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Water sorption and temperature effects on the dynamic mechanical behaviour of epoxy-matrix particulates

G. C. PAPANICOLAOU*, A. PAPPA
University of Patras, Department of Mechanical Engineering, Applied Mechanics Laboratory, Patras, Greece

The effect of water absorption on the dynamic mechanical properties of composite systems consisting of a cold-setting epoxy matrix filled with iron particles was investigated over a wide temperature range. Storage moduli and loss factors of these composites were determined and the effect of water-conditioning temperature was examined. To minimize the complication of higher-order particle–particle interaction, a low filler content was used. Results indicate a strong dependence of dynamic properties on the water-conditioning temperature. It was found that depending on the water-conditioning temperature the presence of water molecules may either enhance or reduce the dynamic properties of the composite. The observed behaviour was similar to the respective behaviour of the same materials when tested in flexure. This behaviour was explained in terms of the dual action of absorbed water as a plasticizer and a crazing agent which deteriorates in a different way the physical and mechanical integrity of the epoxy-matrix composite.

1. Introduction

Epoxy polymers are widely used as glassy matrices for high-performance composites. Particulate composites are, by definition, materials consisting of a polymeric matrix reinforced with particles having dimensions of the same order of magnitude. This kind of reinforcement results in the production of new materials having improved thermomechanical, dynamic, electrical and other properties, when compared to the respective properties of the two main constituents. However, the characterization of a composite system depends on a great number of parameters affecting its overall behaviour. Such parameters are the shape and size of the reinforcing particles, the physicochemical properties of the constituents, the filler volume fraction, the type of dispersion and the amount of agglomeration.

In addition, the adhesion bond between polymer and filler is another parameter affecting greatly the strength of particulate composites. By the term adhesion we refer to the energy of interaction at the interface. For instance, a proper surface treatment of the filler particles can lead to a better adhesion between polymer and filler and, consequently, to a change in the overall behaviour of the composite system. As is well known, in the region between matrix and filler a third phase is developed because of the restriction of the segmental mobility of the macromolecules of the polymeric matrix, due to their adsorption to the filler surface. This area, which is called the *boundary interphase*, is characterized by structural microdefects such as impurities, microcracks and voids. These microdefects lead to localized stress concentrations, which are in excess of the average stress in the bulk of the material. If the localized stresses are sufficiently high, they may lead to a growth of the defect and a failure of the material. In previous publications [1–5] we have described and studied the concept of the boundary interphase and its effect on the overall thermomechanical properties of epoxy composites reinforced either with metallic powders or with glass fibres. In these investigations some theoretical models have been developed, and theoretical results were compared successfully with experimental findings.

The effect of the interphase material is of the upmost importance when studying the overall behaviour of composites. However, although extensive work has been conducted on the effect of the interphase on both the mechanical and thermomechanical behaviour of particulates as well as of fibre-reinforced composites, little is known about its effect on the dynamic behaviour of these materials and especially about the combined effect of interphase, water absorption and temperature on their dynamic properties.

Dynamic mechanical analysis offers a distinct advantage over other property measurements, for it provides the most sensitive response to various subtle physical and chemical transitions in polymers over a wide temperature range. However, the data in the literature often appear to be contradictory, largely

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*To whom all correspondence should be addressed.

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because of the effect of uncontrolled variables such as the adhesion between the two main phases. Also, most of these data give a very indefinite indication of how well the various theories are able to predict the properties.

It is the objective of the present work to study the combined effect of water absorption and temperature on the dynamic properties of iron–epoxy particulate composites.

2. Theoretical background

Concerning the conditions of use, it is well known that immersion of composite materials in aqueous environments results in a degradation of their thermomechanical properties [6–22]. This phenomenon is attributed to moisture-induced plasticization and/or micromechanical damage and matrix cracking [19, 23, 24].

The dual action of absorbed water as a plasticizer and a crazing agent deteriorates in a different way the physical and mechanical integrity of the epoxy matrix material. This is because plasticization is a reversible phenomenon whilst microcavitation damage is a permanent one. Especially for the latter, it has been found that water absorption affects the adhesion efficiency between filler and matrix as well as the extent of the boundary interphase. The action of water results in the abrupt disruption of the bonds between filler and matrix and the formation of additional cavities, which would be filled with water. It has been established [25] that water enters at the interphase at a rate of approximately 450 times more rapidly than in the bulk matrix itself, and consequently any absorbed moisture is concentrated preferentially in the interphase. The influence of water absorption on the extent of the boundary interphase in particulate composites has been studied [26]. It was found that, during the process of water absorption there is a variation of the extent of the interphase closely related to the observed degradation of the mechanical properties of the composites as well as to the amount of absorbed water. However, the influence exerted by the filler particles is probably not limited to the immediate vicinity of the particle surface. The mechanism by which the adsorption of polymer molecules on the filler surfaces reduces the number of possible configurations of the polymer molecules in a substantial portion of the polymer sample is admittedly not clear. It may be likened to a mechanical chain with two ends anchored on two filler particles, each polymer coil being depicted as a link in the chain. The mobility of such a mechanical chain is greatly reduced [27–29].

The effect of the temperature of the aqueous environment is another important parameter when studying the degradation of the mechanical properties of epoxy polymers and composites due to water absorption. It has been found [30, 31] that the critical stress-intensity factor of an unsaturated polyester falls significantly after exposure to hot water; this reduction was attributed to the water extraction of non-bound matter present in the resin. It was also found that thermal treatment of epoxy resins containing sorbed water affects their subsequent water sorption characteristics, elastic modulus and glass transition temperature [6–11].

Also, due to water absorption, the free volume and consequently the molecular mobility of the epoxy matrix macromolecules increases. This enhanced molecular mobility leads to a dissociation of the hydrogen bonds between the water molecules and the active sites within the epoxy matrix. Then, a general decrease of H-bonding develops and a consequent increase in the mobility of the water molecules, which requires a greater free volume than the H-bonds. This increase of the molecular mobility of the epoxy–moisture system causes a lowering of the glass transition temperature.

Concerning the importance of interfacial regions adjacent to the filler particles, Ziegel [32] reported that the immobilized matrix associated with the interphase does not contribute to energy loss, while the relative loss modulus E_c''/E_o'' is given by

$$E_c''/E_o'' = (1 - v_f B)^{-1} \qquad (1)$$

$$B = (1 - \Delta r/R_o)^3 \qquad (2)$$

where R_o is the radius of the particle, Δr is the thickness of the interphase, v_f is the filler volume fraction and E_c'', E_o'' are the loss moduli of the filled and unfilled matrix, respectively [32].

Also Sumita *et al.* [33] have shown in the case of particulate PP composites that the relative loss moduli at 60 °C, where the broad dispersion of these materials is observed, depend upon the content of effective volume fraction, i.e. the fraction of filler volume plus that of the interfacial region.

From the above discussion it follows that the interrelation between sorption and dynamic mechanical data is a difficult task due to the large number of parameters introduced.

The dynamic moduli of a polymeric matrix in which metal particles have been dispersed depend on the stress/strain fields developed around the individual particles under the action of dynamic loads. The latter depend (i) on the shape of the particles and their distribution in the matrix; (ii) on the size of the particles, in particular as compared with the length of the impinging stress wave; (iii) on the interaction between stress/strain fields of neighbouring inclusions; (iv) on the stress fields due to polymerization shrinkage [34, 35] which in many cases are not expected to be uniform; and (v) on thermal stress fields due to different thermal expansion coefficients of the materials at elevated temperatures [36]. The thermal conductivity of the filler is of particular importance in this latter case.

In addition, the nature of the interfacial bond between matrix and filler particles is of crucial importance. In fact perfect adhesion, corresponding to continuity of stresses and displacements at the interface, is a most common assumption for analytical treatments. However, with real composites this condition is very seldom fulfilled, and, in addition, cracks, voids and flaws extensively distort the stress/strain fields developed in the composite. A form of imperfect bonding, depending either on the nature and/or the size of the particles or on the manufacturing process, is agglomeration, when extremely complex situations, hardly susceptible to analytical treatment, develop.

An impedence mismatch, which is a common case with metal-filled plastics, tends to create situations similar to those caused by imperfect bonding. Indeed, a stress wave does not in practice enter the reinforcing particle but simply circumnavigates it. The filler in this case only perturbs the stress fields by restraining the matrix, provided that the strain rate is sufficiently high.

At elevated temperatures more complicated situations develop. Not only does impedance mismatch become more pronounced, but, in addition, the nature of the interfacial bond is greatly affected. Indeed, according to Molotkov *et al.* [37] the bonds are partly disrupted or reinstalled as functions of temperature and in close correlation with the frequency.

As distinct from the storage modulus, what appears to be extremely sensitive towards the complex situations already described is the energy dissipation, i.e. the overall loss factor of the material. Energy dissipation occurs primarily in the viscoelastic matrix and depends on the strains developed. The introduction of non-dissipative fillers decreases the effective dissipation volume, but creates additional strain fields, therefore increasing damping. Indeed, according to theoretical considerations and experimental data, introduction of such a filler produces loss factors increasing with volume fraction up to a certain limit above which the effective dissipation volume decreases faster than the particle-induced strains increase.

It should be emphasized that with real composites energy dissipation occurs at the interfaces by imperfect bonding due to Coulomb friction, as well as in the cases of cracks and flaws and also agglomeration of the particles. Under these circumstances hardly any theoretical predictions are attainable, in particular for filler volume fractions other than very low ones, and also when the combined effect of water absorption and temperature is introduced.

TABLE I Composition and cure cycle of resin and IEC

Resin	IEC
Epoxy: Epicote 828/100[a]	Matrix: Epoxy resin Filler: Iron particles of average diameter 150 μm
Cure accelerator: triethylenetetramine (TETA)/8[a]	Filler volume fraction: 2%
Cure cycle 24 h ambient temperature 7 days temperature rise at 5 °C h^{-1} till 100 °C, to ambient at 1 °C h^{-1}	

[a] 100 and 8 show the weight ratio of composition.

3. Experimental procedure

3.1. Materials

The matrix material was in all cases a cold-setting system based on a diglycidyl ether of bisphenol A resin (Epicote 828, Shell Co.) with the trade name DGEBA, cured with 8% triethylenetetramine (TETA). One particle size (150 μm) of iron powder was used for all specimens. The compositions and the cure cycle of the epoxy resin used as matrix and iron–epoxy composites are given in Table I.

Detailed information on DGEBA–TETA resin cure and iron–epoxy composite (IEC) sample preparation was previously described [38]. The properties of the constituent materials as well as of the IEC materials are given in Table II. The thermomechanical behaviour of the same composites as well as the effect of water absorption on their thermomechanical properties have been previously examined [38].

3.2. Water sorption tests

The specimens were totally immersed in water at 25, 40, 60 and 80 °C for periods up to 10 days, and when removed were wiped thoroughly by using a lint-free towel, and then weighed on a Mettler analytical balance.

3.3. Dynamic mechanical tests

The specimens were subsequently mounted on a Bruel and Kjaer type 3930 complex modulus apparatus. The test piece was in the form of a cantilever which could be excited in a resonance mode of vibration. The vibration was produced with a B & K type 1014 beat-frequency oscillator. Tests could be carried out at different frequencies by altering the free length of the

TABLE II Properties of constituent materials and IEC

Item	TETA-cured DGEBA resin	Iron	IEC
Lamé constants			
λ (GPa)	3.34	112	–
μ (GPa)	1.30	81	13.25
Young's modulus, E (GPa)	3.53	210	3.60
Bulk modulus, K (GPa)	4.21	167	–
Poisson's ratio, ν (–)	0.360	0.290	0.358
Density, ρ (kg m^{-3})	1.19×10^3	7.8×10^3	1.33×10^3
Tensile strength (MPa)	65	–	66
Coefficient of thermal expansion (°C^{-1})			
< 123 °C	65×10^{-6}	15×10^{-6}	58×10^{-6}
> 123 °C	168×10^{-6}	–	165×10^{-6}
Glass transition temperature, T_g (°C)	123	–	121

cantilever. All tests were conducted at room temperature. The properties of each specimen are presented as a storage modulus E' and a loss factor $\tan\delta$, the accuracy being estimated at better than 5%.

Fig. 1 shows the clamping apparatus, while the instrumentation arrangement for the determination of the complex modulus and loss factor is shown in Fig. 2.

4. Results and discussion

Experimental values of water content for various parametric values of water-conditioning temperatures are plotted in Fig. 3 versus time of immersion. The definition used for the percentage water content $M(t)$ of the composite is expressed by the relation.

$$M(t) = \frac{W_m - W_d}{W_d} \times 100 \qquad (3)$$

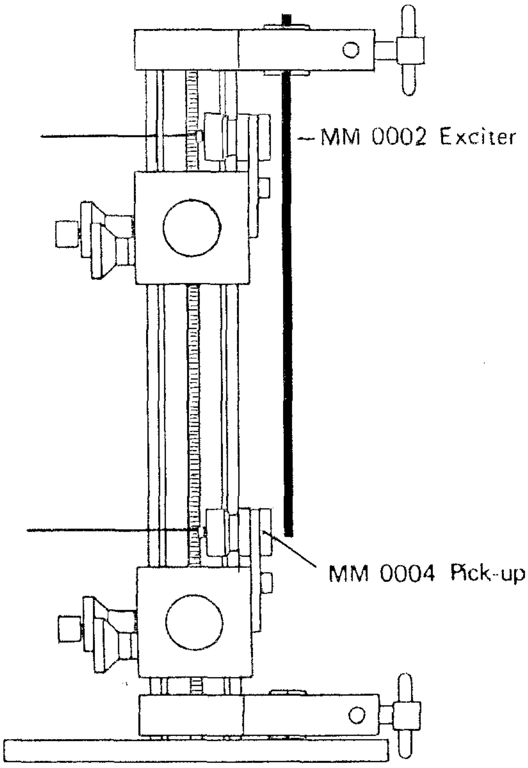

Figure 1 The clamping apparatus.

where W_m and W_d are the moist and dry weights of the specimen, respectively.

Each of these data points corresponds to the average value of about 20 specimens. From these graphs it may be observed that the rate of water absorption increases with water-conditioning temperature, since for temperatures in the range 25–40 °C the time needed for equilibrium is about 20 days, while when the water-conditioning temperature is 80 °C the respective time needed is only 9 days. Also, the value of the percentage water content at equilibrium M_∞ is higher for higher values of water-conditioning temperature.

The above-mentioned observations were well expected since, as is well known, the water absorption mechanism is a thermally activated one and this is the reason why the water absorption rate increases with temperature. However, experimental results in combination with the Arrhenius relation [20, 39] show that an increase in temperature affects the diffusivity D of the material in a direction normal to the lateral faces of the samples and thus, in turn, the rate of water absorption, according to the relation

$$D = D_o \exp(-E_d/RT) \qquad (4)$$

where E_d is the activation energy.

The above relation is an approximate relation which does not give accurate values for the diffusion coefficient of the material and which in the case of composite materials takes a different form. In its new form which has been defined for the case of isotropic materials [23] as well as under different conditions [15], the diffusion coefficient is dependent on several parameters such as the filler volume fraction v_f, the material's structure, the curing conditions, the existence of microcracks and finally the amount of absorbed water. Also, although there is some evidence, it is not well known [40] how the activation energy is affected by the presence of the inclusions, the interphase, the inclusion shape and other parameters.

In general, an increase in temperature results in an increase in the mobility of the macromolecules which, in turn, results in an increase of the volume occupied by the molecules due to their thermally-dependent vibrational movement and consequently of the overall volume occupied by the actual mass of the molecules

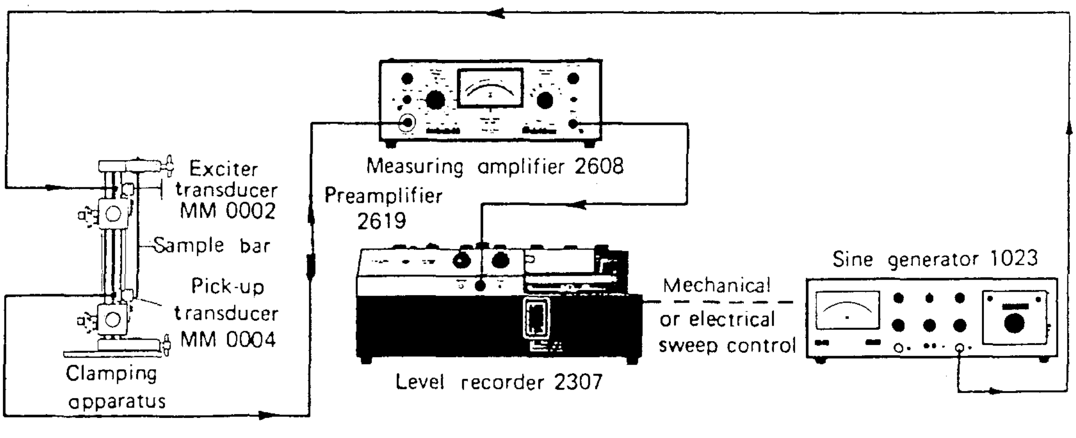

Figure 2 Instrumentation arrangement for the determination of the complex modulus and loss factor.

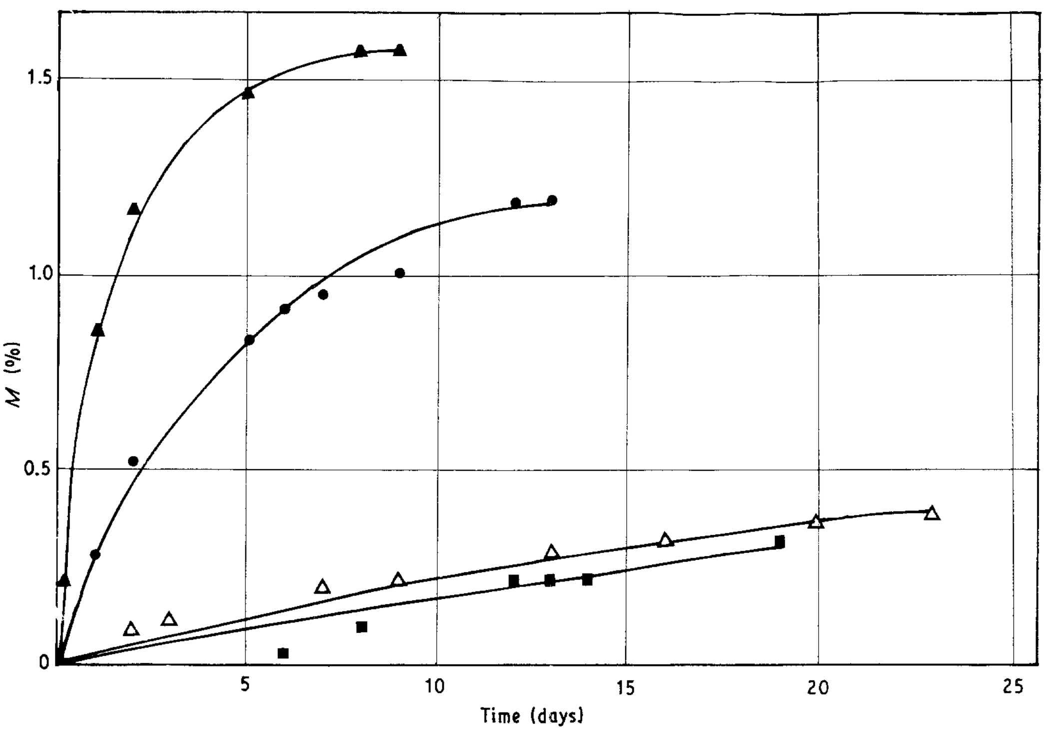

Figure 3 Variation of the percentage water content versus time of immersion for different water-conditioning temperatures: (△) 25 °C, (■) 40 °C, (●) 60 °C, (▲) 80 °C.

[26]. Thus, the material density decreases and the diffusion of the water molecules is enhanced.

From Fig. 3 it may be observed that during the initial period of time (the first 5 days) the rate of water absorption is high enough, while as the time passes, the specimens begin to fill with water, leading to a decrease in water absorption rate. As the equilibrium water content is approached, the rate of absorption becomes extremely low. This kind of behaviour may be explained by the fact that water initially enters rapidly into the specimen, filling the low-density areas of the specimen such as microcracks existing in the surface and inside the material as well as other structural irregularities, one of them being the interphase region. In the above areas water enters without any special difficulty [41, 42].

Fig. 4 shows the variation of the equilibrium water content as a function of water-conditioning temperature. From this figure it may be observed that the water-conditioning temperature has a significant influence on the water uptake. The mobility of penetrant molecules and chain segments increases with temperature and, therefore, a higher diffusion rate is obtained resulting in increased values of water uptake. Also, this kind of behaviour may be due to a combination of other parameters among which one may consider the inhomogeneous distribution of the water molecules within the composite, resulting in craze initiation in the matrix and the formation of water clusters in the polymer. In that case, only the water which is actually dissolved in compact resin is responsible for the plasticizing action [19, 30] creating preferentially highly plasticized zones which would have lower local shear stresses and favour cavitation.

Immediately after this initial period, a decrease in water absorption rate is observed and this may be explained since during this period water enters into denser structural areas while at the same time the –OH groups attract polar water interchain hydrogen bondings with the net effect of increasing the intersegmental hydrogen bond-length [39, 43]. This disruption of interchain hydrogen bonds results in volumetric changes of the specimen, that is the so-called "swelling". Finally, the amount of water absorbed approaches an equilibrium value. At this point

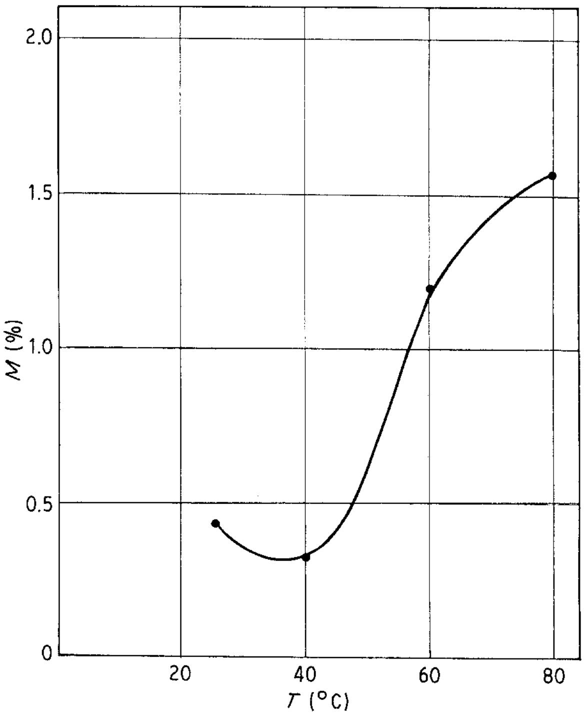

Figure 4 Change in weight of iron–epoxy particulate composites after immersion in water versus water-conditioning temperature.

we must notice that it has been proved [30] that the water absorption behaviour of epoxies is a Fickian one although damage induced in the epoxy or the epoxy composite can cause deviations from Fickian diffusion and accelerate moisture absorption.

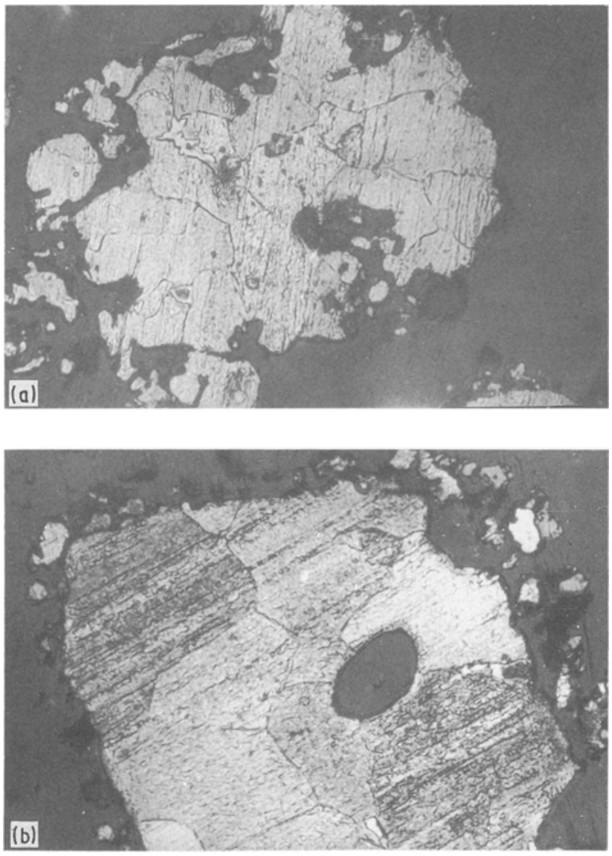

Figure 5 (a, b) Photomicrographs showing structural irregularities of iron–epoxy particulates. Magnification ×250.

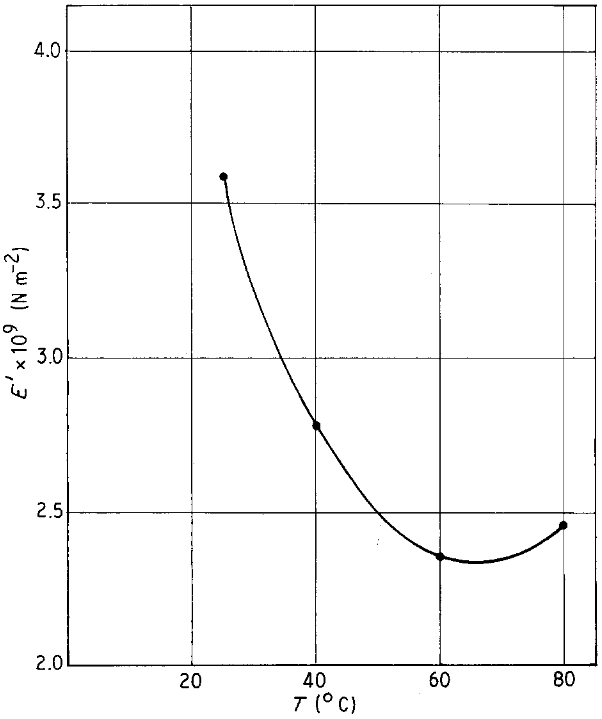

Figure 6 Variation of storage modulus of iron–epoxy composites after immersion in water versus water-conditioning temperature.

The results obtained in the present work show that the amount of water at 25 °C is slightly higher than the respective amount at 40 °C. This may be due to structural irregularities, aggregates and holes formed during the preparation of the specimens, as well as to structural relaxation phenomena. Such structural irregularities for the case of iron–epoxy particulates are shown in the photomicrographs presented in Fig. 5.

Figs 6 and 7 show the effect of water-conditioning temperature on the storage modulus E' and $\tan\delta$, respectively, of the material under investigation.

It may be observed that the storage modulus shows an initial sharp increase, goes through a minimum and then increases. This kind of behaviour can be correlated directly with the respective variations of the

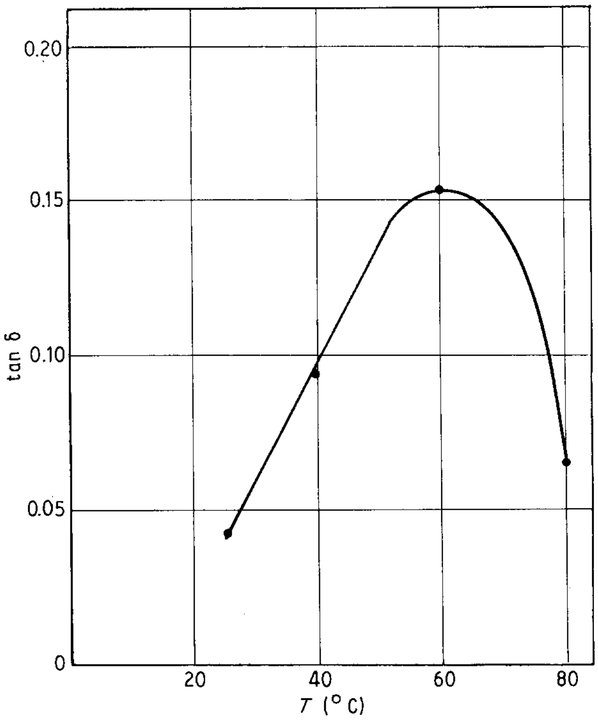

Figure 7 Variation of $\tan\delta$ of iron–epoxy composites after immersion in water for different water-conditioning temperatures.

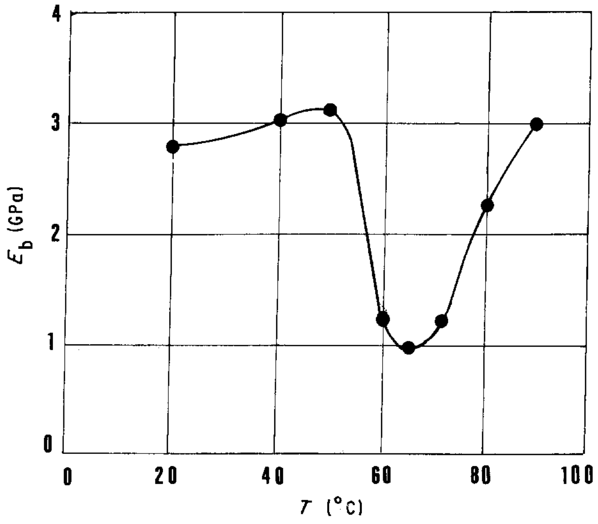

Figure 8 Variation of the flexural modulus of iron–epoxy composites after immersion in water for different water-conditioning temperatures.

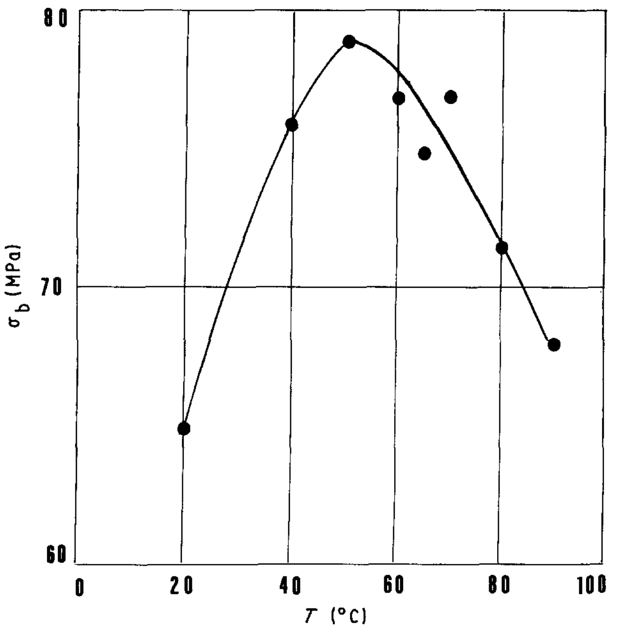

Figure 9 Variation of the flexural strength of iron–epoxy composites after immersion in water for different water-conditioning temperatures.

flexural modulus and flexural strength of the same material when tested under the same conditions [38, 44], and this is shown in Figs 8 and 9, respectively.

The above results can be explained in terms of the theory presented [38] according to which the plasticizing effect of the absorbed water is more pronounced at low temperatures (up to 60 °C) resulting in a lowering of E' and a respective increase in $\tan\delta$. At higher temperatures, microcavities are formed which in turn are filled with water. In that case only the water which is actually dissolved in compact resin is responsible for the plasticizing action, and if the amount of this water at lower temperatures is greater than the respective amount at higher temperatures, a net increase of E' and a simultaneous reduction in $\tan\delta$ must be expected.

5. Conclusions

In the present work a systematic study of the effect of water absorption on the dynamic mechanical properties of iron–epoxy particulate composites was undertaken. A large range of water-conditioning temperatures was employed. From the above investigation the following conclusions may be derived:

1. From the shapes of the isothermal absorption curves plotted in Fig. 3, it follows that during the initial time of immersion the absorption process is diffusion-controlled.

2. Water absorption increases with water-conditioning temperature.

3. The storage modulus decreases as the water-conditioning temperature is raised, having its minimum at about 60 °C, and then increases.

4. Tan δ increases with water-conditioning temperature, having its maximum at about 60 °C, and then decreases.

5. The above behaviour is explained in terms of the well-known theory presented in the literature.

6. The observed variations of E' and $\tan\delta$ can be correlated directly with the respective variations of the flexural modulus and flexural strength of the same material when tested under the same conditions.

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