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Particle-Substrate Collisions, Particle Rebound and Removal*

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Recent progress in particle capture and rebound and its effect on the adhesion force is reviewed in this paper. Particles rebound when the incident velocity is greater than a characteristic critical velocity. Lower impactation velocity particles experience elastic and plastic deformation. Recent models for particle rebound and capture are discussed and evaluated in terms of their restrictive assumptions and results. Recent experimental data of particle rebound and capture is also discussed, as is the hydrodynamic removal of captured particles. The removal of particles occurs when the applied hydrodynamic removal force overcomes the adhesion force. The effect of adhesion-induced deformation on the removal of particles is introduced and discussed.

KEY WORDS particle; adhesion force; removal; rebound; capture; dynamic adhesion; adhesion-induced deformation; polystyrene spheres; silicon substrate.

INTRODUCTION

Adhesion between small particles and solid surfaces is of interest in many technological fields. Particle impactation and capture or rebound are extremely important in aerosol collection and deposition. Particles rebound when the incident velocity is greater than a characteristic critical velocity. Lower impactation velocity particles experience elastic and plastic deformation. Particle impactation is, therefore, important in determining the contact area between the particle and the substrate and, consequently, the magnitude of the adhesion force. The question of particle capture and rebound and its effect on the adhesion force is addressed in this paper. The removal of particles from silicon substrates is very important to the semiconductor industry and other industries that require stringent control of particulate contamination. Many studies have been conducted using various methods to detach particles from surfaces. Few studies, however, have tried to quantify experimentally the removal force. In this paper, a technique for determining the removal force for PSL particles from a silicon substrate is reviewed. Particle removal requires that the adhesion force between particles and substrate must be overcome. In this study, hydrodynamic drag and lift and centrifugal

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forces are used to provide the total removal force. The removal efficiency of particles for several particle diameters is also evaluated.

THEORETICAL TREATMENT

Particle-Substrate Collisions and Rebound

Dahneke¹⁻³ studied particle bounce and presented a theoretical treatment of the particle's rebound considering the loss in kinetic and adhesion energy. He presented a rebound theory and discussed energy loss mechanisms such as plastic deformation, internal friction and surface roughness. Dahneke³ also presented a theory for determining the rate of escape of particles from a surface. The theory uses the Brownian motion to predict the statistical behavior of a large number of particles or the probable behavior of a particle. Cheng *et al.*⁴ presented a semi-empirical criterion for the inception of particle bounce. Rogers and Reed⁵ presented a theory that considered elastic and plastic deformations during large particle impact. Derjaguin⁶ introduced a formula to calculate the rebound and critical speeds (in the elastic range) based on a contact electrification model. The model results (coefficient of restitution) do not agree with experimental data at intermediate incident speeds due to bulk plastic deformation.

Recently, Tsai *et al.*⁷ introduced a new particle bounce model to calculate the coefficient of restitution. The model uses the relationship between contact deformation mechanics and contact surface energy to calculate the energy required to break the contact surface. The model takes into consideration the energy spent to deform local asperities. Some of the important assumptions the model used are: the particle is softer than the substrate, the impact occurs at a right angle to the surface, the particle and surface are electrically neutral before impact, asperities are uniformly distributed hemispheres and the contact surface energy is uniform. Using energy conservation, the relationship between the incident kinetic energy, E_{K1} the rebound kinetic energy, E_{K2} , the energy loss due to breaking of the contact surface, E_{CS} , the energy loss due to bulk plastic deformation, E_p , and local asperity deformation, E_{asp} , is given by:⁷

$$E_{k1} = E_{k2} + E_{cs} + E_p + E_{asp} \quad (1)$$

and the Coefficient of Restitution is defined as:

$$e = \frac{V_2}{V_1} = \sqrt{1 - \frac{2(E_p + E_{cs} + E_{asp})}{mV_1^2}} \quad (2)$$

where V_1 and V_2 are the incident and rebound velocities, respectively, and the critical velocity is defined, by setting V_2 to zero, as :

$$V_{cr} = \sqrt{\frac{2(E_{cs} + E_{asp})}{m}} \quad (3)$$

since the bulk plastic deformation, E_p , is negligible at the critical speed. When the particle leaves the surface with a finite contact area then the incident energy is small. The energy loss due to contact surface energy, considering elastic deformation only, is given by:⁷

$$E_{cs} = \Delta\gamma\pi\left(\frac{a_{\max}}{2^{2/3}}\right)^2 \quad (4)$$

where a_{\max} is the maximum radius of the contact circle which is given by (as defined by Johnson, Kendall and Roberts):⁸

$$a_{\max}^3 = \frac{R}{K} [F + 3\Delta\gamma\pi R + \sqrt{6\gamma\pi RF + (3\delta\gamma\pi R)^2}] \quad (5)$$

where F is the external load in dynes, $\Delta\gamma$ is the surface energy per unit area in ergs/cm², R is the radius of the sphere in cm, K is the material constant in dyne/cm² and defined as:

$$K = \frac{4}{3} \left[\frac{(1 - \nu_1^2)}{E_1} + \frac{(1 - \nu_2^2)}{E_2} \right]^2 \quad (6)$$

where ν_1 and ν_2 are the Poisson's ratios; and E_1 and E_2 are the Young's moduli in dyne/cm² for the particle and surface, respectively. However, according to Johnson and Pollock,⁹ for strictly elastic deformation, E_{cs} is independent of a_{\max} which indicates that equation (4) is wrong. Johnson and Pollock⁹ used the Johnson-Kendall-Roberts-Sperling (J-K-R-S) theory of adhesive elastic contact^{8,10} to examine the interaction between adhesion and inelastic deformation in the impact of predominantly elastic solids. They showed that even perfectly elastic impacting spheres experience some energy loss due to "snap-on" at contact and "snap-off" at separation. The J-K-R-S model predicts that the critical capture velocity varies with particle radius to the power ($-5/6$). However, for very small particles, the model becomes inappropriate and can be better described by the Derjaguin-Muller-Toporov (D-M-T) model.¹¹ Maugis¹² provided an analysis of the transition between the J-K-R-S and D-M-T models.

Equation (4)⁷ can still be used when, as a result of viscoelasticity, the effective value of $\Delta\gamma$ is much greater on peeling (separation) than during attachment (making contact). This can be done only after eliminating the $2^{2/3}$ factor in equation (4)⁷ which is a mistake. The correct equation is shown by Johnson and Pollock⁹ as:

$$E_{cs} = \Delta\gamma'\pi a_{\max}^2 \quad (7)$$

where $\Delta\gamma'$ is the peeling value of $\Delta\gamma$.

For elastic deformation, the energy loss is converted into an electric charge on the rebounding particle due to contact electrification according to Derjaguin.⁶ Tsai *et al.*⁷ estimated the charge on the rebounding particle as

$$n_p = \frac{1}{\epsilon} (2R\Delta\gamma\pi)^{1/2} \frac{a_{\max}}{2^{2/3}} \quad (8)$$

The estimated value compared well with contact charge measurements by Harper.^{13,14} When the incident energy is large, the particle or the surface may deform plastically (E_p becomes significant). This will occur if the incident velocity is larger than V_{\min} (the mean contact pressure is greater than 1.1 times the yield strength according to Johnson)¹⁵ which is defined as:⁷

$$V_{\min} = 0.983 \pi^2 \frac{Y^{5/2}}{\rho^{1/2} k^2} \quad (9)$$

where ρ is the density of the particle, and the contact surface energy is:

$$E_{cs} = \Delta\gamma\pi\left(a_p^2 + \frac{a_e^2}{2^{4/3}}\right) \quad (10)$$

where a_p is the bulk plastic deformation radius and a_e is the radius due to elastic deformation. The energy stored in the bulk plastic zone is given by

$$E_p = \int_0^a P_m \pi a^2 dh \quad (11)$$

where $h = R - (R^2 - a^2)^{1/2}$ and

$$P_m = Y C_1 \ln\left(\frac{3 K a_{\max}}{4 Y R}\right) + C_2 \quad (12)$$

The above relations depend on the assumption that; "the energy loss due to bulk plastic deformation can occur if the incident velocity is greater than a certain value." However, for small enough particles, plastic deformation can occur even at zero incident velocity according to Maugis and Pollock¹⁶ and Pollock.¹⁷

One of the most useful features of Tsai's model⁷ is the inclusion of surface roughness effects. The energy loss due to local asperity deformation, the asperities are modeled as uniformly distributed hemispheres) is given by Tsai as

$$E_{\text{asp}} = Y \left(\frac{2}{3} \pi r_{\text{asp}}^3\right) n_{\text{asp}} (\pi a_{\text{max}}^2) \quad (13)$$

where r_{asp} is the radius and n_{asp} is the total number of the yielded asperities per contact area for particle and surface, and Y is the yield strength of the softer material.

This model as proposed by Tsai *et al.*⁷ provides the coefficient of restitution and critical speed based on the energy loss models and energy conservation principle. The model is compared with the experimental data of Dahneke³ in Figures 1, 2 and 4. The shown experimental data are for the impaction of 1.27 μm PSL particles on a quartz plate. In these data, plastic deformation occurs at an incident speed of 30 m/s. The values of the parameters used in Figures 1 and 2 are $E_1 = 7.5 \times 10^{10}$ dyne/cm², $E_2 = 6.74 \times 10^{11}$ dyne/cm², $Y_1 = 0.081 E_1$, $A_1 = 7.5 \times 10^{13}$ ergs and $A_2 = 10 \times 10^{13}$ ergs,

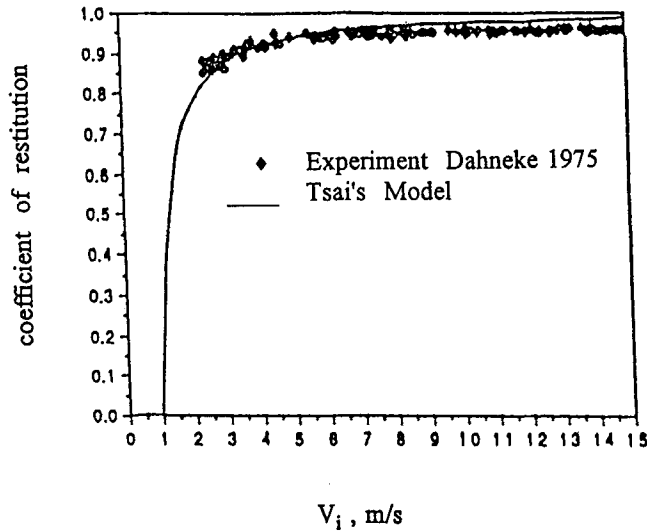


FIGURE 1 Comparison of the coefficient of restitution measured by Dahneke³ and the values predicted by Tsai's model⁷ for incident velocities of 0–15 m/s (from Reference 7). (Reprinted by permission of the publisher from C. Tsai *et al.*, *Aerosol Sci. Tech.*, Vol. 12, p. 497. Copyright 1990 by Elsevier Science Inc.)

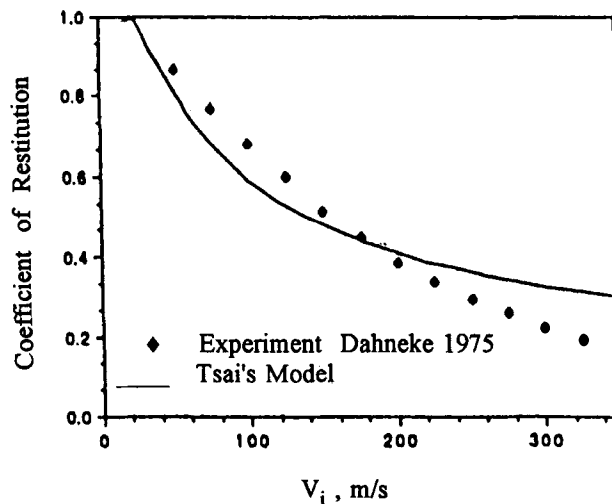


FIGURE 2 Comparison of the coefficient of restitution measured by Dahneke³ and the values predicted by Tsai's model⁷ for incident velocities of 15–350 m/s (from Reference 7). (Reprinted by permission of the publisher from C. Tsai *et al.*, *Aerosol Sci. Tech.*, Vol. 12, p. 497. Copyright 1990 by Elsevier Science Inc.)

where subscripts 1 refers to the PSL particles and 2 refers to the quartz substrate. Figure 3 shows the energy loss due to the different mechanisms considered by the Tsai model.⁷ Figure 4 shows the critical speed predicted by Tsai's model⁷ as compared with Dahneke's data.³

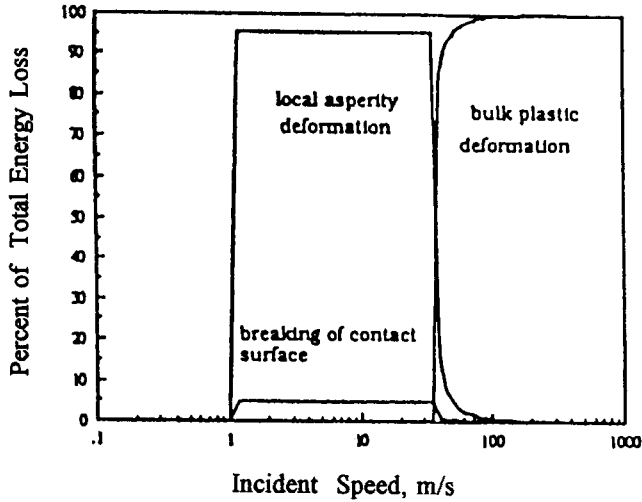


FIGURE 3 Percentage of the total energy loss due to different mechanisms (Tsai's model): $1.27 \mu\text{m}$ PSL on quartz surface, rough surface, $r_{\text{asp}} = 0.01 \mu\text{m}$, $n_{\text{asp}} = 260 \#/\mu\text{m}^2$ (from Reference 7). (Reprinted by permission of the publisher from C. Tsai *et al.*, *Aerosol Sci. Tech.*, Vol. 12, p. 497. Copyright 1990 by Elsevier Science Inc.)

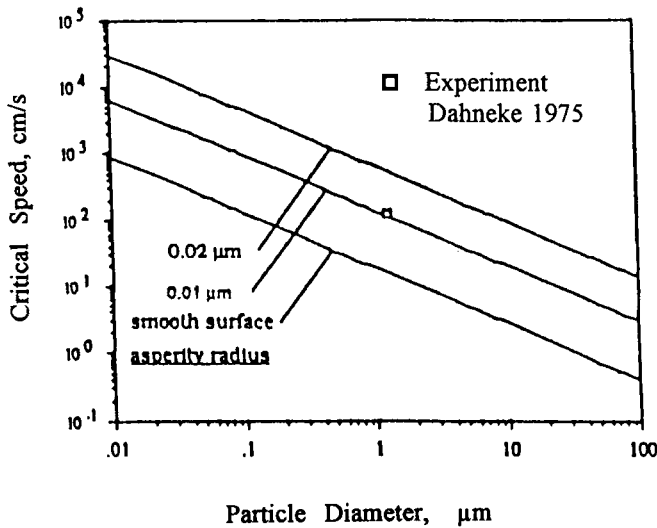


FIGURE 4 Comparison of the critical speed measured by Dahneke³ and the values predicted by Tsai's model for PSL on quartz surface (from Reference 7). (Reprinted by permission of the publisher from C. Tsai *et al.*, *Aerosol Sci. Tech.*, Vol. 12, p. 497. Copyright 1990 by Elsevier Science Inc.)

Hydrodynamic Particle Removal

Most of the physical removal of particles occurs by hydrodynamic forces (drag and lift). These forces can be applied through a boundary layer flow (of gas or liquid), or by moving the fluid by other means such as sound waves (ultrasonic cleaning). The

hydrodynamic forces acting on the particle can be evaluated by considering the flow around a spherical particle. The laminar Stokes drag force on a particle is given by Schlichting¹⁸ and Hiemenz¹⁹ as:

$$\bar{F}_D = 6\pi\mu RV \tag{14}$$

where μ , R , and V are the fluid viscosity, particle radius, and the relative velocity, respectively. In turbulent flow the drag force is given by Cleaver and Yates²⁰ as:

$$\bar{F}_D = 8\rho v^2 \left(\frac{dU}{v}\right)^2 \tag{15}$$

where d is the particle diameter and U is the shear velocity of the fluid. In addition, there is a lift force perpendicular to the wall that can be represented using the equation of Saffman:²¹

$$\bar{F}_L = 6.46\mu R^2 \left(\frac{1}{v} \frac{dV}{dX}\right)^{1/2} V \tag{16}$$

where ν is the kinematic viscosity. Saffman's equation was developed for a small sphere in an unbounded linear shear flow with no wall effects. However, the expression can be used as an approximation for a particle on a surface in a shear flow. In turbulent flow, the lift force is given by Cleaver and Yates²⁰ as:

$$\bar{F}_L = 10\rho v^2 \left(\frac{dU}{v}\right)^3 \tag{17}$$

Figure 5 shows a schematic of the drag and lift forces on the sphere. The total removal force is then given by

$$\bar{F}_L = \bar{F}_D + \bar{F}_L \tag{18}$$

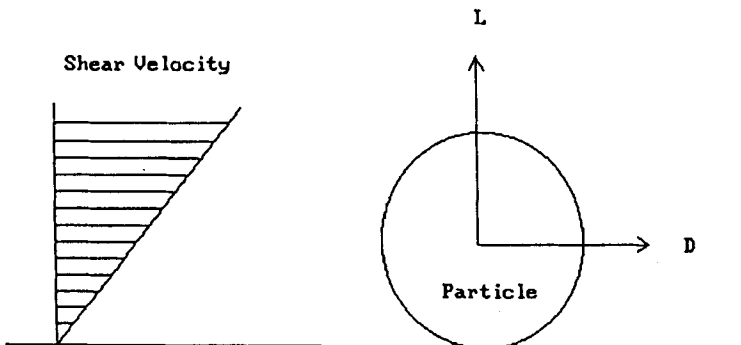


FIGURE 5 Schematic showing the drag and lift forces on a sphere.

Clearly, as particles become smaller in the submicron range, the contribution of the lift force to removal becomes small relative to that of the drag force. In order to calculate the correct drag on the sphere, the sphere is broken into small discrete cylinders. The velocity across the cylinder is arrived at using boundary layer analysis. Using this velocity and expressions for the drag coefficient, a local drag force is found. These local drag forces are then summed over the sphere, producing a total drag force.

The primary forces of adhesion for small particles on a dry surface are the van der Waals forces. These forces are a function of particle-substrate properties, the medium and the contact area between the particle and the substrate. The van der Waals force, therefore, can increase due to particle or surface deformations. The van der Waals force of attraction between an undeformed sphere and a half-space is proportional to the radius of the sphere.²² When the sphere comes into contact with the substrate, the adhesion force causes the deformation of the interface. A circular adhesion area between the sphere and the substrate is then formed (for a soft particle and a hard substrate). The total adhesion force consists of the van der Waals force acting between the adherents before deformation at the instant of first contact, F_{vdW} , and the force acting on the adhesion area due to the deformation, $F_{vdW_{deform}}$:

$$F_{vdW_{total}} = F_{vdW} + F_{vdW_{deform}} \quad (19)$$

It has been shown that deformation can significantly increase the total adhesive force.²³

EXPERIMENTAL TREATMENT

Particle-substrate Collisions and Rebound

Dahneke² measured the incident and rebounding particle velocity directly. He measured the velocity of polymer particles impacting hard surfaces in vacuum. The limitations of the experimental set-up did not allow the measurements of near-critical velocity. High-speed photography was used to determine particle trajectory near the surface by Broom,²⁴ Hiller and Loffler²⁵ and Paw U.²⁶

Broom,²⁴ using glass spheres and an unpolished metal surface, showed that the surface roughness is a major factor in particle rebound. Rogers and Reed⁵ measured the critical impact velocity directly at the surface. The measurements were used to evaluate a thin elastic-plastic model. Other techniques were used by D'Ottavio and Goren,²⁷ Aylor and Ferrandino,²⁸ and Wang and John²⁹ for measuring the high-impact velocity indirectly. The method detects the onset of particle bounce as a decrease in collection efficiency.

Recently, Wall *et al.*³⁰ measured the velocity of incident and rebounding particles within several particle diameters from the substrate surface using laser Doppler velocimetry. The range of the impaction velocity used was 1–100 m/s. The substrates included polished molybdenum and silicon, cleaved mica and a fluorocarbon polymer. The particles were ammonium fluorescein (monodispersed) that ranged in size between 2.6 and 6.9 μm .

Wall *et al.*³⁰ found that the target material affected the coefficient of restitution at low velocity (< 20 m/s). Above 40 m/s the coefficient of restitution becomes insensitive

to the target material (showing that the particles had a lower elastic limit than the substrate) as shown in Figure 6. The figure shows Wall's data fitted using Rogers and Reed adhesion theory for plastic-elastic impacts. The recovered kinetic energy in low-velocity impact is found to depend on particle size. The dependence on particle size stops at velocities near 20 m/s as shown in Figure 7. At higher velocities, half of the impact energy is lost to plastic deformation. Plastic deformation is significant even at the onset of particle bounce (critical velocity). The critical velocity dependency on particle diameter is given by a power law dependency. Experiments also show that electrostatic surface charges do not affect particle adhesion and rebound, as shown in Figure 8.

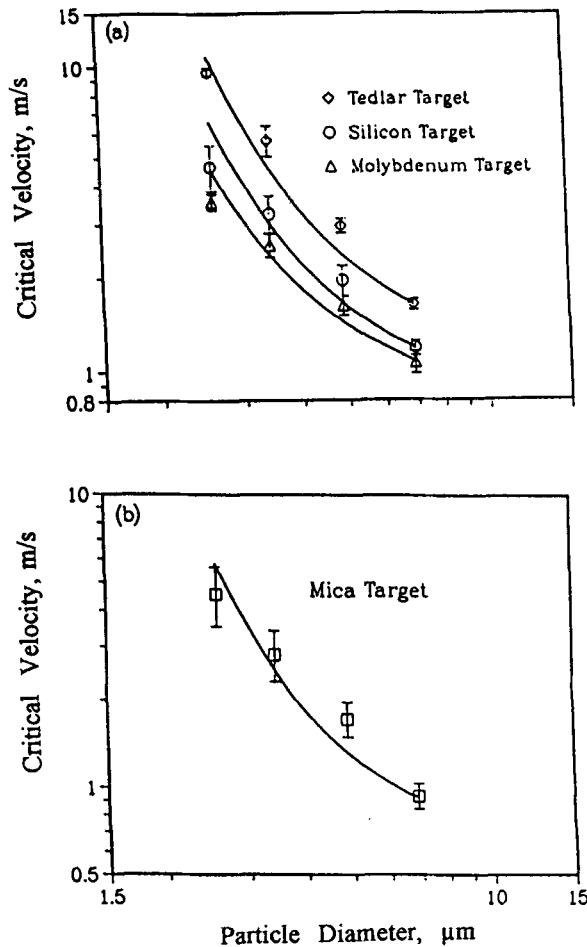


FIGURE 6 Critical velocity versus particle size for several target materials.³⁰ Lines are fits to the data using Rogers and Reed adhesion theory for elastic-plastic impacts (from Reference 30). (Reprinted by permission of the publisher from S. Wall *et al.*, *Aerosol Sci. Tech.*, Vol. 12, P. 926. Copyright 1990 by Elsevier Science Inc.)

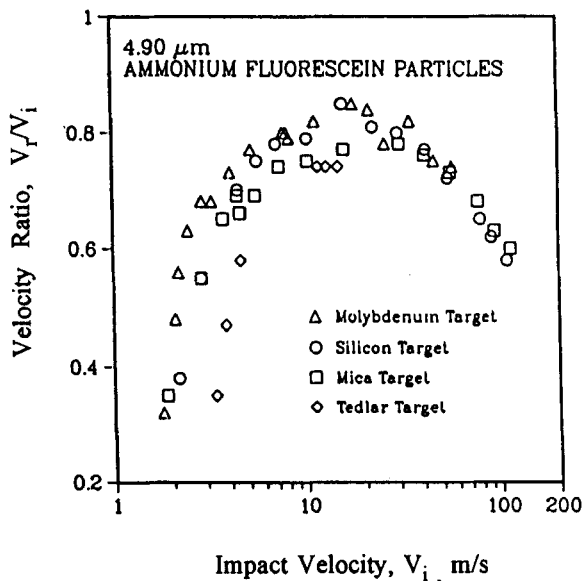


FIGURE 7 Velocity ratio, V_r/V_i , measurements over a full range of impact velocities, V_i , shows a dependence on target materials at low velocity (from Reference 30). (Reprinted by permission of the publisher from S. Wall *et al.*, *Aerosol Sci. Tech.*, Vol. 12, P. 926. Copyright 1990 by Elsevier Science Inc.)

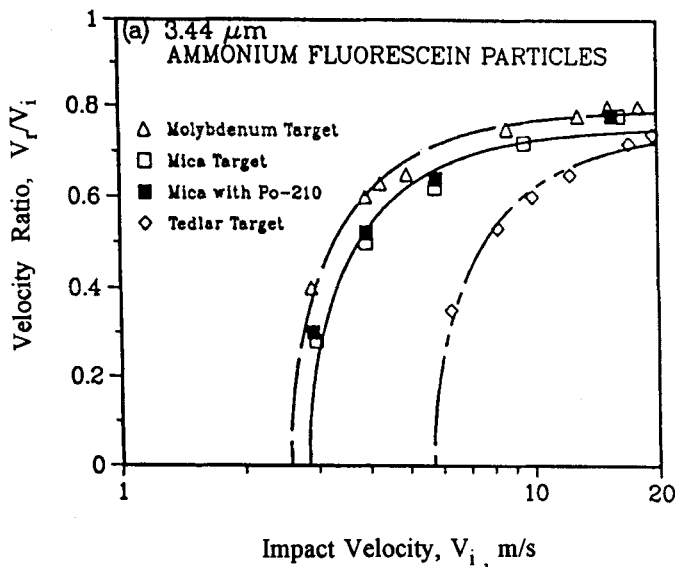


FIGURE 8 A comparison of velocity ratios for impacts on the mica target with and without surface discharge with a ^{210}Po source (from Reference 30). (Reprinted by permission of the publisher from S. Wall *et al.*, *Aerosol Sci. Tech.*, Vol. 12, P. 926. Copyright 1990 by Elsevier Science Inc.)

Hydrodynamic Particle Removal

Visser,³¹ in addition to his later work in theoretical aspects of adhesion, conducted experiments concerning particle removal. The apparatus consisted of two concentric cylinders, the outer one was fixed and the inner one was capable of rotating at a maximum of 5000 rpm. The adhering system involved 0.21 μm carbon black particles deposited on cellulose film on the inner cylinder. Visser developed a criterion for determining the adhesion force. He considered that the adhesion force is equal to the removal force when the 50% of the adhered particles were removed. In addition to the fact that the particle size distribution was not given by Visser, the high initial particle concentration (around 100 per 10^{-4} cm^2) yields (statistically) high removal percentages. Also, the 50% removal criterion used by Visser is impractical since what is needed in industrial applications is the knowledge of the removal force needed to remove 99–100% of the particles (based on the theoretical adhesion force). In other applications, the adhesion force needs to be estimated using a more precise criterion that relates the adhesion force, removal force and removal percentage as will be explained below.

A high-pressure liquid spray technique was used by Stowers³² to remove contaminant particles greater than 5 μm in diameter from large surface areas. The high-pressure and high-fluid-velocity cleaning technique removed 99.9% of 5 μm or larger particles. Spray pressures of 340 kPa, 690 kPa, and 6.9 MPa were used in the experiments.

Musselman and Yarbrough³³ used a model of viscous drag from a high-velocity spray to predict the drag force on particles at different spray nozzle pressures. They predicted the drag *versus* particle size at different nozzle pressures. They explained the difficulties in hydrodynamic drag removal due to what they called “particle hide-out” in the boundary layer. Although free stream velocities may be substantial, the local fluid velocity at the particle is small due to its proximity to the wall. Musselman and Yarbrough predicted the drag *versus* particle size at different nozzle pressures.

Kurz and Busnaina³⁴ used a rotating disk (silicon wafer rotating at 1000–10,000 rpm) to generate hydrodynamic force to remove 1 μm or larger particles. They used PSL spheres on bare silicon in deionized water as the medium. Removal rates above 90% were reported for particles larger than 2.0 μm .

Taylor *et al.*³⁵ measured the magnitude of the removal force (hydrodynamic drag and lift forces) of submicron particles on silicon substrates and correlated it with the theoretical adhesion force. They measured the particle removal percentage as a function of the fluid velocity, particle size and time. The removal percentage is used as a measure of the cleaning efficiency of the technique used. It is defined as

$$\text{Removal Percentage} = \frac{n_{\text{before}} - n_{\text{after}}}{n_{\text{before}}} \times 100 \quad (20)$$

where n is the number of particles. The results indicate that when 90% of the particles are removed, the applied removal forces is comparable with the theoretical adhesion force (van der Waals force), as shown in Figure 9.

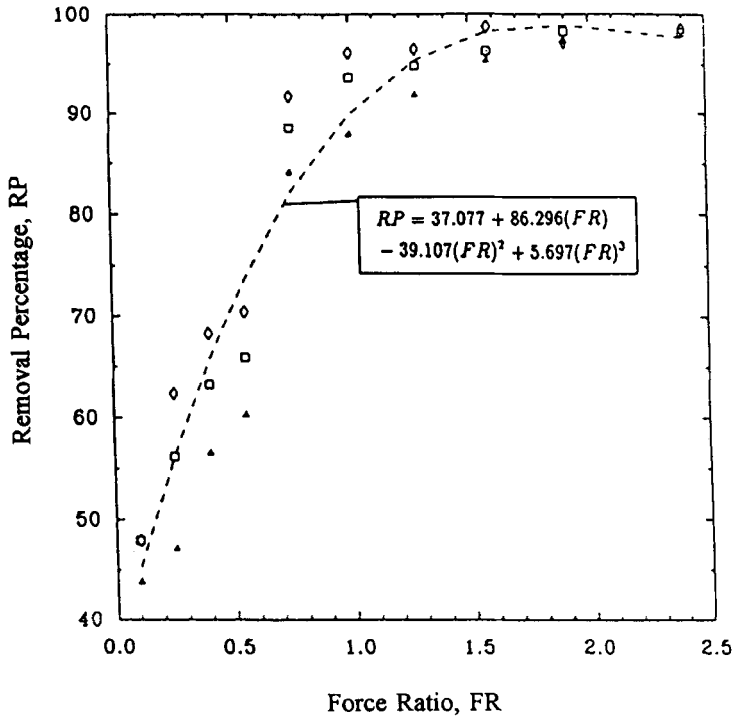


FIGURE 9 Removal percentage versus force ratio $FR = F_{remv}/F_{adh}$ (from Reference 36).

Busnaina *et al.*³⁶ determined an empirical relationship between the hydrodynamic removal force and the adhesion force for PSL submicron monodispersed particles (0.1–1.0 μm) on a silicon substrate. The relation predicts the removal percentage of particles based on the removal force to the adhesion force (van der Waals force) ratio FR (F_{remv}/F_{adh}). The relationship is given by:

$$RP = 37.1 + 86.3 FR - 39.11 FR^2 + 5.7 FR^3 \quad (21)$$

where RP is the removal percentage as shown in Figure 9.

Busnaina *et al.*³⁶ also showed that particle removal is highly dependent on the time the soft PSL particles reside on the surface before cleaning. This is due to adhesion-induced deformation of particles on the silicon substrates. A longer particle residence time significantly lowers the removal efficiency. They also showed that deformation of particles due to the forces of adhesion is a dynamic process over an extended time period up to approximately 72 hours.

The adhesion-induced deformation of particles increases the force of adhesion (through the increase of the contact area) on the particles, consequently increasing the force required for their removal. Thus, the particle removal efficiency is time dependent for a soft particle and hard substrate system or *vice versa*.

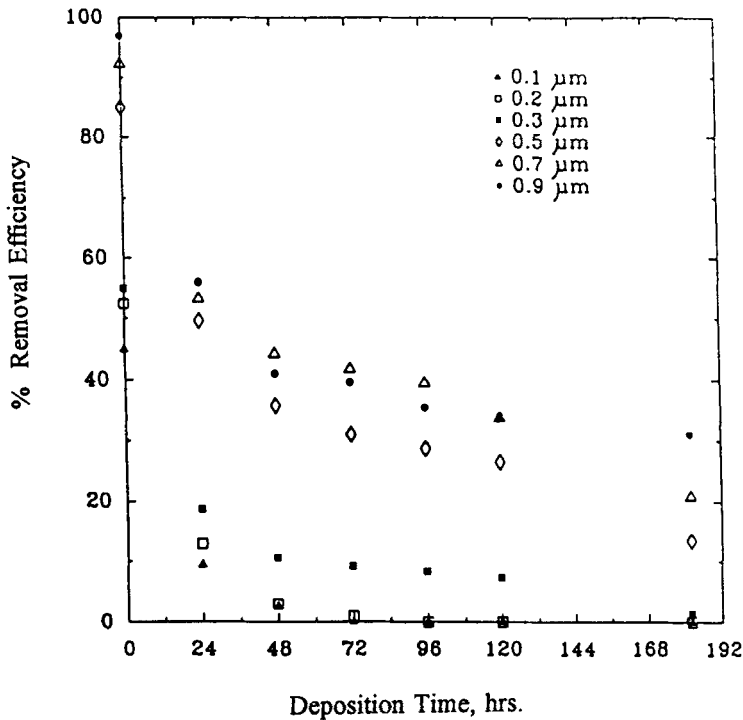


FIGURE 10 Time effect on the removal percentage for different particle diameters (from Reference 36).

CLOSURE

A concise review of recent progress in particle-substrate collisions, rebound and removal has been presented. The subject is extremely complex and has many variables. Although the presented models give reasonable results, they all use very restrictive assumptions. The experimental data of Wall *et al.* is comprehensive and provides a good understanding of the several parameters that affect particle collision and rebound. More theoretical and experimental work, however, is needed to consider the effects of charged particles, their properties, shape, different angles of incidence and humidity, etc. The particle removal review emphasized submicron particle removal using hydrodynamic drag and lift forces. Also, a criterion for experimentally determining the adhesion force for PSL particles on a silicon substrate was presented. The removal efficiency of particles for several particle diameters is also evaluated. The effect of adhesion-induced deformation on the particle removal efficiency and the removal force is also shown. Further investigation of these phenomena is needed to consider different particle and substrate material properties, fluids, and surface energies.

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