



Thermodynamic simulations of SrTiO₃/epoxy nanocomposites with different mass fractions

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Abstract

Molecular dynamics simulations have been used to study the thermomechanical properties of SrTiO₃/epoxy nanocomposites with different mass fractions. The calculation results revealed that thermal conductivity increased with mass fractions and temperature, which was consistent with the measured experiment value. Motion of molecular chain segment of pure epoxy resin became intense with temperature increase. With the increase in mass fractions of incorporated SrTiO₃ nanoparticles, the mobility of molecular chain segment was significantly weakened below 500 K compared with pure epoxy resin model and the model of nanocomposite with a mass fraction of 25 wt% SrTiO₃ showed best structure stability. Dipole autocorrelation functions (DACF) of pure epoxy resin fluctuated slightly under 350 K, but substantial fluctuation happened over 500 K. With the incorporation of SrTiO₃ nanoparticles, the fluctuation of DACF of nanocomposites decreased drastically, maintaining a small value of 0.004 Debye, which demonstrated that structure stability of nanocomposites was strengthened under 500 K. Based on interface model between epoxy resin macromolecular ligands and crystal plane (110) of SrTiO₃, interface interaction energy was calculated to be −235.9 kcal/mol. To sum up, incorporation of SrTiO₃ nanoparticles could significantly improve the thermomechanical properties of epoxy resin and enhance structure stability of nanocomposites.

Keywords Molecular dynamics · SrTiO₃ · Nanocomposite · Thermomechanical properties · Thermal conductivity · Interface interaction energy

1 Introduction

The thermal conductivity of polymer-based composites depends not only on the thermal conductivity of the filler and polymer matrix, but also on the type, content, shape, and distribution of the filler [1–8]. The surface modification of the nanoparticles and their interface interaction with the polymer matrix will greatly affect the thermal conductivity of the composite system [9–11]. According to the different physical properties of the filler, many scholars have established a number of mathematical models to predict the thermal conductivity of the composite system [12–15].

Epoxy resin (epoxy, abbreviated as EP) has excellent electrical properties and thermal stability. It is widely

used in the manufacturing of insulation parts of various electrical equipments. It is experimentally found out that the composite of epoxy resin matrix can exhibit excellent insulation properties. At present, there are many simulation studies on the thermodynamic properties of composite materials. Liu W et al. used molecular dynamics simulation to study the changes of structure and energy during curing and cross-linking of epoxy resin, and predicted the glass transition temperature and mechanical properties of epoxy resin [16]. Chen S et al. used the molecular dynamics simulation method to analyze the interaction between the epoxy resin monomer system and the surface of carbon nanofibers. The results showed that the epoxy resin monomer will form an interface

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phase on the surface of the carbon nanofibers due to attraction effect, and the surface of the carbon nanofibers can inhibit the movement of monomer molecules in the interface phase [17]. Sun W et al. used molecular dynamics to simulate the structure, thermodynamics, and mechanical properties of carbon nanotubes/polyethylene composites and analyzed the variation with simulated temperature and mass fraction of carbon nanotubes. The mechanical properties of carbon nanotube/polyethylene composites show that the elastic constant tensor, elastic modulus and Poisson's ratio are much higher than those of pure polyethylene systems. Values of all mechanics properties become smaller with an increase in temperature and decrease in content of carbon nanotubes [18]. El-Hadek MA found the Young's modulus decreased monotonically over the entire range of microballoon volume fractions. The measurements, quasi-static as well as dynamic, are in very good agreement with Hashin–Shtrikman lower-bound predictions for two-phase mixtures comprising of matrix and randomly distributed spherical voids/pores [19].

Subramanian N et al. investigated the system comprising a combination of crystalline carbon nanotubes (CNTs) dispersed in epoxy polymer molecular based on molecular dynamics (MD). The ultra-high strain rate deformation approach is extended to the CNT-epoxy system at various CNT weight fractions, and the corresponding bond disassociation energy extracted from the simulation volume is used as input to a low-fidelity continuum damage mechanics model to demonstrate the bridging of length scales and to study matrix failure at the microscale [20]. Jung H et al. modified carbon nanotubes by plasma (no solution blending method) and successfully grafted N elements onto carbon nanotubes. The modified carbon nanotubes were then filled into epoxy resin matrix. High thermal stability and mechanical properties of composites were verified by experiments and molecular dynamics simulation methods [21]. Yao X F analyzed the structural properties of SiO₂-filled epoxy resin in terms of nanoparticle dispersion, free volume fraction, and interfacial interaction [22]. Wang Z further simulated the interfacial bond of SiO₂/epoxy composite model with molecular dynamics. The results showed that the chemical bond in the interface region can increase the glass transition temperature of the epoxy resin by 21 K when the graft ratio exceeds 5.8%, which enhances the thermal conductivity of the composite model. In the meanwhile, the interfacial chemical bond can reduce the mobility of epoxy resin molecular chains [23]. Wang Y simulated the interfacial thermal resistance and thermal conductivity of SrTiO₃ particles, showing that the size of the nanoparticles is closely related to the thermal conductivity [24].

Based on the previous research of our group, SrTiO₃/EP composites were prepared, in which the nanoparticles were surrounded by the cross-linked epoxy resin macromolecular structure. The thermal conductivity and dielectric properties of the composites were improved to some extent as the mass fraction of SrTiO₃ increased. Meanwhile, the surface modification of nanoparticles plays a crucial role in the performance improvement of composites [25]. In this paper, the molecular dynamics simulation of SrTiO₃/EP composite model with different filling mass fractions was conducted. The thermodynamic properties of the composite model were studied. The influence of nanoparticle loading amount on the composite structure was analyzed from the perspective of segment mobility and dipole autocorrelation function and the interface interaction energy was calculated, which proved the strong interfacial interaction between the SrTiO₃ nanoparticles and the polymer matrix.

2 Establishment of pure epoxy resin model

Firstly, a pure epoxy resin cross-linking model was established. The matrix molecule was bisphenol A epoxy resin (DGEBA), and the curing agent molecule was an adduct of diethylenetriamine and butyl glycidyl ether dubbed as 593 curing agent. The conventional ring-opening method was selected for cross-linking, and energy minimization was performed for the cross-linked model. The model was constructed with the following steps:

- (1) Monomer molecular models of DGEBA and 593 curing agent were established, respectively. The bisphenol A epoxy resin has an epoxy group on both ends, and the 593 curing agent molecule has a highly active primary amine group at one end. Figures 1 and 2 show the molecular structure of a single DGEBA monomer molecule and a 593 curing agent, respectively.

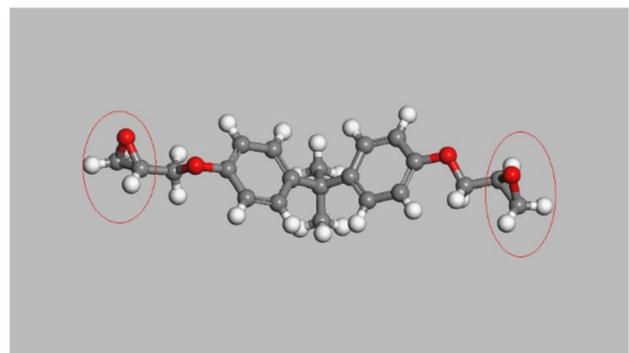


Fig. 1 Structure of a single molecule of DGEBA

groups and the carbon atoms after the ring opening of the epoxy group were named. When the cross-linking reaction was carried out, the initial cutoff distance was set to 3.5 Å, the maximum cutoff distance 7.0 Å, and the step length 0.5 Å. The displacements of the specified carbon and nitrogen atoms changed during the cross-linking process. If the distance between carbon and nitrogen atoms was from 3.5 to 7.0 Å, the activated carbon and nitrogen atoms can bind to form a CN covalent bond. The degree of cross-linking degree and the cutoff distance should be coordinated with each other, which ensures proper degree of cross-linking, and also save the time needed for the cross-linking process. If the degree of cross-linking was found to be relatively low, the cutoff radius value can be moderately increased to reach the preset degree of cross-linking faster. The Perl language script for epoxy resin cross-linking was provided by Accelrys, USA. Attention should be paid to the naming of the model and specific carbon and nitrogen atoms. With relevant physical parameters appropriately set, the Perl language script for epoxy resin cross-linking was coupled by modules including Discover and Forcite to achieve convergence to preset cross-linking value. The three-dimensional model obtained after cross-linking is shown in Fig. 5.

3 Establishment of SrTiO₃/EP composite model

Based on the previous research results of our team [25], it has been proved by experiments that filling the epoxy resin matrix with SrTiO₃ nanoparticles can improve the thermal and electrical properties of the composite.

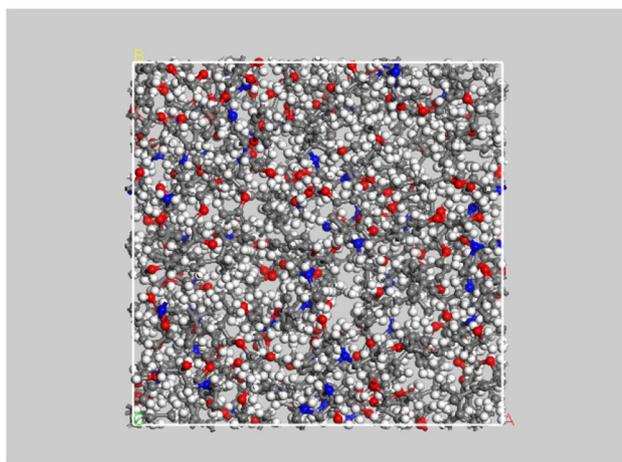


Fig. 5 Cross-linking model structure of pure epoxy

However, study of the microscopic mechanism on thermodynamic properties after filling is still insufficient. Therefore, models with same filling mass fractions of 7 wt%, 15 wt%, 25 wt%, and 40 wt% as in experiment were constructed using amorphous cell tool. Firstly, 20 bisphenol A epoxy resin molecules with polymerization degree of 0 and 20 bisphenol A epoxy resin molecules with polymerization degree of 1 were cross-linked and solidified with 593 curing agent. The target density value was set to 1 g/cm³, and the quality was set as Medium. The pure epoxy resin model after cross-linking and solidification is shown in Fig. 6a. In order to construct epoxy resin composite models with different filling mass fractions, the number of bisphenol A epoxy resin molecules and the number of filled nanoparticle models needs to be adjusted. Final epoxy resin composite models with different filling mass fractions can be obtained as shown in Fig. 6b–e. Then, the composite model was structurally optimized for 5000 steps, followed by 200 ps simulation on NPT ensemble,

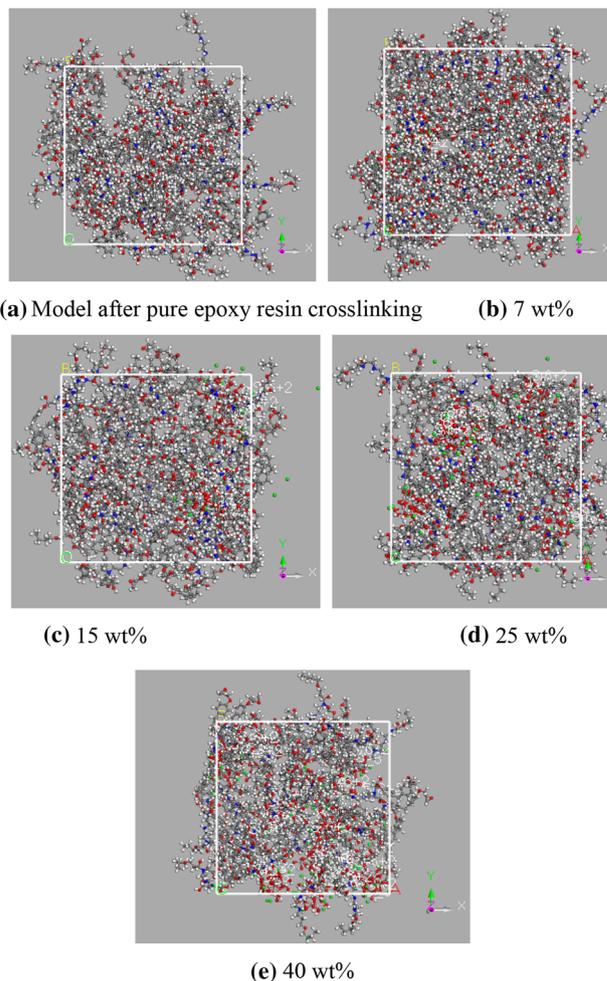


Fig. 6 Models of pure epoxy and SrTiO₃/EP nanocomposites with different mass fractions

and 300 ps simulation on NVT ensemble. The temperature control used the Andersen method and the pressure control adopted the Berendsen method with the pressure set to standard atmospheric pressure. The Newton equation was solved by the velocity Verlet algorithm. During calculation Van der Waals chose Atom Based method and Electrostatic chose Ewald method [5, 7, 9].

4 Simulation results

4.1 Thermal conductivity

The simulation of thermal conductivity was based on the RNEMD (Reverse Non-Equilibrium Molecular Dynamics) method. By changing the moving rate of two different molecules in the model, the molecule with slowest moving rate in one region was used to replace the molecule with fastest moving rate in the other region. The region where molecules move faster will cool down and the temperature of the region where molecules move at a slower rate will rise. The entire system will move the interaction heat from a high-temperature region to a low-temperature region. Finally, when the energy exchange can offset the energy reflux under the temperature gradient of the two regions, the system reaches a steady state, and the thermal conductivity corresponds to the energy flux divided by the temperature gradient.

In the simulation process, Berendsen was selected as the temperature adjustment method and the force field was set as Universal. Different temperature values were considered. The time interval of temperature exchange was 5 fs, and the number of time steps was 1000. Using the Forcite module, the NVT ensemble simulation was performed first, and then the 200 ps simulation was performed under the condition of the heating adjustment method Berendsen to avoid the drift of the module. The NVE ensemble simulation of 200 ps was conducted without the temperature adjustment mode and the system automatically calculated the energy flux (unit GW/m^2). The Forcite module was then used to run the molecular dynamics simulation "dynamics" to obtain the temperature gradient (in GK/m^2). Molecular dynamics simulation was performed using the last frame of the model structure in the model trajectory file. The initial rate was set as Random, the mass center point was picked, the energy exchange between the hot layer and the cold layer was calculated, the heat transfer data were saved, and the average temperature gradient was recorded. The thermal conductivity of the five models at different temperatures is shown in Table 1.

It can be seen in Table 1 that at room temperature of 300 K, the thermal conductivity of the pure epoxy resin

Table 1 Thermal conductivities of four models under different temperatures

Thermal conductivities ($\text{W}/(\text{m K})$)	300 K	350 K	400 K	450 K
Pure epoxy	0.20478	0.21125	0.22951	0.24482
7 wt%	0.28484	0.32487	0.35149	0.36527
15 wt%	0.38014	0.42705	0.47510	0.49254
25 wt%	0.47573	0.51478	0.54280	0.60147
40 wt%	0.58841	0.63418	0.68517	0.71426

and the composite models with filling mass fractions of 7 wt%, 15 wt%, 25 wt%, and 40 wt% was 0.20478, 0.28484, 0.38014, 0.47573, and 0.58841 $\text{W}/(\text{m K})$, respectively. The measured values of the thermal conductivity of the pure epoxy resin prepared in the previous experiment and those with filling mass fractions of 7 wt%, 15 wt%, 25 wt%, and 40 wt% were 0.20, 0.24, 0.29, 0.39, and 0.51 $\text{W}/(\text{m K})$, respectively [25]. The thermal conductivity of pure epoxy resin model at room temperature was consistent with the experimental measurements. With the increase in temperature and filling mass fraction, the thermal conductivity of each model gradually increases. When the filling mass fraction was 7 wt%, the thermal conductivity simulation value 0.28484 $\text{W}/(\text{m K})$ was close to the measured value 0.29 $\text{W}/(\text{m K})$, indicating the satisfactory agreement between simulation and experiment when the filling mass fraction is low. When the mass fraction exceeded 15 wt%, the simulation value was generally higher than the experimental measurement value. The main reason for the difference between the thermal conductivity simulation results and experimental measurements of composite samples is likely to be that the target density value in simulation was different from the density of the experiment sample. The size of the nanoparticles in the experiment sample kept in the range of 500 nm to 1 μm . The nanoparticle model established by the simulation process was small, and the smaller nanoparticles made the heat conduction path difficult to form. The periodic boundary conditions determined the direction of the temperature gradient but the actual measurement of the thermal conductivity of the composite samples did not have an ideal temperature gradient. Therefore, when the filling mass fraction is high, the thermal conductivity simulation value of the sample is higher than the experiment value. It can be seen from the overall simulation results that the simulated values of thermal conductivity show roughly the same trend as the measured values, which proves that the addition of SrTiO_3 nanoparticles can improve the thermal conductivity of the epoxy resin matrix.

4.2 Mobility of molecular segment (MSD)

Mobility of molecular segment can be indirectly reflected by the mean square displacement (MSD) as a function of optimization time at different temperatures. The atoms do not stay in a fixed position, but moves continuously. The MSD mean square displacement correlates with the atom's diffusion coefficient. It is commonly believed that as the degree of cross-linking increases, the MSD of the molecular segment in the system decreases.

The model shown in Fig. 6 was subjected to gradual cooling treatment in the range from 600 to 300 K. 100 ps NVT ensemble simulation was first conducted for the geometrically optimized model, and 200 ps NPT ensemble simulation was performed at the same temperature. The model obtained after NPT ensemble simulation was then used as the input of the next round simulation with same steps as in the first round for the following temperature. The cycle was repeated. In order to ensure the accuracy of the data, the temperature interval was set to 25 K, so that the cooling rate during the cooling simulation was 25 K/300 ps.

The MSD curves of each unit cell model extracted from the unit cell model after 100 ps NPT ensemble optimization at a series of temperatures during the cooling process are shown in Fig. 7. It can be seen that the movement of the pure epoxy resin molecular chain increased with the temperature, and the molecular chain movement increased slowly with simulation time. At room temperature, the molecular chain displacement was small. As the filling mass fraction increased, the molecular chain moved more slowly in the temperature range below 500 K. When the filling mass fraction reached 25 wt% or more, the movement of the molecular chain was almost solidified. It can be speculated that filling of the nanoparticles did not destroy the stability of the three-dimensional cross-linked structure, instead it enhanced the structure stability in the temperature range below 500 K. Under the effect of the thermal field, the movement of the molecular chain depends on the size of the free space, and the nanoparticle hinders the movement of the molecular chain to some extent.

Comparison of the curves at 600 K shows that the structure of the model was destroyed at 600 K, and the filling of the nanoparticles could reduce the mean square displacement at high temperature. However, at this time the epoxy resin had entered a highly elastic state and the filling of nanoparticles could not inhibit the movement of the polymer matrix molecules. Therefore, the MSD curve of 600 K was much larger than the MSD curve at other temperatures under the same simulation optimization time. At this time, the structure of the composite deteriorated, and the mechanical properties got weakened. As the filling

mass fractions increases, the segment mobility gradually decreases, and the three-dimensional network structure of the composite model becomes more stable.

Figure 8 shows the MSD value of five models vs temperature at constant optimization time of 80 ps. With temperature increasing, MSD value increased gradually. The MSD value of pure epoxy resin model at 500 K is 3.89 \AA^2 , while the MSD value of SrTiO₃/EP nanocomposites with mass fraction of 40 wt% is 1.48 \AA^2 that is more than twice value of pure epoxy resin. In summary, the SrTiO₃ nanoparticles significantly reduce the segmental mobility of the composites and enhance the mechanical properties of the composites below 500 K.

4.3 Dipole autocorrelation function

The model of nanocomposites belongs to the hybrid model of organic polymer and inorganic particles, which are mixed by COMPASS force field within a unit cell with the fixed size. The MSD equation reflects the movement of the molecular chain at different temperatures in simulation time. The movement of the molecular chain reflects the molecular expansion and contraction to a certain extent, depending on the size of the free space. If the free space in the composite model is small, the movement range of the molecules is correspondingly reduced. In order to further analyze the change in the structure of the composite after nanoparticle filling, a dipole autocorrelation function (DACF) was introduced. The dipole autocorrelation function can be used to explain the rotation of the dipole in the composite model. The rotation of the dipole reflects the transformation of the polymer conformation at different temperatures. The DACF function compares the positional transformation between clusters in the composite model to describe the change of the dipole moment. The physical meaning of the function can be explained as the self-rotation of the molecule. Figure 9 shows the DACF curves of a pure epoxy resin model with a 15 wt% composite model and a 40 wt% composite model.

It can be seen in Fig. 9a that after the 25 ps NPT ensemble simulation, the pure epoxy resin model had little fluctuation in the DACF curves below 350 K. When the temperature exceeded 400 K, the fluctuation amplitude of the pure epoxy resin model DACF started to increase. During the 25 ps simulation period, the amplitude of the DACF curve fluctuation at the temperature of 400 K was 0.075 Debye, and the fluctuation amplitude reached 0.1 Debye at 500 K, 0.2 Debye at 600 K. The dipole moment of the pure epoxy resin changes greatly at high temperature. As the temperature increases, the molecular chain motion gets intense, and the dipole also begins to shift. When the temperature exceeds 500 K, the dipole moment has significant change. The epoxy resin DACF curve shows that

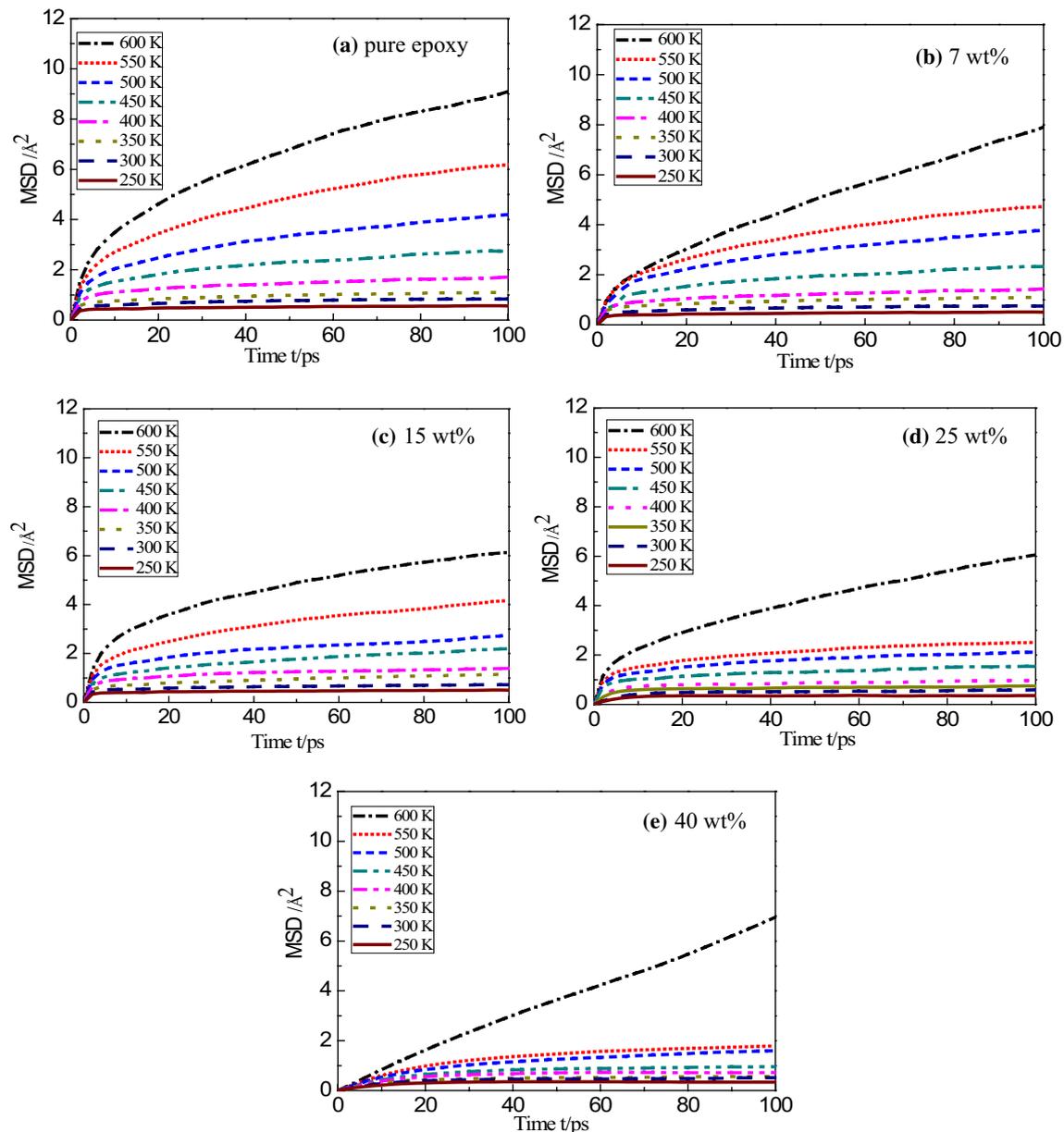


Fig. 7 MSD curves of five models under different temperatures **a** 0 wt%, **b** 7 wt%, **c** 15 wt%, **d** 25 wt%, **e** 40 wt%

when temperature exceeded 400 K, the DACF curve began to fluctuate greatly, and the structure gradually changed on the downward path of deformation.

Compared with the pure epoxy resin model, the composite model with a filling mass fraction of 15 wt% had a DACF curve with smaller fluctuation amplitude, which kept within the range of 0.004 Debye, as shown in Fig. 9b, indicating that the relative displacement of the dipole became smaller after filling of nanoparticles, and the three-dimensional cross-linked structure of the composite model had better structure stability. Figure 7 shows the 15 wt% composite model had weaker segment mobility than the pure

epoxy resin model. The DACF curve further shows that the dipole oscillation was also weakened.

The small fluctuation amplitude of the DACF curve of the composite model also indicates that the dipole polarization of the composite model is weakened to some extent. The dielectric constant of the composite is higher than that of the pure epoxy resin due to the interfacial polarization. Dipole polarization is an energy-consuming process, and pure epoxy resin has poor thermal conductivity. Therefore, under the high-frequency electric field, pure epoxy resin is prone to be overheated and structural stability is deteriorated. After filling SrTiO₃ nanoparticles, the

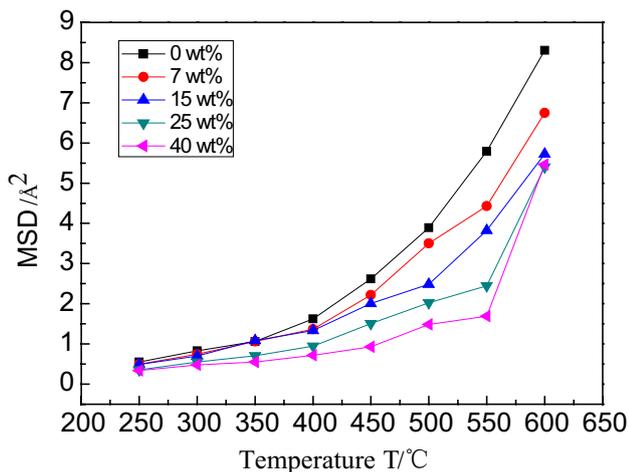


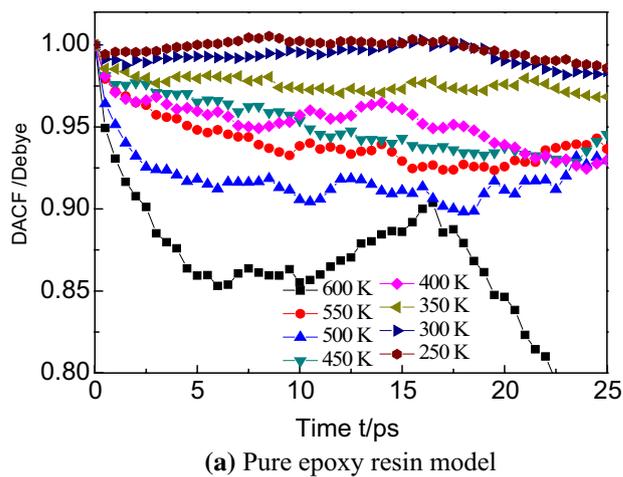
Fig. 8 MSD value of five models vs temperature at constant optimization time of 80 ps

structure of composite model gets compact, the mechanical properties are enhanced, and the segment mobility and dipole polarization are relatively weak. For 40 wt% composite model, the range of DACF curve fluctuation is 0.002 Debye at 550 K within the 25 ps simulation period, as shown in Fig. 9c. The increase in the filling mass fraction made the dipole steering more difficult. When the temperature exceeded 600 K, the composite model with a filling mass fraction of 40 wt% exhibited a fluctuation amplitude of 0.008 Debye, indicating that the highly filled composite model structure became unstable when the temperature exceeded 600 K. On the whole, the MSD curve and the DACF curve analysis of the composite model further indicate that the structure stability of the composite increases with the increase in the filling mass fraction.

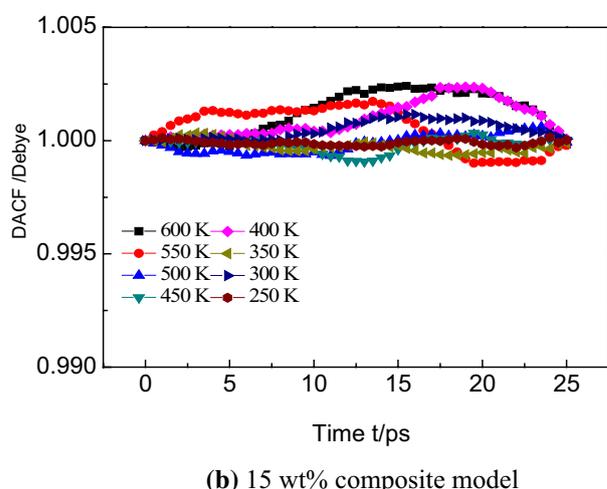
4.4 SrTiO₃/EP interface interaction energy

The interface interaction between epoxy resin and SrTiO₃ nanoparticles needs to be simulated by constructing interface model. Firstly, a bisphenol A epoxy resin with polymerization degree of 1 and a 593 curing agent molecule were used to construct a solidified resin model by manual ring opening, as shown in Fig. 10. Then, homopolymer was selected, the chain length was set to 10, and the number of chains was set to 1. The long-chain epoxy resin model was finally obtained as shown in Fig. 11.

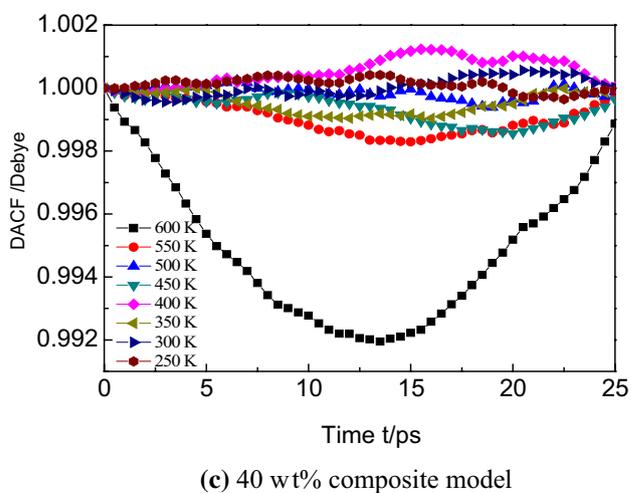
To calculate the interface interaction energy between the (110) crystal plane of SrTiO₃ crystal and pure epoxy resin molecule, the interface model with lattice size matching lattice constant of SrTiO₃ should be constructed first. Because the lattice constant of SrTiO₃ is 0.39 nm, a, b, and c in the cell parameters were set as 39. The temperature was set to 298 K, the cell type sets to confine layer, and the number of



(a) Pure epoxy resin model



(b) 15 wt% composite model



(c) 40 wt% composite model

Fig. 9 DACF curves of pure epoxy model and nanocomposites models with different mass fractions

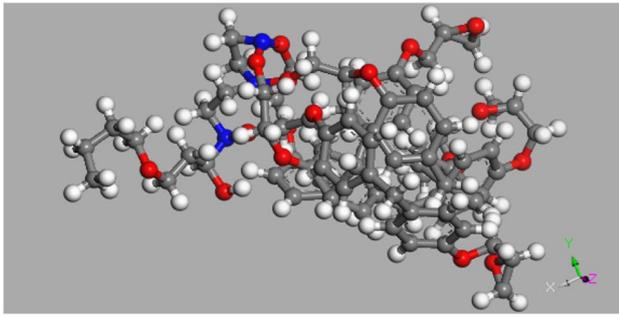


Fig. 10 A single cross-linked model of epoxy resin after reaction of one DGEBA molecule and one curing agent molecule of 593 type

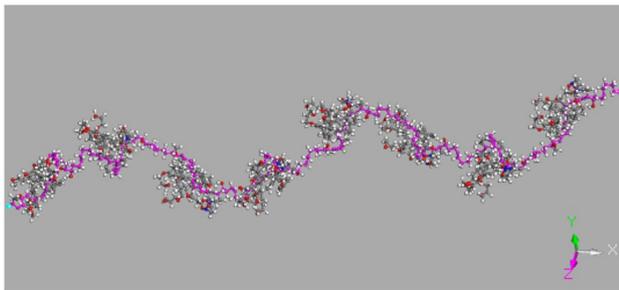
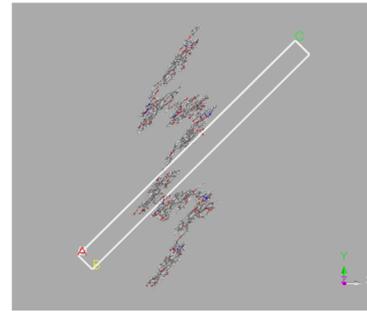


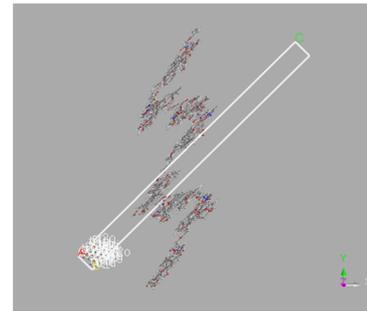
Fig. 11 Model of a long cured epoxy resin molecule with polymerization degree of 10

configurations sets to 1. The long-chain molecular model structure of epoxy resin is shown in Fig. 12a. The polymer was then applied to the surface of the epoxy molecule using a layered molding tool. The vacuum layer thickness was set to be 30, and the epoxy resin interface model was obtained as shown in Fig. 12b. Figure 12c shows the SrTiO₃ (110) crystal plane structure after removal of the polymer.

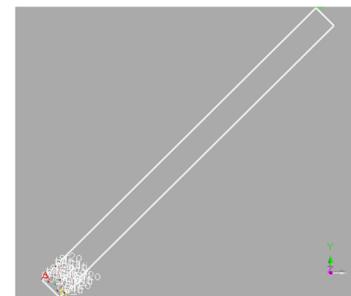
The interface interaction energy between the epoxy resin macromolecular group and the SrTiO₃(110) crystal plane was calculated by the interface model of Fig. 12. The Discover module was selected and the total potential energies of the three models were calculated in the energy tab. The total potential energies of the three models are: $E_{\text{total}} = 43104163.8$ kcal/mol, $E_{\text{polymer}} = 43102625.3$ kcal/mol, $E_{\text{surface}} = 1774.4$ kcal/mol. Then, $E_{\text{interaction}} = E_{\text{total}} - (E_{\text{surface}} + E_{\text{polymer}}) = -235.9$ kcal/mol. The epoxy resin molecule has a strong interface interaction energy with the nanoparticle (110) crystal plane and therefore, the composite material exhibits strong mechanical strength.



(a) Epoxy resin macromolecular chain single crystal model



(b) SrTiO₃/EP interface model



(c) SrTiO₃(110) crystal plane model

Fig. 12 Pure epoxy resin model, SrTiO₃(110) crystal plane model, and SrTiO₃/EP interface model

5 Conclusion

- (1) The thermal conductivity of the SrTiO₃/epoxy composite model is consistent with the experiment value. The thermal conductivity increases with the increase in the filling mass fraction and the temperature.
- (2) The mobility of pure epoxy resin molecular segment increases with the increase in temperature. As the filling mass fraction increases, the mobility of molecular segment gets weaker in the temperature range below 500 K. When the filling mass fraction exceeds 25 wt%, the mobility of molecular segment is dramatically decreased. Besides, with the increase in the filling mass fraction, the fluctuation amplitude of the DACF (dipole autocorrelation function) curve of the

composite model below 500 K is relatively small compared with pure epoxy resin model. On the whole, the MSD curve and the DACF curve analyses of the composite model further indicate that the structure stability of the composite increases with the increase in the filling mass fraction.

- (3) The interface interaction energy between the macromolecular group of epoxy resin and the crystal plane of SrTiO₃(110) was obtained by simulation of interface model, reaching -235.9 kcal/mol. It indicates that filling of the SrTiO₃ nanoparticles enhances the thermodynamic properties of the composite.

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Compliance with ethical standards

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

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