Surface & Coatings Technology xxx (2016) xxx–xxx

Contents lists available at ScienceDirect

Surface & Coatings Technology

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

Molecular dynamics simulations study of nano particle migration by cluster impact

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ARTICLE INFO

Article history:
Received 1 February 2016
Revised 13 April 2016
Accepted in revised form 24 April 2016
Available online xxxx

Keywords:
Molecular dynamics simulation
Gas cluster ion
Surface cleaning

ABSTRACT

Molecular dynamics (MD) simulations are performed in order to investigate the radiation effects of a huge and slow gas cluster for the surface cleaning process. When a large argon cluster with the size ranging from 20,000 to 300,000 is accelerated with a total of 30 keV, each constituent atom carries very low energy ranging from 1.5 eV/atom to 0.1 eV/atom. In many cases, the cluster does not penetrate the solid target surface but is deflected in a lateral direction. This collisional process results in a high density particle flow spreading along the surface plane due to cohesion of the cluster, which suggests the capability to modify the irregular surface structure, without damage in the target. The MD simulations demonstrate that such a huge cluster sweeps a nano particle (NP, 3 nm in radius) attached on a planar silicon target's surface. From the investigation of various conditions of cluster impact, it is found that the migration distance is correlated with the kinetic energy applied on the NP by the impact of cluster atoms. Additionally, the MD results suggest the existence of optimized parameters for the maximum migration distance for the offset distance between the cluster and the NP, and the cluster size for constant total energy (equivalent to energy per atom or kinetic energy density). The optimized offset distance was estimated as the summation of radii of the incident cluster and the NP. The optimized energy per atom was suggested around 0.6 eV/atom, where the cluster efficiently spreads in lateral direction keeping higher kinetic energy density of particle flow.

1. Introduction

The gas cluster ion beam (GCIB) is a unique ion beam technique where a gas cluster, a large aggregation of source atoms/molecules, is generated as a cluster, and then ionized, accelerated, and radiated on the target. Several experiments [1,2] have demonstrated that the glancing angle irradiation of an energetic cluster beam can remove surface roughness without damage, which suggests that the GCIB process can be applied for an efficient surface cleaning process under dry process conditions. The glancing angle incidence of the GCIB is a useful technique to treat a planar surface or the side wall of a trench and pillar structure. On the other hand, the demand for efficient surface cleaning with a dry process arises from the cleaning of semiconductor wafers with a mask structure kept on the surface. Dobashi [3] and his group reported that ultrafine particles are removed without damage on the wafer surface or pattern structure by utilizing a CO_2 GCIB at several tens of keV of incident energy.

In the previous works performed in both experiments [4,5] and simulations [6], it has been demonstrated that low-damage cluster irradiation can be realized by controlling not only the incident energy but also the cluster size or incident angle. The value of the normal component of the incident energy per atom is proposed as a characteristic parameter to determine whether the incident cluster penetrates the target surface to cause crater-like damage. As mentioned before, a large cluster should be selected to reduce the damage under normal conditions.

For the cluster ion beam process, the incident energy per atom is an important parameter to cause surface damage, and it can be controlled by changing the cluster size. For example, when the total incident energy is 10 keV, the X_{1000} and X_{10000} clusters carry 10 eV/atom and 1 eV/atom, respectively. However, it is not clear whether such low-damage irradiation conditions have enough capability to remove surface contamination. In this paper, we demonstrate several molecular dynamics simulations of large and slow cluster impact on a solid target with solid nano particles attached. The motion and energy transfer process were investigated and the mechanisms of surface cleaning were discussed.

2. Simulation method and model

Molecular dynamics (MD) simulations were performed in order to examine the collisional process of a large cluster impacting on a solid surface and its cleaning effect. In this simulation, three different types of atom are prepared to represent an incident Ar cluster, Si(100) target and a nano particle. The interaction between the Si atoms is described

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http://dx.doi.org/10.1016/j.surfcoat.2016.04.053
0257-8972/

by the Stillinger-Weber model in [7]. The other pairs, Ar—{Ar, Si, NP}, NP—{Si, NP}, are governed by the Lennard-Jones 12-6 model described in Table 1, in which the parameters are taken from [8,9].

Additionally, the mass of the NP atom is defined as 12 amu. From these definitions, the bulk properties of the NP are

- density: 1.5 g/cm^3
- structure: fcc
- Young's Modulus: 260 GPa.

These properties imply that the NP is hard enough so that it does not collapse as a result of the cluster impact with all conditions in this study. This fact may simplify the problem to discuss the dynamics among cluster, surface and NP. However, on the other hand, it is noted that the parameter set in Table 1 is artificially designed by the authors so it does not match with real materials. The NP consists of 10,000 NP atoms, and has a spherical structure with 3 nm radius. After the preliminary simulation of the structural optimization of the NP attached on the Si(100) surface, the binding energy between the NP and surface is calculated as about 70 eV.

The target Si(100) surface includes more than 2 million atoms and has 69.5 × 69.5 nm of surface area and 8.6 nm of depth. The atoms in the region 1 unit cell length forming the edge and bottom of the target are fixed to keep a diamond structure, and several layers inside it act as a thermal bath by Langevin dynamics to absorb excess impact energy and keep the target temperature at 300 K. Various sizes of Ar clusters from 20,000 to 300,000 were prepared as the projectiles. In this work, the total kinetic energy of projectile is fixed as 30 keV, so the incident energy per atom of projectiles varies from 1.5 eV/atom to 0.1 eV/atom. These projectiles were radiated on the target along the surface normal, aimed at several points on the target away from the NP center. The incident angle for the projectile to impact on surface was not varied.

3. Results and discussion

Firstly, the dynamics of incident cluster atoms to a planar target surface is examined. Fig. 1 shows the map particle and kinetic energy density of the argon atoms during the impact of Ar_{100000} 30 keV on a planar Si(100) target. The size of the mesh point is 0.5 nm × 0.5 nm.

When the incident cluster makes contact with the target surface, the bottom of the cluster is compressed and the particle density increases at the impact center. On the other hand, the cluster atoms around the impact point do not move anymore or penetrate the target surface, so the kinetic energy density for these atoms decreases and the kinetic energy is transferred to the edge of the contact region, where the atoms can move outside (Fig. 1(b) and (c)). Through the multiple collisions inside the cluster, the initial momentum of the incident cluster along the surface cluster is deflected to the parallel to the surface. As shown in Fig. 1(c) and (d), the most energetic part can be found at the edge of the cluster, and this area also moves outside, spreading and gradually decreasing in value. Meanwhile, the particle density map indicates that the densest area continues to reside around the impact center for long time, but the atoms in this area carry less kinetic energy.

Table 1
Potential models and parameters for MD simulations in this study.

Atomic pairs	ε [eV]	σ [nm]	Reference, note
Ar—Ar	0.01034	0.34	[7]
Ar—Si	0.005	0.375	[8]
Ar—NP	0.005	0.375	Same as Ar-Si
NP—NP	0.5	0.237	Given by authors
NP—Si	0.2	0.20951	ε is given by authors σ is same as Si—Si [6]

The trajectory and time evolution of the most energetic point are shown in Fig. 2. Fig. 2(a) shows the trajectory of the mesh point in cylindrical coordinate (z: height, r: radius) which gives the maximum kinetic energy density at each moment, whereas Fig. 2(b) represents the time and the value of the corresponding point in Fig. 2(a). From Fig. 2 it can be concluded that the highest energy density can be realized at $(t, r, z) = (10\ \text{ps}, 10.75\ \text{nm}, 0.75\ \text{nm})$ with the value of 18 eV/nm^3, as indicated by the arrow symbols. Considering the discussion in Fig. 1, it is supposed that this point is related with the radius of the incident cluster. Additionally, it is interesting that the value at the energetic point may surpass the initial kinetic energy density (11 eV/nm^3), and this effect is expected to contribute to the surface smoothing and cleaning.

As the time proceeds, the energetic point moves outside and its value also diminishes. Fig. 2(a) shows that the most energetic mesh point jumps randomly between the outside and impact center after a long time has passed, which means that the characteristic collective motion due to the cluster impact does not occur.

Fig. 3 shows the MD simulation results of the Ar cluster impacting on the Si(100) target with the attached NP. The snapshots represent the cross-sectional view, which are cut along the center line of the cluster, the target and the NP. The incident cluster consists of 100,000 atoms and is accelerated with a total of 30 keV (namely 0.3 eV/atom). For all conditions, the clusters are radiated along the surface normal of the Si targets, with a varying offset distance from the NP. The offset distance is chosen between 0 and 24 nm. Here, it is noted that the radii of the incident cluster and NP are about 9.5 nm and 3 nm, respectively. Additionally, the time transition of the position and the kinetic energy of the NP are shown in Fig. 4. Fig. 4(a) shows the trajectories of the center of mass of the NPs. The circles in the figure indicate the final position of the NPs at 77 ps, which are equivalent to the bottom snapshots in Fig. 3. On the other hand, Fig. 4(b) represents the time transition of the kinetic energy of NPs.

Figs. 3 and 4 suggest that the motion of the NP differs according to the offset distance. Identically, when the offset distance is 0, the incident cluster atoms hit the both sides of the NP symmetry. Thus, the lateral migration of NP is suppressed and the NP moves only along the surface normal. In this case, from the detailed motion shown as blue line and circle in Fig. 4, the center of mass of the NP first moves downward to reach 2.5 nm from the surface, and is then reflected and remains 2.8 nm, which corresponds that the bottom edge of the NP (with 3 nm radius) moves once to 0.5 nm and remains at 0.2 nm under the surface level, respectively. Additionally, it can be found in Fig. 3(a) that the target surface atoms around the contact point are disordered, which means that damage may be caused with a specific collisional process.

In Fig. 3(b), (c) and (d), the flow of cluster atoms becomes asymmetric and hits the NP on one side, which contributes to the NP motion in a lateral direction. When the offset distance is 6 nm (Fig. 3(b)), a part of the incident cluster atom collides with the NP directly. The NP gains large kinetic energy and moves rapidly. In this case, it is noted that the NP moves to more than 34 nm after 60 ps, which means that the NP is out of range of the target area, making it difficult to discuss the migration distance and energy profile.

For the larger offset distance (cases of Fig. 3(c) and (d)), most of the cluster atoms collide with Si target atoms first, rather than NP atoms, and are then deflected in parallel to the surface plane. Even in such case, the collective flow of deflected atoms shows the capability to apply enough kinetic energy for NP migration. The kinetic energy transfer diminishes as the offset distance increases.

From Fig. 4(b), when the offset distance is 6, 12 or 18 nm, the kinetic energy of the NP rises rapidly and then decreases gradually. This means that the NP gains a large amount of kinetic energy by the impact-like collision and loses it by friction with the target surface. Additionally, it is suggested that there seems to be a threshold energy where the NP starts to migrate and that it is related with the binding energy between the target and the NP.

Please cite this article as: T. Aoki, et al., Surf. Coat. Technol. (2016), http://dx.doi.org/10.1016/j.surfcoat.2016.04.053