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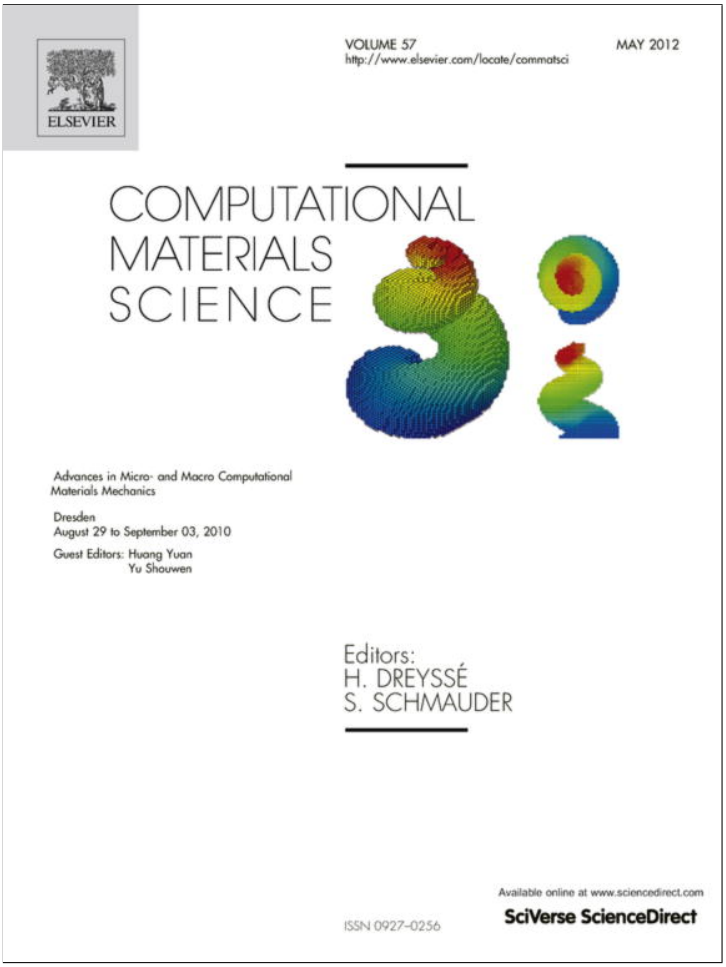

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A multi-scale approach of amorphous polymer from coarse grain to finite element

Jianjun Bian, Gangfeng Wang *

Department of Engineering Mechanics, SVL, Xi'an Jiaotong University, Xi'an 710049, China

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ABSTRACT

In the present paper, a multi-scale method from coarse grain to finite element is developed to simulate the mechanical behavior of amorphous polymer. In the coarse grain scale, the amorphous cell method is adopted to generate the configuration of two-dimensional random polymer. Based on the analysis of a main cell, the displacement mapping relation between finite element nodes and coarse grains is formulated, through which the handshaking region between coarse grains and finite elements is dealt with. The examinations of several typical deformation modes display the new developed multi-scale method can seamlessly transmit deformation across different length scales. As an example, the fracture of polymer is investigated by the multi-scale method, and compared with that simulated by fully molecular simulation. It is shown that the new developed multi-scale method provides a powerful tool to investigate various mechanical behaviors of amorphous polymer.

1. Introduction

The mechanical deformation and failure of solids are inherently interdependent multi-scale phenomena happening in various length scales and time scales, such as the breaking of chemical bond near interface at angstrom scale and the dissipation of strain energy by plasticity and heat at mesoscale. Most recently, many multi-scale computational methods ranging from electronics to continuum have been proposed to simulate the mechanics of crystalline materials. Among them, Quasi-continuum (QC) method [1] is the most fundamental and initiative one. In this method, the whole system has two types of atoms, "local representative atoms" corresponding to continuum finite element (FE) nodes, and "non-local representative atoms" similar to atoms in atomic region. And a handshaking region is employed to transit the deformation and loading between atomic region and continuum region. However, the mismatch between the local assumption in finite element method and the non-local interaction in atomic scale leads to ghost force [2]. Through a non-local continuum formulation near the atomistic/continuum interface, the finite element and atomistic model method (FEA) [3] can correct the spurious interfacial forces to some extent. Recently, Knap and Ortiz advanced a fully non-local QC method [4], in which some atomic positions are constrained to node positions, and forces are calculated from the fully non-local description. To list only few of them, recent progresses on atomistic/continuum method can be referred to the review paper given by Curtin and Miller [5].

Polymer is one important kind of engineering materials, and its mechanical properties have been extensively investigated at different length scales. For examples, at macroscopic continuum scale, Boyce et al. [6], and Wu and Van der Giessen [7] developed an elastic–viscoplastic model for amorphous polymer. At atomic scale, molecular dynamic method has been employed to simulate the mechanical performance of polymer [8]. To reduce computational burdens, a coarse grain (CG) model [9] has also been developed to investigate fracture and craze of polymer [10–12]. Bouvard et al. outlined the hierarchical modeling of polymer from atomic scale to macroscopic scale [13]. However, due to the complex random structure of polymer and the long-range transition of loading along chains, there is yet an absent of multi-scale method for amorphous polymer.

Attention to the coarse grain model (CGM) [9], not only reducing the freedoms in atomic simulation but also adopting identical segments, we propose a multi-scale method of polymer linking from coarse grain scale to continuum scale in the present paper. The basic descriptions of coarse grain method and finite element method are briefly reviewed in Section 2. In Section 3, the displacement mapping relation between finite element nodes and coarse grains is formulated. The methodology to deal with the handshaking region between coarse grain region and finite element region is presented in Section 4. As an example, the fracture of polymer is simulated by the multi-scale method, and compared with that modeled by fully molecular simulation in Section 5.

2. Coarse grain model and finite element model

2.1. Interaction between coarse grains

In the coarse grain method, each individual chain in polymer is modeled as a sequence of spherical "segments" (coarse grains),

* Corresponding author. Tel.: +86 29 8266 8434.
E-mail address: wanggf@mail.xjtu.edu.cn (G. Wang).

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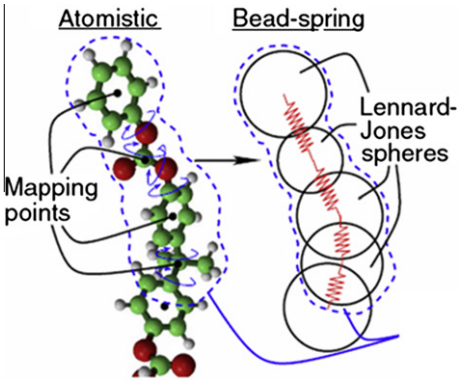

Fig. 1. Schematic of coarse grain method (Ref. [9]).

representing an ensemble of atoms as shown in Fig. 1. The description of interaction between coarse grains is of critical importance, since it is related to whether the coarse grain chains can represent the realistic atomic structure of polymer. Recently, a detailed review paper is addressed on how to link atomic simulation to coarse grain model [14].

Here we use the model proposed by Kremer and Grest [9]. All coarse grains interact with a repulsive Lennard–Jones potential,

$$E_{ij}^{LJ} = \begin{cases} 4\varepsilon\left[(\sigma/r_{ij})^{12} - (\sigma/r_{ij})^{6}\right] & \text{for } r_{ij} \leqslant r_c \\ 0 & \text{for } r_{ij} > r_c \end{cases}. \tag{1}$$

where ε and σ are the characteristic energy and length scales, respectively.

For adjacent coarse grains along the sequence of chain, an attractive finite extensible nonlinear elastic potential is applied,

$$E_{ij}^{CH} = \begin{cases} -0.5kR_0^2 \ln[1 - (r_{ij}/R_0)^2] & \text{for } r_{ij} \leqslant R_0 \\ \infty & \text{for } r_{ij} > R_0 \end{cases}. \tag{2}$$

The cutoff radius of Lennard–Jones potential is $r_c = 2.5\sigma$, and the parameters of the bond attractive potential are $R_0 = 1.5\sigma$ and $k = 30\varepsilon/\sigma^2$.

The force on each coarse grain can be obtained by the negative gradient of interaction potential with respect to the coarse grain position. This model has been widely used to investigate the mechanical properties of polymer [10,11].

2.2. Generation of polymer at coarse grain scale

A realistic polymer structure is necessary for all the later analysis. In the present paper, we adopt the amorphous cell method [15] to generate the configuration of two-dimensional random polymer. According to this method, only a square-shape main cell is needed to be considered, which includes the same number of coarse grains as one chain has. And each coarse grain along a chain can find its mirror part in this cell by translation of an integer times of the main cell's length.

The initial positions of coarse grains are generated randomly in the main cell, and then the conjugate method is adopted to obtain its equilibrium configuration. For a polymer chain with fifty coarse grains, Fig. 2 shows the equilibrium configuration of coarse grains in the main cell. The coarse grains along the same chain are connected by lines. Based on the equilibrium configuration of main cell, then the amorphous cell method is used to produce a polymer sample at coarse grain scale as shown in Fig. 3. By using the main cell, the macroscopic properties of polymer corresponding to this coarse grain structure can be determined, which will be adopted in the finite element region of the multi-scale method.

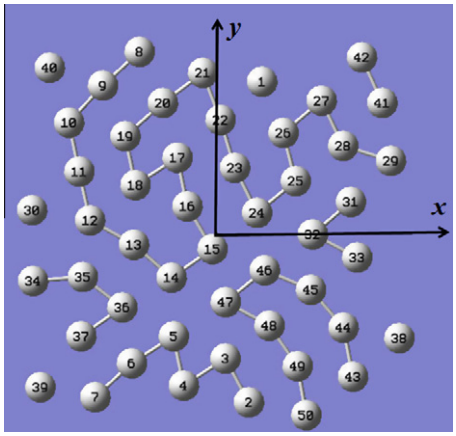

Fig. 2. Equilibrium configuration of coarse grains in the main cell.

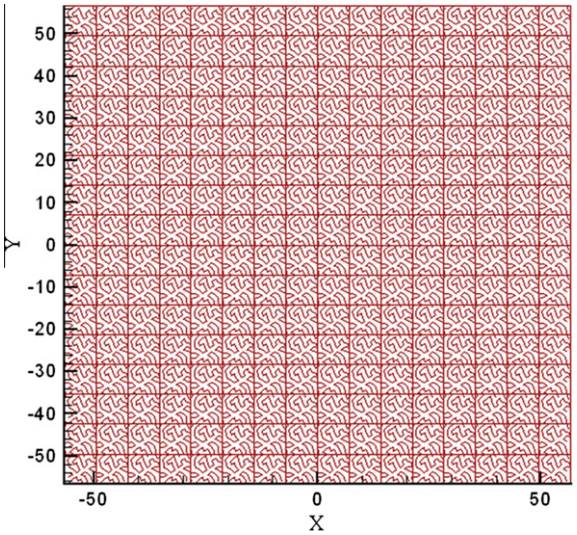

Fig. 3. Polymer at the coarse grain scale.

2.3. Finite element method

Except for some critical regions where are required to be investigated by molecular simulation, the finite element method is used to describe the far-field region, where the deformation is not severe so that the linear elasticity theory is accurate enough to describe its mechanical behavior.

In the classical finite element method, the strain energy V^e of an element can be expressed as

$$V^e = \frac{1}{2}\{u\}^{e^T}[K]^e\{u\}^e, \tag{3}$$

where $\{u\}^e$ is the node displacement matrix, and $[K]^e$ is the element stiffness matrix. Therefore, the forces on the nodes of an element induced by the deformation of this element are

$$\{f\}^m = [K]^m\{u\}^m, \tag{4}$$

where m is the index of element, $\{f\}^m$ are the node forces, $[K]^m$ is the element stiffness matrix and $\{u\}^m$ are the element node displacements. The resultant force on a node is the sum of all the contribution from its adjacent elements who share this node.

Different from the traditional finite element procedure, Broughton et al. [16] adopted a new algorithm in the finite element

region. During the loading process, each node is displaced along the direction of resultant node force. Only when the force on each node is smaller than a very small specified threshold value (i.e. 10^{-12}), it is assumed that the equilibrium system state is obtained, which possesses the minimum potential energy. The conjugate method can also be used to seek the equilibrium state of finite element region.

3. Mapping relation between FE and CGM

In the amorphous cell method, the main cell is the basic block to construct coarse grain region. While in continuum mechanics, the size of finite elements must be big enough to show continuous macroscopic properties. Therefore, it is natural and convenient to treat the main cell as the minimum size of finite elements. In macro-scale, the displacements at any point in an element can be determined by the interpolation of node displacements. However, in micro-scale, the displacements of discrete coarse grains do not follow the same way. When the main cell is treated as a finite element, it is necessary to find the displacement mapping relation between FE nodes and coarse grains for specified deformations by direct molecular simulations, which will be used to deal with the handshaking region between finite element region and coarse grain region.

Consider the main cell as a four-node element as shown in Fig. 4. According the finite element method, the node displacements can be expressed by its coordinates in terms of a_i and b_i,

$$u_i = a_1 + a_2 x_i + a_3 y_i + a_4 x_i y_i, \tag{5}$$

$$v_i = b_1 + b_2 x_i + b_3 y_i + b_4 x_i y_i, \quad (i = 1, 2, 3, 4) \tag{6}$$

where u_i, v_i are the displacements of the i-node, and x_i, y_i the coordinates of nodes. Then a_i and b_i are related to the node displacements by

$$\begin{Bmatrix} a_1 \\ b_1 \\ a_2 \\ b_2 \\ a_3 \\ b_3 \\ a_4 \\ b_4 \end{Bmatrix} = \frac{1}{4} \begin{bmatrix} 1 & 0 & 1 & 0 & 1 & 0 & 1 & 0 \\ 0 & 1 & 0 & 1 & 0 & 1 & 0 & 1 \\ -1 & 0 & 1 & 0 & 1 & 0 & -1 & 0 \\ 0 & -1 & 0 & 1 & 0 & 1 & 0 & -1 \\ -1 & 0 & -1 & 0 & 1 & 0 & 1 & 0 \\ 0 & -1 & 0 & -1 & 0 & 1 & 0 & 1 \\ 1 & 0 & -1 & 0 & 1 & 0 & -1 & 0 \\ 0 & 1 & 0 & -1 & 0 & 1 & 0 & -1 \end{bmatrix} \begin{Bmatrix} u_1 \\ v_1 \\ u_2 \\ v_2 \\ u_3 \\ v_3 \\ u_4 \\ v_4 \end{Bmatrix}. \tag{7}$$

Instead of using the node displacements, we adopt a_i and b_i to characterize the deformation of main cell, not only for their clear physical indications, but also for the convenience to manipulate the deformation of main cell. Among them, a_1 and b_1 correspond to rigid displacements, a_2 and b_3 correspond to uniform stretching, a_3 and b_2 correspond to uniform shear, and a_4 and b_4 correspond to hourglass modes, in the x-axis and the y-axis, respectively.

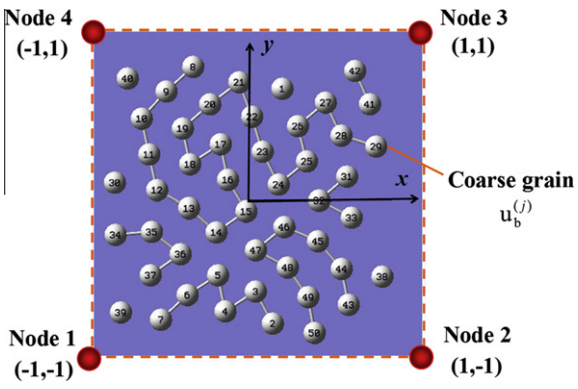

Fig. 4. Geometry of a four-node element.

For each deformation mode corresponding to a_i or b_i, molecular simulation is employed to determine the displacements of coarse grains in the main cell. Denote the displacements of the j th coarse grain by $u_{a_i}^{(j)}$ or $u_{b_i}^{(j)}$ ($j = 1 \ldots k$), and k is the number of coarse grains in the main cell. Since the interaction between coarse grains is non-local, to calculate the forces on coarse grains in the main cell, it is necessary to know the positions of coarse grains in the surrounding cells, as shown in Fig. 5a. It is assumed that the nearest neighboring cells take the same form of deformation as that of the main cell described by Eqs. (5) and (6). Based on the initial equilibrium configuration of main cell, through changing the size of main cell or the coordinate system, one can apply different deformation modes specified by a_i or b_i. Then the conjugate method is used to obtain the corresponding equilibrium configuration after deformation.

For a uniform stretching along the x-axis specified by a_2, Fig. 5b shows the equilibrium configuration before (gray color) and after (red[1] color) deformation. The displacements of coarse grains are given in Fig. 6. It is not surprising to find that coarse grains also displace along the y-axis, though the loading is along the x-axis. In addition, for small deformation, the displacements of coarse grains are linearly proportional to the applied strain. The corresponding results for uniform shear and hourglass deformation along the x-axis are displayed in Figs. 7 and 8, respectively. The mapping relation for deformation related to b_i can be obtained similarly.

Based on the above obtained results of $u_{a_i}^{(j)}$ and $u_{b_i}^{(j)}$, then for any deformation of a finite element described by its node displacements u_i and v_i, we can determine the displacements ($u_b^{(j)}$ and $v_b^{(j)}$) of each coarse grain in it,

$$\begin{Bmatrix} u_b^{(j)} \\ v_b^{(j)} \end{Bmatrix} = \begin{bmatrix} 1 & 0 & u_{a_2}^{(j)} & u_{b_2}^{(j)} & u_{a_3}^{(j)} & u_{b_3}^{(j)} & u_{a_4}^{(j)} & u_{b_4}^{(j)} \\ 0 & 1 & v_{a_2}^{(j)} & v_{b_2}^{(j)} & v_{a_3}^{(j)} & v_{b_3}^{(j)} & v_{a_4}^{(j)} & v_{b_4}^{(j)} \end{bmatrix} \begin{Bmatrix} a_1 \\ b_1 \\ a_2 \\ b_2 \\ a_3 \\ b_3 \\ a_4 \\ b_4 \end{Bmatrix}. \tag{8}$$

According to Eqs. (7) and (8), the mapping relation can be described simply as

$$\mathbf{u}_b^{(j)} = N^{(j)} \mathbf{u}_n, \tag{9}$$

where $\mathbf{u}_n$ are the displacements of four nodes, $N^{(j)}$ is the mapping matrix.

Reversely, if the displacements $\mathbf{u}_b^{(j)}$ of all coarse grains in a finite element are known, then what are the node displacements $\mathbf{u}_n$? It is nature to assume that if we know the node displacements $\bar{\mathbf{u}}_n$, we can obtain the coarse grain displacements following the same relationship as Eq. (9), that is

$$\bar{\mathbf{u}}_b^{(j)} = N^{(j)} \bar{\mathbf{u}}_n. \tag{10}$$

To determine the node displacements, we require the following orientation function is minimized by $\bar{\mathbf{u}}_n$

$$\lambda = \sum_{j=1}^{m} (\bar{\mathbf{u}}_b^{(j)} - \mathbf{u}_b^{(j)}) \bullet (\bar{\mathbf{u}}_b^{(j)} - \mathbf{u}_b^{(j)}), \tag{11}$$

which gives

[1] For interpretation of color in Figs. 1–5, 7–11, the reader is referred to the web version of this article.

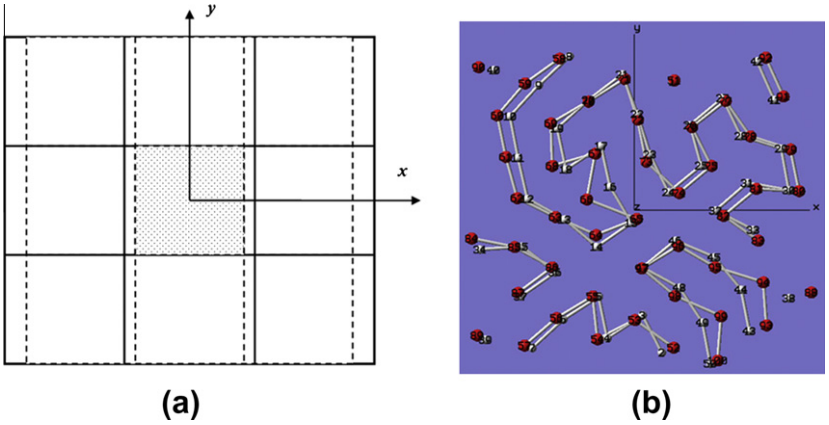

Fig. 5. Deformation of uniform stretching along the *x*-axis.

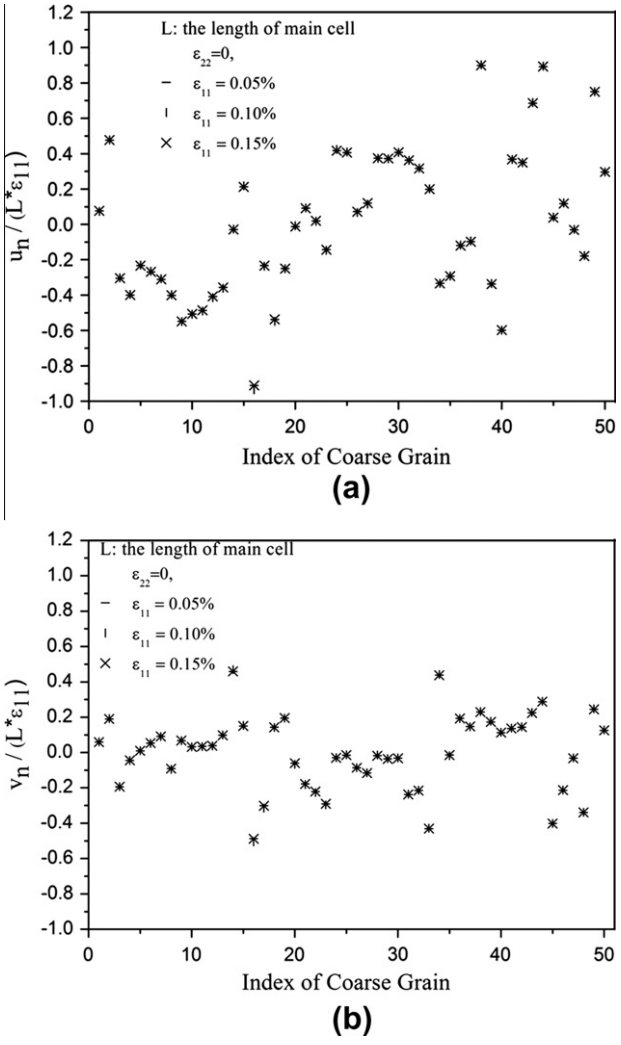

Fig. 6. Displacements of coarse grains along the *x*-axis (a) and the *y*-axis (b).

$$\sum_{j=1}^{m} N^{(j)T}\left(N^{(j)}\bar{\mathbf{u}}_n - \mathbf{u}_b^{(j)}\right) = 0. \tag{12}$$

And the node displacements are approximated by $\bar{\mathbf{u}}_n$. Our numerical examination verified that this method can be used to determine the node displacements, once the displacements of coarse grains in it are known.

4. A multi-scale method for amorphous polymer

With the above preparations, now we develop the multi-scale method for amorphous polymer. In the multi-scale simulation, polymer system consists of coarse grain region, finite element region and handshaking region between them. The coarse grain region and finite element region are dealt with by the methods mentioned in Section 2. The key in the multi-scale method is how to deal with the handshaking region properly, so that the physical and mechanical quantities are transferred smoothly and seamlessly across regions at different scales.

4.1. Handshaking region between CG and FE

Based on the displacement mapping relation in Section 3, we will deal with the handshaking region (as shown in Fig. 9) by the following scheme,

4.1.1. Interaction from FE to CG

The shape and size of the first layer finite elements outside the coarse grain region must be the same as those of main cell, and the boundary coarse grains (as shown in yellow color) are constructed in these elements. The displacements of boundary coarse grains are determined by the mapping relation Eq. (9). Thus the deformation in finite element region can be transferred to the coarse grain region by using the boundary coarse grains.

4.1.2. Interaction from CG to FE

On the other hand, virtual finite elements (nodes in blue and green color) including the same coarse grains as the main cell, are constructed in the coarse grain region adjacent to the finite element region. And the displacements of virtual elements are determined by the reverse mapping Eq. (12). Thereby, the deformation in coarse grain region can be fed back to the finite element region by the boundary nodes (in green color).

4.2. Examination

For uniform stretching and shear, as well as hourglass deformation applied in the remote finite element region, the above handshaking method has been examined, which shows a smooth and seamless transmission of deformation across regions at two length scales. Also, the multi-scale method supplies a great reduction of degree of freedom (DOF), which is related to the number of coarse grains in the main cell. The more the coarse grains in an individual polymer chain, the more efficient the multi-scale method.

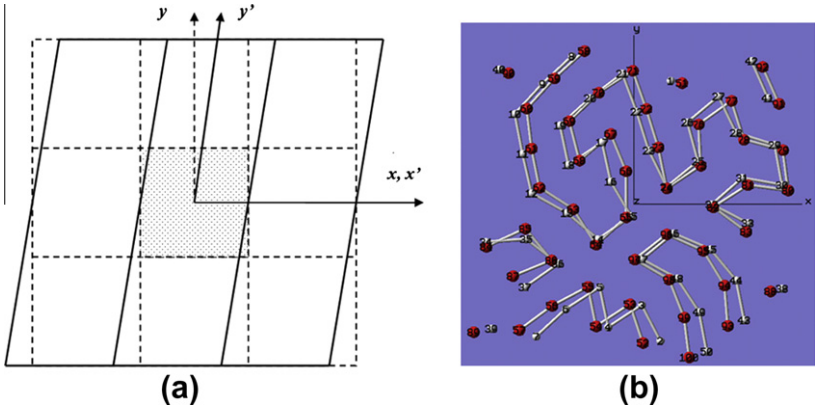

Fig. 7. Deformation of uniform shear along the *x*-axis.

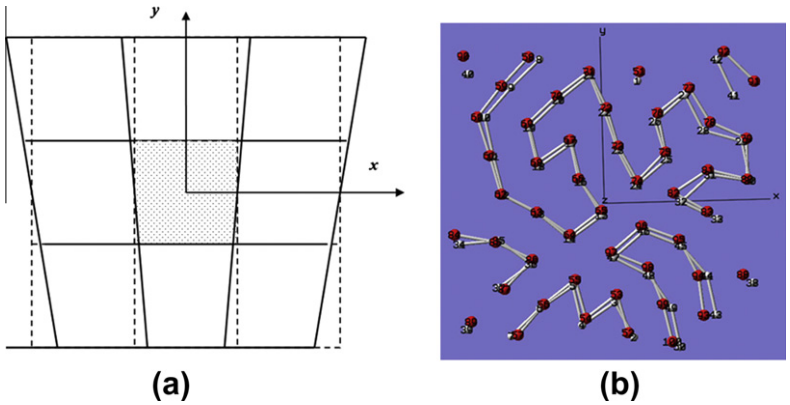

Fig. 8. Deformation of non-uniform stretch along the *x*-axis.

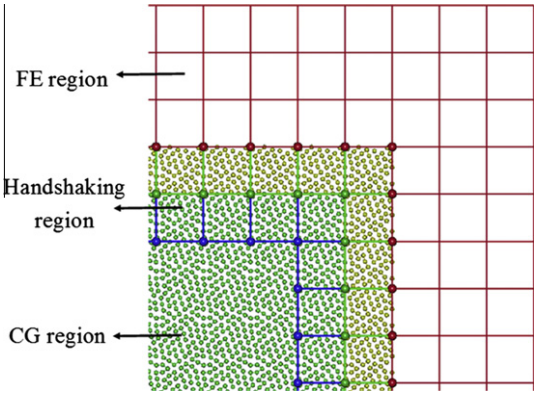

Fig. 9. Handshaking region between FEs and CGs.

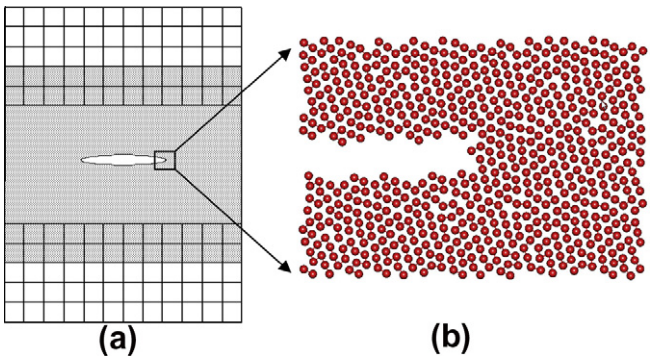

Fig. 10. (a) Multi-scale simulation of polymer fracture, (b) the initial coarse grain configuration near crack tip.

5. Results and discussions

By using the new developed multi-scale method, the fracture of polymer is investigated as shown in Fig. 10a. The coarse grains are used around crack to precisely abstract the deformation characteristics. Fig. 10b displays the configuration of coarse grains around crack tip. The finite element method is employed in the remote field to reduce the computational burden. The handshaking region as mentioned in Section 4 is constructed between CG region and FE region to transmit physical information between them. To investigate the process of deformation, the conjugate method is used to minimize the total potential of the whole system. For uniform stretching in the remote filed, two representative configurations of crack propagation are displayed in Fig. 11a and b with increasing strain. It is found that, for amorphous polymer with random chain structure, the propagation of crack depends strongly on the local micro-structures around crack tip, which is missed in the conventional macroscopic analysis.

To test the accuracy of the present multi-scale method, the same problem is also investigated by fully molecular simulation. For the same level of applied strains, the corresponding configurations of coarse grains near crack tip are shown in Fig. 11c and d. It is found that the configurations of coarse grains obtained by the multi-scale method are almost identical to those simulated by

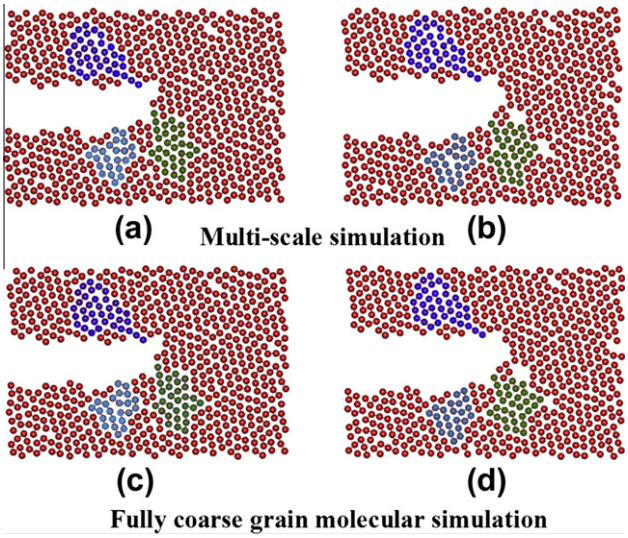

Fig. 11. Deformation around crack tip for two different loadings: (a) and (b) simulated by the multi-scale method, (c) and (d) modeled by the fully coarse grain method.

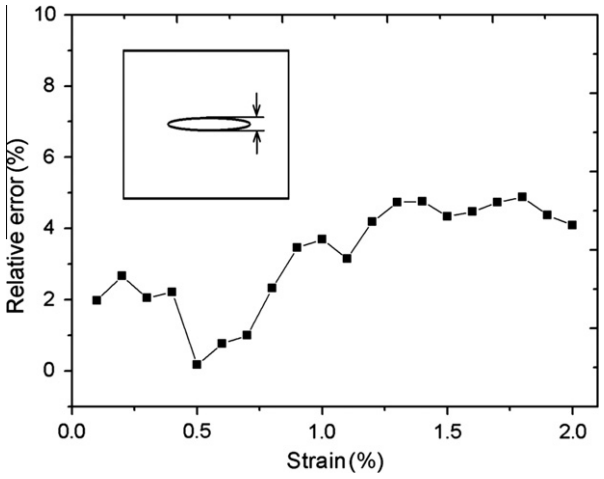

Fig. 12. The relative error of crack opening displacement vs. the applied strain.

the fully molecular simulation. We also check the opening displacement at the middle of crack surface. The relative error of crack opening displacement obtained by the two methods is plotted in Fig. 12, which demonstrates again that the multi-scale method is accurate enough. Therefore, the new developed multi-scale method can achieve almost the same accuracy as that modeled by fully molecular simulation but with less computation cost.

6. Conclusions

In the present paper, through the displacement mapping relation between finite element nodes and coarse grains in the main cell, a multi-scale method is proposed for amorphous polymer. Through tests on several typical deformation modes, the present method shows smooth and seamless transmission of deformation across finite element region and coarse grain region. Then the multi-scale method is used to investigate the fracture of polymer, which can reveal some micro mechanism in polymer. The mapping concept in the present method can also be extended to develop some other multi-scale methods.

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