

Dielectric Properties of an Epoxy Resin and Its Composite I. Moisture Effects on Dipole Relaxation

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Synopsis

Dipole relaxation dielectric loss behavior of a fiberglass-epoxy composite has been studied as a function of moisture uptake. A single widely distributed loss peak, centered at -16°C for 10 kHz measurement, is observed in the dry composite. Very low moisture concentrations ($<0.1\%$) interact with the composite, through either dipole pairing or by inducing chemical changes, to decrease loss intensity. At moisture concentrations from 0.1 to 1.4%, loss peak intensity generally increases with moisture uptake. Over this moisture concentration range water dipoles presumably relax in phase with the epoxy segments or side groups upon which they adsorb. Arrhenius relaxation activation energy decreases, and improved loss peak definition at increasing moisture concentrations is interpreted as indicating resin plasticization by adsorbed moisture. At moisture concentrations above 1.4%, the appearance of a second loss peak suggests formation of a new colloidal or weakly adsorbed moisture phase, as well as the saturation of primary adsorption sites.

INTRODUCTION

Layers of glass-fiber-reinforced epoxy composites have traditionally served as insulators between adjacent copper planes in multilayer circuit boards. Recently, however, the decreased thickness of insulating layers has resulted in an increased probability of both interplane current leakage and dielectric heating, especially during stressful temperature/humidity conditions. These problems, which are directly related to the continuing emphasis on miniaturization of electronic components, have necessitated a more complete understanding of composite dielectric properties.

At least four composite dielectric responses may be detected utilizing alternating current impedance techniques over a wide range of measurement frequencies and temperatures. First, near ambient temperatures, measurements at frequencies between 10 and 4 Hz and 10 Hz typically yield values for the bulk insulation resistance of the composite. At intermediate frequencies (10 Hz–1 kHz), a second process, the long chain polar relaxation process, may yield dielectric loss. Third, glass fiber-epoxy interfacial space charge polarization processes may also contribute to measured loss in this frequency range. The dielectric loss intensities corresponding to these processes are strongly dependent upon composite moisture content and measurement temperature. Fourth, dipolar relaxation of polar side groups or chain segments in the perturbing electric field are generally observed at frequencies ranging from 40 Hz at -60°C to 1 MHz at 40°C . We treat only this dipole relaxation process in the present paper.

Although the dielectric relaxation literature of polymers is extensive,^{1,2} relatively little has appeared concerning relaxation processes in the more poorly defined epoxy resins. The influence of moisture on the dielectric relaxation behavior of an imidazole cured epoxy system was reported by Maxwell and Pethrick.³ The results suggest that water exists both as a bound (hydrogen bonded) phase dispersed throughout the resin matrix, and as a free phase which is adsorbed on or within cavities in the matrix. A freezing out or cessation of water molecule motion at temperatures below -35°C was also reported by these authors. The effects of fillers⁴ and copolymer addition⁵ on the dielectric properties of an epoxy system were recently reported by Daly et al. Dipolar relaxation appeared to be altered primarily by polarity changes in the vicinity of the relaxing group. Several workers^{6,7} have utilized dielectric techniques as an analytical measure of water absorption in epoxy resin.

If dipole polarization (P) may be considered to increase and to decay exponentially upon application and removal of a potential step, then for the application

$$P(t) = P_0(1 - e^{-(t/\tau)})$$

$$P(t) = P_0 e^{-(t/\tau)} \quad \text{for removal}$$

where P_0 = polarization at infinite time and τ = time constant for dipole response to step potential, it may be shown that

$$\tau = e^{-(E_a/kT)}$$

where K = Boltzmann constant, T = temperature, E_a = activation energy, and τ_0 = inverse frequency factor. Activation energies and frequency factors obtained using this method are generally useful only for comparison purposes. Physically meaningful frequency factors will be obtained only if the activation energy is independent of temperature or a slowly varying function of temperature. Dielectric loss intensity represented in the frequency domain yields a peak width at half height of 1.17 decades frequency for an ideal Debye relaxation process. In polymer systems much wider non-Debye frequency distributions are normally observed.⁸ Numerous mathematical models, including nonexponential decay functions for cooperative relaxation processes⁹ as well as the Cole-Davidson and Havriliak-Negami functions,¹⁰ have been derived to explain the observed behavior. In epoxy systems a wider relaxation frequency distribution than that normally observed in polymers may result from a less homogeneous distribution of relaxing functional groups or segments and a variety of relaxation environments.

The effects of adsorbed and clustered water on the dielectric properties of numerous polymer systems have been studied.¹¹ Crofton and Pethrick studied the effect of moisture on cellulose fibers.¹² At water concentrations of less than 4%, the loss amplitude associated with polar side group motion was enhanced. Water molecules apparently relaxed in phase with the substrate side groups on which they were adsorbed. Further water adsorption

generally shifted loss peaks at a given frequency to lower temperatures as anticipated for substrate plasticization by the adsorbed water. Antiplasticization (decreased free volume) effects have been observed using cellulose acetate diluents in poly(methyl methacrylate) systems.¹³ Johnson et al.^{14,15} reported a B loss enhancement associated with adsorbed water in polysulfone, polyvinylacetate, and polycarbonate. A well-resolved loss maximum associated with clustered water was also observed in these systems. The effects of plasticization, clustered water, proton migration, adsorbed water, and increased segmental polarity which may accompany moisture uptake in epoxy systems are not well understood.

The kinetics and modes of moisture adsorption in epoxy systems similar to that of the present work have recently been discussed extensively by Apicella et al.¹⁶ Several modes including H-bond formation, diluent-solution, and adsorption in holes may exist for water uptake in epoxy resin. At low moisture activities, data are usually well described by a Henry's law and/or Langmuir isotherm treatment in which adsorbents fill existing matrix sites. At higher activities, deviations from Henry's law are considered a result of moisture clustering.¹⁷ The availability of sites at which moisture H-bond formation occurs may influence adsorption and desorption characteristics as well as the nature of adsorbed water. Irreversible moisture adsorption and non-Fickian diffusion have been attributed to changes in resin chemistry or morphology accompanying adsorption.¹⁸ Some characteristics of moisture diffusion in the epoxy used for this study were recently discussed by Marsh et al.¹⁹ Correlations of dielectric behavior with these adsorption modes and chemical or morphological changes have not been investigated in epoxy resin systems.

This paper presents the dielectric relaxation spectrum of a dicyandiamide-epoxy resin formulation as a function of moisture concentration. Effects of nonpolar diluents, polar solvents, and exposure to acidic and basic solutions will be presented in following reports.

EXPERIMENTAL

Capacitance and dielectric loss were measured from 40 Hz to 1 MHz using commercially available capacitance and impedance analyzers interfaced with a standard laboratory computer. Most measurements were made using an AC amplitude of 1 V RMS. AC amplitude variation from 0.05 to 5.0 V RMS showed that a linear current-voltage relation existed within this range.

Measurements were generally made at 10°C intervals from -80 to 140°C in a standard temperature control chamber. Samples were allowed to equilibrate for 15 min at each temperature prior to measurement. Liquid nitrogen was utilized to achieve measurement temperatures between -80 and -190°C. Environmental control chambers and standard saturated salt solutions were used to equilibrate samples at various relative humidity and temperature conditions.

Measurements were normally performed on resin or composite samples with uniform thickness of approximately 0.3 mm. The samples were firmly held between identical polished aluminum or copper electrodes of 25 cm

area. Edge effects were insignificant as samples always extended well beyond the measurement area between the electrodes.

The resin used was a typical dicyandiamide-epoxy formulation, the chemistry of which is described elsewhere.²⁰ The resin and composite samples were cured at 180°C and 30 psi for 90 min.

RESULTS AND DISCUSSION

The 10 KHz dielectric constant (ϵ) and loss (D) behavior of a dry composite sample are shown in Figure 1 as a function of measurement temperature. The single, widely distributed loss peak which appears within this temperature range is associated with a composite dielectric constant change from $\epsilon = 2.9$ –3.5. No further maxima were detected between -100 and -180°C . At higher temperatures long chain relaxation and DC conductance contributed to measured losses. This loss peak appears in a temperature-frequency range typically associated with polar side group or segmental relaxation in a polymer matrix. We have studied the response of this loss peak (which we assume to be associated with dipole relaxation) to composite moisture uptake.

Figures 2(a) and 2(b) show typical dielectric loss spectra of the dry epoxy resin and its composite, respectively, plotted in the temperature domain at several measurement frequencies. The samples were dried at 130°C, slightly above the epoxy glass transition temperature, for 100 h. Weight gain and loss measurements show that less than 0.01 wt% moisture remains adsorbed following this treatment. For both the resin and its composite some variability of loss behavior was found for identically prepared samples, although the general magnitudes, temperature dependence and shapes were consistent. Differences observed suggest some minor aging or reorientation processes may occur in this epoxy system following the initial cure.

The loss behavior showed complete reversibility during cycling between -80 and 30°C . Rapid quenching to -80°C also generated no abnormal loss behavior or hysteresis. Since processes and properties associated with long

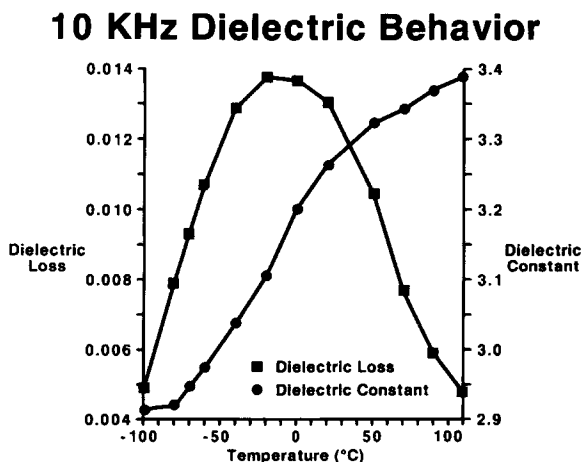
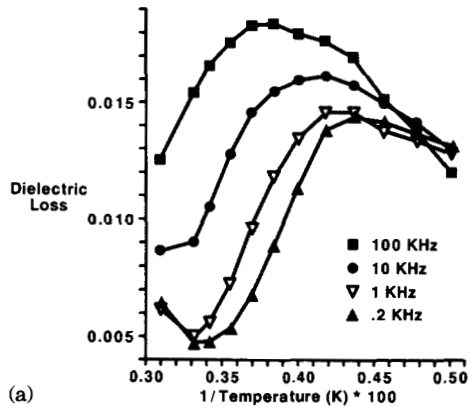


Fig. 1. Ten KHz dielectric loss (□) and dielectric constant (●) as a function of measurement temperature for a composite sample dried at 130°C for 100 h.

Resin Dielectric Loss



Composite Dielectric Loss

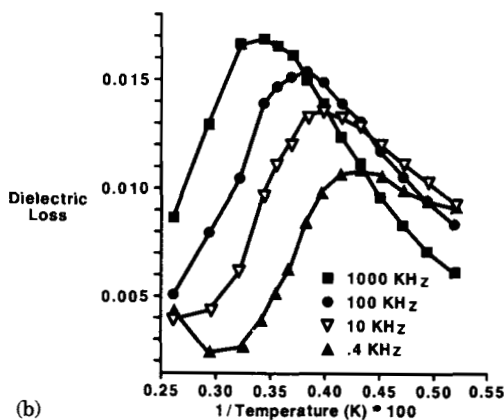


Fig. 2. Temperature domain dielectric loss spectra of composite (a) and resin (b) samples dried at 130°C for 100 h. (a): (□) 100 kHz; (●) 10 kHz; (▼) 1 kHz; (▼) 0.2 kHz. (b): (□) 1000 kHz; (●) 100 kHz; (▼) 10 kHz; (▲) 0.4 kHz.

chain or primary resin structure typically show hysteresis during temperature cycling, these groups are probably not directly involved in the relaxation processes. This supports the contention that the observed relaxation process is due to relatively small functional groups or segments, the properties of which are not strongly dependent on the overall matrix configuration.

An Arrhenius plot of the data in Figures 2(a) and (b) is shown in Figure 3. Relaxation activation energies of 18.6 and 18.1 kcal/mol for the resin and composite, respectively, were calculated from a least squares fit of the data. The frequency factors of 10^{18} obtained from this data are considerably above the physically realistic limit of 10^{13} s. This difference requires that the relaxation activation energy must decrease from the reported value at higher temperatures. The coupling of side group or segmental relaxation to long chain relaxations which become active at higher temperatures has been suggested as a physical explanation of the observed behavior.

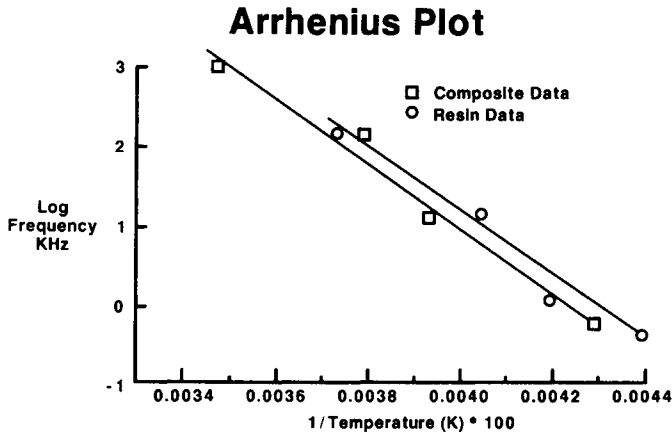


Fig. 3. Arrhenius plots for composite (□) and resin (●) data shown in Figures 2(a) and 2(b).

Since the characteristics of loss behavior in the resin and composite samples are identical, we feel that relaxation may be attributed completely to the resin matrix. The loss intensity of the fiberglass itself was on the order of 10^{-4} over the temperature–frequency range studied. The approximately 35% reduction in relaxation intensity observed in the composite sample simply results from its fiberglass volume fraction. Since the loss magnitude of the fiberglass is small and insensitive to moisture, resin loss behavior in the experimentally convenient composite samples was usually studied.

In the temperature domain (Fig. 2) it is apparent that loss peaks appear to merge to a single curve at lower temperatures, thus suggesting that relaxation intensity becomes distributed over a wider frequency range. The frequency domain loss spectra of the composite sample of Figure 2 is shown in Figure 4 at several measurement temperatures. The unresolved or poorly resolved loss maxima, especially at low temperatures, again reflect a wide distribution of relaxation time constants in the nonhomogeneous epoxy matrix. Both loss maxima, and apparent total capacitance, change from 40

Composite Dielectric Loss

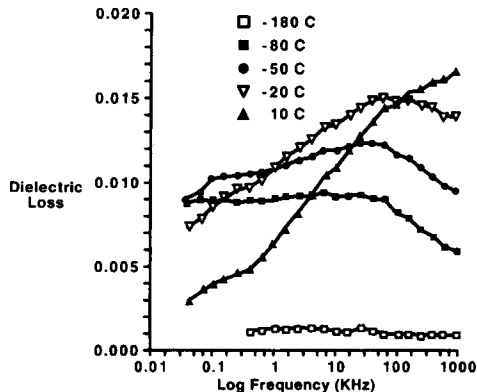


Fig. 4. Frequency domain dielectric loss spectra at several measurement temperatures ($^{\circ}\text{C}$) for a composite sample dried at 130°C for 100 h: (□)-180; (■)-80; (●)-50; (▼)-20; (▲) 10.

Hz to 1 MHz, decreased at lower measurement temperatures. At -180°C the relaxation intensity approached zero over the entire frequency range studied. We believe this behavior indicates a decrease in concentration or "freezing out" of functional groups available for relaxation processes. This freezing out behavior may be analogous to that reported by Maxwell and Petrick.³ However, we observed a gradual decrease in relaxation intensity with decreasing temperature; a distinct drop of relaxation intensity at -35°C was previously reported.³ Using eq. (3), an activation energy for relaxation activity (E_r) may be determined from the temperature dependence of total relaxation intensity (I) (or capacitance change) integrated from zero to infinite frequency.

$$I(T) = I_0 e^{-(E_r/kt)}$$

An E_r of approximately 1 kcal/mol is calculated (Fig. 5) from total relaxation intensities estimated from loss peak intensity and shape using data between 40 Hz and 1 MHz. The physical significance of this activation energy is not clear; however, it may suggest that the resin undergoes changes in its primary structure which allow increased segmental mobility with increasing temperature. As previously described, this behavior is supported by the unrealistically high relaxation frequency factor which was obtained from Figure 2.

Temperature domain relaxation spectra of composite samples equilibrated at 83°C and 60% RH, 83°C and 82% RH, and at 25°C (14.6 mm Hg) are shown in Figures 6(a), 6(b), and 6(c), respectively. These samples contained 0.62, 1.11, and 0.92 wt% water, respectively, following equilibration. Upon uptake of 0.62 wt% moisture, the character of the loss peaks have changed in three ways. First, the relaxation intensity has increased by approximately 75%. Also, loss peak intensity is somewhat less temperature-dependent. Second, the relaxation peaks have become more well defined and no longer merge at lower temperatures. Third, the relaxation activation energy has decreased to 14.1 kcal/mol, and the frequency factor has decreased to $10^{13.5}$ s. The loss peaks at 0.92 and 1.11% water show similar behavior, although DC conductivity or long chain relaxation processes

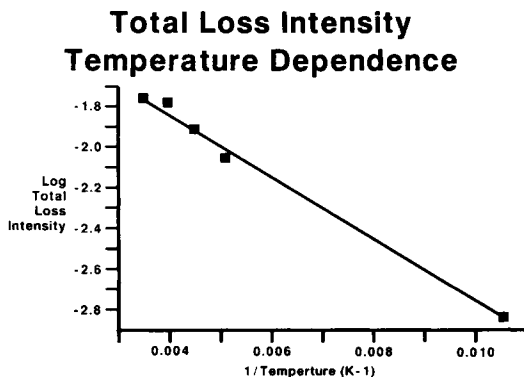


Fig. 5. Log of the estimated total dipole relaxation loss intensity (integrated over infinite frequency) as a function of inverse temperature.

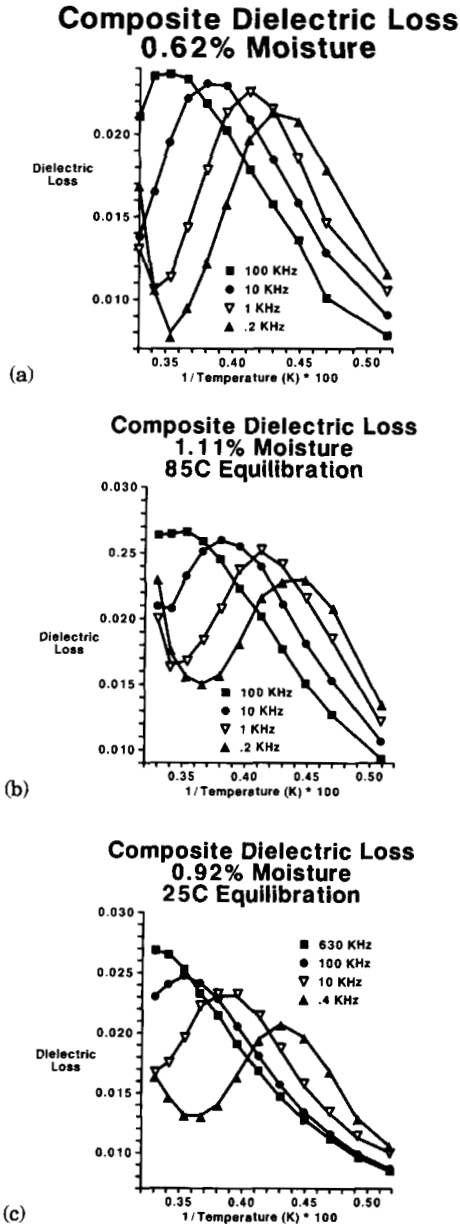


Fig. 6. Temperature domain dielectric loss spectra of composite samples following equilibration at: (a) 83°C and 60% RH (0.62 wt% moisture); (b) 83°C and 82% RH (1.11% moisture); (c) 25°C at 14.6 mm Hg (0.92% moisture). (a,b): (□) 100 kHz; (●) 10 kHz; (▼) 1 kHz; (▲) 0.2 kHz. (c): (□) 630 kHz; (●) 100 kHz; (▼) 10 kHz; (▲) 0.4 kHz.

broaden the peaks by generating a steadily rising loss background at higher temperatures.

Moisture adsorption at concentrations up to 0.1% was frequently observed to decrease relaxation peak amplitudes and apparent integrated total loss intensity. Such behavior was observed during cycling between 130°C drying and equilibration at 0.05% moisture uptake (85°C, 15% RH). Decreases of

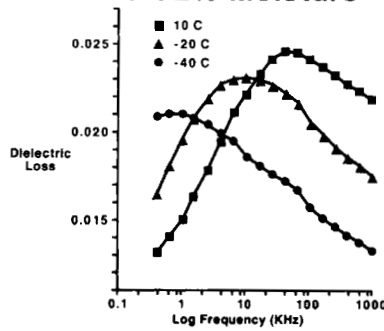
relaxation intensity upon initial moisture adsorption have previously been attributed to pairing of water dipoles with those of substrate adsorption sites such that the total dipole strength is diminished.¹¹ Structural or chemical changes accompanying initial moisture adsorption could also yield the observed behavior. The loss spectra were generally comparable to those of dry composites or resins (Figs. 2 and 3).

In Figures 7 (a)–(d) are shown frequency domain loss spectra of samples equilibrated at 25°C (14.6 mm Hg), 25°C (water submersion), 90°C (183 mm Hg), and 110°C (380 mm Hg). These samples contained 0.92, 1.17, 1.43, and 3.0 wt% water, respectively, following equilibration. Compared to the dry samples, well-resolved frequency domain loss maxima were observed in samples containing between 0.5 and 1.2 wt% water. A reordering of relaxation intensity about a central time constant apparently accompanies moisture uptake. Such reordering of relaxation intensity has previously been reported,¹⁵ and is also frequently observed with increasing measurement temperature (see Fig. 4). A minimum loss peak width at half height of 4–5 decades of frequency was observed for samples containing 0.92% moisture [Fig. 7(a)], still far larger than the ideal 1.17 decade width. Differences this large cannot be accounted for by molecular weight differences among the relaxing groups. A variety of relaxation environments, or a variable coupling of relaxation processes, could lead to the observed peak widths. Frequency domain spectra of samples containing 1.17 and 1.43 wt% water [Figs. 7(b) and 7(c)] show the same relaxation loss characteristics as Fig. 7(a), although the peaks are increasingly obscured by the rising low frequency loss background.

Arrhenius plots of the frequency and temperature domain data of the 0.92% moisture sample [Figs. 6(b) and 7(a)] are shown in Figure 8. Activation energies and frequency factors calculated from the temperature and frequency domain data are in close agreement. It has been reported that frequency and temperature domain data do not necessarily yield equivalent relaxation parameters. However, the close agreement suggests that use of either method provides a comparable measure of the relaxation process in our system. Thus we can compare activation energies over the full moisture adsorption range. At very low moisture concentrations (less than 0.3%), activation energies could be determined only from temperature domain data as peaks were very poorly resolved in the frequency domain. At high moisture concentrations (> 1.2%) the sharply rising loss background at higher temperatures allowed calculation of activation energies only from frequency domain data.

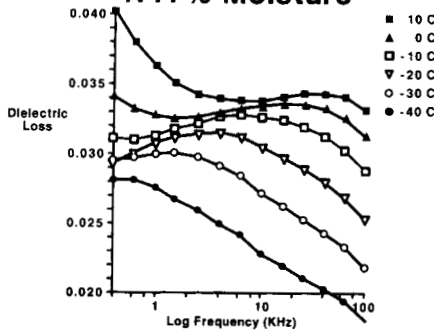
The behavior suggested by Bair et al.¹⁵ in which moisture adsorbs upon and relaxes in phase with polar epoxy segments is consistent with our results obtained at moisture concentrations from 0.5 to 1.5%. According to this argument, increases in relaxation intensity are a result of the larger total segmental dipole moment associated with water adsorption. Slightly lower activation energies (11.3 and 10.7 kcal/mol) and a shift of loss maxima to lower frequencies at a given measurement temperature are observed at 1.17 and 1.43% water concentrations. Such shifts may be attributed to plasticization of the relaxation environment by the adsorbing water molecules.

**Composite Dielectric Loss
0.92% Moisture**



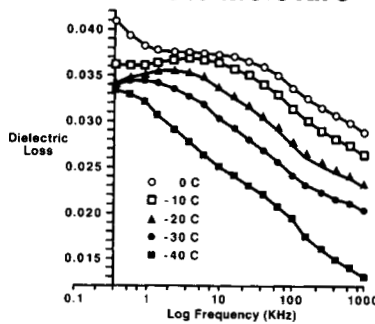
(a)

**Composite Dielectric Loss
1.17% Moisture**



(b)

**Composite Dielectric Loss
1.43% Moisture**



(c)

Fig. 7. Frequency domain dielectric loss spectra of composite samples following equilibration at: (a) 25°C and 14.6 mm Hg (0.92% moisture); (b) 25°C water immersion (1.17% moisture); (c) 90°C and 183 mm Hg (1.43% moisture); (d) 110°C and 380 mm Hg (3.0% moisture). Temperatures (°C): (a): (□) 10; (▲) -20; (▼) -40. (b): (□) 10; (▲) 0; (□) -10; (▼) -20; (○) -30; (●) -40. (c): (○) -40; (□) -30; (▼) -20; (●) -10; (■) 0. (d): (□) 0; (●) -20; (□) -40; (▲) -60; (▼) -80.

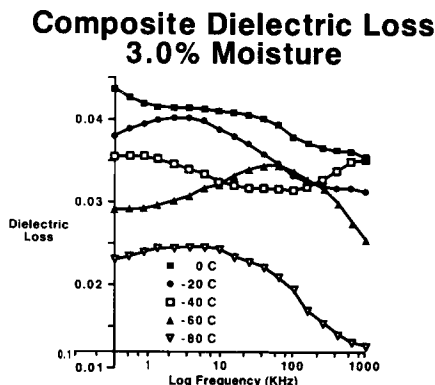


Fig. 7. (continued from previous page)

The composite relaxation spectra observed following 3.0% moisture uptake [Fig. 7(d)] shows both the loss peak observed at lower moisture concentrations and a new peak which occurs at approximately 3 orders of magnitude higher frequency for a given temperature. The new loss peak is of similar intensity to the former, suggesting it is associated with a substantial moisture volume. It appears that a new moisture-composite phase, with considerably different relaxation properties, is formed by moisture concentrations in excess of approximately 1.5%. The onset of colloidal water formation within the epoxy matrix may be indicated by the appearance of this loss peak. The original loss peak remained in the same frequency-temperature region and decreased slightly in activation energy to 8.9 kcal/mol. The loss intensity increased only marginally over that observed at 1.43% water. A saturation of water adsorption sites on the epoxy matrix by approximately 1.5 wt% water, and a corresponding halt to the increasing dipole character of the relaxing segments would explain the observed behavior. No simple correlation between loss peak amplitudes and adsorbed moisture concentration was observed, although amplitudes did generally increase with increasing moisture uptake. The dual phase nature

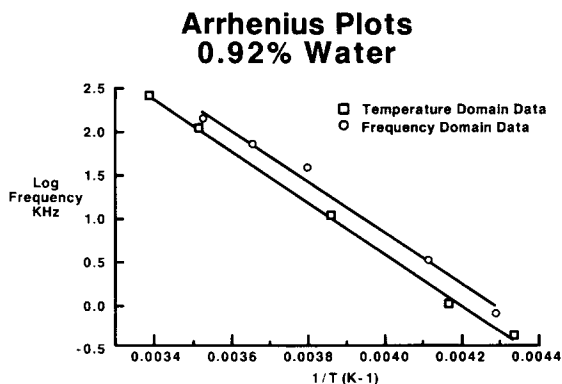


Fig. 8. Arrhenius plots constructed from temperature (□) and frequency (●) domain data [Figs. 6(c) and 7(a)] for composite samples containing 0.92% moisture.

of water observed in the present system is consistent with that observed and discussed by Maxwell and Pethrick.³ Our work suggests that a hydrogen-bonded water phase dispersed throughout the resin matrix forms preferentially to a colloidal moisture phase, with the colloidal phase becoming important only at moisture concentrations greater than approximately 1.4%. The dependence of relaxation activation energies and frequency factors upon moisture concentration are summarized in Figure 9. Activation energies and frequency factors generally decrease with increasing moisture adsorption (0.1–3.0%). Minimum value of approximately 9 kcal/mol and 10^8 s^{-1} . The pattern is consistent with plasticization of the relaxation environment by the adsorbing moisture. However, a more conclusive interpretation of the data is difficult. An activation energy which decreases with increasing temperature is a logical source of the observed behavior. Such an activation energy will lead to smaller frequency factors using activation energies calculated from higher temperature data. In this model the plasticizing effect of moisture is considered to lead to a shift of the relaxation response for a moist sample to a higher effective temperature on an Arrhenius plot, compared to that used to determine the activation energy and frequency factor of a dry sample. Dielectric constants measured at relatively high frequencies and low temperatures will not reflect the contribution of dipolar relaxation processes, while those measured at relatively low frequency and high temperature will reflect these processes. In our system, a dielectric constant independent of moisture concentration was obtained at -80°C and 1 MHz, suggesting no dipolar relaxations are active under these conditions. At 100 Hz and 30°C , loss spectra suggest all polar segmental or small group relaxations are active. The difference between dielectric constants obtained under these conditions will, therefore, be a measure of total dipole activity contributing to the loss process.

The changes in composite dielectric constants from values obtained at 1 MHz and -80°C to those obtained at 100 Hz and 30°C are shown as a function of moisture concentration in Figure 10. A line calculated assuming full moisture dipole contribution to the loss process is also indicated. For the dry composite a relatively small dielectric constant change, reflecting the

Relaxation Rate Parameters

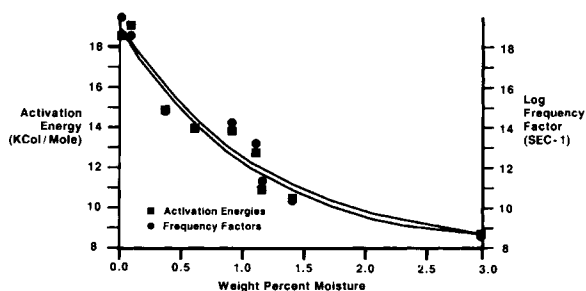


Fig. 9. Activation energies (\square) and frequency (\bullet) factors determined from Arrhenius plots as a function of composite moisture concentration.

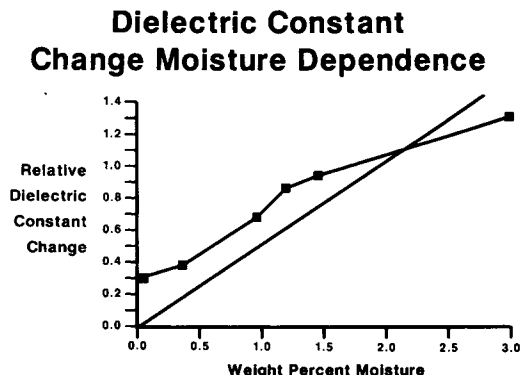


Fig. 10. Total change in composite dielectric constant associated with small molecule, segmental, or side group dipole relaxation processes as a function of composite moisture concentration: (—) the magnitude of dielectric constant change anticipated from 100% moisture dipole participation in the relaxation process.

presence of polar epoxy functional groups, is observed. At low moisture concentrations ($< 0.3\%$) the change is smaller than that anticipated from full moisture dipole participation in relaxation. Some cancellation of effective water dipole strength by adsorption at preferential polar epoxy matrix sites is again suggested. From 0.3 to 1.4% moisture, dielectric constant changes parallel those anticipated from free water relaxation and, therefore, suggest full moisture dipole activity of this moisture volume fraction in the relaxation process. This implies moisture in this concentration range adsorbs at nonpolar epoxy sites or in a way such that its dipole character is not cancelled. At moisture concentrations above 1.4%, the smaller dielectric constant changes indicate that the previously mentioned phase which is formed at moisture concentrations above 1.4% allows only approximately 50% moisture dipole relaxation activity. The nature of this behavior is unclear, but could be associated with colloidal water formation.

CONCLUSION

This study of the dielectric relaxation behavior of a glass-epoxy composite has suggested three modes of moisture interaction with, or adsorption by, the composite. At very low moisture concentration ($< 0.1\%$) decreased loss intensity suggests resin chemistry changes or adsorption involving dipole pairing, and a decrease in total dipole strength. At higher moisture concentrations (0.1–1.4%) increased loss intensity suggests dipoles relax in phase with the segments upon which they adsorb, thus increasing their effective dipole strength. Decreasing relaxation activation energies and increasingly well-defined loss peaks indicate a resin plasticization effect is associated with moisture uptake. At higher moisture concentrations ($> 1.4\%$), the appearance of a second loss peak may correspond to formation of colloidal or weakly adsorbed moisture as well as indicating saturation of primary adsorption sites. The wide frequency distribution of relaxation losses suggests the absence of a unique polar functional group or segment

responsible for the observed losses. Variation of resin chemistry, and relaxation studies involving other polar and nonpolar adsorbing molecules, may yield a better understanding of the nature of observed relaxation losses.

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