Molecule Mechanics Simulation on the Deformation and Damage Process in POSS Nanocomposite

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Abstract

The deformation and damage process of POSS nanocomposite is investigated by molecule mechanics (MM) simulation. Firstly, the nano-scale models of two kinds of homopolymers, pure polystyrene (PS) and polystyrene attached with 5 mol% propyl-POSS (P-POSS-PS) were built. Then the mechanical behaviors of these two kinds of hybrid materials under focused uniaxial tensile loading and the remote uniaxial tensile loading are examined by MM simulations. It is found that a small quantity of POSS can observably increase the tensile modulus of the normal polymers. During tensile loadings, micro voids appear in the polymer matrix. With the increase of deformation, the micro voids become bigger and then interconnect to form the damage in bigger area. The POSS monomers prevent these micro voids from coalescence and thus retarding the formation of the damage. This would be helpful in understanding the reinforcement mechanism of POSS and provide important referential message for further application.

1. Introduction

The POSS nanocomposite is the hybrid of polyhedral oligomeric silsesquioxane (POSS) monomers with polymer molecules. Here the POSS monomer plays the role as a nano-scaled reinforcement. Experiments show that the addition of POSS could improve significantly many properties, including the mechanical properties of polymers. On the other hand, it is desired to know the detailed toughening mechanism of POSS monomers on polymer matrix, so as to make further improvement on materials' molecule design.

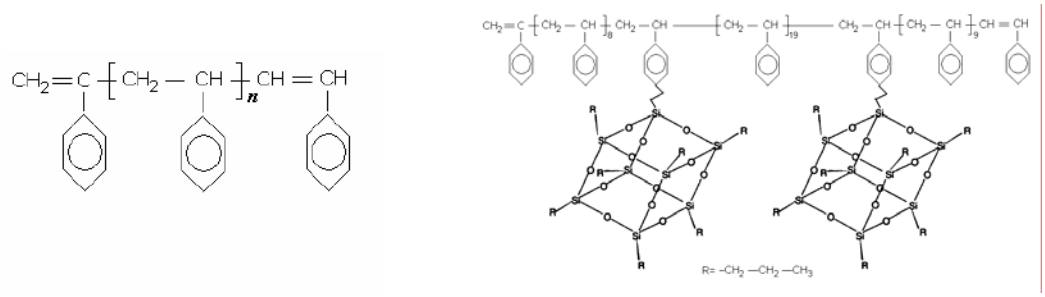
The POSS is depicted by a formula of $(RSiO_{1.5})_n$ or T_n (where n is an even number and R=H, Cl or a variety of organic groups) [1,2]. Comparing with other traditional polymer materials, the inimitable characteristic of organic-inorganic nano-hybrid materials POSS is that POSS monomers are combined to the polymer molecules. The POSS monomers lead to serials of excellent properties such as high thermal stability, high tenacity and high intensity and so on. Not only the POSS monomers can form polymers themselves, but also the monomers can be combined to the polymer chains and reinforce the polymer materials. The polystyrene (PS) is a common engineering plastics and be applied very widely.

The POSS monomers can be combined to the PS molecular chains through condensation reaction [3] to improve the mechanical properties of PS. In the present paper, the deformation and damage behaviors of two copolymers, the pure PS and the PS attached with 5 mol% propyl-POSS (P-POSS-PS, where n=8), will be studied by molecule mechanics simulation method. Attention is focused on the strengthening role of POSS in PS matrix.

2. Model of Simulation

Fig. 1 shows the molecule model of PS and POSS nanocomposite. The PS molecule is constructed by a chain formed from 50 styrene monomers. The molecule model of PS is built by including 12 PS molecules. This atomistic model contains 9 576 atoms and the sizes in the x, y and z directions are 6.09 nm, 4.06nm and 4.06nm, respectively. The P-POSS-PS molecule is constructed by a chain including 40 styrene monomers and 2 P-POSS monomers. The two P-POSS monomers are hanged on the tenth and the thirtieth styrene monomers of the chain respectively. The pole-like atomistic model of P-POSS-PS is built by including 12 P-POSS-PS molecules. This atomistic model contains 9 432 atoms and the sizes in the x, y and z directions are 5.73 nm, 3.82nm and 3.82nm, respectively. The atomistic model densities for the copolymers of PS and P-POSS-PS are selected as 1.05g/cm^3 and 1.25g/cm^3 respectively.

Two types of loading conditions are considered: focused and remote uniaxial tension. For focused uniaxial tension the tensile specimen is simply the block of the atomistic model for MM simulation, and the displacement loading is applied directly on the layer of atoms at the end of the specimen (Fig. 2). While for remote tension the molecule model situated at the center of a uniform tensile specimen (Fig. 3). The tensile models are built by connecting the carbon and silicon atoms on the boundaries of the atomistic models to the corresponding nodes in the finite element areas. The left side of the specimen is kept fixed and the right side is stretched step by step. The central part of the remote tensile model is the pole-like atomistic model of PS and P-POSS-PS.



(a) (b)

Fig. 1 Sketch of the molecule model. (a) PS molecule. (b) P-POSS-PS molecule

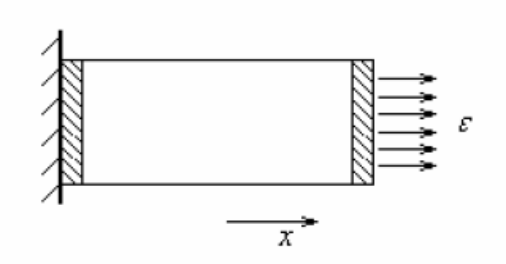


Fig. 2 Focused tension model

In the present paper, only the molecular static simulations have been performed. The MM3[4~6] force field is used. The cutoff radius of 0.8 nm and 1.5nm are selected to compute the Van der Waals forces and the electrostatic forces respectively. For remote tension the model is stretched by a tensile strain loading and the atomistic pole-like model is stretched synchronously by the boundary atoms bonded to the finite element nodes. The L-BFGS[7,8] algorithm is used to optimize the structures and the energies of the molecules and the atomistic models after deformation. There are 593 boundary atoms (Only the C and Si atoms are considered) on the six surfaces of the atomistic model of PS and 597 ones for P-POSS-PS. All the atoms on the right surfaces of the two atomistic models (272 atoms in PS and 320 atoms in P-POSS-PS) are called the tensile atoms and are used to compute the tensile forces of the atomistic models. A displacement of 9 nm at each step and a total of 11 steps are carried out on the right side of the tensile models during the simulations. After each tensile step the atomistic model are optimized. When the optimized structures have been obtained the resultant forces along the tensile direction of the tensile atoms at the right side of the atomistic models can be calculated.

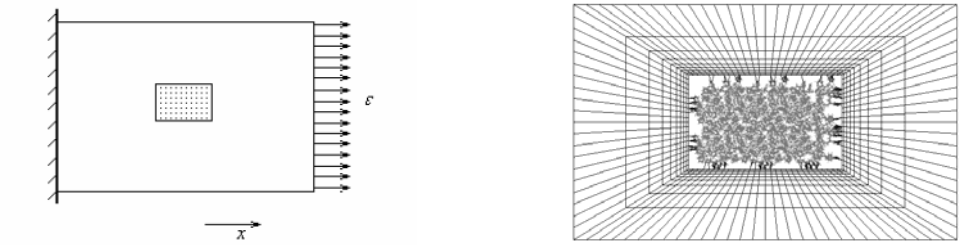


Fig. 3 Remote tension model

3. Stress-strain Relation

The tensile force vs. strain curves and the stress-strain curves of the two kinds of polymers are shown in Fig. 4 and 5. It can be found that the tensile forces increase with increasing strains approximately at the initial tensile stage. After the forces

reach to the maximum values there come the inelastic softening behavior. It is noticed that the inelastic deformation behavior is strongly dependent on the selection of the simulation model. The tensile elastic moduli can be calculated from the maximum stresses and the corresponding strain in Fig.4 and Fig.5. The tensile modulus of PS obtained from this work is 2.26 GPa for focused tension and 4.52 GPa for remote tension. The experimental result is just between these two values [9]. The tensile modulus of P-POSS-PS is 4.13 GPa for focused tension and 5.32 GPa for remote tension. In both cases the reinforcement effect is clear.

The strain at the maximum stress of P-POSS-PS is larger than that of PS. This shows that the P-POSS does not reduce the toughness of PS at the elastic range. Strictly speaking, the elastic zone has been enlarged slightly. In this paper the mole ratio of the P-POSS is only 0.05. If a higher ratio was selected, it can be predictable that the tensile modulus of P-POSS-PS will be more larger. But the influences on the toughness are in need of a further research in such case

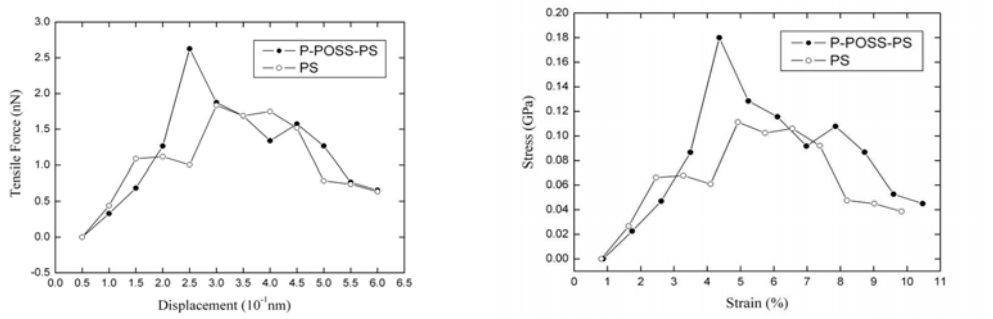


Fig. 4 The tensile force vs. strain of PS and P-POSS-PS under focused tension

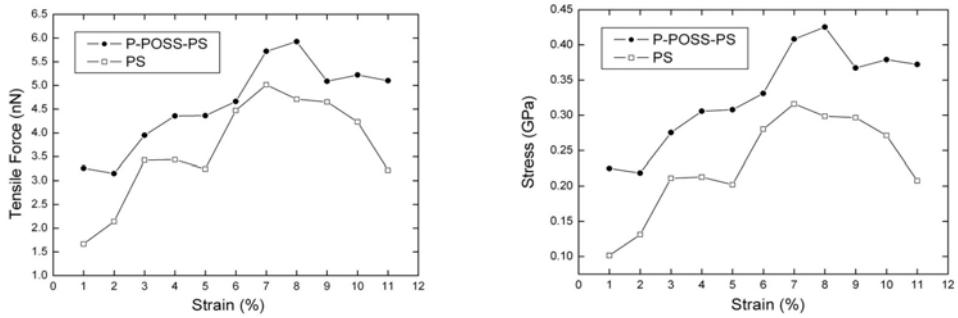


Fig. 5 The stress-strain curves of PS and P-POSS-PS under remote tension

4. Nucleation and Growth of Micro Voids

The structure deformations of the two kinds of polymers under remote tension with different tensile strains are shown in Fig.6 and 7. Here only the atomistic models and the surrounded finite element nodes are displayed. From Fig. 6 we can find that several micro voids appear at the right side when the strain reaches to 0.05. And the diameters of these voids are about 0.4 nm to 0.6 nm. After the micro avoids appear the tensile stress reaches to the maximum value at the strain of 0.07. The sizes of the voids increase with the increasing tensile strains. When the strain reaches to 0.11 the void diameters are about 0.7 nm to 0.9 nm. For P-POSS-PS, the phenomena during the tensile simulations are similar to those of PS. But the micro voids appear at the strain of 0.06. Compare with PS the micro voids in P-POSS-PS appear later and are fewer in number. Consider the factor that the strain at the maximum stress of P-POSS-PS (0.08) is larger than that of PS (0.07) in Fig.4. We can make a conclusion that not only the tensile intensity of polymers can be reinforced clearly but also the toughness can be enhanced slightly by a small mole ration of POSS. For focused tension the results are similar.

5. Discussion

POSS can improve the mechanical properties of polymers effectively. A small mole ration of POSS can make the tensile moduli of the homopolymers improve significantly and the toughness can be reinforced at the same time. Micro voids will appear during the tensile loading. The size of the voids increases with the increasing strain. Only the influences of the 5 mol% POSS on the tensile properties of polymers were studied here, how the higher mol ratios of POSS will influence the toughness of polymers need further research.

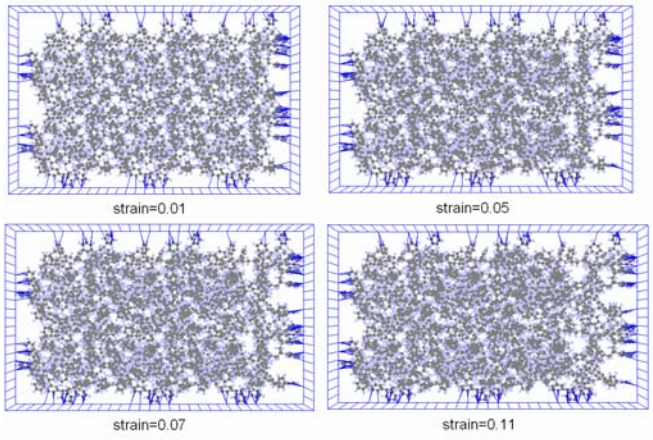


Fig. 6 Structures of PS under different tensile strains

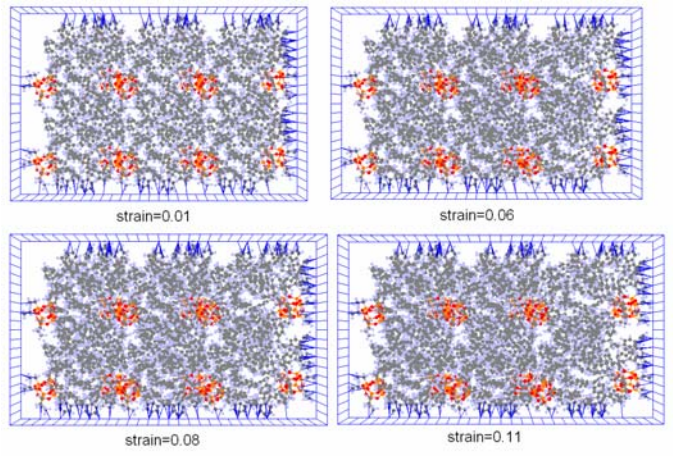


Fig. 7 Structures of P-POSS-PS under different tensile strains

Acknowledgements

The author would like to thank the National Natural Science Foundation (No. 10472028), the Excellent Youth Fund of Heilongjiang Province and the Boshidian Research Funds of the Ministry of Education of China for the financial support of this research.

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