Physics of electrophotography

Our knowledge of the physics underlying this printing and copying technology has advanced considerably since its invention 48 years ago, but there are still important areas in which the principles are incompletely understood.

Donald M. Burland and Lawrence B. Schein

Electrophotographic printing and copying systems are based on two well-known but not well-understood physical phenomena: electrostatic charging and photoconductivity. That some materials can acquire an electric charge by contact or rubbing has been known at least since the time of Thales of Miletus, around 600 B.C., and much work has been done on understanding the phenomenology of the effect, particularly in the 18th and 19th centuries; nevertheless the underlying physics of electrostatic charging of insulators remains unclear. Photoconductivity is a considerably more recent discovery, dating back only to 1873, when Willoughby Smith discovered the effect in selenium. Early work concentrated on crystalline covalent solids; only recently has the photoconductivity of highly insulating amorphous materials been studied. In fact the invention of electrophotography was a major catalyst to research in both electrostatic charging and photoconductivity.

It is a tribute to the genius of Chester F. Carlson, the inventor of electrophotography, that he was able in 1938 to combine such little-understood physical phenomena into a process that is now at the heart of a rapidly growing, $20 billion industry. Commercial products span the range from low-cost personal copiers and printers that produce a few pages per minute to high-speed printers that produce up to 220 pages per minute—corresponding to paper speeds of over three miles per hour. Laser printers, which use electrophotography, are much quieter than impact printers and they can print multiple type fonts and pictures as well as more than one color.

Donald Burland and Lawrence Schein are members of the research staff at the IBM Almaden Research Center in San Jose, California.

Technology of electrophotography

Given all the attention applied to the technology of electrophotography since 1959, when the first commercial copier was introduced, one might expect that the field would have matured to the point where few relevant scientific questions remain. However, a closer look at the process reveals several areas where a deeper scientific understanding could result in significant improvements in print quality, reliability and cost. While a useful understanding of the macroscopic electrostatics involved in the development process has been achieved, other areas are much less advanced. In fact we still lack a microscopic physical understanding of the two major physical phenomena that Carlson combined in his first embodiment of electrophotography, electrostatic charging and photoconductivity of insulators.

The process of electrophotography is shown schematically in figure 1. It is a complex process involving[1,2] in most cases six distinct steps:

- Charge: A corona discharge caused by air breakdown uniformly charges the surface of the photoconductor.
- Expose: Light reflected from the image (in a copier) or produced by a laser (in a printer) discharges the normally insulating photoconductor and produces a latent image—a charge pattern on the photoconductor that mirrors the information to be transformed into the real image.
- Develop: Electrostatically charged and pigmented polymer particles, called toner, are brought into the vicinity of the oppositely charged latent image; they adhere to the latent image, transforming it into a real image.
- Transfer: The developed toner on the photoconductor is transferred to paper by giving the back of the paper a charge opposite to that on the toner particles.
- Fuse: The image is permanently fixed to the paper by melting the toner into the paper as it passes between two rollers, one of which is usually heated.
- Clean: The photoconductor is cleaned of any excess toner using, for example, coronas, lamps, brushes or scraper blades.

Part of the difficulty in perfecting the electrophotographic process comes from its obvious complexity and the interrelated character of the steps. For example, toner materials that optimize development may make fusing difficult, and ozone produced by the various coronas may cause chemical degradation of other parts of the system. A straight paper path on top of a copier is more reliable than a convoluted path deep inside it, but requires a larger overall box to accommodate the longer optical path—because the optics must of course be outside the paper path.

In the following sections we will discuss some of the steps of the electrophotographic process in more detail. We begin by comparing several models of development with experimental data to elucidate the physics of toner-particle behavior during development.

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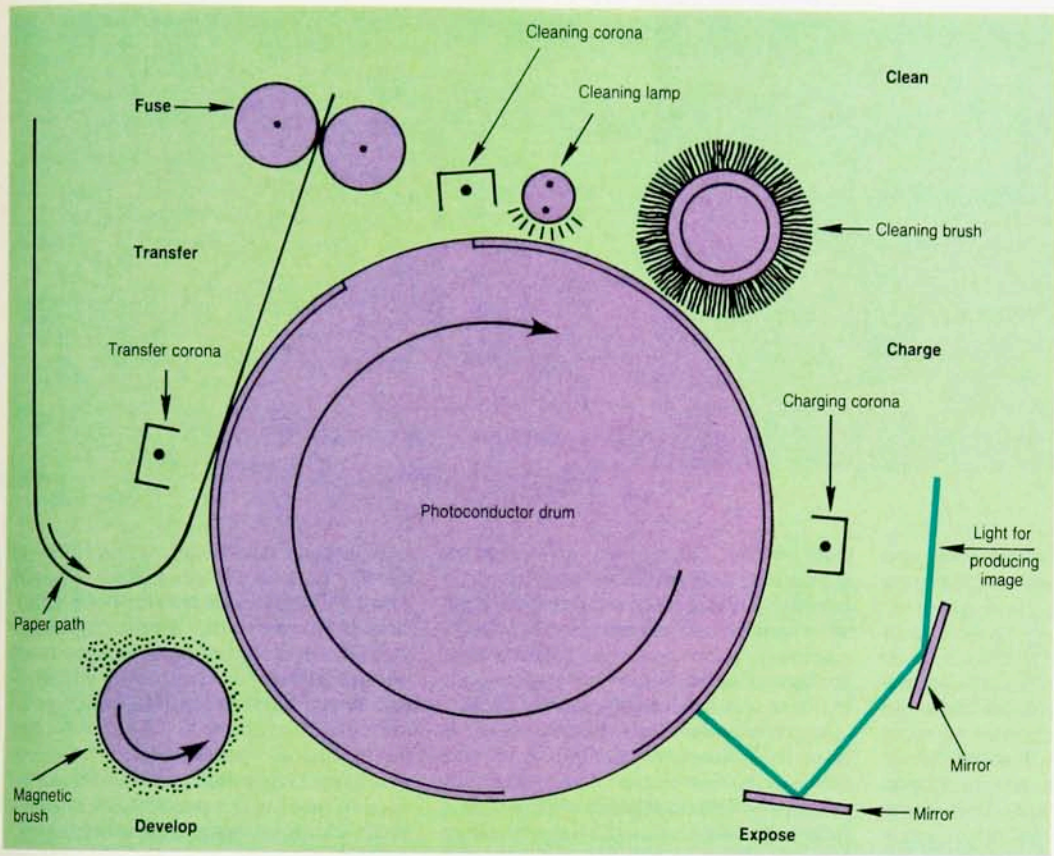


Electrophotography can be separated into six steps: charge, expose, develop, transfer, fuse and clean. The diagram locates the steps schematically in the IBM Series III copier. Figure 1

Next we will discuss how the toner particles become charged during development. Our third and final topic is one of the many interesting phenomena associated with the exposure step: the transport of the photogenerated charge across the photoconductor. By these examples we hope to show that a more complete understanding of the physics underlying these process steps can result in improvements in electrophotographic printers and copiers.

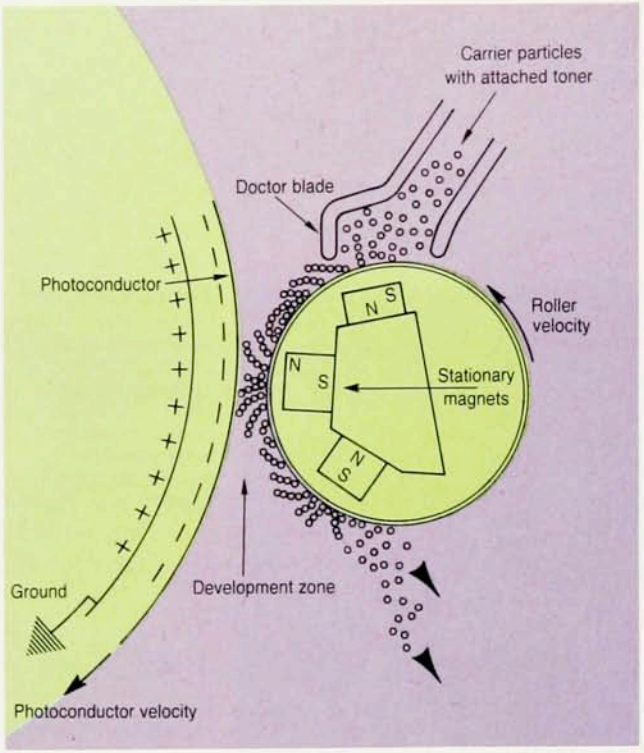
Physics of development

Let us first consider the development of the latent image on the photoconductor, which takes place when toner particles are brought into close proximity with the photoconductor.[2,3] In most development systems, the toner (approximately 10 microns in diameter) is mixed with carrier beads (200 microns in diameter). If the materials are selected properly, the mixing causes both the toner and carrier particles to become oppositely charged, as we will describe below. As a result, the toner electrostatically adheres to the carrier particles. In the development system used in almost all electrophotographic machines today, the carrier beads are made from a soft magnetic material so that they form magnetic "brushes" on a roller that carries them—by a combination of frictional and magnetic forces—past stationary magnets into the development zone, as shown in figure 2. Here, in response to the electric field of the latent image, the toner transfers from the carrier beads to the photoconductor. (Other development systems, used primarily for low-speed copiers, are based on single-component mixes, in which the magnetic material is put directly into the toner particles themselves.)

One measure of the quality of a printed copy is how black the images are. This can be correlated with the mass M of toner developed onto a unit area A of the latent image (and, of course, transferred to the paper). A model of the development process should be able to relate the quantity M/A to other measurable parameters of the development process. At first glance one might imagine that toner continues to develop onto the photoconductor until the charge on the photoconductor has been completely neutralized. This neutralization would occur when the toner charge per unit area σ_T equals the photoconductor charge per unit area σ_P, which is determined by the potential V to which the photoconductor is charged:

$$\sigma_P = K\epsilon_0 V/d$$

where K is the dielectric constant of the photoconductor and d is its thickness. Because the toner charge per unit area equals its charge per unit mass (Q/M)



Magnetic-brush development system. The carrier beads (100–300 microns in diameter) are carried into the development zone by a combination of magnetic and frictional forces exerted by the rotating roller and stationary magnets. The toner particles (around 10 microns in diameter) are electrically charged by being mixed with the carrier beads. Figure 2

times the developed mass per unit area (M/A), the toner mass per unit area is

$$M/A = \frac{\epsilon_0 V}{d/K}\,\frac{1}{Q/M}$$

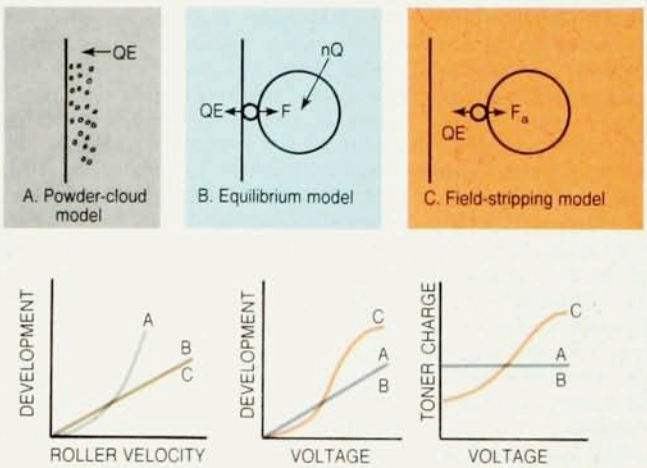
This result overestimates the developed mass per unit area by about an order of magnitude. Experiments show that in general σ_T is only about $^1/_{10}\sigma_P$. Something other than the photoconductor charge must thus limit toner development.

Figure 3 illustrates the three models of solid-area development that have been proposed to describe[2,3] the process by which toner leaves the carrier particles and ends up on the photoconductor surface: the powder-cloud model, the equilibrium model and the field-stripping model. Three sets of measurements can distinguish among the three models: developed mass per unit area as a function of roller velocity, developed mass per unit area as a function of voltage V and toner charge-to-mass ratio as a function of electric field.

In the powder-cloud model, toner is freed from the carrier by inertial forces during carrier–carrier and carrier–photoconductor collisions. The electric field associated with the latent image attracts to the photoconductor, via the Coulomb force, the charged toner out of the resulting cloud of particles.

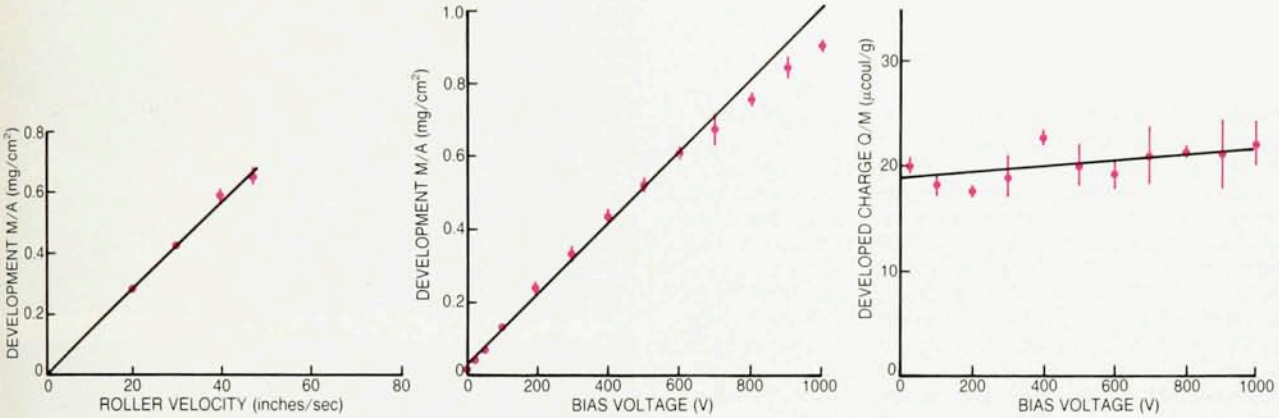
If this model describes development accurately, the developed-toner mass per unit area should be proportional to the flow of toner and a function of carrier-bead agitation. The flow of carrier particles is proportional to the roller velocity, and carrier-bead agitation increases with increasing roller velocity; as a result the developed mass per unit area should exhibit a superlinear dependence on roller velocity, as indicated in figure 3. This behavior distinguishes powder-cloud development from the other models and can be used to test for the presence or absence of this development mechanism.

Predicting the outcomes of the other sets of measurements requires additional assumptions about the forces exerted on the toner particles. If development of toner depends only on the force exerted by the electric field on the toner, then developed mass per unit



Models of development. The three models predict different behaviors of measurable quantities, as shown in the lower three graphs. The powder-cloud model (A) is the only one in which development is a nonlinear function of roller velocity. The equilibrium model (B) and field-stripping model (C) predict different behaviors of developed-toner mass and charge versus voltage. Figure 3

Measurements of development. The graphs show the behavior of developed mass and charge as a function of roller velocity and bias voltage for an IBM 6670 electrophotographic printer. The data are consistent with the equilibrium model.[3] Figure 4



area should be linear in the applied voltage. If the amount of toner freed from the carrier depends on the inertial forces alone, the developed-toner charge-to-mass ratio should be independent of the electric field. On the other hand, if the release of toner from the carrier depends on both inertial forces and toner charge (for example, via toner adhesion), one expects more complicated behavior.

The equilibrium model assumes[3] that toner continues to come off the carrier particles until the Coulomb attraction of the latent image balances the Coulomb attraction of the charged carrier beads, that is, until a force equilibrium is reached. As toner particles jump from the surface of the carrier bead to the latent image, the net charge on the carrier increases. Thus after n particles have left the bead (of radius R), the forces on a toner particle are QE toward the photoconductor and nQ^2/R toward the bead. The force of attraction (electrostatic or van der Waals) of the toner to a neutral carrier is ignored to first order in this model because it is approximately canceled by a similar force of attraction of the toner to the photoconductor during the time the toner particle is in contact with both the bead and the photoconductor.

The equilibrium model predicts that developed mass per unit area depends linearly on roller velocity because development increases linearly with the number of carrier beads brought into proximity with a point on the photoconductor. The model predicts that developed mass per unit area also depends linearly on the voltage because the number of toner particles transferred from each carrier bead is linear in the electric field. Because toner continues to develop until a force equilibrium is reached, independent of the applied voltage, the developed-toner charge-to-mass ratio should be independent of bias voltage.

In the field-stripping model, the Coulomb force due to the latent image on the photoconductor overcomes the forces (the electrostatic image force and van der Waals forces) that attract the toner to essentially neutral carrier beads.[2,3] All particles whose adhesion force is less than QE are developed, so the development curves are integrals over adhesion distributions. Toner particles with lower adhesion or lower charge develop first, so the developed-toner charge-to-mass ratio should increase with applied voltage. Because it takes a minimum force to begin to strip toner off the carrier particles, there should be zero development at low voltages, and because the Coulomb force develops all particles with adhesions less than QE, the development-versus-voltage curve should be nonlinear. The developed mass per unit area should, of course, be linear in roller velocity.

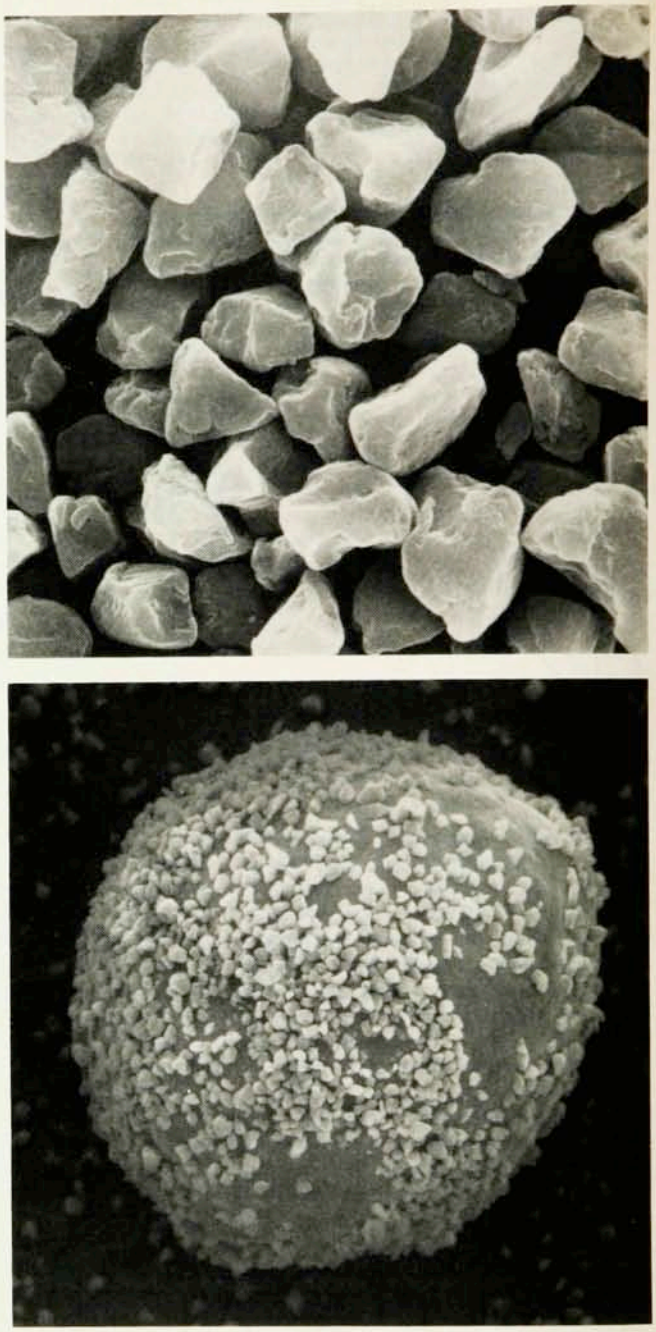
Improving development

In our laboratory at IBM we recently made[3] a series of measurements on a magnetic-brush development system from an IBM 6670 electrophotographic printer. The cover shows the magnetic brush; figure 4 shows the results of the measurements. Comparison of the predicted and observed results clearly shows that for this system the equilibrium model is the appropriate one. The linearity of the developed mass per unit area as a function of roller velocity indicates that powder-cloud development is not occurring, and the linearity of the developed mass with voltage suggests the equilibrium model is the appropriate one; this is confirmed by the observation that the developed-toner charge-to-mass ratio is independent of bias voltage. (The slight nonlinearity observed in the behavior of M/A above 800 volts can be accounted for[3] by a straightforward modification of the theory.)

Knowing the model that describes the development process, we can take a closer look at methods of improving the process suggested by the model. One way of improving an electrophotographic system is to make the printed black areas blacker, that is, increasing the mass of toner per unit area in the fully developed areas. According to the equilibrium model, the mass per unit area depends[3] on properties of the development system as follows:

$$\frac{M}{A} = \frac{V_0}{L}\,\frac{\epsilon_0}{Q/M}\,\frac{RN}{r/K_t + l/K_c}\left|\frac{v_r}{v_p}\right|$$

Here V_0 is the applied bias voltage, L is width of the gap between the photoconductor and the roller, Q/M is the toner charge-to-mass ratio, R and r are the radii of the carrier and the toner



Toner particles. These scanning electron micrographs show toner particles (upper photo) and toner on a carrier particle (below). The toner particles shown have been selected to be 10 ± 2 microns in size; in a commercial electrophotographic system the size variation is typically 1–30 microns. The carrier particle is a little over 350 microns in diameter. Figure 5

particles, l is the carrier coating thickness, N is the number of rollers, K_t and K_c are the dielectric constants of the toner and carrier, and v_r and v_p are the speeds of the roller and the photoconductor. (We have assumed that the photoconductor thickness is small compared with the dielectric thickness of the brush.) This relationship suggests several ways of improving the magnetic-brush development system. One can, for example, narrow the gap L, increase the number of rollers N, decrease the thickness l of the dielectric coating on the carrier beads, increase the carrier radius R or reduce the toner charge-to-mass ratio Q/M.

Each of these possible changes has a penalty associated with it. Narrowing the gap or increasing the number of rollers significantly increases manufacturing costs because parts must be machined to closer tolerances, or more and more complex parts must be put into the system. Other changes may, for example, involve compromises in the materials used in the system. However, the physical model that is now available for the development process provides a basis for systematically examining improvements in the electrophotographic process and a tool for designing new systems for optimum performance.

Charging toner particles

In discussing development, we assumed that the toner is charged. In fact the analysis above shows that one method of increasing the development M/A is to decrease the toner charge-to-mass ratio Q/M. The source of the toner charge is triboelectrification: an electrostatic charge exchange between the toner and carrier surfaces as they rub together in the hopper before being dispensed for developing.[4,5] The phenomenon is familiar to anyone who has touched grounded metal after walking across a rug in a dry room or whose hair clings to a comb in the winter. As a result of this electrostatic—or triboelectric—charging, toner and carrier acquire opposite charges and the small toner particles adhere to the larger carrier beads.

Figure 5 shows scanning electron micrographs of toner particles and of toner particles on a carrier bead. The toner particles are composed of a blend of polymers and carbon-black pigment, and the carrier particles consist of a magnetic core and a thin polymer coating. Depending on the chemical nature of this coating, the charge on the toner particles may be negative or positive.

Improving development thus requires control of the toner charge. Knowledge of the physics of contact electrification is crucial to this effort, yet the physics of electrification of insulators by insulators is very poorly understood.[5] One can appreciate some of the difficulty of studying contact electrification by considering the average amount of charge carried by a toner particle: A 10-micron particle typically carries 10^{10} electron charges per square centimeter but has an atomic surface density of 10^{15}–10^{16} atoms/cm^2—that is, only a few atoms in a million are actually charged. Such low charge concentrations are well below the levels detectable with present-day surface-science tools.

At present the most is known—both experimentally and theoretically—about contact electrification for metal–metal contacts, and successively less for metal–insulator and insulator–

Triboelectric series

Positive*
Silicone elastomer with silica filler
Borosilicate glass, fire polished
Window glass
Aniline–formol resin, acid catalyzed
Polyformaldehyde
Polymethylmethacrylate
Ethylcellulose
Polyamide 11
Polyamide 6-6
Rock salt (NaCl)
Melamine formol
Wool, knitted
Silica, fire polished
Silk, woven
Polyethylene glycol succinate
Cellulose acetate
Polyethylene glycol adipate
Polydiallyl phthalate
Cellulose (regenerated) sponge
Cotton, woven
Polyurethane elastomer
Styrene–acrylonitrile copolymer
Styrene–butadiene copolymer
Polystyrene
Polyisobutylene
Polyurethane flexible sponge
Borosilicate glass, ground surface
Polyethylene glycol terephthalate
Polyvinyl butyral
Formo-phenolique, hardened
Epoxide resin
Polychlorobutadiene
Butadiene–acrylonitrile copolymer
Natural rubber
Polyacrylonitrile
Sulfur
Polyethylene
Polydiphenylol propane carbonate
Chlorinated polyether
Polyvinylchloride with 25% DOP
Polyvinylchloride without plasticizer
Polytrifluorochloroethylene
Polytetrafluoroethylene
Negative

* When two of these materials are rubbed together the upper one in the list becomes positively charged, the lower one negatively charged.

From reference 8

insulator contacts. In electrophotography we are interested in insulator–insulator electrification.

A straightforward model for metal–metal contact electrification involves a simple equilibration of Fermi levels; as the metals are separated, the levels remain in equilibrium out to a distance of 10 Å, where the tunneling currents vanish. Thus two metals whose work functions are ϕ_A and ϕ_B develop a contact potential

$$V_c = (\phi_A - \phi_B)/e$$

The magnitude of the charge exchanged between the metals is equal to the capacitance at 10 Å times V_c. Figure 6 compares the predictions of this model with experimental results. One can obtain even better, quantitative agreement between theory and experiment by taking into account the roughness of the metal surfaces.

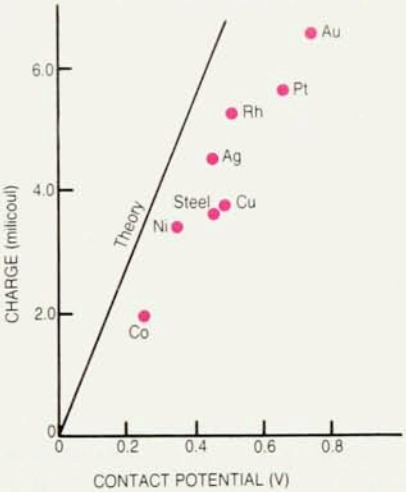
For the metal–insulator system, the situation is not nearly as clear.[5] One of the problems is the considerable uncertainty about how mobile the charges in the insulator are. Also, the concept of a Fermi level may have little meaning in an insulator, where excess charge can be trapped in nonequilibrium situations for long times. Experiments to clarify the situation, however, sometimes produce apparently contradictory results.

For example, consider the two sets of experiments on metal–insulator contact electrification whose results are shown in figure 7. In experiments performed[6] at Xerox, Tom Fabish and Charles Duke repeatedly brought into contact two smooth plates, one a metal, the other a charged insulator. The charge remaining on the insulator was measured periodically. Fabish and Duke found that a given metal produces a specific change in the magnitude of the insulator charge after a sufficient number of contacts. This change is independent of whether the insulator has previously been in contact with other metals and adds to whatever change has resulted from previous contacts. Fabish and Duke explained their results by suggesting that the insulator has a range of spatially localized energy levels near the contacting surface. When the insulator makes contact with a metal, electrons with energies near the Fermi level of the metal are exchanged between them. A given metal thus depletes the available states in the insulator within an energy window about the metal's Fermi level. Because the insulator states are localized they cannot be refilled. Thus each different metal depletes or fills a different set of localized insulator states.

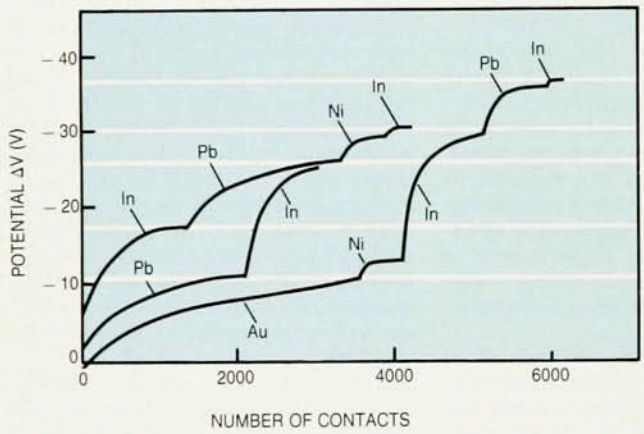
G. Cottrell and his coworkers at Manchester investigated[7] the same problem using a slightly different experimental approach. To ensure that the polymer was contacting the same microscopic region of metal surface each time, they used a hemispherical polymer surface in contact with a plane metal sheet, making an effort to maintain the relative positions of the two surfaces. Their results, shown at lower left in figure 7, do not agree with those of Fabish and Duke: They show no additivity but rather indicate that each metal seeks to establish a specific contact potential. Note, for example, that Pt "wants to" leave the polymer surface neutral, whereas Mg charges it negative. These results are what one would expect if there were some mobility of electrons and holes on the insulator surface.

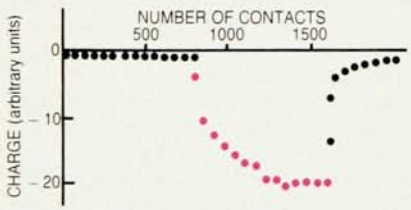
In the case of insulator–insulator contact electrification there are not even such detailed experiments or theoretical ideas to discuss. At the very least one would like to know the sign of the charge produced on insulator *A* when it is brought into contact with insulator *B*. One suggestion[8] is to arrange insulators (along with metals) in an empirical triboelectric series such as the one shown in the table at left: Materials higher up in the series become positively charged on contact with materials lower down in the series. Unfortunately there is no universal agreement on the ordering of insulators. In some cases the series appears[5] to be a ring: For example, while silk charges glass negative and glass charges zinc negative, zinc charges silk negative—much like the old paper–scissors–stone game!

The difficulty of doing definitive experiments in this area should not be underestimated. The experimental results depend on microscopic details of the contacting surfaces, such as surface roughness and chemical composition. Small levels of impurities on the surfaces being investigated can obscure the results. The electronic states in the insulators are not well defined. Nevertheless progress in understanding insulator–insulator charging would be highly useful, not only for electrophotography but also for a variety of processes that involve electrostatic charg-



Electrostatic charging for metal–metal contacts. The charge on a chromium ball ($^5/_{32}$ inch in diameter) touched once to a metal plate is shown as a function of the contact potential developed between the ball and the plate.[4] Figure 6





Repeated charging. Two experiments give apparently contradictory results. In one[6] (above), a polystyrene disk was repeatedly touched to a metal surface; the metal was changed from time to time. The other[7] (left) involved a hemispherical piece of PMMA polymer making contact with platinum, then magnesium and then platinum again. Figure 7

ing, such as dust precipitation, oil beneficiation, spray painting and reduction of sparking.

Charge transport in photoconductors

We now turn to our final example of the physics of electrophotography, the transport of charge carriers across the photoconductor—one of the several processes that occurs in the production of the latent image (during the exposure step). Light striking the photoconductor generates electrons and holes that can travel through the normally insulating material. The photoconductor is initially uniformly charged; exposure to light thus leads to charge dissipation from the illuminated regions to the ground plane, forming the latent image.

Because the processes involve aspects of solid-state physics that are fairly well understood, photoconductor physics has received by far the most attention in the literature on electrophotography. Despite this attention, it is fair to say that we do not understand in detail the microscopic mechanism of charge transport in these systems.[9] Several barriers remain: From a theoretical point of view, it is difficult to handle the asymmetries in systems in which the carrier energies vary from site to site both randomly because of disorder and systematically because of the electric field. From an experimental point of view, it is extremely difficult but imperative to separate the consequences of intrinsic disorder from those of chemical impurities and physical defects.

The overwhelming majority of photoconductors in use today are composed of amorphous materials such as chalcogenide glasses (amorphous selenium and its alloys) and molecularly doped polymers. Recent interest has centered on amorphous silicon as a potential photoconductor because it is much harder than both the organic and the chalcogenide materials. A detailed understanding of charge generation and transport would clearly be of great help in improving current photoconductors and designing new ones.

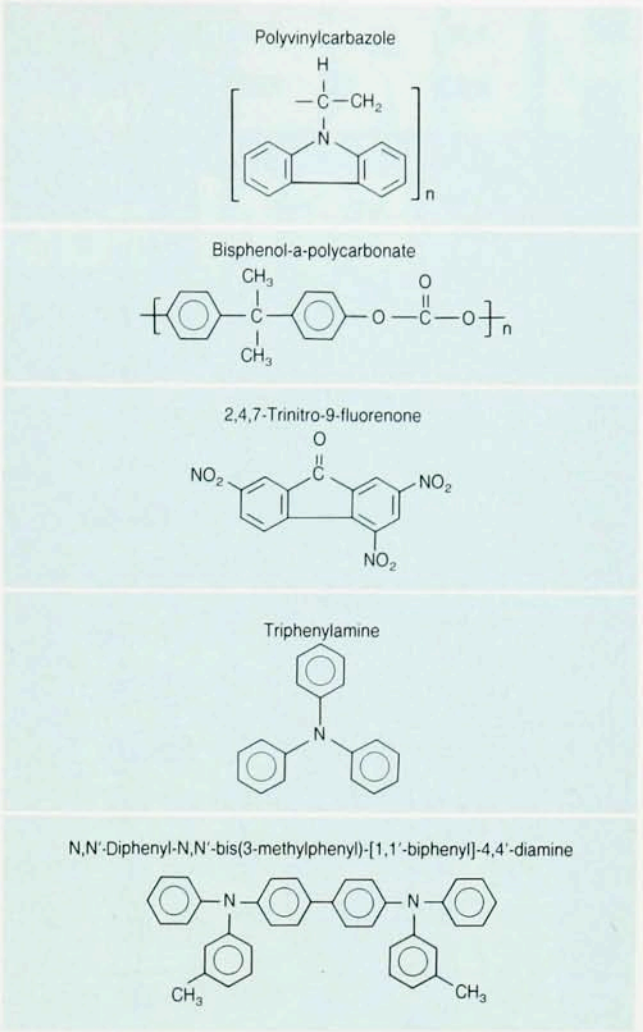
An ideal photoconductor has:

- A high quantum efficiency for conversion of light to separated electrons and holes (close to 100% has been achieved)
- A charge-carrier mobility large enough that the carriers leave the sample in a small fraction of the electrophotographic process time of about 1 sec
- A sufficiently low conductivity in the dark that the latent image remains on the photoconductor longer than the process time
- A low enough concentration of trapping sites that the latent image is not affected by previous latent images.

Here we have only enough space to focus on the physics of charge mobility in molecularly doped polymeric photoconductors. The interested reader can find the other topics discussed in the extensive literature on photoconductivity.[9,10]

The canonical experiment in studying charge transport is transient photoconductivity: A highly absorbed laser flash generates a sheet of mobile charge carriers; one then determines the drift mobility (that is, the carrier velocity per unit electric field) from the average transit time across the sample and the dispersion of the sheet (which can be due to an intrinsic distribution of transfer times between molecules or to extrinsic effects such as trapping).

In molecularly doped polymers such as those whose chemical compositions are shown in figure 8, the carrier mobility depends on temperature, electric field and the identity and



Polymer systems useful as photoconductors. The materials commonly used are polyvinylcarbazole doped with trinitrofluorenone (PVK–TNF) and polycarbonate doped with triphenylalanine (TPA–polycarbonate) or *N,N'*-diphenyl-*N,N'*-bis(3-methylphenyl)-[1,1'-biphenyl]-4,4'-diamine (TPD–polycarbonate). Figure 8

amount of the dopant molecule. The mobility is[9] in general very small, in the range 10^{-4}–10^{-9} $cm^2/V\,sec$, and depends exponentially on the distance between the dopant molecules. Such a mobility is much too small for band transport, and the exponential dependence on distance between molecules suggests that carriers hop among dopant molecules via overlapping wavefunctions, whereas the binder polymer is inert. One can fit[11] empirically the dependence of the mobility μ on electric field E and temperature T with the equation

$$\mu = \mu_0 \exp\left[-(\Delta - \beta E^{1/2})\left(\frac{1}{kT} - \frac{1}{kT_0}\right)\right]$$

where Δ is an activation energy, T_0 is an empirical parameter obtained from the data, k is Boltzmann's constant and β is constant in some systems but varies with dopant concentration in other systems. While an activated process is a very reasonable mechanism for hopping conduction, the nature of Δ remains unclear: It could reflect disorder, molecular vibrations, a polaron binding energy or a trapping energy. The field dependence is also not understood. While the square-root dependence suggests that the electric field lowers the top of the Coulomb potential (by what is called the Poole–Frenkel mechanism), virtually all authors in the field have argued against this mechanism on the grounds that the large number of Coulomb traps required cannot exist undetected in the sample. Concepts based on electric-field-dependent energy shifts between dopant sites either predict stronger field dependences than are observed or cannot account for the puzzling existence of T_0.

Heinz Bässler of the University of Marburg has recently suggested[10] that the carriers are hopping within a Gaussian distribution of energy states. Monte Carlo simulations as well as more recent theoretical work have shown that this mechanism gives a temperature-dependent activation energy. The temperature and field dependences are given by

$$\mu = \mu_0 \exp(-T'/T)^2 \exp(E/E_0)$$

where μ_0, T' and E_0 are empirically determined constants. This relationship appears to fit some of the earlier data, such as those on TNF-doped polyvinylcarbazole and TPA-doped polycarbonate, but it may not be consistent with recent data on TPD in polycarbonate.[12]

Clearly, many questions remain to be answered before we can claim to understand charge transport in molecularly doped polymers. The same holds true for other types of amorphous photoconductors.[9]

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