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Modeling of Hygrothermal Damage of Composite Materials

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ABSTRACT

Composite materials have been used extensively in various applications, such as mechanical engineering, aerospace engineering, and aviation thanks to their interesting mechanical properties. However, a substantial drawback in the use of such composite materials is that they absorb a significant amount of moisture when exposed to severe hygrothermal conditions. This factor dramatically affects the composite material's various physical and mechanical properties. This paper proposed a new model to predict the amount of moisture absorbed by a composite polyester/glass fiber composite material before and after hygrothermal damage. Two damage models were proposed and implemented in ABAQUS. A numerical simulation was used to estimate hygrothermal stresses in a composite plate. The results showed that the moisture absorption followed a Fickian behavior; if the temperature was low, the plate would be damaged, but the moisture diffusion rate accelerated when the hygrothermal parameters (temperature and humidity) were high. Additionally, the magnitude of residual stress, which was at its maximum at the beginning of the absorption, started to decrease until reaching zero when the plate was saturated.

1. Introduction

In the last decades, the use of composites has rapidly expanded in engineering applications. Thanks to their relatively good mechanical properties and especially their low densities (lighter structures), these materials have been used in many areas, such as aerospace and civil engineering. However, moisture in any form has unfavourable effects upon composites and often causes swelling and degradation. As a consequence, the mechanical properties of composites are decreased, especially in the presence of humidity at high temperatures [1-2]. Therefore, it is essential to control the evolution of composite material properties during hygrothermal ageing. For ambient temperatures, the moisture absorption phenomenon closely follows the classical Fickian diffusion law. However, deviations appear at higher temperatures [3], and Fick's Law loses validity. Abnormal increases in specimen weight were observed after prolonged exposure to moisture [4-6]. These phenomena are explained by the fact that humidity may cause damage within composites, mainly in the form of interfacial fiber/matrix micro-cracks. Experimental observations [7] showed that the micro-cracking phenomenon was highly random and difficult to quantify. Such damage occurred in composite samples during the early phases of moisture absorption. Analytical investigations [8] and numerical calculations [6, 7, 9] of the transient stresses caused within a composite material by moisture showed that large stress peaks occurred during moisture absorption. These stressed can be one of the causes of composite failure after a sufficient duration of moisture exposure.

Sharp moisture absorption may induce plasticization, saponification, or hydrolysis, which may lead in turn to reversible and irreversible changes in the microstructure of the polymer, consequently affecting their mechanical, chemical, and thermo-physical characteristics.

In general, there are three primary mechanisms by which water can penetrate a composite material [10]. The first is diffusion of water through the matrix. The second is capillary flow action and diffusion along the fiber/matrix interface. The third

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mechanism is the percolating flow and storage of water in micro-cracks. Water uptake is expected to affect glass/polyester composites and to induce a reduction in strength due to fiber/matrix deterioration [11-15]. For example, Mandell et al. [11] examined the sensitivity of iso- and orthophthalic polyester composites to water immersion. Their study provided strong evidence that composites made using orthophthalic polyester absorb more water as compared to isophthalic polyester composites. Fraga et al. [12] documented, based on SEM (scanning electronic microscopy) analysis, that the immersion of isophthalic polyester/glass composites in water at 80°C may induce a degradation of the fiber/matrix interface. They attributed the degradation to the oligomer extraction and hydrolysis of the silane coupling agents of the fibers. Along similar lines, Mouzakis et al. [13] studied the same kind of composites, and the results of their study showed evidence of micro-cracks in the surface of the materials exposed to hygrothermal degradation. The degradation of the mechanical properties of the composites induced by the water was expected to be reinforced in the presence of a cyclic sorption and desorption process [16]. The type of environment substantially determined the amount of water uptake and its impact on the characteristics of the composites. Akil et al. [17] examined the aging degree of UP/glass composites immersed in three different types of water: distilled water, seawater, and acidic solutions. The composites immersed in the distilled water appeared to be the most impacted by hygrothermal effects.

The impact of moisture absorption on mechanical properties of composite materials has attracted attention in previous studies [18-21]. Moisture effects are strongly marked in interlaminer shear and flexural properties of FRP composites compared to their tensile properties. The low sensitivity of tensile properties to moisture effects can be attributed to the fact that tensile properties are dominated by the material's fibers. On the other hand, glass and carbon fiber reinforcement are insensitive to moisture effects. In these materials, only the fiber-matrix interface constituted a breeding ground for moisture ingression [22, 23].

The aim of this work was to model the moisture absorption kinetics in composites before and after hygrothermal damage. The numerical results were compared with published experimental data [6, 24].

The remainder of this paper consists of several sections.

The next section describes the numerical simulations that were conducted to estimate hygrothermal stresses in a composite plate; while in the subsequent section, a proposed damage model was used to calculate the moisture diffusion in composites before and after hygrothermal damage.

2. Model of Absorption

To predict the amount of moisture absorbed in a composite material prior to the occurrence of damage, Fick's law remains the most widely used because of its simplicity and its ability to predict the humidity diffusion in several types of composite materials.

Let e be the thickness of a plate exposed on its two sides to the same humid environment. Let us consider, moreover, that the plate is infinite in the y and z directions, and the variation of the moisture concentration depends on only the x direction (see Figure 1). Under these conditions, the problem is therefore one-dimensional, and the initial concentration is expected to be uniform throughout the thickness of the material. The moisture concentration within a plate suddenly exposed on its two sides to a constant moisture concentration is accordingly described by the following Fick's equation [9]:

$$\frac{\partial C}{\partial t} = D(\theta)\frac{\partial^2 C}{\partial x^2} \qquad (1)$$

where C denotes the moisture concentration and D is the diffusion coefficient of moisture, depending on the temperature.

However, under certain exposure conditions, the composite materials suffer from degradation due to the deterioration of the quality of the interface fiber/matrix. Figure 2 summarizes the diverse kinetics of moisture absorption. If during the moisture diffusion no damage occurred, then Fick's model is suitable for such behavior (curve *0*).

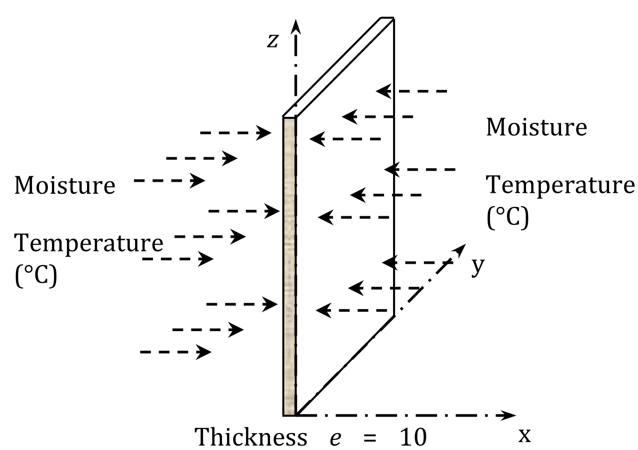

Figure 1. Definition of a plate sample exposed to moisture and temperature.

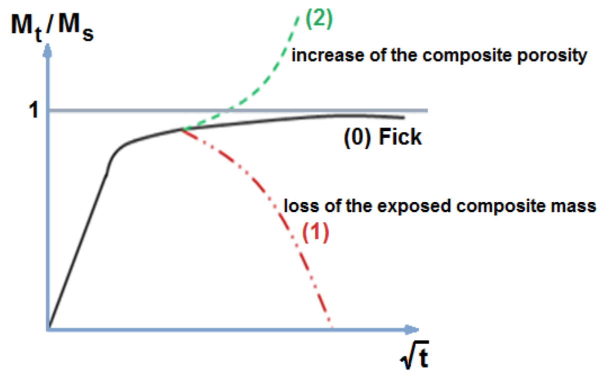

Figure 2. Various kinetic absorptions of moisture into a composite material.

Otherwise, hygrothermal damage occurs with a kinetic absorption in agreement with the following:

- Law 1: loss of the exposed composite mass;
- Law 2: increase of the composite porosity.

To predict these different absorption behaviors, the moisture solubility in the composite depends upon the exposure time. In the case of law 1, the solubility decreased with time and is expressed by [25]

$$S = S_0 \exp\left(-k\frac{\langle t^* - t_s^* \rangle}{t_s^*}\right) \tag{2}$$

where < > are the so-called Macauley brackets, $\langle x \rangle = \frac{1}{2}(x + |x|)$, $t^* = \sqrt{t}/e$, $t_s^* = \sqrt{t_s}/e$, where e stands for the thickness of the sample; k is a material parameter to be calibrated; S_0 denotes the solubility of the moisture in the non-damaged composite, and t_s is the saturation time, at which point damage initiates (onset of damage). We suppose that t_s is the time of exposure corresponding to a concentration reaching 99% of the initial saturation of solubility S_0. For law 2, the solubility increases with time as assumed to be

$$S = S_0 \exp\left(k\frac{\langle t^* - t_s^* \rangle}{t_s^*}\right) \tag{3}$$

3. Transient Hygrothermal Stresses

Using the principle of superposition, the stress state, which is induced by temperature and moisture (hygrothermal stresses), can be expressed by [8]

$$\sigma_{ij} = -L_{ijkl}\varepsilon_{ij} \tag{4}$$

with

$$\varepsilon_{ij} = -\left(\alpha\left(\bar{T} - T\right) + \eta\left(\bar{C} - C\right)\right)\delta_{ij} \tag{5}$$

where L_{ijkl} is the fourth-order isotropic elastic moduli tensor; σ_{ij} is the second-order stress tensor; α_{kl} is the second-order thermal expansion tensor; η_{kl} is the moisture expansion tensor, and the average values of T and C are respectively denoted as $\bar{T}$ and $\bar{C}$. It is clear from equation (4) that the highest value of stress occurred in the transient stage, in which the composite material was subjected to significant gradients of temperature and moisture. These factors can lead to the onset of damage, described by microscopic cracks in the composite material. In the case of an isotropic material, we have $\alpha_{kl} = \alpha\delta_{kl}$ and $\eta_{kl} = \eta\delta_{kl}$

where δ_{kl} is the Kronecker symbol; α is the isotropic coefficient of thermal expansion, and η is the isotropic coefficient of moisture expansion. Therefore, equation (4) is rewritten as [8]

$$\sigma_{ij} = \frac{E}{1+\nu}\left(\varepsilon_{ij} + \frac{\nu}{1-2\nu}\varepsilon_{kk}\delta_{ij}\right) \tag{6}$$

where E is Young's modulus, and ν is the Poisson's ratio.

Equation (6) shows that hygrothermal stresses in the plate were reduced to normal components. With reference to the (x, y, z) coordinate system (Figure 1) and in the case of no external stresses applied to the plate, non-zero stress components are thus given by [8]

$$\sigma_y = \sigma_z = \frac{E}{1-\nu}\left(\alpha\left(\bar{T} - T\right) + \eta\left(\bar{C} - C\right)\right) \tag{7}$$

We suppose that the mechanical parameters are not dependent on both moisture uptake and the range of temperatures. The parameter η, also called the swelling isotropic coefficient, is measured by the relative variation ΔV of the sample volume. In the case of free swelling [25], it is calculated by

$$\eta = \frac{\Delta V}{3V\bar{C}} \tag{8}$$

It is of importance to notice that such a swelling law (8) is observed in many composites with values of η varying from 0.2-0.5 [7, 25].

4. Numerical Simulations

The finite element software program ABAQUS FE 6.10 was designed for general use in both linear and nonlinear structural problems, such as the modeling of a diffusion process. As a transient phenomenon, the process of moisture diffusion refers to the distribution of the moisture concentration throughout a material's profile in proportion to the exposure time. Therefore, the analysis of the transient field constitutes the most appropriate form of analysis for finite element diffusion.

The ABAQUS FE 6.10 software was used for the analysis of moisture diffusion. The FE model was performed with the same dimensions as that of the samples (see Figure 1). Moisture diffusion coefficients obtained from tests (see Table 1) were incorporated as input parameters for the properties of the materials in the FE model. The steps of time integration were controlled automatically. The moisture concentration of the surrounding surfaces' FE models were set to be completely saturated, and these concentration levels were used as the boundary conditions to enable the kinetic of diffusion processes.

The finite elements code within Abaqus was used to simulate the diffusion of moisture in the composite plate of Figure 3, before and after hygrothermal damage. The two damage models given by equations (2) and (3), along with the stresses given by equation (7), were developed and executed by FORTRAN software.

In the beginning, we considered the plate to be in contact with distilled water with dissymmetrical temperatures applied to each face as shown in Figure 4. This test was introduced to verify the effectiveness of the moisture absorption simulation and to estimate the induced hygrothermal stress level. The material used was a polyester/glass fiber composite, where the hygrothermal parameters were obtained by experiments [24] and are summarized in Table 1.

Figure 4 shows the distribution of temperature and moisture concentrations throughout the thickness in a steady state (t = 5000 s). The absorbed amount of moisture increased with an increased temperature. This phenomenon can be explained by the fact that the diffusion coefficient D also increases with an increased temperature.

Figure 5 gives the stress evolution in the different plate layers identified by coordinate x. Clearly, the highest value of the tensile stress, obtained at the earliest stage of absorption, reached a significant value that can cause damage over a longer period of time. The evolution of a moisture concentration average throughout the thickness was predicted and is provided in Figure 6.

Table 1. Moisture diffusion coefficients using distilled water [24]

Temperature θ (°C)	Diffusion D (10^{-3}h/mm^2)	Solubility S_0 (%)
25	5.4	0.26
45	11.7	0.37
65	13.5	0.49
85	16.2	0.57

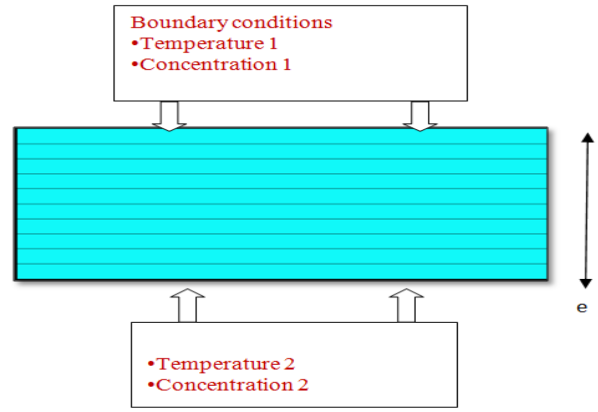

Figure 3. Parameters involved in the diffusion calculation, for a 1-D diffusion.

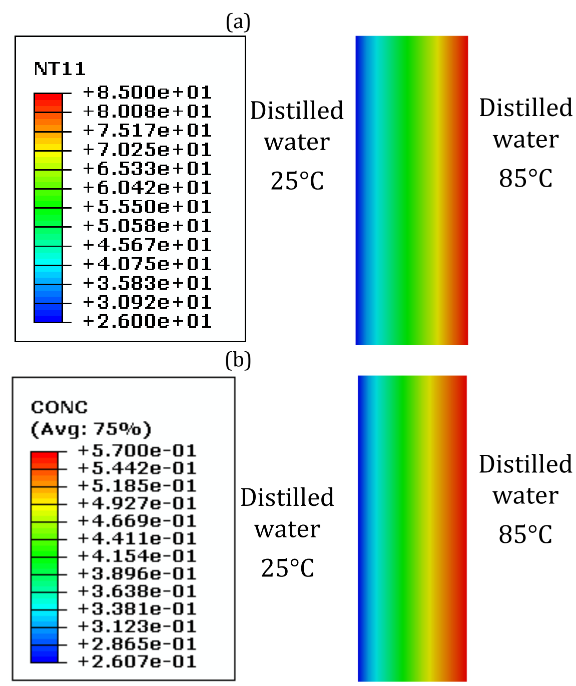

Figure 4. Distribution of temperature (a) and moisture concentration (b) within the plate thickness.

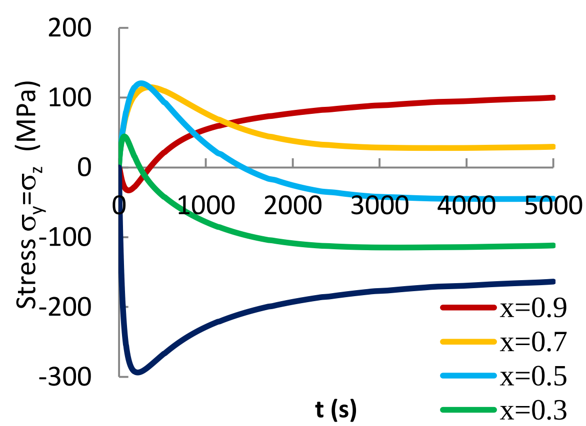

Figure 5. Stress evolution in thickness coordinate x (E =4 GPa, η = 0.2) versus exposure time.

It can be seen that the numerical results are in good agreement with the experiments. It is noticeable, however, that the distilled water does not cause damage to the composite, even with a long exposure time (t = 5600 hours). Next, a plate was immersed in sea water at a temperature of 65°C. The moisture diffusion parameters are given in Table 2.

Table 2. Moisture diffusion coefficients in sea water (θ = 65 °C) [24]

Diffusion D (10^{-6} s/mm^2)	Solubility S_0 (%)	Parameter k	Time of saturation t_s (h)
2.6	0.37	0.7	3802

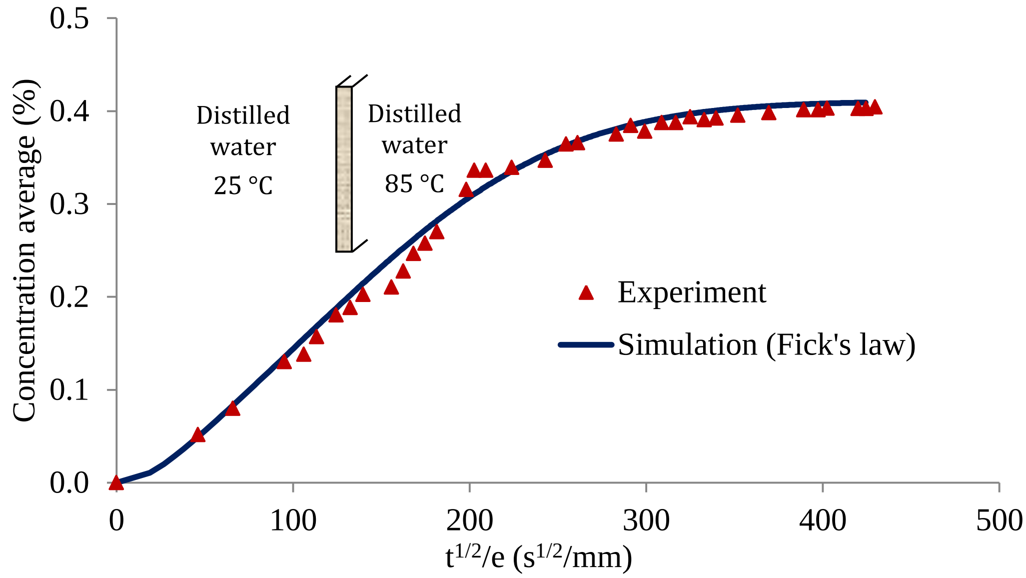

Figure 6. Evolution of the moisture concentration average $\bar{C}$ (%) for the composite immersed in distilled water.

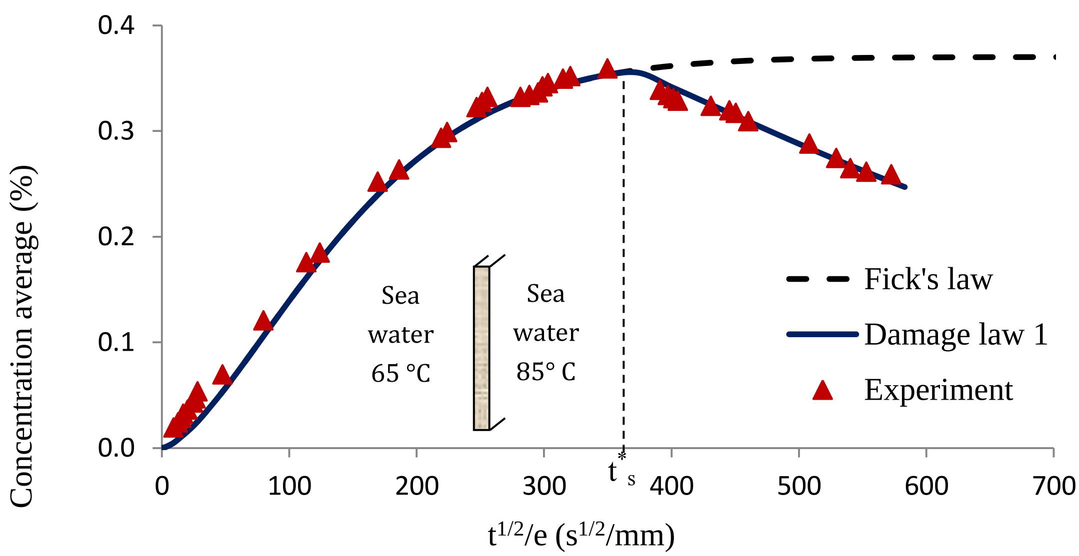

Figure 7. Evolution of the moisture concentration average $\bar{C}$ (%) for the composite immersed in sea water.

For the numerical simulation, damage according to law 1 provided by equation (2) was used. Figure 7 reveals the moisture concentration average evolution for the plate immersed in sea water, obtained by both the experiment and the simulation. This figure clearly shows that there was a loss of composite mass, beginning with the saturation time t_s. It can be seen that a good agreement between the two results were obtained, in particular within the damage part of the plot.

Next, we consider a polyurethane material [6], whose hygrothermal parameters are summarized in Table 3. This material has a high coefficient of diffusion, leading quickly to hygrothermal damage, according to law 2 provided by equation (3). Figure 8 gives the numerical results of the moisture concentration average evolution according to exposure time. It can be seen that the proposed model gives results that are in good agreement with experiments, both before and during damage.

Table 3. Polyurethane absorption parameters at temperature θ = 40 °C [6]

D (10-6 s/mm^2)	S_0 (%)	k	t_s (h)
21.56	2.64	0.1	4

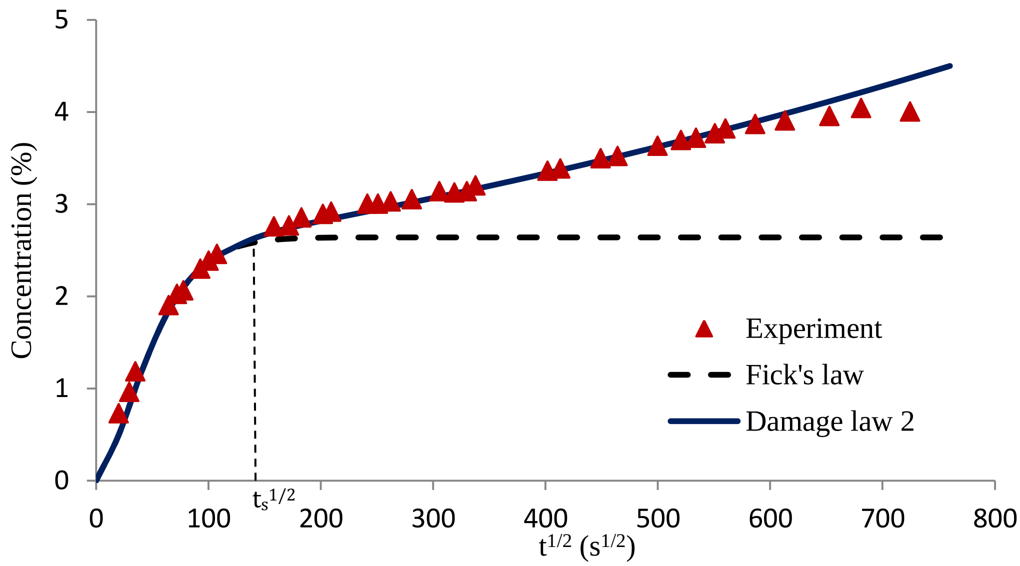

Figure 8. Evolution of the moisture concentration (%) for the polyurethane composite.

5. Discussion

In this work, we can predict that the hygroscopic constraint was spread thanks to either temperature and/or humidity. These factors led to the propagation of damage to the composite materials. Figure 5 illustrated the distribution of these constraints, along the duration of time and within each layer of the composite plate. This figure indicates, in addition, that during the absorption of humidity, the constraints of traction reached a maximum level within the core of the plate. Consequently, these constraints may be the origin of micro-cracks, which were observed in the core of the material and were oriented perpendicular to the surface of the plate ($\sigma_{yy}(x,t) = \sigma_{zz}(x,t)$). Figure 5 also demonstrates that the compression constraints were drawn throughout time until they become stabilized. This reduction was generated by an absence of cohesion in the interface fibers/matrix.

To simulate the diffusion of humidity within the composites before and after damage, a model coupling the diffusion of humidity and mechanics was elaborated.

Figure 6 shows the evolution in the diffusion of humidity in the composite polyester/glass fiber immersed in distilled water at temperature of 25 °C on one face and 85 °C on the other. According to Figure 6, a Fickian behavior can be observed, which, in turn, characterized an evolution of absorption without damage. When this plate was immersed in sea water at a temperature of 65 °C (Figure 7), another type of damage was found; this figure retraces a loss in the weight of the material after a certain duration of time.

In this case, chemical groupings may extract polymer chains and be evacuated in the solvent, explaining the loss of material and the dropping of the global mass, despite the absorption of water.

In Figure 8, where the polyurethane was exposed to distilled water at a temperature T of 40 °C, an increase occurred in the quantity of humidity absorbed (damage type 2). This increase corresponded with a rapid acceleration of water absorption, which was generally accompanied by a significant deformation and damage within the material.

Overall, the simulation results confirmed the experimental results, which were also shown in recent studies by Jiang Xu et al. [6] and Ben Daly et al. [24].

6. Conclusion

In this work, the diffusion of moisture within various composite materials was simulated. Hygrothermal stresses were predicted, and the stress levels were high enough to initiate damage in the composites. Two damage models were proposed and implemented in Abaqus. The obtained numerical results, using the proposed damage laws, fit very closely with the experimental data for the two types of damage. In future research, we intend to develop a hygromecanical model coupled with damage, in which the evolution of the mechanical proprieties will be considered during moisture absorption.

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