

Dielectric Properties of Industrial Polymer Composite Materials

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Abstract

Frequency and temperature dependence of dielectric constant ϵ' and dielectric loss ϵ'' in pure polyester resin and polymer composites with various types of glass fiber are studied in the frequency range 330 Hz–3 MHz and in the temperature range 25–150 °C. The experimental results show that ϵ' and ϵ'' increased with the addition of glass fiber in polyester resin. The value of ϵ' decreased with increasing frequency, which indicates that the major contribution to the polarization comes from orientation polarization. Dielectric loss peaks were also observed in the composite materials at high temperature due to T_g of polyester. The value of ϵ' increased with increasing temperature, and is due to greater freedom of movement of the dipole molecular chains within the polyester at high temperature.

Key Words: Composite materials, orientation polarization, dielectric relaxation, polyester resin.

1. Introduction

It is well known that composites can be produced exhibiting enhanced properties that the constituent materials may not exhibit [1–4]. For instance, from the combination of different fibers or fillers with polymer matrices one can produce polymer-matrix composites, a material important to the electronic industry for its dielectric properties in the use of capacitors [5–7]. The effective utilization of filled polymers depends strongly on the ability to disperse the fillers homogeneously throughout the matrix [8]. The interface properties also strongly affect the characteristics and performance of these composites [9]. One of the most attractive features of these filled composites is that their dielectric properties can be widely changed by choice of shape, size, and the conductivity of filled constituents in the polymeric matrix.

Most of the interesting properties of polymers are attributable to the complex motions within their molecular matrix. In the polymeric system, molecular relaxations exhibit various transitions [10]. As very little work has been reported on double-layer systems, the intention in the present work is to study dielectric properties of such a system. The polymeric interfaces act as charge-carrier trapping sites [11]. Therefore, it has become essential to study the effect of interfaces on the charge-carrier generation, transport and storage in polymeric systems. The study of dielectric constant and dielectric loss, as a function of temperature and frequency is one of the most convenient and sensitive methods of studying polymeric structure.

For polymer composites in the solid or viscoelastic state, the physical structure is of great importance in determining the dielectric behavior [12]. The dielectric properties of polymer composite materials have been studied with a view to modifying the properties of polymer systems for practical applications. The conventional inorganic insulators and dielectrics have to a large extent been replaced by polymers on account of their unique ability to be tailor made for specific needs. Epoxies and polyesters have been used in electronics as insulators, dielectrics, substrates, potting compounds, embedding materials and conformal coatings [13].

Industrial polymer composite materials are developed by using the combination of polyester resin and different types of glass fiber (i.e. E, C and S-glass fiber). Unsaturated polyester resins are the most widely used thermosetting matrices for reinforced plastics, spanning the whole range from basic hand lay-up to complex mechanized molding processes. Polyesters offer a good balance of mechanical, electrical and chemical resistance properties at relatively low cost [14]. Glass fiber is the most widely used reinforcing material both for thermosetting and thermoplastic composites. It has high tensile strength combined with low extensibility, giving exceptional tensile, compression and impact properties.

It has high temperature resistance and low moisture pick up, giving good dimensional stability and weather resistance. Finally, low moisture absorption makes it possible to produce molding with good electrical properties which do not deteriorate, even under adverse weather conditions [15].

There are many types of glass fiber, with the most common being E-glass fiber, C-glass fiber and S-glass fiber. E-glass fiber is the most common in use, because it draws well and has good tensile and compressive strength and stiffness, and good electrical and weathering properties. C-glass fiber has higher resistance to chemical corrosion than E-glass but is more expensive and has lower strength properties. S-glass fiber, developed to meet demand for higher technical performance from the aerospace and defense industry [16], has higher tensile strength and modulus but is more expensive than C and E-glass fiber. Fiber reinforced composite materials have a wide range of applications in aircraft automobile, chemical, medical and electrical industries. In the electrical/electronics industry, these composite materials are used for making panels, switches, insulators and for many insulation purposes. So by way of industrial expectation, all materials will need to be characterized for and prior to such applications.

The electrical response of a normal dielectric can be described by its conductance, dielectric constant and loss factor. Typical compositions of the three well known glasses used for glass fiber in composites are given in Table [17].

Table. Composition of glass used for fiber manufacture (all values in wt%).

| Composition | E-Glass | C-Glass | S-Glass |
|---|---------|---------|---------|
| SiO ₂ | 52.4 | 64.4 | 64.4 |
| Al ₂ O ₃ , Fe ₂ O ₃ | 14.4 | 4.1 | 25.0 |
| CaO | 17.2 | 13.4 | — |
| MgO | 4.6 | 3.3 | 10.3 |
| Na ₂ O, K ₂ O | 0.8 | 9.6 | 0.3 |
| Ba ₂ O ₃ | 10.6 | 4.7 | — |
| BaO | — | 0.9 | — |

2. Experimental Detail

Polyester resin and different types of glass fiber (E, C and S-type fiber) were obtained from Fiber Tech Composite Industry, Lahore (Pakistan) and samples of composite materials were prepared in Materials Research Laboratory, Pakistan Council for Scientific and Industrial Research (PCSIR).

A compression moulding process was used for making the composite materials. A compression moulding process is a low temperature process in which fiber is saturated with polyester resin and then pressed between matched dies. Disc-shaped samples of 3 cm diameter and 2.5 mm thickness were prepared by uniaxial pressing at 5 ton. Four samples, Sample A (pure polyester resin), sample B (polyester + 50% E-glass fiber), Sample C (polyester + 50% C-glass) and sample D (polyester + 50% S-glass) were prepared under the same conditions of temperature and pressure.

Dielectric constants ϵ' and loss factors ϵ'' were measured in the frequency range, 330 Hz to 3 MHz and temperature range 25 °C to 150 °C in all the four samples. The dielectric measurements were made using an Ando TR-10C dielectric loss tester in combination with an Ando BDA-9 null detector and a WGB-9 Oscillator [18]; all brought together into the form of the Ando TRS-10T dielectric loss measurement system. Measured quantities were the capacitance C_x and the conductance G_x of the specimen, from which the dielectric constant ϵ' and dielectric loss factor ϵ'' were calculated as [19]

$$\epsilon' = \frac{C_x}{C_a} \text{ and } \epsilon'' = \frac{G_x}{\omega C_a},$$

where C_a is the capacitance of the cell with air and ω is angular frequency of observation.

3. Result and Discussion

Figure1 gives the variation of the dielectric constant ϵ' with frequency at room temperature for all four samples. At room temperature, marked differences were found in dielectric constant ϵ' between polyester resin and composite materials with different types of glass fiber. An important observation is that ϵ' increases considerably with the addition of glass fiber in polyester resin, which is most likely explained by the glass having a higher dielectric ϵ' than the base polyester resin, thus resulting in the higher dielectric constant of the composites [20]. One further effect may contribute to the rise in dielectric constant: the absorption of moisture at the fiber resin interface, as the dielectric of water is very high.

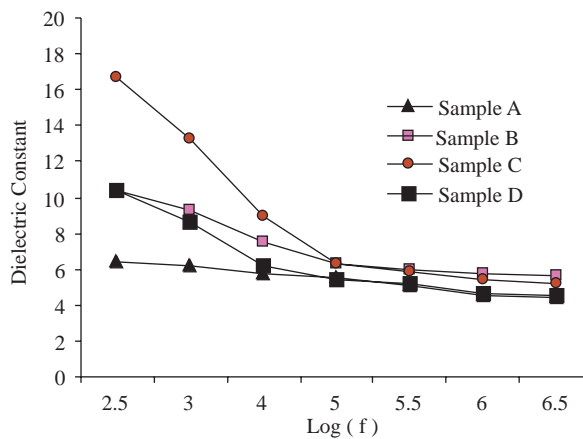


Figure 1. Frequency dependence of dielectric constant at room temperature for sample A, sample B, sample C, and sample D.

Figures 2(a–d) shows the variation of ϵ' with frequency at different temperatures for samples A, B, C and D, respectively. It is evident from these figures that ϵ' decreases with increasing frequency at fixed temperature. It is also evident from these figures that the decrease in ϵ' is very prominent at both low frequencies and at high temperature. The decrease of ϵ' with increasing frequency is the expected behavior

in most dielectric materials. This is due to dielectric relaxation which is the cause of anomalous dispersion. From a structural point of view, the dielectric relaxation involves the orientation polarization which in turn depends upon the molecular arrangement of dielectric to be material. So, at higher frequencies, the rotational motion of the polar molecules of dielectric is not sufficiently rapid for the attainment of equilibrium with the field, hence dielectric constant seems to be decreasing with increasing frequency [20].

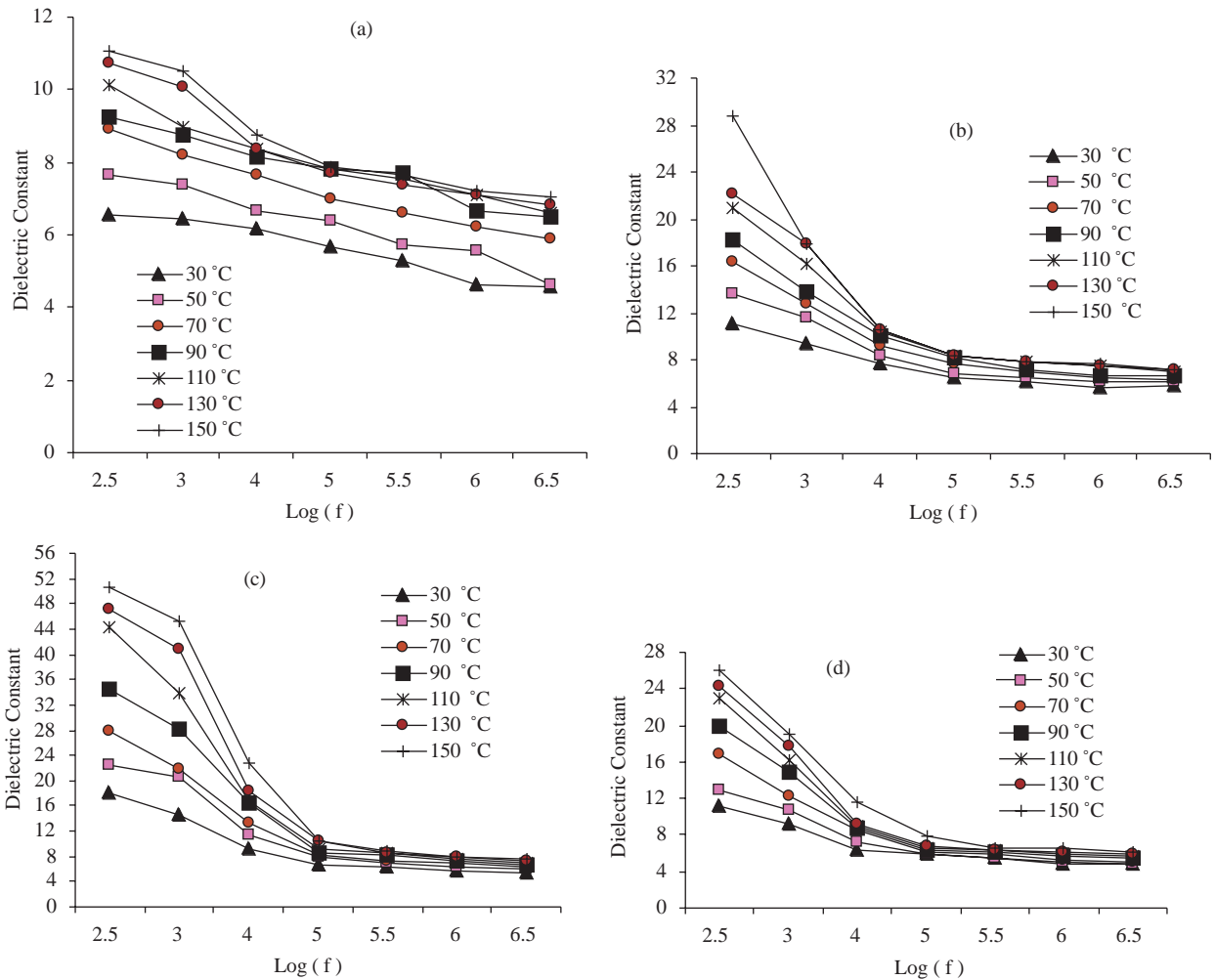


Figure 2. Frequency dependence of dielectric constant at different temperatures for (a) sample A, (b) sample B, (c) sample C, and (d) sample D.

Figures 3(a–d) show the variation of ϵ' with temperature at different frequencies for samples A, B, C and D, respectively. The value of ϵ' increases with temperature at fixed frequency. This is true for all the samples at all frequencies. At lower frequencies this effect is more prominent. The increase in ϵ' with temperature is due to greater freedom of movement of dipole molecular chain of polyester at high temperature. At lower temperature, as the dipoles are rigidly fixed in the dielectric, the field can not change the condition of dipoles. As the temperature increases, the dipoles comparatively become free and they respond to the applied electric field. Thus polarization increased and hence dielectric constant is also increased with the increase of temperature [21].

Figures 4(a–d) show the variation of dielectric loss factor ϵ'' with frequency at different temperatures for all the four samples. The value of ϵ'' decreases with the increase of frequency at fixed temperature in all the four samples. But loss peaks are observed at about 1 KHz at high temperature (150 °C) in composite materials, which may be attributed to T_g , the temperature glass transition in polyester [22]. This effect can be seen in Figures 4(b–d). The loss peak is not observed in pure polyester, as shown in Figure 4(a); it may lie at lower frequency than the lowest observable frequency of our measuring set. In composite materials the absorbed moisture at the fiber-resin interface acts as a plasticizing agent [23] for the polymer, which increases the mobility of the polymer chain and hence brings the loss peak due to T_g of polyester at high frequency value to be detected by our measuring set.

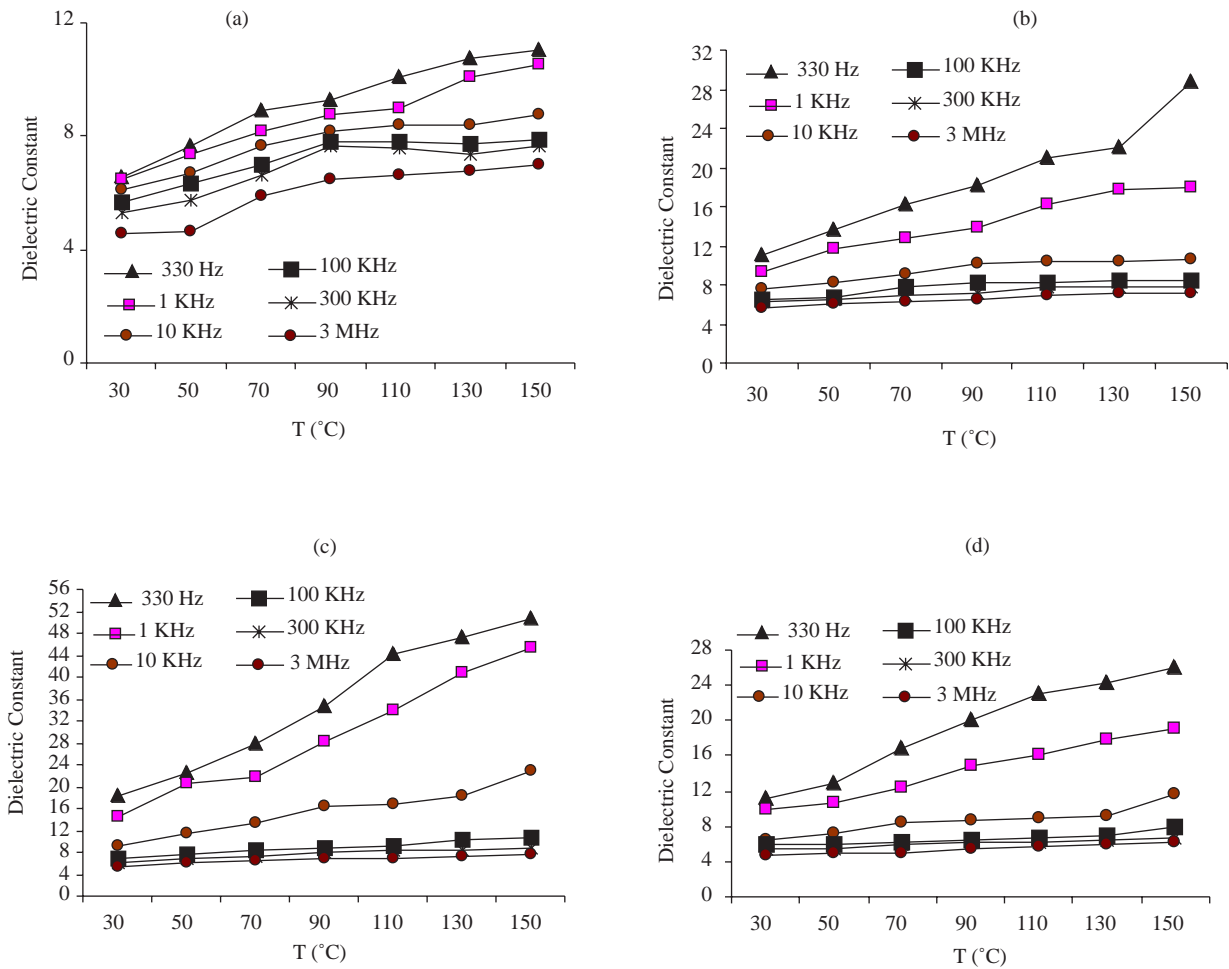


Figure 3. Temperature dependence of dielectric constant at different frequencies for (a) sample A, (b) sample B, (c) sample C, and (d) sample D.

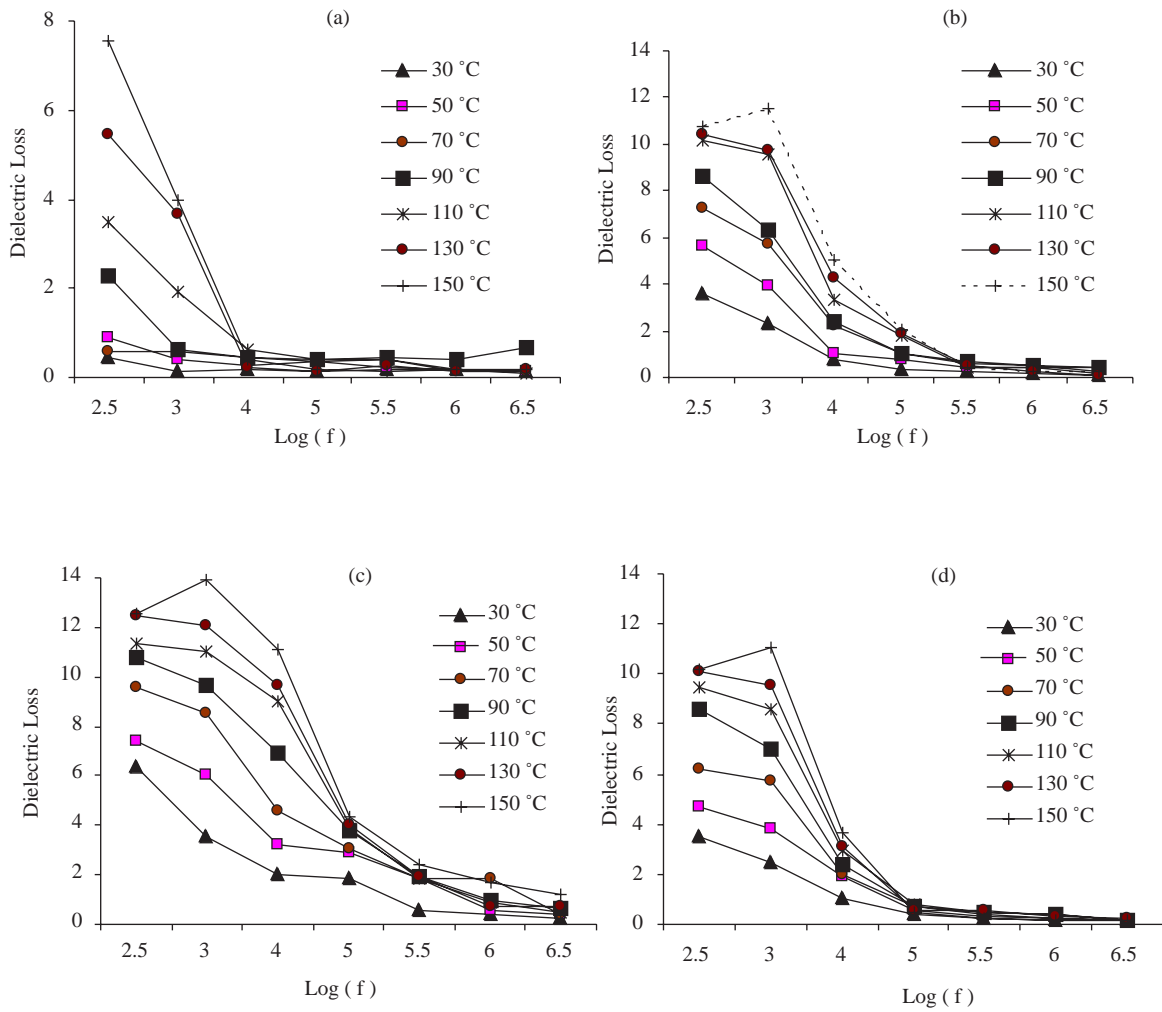


Figure 4. Frequency dependence of dielectric loss at different temperatures for (a) sample A, (b) sample B, (c) sample C, and (d) sample D.

Figures 5(a–d) show the variation of dielectric loss factor with temperature at different frequencies for all four samples. The dielectric loss factor increase with temperature, particularly at lower frequencies at which dielectric loss due to chain motion of polyester is more effective due to the glass transition temperature of the polymer. At high frequencies, however, the dielectric loss factor is low and remains more or less constant with increasing temperature because the orientation polarization due to chain motion of polymer can not keep phase with the rapidly oscillating electric field.

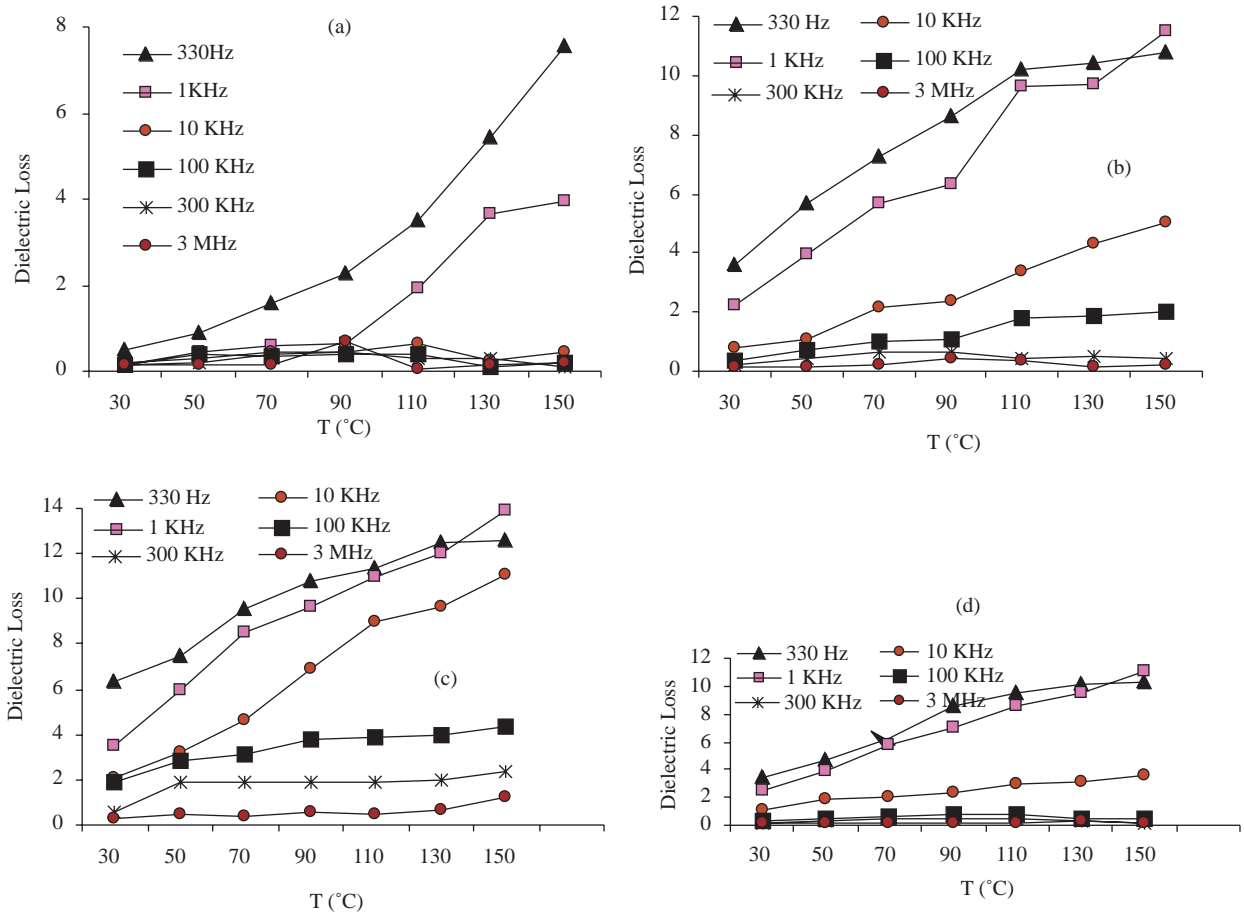


Figure 5. Temperature dependence of dielectric loss at different frequencies for (a) sample A, (b) sample B, (c) sample C, and (d) sample D.

4. Conclusion

Frequency and temperature dependence of dielectric constant ϵ' and dielectric loss ϵ'' in pure polyester resin and polymer composites with various types of glass fiber has been studied in the frequency range 330 Hz–3 MHz and in the temperature range 25–150 °C. The experimental results indicate that ϵ' and ϵ'' increased with the addition of glass fiber in polyester resin. The decrease in ϵ' with increase of frequency is due to the orientation polarization and increase in ϵ' with increase of temperature is due to greater freedom of movement of dipole molecular chain of polyester at high temperature. Dielectric loss peaks in the composite materials at high temperature is due the glass transition temperature of polyester.

References

- [1] Ronald J. Gutmann, *IEEE Trans. Microwave Theory Tech.*, **47**, (1999), 667.
- [2] Nobuyoshi Awaya, Hiroshi Inokawa, Eiichi Yamamoto, Yukio Okazaki, Masayasu Miyake, Yoshinobu Arita, and Toshio Kobayashi, *IEEE Trans. Electron Devices*, **43**, (1996), 1206.
- [3] *The National Technology Roadmap for Semiconductors* (Semiconductor Industry Association, San Jose, California, 2001).

- [4] J. H. Golden, C. J. Hawker, and P. S. Ho, *Designing Porous Low-k Dielectrics* (Semiconductor International, 2001).
- [5] N. Aoi, *Jpn. J. Appl. Phys., Part 1* **36**, (1997), 1355.
- [6] W. Wu, W. E. Wallace, E. K. Lin, G. W. Lynn, C. J. Glinka, E. T. Ryan, and H. M. Ho, *J. Appl. Phys.*, **82**, (2000), 1193.
- [7] M. H. Jo, H. H. Park, D. J. Kim, S. H. Hyun, S. Y. Choi, and J. T. Paik, *J. Appl. Phys.*, **87**, (1997), 1299.
- [8] D. R. Denison, J. C. Barbour, and J. H. Burkart, *J. Vac. Sci. Technol.*, **A14**, (1996), 1124.
- [9] M. Tada, Y. Harada, K. Hijioka, H. Ohtake, T. Takeuchi, S. Saito, T. Onodera, M. Hiroi, N. Furutake, and Y. Hayashi, *IEEE International Interconnect Technology Conference*, (2002), p. 12.
- [10] P. K. C. Pillai, G. K. Narula and A. K. Tripathi, *Polym. J.*, **16**, (1984), 575.
- [11] A. K. Tripathi and P. K. C. Pillai, *Proceedings of the 5th International Symposium on Electrets*, Heidelberg (IEEE, New York. 1985).
- [12] Leyla Aras and Bahattin M. Baysal, *J. Polym. Sci. Polym. Phys. Edn.*, **22**, (1984), 1453.
- [13] D. B. Singh, A. Kumar, V. P. Tayal and B. Sanyal, *Journal of Materials Science*, **23**, (1988), 523.
- [14] Milewski J. V, *Hand book of Fibers and reinforcement*, Van Norstrand Reinhold, (New York. 1979).
- [15] John Murphy, *The reinforced plastic Hand book*, 2nd ed, (Elsevier Science Ltd, UK. 1984).
- [16] G. Lubin, *Hand book of fiber glass and advanced composites*, Van Norstrand Reinhold, (New York. 1982).
- [17] D. Hull, *An introduction to composite materials*, (Cambridge University press, Cambridge. 1981), p. 16.
- [18] Instruction Manual for type TR-10C, Dielectric Loss Measuring set, Ando Electric Co. Ltd., Japan, (1982).
- [19] Instruction Manual for type SE-70, Electrode Ando Electric Co Ltd., Japan (1982).
- [20] C. P. Smyth, *Dielectric behavior and Structure*, McGraw Hill, (New York. 1955), p. 53.
- [21] H. Frohlick, *Theory of Dielectrics*, Oxford University press, (Oxford. 1956), p. 13.
- [22] Mateen A, PhD Thesis, Centre for Solid State Physics , University of the Punjab, Lahore, Pakistan, (1983).
- [23] E. Polac, *Epolac Technical data Bulletin*, Nippon Shokubai Kagaku, Kogyo, Co. Ltd., (Japan. 1985).