



Article The Role of the Accumulated Surface Charge on Nanoparticles in Improving the Breakdown Strength of Liquid and Solid Insulation

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Abstract: In this paper, the role of the accumulated surface charge on the surfaces of nanoparticles on breakdown strength for liquid and solid dielectrics is presented. The breakdown strengths of a nanofilled liquid dielectric and a solid dielectric are evaluated. The evaluation was conducted considering different nanoparticle material types with different nanofiller loadings. Accordingly, the preparation of transformer oil nanofluid and silicone rubber nanocomposites was performed with different nanofillers of the same average particle size. Breakdown voltage was measured for all the prepared samples, both liquid and solid. The interpretation of the obtained results is presented.

Keywords: breakdown strength; transformer oil; silicone rubber; surface charges; nanocomposites; dielectric constant

1. Introduction

Recently, the use of nanotechnology to improve the dielectric properties of liquid and solid insulating materials has increased. The increase in using nanotechnology to improve the dielectric properties of electrical insulation (liquid or solid) is due to the promising properties achieved by the addition of small amounts of nanoparticles. Hence, the addition of semi-conductive nanoparticles to transformer oil results in increased breakdown strength [1–5], as well as improved thermal properties [5–7]. Adding magnetic nanoparticles to transformer oil was also found to increase the breakdown strength of liquid dielectrics [8,9]. Regarding solid dielectrics, nanofillers can also increase their breakdown strength. This was validated with epoxy [10–18], silicone rubber [16,19], ethylene propylene diene monomer rubber (EPDM) [16] and cross-linked polyethylene dielectrics [20]. In fact, breakdown strength is one of the most important dielectric properties for both liquid and solid insulation. Therefore, it is very important to know the role of nanoparticles in improving the breakdown strength for these types of insulation (liquid and solid). In other words, a full understanding of the breakdown mechanisms for nanofluids and solid nanocomposites is required.

Regarding breakdown mechanisms, researchers attribute the increase in the breakdown strength of nanofluids to different reasons. Moisture binding resulting from the hydrophilic nature of nanoparticles [21] is one such reason. The hydrophilic nature of nanoparticles decreases the spread of moisture through the insulating liquid due to binding. Therefore, adding nanoparticles to insulating liquids increases its breakdown strength. Charge trapping is another reason that leads to the improved breakdown strength of nanofluids: this occurs as a result of the free electrons being trapped in oil [22]. The trapping process decreases the energy of free charges due to the ionization process, which leads



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Copyright: © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). to an increased breakdown strength in the nanofluid. Electric field distortion also has a vital role in the percentage increase in nanofilled-oil breakdown strength, as demonstrated in [23]. It was found that the increase in nanofiller content distorts the electric field that leads to a decrease in breakdown strength when it is increased beyond the optimal loading. On the other hand, breakdown strength of solid nanocomposites is mainly improved due to charge trapping that leads to the reduced energy of free electrons which share in the breakdown process, as introduced in [24–27].

According to [3,28], increasing the dielectric constant of nanoparticles results in a higher percentage increase in the breakdown strength of transformer oil nanofluids. However, the increase in the dielectric constant of nanoparticles results in a lower percentage increase in the breakdown strength of polymer nanocomposites [26]. In fact, this effect needs to be further discussed taking into account both liquid and solid insulation and also the effect of the nanofiller dielectric constant on the accumulated nanoparticle surface charge. Therefore, the main objective of this paper is to highlight the effect of the nanoparticle surface taking the mechanism of nanoparticle surface charge accumulation into consideration.

In this paper, the effect of accumulated nanoparticle surface charges and their role in the breakdown process of nanofluids and solid nanocomposites are presented. As the nanoparticle dielectric constant has a very close relation with the formation of surface charges on its surface, the effect of the nanofiller dielectric constant on the breakdown strength of liquid and solid insulation is introduced. This effect was studied in transformer oil as it is a commonly used liquid insulation. Silicone rubber was chosen as a solid insulation due to its widespread use in electric power equipment. The preparation of transformer oil nanofluid and silicone rubber nanocomposite samples was undertaken with nanofillers with different dielectric constants at different concentrations. The breakdown strength for all the prepared samples, either liquid or solid, was measured and evaluated. The evaluation was conducted based on average breakdown strength, as well as on breakdown at 10% and 50% probabilities. The obtained results are interpreted and a statement in terms of the effect of the nanoparticle dielectric constant is given. Finally, this paper makes important recommendations about the selection roles of nanofiller material types for liquid and solid insulation.

2. Sample Preparation and Characterization

In this section, the preparation of samples of transformer oil nanofluids and silicone rubber nanocomposites is presented. The nanofillers used in the present study were spherically shaped (SiO₂, ZrO₂ and TiO₂), such fillers having a particle size of 20–30 nm. Characterization of the prepared silicon rubber nanocomposite samples was performed using field scanning electron microscopy (FESEM) to examine nanoparticle dispersion inside the prepared samples.

2.1. Sample Preparation

The transformer oil nanofluid samples were prepared using highly purified transformer oil (Diala B). The addition of each specified type of nanoparticle to transformer oil was carried out at different concentrations. These concentrations were 0.02, 0.08, 0.14, 0.2 and 0.25 g/L for each nanomaterial. The prepared mixture was stirred in a magnetic stirrer (Model MSH-20D) for 20 min. Next, ultrasonic waves were applied to the mixture using an ultrasonic homogenizer (Model UP400S) for a further 20 min. The prepared samples were degassed in a vacuum chamber for 24 h to prevent the formation of air bubbles during the preparation process and their effect on the breakdown strength results.

On the other hand, preparation procedures for the silicone rubber nanocomposite samples were performed using 10 g liquid silicone rubber mixed homogenously with 10 g of its hardener. Ethyl Methyl Ketone (2-butanone) solvent (with a purity greater than 99%, supplied by ReAgent) was added to the mixture to reduce its viscosity. A quantity of nanoparticles of the same specified types was then added to the mixture and stirred for

10 min (the same stirrer as that used for the transformer oil). In order to achieve a good dispersion of nanoparticles, the mixture was subjected to ultrasonic waves for 30 min (using the same ultrasonic homogenizer used in the transformer oil preparation). The mixture was left in a vacuum chamber at room temperature for 20 days to dry. Again, the vacuum chamber was used to prevent the formation of air bubbles in the prepared samples. These procedures were repeated for all the adopted concentration levels, i.e., 0.1, 0.5, 1 and 2%wt.

2.2. Sample Characterization

The nanomaterials used were purchased from US Research nanomaterials, Inc. The purity of these materials was greater than 99.5%. The nanoparticle sizes were confirmed through transmission electron microscope (TEM) characterization; see Figure 1, which is provided in the supplied material datasheet [29]. In addition, the dispersion of nanofillers in the prepared silicone rubber samples was examined using a JEOL field emission scanning electron microscope (FESEM). SEM images with a 0.5% wt concentration for the adopted nanofillers are shown in Figure 2. A good dispersion of nanoparticles in the silicone rubber nanocomposites samples can be observed for the adopted nanomaterials; however, some agglomerations are observed.



TiO₂

ZrO₂

SiO₂

Figure 1. TEM images of nanoparticles [29].



Figure 2. Cont.



Figure 2. SEM images of silicone rubber nanocomposite samples, (**A**) 0.5% SiO₂, (**B**) 0.5% TiO₂, (**C**) 0.5% ZrO₂.

3. Breakdown Strength Measurements

A breakdown voltage test was performed on the transformer oil nanofluid samples at room temperature (20 ± 5 °C) using a liquid dielectric test set according to ASTM D1816 standard. A schematic diagram of the experimental setup is shown in Figure 3. The breakdown voltage test was carried out using 500 mL of each prepared oil sample (at each concentration) using the standard test cell, also shown in Figure 3. The test cell used in the test had two mushroom-shaped (VDE) electrodes with a 2 mm spacing between them, complying with the adopted standard. During the test, the rate of voltage rise was kept constant at 500 V/s and the frequency of the applied voltage was 50 Hz. Each sample was tested 10 times using the same sample, with a 2 min time interval between each two consecutive breakdowns. The average breakdown strength and breakdown strength at 10% and 50% probabilities were estimated according to Weibull's statistical technique. An evaluation of breakdown strength based on 10% probability was conducted as it gives an

 Liquid Insulation

 Electrode

 High AC Voltage

 Image: Construction of the second s

indication about oil reliability, while the breakdown strength at 50% probability gives the average value using a statistical technique.

Figure 3. Experimental setup for breakdown strength of transformer oil samples.

The breakdown strength for silicone rubber samples was measured at room temperature (20 ± 5 °C) using the same test-cell configuration and the same rate of voltage rise. The schematic diagram of the test setup is shown in Figure 4. During the measurement of breakdown strength, the silicone rubber sample was inserted between the two electrodes through the liquid insulation (transformer oil) to prevent the occurrence of surface flashover. Each sample was tested 10 times (using a new specimen for each breakdown voltage test); average breakdown strength, as well as breakdown strength at 10% and 50% probabilities, was calculated using Weibull's statistical technique.



Figure 4. Experimental setup for breakdown strength of silicone rubber samples.

4. Experimental Results

In this section, the breakdown strengths of nanofilled transformer oil and silicone rubber nanocomposites are evaluated. The evaluation was carried out taking into account the effects of the nanoparticle dielectric constant and nanofiller concentration. The effect of the dielectric constant of nanofillers was studied using the three previously specified nanomaterials, which have different dielectric constants. These materials were SiO_2 (dielectric constant = 4.3), ZrO_2 (dielectric constant = 70) and TiO_2 (dielectric constant = 100). The evaluation was based on the average value of breakdown strength, as well as breakdown strength at 10% and 50% probabilities.

4.1. Breakdown Strength of Transformer Oil Samples

In this section, the breakdown strengths of nanofilled and pure transformer oil were evaluated in terms of average value and breakdown strength at 10% and 50% probabilities. Figure 5 shows the effect of the nanoparticle filler concentration level, as well as the nanoparticle dielectric constant on the average value of breakdown strength. The figure shows that as the nanofiller concentration level increases, the average breakdown strength increases up to an optimal concentration level; the average breakdown strength then decreases. The same behavior is observed for the three adopted nanomaterials. Moreover, the nanoparticle material type (nanomaterial dielectric constant) affects the optimal concentration level. The maximum percentage increase in breakdown strength is about 46% and was obtained by adding $0.14 \text{ g/L} \text{SiO}_2$ (dielectric constant = 4.3). However, adding ZrO₂ (dielectric constant = 70) to transformer oil at 0.08 g/L increases the maximum percentage increase in breakdown strength increases to 84%. Therefore, the increase in the dielectric constant of nanoparticles results in a higher percentage increase in breakdown strength of transformer oil nanofluids.



Figure 5. Average breakdown strength of transformer oil.

Figure 6 shows Weibull's probability curves for the breakdown strength of transformer oil containing TiO_2 , which gives the maximum percentage increase. It demonstrates that, by increasing the TiO_2 concentration level, the breakdown strength increases at all probabilities. Figures 7 and 8 show the effect of concentration level, as well as nanoparticle material types, on the breakdown strength at 10% and 50% probabilities. These figures demonstrate that the breakdown strength at 10% and 50% probabilities increases by increas-

ing the nanoparticle concentration level up to its optimal value, after which it decreases. Furthermore, the nanoparticle material type (dielectric constant) affects the breakdown strength of transformer oil. It can be seen that adding TiO_2 to transformer oil gives a maximum percentage increase in breakdown strength at 10% and 50% probabilities compared with the other two particle material types. This, therefore, validates the increase in the nanoparticle dielectric constant giving a higher increase in the breakdown strength of transformer oil nanofluids.



Figure 6. Weibull probability curves for breakdown strength of TiO_2 /nanofilled transformer oil.



Figure 7. Breakdown strength at 10% probability of transformer oil containing different types of nanoparticles (SiO₂, ZrO₂ and TiO₂).



Figure 8. Breakdown strength at 50% probability of transformer oil containing different types of nanoparticles (SiO₂, ZrO₂ and TiO₂).

4.2. Breakdown Strength of Silicone Rubber Samples

In this section, the breakdown strength of silicone rubber nanocomposites is evaluated. This evaluation was carried out using the same adopted nanoparticle material types, SiO_2 , ZrO_2 and TiO_2 , which were used with transformer oil, as previously mentioned. Figure 9 shows the effect of nanofiller types (dielectric constants) and filler loading on the breakdown strength of silicone rubber. It was found that the average breakdown strength increases with the increase in filler concentration to a certain level and then decreases by increasing the filler concentration. In addition, the nanoparticle dielectric constant significantly affects the breakdown strength of silicone rubber. The results demonstrate that the maximum percentage increase in average breakdown strength by adding SiO₂ (dielectric constant = 4.3) is about 30% at a filler concentration level of 1%wt. However, by adding ZrO₂ (dielectric constant = 70), the maximum percentage increase reaches 19.6% at the same filler concentration level. The maximum percentage increase in average breakdown strength is 16.5% with the addition of TiO₂ (dielectric constant = 100) at the same filler loading of 1%wt. Therefore, the nanofiller dielectric constant significantly affects the breakdown strength of solid nanocomposites. The increase in the nanoparticle dielectric constant results in a lower percentage increase in the breakdown strength of silicone rubber nanocomposites.

Figure 10 shows Weibull's probability curves for breakdown strength of silicone rubber nanocomposite samples containing SiO_2 , which produces the maximum percentage increase. The results show that adding SiO_2 to silicone rubber increases the breakdown strength at all probabilities. Figures 11 and 12 show the effect of nanoparticle material types (dielectric constant) on the breakdown strength of silicone rubber, considering 10% and 50% probabilities. It can be seen that the maximum increase in the breakdown strength at 10% and 50% probabilities is obtained by adding SiO_2 to silicone rubber compared with the other two nanofiller material types (ZrO₂ and TiO₂). This provides another verification of a higher dielectric constant of nanoparticle producing a lower percentage increase in the breakdown strength of solid nanocomposites.



Figure 9. Average breakdown strength of silicone rubber.



Figure 10. Weibull probability curves for breakdown strength of SiO₂/silicone rubber nanocomposites.



Figure 11. Breakdown strength at 10% probability of silicone rubber containing different types of nanoparticles (SiO₂, ZrO₂ and TiO₂).



Figure 12. Breakdown strength at 50% probability of silicone rubber containing different types of nanoparticles (SiO₂, ZrO_2 and TiO_2).

As previously mentioned, the nanoparticle dielectric constant affects the average breakdown strength of both transformer oil nanofluids and silicone rubber nanocomposites. The effect of the dielectric constant on the maximum percentage increase in the average breakdown strength of transformer oil nanofluids and silicone rubber nanocomposites is summarized in Figure 13 and Table 1. From this figure, it can be seen that the increase in nanoparticle dielectric constant results in a higher percentage increase in average breakdown strength of transformer oil nanofluids. In contrast, the increase in nanoparticle dielectric constant results in a lower percentage increase in the breakdown strength of silicone rubber nanocomposites. Therefore, the role of nanoparticle dielectric constants in improving breakdown strength in transformer oil nanofluids (liquid insulation) is significantly different compared with its role in silicone rubber nanocomposites (solid insulation). The interpretation of this point is discussed further in the next section.



Figure 13. Maximum percentage improvement of average breakdown strength (BS) of transformer oil and silicone rubber.

Table 1. Maximum breakdown strength and maximum percentage increase for transformer oil and silicone rubber.

Nanofiller	Maximum BS		Maximum Percentage Increase	
	Transformer Oil	Silicone Rubber	Transformer Oil	Silicone Rubber
SiO ₂	13.87 kV/mm	18.85 kV/mm	46%	30%
ZrO_2	16.42 kV/mm	17.34 kV/mm	72.8%	19.6%
TiO ₂	17.48 kV/mm	16.89 kV/mm	84%	16.5%

5. Discussion and Interpretations

Looking at the obtained breakdown results, the increase in nanofiller concentration level results in an increase in the breakdown strength for both nanofluids and solid nanocomposites until the optimum concentration. However, a reduction in breakdown strength occurs after the optimal concentration level in both nanofluids and solid nanocomposites. This reduction is due to agglomeration and electric field distortion, as reported in [9]. This is because when an external electric field is applied to a nanofluid, the electric field is increased at the nanoparticle/liquid interface. This behavior was validated through the calculation of the electric field distribution, using finite element analysis (FEM). Accordingly, a nanoparticle having a particle size of 10 nm was simulated as a spherical particle. The nanoparticle was simulated as being immersed in a dielectric liquid with a dielectric constant of 2.3, as shown schematically in Figure 14. An external electric field of 10 kV/mm was applied between the electrodes. The nanoparticle dielectric constant was taken to be 4.3, 70 and 100. The presence of the nanoparticle in the dielectric fluid increased the electric field intensity at the nanoparticle/liquid interface, as shown in Figure 15. The electric field at the interface increased with the increase in nanoparticle dielectric constant, as proven from the electric field calculation. This increase in electric field intensity results in the charge trapping of free charges resulting from ionization processes in the dielectric liquid. The increase in the dielectric constant of nanofillers, therefore, results in more improvement in the breakdown strength of nanofluids. However, when the nanoparticle traps the free charges, a distortion in the electric field occurs. The degree of distortion depends on the number of trapped charges. This was validated by using the simulated system in Figure 16, using FEM with a nanoparticle with a dielectric constant of 100. Mixed surface charges (positive and negative with a surface charge density of 10^{-5} C/m²) were also simulated for two cases, as shown in Figure 16. The first case (Case I) had an angle $\theta = 60^\circ$, whereas the second case (Case II) had an angle $\theta = 30^{\circ}$. The presence of surface charges on the nanoparticle surface resulted in a distorted electric field, as shown in Figure 17. This figure (Figure 17) illustrates that the increase in nanoparticle surface charges results in more electric field distortion. With the increase in the nanofiller concentration level, the number of nanoparticles increases. Therefore, the space charges increase, causing a severe electric field distortion. The severe electric field distortion can cause partial discharges at the nanoparticle/liquid interface that leads to breakdown at lower voltages. Furthermore, with the increase in nanofiller concentration level, the distance between the nanoparticles decreases. This can cause agglomerations due to the Coulomb's forces between them. These agglomerations result in lower charge trapping due to the lower resultant surface area, as the increase in particle size reduces its surface area per unit volume. Therefore, because of the agglomerations, the total surface area is reduced with the same filler loading. This effect reduces the capability of charge trapping, which in turns reduces the breakdown strength of nanofluids.

It was also found that the increase in the nanofiller dielectric constant results in a higher percentage increase in the breakdown strength of the nanofluid. However, a lower percentage increase was found to occur when using solid nanocomposites, as previously described in Figure 13. This behavior requires further discussion to achieve a deeper understanding of the role of nanofillers in improving the breakdown strength for both liquid and solid dielectrics. According to the published literature, the increase in breakdown strength considering nanofluids is a result of moisture binding [21] and the charge trapping process [22]. Moisture binding results from the hydrophilic nature of nanoparticles that reduces the spread of moisture in the fluid, leading, in turn, to an increase in breakdown strength. However, charge trapping reduces the spread of free charges in the fluid; therefore, increased breakdown strength is achieved. On the other hand, the increase in breakdown strength in solid nanocomposites mainly results from the charge trapping process, as reported in the published literature [24–27]. Researchers, therefore, attribute the improvement in breakdown strength of both liquid and solid dielectrics to the charge-trapping action of nanoparticles. From the obtained results, the percentage increase in breakdown strength increases with the increase in the dielectric constant of nanofillers with liquid dielectrics; however, it decreased with the increase in the nanofiller dielectric constant with solid dielectrics. In our opinion, this behavior is achieved due to the effect of the nanofiller dielectric constant on the formation of surface charges of nanoparticles, as illustrated in the following subsections.



Figure 14. Simulated nanoparticle in liquid dielectric.



Figure 15. Electric field distribution with no surface charges on nanoparticles having different dielectric constants.



Figure 16. Simulated nanoparticle with mixed charges in liquid dielectric.



Figure 17. Electric field distortion considering accumulation of surface charges on nanoparticle surface (Nanoparticle material dielectric constant = 100).

5.1. Surface Charge Formation on Nanoparticle Surface

When an external electric field is applied to a nanodielectric, either liquid or solid, electric charges on the nanoparticle surface start to accumulate on the surface, as mentioned above. This results from the effect of the polarization of nanoparticle that makes the negative charges in the nanoparticle accumulate on the side facing the positive electrode. However, the positive charges in the nanoparticle accumulate on the opposite particle side, i.e., the side facing the negative electrode. As the external electric field is applied to the dielectric, whether liquid or solid, nanoparticles start to trap free electrons or the positive ions resulting from the ionization process of the dielectric (liquid or solid). The trapping process turns the nanoparticle into a completely negatively or positively charged particle depending on its dielectric constant. Hence, nanoparticles with low dielectric constants become negatively charged; however, nanoparticles with a high dielectric constant become positively charged. This behavior was previously illustrated in [4]. Nanoparticles having high dielectric constants are, therefore, attracted to the negative electrode when a high DC voltage is applied to nanofluids. However, nanoparticles having low dielectric constants are attracted to the negative electrode when the nanofluids are exposed to a high DC voltage. This means that:

- Charges accumulate on nanoparticle surfaces when exposed to an external electric field.
- Charged nanoparticles can move within the fluid due to the effect of an external electric field.
- However, nanoparticles cannot move within a solid dielectric due to the nature of bonding between the particle and the solid material matrix.

Therefore, the difference between nanofluids and solid nanocomposites is the presence of nanoparticle motion in nanofluids; however, the nanoparticle is stationary in solid nanocomposites, as illustrated schematically in Figure 18.





Figure 18. Behavior of nanoparticle inside solid nanocomposites and nanofluids.

5.2. The Role of the Nanofiller Dielectric Constant in Improving Breakdown Strength of Liquid and Solid Dielectrics

As mentioned earlier, surface charges accumulate on nanoparticles surfaces when the nanodielectric, whether liquid or solid, is exposed to an external electric field. The accumulation of surface charges, called trapping, is the major reason behind the improvement in breakdown strength of both liquid and solid dielectrics. Hence, the charge-trapping process reduces the energy of free charges generated by ionization, which leads to a higher breakdown strength. The increase in the dielectric constant of nanofillers results in a higher increase in the breakdown strength of nanofluids. As the increase in the nanofiller dielectric constant results in more surface charges on the nanoparticle surface, this latter increase in surface charge results in a higher attraction force with the electrodes. This results in a higher nanoparticle speed and, therefore, a greater reduction in the kinematic energy, as well as the momentum, of free charges in liquid dielectrics. This leads to a higher nanofluid breakdown strength with the increase in nanoparticle dielectric constant. However, the increase in the nanoparticle dielectric constant leads to higher electric field strength at the nanoparticle/solid interface due to the formed surface charges on nanoparticle surface. This increase in electric field strength allows local discharges at the nanoparticle/solid interface and, therefore, a lower percentage improvement in its breakdown strength. This means that using nanoparticles with high dielectric constants is recommended with nanofluids in order to obtain higher percentage increases in their breakdown strength. However, nanoparticles with lower dielectric constants are recommended with solid nanocomposites to increase their breakdown strength.

6. Conclusions

The role of the nanoparticle dielectric constant on the surface charge of nanoparticles and the breakdown strength of nanofluids and solid nanocomposites were studied. The study was conducted by preparing nanofluid and silicone rubber nanocomposite samples at different nanofiller concentration levels, using three types of nanoparticle materials. The breakdown strength of all prepared samples, both liquid and solid, was measured and the following conclusions can be drawn:

- As the nanofiller concentration increases, the average breakdown strength increases above pure dielectric up to a maximum value and then decreases at higher concentrations for both nanofluids and silicone rubber nanocomposites.
- The percentage increase in breakdown strength increases with the increase in the • dielectric constant of nanofillers with liquid dielectrics; however, it decreases with the increase in the nanofillers dielectric constant with solid dielectrics.
- The increase in the nanofiller dielectric constant results in an increase in the accumulated charges on a nanoparticle surface.

 It is recommended that nanoparticles with higher dielectric constants be used to increase the breakdown strength of nanofluids. However, lower nanoparticle dielectric constants are preferred to increase the breakdown strength of solid nanocomposites.

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