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# Partial Discharge Development in Oil-Based Nanofluids: Inception, Propagation and Time Transition

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**ABSTRACT** Oil-based nanofluids have been indicated to enhance the breakdown strength and dielectric behavior of mineral oil. However, partial discharge (PD) development in these new materials has not yet been clarified. This study aims to deeply investigate PD development in nanofluids considering the role of the electrical double layer (EDL) around nanoparticles. Two types of nanoparticles (TiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub>) with different EDL thicknesses were used. Nanofluids were prepared using the two-step method, and their proper composition was adopted after considering their stability and avoiding the drawbacks that are present when surfactants are used. The prepared nanofluids together with the base oil were tested for PD development. First, the PD inception voltage was evaluated and analyzed using the Weibull distribution. Then, PD parameters including the PD magnitude and repetition rate were obtained for both types of nanofluids; these values were compared with the corresponding results of the base oil. Finally, the PD time transition was acquired over ten minutes of applied voltage using the segmented memory mode of the oscilloscope. Based on obtained results, physical mechanisms behind PD activity are proposed and discussed. It was found that nanoparticles with a large EDL thickness could more effectively suppress PD activity.

**INDEX TERMS** Transformer oil, nanofluids, partial discharge development, electrical double layer.

#### I. INTRODUCTION

Oil-Based nanofluids have become increasingly studied in the dielectric community for use as new insulating and cooling media in power transformers [1], [2]. Nanofluids are composed of a base transformer oil filled with certain types and amounts of nanoparticles. Depending on the type and amount used, one or more properties of the transformer oil can be improved. In terms of improved thermal properties, there are several types that have been successfully used for this purpose. These types include boron nitride [3]–[5], aluminum nitride [6], and carbon- based nanostructures [7], [8], such as carbon nanotubes, graphene, and diamond nanoparticles. Some of these types can maintain acceptable breakdown strength, such as boron nitride [5] and graphene oxide [8], [9], while others exhibit a decrease in breakdown strength, such as aluminum nitride [6] and carbon nanotubes [7].

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On the other hand, metal oxides have been effectively used for improving dielectric properties [10]-[13]. The enhancement percentage depends on many factors related to the nanoparticles themselves or related to the base oil. Regarding different types of nanoparticles, the main factors that affect their dielectric properties are their concentrations, sizes, and shapes [14]-[16]. For the base oil, the enhancement percentage depends on the condition of the oil itself. The better the condition of the oil is, the lower the enhancement percentage. For example, the enhancement percentages obtained for SiO<sub>2</sub> nanoparticles [10] and TiO<sub>2</sub> nanoparticles [12] were 27 and 22% with a base oil having a breakdown voltage above 30 kV, while these enhancement percentages were 171 and 62% for the same nanoparticles, respectively, when using a base oil having a low breakdown voltage below 10 kV [16]. This motivated researchers to use nanofluids to improve the dielectric properties of aged transformer oils [17], [18].

Although many studies have investigated the dielectric and thermal properties of oil-based nanofluids, there are limited studies that have investigated their partial discharge (PD) activity. Clarifying PD activity in these materials will provide two different benefits. The first benefit is to adopt suitable nanofillers that can suppress the PD activity in the oil. The second benefit is to identify the features of PD activity in such materials, which are important for diagnostic purposes.

PD in oil-based nanofluids was investigated by Jin *et al.* [19], where they investigated PD inception voltage (PDIV) and PD magnitude. However, the study was limited to the application of DC voltage, which is not representative of actual transformers. In [20], PD activity was investigated for three different types of nanoparticles under an applied AC voltage. The most conductive nanoparticles exhibited the worst performance for PD activity; in fact, at a certain voltage level, the PD magnitudes and repetition rates with these types of nanoparticles increased above those of mineral oil. A similar conclusion was reached in [21], where PD occurred more easily in nanofluids filled with conductive nanoparticles than in those filled with semiconductive nanoparticles. In [12], the effect of both BaTiO<sub>3</sub> and TiO<sub>2</sub> nanoparticles on PD was investigated under the application of AC voltage. Two different weight fractions of both nanoparticles were considered. For the same tip radius, the PDIV increased in the nanofluid samples compared to that in the mineral oil. However, the PD activity at the PDIV indicated high PD magnitudes in the nanofluids for almost all cases, especially with tips having 20 and 40  $\mu$ m radius.

Most previous studies on PD characteristics in nanofluids focused on the effect of nanoparticle type and neglected the impact of the oil/nanoparticle interface. Since PD activity depends on charge propagation through the fluid, it is believed that the electrical double layer (EDL) formed around nanoparticles will have an important influence on PD characteristics. In addition, previous studies did not discuss the time transition of PD activity. Through the PD time transition, the PD characteristics and mechanisms can be illustrated [22]. Additionally, the time transition represents an important diagnostic parameter, through which the progress of the PD up to the breakdown can be evaluated [23].

From this viewpoint, we aimed to deeply investigate PD characteristics in the present study considering the role

of the EDL at the oil/nanoparticle interface. Two types of nanoparticles with different EDL thicknesses were used. These nanoparticles were  $TiO_2$  and  $Al_2O_3$  nanoparticles. First, proper compositions of these nanofluids were adopted. Then, nanofluids with these compositions were prepared and tested for their PD characteristics. The investigated PD parameters are the PDIV, PD magnitude and PD repetition rate. Additionally, the time transition of PD activity was investigated. Based on the obtained results, the physical mechanisms behind PD activity are proposed and discussed.

### II. NANOFLUID PREPARATION AND COMPOSITION A. NANOFLUID PREPARATION

The nanoparticles used in this study were titania  $(TiO_2)$  and alumina  $(Al_2O_3)$  nanoparticles. Both are obtained from Sigma Aldrich and have a grain size of 20 nm with 99% purity. The transformer oil used was highly refined Shell Diala S2 ZU-I mineral oil.

The preparation of nanofluid samples followed the two-step preparation method recommended in previous studies [24]. These procedures are chronologically illustrated in Fig. 1. First, nanoparticles were dried in a drying oven at 60 °C for 24 hours to eliminate the impact of moisture content. Then, the oil and the dried nanoparticles were mixed using a magnetic stirrer for 5 minutes. After that, the mixtures were placed in an ultrasonic bath for 2 hours with a 5-minute rest after each 30 minutes. Finally, the nanofluid samples and the base oil were degassed for 24 hours in a vacuum oven at a temperature of 45 °C. The degassing process was necessary to remove the absorbed moisture and the formed bubbles during the ultrasonic stage.

#### **B. ZETA POTENTIAL MEASUREMENTS**

Both types of nanoparticles have different EDL thicknesses. To compare these thicknesses, zeta potentials were measured experimentally using a MALVERN ZETASIZER instrument at two different weight fractions, 0.01 g/L and 0.04 g/L, as depicted in Table 1. For both weight fractions,  $Al_2O_3$  nanoparticles exhibited higher zeta potential values than the corresponding zeta potential values for TiO<sub>2</sub> nanoparticles.



FIGURE 1. Preparation of nanofluid samples.

| TABLE 1. | Zeta Pote | ntial measur | ements for | Al <sub>2</sub> O <sub>3</sub> | and TiO <sub>2</sub> | nanoparticles. |
|----------|-----------|--------------|------------|--------------------------------|----------------------|----------------|
|----------|-----------|--------------|------------|--------------------------------|----------------------|----------------|

| Sampla              | Al <sub>2</sub> O <sub>3</sub> na | anofluid | TiO <sub>2</sub> nanofluid |          |  |
|---------------------|-----------------------------------|----------|----------------------------|----------|--|
| Sample              | 0.01 g/L                          | 0.04 g/L | 0.01 g/L                   | 0.04 g/L |  |
| Zeta potential (mV) | 18.9                              | 38.0     | 14.9                       | 27.8     |  |

This result indicates that the EDL thickness for  $Al_2O_3$  nanoparticles is larger than that for TiO<sub>2</sub> nanoparticles.

## C. STABILITY AND NANOFLUID COMPOSITION

Before describing the preparation process of nanofluids, it is essential to point out that the successful application of nanofluids requires a number of rationally designed processes. This includes maintaining a stable suspension of nanoparticles without any chemical changes in the base fluid. The stability of nanofluids can be maintained by using surfactants with polar head groups. However, for dielectric nanofluids, using surfactants has several drawbacks. First, the presence of surfactants increases the amount of acids in the oil, thereby having a negative impact on the dielectric dissipation factor [25]. Additionally, the increase in the amount of acids in transformer oil catalyzes the corrosion of metallic parts and promotes the degradation of cellulose and paper insulation. Second, at temperatures above 60 °C, the surfactant loses its ability to bond with nanoparticles [26], allowing the nanoparticles to agglomerate. In an actual transformer, the oil has a much broad range of application temperatures that may reach 90 °C. Based on these drawbacks, the use of surfactants as stabilizing agents in dielectric nanofluids is unlikely.

To prepare surfactant-free nanofluids and keep them stable, it is important to carefully adopt a suitable weight fraction. To do so, preliminary experiments were carried out to select the suitable weight fractions of the chosen nanoparticles. In these experiments, the nanofluid stability was evaluated using UV/Vis spectroscopy. Usually, the evaluation of nanofluid stability based on UV/Vis spectroscopy is performed by evaluating the decrease in absorbance peaks over time [27]. However, this methodology is not sufficient since it implies that nanofluids after preparation are stable and have a uniform distribution of nanoparticles. Therefore, this study proposed the use of the Beer-Lambert law to directly evaluate the stability after preparation.

According to the Beer-Lambert law [28], the sample absorbance is linearly related to the nanoparticle weight fraction. To obtain this linear representation, a series of samples was prepared using TiO<sub>2</sub> nanoparticles starting from very small nanoparticle weight fractions up to 0.1 g/L. These weight fractions were 0.001, 0.004, 0.007, 0.01, 0.04, 0.07 and 0.1 g/L. The maximum absorbances of these samples were measured and plotted against their weight fraction, as shown in Fig. 2. The linear relationship of absorbance could be realized from the first portion of the graph and was represented by the solid line. It can be observed that



FIGURE 2. Maximum absorbances of the nanofluid samples against the weight fraction.

the absorbance at high weight fractions deviates from the linear relationship. The measured values of absorbance were lower than the expected values from the line. This result indicated that there was a decrease in the total surface area of nanoparticles as a result of agglomeration. As a result, the weight fraction of 0.01 g/L was adopted for the present study since it was the highest weight fraction that exhibited a fully confinement to the linear relationship. For additional verification, the absorbance of the 0.01 g/L sample after 24 hours of preparation was measured and was exactly the same as that measured after preparation. Similar results were obtained for  $Al_2O_3$  nanoparticles.

# **III. EXPERIMENTAL SETUP**

#### A. ELECTRODE CONFIGURATION

The electrode configuration used to generate PD is schematically shown in Fig. 3. It consists of a needle-to-plane configuration to simulate metallic particles in actual transformers. The needle is made from tungsten and has a tip radius of 10  $\mu$ m. The gap spacing between the needle and plane electrodes was kept at 3 mm for all samples. The plane electrode was made from copper and had a diameter of 8 cm and a thickness of 5 mm. In parallel to the electrode system, a 0.1 nF coupling capacitor was used. The electrode setup was connected to a 50 Hz/100 kV testing transformer that was PD free. A protective resistor was used between the testing transformer and electrode setup to prevent large currents in the case of sample breakdown, thereby protecting the PD measuring system.

#### **B. PD MEASURING SYSTEM**

The PD measuring system was based on the electrical sensing of current pulses produced by PD events as described in [29]. These pulses flowed through the impedance detector, which was a 50  $\Omega$  surface-mounted resistor. This resistor was surface mounted to avoid acquiring any noise from the surroundings, while a resistance of 50  $\Omega$  was chosen to achieve a good match with the measuring cables and input impedance of the oscilloscope. A Tektronix DPO7104C oscilloscope was used



FIGURE 3. Electrode configuration and PD measuring system for the nanofluids and the base oil.

with a bandwidth of 1 GHz and a maximum sampling rate of 20 GS/s. The oscilloscope was set to operate in the Fast-Frame segmented memory mode, which enabled the capture of only PD events with a high resolution and eliminated the dead time intervals between them.

#### **IV. PD DEVELOPMENT IN NANOFLUIDS**

## A. PD INCEPTION

For measuring the PDIV, the input voltage was gradually increased until PDs were detected. The PDIV was measured ten times for the base oil and nanofluids. For the performance evaluation of the insulating fluids, it is important to obtain the average PDIV as well as the lowest possible PDIV. To do so, a two-parameter Weibull distribution was used using the following function.

$$f(x) = 1 - e^{-\left(\frac{x}{\alpha}\right)^{\rho}} \tag{1}$$

where x represents the PDIV in kV, f(x) is the cumulative probability,  $\alpha$  is the scale parameter that corresponds to 63.2% probability, and  $\beta$  is the shape parameter that indicates the level of data scattering. Fig. 4 shows Weibull plots for the TiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> nanofluids as well as for the base oil. The shape and scale parameters for these samples are depicted



FIGURE 4. Weibull plots for the PDIVs of the nanofluids and base oil.

TABLE 2. Weibull distribution parameters for the PDIVs of the nanofluids and base oil.

| Sample                                   | Scale<br>parameter | Shape<br>parameter | 10%  | 50%  |
|--|--------------------|--------------------|------|------|
| Al <sub>2</sub> O <sub>3</sub> nanofluid | 26.24 kV           | 23.94              | 24.8 | 26.2 |
| TiO <sub>2</sub> nanofluid               | 24. 62 kV          | 24.12              | 23   | 24.9 |
| Base oil                                 | 22.44 kV           | 63.43              | 21.7 | 22.2 |

in Table 2. It is clear that the PDIVs for both nanofluids were higher than that for the base oil. However, the PDIV enhancement percentage for the  $Al_2O_3$  nanofluid was higher than that for the TiO<sub>2</sub> nanofluid. From these plots, the PDIVs at probabilities of 10 and 50% were deduced and provided in Table 2. The PDIVs at 50% probability were related to the mean PDIV values, while the PDIVs at 10% probability were related to the lowest possible PDIV. The  $Al_2O_3$  nanofluid exhibited an enhancement percentage of approximately 18 and 14% for the PDIV at 50 and 10% probability, respectively.

#### **B. PD PROPAGATION**

To understand the PD propagation characteristics, the PD magnitude and PD repetition rate were obtained for the nanofluids and base oil over ten minutes of applied voltage. Then, these parameters were presented against the phase distribution over a phase step of ten degrees. The voltage was set at 26.3 kV, which was higher than the PDIVs for all samples. The PD magnitude was expressed in pC, and the PD repetition rate was expressed in pulses per second (pps). For the base oil shown in Fig. 5, PDs occurred during the negative polarity of the applied voltage. From the PD repetition rate, it could be recognized that PDs were distributed from 180° to 270°, and reached a maximum around the negative peak. The PD magnitude also attained its highest value around the negative peak. In this case, the highest PD magnitude at the negative polarity was approximately 280 pC.



FIGURE 5. PD propagation characteristics for the base oil.



**FIGURE 6.** PD propagation characteristics for the oil-based  $\mathrm{TiO}_2$  nanofluids.

For the oil-based TiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> nanofluids, the PD activity was effectively suppressed, as observed in Figs. 6 and 7, respectively. For both nanofluids, the PDs were concentrated around the negative peak of the applied voltage. The maximum PD repetition rate of the TiO<sub>2</sub> nanofluid was approximately 0.13 pps, and this value was approximately half of that of the base oil. Additionally, the maximum PD magnitude



FIGURE 7. PD propagation characteristics for the oil-based Al<sub>2</sub>O<sub>3</sub> nanofluids.

of the TiO<sub>2</sub> nanofluid decreased to approximately 200 pC, which was low compared to that of the base oil case. When the Al<sub>2</sub>O<sub>3</sub> nanofluid was used, extreme enhancements in PD activity were observed, especially in the PD repetition rate. The maximum PD repetition rate was 0.002 pps, while the maximum PD magnitude was approximately 200 pC at the negative polarity, which was closer to that of the TiO<sub>2</sub> nanofluid.



FIGURE 8. Comparison of the PD characteristics for the base oil and nanofluids.

To summarize, Fig. 8 depicts the PD measurement results for the nanofluids and base oil. These include the overall PD repetition rate and the total PD magnitude over ten minutes. When using nanofluids, the number of PD pulses and the total PD magnitude were lower than that of the base oil. The comparison between the TiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> nanofluids indicated that the  $Al_2O_3$  nanofluid suppressed PD activity more effectively than the  $TiO_2$  nanofluid.

# C. PD TIME TRANSITION

In this study, the time transition of PD activity was acquired. This includes the time transition for the PD repetition rate. The PD time transition was used to deeply understand and clarify the charging phenomena of the PD in the base oil and nanofluids. Time transition was performed for ten minutes of continuous applied voltage. For each minute, PD parameters were acquired. Fig. 9 shows the time transition of the PD repetition rate for all samples. The experimental results were represented by different markers, while various lines represent the fit of each set of data. For the base oil, it was clear that the number of PD pulses was higher than that for the nanofluid samples. Additionally, the PD repetition rate for the base oil exhibited an increasing trend over time, increasing the risk of breakdown. For both nanofluids, not only were their PD repetition rates lower than the base oil, but they also exhibited a decreasing trend over time. The difference between the trends of the PD repetition rates in the base oil and nanofluids could be explained by the charging process, as will be discussed in the next section.



**FIGURE 9.** Time transition of the PD repetition rate over ten minutes for the nanofluids and base oil.

## **V. PHYSICAL MECHANISMS**

# A. FACTORS AFFECTING PD ACTIVITY

PD activity depends on three factors. The first factor is initial electron generation. For the needle-to-plane configuration, initial electrons are generated from the needle tip during negative polarity and from the oil gap during positive polarity. Initial electrons are generated more easily from the metallic surface of the needle tip by a Fowler-Nordheim electron injection [30]. Thus, PD inception usually occurs at negative polarity for both the base oil and nanofluids.

The second factor affecting PD activity is the charge buildup through the oil gap. When the electric field at the needle tip reaches the critical level, the liquid in the vicinity of the needle is ionized, creating fast-moving electrons toward the anode as well as slow-moving cations. If there is a sufficient time period for charge buildup, a PD event occurs.

The third factor is the applied electric field stress. When the electric field increases, the possibility of PD generation increases. It is important to point out that a high electric field will cause a large charge buildup, thereby enabling the generation of more PD events.

# B. ROLE OF NANOFILLERS

When adding nanofillers into transformer oil, they have a dual role in the charge trapping process. The first role is the charge trapping provided by the nanoparticles themselves. The ability of nanoparticles to trap charges depends on their type. In this regard, semiconductive nanoparticles, such as  $TiO_2$  nanoparticles, have a higher ability for charge trapping than  $Al_2O_3$  nanoparticles. There are four modes of streamer propagation in insulating fluids [31]. The charge trapping process by nanoparticles usually occurs in the first and second modes, which have relatively low velocities. For PD activity, the electric field is extremely high at the needle tip. Thus, PDs propagate with large velocities above the velocity of the first and second modes. Therefore, it is difficult to trap charges by nanoparticles themselves, unless these charges lose part of their energy.

The second role in the charge trapping process is provided by the electrical double layer (EDL) formed around the nanoparticle surface in contact with transformer oil. When nanoparticles come in contact with the oil, they attract counter ions and form a strong bound layer directly adjacent to their surfaces. This layer is called the stern layer. Next to this layer, a diffuse layer is formed, in which the number of counter ions decreases gradually until reaching zero in the free oil volume. Both layers form the EDL. The thickness of the EDL depends on the isoelectric point of nanoparticles and the pH value of oil. This thickness can be evaluated through zeta potential measurements, which measure the potential in a certain plane called the slipping plane and located in the diffuse layer. The higher the zeta potential is, the larger the EDL thickness [32]. Using surfactants can widen the EDL thickness, but due to the drawbacks of surfactants mentioned above in Section II.C, their usage has been avoided in the present study.

When using nanofluids, the EDL works to capture charge carriers. In the PD process, this capture process delays the charge buildup through the oil gap, and thus, high electric fields are needed to produce PDs. Accordingly, the PDIVs for the TiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> nanofluids increase, as shown in Fig. 4. Additionally, the PD repetition rate and the maximum PD magnitude were lower in the nanofluids at the same applied voltage, as shown in Figs. 6 and 7.

The time transition of PD activity is affected by subsequent PD events. When a PD event occurs, residual space charges remain in the vicinity of the needle tip, as schematically shown in Fig. 10. For the base oil, these charges facilitate the occurrence of other PD events. Accordingly, the time transition of the PD repetition rate for the base oil exhibited an increasing trend over time, as illustrated in Fig. 9.



(a) During the PD event

(b) After the PD event

**FIGURE 10.** Time transition of PD activity in the base oil.



FIGURE 11. Time transition of PD activity in the nanofluids.

When using nanofluids, their EDLs could trap residual charges after a certain PD event, as shown in Fig. 11, thereby limiting the impact of these charges on subsequent PD events. Moreover, these charges become part of the EDL and increase its thickness. Thus, the time transition of the PD repetition rate for nanofluids exhibited a decreasing trend over time.

The zeta potential measurements in Section II.B show higher values for the  $Al_2O_3$  nanoparticles when compared to that for the TiO<sub>2</sub> nanoparticles. This means that  $Al_2O_3$ nanoparticles have a large EDL thickness. Thus, better suppression of PD activity is observed with the  $Al_2O_3$  nanofluid compared to that with the TiO<sub>2</sub> nanofluid. This result caused the  $Al_2O_3$  nanofluid to have a high PDIV, low PD repetition rate, and low PD magnitude.

#### **VI. CONCLUSION**

In this study, partial discharge (PD) development in oilbased nanofluids was investigated considering the role of the electrical double layer (EDL) around nanoparticles. Two types of nanoparticles with different EDL thicknesses were used:  $Al_2O_3$  nanoparticles and TiO<sub>2</sub> nanoparticles.  $Al_2O_3$ nanoparticles had a large EDL thickness based on the zeta potential measurements. Finally, the following conclusions were drawn:

1- The PDIVs for both nanofluids were higher than that for the base oil, and the PDIV enhancement percentage for the  $Al_2O_3$  nanofluid was higher than that for the TiO<sub>2</sub> nanofluid.

2- The PD repetition rate and PD magnitude were lower in the nanofluids at the same applied voltage compared to those in the base oil. Similar to the PDIV, Al<sub>2</sub>O<sub>3</sub> nanofluids exhibited the best suppression of PD activity.

3- The enhancements in PD activity for the nanofluids were explained considering the role of the EDL in capturing charge carriers and delaying the PD charge buildup through the oil gap.

4- The time transition of the PD repetition rate for the base oil exhibited an increasing trend over time, while that for nanofluids exhibited a decreasing trend. This result was explained by the ability of EDLs to trap residual charge after a certain PD event, thereby limiting the impact of this charge on subsequent PD events.

The present study may be a guideline for the future application and development of dielectric nanofluids. Furthermore, this study will enable the adoption of suitable nanofillers that can suppress PD activity in oil and may be beneficial for PD-based diagnostics in such new materials.

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