Moisture-dependent interface and bulk behaviour of space charge in DC-stressed oil-impregnated pressboard insulation

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Abstract
In this paper, the effect of moisture on the charge behaviour in oil-impregnated pressboard is analysed based on the characteristics of the interface and the bulk. Space charge behaviour and infrared spectra are acquired from a sample with a high moisture content of 1.92% and another with low moisture content of 0.13%. The results show that a greater number of hydrogen bonds form in the high-moisture sample. The threshold fields of charge injection are 1 and 0.6 kV/mm in the low- and high-moisture samples, respectively. The dynamic charge transport in bulk is significantly accelerated by moisture. The drop in the injection barrier at the interface and the accelerated transport of charge in bulk in the high-moisture sample may be associated with the mediating effect of a water layer at the interface and the lower trap energy in bulk caused by the hydrogen bonds, respectively. The combined changes in the interface and the bulk led to the dominant position of negative charges inside the high-moisture sample, regardless of the distribution area or the charge amount.

1 INTRODUCTION

Oil-immersed transformers are the main components in power systems, and are crucial to voltage conversion and power transmission. The reliability of oil-impregnated pressboard insulation has a significant impact on system stability owing to its important role in the transformer [1]. Research has shown that moisture promotes the formation of traps and accelerates the ageing of insulation [2–6], which increases the risk of serious electric accidents during operation. It is generally known that the majority of moisture in a transformer occurs in the pressboard insulation. Therefore, the parameters that influence the accumulation of space charge in oil-impregnated pressboards should be further researched.

The characteristics of space charge in oil-impregnated pressboard insulation have been recently investigated [7]. Liao et al. claimed that an electric field primarily affects the amount of charge, whereas temperature primarily affects charge distribution and mobility [8]. Chen et al. studied multi-layer samples with moisture concentrations of 0.28%, 1.32%, and 4.96% to show that moisture promotes charge injection, and a higher moisture content is associated with faster charge dissipation [9]. Zhou et al. investigated the effects of moisture and high temperature on charge behaviour. They found that the rate of space charge formation can be increased when the moisture content is lower than 7%, but is suppressed when moisture content is higher than 9% [10]. The results of experiments on accelerated thermal ageing have shown that the moisture absorption capacity of insulation paper decreases with deepening ageing degree whereas that of oil increases [11]. The ratio of moisture content in paper to that in oil decreases with decreasing polymerisation according to similar research [12]. Prevalent studies in the area have mainly focused on the analysis of the interface or the bulk behaviour of space charges in an oil-impregnated pressboard, and the related correlation analysis is weak. However, the characteristics of charge accumulation in bulk are significantly associated with charge injection from electrodes. It is thus important to investigate the correlation between the interface and bulk behaviour to clarify the mechanism of change in parameters influencing charge transport.

In this paper, the correlation in charge transport between the interface and the bulk is investigated for an oil-impregnated pressboard insulation based on the moisture-dependent behaviour of the space charge and infrared spectra. An

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558 wileyonlinelibrary.com/iet-gtd

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Al-water-oil contact model is proposed to explain the influence of moisture on interface properties. The charge transport in bulk is analysed through the relevance between moisture-dependent molecular structure of cellulose and trap depth. The mechanism study of moisture affecting charge transport process in transformer insulation provides guidance on the new method of improving insulation performance in industrial insulation design.

2 | EXPERIMENTAL PROCEDURES

2.1 | Sample preparation

The cellulose pressboard used in this study accorded to the international standard IEC60641-3-1:2008 [13], and was 300 µm thick. Karamay 25# mineral oil was used as fresh insulation oil. Two types of samples, with low and high moisture contents, were prepared according to IEC 60422:2013 [14]. A Buchner funnel was used to filter out impurities from the insulation oil, which was then vacuumed to remove any air that might have mixed with it during filtering. For the low-moisture sample, the insulation pressboards were vacuum-dried for 24 h at 353 K, and the dried pressboards were vacuum-impregnated by treated oil for 48 h at 313 K. The moisture content of the resulting sample was 0.13% as determined by the Karl–Fisher method, which is lower than that specified as the standard moisture content for a new transformer insulation according to CEI/IEC 60422: 2013 [14]. For the high-moisture sample, undried insulation pressboards were directly impregnated by treated oil under the same constraints as above. The moisture content of the resulting sample was 1.92%. The preparation process is shown in Figure 1.

2.2 | Space charge measurement

Space charge experiments were carried out at room temperature based on the pulsed electro-acoustic (PEA) method. The anode was a semi-conductive layer with a 10 mm diameter and 1200 µm thickness, and the cathode was an aluminium (Al) plate. The spatial resolution was about 8 µm. The thickness of the sample after being placed in the PEA device was about 250 µm owing to mechanical stress. The structure of the experiment is shown in Figure 2. Each sample was measured under DC stress of 0.1, 0.3, 0.6, 1, 2, 3, 4, 5, 6, and 12 kV/mm for 1200 s. High voltage stress were applied after 600 s of removed voltage following each polarisation process.

2.3 | Infrared detection

Fourier transform-based infrared detection was performed to analyse different organic functional groups that provided an auxiliary to investigate the influence of moisture content on the dielectric properties of oil-impregnated pressboard insulation. An IR/Nicolet 6700 infrared spectrometer manufactured by American Thermo Fisher Company was used in this work. The wavenumber was set between 4000 and 600 cm$^{-1}$. A background scan was performed before each measurement and each sample was scanned 32 times.

3 | EXPERIMENTAL RESULTS

3.1 | Threshold electric field of charge injection and accumulation

Figure 3 shows the charge distribution characteristics of the two samples at fields ranging from 0.1 to 1 kV/mm during 1200 s of polarisation. For the low-moisture sample, no charge accumulation was observed when the field was lower than 0.6 kV/mm. A small amount of homocharges appear near the cathode and anode when field was 0.6 kV/mm. When it reached 1 kV/mm, a significant number of homocharges accumulated around the electrodes, and the scope of charge distribution gradually extended with time. For the high-moisture sample, no charge accumulation was observed when the field was lower than 0.6 kV/mm. A small amount of homocharges appear near the cathode and anode when field was 0.6 kV/mm. When it reaches 0.6 kV/mm, homocharge accumulation adjacent to the cathode and the anode was non-negligible, and the scope of charge distribution quickly stabilised. The threshold electric
fields of charge injection in samples with moisture contents of 0.13% and 1.92% were ≈1 and 0.6 kV/mm, respectively.

Assuming that the mean volume charge density $\rho_0$ of a sample with no charge injection was background noise, and the apparent depth of the injected charges that migrated inward as a function of time was determined by the charge density $\rho_{0,t}$, the scope of charge distribution was fitted according to a scatter distribution. When the field was increased to the threshold field, the charges were mainly concentrated near the electrodes. A subtraction method has been proposed to remove the influence of capacitive charges on charge distribution characteristics [15]. In the data processing, the charge quantity was obtained by the volume integral of the charge distribution after treatment by the subtraction method:

$$q_a(t, E) = \int_D \int_0^L q_a'(x, t, E) \, dx \, dS \quad (1)$$

where $L$ is sample thickness, $q_a'(x, t, E)$ is the charge density without the capacitive charge, $D$ is the effective area of the electrode, and $a$ represents the type of charge, i.e. positive, negative, and absolute charge density.

The characteristics of charge quantity as a function of the electric field were calculated from the absolute charge density at 1200 s according to Equation (1). As shown in Figure 4, the threshold electric field at which the charge amount began to rapidly increase in the high-moisture sample was slightly lower than that in the low-moisture sample. In the first stage, the charge quantity of the latter remains essentially constant, whereas that of the former showed a slight upward trend. There was no significant difference in the rate of increase between the two samples in the second stage.

### 3.2 Charge accumulation characteristics

Figure 5 shows the charge distributions of the two samples. Because the space charge density obtained from the PEA method was the net charge density, the apparent depth here reflected the predominant charge in the area. For the low-moisture sample, the negative charges migrated to an apparent depth of 85 µm from the cathode. The apparent depth of positive charges extended inward with increasing electric field and pressure time, and ultimately approached the apparent depth of negative charge. The high-moisture sample was filled with charges injected from the cathode and the anode during the initial stage of polarisation, and the negative charges migrated to an apparent depth of 196 µm from the cathode. From the perspective of predominant bulk charge polarity, the low-moisture sample mainly carried positive charges whereas the high-moisture sample mainly carried negative charges. The charge distribution of the high-moisture sample also achieved dynamic balance substantially more quickly.
The quantities of negative and positive charges as a function of pressure time were calculated from Equation (1), as shown in Figure 6. The variation in negative charge was consistent with that in the positive charge in both samples. The charge quantity of the low-moisture sample increased slowly after remaining constant at the beginning of the polarisation, and did not reach stability at 1200 s. The charge quantity of the high-moisture sample increased rapidly during the initial stage and then stabilised. The amount of negative charge in the high-moisture sample was more than twice that of the low-moisture sample, whereas the amount of positive charge in the former was less. Hence, moisture mainly contributed to the accumulation of negative charges and promoted stability in terms of changes in the charge amount.

3.3 Infrared detection

The hydrogen bonds and glycosidic bonds of cellulose have an important influence on the performance of the transformer insulation [16]. The infrared spectra are shown in Figure 7. The results show that no difference is observed in the absorption peaks at 1162, 1106, and 894 cm\(^{-1}\) between the two samples, which indicates that the number of glycosidic bonds remains invariant with increasing moisture content. The absorption peaks at 1647 and 3346 cm\(^{-1}\) are higher in the high-moisture sample than in the low-moisture sample. Moisture therefore presumably contributes to the generation of the hydrogen bonds.

4 DISCUSSION

4.1 Influence of moisture on interfacial property

Contact between materials with different Fermi levels may lead to free carrier flow, and an electric double layer forms near the surface to prevent carrier flow until the Fermi levels equilibrate at contact [17]. During the space charge measurements, an oil layer of macroscopic thickness is thought to be in close contact with the surface of the electrode. Moisture occurs, and not only in the form of molecules in the oil-impregnated pressboard. Because the permittivity of water is higher than that of the insulation oil, the moisture is likely positively charged due to friction. Once DC voltage is applied, a certain number of the charged water molecules may migrate to the surface of the Al electrode and form a water layer.

For low-moisture sample, the schematic of the contact between the electrode and oil is shown in Figure 8(a). \(E_0\) is the vacuum level, \(E_{F_A}\) is the Fermi level, \(E_{C_A}\) is the conduction band, \(E_{V_A}\) is the valence band, \(\phi_a\) is the work function, and \(\chi_a\) is the electron affinity, where subscript \(a\) represents the material type (e.g. Al, water or oil). In this case, the energy \(\phi_m-\chi_s\) is needed for the electrons to be injected into the oil. For high-moisture sample, an Al-water-oil contact structure likely forms when the moisture content is high, as shown in Figure 8(b). The water layer is equivalent to a semiconductor relative to the Al electrode, which is equivalent to a conductor.
FIGURE 6  Charge quantity as a function of pressure time. The arrows indicate the time at which the charge quantity stabilised. (a) Negative charge, (b) Positive charge

FIGURE 7  Infrared spectra of the two samples

relative to the oil layer. In this case, the energy required to pass through a single contact surface decreases relative to the direct contact between Al and oil. Thus, the applied field required for electrons to be injected into the sample decreases, such that moisture reduces the injection barrier of electrons from the electrode to the insulator.

Recently, nanotechnology has become a primary focus in modification of polymeric materials. The performance of paper impregnated with nano-modified oil was investigated in [18]. It shows that the amount of space charges accumulated in the nano-oil impregnated paper is much smaller than that in fresh oil impregnated paper. It is believed that the TiO₂ nanoparticles increase the density of deep traps in the dielectric and thereby suppress the electric field distortion. Thus, it is considered that nanoparticles can be added into transformer insulation to improve the problem of lower trap energy caused by the increased moisture content.

4.2  Influence of moisture on bulk property

The charge dissipation process of the two samples is shown in Figure 9 in logarithmic coordinates. A slightly faster pace is observed in the low-moisture sample in the initial depolarisation stage, which then gradually slows. For the high-moisture sample, the charge amount is substantially attenuated at the onset of depolarisation and essentially reaches a steady state around 20 s. Moreover, the amount of residual charge in the high-moisture sample is significantly less than that in the low-moisture sample in the late stage. Higher applied fields are associated with more residual charges in the low-moisture sample during depolarisation, whereas the applied field has almost no effect on the amount of residual charges in the high-moisture sample and only leads to a faster decay rate.

The trapped charges are released due to thermal vibration after removal of the applied field. At constant temperature, the detrapping process is closely related to the trap energy [19]:

\[
\frac{dn_t}{dt} = -n_t \nu \exp \left(-\frac{U_t}{kT}\right)
\]

(2)

where \(t\) is time, \(U_t\) is the trap energy, \(n_t\) is the trapped charge density, \(\nu\) is the free-electron thermal velocity, \(k\) is Boltzmann’s constant, and \(T\) is the applied temperature.

Hence, the main trap energy levels during different stages can be approximated by the attenuation characteristics. The charge dissipation process is divided into three stages of 1–10, 10–100, and 100–1200 s. The trap energy is calculated by the fitting result of charge density, as shown in Table 1. In the first stage, the trap

| Water content | Trap energy [eV] |
|--------------|-----------------|
|              | I   | II  | III |
| 0.13%        | 0.8154 | 0.9219 | 0.9402 |
| 1.92%        | 0.7546 | 0.9250 | 1.0094 |
energy in the high-moisture sample is lower than that in the low-moisture sample, whereas the opposite is observed in the third stage. The energy levels do not differ much in the second stage. In combination with the amount of charge attenuation at each stage, a trap depth of about 0.75 eV is observed to obtain a dominant position in the high-moisture sample, which results in stronger charge release in the initial stage. A certain amount of charge is released at each stage in the low-moisture sample, indicating that the high-energy trap position is of equal importance. This implies that moisture helps lower the mean trap depth.

In the mixed system of mineral oil and cellulose pressboard, the cellulose with high polarity shows a stronger sorption with water molecules than mineral oil [20]. Moisture and acid are generated in the system during thermal ageing, and can lead to cellulose hydrolysis and accelerated insulation failure [19]. However, in this work, the samples were not placed in a high temperature environment for a long time and sample preparation and experimentation were performed within one week. With this in mind, it is speculated that no significant cellulose hydrolysis is present in the system. Recall that in the infrared spectra, no clear difference is observed in the absorption peaks at $1162$, $1106$, and $894 \text{ cm}^{-1}$ between the two samples, which further verifies this point.

Khan et al. investigated moisture sorption in cellulose by sorption isotherms and dielectric response [21]. The results
show that moisture tends to be absorbed in the regions of amorphous cellulose, as shown in Figure 10, where hydrogen bonds are represented by red dotted lines. There is a large number of hydroxyl groups on the cellulose molecules. In the crystalline region, hydrogen bonds form between the hydroxyl groups, which makes the cellulose molecules close and orderly. Thus, the molecular structure in this region is stable and does not easily react with water molecules. Hydrogen bonds in the amorphous area form easily between the water molecules and hydroxyl groups exposed on the cellulose molecules. This is consistent with the larger number of hydrogen bonds in the high-moisture sample shown by the infrared spectrum. The cellulose molecules in the amorphous region tend to be ordered due to the existence of hydrogen bonds, which may change the trap energy distribution in the system and lower the mean trap energy. Therefore, charge transport changes with increasing moisture content owing to the lower trap energy. Because the low-moisture sample is dominated by negative charges after the short circuit, the effect of moisture on trap energy therefore mainly alters the electron migration.

Much effort has been devoted to suppress space charge injection. An aluminium oxide functional film was deposited on a cellulose insulation pressboard surface to investigate the dielectric properties and space charge behaviour [22]. The results show that the nano-structured Al₂O₃ film sputtered on the fibre surface could act as a functional barrier layer for suppression of the charge injection. It means that the adverse effect of moisture on the interface performance may be improved by sputtering nano-film on the surface of pressboard. More work is need to confirm the specific performance.

4.3 Influence of moisture on space charge transport

According to the above analysis, moisture contributes to lower the electron injection barrier. The threshold electric field of charge injection is clearly lower in the high-moisture sample. Moreover, the number of injected electrons increases because of the lower barrier, based on Schottky injection theory. In addition to providing energy to overcome the Schottky barrier, the electric field provides an initial energy for the charge to migrate inward during injection. The energy of the injected electrons can be expressed as [23]:

\[ E = A \exp \left( -B \frac{(\phi_M - \chi_I)^{3/2}}{F} \right) \]

(3)

where \( \phi_M \) is the work function of the electrons, \( \chi_I \) is the electron affinity of the insulator, \( F \) is the electric field, and \( A \) and \( B \) are constants.

Equation (3) shows that the smaller the injection barrier, the more energy that can be obtained. The injected electrons in the high-moisture sample therefore have a higher initial energy than in the low-moisture sample, which helps the inward movement of electrons.

Apparent mobility is another important parameter for characterising charge migration. The mobility in low fields is mainly affected by the trap energy, which can be simplified to [24]:

\[ \mu = \frac{\lambda \nu}{F} \exp \left( -\frac{E_i}{kT} \right) \]

(4)

where \( \lambda \) is the distance between traps and \( E_i \) is the trap energy.

Equation (4) is reasonable under the condition of \( q\lambda F \ll kT \). Lower trap energies are obviously associated with higher carrier mobility. Thus, charge mobility in the high-moisture sample is higher than that in the low-moisture sample due to the lower trap energy. The charge accumulation in the bulk is enhanced by the combined effects of higher initial energy and charge mobility. The threshold electric field at which the charge amount begins to rapidly increase is lowered by moisture.

Significant charge accumulation can lead to electric field distortion. The electric field distribution after 1200 s of polarisation at 6 and 12 kV/mm is shown in Figure 11. The electric field distribution of the low-moisture sample is relatively uniform, whereas the negative charge distribution in the bulk of the high-moisture sample results in a substantial linear increase in electric field, and there is a clear turning point at the boundary of the negative and positive charges.
results of the maximum electric field distortion rates are shown in Table 2. The distortion rate of the high-moisture sample is two times higher than the low-moisture sample, reaching up to about 70%. According to [25, 26], the region that must be regarded as the most extreme with respect to electrical ageing are those where energy concentration can continue to increase once it has started. Thus, the area in high-moisture sample where the electric field is concentrated due to the large number of negative charge distribution has a high risk of local damage. Insulation ageing could therefore be accelerated by the enhanced field in the high-moisture sample.

In summary of the above analysis, most charges injected from the cathode are trapped near the interface due to the large number of electron traps in the low-moisture sample. The area of positive charge thus holds sway even if the number of injected holes is less than the electrons. With increasing moisture content, a large number of negative charges accumulate in the sample because of more injected electrons and higher charge mobility. Negative charge then holds the dominant position in the high-moisture sample regardless of the distribution area or charge amount.

To verify the above analysis, the simulation of charge transport is carried out based on bipolar charge transport model. Considering a one-dimensional problem, the charge transport process is described by the following equations [27].

\[
\begin{align*}
\frac{\partial j(x,t)}{\partial t} + \frac{\partial j(x,t)}{\partial x} &= s(x,t) \\
\frac{\partial n(x,t)}{\partial t} + \frac{\partial j(x,t)}{\partial x} &= s(x,t) \\
\frac{\partial E(x,t)}{\partial x} &= \frac{\rho(x,t)}{\varepsilon_0 \varepsilon_r}
\end{align*}
\]

where \(x\) is spatial coordinate, \(t\) is time, \(j\) is current density, \(n\) is carrier density, \(\rho\) is the net density of charge, \(\mu\) is the carrier mobility, \(E\) is the electric field, \(D_i\) is the diffusion coefficient, \(s\) is the source term, \(\varepsilon_0\) and \(\varepsilon_r\) are the permittivity of vacuum and relative dielectric constant, respectively.

The parameters used in this numerical simulation refer to [28]. In view of the correlation between the interface and bulk behaviour, four types of conditions are set for the injection barrier and trap depth, as shown in Table 3. The relatively high injection barrier and trap depth are set in condition A. The injection barrier and trap depth are lowered in condition B and condition C, respectively. Both parameters in condition D decrease.

The simulation results are shown in Figure 12. It shows that the result of condition A is in line with the charge distribution of low-moisture sample. The injected electrons accumulate near the interface if the injection barrier is lowered while keeping the trap depth constant. The electric field is reduced due to the electron accumulation near the interface, which prevents further injection of electrons. The injected electrons are more likely to migrate to the bulk of the sample if the trap depth is lowered. However, there is no significant increase in electron amount due to the constant injection barrier. While the injection barrier and trap depth of electron are lowered simultaneously, the electron accumulation holds the dominant position regardless of the distribution area or the charge amount, which is in accord with the charge distribution of high-moisture sample. Thus, it is reasonable to confirm the comprehensive influence of moisture on the interface and bulk behaviour of space charge.

5 | CONCLUSION

In light of the space charge characteristics and infrared spectra of two samples with different moisture contents, the main results are summarised as follows.

| Water content | Electric field distortion rate |
|---------------|------------------------------|
|                | 6 kV/mm | 12 kV/mm |
| 0.13%          | 23.72%  | 27.01%   |
| 1.92%          | 79.54%  | 69.02%   |

**TABLE 3** Simulation parameters

| Types of condition | Carrier | Injection barrier [eV] | Trap depth [eV] |
|--------------------|--------|------------------------|-----------------|
| A (High-moisture)  | Electron | 1.10                  | 0.97            |
|                    | Hole    | 1.10                  | 0.95            |
| B                  | Electron | 1.05                  | 0.97            |
|                    | Hole    | 1.10                  | 0.95            |
| C                  | Electron | 1.10                  | 0.90            |
|                    | Hole    | 1.10                  | 0.95            |
| D (Low-moisture)   | Electron | 1.05                  | 0.90            |
|                    | Hole    | 1.10                  | 0.95            |
(i) The influence of moisture on interfacial properties appears as a decrease in the threshold electric field of charge injection. An Al-water-oil contact model is proposed to explain the reason for the reduction of the contact barrier. The energy required to pass through a single contact surface is lower than that for a direct contact between Al and oil.

(ii) The influence of the moisture on bulk property is reflected in the accelerated transport of charge. In terms of chemical structure, hydrogen bonds formed between the water molecules and hydroxyl groups exposed on the amorphous cellulose lower the mean trap depth.

(iii) The field distortion rate of the high-moisture sample is more than twice that of the low-moisture sample owing to the significant accumulation of negative charge even at low field. Attention should be paid to the electrical ageing of the insulation enhanced by moisture in the transformer design.

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