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# Development of an Image Eraser for Decolorizable Toner

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**Abstract.** *We have developed an image eraser for reuse of paper printed on with decolorizable toner, which is composed of leuco dye and developer. The decolorizable toner loses color when heated. The image eraser enables decolorization of a bundle of paper instead of one sheet at a time. Before designing the image eraser, we studied three problems caused by heating, ghost images, sticking, and color change of paper. We have optimized the heating conditions of the image eraser, i.e., the heating temperature is 130–140 °C, and the heating period is 2 h. © 2006 Society for Imaging Science and Technology.*
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## BACKGROUND

The total paper recycling rate is approximately 60.4% and 40% of trash by volume is said to be paper in Japan,[1] but the rate of recycling of printing paper in offices is less than 30% (Ref. 2) because of shredding for secrecy. The shredded paper is sent to an incinerator where it generates carbon dioxide. In addition, paper reuse has not yet become widespread, except for the use of the reverse side of a sheet of paper, although paper reuse[3] is effective in reducing the amount of paper trash.

In order to reduce the amount of paper trash, several paper reuse systems have been proposed, such as erasable toner which uses far infrared radiation,[4] toner stripping machines,[5] and rewritable marking media.[6] However, a practical paper reuse system has not been widely adopted in actual use. We have already reported erasable ink as a solution to the problem of paper recycling and reuse.[7–10] This ink is originally colored and becomes colorless as a result of heating or the application of an organic solvent such as acetone.

As an application of the erasable ink, we have developed a decolorizable toner for paper reuse in offices. Using the decolorizable toner, the used paper can be easily decolorized. It is possible to reprint and rewrite on the decolorized paper repeatedly. For practical office use, we have selected heat erasing, because it is difficult to use an organic solvent in offices. Hence, an efficient heat eraser is required for the best solution.

Obviously, reuse of something is the most effective way to save transporting and recycling energy. To confirm this concept, we have applied life cycle assessment to a paper reuse system using decolorable toner and heat eraser according to ISO14040 (International Standard). The details of the boundary region of estimation and the life cycle scenario of the paper are cited in the previous paper.[11,12] The definition of the boundary region is the office before fiber recycling. Carbon dioxide emission decreased by 31% when using decolorizable toner with single-side decolorizing, compared with double-side copying. A more moderate reduction of 26% was seen in energy consumption. When the decolorizing process was conducted five times, the reductions were 51% and 39%, for carbon dioxide emission and energy consumption, respectively. Considering the transportation of paper and ease of the deinking, a greater reduction of carbon dioxide is expected for the decolorizable toner system, because the deinking process is a major part of a paper recycling plant and is the system which consumes the most energy.

### *Principles of Erasable Ink and Decolorizable Toner*

Although the erasable ink can be erased completely on a sheet of paper by heating, the decolorizable toner can be read from a certain angle because of remaining resin, which is a main component of the toner. The word "decolorizable" means just becoming colorless by heating. However, the principle of decoloration is as follows. Figure 1 shows the coloring and decolorizing principle of the decolorizable toner. The decolorizable toner is composed of leuco dye colored by developer. The leuco dye, a fluoran compound as shown in Fig. 1, is originally colorless, and conventionally used in thermal paper. The developer is a phenolic compound such as propyl gallate, which is known as a food additive. In colored toner, the lactone ring of the leuco dye is opened and the central double bond is conjugated with two

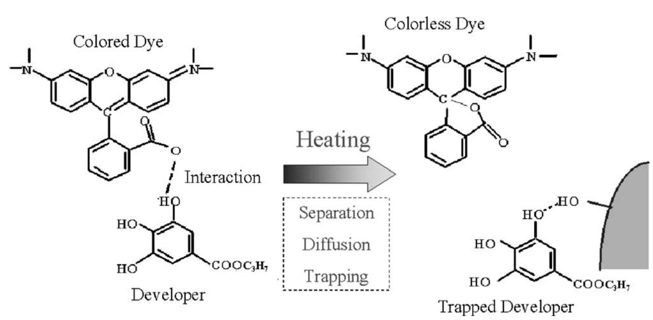


Figure 1. Principle of erasable ink.

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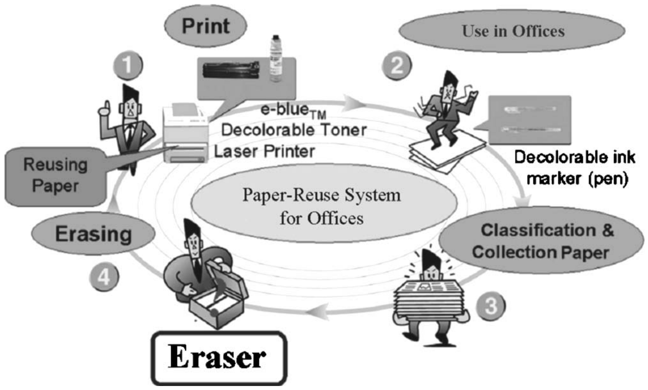


Figure 2. Outline of paper reuse system in offices.

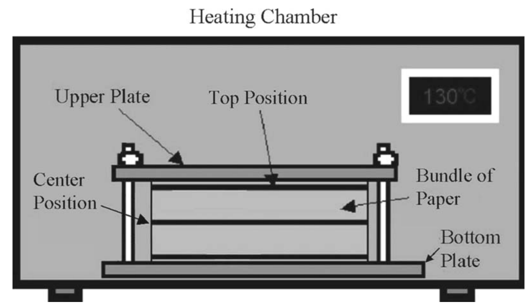


Figure 3. Outline of heating device for a bundle of paper.

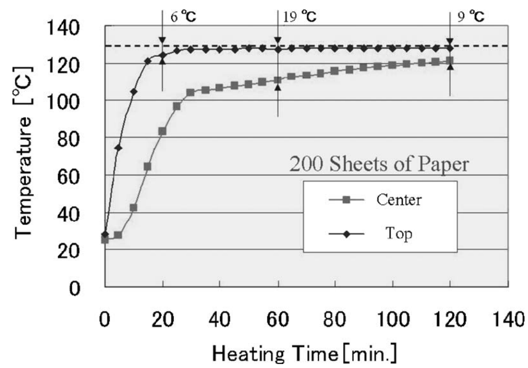


Figure 4. Relationship between heating time and paper temperature.

benzene rings. In the decolorizing process, suitable heating breaks the interaction between the dye and developer. Dissociation of the developer closes the lactone ring to its original state, and the dye becomes colorless. The developer diffuses in the binding polymer and is irreversibly trapped by an absorbing material, such as cholic acid and starch.

### *Properties of Decolorizable Toner*

The decolorizable toner is produced to comply with the specifications for normal toner for laser beam printers of 35 prints per minutes. The fusing temperature is 190 °C at the surface of the fuser. At this temperature and speed, the decolorizable toner maintains its color density. The decoloration process of the leuco dye takes a longer time than the fusing process of the toner binder. The optical density of the decolorizable toner powder was 1.10 when the average size of toner particles was 11 $\mu$m. We have regulated the printed toner to have a specified maximum optical density of 0.7. A rubbing tester designed for ordinary toner evaluated rub resistance, resistance to abrasion, and adherence to paper. After repeated rubbing, 70% of the toner remained on a sheet of paper. The results showed that the decolorizable toner had almost the same rub resistance, as ordinary toner. The degradation of the decolorizable toner in storage was evaluated by the fluidity of the toner after being kept in a bottle at 50 °C for 8 h. We regulated the fluidity to be the same before and after testing by using additives. There was no decline in optical density of printed toner stored in a dark place for 9 years. Because glass transition temperature ($T_g$) of the decolorable toner is 62.5 °C, the toner is considered to be stable below the temperature of $T_g$.

The light fastness was measured by the optical density of the printed toner after 15 000 lux light exposure for 10 h, which decreased to 95.4% of the initial value. The leuco dye is known to have low resistance to light.

### *Eraser for Paper Reuse System*

As an application of the erasable ink, in 2003 Toshiba commercialized "e-blue$^{TM}$",[10] a decolorizable blue toner, for electrophotographic printers. Figure 2 shows an outline of the paper reuse system. After collecting used paper, the image on the paper is decolorized using the heat image eraser. With this system, the paper can be reused several times. Moreover, with the reuse system, seriously damaged paper can be sent to the conventional paper recycling system.

In regard to the design of the heat eraser, the requirements are: (1) maximum safety, (2) minimum energy, (3) minimum time, (4) simplicity, and (5) minimum cost.

We chose a batch treatment method and a structure with two heat plates for the heat eraser.[10]

## EXPERIMENTAL PROCEDURE

### *Test Chart*

Test charts were printed with the decolorizable toner using a Premage 350 duplicator (Toshiba Corp.). The test chart consisted of solid squares of 1 $cm^2$ to measure the original and decolorized optical density. Optical density was measured with a CR300 (Minolta Corp.) densitometer in reflection mode.

### *Thickness of Paper Bundle*

200 sheets were chosen for the bundle thickness, with reference to the previous experiments. For a thicker bundle, the suitable heating time became longer due to the temperature differences in the bundle.

### *Temperature Distribution in the Paper Bundle*

There was a temperature distribution in the paper bundle in the direction of bundle thickness, because of the low thermal conductivity of paper and the air layer generated between paper sheets. Thus, the temperatures at several parts of the bundle of paper were measured, and the changes monitored for a maximum of 10 h. We conducted experiments using

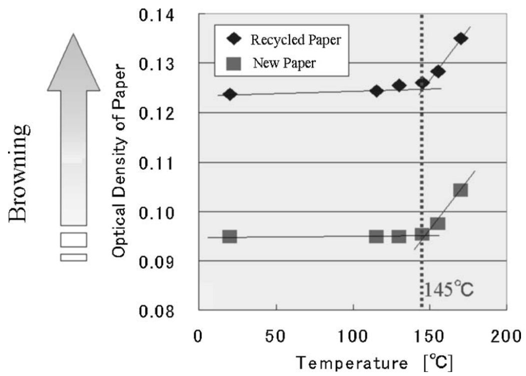


Figure 5. Relationship between heating temperature and optical density of plain paper.

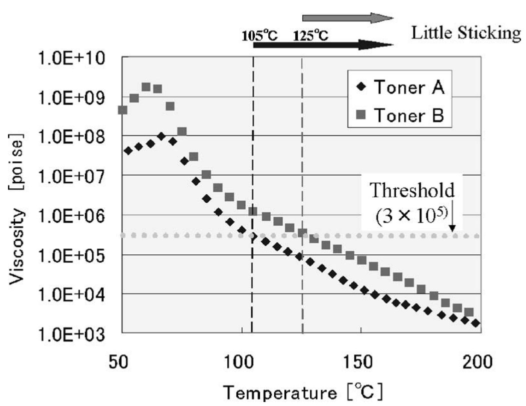


Figure 6. Viscosity of decolorizable toner.

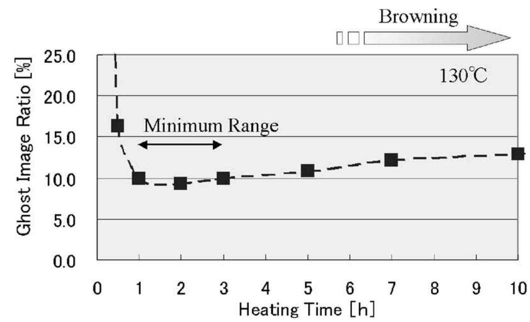


Figure 7. Relationship between heating time and ghost image ratio.

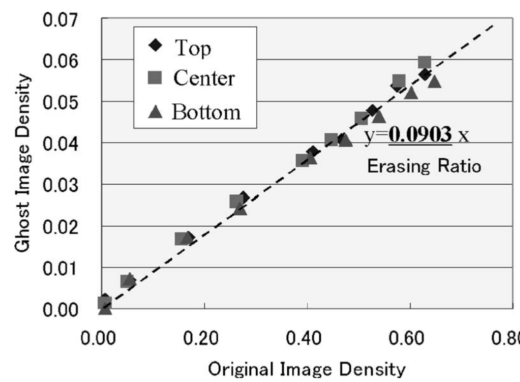


Figure 8. Evaluation of erasing ratio.

the heating chamber and a pressing device as shown in Fig. 3, consisting of two aluminum plates (1.0 cm thick) serving as a heat sink, and four nuts and bolts. An oven with a power rating of 1.3 kW was used for the heating chamber. To avoid the presence of air layers in the paper bundle, the pressing device was used to apply controlled pressure to the paper. After the temperature of the oven and pressing device were stabilized, a bundle of paper was inserted between the aluminum plates and pressed. The pressure was regulated to about 500 Pa.

Based on previous erasing experiments, we have chosen 130 °C as the experimental condition. Figure 4 shows the relationship between heating time and paper temperature in the bundle. After heating for 2 h, the temperature at the center position of the paper bundle was 9 °C lower than temperature at the top position.

### *Problems of Heat Decolorizing Process*

We have identified three problems concerning the heat erasing: (1) color change of paper, white to brown; (2) sticking of paper sheets; and (3) ghost image, a visible trace of the original image after heating. The ghost image is pale yellow or pale blue.

### *Color Change of Paper*

We conducted heating experiments for two kinds of plain paper: new paper made from virgin pulp and recycled paper made from used newspapers. Figure 5 shows the relationship between heating temperature and optical density of the paper after heating for 2 h. We found the color change started at a temperature of higher than 145 °C for both kinds of paper. Therefore, we have determined the maximum heating temperature for the eraser to be 145 °C.

### *Sticking of Paper Sheets*

Paper sticking depends on the heating temperature and the kind of toner and paper. Using one pair of printed sheets of paper, we examined paper sticking with two kinds of toner: a 165 °C fixing toner (toner A) and a 190 °C fixing toner (toner B). There was no sticking of paper above 105 °C for the low temperature fixing toner (toner A) and above 125 °C for the high temperature fixing toner (toner B). In both cases, serious sticking of paper was not found above the threshold temperature. Figure 6 shows the temperature dependence on the toner viscosity. We found that the toner viscosity at the threshold temperature was the same in both cases, about $3.0\times10^5$ poise as shown in Fig. 6. These results suggest that paper sticking depended on the toner viscosity at the heat erasing temperature.

We also identified the following reason for the occurrence of sticking at above the threshold temperature. The toner included a considerable amount of polypropylene wax as an additive. The wax bled out from inside the toner to the surface of the toner by thermal diffusion above the softening temperature of the toner binder. After cooling, the surface bleeding wax easily broke the connection between the paper

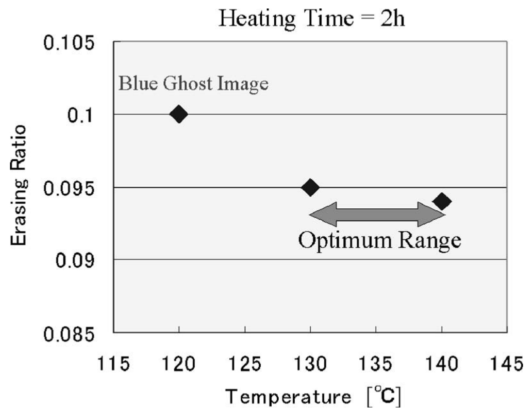


Figure 9. Relationship between heating temperature and erasing ratio.

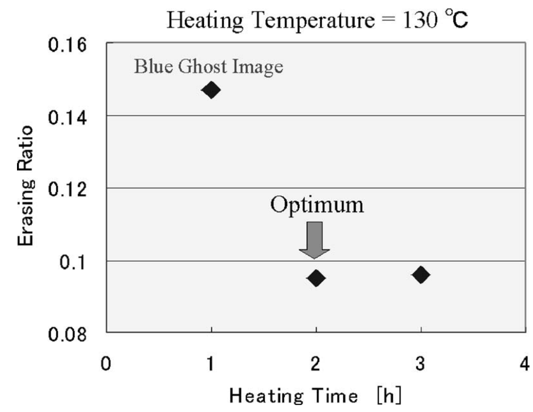


Figure 10. Relationship between heating time and erasing ratio.

surface and toner binder. Hence, the threshold temperature for the paper sticking was the toner softening temperature.

The threshold temperature was 105 °C for the 165 °C fixing toner. Because the paper temperature at the center position in the bundle was about 10 °C lower than that at the top position, we have determined the minimum heating temperature for the eraser without the paper sticking was 115 °C.

### *Ghost Image After Heat Erasing*

The ghost image after heat erasing is the most serious and complicated phenomenon concerning the decolorizable toner. A ghost image is an image that remains after heat erasing.

Figure 7 shows the relationship between the heating time and ghost image ratio in the single-sheet experiment.

The heating temperature was fixed at 130 °C. The ghost image ratio is defined as the ratio of the ghost image density to the original image density. It decreased rapidly with heating time and reached a minimum value in about 2 h. The density increased slowly with time. A change in the color suggested that another color was generated with the passage of time, suggesting decomposition of the leuco dye generated a colored species over a longer heating time. From this experiment, we have confirmed that a suitable heating time is 1–3 h.

In the following erasing experiment, a bundle of paper was placed in the heating chamber. We used the heating chamber and the pressing device shown in Fig. 3. After the temperature of the oven and pressing device was stabilized, a bundle of 200 sheets of paper was inserted between the metal plates and pressed. The pressure was about 500 Pa.

Figure 8 shows the relationship between the optical density of the original image and the ghost image after heat erasing. Three test charts were inserted into the top, center, and bottom of the bundle of paper to obtain data for the three regions. These data were used to obtain a regression line, where we have defined the erasing ratio as the gradient of the regression line in Fig. 8. This erasing ratio represents the decolorizing ability of the toner, using the batch heat eraser. We conducted a series of erasing experiments with different heating temperatures and time. Figure 9 shows the relationship between the heating temperature and the erasing ratio. Based on the erasing ratio, we determined the optimum heating temperature range to be from 130 to 140 °C. Figure 10 shows the relationship between the heating time and the erasing ratio. The heating time should be as short as possible, and we have determined that the optimum heating time is 2 h. The erasing ratio became 10% or less under suitable conditions as shown in Fig. 7. An erasing ratio of 10% corresponds to an optical density of 0.06. When we use the reverse side of printed paper, we can see an image through the paper from the reverse side. This image density can be measured as about 0.06. The results of a questionnaire from 600 people indicated that even a small trace is not acceptable; however, they used the reverse side of printed paper piled in the office for environmental reasons. Therefore, we can use the erased paper in practice because the ghost image density is about the same as that perceived for the reverse side image of printed paper.

## CONCLUSION

We have developed an image eraser for the decolorizable toner, e-blue™, for use in a paper reuse system in offices. The image eraser was a batch heater with two heat plates to decolorize a bundle of paper at once. We found three problems concerning the heat erasing, namely, color change of paper, sticking of paper, and ghost images. We determined suitable conditions for the image eraser: a bundle of paper consisting of 200 sheets of paper, a heating temperature from 130 to 140 °C, and a heating time of about 2 h. Under the optimum conditions, we succeeded in decreasing the ghost image ratio to less than 10%, which is the same level as an image seen through a paper sheet from the reverse side of printed paper.

## REFERENCES

[1] http://www.jpa.gr.jp/en/about/bus/index.html.
[2] Research report about utilization of used paper and environmental impact, National project report of the Paper Recycling Promotion Center in Japan (2001).
[3] K. Isago, *Proceedings of Pan-Pacific Imaging Conference/Japan Hardcopy '98* (The Imaging Society of Japan, Tokyo, Japan, 1998) pp. 281–284.

[4] K. Murohoshi, and Y. Hosada, U.S. Patent No. 5,846.682.
[5] H. Kitazawa, S. Kimura, E. Kawamura, and T. Kawanishi, U.S. Patent No. 5,738,759.
[6] K. Tsutsui, T. Yamaguchi, and K. Sato, Jpn. J. Appl. Phys., Part 1 **33**, 5925–5928 (1994).
[7] S. Takayama, S. Machida, N. Ikeda, and K. Sano, *Proc. IS&T's NIP15* (IS&T, Springfield, VA, 1999) pp. 323–326.
[8] S. Machida, S. Takayama, N. Ikeda, T. Urano, and K. Sano, *218th ACS National Meeting Division of Environmental Chemistry*, No. 39 (ACS, Washington DC, 1999) pp. 220–222.
[9] K. Sano, *Proc. IS&T's NIP17* (IS&T, Springfield, VA, 2001) pp. 24–27.
[10] K. Sano, S. Takayama, S. Machida, and N. Ikeda, Proc. ICIS '02 (The Imaging Society of Japan, Tokyo, Japan, 2002) pp. 622–623.
[11] S. Takayama, N. Ikeda, T. Gotanda, and K. Sano, *2005 Beijing International Conference on Imaging, Preprint of Extended Abstracts* (CSIST, Beijing, China, 2005) pp. 14–15.
[12] I. Tanaka, K. Sano, K. Minakami, N. Takeyama, H. Kagami, and K. Haruki, *Proceedings of 5th International Conference on EcoBalance, Tokyo S1-79* (The Society of Non-Traditional Technology, Tokyo, Japan, 2002) pp. 243–246.