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A Multiscale Model for the Effective Thermal Conductivity Tensor of a Stratified Composite Material[1]

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Thermal modeling of composites has three essential objectives: (i) comprehension of their thermal behavior; (ii) composite scaling in order to satisfy specific requirements; and (iii) optimal analysis of experimental results from thermal characterization. For a complete study of the material, each of these three points must be taken into account at the fiber scale ($\approx 10\,\mu$m), the yarn scale (≈ 1 mm), and the composite scale (≈ 10 cm). This work presents multi-scale modeling of the effective thermal conductivity tensor of a stratified composite material made from carbon fibers, phenolic resin, and carbon loads. The longitudinal and transverse thermal conductivities of the yarn are computed from optical microscopic imaging of the material. The isotropic thermal conductivity of the loaded matrix is computed by the Bruggeman model. Then, the thermal conductivity tensor is determined by a finite element method taking into account the morphology of the fabric. Computed values are close to experimental values measured by classical methods. Finally, analytical relations are proposed to obtain an efficient model which can be used in a multiphenomenon simulation of the composite structure.

KEY WORDS: composite material; effective property; heat transfer; specific heat; thermal conductivity.

1. INTRODUCTION

Woven-fabric/polymer matrix composites have been extensively studied because of the relative ease and low cost of their manufacturing. Many

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industrial issues are due to their very good specific mechanical properties. However, for thermal protection applications, charring ablators are most widely used. Phenolic, epoxy, or silicon resins with glass or carbon fibers are generally used. In this case, the thermal properties of the composites are very important.

The purpose of this paper is to describe one example of modeling of the effective density, effective specific heat, and effective thermal conductivity tensor of a carbon woven-fabric/phenolic matrix composite. The strategy is to take into account the measured properties of each constituent (fibers, matrix, fillers) and the woven-fabric of the composite. Both analytical and numerical approaches are described. The measured data used in the different models and the computation results are presented. Then the difference between these results and experimental data on the composites is discussed.

2. EXPERIMENTAL DATA FOR THE COMPOSITE AND ITS CONSTITUENTS

2.1. Morphological Characteristics of the Composite Material

The material under consideration is made from base rayon carbon fibers (Fig. 1, medium diameter: $d = 12\,\mu\text{m}$; volumetric fraction: $\alpha_{\text{f,C}} = 0.42$) and phenolic resin matrix. Carbon loads (volumetric fraction: $\alpha_{\text{l,C}} = 0.06$) are imbedded into the inter-yarn matrix, but not into the intra-yarn matrix. The porosity ($\varepsilon = 0.02$) is uniformly distributed into the material. The yarns, identical for chain and weft directions (720 fibers per yarn, volumetric fraction of fibers into the yarn: $\alpha_{\text{f,Y}} = 0.6$), are woven in order to make plies of satin 8/3 (Fig. 2). These plies are then put on top of each other, without disorientation, to constitute the stratified composite.

2.2. Thermophysical Properties of the Constituents

The density of the fibers, the resin, and the loads: $\rho_{\text{f}} = 1800\,\text{kg}\cdot\text{m}^{-3}$, $\rho_{\text{r}} = 1300\,\text{kg}\cdot\text{m}^{-3}$, and $\rho_{\text{l}} = 2200\,\text{kg}\cdot\text{m}^{-3}$, are measured by helium pycnometry (commercial apparatus: Accupyc1330, Micromeritics) with an uncertainty less than 3%. The thermal conductivities (longitudinal: $\lambda_{\text{f,L}} = 6\,\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ and transverse: $\lambda_{\text{f,T}} = 1.6\,\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$) of the fibers are obtained from thermal diffusivity measurements, at room temperature, by a photothermal microscopy method [1]. These very difficult measurements are performed with an estimated uncertainty of 20%. The thermal conductivity of the resin, $\lambda_{\text{r}} = 0.4\,\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, is also determined from diffusivity measurements realized on a bulk sample by the flash method [2]. The thermal

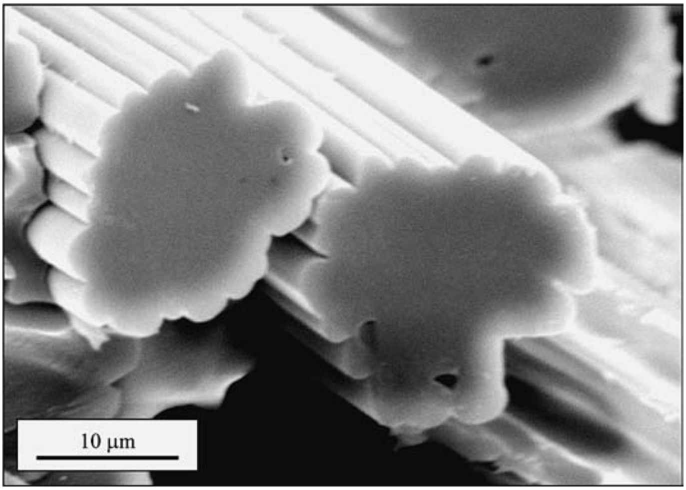

Fig. 1. Rayon-based carbon fiber.

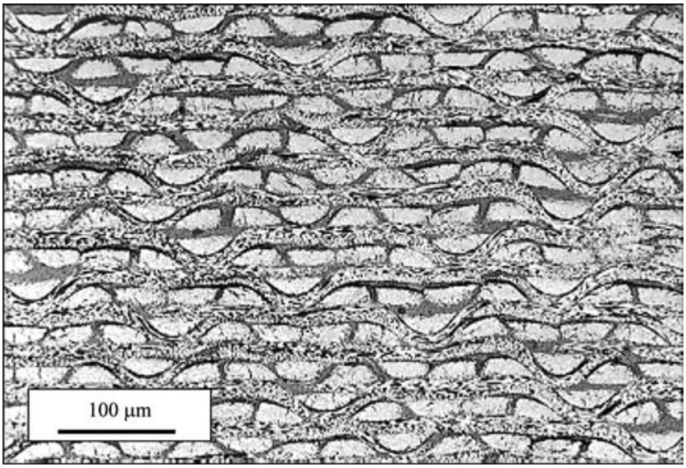

Fig. 2. Composite material under consideration in this work. Chain yarns cross-sections and weft yarns in horizontal direction.

conductivity of the loads, $\lambda_l = 100\,\mathrm{W \cdot m^{-1} \cdot K^{-1}}$, is equally determined from diffusivity measurements on loaded matrix samples at various concentrations. This high conductivity value is the consequence of a very high temperature treatment realized on the carbon loads. These measurements are performed with a large uncertainty of 20%. Finally, the specific heat of carbon fibers, phenolic resin, and carbon loads are obtained by differential scanning calorimetry (commercial apparatus: DSC, Setaram) with an estimated uncertainty of 10%: $c_f = 750\,J\cdot kg^{-1}\cdot K^{-1}$, $c_l = 1050\,J\cdot kg^{-1}\cdot K^{-1}$, and $c_r = 600\,J\cdot kg^{-1}\cdot K^{-1}$.

2.3. Thermophysical Properties of the Composite Material

The thermophysical properties of the composite are determined with the same techniques. The density, $\rho_c = 1490\,kg\cdot m^{-3}$ is measured with an uncertainty of 3%. The thermal conductivities, $\lambda_{c,||} = 1.81\,W\cdot m^{-1}\cdot K^{-1}$ in the parallel direction and $\lambda_{c,\perp} = 1.16\,W\cdot m^{-1}\cdot K^{-1}$ in the perpendicular direction, are determined from diffusivity measurements with an uncertainty of 10%. Finally, the specific heat, $c_c = 900\,J\cdot kg^{-1}\cdot K^{-1}$ is measured by DSC with an uncertainty of 10%.

3. ANALYTICAL MODEL FOR THE EFFECTIVE THERMOPHYSICAL PROPERTIES

3.1. Volumetric Fractions

The volumetric fraction of the fibers into the yarns, $\alpha_{f,Y} = 0.60$, is obtained by analysis of yarn crosssection photographs. Consequently, the volumetric fraction of the resin into the yarns is simply determined by: $\alpha_{r,Y} = 1 - \alpha_{f,Y} = 0.40$. The volumetric fraction of the loads into the matrix is also determined using algebrical relations between the volumetric fractions of all the constituents:

$$\alpha_{l,M} = \frac{\alpha_{l,C}}{1 - \frac{\alpha_{f,C}}{\alpha_{f,Y}} - \varepsilon} \tag{1}$$

or numerically: $\alpha_{l,M} = 0.21$. Consequently, the volumetric fraction of the resin into the loaded matrix is simply determined by: $\alpha_{r,M} = 1 - \alpha_{l,M} = 0.79$, the volumetric fraction of the resin into the composite by: $\alpha_{r,C} = 1 - \alpha_{f,C} - \alpha_{l,C} - \varepsilon = 0.50$, the volumetric fraction of the yarns into the composite by: $\alpha_{Y,C} = \alpha_{f,C}/\alpha_{f,Y} = 0.70$, and the volumetric fraction of the loaded matrix into the composite by: $\alpha_{M,C} = 1 - \alpha_{Y,C} - \varepsilon = 0.28$.

3.2. Effective Density of the Composite

The effective density of a material composed of N constituents of volumetric fraction α_i is given by: $\rho = \sum_{i=1}^{N} \alpha_i \rho_i$. Using this definition to determine the effective density of the yarns and the loaded matrix, the effective density of the composite is given by

$$\rho_C = \alpha_{Y,C}(\alpha_{f,Y}\rho_f + \alpha_{r,y}\rho_r) + \alpha_{M,C}(\alpha_{l,M}\rho_l + \alpha_{r,M}\rho_r) \tag{2}$$

or numerically: $\rho_C = 1538\,\mathrm{kg \cdot m^{-3}}$.

3.3. Effective Specific Heat of the Composite

The effective specific heat of a material composed of N constituents of volumetric fraction α_i is given by: $c = \sum_{i=1}^{N} \alpha_i \rho_i c_i / \sum_{i=1}^{N} \alpha_i \rho_i$. Using this definition to determine the effective specific heat of the yarns and the loaded matrix, the effective specific heat of the composite is given by

$$c_C = \frac{\alpha_{Y,C}(\alpha_{f,Y}\rho_f c_f + \alpha_{r,Y}\rho_r c_r) + \alpha_{M,C}(\alpha_{l,M}\rho_l c_l + \alpha_{r,M}\rho_r c_r)}{\alpha_{Y,C}(\alpha_{f,Y}\rho_f + \alpha_{r,Y}\rho_r) + \alpha_{M,C}(\alpha_{l,M}\rho_l + \alpha_{r,M}\rho_r)} \tag{3}$$

or numerically: $c_c = 864\,\mathrm{J \cdot kg^{-1} \cdot K^{-1}}$.

4. NUMERICAL MODEL FOR THE EFFECTIVE THERMAL CONDUCTIVTY TENSOR

4.1. Effective Longitudinal and Transverse Thermal Conductivities of the Yarns

The fibers being aligned in the yarns, their effective longitudinal thermal conductivity is simply determined by a parallel model:

$$\lambda_{Y,L} = \alpha_{f,Y}\lambda_{f,L} + \alpha_{r,Y}\lambda_r \tag{4}$$

or numerically: $\lambda_{Y,L} = 3.76\,\mathrm{W \cdot m^{-1} \cdot K^{-1}}$. The effective transverse thermal conductivity can be determined by several methods [3]. A range for this property is given by the Hashin–Shtriktman model [4]: $0.85 < \lambda_{Y,T} < 0.98\,\mathrm{W \cdot m^{-1} \cdot K^{-1}}$, whereas the Rayleigh [5] and Bruggeman [6] models can also be used to provide approximate values: $\lambda_{Y,T} = 0.85\,\mathrm{W \cdot m^{-1} \cdot K^{-1}}$ and $\lambda_{Y,T} = 0.89\,\mathrm{W \cdot m^{-1} \cdot K^{-1}}$. Moreover, the cross-section photographs used to determine the volumetric fraction of the fibers into the yarns can be used to compute their effective transverse thermal conductivity by a direct method. In this method, a "hot" temperature $T_H = 1$ is imposed on one boundary of the medium, a "cold" temperature $T_C = 0$ is imposed on the opposite boundary, and an isolation condition is imposed on the other two boundaries (Fig. 3), in order to compute the temperature field in the material and to deduce the effective conductivity by the relation,

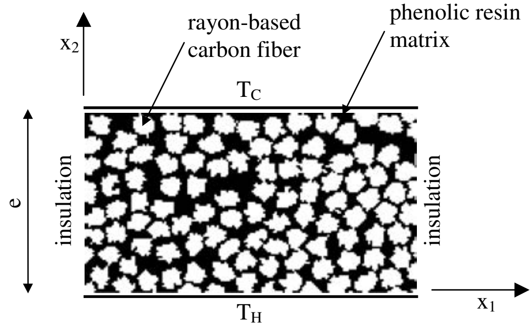

Fig. 3. Determination of the effective transverse thermal conductivity of the yarns by direct method using yarn cross-section photographs.

$$\lambda = \frac{\Phi e}{T_H - T_C} \tag{5}$$

where Φ represents the heat flow and e is the distance between the imposed temperature boundaries. Using this method, the transverse thermal conductivity of the yarn can be evaluated: $\lambda_{Y,T} = 0.89\,W \cdot m^{-1} \cdot K^{-1}$. Finally, the direct method can also be applied on regular arrays of cylinders (Fig. 4) [7]. These computations lead, for a square array,to: $\lambda_{Y,T} = 0.86\,W \cdot m^{-1} \cdot K^{-1}$, and for a hexagonal array,to: $\lambda_{Y,T} = 0.85\,W \cdot m^{-1} \cdot K^{-1}$. All the values computed for the effective transverse thermal conductivity of the yarns are very close. For its simplicity, the Bruggeman model has been chosen to calculate $\lambda_{Y,T}$:

$$\lambda_{Y,T} = \lambda_r \frac{(1-\alpha_{f,Y})^2\left(\frac{\lambda_{f,T}}{\lambda_r}-1\right)^2 + \frac{2\lambda_{f,T}}{\lambda_r} - \sqrt{\left[(1-\alpha_{f,Y})^2\left(\frac{\lambda_{f,T}}{\lambda_r}-1\right)^2 + \frac{2\lambda_{f,T}}{\lambda_r}\right]^2 - \left(\frac{2\lambda_{f,T}}{\lambda_r}\right)^2}}{2} \tag{6}$$

This relation leads to: $\lambda_{Y,T} = 0.89\,W \cdot m^{-1} \cdot K^{-1}$.

4.2. Effective Thermal Conductivity of the Loaded Matrix

The volumetric fraction of loads being relatively low ($\alpha_{l,M} = 0.21$), the effective thermal conductivity of the loaded matrix can be determined by the Maxwell–Eucken model [8] to a good approximation:

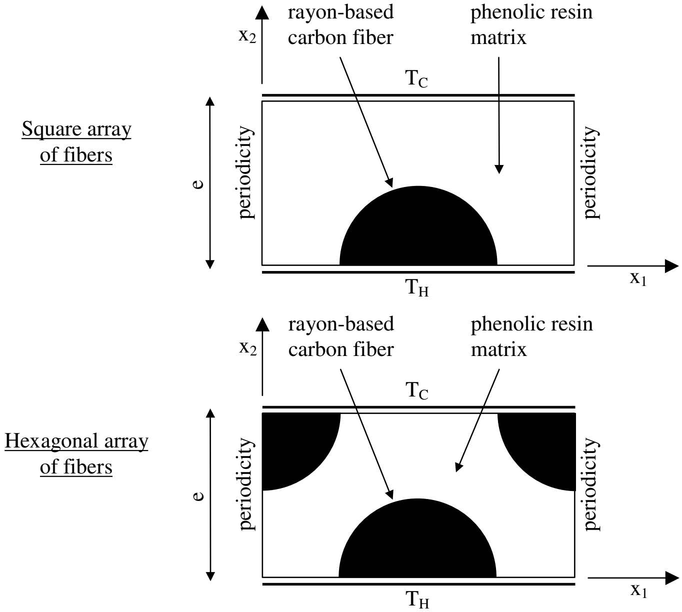

Fig. 4. Determination of the effective transverse thermal conductivity of the yarns by direct method using regular arrays of cylinders.

$$\lambda_M = \lambda_r \frac{1 + 2\alpha_{1,M} \frac{1-\frac{\lambda_r}{\lambda_{ch}}}{1+2\frac{\lambda_r}{\lambda_{ch}}}}{1 - \alpha_{1,M} \frac{1-\frac{\lambda_r}{\lambda_{ch}}}{1+2\frac{\lambda_r}{\lambda_{ch}}}} \tag{7}$$

This relation leads to: $\lambda_M = 0.72\,\mathrm{W \cdot m^{-1} \cdot K^{-1}}$.

4.3. Effective Thermal Conductivity Tensor of the Composite

The calculation of the effective thermal conductivity of the composite is realized in two steps. First, the material is assumed to be non-porous. Then, the volumetric fraction of yarn in the composite becomes: $\bar{\alpha}_{Y,C} = \alpha_{Y,C}/(1 - \varepsilon) = 0.71$. The thermal conductivity tensor is determined by the direct method applied on a periodic pattern of the ply (Fig.5). The morphology (yarn cross section and yarn spacing) of the latest is determined in order to represent the real material and to correspond with the volumetric fraction of the yarns into the composite. The

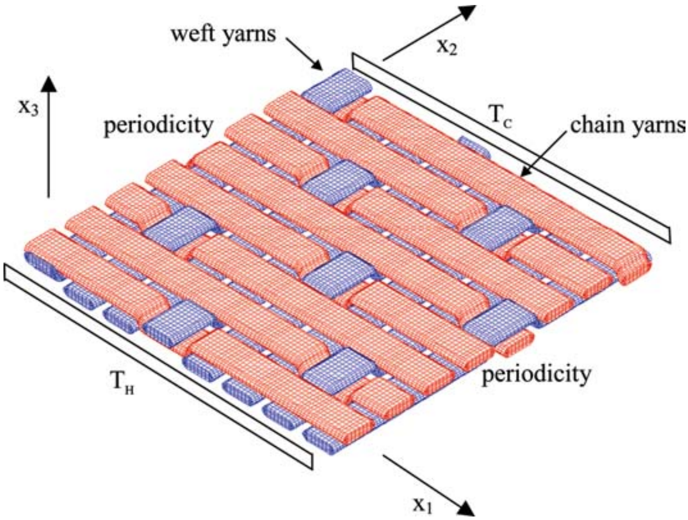

Fig. 5. Determination of the effective thermal conductivity tensor of the composite by direct method applied on one periodic pattern of the ply.

thermal problem is solved by the finite element method (Software CAST3M, CEA, France). The temperature difference $T_H - T_C$ is successively applied along the three directions. From these three numerical experiments, the effective thermal conductivity tensor of the composite is computed by the relation [9],

$$\lambda_{i,j} = \frac{\langle \Phi_i \rangle}{\langle \partial T / \partial x_j \rangle} \tag{8}$$

where $\langle \Phi_i \rangle$ represents the mean heat flow parallel to the x_i direction and $\langle \partial T / \partial x_j \rangle$ is the mean temperature gradient in the x_j direction.

Finally, the material is assumed to be porous. The porosity being low ($\varepsilon = 0.02$), the Maxwell model [10] provides a good approximation of the effective composite thermal conductivities:

$$\lambda_{C,\parallel} = \bar{\lambda}_{C,\parallel} 2 \frac{1-\varepsilon}{2+\varepsilon} \quad \lambda_{C,\perp} = \bar{\lambda}_{C,\perp} 2 \frac{1-\varepsilon}{2+\varepsilon} \tag{9}$$

These relations lead to: $\lambda_{C,\parallel} = 1.78\,\mathrm{W \cdot m^{-1} \cdot K^{-1}}$ and $\lambda_{C,\parallel} = 0.81\,\mathrm{W \cdot m^{-1} \cdot K^{-1}}$.

5. ANALYTICAL MODEL FOR THE EFFECTIVE THERMAL CONDUCTIVITY TENSOR

The model developed in the previous section uses analytical relations to compute the effective thermal conductivities of the yarns and the loaded matrix, and numerical relations to compute the effective thermal conductivity tensor of the composite. The objective of this section is to propose approximate analytical models to compute the effective thermal conductivity tensor of the composite.

5.1. Effective Thermal Conductivity in the Direction Parallel to Ply

First, the material is assumed to be nonporous. The ply is divided into two parts: one containing the yarns parallel to the chain direction, and the other containing the yarns parallel to the weft direction. The thermal conductivity of the first part, in the direction parallel to the chain yarns, is called λ_1, and that of the second part is called λ_2. The thermal conductivity λ_1 can be evaluated by a parallel model,

$$\lambda_1 = \bar{\alpha}_{Y,C}\lambda_{Y,L} + (1-\bar{\alpha}_{Y,C})\lambda_M \tag{10}$$

whereas the conductivity λ_2 can be evaluated by a serial model,

$$\lambda_2 = \frac{\lambda_r\lambda_{Y,T}}{\bar{\alpha}_{Y,C}\lambda_M + (1-\bar{\alpha}_{Y,C})\lambda_{F,T}} \tag{11}$$

The association of both parts in a parallel scheme provides the effective thermal conductivity of the nonporous composite material:

$$\bar{\lambda}_{C,\parallel} = \frac{1}{2}\left[\bar{\alpha}_{Y,C}\lambda_{Y,L} + (1-\bar{\alpha}_{Y,C})\lambda_M + \frac{\lambda_M\lambda_{Y,T}}{\bar{\alpha}_{Y,C}\lambda_M + (1-\bar{\alpha}_{Y,C})\lambda_{Y,T}}\right] \tag{12}$$

Then, using the Maxwell model [10], the effective thermal conductivity of the porous composite material is given by

$$\lambda_{C,\parallel} = \frac{1-\varepsilon}{2+\varepsilon}\left[\bar{\alpha}_{Y,C}\lambda_{F,L} + (1-\bar{\alpha}_{Y,C})\lambda_M + \frac{\lambda_M\lambda_{Y,T}}{\bar{\alpha}_{Y,C}\lambda_M + (1-\bar{\alpha}_{Y,C})\lambda_{Y,T}}\right] \tag{13}$$

These relations lead to: $\lambda_{C,\parallel} = 1.82\,\mathrm{W}\cdot\mathrm{m}^{-1}\cdot\mathrm{K}^{-1}$

5.2. Effective Thermal Conductivity in the Direction Perpendicular to Ply

At the beginning, the material is again assumed to be nonporous. The ply is divided into two parts: one containing the yarns parallel to the chain direction, and the other containing the yarns parallel to the weft direction. Considered in the direction perpendicular to the ply, both parts have the same thermal conductivity: $\bar{\lambda}_{C,\perp}$, which can be evaluated by a parallel model,

$$\bar{\lambda}_{C,\perp} = \bar{\alpha}_{Y,C}\lambda_{Y,T} + (1 - \bar{\alpha}_{Y,C})\lambda_M \tag{14}$$

Then, using the Maxwell model [10], the effective thermal conductivity of the porous composite material is given by

$$\lambda_{C,\parallel} = \frac{1-\varepsilon}{2+\varepsilon}\left[\bar{\alpha}_{Y,C}\lambda_{Y,T} + (1 - \bar{\alpha}_{Y,C})\lambda_M\right] \tag{15}$$

These relations lead to: $\lambda_{C,\parallel} = 0.82\,\mathrm{W \cdot m^{-1} \cdot K^{-1}}$.

6. DISCUSSION

The data of the problem (morphological characteristics and properties of the composite materials) are indicated with associated uncertainties in the first section. These uncertainties on the data lead to uncertainties on the results. For each calculated property, both lower and upper values are computed in order to determine the mean value and uncertainty range (Table I). The bounds of the range are calculated for a minimization or a maximization of the property value due to data uncertainties. For example, a lower value of the composite density is obtained for lower values of constituent densities and volumetric fractions, and a higher value of porosity. As a consequence, the mean value of the uncertainty range may be a bit different from the nominal value calculated in the previous section.

The mean value calculated for the density is a good estimation of the measured value. Nevertheless, due to the relative errors accumulation, the uncertainty attached to this result is twice the measured one. The mean value calculated for the specific heat is also a good estimation of the

Table I. Numerical Results with Absolute Uncertainties

Effective property of the composite material	Measurement	Numerical model	Analytical model
Density, $\rho_c (\mathrm{kg \cdot m^{-3}})$	1490 ± 45	–	1510 ± 89
Specific heat, c_c $(\mathrm{J \cdot kg^{-1} \cdot K^{-1}})$	900 ± 90	–	872 ± 105
Thermal conductivity parallel to ply, $\lambda_{c,\parallel}$ $(\mathrm{W \cdot m^{-1} \cdot K^{-1}})$	1.81 ± 0.18	1.78 ± 0.38	1.82 ± 0.39
Thermal conductivity perpendicular to ply, $\lambda_{c,\perp}$ $(\mathrm{W \cdot m^{-1} \cdot K^{-1}})$	1.16 ± 0.12	0.81 ± 0.13	0.81 ± 0.13

measured value, with a similar uncertainty. For the effective thermal conductivity parallel to ply, the three mean values are similar; the analytical model value is very close to the measured value. Once again, the uncertainties attached to the numerical results are twice that of the measured values.

Finally, for the effective thermal conductivity perpendicular to ply, both mean numerical results are close together but are also lower than the measured value. This difference indicates that the modelled material has a smaller thermal conductivity than the real material. In both analytical and numerical models, as in the real material, the effective longitudinal thermal conductivity of the yarn has very little influence on the effective thermal conductivity perpendicular to the ply; the periodic pattern indicates a few intertwines between chain and weft yarns. This behavior is also observed in the mechanical studies of composite materials [11]. So, the effective thermal conductivity perpendicular to the ply mainly depends on the effective transverse thermal conductivity of the yarns, on the effective thermal conductivity of the loaded matrix, and on the thermal contact between these elements. In both models, the thermal contact between yarns and loaded matrix is assumed to be perfect. Thus, it cannot limit the heat transfer across the material. The relatively small volumetric fraction of the loads in the matrix involves an increase of the resin thermal conductivity but the effective thermal conductivity obtained ($\lambda_M = 0.72\,\mathrm{W \cdot m^{-1} \cdot K^{-1}}$) may not exceed $0.8 - 0.9\mathrm{W \cdot m^{-1} \cdot K^{-1}}$. As a consequence, the difference observed between calculated and measured values of $\lambda_{C,\perp}$ is mainly attributed to a lower transverse effective thermal conductivity of yarns ($\lambda_{Y,T} = 0.89\,\mathrm{W \cdot m^{-1} \cdot K^{-1}}$). This thermal conductivity is calculated with six different models (Section 4.1) that provide similar numerical values (from 0.85 to $0.89\mathrm{W \cdot m^{-1} \cdot K^{-1}}$). Since the thermal conductivity of the resin and the volumetric fraction of fibers into the yarns are better known than the thermal properties of the fiber, it may be concluded that the transverse thermal conductivity of the fiber is probably lower than its true value.

7. CONCLUSION

Both analytical and numerical models have been developed in order to compute the effective density, effective specific heat, and effective thermal conductivity tensor of a composite material. These models were used to calculate the thermophysical properties of a satin 8/3 stratified composite. Good agreement was obtained for all the predicted properties except for the effective thermal conductivity in the perpendicular to ply direction. A discussion has then showed that the difference observed between calculated and measured values may be mainly attributed to a lower transverse thermal conductivity of the yarns or, consequently, of the fibers. This problem will be studied by the thermal characterization of impregnated yarns in future work.

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REFERENCES

1. J. Jumel, F. Lepoutre, J. P. Roger, G. Neuer, M. Cataldi, and F. Enguehart, *Rev. Sci. Instrum.* **74**:537 (2002).
2. A. Degiovanni, *Revue Générale de Thermique* **185**:420 (1977).
3. M. Kaviany, *Principles of Heat Transfer in Porous Media* (Springer, Berlin, 1995).
4. Z. Hashin and S. Shtrikman, *J. Appl. Phys.* **33**:3125 (1962).
5. L. Rayleigh, *Phil. Mag.* **34**:481 (1892).
6. D. Bruggeman, *Ann. Phys.* **24**:636 (1935).
7. J.-M. Goyhénèche, *Congrès SFT 2002,* 03-06/06 Vittel (France) (2002).
8. A. Eucken, *Forsh. Gebiete Ingenieur* **B3**:353 (1932), 6–21.
9. D. Bigaud, J.-M. Goyhénèche, and P. Hamelin, *Composites—Part A: Appl. Sci. Manufacturing* **32**:1443 (2001).
10. J. C. Maxwell, in *A Treatise on Electricity and Magnetism* (Dover Pubs., New York, 1954), pp. 435–441.
11. Cosculluela and J.-M. Goyhénèche, *Congrès Matériaux 2002,* 21–25/10 Tours, France (2002).