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Adaptive and coupled continuum-molecular mechanics simulations of amorphous materials

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Abstract: A method to reduce the degrees freedom in molecular mechanics simulation is presented. Although the approach is formulated for amorphous materials in mind, it is equally applicable to crystalline materials. The method can be selectively applied to regions where molecular displacements are expected to be small while simultaneously using classical molecular mechanics (MM) for regions undergoing large deformation. The accuracy and computational efficiency of the approach is demonstrated through the simulation of a polymer-like substrate being indented by a rigid hemispherical indenter. The region directly below the indenter is modelled by classical molecular mechanics while the region further away has the degrees of freedom (DOFs) reduced by about 50 times. The results of automatically reverting regions of reduced DOFs back to classical MM also demonstrate the capability of performing adaptive simulations.

Keywords: Pseudo amorphous cell, multiscale simulation, molecular simulation, adaptive simulations, amorphous materials.

1 Introduction

Many recent technological advances have been made possible through an understanding of materials and fabrication of structures at the molecular level. At this length scale, where molecular interactions dominate, mechanical analyses are almost always performed using molecular mechanics (MM) simulations. While they have provided important insights into the mechanical properties of material systems at the nanometer scale, MM simulations are limited to systems of at most micrometer dimensions because of the large number of molecules involved. In situations where events at the molecular level are strongly coupled to events at a much larger scale, multiscale simulation techniques to bridge molecular and continuum levels are needed to accurately describe these multi-physics problems. A multiscale simulation technique, formulated to accommodate the amorphous nature of polymer systems, is presented in this paper.

2 Multiscale methods

Literature on multiscale models with regards to coupled atomistics and continua can be categorized as either (i) formulation of new multiscale approaches, (ii) improvements to existing models or (iii) application of multiscale models. Of the various multiscale approaches that have been proposed, those that have seen significant applications can be broadly classified into (i) Coarsed Grained Molecular Dynamics, (ii) handshake models, (iii) Quasicontinuum models and (iv) projection models.

In Coarsed Grained Molecular Dynamics (CGMD), not all molecules are represented independently in the computational model. Instead, clusters of molecules are grouped together to form a bead or a grain. Each bead is then treated as a large molecule in what is then essentially classical Molecular Dynamics simulation [1-3].

Another common approach in multiscale modelling, is through the use of a “handshake” region in the computational model. The handshake region bridges the atomistic domain with the continuum domain of the model [4,5]. The main idea in handshaking is to allow the atomistic domain and the continuum domain to overlap. At the overlap, a field variable (normally the potential energy) is assumed to take on a weighted combination of the magnitudes of the same variable in the continuum and atomistic domains. The weight is normally a function that decreases monotonically from unity to zero through the overlap.

In the Quasicontinuum (QC) method, there is essentially no distinction between atomistic and continuum description of the problem domain. This is achieved by partitioning the domain into non-overlapping cells akin to the finite element method. The cells overlay the molecules of the material with the vertices of the cell coinciding with some (not all) pre-selected representative molecules. The degrees of freedom are the position (or displacement) of the representative molecules. The QC method seeks the configuration of representative molecules that gives the minimum potential energy of the computational domain. The two main simplifications in the QC method are that (i) the position of each molecule in a cell is taken to be a function of the representative molecules of the cell and (ii) the average energy of a cell can be approximated from a group of molecules within the cell [6-9].

The last group of multiscale methods is the projection method. By “projection”, it means that the field variable, usually the displacement of the molecules, is projected onto a reduced solution space. The idea stems from classical works in decomposing a complete solution into fine and coarse scales and solving for the fine scale only in regions that require it [10]. The fine scale solution subspace is orthogonal to the coarse scale solution subspace so that the total solution is simply the addition of the fine and coarse scale solutions. The method starts by defining the solution space for the coarse scale, which again is normally similar to the finite element solution space. The fine scale solution is then defined to be the difference between the complete and coarse scale solution. The coarse scale solution is usually obtained from minimizing some L2 error norm, e.g. minimizing the sum of the squares of error in molecular displacement.

3 The Pseudo Amorphous Cell

While the abovementioned methods have been formulated with general applications in mind, none have been applied to model the mechanical behavior of polymers. The mechanical modeling of polymers even at solely atomistic scale is by itself an area that has not been firmly established. Different methods of modeling polymers are still being proposed. Modeling polymers is complex because they are generally amorphous and molecular interaction comprises pair-wise bonds, long range non-bond interaction, bond bending and bond torsion. Moreover polymer chains can comprise more than 10,000 monomers per chain. Unlike lattices of metal atoms, the equilibration of polymer chains in bulk normally does not end up with the same equilibrated configurations. One way of determining the mechanical properties of polymers in bulk is through the use of the Amorphous Cell [11]. In this method, the bulk polymer model comprises a periodic assembly of repeating parent polymer chains. The repeat intervals in the three spatial directions are conveniently represented by a cuboid or cell. The parent chains are obtained by equilibrating chains of specified number of monomers with the constraints that the chains replicate themselves according to the dimensions of the cell.

The Pseudo Amorphous Cell (PAC) method for multiscale simulation in this paper makes use of the Amorphous Cell as building blocks of polymer domains to be investigated. Rather than solving for the position of all the molecules of the constituent Amorphous Cell, displacement of the vertices of the cells (a.k.a. nodes) are the unknowns. The molecular displacements are then recovered from the nodal displacement through a mapping function, T . The PAC method allows regions where atomistic details are important to be modeled by classical molecular mechanics (MM). The connection between the PAC cell regions and the MM region is seamlessly effected by augmenting the T transformation with unit matrices where the molecular displacements are to be solved directly from MM. The computational effort to solve the coupled system of equations can be shown to be dependent on the size of the MM region if the PAC cell regions do not undergo large deformation. This is in correspondence with the way the polymer domain is expected to be partitioned – PAC cell and continuum methods at regions of small strain gradients with computationally costly MM domains at regions of high deformation. In the PAC method, the T matrix is derived to extract the molecular displacements according to the modes of deformation of the cells (i.e. stretch and shear) which turn out to be very different from linear interpolation.

4 Results and discussion

To demonstrate the PAC cell method, simulations of nanoindentation of a polymer-like substrate were performed. The polymer domain is 5 cells across by 16 cells deep. The PAC cell method is applied to the bottom half of the substrate while molecular mechanics was used for the top half. The parent chain of the amorphous cell consists of 50 monomers. Hence, a 50:1 reduction in degrees of freedom is achieved for the PAC cell domain. The indentation process was also simulated using a fully molecular mechanics simulation for the entire domain for comparison.

Figures 1 and 2 are the strain contours of the substrate obtained from the multiscale and fully molecular mechanics simulations respectively at the same indentation depth. The agreement of both plots show the accuracy of the PAC cell method.

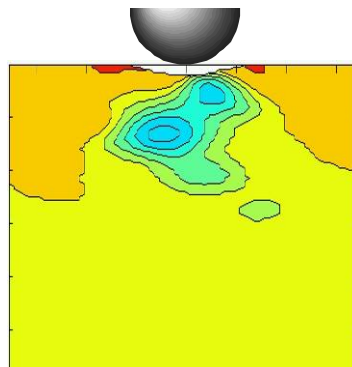


Fig. 1. Strain contours from multiscale simulation

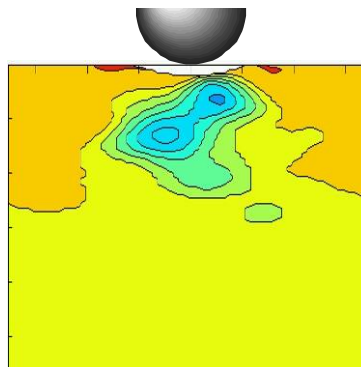


Fig. 2. Strain contours from molecular mechanics simulation

Although the displacements of atoms within PAC cells are not, and need not, be solved during the simulation, individual atom displacement can be easily obtained through the mapping function T at any stage of the simulation. This implies that whenever and wherever higher resolution in displacement is required, PAC cells can selectively be reverted to atomistic domains.

Four sets of simulations are performed – a full molecular mechanics simulation, a coupled MM-PAC simulation without adaptivity as described previously and two adaptive MM-PAC simulations. For the two adaptive simulations, the substrate initially comprises an MM domain in the top four rows of

amorphous cells with the remaining substrate represented by PAC cells. The PAC cells are reverted to atomistic domains when they experience a strain (ϵ_y) of 2% for one of the adaptive simulation and a strain of 3% for the other.

The interatomic potential of atoms which are bonded to each other is given by,

$$f_{bond}(r) = \begin{cases} -0.5kR_o^2 \ln[1 - (r/R_o)^2], & r < R_o \\ \infty, & r \geq R_o \end{cases} \quad (1)$$

and the Lennard-Jones pairwise potential for nonbonded atoms is given by,

$$f_{LJ}(r) = 4e \left(\left(\frac{S}{r} \right)^{12} - \left(\frac{S}{r} \right)^6 \right) \quad (2)$$

with a cutoff of $r = 2.5$.

The parameters of the Lennard-Jones potential are set to unity while the parameters for the bond potential are $k = 30$ and $R_o = 1.5$.

The indentation force versus depth curves from all four sets of simulations are shown in Figure 3. It shows that the indentation force predicted from the multiscale simulation and fully molecular mechanics simulations as the indenter pushes into the substrate are almost identical.

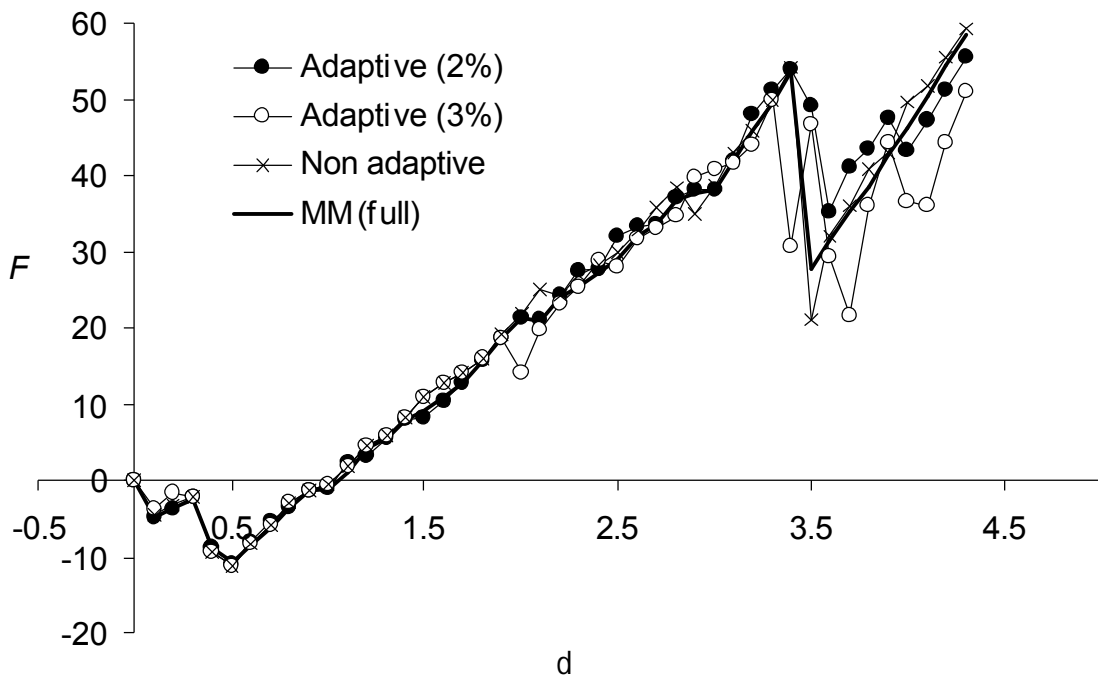


Fig. 3. Comparison of force vs indentation depth graphs from multiscale and molecular mechanics

It should be noted that a fourth fully molecular mechanics simulation was performed but with a substrate only half as thick. The results were very different. This shows that the accuracy achieved by the multiscale simulation is not because deformation is localized and completely resides within the molecular mechanics domain of all simulations.

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