INFLUENCE OF THE FORMING CONDITIONS ON THE RADIATIVE PROPERTIES OF SEMI-CRYSTALLINE POLYMERS

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ABSTRACT. Our objective is to understand the influence of the elaboration conditions of semi-crystalline polymers on their radiative behavior. To reach this goal, we have performed measurements of infrared reflection and transmission, diffuse and specular, on slabs of polypropylene prepared by injection process in our laboratory. Then, we use an inversion method based on the Adding-Doubling concept to determine the volumetric absorption and scattering coefficients as well as the anisotropy factor (mean cosine of the scattering angle) of the compound materials. This work will help provide a data bank for industrial applications concerned by manufacturing of advanced semi-crystalline reinforced systems.

NOMENCLATURE

Exiting angle	ν'	Quadrature points	x_k	
Incident angle	ν	Quadrature weights	H_k	
Normal hemispherical reflectance	R_H	Temperature	T	K
Normal hemispherical transmittance	T_H	Wavenumber	σ	m^{-1}

INTRODUCTION

Polymer composite materials are nowadays widely used in structures where low weight in combination with high strength and stiffness are required. In polymer composites, the individual constituents are polymer matrix and reinforcement [1]. Thermoplastic composites (TPCs) have a number of advantages over traditional thermosetting composites in many fields and becoming the material of choice for replacing traditional materials such as steel and/or aluminum, among others, especially in automotive aerospace, marine and electronic applications. TPCs also offer clear advantages over their thermoset counterparts in terms of improved fracture toughness, potential for repeated recycling and, most notably, the possibility to reshape or remold the product at elevated temperatures. But, nowadays, empiricism is still often the sole way to improve these processes which imply long times for the adjustment of the optimal production rates, the risks of manufacturing of poor quality components (defects, mechanical deficiencies) and/or completely inappropriate use of energy. This is particularly true for industrial processes based on infrared heating (pultrusion, thermoforming,..).
For these cases, the knowledge of the thermal radiative properties of materials [2] is a crucial interest for the design of efficient systems where energy is mainly transported by thermal radiation An accurate control of these properties would allow the optimization of the use of these complex semi-transparent materials in advanced industrial processing related to thermal radiation. So the main objective of our work is to develop innovative experimental and numerical methodologies to increase significantly the understanding of the radiative properties of semi-crystalline polymers. The semi-crystalline polymers are characterized by a particular microscopic texturing. In this work, the term texture stands for the spatial arrangement of the heterogeneities with the host solid matrix

as their respective size, orientation and shape distributions. To achieve the goals we will focus on sample of polypropylene designed in our laboratory by an injection process. We can pilot all the parameters of processing, like the temperature of mold, which will generate samples with parallelepipedic shape (60mm x 60mm x 3mm). Then we characterize by infrared spectroscopy the diffuse and specular transmission and reflection for different samples developed at different elaboration temperatures 40, 80 et 120°C. Finally we present the inverse method called "Adding-Doubling" historically developed by HC Van de Hulst and the first volume radiative properties obtained.

INVERSE ADDING-DOUBLING (IAD)

Inverse Adding-Doubling is a technique developed by Scott Prahl [3] that uses adding-doubling to figure out the optical properties of scattering and absorbing slabs from the observed diffuse reflectance and transmittance. These properties are obtained by repeatedly solving the radiative transport equation until the solution matches the experimental reflection and transmission values. The doubling method was introduced by Van de Hulst for solving the radiative transport equation in a slab geometry [4]. The doubling method assumes that the both the bi-directional reflectance $R(\nu,\nu')$ and transmittance$T(\nu,\nu')$ for light incident at an angle ν and exiting at an angle ν' is known for one layer. The reflection and transmission of a slab that is twice as thick is found by juxtaposing two identical slabs and adding the reflection and transmission contributions from each slab [5]. The adding method extends the doubling method to dissimilar slabs. Thus slabs with different optical properties can be placed adjacent to one another to simulate layered media or internal reflection caused by index of refraction differences.
The adding-doubling method is based on the numerical integration of scattering phase functions with quadrature:

$$\int_0^1 f(\nu,\nu')d\nu' = \sum_{k=1}^{N} H_k\, f(x_k) \qquad (1)$$

The quadrature points x_k and weights H_k are chosen so that the integral is approximated exactly for a polynomial of order $2N-1$ or possibly$2N-2$, depending on the quadrature points (Gaussian quadrature) is equivalent to the spherical harmonic method of order P_{N-1}.

RESULTS AND DISCUSSION

Small pieces of samples were cut from the central part of the polypropylene slabs in order to avoid bolder effect due to the mold cooling. The spectrometer vertex 80V (Bruker Optics) measures direct (normal normal) transmission of light through a sample as a function of wave number. When equipped with an integrating sphere system, the spectrometer can also measure both the diffuse reflection and transmission (normal hemispherical) of a sample. First, we have made spectral measurements (figure 1 and 2) on a sample of polypropylene manufactured at LTN for a mold temperature of 40°C and with a thickness of 3mm. The experimental variables and numerically calculated by "Adding-Doubling" are virtually identical. To obtain these results we have achieved a quadrature type S8 and we have used 20 fictitious films with a thickness of 150 microns.
The inversion method helps us to get the diffusion and absorption coefficients as well as the anisotropy parameter. Figure 3 shows that the material is weakly absorbing in the range of 4000cm^{-1} to 20000cm^{-1}. As in Figure 2, there are three absorption bands for wavenumbers σ = 5900 cm^{-1}, σ = 7100 cm^{-1} and σ = 8200 cm^{-1}. It is also clear from Figure 3 that the scattering coefficient increases with the wave number in the near infrared spectral range and visible. The average anisotropy factor is 0.8 which shows the highly scattering behavior for this spectral range. This can be typical of Mie scattering.

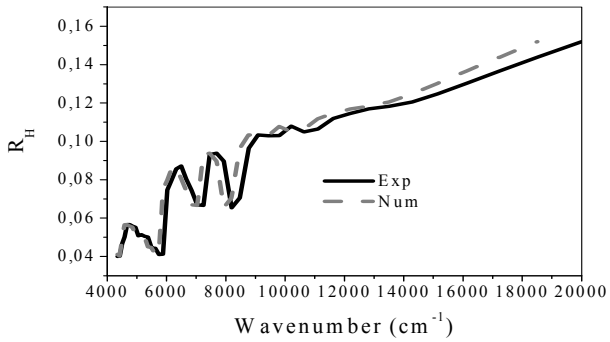

Figure 1 Normal Hemispherical reflectance as a function of the wave number (cm^{-1}) at T=300 K

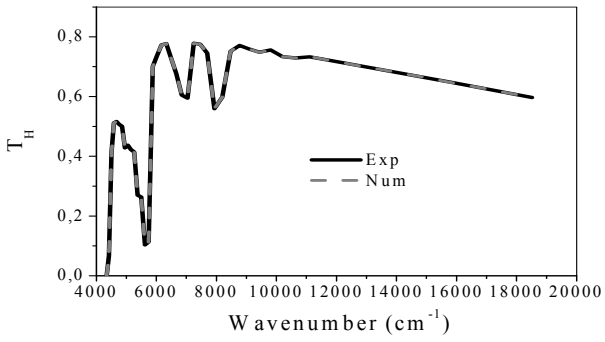

Figure 2 Normal Hemispherical Transmittance as a function of the wave number (cm^{-1}) at T=300 K

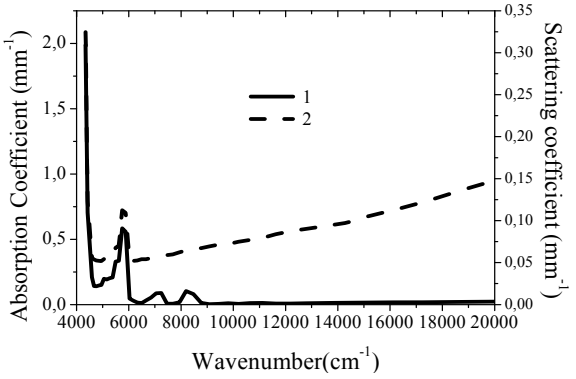

Figure 3 Absorption coefficients (1) and distribution (2) as a function of the wave number (cm^{-1}) at T = 300 K. These coefficients were obtained by the IAD method

CONCLUSION

In this work we have prepared samples of polypropylene by controlling the conditions of forming. Also, we analyze the radiative properties by inverting experimental spectra (transmittance, reflectance) by a method based on the concept of Adding-Doubling. The anisotropy factor for a sample prepared at 40°C is 0.8 which shows the highly diffusive in the Near InfraRed spectral range and visible spectral range. The absorption coefficient is low here and the increase of scattering coefficient with wave number suggests that scattering properties can be explained by the Mie scattering theory. In addition, to better interpret our data, we need to know well the crystallinity of the samples by using the Differential Scanning Calorimetry (DSC). Also, other quadrature schemes under test should be considered for the IAD method. In the poster, more results for an elaboration temperature at 80 and 120°C will be shown.

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