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Assessment of 3D moisture diffusion parameters on flax/epoxy composites

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ABSTRACT

This paper aims at assessing the experimental and analytical parameters of moisture diffusion of quasi-unidirectional and twill flax-fibres reinforced epoxy composites. The tested specimens of these composites were elaborated by hand lay-up technique at room temperature by using a vacuum moulding process. For several days, the samples were subjected to three ageing conditions, in order to evaluate the percentage of moisture uptake according to the ageing time. Next, the collected experimental data were analysed by using an optimisation program of Matlab software in order to identify the 3D diffusion parameters. The obtained results showed that the morphology and the anisotropy of flax fibres had a significant influence on the moisture diffusion. It also showed that moisture diffusion behaviour predicted by the 3D Fick model was in good agreement with the experimental results.

1. Introduction

Under growing pressure from ecological concern and environmental regulations to reduce CO_2 emissions, the market demands more new eco-friendly materials that present low environmental impact and which are recyclable at life end. For instance, according to the European Commission Directive 2000/53/EC, manufacturers of vehicles in the European Union are encouraged to reduce the quantity of waste and to reuse and recover at least 95% of the weight for all end-of-life vehicles by the end of 2015 [1]. Thus, car companies such as Mercedes and PSA Peugeot Citroën have developed several programs with "green materials" and use recycled plastics, natural materials (with vegetable fibres) and bio-sourced materials. For example, PSA Peugeot Citroën is aiming at reducing the proportion of fossil plastics, with an average of 30% per vehicle, and replacing them with green materials by 2015.

The specific tensile properties of vegetal fibres were in some cases better than those of glass fibres [2,3]. This suggests that vegetal fibres, as flax and hemp fibres, have a potential of being used in many applications [4], thanks to their biodegradability, renewability, great performance, low cost per weight [5], and low relative density [6–8]. Unfortunately, several factors can restrain their use in different domains and can influence their properties. In fact, the main drawback with using these green materials remains their degradability under different environmental conditions, in particular their inherent susceptibility to the moisture absorption [9–14]. Therefore, it is necessary to know the kinetics of diffusion processes of these new materials in humid conditions to promote their development.

To deal with this issue, the diffusion of water through these materials should be studied and this requires knowledge about diffusion parameters. Most models which estimate these parameters are based on Fick's laws, mostly used with a one-dimensional (1D) approach [15–19]. For example, Barjasteh and Nutt [15] assessed the moisture absorption behaviour of an unidirectional carbon/-glass hybrid composite. These authors employed the 1D model combined with a 2D diffusion equation in cylindrical coordinates to calculate moisture diffusivities in the longitudinal and the radial directions. These authors also showed that the longitudinal diffusivity along the fibre/matrix interface was four times greater than the radial diffusivity. This low radial diffusivity is attributed to the more tortuous diffusion pathways in the radial direction.

In the case of anisotropic composites, water diffusion depends on many parameters, such as reinforcement geometry, fibre nature and its morphology, temperature, resin and sample dimensions. These factors indicate that the water diffusion has a more complex behaviour and using 1D analysis is most likely not sufficient to study the water diffusion phenomena. A better method is to use three-dimensional (3D) analysis which takes into account the diffusion coefficients D_x, D_y and D_z in three directions x, y and z, respectively. Arnold et al. [20] developed a method to identify the 3D moisture diffusion coefficients of quasi-unidirectional carbon-fibre reinforced epoxy. From an optimisation process on a

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full data set of different exposure conditions, they showed that the full 3D Fick solution is clearly the most rigorous method to determine the diffusion coefficients D_x, D_y and D_z. They also found that the moisture diffusion along the fibres direction was significantly higher than the diffusion in other directions (i.e. in the directions y or z). Similar conclusions, about the interest of using the 3D solution to Fick's law, were drawn by Aronhime et al. [21] in unidirectional Kevlar–epoxy composites with different values of volume fraction. Pierron et al. [22] proposed a novel method for the identification of 3D moisture diffusion coefficients and saturation levels from gravimetric curves for Fickian models. They used the 3D Fick equation and an optimisation process to fit experimental data points. These authors showed that diffusion coefficients and the saturation level can be identified from gravimetric curves obtained from unsaturated coupons. Blikstad [23] investigated the experimental 3D diffusivities by exposing graphite/epoxy laminates specimens of special design to the moist air. He used the 3D Fick model in order to test if it can take into account the moisture absorption behaviour observed in their specimens. First of all, he showed that the diffusion along the fibre direction is about ten times greater than the diffusion in the other directions. Then, he underlined that the experimental results of the 3D diffusion were in good agreement with calculations based on the 3D Fick model. In a recent work, Jiang et al. [24] used specimens of three different forms (square, rectangular and small square) in order to obtain 3D moisture diffusion coefficients D_x, D_y and D_z of glass-fibre reinforced polymer laminates. From the analytical model of 1D and 3D moisture theory, these diffusion coefficients were determined by the best least-square curve fitting to the experimental data. The obtained results showed that the 3D moisture diffusion theory is inevitably needed to develop the optimisation scheme. Furthermore, it is found that the moisture was inclined to mainly diffuse along the fibre direction.

Despite these numerous works for traditional composites, few researchers studied moisture absorption from a 3D analysis on vegetal fibre composites. The main objective of this work is to assess the experimental 3D moisture diffusion parameters (D_x, D_y, D_z) and the maximum moisture uptake (M_∞) of epoxy composite materials. These were reinforced with quasi-unidirectional or twill flax-fibres and were subjected to three ageing conditions. The first and second ageing were characterised by immersion into water at 20 °C and 55 °C, respectively. The third was defined by a Relative Humidity (RH) of 75% and a temperature of 55 °C. Next, the collected experimental data was analysed by using an optimisation program of Matlab software in order to assess the analytical 3D diffusion parameters.

2. Analytical model

Polymeric composite materials absorb water via its surfaces when immersed in water and the water is often assumed to penetrate into the material following the laws of diffusion. In many cases, two categories describe the diffusion behaviour: Fickian diffusion and anomalous diffusion. It is from the shape of the sorption curve (i.e. the water absorption according to the square root of the time) that these two categories can be distinguished. The first based on Fick's law was the classical single free phase model of absorption. In this model, the hypothesis of free displacement of water molecules is correct and water molecules are not combined with the matrix. The second depicted anomalous diffusions, i.e. deviations from Fick's law. The sorption curve can be characterised by the presence of two-stage sorption, by the occurrence of mass loss or by the lack of saturation plateau. In the case of sorption curve without saturation, the Langmuir two phase model was used. It is based on the hypothesis that the penetrant molecules are divided into two natures: a free diffusion phase and a second combined phase which does not involve diffusion. Contrary to the Langmuir's model, the Fick's model predicts that the water absorption increases linearly with the square root of the time, and then gradually slows down until an equilibrium plateau is reached. This is reflected in what was observed in the materials of this study and generally in polymeric composites [15–24]. It is within this context that we used the Fick's model to assess the experimental 3D moisture diffusion parameters of flax-fibre reinforced epoxy composite. For this purpose, the composites materials have been assumed to be assimilated as homogeneous materials and the diffusion to be constant throughout the material in the case of 1D diffusion.

Fick's law, which enables us to predict the moisture diffusion according to time and space, is given by the following equation:

$$\frac{\partial C}{\partial t} = div(-D\overrightarrow{grad}C) \tag{1}$$

where C is the moisture concentration and D the diffusion tensor.

For a thick orthotropic sheet, Fick's diffusion, following x, y and z directions, can be described by the following equation:

$$\frac{\partial C}{\partial t} = D_x\frac{\partial^2 C}{\partial x^2} + D_y\frac{\partial^2 C}{\partial y^2} + D_z\frac{\partial^2 C}{\partial z^2} \tag{2}$$

where D_x, D_y and D_z are the diffusion coefficients in the x, y and z directions, respectively.

The solution of full 3D Fick's model was found by integrating the solution of Eq. (2) relative to the sheet volume, taking into account boundary and initial conditions, and is given by [20]:

$$\frac{M_t}{M_\infty} = 1 - \left(\frac{8}{\pi^2}\right)^3 \sum_{i=0}^{\infty}\sum_{j=0}^{\infty}\sum_{k=0}^{\infty} \frac{\exp\left(-\pi^2 t\left(D_x\left(\frac{2i+1}{L}\right)^2 + D_y\left(\frac{2j+1}{l}\right)^2 + D_z\left(\frac{2k+1}{h}\right)^2\right)\right)}{\left((2i+1)(2j+1)(2k+1)\right)^2} \tag{3}$$

where M_t is the moisture uptake at time t, M_∞ is its maximum moisture uptake at equilibrium state. L, l and h are the sample dimensions along the x, y and z axes respectively.

To identify the diffusion parameters, the 3D Fick analysis can involve a long time of computation. However, several approximations were proposed in literature to reduce this computation time. Based on the approach of Shen and Springer [25], works of Bao and Yee [26] suggested that, in the early stage, the diffusion in x, y and z directions could be treated independently from each other. Consequently, the weight gain of the specimen is the sum of the water diffusion in all directions. The initial weight uptake behaviour is thus predicted by the following equation, called Approximation 1:

$$\frac{M_t}{M_\infty} = \sqrt{\frac{16t}{\pi}}\left(\frac{\sqrt{D_x}}{L} + \frac{\sqrt{D_y}}{l} + \frac{\sqrt{D_z}}{h}\right) (\text{Bao \& Yee [26]}) \tag{4}$$

Unfortunately, this approximation does not take into account the diffusion at the sample edges. That is why Starink and Chambers [27] proposed a better approximation, given by Eq. (5) and called Approximation 2:

$$\frac{M_t}{M_\infty} = \sqrt{\frac{16t}{\pi}}\left(\frac{\sqrt{D_x}}{L} + \frac{0.54\sqrt{D_y}}{l} + \frac{0.54\sqrt{D_z}}{h} + \frac{0.33L}{lh}\sqrt{\frac{D_yD_z}{D_x}}\right)$$
$$(\text{Starink \& Chambers [27]}) \tag{5}$$

For a plane sheet of thickness much lower than the length and the width, the moisture diffusion is mainly carried out in the thinnest part of the specimen, i.e. in the direction following the thickness. In this case, the expression of 3D Fick's solution (Eq. (3)) can be simplified as follows: