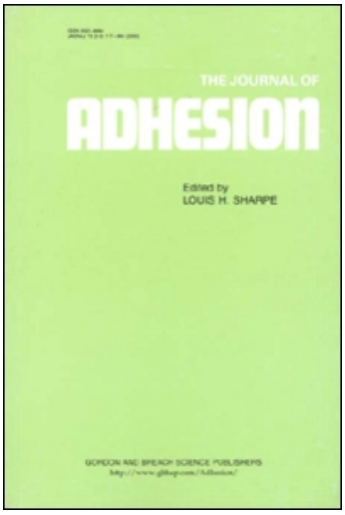

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The Adhesion of Irregularly-shaped 8 μm Diameter Particles to Substrates: The Contributions of Electrostatic and van der Waals Interactions

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The Adhesion of Irregularly-shaped 8 μm Diameter Particles to Substrates: The Contributions of Electrostatic and van der Waals Interactions

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The forces needed to remove irregularly-shaped, 8 μm diameter, polyester particles from a polyester substrate were measured using an ultracentrifuge. Measurements were also made on a second set of similar particles where nanometer-size silica clusters had been placed on their surfaces. These silica clusters acted as spacers, reducing direct contact between the particle and the substrate. It was found that the separation forces for the bare particles were consistent with predictions of the JKR theory of adhesion, but were much larger than could be accounted for from simple electrostatic interactions associated with either uniformly-charged particles or particles with localized charged patches. It was found, however, that the forces needed to effect separation decreased with increasing silica concentration. For particles with 2% by weight silica clusters on their surfaces, the separation force was only about 5% of the separation forces of the bare particles. At this concentration of silica, the estimates of the separation forces obtained from JKR theory, from the uniformly-charged model, and from the localized-charged-patch model are all about equal. The numerical estimates are consistent with the experimentally-obtained values.

Keywords: Adhesion; Irregularly-shaped polyester particles; Polyester substrate; Ultracentrifuge; Charged particles; JKR model; Uniformly-charged model; Localized-charged-patch model; Particles with sharp asperities

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INTRODUCTION

The nature of the interactions controlling the adhesion of irregularly-shaped, dry particles to substrates has been the subject of interest for many years [1, 2]. Early work by Derjaguin [3]; Bradley [4, 5] and Hamaker [6] suggest that the force needed to separate a particle from a substrate is determined by surface energy considerations that are, in turn, related to the density of states of the molecules comprising the contacting materials. Subsequently, Lifshitz [7] argued that the surface forces are due to van der Waals interactions, which arise principally from the correlation of instantaneous dipole fluctuations occurring within the contacting materials [8].

Following the work of Krupp [8], numerous theories of particle adhesion were advanced. Among these, the model proposed by Johnson, Kendall and Roberts, hereafter referred to as the JKR theory [9], seems to have gained a predominance in the literature over the others.

According to the JKR theory, for a spherical particle of radius, R, adhering to a substrate by surface forces, the separation of that particle from the substrate occurs upon the application of an externally-applied load, F_S, such that

$$F_S = -\frac{3}{2} w_A \pi R \tag{1}$$

where w_A is the thermodynamic work of adhesion and is related to the surface energies, γ_P, and γ_S, of the particle and substrate, respectively, as well as their interfacial energy, γ_{PS}, by

$$w_A = \gamma_P + \gamma_S - \gamma_{PS}. \tag{2}$$

Assuming a reasonable value of $w_A = 0.05\,\mathrm{J/m^2}$, one finds that Eq. (1) predicts a separation force of the order of 1000 nN for a particle with a radius of 5 μm. This is in reasonable agreement with reported values [10].

While it might appear at this point that particle adhesion is well understood, this is not the case. Indeed, several complicating factors arise. The first is that particles are rarely perfect spheres. Rather, at best, they tend to be spherical, but with surfaces bearing irregu-

larities or asperities. As discussed by numerous researchers including Krupp [8]; Fuller and Tabor [11]; Schaefer *et al.* [12] and Mizes [13], such asperities can significantly decrease the adhesion forces.

The effects of particle geometry are, however, more complicated than simply treating particles as spheres, perturbed by the occurrence of asperities on the surface. Rather, particles are often formed by fracturing larger materials into smaller pieces. This results in their having highly irregular shapes, appearing somewhat similar to lumps of coal. Such shape factors can have a significant impact on the effects of surface forces.

Another complicating factor is that particles often carry electrical charges and the resulting electrostatic forces can interact with neighboring substrates. The relative contributions of the electrostatic and van der Waals forces have been determined for spherical particles in several recent studies. Rimai *et al.* [14] used electrostatic detachment to measure the separation force between spherical polystyrene particles having diameters between 2 μm and 12 μm and a polyester overcoated conducting substrate and concluded that the contributions of the electrostatic force was substantially smaller than those arising from van der Waals interactions. In that study, it was assumed that the charge was uniformly distributed over the surface of the particles, which is reasonable for the highly-regular and spherical particles.

In another series of experiments, Gady *et al.* [15–18] distinguished between the electrostatic and van der Waals contributions to the force of attraction between polystyrene spheres and a variety of substrates. In these studies, they attached the particle to the cantilever of an atomic force microscope (AFM). By measuring the deflection of the cantilever and its resonance frequency as a function of the particle-to-substrate separation, they were able to determine the power-law dependence of the separation on both the force and force-gradient. As the power-laws for the electrostatic and van der Waals forces are different, they were able to ascertain that, for large separations ($>3-10$ nm), the force of attraction was dominated by a localized charge. This charge was, presumably, due to the contact between the particle and substrate that initially occurred during calibration of the displacement when the particle jumped into contact with the substrate. For smaller separations, the force of attraction was dominated by van der Waals forces. These results were confirmed by measuring the

separation distance where the particle-to-substrate jump-to-contact occurred and relating that distance to the spring constant of the cantilever. It should be noted that, according to these studies, although the separation force was dominated by van der Waals interactions, electrostatic forces also contributed. This was especially noticed when the particle and substrate were comprised of materials having vastly different triboelectric properties. In that case, the separation force was observed to increase monotonically with the number of times the particle had been allowed to contact the substrate. Finally, there are effects that arise from the combination of the irregular shape of particles with their charge. Specifically, particles become charged through triboelectric interactions. As discussed by Hays [10, 19, 29, 30], it would be quite difficult for particles to become charged in their crevasses or other recessed regions, where their surface cannot be contacted by other materials. The occurrence of the resulting nonuniform charge distribution was demonstrated by Hays during an experiment wherein 99 μm diameter particles were electrostatically detached from one electrode and deposited on another, which was parallel to, but separated from, the first by a gap. Associated with the traversal of the charged particles was a current, as would be expected. However, in addition to that current was a second current associated with the particles flipping during the transit. This was attributed by Hays to the occurrence of a dipole moment on the particle caused by a nonuniform charge distribution.

A commonly-used method of measuring the adhesion force between a particle and a substrate involves the use of an ultracentrifuge because of its ability to provide the high accelerations needed to detach micrometer-size particles from surfaces [8]. Goel and Spencer used ultracentrifugation to determine the force needed to detach toner particles, between 5 and 35 μm in diameter, from photoreceptors used in copiers [20]. Subsequently, Mastrangelo [21] used ultracentrifugation to study the effects of smooth and irregular toner particles from hard and soft photoconductors. They found that smooth particles on soft substrates were held more tightly than were irregularly-shaped particles on a hard substrate.

More recently, Lee and Jaffe [22] measured the force needed to separate irregularly-shaped, 20 μm toner particles from a photoconducting substrate using an ultracentrifuge. They found that the

measured forces were in agreement with the values predicted assuming the dominance of van der Waals interactions. However, they argued that an adhesion model *per se* could not possibly be correct for two reasons. First, they claimed that the van der Waals force model overestimates the force of attraction because the irregular shape of the particles resulted in the particles resting on asperities. Second, they also argued that the electrostatically-charged patches actually cause the electrostatic forces to be substantially larger than one would estimate assuming a spherical particle.

However, as discussed by Bowling [23] such a contact may actually increase the effect of van der Waals forces. This is because, if the irregular shape of the particle effectively causes the contact to look planar, rather than the "point" contact of a sphere on a plane, there would be an increase in the size of the van der Waals force adhering the two materials together.

As should be readily apparent, the adhesion of irregularly-shaped particles to a substrate is a complicated problem. This paper examines the roles played by both van der Waals and electrostatic interactions for irregularly-shaped particles, approximately 8 μm in diameter, that had been formed by grinding a larger block of material. This process of formation results in particles that would look somewhat akin to coal particles, rather than highly-irregular particles having sharp asperities. This problem is of both fundamental and practical importance, as the particles closely resemble electrophotographic toner in size, charge, and shape.

EXPERIMENT

The force needed to separate nominal 8.5 μm diameter ground-polyester particles from a polyester substrate was measured using a Beckman LM 70 ultracentrifuge. In addition, similar particles were coated with substantially smaller silica particles, which served as asperities, and the removal force of the coated particles were also measured.

The particles, which were similar to electrophotographic toners, were formed by grinding from a larger block of material and classifying the resulting particles to give the appropriate size. The

volume-weighted diameter of the particles was 8.6 μm, as determined using a Coulter Multisizer. Subsequently, the surfaces of the particles were coated with between 0% and 2%, by weight, of particulate silica (Aerosil R972, produced by DeGussa, Inc.). These are shown in Figures 1A, 1B and 1C, respectively. The fundamental size of the silica, as reported by DeGussa, was approximately 16 nm. However, the silica tended to form agglomerates with a diameter of approximately 60 nm, as determined from observation of field emission scanning electron micrographs. For clarity in this paper, unless otherwise mentioned, the term "particle" will be used to refer to the polyester particle, which is the subject of this investigation.

The substrate consisted of a photoconducting polyester coating, approximately 20 μm thick, on a nickelized Estar (polyethylene terphthalate, produced by Eastman Kodak) support. The particles were deposited onto the substrate in a manner analogous to that used in an electrophotographic development process. In short, the particles were mixed with larger, magnetic carrier particles, against which the

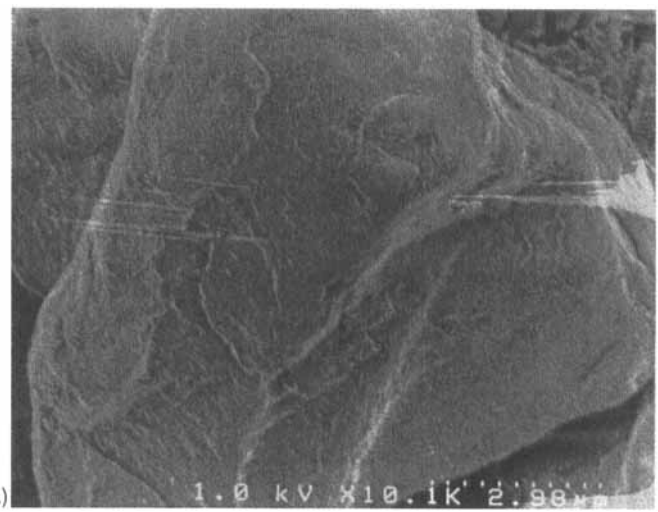
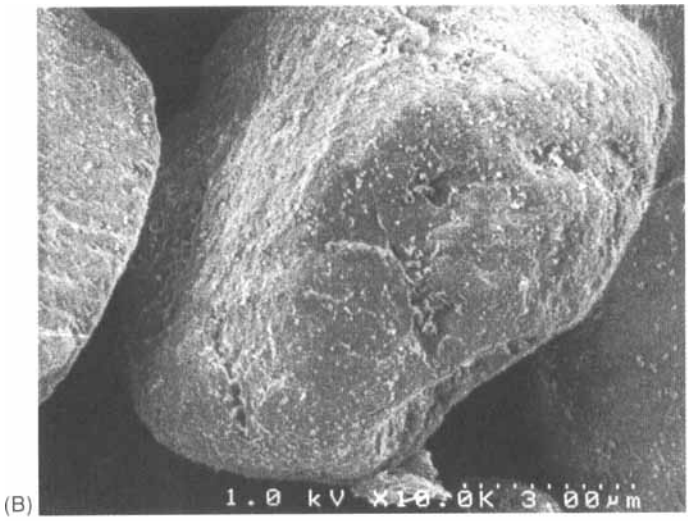


FIGURE 1 Field emission scanning electron micrographs (FESEM) of nominal 8.6 μm, irregularly-shaped, polyester particles overcoated with 0%(A), 1%(B) and 2%(C) silica (nominal cluster size approximately 60 nm).



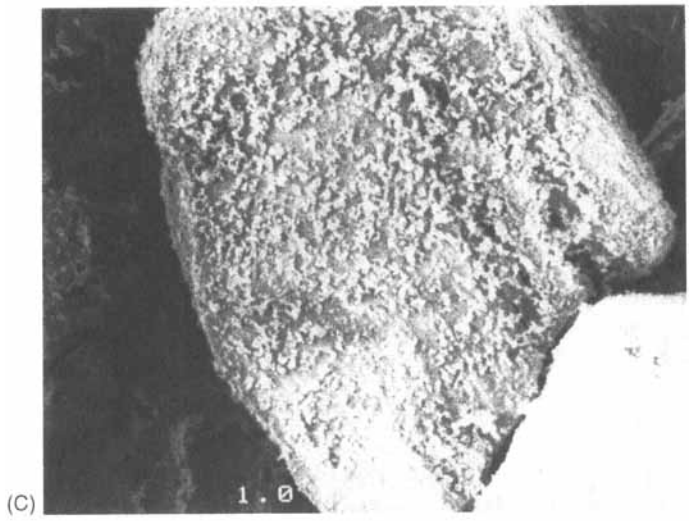


FIGURE 1 (Continued).

particles tribocharged. The charge-to-mass ratio of the particles, measured using standard techniques [24] was approximately $-37 \pm 3\,\mu C/g$ for each of the sets of particles.

Monolayers of particles were deposited onto the substrate by loading the mixture of particles and magnetic carrier particles into a toning station, similar to one used in electrophotographic development, which comprised a rotating stainless steel cylinder and a coaxial cylindrical core comprising a series of magnets. The conductive layer of the substrate was grounded and the shell biased in such a manner so as to result in the appropriate amount of particles being deposited onto the substrate. Typically, coverages of less than a monolayer were used to ensure that particle-substrate, rather than particle–particle, interactions were measured and to facilitate assessing the number of particles present using image analysis methods.

The adhesion of the particles to the substrate was determined by removing the particles in an ultracentrifuge capable of spinning at 70,000 rpm. The initial number of particles on the substrate was determined by counting the particles observed through an optical microscope, using suitable image analysis software. Next, portions of the substrate were placed in the centrifuge and spun at the desired speed. The sample was then removed and the remaining particles counted. This process was repeated for a series of increasing speeds. Centrifugation was performed in a low vacuum of approximately 10^{-2} torr (roughing pump vacuum). Initially, between 50% and 60% of the substrate was covered with particles.

RESULTS

Figure 2 shows the percent of particles removed from the substrate as a function of the mean applied force produced by different centrifuge speeds. Data for three silica concentrations, 0%, 1% and 2%, are shown. The highest force corresponds to a rotational speed of 70,000 rpm on the centrifuge. As can be seen, the general shapes of the curves gradually change for increases in silica concentration. Without silica, the percent removed is nearly linear with the mean applied force over the range investigated. There is no tendency to reach an asymptote. With 2% silica, the curve rapidly rises and then curves to approach asymptotically 100% particle removal as the mean applied force is increased. The result for the particles containing 1% silica is between the other two curves, following the 0% result

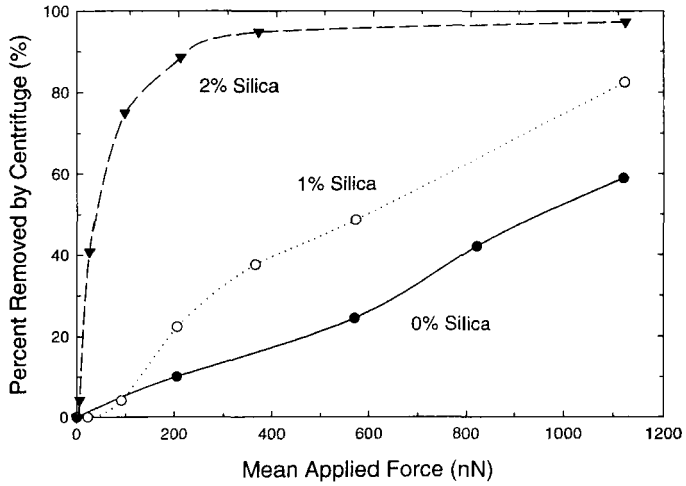


FIGURE 2 The percentage of particles removed by the ultracentrifuge as a function of removal force for three levels of silica: 0%, solid circles; 1%, open circles; and 2%, solid triangles.

TABLE I The measured and estimated forces, for various levels of silica, needed to detach the particles from the substrate

Percent silica	*Measured detachment force (nN)*	*Estimated detachment force from JKR theory (nN)*
0	970	943
1	580	507
2	39	70

initially and then rising as the centrifugation speed and, hence, the mean force is increased. Because there is a distribution in particle sizes, the larger particles would be removed first, as a qualitative view of the samples through an optical microscope suggests.

The mean applied forces reported above were calculated by assuming that the particles were spherical polyester particles with a radius of 4 μm and a mass density of 1.2 g/cm^3. The force needed to remove 50% of the particles, P_S, estimated at the 50% removal point, was determined to be 970 nN, 580 nN, and 39 nN for the 0%, 1% and 2% silica-coated particles, respectively. The measured and estimated detachment forces are listed in Table I.

DISCUSSION

As is well known, there is much debate in the literature as to whether the force of adhesion of irregularly-shaped particles to a substrate arises predominantly from surface forces such as those due to van der Waals interactions or from electrostatic forces such as those arising when charged particles polarize the substrate. For the case of a conducting substrate, this polarization would result in the particle generating its so-called image charge. The actual magnitudes of the two types of interactions depend on a number of factors including the conductivity or polarizability of the materials, as well as the size and shape of the particles. Therefore, it is unlikely that this issue will have a single solution for all possible cases. However, in-so-far as this paper addresses the adhesion of commercially-important, toner-like materials and many other commonly-found particles having similar shapes and properties, it is well worthwhile to attempt to resolve this issue for the present case. This is most readily accomplished by estimating the adhesional forces arising from both mechanisms.

Let us first assume that the uncoated particles are spheres with a radius of approximately 4 μm. The particle removal force, F_S, can be calculated from JKR theory using Eq. (1). Assuming a reasonable value of $w_A = 0.05\,J/m^2$, the particle removal force is estimated to be 943 nN. In light of the approximations made, this value is in reasonable agreement with the experimentally-obtained value of 970 nN.

As previously mentioned, it can be argued that the apparent agreement between the measured force and that calculated from a surface-force model is purely coincidental, due to asperities actually reducing the surface forces. This issue will be discussed more fully later in this paper in relation to the effect of the silica on the separation forces. However, actual measurements of the contact between particles similar to these and planar substrates suggest that the proposed asperities serving as spacers does not actually occur in the present situation. Specifically, for irregularly-shaped particles, independent sets of measurements by Eklund [25] and by Bowen *et al.* [26] both report contact areas being of the order of 10% of the projected cross-sectional area of the particles. In other words, experimental evidence suggests that intimate contact is established between particles similar to the ones used in this study and planar substrates and the particles are not resting on asperities that separate it from the

substrate. This would be in direct contradiction to the assumption that the van der Waals forces are weaker than assumed in the spherical-particle approximation due to the spacer-effect of the asperities. Therefore, it would appear that van der Waals interactions by themselves could readily account for the size of the observed separation force.

It is also necessary to estimate the size of the electrostatic contributions to the total force of adhesion. Charged particles generate fields which are approximately radial, falling off with distance. Such fields will polarize neighboring dielectric materials. When the dielectric constant of the intervening medium is less that those of the particle and substrate, the particle and substrate will attract each other [27, 28], with the force of attraction varying with the difference between the dielectric constants of the materials and intervening medium. If the particle and substrate were in intimate contact, thereby xcluding any intervening medium, the force would vary as the difference between the dielectric constants of the two materials. Unfortunately, as discussed by Jones [28], the problem of calculating either the electrostatic forces of attraction or separation between two dielectrics has not been solved. Therefore, the force of attraction between a charged dielectric particle and a conducting substrate will be approximated instead. Because of the highly polarizable nature of an electrically-conducting material, this should actually overestimate the actual force of attraction arising from the electrostatic attraction between two dielectric materials, as will be seen.

Even with the aforementioned assumption, such estimates are not simple to make, owing to effects arising from polarization and charge distributions. Details of this problem are presented elsewhere [10, 29]. However, for the sake of completeness, these issues will be discussed in brief herein.

Let us now assume, for the time being, that the irregularly-shaped particles used in this study can be approximated as a dielectric sphere of radius, R. Let us further assume that each particle has a charge, q, uniformly distributed over its surface. The electrostatic image force of attraction, F_I, between that particle and a conducting substrate is given by

$$F_I = \alpha \frac{q^2}{4\pi\varepsilon_0(2R)^2}. \tag{3}$$

When $\kappa = 4$, representing a value of the dielectric constant appropriate for a typical polymeric particle, the value of α is 1.9 [30]. In the present situation, however, the particle is not adhered to a conductor. Rather, the substrate comprises a relatively thick polymeric layer whose dielectric constant is similar to that of the particle. Moreover, as previously discussed, experimental evidence argues that the particle-to-substrate contact is intimate and relatively large. In that case, where the dielectric constants of the two contacting materials are equal and there are no air gaps, $\alpha = 1.0$. Presumably, the present case would lie between these extremes. Using the values of charge to mass reported earlier, ($-37 \pm 3\,\mu C/g$, $\rho = 1.2\,g/cm^3$), it is then calculated that F_I would be in the range of 20 to 40 nN, depending on the value chosen for α for the present particles. This value is far less than the measured force needed for detachment shown in Figure 2.

However, as is discussed by Hays [10], the charge on an irregularly-shaped particle may not be uniformly distributed over its surface. Rather, the charge is assumed to reside totally on the high spots of the particle. Accordingly, there would be a finite charge density on these spots and no charge elsewhere on the particle. In that instance, the electrostatic contribution to the force of adhesion, F_E, is related to a surface charge density, σ, and the actual area of contact between the particle and substrate, A_C, by [10]

$$F_E = \frac{\sigma^2 A_C}{2\varepsilon_0}. \tag{4}$$

Using Eq. (4) and assuming that the contact area is approximately 10% of the cross-sectional area [25, 26], one could simply solve for the charge density needed to give the measured removal force. Upon substitution, one finds that $\sigma = 1.85 \times 10^{-3}\,coul/m^2$. Using a parallel-plate-capacitor approximation, one finds that this charge density would result in an electric field of approximately $2.1 \times 10^8\,V/m$.

The problem with a particle having a charge density resulting in this high an electric field is that the air between the particle and substrate, prior to the particle coming into contact with the substrate, cannot support the resulting field. Rather, there is a maximum electric field that air can support, which is generally referred to as the Paschen limit [31, 32]. At fields above the Paschen limit, air spontaneously ionizes and becomes conductive. This limit increases from about

3×10^6 V/m for large air gaps, as would occur when the particle and substrate are widely separated, to approximately 70×10^6 V/m for separations of the order of a micrometer. Exceeding the Paschen limit would result in the electrostatic discharge of the particle. In other words, it would be very difficult, if not impossible, for a particle to maintain this high surface-charge density.

Alternatively, it is worthwhile to estimate F_E within the confines of the Paschen limit. Again, this is not simple to do, as the Paschen limit decreases with increasing air gap. However, one could estimate the field around a charged particle using either a parallel-plate-capacitor approximation when the particle is close to a surface (say a few micrometers) or a spherical particle approximation for larger distances. Moreover, if the field is less than the Paschen limit at a separation distance of approximately 10 μm in air, the particle would be able to approach the surface of the substrate without discharging at other separation distances. In other words, the field associated with the charged particle would increase with decreasing air gap at a slower rate than would the Paschen limit. With a 10 μm gap, air could support a field of approximately 3.5×10^7 V/m [32]. Moreover, the field would fall off sufficiently fast with distance so that the Paschen limit would not be exceeded. The calculated surface-charge density, under this assumption, would be approximately 3×10^{-4} coul/m^2. Therefore, F_E would be of the order of 30 nN, which is consistent with estimates of F_I. This result suggests that the charge on the particle is not highly localized but, rather, uniformly distributed. Therefore, the force of adhesion due to the presence of localized charged patches is much smaller than those contributions that can attributed to van der Waals interactions, and certainly much smaller than the experimentally-determined separation force.

As discussed in the literature [10, 29], if the separation force is determined by electrostatic interactions associated with localized charged patches, it is also necessary to discount the apparent role played by van der Waals forces. This is done by assuming that asperities on the particles separate the particles from the substrate, thereby weakening the van der Waals forces. This proposal can be readily tested by introducing asperities having a controlled size and distribution. This was accomplished in the present study by coating the particles with particulate silica.

Even so, a precise determination of the effect of the silica on the particle-detachment forces will require a detailed knowledge of how the particle and the substrate contact each other and how that contact behaves under the influence of the surface forces. This depends on a number of factors such as the size and distribution of the silica, the shape of the particles, the range of the interactions, and the compliance of the materials. Despite these complications, one may still make some order-of-magnitude estimates of the detachment forces of the silica-treated particles.

The percent of the surface coverage of the particles by the silica can be estimated by assuming both the particles and silica are spherical. For the purpose of this calculation, it was assumed that the weight fraction of the silica is 1%. As previously indicated, the primary particle size of the silica is 16 nm diameter. However, the silica tended to form clusters with an average diameter of about 60 nm. Using $\rho = 1.75\,\mathrm{g/cm^3}$ as the mass density of the silica and $\rho = 1.2\,\mathrm{g/cm^3}$ as the mass density of the particles, and knowing that the particles have a mean diameter of 8.6 μm, the fraction of the surface area of the particles covered by silica clusters is estimated to be approximately 25%. For 2% silica by weight, the area of the particles covered by silica was calculated to be 50%. These estimates are consistent with SEM micrographs of the silica-coated particles.

Again, assuming that the particles are spherical, the contact radius, a_{JKR}, estimated using JKR theory, is given by

$$a_{\mathrm{JKR}} = \left(\frac{6\pi w_A R^2}{\mathrm{E}}\right)^{1/3} \tag{5}$$

where E is the Young's modulus of polyester, approximately 3 GPa [33]. In the absence of silica, $a_{\mathrm{JKR}} = 196\,\mathrm{nm}$. This estimate is of the same magnitude as that reported by Bowen [26] for the contact of an irregularly-shaped particle with a substrate. Assuming a similar contact region exists when silica is present, it was then estimated that approximately 10 silica particles would be in contact with the substrate when the silica concentration is 2%. For the silica-coated particles, the separation force, F_S', is then approximately given by

$$F_S' = n\frac{3}{2}w_A\pi r \tag{6}$$

where $n = 10$ is the number of contacts and $r = 30\,\text{nm}$ is the radius of the silica particle clusters. Assuming that the work of adhesion for silica to photoconductor remains at $w_A = 0.05\,\text{J/m}^2$, upon substitution it is found that $F_S' \approx 70\,\text{nN}$. The experimentally-obtained value of F_S' was approximately 39 nN. In view of the approximations made, the experimentally-obtained value is in reasonable agreement with that estimated. It is interesting to note that these values are also close to the estimated contributions of the electrostatic image charges to the total force of adhesion. This suggests that, for highly irregular particles with many sharp asperities, electrostatic interactions associated with locally-charged patches can be significant factors in determining the separation forces. Indeed, observations have been made of micrometer-size nickel particles on a silicon substrate [34], where the particles do indeed show the sharp asperities discussed. The electrical conductivity of the nickel, as well as the size and shape of the asperities on the nickel particle, suggest that this particle might be a prime candidate for adhesion that is dominated by electrostatically-charged patches.

The detachment force for the particles containing 1% silica was determined by the centrifuge experiments to be approximately 580 nN, or about an order of magnitude larger than the estimated image-charge contributions. One can conclude from this result that the number of asperities present at 1% was insufficient to decrease significantly the van der Waals attraction of an irregularly-shaped particle to a substrate. Indeed, if it is simply assumed that, with a 1% coating of silica, the particles were not totally separated from the substrate so that there is some, but not total, separation of the particle from the substrate, then the separation force can be estimated by simply taking the mean values obtained from JKR theory. Accordingly, the estimated separation force for the particles with 1% silica was determined to be 507 nN, compared with the experimentally-determined value of 580 nN.

Unfortunately, owing to the rather complex nature of the particle-to-substrate contact and the possibility of statistical variations in particle charge density, it is presently not feasible to estimate quantitatively what the separation force should be under these circumstances. Indeed, with recent advances in finite element modeling software, this would be a suitable topic for future studies.

CONCLUSIONS

The force needed to remove electrically-charged, ground 8.6 μm diameter polyester particles from a polyester substrate was measured using an ultracentrifuge. The size of the measured force was in good agreement with that estimated from van der Waals interactions, but was too large to be attributed to either forces arising from a uniform or localized charge distribution. The size of the force was found to decrease with an increasing concentration of asperities, as introduced by coating the surface of the particles with silica. As the concentration of silica approaches 2%, the separation force decreased to approximately 5% of that observed for the uncoated particles. At this concentration, the estimated contributions of the van der Waals and the electrostatic forces become comparable in magnitude. This suggests that, although the localized-charged-patch model does not appear valid for the present type of particle, it may be appropriate to describe the adhesion of highly-irregular particles with sharp asperities.

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References

[1] Goel, N. S. and Spencer, P. R., *Polym. Sci. Technol.* **9B**, 763 (1975).
[2] Mastrangelo, C. J., *Photo. Sci. Engin.* **22**, 232 (1978).
[3] Derjaguin, B. V., *Kolloid Z.* **69**, 155 (1934).
[4] Bradley, R. S., *Philos. Mag.* **13**, 853 (1932).
[5] Bradley, R. S., *Trans. Faraday Soc.* **32**, 1088 (1936).
[6] Hamaker, H. C., *Physica* **4**, 1058 (1937).
[7] Lifshitz, E. M., *Soviet Phys. JEPT* **2**, 73 (1956).
[8] Krupp, H., *Advan. Colloid Interface Sci.* **1**, 111 (1967).
[9] Johnson, K. L., Kendall, K. and Roberts, A. D., *Proc. R. Soc. London Ser. A* **324**, 301 (1971).
[10] Hays, D. A., In: *Advances in Particle Adhesion*, Rimai, D. S. and Sharpe, L. H., Eds. (Gordon and Breach, Amsterdam, 1996), pp. 41–48.
[11] Fuller, K. N. G. and Tabor, D., *Proc. R. Soc. London, Ser. A* **345**, 327 (1975).
[12] Schaefer, D. M., Carpenter, M., Gady, B., Reifenberger, R., DeMejo, L. P. and Rimai, D. S., *J. Adhesion Sci. Technol.* **9**, 1049 (1995).
[13] Mizes, H. A., In: *Advances in Particle Adhesion*, Rimai, D. S. and Sharpe, L. H., Eds. (Gordon and Breach, Amsterdam, 1996), pp. 155–166.

[14] Rimai, D. S., Quesnel, D. J., DeMejo, L. P. and Regan, M. T., Submitted to *Digital Printing: Science and Technology*.
[15] Gady, B., Reifenberger, R., Rimai, D. S. and DeMejo, L. P., *Langmuir* **13**, 2533 (1997).
[16] Gady, B., Schleef, D., Reifenberger, R., DeMejo, L. P. and Rimai, D. S., *Phys. Rev. B* **53**, 8065 (1996).
[17] Gady, B., Schleef, D., Reifenberger, R. and Rimai, D. S., *J. Adhesion* **67**, 291 (1998).
[18] Gady, B., Reifenberger, R. and Rimai, D. S., *J. Appl. Phys.* **84**, 319 (1998).
[19] Hays, D. A. and Wayman, W. H., *J. Imag. Sci.* **33**, 160 (1989).
[20] Goel, N. S. and Spencer, P. R., *Polym. Sci. Technology* **9B**, 763 (1975).
[21] Mastrangelo, C. J., *Photo. Sci. Eng.* **22**, 232 (1978).
[22] Lee, M. H. and Jaffe, A. B., In: *Particles on Surfaces 1: Detection, Adhesion, and Removal*, Mittal, K. L., Ed. (Plenum, New York City, 1988), pp. 169–178.
[23] Bowling, R. A., In: *Particles on Surfaces 1: Detection, Adhesion, and Removal*, Mittal, K. L., Ed. (Plenum, New York City, 1988), pp. 129–142.
[24] Maher, J. C., *IS&T's Tenth International Congress on Advances in Non-Impact Printing Technologies*, IS and T, Springfield, VA, USA, 1994, pp. 156–159.
[25] Eklund, E. A., Wayman, W. H., Brillson, L. J. and Hays, D. A., In: *IS&T's Tenth International Congress on Advances in Non-Impact Printing Technologies*, IS&T, Springfield, VA, USA, 1994, pp. 142–146.
[26] Bowen, R. C., DeMejo, L. P. and Rimai, D. S., *J. Adhesion* **51**, 191 (1995).
[27] Smythe, W. R., *Static and Dynamic Electricity*, 3rd edn. (Hemisphere Publishing Company, New York, 1989).
[28] Jones, T. B., *Electromechanics of Particles* (Cambridge University Press, Cambridge, 1995).
[29] Hays, D. A., In: *Fundamentals of Adhesion and Interfaces*, Rimai, D. S., DeMejo, L. P. and Mittal, K. L., Eds. (VSP, Utrecht, 1995), pp. 61–72.
[30] Hays, D. A., In: *Particles on Surfaces 1: Detection, Adhesion and Removal*, Mittal, K. L., Ed. (Plenum Press, New York, 1988), pp. 351–360.
[31] Paschen, F., *Wied. Ann.* **37**, 69 (1889).
[32] Cobine, J. D., *Gaseous Conductors* (Dover Publications, New York, 1957).
[33] van Krevelen, D. W., *Properties of Polymers* (Elsevier, Amsterdam, 1976).
[34] DeMejo, L. P., Rimai, D. S. and Bowen, R. C., *J. Adhesion, Sci. Technol.* **2**, 331 (1988).