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Effect of graphene on thermal stability and mechanical properties of ethylene-vinyl acetate: a molecular dynamics simulation

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Abstract

Graphene has an important positive impact on improving polymer material properties, making the application of composite materials widely available. This paper investigates the influence of graphene on the thermal and mechanical properties of Ethylene-vinyl acetate (EVA) by the molecular dynamics (MD) simulations. The thermostability and mechanical properties of the graphene/EVA nanocomposites are analyzed in terms of the glass transition temperature ($T_g$), mean-square displacement (MSD), modulus, interfacial binding energy (IBE), stress-strain relationship, yield strength, and tensile strength. The influences of the size of graphene on the thermal stability and mechanical properties of EVA are analyzed and discussed. The simulation result indicated that the glass transition temperature, modulus, yield strength, and ultimate strength of the nanocomposites are higher than that of pristine EVA, which is in good consistent with recent experiments. We attribute this finding to the fact that the strong interfacial bonding of graphene to EVA limits the fluidity of the EVA chains and improves the thermal stability and strength of the graphene/EVA composites. The incorporation of graphene enhanced the thermal stability and mechanical properties of EVA.

1. Introduction

Ethylene-vinyl acetate (EVA) is one of the most commonly used thermoplastic elastomer and plastic materials [1]. Its good physical properties, including light-weight, low moisture absorption and UV resistance, have attracted a huge surge of research interest [2–5]. It has been applied in encapsulating electrical cables, shoes, component packaging, corrosion protection, and water-proofing. However, there are still some shortcomings for the mechanical properties and thermostability for the pure EVA [5, 6]. Defects such as low mechanical strength and poor toughness limit their applications. In order to solve these problems, different types of nanofiller (low-grade magnesium hydroxide, bacterial cellulose, multi-walled nanotubes etc.) have been used to enhance the mechanical properties of the EVA [7–9]. The tensile strength can only be improved by about 3–12 MPa, which still does not meet requirements of the strength and toughness for the cable packaging, device packaging under the extreme environment.

Many studies [10–14] have shown that a small amount of graphene can obviously enhance the thermal stability and mechanical properties of polymer nanocomposites by using molecular dynamics (MD) simulations method. And the modulus, glass transition temperature, tensile strength, and toughness were also be improved. Graphene is a two-dimensional carbon nanofiller, the atoms with the $sp^2$ bonded are densely arranged in a honeycomb lattice [15–17]. The excellent electron surface density [18] and $sp^2$ covalent bond [19] produced the unique features for the graphene, such as the excellent mechanical flexibility, electroconductivity, thermal conductivity, and optical transparency [20–23]. In addition, Lee et al. [19] demonstrated the graphene had a fantastically high ultimate strength of about 130 GPa and Young’s modulus of about 1 TPa. Moreover, the high specific area of the 2D stratiform structure enables the graphene to be a latent enhancement in nanocomposites.
[10, 17, 24]. Therefore, adding the nano-graphene sheet to EVA would be an effective way to improve the mechanical properties and thermostability of EVA.

Recently, some experimental results have also demonstrated that small amounts of graphene in the EVA matrix will improve the elastic modulus and the hardness of the polymer matrix [5, 25, 26]. However, theoretical studies on the internal mechanisms of the effects of graphene on the thermal stability and mechanical properties of EVA are very limited. Meanwhile, the molecular mechanism of studying the effect of graphene on EVA performance is of great significance for the application and design of composite materials. Furthermore, Young et al [27] and Lin et al [28] have shown that monolayer graphene can significantly enhance the mechanical properties of polymers due to its strong interfacial stress transfer capability. Experimental studies by Wang et al [29] show that the toughness enhancement of polymers is closely related to the size of graphene sheets.

Therefore, in this work, the sizes effects of monolayer graphene on the glass transition temperature, mean-square displacement, modulus, interfacial binding energy, yield and tensile strength of EVA are studied by MD simulations. The MD explanations of the influence of graphene on the thermal stability and mechanical properties of EVA is given and discussed, and hope more parameters can be given to experiment.

2. Modeling and simulation details

2.1. Computational method

The MD simulation is applied to research the interaction between graphene and polymer at the atomistic level, it provides available knowledge for the exploitation of graphene/polymer nanocomposites. Here, the MD simulation was on account of the Forcite module in Materials Studio 8.0 (MS 8.0) [30]. The condensed-phase optimized molecular potentials for atomistic simulation studies (COMPASS) [31] was applied to carry out force-field calculations. The COMPASS is a first ab initio force-field that has been tested and verified [32–34], it precisely predicts the multifarious condensed-phase and gas-phase characteristics of most familiar organic and inorganic polymer materials [31, 35–37].

For structure optimization process, the convergence tolerance is utilized with displacement of 5 × 10^{-5} Å and energy convergence of 1 × 10^{-4} kcal/mol. The stress is 0.005 GPa. The maximum number of iterations is 100,000. By employing atom-based van der Waals and electrostatic summation methods, the force field of COMPASS is utilized for simulation at fine standard of quality [14].

2.2. Force field

The quantum-mechanics technique is able to precisely simulate an interacting particles system, but this method is usually feasible only in systems where hundreds of particles interact. The primary purpose of simulating these systems, including a good deal of particles, is usually to acquire the bulk properties of systems. These properties are mainly determined by the position of nucleus [38]. In this case, if we can get a reasonable force field based on physical approximation, we can understand the performance of the system well. Applying such a forcefield can produce a series of system structures, this is corresponding to the statistical description of quantum mechanics [39]. In summary, a key point is the selection of force fields in atomic simulation of multi-particle systems, a simple description of which is showed in the part.

Generally, the total potential energy of the system is expressed as follows [40]:

$$E_{\text{total}} = E_{\text{valence}} + E_{\text{cross-\_term}} + E_{\text{nonbond}}$$

(1)

where the valence energy is expressed by $E_{\text{valence}}$, $E_{\text{cross-\_term}}$ is on behalf of the cross-term interaction energy, the nonbond interaction energy can be represented by $E_{\text{nonbond}}$. The COMPASS forcefield employs following expressions for different constituents of the potential energy [31, 35, 40]:

$$E_{\text{valence}} = \sum_{b} \left[ K_2 (b - b_0)^2 + K_3 (b - b_0)^3 + K_4 (b - b_0)^4 \right]$$

$$+ \sum_{\theta} \left[ H_2 (\theta - \theta_0)^2 + H_3 (\theta - \theta_0)^3 + H_4 (\theta - \theta_0)^4 \right]$$

$$+ \sum_{\phi} \left[ V_1 \left[ 1 - \cos(\phi - \phi_1^0) \right] + V_2 \left[ 1 - \cos(2\phi - \phi_2^0) \right] \right]$$

$$+ V_3 \left[ 1 - \cos(3\phi - \phi_3^0) \right] \right] + \sum_{\chi} K_\chi \chi^2 + E_{UB}$$

(2)
In order to explore the influence of graphene on EVA performance, we use ethylene-vinyl acetate (EVA) as a matrix. The molecular chain, EVA (C260H496O26), with 13 repeating monomers in each EVA molecular chain, are selected in the study. We use a simple model to represent the EVA chain without changing the original structural parameters, as shown in figure 2. Eight ethylene molecules and one vinyl acetate molecule are randomly distributed in the box. Figure 2(a) shows the orthogonal model structure of three EVA chains in EVA composite systems are 0, 5, 9, and 14 wt%, the density is 0.95 g/cm³. This is the same as the sample density of 0.95 g cm⁻³ used by Azizi et al [41] in the experimental study of graphene/EVA composites. The simulation box of EVA consisted of 9384 atoms by expanding the cell. Then we embed graphene sheet into the EVA matrix, the sizes of graphene sheet were changed from 4 × 4 (C48H18), 6 × 6 (C96H26), to 8 × 8 (C160H34), the graphene sheets respectively with dimensions of 19.21, 26.43, and 37.70 Å are displayed in figures 1(b)–(d). The dangling bonds in the graphene pores easily could be saturated with H atoms in the atmosphere [34], and in order to avoid the unsaturated boundary effect, hydrogen atoms are added at the end of graphene [42]. The systems in which the mass fraction of graphene in the four graphene/EVA composite systems are 0, 5, 9, and 14.
wt%, respectively, have been designated as EVA, EVA44Gr, EVA66Gr, and EVA88Gr. The structures of EVA and EVA66Gr are illustrated in figures 2(a) and (b). Table 1 shows the atomic force field types of the four systems in compass force field.

### Table 1. Atomic force field types of the four systems in compass force field.

| System          | h1 | c4 | c43o | o2s | c3' | o1= | c40 |
|-----------------|----|----|------|-----|-----|-----|-----|
| EVA             |    |    |      |     |     |     |     |
| EVAxGr (x = 44, 66, 88) | c3a | h1 | c4   | c43o | o2s | c3' | o1= | c40 |

3. Results and discussion

3.1. Thermal stability of EVA and graphene/EVA polymer nanocomposites

Glass transition temperature ($T_g$) is a physical characteristics of the amorphous polymer translates from a glassy state to an elastomeric state. Since the viscoelasticity of the polymer is greatly sensitive to temperature near $T_g$, $T_g$ is popularly used as the reference standard [43, 44]. This correlation between thermal stability and mechanical properties and $T_g$ of materials has been presented and applied to nanoscale polymers [11, 45, 46]. Differential scanning calorimetry (DSC) is often used in experiments to determine the glass transition temperature ($T_g$) of nanocomposites [29, 47]. In this part, the influence of the graphene on the $T_g$ of polymer nanocomposites was investigated by MD simulations. The temperature was gradually changed from 500 to 125 K with a differential temperature of 25 K. In each step, the system is placed in NPT ensemble and relaxed for 1 ns to achieve an energy minimization structure. The density of the nanocomposites as a function of temperature is indicated in figure 3.

The $T_g$ is considered as the crossing point of the two linear curves of the material by taking a bilinear curve fitting of the data [48].

From figure 3, it can be seen that the $T_g$ of the nanocomposite increases with the growth in concentration of graphene. The current $T_g$ value of pure EVA is about 238 K (−35.0031 °C), which is similar to the DSC experimental value of −33.1 °C of Agroui et al [49] and is within the range of $T_g$ values tested by Stark et al [50]. This means that current cooling rate and method are effective to determine $T_g$ of the EVA systems. The $T_g$ is about 300 K for the EVA66Gr system, increased by about 62 K compared with the pure EVA system. It was found that the $T_g$ increases obviously owing to the existence of graphene.

Normally, the $T_g$ of the polymer is closely related to the mobility of the chains. The main reason for the increase in $T_g$ is that the large interfacial area of graphene affects the behavior of the surrounding polymer matrix, the free movement of polymer chains is limited by graphene [11, 51]. The recent experimental [5, 26, 29] and theoretical [42, 52] results shows that the interaction between the polymer molecules and the embedded graphene can confine polymer chains and reduce the chain mobility. And the similar experimental results of the graphene/EVA system by Kuila et al [5] and Sabet et al [26] show that the good dispersion of graphene limits the segmentation motion of the EVA chains, and enhances the interaction between the graphene and the EVA chains, increasing the glass transition temperature of EVA.

Meanwhile, the behavior of the EVA chains around the graphene can be described by mean-square displacement (MSD). Figure 4 shows the MSD of EVA chains as a function of time for the pure EVA and graphene/EVA composites systems at 350 K, respectively. The results also confirm that the addition of graphene limit the fluidity of the EVA molecular chains. In addition, Zhang et al [53] recently studied the thermophysical
properties of paraffin/EVA/graphene nanocomposites by MD simulations. And their simulation results show that when the content of graphene is higher than 3.6 wt%, graphene showed a limiting effect on the mobility of EVA molecular chains, which is consistent with our simulation results that graphene has a limiting effect on EVA molecular chains mobility. And the limiting effect of graphene is related to its size, which is consistent with the effect of graphene on the T_g of EVA. Among MSD is defined as \[ \text{MSD} = \langle [r(t + t_0) - r(t_0)]^2 \rangle \] [54], where \( r(t_0) \) is the initial coordinate and \( r(t + t_0) \) the position of chain atoms after a time \( t \).

The increased \( T_g \) for the graphene/EVA system upon the addition of graphene is interpreted as the result of the strong interactions between the graphene and EVA chains, and the interactions reduced the molecular mobility and the flexibility of the polymer chain nearby the graphene. As can be seen from figure 3, the increase of glass transition temperature is closely related to the size of graphene. This result is supported by the experimental conclusions of Wang et al [29] and Yousefzade et al [55] Compared to the other two types of nanocomposites, EVA66Gr nanocomposites show higher \( T_g \). Based on the above discussion, this strong interaction leading to high glass transition temperature may be related to the sizes of graphene.

The interaction between graphene and polymer can be assessed by their interfacial binding energy. To explore the impact of graphene size on the interaction between EVA molecules and graphene, the interfacial binding energies of different sizes graphene and EVA were calculated in this part. Normally, the energy of interaction is evaluated by the discrepancy between the total potential energy of nanocomposites and the potential energy for graphene and polymer, as shown below [38, 39]:

![Figure 3. Calculation of glass transition temperature for EVA and graphene/EVA nanocomposites. (a) EVA polymer. (b) EVA44Gr nanocomposites. (c) EVA66Gr nanocomposites. (d) EVA88Gr nanocomposites.](image1)

![Figure 4. The mean-square displacement (MSD) of EVA chains as a function of time for pure EVA and graphene/EVA composites under the temperature of 350 K.](image2)
where the total potential energy of the composites is denoted by \( E_{\text{total}} \). Interfacial interaction occurs when graphene is in contact with EVA polymer. \( E_{\text{polymer}} \) and \( E_{\text{graphene}} \) indicate the potential energy of EVA polymer and graphene sheet respectively. When graphene and EVA polymer are not in contact, they have their own energies. \( E_{\text{polymer}} \) could be computed individually in the simulation unit without the graphene. \( E_{\text{graphene}} \) was calculated without the polymer in a similar way. The total interaction energy can be expressed as \( \Delta E \), which is twice the product of the interface bonding energy \( \gamma \) and contact area \( A \). Because graphene is a lamellar structure, the area of contact \( A \) is twice the superficial area \( S \) of graphene [56]:

\[
\gamma = \frac{\Delta E}{2A} = \frac{\Delta E}{4S}
\]

The total energy of EVA, EVA44Gr, EVA66Gr, EVA88Gr nanocomposites, and the four monomers were calculated, respectively. Firstly, the four systems were simulated in NPT ensemble with an original density of 0.95 g cm\(^{-3}\). The time of MD simulation using NPT ensemble (\( P = 1 \) bar, \( T = 600 \) K) was 3000 ps, and the time step was 1 fs. This step aimed to relaxedly condense the structure of the composites to produce a primal amorphous polymer with the less residual stress and reasonable density. This step could make the original state of the four systems reach equilibrium. Then, the composites were simulated in NVT ensemble and balanced for 1000 ps, and the time step was 1 fs. The step was mainly to construct a zero original pressure state employing NVT ensemble. The temperature was changed from 600 to 200 K gradually with a differential temperature of 50 K applying the relaxation method above. Similarly, the energy of each monomer was calculated only in the NVT ensemble excluding their own influence. Such the energy of the composites was minimized at each step to realize the most powerful combination between the polymer and the graphene. At last, the energies of interaction and interfacial bonding were well computed, given in figures 5(a), (b).

The energy interaction and interfacial bonding energy of the graphene/EVA nanocomposite systems respectively as a function of temperature for are indicated in figures 5(a) and (b). From figure 5(a), for the EVA88Gr polymer, it is observed that the attractive interaction be molecules tween EVA and graphene only decreases slightly with increasing temperature. Ever for EVA44Gr and EVA66Gr polymers, the effect of temperature becomes negligible. It is due to the steady intermolecular interaction between graphene and EVA that cannot be affected by the temperature, in the whole temperature range. However, this effect is more significant for EVA44Gr and EVA66Gr. From figure 5(b), it seems the interfacial bonding energy between EVA molecules and graphene reduce weakly with increasing temperature. The interfacial bonding energy of EVA44Gr and EVA66Gr polymers was less affected by temperature than that of EVA88Gr, relatively. In addition, the interface binding energy of the EVA66Gr is stronger than that of the other two types polymers, it was remained within 0.49–0.52 kcal/molÅ\(^2\) with temperature increasing. The results demonstrate the enhanced adhesion of the interface, which is consistent with the \( T_g \) results [29]. And the physical barrier effect of graphene can also improve the thermal stability of the composite system [57]. Moreover, the high interaction between graphene and EVA may have a noticeable influence on the mechanical properties of nanocomposites, it will be elaborated in the next section.

3.2. Mechanical properties of EVA and graphene/EVA nanocomposites

In order to research the effect of the interaction between EVA molecules and graphene on mechanical properties of composites, the transverse tensile tests of the four polymer systems were carried out in this part by MD.
simulation. The temperature range of tensile simulation is above and below $T_g$ of nanocomposites, in which the temperature was varied from 350 to 200 K with a temperature difference of 50 K. Before stretching, the initial structure is relaxed for 1 ns under the NPT ensemble of 500 K and 1 bar to ensure that the chain is completely relaxed. The time step is 1 fs. Then, using the Nose-Hoover thermostat, the system gradually cooled from 500 to 200 K under the NPT system with a cooling time of 1 ns, the temperature interval is 50 K. Finally, a uniaxial extension is performed at the selected four temperature point above with a constant strain rate. The uniaxial stretching time is 35 ns. The constant strain rate of $10^8$ s$^{-1}$ and the maximum strain of 3.5 were applied to these four systems. Figure 6 shows the stretching process of EVA66Gr cooled to 300 K after high temperature relaxation. The stress-strain curves of these nanocomposites generated from MD simulation are shown in figures 7(a)–(d).

Yield strength and tensile strength are important parameters to measure the mechanical properties of polymer composites [58, 59]. For amorphous polymeric materials, the yield stress is calculated using the 0.2% offset method, and the maximum stress is considered to be the tensile strength [60]. The yield strength of EVA at 350 K is approximately 5 MPa, it is quite close to the experimental data of about 3 MPa [7]. This means that the current strain rate and method can effectively simulate the tensile test of the systems. From figure 7(a), the yield and tensile strength of the pure EVA increase obviously with decreasing of temperature, and the elastic modulus increases clearly. This is caused by a decrease in temperature, a decrease in fluidity of the polymer segment, and an increase in viscoelasticity. From figures 7(b)–(d), with the addition of graphene, the yield strength and tensile strength of nanocomposites increase significantly. This trend has also been supported by the recent similar experiments by Sabet et al [26] and Tayebia et al [24]. That is, a small amount of graphene can limit the fluidity of the EVA chains, improve viscoelasticity and interfacial interaction, and increase the modulus and strength of the composite. Many studies have shown that dispersion and interfacial interaction between graphene and polymer matrix are two key factors for achieving performance enhancement [26, 55, 61–63]. For one thing, the increase in tensile strength owes possibly to the tough between graphene and polar acetate groups in EVA [26, 64]. For another, graphene has a large aspect ratio and specific surface area and its contact geometry is a two-dimensional surface, which leads to better transfer of interfacial stress between graphene and EVA, directly improving the mechanical adhesion of the interface [24, 65, 66]. Moreover, the fluidity of the polymer chains is limited by the interaction with graphene, which enhances the interface toughness and forms a mechanically durable network inside the EVA, thereby improving the modulus and strength of the composite [67–69]. And the results of MSD and IBE also prove that graphene can improve the strength of EVA.

The trend in intensity is clearly observed in figure 8. The results show that the mechanical properties of EVA66Gr composites containing 9 wt% graphene are relatively better than those of the other two composites. This result is consistent with the strong interfacial interaction between graphene and EVA and the results for the improvement of the $T_g$ of EVA by graphene. The trend of tensile strength for EVA88Gr seems different with others, especially around 250 K. This is related to the result in figure 5(b) that the interface binding energy of EVA88Gr material is relatively affected by temperature, especially when it fluctuates greatly around 250 K. Moreover, the contact area of graphene and EVA in EVA88Gr is relatively large. The interface binding energy is
greatly affected by the temperature and is relatively low near 250 K, which may be the reason for its relatively weak strength.

The higher interfacial interaction limits the fluidity of the EVA chains, resulting in an increase in thermal stability, which is the main reason for the improvement of mechanical properties of graphene/EVA nanocomposites.

It is understood that there are some uncertainties in MD simulation. First, although the polymer molecules have reached a relaxation state through isothermal-isobaric ensemble simulation prior to operation, the average thermodynamic parameters of polymer systems in MD simulations cannot avoid the inherent uncertainty of the model that stem from initial conditions [70]. Second, in simulations of glass transition temperatures, simulated density is usually defined as a fixed mass divided by a dynamic average volume. As a result, they exhibit a limited size effect in the form of noise, especially at high temperatures [71]. This can have a slight interference with the results. Moreover, since long MD simulations will generate accumulated deviations in numerical integration
Therefore, the deviations caused by uncertainties in MD simulation can be reduced, but cannot be completely eliminated.

4. Conclusions

In this work, MD simulation was implemented to explore the influence of graphene on the thermal stability and mechanical properties of EVA. The results have proved that the glass transition temperature, modulus, and strength of the graphene/EVA nanocomposites are higher than that of pure EVA. The simulation results are in agreement with the experimental data of EVA and graphene/EVA nanocomposites.

The current MD results have shown that the increase of glass transition temperature of the composite is closely related to the size of graphene. However, with the addition of graphene, the mean-square displacement of the EVA chains is reduced, and the limitation of the flowability of EVA chains by different sizes of graphene is consistent with the improvement of the $T_g$ of EVA by graphene. The increase in $T_g$ and the decrease in mean-square displacement upon the addition of graphene are explained by the strong interaction between graphene and EVA chains, this interaction reduces the molecular mobility and the flexibility of the polymer chains nearby the graphene.

The results on the stress-strain of nanocomposites revealed that EVA66Gr nanocomposites with 9 wt% graphene have superior strength and toughness compared to others. The results show that the strong interfacial interaction between graphene and EVA limits the fluidity of EVA chains and improves the thermal stability of EVA, which may be the reason for the improvement of mechanical properties of graphene/EVA composites. Therefore, the addition of graphene is an effective way to significantly improve thermal stability and the mechanical properties of EVA. Furthermore, it indicates that the addition of graphene may also enhance other types of polymeric materials.

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