The Role of Technical Innovation in the Physics of Electrophotography

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Abstract

Over the years, technical innovation in the physics of electrophotography has had mixed results in influencing the technology. Four examples familiar to the author will be discussed and their current and potential future influences on the electrophotographic technology will be described. (1) The introduction of the theory of magnetic brush development had only limited influence on the invention of improved variants. However, it had an enormous influence on the perception that electrophotography was a logical and predictable technology that could be understood with common scientific tools. (2) The introduction and implementation of the ideas of the Proximity Theory of toner adhesion has allowed the design of the smallest size, lowest cost color electrophotographic engine which will be commercially available soon. (3) The demonstration that toner charging is due to Kondo's high density theory surprisingly has failed to help toner material scientists redesign toners; in particular, it has failed to help them design a toner with a narrower charge distribution. Such a narrow-charge distribution toner would allow the average toner charge-to mass ratio to be reduced, which would be useful in many of the electrophotographic subsystems. Combining a narrow-charge distribution toner with the methods of reducing the toner adhesion based on the Proximity Theory could lead to a new generation of marking engines based on novel marking technologies. Finally, (4) progress has been made recently in identifying the mechanism by which holes move through our photoreceptors. We conjecture that identifying the charge transport mechanism will be beneficial to the design of organic photoreceptors for electrophotographic systems as well as other organic electronic devices. Although today's photoreceptors have adequate mobility, increased process speed will require both higher mobility and a reduction in those factors giving rise to carrier range limitations. For organic electronics higher mobility materials are necessary for practical devices.

Introduction

There exists a tension between empirical methods and technical innovation in every technology, but especially in electrophotography in which many variables in many subsystems contribute to the output (the printed or copied page). The question addressed in this paper is how technical innovation has advanced the physics of electrophotography by examining specific examples. It will be seen that there are some notable successes and failures. However, even those technical innovations which have not yet been implemented in electrophotography may provide future directions for the evolution of the technology into novel marking technologies and advances in organic electronics.

Examples are provided, based on the author's knowledge, including (1) the theory of magnetic brush development, (2) the theory of toner adhesion, (3) the theory of toner charging, and (4) the charge transport mechanisms in molecularly doped polymer films which are used in organic photoreceptors. For each example, the technical innovation will be described, it success in influencing the electrophotographic technology will be noted, and its potential to influence the future direction of the technology will be conjectured.

Magnetic Brush Development

Magnetic brush development systems are used in virtually all electrophotographic engines that operate above 30 pages per minute. It was originally invented at RCA but was developed and used in all electrophotographic copiers and printers after the original Xerox 914 Copier, which was introduced in 1959 and used cascade development [1]. The magnetic brush development system replaced the cascade development system because it allowed higher process speeds and the copying of solid areas. In the late 1960s and early 1970s the design of magnetic brush development systems was almost completely empirical. This was expensive because of the many potential parameters that affected the output, the developed toner mass-per-unit area, such as [1] the toner charge-to-mass Q/M ratio; the toner size; the toner charge and size distributions; the toner concentration (ratio of toner to carrier mass); the carrier size, shape, and magnetic properties; the speed ratio between the development roller and the photoreceptor; the gap between the development roller and the photoreceptor; the voltages on the development roller, the voltages on the charged and exposed areas of the photoreceptor, the design of the magnetic fields, etc. It was felt by many people that a "theory" of magnetic brush development was not possible because of the many different forces acting on the many different types of toner particles (due to the large variation of diameters and charges). For example, models based on field stripping (the Coulomb force overcomes toner-carrier adhesion) or toner powder clouds (the Coulomb force harvests toner particles that are free of the carrier) were qualitatively discussed as possible explanations for particular data sets.

Then in 1974 and 1975 two papers were published in which a technical innovation was introduced: it was demonstrated that the physics of the process was quantifiable and understandable with a simple physical model. In the first paper [2] the toner voltage (the change in voltage across the development gap due to the presence of developed toner on the photoreceptor) was shown to be due to three effects, (1) a constant volume charge density (the charged toner) placed on a dielectric, the photoreceptor; (2) the discharge of the photoreceptor due toner being developed and subsequently removed during the development process; and (3) size classification of the toner as development time increases, effectively changing the average toner charge-to-mass ratio. In the second paper [3] it was shown that the output of the magnetic

brush development system, the developed toner mass-per-unit area, was due to the competition between two forces, the Coulomb force due to the latent image attracting toner to the photoreceptor and the build-up of charge on the carrier beads adjacent to the photoreceptor attracting toner to the charged carrier beads. The build-up of charge on the carrier beads was due to the toner being developed onto the photoreceptor earlier in the development zone. This physical model allowed the derivation for the first time of a simple equation relating developed mass-per-unit area to hardware and material parameters and provided a simple, intuitive picture of how the development system "worked."

This technical innovation, a theory of magnetic brush development, influenced the design of future magnetic brush developed system simply because the first order hardware and materials parameters were identified, allowing for a more straightforward design process. I vividly recall an engineer coming up to me one day saying that he had done a correlation study between developed mass-per-unit area and toner concentration and had found no correlation, to his surprise but consistent with the theory. Many people have said to me that the demonstration that a theory worked for magnetic brush development changed electrophotography from an art to a science and gave them confidence to approach the other development systems and other subsystems in a more scientific and systematic manner.

One would have thought that such a theory would have led to innovative approaches to improving the magnetic brush development system. After all, it is clear that if the mass-per-unit area is limited by the build-up of charge on the carrier beads adjacent to the photoreceptor, reducing this build-up would improve the efficiency of the system. But by 1975, at least 2 and probably 3 or the variants had already been discovered. These include (1) the conductive magnetic brush development [4] in which the carrier charge is shorted to the development roller by making the carrier chain conductive (a Kodak invention), (2) the Kodak SPD system [5], in which permanent magnetized carrier beads are moved out of the development zone with a rotating magnetic shell inside the development roller, and (3) the use of ac electric fields to move toner down the carrier chain to neutralize the carrier charge. It is possible that only the fourth variant [6] (introduced by Xerox in their high speed 9400 series machines) in which the magnetic forces in the development zone were significantly reduced to allow the charged carrier to mix with uncharged carrier, was influenced by the theory.

Proximity Theory of Toner Adhesion

Toner adhesion is one of the critical elements in electrophotography that must be taken into account in the design of three of the subsystems, development, transfer, and cleaning. Lower adhesion would obviously be advantageous, allowing for a simpler design of these subsystems. But an understanding of toner adhesion remained elusive for almost 40 years since the first measurements by Goal and Spencer [7]. Everyone agreed that the simple electrostatic model of adhesion based on the assumption that the toner particles can be modeled as a spherical dielectric, uniformly charged on the surface, was not valid. In this model the charges on the uniformly surface charged toner particle on a conductive surface are attracted to their image charges. Toner adhesion data always gave adhesion forces at least 10 times larger than predicted by this model [8]. This large adhesion was considered a "fact of life," forcing the electrophotographic subsystems designers to design around this high adhesion. It has been argued [9] that innovations in the design of new development systems was basically a process by which this high adhesion was overcome, either by (1) canceling the adhesion of toner to its carrying surface using three-body contact events (magnetic brush development, Ricoh's nonmagnetic monocomponent development), (2) using ac electric fields and low toner charge (Canon's magnetic monocomponent development system), or (3) using horizontal ac electric fields (Xerox's carrier-toner iGen3 system). Techniques to overcome high toner adhesion also were needed in the transfer and cleaning subsystems to make them viable.

Over the years, attempts to understand the physical basis of the experimentally observed high toner adhesion were based on two approaches, one that assumed that van der Waals forces dominated and one that assumed that the toner charge was non-uniformly distributed around the surface of the toner particle, as reviewed in Ref. 10. As almost all measurements showed that toner adhesion depended on the square of the toner's Q/M, it seems clear that an electrostatic model is needed to explain the data. But the non-uniform charge distribution model had two inconsistencies with data (1) the non-uniform charge density needed to be consistent with adhesion data predicted electric fields in excess of Paschen's breakdown, i.e. they would not be stable in air, and (2) it predicted that Q/M would be independent of the toner concentration in toner-carrier mixtures, inconsistent with data [10].

The technical innovation in toner adhesion was initiated by the CEO of the Aetas Corporation who wanted to build the smallest size, lowest cost, color EP engine. In order to accomplish this, the four color toner images had to be accumulated on the photoreceptor. This could only be done if the electric fields of the latent image were sufficient to "develop" the toner from a monocomponent development roller to the photoreceptor, without the use of ac electric fields or contact development. This required that toner adhesion to be lowered by at least a factor of 10 compared to commercially available toner. In the process of simulating toner adhesion based on image forces with finite element methods, it was found that if the discreteness of toner charge were retained in an electrostatic model, then a new force emerged from the calculations both analytically or by adding up all the images forces between all of the charges. This new force was due almost entirely to the forces on those charges in closest proximity to the surface on which the toner rested. This was subsequently named the Proximity Force. This work is summarized in Ref. 11 and 12. This force is active at every contact point between the toner and the surface. The number of contact points could be reduced to a minimum (three, a tripod analogy is useful) by coating the toner particle with about a monolayer of nm size extraparticulates. Such a toner was made; experimentally shown to have a toner adhesion at least 10 times smaller than commercially available toners; shown to have adhesion consistent with three contact points model; and used in the design of the smallest size, lowest cost, color electrophotographic printer, which should be commercially available soon. As discussed below, this approach to lowering toner adhesion may in the future influence the evolution of the electrophotographic technology into novel

marking technologies and it may affect other technologies outside of the printing field.

Kondo's Theory of Toner Charging

What determines a toner's charge? Toner charge, or the more easily measured toner charge-to-mass ratio, Q/M, is of fundamental importance in most of the subsystems in electrophotography. Yet, our control of the toner charge is highly empirical from a material's point of view. Because the toner charge and its distribution has such a strong influence on the behavior of toner particles in the various subsystems, empirical testing of new electrophotographic engines remains a costly, time consuming effort as machine optimization and toner charge distributions are optimized simultaneously and empirically.

The physics/chemistry of toner charging is a subset of the general problem of insulator electrostatic charging. It is one of the oldest areas of solid state physics. The Greek's knew that rubbed amber attracts particles and Benjamin Franklin's experiments on charging and lightening are well known. It is by far one of the least understood areas of solid state physics. It appears to be a surface phenomenon but all attempts to associate electrostatic charging of insulators with any known material parameters has not been successful yet. The reader is invited to read any of the reviews on the subject (see Chapter 4 in Ref. 1 and Ref. 13 and 14) to see how rudimentary our understanding is whenever insulators are involved. (Metal-metal contact charging is well-understood and determined by the difference in work functions.)

A technical innovation came when it was realized that insulator electrostatic charging experiments using toner particles have two advantages relative to typical experiments that use contacts between insulating sheets: (1) toner particles are made reproducible in their charging behavior because the electrophotographic technology demands it and (2) there is an extra variable present in toner-carrier mixtures, namely the toner concentration - the ratio of toner mass to carrier mass, i.e. the number of toner particles on a carrier particle can be varied. This extra variable allowed an experimental test that could distinguish between two suggested surface charging models, the low density and the high density theory of charging. The low density theory is similar to the theory of metal-metal electrostatic charging – it is assumed that there are "surface" work functions on the two insulators making contact which determine how charges move from the surface states of one insulator to the other to equilibrate the work functions. The high density theory assumes that there are so many surface states on the two insulator surfaces that a large voltage drop is created by the movement of charge which shuts off the flow of charge. Lee [15] proposed the equations which describe the low density theory for a toner-carrier mixture. Kondo [16] proposed the equations for the high density theory. Schein and Castle [17-20] suggested an experiment that would distinguish between the two theories, carried out the experiment, and showed that only the electric field theory (the high density theory) is consistent with experiments. The experiment involved comparing the slope-to-intercept ratio in a plot of M/Q vs. toner concentration to theory. It is remarkable to note that quantitative agreement was obtained with the predictions of the high density theory with no adjustable parameters, a result that has not be duplicated in any other electrostatic charging experiment, to the author's knowledge. Toner charging appears to be limited by the build-up of an electric field between the toner surface and the carrier surface; toner stops charging when this electric field reaches a material-dependent value of the electric field. Therefore this theory is sometimes called the electric field theory of toner charging. The value of the electric field at which toner stops charging, called the effective electric field, can be obtained from the slope or intercept of the plots mentioned above. It is about 10 V/μm within about a factor of 4 depending on the materials used. However, no one has been able to associate this electric field with a material property. For example, if one assumes that the electric field is due to the difference in work function between the two materials (which should be on the order of 1 eV) divided by a tunneling distance (1 nm works for the metal-metal charging experiments), then one predicts electric fields 100 times higher than observed in the charging experiments, 1000 V/μm. In addition, it appears from discussion with chemists that it is not clear how to associate an electric field with material properties. There have been recent experiments in which the surface chemistry of polymers was probed for its triboelectric properties by measuring the residence time of probe molecules using inverse gas chromatography [21]. There also exists new theoretical work on the electrification of granular systems of identical insulators [22]. However in all of these models no reasonable explanation of the effective electric field emerges. It appears that that the demonstration that the high density theory explains toner charging has had no influence on toner design. But the identification of a material property that determines the toner charge distribution has enormous potential to influence the technology, as discussed below.

Potential Future Influence of Technical Innovations

One can imagine potential significant influences on the design of electrophotographic subsystems if the electric field theory of toner charging could be used to narrow the toner charge distribution. That would allow the average toner Q/M to be lowered because the charge distribution could be made narrower and the amount of wrong sign toner could be reduced for lower Q/M's. It is obvious that lower Q/M and therefore lower toner adhesion would be beneficial in the development, transfer and cleaning subsystems.

Furthermore, it is interesting to consider the implications of the above technical innovations to novel marking technologies. Novel marking technologies are defined as electrophotographic marking technologies in which one or more of the electrophotographic subsystems have been eliminated. Many have been suggested including Contact Electrography [23], Tonerjet [24], Traveling Electric Waves [25, 26], Direct Imaging Technologies 27], Laser Ablation Technology [28], Electrostatic Powder Transfer Technology [29], Laser Fusion Technology [30], Magnetography [1, 31], and Ionography [1, 31], among others. Very few have been commercialized and none have challenged the electrophotographic technology, yet.

Consider the implications of having a narrow-charge distribution toner combined with methods of lowering its toner adhesion, two technical innovations discussed above. Let us project reductions and/or control of toner adhesion by another factor of 10 beyond what Aetas has achieved.

What would a factor of 10 reduction in toner adhesion control allow? Standard desk-top color electrophotography will not be

made smaller or lower cost. Aetas has achieved the smallest possible electrophotographic color system architecture – one will always need to accumulate the color images on some surface and Aetas' choice, the photoreceptor, eliminates all extraneous subsystems, such as intermediate belts. And the mid range and high end (Xerox's iGen3, Kodak's Nexpress, HP's Indigo liquid system) will obviously continue to evolve, but not in any step-function manner.

Instead consider the many suggestions through the years of novel marking technologies. To be specific consider for example TonerJet [24]. This is a toner marking technology in which toner is "jetted" across an interface between a roller and paper in an image-wise method. A voltage is placed on the back of the paper which attracts toner across an air gap. The toner particles go thought tiny apertures which can be shut off electrically. Such a technology is sometimes called direct marking. It eliminates the charging, exposure and transfer subsystems of electrophotography as well as the need for a photoreceptor. It failed to be commercialized because of some obvious technical problems. Wide toner adhesion distributions led to poor release of toner from the roller, slowing down the toner jetting time. Wrong sign toner contaminated the back of the apertures. And the voltages needed to jet the toner were large due to the large toner adhesion, in the range of at least several hundred volts, making the electronics prohibitively expensive.

But consider a future in which toner charge can be 10 times lower than today and/or overcoming the adhesion of toner particles requires 10 times less force, i.e, lower voltages. Perhaps a direct marking technology such as TonerJet would become feasible. It would be small, very inexpensive, and perhaps capable of high speeds. Other novel marking technologies may also be made viable by our assumed 10X reduction in toner adhesion.

Mechanism of Charge Transport in Molecularly Doped Polymers

The organic photoreceptor OPC used in virtually all electrophotographic engines is made up of a thin charge generation layer and a thick charge transport layer made from a molecularly doped polymer MDP. The mechanism of charge transport in the MDP has been under discussion for many years. Most papers have used the Gaussian Disorder Model GDM to explain experimental data. The GDM envisions the charge carriers hopping in a Gaussian distribution of states through the MDP aided by the imposed electric field. The width of the Gaussian distribution states is directly determined by the disorder in the material.

The technical innovation that has occurred in the last two years is a series of papers which have compared the GDM and other transport theories to the whole body of experimental data, instead of focusing on one material at a time. When this was done, it was found that:

(1) The GDM does not adequately describe charge transport data in MDP because the disorder energy, which can be obtained from the temperature dependence of the mobility, does not change as predicted by the theory when the disorder is changed [32]. The reasoning that led to the conclusion was discussed last year at NIP 24 [33] and in Ref. 32.

(2) The transient current shape is not understandable in terms of any known transport theory [34]. In a plot on linear-linear axes the transient current shape can be described by an initial spike followed by a relatively flat current before the transit time; a mobility that precisely follows the Poole-Frenkel law, being exponential in the square root of the electric field; and a current after the transit time that falls much more slowly than can be accounted for using Gaussian statistics. While the flat plateau perhaps indicates equilibrated transport, the origin of the spike remains unknown.

In addition, the falling current after the transit time is puzzling. Attempts to explain this behavior with all known theories have failed to account for it. It is much wider than Gaussian transport would predict and it is not consistent with the width being determined by the generation region, Rudenko's field diffusion, Coulomb repulsion, or an intrinsic shallow-trap controlled mobility [34]. The data are inconsistent with the predictions of the GDM (putting aside the difficulties that the GDM has explaining the electric field dependence of the mobility and the experimental effects of changing disorder on the activation energy) because the slope of the current before the transit time is observed to be more shallow than predicted and because the field independence of slope is not predicted. Field independent slopes appear to be a general phenomenon of molecularly doped polymers [34]. The data are inconsistent with the Scher-Montrol theory that assumes waiting time distribution functions which are longer than the transit time and its predicted electric field dependence [34].

(3) The activation energies have been derived from the disorder energies [35] and are approximately 0.3-0.8 eV. This is larger than predicted for the disorder or the polaron binding energies [36]. Apparently a higher order energy process is governing transport in molecularly doped polymers.

Given the uniformity of behavior of the mobility in all characterized MDP's (with respect to electric field, temperature, current shape, and sample thickness) we do not find it reasonable that there are two types of mobility characteristics in MPD's, those that have an activation energy that is independent of dopant concentration and those that have an activation energy that depends on dopant concentration. It is argued [35] that the intrinsic mobilities in MDP's are activated, with an activation energy that is independent of dopant concentration, and have a pre-factor that is exponential in ρ, the calculated distance between hoping sites, as expected for a hoping theory. The experimental observations of other behaviors, activation energies that depend on ρ and pre-factors independent of ρ, are attributed to either interactions among the dopant molecules or the failure of the lattice gas model to properly calculate ρ at high dopant concentrations.

It appears that something critical is missing from our understanding of charge transport in MDP and perhaps in all organic materials. While progress is being made in the elucidation of the charge transport mechanism in molecularly doped polymer, much work remains. Our current focus is understanding the origin of the initial spike and using a new experimental technique called TOF1A, in which the hole generation region is varied from the surface to the entire bulk by using electron beam excitation. We conjecture that identifying the charge transport mechanism will be beneficial to the design of organic photoreceptors for electrophotographic systems as well as other organic electronic devices. Although today's photoreceptors have adequate mobility, increased process speed will require both higher mobility and a reduction in those factors giving rise to carrier range limitations.

For organic electronics higher mobility materials are necessary for practical devices.

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