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### **Dielectric spectroscopy of Mixed-Nanoparticle loaded Epoxy Resin**

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#### **ABSTRACT:**

In present work, the dielectric measurements of mixed nanoparticle loaded Bisphenol A-(epichlorhydrin); epoxy resin with hardener N(3-dimethylaminopropyl)-1,3-propylenediamine were carried out at room temperature. Mixture of highly pure grades of commercially available uncoated nanoparticle size compound of SiO<sub>2</sub> (APS-20 nm), and of ZnO (APS-30nm) were used as a filler and sample of the neat epoxy resin and nanoparticle loaded epoxy resin in the form of disc were prepared of different weight fraction (i.e 0.5 wt%, 0.7 wt%, 1 wt%, 1.5 wt%, 1.7 wt%, 2 wt%). Complex permittivity of the prepared samples was measured using Agilent E4980A precision LCR meter in frequency range of 10<sup>3</sup> Hz to 2 MHz. The dependency of dielectric behavior on concentration of mixed nanoparticle and comparative analyses of dielectric properties of all the samples in considered frequency range are discussed in detail.

**KEY WORDS:** Epoxy Resin, Nanocomposite, Dielectrics, Nanoparticle.

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## **INTRODUCTION:**

Epoxy resin is a one of the most commonly used thermosetting epoxide polymer that contains two or more epoxide groups. Because of its important impact on mechanical, chemical and electrical properties, epoxy resin become essential materials for industrial applications and widely used such as dielectric materials, anti-corrosion materials and manufacture of composite materials. In recent time, nanoreinforced technology - method of loading nanoparticle to improve properties of epoxy resin become prominent for epoxy resin modification. Normally 0.1% to 5% nanoparticle loaded polymer nanocomposites have been found to exhibit enhanced thermal property, mechanical property, physical property and dielectric property compared to the traditional polymer materials and that too at low nanofiller concentrations.[1-4] Since the concept of nano-dielectrics based on polymer nanocomposite technology came to be known, limited researcher reported dielectric characteristics of such systems for a better understanding of the behaviors depending on the type and concentration of nanofiller. Clear understanding of the permittivity behaviors in nanoparticle loaded epoxy systems is yet missing and hence this study tries to throw more knowledge into this perspective. [5-8]

In our previous research paper on epoxy nanocomposites we added SiO<sub>2</sub> and ZnO nanoparticle distinctly in an epoxy matrix with 0.5 to 2 wt. % concentration in an attempt to study dielectric behavior over a frequency range of 20 Hz – 2MHz at room temperatures and have seen that the relative permittivity for all the weight percentage is higher than unfilled epoxy resin, for epoxy ZnO composite and for epoxy SiO<sub>2</sub> composite for 0.5, 1 and 1.7 wt. % the dielectric permittivity is lower than the unfilled epoxy resin. This indicated that the presence of nanofillers can lead to higher or lower relative permittivity values compare to the neat epoxy depending on the type and concentration of nanoparticles. [9]

The purpose of the present paper is to study the effects of mixed nano-size fillers on dielectric behaviors of nano epoxy composite. SiO<sub>2</sub> and ZnO mixed nano fillers are used in this study. In this paper we reported complex permittivity of neat epoxy resin and SiO<sub>2</sub> – ZnO mixed nanoparticle loaded epoxy resin in the frequency range of 10<sup>3</sup> Hz to 2 MHz. Influence of filler concentration variation between 0.5 to 2 wt. % on the dielectric behavior of the nanocomposites is studied.

## EXPERIMENTAL PROCEDURE:

In the present study we used, bisphenol A-(epichlorhydrin); epoxy resin with hardener N(3-dimethylaminopropyl)-1,3-propylenediamine. (Supplied by Hindustan Ciba Geigy Ltd., Mumbai, India). We procured highly pure grades of commercially available uncoated nanoparticle size compound of  $\text{SiO}_2$  (APS-20 nm, Product of Otto chemical, India), and  $\text{ZnO}$  (APS-30nm, Product of SRL chemical, India). For processing a pure epoxy resin sample 100 parts by weight of the resin is mixed homogeneously with 80 parts by weight of hardener. And for processing a nanocomposite, the epoxy resin and hardener are taken in two different beakers and then required quantity of mixed  $\text{SiO}_2$  - $\text{ZnO}$  nanoparticle are mixed to the hardener vigorously with hand for few minutes and then resin and nanoparticle filled hardener was mixed by normal hand stirring method for 15 minutes. Finally, the uniformly mixed dough is then slowly decanted into the plastic molds, coated beforehand with wax. The composites were cast in this mold to get disc type specimens. Then they are kept under vacuum desiccation. It was found that a good dispersion could be obtained with this method. [10] The dielectric data of all samples in the frequency range of  $10^3$  Hz to 2 MHz using precision LCR meter were obtained. Dielectric measurements were achieved using an Agilent 4980A LCR meter with solid dielectric test fixture Agilent 16451B. [11, 12]

## RESULT & DISCUSSION:

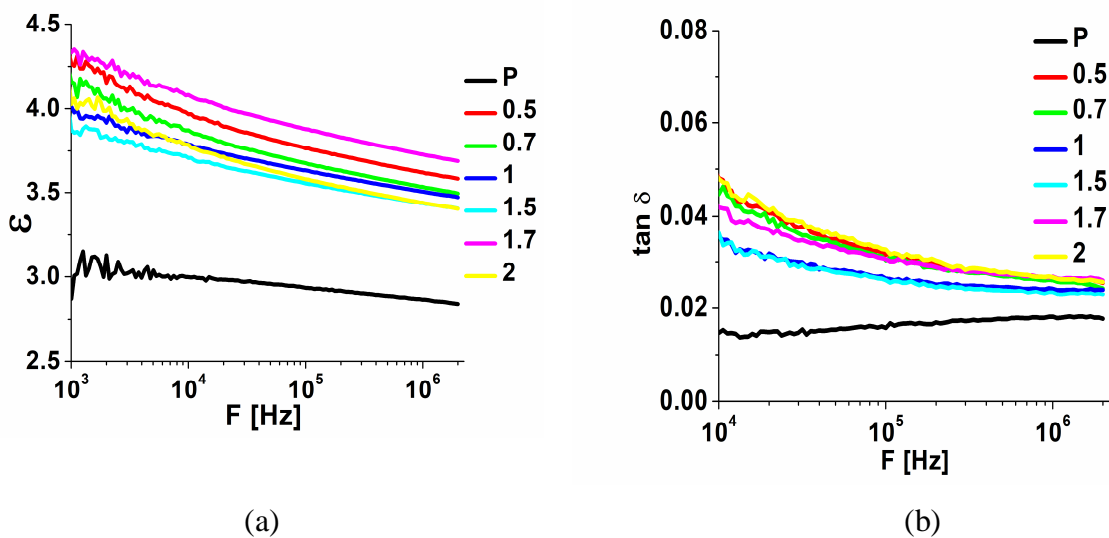


Figure 1- (a) Variations of relative permittivity and (b) Variations of  $\tan \delta$  loss in  $\text{SiO}_2$ - $\text{ZnO}$  mixed nanoparticle loaded epoxy composite in the frequency range of  $10^3$  Hz to 2 MHz.

For epoxy resin loaded with different weight concentration of mixed SiO<sub>2</sub>-ZnO nanoparticles, the variation of dielectric permittivity as a function of frequency is shown in Figure 1 (a). It is clearly seen in figure that effective permittivity decreases with increases in frequency. The same kind of behavior was observed in our earlier study in which SiO<sub>2</sub> and ZnO nanoparticle were mixed separately in epoxy resin. [9]. The values of dielectric constant for an epoxy resin loaded with mixed nanoparticle for all concentrations is higher than the pure epoxy resin. This result can be attributed to the increase in the polarity of all blends due to the increase of filler concentration, which in turn leads to an increase in the orientation polarization and to the presence of interfacial polarization. [11, 13] Compare to the SiO<sub>2</sub> nanoparticle loaded epoxy resin, SiO<sub>2</sub>-ZnO mixed nanoparticle epoxy resin system have high dielectric constant. This is may be due to low dielectric constant of pure SiO<sub>2</sub>(about 3.9 [14]) than that of ZnO (about 9 at 2 MHz frequency [15]). It is shown in figure 1(b) that for pure epoxy resin tan δ slightly increase with increase in frequency where as in mixed nanoparticle loaded epoxy resin tan δ value decrease with the increase in frequency with no evident of relaxation peak. This kind of behavior suggesting low frequency dispersion. [16,17]

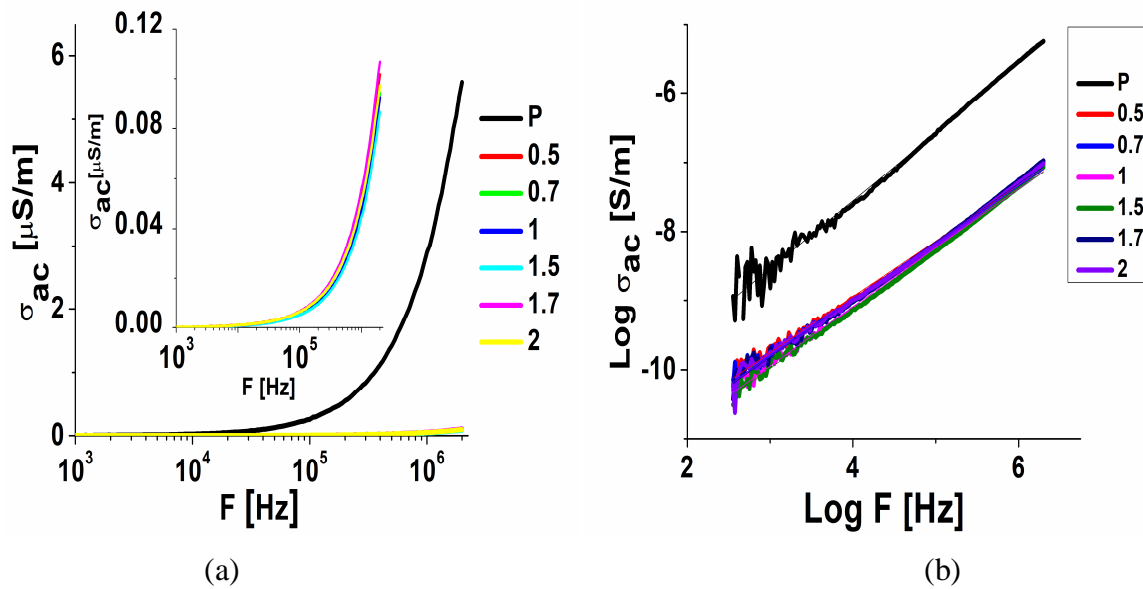


FIGURE 2- (a) Variations of ac conductivity in SiO<sub>2</sub>-ZnO mixed nanoparticle loaded epoxy composite in the frequency range of 10<sup>3</sup> Hz to 2 MHz. and (b) log [σ<sub>ac</sub>] versus log[F]

As shown in figure 2 (a) the value of ac conductivity is highest for unfilled epoxy resin in complete frequency range compare to nanoparticle loaded epoxy resin. Increase in ac conductivity in higher frequency region indicates that the charge carries are sufficiently free to follow the change in electric field. The ac conductivity patterns show a frequency independent plateau in the low frequency region and exhibits dispersion at higher frequencies. This behavior obeys the universal power law,

$$\sigma_{ac} = \sigma_0 + Af^n$$

Analyzing the ac conductivity by plotting  $\log [\sigma_{ac}]$  versus  $\log[F]$  (figure2 (b)) under the condition,  $\sigma_0$  much less than that  $\sigma_{ac}$ , equation can be simplified as

$$\sigma_{ac} = Af^n$$

The values of coefficient A and n were obtained by fitting the  $\sigma_{ac}$  is tabulated in Table 1 and it was found that the value of n is near 1.[18,19,20]

**TABLE No. 1. “A and n values for composites with different concentration of nanoparticle”**

Sr. No.	Nano particle Weight % in epoxy Composites	Fitting parameter	
		n	A
1	Neat Epoxy	0.9937	-11.535
2	0.5 wt%	0.8092	-12.18404
3	0.7 wt%	0.81786	-12.25134
4	1 wt%	0.85496	-12.51077
5	1.5 wt%	0.86222	-12.56701
6	1.7 wt%	0.83649	-12.32883
7	2 wt%	0.82563	-12.463

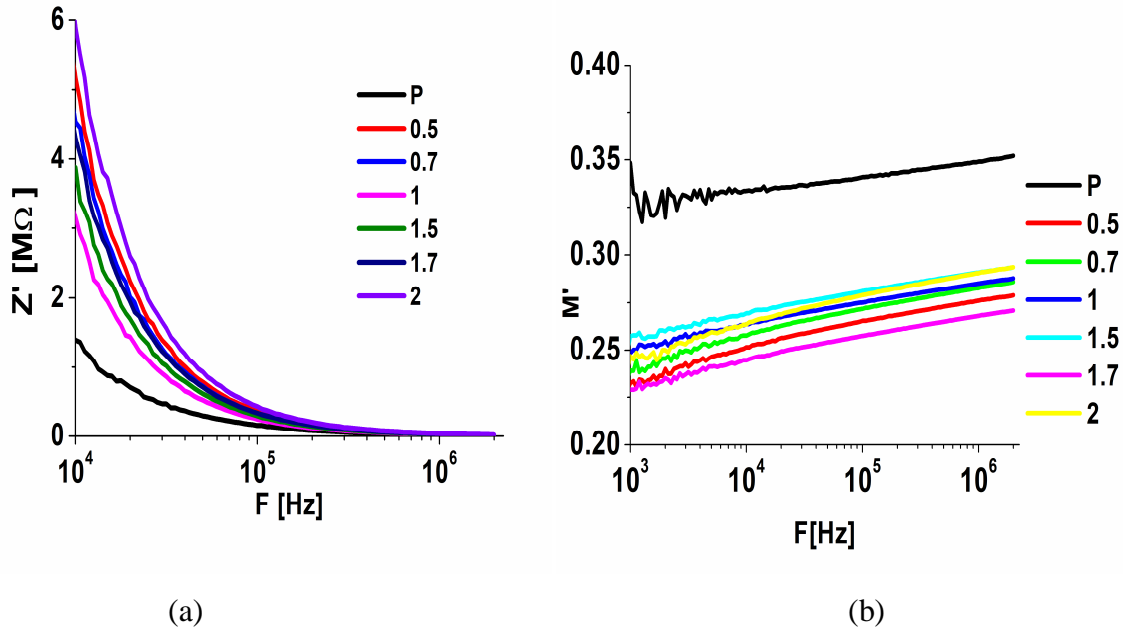


Figure3-(a) Variations of Real part of complex impedance (b) Variations of Electric modulus in SiO<sub>2</sub>-ZnO mixed nanoparticle loaded epoxy composite in the frequency range of 10<sup>3</sup> Hz to 2 MHz.

Figure 3(a) represents the dependence of the real part of complex impedance on the frequency. The figure shows the exponential decrease in the real part of complex impedance with the increase in frequency for all filler weight fractions. This indicates that the material become more conductive. This behavior indicate the cross link between the SiO<sub>2</sub> - ZnO nanoparticle and the epoxy system. [21-22]It can be clearly seen from figure 3(b) that values of electric modulus  $M'$  increased with increases in frequency. It is Because of at higher frequency dipolar groups are not able to orient themselves hence polarization become ineffective at higher frequency. [18]

## CONCLUSION:

The effective dielectric permittivity decreases with increase in frequency and the values of dielectric constants for all weight percentage of SiO<sub>2</sub>-ZnO mixed nanoparticle loaded epoxy resin is higher than neat epoxy resin, which is due the increase of filler concentration, which in turn leads to an increase in the orientation polarization and to the presence of interfacial polarization. Effective  $\tan \delta$  value decreases with the increase in frequency with no evident of relaxation peak suggesting “low frequency dispersion”. The ac conductivity was found to be higher in neat epoxy compare to SiO<sub>2</sub>-ZnO nanoparticle loaded epoxy resin and this ac conductivity behavior obey universal power law model. For all weight percentage of SiO<sub>2</sub>-ZnO mixed nanoparticle loaded epoxy resin, thereal part of complex

impedance were found to be frequency dependent. The values of electric modulus increase with increase in frequency, indicate ineffectiveness of polarization at higher frequency.

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