# A Model for the Charging of Fresh Toner Added to a Two-Component Charged Developer

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# Abstract

In this work we extend the Gutman and Hartmann triboelectric charging model for two-component developers to include the case of uncharged toner added to a charged developer (toner and carrier are charged). In the previous work, we examined the toner charging process when uncharged toner is mixed with uncharged carrier to form a developer. Now we examine the toner charging process when uncharged toner is mixed into a charged developer – a process we call toner admix. We will develop a model and compare it to experimental results. A toner charge spectrograph is used to measure the charge in charge of the incumbent toner as well as the charge of the freshly added, uncharged toner.

#### Introduction

Models of the triboelectric charging of two-component developers usually describe the charge on the toner particles after they have been mixed with carrier to form a charged developer<sup>1</sup>. However, in a copier or printer, toner is dispensed or added into a charged developer. The newly dispensed (or added) toner must gain charge from the charged carrier in the presence of the incumbent charged toner. Due to an appreciable interfacial electric field<sup>2</sup>, the boundary conditions for this charging process are not the same as the initial charging of the uncharged toner mixed with uncharged carrier beads.

We have shown that toner particles in non-image areas (or background) on copies or prints are a result of low charge and wrong sign toners in the two-component developer.<sup>3</sup> A possible source of the low charge and wrong sign toners is the slow charging of the freshly dispensed toner into the developer. The charging of the freshly added toner in the developer can be monitored with a toner charge spectrograph<sup>4</sup>.

In the toner charge spectrograph used in this work, the toner particles are carried in a moving column of air through an electric field perpendicular to the direction of airflow. The toner particles are displaced along the direction of the electric field and collected on a filter paper at the bottom of the spectrograph tube. Measurement of the displacement of each toner particle from the point of zero charge determines the charge-to-diameter ratio (q/d); the size (d=2r) of the each toner particle can be also be measured. From these measurements, the charge

distribution (frequency of particles with a given q/d and d) of the toner in the developer can be obtained.

By "tagging" the dispensed or added toner, one can monitor the charging of this toner as well as the charge on the incumbent toner particles. Gutman and Hollenbaugh<sup>3</sup> have discussed the staining of toner particles with a fluorescent dye as a method of tagging toner particles. An alternate method is to use different color toners. In this work, we will use two different color toners to monitor the charging of toner added to a charged developer. Our results are for bench admix measurements in which a toner and carrier are charged by mixing on a roll mill; then, fresh toner is added to this charged developer and followed by further mixing on the roll mill. The start of the admix test, t=0, is when the toner is added to the charged developer. We have found this stress test is similar to a machine stress test in which the toner is continuously dispensed into the developer for a fixed period of time'.

In the next section we develop a model for the charging of fresh toner added to a charged developer. In subsequent sections we describe the bench admix tests and results. Finally, we compare the model and the charge spectrograph results.

# **Admix Model**

From previous work of Gutman and Hartmann<sup>1,2</sup>, the time rate of change in the charge on the carrier beads is

$$\frac{dQ}{dt} = \sum_{i}^{CS} \sum_{k=1}^{n} \sum_{j}^{TS} P_{i} P_{j} \left\{ \frac{\varepsilon_{0} (\mu_{i} - \mu_{j}) a_{ijk}}{ed_{s}} - \varepsilon_{0} Ea \right\} v \Delta_{ij}$$
(1)

where Q is the carrier bead charge, P is the surface coverage of a constituent material,  $\mu$  is the chemical potential of the material, a is the actual or geometrical area of contact,  $d_s$  is the location of the charge from the surface, E is the interfacial electric field, v is the contact frequency, and  $\Delta$  is the joint density of states. Anticipating toner charge measurements using the charge spectrograph, the time rate of change of the charge, q, on the toner particles is

$$\frac{dq}{dt} = \dot{q} = \frac{a_c \varepsilon_0 v\Delta}{e d_s} \left( \Phi_t - \Phi_C \right) + \left( \varepsilon_0 a v\Delta \right) E \qquad (2)$$

where we have used  $\Phi = \Sigma P \mu$  and Q = -nq.

After integrating Eq. 2 and using the boundary conditions that q=0 at t=0, one obtains the average charge

on a toner particle as a function of time. One can divide by the toner mass, m, to obtain the tribo value, q/m, measured in the total blow-off technique; or, dividing by the toner diameter, d, one obtains the average q/d measured by the charge spectrograph technique.

Eq. 1 is quite general; it can be applied to the case of admix by recognizing that the sum over the toner particles now includes two toner populations, the incumbent toner population with n<sub>i</sub> particles and charge, q<sub>i</sub>, and the added toner population with n<sub>a</sub> particles and with charge, q<sub>a</sub>. The start of the admix experiment, t=0, is when the fresh toner is added into the charged developer. Also, there is no restriction on the composition of the added toner, ie,  $\Phi_{added}$  does not have to be the same as  $\Phi_{incumbent}$ . Usually, in copiers and printers the incumbent and added toners are the same; but in the model they are not required to be the same. For an admix experiment Eq. 2 becomes

$$-n_{i}\dot{q}_{i} - \frac{n_{i}a_{c}v\Delta}{ed_{s}}(\Phi_{C} - \Phi_{i}) + n_{i}(\varepsilon_{0}av\Delta)E =$$

$$n_{a}\dot{q}_{a} + \frac{n_{a}a_{c}v\Delta}{ed_{s}}(\Phi_{C} - \Phi_{a}) - n_{a}(\varepsilon_{0}av\Delta)E$$
(3)

In the admix experiment, another toner charge exchange mechanism may be operative, namely, toner charge sharing. This mechanism has been discussed by Julien et al<sup>5</sup>. When there are regions of local conductivity on the toner surface, the added toner gains charge from the incumbent toner, not by triboelectric charge exchange, but, by the incumbent toner sharing its charge as might happen if an uncharged metallic sphere is touched to another charged metallic sphere. The charge exchange process is such as to bring the two metallic spheres to equipotentials. In the toner case, the assumption is that there are localized conductive regions on the toner surfaces, which if contacted together exchange charge to bring the regions to equipotentials<sup>5</sup>. In our model this mechanism is included by a term of the form

$$\varepsilon_0 a v S(q_i - q_a) n_i$$

where *S* is the strength of the interaction, and, due to the local conductive regions many contacts are required to distribute the charge. Charge sharing occurs as long as there is a difference in charge between the two toner populations. Since there is no change in the net charge of the toner population, this effect is not easily detected by the total blow-off technique that measures the average charge for the whole toner population; but, is detectable with a toner charge spectrograph and can be distinguished from triboelectric charging.

Subtracting the charge sharing term from both sides of Eq. 3, one obtains differential expressions for each toner population. These can differ at most by a constant that we will take to be 0. We now have two coupled differential equations that can be integrated. The solutions for  $q_i$  and  $q_a$  are

For the incumbent toner charge:

$$q_{i}(t) = \frac{k_{m}(\Phi_{i} - \Phi_{c})}{k_{u}\left(1 + \frac{C_{0}}{C}\right)C} \left(1 - e^{-k_{u}\left(1 + \frac{C_{0}}{C}\right)C}\right)$$

$$+ \frac{k_{m}C_{a}(\Phi_{i} - \Phi_{a})\left(k_{u}\left(1 + \frac{C_{0}}{C}\right) - \frac{k_{x}}{C_{a}}\right)}{\left(k_{u}\left(1 + \frac{C_{0}}{C}\right)C\right)\left(\frac{k_{x}C}{C_{a}}\right)} \left\{1 + \frac{k_{u}\left(1 + \frac{C_{0}}{C}\right)Ce^{-\frac{k_{x}C_{i}}{C_{a}}} - \frac{k_{x}C}{C_{a}}e^{-k_{u}\left(1 + \frac{C_{0}}{C}\right)C}\right)}{\frac{k_{x}C}{C_{a}} - k_{u}\left(1 + \frac{C_{0}}{C}\right)C}\right\}$$

$$+ q_{i}(t_{0})\left(\frac{C_{i}}{C}e^{-k_{u}\left(1 + \frac{C_{0}}{C}\right)C} + \frac{C_{a}}{C}e^{-\frac{k_{x}C}{C_{a}}}\right)$$

$$(4a)$$

For the added toner charge:

$$q_{a}(t) = \frac{k_{m}(\Phi_{a} - \Phi_{C})}{k_{u}\left(1 + \frac{C_{0}}{C}\right)C} \left(1 - e^{-k_{u}\left(1 + \frac{C_{0}}{C}\right)C}\right)$$

$$+ \frac{k_{m}C_{i}(\Phi_{a} - \Phi_{i})\left(k_{u}\left(1 + \frac{C_{0}}{C}\right) - \frac{k_{s}}{C_{a}}\right)}{\left(k_{u}\left(1 + \frac{C_{0}}{C}\right)C\right)\left(\frac{k_{s}C}{C_{a}}\right)} \left\{1 + \frac{k_{u}\left(1 + \frac{C_{0}}{C}\right)Ce^{-\frac{k_{s}C}{C_{a}}t} - \frac{k_{s}C}{C_{a}}e^{-k_{u}\left(1 + \frac{C_{0}}{C}\right)C}\right)}{\frac{k_{s}C}{C_{a}} - k_{u}\left(1 + \frac{C_{0}}{C}\right)C} \right\}$$

$$+ q_{i}(t_{0})\frac{C_{i}}{C}\left(e^{-k_{u}\left(1 + \frac{C_{0}}{C}\right)C} - e^{-\frac{k_{s}C}{C_{u}}t}\right)}{\frac{k_{s}C}{C_{a}}}$$

$$(4b)$$

where C=nm/M and  $k_m$ ,  $k_u$  and  $k_s$  are constants which are combinations of the factors in Eq. 3. The q/m or q/d can be obtained by dividing by either m or d. These quantities are incorporated in the constant  $k_m$ . As in the earlier work<sup>1,2</sup>, for either the incumbent or added toner, the parameter  $A_0$  is

$$A_0 = \frac{k_m \left( \Phi_{i(a)} - \Phi_C \right)}{k_u} \tag{5}$$

## Experiment

We used commercially available toners in these experiments. Two toners, which were the same size and had the same external additives but slightly different charging properties due to the pigments, were selected; to further accentuate the triboelectric differences, 0.5% carbon black was blended onto the lower-charging-toner to lower the tribo values; the blending was done in a ball mill for 30 minutes. The carrier was polymethylmethacrylate coated grit-shaped iron beads. The ratio of the carrier radius to the toner radius was an important parameter for the model; for these materials the ratio was 10.

The general procedure for an admix test is to prepare a developer by mixing toner with carrier for some period of time, e.g., 30 minutes. At the end of the mixing time the toner charge distribution is measured; this point becomes t=0 for the admix test. Fresh toner is then added into the developer and mixed. Toner charge distributions are measured at convenient times. Specifically, for this work, developers were prepared by mixing 4 g of toner with 100 g of carrier to yield a 4% toner concentration (all "toner concentrations" are the ratio of toner weight to carrier weight). The developers were mixed on a roll mill for 30 minutes to ensure the tribo values were stable. After the 30 minutes of mixing, the toner charge distribution was

measured with a charge spectrograph. Then 2 g of toner were added to the developer. The jar was tumbled end-overend three times and then mixed on a roll mill for two minutes. Samples for charge spectrograph measurements were taken after 15, 30, 45, 60 and 120 seconds of mixing. The spectra were analyzed using a computer driven optical image analyzer to determine the toner charge distribution, i.e., the frequency of particles having a given q/d and toner size, d. From the toner charge distribution the median q/d of the toner was calculated. A 2X2 matrix of admix tests was done with the two toners, T1 and T2, namely, T1+T1, T1+T2, T2+T1 and T2+T2. When two different color toners were mixed together, optical filters were used to separate the two populations of color toners.

In order to verify 1) the toners had different triboelectric properties; and, 2) the tribo blow-off measurement and charge spectrograph measurement were equivalent, the tribo values and q/d values of the developers were measured as a function of toner concentration. For these experiments, a developer was prepared with about 8 g of toner and 100 g of carrier, about 8% toner concentration, and mixed on the roll mill for 30 minutes; after which time the tribo value and median q/d were measured. The developer was then split into two portions. With one portion of the developer, the toner was removed with an air stream and the "detoned" carrier was added back into the other developer portion; this mixture was put on the roll mill for 5 minutes. We found that the toner particles quickly redistribute themselves among all the carrier beads to form a uniform, lower toner concentration. Likewise, the toner has a new, larger charge. The purpose of this procedure is to minimize changes in additive transfer from the toner particles to the carrier beads thus holding the surface composition of the toners and carriers constant for all the toner concentrations. We measured the tribo values and q/d values for final toner concentrations of approximately 8%, 6%, 4% and 2%.



Figure 1. q/m versus q/d for the two toners. The q/m and q/d values were varied by varying the toner concentration.

#### Results

In Fig. 1, we compare the tribo values measured at different TC's, versus the comparable q/d's determined

from the charge spectrograph measurements. The slope of the line in Fig. 1 is  $d/m=3/(2\pi\rho_t r^2)$ . For the toners used in this work, the slope should be  $3.6X10^6$  cm/g; the experimental value of the slope from the data Fig. 1 is  $3.5X10^6$  cm/g. We concluded that devices were functional and the two charge measurement techniques were equivalent.



Figure 2. Inverse of q/d as a function of toner concentration for the two toners. The lines are caluculated from Ref. 2.

In Fig. 2, we show the inverse of q/d as a function of the toner concentration (a plot analogous to m/q versus toner concentration). The lines were fit to the data using one adjustable parameter,  $A_0$ , for each toner and the model from Eq. 2 and Ref. 2. The toners were selected and adjusted to have different  $A_0$  values, which were confirmed by the data.

Figures 3-6 show the admix data for the four cases in the 2X2 matrix: T1+T1, T1+T2, T2+T1, and T2+T2. The median values of q/d from the incumbent and added toner charge distributions for each case are shown as a function of the admix mixing time. The lines are the calculations from the model, Eq. 4a and b; however, the plots neglect to show the curvature due to the exponential terms.



Figure 3. q/d versus time for the T1 incumbent and T1 added toners. The lines are calculated from Eq. 4.

# Discussion

The equations for the charge on the incumbent and added toner show that after sufficiently long mixing times, only the first term in each equation is important. Consequently, the q/d should be proportional to the difference in the  $\Phi$ 's of the toner and carrier. When the added and incumbent toners are the same, the q/d's are the same. However, when the added and incumbent toners are different, the q/d's of the two populations will be different. The admix cases of T1+T1 and T2+T2, Figures 3 and 6, show this result. The admix time for these two cases is very short – "fast admix" – which is required for commercial developers. These toners may not offer the best test of the admix model because the charging dynamics occur so rapidly.



Figure 4. q/d versus time for the T1 incumbent and T2 added toners. The lines are calculated from Eq. 4.



Figure 5. q/d versus time for the T2 incumbent and T1 added toners. The lines are calculated from Eq. 4.

In the admix model, Eq. 4a and b, the second line contains an interesting term  $(\Phi_i - \Phi_a)$ . When the toners are the same this term vanishes. However, when the incumbent and

added toners are different, this term becomes important for the admix process. In the equations, it appears that the incumbent and added toners are triboelectrically charging against each other. That is not really the case:  $(\Phi_{incumbent} - \Phi_{added})$  is the difference between  $(\Phi_{incumbent} - \Phi_{carrier})$  and  $(\Phi_{added} - \Phi_{carrier})$ . The toners "know" about each other through the carrier beads.

The results in Figs. 4 and 5 show that when the higher charging T1 toner is added to T2, T1 charges higher than the incumbent toner, T2; and, when the lower charging T2 toner is added to T1, T2 charges less than T1.



Figure 6. q/d versus time for the T2 incumbent and T2 added toners. The lines are calculated from Eq. 4.

From the admix model, we can determine  $A_0$  values. We should expect them to be the same as the  $A_0$  values determined from the toner concentration data of Fig. 2. Fig. 7 shows the  $A_0$  values determined from the admix experiments versus the two  $A_0$  values determined from the toner concentration data. The admix  $A_0$  values seem to be slightly higher than those from the toner concentration data from Fig. 2. We were pleased with the agreement from the two different experiments and methods of analysis.



Figure 7. The  $A_0$  values determined from the admix data versus the  $A_0$  values determined from the toner concentration data. There is good agreement between the two sets of measurements.

# Summary

This is first time a model for the admix charging of toner added into a developer with charged toner and carrier has been presented. The mechanisms in the admix model are 1) triboelectric charge exchange between the carrier beads and either the incumbent or added toners; and, 2) toner charge sharing. An interesting outcome of these mechanisms is a term in which the two toners appear to be triboelectrically charging against each other but is actually the result of the toners charging against the carrier bead. The toners "know" about each other through the carrier bead. While in practice this term is not important - the added and incumbent toners are the usually the same - it offers the opportunity for verifying the model by adding a toner that is different than the incumbent toner. Finally, we have shown that the charge spectrograph can be used to characterize the A<sub>0</sub> values of developers, either from the admix data or from variable toner concentration data.

# Acknowledgment

We wish to thank Dr. G. C. Hartmann for helpful comments and suggestions.

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# **Biography**

Edward Gutman received his B.S. and M.S. in Physics from John Carroll University in 1965 and 1967, respectively; and his Ph.D. in Physics from Iowa State University in 1970. Following a post-doctoral fellowship at the University of Illinois, Dr. Gutman joined Xerox in 1972 working on the physics of the xerographic development process. He is currently a Research Fellow in New Toner and Developer Product Delivery and Manufacturing. His current interests are the physics of toners and developers.

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