Experimental study of radiative transfer in semi-transparent composite materials at different temperatures

F Retailleau\textsuperscript{1,2*}, V Allheily\textsuperscript{1}, L Merlat\textsuperscript{1}, J-F Henry\textsuperscript{2} and J Randrianalisoa\textsuperscript{2}

\textsuperscript{1}French-German Research Institute of Saint-Louis, 5 rue du Général Cassagnou, BP 70034, 68301 Saint-Louis CEDEX, France
\textsuperscript{2}Institut de Thermique, Mécánique, Matériaux (ITheMM), EA 7548, Université de Reims Champagne-Ardenne, Campus du Moulin de la Housse, F-51687, Reims, France

E-mail: florent.RETAILLEAU@isl.eu, vadim.ALLHEILY@isl.eu, lionel.MERLAT@isl.eu, jf.henry@univ-reims.fr, jaona.randrianalisoa@univ-reims.fr

Abstract. This study deals with the analysis of the propagation of radiation within a diffusing semi-transparent composite medium with rough boundaries. The two-phase medium (resin matrix and glass fibers reinforcement) is treated as an equivalent homogeneous medium characterized by volumetric radiative properties (extinction coefficient, albedo and phase function) and boundary scattering properties. The aim is to identify the radiative properties at different temperatures ranging from room temperature to 200\textdegree{}C. The identification method (Gauss-Newton) uses bidirectional reflectance and transmittance values. The experimental results are obtained using a spectrophotometer equipped with a goniometer and a heated sample holder. The Monte Carlo method is used to solve the Radiative Transfer Equation (RTE) in order to obtain the theoretical values.

1. Introduction

Composite materials are today essential in many fields of engineering thanks to their structural characteristics (lightness, solidity, rigidity) and the reduction of production costs. In several applications (aeronautics, defense, biomedical, etc.), it is necessary to know their behavior when subjected to a radiation flux. The objective is to understand the propagation of an incident radiation within an absorbing-scattering semi-transparent composite material with rough boundaries at different temperatures.

Many works have been reported on radiative characterization of semi-transparent media \cite{1, 2}. Particular attention has been paid to the study of materials with very high porosity such as foams \cite{2} and fibrous media \cite{3, 4} and materials with low porosity such as bubble glasses \cite{5} and ceramics synthetized by sintering powder \cite{1}. In most of these works, the boundaries of the samples are treated either transparent or smooth at the wavelengths of interest due to their high or low porosity nature. One of the difficulties in studying radiative transfer in composite materials concerns the presence of rough boundaries which may significantly affects the measurements.

To determine the radiative volumetric properties of complex materials, Baillis and Sacadura
[6] have suggested to use an identification method based on a RTE resolution method. The identification of radiative properties is carried out by using hemispherical reflectance and transmittance measurements. In order to identify the extinction coefficient, the albedo and the phase function of disperse materials, Nicolau et al. [7], Henry et al. [8] and Coray et al. [9] used bidirectional transmittance and reflectance measurements.

In a recent study, we have developed a method to simultaneously identify the volumetric radiative properties and boundary scattering properties of rough semi-transparent composite materials [10]. The identification method of Gauss-Newton is based on a combination of hemispherical reflectance and transmittance measurements and bidirectional reflectance and transmittance measurements obtained with a spectrophotometer equipped with a goniometer.

The present work deals with the determination of both volumetric and boundary radiative properties from room temperature to temperature of degradation (200°C). It enables us to follow the evolution of the radiative behavior of this type of material depending on the temperature.

2. Identification of radiative parameters
2.1. Principle of the method
The objective is to identify three volumetric parameters (the extinction coefficient $\beta_\lambda$, the albedo $\omega_\lambda$ and the asymmetry parameter $g_\lambda$ of the Henyey and Greenstein phase function) and three main surface scattering parameters described in the section 2.3, all independent from the direction [10]. The principle of the identification method is based on the minimization of the sum of the squared residuals [10]:

$$S = \sum_{i=1}^{36} (x_{t}(i) - x_{e}(i))^2 + (\tau_{t}^\cap - \tau_{e}^\cap)^2 + (\rho_{t}^\cap - \rho_{e}^\cap)^2$$

(1)

with $x_{t}(i)$ the theoretical value of the bidirectional reflectance or transmittance at the angle of measurement of index $i$ of the quadrature. $x_{e}(i)$ is the corresponding for experimental values.

The Gauss-Newton method is relevant for solving this type of non-linear problem. In addition, the greater the number of meaningful measurement data, the more the identification is accurate. That is why we employed here both hemispherical and bidirectional measurements.

2.2. Experimental measurements
The bidirectional reflectance/transmittance ($\rho^\cap/\tau^\cap$) describes the intensity of the radiation reflected/transmitted by the medium at coordinate $s = 0/s = e$ along a direction of angle $\theta$, in an unit solid angle of divergence $d\omega_0$ [5, 7, 10].

$$\rho^\cap(\theta), \tau^\cap(\theta) = \frac{M(\theta)}{\cos(\theta)\max(d\omega_{f}d\omega_{0})}$$

(2)

$M$ is the ratio of the transmitted (or reflected) signal at the angle of measurement $\theta$ to the measured signal without the sample. $d\omega_{f}$ refer to the solid angle of detection. To obtain an accurate measurement of $M(\theta)$ and $\theta$, a spectrophotometer equipped with a goniometer system (Cary Universal Measurement Accessory of Agilent) is used [10]. The temperature is controlled by a home-made heated sample holder that can reach 300°C. The heating is done by Joule effect and enables a homogeneous heating to ±3.5% over a 20 mm diameter surface. The solid angles $d\omega_0$ and $d\omega_d$ are controlled with diaphragms to determine precisely the bidirectional data. The hemispherical data are obtained by summing the bidirectional data at each solid angle of discretization $\Delta\omega$ over the appropriate hemisphere.

$$\rho^\cap = 2\pi \sum_{i=1}^{18} \rho^\cap(\theta_i)\cos(\theta_i)\Delta\omega_i ; \tau^\cap = 2\pi \sum_{i=1}^{18} \tau^\cap(\theta_i)\cos(\theta_i)\Delta\omega_i$$

(3)
2.3. Theoretical transmittances and reflectances

The theoretical values are obtained by solving the RTE. In this study, the semi-transparent medium is considered to be cold, since up to 200°C, the sample emission domain is outside the spectral range of characterization of the spectrometer (from 0.4 to 2 μm). The RTE for cold samples is written as follows [11]:

\[
\frac{\partial I_\lambda(s, \theta, \varphi)}{\partial s} = -\left(\kappa_\lambda T + \sigma_\lambda T\right) I_\lambda(s, \theta, \varphi) + \frac{\sigma_\lambda}{4\pi} \int_0^{4\pi} \Phi_\lambda(\theta, \varphi, \theta', \varphi') I_\lambda(s, \theta', \varphi') d\omega' \tag{4}
\]

with \(I_\lambda(s, \theta, \varphi)\) the radiation intensity at the abscissa \(s\) along the direction characterized by the polar angle \(\theta\) and the azimuthal angle \(\varphi\), \(\kappa_\lambda\) the absorption coefficient, \(\sigma_\lambda\) the scattering coefficient and \(\phi_\lambda\) the phase function defined here by the Henyey and Greenstein approximation [11]. The boundary conditions are as follows with \(\mu = \cos\theta\):

\[
\begin{align*}
\mu > 0 & \quad I(0, \mu) = (1 - r_{12}(\mu_0, \mu))I(0, \mu_0) + 2\pi \int_0^1 r_{21}(\mu', \mu) I(0, -\mu') \mu' d\mu' \tag{5} \\
\mu < 0 & \quad I(e, \mu) = 2\pi \int_0^1 r_{21}(\mu', \mu) I(e, \mu') \mu' d\mu' \tag{6}
\end{align*}
\]

For rough boundaries, the Fresnel equations are not sufficient to determine the reflectivity involved in Eqs. (5) and (6). One of the solutions employed in the literature is to add a surface scattering factor in the reflectivity and transmittivity calculations of surfaces [10, 12]. This correction factor assesses the size of the roughness of the boundary relative to the wavelength of the radiation beam. The reflectivity of an interface between medium \(i\) and medium \(j\), \(r_{ij}(\mu', \mu)\) can be determined as follows:

\[
r_{ij}(\mu', \mu) = R_{ij}(\mu') P(\mu) C_{fij} \tag{7}
\]

\(R_{ij}(\mu')\) is the reflectivity for a smooth interface calculated with Fresnel equations. \(C_{fij}\) is a correction factor and \(P(\mu)\) is a probability distribution function according to \(\mu\) giving the angular distribution of the scattered beam, both are determined by identification from experimental values. This probability function can ether be a cosine function or a Gaussian function [10, 13].

The collision based Monte Carlo Method (MCM) is well appropriate to model radiative transfer problems, as shown by Fleck [14]. It enables to follow the propagation of photons through a semi-transparent medium. It is not necessary to make approximation of the radiative phenomena which makes the method numerically straightforward.

3. Results

Figure 1 shows the experimental lobes of bidirectional transmittance and reflectance at 1070 nm for a 3 mm thick glass/epoxy sample at different temperatures and Fig. 2 shows the identified radiative properties. The lower left inset is a zoom on the reflection lobe and the black arrow represents the incident radiative flux. The measurements reveal that the radiative behavior of this type of composite is clearly temperature-dependant even before it undergoes thermal degradation. The extinction coefficient \(\beta\) and the scattering albedo \(\omega\) seem much more dependent on the temperature and especially around the temperature of the glass transition, previously measured by DSC around 105°C, while the absorption coefficient exhibits a small temperature dependence. This demonstrates that scattering by fiberglass attenuates around the glass transition temperature due to closeness of the resin and glass refractive indexes in this temperature range [15]. Above 150 °C and as result of the opacification, the absorption coefficient of the resin rises with temperature and explains the decrease of the scattering albedo \(g\). The abrupt decrease of the asymmetry factor at high temperature points out an unexpected change of the scattering distribution, where backward scattering tends to be enhanced.
Figure 1: BRDF/BTDF of a 3 mm glass/epoxy sample at 1070 nm at different temperatures.

Figure 2: Radiative properties of a 3 mm glass/epoxy samples at 1070 nm as a function of temperature.

4. Conclusion
The Monte Carlo method is used to solve the RTE in which the phase function is approximated by the Henyey & Greenstein approximation. Boundaries are assumed as semi-transparent and scattering. The direct model consists of six unknowns: three volumetric properties (absorption coefficient, albedo and asymmetry factor) and three surface scattering properties. A spectrometric device equipped with a goniometer and a heated sample holder enable to measure bidirectional transmittances and reflectances on 36 scattering directions distributed uniformly around the sample at different temperatures. An identification model using the Gauss-Newton method enable to identify the volumetric radiative properties of semi-transparent and rough composite materials. This method enable to follow the evolution of the volumetric properties of a semi-transparent material as a function of its temperature, and possibly identify changes in state as the glass transition.

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