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The Recovery of Latent Fingermarks from Evidence Exposed to Ionizing Radiation*

ABSTRACT: Continual reports of illicit trafficking incidents involving radioactive materials have prompted authorities to consider the likelihood of forensic evidence being exposed to radiation. In this study, we investigated the ability to recover latent fingermark evidence from a variety of substrates that were exposed to ionizing radiation. Fingermarks deposited on common surfaces, including aluminum, glass, office paper, and plastic, were exposed to doses ranging from 1 to 1000 kGy, in an effort to simulate realistic situations where evidence is exposed to significant doses of radiation from sources used in a criminal act. The fingermarks were processed using routine fingermark detection techniques. With the exception of glass and aluminum substrates, radiolysis had a considerable effect on the quality of the developed fingermarks. The damage to ridge characteristics can, in part, be attributed to chemical interactions between the substrate and the components of the fingermark secretions that react with the detection reagents.

KEYWORDS: forensic science, radiological forensics, latent fingermarks, fingermark detection techniques, ionizing radiation, chemical, biological, radiological, and nuclear forensics

Incidents of illicit trafficking of radioactive and nuclear materials have been occurring for some time (1,2). In fact, from January 1993 to December 2007, there have been 1340 confirmed incidents of nuclear materials trafficking registered on the International Atomic Energy Agency's (IAEA) illicit trafficking database (3). The IAEA believes that the number of reported incidents represents a conservative estimate. Furthermore, there are growing concerns that more sophisticated and organized trafficking of nuclear material may be occurring undetected, illustrating the potential threat of uncontrolled radioactive sources falling into the wrong hands (3–5).

Given that the trafficking and illegal possession of radioactive and/or nuclear materials constitutes a breach of state or national law, a thorough criminal investigation into the circumstances of this situation is warranted (6,7). It is also extremely important for authorities, in the context of the criminal investigation, to determine if the seizures are linked (hence providing the first suspicion of an organized and potentially malicious activity) and to assist local authorities to prevent any further theft or diversion of materials from these sources (8–10). Knowing the hazardous nature of the material, it is conceivable that any physical evidence recovered from the crime scene may have been exposed to ionizing radiation, and that any subsequent radiation-induced damage could potentially impact on the quality of the information obtained and/or its interpretation. The main question is whether or not physical evidence can be compromised due to exposure to radiation. And, if yes, up to which dose such evidence may still be exploitable.

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The effects of ionizing radiation on a diverse range of materials have been documented in numerous studies. The results from these studies suggest that the altered physical properties are mainly attributed to the destruction of functional groups such as carbonyl and carboxyl groups, as well as main chain incision and cross-linking of molecules (11,12). Radiation-absorbed doses of up to 50 kGy have induced irreversible changes to the physiochemical and mechanical properties of materials such as polymer-based packaging materials, inks on documents, and archival materials (13–15).

In a recent study into the effects of electron beam radiation on forensic evidence, latent marks on porous and nonporous substrates were exposed to doses of up to 30 kGy. The results indicated that the recovery of latent marks can be significantly compromised by ionizing radiation (16).

Given that the constituents of a latent fingermark are a mixture of organic and inorganic compounds (including water, salts, metal ions, proteins, amino acids, glycerides, fatty acids, and esters) (17,18), it is reasonable to assume that radiolysis (radiation damage mechanisms) of some of the organic constituents and elevated temperatures (from the electron irradiation process) contribute to the accelerated degradation of the latent marks and/or the substrate, and therefore their subsequent recovery (16).

The aim of this study was to assess the impact of ionizing radiation on the detection and enhancement of latent fingermarks on various surfaces. While there have been several studies conducted, examining the effects of ionizing radiation on trace evidence, most of the work has focused on electron beam irradiation processes (commonly used to sanitize mail). The focus of this study was to assess trace evidence (latent fingermarks) inadvertently exposed to radioactive materials found at a crime scene. As such, latent fingermarks were exposed to significantly higher exposures (compared with electron beam irradiators) to simulate a range of doses most likely to be encountered from readily available industrial radioisotopes. The applicability of current latent fingermark enhancement techniques was assessed using several different donors and substrates.

TABLE 1—*The properties and nominal activity ranges for radioactive sources readily available in Australia.*

Source	Half Life ($T_{1/2}$)	*Gamma Constant (Γ)	Activity Range (GBq†)	Common Use
Cesium-137	30.1 years	103 μSv/h/GBq @ 1 m	37–81,400	Teletherapy, blood irradiations, and sterilization facilities
Iridium-192	74 days	160 μSv/h/GBq @ 1 m	1200 or 2700	Industrial radiography and low dose brachytherapy
Cobalt-60	5.3 years	370 μSv/h/GBq @ 1 m	27–185,000	Teletherapy, industrial radiography, and sterilization facilities

*Australian Standards 2243 (Parts 1–10) Safety in Laboratories AS 2243.4–1998 Ionizing radiations.
†1GBq equals 1,000,000,000 Becquerel.

Methods and Materials

Irradiation Parameters

In order to simulate realistic radiation exposures that forensic evidence might be subjected to, in a variety of hypothetical scenarios, several key assumptions were established. These included the proximity of the evidence to the source, the exposure time, and the activity range for certain radioactive sources.

The premise for this research was that the latent fingermarks would be situated in close proximity (i.e., 1, 10, and 100 mm) from an unshielded source for a notional time period ranging from 24 h to 7 days. The third and most important parameter identified a number of radioactive sources, commonly available in Australia, that pose the greatest security risk (19,20). These sources and their nominal activity ranges are identified in Table 1.

The *a priori* absorbed dose values for the forensic samples calculated using the described parameters, ranged from 6.7 Gy to *c.* 11 MGy. However, careful analysis of the data suggested that the irradiation dose values ranging from 1 to 1000 kGy satisfactorily (and realistically) covered the majority of situations. It was determined that if the onset of radiation damage was not evident at 1000 kGy, then the likelihood of damage occurring would be negligible.

A MDS Nordion GammaCell (Model 220 Excel; Ontario, Canada) cobalt-60 research irradiator with a nominal activity of 670 TBq (*c.* 18,000 Curie) was used to simulate *in situ* radiation exposure to the latent fingermarks. The irradiation exposure times (ranging from minutes to days) which were required to attain the appropriate doses of 1, 5, 10, 50, 100, 500, and 1000 kGy were calculated via the use of simple mathematical relationships (21). Irradiation doses were measured and extrapolated using Harwell Perspex PMMA (Batch: 3042s; Oxfordshire, UK) and Ceric-cerous (batch CCT; Lucas Heights, Australia) dosimeters. The ambient temperature in the irradiation chamber was *c.* 37°C.

Latent Fingerprint Deposition and Development

Latent fingermarks from five predetermined "good" donors were deposited on a range of porous and nonporous substrates. These substrates included borosilicate glass (Livingstone™ premium microscope slides, Sydney, Australia), hard plastic (Aptaca™ polystyrene petri dishes, Sydney, Australia), soft plastic (Livingstone™ polyethylene resealable zip lock bags), metal (aluminum sheet), and paper (lined note paper and 80 gsm Reflex™ marker paper, Melbourne, Australia). Eight freshly charged donor marks were carefully deposited on each substrate—that is, a control fingermark (nil exposure) and a mark for each predetermined absorbed radiation dose juncture.

All the reagents and techniques used to develop the latent fingermarks from the various substrates were standard methodologies employed by the Forensic & Technical Laboratory of the Australian Federal Police (22). These are listed in Table 2. Specific details on the reagent formulations can be readily obtained from open literature (23).

TABLE 2—*Reagents and techniques used to process latent fingermarks on the various substrates.*

Reagent/Technique	Glass	Plastic	Plastic	Aluminum	Paper
Cyanoacrylate ester/ Rhodamine 6G	✔	✔	✔	✔	–
Cyanoacrylate ester/ Basic Yellow 40	✔	✔	✔	✔	–
Black powder (nonmagnetic)	✔	✔	✔	✔	–
Ninhydrin	–	–	–	–	✔
1,8-Diazafluoren-9-one	–	–	–	–	✔
1,2-Indanedione	–	–	–	–	✔
Physical Developer	–	–	–	–	✔

Images of developed fingermarks were recorded using a Nikon™ D70 digital SLR camera (Tokyo, Japan) in conjunction with a Tamron™ SP AF Di 90 mm macro lens (Saitama, Japan). General details of the processes used to develop and record the fingermarks on the different substrates are provided below.

Nonporous Surfaces

Cyanoacrylate/Rhodamine 6G/Basic Yellow 40—Cyanoacrylate (CA) fuming was applied to the glass, hard and soft plastics, and the metal substrates. Approximately 20 drops of Loctite® Hard Evidence™ fingermark developer (#17609; Lightning Powder Co. Inc., Jacksonville, FL) was evaporated for 8 min at 120°C. The fingermarks were fumed for 75 min using a Forensic Cyanoacrylate Cabinet (FCC #171; Carter-Scott Design, Melbourne, Australia) and then left overnight before staining.

The CA-treated fingermarks were placed in either a 150 mL bath of Rhodamine 6G (Lightning Powder Co. Inc.) stain solution or a 150 mL bath of Basic Yellow 40 (#12143; Lightning Powder Co. Inc.) stain solution for *c.* 20 sec, removed, and allowed to air-dry for 30 min. The fingermarks were visualized in the luminescence mode using a Polilight (Model PL500; Rofin Australia, Melbourne, Australia) set to the specific wavelength of 505 nm for Rhodamine 6G and 450 nm for Basic Yellow 40, with observation using an orange 590 nm long-pass barrier filter (Hoya, Tokyo, Japan).

Black Powder—Conventional nonmagnetic black powder (Lightning Powder Co. Inc.) was carefully applied to the glass, hard and soft plastics, and the metal substrates using a glass fiber Zephyr® brush (Lightning Powder Co. Inc.). Developed fingermarks were imaged with illumination from a Polilight (Rofin Australia) set to the white light mode.

Porous Surfaces

Ninhydrin—Latent fingermarks on paper were processed by immersion in a 50 mL bath of ninhydrin reagent (Sigma Aldrich Chemicals, Sydney, Australia) for *c.* 30 sec. The treated substrates

were then allowed to air-dry for 2 h. Fingermarks were left to develop for 2 h at room temperature and then imaged with illumination from a Polilight (model PL500; Rofin Australia) set to the white light mode.

1,8-Diazafluoren-9-one—Latent marks on the paper substrates were processed by immersion in a 50 mL bath of 1,8-diazafluoren-9-one (DFO; Lightning Powder Co. Inc.) reagent for *c.* 30 sec. The treated substrates were then allowed to air-dry for 15 min and finally placed in a heat press (Singer™ Magic Steam Press, Sydney, Australia) at a preset temperature of 165°C for 10 sec. Fingermarks were visualized in luminescence mode using a Polilight set to the specific wavelength of 530 nm and imaged using an orange 590 nm long-pass barrier filter (Hoya).

1,2-Indanedione—1,2-Indanedione is a more sensitive reagent than ninhydrin for fingermark detection on porous substrates. The working solution used in this study consisted of 0.5 g 1,2-indanedione (Casali Institute, Jerusalem, Israel), 17.5 mL of ethyl acetate (Chem-Supply, Port Adelaide, Australia), 233.5 mL HFE-7100 (3M™, St. Paul, MN), and 2 mL of acetic acid (Chem-Supply). In a similar method to DFO and ninhydrin treatment, latent marks on paper were processed by immersion in a 50 mL bath of indanedione reagent for *c.* 30 sec. The treated substrates were then allowed to air-dry for 15 min and finally placed in a heat press (Singer™ Elna Magic Steam Press) at a preset temperature of 165°C for 10 sec. Fingermarks were visualized and recorded in luminescence mode using the same conditions as described for DFO.

Physical Developer

The fingermarks deposited on paper were developed using this multistep process. Preliminary steps required the marks to be soaked in distilled water for *c.* 10 min followed by a bath containing maleic acid (CHEM Supply) for 5 min, then placed into a second bath of distilled water for *c.* 10 min. The prepared marks were then placed into a working solution of physical developer (consisting of three components: surfactants, redox solution, and silver nitrate) for 10–15 min (or until there was sufficient ridge detail and contrast with the substrate), removed, and rinsed in distilled water for *c.* 15 min. The air-dried developed fingermarks were imaged with illumination from a Polilight (Rofin Australia) set to the white light mode.

Results and Discussion

Glass

The effects of increasing radiation exposure on the optical attributes for borosilicate glass are clearly seen in Fig. 1. The onset of radiation damage (discoloration) was clearly visible as low as 1 kGy and was consistent with observations described in the literature (12). Irradiating glass containing high percentages of alkaline oxides (>15%) will result in visible absorption peaks (400–600 nm) as a consequence of stable color centers being formed. The color variation (ranging from clear to deep brown) in no way affected the physical properties of the glass and did not result in adverse interaction with the fingermark deposit or the detection techniques. However, it should be noted that substrate discoloration could interfere with standard contrast imaging techniques that rely on the reflection/absorption of light.

Following the CA fuming process, the latent marks on the glass substrates were enhanced using Basic Yellow 40 and Rhodamine 6G luminescent stains. A third set of latent marks was processed using conventional nonmagnetic black powder. Results from all three sets of developed fingermarks suggest that ionizing radiation had little or no effect on the latent marks on this substrate.

Figure 2*a–d* represents the control (0 kGy) and the 1000 kGy exposed fingermarks developed by CA fuming and stained with

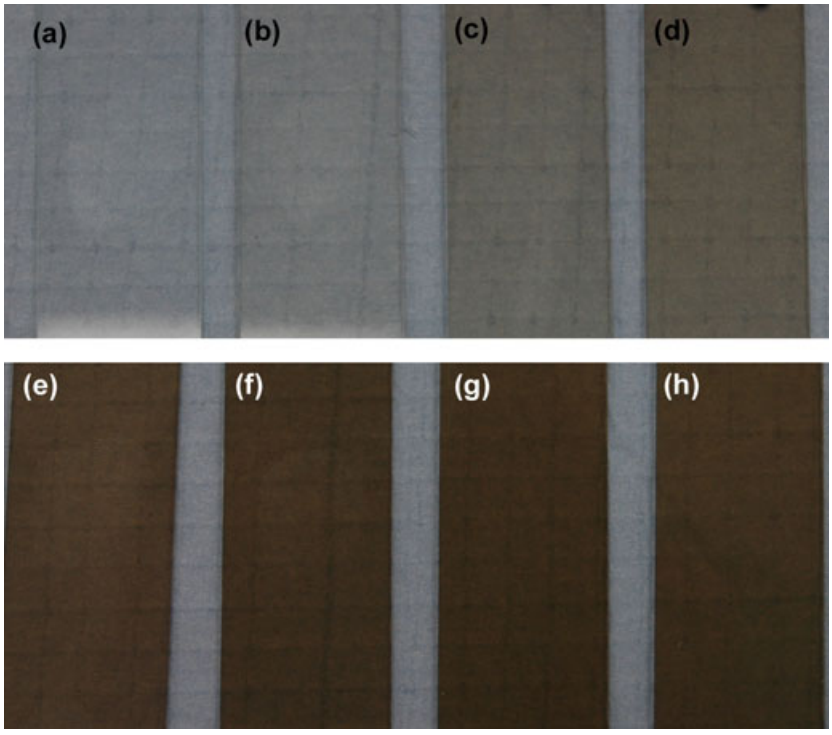

FIG. 1—*The optical degradation (browning) of the borosilicate glass after (a) 0 kGy (control); (b) 1 kGy; (c) 5 kGy; (d) 10 kGy; (e) 50 kGy; (f) 100 kGy; (g) 500 kGy; and (h) 1000 kGy radiation dose.*

Basic Yellow 40 and Rhodamine 6G reagent stains, respectively. The CA developed fingermarks enhanced with Basic Yellow 40 after 1000 kGy radiation exposure exhibited reduced luminescence compared with the control mark though the ridge characteristics still remained distinguishable.

The Rhodamine 6G enhanced fingermarks showed excellent ridge detail even after a 1000 kGy radiation exposure. Furthermore, the ridge-to-substrate contrast in the fingermarks was slightly improved due to the altered optical properties (browning) of the glass substrate. The luminescence intensity along the fingermark ridges was also consistent, suggesting that the CA monomer (CA fuming) was still effective in polymerizing with components of the latent fingermark deposit, and the subsequent binding of the luminescent stain was not inhibited.

Processing using conventional nonmagnetic black powder (Figs. 2*e* and 2*f*) further suggests little or no effects of ionizing radiation on the latent fingermarks. Even after a radiation dose of 1000 kGy, ridge definition was preserved. The optical degradation of the borosilicate glass led to diminished ridge-to-substrate contrast, particularly at higher doses. However, improved visualization techniques and choice of powder can readily overcome this deficiency.

Aluminum

The latent fingermarks on the aluminum substrate were processed similarly to the marks deposited on glass. Following CA fuming, the marks were enhanced using Basic Yellow 40 and Rhodamine 6G luminescent stains. Nonmagnetic black powder was also used as a development technique. All three enhancement techniques (Fig. 3*a–f*) produced clear ridge detail, comparable with that from the control marks, even after a radiation dose of 1000 kGy.

The luminescence intensity produced by the Basic Yellow 40 and Rhodamine 6G stains, along the fingermark ridges, was also consistent with increasing radiation exposure suggesting that there was little or no effect on the physiochemical characteristics of the fingermark secretions that would hinder the CA polymerization process, and subsequent binding of the luminescent stains. The aluminum substrate, as expected, showed no evidence of radiation damage, given that the material is often used as shielding and in radiation-hardened components in the nuclear industry, military applications, and in the space environment (12).

Polyethylene and Polystyrene

The susceptibility of linear chain molecules to radiation damage was well documented. At high doses, polyethylene (PE) and polystyrene (PS) radiolytically induced rupturing of covalent bonds (predominantly C–H bonds) leads to a highly cross-linked molecule overburdened with fragmentation (24). Previous studies suggest that initial signs of radiation damage in polyolefins [linear chain molecules such as PE and polypropylene] occur in the order of 10^4–10^5 Gy (12). Severe physiochemical and mechanical damage was observed in polyolefins at doses nearing 10^6 Gy (25).

The latent fingermarks on the PE and PS substrates were processed using CA fuming and enhanced using Basic Yellow 40 and Rhodamine 6G luminescent stains. A third set of latent marks was processed using nonmagnetic black powder.

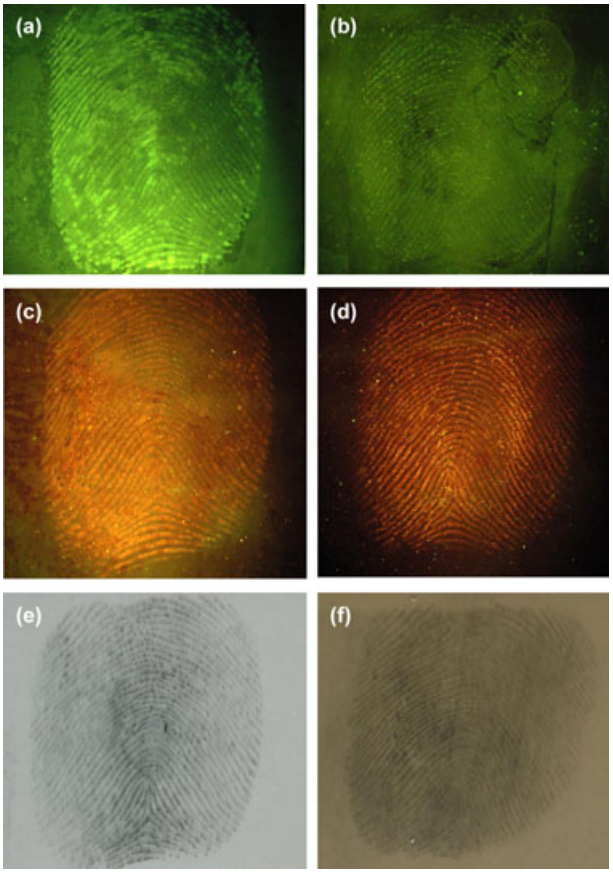

FIG. 2—*Latent fingermarks on glass developed by cyanoacrylate fuming and enhanced with (a) Basic Yellow 40 (control) and (b) after 1000 kGy exposure; (c) Rhodamine 6G and (d) after 1000 kGy exposure; (e) black powder (control); and (f) after 1000 kGy exposure.*

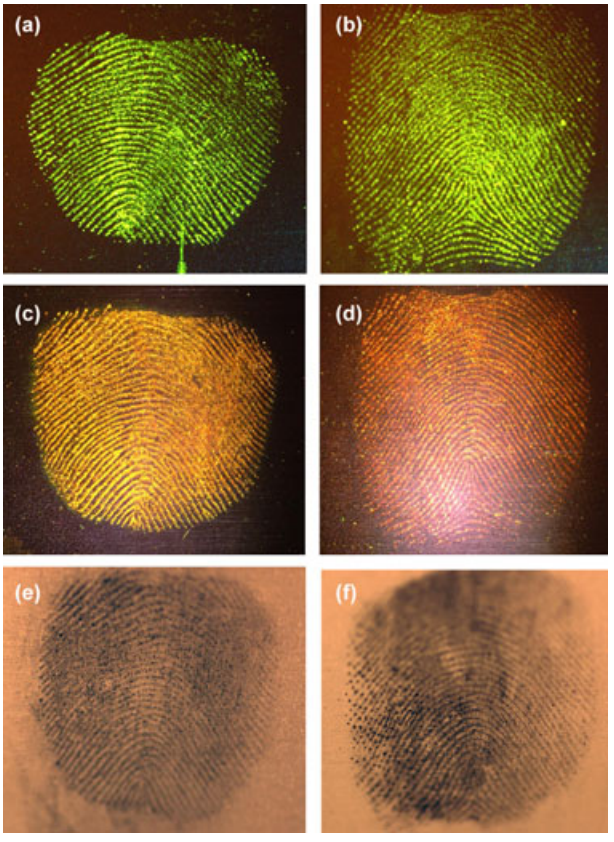

FIG. 3—*Latent fingermarks on aluminum plate developed by cyanoacrylate fuming and enhanced with (a) Basic Yellow 40 (control) and (b) after 1000 kGy exposure; (c) Rhodamine 6G and (d) after 1000 kGy exposure; and (e) black powder (control) and (f) after 1000 kGy exposure.*

Latent marks on the PE and PS substrates (Fig. 4*a–h*, respectively) processed using nonmagnetic black powder indicated a considerable effect of ionizing radiation on the ridge characteristics. Doses greater than 100 kGy showed significant degradation in the overall latent mark definition.

Figures 5*a–d* and 6*a–d* show the fingermark ridge characteristics after CA fuming and with enhancement using Basic Yellow 40 on the PE and PS substrates, respectively. The fingermark ridge features on both substrates were clearly visible up to 100 kGy, and only just visible at doses greater than 500 kGy. The luminescence intensity and emission color also appeared to follow this trend.

Similarly, Figs. 5*e–h* and 6*e–h* highlight the ridge characteristics developed on the PE and PS substrates, respectively, by CA fuming and Rhodamine 6G staining. The ridge features were still well defined after a 100 kGy dose, and only just visible at doses greater than 500 kGy.

The reduction in luminescent dye intensity produced by both reagents with increasing radiation was noticeable, and suggests that the onset of radiation damage to the polymer substrates with increasing dose has irreversibly altered their physiochemical characteristics. It is conceivable that radiolysis has produced reactive polar groups (such as carboxylic acids) at the polymer surface (26) and

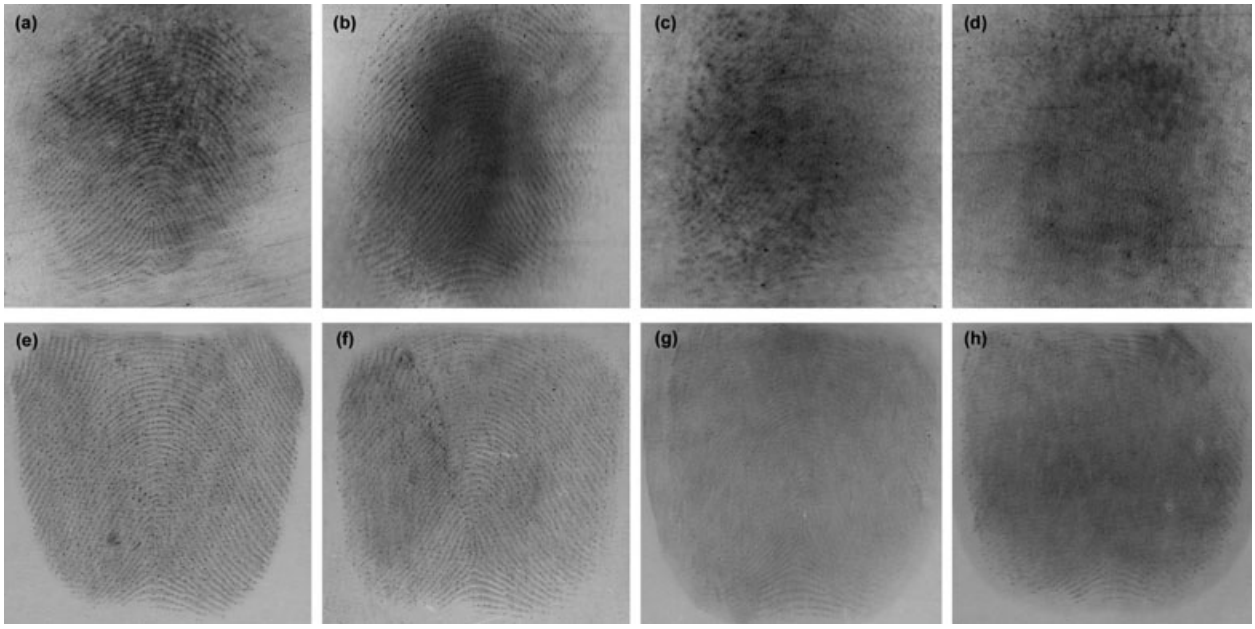

FIG. 4—*Latent fingermarks on polyethylene (a–d) and polystyrene (e–h) plastic substrates developed using cyanoacrylate fuming and enhanced with nonmagnetic black powder after 0 kGy (control) (a,e); 10 kGy (b,f); 100 kGy (c,g); and 1000 kGy exposure (d,h).*

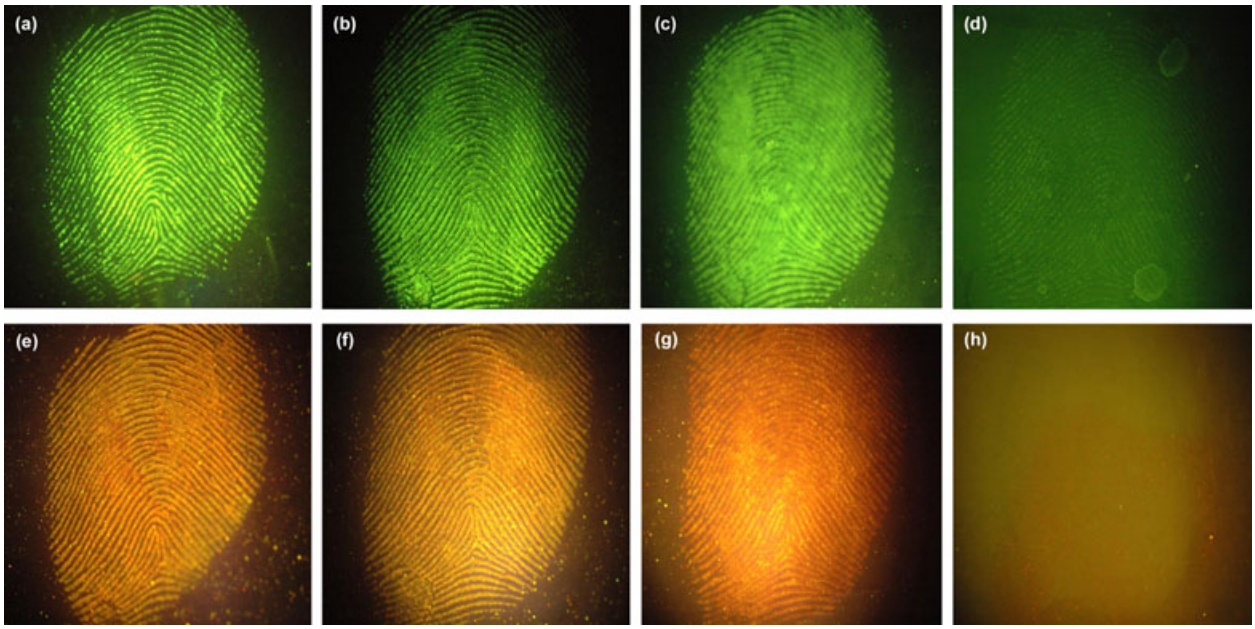

FIG. 5—*Latent fingermarks on polyethylene plastic developed using cyanoacrylate fuming and enhanced with Basic Yellow 40 at (a) 0 kGy (control); (b) 10 kGy; (c) 100 kGy; and (d) 1000 kGy exposure, and enhanced with Rhodamine 6G after (e) 0 kGy (control); (f) 10 kGy; (g) 100 kGy; and (h) 1000 kGy exposure.*

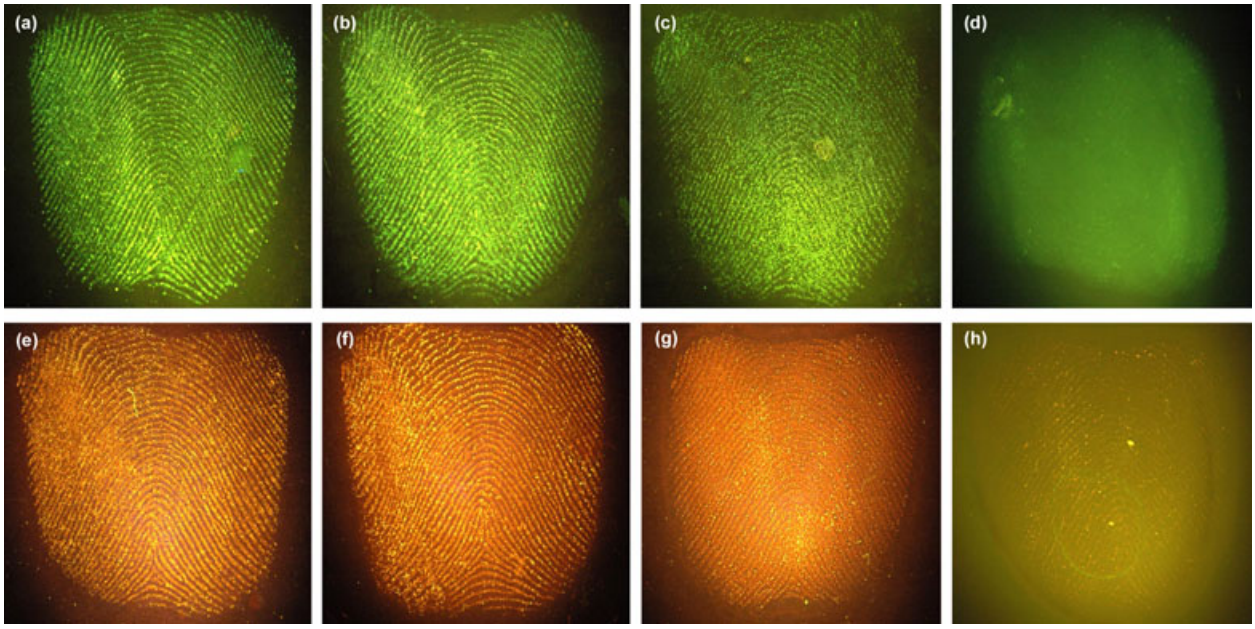

FIG. 6—*Latent fingermarks on polystyrene plastic developed using cyanoacrylate fuming and enhanced with Basic Yellow 40 at (a) 0 kGy (control); (b) 10 kGy; (c) 100 kGy; and (d) 1000 kGy exposure, and enhanced with Rhodamine 6G after (e) 0 kGy (control); (f) 10 kGy; (g) 100 kGy; and (h) 1000 kGy exposure.*

that such groups may have interacted with some of the components of the fingermark secretion, hindering the CA polymerization process and the subsequent uptake of the luminescent stains.

Paper

Cellulose, a major fibrous component in paper (either as wood pulp or as recycled paper) is radiolytically sensitive. Previous studies suggest that moderate to severe radiation damage in cellulose manifests after exposure of tens of kGy (15,25). Furthermore, organic chemical additives and dyestuffs (such as optical brighteners) introduced to improve the properties of the paper, such as whiteness and texture, may also be susceptible to radiolysis.

The latent fingermarks on paper were processed using ninhydrin, DFO, 1,2-indanedione, and physical developer. Particular care was necessary in handling and processing the paper substrate, with the mechanical properties of the paper degraded significantly with increasing dose. Discoloration in the paper samples (browning) was noticeable at doses greater than 50 kGy and like the glass substrates, this aspect had no visible consequence on the fingermark detection process.

The fingermark ridges processed using ninhydrin and physical developer reagents (see Fig. 7*a–f*) were visible and well defined up to doses of 100 kGy exposure. However, the ninhydrin and physical developer results deteriorated quite significantly when exposed to higher irradiation doses. Attempts to develop latent marks on paper in doses greater than 500 kGy failed due to the significant degradation in the mechanical properties of the paper substrate.

Similarly, Fig. 8*a–h* highlights the ridge detail developed on paper using DFO and 1,2-indanedione, respectively. The ridge features using both reagents were visible and well-defined after an irradiation exposure of 50 kGy, but with significant deterioration observed at doses greater than 100 kGy (only partial fingermark features were visible at 100 kGy).

The observations suggest that one or more radiolytic mechanisms may have directly and indirectly interacted with the fingermarks,

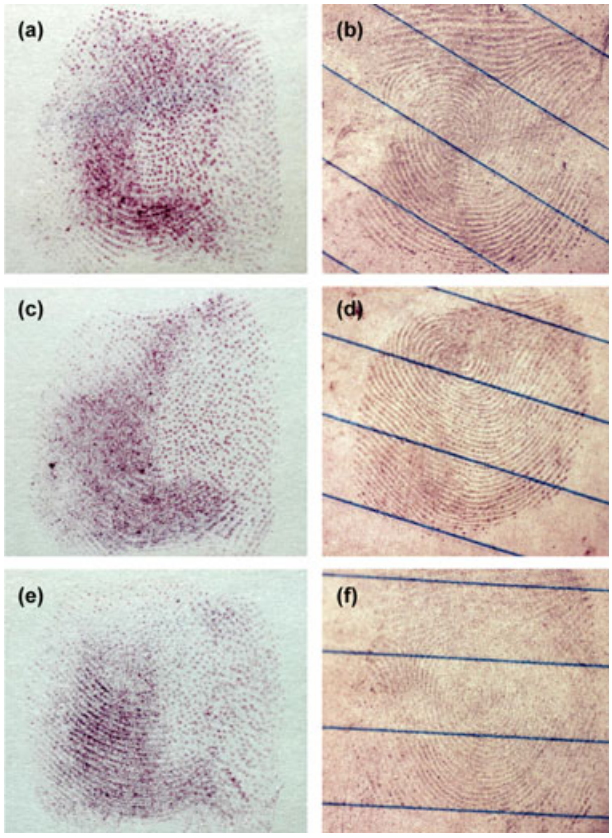

FIG. 7—*Latent fingermarks on copy paper processed with ninhydrin at (a) 0 kGy (control); (c) 10 kGy; and (e) 100 kGy exposure; and fingermarks processed on ruled folio paper with physical developer after (b) 0 kGy (control); (d) 10 kGy; and (f) 100 kGy exposure.*

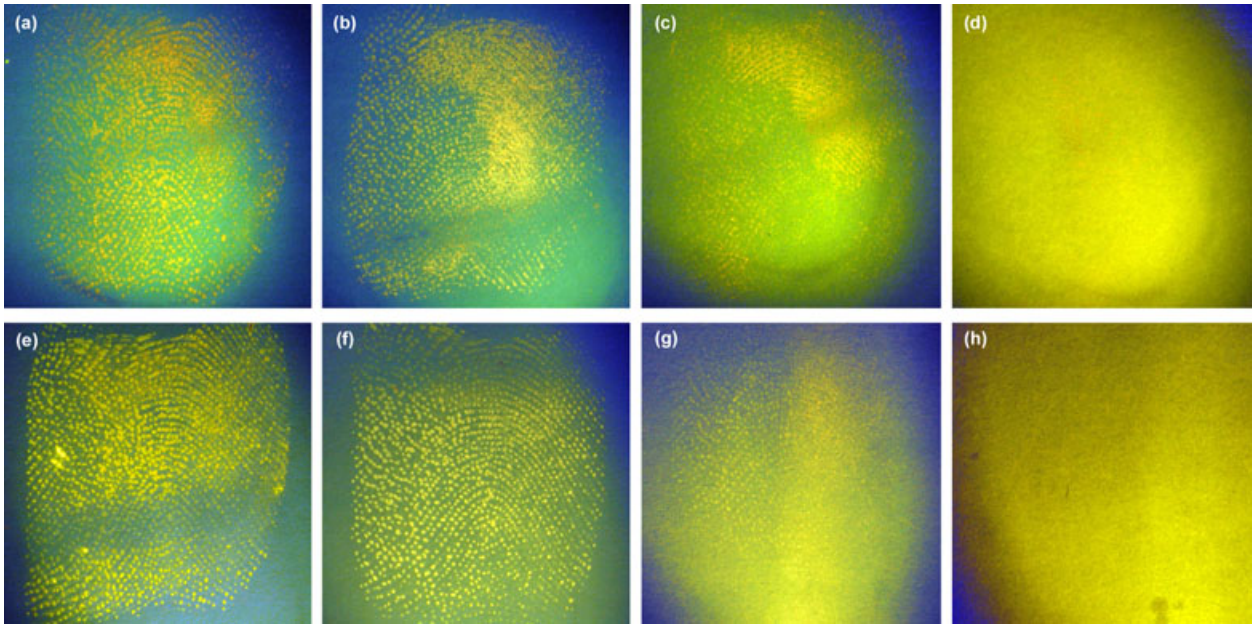

FIG. 8—*Latent fingermarks on copy paper processed with 1,8-diazafluoren-9-one (a–d) and 1,2-indanedione (e–h) and imaged with a 530 nm excitation energy and an orange 590 nm barrier filter at 0 kGy (control) (a,e); 10 kGy (b,f); 100 kGy (c,g); and 1000 kGy (d,h) exposure.*

making them less prone to detection via these amino acid reagents. It is most likely that the formation of reactive ions and or radicals has interfered with the chemistry of some of the latent fingermark compounds, hindering the reaction that normally leads to the formation of a luminescent product.

Furthermore, the effects of radiolytic degradation of the cellulose fibers and the likely formation of reactive polar groups at the surface appear to have altered the properties of the paper (26). As can be seen in Fig. 8*a–h*, where the paper substrates have been exposed to a 1000 kGy dose, both DFO and indanedione have produced a high background luminescence that obscures any fingermark ridge detail. This suggests that the chemical breakdown of the paper has resulted in the formation of amine-containing compounds that have subsequently reacted with the fingermark detection reagents.

Conclusion

Exposure of latent fingermarks to ionizing radiation (typically at doses greater than 100 kGy) can in some circumstances result in considerable damage to the organic fingermark compounds and the substrates on which they are deposited. Nonetheless, processing and visually enhancing latent marks on a variety of substrates exposed to ionizing radiation, using common detection reagents and techniques, was successfully accomplished.

At high radiation doses (>100 kGy), the degradation in the quality of the enhanced fingermarks can mostly be attributed to radiolytically induced interactions of the organic compounds in the fingermark deposits which interferes with the various detection processes. In addition, the radiolytic degradation and subsequent alteration to the physiochemical attributes of the various substrates may have affected the ability of the detection technique to selectively target the latent fingermark components. The decreased mechanical properties, caused by this radiolytic degradation, also make complex multistep enhancement processes problematic.

Finally, it should be noted that there are numerous physical, chemical, and environmental variables which can impact on the quality and quantity of fingermark ridge detail that may remain on a substrate. As demonstrated in this study, ionizing radiation is just one more variable to consider.

Acknowledgments

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