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Breakdown and partial discharges in magnetic liquids

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Abstract

The dielectric properties (permittivity, loss factor, dielectric breakdown strength) of magnetic liquids were investigated. The magnetic liquids were composed of magnetite particles coated with oleic acid as surfactant and dispersed in transformer oil. To determine their dielectric properties they were subjected to a uniform magnetic field at high alternating electric fields up to 14 MV m^{-1}. Nearly constant permittivity of magnetic liquid with particle volume concentration $\Phi = 0.0019$ as a function of electric field was observed. Magnetic liquids with concentrations $\Phi = 0.019$ and 0.032 showed significant changes of permittivity and loss factor dependent on electric and magnetic fields. The best concentration of magnetic fluid was found at which partial current impulse magnitudes were the lowest. The breakdown strength distribution of the magnetic liquid with $\Phi = 0.0025$ was fitted with the Duxbury–Leath, Weibull and Gauss distribution functions.

1. Introduction

Insulating and thermal properties of transformer oils limit the minimal size and maximal transformer power [1, 2]. In comparison with pure transformer oils, magnetic liquids with transformer oil as carrier base provide better heat removal. Magnetic liquids are suspensions of very fine magnetic particles of the order of 10 nm. The presence of foreign particles in transformer oils influences their electric strength. Particles which have dielectric permittivity ϵ_p different from the permittivity ϵ_l of insulating liquid cause local changes of electric field. Electric field E_p in spherically shaped particles occurring in a homogeneous polarized transformer oil is not equal to the electric field E_l in an oil:

$$E_p = \frac{3\epsilon_l}{2\epsilon_l + \epsilon_p} E_l.$$

The decrease of field in particles (in the case of $\epsilon_p > \epsilon_l$) results in an increase of electric field in regions between particles. Changes of electric field result in a decrease of partial discharge inception voltage and breakdown voltage. Changes of local electric fields are higher if clusters of particles are formed. Polarizable particles which are of higher permittivity than the surrounding liquid are attracted to regions of maximum electric field. Aggregation of particles results in the formation of needle-like clusters oriented in the direction of electric field. This can initiate breakdown, for example Duxbury and Leath [3] suggested a theory of electric breakdown in a mixture of metal and dielectric based on metal particle cluster formation. Breakdown voltage is determined by the highest local field in the dielectric which is indirectly proportional to the length of the biggest cluster in this theory. In homogeneous external electric field distribution it is possible to describe the distribution of breakdown electric field by the function $P_{DL}(E)$:

$$P_{DL}(E) = \frac{kcd^3}{E^2} \exp\left(-cd^3 \exp\left(-\frac{k}{E}\right)\right) \exp\left(-\frac{k}{E}\right)$$

or by so called Weibull function $P_W(E)$:

$$P_W(E) = mcd^3 E^{m-1} \exp\left(-cd^3 E^m\right)$$

in this model where k, c and m are constants, d is electrode distance and E applied electric field intensity. Cluster

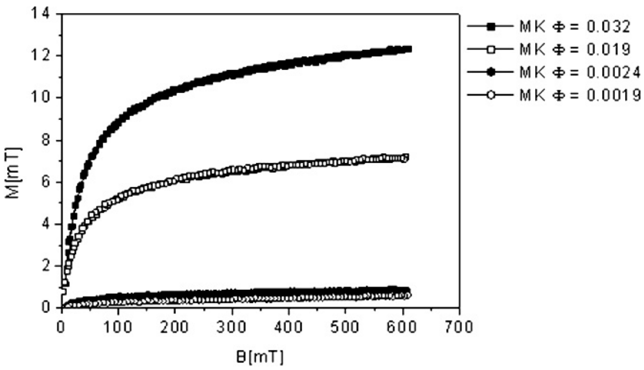

Figure 1. Dependence of magnetization of magnetic liquids on applied magnetic field.

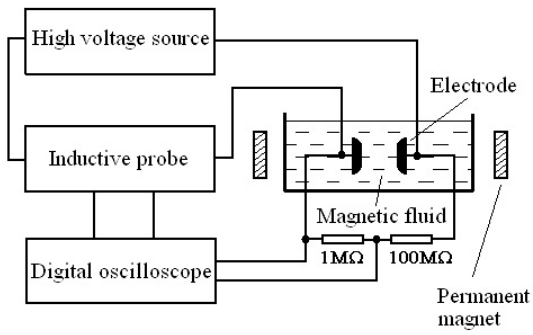

Figure 2. Experimental setup for measurements of time development of breakdown.

formation in a volume of a magnetic liquid influences the permittivity of the liquid. It is known that the permittivity of magnetic liquids is dependent on the direction of applied magnetic field, in what is called the magnetodielectric effect. This effect is also caused by the formation of chain-like clusters of magnetic particles in the presence of magnetic field. They are oriented in the direction of the magnetic field. Magnetic field applied perpendicular to electric field decreases the permittivity of a magnetic liquid but magnetic field applied parallel to electric field increases the permittivity of a magnetic liquid [4]. It is known that also high electric fields initiate cluster formation [5]. The motivation of this work is to investigate dielectric properties of magnetic liquids in high electric fields in the presence of a magnetic field that usually occurs in transformers. Results of the investigation of the magnetodielectric effect in high applied electric fields in a magnetic liquid with magnetite particles dispersed in a transformer oil, dielectric loss factor, breakdown electric field strength distribution and time development of breakdown are reported.

2. Experimental details

The magnetic liquids composed of transformer oil ITO 100 and Fe_3O_4 particles coated with oleic acid were prepared. Magnetic particles were obtained by chemical precipitation of ferrous and ferric salts by NH_4OH. Oleic acid (as a surfactant) was added after washing and water removing at the temperature 70 °C and then ITO 100 was added drop-wise during stirring. Volume concentrations of magnetite particles in magnetic liquids, the average diameter of magnetite particles and the standard deviation of its determination were carried out by vibrating sample magnetometer (VSM) measurements. Magnetization loops were measured in the range of 0–600 mT at room temperature for concentrations up to $\Phi = 0.032$. Superparamagnetic behaviour of magnetic fluids was confirmed for all investigated concentrations of magnetic particles (figure 1). Structuralization effects at $\Phi = 0.0024$ and 0.0019 were not observed by an optical microscope in static magnetic fields up to 50 mT. Permittivity and loss factor were measured by the Schering bridge Tettex 2818 at frequency 50 Hz. A capacitor was composed from parallel plate Cu electrodes placed in a container. The electrodes were 1 cm in diameter and the distance between them was 0.8 mm. Capacity and loss factor of the capacitor were measured as a function of the applied electric field intensity in the range of 0.5–2.5 MV m^{-1}. The experimental error of capacity measurements was 0.05% and of loss factor was 1%. Permittivity of magnetic liquids was determined from capacity measurements as

$$\epsilon_r = \frac{C}{C_0}.$$

where C is the capacity of the capacitor with a magnetic liquid as a dielectric and C_0 is the capacity of the same capacitor filled by air. Two permanent NdFeB magnets were used as a source of homogeneous magnetic field up to 40 mT. Dielectric breakdown strength of samples and time development of current flowing between the electrodes of the capacitor during breakdown development were measured with the equipment shown in figure 2. An alternating voltage source 0–5 kV UH27 with frequency 50 Hz was used. Homogeneous electric field was produced by copper electrodes with Rogowski profile. The electrodes were 1.5 cm in diameter and their distance could be changed in the range of 0.05–1 mm. Time development of discharge current was measured by an inductive probe and recorded by a digital programmable oscilloscope, LeCroy Waverunner LT344.

3. Results and discussion

Magnetite particles prepared by chemical precipitation have nearly spherical shape (figure 3). The hydrodynamic average diameter of magnetite particles and its standard deviation determined by TEM are 9.1 nm and 0.3. Parameters of log-normal distribution of particle sizes (figure 4) were also determined by the method of Chantrell *et al* [6]. The average diameter of particles in the used liquids was 8.5 nm and standard deviation $\sigma = 0.34$. In an electric field of 0.5 MV m^{-1} the permittivity of magnetic liquids with concentrations $\Phi = 0.0019$ and 0.019 is approximately the same as the permittivity of pure transformer oil but an increase of concentration to $\Phi = 0.032$ causes increasing permittivity. It is supposed that permittivity increase is caused by higher magnetic particle concentration which results in cluster creation and space charge formation at the

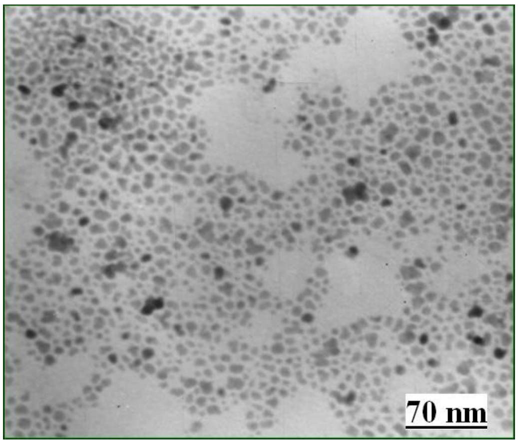

Figure 3. TEM picture of magnetite particles prepared by the chemical precipitation method.

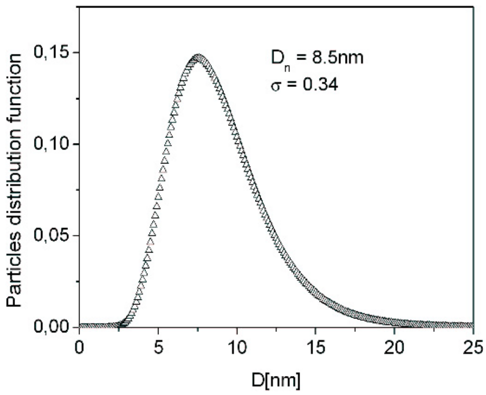

Figure 4. Particle size distribution function of the used magnetic liquid.

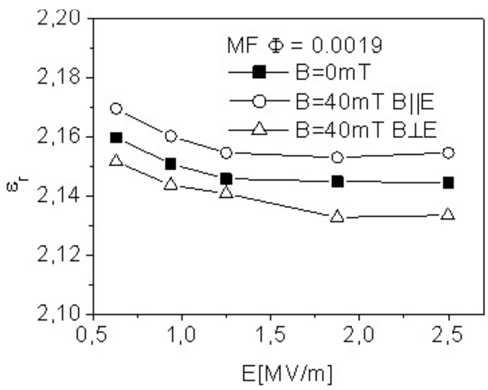

Figure 5. Dependence of permittivity ϵ_r of the magnetic liquid ($\Phi = 0.0019$) on electric field intensity E.

electrode–liquid interface. The investigations showed that the permittivity of the magnetic liquid with the lower concentration $\Phi = 0.0019$ was weakly dependent on the electric field intensity in the range of 0.5–2.5 MV m^{-1} but the permittivity of the magnetic liquids with higher concentrations ($\Phi = 0.019$ and 0.032) increased significantly with increasing electric field intensity in the electric fields in the studied range (figures 5–7).

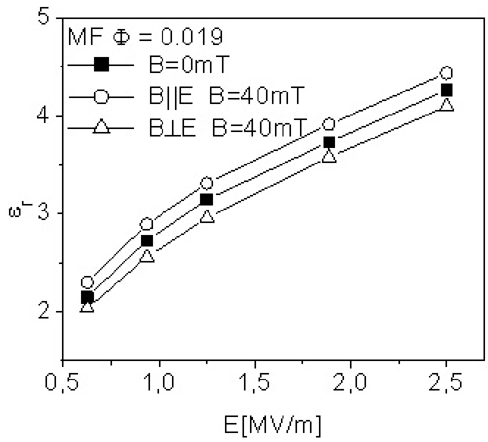

Figure 6. Dependence of permittivity ϵ_r of the magnetic liquid ($\Phi = 0.019$) on electric field intensity E.

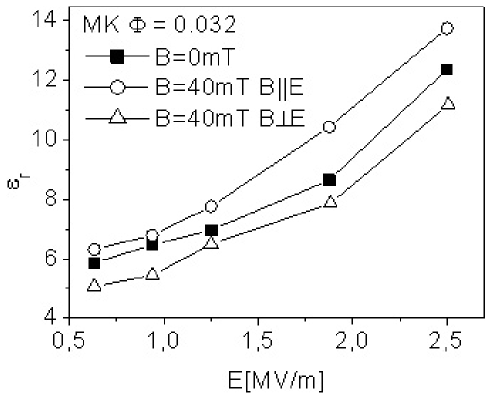

Figure 7. Dependence of permittivity ϵ_r of the magnetic liquid ($\Phi = 0.032$) on electric field intensity E.

We assume that the changes of magnetic liquid permittivity dependence on electric field are caused by cluster formation. Magnetite particles coated with oleic acid as a surfactant are electrically charged by adsorbed ions and counter-ions from the surrounding atmosphere may be attracted to them [7]. Counter-ions are ions oppositely charged to ions adsorbed by particles. Electric dipole moments are induced in electric field.

Increasing electric field increases electric dipole–dipole interaction between particles and supports their agglomeration. The concentration of magnetic liquid $\Phi = 0.0019$ is insufficient to cause aggregation opposite to magnetic liquids with concentrations $\Phi = 0.019$ and 0.032. The decrease (in the case of $\mathbf{B} \perp \mathbf{E}$) and increase (in the case of $\mathbf{B} \parallel \mathbf{E}$) of permittivity in applied magnetic and electric fields was observed by what is known as the magnetodielectric effect. We suppose that particles in magnetic liquids have nearly spherical shape. Our assumption is based on previous investigations, when particles prepared by the same method as used here had approximately spherical shape. In magnetic liquids with spherically shaped particles, there exist critical frequencies above which the magnetodielectric effect appears [4]. The critical frequency of transformer oil based magnetic liquid with spherically shaped magnetite particles coated with oleic acid (specific density of sample is 1470 kg m^{-3} and magnetization

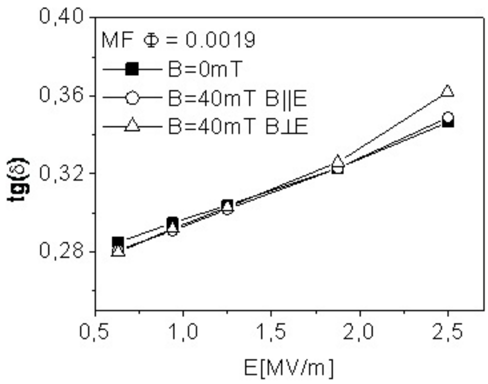

Figure 8. Dependence of loss factor tg(δ) of the magnetic liquid on electric field intensity E.

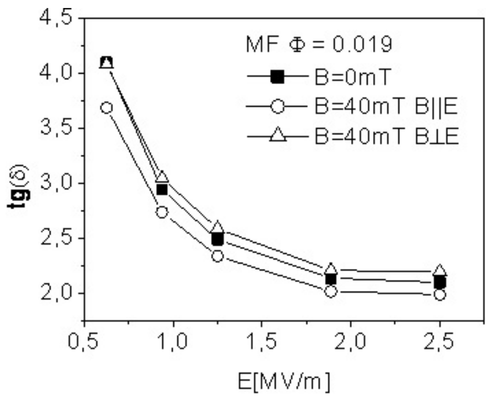

Figure 9. Dependence of loss factor tg(δ) of the magnetic liquid on electric field intensity E.

at $H = 8 \times 10^4$ A m^{-1} is $M = 25.2 \times 10^3$ A m^{-1}) reaches a value of 55 Hz and the critical frequency of kerosene based magnetic liquid is lower at lower specific density [4] therefore we suppose that the critical frequency of our magnetic liquid (specific density is 919 kg m^{-3}, magnetization at $H = 8 \times 10^4$ A m^{-1} is $M = 4.1 \times 10^3$ A m^{-1}) is lower than the used frequency of 50 Hz. As shown in figures 8–10 the loss factor slowly increased at $\Phi = 0.0019$ and decreased with increasing electric field intensity at the higher volume concentrations of magnetic particles. The maximum of loss factor of the magnetic liquid with $\Phi = 0.019$ is in agreement with the investigation of Rădulescu [7], who found that kerosene based magnetic liquid with magnetite particles coated with oleic acid has a very high maxima of tg(δ) at frequencies lying in the range 50–250 Hz and the variation of tg(δ) maxima with Φ has an inverted-U shape. The losses were evaluated using the loss factor tg(δ) and permittivity ϵ_r:

$$P = \omega \epsilon_r \epsilon_0 \,\mathrm{tg}\,(\delta) E^2$$

where ϵ_0 is the permittivity of a vacuum, ω angular frequency and E electric field intensity. It was found that losses increase with increasing electric field nearly quadratically and increase with particle volume concentration (figure 11). Losses of magnetic liquid with $\Phi = 0.0019$ are substantially lower than for magnetic liquid with $\Phi = 0.019$ and 0.032.

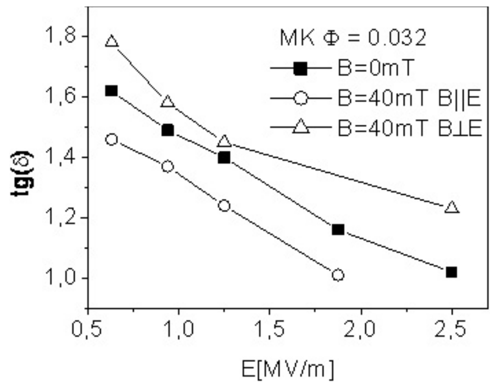

Figure 10. Dependence of loss factor tg(δ) of the magnetic liquid on electric field intensity E.

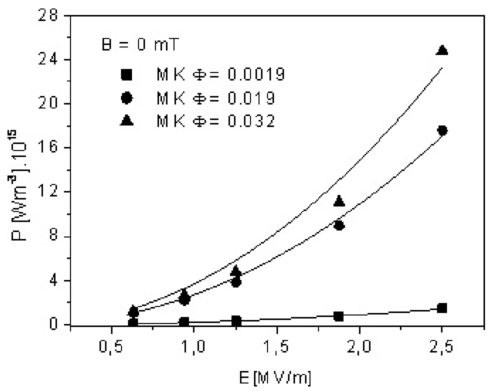

Figure 11. Losses P of the magnetic liquid as a function of the electric field intensity E and fitted curves.

Partial discharges contribute to losses and cause progressive degradation of magnetic liquid. The ratio of current impulses occurring before breakdown ΔI and current impulses at concentration $\Phi = 0.0024$ as a function of magnetic particle concentration Φ is shown in figure 12. Partial discharge measurements show that current impulses are the smallest at concentration $\Phi = 0.0024$ at all investigated electrode distances d. Oil based magnetic liquid has better insulating properties than the pure transformer oil if the concentration of magnetic particles does not exceed the value $\Phi = 0.01$ [2]. To investigate the form of breakdown strength distribution in ac conditions a series of 65 measurements of breakdown strength of magnetic liquid with $\Phi = 0.0025$ was carried out at an operation frequency of 50 Hz and electrode distance $d = 0.1$ mm (figure 13). Breakdown field distribution was fitted with the Weibull, Gauss and Duxbury–Leath function, which was proposed to describe breakdown electric field strength distribution in metal loaded dielectrics. It is not possible to determine from the performed experiments which of the functions (Duxbury–Leath, Weibull, Gauss) better describes the breakdown electric field distribution.

4. Conclusions

The magnetodielectric effect in magnetic liquids was confirmed in high electric fields 0.5–2.5 MV m^{-1} at a magnetic

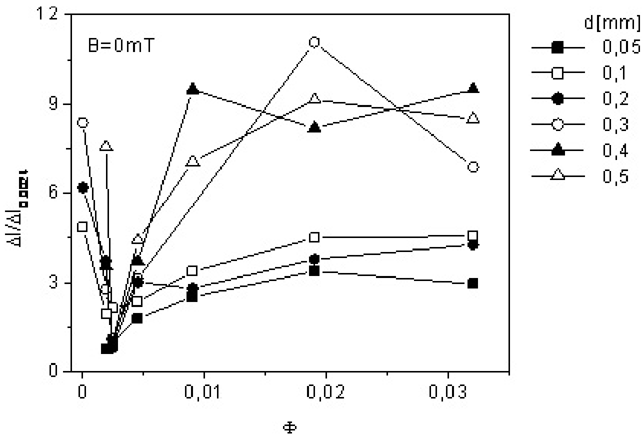

Figure 12. Maximum size of current impulses during breakdown development as a function of particle volume concentration.

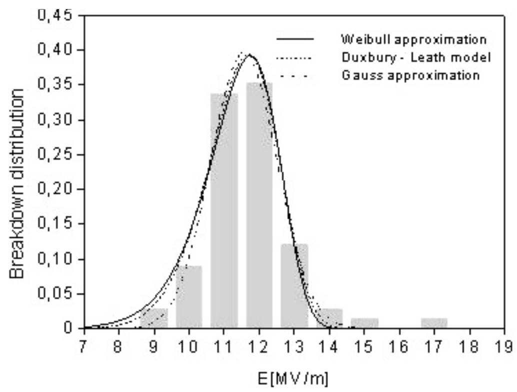

Figure 13. Breakdown electric field distribution fitted with the Weibull, Gauss and Duxbury–Leath functions.

field of value 40 mT. It has been shown that the permittivity of the magnetic liquid with the small volume concentration of magnetic particles ($\Phi = 0.0019$) was nearly constant in the range of the investigated electric field intensity, while the dependence on the electric field intensity was significant for magnetic liquids with higher concentrations ($\Phi = 0.019$ and 0.032). The loss factor was independent of the magnetic field and dependent on the electric field weakly at the concentration $\Phi = 0.0019$. In the case of a magnetic liquid with concentrations $\Phi = 0.019$ and 0.032 a dependence of the loss factor on the orientation of magnetic field and its decrease with increasing electric field were observed. The decrease of permittivity indicates that aggregation effects were presented in magnetic liquids with higher volume concentrations of magnetic particles in the high electric fields. Partial discharge current impulses are the smallest in magnetic liquid with magnetite particle volume concentration $\Phi = 0.0024$. The electric breakdown strength distribution was studied for the first time. It is not possible from the performed experiments to determine which of the used fitting functions (Duxbury–Leath, Weibull, Gauss) is more suitable for describing the distribution of electric breakdown strengths.

Acknowledgments

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