Modeling Fracture of Graphene: A Molecular Dynamics Tutorial

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Abstract

This article describes molecular dynamics simulations of fracture of graphene, which can serve as a tutorial for new researchers in nanomechanics. Graphene was selected for this tutorial by considering its applications in numerous engineering disciplines and the convenience of modeling. All required input files to run the simulations are freely available to the readers.

1. Introduction

In many cases, structural failure initiates at the nanoscale. For example, a crack initiates by breaking individual atomic bonds. Figure 1 depicts the multiscale nature of a typical engineering structure. With the available computational tools, we can start our engineering design at the nanoscale to develop more economical and robust structures.

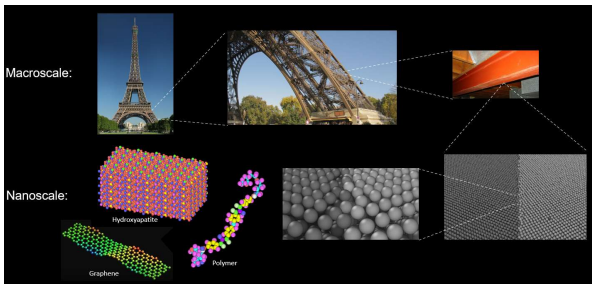

Figure 1: Multiacale nature of an engineering structure. The pictures of the Eiffle Tower were from Wikipedia.

As recently suggested by Munmulla and Weerasekera [1], the field of civil engineering has not greatly benefitted from emerging technologies such as nanotechnology. Their study revealed that 0.03% of graphene-oxide (by weight) could significantly improve the compressive strength of cement. Moreover, they showed that the percentage of oxygen in graphene-oxide has a noticeable influence on the compressive strength of cement. In addition to the oxygen content, structural defects (e.g., cracks and holes) in graphene can influence the performance of graphene itself as well as its composites. In this tutorial, we will use molecular dynamics (MD) simulations to study the effects of missing atoms (in the form of a crack or holes) on the strength of a graphene sheet.

This tutorial aims to guide new researchers by providing them with an overview of conducting proper MD simulations of the fracture of graphene. The required input files to run the MD simulations presented in this tutorial and the codes to post-process MD outputs are available to the readers on the GitHub repository: https://github.com/nuwan-d/fracture_of_grahene. The next section describes the graphene model used for the simulations and some basic simulation parameters and techniques. Results of the MD simulations are presented in the third section.

2. MD Simulation Setup

The uniaxial tensile tests of graphene were simulated using the LAMMPS MD simulator [2]. The interatomic interactions were described by AIREBO potential [3], which contains three sub-potentials: the Lennard-Jones (LJ), torsional, and reactive empirical bond order (REBO) potentials. The LJ and torsional potentials evaluate the energy due to the van der Waals and torsional interactions between atoms, respectively. The REBO potential expresses energy stored in a bond between atom i and atom j as

$$E_{ij}^{REBO} = f(r_{ij})[V_{ij}^{R} - b_{ij}V_{ij}^{A}] \qquad (1)$$

where V_{ij}^{R} and V_{ij}^{A} are the repulsive and attractive potentials, respectively; b_{ij} is the bond order term, which modifies the attractive potential depending on the local bonding environment; r_{ij} is the distance between the atoms i and j, and $f(r_{ij})$ is the cut-off function, which limits the interatomic interactions to the nearest neighbors. In order to avoid the non-physical strain hardening of the stress-strain curves, the cut-off distance of the REBO potential was modified to be 2 Å [4].

The graphene sample used for the simulation is shown in Fig. 2, which contains a central crack (i.e., a row of missing hexagons) and four interacting holes. The crack length is ~1.8 nm. The planar dimensions of the sample are 15 nm × 15 nm. Even though smaller samples (e.g., 5 nm × 5nm) can be used to compute the mechanical properties of pristine graphene, relatively larger samples are required to model defective graphene due to the significant size effects at the atomic scale. For a graphene sample containing a crack, the length and width of the sample must be kept at more than ten times the half initial crack length to avoid the effects of finite dimensions on the simulation results [5]. The MATLAB code "input_files.m" in the GitHub repository generates the required two LAMMPS input files ("grap-in.in" and "grap-data.data") for the simulation. The file named "CH.airebo" is the AIREBO potential file with modified cut-off distances.

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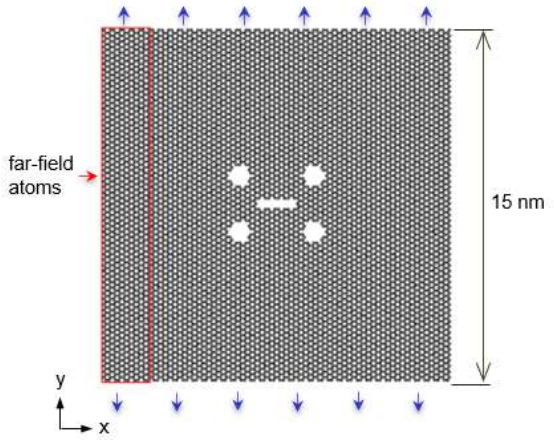

Figure 2: The graphene sample used for simulations, which contains a central crack and four interacting holes. During the simulation, the average stress of the atoms in the region marked as far-field atoms were also computed.

Before simulating the uniaxial tensile tests, the graphene sample was equilibrated for 25 ps; the selected time step being 0.5 fs. Convergence of total energy was used to identify the equilibrium state of the graphene sample. Initial displacement perturbations (~0.001 nm) were imposed on the atoms along the x-, y-, and z-directions to facilitate the equilibration [6]. Periodic boundary conditions were imposed along the x- and y-directions. The simulations were performed with the isothermal–isobaric (NPT) ensemble, and the Nośe-Hoover thermostat was used to keep the temperature at 300 K during the simulation. After the equilibrium period, the graphene samples were subjected to strain along the y-direction (ε_{yy}), at a rate of 0.001 ps^{-1}, until fracture.

The virial theorem was used for the calculation of atomic stress [7]. The averaged virial stress tensor, σ_{ij}, is defined as follows:

$$\sigma_{ij} = \frac{1}{V}\sum_{\alpha}\left[\frac{1}{2}\sum_{\beta=1}^{N}\left(R_i^{\beta} - R_i^{\alpha}\right)F_j^{\alpha\beta} - m^{\alpha}v_i^{\alpha}v_j^{\alpha}\right] \qquad (2)$$

where i and j are the directional indices (i.e., x, y, and z); α is a number assigned to an atom, and β is a number assigned to neighboring atoms of α; R_i^{α} is the position of atom β along the direction i; $F_j^{\alpha\beta}$ is the force on atom α due to atom β along the direction j; m_{α} and v_{α} are the mass and the velocity of atom α respectively; V is the total volume. In volume calculations, the thickness of graphene was assumed to be 3.4 Å [8]. The fracture of the sample was identified by a sudden drop in its stress.

3. Analysis

3.1 Stress Distribution

Figure 3 shows the stress field of two graphene samples when they are subjected to a strain of 0.02. In the case of an isolated crack (Fig. 3a), the stress concentration near the crack tip is significantly high. The presence of holes near the crack tip reduces the crack tip stress field, but a significant stress concentration can be observed at the edges of the four holes (Fig. 3b). We have previously demonstrated that the crack-hole interaction can significantly improve the fracture stress of graphene [9]. The MATLAB code "stress_distribution.m" available in the GitHub repository can be used to plot the stress field obtained from the MD simulations.

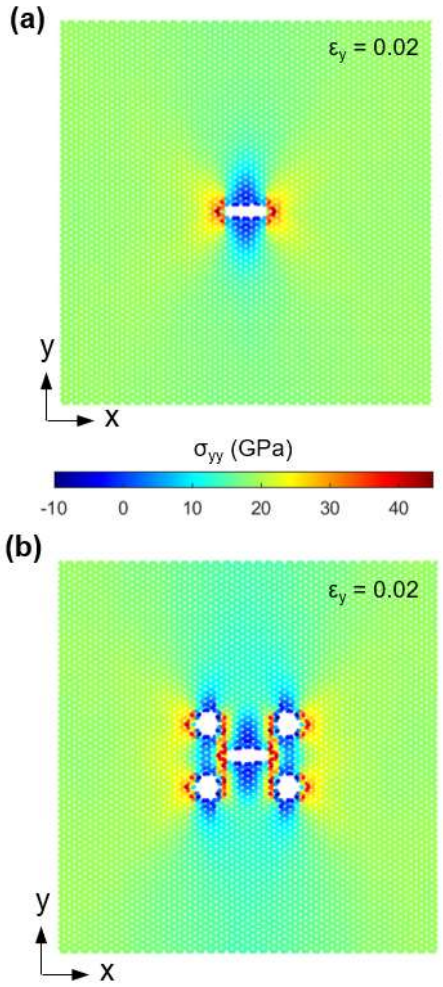

Figure 3: Stress distribution of graphene samples containing (a) a central crack and (b) a central crack and four interacting holes.

3.2 Stress-Strain Curve

Figure 4 shows the stress-strain curves of three graphene samples. Fracture of the pristine sample occurs when the average stress of the far-field atoms is 127 GPa, and the corresponding strain is 0.2. The presence of a central crack with a length of ~1.8 nm decreases the fracture stress down to 58.9 GPa, and the fracture strain down to 0.07. The presence of interacting holes along with the crack further reduces the fracture stress to 50.4 GPa and the fracture strain to 0.059. Please see our

previous publications for more information about the crack-hole interactions in graphene [9–11].

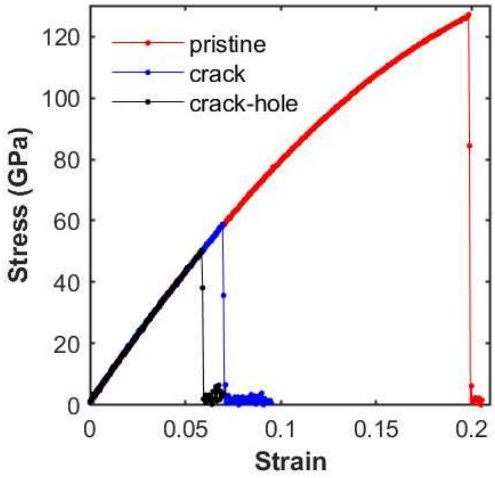

Figure 4: Stress-strain curve of three graphene samples.

3.3 Fracture Toughness

We can compute the mode-I stress intensity factor (K_I) at the onset of crack propagation in the graphene sample containing an isolated crack (see Fig. 3a) as follows:

$$K_I = \sigma_f \sqrt{\pi a} \qquad (3)$$

where a is the half initial crack length, and σ_f is the average far-field stress at the onset of crack propagation.

Figure 5 compares the change in average stress of the graphene sample and the far-field stress. The MATLAB code "stress_strain_curves.m" in the GitHub repository can be used to plot the stress-strain data extracted from the MD simulation.

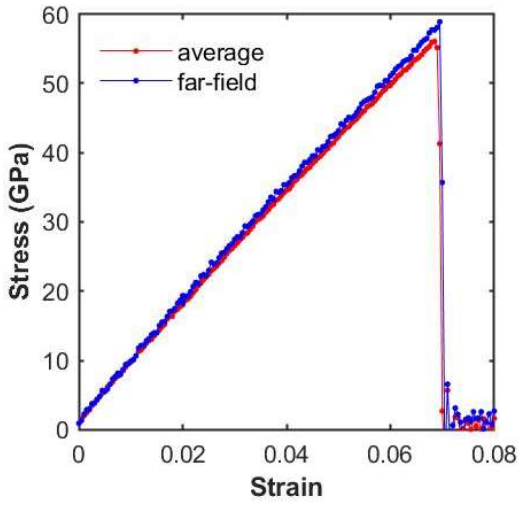

Figure 5: Changes in the average stress of the sheet and the far-field stress.

It can be noticed in Fig. 5 that the average stress begins to drop slightly before the drop of the far-field stress, which indicates the beginning of the crack propagation. The value of far-field stress at the initiation of crack propagation is 57.7 GPa. The computed value for K_I is 3.06 MPa√m, which is significantly below the experimental value (i.e., ~4 MPa√m) [12]. It should be noted that K_I demonstrates a significant size dependence when the crack length is only a few nanometers [10].

Supporting Information

Please see this GitHub repository for LAMMPS input files and MATLAB codes to generate input files and post-process the simulation outputs: https://github.com/nuwan-d/fracture_of_grahene

Acknowledgment

The author would like to thank Professor Nimal Rajapakse for his support during this work.

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