 SL, vol.8, no.2, pp.83-97, 2012

Nondestructive and Destructive Testing of Reinforced Polymeric Materials

E.E. Theotokoglou[1], **E. Sideridis**[1]

Abstract: In this work a class a particle reinforced polymers have been investigated using nondestructive and destructive techniques. The velocities c_l and c_t of the longitudinal and transverse waves were evaluated using ultrasounds. From these and from the density of the material the modulus of elasticity, the shear modulus and the Poisson ratio were calculated by the appropriate relationships. The results were compared with those obtained from destructive tensile experiments and as well as with those derived from theoretical formulae. Also, their attenuation coefficient α and damage parameter D were obtained and an attempt was made to establish a relationship between strength and the effect of frequency, results obtained from dynamic experiments carried out on iron- ultrasonic attenuation in the composite material. Finally, in order to investigate the effect of frequency, results obtained from dynamic experiments carried out on iron-epoxy composites were compared with those obtained from ultrasounds.

Keywords: Reinforced polymeric materials, non destructive technique, dynamic experiments.

1 Introduction

Particulate composites are composites reinforced with particles having dimensions of the same order of magnitude. Particulate composites are produced from a polymeric matrix, into which a suitable metal powder has been dispersed. One role of the matrix is to protect the filler from the corrosive action of the environment and to ensure interactions between the fillers by mechanical, physical and chemicals effects.

Epoxy resins are the most suitable polymers for composite matrixes, and extensive research has been carried out on their rheological behavior [Theocaris(1962)] and their mechanical properties [Hirai and Kline (1973)]. The interrelationship

[1] NTUA, Department of Mechanics Laboratory of Testing and Materials, Zografou Campus 15773, Greece.

of mechanical and optical properties has been investigated previously for various amounts of plasticizer [Theocaris and Prassianakis (1978)] and acoustic properties determined for plasticized epoxies and correlated with the corresponding mechanical properties [Prassianakis (1990) , Konstantelos and E. Sideridis (1994)].

The mechanical and acoustic properties of the epoxy polymers can change by adding different amounts of filler. Metal oxides and metal powders have been used in combination with epoxy matrixes to create composites. The mechanical and thermal properties of such resins filled with iron particles have been investigated, and the effect of particle size on the same properties of iron filled epoxies has been extensively studied [Papanikolaou, Paipetis and Theocaris (1977), Theocaris, Papanikolaou and Sideridis (1982)].

A rigorous description of a composite system consisting of a matrix, in which filler particles have been dispersed, is difficult to undertake. Many geometrical, topological, mechanical, etc., parameters are necessary, the majority of which vary statically or are unknown. Theoretical treatments usually attempt to exploit as much readily available information as possible, which generally consists of the mechanical properties of matrix and filler and the volume fraction of the latter. Appropriate assumptions must be used for the missing data.

Ultrasonic techniques have been widely used for non-destructive inspection and evaluation (NDE) of composite materials. Hale and Ashton (1985) related strength reduction and change in ultrasonic attenuation of progressively damaged glass reinforced plastics. Williams, James, Lee, Samson, Wang and Tony (1987) used the NDE technique of ultrasonics to characterize separation mode and fracture strength for adhesively bonded fibre reinforced plastics. In addition, ultrasonic testing can be used to indicate the fibre direction in a composite material [Dean (1974)] and for general evaluation and quality control of components by attenuation and analysis of frequency or spectroscopy [Teagle (1983), Rose, Carson and Leidel (1973)]. The use of ultrasonic techniques for the determination of the mechanical properties in composite materials has been described by Smith (1972) and by Dean and Locket (1973).

In the present work the acoustic and the mechanical behavior of an epoxy polymer, reinforced with different amounts of iron particles at ambient temperature, was investigated. Acoustic properties were compared with the corresponding mechanical properties obtained from tensile experiments. The comparison revealed large discrepancies depending on frequency. The elastic modulus obtained from ultrasonic tests was then compared with the elastic modulus of the same material obtained from dynamic experiments. Also the attenuation coefficient and the damage parameter were evaluated.

2 Ultrasonic equipment and measurement procedures

Energy pulse propagation through the structure at frequencies above the audible range can be related to the material properties. The velocity propagation can be measured since modulus = density $\times$ velocity [Theocaris and Prassianakis (1978)]. However, the main aim of the ultrasonic testing of materials is to search and evaluate locations in the materials which contain discontinuities and to determine the effects of interaction between sound waves and material properties. The basic parameters required for all ultrasonic measuring methods are sound velocities and sound attenuation through the material in which the sound wave travels. Sound velocities c_l and c_t of the longitudinal and transverse waves, respectively, and the density ρ_c of the material are used for the evaluation of the elastic modulus E_C , the Poisson ratio ν_c, the shear modulus G_C via the following relationship [Krautkramer and Krautkramer (1977)].

$$E_c = \frac{(1+\nu_c)(1-2\nu_c)}{(1-\nu_c)}\rho_c c_l^2, \tag{1a}$$

$$\nu_c = \frac{1/2(c_l/c_t)^2 - 1}{(c_l/c_t)^2 - 1}, \tag{1b}$$

$$G_C = \rho_c c_t^2 \tag{1c}$$

The measuring system consists of a broad band (0.5-15 MHz) ultrasonic pulser-receiver flaw detector [Krautkramer and Krautkramer (1977)] which can generate and receive electric pulses up to 15 MHz. K2G and K2N probes were used as transmitting-receiving transducers of sound waves, producing ultrasounds of 2 and 4 MHz, respectively. Simple machine oil was used as the transducer/specimen interface couplant. A contact load for both probes of 9.88 N was applied to the transducer/specimen interface.

The pulser section produces and injects ultrasonic pulses into the specimen through the transducer and the reflected signals produced and amplified by the receiver section of the equipment and displayed on the oscilloscope.

The sound velocity c_l^x of the longitudinal waves of each specimen was evaluated using the relationship [Krautkramer and Krautkramer (1977)]

$$c_l^x = c_l \frac{d_x}{d_g}, \tag{2}$$

where c_l is the sound velocity of the reference block, d_x is the real specimen thickness, and d_g is the equivalent thickness of the specimen, which is measured on the screen of the oscilloscope.

The above mentioned modules are strongly affected by the frequency of the elastic wave and finally from the two basic properties, which are the viscosity and the elasticity of the materials. This dependence is seemed in the following basic relation given by Perepechko (1975)

$$c^2 = c_0^2 + \int_0^{\infty} \frac{H(t)\omega^2\tau^2}{\rho(1+\omega^2\tau^2)} d\tau, \tag{3}$$

where $\omega = 2\pi f$, f is the frequency of the elastic waves, $\tau = \eta/E$ is the relaxation time, η is the viscosity of the material, $H(\tau)$ is the density of the spectrum of relaxation times and c_0 is the limit velocity which corresponds to the static modulus of elasticity E. From this equation one can see that when $\omega\tau \to 0$ the phase velocity c tends to be equal to c_0 ($c \to c_0$). This means that either the frequency $f \to 0$ (static loading) or the relaxation time $\tau \to 0$, which case corresponds to highly elastic states (low viscosity and high elasticity).

In the other limiting case, when $\omega\tau \to \infty$, from the equation (3) follows that with a growth of frequency f or of the relaxation time τ (high viscosity and low elasticity) the velocity of waves should increase tending to a limiting value.

The attenuation coefficient α, was evaluated for each composition of plasticized epoxy polymer, using the following well known relationship.

$$\alpha = \frac{20}{2d} \log \frac{H_0}{H}, \tag{4}$$

where d is the specimen thickness and H_0 and H are the two successive backwall echo (the first and the second) heights measured on the screen of the ultrasonic equipment. Furthermore, the parameter of internal damage D can be determined. This parameter defined as

$$D = \frac{A - A_0}{A}, \tag{5}$$

where A denotes the initial cross-sectional area and A_0 the effective (undamaged) load transmitting area. Prior to formation of cracks, $A = A_0$ and hence D=0. For the other extreme, A_0 can be zero giving D=1. The inequality limits the range of D

$$0 \le D \le 1 \tag{6}$$

The ultrasonic determination of the internal damage D of a material can be made experimentally using the formula [Dean (1974), Prassianakis (1990)]

$$D = 1 - \frac{H_i}{H_0}, \tag{7}$$

where H_0 is the amplitude of the reflected ultrasonic wave monitored on the screen of the ultrasonic device for the unloaded specimen and H_i the value of the amplitude of the same wave during loading.

When a specimen is loaded by a stress σ_i, the same relation (7) gives the damage versus the loading if we take in consideration that in this case H_0 is the echo height on CRT-screen at zero stress ($\sigma = 0$) and H is the height of the same echo on the screen at stress σ_i .

3 Material and experimental work

3.1 Testing material

The specimen used consisted of a matrix material ,which was a cold setting system based on a diglycidyl ether of bisphenol-A resin having an epoxy equivalent of 185-192, a viscosity of 15 N s m^{-2} at 25^{o}C, and molecular mass between 370 and 384, cured with 8% by wt-triethylenetetramine filled with iron particles of average radius 75 μm. The elastic moduli of the matrix and filler were 3.5 and 210 GNm^{-2}, respectively, and their Poisson ratios were 0.36 and 0.29, respectively.

3.2 Tensile experiments

Dogbone specimens with constant dimensions of measuring area 6 x 3 mm^2 and length 45 mm were used during the tensile tests which were carried out with an Instron type testing machine at room temperature. The specimens were tested at a rate of extension of 1 mm min^{-1}. Five filler volume fractions υ_f and five specimens for each volume fraction were used and the values given correspond to their arithmetic mean value. For the obtention of the stress-strain diagrams, strain gauges (KYOWA type, gauge factor k=1.99) were located on the specimen to measure the strains.

3.3 Ultrasonic experiments

The NDE technique used in the present work was the ultrasonic pulse-echo technique proposed by Krautkramer and Krautkramer (1977). When ultrasonic pulses are introduced into a specimen, they reflect on a discontinuity or on the back wall of the specimen. The magnitude of the echo reflections depends on the changes in the impedance across the specimen.

To determine the velocities of longitudinal and transverse waves, five specimens from each volume fraction of the composite material were tested ultrasonically at ambient temperature. During each experiment the quantities obtained from the oscilloscope screen were the equivalent thickness d_g of the particle filled composite

and the echo heights. Measurements at three different points in each of the five specimens were carried out. From these quantities and using equation (2), the velocity c_l was evaluated. A suitable probe for the longitudinal waves with frequency 4 MHz was used. For the evaluation of the velocityc_t, a suitable probe for transverse waves with frequency 2 MHz was used. From the analogous equation (2) this velocity was calculated

$$c_t^x = c_t \frac{d_x}{d_g}$$

4 Results

Fig. 1 shows the variation of the longitudinal and transverse wave velocities versus the filler volume fraction. From these curves it can be observed that as the amount of inclusions increases, both velocities decrease. Fig. 2 presents the variation of the elastic modulus and shear modulus versus the volume fraction as obtained from the ultrasonic measurements. It can be observed that as the amount of inclusions increase both moduli increase.

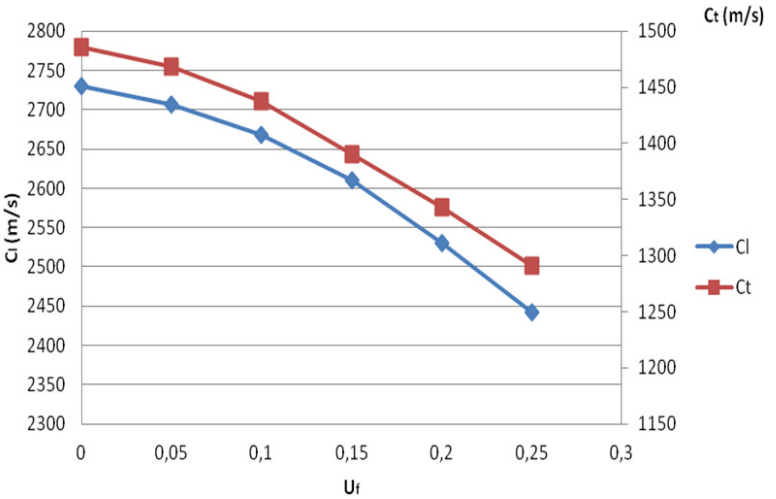

Figure 1: Variation of c_l and $c_t$versus υ_f

Figure 3 shows the variation of the composite density versus the filler volume fraction. It was evaluated experimentally by weighting pieces of given dimensions and dividing the weight by the volume. The theoretical values were calculated using the following formula and the values, $\rho_f = 7800kg/m^3$, $\rho_m = 1190kg/m^3$ for the

filler and matrix respectively.

$$\rho_c = \rho_f \upsilon_f + \rho_m(1 - \upsilon_f) \tag{8}$$

Fig. 4 presents the variation of the attenuation coefficient αas well as the damage parameter D versus filler content of the composite. The values of damage have been normalized to the value of the damage of the epoxy matrix .The damage increase from the value equal to zero as the filler content increases. Similar behavior can be observed in the variation of the attenuation coefficient which increases from the value of $8.2\,dB/cm$, for the epoxy matrix.

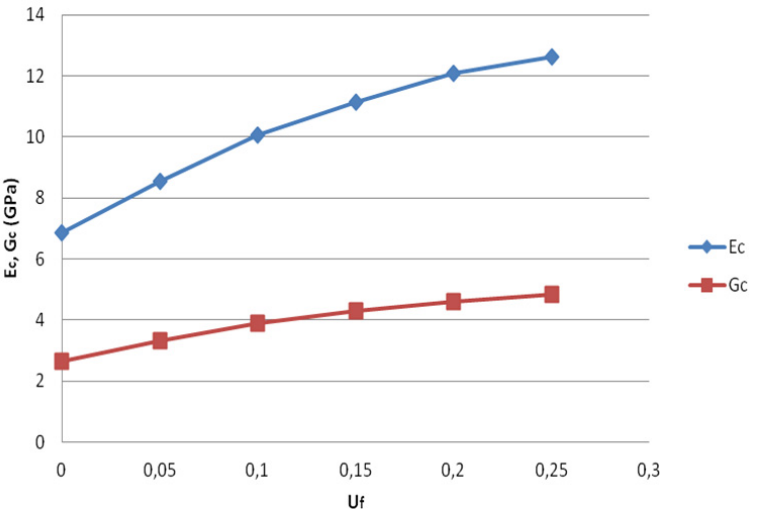

Figure 2: Variation of E_c and $G_C$of the composite versus υ_f

Fig. 5 shows the variation of stress σ_c versus the longitudinal strain $\varepsilon_c$for various filler fractions (0, 0.05, 0.10, 0.15, 0.20, and 0.25) of the iron particle reinforced epoxy polymer as obtained from tensile experiments. By comparing these curves it can be observed that as υ_f increases, the linear portion of the stress-strain curve increases. This mainly is due to the matrix being a viscoelastic material while the fillers are elastic materials. The addition of iron particles reinforces the elastic behavior of the composite, since it becomes more brittle.

Fig. 6 presents the variation of the tensile stress at fracture σ_c versus the filler volume fractionυ_f obtained from the tensile experiments. For comparison Fig. 6 also shows the curves obtained from the theoretical formulae of Nielsen (1966), Nicolais, Mashelkar and Shchrager (1978) given by equations (A_1)-(A_3) (*see* Appendix). It can be observed that σ_c decreases as the filler volume fraction increases. The experimental values are in good agreement with the theoretical ones.

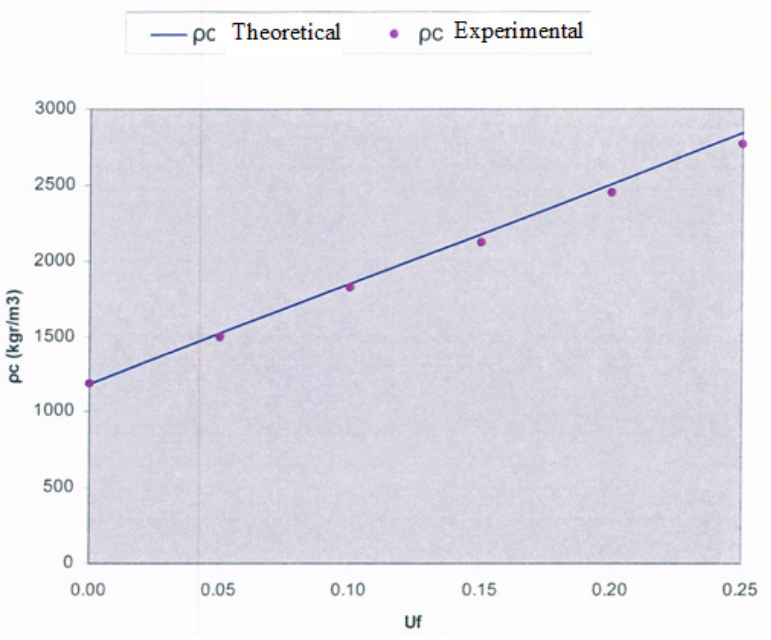

Figure 3: Variation of the composite density ρ_c versus υ_f

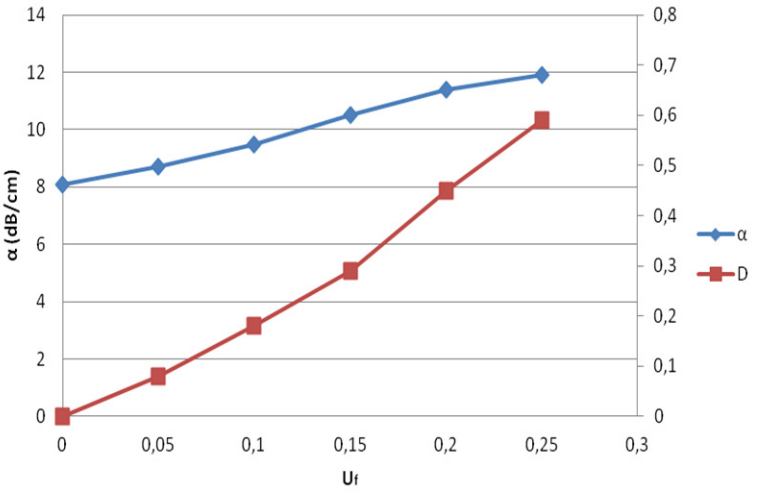

Figure 4: Variation of the attenuation coefficient α and of the damage D versus υ_f

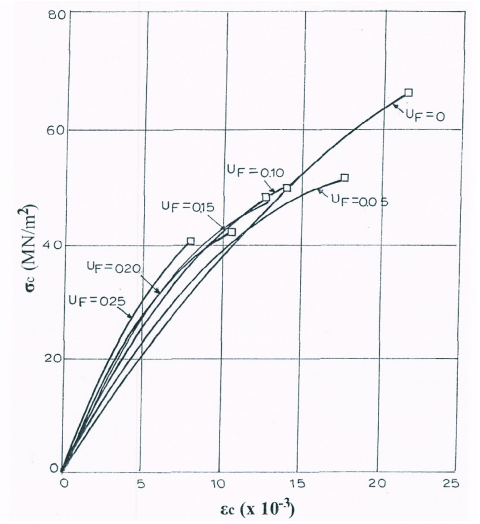

Figure 5: Stress-strain curves obtained from tensile experiments

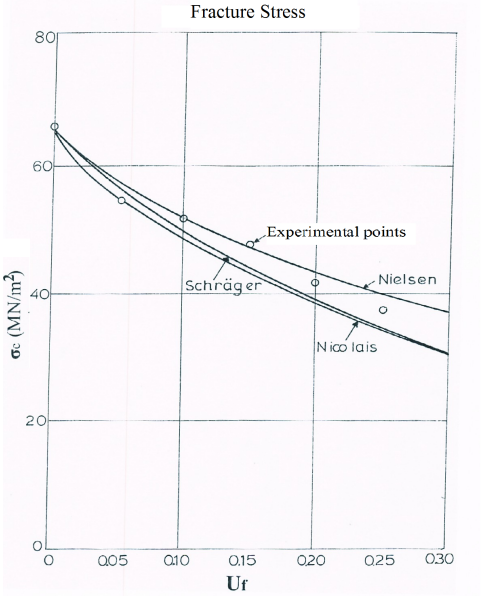

Figure 6: Variation of the tensile fracture stress of the composite

The variation of the elastic modulus E_c of the composite versus the filler volume fraction υ_f obtained from tensile experiments is shown in Fig. 7. For comparison, the curves obtained from the theoretical formulae of Counto (1964) and Kerner (1956) given by equations (A4) and (A5) are also shown (*see* Appendix). It can be observed that the elastic modulus increases as υ_f increases. The experimental values lie between the two theoretical curves.

In Fig. 7 ultrasonic and tensile values of the elastic modulus are also compared. It can be observed that ultrasonic experiments give much higher values than for tensile experiments as expected. This difference appears to result from the viscoelastic behavior of the polymer matrix, which is a strongly viscoelastic material and hence the elastic constant change during the tension test. This is not apparent, for example, in steel for which viscoelastic behavior is insignificant for the tension test and thus the elastic constants measured either ultrasonically or for tension tests are too close.

The behavior comparison with dynamic modulus subjected to sinusoidal varying loading, can be described by the complex frequency dependent moduli $E*(\omega)$ and $G*(\omega)$ where $\omega = 2\pi f$ the angular frequency is and f is the frequency. The following expressions hold

$$E* = E'(\omega) + iE''(\omega), \tag{9}$$

$$G* = G'(\omega) + iG''(\omega), \tag{10}$$

where primed symbols denote the storage modulus and double primed symbols denote the loss modulus of the material.

The procedure, by which effective complex moduli of viscoelastic materials can be determined, on the basis of analytical expressions for effective elastic moduli, is known as the correspondence principle and was developed by Hashin (1970). For isotropic viscoelastic materials a complex Poisson ratio can be introduced

$$\nu*(\omega) = \nu'(\omega) - i\nu''(\omega), \tag{11}$$

on the basis of the relationship between the moduli

$$G* = \frac{E*}{2(1+\nu*)}, \tag{12}$$

where E' which is the real part of the complex modulus, is known as the dynamic modulus and is proportional to the maximum energy stored during each cycle. Thus the expression for the velocity of a longitudinal wave in a medium can also be written

$$c_1 = \left[\frac{E'(1-\nu')}{\rho(1+\nu')(1-2\nu')}\right]^{1/2} \tag{13}$$

Fig. 8 shows the dynamic storage modulus E', of the iron particle reinforced composite material versus frequency, as obtained from dynamic experiments carried out on a

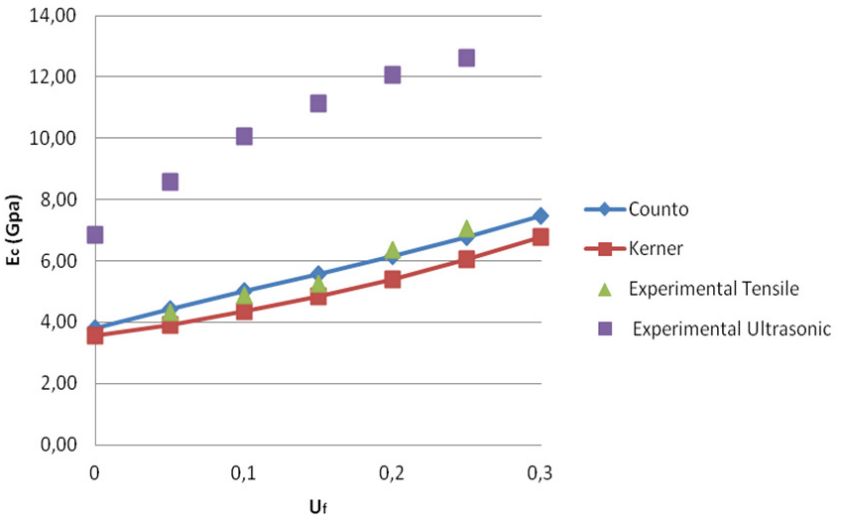

Figure 7: Variation of elastic modulus obtained from tensile experiments and from ultrasonic tests

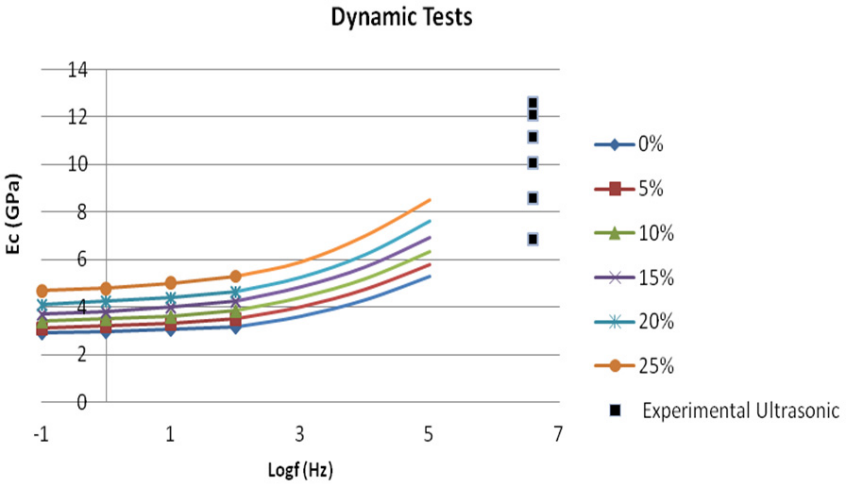

Figure 8: Variation of dynamic elastic modulus versus frequency

Dynastat and Dynalizer apparatus, which can apply a sinusoidal load of maximum amplitude 100 N. The measurements were carried out at frequencies from 0.1 to 100 Hz at ambient temperature and are described in Sideridis (1986). It can be observed that E' increases with frequency and if the curves obtained from the experimental values are extended beyond 100 Hz, as shown by the second part of the curves, the moduli achieved at approximately 2 MHz, will be close to those obtained using ultrasonics. In viscoelastic materials, the results for moduli obtained from ultrasonic measurements appear to be closer to the results obtained from dynamic tests than to those from static tests, owing to the strong variations in the properties of these materials with frequency. This difference can be observed if we take into consideration the elastic modulus dependence from the frequency of the elastic waves according to Eq. (3).

5 Conclusions

A comparison of the mechanical and acoustic properties of iron particle reinforced epoxy resin has been attempted. The velocities of longitudinal and transverse waves were considered as representative quantities of the ultrasonic behavior by using pulse-echo measurements. It was shown that the velocity of ultrasonic longitudinal and transverse waves propagating in the iron particle reinforced epoxy polymer decreases in a non-linear manner as the filler volume fraction increases.

The velocity of longitudinal waves is determined more simply and with greater accuracy than the velocity of transverse waves because these waves, produced by special probes, show high attenuation when passing through the particle filled polymers. The use of this type of waves presents difficulties for the study of the acoustic behavior of the particle reinforced epoxy polymers.

Conversely, the waves velocities are material properties that appear to depend on the discrete nature of the internal structure of the metal particle reinforced epoxy polymer, which may be altered by changes in the filler content. In addition, these materials suffer from microphysical damage accumulation in the form of void formation. The occurrence of a complex series of events at the microstructural level (e.g. microvoids or microcrack accumulation or filler agglomeration) in an initially intact material degrades structural integrity.

Tensile experiments carried out with the iron particle reinforced epoxy resins showed that the tensile strength decreases but the elastic modulus increases with filler volume fraction. Comparison between tensile and ultrasonic determination of the elastic modulus revealed large discrepancies. The values obtained from ultrasonic tests are much higher than those obtained from tensile tests.

Finally, it can be concluded that in materials showing viscoelastic behavior such

as iron particle filled epoxy resins, the results for moduli obtained from ultrasonic measurements are closer to the results obtained from dynamic experiments owing to the strong variations with frequency of the mechanical properties of these materials.

The authors propose to apply this investigation and comparison among static, dynamic and ultrasonic properties in other composite or similar materials such as random fibre composites [Theotokoglou and Sideridis (2011)] and sandwich structures [Theotokoglou and Tourlomousis (2010)].

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Appendix

To determine tensile strength Nielsen (1966) used the relationship

$$\sigma_c = \sigma_{F,m}(1-\upsilon_f^{2/3}) \tag{A1}$$

where σ_c and $\sigma_{F,m}$ denote the tensile stress at fracture of the composite and the

matrix, respectively, and υ_f is the filler volume fraction. However, Nicolais and Mashelkar (1976) use

$$\sigma_c = {}_{F,m}\,(1 - 1.21\upsilon_f^{2/3}) \tag{A2}$$

Finally according to Schrager (1978)

$$\sigma_c = \sigma_{F,m}\exp(-r\upsilon_f), \tag{A3}$$

where *r*is a factor determined experimentally which was found to be 2.66.

The Counto (1964) model uses the relationship for elastic modulus

$$\frac{1}{E_c} = \frac{1-\upsilon_f^{1/2}}{E_m} + \frac{1}{(1-\upsilon_f^{1/2})\upsilon_f^{1/2}E_m + E_f} \tag{A4}$$

where E_c,E_m, and E_f are the elastic moduli of the composite, the matrix, and the filler, respectively, and ν_m is the Poisson ratio of the matrix. To determine the same parameter, Kerner (1956) uses

$$E_c = E_m\{1 + \frac{\upsilon_f}{(1-\upsilon_f)}[\frac{15(1-\nu_m)}{8-10\nu_m}]\} \tag{A5}$$