefficient of the foam. Glicksman et al. [2] concentrated on the radiative contribution to heat transfer foam. Foams scatter radiation due to the interaction with struts/walls such that radiative transfer may be modeled as a diffusion process. Kuhn et al. [3] presented a detailed investigation of a polystyrene (PS) and a polyurethane (PU) foam, and the separation of thermal-transfer modes. In particular they studied radiative transfer theoretically and experimentally. Doermann and Sacadura [4] presented a predictive model for thermal transfer in open-cell foam insulation as a function of foam morphology, porosity, thermal properties of solid and gaseous phases, and optical properties of the solid phase. Caps et al. [5] measured the thermal conductivity of polymide foams in the temperature range 173–323 K for various gas pressures and types (CO_2 and Ar). They also established a quantitative model for predicting the thermal conductivity of polymide foam as a function of density, gas pressure and temperature. Tseng et al. [6] investigated theoretically and experimentally the thermal conductivity of the polyurethane foam in the temperature range between 300 and 20 K for the development of liquid hydrogen storage tanks. In general, energy is transferred through foam insulation by conduction through the solid polymer making up the cell structure, by conduction and radiation through the gas within the cells. Natural convection in such a system can be ignored due to the small pore size, and modified Rayleigh numbers are far below the critical values. The low thermal conductivity can be achieved in evacuated systems because the gaseous conductive and convective heat transfer modes are not present in vacuum condition. Therefore, the dominant heat transfer modes in foam insulation are thermal radiation and solid–solid conduction when the foam insulation system is evacuated to low pressure conditions. Typically, to achieve high insulating performance with a porous medium, it is necessary that an internal panel pressure on the order of 0.1 Torr be maintained for the duration of the intended application.

This work reports on a theoretical and experimental study of the thermal performance of an evacuated polyurethane (PU) foam insulation system. Experimental measurements were carried out on six different PU foams samples with cell sizes in the range of 150–350 μm after removal of the moisture and volatile materials from the PU foams. The PU foams were put into laminated film bags and evacuated down to pressures in the range of 760 torr to 0.014 torr. To identify the contribution of radiative heat transfer, a Fourier Transform Infrared Spectrometer was used to measure the direct transmittance of these samples in the wavelength range 2.5–25 μm. The extinction coefficient was obtained from the transmittance data using Beer's law. Diffusion approximation was used to estimate the radiative thermal conductivity. As solid and radiative contributions are independent of gas pressure, gas conduction at higher pressure was obtained by subtracting the measured results from the total heat transfer.

2. Theoretical analysis

The one-dimensional, steady-state energy equation for a participating medium with no internal heat generation is [7]:

$$-\nabla\cdot(q^{\mathrm{r}}+q^{\mathrm{c}}) = -\left(\frac{\partial q^{\mathrm{r}}}{\partial x}\right)+\frac{\partial T}{\partial x}\left(k_{\mathrm{c}}\frac{\partial T}{\partial x}\right) = 0. \tag{1}$$

For an optically thick medium in which radiation can be treated as a diffusion process, the radiant heat transfer is simply:

$$q^{\mathrm{r}} = -\frac{4}{3\bar{\sigma}_{\mathrm{e}}}\nabla e_{\mathrm{b}} = -k_{\mathrm{r}}(x)\nabla T. \tag{2}$$

Note that the Rosseland mean extinction coefficient, $\bar{\sigma}_{\mathrm{e}}$, for an optically thick medium is used instead of the spectral coefficient, $\sigma_{\mathrm{e}\lambda}$:

$$\frac{1}{\bar{\sigma}_{\mathrm{e}}} = \int_0^\infty \frac{1}{\sigma_{\mathrm{e}\lambda}}\frac{\partial e_{\mathrm{b}\lambda}}{\partial e_{\mathrm{b}}}\partial\lambda. \tag{3}$$

The radiative conductivity, k_{r}, is defined as

$$k_{\mathrm{r}}(x) = \frac{16\sigma T(x)^3}{3\bar{\sigma}_{\mathrm{e}}}. \tag{4}$$

The energy equation is

$$-\nabla\cdot(q^{\mathrm{r}}+q^{\mathrm{c}}) = \frac{\partial}{\partial x}\left[(k_{\mathrm{r}}(T)+k_{\mathrm{c}})\frac{\partial T}{\partial x}\right] = 0. \tag{5}$$

Equation (5) can be solved easily using the control volume method, which was developed by Patankar [8], even though k_r is a complicated function of temperature. The slip boundary conditions, representing effective, linearized, combined diffusion–radiation and conduction boundary conditions [9], are described by the following relationships

$$\left(\frac{1}{\varepsilon_1}-\frac{1}{2}\right)\left(\frac{1}{1+3N_1/4}\right)=\frac{4\sigma T^3_{\mathrm{w},x=0}[T_\mathrm{w}(0)-T(0)]}{q^\mathrm{r}(0)} \tag{6a}$$

and

$$\left(\frac{1}{\varepsilon_2}-\frac{1}{2}\right)\left(\frac{1}{1+3N_2/4}\right)=\frac{4\sigma T^3_{\mathrm{w},x=L}[T(L)-T_\mathrm{w}(L)]}{q^\mathrm{r}(L)}, \tag{6b}$$

where

$$N_{1,2}=k_\mathrm{c}\bar{\sigma}_\mathrm{e}/4\sigma_\mathrm{e}T(x)^3_{x=0,x=L}=4k_\mathrm{c}/3k_\mathrm{r}. \tag{7}$$

An additional boundary condition on the side of the heater was

$$q(0)=-[k_\mathrm{r}(0)+k_\mathrm{c}]\frac{\partial T(0)}{\partial x}. \tag{8}$$

Equations (5) (6a,b) and (8) were solved using the finite-difference method to determine the solid conductivity, k_c, and temperature distribution. The solution procedure was repeated using the newly obtained temperature distribution and solid conductivity. The convergence criterion in all of the calculation was set at:

$$|T_i^\mathrm{new}-T_i^\mathrm{old}|\leqslant 1\times 10^{-5} \tag{9}$$

to meet all mesh points. This iterative procedure was continued until a steady state was reached.

3. Experiments

3.1. Sample description

The fundamental materials in manufacturing PU foams are isocyanates, polyols, and water. The mixture was forced into boxes (40 × 40 × 8 cm^3) using a high pressure injection molding and allowed to rise freely. In order to obtain better reaction rates, pore sizes, and void fractions, it was necessary to preheat the molds to 313 K and include additives, such as blowing agents, catalysts, and surfactants. Moreover, the evacuation time could be shortened by heating during the evacuation process. By changing the mixture of heating temperature and disturbing rate, we were able to obtain samples with different pore sizes. Because of pressure injection, the density near the mold was 2–3 times greater than that near the center during the blowing process, 3 mm edges had to be cut out from all. The samples were cut to 25 × 25 × 1 cm^3, each with a density of about 60 g cm^3 as calculated using the ASTM D-1622 method. The solid volume fraction was 0.042. Foams were completely open cells, and Fig. 1 shows and SEM picture of an open-cell foam. After scanning and accounting for the effective pore-diameter area using image analysis software, mean pore diameters were obtained. The cell sizes for the six samples were from 147–341 μm.

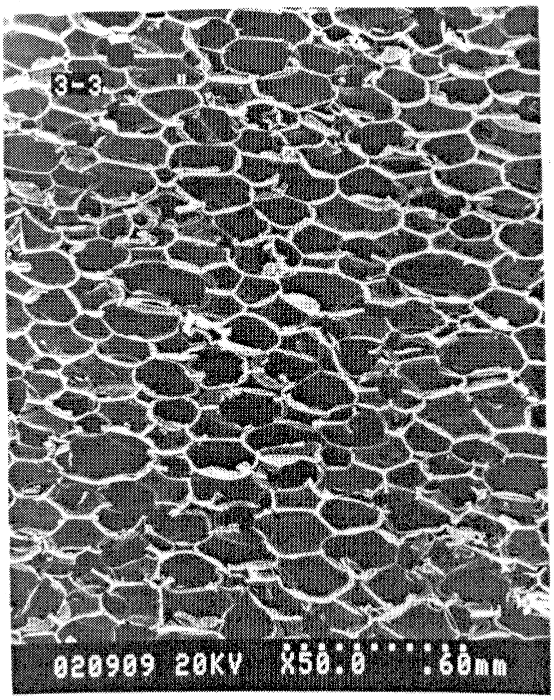

Fig. 1. SEM of an open-cell foam.

3.2. Radiative properties

Radiative conductivity depends on the spectral extinction coefficient $\sigma_{e\lambda}$. Measurements of hemispherical transmission were performed in the wavelength range 2.5–25 μm using a Fourier transform infrared spectrometer (Perkin–Elmer Spectrum 2000). The six samples were measured after removal of moisture and volatiles. Figure 2 shows the transmission of Sample A measurement results for four different thicknesses. As expected, the transmission decreased as media thickness was increased. The transmission data were used to calculate the spectral extinction coefficient σ_λ, by means of Beer's law, $I_\lambda(x)/I_{0\lambda}=\exp(-\sigma_{e\lambda}x)$. The relationship between the extinction coefficient and wavelength for Sample A are shown in Fig. 3. The variation was small; however, the spectral extinction coefficient was large enough for the sample to be treated as an optically thick medium for which radiation could be considered a diffusion process.

3.3. Measurement of thermal conductivity

After the foam was ready, the foam and some getter was put into laminated film bags. The amount and

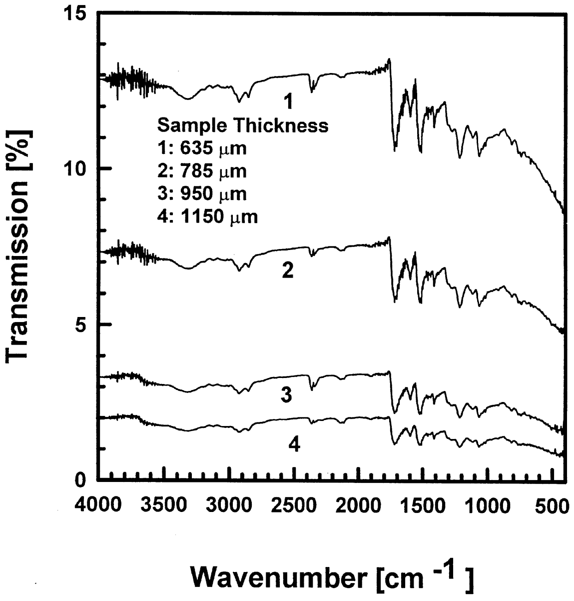

Fig. 2. The measurement of transmission in Sample A.

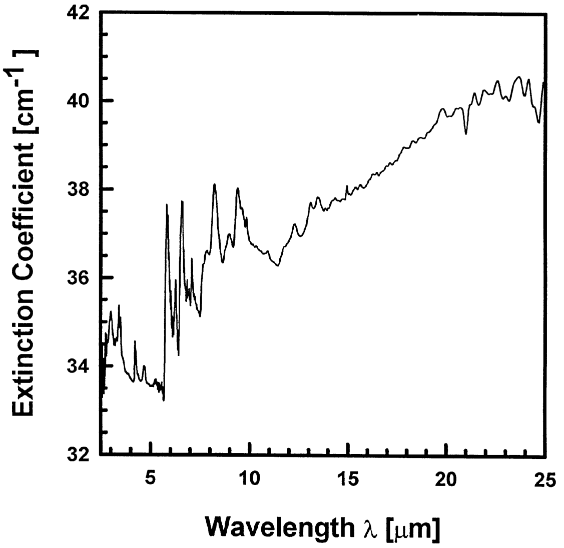

Fig. 3. The relationship between extinction coefficient and wavelength in Sample A.

location of getter that is used to absorb the volatile materials, does not affect the total heat transfer. The laminated film bags, which have low permeability were manufactured by the Japanese company, Takeda. The laminated film bags containing the foams were evacuated to various low pressures, then vacuum packaging machines were used to seal the bags. The device used to measure the effective thermal conductivities of the evacuated foam panels at various evacuation pressures was 'the effective thermal conductivity measuring system' made by EKO (model HC-072). This apparatus consists of thin-film heat-flow meter, hot and cold plates and thickness measurement sensor. The thermocouples were mounted on the center of hot and cold plates to measure the temperature. The measurement theory of the device can be described as

$$k_{\mathrm{eff}} = \frac{E \cdot L}{S \cdot \Delta T}, \tag{10}$$

where E is the output of a heat-flow meter, L is the thickness of the evacuated sample panel, S is the sensitivity of heat-flow meter, and ΔT is the temperature difference between hot and cold side of panel. The measuring uncertainty of this device can be deduced from its definition of effective thermal conductivity:

$$k_{\mathrm{eff}} = q \times \frac{L}{\Delta T}. \tag{11}$$

We applied the relative-uncertainty method to calculate the uncertainty U [10] of the effective thermal conductivity we measured, took Sample A as an example, and denoted it as follows:

$$\begin{aligned} \delta k &= \left[\left(\frac{k_{\mathrm{eff}}}{q}\right)^2 \delta q^2 + \left(\frac{k_{\mathrm{eff}}}{L}\right)^2 \delta L^2 + \left(\frac{k_{\mathrm{eff}}}{\Delta T}\right)^2 \delta \Delta T^2 \right]^{0.5} \\ &= \left[\left(\frac{0.034}{102}\right)^2 (0.02)^2 + \left(\frac{0.034}{0.01}\right)^2 (0.0005)^2 \right. \\ &\qquad \left. + \left(\frac{0.034}{30}\right)^2 (0.1)^2 \right]^{0.5} \\ &= 0.017 \ (\mathrm{W/m\,K}). \end{aligned} \tag{12}$$

So, the effective thermal conductivity uncertainty can be derived from equation (12)

$$U = \frac{\delta k}{k_{\mathrm{eff}}} = \frac{0.017}{0.034} = 0.05. \tag{13}$$

That is, the uncertainty of measuring specimen effective thermal conductivity can be controlled to within 5%.

4. Results and discussion

Experiments were conducted with six samples at a mean temperature of 286 K, as indicated in Table 1. The temperature of heater is higher than the edge of foam because of slip condition [9], and the cold plate temperature is lower. Thus, the temperature differences and the heat fluxes near the centers were greater than the average values. Using these higher temperature differences and the total (average) heat flux (from the heater), yields in k_{eff} (using equation (11)) were smaller than the k_{eff} obtained via one-dimensional analysis. Experimental values of effective thermal conductivity, at a pressure of 0.014 torr, reached 6–9 (10^{-3} W/m K), which means gas

Table 1
Summary of experimental data for six samples at mean temperature 286 K

Parameter/sample		A	B	C	D	E	F
	f_v	0.037	0.041	0.043	0.042	0.038	0.029
	Cell size (μm)	330	341	212	147	214	157
	$\bar{\sigma}_e$ (1/m)	3703	3335	6992	8674	5828	8636
	k_r (mW/m K)	1.91	2.12	1.01	0.82	1.22	0.82
760 torr	k_c (mW/m K)	32.4	32.4	32.5	32.7	32.9	32.5
	k_{eff} (k_r+k_c)	34.3	34.5	33.5	33.5	34.1	33.3
	k_{eff} (measurement, mW/m K)	34	34.2	33.4	33.4	33.9	33.2
	k_r (mW/m K)	1.91	2.12	1.01	0.82	1.22	0.82
0.014 torr	k_c (mW/m K)	7.04	7.03	6.33	6.40	6.76	5.99
	k_{eff} (k_r+k_c)	8.95	9.15	7.35	7.22	7.97	6.82
	k_{eff} (measurement, mW/m K)	8.7	9.0	7.2	7.1	7.8	6.7

thermal conductivity accounted for about 70–80% of k_{eff}. Taking Sample A as an example, if we assume no gaseous heat transfer when the evacuation pressure was 0.014 torr, then the relative contributions of solid, gaseous and radiative modes to total heat transfer at an evacuation pressure of 760 torr should be 19.5, 74.4 and 6.1%, respectively. As the evacuation pressure is decreased, the relative contribution of radiative increases, finally contributing about 25.5% of k_{eff}.

The total conductivity, k_{eff}, of the PU closed-cell foam is about two-thirds of the conductivity of stagnant air because there is low conductivity gas (R-11) inside the foam. The closed-cells age as air components diffuse into the foam, and the R-11 vapor diffuses out. Open-cell foams should have similar problems. Figure 4 shows the relationship between thermal conductivity and heating time at a pressure of 0.1 torr and a preheating temperature of 393 K. It is important to heat samples before packing them into the laminated film bags because moisture and volatile materials increase thermal conductivity, as shown in Fig. 4, the thermal conductivity value without heating is almost two times larger than that with 2 min of heating. Actually, there are two advantages to heating the foam: (1) it shortens the evacuation time because higher temperatures are associated with higher molecular energies and more frequent molecular collisions; (2) it reduces thermal conductivity. The relationship between thermal conductivity and heating temperature at a pressure of 0.1 torr and a heating time of 10 min is shown in Fig. 5. As expected, higher temperature treatments

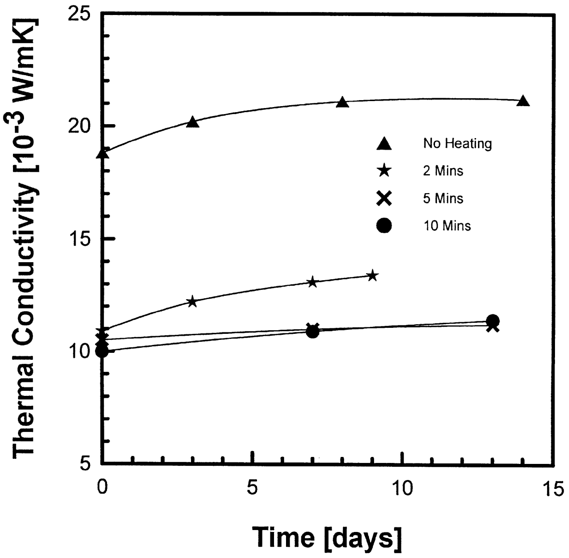

Fig. 4. The relationship between thermal conductivity and heating time.

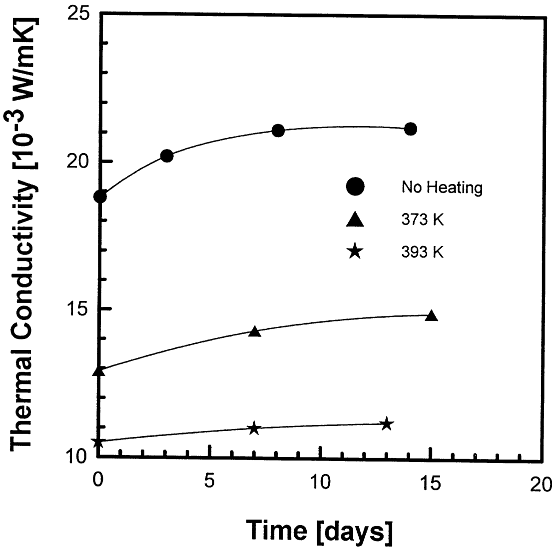

Fig. 5. The relationship between thermal conductivity and heating temperature.

yielded better performances. The melting point of the laminated film bags is about 400 K, so they cannot be heated to that temperature. However, heating to 393 K apparently reduces thermal conductivity to about one-half that of obtained with no heating. Developing optimal processing techniques is, therefore, a very important issue.

Figure 6 shows the relationship between the Rosseland mean extinction coefficient ($\bar{\sigma}_e$) and cell size at a temperature of 286 K. The cell size range is from 147–340 μm. The variation of $\bar{\sigma}_e$ with cell size is rather remarkable. Foams with small cell sizes had larger $\bar{\sigma}_e$ values. In general, the struts and walls contributed considerably to the attenuation of thermal radiation and the extinction coefficient. Furthermore, the foams with small cell sizes had more pores per unit area, which led to more interactions of radiative heat transfer.

Figure 7 shows the relationship between thermal conductivity and evacuation pressure at a temperature of 286 K. These experiments yielded total or effective thermal conductivity. Energy transfer due to gas conduction can be determined by carrying out the experiments at very low pressures, such that energy transfer due to gas conduction is negligible in comparison with other modes. As can be seen with sample D, between 0.1 torr and 100 torr, the thermal conductivity varied in a wide range between 33 and 12 (10^{-3} W/m K). The thermal conductivity at pressure of 0.014 torr was 7.1 (10^{-3} W/m K), or 21% of the total thermal conductivity at atmospheric pressure. Furthermore, according to gas kinetics, the thermal conductivity of air between the panels does not decrease with evacuation initially, because the mean free path is increased at the same rate as the density is being reduced. If the pore diameter becomes smaller than the mean free path, then air thermal conductivity begins to decrease. Therefore, one threshold vacuum level for achieving the thermal conductivity reduction occurs at about 10 torr. As a result, the critical pressure can be defined as the point at which the thermal conductivity increases markedly due to an increasing gas conductivity contribution.

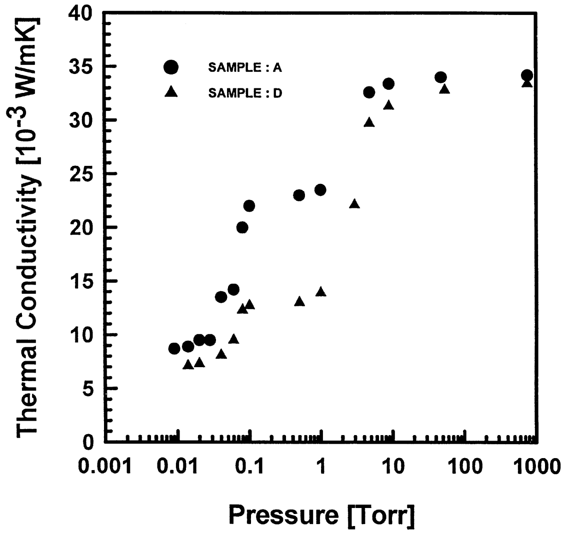

Fig. 7. The relationship between thermal conductivity and vacuum pressure.

Figure 8 shows effective thermal conductivity as a func-

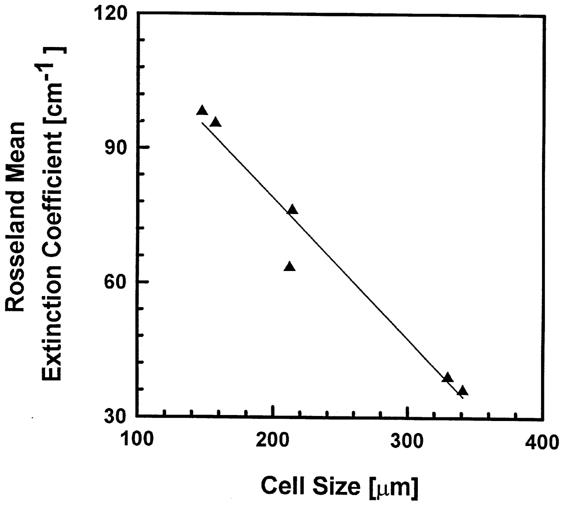

Fig. 6. The relationship between Rosseland mean extinction coefficient and cell size.

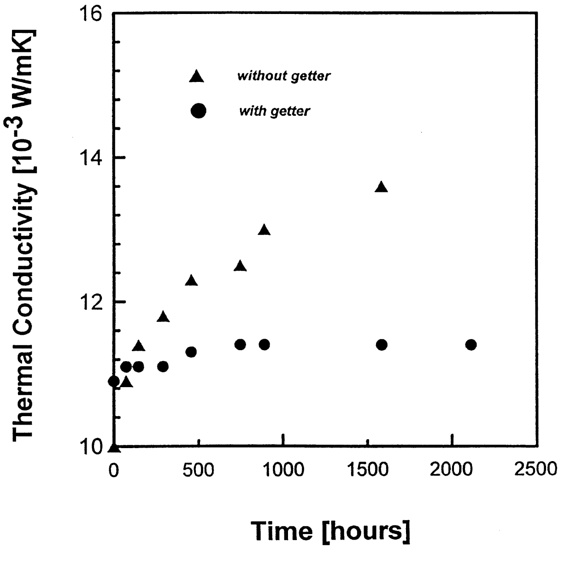

Fig. 8. Endurance test for Sample E foam insulation panel.

tion of time. The performances of PU vacuum insulation panels require not only 100% open-cell foam but also low gas permeability encapsulation films and getters. The laminated film bags were made of one layer aluminum film and two or four layers low gas permeability polymer films. Those films were flexible, thin and low conducting. However, no polymer film currently available can satisfactorily maintain the initial evacuation pressure of a vacuum insulation panel for the duration of intended use. To overcome this pressure increase which will increase thermal conductivity, it is necessary to use getter to absorb any volatiles within the vacuum panels.

5. Conclusions

This work reports on a theoretical and experimental study of the thermal performances of evacuated polyurethane (PU) foam insulation. Experimental measurements of pressures from 760 torr to 0.014 torr, were carried on for six PU foam samples with cell sizes in the range of 150–350 μm. The diffusion approximation was used to estimate radiative thermal conductivity. Solid and radiative contributions were found to be independent of pressure, so gaseous conductivity at higher pressures could thus be obtained by subtracting this measured result from the total heat transfer. Effective thermal conductivity experimental values at specimen pressures of 0.014 torr, can reach 6–9 (10^{-3} W/m K), which means gas thermal conductivity accounts for about 70–80% of the total on effective thermal conductivity. To overcome the volatile material pressure increase that leads to increase thermal conductivity, it is necessary to preheat the foam and use getters to absorb any volatiles within the vacuum panel.

Acknowledgements

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