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MULTISCALE MODELLING OF BIO-COMPOSITES: TOWARDS PREDICTION OF THEIR THERMAL CONDUCTIVITY BASED ON ADEQUATE KNOWLEDGE OF THEIR CONSTITUENTS

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Abstract. To face up to the present energy crisis and the climate challenge, incorporating plant aggregates into reactive or clayey mineral matrices has proved to be an ecological and effective way to improve the thermal performance of buildings. It also helps to reduce both the grey energy involved and the consumption of fossil or non-renewable materials. However, the characterization and optimization of bio-based composites is relatively time-consuming, which may be a hindrance to their development. Our objective is to efficiently predict the thermal conductivity of a hemp particle as this is a fundamental input value for predicting thermal behaviour at the material scale. We found few reliable data on this topic in the literature. This is a major challenge that needs to be addressed as it can lead to erroneous results even when the prediction model is relevant and well developed. An additional difficulty arises from the fact that plant aggregate shows high variability. This paper opens new perspectives by suggesting new procedures for determining the thermal conductivity of hemp shiv using two approaches and considering, in particular, the variability of the raw material. Since existing methods for measuring thermal conductivity do not provide access to the particulate value, an experimental and a theoretical method are proposed to determine it. Both are based on analytical equations obtained by homogenization and both are shown to lead to consistent results. Comparison with the few data available in the literature also highlights a possible overestimation of the values found in previous works.

Keywords: Multiscale modelling, plant aggregates, thermal conductivity

1 Introduction

To address environmental issues, the building sector has to innovate and reduce its energy consumption and greenhouse gas emissions by developing new materials that are

environmentally friendly while ensuring the comfort of users. Using plant aggregates is a way to deal with this challenge, as they are identified as agricultural, renewable and locally-available by-products. The use of plant aggregate for construction limits both transport emissions and fossil energy consumption. Plant-based aggregates also allow large quantities of carbon dioxide to be stored in the materials throughout the building's life-cycle thereby reducing its carbon impact [1] and presenting an additional environmental asset. Last but not least, plant particles possess high insulating and hygroscopic capacities. These multiple factors should lead the building industry to massive use of such raw materials. However, the substantial variety of formulations (nature and dosage of aggregates and binder) induces an equally wide range of thermal performance. Therefore, the characterization and optimization of those types of building materials can be costly and time consuming.

In consequence, several models have been proposed to predict thermal properties of bio-based materials. However, the particulate thermal conductivity remains unmeasurable by current techniques while they are needed as model inputs. In addition, while a homogenization model is currently applied by several authors [2]–[5], considering the particle as an assembly of composite spheres could be questioned, in particular in view of the actual particle geometry. A more faithful representation of microstructure should be investigated. It is also worth noting that hemp shiv is the only plant aggregate with values of particle thermal conductivity available in the literature [3], [6], [7]. The methods by which these values are determined are not always explicitly stated by the authors. Finally, the hygroscopic nature of composites enables ambient hydric variations to be attenuated. However, it makes forecasting their behaviour a more complex task.

Our work is aimed at developing a new tool to guide and optimize bio-based material formulations even before the manufacturing step. This paper presents two different methods for determining the particulate thermal conductivity of dry hemp shiv. In order to validate the suggested procedure and also to limit the raw material variability, only one plant aggregate was chosen for this study: commercial hemp shiv. In addition, to avoid having to consider the effect of water absorption on thermal conductivity, this research was carried out on dry aggregates. Thus, the first method requires measurement of the thermal conductivity of dry particles in bulk, while the second one is more theoretical, being based on the chemical composition and microstructure of the particles. Both methods are founded on analytical equations obtained by homogenization. A protocol to access both thermal conductivity and volume fractions of particles in bulk is therefore established in the following. Indeed, this data is required in the models used. The shape and the microstructure of the particles as well as the anisotropy of the material and its effect on the thermal behaviour are also taken into account. Finally, the values obtained for the thermal conductivity of a hemp shiv particle were compared with those found in the literature. Our models predicted results in reasonable agreement with both isotropic and anisotropic considerations. However, this study may indicate an overestimation of the values determined by the self-consistent scheme [3]. This is discussed in this paper.

2 Materials

2.1 Choice of the plant aggregate

Adapted to temperate climates, hemp is a fast growing plant. Peripheral fibres of the plant are used to manufacture textiles and rope. The central part of the stalk, the porous pith, is crushed to obtain hemp shiv, which has been used for several years now as a building material in hemp concrete. This plant aggregate was selected for our study on different criteria. Firstly, the hemp particle is the one that has been most studied in the literature and for which comparative values of particulate thermal conductivity are available, either in scalar form (isotropic considerations) or as a tensor (anisotropic considerations).

Moreover, the hemp particle has high variability [8], [9]. As it is an agricultural by-product, its intrinsic properties depend on the geographical location and method of production, but also on the climate and the harvesting period. This leads to significant irregularity in the raw material extracted. Thus, variability can be observed in the particulate porosity, the density or the chemical composition of the solid skeleton. It is worth noting that the retting process used to separate fibres has an impact on the chemical composition of the aggregates obtained. In particular, the proportion of crystalline phase in the cellulose of hemp fibres depends on the degree of retting. This degree of crystallinity is known to impact the mechanical properties [5]. A possible effect on thermal conductivity will be highlighted in this paper. Finally, in order to limit intrinsic variability (shape factor, size, density, chemical composition) as far as possible, we chose commercial hemp shiv. To assess the consistency of the proposed models, this aggregate is taken as a reference in this study.

2.2 Physical specificities of hemp shiv

Shape of the hemp particle. The hemp shiv comes in the form of straw. The commercial product chosen for the study was dusted and calibrated (Fig 1). Macroscopically, it can be considered as a kind of elongated parallelepiped or even a cylinder. The shape parameters are available in the thesis of Ratsimbazafy [10]. Finally, hemp shiv particles are assumed to be cylindrical in line with the geometrical values of the aggregate studied and according to the literature [11], [12].



Fig 1. Hemp shiv particle used for this study (size ranging from 2 to 14 mm)

Microstructure of the hemp particle. Many capillaries are present in plant aggregates because they allow sap to circulate when the plant is growing. After drying, this structure naturally fills with air, which causes intra-particle porosity. From SEM observations [6], [7], hemp shiv reveals a tubular microstructure, which can be thus assimilated to a cylinder. In consequence, the solid skeleton of the hemp particle is made of cylindrical pores embedded in a continuous matrix.

Porosity. Hemp shiv is employed in construction for its insulating properties. This insulating capacity is a consequence of the high porosity present both inside the particle (intra-particle porosity) and between the particles themselves in the bulk paste (inter-particle porosity). The particle porosity of our samples was $83.5 \pm 4.8 \%$ [10]. The inter-particle porosity depends on the degree of compaction of the bulk mix.

Density. The density of the particles used in our study was experimentally estimated at $\rho_{\text{part}} = 248 \text{ kg.m}^{-3}$. This result is consistent regarding the high particle porosity and the density of the solid skeleton constituents [10]. Note that the density of the skeleton of hemp shiv and of cellulose in lignocellulosic biomass are both close to the order of 1540 kg.m^{-3} [10], [13].

2.3 Chemical characteristics of hemp shiv

Chemical composition also influences the properties of plant aggregates and cellulose is well known to be the major component of bio-aggregates. The hemp shiv used in our study was composed of more than 55% cellulose. The chemical components of this plant aggregate are listed in Table 1. These values will be useful to justify the assumptions made at the particle level in the homogenization process.

Table 1. Proportions of the constituents of the hemp shiv used expressed in % of their dry mass according Ratsimbazafy [10].

	Cellulose	Hemicellulose	Lignin	Minerals
Dosage (expressed in % of their dry mass)	55.5 ± 0.2	18.9 ± 0.1	16.4 ± 0.4	1.9 ± 0.0

3. EXPERIMENTAL AND THEORETICAL METHODS

3.1 Measurement of the thermal conductivity

Thermal conductivity was measured with the hot wire test since it is very suitable for measuring thermal conductivities ranging from 0.02 to 5 W.m-1K-1. Values around 0.051-0.070 W.m-1K-1 have been reported in the literature for hemp in bulk [10]. Prior testing, the aggregates were dried in an oven at 60°C until a constant weight was achieved and then cooled in a desiccator to ambient temperature. Measurements were made in a temperature-controlled space at 20°C. The samples (described in section 2.2) were weighed again after a sequence of measurements to ensure that water adsorption was negligible.

A Neotim-FP2C hot wire apparatus was used for this study. Since this transient method only allows local measurements, thermal conductivity measurements were repeated 5 times. The samples were stirred after each test because the hot wire caused local heating. A first round of measurements was carried out on a bulk mixture. Then, the aggregates were hand-compacted by 30% of the original volume in order to investigate the effect of compaction on the value of particle thermal conductivity.

3.2 Volume fraction measurement

One of the difficulties of our protocol was to know the volume fraction of the aggregates in the bulk in which the thermal conductivity measurement was carried out. This was because the granular arrangement was different for each bulk mix. To overcome this difficulty, and based on the recommendations of the RILEM TC 236-BBM [14], particles were weighed and placed in a container for thermal conductivity measurement and, at the same time, the total volume occupied by the mixture was marked. After removing the shiv from the container, it was filled with water to the level of the mark. Finally, the container was weighted and the volume of water, i.e. of bulk aggregates, was calculated.

The volume fraction of aggregates in the bulk arrangement, f_g , was calculated as follows:

$$f_g = \frac{V_g}{V_{tot}} \quad (1)$$

where V_{tot} is the total volume of bulk and V_g that of aggregates.

To gain access to this value experimentally, the previous equation was transformed into:

$$f_g = \frac{M_g}{\rho_{part}} \cdot \frac{1}{V_{tot}} \quad (2)$$

where M_g is the mass of aggregates.

3.3 Choice of homogenization schemes

Since it is currently impossible to experimentally measure the thermal conductivity of a single particle, homogenization techniques were used to overcome this difficulty. Note that a self-consistent scheme is generally used in the literature both at the bulk scale [3] and at the composite scale [15]. It is based on an assembly of spherical spheres of various sizes.

Another method was chosen to take the real morphology of the homogenized material into account. Here, we develop the theoretical basis of the techniques chosen for the study. For the heterogeneous material considered, i) the particles in bulk and ii) a single particle, were reduced to an equivalent homogeneous material. A representative volume element Ω (RVE) of the heterogeneous material had to be considered, with $\delta\Omega$ its outer boundary and n the outward unit normal $\delta\Omega$. The RVE was the minimum volume to be considered as representative of the macroscopic behaviour of the heterogeneous medium. Then homogenization required three steps:

Representation, which consisted of describing the material at the microscopic scale (shape, orientation of inclusions in the matrix).

Localization, which allowed the physical relationships to be expressed at the local scale of the material as a function of the different phases considered. Relations between the quantities at the microscopic scale and those defined at the macroscopic scale were defined.

Homogenization, which finally gave the behaviour of the equivalent homogeneous material thanks to averaging operations on the solutions of the previous step.

Thus, for our study, we considered two RVEs (Fig 2. and Fig 3) to work on the thermal conductivity tensor of a hemp shiv particle.

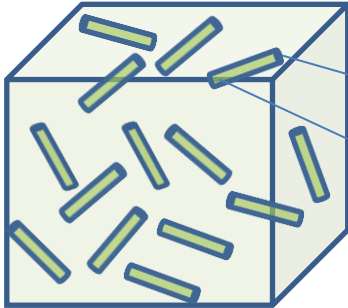


Fig 2. RVE for the particles in bulk

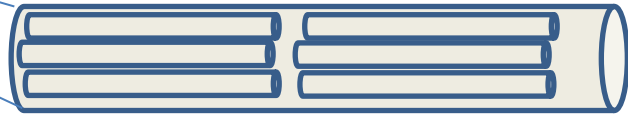


Fig 3. RVE at the particle scale (5-10mm)

The RVE considered for particles in bulk comprised two phases: a matrix of air and hemp cylindrical particles as randomly arranged inclusions. The RVE considered at the particle scale was also composed of a two-phase material containing solid matrix and elongated pore-inclusions. In both cases, the matrix was continuous and the interfaces were assumed to be perfect. Thus, we considered two scales: the macroscopic one (bulk or particle) and the microscopic one, at the scale of heterogeneities (inclusions). The fictitious homogeneous medium had the same geometry as the RVE and, subjected to the same stresses, was assumed to have an identical global response (Fig 4).

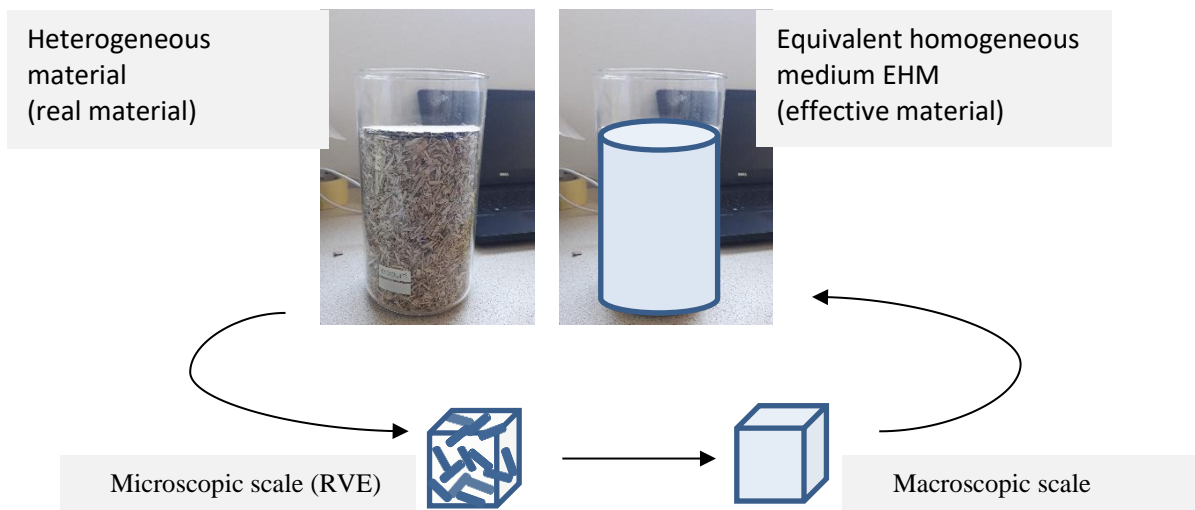


Fig 4. Homogenization process applied to hemp shiv in bulk

In order to provide uniformity in the notations, the following were adopted for this paper: index 0 to for the matrix, index i for the inclusions.

If the heterogeneous material is a matrix-type composite composed of several inclusions, according to classical theory of Eshelby, this homogenized thermal conductivity tensor is given by:

$$\Lambda^{hom} = \sum_{i=0}^n f_i \cdot \Lambda \cdot \mathbb{A}_i = \Lambda_0 + \sum_{i=0}^n f_i \cdot (\Lambda_i - \Lambda_0) \cdot \mathbb{A}_i \quad (3)$$

where Λ_0 is the thermal conductivity tensor of the matrix, f_i is the volume fraction of the inclusion i , Λ_i its thermal conductivity tensor and \mathbb{A}_i its second order concentration tensor.

In this study, only two phases were considered: the matrix (the air or the dry particle) and only one type of inclusions (respectively the hemp shiv particles or the air in the pores).

3.4 Estimation of the effective thermal conductivity of a particle by inverse method: isotropic consideration

Since the plant aggregates made up a relatively large volume fraction in bulk, the Mori Tanaka approach appeared appropriate to consider the interactions between the inclusions [16]. This homogenization scheme considers heterogeneous medium as an isotropic reinforced matrix (air). Its inclusions behave as isolated inclusions in an infinite matrix. Initially, these inclusions (hemp shiv particles) were considered as spherical. In this case, the thermal conductivity tensor was expressed as:

$$\Lambda_{iso}^{hom} = \Lambda_0 + f_i \cdot (\Lambda_i - \Lambda_0) : \mathbb{A}_i \quad \text{with } \mathbb{A}_i = (\mathbb{I} + \mathbb{S} \Lambda_0)^{-1} (\Lambda_i - \Lambda_0) \quad (4)$$

where \mathbb{S} is the Eshelby tensor, which depends on the shape of the inclusions and the properties of the matrix.

In the case of a spherical inclusion, the Eshelby tensor is given by:

$$S_{ij} = \begin{cases} 0 & \text{if } i \neq j \\ 1 & \text{if } i = j \end{cases} \quad (5)$$

This gives the following analytical formula for the expression of the homogenized thermal conductivity tensor:

$$\Lambda_{iso}^{hom} = \Lambda_0 + \frac{3f_i (\Lambda_i - \Lambda_0) \Lambda_0}{(1 - f_i)\Lambda_i + (2 + f_i)\Lambda_0} \quad (6)$$

with $\Lambda_{iso}^{hom} = \lambda_{iso}^{hom} \mathbb{I}$

where \mathbb{I} is the second order unit tensor.

The scalar thermal conductivity λ_{iso}^{hom} can be deduced directly from this formula since the distribution is isotropic. The analytical expression obtained is then inverted to lead to the following result:

$$\lambda_i = \frac{\lambda_0 (2 - 3f_i) - 2 \lambda_{hom}}{\lambda_{hom} - \lambda_{hom} (1 + 3f_i)} \quad (7)$$

where λ_0 is the thermal conductivity of the air ($0.026 \text{ W}\cdot\text{m}^{-1}\text{K}^{-1}$ at 20°C), f_i the volume fraction of the hemp particles, and λ_{hom} the measured thermal conductivity of the bulk.

The relation (6) enabled us to determine the thermal conductivity for a single particle of hemp shiv, considering spherical pores and the solid skeleton as the matrix. Expression (7) was calculated to determine the thermal conductivity for a single particle of hemp shiv by an inverse method.

3.5 Estimation of the effective thermal conductivity of a particle by inverse method: anisotropic consideration

Because of the oriented thermal conduction of the samples studied, a transversely isotropic thermal conductivity tensor $\Lambda_{\text{ani}}^{\text{hom}}$ is considered here. According Nguyen [6], the thermal conductivity tensor at the particle scale, $\Lambda_{\text{ani}}^{\text{hom}}$, is given by:

$$\Lambda_{\text{ani}}^{\text{hom}} = \begin{pmatrix} \lambda_N^{hp} & 0 & 0 \\ 0 & \lambda_T^{hp} & 0 \\ 0 & 0 & \lambda_T^{hp} \end{pmatrix} \quad (8)$$

The components are detailed in the work of Nguyen [6]. For the specific case of dry particles used here and according to the results of the author, a value of 1.57 was taken for the anisotropy parameter. This coefficient characterizes the ratio between the longitudinal and transverse conductivities of the particle expressed as:

$$\gamma_{\text{dry}}^{hp} = \frac{\lambda_N^{hp}}{\lambda_T^{hp}} = 1.57 \quad (9)$$

This method is more advanced than the one presented in section 3.4. as it takes the anisotropy of the particle into account, which is more in agreement with experimental results of the literature [17]. It made it possible to give the values of the components of the tensor $\Lambda_{\text{ani}}^{\text{hom}}$ at the particle scale by applying an inverse method.

3.6 A new method to estimate the effective thermal conductivity of a particle based on its chemical composition

By considering randomly oriented, transversely elongated or isotropic inclusions, the thermal conductivity tensor of hemp shiv particles can be obtained [7]. The conductivity tensor and its components are expressed as:

$$\Lambda_{cyl}^{hom} = \begin{bmatrix} \lambda_N^{hom} & 0 & 0 \\ 0 & \lambda_T^{hom} & 0 \\ 0 & 0 & \lambda_T^{hom} \end{bmatrix} \quad (10)$$

with :

$$\lambda_N^{hom} = f_i \lambda_0 + (1 - f_i) \lambda_i \quad (11)$$

$$\lambda_N^{hom} = \left(\frac{f_i}{\lambda_i + \lambda_0} + \frac{1 - f_i}{2 \lambda_0} \right)^{-1} - \lambda_0 \quad (12)$$

where λ_0 is the thermal conductivity of the solid skeleton, f_i the volume fraction of air (particular porosity), and λ_{hom} the homogenized thermal conductivity of the hemp particle.

In this part, we are concerned with the thermal conductivity of the particulate solid skeleton since it is not referenced in the literature and is difficult to measure. First, we need to focus on the microscopic description of heat transfer modes in solids. In this context, conductive energy transfer occurs mainly through electronic or vibrational excitations. In the case of a dielectric (insulator), as the electrons are strongly bound to the nucleus, thermal energy is only transmitted by vibration of the crystal lattice. These vibratory motions are generally described by elastic waves and associated with quasi-particles called phonons. Thermal conductivity is therefore governed by the mean free path of phonons [18]. The more ordered the atomic lattice (crystalline), the higher the conductivity. According to section 2.3, the solid skeleton of hemp shiv is mainly composed of cellulose, hemicellulose and lignin. However, while the rate of crystallinity observed in cellulose is about 80% [19], hemicellulose and lignin are essentially amorphous polymers [20]. In short, heat energy may be assumed to be transmitted only by cellulose. In the first instance, we considered that the solid skeleton was made of 100% cellulose. We were thus able to apply the formula that relates density and thermal conductivity as a function of temperature [21] for cellulose fibres (CF) from the formula for cellulose nano-fibres (CNF), respectively λ_{CF} and λ_{CNF} :

$$\lambda_{CNF} = (0.0787 + 2.73 \cdot 10^{-3} T - 7.6749 \cdot 10^{-6} T^2 + 8.4637 \cdot 10^{-9} T^3) \left(\frac{\rho_{sample}}{\rho_0} \right)^2 \quad (13)$$

$$\lambda_{CF} = 1.065 \lambda_{CNF} \quad (14)$$

where T is the temperature (in Kelvins) of the sample, $\rho_{sample} = 1543 \text{ kg.m}^{-3}$ its density (average density of cellulose [10]) and $\rho_0 = 1340 \text{ kg.m}^{-3}$ (reference value).

Following through on the strong assumption described earlier, the value of λ_0 will be taken to be equal to λ_{CF} . Thus, we theoretically recovered the value of the thermal conductivity of the particles, using the assumptions of, firstly, spherical pores and, secondly, cylindrical pores.

4. RESULTS AND DISCUSSION

4.1 Hemp shiv considered as a spherical inclusion

The results of the thermal conductivity (as a scalar value λ_{iso}^{hp}) for a single particle of hemp shiv and under the assumption of a spherical inclusions are shown in Table 2.

Table 2. Values of our spherical inclusion models compared to the literature

λ_{iso}^{hp} (W.m ⁻¹ .K ⁻¹)	Reference	Working methods
0.085 ± 0.014	Experimental method	Dry particles in bulk, temperature-controlled environment (relation 7)
0.0953	Theoretical method	From the chemical composition and particulate porosity (relation 6)
0.102	Cérézo [3]	Particles in bulk, other precautions not specified
0.1	Pham [22]	Not specified
0.110	Dartois [23]	Iterative micromechanical modelling

The relative difference between our theoretical and experimental values is 11 %, which indicates that they are consistent. Although, the limited number of references made comparison difficult, we can see that the relative difference between the average literature value of 0.104 W.m⁻¹.K⁻¹ and our values is 9% for the theoretical value and 23 % for the experimental one. This larger discrepancy may have several origins: if the bulk particle measurement by Cérézo [3] was conducted on non-dry particles, the adsorbed water increased the value of the particulate thermal conductivity and the measurement method could also have a significant effect on the measured values [24]. In addition, the origin of the particle and the variability this implies is likely to have had a significant impact on the value and will be discussed in section 4.5.

Nevertheless, whether they result from the theoretical or the experimental method, our mean values are systematically below those of the literature. It should be noted that Pham [23] concludes that homogenization models systematically overestimate the thermal conductivity of hemp concrete. However, these models use the thermal conductivity value of a single particle, as seen in Section 3. It may therefore be possible to suggest that overestimation comes from a non-significant value. Thus, according to our results, the value of 0.1 W.m⁻¹.K⁻¹ for a single dry particle of hemp shiv is probably above the real value.

4.2 Hemp shiv considered as a cylindrical inclusion

The thermal conductivity tensor for a single particle of hemp shiv, under the assumption of cylindrical inclusions, is composed of two types of values: a normal component and a tangential component as detailed in Fig. 5. The transversal conductivity λ_T^{hp} is perpendicular to the plane of the hemp particle and is assumed to be invariant under rotation by angle θ , according to the cylindrical shape. The normal conductivity λ_N^{hp} is oriented parallel to the direction of the tubular pores.

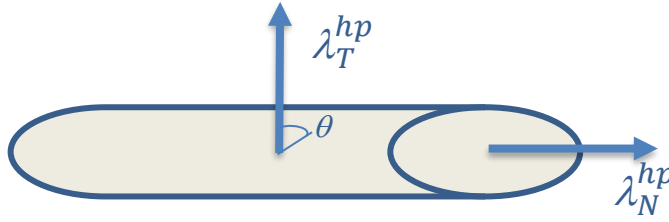


Fig 5. Components of the conductivity tensor in the case of the cylindrical inclusion hypothesis

Our results are summarized and compared with the literature in Table 3.

Table 3. Values of our cylindrical inclusion models compared to the literature

λ_T^{hp} (W.m ⁻¹ .K ⁻¹)	λ_N^{hp} (W.m ⁻¹ .K ⁻¹)	Reference	Working methods
0.067 ± 0.009	0.105 ± 0.013	Experimental method	Dry particles in bulk, temperature-controlled environment
0.0805	0.122	Theoretical method	From the chemical composition and particulate porosity
0.095	0.112	Dartois [23]	Iterative micromechanical modelling
0.088	0.122	Tran-Le[7]	Homogenization from a calibrated value for the conductivity of the solid phase

Transversal component. The relative difference between our theoretical and experimental values is 17 %, which indicates they are fairly consistent. Furthermore, the relative difference between the average literature value of 0.915 W.m⁻¹.K⁻¹ and our values is 14% for the theoretical value and 37 % for the experimental one. Our theoretical

value is therefore rather in line with the results of the literature, while our experimental value is clearly different. This discrepancy, which is not significant for the other values, could be explained by a combination of several factors: impact of the measurement method, value of the anisotropy coefficient of our sample different from the one considered, overestimation of the literature values. Another bias is discussed in section 4.4.

Normal component. The relative difference between our theoretical and experimental values is 14 %, which indicates again that they are fairly consistent. Furthermore, the difference between the average literature value of $0.117 \text{ W.m}^{-1}\text{.K}^{-1}$ and our values is 4% for the theoretical value and 19 % for the experimental one. Our theoretical value is therefore in very good agreement with the results of the literature, while our experimental value is slightly lower.

4.3 Effects of compaction

Thermal conductivity measurements on bulk particles were repeated using loose or compacted samples by a maximum of 30% of the initial volume. The particle thermal conductivity values deduced from these measurements by the inverse method are given in Table 4.

Table 4. Comparison of values with and without compaction

Case studied	$\lambda^{hp} \text{ (W.m}^{-1}\text{.K}^{-1})$ from loose particles	$\lambda^{hp} \text{ (W.m}^{-1}\text{.K}^{-1})$ from compacted particles	Relative Difference
Isotropic	0.083 ± 0.008	0.086 ± 0.014	3%
Transversal	0.067 ± 0.007	0.066 ± 0.009	2%
Normal	0.105 ± 0.010	0.104 ± 0.015	2%

The difference between the particle thermal conductivity values with and without compaction is very small. Furthermore, no general tendency could be identified for any case. We can therefore conclude that the particulate thermal conductivity may not be impacted by the compaction of the aggregates. It is deduced that the solid skeleton is not altered by compaction forces. This is in agreement with the study by Antlauf [21], which demonstrates that no significant irreversible changes occur for a cellulose structure up to a pressure of about 0.9 GPa. What is true for an exclusively cellulosic skeleton could therefore be extended to a solid skeleton consisting mainly of cellulose, hemicellulose and lignin, under the conditions of our study.

4.4 Back to the solid skeleton hypothesis

A general trend emerges from the comparison of our experimental values with the literature references and with our theoretical results. Our experimental results are consistently lower. A first explanation could be that an offset was induced by the measurement method. However, to date, it is difficult to quantify this discrepancy [24]. Another possible explanation is the fact that the solid skeleton was considered to be composed exclusively of cellulose. This polymer, which is semi-crystalline, participates significantly in heat conduction, much more than an amorphous solid [25]. However, hemicellulose and lignin, which are the two other main components of the plant particle skeleton, are amorphous. In other words, we have overestimated the thermal conductivity of the solid skeleton and, by extension, that of the plant aggregate with this assumption. It would be useful to have access to the thermal conductivity values of the other constituents. This would allow the results of our theoretical method to be refined. Nevertheless, our value for the thermal conductivity of the solid skeleton is consistent with the only estimated value we found in the literature (Table 5).

Table 5. Comparison of thermal conductivity values of the solid skeleton

λ_s^{hp} (W.m ⁻¹ .K ⁻¹)	Reference	Working methods
0.6109	Theoretical method	From the results of Stelte [20]
0.5755	Nguyen [6]	Inverse analysis

4.5 Variability of particle thermal conductivity

The key strength of the theoretical method is that it quantifies the influence of certain intrinsic parameters of hemp shiv on its particulate thermal conductivity. Thus, the available data [10] on the variability of the cellulose density and the variability of the intra-particle porosity enabled us to set two bounds for theoretical values (Table 6).

Table 6. Extreme values considered for the calculation of the theoretical bounds

Variable parameter	Maximum	Minimum
Density of cellulose (kg.m⁻³)	1450	1630
Particulate porosity (%)	88.30	78.30

These maximum and minimum values obtained were compared with our own values and with those of the literature. The results for the different cases studied in this paper (isotropic and anisotropic) are presented in Figs. 6, 7 and 8.

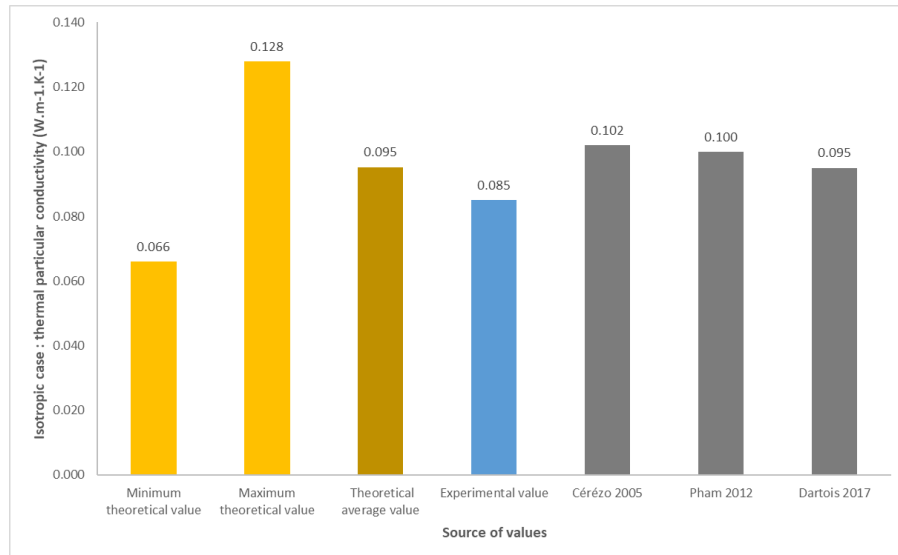


Fig 6. Isotropic case: extreme theoretical values compared to our results and those of the literature [3], [22], [23].

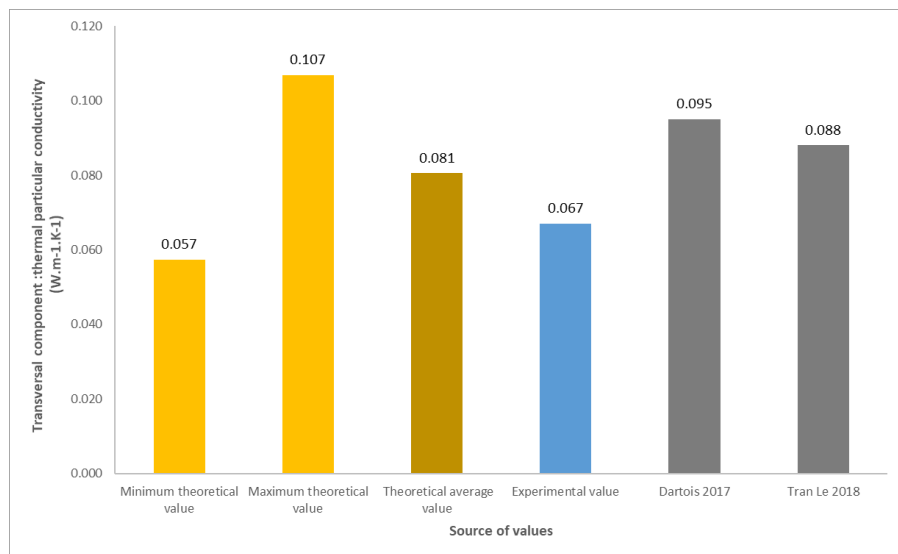


Fig 7. Transversal component: extreme theoretical values compared to our results and those of the literature [7], [23].

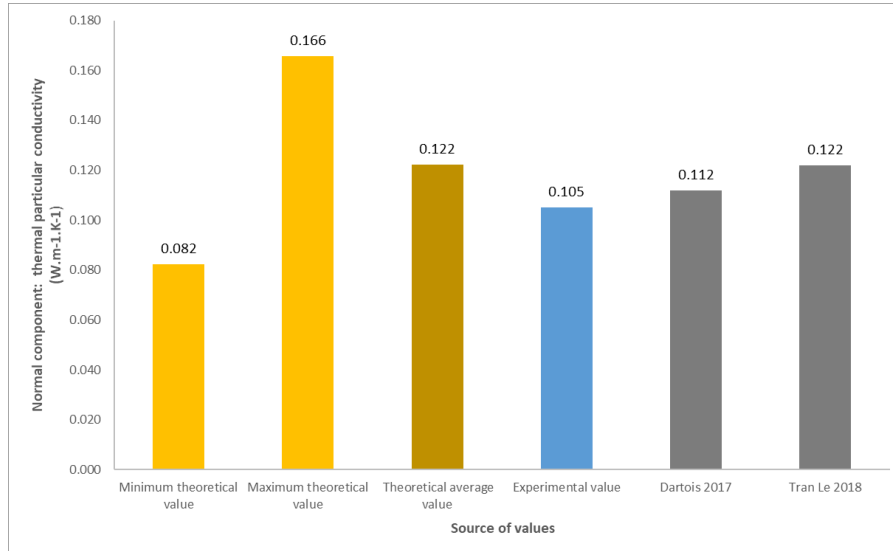


Fig 8. Normal component: extreme theoretical values compared to our results and those of the literature [7], [23].

It is worth noting that the theoretical bounds allow us to include, on the one hand, our experimental value and, on the other hand, those from the literature. Thus, we were able to define a range of particle thermal conductivities for hemp shiv, in both the isotropic and anisotropic cases. This range takes account of the main parameters (particle porosity and density of the solid skeleton), which depend on the very varied conditions of the plant cultivation (growth, place and time of harvesting) and on the nature of the aggregate (cellulosic composition).

Finally, by calculating the ratio of the extreme theoretical thermal conductivities obtained in the anisotropic case, we recover an anisotropy factor consistent with the literature (Table 7).

Table 7. Anisotropy parameter γ^{hp} of hemp shive particle

γ^{hp}	Reference
1.57	Experimental method
1.55	Theoretical method (with maximum values)
1.44	Theoretical method (with minimum values)
1.5	Nguyen [6]
1.18	Dartois [23]
1.53	Tran-Le [7]

Our results are consistent with those of the literature and support a particle anisotropy factor of around 1.5 for hemp shiv particles. This value is important because it determines the anisotropy at the particle scale, but also impacts the anisotropy at the material and wall scale for composites made from hemp shiv. The application process of these materials (sprayed or compacted) leads to a preferential orientation of the particles and thus to anisotropy in the thermal behaviour of the wall.

5. CONCLUSION

In this study, a better knowledge and a better definition of the thermal properties of hemp-based building materials were being sought. We demonstrated that, in order to reliably predict their behaviour, a deeper understanding of the intrinsic properties of the plant aggregate is required. The development of methods to access local and microstructure-specific values is also essential.

Thus, homogenization techniques have been applied to determine the conductivity tensor at the scale of the dry hemp shiv particle from its microstructural characteristics (morphology of the particle and properties of the constituents). The isotropic (which can be seen as a simplified study) and anisotropic cases have been systematically treated. First, an experimental method was proposed. Unlike some values in the literature for which we did not have the test conditions, the protocol here has been described in detail, and the conditions controlled and explained. The aim of this was to be able to propose a reproducible method. Nevertheless, the values obtained experimentally were below the values of the literature. We therefore suspected an overestimation of the latter, since we were unable to assess the methods of determination.

Furthermore, a new method for estimating the particulate thermal conductivity has been developed. Using the results of a recent study [21] the hypothesis of a solid skeleton consisting exclusively of cellulose has been proposed. This assumption has been discussed and could lead to an overestimation of thermal conductivity values. Nevertheless, the strong point of this method is to be able to quantify the impact of the variability of the resource, coming from the plant world, on the particulate thermal conductivity. Our study also shows that the impact of compaction on the particle thermal conductivity value can be negligible. In addition, it demonstrates that the anisotropy parameter at the particle scale, considering heat transfer, is around 1.5. This determining factor has to be taken into account at the scale of the particle used as a bulk insulator or incorporated in a binding matrix, as the insulating layer at the wall scale will also be thermally anisotropic as a result.

Finally, the proposed methods were found to be consistent with the results of the literature. This highlights their relevance. Given the lack of data on the thermal conductivity of hemp shiv particles, this study provides new ways of assessing this value, which is impossible to measure directly with current measurement techniques. The experimental method used here is relatively easy to implement although it uses complex micromechanical modelling results. This inverse method therefore deserves to be applied to other aggregates from agricultural co-products. The theoretical method has also demonstrated its applicability by providing the range of values within which our theoretical, experimental and literature values fall. The same models should be applied first to other types of hemp shiv to evaluate the impact of the variability of this plant-aggregate. It could then be extended to other raw materials provided that a minimum of data is available on their microstructure. Further work will therefore focus on extending these methods to other aggregates. It will also be useful to consider the impact of adsorbed water on thermal conductivity at the particle level. This study provides thus new perspectives for determining the particulate thermal conductivity value, which is essential for predicting the insulating properties of bio-based materials.

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