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Identification of the orthotropic diffusion properties of RTM textile composites for aircraft applications

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A B S T R A C T

The paper focuses on a novel strategy for the rapid identification of the orthotropic diffusion properties of RTM textile composite materials for aircraft applications. The method consists in performing gravimetric tests on composite samples and then employing the Proper Generalised Decomposition method for the identification of a 3D orthotropic Fickian model. The method consists in calculating once and for all the solution for a given range of the diffusion parameters and then in particularising such a solution in accord with the experimental results by an optimisation procedure. Analysis and discussion of results show that the identification can be performed successfully.

1. Introduction

Organic composite materials are sensitive to humid ageing: water absorption taking place by means of chemical mechanisms (bonding of the water molecules with the macromolecules of the organic phase of the composite) may induce reversible or irreversible degradation of the matrix and of the fibre/matrix interfaces and promote a consistent change of the mechanical properties of the composite material [1,2]. Since water diffusion by chemical mechanisms is relatively slow (compared for instance to thermal diffusion or water penetration on pores/cracks by capillarity), species (or bonds) concentration gradients are likely to develop, therefore degradation phenomena take place at a local scale, which is usually microscopic, depending on the extent of the gradient [1,3]. Despite the consistent development of Fourier transform infra-red spectroscopy (FTIR) and nuclear magnetic resonance (NMR) techniques for detecting bound water molecules in polymers [4–7] so far it is not possible to directly measure water concentration in massive composite samples: FTIR spectroscopy is optimised to measure species gradients on samples surfaces or in membrane polymer samples, NMR techniques are particularly adapted to detect free water in porous/cracked materials. Therefore, in order to estimate water concentration and property profiles in materials and structures, it is essential to perform numerical simulations.

The classical Fick's model (the equivalent of Fourier's model for thermal conduction) is often employed within the context of industrial applications. Diffusion anomalies (departure from Fickian behaviour) are sometimes not much marked in carbon fibre reinforced polymers composite materials, since the polymer behaviour is often masked by the presence of fibres, unless damage phenomena take place [8,9]. In other cases, anomalies are visible only after long exposure times [1–3,10]: moreover, in many cases, the true diffusion behaviour of a material can be considered as a linear superposition of a pure Fickian behaviour and of a non-Fickian anomalous, usually the result of chemical reactions appearing after long exposure times. In this context, the Fick's model is more than appropriate to gain a pertinent estimation of the humid behaviour of a material and in particular to establish the diffusion directionality, which is particularly useful for numerical simulation and structural design. However, even in the Fickian context, the identification of parameters must be carried out: in particular the directionality of water diffusion must be established and the diffusion coefficients must be found, only one coefficient for isotropic materials, three a priori distinct coefficients for orthotropic materials and up to six coefficients, for anisotropic materials. This step is not as straightforward as it may seem since, as discussed, the full water concentration field cannot be directly measured, contrary to the case of thermal conduction, in which the full thermal field can be measured in several circumstances of practical interest. To identify the parameters of a diffusion law, a sorption test is often carried out: this test involves performing gravimetric measures of samples exposed to a given humid environment and following

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the specimen mass evolution as a function of time. If the initial mass of the sample is systematically subtracted to such measures, the extent of water mass in the sample is measured. For an ideal Fickian behaviour, the sample mass increases linearly with square root of time, at the start of the conditioning, then reaches a saturation plateau [1–3,10]: by these experimental measures, it is possible to infer the saturation value of water concentration and the diffusivity value. For isotropic materials (one value of the diffusivity coefficient) only one gravimetric curve suffices; for more complex behaviour (orthotropic or anisotropic) more gravimetric curves should be employed and, in general, one curve for one diffusion coefficient.

The employment of gravimetric curves as a tool to identify the diffusion behaviour of a material calls for several comments, listed here:

- water mass measures should be seen as a measure of the average species concentration, over a volume of material. In the presence of Fickian behaviour, usually some elementary reversible mechanisms take place; however the exact nature of such mechanisms is often unknown, the sole information which can be gained is the apparent coefficient of diffusion. It must be noted that – even in the simplest context of Fickian behaviour – the identification of the diffusion properties is not straightforward, since the concentration field calculated by Fick's law is integrated over a volume of material, to get the mass uptake,
- for identification purpose, analytical solutions are available only in few special cases: within the context of Fickian behaviour, only isotropic and orthotropic solutions are available for boundary conditions which are constant [10] or variable with time in a periodic manner [11],
- numerical solutions for instance solutions based on Finite Element Methods (FEM) – can be time consuming: this is particularly true when simulation of non-Fickian behaviour is concerned, but is valuable also for the pure Fickian case [12].

Recent literature reports the employment of analytical methods for the identification of diffusion behaviour [11,13–15]: as mentioned, this methodology is applicable only for special cases. While re-update of FEM solution for identification purposes can be highly time consuming, identification methods based on Proper Generalized Decomposition (PGD) techniques can be successful. This technique pertains to the class of numerical methods based on reduced order models and consists in calculating once and for all the solution for a given set of parameters. The identification can be performed exploring the parametric solution by means of an appropriate optimisation technique. A first step towards this aim has been presented in [12] for the isotropic case, leading to consistent computational time saving with respect to FEM.

The present paper puts forward the analysis carried out in [12] enlarging to the case of orthotropic materials. In this case, the number of parameters to be identified is higher and a specific identification strategy must be developed. With respect to [12] which a commercial Dakota software was employed, in the present analysis a Newton algorithm is implemented. This requires to numerically differentiate the parametric PGD solutions with respect to the parameters to evaluate the Hessian and the Jacobian matrix of the functional.

The method is then employed for the identification of the orthotropic diffusion properties of RTM AS7/RTM6 textile composites for aircraft applications. For this material, the diffusion behaviour of the pure resin has been investigated and discussed in reference [16]. However, no data are available for the composite. This information can be useful for understanding the diffusion behaviour of the material, for performing numerical simulations on structures and for structural design and optimisation.

The present analysis differs from that carried out by Tang et al. [17], since in that case a micromechanical model of moisture diffusion in textile composites is approached, explicitly modelling the diffusion behaviour of the elementary constituents of the textile composite – matrix and fibrous tow. In the present analysis, the textile layer is modelled by hypothesis as an homogeneous orthotropic material with respect to diffusion.

It should be noted that – for a certain class of composite materials for instance composite laminates, for which the degree of orthotropy is presumed a priori gravimetric curves are obtained for very thin samples, enhancing the diffusion behaviour along a given direction. In this case, since diffusion is essentially one-directional, the identification is performed by employing simple one-dimensional solutions or quasi-three-dimensional solutions obtained by imposing simplifying assumptions on the full 3D solutions (see reference [13] for a detailed and interesting account of such methods). This method despite its simplicity is difficult to apply to materials with complex microstructures, for instance to three-dimensional textile composites, since it can be difficult to conceive a thin sample of such materials. In this case, a full 3D orthotropic solution must be employed and an appropriate identification strategy must be developed, like the one proposed in the present paper.

The paper is organised as follows. Section 2 is devoted to the presentation of the material, of the experimental setup and gives results of the gravimetric test. The PGD method is briefly revisited in Section 3 and precedes the presentation of the identification procedure within the PGD framework. This identification strategy is investigated in Section 4 to determine the diffusion parameters of the material described in Section 2. The results are then validated and discussed with respect to experiments. Finally Section 5 gives conclusions and perspectives for future work.

2. Experimental setup: gravimetric curves on composite samples

The composite material is made by a RTM6 matrix by Hexcel [18] and a "high strength" AS7 carbon fibre manufactured by the same supplier [19]: the fibre has circular section with a diameter of 6.9 μm. The preform employed to produce structural components, with reference name HexForce® 48502 XD1200, is a 2D 5-harness satin fabric each tow containing 12,000 filaments of carbon, 12 K, powder-free, 500 $\mathrm{g \cdot m^{-2}}$ weight. The thickness of a ply is 0.47 mm.

The hygrothermal behaviour of the RTM6 resin has been discussed in a recent paper by Simar et al. [16] showing the presence of diffusion anomalies strongly dependent on the test temperature and related to thermo-oxidation phenomena: the anomalous diffusion–reaction behaviour of the resin can be approximately considered as the sum of a Fickian diffusion process and a series of thermally activated chemical reactions which become apparent after long conditioning times. In any case the Fickian part of the diffusion–reaction process can be reasonably well identified, at several values of temperature (T) and relative humidity ($RH\%$) [16]. For $T = 70\,^{\circ}\mathrm{C}$ and $RH = 85\%$ (environmental conditions which are of interest for the scope of the present paper) an apparent diffusion coefficient equal to around $1.6 \times 10^{-2}\ \mathrm{mm^2 \cdot h^{-1}}$ (the behaviour of the pure resin is considered as isotropic) and an apparent water mass at saturation equal to around 1.5×10^{-2} g (corresponding to a mass fraction of around 2.23% and to a saturation concentration of around $2.6 \times 10^{-5}\ \mathrm{g \cdot mm^{-3}}$) can be identified.