Determination of the elastic parameters of thermoplastic composites using the indentation technique

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Abstract—this work deals with the application of the macro-indentation technique to the determination of the elastic parameters of composite with polypropylene matrix and reinforced with ductile and brittle fibers. The aim is to obtain a representatives average values taking into account the influence of the holding time and type of the used reinforcement (ductile or brittle reinforcement) on the overall elastic modulus of the studied polymers. To do this, the diameter of the indenter used is chosen relatively large (diameter of 16 mm) and the load equal to 6 kN. The various results obtained made it possible to see the effects of the holding time and the type of reinforcements used on the evolution of the elastic modulus of the reinforced composite.

Keywords—thermoplastic composite; macro-indentation test; viscoplastic behavior

1. INTRODUCTION

Polypropylene is one of the most widely used polymers in a number of industrial sectors, including packaging, automotive, household appliances, sanitary ware and textiles. This material is easy to implement by various techniques such as compression molding, extrusion and injection, among others. We can then make objects in different forms and it is possible to recycle. Finally, the quality/price ratio of this material is indisputable. Its mechanical properties are relatively medium, especially as it can easily be modified, increasing their property by mixing the polymer with different reinforcements (glass fibres, steel wires), or by adding fillers in particular. Composites are heterogeneous materials that group virtually all materials [1]. They consist of several phases and different compositions. The exact determination of these characteristics is no longer possible, it is therefore necessary to solve the system to maintain only average behaviour, depending on the elementary properties (fibres, matrix) and their geometry.

The classical experiments make it possible to take into account the elastic deformations and the data processing techniques is very heavy with the overall response of the material (stress-strain curves). In the case of low viscosity materials such as fibre reinforced polymers, the mechanical behaviour is composed of elastic components, viscoelastic, plastic and viscoplastic. In the case of indentation tests, the test protocol is composed of three stages, a loading period which corresponds to the first contact of the indenter with the material, a hold time is applied to the maximum load, a period unloading which allows to determine the Young's modulus (dP/dh). The holding charge is necessary to discriminate the reversible and irreversible time-dependent response, i.e., viscoelasticity and viscoplasticity. The macro-indentation test makes it possible to consider a reduced volume of the material, on which we define a heterogeneous behaviour of macro-metric dimension. It gives access to stress and strain fields on a level scale. It is also possible to take measurements at the interface [2, 3]. The mechanical properties are determined from the charge-discharge curve using the Oliver and Pharr model [4-6].

This work explores the potential of instrumented indentation at macroscopic scale to measure the mechanical properties of polypropylene matrix reinforced with the glass-fibre (brittle reinforcement) and steel wires (ductile reinforcement). The sensitivity of the elastic modulus, hardness and creep response of the studied materials is investigated. The applicability of the Oliver-Pharr method [4] is also discussed. The results of this study provide important insights into the viscoplastic behaviour of composites with brittle and ductile reinforcement.

2. INDENTATION TEST

The macro-indentation tests were performed on two glass/polypropylene and steel/polypropylene composites. Local measurements were conducted on an MLS traction machine. This allows the application of a force from 0.5 kN to 50 kN with a depth resolution of 0.001 mm. A spherical steel tool indenter (Z200) is chosen because of its versatility of use and because it is less severe in terms of stress concentration. The tests were carried out at a maximum controlled force $P_m = 6$ kN. This effort is sufficient to obtain a local deformation with dimensions about a few millimeters. The type of performed test consists of two phases which are the rise and descent in strength.

In the area of indentation, the composite will deform in a manner that reflects the mechanical properties of this indented material. The elastic modulus and the hardness can be calculated according to the maximum values of the load and...
depth. The Olivier and Pharr [2] method are used; it allows the description of the upper part of the discharge curve by the law:

\[ P = P_m \left( \frac{h - h_p}{h_m - h_p} \right)^m \]

Where \( P_m \) is the maximum applied load, \( h \) is the spherical indentation depth, \( h_p \) is the residual indentation depth (after full discharge), \( h_m \) is the maximum indentation depth at maximum load \( P_m \) and \( m \) is a constant determined by the fitting of the discharge curve.

The contact stiffness \( S \) is given by the tangent of the unload curve at the point of maximum load.

\[ S = \left( \frac{dF}{dh} \right)_{P=P_m} = m P_m (h_m - h_p)^{-1} \]

The extension of this tangent line gives the depth \( h \) and the contact depth \( h_c \) is then determined by \( h_c = h_m \). The reduced module \( E_r \) is given by:

\[ E_r = \frac{\sqrt{\pi} S}{2 p A_p(h_c)} \]

Where \( A_p \) is the surface of the projected contact at depth \( h_c \). To calculate the Young modulus \( E_{IT} \) of the tested material, it is sufficient to know its Poisson's ratio and the mechanical properties of the indenter. The Young's modulus is given by the following relation:

\[ \frac{1}{E_r} = \frac{1 - \nu_i^2}{E_{iT}} + \frac{1 - \nu_r^2}{E_i} \]

The elastic modulus of the indenter is \( E_i = 210 \) GPa and the Poisson's ratio of the indenter is \( \nu_i = 0.3 \). \( \nu_r \) is the Poisson's ratio of the tested material. The hardness \( H_{IT} \) is identified from the maximum load \( P_m \) and the projected contact surface \( A_p \), which is determined by calibration of the tip of the indenter.

\[ H_{IT} = \frac{P_m}{A_p(h_c)} \]

3. MICROSTRUCTURAL CHARACTERIZATION

Differential scanning calorimetry (DSC) measurements were performed on polypropylene samples to determine their specific enthalpies and melting temperatures and degree of crystallization. The weight of each sample is 7±0.5 mg. The thermal experiments were performed by heating the samples from 25 to 250°C at three different heating rates 1°C/min, 10°C/min and 30°C/min. The value of the specific fusion enthalpy \( \Delta H_f \) was determined from the area under the peak of the melting curves and the reference enthalpy \( \Delta H_f^0 \) at 100% crystalline (\( \Delta H_f^0 = 148 \) J/g for the polypropylene). The degree of crystallinity \( \eta \), is given by the following equation:

\[ \eta = \frac{\Delta H_f}{\Delta H_f^0} \times 100 \% \]

4. RESULTS

A. Local mechanical behavior

The results of the indentation test on the polypropylene/glass and polypropylene/wire-steel composite with the maximum load of 6 kN are shown in Figures 2 and 3.
Curves show nonlinear behavior up to the maximum load. During the discharge, a springback of 1.05 mm is recorded which corresponds to 23% of the total indenter displacement. The depression of the spherical indenter generates the growth of a bead which increases the effective contact area between the indenter and the material and leads to a high hardness value. However, for depths greater than 1 mm, it is found that the Young's modulus stabilizes at a value of 2.35 GPa and the hardness is stabilized at a value of about 15 GPa. However, the presence of the bead in the case of the polypropylene matrix alone leads to an underestimation of the Young's modulus (approximately 2.30 GPa) which is a high value with respect to the theoretical value of the Young's modulus (1.80 GPa). In fact, previous studies have shown that the relationship of Oliver and Pharr, which gives the contact depth $h_c$, remains relevant in the case of materials which have a high work hardening capacity such as the polypropylene matrix. In the fiber-reinforced matrix, the hardening around the impression restricts the movement of material upwards, and the plastic zone will develop rather in depth than on the surface [7]. Mean values of Young's modulus are comparable with those found at the nano-scale. In general, the properties present continuity between the different scales of measurement, since the module is an intrinsic property of the material, thus independent of the force. However, the results show a small difference (7%) between the nano-indentation tests and the macro-indentation tests. This difference is probably related to experimental errors during the tests, may be related to the relationships used for contact depth and contact area.

### Microstructural characterization

Figure 4 shows the thermograms of the samples with different heat treatments and reveals three different degrees of crystallinity (36.12%, 38.48% and 46.74%) and three glass transition temperatures around 40 °C. By comparing the thermograms of the three treatments, the sample cooled from the melt at 2 °C/min exhibits a larger peak of crystallization and a low melting peak. For slower cooling rates, the polymer stays longer in a temperature range where the temperature is low enough for the polymer to begin to solidify, and high enough to provide the energy needed to organize the polymer chains in the polymer. Space and create crystalline networks.

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The results of the thermal programs are shown in table 1.

<table>
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<th>Treatment</th>
<th>$\nu$</th>
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<td>53.4</td>
<td>36.1</td>
</tr>
</tbody>
</table>

### Influence of the holding time at Pmax

The results presented here permit to understand the dependence of the behavior of the polypropylene/wire-steel and polypropylene/glass composites under the macro-indentation with respect to the holding time (creep) (figs. 5 and 6). As shown in the figures, the polypropylene/wire-steel composite has a large capacity to deform under constant load (creep) that the polypropylene/glass composite.

![Fig. 5. Variation of the depth and the indentation load depending on the hold time at maximum load, obtained on polypropylene/wire-steel composite.](image5.png)

![Fig. 6. Variation of the depth and the indentation load depending on the hold time at maximum load, obtained on polypropylene/wire-steel composite.](image6.png)
The calculated reduced modulus contains both the elastic response and a low viscosity effect.

5. CONCLUSIONS

The study of indentation at different scales of measurement may give a better understanding of the phenomena that occur during the indentation test. Macro-indentation tests on composites were carried out and continuity between the different scales of the Young modulus and hardness was observed and these values are perfectly compatible with those from the literature. The elastic parameters calculated from the indent stress-strain behavior of these materials were conclusive for the polypropylene matrix and few inconclusive for the polypropylene/glass and polypropylene/wire-steel composites. However, it has been observed, by examining the flow of matter in the vicinity of the composites contact, that the anisotropic elastic properties are influenced by the presence of the viscosity. The lateral deformations observed on the polypropylene/wire-steel specimens are important, this being explained by the presence of plastic deformation of the steel wires and the matrix. Unlike the polypropylene/glass specimen, a weak lateral deformation is observed and once the deformation exceeds certain limit, the rupture is done brutally.

Références