J Forensic Sci, May 2003, Vol. 48, No. 3
Paper ID JFS2002342_483
Available online at: www.astm.org

Gerald M. LaPorte,[1] M.S.F.S. and Robert S. Ramotowski,[2] M.S.

The Effects of Latent Print Processing on Questioned Documents Produced by Office Machine Systems Utilizing Inkjet Technology and Toner*

ABSTRACT: Counterfeiting of currency and identity documents, death threats, illegitimate business transactions, and terrorist-related activities are some examples of the types of crimes that often involve documents produced from printers and copiers. Although standard protocol typically requires a questioned document (QD) examination prior to latent print (LP) processing, occasionally, items of evidence may be submitted for a QD examination following the application of a series chemicals utilized in the development of latent fingerprints. In such cases, the forensic examiner must take into account any previous treatments prior to initiating an examination on documents produced with a printer or copier. This study was devised to examine the effects of a latent print development technique [ninhydrin, physical developer, and a bleach enhancer] on the physical and chemical examination of documents produced from copiers and printers.

KEYWORDS: forensic science, questioned documents, thin layer chromatography, inkjet printing, electrophotography, toner, ink analysis, latent print development, counterfeiting

With their rapid technological advances and superior performance, office machine systems utilizing inkjet technology and toner have undoubtedly evolved into the most dominant print technologies used in offices and homes. Inkjet printing, typically considered slow and inefficient for high output, has been highly regarded because of its capability to produce a high quality color product at affordable prices making them predominant in households. Laser printers and photocopiers, which are toner based systems that utilize electrophotography, were once considered expensive, but capable of rapid, high quality, black text output, thus making them prevalent in offices. However, continual refinements are eliminating their respective disadvantages, making each system viable for both personal and business use. Laser printers and photocopiers are becoming much more affordable, inkjets are becoming more efficient, and each is capable of color reproduction nearing photographic quality. Their tremendous popularity has paved the way for a significant increase in criminal acts involving inkjet and toner systems.

Forensic examiners are routinely required to analyze questioned documents (QD) produced by printers and photocopiers. They are becoming rampant sources of crime for three reasons. First, they are widely available and easily accessible. Second, because they are used with such frequency, more business documents are generated, which effectively creates more opportunity for fraud. Finally, software advancements, in conjunction with high quality color printing, have allowed users to reproduce and counterfeit documents and currency much more easily. In fact, the U.S. Secret Service maintains a counterfeit document database of nearly 90,000 specimens. Taking into account credit cards, travelers checks, drivers licenses, passports, alien registration cards, social security cards, and birth records, 61.6% of the documents have been created using inkjet, toner, or a combination of the two technologies. Further, records at the U.S. Secret Service show that in 1995, domestically-passed counterfeit U.S. currency (Federal Reserve Notes) produced using toner and inkjet totaled $2,440,229 and $174,924, respectively. In 2001, counterfeit U.S. currency produced using toner and inket technologies accounted for $1,151,791 and $18,403,257, respectively.

Depending on the nature of the crime, the forensic examiner may be requested to authenticate a questioned document, determine if documents were produced using a common source (e.g., associating multiple documents), or ascertain the make, model, and/or manufacturer of the office machine system used. Occasionally, evidence submitted for analysis may be processed for latent prints prior to QD examination. Horton and Shaver (1) acknowledged this and examined the effects of latent print processing on the writing produced by ballpoint pen inks. The authors concluded that “extreme caution should be exercised by a FDE [forensic document examiner] when comparing or differentiating inks” following latent print (LP) processing. LP processing may involve a series of steps which include the application of chemical(s) that may alter the visual appearance of the document and possibly affect the chemistry of inks and toners. Therefore, the forensic examiner

[1] United States Secret Service, Forensic Services Division/Questioned Document Branch, Washington, DC.
[2] United States Secret Service, Forensic Services Division/Research Section, Washington, DC.

* This work was presented in part at the 2002 Mid-Atlantic Association of Forensic Scientists (MAAFS) Annual Meeting, April 25, 2002, Gaithersburg, MD.

All references pertaining to manufacturers and their products do not imply endorsement by the United States Secret Service or the authors.

Received 10 Oct. 2002; accepted 3 Dec. 2002; published 12 Mar. 2003.

must be aware of each aspect of LP processing, as well as consider the impact of chemical treatment on the ink or toner on a QD when conducting physical (e.g., microscopic and optical spectral) and chemical examinations.

To understand the scope of the analysis, it is helpful to briefly describe the fundamental differences between office machine systems utilizing inkjet technology and those that use toner. It is not the intention of this article to provide a detailed description of each process; however, there are a number of sources that can provide a comprehensive description of printers and photocopiers (2–5).

Inkjet Systems

Inkjet printing is a non-impact method that uses tiny nozzles to emit fine droplets of ink onto a document. As the paper passes through the inkjet, a printer head containing the ink moves back and forth horizontally. The ink deposition is controlled digitally via the software and is sprayed so that multiple gradients of dots accumulate to form an image with variable tones of color. Most modern color printers are capable of printing in four colors—cyan, magenta, yellow, and black (CMYK). Some manufacturers (e.g., Canon®) are now producing printers containing more than four ink wells, although the additional colors are variations of the cyan and magenta in order to extend the gamut of colors.

Although several technologies exist based on how the ink droplets are dispersed onto a substrate, two dominant inkjet systems are currently in use: continuous and drop-on-demand. In continuous jet systems, a continuous stream of ink is broken into droplets. The droplets, which are uniformly sized, are then charged and deflected while the excess ink is removed via deflection allowing an image to be created. Drop-on-demand (DOD) systems, most common for home and office use, emit only the droplets necessary to create an image. DOD printer inks are typically formulated to be used on porous or absorbent surfaces. Within the DOD printers, there are four sub-classifications including thermal, piezoelectric, electrostatic, and acoustic (the latter two are uncommon and therefore not acknowledged in detail in this discussion). Thermal and piezoelectric printers operate on the principal that a drop of ink is forced onto the printing media. Thermal technology utilizes high heat to vaporize a bubble, which causes it to burst and emit a drop of ink, while piezoelectric printers use an electrical voltage rather than heat to emit an ink droplet. Because the two systems are fundamentally different, the compositional chemistry of their respective inks varies. The inkjet printers used for this study were all classified as thermal and DOD. These systems typically utilize a fluid, water-based ink; however, it is worthy to note that although these systems are predominant for home and office use at this time, piezoelectric printers are gaining popularity and their use is likely to become significantly more common.

Toner Systems

Laser printers and photocopiers function by means of similar technology, with the exception of the light source. Photocopiers use bright light while laser printers employ light emitting diode (LED). Each contains a rotating drum with a photoconductive coating that permits it to retain an electrostatic charge. The surface of the drum is illuminated with a light source causing the exposed areas to be charged. The light source is controlled and manipulated by a series of signals (digital in laser printing and analog or digital in photocopiers) based on the image to be printed. As the drum rotates, oppositely charged toner is deposited, thus adhering to the illuminated areas that have been charged with a positive potential. The toner mainly consists of organic resins, polymers, and colorants such as carbon black, nigrosines, phthalocyanines, azopiments, and quinacrodone (6). Once the paper enters the system, it is exposed to an electrical current that imparts a static electrical charge onto the paper. The paper then passes through a series of rollers and comes in contact with the rolling drum causing the toner, hence an image, to be transferred to the paper. Since the non-image areas of the drum are charged, they do not attract oppositely charged toner and result in white areas on the paper. Afterwards, heat and/or pressure are applied to permanently fuse the toner. For a multicolored process, the same steps are repeated for each colored component, i.e., cyan, magenta, and yellow.

Forensic Examination

Physical examinations are performed on a printed document to determine the printing process employed to manufacture a questioned exhibit. Additionally, chemical examinations are used to compare two specimens, or to possibly classify or identify the make and model of a possible printer. Indeed, the microscopic examination is a critical part of the process to help the examiner identify the method(s) of production. Using at least 10× magnification, inkjet printing appears as a series of irregular colored dots at planar level with some peripheral bleeding. In contrast, laser printers and photocopiers scatter toner particles over the non-image areas. The image areas do not appear as discrete dots of print like inkjet, and although the toner is heated and flattened, the image still appears to be raised. Figure 1 reveals microscopic comparisons of documents

FIG. 1—*Comparison of documents produced using color inkjet printing (left) and toner (right) at 40× magnification.*

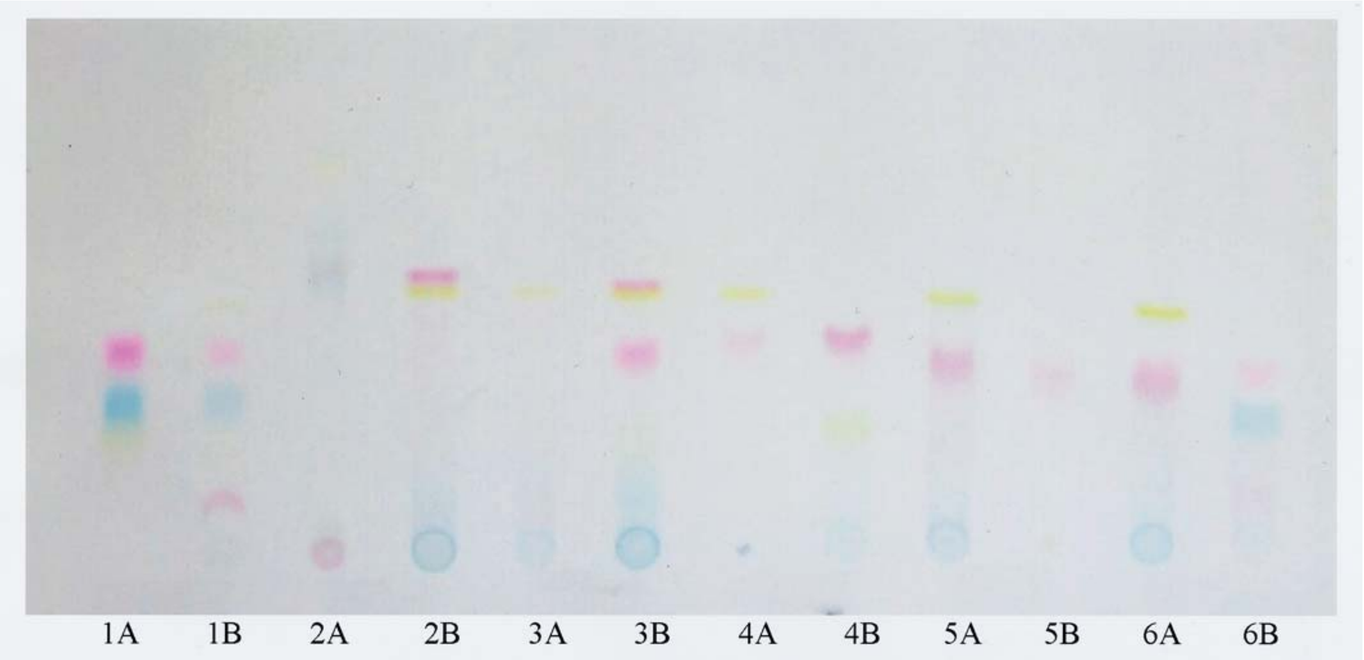

FIG. 2—*Thin layer chromatogram of inkjet printers showing the differences in color components between six different manufacturers (numeric designation) and the differences in models produced by the same manufacturer (alpha designation).*

produced with inkjet ink and toner. Accordingly, the first objective of this study was to determine if latent print processing would preclude the examiner from correctly determining the type of printing process employed.

Chemical analysis of writing inks by means of thin layer chromatography (TLC) has been widely accepted in the field of forensic science since it is very effective for separating and identifying various colored components such as dyes and their by-products (6–9). Undoubtedly, the chemical composition of printing inks can be quite complex since there are many factors to take into consideration when formulating quality inks. With increasing market competition and demand for high quality output, manufacturers have developed inks that must be able to withstand high heat, dry quickly so as not to smear easily, endure some physical handling while maintaining a quality appearance, and have near photographic quality. Since companies allocate significant amounts of resources into research and development, ink formulations are proprietary and difficult to mimic. Hence, the chemistry of color inks and toners does vary from one manufacturer to the next, often making them discernible through forensic analysis. TLC can then be used to determine the colorant composition found in most printers and copiers by comparing with known standards. In fact, the U.S. Secret Service maintains a library of standards consisting of various makes, models, and manufacturers of both color printers and copiers that can be used for comparison and/or identification. Figure 2 demonstrates some examples of the different TLC characteristics of twelve different inkjet printers. Pagano et al. (6) provide an excellent discussion regarding the forensic analysis of inkjet inks based on colorant separation and the comparison with a library of standards. Therefore, the second objective of this study was to ascertain what effects latent processing had on the thin layer chromatography results of inkjet and toner systems.

Methods and Materials

A total of eight color inkjet samples on non-coated paper (Xerox® Multipurpose 4200 8.5 × 11, 20 lb. basis weight, 84 bright) were taken from the following printers: Canon® Multipass C755 Color Bubble Jet™, Canon® S400 Color Bubble Jet™, Hewlett Packard® (HP) DeskJet™ 960C, HP OfficeJet™ K80, Lexmark™ Z53 Color Jetprinter™, Lexmark™ Z82 Color Jetprinter™, Xerox® WorkCentre™ 470cx, and Xerox® M940 Express Mode™. In addition, samples from four toner systems were obtained from the HP Laserjet 1200 series (black only), Brother® MFC 4600 (black only), Xerox® 4900 Color Laser, and Canon® Color Laser. Test samples for physical and chemical examination were obtained prior to latent processing treatment (controls). A similar set of test samples were treated first with ninhydrin reagent, then with the physical developer, and finally with the bleach enhancer. After each treatment, the treated test samples were examined optically (physically) and chemically (chromatographically), and compared with the control samples. The samples were optically examined using a stereomicroscope to determine the printing process and identify any significant morphological changes. The Foster and Freeman® Video Spectral Comparator (VSC) 2000 was used to note any changes in the ultraviolet fluorescence (UVF), infrared reflectance (IRR), or infrared luminescence (IRL).

Thin Layer Chromatography

The chemical examination entailed the use of TLC. The TLC plates were Whatman Polyester plates coated with silica (Whatman® Catalog Number 4410221) and the solvent system utilized was ethyl acetate:ethanol:water in a ratio of 70:35:30 (solvent system I). Three 5 mm hole punch specimens were removed from the standards and each of the printed samples following their respective treatments. An arduous effort to obtain hole punches with equal amounts of each of the four colors (CYMK) from every sample was attempted. The Xerox® WorkCentre™ 470cx printer sample only contained the MYK colors. The inkjet samples were extracted with an ethanol:water solution (1:1) and the toner samples were extracted with 100% chloroform. The samples were spotted on their respective TLC plates and allowed to elute in solvent system I.

Latent Print Processing

The choice of visualization reagents needed to develop latent prints is dependent upon the nature of the substrate. With porous (i.e., paper) substrates, the sequence typically involves a combination of any of the following: visual/white light examination, use of alternate light sources/lasers, iodine, 1,8-diazafluoren-9-one (DFO), ninhydrin, zinc chloride/nitrate, physical developer (PD), multi-metal deposition, silver nitrate, and bleach (10). In this study, each of the inkjet and laser printed samples was subjected to processing, in sequence, with ninhydrin reagent, physical developer, and a bleach enhancement solution. The composition of each of the solutions is provided below. The reagent formulae provided are outlined in the U.S. Secret Service Guide to Chemical, Optical, and Physical Methods for the Visualization of Latent Prints.

The ninhydrin reagent solution (0.6% w/v) was prepared by dissolving 120 g of ninhydrin into one liter of absolute ethanol. Once the ninhydrin had completely dissolved, it was added to 19 L of petroleum ether and mixed thoroughly. The ninhydrin reagent was dispensed into a metal pan into which the various samples were dipped one at a time for a duration of a few seconds. The samples were then allowed to air dry. Development of the visibly colored product, Ruhemann's Purple, was accelerated by placing the samples into a humidity chamber set at 60°C and 80% relative humidity for approximately 30 min.

PD is an aqueous silver nitrate solution that selectively deposits silver particles on latent print residues. The reagent consists of different components, the first being an oxidation-reduction (redox) solution, which is prepared by mixing 30 g of ferric nitrate nonahydrate, 80 g of ferrous ammonium sulfate hexahydrate, and 20 g of citric acid monohydrate into 1 L of distilled water. The second component is a detergent/surfactant solution. This is prepared by mixing 4 g of n-dodecylamine acetate and 4 mL of Synperonic N (nonyl phenol ethoxylate) into 1 L of distilled water. The third component is a 20% w/v silver nitrate solution. This is prepared by mixing 20 g of silver nitrate into 100 mL of distilled water. This solution provides the silver that is converted to colloidal form, which is ultimately deposited on the latent print residue.

The working PD reagent solution is prepared by mixing 900 mL of the redox solution, 40 mL of the detergent solution, and 50 mL of the silver nitrate solution together. Samples were pre-treated by placing them into a malic acid bath for approximately 15 min. This acid pre-wash solution was prepared by mixing 25 g of malic acid into 1 L of distilled water. After the acid pre-treatment, the samples were placed into the PD reagent for approximately 20 min. The samples were then rinsed with tap water to remove any remaining PD reagent solution and then placed on blotters to dry. To speed up the drying process, the samples were placed on an Arkay Stat-Dri (model ST-22) drum-style photographic print dryer.

Household bleach (typically 5% sodium hypochlorite) is used in diluted form after the PD process to darken faintly developed prints. The hypochlorite present in the bleach reacts with the gray colored silver deposited on the prints to produce a dark black colored silver oxide. In addition, the bleach can decolorize any remaining Ruhemann's Purple stains from the ninhydrin process. The bleach solution is prepared by mixing approximately 1 part bleach to 3 parts water.

Results and Discussion

Physical Examination

The application of ninhydrin to the inkjet samples did not impede the microscopic examination, allowing the examiner to accurately determine the printing process used to produce the sample. There was very little microscopic evidence, if any, to indicate that the samples were processed with ninhydrin. However, analysis using the VSC 2000 and a dual wavelength UV source (UV Products®) indicated significant changes in the optical properties of the specimens. All the treated inkjet samples exhibited a quenching fluorescent effect when observed with the dual wavelength UV source (254 nm and 366 nm). In addition, using the VSC 2000, an increased luminescence was observed under visibly-excited IRL, i.e., dequenching, but there was no significant change in IRR. To determine why there was a significant change in the IRL, the authors applied a mixture of ethanol and petroleum ether to a blank paper sample and observed similar IRL dequenching, surmizing that the change was a result of the solvents used in the ninhydrin solution interacting with the paper.

Following the PD step, significant macroscopic, as well as microscopic changes were evident by deposits of silver bead-like material and extreme color fading of the three process colors; however, the cyan exhibited some varying resiliency when comparing the manufacturers. The cyan component is typically made from a phthalocyanine (dyes and pigments containing a tetrabenzoporphyrazine nucleus) complexed with a metal such as copper or cobalt. The use of phthalocyanines is ubiquitous throughout the ink industry because they exhibit excellent color fastness and can be combined to form a wide gamut of brilliant tones (11). These compounds can be very important to the forensic examiner since they can easily be modified to alter their solubility in water by the substitution of sulfonic groups. It is the variation in the ratio of mono-, di-, tri-, and unsubstituted sulfonated phthalocyanines which produces blue bands on the TLC plate with different retardation factors. The black components of the inkjet samples did not appear to be significantly affected and this is likely due to the insolubility of the carbon black pigment in the latent print solutions. Since PD is an aqueous reagent and inkjet inks are approximately 70% water based, the significant loss of color components was not surprising.

The use of a hypochlorite enhancer virtually obliterated the three processing colors, but did not remove significant amounts of black. Ultimately, this could make it very difficult for an examiner to conclude that the questioned exhibit was produced using a color inkjet printer. In addition, although the black inkjet ink is resilient following the PD enhancer step, definitively determining the printing process based on the presence of black only is very difficult. Evidence of PD processing followed with bleach is obvious by the appearance of a gray background accompanied by silver beads. Figure 3 shows microscopic comparisons of an inkjet control and samples following each step of latent print development (i.e., ninhydrin reagent, physical developer, and bleach enhancer).

Unlike the previously described samples produced with inkjet ink, the color components in the toner samples were neither partially nor completely removed following any of the treatments. The processing colors remained glistening and particulate, but indeed,

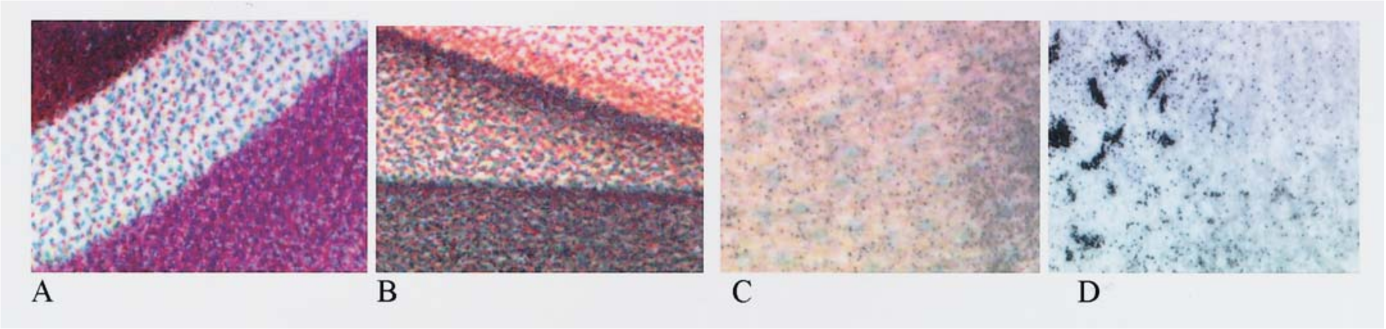

FIG. 3—*Comparison of inkjet samples: A) untreated (control); B) following ninhydrin reagent; C) following physical developer and; D) following bleach enhancer.*

there was microscopic evidence of physical developer and enhancement solution following their respective application.

Chemical Examination

The ninhydrin treatment did not affect the elution and/or position of the dye/pigment components in the thin layer chromatograms. The retardation factor (Rf) of each color band for the respective samples was consistently the same. These results are consistent with Horton and Shaver (1) who found that the ninhydrin process did not reveal any significant changes to 15 ballpoint inks when examined by TLC. However, a faint purple-gray band, which was not evident in the standards, appeared at an approximate Rf of 0.60 in all of the processed inkjet samples. Since the band was not present in the toner samples that had been extracted with chloroform, the hypothesis was that the ethanol and water (1:1) solution was responsible for extracting the ninhydrin and producing the resulting purple-gray band. To test this theory, one microgram and 100 microgram samples of alanine were applied to Whatman® filter paper and reacted quantitatively with ninhydrin to give the characteristic purple color. The samples were extracted with ethanol and water (1:1) and chromatographed under the same conditions as the inkjet samples. As expected, the purple-gray band appeared at an approximate Rf of 0.6 for both samples. It should be noted that these bands significantly fade within a 24 h period, so it is advisable to note and/or document these findings immediately.

As expected, based on the physical results, the color components in the inkjet samples were much more difficult to extract following the PD process, resulting in significant fading of the colored bands when visualized on the chromatograms. However, the color components from all the printers were not chemically altered so as to affect the elution profile of the dye/pigment components on the chromatogram. The Rf values of each colored band for the respective samples were consistently the same. These results are consistent with those of Horton and Shaver (1) who found that LP processing did not cause any significant changes to the fifteen ballpoint inks when examined by TLC. Furthermore, HP and Xerox® incorporate an UV fluorescent magenta (254 nm and 366 nm) into their inkjet formulations. This component did not change following the ninhydrin or PD processing. As mentioned previously, the PD enhancer obliterated the process colors, thus it was not feasible to interpret TLC results for the inkjet inks.

The components of the toner samples were not chemically altered so as to affect the TLC elution profile. All the bands present in the standards were present in the processed samples at the same Rf and there was very little affect on their relative solubilities following each successive treatment.

Conclusion

Occasionally, items of evidence may be submitted for a QD examination following the application of a series of treatments and chemicals utilized in the development of latent fingerprints. The authors have demonstrated that it is feasible to conduct a forensic examination on documents treated for latent prints that were produced using either inkjet or toner-based systems. In cases where inkjet ink was used, it is possible to conduct physical and chemical analyses following the addition of ninhydrin. An examination can be severely hindered by the use of PD, and the addition of a bleach enhancer virtually obliterates the inkjet ink. When examining documents created by a photocopier or laser printer, the examiner can make accurate physical and chemical assessments regardless of the steps employed in the development of latent fingerprints for this study. Nevertheless, caution is warranted when analyzing processed questioned documents and it is critical for the forensic examiner to understand the procedure utilized and the chemicals applied to a processed document prior to beginning physical and/or chemical examinations.

Acknowledgments

The authors wish to gratefully acknowledge Dr. Antonio A. Cantu and Larry F. Stewart from the United States Secret Service, Forensic Services Division for their input and editorial comments. As well, we are thankful to Lorelei Pagano and Anthony Chapa from the United States Secret Service, Counterfeit Division for providing statistical data regarding counterfeit Federal Reserve Notes.

References

1. Horton RA, Shaver LC. The effects of latent print processing on ballpoint pen inks, The Chesapeake Examiner 2000;38(1):5–11.
2. Fenton HM, Romano FJ. On-demand printing: the revolution in digital and customized printing, 2nd edition. Graphic Arts Technical Foundation, Pittsburgh PA, 1997.
3. Pocket pal: a graphic arts production handbook, 17th edition. International Paper Company, Memphis, TN, 1997.
4. Pond SF. Inkjet technology and product development strategies. Torrey Pines Research. Carlsbad, CA, 2000.
5. Andreotolla MA. Inkjet technology. in: diamond AS. Handbook of imaging materials. Marcel Dekker, Inc. New York, NY, 1991.
6. Pagano LW, Surrency MJ, Cantu AA. Inks: forensic analysis by thin-

layer (Planar) chromatography in encyclopedia of separation science. Wilson ID, editor. Academic Press. New York, NY, 2000:3101–9.
7. Aginsky VN. Forensic examination of “slightly soluble” ink pigments using thin-layer chromatography. J Forensic Sci 1993;38:1131–3.
8. Brunelle RL, Pro MJ. A systematic approach to ink identification. J Assoc of Official Analytical Chemists 1972;55:823–6.
9. Kelly JD, Cantu AA. Proposed standard methods for ink identification. J Association of Official Analytical Chemists 1975;58:122–5.
10. Lee HC, Gaensslen RE. Methods of latent fingerprint development. In: Lee HC, Gaensslen RE, editors. Advances in fingerprint technology, 2nd edition. CRC Press LLC. Boca Raton, FL, 2001;105–75.
11. The Society of Dyers and Colourists. Colour Index, 3rd ed. Lund Humphries, Bradford and London, Great Britain. 1971;4:4617–22.

Additional information and reprint requests:
Gerald LaPorte, MSFS
United States Secret Service
Forensic Services Division
950 H Street NW
Washington, DC 20223
E-mail: glaporte@usss.treas.gov