ULTRASONIC STUDY OF THE ANISOTROPIC DAMAGE OF UNIDIRECTIONAL GLASS-EPOXY COMPOSITES DURING HYGROTHERMAL AGEING

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SUMMARY: The purpose of this work is to study using ultrasounds the effects of hygrothermal ageing on the elastic behaviour of an unidirectional glass-fibre reinforced epoxy matrix. Simulated ageing conditions are performed. The degradation processes resulting from 25 days ageing in distilled water at 70°C are considered. The damage is assessed quantitatively through the loss of stiffness in terms of elastic constants. In the framework of the thermodynamic of irreversible processes, a constitutive equation including a tensorial variable describing the damage phenomenon is used. Through the variation of the damage parameters, damage and its anisotropy are quantitatively assessed. This nondestructive characterization is conducted together with a SEM microstructural analysis to correlate ultrasonic macroscopic measurements to microscopic observations

KEYWORDS: elastic properties, non-destructive evaluation, ultrasonics, ageing, damage, anisotropy, interface, polymer composites.

INTRODUCTION

Composite materials can in service be subject to a wide variety of different loading conditions. The most important conditions are mechanical stress and environmental effects. Before to use composite material in structural components, their behaviour under various conditions has to be studied. Concerning glass-fibre reinforced polymers, which are used in structural parts of boats, off-shore equipment and in the nuclear industry as water piping systems, the main environmental attacks are related to effects of ageing at elevated temperatures and humidities. The purpose of this paper is to study the influence of the hygrothermal ageing on mechanical properties of a glass fibre polymer composite. Several investigations in the open literature has shown that hygrothermal ageing influences the properties of reinforced polymers, but such studies only looks at some basic mechanical properties as the in plane Young's modulus [1][2]. The influence of hygrothermal exposure on out of plane Young's moduli and shear modulus as well as Poisson's ratios and the anisotropy of the damage have been less studied. In most of experimental studies, measurements of elastic properties were performed using classical static techniques based on loading and extensometry [3]. These means require a large number of samples with suitable orientations. All needed sample cuts are often not possible from the manufacturing process (thin sheets) and a full evaluation cannot be achieved this way. However, these destructive measurements, using many samples, are therefore mostly not available when the purpose is to follow the changing of materials properties before, during and after damage.

An alternative method to overcome these difficulties is the ultrasonic method. Indeed, ultrasonic evaluation is well adapted to this problem, especially when the immersion technique is used.

In this work, the effect of hygrothermal ageing on the mechanical behaviour of an unidirectional glass fiber epoxy matrix composite is studied by the means of ultrasonic evaluation. The evolution of the elastic behaviour during hygrothermal ageing in water at 70°C is considered and the resulting anisotropic damage is studied. This nondestructive characterisation is conducted together with a SEM microstructural analysis to correlate ultrasonic macroscopic measurements to microscopic observations .

MATERIAL AND AGEING CONDITIONS

The material under investigation is a composite with an epoxy resin reinforced with E-glass fibres. The matrix was obtained from diglycidyl ether of bisphenol A (DGEBA) resin which had been cured by using a dicyan diamide (DDA) hardener. Unidirectional sheet composite (2mm thickness) were fabricated. The cure cycle was 2 h at 125 °C under 3 bars of pressure. The volume fraction of fibre was 68%. The sample (5×5 cm) was placed in a container of distilled water at 70 °C and it was removed at 10, 20 and 25 days for velocity measurements. Before each measurement, the sample was dried in an oven at 60 °C

ULTRASONIC EVALUATION

Theoretical aspects

The ultrasonic evaluation of elastic properties of materials is based on the relationship between the elastic constants of material and the phase velocities of ultrasonic waves. Considering progressive plane waves, the resolution of the wave propagation equation, for a homogeneous elastic and anisotropic media, gives the so-called Christoffel equation [4]:

$$\left| \Gamma_{il} - \rho V^2 \delta_{il} \right| = 0 \tag{1}$$

where $\Gamma_{il} = C_{ijkl} n_j n_k$ is the Christoffel tensor, ρ is the mass density, V is the phase velocity and δ_{il} the Kronecker symbol. C_{ijkl} is the fourth rank stiffness tensor and $\vec{n}$ is the unit vector normal to the planes of wave. Instead of the C_{ijkl} tensor, it is convenient to use the C_{IJ} matrix using the usual contracted notation [4]. The arrangement of material constituents i.e. fibres or particles in composites, determine the structural symmetry at the macroscopic scale. Orthotropic symmetry, which is sufficiently general to describe most of composites, have nine independent elastic constants [4]. The C_{IJ} tensor expressed in the principal axes 1, 2, 3 is showed in figure 1.

$$C_{IJ} = \begin{pmatrix} C_{11} & C_{12} & C_{13} & 0 & 0 & 0 \\ & C_{22} & C_{23} & 0 & 0 & 0 \\ & & C_{33} & 0 & 0 & 0 \\ & & & C_{44} & 0 & 0 \\ & Sym. & & & C_{55} & 0 \\ & & & & & C_{66} \end{pmatrix}$$

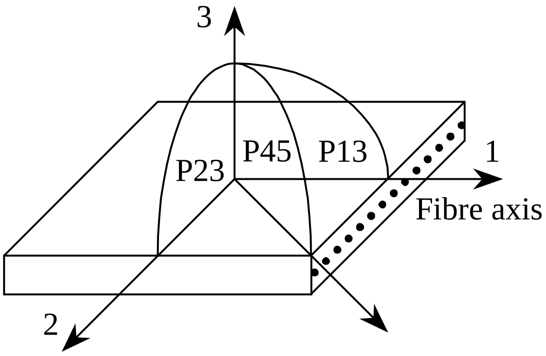

Fig. 1: Stiffness matrix in orthotropic symmetry, reference axes and planes of propagation

Elastic constants determination : the inverse problem resolution

The elastic constants must be recovered from measured ultrasonic velocities. This is typically an inverse problem. The approach we use here is based on measurement of ultrasonic velocities in several accessible planes of propagation. So, there are more velocity measurements than independent elastic constants to determine. To solve this inverse problem, the most efficient method is an optimization process using a non-linear numerical algorithm. In practice, elastic constants determination is performed by minimizing the sum of the squares of the deviations between experimental and theoretical velocities. The minimization algorithm we use is the Levenberg-Marquardt one. The optimisation process is summarised in figure 2 [5, 6].

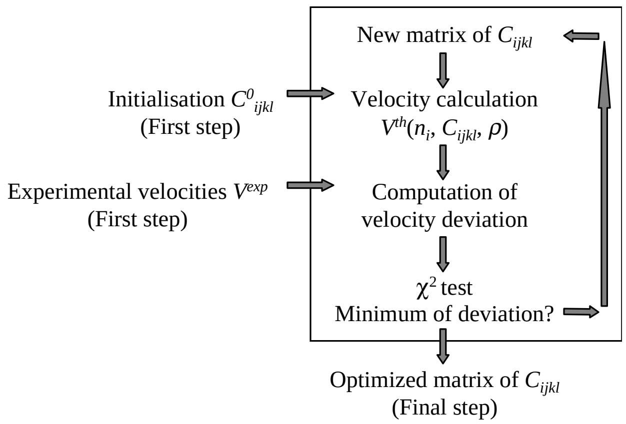

Fig. 2: Optimization process for elastic constants determination

Experimental device

Velocity measurements are obtained by using an immersion technique in simple through transmission assembly. The sample, a plate of uniform thickness with parallel faces, is placed between two transducers fixed on two goniometers. In this work, the central frequency for the transducers is 2.25 MHz and the coupling is water. Variation of the angle of incidence provides several directions and modes of propagation in the material by mode conversion at liquid-solid interface according to the Snell-Descartes's law. The transit time of ultrasonic pulses inside the sample are computed by a cross-correlation function between a reference signal corresponding to the propagation in the water without the sample and a measured signals with the sample at several oblique incidences. The phase velocities *V* in the material is deduced from the transit time and simple geometric considerations. The whole device, immersion tank and data acquisition, is presented in figure 3. Note that rotations and signal acquisition are computer assisted [6].

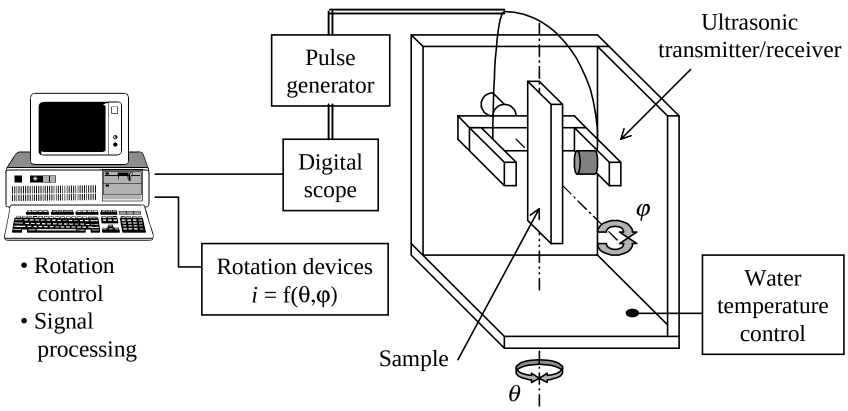

Fig. 3: Experimental device for immersion ultrasonic velocity measurements

RESULTS

Initial state characterization

Velocity measurements have been performed in different planes varying the angle of incidence. We have considered the propagation in plane P23 perpendicular to the fiber direction (axis 1), in plane P13 containing the fiber direction and in P45 as showed in figure 1. From these velocity measurements, the elastic constants of the composite are determined by using the previously described optimization process. Figure 4 represents the velocity fits for longitudinal (resp. shear) waves in two principal planes P23, P13 and P45 (resp. in the planes P23 and P13). The discrete points represents the experimental velocities and the solid curves are the velocities which are computed from the optimized elastic constants.

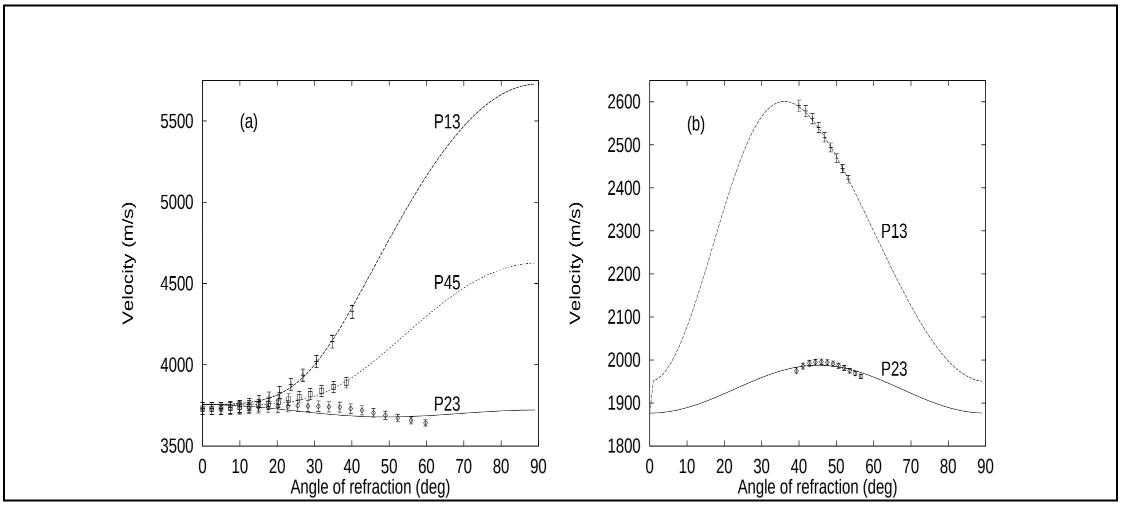

Fig. 4: Ultrasonic velocity versus angle of refraction : (a) QL and (b) QS waves

We can notice the very good fit between experimental and computed velocities. For plane P23, perpendicular to the fiber axis, the ultrasonic measured velocities are practically independent on the direction of propagation. For the other considered planes of propagation P45 and P13 the velocity increase with the direction of propagation. The greater angular dependence is for plane P13. This ultrasonic behaviour agrees with the structure of the material with axis 1 (fibre axis) as symmetry axis. Indeed, the velocity variation is about 600 m/s for a 40° refraction angle. By considering these three planes, seven of the ninths elastic constants were determined as shown in figure 5. C_{12} and C_{66} depends strongly on the shear velocities in the P45 plane. Because of the weak thickness of the sample, the two quasi-shear modes denoted QS1 and QS2 are difficult to identify in this plane. So, the determination of C_{12} and C_{66} could not be achieved with a good accuracy and these two elastic constants are not considered in this work The identified elastic constants with the corresponding uncertainties are reported below :

$$C_{IJ\,(GPa)} = \begin{pmatrix} 71.5 \pm 1.4 & C_{12} & 15.4 \pm 0.2 & 0 & 0 & 0 \\ & 30.2 \pm 0.4 & 13.2 \pm 0.2 & 0 & 0 & 0 \\ & & 30.7 \pm 0.3 & 0 & 0 & 0 \\ & & & 7.7 \pm 0.2 & 0 & 0 \\ & \mathit{Sym.} & & & 8.3 \pm 0.3 & 0 \\ & & & & & C_{66} \end{pmatrix}$$

Fig. 5: Recovered elastic constants from ultrasonic velocities

Damage characterization

First, the initial elastic anisotropy of the composite and its variation with hygrothermal ageing is studied in terms of the angular dependence of velocity for longitudinal and shear waves. In each plane, the ultrasonic velocities versus direction of propagation are measured for the initial state and for aged states after 10, 20 and 25 days. From these velocity measurements, the elastic constants of the material are determined. The consideration of relative variation of stiffness constants C_{ij} from the initial state C^0_{ij} assumed undamaged in the framework of the thermodynamics of irreversible processes leads to a diagonal fourth order damage tensor D_{ij} [7]. The diagonal normalised damage variables which we will consider in this study are then given by the following relation :

$$D_{ii} = 1 - \frac{C_{ii}}{C^0_{ii}} \tag{2}$$

where D_{11} (resp. D_{22} and D_{33}) is the damage along the 1 (resp. 2 and 3) axis, D_{44} (resp. D_{55}) is the shear damage in the plane P23 associated to $C_{44} = G_{23}$ shear modulus (resp. P13 $C_{55} = G_{13}$ shear modulus). The variations of the stiffnesses C_{11}, C_{22}, C_{33}, C_{44} and C_{55} versus time of hygrothermal ageing give the evolution of the five damage components D_{11}, D_{22}, D_{33}, D_{44} and D_{55} using the previous relation. These variations are shown in figure 6.

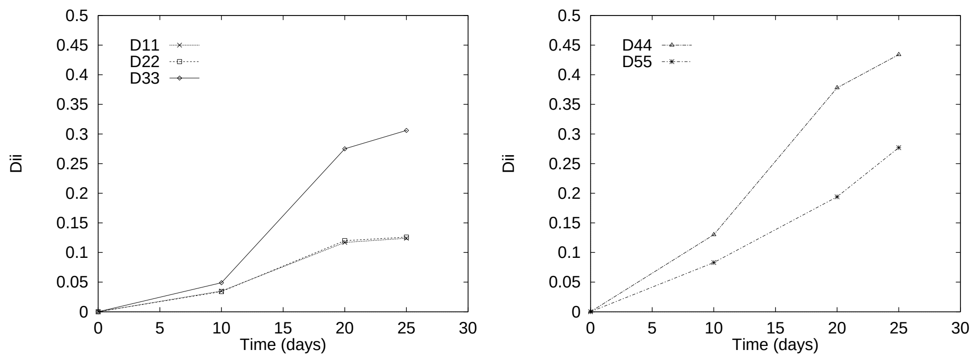

Fig. 6: Damage variables

All internal damage variables increase with the hygrothermal ageing but these variations are different according to the considered directions. This fact clearly demonstrates the anisotropy of the damage. The greatest increase occurs for the shear damage in the plane P23. Where the term D_{44} (related to C_{44} or G_{23}) reaches 0.45. The less important ones are along 1 and 2 axis and concerns the in plane elastic constants C_{11} and C_{22}. We note also an important increase of D_{33} related to C_{33} showing an important damage through the thickness. These macroscopic effects of damage, which are more or less important according the different directions, result from oriented micro-defects within the material. The large increase of D_{44} results certainly from the damage of the fibre/matrix interface.

SEM Observations

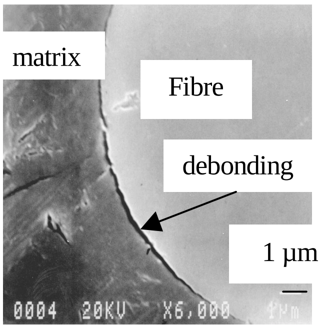

Fig. 7: SEM Observation

A microscopic investigation conducted using scanning electronic microscope (SEM) on the damaged sample confirms the above assumption and shows an important debonding at the fiber-matrix interface mostly located at the pole of the fibres in the thickness direction as shown in fig. 7.

CONCLUSION

According to this study, we can say that ultrasonic evaluation is an efficient tool for the characterisation of the hygrothermal ageing of glass-fibre reinforced epoxy composite. The stiffnesses C_{33} and particularly C_{44} are the elastic constants which are the most affected by the damage. C_{33} is the stiffness according to the direction normal to the sample and C_{44} is the shear elastic constant of plane P23, perpendicular to the fibre direction, this last coefficient depends strongly on the fibre matrix bonding. These ultrasonic results conducted together with SEM Observations show the anisotropy of such a damage and that it affects preferentially the fiber matrix interface.

REFERENCES

1. Chateauminois A., Chabert B., Soulier J.P. and Vincent L., «Study of the interfacial degradation of a glass-epoxy composite during hygrothermal ageing using water diffusion measurements and dynamic mechanical thermal analysis» Polymer, 1994, Vol. 35, N° 22, p. 4766-4774.

2. Selzer, R. and Friedrich K., «Mechanical properties and failure behaviour of carbone fibre-reinforced polymer composites under the influence of moisture» *Composites Part A*, 1997, N° 28A, p. 595-604

3. Naceri A., Surrel Y. and Vautrin A.,«Influence de l'humidité sur le comportement en cisaillement de composites à matrice polymère. *Annales des composites : composites à matrice polymère et humidité* » bulletin de l'AMAC, 1992, p.5-14.

4. Royer D. and Dieulesaint E. «Ondes élastiques dans les solides - Tome 1 : Propagation libre et guidée. » Paris : Masson, 1996. 328 p.

5. Mouchtachi A., El Guerjouma R. and Baboux J.-C. «Nondestructive ultrasonic evaluation of fibrous metal matrix composites» *Applied Composite Materials,* 1995, Vol. 1, p. 387-393.

6. Dubuget M. «Evaluation non destructive des matériaux par ultrasons : caractérisation de l'état initial et suivi sous charge des propriétés d'élasticité linéaire et non linéaire d'alliages d'aluminium». *Thèse de doctorat : Institut National des Sciences Appliquées de Lyon*, 1996. 196 p.

7. Baste S., El Guerjouma R. and Audoin B. «Effect of microcracking on the macroscopic behaviour of ceramic matrix composites : ultrasonic evaluation of anisotropic damage. » *Mechanics of materials*, 1992, Vol. 14, N° 1, p. 15-31.