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Quantification de l'impact de la résistance thermique de contact entre fibres sur la conduction dans les matériaux d'isolation fibreux

Quantifying the impact of fiber-to-fiber thermal contact resistance on conduction in fibrous insulation materials

Clémence Gaunand[1,2,3], Yannick De Wilde[1], Valentina Krachmalnicoff[1], Adrien François[2], Veneta Grigorova-Moutiers[2], Karl Joulain[3]

[1]Institut Langevin, ESPCI Paris, Université PSL, CNRS, 1 rue Jussieu, 75238 Paris Cedex 05 (France), clemence.gaunand@espci.fr, yannick.dewilde@espci.fr, valentina.krachmalnicoff@espci.fr

[2]Saint-Gobain Research Paris, 41 quai Lucien Lefranc, 93303 Aubervilliers Cedex (France), veneta.grigorova-moutiers@saint-gobain.com, adrien.francois@saint-gobain.com

[3]Institut Pprime, CNRS, Université de Poitiers, ISAE-ENSMA, 11 Boulevard Marie et Pierre Curie, 86073 Poitiers Cedex 9 (France), karl.joulain@univ-poitiers.fr

RÉSUMÉ. Nous décrivons une nouvelle approche de simulation nodale, ainsi qu'un modèle théorique correspondant, permettant de quantifier la conduction thermique à travers la phase solide dans des réseaux fibreux tridimensionnels, en considérant l'impact des résistances de contact entre fibres. Nous démontrons que la conductivité solide peut être calculée par une courbe maîtresse, uniquement à partir de paramètres géométriques du milieu, en particulier dans le cas de milieux peu denses. Cela constitue un outil de prédiction puissant à appliquer à l'étude d'isolants réels comme la laine de verre, notamment pour rechercher des stratégies d'optimisation basées sur les paramètres structuraux des matériaux.
ABSTRACT. We describe a new nodal simulation approach, and a corresponding theoretical model, to quantify thermal conduction through the solid phase in three-dimensional fibrous networks, with the impact of fiber-to-fiber thermal contact resistances. We demonstrate that the solid thermal conductivity can be calculated by a master curve, based solely on geometric parameters of the medium, particularly in the case of low-connectivity. This provides a powerful predictive tool to be applied to the study of real insulation materials, to develop in particular optimization strategies based on structural parameters.
MOTS-CLÉS. Matériaux d'isolation fibreux, conduction, résistance thermique de contact, simulation numérique, conductivité solide
KEYWORDS. Fibrous insulation materials, conduction, thermal contact resistance, numerical simulation, solid-phase conductivity

Nomenclature

l	fiber length, m
$k_{cond,solid}$	solid thermal conductivity, $\mathrm{Wm^{-1}K^{-1}}$
θ_i	polar angle of a fiber *i,* rad
φ_i	azimuthal angle of a fiber *i,* rad
$\bar{\theta}$	mean fiber polar angle, rad
d	fiber diameter, m
V_f	fiber volume fraction
T_b	lower boundary temperature, K
ΔT	temperature difference applied between the two heat baths, K
k_{fib}	fiber thermal conductivity, $\mathrm{Wm^{-1}K^{-1}}$
Q	transverse heat flux, W

R_{fib}	fiber thermal resistance, $\mathrm{KW^{-1}}$
R_k	fiber-to-fiber thermal contact resistance, $\mathrm{KW^{-1}}$
r	geometry-modified thermal resistance ratio
H	vertical distance between two centers of fibers in contact, m
N_c	number of contacts per fiber
n_z	areal fiber number density through a horizontal section, $\mathrm{m^{-2}}$
$k_{0,th}$	predicted solid thermal conductivity at $R_k = 0$ by Refs. [VOL 20, ZHA 18], $\mathrm{Wm^{-1}K^{-1}}$
a	fiber aspect ratio
k_0	simulated solid thermal conductivity at $R_k = 0$, $\mathrm{Wm^{-1}K^{-1}}$
h	corrective prefactor for $k_{cond,solid}$
δT_{ij}	corrective term for the temperature difference at a contact, K
∇T_z	temperature gradient applied between the two heat baths, $\mathrm{Km^{-1}}$
$\langle N_c \rangle_l$	minimum average number of contacts per fiber

$\langle . \rangle$ refers to network-scale averaged quantities.

Introduction

Understanding heat transfer in fibrous insulating materials, especially conduction, is a major challenge due to their heterogeneous and multi-scale nature. The thermal performance of a material can be quantified by its effective thermal conductivity, which can be expressed as a sum of the different contributions to heat transfer for sufficiently low densities, according to the additive approximation introduced by Bankvall [BAN 72]. This means that all contributions are assumed to be uncoupled and can be studied separately. In fibrous materials, four mechanisms can be identified : convection, conduction through gas, conduction through solid, and radiation. Although many studies have been conducted to quantify the radiative [TON 80] and convective [LAN 90] contributions, only a few have considered the conduction term, especially the one associated with the solid phase. However, studying this mechanism is essential to understand how the material's structure can impact its final thermal properties and to integrate the relevant parameters into predictive models in order to identify new strategies for minimizing heat transport.

As solid conduction cannot be experimentally measured separately, numerical modeling is the most appropriate tool for its quantitative analysis. Numerical fibrous media can be reconstructed from images of real structures, acquired, for example, by X-ray tomography [MEF 19], or can be generated virtually, which is a faster approach. Representative numerical structures of insulation materials have been produced for example in various studies [ARA 13, KAL 22], using a modified random walk generation method and a specific fiber orientation distribution [SCH 06] representing the directional morphology of these materials. Then, conduction through the solid phase in fibrous media can be numerically computed using different methods. Phonon transport at the atomic level can be investigated using molecular dynamics : this has in particular been used to study the diffusive-ballistic transition of heat conduction in metallic nanotubes and the associated finite-size effects [GU 21]. For microscopic glass fibers, where heat transport is purely diffusive, such small-scale calculations are not necessary, and the most precise approach consists in using finite element analysis [KAL 22], although it requires considerable computing time and resources. However, if only solid conduction is studied, nodal methods can also be used, allowing for an increase in the simulation volumes. It is a common approach in the field of electrical conduction [ROC 15, KIM 18], but it has not been widely applied to thermal conduction [GHI 13].

Using these various methods, the impact of the Biot number on the evolution of solid conduction in fibrous networks has been demonstrated in the presence of fiber-to-fiber thermal contact resistances [FAT 20, VAS 08]. From a theoretical perspective, different models have been proposed but the most advanced to date is that developed for dense networks of straight nanofibers, in 2D by Volkov and Zhigilei [VOL 12, VOL 20], and in 2D and 3D by Zhao et al. [ZHA 18], that includes the impact of morphological parameters. Originally developed to describe carbon nanotube networks, this model must however be compared with simulation results that account for variations in all fiber geometric parameters and thermal contact resistance in the 3D case. Furthermore, its range of validity has not been precisely examined.

In this study, we use a multi-nodal representation to calculate the solid thermal conductivity in virtually generated 3D media with straight fibers. The results obtained are compared with the outputs of the previously mentioned model, and an enhanced theoretical framework is proposed to correct the observed deviations. Using this new model, we introduce an innovative representation of the evolution of solid thermal conductivity in 3D fibrous networks, under the form of a master curve, as four fiber parameters - orientation, length, diameter and density - along with thermal contact resistance, are varied. Finally, we demonstrate the relevance of this predictive approach in the context of studying a real-world insulation material, taking the specific example of glasswool.

1. Numerical methods

1.1. *Numerical generation of a network of connected straight lines*

Three-dimensional fibrous media are numerically generated as assemblies of straight lines, representing fibers, within a cubic computational domain of size L, with the final aim of calculating their solid thermal conductivity $k_{cond,solid}$. These fibers are assigned a prescribed orientation distribution and a constant length l. The orientation is defined by a pair of angles (θ_i,φ_i) in spherical coordinates, as shown in Fig. 1(a) : the azimuthal angle φ_i is chosen from a uniform distribution, while the polar angle θ_i is sampled according to the β-orientation distribution, previously used in various studies to model fibrous insulation materials (see Refs. [ARA 13, KAL 22] for details). The mean value of the polar angle $\bar{\theta}$ at the network scale is determined by one adjustable parameter in this distribution. A constant fiber diameter d is also defined and acts as the distance threshold for detecting contacts between fibers. This means that as a first approximation, fibers are modeled by soft-core, i.e., interpenetrating objects. The fiber volume fraction V_f is defined in this study as the volume fraction obtained after removal of unconnected fibers, that cannot contribute to heat transfers. Periodic boundary conditions are applied at the lateral edges of the cubic simulation domain. Fig. 1(b) shows an example of generated network using our numerical approach with identification of contact points.

1.2. *Resolution of a steady-state heat transfer problem including thermal contact resistances using a nodal method*

The steady-state transverse thermal conductivity through the solid phase $k_{cond,solid}$ - i.e., in the cross-plane direction for the fibrous medium, is computed numerically using the configuration of the guarded hot plate experiment, shown in Fig. 1(c). Two plates with different temperatures $T = T_b$ and

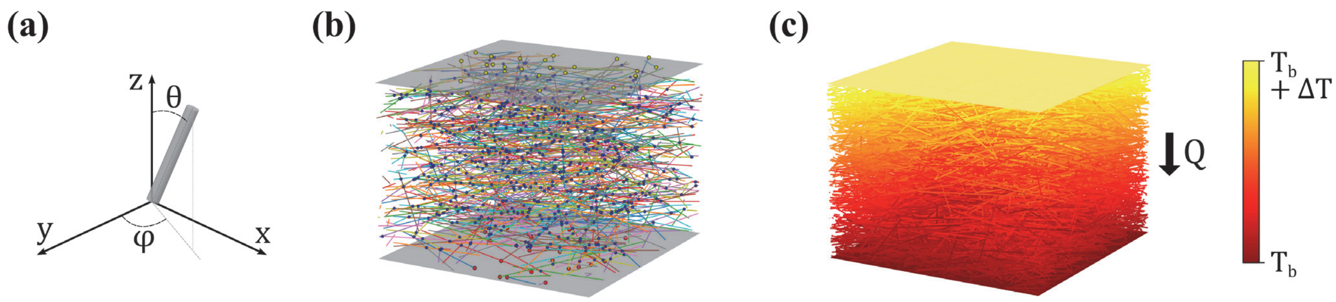

FIGURE 1. *(a) Coordinate system used for fiber generation. (b) Example of a generated network with identification of contact points. (c) Example of a simulated temperature field distribution resulting of a guarded hot plate configuration.*

$T = T_b + \Delta T$ are located at the lower and upper boundaries of the simulation domain, respectively. The four other side boundaries are assumed to be adiabatic, and the fiber thermal conductivity is fixed to $k_{fib} = 1.3$ Wm^{-1}K^{-1}, the value for silica glass.

A nodal method is implemented to compute the transverse heat flux Q in the medium and therefore the associated solid thermal conductivity. It assumes uniform sections in temperature, i.e., unidirectional conductive heat transfer within each fiber : indeed, since only conduction through the solid phase is considered, the external surface of the fibers must satisfy adiabatic conditions. This approach is based on the representation of the fibrous medium by a equivalent thermal circuit of temperature nodes and resistance branches. In particular, the multi-nodal representation is used, considering both the internal thermal resistance of fibers R_{fib} and the thermal contact resistance R_k : each fiber fragment between two contacts and each contact constitutes a branch in the circuit, bounded by two nodes. When fiber extremities do not make contact with any other fiber, they are assumed to be adiabatic and are therefore excluded from the thermal network.

In the steady-state, the resulting thermal network can be described by algebraic graph theory. The temperature in each node and the heat flux in each branch can be therefore obtained from the resolution of a single matrix equation (see Refs. [KIM 18, GHI 13], in which a similar approach is used, for details on the resolution method). Then, the total heat flux Q is computed by adding the contributions from the branches in contact with the upper or the lower edge of the simulation domain, and the solid thermal conductivity is calculated by the following expression :

$$k_{cond,solid} = \frac{QL}{L^2 \Delta T}. \qquad [1]$$

Here, the temperatures are arbitrary fixed to $T_b = 300$ K et $\Delta T = 1$ K; however, these values do not influence the results as the thermal conductivity of individual fibers k_{fib} and the fiber-to-fiber thermal contact resistance R_k are assumed to be independent of the temperature. This hypothesis is valid for narrow temperature ranges ($\Delta T \leq 20$ K), in accordance with the usual configurations for guarded hot plate experiments.

For each set of input parameters l, $\bar{\theta}$, d and V_f, the size of the cubic simulation domain L is fixed so as to reach a Representative Volume Element (RVE), defined in this study as the minimal simulation volume for which the relative error on the solid thermal conductivity $k_{cond,solid}$ over 5 realizations at a 95% confidence interval is lower than 5%. A minimum volume is also fixed to ensure that no single fiber is connected to both heat baths at the same time, and that self-interaction due to finite-size effects is limited. The selected value of solid thermal conductivity is the mean of $k_{cond,solid}$ over 5 realizations at the RVE, associated with its 95% confidence interval.

2. Results and discussion

2.1. *Effect of fiber-to-fiber thermal contact resistance for different network morphologies and comparison with existing conductivity models*

Solid thermal conductivity is simulated for generated networks with different morphologies, varying independently V_f, l, d and $\bar{\theta}$, and with different values of fiber-to-fiber thermal contact resistance R_k. The studied ranges for the morphological parameters are chosen to include the average values estimated for glasswool : volume fraction is lower than 10%, fiber length is of the order of 1 mm, fiber diameter is of the order of 10 µm, and the mean polar angle is around 80° [PRI 24].

Following the theoretical framework previously introduced by Zhao et al. [ZHA 18] and Volkov and Zhigilei [VOL 12], a dimensionless parameter r including the variations of all these parameters is introduced :

$$r = \frac{2l\langle|\cos\theta|\rangle}{\langle H\rangle\langle N_c\rangle}\frac{R_k}{R_{fib}} = \frac{R_k\langle|\cos\theta|\rangle k_{fib}\pi d^2}{2\langle H\rangle\langle N_c\rangle} \qquad [2]$$

where $\langle N_c\rangle$ is the average number of contacts per fiber, $\langle H\rangle$ is the average vertical distance between two centers of fibers in contact, and $\langle|\cos\theta|\rangle$ is the average cosine of the polar angle of fibers. r can be interpreted as a geometry-modified Biot number, showing an interplay between intrinsic fiber thermal resistance $R_{fib} = \frac{4l}{k_{fib}\pi d^2}$ and thermal contact resistance R_k. The authors expressed the solid thermal conductivity in a dense three-dimensional fibrous network as :

$$k_{cond,solid} = \frac{1}{4}k_{fib}\pi d^2\langle n_z\rangle\langle|\cos\theta|\rangle\frac{1}{1+r} = k_{0,th}\frac{1}{1+r} \qquad [3]$$

where $\langle n_z\rangle$ is the average areal number density of fibers through a horizontal section, and $k_{0,th} = \frac{1}{4}k_{fib}\pi d^2\langle n_z\rangle\langle|\cos\theta|\rangle$ is the theoretical value of the thermal conductivity at $R_k = 0$.

Fig. 2 compares the normalized simulated solid thermal conductivities obtained in this present work with the predictions of Eq. 3, varying individually the fiber volume fraction, aspect ratio $a = l/d$, and orientation. Figs. 2(a) and (b) indicate that the agreement between dense network theory and simulation is acceptable for the highest volume fractions and aspect ratios only. In contrast, Fig. 2(c) shows that it is unchanged by the mean angle of the fibers in the studied range. The simulation results obtained in this study therefore form a validation approach and suggest that the model developed by Zhao et al. [ZHA 18] and Volkov and Zhigilei [VOL 12] should include correction terms to treat the case of low to moderate volume fractions or aspect ratios to gain accuracy.

2.2. *Origin of the drop in thermal conductivity at low connectivity and introduction of a general theoretical description*

Colored symbols in Fig. 3 show the evolution of $k_0/k_{0,th}$, the ratio of the simulated thermal conductivity at $R_k = 0$ to its corresponding estimation by the initial model, as a function of the average number of

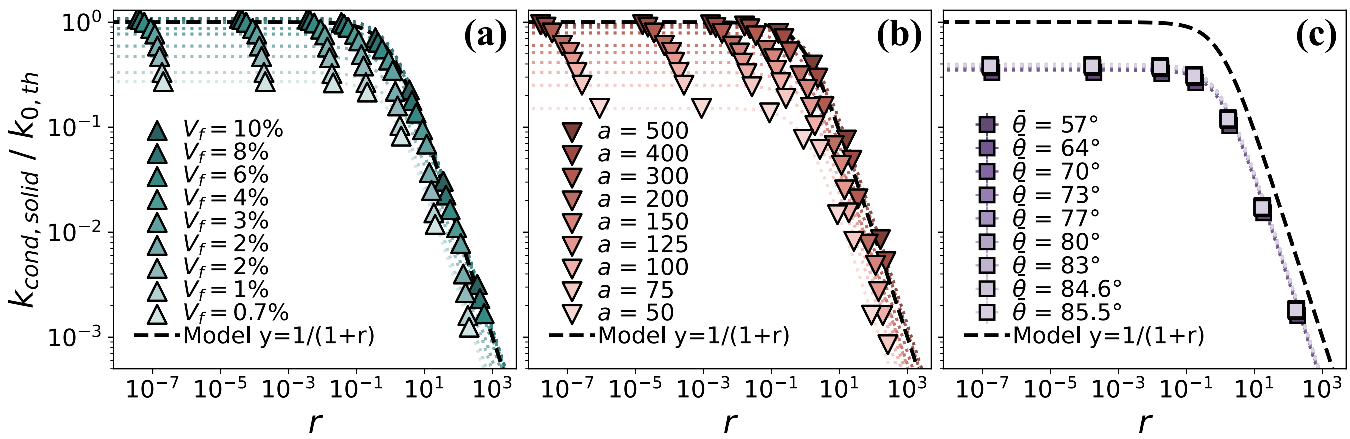

FIGURE 2. *Solid thermal conductivity normalized to $k_{0,th}$, the theoretical thermal conductivity at $R_k = 0$, as a function of the characteristic dimensionless parameter r, for (a) different volume fractions V_f, (b) different aspect ratios a, and (c) different fiber orientations governed by the mean angle to heat flux direction $\bar{\theta}$. The base parameters are $V_f = 0.01$, $a = 100$ and $\bar{\theta} = 57.3°$. The black dashed line shows the predictions of Eq. 3.*

contacts per fiber $\langle N_c \rangle$ for the different combinations of input parameters introduced in Fig. 2, using the same color representation. This figure first demonstrates that unlike fiber orientation, volume fraction and aspect ratio have a significant effect on $\langle N_c \rangle$. This quantity can be interpreted as a measure of the degree of connectivity of the fibrous network. Fig. 3 also highlights that the extent of the observed deviation between model and simulation depends solely on this degree of connectivity, independently of the values of a and V_f. Indeed, in a highly connected network, nearly all fibers participate in heat transfer, whereas in a poorly connected network, only a small fraction contributes : in this case, correlations between fiber temperatures may become significant. To take these phenomena into account, we have developed a new theoretical model, following an approach similar to that described in Ref. [VOL 20] for 2D networks. This model gives this new expression for the solid thermal conductivity :

$$k_{cond,solid} = k_{0,th} h \frac{1}{1+r} \tag{4}$$

with

$$h = 1 + \frac{\langle \delta T_{ij} \rangle}{\nabla T_z \langle H \rangle} \tag{5}$$

where $\langle \delta T_{ij} \rangle$ represents a corrective term for the temperature difference at junctions estimated by the initial model, and $\nabla T_z = \Delta T / L$ is the global temperature gradient imposed in the sample. The term $\langle \delta T_{ij} \rangle$ can therefore be estimated as the difference between the actual average temperature difference at contacts measured directly from simulations and that predicted by the initial model ; it represents the influence of correlations between the temperatures of the fibers in contact. In Fig. 3, round markers represent the evolution of h estimated from our simulation results using Eq. 5 and the method described above : this figure shows a good agreement between this proposed correction and the observed deviation $k_0/k_{0,th}$. Finding $0 \leq h \leq 1$ indicates that $\langle \delta T_{ij} \rangle$ is negative, i.e., that temperature differences at junctions are lower in average in the modified model. Indeed, taking correlations into account reduces temperature differences between closely positioned fibers, particularly those in direct contact. While the ratio $k_0/k_{0,th}$ is expected to approach 1 as connectivity increases, it is observed that the values obtained from simulation results exceed 1 in some cases, suggesting a slight overestimation. However, the direct calculation of h

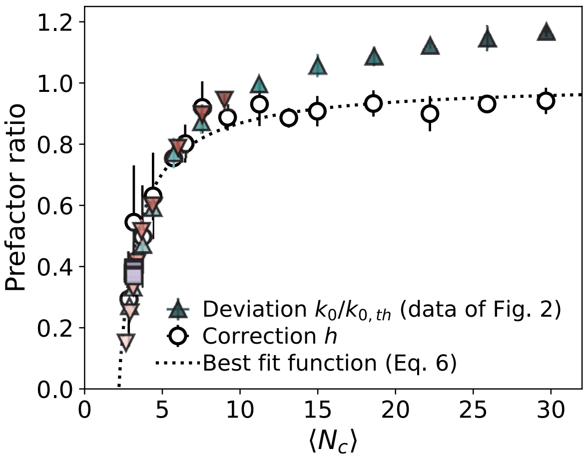

FIGURE 3. *Deviation $k_0/k_{0,th}$ corresponding to the data displayed in Fig. 2 (colored markers), compared to the correction h calculated with Eq. 5 (round markers) and the associated best fit function given by Eq. 6 (dotted line).*

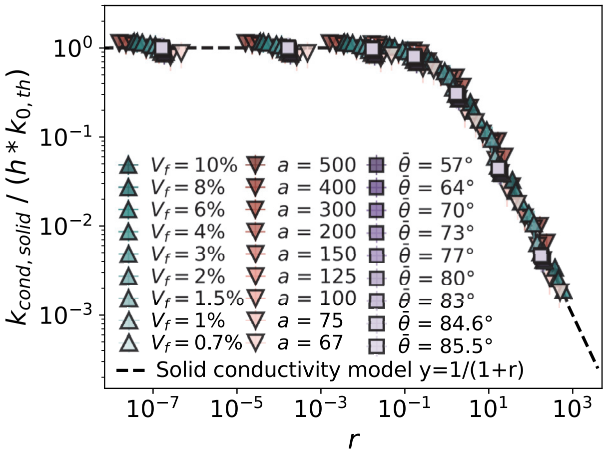

FIGURE 4. *Master curve for the evolution of the solid thermal conductivity, allowing predictions based on the morphological parameters and the thermal resistance ratio. The base parameters are $V_f = 0.01$, $a = 100$ and $\bar{\theta} = 57.3°$.*

seems to have addressed this discrepancy, leading to a higher level of accuracy in the results. The dotted line shown in Fig. 3 represents the best fit to the numerical data for h using a rational function of $\langle N_c \rangle$, chosen of the form of Eq. 5 with three adjustable parameters. The obtained function can be expressed as :

$$h = 1 - \frac{\langle N_c \rangle_l - 1}{\langle N_c \rangle - 1} \qquad [6]$$

where $\langle N_c \rangle_l = 2.18$ is the minimum average number of contacts per fiber for a conductive network, i.e., it corresponds to the percolation threshold. The significance of this function, that correctly describes the evolution of h, lies in its dependence on $\langle N_c \rangle$ only, which is a parameter determined by the geometry of the network. Consequently, it makes the prediction of solid thermal conductivity without solving complex heat transfer problems in fibrous networks possible even at low connectivity, directly using Eqs. 6 and 4, which saves a significant amount of computing time and resources. Fig. 4 shows the result of this improved prediction method under the form of a master curve, which is an innovative way of describing the evolution of solid thermal conductivity. Compared with Fig. 2, it highlights the relevance of the proposed theoretical model even for low to moderate fiber volume fractions and aspect ratios.

2.3. *Application to the case of glasswool*

To practically illustrate the interest of the modeling approach developed in this study, the example of glasswool can be considered. The vast majority of distributed glasswool materials exhibit densities below 70 kg.m^{-3}, which approximately converts into volume fractions below 3%, considering a glass density of 2200 kg.m^{-3}, and the average fiber aspect ratio is estimated around 100 [PRI 24]. The theory developed by Komori and Makishima [KOM 77], supported by more recent studies [VOL 12], establishes a link between the average number of contacts and these input parameters : for 3D networks, $\langle N_c \rangle = 2aV_f$. Using this expression, the maximum number of contacts per fiber in distributed glasswool can be estimated to be approximately 6. The area corresponding to $\langle N_c \rangle < 6$ in Fig. 3 shows that the correction h is lower than 0.8 in this case, and thus begins to significantly deviate from 1. This suggests that glasswool can be considered as a material with low connectivity, for which the proposed modified model is essential to accurately predict the associated solid thermal conductivity. Our approach therefore provides a robust predictive tool that can be applied to the analysis and optimization of real insulation materials.

3. Conclusion

In summary, we have numerically generated 3D fibrous networks and we have developed an original and efficient nodal simulation approach to calculate their steady-state solid thermal conductivity, integrating the effect of fiber-to-fiber thermal contact resistances. We have demonstrated that its evolution is governed by a master curve which depends on a characteristic ratio of thermal resistances ; this approach also constitutes a first validation of the most advanced theoretical model available to date, that shows a deviation in the case of poorly connected networks. This low-connectivity regime particularly applies to the case of insulation materials such as glasswool, in which fibers exhibit relatively low aspect ratios and low densities. We have introduced a novel theoretical model that is particularly well-suited for this case and demonstrated a substantial improvement in accuracy when calculating the resulting solid thermal conductivity, as validated by comparison with our simulation results. This model can be expressed in a simple form, depending entirely on the morphological parameters of the medium, allowing for the prediction of solid thermal conductivity without the need for heat transfer calculations.

This work paves the way for further investigating the impact of thermal contact resistance on the global thermal properties of insulation materials. The relevance of the model developed in this study has been demonstrated for a wide range of thermal contact resistances : next, we plan to perform local experimental thermal contact resistance measurements to select the accurate order of magnitude in this modeling approach. In addition, we are currently investigating the integration of distributions for the input parameters, based on real measurements, to account for a certain heterogeneity in the materials ; this also applies to the thermal contact resistance. In this regard, the correlations that might exist between morphological parameters in real-world materials will also have to be considered.

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