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Review Article

Latent Fingerprint Enhancement Techniques: A Review

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Abstract:

Fingerprint (FP) is a global mark used for personal identification. This study reviews recent latent fingerprint (LFP) enhancement techniques including metal oxides, multi-metal deposition (MMD-I/II/SMD/Au-ASP), optical, chemical, physical, and physicochemical. Furthermore, analytical techniques involved in identification, evaluation, and determination of pieces of evidence, and perpetrators including the SEM, TEM, UV-Vis, IR/NIR, SERS, SKP, DLS, MALDI-MSI, and TD analysis were also discussed. Among numerous LFP enhancement techniques, the application of chemical methods in combination with optical techniques has a greater place to recover FPs with sufficient quality. However, instead of using such a complex and costly enhancing agent, nowadays nanotechnology is using specific techniques used for visualizing, inspecting, gathering, and analyzing trace evidence at the scene of a crime. To indicate using simple metal oxides such as ZnO that have superior fluorescent properties and also that consider both the surface and cost of the materials, enhancement of the LFP is functioning.

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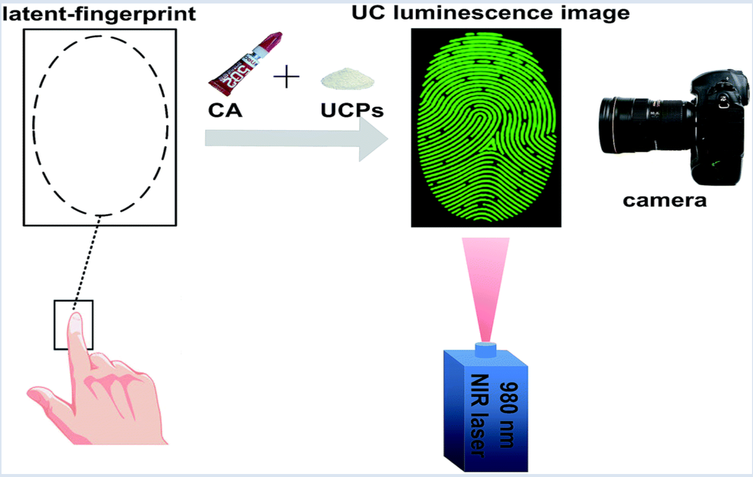
Keywords: Forensic sciences, Latent fingerprints, Enhancement techniques, Analytical techniques, Nanotechnology

Graphical Abstract:



Biography:

Buzuayehu Abebe: Mr. Buzuayehu Abebe is currently pursuing his PhD degree in Material Chemistry stream, Department of Applied Chemistry, Adama Science and Technology University, Adama, Ethiopia. He published more than 10 research articles and also presented papers in national as well as international conferences. His specific research interest mainly includes synthesis and applications of metal oxide composite nanomaterials.

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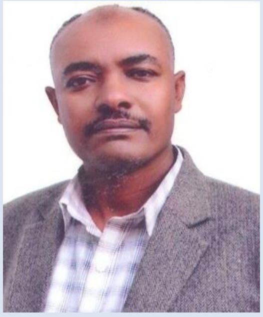

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1. Introduction

FPs have been adopted for physical sign identification of an individual in forensic science [1]. Nowadays, among several LFPs enhancement techniques, depending on the greater surface area of deposition and fluorescence abilities, the role of nanoscience and nanotechnology become an inevitable enhancing agent. In addition to the enhancement, it has also been used to explore genetic profiles, sex, and illicit drug used by perpetrators having little DNA quantities during the deposition of FPs on some surface [2,3].

The deposited FPs at the crime scene is categorized into three parts: (i) the visible (positive/negative), (ii) indented (molded/plastic), and (iii) the latent/invisible prints. Among these, due to the contrast difficulties of the surface or due to the deposition of impurities from the environment on the ridges of the finger/palm of natural secretions, extraction of information from latent prints are challenging. Normally, the enhancement of LFPs depends on the eccrine, sebaceous, and apocrine glands forming a bond with the enhancing agent at a specified condition. The eccrine glands are spread throughout the body and deposited with sweats contain more than 98 % water. Eccrine and sebaceous glands secretions are commonly found in every person FPs during the process of deposition. However, apocrine gland secretions are found in special conditions such as sexual molestation or abuse, it may not found during simple activities such as combing hair and touching the face [4].

In the past, the physical, chemical, optical, electronic, and optoelectronic methods have been employed as LFPs enhancement techniques [4, 5]. In addition, nanomaterial's such as: electrode-position of conducting polymers [6], gold or silver metal deposition [7], electrochemiluminescence [8, 9], multimetal depositions (MMD-I and II/SMD), gold-aspartic acid deposition (Au-ASP) [10], metal oxides deposition such as ZnO [11,12], SiO_2 [13–15], and semiconductors quantum dots [16], have been also tested.

Grades for the evaluation of developed FPs are dependent on the UK Home Office grading system (Table 1). Comparing the print quality of result obtained during enhancement, possible to evaluate the obtained results that helps to give the overview of extracting a piece of useful information from FPs pictured that the details of FPs ridges were visualized by using applied reagents [17].

Table 1. Grades for the evaluation of developed FPs.

Score	Description
0	Absence of development
1	All discontinuous or dotty ridges
2	1/3 of continuous ridges
3	2/3 of continuous ridges
4	All continuous ridges

Environmental factors such as fire soot deposition on the ridge, exposure to drainage water, soil/snow, humidity, light, air circulation, arson, explosion, and other biological factors remains a challenging task in the LFPs recovery procedure. All of them disturb the ridge after deposition of the print, furthermore, the offender may also try to destroy the prints bearing crucial evidence [18, 19]. This review also provides a brief discussion on optical, chemical, physical, and physicochemical techniques in combination with analytical techniques, including transmission electron microscopy (TEM), atomic force microscopy (AFM), ultraviolet-visible spectroscopy (UV-Vis), Near-infrared (NIR), surface-enhanced Raman spectroscopy (SERs), scanning Kelvin probe (SKP), dynamic light scattering (DLS), matrix-assisted laser desorption/ionization mass spectrometry imaging (MALDI-MSI), thermo-desorption (TD) for further enhancement and extracting detail information by going deep into the ridge of the FPs.

2. Morphogenesis of Friction Ridge Skin

The morphogenesis of friction ridge skin begins during the first week of estimated gestational age. In the period between 7th and 8th weeks, volar pads appear on the finger starts to develop. The first interdigital pads that appear include, the 2nd (II), the 3rd (II), and the 4th (IV), along with apex finger pads (Figure 1a). All these pads were found to be visible up to 10 weeks from the beginning of the estimated gestational age. The friction ridge skin significantly develops during the period between the 11th and 20th weeks. At around 10th weeks, the cells start to proliferate. Before ridge development, the epidermal surface becomes thick and smooth on its outer surface to give primary dermal ridges (Figure 1b). These cells in association with a sweat gland, transformed into ridges commonly called “ledges”. These ridges mature by developing downward have a primordial crepe-like structure known as the dermis.

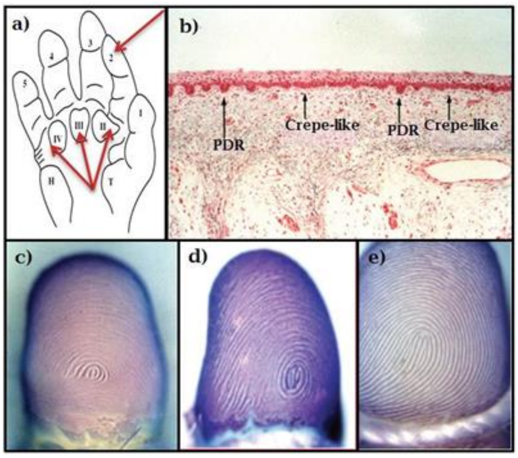

Figure 1. (a) Volar pads; (b) Cross-sectional view of the skin of the fetus; (c & d) Dermal surface of the index finger of a fetus (11 weeks EGA); (e) (14 weeks EGA) [4].

The perfect development of the three fronts of ridges at different rates within the dermal surfaces is shown in (Figure 1c, d, e) [4]. Classification of the ridge detail can also be made by the structure of the print such as a loop, whorl, and arch as level 1, the appearance of small individual characteristics as level 2 and finer details such as the shape and location of pores as level 3. The ridge details and ridge characteristics are shown in Figure 2 [20].

Figure 2. FPs s ridge characteristics: 1st, 2nd, and 3rd level details.

The surface characterization ability for FP on the substrates depends on the nature of the surfaces which could be porous, semi-porous or nonporous. It was also found that surface deposition on porous (paper, cardboard, fabrics, and wood) and nonporous (plastic, glass, metal surfaces, and ceramics) surfaces characteristics were demonstrated in the form of water-soluble deposits (WSD) and non-water-soluble deposits (NWSD), respectively. Very quick adsorption of water-soluble secretions takes place at porous surfaces whereas non-water-soluble components remain at the surface for a long time [4].

3. Mechanism of Multimetal Deposition

Nowadays, due to the high stability and sensitivity, elemental states of noble metals such as gold and silver NPs are used for FPs detection. In MMD, the small particle reagent and physical developers with wet chemical methods were applied to detect the LFPs. The process of LFP enhancement in MMD involves immersing the developed LFP in the metal NPs solution at acidic pH ranging from 2.5 to 2.8, at a pH>3.1 the eccrine, sebaceous, and apocrine glands lose their reactivity. Therefore, it is recommended to work at a pH lower than 2.8. In addition to the pH of the solution optimization of parameters such as particle size, temperature, homogeneity of the solution, and concentration of the developer are also important [21]. It is interesting that, under the acidic environments, due to the protonation proteins, peptides or amino acid-containing components present on the FP ridges carry a positive charge. Moreover, due to the deposition of citrate anions, gold NP (which was reduced from its precursor by citric acid) becomes negatively charged. Therefore, due to electrostatic interaction, the Au NPs easily deposited on the FP ridges. Here, gold NPs on the ridge acts as a catalytic nucleation site [10].

Furthermore, the interaction between gold particles and proteins also further facilitated through hydrophobic (*e.g.*, Vander Waals) interactions [11,22]. To impart a noticeable color to the ridges, silver particles were reduced by iron (II) ions deposit on colloidal gold particles giving black color to the ridges. However, even if the application of MMD is fruitful for developing FPs on nonporous surfaces, most of the time the method is not successful for dark surfaces.

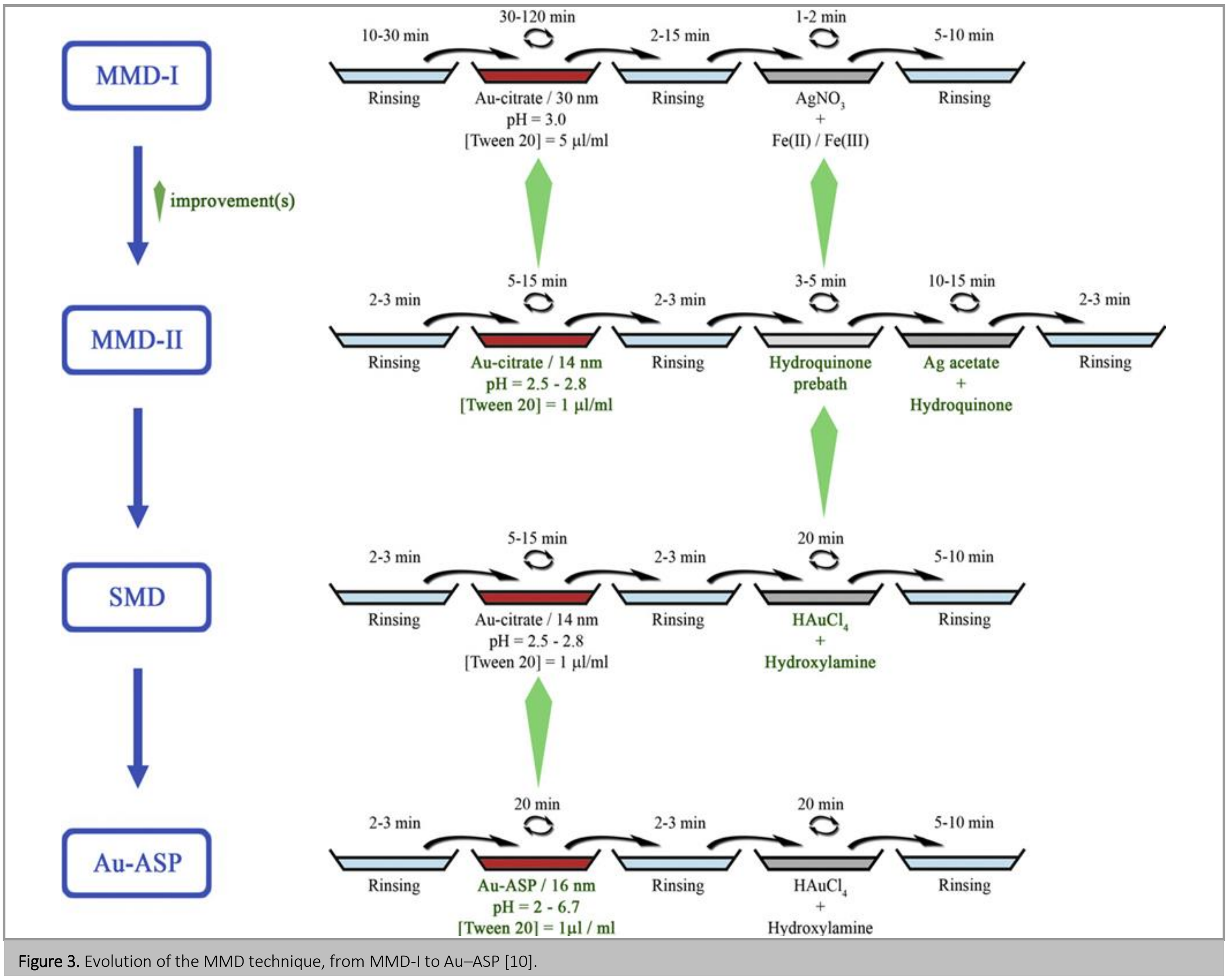

Figure 3. Evolution of the MMD technique, from MMD-I to Au–ASP [10].

To overcome this drawback, MMD was modified to develop new multi-metal deposition II (MMD-II). The gold sol produced in this method was found to possess small particle size and homogeneous in its crystal nature. In this method, silver ions are reduced to metallic silver [23]. However, this method also has some drawbacks, such as following more complex reaction and time-consuming process even more than MMD-I. The other more enhanced techniques next to MMD-II is SMD. SMD LFPs enhancing technique uses only single metal for both deposition and enhancing steps. This method is cheaper and uses fewer reagents with no risk of unwanted background darkening problems [24].

Becue *et al.* [10] have studied the SMD technique using aspartic acid and sodium citrate (Au-ASP) reagents used for improving the efficiency of the enhancement. In addition to enhancement, the SMD technique also extends the working pH range from 2 to 6.7 without affecting the activity of the glands that form a bond with metals. It was found that, that Au-ASP techniques would remove limitations that were observed on the above-discussed methods (i.e. it decreases the labour-intensive synthesis of colloidal gold NPs and increasing working efficiency above pH-3). As illustrated by a green arrow in Figure 3, major modifications such as decreasing the particle size from 30 nm to 14 nm, reducing the working time from 30-120 min to 5 – 15 min, and twining working pH from 3 to 2.5 – 2.8 in MMD-II; using HAuCl4 and hydroxylamine in place of hydroquinone in SMD; and using Au-ASP in place of Au-citrate in Au-ASP were conducted. Zhang *et al.* [25] also confirmed the possibilities of further enhancing the LFPs by MMD techniques combined with scanning electrochemical microscopy (SECM). Here the SECM enhancement process conducted after MMD adsorption of gold NPs on chemically deposited silver metal. However, as suggested by Becue *et al.* [26], due to luminescent properties, high surface area, reducing background contrast problems properties of metal oxides nanomaterial's like ZnO, replacing of them in place of silver/gold blocks reduces the dark substrate visualization problem that was specified on MMD.

4. LFP Detection by ZnO Based Metal Oxides

It is known that, every FP that has a unique and distinctive ridge pattern with a physical evidence for the identification of individuals, analyzing the age and sex. As indicated above, up to now several methods in combination with analytical techniques were practiced as LFPs enhancing agent [27]. Nowadays, nanotechnology in forensic science supposed as the process of using specific techniques and methods for inspecting, gathering, and analyzing trace evidence at the crime scene. Due to the increment of the surface areas of the deposited materials, applying nanotechnologies in forensic science is significantly transform the processes of analysis to be faster, accurate, effective, sensitive, and easy to apply [28]. Generally, the role of nanotechnology for addressing current forensic investigation includes FP identification, explosive residue detection, DNA analysis, ion beam analysis, nano-trackers, screening of drug-facilitated crime, estimation of time since death [29].

Still, relative to optical and chemical enhancement techniques, the physical method deposition techniques using metal oxide nanomaterials do destruct the ridges of the FPs. Among the different physical methods, the powder method is one of the simplest methods that work by sticking the powder on the surface of the FPs. From powder dusting, metal oxide NPs, specifically zinc oxide that has great properties such as high chemical stability in the air, good photo-luminescent properties, and strong spectral absorption behaviour in the UV region give nice enhancement of LFPs [30–32].

Deepthi *et al.* [12] used a hexagonal ZnO NPs prepared by solution combustion methods for enhancement of LFPs. As explained by the workers, the fluorescent property of ZnO supports the material to show good LFPs enhancement competence on varieties of surfaces at normal light illumination. Guzman *et al.* [33] also synthesized 10 nm to 30 nm semi-spherical forms of zinc oxide NPs for recognition of LFPs and got nice results. Yu *et al.* [34] used molybdenum boats thermal evaporator techniques to deposit ZnO on polyethylene terephthalate surface for enhancement of LFPs. Comparing ZnO deposited on Au thin seedling layer, ZnO deposited without the seedling layer gives effective and normal growth of FP. Therefore, their work confirms the possibility of depositing ZnO in the valley between the FP ridges without using common techniques like Au thin film or cyanoacrylate fuming; this is due to the presence of Zn-O bonds that has great electrostatic interaction with organic compounds present in the deposited sweat. Furthermore, the nontoxicity, cheap precursor of the powder makes it advantageous over other commercially available complex and expensive development techniques.

Using both the physical and wet powdering methods as a developing agent, the ZnO-SiO_2 powder synthesized by simple heating techniques by Arshad *et al.* [35] was applied for enhancement of the FPs developed on different semi-porous and nonporous surfaces. Due to the hydrophobic interaction between the ZnO-SiO_2 powder and water-insoluble organic compounds like sebaceous, eccrine and apocrine glands, good development of the powder on the surface was taking places. The obtained result also indicates the development of clear in addition to the 1st, 2nd and, 3rd level ridge details. By stating the misunderstanding of the possibilities of recovering the destroyed FPs, Dhall and Kapoor, [36] also successfully recovered the FPs exposed to different natural destructive conditions using wet powder suspension techniques.

Furthermore, Tiwari *et al.* [38] synthesized Zn, ZnO, and ZnS powder and deposited next to the deposition of Au as a nucleation site. The deposition was done simply by evaporating the material using an oil diffusion pump. The area of the FP obtained using AFM is 40 μm with the average height of 100 nm. The obtained result indicates, the enhancement order of Zn<ZnO<ZnS. As explained by the researchers, the less enhancement potential of Zn is due to its avoidance of forming contact with oily parts of the FP residue. The obtained reflectivity of deposited multilayer using transfer matrix calculations also indicates the occurrence of a parallel homogeneous layer of a stratified medium. The summarized forms of the above discussed and some other additional ZnO based LFP enhancing metal oxides work were present in Table 2.

5. Mechanism and Effect of pH on Modified SiO_2

Metal oxides have also been utilized to enhance the LFPs with superior quality. However, as suggested by Moret *et al.* [41], optimizing the pH and zeta potential, ζ-potential becomes important to understand the detailed mechanism of bonding between functional groups and NPs surface. SiO_2 NPs can influence the physicochemical properties of gold NPs in the MMD process, to indicate, using carboxyethylsilanetriol that offered a negative environment for gold NPs. Therefore, SiO_2 NPs were found to be better than single gold NPs [42]. The strong negative ζ potential value (≤ -30 mV) over the entire pH range confirms the stability of the oxide. Furthermore, when SiO_2 NPs were functionalized with succinic anhydride (SiO_2-$(COOH)_2$), almost similar trends like SiO_2-COOH interactions were observed. Besides, NPs functionalized by methyl-phosphonate and sulfonate ions showed a high negative ζ potential across all pH ranges, however, there was no affinity with the residues of FPs, revealing fewer LFPs enhancing capabilities of the material. To further enhance the linkage between ridge (amine and carboxyl) and enhancing agent, acoupling agent such as carbodiimide groups were also used [41].

Table 2. A reviews on ZnO based oxides for LFP enhancement techniques.

Method & Catalyst	**Pollutant**	**Precursor**		**Properties**		**Conditions**			**Ref.**
	Surfaces	Zn	P2	FE	D, SEM/TEM, BG	Applying Powder	Light	Temp. (°C)	
Precipitation (ZnO)	Non-Porous	$Zn(NO_3)_2\cdot 6H_2O$	LiCl	587& 388	>1000, petal, -	Brush	Uv	25	[37]
(ZnO)	Non-Porous		-	-	-	Thermal evaporator	Optical Microscope	-	[34]
Heating (ZnO-SiO_2)	Semi & Nonporous	$Zn(Ac)_2. 2H_2O$	$Na_2SiO_3. 5H_2O$	-	32.9, agglomerated, -	Wet powder Powder Dusting	Nikon Cameras	700	[35]
ZnO	Nonporous	-	-	-	-, 465, NRs,	Wet Powder Suspension	SLR Camera	-	[36]
combustion ZrO_2/CuO	Porous & Nonporous	$ZrO(NO_3)_2$	$Cu(NO_3)_2$	-	-, -, 1.9	Brushing	UV	- 500	[30]
-	Nonporous	-	-	-		Evaporation	Digital Camera	-	[38]
Precipitation ZnO	Non-Porous	$Zn(Ac)_2. 2H_2O$	-	360	14.75, -, 3.26	Powder Dusting		300	[39]
Combustion ZnO	Porous & Non-Porous	$Zn(NO_3)_2. 6H_2O$	-	502 & 406	-, disks-like, 3.4	Powder Dusting	Digital Camera (UV)	-	[12]
ZnO Fe2O3 (co-precipitation)	porous & nonporous	$Zn(NO_3)_2. 6H_2O$	$FeCl_2 \cdot 4H_2O$	-	-, -, 4 & 2.75	Dusting Method	Digital Microscope With HDMI	110& 80	[37]
N-CDs/ZnONPs	non-porous	$Zn(Ac)_2. 2H_2O$	-	389,443, &472	-, 40–50, -	Powder Brushing	iPhone camera (UV)	450	[40]

NBE: near-band-edge emission (nm), FE: Fluorescence emission, D: crystallite size (nm)

6. Optical Fingerprint Detection Techniques

FP development using an optical spectrometer is a superior systematic technique to detect LFPs composition and morphology at a specific wavelength without other chemicals need.

6.1. Infrared Spectroscopies

Spectroscopic investigations in IR and NIR ranges are relatively fast and non-destructive techniques that can detect LFPs composition and morphology simply by lighting IR radiation sources and capturing the emission from the surface of the analyte. The absorption of the emitted light was accompanied with the help of analytical tools like principal component analysis [43]. During the lighting mechanism, the substrate would appear as light in color while, the FPs appear as a dark color. Williams *et al.* [44] have investigated the chemical nature of LFPs residues of children in between 2 and 11 years age using Fourier-transform infrared micro-spectroscopy analytical technique. The obtained result confirms the presence of different functional groups including carboxylic acid, proteins, and esters in the FPs. Due to the volatility nature of esters, salts residues present in the FPs assists the ridge to be more stable.

Other than specific functional groups, Fritz *et al.* [45] also examined the degradation of FPs using synchrotron-infrared microscopy within 9-month intervals. From the result, small-signal intensity was recorded. However, the lipid composition was found to remain almost the same. To know the rate of aging in FPs, Girod *et al.* [46] measured the initial composition and kinetics of the FPs. After storing the FPs in an open atmosphere (20 °C), the possibilities of grouping FPs based on their age were also confirmed by the chemometric analysis. Crane *et al.* [43] also studied the enhancing capabilities of IR light on the untreated LFPs on varieties of substrates (nonporous and porous), and concluded that IR is efficient to enhance LFPs. Banas *et al.* [47] analyzed the exogenous substances present in LFPs using FT-IR spectro-microscopy with the help of attenuated total reflection (ATR) sampling tool. In addition to enhancement of LFP ridge details, it was confirmed that the possibilities of characterizing, identifying, and quantifying of FT-IR active exogenous compounds deposited within the developed LFPs. It provides evidence of materials touched by the person at the crime scenes.

The NIR techniques are also detection techniques that work only on one donor of FPs at 980 nm radiation. Due to its superior contrast and reduced background luminescence, NIR techniques are better than visible spectral techniques. Maynard *et al.* [48] tested enhancements of LFPs on porous, non-porous and semi-porous surfaces under 650–1100 nm spectral region. After cyanoacrylate fuming followed by staining with NIR dyes, they obtained a considerable NIR absorption for ninhydrin, iodine/benzoflavone, and powdering techniques. Errington *et al.* [49] also confirmed the enhancing capacity of micronized Egyptian blue pigment on non-porous surfaces as near-infrared luminescent impressions.

The test conducted by Li *et al.* [50] using the $NaYF_4$: Yb, Er, Gd fluorescent up-conversion nanorods (UCNRs) and Acid Yellow 7 (AY7) as a comparison confirms the superiority of UCNRs over AY7 on all test substrates. The result indicated that the magnified and detailed image of a blood FP with various second and third levels was detected. In Wang *et al.* [51] work the fluorescent properties and good enhancing capability of $NaYF_4$: Yb, Er up conversion NPs were confirmed with superior efficiency and sensitivity on varieties of substrate materials.

Besides, as a new genre of FP visualization techniques, the NIR-NIR fluorescence enhancement techniques were also reported [52]. They confirmed the possibilities of using finely divided cuprorivaite powder (that has a primary and secondary excitation maximum at 637 nm and 780 nm, respectively). They also revealed the use of these materials to reduce the multi-colored background interferences and raise the fluorescence ability of the ridges. Across a wide range of substrates, King *et al.* [53] validated the LFPs enhancing efficiency of finely divided spirulina platensis dust powder with the help of NIR techniques. The material also showed encouraging results in the field and laboratory cases. To strengthen NIR luminescent FP properties, King and Skros [54] and Chadwick *et al.* [55] synthesized $Zn_3Ga_2Ge_2O_{10}$: 0.5 % Cr^{3+} and styry111 mixed with rhodamine 6G (mixture = StaR11), respectively.

6.2. Long wave UV spectroscopy

LWUV technique is an optical method provides full FP information even on the surface which has various colors that disturb the visualization of the LFPs. However, this technique may destruct the DNA present in the FPs; especially when short wave UV (SWUV) radiation used. King *et al.* [56] conducted the application of long wave UV radiation (LWUVR) for the enhancement of cyanoacrylate developed FPs in the absence of chemical reagents or dye stains. According to the study, LWUVR was simple, versatile, non-destructive compared to (SWUV) and due to the sensitivity of the camera and high-resolution performances; it has the capacity of enhancing the ridge details. Furthermore, with the help of LWUVR, the effectiveness of five FP visualization process sequences(s) on £10 polymer banknotes was tested [57]. After applying each process, they concluded that black magnetic powder with LWUVR has the most effective single process.

The test conducted by metal-organic frameworks (MOFs) containing Lanthanides (Eu and Tb) treated with cyanoacrylate fuming by de Jong and de Puit, [58] shows an attractive luminescent property for both visible as well as UV light, even after persevered for 12 months. Barros *et al.* [59] also utilized Benzazole dyes and rare-earth-based nanopowder for enhancement of LFPs. From the observed results distinct ridge details and enhanced properties were compared with traditional FP powders. In the same year, the research conducted by Sébastien *et al.* [60] and Guo *et al.* [61] confirmed the futuristic enhancement materials of MOFs.

6.3. Raman Spectroscopy

Raman imaging techniques provide more information due to the detection of more spectral bands. In addition to FP analysis, it also serves to identify different chemicals and biological materials present within the ridge. Essentially, Raman and IR techniques might use in tandem as they are a complement to each other. Due to the sensitivity and selectivity, SERS also detect small size samples and small concentrations. For instance, the compositional analysis conducted on ultra-high-purity diatomaceous earth prepared as button-like tablets shows a nice result on SERS techniques [62]. In the presence of Au nanofilm as adhesive, Lin *et al.* [63] also confirmed the sensitivity of SERS techniques. The obtained result shows the full pattern of FPs, including the ridges details, furrows, and sweat pores. Figure 4 demonstrates the collection of LFPs and images of the living FPs on ANF. The undetectable LFPs enhancement was achieved using the SERS analysis [64].

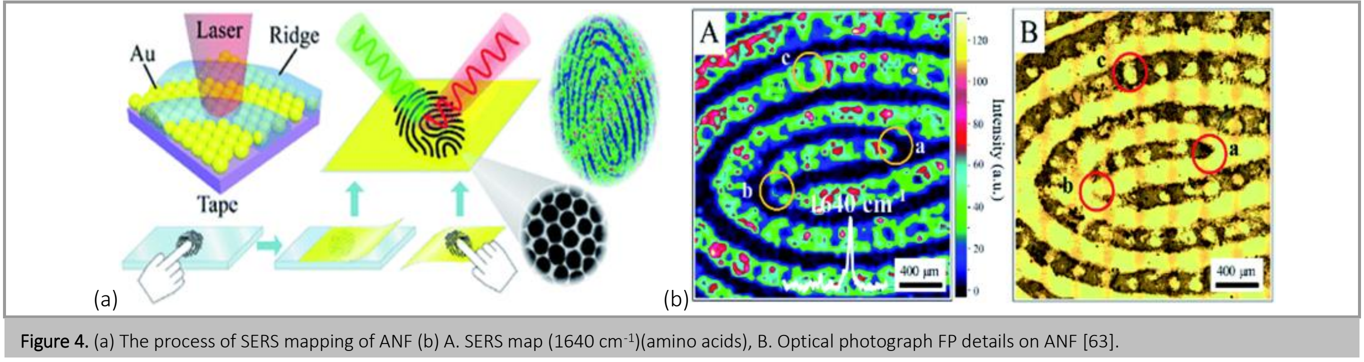

Figure 4. (a) The process of SERS mapping of ANF (b) A. SERS map (1640 cm^{-1})(amino acids), B. Optical photograph FP details on ANF [63].

7. Chemical Methods

Chen *et al.* [65] synthesized ninhydrin embedded NIR fluorescent semiconducting polymer dots. The attained result revealed all details levels on both porous and nonporous surfaces. Barros and Stefani [66] approved the LFPs enhancing abilities of benzazole dye embedded into a silica matrix on porous, non-porous and colored surfaces. Milenkovic *et al.* [67] prepared n-doped carbon dots for LFPs enhancing application. After proofing the size of fluxes blended Y_2O_3:Eu^{3+} being in nano-range by TEM and PXRD characterization techniques and its highest photoluminescence (PL) intensity by PL emission spectra, Venkatachalaiah *et al.* [68] used it for LFPs visualization determination. In this study, it was confirmed that the PL properties of the materials are possibly due to the presence of oxygen vacancies. The result also shows the high efficiency and sensitivity of the materials even after submersion in water. A highly sensitive and low-cost orthorhombic phase nano phosphor ($YAlO_3$: Sm^{3+}) which is synthesized by eco-friendly green combustion techniques from green tea leaf extract were also developed [69]. The result tells the possibility of distinguishing the main features of FPs.

Li *et al.* [70] have reported an orange fluorescent carbon NPs, that displayed as an excellent emission enhancement. The result indicates that HFCNs was an extraordinary tool for LFPs materials. After pointing out the challenges of enhancing bloody FP, Barros *et al.* [59] used benzazole dyes to detect bloodstains and bloody FPs on varieties of surfaces. Compared to Amido Black, luminol and cyanoacrylate, this technique which uses only water as a solvent has high efficiency for LFPs enhancing applications.

Cyanoacrylate fuming is also used as LFPs enhancement. It works on all surfaces and less time-consuming techniques. Especially, one-step luminescent cyanoacrylates did not require using luminescent post-treatment. The polymer cyanoacrylates found that PolyCyano UV and PECA multiband have excellent enhancing properties [71]. It also studied that when a cyanoacrylate glue joined with aggregation such as fluorine, 1, 8-naphthalimide and anthracene (AEE), the hydrophobic interactions between AEE and cyanoacrylate glue led to enhancing the LFPs. As seen in Figure 5, compared to non-pretreated, the SEM images indicated the clear FP ridge details results for the superglue pretreated one [72].

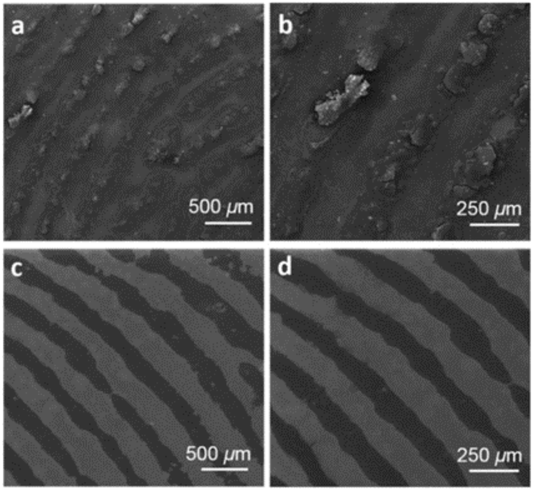

Figure 5. SEM images of LFP incubated with NIFA NPs solution without (a, b) and with (c, d) pre-treated superglue fume [72].

Chen *et al.* [73] synthesized the supper LFPs enhancing material through combining ethyl-2-cyanoacrylate ester fuming as a superglue and poly (p-phenylene vinylene) (PPV) NPs as surface area enrichment using simple fuming and staining procedure. It was found that, using fuming that develops the FPs within the in-situ processes did not alter the properties of the substrate. The confocal microscopy images under bright field, dark field, the overlaid image, and the SEM images [73] are presented in Figure 6.

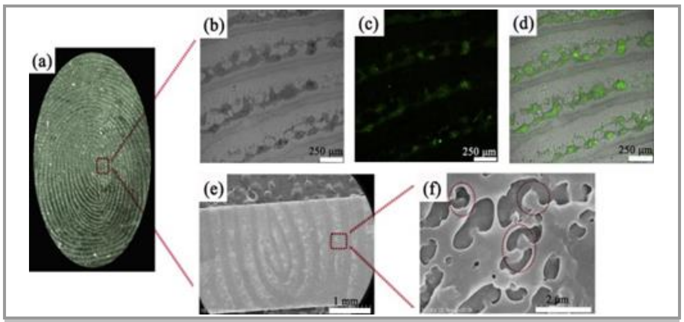

Figure 6. (a) FP of the confocal microscopy images under (b) bright field, (c) dark field (d) the overlaid image, and (e and f) the SEM images [73].

The upconversion particles (UCPs) of ($NaYF_4$: Yb, Er/Ce) coated with polyethyleneimine (PEI) sub-microcrystals and cyanoacrylate-fuming (CA-fuming) was also reported [74]. UCPs revealed a high ability to decrease background interference for obtaining high-contrast FP images, compared to down-conversion materials. The steps used during development of FP and the synthesized UCPs materials are demonstrated in Figure 7. NIR-responsive up conversion fluorescent nanocrystals powder of $NaYF_4$: Yb, Er, and $NaYbF_4$: Er/Tm/Ho was used to develop LFPs on the surfaces of varieties substrates [75].

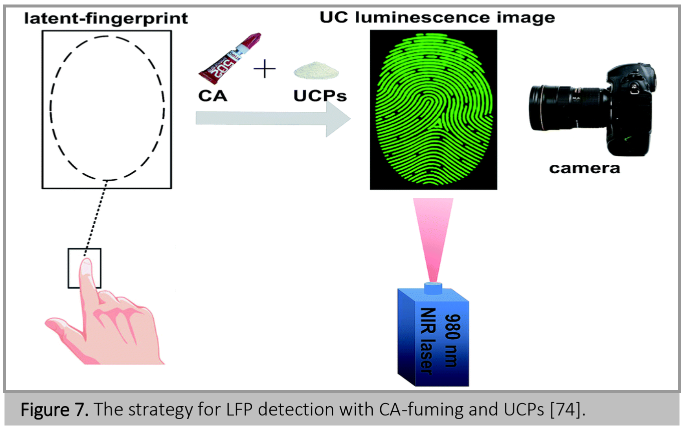

Figure 7. The strategy for LFP detection with CA-fuming and UCPs [74].

Even if enhancing FP present on ammunition is difficult, as suggested by James and Altamimi [76] cold patination fluid containing SeO_2 and HNO_3 was found to be effective. The LFPs enhancement work conducted by Pitera *et al.* [77] explains the role of the nature of the surface and conditions during visualization. Girelli *et al.* [78] also compared the application of powder dusting, fuming of cyanoacrylate, gun bluing solutions and acidified H_2O_2 solutions towards FP enhancing capacity. Liu *et al.* [79] also tested the effects of pH on both treated and untreated cartridge cases. The study indicated that both the acidic and basic range will affect the microscopic striation examination; therefore, the neutral pH is optimum. Furthermore, the degradation abilities of FPs by heat and friction also observed on the fired cartridge cases. Morrissey and Birkett [80] compared the enhancing capacity of (1) superglue fuming followed by basic yellow 40 fluorescent dye staining, (2) superglue fuming followed by gun blue, and (3) gun blue (GB) as a single process on fired and unfired 9 mm brass Luger ammunition casings. Relative to the other, the single GB process enhances the LFPs with the highest quality and clarity.

8. Physical Methods

Relative to optical and chemical methods, the physical method does not destruct the ridges of the FPs. Powder method is one of the simple physical methods which works by sticking the powder preferentially to the surface of the FPs. It involves the application of any one of the following reagents; the metal flake, granular, magnetic flake, and magnetic granular, having their benefit and weakness. Song *et al.* [81] synthesized phenyl-doped graphitic carbon nitride (PDCN) material for enhancement of LFPs. Due to creation of electronic structural change on carbon nitride network, compared to pristine graphitic carbon nitride, doping of phenyl groups shows great enhancement of photoluminescence efficiency and stokes shift.

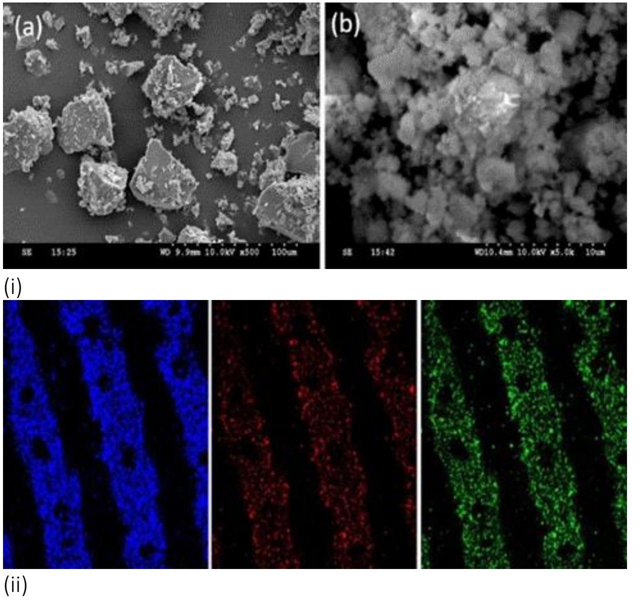

Figure 8. (i) SEM images of PTF formulation, a. before coating and b. after coating and grinding; (ii) Confocal microscopy images of sebaceous FPs showing 3rd level details in FP ridges [84].

Yuan *et al.* [82] applied four cationic dye-diatomite composite powders (the blue MB-diatomite, pink ST-diatomite, green MG-diatomite, and purple CV-diatomite) and concluded that these materials are promising for practical use of LFPs enhancement techniques. Deepthi *et al.* [83] synthesized 3D CeO_2: Eu^{3+} for physical deposition. The result revealed that all three levels of ridge details were observed with superior quality enhancement and minimum interference in the background.

Sharma *et al.* [84] used a 2-(5-(9, 9-diethyl-9H-fluorene-2-yl) thiophen-2-yl)-1-(4isopropylphenyl)-1H-phenanthro [9, 10-d] imidazole (PTF) powder to enhance the fresh and old FPs deposited on varieties of surfaces and concluded that they were successful in enhancing LFPs. The SEM images of PTF indicated the high surface area to volume ratio values of the particle. This facilitated the deposition of the particles on the FPs, improved the contrast (Figure 8). The confocal microscopy images of LFPs demonstrated the possibilities of recording the emission on blue, red or green regions, among these; the blue region shows more intense emission than the other. Other than PTF, experimental work was also conducted on phosphor powder and established better enhancement [85].

Using a sol–gel process and 8 h thermal treatment (1100 ºC), Saif, [86] synthesized lanthanides doped yttrium zirconate NPs (Ln^{3+}: $Y_2Zr_2O_7/SiO_2$) impregnated in the silica matrix. Depending on the different Ln^{3+} ion transition mechanisms, the synthesized nano-sized spherical material provides different color emission that helps for the enhancement of the prints. Wu *et al.* [87] synthesized polyallylamine (PAA) polymer functionalized green (ZnCdS) and red (Cu–ZnCdS)-emitting QDs anchored and embedded in silica NPs, respectively. The result also indicated that both the red and green QDs could effectively stain the LFPs due to electrostatic and hydrophobic interactions, both the level 1 (better ridge configuration) and level 2 (ridge termination, bifurcation, and lake) are displayed. A 20 nm lanthanum oxyfluoride (LaOF) prepared via ultrasound-assisted sonochemical route for enhancements of LFPs and lips was well studied by [88]. During the synthesis process, to raise the surface area of contact between the FPs residues and LaOF enhancing agent, they optimized different experimental parameters including the time of sonication, surfactant volume, pH, and temperature of the bath and sonication power. The result indicated that, the material improved the LFPs without any background interference.

9. Physicochemical Methods

Electro-chromic material is a material that changes color under the influence of the electric field. This material also changes their structure upon the addition and removal of ions, accordingly their light adsorption wavelength [89]. With the help of electro deposition techniques using the cyclic voltammetry method, Ding *et al.* [90] tested LFPs enhancement capacity of compact and uniform film (500 nm) of Prussian blue coated on the ITO electrode. They obtained the enhanced images of bright ridges and the blue substrate surface under optimized humidity (70 %) and applied voltage (30 V). Beresford and Hillman [91] also potentiostatically tested the enhancing abilities of polymer polyaniline. The polymer polyaniline deposited on the stainless steel substrate used as a working electrode (0.9 V).

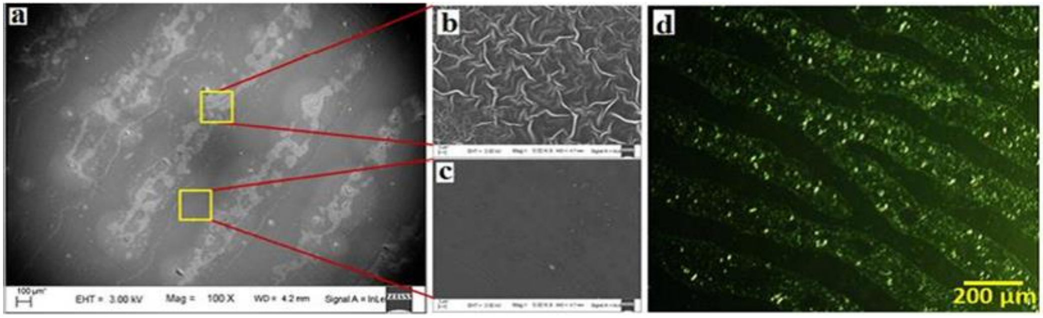

Figure 9. (a) FESEM images LFPs showing adhesion of CPE on ridges (b) and (d) Fluorescence microscopic images of the developed LFPs [92]

During the deposition of polyaniline, a negative visualization of the FPs was generated and clear contrast was obtained by continuously adjusting the applied potential. The FPs on the surface act as an insulator, therefore, deposition of metal NPs happens on the other surface. Malik *et al.* [92] developed highly clear visuals of LFPs using conjugated polyelectrolyte. FESEM image of FP deposited on the surface of PFTPEBT-MI (Figure 9) shows the total adhesion of the electrolyte on the FP ridges. This deposition is due to the hydrophobic fatty acid and the conjugated backbone of electrolyte PFTPEBT-MI. While, the electrostatic interactions is due to interaction of the sweat compounds such as amino acids, proteins, and glucose with the terminal functional groups of the electrolyte. The development of the LFPs on a different surface using ammunition/Cartridge cases were also studied [78,79]. Girelli *et al.* [78] consecutively applied the cyanoacrylate, gun blueing solution and basic yellow 40 for the enhancement of LFPs on both fired and unfired but cycled through the gun cartridge cases. For the former case, low quality of FP ridge details was observed, while for the latter case clear ridge details were obtained without affecting the ridge details due to the cycling. With the help of microscopic examination, Liu *et al.* [78] also tested the effects of heat and fire on the damaging of the ridge details at different pH values. The neutral pH range was chosen as the optimal one.

10. Characterization Techniques

Many analytical tools have been employed in the forensic analysis of varieties of samples and crime scenes. Among these, the SEM, TEM, AFM, DLS, UV-Vis, FTIR, SERs, and MALDI-MSI are the most commonly used methods. SEM is used to evaluate microstructure and density of coverage of the NPs [93, 94]. Similarly, techniques such as AFM [46] for 3-D image detection of FPs ridges, DLS for particle size determination, UV-Vis for enhancement of LFPs with high performance, FT-IR for functional group determination of both ligands and exogenous materials, and SERs for FP analysis as well as identifying chemicals and biological materials present within the ridge. Generally, these techniques assist forensic scientists in two ways: 1) by making it possible to analyze nano-scaled samples and 2) by making use of the specific properties of nanomaterial's to detect and collect evidence, which would not have been possible by previous techniques. With the help of these techniques, the DNA extraction from palm-prints, FPs, gun residues, explosives, and heavy metals are some of the novel approaches that simplify the way for forensic scientists to provide conclusive evidence.

10.1. Scanning and Transmission Electron Microscopy

When compared to optical detection techniques, electron microscopy gives deep information about the morphology and composition of FPs. TEM generates images by detecting primary electrons, whereas, SEM makes images by detecting electrons emitted from the ridges (secondary/back-scattered electrons). With their ability to magnification, SEM and TEM are very beneficial tools in forensic science, which can be applied only for small samples [93,95]. As recommended by Sébastien *et al.* [96] before applying different detection techniques a deeper understanding of the FPs residue (physical and chemical properties) is important. In their study, the ridge morphology (using SEM), simple elements were successfully mapped using energy-dispersive X-ray spectroscopy.

10.2. Atomic Force Microscopy

AFM works with the principle of deflection forces at the FP surface and caterpillar tip. The laser on the top of the cantilever senses the deflection and positional changes of the laser as a result of deflection will be noted and transformed into a 3-D image of the FPs ridges as shown in (Figure 10). AFM tool is very much helpful especially in differentiating various adhesives left behind at crime situations [97]. As studied by Goddard *et al.* [97], it also has the capability of imaging FP ridges and its fine structures.

10.3. Scanning Kelvin Probe

SKP technique works by scanning the metallic surface using a gold electrode which vibrates periodically. The LFPs are scanned by a vibrating gold wire probe by noting the differences in potentials where the FPs are deposited. Challinger *et al.* [98] used the non-contact, non-destructive SKP techniques that work under ambient conditions and the obtained results confirm that FPs are easily imaged without using any other enhancing techniques. Williams and McMurray [99] also used SKP and their work also informs the possibilities of visualizing the FPs indirectly by hidden ridge details.

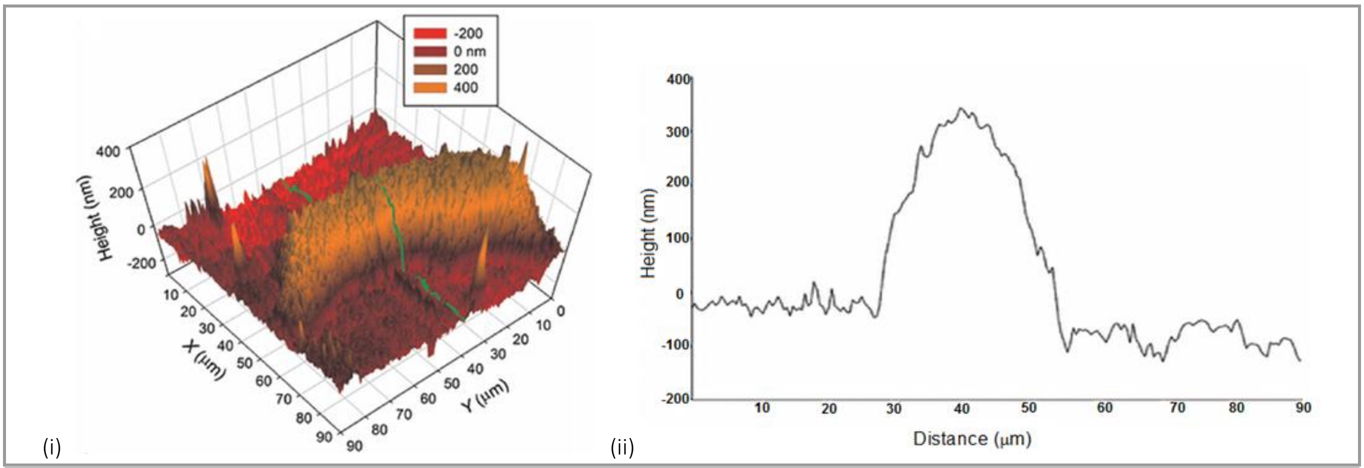

Figure 10. AFM images from the polished and printed brass sample, (i) 3D image showing part of the ridge detail; (ii) 1D lines can across ridge detail, as indicated by a green line across the center of (i) [97].

10.4. Matrix-assisted laser desorption/ionization mass spectrometry imaging

MALDI-MSI is the other FPs imaging technique that works by changing the laser constantly and extracting the exact image with the help of mass spectroscopy. But to do so first, the standard like gluing agent should be used. Bradshaw *et al.* [100] combined known LFP enhancers (powders and powder suspensions, cyanoacrylate fuming), vacuum metal deposition (VMD), Ninhydrin, DFO enhancement with MALDI-MSI and obtained both improved FP development and elemental analysis. Francese *et al.* [101] also suggested the possibility of obtaining information beyond the FPs ridge by taking many images of the same FP in a single analysis. The other recent and accurate method is a time of flight-secondary ion mass spectrometry (TOF-SIMS/SIMS), however, the use of vacuum as its working principle may disturb the ridges of the FP [102].

10.5. Thermo-Desorption (TD)

TD is the other important techniques used for the enhancement of the deposited prints. However, since it uses heat>200 °C, it may damage the point of interest for analysis; it uses on both porous and nonporous surfaces [103, 104]. Vacuum metal deposition also the other method used by depositing metals with the help of a high vacuum chamber [51, 85].

11. Conclusion

FP is the best useful form of evidence required for the identification of an individual in forensic investigation. The three important glands i.e. the eccrine, apocrine and sebaceous glands interact with a catalyst to enhance the prints and give quick results. Up to now, various FPs development techniques including metal oxides, optical, physical, and chemical have been assessed. With these in addition to enhancing, the possibilities of obtaining information about identification and analyzing of the age, sex, etc., have been discussed. Furthermore, among several analytical techniques that are employed in the forensic analysis, the SEM, TEM, AFM, DLS, UV-Vis, FTIR, SERs, and MALDI-MSI are the most commonly used one. For the enhancement purpose, the nature of the surface where the FPs deposited, the stability and sensitivity of the enhancing agent, fluorescence properties of the materials are important factors. The use of elemental states of noble metals such as gold and silver for the enhancement of LFPs may have high stability and sensitivity, however as explained on some papers due to contrast problems these methods may not be successful on dark surfaces. Even if enhancement using chemical methods in combination with different analytical techniques give good enhancement, due to its complex procedure and costly, nowadays enhancement using nanotechnology is becoming a non-destructive, easy, and less costly technique.

Abbreviations

FPs: Fingerprints; LFPs: Latent fingerprints; MMDI/II: Multimetal deposition-I & II; SMD: Single metal deposition; Au-ASP: Gold-aspartic acid; SEM: Scanning electron microscopy; TEM: Transmission electron microscopy; AFM: Atomic force microscopy; UV-Vis: Ultraviolet-visible spectroscopy; NIR: Near-infrared; SERs: Surface-enhanced Raman spectroscopy; SKP: Scanning Kelvin probe; DLS: Dynamic light scattering; MALDI-MSI: Matrix-assisted laser desorption/ionization mass spectrometry imaging; TD: Thermo-desorption.

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No potential conflict of interest was reported by the authors.

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