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Molecular Dynamics Simulation of Epoxy/Al_2O_3 Nanocomposites as Insulation Material for High Temperature Superconducting Insulation

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The insulating materials used in high temperature superconducting equipment need to maintain sound mechanical and electrical properties under large temperature gradient and high field strength. The surface effect of nanoparticles makes nanocomposites have excellent properties which meet the strict requirements of high temperature superconducting equipment for insulating materials. However, the physical mechanism of the effect of nano particle size on the insulation properties of nanocomposites is not clear. In this paper, the effect of Al_2O_3 nanoparticle size on the insulating characteristics of epoxy resin/Al_2O_3 nanocomposites was investigated using molecular dynamics at the microscopic level. Four types of Al_2O_3 nanoparticles with diameters of 1 nm, 1.17 nm, 1.34 nm, and 1.5 nm were doped into the epoxy resin in the same volume fraction, where bisphenol F diglycidyl ether (DGEBF) is the reaction matrix and diethyltoluene diamine (DETDA) is the curing agent. The insulating characteristics and microstructures of pure epoxy and epoxy resin/Al_2O_3 nanocomposites were investigated. MD simulation results show that, compared with Al_2O_3 nanoparticles of other sizes, 1 nm Al_2O_3 nanoparticles are most conductive to improving the thermodynamic and electrical properties of epoxy nanocomposites. The glass transition temperature and elastic modulus of epoxy composites increase significantly when the size of Al_2O_3 nanoparticles decreases, whereas the coefficient of thermal expansion drops. Meanwhile, Al_2O_3 nanoparticles also change the microstructure parameters of the nanocomposites, which improves the cohesive energy density and reduces the mean square displacement of the nanocomposites. Although Al_2O_3 nanoparticles almost has have no effect on the radial distribution function of all atoms, the changes of cohesive energy density and mean square displacement show that doping Al_2O_3 nanoparticles can reduce the movement ability of molecular chain in the system, which provides theoretical basis and guidance for the preparation of epoxy resin nanocomposites for high temperature superconducting insulation.

Keywords: epoxy resin nanocomposites, molecular dynamics, high-temperature superconducting insulation

1. INTRODUCTION

Epoxy resin (EP) has been widely applied in the field of electrical engineering because of its excellent mechanical, thermal and insulation properties, such as basin insulator, dry-type transformer, and superconducting equipment insulation [1], [2]. Among them, bisphenol F epoxy resin is favored by high temperature superconducting (HTS) equipment insulation because of its excellent low temperature performance. Whether it's a high-temperature superconducting current limiter or a high-temperature superconducting cable, the increasingly complex insulation environment, which includes temperature gradients, a strong magnetic field, and irradiation, places greater demands on epoxy resin's thermodynamic and electrical properties. Thermodynamic and electrical characteristics of epoxy resins have been the subject of several investigations. [3-7]. Among them, the nanoparticle-modified epoxy resin has become the focus of the research in the field of insulation because of its high strength, excellent heat resistance and excellent insulation performance, which have been successfully applied in power equipment [8-10]. MD results show that the insulation properties of the materials are closely related to their thermodynamic and dielectric properties. Therefore, the study on the thermodynamic and dielectric properties of bisphenol F epoxy resin/Al_2O_3 nanocomposites is of great significance to expand its application in the field of high temperature superconducting equipment and even electrical insulation. As engineering materials, people mostly pay attention to the application of epoxy resin nanocomposites in practice but ignore the physical mechanism of nanoparticle size on insulation properties of nanocomposites. With the development of computer technology and multi body potential function theory, molecular dynamics simulation technology has become an important means of modern material structure analysis. More importantly, molecular dynamics simulation can provide the molecular scale doping mechanism of nano materials and the influence mechanism of different factors on material properties. In particular, the comparison of different sizes of nanoparticles in microstate is of great significance to study the mechanism of nano doping modified epoxy resin. Based on molecular dynamics, K. Tanaka investigated the influence of nano fillers on the thermal conductivity of epoxy resin [11]. M. Akash discovered that doping Al_2O_3 nanoparticles within certain limitations can greatly improve the breakdown voltage [12]. S. R. Yang determined the thermodynamic and mechanical parameters of the crosslinked epoxy resin using the molecular dynamics approach [13]. Using kinetic modeling, S. Y. Yu investigated the influence of Al_2O_3 nanoparticles on the mechanical characteristics of thermosetting epoxy composites [14]. In conclusion molecular simulation technologies can reveal the link between epoxy resin/Al_2O_3 nanocomposites macro characteristics and microstructure.at molecular level which cannot be satisfied by conventional experiments.

In this paper the pure epoxy resin model and bisphenol F epoxy resin/Al_2O_3 nanocomposite model were constructed by using the reaction matrix of bisphenol F diglycidyl ether (DGEBF) and diethyltoluene diamine (DETDA) as crosslinker. The glass transition temperature, coefficient of thermal expansion, and dielectric constant of the two models were examined, as well as their thermodynamic and dielectric characteristics. The surrounding structural characteristics of each model, such as radial distribution function, mean square displacement, and cohesive energy density, were estimated to better elucidate the mechanism, and the nano doped epoxy resin was described from a molecular perspective.

2. MOLECULAR DYNAMICS MODELING AND SIMULATION

2.1. Model Establishment

Bisphenol-F epoxy resin is widely used in the field of high temperature superconducting (HTS) insulation because of its low viscosity and excellent chemical properties. Therefore, the bisphenol-F diglycidyl ether (DGEBF) was used as the reaction matrix and cross-linked with diethyltoluene diamine (DETDA). Material Studio (MS) software was employed to build pure epoxy resin and epoxy resin/Al_2O_3 models. The Amorphous Cell Tool in MS software was used to build a 3D box structure that contained DGEBF, DETDA and Al_2O_3. The stoichiometric ratio for DGEBF and DETDA in the box structure was 2:1. In order to improve the crosslinking degree of epoxy resin, the DETDA in the model was composed of two isomers, the ratio of the 3,5,2,6-DETDA and 3,5,2,4-DETDA in the DETDA is 1:4. The diameters of spherical Al_2O_3 nanoparticles saturated with surface hydroxyl groups in the epoxy resin model are 1.00 nm, 1.17 nm, 1.34 nm and 1.50 nm, respectively. The volume fraction of Al_2O_3 is 20%. The molecular models of DGEBF, 3,5,2,6-DETDA, 3,5,2,4-DETDA and 1.5 nm Al_2O_3 nanoparticle were shown in Figure 1a, b, c and d.

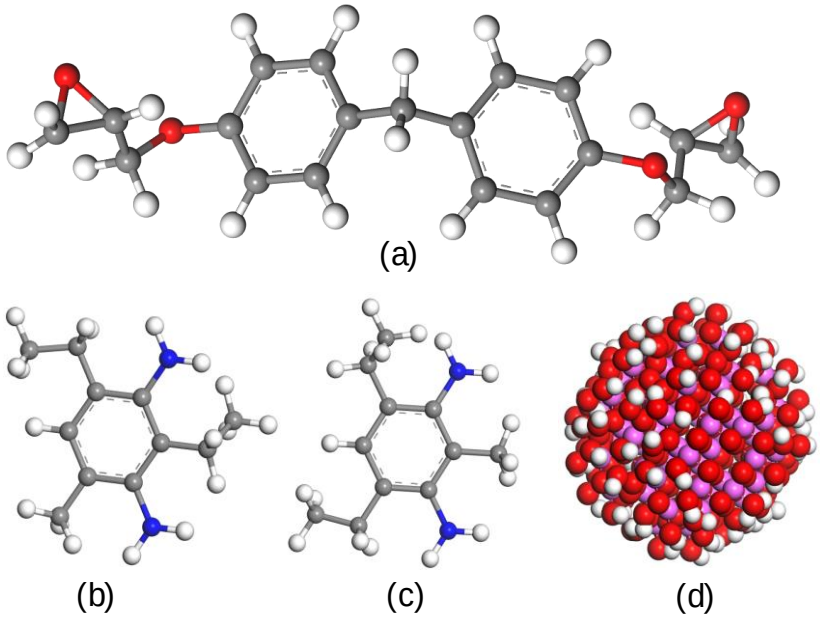
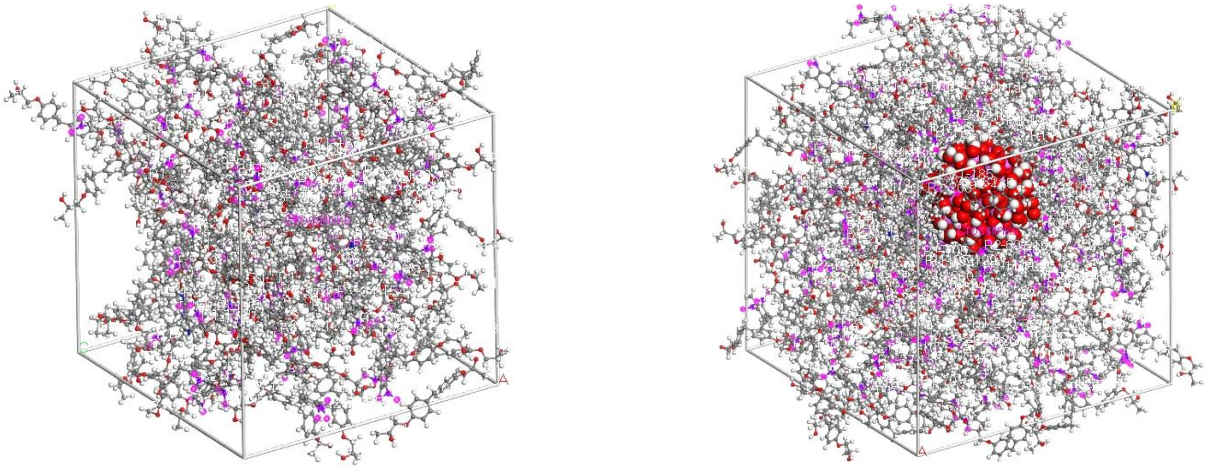


Figure 1. The monomer molecular models.

To carry out the automated crosslinking reaction of EP and EP/Al_2O_3 nanocomposites, the Perl programming language was used for MD simulation. The crosslinking reaction temperature was set at 480 K and the crosslinking distance was set in the range of 0.35 ~ 0.75 nm. The target crosslinking density for both EP and EP/Al_2O_3 nanocomposites was set at 85%. The models were equilibrated at 298 K and 1.013×10^{-4} GPa pressure at NPT ensemble for a total simulation time of 200 ps. The structure was optimized before MD simulation using Geometry Optimization. Besides that, the cyclic annealing treatment was also used to make the structure more reasonable. The COMPASSII force field was

employed throughout the MD simulation. Correspondingly, the Ewald and Atom-based summation control methods were selected for electrostatics (Coulomb) and van der Waals at a cutoff value of 12.5 Å. After crosslinking, the bisphenol F EP and the 1 nm EP/Al_2O_3 nanocomposite were obtained, as illustrated in Figure 2a and b, respectively.



(a) Molecular model of bisphenol F epoxy resin (b) Molecular model of EP/Al_2O_3 nanocomposite

Figure 2. The molecular model.

2.2 Structural Relaxation and Sampling

All the models were relaxed for 100 ps under NPT ensemble where the pressure was set at 1.013×10^{-4} GPa, and the step size was 1 fs. As shown in Figure 3a, the density of the pure epoxy resin model after relaxing was 1.21 g/cm3, which was extremely close to the experimental result. In Figure 3b, the energy of the system gradually tends to be stable with a small fluctuation after 25 ps, which indicates that the system gradually reaches the equilibrium state. To obtain the T_g of EP and EP/Al_2O_3 nanocomposite, the Forcite Anneal module was used to calculate the density of the models from 600 K to 300 K. The annealing temperature interval was 10 K which was carried out at the NPT ensemble. Finally, the T_g can be fitted by temperature-dependent density curve of each model in the cooling process.

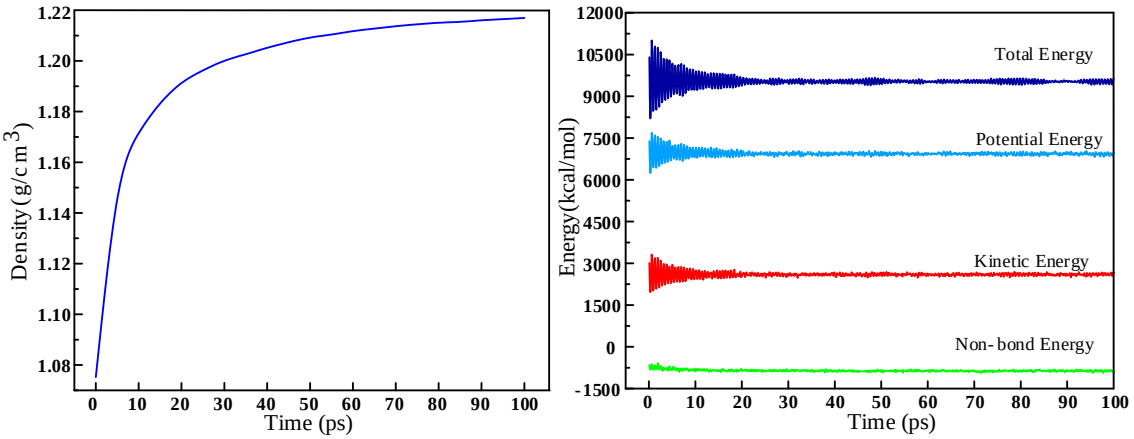


Figure 3. The epoxy resin parameters in relaxation treatment (a) Density of epoxy resin changes over time (b)Energy of epoxy resin changes over time.

3. RESULTS AND DISCUSSION

3.1 Glass Transition Temperature

The temperature at which glass transitions T_g is the lowest temperature at which a polymer macromolecular chain may freely move. Above T_g, the polymer will be elastomeric, while brittleness will be present below the glass transition temperature. In the glassy state, the density of epoxy resin reduces somewhat with increasing temperature, and it decreases rapidly above T_g. Therefore, the high T_g. epoxy resin composites have a better performance in the insulation of high temperature superconducting equipment.

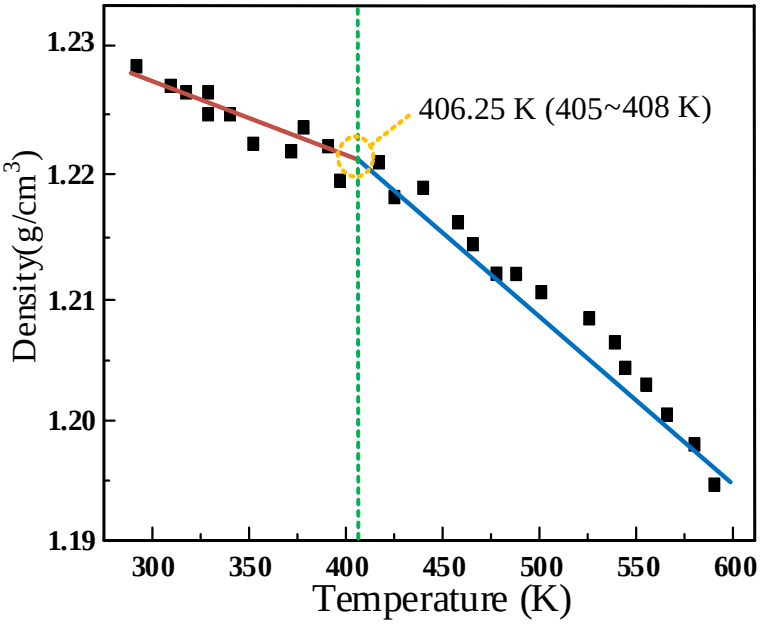


Figure 4. The schematic diagram of linear fitting density-temperature curve.

The density temperature curve of bisphenol F epoxy resin during model annealing is shown in Figure 4. The density of the model grows steadily when the annealing temperature is reduced, but the slope of the density curve changes dramatically. The values on both sides of the inflection point of the density temperature curve are fitted by the least square method, and the temperature corresponding to the intersection of the two fitting lines is the T_g of bisphenol F epoxy resin. The T_g of bisphenol F epoxy resin composite systems with different Al_2O_3 nanoparticle sizes are also obtained by fitting the temperature density curves of the other four models in the annealing process. The results are shown in Figure 5, where different composite systems were represented by the Al_2O_3 nanoparticle size.

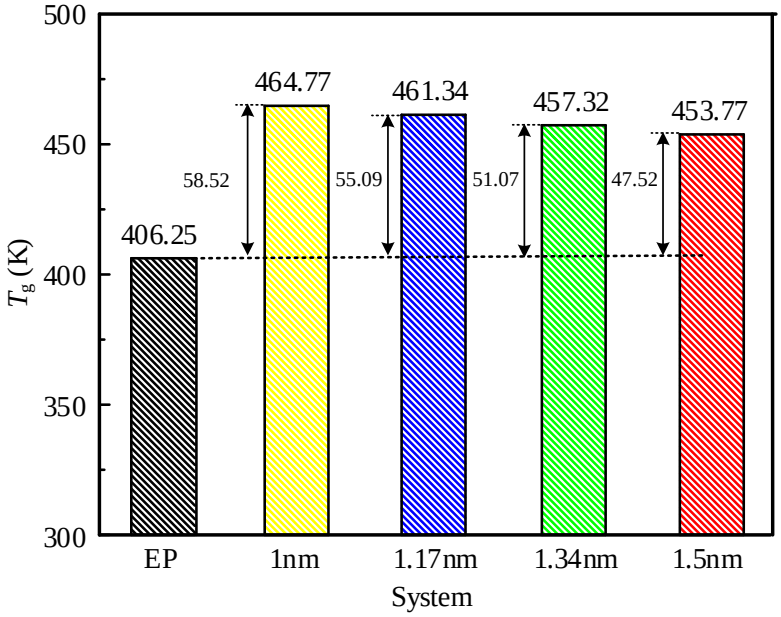


Figure 5. The T_g of different systems.

The results suggest that doping Al_2O_3 nanoparticles in bisphenol F epoxy resin improves the T_g. Among them, 1 nm Al_2O_3 nanoparticle has the largest increase in T_g of bisphenol F epoxy resin with an increase of 14%, while 1.5 nm Al_2O_3 nanoparticle has the opposite performance in T_g of bisphenol F epoxy resin with an increase of 11%. While the epoxy resin is crosslinked with the curing agent, the surface hydroxyl modified Al_2O_3 nanoparticles also react with the epoxy resin and the curing agent, forming a three-dimensional crosslinking structure, reducing the free volume inside the system. According to the free volume theory, the T_g of polymer refers to the limit temperature corresponding to the lowest free volume, so the doping of Al_2O_3 nanoparticles can significantly improve the T_g of epoxy resin [15]. As shown in Figure 4, the glass transition temperature of the composite increases with the decrease of the particle size of Al_2O_3 nanoparticle. As the specific surface area of Al_2O_3 nanoparticles increases with the decrease of particle size, the connection between the system and the filler becomes closer, and the binding force on the molecular chain further increases, which makes the surface effect of nano filler more obvious [16]. From the microscopic point of view, the cross-linking reaction restricts the movement of interfacial molecular chain, thus improving the glass transition temperature of epoxy resin composites. The smaller the particle size of Al_2O_3 nanoparticle is, the larger the unit surface area of interaction between Al_2O_3 nanoparticle and bisphenol F epoxy matrix is, hence the bonding strength between Al_2O_3 nanoparticle and bisphenol F epoxy matrix will be improved. Therefore, compared with the larger size of Al_2O_3 nanoparticle (1.17 nm, 1.34 nm, 1.5 nm), the rigid system formed by 1 nm Al_2O_3 nanoparticle and bisphenol F epoxy matrix is more stable.

3.2 Elasticity Modulus

Based on molecular dynamics, the elastic modulus of two types of polymers were calculated by static method. The specific methods are as follows: uniaxial tensile and uniaxial compressive

deformation of three-dimensional epoxy resin system along X, Y and Z axes respectively, and the corresponding data were counted. Shear deformation was generated in XY, XZ and YZ planes respectively, and the corresponding data were counted. Finally, the relevant parameters of the mechanical properties of the model can be obtained, and the strain relationship satisfies Hooke's law.

$$\sigma_i = C_{ij}\varepsilon_j$$

Bisphenol F epoxy resin and its composites can be approximately regarded as isotropic materials, and its stiffness matrix C_{ij} can be simplified as:

$$Cij = \begin{bmatrix} \lambda+2\mu & \lambda & \lambda & 0 & 0 & 0 \\ \lambda & \lambda+2\mu & \lambda & 0 & 0 & 0 \\ \lambda & \lambda & \lambda+2\mu & 0 & 0 & 0 \\ 0 & 0 & 0 & \mu & 0 & 0 \\ 0 & 0 & 0 & 0 & \mu & 0 \\ 0 & 0 & 0 & 0 & 0 & \mu \end{bmatrix}$$

Where λ， μ are elastic constant, and the formula is as follows:

$$\lambda = \frac{1}{6}(C_{12} + C_{13} + C_{21} + C_{23} + C_{31} + C_{32})$$

$$\mu = \frac{1}{3}(C_{44} + C_{55} + C_{66})$$

Therefore, the bulk modulus K, shear modulus G, young's modulus $E$and Poisson's ratio of epoxy resin are ν can be calculated by follow formula:

$$E = u\frac{3\lambda + 2u}{\lambda + u}$$

$$G = \mu$$

$$K = \lambda + \frac{2}{3}\mu$$

$$\nu = \frac{\lambda}{2(\lambda + u)}$$

The mechanical characteristics of five kinds of bisphenol F epoxy resins and their Al_2O_3 nanoparticle doped composite models were estimated using the methods and formulae described above, and the results are given in Figure 5. The Al_2O_3 nanoparticle may improve the mechanical characteristics of bisphenol F epoxy resin greatly, and the impact becomes more noticeable as the nanoparticle size decreases. Compared with the pure epoxy system, the young's modulus, shear modulus and bulk modulus of the composite system doped with 1nm Al_2O_3 nanoparticle increased by 101.38%, 48.86% and 15.72%, respectively. The results showed that using Al_2O_3 nanoparticles as filler could improve the overall density of the system and reduce the deformation caused by external forces. Because nano materials have larger elastic modulus, when added into epoxy resin as filler to form a composite system, they can play a supporting role in the epoxy resin, increase the deformation resistance of the materials, and improve the mechanical properties of the materials [17]. On the other hand, there is a strong interaction between Al_2O_3 nanoparticles and matrix at high crosslinking degree, and this effect is more obvious with the decrease of Al_2O_3 nanoparticle size, so the mechanical properties of the small particle size composite system are better.

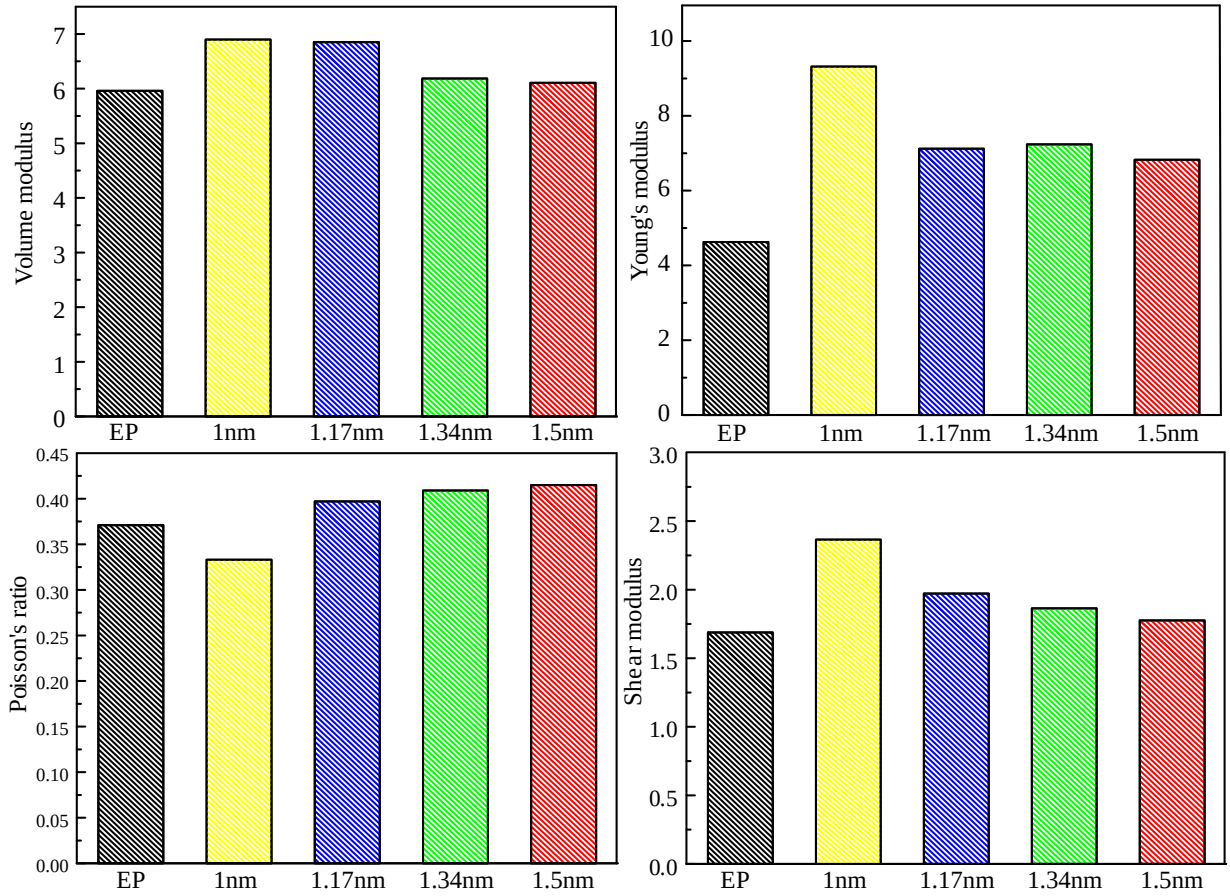


Figure 6. Elastic modulus of different systems.

3.3 Coefficient of Thermal Expansion

Coefficient of thermal expansion β_{CTE} (CTE), also known as linear elastic coefficient, represents the degree of expansion or contraction of materials, and is a critical metric for determining a material's thermal stability.

Table 1. CET of different systems

System	Rubbery State CET/K^{-1}	Glassy State CET/K^{-1}
EP	2.14×10^{-4}	8.92×10^{-5}
1.00 nm Al_2O_3/EP	2.97×10^{-4}	6.88×10^{-5}
1.17 nm Al_2O_3/EP	3.55×10^{-4}	6.97×10^{-5}
1.34 nm Al_2O_3/EP	3.67×10^{-4}	7.34×10^{-5}
1.50 nm Al_2O_3/EP	3.84×10^{-4}	7.95×10^{-5}

The calculation formula is as follows:

$$\beta_{CTE} = \frac{1}{V_0}\left(\frac{\partial V}{\partial T}\right)_p$$

Where: V_0 is the volume of the initial model, p is the pressure of the system, V is the volume of the model, and T is the temperature. In this study, the volume temperature relationships of epoxy resin model and four kinds of epoxy resin/Al_2O_3 model were fitted according to the parameters of annealing process.

The thermal expansion coefficients of each system in glass and rubber states were determined to further examine the changes in physical characteristics of bisphenol F epoxy resin and its composites during thermal shock, and the findings are displayed in Tab. 1. The coefficient of thermal expansion in glass state is less than that in rubber state for both epoxy resin system and epoxy/Al_2O_3 nanocomposite system. The coefficient of thermal expansion of epoxy resin and epoxy resin/Al2O3 composite in the glass state is smaller because the change in free volume of the material with temperature is small when the material is in the glass state. The free volume of the material rises with temperature when it is in the rubber state, resulting in a significant coefficient of thermal expansion of epoxy resin and epoxy resin/Al_2O_3 composite in the rubber state [18]. Similarly, the epoxy resin doped with smaller particle size Al_2O_3 nanoparticles performs better in terms of thermal expansion coefficient, which may be related to the small thermal expansion coefficient of Al_2O_3 itself and the strong interaction between small particle size Al_2O_3 nanoparticles and system molecules.

3.4 Dielectric Constant

The dielectric constant is a key quantity for describing dielectric materials' electrical characteristics [19]. The equilibrium epoxy resin and epoxy resin/Al_2O_3 models were further treated by molecular dynamics at 100 ps in NVT ensemble, and the dipole moments of each system in each MD process were calculated. According to the following formula, the dielectric constants of epoxy resin and epoxy resin/Al_2O_3 nanocomposite models were calculated:

$$\varepsilon = 1 + \frac{1}{3Vk_BT\varepsilon_0}\left[\langle\vec{M}^2\rangle - \langle\vec{M}\rangle^2\right]$$

Where: V is the volume of the model, T is the temperature of the model, k_B is the Boltzmann constant, ε_0 is the vacuum permittivity, $\langle\vec{M}^2\rangle$ is square of the average dipole moment, and $\langle\vec{M}\rangle^2$ is mean of the square of the dipole moment. The dielectric constant of epoxy resin and epoxy resin/Al_2O_3 nanocomposite model is shown in Figure 6. After calculation, the dielectric constant of bisphenol F epoxy resin is 5.9, which is consistent with the data of 2.5 ~ 6 observed by other researchers. The results show that doping Al_2O_3 nanoparticles decreases the composite dielectric constant. Because the dipole polarization of epoxy resin is dependent on the mobility of molecular segments, Al_2O_3 nanoparticle doping will impede this process. Furthermore, the hydrogen bond energy introduced by the modified Al_2O_3 nanoparticles is tightly coupled with the epoxy matrix, significantly reducing the system dipole.

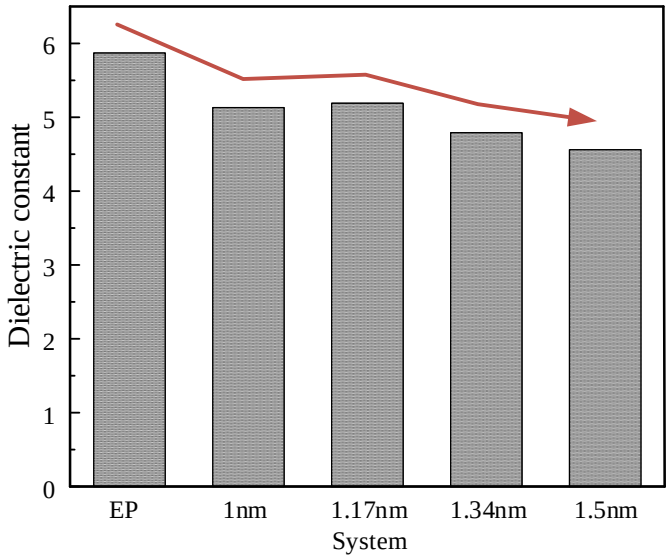


Figure 7. The dielectric constant of different system.

3.5 Radial Distribution Function

On the macro level, the order of matter can be studied by radial distribution function, while the radial distribution function can reflect the microstructure characteristics of the material. The calculation formula is as follows:

$$g_{A-B}(r) = \frac{n_B V}{4\pi r^2 dr N_B}$$

Where: N_B is the number of B atoms and V is the model volume. All atom RDF of bisphenol F epoxy resin and its Al_2O_3 nanoparticle doped composite is shown in Figure 7. The RDF and general trend of pure epoxy resin and nanocomposite systems are nearly same.

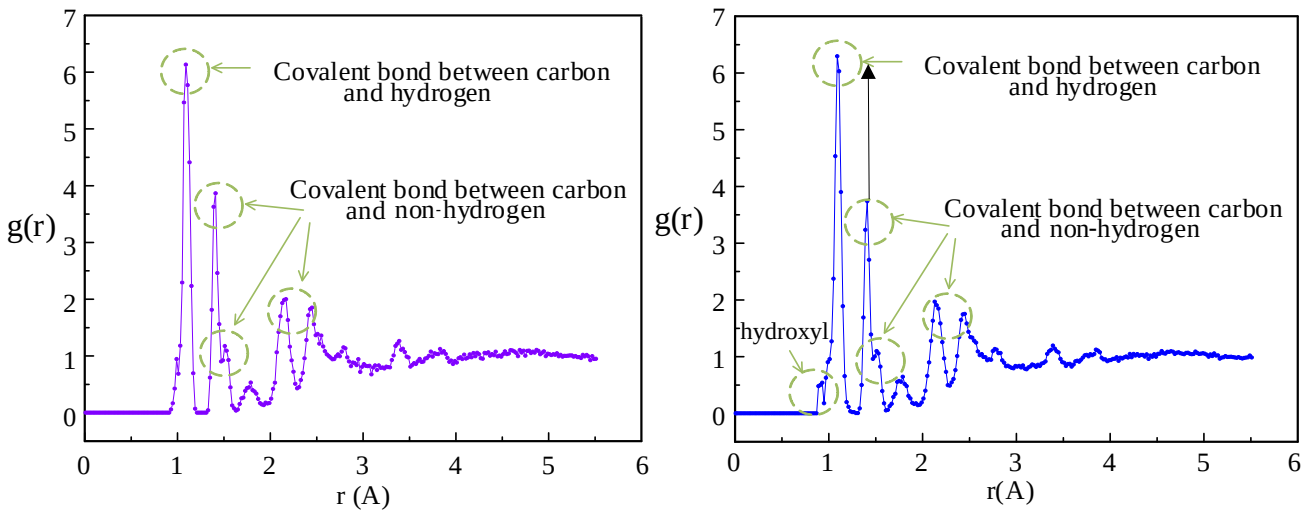


Figure 8. The atom RDF, (a) pure EP (b) EP with 1.5 nm Al_2O_3 nanoparticle.

In this paper, pure epoxy resin and nanocomposite with 1.5 nm Al_2O_3 nanoparticle system are selected for comparison. Among them, about 1.2 Å there is a peak value in each system, which corresponds to the bond length of covalent bond between carbon atom and hydrogen atom in each system and at 1.4 Å, 1.5 Å, 2.3 Å, 2.5 Å the peak value is corresponding to the bond length of covalent bond between carbon and other non-hydrogen elements. In contrast to the pure epoxy resin system, the composite system doped with Al_2O_3 nanoparticle has a peek at about 0.9 Å corresponding to the hydroxyl group in the Al_2O_3 nanoparticle, demonstrating that the hydroxyl group on the surface plays a role in the curing and crosslinking reaction to some extent [20]. In general, Al_2O_3 nanoparticle doping had little effect on the radial distribution of all atoms in epoxy resin.

3.6 Mean Square Displacement

MSD (mean square displacement) is the average of all possible mean square particle position shifts after a certain evolution. The mean square displacement represents the diffusion coefficient of the atom, which can be used to describe the ability of molecular segment movement in the system. In the system, the mean orientation shift can be expressed as:

$$d_{MSD} = \frac{1}{N}\sum_{i=1}^{N}(|R_i(t) - R_i(0)|^2)$$

Where: N is the number of atoms, R is the number of atoms, i is the displacement vector of any atom at the initial time, $R_i(t)$ is the displacement vector of any atom at time t.

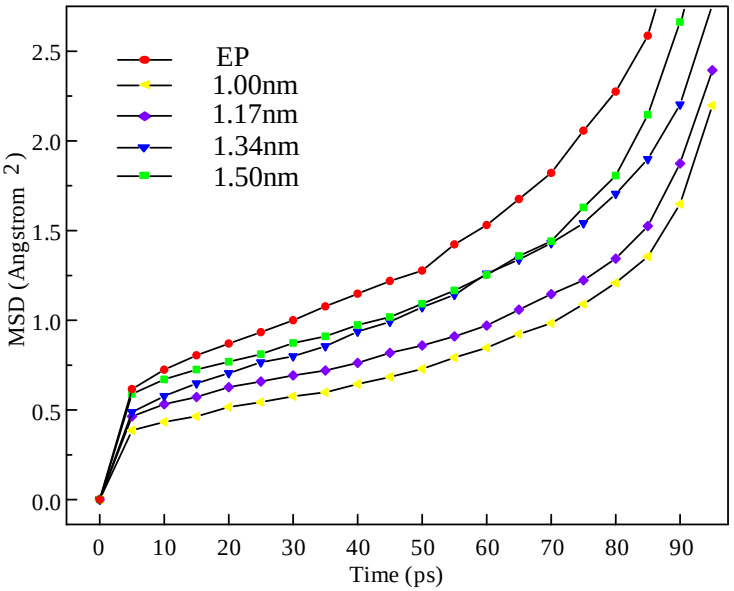


Figure 9. The MSD of different epoxy resin systems.

To study the influence of Al_2O_3 nanoparticle doping on the movement ability of molecular segments in bisphenol F epoxy resin composites, the MSD curves of each system were calculated. The

MSD results are shown in Figure 8. With the increase of time, MSD of each system showed the same upward trend. With the decrease of Al_2O_3 nanoparticle size, the MSD of the system also decreases and the MSD of the system with filler particle size of 1 nm is the smallest. This is because the size effect of Al_2O_3 nanoparticles becomes more apparent as particle size decreases and the 1 nm Al_2O_3 nanoparticle filler has the highest limitation on molecular chain mobility, which makes the movement ability of the system becomes weaker [21].

3.7 Cohesive Energy Density

Cohesive energy density is a physical quantity to evaluate the force between molecules, which reflects the interaction between groups. The stronger the contact force of the molecular segments and the higher the cohesive energy density, the higher the polarity of the groups in the molecule. The doping of Al_2O_3 nanoparticles may effectively increase the glass transition temperature and elastic modulus of epoxy resin, according to the prior estimate. To further explain this problem from the microscopic point of view, the cohesive energy density of each system was calculated.

$$\rho_{CED} = \frac{Ecoh}{V}$$

Where: $Ecoh$ is the cohesive energy of the molecular chain in the system, and V is the volume of the system. The cohesive energy density of each system was calculated by force module, and the results are shown in Figure 9.

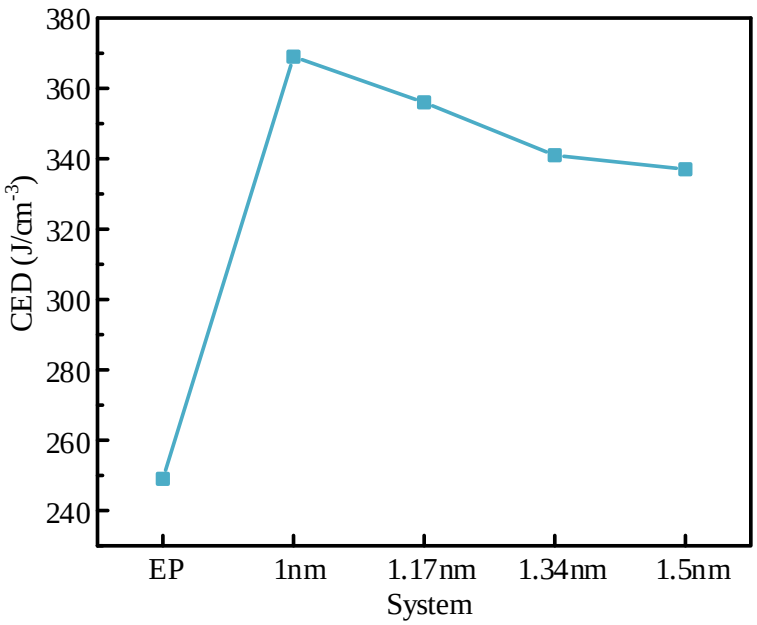


Figure 10. The CED of different epoxy resin systems.

Compared with pure epoxy system, the cohesive energy density of the system can be improved by doping Al_2O_3 nanoparticle, and the cohesive energy density of epoxy resin/Al_2O_3 nanocomposite system can be improved with the decrease of Al_2O_3 nanoparticle size. The results match those of the

glass transition temperature and elastic modulus tests. Therefore, the doping of Al_2O_3 nanoparticle can limit the movement of molecular chain of bisphenol F epoxy resin, make the molecular chain closely combined, and enhance the interaction of molecular chain [22]. From the microscopic point of view, it makes a reasonable explanation for Al_2O_3 nanoparticle doping to improve the performance of epoxy resin.

4. CONCLUSIONS

In this study, to analyze the thermal, mechanical, and electrical properties of bisphenol F epoxy resin nanocomposites for high temperature superconducting equipment insulation from a microscopic point of view, the model of bisphenol F epoxy resin was constructed, and the influence of Al_2O_3 nanoparticle doping on the insulation properties of bisphenol F epoxy resin was discussed under the condition of the same volume fraction. The improvement of thermodynamic properties of epoxy resin doped with Al_2O_3 nanoparticle was characterized by glass transition temperature, elastic modulus, thermal expansion system and dielectric constant. To explore the root cause, the microstructure parameters of bisphenol F epoxy resin and its Al_2O_3 nanoparticle doped composites were calculated: radial distribution function, mean square displacement and cohesive energy density. The following are the conclusions:

(1) The doping of Al_2O_3 nanoparticle can improve the glass transition temperature and elastic modulus of the composites and reduce the thermal expansion coefficient and dielectric constant. However, doping a single Al2O3 nanoparticle will not improve the electrical characteristics of the composites much.

(2) The beneficial effect on the thermodynamic characteristics of composites becomes more substantial as the size of Al_2O_3 nanoparticles decreases. The glass transition temperature, Young's modulus, shear modulus and bulk modulus of 1 nm epoxy resin/Al_2O_3 nanocomposites increase by 14%, 101.38%, 48.86% and 15.72%, respectively.

(3) Al_2O_3 nanoparticle doping alters the microstructure of epoxy composites at the microscopic level. Although Al_2O_3 nanoparticle doping does not significantly change the all-atom RDF of the nanocomposites, the changes of cohesive energy and mean square displacement indicate that Al_2O_3 nanoparticle doping can make the molecules bond more closely and effectively limit the movement of molecular segments in the composites.

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