Experimental and simulation analysis of the influence of moisture on dielectric and breakdown properties of insulation pressboard used in power transformer

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Abstract. The influence of water molecules on cellulose has attracted much attention. The influence of water molecules on the dielectric property and breakdown property of cellulose requires in-depth research. In this paper, firstly, the effect of water on the dielectric spectrum in the frequency domain dielectric spectrum and the breakdown voltage of cellulose were studied. Then the parameters such as free volume, hydrogen bond, energy level and state density of cellulose under different moisture content were analysed by molecular simulation. The changes of dielectric properties and breakdown properties of cellulose under different water content were studied and summarized. The variation mechanism of dielectric property and breakdown property of different moisture content was explained by molecular simulation.

1. Introduction
Transformer is one of most important equipments in the power system which plays a major role in safe operation of power system. Oil-paper insulation, as a classical insulation structure, has been successfully used in transformers for many years. Transformer insulation board is an important part of oil-paper insulation, which is mainly composed of cellulose. In the normal operation of power equipment, moisture is one of the most important factors leading to cellulose insulation degradation, which is not only the product of cellulose insulation degradation, but also the catalyst of aging [1, 2]. Therefore, it is necessary to analyse the influence of water content on the dielectric properties and power frequency breakdown properties of paperboard, and to analyse the mechanism of water influence from the micro perspective.

A large number of scholars have studied the dielectric property and power frequency breakdown property of oil-paper insulation system of transformer. Yang L. et al studied the frequency domain dielectric properties of oil-impregnated paperboard under different moisture content and different degree of polymerization [3]. Mu Z. studied the dielectric properties of oil-impregnated paper samples with moisture content of 0.69%~6.43% [4]. The results show that the higher the water content is, the higher the true dielectric constant is, and the higher the water content is, the higher the loss is. At the same time,
the breakdown characteristics of oil-impregnated paperboard have also been widely reported [5, 6]. The results show that the breakdown characteristics of oil-impregnated paperboard vary with the influencing factors. In this case, due to the technical limitations in the laboratory, most studies mainly focused on the experimental results, few studies analysed the micro-mechanism of dielectric and breakdown characteristics of insulating pressboard.

Recently, molecular simulation technology and its application have attracted enormous attention. Y. Mo et al. studied the mean square displacement and polarization strength of both cellulose and modified cellulose by using molecular dynamic simulation [7]. R. Liao et al. showed the water diffusion behaviour in oil-paper insulation by analysing interaction energy, hydrogen bond, free volume and diffusion coefficient [8]. So far, few people have studied the micro mechanism of dielectric properties and breakdown properties of insulating pressboard.

In this paper, firstly, three kinds of mineral oil impregnated paperboards with water content were prepared and their frequency dielectric properties and power frequency breakdown properties were measured. Then the parameters such as free volume, hydrogen bond, energy level and state density of cellulose under different moisture content were analysed by molecular simulation.

2. Experiment and molecular simulation

2.1. Experiment part

2.1.1. Sample preparation.
In this paper, 2mm thick ordinary cowhide insulating board and commercial 25 # naphthenic mineral oil were selected as test materials. The parameters of insulating board and insulating oil are shown in Table 1. Before the test, the cellulose board is cut into disks of 160mm diameter. In order to simulate the actual drying process of transformer, it is necessary to pretreat insulating board and insulating oil. The pretreatment process is as follows: First, the insulating board and insulating oil are dried for 48h in a vacuum environment of 105 °C and less than 1mbar, respectively. Pour the dried insulating oil into the stainless steel tank containing insulating cardboard and vacuum pump it for 48h at 60 °C. The proportion of insulation board and oil is 1:10. Finally, the moisture content of the dried insulating board in different positions was tested to ensure that the moisture content was at a lower level and the value was equal.

Table 1. Parameters of insulation pressboard and insulation oil

| Cellulose pressboard | Insulation oil |
|----------------------|---------------|
| Density 1.2g/cm³     | Density 884.6 kg/m³ |
| thickness 2 ± 0.15 mm | Flash point 143 ºC |
| diameter 160 mm      | Pour point <-24 ºC |

In order to prepare oil-impregnated pressboards with different moisture contents, the treated oil-impregnated pressboard was placed in the air for natural moisture absorption. The moisture absorption test procedure of oil-impregnated cellulose pressboard is as follows. A pressboard sample with insulation oil removed from the surface was placed on a precision electronic balance, and its initial mass was recorded. Then, the oil-impregnated pressboard was put in the air to naturally absorb moisture. The pressboard was weighed according to the moisture absorption time until the pressboard quality after absorbing moisture met the expected requirements. Finally, the pressboard was placed in the insulation oil for oil-moisture balance. In the above process, the Karl Fischer Moisture Analyzer was used to measure moisture content of the oil-impregnated pressboard. The average of the three test results was selected as the moisture content of the oil-impregnated pressboard. In this study, the moisture content of unaged oil-impregnated pressboard is 1%, 3%, and 5%, respectively.

2.1.2. Frequency domain dielectric response test.
The three-electrode test system was used to perform time-frequency domain dielectric response tests on oil-impregnated pressboards containing with different moisture contents. The instrument used in the
FDS test was the IDAX300 produced by the Swedish Megger Company. The effective value of test voltage was 140 V and the test frequency range was from $10^{-3}$ Hz to $10^4$ Hz. The test temperature is 45 °C. Test platform of the frequency domain dielectric response is shown in figure 1(a).

2.1.3. Power frequency breakdown test.
The AC breakdown voltage of the oil-immersed insulation paper (pressboard) was tested using an equal diameter electrode in accordance with IEC 60243. The rise rate of voltage chose 1kV/s and the interval between the two breakdown tests is about 3 minutes. The average value of the five measurement results for each sample at each temperature was analyzed in this paper. The test temperature is 45 °C. The breakdown test electrode is shown in figure 1(b).

2.2. Molecular simulation

2.2.1. Model building.
The models' establishment and molecular simulations were both accomplished by using Material Studio software. The amorphous cell tool was used to construct insulation pressboard model with water content of 1%, 3% and 5%. The insulating board is mainly composed of cellulose. Figure 2 shows the combination of water molecules with different mass fractions and cellulose molecules, while the pink part represents water molecules.

2.2.2. Molecular dynamic simulation process.
To make the model reasonable, geometry and energy optimization were required. The geometry of two kinds of models were optimized by the “Smart” method with 1000 steps. Annealing optimization was performed with 5 cycles in the range from 300K to 500 K. Under PCFF forcefield, NPT was used to balance each model and to make the model more reasonable with 500 ps. Finally, NVT ensemble was
used for molecular dynamics simulation with 500 ps. The obtained trajectories were used to analyse the models’ microscopic parameters. The simulated temperature is 45 °C.

2.2.3. Density functional theory (DFT) simulation.
Based on the density functional theory (DFT), the calculations for two natural esters were analyzed by DMol3 Tool in Materials Studio. The geometry optimization, molecule orbitals, energy band and density of states (DOS) were calculated by employing the BLYP function under the generalized gradient approximation (GGA) exchange-correlation term. The Tkatchenko-Scheffle DFT–D (density functional theory- dispersion) dispersion correction method was adopted.

3. Results and discussions

3.1. Influence of moisture on dielectric response of oil-impregnated paperboard
As shown in figure 3, the real and imaginary parts of the relative permittivity of oil-impregnated pressboard containing different moisture contents are displayed. From figure 3 (a), the low frequency region (2.2×10⁻⁴-10¹ Hz) of the real part of relative permittivity of oil-impregnated pressboard is seriously affected by moisture. The curve of the real part of relative permittivity gradually moves to the upper right with the increase of the moisture content. However, in the high frequency region (10¹-10⁴ Hz), the real part of relative permittivity is basically not affected by moisture. From figure 3 (b), the imaginary part of relative permittivity moves up to the upper right with the increase of water content.

3.2. Influence of moisture on AC breakdown voltage of oil-impregnated paperboard
Figure 4 shows breakdown voltage of oil-impregnated pressboard with different moisture contents. When the water content in paperboard is 0.98%, the breakdown voltage is 46.3kV. When the water content in paperboard is 5.02%, the breakdown voltage is 42.5kV. It can be seen that with the increase of the moisture content in the insulating pressboard, the breakdown voltage presents a downward trend.

Figure 3. The real and imaginary parts of the relative permittivity of oil-impregnated pressboard with different moisture contents

Figure 4. Breakdown voltage of oil-impregnated pressboard with different moisture contents
3.3. Micromechanism analysis of influence of water content on dielectric property

The free volume of two types of celluloses were calculated, as shown in figure 5. The grey part shows the occupied volume, and the blue one stands for free volume. The fraction of free volume (FFV) of two celluloses were calculated, as demonstrated in Table 2. It can be seen that the free volume of cellulose with more water content is larger. The greater the free volume fraction in cellulose, the greater the carrier movement space in cellulose, so the conductance loss will increase.

![Figure 5. Free volume of cellulose with different moisture contents](image)

Table 2. FFV of cellulose with different moisture contents

| Model             | Free volume(Å³) | Occupied volume(Å³) | FFV(%) |
|-------------------|-----------------|---------------------|--------|
| Cellulose +1 % water | 1024.01         | 8773.77             | 10.45  |
| Cellulose +3 % water | 1112.12         | 9013.54             | 10.98  |
| Cellulose +5 % water | 1259.97         | 9227.85             | 12.01  |

Table 3 summarizes the changes in the number of hydrogen bonds in the whole model, pure cellulose, pure water and between water molecule and cellulose. The snapshot of hydrogen bonds between water molecule and cellulose is shown in figure 6. With the increase of water concentration in cellulose, the number of hydrogen bonds in water molecule-cellulose increases rapidly, while the number of hydrogen bonds in pure cellulose decreases. The hydrogen bond formed by water and cellulose reduces the hydrogen bond formed by cellulose itself, thus reducing the stability of cellulose. The less stable the cellulose is, the more dynamic it is and the more likely it is to turn under the action of an electric field, thus increasing dipole turning polarization.

![Figure 6. Snapshot of hydrogen bonds between water molecule and cellulose](image)
The polarizability of the medium in the model is obtained by dividing the total dipole by the model volume, which is summarized in Table 4. It can be seen from Table 5 that the polarizability of the amorphous region increases from $9.11 \times 10^{-3}$ to $12.86 \times 10^{-3}$ with the increase of moisture content. Under the action of external alternating electric field, the internal polarization ability of cellulose insulation is enhanced, which makes the polarization current, complex permittivity and dielectric loss increase.

### Table 4. Volume, total dipole moment and polarizability of different models

| Model              | Volume ($Å^3$) | Total dipole (D) | Polarizability (D/Å³) |
|--------------------|---------------|-----------------|-----------------------|
| Cellulose + 1 % water | 10021        | 91.36           | $9.11 \times 10^{-3}$ |
| Cellulose + 3 % water | 9923         | 106.26          | $10.71 \times 10^{-3}$ |
| Cellulose + 5 % water | 10350        | 133.08          | $12.86 \times 10^{-3}$ |

3.4. Micromechanism analysis of influence of water content on breakdown property

The energy band structure of cellulose is a good description of the ability of cellulose to gain and lose electrons and the state density of cellulose can be used to analyze the electrical conductivity of cellulose. Figure 7 shows the results of energy band and state density of cellulose under different water content. As shown in figure 7(a), with the increase of water content in cellulose, the LUMO level of cellulose gradually decreased, while the HOMO level gradually increased. This phenomenon indicates that the ability of cellulose to gain or lose electrons increases with the increase of water content in the cellulose, leading to more ionization and breakdown.

On the other hand, it can be seen from figure 7(b) that the increase of water content of cellulose resulted in narrow forbidden band gap width, which was 4.885, 4.718 and 4.712, respectively. Water, as a highly polar substance, enhances the electrical conductivity of cellulose. Therefore, as the moisture content increases, the breakdown voltage of cellulose decreases.

![Figure 7. The energy band and density of state of cellulose with different moisture contents](image-url)
Figure 8 illustrates the electrostatic potential of cellulose with different moisture contents. The red is the positive electrostatic potential and the blue is the negative electrostatic potential. In the model with 1% water content of cellulose, there are more positive electrostatic potential parts. These parts have a large electrostatic effect on the electrons, trapping them and preventing them from taking part in the breakdown. As the water content increases, these red parts become less and less, and the cellulose's ability to bind electrons becomes less and less, leading to an increase in the breakdown voltage.

4. Conclusions
The curve of the real part of relative permittivity gradually moves to the upper right with the increase of the moisture content, while the imaginary part of relative permittivity move in the direction of high frequency. With the increase of water content, the breakdown voltage of pressboard presents a trend of decreasing. The result of molecular simulation shows that moisture increased the free volume in cellulose and increased the conductance loss of cellulose. At the same time, water and cellulose form a large number of hydrogen bonds, which reduces the stability of the cellulose itself and makes the cellulose more prone to polarization under the action of electric field. On the other hand, with the increase of water content in the cellulose, the ability of cellulose to gain or lose electrons increases, the forbidden band declines and the electrostatic potential of cellulose decreases, leading to more ionization and breakdown.

References
[1] Seytashmehr A., Fofana I., Eichler C., Akbari A., Borsi H., Gockenbach E. (2008) Dielectric spectroscopic measurements on transformer oil-paper insulation under controlled laboratory conditions. IEEE Trans. Dielectr. Electr. Insul. 15: 1100-1111.
[2] Zhang Y., Liu J., Zheng H., Wei H., Liao R. (2017) Study on quantitative correlations between the ageing condition of transformer cellulose insulation and the large time constant obtained from the extended debye model energies 10: 1842, 2017.
[3] Yang L., Zou T., Deng B., Zhang H., Mo Y., Peng P. (2019) Assessment of oil-paper insulation aging using frequency domain spectroscopy and moisture equilibrium curves. IEEE Access 7: 45670-45678.
[4] Mu Z., Yang Y., Wang Z. Effect of moisture on frequency domain spectroscopy (FDS) for oil-impregnated-paper insulation. In: 2018 IEEE 2nd International Conference on Dielectrics. Hungary. pp. 1-4.
[5] Feng D., Hao J., Yang L., Liao R., Chen X., Li J. (2020) Comparison of AC breakdown characteristics on insulation paper (pressboard) immersed by three-element mixed insulation oil and mineral oil. High Voltage 5: 298-305.
[6] Li G., Hao J., Li S., Liao R., Zhao X., Yang L. (2017) AC breakdown and frequency dielectric response characteristics of the mixed oil-paper insulation with different moisture content. In: 2017 1st International Conference on Electrical Materials and Power Equipment. pp. 478-481.
[7] Mo Y., Yang L., Hou W., Zou T., Huang Y., Zheng X., Liao R. (2019) Preparation of cellulose insulating paper of low dielectric constant by OAPS grafting. Cellulose 26: 7451-7468.

[8] Liao R., Zhao M., Yang L., Zhou X., Gong C. (2008) Molecular dynamics study of water molecule diffusion in oil–Paper insulation materials. Phys. Rev. Lett. 406: 1162-1168.