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# Effect of MgO Nanoparticles on Material Properties of Cold-Curing Epoxy and Polyurethane Mixtures

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**Abstract.** Potting compounds are an integral part of a large number of electrical devices, in which play the role of mainly mechanical or photosensitive protection. So-called cold-curing resins are one of the subgroups of potting mixtures. This article focuses on this type of potting compound based on epoxy (EPs) and polyurethane (PUs). In addition to the study of their electrical properties, structural properties are also introduced. The main focus of research is on the incorporation of nanometric magnesium oxide (MgO) in different weight ratios and the study of its effect on the properties of synthesized composites. The electrical properties analyses include the changes in relative permittivity, dielectric losses in the wide frequency range, and volume resistivity estimated at DC conditions. In terms of structural analyses, the effect of MgO incorporation on glass transition temperature changes was studied. From the performed investigations it is evident that the selected basic materials can be partially modified to improve their properties, especially in lower levels of filling, where there is a positive decrease of relative permittivity close to industrial frequency ( $\approx 3$  % for EP+1MgO;  $\approx 11$  % for PU+1MgO). For investigated composites is also evident a trend in the growth of dielectric losses. MgO affects also the volume resistivity and glass transition temperature of EPs-MgO and PUs-MgO composites.

## INTRODUCTION

Epoxy resins (EPs) and polyurethane mixtures (PUs) represent a significant group of synthetic thermosets, that are used in many industrial applications. Epoxy resins excel especially in superior adhesion to various materials (metal, porcelain, wood, glass, or selected plastics). Other interesting properties include low water vapor diffusion or resistance to moisture and mold. From the point of view of most important properties in electrical engineering, it is necessary to mention high thermal resistance and stability, high electrical and mechanical strength or resistance of their surface to creeping currents [1] - [3]. Due to these facts, EPs are suitable for a wide range of electrical and other applications. As an example: layered insulators, molding compounds, adhesives, or sealants. EPs are also an inseparable part of the impregnation processes of motors, generators, transformers, coils, or insulators. The potting properties are used especially in the encapsulation of integrated circuits, hybrid circuits, various modules, sensors, optical elements, including LEDs, or also for the encapsulation of entire printed circuit boards. It is the encapsulation of sensitive components that can be crucial for the reliability of the device as a whole [4]. And because some electronic components are temperature sensitive, conventional potting materials cured at elevated temperatures cannot be used. In this case, the encapsulation must be performed with cold-curing epoxy resins [5]. These resins are cured at ambient

temperature and the possibility of curing without additional equipment (curing oven) is also advantageous. The limiting parameter for the usage of cold-cured epoxy resins is usually the low glass transition temperature [6].

Polyurethanes are an increasingly used polymeric material, mainly due to their versatility. Generally, PUs are based on the reaction of a polyol with a diisocyanate or polymeric isocyanate basis while using suitable catalysts and additives [7]. Good material properties make them interesting materials not only for electrical applications. They can be used in almost the same portfolio of applications as epoxy resins. However, an interesting feature of polyurethanes can be their high flexibility compared to EPs. On the contrary, the disadvantage may be a relatively low tensile strength with minimal elongation. Even polyurethanes can be cured at room temperature. The polyurethane thus hardened then has sufficient properties, for example, for encapsulating components on printed circuit boards or as potting compounds for low-voltage components. Polyurethanes cured in this way have also a relatively low water absorption or low cure shrinkage [8].

## MATERIALS

As has been already mentioned, the cold-curing mixtures were used for this experiment. Epoxy resin is based on Bisphenol A + Epichlorohydrin (one of the most common types of EPs). It has to be mixed together with an amine hardener in a 1:0.45 ratio, as is specified by the manufacturer. This mixture is fully cured at room temperature in the interval from 48 to 72 hours. After curing, the mixture is colorless and transparent (without additives).

The composition of the selected polyurethane matrix is not available, because this mixture is not classified as dangerous according to (EC) No 1272/2008, and individual substances are not shown in the material safety data sheet. The curing reaction starts after the addition of the hardener, and there is a relatively short processing time (28 min). The mixing ratio is recommended by the producer as 1:0.45. This mixture is cured after 12 – 16 hours at room temperature (or 1 hour at 60 °C). The application of mechanical or electrical stress must be performed after 3-4 days. This mixture is colorless and transparent.

The magnesium oxide nanoparticles with the purity of 99 % (rest are impurities Ca, K, Na) were used for this investigation. Selected particles have an ellipsoidal and spherical shape and an average size of 20 nm. Specific surface area is lower than 60 m<sup>2</sup>/g and bulk density is between 0.1 and 0.3 g/cm<sup>3</sup>. The color of MgO powder is white.

## SAMPLE PREPARATION

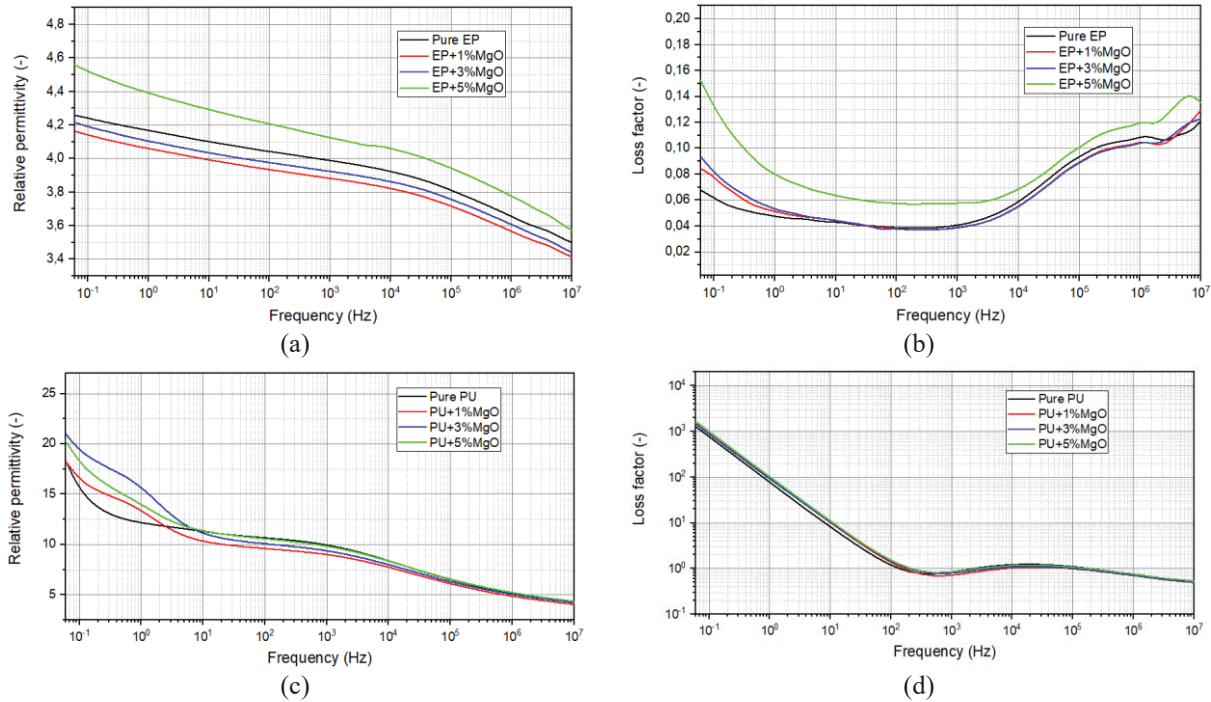
The composite samples were prepared in laboratory conditions by the direct dispersion method. Magnesium oxide particles have been dried in a laboratory hot air oven for 24 h to lose their surface moisture before their incorporation into the polymer matrix. To get the optimal concentration, the precise weight of the MgO nanofiller has been added (0, 1, 3, and 5 wt.%). The following procedure has been carried out for both compositions (based on EP and based on PU). The base material and MgO nanoparticles were mixed together for 5 h by a magnetic stirrer (700 rpm) and then combined with a vacuuming process (8 mbar, 60 rpm). During this time a great number of air bubbles were removed from the mixture. This method well guarantees the dispersion of nanoparticles in the polymer matrix, even at very low concentrations. Then has the hardener has been added to the mixture in the recommended ratio (1:0.45). The finished mixture has been vacuumed and then has been poured into flat square-shaped molds with dimensions 100 × 100 × 1.0 ± 0.2 mm. Then the samples were cured at laboratory conditions (24 °C, 53% RH) minimal for 48 h. Each set of prepared samples included 3 pieces with a specific concentration of nanoparticles.

## EXPERIMENT

### Relative Permittivity and Loss Factor Measurement

The real (relative permittivity) and imaginary (loss factor) parts of the complex permittivity were investigated using broadband dielectric spectroscopy (BDS). The aim of the broadband dielectric analysis is primarily to determine the general trends or changes in the development of relative permittivity and loss factor, as the results of measurements at a variable frequency of the applied electric field. A high-performance modular measuring system (Alpha-A, Novocontrol Technologies) with an active electrode system (type ZGS) was used for this purpose. The sample cell consists of two parallel cylindrical gold-plated electrodes, and test flat samples were placed between them. The diameter of the electrodes in direct contact with the sample was 20 mm and applied voltage was 1 V<sub>RMS</sub>.

The results of the loss factor and relative permittivity measurements are shown in Fig. 1. In the case of EPs, certain changes in relative permittivity can be observed. There is visible a positive effect of MgO addition, which results in a slight decrease in relative permittivity in the whole frequency range. Due to the proper incorporation of the particles into the epoxy resin, there is a strong adhesion. The dipolar groups of the nearest epoxy resin have reduced mobility and this fact reflects on the reduction of the influence of polarization processes [9]. We can see in Fig.1 - a), that the addition of higher filling content causes the increase of relative permittivity of the whole composite. The trend is visible in the loss factor behavior, as well. The higher the filling level the greater dielectric losses. For PUs, the behavior is different. It is most like due to the fact, that this mixture does not have perfect insulating properties. It is visible primarily in the region of low frequencies, where a higher conductivity of the insulation system is noticeable. However, the relative permittivity close to industrial frequency (50 Hz) was decreased by the addition of 1wt% of MgO. According to the results, the selected potting PU compound has a very limited range of applications.



**FIGURE 1.** Frequency dependence of dielectric parameters: a) relative permittivity of EPs + MgO composites, b) loss factor of EPs + MgO composites, c) relative permittivity of PUs + MgO composites, b) loss factor of PUs + MgO composites

## Volume Resistivity Measurement

The measurement of volume resistivity was performed according to Standard IEC 62631-3-1:2016. Prior to the measurements of absorption currents, the samples were short-circuited for 24 hours to eliminate the electrostatic charge. Precise multimeter 6517A and 8009 Resistivity Test Fixture (both Keithley Instruments) was used for this investigation. From the results presented in Fig. 2 - a) show, that incorporation of MgO has not a negative effect on the volume resistivity of EPs. This indicates proper sample preparation. Since no ultrasonic mixing was performed, there is a presumption of the presence of agglomerates, which, however, are evenly distributed and have not to impact on the charging (from the point of view of long-term stable conductivity current). The results for PUs composites presented in Fig. 2 – b) confirm the previous statements. For both cases, there is a change in volume resistivity in one order. Importantly, the addition of higher amounts of nanoparticles slightly reduces the internal resistivity in comparison with the previous filling level (presence of particles changes the conditions for trapping and de-trapping of charge in the internal structure [10]).

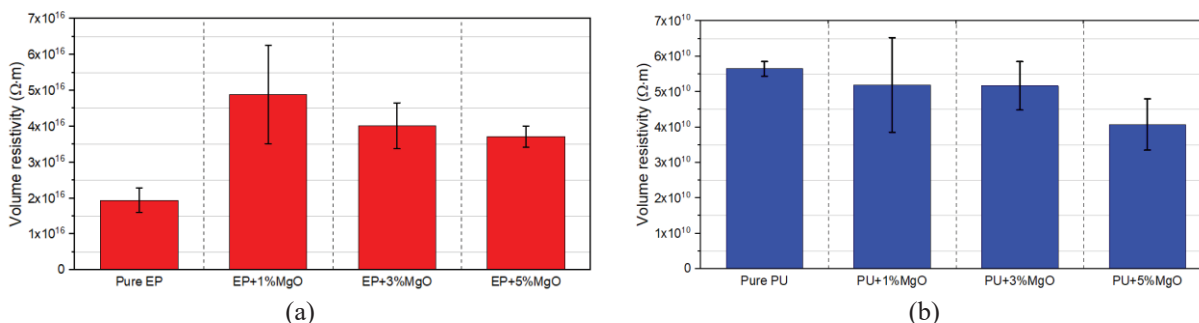


FIGURE 2. Changes in volume resistivity: a) EPs + MgO composites, b) PUs + MgO composites

### Differential Scanning Calorimetry Measurement

Differential Scanning Calorimetry (DSC Q2000 thermal analyzer, TA Instruments) was used to evaluate the glass transition of samples. Samples (approximately 7 mg) were placed into a pinhole aluminum cup and exposed to a linear heating rate of  $10\text{ }^{\circ}\text{C}\cdot\text{min}^{-1}$  ( $0\text{ }^{\circ}\text{C}$  to  $100\text{ }^{\circ}\text{C}$  for EPs, and  $-90\text{ }^{\circ}\text{C}$  to  $250\text{ }^{\circ}\text{C}$  for PUs) under a nitrogen atmosphere (flow rate of  $50\text{ ml}\cdot\text{min}^{-1}$ ). Measurement in reheating mode was performed for all samples. Cooling and reheating allow the original thermal history of the sample to be replaced by a standard and more controlled thermal history, thereby providing a more sensitive means of making distinctions between materials. The glass transition results in a step change in the heat versus temperature plot due to the change in heat capacity. In a plot of heat flow versus temperature, it is a gradual transition that occurs over a range of temperatures (step change). It is because amorphous materials are not in thermodynamic equilibrium but, with time, do relax and move towards equilibrium. It could be released during the first heating run (enthalpic recovery – occurs as the sample is heated to a temperature above  $T_g$ ). As has been already mentioned, the glass transition can be observed by a step change in the baseline of the measured signal. Generally, it could be characterized by its onset, midpoint, inflection, and endset temperature. The midpoint of step-change was used for the evaluation in this paper. The results presented in Fig. 3 show that the addition of MgO caused only minimal the changes in  $T_g$ .

According to the values of  $T_g$  and to the heat flow vs. temperature dependencies, more pronounced change in  $T_g$  was observed in the case of PU samples. In the case of PUs+MgO composites, nanostructured particles only slightly affect the soft segment glass transition temperature. According to study [11], it is probably due to the relatively low concentration of MgO nanoparticles in composites. For EPs+MgO can be stated, that the increased resin-filter interface created extra free volume and therefore aided in the large-scale segmental conversion of the polymer [12]. As a result, the glass transient temperature of EPs-MgO composites decreased with increasing filter loading, excepting 5 wt%.

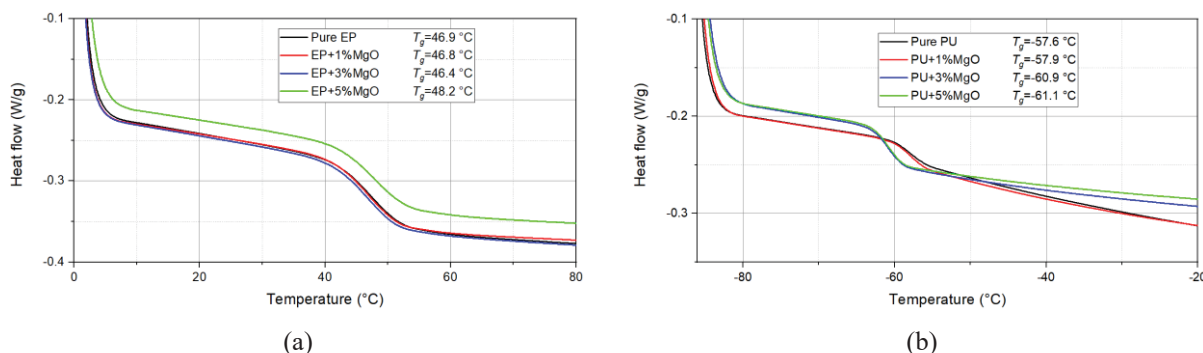


FIGURE 3. DSC curves for 2<sup>nd</sup> heating: a) EPs + MgO composites, b) PUs + MgO composites

## CONCLUSION

The main focus of this research was on the incorporation of nanometric magnesium oxide (MgO) in different weight ratios and the study of its effect on the properties of synthesized composites. From the performed investigations it is evident that the selected basic materials can be partially modified to improve their properties, especially in lower levels of filling, where there is a positive decrease of relative permittivity close to industrial frequency ( $\approx 3$  % for EP+1MgO;  $\approx 11$  % for PU+1MgO). For investigated composites is also evident a trend in the growth of dielectric losses. MgO also affects the volume resistivity and glass transition temperature.

## ACKNOWLEDGMENTS

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