INFLUENCE OF EPOXY/GLASS FIBRES INTERFACE/INTERPHASE ON WATER DIFFUSION KINETICS: EXPERIMENTAL AND NUMERICAL APPROACH

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Abstract

This study deals with the water diffusion in unidirectional composite by three approaches: experimental measurements, analytical and numerical models. In the initial state, localized measurements at fibre/matrix interphases show a decrease of the stiffness at decreasing distance from the fibre surface. An interphase with a higher molecular mobity is then defined around each monofilament. A Fickian diffusion kinetic was determined from water absorption measurements on both the resin and the UD composite. The numerical simulation shows that the matrix is strongly modified by the presence of fibres, compared to the resin alone. A matrix diffusion coefficient higher than the bulk resin one must be used to fit correctly the experimental data. This is consistent with the decrease of stiffness highlighted by AFM measurements in the interphase.

1. Introduction

High mechanical characteristics are observed at the initial stage for composite used in aeronautical applications. Under wet service conditions, the decrease of the mechanical properties can be explained by the degradation of the interfaces and/or interphases present around each fibre [1–3]. The origin of the interphase can be explained by an incompleted curing stage of the matrix [4–6]or to plastizing effects due to fibres surface treatment [7–10]. These specific areas generally are generally softer than the bulk matrix. Due to small sizes of the interphases in the composite, submicron scale methods must be used [11,12]. In presence of humid environment, the water diffusion kinetics will be modified in the interphase. An increase of water diffusivity was observed [13] due to the higher molecular mobility inside the interphase than inside the bulk matrix.

In a first part, experimental investigations at sub-micron scale will precise the size and properties of theses interphases. A second part will be dedicated to the numerical models to describe the behaviour of the composite in immersion and the consequences of water diffusion on the stress levels inside the material.

2. Materials

Resin plate based on DGEBA-based epoxy (i.e. without fibres) and unidirectional composite based on the same resin with E-glass fibres treated with a commercial sizing. Details are described in a previous work [14].

3. Experimental measurement of the interphase size

Micro thermal analysis is a local probe technique which consists in heating the sample from room temperature to a maximum temperature with a high rate. The aim is to determine local transition temperatures such as the glass transition temperature (i.e. softening temperature T_s). Localized measurements on the resin samples give T_s values between 110°C and 127°C as represented between dashed lines in Figure 1. The results show a decrease of the T_s when the distance from fibres surface decreases [8]. The interphase region is then defined by the area where the T_s is lower than the T_s of the bulk. So, Figure 1 shows an interphase around 10 µm width. This interphase area can be divided in two parts:

- T_s is slightly modified (less than 10% of the bulk resin values);
- T_s is highly modified (more than 10% of the bulk resin values).

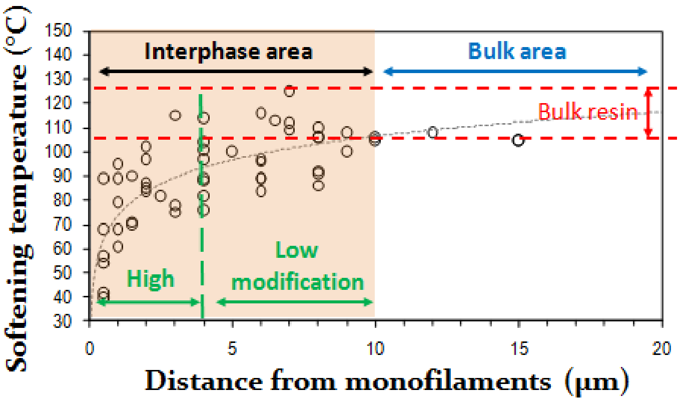


Figure 1. Ts evolution vs. distance from monofilament for epoxy/glass fibre composite

4. Hydrothermal ageing

Composite and resin plates (75 x 75 x 2 mm3) were immersed at 70 °C in deionized water up to 16 weeks according to European aerospace standard NF EN 2489 which is similar to ASTM D5229. The water uptake was measured regularly during ageing and the water absorption was then reported vs time. The kinetic diffusion of the water is well described by a fickian law (Figure 2).

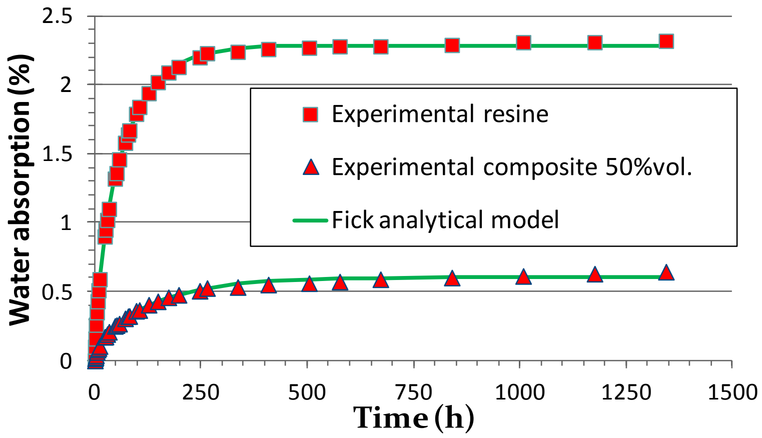
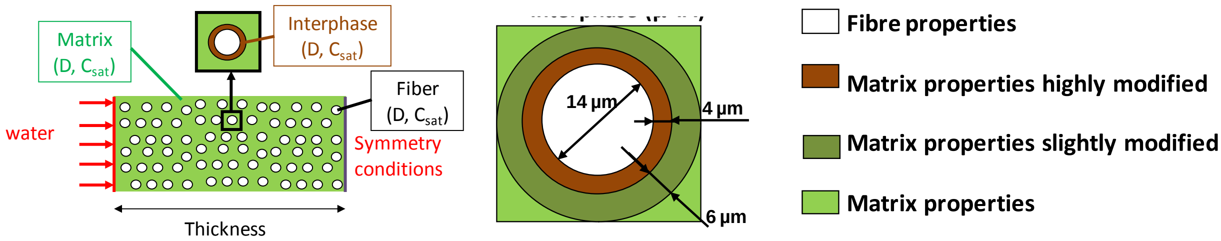


Figure 2. Water absorption curve vs time - experimental and Fick values

5. Numerical models

Numerical model is based on a coupled diffusion/mechanical approach. Parameters used in the model come from experimental characterization or literature [15]. The contact interactions between each part are done by a permanent contact (Tied). Boundaries conditions and interphases of the numerical model are shown by the Figure 3. Composite geometry is discretized with quadratic triangular elements.



(a) (b) (c)

Figure 3. Water diffusion model (a) and interphase (b)

As expected the water absorption generates stress in the composite due to the water expansion coefficient of the matrix. The stress is especially high in the confined areas (matrix areas where fibres are close. As shown in the Figure 4, the stress can be increased up to 18 MPa. Moreover, no cracks have been observed by scanning electron microscopy on the aged samples at the interface or in the matrix.

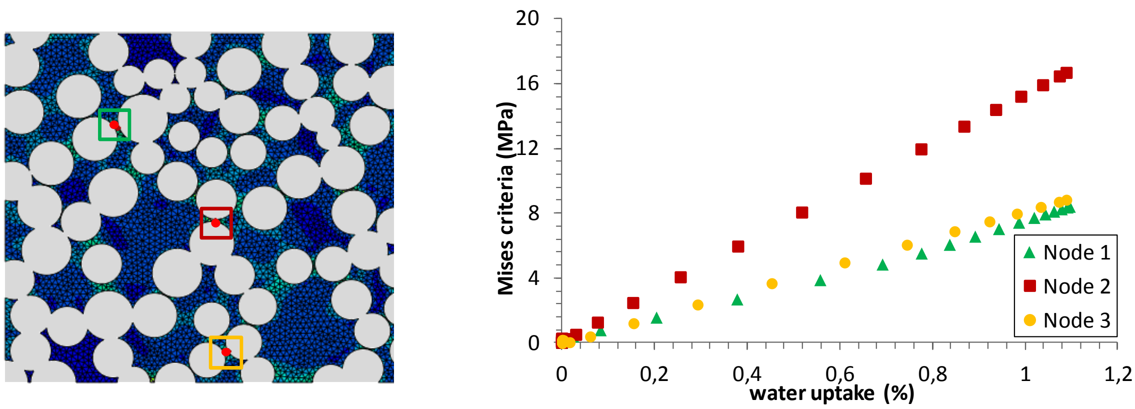


Figure 4. Stress values in matrix composite due to water absorption

6. Conclusion

Localized measurements inside the UD composite show an interphase with a gradient of properties around each glass monofilament. Below 4 µm from fibres surface, the molecular mobility is much higher than the bulk resin one. This is consistent with an important decrease of the elastic modulus determined by AFM force measurements. It is interpreted by a decrease of the crosslink density and/or by plasticizing effects due to fibre surface treatments.
The influence of this modified area on water diffusion kinetics is then studied by gravimetric measurements and finite element analysis. The kinetics of water uptake in the neat resin and composite are correctly modeled by a fickian law. The corresponding diffusion parameters are then implemented in numerical models. To fit correctly with experimental results, a realistic microstructure with the presence of the interphase was considered in numerical models. In a first approach, the interphase is defined by a layer of 4 µm width with homogeneous properties around each monofilament. The heterogeneity of fibre diameters and distribution including some contacts between fibres were also taken into account into the model. The confrontation between experimental data and the numerical model leads to the determination of a diffusion coefficient five times higher for the interphase than for the bulk resin. This is consistent with the increase of molecular mobility measured by µTA in that area. Moreover, the stress-diffusion model enables the determination of stresses generated by water uptake at fibre-matrix interfaces.
The evolution of interphases properties during ageing is under process. The aim is to correlate micro- to macro- mechanical properties of the aged composite.

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