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ABSTRACT

Nanoparticles enhance the positive and negative lightning breakdown voltages of vegetable insulating oils due to a reduced streamer velocity. Pre-breakdown streamers in vegetable oil based h-BN and Fe$_3$O$_4$ nanofluids were observed using the shadowgraph method under a lightning impulse voltage. The propagation of streamers until their stopping length is analyzed using shadowgraph images for the different nanofluids. The results suggest that these nanofluids present a higher breakdown voltage per unit length compared with pure oil. Secondary reverse streamers are observed to appear at the dissipation stage of the streamer. In particular, the secondary reverse streamer of the nanofluids appeared earlier and seemed to be shorter than that of pure oil under positive impulse voltages. The underlying mechanism was analyzed using space charge theory.

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I. INTRODUCTION

Insulating oil, which also serves as a cooling medium, is an important part of power transformer insulation system. Mineral insulating oil, the traditional oil used for transformer insulation, has a low fire point of approximately 170 °C and possesses poor bio-degradability, allowing contaminating soil and water leakage to occur. Therefore, vegetable insulating oil, which is an environmentally-friendly liquid dielectric, has drawn significant attention as a potential alternative to traditional mineral oils for power transformers.

Nanofluids are formed from adding nanoscale particles to a base fluid and offer notable advantages to insulation enhancements. Research on the breakdown process of insulating oils has mainly focused on the streamer development at the pre-breakdown phase. The liquid dielectric produces a weak discharge channel under the action of a local high field strength and a "treelike" streamer is generated over a relatively short time under an external energy injection. Recently, streamer initiation and propagation at the pre-breakdown stage of discharge processes in mineral oil based nanofluids have been reported by Hwang et al. The results have shown that nanoparticles hinder the streamer propagation by quenching electrons and reducing the negatively charged ion mobility. A secondary back discharge was discovered from the current waveform after the main discharge and the phenomenon of a secondary reverse streamer was revealed. However, the secondary reverse streamer phenomenon has not yet been reported yet in vegetable oils.

In this paper, positive and negative breakdown voltage of vegetable oil based h-BN and Fe$_3$O$_4$ nanofluids under lightning impulse voltage is reported. Shadowgraph images are utilized to analyze the streamer propagation and dissipation stages of the pre-breakdown with a 25mm electrode gap. In addition, the generation mechanism for the secondary reverse streamer at different dissipation stages is explained using space charge theory.
II. EXPERIMENTAL METHODS AND PROCEDURES

A. Preparation of nanoparticles and nanofluids

The nanofluids were generated using a two-step method. The h-BN nanoparticles were prepared with a liquid exfoliation method. Micrometer-sized h-BN (Aladdin, 1–2 μm, 98%) were sonicated for 24 hours in isopropyl alcohol at room temperature and centrifuged at 3000 rpm for 10 min to collect the supernatant, which was vacuum-dried in an oven for 48 hours. Particles in the form of a white powder were then collected and ground. The ferroferric oxide (Fe$_3$O$_4$) nanoparticles were prepared with a high temperature decomposition method. A total of 6.48 g of iron (III) chloride hexahydrate and 21.9 g of sodium-oleate-vigorous were mixed with 48 ml of ethanol and 84 ml of n-hexane in a beaker and stirred for 6 hours in a 60 °C water bath. After washing three times with deionized water, 2.1 g of the iron oleate precursor, 10 ml octadecene mixed with 0.64 ml oleic acid and added under stirring at 800 r/min before heating to at 200 °C for 2 hours and followed by continuous heating at 320 °C for 48 hours. After cooling to room temperature, the mixture was subsequently centrifuged at 3000 rpm with ethanol and cyclohexane and washed several times.

Two types of nanoparticles were dispersed in the commonly used vegetable insulating oil FR3, mechanized by Cargill, using an ultrasonic dispersion technique. Before the electrical characterization, three nanofluids and pure FR3 were dried at 85 °C under 50 Pa for 72 hours.

B. High-voltage experimental platform and test electrodes

The lightning impulse breakdown voltage testing platform is shown in Fig. 1. The needle to plate electrode configuration was used and a high voltage stress was applied at the needle electrode while the plate electrode was grounded. The radius of curvature of the tungsten needle was 50 μm ± 5 μm, and the diameter of the plate electrode was 20 cm. The gap distance between the electrodes was adjustable with a maximum of 50 mm. The entire system consisted of three parts: test cell, impulse generator and measuring components, and the discharge recording equipment.

The lightning impulse breakdown, which is a key parameter to represent the insulating properties of dielectric liquids, was measured in accordance with the IEC 60897 standard. The nano-oil was subjected to a lightning stroke test using the rising voltage method. The lightning impulse was provided from a six-stage impulse generator, with maximum voltage was of 900 kV to provide for an energy of 33.74 kJ. This impulse generator was used to deliver a standard lightning impulse of 1.2 (±30%)/50 μs (±20%) and a switching impulse of 250 (±20%)/2500 μs (±60%). The voltage waveform was measured using a high-voltage capacitive resistor voltage divider and was recorded with an oscilloscope (LeCroy WaveRunner 610zi). The sampling rate was adjusted to 10 GS/s, with a bandwidth of 1 GHz.

III. RESULTS AND DISCUSSION

A. Characterization of nanofluids

Transmission Electron Microscopy (TEM) images and the Atomic Force Microscope (AFM) of the exfoliated h-BN samples are presented in Fig. 2. It can be seen that the sizes of the exfoliated h-BN nanoparticles are mostly irregular, and the lengths ranges from a few hundred nanometers to one micrometer. The Fe$_3$O$_4$ nanoparticles are evenly distributed with an average size of about approximately 20 nm.

B. Lightning impulse breakdown voltage

The results of the positive and negative lightning breakdown voltages for the oil samples under different gap lengths are shown in Fig. 3. It can be seen that the two kinds of nanoparticles enhanced the breakdown voltage of the vegetable oil. Under a 15 mm gap, the positive lightning breakdown voltage of the Fe$_3$O$_4$ and h-BN nanofluids is increased by 28% and 26%, respectively, with similar enhancements of approximately 18% and 16% observed under...
a 50 mm gap. In contrast, the negative lightning breakdown voltage of the Fe3O4 and h-BN nanofluids at a 15 mm gap increased by 6.8% and 9.9%, respectively, and similar improvements of 3.2% and 7.3% were observed for a 50 mm gap.

### C. Streamer propagation and dissipation

The overall process of streamer propagation and dissipation in insulating oils can be considered when the electric field near the tip of the needle increases and the liquid molecules and bubbles near the needle tip begin to ionize, as show in Fig. 4. The ionization region expands with an increasing applied voltage, and finally reaches the area near the tip of the needle to form streamers while a corona erupts. Thus, a small tip streamline is formed at the needle tip. If the voltage is further increased, the streamer continues to develop at the core of the needle tip region.

The maximum electric field $E$ at the needle tip can be expressed by using Mason’s equation as

$$E_{\text{max}} = \frac{2 \times V}{r \times \ln(1 + (4d/r))}$$

where $V$ is the voltage of the needle tip, $r$ is the tip radius of the needle electrode, and $d$ is the gap distance.

Fig. 5 provides shadow photographs of the pre-breakdown positive streamer propagation and dissipation of the an oil samples under applied voltage of 60 kV at a 25 mm gap distance. As the applied voltage increases, the positive streamer begins propagating towards the ground electrode. Its morphology is transformed from a small dispersion and a filament distribution to more branched with a longer filament near the tip of the needle electrode. The initial direction of the streamer is random and partially branched at the Initial state of the streamer as shown in Fig. 5a. As the branches of the streamer are induced by the electronic avalanche, the mutual exclusion and shielding effect of the same polar electron avalanche leads to the development of the streamer branching, which becomes more obvious during its propagation process with an increased discharge time. In addition, the streamer propagation process is limited by the applied voltage and the dielectric liquid, causing the streamer to reach a stopping length without further development. The stopping length of streamer in vegetable oil was 21.9 mm as demonstrated in Fig. 5(b). If the applied voltage is not sufficiently strong to cause a breakdown, the streamer will dissipate and taper with a significant increase of the stopping length in the vegetable oil (Fig. 5(c)). After 230 μs, a short streamer may appear at the needle tip, called a secondary reverse streamer, which is generated due to the reverse electric field from the residual space charges.

Compared with pure oil, the shape, length and time of the secondary reverse streamer for the two types of nanofluids are different. The branches of the nanofluids near the needle tip have obvious developments. The stopping length of the nanofluids are shorter than for pure oil in Fig. 5(b) (h-BN nanofluids ∼15.8mm and Fe3O4 nanofluids ∼13.9mm). Moreover, the secondary reverse streamer appears at 170 and 190 μs for the h-BN and Fe3O4 nanofluids respectively, which is significantly shorter than for the pure vegetable oil.
Figure 5 shows shadowgraphs of the pre-breakdown propagation and dissipation of the negative streamers in the tested oil samples under an applied voltage of 80 kV. Multiple branched streamers develop in the nanofluids that exhibit a shrub-like structure. The pure oil has a main branch and several lateral branches. At 10 μs, the streamer in the h-BN and Fe$_3$O$_4$ nanofluids have stopping lengths of 12.7 and 13.0 mm respectively, which are 89% and 91% that of the pure oil (14.2 mm). For all considered oils, the streamers began to dissipate and the secondary reverse streamers appeared at approximately 150 μs.

In Fig. 7, curve fitting describes the relationship between the (a) positive and (b) negative applied voltages and the stopping lengths for oil gaps less than 25 mm for tested oil samples. Initially, all oil samples stopped at approximately 5 mm, while voltages closer to the breakdown voltage produced a maximum stopping length of 20 mm for the streamers. For the positive streamers, the growth rate in pure oil was 0.85 mm/kV, and the growth rates for BN and Fe$_3$O$_4$ nanofluids were lower at approximately to 0.63 and 0.57 mm/kV respectively. Assuming the effect of side branches are negligible, each unit streamer length required for the voltage was calculated using Fig. 7(a), which were 1.17, 1.58, and 1.75 kV/mm for the pure oil, BN nanofluid, and Fe$_3$O$_4$ nanofluid, respectively. Therefore, nanofluids need a higher breakdown voltage per unit length of the streamer compared with pure oil, indicating that nanoparticles can inhibit the development of positive streamers and increase the breakdown voltage. For negative streamers, the growth rates of the pure oil, BN nanofluid, and Fe$_3$O$_4$ nanofluids were 0.64, 0.61, and 0.6 mm/kV, respectively, and the voltages required for each unit length were 1.56, 1.64, and 1.67 kV/mm, respectively.
D. Secondary reverse streamer

Fig. 8 shows the secondary reverse streamer images for the pure oil and the nanofluids after clearing the background and any previous streamers. For a positive voltage, the length of the streamer in the nanofluids is shorter than that of the pure oil. This indicates that the nanoparticles inhibit the development of the secondary reverse streamer. For a negative voltage, the secondary reverse streamer lengths of the nanofluids are slightly smaller than that of the pure oil. Compared to the positive secondary reverse streamer, the negative secondary reverse streamer of the nanofluids is more pronounced. At the same time, the statistics of the secondary reverse streamer development time for all the oil samples were analyzed and are presented in Fig. 9. The results show that the positive secondary reverse streamer of the h-BN nanofluid was the fastest with an average time of approximately 172 μs, whereas, those for the Fe₃O₄ nanofluids and pure oil were 183 and 235 μs, respectively. This indicates that the nanoparticles cause the secondary reverse streamer to appear sooner while tending to shorten the streamer dissipation time. The average time of the negative secondary reverse streamer for the pure oil, h-BN nanofluids and Fe₃O₄ nanofluids are 152, 148 and 155 μs, respectively. The results suggest that the streamer times of the nanofluids were less influenced by the negative secondary reverse streamer.

The secondary streamer maybe generated from the reverse electric field, which is caused by the residual space charge resulting from the primary streamer.
The mechanism for the formation of the negative secondary reverse streamer is shown in Fig. 11. As the primary streamer dissipates, there is a significant amount of positive charges and negative ions near the needle electrode, resulting in the generation of an electric field $E_q$ and the secondary reverse streamer. However, the nanoparticles with negative charges in the nanofluids neutralize the positive charge near the needle electrode and strengthen the negative charge in the middle of oil gap. Thus, the nanoparticles have little effect on the negative secondary reverse streamer of the vegetable oil.

IV. CONCLUSIONS

Enhancements of the positive and negative lightning breakdown voltage in vegetable insulation oil proposed using h-BN and Fe$_3$O$_4$ nanoparticles under large electrode gaps ($15$–$50$ mm). With a $15$ mm gap, the positive lightning breakdown voltage of the Fe$_3$O$_4$ and h-BN nanofluids increased by 28% and 26%, respectively. Similarly, the negative lightning breakdown voltage increased by 6.8% and 9.9%, respectively. The streamer propagation and dissipation was observed using shadowgraphs under a lightning impulse voltage to find the stopping lengths of the bush-like streamer structures. For the streamer propagation, the nanofluids developed into several main discharge channels near the needle tip, showing a shrubby-type shape, and the stopping lengths of the nanofluids were found to be shorter. For the streamer dissipation, a shorter streamer, called the secondary reverse streamer, appeared at the needle tip. Since the nanoparticles produce charge carrier trapping states that capture migrating electrons to later form negative ions, the secondary reverse streamer of nanofluids appeared earlier and was shorter than that of the pure oil under a positive impulse voltage. In general, nanoparticles can improve the insulation performance of vegetable insulating oils and inhibit the propagation of streamers. Therefore, these nanofluids are favorable for applications with vegetable insulating oil in transformers.

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