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The Influence of ZnO nanoparticles in the epoxy resin on the complex permittivity and dissipation factor

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Abstract

Epoxy resins are used as electro-insulating materials because they have great electro-insulation properties, adhesion, chemical resistance, and minimal shrinkage during curing. Epoxy nanocomposites have gained much interest in the area of nanotechnology, because of the ease of manufacture and the significant gain in properties. The influence of the various concentration of ZnO nanoparticles in epoxy resin Vukol 022 on the changes of the complex permittivity and dissipation factor has been measured at the temperature range from 20 °C to 120 °C. Frequency dependences of these parameters were measured within the frequency ranges from 1 mHz to 1 MHz by a capacitance method. The 0,5 wt.% nanoparticles caused a decrease of the real part of the complex relative permittivity. The higher concentration of nanoparticles for frequencies above 10 Hz had higher real relative permittivity as pure epoxy resin. At the study of the influence of temperature on dissipation factor, α-relaxation process and its shift to lower frequencies with ZnO fillers were observed.

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Keywords: epoxy resin, nanoparticles, permittivity

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1. Introduction

Epoxy resins (below ER) are widely used in various parts of the industry due to their high adhesion and ability to fill full space of a sample without the creation of air bubbles. Due to these properties, they have excellent electrical insulation properties, so they are widely used as electrical insulating material Becker and Simon (2006), Ramprasad et al. (2010). The long chain molecular structure is characteristic of ER. One of the possibilities of their further improvement is their combination with nanoparticles (NPs). The development of ER-based nanocomposites (NCs) has opened up new possibilities in the field of nanotechnologies in order to obtain new functions suitable for industry. They are promising materials, which are designed and produced in a manner with a focus on the improvement of the thermal or mechanical properties. The epoxy chains are connected to NPs so interfacial regions Nelson et al (2002), Tanaka (2005) are formed around them, which has a significant influence on the dielectric properties of the final NCs. Studies of the complex relative permittivity of NC as a function of the frequency of electric field and temperature are one of the basic features of dielectrics Singha and Thomas (2008), Mallakpour and Behranvand (2016). In this work, we present a study of the influence of a various concentration of ZnO nano-filler in on the dielectric properties of the epoxy resin.

2. Experimental results

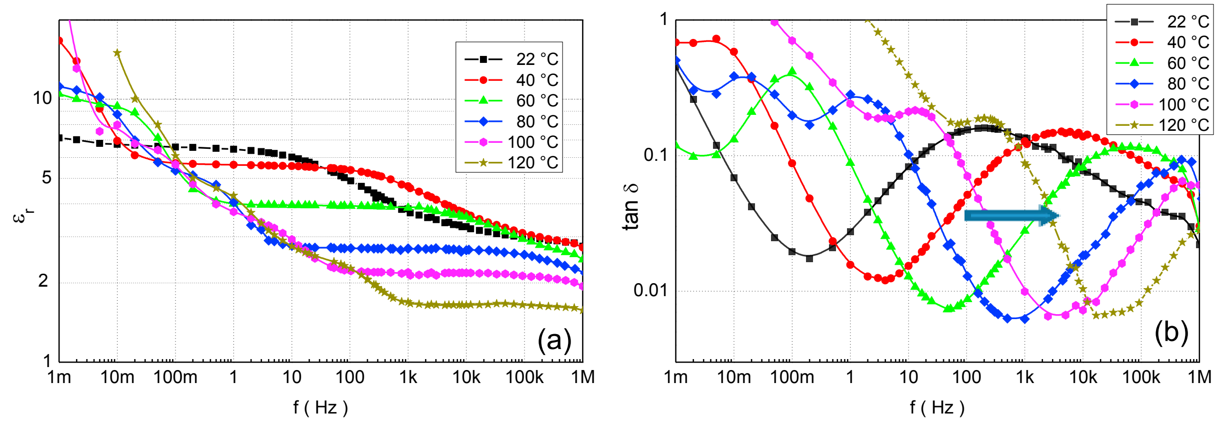

Fig. 1. The frequency dependence of the real part of the complex relative permittivity (a) and dissipation factor (b) for the epoxy resin with 1 wt% ZnO nanoparticles at various temperatures.

Fig. 1 shows the frequency dependence of the real part of the complex relative permittivity and dissipation factor of the ER with 0.5 wt% ZnO at various temperatures. Within the studied the frequency range, the change in the dielectric parameters is strongly depended on the frequency and the temperature. The real part of the complex relative permittivity (ε_r) for the temperature 20 ^{o}f decreased with increasing frequency (Fig. 1a). This decrease is not monotonic, but there are important parts. At frequencies below 1 Hz the permittivity is almost constant then slightly decreases with frequency and from 100 kHz it reaches a constant value. The decrease corresponds to the local maximum of tan δ around frequency 200 Hz. With increasing temperature and for frequencies higher than 100 mHz values of ε_r decrease. In the case of dissipation factor, its temperature and frequency development is more interesting. At the temperature 20 ^{o}C we can observe only one local maximum around 200 Hz. This maximum on the base of literature Jonscher (1999), Kochetor (2012) can be caused by the β-relaxation process. With increasing temperature, it moves to higher frequencies and for 80 ^{o}C its position has out of our study frequency range. At temperature to 60 ^{o}C also the next local maximum of *tan δ* appears at 0.1 Hz. This local maximum is caused by the α-relaxation process, which was also observed in various works, Soulintzis et al (2009), Kontos (2007), Klampár and Liedermann. (2012) and it also moves to a higher frequency as is shown by the arrow in Fig. 1b).

In Fig. 2a is shown the frequency dependence of the real part of the complex relative permittivity of ER and its mixtures with various concentrations of ZnO filler at 60 ° C. Within the given frequency range, we observe a significant change in ε_r of NCs with a concentration of NPs. Firstly, 0.5 wt% of ZnO filler caused a significant decrease in ε_r relative to pure ER for the whole frequency range. At 2 wt% we see an increase in the real permittivity, and from the frequency of 10 Hz, its value is higher than the pure ER. At frequencies below 10 mHz, ε_r of NCs reaches a similar value. In the case of dissipation factor (Fig. 2b), we notice a significant shift of the local peak due to α-relaxation to lower frequencies (50 and 100 mHz). The position of the second local peak is almost unchanged for 0.5 wt% of ZnO filler, but there is a big local minimum around 100 Hz. At higher concentration, the changes of *tan* δ were smaller and the position of the second peak is shifted to 10 kHz.

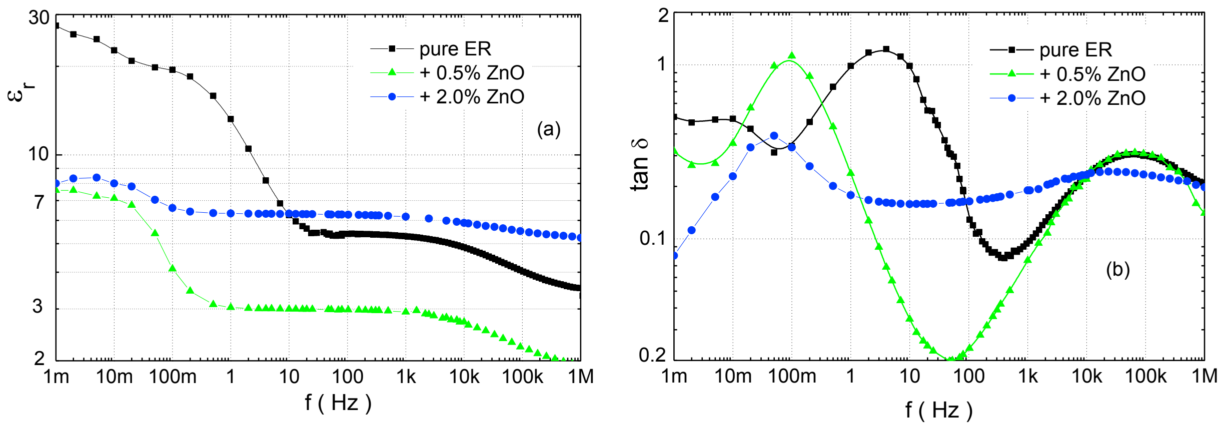

Fig. 2. The frequency dependence of the real part of the complex relative permittivity (a) and dissipation factor (b) at the temperature 60°C for the epoxy resin and its mixtures with 0.5 wt% and 2 wt% ZnO filler.

3. Discussion

The changes of the real part of the complex relative permittivity and dissipation factor for studied nanocomposites at different temperatures by the dielectric frequency spectroscopy were studied. Firstly, the frequency dependence of these parameters of ER with 0.5 wt% ZnO filler at various temperatures was presented. For *tan* δ it can be clearly observed two peaks (Fig. 1b), at low frequency caused by α-relaxation process and at high frequency caused by the β-relaxation process. There is a visible sift of these maximums to higher frequencies with the temperature. With the temperature increases thermal vibration and oscillation of nanoparticles and epoxy chains, so there are able to follow changes in the electric field to higher frequencies. At higher concentration the changes of ε_r was smaller, so we can suppose that NP caused less dependence on the frequency.

The results on Fig. 2 showed that the presence of nanoparticles in the epoxy resin had an important influence on their dielectric properties Jahoda (2018), Kudelcik (2018). These changes were influenced by the interfacial region around the NP and the epoxy matrix. The most significant impact for 0.5 wt% ZnO filler in the ER (Fig. 2a) was measured. There was the evident decrease in ε_r in the whole frequency range. The reason for this decrease was the presence of highly immobile epoxy chains in the interfacial regions (bonded and bond layers). With increasing concentration of the filler, ε_r increased due to the dominance of the NPs permittivity.

The dissipation factor for various concentrations of ZnO filler at the temperature 60 ºC is depicted in Fig. 2b). There are, again, two local maxima at frequencies between (50 mHz, 4 Hz) and around 60 kHz, respectively. At higher temperatures the developments of the dissipation factor were similar, but maxima's were shifted to higher frequencies. As was said the maximum at low frequency was caused by the α-relaxation process. In the interfacial region of NP and epoxy matrix are captured charges – electrons, which generate a local electric field around NP. This field is higher than Laplacian or geometric electric field so it has an influence on the reorientation of electric dipoles of epoxy chains bound in layers around NP. The higher electric field causes quicker transfer of dipole charges, what is reflected by shifting of this local maximum to lower frequencies. With an increase of filler concentration, the inter-particle distance between NPs decreased and dimer or oligomers from NPs can be created. These structures with the overlapping layers led to increased interfacial polarization and there is a better transfer of charges within the structure. These effects cause a slight increase in relaxation time with concentration. The position of the second local maximums, the β-relaxation process, was slightly influenced by the fillers.

4. Conclusion

The dielectric spectroscopy for the study of the epoxy nanocomposite systems with ZnO nanoparticles as fillers were used. The multi-layer model for the description of the observed dielectric properties of NCs was applied. The complex permittivity of nanocomposites measured within the frequency range from 1 mHz to 1 MHz was dependent on the concentration of nanoparticles. In the case of the concentration of 0.5 wt% ZnO filler, the permittivity was smaller for the whole frequency range. The decrease of the real permittivity was caused by the presence of highly immobile epoxy chains in the interfacial regions around nanoparticles. From the development of dissipation factor at various temperatures revealed the presence of the α- and β-relaxation processes. The shift of peak related to the α-relaxation process to lower frequencies for all type of nanoparticles was observed.

Acknowledgements

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References

Mleziva, J. 1993. Polymery – výroba, Struktura, Vlastnosti a použití. Sobotáles, Praha.

Becker, O. and Simon, G.P. 2006. Epoxy nanocomposites based on layered silicates and other nanostructured fillers. In Y-W. Mai and Z. Z. Yu (Eds.), Polymer Nanocomposites (pp. 29 -56). Woodhead Publishing Ltd, Cambridge, UK.

Ramprasad R., Shi N. and Tang C. 2010. Modeling the Physics and Chemistry of Interfaces in Nanodielectrics. in Nelson, J.K (Ed.), Dielectric Polymer Nanocomposites, Springer-Verlag, US.

Nelson J.K., Fothergill J.C., Dissado L.A. and Peasgood W. 2002. Towards an understanding of nanometric dielectrics. IEEE Conference on Electrical Insulation and Dielectric Phenomena, Cancun, Quintana Roo, Mexico, 295-298.

Tanaka, T., 2005. Dielectric nanocomposites with insulating properties. IEEE Trans. Dielectr. Electr. Insul., 12, 914.

Singha, S., Thomas, M.J., 2008. Dielectric properties of epoxy nanocomposites, IEEE Trans. Dielectr. Electr. Insul., 15(1), 12.

Mallakpour, S., Behranvand, V., 2016. Polymeric nanoparticles: Recent development in synthesis and application, eXPRESS Polym Lett., 10(11), 895.

Jonscher, A.K., 1999. Dielectric relaxation in solids. Journal of Physics D Applied Physics, 32(14), R57.

Kochetov, R. 2012. Thermal and Electrical Properties of Nanocomposites, Including Material Processing. Ph.D. thesis, Wöhrmann Print Service, Zutphen, the Netherlands (2012).

Soulintzis, A. , Kontos, G., Karahaliou, P., Psarras, G.C., Georga, S.N., Krontiras, C.A., 2009. Dielectric relaxation processes in epoxy resin—ZnO composites. Journal of Polymer Science B Polymer Physics, 47, 445.

Klampár, M., Liedermann, K., 2012. Dielectric relaxation spectroscopy of epoxy resins with TiO_2, Al_2O_3, WO_3, and SiO_2 nanofillers, *19th* IEEE International Symposium on Electrical Insulation, San Juan, Puerto Rico, 12907735.

Kontos, G.A., Soulintzis, A.L., Karahaliou, P.K., Psarras, G.C., Georga, S.N., Krontiras, C.A., Pisanias, M.N., 2007. Electrical relaxation dynamics in TiO2 – polymer matrix composites. eXPRESS Polymer Lett., 1(12), 781-789.

Jahoda, E., Kúdelčík, J., Hornak, J., Trnka, P., (2018). The influence of nanoparticles in the epoxy resin on dielectric parameters and partial discharges. ELEKTRO 2018 conference proceedings, Mikulov, Czech Republic, 1-5.

Kúdelčík J., Jahoda E., Hornak J., Michal O. and Trnka P., 2018. Partial Discharges and Dielectric Parameters of Epoxy Resin filled with Nanoparticles. Electric power engineering 2018: proceedings of the 19th international scientific conference, Brno, Czech Republic, 1-6.