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Electrical properties evaluation of double-layer nano-filled oil-paper composites

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Abstract
Oil-paper composite is the main insulation system in oil-filled transformers. In recent years, the use of nanotechnology to improve the electrical and thermal properties of transformer oil has become a field of research interest. In this paper, the electrical properties such as breakdown strength, volume resistivity, and dielectric response of nano-filled oil-paper insulation are evaluated. The evaluation is carried out on a double-layer of oil-paper composites insulation. The double-layer samples are prepared considering different nano-filler types. The studied nano-filler types are Pb3O4, Al2O3, and SiO2 with average particle size 25 nm of each. For each nano-filler type, oil-paper samples with different concentrations (0.04, 0.06, 0.1, 0.2, and 0.4 g/L) are prepared. The effect of nano-filler types on the breakdown strength, volume resistivity, and dielectric response of oil-paper composites are highly evaluated. The results show that the filler types as well as filler permittivity affects the breakdown strength, volume resistivity, and dielectric response of oil-paper composites. Also, the results show that nano-filler with high permittivity gives higher improvement in breakdown strength of Oil-paper composite compared nano-filler with low permittivity. Finally, the interpretation of all obtained results considering breakdown strength, volume resistivity, and dielectric response is highly reported.

1 | INTRODUCTION

Oil-filled transformers are commonly used in transmission and distribution systems due to their high cooling efficiency. The insulation systems in these transformers consist of cellulose paper impregnated in oil (oil-paper composites). Therefore, the condition of the oil-paper composites system affects the transformer lifetime [1]. Furthermore, it is difficult to achieve the demands of small size using traditional oil-paper composite with increasing transmission voltage levels [2]. Using nanotechnology, the development of new composite materials with good electrical and thermal properties can be generated.

In [3–14], the nanotechnology has been utilized to improve the electrical properties of transformer oil. They showed that the nano-filled transformer oil has breakdown strength higher than base transformer oil. The transformer oil has improved by adding a small amount of nanoparticles. The presence of paper insulation in nano-filled transformer oil can affect the dielectric properties of the composites insulation system. Therefore, in order to simulate the real field, the evaluation of the nano-filled oil-paper composites system should be carried out.

Limited studies as in [2,15,16] have performed on nano-filled oil-paper composites. In [2], different aspects are evaluated such as the effect of temperature and moisture content on dielectric response. Also, the study included the partial discharge inception voltage of base and TiO2 nanoparticles modified oil-paper composites. In [15], the space charge, as well as the field distortion, is reduced by adding TiO2 nanoparticles to oil-paper composites. Also, in [16], the space charge behaviour in oil-paper composites modified by different concentration levels of TiO2 when subjected to different electric fields. The effect of TiO2 nanoparticles on dielectric response and charge trapping characteristics of oil-paper composites is studied in [17]. Therefore, most of the studies are limited to evaluation of only one type of nano-filler for improving oil-paper insulation system. Until now, the impact of nano-filler types as well as
nano-filler permittivity on the electrical properties of oil-paper composites is not considered. Furthermore, important properties such as breakdown strength and volume resistivity of oil-paper composites insulation is not highlighted. Therefore, it is important to study the effect of nano-filler types on the breakdown strength, volume resistivity and dielectric response of oil-paper insulation system. Accordingly, three nano-filler types with a wide range of permittivity ($\text{Al}_2\text{O}_3$, $\text{Pb}_3\text{O}_4$, and $\text{SiO}_2$) are selected in this study. This is to study the impact of relative permittivity on the electrical properties of oil-paper composites.

In this paper, the effect of nano-filler types and filler concentration levels on the breakdown strength of nano-filled oil-paper composites are studied. Also, the evaluation of volume resistivity and dielectric response of nano-filled oil-paper composites are carried out. These evaluations are performed on the double-layer of oil-paper composites samples modified by nanoparticles. The types of nanoparticles are $\text{Pb}_3\text{O}_4$, $\text{Al}_2\text{O}_3$, and $\text{SiO}_2$ with the same average particle size of 25 nm. The obtained results are highly discussed. Finally, this study gives some recommendations about the selection rules of nanoparticle material types for improving oil-paper insulation systems.

2 | EXPERIMENTAL WORK

The preparation procedures of the nano-filled oil-paper composite samples are carried out considering different material types. The breakdown voltage test of the oil-paper insulation samples is presented. The measurement method of volume resistivity and dielectric response (dielectric constant and $\tan \delta$) of the oil-paper composites are introduced.

2.1 | Preparation of nano-filled oil-Paper composite sample

First, in order to decrease moisture content, the cellulose paper as well as transformer oil are dried in oven at 40 $^\circ$C for 48 h. Figure 1 shows the preparation procedures of the nano-filled oil-paper composites sample. A weighted nanoparticles is inserted in a half-litre of Nynas mineral oil and stirred 30 min. In order to get homogenous dispersion of the nanoparticles in the oil, the mixture is subjected to an ultrasonic wave using an ultrasonic homogenizer 30 min. Now, the name of this mixture is a nano-filled oil sample. The double layers of circular cellulose papers (40 mm in diameter) with thicknesses of 0.1 mm of each are inserted in the nano-filled oil mixture. Finally, this nano-filled oil-paper composites sample is left in a vacuum chamber 24 h to extract any generated bubbles [17]. In this study, different nano-filled oil-paper composites samples are prepared considering different nano-filler types such as $\text{Pb}_3\text{O}_4$, $\text{Al}_2\text{O}_3$, and $\text{SiO}_2$ with the same average particle size of 25 nm. The adopted nanoparticles are spherical in shape. $\text{Pb}_3\text{O}_4$ and $\text{Al}_2\text{O}_3$ nano-fillers are obtained from SUVCHEM laboratory chemicals. However, the $\text{SiO}_2$ is obtained from SIGMA-ALDRICH Company. Different samples with different nano-fillers concentration levels are prepared. These concentrations are 0.04, 0.06, 0.1, 0.2, and 0.4 g/L.

2.2 | AC breakdown strength test

The AC breakdown strength is measured to evaluate the quality of oil-paper composites insulation. Usually, the average breakdown strength values are evaluated to determine the quality of an insulating liquid. However, the power transformers are designed according to the minimum withstand voltage level of the insulation, rather than the average breakdown values. Weibull statistical techniques can be applied to evaluate the breakdown strength that can be obtained from breakdown data for lower probabilities [5]. Therefore, in this study, the evaluation of nano-filled oil-paper composites is carried out considering average breakdown strength as well as the breakdown at 10% and 50 % probabilities considering Weibull statistical technique. The AC breakdown strength test for all prepared nano-filled oil-paper composites samples is carried out considering ASTM standard. The sample of double-layer of the cellulose papers that impregnated in nano-filled oil is attached between two electrodes as declared in Figure 2. The selected voltage rate of rise is 500 V/s at 50 Hz. Ten times of breakdowns are performed and recorded for each sample. The breakdown strength test is carried out at room temperature (average temperature is 24 $^\circ$C).
2.3 Volume resistivity test

The volume resistivity of the oil-paper composites is a property that determines how the material can resist the electric current in the internal volume of the composites. In this study, the volumetric resistance is measured using Megohmmeter. The double layers of each sample are extracted from the nano-filled oil and then directly measured considering a three-electrode test cell is used. The three electrodes are made of copper and they have smooth surfaces. The measurement of volumetric resistance of all samples is carried out by applying a 500 V with time duration of 10 min. The test is performed at room temperature (average temperature is 24°C). Figure 3 shows a schematic diagram of the volume resistivity test for the double-layer oil-paper composites. Finally, based on the sample dimensioned and the measured volumetric resistance, the volume resistivity is calculated using Equation (1).

\[
\rho = \frac{R \times a}{\tau}
\]

where, \(\rho\) is the volume resistivity in G\(\Omega\) m, \(R\) is the measured volumetric resistance in G\(\Omega\), \(a\) is the current measuring electrode cross section area in m\(^2\) (see Figure 3), and \(\tau\) is the sample thickness in m. The dimension of adopted electrode and sample thickness is declared in Figure 3.

2.4 Dielectric responses test

The dielectric response is a technique that is used in insulation diagnosis and first presented in the 1980s [2]. The relative permittivity indicates how easily the oil-paper composites can become polarized under the application of the electric field. However, the dissipation factor indicates the presence of contamination or deterioration in the oil-paper composites. In this study, the double layers sample are extracted from the nano-filled oil and directly measured using an accurate LCR meter.

3 RESULTS AND DISCUSSIONS

In this section, evaluating the electrical properties of nano-filled oil-paper composites samples is presented. The evaluation is carried out considering breakdown strength, volume resistivity, and dielectric response. The effect of concentration levels of nano-fillers and nano-filler material types is presented. Finally, the discussions of the obtained results are presented.

3.1 Breakdown strength

Evaluating the breakdown strength of nano-filled oil-paper composites is carried out considering the double-layer samples. The average breakdown strength as well as breakdown strength at 10%, and 50% probabilities are evaluated. Figure 4 shows the average breakdown strength of nano-filled oil-paper composites insulation at different concentrations of Pb\(_3\)O\(_4\), Al\(_2\)O\(_3\), and SiO\(_2\). The average breakdown strength for the base oil-paper sample is 28.3 kV mm\(^{-1}\). However, the average breakdown strength of oil-paper composites modified by Pb\(_3\)O\(_4\), Al\(_2\)O\(_3\), and SiO\(_2\) is increased by increasing the concentration level to the maximum value and then decreased. The maximum breakdown strengths of nano-filled oil-paper composites considering Pb\(_3\)O\(_4\), Al\(_2\)O\(_3\), and SiO\(_2\) are 38.75, 37.75, and 36.8 kV mm\(^{-1}\), respectively. The maximum percentage increases in average breakdown strength of nano-filled oil-paper modified by Pb\(_3\)O\(_4\), Al\(_2\)O\(_3\), and SiO\(_2\) are 38.9%, 33.4%, and 30%, respectively. This means that the maximum percentage enhancement is 36.9% and obtained by adding 0.2 g/L of Pb\(_3\)O\(_4\).

Figures 5–7 show the Weibull distribution curves for oil-paper composites modified by Pb\(_3\)O\(_4\), Al\(_2\)O\(_3\), and SiO\(_2\). From these figures, it can be seen that the concentration levels, as well as the nano-filler types, affect the breakdown strength of the oil/kraft paper insulation system. The breakdown strength at 10% and 50% probabilities are obtained and reported in...
FIGURE 5  Weibull distribution curves for oil-paper modified by different concentrations of Pb$_3$O$_4$ nanoparticles

Tables 1 and 2, respectively. The results show that breakdown strength at 10% and 50% probabilities are improved by adding a small amount of Pb$_3$O$_4$, Al$_2$O$_3$, and SiO$_2$ nanoparticles to the oil-paper insulation system. Adding Pb$_3$O$_4$ to oil-paper gives high enhancement in breakdown voltage at 10% and 50% probabilities.

3.2 Volume resistivity

In this section, the effect of nano-filler types, as well as its concentration level on the volume resistivity of the double-layer of the oil-paper samples, is carried out. Figure 8 shows the volume resistivity results of oil-paper composites considering the three nanoparticle material types, Pb$_3$O$_4$, Al$_2$O$_3$, and SiO$_2$. From the figure, it can be seen that adding a low amount of nano-filler significantly affects the volume resistivity of the oil-paper composite system. The volume resistivity of the base oil-paper insulation sample is 2.15 GΩ m. The volume resistivity increases with increasing nanoparticle concentration level up to a maximum value of 3.34 and 3.05 GΩ m by adding 0.2 g/L of Pb$_3$O$_4$ and SiO$_2$, respectively. Then it decreases by increasing the nanoparticle concentration level. The maximum percentage increase of volume resistivity is 55.3% and 41.9% obtained by adding Pb$_3$O$_4$ and SiO$_2$, respectively. Therefore, the highest percentage increase in volume resistivity is obtained by adding 0.2 g/L of Pb$_3$O$_4$. Negative impact on volume resistivity of oil-paper composites is occurred by adding Al$_2$O$_3$ nanoparticles. The volume resistivity of oil-paper composites decreases with increasing Al$_2$O$_3$ nanoparticles.

TABLE 1  Breakdown strength of oil-paper at 10% probabilities

| Particle type | Base oil-paper | 0.04[g/L] | 0.06[g/L] | 0.1[g/L] | 0.2[g/L] | 0.4[g/L] |
|---------------|----------------|-----------|-----------|----------|----------|----------|
| Pb$_3$O$_4$   | 25             | 26.8      | 27.9      | 27.2     | 36.2     | 29       |
| Al$_2$O$_3$   | 25             | 28        | 27.5      | 28       | 34.7     | 28.7     |
| SiO$_2$       | 25             | 33.7      | 34.5      | 28.5     | 33       | 29.3     |
TABLE 2  Breakdown strength of oil-paper at 50% probabilities

| Particle type | Base oil-paper | 0.04 [g/L] | 0.06 [g/L] | 0.1 [g/L] | 0.2 [g/L] | 0.4 [g/L] |
|---------------|----------------|------------|------------|------------|------------|------------|
| Pb$_3$O$_4$   | 28.7           | 28         | 29.1       | 29.6       | 38.6       | 37         |
| Al$_2$O$_3$   | 28.7           | 29.1       | 30.5       | 30.9       | 38.8       | 32.3       |
| SiO$_2$       | 28.7           | 37.3       | 37         | 35.3       | 34         | 34.7       |

3.3  Dielectric response

The dielectric response of oil-paper composites is evaluated. Accordingly, both relative permittivity and dissipation factor of oil-paper composites are evaluated considering different concentrations of Pb$_3$O$_4$, Al$_2$O$_3$, and SiO$_2$. Figure 9 shows the effect of adding Pb$_3$O$_4$ on the relative permittivity of oil-paper composites considering different concentration levels. From this figure it can be seen that, adding Pb$_3$O$_4$ nano-filler can significantly increase the relative permittivity of the oil-paper composites. Figure 10 shows the effect of filler type on relative permittivity of oil-paper composites considering different concentration levels. The results show that the nano-filler types, as well as its concentration levels, have significantly affected the relative permittivity of oil-paper composites. The relative permittivity of the base oil-paper sample is 5.5. It is increased by increasing the nano-filler concentration level. The maximum relative permittivity of oil-paper composites modified by Pb$_3$O$_4$, Al$_2$O$_3$, and SiO$_2$ are 5.85, 5.73, and 5.6, respectively.

Figure 11 shows the dissipation factor of oil-paper composites modified by different concentration levels of Pb$_3$O$_4$ with the variation of the frequency. The results show that adding 0.2 g/L of Pb$_3$O$_4$ decreases the dissipation factor of oil-paper composites. Figure 12 shows the dissipation factor of oil-paper composites modified by different concentration levels of Pb$_3$O$_4$, Al$_2$O$_3$, and SiO$_2$. The results show that adding nanoparticles of Al$_2$O$_3$, and SiO$_2$ can significantly increase the dissipation factors of the oil-paper composites. However, the dissipation factors are slightly affected by adding Pb$_3$O$_4$ nanoparticles. It is decreased by increasing the concentration of Pb$_3$O$_4$, then slightly increased. Therefore, the dissipation factor is lower by adding Pb$_3$O$_4$ compared to the other two nano-fillers (Al$_2$O$_3$, and SiO$_2$).
4 | DISCUSSIONS

In this section, the interpretations of the effect of nano-filler types and concentration levels on the breakdown strength, volume resistivity, and dielectric response are presented.

4.1 | Breakdown strength

From the aforementioned results, adding a small amount of Pb$_3$O$_4$, Al$_2$O$_3$, and SiO$_2$ nanoparticles can significantly improve the breakdown strength of nano-filled oil-paper composites. The reasons for improving breakdown strength by adding nanoparticle is discussed as follows.

Based on the mechanism proposed in [14], adding nanoparticle to transformer oil can decrease the spread of moisture in oil as illustrated in Figure 13. This is due to the hydrophilic effect of nanoparticle that is adsorbed and collected water on its surface. This is reasonable for decreasing the spread of moisture in oil. As a result, the breakdown strength of transformer oil is increased by adding nanoparticles. Also, here adding hydrophobic nanoparticles to oil-paper composites decreases the spread of moisture content in the insulation system. As the moisture content decreases, the breakdown strength of oil-paper composites insulation system is increased.

Regarding the impact of three nano-filler types on the improvement of the breakdown strength, different percentages increase is attained. The maximum percentage increases in breakdown strength of nano-filled oil-paper composites are 36.9%, 33.4%, and 30% for Pb$_3$O$_4$, Al$_2$O$_3$, and SiO$_2$, respectively. The reasons for this point are related to the charge trapping process as discussed as follows. Based on the study reported in [13] that conducted on bulk nano-filled transformer oil, the percentage increase in breakdown strength is very high and significantly depends on the particle permittivity. They have shown that nanoparticle with higher permittivity can hold more surface charges compared to the particle with lower permittivity. More charges on the particle surface contribute to further free electrons that can be trapped. Furthermore, the electric field strength at nano-filler surface is significantly increases by increasing its permittivity. This point is validated using finite element in [8]. Accordingly, a force ($F$), see Equation 1, can attract more free electrons from surrounding oil. This called the charge trapping process. These results in a high percentage increase in breakdown strength of nano-filled oil.

\[ F = qE \]  

(1)

where, $q$ is the charge magnitude in Coulomb, and $E$ is the electric field strength in V/m.

In the case of the nano-filled oil-paper composites insulation system, there are micro-pores in the interface between the double layers as well as in each layer itself as shown in Figure 14. These micro-pores are filled with nano-filled transformer oil as declared in Figure 14. Under applying the electric field, the nanoparticles in the micro-pores are charged and therefore can trap the free electron. The particles with higher permittivity (Pb$_3$O$_4$) can hold more charge compared to the other two particle types (Al$_2$O$_3$, and SiO$_2$). Therefore, the percentage increase in breakdown strength of Pb$_3$O$_4$ (36.9%) is higher than the other particle types Al$_2$O$_3$ (33.4%) and SiO$_2$ (30%). The percentage increase in breakdown strength of nano-filled oil-paper composites is seemed low in the three particle types. However,
the percentage increase in breakdown strength of bulk nano-filled oil is high and reached 170% as reported in [13]. The reasons for this point come due to the presence of low nano-filled oil content in micro-pores, as well as less charge trapping process in the gap between electrodes, compared to the bulk nano-filled oil.

4.2 | Volume resistivity

Based on the obtained results, adding Pb₃O₄ and SiO₂ to the oil-paper composites increases the volume resistivity of oil-paper composites by 55.3% and 41.9%, respectively as declared in Figure 8. This is returned to the decreasing of moisture content in oil-paper composites by adding nanoparticles which have a hydrophilic effect. On the other hand, the volume resistivity of oil-paper composites is decreased by adding Al₂O₃. This comes due to the semi-conductive nature of Al₂O₃ nanoparticles that responsible for the reduction of volume resistivity. Therefore, due to this negative impact, it is not recommended to use Al₂O₃ in improving oil as well as oil-paper composites.

4.3 | Dielectric response

The obtained results show that nano-filled oil-paper composites have a higher relative permittivity compared to base oil-paper insulation considering Pb₃O₄, Al₂O₃ and SiO₂ nanoparticles as illustrated in Figure 9. Nanoparticles generated new electron–hole pairs in oil-paper composites. In nanoparticle, electrons can move from valence band to conduction band produces more electron-hole pairs. The hydrophilic nature of the adopted nanoparticles increases the absorbance of the moisture (water molecules) from the surrounding oil. The presence of ionic charges in water molecules increases the relative permittivity. Also, the results show that adding Pb₃O₄ nanoparticles increases the relative permittivity of the oil-paper insulation system compared to the other two nanoparticle material types (Al₂O₃ and SiO₂) as shown in Figure 9. This is due to the relative permittivity of the nanoparticle itself. The relative permittivity of Pb₃O₄ is higher than Al₂O₃ and SiO₂. The relative permittivity of Al₂O₃ nanoparticle is higher than SiO₂ nanoparticle. Nanoparticle with high permittivity such as Pb₃O₄ has more surface charge (electron–hole pairs) on its surface. Therefore, the relative permittivity of oil-paper insulation modified by Pb₃O₄ is higher than the insulation modified by the other two nanoparticle material types (Al₂O₃ and SiO₂).

5 | CONCLUSIONS

In this paper, the evaluation of electrical properties of nano-filled oil-paper composites has been carried out considering different three material types and concentration levels. The evaluation is carried out considering double layers of oil-paper composites. The breakdown strength, volume resistivity, the dielectric response is experimentally investigated. Based on the obtained results, the following points can be concluded:

1. The maximum percentage improvement of breakdown strength of nano-filled oil-paper is quite different by comparing the three filler material types. The improvement of oil-paper breakdown strength is quite higher by adding Pb₃O₄ nano-fillers compared to the other nano-filler materials (Al₂O₃ and SiO₂).

2. The maximum percentage improvement of volume resistivity of oil-paper composites is occurred by adding Pb₃O₄ nanoparticles as compared to the Al₂O₃ and SiO₂.

3. Adding Al₂O₃ to oil-paper composites is significantly decreased the volume resistivity of oil-paper composites. So, it is not recommended to use Al₂O₃ nanoparticle to improve the insulation system in oil-immersed power transformers.

4. The relative permittivity of the oil-paper insulation system is increased by adding the nanoparticles considering Pb₃O₄, Al₂O₃, and SiO₂. Adding Pb₃O₄ nanoparticles can increase the relative permittivity of oil-paper insulation compared to the other nanoparticle types.

5. The dissipation factor of nano-filled oil-paper composites modified by Pb₃O₄ is lower compared to the oil-paper composites modified by Al₂O₃ and SiO₂.

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