Molecule diffusion behaviours of waterproof sealants into silicone rubber insulation for submarine cable joint based on molecular dynamics simulations

Zhenpeng Zhang\(^1\) | Heyu Wang\(^2\) | Zhonglei Li\(^2\) | Jiankang Zhao\(^1\) | Shaohua Wang\(^3\) | Wenlin Pan\(^4\) | Wenjie Li\(^1\) | Te Li\(^3\) | Boxue Du\(^2\)

\(^1\)China Electric Power Research Institute, Wuhan, China
\(^2\)Key Laboratory of Smart Grid of Ministry of Education, School of Electrical and Information Engineering, Tianjin University, Tianjin, China
\(^3\)State Grid Zhejiang Electric Power Co., Ltd., Hangzhou, China
\(^4\)Hengtong Submarine Power Cables Co., Ltd., Changsha, China

Abstract

The repair joints of high-voltage cables play a significant role in the urgent cable maintenance, by recovering cable conductor, insulation, shielding layer and waterproof layer especially for submarine cables. However, the molecule diffusion and permeation processes from waterproof sealant into the cable joint insulation occur, leading to the insulation ageing and deterioration. In this article, the molecule diffusion behaviours of six typical molecules from the asphalt-based and polyurethane-based waterproof sealants into silicone rubber insulation are investigated. The results show that the order of diffusion coefficients is as follows, phenanthrene > fluoranthene > pyrene > benzo [k] fluoranthene > indeno [1,2,3-cd] pyrene > polyurethane (PU). The relative molecular mass and binding energy are pivotal factors affecting the diffusion behaviour, while there is a linear relationship between the logarithm of the diffusion coefficient and the ratio of binding energy to relative molecular mass. Besides, the rising ambient temperature increases the fraction of free volume of silicone rubber and decreases the binding energy, thus resulting in a higher diffusion coefficient and an enhanced permeation process of the asphalt-based sealant. In summary, the PU-based waterproof sealant, with a much lower permeability than the asphalt-based material, has a potential application in the waterproof layer for high-voltage submarine cable repair joints.

1 | INTRODUCTION

High-voltage alternating-current (HVAC) submarine cables play a vital role in the cross-sea interconnection project, which transmits the power between the mainland, islands and offshore platforms [1–4]. The repair joint is a crucial component of the cable and has a significant effect on the rapid maintenance and emergency repair, ensuring the reliability of the operation, especially for submarine cables [5]. The accessories for HVAC 500 kV crosslinked polyethylene submarine cable in Zhoushan, China, have completed the type tests and pre-qualification tests [6]. The silicone rubber reinforced insulation of the submarine cable repair joint, which goes through the pre-qualification tests, occurs the discoloration phenomenon, leading to the insulation ageing and deterioration [6, 7]. The current researches declare that the discoloration is not only caused by the ageing of silicone rubber insulation in the electro-thermal coupling field but also attributed to the permeation process of small organic molecules from the organic waterproof layer into silicone rubber insulation. At present, there are still few studies on this molecule diffusion behaviour, whose mechanism is not yet clear and influencing factors are still unknown.
For submarine cable repair joints, the waterproof layer, between the outer semi-conductive layer and the metal shell, is filled with sealants. Due to the advantages of easy doping, high viscosity and low cost, the asphalt-based sealants are employed as the waterproof materials for submarine cable joints, which contains benzene, toluene, ethylbenzene, xylene, and polycyclic aromatic hydrocarbons with abundant hydrophobic groups [8]. Polyurethane (PU) is also an important waterproof material due to such excellent properties as good toughness, abrasion resistance and easy preparation [9]. Thus, asphalt-based and PU-based waterproof sealants are now widely used in the waterproof layer of submarine cable joints [6].

Molecular dynamics (MD), derived from molecular mechanics, is a simulation method that describes the potential energy surface of particles through empirical force fields and numerically solves Newton's differential equations of motion [10]. An extensive number of researches have proved that MD can accurately simulate the permeation and diffusion behaviour of molecules, such as gas, aromatic hydrocarbons or polymers [11, 12]. Liao et al. investigated the diffusion behaviour of water molecules in oil–paper insulation and found that the moisture in cellulose diffuses isotropically by analysing the interaction energy, radial direction functions and mean square displacement [13].

Zhu et al. studied the effect of water molecules on the mechanical properties of cellulose, and propose that the increasing moisture causes a decrease in cohesive energy density, leading to a decline in mechanical properties [14]. Zhou et al. simulated the thermal decomposition of insulating oil and proved that the rising temperature enhances the decomposition of the oil, leading to thermal ageing and faults [15]. Tang et al. researched the thermal stability of the insulation paper modified with polyphenylsilsesquioxane, which showed that the modified paper has better thermal properties due to the stronger bonds and the greater mutual permeability between the molecular chains [16]. Ye et al. focuses on the diffusion behaviour of gas molecules in mineral oil and natural esters and adopt the Pearson correlation coefficient to evaluate the correlation between diffusion coefficients and influencing factors. In addition, the Arrhenius equation is used to fit the diffusion coefficients varying with temperature, in which the activation energy is used to evaluate the temperature sensitivity of diffusion behaviour [17].

In this article, based on MD, the molecular diffusion processes of asphalt-based and PU-based waterproof sealant at different temperatures are simulated, in order to investigate the mechanism of permeation behaviour and the effect of molecular species and temperature on it. This article aims to compare and recommend a preferential waterproof sealant material with low permeability suitable for high voltage submarine cable repair joints.

2 | MD SIMULATION

2.1 | Molecular models establishment

Molecular models for simulation of silicone rubber insulation and waterproof sealant are established based on Materials Studio (MS) software. The silicone rubber model consists of methyl silicone rubber, which is shown in Figure 1. The formula of methyl silicone rubber is C$_{22}$H$_{126}$O$_{19}$Si$_{20}$, with a relative molecular mass ($M_r$) of 1497.15, which is obtained from MS software. Next, asphalt and PU, which are the two primary materials of waterproof sealant for submarine cable joints, are established, respectively [18]. The asphalt, consisting of asphaltene, saturates, naphthene aromatics and polar aromatics, is characterized by phenanthrene (Phe), fluoranthene (Flu), pyrene (Py), benzo [k] fluoranthene (BkF), and indeno [1,2,3-cd] pyrene (IcdP), which are abundant in asphalt [19–21]. The information of waterproof sealant molecular models is shown in Table 1.

An amorphous periodic cell of silicone rubber is built. Meanwhile, six molecular layers of asphalt-based and PU-based waterproof sealants are also built, including Phe, Flu, Py, BkF, IcdP and PU. Then, each molecular layer of asphalt or PU is combined with an amorphous periodic cell of silicone rubber to construct a waterproof sealant–silicone rubber double-layer structure model.

Because of the extremely high energy and local stress of the initial double-layer structure models, it has to perform some tasks to make their structures reasonable, such as geometry optimization, annealing and volume relaxation. The constant-pressure and constant-temperature (NPT) ensemble is selected for volume relaxation lasting 100 ps, making the simulation models in a state of equilibrium. After the above treatments, the waterproof sealant–silicone rubber double-layer structure models are constructed, which will be used for permeation process simulation.

2.2 | MD simulation method

The forcefield potential energy function is the foremost parameter in MD simulation. The polymer consistent forcefield (PCFF) has been proven to be suitable for MD simulation of polymers and organic molecules [13, 14, 17]. In this article, PCFF, whose potential energy surface is composed of bond stretching, bending, torsions, inversion, cross terms and non-bond energy, has been adopted to describe the potential energy of particles.

The diffusion behaviour simulation of waterproof sealant for submarine cable joints is performed based on the Dynamics task of Forcite module. The canonical (NVT) ensemble is selected to perform the simulation for 500 ps, with a constant volume and a constant temperature. In the simulation process of 500 ps, the first 100 ps are used to balance the system, and the last 400 ps are used to calculate the simulation results. During the simulation process of molecule diffusion behaviour, the Nose method is chosen to control the temperature at 0, 30, 60, 90 and 120°C [22, 23]. The initial velocity of particles satisfies the Boltzmann distribution. The Velocity Verlet integral algorithm is used for solving Newton's differential equations of motion [24].
2.3 MD simulation parameters

2.3.1 Free volume

Free volume is the unoccupied volume in the material, which provides voids for molecular thermal movement. The existence of free volume is a critical factor that enables small molecules to permeate or diffuse in it. Based on Fox and Florý's free volume theory [25], the total volume is composed of free volume and occupied volume. In order to evaluate the free volume, the fraction of free volume (FFV) is calculated to represent the proportion of free volume in the total volume, according to Equation (1),

\[
FFV = \frac{V_F}{V_F + V_O} = \frac{V_F}{V_T}
\]

where, \(V_F\) is the free volume, \(V_O\) is the occupied volume and \(V_T\) is the total volume.

2.3.2 Binding energy

Binding energy, which represents the attractive trend between two materials, plays a crucial role in molecular diffusion behaviour [26, 27]. Positive binding energy indicates that there is a tendency to attract between the particles. On the contrary,
The trajectories are extracted from each frame of the simulation system, and the projection on the plane shows the range of molecular diffusion. The trajectories are extracted from each frame of the simulation system based on the Perl script.

2.3.3 | Diffusion trajectory

The diffusion trajectory intuitively reflects the path of the molecular movement in the simulation system, and its projection on the plane shows the range of molecular diffusion. The trajectories are extracted from each frame of the simulation system based on the Perl script.

2.3.4 | Diffusion displacement

The diffusion displacement is the distance of particles from the initial coordinates at time \( t \), reflecting the strength of molecular diffusion, and is calculated by Equation (3),

\[
S(t) = \sqrt{\left| r(t) - r(0) \right|^2}
\]

where, \( S(t) \) is diffusion displacement at time \( t \), \( r(t) \) and \( r(0) \) are the coordinates at time \( t \) and 0, respectively.

2.3.5 | Mean square displacement

The mean square displacement (MSD), which is the average displacement of all particles at time \( t \), is a pivotal parameter to evaluate the diffusion behaviour of small molecules from waterproof sealant into silicone rubber. The MSD is measured over time to determine the statistical strength of molecular diffusion behaviour, and is expressed as Equation (4),

\[
MSD = \left\langle \left| \vec{r}_i(t) - \vec{r}_i(0) \right|^2 \right\rangle
\]

where, \( \vec{r}_i(t) \) and \( \vec{r}_i(0) \) are the position vector of particle \( i \) at time \( t \) and 0, respectively, and \( \left\langle \right\rangle \) means calculating the average for all particles.

2.3.6 | Diffusion coefficient

The diffusion coefficient, quantifying the diffusivity of particles, is also a key parameter to evaluate the diffusion behaviour of waterproof sealant molecules, which is derived from the MSD curve. According to the Einstein formula [29], the diffusion coefficient can be expressed by Equation (5)

\[
D = \frac{1}{6N} \lim_{t \to \infty} \frac{d}{dt} \sum_{i=1}^{N} \left[ \left( \vec{r}_i(t) - \vec{r}_i(0) \right)^2 \right] = \frac{a}{6}
\]

where, \( D \) represents the diffusion coefficient, \( N \) is the total number of particles, and \( a \) is the slope of the linear fitting of the MSD curve.

3 | RESULT

3.1 | Influence of molecular species on diffusion behaviour

3.1.1 | Free volume

According to the free volume theory, the free volume is a critical factor that can cause the permeation and diffusion behaviour of molecules, in which small molecules can be in motion. Based on the calculation of the simulation results, the silicone rubber has a free volume fraction of 30.84% at 30°C, as shown in Figure 2, where the blue part is the free volume (17194.75 Å), and the grey part is the occupied volume (38559.75 Å). It can be observed that the free volume is evenly dispersed in the silicone rubber model, suggesting that the volume relaxation of the silicone rubber model has been fully completed.

3.1.2 | Binding energy

The binding energy between waterproof sealant molecules and silicone rubber is calculated based on the Forcite module and Perl script, whose results are displayed in Figure 3. It could be seen that the binding energy of each molecule is positive, indicating that there is a binding force between the waterproof sealant and silicone rubber, and a tendency to attract each other. PU molecules have stronger binding energy than other molecules, meaning that it has greater non-bond energy with silicone rubber.

In order to facilitate the comparison of the interaction of various waterproof sealant molecules, the arithmetic mean of binding energy is calculated and shown in Figure 4. PU molecules have average binding energy greater than other molecules, which is 1853.24 kcal/mol. The order of average binding energy is as follows, PU > BkF > Flu > IcdP > Phe > Pyr. Compared with poly cyclic aromatic hydrocarbons, PU molecules are weaker in polarity, resulting in stronger binding energy with silicone rubber. Moreover, the N–H bond creates a hydrogen bond between PU and silicone rubber, which further
enhances the attraction. Therefore, compared with the asphalt-based, the PU-based material has a stronger binding force with silicone rubber, making it more difficult to overcome the energy barrier to permeate.

3.1.3 | Diffusion trajectory and displacement

Figure 5 presents the molecule diffusion of asphalt- and PU-based waterproof sealants in silicone rubber insulation at 0 and 500 ps. Figure 6 displays the diffusion trajectories of small molecules from 0 to 500 ps, where the red lines are three-dimensional diffusion trajectories, while the black, green, and blue lines are the projections of the trajectories on the X–Y, X–Z, and Y–Z planes. Meanwhile, the diffusion displacements of molecules are shown in Figure 7.

Based on the simulation model in Figure 5 and the diffusion trajectory in Figure 6, PU molecules are adsorbed on the surface of silicone rubber and not permeate deeper into silicone rubber, which is related to the tremendous binding energy. Besides, focussing on Figure 7, the diffusion displacement of the PU molecules does not tend to rise with time, which is always below 10 Å, while other aromatic molecules of asphalts have a trend to permeate deeper. In a word, PU-based waterproof sealant has a weaker diffusion behaviour into the silicone rubber insulation than that of asphalt-based waterproof sealant.

3.1.4 | MSD and diffusion coefficient

The MSD statistically evaluates the strength of molecular diffusion behaviour. In order to quantitatively evaluate the permeation behaviour of the waterproof sealant, the MSD curves are measured and shown in Figure 8, and the diffusion coefficients are calculated by Equation (5) and listed in Table 2.

The separation of each MSD curve is obvious, showing a trend of linear growth. The order of diffusion coefficients of small molecules from waterproof sealants is as follows, Phe > Flu > Pyr > BkF > IcdP > PU. It is indicated that PU has a smaller diffusion coefficient than any kind of small molecules of asphalt, proving that the permeation behaviour of PU-based waterproof sealant into silicone rubber is weaker and has less impact on silicone rubber insulation. PU, with a greater relative 

3.2 | Influence of temperature on diffusion behaviour

3.2.1 | Free volume

The free volume distributions in silicone rubber at 0, 60, 90, and 120°C are depicted in Figure 9. The FFV varying with temperature is presented in Figure 10. The FFV of silicone rubber increases linearly with temperature, which is related to the change in the density. Based on the free volume theory, the volume expansion of the material caused by heating is mainly contributed by the free volume [30]. The high temperature intensifies the thermal movement of silicone rubber molecules, which tends to expand the molecular distance. The higher FFV of silicone rubber, which means that small molecules have more space for free
FIGURE 5  Molecule diffusion of asphalt- and PU-based waterproof sealants in silicone rubber insulation at 0 and 500 ps. BkF, benzo [k] fluoranthene; Flu, fluoranthene; IcdP, indeno [1,2,3-cd] pyrene; Phe, phenanthrene; PU, polyurethane; Pyr, pyrene

FIGURE 6  Diffusion trajectories of small molecules at 30°C
movement, is more likely to cause small molecules to permeate and diffuse into it.

### 3.2.2 Binding energy

The binding energy between the various molecules in the waterproof sealant and the silicone rubber varying with temperature is exhibited in Figure 11. It is illustrated that the binding energy between the waterproof sealant and silicone rubber is positive, meaning that there is an attraction between them. And it is seen that as the temperature rises, the binding energy between the waterproof sealant and silicone rubber decreases so that the tendency to attract each other is weakened. The rising temperature enhances the random movement of molecules, indicating that the increase in internal energy leads to a decrease in binding energy. For the Phe, the binding energy is 1730.82 kcal/mol at 0°C, while it is 1426.61 kcal/mol at 120°C with a decrease of 17.58%. For the PU, it has a binding energy of 1964.50 kcal/mol at 0°C, and 1664.72 kcal/mol at 120°C, which decreases by 14.48%

Moreover, it can be seen from Figure 11 that Phe and Pyr have similar binding energy, while Flu, BkF and IcdP have another similar binding energy, which is related to the molecular structure. Phe and Pyr have a planar molecular structure, while Flu, BkF, and IcdP have a non-planar molecular structure, resulting in a more powerful polarity, so their binding energy with silicone rubber is greater.

### 3.2.3 Diffusion trajectory

Figures 12 and 13 display the diffusion trajectories of Phe and PU molecules at 0, 60, 90 and 120°C, respectively. Similarly, the red lines represent the three-dimensional trajectories, while the black, green and blue lines represent the projections of the trajectories on the X–Y, X–Z and Y–Z planes.

Based on Figure 12, it is indicated that the diffusion trajectories of Phe molecules become more dispersed at higher temperatures, whose diffusion behaviour is much intense. And Figure 13 is shown that the diffusion trajectories of PU molecules have no obvious change at different temperatures. After being adsorbed on the surface of silicone rubber, PU molecules do not permeate deeper into the silicone rubber.

To sum up, the diffusion behaviour of aromatic molecules from asphalt-based waterproof sealant into the depth of silicone rubber is much stronger than that of PU-based waterproof sealant.

### 3.2.4 MSD and diffusion coefficient

The MSD curves of Phe molecules at different temperatures are presented in Figure 14. The slope of the curves becomes steeper as the temperature rises, which means that the average displacement of the Phe molecule is greater, making a more powerful permeation process into silicone rubber insulation.
By measuring the MSD curves of various molecules at different temperatures, the diffusion coefficients of each molecule are calculated, as shown in Figure 15. The rising temperature does not affect the order of diffusion coefficients, which is always as follows, Phe > Flu > Pyr > BkF > IcdP > PU. For the Phe, the diffusion coefficient is 0.11216 Å²/ps at 0°C, while it is 0.41486 Å²/ps at 120°C with an increase of 269.88%. Meanwhile, for the PU, it has a diffusion coefficient of 0.02551 Å²/ps at 0°C, and 0.14366 Å²/ps at 120°C, which increases by 463.16%.

In general, at any temperature, the PU molecules always have a smaller diffusion coefficient than other aromatic molecules from asphalt, suggesting that the diffusion behaviour of PU-based waterproof sealants is much weaker than that of asphalt-based waterproof sealants, which is consistent with the diffusion trajectory analysis.
4 | DISCUSSION

4.1 Relationship between the diffusion coefficient, binding energy and molecular mass

Current researches have shown that relative $M_r$ and binding energy ($E_{\text{bind}}$) are key factors affecting molecular diffusion behaviour [17, 31]. In order to reveal the influence of molecular species on diffusion behaviour, the relationship between relative $M_r$, interaction energy, and diffusion coefficient needs to be established. Some studies described diffusion behaviour by using a single-factor model involving with $M_r$ or $E_{\text{bind}}$ and evaluated the relationship between the single-factor and the diffusion coefficient through the Pearson correlation coefficient [17, 32, 33]. Hao et al. investigated the correlation between diffusion coefficient and interaction energy and relative $M_r$, whose Pearson coefficient are 0.9488 and −0.8499, respectively [17]. Therefore, a multi-factor model needs to be established to describe the diffusion behaviour of waterproof sealant molecules.

In this article, a single-exponential equation is proposed to reveal the relationship between $M_r$, $E_{\text{bind}}$ and diffusion coefficient, which is expressed as Equation (6),
\[ D = A \cdot \exp \left( B \cdot \frac{E_{\text{bind}}}{M_r} \right) \tag{6} \]

where, \( A \) and \( B \) are the fitting parameters. According to Equation (6), the \( D \cdot (E_{\text{bind}}/M_r) \) curves at different temperature are fitted, whose results are displayed in Figure 16.

Based on Figure 16, it is suggested that the molecular diffusion coefficient is affected by the binding energy and relative \( M_r \), while there is a linear relationship between the ratio of binding energy to relative molecular mass \( (E_{\text{bind}}/M_r) \) and the logarithm of the diffusion coefficient \( \ln D \). Thereby, the Pearson correlation coefficient, which is calculated by Equation (7), is adopted to describe the linear correlation between \( \ln D \) and \( E_{\text{bind}}/M_r \).

\[ \rho = \frac{\sum (P - \overline{Q})(Q - \overline{Q})}{\sqrt{\sum (P - \overline{Q})^2 \sum (Q - \overline{Q})^2}} \tag{7} \]

where, \( \rho \) is the Pearson correlation coefficient, \( P \) and \( Q \) are two sets of variables. The Pearson correlation coefficients between \( D \) and \( E_{\text{bind}}/M_r \) at different temperatures are listed in Table 3.

The absolute values of all correlation coefficients in Table 3 are greater than 0.96, which proves that there is a tremendous linear correlation between the two variables, indicating that the single-exponential equation multi-factor model can describe the relationship between relative \( M_r \), \( E_{\text{bind}} \) and diffusion coefficient.

### 4.2 Relationship between the diffusion coefficient and temperature

In order to evaluate the effect of temperature on the diffusion behaviour of waterproof sealants, a function, which can describe the relationship between the temperature and the diffusion coefficient, needs to be investigated. The Arrhenius equation describes the relationship between the reaction rate and temperature, which can also characterize the relationship between diffusion coefficient and temperature.
between temperature and diffusion coefficient based on current research [34]. Arrhenius equation is expressed by Equation (8),

$$\ln D = \ln C - \frac{1000E_a}{RT}$$  \hspace{1cm} (8)$$

where, $D$ is the diffusion coefficient, $T$ is the Kelvin temperature, $R$ is the molar gas constant whose value is 8.314 J/(mol·K), $C$ is the pre-exponential factor, and $E_a$ is the activation energy that describes the sensitivity of the diffusion behaviour to temperature.

Based on Equation (8), a non-linear fitting of the diffusion coefficient–temperature curve is performed, whose result is shown in Figure 17. And Table 4 lists the pre-exponential factor $A$ and the diffusion activation energy $E_a$. The order of the activation energy of various molecules is as follows, PU > IcdP > BkF > Pyr > Phe > Flu, which is almost inverse to the order of diffusion coefficient, except for Phe and Flu. The PU molecule has the largest activation energy, suggesting that its diffusion coefficient is the most sensitive to temperature. However, the high sensitivity to temperature does not affect the lowest diffusion coefficient of PU molecules at any temperature, which is inferred that PU-based waterproof sealant has a weaker permeation behaviour into silicone rubber.

### 5 | CONCLUSION

In this article, the molecular diffusion behaviour of waterproof sealant for submarine cable repair joints and the effect of molecular species or temperature on it are investigated, based on MD simulation. The simulation parameters are calculated and analysed in order to evaluate the permeation processes of small molecules from asphalt-based and PU-based waterproof sealant into silicone rubber insulation. The main conclusions of this article are as follows,

1. Compared with the PU-based material, the asphalt-based waterproof sealant, containing polycyclic aromatic hydrocarbons, significantly permeates into the silicone rubber insulation, thus introducing impurities with polar groups into silicone rubber.
2. The free volume in silicone rubber provides voids for the small molecules of the waterproof sealant to diffuse. The relative molecular mass of small molecules and the binding energy between the waterproof sealant and silicone are the pivotal factors affecting the diffusion behaviour. There is a linear relationship between the logarithm of the diffusion coefficient ($\ln D$) and the ratio of binding energy to relative molecular mass ($E_{\text{total}}/M_r$).
3. The rising ambient temperature increases the fraction of free volume of silicone rubber and decreases the binding energy, leading to a higher diffusion coefficient and a stronger permeation behaviour into the joint insulation. Furthermore, the change of temperature does not affect the order of diffusion coefficients, which is always as follows, Phe > Flu > Pyr > BkF > IcdP > PU. According to the Arrhenius equation, the order of the diffusion activation energy is as follows, PU > IcdP > BkF > Pyr > Phe > Flu.

To sum-up, the PU-based waterproof sealant, with lower permeability, has a great potential for the waterproof layer of high-voltage submarine cable repair joints.

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