3rd International Non-Destructive Testing Symposium and Exhibition, Istanbul Turkey, April 2008
For all papers of this publication click: www.ndt.net/search/docs.php3?MainSource=58

Acoustic and Mechanical properties of particulate composites

E. Sideridis, V.N. Kytopoulos, J.N. Prassianakis and I. Sakellaris

The acoustic and mechanical properties of a class of particle filled polymers have been investigated using destructive and non-destructive testing techniques. The velocities Cl

and Ct

of the longitudinal and transverse waves were evaluated from the acoustic properties. From these and from the density of the material, the modulus of elasticity, the shear modulus, and the Poisson ration were evaluated by the appropriate relationships. The results were compared with those obtained from tensile experiments and also with those obtained from theoretical formulae. Finally, to investigate the effect of frequency, the results obtained from ultrasonic experiments were compared with those obtained from dynamic experiments carried out on iron-epoxy composites. It was found that the results from ultrasonic tests are closer to those obtained from dynamic experiments than to those from static ones.

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 chemical effects.

Epoxy resins are the most suitable polymers for composite matrixes, and extensive research has been carried out on their rheological behaviour [1,2] and their mechanical properties [3,4]. The interrelationship of mechanical and optical properties has been investigated previously for various amounts of plasticiser [5] and acoustic properties determined for plasticised epoxies and correlated with the corresponding mechanical properties [6,7]. 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 [8,9].

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 [10] related strength reduction and change in ultrasonic attenuation of progressively damaged glass reinforced plastics. Williams et al. [11] used the NDE technique of ultrasonics to characterise separation mode and fracture strength for adhesively bonded fibre reinforced plastics. In addition, ultrasonic testing can be used to indicate the fibre directions in a composite material [12] and for general evaluation and quality control of components by attenuation and analysis of frequency or spectroscopy [13,14]. The use of ultrasonic techniques for the determination of the mechanical properties in composite materials has been described by Smith [15] and by Dean and Locket [16].

In the present work the acoustic and mechanical behaviour 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.

ULTRASONIC EQUIPMENT AND MEASUREMENT PROCEDURES

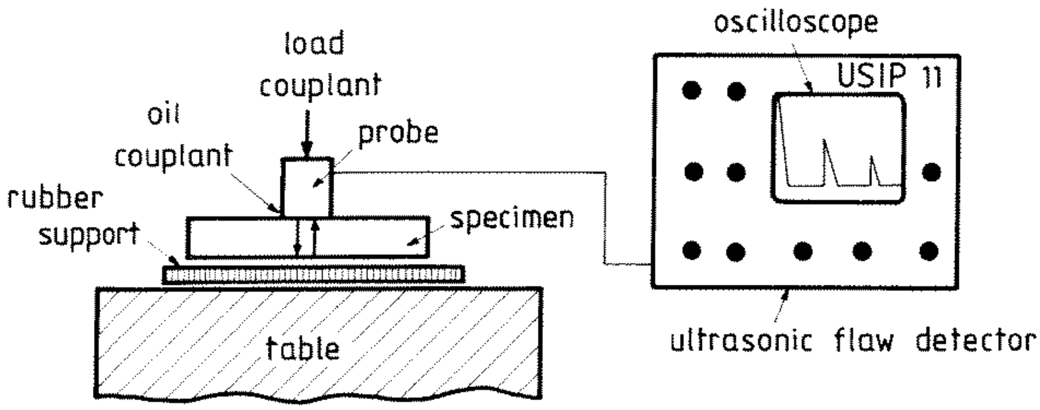
Energy pulse propagation through the structure at frequencies above the audible range, can be related to the material properties. The velocity propagation can be measured since modulus = density x velocity (Ref. 5). However, the main aim of the ultrasonic testing of materials is to search and evaluate 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 v_c , and the shear modulus G_c via the following relationships

$$E_c = \frac{(1+v_c)(1-2v_c)}{(1-v_c)}\rho_c c_1^2 \quad (1a)$$

$$v_c = \frac{1/2(c_l/c_t)^2 - 1}{(c_l/c_t)^2 - 1} \quad (1b)$$

$$G_c = \rho_c c_t^2 \quad (1c)$$

Figure 1 shows a schematic diagram of the ultrasonic pulse-echo measuring system used. The system consists of a broad band (0·5-15 MHz) ultrasonic pulser-receiver flaw detector (Krautkramer) 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. A 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.



1 Schematic diagram of ultrasonic pulse–echo measuring system used

The pulser section produces and injects ultrasonic pulses into the specimen through the transducer and the reflected signals produced are 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

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

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.

MATERIAL AND EXPERIMENTAL WORK

Testing material

The specimens 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 15Nsm^{-2} at 25℃, and molecular mass between 370 and 384, cur ed with 8 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.

Tensile experiments

Dogbone specimens with constant dimensions of measuring area 6x3 mm 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.

Ultrasonic experiments

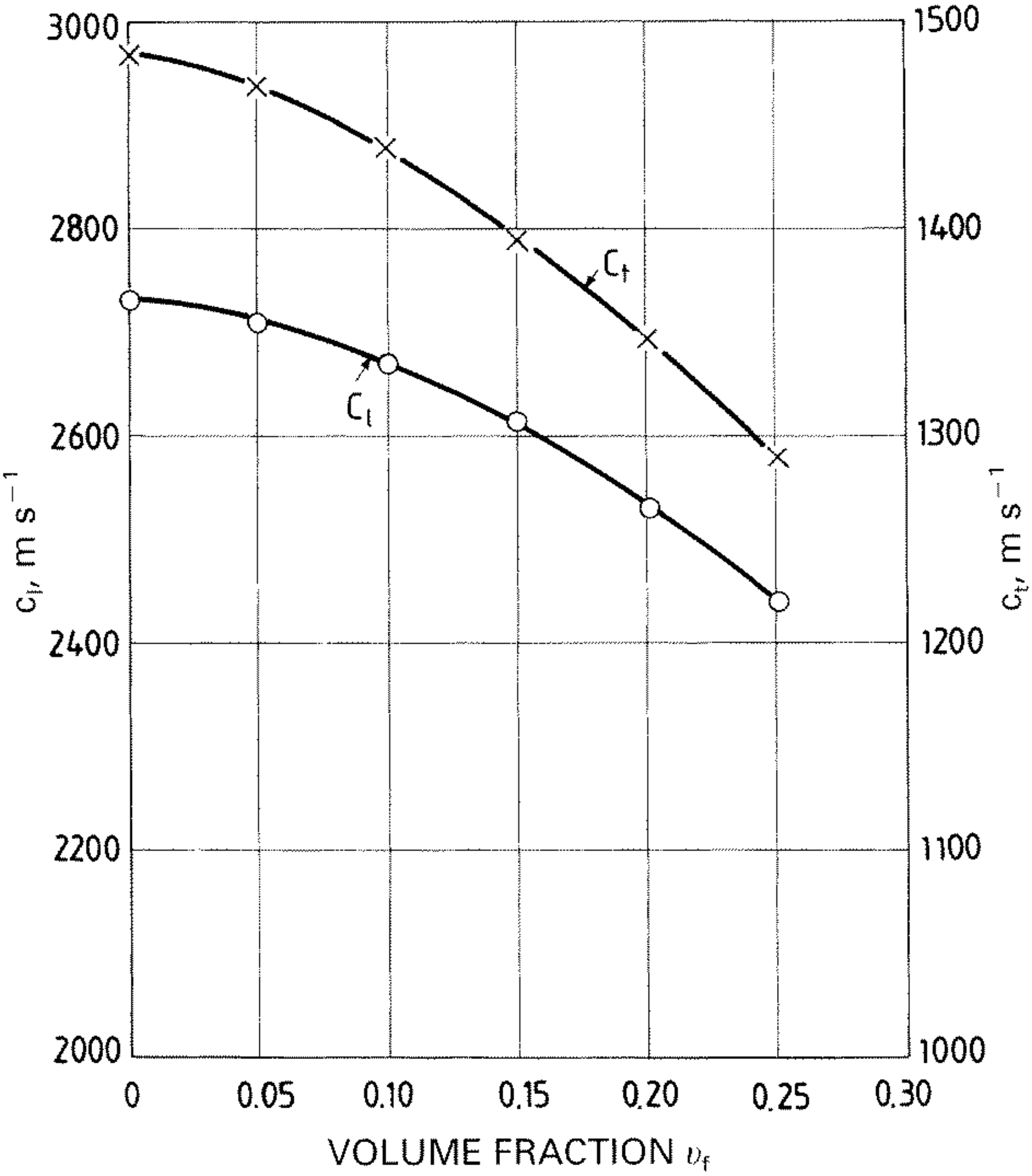
The NDE technique used in the present work was the ultrasonic pulse-echo technique [17]. When ultrasonic pulses are introduced into a specimen, they reflect on a discontinuity or on the back wall of the specimen. The magnitude of the echo reflections depends on the changes in the impedance across the 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 velocity c_t, a suitable probe for transverse waves with frequency 2 MHz was used. From the analogous equation (2) this velocity was calculated

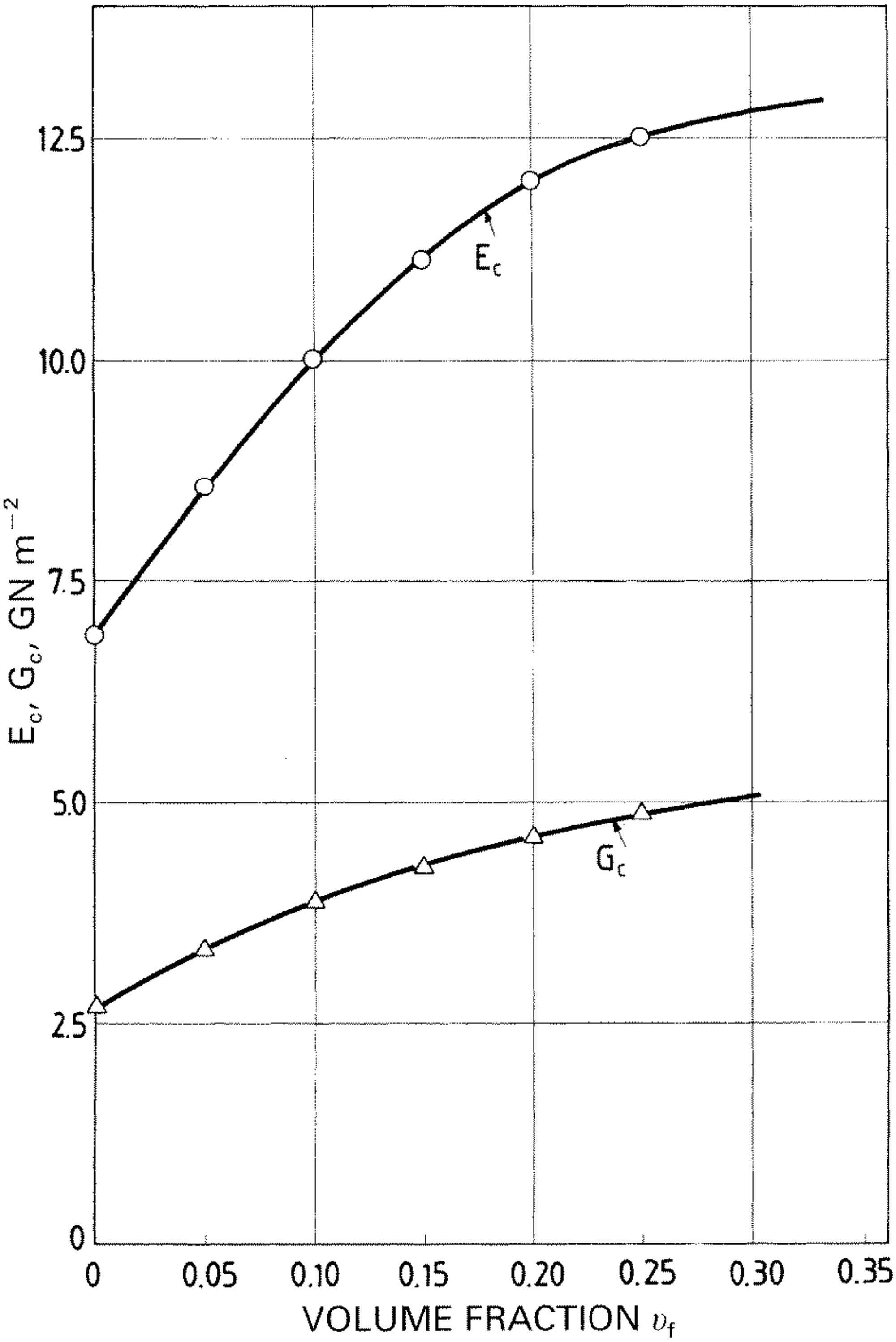
$$c_t^x = c_t \frac{d_x}{d_g} \tag{3}$$

RESULTS

Figure 2 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. Figure 3 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 increases both moduli increase.

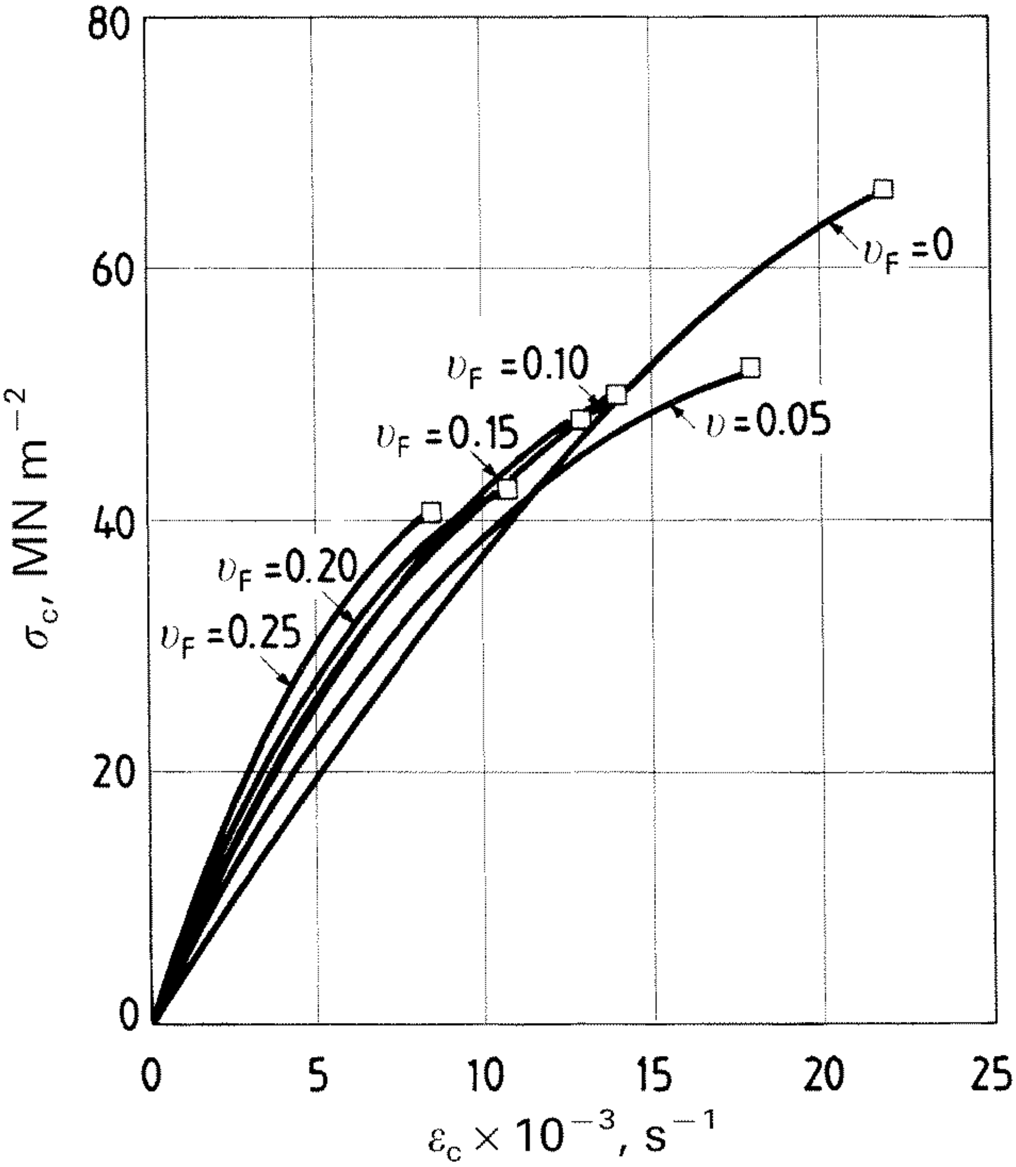


2 Variation of longitudinal c_l and transverse c_t wave velocities versus filler volume fraction v_f



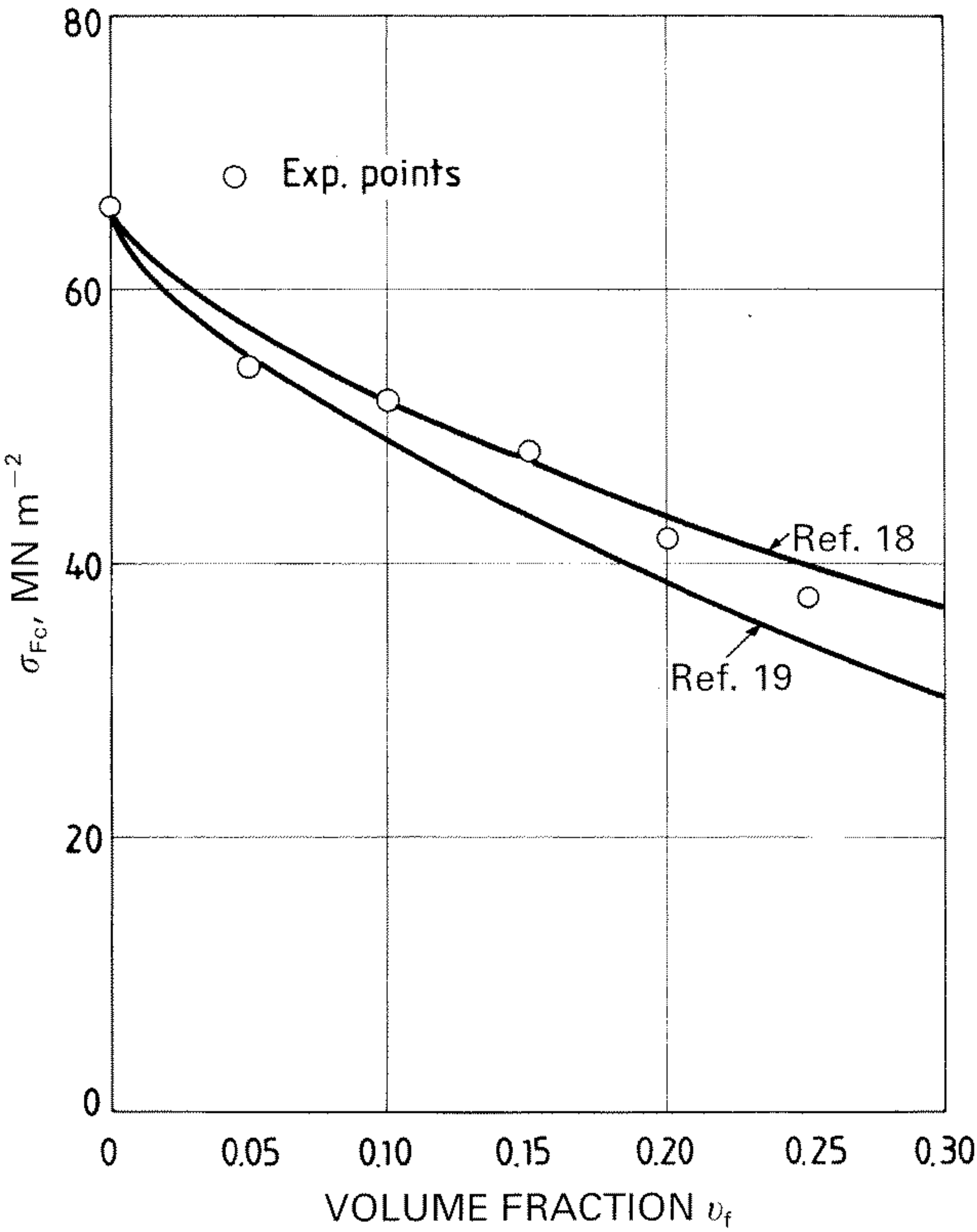
3 Variation of elastic modulus E_c and shear modulus G_c of composite versus filler volume fraction v_f obtained from ultrasonic measurements

Figure 4 shows the variation of stress σ_c v. the longitudinal strain ε_c for various filler fractions (0, 0·05, 0·10, 0·15, 0·20, and 0·25 vol.-%) 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 is due to the matrix being a viscoelastic material while the fillers are elastic materials. The addition of iron particles reinforces the elastic behaviour of the composite.



4 Stress–strain curves obtained from tensile experiments for different filler volume fractions

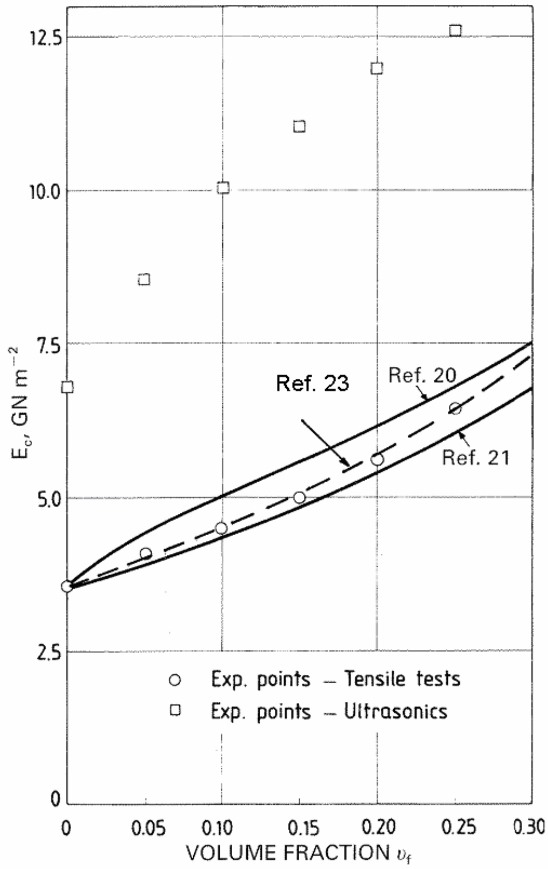
Figure 5 presents the variation of the tensile stress at fracture $\sigma_{F,c}$ versus the filler volume fraction υ_f obtained from the tensile experiments. For comparison the figure also shows the curves obtained from the theoretical formulae of Nielsen [18] and Nicolais and Marshelkar [19] given by equations (9) and (10): *see* Appendix. It can be observed that $\sigma_{F,c}$ decreases as the filler volume fraction increases. The experimental values are in good agreement with the theoretical ones.



5 Variation of tensile stress at fracture of composite $\sigma_{F,c}$ versus filler volume fraction v_f

The variation of the elastic modulus E_c of the composite versus the filler volume fraction v_f obtained from tensile experiments is shown in Fig. 6. For comparison, the curve obtained from a theoretical expression of the authors [23] given by equations (14) and (15) and the curves obtained from the theoretical formulae of Counto [20] and Kerner [21] given by equations (11) and (12) are also shown, *see* Appendix. It can be observed that the elastic modulus increases as v_f increases. The experimental values lie between the two theoretical curves.

In Fig. 6 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. This difference appears to result from the viscoelastic behaviour of the polymer matrix, which is a strongly viscoelastic material and hence the elastic constants change during the tension test. This is not apparent, for example, in steel for which viscoelastic behaviour is insignificant for the tension test and thus the elastic constants measured either ultrasonically or from tension tests are very similar.



6 Variation of elastic modulus *E* versus filler volume fraction v_f obtained from tensile experiments; comparison with values obtained from ultrasonic experiments

The behaviour of a viscoelastic material, when subjected to sinusoidally varying loading, can be described by the complex frequency dependent moduli $E^*(\omega)$ and $G^*(\omega)$ where $\omega = 2\pi f$ is the angular frequency and f is the frequency. The following expressions hold

$$E^* = E'(\omega) + iE''(\omega) \tag{4}$$

$$G^* = G'(\omega) + iG''(\omega) \tag{5}$$

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 [22]. For isotropic viscoelastic materials a complex Poisson ratio can be introduced

$$v^* = v'(\omega) + iv''(\omega) \tag{6}$$

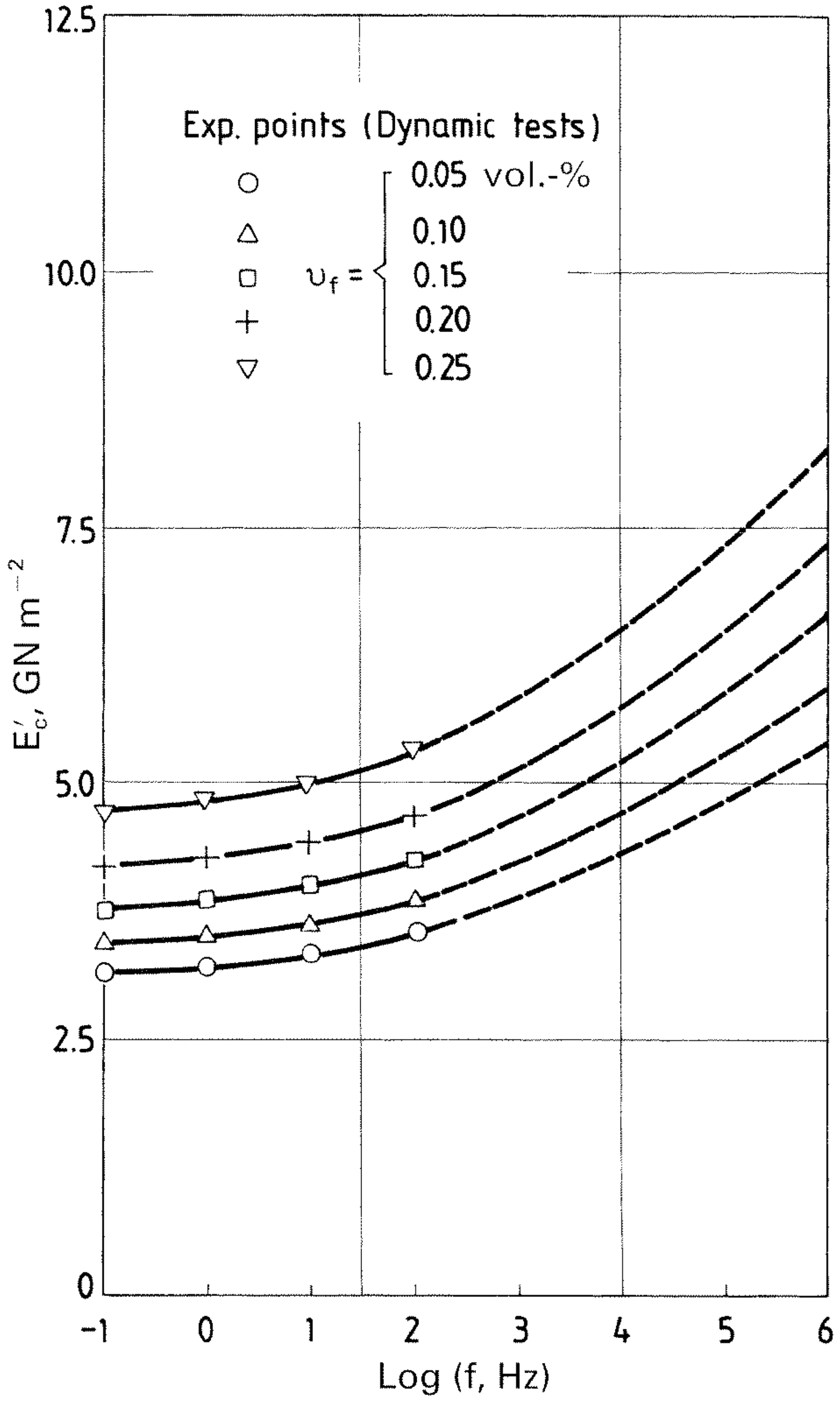
on the basis of the relationship between the moduli

$$G^* = \frac{E^*}{2(1+v^*)} \tag{7}$$

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

$$c_l = \left[\frac{E'(1-v')}{\rho(1+v')(1-2v')} \right]^{1/2} \tag{8}$$

Figure 7 shows the dynamic storage modulus E' of the iron particle reinforced composite material versus. frequency, as obtained from dynamic experiments carried out on a 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 Ref. 23. 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 discontinuous part of the curve, the moduli achieved at approximately 2 MHz, are 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.



7 **Variation of dynamic storage modulus of composite E'_c versus frequency for different values of filler volume fraction**

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 behaviour 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 rilled polymers. The use of this type of wave presents difficulties for the study of the acoustic behaviour of the particle reinforced epoxy polymers.

Conversely, the wave 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 behaviour 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.

APPENDIX

TENSILE STRENGTH

To determine tensile strength Nielsen [18] used the relationship

$$\sigma_{F,c} = \sigma_{F,m}\left(1-\upsilon_f^{2/3}\right) \tag{9}$$

where $\sigma_{F,c}$ 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 [19] use

$$\sigma_{F,c} = \sigma_{F,m}\left(1-1\cdot 21\upsilon_f^{2/3}\right) \tag{10}$$

ELASTIC MODULUS

The Elastic Modulus of the composite according to the procedure derived by one of the authors in Ref. 23 can be calculated as:

$$\frac{2\left(1-2\nu_c\right)}{E_c} = \frac{2\lambda^2\upsilon_f\left(1-2\nu_f\right)}{E_f} + \frac{1}{E_m}\left\{\frac{\upsilon_f\left(1-\lambda\right)^2\left(1+\nu_m\right)+2\left(\lambda\upsilon_f-1\right)^2\left(1-2\nu_m\right)}{1-\upsilon_f}\right\} \tag{11}$$

where the parameter λ is given as:

$$\lambda = \frac{3\left(1-\nu_m\right)E_f}{\left[2\upsilon_f\left(1-2\nu_m\right)+1+\nu_m\right]E_f+2\left(1-2\nu_f\right)\left(1-\upsilon_f\right)E_m} \tag{12}$$

The Poisson ratio v_c of the composite may be calculated with a great accuracy by the simple relation derived from the inverse law of mixtures, which is expressed by:

$$\frac{1}{v_c}=\frac{\upsilon_f}{v_f}+\frac{\upsilon_m}{v_m} \tag{13}$$

where v_f denotes the Poisson ratio of the filler, and v_m the Poisson ratio of the matrix.

Also the Counto [20] model uses the relationship for elastic modulus

$$\frac{1}{E_c}=\frac{1-\upsilon_f^{2/3}}{E_m}+\frac{1}{\left(1-\upsilon_f^{1/2}\right)\upsilon_f^{1/2}E_m+E_f} \tag{14}$$

where E_c, E_m, and E_f are the elastic moduli of the composite, the matrix, and the filler, respectively. To determine the same parameter, Kerner [21] uses

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

REFERENCES

1. P. S. THEOCARIS: *Rheol. Acta*, 1962, **2**, (2), 92.
2. P. S. THEOCARIS: *J. Appl. Polym. Sci.*, 1964, **8**, 399.
3. T. HIRAI and D. E. KLINE: *J. Appl. Polym. Sci.*, 1972, **16**, 3145.
4. T. HIRAI and D. E. KLINE: *J. Appl. Polym. Sci*., 1973, **17**, 31.
5. P. S. THEOCARIS and J. PRASSIANAKIS: *J. Appl. Polym. Sci*., 1978, **22**, 1725.
6. J. N. PRASSIANAKIS: *J. Appl. Polym. Sci.*, 1990, **39**, 2031.
7. B. KONSTANTELLOS and E. SIDERIDIS, *Plast. Rubber Compos. Process. Appl.*, 1994, **22**, 261.
8. G. C PAPANIKOLAOU, S. A. PAIPETIS, and P. S. THEOCARIS: *J. Appl. Polym. Sci*., 1977, **21**, 689.
9. P. S. THEOCARIS, G. C. PAPANICOLAOU, and E. SIDERIDIS: *J. Reinf. Plast. Compos*., 1982, **1**, 92.
10. J. M. HALE and J. N. ASHTON, Br. J. Non-Destr. Test., 1985, 1, 83.
11. J. H. WILLIAMS, J. R. SAMSON, S. LEE, and T. K. ANG: *Compos. Mater.,* 1987, **21**, 14.
12. G. DEAN: 'Characterisation of fibre composites using ultrasonics, composite-standards, testing and design', 126; 1974, National Physical Laboratory, UK.
13. P. R. TEAGLE: *Composites*, 1983, **14**, (2), 115.
14. J. L. ROSE, J. M. CARSON, and D. L. LEIDEL: 'Analysis of the test methods for high modulus fibers and composites', STP 521, 311; 1973, Philadelphia, PA, ASTM.
15. R. E. SMITH: *J. Appl. Phys*., 1972, **43**, (6), 2555.
16. G. D. DEAN and F. J. LOCKET: 'Analysis of the test methods for high modulus fibers and composites', STP 521, 236; 1973, Philadelphia, PA, ASTM.
17. J. KRAUTKRAMER and H. KRAUTKRAMER: 'Ultrasonic testing of materials'; 1977, Berlin, Springer-Verlag.
18. L. E. NIELSEN: *J. Appl. Polym. Sci*., 1966, **10**, 97.
19. L. NICOLAIS and R. A. MASHELKAR: *J. Appl. Polym. Sci*., 1976, **20**, 561.
20. U. J. COUNTO: *Mag. Concr. Res*., 1964, **16**, 129.

21. E. H. KERNER: *Proc. Phys. Soc*, 1956, **69B**, 808.
22. Z. HASHIN: *Int. J. Solids Struct*., 1970, **6**, 539.
23. E. SIDERIDIS: *Compos. Sci. Technol*, 1986, **27**, 305.