Plasma-surface Interactions in Material Processing

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Abstract. In plasma processes such as reactive ion etching and thin film deposition for microelectronics device fabrication, atomic-level control of surface morphologies and compositions of processed materials has become increasingly important as the device sizes diminish to the nano-meter range. While various species such as ions, neutral radicals, electrons and photons simultaneously hit the material surface in a plasma, the plasma-surface interactions can be best understood if individual elementary processes such as interaction of specific species with the surface at specific incident energy are studied separately under well controlled conditions. In this article, a molecular dynamics (MD) simulation technique is reviewed as a means to analyse plasma-surface interactions in such a manner and some sample simulation results for polymer etching and diamond-like carbon (DLC) deposition are presented.

1. Introduction
As the dimensions of semiconductor devices diminish, surface irregularities in less-than-a-nanometer scale have increasingly higher chances to affect the performance of such devices. In plasma processing used for semiconductor fabrication, therefore, etching and deposition processes that are less damaging to the surface are now in high demand. One of the most significant causes of surface damages in plasma etching processes is the kinetic energy of ions bombarding the surface. For example, in a plasma process that contains hydrogen in the gas phase, hydrogen ions, even if their kinetic energies are relatively low, can penetrate deeply into the material and damage or modify the material structures (e.g., the crystalline structure of silicon at the gate channel) below the surface. In the semiconductor industry, such damages are now carefully studied mostly with the use of plasma processing tools that are similar to, or essentially of the same type as, those used in the manufacturing lines. In such a plasma processing tool, however, it is generally extremely hard, if not impossible, to control ion and/or neutral fluxes generated from the plasma so that one could obtain detailed information on plasma surface interactions. If one wishes to understand better the atomic processes on or near the surface during processing, more carefully controlled studies on surface processing are needed.

As a means for studies on plasma-surface interactions, molecular dynamics (MD) simulation techniques are discussed in this article. Although some approximations and assumptions are used for interatomic potential functions, classical MD simulations are generally considered to be a useful tool to analyze atomic-level phenomena associated with plasma surface interactions. The results of such MD simulations can be directly compared with results obtained from corresponding beam experiments and are often found in good agreement. In the rest of this article, after briefly reviewing MD simulation techniques and describing some of its historical development up to today, MD simulation
studies on reactive ion etching (RIE) of organic polymer and formation of sp$^3$ structures during diamond-like carbon (DLC) deposition processes will be presented as sample simulations.

2. Molecular dynamics simulations

The first MD simulation study that the author is aware of on the effect of energetic impact of incoming atoms on a solid material is that by Gibson, Goland, Milgram, and Vineyard in 1960 [1], where the “radiation damage” of metallic copper, i.e., how Cu atoms in crystalline sites are displaced by an incoming ion, was examined. In 1968 the first study on direct MD simulation of sputtering (i.e., particle emission from a surface by injected particles) was reported by Harrison, Levy, Johnson, and Effron [2], where Cu sputtering yields by Ar impact were evaluated from the simulations. Subsequently a series of MD simulations studies on Cu sputtering by Ar impact based on the same or similar potential functions were published by Harrison and his coworkers, as in [3] and [4].

The studies mentioned above are exclusively on physical sputtering, in the sense that neither incoming atoms/ions nor substrate atoms form new chemical bonds among them that did not exist prior to the beam injections. In 1980, Winograd, Garrison, and Harrison examined interaction of incoming carbon monoxide (CO) with a Ni substrate [5] that involves some surface chemical reactions. Until late 80s, essentially all atomic interaction models used in MD simulations for sputtering studies were of two-body short range. However, since mid 80s, various multi-body classical interatomic potential functions have been developed for more accurate MD simulation studies on material properties. For example, in 1984, Daw and Baskes developed a method to represent multi-body interaction among metallic atoms called Embedded Atom Method (EAM), which is based on density functional theory for atomic orbitals [6]. The EAM potential functions were later generalized to represent interactions among non-metallic atoms [7]. Based on cluster expansion, Stillinger and Weber developed functional forms for multi-body interactions for covalent bonds in 1985 [8-11]. On the other hand, in 1988, Tersoff proposed different multi-body interatomic function models to represent covalent bonds [12-14], based on a general expression for bond energies derived by Abell [15]. Multi-body interatomic potential functions models used today in classical MD simulations for beam-surface interactions mostly have functional forms similar to any one or combination of those mentioned above.

To the best of the author’s knowledge, the first published MD simulation study for sputtering based on multi-body interatomic potential functions was that by Reimann, Walzl, El-Maazawi, Deaven, Garrison, and Winograd in 1988 [16], in which energetic impact of Ar on Rh was examined using EAM interatomic potential functions.

On the other hand, the first MD simulation study on semiconductor sputtering using multi-body potential functions was reported by Stansfield, Broomfield, and Clary in 1989 [17], where Si sputtering by energetic Ar impact was examined based on Stillinger-Weber-type potential functions. A similar MD simulation study was also reported by Smith, Harrison, and Garrison [18] in the same year, in which Ar impact on Si was studied based on Tersoff-type potential functions.

As to surface chemical reactions associated with semiconductor etching, Schoo<text>lcraft and Garrison studied reactions of fluorine atoms impinging on a Si surface at very low energies (in the range of 0.5 – 3eV) in early 90s [19,20], using MD simulations based on the potential functions proposed by Stillinger and Weber [10]. Using similar MD simulations with a much higher injection energy (200eV), Feil, Dieleman, and Garrison [21] reported in 1993 MD simulation of reactive ion etching for Cl passivated Si surface by energetic Ar beams, using Stillinger-Weber type potential functions developed by the same authors for the system consisting of Si, Cl, and Ar atoms.

From mid 80s to mid 90s many experimental data were accumulated on surface reactions associated with reactive ion etching of Si surfaces by Cl$_2$ gas and energetic Ar ion bombardment, obtained from well controlled beam experiments [22-27]. These experimental results prompted the interest in the mechanisms of reactive ion etching in general and the simulation studies mentioned above demonstrated that classical MD simulation is an ideal tool to elucidate such mechanisms at the
atomic level. In addition, the proliferation of inexpensive but powerful computers drove the cost of high-performance computing significantly down in 90s. Due to all such combined factors, the time became ripe in mid 90s for extensive classical MD simulation studies for nonequilibrium surface reactions caused by impinging beams.

Various MD simulation studies on Si etching by Cl or F based plasmas began to be performed from early 90’s by Weakliem, Wu, and Carter [28,29], and by Barone and Graves [30] for Si-F systems, Feil [22,31] for Si-Cl-Ar systems, by Athavale and Economou [32] for layer-by-layer etching of Cl passivated Si surface by Ar ion injections. Other studies include those by Hanson, Voter, and Kress [33-35], Helmer and Graves [36,37], Kubota, Economou, and Plimpton [37]. On the other hand, SiO2 etching were studied by Ohta and Hamaguchi [39,40] for Cl or F based plasma processes and by Smirnov et al. [41-43] and by Ohta and Hamaguchi [44] for fluorocarbon (CxFy) plasma processes.

Similarly etching and deposition processes for organic polymers have been studied with MD simulations since early 90s. Due to the limited space, we here only refer to Ref. [45] for references on work performed by mid 90s. In the following sections, some examples of MD simulations by the author and co-workers for polymer etching and deposition will be presented.

3. Outline of MD simulations
Figure 1 shows an example of substrate material used for MD simulation [46]. In this example, poly paraphenylene (PPP) is placed in a simulation box, whose perpendicular edges are aligned to the x, y, and z coordinates. In MD simulations, motion of each atom in the simulation is obtained from the integration of its equation of motion. In this particular example, the interatomic potential functions based on Brenner [47] are employed for covalent bonds and Lennard-Jones type two-body potential functions for weak Van der Waals interactions [46]. For the simulation box shown in Fig. 1, periodic boundary conditions are imposed in the horizontal directions and atoms near the bottom were assumed immobile so that the substrate will not drift when it is subject to ion bombardment. The substrate system is initially in thermal equilibrium at 300K.

Interatomic potential functions used in the MD simulation studies presented in this article are those for charge-neutral atoms and therefore all particles are assumed to be charge neutral for the sake of simplicity. In other words, energetic ions that are typically seen in plasmas are represented in this MD model by fast neutrals. This assumption may be justified under some conditions, where impinging ions are neutralized right before the impact by the transfer of electrons from the surface (Auger neutralization) [48]. However, the validity of use of charge neutral interatomic potential functions for ions, which has been commonly practiced in MD simulation studies for beam-surface interactions, has not been closely examined. In what follows, however, we do not address this question directly, either, and instead examine the validity of MD simulations by comparing simulation results with
In MD simulations, atoms or clusters of atoms are injected into the model substrate. After each injection, the system is left to evolve under the microcanonical conditions (i.e., conditions where the total energy of the system is set to be constant) for about 1 pico second (in the simulation time scale) and then rapidly cooled to a room temperature. Once the substrate system reaches the initial temperature, the next injection process will start. Such cycles of injections are repeated for hundreds or thousands of times and then physical properties such as sputtered species, surface roughness, atomic compositions near the top surface, etc. obtained from the simulations are examined. Especially, by counting the number of atoms removed from the surface, one can obtain sputtering yields of the system.

4. Sample simulations: polymer etching

Figure 2 shows atomic morphologies of the surface after injecting (a) C atoms, (b) H₂ molecules, and (c) CH₄ molecules into the PPP model substrate shown in Fig. 1 at the kinetic energy of 50 eV [49,50]. The angle of incidence was 90° and the injection dose in each was 2.4 × 10¹⁶ cm⁻² and 50 eV and the angle of incidence is normal to the surface. The z coordinate represents the height of the substrate and z = 0 represents the bottom position of the initial substrate. The portion indicated as “ordered PPP” in the figure represents PPP that are assumed to be unaffected by beam injections. [46]
erosion yield" for an atomic species A as the number obtained by subtracting the number of injected A atoms from the corresponding sputtering yield.

Figure 3 shows the net erosion yields for carbon and hydrogen as functions of injection dose for 50eV CH₄ molecules. The injection conditions for (a) (b) and (c) are the same as those given of Fig. 2(c). The only difference among the three cases (a)-(c) here is the random number sequence employed in each case that determines the lateral locations of injection. [50]

Figure 3. The net erosion yields for carbon and hydrogen $Y_C$ and $Y_H$ as functions of the injection dose for 50eV CH₄ molecules. The injection conditions for (a) (b) and (c) are the same as those given of Fig. 2(c). The only difference among the three cases (a)-(c) here is the random number sequence employed in each case that determines the lateral locations of injection. [50]

Similar polymer etching simulations by nitrogen-based NxHy radicals are found in Ref. [51].
5. Sample simulations: diamond like carbon deposition

Amorphous carbon with a high content of sp\(^3\) structures is often called diamond-like carbon (DLC) and known to have extraordinary material properties such as high mechanical hardness, high wear resistance, low friction coefficients, high chemical inertness, good infrared (IR) transparency, high electrical resistivity, good field emission properties, and low dielectric coefficients [52-58]. Heterogeneous growth of DLC films on various materials at low deposition temperatures can be achieved by plasma enhanced chemical vapor deposition (PECVD). In most PECVD processes, the growth of DLC, as opposed to amorphous carbon, can be achieved by applying a certain amount of bias voltage to the substrate, which makes ionized deposition precursors reach the surface with finite kinetic energies. Typically, if the impact energy is too low, the deposited carbon is amorphous whereas, if the energy is too high, sputtering of the surface becomes significant. DLC differs from amorphous carbon as it contains a large number of sp\(^3\) hybridized bonds. The formation mechanism of sp\(^3\) hybridized bonds in carbon deposition process is of major interest in this section. Especially we focus on the effects of impact energy and presence of hydrogen.

As in the previous section, MD simulations can reveal some aspects of the formation of sp\(^3\) hybridized bonds at the atomic level when carbon deposition processes are simulated. In actual PECVD based on hydrocarbon gases, various ions and free radicals can form deposition precursors. Here we simplify the process and simulate only single-species incidence of CH\(_3\) or CH on an a-C film that is initially hydrogen-free [59,60]. To further simplify the study, we also focus just on the initial phase of deposition. A better understanding of individual reaction processes such as those examined here will serve as the basis for a more complete understanding of real PECVD processes.

The interatomic potential functions used in the simulations shown here are those given in Ref. [61], which are similar to Stillinger-Weber functions with bond-order corrections. In this set of potential functions, the bond order of a carbon-carbon (CC) bond is determined from the number of other carbon (C) and hydrogen (H) atoms that are present in the neighborhoods of the carbon atoms that forms the bond in consideration. For example, if there are three H atoms in each neighborhood of the C atom, the CC bond is a single bond whereas if there are two H atoms in each neighborhood, the CC bond is a double bond. A function that counts the neighboring atoms is built in each interatomic potential function.

The model substrate used in the simulation is a hydrogen-free amorphous carbon (a-C) cube with an edge length of about 2nm. The system is in thermal equilibrium at 300K. The initial substrate consists mainly of sp\(^2\) hybridized bonds; the ratio of the number of sp\(^3\) C atoms to that of sp\(^2\) atoms in the model substrate is about 0.4%. As in the model in the previous section, the periodic boundary conditions are imposed in the lateral directions of the system. In the bottom layer of the substrate cube, the atoms are also set immobile.

![Atomic structures of the substrate and deposited atoms obtained from MD simulations.](image)

**Figure 5.** Atomic structures of the substrate and deposited atoms obtained from MD simulations. In each case of (a) CH\(_3\) and (b) CH incidences, the beam dose and kinetic energy are 2.0×10\(^{16}\)cm\(^{-2}\) and 20eV. The large black and grey spheres represent immobile C atoms in the bottom layer and mobile C atoms of the initial substrate. The large white and small black spheres represent injected C and H atoms. [59]
Figure 5 shows atomic structures of the system at an injection dose of $2.0 \times 10^{16} \text{cm}^{-2}$ of (a) CH$_3$ and (b) CH at 20 eV impact energy [59]. Here large black spheres represent C atoms in the bottom layer which are assumed to be immobile. The grey spheres represent mobile C atoms that initially constitute the substrate. The large white spheres represent injected C atoms. Injected H atoms are represented by small black spheres. Clearly the deposited film is thicker for CH incidence as CH has a higher sticking probability than CH$_3$.

Figure 6 shows the depth profiles of carbon and hydrogen atomic species at a dose of $2.0 \times 10^{16} \text{cm}^{-2}$ for CH$_3$ incidence with incident energies of (a) 20eV and (b) 50 eV [59]. The profiles of substrate C atoms, deposited C atoms, and deposited H atoms are given by the thick solid, broken, and thin solid curves, respectively. The horizontal axis represents the film thickness measured from the bottom of the model substrate. In this figure, the atomic density at each depth is obtained from the total number of the atoms that are present in a 1Å thick layer at the indicated depth. The figure clearly shows that deposited C and H atoms penetrate the substrate more deeply at higher energy. Furthermore, H atoms penetrate the substrate more deeply than C atoms because a H atom has a smaller interaction length (i.e., atomic radius) than a C atom.

In this study, the “carbon-to-carbon (C-to-C)” coordination number is defined as the number of C atoms that exist in the neighborhood of a particular C atom that we consider. For example, in crystalline diamond, the C-to-C coordination number of every C atom is 4. If the C-to-C coordination number of a C atom is 4, then the C atom forms sp$^3$ hybridized bonds with the surrounding C atoms. Similarly, if the C-to-C coordination number for a C atom is 3, it forms sp$^2$ hybridized bonds with the surrounding C atoms. Figure 7 shows the ratio of the number of C atoms with each C-to-C coordination number deposited on the substrate to the number of all deposited C atoms as functions of the incident energy at a dose of $2.5 \times 10^{16} \text{cm}^{-2}$ for (a) CH$_3$ and (b) CH beam incidences [59]. Especially, the ratio for the C-to-C coordination number being 4 represents the fraction of fully carbon terminated sp$^3$ C atoms (each of which is bonded with 4 other C atoms only, not more and not less) for all deposited C atoms. This fraction represents the “diamond-likeness” of the deposited film. Note that C atoms that constitute the initial substrate (which are mostly of sp$^2$, as mentioned earlier) are not counted in Fig. 7. However, in the C-to-C coordination numbers listed here, bonds connecting deposited C atoms and substrate C atoms are counted. The “others” in Fig. 7 include deposited C atoms bonded with either H atoms or those bonded with five or more other C atoms (although the latter are scarce).

**Figure 6.** Depth profiles of atomic species at a dose of $2.0 \times 10^{16} \text{cm}^{-2}$ of CH$_3$ incidence with incident energies of (a) 20eV and (b) 50 eV, obtained from MD simulations. The profiles of substrate C atoms, deposited C atoms, and deposited H atoms are given by the thick solid, broken, and thin solid curves, respectively. The horizontal axis represents the film thickness measured from the bottom of the model substrate. [59]
It is clearly seen in Fig. 7 that, for CH$_3$ incidence, deposited C atoms are more likely to form sp$^3$ hybridized bonds with other C atoms at higher incident energy under the conditions studied here. A similar tendency is also seen for CH incidence up to 20eV. However, at 50eV for CH incidence, the sp$^3$ fraction is significantly lower, which is likely caused by relative scarcity of hydrogen in the substrate. The sticking coefficient of H atoms for CH incidence is low at high energy.

Figure 8 shows the density profile of fully carbon terminated sp$^3$ C atoms at a dose of 2.0×10$^{16}$cm$^{-2}$ for 50 eV CH$_3$ incidence (depicted by the grey curve denoted by “sp3 C”) [59]. For reference, the density profiles of C and H atoms are also plotted here, which are the same as those given in Fig. 6(b). Note that “the fully carbon terminated sp$^3$ C atom” means a C atom that is bonded with four other C atoms only, as defined in Fig. 7. It is clearly seen that the sp$^3$ C atoms are mostly located in a level below the top surface (i.e., subsurface level), typically around 1 nm below in this case.

6. Conclusions
A technique to apply MD simulation for the analysis of plasma-surface interactions has been reviewed with some sample simulation results. For example, in semiconductor manufacturing, sizes of electronic devices on a chip, such as transistors and interconnect wires, continue to diminish and damages on a processed surface affect device performance more severely as the miniaturization continues. Atomic-scale analyses for plasma-surface interactions may provide useful information for the development of technologies that alleviate such problems.

The simulation results presented in this article have not been compared with experimental results as there has been no experiment data with which the simulation data given here can be directly compared. However, in many cases, MD simulations have been found to provide macroscopic data such as sputtering yields and atomic compositions on the surface that are in reasonably good agreement with experimental observations (e.g., [39,40]). Some may find this counterintuitive as chemical reactions on the surface often play important roles in interaction between a chemically reactive plasma and a surface whereas a classical model of atomic interactions is known to be nearly incapable of representing actual chemical reactions in thermal equilibrium. The reason that classical MD simulations can predict some characteristics of chemical sputtering and/or deposition processes is that, in such processes, the injection energy (which is in the order of a 100eV or so) is far higher than bond energies and therefore the outcome, including some chemical reactions, is less sensitive to the detailed structures of interatomic potential functions. It is mostly likely that only a small number of parameters

Figure 7. The ratio of the number of C atoms with each C-to-C coordination number deposited on the substrate to the number of all deposited C atoms as functions of the incident energy at a dose of 2.5×10$^{16}$cm$^{-2}$ for (a) CH$_3$ and (b) CH beam incidences, obtained from MD simulations. The “others” include deposited C atoms bonded with one or more H atoms or those bonded with five or more other C atoms (although the latter are scarce). [59]
such as the bond energy and bond length are the essential parameters that determine macroscopic outcomes of sputtering or deposition phenomena.

In this article, only the cases of ion injections (i.e., injections of energetic species) into the surface have been discussed and no neutral radical flux effect has been presented. However, effects of neutral radical fluxes can be incorporated in MD simulations in a relatively straightforward manner by the addition of low-energy fluxes of reactive species, as shown in, e.g., Refs. [62-64].

Experiments that can be directly compared with MD simulation are beam experiments. The reader is referred to, e.g., Refs. [65-69], in which mass-selected ion beam experiments and some sputtering yield data are presented in detail.

Figure 8. The depth profile of fully carbon terminated sp$^3$ C atoms at a dose of $2.0 \times 10^{16}$cm$^{-2}$ for 50 eV CH$_3$ incidence (depicted by the grey curve denoted by “sp$^3$ C”), obtained from MD simulations. For reference, the profiles of C and H atoms are also plotted here, which are the same as those in Fig. 6(b). [59]

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