Investigation on Charge-Carrier Transport Characteristics of Transformer Oil-based Nanofluids

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ABSTRACT

Adding nanoparticles to traditional transformer oil can improve their heat exchange properties as well as enhance their dielectric withstand characteristics. Investigations into the charge-carrier transport characteristics of these transformer oil-based nanofluids (TNFs) can explain their improved insulating characteristics, and clarify the mechanisms dictating these modifications. In this study, we measured the conduction current and velocity field of TNFs under high electric field excitation for the first time, for analysis of charge-carrier transport. We describe different transport processes according to the magnitude of the applied electric field, characterized as ohmic, tunneling, and space charge limited current (SCLC) stages. In the ohmic stage, characterized by very low electric field strengths, the increase in conduction current is due to the increase in the number of carriers from the addition of nanoparticles. In the tunneling stage (medium-to-high electric field strengths), the predominant charge carriers in the TNF change from ions and colloidal particles, to electrons emitted from the electrode. The thickness of the Schottky barrier at the metal-liquid interface increases on the addition of nanoparticles, which reduces the number of electrons that pass through to the interface region. The field strength required for electron transmission is enhanced, and the insulation strength is improved. In the SCLC stage (very high electric fields), the carrier mobility is reduced because of the larger trap density of TNFs and the electrical discharge is suppressed.

Index Terms — transformer oil-based nanofluids, carrier transport, ohmic stage, tunneling stage, space charge limited current stage

1 INTRODUCTION

WITH the increasing demand for electrical power, oilimmersed-paper equipment, especially transformers and reactors, are required to be capable of secure and reliable operation at high voltages, in addition to possessing large transmission capacities. The concept of nanofluids, proposed by Choi of Argonne National Laboratory in 1995 [1], and the subsequent development of transformer oil-based nanofluids (TNFs) by Segal *et al* in 1998 [2], has helped improve the performance of these devices. TNFs are stable colloidal systems, obtained by dispersing nanoparticles with diameters of less than 100 nm into traditional transformer oil, using methods such as magnetic stirring and ultrasonic oscillation. Research indicates that TNFs exhibit better characteristics than traditional transformer oil in terms of their suitability for use as liquid dielectrics, with respect to parameters such as withstand voltage, thermal conductivity, and anti-aging and moisture resistance properties [3–5]. Electrical characterization, including investigations determining AC/DC breakdown voltage, lightning impulse voltage, and PD voltage [6, 7], also reveals that the insulating characteristics of TNFs are considerably better than those of traditional transformer oils.

Charge carriers are particles that move freely within a liquid dielectric, with the conduction current being generated by this directional motion. The carrier transport characteristics of liquid dielectrics thus affect their insulating properties [8], and can be analyzed through measurement of the conduction current. Carrier mobility, which is the ratio of the carrier migration velocity to the electric field strength, can also be used in analysis of the transport characteristics of charge carriers.

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In 1934, Onsager proposed a conduction model for liquids based on ionic dissociation and recombination [9], from which the prevailing theories on ion mobility have been developed. The research on the conductivity of transformer oil-based nanofluids are mainly focused on the measurement of electrical conductivity and the establishment of the model. Most researches are carried out under low electric field and can't be connected with the breakdown process of transformer oil. For the measurement of the conductivity of liquid dielectric under high electric field, W. F. Schmidt first summed up the elementary processes which is closely linked with the initiation and the propagation of electric breakdown processes in liquids, including: electrode processes, ionization in the liquid phase, bubble formation, and evolution of the breakdown phenomenon [10]. Butcher tested and analyzed the conduction process of the pure transformer oil under the high field [11]. The conductivity process is divided into three stages, which are resistance stage, tunnel effect stage and space charge limiting current saturation stage, respectively. The change of conduction current of transformer oil and the influence of various factors on the transformer oil were discussed in detail by Zhou et al [12]. But the influence of tunneling effect on the conduction process is ignored. Negri and others have studied the conductivity of the magnetic fluid [13], but due to the addition of ferromagnetic nanoparticles, the stability is greatly influenced by the voltage level, and the conductivity characteristics of the space charge limiting current saturation stage are not measured.

At present, explanations for the influence of nanoparticles on the insulating properties of transformer oils, and mechanisms controlling these modifications, are disputed. The behavior of a dielectric in an electric field, from electrical conduction to electrical breakdown processes, is strongly related to carrier transport characteristics [14]. Carrier motion under the influence of a DC electric field can be used to explain the pre-breakdown process. Hence, in this study, we compared the conduction currents and carrier flow fields of TNFs and a traditional transformer oil at different electric field strengths, prior to breakdown. We analyzed the carrier transport characteristics based on this information, to explain the enhancement in insulation resulting from the addition of nanoparticles, and clarify the mechanisms controlling this modification.

2 EXPERIMENT

2.1 PREPARATION OF TRANSFORMER OIL-BASED NANOFLUIDS

In this study, we used ZnO nanoparticles as the colloidal component of the TNFs. The surfaces of the nanoparticles were treated with oleic acid, to reduce agglomeration. In order to observe the morphology and uniformity of ZnO prepared by hydrothermal synthesis method, a representative transmission electron microscopy (TEM) image of the ZnO nanoparticles is shown in Figure 1, revealing a uniform size distribution. The average particle diameter is approximately 20 nm. The X-ray diffraction (XRD) patterns shown in Figure 2 illustrate the purity of the ZnO particles, which were grown along the Caxis via a hydrothermal reaction. In addition, we observe the standard hexagonal wurtzite crystal structure in these particles.



Figure 1. Transmission electron microscopy (TEM) image of the ZnO nanoparticles.



Figure 2. X-ray diffraction (XRD) pattern of the ZnO nanoparticles.

We used Karamay 25 #, a mineral transformer oil, as the base fluid for the TNFs. Prior to nanoparticle dispersion, this was dried in a vacuum oven at 85 °C for 48 h. The water content was 8 ppm, measured using a Karl Fischer micro water tester. A flowchart of the nanoparticle dispersion process is shown in Figure 3. In summary, varying concentrations of ZnO particles were added to a transformer oil base. The solution was mixed for 10 min using a magnetic stirrer, followed by 30 min of ultrasonication, to obtain TNFs with a uniform distribution of nanoparticles.



Figure 3. Flowchart of nanoparticle dispersion process.

We produced TNFs with nanoparticle concentrations of 0.05, 0.1, and 0.2 g/L, suitable for obtaining electrically insulating characteristics. The TNFs were treated in a vacuum oven at 80 °C for 48 h, to reduce the influence of dissolved water and air on the behavior of the dielectrics, and the water content was 6 ppm.

2.2 CONDUCTION CURRENT MEASUREMENT PLATFORM

We measured the conduction current of the samples in a parallel plate configuration, using copper electrodes with a diameter of 25 mm. The two electrodes were separated by a distance of 1 cm. We used a Keithley 6517B electrometer with an operational range from 10 mA to 21 mA for this electrical characterization. In addition, we included an overload protection circuit, composed of two antiparallel diodes, to the system, to prevent damage to the electrometer caused by the breakdown of the test sample. A schematic of the conduction measurement system is shown in Figure 4.



Figure 4. Schematic of conduction measurement system.

Different DC voltages were applied to the sample during the measurement of dielectric conduction current, according to the IEC 60247 and IEC 61620 standards. As space charge migrates to the positive and negative poles under the influence of an electric field, the current flowing through the sample is gradually reduced to a limit value following DC voltage excitation. The time-dependent changes in the conduction current of the traditional transformer oil and a TNF (0.1 g/L), measured at room temperature (25 °C) at different electric field strengths, are shown in Figure 5. As we observed a stable current after 30 s of excitation, in subsequent experiments, we defined the conduction current as the average of the values measured between 60 and 100 s.

DC voltage was applied using a step-up method, as shown in Figure 6. The voltage was increased from a minimum (500 V) to a maximum (9–13 kV), in increments of 500 V. The duration of excitation at each voltage increment, t, selected as two minutes, should be greater than the time required to establish a stable leakage current. Δt is the boost time, i.e., the length of the transition between successive voltage increments, which was typically 3 s long

3 RESULTS AND DISCUSSION

As previously stated, the directional motion of chargecarriers under the influence of an electric field generates a conduction current. The conduction processes of TNFs can thus be classified according to the electric field strength. In a weak electric field, TNF conduction is primarily dependent on ionic mobility and the electrophoretic mobility of nanoparticles. In contrast, in a strong electric field, carrier motion is connected to processes occurring at the interface between the electrode and the dielectric.



Figure 5. Time-dependent change in conduction current of (a) a traditional transformer oil, and (b) a transformer oil-based nanofluid (0.1 g/L), measured at different electric field strengths.



Figure 6. Illustration of changes in excitation used in step-up voltage method.

Conductivity γ is defined as the ratio of the current density of a material *j* to the electric field strength *E*, as below [15]:

$$\gamma = \frac{J}{E} \tag{1}$$

The relationship between the conductivity of a liquid dielectric, as measured by the conduction current, and the

electric field strength is shown in Figure 7. Here, we mainly analyze the difference between traditional transformer oil and TNFs with nanoparticle concentrations of 0.1 g/L. We note a gradual increase in conduction current as the electric field strength increases.



Figure 7. Characteristic E-I curves of transformer oil based dielectrics.

From Figure 7, we note a negligible change in conduction current, for weak electric fields. In contrast, in a strong electric field, the rate of the increase in current gradually accelerates, and the current becomes significantly high. This discrepancy is a result of the different processes governing carrier motion.

The current density of a dielectric material is determined by the density of carriers in the medium, n_0 , the migration velocity of the carriers, v, and the charge of the carriers, q, as below:

$$j = q \cdot n_0 \cdot v \tag{2}$$

The addition of nanoparticles to a transformer oil primarily increases the carrier density, resulting in the enhancement of the conduction current of TNFs in a weak electric field. Nevertheless, we note that, in a strong electric field, the increase in conduction current is slower with the TNFs than it is with traditional transformer oil, as the reduced mobility of carriers in TNFs suppresses the development of discharge, leading to an improvement in the insulating strength.

To further characterize the effect of nanoparticles on carrier transport, we analyzed the experimentally derived data using the Fowler–Nordheim plot method [16–18]. Based on our observations, the entire process of conduction can be divided into three stages: the ohmic stage, tunneling stage, and space charge limited current (SCLC) stage, as shown in Figure 8 [19]. Then, we analyzed the specific laws of different stages and tried to explain the phenomena from the motion of microscopic charged particles.

3.1 OHMIC STAGE

Figure 9 shows the relationship between the conduction current of the traditional transformer oil and the TNFs, and electric field strength, for weak excitation. In accordance with Ohm's law, the current is proportional to the field strength.



Figure 8. Relationships between I and E in a Fowler–Nordheim plot for (a) the transformer oil, and (b) TNFs, illustrating the different conduction stages.



Figure 9. E-I curves of transformer oil-based dielectrics in the ohmic stage.

The electrical conductivity of the TNFs is much larger than that of the traditional transformer oil, and increases with increasing nanoparticle concentration. This relationship can be inferred from the conduction current, as conductivity is the ratio between the current density and the electric field strength, and it is equivalent to the product of carrier density, carrier charge, and carrier mobility μ [20, 21]:

$$\gamma = q \cdot n_0 \cdot \mu \tag{3}$$

For weak excitations, the average migration velocity of the carrier is related to the electric field intensity through carrier mobility, which typically remains constant, as follows:

$$v = \mu \cdot E \tag{4}$$

Under the low electric field, ions and electrophoresis play a leading role in conduction current of traditional transformer oil. After adding nanoparticles, electrical charge can be adsorbed onto the surface of nanoparticles in TNFs. Hence, as well as molecules of alkanes, or ions dissociated from impurities, charge carriers in TNFs include the colloidal nanoparticles. Thus, carriers of transformer oil-based nanofluids are much larger, and increasing the nanoparticle concentration increases the carrier density and conductivity of a TNF. The diversity of charge carriers means that the mobility of ions and the electrophoretic mobility of nanoparticles in TNFs are both involved in the process of conduction in this stage.

3.2 TUNNELING STAGE

Under low electric field, the main conduction is ionic conduction and electrophoretic conduction. Therefore, at the ohmic stage, the increase of nanoparticles is actually increasing the electrophoretic conduction and increasing the number of carriers. However, it is no longer only ionic conduction under high electric field, but the simultaneous effect of ions, electrophoretic and electrons. The difference between pure oil and transformer oil-based nanofluids is mainly caused by the effect of nanoparticles on the electrons.

In view of the general law of conduction of pure oil, electric current increases exponentially with electric field under high electric field. That is to say, there are lots of charged particles in the liquid. Assuming that the ionic conduction still plays a major role under high electric field without the effect of electrons, it can be thought that the increase of electric current is caused by the increase of ionic mobility or ions dissociation. Now these two possibilities are discussed.



Figure 10. Schematic diagram of ion migration.

When the positive ions A, B, C vibrate at the lowest potential energy, the vibration frequency is v. If the thermal vibration energy exceeds the binding barrier of the molecule, then the ion migrates. When E=0, ion concentration n_0 has six directions, and the ion concentration of each transition may be n_0 /6. In one direction, the number of ions *n* that can

overcome the potential barrier transition to the new equilibrium position every second can be calculated.

$$n = \frac{n_0}{6} v e^{\frac{u}{kT}}$$
(5)

Take the x axis as an example. When $E_m \neq 0$, A migrates to B, and barrier potential is reduced by $\Delta u = \frac{1}{2}qE\delta$.

$$n_{AB} = \frac{n_0}{6} v e^{-\frac{\bar{u} - \Delta u}{kT}}$$
(6)

B migrated to A, and barrier potential is increased by Δu .

$$n_{BA} = \frac{n_0}{6} v e^{-\frac{\overline{u} + \Delta u}{kT}}$$
(7)

The number of excess migration ions Δn in the X direction can be obtained.

$$\Delta n = n_{AB} - n_{BA} = \frac{n_0}{6} v e^{-\frac{u}{kT}} (e^{\frac{\Delta u}{kT}} - e^{-\frac{\Delta u}{kT}})$$
(8)

When the electric field is not too high and satisfy $\Delta u = kT$, formula 8 can be simplified.

$$\Delta n = \frac{n_0}{6} v e^{-\frac{\bar{u}}{kT}} \left[I + \frac{\Delta u}{RT} - (I - \frac{\Delta u}{RT}) \right]$$

$$= \frac{n_0}{6} v e^{-\frac{\bar{u}}{kT}} \left[2 \frac{\Delta u}{RT} \right]$$

$$= \frac{n_0}{6} v e^{-\frac{\bar{u}}{kT}} \left[2 \frac{\frac{I}{2} qE\delta}{kT} \right]$$

$$= \frac{n_0 q \delta v}{6kT} e^{-\frac{\bar{u}}{kT}} E$$
(9)

The average velocity of the ions in the X direction can be calculated.

$$v = \frac{\Delta n\delta}{n_0} = \frac{n_0 q \delta^2}{n_0 6kT} v e^{-\frac{u}{kT}} E = \frac{q \delta^2}{6kT} v e^{-\frac{u}{kT}} E = \mu E$$
(10)

$$\mu = \frac{q\delta^2}{6kT} v e^{-\frac{\bar{u}}{kT}}$$
(11)

Formula (8) indicates that mobility of ions is independent of electric field when the electric field satisfy $\Delta u = kT$.

When the electric field is too high and satisfy $\Delta u = kT$, Equation 9 can also be simplified.

$$\Delta n = \frac{n_0 v}{6} e^{-\frac{u}{kT}} \cdot e^{\frac{q\delta E}{2kT}}$$
(12)

$$v = \frac{\Delta n\delta}{n_o} = \frac{\delta v}{6} e^{-\frac{\tilde{u}}{kT}} e^{\frac{q\delta E}{2kT}}$$
(13)

$$\mu = \frac{v}{E} = \frac{\delta v}{6E} e^{-\frac{u}{kT}} e^{\frac{g\delta E}{2kT}}$$
(14)

Here, it seems that the mobility increases exponentially with the increase of E. The strength of the critical electric field can be calculated by the condition $\Delta \mu = \frac{q \delta E_m}{2} = kT$, then $E_m \approx 5 \times 10^8 kV$. But the transformer oil is basically broken

down under this condition. Then it can be considered that the mobility does not change with the electric field.

For ionic dissociation, concentration of ions can be calculated by Equation 15. It does not change exponentially with the electric field.

$$n_0 = \sqrt{\frac{N_0 v_0}{\zeta}} e^{-\frac{u_a}{2kT}} e^{\sqrt{\frac{e^3}{\pi \varepsilon_0 \varepsilon_r} \frac{\sqrt{E}}{2kT}}} = A e^{a\sqrt{E}}$$
(15)

Therefore, the conduction under high electric field is different from that in low electric field, and it is no longer just effected by ions. Based on previous studies, we believe that electrons emitted from the electrodes will have an effect on conduction at high electric field [20]. In this stage, the current and electric field strength satisfy the following equation:

$$I = K \cdot E^2 \exp(-B/E) \tag{16}$$

where K and B are constants of proportionality. The relationship between $ln(I/E^2)$ and E^{-1} can be plotted as a straight line. For TNFs, the gradient is -16, which is steeper than the gradient calculated with the traditional transformer oil (-2.455). This gradient can be determined analytically, as follows:

$$B = -\frac{3\chi\varepsilon}{2\Delta d} \tag{17}$$

where χ is the electron affinity barrier, Δd is the barrier thickness, and ε is the relative permittivity of the dielectric.

The electron affinity barrier reflects the energy required to free electrons at the bottom of the conduction band of an electrode. As the electrodes are surrounded by transformer oil, and the electrode material does not change, the electron affinity barrier remains constant. An approximately 100-nm-thick interface is formed when a metal electrode comes in contact with a dielectric. Interface barriers, A and C, also appear, as shown in Figure 11 [22]. The interaction of charges in the interface creates a double layer, in which the electric field can be as high as 10^3 MV/m [23].



Figure 11. Schematic of the interface between the electrode and transformer oil.

For transmission to the dielectric, the energy of the electrons should be greater than the height of barrier A. Alternatively, electrons can tunnel through this region when their de Broglie wavelength is greater than the thickness of the barrier. These electrons increase the conduction current of the transformer oil. Hence, in moderate electric fields, electronic conductivity from field emission is the primary factor that dictates the conduction current of transformer oils. The primary effect of the addition of nanoparticles is to increase the thickness of the potential barrier. As a result, the number of electrons transmitted to the interface region is reduced, and breakdown is suppressed.

Analysis of the transition from the ohmic stage to the tunneling stage revealed that the electric field strength required to induce tunneling increased from 0.6 kV/mm to 1.5 kV/mm, following the addition of the nanoparticles. This discrepancy can be explained using the interaction between ZnO particles and electrons. As the nanoparticles in the TNF are capable of electron capture, the electric field in the region of the electrodes is weakened and the velocity of the electrons is subsequently reduced. A higher electric field is thus needed to provide the electrons with sufficient kinetic energy to leave the electrode and enter the interface, delaying the onset of the tunneling stage.

As in the ohmic stage, under the influence of an electric field, nanoparticles in the TNF act as carriers, as they are charged through electron capture. The kinetic energy of these particles is transferred to the oil molecules in the process of collision. This transfer of momentum moves the liquid molecules in the direction of the carriers. To analyze carrier motion more intuitively, we observed the velocity field of TNFs under DC excitation, using particle image velocimetry. Details regarding the parameters used in this experiment are available from our previous work [24].



Figure 12. Speed distribution of TNFs at different excitations.

Based on the results of velocimetry, the average speeds of TNF flow in the electrode region were approximately 2.156, 4.317, and 7.962 mm/s, for DC excitations of 10, 15, and 20 kV, respectively. The relationship between the voltage and velocity is plotted in Figure 13.

From Figure 13, we note that the velocity of TNF flow increases with increasing electric field strength, as suggested by our considerations of transfers in momentum. This increase in velocity is accelerated when the conduction phase changes from the ohmic stage to the tunnel stage, indicating an increase in carrier mobility with electric field strength. The carrier concentration also increases, as the number of electrons injected into the dielectric is increased.



Figure 13. Analysis of flow rate of TNFs with respect to electric field strength.

3.3 SPACE CHARGE LIMITED CURRENT STAGE

In addition to increases in the free carrier migration velocity and charge injection from the electrode, application of a strong electric field causes carrier multiplication in the dielectric and space charge accumulation. As the applied voltage is increased, if the electrode injection current is not equal to the body drift current I_b, excess charges are generated in the dielectric, which are trapped by carriers to form the space charge. When the electric field generated by the space charge is balanced with the external electric field, the injection current reaches a steady state. In this region, the current in the dielectric is termed SCLC. For traditional transformer oil, if electron capture by space charge traps is ignored, the current and electric field satisfy the Mott–Gurney Law [25]:

$$I = \frac{9}{8} \cdot \left(\frac{\mu \cdot \varepsilon \cdot \varepsilon_0 \cdot S}{d}\right) \cdot E^2$$
(18)

where ε is the dielectric constant of the transformer oil (2.2), *d* is the size of the oil gap (1.5 mm), and μ is the apparent carrier mobility.

To verify the transition to the SCLC stage, we plotted the current with respect to the square of the electric field, as in Figure 14.

We note that the current is directly proportional to the square of the electric field strength, as is typical of SCLC. The carrier mobility in this region was calculated to be 2.93×10^{-5} cm²/(V·s). The inclusion of nanoparticles increases trapping density, such that the effects of electron capture in TNFs cannot be ignored. Instead, a coefficient θ_a is introduced as a correction factor related to trap density, given as

$$\theta_a = \frac{n}{n+n_r} \tag{19}$$



Figure 14. Plot of E²-I for the traditional transformer oil under large electric field excitation.

where n_t is the density of traps introduced by the nanoparticles, and *n* is the intrinsic trap density of the base fluid. When the space charge trap density is less than 1, the mobility of carriers in TNFs can be calculated as

$$\mu_{\rm n} = \theta_a \cdot \mu_0 \tag{20}$$

where μ_0 is the carrier mobility of the base fluid.

With TNFs, we observe a proportional relationship between current and the square of the electric field, as shown in Figure 15. The carrier mobility in this region was calculated to be approximately 2.2×10^{-5} cm²/(V·s). The trap density is 1.5 times that of traditional transformer oil, which is consistent with the results presented by Yuefan et al. [26].



Figure 15. Plot of E²⁻I curve for TNFs under high electric field excitation.

The addition of nanoparticles increases the density of traps in TNFs. In a strong electric field, electrons produced by ionization are constantly being trapped and released in the course of motion, impeding their migration. Thus, nanoparticles inhibit carrier mobility, explaining the differing behaviors of TNFs and transformer oil in the SCLC stage.

4 CONCLUSIONS

In this study, the transport characteristics of carriers in TNFs, under the influence of electric fields of different

magnitudes, were obtained, by analyzing the conduction current and the velocity field of the fluids. From these analyses, we explained the effect of nanoparticles on the electrical properties of transformer oil, and clarified the mechanisms dictating these modifications. We identified three distinct conduction mechanisms in TNFs, depending on the magnitude of the electric field. In considering these mechanisms, we divided the evolution of conduction with electric field into three stages: ohmic, tunneling, and SCLC.

In the ohmic stage, the current (I) is primarily dependent on free carrier motion, and it is proportional to the electric field strength (E). The addition of nanoparticles increases the number of electrophoresis, resulting in an increase in the conduction current.

In the tunneling stage, the relationship between electric field strength and current is also more complex: $\ln(I/E^2)$ is proportional to E^{-1} . With the increase in electric field strength, the effect of electrons emitted by the electrodes can't be ignored. And the carrier mobility gradually increases, in contrast to the ohmic stage. The addition of nanoparticles increases the thickness of the barrier at the electrode-dielectric interface, reducing the number of electrons transmitted to this region. This increase in barrier thickness increases the field strength of the transition from the ohmic stage to the tunneling stage.

In the SCLC stage, the current (I) is proportional to the square of the electric field strength (E^2). The addition of nanoparticles increases the density of traps in the transformer oil, which impede carrier motion. These traps thus reduce carrier mobility, suppressing the occurrence of discharge.

From the above three stages, we can find that although the nanoparticles increase the conductivity at the low field strength stage, the contribution of nanoparticles to the inhibition of breakdown is more prominent at the higher electric field. In order to improve the mechanism of nanoparticle modification, more comprehensive tests from different angles of materials are certainly necessary in future works, including the inhibition effect of nanoparticles on the motion charge in transformer oil, the boundary effect of antimoisture, anti-aging and strengthening effect in the field of heat conduction.

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