Effects of Carbon Black on Toner Tribocharging in Two-Component Electrophotographic Developers

J. H. Anderson▲

Heidelberg Digital, LLC. Rochester, New York

Carbon black is the most widely used pigment for black electrophotographic toners. In this article the effects of carbon black on tribocharaging of two-component developers reported in the published literature are reviewed and compared to experemental work done recently. Carbon black is found to have the following effects on the triboelectric properties of model toners made from carbon black in a polymer binder: (1) In toners compared at the same carbon black loading, toners containing carbon blacks with greater work function (or surface acidity) charge more negatively. (2) Increasing the carbon black content causes the absolute value of the charge-to-mass of model toners to decrease when the toner is charged either positively or negatively using poly(methylmethacrylate) coated carrier or poly(vinylidine fluoride) coated carrier. (3) Q/m when the toner is charged negatively is a linear function of q/m when the same toner is charged positively. (4) Increasing the carbon black content causes the absolute value of the slope and the intercept of linear plots of mass-to-charge as a function of the mass ratio of toner to carrier to increase. A steady-state version of the surface state model of tribocharging is developed and compared to experimentally observed tribocharging behavior. In this model, the rate of charging of the toner and the rate of discharging of the toner due to increased conductivity related to the carbon content of the toner are assumed to be equal at steady state. The model predicts all four of the observed behaviors.

Introduction

Black electrophotographic toners most frequently contain 5 to 10% of carbon black as a pigment in a polymer matrix. Despite the extensive application of black toner, only a few studies have appeared in the published literature describing the influence of carbon black on the triboelectric properties of black toners. The results presented in these studies differ widely. The purpose of this report is to survey the published work and to compare it to new experimental work in order to understand better the role of carbon black in tribocharging of two-component electrophotographic developers. As a part of this study, a model is presented which is consistent with the majority of the experimental results that were reviewed.

The early work of Julien is the most often quoted study of the influence of carbon black on tribocharging of toners.[1] He made toners consisting of polymer and carbon black only. He used two different polymer matrices, a polyester and an epoxy. He selected a variety of carbon blacks which had different levels of surface acidity and characterized them by measuring their contact potential relative to a gold reference. He found a linear relationship between charge to mass, q/m, and contact potential. More positive contact potentials gave more positive q/m. Pethrick and co-workers observed behav-

ior similar to Julien.[2] They deliberately oxidized the surface of Regal 300 carbon black, a neutral carbon, to produce carbon blacks of varying surface acidity. They found that q/m of toners made from these carbon blacks became more negative as the degree of oxidation increased. Fabish and Hair have shown that the work function of carbon black is related to the degree of oxidation of the carbon black surface. More oxidized surfaces are more acidic and have a higher work function.[3] Duke and Fabish also report that addition of an acidic carbon black to a styrene/methylmethacrylate copolymer (15/85) caused q/m to become more negative.[4]

Gutman and Hartmann interpreted Julien's data in terms of the "high density of states limit" of a surface state model of tribocharging originally developed by Kondo.[5,6] As charge is exchanged, an electric potential, V, develops between the toner and the carrier particles. Charge exchange continues until V becomes equal to the difference between the work functions of the two surfaces, $\phi_t - \phi_c$. Anderson and Castle and Schein have shown that, in the high density of states limit, the reciprocal of q/m, m/q, can be represented as:[7,8]

$$m/q = -\frac{d}{3\varepsilon_0(\varphi_t - \varphi_c)} \left[r_t \rho_t - T/C \bullet r_c \rho_c \right] \quad (1)$$
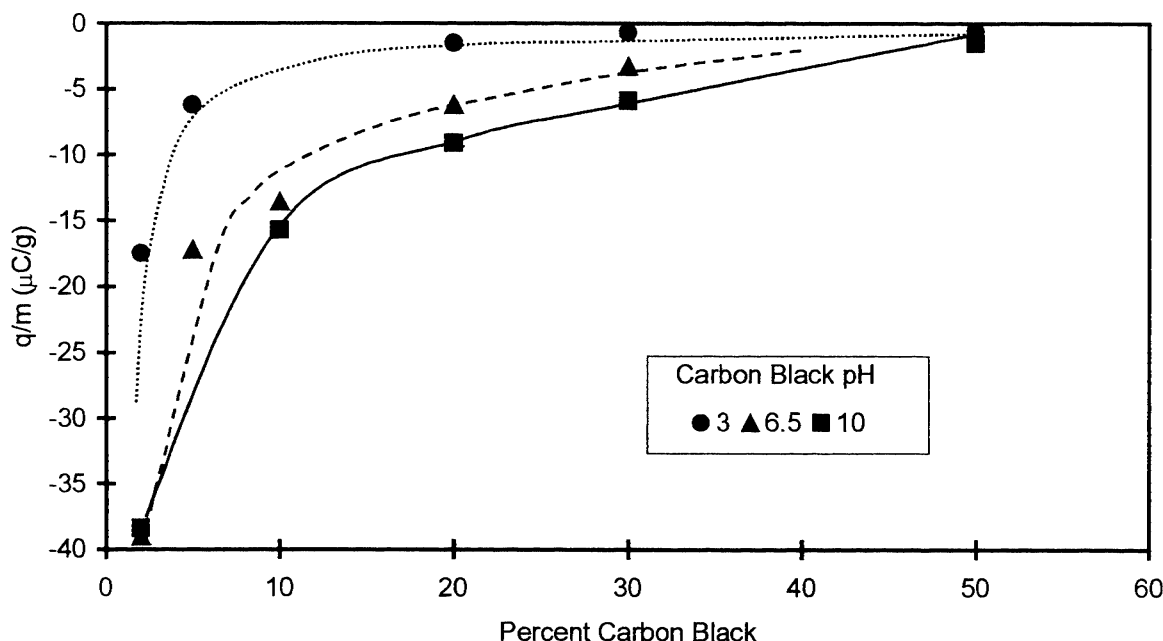
In Eq. 1, d is a characteristic separation distance between the particle surfaces during charging, ε_o is the permittivity of free space, r is the particle radius, T/C is the mass ratio of toner to carrier in the developer, and ρ is the mass density of the particles. The subscripts, t and c refer to the toner and carrier respectively. Equa-

Figure 1. Charge to mass measurements of Matsui and Oka.[10] These workers examined both the effect of the surface acidity of carbon blacks and their concentration on q/m of toners. In this figure, taken from their study, it can be clearly seen that increasing the carbon content of the toner causes q/m to move toward zero. The effects of acidity of the carbon black are similar to those found by Julien.[1]

tion 1 predicts that m/q is a linear function of T/C. (Plots of m/q versus T/C are referred to below as "TC series plots.") This prediction has been thoroughly verified for fully charged mixtures.[7,8] This and other confirmations of the surface state model have recently been reviewed by Schein.[9]

The terms in the square brackets are determined only by the physical characteristics of the toner and the carrier, i.e., particle size and mass density. Thus, in this charging model, the only way in which the chemical composition of the toner affects q/m is through the work function of the toner. The work function of the toner depends in some way upon the carbon black concentration. Gutman and Hartmann assumed that the work function was a linear combination of the work function of the carbon black and the toner polymer:[5]

$$\phi_t = w_{CB} \bullet \phi_{CB} + w_P \bullet \phi_P \qquad (2)$$

In Eq. 2, w is the mass fraction of a component in the toner. The subscripts CB and P refer to carbon black and the toner polymer respectively. If $\phi_{CB} > \phi_P$, then the toner charge will become more negative, while if $\phi_{CB} < \phi_P$, then the toner charge will become less negative. This is consistent with Julien's observation that more electronegative carbon blacks (higher ϕ_{CB}) caused the toner to charge more negatively.

Gutman and Hartmann also noted that the carbon black content of the toner will have no effect on the toner charge if $\phi_{CB} = \phi_P$.[5] However, if $\phi_{CB} \neq \phi_P$, then q/m should depend upon the weight fraction of carbon black in the toner. They tested this hypothesis experimentally. They made two series of toners using an *n*-butyl methacrylate polymer and two different carbon blacks. One black was unoxidized and had approximately the same work function as the polymer. The other was an oxidized black with a work function more negative than the polymer. They found that the concentration of the first black had

no effect on the toner charge, while the charge of the toner containing the second black became more negative as the concentration increased. Above about 15 wt%, q/m leveled off. They interpreted this latter behavior as resulting from high electrical conductivity of the toner at the higher carbon concentrations.

These experiments and their interpretation by Julien, Gutman and Hartmann suggest that the main influence of carbon black in toners is through their work function. Other workers, however, have reported different experimental results. Matsui and Oka made styrene-acrylic based toners of varying carbon black content from three carbon blacks of widely different degrees of acidity.[10] They measured q/m of these toners against a metallic shot carrier, which charged the toners negatively. They found that in all cases, contrary to Gutman and Hartmann and Duke and Fabish's results, q/m became less negative as the carbon black concentration increased (see Fig. 1, taken from their study.) At any particular carbon black loading, for example 10% carbon black, q/m is most negative for the acidic carbon black, intermediate for the neutral black, and least negative for the basic black. This order is in agreement with the interpretation of Julien and Gutman and Hartmann.[1,5] However, the effect of carbon concentration on q/m is not.

Matsui and Oka interpreted their results using a kinetic model rather than the equilibrium model used by Gutman and Hartmann.[5,10] As with the "high density of states limit" of surface state models, they assumed that the driving force for exchange of charge is the difference in work functions between the toner and the carrier. In addition they assumed that the maximum charge achieved during mixing of toner and carrier is a steady state, not an equilibrium. In their view, the charge is determined by a balance between the rate of charging and the rate of loss of charge due to the low resistance of the toner caused by the presence of the carbon black:

Charging rate =
$$k_c (\phi_c - \phi_t) = \text{Discharge rate} = (q/m)/\tau \qquad (3)$$

where k_c is the rate constant for charging and τ is the time constant for discharging due to electrical conduction. Therefore, at steady state

$$q/m = k_c \tau (\phi_c - \phi_t) \qquad (4)$$

Equations 3 and 4 predict:
1. Because discharge becomes more rapid (i.e., t decreases) as more carbon is added to the toner, q/m will decrease as the carbon black concentration in the toner increases.
2. At the same carbon black concentration, toners made from carbon blacks with higher work functions (higher surface acidity) will charge more negatively.

Thus, Equations 3 and 4 are in agreement with their experimental results and with the results of Julien. Equation 4 is not in agreement with Eq. 1, however, because Eq. 4 predicts that q/m should be infinite for a perfectly insulating toner. This is because Matsui and Oka neglected the effects of the electric potential generated by the transfer of charge that limits q/m. Thus, their model is correct only for cases where the electric potential is negligible compared to $(\phi_c - \phi_t)$.

Extension of the Surface State Model

In this section, the high density limit of the surface state model as used by Gutman and Hartmann[5] is combined with the steady-state charging model of Matsui and Oka.[10] The effects of electric potential generated by exchange of charge are included.

The rate of charge generation, dQ/dt can be written as a balance between charged species moving between the toner and the carrier over an energy barrier and ohmic back-flow of the charged species:

$$\frac{dQ}{dt} =$$
$$-e\beta \exp\left[-\frac{(\varepsilon - \varphi_t)}{kT}\right] + e\beta \exp\left[-\frac{(\varepsilon - \varphi_c) + V}{kT}\right] - V\sigma \qquad (5)$$

In Eq. 5, Q is the charge transferred to a single carrier particle: t is the time, β is a kinetic parameter, ε is the height of the energy barrier to charge transfer, ϕ is the work function, V is the potential generated by the transfer of charge between the toner and the carrier, σ is the conductivity of the contact ($1/\sigma$ is the resistance to back-flow of charge), k is Boltzmann's constant and T is the absolute temperature. The subscripts, t and c, refer to toner and carrier respectively.

At steady state, $dQ/dt = 0$, and

$$V\sigma = -e\beta \exp\left[-\frac{(\varepsilon - \phi_t)}{kT}\right] + e\beta \exp\left[-\frac{(\varepsilon - \phi_c + V)}{kT}\right] \qquad (6)$$

In the high density of states limit of the surface state model, V is the electrical potential between the toner and the carrier as a result of the transfer of charge and is given in Ref. 7 as:

$$V = -\frac{d}{3\varepsilon_0} \frac{q}{m} \left(r_t\rho_t + \frac{T}{C} r_c\rho_c\right) \qquad (7)$$

Equation 7 can be substituted into Eq. 6 and rearranged:

$$\frac{q}{m} = \frac{3\varepsilon_0 e\beta'}{d\sigma\left(r_t\rho_t + \frac{T}{C} r_c\rho_c\right)} \bullet \left\{-\exp\left(\frac{-\phi t}{kT}\right)\right.$$
$$\left. + \exp\left[-\left(\phi_c - \frac{q}{m} \bullet \frac{d}{3\varepsilon_0} \bullet \left(r_t\rho_t + \frac{T}{C} r_c\rho_c\right)\right)\bigg/kT\right]\right\} \qquad (8)$$

In Eq. 8 the charging rate, $e\beta \exp[-\varepsilon/kT]$, has been replaced by β'. This equation can be solved for q/m numerically.

However, in order to get a clearer picture of the behavior of q/m as a function of σ and T/C, the exponential expression can be approximated by the first two terms of their series expansion. Solving for q/m:

$$\frac{q}{m} \cong -\frac{3\varepsilon_0}{d}\left(\frac{\phi_t - \phi_c}{r_t\rho_t + \frac{T}{C} r_c\rho_c}\right) \bullet \left(\frac{1}{\frac{\sigma kT}{e\beta'} + 1}\right) \qquad (9)$$

Numerical calculations confirm that charging behavior predicted by Eq. 9 is similar to that predicted by Eq. 8. The predicted q/m values differ by less than a factor of 2 when $\sigma kT/e\beta' \ll 1$ and converge as $\sigma kT/e\beta'$ increases.

Equation 9 is the reciprocal of Eq. 1, the high density of states limit of the surface state model, except for the factor $1/(\sigma kT/e\beta' + 1)$. In the limit of $\sigma kT/e\beta' \ll 1$, Eq. 9 gives the same relationship between q/m and T/C as the high density of states limit of the surface state model, Eq. 1. That is, m/q is a linear function of T/C, and the slope and intercept depend only on the particle sizes and mass densities. This is to be expected since large values of $\sigma kT/e\beta'$ means that there is no back-flow of charge. As $\sigma kT/e\beta'$ increases but is still smaller than 1, q/m will decrease as linearly as $1 - \sigma kT/e\beta'$. In the limit of $\sigma kT/e\beta' \gg 1$, Eq. 9 has the same form as Matsui and Oka's model, Eq. 4. That is, q/m is proportional to $\sigma kT/e\beta'$. Because $\sigma kT/e\beta'$ is the quotient of a parameter related to charging rate and a parameter related to the back-flow of charge, it is similar to the factor $k_c\tau$ in Eq. 4. When $\sigma kT/e\beta' \gg 1$, Eq. 9 predicts the same charging behavior as Eq. 4.

In addition, Eq. 9 predicts a behavior not predicted by Eq. 4, namely that as carbon black concentration increases, the absolute value of the slope and intercept of the TC series plots should increase. This prediction is shown to be correct by the experimental results presented below.

If two different carriers of the same particle size are used to charge the same toner (one positively and the other negatively), the relationship between the charges is given by

$$\left(\frac{q}{m}\right)^- = \frac{\phi_t - \phi_c^+}{\phi_t - \phi_c^-}\left(\frac{\frac{\sigma kT}{\beta_+'} + 1}{\frac{\sigma kT}{\beta_-'} + 1}\right)\left(\frac{q}{m}\right)^+ \cong$$
$$\frac{\phi_t - \phi_c^+}{\phi_t - \phi_c^-}\left(\frac{\beta_+'}{\beta_-'}\right)\left(\frac{q}{m}\right)^+ \qquad (10)$$

In Eq. 10 the "+" and "−" signs refer to carrier which charges the toner positively and negatively, respectively.

Eq. 10 predicts that, when $\sigma kT/e\beta' \gg 1$ (expression on the far right side of Eq. 10), the ratio of q/m when the toner is charged positively to q/m when the toner is independent of the conductivity, σ. Thus, Eqs. 9 and 10 predict:

1. Toner concentration plots are linear regardless of the polarity of q/m or the magnitude of the conductivity parameter, σ.
2. At the same concentration in the toner, carbon blacks with larger work functions (higher surface acidity) cause q/m to be more negative.
3. As $\sigma kT/e\beta'$ increases, the absolute value of the slope and intercept of TC series plots increases.
4. As $\sigma kT/e\beta'$ increases, q/m decreases. Thus, the presence of carbon black in toner should cause a decrease in q/m, if it causes an increase in conductivity.
5. The relationship between $(q/m)^-$ and $(q/m)^+$ is linear with a negative slope and zero intercept if the work functions of the toners are independent of carbon content.

Experimental

A series of toners was made at different concentrations of Black Pearls 430 carbon black in a styrene-butylacrylate copolymer (75/25) matrix. Carbon black was melt-compounded in the polymer at 150°C. The toner was ground to approximately 10 µm volume average particle size. Ten toners were made ranging in carbon content from 0 to 40 wt%.

These toners were charged against two carriers. The first was a poly(vinylidine fluoride) coated iron-strontium ferrite (30 µm average volume diameter). The second carrier was made from the same ferrite but coated with poly(methylmethacrylate). The former carrier caused the toners to charge positively, while the latter caused them to charge negatively.

The toners were mixed with the carriers at a series of toner concentrations ranging from a toner-carrier mass ratio (T/C) of 0.05 to 0.30. The mixtures were shaken for two minutes in a wrist shaker and then mixed for 30 seconds in a magnetic stirrer at 2000 rpm. This time was determined to be sufficient to bring the toner charge to its maximum value. The toner charge-to-mass ratio, q/m, was determined by removing the toner electrostatically.[7] TC series plots (m/q versus T/C) were then made for each toner and carrier combination. All toner concentration series plots were linear. All but one of those measured using the PMMA-coated carrier had correlation coefficients greater than 0.95. Seven of the ten series measured using the poly(vinylidine fluoride)-coated carrier had correlation coefficients of 0.89 or greater.

Bulk DC conductivity was measured using a Keithly 617 Electrometer provided with an external variable voltage supply to generate voltages up to 2kV. Current response as a function of time was measured during application of the electric field of constant magnitude. The value of the current was recorded at a time, t_m, where $t_m \gg \varepsilon_0 \varepsilon_r \rho$. ε_r is the effective relative permittivity of the material, and ρ is the resistivity of the toner.[11]

Results and Discussion

The electrical conductivity of polymer-carbon black composites depends strongly on the concentration of carbon black. At low carbon black loading, the carbon particles are separated from each other by the polymer and the conductivity of the composite is essentially that of the polymer matrix. As the carbon black loading increases, the particles come closer together and quantum mechanical tunneling of electrons can occur between particles causing the conductivity to increase. At still higher concentrations, the carbon particles are numerous enough that there is a continuous contacting network of carbon particles. This point is referred to as the percolation limit. At concentrations higher than the percolation limit, the network of carbon particles becomes more interconnected, and the conductivity increases until it approaches that of pressed pellets of pure carbon black, of the order of 10^3 S/cm.[12]
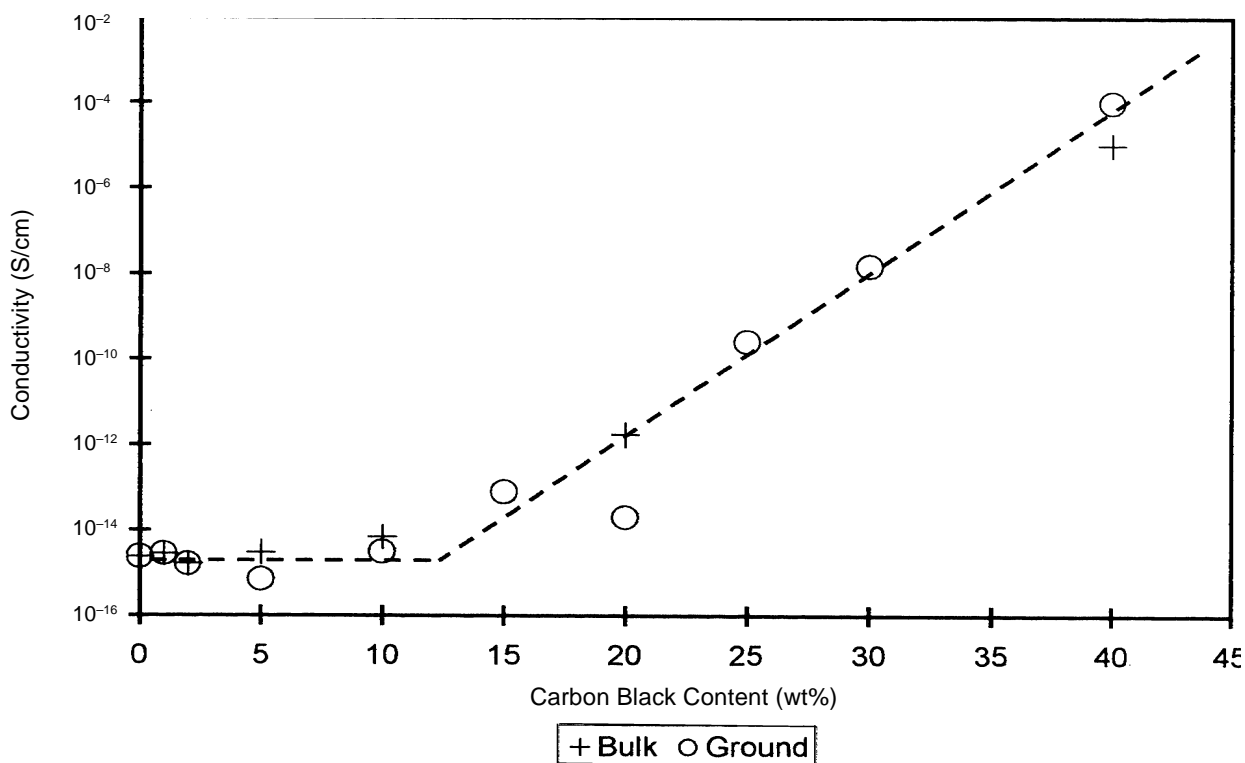
Figure 2 shows the DC conductivity of the toners used in this study as a function of carbon black concentration. The data show that bulk conductivity begins to increase when the carbon concentration exceeds about 10 wt %. Thus, the percolation limit for these toners is above 10% carbon black loading. Below this level, carbon black particles or clusters of particles are separated widely enough that they do not contribute to the bulk electrical conductivity. Above this level, the number of conductive pathways increases, and the conductivity increases.

Figure 3 shows the toner concentration series for a representative subset of the toners charged positively against the poly(vinylidine fluoride)-coated carrier. Figure 4 is similar to Fig. 3 but for a subset of the toners charged negatively against PMMA-coated carrier. All of the plots are linear. The absolute values of the slopes and intercepts of the TC series plots increase as the carbon black concentration is increased.

Figure 5 is a plot of q/m all ten toners charged negatively as a function of q/m of the same toners charged positively. The q/m values were interpolated to $T/C = 0.15$ using least squares linear fits to TC series. There is an excellent linear correlation between the positive and negative q/m measurements. The slope is –0.63. The intercept is nearly zero as expected from Eq. 10 if the work function of the toner did not change substantially as a function of carbon black concentration (prediction 5 of the extended model).

Figure 6 shows q/m values interpolated to $T/C = 0.15$ using least squares linear fits to TC series as a function of carbon black concentration for the toners charged both positively and negatively. The absolute values of q/m in either polarity decreased as the carbon content increased. The charging behavior of toners with carbon black loading below the percolation limit is particularly interesting, because typical commercial toners contain 5 – 10 wt% carbon black. At 1.0 wt% loading, $|q/m|$ is less than for the polymer alone. At 10 wt% carbon loading, $|q/m|$ is only about 40% of $|q/m|$ of the unpigmented polymer. Thus, at loading below the percolation limit, carbon black has a major effect on charging even though it does not affect bulk conductivity.

There is not a sharp break in charging behavior corresponding to the sudden onset of electrical conductivity at the percolation limit. Carbon black concentrations below that necessary to influence the bulk electrical conductivity of the toner have a significant effect on q/m, and toner with carbon content above the percolation limit still retains a charge. This is puzzling at first. One explanation for this behavior is that localized clusters of carbon black particles form even at low carbon black concentrations. Regions of locally high carbon content can give rise to local regions of higher conductivity that are electrically isolated from one another by the polymer. Thus, the local conductivity of the toner may be high at some points of contact between the toner and the carrier, even though bulk conductivity of the toner

Figure 2. The measured bulk electrical conductivity of the toners consisting of carbon black and polymer only. Below about 10%, carbon black content has no effect on bulk conductivity. At higher carbon black concentrations, the conductivity increases markedly. Triboelectric changes, on the other hand, are observed even at the lowest carbon black concentrations.
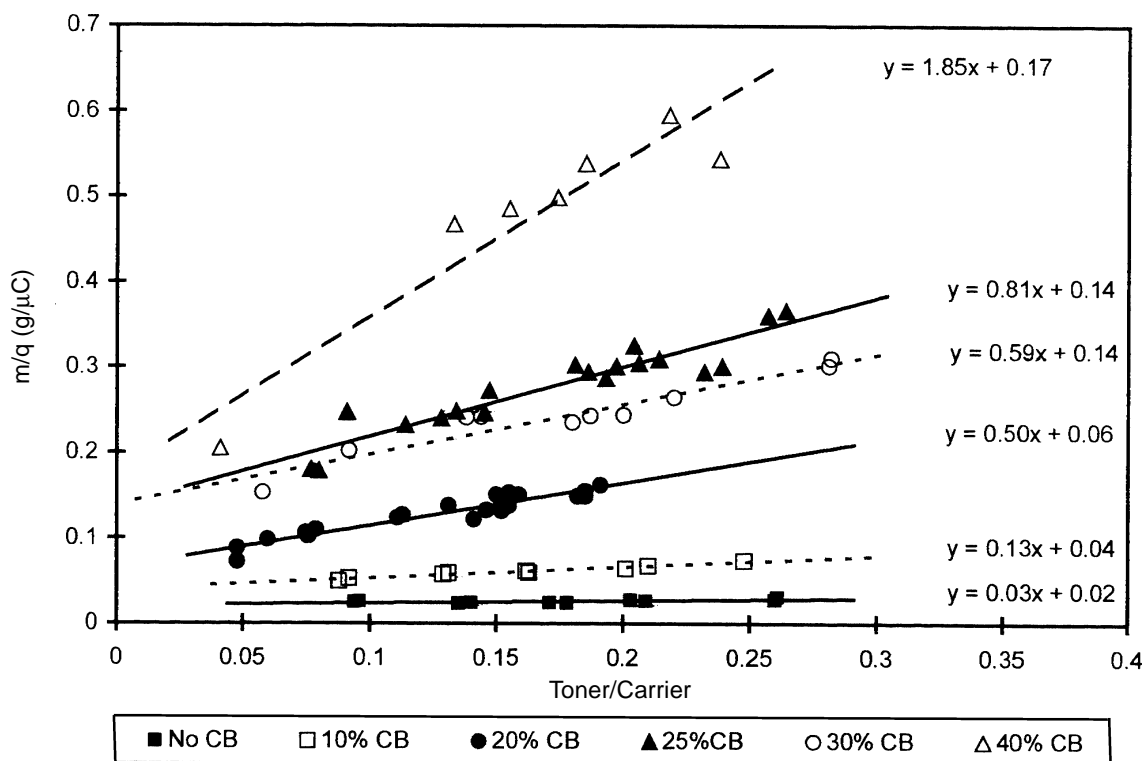

Figure 3. A representative set of T/C series plots. These toners charged positively against the poly(vinylidine fluoride)-coated carrier in agreement with prediction 1 of the model presented above. The plots are linear in agreement with prediction 2 of the model. As the carbon concentration increased, m/q of the toner increased. This behavior is in agreement with prediction 3 of the model.
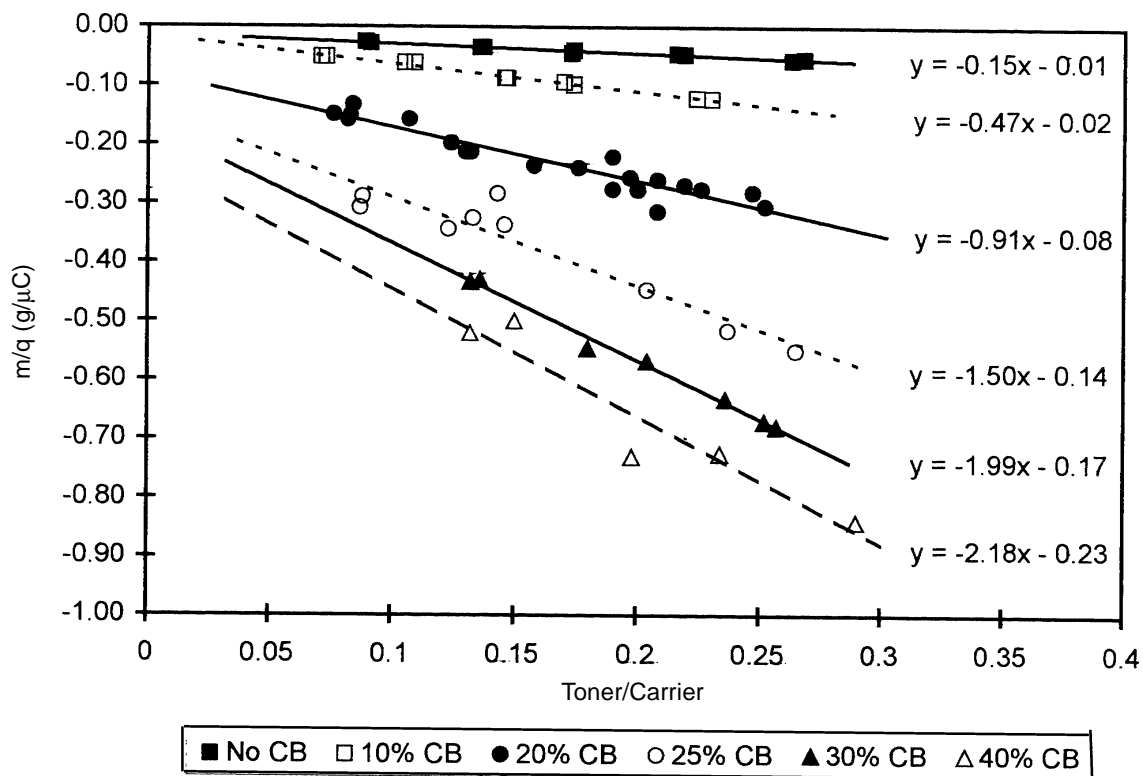
Figure 4. A representative set of T/C series plots for toners charged against the PMMA-coated carrier. The toners charged negatively against this carrier in agreement with prediction 1 of the model. The plots are linear in agreement with prediction 2, and as the carbon concentration increased, the absolute value m/q increased as predicted by the model.
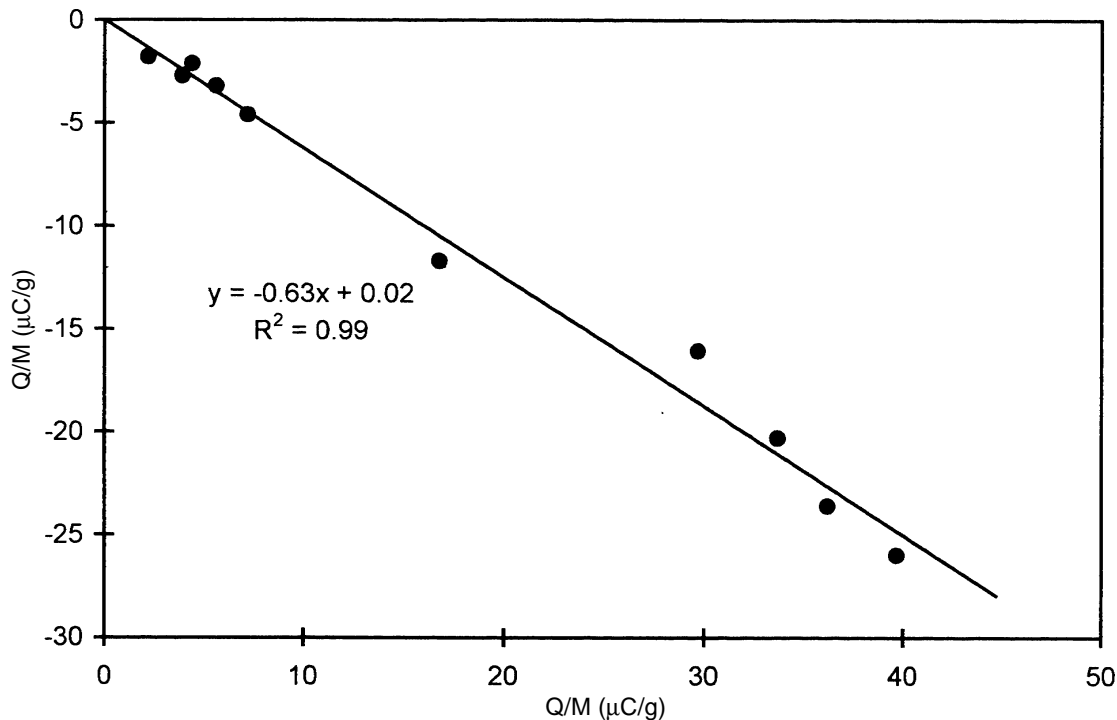
Figure 5. Q/m at T/C = 0.15 of all ten toners charged negatively as a function of q/m of the same toners charged positively. The q/m values were interpolated to T/C = 0.15 using least squares linear fits to TC series. As predicted by the extended model, there is an excellent linear correlation between the positive and negative q/m measurements. The slope is negative, and the intercept is nearly zero in agreement with prediction 5 of the model.
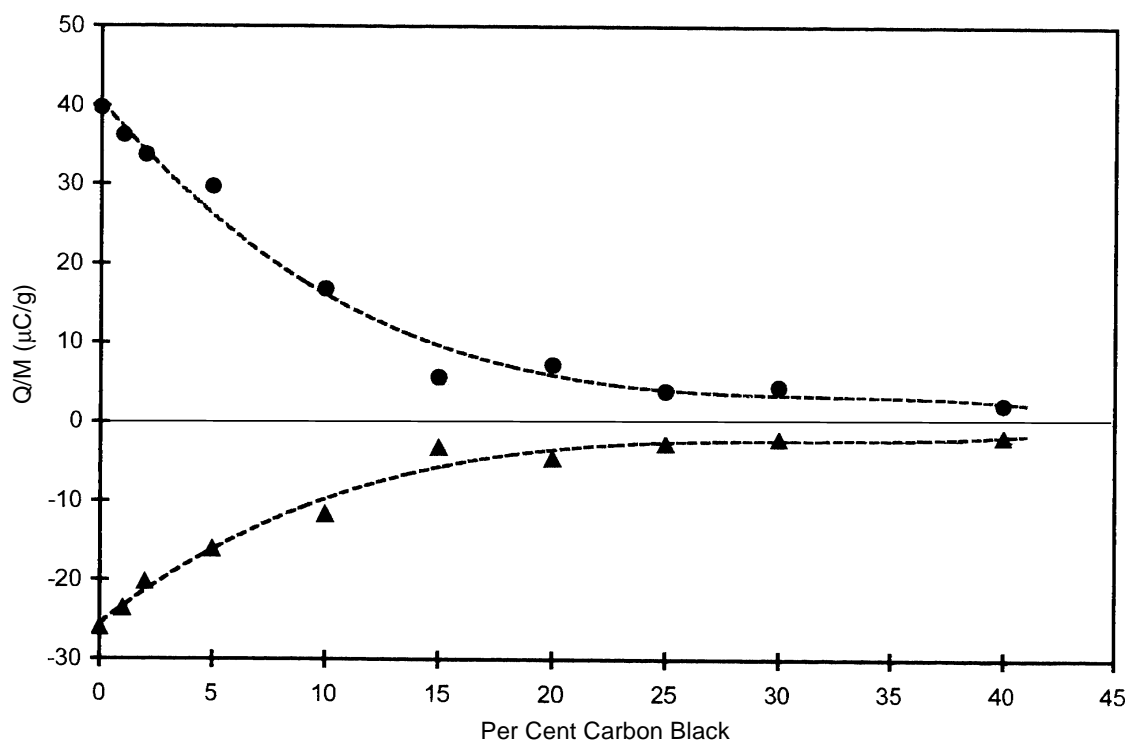
Figure 6. Q/m values interpolated to $T/C = 0.15$ using least squares linear fits to TC series as a function of carbon black concentration for the toners charged both positively and negatively. The absolute values of q/m in either polarity decreased as the carbon content increased. The decrease of $|q/m|$ at carbon black concentration below 10 wt% may result from local areas of high conductivity related to clustering of the carbon black. Non-zero charges for toners with carbon content above the percolation limit may result from the presence of areas on the toner surface with local carbon content below the percolation limit that can retain charge like an insulating toner.

is low. These areas would have lower charge density than insulating regions of the toner surface. At the percolation limit and above, continuous interconnecting networks of carbon black particles exist, giving rise to increased bulk conductivity. As more carbon is added, interconnected pathways proliferate, and the bulk conductivity increases. However, even above the percolation limit there may be local regions that are not conductive. These regions behave like an insulating toner and can retain charge without it leaking back to the carrier. Thus, there can be loss of charge at carbon levels below the percolation limit; there need not be a sudden, dramatic drop in q/m at the percolation limit, and toner with carbon content above the percolation limit can still charge.

Conclusions

Matsui and Oka's charging model has been extended to include the toner concentration dependence of tribocharging and the charge-limiting effects of electric fields generated by charge transfer. The published data and the charging measurements presented in this report are generally in agreement with the predictions of the extended model: The work of Julien and subsequent analysis by Anderson show that carbon blacks with larger work functions caused toner to charge more negatively (prediction 2).[1,13] In this study, m/q was found to be linearly related to toner concentration at all levels of carbon concentration and for toners of a wide range of conductivity (prediction 1). The absolute values of the slopes and intercepts of the linear relationship between

m/q and T/C increased as the carbon concentration of the toner was increased (prediction 3). Whether the toner was charged positively or negatively, the absolute value of q/m decreased as the carbon content and conductivity of the toner was increased (prediction 4). Finally, q/m of toners charged negatively was linearly related to q/m of the same toners charged positively. The intercept was near zero indicating that the work function of the toners in this study was not greatly affected by the presence of the carbon used to fabricate the toners (prediction 5).

Some of the results of Gutman and Hartmann[5] and of Duke and Fabish[4] differ from the results of Matsui and Oka[10] and the experiments reported in this investigation, however. Gutman and Hartmann report experiments in which acidic carbon black causes q/m to become more negative when added at concentrations below the percolation limit. Above the percolation limit, the magnitude of the charge levels off and begins to decrease. Gutman and Hartmann also report a case in which addition of carbon black has no effect on q/m. They suggest that this is because the work functions of the carbon and the polymer matrix are the same and that carbon black at concentrations below the percolation limit does not contribute to conductivity of the toner. These results are at odds with the results reported in this article where even small amounts of carbon black cause $|q/m|$ to decrease. This suggests that there may be less clustering of the carbon black particles in their experiments than in those reported here. It would be interesting to see if the addition of a carbon black, with the same work function as the polymer matrix at concentrations above the percolation limit, would cause $|q/m|$ to decrease.

The model, represented by Eq. 9 and Eq. 10, may have another application. It is often said that high relative humidity causes tribocharging to be reduced because water adsorbed on the contacting surfaces leads to increased surface electrical conductivity. If this is so, then Eq. 9 predicts that the absolute values of both the positive and negative tribocharge should be reduced. Furthermore, the absolute values of the slopes and intercepts of TC series plots using carriers that charge the same toner positively and negatively should increase. This prediction should be tested experimentally. ▲

References

1. P. C. Julien, in *Carbon Black-Polymer Composites*, E. Sichel, Ed., Marcel Dekker, NY, 1982.

2. J. H. Daly, D. Hayward and R. A. Pethrick, *J. Phys. D. Appl. Phys.* **19**, 885 (1986).
3. T. J. Fabish and M. L. Hair, *J. Colloid and Interface Sci.* **62**, 16 (1977).
4. C. H. Duke and T. J. Fabish, *J. Appl. Phys.* **49**, 315 (1978).
5. E. J. Gutman and G. C. Hartmann, *J. Imag. Sci. and Technol.* **36**, 335 (1992).
6. A. Kondo, Triboelectrification Mechanism of Powder Developers, in *Proc. of the Technical Association of the Pulp and Paper Industry, 1980 Printing and Reprography Conference*, TAPPI Press, Atlanta, GA 1980, pp. 153–157.
7. J. H. Anderson, *J. Imag. Sci. and Technol.* **38**, 378 (1994).
8. G. S. P. Castle and L. B. Schein, *J. Electrostatics* **36**, 165 (1995).
9. L. B. Schein, *J. Imag. Sci. and Technol.* **44**, 475 (2000).
10. N. Matsui and K. Oka, *Denshi Shashin Gakkai-shi* [Electrophotography], **27**, 307 (1988).
11. T. Binga, J. H. Anderson, J. Fitzgerald, and J. Glatz-Reichenbach, The Influence of Carbon Black filler on the Molecular Relaxation of a (75/25) Styrene-Butylacrylate Random Copolymer, Presented at Am. Phys. Soc. Spring Meeting, Mar. 22–26, 1993, Seattle, Washington.
12. E. K. Sichel and J. I. Gittleman, *J. Electronic Mats.* **11**, 699 (1982).
13. J. H. Anderson, *J. Imag. Sci. and Technol.* **44**, 129 (2000).