SCIENCE CHINA
Chemistry

• MINI REVIEWS •

July 2015 Vol.58 No.7: 1090–1096
doi: 10.1007/s11426-014-5294-5

Advances in the development and component recognition of latent fingerprints

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Received September 5, 2014; accepted September 25, 2014; published online January 30, 2015

Fingerprints have been used as an indispensable tool for personal identification in forensic investigations since the late 19th century. At present, fingerprinting technology has moved away from its forensic roots and is incorporating a broader scientific range, e.g., material science, spectroscopy and spectral analysis, and even *in vitro* diagnosis. After a brief introduction to latent fingerprints, this mini-review presents the pioneering progresses of fingerprinting technologies including (i) material and electrochemical techniques, and (ii) spectral and spectroscopy imaging techniques and immunological techniques capable of both the visualization of a fingerprint and the detection of chemicals present in it. Finally, perspectives on this rapidly developing field are discussed.

latent fingerprints, recognition, immunoassay, aptamer, mass spectrometry

1 Introduction

The corrugated skin at the ends of human fingers is characterized by a pattern of raised ridges and recessed furrows that is truly unique to an individual, invariable throughout a person's lifetime, and cannot be easily forged. Whenever a fingertip touches the surface of an object, materials on the protuberant ridges are transferred to the surface and will leave a fingerprint [1,2]. This process represents a typical example of the classic Locard's principle commonly expressed as "every contact leaves a trace" [3]. Therefore, a fingerprint can incontrovertibly demonstrate the presence of an individual at the scene of a crime. Since the first use of fingerprints to obtain a criminal conviction [4], the identification of fingerprints has been the cornerstone of modern criminal investigations despite the rise of other methods such as those based on DNA [5].

The most common form of fingerprint evidences at crime scenes is latent fingerprint (LFP), which is invisible to the naked eye and requires the application of "development" techniques to enable its visualization [1]. The principle of fingerprint development is to generate a distinct contrast between the secretion residue constituting the fingerprint pattern and the underlying substrate [6]. Because the success of this endeavor depends heavily on the chemicals within the fingerprint residue, precise knowledge about these components is particularly useful. The chemicals of LFPs mainly originate in the pores along the skin ridges through which human sweat is excreted [6,7]. There are three types of secretion glands distributed throughout the human body: eccrine, sebaceous, and apocrine [8,9]. Eccrine glands are present all over the body but have higher density in hands, sweat from which contains 98%–99% water, a number of inorganic salts (e.g., chloride and phosphate), and organic materials (e.g., amino acids, urea and polypeptides). Sebaceous glands are found all over the body except on the surfaces of hands and feet. They secrete sebum, which consist of fatty acids, wax esters, and squalene [10]. Apocrine glands are found primarily in the genital,

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breast, inguinal and axillary regions, and thus play a minor role in fingerprint composition [6]. Therefore, fingerprint residue is composed mostly of eccrine sweat, as well as sebaceous secretions due to the contact of bare hands with faces or hair. In addition, contaminants such as cosmetics or other exogenous components can also be found in fingerprint residues.

To date, a range of chemical and physical methods based on chemical or physical interactions of the chosen reagent with the fingerprint residue have been developed for the visualization of LFPs [11,12]. These include powder dusting, fluorescent dyes, ninhydrin/1,2-diazafluoren 9-ONE staining, cyanoacrylate/iodine fuming, and vacuum metal deposition [13,14]. Synthetic techniques [11,15–18], metal [19,20] and semiconductor nanoparticles (NPs) [13,21,22], as well as electrochemical methods [23–25], have also been exploited. In addition, the past few years have witnessed a renewal of research interest in the possibility that a fingerprint can provide additional information about the donor than mere identification [7]. To this end, numerous methods including infrared [26] and Raman spectroscopy [27], mass spectrometry (MS) [28], and immunological techniques [29,30] have been utilized.

In this mini-review, we divide the current research interest in the fingerprinting technology into two categories:

(i) The improvement of enhancement techniques for exploiting more portable and efficient manners; and (ii) the development of component recognition for elucidating advanced knowledge about fingerprint composition, such as gender [31], diet [32], the presence of human metabolites with diagnostic values [33] and the evidence of contact with explosives or illicit drugs [34].

The aim of this mini-review is to highlight the methods and techniques that have been focused on these two objectives since 2007. Use of immunological techniques in the recognition of components in LFPs are emphasized.

2 New methods for the enhancement of latent fingerprints

Among various processes that have already been proposed for the development of LFPs, fingerprint powdering is still the most widely used method at crime scenes. However, brushing magnetic or luminescent powders onto LFPs has some drawbacks, such as the inevitable destruction of the fingerprint details and the health hazard it poses to the examiners. Therefore, current studies are focusing on extending the ability to enhance the visualization of LFPs in simple, rapid, cost-effective, and user-friendly ways.

2.1 New materials

Rapid advances in nanoscience have increased research interest in the application of NPs for fingerprint enhancement [13]. Sametband *et al.* [22] found that CdSe/ZnS quantum dots (QDs) could develop LFPs deposited on silicon wafers under UV illumination. Xu *et al.* [35] recently reported the rapid imaging of eccrine LFPs by using nontoxic Mn-doped ZnS QDs. The multimetal deposition (MMD) introduced by Saunders *et al*. [36] in 1989 is a very sensitive method for visualizing LFPs on a range of surfaces. Since then, a great deal of effort has been devoted to reducing the labor-intensity of this procedure [37–39]. Recently, MMD has been performed in a reverse manner by developing a paper substrate rather than fingerprint residue [40,41]. A review of applications to nanomaterials in LFPs' development was recently published [13].

Fluorescence is a general approach for the detection of LFPs because of its high sensitivity. For example, a highly fluorescent conjugated polymer film has been used for nondestructive fingerprint detection based on swelling-induced emission created by the readily migration of oily components in LFPs into the film through the microvoids [15]. Yang *et al.* [17] have demonstrated a novel method for the ultrafast recognition of LFPs on various surfaces by using commercial thermoplastic polyurethane resin and an electrospinning technique with a fluorescein release-induced response. Our group has reported an alternative fluorescent approach to enhance the visualization of LFPs, based on aggregation induced emission (AIE) [18,42]. Because the overall process is performed in solution and does not involve powder treatment, it is more user-friendly.

2.2 Electrochemical techniques

Shan *et al.* [43] have demonstrated an electrochemical microscopy technique for visualizing LFPs and analyzing trace chemicals, by detecting the variation in local electrochemical current from optical signals of surface plasmon resonance (SPR). Scanning Kelvin probe technique [25] and scanning electrochemical microscopy (SECM) [23,44,45] with microscale resolution have also been employed for visualizing LFPs on a variety of surfaces. Hillman *et al.* [11,12] have demonstrated the visualization of LFPs by means of spatially selective deposition of electrochromic polymers (e.g. polyaniline and poly(3,4-ethylenedioxythiophene)). Recently, our group developed an electrochemiluminescence (ECL) imaging method for visualizing LFPs by the spatially selective control of the location of ECL generation (Figure 1), using $Ru(bpy)_3^{2+}$ [46] or rubrene [47] as the ECL luminophore. This method is particularly useful in visualizing LFPs on metal surfaces, e.g., stainless steel [48], that are often found in felony cases. Recently, Tan *et al.* [49] demonstrated the visualization of oily LFPs and *in situ* detection of TNT in fingerprints by using an ECL-based image-contrast technology on the surface of porous silicon. Unlike the ECL based on molecular luminescence, this technology does not require a co-reactant.

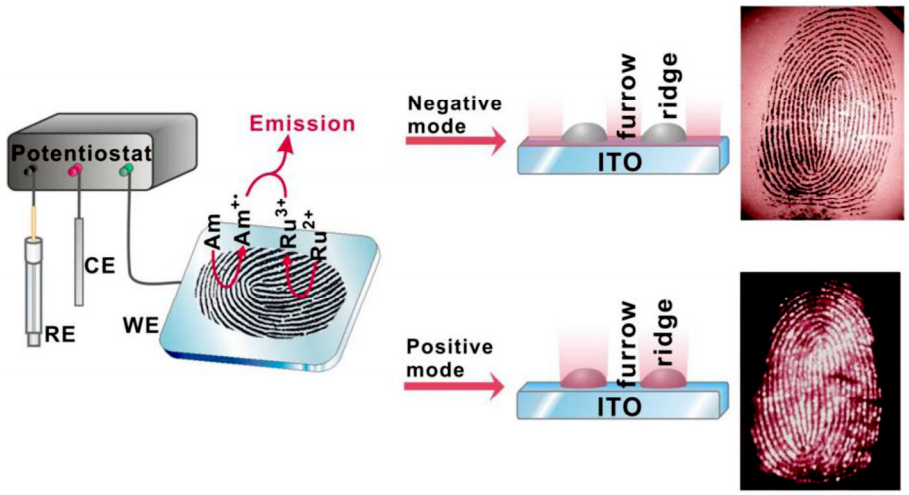

Figure 1 A diagram of a typical ECL imaging system and the imaging strategy for visualizing LPFs in negative and positive modes. Ru^{2+} and Am respectively represent the ECL-generating luminophore and the amine co-reactant. The negative and positive modes were performed by selectively controlling the ECL generation at the bare electrode surface uncovered by the fingerprint (negative mode), or at the fingerprint ridges (positive mode). Reproduced from Ref. [46] with permission from John Wiley and Sons.

3 Component recognition of latent fingerprints

Because a fingerprint can provide additional information about a person than mere identification [7], interest has been increasing in extending the range of instrumental and immunological techniques in order to find new ways for the sensitive detection of components that are present in LFPs.

3.1 Mass spectrometry

Various compounds that originate in eccrine and sebaceous glands, such as lactic acids, amino acids, urea, fatty acids, wax esters, diglycerides, triglycerides, monoglycerides, cholesterol esters, squalene, and cholesterol, have been quantitatively detected by gas chromatography coupled with MS (GC-MS) in early studies [10]. However, GC-MS is a destructive technique because fingerprints need to be swabbed, extracted, and derivatized for analysis. Desorption electrospray ionization (DESI) MS is a minimally destructive and ambient technique that features the advantage of *in situ* analysis without sample preparation from ordinary samples in their native environment [5]. In 2008, the Cooks's group [34] that originally developed this technique showed that DESI MS can be used for the chemical imaging of endogenous components such as *cis*-hexadec-6-enoic acid, stearic acid, *cis*-octadec-8-enoic acid, palmitic acid, pentadecylic acid, myristic acid, and triacylglycerols in sebum-rich LFPs, as well as small amounts of illegal drugs such as cocaine and THC, and explosives such as RDX in contaminated LFPs. Recently, chemical imaging techniques utilizing surface-assisted laser desorption/ionization time-of-flight MS (SALDI-TOF-MS) and matrix-assisted laser desorption ionization MS (MALDI-MS) have significantly improved the study of fingerprint composition because of their ability to identify and map the substances present in LFP residues [50–55]. Most studies are devoted to finding new substances that are capable of increased photon-energy absorption from the incoming laser in place of traditional matrixes. For example, silica dusting agent [56], gold NPs [28], curcumin [57], and silver clusters [58] have been integrated with MS imaging for both the visualization and molecular imaging of LFPs. Recently, a review on the multi-informative analysis of LFPs by MS was published [5]. Future research should be devoted to identifying proteins/polypeptides using advanced MS techniques, because they represent the most abundant group of compounds in the fingerprint residues that originate in eccrine glands [10].

3.2 Vibrational spectroscopic imaging

Fourier transform infrared (FTIR) spectroscopy and Raman spectroscopy can produce visual images of fingerprints by mapping the functional groups of the molecules. Ricci *et al.* [59] have studied the distribution of lipids and amino acids in the fingerprints, using an *in situ*-attenuated total reflection FTIR (ATR-FTIR) imaging approach. Changes in the spectra of lipids with temperature and time have been detected, which are potentially important in understanding the aging process of LFPs. Bhargava *et al.* [60] have reported the use of reflection-absorption mode for the distinction of overlapping fingerprints based on differences in their chemical origins. FTIR imaging has been used to detect exogenous trace residues associated with forensic evidence, e.g., explosives [60–62] and illegal substances [63].

Raman spectral mapping has also been used to extract chemical information from LFPs and to identify trace amounts of substances via their unique Raman spectral signatures [27]. Compared to the conventional Raman tech-

nique, surface-enhanced Raman spectroscopy (SERS) can give more detailed chemical information about target analytes because of its enhanced sensitivity of 10^4–10^6 [64]. For example, SERS has been utilized to recreate visual images of fingerprints that are undetectable by conventional methods [65]. Although FTIR and Raman spectroscopy are the least-destructive spectroscopic techniques, they have much poorer chemical specificity than MS. To resolve this limitation, Song *et al.* [64] demonstrated an indirect approach for the identification of specific proteins deposited within an LFP by employing an antibody/silver NPs/Raman probe conjugate. This conjugate could indirectly provide chemical information on the targeted proteins through specific immune recognition and thereby provides the possibility of detecting biomolecules in LFPs, such as those valuable for medical diagnostics or criminal investigations.

3.3 Immunological techniques

The first use of immunoassay for the detection of forensic secretions within the sweat deposited in LFPs was reported by Leggett *et al*. [33] and then highlighted by Wolfbeis [66]. As shown by these studies, smokers' fingerprints can be visualized by incubation with gold NPs functionalized with antibody specific to cotinine and secondary antibody tagged by a dye. Fluorescence image of fingerprints treated in this way can clearly reveal second- and third-level details, which shows the capability to both identify an individual and to simultaneously determine the sweat chemicals deposited in that individual's fingerprint. Later, this group used magnetic particles functionalized with a range of antibodies for the detection of metabolites for controlled drugs such as methadone, cocaine, and heroin [29,67]. A key advantage of the detection of drug metabolite is that it can prove the sumption of a particular drug, whereas detection of a drug in a fingerprint can only prove that an individual touched it. In further studies, anti-cotinine/magnetic particle conjugates were successfully applied to fingerprints deposited on highly reflective white porcelain surfaces [68] and fingerprints were aged under various conditions [69].

Spindler *et al.* [32] have shown that gold NPs conjugated with anti-*L*-amino acid antibodies can visualize LFPs on non-porous surfaces. Using dermcidin as the antigen of interest, van Dam *et al.* [70] investigated the compatibility of immunolabeling with two commonly used fingerprint visualization techniques: magnetic powdering and ninhydrin staining. It was shown that LFPs pre-developed by magnetic powder or ninhydrin did not inhibit the specific detection of dermcidin.

Our group has been working on new methods for the detection of various secretions within eccrine fingerprints by combining immunoassay with portable and efficient imaging techniques. Figure 2(a) illustrates the general principle of the enzyme immunoassay and the ECL imaging for the detection of secretions in LFPs [71]. First, the primary antibodies specific for target analytes were incubated with the fingerprint sample. Second, the sample was sequentially processed by incubation with biotinylated secondary antibodies and HRP-labeled streptavidin. The application of a sufficiently negative voltage to the electrode carrying the fingerprint led to the electrochemical reduction of dissolved oxygen to H_2O_2. Then HRP can catalyze the ECL reaction of H_2O_2 with luminol, thus yielding an ECL image of the fingerprint. Specifically, we detected epidermal growth factor (EGF), lysozyme, and dermcidin, all of which are polypeptides secreted by human eccrine sweat glands. The obtained ECL images (Figure 2(b–d)) not only reflect the unique ridge pattern of a fingerprint but also provide the chemical evidence of the presence of specific secretions. Because the light emission is triggered by the electrochemical reaction, the ECL imaging approach allows spatial and temporal control of the light-emitting reaction; in addition, because it does not involve a light source, it eliminates the interference of scattered light and luminescent impurities.

In another work, we have developed a modified MMD method, termed immunological MMD (iMMD), by combining the immunoassay with the conventional MMD [72]. As shown in Figure 3(a), iMMD uses antibody-modified gold NPs that bind with the corresponding antigens in the fingerprint residue via specific immunoreactions. The AuNPs then serve as the catalytic nucleation sites for the metallic deposition of silver particles from the silver staining solution, a process that eventually enhances a black fingerprint on a lighter background for visual detection. With this formulation, we successfully accomplished the visualization of LFPs via the detection of two secreted polypeptides: EGF (Figure 3(b)) and lysozyme (Figure 3(c)). Unlike the conventional MMD, AuNPs used in iMMD served not only as the nucleation sites for the deposition of silver particles, but also as the carriers of recognition molecules that could verify the existence of specific components. Another significant advantage of iMMD is that the developed fingerprints can be directly observed by the naked eye, without involving any sophisticated imaging equipment.

3.4 Aptamer-based reagents

Aptamers are short single-stranded nucleic acids that bind to a number of targets such as metal ions, proteins, and even whole organisms [73]. In 2012, Wood *et al.* [30] reported for the first time the use of the unique recognition power of aptamers to detect lysozyme in LFPs. When incubated with the fingerprint, the fluorophore-tagged aptamers folded into specific 3D structures and subsequently bound to lysozymes. Recently, Li *et al.* [74] proposed a nanoplasmonic method by employing aptamer-bound AuNPs to visualize LFPs and detect contact residues of cocaine. Because the localized SPR of AuNPs is highly dependent on the interparticle distance, the cocaine-induced aggregation of aptamer-bound AuNPs led to a true green-to-red color change of the scatter-

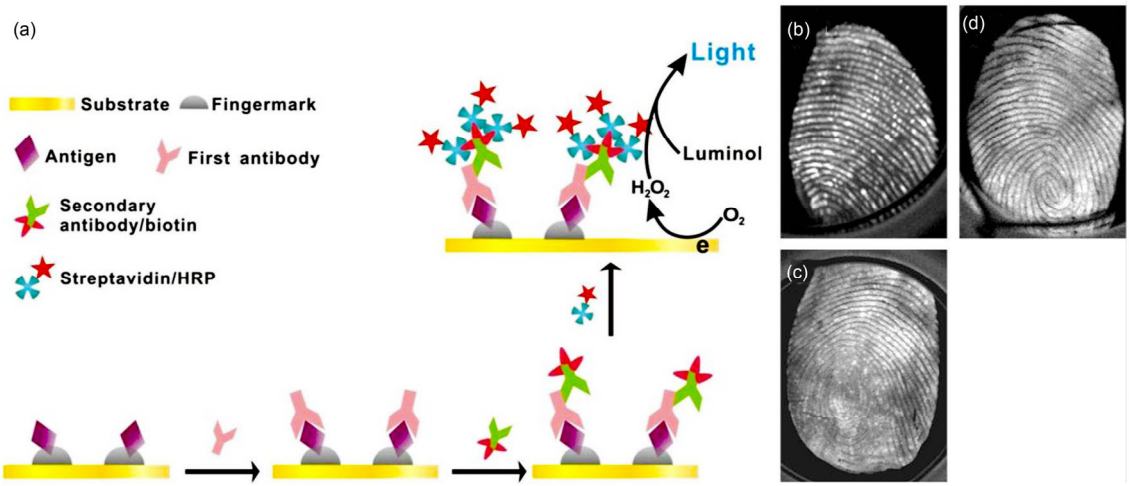

Figure 2 Schematic illustration of the detection of antigenic secretions deposited within a fingerprint using enzyme immunoassay and ECL imaging (a), and ECL images of the as-treated eccrine fingerprints for the detection of EGF (b), lysozyme (c), and dermcidin (d). Reproduced from Ref. [71] with permission from the Royal Society of Chemistry.

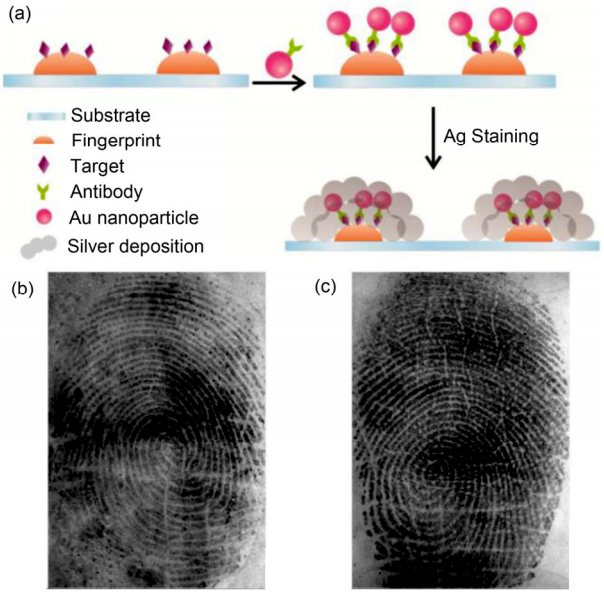

Figure 3 Illustration of the iMMD process involving the binding of AuNPs/antibody conjugates with specific secretions and silver staining enhancement (a), and optical images of eccrine fingerprints via the detection of EGF (b), and lysozyme (c). Reproduced from Ref. [71] with permission from John Wiley and Sons.

ing in the dark-field image, thus providing the molecular recognition of cocaine loaded into the LFPs. The minimally detectable cocaine loaded was calculated as 90 ng. Yuan *et al.* [75] employed upconversion NPs (UCNPs) functionalized with aptamers for the detection of fingerprints through recognizing lysozyme. The fingerprints treated with UCNPs/aptamer conjugates displayed a better luminescence image than those treated with fluorescein or CdTe QDs, due to the ability of UCNPs to suppress background fluorescence.

Recently, a review has been published that details the immunogenic and aptamer techniques for the selective detection of fingerprints [76].

4 Summary and outlook

This mini-review highlights the advanced applications of new materials and analytical techniques in the field of latent fingerprint detection. A variety of functional materials such as QDs, gold NPs, conjugated polymer film, thermoplastic polyurethane resin, and fluorophores with the AIE effect, have been extensively studied in order to enhance the visualization of LFPs in a more portable and efficient manner. Taking into account the advantages of low cost, high sensitivity, and good controllability of reactions, pioneering electrochemistry techniques including electrochemical microscopy in conjunction with SPR, scanning Kelvin probe, SECM, electrodeposition, and ECL imaging, have been applied to the detection of LFPs. Another important area of fingerprint detection is the use of chemical imaging techniques (e.g., mass spectrometry imaging, FTIR, and Raman spectroscopic imaging), immunological techniques, and aptamer-based reagents. These approaches have shown great potential not only for the effective visualization of LFPs, but also for the detection of various kinds of endogenous metabolites and exogenously doped species such as illegal drugs and explosive materials, which has promise for diagnostic and safety purposes. Although considerable achievements have been reached in fingerprinting technology, further research is still required to (i) implement these new approaches in routine forensic investigations; (ii) detect many more species and gain their quantitative information from fingerprint residues, particularly the polypeptide/protein components with the help of advanced mass spectrometry techniques; (iii) extract more valuable information

from fingerprint composition such as the aging process of a fingerprint; the person's gender, diet, and medical conditions; and the evidence of a person's contact with explosives or illegal drugs. Finally, we envisage that there is enormous room for chemical researchers to extend the capability of fingerprint detection in both analytical and forensic areas in ways that take advantage of rapid progress in the areas of material science, synthetic techniques, advanced spectroscopy, and immunological and aptamer techniques.

This work was supported by the National Natural Science Foundation of China (21222504, 21335001), the Program for New Century Excellent Talents in University, the Scholarship Award for Excellent Doctoral Student granted by the Ministry of Education, the Nature Science Foundation of Zhejiang Province (R14B050003) and the Fundamental Research Funds for Central Universities (2014XZZX003-04).

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