Computational Materials Science 47 (2009) 1–11

Contents lists available at ScienceDirect

Computational Materials Science

journal homepage: www.elsevier.com/locate/commatsci

Nanoindentation of model diamond nanocomposites: Hierarchical molecular dynamics and finite-element simulations

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ARTICLE INFO

Article history:
Received 25 August 2008
Received in revised form 2 March 2009
Accepted 10 June 2009
Available online 15 July 2009

PACS:
62.25.−g
61.43.Bn
62.23.Pq
02.70.Dh

Keywords:
Molecular dynamics (MD)
Finite-element (FE)
Simulations
Nanoindentation
Nanocomposite
Model nanocomposite diamond
Hierarchical MD–FE modeling
Surface roughness
Diamond grains

ABSTRACT

Complementary molecular dynamics (MD) and finite-element (FE) simulations of model diamond nanocomposites (MDN) subjected to nanoindentation were undertaken to understand how contact behavior pertaining to the surface morphology of MDN surfaces can be spanned from the molecular to the continuum scale. The MD simulations determined that the behavior inside the contact region is influenced by atomic-scale features on the tip and surface, indent location, grain tilt and roughness of the MDN surfaces. In addition, if the atomic-scale surface morphology is treated as a surface roughness within the FE simulations, the same grain orientations, and similar elastic properties are used for both MD and FE simulations, there is reasonable agreement between the contact pressures for relatively low indentation loads and shallow substrates. For larger loads, the contact pressures from the FE simulations deviate somewhat from the MD results near the center of the contact. The contact behavior for length scales that are prohibitive for MD models (e.g., deep substrates) was also examined using FE simulations. This allowed for a detailed investigation of how contact conditions and stick-slip events within the contact evolve as a function of contact pressure and continuum surface stresses.

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1. Introduction

Since the ground-breaking molecular dynamics simulations of the indentation of Au(1 1 1) [1] and of a two-dimensional Lennard–Jones crystal [2], MD simulations have been used extensively to model nanoindention. MD simulations of nanoindentation have been performed on a wide variety of materials using various empirical potentials. Some examples include the indentation of metals [1,3–9], of diamond [10–12], of Si [13–18], of non-metals and ceramics [19–26], of nanocrystalline materials [8,27], of thin films on substrates [28–31], and of self-assembled monolayers [32–35]. For more complete reviews of the use of MD to model the indentation of various materials, the reader is referred to Refs. [36–38].

Despite the inherent length-scale differences between MD simulations and scanning probe experiments, significant insight has been gained into the nanoindentation process and, at times, surprising agreement obtained between simulation and experiment. For example, force curves from the MD simulations of a gold tip interacting with a gold surface had the same qualitative trends as the force curves obtained from the AFM experiments [1]. The force curves from these simulations also showed discontinuities associated with the necking of the gold upon tip withdrawal, which could not be observed experimentally.

While MD simulations have enjoyed some success in modeling atomic-scale phenomena, FE simulations have proven effective in modeling the contact behavior and long-range elasticity in large-scale contacts. FE simulations have been applied successfully to modeling the nanoindentation of multilayered composites and long-range behavior, while MD simulations were more suitable for modeling dislocation nucleation and other processes that occur on short-time scales [36,39]. In an effort to bridge the gap between the atomic-level and macroscopic methods, several investigators have attempted to couple atomistic-based methods with continuum methods. In general, there are two approaches to coupling

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0927-0256/$ - see front matter Published by Elsevier B.V.
doi:10.1016/j.commatsci.2009.06.007

calculations at different length scales, namely the concurrent and the hierarchical approach.

Concurrent methods contain calculations based on different length scales and physical models which are coupled and evolve simultaneously. Despite the fact that this approach to multiscale modeling is more challenging, several schemes for concurrent coupling have been developed [40–46]. These methods incorporate atomistic effects within a continuum entity, such that a single computational framework can be utilized. Near the indenter, an MD analysis is utilized within a predefined region, and outside of this region, an FE technique is utilized. Miller et al. adopted an atomic-scale model based on a nonlinear finite-element formulation to analyze the stress and strain induced in a very thin film during nanoindentation [47]. Their results indicated that the microscopic plastic deformation in thin films is due to the instability of its crystalline structure, and that the magnitude of the nanohardness varies with the maximum indentation depth and the geometry of the indenter.

FE modeling and MD simulations were used by Choi et al. to model the nanoindentation of aluminum on a silicon substrate [48]. It was concluded that FE analyses have limited utility in rationalizing nanoindentation experiments in which discrete discontinuities are dominant, and that MD simulations can be used to simulate experimental observations related to defect nucleation. Ma et al. used a hierarchical model for nanoindentation based on scaling strains from MD simulations to FE models to track the evolution of dislocation densities [49]. Ortiz et al. used a quasi-continuum method, which combined orbital-free density functional theory in a finite-element framework, to model nanoindentation. Through comparisons of dislocation initiation criteria based on stress, strain, and energy, they noted the need for multiscale models that can handle larger systems [50].

In the hierarchical approach, information from an atomistic, or the fine-scale approach, is used to determine parameters for the larger-scale model. In turn, the larger-scale model, such as FE, provides information that may not be available to the fine-scale model, such as, long-range elasticity information. This approach has been used successfully to determine flow boundary conditions at solid–fluid interfaces [51,52] and constitutive laws for continuum calculations [53–55]. It is the hierarchical approach to multiscale modeling that is utilized in this work.

Recently, Robbins and coworkers examined the applicability of continuum mechanics to nanoscale contacts by comparing analytic continuum solutions to MD simulations [74,75]. Those studies showed that the roughness that arises from the presence of discrete atoms can lead to dramatic deviations from analytical continuum solutions during indentation and sliding. Contact areas and stresses can be changed by a factor of two, whereas friction and lateral contact stiffness change by an order of magnitude compared to the analytic continuum solutions. These findings underscore the importance of utilizing methods that encompass atomic-scale information in conjunction with continuum methods. In this investigation, we show that continuum scale modeling can provide qualitative predictive capabilities if molecular representations from a fine-scale model of atomic-scale surface roughness and similar elastic properties are used as input.

The objectives of this study are to elucidate the effects of surface roughness, which arises from the tilt of diamond grains, and contact point on the contact behavior of model diamond nanocomposites (MDN) at the atomic and continuum scales. The structure of the model diamond nanocomposites used here, was loosely based on the structure of nanocrystalline diamond (NCD) [56–60]. NCD is being used in a wide range of applications, such as machine tools and microelectromechanical systems (MEMS) because it has robust mechanical and desirable tribological properties [60–62]. An additional objective of this work is to understand why, and to predict how, deviations and agreement between MD and FE simulations pertaining to the indentation model diamond nanocomposite (MDN) arise. These hierarchical MD–FE studies were carried out by incorporating the surface morphology and crystalline orientations that were used in the MD simulations into the FE mesh, and by utilizing the experimentally determined elastic properties of diamond and amorphous carbon in the FE approach. Once the differences and agreement between the different computational methods are quantified, then the hierarchical computational approach can be used to investigate systems that are too computationally intensive to be examined using MD.

2. Method

Complementary FE and MD simulations of the contact of MDN using a curved indenter were carried out. Two MDN systems were created for use in the MD simulations. To facilitate comparison of the data obtained using the two methods, one of these model systems was used as a template to create the FE mesh. Care was taken to ensure that the simulated systems using the two techniques were as similar as the methods allow. The creation of the MDN for the MD simulations and the FE mesh is described in detail below.

The MDN systems for the MD simulations were created by embedding 16 hydrogen-terminated diamond grains, or crystals, in an amorphous carbon matrix that is supported by six layers of diamond (1 1 1). While the predominate facets in experimentally grown NCD films are a function of deposition conditions [63], it is possible to create films where the (1 1 1) and (1 1 0) grains predominate [60]. The model diamond nanocomposites created here are generally based on the structure of NCD films, which contain diamond grains separated by an amorphous carbon matrix [60]. It should be noted, however, that the width of the amorphous carbon region in the model diamond nanocomposites used here is larger than the amorphous regions obtained experimentally. In the MDN, 11 of the diamond grains embedded in the carbon matrix had the (1 1 1) orientation and the remaining five had the (1 1 0) orientation (Fig. 1). The predominance of the (1 1 1) orientation of the grains is observed experimentally [60]. The exposed face of the hydrogen-terminated (1 1 1) grain in the MDN was 1.13 nm $\times$ 1.24 nm and its thickness was 0.61 nm. The exposed surface of the hydrogen-terminated (1 1 0) crystal was 0.88 nm $\times$ 0.98 nm and its thickness was 0.73 nm. The entire simulation system containing the diamond grains had surface dimensions of 6.43 nm $\times$ 6.48 nm and a total thickness of 2.02 nm.

Roughness of NCD films is also a function of the growth conditions [56,57,60,64]. NCD films can have roughnesses in the range of 26–78 nm [60]. In this work, two MDN films with different roughnesses were created for use in the MD simulations. Roughness was introduced into the MDN by placing the diamond grains within the amorphous carbon matrix after adjusting their Euler angles (α, β, γ), i.e., tilt of the grain. The range of selected angles fixes the surface roughness. In particular, the centers of the diamond grains were located on a 4 $\times$ 4 lattice. The angle α was randomly selected from the range 0° to 180°. The angles β and γ were set equal to each other and randomly chosen from the range 0° to 10° for film 1 (MDN1) and 0° to 20° for film 2 (MDN2). Thus, MDN2 is rougher.

Empty spaces between the diamond grains were filled with carbon atoms to create an amorphous carbon matrix. The amorphous carbon atoms were then heated to a high temperature (approximately 8000 K), while holding the diamond substrate and the diamond grains rigid. The heating phase was approximately 2 ps. After the heating, the system temperature was lowered rapidly to 100 K in another 2 ps interval. The resulting MDN films (Fig. 1) contained the diamond grains embedded in an amorphous carbon