RADIATIVE TRANSFER THROUGH A PMMA SAMPLE. PART 1 : SPECTROSCOPIC STUDY AND OPTICAL INDICES

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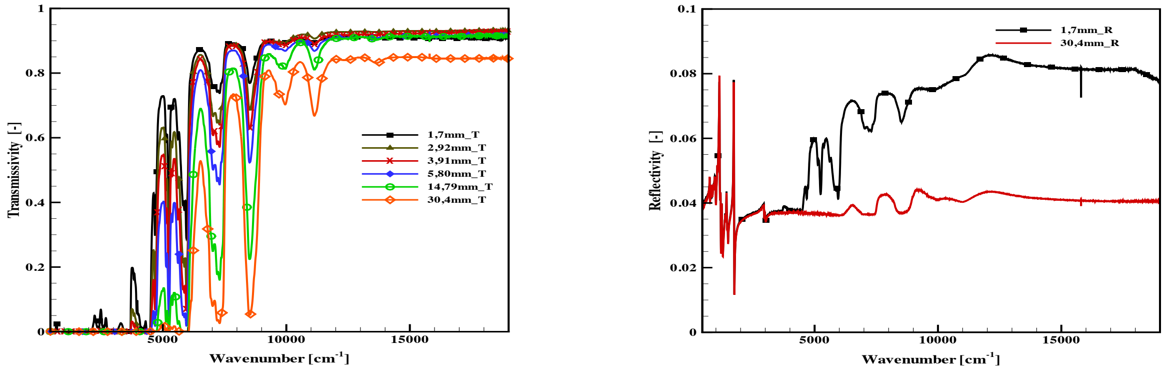
ABSTRACT. This work has been carried out in order to determine the optical properties of a poly-methyl-methacrylate (PMMA) sample, which is known to be a highly non grey participating medium. Spectral transmissivity and reflectivity have been measured in a wide range from visible to infrared. The optical indices have been determined combining several methods suitable for ranges where either reflectivity or transmissivity can be measured with sufficiently high accuracy. The particular behaviour of PMMA is discussed, before application to samples submitted to high incident fluxes, which is the topic of a second part of the present study.

INTRODUCTION

Poly-methyl-methacrylate (PMMA) is a material involved in numerous academic studies aimed at characterizing the thermal degradation of solids (see [1, 2] among others). Such studies are usually undertaken by submitting the sample to high radiative fluxes provided by external sources. PMMA is a non-charring material, often involved in experimentations, since it is easily ignited and leads to tests with a very long stationary step, with very good repeatability. However, it presents a highly non-grey behaviour which is not always accounted for. This behavior might cause strong discrepancies in the analysis of researchers depending on the way they are producing the radiative flux incident on the slab. The most often-used devices are [3]: (i) the Fire Propagation Apparatus (FPA) which produces a strong incident irradiation in the visible and the near infrared thanks to tungsten lamps, or (ii) the cone calorimeter which almost behaves like a blackbody with typical temperatures ranging between 700 and 1000 K depending on the expected flux, but always involving larger wavelengths than the FPA. For a better understanding of the influence of the source type on the sample heating, a complete characterization of the optical and radiative properties of the PMMA is required. The corresponding study is presented here with the data for the spectral reflectivity and transmissivity, followed by an analysis providing the optical indices and a verification of the data relevance (Part 1); then, the radiative behaviour of a PMMA sample submitted to a high incident flux and the related thermal degradation is simulated, using a Monte Carlo Method for the radiative part of the problem (Part 2).

REFLECTIVITY AND TRANSMISSIVITY MEASUREMENTS

Spectroscopic measurements have been carried out using a Vertex 80 FTIR spectrometer by BRUKER. Seven samples with thickness equal to 1.7 mm, 2.9 mm, 3.9 mm, 5.8 mm, 14.8 mm, 30.4 mm and 100 mm have been studied (all measurements are not reproduced here). Typical results are plotted in Figure 1 as a function of the wavenumber. The non grey behaviour is obvious with high variations in the transmissivity in particular, the sample being almost transparent (transmissivity near 90%) in the high wavenumbers, while being opaque in the mid infrared. The sample thickness naturally strongly affects the transmission signal. The reflectivity variation is limited to the range between 2% and 8%, also with sharp spectral variations. Two different thicknesses are presented for the reflectivity, indicating the influence of multiple reflections inside the slab (highest reflection for the thinnest sample because of further order reflection captured by the detector), especially in the range where the transparency is the highest of course. Such high non grey behavior obviously explains why discrepancies have been observed in the sample degradation depending on the irradiation source.



(a) Transmissivity (b) Reflectivity

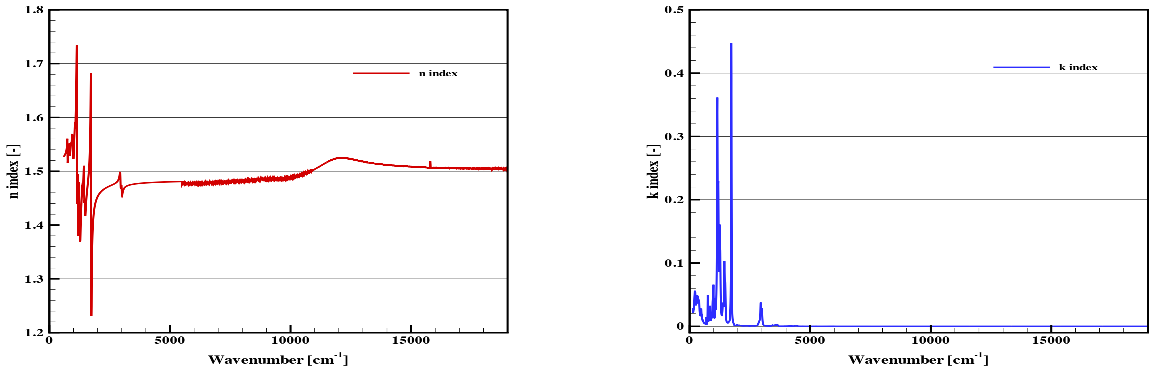
Figure 1: Transmissivity and reflectivity of various PMMA slabs

IDENTIFICATION OF THE OPTICAL INDICES

The radiation path is easily modeled in the present slab, even accounting for multiple reflection [4]. Straightforward identification of the indices can be sought on this basis, or more usual methods (Kramers-Kronig, Lorentz oscillators) can be applied. All these methods have been used and combined here. One difficulty arises from the strongly varying transmission and reflection data which may provide high uncertainty on the index values in given ranges, when transmission or reflection levels fall below a correct sensitivity threshold. Therefore a combination of several methods has been considered in complementary ranges with sufficient overlappings in order to ensure a continuous determination of the indices and a check of the identified values with the different methods whenever possible. A model based on Lorentz oscillators has been used especially in the ranges where transmissivity is too small to ensure a good accuracy when identifying the indices in a straightforward manner, while reflectivity is measured with a good sensitivity. An optimization algorithm has been used (based on genetic algorithm formulation) which provides the oscillator parameters in definite bands and the optical indices in a second step (results have been compared with a usual Kramers-Kronig method in the corresponding ranges, showing a quite perfect agreement for the identified indices). For the other spectral ranges an optimization algorithm has been used, based on a complete model for the slab transmission and a least mean squares method, making use of the data gathered for all the thicknesses. The indices are plotted in Figure 2 as a function of the wavenumber. The refraction index presents sharp variations, with average value close to 1.5 (which is in agreement with the few available data [5]). The absorption presents very high contrasts with near-zero values in some ranges and maximum values larger than 0.4 in definite bands (which also explains the strong variations on the observed spectroscopic data).

INDEX VERIFICATION AND RAY TRACING APPROACH VALIDATION

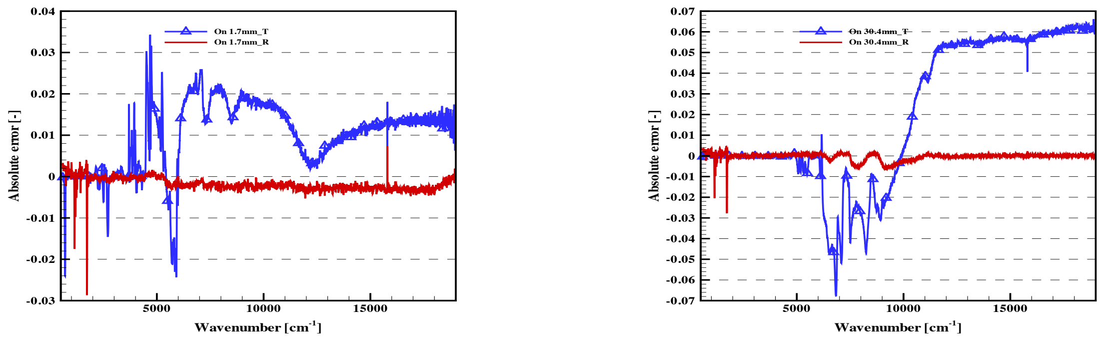
A verification of the acccuracy of the identified indices has been carried out based on an analysis of the radiation path inside the slab and accounting for multiple reflections. A ray tracing approach has been used to simulate the reflectivity and the transmissivity of PMMA samples with thickness 30.4 m and 1.7 mm. For the thickest sample, multiple reflections are not included in the simulation as the rays exit the sample on the irradiated face too far from the



(a) n index (b) k index

Figure 2: n and k indices

area observed by the detector, while first order reflections are accounted for in the simulation of the 1.7 mm sample (actually, larger order reflections are not captured by the detector). Figure 3 shows the discrepancies obtained on transmissivity and reflectivity. The agreement is satisfactory for the present preliminary test, with average discrepancies computed between 1.6 and 6.0% depending on the observed curve. The obtained indices and the ray tracing code will be now involved in the simulation of a sample degradation test (see Part 2).



(a) Error, sample 1.7 mm thick (b) Error, sample 30.4 mm thick

Figure 3: Error on the simulated reflectivity and transmissivity

CONCLUSION

Various PMMA samples have been studied, measuring their reflectivity and transmissivity. Measurements confirm the high non-grey behavior of PMMA and explain the dispersion of the degradation results observed by various groups depending on the source used for the sample heating. These data will be used in a companion study aimed at modeling the combined heat transfer inside the sample during thermal degradation.

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