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The influence of barium titanate as a filler in impregnating epoxy resin on chosen electrical parameters of obtained material

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The paper describes the influence of barium titanate $BaTiO_3$ used as a filler in impregnating epoxy resin on electrical parameters of obtained material. The results of computer simulation of electric field intensity distribution in area of 220 kV ac composite insulator show that in order to reduce the maximum value of this field it is necessary to use the rod with the highest possible permittivity. It was assumed that the increase of rod permittivity is possible by doping the epoxy resin, being one of glass-epoxy rod components, with a filler having high permittivity. Barium titanate $BaTiO_3$ in micro- and nanograins was used as a filler. Presumably, such a change in material structure, may negatively influence other electrical parameters of the material, important in composite insulators construction. In order to check the filler dispersion in epoxy matrix, microscopic observations of fractions of the obtained materials were performed. In a wide range of frequencies, the permittivity and volume resistivity of the materials were determined. The inception voltage of partial discharges on surface of materials was determined by the electric, acoustic and antenna methods.

Key words: *nanofillers; $BaTiO_3$; epoxy resin; permittivity*

1. Introduction

Usually, a composite insulator is made of glass-epoxy resin rod with the relative permittivity 5 and covering material (silicone rubber) with the relative permittivity 3. The results of last years investigations show that a suitable modification of dielectrics with nanofillers will presumably make steering of material permittivity possible.

The modification of dielectrics relies on introduction of suitable nanofillers to material matrix. A nanofiller is a polycrystalline substance built-up of grains with

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a diameter of at most hundred nanometres. The popularity of use of nanofillers grows thanks to specific nanoscale phenomena and not present in materials with fillers of micrometric grains. Thus, the use of microfillers may lead to obtainment of materials with properties better or other than the properties of microfilled materials with the same chemical composition. The right choice of a nanofiller permits to improve, among others, electrical, mechanical or thermal properties of dielectrics. The best known nanofillers are alumina Al_2O_3, titania TiO_2, silica SiO_2, barium titanate $BaTiO_3$, zinc oxide ZnO and ferric oxide Fe_2O_3.

Electrical strength, resistance to partial discharges and appearance of conducting paths due to partial discharges, dielectric loss factor, resistivity or permittivity are the electrical properties which may be changed by nanofiller addition [1–12].

2. Objective and scope of investigations

The increase of permittivity, with preservation or improvement of other electrical parameters and remaining of impregnating properties of epoxy resin, was the goal of the researches. The increase of permittivity was realised by addition of barium titanate in form of micro- and nanograins as a filler. In this way, three materials were disposed: epoxy resin without filler, epoxy resin with micrograins of barium titanate and epoxy resin filled with nanograins of $BaTiO_3$.

A stress should be put on fact that the change of dielectrics permittivity in composed insulating systems ameliorates electric field distribution and improves work conditions of these systems. Still, it is important to remember that in the case of simple insulating systems, the conditions in triple junction (electrode–dielectric–air) may become worse. An increase of electric field intensity in triple junction should be expected. It consequences in partial discharges inception voltage and flashover voltage decrease. In order to carry out quantitative estimation of these effects, surface partial discharges and flashover voltage of investigated materials were determined.

The permittivity and resistivity of obtained materials were determined in a wide frequency range (from 0.001 up to 1000 Hz). On surfaces of obtained materials flashover voltage and partial discharges inception voltage were measured. The latter was measured with the use of classical electric, acoustic emission and ultra high frequency methods.

The observations of material fractures were performed with scanning electron microscope (SEM) in order to check the fillers dispersion uniformity in epoxy resin.

3. Investigation objects

The samples of impregnating epoxy resin without a filler, filled with micrograins of barium titanate (64 wt. %), and filled with nanograins of barium titanate (32 wt. %) were investigated. In order to measurably increase the permittivity epoxy resins, addition of the highest possible quantity of barium titanate was overtaken, at condition that impregnating properties of the resin were preserved. As a consequence, epoxy resin samples with 64 wt. % of micrograins of barium titanate and with 32 wt. % of nanograins of barium titanate were obtained.

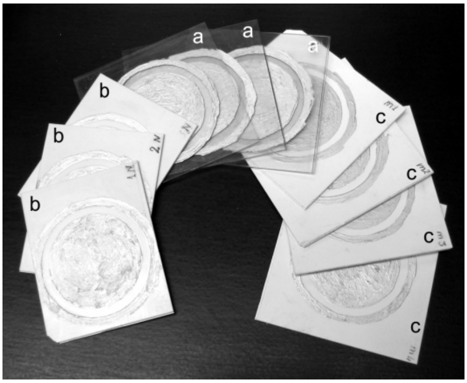

Fig. 1. Samples of impregnating epoxy resin: without filler (a), with nanograins (b) and micrograins (c) of barium titanate

At least three samples of each material were fabricated. They were 2 mm thick with the dimensions of 10×8 cm^2 or 10×10 cm^2 (Fig. 1).

4. Results

The main goal of researches was to obtain materials with increased permittivity at 50 Hz. However, the permittivity was determined with Programma IDA 200 device in much wider range of frequency (from 0.001 Hz up to 1000 Hz). Average values of the permittivities are shown in Fig. 2. As can be seen, they are steady in the whole range of frequencies. At 50 Hz, unfilled epoxy resin has the relative permittivity equal to 3.9. The addition of nanofiller to epoxy resin caused the increase of permittivity to 5.4, and the addition of microfiller increased the permittivity to 12.7.

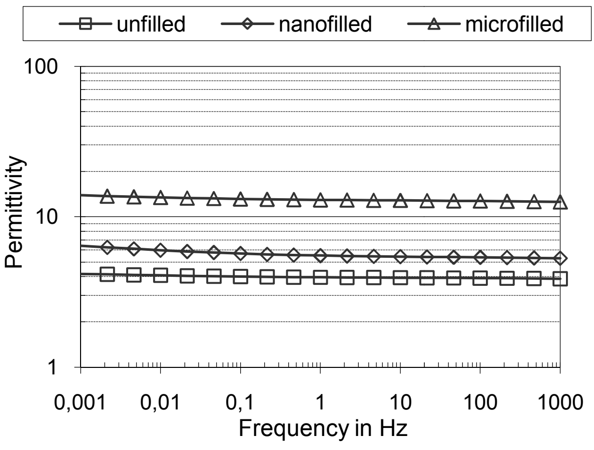

Fig. 2. Permittivity of investigated materials as a function of frequency

The results obtained for samples of epoxy resin with the nanofiller are surprising for two reasons. Firstly, expected growth of permittivity, usually caused by the use of nanofillers, was not observed. Secondly, the increase of permittivity was not comparable even to expected growth caused by use of the same amount of microfiller. Unexpected results of permittivity measurements inclined observations of the filler dispersion in the epoxy matrix. Inadequate dispersion of the filler in matrix influences the properties of the material. The simplest microscopical method, observation of fractures in SEM, was employed. A fracture of epoxy resin without a filler, being a basis for comparison of the filler distribution, is shown in Fig. 3.

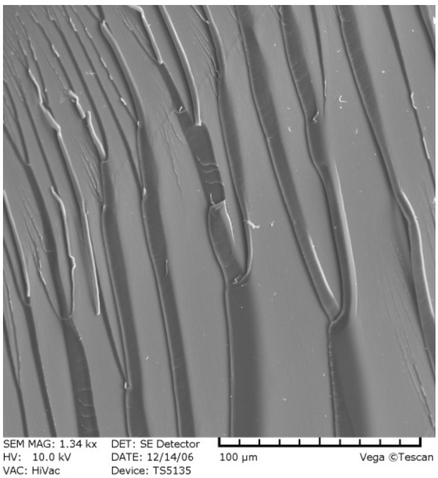

Fig. 3. Fracture of epoxy resin without fillers

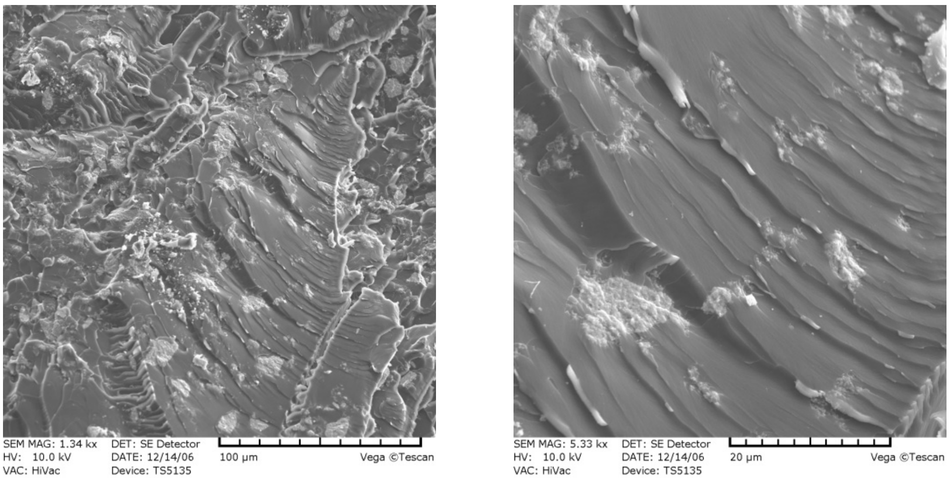

Fig. 4. Fracture of epoxy resin with barium titanate nanograins at two different magnifications

The fracture of the sample containing nanofiller grains is presented in Fig. 4. It is evident that the distribution of barium titanate is nonuniform and nanograins form agglomerates of greater dimensions, up to more than ten microns. This phenomenon was probably caused by lack in equipment used for the sample preparation. It was not the case for epoxy resin with the microfiller, where distribution of grains is quite uniform as is shown in Fig. 5. Nonuniform distribution of filler nanograins has evidently caused the surprising results of the measurement of electric permittivity.

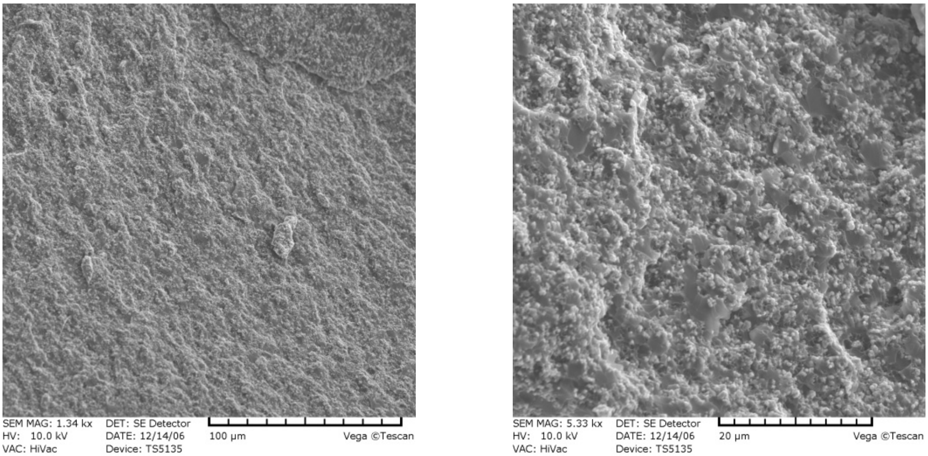

Fig. 5. Fracture of epoxy resin with barium titanate micrograins at different magnifications

The measurement of dielectric loss factor and volume resistivity was carried out with electrodes sputtered on surface of material as is shown in Fig. 1.

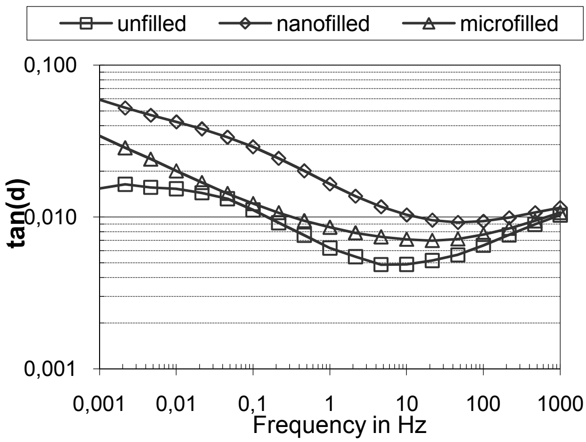

Fig. 6. Average tan(δ) of investigated materials as a function of frequency

The measured values of average tan(δ) of three analysed materials are shown in Fig. 6. At 50 Hz, the loss factor of unfilled resin was 60×10^{-4}. The addition of the microfiller caused a slight growth up to 70×10^{-4}, while the addition of the nanofiller increased tan(δ) up to 90×10^{-4}. As can be seen, the addition of fillers caused only a small growth of the dielectric loss factor.

The results of measurements of the volume resistivity are shown in Fig. 7. As can be seen, irrespective of the filler grain size, their introduction to epoxy resin matrix caused the decrease of the volume resistivity at most of one order of magnitude.

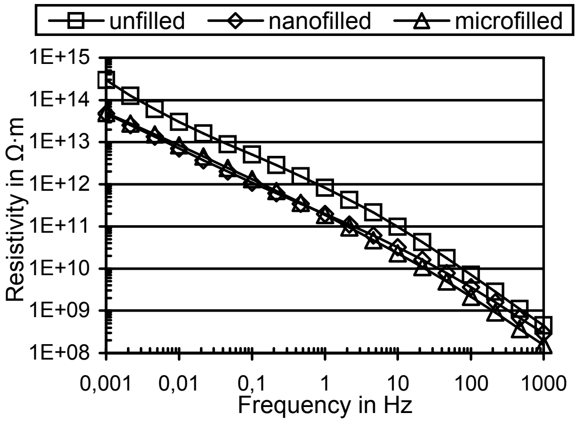

Fig. 7. Volume resistivity of investigated materials in function of frequency

In systems layered in series, permittivity influences distribution of the electric field. Basically, its intensity is higher in materials with lower permittivity. Thus, in simple systems, the increase of permittivity of epoxy resin should cause the raise of electric field intensity in air. Consequently, surface partial discharges inception voltage and flashover voltage should decrease. For these reasons, partial discharges inception voltage and flashover voltage were determined.

Both in the case of partial discharges inception voltage and flashover voltage, cylindrical electrodes were placed in the middle of the investigated sample, 20 mm one from another, as shown in Fig. 8.

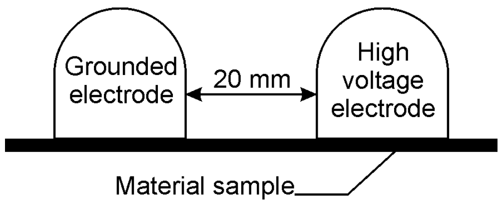

Fig. 8. Electrodes set-up for partial discharges measurement

Flashover voltage was equal to 19.5 ± 0.7 kV on samples of epoxy resin without a filler, 20.0 ± 0.8 kV for samples with the nanofiller and 16.3 ± 0.6 kV with the microfiller. As can be seen, the introduction of barium titanate nanograins did not cause changes of the flashover voltage, whereas the use of microfiller reduced it by ca. 15%.

In partial discharges inception voltage measurements, for the sake of security, the maximum voltage value was equal to 13 kV, which is 80% of the lowest flashover voltage on the surface of investigated materials.

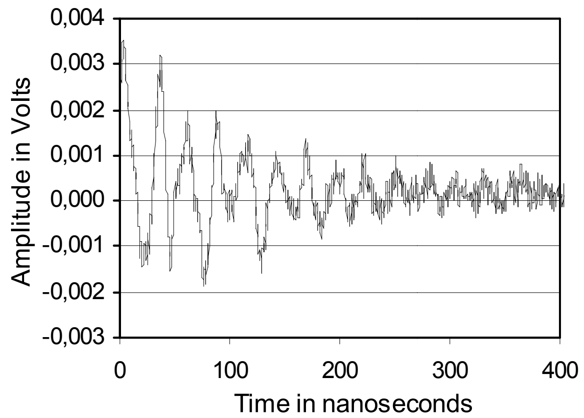

Fig. 9. Example of a partial discharge impulse on surface of epoxy resin with microfiller, registered with UHF probe

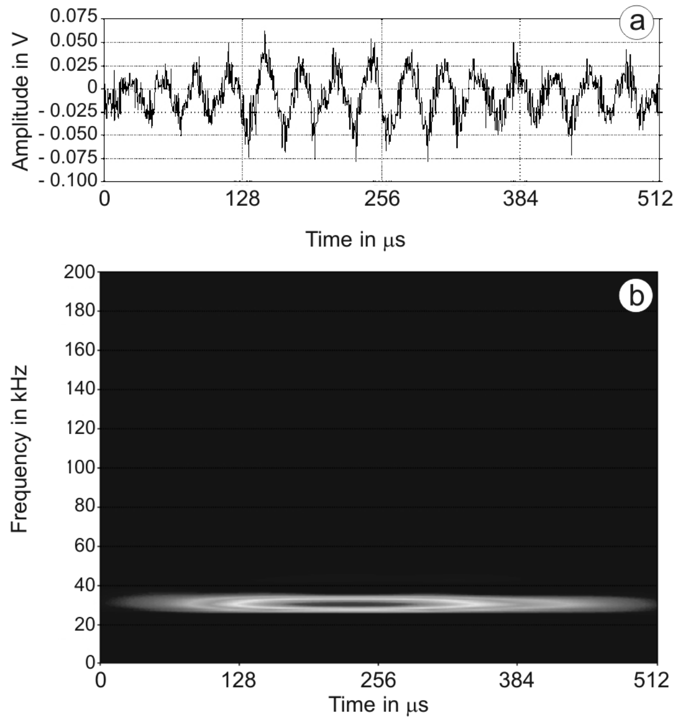

Fig. 10. Chosen partial discharge impulse recorded with an acoustic sensor (a) and its analysis by continuous wavelet transform (b)

The inception voltage of partial discharges developing on surfaces of investigated materials was determined by three independent methods: classical electric, acoustic emission and UHF. In the cases of epoxy resin without filler and filled with nanograins of barium titanate, up to 13 kV no partial discharges were observed. In the case of epoxy resin with a microfiller, some partial discharges were recorded. The inception voltage, measured by the applied method, was equal to 12.0 ± 0.9 kV. An example of a partial discharge impulse, recorded with a UHF probe is shown in Fig. 9. Evaluated time parameters of the impulses are typical of surface partial discharges [13].

A chosen impulse recorded with an acoustic sensor is shown in Fig. 10a, and its joint time-frequency analysis (continuous wavelet transform spectrogram) is shown in Fig. 10b. The frequency parameters of the signal confirm that it was generated by a surface partial discharge [14].

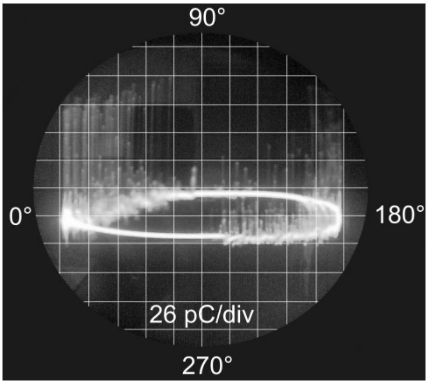

Fig. 11. Oscillogram of partial discharges obtained with classical electric method

An oscillogram of partial discharges obtained by the classical electric method is shown in Fig. 11. It can be seen that the current impulses were generated on leading edge of sinusoid positive and negative halves, which confirms that they were generated on material surface [14]. The biggest charges carried by a single discharge were up to 100 pC. The partial discharges inception voltage and flashover voltage were the lowest in the case of material with the highest measured permittivity (epoxy with 64 wt. % of the microfiller).

5. Conclusions

Doping of impregnating epoxy resin with barium titanate came out an efficient way of increasing the permittivity. Addition of 64 wt. % of micrograins of the filler caused growth of relative permittivity from less than 4 up to almost 13. Doping the epoxy resin with micrograins of barium titanate caused higher increase of permittivity than in the case of addition of nanograins. It was caused probably by the fact that micrograins were dispersed better than nanograins. An additional factor influencing the permittivity of materials under investigation was a possibility of introducing a higher amount of micrograins (64 wt. %) than that of the nanograins (32 wt. %) of barium titanate to epoxy matrix with its impregnating properties preserved. However, the most important factor was probably the increase of the density of polymer–filler bonds in the nanocomposite, related to the growth of the matrix–filler interface. It could impede movements of chain segments and of lateral chain segments, causing the decrease of permittivity resulting in considerably higher increase of permittivity of obtained material when the microfiller was used.

The results of permittivity measurements were confirmed by the determined values of the partial discharges inception voltage and flashover voltage. In the case of microfilled epoxy resin, these two quantities measurably decreased. This means that the permittivity of this material was higher than the permittivity of remaining two ones. Basic electrical parameters, important in aspect of insulating techniques, were not considerably decreased.

Dielectric loss factor tan(δ) increased from 60×10^{-4} (epoxy without filler) to 70×10^{-4} (epoxy filled with barium titanate micrograins) or 90×10^{-4} (epoxy with barium titanate nanograins). Volume resistivity decreased by about half order of magnitude.

Acknowledgements

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