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Production of artificial fingermarks. Part II – The use of a modified inkjet printer for the deposition of synthetic secretions

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ABSTRACT

This study is the second part of a larger body of research dedicated to the production of synthetic secretions and the use of an inkjet printer to deposit realistic artificial fingermarks. An artificial emulsion combining eccrine and sebaceous compounds, which was described and tested in the first part of this research, was used as it showed a promising compatibility with common detection techniques. An inkjet printer was modified to print the emulsion on two different substrates: paper (porous) and acetate (non-porous). After optimisation of the printing parameters, multiple fingermarks were printed and processed with a range of standalone detection techniques: 1,2-indanedione-zinc, ninhydrin, Oil Red O, and physical developer on paper, and cyanoacrylate fuming, rhodamine 6G, gold/zinc vacuum metal deposition, and silver black powder on acetate. The detection techniques were also applied in sequence, which is considered one of the biggest advantages of the emulsion over simpler amino acid mixtures that are usable with amino acid reagents only. Natural fingermarks deposited by a single donor were processed with the same techniques for comparison. The effect of water immersion was also investigated, where fingermarks printed on paper were immersed in water for 15 min, before being processed with 1,2-indanedione-zinc and Oil Red O. The results showed that realistic-looking fingermarks could be printed on paper and that printing on acetate was also possible albeit of lower quality due to the nature of the substrate. The artificial fingermarks were successfully enhanced by all the detection techniques tested, at the notable exception of physical developer. The results obtained were very similar to what is generally observed with real fingermarks, and it was observed that the impact of water immersion on the artificial fingermarks was comparable as well. These findings open new perspectives for the development of multi-target quality control test strips or for the standardisation of proficiency testing and interlaboratory collaborative exercises where ground truth is crucial to guarantee comparable results and objective assessment.

1. Introduction

Fingermark variability is a consequence of the natural variations in the chemical matrix found on fingertips between individuals (inter-variability) and for a same individual at different times (intravariability) [1–5]. It is also influenced by several deposition factors such as the quantity of secretions on the fingertips during deposition, as well as the pressure, angle, and duration of contact [6–12]. This variability can sometimes be challenging when developing and comparing fingermark detection methods, especially when poor results are obtained after processing. As the quality of detection relies on both the performance of the technique used and the quality of the fingermarks deposited, it is therefore impossible to unambiguously attribute any failure in detection to one or the other factor or a combination of them. Another area of concern is when items bearing fingermarks are produced for proficiency testing or collaborative exercises purposes. In such cases, fingermark deposition should be controlled to ensure that all participants receive fingermarks with equivalent qualities, which is not possible with natural depositions. This was recently recognised by the Fingerprint Working Group of the European Network of Forensic Science Institutes (ENFSI) when presenting the results of their latest collaborative exercise [13]. In such situations, the use of artificial secretions would be beneficial as they allow for the deposition of reproducible fingermarks.

The chemical composition of eccrine sweat and sebum, the two main

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constituents of fingermark residue, is well documented [3,14] and this knowledge has led to the development of synthetic secretions containing a range of compounds known to be targeted by common detection techniques [15–21]. However, most of those formulations misrepresent the complexity of the matrix that forms fingermark secretions. Studies on fingermark residues have shown that sweat and sebum are present together in the form of water-in-oil (w/o) or oil-in-water (o/w) emulsions, depending on the nature (eccrine-enriched or sebum-enriched) of the fingermark [22]. These observations provide further evidence of the chemical complexity of fingermark residues, which cannot simply be reproduced using eccrine-only or sebaceous-only mixtures.

In the first part of this research, two promising formulations of synthetic residues were reproduced and compared [23]. Both of these simulants consisted of emulsions of eccrine and sebaceous compounds and were expected to react with a range of detection techniques larger than simpler mixtures [24,25]. The emulsions were pipetted on two different types of papers and were then processed with common detection techniques for porous substrates. The results showed that both simulants were reactive towards the amino acid reagents 1,2-indanedione/zinc (IND-Zn) and ninhydrin (NIN), as well as the lipid stain Oil Red O (ORO). Preliminary tests have also demonstrated compatibility with the sequence IND-Zn → NIN → ORO. However, important issues in relation with the pipetting method were highlighted, indicating that the process used to create artificial fingermarks should be carefully chosen to guarantee the best possible reproducibility while producing fingermarks that interact with detection techniques in a similar way as natural depositions.

Several papers have described the use of inkjet or chemical printers for the production of control test strips [13,15,16,18–20,26,27]. These tests take the form of strips of paper on which fields containing specific chemicals with a known concentration are printed. They can then be used to quickly test the presence of a substance or as a quality control for the validity of detection techniques. Printers are ideal to produce test strips as they guarantee a good degree of reproducibility between each strip, are easy to use, relatively fast, and allow the creation of personalised patterns. Some authors used a modified inkjet printer, where the ink in the cartridges was replaced with a particular synthetic solution [13,15,16,19,26–28], while others worked with a specialised chemical printer [18,20,21]. All agree that such test targets can be used for routine reagent quality control or for proficiency testing purposes (and are even recommended for the latter). Most of these studies rely on the use of amino acid solutions that are quick and easy to prepare and generally do not interfere with the printing process. Generally, the amino acid solution formulations are based on natural abundance of amino acids in real fingermarks, with some variations [14]. Kupferschmid et al. [16] used an inkjet printer to produce test strips with fields of ascorbic and oleic acid that can be used as a quality control for physical developer (PD) solutions. Gorka et al. [21] went one step further by printing a synthetic sebum solution with a chemical printer. By doing this, paper strips containing fields of synthetic sweat and sebum were produced, hence expending the range of compatible detection techniques beyond amino acid reagents. By doing so, they could successfully develop the printed patterns with NIN and ORO. However, they were not able to print a proper emulsion and the eccrine and sebaceous fractions were therefore printed on superimposed layers, which doubled the printing time and did not truly represent the mechanism through which natural secretions are formed and transferred onto a substrate.

The printing method is particularly promising because it allows the reproduction of ridge detail that may be used for effective quality assessment [5]. The methodology presented in this paper builds upon [5] to extend to the printing of a synthetic emulsion. To the authors' knowledge this has never been published before. The aim of the study was to assess if this emulsion could be reliably printed and to test its compatibility with detection sequences. Including a synthetic emulsion in the printed matrix would enable the development of multi-targets test strips or proficiency tests that are more representative of real fingermarks than the solutions proposed so far.

2. Materials and methods

2.1. General methodology

The synthetic emulsion used in this research is based on de la Hunty's work [25] and has been presented in the first part of this research [23]. Before printing the emulsion, an inkjet printer had to be modified by opening and emptying the black cartridge and replacing the ink with the emulsion. Different printing parameters were tested to optimise the printing quality. The emulsion was then printed on porous (paper) and non-porous (acetate) substrates and the recommended detection techniques were subsequently applied to each of the substrates, either as standalone or following the recommended sequence for porous and non-porous substrates. The results were recorded and compared to those obtained with real fingermarks.

2.2. Chemicals

The chemicals used for the artificial secretions were detailed in Steiner et al. [23]. All amino acids, as well as sodium chloride, squalene, stearic acid, palmitic acid, oleic acid, stigmasterol, hexane (anhydrous), and dichloromethane were purchased from Sigma-Aldrich. Cholesterol was purchased from Ajax Finechem. Vegetable oil (Crisco – Coles Australia) and vitamin E oil (Plunkett's – Priceline Australia) were used as sources of triglycerides and tocopheryl acetate, respectively. For the detection techniques, ethyl acetate, ninhydrin, Oil Red O, methanol, nitric acid, maleic acid, and rhodamine 6G were purchased from Sigma-Aldrich. Sodium carbonate, ferric nitrate nonahydrate, silver nitrate, citric acid monohydrate, and Tween® 20 were purchased from ChemSupply Pty Ltd. Ammonium ferrous sulfate hexahydrate and methyl ethyl ketone (2-butanone) were purchased from Ajax Finechem. Absolute ethanol was purchased from VWR Chemicals. Isopropanol was purchased from Honeywell. Glacial acetic acid was purchased from RCI Labscan Ltd. 1,2-indanedione was purchased from RedyChemtech. Sodium hydroxide was purchased from Rowe Scientific Pty Ltd. Zinc chloride was purchased from Scharlau. N-dodecylamine acetate was purchased from MP Biomedicals. HFE-7100 was purchased from 3M Novec. Cyanobloom was purchased from Foster+Freeman Ltd. Silver black fingerprint powder and white gelatin lifters were purchased from Sirchie. Gold wires (0.25 mm diameter) and zinc spheres (3 mm diameter) were purchased from West Technology Ltd. All chemicals were stored as recommended by the provider, were analytical reagent grade, and used as supplied.

A detailed list of the chemicals and products used, as well as their purity, can be found in the supplementary material (Table S1 and S2).

2.3. Preparation of the synthetic secretions

The emulsion was obtained after emulsification of synthetic solutions of sweat and sebum, based on de la Hunty's formulation [25]. The detailed formulation and emulsion preparation were described in the first part of this research [23]. The emulsion used in this study has a doubled concentration of sebum, as it was shown to produce better results.

2.4. Inkjet printer and cartridge preparation

An HP Deskjet 3630 drop-on-demand thermal inkjet printer, equipped with disposable HP 63 cartridges, was used to print the emulsion. The print head integrated into the cartridge presented 336 nozzles organised in two rows of 168 nozzles, with each nozzle having a diameter of about 25 μm. Each new cartridge was cut open with a hacksaw and the ink-soaked sponge found inside was discarded; the cartridge was then thoroughly rinsed with deionised water until no

visible ink was seen leaking from the print head; the cartridge was then sonicated for 15 min in a water/isopropanol (50:50, v/v) mixture and dried on absorbing paper; finally, the cartridge was cleaned with ethanol and dried with compressed air until no traces of liquid were left inside the ink chamber. Before each series of printings, the cartridge was filled with up to 3 mL of the emulsion and the lid was replaced. After use, the cartridge was sonicated for 5 min in a water/isopropanol mixture to prevent any drying of the solution that could clog the nozzles. As black and white templates were printed, only black cartridges were used.

2.5. Template creation and printing settings

Fingermark templates were created from inked fingerprint references. The inked standards were taken from two donors (ethics validated under UTS Research Program Ethics ETH182521) and deposited on personal record cards (purchased from Sirchie). The record cards were then scanned in 1200 dpi with a CS9000F Mark II (Canon) scanner. Two fingerprints were selected and converted into 2-bits black and white images using Adobe® Photoshop® CS6. The contrast was then enhanced to obtain ridges as black as possible and the templates were saved at a 1:1 scale.

An A4 document was created on Adobe® Photoshop® CS6 on which each of the fingermark template was copied 3 times at a 1:1 scale (six fingermarks in total). The templates were placed in different areas of the document and in different orientations. The artificial fingermarks were printed in real life size to be as similar to real fingermarks as possible and to allow quality assessment of the printing of very fine ridge details. The A4 document was saved in Portable Document Format (PDF).

The document was first printed with an unmodified black ink cartridge to assess the printing quality under optimal conditions using genuine ink, as well as to ensure that the artificial fingermarks were printed at the correct size. The settings used on the HP Printer are described in Table 1. The printer allowed printing in standard and maximum dots per inch (DPI). Fingermark were printed with both available resolutions before being treated with a solution of IND-Zn to highlight any quality difference between those two resolutions.

2.6. Assessment of printing performance and comparison with real fingermarks

All printings were made in Max DPI using the 6-fingermark template. A porous and a non-porous substrate were used in this study (Table 2). IND-Zn, ORO, PD, and rhodamine 6G (R6G) were prepared and applied on the substrate according to Stoilovic & Lennard [29]. NIN was prepared following the same guidelines, but the development was made using a heat press at 165 °C for 10 s, and steam was added to accelerate the visualization of results. Cyanoacrylate (CA) fuming was undertaken in an MVC1000 (Foster + Freeman) fuming cabinet, the relative humidity was set to 80% and 0.5 g of Cyanobloom was heated to 120 °C for fuming. The humidity cycle was set to 10 min. Fumigation was carried on until a satisfying contrast was obtained, usually 10–15 min of glue cycle. A purge cycle of 15 min was launched after each glue cycle. Gold/Zinc vacuum metal deposition ($VMD_{Au/Zn}$) was carried out in a VMD360 (West Technology) vacuum chamber. A small filament of gold (about 4 mg) was completely evaporated under a high vacuum (2×10^{-4} mbar), followed by zinc until a sufficient contrast was obtained. Silver black powder was carefully applied using a squirrel hair brush and the powdered fingermarks were lifted using white gelatin lifters.

Table 1
Printing settings selected on the HP Deskjet 3630.

Parameter	Value
Media	Plain Paper
Color	Black & White
Paper size	A4
Print in Max DPI	Yes

Table 2
Substrates and compatible detection techniques and sequences used to detect artificial and real fingermarks.

Substrate type	Substrate name	Techniques applied	Sequence
Porous	A4 copy paper 80 gsm - Paperclick	IND-Zn NIN ORO PD $VMD_{Au/Zn}$	1) IND-Zn 2) NIN 3) ORO
Non-porous	A4 transparency film (acetate) - Nobo	CA + R6G $VMD_{Au/Zn}$ Silver black powder	1) CA 2) $VMD_{Au/Zn}$ 3) R6G

Compatible detection techniques and sequences were applied to the fingermarks printed on each substrate (Table 2) [30–32]. The results obtained with the standalone techniques were compared to those obtained with the detection sequences and the quality of detection was compared with natural fingermarks deposited by a single donor and treated with the same techniques. The donor was asked to not wash their hands in the 30 min prior to deposition, as recommended [33,34], and the fingermarks were left to age on a laboratory bench at room temperature for 24 h before any treatment. At least three replicates of each printed fingermark were processed with the chosen techniques or sequences. Two natural fingermarks, deposited at the same time as the printed marks, were processed in the same batch.

The visual appearance of the artificial fingermarks once printed on the acetate, before any treatment with detection techniques, was observed and recorded with a VSC8000 (Foster+Freeman) in coaxial illumination mode. Comparisons were made with freshly deposited fingermarks on the same substrate.

To further understand the emulsion and its reaction to adverse conditions, a short immersion study was undertaken. Artificial fingermarks were printed on paper and then immersed in a tray filled with tap water for a period of 15 min under orbital shake at 80 rotations per minute. The substrates were then removed from the water and left to dry on absorbent paper at room temperature overnight. The fingermarks were then processed with Ind-Zn and ORO (individually and in sequence) and the results compared with artificial fingermarks printed on paper that was not immersed before being processed with the same detection techniques.

3. Results and discussion

3.1. Artificial fingermarks on paper

Before artificial fingermarks were processed and compared to real depositions, the quality of the inkjet printing had to be ensured. Differences in quality were observed between fingermarks printed in Standard and Max DPI (Fig. 1). Ridge detail of artificial fingermarks printed in lower resolution appeared blurrier and more diffused compared to the Max DPI equivalents. Printings in Max DPI also tended to be more consistent, in terms of contrast, than those in Standard DPI. The time required to print 6 fingermarks on an A4 sheet in Max DPI was 140 s, while printing in Standard DPI was about 9 times quicker but had an impact on the quality of the marks. A standard resolution can be considered if quality is not an important factor, but a 2-minute printing time was still considered reasonable as it allowed the printing of artificial fingermarks in large quantities in a single day (> 1000 fingermarks). For these reasons, all the fingermarks were printed in Max DPI for the rest of this study.

Results obtained with artificial fingermarks printed on paper can be found in Table 3. The emulsion could be printed successfully, but a cartridge cleaning was necessary on a regular basis (before every

Fig. 1. (top) 2-bit image of one of the fingermark models used in the study. (bottom) Black and white images (inverted) of an artificial fingermark printed with this model in Standard DPI (left) and Max DPI (right) and treated with IND-Zn. Ridge detail is better defined on the fingermark printed in Max DPI.

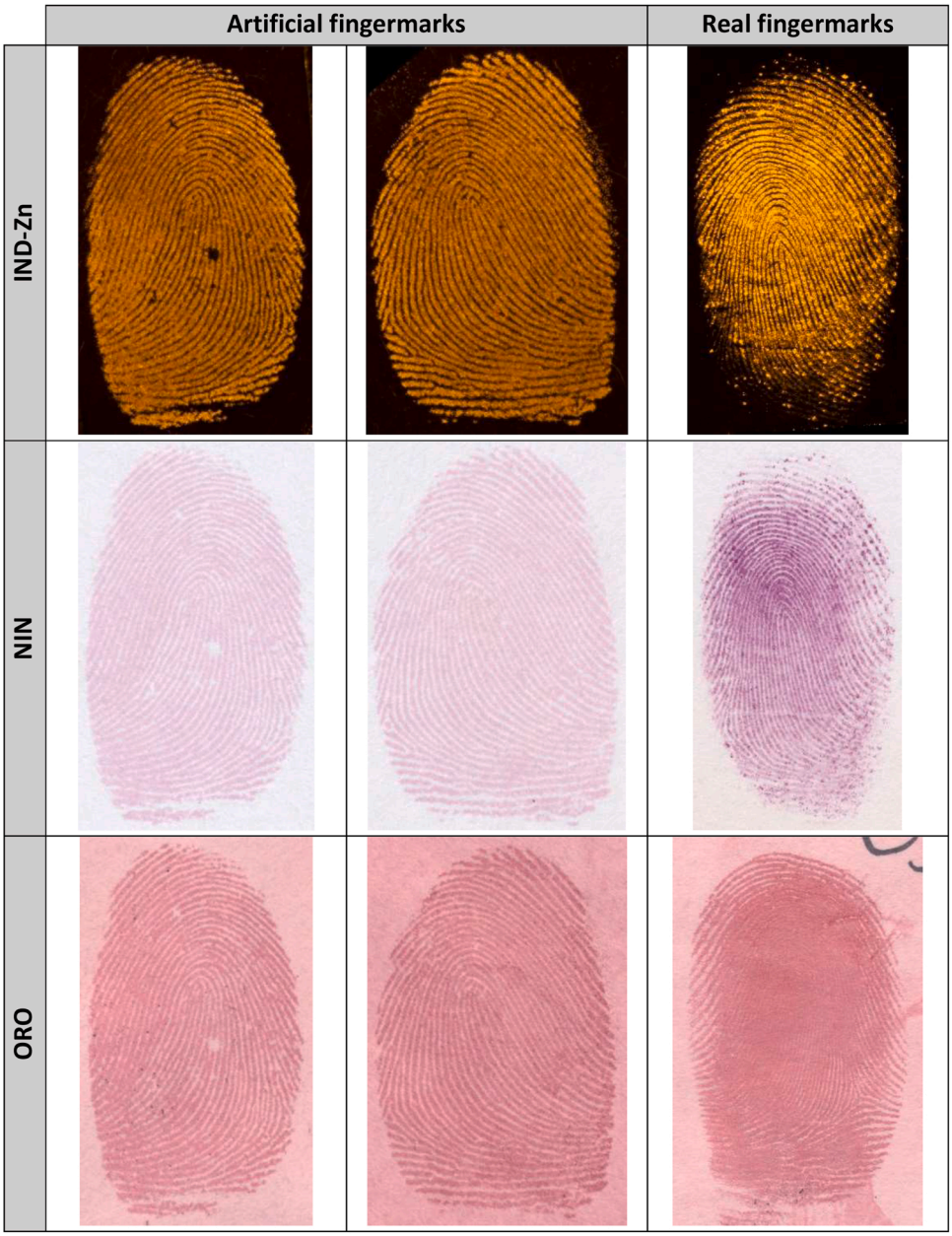
cartridge refill or at the end of a day of printing if the cartridge was not refilled) to avoid dried emulsion clogging the nozzles and impacting the quality. The artificial fingermarks detected with NIN were of visually similar color as the one obtained when real fingermarks were processed, but their contrast was slightly inferior because the amino acid concentration in the emulsion (255 mg/L) was probably lower than the one in the real fingermarks deposited for these comparisons.

When the artificial fingermarks printed on paper were processed in sequence (IND-Zn → NIN → ORO), the NIN reaction was quite faint compared to that observed when the technique was applied individually (Table 4). This decrease in contrast was attributed to a decrease in the amount of amino acids available for the reaction after IND-Zn processing, as both techniques target amino acids. The same phenomenon was observed with real fingermarks, even if the loss in contrast was less pronounced. A drop in quality was observed after ORO processing compared to when the technique was used on its own. Some ridge detail was observed but was not as defined as the artificial fingermarks processed with ORO only. It seemed that detection with the amino acid reagents somewhat had an impact on the performance of ORO, even though the target compounds are known to be different.

This loss of quality was also observed with real fingermarks but was not as pronounced and depended mainly on the quality on the fingermarks themselves (Table 4). Interestingly, this effect was mitigated when a shorter NIN → ORO sequence was used on artificial fingermarks (Fig. 2*a*), with a quality of ridge detail being superior compared to the full sequence. When only IND-Zn was applied before ORO, the contrast and ridge detail were comparable to those obtained with the full sequence (Fig. 2*b*). It appeared that the quality of the artificial fingermarks treated with ORO depended on the detection techniques applied previously, the full sequence leading to the poorest results in terms of quality.

The adverse effect of amino acid reagents when applied before ORO had been previously reported [35–37]. The relatively nonpolar carrier solvents used in IND-Zn and NIN processing may have an impact on the lipid fraction contained in the residue and, hence, on the efficiency of lipid stains such as ORO or Nile Red [36,38], especially when the fingermarks are immersed in the working solutions for extended times. The heat press treatments following IND-Zn and NIN may also be detrimental to the lipid fraction. It is important to note that the working solutions for IND-Zn and NIN used in this project contained HFE-7100 as the carrier solvent and that it was shown to be less detrimental to the lipid fraction than hydrocarbon solvents such as n-hexane or petroleum ether. However, increasing the number of steps in the sequence meant that the fingermarks were immersed in a working solution containing HFE-7100 twice, which might further increase the effect on the subsequent ORO treatment [39]. This would explain why processing the artificial and real fingermarks with only one amino acid reagent increased the quality of the subsequently ORO-treated marks. NIN reaction was accelerated by using a heat press and steam, which might also have had a detrimental impact on the lipids present in the residue. It is possible that this effect would be limited by developing the NIN-treated fingermarks in an oven at 65 °C and 80% RH, as recommended in the CAST Fingermark Visualisation Manual [31]. The fact that this effect was more pronounced on artificial fingermarks than on real ones may be explained by the type of emulsion in both matrices (o/w for the emulsion and w/o for real fingermark residues) and the number of fatty acids available for ORO staining. The carrier solvent HFE-7100 contained in the working solutions might be more detrimental to the fatty acids dispersed in the emulsion compared to real fingermarks, which probably also have a higher proportion of lipids. Adding more lipids to the emulsion could be detrimental to its stability and to the printing process, but further research is needed to optimise the lipid content of the emulsion.

Table 3
Artificial fingermarks printed on paper and treated with IND-Zn, NIN, and ORO (all applied individually). Real fingermarks are shown for comparison.



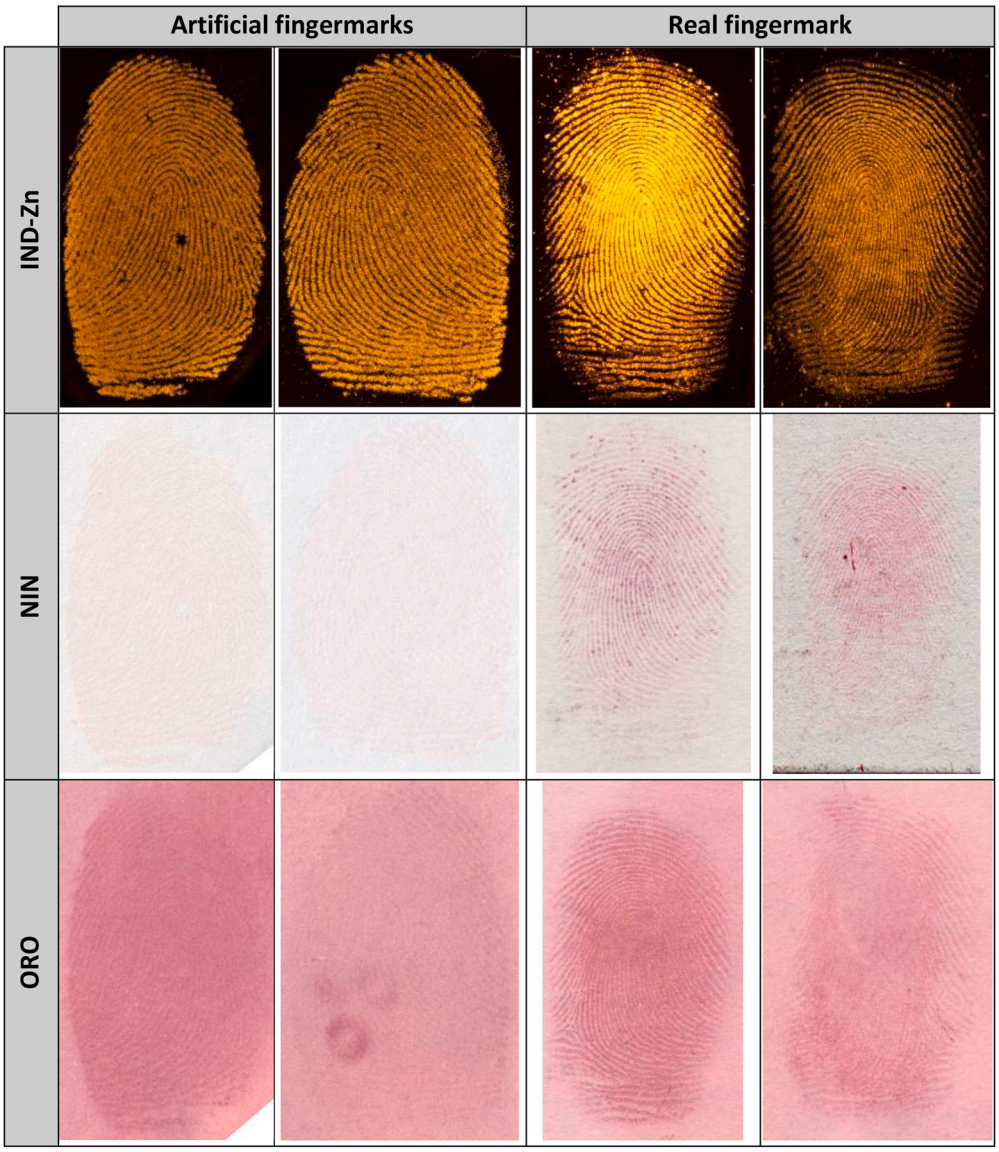
IND-Zn, NIN, and ORO are common techniques used to detect latent fingermarks on paper. However, to demonstrate that the emulsion can lead to positive results with a wider range of techniques, $VMD_{Au/Zn}$ was also applied to the artificial fingermarks printed on paper. This technique is generally used to detect fingermarks on non-porous substrates such as glass or different kinds of plastics, but was originally introduced to detect latent fingermarks on paper [40]. Artificial fingermarks printed with the emulsion and detected with $VMD_{Au/Zn}$ showed a very good quality of ridge detail but with a limited contrast (Fig. 3). $VMD_{Au/Zn}$ is known to work well on fingermarks that have been exposed to water [41], which is indicative of a preferential affinity towards the non-soluble sebaceous compounds found in fingermark secretions. Therefore, this faint contrast is not surprising because the hydrophilic fraction of the emulsion is thought to be dispersed in the continuous aqueous phase and, hence, less concentrated.

It is important to mention that, similarly to what was observed with spot tests [23], poor results were obtained when artificial fingermarks were processed with PD, with almost no signs of silver deposition observed. Further research will look into adding proteins to the emulsion, as they are thought to play a role in silver reduction [42]. The absence of results with the PD remains an issue that will have to be investigated and once again highlights the complexity of the mechanisms involved in silver deposition.

3.2. *Artificial fingermarks on acetate*

When the artificial fingermarks printed on acetate were observed with coaxial illumination before any treatment, a very good contrast was visible and comparable to real fingermarks, even though the ridges were not as defined (Fig. 4). Darker spots were seen along the ridge pattern and were the consequence of very small droplets that were not absorbed in the matrix of the non-porous substrate, unlike paper. This characteristic was inherent to the printing process and could not be avoided. For this reason, the artificial fingermarks printed on acetate were of

Table 4
Artificial fingermarks printed on paper and detected with IND-Zn → Nin → ORO (in sequence). Real fingermarks processed with the same sequence are shown for comparison.



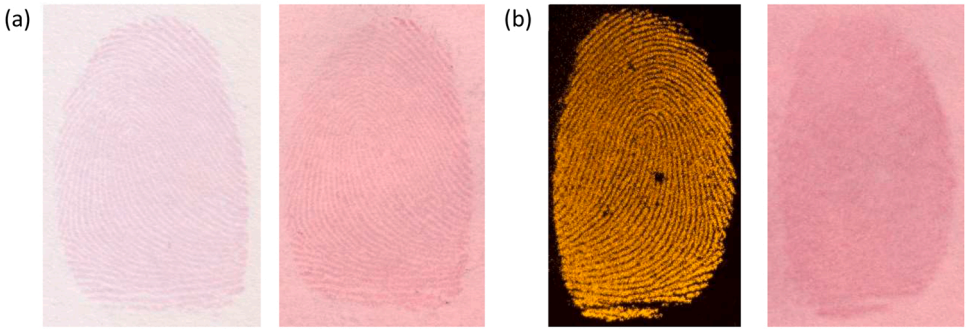


Fig. 2. Two artificial fingermarks treated with the sequence (a) NIN → ORO and (b) IND-Zn → ORO.

poorer quality than the ones printed on copy paper.

The results obtained after treatment with CA, R6G and $VMD_{Au/Zn}$ individually were similar to what was observed on real fingermarks. CA polymerization was observed in a similar way on both artificial and real fingermarks deposited on the acetate. R6G staining led to a clear contrast improvement and the luminescence intensity across the same fingermark was more homogeneous when compared to natural depositions. $VMD_{Au/Zn}$ results were more contrasted than what was observed on paper, but ridge definition was not as good. Empty marks (shape of the fingermark observed but no ridge detail) were observed in

Fig. 3. Two artificial fingermarks detected with $VMD_{Au/Zn}$ on paper.

some cases for both printed and real fingermarks.

While techniques such as CA, R6G and $VMD_{Au/Zn}$ are based on chemical reaction or physico-chemical interactions with the residue, fingermark powders are adhering to the sticky components of secretions by physical interactions. When silver black powder was dusted on artificial fingermarks printed on acetate, results were obtained with a good ridge resolution, but with the same dotted aspect resulting from the jetting process. Lifting the artificial fingermarks using a gelatine lifter presented no detrimental effect on the quality or the contrast of the marks. The results were very similar, in terms of contrast, to what was obtained when real fingermarks were dusted and lifted (Table 5).

The results of the non-porous sequence CA → $VMD_{Au/Zn}$ → R6G are shown in Table 6. Once again, cyanoacrylate polymerization was observed and was similar to what could be observed on real depositions. It is known that the main initiators of CA polymerization are found in eccrine sweat [43], which forms the continuous phase of the emulsion. Therefore, it was not surprising to see such a good level of polymerization, with well-defined ridges.

When $VMD_{Au/Zn}$ was applied after CA, some ridge detail was visible on some of the fingermarks, but the quality was quite variable, with a high proportion of empty marks. These empty marks were hardly avoidable as the technique is known for being very sensitive and producing empty mark when the deposit is too rich. In these cases, residues from the ridges will tend to seep into the valleys of the mark, hindering condensation of the zinc. In the case of the printed fingermarks, it could not be excluded that some of the emulsion seeped into the inter-ridge spaces after printing, due to its lower viscosity compared to real fingermark residue. It was previously reported that this phenomenon was more likely to occur on certain polymer surfaces [44,45]. As this phenomenon of empty marks was observed on some real fingermarks as well, it is thought that the type of acetate used for those experiments was prone to empty marks.

The artificial fingermarks were strongly luminescent after R6G staining and the intensity of the luminescence was comparable to what was observed with real fingermarks. The ridges were well defined but luminescent spots along the ridges, attributed to the printing process, had a detrimental impact on the overall quality.

3.3. *Artificial fingermarks after immersion*

To further study the emulsion and its behavior when exposed to adverse conditions, a short immersion stud was undertaken. An absence of IND-Zn reaction with the artificial fingermarks that were immersed was expected as the amino acids in the synthetic sweat are known to be washed away by water. On the other hand, ORO was expected to perform well as it targets lipids, which are not water-soluble. The results obtained after applying IND-Zn and ORO (individually and in sequence) were in line with those expectations (Table 7). Immersion had an impact on the eccrine fraction of the artificial fingermarks, as no visible marks were obtained after treatment with IND-Zn. Conversely, the artificial fingermarks presented a good contrast and quality when processed with ORO, which successfully demonstrated that the lipids in the emulsion were not washed away by water immersion. Applying IND-Zn before ORO had a negative impact on the quality of the final enhanced marks which showed diffused ridges, as opposed to the well-defined and contrasted artificial fingermarks that were obtained after applying ORO individually. Real fingermarks immersed in water and treated with the sequence IND-Zn → ORO also showed a drop in quality compared to the results obtained with ORO alone. However, this decrease in quality seemed to be dependent on the quality of the fingermark itself and the abundance of lipid material within the matrix (Fig. 5).

4. General discussion

The inks used for inkjet printing are critically formulated to ensure a good quality of printing as heat-sensitive inks cannot be used in thermal inkjet printers for instance [46]. It was a crucial step to choose the right printer to create artificial fingermarks as it should allow the printing of solutions with different properties while maintaining a sufficient quality. It should also be easy to use and clean, and be reproducible. This is especially true if the printer is to be used to produce tests strips or proficiency tests. It is important to note that, according to the printer

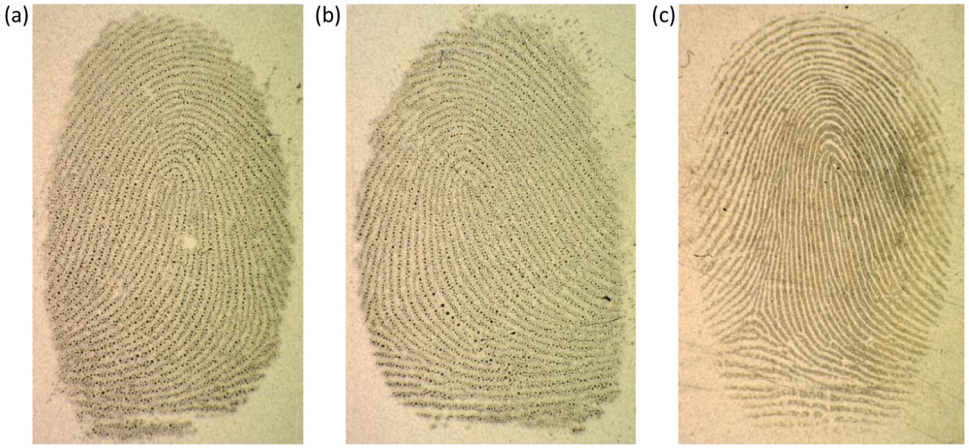
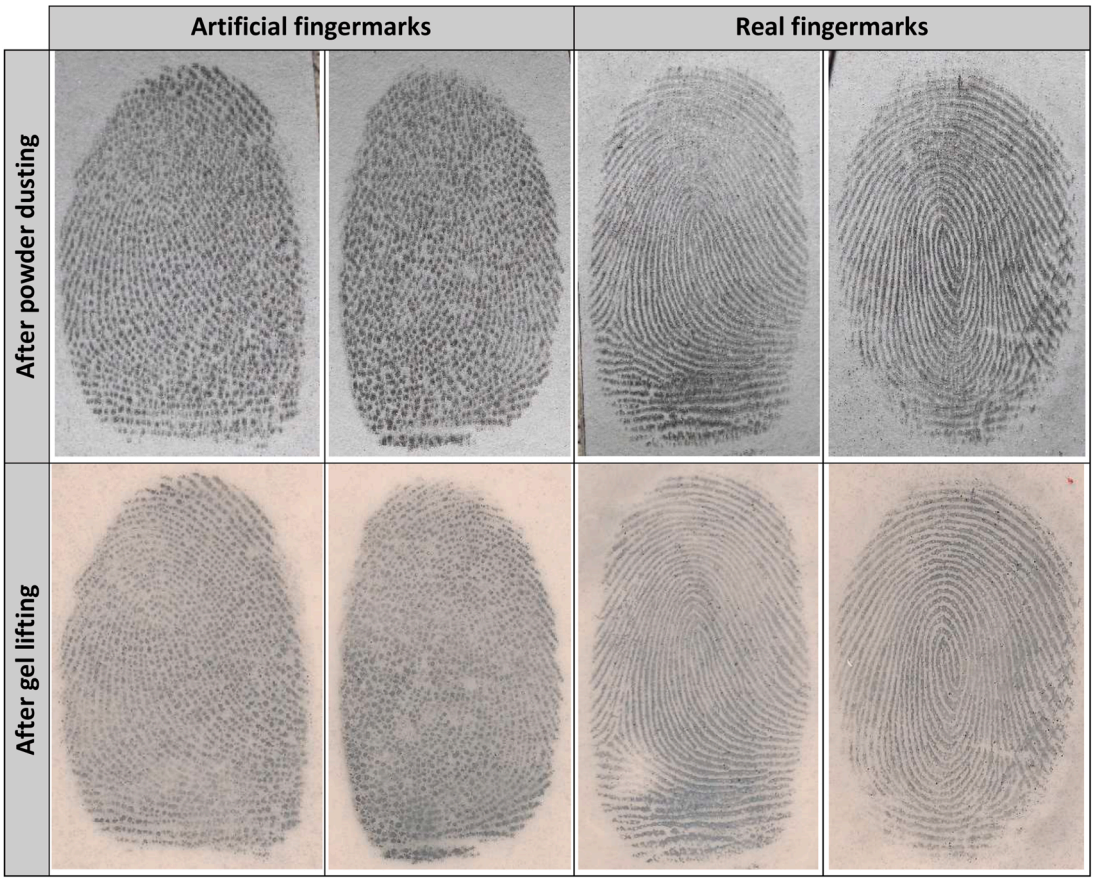


Fig. 4. (a and b) unprocessed artificial fingermarks and (c) a real fingermark, all deposited on acetate and observed in coaxial illumination.

Table 5
Artificial fingermarks dusted with silver black powder within an hour after deposition. Results are shown before and after lifting with a gelatine lifter (images of the gel lift were flipped horizontally). Results obtained with real fingermarks dusted and lifted in the same conditions are shown for comparison.



specifications, the maximum resolution that could be achieved with mono-color printing was 1200 × 1200 dpi. However, this value was only true when genuine ink was used. This resolution could not be guaranteed when solutions with different properties were printed. Factors such as the viscosity and surface tension could interact with the printing process, printing head, and general flow in the cartridge. It could not be excluded that the heat produced during the jetting process had some adverse effect on the emulsion, and other technologies, such as piezoelectric inkjet printing or continuous inkjet printing, should also be tested in the future.

When it came to choosing the substrates, transparent acetate sheets were deemed an ideal choice because they are flexible enough to be loaded in the paper tray of the printer. Also, printing artificial fingermarks on a non-porous substrate was expected to lead to differences in the quality of the depositions and the interaction with the substrate, due to the surface properties. As the ambition was to demonstrate the feasibility of the inkjet printing method on more than one type of surface, a flexible plastic was considered the best solution.

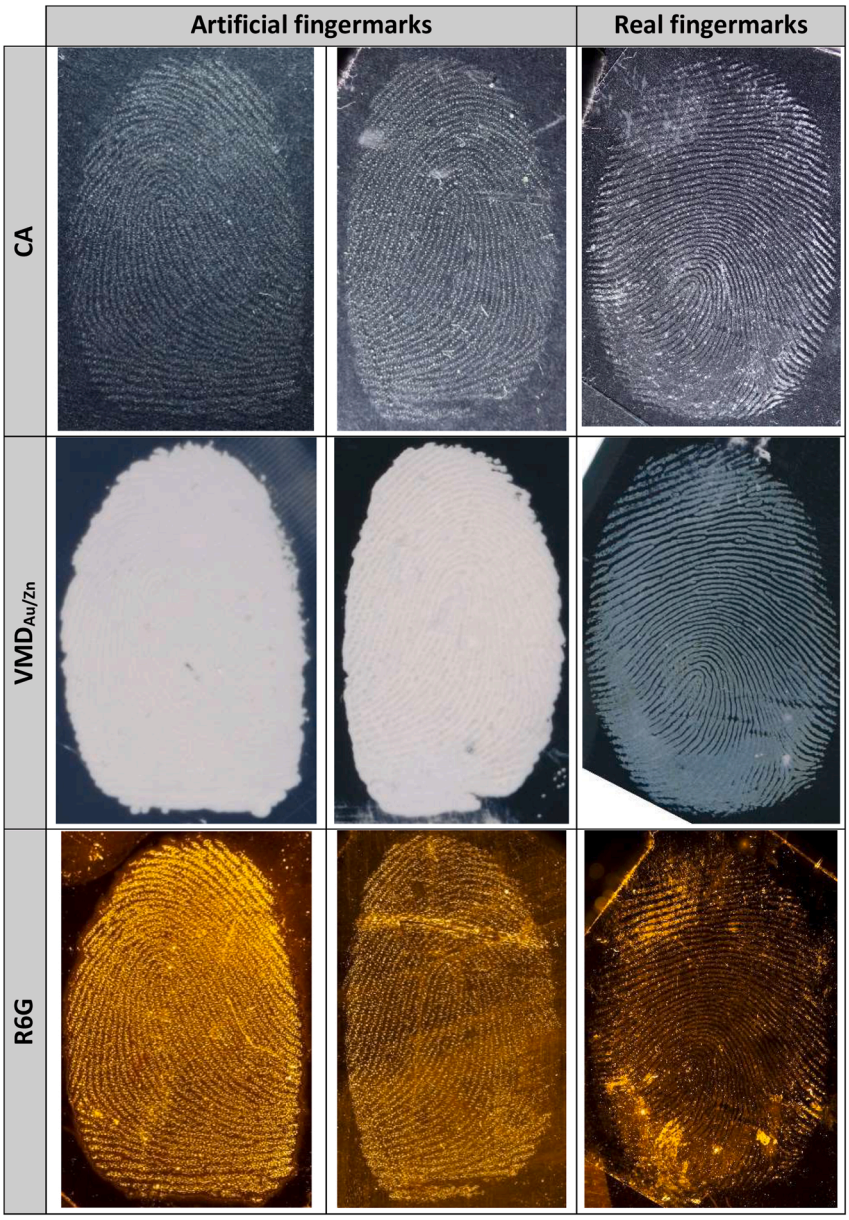
The expected differences in quality between the two substrates were indeed observed in practice. Printings made on paper were extremely well defined and no bleeding of emulsion was observed for any of the fingermarks printed. On acetate, the droplets produced by the jetting processed were seen to dry on the surface as the emulsion did not diffuse in the substrate. The resulting fingermarks had a dotted aspect that did not look as natural as those printed on the porous substrate. However, it should once again be highlighted that the emulsion was reactive towards all the detection techniques tested (at the notable exception of PD on paper), either applied individually or in sequence, on both substrates studied. Also, the results obtained were fairly consistent.

It was observed that the emulsion tended to break down over time, with the dispersed liquids coming out of suspension to flocculate at the surface of the eccrine continuous phase. This phenomenon is well known and is a sign that the emulsion is not stable enough over time [47]. Issues were noted when an aged emulsion was used, particularly when it was printed on acetate (no traces of deposition were seen in coaxial illumination). For this reason, the emulsion was always used within 1 month of preparation. Further studies are required to determine the size of the lipid micelles and the stability of the synthetic fingermark residue over time. The influence of storage conditions will also be investigated.

A specialised Fujifilm printer (Dimatix Materials Printer DMP-2850) described and used by some researchers [18,20,21] was also tested with the emulsion. Despite the fact that the printer shows a promising potential regarding the range of substrates that can be printed on, the quality of the printed fingermarks was nowhere close to what was obtained with the HP inkjet printer. Reproducibility issues were highlighted and led to visible differences in the amount of synthetic residue printed between series. However, it should be mentioned that this printer had never been tested with an emulsion and that the published studies were using synthetic sweat solutions. A change in the printing parameters could improve the quality, but further work is required to provide a robust comparison between the two types of printers.

Overall, the main aim of this study was successfully achieved as it was shown that a modified inkjet printer can be used to print synthetic secretions in a reproducible way on paper, and promising results were obtained on a non-porous substrate. Future developments will focus on optimising the emulsion to improve its shelf life and make it compatible with even more detection techniques. PD compatibility will be particularly investigated as it is usually the third technique in the sequence for porous substrates, following amino acid reagents, and is considered the only technique that works consistently on porous substrates that have

Table 6
Artificial fingermarks printed on acetate and detected with CA → $VMD_{Au/Zn}$ → R6G (in sequence). Real fingermarks processed with the same sequence are shown for comparison.



been exposed to water. Analytical techniques should also be used to get a better understanding of the form the emulsion takes and to determine if any compounds are lost during the printing process, for example due to the filter found inside the cartridges to avoid ink leakage.

It is important to note that artificial fingermarks are not intended to completely replace the use of real fingermarks in research & development, but could be an alternative in cases where fingermark variability is an issue. The method presented has beneficial applications in proficiency testing by ensuring a proper quality control before the marks are sent to participants. It would also be a helpful tool in the first phases of development of new detection techniques. Also, artificial secretions could be useful for quality control purposes where multi-targets test strips could be used to test the performance of detection techniques before application. It is acknowledged that natural fingermarks still need to be used in cases where variability is needed, such as (pseudo-) operational and validation trials.

5. Conclusions

This study investigated a method focused on a modified inkjet printer to create artificial fingermarks using a synthetic emulsion. The aim was to demonstrate the feasibility of the inkjet printing, to assess the quality of the printed fingermarks once processed with detection techniques, and to compare the results with natural fingermarks.

An HP Printer was used to print the emulsion on copy paper and transparent acetate sheets. The printed fingermarks were then treated with some of the most common visualization techniques used on each of the substrates to assess the quality of the printings, as well as the quality of detection. The emulsion was successfully printed and no clogging of the cartridges was observed as long as they were cleaned on a regular basis. High quality fingermarks were obtained on paper and successfully detected with the amino acid reagents IND-Zn and NIN, as well as the lipid stain ORO, which confirmed the combined presence of eccrine and sebaceous material in the printed matrix. When the same three detection techniques were applied in sequence, a loss in quality was observed after

Table 7
Artificial fingermarks printed on paper and immersed in water for 15 min compared to dry artificial fingermarks. Fingermarks were treated with IND-Zn and ORO (individually and in sequence).

	Detection techniques individually		Detection techniques in sequence	
	Dry	Wet	Dry	Wet
IND-Zn		*Undetected*		*Undetected*
ORO				

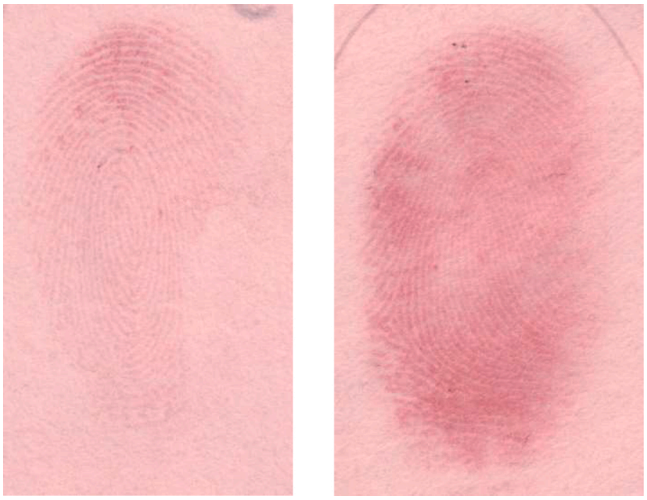


Fig. 5. Real fingermarks immersed in water for 15 min and treated with ORO (left) individually and (right) in sequence after IND-Zn.

ORO treatment, with fingermarks appearing fainter and less detailed compared to the depositions processed with ORO only. This phenomenon could be mitigated by using a single amino acid reagent prior to ORO but the impact on the quality was more important than what was observed on real fingermarks. It was also shown that artificial fingermarks that were immersed for 15 min behaved in the same manner as real marks and were properly detected with ORO while amino acid reagents were unsuccessful.

Satisfying results were obtained when artificial fingermarks were processed with CA + R6G or $VMD_{Au/Zn}$ and with the sequence for non-porous substrates (CA → $VMD_{Au/Zn}$ → R6G). A visible CA polymerization was observed on the artificial fingermarks ridges and the luminescence resulting from R6G staining was similar to what was seen with real fingermarks. Most of the marks processed with $VMD_{AU/Zn}$ were empty with a well-defined print area but no ridge detail. This phenomenon was also observed on some real fingermarks, especially when they were rich in secretions, and was thought to be caused by the type of acetate chosen. The artificial fingermarks printed on acetate appeared as dotted lines due to the inherent printing process where droplets jetted by the cartridge could not be sufficiently absorbed into the substrate and eventually dried on the surface. The quality of the marks on acetate was thereby not as good as what could be achieved on a porous substrate where the emulsion was easily absorbed into the fibres of the paper. It should be noted that printing on paper, which is a very common substrate found in casework, was the focus of the project and that the emulsion was designed to be printed with a high quality on this substrate. Optimization of the emulsion to improve its jetting and adhesion to plastic surfaces would undoubtedly improve the quality of the artificial fingermarks printed on such substrates.

The methodology presented in this paper is an important step towards a more generalised use of artificial fingermarks in routine laboratory work and for improved quality control. Using an emulsion that combined eccrine and sebaceous compounds was shown to mimic natural fingermarks more realistically than simpler synthetic sweat secretions. The inkjet printing method was shown to be fast and reliable to print multi-target test strips or realistic latent marks for proficiency testing purposes. It is hoped that these results will lead the way to further research and optimization of synthetic emulsions, which would allow even more flexibility and potential applications.

CRediT authorship contribution statement

Romain Steiner: Conceptualization, Methodology, Investigation, Resources, Writing – original draft, Writing – review & editing, Visualization. **Sebastien Moret**: Conceptualization, Methodology, Validation, Writing – review & editing, Supervision. **Claude Roux**: Validation, Writing – review & editing, Supervision, Project administration.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.forsciint.2023.111804.

References

[1] R.S. Croxton, M.G. Baron, D. Butler, T. Kent, V. Sears, Variation in amino acid and lipid composition of latent fingerprints, Forensic Sci. Int. 199 (1) (2010) 93–102.
[2] C. Weyermann, C. Roux, C. Champod, Initial results on the composition of fingerprints and its evolution as a function of time by GC/MS analysis, J. Forensic Sci. 56 (1) (2011) 102–108.
[3] A. Girod, R. Ramotowski, C. Weyermann, Composition of fingermark residue: a qualitative and quantitative review, Forensic Sci. Int. 223 (1) (2012) 10–24.
[4] S. Chadwick, S. Moret, N. Jayashanka, C. Lennard, X. Spindler, C. Roux, Investigation of some of the factors influencing fingermark detection, Forensic Sci. Int. 289 (2018) 381–389.
[5] R. Steiner, C. Roux, S. Moret, Controlling fingermark variability for research purposes: a review, Wiley Interdiscip. Rev.: Forensic Sci. 1 (4) (2019).
[6] G.L. Thomas, T.E. Reynoldson, Some observations on fingerprint deposits, J. Phys. D: Appl. Phys. 8 (6) (1975) 724–729.
[7] B. Scruton, B.W. Robins, B.H. Blott, The deposition of fingerprint films, J. Phys. D: Appl. Phys. 8 (6) (1975) 714–723.
[8] G.L. Thomas, The physics of fingerprints and their detection, J. Phys. E: Sci. Instrum. 11 (8) (1978) 722–731.
[9] S. Mil'shtein, U. Doshi, Scanning the pressure-induced distortion of fingerprints, Scanning 26 (6) (2004) 270–272.
[10] S. Fieldhouse, Consistency and reproducibility in fingermark deposition, Forensic Sci. Int. 207 (1–3) (2011) 96–100.
[11] S. Fieldhouse, A comparison of fingermark deposition methodologies, Fingerpr. Whorld 37 (143) (2011) 94–101.
[12] S. Fieldhouse, An investigation into the effects of force applied during deposition on latent fingermarks and inked fingerprints using a variable force fingerprint sampler, J. Forensic Sci. 60 (2) (2015) 422–427.
[13] F. Zampa, M. Hilgert, J. Malmborg, M. Svensson, L. Schwarz, A. Mattei, Evaluation of ninhydrin as a fingermark visualisation method – a comparison between different procedures as an outcome of the 2017 collaborative exercise of the ENFSI fingerprint working group, Sci. Justice 60 (2) (2020) 191–200.
[14] R. Ramotowski, Composition of Latent Print Residue, in: H.C. Lee, R.E. Gaensslen (Eds.), Advances in Fingerprint Technology, second ed., CRC Press, Boca Raton, 2001, pp. 63–104.
[15] L. Schwarz, An amino acid model for latent fingerprints on porous surfaces, J. Forensic Sci. 54 (6) (2009) 1323–1326.
[16] E. Kupferschmid, L. Schwarz, C. Champod, Development of standardized test strips as a process control for the detection of latent fingermarks using physical developers, J. Forensic Identif. 60 (6) (2010) 639–655.
[17] N. Thiburce, A. Bécue, C. Champod, F. Crispino, Design of a control slide for cyanoacrylate polymerization: application to the CA-bluestar sequence, J. Forensic Identif. 61 (3) (2011) 232–249.
[18] R. Janssen-Bouwmeester, C. Bremmer, L. Koomen, S. Siem-Gorre, M. de Puit, Positive control tests for fingermark development reagents, Forensic Sci. Int. 310 (2020), 110259.
[19] R. Croxton, T. Kent, A. Littlewood, M. Smith, An evaluation of inkjet printed amino acid fingerprint test targets for ninhydrin process monitoring – and some observations, Forensic Sci. Int. 321 (2021), 110741.
[20] A. Jeanneret, A. Anthonioz, A. Bécue, Printed artificial sweat as replacement for natural fingermarks: qualitative and quantitative approach considering an amino acid reagent, Sci. Justice 61 (3) (2021) 249–259.
[21] M. Gorka, A. Thomas, A. Bécue, Development of a printed quality control test strip for the analysis and imaging of fingermark composition, Forensic Sci. Int. 329 (2021), 111063.
[22] B.N. Dorakumbura, R.E. Boseley, T. Becker, D.E. Martin, A. Richter, M.J. Tobin, W. van Bronswjik, J. Vongsvivut, M.J. Hackett, S.W. Lewis, Revealing the spatial distribution of chemical species within latent fingermarks using vibrational spectroscopy, Analyst 143 (17) (2018) 4027–4039.
[23] R. Steiner, C. Roux, S. Moret, Production of artificial fingermarks. Part I – synthetic secretions formulation, Forensic Sci. Int. 331 (2022), 111166.
[24] E. Sisco, J. Staymates, K. Schilling, A chemically relevant artificial fingerprint material for the cross-comparison of mass spectrometry techniques, J. Can. Soc. Forensic Sci. 48 (4) (2015) 200–214.
[25] M. de la Hunty, An Investigation of Latent Fingermark Residues and Their Development on Porous Substrates Using Physical Developer and Nile Red, University of Technology Sydney, Australia, 2017.
[26] L. Schwarz, I. Klenke, Enhancement of Ninhydrin- or DFO-treated latent fingerprints on thermal paper, J. Forensic Sci. 52 (3) (2007) 649–655.
[27] L. Schwarz, M. Baisei, Erster ringversuch zur sicherung latenter daktyloskopischer spuren mit reproduzierbaren testspurenträgern, Kriminalistik 62 (8–9) (2008) 500–505.
[28] S. Hong, I. Hong, A. Han, J.Y. Seo, J. Namgung, A new method of artificial latent fingerprint creation using artificial sweat and inkjet printer, Forensic Sci. Int. 257 (2015) 403–408.
[29] M. Stoilovic, C. Lennard. Fingermark Detection & Enhancement, sixth ed., National Centre for Forensic Studies, Canberra, Australia, 2012.
[30] B.J. Jones, R. Downham, V.G. Sears, Nanoscale analysis of the interaction between cyanoacrylate and vacuum metal deposition in the development of latent fingermarks on low-density polyethylene, J. Forensic Sci. 57 (1) (2012) 196–200.
[31] H.L. Bandey, S.M. Bleay, V.J. Bowman, R.P. Downham, V.G. Sears. Fingermark Visualisation Manual, first ed., Home Office Centre for Applied Science and Technology, 2014.
[32] C. Champod, C. Lennard, P. Margot, M. Stoilovic. Fingerprints and Other Ridge Skin Impressions, second ed., CRC Press, Boca Raton, 2016.
[33] V.G. Sears, S.M. Bleay, H.L. Bandey, V.J. Bowman, A methodology for finger mark research, Sci. Justice 52 (3) (2012) 145–160.
[34] International Fingerprint Research Group (IFRG), Guidelines for the assessment of fingermark detection techniques, J. Forensic Identif. 64 (2) (2014) 174–200.
[35] A.A. Frick, P. Fritz, S.W. Lewis, W. van Bronswijk, Sequencing of a modified Oil Red O development technique for the detection of latent fingermarks on paper surfaces, J. Forensic Identif. 63 (4) (2013) 369–385.
[36] K. Braasch, M. de la Hunty, J. Deppe, X. Spindler, A.A. Cantu, P. Maynard, C. Lennard, C. Roux, Nile red: alternative to physical developer for the detection of latent fingermarks on wet porous surfaces? Forensic Sci. Int. 230 (1–3) (2013) 74–80.
[37] C. Marriott, R. Lee, Z. Wilkes, B. Comber, X. Spindler, C. Roux, C. Lennard, Evaluation of fingermark detection sequences on paper substrates, Forensic Sci. Int. 236 (2014) 30–37.
[38] M. de la Hunty, X. Spindler, S. Chadwick, C. Lennard, C. Roux, Synthesis and application of an aqueous nile red microemulsion for the development of fingermarks on porous surfaces, Forensic Sci. Int. 244 (2014) e48–e55.
[39] J. Salama, S. Aumeer-Donovan, C. Lennard, C. Roux, Evaluation of the fingermark reagent Oil Red O as a possible replacement for physical developer, J. Forensic Identif. 58 (2) (2008) 203–237.
[40] P. Theys, Y. Turgis, A. Lepareux, G. Chevet, P.F. Ceccaldi, Nouvelle technique de révélation de traces papillaires latentes (sur le papier) par métallisation sous vide, Rev. Int. Police Crim. 23 (217) (1968) 106–108.
[41] R. Steiner, A. Bécue, Effect of water immersion on multi- and mono-metallic VMD, Forensic Sci. Int. 283 (2018) 118–127.
[42] S. Houlgrave, M. Andress, R. Ramotowski, Comparison of different physical developer working solutions - Part I: longevity studies, J. Forensic Identif. 61 (6) (2011) 621–639.
[43] S.P. Wargacki, L.A. Lewis, M.D. Dadmun, Understanding the chemistry of the development of latent fingerprints by superglue fuming, J. Forensic Sci. 52 (5) (2007) 1057–1062.
[44] N. Jones, D. Mansour, M. Stoilovic, C. Lennard, C. Roux, The influence of polymer type, print donor and age on the quality of fingerprints developed on plastic substrates using vacuum metal deposition, Forensic Sci. Int. 124 (2–3) (2001) 167–177.
[45] N. Jones, M. Stoilovic, C. Lennard, C. Roux, Vacuum metal deposition: factors affecting normal and reverse development of latent fingerprints on polyethylene substrates, Forensic Sci. Int. 115 (1–2) (2001) 73–88.
[46] H. Kipphan, Handbook of Print Media, Springer-Verlag Berlin Heidelberg, Berlin, Germany, 2001, p. 1207.
[47] T.F. Tadros, Applications of Surfactants in Emulsion Formation and Stabilisation. Applied surfacants, Wiley-VCH, Great Britain, 2005.