Molecular Dynamics Simulation of Low-Energy Sputtering of Molybdenum with Xenon Ions*+

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A study has been initiated to investigate interactions between low-energy xenon ions and molybdenum using molecular dynamics (MD) simulation. An MD code, Simulation Kit, designed for simulation of atomic collisions in solid lattices was used in this study. The ion energies ranged from 150 to 500 eV. The ions impinged on the target normal to the (110) plane. The target consisted of 6 layers, each nominally consisting of 306 atoms. The standard ZBL potential with a screening length correction factor of 1.1 has been used in this investigation. Each simulation was terminated after 1000 fs. The sputtering yields obtained from these simulations are in reasonable agreement with existing data.

Introduction

The principle life-limiting element in an ion propulsion system is the thruster ion optics [1]. In a thruster with two grid optics, the negatively biased accelerator grid is subject to the impact of charge exchange ions produced just downstream of the grid [2]. The energy of these charge exchange ions is approximately 200 eV. The positive grid is bombarded on the upstream side by low-energy ions produced in the discharge chamber. Most of the sputter erosion is due to the doubly charged ions peaked at the center of the thruster. The energy of the doubly charged ions is twice that of the singly charged ions, which is approximately equal to the discharge chamber voltage of about 25 V.

Sputtering yields have been measured at low ion energies for various metal-ion combinations. However, there are uncertainties about the low-energy yield data, particularly below 200 eV. There are two reasons why it is difficult to measure sputtering yields accurately below 200 eV. First, the ion current density is very low at these energies. Second, the sputtering yield begins to drop off significantly around 200 eV. Thus hundreds of hours of sputtering time is needed to collect statistically significant yield data.

Several methods have been tried successfully to measure sputtering yields at low ion energies. These are:

1. Weight loss method [3,4]
2. Optical spectroscopy method [5]
3. Radioactive tracer method [6,7]
4. Secondary neutral mass spectrometry [8,9]
5. Rutherford backscattering spectrometry (RBS) [10,11]
6. Quartz crystal microbalance method [12]

Each method has its disadvantages [10]. However, it appears that the RBS method is sensitive enough to measure sputtered film thickness of 1 monolayer and has the potential of providing accurate sputtering yield data at energies below 200 eV [11].

Besides conducting experiments, investigations in sputtering can be carried out in two other ways, analytically and through computer simulation. The analytical approach that is currently accepted has been developed by Sigmund [13]. It assumes the formation of collision cascades inside the target due to the

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passage of a high-energy ion. Hence, it is not applicable to low-energy sputtering where no collision cascades are formed. To determine sputtering yields more accurately at low ion energies, semi-empirical correction factors to Sigmund's formula have been proposed [14,15]. Although these formulations provide a reasonable agreement with the measured sputtering yields, they both use very high values of threshold energies. Most estimates of sputtering threshold energies have indicated values 2 to 4 times the sublimation energy of the targets. These are significantly below the threshold values used in the theoretical calculations [16].

Simulation efforts have led to the emergence of two distinct computational approaches, the binary collision (BC) approximation model and the molecular dynamics (MD) model [17]. The BC codes treat a collision sequence as a series of binary collisions between two atoms. The BC codes are more successful in modeling phenomena in high-energy sputtering. However, these codes have been used in investigating low-energy sputtering processes as well [18].

In contrast, the MD model treats the collision system as a classical many-particle ensemble and takes into account multiple interactions involving both projectile and target atoms. MD codes solve Newton's equations of motion numerically with forces derived from potential functions. To accurately portray the physical processes, these codes use extremely small time steps, usually of the order of 1 fs. Hence they need large computer times for each simulation. With the easy availability of fast computers, MD methods are beginning to be used more widely in simulating ion-solid interactions [19].

The strength of the MD simulation lies in its greater accuracy, particularly in modeling many-particle interactions at low energies. In view of this, we have initiated a systematic investigation of the applicability of the MD simulation technique to low-energy sputtering of Mo by Xe ions. A PC software package designed for MD simulation of atomic collisions in solids was used in this study. The suite of programs in this package is collectively known as the Simulation Kit (SK) [20]. The physical model, and the assumptions underlying the MD simulation performed by the SK code are described in the literature [17]. The simulation techniques used by SK closely follows the methodology used by Smith and Harrison [21]. Results of the first part of this investigation involving the sputtering of molybdenum with xenon ions in the energy range 150 to 500 eV are reported in this paper.

Theory

When an ion is impacted on a target atom (here referred to as an anchor atom), not only it but also the surrounding atoms are set in motion. An MD simulation follows the movement of these atoms as a function of time. The motion of this ensemble of N atoms is described by a set of coupled differential equations of classical dynamics:

$$m_i \frac{d^2 r_i}{dt^2} = -\nabla V \left(i = 1,2,.....N\right) \tag{1}$$

where m_i and r_i are the mass and position of the ith atom respectively and V represent the potential function which describes the interaction between the particles. To keep the computation time to a reasonable level, one usually assumes that the motion of any particle is influenced by the interaction of only those atoms that reside within a cutoff radius, R_c, of the particle, i.e.

$$\left|r_i - r_j\right| \le R_c \tag{2}$$

Usually R_c is chosen such that it lies between the 1st and 2nd or 2nd and 3rd nearest neighbors.

In this simulation it is further assumed that all interactions could be expressed by pairwise potentials which could be summed over all particles, i.e.

$$\nabla V = \sum \nabla_i V\left(\left|r_i - r_j\right|\right) \tag{3}$$

where the summation runs over the index j (j = 1,….N and $j \neq i$).

The interaction between two atoms due to repulsive forces is described by an interaction potential which depends only on the nuclear charges and the internuclear distance. The interaction potentials commonly used are screened Coulomb potentials. Interaction between target atoms will also include attractive potentials. We have used the Ziegler-Biersack-Littmark (ZBL) screened Coulomb potential

to describe the projectile-target interaction. The ZBL potential represents a mean value as it was constructed from determination of interaction potentials of about 500 ion-target combinations [17]. The interaction between target atoms was modeled using the ZBL repulsive potential at short internuclear separations and a Morse attractive potential at large separations. A cubic polynomial spline function joins the two potentials in an intermediate region. A switching function smoothly brings the potential to zero at the cut-off distance.

Simulation Parameters

Ions are impacted on the (110) plane of a Mo microcrystal target at normal incidence. This surface was chosen as atoms are densely packed on this plane. Mo has a BCC crystal structure with a lattice constant of 0.31469 nm. The target consists of 6 atomic layers, each layer nominally consisting of 306 Mo atoms. The edge atoms have been trimmed off, leaving a total of 1,734 atoms in the target. The projectile is normally incident on the target on the x-y plane. The impact parameter on the anchor atom is varied in each run at a given ion energy. As shown in Fig. 1, the projectile impact zone on the target surface is triangular in shape. In this simulation, 712 impact points have been used for a given ion energy.

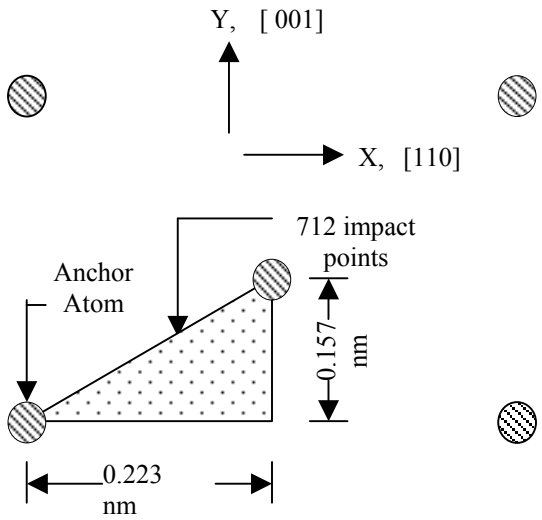

Figure 1 – Projectile impact zone. The target surface lies in the (110) plane.

The 2nd and 3rd nearest neighbors of a Mo atom lie at distances of 0.315 and 0.445 nm respectively. The potential is cut-off at 0.38 nm. The switching function is applied beyond 0.36 nm to terminate the potential smoothly at the cut-off distance. The standard ZBL potential was used to describe repulsive interactions and a Morse potential was used to describe the attractive interactions between Mo atoms. A spline was used to join the Morse and ZBL Mo-Mo potentials in the region 0.17 to 0.22 nm. A surface cohesive energy of 8.12 eV was used in this study. Vibrational displacements of target atoms have been included assuming a target temperature of 300 K. The rms value of these displacements is about 0.02 nm.

No inelastic losses were used in this simulation. A 500 eV Xe ion is not moving very fast, so inelastic losses are expected to be a small fraction of its kinetic energy.

The target surface lies in the x-y plane. The anchor atom is placed at the origin of the coordinate system. The simulation is initiated with the projectile located 0.4 nm above the surface. At 500 eV, a Xe ion travels 0.01 nm in 0.37 fs. Hence, each simulation is initiated with a slightly smaller time step of 0.3 fs. Verlet's integration algorithm was used to solve the differential equations.

Results and Discussion

Each simulation was run for 1000 fs as this provided the optimum result. Projectile energies ranged from 150 to 500 eV in steps of 50 eV. Below 150 eV, no sputtering was observed with the parameters used in these simulations.

It is the usual practice to adjust the interaction potential by incorporating a screening length correction factor to fit simulated results to experimental values. The correction factor scales the screening length parameter in screened Coulomb potentials. We have used the ZBL potential with a screening length correction factor of 1.1 for both projectile-target atom and target atom-target atom interactions. This value provides the best sputtering yield over the energy range investigated.

When a projectile is impacted on the anchor atom, it sets the atoms of the target in motion. At the termination of simulation, it is observed that a large

number of atoms are above the surface and moving away from the surface. Figure 2 shows the total number of atoms located beyond a certain distance above the surface at the end of simulation when 500 eV Xe ions were used as projectiles. It can be seen that below 0.3 nm, the number of atoms above the original surface begins to rise steeply. Between 0.3 and 1.0 nm, the number of atoms remains constant and beyond 1 nm, the number begins to fall rapidly.

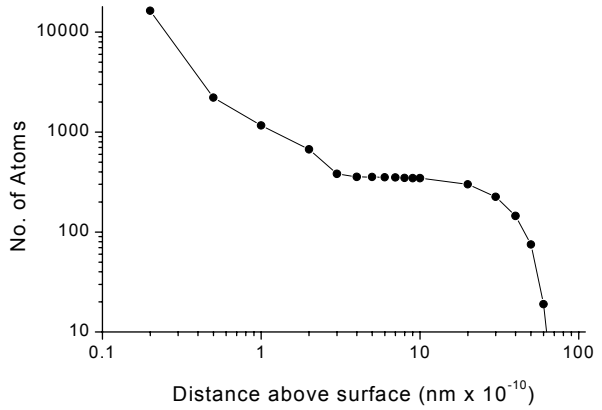

Figure 2 - Distribution of Mo atoms above the surface for a Mo(110) target bombarded with 500 eV Xe ions.

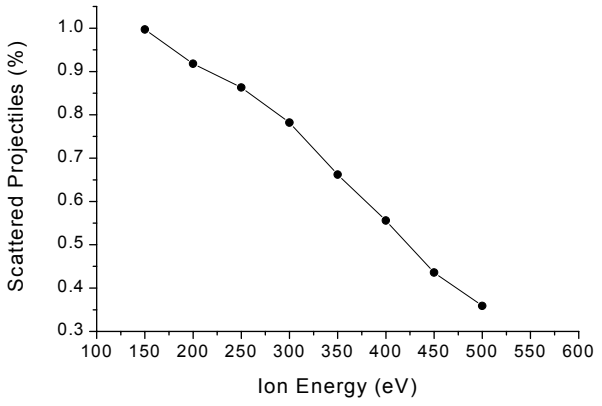

Figure 3 – Percentage of scattered projectiles as a function of ion energy

Most of the atoms located below 0.3 nm have insufficient upward energy component to allow them to escape. These atoms eventually fall back on the surface of the target. To avoid counting these atoms as sputtered atoms, we have defined sputtered atoms as those which are more than 2.5 interlayer distance from the surface. The interlayer distance in Mo(110) is 0.2225 nm. Hence, only those atoms found beyond 0.556 nm and moving away from the surface at the termination of simulation, were counted as sputtered atoms in this study. This distance lies in the middle of the plateau in Fig. 2.

Many projectiles were found to turn around after penetrating 1 to 2 atomic layers and escape the target surface (Fig. 3). Nearly all projectiles were scattered back at 150 eV. The percentage of scattered projectiles decreased almost linearly with increasing ion energy. At 500 eV, 36% of the projectiles were found to scatter back from the target.

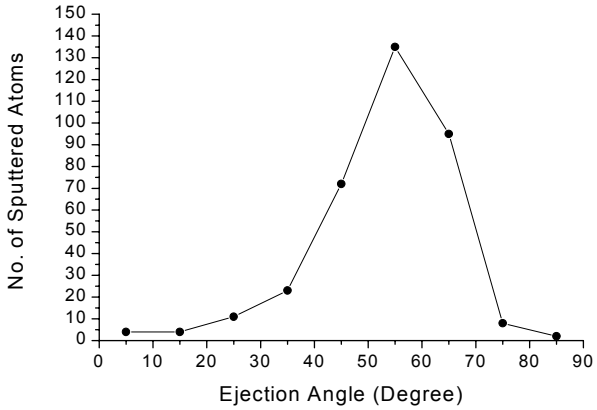

Figure 4 – Number of sputtered atoms as a function of ejection angle.

The angular distribution of the sputtered atoms is shown in Fig. 4. The distribution is under-cosine and it peaks at 55^{o}. This compares favorably to the measured peak of the angular distribution at 45^{o} [10].

Table 1. Low-Energy Sputtering Yields of Mo with Xenon, Measured and Simulated

Energy (eV)	Ref. 3	Ref. 4	Ref. 9	Ref. 10	Ref. 12	This work
150			0.09			0.004
200	0.28	0.20	0.30	0.18	0.13	0.052
250					0.22	0.112
300	0.51	0.40	0.60		0.50	0.211
350						0.288
400	0.70	0.60	0.65		0.73	0.369
450						0.442
500		0.69	0.70	0.73	0.72	0.497
600	1.06	0.88	0.78		0.90	

Sputtering yields obtained from this simulation are presented in Table 1 and Fig. 5. Low-energy Mo sputtering yield data measured by other researchers with Xe ions are also presented in Table 1 and Fig. 5 for comparison [3,4,9,10,12]. However, only a few experimental data points are shown in Fig. 5 to avoid clutter.

The results indicate that the sputtering yields obtained from this simulation are lower than those obtained from various measurements. The difference in sputtering yields is observed to be larger at lower ion energies. For example, at 500 eV, the difference is about 40% whereas at 200 eV, the simulated yield comes out lower by a factor of about 4. It is clear that the same interaction potential can not be used over the entire energy range.

The simulated sputtering yields are expected to be somewhat different from experimental data. The simulation uses a perfect crystal configuration for each ion impact whereas in experiments polycrystalline targets are used. Experiments also involve prolonged bombardment of the surface. This destroys much of the crystal order.

No sputtering was observed with these simulation parameters at 100 eV. It is interesting to note that the same observation was made by another research group when Xe was incident normally at Ni (which has a FCC crystal structure) [22].

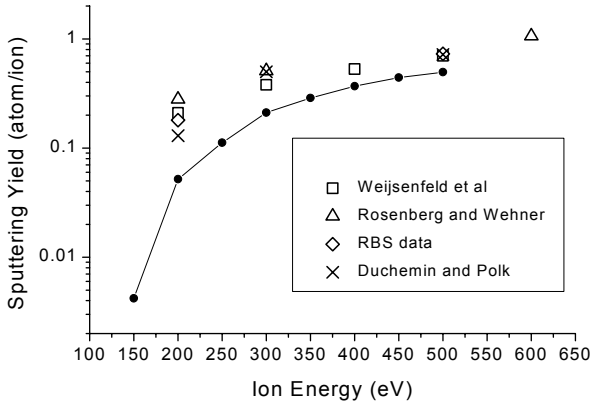

Figure 5 – Sputtering yield of molybdenum as a function of xenon ion energy. Results obtained from computer simulation are presented as solid circles.

Conclusions

Sputtering of Mo by low-energy Xe ions was investigated using MD computer simulation method. Xe ions with energies ranging from 150 to 500 eV were normally incident on the (110) plane of a Mo microcrystal target. The sputtering yields obtained from this simulation were found to be lower than those obtained from experiments but the correct trend of the simulated results is encouraging. The simulation reveals that a different interaction potential is needed at lower ion energies where discrepancies between the measured and simulated data become large.

Acknowledgements

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