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Effects of Nanoparticle Materials on Prebreakdown and Breakdown Properties of Transformer Oil

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Featured Application: This work is of practical value in predicting the breakdown accidents and of theoretical importance in reducing the volume of power equipment.

Abstract: In order to reveal the effects of nanoparticle materials on prebreakdown and breakdown properties of transformer oil, three types of nanoparticle materials, including conductive Fe_3O_4, semiconductive TiO_2 and insulating Al_2O_3 nanoparticles, were prepared with the same size and surface modification. An experimental study on the breakdown strength and prebreakdown streamer propagation characteristics were investigated for transformer oil and three types of nanofluids under positive lightning impulse voltage. The results indicate that the type of nanoparticle materials has a notable impact on breakdown strength and streamer propagation characteristics of transformer oil. Breakdown voltages of nanofluids are markedly increased by 41.3% and 29.8% respectively by the presence of Fe_3O_4 and TiO_2 nanoparticles. Whereas a slight increase of only 7.4% is observed for Al_2O_3 nanofluid. Moreover, main discharge channels with thicker and denser branches are formed and the streamer propagation velocities are greatly lowered both in Fe_3O_4 and TiO_2 nanofluids, while no obvious change appears in the propagation process of streamers in Al_2O_3 nanofluid in comparison with that in pure oil. The test results of trap characteristics reveal that the densities of shallow traps both in Fe_3O_4 and TiO_2 nanofluids are much higher than that in Al_2O_3 nanofluid and pure oil, greatly reducing the distortion of the electric field. Thus, the propagations of positive streamers in the nanofluids are significantly suppressed by Fe_3O_4 and TiO_2 nanoparticles, leading to the improvements of breakdown strength.

Keywords: nanoparticle; impulse breakdown strength; streamer propagation; trap characteristic; electric field distribution

1. Introduction

Breakdown characteristics of transformer oil have been widely investigated for its important significance in the safe operation of power equipment [1,2]. Breakdown phenomena in transformer oil are caused by the initiation and propagation of charged gaseous channels called "streamer" under the high local electric field [3]. Positive streamers have been proved to initiate at lower applied voltages and propagate faster and further than negative streamers [4–6], which constitute a greater risk to oil insulated high voltage electrical equipment than negative streamers. It has been found that the positive impulse breakdown strength of transformer oil depends on the prebreakdown phenomenon [7–9] and it can be considerably influenced by adjusting the propagation characteristic of prebreakdown streamers with the addition of additives [10,11].

Many efforts have been devoted to improve the positive breakdown voltage of transformer oil. In the past two decades, nanofluids (NFs) have attracted much attention due to their good insulating and thermal performance [12,13]. A variety of nanoparticle (NP) materials have been used to prepare transformer oil-based nanofluids, exhibiting different modification effects. It is found that the positive lightning impulse breakdown voltage of mineral transformer oil can be improved by 82.6% with the addition of Fe_3O_4 nanoparticles [13]. For TiO_2 nanofluid, the breakdown voltage is increased by 23.6% [14]. However, with the addition of Al_2O_3 and SiO_2 nanoparticles, the breakdown voltage is decreased by 11.6% and 8.8%, respectively [15]. Recently, three types of nanoparticle materials with the same particle size were used to modify the switching impulse breakdown voltage of transformer oil. Based on the simulation of the potential well distribution on the nanoparticles, a possible mechanism was proposed to explain modification effects of the three kinds of nanoparticles [16]. However, there has been still dearth of experimental evidence that the type of nanoparticle materials has an effect on prebreakdown process in transformer oil, especially for nanoparticles with the same size, surface modification and concentration. The propagation process of prebreakdown streamer will give a valuable insight into the event leading to breakdown and provide a bridge between macroscopic breakdown phenomena and microscopic effect mechanism. Therefore, it is helpful to study the prebreakdown process in nanofluids for an improved understanding of the effect mechanism of nanoparticle materials.

In this paper, three types of nanoparticle materials, including conductive Fe_3O_4, semiconductive TiO_2 and insulating Al_2O_3 nanoparticles, were prepared with the same size and surface modification. The breakdown strengths of transformer oil and nanofluids with the same nanoparticle concentration were measured. The propagation characteristics of positive streamers in the oil samples are studied by comparing their shapes, propagation lengths and velocities with the help of the shadowgraph technique. Moreover, the effects of nanoparticle materials on the breakdown and prebreakdown characteristics of nanofluids are discussed.

2. Materials and Methods

2.1. Nanofluid Preparation

In this work, naphthenic transformer oil (25# Karamay) was used as the base oil, which was filtered in order to remove the impurity particles and meet the demand of pure oil defined by CIGRE working group 12.17 [17]. Fe_3O_4, TiO_2 and Al_2O_3 nanoparticles modified by oleic acid were synthesized in our lab, which were tested to be the same diameter of 10 nm, as shown in Figure 1. Nanofluids were prepared by dispersing the three types of nanoparticle materials into the pure oil with a same volume percentage concentration of 0.025% by stirring and ultrasonic treatment. This concentration is the optimal choice considering both the modification effect and dispersion stability, because the nanoparticle sedimentation happens easily under higher concentrations. The nanofluids and the pure oil were degassed at less than 1 kPa for 24 h before testing and the moisture content of each sample was around 10 ppm.

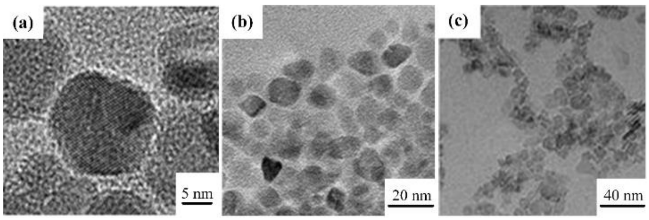

Figure 1. HRTEM (High Resolution Transmission Electron Microscopy) images for three types of nanoparticles: (**a**) Fe_3O_4; (**b**) TiO_2; (**c**) Al_2O_3.

2.2. Experimental Setup and Measurements

Figure 2 shows the schematic of experimental setup used to observe the propagation characteristics of prebreakdown streamers. A needle-sphere electrode system, with a high voltage tungsten needle (tip radius of 35 ± 5 μm) opposing a grounded sphere, was located within a test cell made of transparent Perspex sheet, which facilitates the streamer observation. A ten stage impulse generator with an energy of 27.5 kJ was used to provide 1.2/50 μs standard lightning impulse voltage. The positive impulse breakdown strength were measured with an electrode gap of 25 mm according to the standard procedures for testing lightning impulse breakdown strength (IEC60897-1987). Then, an Intensified Charge Coupled Device (ICCD) camera was used to capture streamer propagation images, which were performed with an electrode gap of 45 mm under peak voltage of 75 kV. The camera works with the help of the laser to meet the need of the shadowgraph technique. A trigger unit was used to synchronously trigger the impulse generator and camera.

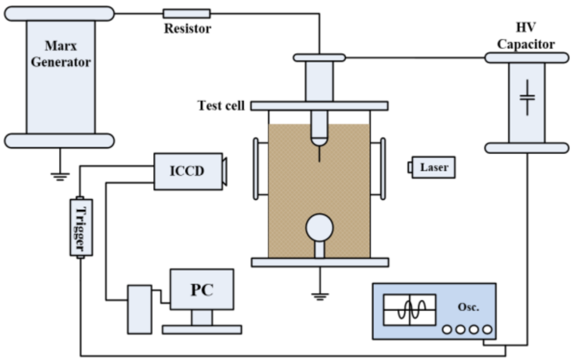

Figure 2. Schematic of experimental setup for measurement of breakdown strength and prebreakdown streamers.

To further investigate the microscopic effect mechanism of different types of nanoparticle materials, the charge transport processes of pure oil and nanofluids were measured by the thermally stimulated current (TSC) method. The experimental setup used was shown in Figure 3. At atmosphere pressure, switch S_3 was closed and a negative direct current (DC) field of 2 kV/mm was applied to the oil sample with a thickness of 0.2 mm for 20 min at 313 K. Then, after the temperature was lowered down to 248 K, switch S_3 was opened and switch S_1 was closed to discharge the equivalent capacitor of electrodes for one minute. Finally, switch S_1 was opened and switch S_2 was closed to begin the measurement of TSC by raising temperature at a rate of 2 K/min. A Keithley 6514 electrometer provides an accuracy of 10^{-16} A on the current measurement, and the accuracy of temperature measurement is about ±0.2%.

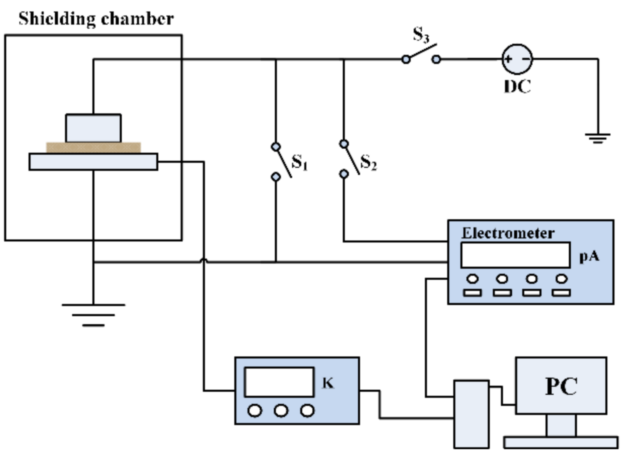

Figure 3. Schematic of experimental setup for measurement of thermally stimulated current.

3. Results

3.1. Breakdown Strength

The positive breakdown voltage and time to breakdown of pure oil and nanofluids are shown in Figure 4. Six repeat breakdown measurements for each sample were performed to generate the breakdown voltage and time to breakdown. The breakdown voltages of nanofluids are improved by the addition of three types of nanoparticles. It is worth noting that the improvements for breakdown voltage of Fe_3O_4 and TiO_2 nanofluids are up to 41.3% and 29.8% respectively, much higher than that for Al_2O_3 nanofluid of 7.4%. Especially, the times to breakdown in Fe_3O_4 and TiO_2 nanofluids are significantly increased by 76.3% and 69.6%, respectively. As for Al_2O_3 nanofluid, the time to breakdown is almost the same as that of pure oil. These indicate that the average propagation velocities of positive streamers are greatly slowed down due to the addition of Fe_3O_4 and TiO_2 nanoparticles, which is of great importance since it is closely related to the breakdown strength.

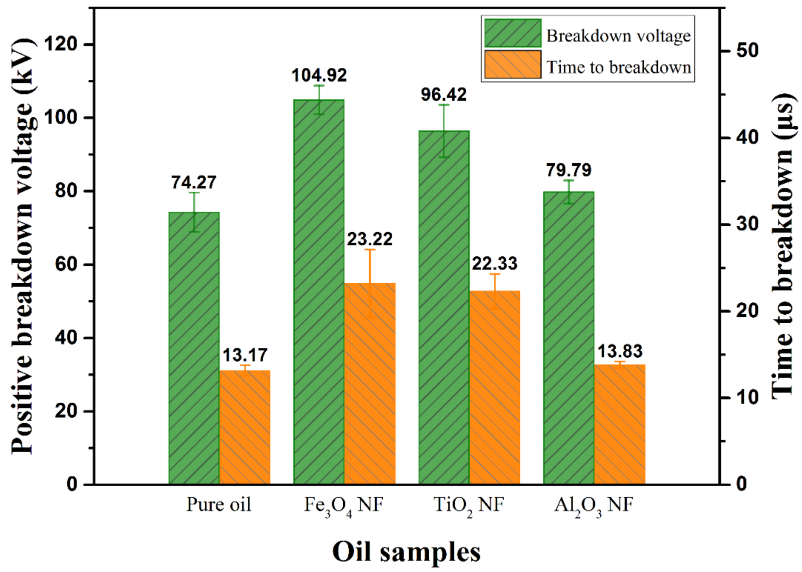

Figure 4. Positive breakdown strengths of pure oil and nanofluids.

3.2. Prebreakdown Streamers

The breakdown strength of transformer oil closely depends on the propagation process of prebreakdown streamers, including streamer shape and velocity [18–20], which is helpful to understand the fundamental modification effect of nanoparticles.

3.2.1. Shape

The propagation process of positive streamers can be divided into two stages in view of the shape development characteristics. In the initial stage, as shown in Figure 5, the streamers in pure oil and nanofluids behave similarly, exhibiting dense bush-like structures with slow velocities. While in the propagation stage, remarkable differences in the streamer shape are emerged in Fe_3O_4 and TiO_2 nanofluids. In pure oil, certain filaments develop into main channels with a rapidly increased length accompanied by other filaments fading away. But in the case of Fe_3O_4 and TiO_2 nanofluids, the main channels become much thicker with more lateral branches than that in pure oil during the whole propagation process, resulting in a much denser structure. In addition, the streamer propagation length is much shorter in Fe_3O_4 and TiO_2 nanofluids, which is of notable importance due to its relationship with electric breakdown event. Whereas in the Al_2O_3 nanofluid, streamers have no obvious change with a tree-like shape similar to that in pure oil.

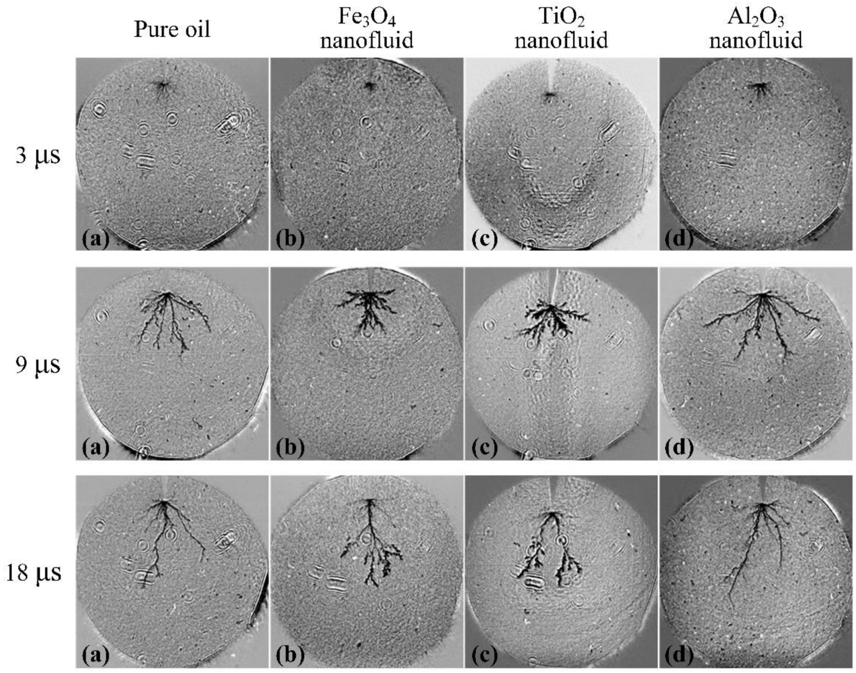

Figure 5. Prebreakdown streamers in pure oil and nanofluids. (a: pure oil, b: Fe_3O_4 nanofluid, c: TiO_2 nanofluid and d: Al_2O_3 nanofluid).

3.2.2. Propagation Length and Velocity

Figure 6 indicates that the propagation length of streamers varies considerably across pure oil and nanofluids. It is obvious that streamers in Fe_3O_4 and TiO_2 nanofluids keep shorter than that in pure oil during the whole development process. Moreover, it should be noted that the presence of nanoparticles extends the time to reach the maximum length, leading to a decrease of streamer velocity. The value of average velocity is calculated by dividing maximum length by arrival time. The streamers in pure oil develop fastest with a velocity of 1.40 km/s, followed by that in Al_2O_3, TiO_2 and Fe_3O_4 nanofluids, with velocities of 0.90 km/s, 0.73 km/s and 0.72 km/s, respectively. The values are in accordance with the propagation velocity of 2nd streamers in mineral transformer oil [21,22]. This indicates that the addition of TiO_2 and Fe_3O_4 nanoparticles dramatically affects the propagation velocity of streamers, which is lowered into around one half of that in pure oil.

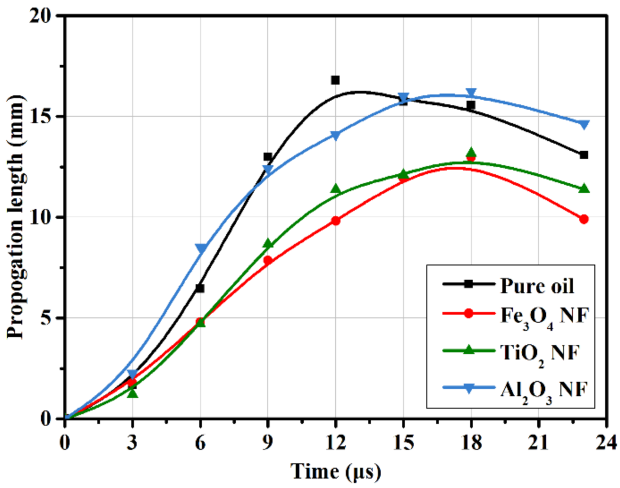

Figure 6. Prebreakdown streamer propagation length in pure oil and nanofluids versus propagation times.

4. Discussion

The electric field dependent molecular ionization has been proved to be the dominant contributor to streamer development in transformer oil, which has a great effect on breakdown strength of transformer oil [22–24]. In transformer oil, ionization occurs at streamer tip with a high electric field, generating positive ions and electrons. The highly mobile electrons swept back towards the positive electrode, leaving positive ions to form a net space charge region near the streamer tip, as presented in Figure 7a. These space charges slowly propagate towards the ground electrode and enhance the electric field towards the ground electrode [25]. So, the streamers tend to propagate straightly towards the ground electrode showing filamentary pattern, and breakdown happens when streamers extend throughout the gap. It is clear that the breakdown event is closely associated with charge transport process, which in turn, is influenced by the trap characteristics of transformer oil.

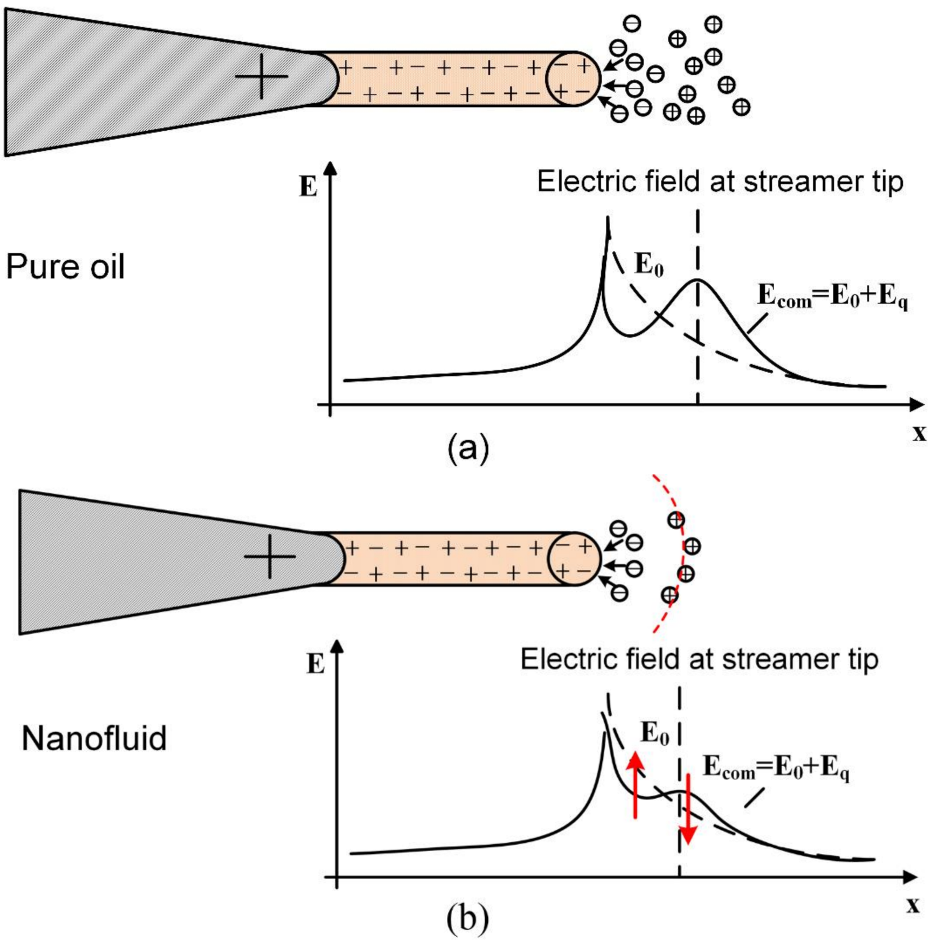

Figure 7. The electric field distribution in pure oil (**a**) and nanofluids (**b**).

To investigate the effects of nanoparticle materials on the trap characteristics of transformer oil, thermally stimulated currents (TSC) were measured. It is believed that TSC measurements can be used to investigate the nature and origin of charge carrier traps in dielectric, including the change of the density and energy level of the trap sites [26]. Figure 8 presents results on TSCs of pure oil and three types of nanofluids. It can be seen that the thermally stimulated currents are increased with the addition of nanoparticles. The trap characteristics including energy level and total trapped charges were calculated as shown in Table 1 [26,27]. The trap density is proportional to the total trapped charge if one-level of trap distribution is assumed [26]. It is indicated that the trap energy level of transformer oil is nearly unchanged by the addition of nanoparticles, which is considered to be shallow trap [14]. However, the trap densities are significantly increased by 65.5% and 59.0% in Fe_3O_4 and TiO_2 nanofluids, respectively. In Al_2O_3 nanofluid, the increase is only 14.6%. These results agree well with the effects of nanoparticle materials on breakdown and prebreakdown characteristics of nanofluids presented above.

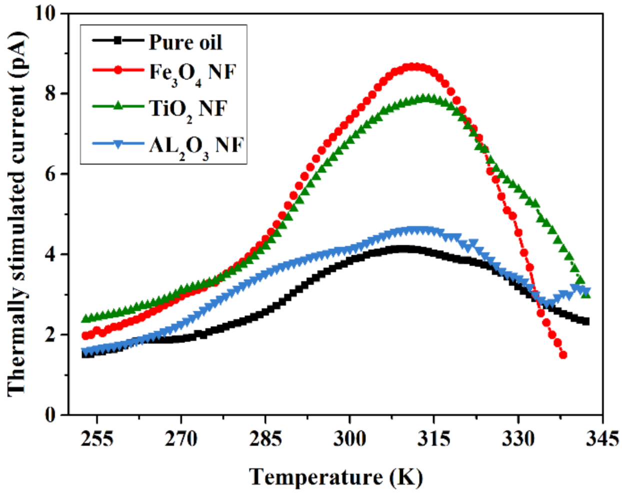

Figure 8. Thermally stimulated currents of pure oil and nanofluids.

Table 1. Trap characteristics of pure oil and nanofluids.

Oil Samples	Total Trapped Charges (nC)	Trap Level (eV)
Pure oil	7.86	0.340
Fe_3O_4 NF	13.01	0.345
TiO_2 NF	12.50	0.346
Al_2O_3 NF	9.01	0.346

By introducing more shallow traps, the probability of capturing electrons is more likely increased, meanwhile decreasing the energy of fast electrons during these trapping and de-trapping process in shallow traps. In all, the presence of Fe_3O_4 and TiO_2 nanoparticles greatly increases the density of shallow traps and forms more negative charges in the oil. As a result, the electric field is changed due to the superposition of space-charge field created by these negative space charges with applied electric field, as presented in Figure 7b. The electric field towards ground electrode is weakened, making it hard for positive streamers in Fe_3O_4 and TiO_2 nanofluids to propagate towards ground electrode. Moreover, the electric field between the space charge region and the positive electrode is enhanced, contributing to generate lateral branches. In comparison with streamers containing one or two main filaments, streamers with more branches can reduce the tip field due to mutual electrical shielding [5,28], which further slows down the propagation velocity of streamers, improving the positive breakdown strength. Whereas in Al_2O_3 nanofluid, the increase of breakdown strength is slight due to limited increase of trap density.

5. Conclusions

In this paper, the effects of nanoparticle materials on prebreakdown and breakdown characteristics in nanofluids under positive lightning impulse voltage were studied, and the conclusions are drawn as follows:

(1) Prebreakdown and breakdown characteristics of nanofluids are considerably affected by three types of nanoparticle materials. In Fe_3O_4 and TiO_2 nanofluids, positive streamers with more thicker and denser branches present greatly lowered propagation velocity, resulting in much higher

breakdown voltage than that in pure oil. Whereas in Al_2O_3 nanofluid, streamers are similar with that in pure oil, just exhibiting a slight increase of only 7.4% in breakdown voltage.

(2) Densities of shallow traps in Fe_3O_4 and TiO_2 nanofluids are significantly increased by the addition of nanoparticles. The rather higher density of shallow traps in nanofluids provides more opportunity to capture the fast electrons and convert into negative charges, inhibiting the propagation of positive streamers and improving the breakdown strength. But for Al_2O_3 nanofluid, the density of shallow traps is only slightly increased, making a less obvious improvement of breakdown strength.

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Conflicts of Interest: The authors declare no conflict of interest.

References

1. Wedin, P. Electrical breakdown in dielectric liquids—A short overview. *IEEE Electr. Insul. Mag.* **2014**, *30*, 20–25. [CrossRef]
2. Devins, J.C. Breakdown and prebreakdown phenomena in liquids. *Jpn. J. Appl. Phys.* **1981**, *52*, 4531–4543. [CrossRef]
3. Linhjell, D.; Lundgaard, L.; Berg, G. Streamer propagation under impulse voltage in long point-plane oil gaps. *IEEE Trans. Dielectr. Electr. Insul.* **1994**, *1*, 447–458. [CrossRef]
4. Massala, G.; Lesaint, O. A comparison of negative and positive streamers in mineral oil at large gaps. *J. Phys. D Appl. Phys.* **2001**, *34*, 1525. [CrossRef]
5. Dung, N.V.; Hoidalen, H.K.; Linhjell, D.; Lundgaard, L.E.; Unge, M. Effects of reduced pressure and additives on streamers in white oil in long point-plane gap. *J. Phys. D Appl. Phys.* **2013**, *46*, 25501. [CrossRef]
6. Liu, Q.; Wang, Z.D. Streamer characteristic and breakdown in synthetic and natural ester transformer liquids under standard lightning impulse voltage. *IEEE Trans. Dielectr. Electr. Insul.* **2011**, *18*, 285–294. [CrossRef]
7. Chadband, W.G. The electrical breakdown of insulating oil. *Power Eng. J.* **1992**, *6*, 61–67. [CrossRef]
8. Beroual, A.; Tobaz, R. Prebreakdown Phenomena in Liquid Dielectrics. *IEEE Trans. Dielectr. Electr. Insul.* **1986**, *21*, 613–627. [CrossRef]
9. Babaeva, N.Y.; Kushner, M.J. Effect of inhomogeneities on streamer propagation: II. Streamer dynamics in high pressure humid air with bubbles. *Plasma Sources Sci. Technol.* **2009**, *18*, 35010–35013. [CrossRef]
10. Dung, N.V.; Hoidalen, H.K.; Linhjell, D.; Lundgaard, L.E.; Unge, M. Influence of Impurities and Additives on Positive Streamers in Paraffinic Model Oil. *IEEE Trans. Dielectr. Electr. Insul.* **2012**, *19*, 1593–1603. [CrossRef]
11. Lesaint, O.; Jung, M. On the relationship between streamer branching and propagation in liquids: Influence of pyrene in cyclohexane. *J. Phys. D Appl. Phys.* **2000**, *33*, 1360. [CrossRef]
12. Lue, Y.-F.; Hung, Y.-H.; Li, F.-S.; Teng, T.-P.; Chen, S.-Y.; Wu, C.-H.; Ou, Y.-C. Performance Assessment and Scooter Verification of Nano-Alumina Engine Oil. *Appl. Sci.* **2016**, *6*, 258. [CrossRef]
13. Abhishek, R.; Hamouda, A.A. Effect of Various Silica Nanofluids: Reduction of Fines Migrations and Surface Modification of Berea Sandstone. *Appl. Sci.* **2017**, *7*, 1216. [CrossRef]
14. Du, Y.; Lv, Y.; Li, C.; Chen, M.; Zhou, J.; Li, X. Effect of electron shallow trap on breakdown performance of transformer oil-based nanofluids. *J. Appl. Phys.* **2011**, *110*, 619–622. [CrossRef]
15. Liu, R.; Pettersson, L.A.A.; Auletta, T.; Hjortstam, O. Fundamental research on the application of nano dielectrics to transformers. *IEEE Conf. Electr. Insul. Dielectr. Phenom.* **2011**, 423–427.
16. Sima, W.; Shi, J.; Yang, Q.; Huang, S.; Cao, X. Effects of conductivity and permittivity of nanoparticle on transformer oil insulation performance: Experiment and theory. *IEEE Trans. Dielectr. Electr. Insul.* **2015**, *22*, 380–390. [CrossRef]
17. Council on Large Electric Systems (CIGRE). *Effect of Particles on Transformer Dielectric Strength*; Working Group 17 of Study Committee: Paris, France, 2000.

18. Peppas, G.D.; Charalampakos, V.P.; Pyrgioti, E.C.; Gonos, I.F. Electrical and optical measurements investigation of the pre-breakdown processes in natural ester oil under different impulse voltage waveforms. *IET Sci. Meas. Technol.* **2016**, *10*, 545–551. [CrossRef]
19. Mccluskey, F.M.J.; Denat, A.; Lesaint, O. Breakdown and prebreakdown phenomena in liquids under positive impulse voltages. *IEEE Trans. Dielectr. Electr. Insul.* **1994**, *1*, 377–382. [CrossRef]
20. Sima, W.; Liu, C.; Yang, M.; Shao, Q.; Xu, H.; Liu, S. Plasma model of discharge along a dielectric surface in N2/O2 mixtures. *Phys. Plasmas* **2016**, *23*, 63508. [CrossRef]
21. Torshin, Y.V. Schlieren registration of electrohydrodynamics phenomena in dielectric liquids under lightning impulse. *IEEE Trans. Dielectr. Electr. Insul.* **2009**, *16*, 470–474. [CrossRef]
22. Hwang, J.G.; Zahn, M.; Pettersson, L.A.A. Mechanisms behind positive streamers and their distinct propagation modes in transformer oil. *IEEE Trans. Dielectr. Electr. Insul.* **2012**, *19*, 162–174. [CrossRef]
23. Jadidian, J.; Zahn, M.; Lavesson, N.; Widlund, O.; Borg, K. Impulse breakdown delay in liquid dielectrics. *Appl. Phys. Lett.* **2012**, *100*, 909. [CrossRef]
24. Akishev, Y.; Arefikhonsari, F.; Demir, A.; Grushin, M.; Karalnik, V.; Petryakov, A. The interaction of positive streamers with bubbles floating on a liquid surface. *Plasma Sources Sci. Technol.* **2015**, *24*, 65021. [CrossRef]
25. O'Sullivan, F.; Hwang, J.G.; Zahn, M.; Hjortstam, O. A Model for the Initiation and Propagation of Positive Streamers in Transformer Oil. In Proceedings of the 2008 IEEE International Symposium on Electrical Insulation, Vancouver, BC, Canada, 9–12 June 2008; pp. 210–214.
26. Chen, R.; Kirsh, Y. *Analysis of Thermally Stimulated Processes*; Pergamon Press: Oxford, UK, 1981; pp. 78–92.
27. Lu, J.; Ding, L.; Li, C.R.; Wang, W. Study on the trap distribution characteristic of alumina ceramics. In Proceedings of the IEEE Conference on Electrical Insulation and Dielectric Phenomena, Kitchener, ON, Canada, 14–17 October 2001; pp. 381–384.
28. Unge, M.; Singha, S.; Van Dung, N.; Linhjell, D.; Ingebrigtsen, S.; Lundgaard, L.E. Enhancements in the lightning impulse breakdown characteristics of natural ester dielectric liquids. *Appl. Phys. Lett.* **2013**, *102*, 172905. [CrossRef]