Impact of adding activated bentonite to thermally aged ester-based TiO$_2$ nanofluids on insulation performance

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Abstract
The authors report the key findings from an experimental study that explored the use of activated bentonite for the reclamation of thermally aged ester-based transformer nanofluids to improve their insulation performance. Bentonite activated with acid treatment caused an increase in the specific surface area and pore volume of bentonite compared to the bentonite sample before treatment, thus imparting an improved adsorption capability. Physico-chemical diagnostic studies were carried out to characterise the activated bentonite. The insulation performance of the reclaimed natural ester and nano-filled ester fluid samples was assessed by measuring the corona inception voltage and breakdown voltage of each fluid sample, apart from measuring the flow electrification current using the spinning disk method. The results revealed that the reclamation process improved the corona inception voltage, dissipation factor and the breakdown voltage of the base ester fluid sample due to attraction of carbon particles to activated bentonite, but no significant variation was observed with nanofluids due to the depletion of the electrical double layer. The flow electrification current of ester and ester nanofluids reduced after treatment with activated bentonite, may be attributed to the interaction between copper and bentonite that alters the double layer formation responsible for the separation of charges.

1 INTRODUCTION

Transformers are major components in the power system network. Most of the medium and high voltage transformers are insulated using a liquid insulant (oil), which also acts as a coolant and paper/pressboard insulant (solid insulation), and used as a spacer and winding insulation material. The most widely used liquid insulation is of petroleum origin, but there is now considerable interest on the ester fluid which as an alternative dielectric liquid. Ester fluid has advantages over mineral oil due to its high flash point, high fire point and bio-degradability [1].

Recently, the use of nanoparticles to improve the insulation performance of natural esters has been the focus of much research [2-5]. Natural esters have the benefit of increased electrical breakdown voltage and retention of heat transfer property [6]. Different types of nanoparticles were considered for the study of nanofluids including conductive, semi-conductive and insulating nanoparticles [7, 8]. Titanium oxide (TiO$_2$) nanoparticles have frequently been used with esters and their positive effect on dielectric properties were demonstrated [2-5]. For example, such nanoparticle addition was found to result in an increase in the impulse breakdown voltage, which was attributed to a reduction in the stremmer velocity due to an increased relaxation time [2]. Other researchers investigated the effects of nanoparticles on the breakdown characteristics of oil by focusing on the differences in streamer patterns between pure oil and nanofluid [9].

The liquid insulant in transformers undergoes multiple stresses that contribute to its degradation. These are mainly due to electrical, thermal and chemical reactions such as oxidation, formation of hydrolysis and acidic derivatives which reduce lifetime [10]. The sludge produced as a result of such reactions also reduces the heat transfer property of the oil and increases the operating temperature of the transformer. The reclamation of degraded oil is usually carried out using adsorbents, such as activated carbon, Fuller’s earth, bentonite and alumina, to restore breakdown and dielectric properties [11]. Bentonite is mainly a clay mineral particle consisting of one aluminium octahedral sheet sandwiched between two silica octahedral sheets. The classification of bentonites is based on their chemical makeup, crystallographic properties and particle size.
factors including ion exchange property, swelling index and pH. Typically, bentonite mineral crystals are negatively charged according to the electrical double layer theory, and sodium bentonite is used in all applications due to its higher adsorption compared to calcium bentonite. Recently, sulphuric acid with different concentrations was used for the activation of bentonite clays to increase the adsorption capability by exchanging the metallic ions in its outer layer with hydroxyl ions [12]. The bleaching property of bentonite can be exploited for the reclamation of aged ester fluids to improve breakdown strength, flash point and viscosity.

Another major concern with power transformers is static electrification which may lead to unwanted electrostatic discharges. The use of insulating oils involves charge separation at the interface between two dissimilar materials forming an electrical double layer and the spinning disc system is one of the techniques adopted in flow electrification studies [13, 14]. Important factors influencing the inducement of static charges include temperature, moisture, viscosity and flow velocity. Natural ester involves a larger quantity of static charges when compared with mineral oil [15]. Research on nanofluids used for improving the dielectric strength of esters and the study of their flow electrification tendency are still at an early stage and the in-service performance of such nanofluids is yet to be fully demonstrated on operational transformers.

Here, the authors present a study on the effect of TiO$_2$ nanoparticles on the insulation characteristics of ester fluid with added cetyl trimethyl ammonium bromide (CTAB) as a surfactant, to help achieve a more uniform dispersion of nanoparticles and high stability. Furthermore, the impact of thermal and impulsive stresses was investigated to explore the physiochemical change of the nanofluid and correlate it with the electrification current. This was done to help clarify the issues relating to the charging tendency of ester. The influence of bentonite on flow electrification of ester fluid is also examined. In particular, the ability of bentonite to recover the dielectric properties of aged ester-based nanofluids is analysed, which forms an important part of the study.

Considering the above aspects of the reclamation process of ester-based nanofluids, the following studies were carried out in the present work: (i) characterisation of bentonite through elemental analysis, (ii) measurement of thermal and chemical changes on the bentonite after modification, (iii) corona inception and breakdown voltage tests of ester-based fluids before and after reclamation, and (iv) tests to determine the effect of thermal ageing and the impulse stress of ester-based fluid on its charging current before and after reclamation.

2 | EXPERIMENTAL STUDIES

2.1 | Material preparation

2.1.1 | Surface modification of bentonite

Bentonite occurs in its natural form as a montmorillonite (ash mineral). It is mainly used for bleaching of oils and fats. Bentonite (AlZ03.4SiO2.H2O) or hydrated aluminium silicate contains minerals such as potassium, calcium and sodium depending on the source. In the present study, sodium bentonite was used which offers a higher adsorption capability when compared with calcium bentonite [16]. The bentonite sample (50 g) was first dried in vacuum at 110°C and 20 g of the sample was treated with sulphuric acid (H$_2$SO$_4$) using a wet chemical method. The sample was mixed with 400 ml of H$_2$SO$_4$ (7 N) at 80°C for 6 h to improve its adsorption capability [17]. After the completion of acid treatment, the suspended bentonite powder was filtered under vacuum, and then the precipitate was washed with distilled water until it was free from SO$_4^{2−}$ ions. Such condition was confirmed by the formation of a white precipitate on addition of BaCl$_2$ solution to the acid-treated samples. Finally, the activated bentonite was dried at 110°C for 4 h and stored in a closed vessel.

2.1.2 | Characterisation of bentonite

X-ray diffraction (XRD) pattern of the samples was studied using a Bruker D8 diffractometer with Cu-Kα radiation with a wavelength of 1.5425 Å at a scan rate of 9º/min. Rietveld refinement was adopted to analyse the XRD data using PANalyticalX’pert High score plus software package. The nitrogen adsorption–desorption measurements were carried out at 77 K using an accelerated surface area and porosimeter system (ASAP-2020, Micromeritics). The specific surface areas (S$_{BET}$) were determined based on Brunauer-Emmett-Teller (BET) model and their pore size distributions were obtained from the desorption branch of the isotherm using Barrett–Joyner–Halenda (BJH) procedure. The thermal analysis of bentonite and activated bentonite was carried using an SDT Q600 thermal analyser under a nitrogen ambiene with a heating rate of 5°C/min. The flow rate of the inert gas was maintained at 100 ml/min and the crucible was made of alumina. The Fourier transform Infrared (FTIR) spectroscopy analysis was performed using the Agilent Cary 630 FTIR spectrometer and its vibrational functional groups were analysed.

2.2 | Preparation of ester-based nanofluids: ageing and reclamation process

Natural ester (MIDEL 1215) was used as the base fluid in the present study. This was mixed with TiO$_2$ (procured from Nanostructured and Amorphous Materials Inc., 99.8 % purity, 5 nm) together with CTAB as a surfactant. The adopted concentrations of TiO$_2$ nanoparticle and CTAB surfactant used were 0.05 g/L and 0.001 g/L, respectively, based on the optimal performance of inception voltage observed from our previous studies [18]. In order to remove moisture, the TiO$_2$ sample was placed in an oven for 8 h at 150°C. The base fluid was mixed with the surfactant using a magnetic stirrer for 30 m after which the nanoparticle quantity was added. This colloidal solution was sonicated for 180 m using Sonics Vibra-cell
sonicator (500 W, 20 kHz) at 40°C. After completion of this process, the nano-filled ester fluid was left for 2 h in a vacuum desiccator to remove micro-bubbles formed during the ultrasonication process. The types of samples used in the present study are described in Table 1.

The pressboard material with a thickness of 2 mm was preheated at a temperature of 110°C for 4 h in order to remove the moisture associated with the solid insulation. After the completion of this process, the dried pressboard insulation was wrapped into a copper sheet and then immersed into a beaker containing 1 L of ester-based nanofluids. The weight ratio of pressboard, copper and ester fluids were maintained at 1:1:10. Further, the thermal ageing was carried out at 140°C in a temperature-controlled thermostat under air medium. Based on the thermal class of fluid [19], the ageing of each fluid/pressboard sample was carried out in a convection oven with its normal internal air circulation for 50 days. Furthermore, the impulse stress was applied using negative lightning with an amplitude of 60 kV, which was applied to each test sample 120 times. The reclamation process of aged ester nanofluids is shown in Figure 1. A quantity of 5 g of activated bentonite was dissolved in 500 ml of aged fluid and then mixed using a magnetic stirrer at 800 rpm while heating the fluid to 80°C. The resultant mixture was filtered through Whatman filter paper (diameter 12.5 cm and pore size 11 µm). This particular pore size was selected in order to filter out micro carbonized particles and pressboard material which are responsible for ageing of the fluid. Figure 2 shows the filter paper before and after the filtration process where the carbon particles adsorbed on the activated bentonite powder are evident.

**TABLE 1** Types of samples used for test and analysis

| Sample | Insulating liquid          |
|--------|---------------------------|
| A      | Ester                     |
| B      | Ester + TiO₂              |
| C      | Ester + TiO₂ + CTAB       |

**FIGURE 1** Steps involved in the reclamation of ester-based nanofluids

2.3 | High-voltage experimental set-up

The experimental setup used for generating corona and for breakdown studies is shown in Figure 3. The test arrangement includes a high voltage source, a test electrode and a UHF sensor. The UHF plate sensors were used where the active coupler face contains a broadband sensing element with a diameter of 175 mm [20]. The sensitivity plot of the UHF sensor across its frequency range of 200 MHz to 2 GHz is shown in Figure 4 [21]. The AC voltage was generated using a test transformer (100 kV, 5 kVA), which on addition to a voltage doubler circuit generates DC voltage.

Measurement of these two voltage energizations was made using a capacitance and resistance divider. The applied voltage was increased at a constant rate of 300 V/s. The standard lightning impulse voltage (1.2/50 µs) was generated with a single stage impulse generator and measured using a capacitance divider. The test cell consisting of a needle-plane electrode configuration was used for both corona and breakdown studies. The needle and the grounded plane electrode were maintained at a distance of 2.5 mm. The radius of curvature of the needle tip connected to the high voltage was 50 µm and the plane electrode used was 5 cm in diameter. After five breakdowns, the needle and liquid being used for the test were removed and then replaced with new ones.

2.4 | Flow electrification

The streaming electrification of the nanofluid was carried out using a spinning disk system housed in a cylindrical aluminium vessel of 85 mm diameter, 100 mm height where the disk is covered on both sides by cellulose material as shown in Figure 5. The aluminium disc has a diameter of 40 mm and a thickness of 6 mm. As the disk rotates, centrifugal forces cause double layer charge formation on the surface of the disc and the charged fluid moves towards the inner walls of the measuring vessel causing a current flow. The spinning system as well as the electrometer were enclosed in a Faraday cage to
prevent the stray currents. The rotation of the disc was varied between 0 to 600 rpm by a motor equipped with a speed regulator.

2.5 | Dielectric response measurement

The dielectric response measurements were performed using OMICRON-DIRANA unit which generates a sinusoidal signal of desired frequency. A three terminal based test cell with voltage, measuring and guard electrode was used for the oil samples. The heating system was provided along with the test cell and the temperature equilibrium was maintained during the measurement period.

![FIGURE 2](image) Whatman filter paper (a) before filtering and (b) after filtering

![FIGURE 3](image) High-voltage experimental setup

![FIGURE 4](image) Sensitivity plot of UHF sensor

![FIGURE 5](image) Spinning disk system used for studying electrification of insulating liquid

3 | RESULTS AND DISCUSSIONS

3.1 | Material characterisation

3.1.1 | XRD analysis

The XRD patterns of bentonite and acid-activated bentonite are shown in Figure 6. The major mineral constituents that have been observed are montmorillonite (M), gypsum (G), quartz (Q) and calcite (C). Bleaching of the bentonite sample with sulphuric acid (7 N) did not show any major variation in mineral content except calcite [17]. There was a slight reduction in the intensity of montmorillonite indicating its partial attack due to acid treatment. The calcite peaks disappeared after acid treatment which could be due to the replacement of exchangeable cations of the montmorillonite lattice with hydrogen ions, leading to a significant reduction of calcium, and it is shown later in Table 3 where the elemental compositions of the bentonite samples are compared. The peaks of gypsum and quartz were relatively unchanged after activation indicating their resistance to acid.

3.1.2 | Brunauer-Emmet-Teller analysis

The specific surface area, pore volume and pore size of bentonite samples are shown in Table 2. The surface area of bentonite is increased to almost three times after acid
treatment. This is related to the unoccupied octahedral cation spaces left in the clay minerals. The acid-activated samples increased the pore volume due to the change in the structure of minerals and their decomposition has caused a reduction in the pore size.

3.1.3 | Thermal analysis

The mass loss profile of bentonite (Figure 7a) and the maximum rate of decrease in its weight percentage were analysed by the derivative weight percentage as shown in Figure 7b and it was observed that there are two levels of degradation taking place in the temperature range of 0–1000°C. The initial ambient temperature was at 25°C during the start of the experiment. Referring to Figure 7b, the first peak occurs at around 30°C which could be due to adsorbed water losses from the interlayer of mineral. The decomposition temperature at which this peak occurs was slightly lower for activated bentonite samples compared with the pure bentonite. The second peak takes place at a temperature of 485°C due to the loss of hydroxyl groups from the minerals and it was found to be the same for both samples. The residue after thermal decomposition was higher for pure bentonite samples compared with acid-activated samples.

3.1.4 | FTIR spectroscopic analysis

The FTIR spectra of the vibrational functional groups of the samples under infrared beam are shown in Figure 8. Major peaks were observed at adsorption bands of 3622, 1430, 1150, 792, 915 and 836 cm⁻¹. The stretching and bending vibrations of OH groups in the crystal were detected (3622 and 1644 cm⁻¹) due to adsorbed water molecules. The band at 3622 cm⁻¹ could be further related to the OH groups in the silicate layers.

The calcite peaks that were found to disappear after acid activation (Figure 6) were also confirmed by the decrease in the intensity band at 1430 cm⁻¹. This clearly indicates that calcite impurities are removed following acid-activation of bentonite. The band adsorbed at 1150 cm⁻¹ indicates the stretching of Si-O presence in the montmorillonite and no variation was observed in its silica layers after acid activation. The bands observed at 915 and 836 cm⁻¹ indicate a reduction in intensity due to the partial depletion of Al, Mg and Fe from the bentonite clay which is evident from the variation of its chemical structure after acid activation.

3.1.5 | Scanning electron microscopy analysis

The surface of the bentonite and activated bentonite was studied using high resolution, field emission gun-based scanning electron microscopy (FE-SEM). A better resolution on the image was obtained by applying a gold sputter coating before examining the samples. Figure 9 shows the FE-SEM images of bentonite and activated bentonite. It was observed that the two samples had a dissimilar structure, with the bentonite showing a smaller inter-particle distance than the activated bentonite. This could be the reason for the higher adsorption capability of bentonite after subjecting it to acid treatment.

3.1.6 | Energy dispersive spectroscopy analysis

The elemental composition of bentonite and activated bentonite samples shown in Table 3 was studied to calculate percentage change in mineral composition. It was observed that there are some chemical modifications to the structure of

| TABLE 3 | Elemental composition of bentonite and activated bentonite samples |
| --- | --- |
| Elements | Atomic composition (%) | Bentonite | Activated bentonite |
| O | 52.25 | 61.73 |
| Si | 9.3 | 17.96 |
| Al | 7.06 | 4.7 |
| Mg | 0.933 | 0.356 |
| Ca | 0.2 | 0.016 |
| Fe | 1.33 | 1.267 |

| TABLE 2 | Specific surface area and pore structure of bentonite and activated bentonite samples |
| --- | --- |
| Sample | S_{BET} (m²/g) | Pore volume (cm³/g) | Pore size (nm) |
| Bentonite | 52.75 | 0.09 | 7.44 |
| Activated bentonite | 164.08 | 0.22 | 5.42 |

| FIGURE 6 | XRD pattern of bentonite and activated bentonite |
bentonite after acid activation. An increase (18%) was seen in oxygen and significant reduction in metals (Ca, Mg and Al). This could be due to the exchange of cations present in the outer layer of the montmorillonite that causes its replacement with hydroxyl ions from the sulphuric acid easily [17]. The removal of calcite peaks observed in Figure 6 could be related to the large decrease in the percentage of calcium peaks (92%). The other metallic ions such as Al, Mg and Fe come under the second group belonging to the octahedral sheet. It is seen that the cations present in the octahedral group reduce much more slowly than the exchangeable cations present in the interlayer of montmorillonite lattice. It is more difficult for the acid activation to diffuse its ions into the octahedral cations compared with the interlayer cations where the ion exchange can take place at a much lower concentration. The reductions in the percentage of aluminium and magnesium elements were 33% and 62%, respectively. The small effect on the iron elements indicates a high resistance to acid attack. The increase in the Si\(^{4+}\) cations could be related to the decrease in the cations from interlayer and octahedral sheets of the bentonite structure. Therefore, it can be concluded that interlayer cations are more prone to the acid activation than octahedral cations and the tetrahedral site.

3.2 | Characterisation of fluid samples

3.2.1 | Stability and moisture content

Zeta potential analysis was carried out using a Horiba SZ-100 nano particle to understand the stability of titania nanoparticles in natural ester fluid with and without surfactant. The standard deviation in the measured value of zeta potential was less than ±5 mV. The electrophoretic behaviour was used to determine the stability with the repulsive forces between the two nanoparticles greater than the attractive forces. The zeta potential can be either positive or negative depending on the surface interaction between the two phases. Zeta potentials greater than 30 mV are considered to be stable and greater than 60 mV are found to be excellently stable [22]. A marginal increase (8%) in the zeta potential was observed in the unaged ester nanofluid with CTAB surfactant compared to the fluid without surfactant (Figure 10). After thermal ageing, the zeta potential decreases for both samples. The zeta potential of the aged nanofluid with CTAB surfactant was higher than that without the surfactant by approximately 34%, indicating excellent stability.

The moisture content of ester and ester nanofluids were performed using the 899-Coulometer Karl Fischer titrator, and the results before and after thermal ageing are shown in Table 4. It was observed that ester-based nanofluids exhibit a higher moisture content compared to the base fluid under both unaged and aged conditions. The addition of nanoparticles may play a role in enhancing the water formation [4]. The degradation of oil/pressboard insulation with ageing has caused further increase in the amount of moisture content.

3.2.2 | Corona inception voltage

Figure 11 shows the measured corona inception voltage (CIV) for the base, the thermally aged and the reclaimed nanofluid samples. The CIV was taken as the magnitude corresponding to the first discharge captured by the UHF sensor. The standard deviation in all the measured values of 30 readings was less than ±1.5 kV. It was observed from Figure 11a that
negative DC CIV was higher than that under the positive DC and AC. The localised electric field at the tip of the needle electrode has a high influence on the inception voltage. The ionization that takes place during positive polarity aids the applied electric field, thereby causing discharges at a lower voltage magnitude compared to negative polarity. Sample C shows a higher inception voltage compared with Samples A and B.

The high stability of nanofluids with surfactant (Sample C) may have inhibited the propagation of streamers due to Van der Waals force, by trapping all the local charges due to its electrical double layer formation around the nanoparticles [23]. It is inferred from Figure 11b that there is a decrease in the inception voltage after thermal ageing for all samples. The ageing contaminants such as copper intruding into the fluid, pressboard contaminants along with the ambience can alter the conductivity, thereby creating discharges at voltage magnitudes lower than that of the base nanofluids. The effect of an interfacial zone on Sample C would have caused higher inception voltage even under thermal ageing. Furthermore, the reduction in the inception voltage due to thermal ageing was around 30% in case of Samples A and B. The reclamation of aged fluid using activated bentonite showed a noticeable improvement in the inception voltage under all voltage types for Sample A as shown in Figure 11c, where an average of 20% increase was observed, compared with the aged fluid. This increase may be related to the absorption of carbon particles and copper ions by the bentonite layer [24, 25]. On the other hand, the depletion in the electrical double layer formation after thermal ageing could not be reclaimed with activated bentonite and hence a comparatively smaller increase in CIV was observed in the case of nanofluids (Samples B and C) under AC and positive DC voltages. No noticeable increase in the CIV under negative DC voltage was observed for these samples.

3.2.3 Breakdown voltage

Figure 12 shows the breakdown voltage measured under DC, AC and lightning impulse voltages. As expected, the lightning impulse breakdown voltage was higher than that under AC and DC voltages (Figure 12a), with the negative polarity impulse breakdown voltage higher than the positive polarity voltage. Electrons in the fluid are attracted towards the positive needle (anode) electrode creating ionization that causes localised heating. This creates a space charge field due to positive ion formation that causes a reduction in the applied electric field near the needle tip [26].

The reduction in the breakdown voltage after thermal ageing (Figure 12b) was much lower in Samples B and C as compared to Sample A. This could be due to trapping of electrons by the titania nanoparticles reducing their streaming velocity and increasing their propagation time [6]. Also, the antioxidant additive such as tocopherol which is present in natural ester fluid inhibits the ageing process and may not lead to much variation in breakdown voltage [27]. The activated bentonite slightly improves the breakdown voltage after reclamation of the aged fluid, as shown in Figure 12c.

**F I G U R E 9** FE-SEM image of (a) bentonite and (b) activated bentonite samples

**F I G U R E 10** Stability of ester-based nanofluids
3.2.4 | Dielectric dissipation factor

The dielectric dissipation factor of ester nanofluids for the base, aged and reclaimed samples is shown in Table 5. Before ageing, the nano fluid samples showed a relatively higher loss tangent compared to base ester fluid which may be due to increased number of conductive ions surrounding the electrical double layer [16]. Upon ageing, there could be a formation of various acidic and alcoholic derivatives along with solid contaminants from copper and pressboard material affecting the dielectric properties of insulating fluid which in turn causes an increase in tan δ [28]. Samples B and C observed a higher loss tangent compared to Sample A due to thermal dissociation of different layers of nanoparticles caused by ageing. After reclamation of aged fluid samples using activated bentonite, only a minor change in tan δ was observed due to the use of single adsorbent [29]. The complete depletion in the electrical double layer of nanofluids after thermal ageing could be the reason for the negligible effect observed on the dissipation factor even after the reclamation process.

TABLE 4  Moisture content of ester-based nanofluids

| Sample | Moisture content (ppm) |
|--------|------------------------|
|        | Before ageing | After ageing (50 days) |
| A      | 65             | 245                   |
| B      | 106            | 274                   |
| C      | 112            | 283                   |

3.2.5 | Flow electrification

(A) Streaming current of base fluid

Figure 13a shows the variation with time of streaming current for ester fluid (Sample A) and ester-based nanofluids (Samples B and C). A steep increase in the streaming current was observed during the initial 10 s of the disc rotation, which then attained a steady state. In the authors' work, the current was recorded for 300 s such that steady state conditions were reached. An increase in the current is observed with spinning velocity due to the effect of the variation of the Debye length. This increase may also be attributed to the friction exerted between the solid and liquid insulation [30]. A positive streaming current is observed in all oil samples (Figure 13b) and it was higher for Sample C compared with Samples A and B. This may be attributed to the presence of nanoparticles (TiO₂) added to the virgin ester. The addition of nanoparticles may have altered the composition of the ester fluid with their semiconductive nature causing an increase in the current. In addition, Sample C with the added surfactant showed an even higher streaming current due to its cationic nature which could have caused a large separation of charge at the interface of the two different dielectrics.

(B) Effect of impulse stress on charging tendency of oil

The variations of the streaming current with time and with disk velocity for the three impulse-stressed samples are shown in Figure 14a and b. It was noticed that flow current in Sample B
**FIGURE 12** Breakdown voltage for (a) base fluid, (b) thermally aged fluids and (c) reclaimed fluid

![Graph of Breakdown Voltage vs Sample](image)

| Sample | Before ageing | After ageing (50 days) | After reclamation |
|--------|---------------|------------------------|------------------|
| A      | 0.016         | 0.027                  | 0.024            |
| B      | 0.035         | 0.051                  | 0.049            |
| C      | 0.042         | 0.057                  | 0.056            |

**TABLE 5** Dissipation factors of ester-based nanofluids

**(C) Effect of thermal ageing on charging tendency of fluid**

The measured post-thermal ageing streaming current with respect to time is shown in Figure 15a. It can be seen that there is a large reduction in streaming current for all samples compared with base fluid samples. The streaming current associated with Sample A after thermal stress (Figure 15b) is higher than that measured after impulse stress, which may be due to the effect of copper dissolution in the fluid. The presence of antioxidant additives in base ester fluid [27] does not allow much thermal dissociation on ageing, preventing further charge formation at the interface. Samples B and C showed a much larger reduction in streaming current compared to the case without the application of stress. For Sample B, the tetrahedral structure of titania nanoparticles (anatase) breaks, which allow negative oxygen ions into the fluid, reducing streaming current. Sample C showed the largest reduction in streaming current which could be related to the depletion of the stabilising electrical double layer created by the surfactant. As a result, unlike under impulse stress, it appears that thermal ageing negatively affects the fluid stability. The effect of thermal stress is to introduce negative ions in the fluid causing a reduction in the streaming current for ester-based nanofluids.

**(D) Effect of charging current after fluid reclamation process**

Figure 16 shows the variation in the streaming current for the samples that were reclaimed after thermal ageing and impulse stressing. Also, dotted lines represented in the Figure 16a and b is an indication of stressed samples (Figures 14b and 15b) which is...
FIGURE 13  (a) Variation of streaming current versus time at \( \omega = 500 \) rpm and (b) steady state current for different disk velocity

FIGURE 14  Measured streaming current after impulse stress application (a) variation with time at \( \omega = 300 \) rpm and (b) steady state current for different disk velocity

FIGURE 15  Measured streaming current after thermal ageing (a) variation with time at \( \omega = 300 \) rpm and (b) steady state current for different disk velocity

FIGURE 16  Streaming current after reclamation process following (a) impulse stress and (b) thermal ageing
shown for comparison. The fluid reclamation using bentonite does not cause much variation in the streaming current, except for Sample B reclaimed after impulse stress (Figure 15a), where a noticeable reduction is observed.

There was no change in the trend and in the polarity of the streaming current for sample B after reclamation. This may be due to the absorption of carbon particles and non-modification of physico-chemical properties after reclamation. The samples reclaimed after thermal ageing (Figure 16b) showed similar results for Samples B and C. For sample C, the severe depletion of the electrical double layer as a result of thermal ageing gave a streaming current behaviour similar to that of Sample B. Sample A did not show much variation in its current even after reclamation.

4 | CONCLUSIONS

- The surface modification of bentonite with acid treatment showed an enhanced adsorption capability, with the removal of calcite peaks and octahedral cations from the montmorillonite structure and increased silica in the tetrahedral layer.
- The activated bentonite indicated a lower decomposition temperature and residue compared with pure bentonite. The stretching of Si-O bonds and the removal of Al, Fe and Mg ions in the octahedral layer were observed using the FTIR analysis which was also confirmed through its chemical composition.
- The corona inception voltage of ester and its nanofluids was higher under negative applied DC voltage compared to positive DC and AC voltages. The retardation in the streamer propagation due to electrical double layer formation increased the corona inception voltage in Samples B and C compared with Sample A. Thermal and impulse stress applied to these samples indicated a similar trend in inception voltage. A slight improvement in the inception voltage of about 20% was observed after reclamation which was related to the adsorption of carbon and copper ion interaction with bentonite.
- The breakdown voltage of ester and its nanofluids was higher for negative lightning impulse than the positive lightning impulse. The change in breakdown voltage was not significant due to the antioxidant additive which retards its degradation upon ageing. Also, the reclaimed fluid maintained the same level of breakdown voltage as that of aged samples.
- The dissipation factor of Samples B and C was higher than Sample A after thermal ageing and only a minor change was observed after reclamation due to depletion in the double layer.
- The flow electrification of Sample A was lower when compared with Samples B and C. The semiconductive nanoparticles added to ester fluids increased their streaming current. A negative polarity of streaming current was observed in Sample B due to dissociation of oxygen atoms following impulse stressing. A lower streaming current was observed for Samples B and C compared with Sample A upon thermal ageing. The reclamation of aged fluid did not reduce its flow current magnitude.

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CONFLICTS OF INTEREST

The authors declare that there is no conflict of interest.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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