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MECHANICAL CHARACTERIZATION AND SIMULATION UNDER MULTIAXIAL CONDITIONS OF POLYHYDROXYALKANOATES

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Abstract – This study focuses on the mechanical properties characterization of biodegradable and bio-based materials: the Polyhydroxyalkanoates (PHAs). An original experimental set-up has been developed to perform multiaxial tests on a single shape of cylindrical specimen. It is composed of a load cell and a mechanical assembly allowing successive or simultaneous loading cycles of tensile, compression and torsion. Classical shear and tensile tests, besides the fact they use different specimen shapes, seems not sufficient to get a suitable 3D constitutive model. The aim is so to perform complex loading paths with direction changing and cycles during a test, allowing to scan all the deviatoric plan of deformation and taking into account the loading history. This database is used for the identification of an Hyperelasto-Visco-Hysteresis (HVH) model which is used to simulate the mechanical behavior of food packaging structure in PHAs.

Keywords: Multiaxial mechanical properties, biopolymers, Polyhydroxyalkanoates, Finite Element Analysis, Food packaging

Introduction

Nowadays, plastic materials are everywhere. Used in major activities as electronic, automotive, building and food packaging industries, the demand of this kind of material is constantly growing. Largely petroleum based, “classical” plastics are no longer a future considering the petroleum resource depletion and their environmental impact. Bio-based and biodegradable polymers are a good answer to these issues and have been consequently much more investigated since the last decade.

Polyhydroxyalkanoates (PHAs), a polyester family which fit on this category, can be easily found in many different bacteria (*Ralstonia*, *Pseudomonas*, etc.) as intracellular reserves of energy [1]. PHAs have also the advantage to be compostable under specific conditions of temperature and humidity. This material has been chosen in this study because it shows an analogous mechanical behavior with synthetic polymers, as polypropylenes, and great similarities in terms of thermal and barrier properties [2]. These properties allow PHAs to be used in food packaging industry.

Nevertheless, their elaboration remains hard to control bringing to a high production cost and impedes a greater using of them. To make this kind of polymer more attractive for food packaging industry, it is necessary to optimize their mechanical properties and their shapes as finished products. An investigation of these properties, still not very well-known, has to be done to allow Finite Element Analysis (FEA) of industrial parts.

In this work, we present an original specimen and experimental device allowing to study the 3D mechanical behavior of a grade of PHA. Firstly, a presentation of the experimental setup and explanations of its purpose are done. Then, the results of tests with multiaxial loading conditions are given. Finally, this database is used to simulate food packaging structure by FEA.

Experimental methodology

The PHA used is a Mirel F3002 supplied by Telles (USA). The mechanical behavior as well as the thermal and barrier properties of this material make it a possible substitute in applications using Polypropylene materials [2]. Consequently, a Polypropylene (PPC7712 supplied by Total Petrochemicals, FR) is used to be compared with the F3002 results.

The specimens were molded using an injection-molding machine (Battenfeld) on a specially design mould. The injection for the F3002 and the PPC7712 has been carried out with a pressure of 75 MPa and 125 MPa and a temperature of 180°C and 240°C, respectively at the injection nozzle. An annealing process at 120°C was performed on all samples in order to achieve the best crystallinity ratio.

An original experimental setup (Fig. 1) has been especially designed to the characterization of the 3D mechanical behavior of the F3002. The main goal of this device is to perform multiaxial loading tests to have a better comprehension of the studied material behavior than we could have with uniaxial tests. The opportunity to have tests with complex trajectories (tension, compression and torsion), including proportional and non-proportional cycles, allow to scan all the deviatoric space and to study the influence of a complicated loading history thanks to direction changing.

All tests were carried out using Zwick/Roell axial and torsional testing machine. Both tensile force and torque were measured by a 25kN/200Nm load cell (Applied Measurements Ltd). The deformation measurement was done by a 3D Digital Image Correlation (DIC) technique (Aramis 4M, GOM, Germany) to analyze the homogeneity of the deformations. An axial/torsional extensometer (Epsilon Technology Corp, 3550 model) was also used to drive tests with the strain rate.

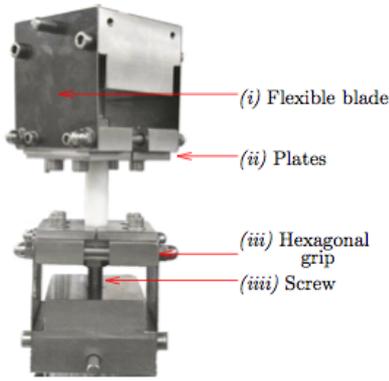


Fig. 1: Experimental device for multiaxial tests

To be useful, this experimental device needs an appropriate specimen. This specimen (Fig. 2) is designed with the objective to decrease the initial mechanical stresses by using blocking solutions to drive the sample instead of squeezing method used in standard tests as tensile ones. These solutions are visible in Fig. 1. The flexible blades are able to take into account the size of variation of specimen heads. The plates maintain the heads during the tensile loading when the screws have the same role during the compression loading. The hexagonal grips fit with the shape of the specimen heads to allow a torsion load.

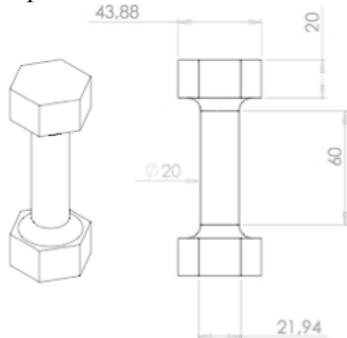


Fig. 2: Specimen for multiaxial tests

This specimen has also the advantage to have a single shape to perform all the tests and to avoid the use of sample shapes specially dedicated to tensile, compression or torsion tests. Indeed, if the sample's shape changed, it is difficult to be sure that the structure is still the same, because the structure of a polymer is strongly influenced by the specimen elaboration conditions and the cooling process management.

Experimental results

This section shows some examples of tests which are possible to get from this experimental set-up. These stress and strain are plotted in different ways according to the test studied. The Cauchy stress and the logarithmic strain are used for the tensile test. As the transformation is assumed isochoric, they are calculated with:

$$\sigma_{cauchy} = \frac{F}{S_0} (1 + (l - l_0)/l_0) \quad \text{Eq.1}$$

$$\varepsilon_{\log} = \ln(1 + (l - l_0)/l_0) \quad \text{Eq.2}$$

Where F , S_0 , l_0 , l are respectively the force, the initial surface of the specimen, the initial length and the current length of the extensometer.

The torsion test is based on the maximum shear stress and shear strain:

$$\tau_{max} = \frac{M_T}{J_T} r \quad \text{Eq.3}$$

$$\gamma = 2\varepsilon_{xy} \quad \text{Eq.4}$$

Where M_T , J_T and r are respectively the torque, the second moment of torsion and the specimen's radius. For tests combining tensile, compression and torsion loading, the von Mises equivalent stress and strain are used:

$$\sigma_{mises} = \sqrt{\frac{3}{2} \bar{\sigma} : \bar{\sigma}} \quad \text{Eq.5}$$

$$\varepsilon_{mises \log} = \sqrt{\frac{2}{3} \bar{\varepsilon}_{\log} : \bar{\varepsilon}_{\log}} \quad \text{Eq.6}$$

Tensile tests

The first experiment performed is a tensile test until the specimen's rupture. Fig. 3 gives the results of a Mirel F3002 and a PPC7712. Both materials appears to have a value of Young modulus very close. The PHA presents a behavior more rigid but also more fragile than the Polypropylene as the strain at break is about 7.7% against 28.1% for the PPC7712.

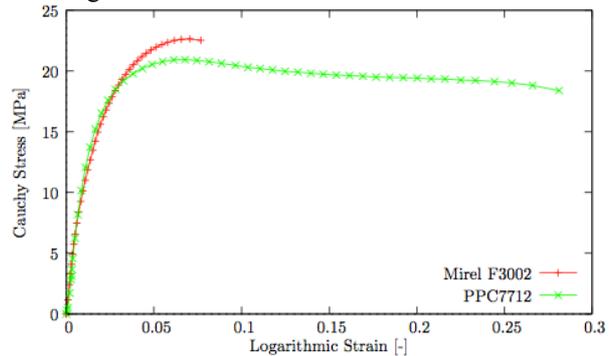


Fig. 3: Comparison between the F3002 and the PPC7712 for a tensile break test

Such test can be interrupted by relaxation steps as seen in Fig. 4. In this case, the relaxation steps are at 1% and 2% of strain with duration of 15 minutes each. A tensile test is also plotted to compare with the previous one. The loads of both tests were carried out at a strain rate of $4.10^{-4} s^{-1}$. The figure shows a discrepancy of the results which can occur with these specimen shapes since the load of both tests should coincide. This kind of test gives also information about the viscosity of the F3002, integrated in the 3D constitutive model which is presented in the next section.

Torsion test

The torsion test (Fig. 5) was carry out to approximately reach a shear strain of 5% at a shear strain rate of $4.10^{-4} s^{-1}$. This figure presents the results obtained for the PHA and the Polypropylene in considering the stress

distribution as linear along the radius. The F3002 and the PPC7712 shows a great similarity of shear behavior and the hysteresis area seems almost identical.

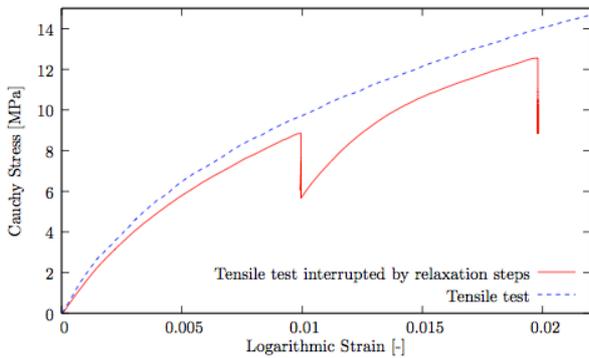


Fig. 4: Tensile test interrupted by relaxation steps of 900s at 1% and 2% of strain

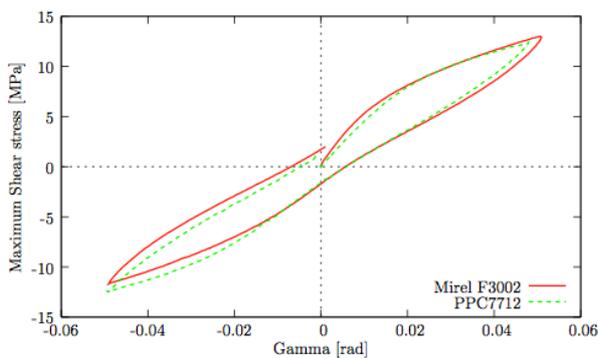


Fig. 5: Comparison between the F3002 and the PPC7712 for a torsion test

Test with complex loading paths

All combinations including tensile, compression or torsion loading simultaneously or successively are possible. The Fig. 6 illustrates one of these possibilities on a F3002 specimen. This test is composed of three steps. The first step consists in a compression test of -3mm (crosshead displacement) combined with a torsion test of -5° (Step 1 in the figure). This step is immediately followed by a second one (Step 2) with a tensile displacement of +10mm and a angle of torsion of $+17^\circ$. The last step (Step 3) corresponds to an unloading until the starting position.

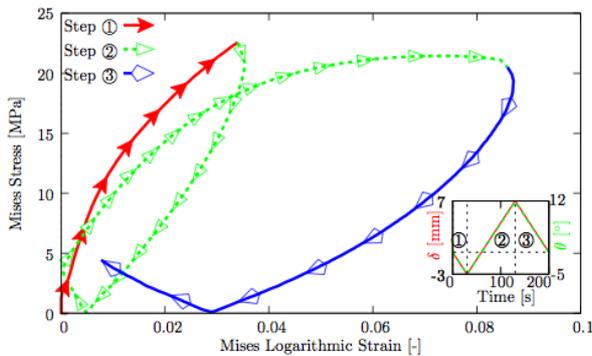


Fig. 6: Cycle combining Compression/Torsion then Tensile/Torsion loading and unloading

These tests allow to have an overview of the experimental set-up possibilities but also to note that

the F3002 has a similar behavior with the PP which seems to confirm that this PHA could be an interesting alternative to this Polypropylene. All these tests allow to have a large experimental database in order to get a 3D constitutive model able to simulate the F3002 behavior.

Numerical Simulation

To simulate the mechanical behavior of PHA, a constitutive model has to be chosen. The HyperVisco-Hysteresis (HVH) law [3,4] has a different approach than other Visco-Plastic models. This model is based on a superposition of stress contributions (Fig. 7) instead of splitting the strain. Three stress contributions are taken into account and assumed existing: the hyperelasticity (σ_e), the viscosity (σ_v) and the pure hysteresis (σ_h).

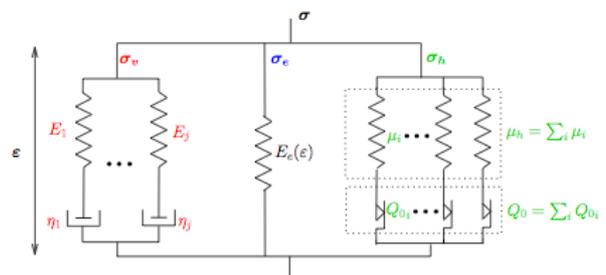


Fig. 7: The rheological view of the HVH law

The mechanical parameters needed for this model are then identified thanks to the experimental database. Once this identification is done, simulations are performed by FEA using an in-house finite element code Herezh++ [5]

The Fig. 8 gives an example of what it is possible to obtain with the simulation of food packaging structures in PHAs. The structure chosen is a quarter of bowl due to its symmetries and a localized force is imposed. The figure shows the von Mises stresses which occur with this loading condition.

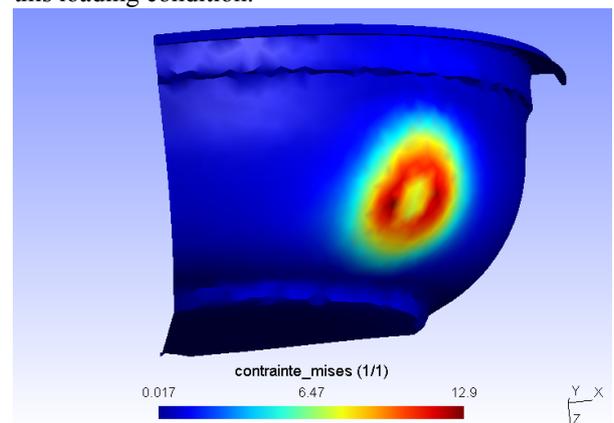


Fig. 8: Visualization of the von Mises stresses on a quarter of bowl under a point load

The next step of this work is now to optimize the numerical simulation and to be able to simulate other PHAs food packaging structures.

Conclusions

The goal of this work is to predict the mechanical behavior of food-packaging structures made in Polyhydroxyalkanoates, a biodegradable and bio-based polymer promoted to a growing interest in the plastic industry. To have such results, tests on this material have to be carried out in order to characterize its mechanical properties. The tests (tension, compression, torsion) presented here are original since they use one single shape of specimen to perform complex loads. Consequently, all these tests, performed with a new experimental device give us a better understanding of the 3D mechanical behavior of the material and show great similarities with a polypropylene used in the food-packaging industry. The next objective is to optimize the numerical simulations of food-packaging structures with the Hyper-Visco-Hysteresis constitutive model by improve the identification of the material parameters thanks to the experimental database. The final aim of this work is to promote Polyhydroxyalkanoates, innovative materials in terms of environmental impact.

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