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Pamela J. Forness
Program Manager and Managing Editor of *JIST*:
The Journal of Imaging Science and Technology
IS&T: The Society for Imaging Science and Technology
7003 Kilworth Lane
Springfield, Virginia USA 22151
Voice: 703-642-9090 extension 16
FAX: 703-642-9094
Email: pam@imaging.org <mailto:pam@imaging.org>
WEB: http://www.imaging.org

Experimental Verification of the Proximity Theory of Toner Adhesion

L.B. Schein✦,*, W. Stanley Czarnecki†, Bruce Christensen, Taomo Mu, and Graham Galliford▲,‡

Aetas Technology Incorporated, Irvine, California, USA

Recently a new theory of toner adhesion was suggested. In this new theory an electrostatic proximity force of adhesion is taken into account. The proximity force is due to the attraction of charges on the toner particle in close proximity to a conductive plane to their respective image charges in the conductive plane. In this paper experimental verification of this new theory is presented using a 16 μm diameter, ground toner with silica additives. The theory is fit to complete curves of the developed and residual (1) mass per unit area, (2) charge-to-mass ratio, and (3) size distribution in an electric field detachment experiment, a much more stringent test of an adhesion theory than has been published before (to the authors' knowledge). The observed adhesion and ratio of the measured toner adhesion to theory are the lowest ever observed (to the authors' knowledge).

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Introduction

Toner adhesion is active in the subsystems of electrophotography in which toner particles are moved between surfaces, including development, transfer, and cleaning. Yet the source of toner adhesion remains under discussion, despite intensive investigations starting with the early work of Goel and Spencer,[1] a review by Hays,[2] and papers by Gady and co-workers[3] and others.[4,5] Measurements[1–5] generally indicate that the force of adhesion is at least an order of magnitude larger than expected by simple image force calculations that model the toner charge by locating it in the center of toner particle. In order to account for this large discrepancy, Hays[2] proposed that one must take into account non-uniform surface charge distributions and Gady and co-workers[3] applied classical JKR theory to calculate the surface adhesive van der Waals forces.

Recently a new electrostatic force of adhesion was suggested.[6] It was pointed out that a spherically symmetric distribution of charge points can only be modeled as having its charge in its center in free space. In contact with a conductive plane there is an additional contribution to the force of adhesion, called the proximity force, which equals $4/\pi$ times the usually assumed image force. In this new theory of toner adhesion, it is suggested that at every contact point this proximity force is active.[7] Therefore the force of adhesion has three terms, the usually assumed image force, the proximity force times the number of contact points, and the van der Waals force times the number of toner (or silica) asperities which contribute to the adhesion. Experimental verification of this new theory is presented in this paper for a 16 μm diameter, ground toner with silica added to its surface.

Theory of Toner Adhesion

Throughout the literature on toner adhesion it is always assumed that the adhesion of a charged toner particle in contact with a conductive plane can be modeled as though the charge is in the center of the particle. This gives for the adhesion of the particle (to its image) an electrostatic toner adhesion of

$$F_b = \frac{1}{4\pi\varepsilon_0}\frac{Q^2}{d^2} \tag{1}$$

where Q the toner charge and d is the toner diameter (ignoring the dielectric properties of the toner). This equation results from applying Gauss' Law to a spherically symmetric charge distribution in free space. However, when a spherically symmetric distribution of charge points is in contact with a conductive plane, a simple geometry (a sphere) cannot be found over which the electric field is normal. The electrostatic problem of a spherically symmetric distribution of charge points in contact with a conductive plane has recently been solved by finite element analysis using the method of images. It was shown[6] that near a conductive plane there is an additional force, which is called the proximity force

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✦ IS&T Fellow

▲ IS&T Member

* Independent Consultant, 7026 Calcaterra Dr., San Jose, CA 95120
† Torrey Pines Research, 6359 Paseo Del Lago, Carlsbad, CA 92009 Current address: IBM Corporation, 5600 Cottle Rd., Building 13, San Jose, CA 95193
‡ Independent Consultant, P.O. Box 217, 3844 West Channel Islands Boulevard,Oxnard, CA 93035

(because it is due to the charge in proximity to the contact point), which equals 4/π times the usual force

$$F_p = \frac{4}{\pi}\frac{1}{4\pi\varepsilon_0}\frac{Q^2}{d^2} \tag{2}$$

The total electrostatic force of adhesion is just the sum of these two forces.

Having shown that a spherically symmetric distribution of charge points has a force of adhesion to a conductive plane that is composed of two parts, one of which is due to a newly identified proximity force, we now apply this result to toner.[7] Assume that toner particles are not perfect spheres and consequently have many contact points. We suggest that at each contact point the proximity force is active. If there are n_p contact points, the electrostatic force of adhesion is then

$$F = \frac{1}{4\pi\varepsilon_0}\frac{Q^2}{d^2} + n_p\frac{4}{\pi}\frac{1}{4\pi\varepsilon_0}\frac{Q^2}{d^2} \tag{3}$$

At each contact point there is in addition the possibility of van der Waals forces.[3] Therefore the total adhesion force is predicted to be

$$F = \frac{1}{4\pi\varepsilon_0}\frac{Q^2}{d^2} + n_p\frac{4}{\pi}\frac{1}{4\pi\varepsilon_0}\frac{Q^2}{d^2} + n\frac{3}{2}\omega_A\pi R \tag{4}$$

where ω_A is the thermodynamic work of adhesion and R is effective radius of the asperities of the particles contacting the plane (either toner or silica asperities) and n is the number of toner (or silica) asperities which contribute to the adhesion. It should be clear that the large discrepancy between reported measurements and calculated adhesion based on Eq. (1) is easily resolved if n_p is on the order of 10 to 40 contacts, which is not unreasonable. Further, large distribution of forces, which are also observed, can easily be ascribed to large distributions of contact points among the toner particles. Quantitative experimental verification of this theory is presented in the next section.

Experimental Verification

An electric field detachment experiment was carried out in a single component development system in which the toner is charged against an aluminum development roller after passing under a counter rotating supply roller and a polyurthethane doctor blade (all tied together electrically). A metal cylinder was spaced 150 μm from the development roller moving at the same speed (usually 55 mm/s, although experiments at 2x, 0.5x, and at zero speed gave the same results). A bias voltage between the metal cylinder and the development roller allowed for the electric field detachment experiment. The toner mass per unit area M/A and charge to mass ratio Q/M can be measured with standard[8] vacuum pencil techniques both on the cylinder and remaining on the development roller ("residual"). Collected toner in the filter of the vacuum pencil can be analyzed by Coulter Counter for toner size distributions.

Toner "develops" from the development roller to the metal cylinder when the Coulomb force QE exceeds the force of adhesion where E is the applied field. Expressing E as V/L where L is the gap (150 μm in these experiments), toner develops when V is larger than (using Eq. (4))

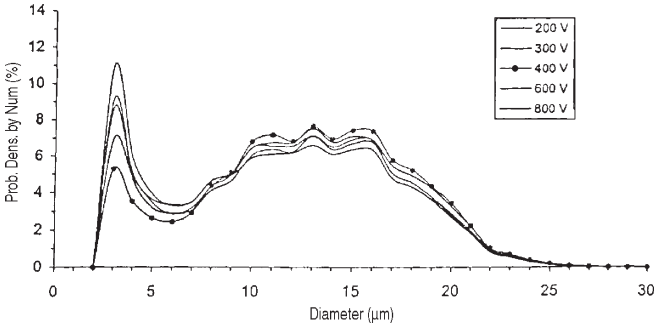

Figure 1. The size distribution of the toner on the development roller as measured by Coulter Counter. The size of the diameter buckets has been altered to be a uniform 1 μm increments to facilitate comparison with theory. Some variation seen with development roller voltage, which is probably due to the supply roller not being 100% efficient, is averaged out.

$$V = \frac{L}{4\pi\varepsilon_0}(1+n_p\frac{4}{\pi})\frac{Q}{d^2} + 1.5\frac{L}{Q}n\omega_A\pi R \tag{5}$$

or using Q/M and retaining the R in the van der Waals expression

$$V = \frac{L\rho}{24\varepsilon_0}(1+n_p\frac{4}{\pi})\frac{Q}{M}d + 9\frac{L}{\rho}n\omega_A\frac{R}{\left(Q/M\right)d^3\right)} \tag{6}$$

where ρ is the toner density.

Note that the existence of the van der Waals term implies that at $Q = 0$, infinite V is required to develop toner. This is a standard optimization problem. There exists an optimized Q/M

$$\left(\frac{Q}{M}\right)_{opt} = \frac{6}{d^2\rho}\sqrt{\frac{6Rn\omega_A\varepsilon_0}{(1+n_p\,4/\pi)}} \tag{7}$$

which produces a threshold voltage for development which is determined by the magnitude of the van der Waals force

$$V_{th} = \frac{L}{2d\varepsilon_o}\sqrt{\left(1+n_p\,4/\pi\right)6Rn\omega_A\varepsilon_0} \tag{8}$$

for single values of Q/M and d. (Van der Waals forces can also be separated from electrostatic forces by examining the y-axis intercept in plots of F versus $(Q/M)^2$ as can be seen by inspecting Eq. (4). This procedure has been carried out by many workers, most recently by both papers in Ref. 4, with similar results as obtained here, i.e., the van der Waals force is small.)

Of course Q/M and d are distributed properties. Our procedure is to measure the size distribution of the toner on the development roller (see Fig. 1 which is a plot of the number distribution) and then to use the experimental charge distributions reported in Refs. 9 and 10 to guide a choice of charge distribution which will give the measured average Q/M. Figure 2 is a scatter plot of the Q/M and d of each particle used in this Monte Carlo simulation. This results in a set of particles each with its own Q/M and d, which can be tested against Eq. (6)

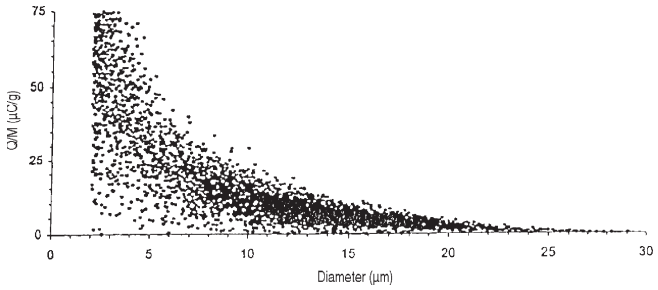

Figure 2. A scatter plot of the 4000 particles used in the simulation, indicating their Q/M and d. Note that the normal distribution at each d can be seen by inspection.

to determine at what voltage the particle develops. The developed and residual M/A, Q/M and size distribution can then be predicted. The prediction of a threshold behavior remains even in the presence of the distributions if van der Waals forces exist. (If van der Waals forces are zero, then the threshold is zero and development starts at zero voltage, as long as the charge distribution has some charged particles with very low charge, as observed in Refs. 9 and 10 and assumed in our distributions, shown in Fig. 2).

The experiment and theoretical fit for a 16 μm diameter, ground toner with Q/M = 5.3 μC/g are shown in Fig. 3 and Fig. 4. The toner was optimized to have minimum adhesion and had approximately one monolayer of silica on the surface. The data are shown in Fig. 3. On the top left is development efficiency, scaled to 100% of the toner on the development roller. The top right is a plot of developed and residual Q/M. The bottom left is the developed particle size distribution for 5 voltages (derived from Coulter Counter data by converting the diameter axis to uniform increments to make it easier to compare to theory). The bottom right is the residual particle size distribution. The best fits to the data are given in Fig. 4.

The procedures and values to the parameters chosen for the theory are as follows:

1. The Q/M distribution is assumed to follow a simple function $A/(d + B)$ where A and B are constants to be found, such that the measured average Q/M is obtained. The distribution is assumed normal at each diameter such that 5σ (statistically speaking) of the particles falls between 0 and $A(d + B)$. There is a range of values of A and B which can produce the correct measured Q/M. Within this range of acceptable values, the Q/M distribution is iterated to obtain the best fit to the data.
2. The scaling parameter n, of the van der Waals force defines the number of asperity contact points. In these experiments the amount of silica was deliber-

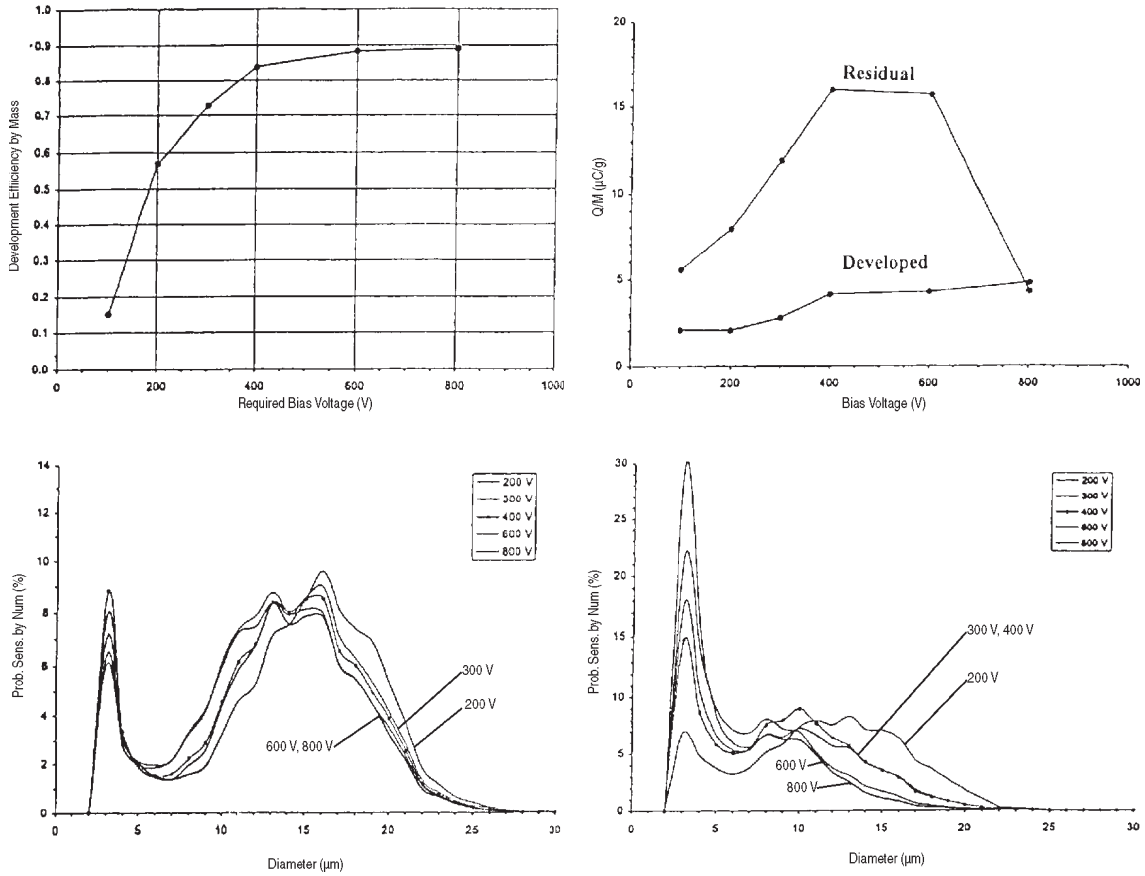

Figure 3. Experimental data taken on 16 μm toner which has about one monolayer of silica on the surface. The upper left is the development efficiency. The upper right is the developed and residual Q/M. The lower left is the size distribution of the toner developed at each voltage. The lower right is the size distribution of the toner left behind (residual) at each voltage.

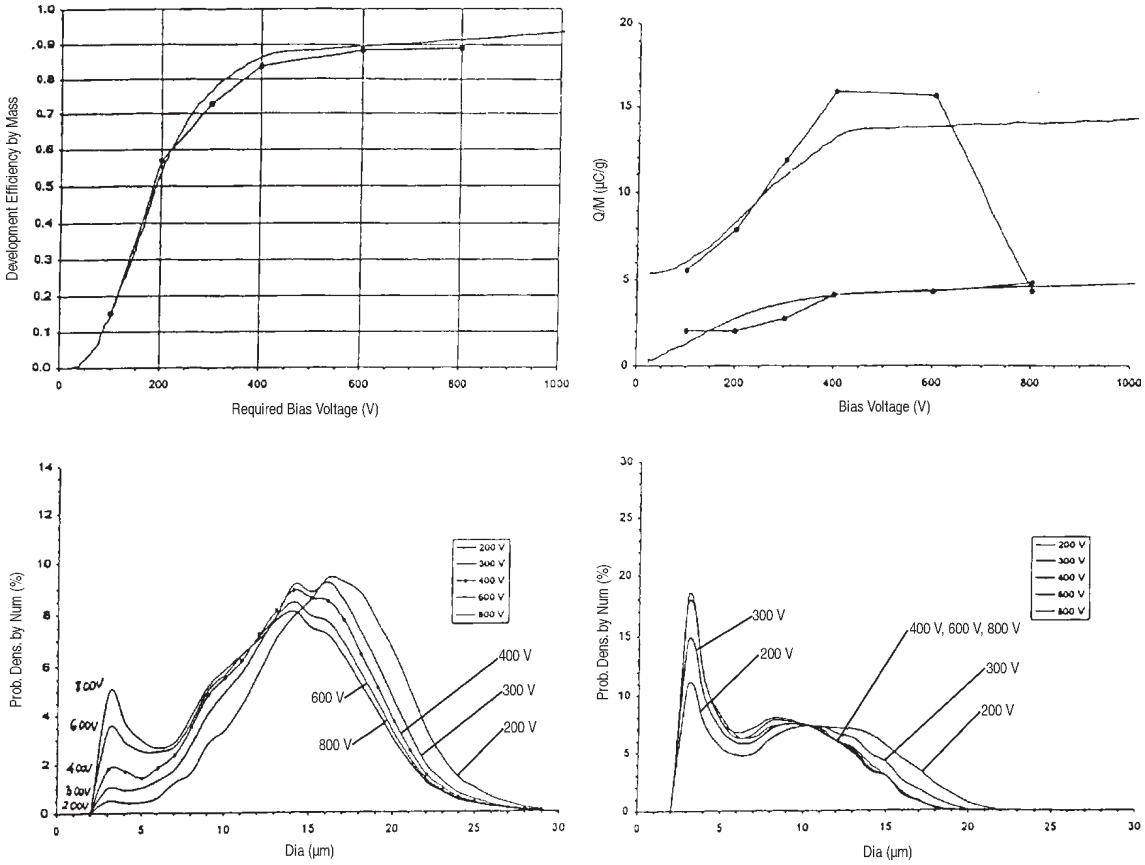

Figure 4. Best fit to the data (see text)

ately chosen to be high enough so that only silica contacts to the substrate are possible, i.e., toner resin contacts are eliminated. The value of n is chosen in the theory to give (1) the intercept seen in the development efficiency and (2) to obtain the correct amount of small particles observed to develop at low biases. For this best fit n is [0.01 + *Rnd* (0.4)] where *Rnd* is a computer function that generates a random number between 0 and 1. This means that n is a random number between 0.01 and 0.41. The fact that it is less than unity simply means that the other parameters in the van der Waals theory, the radius of the silica asperities (15 nm) or thermodynamic work of adhesion (0.05 J/m^2), probably should be chosen to be lower values. As can be seen by comparing Figs. 4 and 5 (in which single values of Q/M and d are used and the van der Waals force is assumed zero) the van der Waals force controls the intercept.

3. A uniform or single value for n_p was not successful in fitting the data. It appears necessary to postulate a distribution for n_p, and that those toners with higher $(Q/M)d$ products have higher number of contact points. For the best fit, we choose

n_p = [1 + *Rnd* (1.8)] for $(Q/M)d$ = 0–80 μm – μC/g

n_p = [1 + *Rnd* (2.0)] for $(Q/M)d$ = 80–116

n_p = [1 + *Rnd* (9.0)] for $(Q/M)d$ = 116 and above.

For example, [1 + *Rnd* (1.8)] means the values are randomly and uniformly distributed between 1 and 2.8.

Note that most of the toner is characterized by n_p being randomly distributed between 1 and 2.8. This can be rationalized as follows: since 3 points define a plane, most of the toner is balanced on the planar conductive surface by three points, probably three silica particles. Some however are balanced against neighboring toner, so n_p is smaller for those particles, giving a distribution from 1 to 3 (actually 2.8). This suggest that the minimum electrostatic adhesion force is characterized by n_p = 3.

In comparison with prior adhesion measurements, this toner has the lowest adhesion and the lowest ratio of observed adhesion compared to theory Eq. (1) ever reported, to the author's knowledge. Using the d (16 μm) and Q/M (5.3 μC/g) values for this toner, the adhesion force predicted from Eq. (1) is 4.5 nN. The observation at the 50% point is 190 volts, which corresponds to an adhesion force of 14.2 nN, or a ratio of observed to predicted adhesion of 3.2, which is lower than previously reported.[2,7] Of course this theory suggests that the data can be analyzed more physically: the approximately 100 volt intercept in Fig. 3 is due to van der Waals forces and the electrostatic component of adhesion is characterized by n_p = 3, which we have suggested above is a minimum value.

This choice of different values of n_p for different values of $(Q/M)d$ can be rationalized as follows: $(Q/M)d$ is actually Q/A which is proportional to the effective electric field in the electric field theory of toner charging.[11] The effective electric field is a material determined parameter which accounts for all known charging behaviors of toner particles. Particles with different effective electric fields have different surfaces, which may be related to why they have more contact points. Examples

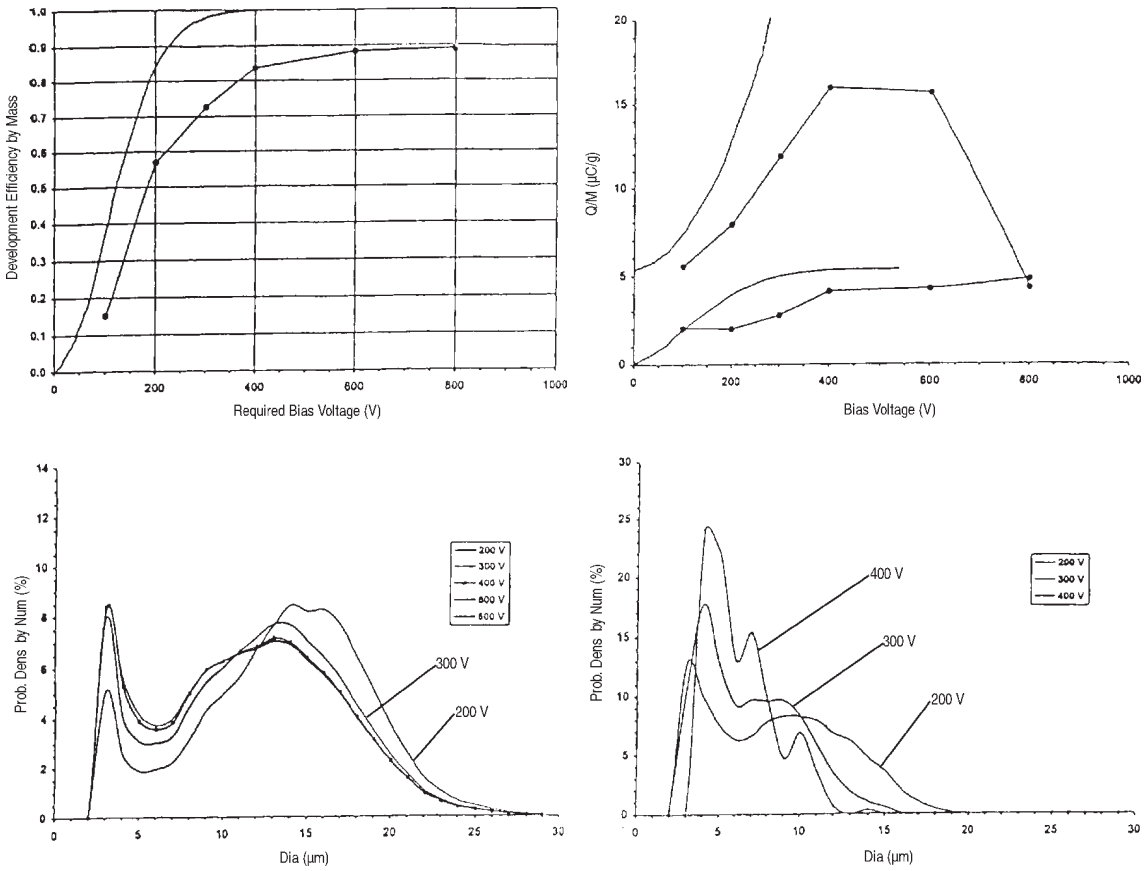

Figure 5. Fit to the data assuming only electrostatic adhesion and only one contact point per toner particle.

of fits to the data with other distributions of n_p are shown in the subsequent graphs. These larger values of n_p (which are needed to account for 15% of the toner particles) are a way to characterize a small fraction of the toner with higher adhesion, something not unexpected in a manufactured ground toner.

To allow the reader to better understand the rationale for our choices of the parameters and the sensitivity of the data fitting to the parameter choices, fits to the data with other choices of the parameters are shown in the subsequent figures. In these figures, the A (0.56 μC/g) and B (–3.99262 μm) parameters are not changed, i.e., the charge distribution is not changed.

In Fig. 5, only the minimum electrostatic adhesion is assumed (Eq. (6) with $n_p = 1$ which would be the result of one contact per toner particle). Note that this is the steepest curve that can be expected with this particular size toner and charge distribution. It takes approximately 300 volts to obtain full development. The intercept is zero, which is expected because van der Waals forces are assumed absent.

In Fig. 6, the effect of a nonuniform charge distribution is considered. Zero van der Waals force is assumed. The proximity force is present but a nonuniform charge distribution is assumed instead of the normal spherically symmetric charge distribution. The nonuniformity is characterized by a parameter f, such that the bottom of the toner particle has f times the charge per unit area of the top of the particle, $f = 2$ is used in the fit shown. The transition point between the top and bottom is chosen so that the total charge is unchanged compared to the other graphs. Clearly the fit is poor. (For this nonuniform distribution, the electric field at the interface between the toner and the metal ground plane was verified to be less than 3 V/μm, the macroscopic electric field at which air breakdown occurs). Other values of f were examined, but fits to the data were also unsatisfactory.

In Fig. 7, van der Waals force is zero. The n_p value is chosen to be [1 + *Rnd* (1.8)] throughout the whole distribution. Clearly the fit is not good at high voltages.

In Fig. 8, van der Waals force is again zero. The n_p value is chosen to be [1 + *Rnd* (1.8)], [1 + *Rnd* (3.2)], [1 + *Rnd* (3.2)] in the same three ranges used in Fig. 4. Again the fit is not good, but it is improved.

In Fig. 9, van der Waals force remains zero; n_p values are [1 + *Rnd* (1.8)], [1 + *Rnd* (3.2)], [1 + *Rnd* (9)] in the same three ranges used in Fig. 4. Fit is further improved.

In Fig. 10, van der Waals force is added; n = [0.01 + *Rnd* (0.4)] and n_p = [1 + *Rnd* (1.8)], [1 + *Rnd* (3.2)], and [1 + *Rnd* (9)]. The addition of the van der Waals force accounts for the intercept.

Figure 4 shows the final fit, n = [0.01 + *Rnd* (0.4)] and n_p = [1 + *Rnd* (1.8)], [1 + *Rnd* (2)], [1 + *Rnd* (9)]. This is the best fit.

Summary

A new theory of toner adhesion has been proposed based on a recent electrostatic result that suggests that there is an electrostatic force of adhesion at every contact point, called the proximity force. Because of the many possible contact points between a toner (or toner with silica on the surface) and a conductive plane, this theory can naturally account for the large magnitude of toner

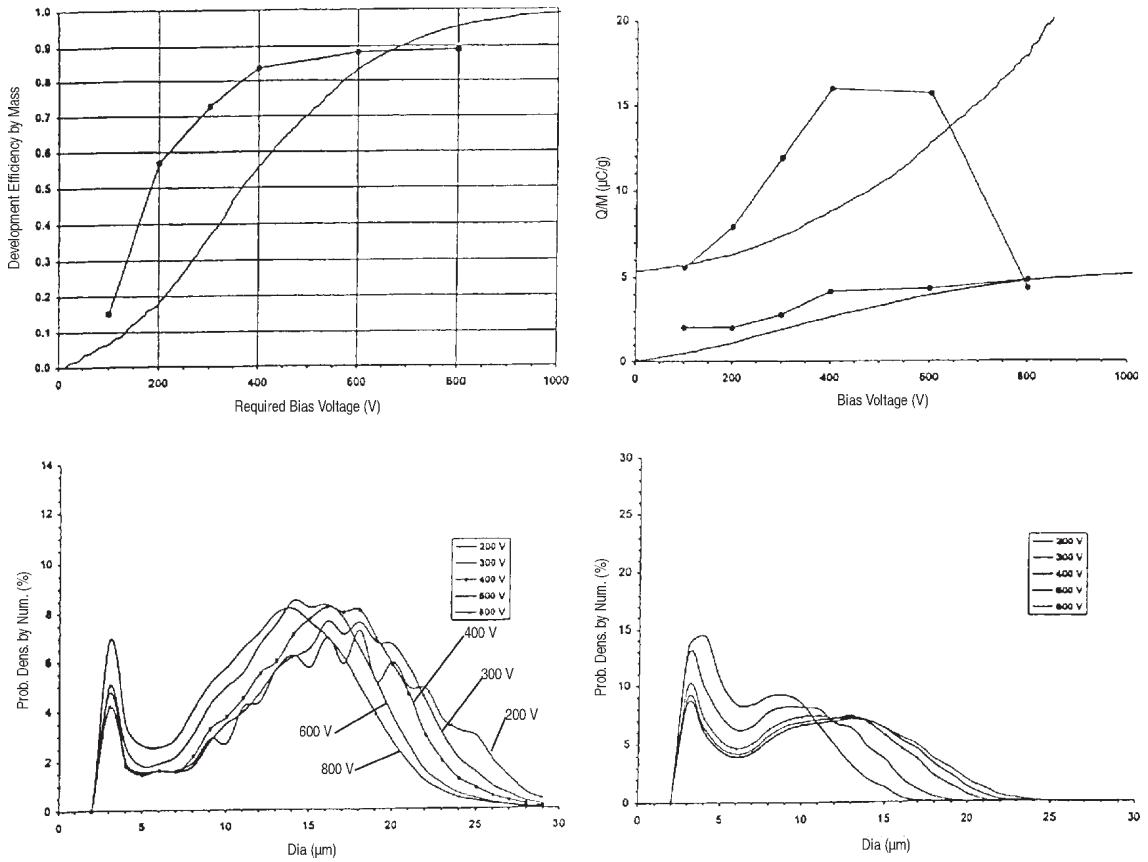

Figure 6. Fit to the data assuming a non uniform charge distribution. The nonuniformity is characterized by a parameter f, such that the bottom of the toner particle has f times the charge per unit area of the top of the particle. $f = 2$ is used in this fit. The transition point between the top and bottom is chosen so that the total charge is unchanged as compared to the other graphs. Zero van der Waals force is assumed.

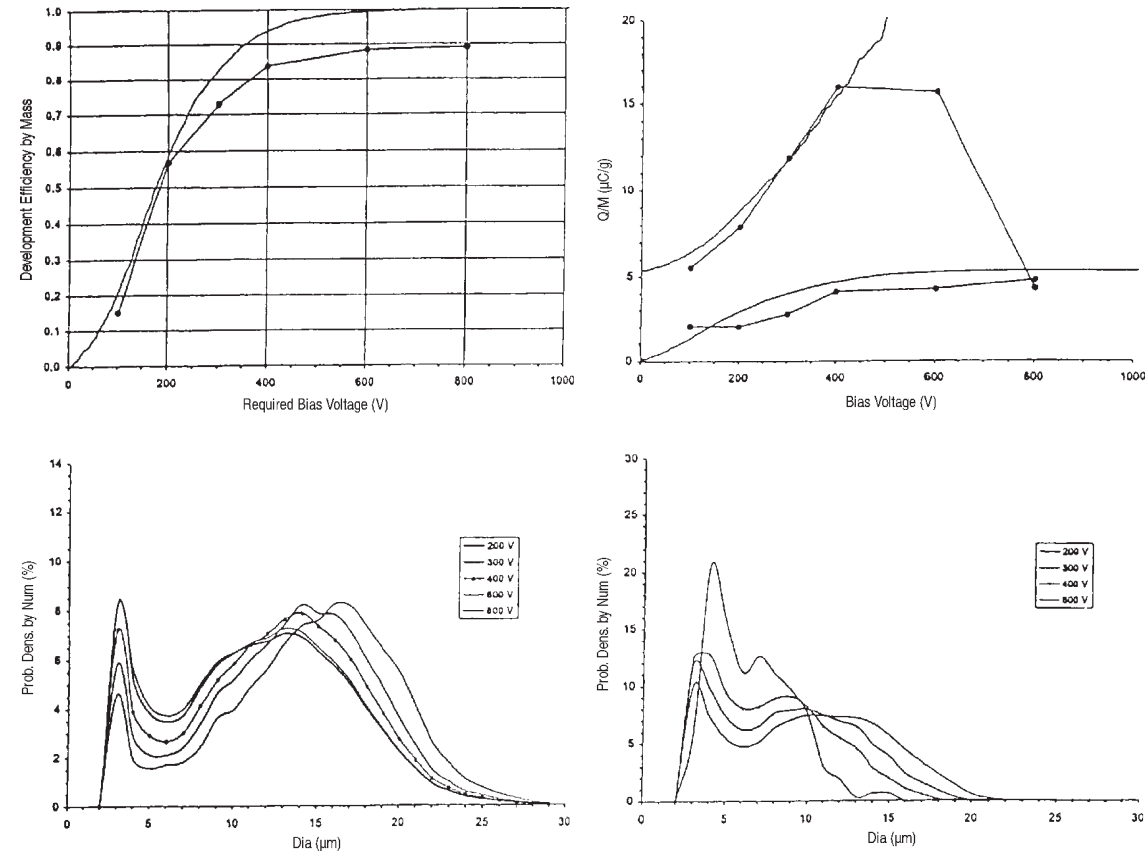

Figure 7. Fit to the data assuming a uniform $n_p = [1 + Rnd\ (1.8)]$. Zero van der Waals force is assumed.

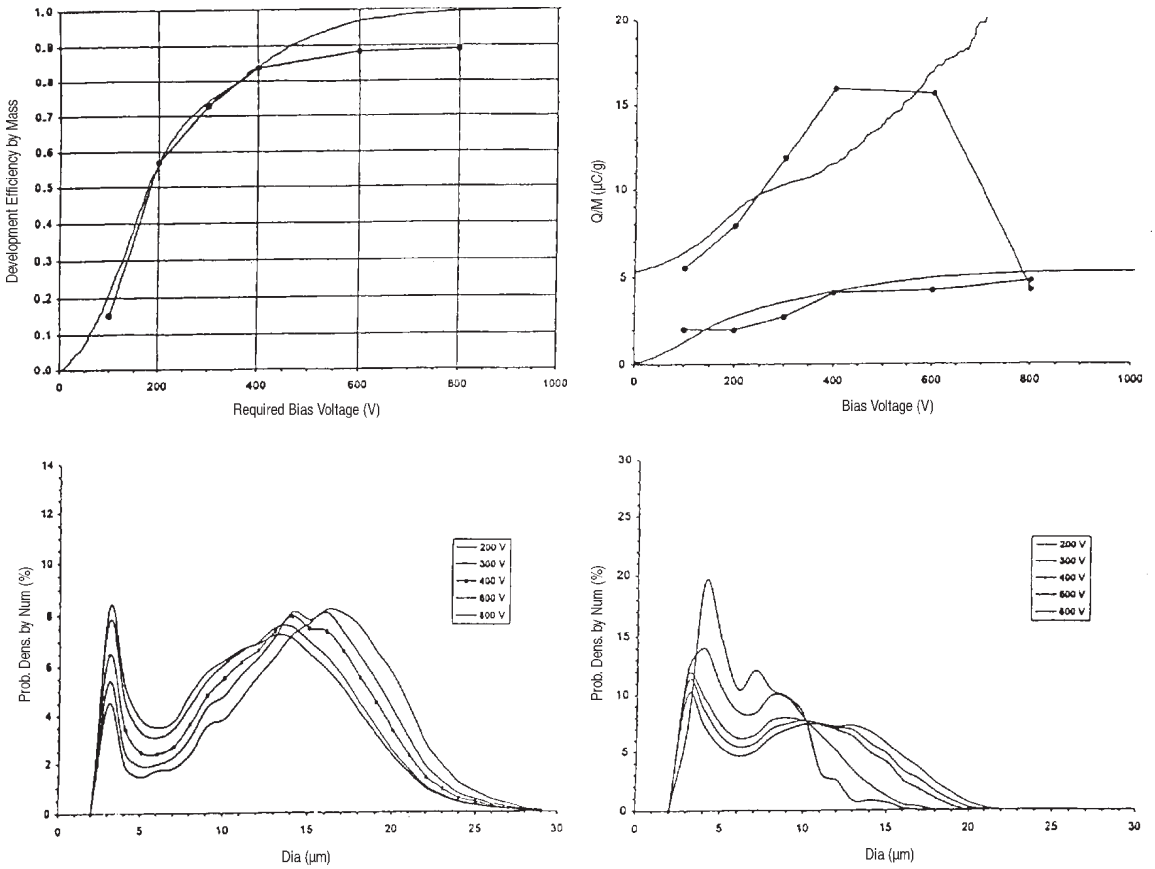

Figure 8. Fit to the data assuming n_p = [1 + *Rnd* (1.8)], [1 + *Rnd* (3.2)], and [1 + *Rnd* (3.2)] in three regions (see text). Zero van der Waals force is assumed.

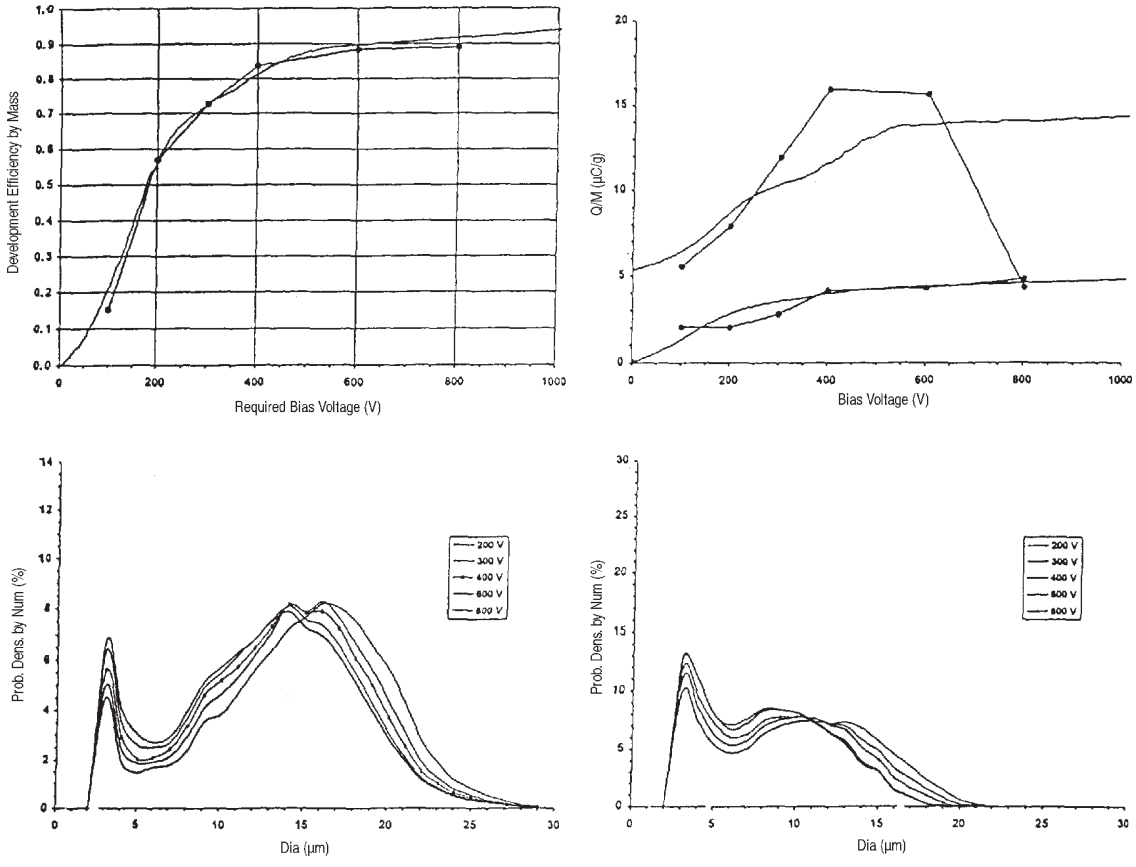

Figure 9. Fit to the data assuming n_p = [1 + *Rnd* (1.8)], [1 + *Rnd* (3.2)], and [1 + *Rnd* (9)] in three regions (see text).

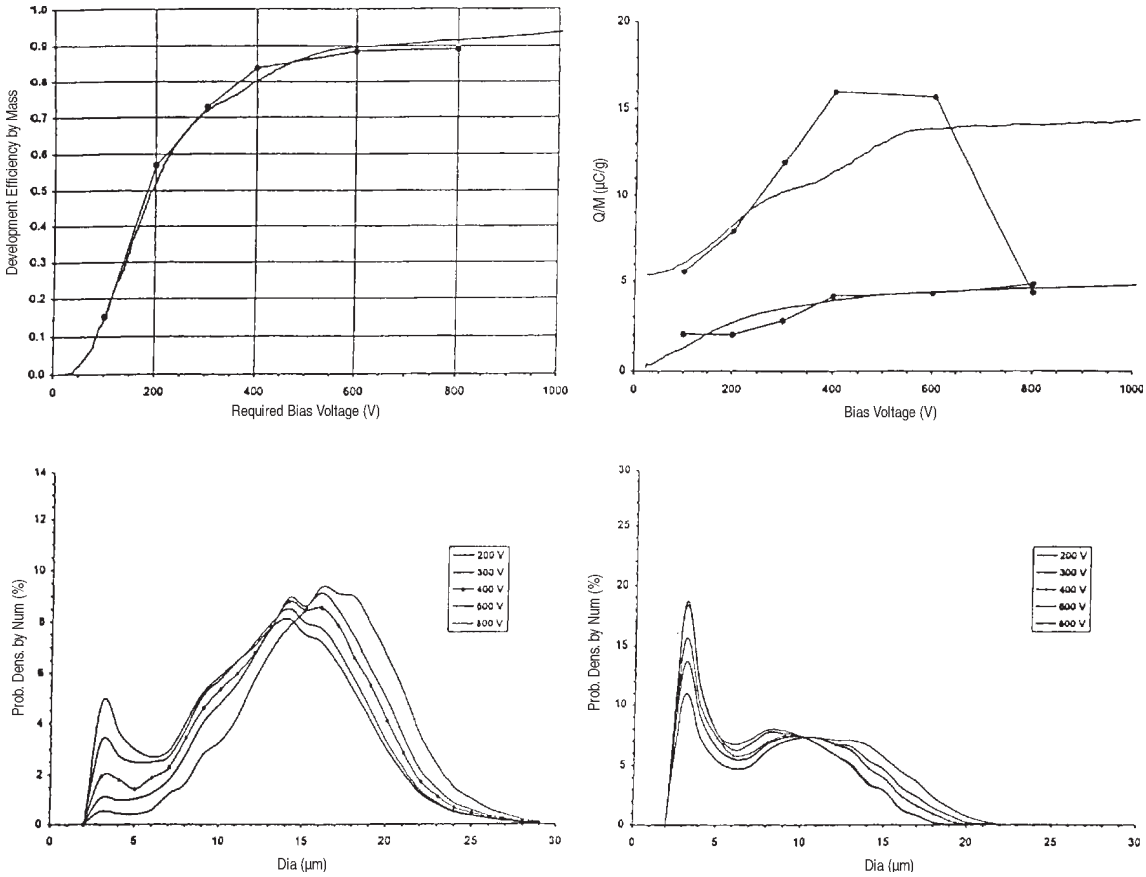

Figure 10. Fit to the data assuming both van der Waals (n = 0.01 + *Rnd* 0.4) and n_p = [1 + *Rnd* (1.8)], [1 + *Rnd* (3.2)], and [1 + *Rnd* (9)] in three regions (see text).

adhesion reported in the literature, the wide width of the observed toner adhesion distributions, and the dependence of toner adhesion on Q/M.

To test this theory experimentally, it has been compared to electric field detachment data for a 16 μm diameter ground toner with silica additives on the surface. The data include developed and residual M/A, Q/M and size distributions The theory is fit to the complete curves of these data sets, a much more stringent test of an adhesion theory than has been presented before, to the author's knowledge (usually only one point, the 50% development efficiency value is fit). The agreement between theory and experiment is excellent over all of these curves with one set of parameters, all of which turned to be reasonable:

- the A and B parameters, which characterize the charge distribution, are highly constrained by the requirement that the charge distribution be similar to published curves and the charge distribution give the measured Q/M.
- The value of the van der Waals force is small, which results directly from the observation of a finite but small intercept (consistent with conclusions reached by previous workers,[4] who noted low intercepts when the adhesion force was plotted versus $(Q/M)^2$). The result that the van der Waals force is small is almost surely due to the addition of silica to the surface of the toner.
- n_p for most of the toner was between 1 and 2.8, which can be rationalized as follows: as 3 points define a plane, most toner is balanced on the planar conductive surface at three points, probably three silica particles. Some, however are balanced against neighboring toner, so n_p is distributed between 1 and 3 (actually 2.8).

We have pointed out that the adhesion and the ratio of the observed adhesion force of this toner to theory is the lowest ever reported and have argued that the physical significance of n_p suggests that the electrostatic adhesion force has been minimized. (4) About 15% of the toner has much higher adhesion, as might be expected of a ground, manufactured toner.

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