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## Effect of Electron Beam Irradiation on Forensic Evidence. 2. Analysis of Writing Inks on Porous Surfaces

**ABSTRACT:** The effect of electron beam irradiation on a series of different writing inks is described. As the anthrax-tainted letters were discovered in October 2001, the U.S. government began to experiment with the use of the electron beam irradiation process for destroying such biological agents. Plans initially considered a large-scale countrywide use of this technology. However, over time the scope of this plan as well as the radiation dosage were reduced, especially when some adverse consequences to mailed items subjected to this process were observed. Little data existed at the time to characterize what level of damage might be expected to occur with common items sent through the mail. This was especially important to museums and other institutions that routinely ship valuable and historic items through the mail. Although the Smithsonian Institution initiated some studies of the effect of electron beam irradiation on archived materials, little data existed on the effect that this process would have on forensic evidence. Approximately 97 different black, blue, red, green, and yellow writing inks were selected. Writing ink types included ballpoint, gel, plastic/felt tip, and rollerball. All noncontrol samples were subjected to standard mail irradiation conditions used by the U.S. Postal Service at the time this experiment was performed. A video spectral comparator and thin-layer chromatography (TLC) analysis were used to evaluate both the control and the irradiated samples. Some published studies reported changes in the presence/absence of dye bands in the chromatograms of irradiated writing inks. Some of these studies report the formation of additional dye bands on the chromatogram while others report missing dye bands. However, using standard testing guidelines and procedures, none of the 97 irradiated inks tested were found to show any significant optical or chemical differences from the control samples. In addition, random testing of some of the ink samples using a second solvent system did not reveal any changes. However, one control ink did show some minor changes in optical properties and dye characteristics over time (but not TLC) while the irradiated sample remained stable. Significant changes in the ultraviolet fluorescence characteristics of the irradiated paper samples themselves (not inks) were also observed.

**KEYWORDS:** forensic science, writing inks, inks, ink analysis, thin-layer chromatography, mail irradiation, electron beam irradiation

The routine irradiation of mail is a relatively recent development and has a limited scope; therefore, little data exist on the effects of such treatments on forensic evidence. In a previous paper, the authors investigated the effects of such conditions on the ability to visualize latent prints on porous and nonporous surfaces (1). Significant degradation of latent print residue was found to occur on both surface types. As the U.S. Secret Service routinely performs chemical analyses of writing ink samples on questioned documents, a decision was made to investigate what effects the irradiation process would have on such examinations.

Several relatively recent studies have investigated the impact of irradiation treatments on archival materials (2) and writing inks (3,4). These studies were conducted primarily by the Smithsonian Center for Materials Research and Education (Suitland, MD). In one study, seven of 26 inks tested showed changes in the TLC chromatograms (3). The inks affected were red and blue ballpoint. With six of the seven inks, a new dye band was detected after chromatographic analysis. With the seventh ink, the relative mobilities (i.e., retardation factors) of the dye components were altered. Radiation dosages in this study were estimated to be up to five times the dosage encountered in the routine mail irradiation

process. The second study found additional ink changes, including altered mobilities and new dye components even at the lower radiation dosages associated with mail irradiation (4). It should be noted that these studies used different extraction solvents (acetone and ethyl acetate) and solvent systems for eluting TLC plates. The authors examined 97 different writing inks of various colors (132 samples total, including 35 aged samples) before and after irradiation and compared their thin-layer chromatograms.

The U.S. Secret Service maintains an International Ink Library that contains samples of more than 9000 inks from around the world, some dating back to the 1920s (5). Although the library contains primarily writing ink samples, it also contains printing, marking, typewriter ribbon, and stamp pad inks. The origin of this ink library dates back to the early 1960s, when Werner Hofmann of the Zurich Cantonal Police in Switzerland established a modest collection of black and blue European ballpoint inks (L. Olson, personal communication, 2005). This collection was eventually incorporated into what would become the International Ink Library. Additional ink samples were contributed by the Federal Bureau of Investigation, United States Postal Service, and several private document examiners. In 1968, Richard Brunelle, a chemist with the Internal Revenue Service's (IRS) Alcohol and Tobacco Tax Division Laboratory (which later separated from the IRS and became the Bureau of Alcohol, Tobacco, and Firearms [BATF]), began to contact domestic ink producers and significantly expanded the library's contents. In the first year of the effort, more than 2000 samples were collected.

In an effort to relieve the BATF ink analysis case load (which was largely due to submissions from the IRS), the IRS laboratory and BATF began to share the resources of the ink library in June

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of 1987. This mutual agreement also involved a decision by the BATF to train an ink chemist for the IRS laboratory as well. Initially, the IRS was interested in only black, blue, red, and green ballpoint ink samples from the BATF ink library. Subsequently, the IRS collected over 1500 open market (i.e., commercial off-the-shelf) ink samples and contributed them to the BATF library. To date, the IRS laboratory has contributed a total of approximately 2300 open market samples to the library. In 1988, the ink library was ultimately transferred from the BATF to the U.S. Secret Service. These two libraries are now constantly being updated with samples submitted from the major ink manufacturers (primarily within the United States, Europe, and Japan) as well as with open-market purchases from domestic and foreign sources.

## Materials and Methods

A total of 97 different inks were tested and evaluated during this study, of which 27 were black, 33 blue, 18 red, 12 green, and seven yellow specimens. Appendices A–E contain lists of all of the inks that were chosen for this study. A series of sample sheets were prepared by scribbling each ink within two similar rectangular boxes. The boxes were numbered and then separated, one to act as a control and the other to be irradiated. Thirty-five additional ink samples were included as well. These samples were duplicates of some of the original 97 that had been deposited on paper (typically Whatman filter paper) at various times within the past 26 years. These samples were included to determine whether there would be any differences with samples that had been allowed to dry on paper for a number of years. Two types of paper were used: Xerox Premium Multipurpose 4024 Paper (8.5" × 11", 20lb, 88 brightness) and white, blue-lined notepad paper (8.5" × 11", 16lb, 50% recycled content, purchased from GSA (Washington, DC), NSN 7530-01-124-5660).

The electron beam irradiation process was performed at a facility in New Jersey. The exact radiation dosage and conditions will not be specified for operational security reasons. As a point of comparison, typical irradiation dosages for foods can vary from 1 kGy for fruit, 3 kGy for poultry, and up to 30 kGy for spices and seasonings (6). The unit of measurement for absorbed radiation dosages is the Gray, which is abbreviated Gy, and is equivalent to 100 rads (7). The U.S. Environmental Protection Agency estimates that mail irradiation dosages are equivalent to approximately two million times that of a chest X-ray, which they claim is more than sufficient to kill any biological agent (8). It is important to note that it is nearly impossible to expose all of the samples to a specified dosage of radiation; rather, the U.S. Postal Service guarantees that all specimens will receive a minimum radiation dosage.

The Foster & Freeman Video Spectral Comparator 2000 high resolution (VSC [Evesham, Worcestershire, UK] 2000 HR) was used to evaluate the optical properties of both the control and irradiated ink and paper samples. Any differences in infrared reflectance (IRR), infrared luminescence (IRL), and ultraviolet fluorescence were noted. In addition, absorbance spectra of all ink samples were recorded. It should be noted that the VSC 2000 HR refers to these graphs as "absorption" rather than "absorbance." These two terms are essentially interchangeable, but should not be confused with the related term absorbance. To gain a better understanding of this concept, it should be noted that the radiant flux incident upon a surface or medium undergoes three processes: transmission, reflection, and absorption (9). The application of the conservation of energy leads to the statement that transmission ( $\tau$ ), reflection ( $\rho$ ), and absorption ( $\alpha$ ) of the incident

flux are equal to unity, or  $\tau + \rho + \alpha = 1$ . When dealing with opaque items,  $\tau$  is generally assumed to be 0; thus, absorption (or absorbance) is defined by  $1 - \rho$ . Thus, absorbance is the fraction of radiant flux that is absorbed ( $\alpha = \Phi_a/\Phi_i$ , where  $\Phi_a$  is the radiant flux absorbed and  $\Phi_i$  is the incident radiant flux) and is expressed as a fraction or percentage.

The TLC analyses were performed in accordance with ASTM guideline E1422-01 (10), except where noted below. Silica gel glass plates (Merck Kieselgel 60, EM Science #5721-7) were used for all TLC analyses. Approximately five to eight microplugs (1 mm diameter) of ink were removed from each ink sample. All ballpoint inks were extracted with pyridine and all nonballpoint inks were extracted using a 50:50 mixture of ethanol and distilled water. After spotting, the plates were placed in a 100°C oven for *c.* 10 min to drive off any residual extraction solvent(s). It should be noted that ASTM guideline E1422-01 recommends that the plates be air-dried. However, internal research has found that heating plates at 100°C for short periods of time does not induce changes in the ink's colorants. All plates were developed using solvent system 1, which is a 70:35:30 mixture of ethyl acetate, ethanol, and distilled water, respectively. The solvent front was allowed to elute a distance of 4 cm from the origin of the ink spots.

A secondary set of TLC analyses were performed on random samples from each ink color set using solvent system 2. This solvent system, described in the aforementioned ASTM guideline, is a 50:10:15 mixture of *n*-butanol, ethanol, and water. This solvent system was used in the previous Smithsonian studies as well (3,4). The authors decided to see whether some of the observations noted in these studies could be reproduced using this solvent system. With the exception of the solvent system, all other procedures for analyzing the inks were the same as described in the preceding paragraph.

## Results

### Optical Properties

Overall, the optical properties (e.g., IRR, IRL) of all of the ink samples did not appear to change after the irradiation process. Although there were some minor intensity differences noted in the absorbance spectra (most likely due to minor differences in ink concentrations of the control and irradiated samples), the shapes of the curves for those inks were not significantly altered after irradiation.

There was one minor exception to this trend. Although one particular blue ink (ink 55) initially did not show any differences between the control and irradiated samples, the color of these two ink samples on blue-lined notepad paper changed noticeably over time. Interestingly, it was the color of the irradiated sample (55A) on lined paper that remained stable while the control sample (55B) became more gray/black in color over a 12-month period. The color difference can be observed in Fig. 1. This color change did not occur with the same ink on unlined photocopy paper (Fig. 2). One possible explanation for this may involve a slight difference in pH noted between the lined and unlined papers. A crude estimate of the pH (using a pHydriion Insta-check Surface pH Pencil, Micro Essential Laboratory, Inc., Brooklyn, NY) indicated that the lined paper was *c.* 1 pH unit below that of the unlined photocopy paper (pH range 6–7 vs. 7–8, respectively). The ink industry has at times added acids (e.g., oleic acid) to some ink formulations to stabilize certain basic dyes (Dr Ben Fabian, National Ink Inc., Santee, CA, personal communication, May 2006). It is possible that the decrease in pH observed in the lined

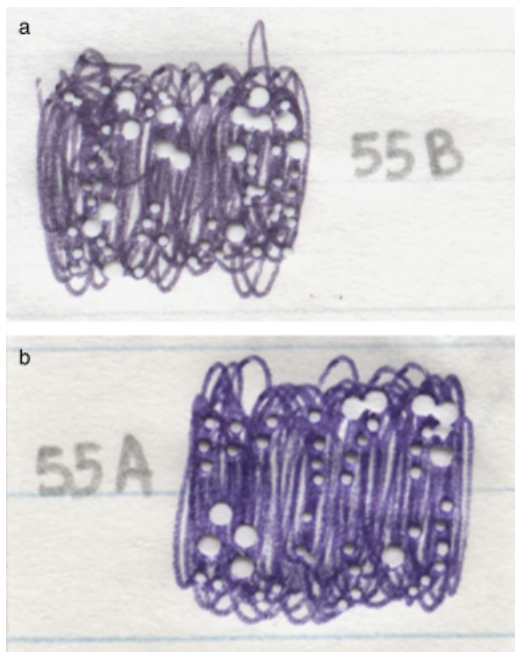


FIG. 1—(a) An image of the irradiated ink (55A) on blue-lined notepad paper. (b) An image of the control ink (55B) on blue-lined notepad paper. Although the images appear to be identical as reproduced in black and white, the differences can be discerned in the online color version of this article.

paper sample may have stabilized the particular basic dyes contained in this ink formulation while the unlined paper did not.

Upon further study, it was noted that the color change on lined paper was reflected in the absorbance spectrum as well as in the TLC analysis. An absorbance spectrum taken just after the irradiation process can be seen in Fig. 3a, while a more recent one can be seen in Fig. 3b. Note how the absorbance curve tends to flatten out in the visible region, which in effect makes it begin

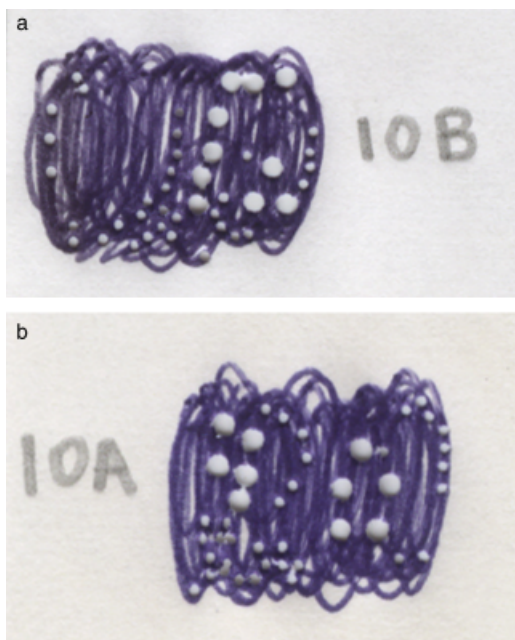


FIG. 2—(a) An image of the irradiated ink (10A) on photocopy paper. Sample 10B is the same ink formulation as sample 55A. (b) An image of the control ink (10A) on photocopy paper. Sample 10A is the same ink formulation as sample 55A.

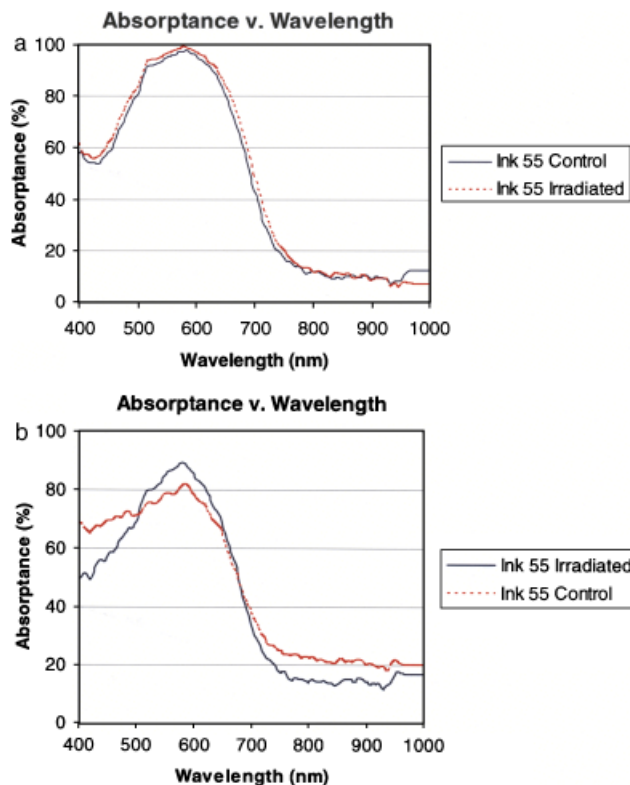


FIG. 3—(a) A graph of absorbance versus wavelength for ink 55 before and after irradiation. These data were obtained soon after the ink sample was exposed to the irradiation process. (b) A graph of absorbance versus wavelength for ink 55 before and after irradiation. These data were obtained c. 1 year after the ink sample was exposed to the irradiation process. Although the images appear to be identical as reproduced in black and white, the difference can be discerned in the online version of this article.

to resemble the spectral characteristics of a black ink rather than a blue one. An image of the TLC plate run c. 9 months after irradiation can be seen in Fig. 4. The two dark blue bands are clearly not present in the control (refer to the upper cluster of bands c. 3 cm from the origin in lane 5 in Fig. 4), but they are present in the irradiated sample. The TLC analysis of this sample was repeated twice and each time the dark blue bands were not

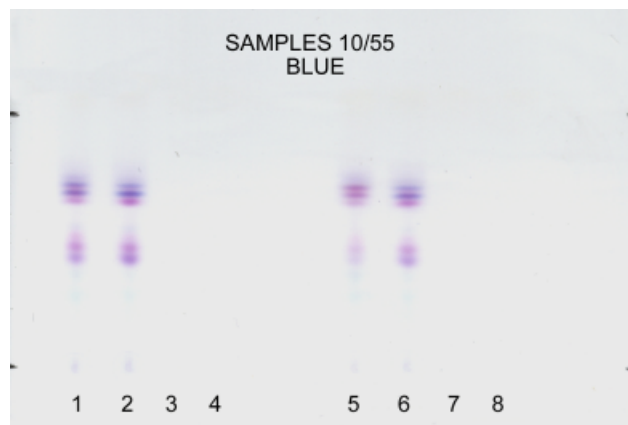


FIG. 4—An image of a thin layer chromatography (TLC) plate for ink samples 10 and 55. The two ink samples have the same formulation; however, ink 10 was deposited on photocopy paper, whereas ink 55 was deposited on blue-lined notepad paper. This TLC plate was developed c. 9 months after the samples were exposed to the irradiation process.



FIG. 5—An image of two inks deposited on photocopy paper. The paper on the left contains the irradiated samples for those two inks. The paper on the right contains the control samples for those two inks. Note the decreased intensity of the ultraviolet fluorescence of the paper on the left (irradiated) compared with the paper on the right (control).

present in the control sample. It should be noted that for the TLC plates in this and subsequent figures, a total of four spots for each ink were deposited. When viewing a standard two-ink sample plate from left to right, the spot sequence would be as follows: unlined paper ink control (lane 1); unlined paper ink irradiated (lane 2); unlined paper control blank (lane 3); unlined paper irradiated blank (lane 4); lined paper ink control (lane 5); lined paper ink irradiated (lane 6); lined paper control blank (lane 6); and lined paper irradiated blank (lane 8). For the aged sample studies, there would be the following additional four spots on the TLC plate: aged paper ink control (lane 9); aged paper ink irradiated (lane 10); aged paper control blank (lane 11); and aged paper irradiated blank (lane 12). It should also be noted that Adobe<sup>®</sup> Photoshop<sup>®</sup> was used to make minor corrections to the contrast and brightness of the ink bands shown on all of the TLC plates for better visualization.

A noticeable change in the ultraviolet fluorescence properties of the paper after irradiation was observed. Figure 5 shows the difference before and after irradiation for the unlined paper. Visibly, there was also a slight yellowing of those samples. It has been reported that a minimum radiation dosage as small as 2 kGy can lead to unacceptable damage to cellulose-based paper

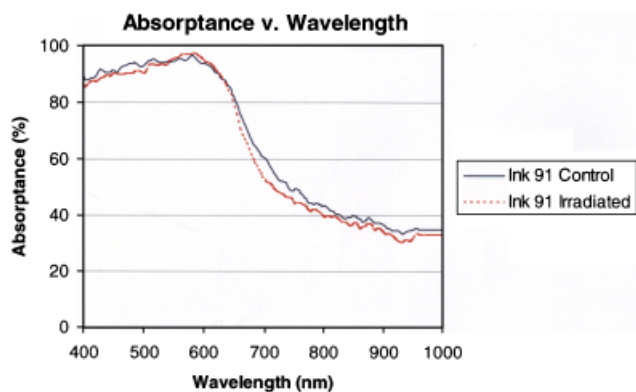


FIG. 6—A graph of the absorbance versus wavelength for ink 91 before and after irradiation. There are no significant differences between the absorbance values in these two graphs.

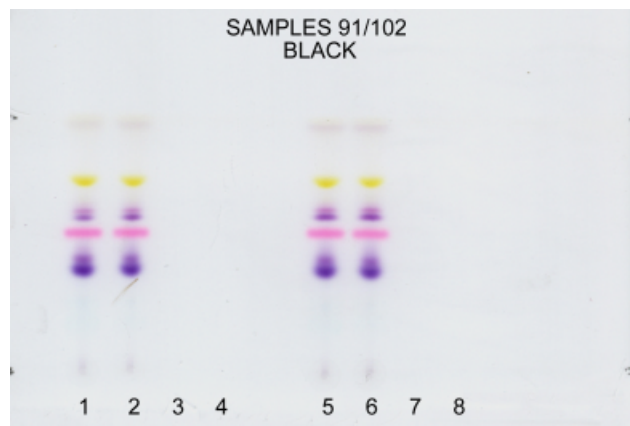


FIG. 7—An image of the TLC plate for ink samples 91 and 102. No detectable differences were observed between the control and irradiated ink samples.

(11). Dosages of 7 kGy or higher were found to result in extensive oxidation and depolymerization. At higher dosages, additional reactions can occur including acidification, loss of strength, embrittlement, and discoloration of the paper.

#### Black Inks

As mentioned before, none of the black inks showed any detectable changes in any of the optical properties evaluated after irradiation. Figure 6 shows a graph of the absorbance spectra for the control and irradiated samples of one of the black inks (ink 91). Using solvent system 1, neither the irradiated nor the control samples showed any additional or missing bands or altered mobilities when analyzed by TLC. Figure 7 shows an image of the chromatogram for ink 91.

#### Blue, Red, Green, and Yellow Inks

As with the black and blue inks, none of the blue red, green, or yellow irradiated samples showed any detectable changes in any of the optical properties. Nor were any additional or missing bands or altered mobilities detected when these samples were analyzed by TLC.

#### Aged Samples

Thirty-five inks that had been deposited on Whatman filter paper at varying times over the past 25 years were also analyzed using TLC. Using solvent system 1, neither the irradiated nor the control samples showed any additional or missing bands or altered mobilities when analyzed by TLC.

#### Solvent System 2

Approximately two dozen of the ink samples were re-analyzed using solvent system 2. In some cases, the use of an alternate solvent system can improve band separation and resolution and assist in highlighting discrepancies. Solvent system 2 replaces ethyl acetate (solvent strength on silica gel of 4.4) with *n*-butanol (solvent strength on silica gel of 3.9). In addition, the solvent ratios in the system are different. In solvent system 1, ethyl acetate comprises 52% of the system by volume, ethanol 26%, and water 22%. In solvent system 2, *n*-butanol comprises 67% of the system by volume, ethanol 13%, and water 20%. None of the inks evaluated using solvent system 2 showed any unexplained differ-

ences (e.g., discrepancies in band/spot intensities due to slight differences in spot concentrations on the TLC plate).

## Conclusions

Unlike the author's previous experience with latent prints on paper (1), the irradiation process appears to have very little effect on the chemical analysis of writing inks. Of the 97 different inks evaluated in this study, none was found to have changed with respect to dye composition as a result of the irradiation process. These results appear to contradict earlier studies conducted on a more limited sample set (3,4). It is difficult to compare directly the results from these previous studies as it is not possible to determine accurately the exact radiation dosages that the inks were exposed to during processing. In addition to internal variations in the radiation dosages possible during a single processing run, there have undoubtedly been changes (most likely decreases) in the overall radiation intensities implemented immediately after the anthrax letter attacks in October 2001. Initially after the attacks, there were considerably more reports of irradiation-induced damage to materials sent through the U.S. Postal Service (12,13). There appear to be fewer of these reports now, which may support the theory of lower irradiation levels.

The overall results indicate that the paper type, solvent system, and the age of the ink samples did not have a significant effect on the TLC profiles for either the control or irradiated inks. Of the five ink colors tested, none appeared to be more susceptible to the irradiation process than any other. However, in one instance, the control sample from one ink was found to change color with time while the irradiated one remained stable. It is interesting to note that this phenomenon was observed only on lined paper and not with the unlined photocopy paper. The lined paper had a slightly acidic pH when tested, whereas the unlined photocopy paper had a slightly basic pH. As most of the dyes used in the manufacture of ballpoint inks are basic dyes, it is possible that a chemical reaction may have occurred on the paper surface.

## Acknowledgments

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## APPENDIX A—A summary of black inks used in this study.

Sanford Sharpie Ultra Fine Point	Felt-tip
Sanford Vis-à-vis Wet Erase Fine Point	Felt
Papermate Gelstick	Gel
Papermate Widemate	Ballpoint
Papermate Erasable Ink Medium Point	Ballpoint
Papermate Erasable Ink Medium Point (10/78)*	Ballpoint
Papermate Rollerstick	Rollerball
Papermate Flair	Felt-tip
Gillette Papermate	Ballpoint
Gillette Papermate (11/86)	Ballpoint
Bic	Bulk Ink
Bic (8/9/79)	Bulk Ink
National	Bulk Ink
National (10/25/83)	Bulk Ink
Pentel Energel 0.7 mm	Gel
Pilot Dr. Grip	Gel
Pilot Dr. Grip (11/18/02)	Gel
Hi-Point RTC Free Flow Rolling Pen	Gel
Hi-Point RTC Free Flow Rolling Pen (11/21/02)	Gel
Pilot Erase-a-Gel	Gel
Uniball Gellyz Scents UM 100	Gel
Papermate Gel Roller	Gel
Papermate Gel Roller (11/18/02)	Gel
Luxoz Ace	Ballpoint
Luxoz Ace (11/15/02)	Ballpoint
Luxoz Sprint	Ballpoint
National Ink Souvenir Pen WIMA 2000	Ballpoint
National Ink Souvenir Pen WIMA 2000 (6/26/02)	Ballpoint
Hunt Elegant Writer Fine Permanent	Felt-tip
Hunt Elegant Writer Fine Permanent (3/18/95)	Felt-tip
Skillcraft Cushion Grip Medium	Ballpoint
Papermate Tattoo Stick Medium	Gel
Pilot Explorer Extra Fine	Rollerball
Pilot Easytouch Medium	Ballpoint
Papermate Medium Point	Ballpoint
Skillcraft U.S. Government	Ballpoint
Pentel Marker MS50	Felt-tip
Pentel Marker MS50 (3/78)	Felt-tip

\*The date in parentheses denotes the actual date of that sample's deposition. The samples without such notations were prepared just before the beginning of this study.

## APPENDIX B—A summary of blue inks used in this study.

Bic Blue	Ballpoint
BKA Souvenir Pen	Ballpoint
Eppendorf Corporation Souvenir Pen	Ballpoint
Sanford Uni-ball Fine	Rollerball
Netherlands Forensic Institute Souvenir Pen	Ballpoint
University of Teeside Souvenir Pen	Ballpoint
Pilot Supergrasp Medium	Ballpoint
Pilot Easytouch Fine	Ballpoint
Bic Intensity Medium	Gel
Luxoz Ranger	Ballpoint
Luxoz Ranger (11/15/02)*	Ballpoint
Luxoz Ace grip	Ballpoint
Pentel R100 Rolling Writer	Rollerball
Pentel Superball 02	Rollerball
Pentel MS50	Felt-tip
Pentel MS50 (9/77)	Felt-tip
Pentel SF50	Felt-tip
Pilot Ball Liner	Rollerball
Pilot Superfine BP	Ballpoint
Pilot Superfine BP (7/86)	Ballpoint
Pilot Fineliner	Plastic-tip
Faber Castell Uniball	Rollerball
Faber Castell Wonderiter	Plastic-tip
Faber Castell Wonderiter (3/10/88)	Plastic-tip
Papermate Write Brothers Medium	Ballpoint
Pilot Explorer Extra Fine	Rollerball
Skillcraft U.S. Government Fine	Ballpoint
Hunt Elegant Writer Medium	Felt-tip
Hunt Elegant Writer Medium (5/16/95)	Felt-tip
Forensic Alliance Limited Souvenir Pen	Ballpoint
Sakura Gelly Roll	Gel
Sakura Gelly Roll (6/12/02)	Gel
Pilot Precise Extra Fine	Rollerball
Sanford Sharpie Ultra Fine Point	Felt-tip
Sanford Vis-à-vis Wet Erase Fine Point	Felt-tip
Papermate Ultra Fine Flair	Felt-tip
Papermate Rubberstick Medium Point	Ballpoint
Papermate Rubberstick Medium Point (6/17/94)	Ballpoint
Papermate Accu-point	Rollerball
Papermate Tattoo Stick Medium Point	Gel
Papermate Gel Stick	Gel
Papermate Widemate	Ballpoint
Papermate Widemate (7/15/02)	Ballpoint

\*The date in parentheses denotes the actual date of that sample's deposition. The samples without such notations were prepared just before the beginning of this study.

## APPENDIX C—A summary of red inks used in this study.

Pentel Flowing Writer SF50	Felt-tip
Pentel B76	Ballpoint
Pentel Sign Pen Chisel Tip	Felt-tip
Pentel Sign Pen Chisel Tip (3/23/88)*	Felt-tip
Pentel for Film PM2	Felt-tip
Pilot Easytouch Medium	Ballpoint
Pilot Razor Point II	Felt-tip
Pilot Razor Point II (7/86)	Felt-tip
Faber Castell Wonderiter	Felt-tip
Faber Castell Superiter	Felt-tip
Faber Castell Superiter (2/19/90)	Felt-tip
Faber Castell Uniball Micro	Ballpoint
Pentel Marker MS50	Felt-tip
Sharpie Ultra Fine Permanent Marker	Felt-tip
Pilot Explorer Extra Fine	Rollerball
Bic Intensity Medium	Ballpoint
Crayola Red Marker	Felt-tip
Crayola Red Marker (1/24/90)	Felt-tip
Speedball Elegant Writer Medium Point	Felt-tip
Sakura Gelly Roll Red	Gel
Sakura Gelly Roll Red (7/9/02)	Gel
Sanford Vis-à-vis Wet Erase Fine Point	Felt-tip
Papermate Medium Point	Ballpoint

\*The date in parentheses denotes the actual date of that sample's deposition. The samples without such notations were prepared just before the beginning of this study.

## APPENDIX D—A summary of green inks used in this study.

Pentel for Film PM2	Felt-tip
Pentel for Film PM2 (4/77)*	Felt-tip
Sanford Vis-à-vis Wet Erase Fine Point	Felt-tip
Pentel Rolling Writer	Ballpoint
Pentel S510	Felt-tip
Pilot Razor Point II	Rollerball
Pilot Razor Point II (7/86)	Rollerball
Pentech Metallicz	Gel
Pentech Metallicz (11/19/02)	Gel
Pentech Hot Sparklez	Gel
Sakura Gelly Roll Green	Gel
Sakura Gelly Roll Green (7/9/02)	Gel
Bic	Ballpoint
Bic (8/8/00)	Ballpoint
Scillcraft U.S. Government Medium Point	Ballpoint
Bic Intensity Medium Point	Gel
Bic Intensity Medium Point (8/8/00)	Gel
Crayola Green	Felt-tip

\*The date in parentheses denotes the actual date of that sample's deposition. The samples without such notations were prepared just before the beginning of this study.

## APPENDIX E—A summary of yellow inks used in this study.

Fluorescents 54-32-A1	Ballpoint
Pentel for Film PM2	Felt-tip
Pentel for Film PM2 (4/77)*	Felt-tip
Crayola Yellow	Felt-tip
Crayola Yellow (8/89)	Felt-tip
Crayola Fluorescents	Felt-tip
Staedtler Textsurfer Classic	Felt-tip
Staedtler Textsurfer Classic (11/18/02)	Felt-tip
Gelly Roll Yellow	Gel
Crayola Pastels	Felt-tip

\*The date in parentheses denotes the actual date of that sample's deposition. The samples without such notations were prepared just before the beginning of this study.

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