

TECHNICAL NOTE

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Detection of Visible and Latent Fingerprints Using Micro-X-ray Fluorescence Elemental Imaging*

ABSTRACT: Using micro-X-ray fluorescence (MXRF), a novel means of detecting fingerprints was examined in which the prints were imaged based on their elemental composition. MXRF is a nondestructive technique. Although this method requires *a priori* knowledge about the approximate location of a print, it offers a new and complementary means for detecting fingerprints that are also left pristine for further analysis (including potential DNA extraction) or archiving purposes. Sebaceous fingerprints and those made after perspiring were detected based on elements such as potassium and chlorine present in the print residue. Unique prints were also detected including those containing lotion, saliva, banana, or sunscreen. This proof-of-concept study demonstrates the potential for visualizing fingerprints by MXRF on surfaces that can be problematic using current methods.

KEYWORDS: forensic science, fingerprint, latent, XRF, X-ray fluorescence, micro-XRF, MXRF, imaging, spectroscopy

Despite the numerous methods available to forensic scientists, fingerprints left on certain types of substrates or made under certain conditions are sometimes difficult to detect with conventional contrast methods. Examples of potentially troublesome substrates include: fibrous papers, textiles, woods, leather, plastic, adhesives, and human skin (1,2). Prepubescent children's fingerprints can also be particularly difficult to detect (3) due to the absence of sebum and the presence of relatively volatile free fatty acids (4). Another category of latent fingerprints that can prove troublesome are those present on dark or multicolored backgrounds (5,6). Hence, any new fingerprint detection methods that can provide forensically useful information to complement or supplement current methods would be beneficial.

Chemical imaging, in which an image is separated into its component colors, has recently been shown to be effective at enhancing subtle visual contrast between a print and the background (5). Recently, a few spectroscopic methods have also been used to analyze fingerprints. Spectroscopy can be used to examine the chemical components of a fingerprint instead of relying on the visible contrast between the print and substrate. However, very few published studies have used spectroscopy to actually detect an image of a fingerprint; most of the spectroscopic studies present data acquired from one or several spots on the sample.

Wilkinson et al. (7–10) have published several abstracts in which they used synchrotron infrared (IR) microspectroscopy to map chemical heterogeneities in human fingerprints, and Williams

et al. (3) utilized a lab-based IR microspectroscopic instrument to study the chemical composition of fingerprints by collecting spectra from spots along the friction ridges.

Koch et al. (11) imaged sections of fingerprints on gold and stainless-steel surfaces based on elemental composition using SEM energy-dispersive X-ray spectroscopy (EDS) and imaging Auger spectroscopy (AES). The paper also discusses the possibility of imaging an entire fingerprint by modifying the instrument detectors. Migron and Mandler (12) also used AES and SEM/EDS to analyze fingerprints. A novel method was also developed by Nolan et al. (13) in which they treated prints with a physical developer and imaged the prints with backscattered electron SEM.

In the present work, micro-X-ray fluorescence (MXRF) was used to acquire elemental spectra and to directly image both visible and latent fingerprint friction ridges via inorganic elements present in the print residue (sodium and higher atomic number elements). MXRF is currently capable of imaging elemental heterogeneities across surfaces up to several hundred cm² in size with a spatial resolution ranging from a few micrometers to a few 100 μm (instrument dependent) (14,15). Thus, the MXRF imaging resolution and macroscopic capabilities are ideal for detecting fingerprints. X-ray fluorescence (XRF) entails irradiating a sample with a primary X-ray beam and detecting X-rays emitted from the sample. The energies of these emitted X-rays are characteristic of the elements present in the sample (16). In MXRF, a small spot on a sample surface is irradiated with X-rays, and fluorescence X-rays emitted from the spot are detected as a function of location on the sample surface. By moving the X-ray beam across the sample surface, a two-dimensional elemental distribution image is generated by detecting variations in X-ray intensity versus position. Such intensity differences correlate with the relative concentration changes of a given element dispersed across the sample (14,17).

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In addition to various organic components, fingerprint residue also contains salts (1). Therefore, MXRF could be a viable method for detecting a fingerprint based on elements present in the friction ridges (sodium and higher atomic numbers). To the best of the authors' knowledge, only one XRF study of human fingerprints has been published to date (18). In that study, the authors utilized grazing angle XRF to collect elemental spectra from a variety of forensic samples including human fingerprints. In the current work, MXRF was used not only to study elemental spectra, but to image visible and latent fingerprints directly via X-ray emission from the elements present.

Because the elemental residue present in a fingerprint is detected with MXRF, it offers a unique means of visualizing fingerprints that might prove difficult to identify by conventional methods. Energy-dispersive MXRF used here is a nondestructive technique (19) if the sample can fit inside the spectrometer, so the sample is available for further analyses or archiving. Additionally, no developing agents are required. Some fingerprint examples where MXRF may prove uniquely valuable include analyzing young children's prints, fingerprints on dark, multicolored, or porous surfaces, and prints containing special residues such as saliva, gunpowder, or sunscreen.

In the current work, MXRF was used to image regular sebaceous fingerprints as well as special prints that could be present in real-world forensic cases but potentially undetectable by contrast methods. Because this work is focused on demonstrating the feasibility of MXRF for elemental imaging, we have not placed significant emphasis on obtaining the maximum resolution possible from the X-ray maps. Nonetheless, obtaining image resolution compatible with the NIST biometric standards (20) is straightforward using off-the-shelf commercial equipment.

Materials and Methods

Instrumentation

Two MXRF instruments were used in this work to demonstrate the efficacy of our approach regardless of the brand of instrument or type of X-ray source used. A Kevex Omicron instrument was used to analyze a majority of the samples. (Kevex is now owned by Thermo Electron, Waltham, MA) This is an energy-dispersive XRF spectrometer with a 100 W molybdenum X-ray source. To irradiate a small analysis spot on the sample with a high flux of X-rays, the instrument was retrofitted with a monolithic polycapillary optic (designed by X-ray Optical Systems, Inc., East Greenbush, NY). This optic provided an analysis spot $\sim 190 \mu\text{m}$ in diameter for these studies (i.e., 134 pixels per inch; ppi). The instrument uses a Si(Li) detector with an energy resolution of $\sim 160 \text{ eV}$. The sample stage was moved laterally in the x and y directions across a stationary X-ray beam. The sample chamber was flushed with helium for the analyses. All MXRF spectra and images were acquired using X-ray source power settings that maximized the total count rates without exceeding a detector dead time of 50%. The XRF elemental $K\text{-L}_{2,3}$ ($K\alpha$) peak intensities were used to collect images for elements other than the source molybdenum scatter peak ($L_3\text{-M}_{4,5}$ peak ($L\alpha$) used). Note: the IUPAC nomenclature for labeling XRF elemental lines is used here (21). Unless otherwise specified, the images collected with this instrument consisted of 256×256 pixels.

An EDAX (Mahwah, NJ) Eagle II XPL MXRF instrument was used to compare to or supplement the Kevex Omicron MXRF images collected from several samples. This is an energy-dispersive XRF spectrometer using a 40 W rhodium X-ray source. This

instrument uses a monolithic polycapillary optic (designed by X-ray Optical Systems, Inc.) for producing a small X-ray analysis spot with a minimum diameter of $\sim 40 \mu\text{m}$ (i.e., 635 ppi). The instrument uses a Si(Li) detector with an energy resolution of $\sim 155 \text{ eV}$. The sample chamber was purged with vacuum for all analyses. All images were acquired using an X-ray source power setting of 20 kV and a current setting that resulted in a net count rate of $\sim 10,000$ cps. The same elemental mapping process used with the Kevex system was also used with the EDAX instrument.

A Perkin-Elmer (Boston, MA) Spectrum Spotlight 300 FT-IR imaging microscope was used to examine one sample in this paper and supplement the MXRF image data. This system used a $25 \mu\text{m}$ diameter IR beam (i.e., 1016 ppi), and the image was collected in the IR absorbance mode at an energy resolution of 16 cm^{-1} . Eight spectra were acquired per pixel and coadded. The image resolution was 300×300 pixels.

Sample Preparation

Two subjects supplied the fingerprints examined in this work. All fingerprints prepared on polypropylene were made using 4- μm -thick film sold by Chemplex (Palm City, FL; trade name Prolene[®]).

Sebaceous Fingerprints

Subject #1 made a fingerprint on a polypropylene film after touching the forehead. The subject's hands were not washed prior to touching the forehead. The fingerprint sample was imaged with the EDAX MXRF instrument using a $1000 \mu\text{A}$ X-ray source current. The following elemental relative intensity images were collected: Na, Mg, Si, S, Cl, K, Ca, Ti, Mn, Fe, Cu, and Rh (source scatter). Each elemental image consisted of 512×400 pixels corresponding to an area from the middle of the fingerprint. The intensity collection time per pixel was 200 msec.

Fingerprints after Perspiring

After wearing a latex glove for ~ 2 h to induce perspiration on the hand, subject #2 removed the glove and left a fingerprint on polypropylene. The subject's hands were not washed prior to perspiring and making the print. The sample was imaged with the Kevex MXRF instrument using an X-ray source power of 21 kV and 3 mA. The following elemental relative intensity images were collected: Na, Mg, Al, Si, P, Cl, K, Ca, Fe, and Mo (source scatter). The elemental images were acquired from a $17.4 \text{ mm} \times 17.4 \text{ mm}$ region of the sample. The elemental intensity collection time per pixel was 1.06 sec.

Lotion

Subject #2 applied Eucerin lotion (Beiersdorf Inc., Wilton, CT) to unwashed hands and rubbed the lotion in until the white color was no longer visible. A fingerprint was made on polypropylene and was examined by both MXRF instruments and the IR microscope. The sample was imaged with the Kevex MXRF instrument using source power settings of 15 kV and 3 mA. The following elemental relative intensity images were collected: Na, Mg, Al, Si, P, Cl, K, Ca, Fe, and Mo (source scatter). Each elemental image consisted of 512×512 pixels corresponding to a $16.8 \text{ mm} \times 19.4 \text{ mm}$ (x, y) region of the sample. The elemental intensity collection time per pixel was 249 msec. XRF spectra were also collected at the same power for 100 sec count times from a film of lotion dried on polypropylene and from a film blank.

The sample was imaged with the EDAX instrument using a 1000 μA source current. The following elemental relative intensity images were collected: Na, Mg, Si, S, Cl, K, Ca, Ti, Mn, Fe, Cu, and Rh (source scatter). Each elemental image consisted of 256×200 pixels. The intensity collection time per pixel was 200 msec.

A small section of the fingerprint was imaged with the IR microscope in the total absorbance relative intensity mode. An image corresponding to the relative intensity of the 2842 cm^{-1} IR peak was produced.

Saliva

After washing the hands with soap and tap water, subject #2 applied saliva to a finger by licking it and made a fingerprint on polypropylene. The sample was imaged with the KeveX MXRF instrument using source power settings of 15 kV and 3 mA. The following elemental relative intensity images were collected: Na, Mg, Al, Si, P, Cl, K, Ca, Fe, and Mo (source scatter). Each elemental image corresponded to a $24.6\text{ mm} \times 24.6\text{ mm}$ region of the sample. The elemental intensity collection time per pixel was 906 msec.

Banana

Subject #2 touched the moist interior of a banana after washing the hands with soap and tap water and left a fingerprint on polypropylene. The sample was imaged with the KeveX MXRF instrument using source power settings of 21 kV and 3.1 mA. The following elemental relative intensity images were collected: Na, Mg, Al, Si, P, Cl, K, Ca, Fe, and Mo (source scatter). Each elemental image corresponded to a $25.9\text{ mm} \times 25.9\text{ mm}$ region of the sample. The elemental intensity collection time per pixel was 1.19 sec.

Sunscreen on Black Substrate

A piece of black paper was cut from a two-pocket-type presentation folder. Analysis of this paper by XRF indicated that it contained only low levels of Al, Si, Ca, and K. After washing the hands with soap and tap water, subject #2 applied Neutrogena (Los Angeles, CA) sensitive skin sunscreen SPF 30 to the hands and rubbed it in until the white color could not be seen. This sunscreen contained 9.1% TiO_2 . A fingerprint was then made on the black paper.

The sample was imaged with the KeveX MXRF instrument using source power settings of 21 kV and 3.1 mA and in air instead of helium. Titanium and molybdenum relative intensity images were collected. Each elemental image corresponded to a $17.6\text{ mm} \times 17.6\text{ mm}$ region of the sample. The elemental intensity collection time per pixel was 805 msec.

Fingerprint Longevity

The fingerprint prepared by applying Eucerin hand lotion was analyzed a second time with the KeveX instrument ~ 7.5 months after it was first prepared and analyzed to determine if it could still be detected by MXRF after aging. The sample was imaged using source power settings of 21 kV and 3.1 mA. The following elemental relative intensity images were collected: Na, Mg, Al, Si, P, Cl, K, Ca, Fe, and Mo (source scatter). Each elemental image corresponded to an $18.1\text{ mm} \times 18.1\text{ mm}$ region of the sample. The elemental intensity collection time per pixel was 828 msec.

Results and Discussion

This work demonstrates the feasibility of analyzing fingerprints by MXRF. There are also limitations to detecting fingerprints by

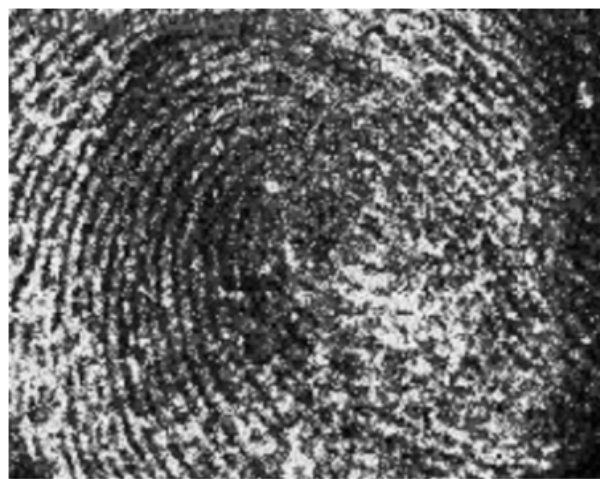
this method. One is that knowledge of the general location of a print would be required in order to examine the sample with the MXRF instrument. However, if a faint or partial print could be located by conventional powder or chemical development, MXRF could be used to scan the location of interest in an attempt to generate a complete print, provided that the conventional visualization treatments did not completely remove the MXRF mappable elements. Additionally, *a priori* knowledge about possible print locations might be available. For example, commonly handled items such as a firearm handle or a book of matches would be logical choices for examination. Alternatively, a large sample area could be imaged quickly at low resolution to identify a promising region, and slower, high-resolution images could be acquired from any promising regions.

The other limitation would be the sample size. If it were too large to fit into the spectrometer, then either a smaller specimen would need to be extracted from the original sample, or a print could potentially be lifted with tape and analyzed through the tape. The KeveX system used in this work can accommodate fairly large specimens up to approximately $15\text{ cm} \times 15\text{ cm}$, and a custom-built instrument could be made to house much larger specimens. With method development a portable instrument might be a possibility for examining any surface in the field.

Sebaceous Fingerprints

Fingerprint friction ridges were evident in the chlorine image acquired from the middle portion of the fingerprint made by subject #1 (Fig. 1). Less-pronounced friction ridges were also detected in the potassium and sulfur maps. Multiple sebaceous fingerprints were prepared by subject #2 both with and without touching the forehead, but none were detectable by either MXRF instrument. Therefore, the salt content by which regular, oily prints are identified with this technique may be person and/or diet dependent.

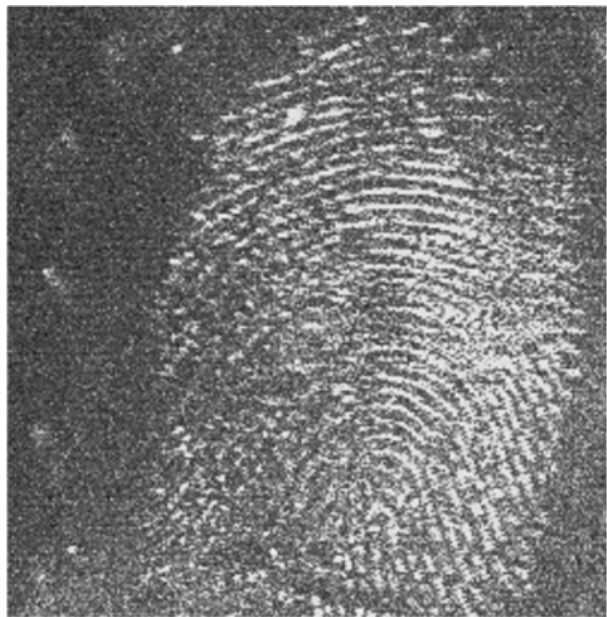
Use of a high-flux synchrotron X-ray source would be a possible means of improving the MXRF signal to noise enough to image any person's sebaceous fingerprint based on the trace amount of salts present. However, a laboratory-based instrument would be the only practical approach for routine forensics work.



0.25 cm

FIG. 1—Micro-X-ray fluorescence chlorine $K-L_{2,3}$ relative intensity image acquired from the middle portion of a sebaceous fingerprint left on polypropylene.

To improve fingerprint image detail obtained with a laboratory-type instrument, the signal to noise could potentially be amplified with effective image-processing software.



0.25 cm

FIG. 2—Micro-X-ray fluorescence potassium $K-L_{2,3}$ relative intensity image acquired from a fingerprint left on polypropylene using perspiration.

Fingerprints after Perspiring

Some real-world prints will contain significant levels of salts from sweat. Prints made after perspiring were therefore tested. Friction ridges were detected in the potassium (Fig. 2) and chlorine images, and subtle ridges were seen in the silicon and source molybdenum scatter images. The chlorine and potassium likely originated from salts present in sweat. Due to poor instrumental detection limits for sodium, only an outline of the print was observed in the sodium map.

Lotion

Many people use hand lotion. Thus, lotion-containing fingerprints were examined by MXRF. Figure 3 shows a visible image of the sample and the MXRF elemental relative intensity images for potassium, chlorine, silicon, calcium, and aluminum collected with the Kevex instrument. Pronounced fingerprints were found in the potassium and chlorine maps, indicating that significant levels of these elements were present in the print material. Friction ridges were very apparent, and other minutiae were easily evident such as ridge terminations and bifurcations.

A spectrum collected from a film of dried lotion only indicated the presence of low levels of magnesium and aluminum; no potassium or chlorine was detected in the lotion. This seems to indicate that the lotion served as a carrier to transfer efficiently any trace amounts of salts present on the finger (or in the outer skin layers) to the substrate. Also, any other residue present on the finger would be transferred by the lotion. Not all lotion prints made by the same subject (on different days) were detectable. Therefore, the person's diet and/or amount of sweat present on the

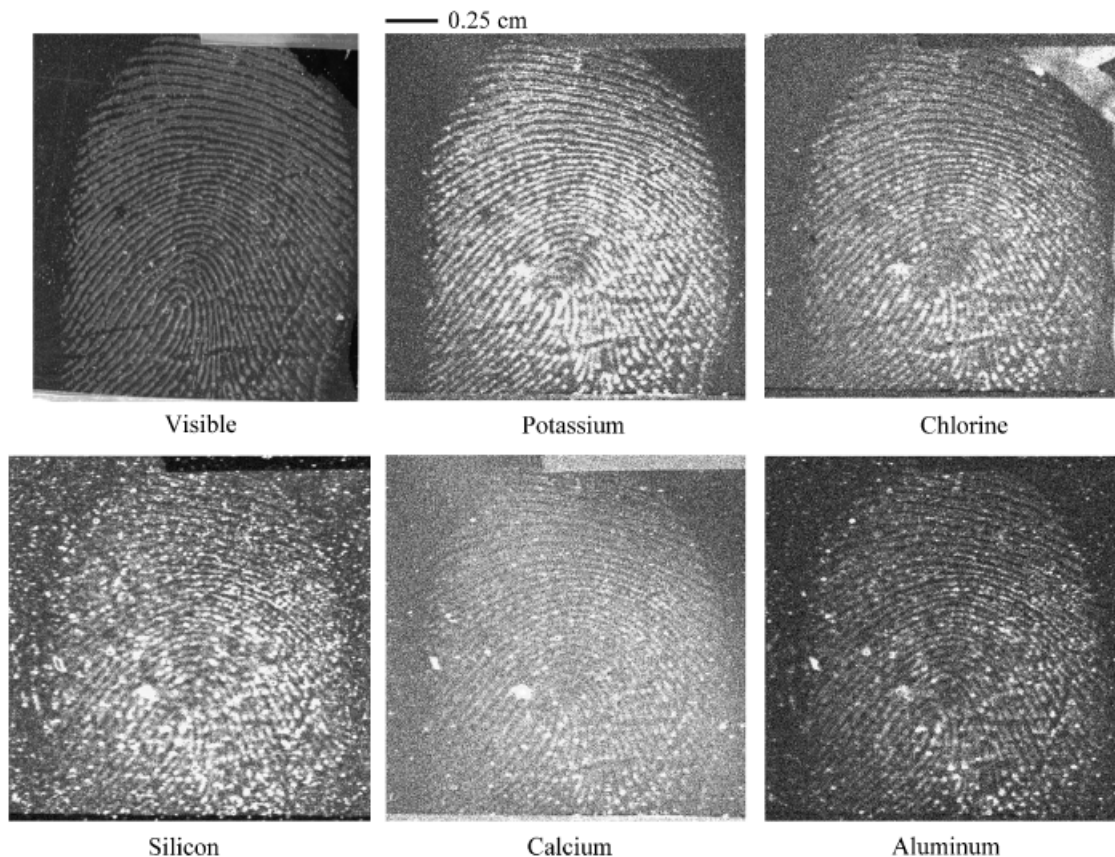


FIG. 3—Visible image and the micro-X-ray fluorescence $K-L_{2,3}$ relative intensity images for potassium, chlorine, silicon, calcium, and aluminum collected from the fingerprint made on polypropylene after applying Eucerin lotion to unwashed hands.

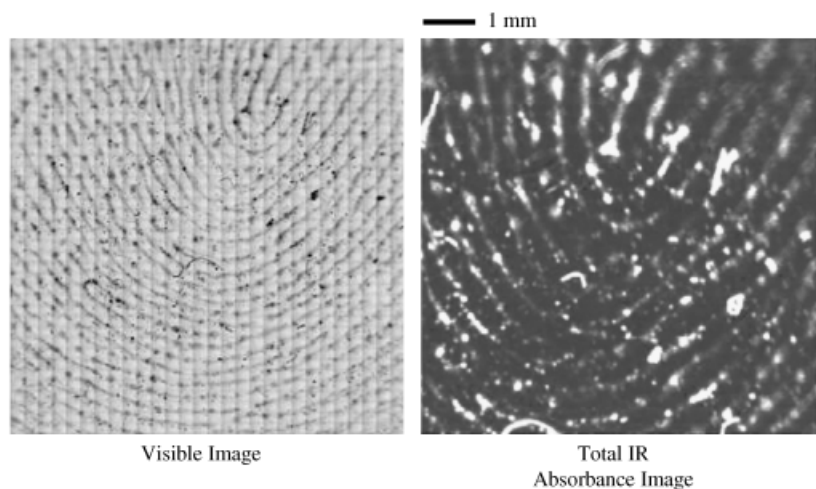


FIG. 4—Visible image and total infrared absorbance image acquired from a small section of the Eucerin lotion fingerprint prepared on polypropylene with unwashed hands.

fingers are probably critical in determining whether the print will be detected by MXRF.

As seen with the Kevex instrument, analysis of the sample with the EDAX instrument revealed a prominent fingerprint image in the potassium map. The sulfur map also showed pronounced fingerprint detail. Also, analysis of a small portion of the print using the IR microscope for both total absorbance and the 2842 cm^{-1} peak intensity (C–H stretch) revealed similar features; definitive friction ridges were detected as depicted in the total IR absorbance image (Fig. 4). IR microscopy could also be a very powerful method for detecting fingerprints and could complement MXRF results. In certain cases where fingerprints are not evident using MXRF, they might be identifiable by IR microscopy. One IR requirement is that the specimen must be at least partially transparent or reflective, but MXRF is not constrained by this particular limitation.

Saliva

Since young children often stick their fingers in their mouth, the ability to identify fingerprints based solely on saliva residue content would be valuable in missing child cases. Here, a subject's fingerprint was detected from the potassium (Fig. 5), chlorine, and phosphorus present in saliva transferred to the substrate. Several regions of excess saliva material were present in the print that hindered the identification of all of the friction ridges, but even partial ridge information might be useful. Ideally, if the saliva were transferred to the surface more uniformly, a complete fingerprint would be observed by MXRF. Multiple specimens were not studied in this preliminary work. However, preparing several prints from a licked finger would remove excess saliva initially and likely provide at least one specimen containing a more ideal amount of material, as would be the case if a child touched a surface multiple times.

Banana

Young children often have food on their fingers after eating, so MXRF might be capable of discerning a fingerprint based on elements present in food residue left by a child touching an object. Since bananas contain significant levels of potassium, a fingerprint was made after touching the moist interior of a banana. The sample consisted of some aggregates of banana residue that masked a portion of the print ridges in the potassium image (Fig. 6), but ridge detail could be detected in other regions. Similar

aggregates were observed in the sodium, magnesium, phosphorus, chlorine, and molybdenum maps, but no ridge detail was present. The bright patch on the lower left corner of the potassium image was due to source scatter from the side of the plastic cup used to mount this particular sample film. As was mentioned with the saliva print, touching a surface multiple times with the same finger would sometimes provide a print containing a more ideal amount of residue for detecting the entire print. Even locating partial friction ridges could be helpful in forensic cases.

Sunscreen on Black Substrate

As with the other substances applied to the hands for study, sunscreen is a fairly common material that could potentially be found in fingerprint residue. A fingerprint consisting of sunscreen with an organic active agent such as para-aminobenzoic acid (PABA) is not detectable by XRF, but identifying sunscreen prints containing titanium or zinc oxide is straightforward.

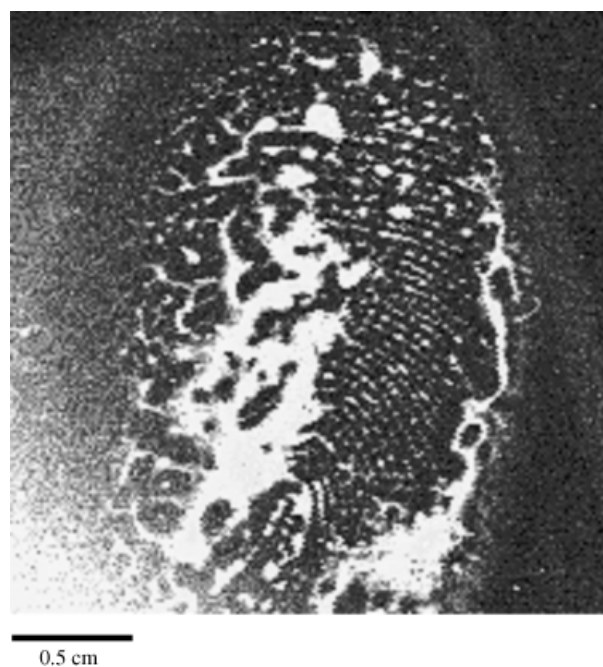


FIG. 5—Micro-X-ray fluorescence potassium $K-L_{2,3}$ relative intensity image from a saliva-containing fingerprint prepared on polypropylene.

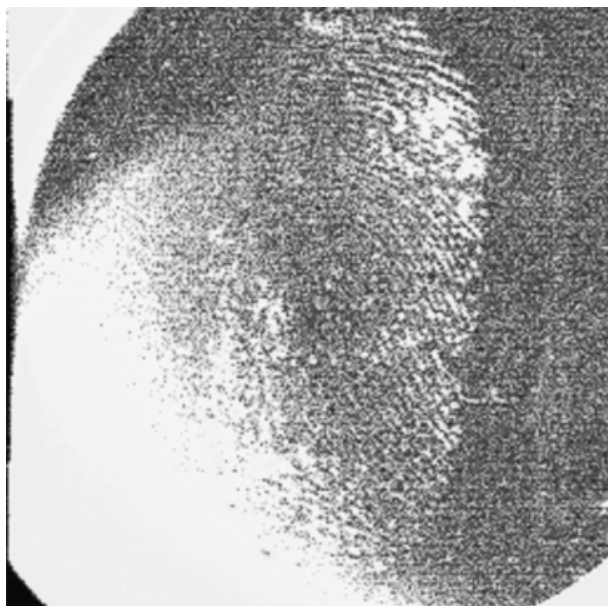


FIG. 6—Micro-X-ray fluorescence potassium $K-L_{2,3}$ relative intensity image from a fingerprint left on polypropylene after touching the interior of a banana.

Figure 7A shows a picture of the sample. Only a black ink mark outlining the print location was evident; the fingerprint was invisible. The MXRF titanium image (Fig. 7B) revealed a very prominent fingerprint with discernible friction ridges and other minutiae (ridge terminations, bifurcations, etc.).

The fingerprint sample was prepared on black paper to demonstrate a potentially challenging real-world case where the substrate was porous and black in color (in addition to the print consisting mainly of sunscreen). Cyanoacrylate fuming will provide a white-colored print (22,23), so a fingerprint located on a black surface would be revealed if fuming were effective for a particular specimen. However, cyanoacrylate works best on smooth surfaces rather than porous specimens (24,25). For fingerprints on porous surfaces such as the paper studied here, MXRF would provide an alternative means to image the print.

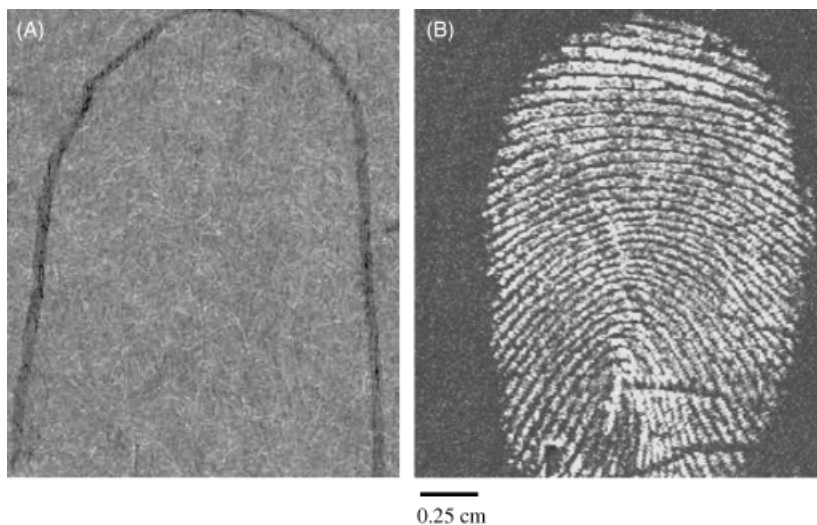


FIG. 7—Picture of a latent fingerprint specimen on black paper (A) left after applying sunscreen to the hand, and the micro-X-ray fluorescence titanium $K-L_{2,3}$ relative intensity image (B).

Fingerprint Longevity

Because nonvolatile elements such as potassium are detectable by MXRF, a fingerprint should not degrade over time when examined by MXRF, which is not always the case with contrast-enhancing techniques (1). After aging for ~ 7.5 months, the fingerprint composed of hand lotion was still readily apparent in the potassium (Fig. 8A) and chlorine maps. The potassium image was not identical to the image collected 7.5 months earlier (Fig. 8B), but the X-ray source was replaced before the second analysis was performed, and the capillary optic was realigned less successfully prior to the second analysis, resulting in a lower X-ray flux irradiating the sample. However, successful detection of the fingerprint the second time proved that a significant amount of potassium was still present. In theory, this fingerprint should be detectable indefinitely if left undisturbed. Therefore, MXRF could allow one to identify latent prints many years old if they contain significant levels of body salts or other inorganic residue.

Future Directions

Future plans include studying a larger pool of subjects. Determining whether certain people consistently generate prints detected by MXRF is one area deserving further study. Also, we will determine whether there is a correlation between a person's diet and the ability to detect their fingerprints. Another area of study will be fingerprints made after handling various types of food. This will be especially pertinent in determining the viability of identifying young children's prints by MXRF. Aging effects with exposure to sunlight and ambient light, heat, and humidity will also be inspected. Finally, examination of fingerprints left on a variety of materials will be pursued, and efforts are underway to study prints using XRF-detectable inorganic powders and chemical developers. Even if a print cannot be visualized by MXRF, treating it with a physical developer (e.g., silver) should create a print that can be detected.

Conclusion

The potential of MXRF for detecting both visible and latent fingerprints has been demonstrated. While MXRF would not replace established contrast enhancement methods, there are a number of niche areas where it could provide data that comple-

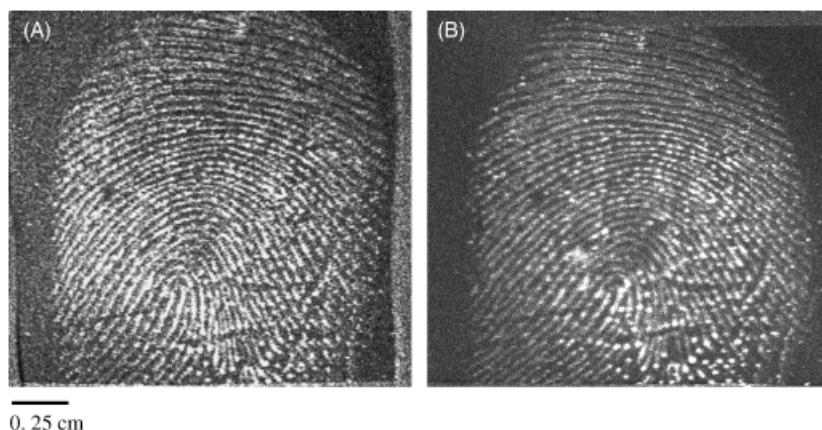


FIG. 8—Micro-X-ray fluorescence (MXRF) potassium $K\text{-}L_{2,3}$ relative intensity image from the fingerprint made on polypropylene after applying Eucerin lotion to unwashed hands and acquired ~ 7.5 months after the specimen was prepared (A), and the original MXRF image (B) collected shortly after the specimen was originally made.

ment that acquired using traditional methods. For example, a print left from a finger coated with a residue such as gunpowder might be detectable from the sulfur and potassium content. Other distinctive fingerprint examples such as those containing lotion, sunscreen, saliva, or certain food residue can be detected by MXRF based on the inorganic elemental constituents.

While this method currently requires *a priori* knowledge of the general location of a print, it clearly has some advantages over contrast-based techniques for special cases. Because it is a spectroscopic method, the elemental composition of a fingerprint is examined, and visual contrast with the substrate is irrelevant. Thus, if sufficient detectable residue is present, the print can be identified regardless of the background color. It is also a noninvasive tool, so a sample remains intact for other analyses or archiving. Volatilization of water, oils, and other organic components over time should not hinder print identification since only inorganic elements are detected. In summary, MXRF offers a novel means to image fingerprints that should complement the current list of tools available to forensic investigators.

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