Journal of Imaging Science and Technology® 50(3): 282–287, 2006.

A Bidirectional Acid-Base Charging Model for Triboelectrification: part 1. Theory

Richard P. N. Veregin▲, Maria N. V. McDougall▲, Michael S. Hawkins, Cuong Vong and Vladislav Skorokhod
Xerox Research Centre of Canada, Mississauga, Ontario L5K 2L1, Canada
E-mail: rick.veregin@xrcc.xeroxlabs.com

Henry P. Schreiber
Department of Chemical Engineering, École Polytechnique, Université de Montréal, Montréal, Québec H3C 3A7, Canada

Abstract. *A model for charging of insulative materials has been developed which quantitatively links the surface chemistry of materials to their work functions, and to the triboelectric charge exchange charge in two-component developers. The proposed model fits within the standard high-density of states model for charging, but proposes bidirectional electron transfer from basic sites on one contacting material to acidic sites on the second contacting material, and equally, from the basic sites on the second material to acidic sites on the first. The result is an "effective" work function for the material surface, the average of the work functions of the surface acidic and basis sites. This model predicts that both surface work functions and negative toner charge increase linearly with the logarithm of the ratio of the toner acid/base constants, K_a/K_b, which can be experimentally measured by inverse gas chromatography. Charge exchange is zero when the toner and carrier acid-to-base constants are identical. The model implies that an insulator's surface Lewis base and acid functional groups are responsible for triboelectric charge exchange. The model can also be applied to insulator to metal charge exchange.* © *2006 Society for Imaging Science and Technology.*
[DOI: 10.2352/J.ImagingSci.Technol.(2006)50:3(282)]

INTRODUCTION

Surface state models of triboelectric charging in insulators have successfully accounted for many of the salient features of charging in two component xerographic charging, including the effects of toner concentration, as well as toner and carrier particle size.[1–7] These models have also enabled establishing quantitative triboelectric series[1,6,7] for some xerographic developer materials. Despite the success of the surface state model for insulators, there have been few studies that have been able to link measured apparent work functions of insulator surfaces, a key parameter in surface state models, with measured charging in insulative xerographic developers. Further, there have been few examples to show how the specific surface chemistry of materials quantitatively determines the work function of the surface states, and how the work functions in turn relate to triboelectric charging. Smith[8] has shown that measured work functions of a series of ion binding polymers correlated with triboelectric charge. Work of Gibson and work of Shinohare et al. showed that work function surrogates—ionization potentials, electron affinities, Hammet σ functions, and the like—correlate to triboelectric charging.[9] A good example of a complete model—from surface chemistry, to work function, to triboelectric charging—is the early work with toners where charge is controlled by the carbon black included in the formulation. Fabish and Hair[10] showed that more oxidized carbon black surfaces are more acidic and thus have higher work functions. Julien et al.[11] showed that toners prepared with carbon blacks whose surface had been deliberately oxidized have more negative charge. Gutman and Hartmann[1] then showed that Julien's data could be interpreted in terms of the surface state model. However, because this work was based on conductive carbon blacks, its does not validate the surface state model for insulators. Nash et al.[6] showed that toner charging with insulative metal oxide surface additives did quantitatively fit a work function model; however, the work functions were a result of a fit to the model, and were not measured values. Veregin et al.[12] showed that measured work functions of insulative metal oxide additives explained quantitatively the charging of toners with those additives, although a connection to the specific surface chemistry of the oxides was not shown.

The purpose of this paper is to develop a quantitative link between the work functions of insulative materials and their surface chemistry. Using the surface state model, it will be shown that the surface chemistry and associated work functions should then predict the charging of two component developers. In a companion paper, the model will be verified for the example of insulative metal oxide surface additives.[13]

Lee[14] first proposed a surface state model for triboelectric charging of two-component xerographic developers. He assumed both toner and carrier surfaces have partially filled surface states. When a developer is mixed, charged species are transferred from higher energy filled states on one of the surfaces to lower energy unfilled sites on the other surface. Charging continues until the energies of the highest occupied states of the two surfaces are equal. Kondo[15] incorporated the idea that charge exchange creates an electric field

---

▲IS&T Member

Received Mar. 21, 2005; accepted for publication Jul. 27, 2005.
1062-3701/2006/50(3)/282/6/$20.00.

that opposes further charge exchange, so that charging ceases when the electric potential caused by charge exchange is equal to the work function difference between the toner and the carrier. The electrical potential is determined by the electrical capacitance of the associated toner and carrier particles, with a larger capacitance reducing the developed potential difference, and thus increasing the charge exchange. The capacitance is determined by the size of the toner and carrier particles as well as the number of toner particles associated with the carrier particles. Gutman and Hartmann[1] and Schein[2] combined Lee's and Kondo's models to consider the concentrations of charging sites on surfaces, the work function differences between the surfaces, and the effects of the electric fields created by charge exchange.

The surface state model has two different aspects, the two different limits on the density of states, depending on whether one assumes either that there is a high density of the surface states per unit energy, sufficiently high that the density of toner and carrier surface states does not affect charge exchange, or a low density of states, where the density of surface states does affect the magnitude of charge exchange. Castle and Schein[4] and Anderson[5] have shown a generalized equation for the surface states model, where the low-density and high-density limits are special cases, as shown in Eq. (1) for charge-to-mass ratio for the toner in a two-component development system.

$$\frac{m}{q}=\frac{\left[\frac{1}{eN_c}+\frac{d}{\varepsilon_0}\right]C_tR\rho_c+\left[\frac{1}{eN_t}+\frac{d}{\varepsilon_0}\right]r\rho_t}{3\Delta\phi}. \tag{1}$$

Here, $-e$ is the charge on an electron, ε_0 is the permittivity of free space, N_c and N_t are the carrier and toner density of states, respectively, ρ_c and ρ_t are the density of the carrier and toner, respectively, d is the distance between insulators at which the charge exchange ceases, R and r are the carrier and toner radii, respectively, C_t is the toner concentration in the developer (toner mass divided by carrier mass) and $\Delta\phi=\phi_c-\phi_t$, where ϕ_t and ϕ_c are toner and carrier work functions. A meta-analysis was done by Castle and Schein[16] of data based on the dependence of charge exchange on toner concentration, collected by many workers for two-component xerographic developers. While not completely conclusive, the meta-analysis does suggest that the charge transfer is not limited by the presence of finite surface states on the insulator surfaces, the low density limit, but primarily by the electric field generated by the charge transfer at the point of contact, the high density limit.

It is not the purpose of this paper to delve into the relative merits of the high-density of states or the low-density of states models. For the purposes of this paper, a high-density of states model will be assumed. Not only is the high-density of states model currently better accepted than the low-density of states, it also allows a clear interpretation for the charging model proposed herein. *Vide infra*, it will be shown in a companion paper, that the high density of states assumption is in quantitative agreement with work function, charging, and inverse gas chromatography (IGC) data for metal oxides as toner surface additives in xerographic developers. An interpretation for a low density of states model is also possible, although the fit of the low density of states model with our bidirectional charge model is not very satisfying. This interpretation will be briefly discussed as well.

THEORY

The fundamental question to be answered here is: What is the surface chemistry that defines a material's work function, and thus a material's ability to exchange charge? Given this question, the second question that needs to be answered is: How do we quantitatively define the chemistry of a surface? One method to define surface chemistry of a solid is in terms of its interaction with a liquid or gas phase. Thus one can probe the solid surface with a series of liquids or gases to measure the interaction parameters. Contact angles are one example of this type of measurement, where a solid is probed with liquids of varying chemistry to determine a contact angle, and thus to determine the associated interaction energy. Inverse gas chromatography (IGC) is another, more powerful example, where instead of determining a contact angle, one determines a retention volume—and thus the free energy—for the interaction of the solid with probe molecules. In either technique, the measurement depends on the interaction of the known chemistry of a selected probe and the unknown surface chemistry of the solid.

Quantification of Surface Chemistry by Inverse Gas Chromatography

The quantification of the surface chemistry of a solid by IGC is well established and has been widely applied.[17] In order to analyze that chemistry of a solid surface, the approach of Fowkes has proven valuable.[18] Consider a probe interacting with a solid surface. According to Fowkes the work of adhesion is the sum of the long-range Lifschitz-van der Waals forces, W_A^d, and the short-range acid-base interaction, W_A^{ab}:

$$W_A = W_A^d + W_A^{ab}. \tag{2}$$

The term W_A^{ab} sums up all short-range interactions as Lewis acid-base interactions, where a Lewis acid is defined as any material whose atoms can function as electron acceptors, and a Lewis base is defined as any material whose atoms can function as electron donors.

Equation (2) may be used to describe a surface, provided an appropriate method is available for the experimental determination of the quantities involved. The requirement is met by the IGC method, as a well established and widely applicable tool for the characterization of interactions with solid surfaces. In IGC, a solid of interest is packed in a chromatographic column. The solid needs to have a high surface area, such as a finely divided powder, or a thin film cast on a solid particulate support. An inert gas flows through the column, into which different probe liquids are injected at "infinite dilution" (i.e., by extrapolation from greater and greater dilution of the probe in the carrier gas). The probe molecule adsorbs and desorbs from the solid under study as determined by Eq. (2). The volume of gas required to transport the probe molecule through the column is the retention volume, which can be related back to the work of adhesion. As the chemistry of the probe varies, the work of adhesion will change, controlled by the interaction between the probe and the surface.

The first step in the IGC measurement procedure is to utilize probes that have only long-range van der Waal's interactions, for example a series of n-alkanes. For these materials, there are no short-range interactions, and thus the IGC data can be analyzed using the developments of Papirer, Schultz et al.:[19]

$$-\Delta G_A = RT\ ln\ V_n = 2Na(\gamma_l^d)^{1/2}(\gamma_s^d)^{1/2} + c. \tag{3}$$

Here ΔG_A is the free energy of the alkane probe adsorption, N is Avogadro's number, a is the cross-sectional area of the vapor molecule in the adsorbed state, V_n is the retention volume, γ_l^d is the dispersive surface energy of the liquid alkane probe, γ_s^d is the dispersive surface energy of the solid, and c is a constant.

The second step is to probe the surface with respect to the acid-base interaction parameter, by choosing differing probe molecules of varying acid-base character. This calls for a quantitative definition of the acid-base character of a material. Among the various acid-base theories which, in principle, would be applicable, that of Gutmann[20] is particularly appropriate. In Gutmann theory, organic fluids, and hence vapor probes, are identified as acids and bases according to their electron donor or acceptor numbers, AN and DN, which are obtained experimentally from calorimetric and nuclear magnetic resonance (NMR) investigations in which test materials are reacted with defined reference acids and bases. Values of AN and DN have been tabulated by Gutmann and used in experiments relevant to this account.[20] A series of probes of varying and known DN, AN, and γ_l^d are then utilized to measure a series of retention volumes with respect to temperature. To extract the acid-base interaction, the retention volume for an acid-base probe, $(V_n)^{ab}$ is compared to the retention volume of a probe that interacts only through long-range forces, $(V_n)_{ref}$, where both probes have the same γ_l^d. The result is the free energy for the acid-base interaction, ΔG^{ab}:

$$\Delta G^{ab} = -RT\ ln(V_n)^{ab}/(V_n)_{ref}. \tag{4}$$

From the temperature dependence of $\Delta G^{a,b}$ the enthalpy can be calculated:

$$\Delta H^{ab}/T = \Delta G^{ab}/T - \Delta S^{ab}. \tag{5}$$

Finally, the enthalpy of interaction can be written in terms of the probe AN and DN values:

$$\Delta H^{ab} = K_a DN + K_b AN. \tag{6}$$

Here K_a and K_b are the temperature averaged acid-base interaction constants for the solid stationary phase. The values are found by plotting Eq. (7), a rearrangement of Eq. (6):

$$\Delta H^{a,b}/AN = K_a DN/AN + K_b. \tag{7}$$

Thus, a plot of $\Delta H^{ab}/AN$ with DN/AN for a series of acid-base probes enables the calculation of K_a and K_b. Using these techniques, any solid surface can be defined by its surface interaction constants: γ_s^d, K_a and K_b.

A Bidirectional Acid-Base Triboelectric Charging Model for Insulators

Now that we can quantitatively define a solid surface, how can we relate this to surface work functions, as well as to triboelectric charge exchange? To illustrate the model, we will use the high density of surface states approximation, $1/(eN_{c,t}) \ll d/\varepsilon_0$, where toner charge can be described by:[1]

$$Toner\ q/m = A[(\phi_c - \phi_t)]. \tag{8}$$

While ϕ_t and ϕ_c are toner and carrier work functions, respectively, in a two-component development system, they could equally represent two other insulative solid particulate materials brought into contact. The factor $A = 3(\varepsilon_o/d)\ (RC_t\rho_c + r\rho_t)$ using the formalism in Eq. (1), although additional terms have also been shown to be important to correct for effects of multiple toner layers and non-equilibrium charge.[1] For the purposes of this work we need not concern ourselves with these additional terms, as they do not change the analysis, only the value of the A term. In order to illustrate the model, we shall assume fixed toner/carrier geometry, a fixed TC, and equilibrium charge levels, so that the factor A is a constant. This is not required for the derivation to follow, and is not a limitation of the model, but simplifies the illustration. Note that the work functions in this paper are defined here as positive numbers, so that a larger number represents lower energy. Thus, a more positive work function for toner compared to carrier gives a negative toner charge.

Following the surface model described above, the chemistry of a surface can be fully defined by the γ_d, K_a, and K_b. Since γ_s^d represents long-range forces, and ignores any Lewis acid-base interactions which would exchange charge, γ_s^d cannot be involved in triboelectric charge exchange. There is some experimental confirmation of this assumption. Schreiber[21] has measured K_a and K_b for a polyethylene, a polymeric long-chain alkane, and has shown that both acid and base constants were zero. In agreement with the IGC data, and our model, Hays[22] has shown that polyethylene shows very little ability to exchange charge, unless it is deliberately treated to create oxygen containing surface species. Thus polyethylene, which does not show K_a or K_b interactions, also does not show significant charge exchange.

To explain charge exchange, we are then left with the interaction constants K_a and K_b, which represent short-range interactions between donor and acceptor sites, and which do involve electron charge exchange. Thus, material work functions and charge exchange should be directly related to both acid *and* base properties of materials, K_a and K_b. Together the acceptor *and* donor sites must somehow collectively determine an effective work function for the sur-

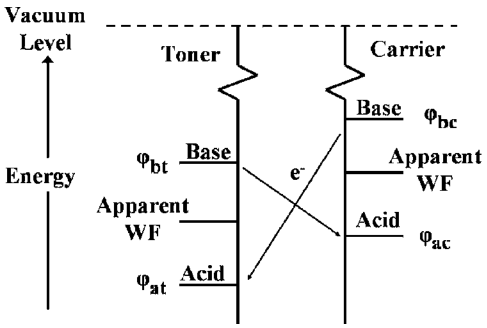

Figure 1. Toner-carrier charge exchange interactions. Arrows show the direction of the electron flow.

face of a material. This model predicts charge exchange in both directions, perhaps simultaneously on contact of the two surfaces:

(1) Transfer from toner donor (basic) sites to carrier acceptor (acidic) sites, which results in positive toner charge; and
(2) transfer from carrier donor sites to toner acceptor sites, which results in negative toner charge.

The model is illustrated in Fig. 1.

Thus, associated with the acidic surface sites of a material, we can define a work function, ϕ_a, and with the basic sites of a material, a work function, ϕ_b. Note that since we can only measure the average K_a and K_b for *all* acid and basic sites, $\phi_{a,b}$ are themselves effective work functions, averaged over all acid and base sites, respectively: $\phi_{a,b}=\Sigma\{(\phi_{a,b})_1+(\phi_{a,b})_2+(\phi_{a,b})_3+\cdots+(\phi_{a,b})_n\}$. The effective work function of a surface, ϕ_s, will be an average over the work function of the acid sites and the work function of the basic sites, as shown in Eq. (9):

$$\phi_s = (\phi_a + \phi_b)/2. \tag{9}$$

Substitution of the average work functions in the high-density of states charging model, Eq. (8), gives:

$$Toner\ q/m = (A/2)[(\phi_{ac} + \phi_{bc}) - (\phi_{at} + \phi_{bt})]. \tag{10}$$

Here the work functions are: ϕ_{at} for acidic toner sites, ϕ_{bc} for basic carrier sites, ϕ_{bt} for basic toner sites, and ϕ_{ac} for acidic carrier sites. Note that this model implies that the average work function for a *pure* material does *not* correspond to an existing physical state. Only the work functions for individual acid sites and the basic sites in an insulative material correspond to actual surface states. Thus the observed work function is only a global average over the work functions of the individual acid and base sites.

Using free energy relationships, we can relate the acid constants, K_a, to corresponding chemical potentials, where the more acidic the site, the lower the energy, and thus the higher the work function:

$$\phi_a = \phi_a^o + kT\ ln\ K_a. \tag{11}$$

Similarly, we can relate the acid constants, K_b, to corresponding chemical potentials, where the more basic the site, the higher the energy and thus the lower the work function:

$$\phi_b = \phi_b^o - kT\ ln\ K_b. \tag{12}$$

Here, ϕ_a^o and ϕ_b^o are the work functions of a chosen reference state for the acid and base constants, respectively, k is Boltzmann's constant, and T is temperature in degrees K. We use Boltzmann's constant here, rather than the molar gas constant, as our energy units are in eV. There are some assumptions behind the formulations in Eqs. (11) and (12) that need to be addressed. First, these equations assume that we have chosen an appropriate reference state for K_a and K_b, such that we can define an associated ϕ_a^o and ϕ_b^o. The choice of those reference states is arbitrary, though some choices provide simplicity, or at least a clearer definition. For Eq. (11) the best choice of a reference state is clear, it should be the state that corresponds to $\phi_a^o=0$, the vacuum level. With this choice ϕ_a will increase linearly with the logarithm of K_a at lower energies than the vacuum level. Thus, the chosen reference state will result in $K_a=1$ at the vacuum level. For the base sites, an appropriate choice could also be the vacuum level, or it could be a state with a large positive work function, so that as K_b increases, from a value of 1, the work function of the base site decreases. The important point to make is that the K_a and K_b determined by IGC are referenced to an arbitrarily selected reference material (defined by AN and DN) in an aqueous medium, and thus we do not know, *a priori*, the values of ϕ_a^o and ϕ_b^o. Clearly, they are unlikely to be zero just by chance, and so we will retain them in the ensuing calculations.

Comparing Eqs. (9), (11), and (12) gives rise to Eqs. (13) and (14), explicit quantitative relationships between the toner or carrier surface's work function and the acid and base constants of those surfaces can be obtained:

$$\phi_t = \tfrac{1}{2}\phi^o + \tfrac{1}{2}kT\ ln(K_{at}/K_{bt}), \tag{13}$$

$$\phi_c = \tfrac{1}{2}\phi^o + \tfrac{1}{2}kT\ ln(K_{ac}/K_{bc}). \tag{14}$$

Here we define $\phi^o=\phi_a^o+\phi_b^o$. Thus, the *apparent* toner work function of both toner and carrier is directly related to the ratio of the K_a to K_b values for acid and base sites of the toner and of the carrier, respectively.

Substituting Eqs. (13) and (14) into Eq. (8) gives the final result for toner charge based on the bidirectional acid-base charge model:

$$Toner\ q/m = -(A/2)kT[ln(K_{at}/K_{bt}) - ln(K_{ac}/K_{bc})]. \tag{15}$$

Note that ϕ^o cancels out in deriving Eq. (15), and thus does not affect the observed toner charge. Analogous to Eqs. (13) and (14), Eq. (15) predicts a toner charge that is directly related to the ratio of K_a to K_b values for acid and base sites of the toner, and the ratio of the acid base value of the carrier.

Predictions of Bidirectional Acid-Base Triboelectric Charging Model for Insulators

What predictions can be made from the bidirectional charge model, and how do those predictions compare with the standard high-density of states charging model? The following predictions can be made:

(1) Equation (15) predicts that a plot of toner q/m versus $ln(K_{at}/K_{bt})$ for a fixed carrier will be linear with a slope of $-AkT/2$. That is, negative toner charge increases with the increasing ratio of toner acid to toner base constants.

(2) Similarly, a plot of toner q/m versus $ln(K_{ac}/K_{bc})$ for a fixed toner will be linear with a slope of $-AkT/2$: Positive toner charge increases with increasing ratio of carrier acid to carrier base constants. Generally, when conceptualizing the standard model, an implicit assumption is made that charge exchanges from one surface to the other surface in one direction. That being said, the standard model does not prohibit bidirectional exchange. On the other hand, the acid-base model described herein shows that bidirectional exchange must be considered, because nearly all surfaces have both donor and acceptor properties (i.e., both K_a and K_b are nonzero).

(3) Eq. (15) also predicts that toner $q/m=0$, if $K_{at}/K_{bt}=K_{ac}/K_{bc}$, i.e. when toner and carrier acid/base ratios are equal. This is a very interesting result, as it shows that the toner and carrier surfaces do not need to have the same K_a and K_b to show no charge exchange, it is only necessary that their ratios are equal.

(4) Equations (13) and (14) predict that the work functions of the two surfaces are linearly related to the logarithm of the ratio of acid to base constants for the surface, with a slope of $kT/2$.

(5) The model predicts that γ_d does not affect work functions or charging.

(6) Finally, since the proposed model predicts that the material work functions and charging are dependent on K_a and K_b values, and since the acid and base constants are based on Lewis acid-base concepts, the model is implicitly implying that the species involved in charge exchange is an electron. While the standard model is usually thought of in terms of an electron as the exchange charged species, the standard model can apply to ion exchange as well, as long as the exchange is between discrete sites.

Extensions of Bidirectional Acid-Base Triboelectric Charging Model

The derivation of the acid-base charging model above is for the case of an insulator-insulator charge exchange. However, it is straightforward to extend this model to an insulator-conductor charge exchange. Thus, for example, in the case of an insulative toner material contacting a carrier that is a conductor, substitution of Eq. (8) in Eq. (8) gives:

$$Toner\ q/m = A\left[\phi_c - \tfrac{1}{2}\, ln(K_{at}/K_{bt})\right]. \qquad (16)$$

In this case, Eq. (13) will still predict the toner work function.

The insulator-insulator acid-base charge model was derived for the case of a high-density of states. Is there any way to interpret the acid-base model in terms of a low-density of states? The answer is there is not a satisfactory method to do this. The problem is that the low-density of states, where $1/(eN_{c,t}) \gg d/\varepsilon_0$, has two parameters for each surface, since we are left with the $1/(eN_{c,t})$ terms being important. Thus, in addition to the work functions, ϕ_t and ϕ_c, that we had for the high-density of states model, there are the density of states, N_t and N_c. Unfortunately, the acid-base model has only one parameter for each surface, the ratio of K_a/K_b. Also, the work function terms and density of state terms for each surface are not linear combinations, as expansion of Eq. (1) in the low-density of states model gives cross-terms. There are two obvious approaches to attempt to deal with the low-density of states. One is to identify the work function term, in the low-density of states model, exactly as done in the high-density of states model, to the K_a/K_b values [Eqs. (11) and (12)]. The model would look exactly like Eq. (16), except that the constant, A, would no longer be a constant for different materials, as it would now include the density of states, which would need to be determined by fitting to a set of data taken under varying conditions. The second approach would instead identify the density of states with the $K_{a,b}$ values. In other words, an IGC measurement of $K_{a,b}$ values would actually be measuring the density of states, rather than the apparent work functions of those states. The density of donor states would increase proportionally to K_a, and the density of acceptor states would increase proportionally to K_b. In this approach, the work functions would not be explicitly determined by the acid-base chemistry. Neither model is fundamentally satisfying, as the measured acid-base chemistry, which does explain donor-acceptor interactions in many materials applications, would not be sufficient to predict the magnitude of triboelectric charge exchange. Fundamentally there is not enough information in acid-base interactions to explain both work functions and density of states. Overall, only the high-density of states give a completely satisfactory model that relates acid-base parameters to work functions and charge exchange. Thus, the low density of states incorporates additional information that is not substantiated by the data measured by IGC.

CONCLUSIONS

A model for charging of materials has been developed, which quantitatively links the surface chemistry of a material, as defined by IGC determined K_a and K_b acid constants for the surfaces, to the surface work function, and thus to the ability to exchange charge in a two-component developer. The model proposes bidirectional electron transfer from basic sites on one material to acidic sites on a second material, and equally important, from the basic sites on the second material to acidic sites on the first material. The result is an observed "effective" work function that is the average of the work functions of the acid and base sites of each material in the charge exchange. Thus, the observed work function and toner charge are predicted to be determined by the relative amounts of these two electron exchange processes, which are linearly related to the logarithm of the ratio of the IGC determined K_a/K_b values. The higher the acid-base ratio the higher the material work function and the more negative the materials charge. The model implies that the electron donor and acceptor sites in developer materials, which are responsible for developer triboelectric charging, are the Lewis base and acid functional groups, respectively, in the materials, and that the species responsible for the charge exchange is the electron. Charge exchange is zero when the toner and carrier acid-to-base ratios are identical.

REFERENCES

[1] E. J. Gutman and G. C. Hartmann, J. Imaging Sci. Technol. **36**, 335 (1992).

[2] L. B. Schein, J. Imaging Sci. Technol. **37**, 1 (1993).

[3] J. H. Anderson and E. Fox, J. Adhes. **51**, 27 (1995).

[4] G. S. P. Castle and L. B. Schein, J. Electrost. **36**, 165 (1995).

[5] J. H. Anderson, J. Imaging Sci. Technol. **38**, 378 (1994).

[6] R. J. Nash and R. N. Muller, *Recent Progress in Toner Technology* (IS&T, Springfield, VA, 1997) p. 267.

[7] J. H. Anderson, J. Imaging Sci. Technol. **43**, 460 (1999).

[8] T. Smith, Polym. Prepr. (Am. Chem. Soc. Div. Polym. Chem.) **30**, 342 (1989).

[9] H. W. Gibson and F. C. Bailey, Chem. Phys. Lett. **51**, 352 (1977); H. W. Gibson, J. Am. Chem. Soc. **97**, 3852 (1975); I. Shinohara, F. Yamamoto, H. Auzai, and S. Endo, J. Electrost. **2**, 99 (1976).

[10] T. J. Fabbish and M. L. Hair, J. Colloid Interface Sci. **62**, 16 (1977).

[11] P. Julien, in *Carbon-Black Polymer Composites*, E. Sichel, Ed. (Marcel Dekker, NY, 1982), p. 410.

[12] R. P. N. Veregin, D. Powell, C. P. Tripp, M. N. V. McDougall, and M. Mahon, J. Imaging Sci. Technol. **41**, 192 (1997).

[13] R. P. N. Veregin, M. N. V. McDougall, M. Hawkins, C. Vong, V. Skorokhod, and H. P. Schreiber, J. Imaging Sci. Technol. **50**, 288 (2006).

[14] L. H. Lee, Photograph. Sci. Eng. **22**, 228 (1978).

[15] A. Kondo, *TAPPI Reprographics Conference Proceedings*, 153 (1980).

[16] G. S. P. Castle and L. B. Schein, *Recent Progress in Toner Technology* (IS&T, Springfield, VA, 1997), p. 149.

[17] H. P. Schreiber and D. R. Lloyd, in *Inverse Gas Chromatography; ACS Symposium Series 391*, D. R. Lloyd, T. C. Ward, and H. P. Schreiber, Eds., American Chemical Society, Washington, DC, 1989, p. 1; P. Mukhopadhyay and H. P. Schreiber, Colloids Surf., A **100**, 47 (1995).

[18] F. M. Fowkes, J. Adhes. Sci. Technol. **4**, 669 (1990).

[19] C. Saint Flour and E. Papirer, J. Colloid Interface Sci. **91**, 69 (1983); J. Schultz, L. Lavielle, and C. Martin, J. Adhes. **45**, 23 (1987).

[20] V. Gutmann, *The Donor-Acceptor Approach to Molecular Interactions* (Plenum Press, New York, 1978).

[21] H. P. Schreiber and A. Ouhlal, J. Adhes. **79**, 141 (2003).

[22] D. A. Hays, J. Chem. Phys. **61**, 1455 (1974).