

**Technical note**

## AN ANALYSIS OF RELATIONS FOR DETERMINING THE THERMAL CONDUCTIVITY OF RIGID POLYMER FOAMS

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In the paper the authors present the effectiveness of the generalized thermal conductivity method for polymer foams, including modelling their geometrical structure. Calculations of the effective thermal conductivity coefficient  $\lambda$  are based on the generally accepted assumption of the additivity of different thermal exchange mechanisms in porous media and this coefficient is presented as a sum the coefficients of conductive  $\lambda_q$ , radiative  $\lambda_p$ , and convective  $\lambda_k$  thermal conductivity. However, in literature not enough attention is given to relations determined by means of the theory of generalized conductivity, including modelling the geometrical structure. This paper presents an analysis of these relations and verifies their ability to predict experimental data in comparison with the best formulae included in the paper [2].

**Key words:** thermal insulation, effective thermal conductivity, rigid polymer foam, mathematical model, structure of used polymer foams, analysis of experimental data.

### 1. Introduction

The problem of reducing expenses for heating residential, public and industrial buildings in a cold and moderately cold climate will probably never lose its importance. At present, in order to solve this problem (at least partially), it is fully justified to use thermal insulation layers of effective polymeric materials with cellular structure in partition walls. The insulating characteristics of thermal insulation layers are usually described by the effective thermal conductivity coefficient, the value of which can be determined

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empirically or computationally from mathematical models describing the structure of a porous material and the heat exchange processes occurring in it.

Numerous publications, for example [1], emphasize the need to use two interrelated research approaches, and to consider their strengths and weaknesses.

When calculating the effective thermal conductivity coefficient  $\lambda$ , it is generally accepted to assume the additivity of various heat exchange mechanisms in porous media and to presents this coefficient as the sum of the coefficients of conductive thermal conductivity  $\lambda_q$ , radiation thermal conductivity  $\lambda_p$  and convective thermal conductivity  $\lambda_k$  as in [13, 14].

According to [1, 2, 3] and other papers, convective heat transfer in polymer foams may be disregarded in most cases because relatively small cell dimensions of the porous structure prevent the formation of a convective flow as a result of the temperature gradient. For polymer foams whose cells are filled with air, the maximum limit cell size is 4 mm [2]. When the pores are filled with freon, the limit value is reduced to 1.5 mm. According to the papers [3, 4] for polyurethane and phenolic foams with density of up to 115 kg/m<sup>3</sup>, the maximum cell size reaches 0.6 mm and it is significantly lower than the above-mentioned limit values. For this reason, it is possible to omit the convective component when determining the thermal conductivity coefficient.

When determining the thermal insulation characteristics of insulation layers made of cellular polymers in protective building partitions, it is also often possible to omit the radiation heat transfer component. According to the paper [3], the contribution of the radiation component to the total heat transport in cellular polymers is only 7-10%. If it is necessary to include the radiation component, it can be determined from the formulas presented, for example, in [1, 2, 3, 5], which to some extent (differently) take into account the average cell size and energy absorption or reflection by the walls of polymer foam cells. However, having considered the results of the calculations carried out according to different models presented in the review [2], it is not possible to recommend one of these formulas as the best one.

If we omit the radiative and convective component of thermal conductivity, then the effective thermal conductivity coefficient will be determined only on the basis of the conductivity of the solid polymer skeleton and the pore substance. In order to calculate the thermal conductivity of this type of a two component system, it is possible to use a number of relations obtained with various methods over the last one hundred years, beginning with Maxwell and Rayleigh [1, 3, 5]. In the review [2], many of these relations, with varying levels of complexity, were considered and analysed, and several that provided the best estimates of experimental data were selected. However, neither in this review nor in other publications was enough attention paid to the relations determined with methods based on the generalized theory of conductivity in combination with the geometric structure models [5] that enable a uniform analysis of the thermal conductivity of foams. In this paper, these relations were analysed and their ability to predict experimental data was verified by comparison with the best models presented in the paper [2].

## 2. Structure of polymer foams and its simplified model

Visualizations of various structures of polymer foams, presented in a number of papers [2, 3, 4, 6], suggest that the rigid polymer skeleton consists of polyhedral cells (bars and membranes) with walls mainly in the form of quadrangles, pentagons and hexagons. It is emphasized [3] that the main part of the polymer consists of straight bars (ribs), having a practically constant cross-section, with a slight increase in thickness at the nodes, which usually occurs as a result of four bars coming together. Schuetz and Gliksman [2] estimated that 80-90% of the polymer is found in the cell bars. This can be confirmed by approximate calculations using experimental data with an average cell size and wall (membrane) thickness of polymer foams presented in the paper [2]. If we assume that the polymer skeleton consists of closed cubical cells and the cross-section of the bars is square, then for polymer foams with an apparent density of 50 kg/m<sup>3</sup>, an average cell size of 410 μm and a membrane thickness of 0.4 μm [2], 86% of the polymer is in the bars. At the same time, the length of the cross-section of the bar is 60 times greater than the thickness of the membrane. As the apparent density of the polymer foam increases, the thickness of the membranes increases as well and the amount of polymer deposited in the cell bars decreases.

Since the continuity of the membranes is often interrupted, a large number of cells are open and their pore space is connected. Depending on the type of polymer and the technology used to turn it into foam, the content of open cells varies considerably. According to the paper [3], the structure of urea foams obtained with the air-mechanical method is formed almost exclusively of open cells, the content of which reaches 98%. The structure of phenolic foams is also formed mainly of open cells. A lower content of open cells is found in polyurethane foams.

The polymer skeleton degrades due to environmental factors. In the case of the thermal insulation of buildings, the degradation of polymers occurs mainly as a result of the joint interaction between the cyclically changing temperature and humidity. Hydrothermal degradation results mainly from the plasticization and swelling of the polymer skeleton, the formation of micro-pores and surface micro-cracks [7, 8]. As can be seen from [9, 10], micro-cracks with a width of  $0.01$  to  $0.1\mu\text{m}$  appear in polymeric materials. Over time, the concentration of micro-cracks increases, and when it reaches a certain critical value, macro-cracks begin to appear, the width of which is  $100\mu\text{m}$  or more, and it significantly exceeds the thickness of the membranes and even the cell bars. In addition, the distance between macro-cracks can be almost the same as their width. This type of damage reduces the number of closed polymer foam cells, increases their sorption capacity, accelerates moisture diffusion and increases effective thermal conductivity. The data presented in [10] show that the thermal conductivity of polyurethane foam samples with initial humidity not exceeding the maximum hygroscopic humidity increased on average 1.7 times after they were subjected to 160 cycles of freezing and thawing.

Therefore, it can be said that the structure of used polymer foams consists mainly of open cells and should be regarded as a structure with mutually interpenetrating, geometrically equivalent components [5]. A characteristic feature of this structure is the uninterrupted continuity of any component in all directions. When modelling this kind of structure, it is possible to use the most developed method of transition to the elementary cell. In this case, the actual structure of the material with interpenetrating elements, which is considered to be isotropic, is replaced with an adequate model with an ordered structure consisting of identical elementary cells. For polymer foams, the simplest but fully justified shape of the elementary cell seems to be a cube. The symmetry of the elementary cell in two mutually perpendicular directions makes it possible to describe the process under research in  $\frac{l}{2}$ ,  $\frac{l}{4}$  and  $\frac{l}{8}$  part of the cell.

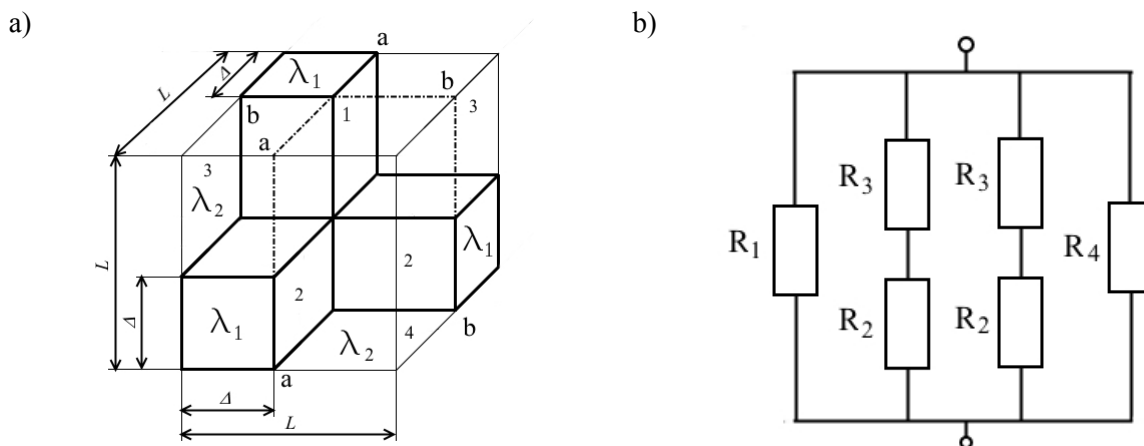


Fig.1. An eighth part of the elementary cell (a) and a diagram of the resistance of its different parts (b).

Figure 1 shows an eighth part of the elementary cell from the simplest structural model with interpenetrating components with bars having a constant cross section. On the basis of this figure, it is easy to determine the relationship between the geometric parameters of the model and the volumetric concentration of the components.

The volume of the first component is

$$V_1 = 3 \cdot \Delta^2 \cdot (L - \Delta) + \Delta^3 = 3 \cdot \Delta^2 \cdot L - 2 \cdot \Delta^3. \quad (2.1)$$

After dividing this expression by the cell volume  $V = L^3$ , we determine the volumetric concentration of the first component

$$m_1 = V_1 / V = c^2 \cdot (3 - 2 \cdot c) \quad (2.2)$$

where  $c = \Delta / L$  - the absolute dimension of the bar. By definition, the concentration of the second component is

$$m_2 = 1 - m_1. \quad (2.3)$$

In order to calculate the parameter  $c$ , we will use the positive root of the cubic Eq.(2.2), which produces physically justified results and has the form

$$c = 0.5 + \sin \left[ \frac{\arcsin(2 \cdot m_1 - 1)}{3} \right]. \quad (2.4)$$

The same result is obtained when the other, less convenient positive root of Eq.(2.2), shown in the paper (2.5), is used in the calculations.

### 3. Relations for determining the effective thermal conductivity coefficient and their experimental verification

During analysis of heat transfer through the elementary cell, different ways of dividing the cell into parts are used. The paper [5], presents three possible methods of dividing cells: through adiabatic or isothermal planes, and combined decomposition through adiabatic and isothermal planes. Each decomposition method has its own scheme for connecting the resistances that are encountered by the thermal flux as well as the corresponding relation for determining the effective thermal conductivity. The thermal resistance of individual parts of the elementary cell is calculated from the formula [5]

$$R_i = \frac{l_i}{\lambda_i \cdot S_i} \quad (3.1)$$

where  $l_i$  and  $S_i$  - the length and the area of the cross-section of the heat flux of the  $i^{\text{th}}$  part;  $\lambda_i$  - the thermal conductivity coefficient of the  $i^{\text{th}}$  part.

Let us repeat the derivation of the formula obtained by G.N. Dulnev in 1965, used to determine the effective thermal conductivity of the elementary cell shown in Fig.1a, divided into parts 1, 2, 3 and 4 with adiabatic planes a-a and b-b, parallel to the general direction of the heat flux. According to the formula (5), the thermal resistance of these parts is as follows

$$R_1 = \frac{L}{\lambda_1 \cdot \Delta^2}, \quad R_2 = \frac{\Delta}{\lambda_1 \cdot \Delta \cdot (L - \Delta)}, \quad R_3 = \frac{L - \Delta}{\lambda_2 \cdot \Delta \cdot (L - \Delta)}, \quad R_4 = \frac{L}{\lambda_2 \cdot (L - \Delta)^2}. \quad (3.2)$$

On the basis of the diagram of the thermal resistance position shown in Fig.1b, the total thermal resistance  $R = L/(\lambda \cdot L^2) = 1/(\lambda \cdot L)$  of a cell with the volume  $V = L^3$ , filled with a homogeneous substance with the effective thermal conductivity  $\lambda$  can be represented as follows

$$\frac{1}{R} = \frac{1}{R_1} + \frac{1}{R_2 + R_3} + \frac{1}{R_4}. \quad (3.3)$$

From the relation (3.3), the formula (3.2) and after some simple transformations, we obtain an expression for determining the relative effective thermal conductivity of a structure with interpenetrating components when the elementary cell is divided with adiabatic planes

$$\frac{\lambda}{\lambda_1} = c^2 + v \cdot (1-c)^2 + 2 \cdot v \cdot c \cdot (1-c) \cdot (v \cdot c + 1-c)^{-1} \quad (3.4)$$

where  $c$  is calculated from the formula (2.4), proposed by the authors of this paper, wherein  $v = \lambda_2 / \lambda_1$ . In 1935 Van-Frey divided the elementary cell (Fig.1a) with isothermal planes and obtained the relation

$$\frac{\lambda}{\lambda_1} = \left[ \frac{1-c}{c^2 + v \cdot (1-c^2)} + \frac{c}{c \cdot (2-c) + v \cdot (1-c)^2} \right]^{-1}. \quad (3.5)$$

In 1977 Dulnev and Novikov used the method of combined division of the same cell with adiabatic and isothermal planes and obtained the following relation [5]

$$\frac{\lambda}{\lambda_1} = \frac{c^2 + v \cdot c \cdot (1-c)}{v \cdot c \cdot (1-c) + (1-c + c^2)} + v \cdot \frac{c \cdot (1-c) + v \cdot (1-c)^2}{c \cdot (1-c) + v \cdot (1-c + c^2)}. \quad (3.6)$$

Since both components in the structure with mutually interpenetrating components are equal, it does not matter which of the two is adopted first for calculations using the relations (3.4), (3.5) and (3.6).

In a moist polymer foam, the liquid may completely or partially wet the polymer skeleton. In the first case, the liquid and the vapour-gas mixture form a structure with inter-penetrating components. Then, one of the three relations presented here is enough to calculate the effective thermal conductivity of the entire material. In the other case, isolated liquid drops (small or large) can be regarded as an inclusion in the vapour-gas mixture. It is emphasized [2, 5] that the shape of the inclusions does not significantly affect the effective thermal conductivity. Therefore, for the sake of simplicity, it is advisable to assume that the inclusions are cubical. Then, the elementary cell of the simplest structure model with isolated inclusions will have the form of a cube in a cube and will remain symmetrical in two mutually perpendicular directions. Division of this kind of cell with adiabatic planes leads to the formula [5]

$$\frac{\lambda}{\lambda_1} = \frac{v - (v-1) \cdot (1 - m_2^{2/3}) \cdot m_2^{1/3}}{v - m_2^{1/3} \cdot (v-1)} \quad (3.7)$$

where  $m_2$  – the volumetric concentration of the inclusion,  $v = \lambda_2 / \lambda_1$ . The derivation of this formula was also repeated by the authors of this paper.

It should be emphasized that in the structure with closed inclusions, geometrical components are not equivalent and their positions cannot be changed, and the continuous component in the formula (3.7) has to always come first.

When the elementary cell is divided isothermally in the form of a cube in a cube, the relation is as follows

$$\frac{\lambda}{\lambda_1} = \frac{I + (v - I) \cdot m_2^{2/3}}{I + (v - I) \cdot m_2^{2/3} \cdot (1 - m_2^{1/3})}. \quad (3.8)$$

Combined division with adiabatic and isothermal cross sections leads to the formula

$$\frac{\lambda}{\lambda_1} = I - \frac{m_2}{(1 - v)^{-1} - (I + m_2) \cdot (1 - m_2^{1/3}) \cdot (1 + m_2^{1/3})^{-1}}. \quad (3.9)$$

In order to calculate the effective thermal conductivity of a two-component system with closed inclusions, a simple formula by Odelevsky [11] is widely used in literature and is also cited in the papers [2, 3, 5]

$$\frac{\lambda}{\lambda_1} = I - m_2 \cdot \left( \frac{I}{I - v} - \frac{I - m_2}{3} \right)^{-1}. \quad (3.10)$$

The results of the calculations based on the above formula are practically identical to the results of the calculations based on the formula (3.9) and appear to be smaller than the results obtained from the formula (3.8).

The authors of this paper [2] considered about thirty models containing empirical coefficients without taking into account the radiation component of thermal conductivity. In their opinion, only two of them provide results that are very similar to the experimental values. If we take into account the radiation component of thermal conductivity, then the results of calculations using these formulas will be much greater than the experimental values. At the same time, it turned out that two consecutive patterns could correctly predict experimental data. Therefore, it made sense to include one of them in a later analysis carried out by the previously mentioned Schultz and Glicksman. Their relation for determining the effective thermal conductivity of a two-component system was obtained using the volumetric law of mixtures for modelling the structure by means of identical cubic cells in bars with about 80-90% polymer concentration. This relation has the form [2]

$$\lambda = \lambda_1 + \left( \frac{2}{3} - \frac{I}{3} \cdot f_s \right) \cdot (I - m_1) \cdot \lambda_2 \quad (3.11)$$

where  $\lambda_1$  and  $\lambda_2$  – the thermal conductivity coefficients for gas and the polymer skeleton respectively;

$m_1$  – the volumetric gas concentration in polymer foam ( $m_1 + m_2 = I$ ),

$f_s$  – the coefficient including the polymer concentration in the cell bars.

The authors of the formula (3.11) recommend adopting the value of the coefficient  $f_s$  as 0.8. In order to calculate the radiation component of thermal conductivity, we use the frequently cited relation [1, 2, 3]

$$\lambda_p = 4 \cdot F \cdot \sigma \cdot T_m^3 \cdot \delta \quad (3.12)$$

where  $F$  – the parameter including the form of the cell and the blackness extent of its walls;

$\sigma$  – the Stefan-Boltzmann constant;  
 $T_m$  – the average sample temperature, K;  
 $\delta$  – the average cell size,  $m$ .

According to data collected by different authors [1, 2, 3], the value of the parameter  $F$  is from 0.6 to 0.85.  $F = 0.7$  was used in our calculations.

Next, it was necessary to verify the ability of the presented relations to predict experimental values of the thermal conductivity coefficient of polymer foams  $\lambda_o$ . In order to do this, the experimental data [2] were used for samples of dry polyurethane foams with different density and a constant thickness of 50mm. The measurements  $\lambda_o$  were carried out at the average temperature  $T_m=297K$ , with the thermal conductivity coefficient of the gas filling the pore space  $\lambda_1=0.0143W/(m\cdot K)$  and the thermal conductivity coefficient of solid polyurethane  $\lambda_2= 0.25W/(m\cdot K)$ . The results of the experiments and calculations based on the formulas analysed in the paper are presented in Tab.1.

Table 1. Experimental  $\lambda_o$  [2] and calculated values of the thermal conductivity coefficient  $\lambda$  of polyurethane foam samples with the apparent density  $\rho$ , the porosity  $m$  and the average cell size  $\delta$ .

Sample No.	$\rho, kg/m^3$	$m$	$\delta, \mu m$	$\lambda_o \cdot 100, W/(m\cdot K)$	Calculated $\lambda \cdot 100, W/(m\cdot K)$								
					Formula No.								
					(8)	(9)	(10)	(11)	(12)	(13), (14)	(15)	(16)	(8)+(16)
1	31	0.973	320	1.8	1.68	1.78	1.77	1.87	1.87	1.87	1.69	0.13	1.81
2	50	0.957	410	2.0	1.84	1.97	1.97	2.13	2.14	2.14	1.84	0.17	2.01
3	80	0.930	220	2.2	2.11	2.33	2.33	2.57	2.60	2.59	2.10	0.09	2.20
4	96	0.917	330	2.8	2.25	2.51	2.51	2.79	2.83	2.81	2.22	0.14	2.39
5	173	0.850	340	3.2	2.99	3.47	3.46	3.92	4.03	3.98	2.87	0.14	3.13
6	234	0.797	240	3.6	3.67	4.33	4.29	4.83	5.04	4.93	3.37	0.10	3.77

Since the measured values of the thermal conductivity coefficient  $\lambda_o$  are a random variable, it is advisable to take into account the random measurement error while comparing the experimental and computational data. In the case under analysis, on the basis of  $n = 6$  of the experimental values  $\lambda_o$ , we will construct an approximate, adequate regression equation and we will estimate the experimental error. The experimental data from Tab.1 adequately describe the linear regression equation

$$\hat{\lambda}_o = 0.0161 + 0.1028 \cdot (1 - m_j), \tag{3.13}$$

from which, by analogy to Eq.(3.11), we conclude that with known values of the thermal conductivity coefficients of the components  $\lambda_1$  and  $\lambda_2$  and the target coefficient  $f_s$ , the effective thermal conductivity coefficient only depends on the volumetric concentration of one the components.

The sum of the squares of experimental data deviations from the straight-line regression (3.13) is  $SS=1556 \cdot 10^{-8}$ . The variance  $s^2 = SS / (n - 2) = 389 \cdot 10^{-8}$  can be adopted as the experimental error estimation with the degrees of freedom  $\nu = n - 2 = 4$ , and the standard deviation value  $s = \sqrt{s^2} = 1.972 \cdot 10^{-3} W/(m\cdot K)$  can be determined.

A more accurate estimation of the standard deviation, based on experimental data, is presented in the paper [12]. On the basis of the results of 3 tests on polyurethane samples, with a different number of tests  $n_i$  ( $i=1..3$ ), characterized by homogenous variances ( $n_1 = 148, s_1^2 = 248.43 \cdot 10^{-8}$ ;  $n_2 = 111, s_2^2 = 158.4 \cdot 10^{-8}$ ;  $n_3 = 14, s_3^2 = 229.39 \cdot 10^{-8}$ ), the authors of this paper determined the standard deviation for all tests

$s = 1.535 \cdot 10^{-3} \text{ W/(m}\cdot\text{K)}$  with the degrees of freedom  $\nu=270$ . When the first estimation is taken into account, we finally have  $s = 1.542 \cdot 10^{-3} \text{ W/(m}\cdot\text{K)}$  with  $\nu=274$ . This estimation practically reflects the standard deviation of the general population and it will be used in further analyses.

When we look at Tab.1, it is easy to notice that the experimental data are predicted most accurately by the relation (3.4), which includes the radiation component of thermal conductivity (3.12). An exception is the result obtained from sample No.4 with the density  $\rho=96 \text{ kg/m}^3$ . In this case, the difference between the experimental data and the calculated data exceeds the double value of the standard deviation ( $2\sigma = 3.08 \cdot 10^{-3}$ ). It is possible that in this sample the number of closed cells was greater than in the other samples. When the radiation component is included (3.12) quite a good result is also given by the formula (3.11) with the coefficient  $f_s=0.8$ . By changing this coefficient, it is possible to improve the accuracy of data prediction by this relation. In comparison to the formula (3.11), the relation (3.4) does not include empirical coefficients, and for this reason it is much easier to use. Moreover, it is emphasized [5] that a comparison between the results of the calculations based on the formula (3.4) and a number of experimental data brings good results, so the widespread use of this formula is fully justified.

Let us note that according to the formula (3.12) the greatest participation of the radiation component in the total thermal conductivity was obtained for samples with the apparent density of 31 and 50  $\text{kg/m}^3$ , and it only amounts to 7.2 and 8.4%. In the case of the samples with higher density this participation is even lower and it does not exceed 5.8%.

When the radiation component of thermal conductivity is taken into account, the other formulas will predict experimental data with deviations exceeding the double standard deviation  $2\sigma$ . With the degree of significance  $\alpha = 0.05$ , such deviation should be regarded as statistically significant. The formulas (11) - (14), including structures with isolated inclusions, bring results that are close to each other and significantly exceed the results of the calculations based on the formulas (3.4) - (3.6), obtained for structures with mutually interpenetrating components. For example, the difference in values calculated for the experimental conditions according to the formulas (3.4) and (3.7) (adiabatic cell division), in four cases out of six exceeds  $2\sigma$ . This indirectly indicates that the structure of the polyurethane foam samples in question mainly consists of open cells.

#### 4. Conclusions

An analysis of a number of publications leads to the assumption that the structure of used polymer foams mainly consists of open cells. On the basis of the analysis of experimental data and the relations for determining the effective thermal conductivity of dry polymer foams consisting mainly of open cells, it was demonstrated that the proposed relation (3.4), describing heat transfer in a structure with mutually interpenetrating components with adiabatic division of elementary cells is fully justified. It is also possible to use a very simple formula (3.11), which is based on the law of mixtures and includes the empirical coefficient. For the analyzed polyurethane foam samples, the calculated value of the radiation component did not exceed the double standard deviation value and at the significance level  $\alpha= 0.05$  is not statistically significant. In comparison with the structure with closed cells, the structure of polymeric foams with fine, open cells makes it possible to reduce the thermal conductivity of the material.

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