COMPUTATIONAL EFFECTIVE THERMAL CONDUCTIVITY OF POLYURETHANE MIXED CELL FOAMS

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Abstract. The objective of this paper was to calculate effective thermal conductivity of polyurethane foam used in thermal insulation of buildings with a new modeling approach. The proposed approach was more realistic as it simulated the real fraction of closed cells and open cells in the foam and it modeled the real physical phenomena that happen when both types of cells are present. This study was investigated for polyurethane foam with 70 % of closed cells and 30 % of open cells, by using finite element method and numerical homogenization. The result showed that there was a systematic change in thermal conductivity when the type of cell (closed, open and mixed cell) varied at fixed volume fraction. Also the effective thermal conductivity of mixed cell of this PU foam was about $\lambda_{cm} = 0.07$ W.m$^{-1}$.K$^{-1}$. Simulation proved the interest of this approach. Indeed, it brought new factor that influence the effective parameters which was fraction of closed cells and open cells. They suggested new method for computing thermal conductivity as a function of thermal conductivity of closed and open cell foam.

Nomenclature

| QUANTITY | SYMBOL | SI UNIT |
|----------|--------|---------|
| Density of PU foam, solid matrix and air | $\rho, \rho_s, \rho_g$ | Kg/m$^3$ |
| Porosity | $\xi$ | % |
| Solid volume fraction | $f_s$ | - |
| Fraction of closed cell | $f_c$ | - |
| Length of the struts | $l$ | m |
| Diameter of the strut | $d$ | m |
| Directional nodal Temperature | $T_i, i=x,y,z$ | K |
| Directional nodal heat flux | $q_i, i=x,y,z$ | W/m² |
| heat conductivity | $\lambda, \lambda_m, \lambda_{oc}, \lambda_c$ | W·m$^{-1}$·K$^{-1}$ |
| Total volume, volume of solid matrix, volume of air | $V, V_s, V_g$ | m$^3$ |
1. Introduction

The material behavior is determined by its characteristics parameters, balance equation and its constitutive laws. The coefficients such as the thermal conductivity, permeability and Young’s module are intrinsic to the material and help to identify the material application. This macroscopic description can be obtained either by experiment, or by theory. In the case of heterogeneous material, the experimental characterization cannot give intrinsic coefficients because this kind of study is limited to the continuous medium [1]–[3], and experimental conditions impact the results. Analytical methods are appropriate to the case of heterogeneous materials having a simple microstructure like composite [4]. This analytical description is delicate in the case of porous materials, because they have a random geometry structure (distribution, orientation, shape and volume fraction). Unlike theoretical and experimental studies, numerical ones overcome those problems, so we can reconstruct the complex morphology and compute effective coefficients of this kind of materials using numerical simulation.

The geometry structure of the pores is the main parameter affecting the properties of heterogeneous material. KANAUN [5] [6] studies the effect of struts and cell size distribution on effective thermal conductivity of metallic open cell foam using computational homogenization. The numerical analysis of effective thermal conductivity was investigated for plastic closed cell foams in [7]. At fixed values of porosity and number of struts per cell of open cell foam, if the size of cells increase, radiative conductivity increases as well [8]. The influence of microstructure also appears on the properties of convection for open cell foam, as the number of Nusselt increases simultaneously with increasing porosity at constant cell density [9] [10]. Also Marvi-Mashhadi an al. [11] [12] study mechanical behavior of polyurethane closed cell foams by means of computational homogenization.

Most of geometrical modeling of metallic or plastic foams in the literature consider only open cells or closed cells. However, current manufacturing processes cannot reach foams with 100% of only closed cells [13], also in order to have combined characteristics of both types of foam (for example thermal and acoustic insulation) it is necessary to introduce both open and closed cells. Also According to the fraction of closed cell, PU foam is classified in 4 categories [14] CCC1(fr <20%), CCC2(20% ≤ fr ≤ 80%), CCC3 (80%< fr ≤89%) and CCC4(fr ≥ 90%). Therefore we propose to model the structure of the foam with the real fraction of closed cells and open cells.

2. ANALYSIS AND MODELLING

2.1. Geometrical modeling of the microstructure

Polyurethane foam is a porous heterogeneous material; the microscopy parameters vary in the material. It is made of thin struts which compose the solid phase and cavities. The cavities are polyhedral cells with polygonal faces (in the case of closed cells) and edges.

In this study, the geometry of the polyurethane foam is considered as a periodic structure and characterized by a representative elementary volume (REV). The construction of the REV is based on the kelvin cell method [15] and has a tetrakaidecahedron shape. This method is an idealization of geometrical modeling of the microstructure [16]. Three types of microstructures are considered:

- Microstructure 1: with open cell only
- Microstructure 2: with closed cell only
- Microstructure 3: with mixed cells

Figure 1 illustrates Kelvin multi-cells models in the three cases for 20 cells.
Figure 1 Geometrical modeling of PU foam using 20 cells:  a. Closed cells PU foam (CC), b. Open cells PU foam (CO), c. Mixed cells PU foam (CM) with 70 % of closed cells and 30 % of open cells.

The parameters of geometrical modeling of the kelvin cells (the length $l$ and the width $d$ of the struts (see Figure 2) are taken from the morphology analysis of the microstructure of PU foam in [17]. The fraction considered of closed cell is 70%, the diameter of ligament is about 48 μm and the length is about 160 μm. The experimental porosity of this material is 98% [17].

Figure 2 Geometry parameters of single Kelvin cells: a. open cell, b. closed cell

2.2. Homogenization theory

The homogenization theory was introduced by Enrique Sanchez-Palencia. It assumes the existence of homogeneous material which is macroscopically equivalent to the heterogeneous material. It is continuous medium that behaves in "Average" as the heterogeneous material [18]. So thermal conductivities of PU foams can be determined by solving Fourier's law:

$$\langle \dot{q} \rangle = -\langle \lambda \nabla T \rangle \quad (1)$$
The Hill-Mandel criteria [19] give the equivalence of the average heat dissipation and the heat dissipation of the averages:

\[-\langle \dot{q} \cdot \nabla T \rangle = -\langle \dot{q} \rangle \langle \nabla T \rangle = \langle \dot{q} \rangle (-\nabla T) \quad (2)\]

The assumption of the unique equivalent continuous medium introduces thermo-physical parameters associated to the medium porous and defined as macroscopic effective coefficients. So the effective thermal conductivity \( \langle \lambda \rangle \) corresponding to the average of the local value at \( \lambda(P) \) inside the REV:

\[
\langle \lambda \rangle = \frac{1}{\text{REV}} \int_{\text{REV}} \lambda(P) dV \quad (3)
\]

2.3. Computational thermal simulation

The computation of the effective thermal conductivity of PU foam requires studying thermal conduction in the direction of the thickness. So we apply Uniform Dirichlet conditions in both boundaries: \( T_L = 277.15 \text{K} \) & \( T_R = 317.15 \text{K} \) in order to generate conduction. Pure polyurethane and air have a constant thermal conducting in this temperature range: the considered thermal conductivity for matrix phase is 0.29 \( W.m^{-1}.K^{-1} \) and for pores 0.024 \( W.m^{-1}.K^{-1} \). The value used are taken from [20].

The technique used for meshing is finite element method with unstructured meshing. It is the most popular method for discretization of complex geometry in the case of linear problem as heat equation in the case of steady-state conduction.

For that purpose microstructure parameters are unchanged, but different number of cells (1, 10, 20 and 30 cells) and different mesh resolutions are used in the three cases open, closed and mixed cell types.

The parameters of PU foam are deduced from calculating directional heat flux \( q_x, q_y, q_z \) (Figures 3, 4, 5 Directional heat flux for 20 mixed cell) and directional temperature gradient \( \nabla_x T, \nabla_y T, \nabla_z T \) (Figures 6, 7, 8 directional temperature gradient for 20 mixed cell) over the nodes of the domain by using ANSYS workbench, then computing the averages.
So if we assume that the macroscopic equivalent is an isotropic and homogeneous material, the effective thermal conductivity is given by [21]:

\[
\lambda = \frac{\langle q_x \rangle \langle \nabla_x T \rangle + \langle q_y \rangle \langle \nabla_y T \rangle + \langle q_z \rangle \langle \nabla_z T \rangle}{\langle \nabla_x T \rangle^2 + \langle \nabla_y T \rangle^2 + \langle \nabla_z T \rangle^2}
\]  

(4)

3. Validation of the geometrical modeling and validation of the results

In order to confirm the precision relevance suitability of the geometrical modeling the PU foam, we compare apparent porosities of the model with the theoretical and the experimental ones. There are two methods of computing porosity \(\xi\): the first one is established the relation between porosity, the density of the PU foam \(\rho\) and the density of the pure polyurethane \(\rho_s\):

\[
\xi = 1 - \frac{\rho - \rho_s}{\rho_s - \rho_g} \approx 1 - \frac{\rho}{\rho_s} \quad \text{si} \quad \rho \gg \rho_g
\]

(5)

Gibson and Ashby approaches [16], gives a density of a package of Kelvin cells as a function of the struts dimensions, to compute theoretical porosities in the 3 cases:

- Theoretical porosity of a tetrakaidecahedron open cell:
  \[
  \xi_o = 1 - 1.06 \times \frac{d^2}{T^2}
  \]
  \[
  \xi_o = 90.46\%
  \]

- Theoretical porosity of a tetrakaidecahedron closed cell:
  \[
  \xi_c = 1 - 1.18 \times \frac{d}{T}
  \]
  \[
  \xi_c = 64.6\%
  \]

- And we can find that theoretical porosity of a tetrakaidecahedron mixed cell:
  \[
  \xi_m = (1 - f_r) \cdot \xi_o + f_r \cdot \xi_c
  \]
  \[
  \xi_m = 72.36\%
  \]
The second method gives the porosity $\xi$ as a function of the solid fraction. It is used to compute apparent porosities of the models:

$$\xi = 1 - f_s = \frac{V_s}{V} \quad (9)$$

So we evaluate apparent porosity for the different models and we calculate the relative error. We notes that it is lower than 16% in comparing with theoretical values, and it is lower than 18% in comparing with the experimental ones (see Table 1 for more details).

### Table 1
Comparison between Apparent, Experiment and theoretical porosity

| Cell types       | Apparent porosity | Relative error compared to the experimental | Relative error compared to the theory |
|------------------|-------------------|--------------------------------------------|--------------------------------------|
| Closed cell      | 74.9%             | -                                          | 16%                                  |
| Open cell        | 93.6%             | -                                          | 3.4%                                 |
| Mixed cell (fr=70%) | 80%              | 18%                                        | 11%                                  |

Thermal error is an important parameter to check the validation of results, it evaluate the continuity of flux and temperature gradient. High value requires refinement of the mesh. It is based on error estimation technique for adaptive finite element analysis of heat conduction problems given by Huang and Lewis [22].

Thermal error for each element $i$ is [23]:

$$e_i = \frac{1}{2} \int_{V_e} \{\Delta q\}^T [D]^{-1} \{\Delta q\} dV_e \quad (10)$$

Where:

- $e_i$: energy error for element
- $V_e$: volume of element
- $[D]$: conductivity matrix
- $\{\Delta q\}$: thermal flux error vector

The thermal error of geometrical modeling of PU foam at converged value of thermal conductivity are reported in Table 2. Figure 9 show exemple of thermal error of 20 mixed cell PU foam.

### Table 2
Thermal error at converged value of thermal conductivity

| Cell types       | Number of cells | Average thermal error |
|------------------|-----------------|-----------------------|
| Closed cell      | 1               | 7.04E-10              |
|                  | 10              | 2.1975                |
|                  | 20              | 53.6901               |
|                  | 30              | 9.79E-09              |
| Open cell        | 1               | 1.75E-08              |
|                  | 10              | 38.1283               |
|                  | 20              | 8.83E-09              |
|                  | 30              | 4.46E-09              |
| Mixed cell (fr=70%) | 10              | 25.4981               |
|                  | 20              | 1.18E-08              |
|                  | 30              | 23.0786               |
4. RESULTS AND DISCUSSION

In this part, we verify the convergence of effective thermal conductivities. The results should be more accurate when mesh density and/or number of cells increases, so the results converge to a single value.

The numerical results for the three cases are compared with different analytical bounds: Hashin–Shtrikman bounds (H.S bound+ & bound-), and models such as: series model, parallel model, Self-consistent model (S.C) and the two forms of Maxwell-Eucken models (M.E model 1 & 2) (Figures 10, 11 and 12). These bounds and models don’t consider distribution, shape, and orientation of cells. The only morphological parameter used is volume fraction. For computing the analytical effective thermal conductivity, we use the forms described in [4], [21]. And the apparent volume fraction.

The analytical models and bounds used for thermal analysis are given by following equations:

- **Series model:**
  \[ \lambda = \frac{\lambda_a \lambda_g}{(1-f_o)\lambda_a + f_o \lambda_g} \]  
  (11)

- **Parallel model:**
  \[ \lambda = f_o \lambda_a + (1-f_o)\lambda_g \]  
  (12)

- **Self-consistent model:**
  \[ \lambda = \frac{1}{4} (3f_v - 1)\lambda_a + \lambda_g (3(1-f_v) - 1) + \sqrt{[(3f_v - 1)\lambda_a + \lambda_g (3(1-f_v) - 1)]^2 + 8\lambda_a \lambda_g} \]  
  (13)

- **Maxwell-Eucken model 1:** Air considered as continuous phase:
  \[ \lambda = \frac{\lambda_a + \lambda_g - 2(\lambda_g - \lambda_a)f_v}{2\lambda_a + \lambda_g + (\lambda_g - \lambda_a)f_v} \]  
  (14)

- **Maxwell-Eucken model 2:** pure polyurethane considered as continuous phase:
  \[ \lambda = \frac{\lambda_a + \lambda_g - 2(\lambda_g - \lambda_a)(1-f_v)}{2\lambda_a + \lambda_g + (\lambda_g - \lambda_a)(1-f_v)} \]  
  (15)

- **Hashin–Shtrikman bound +:**
  \[ \lambda = \lambda_a + \frac{f_o}{\frac{1}{\lambda_a} - \frac{f_o}{\lambda_g} + \frac{1-f_o}{\lambda_g}} \]  
  (16)

- **Hashin–Shtrikman bound -:**
  \[ \lambda = \lambda_g + \frac{f_o}{\frac{1}{\lambda_g} - \frac{f_o}{\lambda_a} + \frac{1-f_o}{\lambda_a}} \]  
  (17)

The convergence value of effective thermal conductivity of closed cell of this PU foam is about \( \lambda_c = 0.08 \, W \cdot m^{-1} \cdot K^{-1} \) (Figure 10). The numerical results are approximately between parallel model and H.S bound- (which is equal to M.E model 2).
Figure 10 Effective thermal conductivity of closed cell PU foam \( (W \cdot m^{-1} \cdot K^{-1}) \) versus Mesh size (um).

The convergence value of effective thermal conductivity of open cell of this PU foam is about \( \lambda_a = 0.05 W \cdot m^{-1} \cdot K^{-1} \) (Figure 11). The numerical results are close to parallel model.

Figure 11 Effective thermal conductivity of open cell PU foam \( (W \cdot m^{-1} \cdot K^{-1}) \) versus Mesh size (um).

The convergence value of effective thermal conductivity of mixed cell of this PU foam is about \( \lambda_m = 0.07 W \cdot m^{-1} \cdot K^{-1} \) (Figure 12). The numerical results are approximately between parallel model and H.S bound- (which is equal to M.E model 2).
Figure 12 Effective thermal conductivity of mixed cell PU foam (W. m$^{-1}$. K$^{-1}$) versus Mesh size (um).

- Thermal conductivity converge rapidly for finer meshes and large volumes for the three cases.

- We can perceive that the thermal conductivity of mixed cells (which represents the real morphology of the PU foam) is a linear function of thermal conductivity of open and closed cells:

$$\lambda_m = \lambda_o (1 - f_r) + \lambda_c f_r$$

- The numerical results are based on the assumption of averaging values of thermal conductivities, these averages are computing in nodes which describe the geometry of the material and this explains the reason that the closest analytical model to the numerical results in the three cases (close, open and mixed cell) is parallel model.

- Maxwell-Eucken model 2 is also close to the results because it includes the distinction between the dispersed phase and the continuous phase and this is the reason make Maxwell-Eucken model 1 is far from the numerical results.

- The analytical homogenization of random heterogeneous materials is introduced by Kroener [24]. Self-consistent model is used when the material has perfectly random material and anisotropic. Hashin–Shtrikman bounds are used in the case of random and isotropic material [25]. The material studied is considered as periodic and isotope. So the difference between numerical results and analytical ones may be caused by this reasons.

CONCLUSIONS

This study is a computational method for calculating thermal conductivity of PU foam with the new approach. Two ways investigated for computing effective thermal conductivity of PU mixed cell foam: First method, the effective thermal conductivity of mixed cell foam is a linear function of thermal conductivity of open and closed cell foam. Second method is to compute directly the effective thermal conductivity from the model of mixed cell foam.

This study does not consider some material characteristic such as the aspect of randomly and anisotropic, so in order to develop the numerical model of the new approach we must include these parameters.

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