Enhanced voltage distribution and breakdown strength performances of oil-paper composite insulation by adding TiO₂ nanoparticles

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Abstract
Oil-paper composite insulation (OPCI) is used as the main insulation material in the converter transformer. However, the dielectric properties of insulating oil and oil-impregnated paper are markedly mismatched under direct current (DC) voltage, resulting in an unbalanced distribution of voltage, which is considered to be a major cause of large insulation margin but frequent failures. Therefore, it is necessary to adjust the voltage distribution in OPCI to enhance its breakdown strength. In this work, surface-treated TiO₂ nanoparticles were employed to modify OPCI. The polarisation current tests revealed that by changing the additive amount of nanoparticles, the polarisation intensities of both oil and oil-impregnated paper could be adjusted. Based on this, a model taking relaxation polarisations into full consideration was established to analyse the voltage distribution in OPCI and predict the optimal concentration. The prepared OPCI with the concentration of 0.02 g/L has desired balanced voltage distribution due to the influence of nanoparticles on interface charge, and therefore high breakdown strengths under both DC voltage and polarity reversal voltage, which can be increased by 22.6% at most. This work puts forward a novel idea to improve insulation property of OPCI and provides a reference for broader application of nanomaterial in power equipment.

1 | INTRODUCTION

Oil–paper composite insulation (named as OPCI), which consists of mineral oil and pressed paper or board made from cellulose, has been widely used for electric isolation in the transformer due to its excellent electrical and mechanical properties [1,2]. The insulating paper has to be impregnated with oil in practice to enhance its dielectric strength [3], so for the sake of simplicity, OPCI can be seen to be composed of layers of oil and oil-impregnated paper (named as OIP) [4].

In converter transformers, a direct current (DC) voltage is applied to OPCI at runtime [5,6]. To maintain the required potential difference, both oil and OIP have to endure part of the voltage. For multilayer composite material, once the distribution voltage in any layer reaches its breakdown strength, partial discharge or even breakdown happens, which is not allowed in electrical equipment [7]. Therefore, if the applied DC voltage is distributed proportionally according to the breakdown strength of each layer, the insulation property of material can be better utilised to enhance the breakdown strength of the whole system. It is known that a resistive field is established under DC voltage. The OIP’s resistivity can be 100 times as large as that of oil, hence the voltage in OIP is much larger than that in oil [8]. However, the DC breakdown strength of OIP is about 200 kV/mm, and just 7 times oil’s (approximately equal to 30 kV/mm) [7,9], so the DC voltage distribution in OPCI is not as balanced as expected.
Moreover, a unique but complex DC voltage, the polarity reversal voltage, which emerges with power flow reversal, often occurs in high voltage DC transmission system. The reversing external electric field is superimposed with the electric field formed by accumulated charge, resulting in a distorted transient voltage distribution [10]. As a result, the DC breakdown strength of OPCI is weakened and thus large margin is obliged in insulation design. In spite of that, there are still many insulation faults. Hence it is very significant to balance the voltage distribution and enhance the breakdown strength of OPCI under DC voltage.

There are many studies focusing on the property improvement of OPCI by nanoparticles [11–16]. It has been demonstrated that TiO$_2$ nanoparticles can not only enhance the breakdown voltages of transformer oil under both AC and lightning impulse voltage but also suppress the deterioration effects of moisture and ageing on insulating performance of oil [11–13]. Liao et al. [14] have pointed out that the AC breakdown strength of oil-impregnated insulation paper increased by 20.8% when the nano-TiO$_2$ content reached 3%. Chen et al. [15] added nano-Al$_2$O$_3$ into insulating pressboard, and the test results have shown that the conductivity and permittivity of nanocomposites initially declined and then rose with the increase of concentration. However, most of these studies just singly study either oil or OIP, only a few studies take them as a whole and focus on the effect of nanoparticles on multilayer OPCI. Under AC voltage, it was found that nanoparticles can improve the matching degree of the relative dielectric constant between oil and OIP, thereby generate uniform electric field distribution and increase the breakdown and flashover voltage of OPCI [14,16]. However, for DC voltage, the mechanism of nanoparticles’ influence is not clear, which greatly limits the application of nanoparticles in the converter transformer.

In this paper, the OPCI was modified by TiO$_2$ nanoparticles. We first tested the polarisation currents of nanocomposites with different concentrations. Then a model was put forward to analyse the transient behaviour of OPCI modified by nanoparticles. Based on the model, the voltage distribution in OPCI was studied, thus the DC breakdown voltage and the starting layer of breakdown were predicted, and the estimated results are in good agreement with the experimental results. The prepared OPCI with concentration of 0.02 g/L possesses improved voltage distribution and enhanced breakdown strength performances to better meet practical requirements, offering a possibility of reducing the size and increasing the reliability of converter transformer. Finally, the mechanisms of nanoparticles’ influence were discussed by analysing the interface charge of OPCI.

2 | EXPERIMENTAL SECTION

2.1 | Sample preparation

The base transformer oil (mineral oil, KI-25X) and cellulose paper (with thickness of 0.12 mm) were used in tests. TiO$_2$ nanoparticles were synthesised by solvothermal method [17]. Tetrabutyl titanate was used as titanium source and oleic acid was chosen to modify the surface of nanoparticles to suppress agglomeration. First, tetrabutyl titanate and oleic acid (the molar ratio was 1:24) were mixed into cyclohexane. Then, a small amount of ammonium bicarbonate (used as hydrolytic agent) and triethylamine (used as catalyst) were added to the mixture with vigorous stirring, and the reaction was maintained at 150°C for 24 h. Finally, the specified TiO$_2$ nanoparticles were obtained after washing products with absolute ethanol for several times.

The transformer oil modified by TiO$_2$ nanoparticles was manufactured by dispersing nanoparticles into the base oil by ultrasonic oscillation, and the TiO$_2$ nanofluids have been proved to possess good stability in our previous studies [12,13]. All oil samples were dried in vacuum at 85°C for 48 h to remove moisture. The papers were cut into pieces of 50 mm × 50 mm and dried in a ventilated oven at 105°C for 48 h, and then impregnated in the base oil and nanofluids, respectively at 100 Pa/85°C for 48 more hours. The double-layer OPCI consisted of one layer of oil and the other layer of the corresponding OIP, wherein the thickness of oil was set as 1 mm.

The preparation process and microstructure of nanocomposites are illustrated in Figure 1. As seen in Figure 1a, tetrabutyl titanate first reacts with water to form hydroxyl complexes, which then condensates to titanium dioxide nucleus. During this process, oleic acid is bonded to the titanium atom through chelation to adjust the morphology of nanoparticles [18]. Transmission electron microscope (TEM; FEI Tecnai G2 F30; USA) was used to observe the microstructure of nanoparticles, as can be seen, the diameter of nanoparticles was about 10 nm. Figure 1b is the preparation process of nanofluids, and the morphological observations of nanoparticles dispersed in oil were by another TEM (JEOL JEM-2010; Japan). The average diameter of nanoparticles in nanofluids is 10.3 nm, indicating that agglomeration hardly occurs. Meanwhile, the morphology of OIP was photographed by the scanning electron microscope (SEM; Hitachi S-4800; Japan), as shown in Figure 1c, a small amount of nanoparticles can be found inside the paper after nanofluid impregnation. Although the nanoparticles concentration of OIP is much lower than that of nanofluids, the OIP is still marked as the concentration of corresponding oil. For example, the OIP, which was impregnated in the nanofluids with a concentration of 0.1 g/L, is named as OIP-0.1 g/L; similarly, the corresponding OPCI is named as OPCI-0.1 g/L.

2.2 | Characterisation of dielectric performance

The polarisation currents of samples were measured with Keithley 6517B electrometer. That of OIP was tested with the three-electrodes structure (the diameter of test electrode was 30 mm) under a DC voltage of 600 V. Parallel plate electrodes with a diameter of 38 mm and distance of 1 mm were used for that of oil, and the applied voltage was 1 kV. The latter electrode set was also used to test the polarisation current of OPCI, but the electrodes were separated by 1.12 mm, as an
extra layer of corresponding OIP was inserted and appressed to the ground electrode. The polarisation currents of OPCI were measured under both positive and negative 1 kV voltages owing to the polarity effect of interface charge [5,10].

The positive and negative DC breakdown strengths of OPCI were obtained according to IEC 60243 with equal diameter (25 mm) electrodes (see Figure 2). The breakdown strengths of both oil (the thickness was 1 mm) and OIP (sandwiched between two electrodes) were also gained with the same electrode set. The boosting rate was 500 V/s, and at least ten effective breakdown results were collected for each experiment condition. Optical microscope (OLYMPUS CX23; Japan) was used to investigate the discharge tracks on the surface of OIP after the breakdown.

3 | RESULTS AND DISCUSSION

We have studied the breakdown strength of OPCI with different nanoparticles concentrations, as well as the cause. As seen in Figure 3, when the concentration is low (0.004 g/L), the breakdown voltage of OPCI is similar to that of OPCI without modification. With the increase of concentration, the breakdown strength reaches the maximum value when the concentration is 0.02 g/L, and the breakdown voltage of OPCI-0.01 g/L is very close to that of OPCI-0.02 g/L. Then the breakdown strength of OPCI decreases as the concentration rises (0.05 and 0.1 g/L). Meanwhile, these properties can also be divided into three categories from the viewpoint of the voltage distribution and breakdown mechanism. As a result, the typical
concentrations of 0.02 and 0.1 g/L were chosen for detailed illustration in the following, together with the base samples.

3.1 Polarisation current performance

When a step DC voltage is applied to dielectrics, the polarisation current $i$ through the dielectric decreases with time and finally reaches a constant value, wherein the attenuation part is the absorption current caused by relaxation polarisation and the constant part is known as the conduction current $I_R$ [19]. As shown in Figure 4, the initial value of polarisation current has a sharp enhancement with the addition of nanoparticles, while the steady-state value just increases a little.

Modelling of insulation system helps to understand the dielectric response, and the extended Debye model has been adopted for deriving the equivalent circuit of oil–paper insulation in most of the cases [2,20], as shown in Figure 5. The $C_s$ is the geometric capacitance of dielectric and the conduction current is expressed by resistance $R_s$ [2]. The series $R–C$ branches stand for different relaxation polarisation processes and can be determined by fitting the polarisation current with (1)–(3) [20],

$$i = I_R + \sum_{k=1}^{n} A_k e^{-t/\tau_k}$$  \hspace{1cm} (1)

$$A_k = U/R_k$$  \hspace{1cm} (2)

$$\tau_k = R_k \times C_k$$  \hspace{1cm} (3)

**FIGURE 2** Schematic diagram of electrodes and specimen placement in breakdown strength measurement

**FIGURE 3** DC breakdown voltages of OPCI with different nanoparticles concentrations

**FIGURE 4** Polarisation currents of oil and OIP with different nanoparticles concentrations (a) Oil, (b) OIP
where $U$ is the applied voltage, $R_k$ and $C_k$ are the resistance and capacitance values of branch $k$ respectively, $\tau_k$ is the relaxation time of branch $k$.

The equivalent circuit parameters of oil and OIP are separately given in Tables 1 and 2, where $C_s$ was measured by an IDAX 300 Insulation Diagnostic Analyzer; $R_s$ was calculated by the conduction current $I_c$ with (2); $R_k$ and $C_k$ were obtained by fitting the polarisation current with (1)–(3). As can be seen, for both oil and OIP, nanoparticles can lead to a slight decrease in $R_s$ while a little increase in $C_s$. On the contrary, nanoparticles can not only significantly intensify the relaxation polarisation intensity in nanofluids but also generate more polarisation processes, as expressed by the series $R$–$C$ branches [20]. The polarisation current of pure oil can be fitted by two $R$–$C$ branches, which are mainly attributed to the inevitable impurities, in contrast, four branches are needed for nano fluids, due to the interfacial polarisation of nanoparticles [21]. Even the addition amount is small, the surface area of the nanoparticle is significantly large and this surface effect increases with the decrease of diameter. The average diameter of TiO$_2$ nanoparticles in the nano fluids is merely 10.3 nm, hence the surface effect is remarkable and the total interface area is very large, which leads to an obvious interfacial polarisation process. As a result, the absorption current of nano fluids is increased exponentially relative to that of pure oil, as shown in Figure 4a. Compared with the effect of nanoparticles on oil the effect on OIP is similar but slight, because of the lower concentration of nanoparticles in OIP.

As shown in Tables 1 and 2, the nanoparticles have little effect on both $R_s$ and $C_s$. According to the resistive–capacitive model, which is widely accepted to analyse the interlayer polarisation in double-layer dielectric and only depends on $R_s$ and $C_s$ of each layer [6,10], the polarisation currents of OPCIs should be similar throughout the process. However, Figure 6 displays the measured polarisation currents of OPCIs (take OPCI-0 g/L and OPCI-0.02 g/L for instance). It can be seen that the initial value of polarisation current of OPCI-0.02 g/L increases markedly in contrast with that of OPCI-0 g/L, while the constant value just experiences a subtle enhancement. These differences between the test results and theoretical analysis indicate that the RC model may not be suitable for multilayer nanocomposites. This is because the RC model assumes that all layers in the insulation system are weak-polar dielectrics and ignores the relaxation polarisation of each layer. This assumption is reasonable for traditional oil–paper
insulation whose relaxation polarisation is quite slight. However, for the oil–paper insulation modified by nanoparticles, the interface polarisation by nanoparticles is very intense and thus cannot be ignored [21].

Considering the limitation of RC model, we put forward a double-layer extended Debye circuit model (see Figure 7), which is more credible to analyse the transient behaviour of OPCI modified by nanoparticles. The circuit parameters of Oil and OIP are determined by the data in Tables 1 and 2, respectively. It is necessary to adjust the capacitance and resistance values in Tables 1 and 2 to the same case when calculating the parameters in Figure 7, owing to the difference in the area of test electrodes.

### 3.2 Properties of OPCI under DC voltage

#### 3.2.1 Voltage distribution and breakdown strength

The voltage distribution in OPCI under ramp DC voltage is described with the proposed model (see Figure 7). When the positive DC voltage increases linearly at a rate of 500 V/s, the distribution voltages in both oil ($u_o$) and OIP ($u_p$) should satisfy (4) [22]

$$
\begin{align*}
C_o \frac{du_o}{dt} + \frac{u_o}{R_o} + \sum_{j=1}^{n} \frac{1}{R_{oj}} \left[ \frac{u_o}{R_{oj}} - \frac{e^{-t/(R_{oj}C_{oj})}}{R_{oj}^2 C_{oj}} \int u_o e^{t/(R_{oj}C_{oj})} dt \right] = i_o \\
C_p \frac{du_p}{dt} + \frac{u_p}{R_p} + \sum_{k=1}^{n} \frac{1}{R_{pk}} \left[ \frac{u_p}{R_{pk}} - \frac{e^{-t/(R_{pk}C_{pk})}}{R_{pk}^2 C_{pk}} \int u_p e^{t/(R_{pk}C_{pk})} dt \right] = i_p
\end{align*}
$$

where $C_o$ and $R_o$ are geometric capacitance and insulation resistance of oil, respectively; $C_p$ and $R_p$ are geometric capacitance and insulation resistance of OIP. $C_{oj}$ and $R_{oj}$ are parameters in the branch $j$ of oil; $C_{pk}$ and $R_{pk}$ are parameters in the branch $k$ of OIP. $u$ is the applied voltage. $i_o$ and $i_p$ are the total currents through oil and OIP, respectively.

Figure 8 shows the voltage distribution in OPCI till breakdown, and the measured breakdown strengths of OPCI are illustrated in Figure 9. The marked lines in Figure 8 mean that if the distribution voltage in any layer (oil or OIP) first reaches its own breakdown voltage (see Figure 9), entire breakdown of OPCI is supposed to take place immediately and the value of the applied voltage at this point is estimated as the breakdown voltage. As marked on the upper axis in Figure 8,
the predicted breakdown voltages of OPCI are 37.0, 42.4 and 23.7 kV, respectively with the increase of nanoparticles concentration. The OPCI-0.02 g/L, which shows a balanced voltage distribution, is predicted to possess high breakdown strength. Furthermore, as seen in Figure 8, the breakdown occurs in oil first for OPCI-0 g/L, while the insulation failures start at OIP for both OPCI-0.02 g/L and OPCI-0.1 g/L.

According to Figure 9, the breakdown voltage of OPCI varies with polarity, but it rises first and then undergoes a huge drop with the increase of nanoparticles concentration whatever the polarity is. As the concentration increases, the average positive/negative breakdown voltages of OPCIs are 36.9/39.0 kV, 45.2/40.4 kV and 25.7/25.1 kV respectively. When the polarity effect is neglected, the measured voltages are in good agreement with the estimated values.

Furthermore, the discharge tracks on the surface of OIP are presented in Figure 10. Three breakdown channels with obvious creeping discharge traces can be found for OIP in OPCI-0 g/L (see Figure 10a and Figure 10a-1, a-2, a-3), which is distinctly abnormal for the breakdown of solid dielectric itself. The most likely explanation is: the discharge channels occur first in oil, and then some channels develop along the surface of OIP rather than develop into the interior like others [23], remaining creeping discharge traces at the surface and multiple breakdown channels. Owing to the breakdown occurs first in OIP for OPCI-0.02 g/L, merely single breakdown channel can be observed and no creeping discharge occurs (see Figure 10b). The results for OPCI-0.1 g/L are the same as that of OPCI-0.02 g/L and thus not repeated here.

It can be seen that the breakdown voltage and initiating layer of breakdown are well predicted by the proposed model. Same experiments were applied to another double-layer OPCI, with the thickness of the oil gap being 0.5 mm. The predicted breakdown voltages are 29.5 and 34.3 kV for OPCI-0 g/L and OPCI-0.02 g/L, respectively in this case, and the corresponding measured positive/negative breakdown voltages are 29.0/31.4 kV and 37.8/35.3 kV, indicating that the proposed model is reasonable and effective. Owing to the limitation of length no more tautology is considered here.

### 3.2.2 Interface charge and polarity effect

During voltage boosting, interlayer polarisation occurs and charges accumulate at the oil–OIP interface, due to the discontinuities of both conduction current and absorption current [6,22]. The interface charge density $\sigma$ can be obtained by

$$\sigma = \frac{1}{5} \int_0^1 \left( \frac{\mu_o}{R_o} - \frac{\mu_o}{R_p} \right) + \left( \sum_{j=1}^m \frac{\mu_o}{R_{ij}} - \frac{e^{-\left( t/\tau_{ij} C_{ij} \right)}}{R_{ij}^2 C_{ij}} \int \mu_o e^{\left( t/\tau_{ij} C_{ij} \right)} dt \right) - \left( \sum_{k=1}^n \frac{\mu_o}{R_{pk}} - \frac{e^{-\left( t/\tau_{pk} C_{pk} \right)}}{R_{pk}^2 C_{pk}} \int \mu_o e^{\left( t/\tau_{pk} C_{pk} \right)} dt \right) dt$$

where $S$ is the electrode area.

As shown in Figure 11, the interface charge increases significantly with the addition of nanoparticles and its polarity is the same as that of applied voltage, i.e. the charges will weaken the electric field in oil and intensify the electric field in OIP. Thus, a lower distribution voltage in oil while a higher distribution voltage in OIP occurs with the increase in concentration, which agrees with the results in Figure 8. Therefore, the influence of nanoparticles on polarisation current further affects the interface charge is attributed to be the fundamental reason for the difference in voltage distribution, which determines where breakdown initiates as well as the breakdown voltage of OPCI.

There is the polarity effect in the breakdown voltage of OPCI, resulting in a slight difference from the predicted value, meanwhile, the influence of polarity effect varies with the concentration (see Figure 9). This is because the analysis of
voltage distribution and interface charge is based on the Maxwell–Wagner effect without considering any polarity effect [23], i.e. the same amount of negative interface charges should be also induced under negative applied voltage by this way. However, the polarisation currents under negative voltage are larger than that under positive voltage for OPCI in actual (see Figure 6), showing that more interface charges can be induced under negative voltage. Previous studies have indicated as well that extra negative charges can accumulate at the interface between oil and cellulose paper under either polarity [5,24]. Then the polarity effect of breakdown voltage can be consequently explained. For example, more accumulated negative interface charges weaken the electric field in oil under negative voltage. As a result, a higher negative voltage is required for breakdown than positive voltage, because the breakdown occurs first in oil for OPCI-0 g/L. On the contrary, the breakdown starts at OIP for OPCI-0.02 g/L, here a lower negative voltage is needed. Nevertheless, the polarity effect is not evident for OPCI-0.1 g/L, due to the gigantic amount of charges induced by Maxwell–Wagner effect (see Figure 11) which have basically determined the voltage distribution. It is obvious that the trapped charge accounts for a small proportion of total interface charge, so the analysis above of voltage distribution and interface charge of OPCI is still accurate though the polarity effect is neglected.

3.3 Voltage distribution under polarity reversal voltage

The effect of nanoparticles on the voltage distribution performance of OPCI under polarity reversal voltage further needs to be discussed. The previously proposed equivalent circuit is adopted again. The applied voltage starts at -10 kV and lasts long enough, thus the voltage distribution in OPCI reaches a steady-state, which is only determined by $R_p$. Then, the applied voltage gradually decreases and reverses to +10 kV within 60 s. After that, the +10 kV is applied for a long time as well to reach another steady-state. Since voltage distribution is less dependent on the trap charges, polarity effect is ignored again.

Figure 12 shows the voltage distribution in OPCI under polarity reversal voltage. After polarity reverse, the distribution voltages of oil in OPCI-0 g/L and that of OIP in OPCI-0.1 g/L reach a maximum of +12.7 and +10.7 kV, respectively, which are even higher than the amplitude of the applied voltage. In contrast, the voltage distribution in OPCI-0.02 g/L is very uniform without the appearance of distorted high voltage throughout the duration. Hence it can be expected that the obtained property enhanced OPCI (OPCI-0.02 g/L) does also have an improved voltage distribution performance to decrease the possibility of discharge under polarity reversal voltage.

4 CONCLUSION

In summary, we have shown that the voltage distribution and breakdown strength performances of OPCI could be improved by adding TiO$_2$ nanoparticles. In comparison with that of unmodified OPCI, the positive and negative DC breakdown voltages of OPCI-0.02 g/L were increased by 22.5 and 3.6%, respectively, as where breakdown started changed from oil to OIP. A model was proposed for predicting and analysing these performances of OPCI. Based on the model, the results were ascribed to the influence of nanoparticles on interface charge, and the negatively charged cellulose was considered to be the root of the polarity effect. Furthermore, the OPCI of 0.02 g/L was expected to have an enhanced breakdown strength under polarity reversal voltage as well. Our research has revealed the mechanisms of how nanoparticles affect voltage distribution and electric breakdown in OPCI, providing a reference for the practical application of
nanomaterial in converter transformer improvement, but the long-term performance needs further study.

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