Analysis of the Printing Process with Transfer of Toner Particles by Direct Light Irradiation

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We analyzed a printing method using direct toner transfer by a light flash in which the image-forming process was expected to be considerably simplified. First, we found the transferred amount of toner particles increased linearly with the irradiated energy and abruptly decreased at long pulse length. Next, we observed the toner movement using a high-speed camera and measured the transmissivity and current transition to clarify the toner transfer process. The toner transfer was caused by a thermal event, and from a temperature simulation we concluded was the ablation of toner occured. Comparison of experiment and simulation results showed the toner transfer started at the time when the inside maximum temperature reached a specific temperature, i.e., the decomposition temperature of the toner material.

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Introduction

Today's major printing technologies are based on electrophotography that has advantages of high resolution and high printing speed, but the disadvantage of complicated processes such as charging, exposure, developing, transfer, fixing, and cleaning. To eliminate these complications, a printing method that addresses the toner particles by meshed electrodes was studied.[1] In this process, toner particles were selected by the voltage supplied to the corresponding electrodes and transferred by electrostatic force. The size of the electrode mesh determines the process resolution.

Using light or laser beams to heat an ink ribbon on a thermal transfer printer is another approach to reducing the printing process complexity. A photoduplicating procedure was proposed in which pigment particles such as carbon black on a base, for example, a typewriter ribbon, were irradiated by high-intensity light and transferred to a receptor sheet.[2] Dye diffusion,[3] dye transfer,[4] and laser ablation transfer[5,6] printing were also studied. High-resolution images are formed by these thermal transfer methods. The ink ribbons, however, can be used only once.

Recently, we found a printing method in which toner particles on a transparent plastic sheet can be transferred by direct irradiation of high-intensity light.[7,8] The toner layer is irradiated by the flash of a xenon flash lamp through a mask. The toner particles irradiated in the flash are transferred to a paper facing the toner layer and patterns are formed on the paper. If we use a transparent endless belt or drum on which toner is carried and a controllable spatial modulator such as an LCD panel in place of the pattern mask, we can make a printing system.

The printing process will be as follows: (1) A toner layer is formed uniformly on a transparent belt or drum. (2) The toner layer is exposed to the high-power light image pattern formed by the controllable spatial modulator. The part of the toner exposed to light is transferred to the paper and a negative image pattern of the mask is formed. (3) The image pattern is fixed on the paper. The

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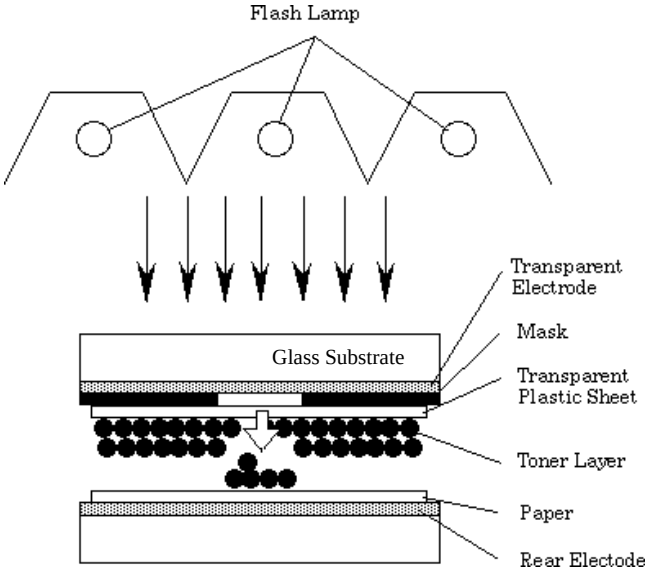


Figure 1. Experimental setup.

light energy that forms image patterns is high enough to melt toner, so the light can also be used to fix the toner by irradiating again. This process was proposed in a patent issued to Haas et al.[2] in 1970; we have added use of electrophotographic toner to deposit the layer on the transparent base by charging. Electrophotographic toner is easy to fix by heat and to supply using electrostatic force. This printer principle, using electrophotographic toner, would have many advantages. Light is used to address image pixels, so concentration onto a small point is possible. Because toner is exposed directly to the light modulated on the image information through the transparent belt or drum, no photoconductive member is needed. Moreover, charging and transfer devices are also unnecessary. Unlike the thermal transfer printers that use an ink ribbon, the toner layer can be regenerated cyclically.

In a preliminary study, we showed that our toner transfer was caused by light absorption of toner particles,[6,7] but the details of the transfer were not clarified. To elucidate the toner transfer process, we have measured the dynamics of the toner transfer and analyzed the toner temperature when irradiated by pulsed light.

Experiments

Evaluation of Amount Transferred . Figure 1 shows the experimental setup for the image-forming experiment.[7,8] A toner layer was formed uniformly on a transparent plastic sheet. A glass substrate with a transparent electrode was used to support the transparent plastic sheet on which the toner layer had been deposited. This toner layer was formed using a conventional copy machine from which we had removed the fixing device. Black copy machine toner was used. In the developing process of the copy machine, toner particles were charged. After the layer of toner particles was uniformly formed on the photoconductor drum, the toner particles were transferred to the plastic sheet. A piece of paper or another plastic sheet was placed on the rear electrode at a distance of about 200 μm from the toner layer surface. Toner was exposed to light through the substrate and the transparent sheet. A long tube-type xenon flash lamp was used to form large area images and evaluate the printing characteristics. The flash lamp (emission efficiency: 30%) was supplied with 200 to 440 J of electrical energy. The optical energy density was 0.32 to 0.73 J/cm^{2,} and the exposure time was 0.43 to 0.7 ms full width at half maximum (FWHM).

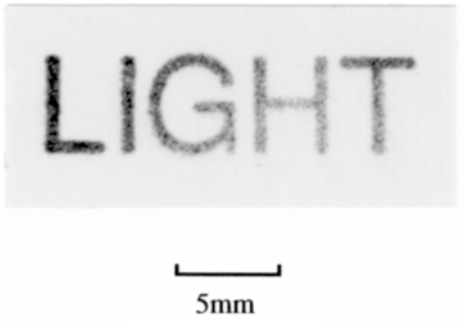


Figure 2. Image sample.

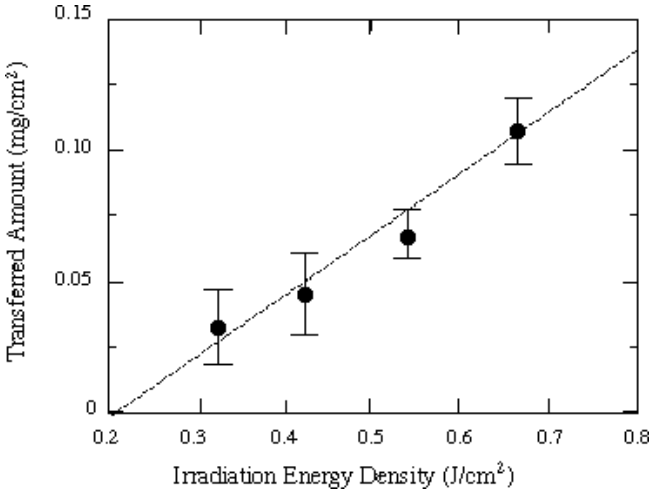


Figure 3. Amount of transferred toner as a function of the irradiation energy density.

A pattern mask was sandwiched between the glass substrate and the transparent plastic sheet to form images. The light image was irradiated onto the toner layer through the sheet. A part of the irradiated toner was transferred to the piece of paper that faced the toner layer, and a negative image pattern of the mask was formed on it. Figure 2 shows a sample image formed by the above method when irradiated for 0.43 ms (FWHM) by light with an energy density of 0.73 J/cm^2.

We measured the transferred amount of toner when the toner was uniformly irradiated by the flash lamp without the mask. The toner transferred to the plastic sheet facing the toner layer was moved by vacuum and weighed. Figure 3 shows the amount of transferred toner as a function of the irradiated energy density. The transferred amount increased linearly with irradiation energy density. The initial sticking amount was 0.76 mg/cm^2, so the maximum transfer toner ratio was about 15% at 0.73 J/cm^2. Because the transferred amount increased linearly with the irradiation energy density, gradation of images could be made by changing the irradiation energy density. This figure also shows that the threshold energy density at which toner could be transferred was 0.22 J/cm^2.

Figure 4 shows the amount of transferred toner as a function of the irradiation time in FWHM at the optical energy density 0.73 J/cm^2. The transferred amount was almost constant below 0.67 ms, but abruptly dropped at 0.7 ms.

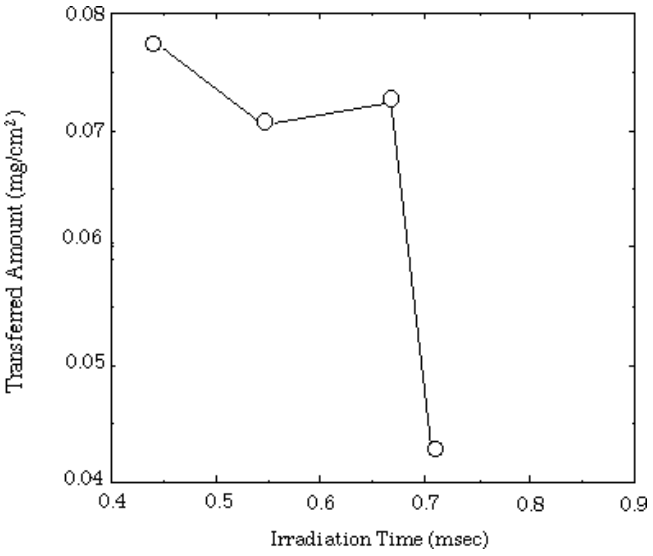


Figure 4. Amount of transferred toner as a function of the irradiation time.

Time-Resolved Measurement. Clarifying the mechanism of toner transfer was important for improvement of image quality. To confirm the time dependence of the toner movement, we performed two experiments: one involved observation of the irradiated part of the toner layer with a high-speed video camera and the other was transmissivity measurement. Figure 5 shows the transmissivity measurement and high-speed observation setup. In these experiments, we used a laser diode with a wavelength of 830 nm as a light source. A pulsed laser beam was focused on the toner layer and the laser intensity transmitted from the toner layer measured. The transmitted light intensity should increase as toner particles are transferred. Furthermore, a side view of the irradiated part of the toner layer was observed with a microscope and recorded on high-speed video tape (maximum recording speed: 1000 frame/s). The beam diameter was about 50 μm with average power density of about 2.5×10^3 W/cm^2 when the irradiation power was 50 mW, and the energy density on the sample for 0.3 ms irradiation was about the same as that from the flash lamp.

First, we observed the irradiated part by using the high-speed camera to reveal the motion of the toner particles when irradiated by light. Figure 6 shows three photographs recorded on the high-speed video tape. The white particles on the right side of Fig. 6(a) are toner particles. The laser pulse was irradiated from the right side through the transparent sheet (the transparent sheet was not shown in the figures). By irradiating a laser pulse onto the toner layer, we observed a jet of smokelike matter given off from it [Fig. 6(b)]. As time passed, the jet expanded. After irradiation, toner particles at the irradiated part disappeared [Fig. 6(c)]. The toner particles were presumably pushed by this jet from the toner layer.

Next, we measured the time dependence of the transmissivity to evaluate the toner transfer. Figure 7 shows the time dependence of the transmissivity at two different irradiation powers when the exposure was for 3 ms. A flat part in the transmissivity curve occurs just after irradiation commences. As the irradiation power increased, the duration of the flat part became shorter. After the flat part, transmissivity increased. This increase indicated that toner moved from the transparency sheet to the paper. We measured the duration of the flat part, which we defined as the transfer starting time, with varying irradiation power.

In Fig. 8, the circles show the experimental transfer starting time as a function of the irradiation power. When

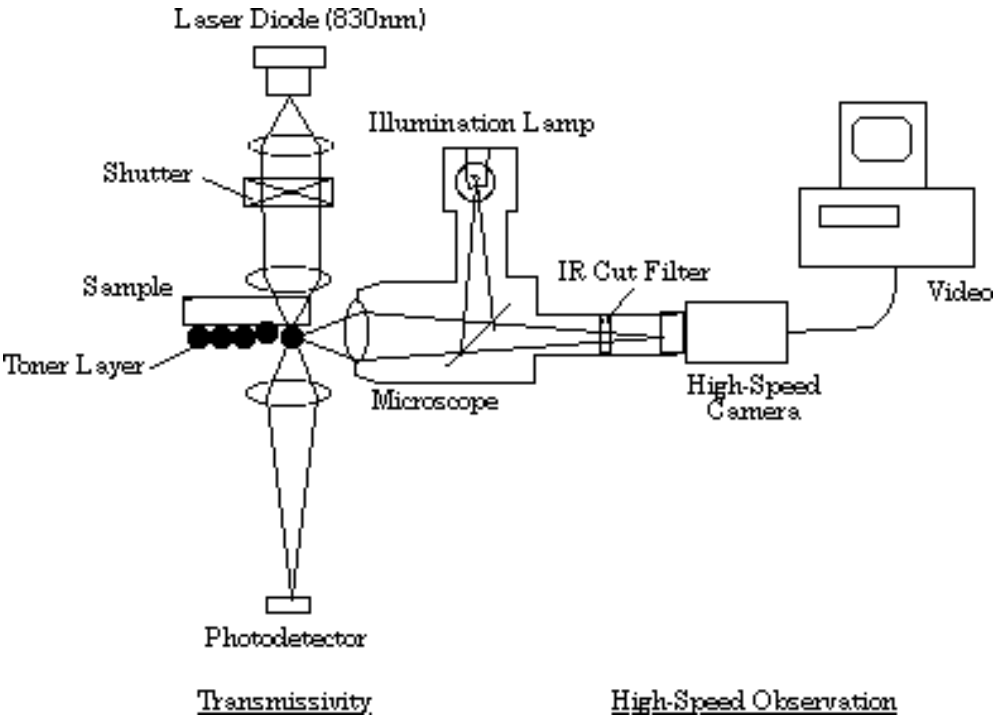
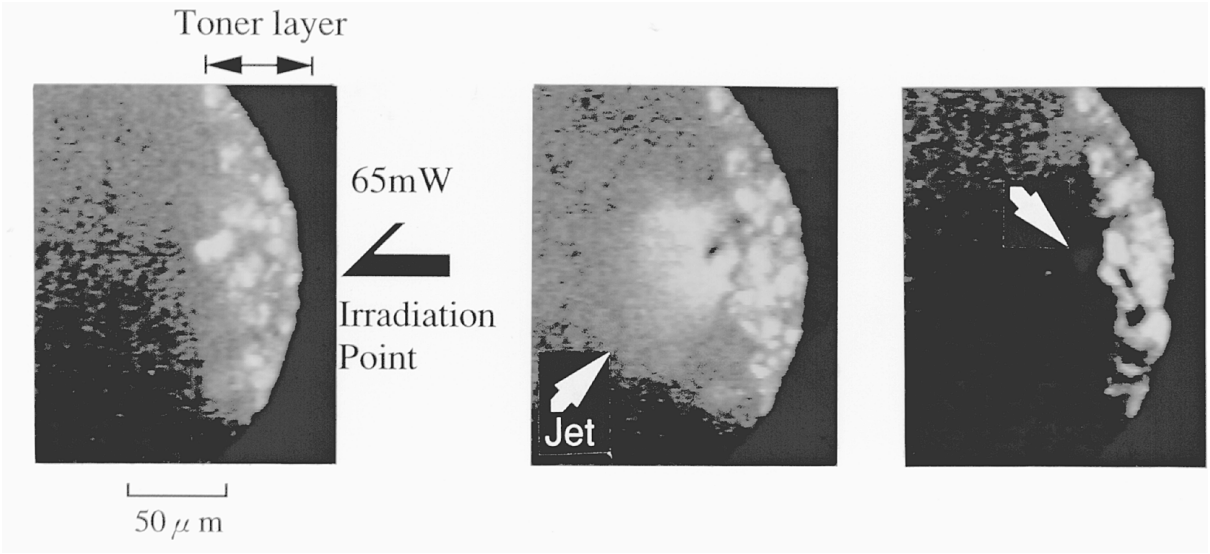


Figure 5. Transmissivity measurement and high-speed observation setup.



(a) Before irradiation **(b) 2 msec** **(c) After irradiation**

Figure 6. Pictures of high-speed observation (negative images).

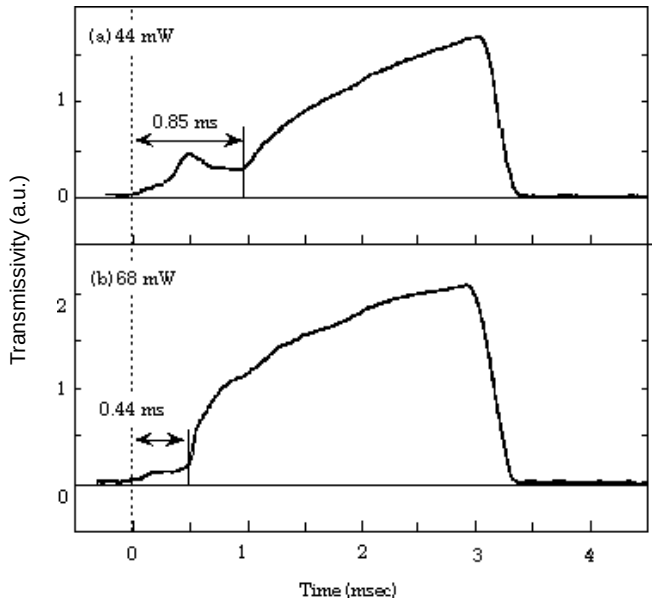


Figure 7. Time dependence of the transmissivity.

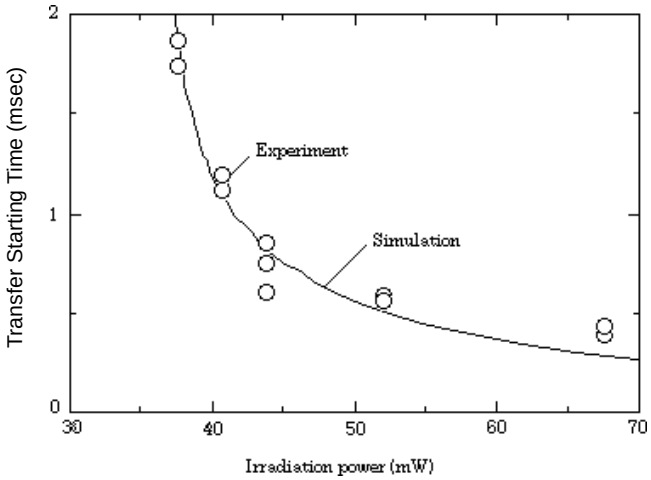


Figure 8. Transfer starting time as a function of the irradiation light power.

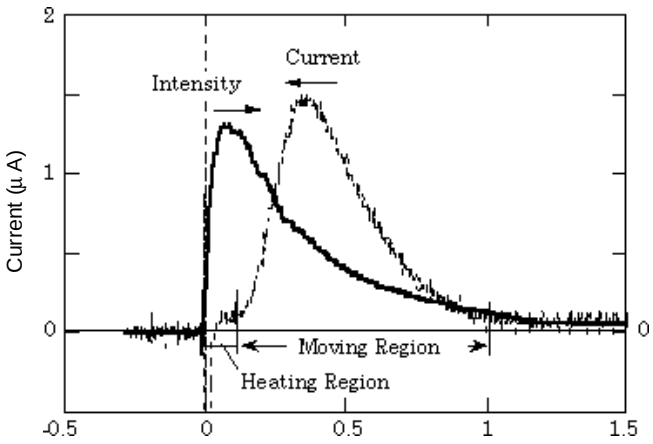


Figure 9. Time dependence of the current due to toner transfer and irradiated light intensity.

the irradiation power was low, the time interval before departure became long. The departure time increased steeply when irradiation power was less than 40 mW and seemed to become infinite at around 30 mW. This agreed with the result that a threshold energy exists for toner transfer as shown in Fig. 3.

Current Measurements. To evaluate the time dependence of the toner transfer when the toner layer was irradiated by the flash lamp, we measured the current flowing between the transparent electrode and the rear electrode (Fig. 1). The toner particles were charged in the process of toner layer formation. Corresponding to the movement of the charged toner particles (charge: – 5.0 μC/g), current flows between the electrodes. A short arc type of flash lamp was used in this experiment. Conditions were as follows: electrical energy supplied, 50 ~ 200 J; emission efficiency, 1%; exposure time, 0.18 to 0.36 ms (FWHM).

Figure 9 indicates the time dependence of the current and light intensity. There is a time delay between the onset of irradiation and the current increase. Again we saw a delay in the start of toner particle transfer as in the transmissivity measurements (Fig. 7). The current peak corresponding to the most toner particle transfer also was delayed relative to the light intensity peak.

Process of Toner Transfer. Time delay of transfer of the toner particles showed that a thermal event was occurring; i.e., the time delay showed the interval during which the toner particle was heated. We at first assumed the toner particles were pushed by the heated surrounding air, which was heated by conduction from the high-temperature particles. We, however, confirmed that the toner transfer occurred in a vacuum, 0.2 Torr, so there was also another cause, which we concluded was as follows: Toner particles absorbed the light energy and were heated. When the internal temperature of the toner particle reached the decomposition temperature of the toner's base material, the toner particles underwent ablation and started to give off a vapor jet as shown in Fig. 6. They were then pushed by the jet. In short, we theorized that one cause of toner transfer was the ablation of toner. Accordingly, the internal temperature profile of the toner particle was important to analyze the toner transfer process.

Simulations

Temperature Simulation Method. As mentioned in the "Experiments" section, the temperature profile of the toner particle was the most important parameter for analyzing the toner transfer process. We solved the heat conduction equation to simulate the internal temperature of a toner particle. The heat conduction equation is expressed as,

$$\frac{\partial T}{\partial t} = D\Delta T + \frac{1}{\rho c_p} Q(r,t), \tag{1}$$

where D is the thermal diffusivity, ρ is the density, c_p is the specific heat, and $Q(r,t)$ is the heat generated at position r and time t. The heat is generated by light absorption, so the heat generation is in proportion to the light intensity in the particle and the absorption coefficient of the particle.[9] The exact solution of the internal and external field of a spherical particle irradiated by parallel light is given by as Mie theory.[10,11] The Mie equation is expressed using the complex refractive index of toner, the radius of the toner particle, and the wavelength of the light. The details of the heat conduction equation and the Mie equation are given in the Appendix.

We simulated the internal temperature of the toner particle in three steps: First, we sought the complex refractive index of the toner material. To this end we applied the effective medium approximation to estimate the mean complex refractive index of the mixture.[12] Second, we calculated the Mie equation to get the internal electric field of the toner particle using the estimated value of the complex refractive index (Eqs. A4 and A5). We assumed toner particles were spherical and light parallel. Third, we solved the heat conduction equation using the distribution of the internal electric field. We solved Eq. A1 with Eq. A2 by the implicit finite-difference scheme. The initial temperature was set to zero. The temperature at a distance of 140 μm from the particle surface, which was the thermal diffusion length of air 1 ms after the start of irradiation, was set to zero as the boundary condition. We divided the radius r and the angle θ in spherical coordinates each into 21 parts. The time interval for calculation was 1 μs.

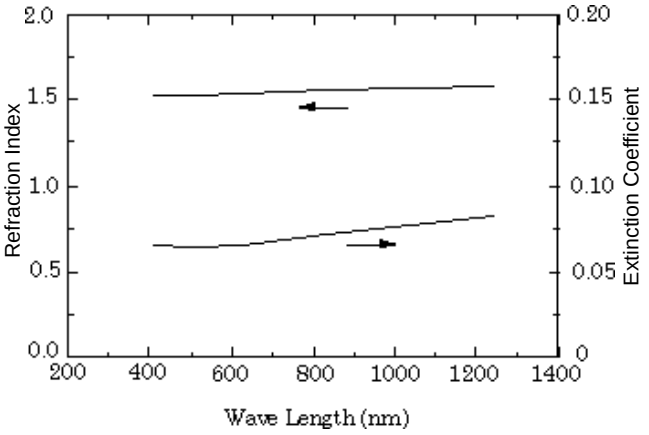


Figure 10. Calculation of complex refraction index using the mean medium approximation. Comparision with extinction coefficient, estimated from the reflectivity of fused toner measured using a spectrophotometer.

In the following sections, we explain the effective medium approximation and estimation of the complex refractive index of the mixture. After that, we simulate the internal temperature of the toner particle.

Complex Refractive Index of Toner. To calculate an internal electric field using the Mie equation, we needed the complex refractive index of toner, which was unknown. We estimated a value based on the toner composition. Black toner is a mixture of resin and submicrometer particles of carbon black. Carbon black particles are smaller than the main wavelength of the light of flash, so we can apply the effective medium approximation to estimate the mean complex refractive index of the mixture.[12] When medium A (dielectric constant: ε_A, volume ration: v_A) is mixed with medium B (dielectric constant: ε_B, volume ration: v_B), the average dielectric constant ε_m is expressed as,

$$v_A \frac{\varepsilon_A - \varepsilon_m}{\varepsilon_A + 2\varepsilon_m} + v_B \frac{\varepsilon_B - \varepsilon_m}{\varepsilon_B + 2\varepsilon_m} = 0. \tag{2}$$

Light incident on this medium experiences the average dielectric constant ε_m. The complex refractive index N is related to the dielectric constant ε as,

$$N = \varepsilon^{1/2}. \tag{3}$$

We needed to know the dielectric constants of the resin and the carbon black to calculate the complex refractivity of the toner particle by the effective medium approximation as expressed in Eq. 2. We assumed the resin's refractive index was 1.5 independent of the wavelength. This assumption is proper for the flash lamp of which the main wavelengths are in the visible. Using Eq. 3, we obtained 2.25 as the dielectric constant of the resin. As the dielectric constant of carbon black, that of "glassy" carbon[13] consisting of very small crystallites was used because carbon black was considered to be disordered graphite.

Figure 10 shows the complex refractive index as a function of wavelength of light calculated by using Eq. 2. The volume ratio of the carbon black was 10%. The refractive index and the extinction coefficient had little dependence on wavelength. To confirm the accuracy of this calculation,

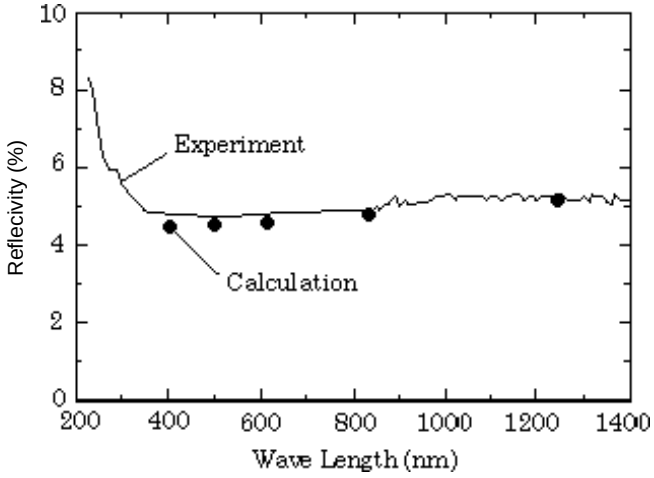


Figure 11. Comparison of calculated and experimental reflectivity.

we compared the measured reflectivity of toner with that estimated by using the calculated complex refractive index.

Figure 11 shows the measured and calculated reflectivities. The calculations had good agreement with the measurements except below 300 nm. Because the resin absorbs light below 300 nm, where the assumption that the complex refractive index was constant does not hold true. The main wavelengths of the xenon flash lamp are in the visible and infrared regions so the complex refractive index was not very dependent for these wavelengths. As a result, we used $1.55 - 0.07i$ as the complex refractive index of toner to calculate light absorption of toner.

Temperature Simulations at Constant Power Irradiation. First, the internal light intensity of the toner particle was calculated by using the Mie theory (Eqs. A4 and A5). Figure 12(a) shows the internal light intensity distribution when the toner particle's diameter was 10 μm and the dielectric constant was $1.55 - 0.07i$. The light was propagated in the z direction and the intensity was shown on the x-z (or y-z) plane. The intensity of incident light was set to 1. The penetration length of light into toner material is less than the toner diameter, so the light was strongly absorbed at the incident side of the particle. By using this intensity distribution, the heat conduction Eq. A1 was solved with Eq. A2 and the internal temperature distribution was calculated. The thermal and physical constants of polystyrene were used for those of toner particle: $c_p = 1.3 \times 10^3$ J/kg • K, $\rho = 1.2$ kg/m^3, $D = 7.0 \times 10^{-8}$ m^2/s.

The light intensity was set constant, 1.5×10^3 W/cm^2. Figures 12(b) and 12(c) show internal temperature distributions at two different times. The heat generated on the light incident side was conducted to the inside of the toner particle as time passed. The position of the maximum temperature was slightly inside the toner particle because of the heat conduction to the air from the particle surface. The temperature increased to more than 600°C at 0.5 ms in the calculation. We confirmed that

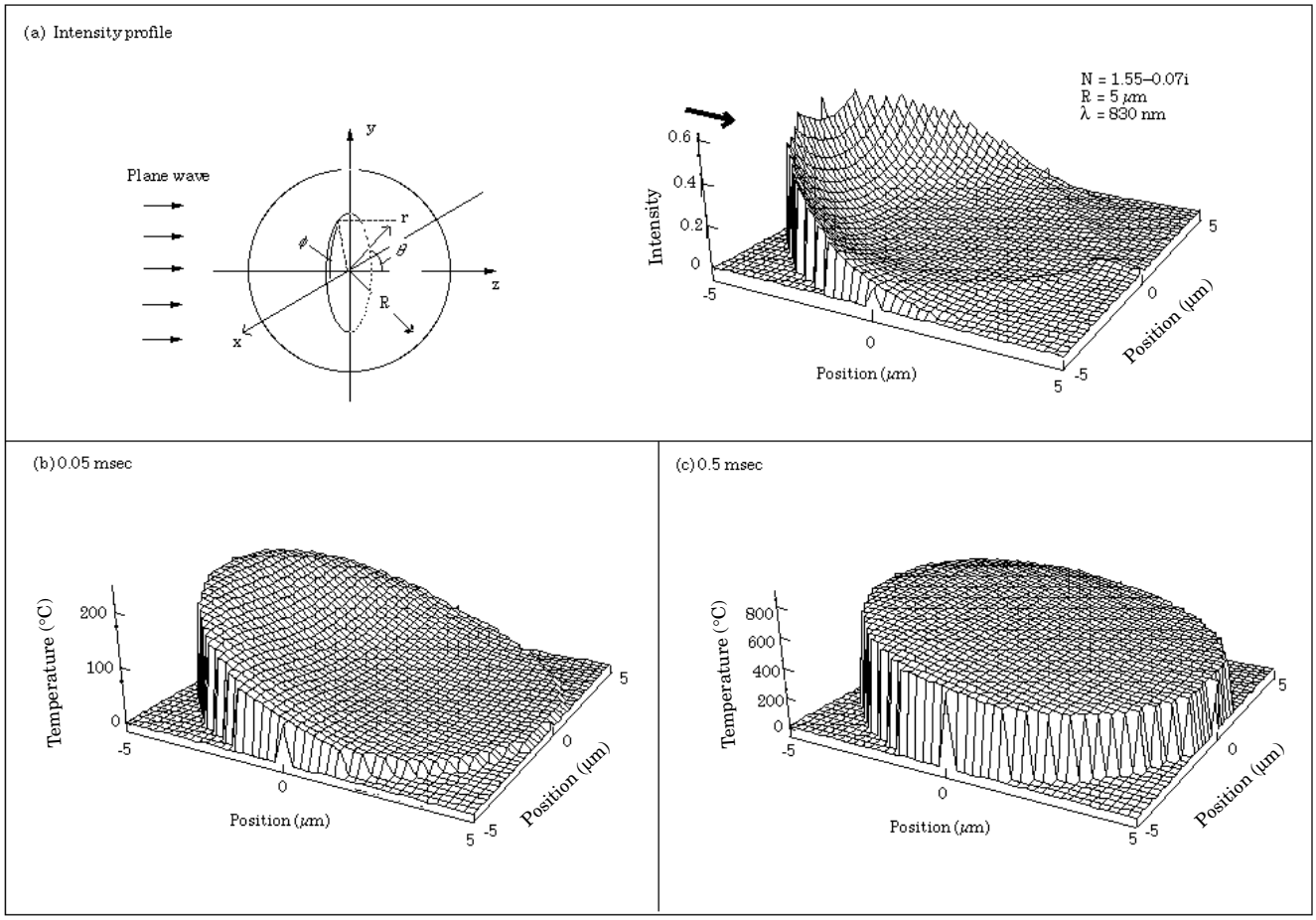


Figure 12. Simulation of temperature profiles of toner irradiated by pulse light.

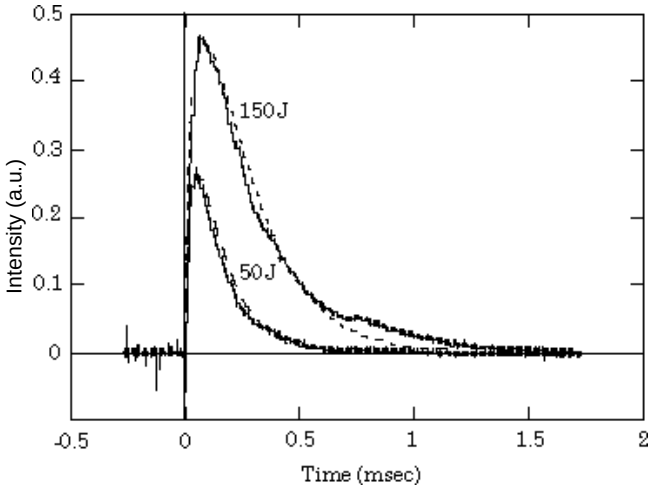


Figure 13. Profile of the intensity of the flash lamp.

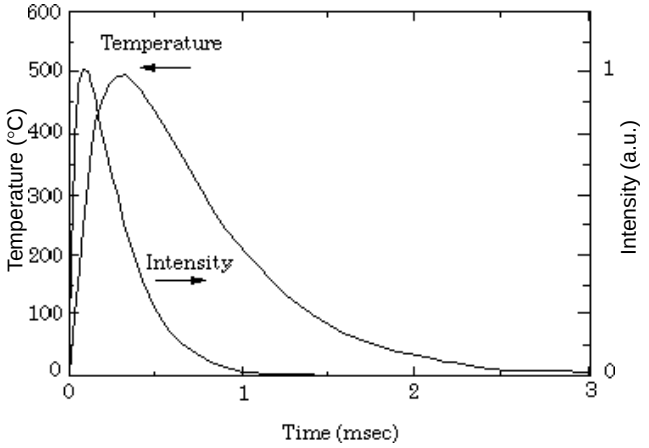


Figure 14. Simulation of the time-dependent internal temperature and intensity.

the maximum temperature when the particle was irradiated by various powers was proportional to the power density at the same irradiation time. Using this confirmation, the time when the toner maximum temperature reached the particular constant temperature as a function of the power density was determined.

If the toner particle was moved by ablation, its transfer would start when the maximum temperature reached the particular temperature at which ablation commenced. We sought to compare the experiment on the toner departure interval to the time when the toner was heated to a certain temperature; i.e., the decomposition temperature. The toner material starts to decompose at about 300°C, so we calculated the time when the maximum temperature of the toner particle reached 300°C.

We fit this temperature simulation result to the data of the transmissivity experiment by using the beam size as a fitting parameter. The solid line in Fig. 8 shows the power dependence of the time when the internal maximum temperature reached 300°C, setting the diameter of the beam as 51 μm. The circles were the data of the transmissivity experiment. The simulation curve fitted the experiment points well, which meant that toner particles start to transfer when they are heated to a specific temperature; probably the decomposition temperature of toner. It was thus confirmed that the toner transfer was caused by the ablation.

Using the same flash lamp, Mitsuya et al.[14] measured the time-dependence profile of the surface temperature of the toner layer when directly irradiated from the air side and showed that the peak surface temperature of toner irradiated by the light from the flash lamp supplying 440 J of electric power was more than 300°C.

Temperature Simulation at Flash Irradiation. To calculate the temperature under conditions of flash irradiation, we tried to estimate the intensity profile of the light flash by assuming the time profile of flash lamp radiation as,

$$I(t) = at^n \exp(-bt). \tag{4}$$

We determined the parameters n, a, and b from fitting to measurements. Figure 13 shows the time dependence of the intensity of the short arc flash lamp (the solid line) and the fitting result when n was 0.5 (the dashed line). Data for intensities electrical energy supplied to the flash lamp of 50 and 150 J are shown. The form of the intensity fit well with n = 0.5. By integrating Eq. 4 from time 0 to infinite time, the irradiation energy E in infinite time is

$$E = a\frac{\Gamma(n)}{b^{n+1}}, \tag{5}$$

where $\Gamma(n)$ is the Gamma function. The coefficient b is in proportion to the inverse of the light pulse width. We confirmed that the E calculated from Eq. 5 using the fitting values of a and b with n = 0.5 was proportional to the input electric energy of the flash lamp.

Figure 14 shows the simulation results of the time dependence of the light intensity and the maximum temperature of the inside of the toner. The light intensity was normalized by setting the maximum intensity to 1. The temperature peak came later than the light intensity peak. The time at which the maximum temperature of the inside of the toner was more than 300°C was delayed from the irradiation start. The toner transfer region in which temperature of the toner was more than 300°C from 0.2 to 0.8 ms in Fig. 14 almost agreed with that of the current measurements (Fig. 9).

Discussion

In the temperature simulations of the particle, we checked that the maximum temperature inside was proportional to the irradiated light energy at the same irradiation time. To increase the toner temperature above that of the toner decomposition when irradiated by a pulsed light of a constant length, there was a minimum irradiation energy. The threshold of energy density at which toner can be transferred, as shown in Fig. 4, thus represented the lowest irradiation energy that exceeds the decomposition temperature. The rapid increase of transfer time at low irradiation power in Fig. 8 also corresponded to the threshold of the irradiation energy in Fig. 4.

In Fig. 5 the rapid drop of the transferred amount at long irradiation time was shown. This phenomenon could also be explained in relation to the toner temperature. Figure 15 shows the inside maximum temperature of the toner particle versus the half value of the irradiation time for the flash of light or a pulse with the same irradiation energies. As the irradiation time increased, the maximum temperature decreased owing to the increase of heat conduction to air. In this calculation, the maximum temperature dropped

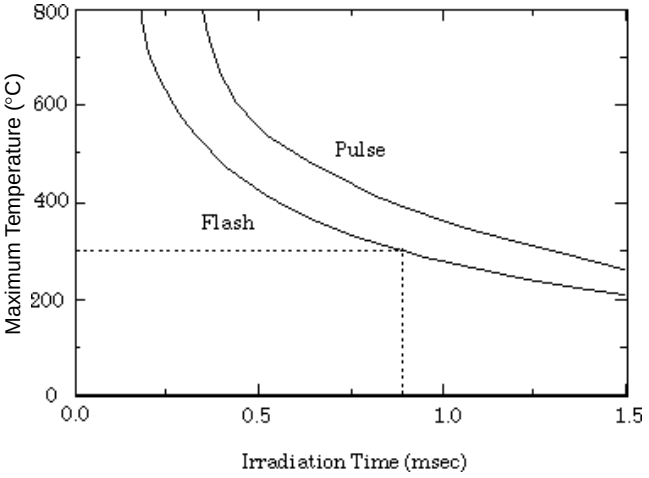


Figure 15. Simulation of the inside maximum temperature as a function of the light irradiation time.

below 300°C after 0.8 ms, under conditions of flash irradiation and the toner particles, consequently, would not be transferred. We considered the rapid drop of the transferred amount in Fig. 5 to correspond to this threshold of irradiation time.

From the above study, we identified desirable conditions for this printing method. The irradiated light pulse should be short because the energy loss caused by heat conduction would be lowered as shown in Fig. 15. The pulse form is also important. Figure 15 shows the maximum inside temperature of the toner particle versus the irradiation time of a rectangular-shaped pulse with the same irradiation energy as the flash of light. The rectangular-shaped pulse needed less energy than the flash of lamp to heat to the same temperature. In regard to the toner material to the decomposition temperature, the decomposition temperature should be low because less energy would be needed to heat the toner material.

As shown in Fig. 12, the position where the inside temperature of the toner particle registered a maximum was a little bit inside the surface of the toner particle. As a result, the portion between the position of the maximum temperature and the light incident surface was blown off by ablation. Toner particles were repelled by the mass blown off, so the driving force of toner transfer became large;[6] i.e., on the order of 10^{-9} *N*.

It has been well-established that ablation occurs by a thermal or chemical process. Laser ablation transfer of an ink ribbon caused by irradiation with a YAG laser beam has been studied.[5,6] When irradiated by a laser beam, the ink transfer was seen to be a thermal process. The study used a 150 ns (FWHM) pulse, but ablation occurred at around 200 ns from the start of irradiation. In contrast with this work, we used a much longer pulse of the flash lamp, typically an 0.5 ms pulse. We considered that one reason for the difference in observations was the form of the material; particles versus ribbon. In comparsion irradiation of the flat face, the spherical particle reaches a higher temperature at the same radiant energy.

Toner particles exposed to the high-power light and not transferred were fused on the plastic sheet. The fused toner particles could be scraped off. In the case where a heat-resistant belt or drum is used to carry the toner layer, the drum or belt can be regenerated cyclically.

Concerning the image quality of this process, the transferred amount was small as shown in Fig. 3. And the resolution of images formed by irradiation was only about 180 dpi, perhaps because charged particles repel each other. However, these charge properties can be utilized for improvement of the image quality. We supplied a voltage between the transparent electrode and the rear electrode to attract charged toner particles to the paper in Fig. 1. By applying an electric field to the charged toner particles, the amount transferred was increased and the image resolution was improved.[8]

Conclusion

We analyzed a printing method based on direct toner transfer by a flash of light. This method considerably simplifies the imaging process. The transferred amount of toner particles increased linearly with the irradiated energy and was about 15% of the initial sticking amount at the irradiation light energy density of 0.73 J/cm^2. The amount abruptly decreased at long pulse lengths of more than 0.7 ms. Next, we observed toner movement using a high-speed camera and measured the transmissivity and current transition to clarify the toner transfer process. In the high-speed observation, we confirmed that a jet of smokelike matter was given off from the toner layer because of the laser pulse irradiation. In the transmissivity and current measurements, we found a time interval existed in which the toner transfer started, and the interval became shorter as the light power increased. We concluded from these experiments that the toner transfer was caused by a thermal event and the process was ablation of the toner. The hypothesis was checked by temperature simulations. We solved the heat conduction equation using the Mie theory. For the simulation, the complex refractivity of toner material was estimated using the mean effective approximation. Comparison with experimental and simulation values showed the toner transfer started when the inside maximum temperature reached a specific temperature, which was the decomposition temperature of the toner material. The abrupt decrease of the transferred amount as the pulse length was increased was explained well by the temperature simulations.

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Appendix

The heat conduction Eq. 1 for a spherical particle in uniform parallel light is expressed in spherical coordinates as,

$$\frac{\partial T}{\partial t} = D\left[\frac{1\partial}{r^2\partial r}\left(r^2\frac{\partial T}{\partial r}\right)+\frac{1\partial}{r^2\sin\theta\partial\theta}\left(\sin\theta\frac{\partial T}{\partial\theta}\right)\right]+ +\frac{1}{\rho c_p}Q(r,\theta,t). \quad \text{(A1)}$$

The heat is generated by light absorption, so the heat generation is in proportion to the light intensity in the particle and the absorption coefficient of the particle. The heat generation Q is related to the normalized intensity S as [8]

$$Q(r,t) = \frac{4\pi n\kappa I_0}{\lambda}|S(r,t)|, \quad \text{(A2)}$$

where $n - i\kappa = N$ is the complex refractive index of the particle, n is the refractive index, and κ is the extinction coefficient of the particle; λ is wavelength of the incident light; and I_0 is the incident light intensity. The normalized intensity $S(r,t)$ is expressed as

$$S(r,t) = |E(r,t)/E_0|^2, \tag{A3}$$

where $E(r,t)$ is the internal electric field of the particle and E_0 is the electric field of the incident light.

For Eqs. A2 and A3 to be able to calculate the heat generation Q in the toner particle, the internal electric field $E(r,t)$ in the particle is needed. The exact solution of the internal and external field of a spherical particle irradiated by parallel light is given by Mie theory.[9–11] The internal electric field of the spherical particle is expressed in spherical coordinates (r,θ,ϕ) as,

$$\begin{aligned}
E_r &= \sum_{m=1}^{\infty}\left[-i^{m+1}(2m+1)\frac{1}{Nkr}\,j_m(Nkr)\cos\phi P_m^1(\cos\theta)d_m\right],\\
E_\theta &= \sum_{m=1}^{\infty} i^m\cos\phi\left[\frac{2m+1}{m(m+1)}\,j_m(Nkr)\frac{P_m^1(\cos\theta)}{\sin\theta}c_m - i\cos\phi A_m B_m d_m\right],\\
E_\phi &= \sum_{m=1}^{\infty} i^m\sin\phi\left[\frac{2m+1}{m(m+1)}\,j_m(Nkr)B_m c_m + iA_m\frac{P_m^1(\cos\theta)}{\sin\theta}d_m\right],
\end{aligned} \tag{A4}$$

where the coefficients A_m, B_m, c_m, and d_m are expressed as

$$\begin{aligned}
A_m &= j_{m-1}(Nkr) - \frac{j_m(Nkr)}{Nkr},\\
B_m &= m\cos\theta\frac{P_m^1(\cos\theta)}{\sin\theta} - (m+1)\frac{P_{m-1}^1(\cos\theta)}{\sin\theta},\\
c_m &= \frac{i}{x^2\left[j_m(Nx)h_{m-1}^1(x) - Nj_{m-1}(Nx)h_m^1(x)\right]}\\
d_m &= \frac{Ni/x}{x\left[N^2 j_m(Nx)h_{m-1}^1(x) - Nj_{m-1}(Nx)h_m^1(x)\right] - m(N^2-1)j_m(Nx)h_m^1(x)},
\end{aligned} \tag{A5}$$

and N is the complex index of refraction of the particle, k is the wave number of the incident light, and $i = \sqrt{-1}$. The parameter x is called the size parameter, which is related to the radius of the particle R and the wavelength λ, as $x = \pi R/\lambda$. The functions $j_m(x)$, $h_m^1(x)$, and $P_m^1(\cos\theta)$ express the spherical Bessel function of the first kind, the spherical Hankel function of the first kind, and the associated Legendre function, respectively. The light flash is not polarized, so we average the electric field as the coordinate ϕ.

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