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Effect of ZnO Nanoparticles on the Dielectric Properties of Polyurethane and Epoxy Resins

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Abstract. This study aims to present the influence of Zinc Oxide (ZnO) filler on the dielectric properties of two types of commercial potting compounds. For this purpose, the methods of dielectric spectroscopy were used to measure the changes in dielectric responses of polyurethane and epoxy by 1.0 wt. % ZnO over the frequency and temperature range, respectively. Two representatives of polyurethane and epoxy mixtures (VUKOL 022 and EPOXYLITE 3750 LV), that are commercially used in industry for filling cavities of all kinds (construction of electrical equipment), were used for this investigation. The ZnO nanoparticles in EPOXYLITE 3750 LV caused an increase, and in VUKOL 022 a decrease of the real permittivity compared to the pure matrix. Its decrease was most likely caused by the presence of highly immobile polyurethane chains in the interfacial regions around nanoparticles. The shift of a low relaxation maximum to higher frequencies for nanocomposite base on EPOXYLITE 3750 LV was affected by a smaller local electric field and electric double layers around the nanoparticles.

1. INTRODUCTION

On the transmission of energy of high-quality increasing demands are being made, whereas electronic equipment has become more sensitive to fluctuations of voltage [1]. The electrical equipment of the insulation systems (e.g., in transformers, bushings, cable) is formed by several components with different types of insulating materials (e.g., oil, impregnated paper/board, polymeric films, epoxy resin, silicone, or air). From the available statistics, the lifetime of oil transformers due to their failure (even though dielectric insulation issues originated 75% of all collapse) is not even 18 years [2]. One of the possibilities for a longer life of the transformer is its encapsulation by epoxy (EPs) or polyurethane (PUs). Polyurethanes, as one of the classes of polymers, is nowadays the most widely adopted primary insulating material in the industry of power engineering. Its properties, like mechanical strength, thermal stability, barrier resistance, and adhesive, are improved due to many researchers worldwide.

In comparison with other types of insulating materials, the polymers have as free of maintenance, higher reliability, and low dissipation factor [3]. Their excellent mechanical, electrical, and thermal properties are objects of interest for academic and industrial researchers. They have many applications in various industries due to their high adhesive strength and mechanical properties. They have excellent electrical insulating properties, which predisposes them to be used as electrical insulating materials [5, 6]. Nowadays, polymer-based nanodielectric systems are more popular

investigated for their electrical properties because the introduction of fillers of nano-sized provides several advantages in their properties compared with the similar properties obtained for systems without any particles or doped by the micrometer-sized fillers. Many studies clearly shown that adding some fraction volume of nanoparticles in EPs or PUs improves several characteristics as dimensional stability, dielectric properties, energy dissipation capabilities, and finally, the high-performance stiffness of polymer composite. Modification of the polymers, or in our case EPs or PUs by nanoparticles, is one of the possibilities to develop to acquire better flexibility, mechanical or electrical properties. The studies of the complex permittivity of nanocomposites (NCs) as a function of temperature, the intensity of the electric field, or frequency are part of the fundamental characterizations of dielectrics [7-10].

2. PREPARATION OF SAMPLES

The goal of this study is to present the influence of the incorporation of ZnO nanoparticles into two types of commercially used potting compounds. Two-component PU (the VUKOL 022 with hardener agent Vukit M from VUKI a.s. [11]), and single-component EP (EPOXYLITE 3750 LV produced by Elantas), respectively, were used for this experiment. The final nanocomposites consist of base matrices of PU, which are doped with 1 wt.% ZnO nanoparticles. A very detailed description of preparing NCs based on the VUKOL 022 was described in [12]. In the case of NCs based on the EPOXYLITE 3750 LV, the process of preparing samples is almost the same in the case of VUKOL 022. Since the EPOXYLITE 3750 LV is not a two-component, the sample preparation does not need any additional hardener. Thus, the curing process occur in a laboratory hot air dryer at elevated temperature (140 °C, 6 h).

3. EXPERIMENTAL RESULTS

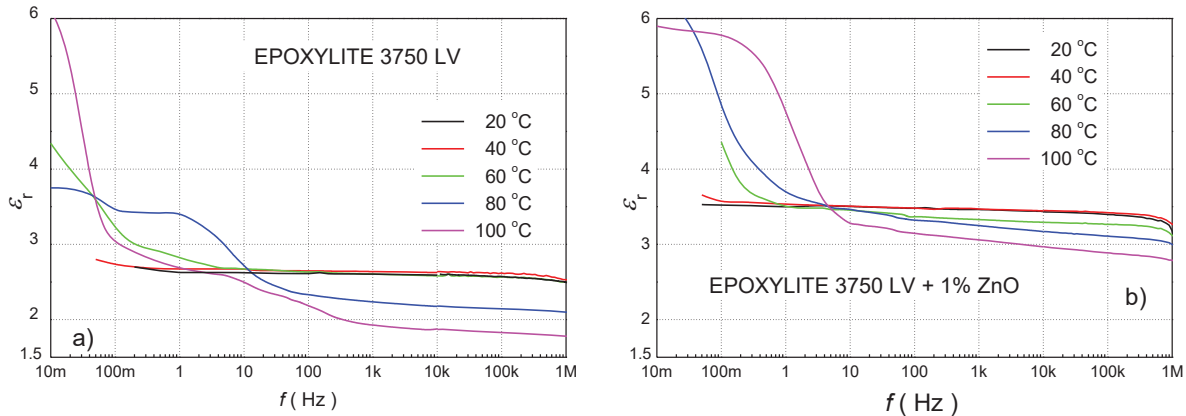


FIGURE 1. The frequency dependence of the real parts of the complex relative permittivity (a) of pure EP and (b) EP with 1 wt. % of ZnO nanoparticles at various temperatures.

The frequency dependences of the real relative permittivity (ϵ_r) of pure EP at various temperatures are shown in Fig. 1a). The ϵ_r was almost frequency independent at temperatures 20 and 40 °C, and it increased for low frequencies at higher temperatures. This increase is the effect of the α -relaxation process, which was also observed in various works, [12-14]. α -relaxation is the result of the micro-scaled Brownian motion of the whole chain of polymers. The decrease of ϵ_r for frequencies higher than 10 Hz at temperatures 80 and 100 °C. The influence of the addition of 1 wt.% of ZnO fillers to EP on ϵ_r is depicted in Fig. 1b). From comparison with pure EP, it can be seen that nanoparticles increased ϵ_r of the studied nanocomposite. This effect was caused by the higher permittivity of ZnO fillers than EP. At temperature 60 °C and higher, again increase of permittivity for low frequencies was observed. This increase due to the influence of the interfacial polarization around nanoparticles was more pronounced. At this type of polarization, the space charges accumulated at the interfaces between matrix and fillers create large dipoles able to follow the changes of the external electric field [13-16].

In Fig. 2a is shown a comparison of the frequency dependencies of the real parts of the complex relative permittivity of PU and EP and their mixtures with 1 wt.% of ZnO fillers at 80 °C. Within the given frequency range, we observe a various significant change in ϵ_r of nanocomposites by ZnO. Firstly, ZnO filler caused a significant

decrease in ϵ_r relative to pure VUKOL 022 for the frequencies higher than 2 Hz [12]. In the case of EPOXYLITE 3750 LV, the ZnO filler caused an increase in the real permittivity in the whole frequency range. In the case of the dissipation factor (Fig. 2b), the frequency developments are more interesting. At low frequencies, the first local maximum caused by the α -relaxation process was observed. The addition of ZnO filler to studied polymers caused a significant shift of this local peak. At high frequencies for PU and its mixtures with ZnO, the second local maximum by the intermediate dipolar effect: IDE-relaxation process was observed [15, 16]. For EP and its mixture with ZnO, no second local maximum was observed.

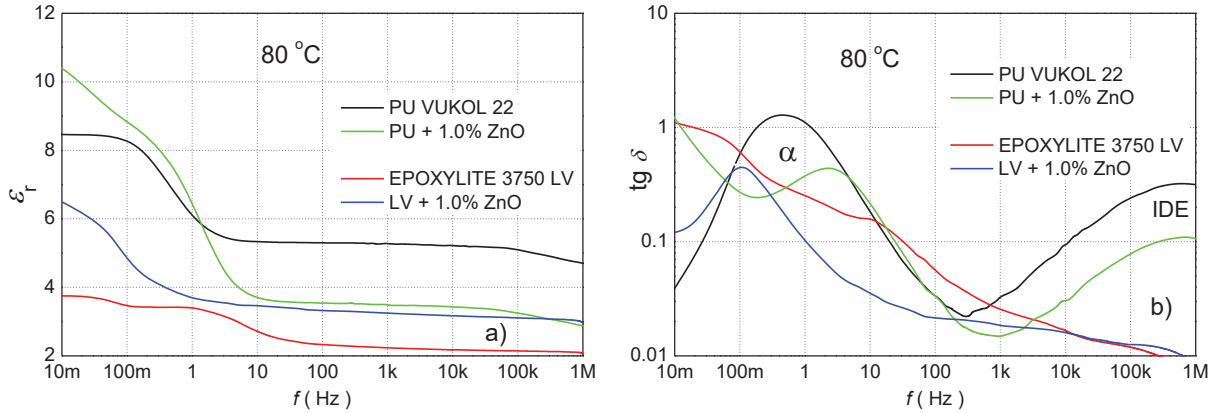


FIGURE 2. The frequency dependence of the real parts of the complex relative permittivity (a) and dissipation factor (b) at the temperature 80°C for the PU and EP and their mixtures with 1 wt.% ZnO filler.

4. DISCUSSION

The influences of ZnO filler on the dielectric parameters for two types of polymer resins are shown in Fig. 2. These changes were caused mainly by three factors: the interfacial region around the NPs, their relative permittivity, and the α -relaxation process. The decrease of ϵ_r in the PU + ZnO for frequencies higher than 2 Hz (Fig. 2a) was caused by the effect of the interfacial region (bonded and bond layers) around the NP. In this region, the mobility of polymer chains decreased, which reduced the effect of polarization and ϵ_r decreased [12]. In the case of EP was the situation opposite. The polymer chains were not so bounded in the interfacial region of NP, so ϵ_r increased due to the dominance of the NPs permittivity.

The influence of ZnO filler on the frequency dependence of dissipation factor is shown in Fig. 2b). In all cases, the low-frequency local maximum caused by the α -relaxation process was observed. This process is mainly influenced by a local electric field. NPs in polymer matrix capture electrons so their interfacial region can be charged, and a high local electric field around NP is generated. This field changed the orientation of electric dipoles of polymer chains bound in layers around NP and caused a quicker transfer of dipole charges. This effect influenced the increase of interfacial polarization, and there is a better transfer of charges within the matrix by α -process. These effects caused a rapid decrease in relaxation time and a shift of the local maximum to higher frequencies for PU with ZnO. For EP+ZnO, the polymer chains were weakly bounded, so the effect of a local electric field was minor. The relaxation time increased, and the shift of the local maximum was to lower frequencies. The main difference between studied polyurethanes was at high frequency. ZnO filler, usually in polymers, caused highlights IDE relaxation, but for EP, this effect was not observed [17].

5. CONCLUSION

The nanocomposites insulation materials prepared from EPOXYLITE 3750 LV (solvent-free resin, commonly used in industry to impregnate rotating machines) and VUKOL 022 (used in industry for filling cavities of all kinds) by addition of 1 wt.% ZnO fillers were studied by the dielectric spectroscopy. The real permittivity of the nanocomposite base on EPOXYLITE 3750 LV was higher than pure EP due to the high permittivity of ZnO fillers. In the case of VUKOL 022 with 1 wt.% ZnO fillers, it decreased for frequency higher than 2 Hz, as the effect of the immobile

polyurethane chains in the interfacial region around the NP. Nanoparticles in the base matrix captured electrons, so a high local electric field around them was generated, which influenced the α -relaxation process.

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