Transient Thermal Behavior of Directional Reinforced Composites: Applicability Limits of Homogeneous Property Model

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The parameters governing the transient thermal behavior of directional reinforced composites are identified. The limits of applicability of the homogeneous property model are analyzed considering two typical situations: the diffusivity measurement using the Parker method, and the convective heating of the composites. In these two situations the homogeneous property model is erroneous and may lead to significant errors, which are evaluated.

Nomenclature

a = thermal diffusivity, m^2s^{-1}
b = effusivity, $Jm^{-2}s^{-1/2}K^{-1}$
b_0 = effusivity defined by Eq. (4)
c = volumic heat, $Jm^{-3}K^{-1}$
$\bar{c}$ = mean volumic heat, $Jm^{-3}K^{-1}$
h = convective heat transfer coefficient, $Wm^{-2}K^{-1}$
k = thermal conductivity, $Wm^{-1}K^{-1}$
$\tilde{k}$ = equivalent thermal conductivity, $Wm^{-1}K^{-1}$
L = sample thickness, m
q = heat flux density, Wm^{-2}
R = contact thermal resistance per unit surface of matrix/reinforcement interface, $W^{-1}m^2K$
T = temperature, K
t = time, s
ω = space period of the reinforcement, m
ρ = specific contact thermal resistance of the reinforcement/matrix interface
Σ = matrix/reinforcement contact area per unit volume of composite, m^{-1}
σ = specific contact surface of the reinforcement/matrix interface
τ = reinforcement volume content

Subscripts

c = convection
lim = end of the heating period
M = matrix
R = reinforcement
r = recovery conditions
R/M = reinforcement/matrix
stat = steady-state
w = wall conditions
0 = initial conditions

Introduction

DIRECTIONAL reinforced composites are heterogeneous materials consisting of reinforcement fibers embedded in a matrix. The matrix may be considered homogeneous and isotropic, but not the reinforcement, which consists of highly anisotropic fibers. These fibers are grouped into yarns laid in specific directions. The thermal properties of the matrix and the reinforcement are usually extremely different. The fibers are generally much more conductive than the matrix and play a dominant role in the heat transfer, especially when the main heat flux resulting from the boundary conditions is parallel to one of the reinforcement directions. Carbon-resin composites are probably the most extreme example of this, as the ratio between the conductivity of the two constituents can reach 500. Under these conditions, very large temperature heterogeneities can be observed under severe heating. Figure 1 gives a qualitative example: it is a frame taken from a film showing the isolated rear face of a 4-mm-thick specimen of three-dimensional carbon-carbon composite subjected to a 60-MW m^{-2} laser flux. Warm areas (reinforcement) show up light and cold areas (matrix) show up dark.

The thermal protection materials currently used in missiles are practically all directional reinforced composites, whether they are used as external heat shields for atmospheric re-entry or as nozzles directing the rocket engine combustion gases. These composites are, for example, carbon-carbon, carbon-resin, silica-resin or, more recently, ceramic-ceramic composites made of silica, alumina, zirconia, boron nitride, silicon carbide, etc. While a great deal of mechanical modeling has been done, little has been done about thermal modeling, especially since the transient regime is almost the only way in which these materials are expected to operate. We are concerned only with the transient regime hereafter. In this field, theoretical studies have been going on for about 10 years. This subject is covered in Refs. 1-12, which are only the most recent publications by the authors. On the other hand, experimental data are very scarce.[5,7,12-14]

In practical applications, such as measuring thermal properties or carrying out calculations to predict heating, the engineer assumes that this type of material can be simulated by an equivalent homogeneous medium, the transient-regime thermal properties of which (effusivity and diffusivity) can be deducted from the steady-state properties (volume specific heat and thermal conductivity). Certain of the references mentioned above show that this is an oversimplification. The purpose of the present publication is to emphasize the limits of such a homogenization and to summarize our current knowledge of the transient thermal behavior of the directional reinforced composites. The overriding goal is to create awareness of these problems and to set forth some ideas that will guide future studies in this field.

Steady-State and Transient Thermal Properties

The thermal behavior of a medium in steady-state regime depends on only two independent quantities: thermal conductivity k and volume specific heat c. Homogenizing a

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heterogeneous medium such as the directional reinforced composites consists of measuring or calculating the equivalent thermal conductivity $\bar{k}$ and the mean volume specific heat $\bar{c}$. The first one may be evaluated theoretically by using an electrical analogy, for simple configurations, or by solving the steady-state heat equation for the heterogeneous system. This procedure is universally recognized, although it is subject to caution when Neuman or Fourier conditions are encountered. This particular point will not be covered here. The quantities governing the thermal behavior in the transient regime are thermal diffusivity a and effusivity b. These properties are related to steady-state properties by the relations $a=k/c$ and $b=\sqrt{kc}$. They characterize two transient phenomena, the diffusion and effusion of heat.

Diffusion of Heat in Directional Reinforced Composites

The diffusion of heat in a homogeneous medium is characterized by diffusivity a, which is the only thermal property appearing in the heat equation when the temperature variations in the conductivity are neglected:

$$a\nabla^2 T=\frac{\partial T}{\partial t} \tag{1}$$

Diffusivity used in composite heating calculations very rarely comes from independent measurements of the volume specific heat and thermal conductivity but rather from direct measurements. Most current diffusivity measurements are made by the flash method initially proposed by Parker et al.[15] This method is applied to directional reinforced composites on the assumption that the composites effectively behave as homogeneous media.[16-18] If this procedure is subject to caution, it is of prime importance to study the behavior of these materials under the particular conditions of heating by a pulse heat flux. Parallel studies have been going on in the United States and France for several years, with Taylor[14,19] following an experimental approach and Lafond and Bransier[5,20] and Balageas and Luc[7,21-23] taking a theoretical and experimental approach.

The many numerical simulations (see, especially, Ref. 7) carried out on unidirectional reinforced composite models of the layered type or of the chessboard pattern type (Fig. 2) give rise to the following conclusions:

1) The diffusivity determined by a flash experiment is not constant but varies during the time the measurement is made. It also depends, among other things, on the thickness of the sample.

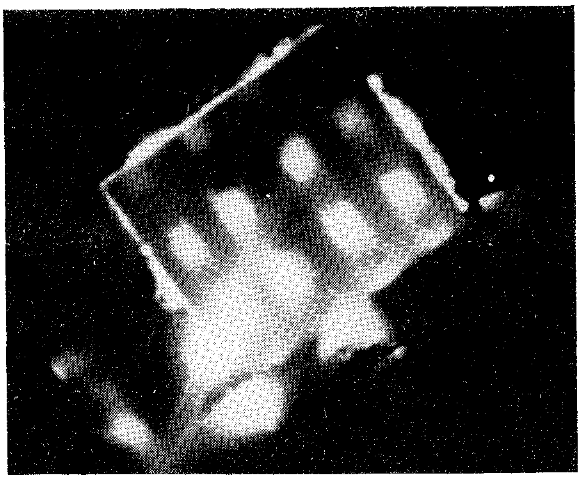

Fig. 1 Frame from a motion picture showing the temperature heterogenities in the insulated rear face of a 4-mm thick three-dimensional carbon-carbon specimen subjected to a laser flux of 60 MW m^{-2}.

2) As a first approximation, the transient thermal behavior of the unidirectional reinforced composites depends only on the following parameters: the volume content of the reinforcement τ; the reinforcement-to-matrix ratios of thermal conductivity ($k_{R/M}$) and of volume specific heat ($c_{R/M}$); and the specific contact surface σ and the specific contact thermal resistance ρ, which characterize the thermal coupling of the two components and are expressed as: $\sigma=L\Sigma$ and $\rho=Rk_{R/M}\Sigma$. Here R is the contact thermal resistance per unit surface of matrix-reinforcement interface, L the thickness of the specimen, and Σ the matrix-reinforcement contact area per unit volume of composite. Σ depends on the shape of the cross section and on the space period ω of the reinforcement. For the composites shown in Fig. 2, $\Sigma=2/\omega$ for the layered composite, $\Sigma=4/\omega$ for a composite with a jointed chessboard reinforcement, and $\Sigma=2\sqrt{\tau}/\omega$ for a composite with an unjointed chessboard reinforcement.

The variation of the apparent diffusivity with the temperature rise of the sample rear face $T/T_{\lim}$ during the measurement is plotted in Figs. 3 and 4 for a directional reinforced composite with a reinforcement volume content of 0.5 and ratios $k_{R/M}=100$ and $c_{R/M}=1$. Figure 3 shows the effects of the specific area σ and Fig. 4 the effect of specific thermal resistance ρ. Reference 7 gives other examples of such curves and specifies the influence of the various parameters.

As the thickness of the specimen becomes large compared to the reinforcement space period (i.e., the mean distance between reinforcements), the apparent diffusivity tends toward the diffusivity deduced from the steady-state properties $a_{\text{stat}}=\bar{k}/\bar{c}$. This seems to be the case for the experimental data given in Ref. 24. The idea of the thick specimen, for which homogenization is justified, thus emerges. On the other hand, the greater the specific contact thermal resistance, the more the components are uncoupled and the less the homogenization is justified. These phenomena become more and more pronounced as the ratio of conductivities $k_{R/M}$ increases, while the ratio of the volumic heats cannot, in practice, vary greatly

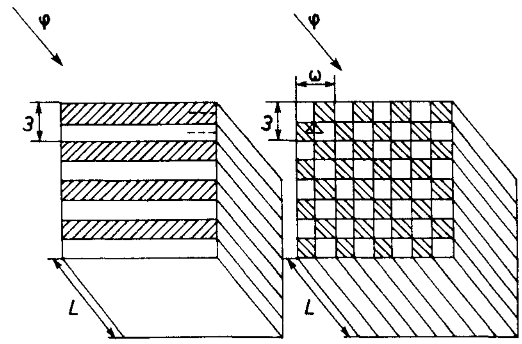

Fig. 2 Types of unidirectional reinforced composites modeled in Refs. 5-7 and 20-23.

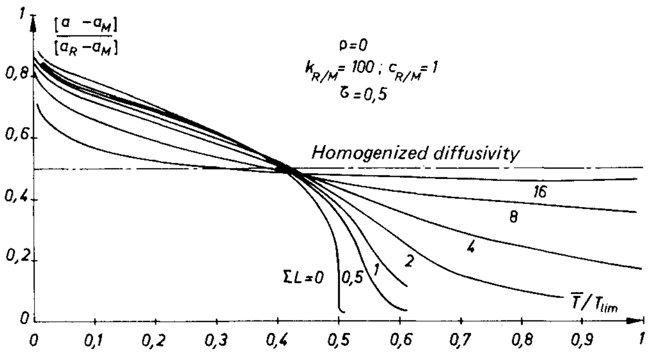

Fig. 3 Apparent diffusivity dependence on temperature of specimen rear face. Influence of the specific contact area σ for a layered composite characterized by $\tau=0.5$, $k_{R/M}=100$, $c_{R/M}=1$, and $\rho=0$.

from unity. This is illustrated in Fig. 5, where the difference between the Parker diffusivity, which corresponds to half the total rise in temperature of the rear face $T/T_{\rm lim}=0.5$, and the diffusivity of the equivalent homogeneous material $a_{\rm stat}$ is shown as a function of the ratio $k_{R/M}$ and for $\rho=2$, with variable σ.

These data, obtained using unidirectional reinforced composite models, are applicable to multidirectional composites, particularly to orthogonal three-dimensional composites. This was established in theory,[7,22,23] verified on a three-dimensional model[25] reproducing the exact geometry of such a composite (see Fig. 6) and, finally, proved experimentally.[7,14,26] On this last point, experiments showed that the time variation of the apparent diffusivity of a three-dimensional carbon-carbon reinforced composite was quite analogous to the variations obtained with numerical simulations of one-dimensional reinforced composites. Figure 7 gives an example of experimental data for the variation of the apparent diffusivity of a three-dimensional carbon-carbon composite, measured in our laboratory.[26] Methods were developed for deducing from such curves the real diffusivity of the reinforcement a_R, that of the matrix a_M, and the thermal resistance R.[26,27]

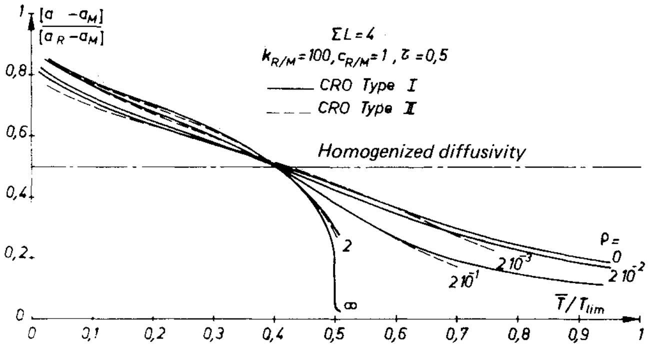

Fig. 4 Apparent diffusivity dependence on the specific contact thermal resistance ρ for undirectional reinforced composites characterized by $\tau=0.5$, $k_{R/M}=100$, $c_{R/M}=1$, and $\sigma=4$. —— layered directional reinforced composite; --- jointed chessboard directional reinforced composite.

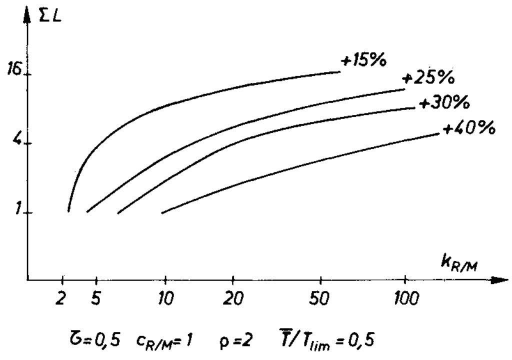

Fig. 5 Difference between the Parker diffusivity and the homogenized diffusivity as a function of ratio $k_{R/M}$ and of σ, for a unidirectional reinforced composite characterized by $\tau=0.5$, $c_{R/M}=1$.

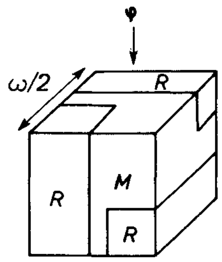

Fig. 6 Example of an orthogonal three-dimensional reinforced composite model.

In conclusion, the flash method measurement of the thermal diffusivity of a directional reinforced composite is difficult to interpret and results in the diffusivity of the equivalent homogeneous material only if certain precautions are taken, particularly as concerns the thickness of the specimen. Finally if homogenization is found faulty in the flash experiments, it may be faulty under conditions of actual use, e.g., when subjected to rapidly varying fluxes encountered during re-entry into Earth's atmosphere. This point is analyzed in the following section.

Effusion of Heat in Directional Reinforced Composites

The passage of heat through a medium, when it comes into contact with another medium, is subjected to the convection of a fluid, or receives radiations emitted from another medium, is characterized by effusivity b, or thermal inertia. Thus, the surface temperature increase of a semi-infinite medium subjected to a step heat flux can be written

$$T_w - T_0 = (2q\sqrt{t})/(b\sqrt{\pi}) \tag{2}$$

This property can be measured directly on an effusivimeter either of the Touchau type,[28-30] or others.[31] However, this measurement has not been made on directional reinforced composites until now. Thus, in contrast to diffusivity, analysis of the idea of the equivalent effusivity of a homogenized material applicable to directional reinforced composites has been approached only from the theoretical point of view.

Among the possible boundary conditions, the Fourier type is the most sensitive to the inhomogenity of a medium such as a directional reinforced composite. This is why the convective heating of such media has been covered by a numerical study.[6] The simulated directional reinforced composite is a layered type, as in Fig. 2, subjected to a convective step heat flux of the form

$$q_c = h(T_w - T_r) \tag{3}$$

where T_r is the recovery temperature of the gas. This study pointed out the following conclusions:

1) For convective heating, assimilating a directional reinforced composite to a homogeneous medium with effusivity $b_{\rm stat}=\sqrt{\bar{k}\bar{c}}$ is justified only for wall temperatures near the recovery temperature ($\bar{T}_w \simeq T_r$); that is, when the convective heat flux tends to cancel out and the phenomenon comes to resemble the steady state.

2) When the wall is cold, that is, when $T_w \ll T_r$, the overall behavior of the composite is closely approximated by an equivalent homogeneous medium having effusivity b_0 equal to

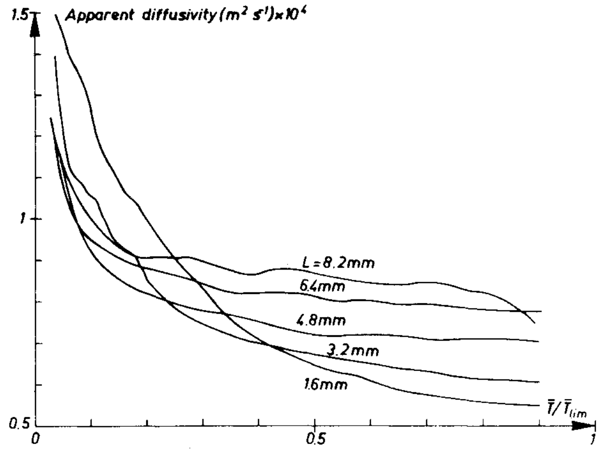

Fig. 7 Apparent flash diffusivity of an orthogonal three-directional carbon-carbon composite measured at ONERA.[26,27] In this case, $\sigma=L\Sigma=2,4,6,8$, and 10.2.

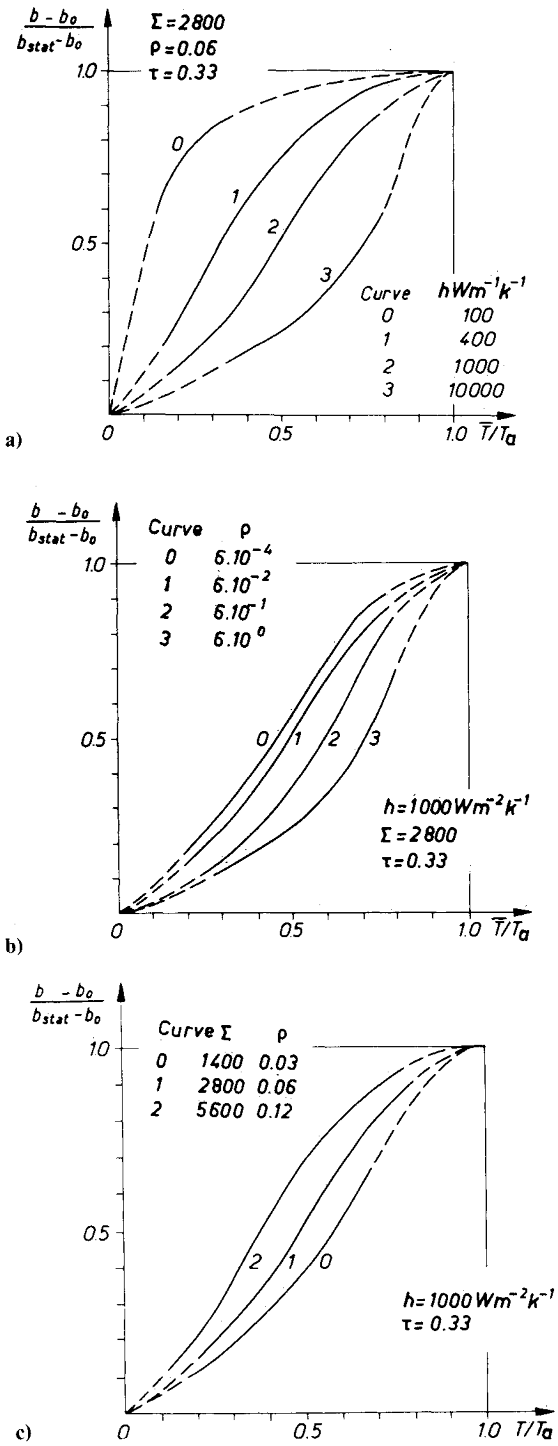

Fig. 8 Variation of the mean apparent effusivity of a directional reinforced composite characterized by $\tau = 0.33$, $k_{R/M} = 6.33$, and $c_{R/M} = 1.08$, subjected to a convective step heat flux. Influence of the transfer coefficient h (Fig. 8a), of ρ (Fig. 8b), and of Σ (Fig. 8c).

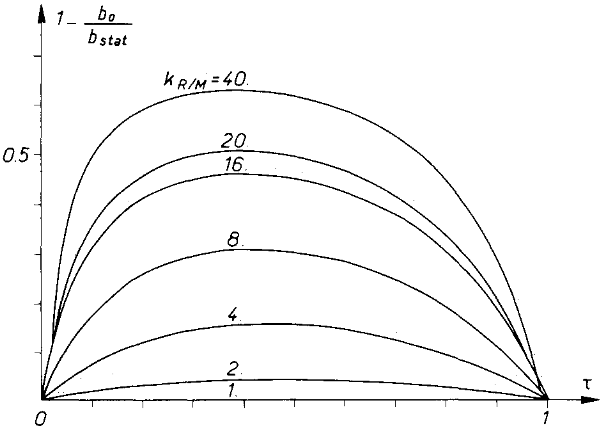

Fig. 9 Influence of τ and of $k_{R/M}$ on the apparent effusivity of a reinforced composite with ratio $c_{R/M} = 1$, subjected to a convective step heat flux.

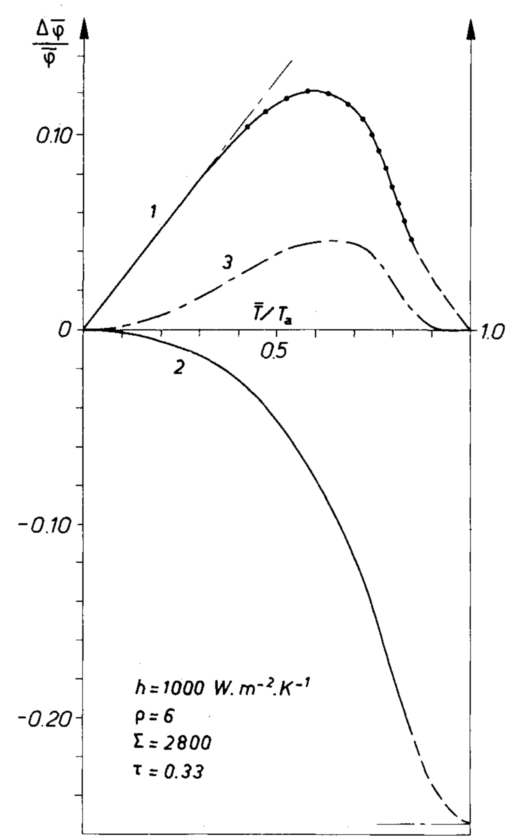

Fig. 10 Error appearing in the estimation of the net heat flux entering a directional reinforced composite subjected to a convective step heat flux. The computation conditions are: $h = 1000$ W m^{-2}K^{-1}, $\rho = 6$, $\Sigma = 2800$ m^{-1}, $\tau = 0.33$, $k_{R/M} = 6.33$, and $c_{R/M} = 1.08$. Curve 1: $b = b_{stat}$; curve 2: $b = b_0$; curve 3: $b = b_0 + (\bar{T}_w/T_r)(b_{stat} - b_0)$.

the harmonic means of the effusivities of the two components:

$$1/b_0 = (\tau/b_R) + [(1-\tau)/b_M] \tag{4}$$

3) Neither of the two homogeneous models is entirely satisfactory for wall temperatures between these two extreme cases. In fact, the whole directional reinforced composite, as far as its wall temperature and net input flux are concerned, behaves as a medium with an effusivity that varies as a function of wall temperature. This apparent effusivity is shown in Fig. 8 for a silicophenolic-type material with a reinforcement volume content of 0.33, characterized by the ratios $k_{R/M} = 6.3$ and $c_{R/M} = 1.08$. The parameters governing the variation, with $\bar{T}_w/T_r$, of the apparent effusivity are 1) the previously defined parameters τ, $k_{R/M}$, $c_{R/M}$, Σ, and ρ, and 2) the convective transfer coefficient h.

The effect of h, Σ, and ρ, is shown in Fig. 8; the effect of τ and $k_{R/M}$ is shown in Fig. 9. As for the diffusivity, the increase in the ratio of conductivities enlarges the phenomenon and the deviation with respect to the steady-state homogeneous model.

Figure 10 shows the errors that appear in the evaluation of the net flux entering the silicophenolic material when the two homogeneous models are used, defined by effusivities b_0 and b_{stat}. Curve 3 is an evaluation using an effusivity that varies linearly between b_0 and b_{stat}, with reduced temperature $\bar{T}_w/T_r$:

$$b = b_0 + (\bar{T}_w/T_r)(b_{stat} - b_0) \tag{5}$$

In conclusion, numerical simulation shows that the evaluation of convective heating, and particularly the total quantity of heat absorbed by the medium, can be marred with non-negligible errors when the homogeneous model is used with the properties of the steady-state regime.

Conclusions

The examples given herein show that the directional reinforced composites pose problems in determining their thermal properties and also in predicting their behavior under severe heating conditions. It would be presumptuous to expect quick, complete, and effective solutions to these two problems. What can be done, though, is to spell out some rules dictated by common sense.

As far as the methods of thermal characterization are concerned, experiments must be aware of the limiting conditions under which the idea of an equivalent homogeneous material become faulty, and they must conceive new methods that will enable them to determine firsthand the thermal properties of the components and of the interface. On this point, we do not mean to question the value of proven experimental techniques, but rather to conceive new interpretations that take into account the real structure of the composite. An example of adapting a conventional method to this new type of material is presented in Refs. 26 and 27. The flash method is reviewed in these references, without any modification of the experimental technique, but a new interpretative process makes it possible to start with the mean temperature on the rear face and to deduce the in situ diffusivity of the reinforcement parallel to the heat flux, that of the equivalent matrix (real matrix plus transverse reinforcements), as well as the value of the thermal contact resistance R. In Ref. 26, the method is applied to various three-dimensional carbon-carbon composites. The axial in situ diffusivity of Toray fiber yarns is determined in this manner. Very high values of diffusivity are found: 3.10^{-4} m^2s^{-1}, three times higher than the homogenized diffusivity of the composite. A somewhat different procedure is presented in Refs. 14 and 19, in which the diffusivity of the reinforcement is likewise approached by starting with the flash measurement. Both methods seem to lead to results that agree.

As to the modeling of the directional reinforced composites for the purpose of predicting their behavior under severe transient heating, here, too, the engineer should first determine the limits of applicability of the equivalent homogeneous model and, if this is not satisfactory, should then consider a more sophisticated model. The optimum complexity needed must then be determined. Going from the simple to the complex, we can consider:

1) The model of the equivalent homogeneous medium, the properties of which are deduced from the steady-state regime.

2) Equivalent homogeneous models specially adapted to a particular situation (refer to the example of the model with effusivity b_0 described above).

3) Models in which each phase is homogenized separately, with the interface characterized by a coupling term that appears in the equation for each component.[8,10,11]

4) Replacement of the multidirectional reinforced composite with a unidirectional reinforced composite, with reinforcement parallel to the main direction of the flux, the behavior of which is equivalent in the steady-state regime as well as in the transient regime.

5) A model taking the exact geometry of the multidirectional reinforced composite into account. The volume of the calculations to be made then becomes enormous, requiring the use of such methods as Schwartz overlapping subdomains,[32] which provide effective processing on parallel processors. Work is currently under way in this field in a cooperative effort between Laboratoire d'Informatique pour la Mécanique, et les Sciences de l'Inqénieur Centre National de la Recherche Scientifique and ONERA.

The Fourier-type boundary conditions are the most frequently encountered in practical applications. The difficulties that arise from modeling directional reinforced composites subjected to such boundary conditions were shown. In the case of convective heating, it would be useless to attempt anything on the internal response of the material without a correlative improvement in the modeling of the boundary layer. To do this, it seems necessary to take into account the large periodic variations in the wall temperature arising from the reinforcement/matrix alternation for airstreams in the boundary layer. No studies seem to have been carried out in this field yet.

In summary, three lines of action emerge: 1) adaptation of the thermal characterization methods to the directional reinforced composites, 2) improvement of the models along with better knowledge of the limits of their applicability, and 3) in the special but important case of convective heating, the inclusion of the heterogenity of the material in the treatment of the boundary layer.

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