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Microwave Dielectric Relaxation Spectroscopy of Nano Filler Loaded Epoxy Composite

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Present work reports the result of spectroscopic dielectric relaxation study of (Bisphenol A-(epichlorhydrin): epoxy) and hardener (N(3-dimethylaminopropyl)-1,3-propylenediamine: hardener) doped with a range of concentrations of inorganic nano-fillers (SiO_2, Al_2O_3, TiO_2 and ZnO) and their mixtures. Measurements of complex permittivity of neat epoxy (epoxy + hardener), nano-epoxy composite (nano filler + neat epoxy) and mixed-nano epoxy composites (mixed nano filler + neat epoxy) are carried out using vector network analyzer along with SPEAG dielectric assessment kit over the frequency range of 200 MHz to 20 GHz at a constant temperature of 300.15 K. Obtained results are analyzed in order to attain the structural information and polarization mechanisms exhibited in these composites. Influence of varying concentrations of inorganic nano-fillers on the dielectric behavior of neat epoxy is explicitly conferred. From the obtained dielectric properties; other microwave energy parameters like power reflected (P_r), power transmitted (P_t) and penetration depth (d_p) are also determined at a spot frequency of 2.45 GHz and examined to gain additive information in view of their specific industrial and medical applications.

Keywords: Vector network analyzer, Dielectric relaxation spectroscopy, Epoxy resin, Nano-fillers, Penetration depth

1 Introduction

Epoxy resin developed an intense research focus as a base polymer because of its low cost, simple processing steps and versatility. Epoxy resin offers an excellent electrical, mechanical, chemical and thermal properties. Therefore, it is utilized as a thermosetting polymer polymer in multifunctional industries[1–5]; epoxy resin has been appointed for the present study. Enhancement in the aforementioned properties of epoxy resin can be accomplished by doping a range of concentrations of various inorganic nano-fillers. Utilization of inorganic nano-fillers such as SiO_2, ZnO, Al_2O_3 and TiO_2 is extremely suitable for doping in neat epoxy in order to accomplish reinforcement of epoxy resin. Dielectric, thermal, mechanical and physical properties of epoxy composites loaded with such nanofiller in the concentration range of 0.1 to 5 wt. % are extensively studied[6–9].

Study of dielectric properties in the microwave frequency region is more significant in various aspects such as in the manufacturing of insulating polymer matrix loaded wireless telecommunication systems, radar, *etc*. [10–13]. A group of researchers studied the dielectric properties of epoxy composites doped with various nano-fillers over the frequency range of 1 kHz to 10 MHz[8,14–18]. Apart from this; study of dielectric behavior of epoxy resin loaded nano-fillers in the microwave frequency region is also reported in the literature. Wang Yuan *et al.*[10] studied the complex dielectric permittivity of TiC powders loaded epoxy resin in the frequency range of 2.6–18 GHz and observed that TiC/epoxy composites are excellent microwave absorbers that exhibits wide bandwidth and thin breadth. Cheng Kuo-Chung *et al.*[19] reported the dielectric properties of epoxy resin–barium titanate composites over the frequency range of 1MHz to 1 GHz. However; an actual understanding for the influence of type, size and concentrations of various nano-fillers on the dielectric behavior of neat epoxy in the radio and microwave frequency regions is yet missing.

Recently, we reported the dielectric and electric properties of neat epoxy and nano-epoxy composites doped with a range of concentrations of SiO_2, ZnO, Al_2O_3 and TiO_2 nano-fillers over the frequency range of 1 kHz to 2 MHz at a constant temperature of 300.15 K and observed that the presence of nano fillers in neat epoxy can lead to higher or lower values of permittivity compared to that of neat epoxy depending upon type and concentration of nano fillers[20–22]. In the present work; we extend our study to investigate the effect of

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SiO_2, ZnO, Al_2O_3 and TiO_2 inorganic nano-fillers (of varying range of concentrations) and their mixtures on the dielectric properties of neat-epoxy over the frequency range of 200 MHz to 20 GHz at a constant temperature of 300.15 K.

2 Materials and Methods

2.1 Sample preparation

The epoxy resin (Bisphenol A-(epichlorhydrin): epoxy) and hardener (N(3-dimethylaminopropyl)-1,3-propylenediamine) were procured from Hindustan Ciba Geigy Ltd., Mumbai, India and were used to prepare neat epoxy without further purification. For the preparation of nano-epoxy composites; we procured high purity grades of commercially available uncoated inorganic nano-fillers: SiO_2 (APS-20 nm, from Otto Chemicals, Mumbai, India), ZnO (APS-30 nm, product of SRL Chemical, India), TiO_2 (APS-50 nm, product of SRL Chemical, Mumbai, India) and Al_2O_3 (APS-30 nm, SRL Chemical, Mumbai, India). A neat epoxy sample was prepared by mixing 100th part of the weight percentage (wt. %) of the epoxy homogeneously with 80th part of weight fraction of hardener. Similarly; for the preparation of nano-epoxy composites a range of concentrations (0.5, 0.7, 1, 1.5, 1.7 and 2 wt. %) of procured nano-fillers was dispersed through continuous hand stirring into neat epoxy. Finally, the uniformly mixed dough (neat epoxy filled with nano fillers) was slowly decanted into the plastic molds, coated earlier with wax. The composites were then casted in this mold in order to get disc type specimens. Similar technique was employed for the preparation of mixed nano-epoxy composites, where the mixtures (over a range of concentrations) of two different nano-fillers were doped in neat epoxy. Prepared neat epoxy, nano-epoxy composite and mixed nano-epoxy composites were kept under vacuum desiccation prior to utilize for the experiment.

2.2 Experimental techniques

Anritsu Shockline Vector Network Analyzer (VNA) model no. MS46322A along with SPEAG DAK-TL (Dielectric Assessment Kit) as represented in the block diagram (Fig. 1) were used to measure the dielectric constant and dielectric loss of the prepared epoxy composites over the frequency range of 200 MHz to 20 GHz (over 397 distinct data points with a frequency span of 50 MHz) at a constant temperature of 300.15 K. Prior to initiate the experiment; VNA was calibrated through Open–Short–Load technique [23] in order to eliminate the systematic measurement error. Dielectric measurements of the prepared composites were carried out over the considered frequency range following the technique described in the VNA and SPEAG-DAK manual[24,25]. Dielectric constant and dielectric loss were calculated by the DAK software itself from outcomes of VNA S-parameters at each frequency point of measurement.

3 Results and Discussion

Dielectric constant (ε′) and dielectric loss (ε″) spectra for neat epoxy and nano-epoxy composites doped with varying wt. % of Al_2O_3, SiO_2, TiO_2 and ZnO nano fillers over the frequency range of 0.2 to 20 GHz are represented in Fig. 2-5, respectively. It is

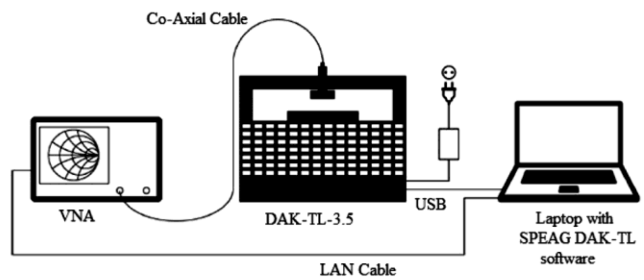

Fig. 1 — Block diagram of experimental setup of complex dielectric function measurements using VNA with SPEAG DAK-TL

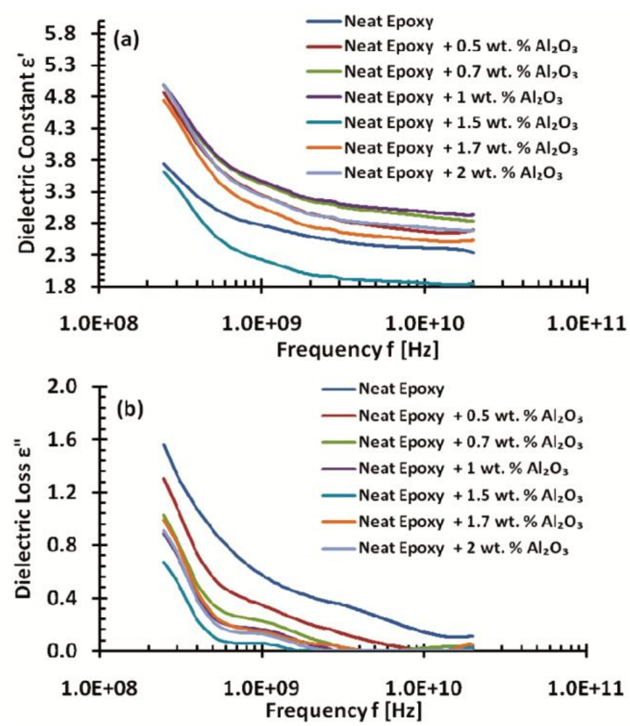

Fig. 2 — Frequency dependent spectra of (a) dielectric constant (ε′) and (b) dielectric loss (ε″) for neat epoxy and nano-epoxy composites consisting of Al_2O_3 nano fillers of various weight percentage concentrations over the frequency range of 0.2 to 20 GHz

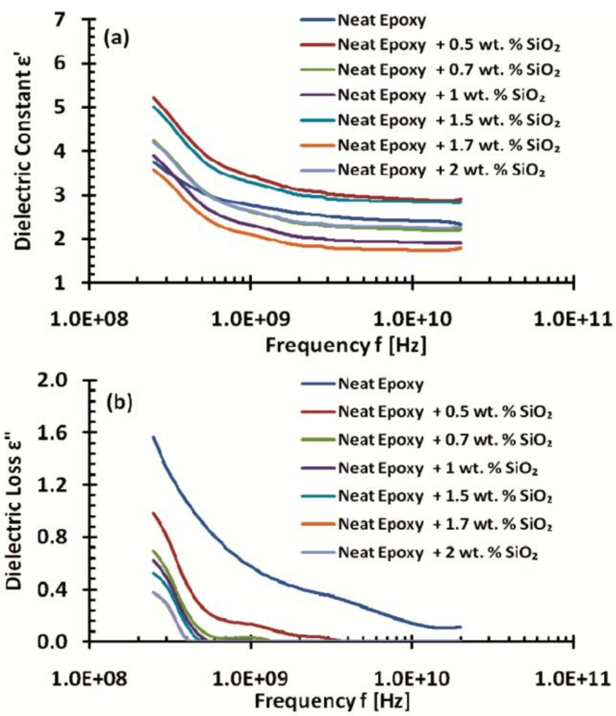

Fig. 3 — Frequency dependent spectra of (a) dielectric constant (ε′) and (b) dielectric loss (ε″) for neat epoxy and nano-epoxy composites consisting of SiO2 nano fillers of various weight percentage concentrations over the frequency range of 0.2 to 20 GHz

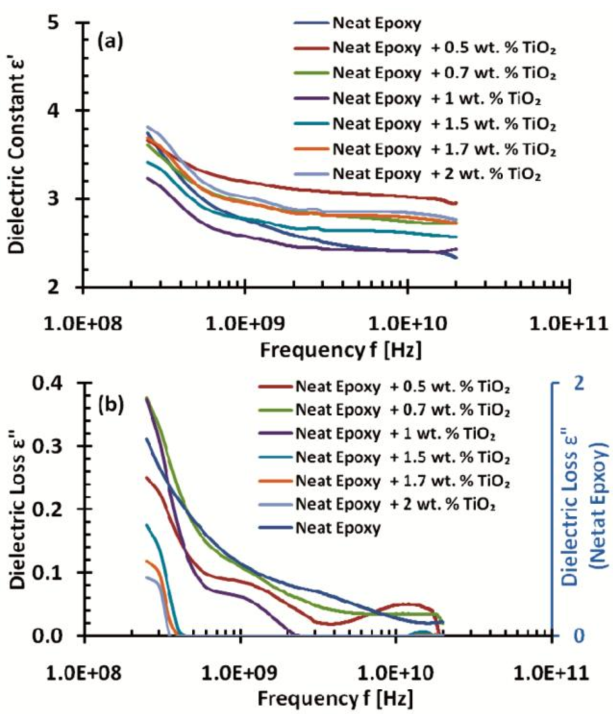

Fig. 4 — Frequency dependent spectra of (a) dielectric constant (ε′) and (b) dielectric loss (ε″) for neat epoxy and nano-epoxy composites consisting TiO2 nano fillers of various weight percentage concentrations over the frequency range of 0.2 to 20 GHz

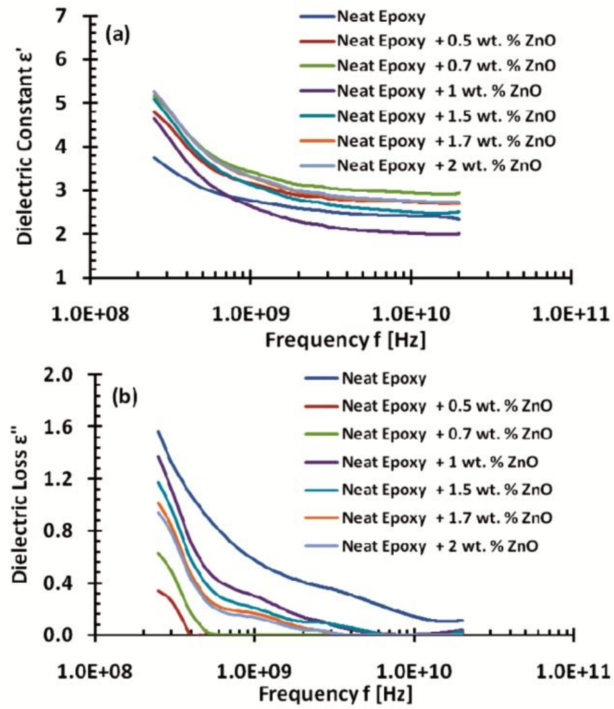

Fig. 5 — Frequency dependent spectra of (a) dielectric constant (ε′) and (b) dielectric loss (ε″) for neat epoxy and nano-epoxy composites consisting ZnO nano fillers of various weight percentage concentrations over the frequency range of 0.2 to 20 GHz

observed that the frequency dependent variation of dielectric constant and dielectric loss for various nano-epoxy composites exhibited identical behavior to that of the neat epoxy. For the lower frequency region i.e. $0.2 \leq f/\text{GHz} \leq 0.8$; values of ε′ and ε″ decrease rapidly with increase in frequency which can be attributed to the electrical conduction phenomena. [26] Beyond this frequency range i.e. for $0.8 \leq f/\text{GHz} \leq 20$; values of ε′ level off marginally and attained frequency independent behavior. Perceptible variation in dielectric constant and dielectric loss values with variation of nano-fillers can be understood from the actual chemical cross-linking process between neat epoxy and hardener, depicted schematically in Fig. 6[27]. The primary amine reacts with the epoxide group to form tertiary amine then as a result of catalytic effect of tertiary amine, poly ether is formed due to self-polymerization of the epoxide group. When nano-fillers are added during the curing process of amine and hardener; enormous variation in structural changes such as disappearance of epoxy dipoles, modification of amine dipoles, appearance of hydroxyl dipoles, mobility of dipoles and total number of dipoles per unit volume [28,29] takes place

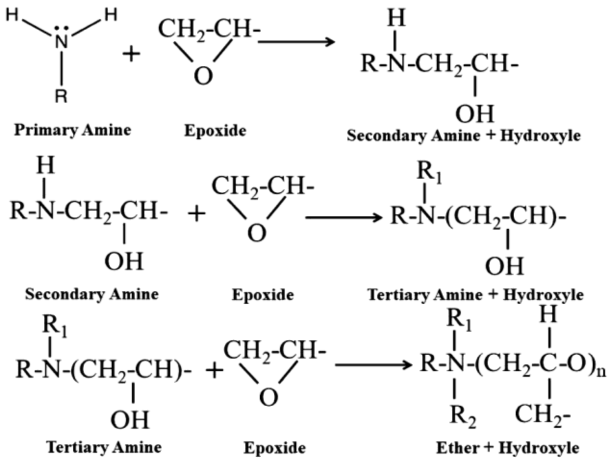

Fig. 6 — Possible cross-linking chemical interaction between epoxy and hardener

depending upon the type and amount of nano-filler added, the effective change in the value of complex dielectric were observed. Moreover; anomalous variation in the dielectric properties of neat epoxy due to the addition of varying wt. % of nano-fillers can be attributed to the existence of large number of active trailing bonds upon the surface of nano-fillers which are dispersed into the neat epoxy at different chains and interfaces[30]. Thus; formation of steady nano-layers takes place due to strong interaction between polar groups of epoxy and nano-fillers which may restrict the mobility of side/end chain of epoxy[31]. It is also observed that for some wt. % of nano-fillers, dielectric constant values are lower than that of neat epoxy over the considered frequency range, because the bigger dipole chains in the epoxy are restricted by the immobile nano layers around the nano-fillers.

From the Fig. 2 (b), it can be seen that in the dielectric loss (ε'') spectra of all Al_2O_3 wt.% loaded nano-epoxy composites, relaxation peak appears around 1 GHz. Whereas, in case of SiO_2, TiO_2 and ZnO the relaxation peak is not observed at all concentrations (Fig. 3 (b), Fig. 4 (b) and Fig. 5 (b)), but it appears only for 0.5 and 0.7 wt.% of SiO_2, 0.5, 0.7 and 1 wt. % of TiO_2,and 1, 1.5, 1.7 and 2 wt. % of ZnO loaded nano-epoxy composites around 1 GHz. Dielectric loss spectra for neat epoxy exhibited very weak relaxation peak at 2.75 GHz frequency. For such complex epoxy composite systems; several relaxation processes *i.e*. primary relaxation α and secondary relaxation β, γ may be observed, if the dielectric relaxation spectroscopic study is conducted over a broad range of frequency (a few Hz to several hundreds of GHz)[32,33].These relaxations have different frequency scales, depending upon the structure and bulkiness of molecular groups[34,35]. Dielectric loss peak of α-relaxation mechanism due to relaxation of whole molecules, generally appears in the lower frequency region (radio waves) and at high temperature [(32,33,36)]. In the present work the measurements are done over a limited frequency range, $0.2 \leq f/\mathrm{GHz} \leq 20$, at room temperature. Furthermore, there exist cross linking interactions between epoxy and hardener, hindering the relaxation of the whole molecule of the system. Therefore, the observed loss peak cannot be assigned to the α-relaxation mechanism. The dielectric loss peak of β-relaxation is attributed to the rotation of hydroxyl groups. But due to the lower concentration of hydroxyl groups in the studied composites, its contribution to the exhibited relaxation processes is negligible [29,34]. Therefore, the relaxations observed in these composites are mainly the γ-relaxations. The γ-relaxation is attributed to the motion of dipolar groups of atoms, including neat epoxy, amine groups with the unreacted epoxy + hardener and dipolar groups with the intermediate products as well as final polymers [33–35,37].

Dielectric constant (ε') and dielectric loss (ε'') spectra for neat epoxy and mixed nano-epoxy composites consisting of different weight percentage i.e.0.5, 0.7, 1, 1.5, 1.7 and 2 wt. % of Al_2O_3-ZnO, Al_2O_3-SiO_2, Al_2O_3-TiO_2 and SiO_2-ZnO nano-filler mixtures over the frequency range of 0.2 GHz to 20 GHz and at a constant temperature of 300.15 K are represented in Fig. 7-10, respectively. It is observed that the dielectric constant (ε') and dielectric loss (ε'') of various mixed nano-epoxy composites showed similar frequency dependent response to that of the single nano-filler loaded epoxy resin. However; mixed nano-filler loaded epoxy nanocomposites exhibited dual relaxation peaks except in 1.7 and 2 wt. % of Al_2O_3-ZnO, 0.5 wt. % of Al_2O_3-SiO_2 as well as 1.7 and 2 wt. % of SiO_2-ZnO nano-filler mixtures loaded epoxy composites. Dual relaxation peaks observed for mixed nano-epoxy composites can also be attributed to γ-relaxation mechanisms, which existed in individual nano-fillers doped epoxy composites. Nwigboji *et al.* reported similar behavior of dielectric loss (ε'') for carbon nanotube (CNT) - epoxy composite over the frequency range of 1 GHz to 26.5 GHz. [38]Spectral lines of dielectric constant (ε') and dielectric loss (ε'') of neat epoxy, nano-epoxy composite and mixed nano-epoxy composites exhibited anomalous behavior with respect to the nano-filler concentrations.

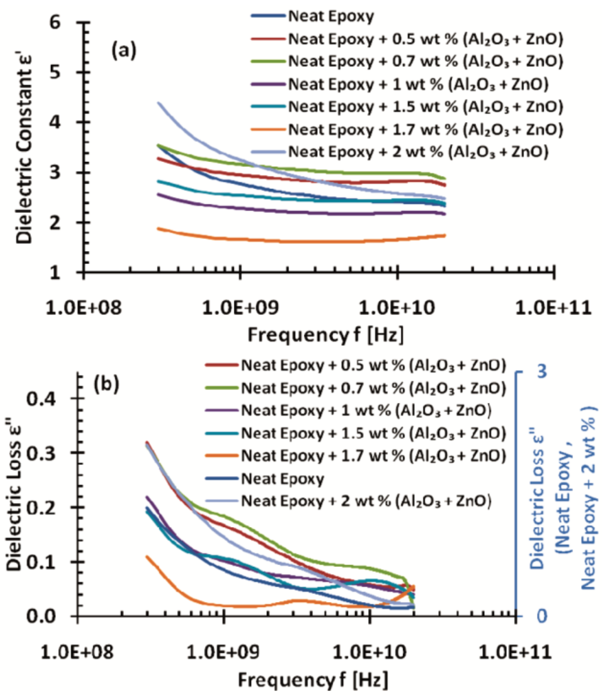

Fig. 7 — Frequency dependent spectra of (a) dielectric constant (ε′) and (b) dielectric loss (ε″) for neat epoxy and nano-epoxy composites consisting of mixed nano fillers Al_2O_3-ZnO of various weight percentage concentrations over the frequency range of 0.2 to 20GHz

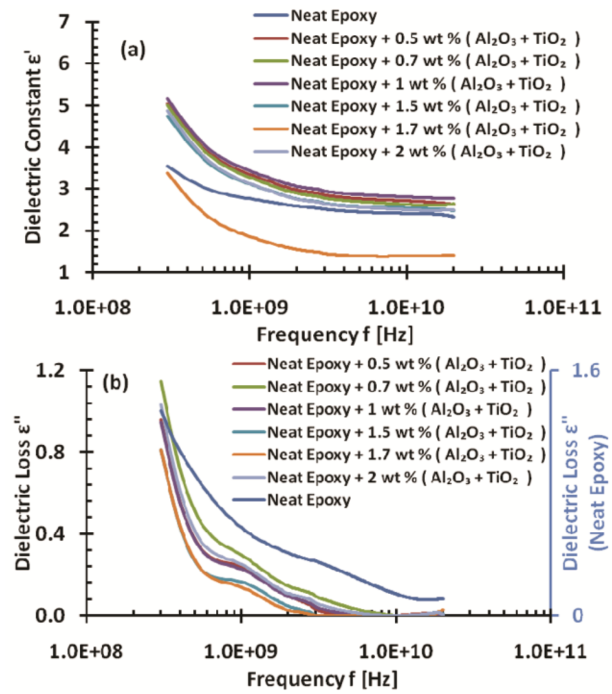

Fig. 9 — Frequency dependent spectra of (a) dielectric constant (ε′) and (b) dielectric loss (ε″) for neat epoxy and nano-epoxy composites consisting of mixed nano fillers Al_2O_3-TiO_2 of various weight percentage concentrations over the frequency range of 0.2 to 20 GHz

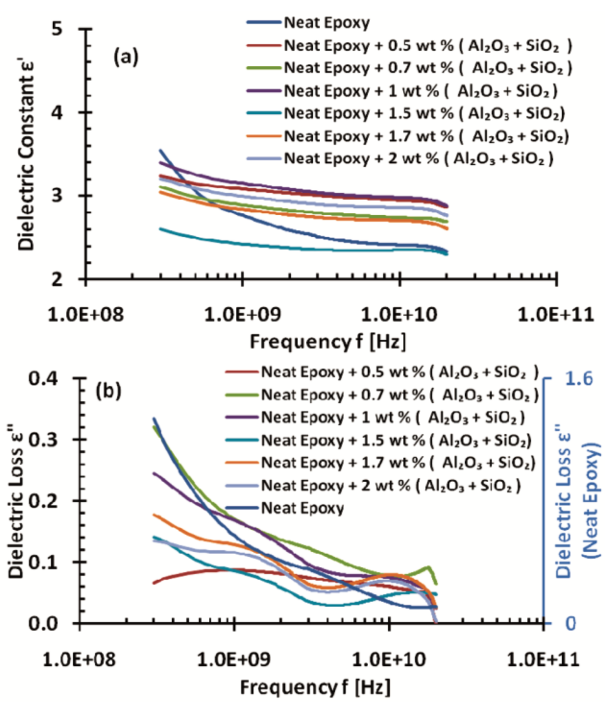

Fig. 8 — Frequency dependent spectra of (a) dielectric constant (ε′) and (b) dielectric loss (ε″) for neat epoxy and nano-epoxy composites consisting of mixed nano fillers Al_2O_3-SiO_2 of various weight percentage concentrations over the frequency range of 0.2 to 20 GHz

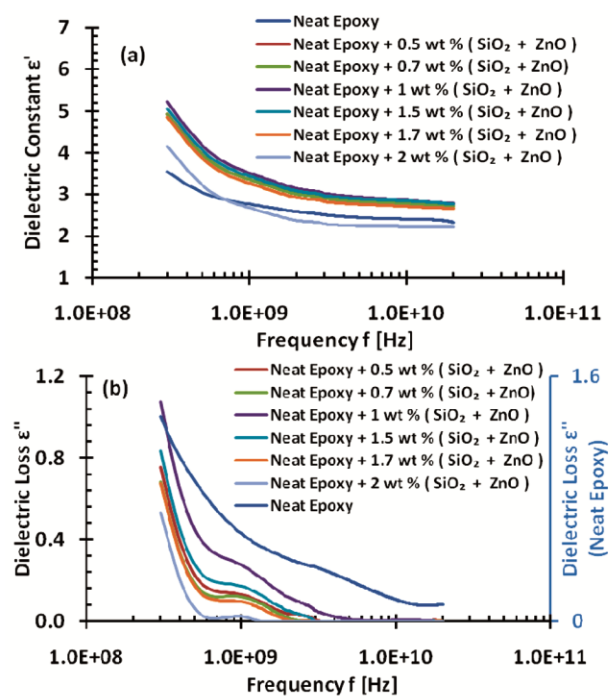

Fig. 10 — Frequency dependent spectra of (a) dielectric constant (ε′) and (b) dielectric loss (ε″) for neat epoxy and nano-epoxy composites consisting of mixed nano fillers SiO_2-ZnO of various weight percentage concentrations over the frequency range of 0.2 to 20 GHz

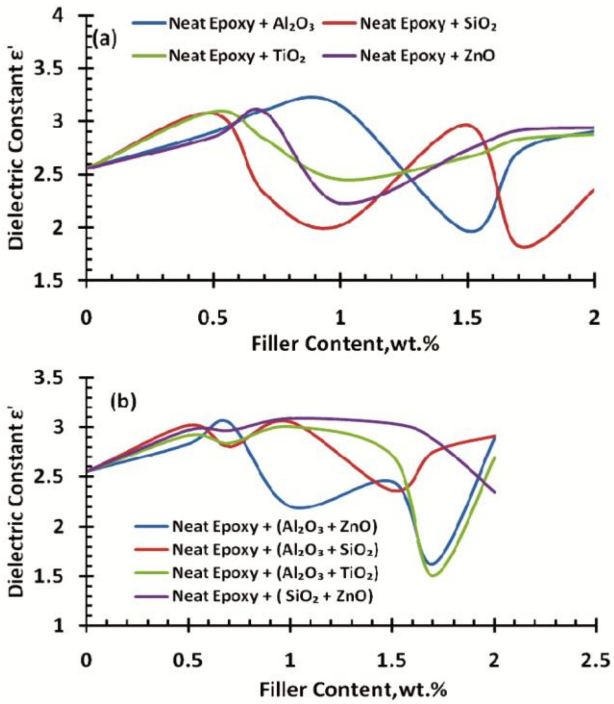

Fig. 11 — Variation of dielectric constant of nano-epoxy and mixed nano-epoxy composites with respect to the wt. % of doped nano-fillers at 2.45 GHz frequency.

The anomalous response of dielectric constant in nano-epoxy and mixed nano-epoxy composites with respect to the wt. % of doped nano-fillers at a spot frequency of 2.45 GHz frequency is represented in Fig. 11. Nano-epoxy composites loaded with Al_2O_3, TiO_2 and ZnO nano-fillers exhibited one fold decrease (dip) in ε′ at the concentrations of 1.5, 1 and 1 wt.%, respectively. SiO_2 loaded nano-epoxy composite exhibited two fold decrease (dip) at 1 and 1.7 wt.% concentrations. In the case of SiO_2 – ZnO mixed nano-filler loaded epoxy composites; one fold decrease is observed at 0.5 wt.% concentration whereas for Al_2O_3-ZnO, Al_2O_3 - SiO_2 and Al_2O_3 - TiO_2 mixed nano-fillers loaded epoxy composites; two fold decrease is observed at the concentrations of 1 and 1.7 wt.%, 0.7 and 1.5 wt.%, 0.7 and 1.7 wt.%, respectively. Reasons for the observed anomalous response of dielectric constant as a function of doped nano-fillers concentration are still unclear. Muradyan *et al.* reported such anomalous behavior of dielectric constant for carbon loaded nano-epoxy composite at 2.75 GHz frequency and suggested that difference in the surface energies of nano filler results in several anomalous features in the complex dielectric permittivity[39].

Table 1 — Microwave energy parameters: power reflected (Pr), power transmitted (Pt) and penetration depth (dp) for nano-epoxy composite at 2.45 GHz frequency.

Wt.% of nano-fillers	ε'	ε" (x 10^{-3})	P_r (%)	P_t (%)	d_p (m)
Al_2O_3 nano-filler loaded epoxy composite					
Pure	2.55	378.05	13.68	86.32	0.08
0.5	2.90	170.80	24.74	75.26	0.19
0.7	3.11	64.90	36.99	63.01	0.53
1.0	3.15	21.93	45.63	54.37	1.58
1.5	1.96	0.11	15.81	84.19	253.93
1.7	2.71	47.97	28.03	71.97	0.67
2.0	2.90	3.11	44.42	55.58	10.68
SiO_2 nano-filler loaded epoxy composite					
0.5	3.07	39.94	39.44	60.56	0.86
0.7	2.33	0.16	26.88	73.12	182.65
1.0	2.02	0.06	17.37	82.63	463.15
1.5	2.97	0.10	51.16	48.84	335.35
1.7	1.83	0.09	12.15	87.85	298.43
2.0	2.35	0.13	27.85	72.15	231.63
TiO_2 nano-filler loaded epoxy composite					
0.5	3.09	37.47	40.43	59.57	0.91
0.7	2.83	58.13	30.30	69.70	0.56
1.0	2.45	0.25	30.94	69.06	120.70
1.5	2.66	0.15	38.91	61.09	213.71
1.7	2.83	0.09	45.69	54.31	347.31
2.0	2.88	0.16	47.26	52.74	201.58
ZnO nano-filler loaded epoxy composite					
0.5	2.85	0.14	46.44	53.56	237.69
0.7	3.09	0.32	55.59	44.41	106.81
1.0	2.23	112.53	13.58	86.42	0.26
1.5	2.73	96.77	24.78	75.22	0.33
1.7	2.91	37.54	35.04	64.96	0.89
2.0	2.94	36.76	36.09	63.91	0.91

The Federal Communications Commission (FCC) allocated a range of microwave frequencies ($896 \leq f/\text{MHz} \leq 915$ and 2.45 GHz) for Industrial, Scientific and Medical (ISM) applications, among which 2.45 GHz is the major operating and widely utilized frequency globally[37,40–42]. In view of the significance of this frequency; microwave energy parameters such as power reflected (P_r), power transmitted (P_t) and penetration depth (d_p) are evaluated, for nano-epoxy and mixed nano-epoxy composites, using Equations (1) to (3)[43–45].They are reported in Table 1 and 2.

$$P_r(\%) = \left(\frac{\sqrt{\varepsilon'}-1}{\sqrt{\varepsilon'}+1}\right)^2 \quad \text{... (1)}$$

$$P_t(\%) = 100 - P_r \quad \text{... (2)}$$

$$d_p = \frac{\lambda_0}{2\pi}\frac{\sqrt{\epsilon'}}{\epsilon''} \quad \text{... (3)}$$

Where; λ_0 is free space wave-length.

Table 2 — Microwave energy parameters: power reflected (P_r), power transmitted (P_t) and penetration depth (d_p) for mixed nano-epoxy composite at 2.45 GHz frequency.

Wt.% of nano-fillers	ε'	ε" (x 10^{-3})	P_r (%)	P_t (%)	d_p (m)
Al_2O_3-ZnO mixed nano-fillers loaded epoxy composite					
Pure	2.55	378.05	13.68	86.32	0.28
0.5	2.84	116.04	26.05	73.95	0.27
0.7	3.05	127.40	30.15	69.85	0.38
1.0	2.20	75.41	14.31	85.69	0.47
1.5	2.45	64.58	20.34	79.66	1.01
1.7	1.62	24.66	5.53	94.47	0.05
2.0	2.90	655.63	15.03	84.97	0.28
Al_2O_3-SiO_2 mixed nano-fillers loaded epoxy composite					
0.5	3.02	78.79	33.15	66.85	0.43
0.7	2.81	129.70	24.68	75.32	0.25
1.0	3.06	112.15	31.39	68.61	0.30
1.5	2.36	47.40	19.48	80.52	0.63
1.7	2.75	78.77	26.52	73.48	0.41
2.0	2.91	71.41	31.08	68.92	0.47
SiO_2-ZnO mixed nano-fillers loaded epoxy composite					
0.5	2.97	29.60	38.12	61.88	1.14
0.7	2.97	4.21	46.05	53.95	7.98
1.0	3.09	91.01	33.84	66.16	0.38
1.5	3.03	33.12	39.38	60.62	1.02
1.7	2.89	1.62	45.12	54.88	20.44
2.0	2.34	0.14	27.49	72.51	207.54
Al_2O_3-TiO_2 mixed nano-fillers loaded epoxy composite					
0.5	2.92	68.91	31.48	68.52	0.48
0.7	2.84	123.26	25.74	74.26	0.27
1.0	3.01	73.51	33.37	66.63	0.46
1.5	2.71	27.63	30.80	69.20	1.16
1.7	1.50	13.59	4.04	95.96	1.76
2.0	2.70	90.39	24.48	75.52	0.35

Power reflection and transmission co-efficient, also known as the reflectance and transmission; denotes the percentage of the incident power that gets reflected and transmitted. Higher values of power transmission (> 50%) and lesser power reflection are observed for all composites. According to Eq. 3; it is obvious that the materials with low dielectric loss factors have a very large penetration depth suggesting that all the composites studied in the present work are suitable for microwave assisted processing for industrial and medical applications. This means that very little of the energy is absorbed in the prepared composites, and the material is transparent to microwave energy[46]. Most of the polymeric materials hold large penetration depth values such as penetration depth values for polyethylene, polystyrene, natural rubber and teflon are 2953 m, 3809 m, 288 m and 2822 m respectively at 2.45 GHz frequency[47]. Different values of penetration depth for neat epoxy are reported in literatures ranging from 30 cm to 4100 cm at a spot frequency of 2.45 GHz[48–50]. However, such variance can be attributed to the dissimilarity in the sample preparation and experimental techniques utilized. In present work, determined value of neat epoxy is 8 cm which is beyond the range of reported values due to the reasons mentioned above. For nano-epoxy composites; values of penetration depth are varying from 19.43 cm to 463.14 m when varying wt. % of nano-fillers in neat epoxy. Similarly; for mixed nano-epoxy composites values of penetration depth are ranging from 5.05 cm to 207.53 m when varying wt. % of mixed nano-fillers in neat epoxy. Thus, it can be concluded that; addition of single and mixed nano-fillers in neat epoxy exhibited stronger influence on the penetration depth of nano-epoxy and mixed nano- epoxy composites. Non-linear variation in the values of penetration depth with respect to concentrations of single and mixed nano-fillers can be attributed to differences in their dipole strength, mobility of the dipole, mass of the dipole, *etc*.[47,51,52]. Higher values of penetration depth for nano-epoxy composite as well as mixed nano-epoxy composites signified that the microwave energy can easily penetrate through the composites and also allows uniform temperature distribution results into rapid microwave processing (for specific industrial, medical applications) because of faster heating rates[51].

4 Conclusions

Complex permittivity spectra of neat epoxy as well as inorganic nano-filler and mixed nano filler loaded epoxy composites over the microwave frequency region of 0.2 to 20 GHz at a constant temperature of 300.15 K are reported. Effect of type and concentrations of doped nano-fillers on the complex dielectric function values of neat epoxy is clearly observed. Spectral lines of dielectric constant (ε′) and dielectric loss (ε″) of neat epoxy, nano-epoxy composite and mixed nano-epoxy composites exhibited anomalous behavior with respect to the nano-filler concentrations. Dielectric loss peak observed in the dielectric loss spectra of nano-fillers loaded epoxy composites is attributed to the γ-relaxation mechanism. Determined microwave energy parameters such as power reflected, power transmitted and penetration depth for all composites, at a spot frequency of 2.45 GHz, are also reported and discussed in view of their industrial and medical applications.

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