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Impact of Ultrasonic Mixing on the Electrical Properties of PEI/SiO_2 Nanocomposites

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Abstract. The usage of nanoparticles in various applications is a widely discussed topic in the science community. One of these applications is dielectrics and the proposition of nanoparticles as modification of matrix material in composites. Epoxy resins are highly used resin material. However, a new type of resins composes of polyester-imide (PEI) is starting to take attention thanks to their advantageous base properties such as low viscosity under laboratory temperatures or electrical properties brought near the epoxy resins. This type of resin is still not examined as a matrix material with nanoparticles. This article deals with an experiment in which the main task was to determine the effect of SiO_2 nanoparticles added to Polyester-imide resin on the final composite properties. The effect of purely mechanical mixing was further investigated, followed by mixing using an ultrasonic tip. Based on the results of the experiment, it can be stated that the samples to which the ultrasonic tip was applied had visibly improved dielectric properties. Also, the production process was accelerated due to the reduction of air bubbles during sample casting. The investigated parameters were primarily frequency dependences of relative permittivity and dissipation factor. Electron microscope images are added to see the reduction of nanoparticle agglomerates.

INTRODUCTION

New materials are fundamental for developing new ways to produce power, make new products, or move forward as humanity. For the effective use of new materials, their properties must be deeply investigated. Suppose we focus on high voltage electrical machines. In that case, we find that composite materials formed of the matrix (commonly epoxy resin) and fillers (typically a combination of glass fibers and mica tapes) are only used insulating materials. [1-3] The investigations mainly focus on the variety of a new type of materials. One of them is nanocomposites, in which one of the fillers are the nanoparticles of one of the metal oxides like SiO_2, Al_2O_3, and others. [4-6] One of the main problems of the current insulating materials is their high thickness due to the still increasing demands on the operating voltage or higher operating temperature. This leads to an increased price of the overall insulating system. Current studies are showing that nanocomposites may be the answer to these problems. The compactness of the design would be another advantage of the nanocomposites. One of the unique properties of nanodielectrics is the specific surface area. This is the total area of the particle per unit mass or volume. This means that as the particle size decreases, the interaction zone increases. This mainly influences the bonding with the matrix material and the overall improvement of the final material properties. Problems start with the blending of nanoparticles into the matrix. The primary influence on the properties is the correct distribution of the nanoparticles. Agglomerates may influence the space charge phenomena and decrease the overall dielectric properties. [7]

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In this study, the nanoparticles of the SiO_2 were incorporated into polyester-imide resins, one of the new uninvestigated resin. One of the main goals of this is study was to investigate influence of the nanoparticles on the properties of the nanocomposite material and also find out if the ultrasonic addition into the preparation process influence the distribution of those nanoparticles in the matrix and final dielectric properties as well.

EXPERIMENT DETAILS

Nanocomposite material prepared and investigated in this study was consisted of two materials. Matrix was one component polyester-imide resin (ELAN-protect UP 343, company Elantas Italia) cured by temperature. This resin was selected because of its advantageous electrical and thermal properties and low viscosity property, which is beneficial for electrical rotation machines.

Based on the previous experiments, the nanoparticles of SiO_2 have been selected as the first filler to enhance dielectric properties. Commercially called AEROSIL R974 is a silicon dioxide particle with an average size of 10 nm. Nanoparticles of AEROSIL R974 have the chemical surface treatment by siloxane groups (Si-O-Si), which adds the hydrophobic properties and positively influences the dispersion of nanoparticles in resin. Preparation of samples includes mixing resin with nanoparticles and vacuuming to eliminate added air bubbles. After pouring the mixture into molds, they are placed in a dryer for gelation and curing process. In the first set of samples, the nanoparticles were mixed under 500 rpm and 60 °C for 4 hours. Then vacuumed for 2 hours under 50 rpm and 60 °C. In the second set of samples, the nanoparticles were mixed under the same conditions, but at the end of the mechanical mixing, the ultrasonic tip was added for 30 minutes. All samples were cured under 155 °C for 1 hour. Three sets of 5 samples were prepared, neat resin, resin without ultrasonic mixing, and resin with ultrasonic mixing. The 1 wt. % of SiO2 was used. Sets were named as UP (neat resin), UP4h (resin + mechanical mixing), UP4hU (resin + mechanical mixing + addition of ultrasonic). The diagram of sample preparation is visible in Fig. 1.

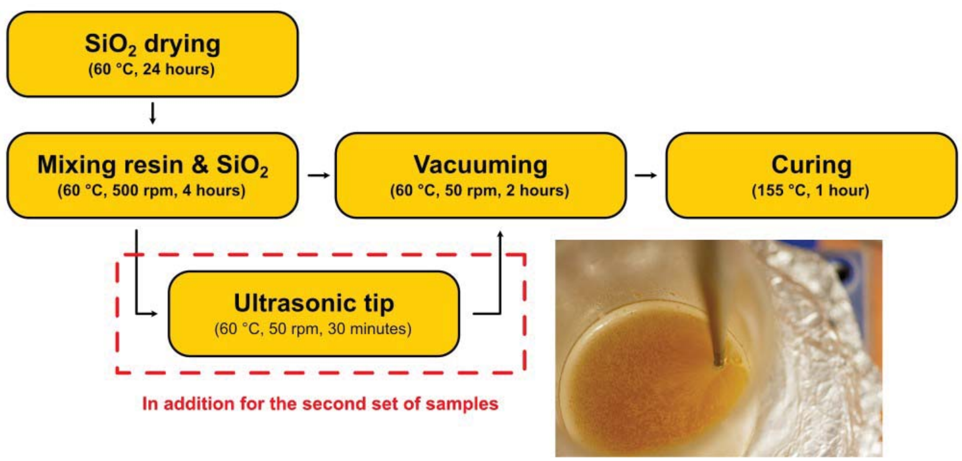

FIGURE 1. Diagram of the sample set preparation

Samples were diagnoses for multiple parameters. In this paper, the frequency dependence of the relative permittivity and dissipation factor is presented. Also, the SEM pictures are presented to compare the agglomeration dissipation on both sample sets. The frequency dependence of dielectric losses was measured by the broadband dielectric spectrometer from Novocontrol Industries was used applying 1 V. The frequency spectrum was 0.1 Hz to 10^6 Hz, and under various temperatures, the investigation was conducted (-50 °C to 90 °C). Absorption and resorption characteristics were also measured according to Standard IEC 62631-3-1:2016. Before the measurements, the samples were short-circuited for 24 hours to eliminate the electrostatic charge. An accurate multimeter Keithley 6517A and 8009 Resistivity Test Fixture was used for this measurement. The volume resistivity, polarization indexes (PI_1, PI_{10}), and area under the resorption curve (AURC) were calculated from these characteristics.

RESULTS AND DISCUSSION

The frequency dependence of the dielectric losses shows interesting results. Overall properties of the selected components are excellent. The hypothesis that the nanoparticles with only mechanical mixing incorporate the nonhomogenous parts into the samples, such as air bubbles or the agglomerates of the nanoparticles, was confirmed. The SEM images in Fig. 2 show the visible agglomerates even when we use 50 µm microscope resolution. Even with only mechanical mixing, the surface modification delivers a relatively good distribution of nanoparticles in the sample. However, when we use the ultrasonic tip, the agglomerates fully disperse.

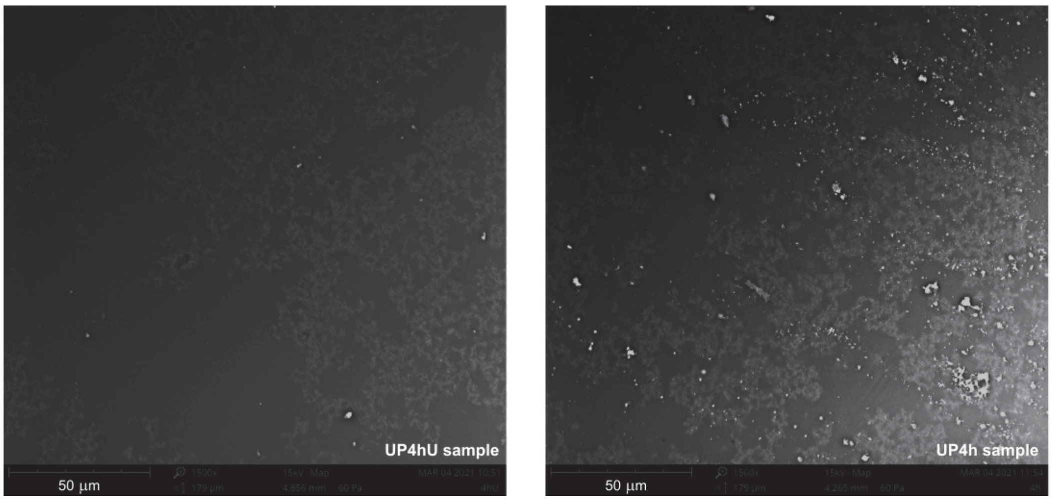

FIGURE 2. SEM of the samples (Left – UP4hU sample, Right – UP4h sample)

However, when we investigate the results of the dielectric parameters (Figs. 3, 4, 5), we see that the only mechanical mixing for four hours is not enough. Overall properties of this type of resin material (volume resistivity $1.9 \cdot 10^{15}$, relative permittivity 3, dissipation factor 10^{-3}, and dielectric strength 32 kV/mm) are suitable for high voltage application. The ultrasonic tip was able to disperse nanoparticles and remove agglomerates that deformed the electrical field and incorporate unwanted space charges, increasing dielectric losses. From the Fig. 3 and Fig. 4 we can see that nanoparticles, processed through the ultrasonic tip, significantly reduce the values of dissipation factor and relative permittivity under higher temperatures.

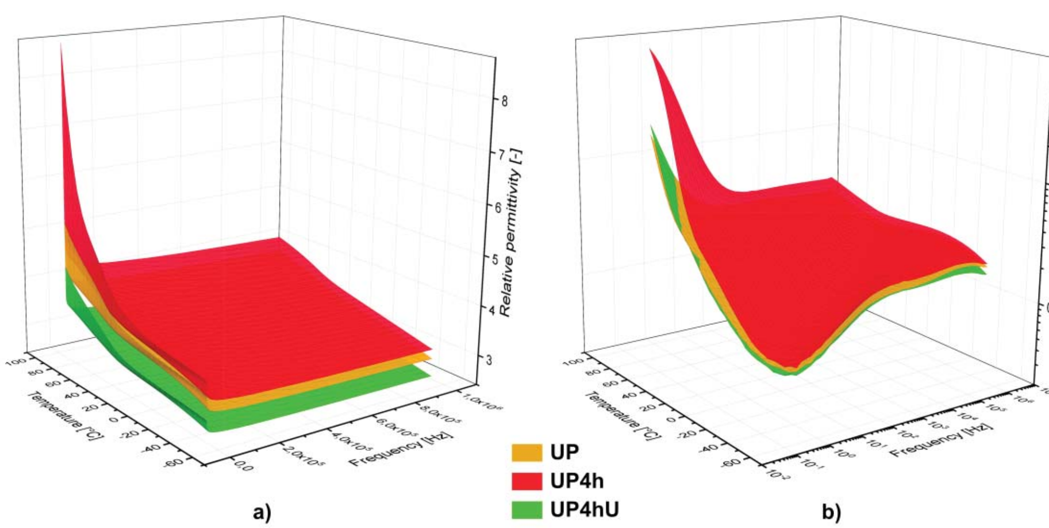

FIGURE 3. 3D plots showing the difference while the ultrasonic tip is incorporated into the process. a) relative permittivity; b) dissipation factor

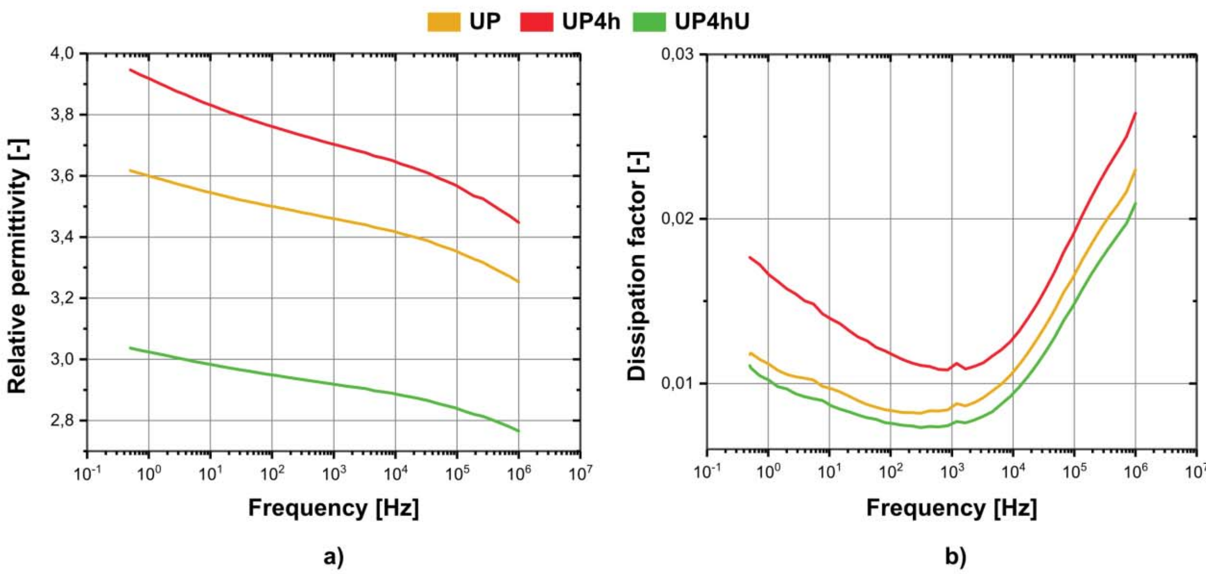

FIGURE 4. Frequency dependence of the dielectric parameters under laboratory conditions. a) relative permittivity; b) dissipation factor

If we investigate the results of the absorption and resorption characteristics, visible in Fig. 5, the similarities can be seen with the frequency dependencies. By incorporating the nanoparticles, we can see that the volume resistivity decreases by applying only the mechanical mixing, but the volume resistivity increases if the ultrasonic tip is included. We can conclude that the agglomerates have a significant role in decreasing the volume resistivity and polarization indexes. For the UP4h, the volume resistivity is $7.1 \cdot 10^{14}$, PI_1 is 2.3, and PI_{10} is 3.2. For the UP4hU, the volume resistivity is $5.03 \cdot 10^{15}$, PI_1 is 3.05, and PI_{10} is 5.95. Based on the results of the resorption characteristics, we can conclude that the agglomerates increase the discharge time of the samples. AURC of the UP4h is more than double of the UP4hU, 3.5 and 1.3, respectively. The lower the values, the faster the discharge is done.

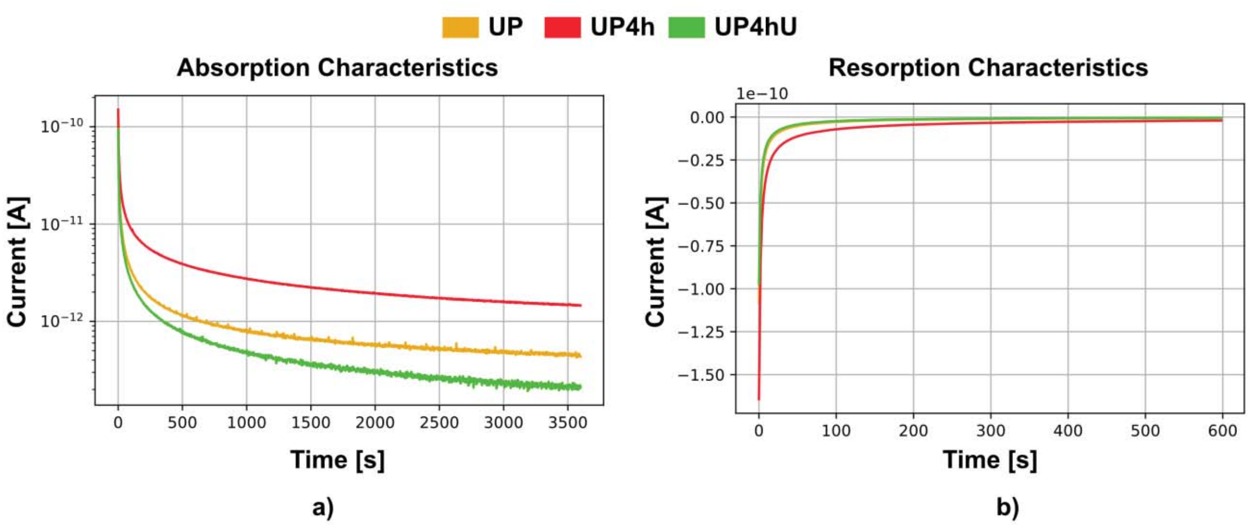

FIGURE 5. Change of the characteristics based on the material under laboratory conditions. a) absorption characteristics; b) resorption characteristics

CONCLUSION

The main focus of this experiment was the confirmation of the positive effect of the application of ultrasonic tip on the samples of nanocomposite material composed of a modern type of Polyester-imide resin and the SiO_2 nanoparticles. Based on the presented results of the dissipation factor, relative permittivity, and the absorption and resorption characteristic, supported by the SEM images, we can confirm that the ultrasonic tip decomposes the agglomerates of the nanoparticles. This has a visible influence on the dielectric properties of the resin samples. Also, it helps to reduce the amount of air bubbles during the curing process, which also influences all mentioned dielectric parameters. By applying the ultrasonic, we can also reduce the time for sample preparation by decreasing the mechanical mixing of the nanoparticles and the resin.

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