IPPI

Polyolefins Journal, Vol. 6, No. 1 (2019)
DOI: 10.22063/poj.2018.2185.1113

ORIGINAL PAPER

Thermal-insulation performance of low density polyethylene (LDPE) foams: Comparison between two radiation thermal conductivity models

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Received: 30 April 2018, Accepted: 4 August 2018

ABSTRACT

The loss of energy, especially in industrial and residential buildings is one of the main reasons of increased energy consumption. Improving the thermal insulation properties of materials is a fundamental method for reducing the energy losses. Polymeric foams are introduced as materials with excellent thermal insulation properties for this purpose. In the present study, a deep theoretical investigation is performed on the overall thermal conductivity of low-density polyethylene (LDPE) foams. The thermal conductivity by radiation is predicted using two different methods. The most appropriate model is selected through comparing results with experimental data. The results show that the theoretical model has an appropriate agreement with the experimental results. The effects of foam characteristics including foam density, cell size, and cell wall thickness on the overall thermal conductivity are investigated. The results indicate that by decreasing the cell size and increasing the cell wall thickness, the overall thermal conductivity is decreased significantly. Also, there is an optimum foam density in order to achieve the smallest thermal conductivity. The lowest overall thermal conductivity achieved in the studied range is 30 mW/mK at a foam density of 37.5 kg.m^{-3}, cell size of 100 μm, and cell wall thickness of 6 μm. **Polyolefins J (2019) 6: 13-21**

Keywords: LDPE; radiated thermal conductivity; thermal-insulation; polymeric foams.

INTRODUCTION

Nowadays, the application of polymeric components is dramatically increasing in scientific and industrial fields. Therefore, the saving in the consumption of these materials is an environmental requirement, which aims to improve the use of resources. In this regard, one of the effective ways to save consumable polymer materials is foaming i.e. making cellular structures. While reducing the amount of material consumed [1], this method can improve the properties such as mechanical [2, 3], physical [4] and chemical [5] properties as well as sound [6] and thermal [7] insulation.

The development of polymeric foams began in the 1930s with the production of macrocellular polystyrene foams with cell sizes greater than 100 μm [8]. The progress in the field of polymeric foams was continued with the development of solid-state batch foaming, and

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fine-celled foams with cell sizes smaller than 100 µm were introduced in the 1980s [9]. The aim of the production of polymeric foams was to reduce the density of the materials without damaging their properties and to lower the cost of consumable materials.

The international community faces many demands on reducing the consumption of fossil fuels, because these resources are coming to an end and, on the other hand, they release a massive amount of harmful pollutants to the environment. There are two different ways to respond to the aforementioned demands: 1) the development of renewable energies and 2) saving and maintaining energy. Despite the special investments, renewable energies have not been proven profitable as expected because they currently account for almost 15% of total global energy demand [10-12]. Therefore, the development of energy saving methods is a fundamental way for researchers to effectively reduce energy consumption.

Polymeric foams are one of the most commonly used thermal-insulation materials. These materials also have many other advantages in comparison to other thermal-insulation materials. Excellent mechanical properties, easy installation, low cost, etc. are some of the advantages of polymeric foams.

The thermal conductivity behavior in polymeric foams is arisen from four different mechanisms, including thermal conductivity due to the gaseous phase, thermal conductivity due to the solid phase, thermal conductivity by radiation, and thermal convection within the cells [13, 14].

Despite the empirical achievements of researchers in recent years, almost all of the empirically presented studies have not made it possible to investigate the thermal insulating mechanism of polymeric foams, thoroughly and extensively. One of the most important reasons for this shortcoming is the numerous factors affecting the thermal insulation properties of polymeric foams. In addition, these parameters have different effects on different mechanisms associated with the thermal conductivity of polymeric foams. On the other hand, simultaneous control of different influential parameters and taking the interactions of these parameters into account in empirical terms are very difficult and challenging. For instance, Gong et al. [15] produced foam samples with same cell sizes and densities in order to investigate the effect of the addition of multi-walled carbon nanotubes (MWCNTs) on the thermal-insulation properties of polystyrene foams. Elimination of the effect of cell size and foam density was their aim to produce the same samples. Also, Wang et al. [16] produced poly methyl methacrylate (PMMA) foam samples with similar densities to investigate the effect of cell size on the thermal conductivity. Some experimental efforts have been made on thermal-insulation of polymeric foams field. Zhao et al. [17] studied the thermal-insulation performance of polypropylene foams produced by high-pressure foam injection molding and mold opening. Experimental investigation on the thermal conductivity of polyurethane foams was conducted by Zhang et al. [18]. They studied the effect of temperature, humidity and water uptake on the thermal conductivity. Zhao et al. [19] developed high thermal-insulation branched polypropylene (BPP) containing polytetrafluoroethylene (PTFE) nanofiber and obtained a thermal conductivity value as low as 32.4 mW/mK. It is necessary to mention that the overall thermal conductivity is measurable experimentally, but measuring different mechanisms is not feasible or it needs special measuring methods such as thermal conductivity measurement in vacuum conditions [20, 21]. For these reasons, a theoretical model to estimate the thermal conductivity of polymeric foams seems to be necessary.

So far, some theoretical models have been presented to measure the thermal conductivity of polymeric foams. The gaseous and solid thermal conductivities are easily and precisely predictable, but the radiated thermal conductivity, especially in theory, still faces serious challenges. To this point, several efforts have been made to predict the thermal conductivity by radiation. Rosseland model [22] and Williams and Aldao model [23] are two most well-known models for estimating the thermal conductivity by radiation. The first goal of this study is to compare the accuracy of the results obtained by these two theoretical methods and by using experimental results achieved for low-density polyethylene (LDPE) foams. After ensuring the accuracy of theoretical results, the best model will be selected. Comprehensive investigation of the effect of structural properties on the thermal conductivity of LDPE foams is another aim of the present research.

THEORETICAL PROCEDURE

The overall thermal conductivity (λ_t) is consisted of four different mechanisms including thermal conductivity through gaseous phase (λ_g), the thermal conductivity through the cell walls and struts of solid phase (λ_s), the radiation thermal conduction (λ_r), and thermal convection within the cells (λ_c). It should be noted that the thermal convection is neglected due to the small cell sizes [24, 25]. The overall thermal conductivity can be expressed by a superposition of each mechanism taken separately as Equation (1).

$$\lambda_t = \lambda_g + \lambda_s + \lambda_r \tag{1}$$

The thermal conductivity due to the gaseous and solid phases can be obtained using Equations 2 and 3, respectively [26, 27].

$$\lambda_g = V_{gas}\lambda_{gas}\left(\frac{1}{1+2\beta K_n}\right) \tag{2}$$

$$\lambda_s = V_{solid}\lambda_{solid}\left(\frac{2}{3}-\frac{f_s}{3}\right) \tag{3}$$

where λ_{gas} and λ_{solid}, respectively, are the thermal conductivities of air (0.0263 W/mK at ambient conditions) and base polymer which is here LDPE. Also, V_{gas} and V_{solid} are the volume fractions of gaseous and solid phases which can be obtained as:

$$V_{gas} = 1 - \frac{\rho_f}{\rho_s} \tag{4}$$

$$V_{solid} = \frac{\rho_f}{\rho_s} \tag{5}$$

where ρ_f and ρ_s are the foam and solid densities, respectively.

In Equation 2, β is the efficiency of the energy transfer between gas molecules and cell walls and is equal to 2 for polymeric foams [28]. Also, K_n is the Knudsen number and can be calculated using Equation 6 [20]:

$$K_n = \frac{l_{mean}}{\varphi_c} \tag{6}$$

where l_{mean} is the mean free path of gas molecules and is equal to 70 nm for air at ambient conditions and also, φ_c is the cell size [20].

In Equation 3, f_s is the struts fraction and can be calculated using Equation 7.

$$f_s = \frac{V_{struts}}{V_{struts}+V_{walls}} \tag{7}$$

where V_{struts} and V_{walls} are the volumetric fraction of struts and cell walls, respectively, and can be obtained using Equations 8 and 9 [29]:

$$V_{struts} = 2.805\varphi_s^2\varphi_c \tag{8}$$

$$\begin{aligned} V_{walls} = {} & [1.317\varphi_c^2 - 13.4284\varphi_s\varphi_c + 34.2374\varphi_s^2].d_w \\ & + 4.639\varphi_c d_w^2 - 17.976\varphi_c d_w^2 \end{aligned} \tag{9}$$

where φ_s and d_w are the strut diameter and cell wall thickness, respectively.

This procedure is a well-known method for estimating the gas and solid conductivities and the literature review shows that it has an acceptable estimation in comparison to experimental results. But the radiation thermal conductivity is still facing challenges. There are some theoretical approaches in order to estimate this quantity. There are two methods that are most used: Rosseland [22] and Williams and Aldao [23].

Rosseland Equation is expressed as Equation 10 [22]:

$$\lambda_r = \frac{16n^2\sigma T^3}{3K_R} \tag{10}$$

where n is the effective index of refraction and is close to 1 for polymeric foams. T is the mean temperature (300 K at ambient conditions) and is the Stefan-Boltzmann constant and is equal to 5.67×10^{-8} $W.m^{-2}.K^{-4}$. K_R is the Rosseland mean extinction coefficient and can be approximately estimated using the following Equation [30]:

$$K_R = K_{structs} + K_{walls}K_{solid} \tag{11}$$

where $K_{structs}$ and K_{walls} are the extinction coefficient due to the struts and walls, respectively. K_{solid} is the extinction coefficient of the solid polymer. For instance, this value is equal to 80 cm^{-1} [26] and 140 cm^{-1} [27] for polystyrene and polyethylene, respectively. K_{struts} and K_{walls} can be obtained as follows [29]:

$$K_{struts} = 4.10\frac{\sqrt{f_s\frac{\rho_f}{\rho_s}}}{\varphi_c} \tag{12}$$

$$K_{walls} = (1-f_s)\frac{\rho_f}{\rho_s} \tag{13}$$

Williams and Aldao model is as Equation 14 [23]:

$$\lambda_r = \frac{4\sigma T^3 L}{1+(\frac{L}{\varphi_c})(\frac{1}{T_N}-1)} \tag{14}$$

where L is the sample thickness, and T_N is the net fraction of radiant energy sent forward by a solid membrane of thickness d_w, which this fraction of energy is given by:

$$T_N = \frac{(1-r)}{(1-rt)}\left\{\frac{(1-r)t}{(1+rt)}+\frac{(1-t)}{2}\right\} \tag{15}$$

where r is the fraction of incident energy reflected by each gas–solid interface and t is the fraction of energy transmitted through the solid membrane of thickness dw. These quantities can be calculated using Equations 16 and 17:

$$r = \left\{\frac{\omega-1}{\omega+1}\right\}^2 \tag{16}$$

$$t = \exp(-a\cdot d_w) \tag{17}$$

where ω and a are the refractive index and absorption coefficient of the solid matrix, respectively.

RESULTS AND DISCUSSION

Validation of theoretical approaches

In order to validate the theoretical results, we collected experimental data from different researches. Table 1 shows the structural properties of various low-density polyethylene (LDPE) foams. The theoretical and experimental results and the difference between them are presented in Table 2. First model is a theoretical model in which the thermal conductivity by radiation is obtained using Rosseland model (Equation 10). Also, model No. 2 is a theoretical model in which the thermal conductivity by radiation is calculated using Williams and Aldao model (Equation 14).

Table 1. Experimental structural properties of LDPE foams.

LDPE Foam	r_f (kg.m^{-3})	j_c (µm)	d_w (µm)	L (cm)
1 [30]	61.0	162	2	1.00
2 [30]	36.9	214	1.5	1.00
3 [30]	25.5	255	1.2	1.00
4 [31]	16.7	313.5	1.4	1.12
5 [31]	22.5	879.7	5.8	0.96
6 [31]	24.6	311.9	1.9	1.02
7 [31]	30.7	528.1	4.2	1.11
8 [31]	32.0	424.4	3.6	1.11
9 [31]	32.5	396.9	2.5	1.08

The results of Table 2 indicate that both models estimate the thermal conductivity with a good difference, especially, model 2 which predicts the thermal conductivity with an error smaller than 10% in all cases. Figure 1 shows the efficiency of both models versus cell size. Both models have a good agreement with experimental results in small cell sizes. By increasing the cell size, the accuracy of model 2 will be more acceptable than that of model 1. Therefore, in the following, model 2 is used for all investigations. Figure 1 shows that the model 1 underestimates the thermal conductivity whereas model 2 overestimates this property in most cases.

Cell size, foam density and cell wall thickness are considered as the variable parameters. The effects of considered parameters on the thermal-insulation per-

Table 2. Validation of theoretical models through comparing results with experimental data.

LDPE Foam	l_t (mW/mK) Experimental	l_t (mW/mK) Model No. 1	Error (%)	l_t (mW/mK) Model No. 2	Error (%)
1 [30]	42.0	39.2	6.7	43.6	-3.8
2 [30]	38.4	36.0	6.3	41.7	-8.6
3 [30]	37.5	35.1	6.4	41.5	-10.7
4 [31]	37.4	36.6	2.1	40.1	-7.2
5 [31]	43.3	44.5	-2.8	43.7	-0.9
6 [31]	37.2	34.5	7.3	39.5	-6.2
7 [31]	44.1	37.0	16.1	41.5	5.9
8 [31]	40.7	36.1	11.3	40.4	0.7
9 [31]	39.8	34.2	14.1	41.0	-3.0

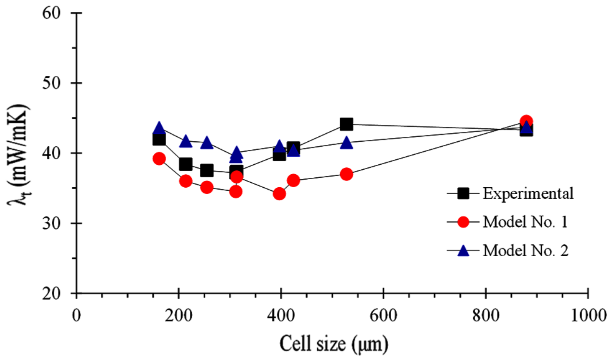

Figure 1. Efficiency of models no. 1 and 2 versus cell size.

formance of LDPE foams are investigated.

It should be noted that all the statistical analyses are performed in Minitab software.

Effect of parameters

In the following, the effects of considered parameters on the overall thermal conductivity are investigated. Figure 2 shows the effect of foam density on the overall thermal conductivity. The results show that there is an optimum foam density in order to achieve the smallest amount of overall thermal conductivity. In other words, the overall thermal conductivity shows a decreasing behavior followed by an increasing behavior with foam density. At low densities, the thermal conductivity by radiation is decreased by increasing the foam density. Therefore, the overall thermal conductivity is decreased by increasing the foam density. In contrast, at higher densities, i.e. higher than the optimum foam density, the solid thermal conductivity is increased by increasing the foam density. Hence, the overall thermal conductivity is increased.

Figure 3 indicates the effect of cell size on the overall thermal conductivity. It is observable that the overall thermal conductivity is decreased significantly by decreasing the cell size. This phenomenon can be explained by two different mechanisms. By decreasing the cell size, on the one hand, the Knudsen number is decreased (see Equation 6), and consequently, the gaseous thermal conductivity is decreased (see Equation 2); although the experimental studies demonstrate that the Knudsen effect is activated in microcellular and nanocellular ranges [15, 20, 32]. On the other hand, λ_r is changed by variation of cell size. This effect is presented in Figure 4. As it can be seen, λ_r is reduced by decreasing the cell size. By reducing the cell size, the number of cell walls and struts are increased. Thus, the radiation is more attenuated.

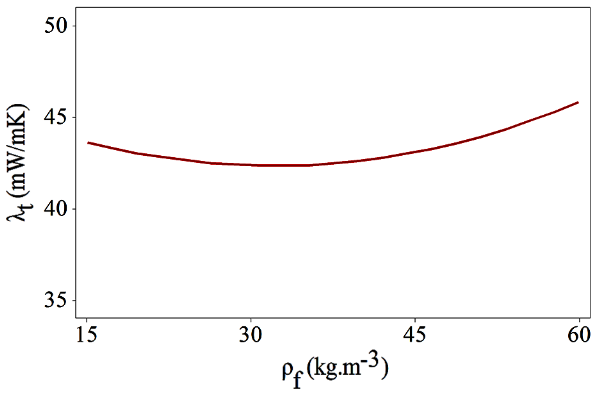

Figure 2. Effect of foam density on the overall thermal conductivity.

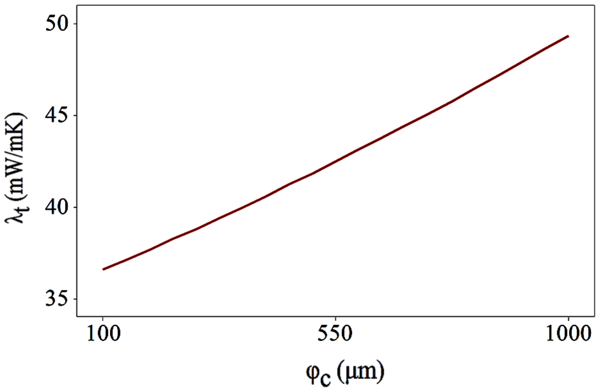

Figure 3. Effect of cell size on the overall thermal conductivity.

The effect of cell wall thickness on the overall thermal conductivity is investigated and presented in Figure 5. As it can be seen, the overall thermal conductivity is significantly reduced by increasing the cell wall thickness in the studied range. By increasing the cell wall thickness, more radiation is attenuated. There-

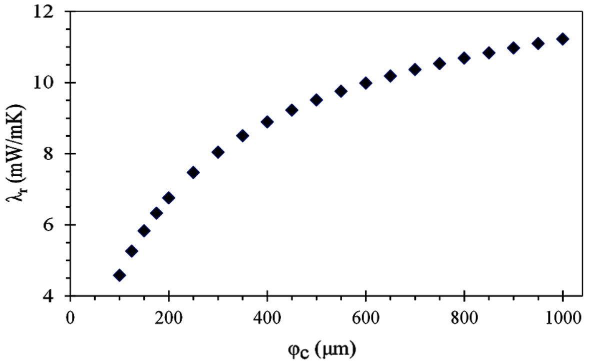

Figure 4. Effect of cell size on the radiated thermal conductivity.

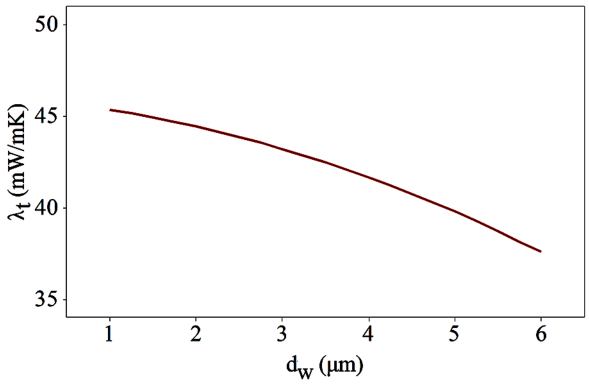

Figure 5. Effect of cell wall thickness on the overall thermal conductivity.

fore, the radiation thermal conductivity is decreased. Hence, the overall thermal conductivity is reduced.

The interaction effect of foam density and cell sizes on the overall thermal conductivity is presented in Figure 6. The results reveal that the maximum overall thermal conductivity is achieved when both cell sizes and foam densities are simultaneously high. The value of thermal conductivity in this condition is higher than 50 mW/mK. Also, it is observable that in all foam densities, the overall thermal conductivity is smaller than 40 mW/mK in cell sizes approximately below 200 µm. Also, an optimum foam density is observable for each cell size in order to achieve the smallest overall thermal conductivity which is in accordance with the previous results (see Figure 2).

Figure 7 shows the interaction effect of foam density and cell wall thickness on the overall thermal conductivity. The results indicate that the minimum overall thermal conductivity is seen at high cell wall thickness for all foam densities. In these conditions, the overall thermal conductivity is smaller than 40 mW/mK. Also, at low cell wall thicknesses and high foam densities, the overall thermal conductivity is higher than 46 mW/mK.

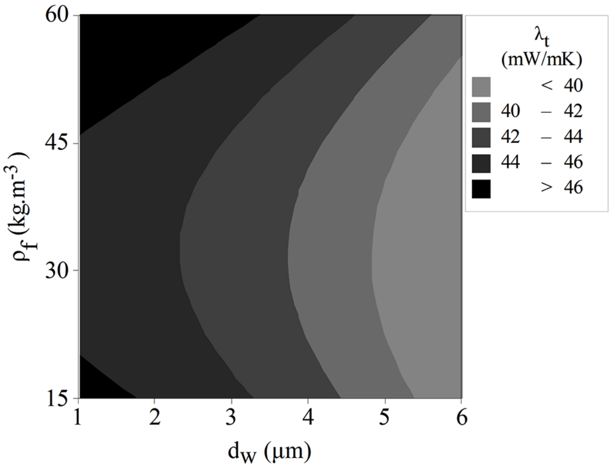

Figure 7. Interaction effect of foam density and cell wall thickness on the overall thermal conductivity at φ_c = 550 µm.

Figure 8 shows the interaction effect of cell size and cell wall thickness on the overall thermal conductivity. The results illustrate that the lowest thermal conductivity i.e. smaller than 35 mW/mK is achieved at high cell wall thicknesses and low cell sizes whereas in the opposite situations i.e. low cell wall thicknesses and high cell sizes, the overall thermal conductivity is higher than 50 mW/mK.

Contribution of heat transfer mechanisms

In the following, the contribution of each heat trans-

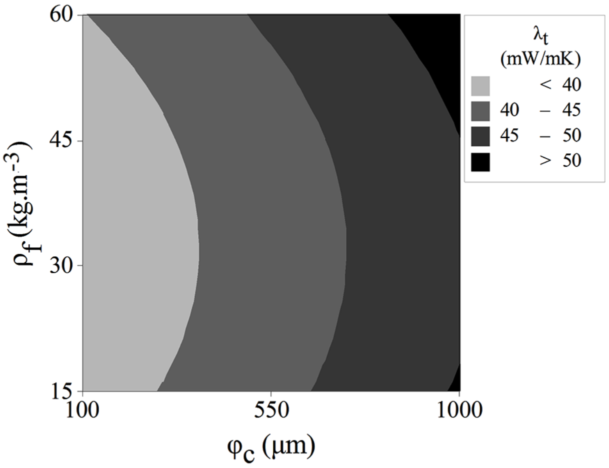

Figure 6. Interaction effect of foam density and cell size on the overall thermal conductivity at d_w =3.5 µm.

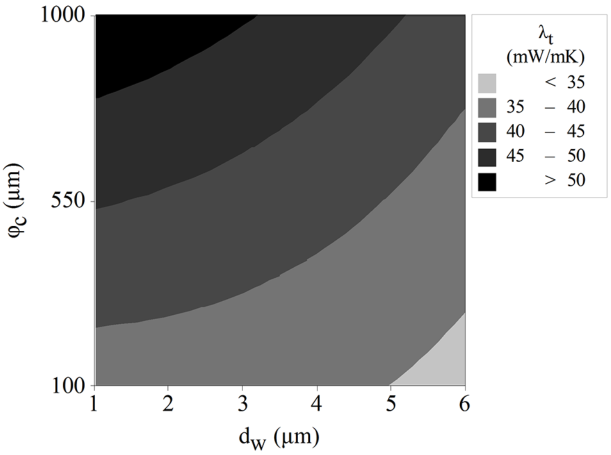

Figure 8. Interaction effect of cell size and cell wall thickness on the overall thermal conductivity at ρ_f =37.5 kg.m^{-3}.

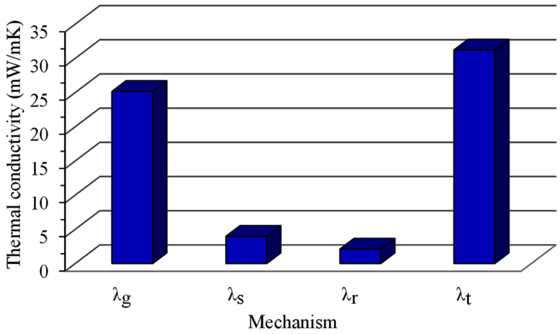

Figure 9. Contribution of each mechanism of heat transfer in the overall thermal conductivity at ρ_f = 37.5 kg.m^{-3}, φ_c = 100 µm, and d_w = 6 µm.

fer mechanism on the overall thermal conductivity is investigated (see Figure 9). The results show that the gaseous thermal conductivity has the most contribution on the overall thermal conductivity. The solid and radiated thermal conductivities are, respectively, the second and the third parameters in terms of their contribution to the overall thermal conductivity. The lowest overall thermal conductivity that could be achieved was almost 30 mW/mK at the conditions of foam density of 37.5 kg.m^{-3}, cell size of 100 µm, and cell wall thickness of 6 µm according to the multi-objective optimization tool of response surface methodology.

CONCLUSION

The overall thermal conductivity of low-density polyethylene is predicted using theoretical models. Two different models are used in order to estimate the thermal conductivity by radiation. The conclusions of this research can be summarized as follows:

- Williams and Aldao model is more accurate than Rosseland model in predicting the thermal conductivity of LDPE foams, especially, at high cell sizes.
- Williams and Aldao model overestimates the radiated thermal conductivity of LDPE in most cases whereas the Rosseland model underestimates it.
- The thermal conductivity is significantly decreased by decreasing cell sizes and increasing cell wall thickness.
- There is an optimum foam density in order to achieve the smallest thermal conductivity.
- 30 mW/mK is the smallest thermal conductivity, which in this study can be obtained in the considered ranges of structural properties.

It should be noted that this study is performed at the macrocellular foam structure state and more deep investigation is needed, especially in microcellular and nanocellular ranges.

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