

MECHANICAL BEHAVIOR OF GRAPHENE USING ATOMIC-SCALE FINITE ELEMENT METHOD AND MOLECULAR DYNAMICS

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Summary We show that atomic-scale finite element method (AFEM) is computationally efficient and as accurate as molecular dynamics by comparing the force-strain curve for a graphene sheet. We also demonstrate that AFEM is able to capture the effect of free edges on the equilibrium configuration of graphene.

Graphene has attracted significant attention of the scientific community due to its unique electronic and mechanical properties [1, 2]. Researchers have used a number of computationally efficient continuum models to explain the behavior of nanostructures. However, graphene and carbon nanotubes are essentially discrete structures, and continuum models cannot account for such discreteness. In this paper, we demonstrate that the atomic-scale finite element method (AFEM), originally proposed in [3], is a computationally efficient and accurate method that can be used to model graphene structures.

AFEM is an atom based finite element method (FEM), where the stiffness of the atoms is derived from a molecular mechanics force field. AFEM is much more superior compared to the most commonly used bond based FEM for nanostructures [4]. Fig. 1(a) shows the two basic AFEM elements, which we used in this study. The element type 2 can be obtained by rotating the element type 1 by $\pi/3$ rad around central atom (No. 1) in clock-wise direction. The central atom of each element interacts with three nearest neighboring atoms (No. 2 - No. 4) and six second nearest neighbors (No. 5 - No. 10).

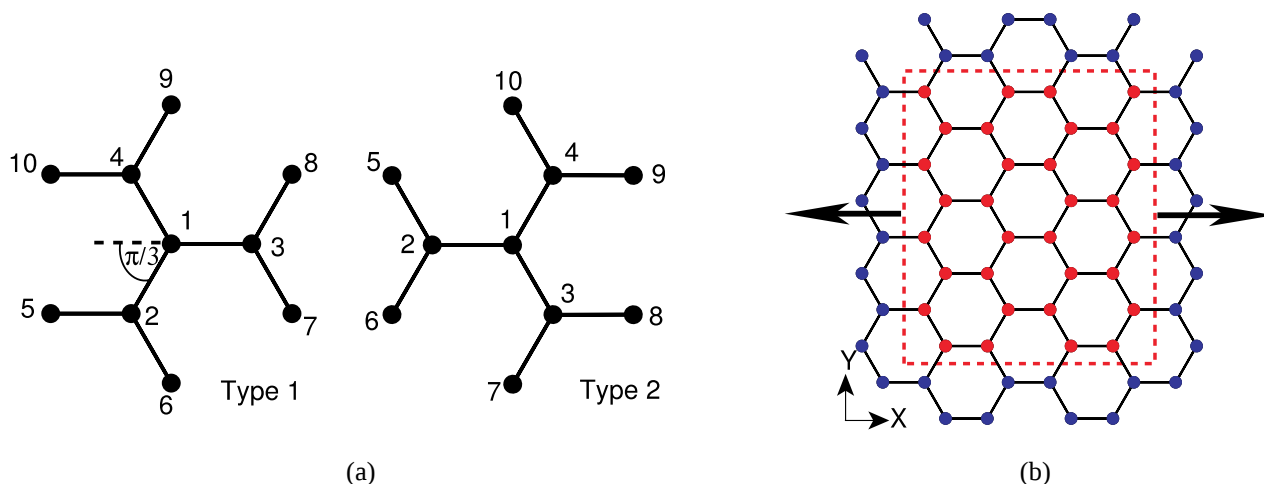


Figure 1: (a) Basic AFEM elements used for the study, (b) The graphene sheet used to calculate the strain-force relation from AFEM and molecular dynamics (MD).

The total energy (E_{tot}) of the central atom is obtained from reactive bond order (REBO) potential [5]. Taylor series expansion of E_{tot} around an initial configuration (need not be stable) gives the non-equilibrium force vector ($\mathbf{P}_{\text{ele}}$) and the element stiffness matrix ($\mathbf{K}_{\text{ele}}$) [3].

We compare the average force on a carbon atom of the graphene sheet shown in Fig. 1(b), calculated at different strains using AFEM and molecular dynamics (MD). The MD simulation is done using LAMMPS [6] with REBO potential at a temperature of 1 K. Periodic boundary conditions (PBC) are used in both x and y directions for the system inside the dashed box (in red) to eliminate edge effects. Isothermal isobaric (NPT) time integration scheme is used with a time step of 0.5 fs and a strain rate of 0.001 ps⁻¹. The cut-off radius of REBO potential is set to 2 Å in order to avoid non-physical strain hardening at higher strains. The average force acting on an atom is obtained by the gradient of the strain energy-strain curve. In AFEM, the direct tangent stiffness method is used to calculate the average force on an atom and PBC are implemented on the inside atoms by considering energy contribution from atoms outside the box (in blue). The arrows indicate the strain direction. The stiffness matrix is evaluated at strain intervals of 0.005.

Fig. 2(a) shows an excellent agreement between AFEM and MD at 1 K at lower strains. AFEM is not able to capture the softening of graphene at higher strains, but the overall agreement between the two curves is quite acceptable. The AFEM in the current form also does not account for temperature effects and is applicable only at very low temperatures

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(≈ 0 K), while MD can be used to model structures at different temperatures. We study the effect of temperature on the force-strain curve of the graphene sheet in Fig. 1(b) by performing a MD simulation at 300 K. All other MD parameters are kept at the values mentioned before. Fig. 2(a) shows that temperature has a minor effect on the force-strain curve, but it affects the failure point significantly.

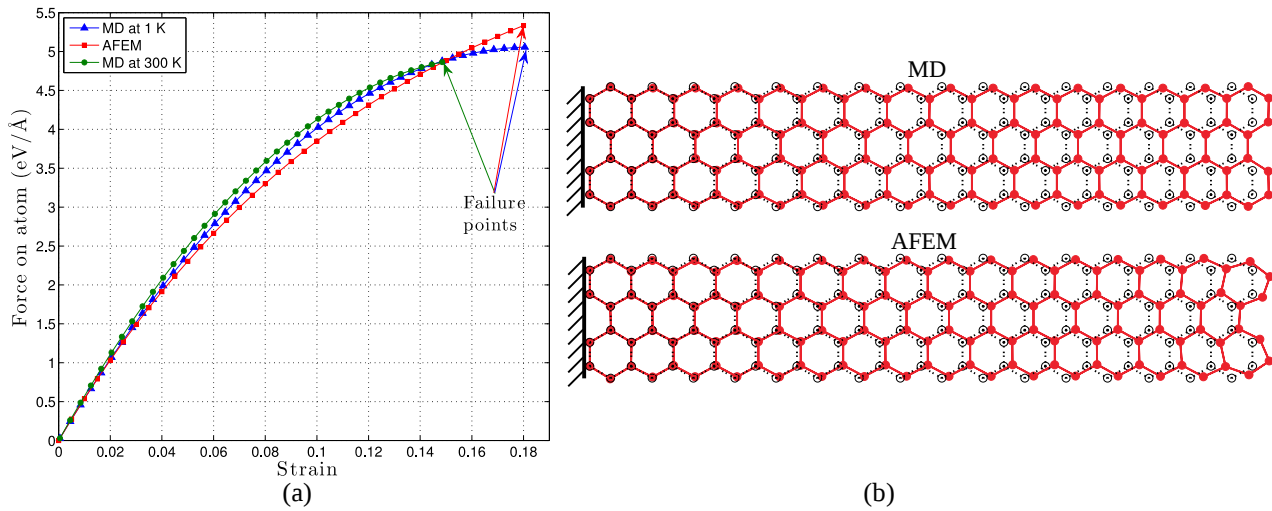


Figure 2: (a) Comparison of force on atom-strain curves of the graphene sheet in Fig. 1(b), (b) Comparison of the effect of edges on the equilibrium configuration of a graphene sheet. Hollow circles indicate the initial configuration of atoms. Fixed boundary condition is imposed at the left edge, while the other three edges are free in both MD and AFEM.

We also compare the equilibrium configuration of a narrow graphene sheet given by AFEM and MD. It has been observed that the compressive force at free edges put graphene sheet into a different equilibrium configuration [7]. $\mathbf{P}_{ele}$ of AFEM directly gives the compressive force acting on the edge atoms, and we find that $\mathbf{P}_{ele}$ is exactly equal to the edge force given by MD. In MD simulation, we allow a graphene sheet to reach its equilibrium at 1 K over a time period of 30 ps with a time step of 0.5 fs. Fig. 2(b) shows a comparison of the equilibrium configurations given by AFEM and MD. Apart from the distortion at the right edge, the deformation given by AFEM quite well agrees with the results obtained from MD. MD is an iterative method, and it took 60,000 iterations to get into the final configuration whereas AFEM gives the equilibrium in only one step.

Fig. 2(a) shows that AFEM accurately explains the global behavior of graphene, while Fig. 2(b) indicates that AFEM can also capture local behavior at individual atom level, which continuum approaches fail to do. This validation confirms the potential of AFEM in the study of more complex problems related to graphene such as buckling, fracture, defects and wave propagation.

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