Detection of fingermarks – applicability to metallic surfaces: A literature review

ABSTRACT:

There are many different fingermark visualization techniques available and the choice of methodology employed may be dependent on the surface type. This comprehensive review of the scientific literature evaluates the methodologies of fingermark enhancement methods that are applicable to metallic surfaces; optical, physical, chemical and physico-chemical methods are critically discussed. Methods that are currently used and those that have the potential to reduce the cost and time required to process evidence and increase the recovery rates are considered, and are assessed against the Centre for Applied Science and Technology (CAST) and the International Fingerprint Research Group (IFRG) guidelines. The use of chemical imaging techniques in particular, has increased the potential to recover fingermarks of sufficient quality for identification purposes. Presently, there appears to be a lack of detailed research pertaining to validation and thorough casework studies for fingermark enhancement techniques. Further studies incorporating these guidelines are recommended.

KEYWORDS

Forensic science, Fingermarks, Fingerprints, Enhancement techniques, Metallic Surfaces, Latent Fingermarks

Fingermarks are considered one of the most valuable types of physical evidence recovered from a crime scene. Fingermark casework is still the most common casework for forensic scientists despite the increase in use of DNA(1) to identify/exonerate suspects via bodily fluids(2). The ridge patterns found in fingermarks are permanent and can be used to individualize or exclude suspects from an investigation.

Detection of fingermarks on metallic surfaces will be dependent upon the compounds remaining from the fingermark residue. Several detailed reviews have already been published on fingermark composition, but none have focused on fingermarks deposited on metallic surfaces(3, 4).

Generally, there are two types of fingermarks found at crime scenes: Visible fingermarks, usually formed by fingers contaminated by a substance such as oil, fruit, grease, paint or blood which are deposited on a surface; and latent fingermarks, which are invisible to the naked eye and are the most abundant type of fingermarks at crime scenes. Metallic surfaces are also commonly encountered in crime scenes, and are ubiquitous in the environment. Surfaces such as the door of a car, objects like handles, weapons and tools are usually made of steel. Cartridge cases are frequently recovered from crime scenes and are usually made of brass or nickel(5). There are a number of different techniques that can be applied to a surface to visualize latent fingermarks. The selection of a suitable technique depends on different factors, such as the expected composition of a fingermark, the ability of the chosen technique to be used in tandem with other techniques, and the nature of the substrate. In this review, the focus will be on fingermarks deposited on metallic surfaces, and the optical, physical, chemical and physico-chemical visualization techniques that can be used for detection of fingermarks on these surfaces. The UK Home Office’s grading system (Table 1) is considered when assessing the quality of the development of fingermarks of all the different enhancement methods. It should be noted, that the assessment of the quality of fingermark development, was performed by the authors, based only on the data that was disclosed in each different study. Moreover, the majority of the scientific papers included in this review, studied fingermark development under laboratory conditions, thus may not replicate the ‘real’ conditions seen in actual casework.

Additionally, in order to provide extra information about the stage of the fingermark study of each individual paper examined in this review, the International Fingerprint Research Group (IFRG) guidelines will be introduced and used when reviewing a study. Specifically, there are four main research phases in fingermark research (7):

- **Phase 1 (Pilot Study)** involves initial pilot or proof-of-concept investigations of novel fingermark detection methods (reagents or techniques) or major modifications to existing methods. These projects are often the domain of universities and dedicated government research facilities.

- **Phase 2 (optimisation and comparison study)** is a more detailed investigation and evaluation of a method. The optimisation of relevant parameters is generally a first step in this phase. The relative performance of the new or modified method then needs to be compared to that of established operational techniques and the performance of the method across a number of variables (substrates, donors and ageing periods, for example) assessed under reasonably controlled conditions. Consideration may also be given to how the new method performs in sequence with relevant routine detection techniques. Phase 2 projects may be undertaken by universities, government research facilities, or operational casework facilities.

- **Phase 3 (Validation)** studies are designed to introduce successfully optimised techniques to more realistic, pseudo-operational scenarios using simulated casework material. This phase is a rigorous evaluation of the performance of the new technique against current methods in order to assess suitability for potential operational use. The position of the new method in relevant detection sequences must also be thoroughly tested as part of the validation. Phase 3 research may be done by universities or government research agencies but should at least be undertaken in close collaboration with an operational casework facility.

•**Phase 4 (operational, evaluation and casework trials)** focuses on eventual casework implementation via inclusion into standard operating procedures (SOPs). Phase 4 must be undertaken as a live casework trial by an operational facility intending to introduce the method. For accredited facilities, Phase 4 evaluations should be undertaken in a manner that will facilitate the subsequent formal method validation processes required to meet relevant international standards (e.g., ISO 17025). A typical Phase 4 project includes the assessment of a new technique across a large number of cases – and possibly across a large number of laboratories in the case of national agencies or geographically broad jurisdictions – during a designated trial period. During this period, the performance of the new technique is compared to the performance of current methods. For studies undertaken across multiple locations, the ambient laboratory conditions can differ significantly and may impact on the results. If this can be an issue, it is recommended that the temperature and humidity in each laboratory and storage conditions be recorded. If field-based methods are being evaluated, environmental conditions should be recorded to determine if these may be impacting on the results achieved. This information can be invaluable for assessing and documenting the robustness of a new technique.

The IFRG guidelines also discuss parameters such as number of donors, substrates, donation variables for each study phase and assessment methodologies (7).

Optical fingermark development methods

Optical fingermark development methods can detect a fingermark without the use of any other chemical enhancement technique. This is possible by exploiting the different optical behaviour of the fingermarks compared to the substrate they are deposited on(8).The advantage of these methods is that they can provide information about the composition and the morphology of a fingermark simultaneously. These already established fingermark visualization techniques revolve around exciting certain compounds present in the latent fingermark in a narrow range of wavelengths and then detecting the data at one specific wavelength. Chemical imaging can analyse an image into its component colours at many different wavelengths quantitatively. This feature enables the forensic examiner to discriminate usable information from a background interference pixel-by-pixel. Unwanted interference including fluorescence, texture, and colours can be efficiently minimized, leading to a detailed fingermark image. Although these techniques may be considered complex, there is no relevant literature available on simpler optical methods in relation to metallic surfaces.

Infrared reflection/Infrared imaging

Infrared imaging is a process in which the (latent) fingermark and the surface upon it is deposited are illuminated with infrared radiation and the outcome is captured by an infrared sensitive viewing system. When a substrate is illuminated, the substrate obtains a light color whilst the fingermark has a black/dark color(8). It is a non-destructive procedure that can provide information about the morphology and the composition of a fingermark on almost any surface (except for substrates that absorb infrared light). The visualization of the fingermark is also dependent on the fingermark itself and on whether other enhancement techniques have been used prior to Infrared imaging. For instance, a fingermark deposited on a metallic surface which is untreated (the fingermark), or can be enhanced with deposition of a metal containing powder is an ideal candidate for this method. Fourier Transform Infrared Spectroscopy (FTIR) imaging has the added benefit of obtaining better images of the fingermarks by using various data analysis techniques (i.e. Principle Component Analysis - PCA)(9). Additionally, information about other compounds (drugs, explosives) present on fingermarks can be gathered by using this technique but only in cases where the deposition happened on a flat metal substrate (aluminium coated slides) or on a substrate where the fingermark can be easily lifted(10). Finally, Tahtouh et al.(11) in their study state that this method is also relatively fast, reporting that an experienced user can develop a fingermark within 4 hours.

Ultraviolet reflection

Fingermarks absorb the largest part of incident visible light, while reflecting the largest part of ultraviolet light, a phenomenon which can be exploited especially on smooth non-porous surfaces. This method is especially helpful for fingermark detection on surfaces that have multiple colorations and thus many visual disruptions (i.e. aluminium can).

One disadvantage of this technique is that it damages the DNA present in fingermarks. Gibson et al. state that although the different UV detection procedures can visualize fingermarks, only a small percent around 7-14% of these fingermarks will be of a Home Office's grade of a 3 or 4 (12). Additionally, the visualization quality depended on the illumination angle, which can vary when dealing with different samples and different substrates The UV-C radiation wavelength range used was from 100 to 280 nm and the systems evaluated were: a UV-C-sensitive, back-thinned CCD and camera system , RUVIS (reflected ultraviolet imaging system) which is a UV-C-sensitive image intensifier (254nm) and a flatbed scanner fitted with a UV-C light source. UV reflection has also been used sequentially with specially modified cyanoacrylate. A pilot study by Khuu et al.(13) using one –step luminescent cyanoacrylates, where the authors used a VSC6000 has shown some potential, but further work is still needed.

Fluorescence

Recent research has demonstrated excellent visualization (without damaging the DNA present in a fingermark)(14) on many different metallic substrates (stainless steel sheets, aluminium foil) can be achieved when using a Near Infrared (NIR) radiation source at 980 nm for excitation of latent fingermarks that have been powdered with $NaYF_4$:Yb, Er upconversion nanoparticles. One limitation of this research is that it was only tested with one donor of fingermarks(14). The efficacy of this technique is yet to be determined on a wider range of metallic surfaces. Another study by King et al.(15) capitalizing on fluorescence, yielded good results on metallic surfaces (such as metals used for aerosol cans). A modification of a Cuprorivaite fingermark powder was used (by applying with a brush rotationally) and the image was captured with modified Nikon D810 with the IR filter removed. The excitation wavelength applied was 780 nm and took the form of a 9 × 780 nm LED array, fitted with an 800 nm short-pass filter. Despite the reasonable donor pool in this study (10), the focus was mainly on non-metallic surfaces (banknotes, plastics), which means more data is required before drawing any conclusions.

Raman Imaging

Connatser et al.(16) used Surface Enhanced Raman Spectroscopy (SERS) imaging in an attempt to visualize eccrine latent fingerprint that would otherwise remain undetected (due to exposure to heat or other degrading factor). For their experiments, they also used an artificial fingermark solution, which was made after evaluating all the methods that were available, and creating a single optimized solution. The solution consisted of a large amount of the known components of eccrine sweat. Chemical imaging of fingermarks was based upon the hydrocarbon bond from skin oils at 2900 cm^{-1}. Imaging was successful on high contrast surfaces (including metals). In the same study, a sebaceous fingermark with drug remnants on it was used and successfully detected. Raman Imaging and IR imaging could be used in tandem as they complement one another, but in order for SERS to be used in practice a lot more research has to be done, specifically with a large donor size and the optimization of the enhancement methodology.

Song et al.(17) also used SERS to detect specific biomolecules in latent fingermarks. In their study, they used the aromatic ring vibrations to detect the molecules and obtain clear SERS photos of the fingermarks Although in this study the experiments were not performed on a metal substrate the potential of Raman imaging is clearly demonstrated and the added benefit (compared to fluorescence) is that the spectral bands are narrower allowing for a more precise assignment to the bonds. A performance table (Table 2) provides a summary of the optical methods discussed in this section. Although, in some of the studies examined in this section the surfaces and items that were included resembled items that can be encountered in crime scenes the lack of a large number of donors and fingermarks aged for different time limits make the classification of the research as phase 1 according to IFRG guidelines.

Physical methods

Physical methods are usually methods that employ solid-state reagents and in most cases, the fingermarks are not significantly altered.

Powder methods

The powders used for developing latent fingermarks at a crime scene adhere preferentially to specific components of the fingermarks. The concept of these methods is quite simple; application of the powder to the surface of interest and observation and monitoring of the progress of the deposition(8). With ‘fresh’ fingermarks, the powder adheres to the aqueous components. In aged fingermarks, the adhesion happens mostly due to the fatty acids and other greasy substances present in sebaceous sweat(19).

Powder methods can be divided in to four categories: metal flake, granular, magnetic flake, and magnetic granular. Each have different advantages and disadvantages (work better on different surfaces etc.). A general rule for powders is that they can be detrimental for further analysis, and that the forensic practitioner must know which powder to use on each surface. An advantage of powder use is that their application is simple, thus no extensive training is needed. All of them are routinely used in casework for developing fingermark on metallic surfaces (i.e. car doors)(20). Bandey(20) states that studies have been conducted to find out the suitability of each powder on different (metallic) surfaces. Specifically, brass flake powder should be used on smooth silver surfaces. Aluminum flake powder can also be the powder of choice for certain smooth metal surfaces as it is easy to apply and gives good contrast. Black granular powder can be used on smooth surface only, while the black magnetic powder can also be used on textured surfaces. This study illustrates that there is not one powder that is deemed the most effective across all surfaces, including metals. Recently, with the advances in nanotechnology, hybrid (inorganic and organic) powders are being developed, with the ultimate goal of creating a powder that can develop high quality fingermarks in the majority of the cases, despite the variation in the fingermark itself (in terms of compounds), and the substrate it is deposited on.

Silica based nano-particles

From 2008 and onwards, silica based composites for the development latent fingermarks were developed and studied. In the work of Liu et al.(21) Eu^{+3} was entrapped in a SiO_2 matrix and a series of different sensitizers was used with 1,10-Phenanthroline providing the best results in an array of substrates by dusting. J. Dutta et al.(22) modified latent fingermarks with columnar thin films consisting of SiO_2 and CaF_2. The procedure involved depositing the columnar films with the physical vapor deposition technique and then spin coating with rhodamine 6G solution. Fingermarks on non-porous substrates were developed with this method. Luminescent nanophosphores embedded in silicate matrix have been synthesized and tested on aluminium foil and demonstrated that fingermarks of 7 days old could be developed. Another group of composite compounds are the porous phosphate heterostructures. These developing powders are made of zirconium phosphate on silica galleries. PPH-S-CdS nanocomposite was used to developed latent fingermarks on a steel surface successfully(23). Although the silica based nano-particles seem to adhere well on the ridges of fingermarks on various substrates (including aluminium and steel), their visualization is achieved by exploiting their optical properties (light absorption, photoluminescence), and that is not always feasible in surfaces that their background luminescence is strong.

Lanthanide-based powders

Europium Strontium Aluminate (ESA) Phosphors have revealed long afterglow properties. Liu et al.(21) examined their use in substrates that may fluoresce themselves by using ESA Phosphors and UV-lighting and development of fingermarks was achieved on a range of surfaces. Favorable results were achieved on aluminium foil for both fresh and a few days old fingermarks. Saif(24) examined the use of Ln^{3+}:$Y_2Zr_2O_7/SiO_2$ (Ln^{3+}=Eu^{3+}, Tb^{3+}, Sm^{3+}, Dy^{3+} or Pr^{3+}) for development of fingermarks also on aluminium foil with Terbium (Tb^{3+}) powder found to be the most effective.

The use of this method might be detrimental for DNA evidence examination, which means that it needs to be performed after all DNA related methods in a sequential treatment. Despite the fact that only one donor was used in both studies, the method seems promising, due to its low-cost, low-risk nature along with its fast and successful development of fingermark.

Scanning electron microscopy (SEM)

SEM has some advantages compared to optical microscopy. Information about the morphology and the composition of the sample can be obtained at the same time and textured substrates that are not transparent can be examined. However, SEM may not be the best method for analyzing a large bulk of samples, due to the difficulty of locating accurately the area of interest (in our case the area of the latent fingermark). SEM works best with a pre-treated mark (i.e. cyanoacrylate)(25) because this way the area of interest becomes easy to visualize. Fingermarks on a wide range of metallic surfaces (brass, copper, steel, aluminium, stainless steel foils) have been studied and visualized with SEM along with their interactions with the substrates (corrosion etc.)(5, 26).

Scanning Kelvin Probe

A metallic surface is scanned with a vibrating gold wire probe measuring the Volta potential of the surface and latent fingermarks are scanned by observing the differences in Volta potentials where the fingermarks are deposited. When fingermarks are deposited on metallic surfaces, usually corrosion or insulation occurs, resulting to a different Volta potential compared to the regions of the surface where no fingermarks are deposited (8, 27). The whole procedure can take hours to complete but the advantage is that the fingermarks remain intact. Another limiting factor of this method is that the shape of the object/surface under analysis has to be relatively simple for this method to work effectively.

Williams et al(27) have demonstrated in their study that this method has potential in detecting eccrine fingermarks (figure 1), and fingermarks on metallic (i.e. iron) surfaces that were previously cleaned/rubbed or polished.

Matrix assisted laser desorption ionization – Mass spectrometry imaging (MALDI-MSI)

Matrix assisted laser desorption ionization – Mass spectrometry (MALDI-MS) is an established technique when it comes to analyzing human tissue due to its specificity and speed of analysis. The added benefit of MALDI-MSI compared to MALDI-MS is the ability to extract information about the distribution of compounds in a sample. When MALDI-MSI is used for fingermarks, no other sample preparation has to be performed except for the application of the matrix solvent. By using a matrix, the different constituents of the sample can be extracted into the matrix solvent with the help of a laser beam. The laser constantly changes position and a mass spectrum is acquired in each position during the analysis to ensure homogeneity and to capture a precise image of the fingermark and its chemical composition(8). It has to be noted here that MALDI-MSI can be used after a standard enhancement method (i.e. superglue) has been used first as Bradshaw et al. and Wolstenholme et al. report in their studies(28, 29). Moreover, Wolstenholme et al. demonstrated the versatility of MALDI-MSI as a stand-alone method for fingermark visualization in a range of substrates, including aluminium foil(29). Despite the promising results on aluminium foil, research on more metallic substrates is required to be able to perform a thorough assessment of this technique.

Time of flight – Secondary Ion Mass spectrometry (ToF-SIMS/SIMS)

Bailey et al.(30) demonstrated the improvement ToF-SIMS can offer in cases where the conventional methods produce unsatisfactory results, although the number of donors and substrate was not large (with aluminium foil being the only metallic one). A disadvantage of ToF-SIMS is that the use of vacuum has been proven to alter the composition of fingermarks (a reduction in the lipid composition of a fingermark)(31), which can be detrimental for a reliable analysis, since the original content of compounds is altered.

Thermal development

Most of the research concerning thermal development in the literature studies the development of fingermarks on paper(32). However, Wightman et al.(33) used this technique to develop fingermarks on metallic surfaces, namely brass, aluminium and steel which are materials used for ammunition casings. They used a low thermal mass furnace and acquired favorable results especially for the brass samples that were heated at 200 degrees Celsius (figure 2). The effect of time of deposition was also tested, and encouraging results were also obtained. However, only three donors were included in this study and further research is needed to produce reliable results. Moreover, this method is destructive and a lot of donor-specific information could be lost when it is employed.

Metal containing nano-particles and small particle reagents

Metal containing nano-particles are also used for the enhancement of latent fingermarks. These reagents rely on their affinity with the oily part of latent fingermarks and can be classified as powdering methods. ZnO was studied by Choi et al.(34) on its effectiveness as a fluorescent pigment on aluminium foil and favorable results were produced when using long wave UV-light for visualization. Rohatgi et al.(35) created a suspension powder consisting mostly of zinc carbonate hydroxide monohydrate and successfully enhanced fingermarks up to 25 days old on wet aluminium foil, while achieving high quality development on fingermarks that were 6-15 days old. The applicability of this technique to other metallic surfaces needs to be given further attention.

Vacuum metal deposition

The presence of fingermarks is detected by the different rate of metal growth on the surface. This is a sensitive method, which employs metal evaporation and exploits the difference in the rate of deposition on a surface with disturbances (i.e. fingermarks). A high vacuum chamber is needed for this method. Wetted surfaces and surfaces that have been exposed to high temperatures are not problematic when this technique is used. Gold/Zinc vacuum metal deposition is the most effective option in a wide range of substrates. First gold is deposited to form a thin film; this deposition facilitates the visualization of the marks. Gold diffuses in the areas where fatty components of the fingermarks are present. When zinc is deposited, it condenses in the areas where the non-diffused gold is (figure 3). Lately, vacuum metal deposition has been successfully used for the visualization of fingermarks on fired cartridge cases(36) but still needs to be studied further before it can become a standard casework procedure. A performance table (Table 3) provides a summary of the physical methods discussed in this section. In general, the studies examined in this section fall into the Phase 1 IFRG category (with the exception of the powdering techniques) and fail to place on a higher phase due to the lack of a higher number of substrates examined and donors employed or the lack of ‘’natural’’ fingermarks. However, that does not necessarily mean that the examined methods shown here cannot progress to a higher phase.

Chemical methods

In the chemical development processes, the fingermarks are usually treated with an aqueous reagent and their composition may be altered.

Acid dyes

Acid dyes target the proteins or protein rich compounds that are present in blood and give a coloured or fluorescent background. This is a method that has to be used in sequence with other fingermark enhancing techniques when a bloody metallic item is examined(8). Barros et al.(40) tested several acid dyes (as a part of a sequential treatment) on painted aluminium, and were able to develop/enhance bloody fingermarks. It is not clear however, how many of the developed fingermarks were of identifiable quality. Additionally, their donor pool consisted of only 3 donors.

Cyanoacrylate fuming

Farrugia(41) performed a comparative study on metallised plastic films, to find out the effect of cyanoacrylate fuming under vacuum. This method is performed in a fuming cabinet. It is noteworthy that eccrine fingermarks produce the best results since the polymerization produces fibrous noodle-like deposits on the ridges. Sebaceous fingermarks form different sphere-like structures (8, 42). The main advantage of this method is that it can be effectively used in almost all surfaces and it is not time consuming (20-30 minutes). However, often an extra enhancement of fingermarks is required after superglue fuming, additionally, this method is significantly hindered when the surface is wetted.

A modification of cyanoacrylate, Lumicyano (figure 4), which is a fluorescent reagent was studied by Prete et al.(43). Results on aluminium foil showed that Lumicyano could enhance fingermarks slightly better than the two-step method of cyanoacrylate and

Basic Yellow 40. Khuu et al.(13) evaluated the performance of commercially available fluorescent cyanoacrylate compared with the conventional Cyanobloom-Rhodamine 6G method. Slightly better enhancement was achieved overall with the majority of the Fluorescent cyanoacrylate methods. Greater enhancement was achieved specifically on polystyrene substrates and also on older fingermarks where Cyanobloom-Rhodamine 6G was underperforming. However, in both studies the number of donors was not high (3-4) thus, no definitive conclusions can be drawn yet. Although, cyanoacrylate fuming is routinely used in police casework and thus it has the potential to be considered for a phase 4 IFRG study, the articles examined in this section will be ranked as phase 1 study mostly due to the lack of donors included in each study.

Patination fluid and Gun Blue

Patination fluid consists of selenium dioxide and nitric acid. A black patina is developed when patination fluid is applied to metallic surfaces leaving the latent fingermarks intact. The process is time- and cost-efficient, but can be harmful due to the formation of selenious acid when selenium oxide is dissolved in water, additionally unfavorable results were obtained on aluminium surfaces. James et al. have studied this process on cartridge cases of brass and found promising results especially with sebaceous fingermarks (figure 5)(44). In their study, they also had a stable development time of 90 seconds per sample, which can be an advantage compared to the Gun Blue method, where the development time seems to be highly variable and user specific. This study was a proof of concept study and only one donor provided fingermarks, thus only limited data currently exists on the efficacy of these fluids.

Gun blue is a solution consisting of selenious acid, nitric acid, cupric sulfate and water. When applied on a metallic surface, which has latent fingermarks present, the surface becomes dark blue while the part of the surface where the fingermarks are deposited remains unstained due to the presence of fatty components (figure 5).

Gun bluing develops fingermarks rapidly and is a cost efficient procedure, but it has not been yet fully studied and has a questionable success rate on latent fingermarks(8). However, research so far using this technique has produced inconsistent results, possibly due to the use of different commercial products and different dilutions of those products used in each different study. Gun bluing appears to be the best approach when one needs an enhancement method for latent fingermarks deposited on surfaces made of brass (brass cartridge cases were mostly used in studies). Other advantages of this method are that it can be used as a part of a sequential fingermark (i.e. Superglue-Gun Blue) treatment, therefore a better development can be achieved and that it has shown promising results even for corroded and fired cartridge cases. (45-48). The application of this and other techniques to enhance fingermarks on cartridge cases is discussed separately.

Palladium deposition

Electrochemical deposition of palladium onto metallic (copper, brass, bronze) surfaces occurs leaving the area where the latent fingermark is deposited, intact, thus a negative fingermark is developed. The article with the latent fingermark is immersed in a solution and subsequently washed with water to halt the reaction (figure 6). Palladium deposition is a method not yet fully explored, although preliminary research has shown some potential(49). Dominick et al.(46) compared sequential cyanoacrylate (CA) and palladium deposition to sequential cyanoacrylate, gun bluing and Basic yellow 40 and found that the number of fingermarks that were successfully developed were similar. An advantage of CA and palladium deposition was that the fingermarks were developed after 1 day whilst the other method needed 2 days. However, the cost of palladium chloride makes this method less cost-efficient than others.

Development by Electrolysis

Nizam et al.(50) in their study found favorable results when using short electrolysis times (5 minutes) and a 37% HCl as solvent. However, developing fingermarks on the fired cases proved to be challenging with only one out of four cases yielding sufficient detail, while fingermarks in all of the unfired cases were successfully developed. When the time between deposition of fingermarks and development increased, there was a decrease in ridge detail developed, with absolutely no ridge detail developed after 7 days. This was a proof of concept study as only one male donor was utilized.

Development by aqueous electrolytes

Jasuja et al.(51) examined the development of eccrine and sebaceous fingermarks on metallic surfaces. The experiments were carried out by immersion (10-20min) of the metal substrate with the latent fingermark, in solutions of variable pH. In some instances, a second metallic surface (without latent fingermarks) was immersed in the solution to accelerate the reaction. Favorable results were obtained in most surfaces. Aluminium, zinc, copper, and brass were some of the surfaces examined, for some metals slight acidic solutions gave off the best development while others needed a more basic environment. However, the authors have used their own grading scale, with grade (3) being somewhere between a grade 2 or 3 compared to the Home Office’s grading system, with the ideal grade for an identification by the Home Office’s standards being 3 or higher. Nevertheless, the effectiveness of this method was proven, since 10 different donors were utilized for this study and ‘’older’’ fingermarks of 10 days old were successfully developed. This method can develop fingermarks fast while keeping the cost low. An example of the enhancement aqueous electrolytes can achieve is shown in figure 7.

Liu et al.(52) investigated how this method performs on fired and unfired cartridge cases using an adaptation of the technique used by Jasuja et al. For the unfired cases, sufficient development was possible for the vast majority of them, however only a few samples (4 out of 60) were successfully developed on fired cases. It appears that the heat and friction during the shooting have a detrimental effect on the fingermarks. Liu et al. also suggests that the longer a fingermark is deposited on a case the better its visualization will be due to the greater corrosion time allowed. In this study, the immersion time of the cases was 24h, which is significantly longer that reported by Jasuja et al.(51) (10 to 180 minutes) and could have played a role in the quality of the fingermarks that were developed. According to IFRG guidelines the studies examined here can be considered as phase 2, although a higher phase seems within reach since fired cartridge cases were also examined.

A performance table (Table 4) provides a summary of the chemical methods used for fingermark detection. Although destructive, these techniques do appear to be able to provide fingermark detection of a high quality. Using the IFRG guidelines, with the exception of the studies on the aqueous electrolytes methods which can be ranked as phase 2 the majority of the methods discussed in this section can be regarded as phase 1 studies. Cyanoacrylate fuming is extensively used in casework; however, the research examined in this review has been phase 1 or 2.

Physico-chemical methods

Electrostatic enhancement

Exploiting the corrosion that occurs even under ambient conditions when fingermarks are deposited on a metallic surface (brass, aluminium, steel), Bond et al.(56, 57) applied electrical potential to metal containing deposited fingermark and subsequently applied black powder to it. The black powder was applied with the help of spherical beads, which were rolled back and forth on the substrate. The conducting black powder was found to adhere on the corroded areas of the surface, thus assisting visualization. This technique was found to be useful when conventional techniques produced a grade 1 or 2 fingermark. When used sequentially, the fingermarks developed improved to a 3-4 grade. This is one of the few studies that included a large pool of donors (forty) and the results can be regarded as reliable. Nevertheless, the method was not effective when used on aluminium and steel surfaces.

Multi metal deposition (MMD)

MMD is a two-step metallic deposition. Colloidal gold is deposited, after the pH of the environment is adjusted, the amino and fatty acids of fingermarks become charged making it possible for the colloidal gold particles to be deposited on to them. After the initial gold deposition silver is deposited, which is selectively reduced and a dark brown color is obtained. Conventional MMD is ideal for developing fingermarks on non-porous surfaces, but the method failed to produce favorable results on dark surfaces(8).

Recently a modification on MMD was studied on aluminium foil(58). In this study instead of using silver, ZnO was deposited after the initial gold deposition. ZnO-based structures have luminescent capabilities, which can tackle the dark substrate visualization problem. In the case of aluminium foil, reverse deposition happens (the gold and ZnO nanoparticles have greater affinity with the aluminium foil rather than the fingermark). This irregularity does not appear to hinder the visualization of the fingermark (figure 8) and the method appears to work very well on aluminium foil, but no other metallic surface was tested in this study. Finally, only three donors were used in these experiments.

Single metal deposition (SMD)

Single metal deposition is an improvement of MMD. Gold particles are used both for the deposition and for enhancement steps. SMD can develop more samples in one solution bath, without any major detriment on the quality of the fingermarks while also enhancing 50% more fingermarks than MMD. Newland et al.(59) reported producing grade 2 fingermarks (figure 9) when using SMD on aluminium foil (among others), adding also that the method does not seem to be hindered by the environmental conditions (i.e. heat). One disadvantage of the method would be the increased use of gold, which can be costly. SMD compared to other enhancement methods remains a tedious method that requires keeping constant the pH value of the gold bath, and creating nano-particles of the suitable size. Therefore, trained personnel will be necessary to achieve optimal results.

Deposition of electrochromic films

A conducting polymer is deposited on the surface of interest. A negative fingermark is developed when using this method since latent/superglued fingermarks act as protection and deposition only occurs on the background. After deposition, the sample is placed in a second solution resulting in a change of colour in the polymer (decided by the examiner) depending upon what the best contrast would be. An advantage of this method is the ability to develop fingermarks on an array of metal substrates such as bronze, brass, lead, copper and nickel, all metals used for cartridge cases. The main disadvantages of this method are its destructive nature since further enhancement after applying this method is not possible, and the fact that it requires highly trained personnel to be able to carry out this process. Sapstead et al.(60) studied the insulating effect fingermarks have on metallic surfaces using electro-oxidation of copolymers on stainless steel films and improvement of the visualization of the negative fingermark based on film color, composition and topography. This was a proof of concept study but the authors reported the visualized fingermarks to be grades 3 to 4.

Beresford(61) achieved similar results by using polyaniline films on stainless steel plates, and it was equally effective on both old and fresh fingermarks. The method was tested with only one donor, but a range of fingermarks was produced under different sweat inducement times and deposition pressure. However, only 40% were usable for forensic purposes. Brown et al.(62) studied the enhancement of fingermarks with Poly(3,4-ehtylenedioxythiophene) also known as PEDOT. Different donors and different deposition times were used in this research to assess the efficacy of the method; additionally, the samples were also enhanced with one of the already established techniques to obtain a better understanding on which cases electrochromic deposition is superior to the other methods. The method achieved over 50% successful enhancements and specifically for samples of 7 days old, the success rate was 60%. The results demonstrate that this method can be complementary to the superglue method due to its superiority in substrates where superglue was unsuccessful. Brown et al. believe that this method could become a staple of fingermark visualization due to its relative low cost (similar to a cyanoacrylate development chamber) and relatively easy development technique.

Electrodeposition of metal nanoparticles

When a fingermark is deposited on a metallic surface, it can act as an insulator to an electrodeposition process. Deposition of metal nano-particles happens only on fingermark-free areas, thus obtaining a negative image of the fingermark with high contrast. Qin et al.(63) examined this method on a range of substrates and achieved high quality enhancement of fingermarks on both eccrine and sebaceous deposits (figure 10). Additionally, this method is faster (only five minutes deposition time) than MMD methods that require multiple bath immersions of the samples. Despite the promising results, this was a proof of concept study and only one donor was utilized.

Zhang et al.(64) used a similar method with the difference being the use of silver and copper particles. They achieved development of grades 3 to 4 for their fingermarks in an array of metallic surfaces (figure 11), they also underline that for each different metallic surface a different deposition time and potentiostatic parameters were ideal. Aged fingermarks of identifiable quality were also developed using this method from three different donors. A performance table (Table 5) summarizes the physico-chemical methods used for fingermark detection. According to IFRG guidelines all the studies examined in this section can be classified as phase 1 studies.

Ammunition/Cartridge cases

Several studies have focused on achieving fingermark enhancement on cartridge cases. In some of them, a comparative study is performed between fired and unfired cartridge cases. Dominick et al.(46) studied six different methods for the development of fingermarks on unfired cartridge cases. With the exception of powder suspension, the rest of the methods performed well, with Cyanoacrylate-Palladium and Cyanoacrylate-Gun bluing-Basic Yellow 40 giving the best results. Girelli et al. examined many sequential treatments like Cyanoacrylate-Gun bluing, Cyanoacrylate-powders, and achieved high quality enhancement (grade 3-4) on both metal discs and unfired cartridge cases. Liu et al(52). in their study, successfully recovered identifiable fingermarks on unfired cartridge cases with electrolyte deposition in a variety of pH ranges. Williams et al.(27) also, achieved visualization of fingermarks on unfired brass cartridges with a scanning kelvin probe. Generally, it could be concluded that visualization of fingermarks on unfired cartridge cases will be achievable in the majority of instances.

However, in a crime scene it is more likely that fired cartridge cases will be recovered and thus optimizing the recovery process of a fingermark from fired cartridge cases is of crucial importance. Development of fingermarks appears to be significantly more difficult on fired cartridge cases due to the heat and the friction of the expanded case against the barrel of the gun while it is being fired(67). Such processes are likely to degrade or chemically alter any remaining print residue. Contamination with gunshot residue and associated material may also be present on the cases after the firing of the projectile, which might be problematic for some development of any surviving fingermarks. In certain firearms (submachine guns, pistols), it is believed that a part of the fingermark material is damaged during the loading/reloading phase(68).

Girelli et al.(45) also studied the development of fingermarks on fired cartridge cases. The results show that the most efficient methods on unfired cases (Cyanoacrylate-Gun bluing, Cyanoacrylate-Basic Yellow 40) were the most efficient on the fired cases as well. However, the recovery was of a much poorer quality, acquiring fingermarks of Home's office's grade 3 in less than 20% of the cartridge cases examined. In order, to fully understand the inhibiting factors contributing to the added difficulty of fingermark enhancement on fired cartridges, the authors tried to pinpoint the cause by heating metal discs prior to enhancement and loading and unloading the cartridges. In both instances, development of fingermarks was successful indicating that there are other factors during the firing process that affect the fingermark recovery, such as the blowback of the hot gasses produced from the burning of the propellant powder, or some byproducts of the propellants that adhere to the surface of the cartridge and hinder visualization. Although, Girelli et al.'s study approaches the subject as close to real crime scene samples as possible and included many cartridge cases (100), the small number of donors (2) makes the drawing of conclusions difficult. Nizam et al.(50) used Electrolysis only, which successfully developed fingermarks in 1 out of 4, spent cartridge cases, and used only one donor. Additionally, other parameters of the method itself were not optimized (voltage, catalyst). Palladium deposition was one of the methods that has also yielded results on fired cartridges. Migron et al.(49) achieved partial development of fingermarks on fired cartridges while using palladium deposition, however partial development has limited forensic value. Leintz et al.(69) tried an optical approach in their study. A reflected UV imaging system was employed in an attempt to visualize the corrosion pattern from the sweat included in the fingermark, which has been proven to be detectable(57) even after a fingermark has been wiped off from the surface. However, this attempt did not result in high quality enhancement possibly due to the monochromatic wavelength source of the RUVIS system.

Summary

The complexity of visualizing a fingermark that has been exposed to various environmental factors becomes greater if one takes into account the alterations that occur on to the substrate; especially when dealing with metallic substrates (corrosion, change of texture).

In terms of choosing and investing in a fingermark visualization/enhancement method, chemical imaging techniques seem like the ideal technique for forensic applications due to their non-destructive nature. However, they can incur higher costs than other development techniques. A more realistic approach would be to focus on validation studies on enhancement techniques like palladium deposition, gun bluing, development with patination fluid, and/or aqueous electrolytes. All of the aforementioned techniques have shown their potential for fingermark development on metallic surfaces in proof of concept studies and in some cases partial development on fired cartridge cases and other metallic surfaces has been achieved. These techniques are simple to apply and their cost is minimal, which means that they could be incorporated into standard operating procedures in police laboratories once they have been validated.

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Table 1. Fingermark grading system (6)

Grade	Criteria
0	No ridge detail visible
1	Weak development; evidence of contact but no ridge details
2	Limited development; about 1/3 of ridge details are present but probably cannot be used for identification purposes
3	Strong development; between 1/3 and 2/3 of ridge details; identifiable finger mark
4	Very strong development; full ridge details; identifiable finger mark

Table 2. Summary of optical fingermark enhancement methods

Technique	Destructiveness	Evaluation of enhancement Quality	Limitations	Strengths	Literature
Infrared reflection	Non-destructive	Home Office's grade 3-4 can be achieved	-Works better when one knows where to look for latent fingermarks. -Only a few studies available	Information of other exogenous substances can be obtained (i.e. drugs)	(8-10)
Ultraviolet reflection	Non-destructive	Home Office's grades can be achieved but not consistently	-DNA on fingermarks can be damaged. -Process might be harmful for the user of the method.	Applicability on a wide range of substrates	(12, 18)
Fluorescence	Non- destructive	Home Office's grade 3-4 can be achieved	-Pre-treatment of the sample might be needed. -Only a few studies available	Achieves the best visualization compared to other optical techniques	(14, 15)
Raman Imaging	Non-destructive	Home Office's grade 3-4 can be achieved	- Only a few studies with artificial fingermarks available. -Pre-treatment is necessary for high quality visualization	Information of other exogenous substances can be obtained (i.e. drugs)	(16)

Table 3. Summary of physical fingermark enhancement methods

Techniques	Destructiveness	Evaluation of enhancement quality	Limitations	Strengths	Literature
Powder methods	Destructive	Usually a Home Office's 3-4 grade can be achieved	-Can be detrimental for further analyses -The examiner must know which powder to choose	Simple application, Minimal training needed	(14, 21, 34)
Scanning Electron Microscopy	Non-destructive	A high grade fingermark can be visualized	Usually better after the fingermark has been enhanced with other method	Morphology and composition of fingermark can be obtained	(5, 25, 26)
Scanning Kelvin Probe	Non-destructive	A high grade fingermark can be visualized	A flat surface is preferable	Can enhance fingermarks that have been wiped off. Can enhance eccrine fingermarks	(27)
MALDI-MSI	Destructive	A high grade fingermark can be visualized	Fingermark Requires pre-treatment	Can work as a sequential or standalone technique	(28, 29)
ToF-SIMS	Potentially destructive	Usually a Home Office's 3-4 grade can be achieved	Relatively unexplored	Has shown applicability in many substrates	(30, 37)
Thermal development	Destructive	Mediocre results most of the times	Detrimental to further fingermark analyses	Older fingermarks could be visualized	(33)
Metal-containing nano-particles/ small particle reagents	Destructive	Usually a Home Office's 3-4 grade can be achieved	Often extra illumination with UV light is needed	Also applicable on older fingermarks	(34, 35)
Vacuum metal deposition	Destructive	Usually a Home Office's 3-4 grade can be achieved	Competent Personnel is needed	Can be applied on wetted surfaces	(8, 38, 39)

Table 4. Summary of chemical fingermark enhancement methods

Technique	Destructiveness	Evaluation of enhancement quality	Limitations	Strengths	Literature
Acid dyes	Destructive	Usually a Home Office's 3-4 grade can be achieved	Has to be used with other enhancement methods Not thoroughly explored method	Can enhance fingermark contaminated with blood	(8)
Cyanoacrylate fuming	Destructive	Usually a Home Office's 3-4 grade can be achieved	Can be problematic in wet surfaces.	Can be used almost in all substrates	(41, 43, 53)
Gun bluing	Destructive	Usually a Home Office's 3-4 grade can be achieved	Still relatively unexplored	Enhances the substrate thus leaving the fingermark intact for further enhancement	(45, 46, 54)
Palladium deposition	Destructive	Usually a Home Office's 3-4 grade can be achieved	Still relatively unexplored	Has shown potential for developing fingermarks on fired cartridge cases	(46, 49)
Patination fluid	Destructive	Some of the samples gave off high quality fingermarks	Still relatively unexplored (only 1 study available)	Simplicity of use	(44)
Electrolysis	Destructive	Usually a Home Office's 3-4 grade can be achieved	Only 1 study available	Enhancement was successful in one fired cartridge case	(50)
Aqueous electrolytes	Destructive	Usually a Home Office's 3 grade can be achieved	Only 2 studies available	Successful with older fingermarks and in many metallic substrates	(51, 55)

Table 5. Summary of physico-chemical fingermark enhancement methods

Technique	Destructiveness	Evaluation of enhancement quality	Limitations	Strengths	Literature
Electrostatic enhancement	Destructive	Converted to Home Office's grade 2-3	Effective in a few metallic surfaces	A large donor pool was tested Effective when conventional techniques failed	(56)
Multi metal deposition	Destructive	Usually a Home Office's 3-4 grade can be achieved	Proven applicability only on aluminium foil.	Potentially works well on most metallic surfaces	(65, 66)
Single metal deposition	Destructive	Usually a Home Office's 3-4 grade can be achieved	Trained personnel needed	Enhancement is not hindered by environmental conditions	(59)
Deposition of electro-chromic films	Destructive	Usually a Home Office's 3-4 grade can be achieved	Trained personnel needed	Complementary to cyanoacrylate fuming	(62)
Electro-deposition of metal nano-particles	Destructive	Usually a Home Office's 3-4 grade can be achieved	Trained personnel needed	Successful enhancement for eccrine and sebaceous fingermarks and in many metallic surfaces	(63)

Figure legends

Figure 1. Image created by scanning kelvin Probe on iron substrate (27)

Figure 2. Fingermark thermally developed on brass surface

Figure 3. Fingermark developed with Vacuum metal deposition (Gold/Zinc) on brass surface

Figure 4. Fingermark developed with Lumicyano (left) and normal Cyanoacrylate (right) on brass surface

Figure 5. Fingermark on a cartridge case developed using Gun Blue

Figure 6. Fingermark developed with Palladium Chloride on brass surface

Figure 7. Fresh sebaceous fingermark on brass developed by aqueous electrolytes (NaOH)

Figure 8. Fingermark developed with MMD/ZnO on aluminium foil under 300-400nm excitation light (58)

Figure 9. Fingermark on aluminium foil developed with single metal deposition (59)

Figure 10. Fingermark developed with the electrodeposition of metal nanoparticles method (63)

Figure 11. Electrodeposition of metal nanoparticles on different metal surfaces (Left: Zinc, Right: Copper) (64)