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Diffusion Mechanisms of Dissolved Gases in Transformer Oil Influenced with Moisture Based on Molecular Dynamics Simulation

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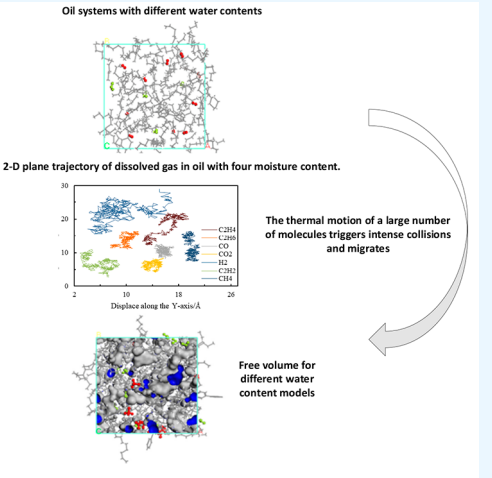
ABSTRACT: Dissolved Gas Analysis (DGA) of insulating oil is widely used for diagnosing transformer incipient faults. Moisture is a major contaminant and degradation byproduct of transformer insulating oil. In this paper, molecular dynamics simulation was used to study the influence of moisture on the diffusion movement of dissolved gases in the insulating oil. Cycloalkanes ($C_{20}H_{42}$), alkanes ($C_{20}H_{38}$), and aromatic hydrocarbons ($C_{20}H_{26}$) are selected as the basic structural units in the constructed transformer oil simulation system. 0%, 1%, 3%, and 5% moisture molecules are added to insulating oil, respectively, and the insulating oil generates seven kinds of gas molecules through cracking. With an anhydrous model used as a benchmark, we researched the diffusion trajectory, the diffusion coefficient (D), free volume (V_F), and the moisture−gas interaction energy of each gas molecule as a function of moisture content. Through this study, we found that the increase of moisture content enlarges the V_F value of dissolved gas in insulating oil, which makes the gas in oil easier to diffuse. Besides, the moisture can also alter the mean square displacement (MSD) of dissolved gases; the mutual energy of molecules is mainly affected by the electrostatic interaction energy. This study can contribute to a better understanding of the influence of different moisture contents on the diffusion movement of dissolved gas in transformer oil from the micro level.



1. INTRODUCTION

The power transformer is the electrical equipment used to transform alternating current (AC) voltage and current and transmit AC energy.[1,2] The safe and stable operation of a power transformer plays a significant role in modern power systems.[3] Generally, insulating oil makes up the internal insulation system of a transformer; their state affects the service life of the transformer directly.[4−7] In the long term operation, a transformer will be affected by various stresses such as electrical stress, thermal stress, and mechanical stress. Insulation oil will be aging and decomposing gradually, produce dissolved gas in oil, and even lead to thermal failure or electrical failure of the transformer.[8−10] H_2, CH_4, C_2H_2, C_2H_4, C_2H_6, CO, and CO_2 will be produced in oil after the transformer fault, which will diffuse in the transformer oil.[11−13] In addition, the types and proportions of dissolved gases in oil are also related to temperature and failure degree.[14] Therefore, the gas volume in insulating oil can be analyzed by the Dissolved Gas Analysis (DGA) method to evaluate the insulation aging degree and diagnose the fault type of the transformer.[15,16]

Traditional insulation aging and transformer failure testing methods can only study observable surface phenomena but cannot explain objective facts from a microscopic perspective.[17−19] Molecular simulation can not only analyze the static characteristics but also study the dynamic behavior of molecules.[20−22] The research methods used in this paper can also make up for the defect that traditional experiments can only analyze the problem from a visual perspective. In recent years, molecular simulation technology has made many important achievements in the aging of transformer insulating oil. At the

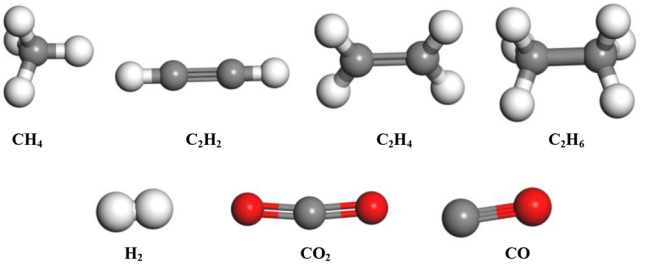


Figure 1. Small gas (CH_4, C_2H_2, C_2H_4, C_2H_6, H_2, CO_2, and CO) molecule model.

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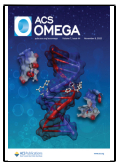

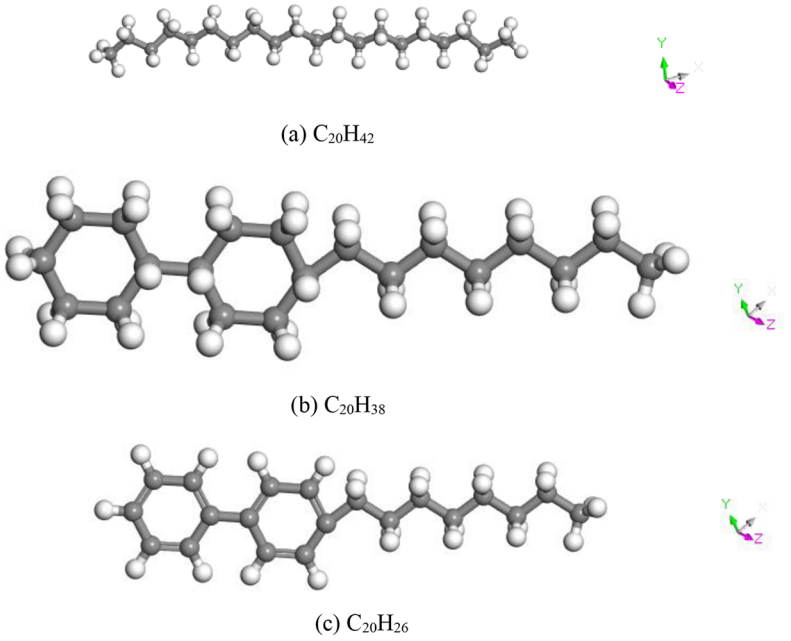

Figure 2. C_{20} monomer model.

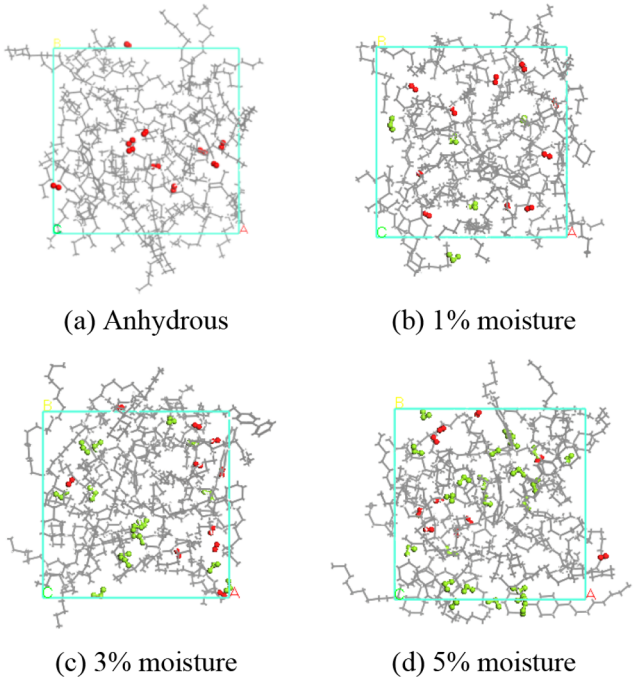

Figure 3. Transformer oil system with different moisture contents.

same time, great progress has been made in the study of molecular diffusion mechanisms from the microperspective.[23] For example, Koirala et al. studied the water diffusion in mixed insulating oil at different temperatures by molecular dynamics simulation.[24] Wang et al. studied the structures of graphene-reinforced epoxy coatings and the dynamic diffusion of guest water.[25] Zhu et al. investigated the adsorption and diffusion of methane and liquid water in the mesopores of coal.[26] Yang et al. studied the effect of water volume on methane adsorption capacity under high pressure.[27] Urbina et al. studied rotational diffusion in methanol by means of molecular dynamics simulations at several temperatures.[28] He et al. studied

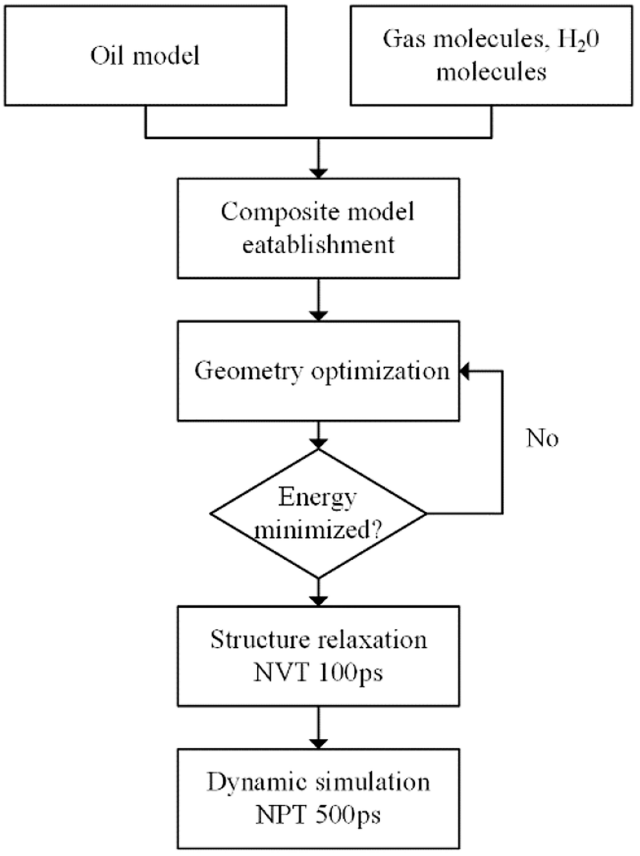

Figure 4. Flowchart for the simulation.

tortuosity of the kerogen pore structure to gas diffusion at molecular and nano scales by molecular dynamics simulation.[29] Zhou et al. studied the effect of the concentration and the size of the silica particles on diffusion coefficients of gases and the changes in Free Volume (V_F) and translational dynamics and intermolecular energies.[30] Mozaffari et al. studied molecular

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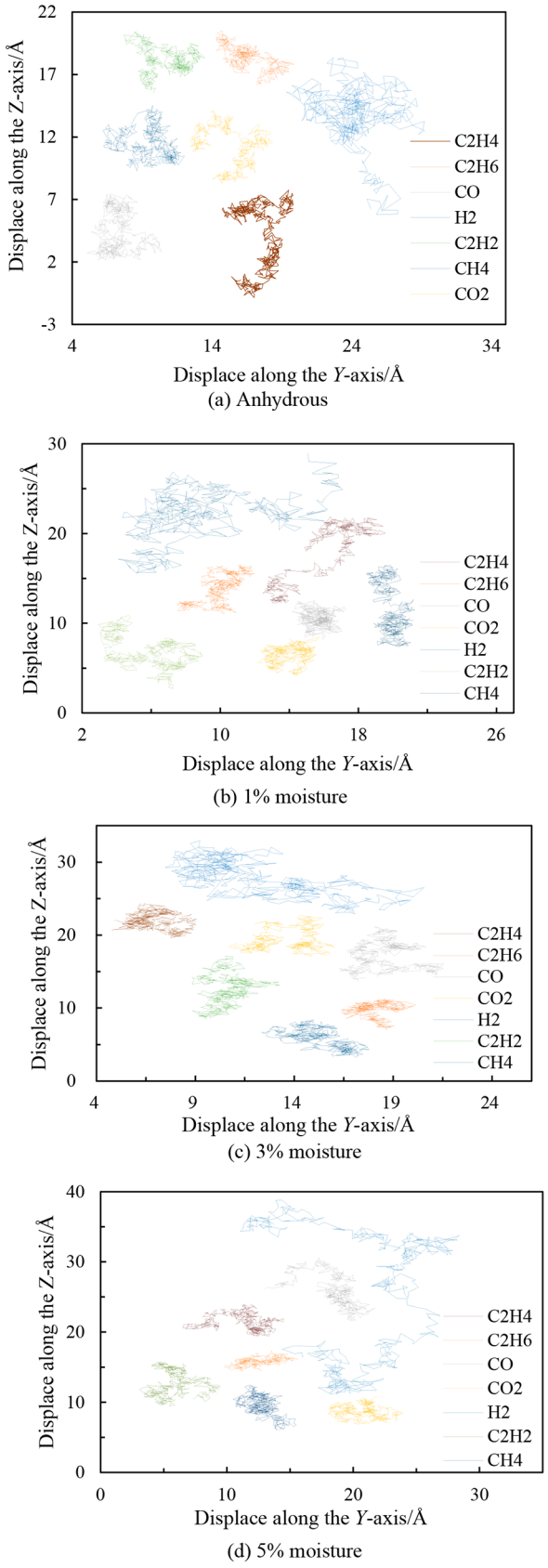

Figure 5. 2-D plane trajectories of seven gas molecules are in transformer oil with moisture contents of 0%, 1%, 3%, and 5%.

dynamics simulation of diffusion and permeation of gases in polystyrene.[31]

Although the diffusion activities have been explored by some experiments, the diffusion mechanisms on a molecular and atomic basis have still not been fully studied. The diffusion movement between molecules needs to be explained at the microlevel. Consequently, this paper reports on studies on the diffusion characteristics of dissolved gases in insulating oil. It is meaningful that findings from molecular dynamics simulations could be utilized to study diffusion properties and mechanisms, by which the obtained result is consistent with the actual experiments.

In this paper, Materials Studio (MS) software is used to carry out the molecular dynamic simulation of the composite material model. The mean square displacement (MSD), diffusion coefficient, free volume, and interaction energy were studied to reflect the distribution and diffusion of dissolved gas in the oil. The present findings are expected to provide support for studying the diffusion mechanism of dissolved gas in oil, which is valuable for evaluation of the aging condition of oil insulation.

2. SIMULATION MODEL

2.1. Establishment of Small Gas Molecule Model. During the long-term operation of the transformer, the insulating oil system will be decomposed into small molecule gases, such as CH_4, C_2H_2, C_2H_4, C_2H_6, H_2, CO_2, and CO. In this manuscript, MS was used to establish the seven small molecule models, as shown in Figure 1.

2.2. Construction of Transformer Oil Model. Transformer oil is a mixture of various hydrocarbons. C and H are two elements that accounted for 95% to 99% of the total weight.[32] The main hydrocarbons are cycloalkanes ($C_{20}H_{42}$, proportion more than 50%), alkanes ($C_{20}H_{38}$, the proportion is about 10−40%), and aromatic hydrocarbons ($C_{20}H_{26}$, the proportion is about 5−15%). After reviewing the literature, $C_{20}H_{42}$, $C_{20}H_{38}$, and $C_{20}H_{26}$ are selected as the basic structural units in the constructed transformer oil simulation system. The Amorphous Cell module in MS is used to construct three kinds of C_{20} alkane monomers, as shown in Figure 2.

Model of transformer oil containing 30 molecules at a ratio of 6:3:1. The linear model and the baseball model were used to represent the transformer oil, gas, and H_2O molecules (red represents H_2 molecule; green represents H_2O molecule), respectively. This paper takes the mixture system containing 10 H_2O molecules as an example to construct a transformer oil model without moisture, 1%, 3%, and 5% moisture, respectively. The oil density was set to 0.9 g/cm^3, and the side length of the box was 25.1 Å. The simulation model is shown in Figure 3.

3. MOLECULAR DYNAMICS SIMULATION METHODS

The periodic mineral oil model is established, and the initial density is set to 0.3 g/cm^3. All the simulation steps contain three parts, including model optimization, structure relaxation, and molecular dynamics. For model optimization and molecular dynamics, all force fields were based on the Polymer Consistent Force Field (PCFF). The Steepest Descent optimization algorithm was selected, the cutoff radius was set to 12.5 Å, and the model was optimized based on the electrostatic interaction and van der Waals algorithm by group. After these steps, the constant-pressure, constant-temperature (NPT) ensemble was selected for 100 ps internal relaxation with 0.01 GPa pressure, so that the entire system could achieve a stable

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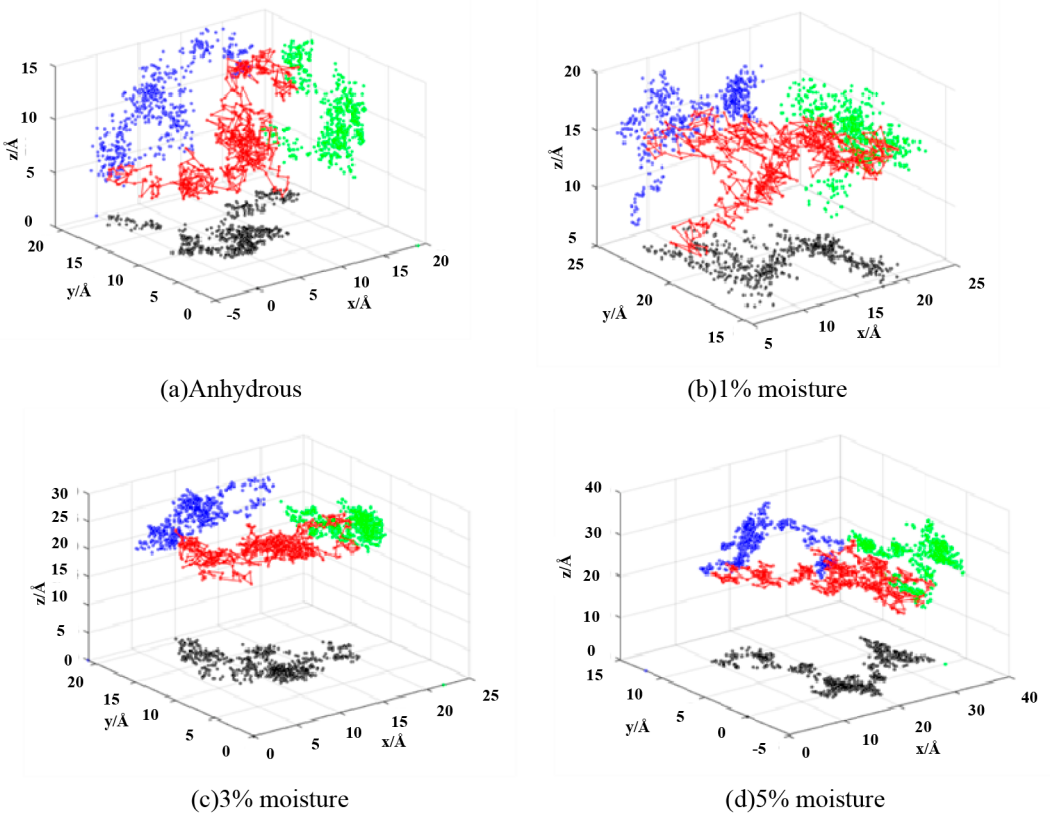

Figure 6. 3-D diffusion trajectory of H_2 in transformer oil systems with four moisture contents.

Table 1. Molecular Weight of Each Dissolved Gas in Oil

gas type	H_2	CH_4	C_2H_2	C_2H_4	C_2H_6	CO	CO_2
molecular weight	2.01	16.04	26.4	28.06	30.07	28.01	44.00

state, and the temperature was set to the normal operating temperature (343 K) of the transformer. Then, the constant-volume, constant-temperature (NVT) ensemble was selected for the 500 ps molecular dynamics simulation of the oil model, the first 100 ps was used for the equilibrium of the system structure, and the last 400 ps was used for the molecular dynamics calculation of the system. A frame was output every 1000 steps, with a total of 500 data points. The Smart optimization algorithm was adopted: the resulting unit cell edge length was 21.4 Å and the insulating oil system density was 0.85 g/cm^3. Periodic boundary conditions are used throughout the simulation; the simulation flowchart is shown in Figure 4.

4. SIMULATION RESULTS AND DISCUSSION

4.1. Diffusion Trajectory. From a microscopic point of view, molecular diffusion is a migration phenomenon caused by intermolecular interactions; temperature makes molecules move randomly, resulting in collisions;[33] molecular dynamics simulation can intuitively reflect the diffusion behavior of molecules. In this paper, the diffusion trajectory of dissolved gas in oil on the *YZ* plane is extracted by a self-compiled Perl script; different colors represent the diffusion trajectories of different molecules, as shown in Figure 5. In order to avoid the overlap of the trajectory of dissolved gas in oil, the diffusion trajectory is reasonably translated; this is overlapping in the actual movement.

There are significant differences in the diffusion trajectories of various characteristic gas molecules; the results of the diffusion trajectory show that the moisture content in transformer oil is positively correlated to the range of gas diffusion trajectory in oil. Among the seven gas molecules, the diffusion motion of the H_2 molecule is the most intense, and the range of motion trajectory is the largest; there are many examples of jump-style diffusion, displaced along the *Y*-axis increasing from 10 to 24 Å, with randomness of the diffusion trajectory. This is because H_2 has the smallest molecular weight and is less constrained by its mass. In comparison with H_2, the movement range of CH_4, C_2H_2, C_2H_4, C_2H_6, CO, and CO_2 is relatively concentrated.

To visually display the diffusion trajectory of molecules in transformer oil systems with different moisture contents, with H_2 as an example, the 3-D diffusion trajectory of H_2 in a transformer oil model with four moisture contents is given, as shown in Figure 6: red is the diffusion trajectory of the 3-D plane. Blue, green, and black represent trajectory projections on the *XZ*, *YZ*, and *XY* planes, respectively. The results show that with the increase of moisture content in the transformer oil system, the maximum movement displacement range of H_2 on the *X* axis increases from 16 to 25 Å, that on the *Y* axis increases from 13 to 25 Å, and that on the *Z* axis increases from 15 to 30 Å.

Discussion of Diffusion Trajectory. The conclusion accords with Graham's law of diffusion: at the same temperature and pressure, the diffusion range of various gases is inversely proportional to the square root of the gas weight.[34] The

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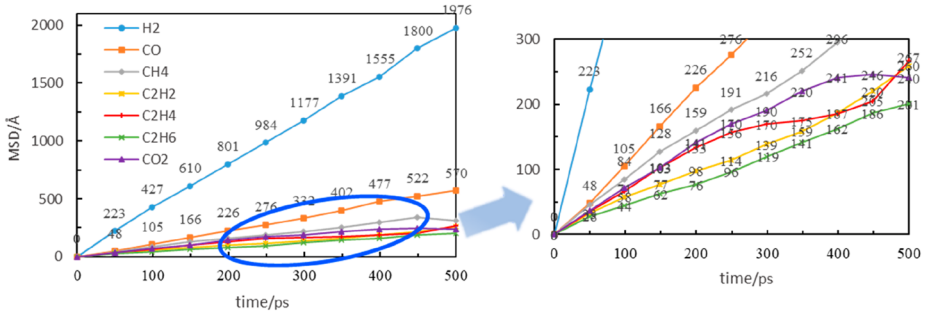
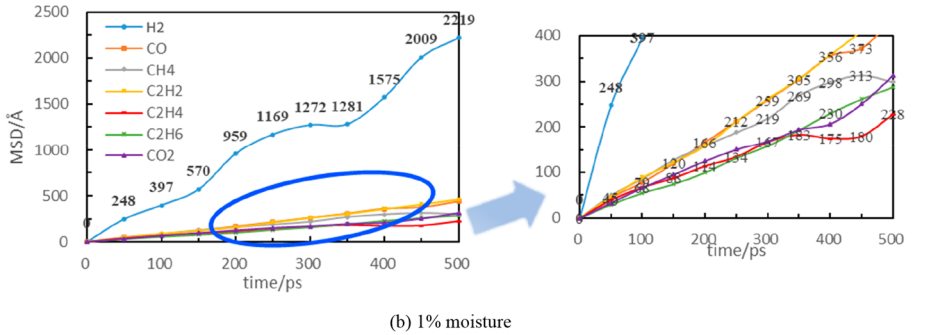
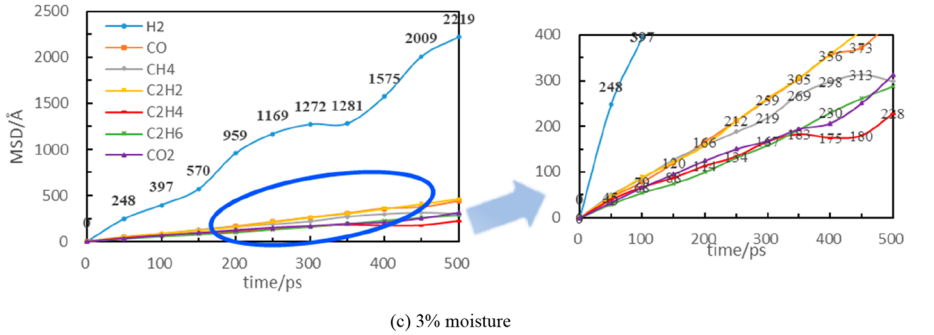


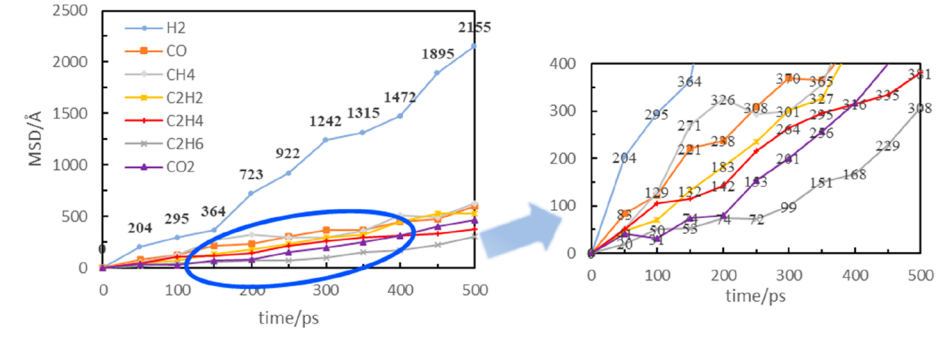
(a) Anhydrous



(b) 1% moisture



(c) 3% moisture



(d) 5% moisture

Figure 7. MSD curve of dissolved gases in oil with four moisture contents.

Table 2. Slope of Mean Square Displacement Curve Value ($cm^2\ s^{-1}$)

	H_2	CO	CO_2	CH_4	C_2H_4	C_2H_6	C_2H_2
without moisture	3.8645	0.7639	0.5337	0.6951	0.4431	0.3952	0.4813
1% moisture	3.9632	0.9175	0.4945	0.7074	0.431	0.5462	0.8381
3% moisture	5.3578	1.0419	0.7676	0.9523	0.4635	0.5785	0.8856
5% moisture	7.2068	1.2418	0.8567	1.1143	1.0022	0.5968	1.2159

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Table 3. Fitting Relation Coefficient Value ($cm^2\ s^{-1}$)

	H_2	CO	CO_2	CH_4	C_2H_4	C_2H_6	C_2H_2
Without moisture	0.9944	0.9599	0.9387	0.984	0.933	0.9853	0.9819
1% moisture	0.9695	0.9945	0.9786	0.989	0.973	0.9883	0.938
3% moisture	0.9553	0.9126	0.9733	0.9463	0.9316	0.8523	0.9945
5% moisture	0.9776	0.9918	0.9504	0.9463	0.9647	0.9572	0.9703

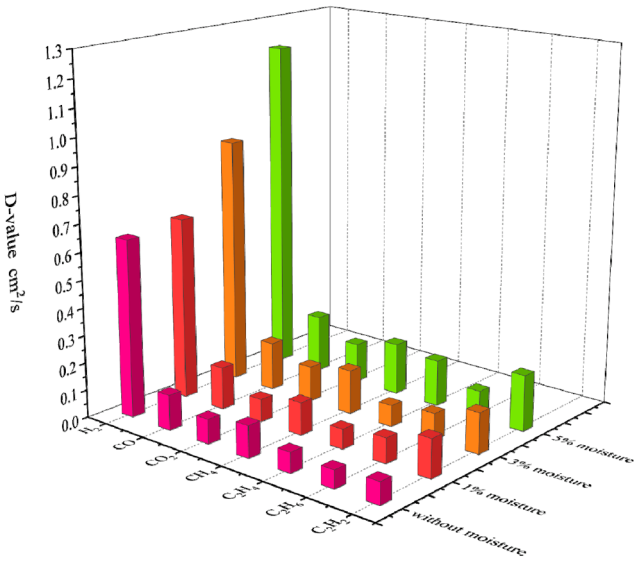

Figure 8. Diffusion coefficient D of dissolved gases in oil with four moisture content.

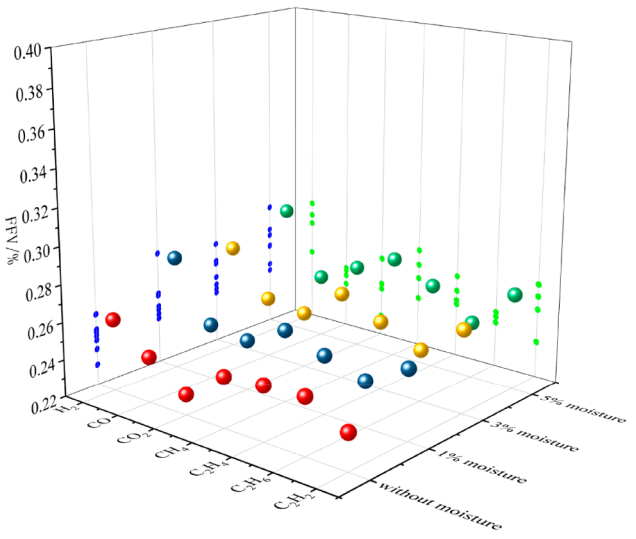

Figure 9. Free of Volume Fraction of seven gas molecules in transformer oil with different moisture contents.

molecular weight of each gas molecule is shown in Table 1. It shows that H_2 has the smallest weight, followed by CH_4 < C_2H_2 < CO < C_2H_4 < C_2H_6 < CO_2.

4.2. Diffusion Coefficient. The Mean Square Displacement (MSD) is the positional deviation of a particle relative to a reference position over time and is an important parameter to characterize diffusion. MSD is defined as an average of time geometry, as shown in eq 1.[35]

$$\mathrm{MSD} = \langle |x(t) - x(0)^2| \rangle = \frac{1}{N}\sum_{i=1}^{n} |x^{(i)}(t) - x^{(i)}(0)|^2 \quad (1)$$

where N is the total number of molecules in the whole transformer oil system, and $x^{(i)}(t)$ and $x^{(i)}(0)$ are the position of the first atom at time t and time 0, respectively.

The diffusion coefficient is a physical quantity that describes the degree of gas diffusion. The diffusion coefficient can be indirectly calculated by MSD; that is to say, the diffusion coefficient is 1/6 of the slope k of the MSD curve. Taking any point in the simulation as the starting point of the data, the diffusion coefficient D is calculated according to the Einstein relation, as shown in eq 2:

$$D = \frac{1}{6N}\lim_{t\to\infty}\frac{\mathrm{d}}{\mathrm{d}t}\sum_{i=1}^{N}\{[\vec{r}_i(t) - \vec{r}_i(0)]^2\} \quad (2)$$

where D is the diffusion coefficient of gas molecules. Since the MSD curve takes the average number of diffusion atoms N, the diffusion coefficient D can be simplified as in eq 3:

Table 4. Occupied Volume (Å)

	H_2	CO	CO_2	CH_4	C_2H_4	C_2H_6	C_2H_2
Without moisture	14481.99	14422.95	14708.08	14566	14223.35	14357.53	14292.97
1% moisture	14044.91	14499.05	14053.83	14016.71	14235.52	14422.95	14162.82
3% moisture	16770.26	14191.11	14248.29	14629.7	14837.08	14941.55	14720.08
5% moisture	16997.23	14080.83	14659.08	14224.39	14560.76	14357.53	14698.67

Table 5. Free Volume (Å)

	H_2	CO	CO_2	CH_4	C_2H_4	C_2H_6	C_2H_2
Without moisture	5053.45	4669.96	4419..42	4778.24	4721.15	4795.9	4504.27
1% moisture	5490.53	4825.23	4610.9	4890.34	4781.7	4669.96	4913.09
3% moisture	7752.55	4796.3	4750.88	5312.75	5131.04	4922.4	5299.45
5% moisture	7923.59	4795.9	5281.49	5389.02	5254.65	4795.90	5474.63

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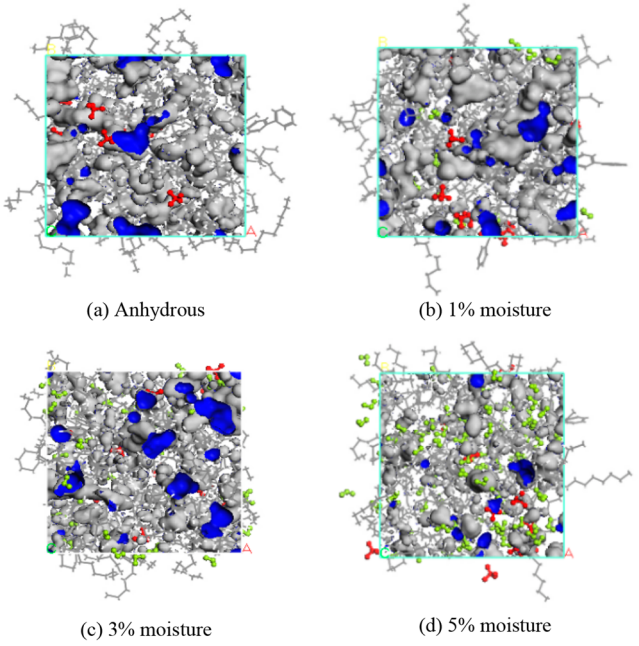

Figure 10. Free volume of different water content models.

$$D = \frac{k}{6}D = \frac{1}{6N}\lim_{t\to\infty}\frac{\mathrm{d}}{\mathrm{d}t}\sum_{i=1}^{N}\{[\vec{r}_i(t) - \vec{r}_i(0)]^2\} \quad (3)$$

Figure 7 is the MSD curve of dissolved gas in oil with four moisture contents; because the mean square displacements of the H_2 molecule in the mixed oil system with the moisture contents of 3% and 5% is too large, 1/2 of the actual value is drawn. As shown in Figure 7, significant differences in the MSD curves of seven gases can be found: the MSD of H_2 is the largest and that of C_2H_6 is the smallest. The MSD curve of molecules in an anhydrous system is approximately linear, and the MSD curves of each molecule at 1%, 3%, and 5% moisture content are tortuous and nonlinear; the addition of moisture intensifies the intermolecular motion, thus enhancing the molecule diffusion performance, with relatively significant effects on H_2 molecules.

The data of 50−450 ps are selected to calculate the diffusion coefficients of the seven gases because of the good linearity, and the slope k and fitting correlation coefficient R^2 are obtained by linear fitting; the data for k and R^2 are shown in Tables 2−3. R^2 is above 0.9, indicating that the results have high reliability. The D of dissolved gas in oil is calculated, and the data for D are shown in Figure 8.

According to the molecular structure and properties of the gas, the seven gases can be divided into 3 categories, the first category is elemental, including H_2; the second category is oxides, including CO and CO_2; the third category is hydrocarbon gases, including CH_4, C_2H_4, C_2H_6, C_2H_2. In the transformer oil model with four moisture content, the diffusion coefficient of H_2 is greater than that of any other molecule. In oil with the same moisture content, the larger the molecular weight, the smaller the diffusion coefficient is; there is a negative correlation between molecular weight and diffusion coefficient. In the hydrocarbon gas and oxide gas, the order of diffusion coefficient is $CH_4 > C_2H_2 > C_2H_4 > C_2H_6$ and $CO > CO_2$.

Since the diffusion of the gas molecules is disordered, the arrangement of gas molecules with little difference in the diffusion coefficient appear certainly different: the diffusion coefficients with moisture 1% are arranged in the order $H_2 > CO > C_2H_2 > CH_4 > CO_2 > C_2H_6 > C_2H_4$, but the diffusion coefficients with moisture 3% are in the order $H_2 > C_2H_2 > CH_4 > CO > CO_2 > C_2H_6 > C_2H_4$.

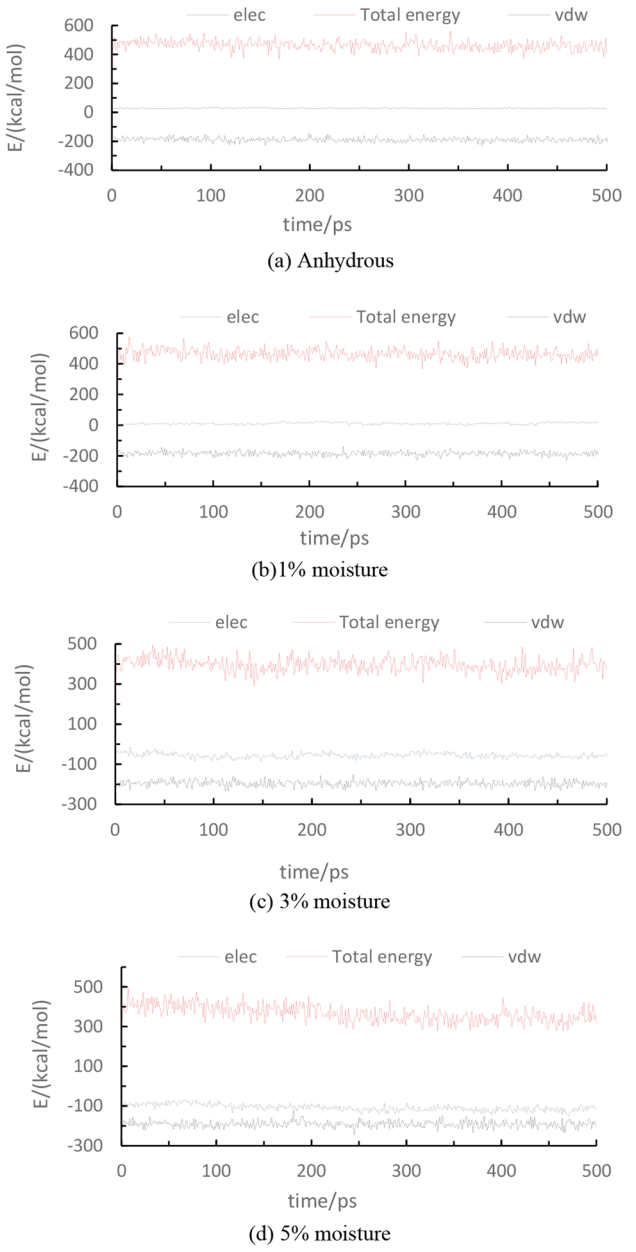

Figure 11. Interaction energy of H_2 in transformer oil systems with different moisture contents.

Discussion of Diffusion Coefficient. Moisture increases the spatial evacuation of transformer oil and increases the disorder and uncertainty in the diffusion of gas molecules. The diffusion ability of H_2 molecules is stronger than that of other gas molecules, indicating that moisture has the greatest influence on H_2. On the other hand, by comparing the same gas molecule in transformer oil with different moisture contents, moisture and diffusion coefficient are positively correlated: the higher the moisture content, the greater the diffusion coefficient. However, the coefficient change is not obvious, which is similar to the MSD description.

4.3. Free Volume. V_F theory is proposed by Cohen et al. and developed by Duda et al. It is of great significance in studying the

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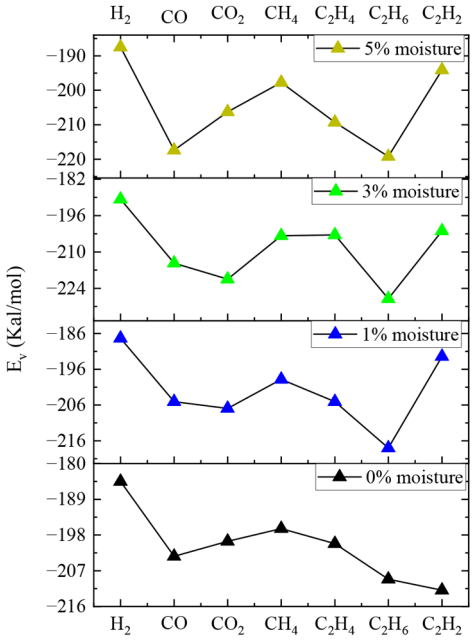

Figure 12. Van der Waals interaction energy (Kal/mol).

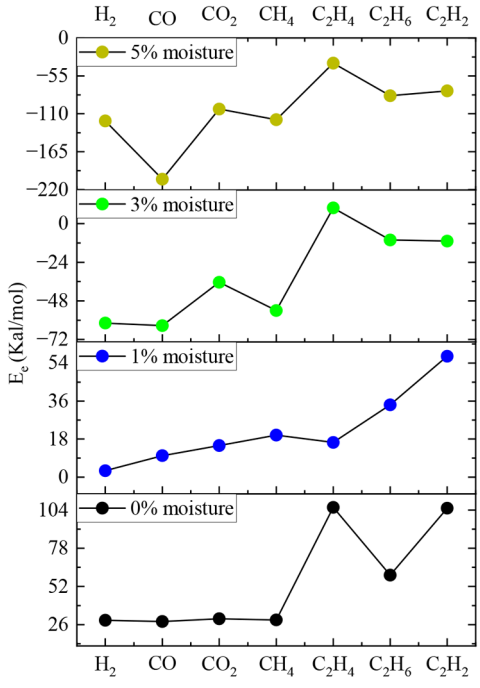

Figure 13. Electrostatic interaction energy (Kal/mol).

diffusion phenomenon in simple polymer systems.[36−39] Occupied Volume (V_O) and V_F constitute the Total Volume (V_T) of the polymer. Free of Volume is the ratio of V_F to V_T.[40] The V_O value is shown in Table 4, and the V_F value is shown in Table 5. In this paper, the Atom Volume and Surface tool is used to calculate the V_F of various gas-insulating oil mixture models at 343 K, and the Connolly surface method is used to quantitatively calculate the V_F of each system. Figure 9 is the Free of Volume Fraction of seven gas molecules in transformer oil with different moisture contents. To compare the value of the Free of Volume Fraction, the Free of Volume Fraction value is projected onto *XZ* and *YZ* axes (blue and green represent value projections on the

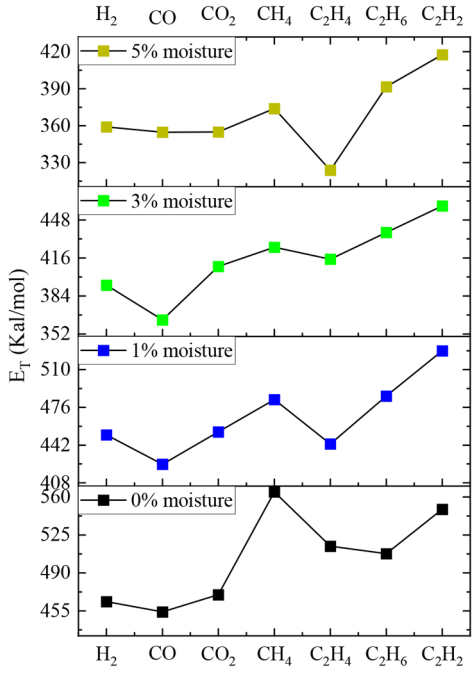

Figure 14. Total energy of the whole mineral oil mixing system (Kal/mol).

XZ and *YZ* planes, respectively). The results show a positive correlation between the Free of Volume Fraction and the moisture content; after adding moisture to the transformer oil, the Free of Volume Fraction increases in the model, but the change is not obvious.

A positive correlation exists between Free of Volume Fraction and moisture content; the increase in moisture content makes the Free of Volume Fraction value larger. This is because the increase of the polarity strength of transformer oil is small, and the bonding ability of moisture is not large enough, which is low spatial evacuation; the V_F change in the polymer system is small.

Due to space limitations, this paper only gives examples of the V_F of CH_4 in transformer oil systems with four moisture contents. As shown in Figure 10, the V_T of transformer oil is the sum of the V_O and the V_F (V_O by the transformer insulating oil molecules and the V_F is formed around the molecules due to thermal fluctuations). The V_F is divided into gap free volume and holes free volume. When the redistributed volume needs to provide energy, gap free volume turns into holes free volume. When the gas diffuses, the unoccupied volume is the holes free volume, and the redistribution of this part of the V_F does not need to provide energy. Effective gas diffusion occurs when enough free volume holes left by diffusion molecules are filled by another adjacent molecule.

Discussion of Free Volume. H_2O molecules are polar molecules, while transformer oil is a nonpolar substance. Due to polar attraction, most of the moisture molecules exist in the transformer oil. Moisture leads to the polarity of transformer oil increasing, which reduces the viscosity and increases the fluidity. This causes the diffusion barrier of the small molecule in the transformer oil to become smaller, which promotes the diffusion activities of small molecules.

4.4. Interaction Energy. The interaction energy between oil medium and gas small molecules is an important factor affecting the diffusion of gas in the oil.[41] The interaction energy obtained by simulation is composed of van der Waals Interaction

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Energy (E_v) and Electrostatic Interaction energy (E_e). Figure 11 is the curve of the interaction energy between H_2 and oil medium with time in different moisture contents. As shown in Figure 11, the interactions between different substances can fluctuate at a given value; the molecular diffusion process is dynamically balanced. The change waveform of the interaction energy between the six gases and the oil medium is similar to that of H_2. If $E = 0$, there is no interaction between the substances. If $E > 0$, there is a repulsive interaction. If $E < 0$, there is an attractive interaction. In all models, the E_T values are always positive numbers, which shows that the interaction of oil and gas molecules is repulsive. There is little difference in E_v of gas molecules, but the E_e has an obvious change with the increase of moisture content, which indicates that E_e plays a major role in the process of small molecule diffusion.

Figures 12−14 represent the interaction energy of seven kinds of gas molecules and oil medium. With the increase of moisture content, the E_e value changes from positive to negative, which indicates that the interaction changes from repulsive to attractive. If $E_e < 0$, the attraction between molecules increases, the diffusion of molecules should be restrained by the E_e effect, but the result is that the diffusion is exacerbated with the increase of moisture; the reason is that pure gas does not produce electrostatic effects. Due to the combined action of moisture and thermal motion, collisions and friction occur between molecules, resulting in the electrostatic interaction that occurs between H_2O molecules and oil, and this E_e easily to reaches saturation. Therefore, the total electrostatic action is manifested as repulsive.

Discussion of Interaction Energy. Van der Waals force is the function of distance and decays according to 12 times the distance. In the NVT system, the distance between the gas molecules and the oil medium will be shortened with the addition of moisture, which causes the van der Waals force to weaken and offset the effect of electrostatic force increase.

5. CONCLUSION

In this paper, for the dissolved gas in oil under four moisture conditions, the diffusion mechanism of dissolved gas in oil was analyzed from the aspects of diffusion coefficient, diffusion trajectory, free volume, and interaction energy. The present findings and analysis lead to the following conclusions:

(1) It can be observed from the diffusion coefficient of dissolved gas in oil and the trajectories of different planes that the diffusion trajectory of gas in oil is positively correlated with moisture content; the moisture promotes the diffusion of dissolved gas in oil, the greatest impact on the H_2 molecule.

(2) Polar moisture is added to nonpolar transformer oil so that the polarity of the moisture−oil medium increased. Moisture induces transformer oil medium space rarefaction, increases mobility, and reduces the viscosity. That further makes the diffusion obstacles smaller for dissolved gas in oil and increases the free volume. It provides the necessary conditions for the diffusion of dissolved gas in the oil.

(3) Due to thermal motion, collisions and friction occur between molecules, resulting in electrostatic interaction (the mutual energy of molecules is mainly affected by the E_e). However, the electrostatic interaction energy easily reaches saturation, and the van der Waals force weakens and offsets the effect of electrostatic force increase. In general, the interaction of oil and gas molecules is repulsive.

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Notes

The authors declare no competing financial interest.

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