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The Effect of Nano-Silica Modified With Silane Coupling Agents on the Diffusion Behavior of Water Molecules in Palm Oil Based on Molecular Simulation

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ABSTRACT To enhance the compatibility of nano-SiO_2 and palm oil, four different types of silane coupling agent (KH550, KH560, KH570 and KH792) were adopted to modify the surface of silica. It turns out that when using the KH792 to modify the surface of nano-SiO_2, the interfacial bonding between silica and palm oil was maximum and their overlapping area was the largest, up to 12 Å. Thus, it can be concluded that the relatively optimal silane coupling agent is KH792. And when nano-SiO_2, whose surface was modified with KH792, was added to the palm oil, the interaction energy between palm oil and water molecules was enhanced and the diffusibility of water molecules in oil was decreased. In addition, the addition of nano-SiO_2 to the oil reduced the diffusible space of water molecules provided by oil and further restricted the diffusion of water molecules in oil. Therefore, it can be seen that the addition of nano-SiO_2 to palm oil may cause the restriction of the diffusion of water molecules in it.

INDEX TERMS Palm oil, molecular simulation, surface modification, silane coupling agent, interaction energy.

I. INTRODUCTION

Recently, in the dielectric materials field, the usage of nanomaterial modification technology has been the research hotspot [1], [2], and it can be used to improve the electricity [3], [4], heat conduction [5], [6] and aging [7], [8] of insulation oil. Yuxiang Zhong and his team found that TiO_2 nanoparticles not only enhance the AC breakdown voltage of ester-based nanofluid, but also improve the partial discharge performance of ester-based nanofluids compared to natural esters [9]. The addition of Al_2O_3+ZnO nanocomposites to the vegetable oil enhanced the thermal and electronic properties of it, which was better than to the mineral oil, founded by Zhong *et al.* [9]. However, direct doping of inorganic nanoparticles into insulating oils is not conducive to the performance of nanocomposites due to the incompatibility of inorganic additive particles with organic matrix materials. Surface modification of nanoparticles can enhance the interfacial binding of nanoparticles to the substrate. Many researchers have used surface organic modification of nanoparticles by using surface modifiers [11], [12] to reduce the agglomeration of nanoparticles and enhance the performance of nanocomposites [13], [14]. For the improvement of compatibility between nanoparticles and matrix materials, silane coupling agent was utilized to modify the surface of nano SiO_2. Jieyuan Zheng *et al.* adopted eugenol group epoxy silane coupling agent to modify the surface of SiO_2, showed that after surface modification, the compatibility between SiO_2 and epoxy resin was increased, thus the perfermance of cured epoxy products was enhanced [15]. ShujieTian *et al.* used silane coupling agent (KH570) for surface modification of silica to improve the dispersion stability of nanoparticles in the matrix material [16]. Wei Zheng *et al.* also made a research on the microscopic effect of nano-silica

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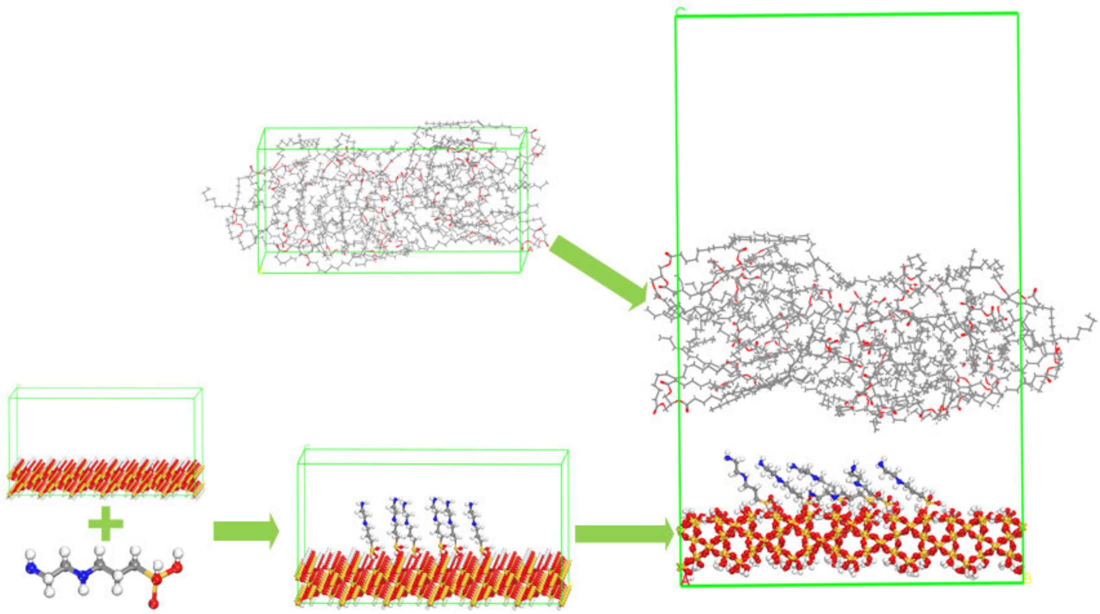


FIGURE 1. The layer model of surface-modified SiO_2 and palm oil.

modification by using different silane coupling agents on cellulose/nano-silica interface. The results showed that nano-SiO_2 modified with silane coupling agent (KH792) could obtain the highest interfacial binding energy and binding energy density [17]. However, the effect of silane coupling agent modified silica on the interfacial interaction between silica and palm oil has rarely been reported from a microscopic perspective.

Water molecule is considered as the ''Enemy Number One'' of transformers in addition to temperature. Because the presence of moisture can greatly reduce the breakdown voltage and partial initial discharge voltage of the insulating oil, and can form a synergistic effect with temperature during the operation of the transformer to accelerate the aging of the oil-paper insulation system [18]–[21]. With the increase of water molecules, the AC breakdown voltage of palm oil went down almost exponentially [22]. Most studies on nano-modified vegetable oils have paid less attention to the diffusion and influence mechanisms of water in nano-modified insulating oils. But the diffusion mechanism of the nanocomposites is more complicated due to the addition of nanoparticles. And in previous works, we have confirmed that adding nano-SiO_2 particles can inhibit water diffusion in mineral insulation oil [23]. In order to further study the influence of nanoparticles on the diffusion of water molecules in palm oil. In this paper, the effect of SiO_2 nanoparticles on the diffusion of water molecules in palm oil was investigated.

Therefore, in this paper, the effect of different silane coupling agents modified silica on the interface between silica and palm oil was investigated by molecular simulation method. Through analysis of interfacial interaction and concentration distribution, determined the relatively optimal silane coupling agent; besides, surface-modified silica was added to palm oil, and the effect of nanoparticles on the diffusion of water molecules in palm oil was investigated by analyzing the mean square displacement of water molecules in palm oil containing nanoparticles, the free volume, and the interaction energy between them.

II. MODELING AND SIMULATION

A. INTERFACE MODELING

The surface model of lower SiO_2 was established, and the upper surface was hydroxylated. The hydroxyl location on the same position on the SiO_2 surface was selected to graft the silane coupling agent molecule, thus forming modified nano-SiO_2 surface. The upper palm oil model was built according to the proportion of each triglyceride molecule in palm oil and lastly the layer model containing SiO_2 and palm oil was constructed, as shown in Fig.1.

B. THE BLENDING MODEL OF PALM OIL, NANO-SiO_2 AND WATER MOLECULES

Palm oil is composed of various triglyceride molecules, each of whose proportion is shown in Tab. 1 [24]. The content of surface-modified SiO_2 nanoparticles added in the oil is 1%, and the water molecules is 5%. The mixing model of palm oil, nano-SiO_2 and water molecules is demonstrated in Fig.2.

III. RESULTS AND DISCUSSION

A. SELECTION OF RELATIVELY OPTIMAL SILANE COUPLING AGENT

1) INTERFACIAL INTERACTION ENERGY

The interface binding energy can be used to measure the interface binding strength. The greater the interface energy, the more work needs to be done to damage the

TABLE 1. The components of palm oil.

Ingredient	Palmitic acid ($C_{51}H_{98}O_6$)	Stearic acid ($C_{57}H_{110}O_6$)	Oleic acid ($C_{57}H_{104}O_6$)	Linoleic acid ($C_{57}H_{98}O_6$)	Total
ω_B(%)	44.4	4.3	39.9	9.4	98

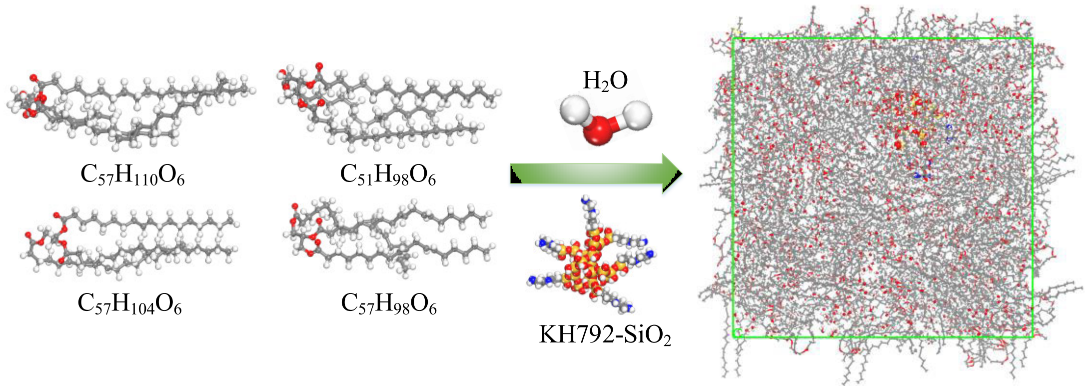


FIGURE 2. The mixing model of palm oil, nano-SiO2 and water molecules.

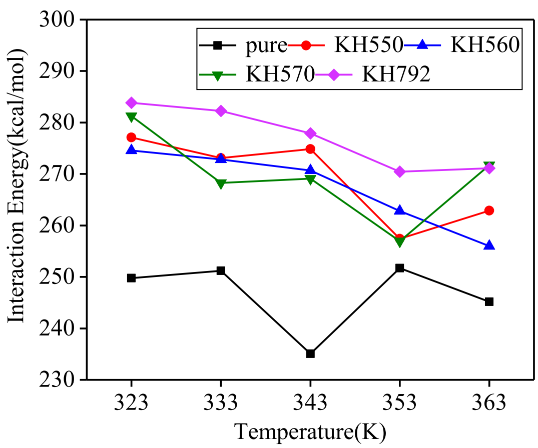


FIGURE 3. Interfacial binding energy between surface-modified nano-SiO_2 and palm oil.

interface. As shown in Fig.3, when nano-SiO_2 is modified with KH792, the interface energy between nano-SiO_2 and palm oil is the largest. The reason for the different interaction between nano-SiO_2 modified by different silane coupling agents and palm oil is that different silane coupling agents contain different groups. The corresponding groups of KH550, KH560, KH570 and KH792 selected in this paper are amino group (-NH_2), epoxy group ($-O-CH_2-CH(O)CH_2$), methylacrylloxyl group ($-O-CO-C(CH_3)=CH_2$) and N-(β-aminoethyl)-γ-aminopropyl group ($-NH-CH_2-CH_2-NH_2$). Amino ($-NH_2$) and imino ($-NH-$) groups can form hydrogen bonds (O-H—N and N-H—O) with the ester groups on the triglyceride molecule. KH550 only contains amino group(-NH_2), while KH792 has amino group(-NH_2) and imino group (-NH-), so the amounts of nano-SiO_2 modified with KH792 and hydrogen bonds in palm oil are larger than that of nano-SiO_2 modified with KH550 and hydrogen bonds in palm oil. Owing that KH92 contains two groups, amino ($-NH_2$) and imino ($-NH-$), the formation of hydrogen bond is beneficial to enhance the electrostatic force between the two, thus promoting the interfacial interaction energy between nano-SiO_2 modified with KH792 and palm oil. As a result, it can be seen that the interfacial interaction force is the largest between the nano-SiO_2 modified with KH792 and palm oil. Thus, from the microscopic perspective, this paper has verified the usage of silane coupling agents to modify nano-SiO_2 can improve the interfacial interaction between palm oil and nano-SiO_2, and it thus effectively increases the compatibility between them.

2) INTERFACIAL RELATIVE CONCENTRATION

Relative concentration distribution at the interface can determine the spatial position and directional arrangement of molecules or atoms in the system [25]. As can be seen in Fig.4, there is an overlapping area of the concentration distribution of nano-SiO_2 and palm oil along the Z direction, which demonstrates that adsorption exists between nano-SiO_2 and palm oil. In the layer model of unmodified nano-SiO_2 and palm oil, the overlapping distance is 3.1Å, while in the layer model of nano-SiO_2 modified with KH550, KH560, KH570 and KH792 and palm oil, the overlapping distance are respectively 8.5 Å, 9.7 Å, 10.4 Å and 12 Å. In conclusion, nano-SiO_2 modified with silane coupling agents can increase the overlapping distance between the interface of nano-SiO_2 and palm oil, and improve the compatibility between nano-SiO_2 and palm oil. The order of overlap distance between the interface of nano-SiO_2 modified with silane coupling agents and palm oil is as follows: KH792>KH570>K560>K550>Pure.

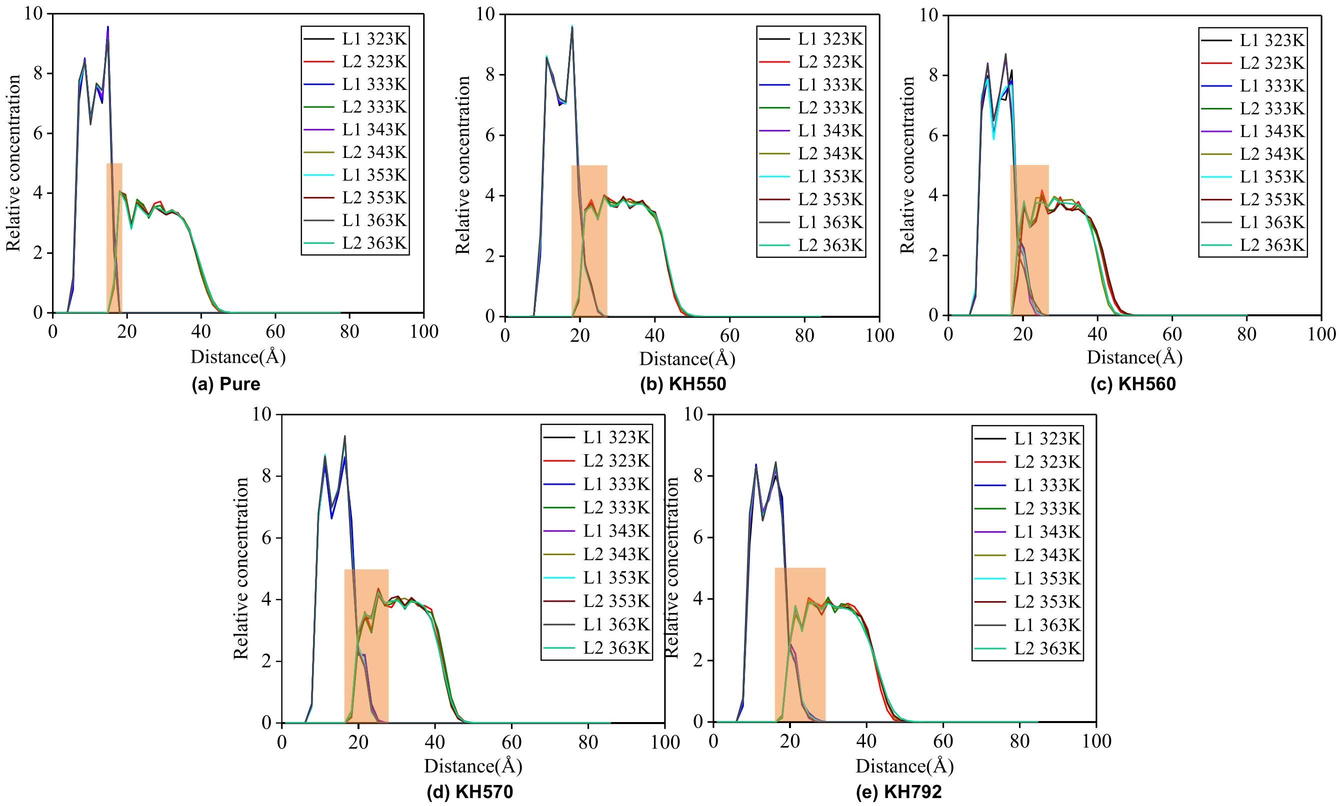


FIGURE 4. The Interfacial concentration distribution of nano-SiO_2 modified with different silane coupling agents and palm oil.

In summary, the relatively optimal silane coupling agent was selected as KH792, which was used to modify the surface of nano-SiO_2, and then added into palm oil to build a nano-modified palm oil model.

B. DIFFUSION BEHAVIOR OF WATER MOLECULES IN NANO-MODIFIED PALM OIL

1) FREE VOLUME

The free volume of water molecules in oil is an important factor affecting the diffusion of it. According to the Free Volume Theory of Fox and Flory [26], the Total Volume(V_t) inside the material can be divided into Occupied Volume(V_o) and Free Volume(V_f). The formula of Fraction of Free Volume (FFV) is as follows:

$$FFV = \frac{V_f}{V_f + V_o} \times 100\% \tag{1}$$

In Fig.5, the Occupied Volume is represented in gray and the Free Volume is in blue. We can see that the blue area grows gradually with the increase of the temperature, which means that the Free Volume in the model grows gradually as the temperature increases. In addition, the Free Volume in the model without nanoparticles is larger than that in the model with nanoparticles at the same temperature. While the growth of the Free Volume in the model helps raise the free space for the diffusion of water molecules, which intensify the diffusion of water molecules in the oil medium. Therefore, the addition of nanoparticles can help inhibit the diffusion of water molecules in palm oil.

As can been seen from Tab. 2 and Tab. 3, at the same temperature, The FFV of water molecules in nano-modified palm oil is smaller than that of water molecules in palm oil. This is because when nanoparticles are added to the oil, the nanoparticles occupy part of the free volume in the model, reducing the free volume available for the diffusion of water molecules. Thus, the FFV of water molecules in nano-modified palm oil is less than that in palm oil.

2) INTERACTION ENERGY

The energy of interaction between water molecules and palm oil plays an important role in influencing the diffusion of water molecules. The energy of interaction can be figured out by the following formula:

$$E_{int} = E_{total} - (E_{oil} + E_{water}) \tag{2}$$

E_{int} is the interaction energy of oil and water, and E_{total} is the total potential energy of the whole model. E_{oil} is the potential energy of oil and E_{water} is the potential energy of water. If the interaction energy is positive, it indicates that substances are mutually exclusive; if the interaction energy is negative, it indicates there is mutual attraction between substances, and the larger the negative value is, the stronger the binding effect between substances will be.

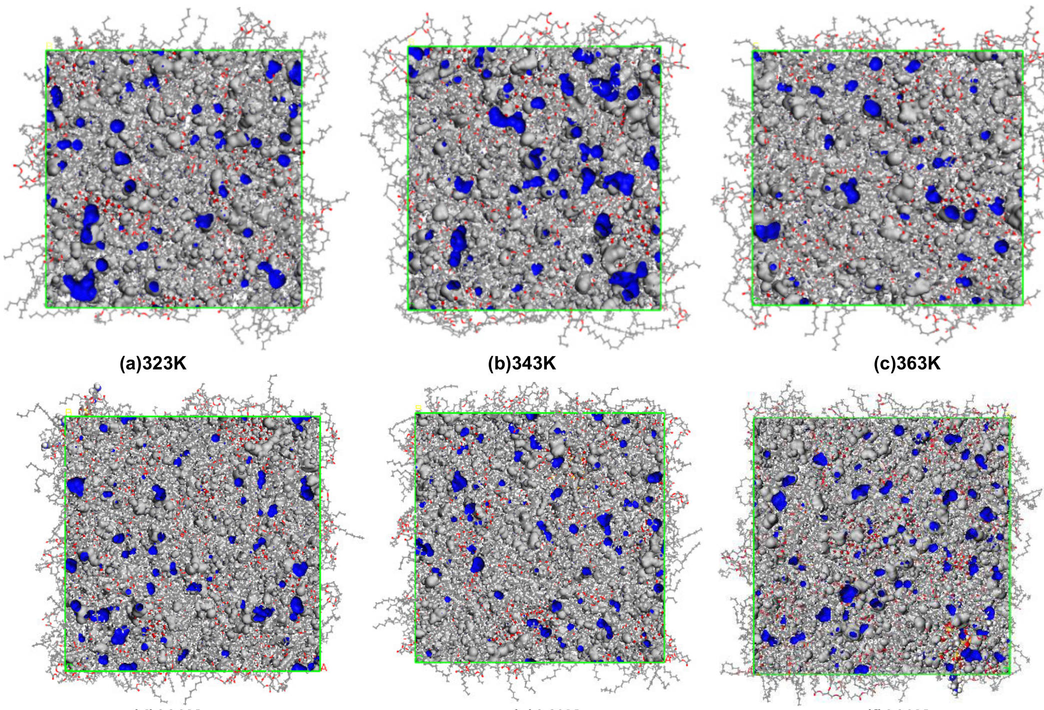


FIGURE 5. Free volume of water molecules in oil at different temperatures: (a)~(c) Free volume of water molecules in palm Oil; (e)~(f) Free volume of water molecules in nano-modified palm Oil.

TABLE 2. Fraction of free volume of water molecules in palm oil.

	323K	333K	343K	353K	363K
V_O(Å^3)	180559.36	180596.98	180146.52	180107.22	179949.91
V_f(Å^3)	14874.29	15152.84	15419.64	15577.37	15650.9
FFV(%)	0.0761	0.0774	0.0788	0.0796	0.08

TABLE 3. Fraction of free volume of water molecules in nano-modified palm oil.

	323K	333K	343K	353K	363K
V_O(Å^3)	382146.45	382608.69	381768.95	381548.89	381815.83
V_f(Å^3)	30577.62	30941.16	30983.29	32000.96	32334.02
FFV(%)	0.0741	0.0748	0.0751	0.0774	0.0782

TABLE 4. Interaction energy between water molecules and palm oil.

	323K	333K	343K	353K	363K
Evdw(kcal/mol)	-316.1518	-304.8219	-297.027	-277.6865	-290.4105
Eelec(kcal/mol)	-458.3072	-386.0354	-359.7754	-348.6581	-316.3299
Eint(kcal/mol)	-774.459	-690.8573	-656.8024	-626.3446	-606.7404

According to Tab. 4 and Tab. 5, the energy of interaction between water molecules and the two oil media gradually reduces as the temperature raises, which is because the increase of temperature accelerates the thermal motion of water molecules, and also weakens the binding capacity of oil medium to water molecules, thus the energy of interaction between water molecules and oil gradually decreases. The decline of interaction energy helps increase the diffusion of water molecules in the oil.

TABLE 5. Interaction energy between water molecules and nano-modified palm oil.

	323K	333K	343K	353K	363K
Evdw(kcal/mol)	-649.665	-632.211	-637.149	-628.832	-619.914
Eelec(kcal/mol)	-895.297	-886.461	-820.67	-788.087	-769.633
Eint(kcal/mol)	-1544.96	-1518.67	-1457.819	-1416.92	-1389.55

By making the contrast of the two groups data, we can see that the energy of interaction between nano-modified palm oil and water molecules is greater than that between palm oil and water molecules. The reason is that the existence of nanoparticles promotes the adsorption of palm oil on water molecules, thus helping increase the energy of interaction between palm oil containing nanoparticles and water molecules. The surfaces of the SiO_2 nanoparticles, used in this paper, are modified with KH792. Because KH792 can form hydrogen bonds together with palm oil, the surface-modified SiO_2 nanoparticles can be stably dispersed in palm oil, conducive to improving the insulation performance of nanofluids. In addition, KH792 can also form hydrogen bonds together with water molecules, which is beneficial to grow the electrostatic force between the nanofluid and water molecules, thus improve the energy of interaction between the nano-modified palm oil and water molecules.

3) MEAN SQUARE DISPLACEMENT

The motion state of water molecules can be expressed by mean square displacement (MSD), which describes the average distance of all particles from their initial point at time t. Its formula is as follows:

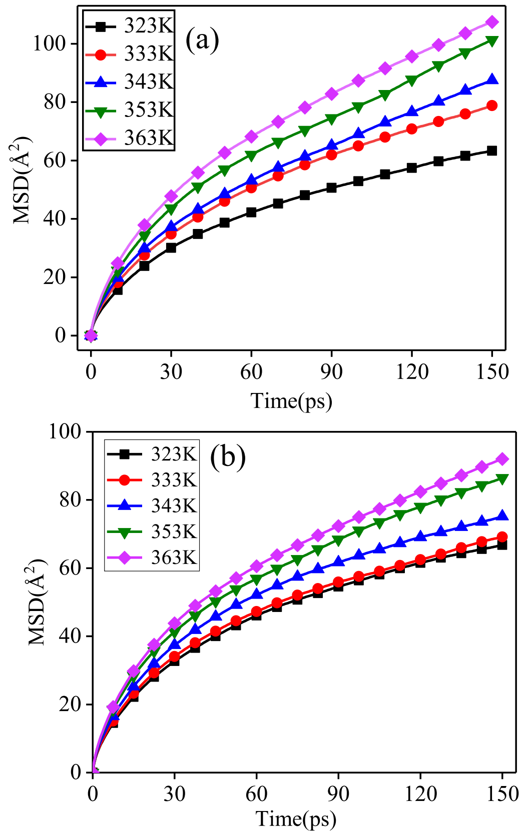
$$\mathrm{MSD} = \langle |\vec{r}_i(t) - \vec{r}_i(0)|^2 \rangle \tag{3}$$

$\vec{r}_i$ (t) and $\vec{r}_i(0)$ respectively represents the position vectors of particles at time t and time 0. <> indicates the average value of the calculated results. Diffusion coefficient is an important parameter to characterize the diffusion ability of a substance. The larger the diffusion coefficient is, the smaller the binding effect of the medium on the particle will be. The diffusion coefficient of water molecules in oil medium is calculated by the following equation [18]:

$$D = \frac{|\vec{r}_i(t) - \vec{r}_i(0)|^2}{6t} = \frac{\alpha}{6} \tag{4}$$

In the above equation, α is the slope of the MSD fitting curve.

According to Fig.6, as the temperature raises, MSD of water molecules in the two oil media gradually increase. The reason is the raise of temperature accelerates the thermal motion of water molecules, which results in the acceleration of the motion of water molecules. In palm oil, when the temperature is from 323K to 363K, the MSD of water molecules is between 0Å^2 and 108Å^2. In the nano-modified palm oil, the MSD of water molecules is between 0Å^2 and 91Å^2 when the temperature is from 323K to 363K. The



MSD curves of water molecules in palm oil; (b) MSD curves of water molecules in nano modified palm oil

FIGURE 6. MSD curves of water molecules at different temperatures: (a) MSD curves of water molecules in palm oil; (b) MSD curves of water molecules in nano modified palm oil.

MSD of water molecules in the nano-modified palm oil is significantly smaller than that in the unmodified palm oil. As a result, the addition of nano-particles can help inhibit the diffusion of water molecules in the palm oil, conducive to improving the insulation performance of the palm oil.

In order to make a further analysis of the diffusion capacity of water molecules in the two oil media, the diffusion coefficient of water molecules in two oil media was researched. According to Tab.6 and Tab.7, as the temperature grows, the diffusion coefficient of water molecules in the two oil raises gradually. Meanwhile, the diffusion coefficient of water molecules in the unmodified palm oil is greater than that in the nano-modified palm oil. The reason is that the surface-modified SiO_2 nanoparticles can form hydrogen bonds together with water molecules, which

TABLE 6. Diffusion coefficient of water molecules in palm oil.

	323K	333K	343K	353K	363K
α	0.339	0.4334	0.4777	0.547	0.585
r	0.933	0.9452	0.9623	0.9569	0.9476
D(Å²/s)	0.0565	0.0722	0.0796	0.0912	0.0975

TABLE 7. Diffusion coefficient of water molecules in nano-modified palm oil.

	323K	333K	343K	353K	363K
α	0.3542	0.3587	0.3936	0.4557	0.4835
r	0.9206	0.9186	0.913	0.9351	0.9337
D(Å²/s)	0.059	0.0598	0.0656	0.076	0.0806

makes the amount of hydrogen bonds that is between water molecules and nano-modified palm oil greater than that between water molecules and unmodified palm oil, and the nanofluid can bound more water molecules. At the same temperature, the energy of interaction between water molecules and nano-modified palm oil is greater than that between nano-modified palm oil and unmodified palm oil, which shows that the adsorption capability of nano-modified palm oil for water molecules is stronger. Thus, the diffusion coefficient of water molecules in nano-modified palm oil is smaller.

IV. CONCLUSION

In this paper, molecular simulation method was utilized to make a research on the effects of nano-SiO_2 modified with different silane coupling agents on the interface between palm oil and nano-SiO_2, and the relatively optimal silane coupling agent was determined to modify nano-SiO_2 particles; secondly, the diffusion of water molecules in the palm oil modified with nano-SiO_2, whose surface is modified, was researched. The following conclusions have been drawn:

(1) In this paper, the silane coupling agents selected are KH550, KH560, KH570 and KH792, whose corresponding groups are: amino group ($-NH_2$), epoxy group ($-O-CH_2-CH(O)CH_2$), methylacrylloxyl group ($-O-CO-C(CH_3)=CH_2$) and N$-$(β-aminoethyl)-γ-aminopropyl group ($-NH-CH_2-CH_2-NH_2$). KH792 contains two groups, amino ($-NH_2$) and imino ($-NH-$), and the two groups can help form hydrogen bonds with the ester group on a triglyceride molecule. The formation of hydrogen bond is conducive to promoting the electrostatic force between the two objects, thus making the interfacial binding energy between KH792-modified nano-SiO_2 and palm oil maximum. Meanwhile, by comparing the concentration distribution of the interface between nano-SiO_2 modified with four types of silane coupling agents and palm oil, it was found that the interfacial overlapping distance between nano-SiO_2 modified with KH792 and palm oil was the largest. To sum up, when nano-SiO_2 is modified with KH792, the interfacial binding between nano-SiO_2 and palm oil may be promoted.

(2) The diffusion ability of water molecules in palm oil was inhibited by adding nano-SiO_2 modified with KH792. The reason is that after the SiO_2 nanoparticles were added, it helps raise the energy of interaction between oil medium and water molecules, thus enhancing the binding capacity of oil for water molecules and weakening the diffusion of water molecules in oil media. In addition, the FFV of water molecules is smaller in the model containing SiO_2 nanoparticles, which shows that the diffusing space for water molecules is smaller after the SiO_2 nanoparticles are added, and further restricting the diffusion of water molecules in the oil.

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