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Experimental and Theoretical Study on Hygrothermal Aging Effect on Mechanical Behavior of Fiber Reinforced Plastic Laminates

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***Abstract*—**The manufacture of composite parts is a major issue in many industrial domains. Polymer composite materials are ideal for structural applications where high strength-to-weight and stiffness-to-weight ratios are required. However, exposition to extreme environment conditions (temperature, humidity) affects mechanical properties of organic composite materials and lead to an undesirable degradation. Aging mechanisms in organic matrix are very diverse and vary according to the polymer and the aging conditions such as temperature, humidity etc. This paper studies the hygrothermal aging effect on the mechanical properties of fiber reinforced plastics laminates at 40 °C in different environment exposure. Two composite materials are used to conduct the study (carbon fiber/epoxy and glass fiber/vinyl ester with two stratifications for both the materials [904/04] and [454/04]). The experimental procedure includes a mechanical characterization of the materials in a virgin state and exposition of specimens to two environments (seawater and demineralized water). Absorption kinetics for the two materials and both the stratifications are determined. Three-point bending test is performed on the aged materials in order to determine the hygrothermal effect on the mechanical properties of the materials.

***Keywords*—**FRP laminates, hygrothermal aging, mechanical properties, theory of laminates.

I. Introduction

COMPOSITE materials are increasingly used in many industrial sectors: automotive, aeronautics, aerospace, construction industry ... in view of their advantages (elaboration depending on application, incorporation of fillers and additives for resistance to a specific environment ...). However, these materials have limitations on their resistance to extreme conditions, we can cite as an example the structures or mechanisms working at high temperatures, offshore structures exposed to the marine environment that is known to be harmful for composite materials organic matrix.

In this work, particular attention is paid to the effect of hygrothermal aging on the mechanical properties of composite carbon/epoxy and glass fiber E(GFE)/vinyl ester, which are among the most commonly used materials in their category. An experimental study was conducted to evaluate the tolerance of the materials to absorb the solvent in the two exposure environments as well as the effect of aging on the mechanical properties by means of three point bending tests. The laminate theory is used to determine the effect of aging on the different layers constituting the materials studied.

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II. Theoretical Aspect

The composite materials aging mechanisms are diverse and vary according to the polymer matrix of the aging conditions, such as temperature, relative humidity, but also of the reference state (residual stresses, stress concentrations). Natural aging is often too slow this is why we often opt for accelerated aging in terms of simulations and after resorting to feedback to best correlate natural aging.

The simplest model for the diffusion of a solvent is the model of Fick [1]. Its first law is given by (1).

$$\bar{\varphi} = -D\overrightarrow{grad}C \tag{1}$$

The second Fick's law is time dependent and is illustrated by (3); the latter is obtained by combining (1) and (2):

$$\frac{\partial c}{\partial t} = div(\overrightarrow{\varphi}) \tag{2}$$

$$\frac{\partial c}{\partial t} = div(-D\overrightarrow{grad}\,C \tag{3}$$

when the diffusion is unidirectional and that D is independent of concentration, we have:

$$\vec{\varphi} = -D\frac{\partial c}{\partial x} \tag{4}$$

$$\frac{\partial y}{\partial x} = D\frac{\partial^2 y}{\partial x^2} \tag{5}$$

The effect of hygrothermal aging on the mechanical properties of polymers and composites organic matrices is the subject of several research studies [1]-[10]. Some authors evaluated the ability of the composite to be rehabilitated after hygrothermal aging through drying [11]. At high temperatures, a different absorption kinetics than the Fick model can be observed [12] as the absorption of the solvent can be considered a thermally activated process [13]. The solvent absorption and its effect on the mechanical properties of the composite material differs de-pending on the orientation of the reinforcement [14].

III. Experiments and Results

This study concerns two composite materials with two different laminations for each material. The specimens are cut from plates and their dimensions are consistent with the ISO standard 14125. The aging conditions (exposure environment, temperature) are described in [15]. Weight gain tests are conducted over a period of 3000 hours which is considered a relatively long period to assess the irreversible degradation that can undergo these materials and possibly identify the causes. Kinetics absorptions at 40 °C are given in Figs. 1 and 2 for carbon/epoxy material, and in Figs. 3 and 4 for the GFE/vinyl ester material for both stratifications and two exposure environments, which give four curves for each material.

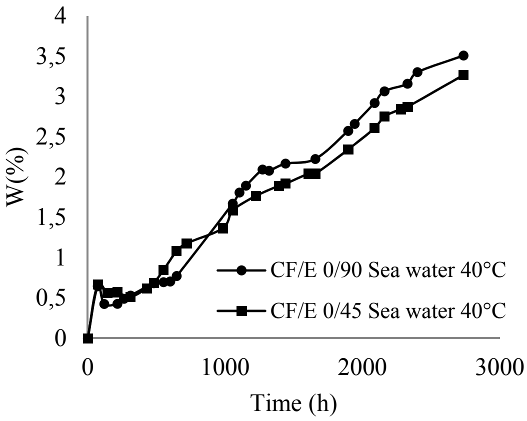

Fig. 1 Kinetic Absorption of Carbone fiber/Epoxy in seawater at 40°C

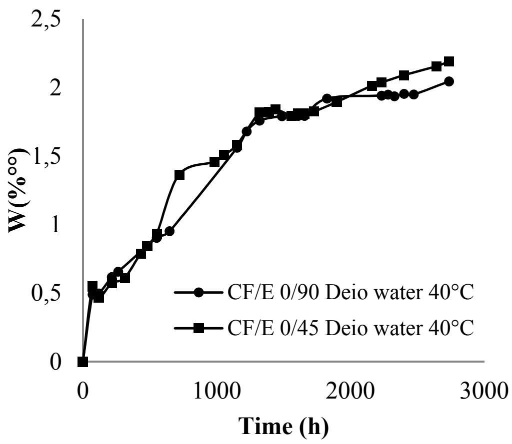

Fig. 2 Kinetic Absorption of Carbone fiber/Epoxy in deionized at 40°C

In this study, the sea water absorption is higher than that in deionized water, in [16], [17], the authors warrant it by a significant pH of the sea water causing the degradation of the matrix (formation of micro cracks) that fill in the solvent away, hence the absorption continuously. In terms of the effect of hygrothermal aging on the mechanical properties of materials, it is illustrated by the Table I.

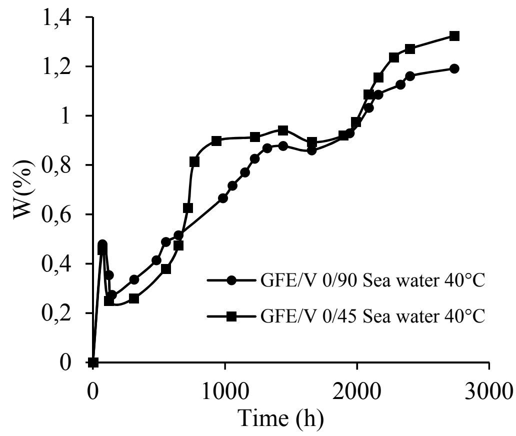

Fig. 3 Kinetic Absorption of GFE/Vinyl ester in seawater at 40 °C

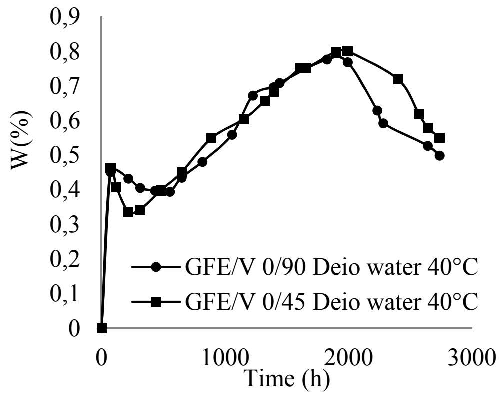

Fig. 4 Kinetic Absorption of GFE /Vinyl ester in deionized water at 40 °C

TABLE I
Effect of Aging on Mechanical Properties of Materials

Mat.	Arch.	Environment	E_f(MPa)[a]	σ_r (MPa)[b]	ε_r (MPa)[c]
GFE/Vinyl ester	[$90_4/0_4$]	Deionized Water	-39.4%	-42.37%	+11.84%
		Sea Water	-49.94%	-47.82%	-10.97%
	[$45_4/0_4$]	Deionized Water	-41.22%	-25.32%	+12.40%
		Sea Water	-15.62%	--2.68%	-11.97%
CF/Epoxy	[$90_4/0_4$]	Deionized Water	-26.3%	-52.48%	-17.00%
		Sea Water	-38.42%	-49.83%	-23.93%
	[$45_4/0_4$]	Deionized Water	-25.76%	-41.46%	-12.98%
		Sea Water	-16.65%	-43.36%	-31.18%

Mat.: Material of the specimen, Arch.: Architecture of composite material, GFE: Glass fiber E, CF: Carbone fiber, [a] Flexion modulus, [b] Stress at rupture, [c] Strain at rupture.

As reported by most studies [4]-[10], we find here that the hygrothermal aging a negative affect the behavior of reinforced plastics, this is illustrated in particular by a loss of rigidity and resistance to breakage. In some cases, there is a ductility of gain with a larger strain at break for an aged material, this may be due to the swelling of the matrix because it has a greater elongation at break.

IV. Effect of Aging on the Laminated Layers

The usefulness of this part of the investigation is to evaluate the effect of hygrothermal aging on the various layers constituting materials of the study. The stiffness of a laminate of matrix is composed of three sub matrices made as follows:

$$A_{ij} = [A] = \sum_{k=1}^{n} (h_k - h_{k-1})\, E'_k \qquad (6)$$

$$D_{ij} = [D] = \frac{1}{3}\sum_{k=1}^{n} ({h^3}_k - {h^3}_{k-1})\, E'_k \qquad (7)$$

$$B_{ij} = [B] = \sum_{k=0}^{n} \frac{1}{2} ({h^2}_k - {h^2}_{k-1})\, E'_k \qquad (8)$$

where h is the thickness of the respective bend, is the stiffness matrix of the respective bend, [A] is the membrane stiffness matrix, [B] is the membrane bending-twisting coupling matrix, and [D] is the matrix bending stiffness. The stiffness matrix in its entirety is written as follows:

$$[ABD] = \begin{bmatrix} A_{11} & A_{11} & A_{16} & B_{11} & B_{11} & B_{16} \\ A_{12} & A_{22} & A_{26} & B_{12} & B_{22} & B_{26} \\ A_{16} & A_{26} & A_{66} & B_{16} & B_{26} & B_{66} \\ B_{11} & B_{11} & B_{16} & D_{11} & D_{11} & D_{16} \\ B_{12} & B_{22} & B_{26} & D_{12} & D_{22} & D_{26} \\ B_{16} & B_{26} & B_{66} & D_{16} & D_{26} & D_{66} \end{bmatrix} \qquad (9)$$

Before calculating the laminate stiffness matrix as a whole, it is necessary to evaluate the stiffness matrixes of the folds; this is done using the stiffness matrix of the ply oriented at zero degree through the transition matrix, and the relationship is written as follows:

$$\begin{Bmatrix} Q_{11} \\ Q_{22} \\ Q_{12} \\ Q_{66} \\ Q_{16} \\ Q_{26} \end{Bmatrix} = \begin{bmatrix} C^4 & S^4 & 2C^2S^2 & 4C^2S^2 \\ S^4 & C^4 & 2C^2S^2 & 4C^2S^2 \\ C^2S^2 & C^2S^2 & C^4+S^4 & -4C^2S^2 \\ C^2S^2 & C^2S^2 & -2C^2S^2 & (C^2-S^2)^2 \\ C^3S & -CS^3 & CS^3-C^3S & 2CS^3-2C^3S \\ CS^3 & -C^3S & C^3S-CS^3 & 2C^3S-2CS^3 \end{bmatrix} \begin{Bmatrix} Q_{xx} \\ Q_{yy} \\ Q_{xy} \\ Q_{ss} \end{Bmatrix} \qquad (10)$$

where C= Cos θ and S= Sin θ. The indices 1, 2, 6 denote the ply oriented at an angle different from zero degree. The indices x, y, s denote stiffness matrix oriented at an angle θ different from zero degree.

The mentioned procedure is followed to assess the resistance of laminated layers before and after aging. The calculation was done for all the materials of the study using experimental test data. A script is produced via the MATLAB software for the calculation to be done automatic changing only the data of the constituents of the material in question as well as the orientation of the reinforcing manner and the volume fractions of the components. Additional data are considered, it is the thickness of the layers, in this case, this thickness is considered to be the same. The reference pattern is given by the calculation, Fig 5.

After execution of the steps mentioned previously separately for each sequence, we have led to the evaluation of the resistance of creases in different laminates in a virgin state and the reductions in% after exposure to both considered media, results are given in Table II for the carbon / epoxy and Table III for fiber glass / vinyl ester.

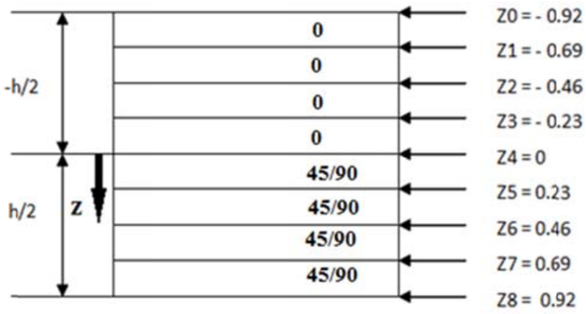

Fig. 5 Schematic architecture of the used materials

TABLE II
Aging Effect on Carbone/Epoxy Layers Resistance at 40 °C

Mat	Arch	Stress (MPa)	Virgin (MPa)	D.W.D (%)	S.W.D (%)
		σ^0_{XX}	116.76	-44.62%	-55.80%
	$[90_4/0_4]$	σ^0_{YY}	4.43	-44.65%	-55.82%
		σ^0_{XY}	0	0	0
		σ^0_{XX}	18.028	-44.62%	-55.80%
	$[90_4/0_4]$	σ^0_{YY}	-4.27	-44.61%	-56.27%
		σ^0_{XY}	0	0	0
CF/Epoxy		σ^0_{XX}	164.94	-44.73%	-65.55%
	$[45_4/0_4]$	σ^0_{YY}	-2.91	-44.77%	-65.43%
		σ^0_{XY}	-3.574	-44.71%	--65.43%
		σ^0_{XX}	30.636	-44.73%	-65.56%
	$[45_4/0_4]$	σ^0_{YY}	2.91	-44.73%	-65.53%
		σ^0_{XY}	3.574	-44.71%	-65.53%

TABLE III
Aging Effect on GFE/Vinyl Ester Layers Resistance at 40 °C

Mat	Arch	Stress (MPa)	Virgin (MPa)	D.W.D (%)	S.W.D (%)
		σ^0_{XX}	45.582	-32.31%	-32.02%
	$[90_4/0_4]$	σ^0_{YY}	1.585	-32.30%	-31.98%
		σ^0_{XY}	0	0	0
		σ^0_{XX}	8.468	-32.31%	-32.01%
	$[90_4/0_4]$	σ^0_{YY}	-1.585	-32.30%	-31.98%
		σ^0_{XY}	0	0	0
GFE/Vinyl ester		σ^0_{XX}	49.84	-31.14%	-8.08%
	$[45_4/0_4]$	σ^0_{YY}	-1.167	-31.11%	-8.05%
		σ^0_{XY}	-1.268	-31.15%	-8.04%
		σ^0_{XX}	10.77	-31.14%	-8.09%
	$[45_4/0_4]$	σ^0_{YY}	11.67	-31.11%	-8.05%
		σ^0_{XY}	1.268	-31.15%	-8.04%

By superposing the results obtained, we can see the reductions in the strength of the laminated layers, for exposed materials in seawater and deionized water connection with virgin materials. Some information has been confirmed as the best strength of materials $[45_4/0_4]$ relative to $[90_4/0_4]$ as was the case during experimental testing. We also deduce that the composite carbon/epoxy been the most affected by sea water as distilled water in contrast to composite GFE/vinyl ester in which the layers have known a greater reduction of resistance in distilled water. We can conclude that the effect of hygrothermal aging differs depending on the orientation of the reinforcement of the material and the exposure environment.

V. Conclusion

The purpose of this study was to determine the effect of hygrothermal aging on the mechanical properties of carbon fiber/epoxy and GFE/vinyl ester with two different stratifications for each material. We can conclude:

1- The kinetics of absorption is not following the Fick's model in the beginning of process, and this is due to irreversible degradation of the material.
2- The hygrothermal aging tends to adversely affect the behavior of laminates. This is illustrated by the deterioration of mechanical properties in a major way.
3- The effect of hygrothermal aging is different depending on the orientation of the reinforcement in the ply; this is due to the barrier effect that may carry the fibers according to their orientation

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