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Title

Effect of water immersion on multi- and mono-metallic VMD

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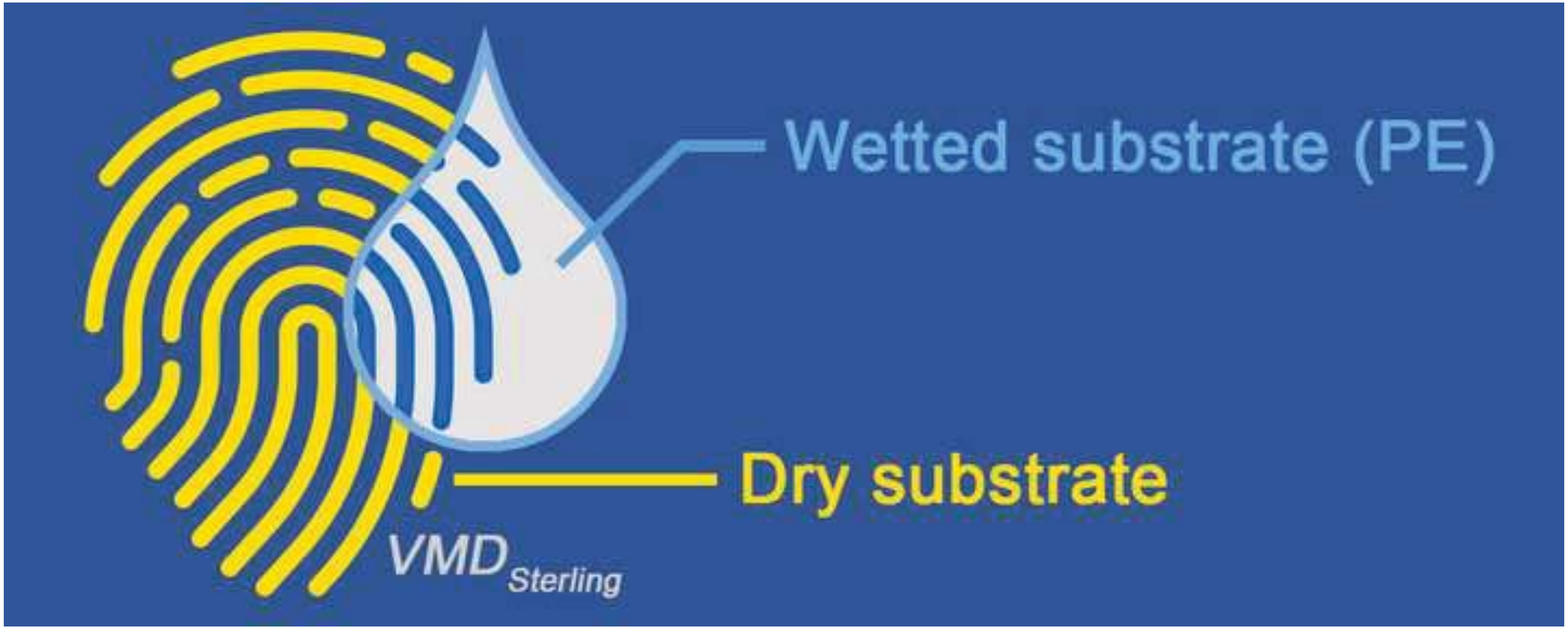
Graphical Abstract (for review)



Highlights

* Quantitative/Qualitative study involving silver, sterling silver, and gold/zinc VMDs

* Immersion of items has limited impact on the quality of VMD-processed fingermarks

* Immersion of items may lead to contrast reversal for polyethylene (conventional VMD)

* Immersion of items leads to changes in fingermark color shades (monometallic VMDs)

* Monometallic VMDs appear more adapted to PVC than conventional VMD

Abstract

The use of vacuum metal deposition (VMD) for fingermark detection has been known for almost 40 years. The technique is applicable on a wide variety of substrates and on wetted items. Several publications compare the relative efficiency of VMD (conventionally based on a successive vaporization of gold followed by zinc) with other detection techniques, or its ability to detect marks on difficult substrates, but few are known about the application of monometallic VMDs and about the impact of immersion on the detection performances. This study aims at partially filling that gap by offering a quantitative and qualitative glance at three VMD processes (*i.e.*, gold/zinc, silver, and sterling silver) applied to dry and wetted substrates. The impact of immersion on the detection process has been studied by using split marks (one half kept dry, the other one wetted). On immersed substrates, a modification of color shades has been observed with monometallic VMDs (on all substrates considered) and of contrast with conventional VMD (on polyethylene). In terms of ridge details, a relatively good resistance of secretion residue towards immersion has been emphasized (in regards with VMD). This study provides original data, which will hopefully help getting a better understanding of the VMD detection mechanism.

Keywords - Forensic science, Fingermark, Detection, Vacuum metal deposition, Contrast

Introduction

Vacuum metal deposition (VMD) is part of the currently available fingermark detection techniques [1]. It is mostly characterized by its versatility of application (*i.e.*, range of compatible substrates) and its efficiency, especially regarding difficult cases (*e.g.*, problematic substrates, adverse conditions). The technique is based on the vaporization of one or two metal(s) under vacuum, towards the item to be processed. Fingermarks becomes visible by the formation of a metallic film on the substrate (normal development) or on the secretion residue (reverse development), most likely due to a differentiated condensation mechanism. VMD was initially introduced in the forensic field in 1968 to detect fingermarks on paper [2], and was then optimized to be fully operational in the late seventies [3]. The conventional VMD process is based on the successive vaporization of gold and zinc ($VMD_{Au/Zn}$). Monometallic alternatives were also developed and offer the advantage of establishing a visible contrast in one step. They are complementary to $VMD_{Au/Zn}$ for they can develop fingermarks on substrates for which $VMD_{Au/Zn}$ results in poor performances. Monometallic VMDs can be based on silver (VMD_{Ag}) [4-8], copper (VMD_{Cu}) [8, 9], aluminium (VMD_{Al}) [7, 10], or palladium (VMD_{Pd}) [7], to cite a few.

In terms of contrast, $VMD_{Au/Zn}$–processed fingermarks will most likely results in transparent ridges opposed to a metal-coated substrate (Figure 1a). This kind of contrast is not common in the field of fingermark detection, since detection techniques generally result in stained ridges (colored or luminescent) opposed to a passive substrate. In some cases, $VMD_{Au/Zn}$ can result in fingermarks presenting ridges coated with a metal film. In that case, we speak of a "reverse" development, in regards with VMD. Finally, some processed fingermarks may present a normal contrast but no inner ridge details ("empty marks") – Figure 1a. In this paper, the obtained contrasts (*i.e.*, normal or reverse) are qualified in regards with a conventional $VMD_{Au/Zn}$ result, that is, "coated substrate vs transparent ridges". This distinction hardly applies to monometallic VMDs, which mostly result in colored contrasts (Figure 1b).

< INSERT FIGURE 1 HERE>

A strength of the VMD is its versatility of application, for it is compatible with an extended range of substrates (*e.g.*, porous, non-porous, metals, adhesives, wetted substrates) among which challenging ones, such as banknotes [9, 11, 12] or fabrics [5, 13, 14]. The use of VMD is compatible with "touch DNA" profiling [15, 16] and it complements the conventional techniques as it can be introduced in detection sequences; even if no consensus does exist regarding its relative position with other techniques, especially cyanoacrylate fuming [12, 17-19]. The technique nevertheless suffers from its cost (*i.e.*, a specific and costly equipment is required), the necessity to gain experience with its handling before obtaining acceptable detection results, and a detection mechanism which remains partially understood [20, 21]. This results in substantial variations of efficiency according to the substrate composition, especially polymers/plastics and surface treatments [19, 20, 22-24]. Guidelines and best practice recommendations can be provided to users but they don't overcome all these difficulties [17, 19, 25]. Research in the field of VMD is consequently a valuable source of information for people willing to gain a better understanding of the technique.

This study originated from a detection course we organized about mono-/bi-metallic VMD. During this course, a hand mark (fingers and palm) was left on a PVC plastic sheet that was then briefly and partially immersed in water (half the substrate remained dry). Once dried, the whole plastic sheet was processed with VMD_{Ag}. As a result, half of the hand mark appeared with yellow/blue color tones (dry half) while the other half appeared with blue/purple tones (wetted half). This change of color upon immersion has not been reported in the literature yet, to the authors' knowledge. Some research has been performed on wetted items processed with $VMD_{Au/Zn}$ [26], but no systematic study regarding the impact of immersion or the use of monometallic VMD in this context. This contribution consequently aims at exploring this phenomenon and providing original data that may help getting a better understanding about the intrinsic VMD detection mechanism.

Materials and methods

- Substrates and fingermark collection

Three non-porous substrates were chosen: white polyethylene (PE containing 50% recycled material; official state garbage bag), transparent polyvinylchloride (HiClear PVC; GBC), and glass (microscopy slides; VWR). Fingermarks were collected from three donors who were asked to leave natural marks [27]. Natural marks were exclusively used in this study, to offer a more realistic approach since secretions are not artificially enriched with sweat or sebum. The only recommendations that the donors received were to act normally, at the exception of washing their hands (prohibited 30 minutes before the deposition). To allow a direct comparison (*i.e.*, Situation A *vs* Situation B), halved marks were used. For that, fingermarks left on plastics (PE and PVC) were cut after deposition; for glass, donors were asked to leave fingermarks between two slides put aside. Finally, replicates were also considered in that sense that donors were asked to give more than one fingermark for a specific comparative study (all other parameters being set). All the substrates bearing fingermarks were stored in the dark until being used (immersed and/or processed with VMD).

- Detection techniques

Three kinds of VMDs were considered in this study: the conventional one based on the successive vaporization of gold and zinc ($VMD_{Au/Zn}$) and two monometallic ones based on silver (VMD_{Ag}) and sterling silver ($VMD_{Sterling}$). The vaporization chamber was a VMD 360 from West Technology (Bristol, UK). All metals were of high purity and provided by West Technology (gold wire ∅ 0.25mm, zinc spheres ∅ 3mm, silver wire ∅ 0.5 mm, and sterling silver wire ∅ 2mm).

- Immersion procedures

Three studies were conducted: (Study 1) Influence of the immersion time; (Study 2) Influence of the age of the fingermarks upon immersion; (Study 3) Difference between VMDs when immersed items are to be processed. Experimental details are summarized in Table 1.

< INSERT TABLE 1 HERE >

For the influence of the immersion time (Study 1), all the marks were aged for 48H. Then, one half of each fingermark was kept dry (reference) whereas the corresponding half was immersed for 1H or 24H in a dish filled with tap water. When the immersion time was reached, all wet halves were removed from water and left for drying under ambient temperature for 24 hours at least before being processed for detection using VMD. For the influence of the age of the marks upon immersion (Study 2), one half of each fingermark was immersed 24H after deposition (1D-old) whereas the corresponding half was immersed 1 week after deposition (1W-old). After being immersed for 24H, the marks were removed from water and dried for 24 hours at least before being processed with VMD. For the influence of the metal (VMD_x, x being Ag, Sterling or Au/Zn; Study 3), all halved fingermarks were aged for 48H before being immersed for 24H in tap water. Afterwards, the marks were dried for 24 hours at least and processed in accordance with the scheduled comparisons. For all the experiments: half marks were processed using either VMD_{Ag}, $VMD_{Sterling}$, or $VMD_{Au/Zn}$. After VMD processing, the corresponding halves were put aside and captured under white light for further characterization.

- Characterization of the detected fingermarks

The quality of all half fingermarks was assessed in terms of ridge quality (identification purposes) by considering an absolute scale (Table 2) [28]. To avoid any bias during the scoring procedure, each reconstructed fingermark was enhanced (*i.e.*, contrast, levels) and converted into greyscales. The images were then cropped to allow the recording of each half independently from the other. Finally, right halves were horizontally inverted so that they look like left halves. At the end of this process, the assessors were provided with a series of left-handed half-marks in greyscales, which were beforehand shuffled. This way of doing prevents an assessor to associate a half mark with a specific comparative study or a specific VMD. The scoring procedure was conducted by two independent assessors familiar with fingermark detection and identification, each one assessing the totality of the marks. At the end of the process, the scores were averaged to provide conclusions about each study.

< INSERT TABLE 2 HERE >

Besides the quality of their ridges, the marks processed with VMD_{Ag} and $VMD_{Sterling}$ were characterized by their color shades (ridges *vs* background). For $VMD_{Au/Zn}$, the contrast type was reported (*i.e.*, normal or reverse).

Results

After their detection, all the half marks were assessed quantitatively (quality score) and qualitatively (color shades or contrast). This characterization step allowed to get an overview of the effect of immersion on the VMD results (dry *vs* wet), but also of the differences between substrates (for a same VMD) and between VMDs for a same substrate (VMDx *vs* VMDy).

- Fingermark quality

The results of the three studies are illustrated in Figures 2 to 4 and summarized in Table 3.

< INSERT FIGURES 2 TO 4 HERE >

< INSERT TABLE 3 HERE >

Study 1 provides information about the effect on the fingermark quality of an immersion (1H-long or 24H-long) compared to non-wetted halves (Figure 2). On PE, the overall impact of the immersion is very limited, with scores similar to the dry halves (exception: pronounced detrimental effect observed for VMD_{Ag} after a 24H immersion). On PVC, the overall impact is negative, with decreasing scores (exception: VMD_{Ag} after a 1H immersion). About PVC, the processed of wetted items with $VMD_{Au/Zn}$ resulted in several empty marks. Finally, the effect is mixed for glass: increase of quality after a 1H immersion for VMD_{Ag}, decrease of quality for $VMD_{Sterling}$, and limited effect for $VMD_{Au/Zn}$.

Study 2 aimed at assessing if older marks (*i.e.*, 1W-old) resist better to immersion compared to fresh ones (*i.e.*, 1D-old), both being immersed for 24H before being processed. By averaging the difference of scores between corresponding halves ($Score_{1W}$ – $Score_{1D}$), an overall trend can be obtained for each substrate and VMD (Figure 3). A negative value means that older halves (*i.e.*, 1W-old) led to lower quality scores compared to the corresponding fresh halves (*i.e.*, 1D-old). On PE, older marks resulted in lower quality marks compared to fresh ones, for all three VMDs. On PVC, the trend is mixed: better quality for fresh marks with $VMD_{Sterling}$, negligible impact of the age with VMD_{Ag}, and better quality for older marks with $VMD_{Au/Zn}$. On glass, the overall trend is the opposite as for PE, for older marks led to higher quality scores compared to fresh ones.

Study 3 provides a direct comparison between the three VMDs as they were applied to half marks having followed the same detrimental process (*i.e.*, aged for 48H, immersed for 24H, then processed with VMD). Figure 4 illustrates the trends that were obtained by averaging all the quality scores associated with a specific substrate and VMD process. On PE, all three VMDs performed equally in terms of overall quality of development. On PVC, $VMD_{Au/Zn}$ led to several empty marks, explaining why its scores are so low. VMD_{Ag} and $VMD_{Sterling}$ performed well, with a preference for VMD_{Ag}. On glass, the trend is the opposite: $VMD_{Au/Zn}$ gave better results compared to VMD_{Ag} and $VMD_{Sterling}$.

- Contrast and color shades

While the conventional VMD ($VMD_{Au/Zn}$) is monochromatic, monometallic VMDs offer a range of color shades influenced by the substrate, the donors, and the adverse conditions (Figure 5). An overview of the categories of contrasts that were observed after having processed all the fingermarks with VMD_{Ag}, $VMD_{Sterling}$, and $VMD_{Au/Zn}$ is provided in Figures 6 to 8, respectively. In addition to the qualitative information, the treemap representations provide a visual information about the proportion of marks presenting a specific contrast (*i.e.*, a sub-area is proportional to the % of marks characterized by the illustrated contrast, each main rectangle being equal to 100%) [29]. It should be noted that these charts provide information about the observed contrasts but not about the quality of the fingermarks.

< INSERT FIGURE 5 HERE>

< INSERT FIGURES 6 to 8 HERE>

The following explains some of the images of Figure 6, providing some clues to decipher the treemap representations: on PE not exposed to water (*i.e.*, "Dry"), 100% of the marks processed with VMD_{Ag} were characterized by yellow-orange ridges on a purple background; on dry PVC (dry glass), 50% (ca. 60%) of the marks were characterized by the same contrast and 50% (ca. 40%) were of yellow-orange ridges on blue background. When PE exposed to water (*i.e.,* "Wetted") was processed with VMD_{Ag}, various contrasts were obtained among which purple ridges on colorless background (ca. 30%), yellow-orange ridges on purple (ca. 20%), light yellow on purple (ca. 20%), and light blue on purple (ca. 20%) to cite the four main classes. The other rectangles (and sub-rectangles) should be read on the same basis. When referring to Figures 6 to 8, it appears that monometallic VMDs applied to dry substrates generally result in one or two contrast configurations: "yellow-orange ridges on purple/blue background" for VMD_{Ag} and "yellow-orange/brownish ridges on blue/purple background" for $VMD_{Sterling}$ (Figures 6 and 7). However, when the substrates bearing fingermarks are immersed in water, the set of tonalities is increased (Figures 6 and 7). It should also be noted that: (i) some cases of reverse developments (*i.e.*, stained ridges on light background) were observed with VMD_{Ag} and $VMD_{Sterling}$ on wetted PE; (ii) purple ridges on light background were observed with VMD_{Ag} for wetted PE only; (iii) blue ridges on light background were observed $VMD_{Sterling}$ for wetted PE only; (iv) if all fingermarks left on dry substrates were detected, some wetted samples resulted in no ridge detection, especially on glass (crosses in Figures 6 and 7).

Regarding $VMD_{Au/Zn}$, reverse development was systematically obtained with the dry PE bags (Figure 8). For wetted PE, a range of mixed developments were obtained (*e.g.*, partially stained ridges, dark ridges on dark background). For all other substrates and conditions (dry or wet), a normal contrast was obtained.

Discussion

On overall, our results are in good agreement with the studies dedicated to VMD (applied to dry substrates, for most of them). The color tones obtained with VMD_{Ag} are in accordance with the findings of Philipson and Bleay [8], who have compared $VMD_{Au/Zn}$ with various monometallic VMDs using plastics (among which PVC- and PE-based cling films, uPVC, low-density PE, and high-density PE). Yellow ridges on a pink/purple/blue background were obtained with VMD_{Ag}. They explained the difference between purple and blue background by the quantity of silver used during the detection process [8]. A purple background was associated with an optimal detection process, while a blue background may be the indication of an over-development. Yellow ridges on purple background were also obtained by Lucius when VMD_{Ag} was applied on glass [7]. No information being published about $VMD_{Sterling}$ yet, it is only possible to discuss the fact that its behaviour is quite close to VMD_{Ag}, which is awaited given that sterling silver is mostly composed of silver.

On PVC and glass, $VMD_{Au/Zn}$ resulted in normal development (*i.e.*, metallized substrate and transparent ridges), as expected. Reverse contrast was observed with the PE-based garbage bags only (Figure 8). In their study, Jones *et al.* observed that reverse development occurred with $VMD_{Au/Zn}$ on LDPE, but never on HDPE [23]. The same observation was made by Grant *et al.* when processing drug-related PE plastic bags [24]. In regards with the obtained results, we can emit the supposition that our PE-based garbage bags were made of LDPE. The poor performances of $VMD_{Au/Zn}$ on PVC (*i.e.*, average value close to 1 in Study 1, after immersion, and close to 0 in Study 3) were mostly due to empty marks. This phenomenon was reported in the literature [7, 8, 19]. In their study, Jones *et al.* discussed this phenomenon for PET and PVC [19]. They explain the obtaining of empty marks by the fact that a fraction of the secretion residue may migrate into the inter-ridge area, preventing the condensation of zinc. They add that it is more likely to occur with fresh and rich fingermarks. The fingermarks used in this study were not artificially enriched and were aged for 24 hours to 1 week before being processed. We are consequently not strictly speaking about "fresh and rich" secretion residue. Nevertheless, fingermarks resulting from a controlled deposition may lead to fingermarks richer than those obtained in pseudo-operational trials or in caseworks. About empty marks, Philipson and Bleay reported that ridge details could be retrieved if VMD_{Ag} is applied subsequently to $VMD_{Au/Zn}$ [8]. This option has not been explored in this study. On overall, these observations reflect the difficulty to detect fingermarks on PVC using $VMD_{Au/Zn}$, whereas monometallic VMDs seem more appropriate (quality scores close to 2).

- Impact of the immersion on the quality of the fingermarks

The quality scores on dry substrates are close to 2 on average, for all three VMDs (Figure 2), meaning that ridges are visible on almost the whole area of the marks and that second-level characteristics can be retrieved. In Study 1, it was observed that immersion in water resulted in a negligible impact or in a limited quality decrease (Table 3). This observation is in good agreement with the secretion residue fraction that is supposedly involved in the VMD detection mechanism, that is, the non-water-soluble fraction of the secretion residue (NSW fraction) and more specifically the lipids. The limited impact of immersion on the performance of VMDs is consequently explained by the fact that the NWS fraction (and lipids) persist after immersion. This is quite logic since VMD is known for its ability to process wetted items. However, the non-negligible ratio of undetected marks (especially on glass – Figures 6 to 8) indicates that immersion remains a detrimental event and that secretion residue can still be washed out during the process. In their study, Nic Daéid *et al.* reported the use of $VMD_{Au/Zn}$ and white powder suspension (WPS) to process wetted non-porous dark substrates [26]. 24H-Old natural fingermarks were immersed for 6 hours in still tap water, dried and further aged (from 2 days to 28 days) before being processed with $VMD_{Au/Zn}$ and WPS. Comparable results were obtained for both techniques, with recovery rates varying according to the substrates (*i.e.*, "identifiable" marks recovered by $VMD_{Au/Zn}$: 86% of for sandwich bags, 66% for black bin bags, 58% for carrier bags, and 18% for cowlings). It should be noted that most of the development contrasts were normal, in accordance with our observations with immersed items (Figure 8). Finally, no clear trend was obtained regarding the age of the fingermarks upon immersion. One could think that older marks would be more resistant to immersion (due the hardening of the secretion residue). However, as illustrated in Figure 3, it is not the case for all the substrates/experiments. This observation should however be weighted by the fact that the differences are less than one unit (+ or -) and that 1-day-old half marks were compared to 1-week-old ones, limitating by the same way the impact of the age. Extended research are required if this aspect is to be investigated.

- Impact of the immersion on the contrast/color tones

The impact of immersion on color tones (VMD_{Ag} and $VMD_{Sterling}$) and contrast ($VMD_{Au/Zn}$) is certainly the most stricking observation of this study. Indeed, pronounced changes of tonalities were observed for monometallic VMDs on all substrates, as well as a contrast reversal on wetted PE (Figures 6 and 7). For $VMD_{Au/Zn}$, contrast reversal was also observed for wetted PE (Figure 8). As a matter of fact, the immersion step modified the secretion residue and/or the underlying substrates in such a way that it affected the VMD outcome. These observations are extremely valuable for they have not been reported in the literature yet. The mechanisms leading to the condensation of metallic vapour atoms under vacuum constitute a starting point to try bringing elements of answers. First, VMD is a physical vapour deposition process. Readers interested in detailed information about PVD can refer to [30]. Briefly: such process starts with the vaporization of solid metal under vacuum, using high temperature, followed by a rectilinear motion of the vapour atoms in the vacuum chamber and their condensation on the target surface. The formation of a metal film on a surface is the result of three successive steps [31]: (1) condensation (after impact and heat releasing), (2) nucleation into clusters, and (3) film growth. An incident vapour atom reaching the surface can be reflected from its impact location, be physically or chemically adsorbed on the surface, or associate itself with atoms already present to create metal clusters (involving some lateral migration). Consequently, a metal cluster appears only if the right energetic conditions are met. Otherwise, an incident atom or an existing cluster may desorb the surface. The main parameter driving the stabilization of an incident atom or existing cluster is consequently the surface energy [31]. If the surface energy is not adequate, the formation of a metal film may be prevented or deeply impacted. This explains why most of the traditional vacuum coating processes (e.g., to produce reflective metal films or anti-reflection coatings on glass) are carried out on cleaned surfaces, ensuring a uniform and predictable coating. The presence of contaminants (e.g., fats/grease, salt) can substantially modify the surface energy and hence locally modify the rate of growth and the size of stable clusters. In that context, different parameters have been identified as having a major role in the growth and structure of a metal film [31], among which: the nature of the substrate, the presence of impurities or defects on the surface, and the presence of electrostatic charges. In the field of fingermark detection, the target surface is not "clean" as it encompasses the substrate bearing secretion residues. In addition to that, some substrates can be treated during the manufacturing process (e.g., silicon release agents or added properties, such as facilitated surface printing and stability to UV). The difference in surface energy between the substrate and the fingermark explains the ability of VMD to detect fingermarks. This explanation is in agreement with Jones' observations [19], who characterized the deposition of gold and zinc on plastic-based items in terms of clusters and gold counts. Gold clusters of the adequate size and density can be formed on the substrate (e.g., plastic) and not necessarily on the secretion residue, leading to (or preventing) the subsequent deposition of zinc. The display of colours reflected by a metal film can also be influenced by the cluster sizes, the presence of defects, and the film thickness [32], mostly due to Rayleigh or Mié scattering. It explains the difference of colors between the substrate and the fingermarks observed for monometallic VMDs. Regarding this study, water immersion can consequently be seen as a major event affecting both the substrate and the secretion residue. The solubilization of impurities or of water-soluble components from the secretion residue and coated surfaces (such as PE) induces a modification of their composition and of their surface energy. When comparing both situations (i.e., non-wetted items and items having been wet), the respective items are modified in such a way that the formation of metal films is impacted as well as the resulting colours. These observations assuredly constitute supplemental data towards a better understanding of the way the vaporized metals interact with the fingermarks and the underlying substrates.

- IFRG Guidelines

The aim of this study was to explore the impact of immersion on VMD-processed items. The limiting parameters were: the size of the VMD chamber (VMD 360 from West Technology; max useable area: 28.5 cm x 48.0 cm), the fact that three different VMDs were compared (which requires to modify the heating boats accordingly), and the will to provide a methodology fulfilling as best as possible the recommendations of the International Fingerprint Research Group (IFRG) regarding fingermark detection [27]. In regards with these latest, were considered: only natural secretions (to avoid any bias caused by artificial enrichment), fresh and older marks (1-day-old and 1-week-old), split marks (to allow a direct comparison), three different donors (maximum possible in regards with the workload), fingermark replicates, and two independent assessors. No sensitivity assessment was scheduled, explaining why no depletion series were considered in this study.

About the assessors, it should be noted that they were consistent in their scoring (performed independently from each other): 72.3% of their grades were identical for a given half mark, and 99.4% of their grades were contained in a ± 1 interval. These values are in accordance with the findings of Fritz *et al.* about the reliability of assessors in a fingermark grading process [33]. Asking 11 evaluators to assess 80 fingermarks, they observed that 67% of the scores were the same as the median grade and 99% within 1 unit. The concordance between assessors in this study is worth being cited.

Conclusions

This study aimed at gathering information about the impact of immersion on the performance of three kinds of VMDs: $VMD_{Au/Zn}$ (conventional), VMD_{Ag} and $VMD_{Sterling}$ (monometallic). The methodology was based on the use of half marks which allowed a direct comparison between two distinct situations (*e.g.*, dry *vs* wetted, fresh *vs* old marks, VMDx *vs* VMDy). All the processed marks were quantitatively and qualitatively characterized (quality score and color shades/contrast, respectively). As a result, it was shown that the immersion has a limited impact on the quality of fingermarks (no impact or slight degradation), but a more pronounced effect on their appearance (change of color tones/contrast between dry and wetted halves). Additionally, monometal VMDs were shown to be more adapted to the processing of PVC, as opposed to $VMD_{Au/Zn}$. Overall, the obtained results are in agreement with what has been published about VMD so far (recalling that a majority of these papers deal with dry substrates).

This study brings original and valuable data to the community: impact of the immersion on the quality/contrast of fingermarks left on non-porous substrates, use of monometallic VMD on wetted items, use of monometallic VMD based on sterling silver. Even if this study could not provide answers to all the emphasized effects, it participates to the strengthening of the fingermark detection field. It is expected that these results will provide a step forward towards a better understanding of the VMD detection mechanism.

Figure captions

Figure 1 – (a) Illustration of the three main results obtained with $VMD_{Au/Zn}$ - From left to right: normal contrast (substrate = glass), reverse contrast (substrate = polyethylene), and hollow mark (substrate = polyvinylchloride); b) Example of color shades that may result from the application of monometallic VMDs: VMD_{Ag} (substrate = glass) and $VMD_{Sterling}$ (substrate = polyvinylchloride).

Figure 2 – Radar representation linked to Study 1. Each radar map is associated with one kind of VMD (top labels). The reported values are the averaged scores obtained from the half marks associated with the different configurations of substrates (*i.e.*, polyethylene – PE, polyvinylchloride – PVC, and glass) and adverse conditions (*i.e.*, dry, immersed for 1H, and immersed for 24H). For details about Study 1, please refer to Table 1.

Figure 3 – Chart linked to Study 2, reporting the difference of scoring (average) associated with 1-week-old half-marks (1W-old) compared to the corresponding 1-day-old ones (1D-old). For details about Study 2, please refer to Table 1.

Figure 4 – Chart linked to Study 3, reporting the averaged scores obtained from the half marks associated with the different configurations of substrates (*i.e.*, polyethylene – PE, polyvinylchloride – PVC, and glass) and VMDs (*i.e.*, VMD_{Ag}, $VMD_{Sterling}$, and $VMD_{Au/Zn}$). For details about Study 3, please refer to Table 1.

Figure 5 – Non-exhaustive set of contrasts and color shades observed during these studies: (a) VMD_{Ag} (PE, dry) vs VMD_{Ag} (PE, wet-1H), (b) $VMD_{Sterling}$ (PE, dry) vs $VMD_{Sterling}$ (PE, wet-1H), (c) $VMD_{Au/Zn}$ (PE, dry) vs $VMD_{Au/Zn}$ (PE, wet-1H), (d) VMD_{Ag} (PE, wet-24H) vs $VMD_{Sterling}$ (PE, wet-

24H), (e) VMD_{Ag} (glass, wet-24H) vs $VMD_{Sterling}$ (glass, wet-24H), (f) $VMD_{Sterling}$ (glass, 1D-old, wet-24H) vs $VMD_{Sterling}$ (glass, 1W-old, wet-24H).

Figure 6 – Treemap representation of the color shades observed after processing the fingermarks with VMD_{Ag}. Notes: the areas are proportional to the % of marks sharing the illustrated contrast (whole rectangle area = 100%); fingermark icon made by Freepik from www.flaticon.com.

Figure 7 – Treemap representation of the color shades observed after processing the fingermarks with $VMD_{Sterling}$. Same remarks as for Figure 6.

Figure 8 – Treemap representation of the contrasts observed after processing the fingermarks with $VMD_{Au/Zn}$. Same remarks as for Figure 6.

Table captions

Table 1 – Experimental details of the three studies. The total number of fingermarks considered for each study (*) is obtained by multiplying the number of comparative studies with the number of donors (3), the number of substrates (3), the VMDx considered (if applicable), and the number of replicates (*i.e.*, number of fingermarks a donor is asked to leave for a specific comparison).

Table 2 – Quantitative scale used to characterize the quality of detection of all marks processed in this study. Source: [28]

Table 3 – Summary table of the conclusions of Studies 1 to 3. The symbols correspond to an increase of efficiency/score (↗), a decrease (↘), or similar values/no effect (≅) in regards with the purpose of the study (top labels). PE and PVC stand for polyethylene and polyvinylchloride, respectively. For details about each study, please refer to Table 1.

Bibliography

1. Champod C., C. Lennard, P. Margot, and M. Stoilovic, *Fingerprints and Other Ridge Skin Impressions - Second Edition*. 2016, Boca Raton, Florida: CRC Press LLC. 427.
2. Theys P., Y. Turgis, A. Lepareux, G. Chevet, and P.F. Ceccaldi, *Nouvelle technique de révélation de traces papillaires latentes (sur le papier) par métallisation sous vide.* Revue Internationale de Police Criminelle [International Criminal Police Review], 1968. **23**: p. 106-108.
3. Kent T., G.L. Thomas, T.E. Reynoldson, and H.W. East, *A Vacuum Coating Technique for the Development of Latent Fingerprints on Polythene.* Journal of the Forensic Science Society, 1976. **16**(2): p. 93-101.
4. Gaskell C., S.M. Bleay, H. Willson, and S. Park, *The Enhancement of Fingermarks on Grease-Contaminated, Nonporous Surfaces: A Comparative Assessment of Processes for Light and Dark Surfaces.* Journal of Forensic Identification, 2013. **63**(3): p. 286-319.
5. Knighting S., J. Fraser, K. Sturrock, P. Deacon, S. Bleay, and D.H. Bremner, *Visualisation of Fingermarks and Grab Impressions on Dark Fabrics using Silver Vacuum Metal Deposition.* Science and Justice, 2013. **53**: p. 309-314.
6. Dominick A.J., N. Nic Daéid, and S.M. Bleay, *The Recoverability of Fingerprints on Nonporous Surfaces Exposed to Elevated Temperatures.* Journal of Forensic Identification, 2011. **61**(5): p. 520-536.
7. Lucius C., *Single-metal vacuum deposition: An alternative to the traditional Gold/Zinc process?* Master degree in Forensic Science; École des Sciences Criminelles, University of Lausanne. Lausanne-Dorigny (Switzerland). 2008, pp. 70.
8. Philipson D. and S. Bleay, *Alternative metal processes for vacuum metal deposition.* Journal of Forensic Identification, 2007. **57**(2): p. 252-273.
9. Davis L.W.L., P.F. Kelly, R.S.P. King, and S.M. Bleay, *Visualisation of latent fingermarks on polymer banknotes using copper vacuum metal deposition: A preliminary study.* Forensic Science International, 2016. **266**: p. e86-e92.
10. Gunaratne A., C. Knaggs, and D. Stansbury, *Vacuum metal deposition: Comparing conventional gold/zinc VMD to aluminium VMD.* Identification Canada, 2007. **30**(2): p. 40-62.
11. Lam R., D. Wilkinson, T. Tse, and B. Pynn, *Recommended Protocols for Fingerprint Detection on Canadian Polymer Banknotes - Part I: Chemical Development.* Journal of Forensic Identification, 2014. **64**(4): p. 375-401.
12. Jones N., M. Kelly, M. Stoilovic, C. Lennard, and C. Roux, *The Development of Latent Fingerprints on Polymer Banknotes.* Journal of Forensic Identification, 2003. **53**(1): p. 50-77.
13. Fraser J., P. Deacon, S. Bleay, and D.H. Bremner, *A comparison of the use of vacuum metal deposition versus cyanoacrylate fuming for visualisation of fingermarks and grab impressions on fabrics.* Science and Justice, 2014. **54**: p. 133-140.
14. Fraser J., K. Sturrock, P. Deacon, S. Bleay, and D.H. Bremner, *Visualisation of Fingermarks and Grab Impressions on Fabrics. Part 1: Gold/Zinc Vacuum Metal Deposition.* Forensic Science International, 2011. **208**: p. 74-78.
15. Bhoelai B., B.J. de Jong, M. de Puit, and T. Sijen, *Effect of Common Fingerprint Detection Techniques on Subsequent STR Profiling.* Forensic Science International: Genetics, 2011. **3**(1): p. e429-e430.

16. Raymond J.J., C. Roux, E. Du Pasquier, J. Sutton, and C. Lennard, *The effect of common fingerprint detection techniques on the DNA Typing of fingerprints deposited on different surfaces.* Journal of Forensic Identification, 2004. **54**(1): p. 22-44.
17. Home Office - Centre for Applied Science and Technology (CAST), *Fingermark Visualization Manual*, Bandey H., Editor. 2014.
18. Jones B.J., R. Downham, and V.G. Sears, *Nanoscale Analysis of the Interaction Between Cyanoacrylate and Vacuum Metal Deposition in the Development of Latent Fingermarks on Low-Density Polyethylene.* Journal of Forensic Sciences, 2012. **57**(1): p. 196-200.
19. Jones N., D. Mansour, M. Stoilovic, C. Lennard, and 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 Science International, 2001. **124**(2-3): p. 167-177.
20. Jones N., M. Stoilovic, C. Lennard, and C. Roux, *Vacuum metal deposition: developing latent fingerprints on polyethylene substrates after the deposition of excess gold.* Forensic Science International, 2001. **123**(1): p. 5-12.
21. Dai X., M. Stoilovic, C. Lennard, and N. Speers, *Vacuum metal deposition: Visualisation of gold agglomerates using TEM imaging.* Forensic Science International, 2007. **168**(2-3): p. 219-222.
22. Downham R.P., S. Mehmet, and V.G. Sears, *A Pseudo-Operational Investigation into the Development of Latent Fingerprints on Flexible Plastic Packaging Films.* Journal of Forensic Identification, 2012. **62**(6): p. 661-682.
23. Jones N., M. Stoilovic, C. Lennard, and C. Roux, *Vacuum metal deposition: factors affecting normal and reverse development of latent fingerprints on polyethylene substrates.* Forensic Science International, 2001. **115**(1-2): p. 73-88.
24. Grant H., E. Springer, and Z. Ziv. *Vacuum Metal Deposition lnhibition on Polythene Bags*. in *International Symposium on Fingerprint Detection and Identification*. 1995. Ne'urim (Israel).
25. Hilgert M. *Standardization and optimization of the VMD-method for the development of latent fingermarks*. in *ENFSI Fingerprint Working Group (EFPWG)*. 2011. Riga (Latvia).
26. Nic Daéid N., S. Carter, and K. Laing, *Comparison of vacuum metal deposition and powder suspension for recovery of fingerprints on wetted nonporous surfaces.* Journal of Forensic Identification, 2008. **58**(5): p. 600-613.
27. International Fingerprint Research Group (IFRG), *Guidelines for the Assessment of Fingermark Detection Techniques.* Journal of Forensic Identification, 2014. **64**(2): p. 174-200.
28. Moret S. and A. Bécue, *Single-Metal Deposition for Fingermark Detection - A Simpler and More Efficient Protocol.* Journal of Forensic Identification, 2015. **65**(2): p. 118-137.
29. Wikipedia. *Treemapping - Wikipedia, The Free Encyclopedia*. 2017 Online; accessed November 1, 2017]; Available from: https://en.wikipedia.org/w/index.php?title=Treemapping&oldid=804759138.
30. Mattox D.M., *Chapter 6 - Vacuum Evaporation and Vacuum Deposition*, in *Handbook of Physical Vapor Deposition (PVD) Processing (Second Edition)*. 2010, William Andrew Publishing: Boston. p. 195-235.
31. Chaliha S., *Studies of ZnSe and indium tin oxide based thin film schottky barriers and heterojunctions for their electrical and optical properties.* Doctor of Philosophy; Department of Physics, Gauhati University. 2008, pp.
32. Furrer A., *Colours in thin metallic films based on precious metals and their intermetallic phases.* Doctor of Sciences, ETH Zurich. 2013.

33. Fritz P., A.A. Frick, W. van Bronswijk, S.W. Lewis, A. Beaudoin, S. Bleay, and C. Lennard, *Variability and Subjectivity in the Grading Process for Evaluating the Performance of Latent Fingermark Detection Techniques.* Journal of Forensic Identification, 2015. **65**(5): p. 851-867.

Figure 1

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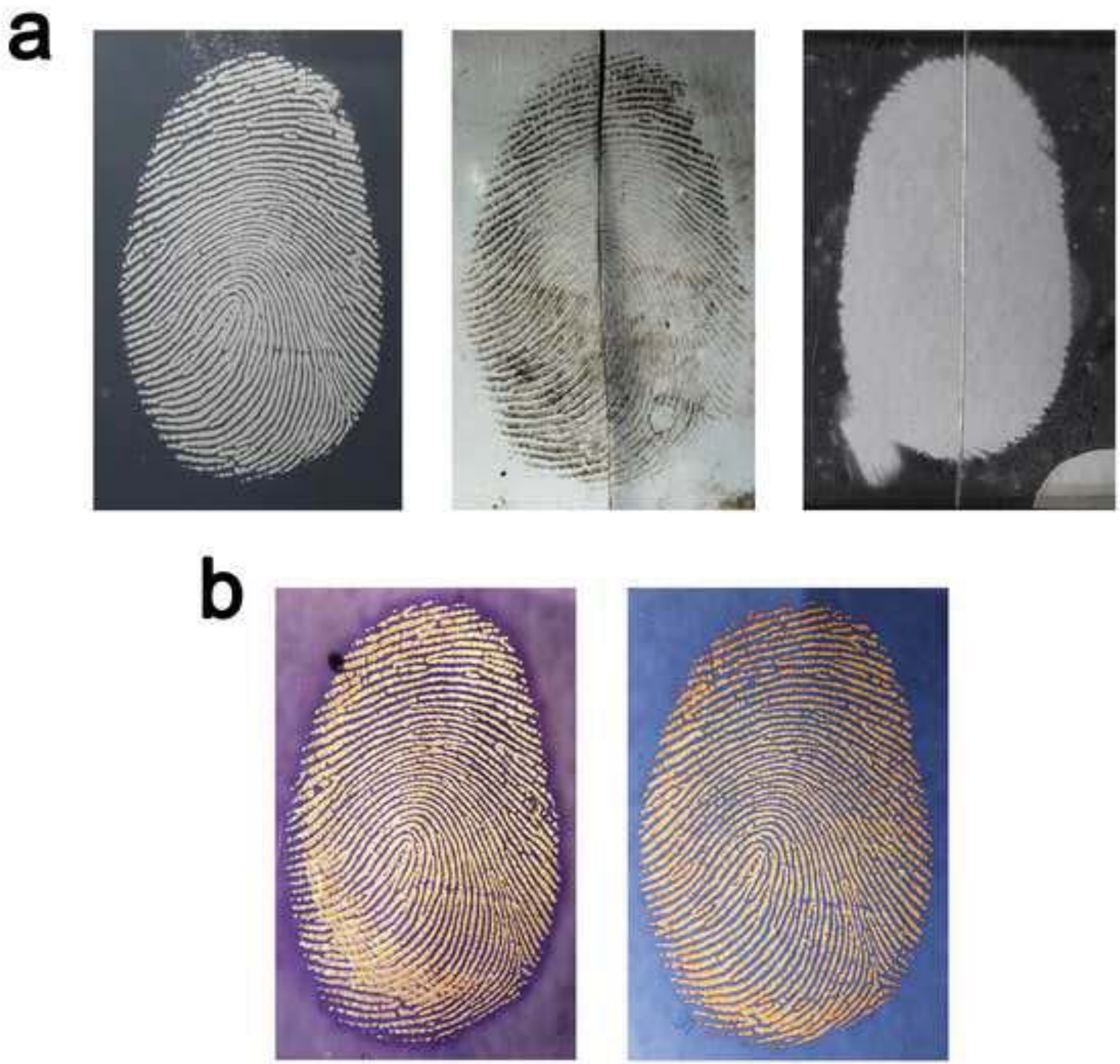

Figure 2
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Figure 3

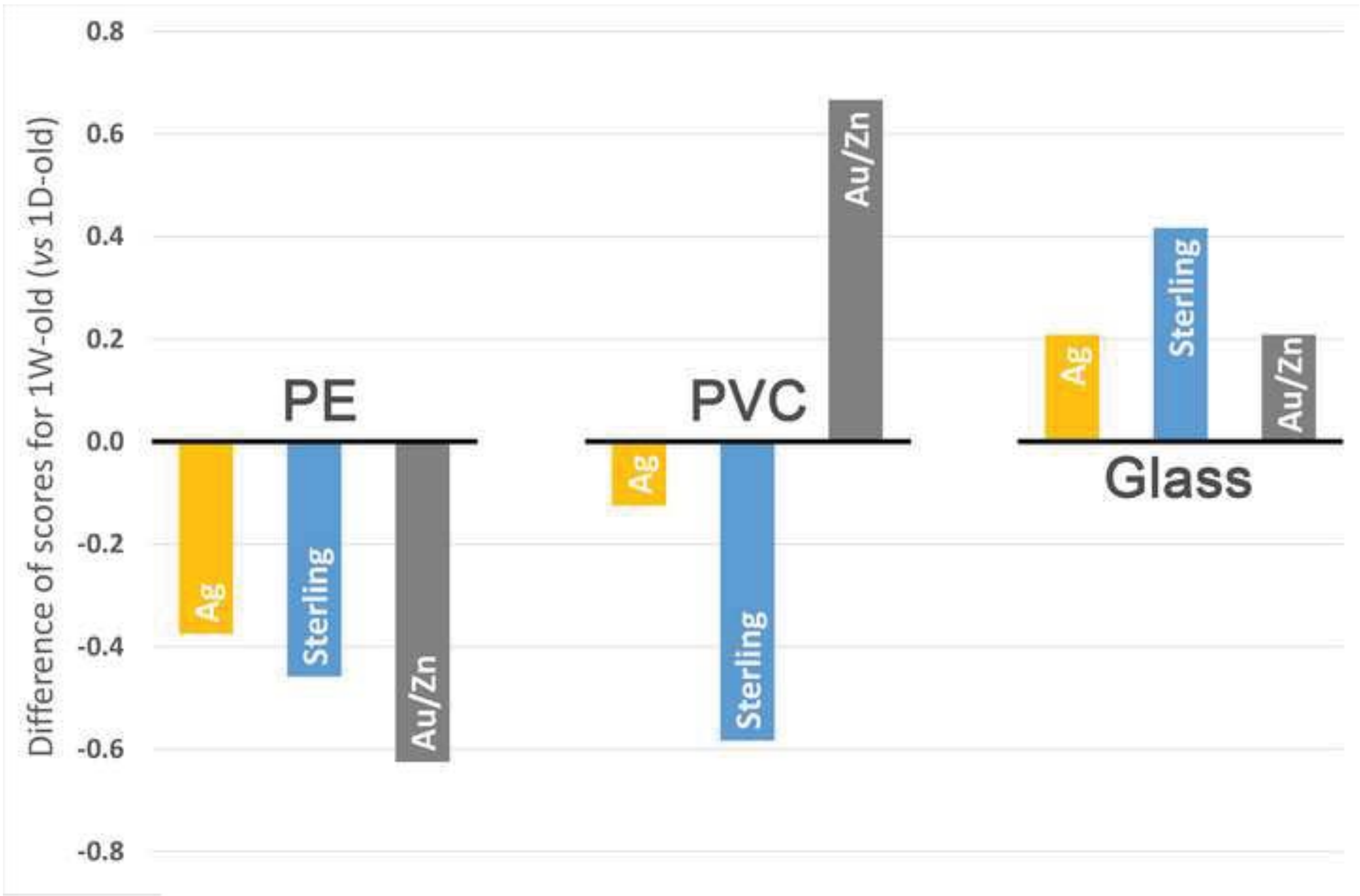
Difference of scores for 1W-old (vs 1D-old)
0.8
0.6
0.4
0.2
0.0
-0.2
-0.4
-0.6
-0.8
PE
Ag
Sterling
Au/Zn
PVC
Ag
Sterling
Au/Zn
Glass
Ag
Sterling
Au/Zn

Figure 4
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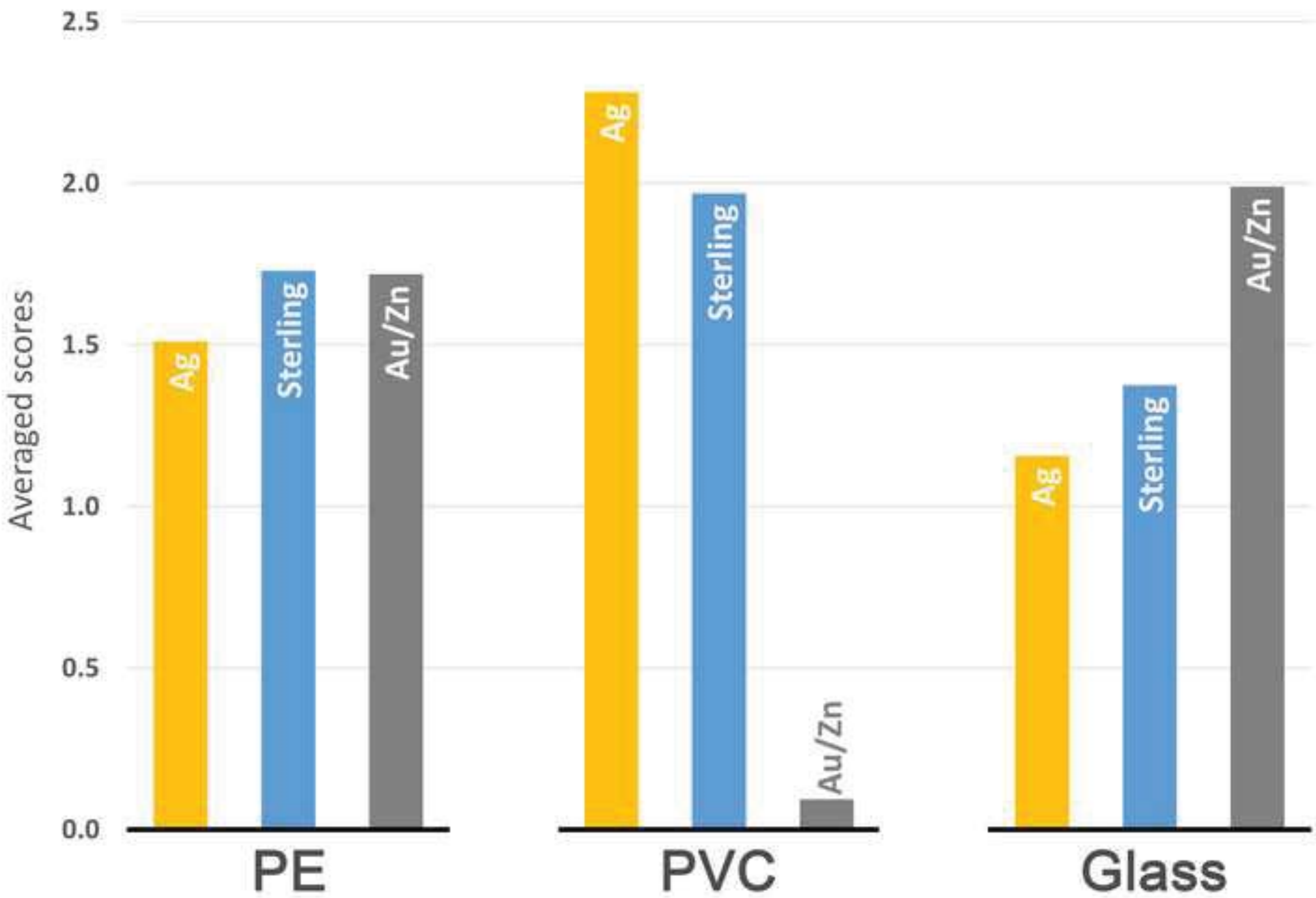

Figure 5

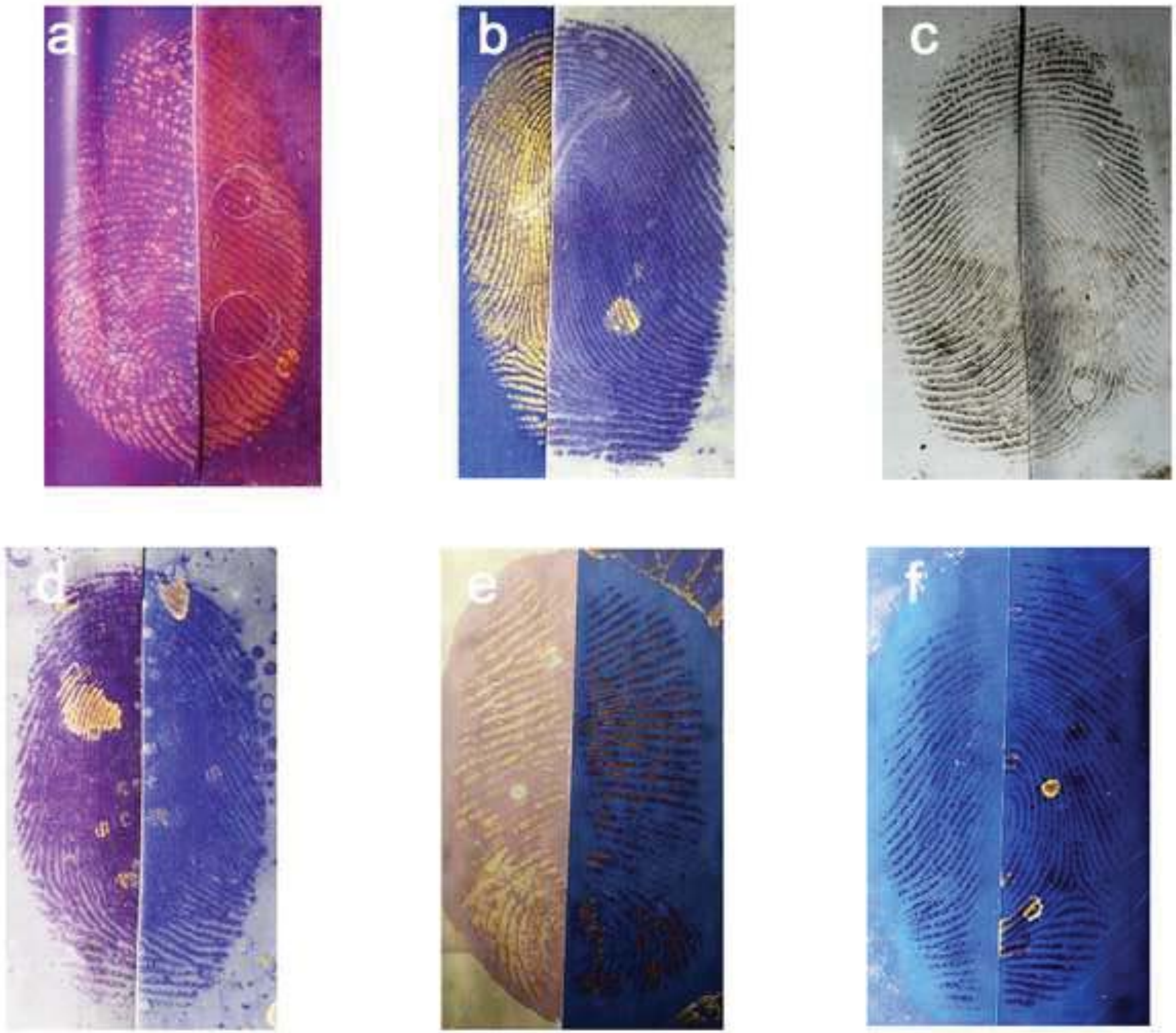

Figure 7

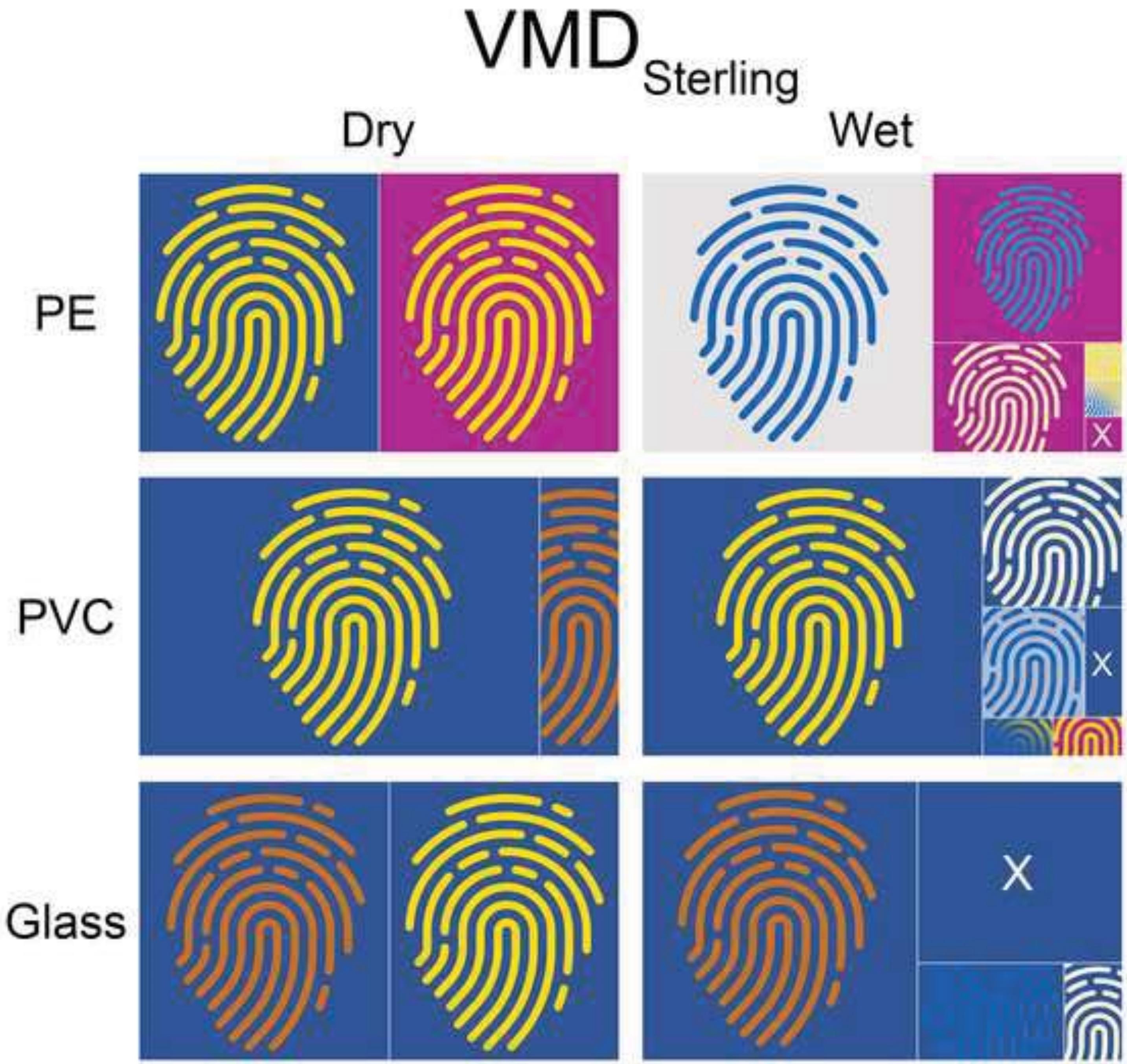

Figure 8

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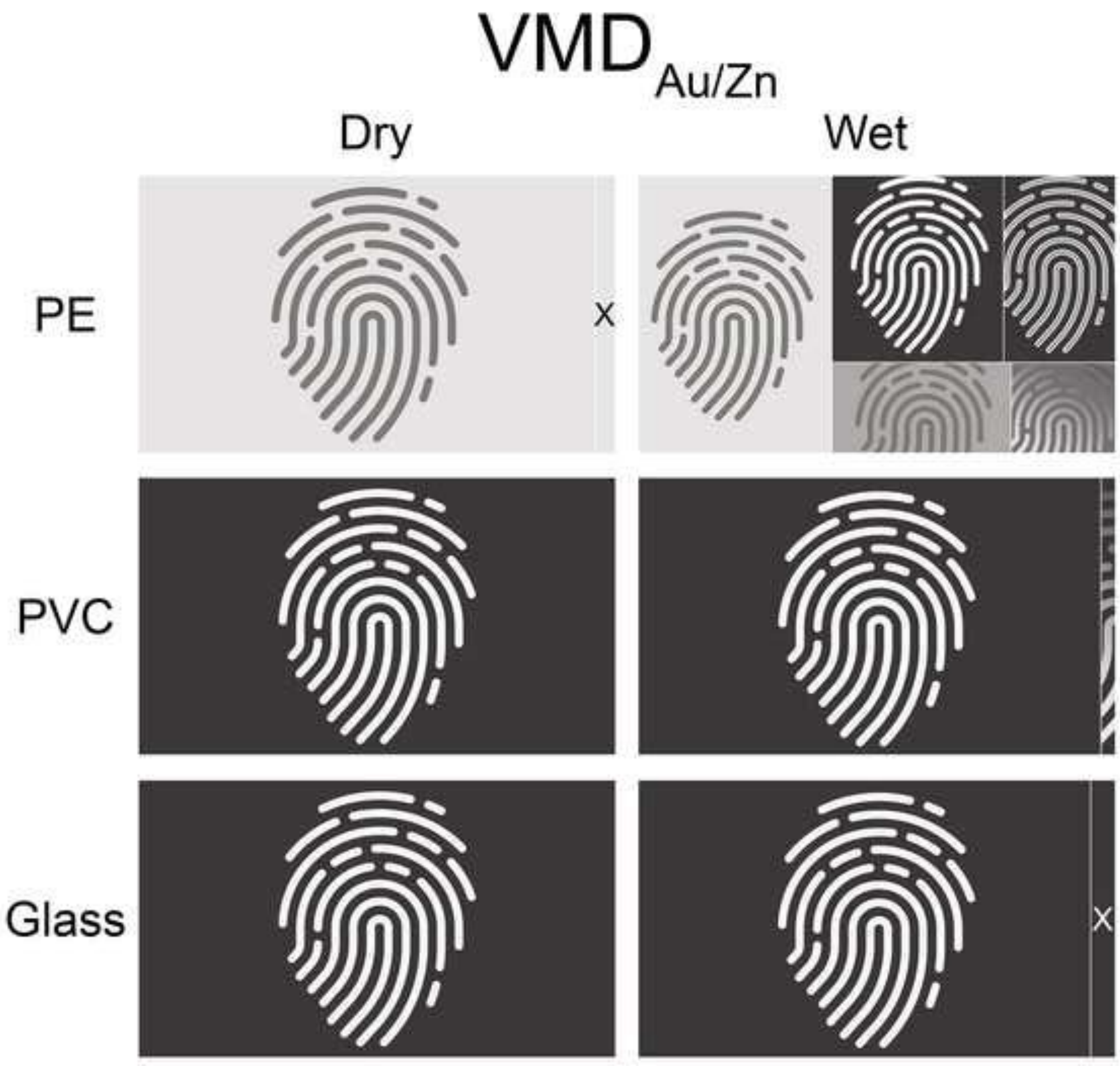

Figure 6
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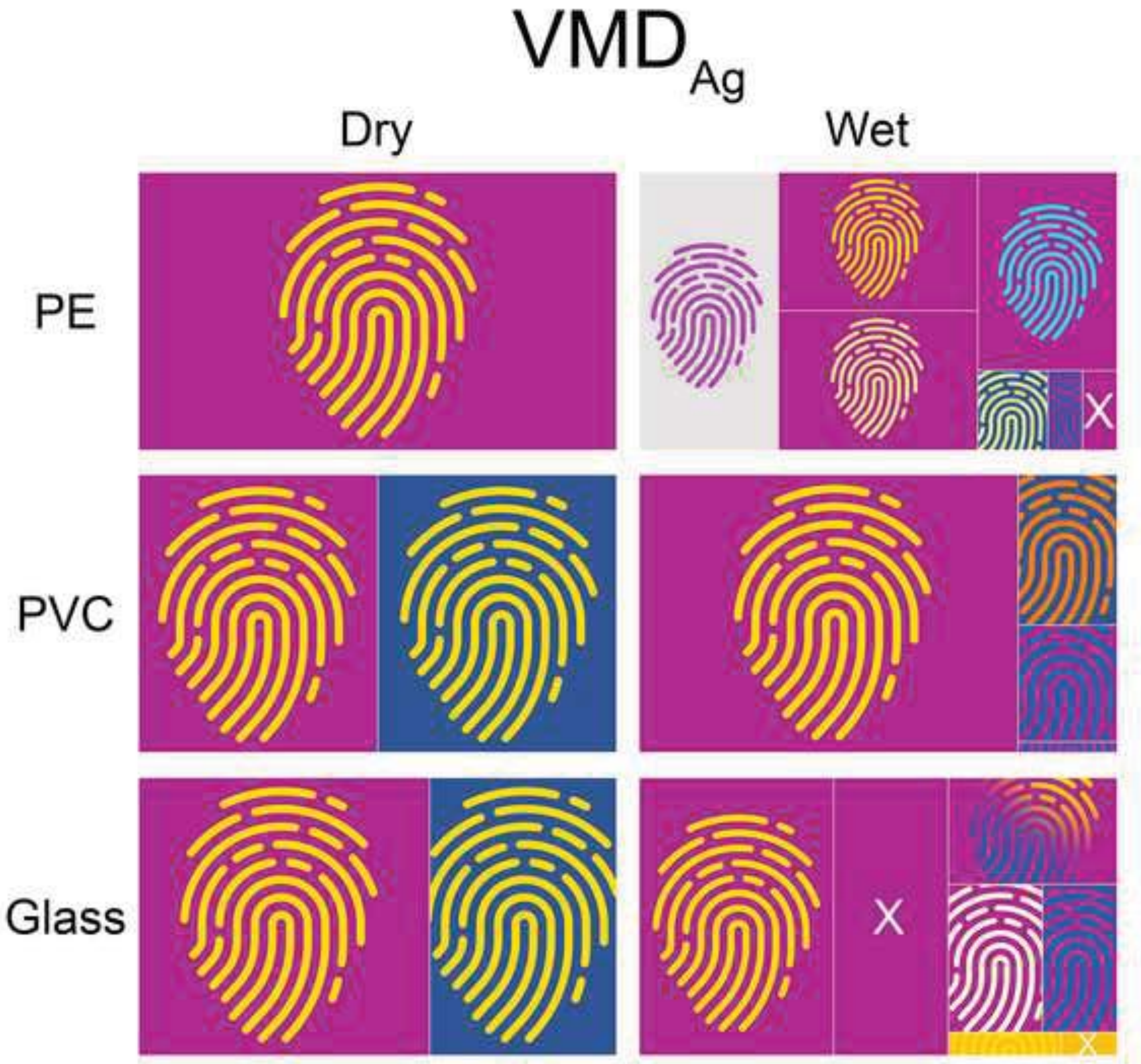

Table 1

Study # / Influence of	**Age of the fingermarks (upon immersion)**	**Immersion time**	**Comparative studies (L-half *vs* R-half)**	**Replicates**	**Total number of fingermarks (*)**	**Total number of half marks**
1/ Immersion time	48H	1H or 24H	Dry (reference) *vs* Immersed (1H) Dry (reference) *vs* Immersed (24H)	4	216	432
2/ Age of the fingermarks	24H or 1 week	24H	"24H" *vs* "1W"	4	108	216
3/ Metal (VMD_x)	48H	24H	Au/Zn *vs* Ag Au/Zn *vs* Sterling Ag *vs* Sterling	8	216	432

Table 2

Score	Description
0	No ridges are visible at all, no sign of fingermark.
1	Ridges are visible over a small area of the mark or over the whole mark, but it is extremely difficult to retrieve second-level characteristics (such as minutiae) due to extremely poor ridge details.
2	Ridges are visible on almost the whole area of the mark, and second-level characteristics can be retrieved. Nevertheless, the quality is not optimal due to a low contrast (strong background staining or faint ridges).
3	Ridges are very well defined on the whole mark. Second-level characteristics can easily be retrieved. The contrast is optimal with no (or extremely faint) background staining.

Table 3

Table 3

Study 1	**Effect of 1H immersion (*vs* Dry)**	**Effect of 24H immersion (*vs* Dry)**
PE	Ag ≅ ; Sterling ≅ ; Au/Zn ≅	Ag ↘ ; Sterling ≅ ; Au/Zn ≅
PVC	Ag ≅ ; Sterling ↘ ; Au/Zn ↘	Ag ↘ ; Sterling ↘ ; Au/Zn ↘
Glass	Ag ↗ ; Sterling ↘ ; Au/Zn ≅	Ag ≅ ; Sterling ↘ ; Au/Zn ≅
Study 2	**Relative scores for 1W-old marks (*vs* 1D-old) (wetted items)**	
PE	Ag ↘ ; Sterling ↘ ; Au/Zn ↘	
PVC	Ag ≅ ; Sterling ↘ ; Au/Zn ↗	
Glass	Ag ↗ ; Sterling ↗ ; Au/Zn ↗	
Study 3	**Comparison between metals (wetted items)**	
PE	Ag ≅ Sterling ≅ Au/Zn	
PVC	Ag ≥ Sterling >> Au/Zn (hollow marks)	
Glass	Ag ≅ Sterling < Au/Zn	