BEAM
INTERACTIONS
WITH
MATERIALS
AND ATOMS

Plasma-surface interactions of hydrogenated carbon

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ABSTRACT

We present a review of our study of interactions of plasma particles (atoms, molecules) with hydrogenated amorphous carbon surfaces typical of plasma-facing divertor tiles and deposited layers in magnetic-fusion reactors. Our computer simulations of these processes are based on classical molecular dynamics simulations, using the best currently available multibody bond-order hydrocarbon potentials. Our research in this field has been focused on the chemical sputtering of carbon surfaces at low impact energies, the most complex of the plasma-surface interactions (PSI). Close collaboration with beam-surface and plasma-surface experiments provides not only theoretical support for the experiments, but also builds suitable benchmarks for our methods and codes, enabling production of theoretical plasma-surface data with increased reliability.

1. Introduction

Since plasma-boundary physics encompasses some of the most important unresolved issues in future energy production for fusion reactors, there is a strong interest in the fusion community for better understanding and characterization of plasma-surface interactions (PSI). Chemical and physical sputtering cause the erosion of the limiter/divertor plates and vacuum-vessel walls, whether these are made of C, Be, W, or some combination (i.e. mixed materials), and degrade fusion performance by diluting the fusion fuel and excessively cooling the core. Hydrocarbon re-deposition onto plasma-facing components can lead to long term accumulation of large in-vessel tritium inventories.

A major challenge in the production of D–T fusion power is the development of materials for the first wall and internal components. The choice of wall material has profound effects on confinement of fusion-grade plasmas. Although carbon-based materials have superior thermo-mechanical properties, they could trap high levels of tritium by co-deposition with eroded carbon and thereby severely constrain plasma operations. Thus, a mix of several different plasma-facing materials is now proposed in ITER to optimize the requirements of areas with different power and particle flux characteristics. The slow rate of progress in the area of tritium removal, together with favorable results from divertor tokamaks with high atomic number (e.g. tungsten) walls, suggest that interest in all-metal machines will increase.

The key issues for surface experiments on carbon-based materials are erosion, reflection, impurity transport in the plasma, deposition, T uptake and removal. There is a need to determine the composition of eroded species such as hydrocarbon molecules and radicals, their rovibrational state and energy spectra, and their sticking coefficients to surfaces as a function of energy. An expansion of the available erosion database towards very low energies (5 eV) is needed in order to narrow the gap in erosion data between energetic hydrogen ions and thermal atomic hydrogen, both for pure carbon as well as mixed materials systems. Properly benchmarked molecular dynamics (MD) simulations could provide comprehensive databases for boundary plasma modeling, providing details often not accessible by experiment.

Chemical sputtering is a process where bombardment by atoms or molecules induces a chemical reaction which produces a particle that is weakly bound to the surface and hence is easily desorbed into the gas phase [1]. For carbon surfaces, it is hypothesized that incident particles break bonds within the collision cascade [2]. The broken bonds are rapidly passivated by the abundant flux of atomic hydrogen from the hydrogen fusion plasma environment. This leads to the formation of stable hydrocarbon molecules

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underneath the surface, which diffuse to the surface and desorb thermally.

In our MD simulations the classical motion of a collection of atoms is followed using forces obtained from predefined analytic ground electronic state potentials, which for hydrocarbon systems have reached maturity (empirical bond-order potentials of the Tersoff–Brenner type, such as REBO [3,4] and AIREBO [5]) and their predictive strength is improving. The parameters of the phenomenological potential functions are taken either from parameter-free quantum-mechanical calculations or experimental data. Dynamic electronic effects are not included in classical MD. Non-local effects, which take into consideration the bonding structure further away than the nearest neighboring atoms, may be included in the potential to improve the accuracy of the calculations. This is especially important for carbon, where heterogeneous bonding configurations arise from the presence of hybridized bonding. The REBO potential has a reasonably realistic description of pure carbon and hydrocarbon molecular structures, as well as dynamic effects, such as bond forming and breaking. Hence, it has been adopted by most MD studies of sputtering [6–9] including our own [10–15]. The AIREBO potential overcomes some limitations of the REBO potential, but requires significantly faster computer resources, due to the longer range of the potential.
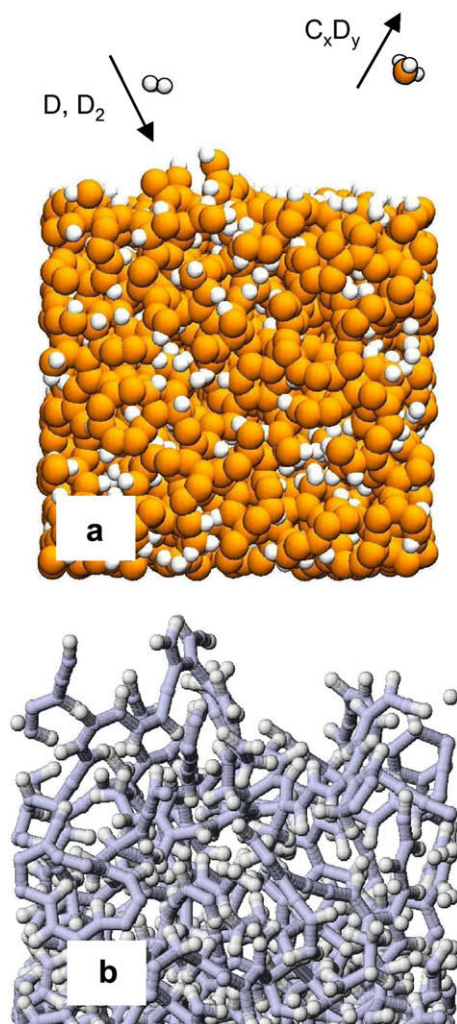
The numerical cell must be larger than the characteristic region of atom impact induced perturbation, to secure scattering-free cell boundaries. In the range we have studied [10–15], below 30 eV impact energy, a realistic cell of deuterated carbon consists of a cubic random C:D network of several thousand atoms, with edge dimension of 2.5 nm (Fig. 1(a)) and two-dimensional periodic boundary conditions in the planes perpendicular to the surface plane. Saturated surfaces are prepared by cumulative bombardment of several thousand projectiles during creation times up to 10 ns until a saturation regime is reached [10,15]. Cumulative bombardment not only erodes the surface, but also creates surface swelling producing a lower-density interface that is inhomogeneous and contains filamentous structures [14] of hydrocarbon chains (Fig. 1(b)), which play an important role in the PSI dynamics. Bombardment also changes the D/C ratio close to the surface interface (extending to increasing depths with increasing impact energy), where it reaches values $\geqslant 1$, as seen in Fig. 2. Sputtering yields typically reach a nearly stationary regime as a function of fluence. However, this stationarity can never be a true steady-state since the finite simulation cell is continually changing due to erosion. In any case, chemical sputtering is strongly dependent on the accumulation of hydrogen in the surface and, consequently, depends on the concentration of carbon atoms with $sp/sp^2/sp^3$ hybridization states, which also changes with fluence. This explains the change in the total carbon yield as a function of the deuterium fluence, shown in Fig. 3, which is also seen experimentally [16]. Most of our simulations were done with surfaces prepared in the range of 1000–2000 impacts (at or beyond the point where the sputter yield has reached an approximate stead state in Fig. 3).

After creating the surfaces by cumulative bombardment, sputtering yields are subsequently calculated by independent random impacts of several thousand projectiles on the saturated surfaces [10]. At the lowest energies the time duration of the collisional cascade for each impact is less than 5 ps, but approaches 30 ps for 30 eV deuterium impacts [15]. Calculations are performed using 2000–4000 independent trajectories for 5–30 ps. Such level of statistics is essential for meaningful calculation of small yields. Our recent calculations [10–15] of impact of deuterium atoms and molecules have shown good agreement with the experimental results [17] for sputtering of methane and acetylene. This good agreement for selected hydrocarbon yields is the result of our computational methodology: we mimic the cumulative saturation conditions of
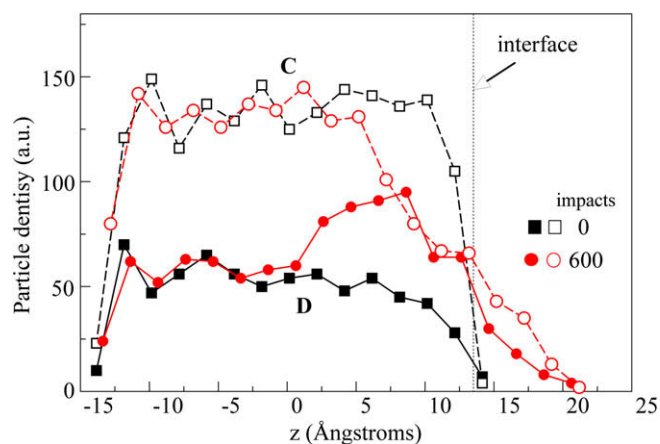

Fig. 1. (a) Chemical sputtering upon impact of D and D_2 projectiles on deuterated amorphous carbon and (b) filamentous low-density structures at the interface after prolonged bombardment with 20 eV D.

Fig. 2. Depth profile for the density of carbon (open symbols) and deuterium (solid symbols) in the simulation cell, both for the initial surface (black squares) and for the surface after 600 impacts (red circles). Initially, the cell was created from a random distribution with a D/C ratio of 0.4, homogenized by a heating and annealing. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)