Molecular Dynamics (MD) Simulation on the collision of a nano-sized particle onto another nano-sized particle adhered on a flat substrate

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ABSTRACT

Adhesion of a nano particle on a flat substrate both with and without deformation and also the behaviors of bullet and target particles after collision are simulated using the MD technique. The bullet particle, a low temperature solid Argon, is modeled by a Lennard-Jones(LJ) potential, and the target particle is modeled by a strong LJ potential. Parameters varied are the size of bullet and contaminant particles, adhesion force between the target particle and the substrate, and the velocity and collision angle of the bullet particle. Removal characteristics are different between weakly adhered and strongly adhered particles. For soft target particles high velocity at small angle favors removal. For hard particles the particle-substrate adhesion is the determining factor, and the ineffective angle is observed

Keywords: nano particle, nano particle adhesion, nano particle removal, molecular dynamics

1 INTRODUCTION

Particulate contamination seriously affects the manufacturing yield of micron or submicron scale devices, and semiconductor device feature is expected to decrease continuously, reaching 0.75μm by 2007 and 0.05μm by 2011[1]. Since the adhesion force is proportional to the size and drag force to the area of a particle, the use of drag force for cleaning becomes less efficient as the contaminant size is decreased, and it is generally agreed that these techniques work poorly for submicron particles [2,3] and other disadvantages with wet chemistry cleaning become more apparent at smaller scales [4].

Aerosol cleaning is a promising alternative to the classical cleaning methods. This technique has matured in industry for large particles in the micron range, but not for submicron or nano particles. Furthermore, the mechanism of contaminant removal by aerosol bombardment is not completely understood yet [5].

The objective of this study is to simulate the collision of a nano-sized particle with a rigid surface or a hard particle on a surface, in order to see the effect of various factors on the particle removal characteristics. The adhesion force between a particle and a flat substrate is dominated by the electrostatic force for the separation distance z_0>30nm but by the Van der Waals force for $z_0 \leq$ 30nm, only the Van der Waals interaction is considered in this study [6].

2 MOLECULAR DYNAMICS

Techniques used for simulation in this study is the standard MD algorithm in which molecules interact via a pairwise Lennard-Jones potential:$V(r)=4\varepsilon\ [(\sigma /r)^{12}-(\sigma /r)^{6}]$, characterized by energy and distance scales ε and σ, respectively. All parameters are nondimensionalized with ε, σ and molecular mass *m.* The natural time unit is then $\tau = \sigma\sqrt{m/\varepsilon}$. Common values of the parameters for Argon are σ =3.405Å, ε/k_B=120°K, and mass *m*=40amu, where k_B is the Boltzmann constant [7]. Argon particle which is usually used as the bullet(cleanser) particle was simulated with the basic LJ potential. The contaminant particle, which may be a solid particle, an organic droplet, a fiber, or a metal ion and the like was simulated in three different forms: an LJ particle with number density(ρ) 1.0 and binding energy ε=10.0, an LJ particle with ρ=1.0 and binding energy ε=30.0, or an LJ particle with ρ=2.0 and binding energy ε=30.0. The substrate is assumed a rigid solid, considering the rather low level of kinetic energy and fragility of the bullet argon particles.

3 RESULTS AND DISCUSSION

When a particle on a substrate is hit by another particle, it can be detached from the surface, move along the surface, or stay at the same spot with some degree of vibration. The head-on collision of a soft nano particle onto a hard nano particle on a surface is simulated, and the post-collision motion of the surface particle is monitored (Figure 1).

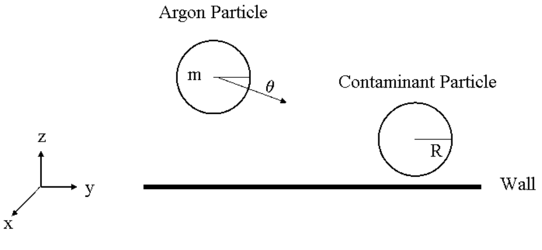


Figure 1: Schematic representation of collision of a bullet particle on a target particle on a substrate

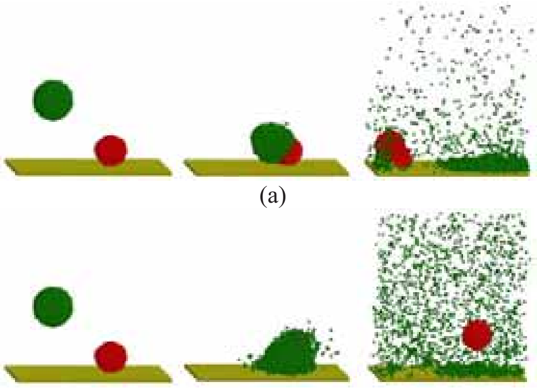
The size of the computational domain is 60σ x 31σ x 71σ, and periodic boundary conditions are imposed in the x- and y- direction. The reflective boundary condition is used for the upper z boundary, and the domain size is set a little larger in the z-direction at 71σ(24.2nm) so that the effect of the reflective boundary condition can be reduced. Time step(Δt) was 0.002~0.005τ, and the total duration of the collision process was 30000~40000Δt. Cut-off radius was 2.5σ.

Initially, the system was equilibrated for 5000Δt, and then the contaminant particle is moved toward the substrate until the distance between the particle and substrate is smaller than one lattice parameter, 1.587. Then the particle is attracted to the substrate by the attractive Van der Waals force. After the contact is settled, additional 10000Δt is allowed for equilibration, and then the argon particle is shot toward the contaminant particle. The dimensionless bullet velocity is $v(\varepsilon/m)^{1/2}$, with v=1.0 corresponding to v=158 m/s in real dimension.

3.1 LJ Solid Particle of Medium Density and Binding Energy

The LJ solid particle with ε=10.0 is a little softer than Al or Cu particle, and sample results for a shooting angle of θ=45° are shown in Figure 2. The left figures show the states after contact equilibration. When $V_x = V_z = V_0 = 2.0(\varepsilon/m)^{1/2}$ (316m/s) with total kinetic energy of $1.38\text{x}10^{-19}$J as in Figure 2(a), the argon particle is partially broken up, but the contaminant particle just slides and rolls to the +x direction, without departing from the surface. When the particle velocity is increased by 50% with total kinetic energy of $3.10\text{x}10^{-19}$J as in Figure 2(b), the argon particle becomes totally broken up, and the contaminant particle becomes detached from the surface after collision.

It seems clear that a higher kinetic energy of the bullet particle will be effective in removing the target particle, but it is not clear whether the determining factor for particle removal is the kinetic energy or the momentum. In order to clarify this question, simulations are done with twice the kinetic energy as in Figure 2(a) but with different mass and velocity combinations: (a) 2m and V_0; (b) m and $\sqrt{2}\ V_0$; (c) m/2 and $2V_0$. At the large mass and low velocity condition, particle breakup is not complete, and the contaminant particle moves with sliding and rolling but stops after some distance. At the intermediate mass and velocity condition, particle breakup is almost complete, and the contaminant particle becomes detached after moving some distance with sliding and rolling. Small amount of contaminant molecules get disintegrated. At the small mass and high velocity condition, particle breakup is complete, and the contaminant particle gets detached from the surface with a high velocity soon after collision. Of the three cases (a) has twice as high and case (b) √2 times as high a momentum as case (c). From the simulation results it can be concluded that momentum(~ mv) cannot be an indicator for particle removal, and kinetic energy(~ mv^2) is not a proper indicator, either. Particle removal seems to be determined by a new parameter with a much stronger dependence on velocity than mass like mv^3. It follows that an increased velocity is much more effective for particle removal than an increased mass.


(a)

(b)

Figire 2**:** Snapshots at 25τ, 65τ, and 100τ for LJ particle: (a) $V_x=V_z=2.0(\varepsilon/m)^{1/2}$(316m/s);(b) $V_x=V_z=3.0(\varepsilon/m)^{1/2}$ (474m/s).

The behavior of contaminant particle after collision can be classified into three modes: 1) just oscillates about a fixed point but does not move at all; 2) keeps moving with rolling and sliding but does not detach from the surface; 3) gets detached. The post-collision behaviors observed at various velocities and shooting angles are summarized in Table 1, where '×' means no motion, '□' moving without detachment, '●' detached, and '□×' stopping after sliding and rolling. It is clear from the table that a higher velocity gives a better removal, and shooting angles between 15° and 45° gives optimum performance. Particles in the cases marked □ or □× are thought to be removed in real situations due to the thermophoretic effect or by the carrier gas flow.

The argon particle breaks up when it collides with the substrate or the contaminant particle. To distinguish the effect of center-of mass kinetic energy from that of the burst induced by breakup, one artificial case is tried where the argon particle is shot normal to the substrate just ahead of the contaminant particle. In this case, the contaminant particle is affected solely by the breakup, not by the overall kinetic energy, of the bullet particle. This case is included as 'Burst only' in Table 1. When the velocity of the argon particle is high enough, the contaminant particle can be removed by the burst alone, but using the burst effect alone is not an efficient way of particle removal.

3.2 Strong Attraction : Deformation

Two different levels of deformation are considered. In weak deformation with ε=2.5, the adhesion energy is increased by a factor of 1.18, and in strong deformation with ε=5.0. the adhesion energy is increased by a factor of 2.28.

	Burst only	15°	30°	45°	60°	75°	K.E. (10^{-17}J)
V=67(m/s)	×	×	×	×	×	×	0.0313
V=223	□×	□	□	□×	□×	×	0.347
V=316	□	□	□	□	□	×	0.696
V=447	□	□	□	□	□	×	1.39
V=632	□	●	●	●	□	□	2.78
V=670	□	●	●	●	●	●	3.13
V=893	●	●	●	●	●	●	5.56

Table 1:Post-collision behavior of an LJ particle of medium binding energy at various velocities and angles of collision

Post-collision behavior of the strongly-bound target particle is much different from that of weakly-bound particle without deformation. When the bullet velocity is low and at low shooting angle, the contaminant particle starts to move but stops very soon, but at high shooting angles it does not move but gets deformed by collision. When the bullet velocity is high, the weakly-deformed particle gets detached if shot at low angle, but breaks up without moving if shot at high angle. The strongly-deformed particle breaks up, partially or fully, irrespective of the shooting angle, because the strong adhesion force prevents the contaminant particle from moving.

In generating Argon particles through supersonic expansion, the velocity of the argon particle cannot become much higher than 1000m/s. Then the results of Table 2 imply that particle bombardment may not be an effective means of removing deformed particles.

3.3 Effects of Higher Density and Binding Energy

In order to elucidate the behavior of a contaminant particle with a higher density and binding energy, simulations are tried with another artificial LJ particle with number density = 2.0, binding energy = 30.0 and σ = 0.80. Because Hamaker constant is proportional to density, the adhesion energy is increased due to the higher density, and a higher energy will be required to dislodge the target particle. Resultant behaviors of the new contaminant particle are summarized in Table 3.

When the results of Table 3 are compared with those of Table 1, it's evident that the removal efficiency for particles of higher density and binding energy is generally lower, particularly for the conditions of low velocities and small collision angles. At an intermediate collision angle of 45° the removal performance stays the same, and at higher angles of 60° and 75° removal even gets enhanced at high velocity conditions. This removal enhancement at high collisions angles and velocities can be attributed to the elastic repercussion of the elastic target particle against the hard substrate.

Weakly deformed	15°	30°	45°	60°	75°	K.E. (10^{-16}J)
V=894(m/s)	□×	□×	×	×	×	0.551
V=1117	□	□×	×	×	×	0.861
V=1340	□×	□×	×	×	×	1.24
V=1564	□	□	□	□	□	1.71
V=1787	□	●	●	□	▲	2.23
V=1974	●	●	●	▲	▲	2.72
V=2070	●	●	●	▲	▲	3.00

strongly deformed	15°	30°	45°	60°	75°	K.E. (10^{-16}J)
V=1117	×	□×	×	×	×	0.861
V=1340	□×	□×	×	×	×	1.24
V=1564	□×	□▲	□▲	×	×	1.71
V=1787	□×	□▲	□▲	△	△	2.23
V=1974	□▲	□▲	□▲	▲	▲	2.72
V=2070	●▲	▲	▲	▲	▲	3.00

Table 2: Summary of removal characteristics for weakly (ε=2.5) and strongly (ε=5.0) deformed particles. '▲' denotes particle breakup. '△' denotes partial break up

In order to discriminate the effect of the target particle hardness, a little softer target particle with number density = 2.0 and binding energy = 10.0 is simulated. Then the general behavior after collision of this softer particle is seen nearly the same as that of a harder particle with number density = 2.0 and binding energy = 30.0. It implies that the removal characteristics of hard particles are dominated by the particle-substrate adhesion energy, not by the intra-particle binding energy.

3.4 Effects of Contaminant Particle Size

As a final example of particle removal problem the case of a larger contaminant particle size is simulated. The new target particle has a diameter of 16.2(5.5nm) instead of 12.7(4.6nm), and the density and binding energy is 1.0 and 30.0, respectively. Under these conditions the number of molecules and the total adhesion energy are the same as in the previous section 3.3 for a hard target particle, but only the diameter is changed. Then even at the same kinetic energy (or linear momentum) and collision angle of the

	Burst only	15°	30°	45°	60°	75°	K.E. (10^{-17}J)
V=67m/s	×	×	×	×	×	×	0.0313
V=223	×	□×	□×	□×	×	×	0.347
V=316	□×	□	□	□	×	×	0.696
V=447	□	□	□	□	×	×	1.39
V=632	□	□	□	●	●	●	2.78
V=670	●	□	□	●	●	●	3.13
V=893	●	●	●	●	●	●	5.56

Table 3: Removal behavior for a higher binding energy and density particle. ε = 30.0, σ = 0.80 and density = 2.0. Shaded and underlined symbol denotes the removal is deteriorated and enhanced compared with Table 1, respectively.

	Burst only	15°	30°	45°	60°	75°	K.E. (10^{-17}J)
V=67m/s	×	×	×	×	×	×	0.0313
V=223	×	□×	□×	□×	×	×	0.347
V=316	□×	□	□	□	×	×	0.696
V=447	□	●	□	□	□●	×	1.39
V=632	●	●	□	●	●	●	2.78
V=670	●	●	□●	●	●	●	3.13
V=893	●	●	●	●	●	●	5.56

Table 4: Removal behavior for a large contaminant particle whose radius is 8.1(2.75nm). ε= 30.0, σ = 1.0 and density = 1.0

bullet particle, the forcing effect on the target particle may change due to the difference in radius of rotation.

The change of removal behavior with shooting velocity and angle shows some anomalous pattern at an intermediate velocity and angle (Table 4). Under these conditions, the particle keeps in motion with sliding and rolling but does not get detached. This seemingly anomalous behavior has a theoretical ground. As Wang [8] formulated for a perfectly rigid particle, a particle adhered to a solid surface comes to move, upon impaction by another particle, in any of the three modes - lift-off, sliding, and rolling -, and among the three modes the onset force for rotation is usually the smallest. However, under some conditions, there appears a range of forcing angle over which particle rotation is very difficult to induce. Then particle removal becomes greatly reduced when the forcing angle is in this range, and he found that this ineffective zone for rotation extends from 42.1° to 47.9° of forcing angle. The seemingly ineffective zone observed in this study is about 30-45°, which is quite close to the theoretical prediction.

And the difference in forcing – this study considers only the head-on collision – may be the reason for the difference in the observed ineffective zone. Such a behavior was not observed with soft particles as in Table 1, and the reason for that is not clear so far. It may be that the behavior is predicted only for a perfectly rigid particle as is assumed in the theory, or that such a behavior is expected even for a soft particle but the small number of angles studied missed the narrow zone.

4 CONCLUSIONS

Higher kinetic energy is favored in particle removal, but from the view point of removal effectiveness an increased particle velocity is more efficient than an increased particle mass. Smaller particle with higher velocity is favored than large particle with low velocity. For particles adhered without strong deformation small shooting angles facilitate removal, and optimum performance is observed for shooting angles in the range of 30-45°. For strongly bound particles as in the presence of deformation, the target particle does not get detached but gets broken-up frequently. Then particle removal by particle bombardment is not effective. The effect of bullet particle breakup should be considered when the shooting velocity is high. Ineffective zone for rotation was observed for hard and large target particles, in close agreement with classical theory. The removal characteristics of hard particles are dominated by the particle-substrate adhesion energy, not by the intra-particle binding energy.

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