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New Model of Charge Generation and Latent Image Degradation in Single Layer Organic Photoconductor

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Abstract. *A new model of charge generation process in the single layer organic photoconductor (OPC) was proposed where the charge generation took place at the surface of the photoconductive layer after phonon diffusion from within the photoexcited bulk photoconductive layer. The degradation of the edge sharpness of the electrostatic latent image was simulated theoretically according to this model and the parameters that dominated the resolution degradation in the single layer OPC due to the energy diffusion were determined. The analytical result suggested the model of edge degradation in this system depended strongly on the phonon diffusion length. Accordingly, an experimental analysis was performed that explained the dependence of diffusion length on the photoreceptor formulation. The experimental result showed the validity of the principle in the proposed model for all OPC formulations tested, and the existence of a significant dependence of diffusion length on the CGM concentration in the photosensitive layer. The obtained results numerically showed the superiority in resolution of the single layer OPC compared with those of the conventional dual layer OPC, suggesting the possibility of true* 2400 dpi *imaging. The results provided a guideline for realizing higher resolution imaging via the single layer OPC formulation.* © *2006 Society for Imaging Science and Technology.* [DOI: 10.2352/J.ImagingSci.Technol.(2006)50:6(503)]

INTRODUCTION

One of the major trends in the recent electrophotographic apparati, such as laser beam printers (LBPs), must be high resolution imaging,[1] having advanced from the 240 or 300 dpi resolution levels of the early generation LBP to the current main stream of values of around 600 dpi. Besides, every company in this commercial area is aggressively pursuing the development of true 1200 dpi or higher resolution.[2] However, with the increase of the mechanical resolution in the apparatus, more attention is being devoted to the problem of the electrostatic latent image degradation on the organic photoconductor (OPC) surface due to the carrier diffusion. In the case of conventional dual layer OPC, the latent image is formed after the holes that are generated at the charge generation layer migrate through the thick charge transport layer (CTL) and neutralize the negative surface charges. Therefore, the resolution of latent image may become degraded because of the lateral migration of carriers due to the Coulomb repulsion of charges in the carrier cloud and lateral attraction by surface charges.[3,4]

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Single layer OPC is advantageous in this regard because of shorter distance between the charge generation site and the surface charge to degrade the electrostatic latent image.[5,6] This viewpoint is based on the conventional hypothesis that charge generation occurs at the photoexcited CGM pigments that are mostly located in a region close to the surface and based on distribution of light absorption in the layer. In this case, the short migration distance of electrons from the excited CGM particles in the layer to the positive surface charges determines image degradation, like a positively charging single layer OPC construction consisting of CGM particles dispersed in CTM/polymer matrix is employed.

As we discussed in the previous paper,[7] we propose a different process of charge generation in single layer OPC, where the energy of incident light absorbed in the bulk layer propagates to the charge generation site at different locations and charge generation occurs near the surface of the OPC. According to this assumption, even the unexposed area of the surface would inevitably receive the influence of energy propagation from bulk, and the electrostatic latent image on the surface would be degraded to a certain extent, even though such energy propagation may be electrically neutral and not affected by the electric field. It is necessary to review the degradation mechanism of latent image according to this model and to correctly estimate the underlying factors of image degradation to identify a formulation to obtain high resolution, making this basic concept quite different from the model describing image resolution conventional OPC.

This paper will first discuss the mechanism of edge degradation of latent image in single layer OPC using a theoretical analysis based on the assumed model, prove the important role of phonon diffusion length in the resolution, and then clarify the compositional dependence of phonon diffusion length by analyzing experimental results on spectral sensitivities of various formulations. Finally, a discussion is presented on formulation optimization to obtain a high resolution, single layer OPC and the resolution limit by the structural potential of single layer OPC.

THEORY OF IMAGE DEGRADATION

A simplified light exposure model is proposed to describe theoretical image degradation process. As a first approximation, the energy distribution of incident light is assumed to produce a square shape on the surface of OPC layer and that

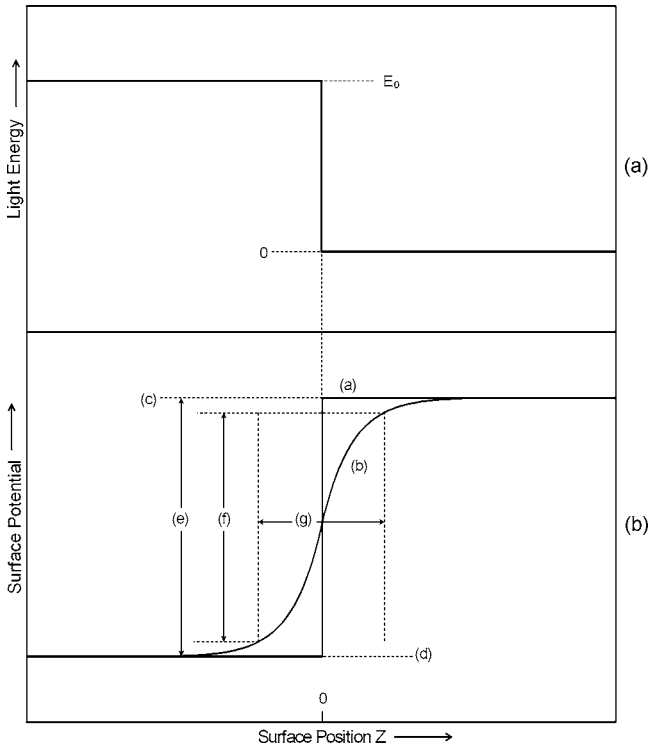

Figure 1. (a) Schematic illustration of light energy distribution at OPC surface, (b) illustration of edge degradation of latent image: (a) ideal potential profile, (b) degraded potential profile according to Eqs. (5) and (6), (c) dark potential, (d) light potential, (e) contrast potential, (f) practical contrast potential, and (g) edge width.

the light penetrates the layer orthogonal to the layer surface without scattering. The boundary point of light exposure is at the surface position $z=0$ on the layer, where light energy $E=0$ when $z\geqslant 0$, and $E=E_0$ when $z<0$, as illustrated in Fig. 1(a).

The phonon number at any surface position, z, transferred from the bulk layer can be determined utilizing the same energy transfer mechanism proposed in the previous paper.[7] The phonon number at any place on the surface, due to the exposure by a small beam of incident light, is determined by Eq. (1)

$$n(a)=\int_0^l A\exp(-\alpha x)\exp\left(-\frac{\sqrt{x^2+a^2}}{L}\right)dx, \qquad (1)$$

where the parameter, a, denotes the distance of the surface position from the incident position, depth position, x, the distance from the surface into the bulk layer, the phonon diffusion length L, absorption coefficient α, and a constant A.

For simplicity, $\sqrt{x^2+a^2}$ is replaced by $x+a$. The integral in Eq. (1) can be resolved using inversed diffusion length β as follows:

$$\begin{aligned} n(a) &\approx \int_0^l A\exp(-\alpha x)\exp\{-\beta(x+a)\}dx \\ &= \int_0^l A\exp(-\beta a)\exp\{-(\alpha+\beta)x\}dx \\ &= \frac{A}{\alpha+\beta}\exp(-\beta a)[1-\exp\{-(\alpha+\beta)l\}] \\ &\approx \frac{A}{\alpha+\beta}\exp(-\beta a). \end{aligned} \qquad (2)$$

When $z\geqslant 0$ in the case of Fig. 1(a), the total phonon number $N(z)$ at the position, z, is obtained by the integration

$$N(z)=\int_z^\infty n(a)da=\frac{A}{(\alpha+\beta)\beta}\exp(-\beta z). \qquad (3)$$

On the other hand, when $z<0$, it will be as follows:

$$N(z)=\int_0^\infty n(a)da+\int_z^0 n(-a)da=\frac{A}{(\alpha+\beta)\beta}\{2-\exp(\beta z)\}. \qquad (4)$$

Figure 1(b) illustrates the potential profile at the image edge using the above equations, where the surface potential at each area can be determined from Eqs. (3) and (4) as follows, assuming that the discharge of surface charge is simply proportional to phonon number $N(z)$

$$V(z)=V_0-\frac{V_0-V_R}{2}\exp(-\beta z) \qquad (5)$$

when $z\geqslant 0$, and

$$V(z)=V_R+\frac{V_0-V_R}{2}\exp(\beta z) \qquad (6)$$

when $z<0$.

In Fig. 1(b), undisturbed ideal potential profile (a) has a square edge reflecting the light power distribution model on the surface. Actual potential profile (b) receives the influence of energy diffusion and has some slope in accordance with Eqs. (5) and (6) disturbing edge distinctness. Here, the resolution is defined as the edge width (g) which can hold the practical potential (f) to a predefined ratio with the maximum contrast potential (e). It must be noted that the resolution is determined only by the combination of surface positive charges and the generated negative charges, so the usual parameters like carrier mobility and layer thickness, which affect the resolution in the dual layer OPC, may not take part in determining the resolution of supposed model, because the generated electron combines with the surface charge almost in place. These parameters can only affect the discharging speed in our model.

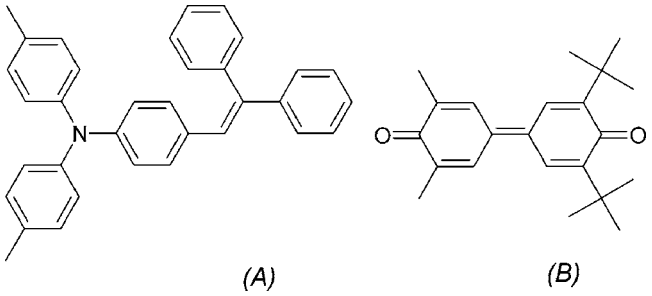

Figure 2. Employed charge transport materials.

Defining p to be the ratio of the practical contrast potential to the maximum obtainable value ($p=(f)/(e)$), then the resolution R can be determined from Eqs. (5) and (6) in the following equation:

$$R = -2L\ln(1-p). \tag{7}$$

This estimate would give an approximate edge structure of the latent image for consideration of the resolution limit. The actual laser beam has a Gaussian profile, not the square shape as described here, making the discussion of actual resolution a little more complicated.

As discussed above, the edge degradation by energy transfer from the bulk layer in single layer OPC can be described as the function of phonon diffusion length. Better resolution can be obtained by employing an OPC with shorter diffusion length. The diffusion length is dependent on the physical parameters of the layer matrix, which is related to the layer formulation. Since the phonon diffusion length is an important factor predicting the image degradation problem in the proposed model, then a better understanding of this dependence on OPC formulation is necessary. Among various parameters of the OPC formulation, the most effective parameter for diffusion length is the CGM pigment concentration because of its phase discreteness in the matrix. Accordingly, the following experiment was carried out to estimate the dependence of diffusion length on CGM concentration.

EXPERIMENT

Single Layer OPC

The OPC formulation was composed of X-form metal-free phthalocyanine (X–H_2Pc) (Ref. 8) CGM pigment, hole transport material (HTM) formula (*A*) (Ref. 9) and electron transport material (ETM) formula (*B*) (Ref. 10) dissolved with polycarbonate-Z resin solution to make a macroscopic homogeneous transparent active matrix (Fig. 2). The purpose for the ETM addition in this system is to keep the electrical performance of the OPC stable by allowing the generated electrons to be released from charge generation site and to combine with surface positive charges. The amount of ETM was limited for the least concentration required for this purpose.

The samples of single layer OPC were coated on bare aluminum drums in the same manner as described in the previous paper,[7] with the following four formulations containing different CGM concentrations:

(i)
CGM/HTM/ETM/binder=0.6/40/10/49.4,
(ii)
CGM/HTM/ETM/binder=1/40/10/49,
(iii)
CGM/HTM/ETM/binder=2/40/10/48, and
(iv)
CGM/HTM/ETM/binder=4/40/10/46.

The dried film thickness for these samples was 14–15 μm.

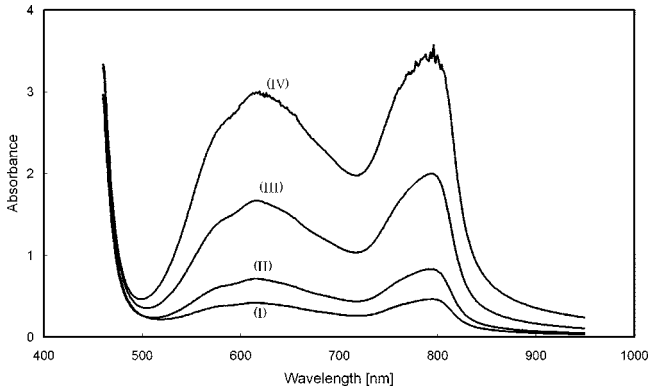

Figure 3. Absorption spectra of OPC films for formulations (i)–(iv).

Spectral Sensitivity

The measurements of spectral sensitivities were carried out for all four samples using an electrophotographic drum analyzer, Cynthia model 91 of Gentec Co., Japan. The details of the measurement conditions are described in the previous paper. In the measurement, the sensitivity is defined as the photodischarging rate for 2 μW/m^2 monochromatic light when the sample was charged to 500 V in absolute value, as expressed by following equation:

$$S = \left| \frac{dV_D}{dt} - \frac{dV_L}{dt} \right|, \tag{8}$$

where V_D and V_L mean the dark and light potentials just before and after the start point of light exposure, respectively.

Absorption Spectra

The UV-visible spectra of OPC films were directly measured by a UV-visible spectrometer, model 8453 from Agilent Technologies, for 17–18 μm thick samples prepared by spin-coating solutions on a glass substrate.

RESULTS

Figure 3 shows the absorption spectra of the OPC layers of four single layer formulations. The absorbance increased proportionally to CGM concentration in the wavelength range longer than 550 nm.

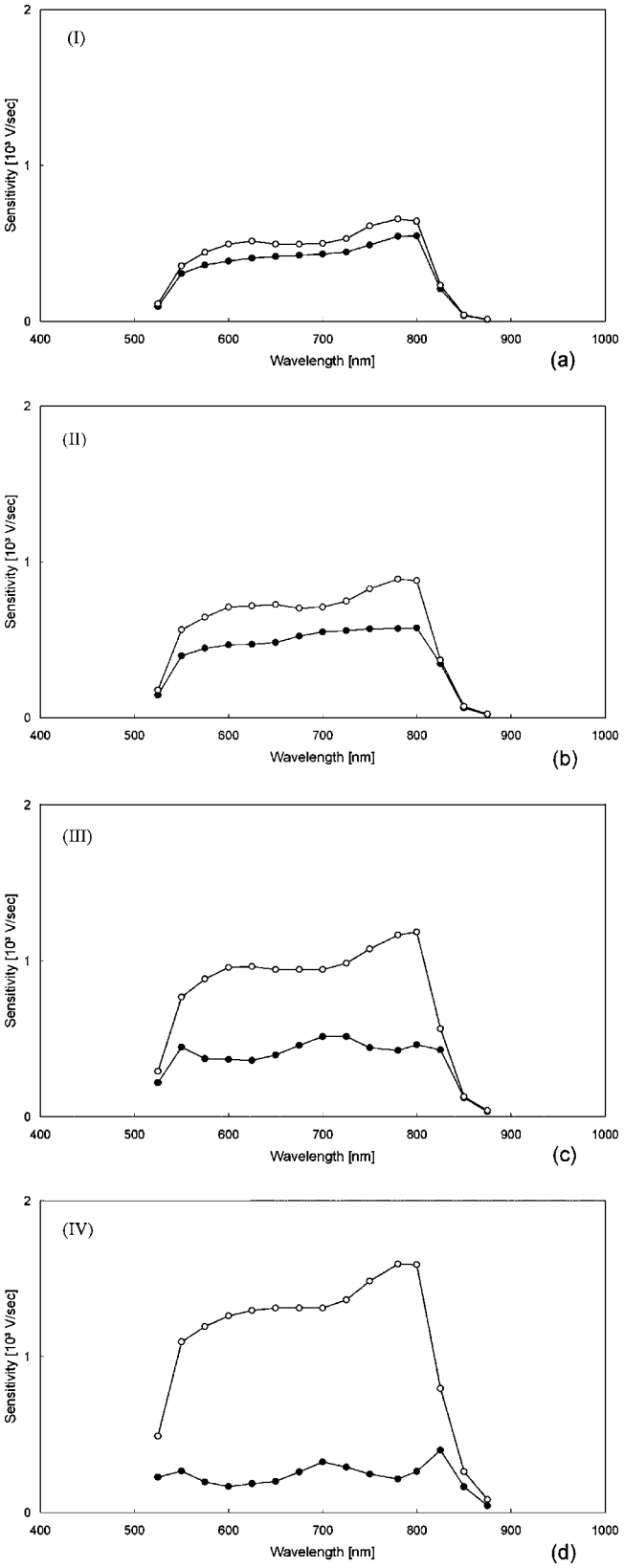

Figure 4. (a)–(d) Spectral sensitivities of both polarities: Open circles show the positive charging sensitivities and solid circles show the negative charging sensitivities; CGM concentration increases from (a) through (d).

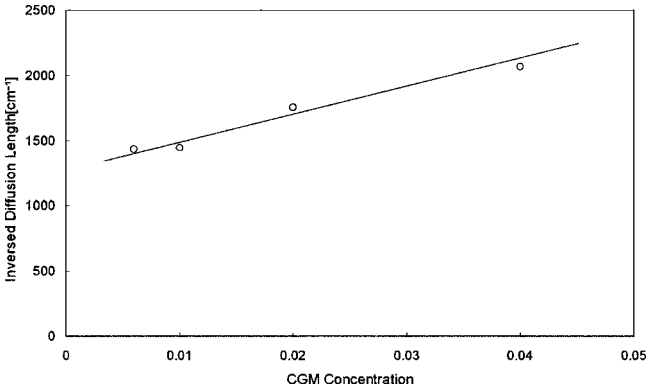

Figure 5. Inverse diffusion length β as the function of CGM concentration from date of Fig. 4.

Figures 4(a)–4(d) are the spectral sensitivities of all samples for both polarities. In the very low CGM concentration sample, both sensitivities were comparable in absolute values and the spectral dependences were similar to each other. However, the difference of both sensitivities became more pronounced by increasing CGM concentration, where the positive charging sensitivity increased almost linearly without changing spectral shape. On the contrary, the negative charging sensitivity decreased in absolute value with increasing CGM concentration and, simultaneously, the characteristic spectral dependence of negative charging sensitivity appeared significantly different at high CGM concentration samples showing peaks at the absorption valley and shoulder slopes as described in the previous paper. These results prove the important role of the light transmittance of the layer in realizing the negative charging sensitivity and different locations of charge generation sites by the charging polarities as suggested in the previous paper.

DISCUSSION

Diffusion Length Dependence

According to our previous paper,[7] we can estimate the phonon diffusion length by solving Eq. (9)

$$\frac{S_{n1}S_{p2}}{S_{n2}S_{p1}} = \frac{(\beta+\alpha_1)(\beta-\alpha_2)\{\exp(-\alpha_1 l) - \exp(-\beta l)\}}{(\beta-\alpha_1)(\beta+\alpha_2)\{\exp(-\alpha_2 l) - \exp(-\beta l)\}}, \tag{9}$$

where l is the layer thickness, and S_{n1}, S_{p1}, α_1 and S_{n2}, S_{p2}, α_2, are the set of negative and positive sensitivities and absorption coefficients at two different wavelengths, respectively. Substituting those values at 600 and 725 nm for each result of tested samples, we could obtain the value β for all OPC formulations. Figure 5 shows the dependence of β on CGM concentration thus obtained, where a clear correlation can be observed showing linear increase of β value on increasing CGM concentration.

This result suggests that the phonon propagation in the CTM/binder matrix is strongly disturbed by the presence of CGM pigments, where the magnitude of the disturbance

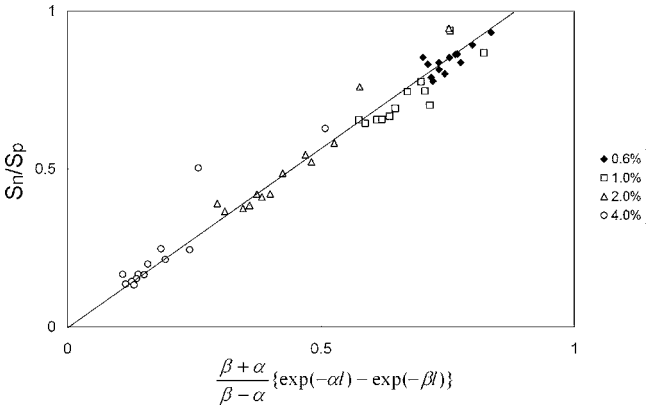

Figure 6. Correlation plot of (S_n/S_p) vs $[((\beta+\alpha)/(\beta-\alpha))\{\exp(-\alpha l)-\exp(-\beta l)\}]$. Dots are the calculated values from the data of spectral sensitivities of both polarities. Line is the least-squares approximation.

seems to be proportional to the density of CGM, so that the phonon diffusion length decreased with increasing CGM concentration.

Here, let us assume the existence of two different disturbance coefficients for phonon propagation, D_M of the CTM/binder matrix and D_P of CGM pigment and naturally $D_M < D_P$. The transmittance of phonon for the length d would be proportional to $\exp[-\{(1-\rho)D_M+\rho D_P\}d]$ using the value of CGM density ρ, so that the diffusion length L should be determined in the following equation:

$$L = \frac{B}{(1-\rho)D_M + \rho D_P}, \tag{10}$$

where B is a constant value. The above equation can be modified as follows by replacing L by β^{-1}

$$\beta = \frac{1}{B}\{D_M + (D_P - D_M)\rho\}. \tag{11}$$

Thus, the linear dependence of β on CGM concentration can be simply explained.

The adaptability of the values obtained for the supposed model of charge generation is verified by the following equation:

$$\frac{S_n}{S_p} = r\frac{\beta+\alpha}{\beta-\alpha}\{\exp(-\alpha l) - \exp(-\beta l)\}, \tag{12}$$

where r denotes the ratio of charge separation efficiency of negative charging to that of positive charging.

Figure 6 exhibits the correlation of (S_n/S_p) vs $[((\beta+\alpha)/(\beta-\alpha))\{\exp(-\alpha l)-\exp(-\beta l)\}]$ by substituting obtained values of β, sensitivities of both polarities, and absorption coefficients for all samples and wavelength.

As shown in Fig. 6, data of different formulations and wavelength are aligned on one line through the origin, which would indicate that relation (12) is consistently valid in the different formulations of single layer OPCs evaluated in this study. The parameter r is defined as the ratio of charge separation efficiency of negative charging to that of positive charging and is determined from the slope of the regression line in the graph. A slope that is larger than one suggests that the separation efficiency for negative charging is larger than for positive charging due to the different locations of charge generation sites as suggested in the previous paper.[7] In this case, the charge generation site is supposed to exist in the CGM pigments at the substrate-OPC layer interface, where portion of the pigments may directly contact the substrate, causing a large local field in the Schottky barrier at the contact point of X−H_2Pc and Al substrate[10] which assists the charge generation.

Resolution Limit of Latent Image

From the result of the experiment, it is whown that the phonon diffusion length is dependent on the CGM concentration and becomes shorter with increasing concentration, which means the edge degradation should be smaller. This result suggests that small edge degradation can be attained by increasing the CGM concentration, which correlates well with the experimental results obtained on latent image resolution in single layer OPC.[6] This agrees with the general point of view for this issue that is based on a conventional charge generation model and considers a shorter electron migration length from photoexcited CGM to the surface charge in higher CGM concentration systems.

In the prior study on dual layer OPC,[3,4] it was found that the resolution of dual layer OPC was strongly dependent on layer thickness, carrier density, mobility, image pattern, and it has been claimed that CTL thickness must be 10 μm or less in order to realize 1200 dpi resolution.[1,3] This thickness is insufficient to give proper charge acceptance in usual electro photographic (EP) system and to fulfill another technical demand for extending the usage life of OPC due to mechanical wear, so that the practical resolution limit of conventional dual layer OPC is ~1200 dpi or less.

On the contrary, the single layer OPC as discussed here is independent of those factors. The main factor affecting resolution is only phonon diffusion length according to our proposed model. Equation (7) shows the linear proportionality of edge degradation to diffusion length and it illustrates the importance for determination of necessary contrast potential (g) in estimating the resolution.

If we assume as a first approximation that the contrast potential should be more than 70% ($p=0.7$) of the maximum value for the single layer formulation with a 5 μm diffusion length, then R would become approximately 12 μm, calculated using Eq. (7). For the realization of 2400 dpi imaging, approximately 10 μm dot resolution may be necessary and this value seems to be a critical resolution limit for 2400 dpi imaging. However, considering that the developing potential is usually set much lower than the surface potential of the OPC, the actual contrast potential is supposed to be much smaller so that this resolution would be enough to realize 2400 dpi. This result corresponds well to the experimental value observed in a similar formulation single layer OPC.[6]

Let us suppose that the diffusion length is around 5 μm

(β=2000 cm^{-1}; cf. Fig. 5), and then the single layer OPC has the potential of achieving 2400 dpi or more with a thick OPC layer. Moreover, considering the dependence of diffusion length on CGM concentration, the higher resolution can be attained in the formulation with higher CGM concentration system, although it is not possible to increase the CGM ratio indefinitely because excessive amount of CGM ratio would usually degrade EP properties, such as charge acceptance, cycling stability, etc.

It must be noted that the analysis was based on the assumption that all of the charge generation occurred at top surface region and neglects electron transport from the bottom of the layer. This assumption is valid because electron mobility is much slower than hole mobility in the usual single layer OPC of this formulation. However, when ETM concentration is increased and electron mobility becomes comparable to hole mobility to some degree, then carrier migration from the substrate may not be negligible. In that case, the phenomenon of resolution degradation by lateral carrier migration as observed in conventional dual layer OPC will also appear even in the single layer OPC.

CONCLUSIONS

According to the new model of charge generation in the single layer OPC, the edge degradation effect in electrostatic latent image was discussed theoretically and experimentally. The results can be summarized as follows.

1. The potential profile of at the edge of latent image can be expressed as the exponential function of phonon diffusion length, where the edge width (or expansion) is proportional to the diffusion length.
2. The phonon diffusion length is dependent on the CGM density and becomes shorter with increasing CGM concentration.
3. The proposed model of charge generation has been consistently verified for various formulations of single layer OPC.
4. The resolution limit in a usual single layer OPC can be estimated to be 2400 dpi or more considering degradation width, which is a significient advantage in comparison to conventional dual layer OPC with the resolution limit around 1200 dpi.
5. Higher resolution is achieved by increasing CGM concentration in so far as the EP characteristics remain in practical working range.

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