COMMUNICATIONS

UNDERSTANDING THE MECHANICAL PROPERTIES OF EPOXY NANOCOMPOSITE INSULATING MATERIALS

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In recent times, epoxy nanocomposites are used as electrical insulating materials. It is essential to understand the mechanical properties of such electrical insulating materials for proper design of insulation structure of power apparatus. Fundamental characteristics of epoxy nanocomposites are analysed through wide angle X-ray diffraction (WAXD) studies. The results of the study indicates that intercalated structures are formed above 5 wt % of clay in epoxy resin. Hydrophobic characteristics are analyzed through contact angle measurement. Characteristic variations in material properties of epoxy nanocomposites due to water ageing are analysed through vibration studies. Vibration test results indicate that natural frequency of vibration and damping factor of the material increases by adding up to 5 $wt\%$ of nano clay to base resin.

K e y w o r d s: nanocomposites, epoxy resin, insulation, vibration test, contact angle

1 INTRODUCTION

With the advancement in power transmission level, it has become essential to design and develop compact insulating materials. Polymer nanocomposites are emerging as a new class of materials for its demanding applications as insulating material [1-4]. During operation of power equipment, the insulation is not only subjected to electrical stress but also to thermal stress due to heat generated by the current carrying conductor and mechanical stress due to force exerted by the flow of high current and/or vibration stress arise from the mechanical movement of electrical machines causing multi-stress ageing of insulating material. However, because of the polymer matrix, the insulating material must withstand the mechanical strength and tribological loads, it is usually reinforced with fillers forming composite material. *Nanocomposites* are named when the disperse phase particle size is less than 100 nm. Reinforcement of polymeric resin with nanoclay platelets as fillers has resulted in light weight materials with increased modules and strength, decreased permeability, less shrinkage with increased heat resistance.

Epoxy resin is an indispensable material for insulation systems in power equipment such as dry type transformers and rotating machines. The epoxy resin is used not only as insulating material but also as structural material because it is cost-effective. In recent times epoxy resin added with organically modified clay as filler finds major application as insulating material [5–9]. The researchers in the world over are trying to understand the electrical, thermal and mechanical properties of epoxy nanocomposites insulating materials and the database is scanty.

Having known all this, in the present work, it is planned to carry out a methodical experimental study, to evaluate the performance characteristics of the epoxy nanocomposite, an epoxy resin containing different proportion of Montmorillonite (MMT) clay. The results of the study were compared with epoxy resin without filler content. The basic characteristics of the epoxy nanocomposite were analyzed through certain physico-chemical diagnostic techniques especially through Wide angle X-ray Diffraction (WAXD) studies. Contact angle (θ) measurement was used as a measure of hydrophobicity of the material. Water aging was carried out to understand the mechanical properties of the material through vibration tests.

2 EXPERIMENTAL STUDIES

Epoxy nanocomposite was synthesized by mechanical shear mixing of organo clay in the resin bath at room temperature. The clay mineral used in this study was organophillic MMT procured from southern clay products Inc. (Gonzales, Texas) under the trade name of garamite 1958 [10]. After uniform mixing of clay particles in epoxy resin (DGEBA, CY205, Giba-Geigy Inc), Tri Ethylene Tetra Amine (TETA) hardener is added and then cast in a mold of required dimensions. Montmorillonite (MMT) clay belongs to a family of 2:1 layered silicates. The crystal structure of MMT clay consists of two fused silica tetrahedral sheets sandwiching an edge shared octahedral sheet of either aluminium or magnesium hydroxide. Epoxy nanocomposites with different percentage of clay ($\sim$1–10$wt\%$) were prepared. The vibration test test were carried out with epoxy resin and with 1,3,5 and 10 $wt\%$ of nanoclay included epoxy nanocomposite material. The

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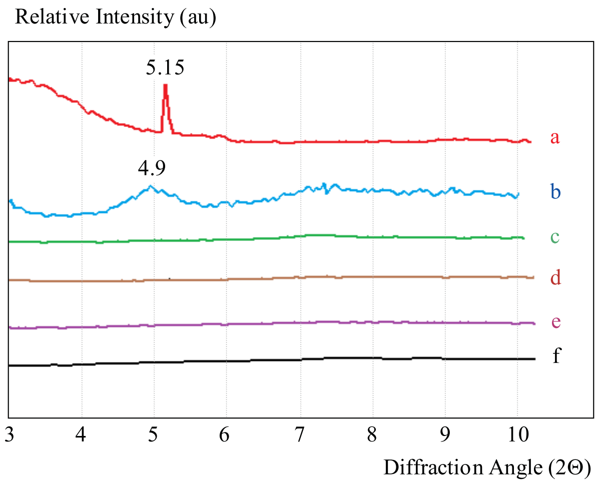

Fig. 1. WAXD Pattern of Epoxy NanoComposite: (a) clay, (b) 10 *wt*%, (c) 5 *wt*%, (d) 3 *wt*%, (e) 1 *wt*%, (f) epoxy

size of the samples which are used for water ageing were of $3 \times 3 \times 0.3$ cm.

WAXD measurements were done using Phillips X-ray diffractometer. In the present work, the free vibration test was carried out to understand the natural frequency and the damping factor of the epoxy nanocomposite.The dimension of the specimen used for carrying out vibration test followed as specified by the standards. The first four modes of natural frequency of vibration obtained by carrying out a free vibration test. The accelerometer (Bruel and Kjaer 437) is used to measure the displacement through the charge amplifier and the response of the system measured using a dynamic signal analyzer (Agilant 35670).

3 RESULTS AND DISCUSSION

3.1 WAXD Studies of epoxy nanocomposite

WAXD pattern of the epoxy nanocomposites is shown in Fig. 1. It is observed that MMT clay shows a peak at 5.2° of the 2θ value correspond to the d spacing of 17Å, which is attributed to the reflection from the d_{001} plane (Fig. 1a). It is observed that neat epoxy and the epoxy nanocomposite (up to 5 *wt*% of nano clay, Fig. 1(c–f)) did not show any sharp diffraction peaks in the WAXD pattern, indicating that the structure is exfoliated [8]. As the weight percentage is increased above 5 %, the diffraction patterns show the formation of intercalated nanocomposites (Fig. 1b).

3.2 Hydrophobicity Characterization of Epoxy Nanocomposite

The measure of contact angle of a water droplet is an indirect measure of hydrophobicity of the material. In the present work, the static contact angle was measured by liquid droplet method [13]. The size of the drop was about 1.5mm in diameter. The contact angle is measured using the following equation

$$\theta = 2\tan^{-1}\Big(\frac{2h}{d}\Big) \tag{1}$$

where d is the diameter of the liquid drop and h is the height of the liquid drop. After the solution is placed over the surface of the specimen, the contact angle (θ) is measured within 5 seconds. For each specimen, the contact angle was measured at six different locations and average of it considered as the contact angle. Fig. 2 shows the variation in contact angle of the specimen aged in water at different temperatures with time. It is observed that increase in aging time of the specimen shows a reduction in the contact angle, irrespective of temperature of the water bath. It is also noticed that the contact angle variation is less as the temperature of the water bath is increased. The cause for reduction in contact angle could be due to dissolvement of compounds and the particles which are not properly adhered to the base resin. Comparing the contact angle of pure epoxy resin with epoxy nanocomposite, it is observed that epoxy nanocomposites have high contact angles. Also, increase in weight percentage of nano particle in epoxy resin shows increase in contact angle, indicating an increase in hyrdophobicity of the material.

Figure 3 shows a weight gain in epoxy/epoxy nanocomposites due to ageing of specimen in water at room temperature. In the present work, the increase in weight of the specimen measured based on the ratio between increase in weight of specimen (difference between final weight of the specimen and the initial weight) and the initial weight of the specimen. When the material is aged in water, this would result in mass change and the rate of absorption will be initially linear with $t^{0.5}$, where t is the time of absorption. Hence

$$\frac{\Delta m(t)}{\Delta m_\infty} = 2\sqrt{\frac{Dt}{l^2}}\Big\{\sqrt{\frac{1}{\pi}} + 2\sum_{n=1}^{\infty}\Big[\Big((-1)^n \operatorname{ierfc}\Big(\frac{nl}{2\sqrt{Dt}}\Big)\Big]\Big\} \tag{2}$$

where D is the diffusion coefficient, l is the thickness of sheet, n is the number of sheets ierfc is the integrated complimentary error function, $\Delta m(t) = m(t) - m(0)$ and $\Delta m_\infty = m_\infty - m(0)$. In this, $m(t)$ is the mass at time t, $m(0)$ and m_∞ are the initial mass (at time $t = 0$) and after infinite time, respectively. Crank [11] formulated the final equation to calculate diffusion coefficient as

$$D = \frac{\pi L_{0.5}^2}{64 t_{0.5}} \tag{3}$$

where L is thickness of the specimen. Table 1 shows the variation in the diffusion coefficient of the material aged in water at different temperature. It is observed that an increase in ageing temperature relatively increases the diffusion coefficient of the material. Also increase in percentage of nanofiller in epoxy shows a reduction in diffusion coefficient of the material, for a given temperature. This indicates that the epoxy nanocomposite is hydrophobic in nature with increase in percentage content of nano filler in the base resin material [12].

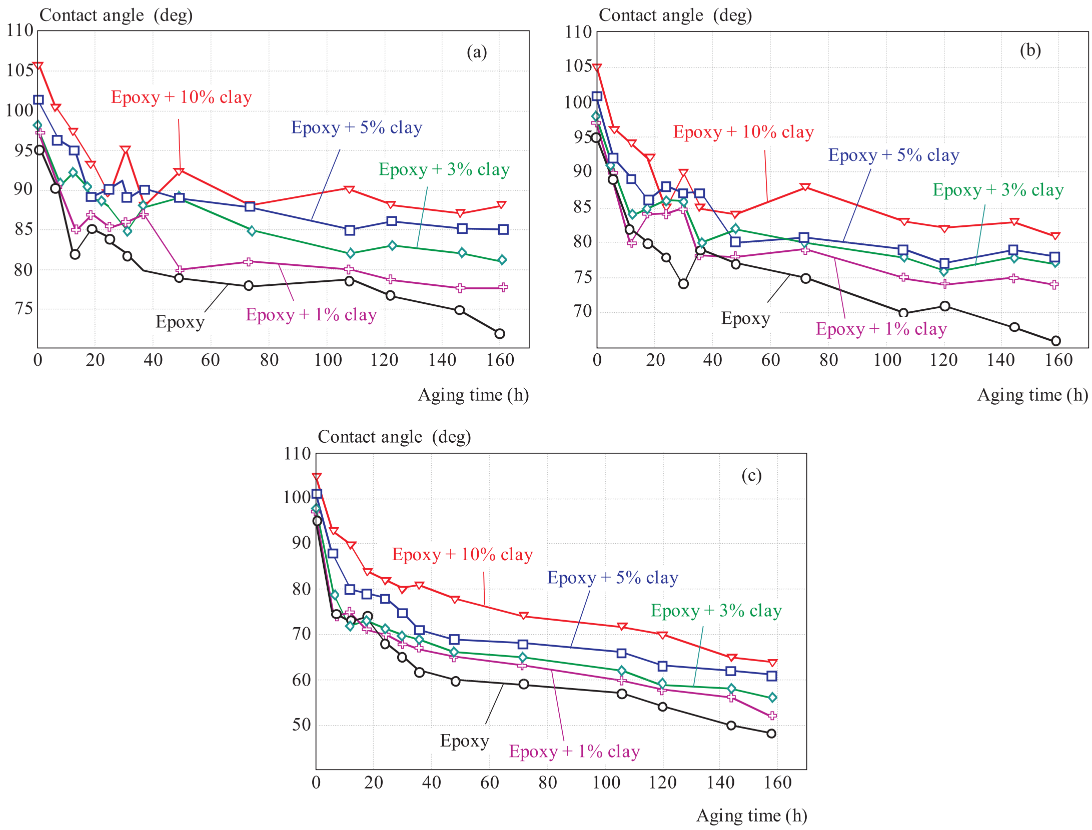

Fig. 2. Variation in contact angle of the specimen aged in water at different temperatures with time: (a) 30 °C, (b) 60 °C, (c) 90 °C

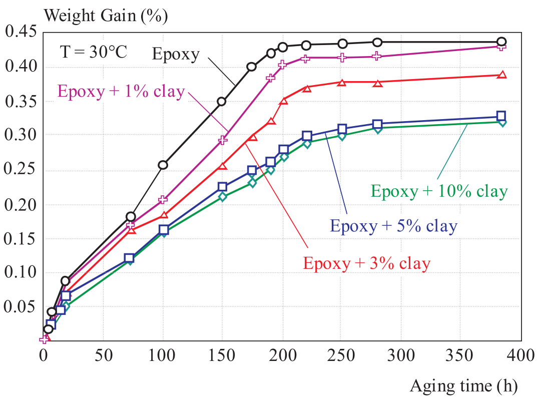

Fig. 3. Weight gain characteristics of epoxy nanocomposites due to water ageing

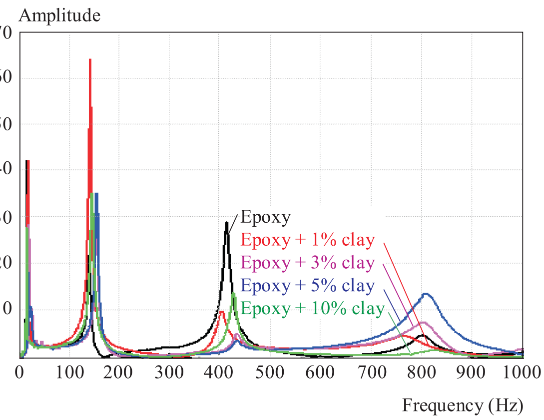

Fig. 4. Frequency response of epoxy nanocomposite material in vibration test

3.3 Vibration Test on Epoxy Nanocomposites

Figure 4 shows the frequency response function of epoxy nanocomposite material obtained through hammer impact test. The characteristic peaks obtained are the natural frequency of vibration [13]. Table 2 shows the first four modes of vibration frequency of epoxy nanocomposite materials. It is observed that increase in clay content (say up to 3 $wt\%$ of clay) in epoxy resin shows an increase in its natural frequency. In the present study, the natural frequency of vibration, not much characteristic variation in natural frequencies is observed between 3 and 5 $wt\%$ of clay in epoxy material. Above 5 % reduction in its value is observed but much higher than the pure epoxy resin material. The cause for it could be due to agglomeration of the nano clay material. Table 3 shows the damping factor of the epoxy nanocomposite material. It is realized that increase in percentage content of nanoclay shows increase in damping factor and above a certain percentage it shows a reduction in its value. The cause for the reduction in damping factor could be due to intercalation and agglomeration of material. The result matches with the XRD results where above 5 % intercalation characteristics is observed in epoxy nanocomposites. As a general remark, it may can be said that the present study's results confirm earlier findings, especially regarding the diffusion of water in epoxy nanocomposites and the contact angle as compared with those of neat epoxy [14, 15].

Table 1. Diffusion Coefficient of Epoxy Nanocomposites

Material	At 30 °C (m^2/sec) $\times 10^{-12}$	At 60 °C (m^2/sec) $\times 10^{-12}$	At 90 °C (m^2/sec) $\times 10^{-12}$
Epoxy	0.390	3.477	10.295
Epoxy +1%clay	0.274	2.841	7.443
Epoxy +3%clay	0.217	1.227	5.663
Epoxy +5%clay	0.124	0.963	4.721
Epoxy +10%clay	0.1219	0.912	4.091

Table 2. Natural frequency of epoxy nanocomposites

Material	Mode I	Mode II	Mode III	Mode IV
Epoxy	17.98	141.97	414.2	800.8
E+1%MMT	20.98	143.97	403.0	764.1
E+3%MMT	22.00	156.04	434.9	802.9
E+5%MMT	21.92	156.04	433.7	808.9
E+10%MMT	18.01	141.97	414.5	823.9

Table 3. Damping Characteristics of Epoxy nanocomposites

Material	Mode I	Mode II	Mode III	Mode IV
Epoxy	0.023	0.017	0.015	0.027
E+1%MMT	0.039	0.014	0.022	0.041
E+3%MMT	0.044	0.016	0.021	0.036
E+5%MMT	0.026	0.017	0.024	0.038
E+10%MMT	0.024	0.015	0.017	0.031

4 CONCLUSIONS

The important conclusions arrived at, based on the present study, are the following

- WAXD studies indicate that intercalated structures are formed above 5 $wt\%$ of clay in epoxy resin.
- Increase in percentage of nano clay in epoxy resin shows an increase in hydrophobic characteristics of the material. This is realized from the contact angle measurement. Diffusion of water in epoxy nanocomposite is less compared to pure epoxy resin material.
- The free vibration test results indicate that natural frequency of vibration and damping factor of the material increases by adding up to 5 $wt\%$ of nano clay to base resin.

The above indicate that the addition of nano clay into the base material has some beneficial effects.

References

[1] LEWIS, T. J.: Nanometric Dielectrics, IEEE Trans. Diel. Electr. Insul., 1 (1994), 812-825..

[2] KOZAKO, M.—FUSE, N.—OHKI, Y.—OKAMOTO, T.—TANAKA, T.: Surface Degradation of Polyamide Nanocomposites Caused by Partial Discharges Using IEC (b) Electrodes, IEEE Trans. Dielectr. Electr. Insul. **11** (2004), 833–839.

[3] TANAKA, T.—MONTANARI, G. C.—MULHAUPT, R.: Polymer Nanocomposites As Dielectrics and Electrical Insulation-Perspectives for Processing Technologies, Material Characterization and Future Applications, IEEE Trans. Dielectr. Electr. Insul. **11** (2004), 763–784.

[4] IMAI, T.—HIRANO, Y.—HITAI, H.—KOJIMA, S.—SHIMIZU, T.: Preparation and Properties of Epoxy-Organically Modified Layered Silicate Nanocomposites, Conf. Record of the 2002 IEEE Int. Symp. Electr. Insul., 379–382, 2002.

[5] LAN, T.—PINNAVAIA, T. J.: Clay-Reinforce Epoxy Nanocomposites, Chem. of Mater. **6** (1994), 2216–2219.

[6] PINNAVAIA, T. J.—BEALL, G. W.: Polymer Clay Nanocomposites, Wiley Series in Polymer Science, New York, 2000.

[7] MESSERSMITH, P. B.—GIANNELIS, E. P.: Synthesis and Characterization of Layered Silicate Epoxy Nanocomposites, Chem. of Mater. **6** (1994), 1719–1725.

[8] ALEXANDRE, M.—DUBOIS, P.: Polymer-layered Silicate Nanocomposites: Preparation, Properties and Uses of a New Class of Materials, Mater. Sci. and Eng. **28** (2000), 1–63.

[9] ZILG, C.—MULHAUPT, R.—FINTER, J.: Morphology and Toughness/Stiffness Balance of Nanocomposites Based Upon Anhydride-Cured Epoxy Resin and Layered Silicates, Macromol. Chem. Phys. **200** (1999), 661–667..

[10] Southern Clay Products, Inc, Technical Data.

[11] CRANK, J.: Mathematics of Diffusion, 2nd edition, Eds., Clarendon, Oxford, 1975.

[12] MURATA, Y.—MURAKAMI, Y.—NEMOTO, N.—SEKIGUCHI, Y.—INOUE, Y.—KANAOKA, M.—HOZUMI, N.—NAGAO, M.: Effect of Nano-Sized MgO-Filler on Electrical Phenomena under DC Voltage Application in LDPE, 2005 Ann. Rep. Conf. Electr. Insul. Diel. Phen., 158–161, 2005.

[13] SUAREZ, S. A.—GIBSONB, R. F.—DEOBALD, L. R.: Random and Impulse Techniques for Measurement of Damping in Composite Materials, Experim. Techn. (Oct 1984), 19–24.

[14] SARATHI, R.—SAHU, R. K.—TANAKA, T.: Understanding the Hydrophobic Characteristics of Epoxy Nanocomposites Using Wavelets and Fractal Technique, IEEE Trans. Diel. Electr. Insul. **15** (2008), 178–186.

[15] SARATHI, R.—DANIKAS, M. G.: Understanding Discharge Activity Due to Water Droplet in Epoxy Nanocomposites Using Acoustic Emission Technique, J. Electr. Eng. **59** (2008), 294–301.

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