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Review:

Resuspension of wall deposits in spray dryers

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Abstract: Wall deposition occurs in spray dryers when dried or partially dried particles contact and adhere to the walls during operation, thus reducing the yield of product collected. Wall deposits also present a product contamination risk and a fire or explosion risk when spray drying products that oxidize exothermically, such as milk powder. Re-entrainment is the resuspension of spray dryer wall deposits into the main gas stream for collection as product. Literature suggests that the process for re-entrainment of particles from spray dryer wall deposits is strongly dependent on particle size and gas velocity.

Key words: Wall deposition, Spray dryers, Dried particles, Re-entrainment, Adhesive forces
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INTRODUCTION

Spray dryers are used to transform a feed solution or suspension from a liquid state into a solid particulate state in a single step process. The feed liquid is dispersed into a fine mist and passed through a hot gas to evaporate the fluid. As the fluid evaporates, only solid particles remain. These particles are then separated from the gas stream and collected.

Drying operations are required for many different applications in a variety of industries. It is estimated that there are more than 15000 spray dryers of industrial size in operation throughout the world, with approximately double that number used in pilot plants and laboratories (Masters, 1996). The simplicity of the spray drying process and the ease with which the operation can be scaled up and scaled down contribute to the suitability of spray dryers for many drying operations. These range from applications processing several hundred thousand litres of fluid per day to applications where only millilitres of fluid are dried. Industries in which spray dryers are used include the chemical industry (for example in the production of fertilizers and oxide ceramics), the food industry (in the production of milk powder and instant coffee) and the pharmaceutical and biochemical industry.

As with any processing operation, yield is an important consideration. Yield from a spray-drying operation may be defined as the ratio of the actual amount of solid powder produced to the maximum amount of powder achievable. The deposition of powder on the walls of a spray dryer reduces the yield. A low yield decreases the efficiency and profitability of an operation. There is a large variation in the yield of spray drying operations. Marquez (2005) obtained yields ranging from 10% to 83% in a Buchi B-290 spray dryer. The yield of product from a spray drying operation may be influenced by factors such as spray dryer geometry atomizer characteristics, operational conditions and final gas-particle separation efficiency (Masters, 1976). The main loss of product typically occurs due to particle deposition. Particle deposition is a common occurrence in all spray drying chambers. Dried or partially dried particles contact and adhere to the walls of the spray drying chamber, thus reducing the yield of product collected. Particles that deposit and remain on the spray dryer walls degrade as the particles undergo oxidation (and potentially browning or scorching). If these particles eventually fall into the product, product contamination occurs [(Raemy *et al*.,

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1994), cited in (Ozmen and Langrish, 2003)]. There is also a fire or explosion risk associated with the deposits of potentially combustible materials. For example, milk powder oxidizes exothermically and combustion of the milk powder deposits may cause fires or explosions in spray dryers [(Pisecky, 1997), cited in (Ozmen and Langrish, 2003)].

Many factors affect the degree of wall deposition. These include operating parameters such as airflow patterns (including swirl) (Ozmen and Langrish, 2003). The properties of the solution and subsequent particles that are spray-dried also affect the rate and extent of deposition in the spray dryer. Sticky particles, such as whey, and sugar compounds, for example, lactose, sucrose, glucose and fructose, have a high tendency to form deposits since the particles stick to surfaces and other particles (Boonyai *et al*., 2004). Emphasis should be placed on the prevention of particle deposition on surfaces rather than on subsequent removal, but the full prevention of deposition cannot be achieved in practice [(Bowling, 1988), cited in (Ziskind *et al*., 1995)]. Consequently it is necessary to consider means for particle removal, including removal by fluid flow over the surface (Ziskind *et al*., 1995).

Re-entrainment is the removal of particles from a deposit back into suspension in the gas stream. The particles may become fully suspended in the gas stream and thus available for collection in the final gas-particle separation process. To achieve re-entrainment, the magnitude of disruptive forces (primarily drag force) [(Yung, 1987), cited in (Yung *et al*., 1989)] must exceed the magnitude of adhesive forces. The adhesive forces are primarily Van der Waals forces under most conditions (Hein *et al*., 2002) but the adhesive force of liquid and/or solid bridges are significant for systems with wet or semi-dry particles (Farber *et al*., 2003).

A common occurrence in general particle-wall interaction is a series of depositions and re-entrainment as the particles, in effect, bounce along the wall. The particles have a characteristic length that describes the average distance from lift-off to touch-down on a surface. This characteristic length is generally of the order of millimetres for small particles (18~34 μm in diameter) (Braaten, 1994). Re-entrainment may be a means of reducing wall deposition after the deposit has formed on the walls of a spray dryer. The re-entrained particles would become available for collection and thus increase the yield of the spray drying operation.

This work discusses the criteria for the re-entrainment of particles from deposits and includes an overview of the attractive forces and disruptive forces acting on deposited particles. A simple view of re-entrainment suggests that, for re-entrainment to occur, the disruptive forces acting on a particle must exceed the attractive forces holding particles to a surface or to other particles. Operating parameters and environmental factors that influence re-entrainment, such as particle diameter, surface roughness, gas relative humidity and gas velocity, are introduced and discussed. Other perspectives describing aspects of the re-entrainment processes, such as fluid flow near walls with deposits and characteristic re-entrainment path lengths, are also reviewed below.

CRITERIA FOR RE-ENTRAINMENT

In order for particles to be re-entrained from deposits formed on a surface, the magnitude of disruptive forces acting on a deposited particle must exceed the magnitude of the forces holding the deposit in place.

In a wall deposit there are two different contacting regions: particle to surface contacting and particle to particle contacting. Particle to surface contacting is shown as Region 1 in Fig.1. In Region 1 the particles are held in place due to forces between the particles and the surface. Particle to particle contacting is shown as Region 2 in Fig.1. In Region 2 the particles are held in place due to inter-particle forces.

The main sources of adhesive forces in a particle

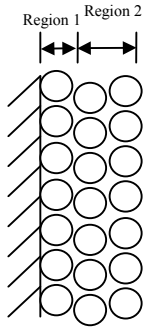


Fig.1 The two contacting regions in a wall deposit

deposit are Van der Waals forces, electrostatic forces and liquid bridge forces (Gotoh *et al*., 1997). The main disruptive forces acting on particles in a particle-gas system, such as in a spray dryer, are drag forces, lift forces and impact forces.

ADHESIVE FORCES

Van der Waals force

The Van der Waals force is a short-range electromagnetic force interacting between two molecules (or atoms). However, the force also acts between two macroscopic bodies, such as between particles and between a particle and a surface (Gotoh *et al*., 1997).

The magnitude of the Van der Waals force between two particles may be estimated from the London-Van der Waals theory which assumes that the force is acting between two symmetrical and electrically neutral molecules (or atoms). The equations for Van der Waals force, as derived from London-Van der Waals theory, are shown in Table 1.

Table 1 Van der Waals force expressions (Gotoh *et al.*, 1997)

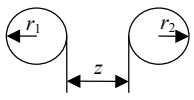
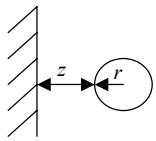
Interaction	Van der Waals force (F_{VDW})
Particle to particle: 	Without retardation effect: $-\frac{Ar_1r_2}{6(r_1+r_2)z^2}$ With retardation effect: $-\frac{Ar_1r_2}{6(r_1+r_2)}\left[\frac{r}{z^2}-\frac{b}{\lambda}\left(\frac{b}{zb+\lambda}-\frac{1}{z}\right)\right]$
Particle to surface: 	Without retardation effect: $-\frac{Ar}{6z^2}$ With retardation effect: $-\frac{A}{6}\left[\frac{r}{z^2}-\frac{b}{\lambda}\left(\frac{b}{zb+\lambda}-\frac{1}{z}\right)\right]$

Table 1 also shows the Van de Waals equations incorporating the retardation effect, where the constant, b, is equal to 11.1, λ is termed the "London characteristic wavelength" of the interaction and is often taken to be about 100 nm, and A is the Hamaker constant. The Hamaker constant is given by $A=\pi^2q_1^2/\beta_{11}$ where q_1 is the number of molecules per unit volume in the body and β_{11} is a constant which depends on the molecular (or atom) characteristics. The Hamaker constant, A, has a value of around 10^{-19} N·m and depends on the surface properties of the particle. The typical separation distance for "contact", z, is determined by Born's repulsion force and is usually taken as 0.4 nm in air (Gotoh *et al*., 1997). The retardation effect should be taken into consideration when the distance between the surfaces of two bodies (z) is greater than 100 nm.

As seen from the equations in Table 1, the magnitude of Van der Waals force is dependent on the size of the particle and the separation distance.

Liquid bridge force

If the surface of a particle has a film of mobile liquid then, at the points of contact with other particles and surfaces, 'liquid bridges' will form (Seville *et al*., 1997). Zimon (1969) states that this is considered to occur when the relative humidity of the atmosphere is greater than 65%. For a completely wettable surface of a particle, the adhesive force caused by the liquid bridge can be obtained as the sum of the capillary force: $F_L=\pi r_2^2P_L+2\pi\sigma r_2$, where r_2 is the radius of the liquid bridge (as seen in Fig.2), σ is the surface tension of the liquid, and P_L is the capillary pressure inside the liquid bridge (Gotoh *et al*., 1997).

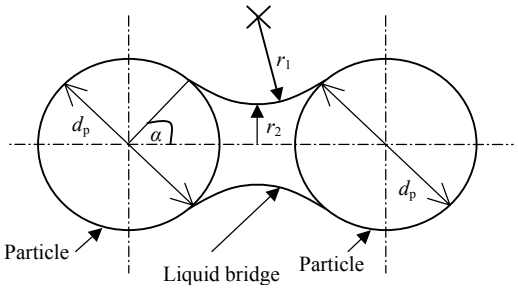


Fig.2 Liquid bridge formed between two particles (from Gotoh *et al.*, 1997)

If the cross section of the liquid bridge is approximated by a circular arc, the capillary pressure is expressed by $P_L=\sigma(1/r_1-1/r_2)$. The liquid bridge force can be calculated as follows:

$$F_L=\pi r_2^2\sigma(1/r_1-1/r_2)+2\pi\sigma r_2. \qquad (1)$$

The geometric relationships between particle diameter, d_p, r_1 and r_2 are $r_1=d_p(\sec\alpha-1)/2$, $r_2=d_p(1+$

tanα−secα)/2 (Gotoh *et al*., 1997). If r_1 is much smaller than r_2, and $\alpha\to 0$, using the above geometric relationships, the liquid bridge force for contacting spheres of the same size may be approximated by $F_L \sim \pi\sigma d_p$ [(Zimon, 1982), cited in (Gotoh *et al*., 1997)]. For a spherical particle on a plane wall, it becomes $F_L \sim 2\pi\sigma d_p$.

Alternatively, r_1 and r_2 can be correlated with a vapour pressure, P_d in the vicinity of the surface by the Kelvin equation as follows [(Carman, 1940), cited in (Gotoh *et al*., 1997)]:

$$\frac{P_d}{P_{s0}} = \exp\left[-\frac{M\sigma\cos\theta}{RT\rho_L}\left(\frac{1}{r_1}-\frac{1}{r_2}\right)\right], \tag{2}$$

where P_{s0} is the saturation vapour pressure, M is the molecular weight, R is the gas constant, T is temperature, ρ_L is the density of the liquid and θ is the contact angle.

If liquid bridges are subjected to conditions that promote drying (such as those in a spray dryer), solid bridges may form. As liquid evaporates from the bridges, it leaves behind solid bridges that impart mechanical strength to the dry granule (Tardos *et al*., 2006). During bridge formation, particle surfaces deform, melt, dissolve and/or recrystalise and a new solid bridge forms (Bika *et al*., 2005). The force required to break solid bridges is several orders of magnitude greater than the force required to separate two particles joined by a liquid bridge (from which the solid bridge was formed).

Electrostatic force

The electrostatic force in the gas phase arises from the particle-charge interaction known as the double layer force, the image-charge effect caused by charging of particles by an external field and the electrostatic contact potential difference (Gotoh *et al*., 1997).

The surface charge of a particle attracts oppositely charged ions from the surrounding fluid, forming a layer as seen in Fig.3. This layer is known as the electrical double layer (EDL). Consequently if the particle approaches a surface or particle of opposite charge to the electric double layer, an attractive force occurs.

The EDL force depends on the particle size and

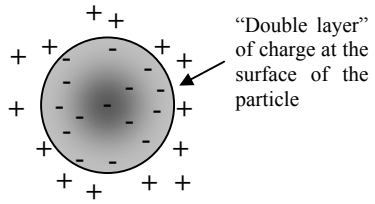


Fig.3 The electrical double layer

the separation distance, the Debye length and the zeta potentials of the particles and the walls. Hogg *et al*.(1965) derived an approximately analytical solution for EDL interaction potential between spheres, which is called the HHF formula. It can be extended to the interaction between a sphere and a plane surface by allowing the radius of one sphere to be infinite (Ye and Li, 2002).

Taking the derivative of the EDL interaction potential between a particle and a wall with respect to separation distance gives the following equation:

$$F_{EDL} = \frac{k_b T}{r_P} D_l \tau\left[\frac{\exp(-\tau X)}{1+\exp(-\tau X)} - D_a\frac{\exp(-2\tau X)}{1-\exp(-2\tau X)}\right], \tag{3}$$

where k_b is the Boltzmann constant (1.381×10^{-23} J/K), T is the absolute temperature, τ is the reduced particle radius ($r_P/k^{-1}=kr_P$, k^{-1} is the Debye length or EDL thickness). And D_l and D_a are an EDL parameter and the EDL asymmetric parameter, respectively, given by the following equations:

$$D_l = \frac{4\pi\varepsilon_r\varepsilon_0 r_P\zeta_P\zeta_S}{k_b T}, \tag{4}$$

$$D_a = \frac{(\zeta_P-\zeta_S)^2}{2\zeta_P\zeta_S}, \tag{5}$$

where ζ_P is the zeta potential of the particle, and ζ_S is the zeta potential of the surface. The relative dielectric permeability of the fluid medium is ε_r, and the permittivity of a vacuum (ε_0) is 8.85×10^{-12} C/(V·m).

Bowling (1988) [cited in (Ziskind *et al*., 1995)] suggests that, for particles less than 20 μm in diameter, double layer electrostatic forces generally predominate over image electrostatic forces. For larger particles, image force becomes more important.

DISRUPTIVE FORCES ACTING ON DEPOSITED PARTICLES

The dislodging forces acting on a solid particle adhering to a surface, when subjected to a gas flow, are the drag and lift forces. Within the spray dryer environment, wall deposits may also be subject to impacts from particles. As the liquid in the feed stream is evaporated, the particle trajectory may cause the particle to impact on the spray dryer wall and thus hit the wall deposit. The magnitude of the force is dependant on the mass and acceleration of the impacting particle.

Drag force

A passing fluid exerts a force on the body (particle or surface) in the direction of flow of the gas stream. This is known as the drag force and is the sum of all aerodynamic and hydrodynamic forces. Corn and Stein (1965) state that the drag force acting on a particle moving through a fluid (with a uniform velocity field) is as follows:

$$F_{\text{drag}} = \frac{(C_{\text{D}}\rho_{\text{f}}u^2 A_{\text{p}})}{2}, \tag{6}$$

here, C_{D} is a unique function of the particle Reynolds number (Re_{p}):

$$Re_{\text{p}} = \frac{d_{\text{p}}\rho u}{\mu}. \tag{7}$$

For creeping flow (Stokes' law region): $Re_{\text{p}}<0.3$, $C_{\text{D}}=24/Re_{\text{p}}$.

For inertial flow (intermediate region) [(Schiller and Naumann, 1933), cited in (Rhodes, 1998)], for $0.3\leq Re_{\text{p}}<500$, $C_{\text{D}} = \frac{24}{Re_{\text{p}}}(1+0.15Re_{\text{p}}^{0.687})$.

In a non-uniform velocity field, F_{drag} must be integrated over the particle projected area. Alternatively, an approximation may be obtained by calculating F_{drag} using u_{e}, the fluid velocity at the centre of the particle and influencing the entire area of particle (A_{p}) (Corn and Stein, 1965).

For deposited particles on a wall, where the particle is in creeping flow ($Re_{\text{P}}<1$) and in contact with a wall, the viscous drag force parallel to the wall (F_{drag}) is given as follows:

$$F_{\text{drag}} = 6\pi f\mu_{\text{f}}r_{\text{P}}^2\frac{\text{d}u_{\text{r}}}{\text{d}z} = 6\pi f\tau_{\text{w}}r_{\text{P}}^2, \tag{8}$$

here f is a factor compensating for wall effects, which O'Neill (1968) suggests has a value of 1.7009, and τ_{w} is the wall shear stress.

The drag force acts on a particle at a distance of 1.399 particle radii from the wall (Sharma *et al*., 1992), so the torque on a deposited particle at the point of contact (T_{contact}) is given by the following equation:

$$T_{\text{contact}} = 1.399F_{\text{drag}}r_{\text{P}} = 44.85\tau_{\text{w}}r_{\text{P}}^3. \tag{9}$$

The torque, then, is proportional to the wall shear stress (τ_{w}) multiplied by the particle radius (r_{P}) cubed.

Lift force

Lift force is the sum of all aerodynamic and hydrodynamic forces in the direction perpendicular to the gas flow. If C_{L} is the lift coefficient, the lift force (F_{lift}) is found as follows (Hamill, 1995):

$$F_{\text{lift}}=C_{\text{L}}\rho_{\text{f}}u^2A_{\text{p}}/2. \tag{10}$$

The lift force on a stationary particle deposited on a wall (F_{lift}) is given in terms of wall shear stress by the following equation (Hubbe, 1984):

$$F_{\text{lift}} = 81.2\mu_{\text{f}}^{-1/2}\nu_{\text{f}}^{-1/2}\tau_{\text{w}}^{3/2}r_{\text{P}}^3. \tag{11}$$

This equation suggests that the inertial lift force is proportional to the wall shear stress (τ_{w}) raised to the power of 1.5, multiplied by the particle radius (r_{P}) cubed. For creeping flow ($Re_{\text{P}}<1$), the effect of inertial lift on deposited particles is suggested to be negligible (Hubbe, 1984). It has also been shown that for particles with near spherical shapes, particle lift contributes negligibly to the dislodging force (Corn and Stein, 1965).

FACTORS AFFECTING THE RELATIVE MAGNITUDE OF FORCES

At system conditions that allow liquid bridges to form, and Van der Waals interactions and electro-

static charge to be present, the liquid bridge force is generally of the greatest magnitude. In the absence of liquid bridges, Van der Waals forces dominate. Electrostatic forces, such as Coulombic forces, are generally several orders of magnitude smaller than Van der Waals forces (Gotoh *et al.*, 1997).

Krupp (1967) concludes that, in general, the possible contribution of electrostatic forces does not exceed the Van der Waals force contribution. Electrostatic forces need large non-equilibrium charges on particles to be of significance when compared with Van der Waals forces. The findings of Bowling (1988) [cited in (Ziskind *et al*., 1995)] confirm this, stating that, for small particles of less than 50 μm in diameter, Van der Waals forces dominate electrostatic forces.

An analysis performed by Hays (1991) finds that the magnitude of electrostatic forces is comparable with Van der Waals forces when the surface charge density of particles exceeds 16000 $\mu C/m^2$. On this basis, Hays (1991) suggests that the importance of electrostatic forces relative to other adhesive forces is uncertain. Gotoh *et al.*(1997) concur in stating that the relative magnitude of electrostatic forces, with respect to Van der Waals force, is dependant on the surface charge density of the particles. Lee and Ayala (1985) [cited in (Gotoh *et al.*, 1997)] state that, for the charged particles, such as toner powders, the coulombic force (a type of electrostatic force) becomes important.

In processes such as spray drying, dry particles are transported through a drying chamber, a system of tubes and a cyclone. The particles may become charged by friction with the walls of the equipment [(Jonassen, 1998) cited in (Ozmen and Langrish, 2003)]. In practice, however, the effect of electrostatic forces on wall deposition in spray dryers has been found to be negligible. A preliminary study by Chen *et al*.(1994) found that either charging or earthing plates had no effect on the amount of deposit build up per unit area of a plate. A study performed by Ozmen and Langrish (2005) found that the average deposition flux did not change significantly when the spray dryer was earthed or not earthed. The lack of influence of electrostatic change of deposition in spray dryers may be attributed to the short range nature of electrostatic forces.

Effects of humidity

Using an indirect method for the measurement of adhesion force between a particle and a wall, Hein *et al*.(2002) found that, for glass spheres (20~30 and 60~70 μm), tin spheres (20~30 μm) and corn starched-fumed silica mixtures (10~20 μm) in a relative humidity of 8%~14%, the adhesive force agreed approximately with the Van der Waals force. Neither the formation of liquid bridges between contacting surfaces (and the force associated with liquid bridges) nor the influence of electrostatic adhesion were observed during their investigation. Hein *et al*.(2002) also proposed that the relatively humidity (8%~14%) contributes enough water to be adsorbed by the particle and to dissipate charges possibly accumulated on the surface. From these experimental results of Hein *et al*.(2002), it may be concluded that for particles as small as 10 μm at 8% humidity, electrostatic forces are not significant.

The experimental results of Akiyama and Tanijiri (1989) similarly suggest that the contribution of electrostatic forces at any humidity is negligible. They performed experiments using fly ash (15 μm), talc (16 μm), alumina (15~80 μm) and glass beads (16~33 μm). The magnitude of re-entrainment witnessed was unaltered between different experimental conditions within the humidity range of 0~60%.

When the relative humidity is high (greater than 65%) [(Zimon, 1982), cited in (Gotoh *et al.*, 1997)], liquid bridges may form. The magnitude of the liquid bridge force in comparison with the other adhesive forces (Van der Waals forces and possibly electrostatic forces) is high. As a result, the magnitude of the sum of all adhesive forces increases for systems of high relative humidity. Evidence of this was witnessed in experiments by Akiyama and Tanijiri (1989). They found that, for a constant air flow rate, the quantity of particles that became entrained sharply decreased as the relative humidity exceeded the 50%~70% level. The actual value at which the sharp increase in cohesive forces is witnessed depends on the particle size, the particle shape, the surface roughness and the chemical properties of the particle. The decrease in re-entrainment at high relative humidity may thus be attributed to the formation of liquid bridges. Because the liquid bridges contribute significantly to the magnitude of adhesive forces, the amount of re-entrained particles (for constant disruptive force) is decreased.

As seen in experiments by Akiyama and Tanijiri

(1989) and Hein *et al*.(2002), the relative magnitudes of inter-particle forces are highly dependant on the system conditions. Humidity contributes significantly to the relative strength of the adhesive forces. In particular, at high relative humidity (greater than 50%), liquid bridges form and increase the magnitude of adhesive forces, thus decreasing the magnitude of re-entrainment witnessed. The experiments by Akiyama and Tanijiri (1989) and Hein *et al*.(2002) suggest that the contribution of electrostatic forces to the total adhesive forces of particles to surfaces are negligible.

Effects of surface roughness

From the equations in Table 1, it is evident that the magnitude of Van der Waals forces is dependent on distance between the particles or between the particle and the surface. A consequence of this is that the magnitude of adhesive forces is dependant on surface roughness. Rough surfaces, in effect, increase the distance between the two contacting surfaces, and thus the magnitude of adhesive forces decreases with increasing surface roughness.

Krupp (1967) [cited in (Ziskind *et al*., 1995)] suggests that, in the presence of surface asperities, the calculation of Van der Waals force should be based on the radius of curvature of the surface elevation at which adherents are in contact, rather than on the full radius of the particle itself. Surface roughness also affects the magnitude of the electrostatic adhesive forces. Krupp (1967) finds that the surface roughness significantly reduces the magnitude of electrical double layer forces. Surface roughness does not affect the magnitude of image force between a surface and spherical particles, but the image force between a surface and a particle of low sphericity may be affected. As a result of these findings, Krupp (1967) suggested that electrostatic forces may be greater than Van der Waals forces for particles adhering to rough surfaces.

Experiments performed by Tabor (1977) found that, when a flat surface was roughened, the amount of adhesion decreased as the surface roughness was increased. For extremely small surface roughnesses, the adhesion hardly changes but thereafter adhesion decreases rapidly with greater surface roughness. High asperities separate the surfaces and the amount of adhesion falls to a low value. Tabor (1977) noted that, even for asperities that are quite small in comparison with the overall bulk deformation of the particle, adhesion decreased substantially. Depressions on the particle surface at the centre of the region of contact were of order 20 μm. When surface asperities were approximately 1 μm, adhesion fell to almost zero. A qualitative representation of the effect of surface roughness on adhesive forces is shown in Fig.4.

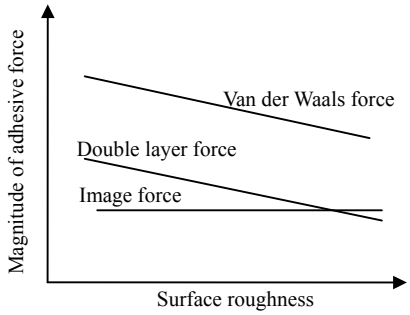


Fig.4 Qualitative representation of the effect of surface roughness on adhesive forces

Interparticle forces are not the only mechanism by which particles adhere to a surface. Rough particles may adhere to rough surfaces due to mechanical interlocking. The size of asperities on rough surfaces effects both the relative contribution of interparticle forces and mechanical interlocking effect and thus will affect the disruptive force required to separate a particle from a surface.

Effects of particle diameter and threshold re-entrainment velocity

Attractive forces, regardless of whether they are Van der Waals forces, liquid bridge forces or electrostatic forces such as the electrical double layer force, are proportional to particle diameter (d_p). As shown in Eqs.(6), (8), (10) and (11), disruptive forces (drag force and lift force) are proportional to d_p^x, where $x>1$. This theory then suggests that large particles are easier to re-entrain due to the larger ratio of disruptive to adhesive forces. This effect was found experimentally by Corn and Stein (1965). The entrainment of particles 5.3 to 42.4 μm in diameter of air velocities ranging from 30 to 117 m/s was observed. In this particle size range, Corn and Stein (1965) found that the percentage removal of particles from a

deposit increased with increasing particle size. Particles less than five microns in diameter were not dislodged from deposits, even at velocities as great as 150 m/s.

At a constant gas velocity, the quantity of small particles re-entrained is less than that of large particles re-entrained. As a result, small particles require a higher gas velocity to facilitate re-entrainment. Bagnold (1941) found that the reverse is true for particles greater than 80 μm in diameter, where larger particles have a higher threshold re-entrainment velocity. Through experiments using individual, non-adhering sand grains, larger than 80 μm in diameter, Bagnold (1941) found that the threshold re-entrainment velocity is directly proportional to the particle diameter as follows:

$$u_{*\mathrm{th}} = A\left[\left(\frac{\rho_\mathrm{p}-\rho_\mathrm{a}}{\rho_\mathrm{a}}\right)gd_\mathrm{p}\right]^{0.5}, \qquad (12)$$

here A is a constant, ρ_p and ρ_a are the particle and air densities, respectively, g is the acceleration due to gravity and d_p is the particle diameter. For $d_\mathrm{p}\geq 250$ μm, $A\cong 0.1$; for d_p 80~250 μm, $A\cong 0.2$.

Corn and Stein (1965) sought to develop a relationship between particle diameter and threshold re-entrainment velocity for particles smaller than fifty microns in diameter. The experimental results were that, for particles less than 50 μm in diameter, the threshold re-entrainment velocity was inversely proportional to particle diameter, that is, $u_{*\mathrm{th}}\alpha/d_\mathrm{p}$. Corn and Stein did not, however, quantify this relationship in more detail. Their results did, however, suggest that there is a significant change in the relationship between the threshold re-entrainment velocity and the particle diameter somewhere in the range of particle diameters between 50 and 80 μm.

The change in relationship is shown qualitatively in Fig.5. This significant change in relationship at a particle diameter between 50 and 80 μm is due to cohesion effects. When particle diameter is less than 50~80 μm, cohesion of particles can no longer be ignored, and the physical properties of particles become of great importance. As the particle size decreases, the particle sinks deeper into the viscous boundary layer at the surface. Engulfed in the viscous boundary layer, the particle is out of the range of the mainstream turbulence eddies that could dislodge it. Consequently it requires a higher bulk air stream velocity to thin this layer in order to reach the particle (Corn and Stein, 1965).

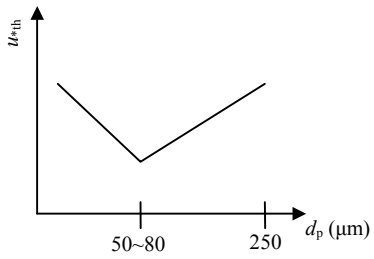


Fig.5 Qualitative diagram of relationship between particle diameter and threshold friction velocity for re-entrainment

Braaten (1994) confirmed the findings of Corn and Stein (1965) that, for particles smaller than 50~80 μm in diameter, the threshold re-entrainment velocity is inversely proportional to the particle diameter. Braaten (1994) developed an empirical formula for particles smaller than 50 μm: $u_{*\mathrm{th}}=A(\nu^2/d_\mathrm{p}^2)^{0.5}$ where A is a constant and equal to 0.26 (in CGS units), and ν is the kinematic viscosity of the gas. This relationship was established and validated for (effectively) single particles, thus cohesion forces between particles are not accounted for. A quantitative relationship for particle deposits (that is a model accounting for cohesion forces) was not developed. The particle diameter, as referred to in the literature, is the effective particle diameter. Under some conditions, particles in spray dryers may agglomerate, thus enlarging the effective size of the particles.

Both Braaten (1994) and Corn and Stein (1965) agree that threshold re-entrainment velocity is inversely proportional to particle diameter. The actual value of threshold re-entrainment velocity presented by each of the authors differs substantially. As seen in Table 2, Braaten (1994) shows that substantially lower air velocities are required to achieve entrain-

Table 2 Re-entrainment gas velocities presented by different authors

Particle type and author	u_th (m/s)	$u_{\mathrm{FS}\,50}$ (m/s)
21.2 μm spherical glass particles; Corn and Stein (1965)	30	90
20 μm spherical glass particles; Braaten (1994)	7.51	22.06

u_th is the threshold re-entrainment air velocity, $u_{\mathrm{FS}\,50}$ is the air velocity which re-entrains 50% of the particle bed

ment of particles that are 1.2 μm smaller in average diameter. There is thus some conflict regarding the gas velocity required to re-entrain particles.

RE-ENTRAINMENT OF AGGLOMERATED PARTICLES

Spray drying generally produces amorphous solids, since the speed of drying within the spray dryer is such that the complex crystalline structure does not have time to form (Goula and Adamopoulos, 2005). It has been found, however, that the particles formed in spray dryers may form amorphous agglomerates (Verdurmen *et al*., 2004). Re-entrainment of an agglomerate particle from a plane surface by an air stream will occur when the external force acting on the particle exceeds the adhesive force of the particle to the surface, or exceeds the strength of the aggregate particle (Kousaka *et al*., 1980).

As a result, consideration must be given to not only the external forces that act to dislodge the particle (or aggregate) but also the stresses that act to oppose the force holding the aggregate together as a unit. The stresses acting upon an aggregate particle subjected to a gas stream are bending stress and shearing stress (Kousaka *et al*., 1980). Kousaka *et al*.(1980) state that the bending stress imposed by the gas stream is greater in magnitude than the shearing stress imposed by the gas stream. The bending stress is induced by the drag force at small (micron) distances above the flat surface. Bending stress is composed of tensile and compressive stress. Compressive stress is unlikely to contribute to the break-up or re-entrainment of an aggregate. Thus the re-entrainment or break-up of the aggregate can be primarily attributed to the tensile stress induced by the flow of fluid past the aggregate (Kousaka *et al*., 1980).

Within the aggregate itself, the tensile strength is expected to be of the same order of magnitude as the shearing strength, since these strengths are the result of cohesive and frictional forces between particles. Thus any re-entrainment of aggregates is likely to be caused principally by bending forces on particles (Kousaka *et al*., 1980).

Due to the existence of electrical forces, the strengths of the forces holding the aggregate together are typically smaller in magnitude than the forces holding particles to a flat surface. Thus, as the aggregate particles are re-entrained, separation occurs not at the flat surface but inside the aggregate itself, as shown in Fig.6a. This statement is supported by the experimental results of Kousaka *et al*.(1980). Kousaka *et al*.(1980) observed that, following the re-entrainment of the bulk of an aggregate particle, smaller particles remained on the surface. A diagram of the pattern of aggregates observed before and after re-entrainment is shown in Fig.6b.

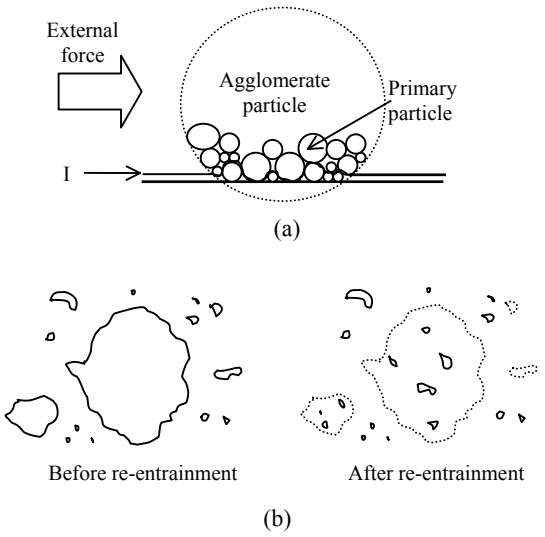


Fig.6 (a) Position where an aggregate particle is separated (represented by I); (b) Typical pattern of aggregates observed before and after re-entrainment (from Kousaka *et al*., 1980)

RE-ENTRAINMENT PROCESS

Ziskind *et al*.(1995) state that experimental measurements of resuspension indicate that particle removal from a surface is not instantaneous but takes place over a period of time. As a result, it is assumed that resuspension has a statistical origin associated with the turbulent flow character. In order to describe the random motion of a fluid near a surface, the concept of coherent boundary layer structures or "bursts" has been proposed. The boundary layer flow includes "renewal" of low-momentum fluid near the surface with fluid from a turbulent dominated region.

The concept of boundary layer structures may be represented by a laminar sublayer at the surface followed by a transition to turbulent flow at greater dis-

tances from the surface. Early models suggest that small particles are close to the wall and deeply buried in the viscous sublayer. Such models are, however, considered inaccurate and, as suggested by Corn and Stein (1965) and O'Neill (1968), lift forces are generally small. Slow and steady flows parallel to the surface are unlikely to generate enough lift to dislodge particles from the viscous sublayer as described in early models, but some experimental results showed that, even with laminar flows, dislodging is witnessed. The laminar sublayer is a rather simple idealization of conditions in the boundary layer. The sharp transition from laminar to turbulent does not exist in reality (Corn and Stein, 1965).

Bagnold (1941) noted the random movement of particles in close proximity to surfaces in turbulent flows. Particle movement, rolling along the surface and then suddenly moving away from the surface, almost at right angles to the mean flow, was observed. This prompted Bagnold (1941) to suggest that there are unsteady flow fluctuations at close proximity to walls.

Fluid motion in the viscous sublayer of a turbulent boundary layer is identifiable in terms of a definite sequence of coherent structures [(Corino and Brodkey, 1969), cited in (Reeks *et al*., 1988)]. Fluid flow at the boundary layer is a definite sequence of 'ejections' followed by downward 'sweeps' with occasional 'interactions' also known as turbulent bursts. The downward sweeps are responsible for particle deposition, while the ejections or bursts re-entrain particles from the walls. A schematic diagram of a turbulent burst is shown in Fig.7. In all these processes, transfer to the wall is more efficient than transfer away from the wall. This has been shown in theoretical studies by Cerbelli *et al*.(2001), in experimental studies by Kaftori *et al*.(1995a; 1995b) and using numerical simulations by McLaughlin (1989) and Ounis *et al*.(1993).

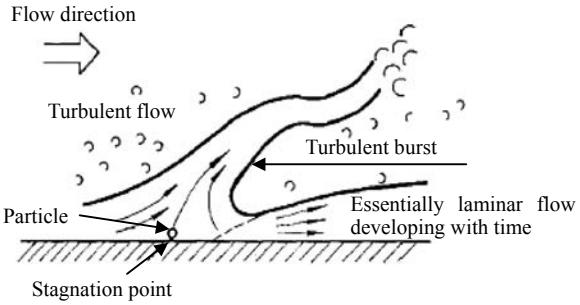


Fig.7 Schematic diagram of a turbulent burst in the wall region (from Cleaver and Yates, 1973)

It has been suggested (for example, by Corn and Stein (1965)) that the frequency of turbulent bursts is of statistical origin. An analysis of the spatial and temporal distribution of turbulent bursts was developed by Cleaver and Yates (1976). The positions of downward sweeps and bursts vary in a stochastic manner so that deposited particles may experience a burst later in time to the time when the particle was deposited. Additionally, for an initially clean surface, the number of particles per unit surface area is initially dependant on gas velocity, kinematic viscosity and time. However, as time progresses, a limiting particle concentration per unit surface area is reached and the number of particles deposited reaches a constant value (Cleaver and Yates, 1976).

Reeks *et al*.(1988) diverged slightly from the force balance models described previously by shifting the focus to energy transfer in interactions. In this model, the influence of the transfer of turbulent energy to a particle on a surface from the resuspending flow was recognized. The transfer of turbulent energy from the resuspending flow to the particle causes the particle to remain in motion, even within the viscous sublayer (described as a surface adhesive potential well). The particle detaches from the surface when it has accumulated enough vibrational energy to escape from the well (Reeks *et al*., 1988).

Characteristic re-entrainment path length

Upon criteria for re-entrainment of particles being developed, particles become re-suspended in the gas phase. It has been suggested by Braaten (1994) that particles typically do not remain in the gas phase following the initial re-entrainment period. Instead, the particles undergo a succession of depositions and entrainments. This view is supported by the suggestion of Cleaver and Yates (1976) that turbulent bursts occur in a stochastic manner.

The concept of characteristic path length, the average distance from the point at which a particle departs from a deposit on the surface into the gas phase to the point when the particle reconnects with a surface, was introduced by Bagnold (1941). A relationship relating the characteristic path length to the friction velocity for a saltating sand grain was subsequently developed by Braaten (1994). Generally,

friction velocity is defined as the square root of kinematic stress (stress per unit density): $u_* = \sqrt{\tau_w / \rho}$ and is representative of the near surface velocity value. Braaten (1994) calculated the friction velocity (u_*) as the square root of the absolute value of the velocity covariance. That is, $u_* = \sqrt{|u'w'|}$ where u' is the streamwise velocity perturbation and w' is the vertical velocity perturbation as obtained by probe measurements.

The scaling relationship for re-entrainment characteristic path length is similar to that using a Froude number (Fr). The Froude number is defined as the ratio of inertial to gravitational force and is applied to surface behaviour. The Froude number is found by $Fr=u^2/(gL)$, where u is a representative velocity, g is the acceleration due to gravity, and L is a length. A scaling relationship for re-entrainment characteristic path length was developed by Owen [1980, cited in (Braaten, 1994)] and is given by $L_* = Cu_*^2 / g$ where L_* is a characteristic re-entrainment path length, u_* is the friction velocity and C is a constant equal to 10.3 [(Owen, 1980), cited in (Braaten, 1994)]. This relationship is only applicable to saltating particles of diameter greater than 50 μm and is equal to zero when the friction velocity is less than the threshold friction velocity required for initial particle movement. An empirical relationship for determining the characteristic re-entrainment path length for particles having diameters less than 50 μm was developed by Braaten (1994). Braaten (1994) found that the experimentally-determined characteristic re-entrainment length of these particles was in fact directly proportional to both u_* and particle diameter. For particles less than 50 μm in diameter, the characteristic re-entrainment path length may be found by $L_* = Cu_*^3 d_p /(g\nu)$ where u_* is the friction velocity, ν is the kinematic viscosity and C is a constant and equal to 0.8 (in CGS units).

CONCLUSION

Particle deposition reduces the yield of spray drying operations. Many studies have been performed in an attempt to prevent the formation of wall deposits. Although these studies have resulted in effective, practical means of reducing the formation of deposits, complete prevention of deposit formation has not been achieved, and is unlikely [(Bowling, 1988), cited in (Ziskind *et al*., 1995)]. Consequently, the possibility of reducing or eliminating wall deposits by re-entraining the particles into the main gas stream is being considered. Re-entrainment of particles from spray dryer wall deposits may increase the yield of spray drying operations and (if re-entrainment is instantaneous) may reduce issues associated with contamination. Physical principles state that, to remove particles from a deposit, the magnitude of disruptive forces must exceed the magnitude of adhesive forces. Within the spray dryer environment, the primary adhesive forces that may act on particles in wall deposits are Van der Waals forces, liquid bridge forces and electrostatic forces. Several authors have studied the impact of variations in system parameters on the magnitude of cohesive forces. Parameters that affect the magnitude of adhesive forces between particles and surfaces include particle size, surface roughness and gas humidity. Literature suggests that under all operational conditions, the contribution of electrostatic forces to the sum of adhesive forces is negligible. A large factor contributing to the magnitude of adhesive forces is liquid bridges. Liquid bridges may substantially strengthen adhesive forces, thus making particles more difficult to re-entrain. Literature suggests that liquid bridges form only in environments where relative humidity is greater than 65%. At all other conditions, adhesive force is mainly due to the Van der Waals force. The primary disruptive forces acting on particle deposits in spray dryers are drag forces, lift forces and impact forces. The magnitude of drag and lift forces increases with increasing gas velocity. This has been demonstrated in research by the increase in re-entrainment of particles with increasing gas velocity. The spray dryer environment incorporates an additional factor that may assist in re-entrainment of particles. This is impact force. As the particles dry, particle trajectory may lead to impact of particles with particles deposited on the spray dryer walls. The impact force may contribute to overcoming adhesive forces, thus prompting detachment of particles from the deposit and promoting re-entrainment. Quantification of the effect of impact force on the re-entrainment of particles from spray dryer wall deposits would be useful in exploring the

potential to promote re-entrainment of particles from spray dryer wall deposits.

References

Akiyama, T., Tanijiri, Y., 1989. Criterion for re-entrainment of particles. *Powder Technology*, **57**(1):21-26. [doi:10.1016/0032-5910(89)80100-7]

Bagnold, R.A., 1941. The Physics of Blown Sand and Desert Dunes. Methuen and Co. Ltd., London, p.57-76.

Bika, D., Tardos, G.I., Panmai, S., Farber, L., Michaels, J., 2005. Strength and morphology of solid bridges in dry granules of pharmaceutical powders. *Powder Technology*, **150**(2):104-116. [doi:10.1016/j.powtec.2004.11.024]

Boonyai, P., Bhandari, B., Howes, T., 2004. Stickiness measurement techniques for food powders: A review. *Powder Technology*, **145**(1):34-46. [doi:10.1016/j.powtec.2004.04.039]

Braaten, D.A., 1994. Wind tunnel experiments of large particle reentrainment-deposition and development of large particle scaling parameters. *Aerosol Science and Technology*, **21**(2):157-169. [doi:10.1080/02786829408959705]

Cerbelli, S., Giusti, A., Solidati, A., 2001. ADE approach to predicting dispersion of heavy particles in wall bounded turbulence. *International Journal of Multiphase Flow*, **27**(11):1861-1879. [doi:10.1016/S0301-9322(01)00036-2]

Chen, X.D., Rutherford, L., Lloyd, R.J., 1994. Preliminary results of milk powder deposition at room temperature on a stainless steel surface mimicking the ceiling of a spray dryer. *Transactions of the Institute of Chemical Engineers*, **72**(C):170-175.

Cleaver, J.W., Yates, B., 1973. Mechanism of detachment of colloidal particles from a flat substrate in a turbulent flow. *Journal of Colloid and Interface Science*, **44**(3):464-474. [doi:10.1016/0021-9797(73)90323-8]

Cleaver, J.W., Yates, B., 1976. The effect of re-entrainment on particle deposition. *Chemical Engineering Science*, **31**(2):147-151. [doi:10.1016/0009-2509(76)85049-X]

Corn, M., Stein, F., 1965. Re-entrainment of particles from a plane surface. *American Industrial Hygiene Association Journal*, **26**(4):325-336.

Farber, L., Tardos, G.I., Michaels, J.N., 2003. Evolution and structure of drying material bridges of pharmaceutical excipients: studies on a microscopic slide. *Chemical Engineering Science*, **58**(19):4515-4525. [doi:10.1016/j.ces.2003.07.003]

Gotoh, K., Masuda, H., Higashitani, K. (Eds.), 1997. Powder Technology Handbook (2nd Ed.). Marcel Dekker, New York, p.133-154.

Goula, A.M., Adamopoulos, K.G., 2005. Spray drying of tomato pulp in dehumidified air: I. The effect on product recovery. *Journal of Food Engineering*, **66**(1):25-34. [doi:10.1016/j.jfoodeng.2004.02.029]

Hamill, L., 1995. Understanding Hydraulics. Macmillan Press, London, p.106.

Hays, D.A., 1991. Role of Electrostatics in Adhesion. *In*: Lee, L.H. (Ed.), Fundamentals of Adhesion. Plenum Publishing Corporation, New York, p.249-278.

Hein, K., Hucke, T., Stintz, M., Ripperger, S., 2002. Analysis of adhesion forces between particles and wall based on the vibration method. *Particle and Particle Systems Characterization*, **19**(4):269-276. [doi:10.1002/1521-4117(200208)19:4<269::AID-PPSC269>3.0.CO;2-T]

Hogg, T.W., Healy, T.W., Fuerstenau, D.W., 1965. Mutual coagulation of colloidal dispersions. *Transactions of the Faraday Society*, **62**(1):1638-1651.

Hubbe, M.A., 1984. Theory of detachment of colloidal particles from flat surfaces exposed to flow. *Colloids and Surfaces*, **12**(1):151-178. [doi:10.1016/0166-6622(84)80096-7]

Kaftori, D., Hetsroni, G., Banerjee, S., 1995a. Particle behaviour in the turbulent boundary layer. I. Motion, deposition, and entrainment. *Physics of Fluids*, **7**(5):1095-1106. [doi:10.1063/1.868551]

Kaftori, D., Hetsroni, G., Banerjee, S., 1995b. Particle behaviour in the turbulent boundary layer. II. Velocity and distribution profiles. *Physics of Fluids*, **7**(5):1107-1121. [doi:10.1063/1.868552]

Kousaka, Y., Okuyama, K., Endo, Y., 1980. Re-entrainment of small aggregate particles from a plane surface by air stream. *Journal of Chemical Engineering of Japan*, **13**(2):143-147.

Krupp, H., 1967. Particle adhesion—Theory and experiment. *Advances in Colloid and Interface Science*, **1**(2):111-239. [doi:10.1016/0001-8686(67)80004-6]

Marquez, N., 2005. An Investigation and Assessment of Process Design in Spray-Dried Milk Production. Undergraduate Thesis, School of Chemical and Biomolecular Engineering, University of Sydney, Australia.

Masters, K., 1976. Spray Drying: An Introduction to Principles, Operational Practice and Applications (2nd Ed.). George Goodwin Limited, London.

Masters, K., 1996. Deposit-free Spray Drying: Dream or Reality. Proceedings of the 10th International Drying Symposium, IDS'96. Krakow, Poland, **A**:52-60.

McLaughlin, J.B., 1989. Aerosol particle deposition in numerically simulated channel flow. *Physics of Fluids A*, **1**(7):1211-1224. [doi:10.1063/1.857344]

O'Neill, M.E., 1968. A sphere in contact with a plane wall in a slow linear shear flow. *Chemical Engineering Science*, **23**(11):1293-1298. [doi:10.1016/0009-2509(68)89039-6]

Ounis, H., Ahmadi, G., McLaughlin, J.B., 1993. Brownian particle deposition in a directly simulated turbulent channel flow. *Physics of Fluids A*, **5**(6):1427-1432. [doi:10.1063/1.858578]

Ozmen, L., Langrish, T.A.G., 2003. An experimental investigation of the wall deposition of milk powder in a pilot-scale spray dryer. *Drying Technology*, **21**(7):1253-1272. [doi:10.1081/DRT-120023179]

Ozmen, L., Langrish, T.A.G., 2005. An experimental investigation into wall deposition of milk powder in spray dryers. *Developments in Chemical Engineering and Minerals Processing*, **13**(1-2):91-108.

Reeks, M.W., Reed, J., Hall, D., 1988. On the resuspension of small particles by a turbulent flow. *Journal of Physics D: Applied Physics*, **21**(4):574-589. [doi:10.1088/0022-3727/21/4/006]

Rhodes, M., 1998. Introduction to Particle Technology. John Wiley & Sons, New York, p.2-4.

Seville, J.P.K., Tuzun, U., Clift, R., 1997. Processing of Particulate Solids. Blackie Academic and Professional, London, p.99-125.

Sharma, M.M., Chamoun, H., Sita Rama Sarma, D.S.H., Schechter, R.S., 1992. Factors controlling the hydrodynamic detachment of particles from surfaces. *Journal of Colloid and Interface Science*, **149**(1):121-134. [doi:10.1016/0021-9797(92)90398-6]

Tabor, D., 1977. Surface forces and surface interactions. *Journal of Colloid and Interface Science*, **58**(1):2-13. [doi:10.1016/0021-9797(77)90366-6]

Tardos, G.I., Farber, L., Michaels, J.N., 2006. The Morphology and Strength of Solidifying Inter-Particle Bridges in a Granule. Retrieved July 31, 2006, from http://aiche.confex.com/aiche/s06/techprogram/p33707.HTM

Verdurmen, R.E.M., Menn, P., Ritzert, J., Blei, S., Nhumaio, G.C.S., Sonne Sorensen, T., Gunsing, M., Straatsma, J., Verschueren, M., Sibeijn, M., *et al*., 2004. Simulation of agglomeration in spray drying installations: The EDECAD project. *Drying Technology*, **22**(6):1403-1461. [doi:10.1081/DRT-120038735]

Ye, C., Li, D., 2002. Electrophoretic motion of a sphere in a microchannel under the gravitational field. *Journal of Colloid and Interface Science*, **251**(2):331-338. [doi:10.1006/jcis.2002.8438]

Yung, B.P.K., Merry, H., Bott, T.R., 1989. Effects of particle-surface interactions on deposition and re-entrainment of a particulate fouling system. *Geothermics*, **18**(1-2):327-335. [doi:10.1016/0375-6505(89)90043-6]

Zimon, A.D., 1969. Adhesion of Dust and Powder. Plenum Press, New York, p.110.

Ziskind, G., Fichman, M., Gutfinger, C., 1995. Resuspension of particulates from surfaces to turbulent flows—Review and analysis. *Journal of Aerosol Science*, **26**(4):613-644. [doi:10.1016/0021-8502(94)00139-P]