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ABSTRACT
Thermodynamic properties of cellulose insulation paper are vital factors affecting the life of a transformer; in order to obtain cellulose insulation paper with better thermodynamic properties, three types of silane coupling agents—3-aminopropyltriethoxy silane (KH550), 3-glycidoxypropyltrimethoxy silane (KH560), and 3-methacryloyloxypropyltrimethoxy silane (KH570)—were grafted on the surface of nano-SiO$_2$, and thermodynamic properties of cellulose modified with nano-SiO$_2$ were explored. The molecular dynamics method was used to establish a composite model of nano-SiO$_2$/cellulose. Also, different silane coupling agent grafted nano-SiO$_2$/cellulose models were established to explore the effect of mechanical properties, interaction energy, free volume, and hydrogen bonds on thermodynamic properties. The results showed that KH550 was the best modification of the nano-SiO$_2$/cellulose system among the three grafted silane coupling agents because KH550 grafted on the surface of nano-SiO$_2$ formed more hydrogen bonds in the cellulose system. The interfacial bonding strength between the nano-SiO$_2$ and the cellulose chains can effectively improve the thermal stability of the cellulose insulating paper.

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I. INTRODUCTION

Ensuring the safety of power supply in a transmission grid is an important prerequisite for national security and stable economic growth. However, the electrical strength of cellulose insulating paper in a transformer oil-paper insulation system will decrease due to hydrolysis of the cellulose chain. Therefore, it is of great significance to improve the thermal stability of insulating paper.

Physical and chemical modifications are usually used to improve the properties of insulating paper. Liao modified cellulose insulating paper with nano-$\text{Al}_2\text{O}_3$ and proposed that nano-$\text{Al}_2\text{O}_3$ could effectively absorb water and small molecular acids into cellulose insulating paper. Zhang’s research shows that nano-$\text{SiO}_2$ microspheres can effectively reduce the dielectric constant of insulating paper and improve the electrical performance of insulating paper. Wang grafted cellulose with polysiloxane, and the results indicated that the thermal stability of grafted cellulose was improved effectively. It can be demonstrated that by grafting cellulose, it not only has the advantages of its own properties but also has the advantages of grafting particles.

With the development of nanotechnology, nanoparticles have been widely used in the research of polymer because of their advantages of small particle size and high surface activity. Huang used elastic nanoparticles to modify epoxy resin and found that the toughening effect of modified epoxy resin is more than that of traditional rubber. Kotchetov investigated how nanoparticles can reduce the dielectric constant of insulating composites. Zhang showed that nanocomposites play an important role in power insulation. Nano-$\text{SiO}_2$ is one of the most productive nanomaterials in the world. However, the surface of nano-$\text{SiO}_2$ reacts with water...
molecules in the air with hydroxyl groups, which cannot be dispersed and combined with composite materials, leading to the degradation of insulating material properties. Thus, modification of nano-SiO$_2$ is used to improve the performance of nano-SiO$_2$. Min$^{19}$ grafted PAAM onto the surface of nano-SiO$_2$ particles and added it into the polypropylene matrix, which significantly improved the mechanical properties of the composites. Li$^{12}$ prepared an $m$-LLDPE/nano-SiO$_2$ composite by the melt blending method and found that the tensile properties and insulation impact strength increased, indicating the importance of nano-SiO$_2$ modification in improving its mechanical properties and stability. There is abundant evidence that shows that modification of nano-SiO$_2$ to improve material properties has been widely used, but still there is no systematic study on the influence of the modification of nano-SiO$_2$ on the thermodynamic properties of cellulose insulating paper.

In this paper, the molecular dynamics simulation studies on the mechanical properties and thermal stability of cellulose insulating paper of three different grafting nano-SiO$_2$ silane coupling agents, the composite model of nano-SiO$_2$/cellulose, and different silane coupling agent grafted composite models of nano-SiO$_2$/cellulose are established. Mechanical properties, free volume, interaction energy, and the effect of hydrogen bonds between the different types of silane grafting coupling agents on the nano-SiO$_2$/cellulose are studied from a microscopic view. Then, silane coupling with agents for better performance in a composite model of nano-SiO$_2$/cellulose is chosen in this paper.

II. MODEL BUILDING AND DYNAMICS SIMULATION DETAILS

Based on the amorphous cell (AC) module of the materials studio (MS), different composite models with cellulose DP = 10$^{13}$ and an initial density of 0.6g/cm$^3$ are built, which are doped with 12.5 wt. % nano-SiO$_2$.

Models with a nano-SiO$_2$ surface grafted with 3-aminopropyltriethoxy silane (APTES, KH550), 3-glycidoxypropyltrimethoxy silane (KH560), and 3-methacryloyloxypropyltrimethoxy silane (KH570) are expressed as KH550, KH560, and KH570, respectively. Also, the model without grafting treatment of the nano-SiO$_2$ surface is expressed as pure.

As shown in Fig. 2, hydroxylation of nano-SiO$_2$ was performed before building the model$^{14}$ because the silane coupling agent will be hydrolyzed during the actual grafting process. The silane coupling agent first reacts with water and then reacts with nano-SiO$_2$. Three types of silane coupling agents were grafted onto nano-SiO$_2$ with a radius of 5 Å.$^{16}$

First, geometric optimization was performed on the model through 5000 steps in a Forcite module. Second, annealing was performed to optimize the mechanical properties.
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illustrate the influence of different silane coupling agents on the thermodynamic parameters of a nano-SiO₂/cellulose model.

A. Mechanical properties

The strain-stress relationship of solid materials can generally be expressed by Hooke’s law by the following equation:\[\sigma_i = C_{ij} \epsilon_j.\] (1)

In Eq. (1), \(C_{ij}\) represents the elastic matrix coefficient, \(\sigma_i\) represents the stress vector, and \(\epsilon_j\) represents the strain vector. The stress component is calculated by the following equation:

\[
\sigma_{ij} = -\frac{1}{\nu} \sum_k \left[ m^k (u^k_i u^k_j) + \frac{1}{2} \sum_{j=k}^{n} (\tau^{ik}_{jk}) f^{jk}_{ik} \right].
\] (2)

In Eq. (2), \(\nu\) represents the volume, \(m^k\) and \(u^k\) represent the mass and velocity, respectively, of particle \(k\), \(\tau^{ik}_{jk}\) represents the distance between the first particle and particle \(k\), and \(f^{jk}_{ik}\) represents the force applied between the first particle and particle \(k\); the elastic constant coefficients \(\lambda\) and \(\mu\) are calculated by the following equations:

\[
\lambda = \frac{1}{6} (C_{12} + C_{13} + C_{21} + C_{23} + C_{31} + C_{32}) = (C_{12} + C_{13} + C_{23}),
\] (3)

\[
\mu = \frac{1}{3} (C_{44} + C_{55} + C_{66}),
\] (4)

\[
\lambda + 2\mu = \frac{1}{3} (C_{11} + C_{22} + C_{33}).
\] (5)

Supposing the column vectors formed by the side lengths of hexahedral deformation at the reference time are \(a\), \(b\), and \(c\), the inverse matrix of \(C_{ij}\) is expressed as \(S_{ij}\), Poisson’s ratio as \(\nu\), elasticity modulus as \(E\), bulk modulus as \(K\), and shear elasticity as \(G\). The calculations are shown in the following equations:

\[
\nu = \frac{\lambda}{2(\lambda + \mu)},
\] (6)

\[
K = \left[ 3(a + 2b) \right]^{-1},
\] (7)

\[
G = \frac{5}{4a - 4b + 3c},
\] (8)

\[
E = 2G(1 + \nu) = 3K(1 - 2\nu).
\] (9)

Mechanical properties are an important parameter to evaluate the mechanical strength of polymer materials.\[23\] Elasticity modulus \((E)\) is the parameter that measures the rigidity of a material, which is positively correlated with the ability of a material to resist deformation. Shear modulus \((G)\) is the ratio of shear stress to strain. Bulk modulus \((K)\) is a kind of elastic modulus reflecting the macroscopic properties of materials and that characterizes the incompressibility of materials. Poisson’s ratio \((\nu)\), also known as the transverse deformation coefficient, reflects the degree to which a material resists transverse deformation. \(K/G\) is the ratio of volume modulus to shear modulus, which is used to evaluate the toughness of materials. Cauchy pressure is used to measure the ductility of a material, and a negative value indicates that the material is brittle. In this paper, the different parameters mentioned above of different models under 343 K were calculated. Table 1 shows the mechanical parameters.

III. RESULT ANALYSIS

This section explores the influence of mechanical properties, interaction energy, free volume, and hydrogen bonds on the thermodynamic properties of the model. The mechanical properties of polymer materials characterize the macroscopic mechanical properties of the materials. At the same time, the excellent degree of mechanical properties is also related to the thermal motion of molecules; the decrease in the space of molecular motion and free volume fraction and the intermolecular interaction indicate the bonding state of molecules. The closer the bonding between molecules is, the greater is the interaction energy between molecules, which shortens the bonding distance between molecules and facilitates the formation of hydrogen bonds. These thermal parameters can be further supplemented with macroscopic mechanical parameters to jointly
TABLE I. Mechanical properties of different models.

| Model       | Bulk modulus (Gpa) | Shear modulus (Gpa) | Poisson’s ratio | Elasticity modulus (Gpa) | K/G | Cauchy pressure (Gpa) |
|-------------|--------------------|---------------------|-----------------|--------------------------|-----|-----------------------|
| Pure        | 6.8628             | 2.6021              | 0.2412          | 9.9304                   | 2.6374 | 0.7822            |
| KH550       | 7.9698             | 4.986               | 0.3317          | 12.3769                  | 1.5984 | 0.7618            |
| KH560       | 7.0627             | 4.0172              | 0.2609          | 10.1308                  | 1.7581 | −0.3969          |
| KH570       | 7.6475             | 4.7314              | 0.2435          | 11.7674                  | 1.6163 | −0.3665          |

of these models. The volume modulus, shear modulus, Poisson’s ratio, elastic modulus and K/G values of KH550 at 343 K were all higher than those of the nano-SiO$_2$ cellulose composite model without grafting or when grafted with KH560 and KH570. It indicates that the deformation resistance and ductility of the model grafted with KH550 are the best. This is due to the introduction of an Si–O bond in the cellulose chain, which improved the performance of the cellulose model with nano-SiO$_2$ particles. Comparing the different SiO$_2$/cellulose models grafted with silane coupling agents, KH550, KH560, and KH570 increased the bulk modulus of pure nano-SiO$_2$/cellulose models by 16.1%, 2.9%, and 11.4%, shear modulus by 91.6%, 54.3%, and 81.8%, Poisson’s ratio by 37.5%, 8.2%, and 1%, and elastic modulus by 24.6%, 20.1%, and 18.5%, respectively. According to the proportion of improvement in mechanical properties, it can be concluded that the mechanical properties of KH550 grafted nano-SiO$_2$ modified cellulose can be improved the most.

B. Free volume

The volume of polymer materials is divided into occupied volume and free volume. The volume of molecules or atoms in the material is occupied volume, and the system of interstitial space in the material is free volume. Free volume refers to the proportion of unfilled space in the polymer in the total volume, which is often used to characterize the material’s plasticity and sensitivity to deformation. In this paper, we define the plane ABC as a plane parallel to plane oxy and perpendicular to the z axis and select it as a reference plane. Figure 4 shows the free volume distribution of plane ABC of the four models. The blue part is the free volume, and the gray part is the occupied volume. Table II shows free volume fractions of different models at 343 K. The relation between free volume and occupied volume can be expressed by the following equation:

$$\zeta = \frac{V_{\text{free}}}{V_{\text{free}} + V_{\text{occupy}}} \times \%. \quad (10)$$

$V_{\text{free}}$ represents the free volume, and $V_{\text{occupy}}$ represents the occupied volume.

Figure 4 shows the schematic diagram of the pure model [Fig. 4(a)], KH550 [Fig. 4(b)], KH560 [Fig. 4(c)], and KH570 [Fig. 4(d)]. The fraction of the unmodified model is larger than that of the three modified models, which shows that the unmodified model has more space for free movement of cellulose chains than the modified models. At high temperature, the structure of cellulose is more easily destroyed, and the thermal stability of the polymer material is reduced. Due to the formation of more hydrogen bonds, the silane grafting coupling agent model enhanced the adsorption capacity and intermolecular interaction between nano-SiO$_2$ and cellulose. Among the three different silane coupling agents, the model grafted with KH550 has the smallest free volume fraction because

![Free volume distribution](image)

TABLE II. Free volume fractions of different models at 343 K.

| Model       | $V_{\text{free}}$ | $V_{\text{occupy}}$ | $V_{\text{free}} + V_{\text{occupy}}$ | $\zeta$ |
|-------------|-------------------|---------------------|-------------------------------------|--------|
| Pure        | 8 344.07          | 5 292.65            | 13 636.72                           | 0.248  |
| KH550       | 25 272.14         | 26 569.45           | 51 841.59                           | 0.166  |
| KH560       | 33 616.21         | 31 862.10           | 65 478.31                           | 0.243  |
| KH570       | 34 715.13         | 32 930.86           | 67 645.99                           | 0.218  |
KH550 also formed the most hydrogen bonds between the cellulose chain, which increased the intermolecular force and reduced the space of cellulose chain movement.

C. Interaction energy

The total energy of the nano-SiO$_2$/cellulose composite model is composed of nonbond energy, valence energy, and cross interaction energy, van der Waals force and electrostatic interaction energy play a dominant role in nonbond energy.\textsuperscript{27,28} Interaction energy can characterize the strength of intermolecular interactions.\textsuperscript{9–31} In order to study the scale and interface stability of the interaction between cellulose and nano-SiO$_2$, the energies of the nano-SiO$_2$/cellulose system in four models were calculated by means of molecular dynamics simulation, as shown in Table III. The energy can be calculated by the following equations:

$$E_{\text{inter}} = E_{\text{total}} - (E_{\text{cellulose}} + E_{\text{SiO}_2}),$$  \hspace{1cm} (11)

$$E_{\text{inter}} = E_{\text{coulomb}} + E_{\text{vdw}} + E_{\text{H-bond}},$$  \hspace{1cm} (12)

where $E_{\text{total}}$ represents the total energy of the complex system, $E_{\text{inter}}$ represents the interaction energy between the cellulose and nano-SiO$_2$, $E_{\text{vdw}}$, $E_{\text{coulomb}}$, and $E_{\text{H-bond}}$ represent the van der Waals energy, coulomb electrostatic energy, and hydrogen bond interaction energy of the hydrogen bond, respectively.

The larger the interaction energy, the more is the energy that is required to break down the bond between molecules; Table III calculates the average energy of the last 20 frames in the different models, and it can be seen that the order of interaction energy in cellulose models grafted with different silane coupling agents is KH550 > KH560 > KH570 > pure. The interaction energy of the models grafted with silane coupling agents was larger than the ungrafted model. Because of the silane coupling agent containing different groups (amino, epoxide, and methyl acryloyloxy groups), all three kinds of silane coupling agent grafted nano-SiO$_2$ in the model can improve the van der Waals energy and electrostatic energy between nano-SiO$_2$ and cellulose. Among different silane coupling agent models, the nano-SiO$_2$/cellulose model grafted with KH550 has the highest interaction energy, so the model grafted with KH550 has the best performance.

D. Hydrogen bond analysis

The hydrogen bond is a kind of intermolecular force. Hydrogen bonding can be either intermolecular or intramolecular. If the intermolecular interaction force reaches a certain level, the existence of a hydrogen bond between molecules is proved; \textsuperscript{1–10} hydrogen bond exists between hydrogen atoms that are already covalently bonded to one electronegative atom X and another electronegative atom Y (X–H⋯Y),\textsuperscript{29} where X stabilizes the negative charge and dissociates the hydrogen atom; on the other hand, Y has a higher electron density, which attracts hydrogen atoms to form a three-center, four-electron bond. Figure 5 shows two different types of hydrogen bonds in the nano-SiO$_2$/cellulose model grafted with KH550.

As shown in Fig. 6, the number of hydrogen bonds in the nano-SiO$_2$/cellulose model grafted with a silane coupling agent at 343 K was higher than that in the nano-SiO$_2$/cellulose model grafted without a silane coupling agent, while the number of hydrogen bonds in models grafted with KH550 was higher than that in models grafted with KH560 and KH570. The number of hydrogen bonds formed between cellulose chains in different models basically remains unchanged, but nano-SiO$_2$ grafted with a silane coupling agent forms more hydrogen bonds with cellulose, increasing the total number of hydrogen bonds in the model. Hydrogen bonds formed by nano-SiO$_2$ and cellulose chains strengthen the interaction force between SiO$_2$ and cellulose, constrains the movement of the cellulose chain, and stops the destruction of cellulose.

Nano-SiO$_2$ grafted with KH550 can form N–H⋯O hydrogen bonds, as shown in Fig. 7, which increases the number of hydrogen bonds.

![Figure 5](image-url)

**FIG. 5.** An example of hydrogen bonding in the models.

![Figure 6](image-url)

**FIG. 6.** Hydrogen bond number statistics of different models at 343 K.
bonds between nano-SiO$_2$ and cellulose chains, thereby improving the thermal stability of the nano-SiO$_2$/cellulose model and slowing down the chain motion of cellulose in the model. Therefore, KH550 is the relatively best choice for improving the thermal stability of the nano-SiO$_2$/cellulose model with the same graft density. It will relatively be the best choice for improving the thermal stability of the nano-SiO$_2$/cellulose model by grafting KH550 on nano-SiO$_2$.

IV. CONCLUSIONS

A molecular simulation was used to study the mechanical properties, free volume, interaction energy, and hydrogen bonds for cellulose models modified with nano-SiO$_2$ grafted 3-aminopropyltriethoxysilane (KH550), 3-glycidoxypropyltrimethoxysilane (KH560), and 3-methacryloxypropyltrimethoxysilane (KH570) under the same temperature and same grafting density. The conclusions are as follows:

1. By grafting three different silane coupling agents, cellulose doped with nano-SiO$_2$ could improve the thermodynamic properties of cellulose and enhance the toughness and deformation resistance of the material. The interaction energy in the nano-SiO$_2$/cellulose model rises, the free volume decreases, and the number of hydrogen bonds between nano-SiO$_2$ and cellulose increases. The results show that it is necessary for nano-SiO$_2$ to be grafted with a silane coupling agent in a nano-SiO$_2$/cellulose system.

2. Among three different silane coupling agents, the degree of enhancing the thermodynamic performance of nano-SiO$_2$/cellulose composites is as follows: KH550 > KH560 > KH570; this is because the amino groups in KH550 form more hydrogen bonds with cellulose chains, which can enhance the interaction energy between nano-SiO$_2$ and cellulose chains, increasing the adsorption between nano-SiO$_2$ particles and cellulose chain. The thermal motion of O⋯H–O hydrogen bonds with cellulose, but also cellulose at high temperature, is slowed down, so as to improve the thermodynamic performance of nano-SiO$_2$/cellulose model.

After grafting silane coupling agents on nano-SiO$_2$, especially KH550, its thermodynamic properties were significantly improved, which provided reference for subsequent research on material properties improved by silane coupling agent grafting nano-SiO$_2$.

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