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Research paper

On the identification of the coefficient of moisture expansion of polyamide-6: Accounting differential swelling strains and plasticization

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A B S T R A C T

Polyamide based composites are increasingly used in automobile industry. During their lifetime, they are subjected to aggressive environments including moisture which acts as a plasticizer of the polymer network, affecting its mechanical behaviour. Besides, the heterogeneous in-depth moisture content induces a differential hygroscopic swelling that may eventually cause damage. In practice, hygroscopic expansion depends on both the moisture content and coefficient of moisture expansion. Often, the polymer database do not provide detailed information on the dependence of the coefficient of moisture expansion on the moisture content. This work describes a multiscale and multiphysics approach enabling to identify the local evolution of the materials parameter of the hygro-mechanical behaviour law of a neat PA6 polymer, occurring during the transient stage of humid aging tests, from the numerical simulation of the dimensional change experienced by macroscopic samples. This study highlights the nonlinear evolution of the measured hygroscopic strain at the beginning of the moisture diffusion, as well as the need to account for the water induced plasticization effects (i.e. the fall down of the Young's modulus of the polymer material), for building an appropriate, effective modelling of the material behaviour.

1. Introduction

Passenger cars are still mainly propelled owing to fossil fuels. Nevertheless, fossil fuels supply became a critical geopolitical issue since World War II (Patterson, 1964). Moreover, fossil fuels combustion is a critical source of greenhouse gas emission. Greenhouse gases such as carbon dioxide (CO_2) contribute to the global warming (Judkins et al., 1993). Fossil fuel consumption by passenger cars is strongly related to the mass of the vehicles (Cheah, 2010). According to regulation (EC) No 443/2009 of the European Parliament and of the Council, the passenger car standards are 95 g/km of CO2, phasing in for 95% of vehicles in 2020 with 100% compliance in 2021. Consequently, in Europe, the automobile industry is looking for cutting dramatically the weight of new vehicles. In order to reach that goal, lightweight materials such as magnesium alloys, aluminium alloys, high strength steels, or polymer composites can be used instead of the more traditional cast iron and conventional steels (Kulekci, 2008; Hirsch, 2014). In that context, materials selection is thereby determined by economic issues as much as by materials and components characteristics or properties.

In this field of applications, glass reinforced thermoplastics tend to concentrate on polypropylene or Nylon as the base resin. Among the engineering thermoplastics, these materials are actually inclined to be the most inexpensive whereas they are the most easily processed. Nevertheless, both these materials are sensitive to environmental conditions (temperature and moisture) relative to vehicle requirements for structural parts (Beardmore and Johnson, 1986).

During their service life, passenger cars are often submitted to variable hygroscopic environments. Polymer matrices such as polyamides may absorb significant amounts of water when exposed to humid environments. Absorbed water induces dimensional change during both the transient stage (differential swelling), and the permanent stage of the diffusion process (Kawasaki et al., 1962; Monson et al., 2008; Starkweather, 1959). Besides, moisture absorption can adversely affect most physico-mechanical properties of polymers. One of the main effects of water on polymer matrices is, among others, plasticization (Chang et al., 2000; Mali et al., 2005; Myllytie et al., 2010; Abacha et al., 2009; Dlubek et al., 2002). Water can be associated to specific sites of the macromolecular backbone, through hydrogen bonding or polar interactions (McBrierty et al., 1999). This sorption mechanism induces a reversible reduction of both the glass transition of the polymer (Kaimin' et al., 1975) and its mechanical properties such as the

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Young's modulus (Nielsen and Toftegaard, 2000; Yakimets et al., 2007), as well as the mechanical strengths (Selzer and Friedrich, 1977; Hassan et al., 2012). The effect of moisture can be critical in products that may be exposed to different weather conditions due to change in seasons or geographic locations. Thus, one needs to take into account this factor in material pre-selection stage, plastic parts design, mechanical performance prediction and optimization.

The present work will be devoted to the study of a PA 6 resin intended to be used in passenger vehicles, as the matrix of structural composite parts. Experimental investigations will be realized in order to characterize the kinetics of diffusion. Besides, the fall down of the macroscopic tensile modulus as well as the dimensional change experienced by the samples, will be characterized during the transient part of the diffusion process. In order to achieve the identification of the hygro-mechanical Hooke's law of the studied material, an original methodology, featuring a multi-scale, multi-physics model, will be presented. Owing to numerical simulation, we will show that this method makes it possible to find the local behaviour law enabling to appropriately reproduce the results gathered, at macroscopic scale, during the experimental characterization of laboratory-size specimen which experience a non-uniform moisture content profile during the transient part of the diffusion process. In this context, the necessity to account for plasticization effects (i.e. the softening of the material) for an improved prediction of the global strains experienced by the samples, in particular at the beginning of the diffusion process, will be underlined. Another section of the present work will be devoted to purely numerical examples, focused on discussing the fields predicted by the identified model for various quantities that cannot be directly compared to experimental quantities, namely, the evolutions of the local properties (Young's modulus and coefficient of hygroscopic expansion), along with others quantities of interest, such as the moisture content and the mechanical states.

2. Experiments

2.1. Materials

This study was made on an unreinforced PA6 resin, provided by Solvay (a project partner). Polyamides have interesting mechanical properties such as a strong fracture toughness compared to epoxies (Mark, 2007). Besides, they show a good resistance to various chemical products. Polyamides and their composites are used in several application fields for mechanical parts designed for being subjected to mechanical stress and high temperatures. They can also be used in mechanical automotive construction for various parts such as: plain bearing, body coils, guide and coupling parts, nuts and slides.

2.2. Samples

The samples were cut with a water jet from plates (of 2 mm thickness), obtained by injection molding. The resulting specimen present the following geometries: a dumbbell shape with rounded junction according to norm (ISO 3167) for tensile tests (30 samples), or a parallelepiped shape of dimensions $200 \times 20 \times 2$ mm^3 for moisture diffusion kinetics as well as swelling hygroscopic swelling characterizations. 5 samples were dedicated to the kinetics and swelling identification.

These samples were referenced and undergone a polishing with sandpapers with 4 grains sizes (mean diameters of 68, 25.8, 18.3 and 15.3 µm, respectively). Thereafter, a final micro polishing by alumina powder was achieved. Preliminary to the aging tests, the samples were conditioned under a vacuum for 24 days. The water loss was followed during this stage, until a constant mass was obtained: the conditioned samples experience, as a result, an initial homogeneous in-depth moisture content, prior to the beginning of the aging tests.

Finally, the specimens intended to be aged are placed in a controlled climatic chamber at 80% relative humidity (RH) and at a temperature of 24 °C.

2.3. Water sorption

Five specimens undergo a gravimetric monitoring to measure the water uptake (by using a laboratory balance with precision of 10^{-4} g). The time interval between two measures increases, as the rate of moisture absorption decreases with time, to reach a stage when one measurement by week becomes sufficient. The purpose of monitoring the water uptake is to identify the diffusion kinetics of investigated polymer.

The absorption of moisture in a hydrophilic material is primarily characterized by two quantities:

- The maximum moisture absorption capacity C^{∞}. It corresponds to the water content that the material can absorb in given environmental conditions, at the saturation of the diffusion process.
- The diffusion coefficient D, which represents the velocity rate at which moisture enters the material.

In order to identify both parameters, one often draws the classical sorption curve, i.e. the macroscopic water content $\overline{C}(t)$ as a function of the root square time t. An example of diffusion curves is presented on Fig. 1, where the water content is calculated by the following equation

$$\overline{C}(t) = 100\,\frac{m(t) - m_0}{m_0} \tag{1}$$

In expression (1), $m(t)$ stands for the weight of a specimen at time t, whereas m_0 is the weight of the same sample, at the initial time. This gravimetric monitoring enables to determine the macroscopic (overall) moisture content but does not make it possible to obtain the spatial distribution of this quantity during the absorption process. A model is thus required to predict the local fields of moisture within the sample, during the transient stage of the aging test. There are various laws describing the sorption of a material. We will only focus on the classical uncoupled Fick model (Fick, 1855), since the studied samples present typical Fickian kinetics of diffusion.

The kinetics of diffusion is identified by the mean of the optimization problem (minimizing the least-squared difference between the experimental data $\overline{C}(t)$ and the exact solution of Fick (Crank, 1978)), which is solved using a classical algorithm of optimization (Lagrias et al., 1998) based on some assumptions on the material properties and the geometrical definition.

An example of a diffusion parameters identification on the studied neat resin samples is presented in Fig. 1. The very good agreement between the identified Fick model and the experimental data shows that the studied materials absorb moisture according to a Fickean kinetics. The identified parameters are presented in Table 1.

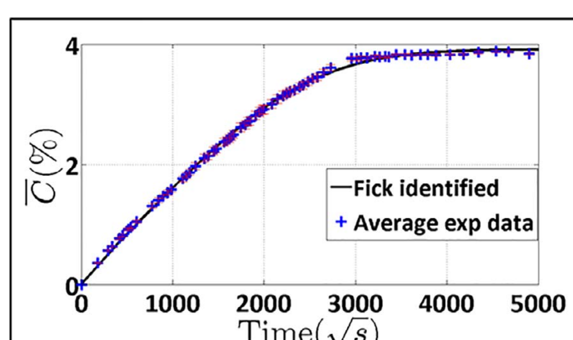

Fig. 1. Average experimental and identified data with Fick solution for one specimen of neat resin.