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Deformation and Non-uniform Charging of Toner Particles: Coupling of Electrostatic and Dispersive Adhesion Forces

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Abstract. *Both electrostatic and dispersive (van der Waals) forces contribute to particle adhesion, which has a significant effect on toner transfer in the electrophotographic process. Several approaches to adhesion measurements have yielded a large range of results for a variety of particle and environmental conditions. We present adhesion measurements taken in different environments using the metered air pulse method. They yield significantly different removal forces as a function of temperature for the same average particle charge. Particle deformation due to a combination of changes in particle stiffness with temperature and compressive electrostatic forces can predict the resulting adhesion increase. The morphology change is one of several factors which can contribute to the measured adhesion, which is significantly higher than values obtained by considering only the charged particle monopole and its image. Additionally, non-uniform charging in controlled adhesion experiments provides further muddling between the electrostatic and dispersive forces. This result is due to the electrostatic force having a component which is independent of the nominal charge under certain conditions. We find that the adhesion forces can be fully cubic with respect to the average particle charge, and that the components of the adhesion force may be much more difficult to decouple than previously thought. © 2013 Society for Imaging Science and Technology.*
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INTRODUCTION

Toner adhesion is a significant issue in electrophotography which affects development, transfer, and cleaning.[1] Discrepancies between predictions and measurements have ignited a debate over toner particle adhesion.[2] A central controversy in the debate relates to the relative contributions of long-range electrostatic or short-range dispersion forces. Understanding the dominant mechanism of adhesion would significantly benefit the electrophotographic printing system and also have implications for other industries using powders.

Several methods have been applied to measure microparticle adhesion to a flat substrate. Ultracentrifuge,[3–7] electrostatic removal,[7–9] microcantilever,[7,10–12] and blow-off devices[4,13,15] have all been used. Each method has yielded information about adhesion for particles ranging from 6 to 20 μm in diameter. In each case, the measured average adhesion value for particles with minimal contact area was on the order of a few tens to a few hundred nanonewtons.[3,5,6,12,13]

In this article, we present the results of experimental and theoretical studies that reveal macroscopic coupling between electrostatic and dispersive forces, which is due to increased particle sagging while under electrostatic load. The measurements of adhesion presented herein result from the use of the metered air pulse method of toner removal in different environmental conditions.[13,14] This blow-off method is used to quantify the distribution of adhesion forces acting on a halftone image sample of charged toner developed on a flat transfer belt by an electrophotographic process. It produces average adhesion measurements comparable to those of other published techniques, but is more controllable. The theoretical method applied is an analytical field model of electrostatic forces which takes into account multiple particle interactions and the substrate. The model includes independent assignment of the toner particle charge, the dielectric constant, and a non-uniform charge distribution. The calculated adhesion force has been shown to be nearly an order of magnitude higher than that from the simple Coulombic attraction model.[16] The implications of the results presented herein are that the adhesion force has an additional term proportional to the cube of the average particle charge, and that the components of adhesion force may be much more difficult to decouple than previously thought.

ELECTROSTATIC AND DISPERSIVE FORCES

The controversy surrounding toner adhesion has centered on the assumption that there are two key mechanisms contributing to toner adhesion to a substrate. These two forces are long-range electrostatic attraction and close-range dispersion forces; the latter are also known as van der Waals forces, London–van der Waals forces, or close-range dipole forces.[3]

The simplest model of electrostatic forces is Coulombic attraction. The attraction force between two charged particles at a distance is known to be a function of the charges on the two particles and the distance between them. Known as Coulomb's law or Coulomb's inverse square law, it suggests

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that at a reasonable distance the force between two particles not on a substrate would be

$$F = \frac{Q_1 Q_2}{4\pi\epsilon_0 r^2}, \tag{1}$$

where Q_1 and Q_2 are the total net charge on the two particles, r is the separation distance, and ϵ_0 is the permittivity of vacuum. For a particle with radius R, unit dielectric constant, and total charge Q resting on a conductive substrate, the electrostatic image force can be modeled as Coulombic attraction between the charge and its image:

$$F = \frac{Q^2}{16\pi\epsilon_0 R^2}. \tag{2}$$

Coulomb's law defines these electrostatic distance forces to be a function of the square of the charge, so it would seem that any term in an adhesion equation resulting from these forces should also be a function of the square of the charge.

When the distance between adjacent molecules of a particle and the substrate is on the order of a few nanometers, intermolecular forces greatly enhance adhesion. They are thought to be a result of atomic instantaneous dipole interactions of the materials involved and are a function of the material properties and the contact area. For a spherical particle, the van der Waals force is modeled as

$$F_{van\ der\ Waals} = \frac{3}{2} W_a \pi R_c, \tag{3}$$

where W_a is the work of adhesion and R_c is the contact radius. Given a printer system with a constant toner, environment, speed, and field parameters, the adhesion of toner to a substrate like a transfer belt is expected to be a function of electrostatic and dispersive forces.

$$F_a = F_{van\ der\ Waals} + F_{electrostatic} = A + Bq^2. \tag{4}$$

Kemp and Whitney analytically expanded the electrostatic field forces from multiple particles and demonstrated that the coefficient B should be on the order of 1 nN/fC^2 for a single uniformly charged dielectric particle resting on an image plane.[17,18] As has been reported in the literature, the actual values of F_a measured are about an order of magnitude higher than theoretically predicted by an electrostatic attraction model consisting only of the particle monopole and its image. The predicted electrostatic adhesion force is increased by nearly 10× by including particle dielectric polarization, multiple particle interactions, and non-uniform charge.[16]

In a two-transfer system, it is possible to change the charge on the toner independently of other variables by recharging under additional first transfer nips.[1] Figure 1 shows a graph of toner adhesion as a function of toner charge that was created in this way. The toners used in this study are chemically processed toner of nearly spherical shape with average diameter 5–8 μm. The average toner charge is calculated from the measured charge per mass, printed mass per area, and toner diameter. The substrates used herein

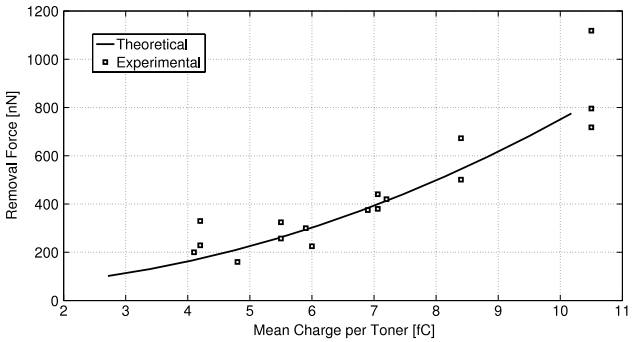

Figure 1. Median toner removal force as a function of average per toner charge. The removal force is for 50% toner removal at 60°F and 8% relative humidity. The theoretical curve represents a two-term prediction based on electrostatic and van der Waals attractions from Eq. (4) with $A = 50$ nN and $B = 7.0$ nN/fC^2.

are intermediate transfer belts. These belts are electrically semi-insulating, but they behave as image planes as long as the time between toner attachment and detachment is longer than the electrical relaxation time.[16] Each measurement taken for differently charged samples produced a distribution of adhesion, and the force for 50% removal is shown. Superimposed on that data is a best-fit curve of the form described in Eq. (4), where $A = 50$ nN and $B = 7.0$ nN/fC^2. In this theoretical prediction, $A = 50$ nN is interpreted as the mean van der Waals adhesion force for the samples. The slope factor on the charge-squared term, $B = 7.0$ nN/fC^2, is higher than the theoretically predicted value of 1 by almost an order of magnitude. The quality of fit between theoretical and experimental results is acceptable, although it appears that the experimental curve may be more sensitive to charge than is predicted by the two-term model. The model in Eq. (4) assumes separate van der Waals and electrostatic contributions to particle adhesion. In what follows, we show that this assumption forms a poor theoretical basis for interpreting particle adhesion measurements and is critical to resolving the debate over particle adhesion.

Impact of Non-uniform Charge

It has long been suspected that non-uniform charge distributions on toner particles increase the adhesion forces.[19] In printer systems, recharging typically occurs on the top of the particle by electrostatic breakdown of air, for example during the passage of toner through additional transfer nips. Such recharging commonly occurs post nip due to Paschen discharge at downstream first transfer stations in color transfer systems. This results in the toner having additional charge on the air side of the toner particles post nip. However, it has been pointed out that such charge imbalances will result in a net torque on the particle causing a sufficiently spherical one, such as those used here, to rotate so that the majority of charge will be oriented closest to the image plane.[20] The chemically processed toners used in this study have a mean circularity of 0.97. In our adhesion measurement experiments, we increase the charge by adjusting the first transfer voltage on the downstream

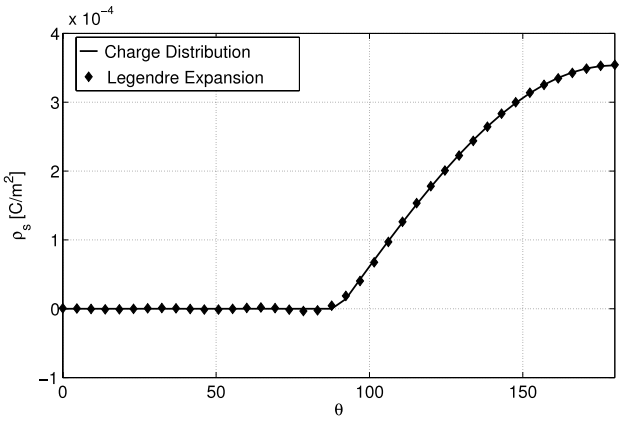

Figure 2. Surface charge distribution ρ_s as a function of zenith angle θ as used in the model along with the spherical mode representation ($N = 10$ modes) using a summation of Legendre functions.[16] For this case, the uniform portion of the charge density is $\rho_0 = 0$ and the total charge per particle is $Q = 10$ fC.

first transfer rollers. The measured average charge values presented herein confirm that this approach increases the average charge on the printed sample.

To study the effect of non-uniform charge on particle adhesion, an analytical model has been employed.[16] This model has also been used to explain the nonlinear relationship between the charged particle detachment force and applied electric field, which is consequential to toner transfer.[18] The model consists of expansion of the electrostatic fields as analytical solutions to the Laplace equation in spherical coordinates near each particle. Likewise, the surface charge density $\rho_s(\theta, \phi)$ is also expanded in the spherical basis. The boundary conditions are satisfied by use of a translational theorem with the center of each particle representing the origin of a spherical coordinate basis. The force on each particle is then calculated from the divergence of the Maxwell stress tensor,[21] which reduces to a simple sum of the mode coefficients.[22] As the model is applied here, a toner particle is assumed to be charged uniformly to some base charge density ρ_0. Additional charging occurs via a uniform ion stream which impacts only the exposed top half of the particle. Because the particle is spherical, the resulting additional charge distribution $\Delta\rho$ on the top half will be a cosine function with respect to the zenith angle. If the particle then rotates under the additional force imbalance, the charge on the bottom of the particle ($\pi/2 < \theta < \pi$) is then described by

$$\rho = \rho_0 - \Delta\rho \cos(\theta), \tag{5}$$

where the charge ρ_0 on the top of the particle ($0 < \theta < \pi/2$) and θ is the usual spherical coordinate. This charge distribution is shown in Figure 2 along with the spherical mode expansion used to represent the charge density.[16] In the case shown, the total charge is $Q = 10$ fC, obtained by integrating the charge density over the surface of the sphere.

While studying the outputs of the electrostatic adhesion model,[16] it became obvious that the electrostatic adhesion

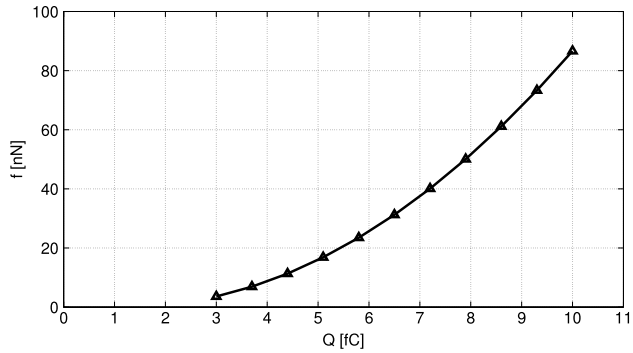

Figure 3. Force versus charge for a single dielectric particle with $\epsilon_p = 3$. The particle has a uniform base charge density ρ_0 providing 3 fC of charge plus an additional charge according to Eq. (5) to bring the total charge to the value indicated on the graph. The resulting curve has constant, linear, and squared dependence. The markers show the output of the analytical adhesion model and the line indicates the best fit curve.

force is not only a function of Q^2 as predicted by Coulomb's law, but rather

$$F_{electrostatic} = B_0 + B_1 Q + B_2 Q^2. \tag{6}$$

It should be pointed out that B_1 is non-zero only when the magnitude and distribution of the particle surface charge are varied at the same time. Such a variation can occur unintentionally, for example during post-nip ionization. As an example, Figure 3 shows a theoretical plot of adhesion force versus charge for a single dielectric particle with base uniform charge of 3 fC and additional charging up to a total of 10 fC using the previously described charge distribution. The polynomial $f = 2.01 - 2.87Q + 1.13Q^2$ fits the model output exactly, with the Q and constant terms arising from the non-uniform surface charge distribution. The coefficients B_0 and B_1 are due to non-uniform charging.

The existence of non-quadratic terms in the electrostatic adhesion force is a novel contribution. The coefficients B_0 and B_1 arise due to non-uniform charging, and B_2 gives the proportion of electrostatic force due to all Coulombic interactions. To illustrate why this is, consider the simple case where a particle has discrete charges q_1 and q_2 on the top and bottom, respectively, as shown in Figure 4. The total charge is $Q = q_1 + q_2$. The total force is obtained by summing forces between the charges and the images

$$\bar{F} = -\hat{z}\frac{1}{4\pi\epsilon_0}\left[\frac{q_1^2}{(4R)^2} + 2\frac{q_1 q_2}{(2R)^2} + \frac{q_2^2}{(2\delta)^2}\right]. \tag{7}$$

The method of increasing toner charge by air ionization at transfer zones tend to increase q_2 while maintaining q_1 constant. Therefore, $q_2(Q) = Q - q_1$, and the terms in the brackets can be written as

$$\left[\left(\frac{q_1^2}{4\delta^2} - \frac{7q_1^2}{16R^2}\right) + Q\left(\frac{q_1}{2R^2} - \frac{q_1}{2\delta^2}\right) + Q^2\left(\frac{1}{4\delta^2}\right)\right]. \tag{8}$$

Therefore, a plot of force versus charge will include a linear term and a constant term (q_2 is constant). The significance

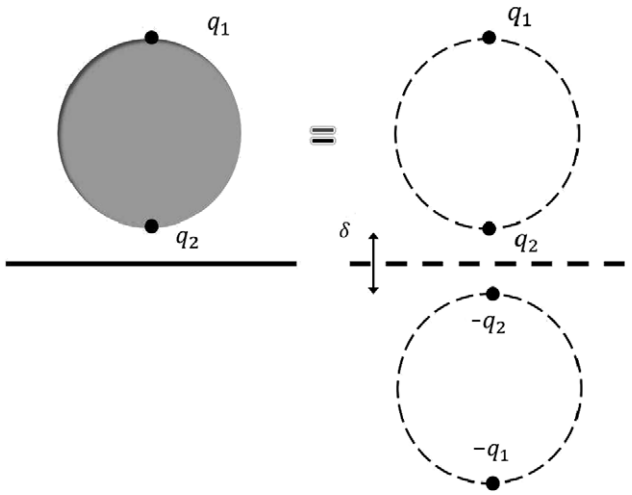

Figure 4. Example non-uniform charge distribution for illustration. Left: A particle above a conductive substrate with dielectric constant $\epsilon_p = 1$ and radius R has discrete charges q_1 and q_2 on the top and bottom, respectively. Right: Image theory equivalent of the charge distribution.

of this is that (i) the dependence of adhesion force on total charge under experimental conditions of increasing the charge by air ionization may not just be total charge squared as obtained from Coulomb's Law, and (ii) the emergence of a constant term B_0 indicates that decoupling of electrostatic and dispersion forces may be very difficult. We stress that this effect is a result of the experimental method used to increase the toner charge. It occurs because several measurements of adhesion force were made with different toner charges where the toner charge was increased in a non-uniform manner.

Impact of the Environment

Given the model described above, toner adhesion for the same system should be predictable for different environments. Electron microscopy reveals loosely held toner sitting upon a few silica nodules or resting on a small portion of the toner. These toner particles represent a high percentage of toner that is removed with little force. The toner that is tightly held can frequently be seen to have large contact area with the substrate.[9,13,14] It is reasonable that the initial constant A might change as the work of adhesion changes, for example with the presence of water bridges at higher humidity. However, the electrostatic portion should be a function of toner charge only. This is not what actually happens when the system is tested. Figure 5 shows data for toner adhesion to an intermediate transfer belt for three different environments 60°F, 8% relative humidity; 75°F, 40% relative humidity; and 78°F, 80% relative humidity. For the same toner charge, there are different adhesion values depending on ambient conditions. The two-term model can be modified to give a good general prediction of the adhesion force measured. However, the model is not capable of reflecting the change in adhesion response to charge for different environmental conditions.

The existence of a change in charge-response in different environment conditions suggests an additional physical

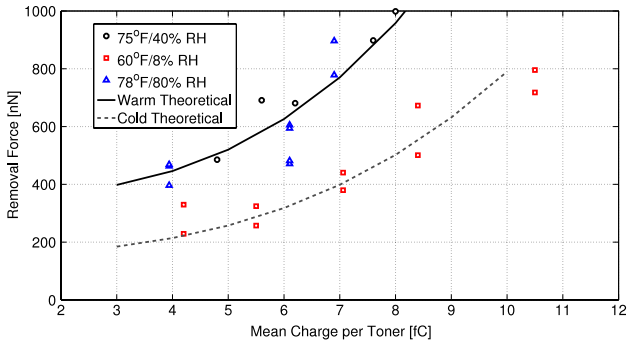

Figure 5. Average removal force versus average total charge per toner for different environments. Temperature and relative humidity are shown in the inset. The adhesion data is for toner adhered to an intermediate belt where the printer was acclimated and stopped during the printing process at each of three different environments shown. The curves are given by Eq. (10) for warm data ($\alpha_0 = 350$ nN, $\alpha_1 = 4.0$ nN/fC, $\alpha_2 = 1.0$ nN/fC^2, and $\alpha_3 = 1.0$ nN/fC^3) and cold data ($\alpha_0 = 150$ nN, $\alpha_1 = 4.0$ nN/fC, $\alpha_2 = 1.0$ nN/fC^2 and $\alpha_3 = 0.5$ nN/fC^3).

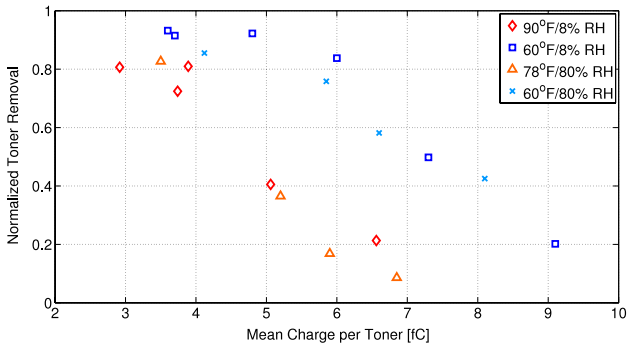

Figure 6. Normalized toner removal (toner removed/toner originally present) versus average charge magnitude in fC for four environments. Temperature and relative humidity are shown in the inset. The matrix of data shows that the toner adhesion changes with environment are a function of temperature.

mechanism. The first step in identifying and understanding this mechanism is to separate the temperature effect from the humidity effect to see which of these is causing the change in charge sensitivity, or indeed if both are. Figure 6 shows the results of tests for the adhesion dependence of charge to separate out the humidity and temperature effects. Toner adhesion was tested at a matrix of high and low temperatures and high and low relative humidity to determine the relative impact of each parameter. The graph shows that the data falls into two groups divided by temperature. Warm temperatures have higher adhesion than cooler temperatures across a wide range of relative humidity levels.

There are several possible mechanisms for heat to impact toner adhesion. If the temperature simply increased the dispersive adhesion force, the expected outcome would be a curve with the same sensitivity to charge, but a higher offset. There is an offset difference seen in the data above, suggesting that potentially the "wetability" of the material could be changing with temperature. This offset can be compensated for by altering the coefficient A in the two-term model of Eq. (4), but coefficient B should not be sensitive to changes

in material properties and thus the slope of the data. As a result the slope of the data at any charge level should be independent of the environmental conditions. This is not the change seen in the data in Fig. 5.

Another potential mechanism could be the heat causing a small relaxation in the toner particle resulting from a change in modulus of elasticity. A decrease in toner modulus would cause a small deformation of the particle as electrostatic forces pulled it toward the transfer belt. This could change the adhesion of the toner to the substrate in two ways. The first mechanism would be the decrease in distance between the toner and its image charge, and the second mechanism would be an increase in contact area caused by the modest deformation.

Jurgen Tomas' work describes the effect of deformation of a particle under the pull of gravity on adhesion.[23] Using his work and substituting the Coulombic attraction for gravity, one can make an estimate of the relative potential contribution of each of these mechanisms. The normal force on a particle from the substrate is a function of Coulombic attraction, which is a function of toner charge squared. The adhesion measured by rolling resistance of a spherical particle is a function of the normal force to the 3/2 power.[23] Inserting a charge-squared term in for normal force yields a pull-off force that is sensitive to the charge of the toner cubed.

The first-order impact of the adhesion of a particle as a function of the modulus of elasticity has been suggested many times.[23–25] Actual changes in toner modulus with temperatures lower than the toner softening point have not been published. Nano-dynamic mechanical analysis testing on individual toner particles was performed to investigate particle softening below the glass transition temperature. These measurements showed an increase in loss modulus (i.e. the measure of elastic stored energy) for toner at 38°C as compared to toner at 25°C. The tan Delta value, or the ratio of the loss modulus (i.e. the measure of dissipated energy) to the storage modulus, increased from an average value of about 0.03 at 25°C to about 0.05 at 38°C, indicating more energy lost to permanent deformation given a force input. These values agreed well with those from macro samples of the same material, suggesting that the gentle drop in storage modulus below the official melting temperature was impacting the contact area and therefore the adhesion of the particles. When these values are used to predict contact area changes they can account for part of the difference in adhesion seen between the different environments.

The elastic radius of contact under load can be approximated by[23]

$$R_{el} = \left(\frac{3}{2}\frac{RF_n}{\mathcal{E}}\right)^{\frac{1}{3}}, \tag{9}$$

where F_n is the normal force causing the deformation and $\mathcal{E}$ is the modulus of elasticity. Consider, for example, a 6 μm ($R = 3$ μm) toner charged to 4 fC at 16°C (60°F) without an applied field. The Coulombic attraction in Eq. (2) predicts a normal force of 4 nN to the substrate. However, the analytical expansion model reveals that the electrostatic component adhesion will actually be about ten times that value.[16] From Fig. 5, we see that the total adhesion measured for 90% removal at 60°C is around 200 nN. Given that the van der Waals force for this particle is about 150 nN, the contact radius should be close to 0.013 μm, as calculated from Eq. (3). Under the additional load of a transfer field $E_0 = 1E7$ V/m, the additional normal force is $QE_0 = 40$ nN, and the contact radius will climb to 0.028 μm according to Eq. (9), where $\mathcal{E} \approx 3046$ N/mm^2 for polymers. According to tan Delta measurements, about 5% of the deformed radius will remain after the additional electrostatic force is removed at 90–100°F. If this warm toner particle undergoes two or more transfer fields, the contact area could easily remain at 0.020 μm toward the end of the printing process. The associated adhesion forces would be 150 + 40 = 190 nN for the cold toner at the second transfer and 220 + 40 = 260 nN for the warm toner of the same charge at second transfer. These calculations partially explain the difference in adhesion between warm (90°F) and cold (60°F) toner. Since the data presented herein (see Fig. 4 for example) represents 90% removal, we expect some toner removed to be mechanically deformed by the printing process.[14] Of course, compression of this nature will likely have some temperature dependence as well, and is expected to effect the toner contact area.

DISCUSSION

Examining the adhesion of toner in different environments sheds light onto the actual physics of particle adhesion. The proposed model is now a cubic function of charge.

$$F_{adhesion} = \alpha_0 + \alpha_1 Q + \alpha_2 Q^2 + \alpha_3 Q^3. \tag{10}$$

In reviewing this model, coefficient α_2 is responsible for all Coulombic interactions, while non-uniform charging can contribute to α_0, α_1, and α_2. In fact, the manner in which charging occurs can couple the constant terms so that $\alpha_0 = A + B_0$. Thus, the constant component represents an upper bound for the dispersive forces. The α_0 and α_3 coefficients are likely to change as a function of temperature, while small changes in α_1 and α_2 may occur as the particle charge distribution moves closer to the substrate during deformation. Coefficient α_0 can change as the toner or the substrate wetability changes with increased molecular energy. Coefficient α_3 can change with the modulus of elasticity as the material softens slightly with temperature. The cubic dependence on charge stems from the fact that, due to particle deformation, the adhesion force is proportional to the normal force to the 3/2 power, which in turn is proportional to Q^2 due to Coulombic attraction with the image charge. If this model is capturing the main features of the physics, it should be able to track the change seen across environments.

Adhesion as a Function of Charge

For the warm curve shown in Fig. 5, the coefficients of Eq. (10) are $\alpha_0 = 350$ nN, $\alpha_1 = 4.0$ nN/fC, indicating the effect of non-uniform charge, $\alpha_2 = 1.0$ nN/fC^2 (for the Coulomb attraction), and $\alpha_3 = 1.0$ nN/fC^3. For the cold curve, the coefficients are $\alpha_0 = 150$ nN, $\alpha_1 = 4.0$ nN/fC, $\alpha_2 = 1.0$ nN/fC^2, and $\alpha_3 = 0.5$ nN/fC^3. Non-uniform charge was enhanced in all of these data sets due to the manner in which the higher charge samples were obtained, namely increasing the transfer voltage downstream of the initial transfer station to shower the toner with charged ions. The model above indicates a more complex interaction between charge and adhesion than had been previously suggested.

This complexity is partly responsible for the controversy over how the charge on a small particle can impart such a strong adhesion force. The combination of charge on the surface of a finite particle being non-uniform, expandable to multiple particles, and having a strong impact on the resulting contact area all contributes to high charge-based adhesion forces. Of these, the contact portion is a cubic function of charge, and therefore is very significant for actual particle adhesion.

Conclusions

A toner adhesion measurement tool has been applied to measure toner as a function of environmental conditions, and an electrostatic adhesion model has been applied to study the effect of non-uniform toner charging. Both electrostatic and non-electrostatic contributions to the adhesion are observed in the toner samples, and increased charge or increased contact area causes measurable increases in toner adhesion. Changes in environment demonstrate a more complex interaction between charge and adhesion, resulting in a cubic term based on increased contact area with increased electrostatic force. Additionally, non-uniform charging results in a linear term and an electrostatic contribution to the constant term when plotting the adhesion force versus charge. The implication of these results is that the complex interactions between the electrostatic and dispersive force contributions make it difficult to decouple these contributions. This complexity is one reason behind the persistence of the toner adhesion controversy.

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