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Fluorescent Nanomaterials for the Development of Latent Fingerprints in Forensic Sciences

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Abstract

This review presents an overview on the application of latent fingerprint development techniques in forensic sciences. At present, traditional developing methods such as powder dusting, cyanoacrylate fuming, chemical method, and small particle reagent method, have all been gradually compromised given their emerging drawbacks such as low contrast, sensitivity, and

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selectivity, as well as high toxicity. Recently, much attention has been paid to the use of fluorescent nanomaterials including quantum dots (QDs) and rare earth upconversion fluorescent nanomaterials (UCNMs) due to their unique optical and chemical properties. Thus, this review lays emphasis on latent fingerprint development based on QDs and UCNMs. Compared to latent fingerprint development by traditional methods, the new methods using fluorescent nanomaterials can achieve high contrast, sensitivity, and selectivity while showing reduced toxicity. Overall, this review provides a systematic overview on such methods.

1. Introduction

The corrugated skin at the end part of human fingers is characterized by a complex pattern of raised papillary ridges and depressed furrows.[1] The papillary ridge patterns, which remain topologically unchanged from birth of an individual, differ not only from one individual to another but also from one finger to another.[2] When the surface of an object is touched by a finger, aqueous ectocrines such as sweat and oily substances such as sebum can be transferred and deposited onto the surface, resulting in the formation of a fingerprint.[3] Therefore, fingerprints represent the contact impression of the lifted papillary ridge of skin.[1] Due to the complexity, uniqueness, and stability of the papillary ridge patterns, fingerprints formed through papillary ridges have been and still are considered the best reference for personal identification in forensic sciences.[4] Since the first use of fingerprints for personal identification suggested in the late 19th century, fingerprints have become a well-established signature for criminal investigation and personal identification.[5]

Generally, three types of fingerprint evidence are common at crime scenes, including impression (or indented) fingerprints, visible (or patent) fingerprints, and latent fingerprints.[6] Among them, latent fingerprints are the most common at crime scenes, namely, they are not obviously visible to the naked eye. However, the latent fingerprints can be made visible with the use of certain developing techniques.[6] If a distinct contrast can be generated between the fingerprint residues and the underlying substrate, the fingerprint can be developed. Over the past century, many fingerprint development approaches have been studied, including optical, physical, and chemical processes.[7,8] If a latent fingerprint left at a crime scene cannot be developed clearly by a valid method, proper fingerprint analysis and identification will be difficult to achieve. Therefore, latent fingerprint development is crucial to identifying individuals in forensic sciences.

Over time, many researchers have explored a variety of methods to promote and improve latent fingerprint developing to assist with the efficiency of criminal investigations and personal identifications. Among them, powder dusting, cyanoacrylate fuming, silver nitrate method, ninhydrin method, 1,8-Diazafluoren-9-one method, and small particle reagent method, are the most widely used due to their simplicity, efficiency, and ease of operation.[9] However, these traditional developing methods have drawbacks. For instance, the contrast, sensitivity, and selectivity of the development are low while the toxicity of the developing reagents is high. Recently, the employment of new techniques such as electrochemiluminescence and immunological detection for the highly selective development of latent fingerprints have been used.[10–16] However, the actual operation of

these two techniques is relatively complex, thus dampening their practical use at crime scenes. Therefore, there is an urgent need in seeking a simple and efficient method for developing the latent fingerprints with improved contrast, sensitivity, and selectivity, as well as low toxicity.

Fluorescent nanomaterials (NMs) such as quantum dots (QDs) and rare earth upconversion fluorescent NMs (UCNMs) have emerged as new agents for developing latent fingerprints, due to their unique optical and chemical properties. They exhibit many advantages such as small size, high fluorescent intensity, good chemical and photo stability, facile surface modification, and low toxicity. We will start with a brief overview on the current state of traditional developing methods as shown in Scheme 1, then highlight recent advances of latent fingerprint development techniques by using various fluorescent NMs including QDs and UCNMs.

2. Traditional Methods for Latent Fingerprint Development

2.1. Powder Dusting Method

The powder dusting method is one of the oldest and more prevalently applied methods for latent fingerprint development on nonporous substrates.[6,9] It has been in use since the late 19th century.[6,9] In the powder dusting method, the fingerprint powder particles are mechanically or physically adhered to the aqueous or oily components present in the latent fingerprint residues.[6,9]

Regular powder, metallic powder, and fluorescent powder are generally used at crime scenes.[7] Regular powders usually consist of both resinous polymer materials (e.g., starch, kaolin, rosin, silica gel, etc.) for adhesion and colorants (e.g., bronze flake, aluminum flake, dolomite powder, etc.) for developing contrast. Magnetic powders are usually composed of both magnetic particles as a carrier and nonmagnetic colorants (e.g., carbon black powder, bronze flake, aluminium flake, fluorescent powder, etc.) as a developer. In 1961, MacDonell et al. first reported the use of magnetic powders for latent fingerprint development.[17] Fluorescent powders were introduced for latent fingerprint development in the late 20th century, after the laser was used for visualizing latent fingerprints. In 1977, coumarin-6 was first employed as a fluorescent powder for developing the latent fingerprints along with the use of an argon ion laser.[18] Fluorescent dyes such as acridine yellow, acridine orange, crystal violet, Nile blue, Rhodamine B, and Rhodamine 6G were commonly used as fingerprint powders, with the excitation of forensic light sources and the appropriate use of barrier filters. Fluorescent powders have been proposed for use on surfaces with complex background color or texture, making it difficult to visualize fingerprints developed by other non-fluorescent fingerprint powders, in particular on multicolored surfaces.

The simple application of powder to latent fingerprints is done with careful brushing; it is efficient and inexpensive, and can yield apparent fingerprints instantly on almost all smooth and nonporous substrates. However, the powder dusting method has its own drawbacks. For example, it may bring about a potential health hazard at a crime scene. In addition, there is a concern regarding possible contamination of DNA samples via transfer from the fingerprint brushes.

2.2. Cyanoacrylate Fuming Method

The Cyanoacrylate fuming method, also known as the Super Blue fuming method, is commonly applied for developing latent fingerprints on nonporous surfaces. It was almost simultaneously devised in the late 20th century.[6,9] The method involves the quick polymerization of cyanoacrylate ester monomers on latent fingerprint residues. Generally, the vaporized monomers are introduced to the latent fingerprints and they then form a rapid bond with initiators (e.g., water, acid, alkali, etc.) in the fingerprint residues, and then the previously introduced monomers react with the rest of monomers in the vapor to form a white-color, durable polymer that covers the raised papillary ridges.[6,9]

There are some effective techniques to reduce the fuming time by promoting the volatilization of cyanoacrylate ester monomers, including fume circulation, heat acceleration, chemical acceleration, and vacuum acceleration. Fully developed latent fingerprints by cyanoacrylate fuming appear as a white three-dimensional matrix, which often lacks contrast and making visualization challenging. The fumed fingerprints can be further enhanced via a multitude of methods. The most commonly used practice is simply to dust the fumed fingerprints with fingerprint powders, especially fluorescent powders.

The cyanoacrylate fuming method is a versatile and effective technique to develop the latent fingerprints on virtually all nonporous substrates, especially rough surfaces. However, there are a number of health and safety issues associated with this method. For instance, the liquid cyanoacrylate esters and their vapors have the ability to cause severe damage to the skin, eyes, and mucous membranes.[19] In addition, highly toxic hydrogen cyanide gas could be formed when cyanoacrylate ester monomers or polymers are heated.[13]

2.3. Silver Nitrate Method

The silver nitrate method is an old approach to developing latent fingerprints on the porous surfaces. This method has been in use since the late 19th century.[6,9] It involves the reaction of silver nitrate with chloride in the fingerprint residues, resulting in the formation of silver chloride. Then, upon light irradiation, the stains become black because silver ions in the chloride are reduced to elemental silver, enabling the visualization of the fingerprints.

Initially, silver nitrate solution was applied for developing fingerprints on porous surfaces such as paper and wood. Later, Trozzi et al. utilized ethanol-based 3% silver nitrate solution for developing latent fingerprints on water-repelling materials (e.g., waxed paper).[20] In their work, ethanol was used for decreasing the dissolution rate of NaCl in the residues of the fingerprints in order to achieve surface wetting and subsequent accelerated evaporation. Under an ordinary interior luminous environment, the conversion from silver chloride to elemental silver via photo-reduction is slow. However, the photo-reduction can be hastened under ultraviolet (UV) lights and the reaction efficiency is inversely proportional to its wavelength (e.g., a shorter wavelength of 254 nm will outperform a longer one of 365 nm).

The silver nitrate method is a simple and effective technique to develop latent fingerprints on normal porous substrates and some water-repelling surfaces. However, it is suggested that the age of latent fingerprints should not be older than one week. The major drawback of the silver nitrate method lies in the potential decrease in contrast due to background stains.

2.4. Ninhydrin Method

The ninhydrin method is now commonly used in developing latent fingerprints present on the surface of porous materials (e.g., paper, cardboard, raw wood, and plasterboard). This technique was first proposed in the mid-20th century.[6,9,21] In this technique, ninhydrin reacts with amino acids that are usually present in fingerprint residues. The reaction results in the formation of a product (Ruhemann's purple) with deep color, rendering the latent fingerprints visible.[6,7,9]

On contact with porous substrates such as paper, the amino acids impregnated in the surface of substrates are stable and do not significantly migrate over the time,[22] indicating that amino acids are desirable targets for the development of aged fingerprints. Therefore, this method aims to develop aged fingerprints. The ninhydrin reaction in fingerprint development often requires strict control of reaction conditions: (i) the pH of the reaction must be above 4, and the ideal pH range should be 4.5–5.2; (ii) a high-humidity (50%–80%) environment is also required because water is a necessary reactant; and (iii) the treated fingerprints should remain cool in the dark since exposure to light and oxygen will degrade Ruhemann's purple. Heat or steam treatment is often carried out to accelerate the rate of ninhydrin reaction. At the same time, the elevated temperature can cause serious background discoloration, leading to a decreased developing sensitivity and contrast.

The ninhydrin method is now most often used for developing the latent fingerprints on paper and other porous substrates because it is relatively simple, effective, and has low toxicity.[7] In addition, aged latent fingerprints can be developed by this method. However, optimal outcomes always result from skillfulness and experience due to the relatively rigorous reaction conditions.

2.5. 1,8-Diazafluoren-9-one Method

The 1,8-diazafluoren-9-one (DFO) method is an efficient method with a high sensitivity for latent fingerprint development on various porous substrates. This approach was proposed in the late 20th century.[6,9,23,24] Similar to the ninhydrin method, the DFO method works by allowing DFO to react with the amino acids present in fingerprint residues, forming a faint red or pink colored product. This product is intensely fluorescent under green light, rendering the latent fingerprints visible.[6,7,9]

Unlike the ninhydrin method, the DFO reaction in fingerprint development often requires a high-temperature, low-humidity environment. The fingerprints developed by the DFO method can be fluorescently detected at room temperature. Post-treatment is not needed to induce fluorescence at a low temperature, and the total number of identifiable fingerprints developed by the DFO method is considerably higher than that of the ninhydrin method. Therefore, this method is a more sensitive and efficient developing technique than the ninhyrin method. Despite this, if ninhydrin is used after DFO treatment, further development is achieved due to the production of Ruhemann's purple. Because DFO cannot react thoroughly with all the amino acids in the fingerprint residue, this leaves some of the amino acids free to react with ninhyrin. Integrating the DFO and ninhydrin method can reach an improved development sensitivity compared to the DFO or ninhydrin method alone.

Therefore, the most current developing sequence recommends the use of DFO prior to ninhydrin for porous surfaces.

At present, the DFO method is considered very suitable for fluorescently developing latent fingerprints on porous substrates due to its high sensitivity, high efficiency, and easy operation.[7] However, the toxic and carcinogenic properties of DFO should not be ignored.[25]

2.6. Small Particle Reagent Method

The small particle reagent method is efficient for latent fingerprint development in some particular cases and was invented in the late 20th century.[6,9] This method relies on the interaction of fine particles with the oily or fatty fingerprint residue; the particles are suspended in a treatment solution and the adherence of the particles to the residue allow the latent print to be developed.[6,9]

Generally, the small particle reagent is a suspension containing fine particles, surfactant, and water. For this method, regular powders and fluorescent powders are often used. Surfactants such as sodium dodecyl sulphate (SDS) can be used in this method. The concentration of the surfactant is found to be critical: a lower concentration results in random particle deposition, but a higher concentration could cause the fingerprint residues to be washed away by water. The small particles bind to the fatty/oily components of the fingerprint residues due to the addition of a surfactant; this is only feasible due to the relative stability of these components maintained over time. Therefore, aged fingerprints can be developed by using the small particle reagent method. In addition, latent fingerprints on nonporous surfaces that have been wetted or immersed in water for a long time can be developed by using this method.

The small particle reagent method is an effective supplement to the powder dusting method for developing fingerprints, such as wet fingerprints, water-immersed fingerprints, aged fingerprints, and other ones on nonporous substrates under particular circumstances.

3. Problems of Traditional Methods for Latent Fingerprint Development

3.1. Low Contrast

The concept of contrast in fingerprint development, similar to the meaning of signal-to-noise ratio, means the contrast between the substrate background and the fingerprints. Figure 1 shows the images of latent fingerprints developed with a high (Figure 1a) and a low (Figure 1b) contrast. There are two main pathways to improve the developing contrast: (i) to improve the signal and (ii) to reduce the background (noise). To improve the signal, fluorescence enhancement is the most widely used technique. To reduce the noise, avoiding background color distraction and decreasing background fluorescent interference are the two effective approaches.

In the powder dusting method, the practicality of nonfluorescent fingerprint powders, such as regular powders and metallic powders, is seriously limited by the color of the substrate. Generally, fluorescent powders are often used to enhance the developing signal due to their strong fluorescent emission. However, the excitation of fluorescent powders usually requires

UV light radiation. Such high-energy UV light radiation can easily cause the substrates to emit fluorescence, leading to an increased background fluorescence interference. In the cyanoacrylate fuming method, the fumed fingerprints with white coating provide a low developing contrast on white-colored, or some light-colored, surfaces. In chemical methods including the silver nitrate method and ninhydrin method, the problem of low developing contrast still exists. In addition, the substrates can also be stained by improper operations, leading to an increased background interference. In the small particle reagent method, the problem of low developing contrast is quite similar to that in the powder dusting method due to the use of traditional powders in both approaches.

3.2. Low Sensitivity

The concept of sensitivity can be described by the visibility and clearness of the ridge details. Figure 2 shows the images of latent fingerprints developed with a high (Figure 2a) and low (Figure 2b) sensitivity. Many complex factors can influence the developing sensitivity, such as the size, shape, and tackiness of the powder particles, the component and properties of the chemical reagents, the developing skills of the operator, and the developing condition controls. There are some pathways to improve the developing sensitivity: (i) to control the synthesis of high-quality fingerprint powders with proper size, shape, and tackiness; (ii) to promote the operating skills; and (iii) to strictly control the developing conditions.

In the powder dusting method, almost all of the used fingerprint powders were micron-sized or even larger in size. One of the drawbacks for using micron-sized or larger-sized powders is that some of the tiny ridge details, especially the sweat pores, tend to be thickly coated by the larger particles, resulting in a decreased sensitivity. In addition, the powders with high tackiness or/and flake-like shape, such as gold powder and silver powder, are inclined to cover or fuzz up the tiny ridge details of the fingerprints. In the cyanoacrylate fuming method, the fingerprints tend to be over-fumed, and the tiny ridge details of the fingerprints will be heavily covered by the white polymers, resulting in a decreased developing sensitivity. In chemical methods including the silver nitrate method, ninhydrin method, and DFO method, the fingerprint residuals can be easily dissolved on the surfaces of the substrates by some organic solvents, which results in a decreased developing sensitivity. In addition, the sweat pores are rarely developed on porous substrates because the sweat pores cannot be reflected well on porous substrates with infiltrating properties. In the small particle reagent method, the problems of low developing sensitivity are quite similar to the situations in the powder method because traditional powders are often used in small particle reagent methods. The immersion time may be a key factor in affecting the developing sensitivity. In other words, immersion for too long can cause the heavy coverage of the tiny ridge details, which reduces developing sensitivity.

3.3. Low Selectivity

The concept of selectivity in fingerprint development refers to the specificity of developing materials (e.g., chemical reagents, fingerprint powders, etc.) in adhering or reacting only with the papillary ridges in latent fingerprints, but not with the furrows on substrates. Figure

3 shows images of latent fingerprints developed with a high (Figure 3a) and a low (Figure 3b) selectivity.

In the powder dusting method, the developing selectivity is usually not high, due to the relatively low selective physical adherence of fingerprint powder particles to latent fingerprint residues. Particularly, when fingerprint powders are applied onto the humid substrates, the developing selectivity will be quite low, due to strong adherence of fingerprint powders to humid surfaces. In the cyanoacrylate fuming method, the developing selectivity is typically high under proper operation, due to the highly selective polymerization reactions between the cyanoacrylate ester monomers and the fingerprint residues. In chemical methods including the silver nitrate method, ninhydrin method, and DFO method, the furrows on the substrates can be easily stained due to the diffusion of active components, such as chloride ions and amino acids, on some infiltrating substrates spontaneously or due to improper operation. This usually results in a decreased developing selectivity.

3.4. Low Toxicity

The concept of toxicity in fingerprint development refers to not only the toxicity of the developing materials and the corresponding equipment to humans but also the damage to DNA in fingerprint residues. Nowadays, with the rapid development of the DNA detection technique, the extraction and detection of DNA is becoming more and more effective for personal identification.[26] The detection techniques of both fingerprints and DNA are two powerful tools for personal identification. However, the above two techniques seem to be incompatible. The latent fingerprint development cannot be accomplished if the DNA extraction is conducted beforehand, and the DNA extraction is almost impossible to achieve when latent fingerprint development is attempted before the extraction is performed. Therefore, it is reasonable, albeit difficult, to propose an improved or innovative method for latent fingerprint development without DNA damage.

In the powder dusting method, the most flagrant drawback is dust blowing, because most of the chemical substances used in this method are toxic. In the cyanoacrylate fuming method, the liquid cyanoacrylate esters and their vapors can cause acute damage to the skin, eyes, and mucous membranes. In chemical methods, the developing reagents, including silver nitrate, ninhydrin, and DFO, are all toxic. The use of silver nitrate can also cause DNA damage.[27] In the small particle reagent method, the toxicity is relatively lower than the powder dusting method due to the avoidance of dust blowing. However, for all liquid-based methods, including chemical methods and the small particle reagent method, DNA in fingerprint residuals can be easily damaged or rinsed off. In addition, the use of UV illumination in fingerprint develpment can not only do harm to human eyes and skin but also reduce the possible extraction yield of DNA.[28]

4. Advantages of Using Fluorescent Nanomaterials for Developing Latent Fingerprints

Fluorescent NMs can emit strong fluorescence by excitation of specific lights. One of the most successful applications of fluorescent NMs is biolabelling and bioimaging.[29,30]

Inspired by this, scientists have proposed the use of florescent NMs as a way to develop latent fingerprints.[31–34] Recently, the use of fluorescent NMs such as QDs and UCNMs for the development of fingerprints has garnered a tremendous amount of attention due to their unique optical properties. There are several superior advantages when fluorescent NMs are used for latent fingerprint development, such as high developing contrast, high developing sensitivity, high developing selectivity, and low toxicity.[31,32]

4.1. High Contrast

QDs and UCNMs as the best-studied examples of fluorescent NMs; they can emit visible fluorescence with high intensity under UV and near-infrared (NIR) light irradiation, respectively.

Such strong fluorescent emissions can dramatically promote the developing signal and reduce the background color distraction, leading to a high developing contrast.[31,32] More importantly, featured with low energy radiation, NIR lights would not trigger the emission of background fluorescence from substrates, thus circumventing the risk of background color distraction, and finally warranting a high developing contrast.[32] Therefore, the use of fluorescent NMs including QDs and UCNMs for latent fingerprint development can obtain a high developing contrast through an increased developing signal and a decreased background color distraction, due to their strong fluorescent properties.

4.2. High Sensitivity

The size of fluorescent NMs such as QDs and UCNMs is typically small. Generally, the diameter of UCNMs is no more than 100 nm, and the size of QDs is even smaller, not exceeding 10 nm in diameter. Using these small-sized fluorescent NMs for latent fingerprint development, the ridge details such as arches, termination points, and sweat pores, will not easily be heavily covered, giving rise to high developing sensitivity.[31] In addition, these fluorescent NMs are spherical in shape, and their shape can be tuned during the synthesis. The tackiness of these fluorescent NMs can also be adjusted by surface modification.[35] Thus, the adsorption of the fluorescent NMs onto the fingerprint residues can be adjusted, and the developing sensitivity can be further improved. Therefore, the use of fluorescent NMs including QDs and UCNMs for latent fingerprint development can obtain a high developing sensitivity due to their small size, suitable shape, and tunable tackiness.

4.3. High Selectivity

The surface modification of fluorescent NMs such as QDs and UCNMs is a flexible and effective approach to reaching a high selectivity of fingerprint development. The electric charge of these fluorescent NMs can be adjusted by surface modification, allowing the NMs to bind to some specific residues in fingerprints by electrostatic adsorption for high developing selectivity.[36] In addition, the surfaces of these fluorescent NMs can be modified with a variety of functional groups (e.g., carboxyl, amino, and aldehyde). The resultant fluorescent NMs can selectively label a specific component in fingerprints by chemical reactions to reach an increased developing selectivity.[37] The fluorescent NMs can also be modified with some special molecules such as lysozyme-binding aptamer, which could conjugate with the lysozymes in fingerprint residues providing high selectivity.[38]

4.4. Low Toxicity

Some elements in QDs such as Cd can raise the toxicity concern.[39] Besides, some QDs could also cause inflammation and allergy.[40–43] It was reported that the toxicity of QDs could be reduced to a great extent by surface modification with a layer of silica dioxide. Despite this, it is generally recommended to wear personal protective equipment such as protective eyewear, N95 masks, and nitrile gloves while operating. It has been proven that surface-modified UCNMs exhibit a low toxicity or even become nontoxic in biolabelling and bioanalysis.[44] More importantly, the use of NIR light for excitation of UCNMs shows that it is less harmful to DNA in fingerprint residues, which is beneficial for the subsequent DNA analysis.[45] Therefore, the use of fluorescent NMs including QDs and UCNMs for latent fingerprint development can obtain a low toxicity or even nontoxicity.

5. Latent Fingerprint Development Using Quantum Dots

QDs can emit strong visible light under the excitation of blue or UV light (Figure 4).[46,47] They have optical advantages such as broad adsorption spectra, size- and composition-tunable emission, narrow emission spectra, high quantum yield, good photo-stability, and strong intensity.[48,49] In addition, QDs can be flexibly chemically modified with a variety of functional ligands.[50,51] Therefore, QDs are raising new possibilities for latent fingerprint development with a high level of contrast, sensitivity, and selectivity. So far, CdS, CdSe, and CdTe QDs have been used for developing latent fingerprints.[52]

5.1. Quantum Dots Used in Powder Dusting Method

Generally, when QDs are used in the powder dusting method, they are allowed to adhere to the aqueous or oily components that can be found in the latent fingerprint residues, largely resembling the adherence manner of the traditional powder dusting method. The adsorption interaction between QDs and latent fingerprint residues mainly depends on physical adsorption and electrostatic attraction. Due to the high oxidation behavior of QDs exposed in air after adhering to the fingerprints, the fluorescence emitted from QDs is liable to decrease, leading to a low developing contrast. This indicates that the long-time preservation of fingerprint specimens is another problem. To overcome this oxidation problem of QDs powders in air, QDs were often conjugated or covered with a specific material to form the QDs based nanocomposites (NCs).

In 2009, Dilag et al. reported the use of CdS/chitosan NCs (CdS QDs encapsulated in polymeric chitosan matrix) for developing the latent fingerprints deposited on aluminum substrates.[53] They converted the synthesized NCs into dry powders using a freeze drier. The latent fingerprints were fumed first with cyanoacrylate ester. Subsequently, the as-prepared NCs were applied to latent fingerprints by using a squirrel hair brush. When the unfumed latent fingerprints on aluminum were directly developed by using the dry powders of CdS/chitosan NCs, good results could be observed (Figure 5). Compared with the latent fingerprints developed by conventional micron-sized powders, including white powders (containing titanium dioxide) and aluminum powders, the latent fingerprints developed by CdS/chitosan NCs exhibited an increased developing contrast but a decreased developing sensitivity and selectivity. They explained that the freeze-drying process could certainly

cause the CdS/chitosan NCs to become aggregated, which can deteriorate the final quality of latent fingerprint development.

In 2011, Algarra et al. similarly reported the use of CdS/PPH NCs (CdS QDs bonded to porous phosphate heterostructure materials (PPH)) for developing the latent fingerprints deposited on various types of substrates.[54] The hybrid PPH materials were chemically functionalized with mercaptopropyl groups, and then used them to synthesize the CdS/PPH NCs. Subsequently, the as-prepared NCs were directly used for developing the latent fingerprints on plastic, glass, steel, ceramic, and wood surfaces. As shown in Figure 6, the latent fingerprints deposited on steel (Figure 6a) and glass (Figure 6b) were developed with a good contrast and could be further observed under the excitation of 450 nm light and imaged with orange goggles (edge wavelength of 550 nm). However, the developing sensitivity and selectivity should be further improved. The attempt to develop latent fingerprints on paper seems unsuccessful due to the background fluorescent interference of paper.

In 2011, Gao et al. used CdTe/MMT NCs (CdTe QDs inserted into montmorillonite (MMT)) for developing the latent fingerprints deposited on various types of substrates.[35] As shown in Figure 7, the CdTe/MMT NCs-developed fingerprints under the excitation of 365 nm light were well-defined in terms of papillary ridge details, resulting in high developing sensitivity. In addition, the fluorescence could be observed only at the papillary ridges rather than on the furrows or the background, generating high developing selectivity. However, the developing contrast for painted wood (Figure 7b) and leather (Figure 7d) was much lower than that for polymer (Figure 7a) and glass (Figure 7c), due to the strong background fluorescent interference from paper and the weak fluorescence for leather, respectively.

In 2012, Gao et al. further used CdTe@SiO_2 NMs (CdTe QDs coated with a layer of silicon dioxide) for developing the latent fingerprints deposited on different substrates.[55] In the synthesis, bare CdTe QDs were modified with SiO_2 coating to form CdTe@SiO_2 NMs via a typical Stober-based method. Compared with bare CdTe QDs, the resultant NMs with a core-shell structure could possess a better adhesive ability, higher chemical stability, stronger fluorescent intensity, and lower toxicity, due to the stable, transparent, and non-toxic SiO_2 shell. Subsequently, the as-prepared CdTe@SiO_2 NMs were directly applied to develop the latent fingerprints on glass, polymer, plastic, rubber, marble, aluminum, and paper. Figure 8 shows that the latent fingerprints developed by CdTe@SiO_2 NMs exhibited a high sensitivity and selectivity. However, the developing contrast for black rubber (Figure 8e) and paper (Figure 8f) was slightly lower than that for others, due to the unavoidable background fluorescent interference from the substrates under the excitation of 365 nm UV light.

5.2. Quantum Dots Used in Liquid Method

Using QDs in the liquid method for latent fingerprint development is made possible by allowing the QDs to selectively adhere to some certain components in fingerprint residues, which occurs in a liquid environment. The selective absorption interaction between QDs and latent fingerprint residues is determined by chemical reactions. However, some physical processes such as physical adsorption and electrostatic attraction may occur at the same

time. To accomplish the absorption interaction, QDs were often modified by various specific ligands with functional groups.

In 2000, Menzel et al. first reported the use of CdS/DSS NCs (CdS QDs modified with two-branched molecules called dioctyl sodium sulfosuccinate (DSS)) dissolved in heptane or hexane for developing the latent fingerprints on metallic and sticky substrates.[56] Before treatment using CdS/DSS NCs solution, the latent fingerprints were pre-fumed with cyanoacrylate ester. The fumed fingerprints were then immersed in the CdS/DSS NCs organic (heptane or hexane) solution for times ranging from a few seconds to a few minutes. The resultant samples had to be gently rinsed with hexane to remove the excess CdS/DSS NCs, and thus the heavily covered details of the fingerprints were made visible. The samples were left to dry and then observed under UV light. As shown in Figure 9, the latent fingerprints deposited on aluminum foil (Figure 9a) and soft drink can (Figure 9b) could be developed with a high contrast and selectively, using the cyanoacrylate ester fuming followed by CdS/DSS NCs staining technique. Unfortunately, attempts to develop the unfumed bare fingerprints deposited on metal, glass, and plastics using such a procedure turned out unsuccessful because the oily fingerprint residues could be obliterated by the nonpolar organic solvents. However, bare fingerprints were deposited on the sticky side of black electrical tape and could then be developed by a similar procedure, where fingerprints were stained by CdS/DSS NCs solution without prefuming.

In 2008, Jin et al. reported the use of CdS/PAMAM NCs (CdS QDs modified with polyamidoamine dendrimers (PAMAM)) for developing latent fingerprints deposited on aluminum foil.[57] Before treatment using CdS/PAMAM solution, the latent fingerprints were prefumed with cyanoacrylate ester; the fumed fingerprints were then immersed into the CdS/PAMAM solution overnight at room temperature. As shown in Figure 10, the latent fingerprints deposited on aluminum foil could be developed by CdS/PAMAM NCs dissolved in both methanol (Figure 10a) and methanol-water mixtures (Figure 10b), with a high developing contrast, sensitivity, and selectivity, mainly due to the strong fluorescence emitted from CdS/PAMAM NCs and the selective binding between the NCs and the fumed fingerprints.

In 2009, Wang et al. reported the use of thioglycolic acid (TGA)-modified CdSe and CdSe@CdS QDs NPs dissolved in aqueous solution for developing latent fingerprints.[58] The developing procedure was simple, efficient, and effective. It involved the direct immersion of the unfumed fingerprints into CdSe QDs aqueous solution for about 15 min. The resultant fingerprints were left to dry and then observed under 380 nm UV light. It was found that the pH value of this CdSe QDs solution exhibited an obvious impact on the result of fingerprint development. Fingerprint development carried out under weakly basic conditions (e.g., pH 8) could obtain more ridge details. As shown in Figure 11, they found that the latent fingerprints developed by CdSe@CdS QDs aqueous solution (Figure 11b) showed a better result than that using bare CdSe QDs (Figure 11a). They also discovered that an improved fingerprint image can be captured under 380 nm UV light (Figure 11b), exhibiting high developing contrast, sensitivity, and selectivity.

In 2010, Liu et al. employed CdTe QDs NPs to develop the latent fingerprints.[37] The CdTe QDs could be selectively adsorbed by the ridges of the fingerprint. The latent fingerprints were then developed when CdTe QDs were excited (Figure 12) with a sufficient contrast, sensitivity, and selectivity. However, some ridge details were still covered by CdTe QDs, probably because excess CdTe QDs were not rinsed thoroughly.

In 2011, Gao et al. reported the use of both negatively and positively charged CdTe QDs dissolved in aqueous solution for developing the latent fingerprints deposited on various smooth substrates.[36] During the synthesis, the negatively charged CdTe QDs were prepared by surface modification with TGA in an aqueous solution at room temperature, while the positively charged CdTe QDs were prepared by surface modification with hydrazine hydrate, based on the TGA-modified CdTe QDs. The developing procedure was relatively simple, including the following consecutive steps: (1) applying small aliquots (1 ml) of CdTe QDs aqueous solution (pH 7–11) on the latent fingerprints for 30 min to 1 h; (2) removing the excess CdTe QDs by rinsing with distilled water; and (3) observing the fingerprints under 365 nm UV light. It was found that the use of the positively charged CdTe QDs for latent fingerprint development possessed an enhanced affinity and effectivity, compared with the use of negatively charged CdTe QDs. They used the positively charged CdTe QDs to develop the latent fingerprints deposited on a verity of substrates. As shown in Figure 13, the latent fingerprints developed by positively charged CdTe QDs aqueous solution and then excited under 365 nm UV light could exhibit a sufficient developing contrast, sensitivity, and selectivity, except for the applications on rough plastic sheet (Figure 13e), due to the rough surfaces. In addition, some ridge details were still covered by CdTe QDs, probably because excess CdTe QDs were not rinsed thoroughly.

In 2014, Wang et al. employed CdSe QDs NPs dissolved in aqueous solution for developing latent fingerprints.[59] The developing procedure included the direct immersion of the fingerprints in CdTe QDs aqueous solution, followed by rinsing excess CdSe QDs with pure water and observation under 365 nm UV light. The optimization of developing parameters including the developing time, the pH of aqueous solution, and excitation light source was also studied in detail. To obtain a good, uniform development, the samples should be immersed in the CdTe QDs aqueous solution for more than 15 min, and as for one-day-old fingerprints, the immersion time should be prolonged to more than 30 min. Wang et al. also found that the optimized pH value for latent fingerprint development was 8, a weakly basic condition. As shown in Figure 14, the latent fingerprints developed by CdSe QDs aqueous solution and then excited under either 365 nm UV light (Figure 14b) or 440 nm blue light (Figure 14c) could exhibit a sufficient developing contrast, sensitivity, and selectivity. However, the whole fingerprints were still not uniformly developed with some regions exhibiting over-bright or over-dark fluorescence (Figure 14d). In addition, some tiny filiform obits were found on some ridge details, which could emit blue background fluorescence under UV light and thus caused obvious background interference (Figure 14d).

5.3. Quantum Dots Used for Developing Blood Fingerprints

Generally, the use of QDs for blood fingerprint development involves the adherence of QDs to some certain components in blood fingerprint residues such as hemoglobin.[60] To

increase the interaction between QDs and hemoglobin in blood fingerprint residues, QDs were often modified by various specific ligands with functional groups.

In 2009, Becue et al. utilized TGA-modified CdTe QDs NPs to develop the blood fingerprints deposited on various nonporous substrates, including transparent polypropylene, black polypropylene, glass, and aluminum foil.[61] The method first involved immersion of the blood fingerprints in an aqueous solution of 5-sulfosalicylic acid for 10 min, then they briefly rinsed the fingerprints with water. Finally, the fingerprints were immersed in the CdTe QDs aqueous solution (pH 3.5) for 20 min and the excess of CdTe QDs were removed by rinsing with distilled water for 2–3 min. As shown in Figure 15, the blood fingerprints developed by CdTe QDs aqueous solution and then excited under 300–400 nm UV light could exhibit a sufficient developing contrast and selectivity. The ridges were clearly defined, presenting a high developing sensitivity. In addition, the blood fingerprints developed by CdTe QDs were equally recognizable in the presence of acid yellow 7 (AY7) on glass (Figure 15a, a′) and polyethylene materials (Figure 15b, b′, c, c′), where an inferior image under AY7 on aluminum was developed (Figure 15d, d′). It is apparent in some regions, however, that fluorescence was either too bright or too dark.

In 2013, Moret et al. similarly reported the use of 3-mercaptopropionic acid (MPA)-modified ZnS:Cu (copper doped zinc sulphide) QDs NPs dissolved in aqueous solution for developing the blood fingerprints deposited on various nonporous substrates, including transparent polypropylene, black polypropylene, glass, and aluminum foil.[62] The developing mechanisms and procedures were quite similar to that reported by Becue et al. in 2009.[61] In addition, both AY 7- and TGA-modified CdTe QDs (as reported by Becue et al.[57] in 2009) were used to develop the blood fingerprints for comparison. As shown in Figure 16, the blood fingerprints, which were developed by ZnS:Cu QDs (Figure 16a–h), AY 7 (Figure 16a′–d′) and CdTe QDs (Figure 16e′–h′), respectively, and then excited under 300–400 nm UV light, presented a sufficiently high contrast, sensitivity, and selectivity. It could be concluded that the development of blood fingerprints using ZnS:Cu QDs was comparable to that using AY 7 and CdTe QDs. However, the fingerprints developed by ZnS:Cu QDs on the transparent polypropylene materials appeared less visible, due to the background fluorescent interference (Figure 16b, f′).

6. Latent Fingerprint Development Using Rare Earth Upconversion Fluorescent Nanomaterials

Rare earth upconversion fluorescent nanomaterials (UCNMs) are rare earth-doped materials. When excited by a longer wavelength light, they emit a light of a shorter wavelength.[63,64] For example, they emit visible light when excited by NIR light (Figure 17).[65] UCNMs have many characteristic advantages such as narrow emission spectra, low toxicity, and strong intensity.[66] In addition, they can be functionalized chemically.[67] Because they can be excited by using NIR light (e.g., 980 nm), the background fluorescent interferences are avoided,[68,69] leading to an increased developing contrast. Therefore, UCNMs hold promise for latent fingerprint development with very high contrast, sensitivity, and selectivity.

Currently, $NaYF_4$ co-doped with Yb^{3+}–Er^{3+} ions ($NaYF_4$:Yb,Er) is the most commonly used UC material, which can emit the brightest UC fluorescence.[70]

6.1. Rare Earth Upconversion Fluorescent Nanomaterials Used in Powder Dusting Method

When UCNMs are used in the powder dusting method, they adhere to the latent fingerprint residues to enable the development. In this sense, the operation is quite similar to the traditional powder dusting method.

In 2011, Ma et al. first reported the use of commercially available $NaYF_4$:Yb,Er UC microparticles for developing the latent fingerprints on various nonporous and semiporous substrates.[71] The developing process only involved the application of $NaYF_4$:Yb,Er powders to the latent fingerprints using a squirrel brush. In their work, the attempts to develop the latent fingerprints on various common substrates including glossy magazine papers, beer cans, and plastic labels proved to be successful. In addition, good results were obtained on some special substrates such as the Australian five-dollar polymer banknote, which could emit strong fluorescence under UV radiation to cause a serious background fluorescent interference (Figure 18a). In 2012, Ma et al. similarly reported the use of YVO_4:Yb,Er UC particles for developing latent fingerprints on various nonporous and semiporous substrates.[72] The developing procedure was quite similar to their previously reported methods. In addition, similar results were obtained when developing the latent fingerprints on various substrates (Figure 18b). By the excitation of 980 nm NIR light, the $NaYF_4$:Yb,Er (Figure 18a) and YVO_4:Yb,Er (Figure 18b) UC powder used to develop latent fingerprints on Australian five-dollar polymer banknote emitted strong visible fluorescence, and the ridge details were remarkably clear almost without any background fluorescent interferences. Although the $NaYF_4$:Yb,Er and YVO_4:Yb,Er particles applied in the above two studies were micrometers in size rather than nanometers in size, it opened up a new direction on the use of nano-sized UC particles for latent fingerprint development.

In 2015, Wang et al. employed nano-sized $NaYF_4$:Yb,Er particles to develop latent fingerprints on a variety of substrates.[73] In the synthesis, oleic acid-modified $NaYF_4$:Yb,Er UCNMs were prepared via a solvothermal approach. Subsequently, the as-prepared $NaYF_4$:Yb,Er UCNM dry powders were carefully applied to the latent fingerprints with a light brushing action, and the developed fingerprints were observed under 980 nm NIR light radiation. The $NaYF_4$:Yb,Er UCNMs-developed fingerprint on glass (Figure 19a) presented even the detailed features such as sweat pores. Such a high developing sensitivity is impossible to achieve by using the traditional powders-even the fluorescent powders. In addition, the green fluorescence could be observed only at the papillary ridges rather than the furrows or the background (Figure 19a), resulting in a high developing selectivity. It can be seen from Figure 19b–i that good results were also obtained on various substrates, and well-defined papillary ridges could be clearly defined without background interference, exhibiting a high developing contrast. It can be concluded that $NaYF_4$:Yb,Er UCNMs are a versatile fluorescent label for the facile development of fingerprints on virtually any non-infiltrating substrates with very high developing contrast, sensitivity, and selectivity.

In 2015, Wang et al. also used $NaYF_4$:Yb,Er UCNMs for developing the latent fingerprints on three types of substrates, including substrates with a single background color (transparent

glass, white ceramic tiles, and black marbles), substrates with multiple background colors (marbles with different complex patterns), and substrates with strong background autofluorescence (note papers, Chinese paper money, and plastic plates).[74] As shown in Figure 20, the latent fingerprint development procedure using $NaYF_4$:Yb,Er UCNMs on various substrates exhibited very high developing contrast, sensitivity, and selectivity. It should be noted here that the fingerprint development on Chinese paper money, note papers, and plastic plates that could emit strong autofluorescence under UV lights showed a very high developing contrast with no background interference.

6.2. Rare Earth Upconversion Fluorescent Nanomaterials Used in Liquid Method

UCNMs suspensions are used for latent fingerprint development through their selective adherence to some certain components in latent fingerprint residues in a liquid environment.

In 2014, Wang et al. first employed lysozyme-binding aptamer (LBA)-modified $NaYF_4$:Yb,Er UCNMs ($NaYF_4$:Yb,Er/LBA) for developing latent fingerprints based on the molecular recognition technique.[38] A lysozyme, one of the polypeptides found in human perspiration, could serve as a universal target in fingerprint residue; LBA, a DNA aptamer-targeting lysozyme, could specifically recognize the lysozyme. In the developing procedure, a suspension of $NaYF_4$:Yb,Er/LBA UCNMs was first applied to the latent fingerprints, and then the samples were incubated for 30 min. During the incubation, the LBA molecules on the surface of the UCNMs were selectively conjugated with the lysozyme molecules in the fingerprint residuals, triggering the selective recognition and adsorption between $NaYF_4$:Yb,Er UCNMs and latent fingerprints. For comparison, LBA-modified fluorescein amidite (FAM/LBA) and LBA-modified CdTe QDs (CdTe/LBA) were used in a similar manner and the corresponding results are presented in Figure 21. The marbles treated with FAM (Figure 21b1) or CdTe QDs (Figure 21c1) showed no fingerprints but only strong purple background fluorescence under excitation with 365 nm UV light. A similar phenomenon was obtained when using the suspension of $NaYF_4$:Yb,Er UCNMs; however, there was not background fluorescence at all under excitation with 980 nm NIR lights (Figure 21d1). When FAM/LBA (Figure 21b2) or CdTe/LBA (Figure 21c2) was used for incubation, the fingerprints were developed with a low developing contrast, due to the strong background fluorescent interferences. When $NaYF_4$:Yb,Er/LBA UCNMs were used for incubation, a clear fluorescent image without any background fluorescent interference was obtained (Figure 21d2); it exhibited a high developing selectivity and conrast. In addition, the arches and termination points of the fingerprints had easily discernable details when observed in the magnified image (Figure 21d3), showing high developing sensitivity. This strategy could further serve as a robust approach to latent fingerprint development; however, a relatively long time of 30 min for incubation was required.

In 2016, Wang et al. further used $NaYF_4$:Yb,Er UCNMs-based suspension for developing latent fingerprints on many substrates.[32] In the suspension, $NaYF_4$:Yb,Er UCNMs and a surfactant of sodium dodecyl sulfonate (SDS) were mixed together and dispersed with water. The whole developing process was relatively simple; the fingerprint was first immersed into the suspension for several minutes, and then gently rinsed with water. The hydrophobic chain of the SDS on the UCNMs allowed them to stick to the grease in the fingerprint

residues. The hydrophobic interactions among the alkyl chains allows for the absorption, thus staining fingerprints with high developing selectivity. Then, an excitation of the UCNMs by 980 nm NIR light enabled the detection of the fingerprints (Figure 22). In particular, old fingerprints (Figure 22i), wet fingerprints, and fingerprints on multi-colored and auto-fluorescent background (Figure 22h) could also be clearly observed through this method.

7. Conclusion and Outlook

This review places special focus on the application and performance of fluorescent NMs, including QDs and UCNMs on latent fingerprint development. We included a brief overview of the most widely used traditional developing methods, including powder dusting, cyanoacrylate fuming, silver nitrate method, ninhydrin method, DFO method, and small particle reagent method. Then, a summary is presented on the drawbacks of these traditionally used methods, such as low developing contrast, low developing sensitivity, low developing selectivity, and high toxicity. To overcome these problems, fluorescent NMs, especially QDs and UCNMs, which possess excellent optical properties, have emerged as a new class of fluorescent probes for latent fingerprint development. A discussion was then presented on the advantages of fluorescent NMs for latent fingerprint development. Finally, we summarized the recent advances of latent fingerprint development using QDs and UCNMs in detail.

As discussed above, the uses of fluorescent NMs including QDs and UCNMs for latent fingerprint development exhibited four main advantages: (i) high developing contrast due to enhanced developing signal and reduced background noise; (ii) high developing sensitivity due to the small size, suitable shape and tunable stickiness; (iii) high developing selectivity due to various surface modifications of fluorescent NMs; and (iv) low toxicity to the operator and DNA in fingerprint residuals, particularly due to the use of NIR light in the case of UCNMs. Therefore, the fluorescent NMs, especially QDs and UCNMs, could serve as efficient probes for latent fingerprint development, which has been proven to be a competitive supplement for traditional developing techniques.

However, there are still some higher requirements of fluorescent NMs including QDs and UCNMs in latent fingerprint development. First, the reported works are still being developed and have not yet transitioned to real-world applications, which means that the possibility of a high developing contrast, sensitivity, selectivity, and low toxicity has yet to be transformed into any simplified and efficient practice. Second, most of the reported works only deal with the development of normal fingerprints such as sweat fingerprints and sebum fingerprints, but hardly touch on special fingerprints such as blood fingerprints and aged fingerprints. Third, most of the current studies only focus on the development of latent fingerprints on normal substrates such as smooth, nonporous, and nonfluorescent surfaces, while special surfaces such as porous, wet, and fluorescent surfaces are barely touched upon. Fourth, the techniques need to be developed for selectively targeting fingerprints and simultaneously detecting fingerprint residue by using immunogenic methods.[75] Finally, there is a necessity for further development of DNA extraction following latent print development in order for the research to move forward into field testing.

Nowadays, nanotechnology is gradually becoming a powerful tool, and applied in forensic sciences, has great potential to expand and change the concept of trace examination. Most research and development of nanotechnology in forensic sciences have been focused on the fluorescent nanomaterials-based development of latent fingerprints. Since little is known about utilizing nanotechnology on other types of trace evidence, at the end of this review, we put forward some challenges in the forensic applications of nanotechnology: (i) analysis of explosive residues, (ii) analysis of gunshot residues, (iii) analysis of drugs and poisons, (iv) analysis of trace material evidences, (v) extraction and analysis of DNA, and (vi) analysis of human secretions. We firmly believe that nanotechnology will change the traditional concept of trace examination and open up a new field in forensic sciences.

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Biographies

Meng Wang received his BS degree in Applied Chemistry from Northeastern University in 2005 and his PhD in Analytical Chemistry from Northeastern University in 2010, under the direction of Prof. Shukun Xu. He joined the faculty of Department of Trace Examination, National Police University of China in 2011, and became an associate professor in 2013. His current research is focused on the synthesis, characterization, and application of rare earth fluorescent nanomaterials.

Mingying Yang received her PhD in Biotechnology in 2005 from Tokyo University of Agriculture and Technology. She then conducted postdoctoral studies in biomaterials at the

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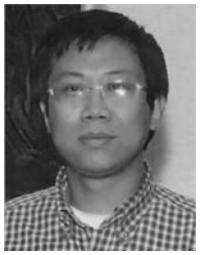

Chuanbin Mao received his PhD in 1997 from Northeastern University in China. He completed postdoctoral studies at Tsinghua University, followed by a faculty position in the same university in 1999. After a short stay at the University of Tennessee at Knoxville, he moved to the University of Texas at Austin in 2000 and continued his postdoctoral studies until he took a faculty position at the University of Oklahoma in 2005. His research is now focused on nanobiomaterials, nanobiotechnology, and nanomedicine.

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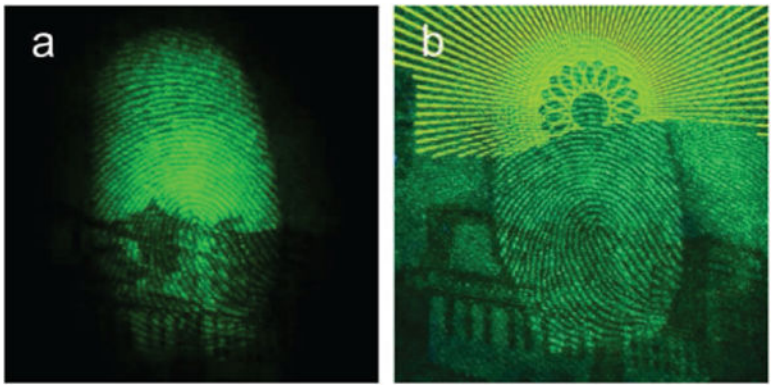


Figure 1.

Latent fingerprint development on Chinese paper money through the fluorescent property of different developing powders: a) $NaYF_4$:Yb,Er UCNMs, in dark field and under 980 nm NIR excitation; and (b) green fluorescent powders, in dark field and under 254 nm UV excitation. Reproduced with permission.[74] Copyright 2015, American Chemical Society.

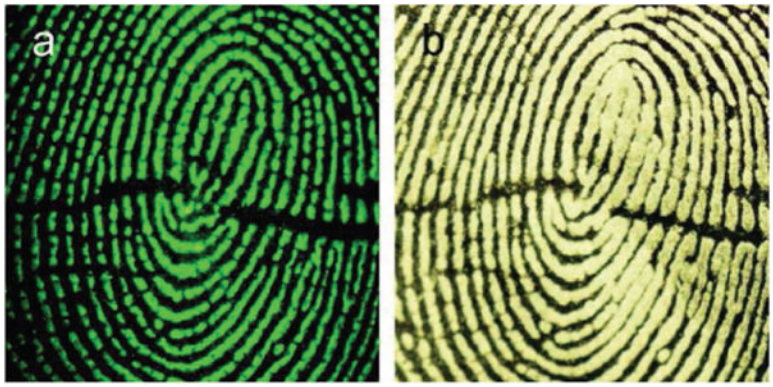


Figure 2.

Latent fingerprint development on glass by using different types of developing powders: a) $NaYF_4$:Yb, Er UCNMs, in dark field and under 980 nm NIR excitation; and (b) bronze flake, in bright field. Reproduced with permission.[73] Copyright 2014, Springer.

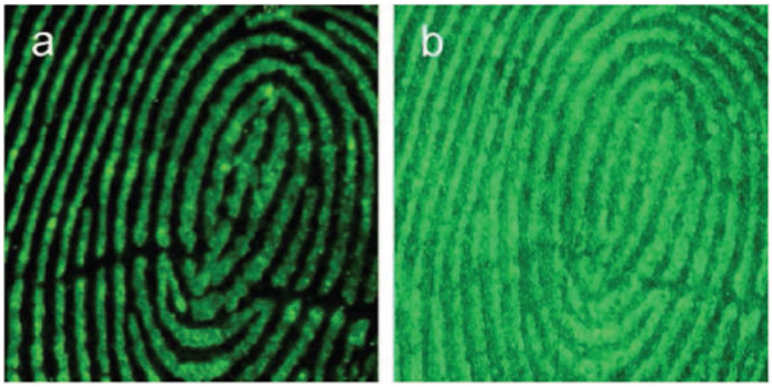


Figure 3.

Latent fingerprint development on glass by using different types of developing powders: a) $NaYF_4$:Yb, Er UCNMs, in dark field and under 980 nm NIR excitation; and (b) green fluorescent powders, in dark field and under 254 nm UV excitation.

Figure 4.

Aqueous suspensions of CdSe@ZnS QDs with ten distinguishable fluorescent emission colors excited with a 350 nm UV radiation. From left to right, the maximum emissions are located at 443, 473, 481, 500, 518, 543, 565, 587, 610, and 655 nm. Reproduced with permission.[47] Copyright 2001, Nature Publishing Group.

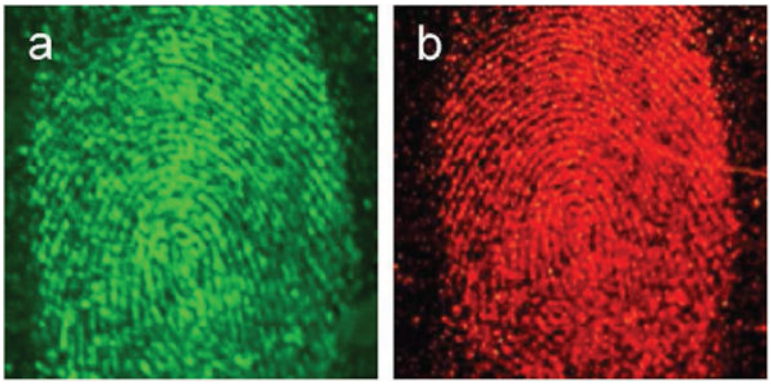


Figure 5.

Latent fingerprint development on aluminum by using CdS/chitosan NCs under the 450 nm light excitation and imaged with a) 550 nm long pass barrier filter and b) 565 nm band pass barrier filter. Reproduced with permission.[53] Copyright 2009, Elsevier.

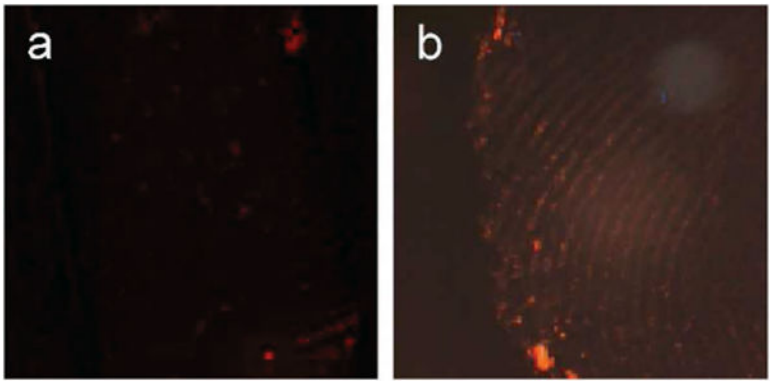


Figure 6.

Latent fingerprint development by using CdS/PPH NCs under the 450 nm light excitation on the different substrates: a) steel tweezers, and b) glass. Reproduced with permission.[54] Copyright 2011, Elsevier.

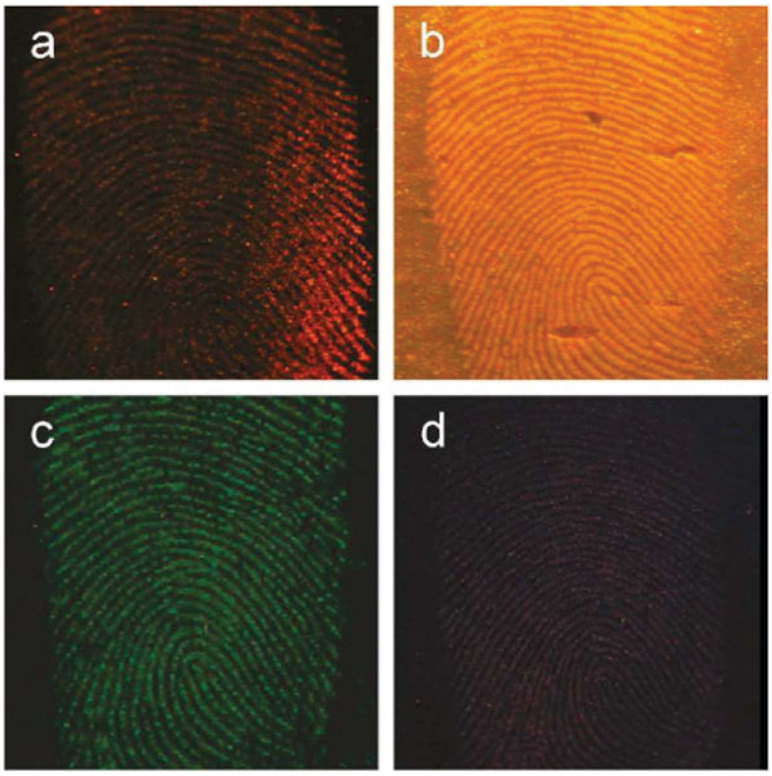


Figure 7.

Latent fingerprint development by using CdTe/MMT NCs under the 365 nm UV excitation on a variety of substrates: a) polymer, b) painted wood, c) glass, and (d) leather. Reproduced with permission.[35] Copyright 2011, American Chemical Society.

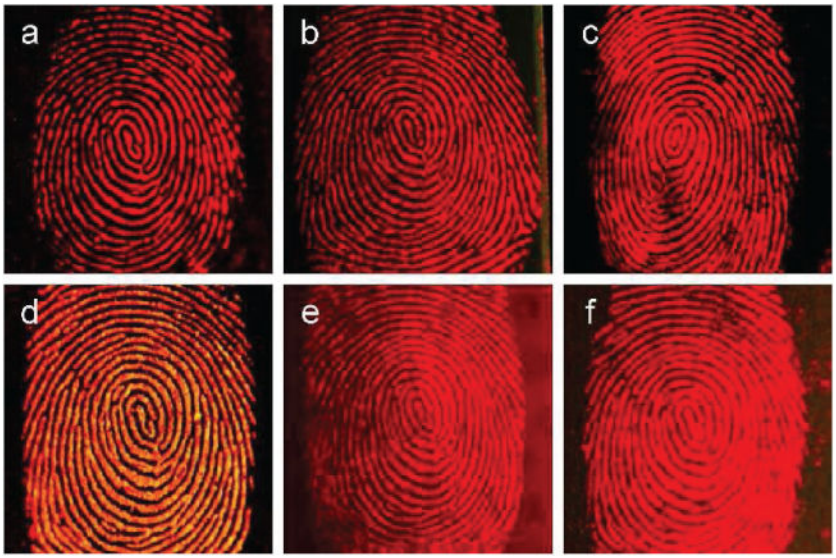


Figure 8.

Latent fingerprint development by using CdTe@SiO_2 NMs under the 365 nm UV excitation on a variety of substrates: a) glass, b) polymer materials, c) aluminum foil, d) black ceramic, e) black rubber, and f) paper. Reproduced with permission.[55] Copyright 2012, Springer.

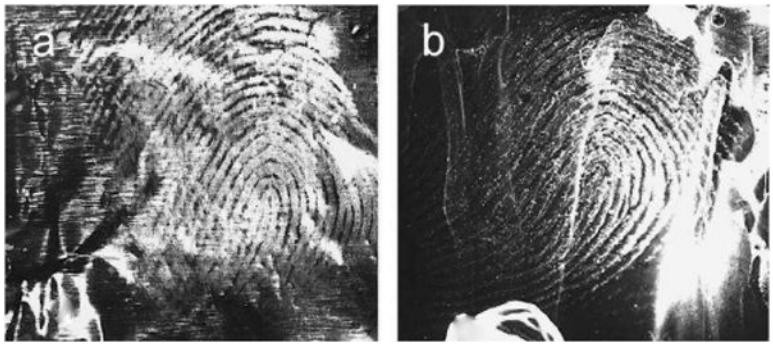


Figure 9.

Latent fingerprint development by using CdS/DSS NCs under the UV excitation on different substrates: a) aluminum foil, and b) soft drink can. Reproduced with permission.[56] Copyright 2000, ASTM International.

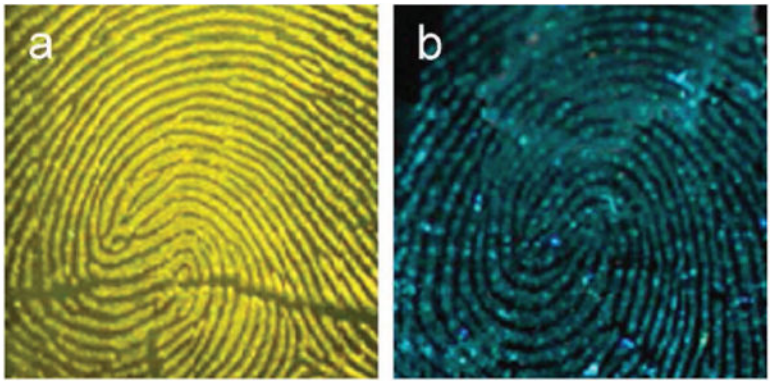


Figure 10.

Latent fingerprint development on the aluminum foils by using CdS/PAMAM NCs dissolved in a) methanol, and b) 1:9 methanol-water solutions, under the 365 nm UV light excitation with the assistance of a yellow light filter and a blue light filter, respectively. Reproduced with permission.[57] Copyright 2008, Elsevier.

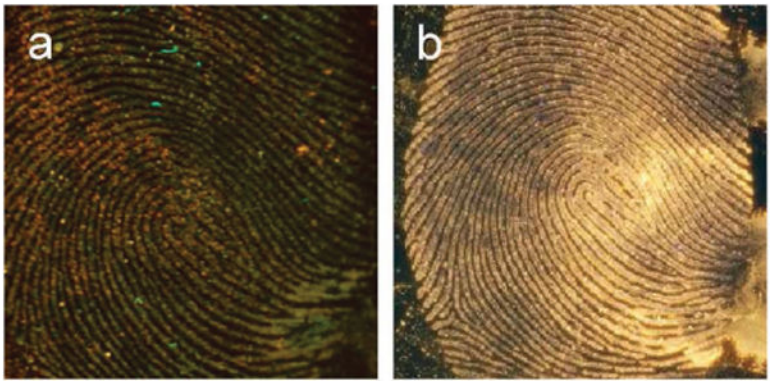


Figure 11.

Latent fingerprint development on the sticky side of tapes by using a) CdSe and b) CdSe@CdS QDs under the 380 nm UV excitation. Reproduced with permission.[58] Copyright 2009, Elsevier.

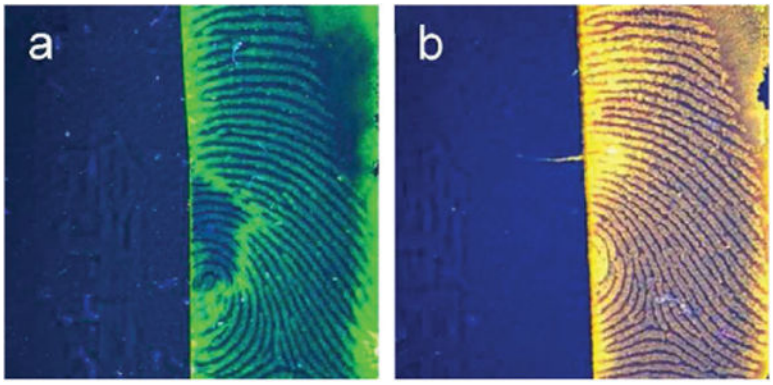


Figure 12.

Latent fingerprint development on the sticky side of adhesives by using CdTe QDs with multi-colors under the 365 nm UV excitation: a) CdTe QDs with green fluorescence, synthesized without refluxing, b) CdTe QDs with yellow fluorescence, refluxed for 2 h. Reproduced with permission.[37] Copyright 2010, Elsevier.

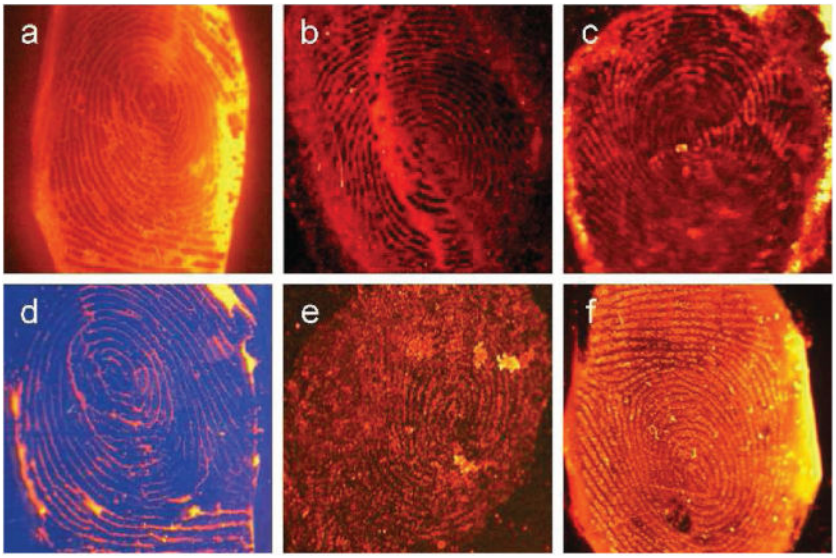


Figure 13.

Latent fingerprint development by using positively charged CdTe QDs under the 365 nm UV excitation on a variety of substrates: a) glass, b) black ceramic, c) painted polymer material, d) transparent plastic sheet, e) rough plastic sheet, and f) black rubber. Reproduced with permission.[36] Copyright 2011, IOP Publishing.

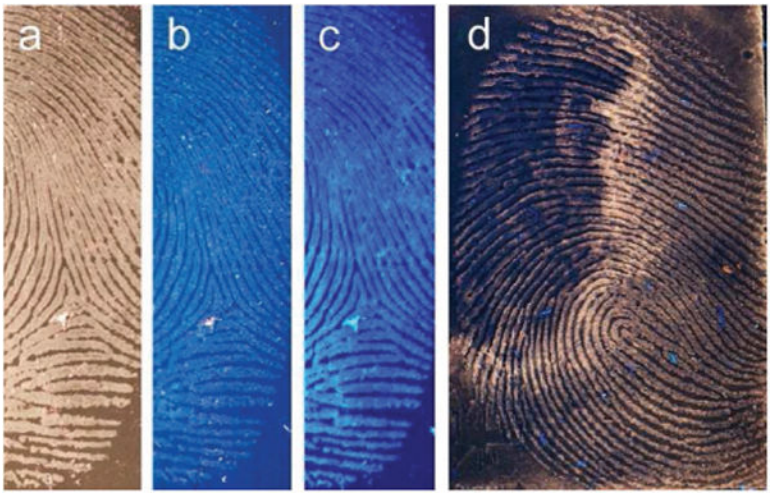


Figure 14.

Latent fingerprint development on the sticky side of black electrical tape by using CdSe QDs: a) in the bright field, b, d) in the dark field with 365 nm UV excitation, and c) in the dark field with 440 nm blue light excitation. Reproduced with permission.[59] Copyright 2014, Elsevier.

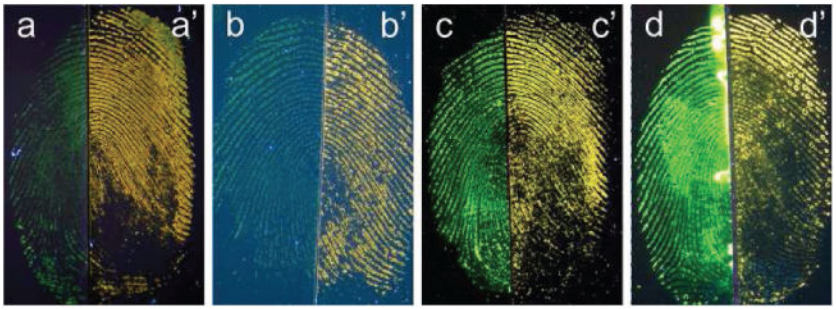


Figure 15.

Blood fingerprint development by using CdTe QDs (left halves, a–d) and AY7 (right halves, a′–d′) under the 300–400 nm UV excitation on a variety of substrates: (a, a′) glass, (b, b′) transparent polypropylene, (c, c′) black polyethylene, and (d, d′) aluminum. Reproduced with permission.[61] Copyright 2009, Elsevier.

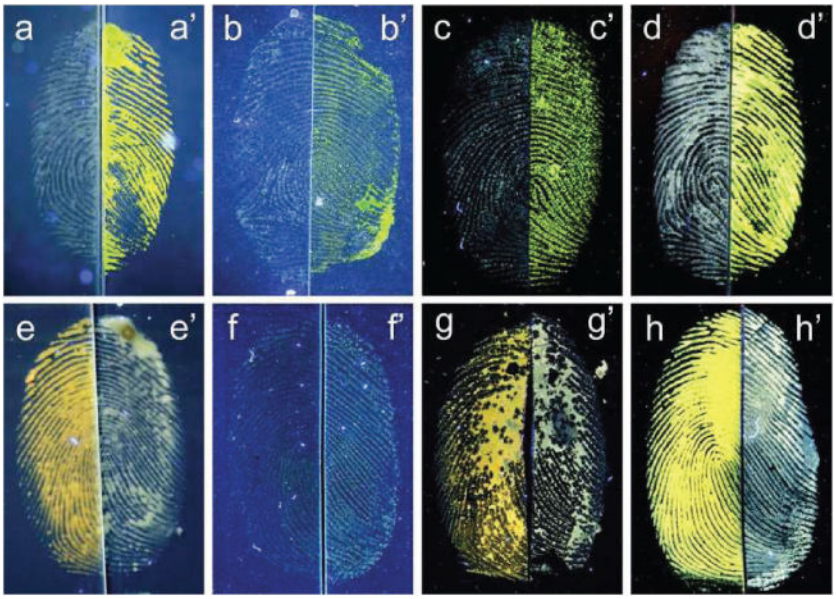


Figure 16.

Blood fingerprint development by using ZnS:Cu QDs (left haves, a–d; right halves, e′–h′), AY7 (right halves, a′–d′), and CdTe QDs (left halves, e–h), under the 300–400 nm UV excitation on a variety of substrates: (a, a′, e, e′) glass, (b, b′, f, f′) transparent polypropylene, (c, c′, g, g′) lack polyethylene, and (d, d′, h, h′) aluminum. Reproduced with permission.[62] Copyright 2013, Elsevier.

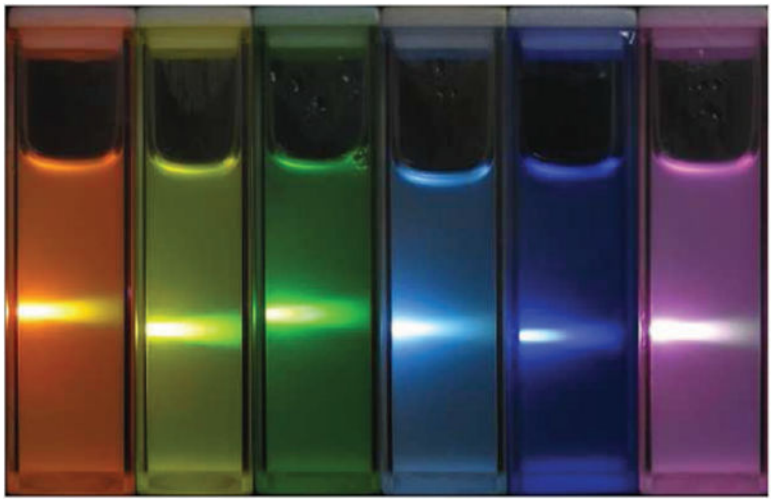


Figure 17.

Aqueous suspensions ofRE^{3+} ions-doped $NaYbF_4$ UCNMs with six distinguishable fluorescent emission colors excited with a 980 nm NIR radiation. From left to right, the UCNMs are $NaYbF_4$:Er, $NaYbF_4$:Er,Ho, $NaYbF_4$:Ho, $NaYbF_4$:Tm,Ho, $NaYbF_4$:Tm, and $NaYbF_4$:Er,Tm. Reproduced with permission.[65] Copyright 2009, American Chemical Society.

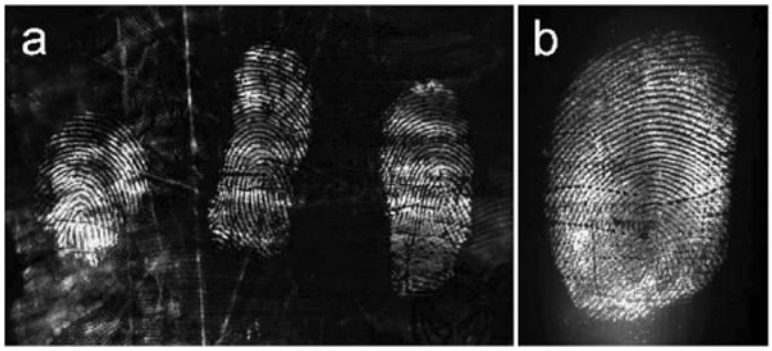


Figure 18.

Latent fingerprint development on Australian five-dollar polymer banknote by using a) $NaYF_4$:Yb,Er UC powders and b) YVO_4:Yb,Er UC powders under the 980 nm NIR excitation. (a) Reproduced with permission[71] Copyright 2011, Elsevier. (b) Reproduced with permission.[72] Copyright 2012, Elsevier.

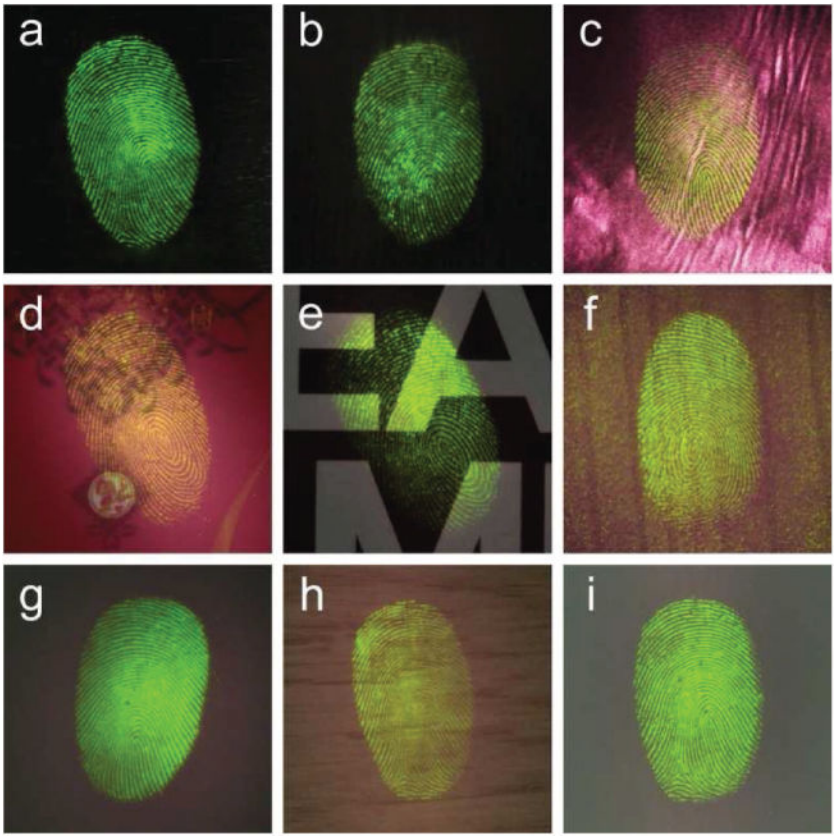


Figure 19.

Latent fingerprint development by using $NaYF_4$:Yb,Er UCNMs under the 980 nm NIR excitation on a variety of substrates: a) aluminum alloys sheets, b) stainless steel sheets, c) aluminum foils, d–e) plastic cards, f) floor leathers, g) ceramic tiles, h) wood floor, and i) painted wood. Reproduced with permission.[73] Copyright 2014, Springer.

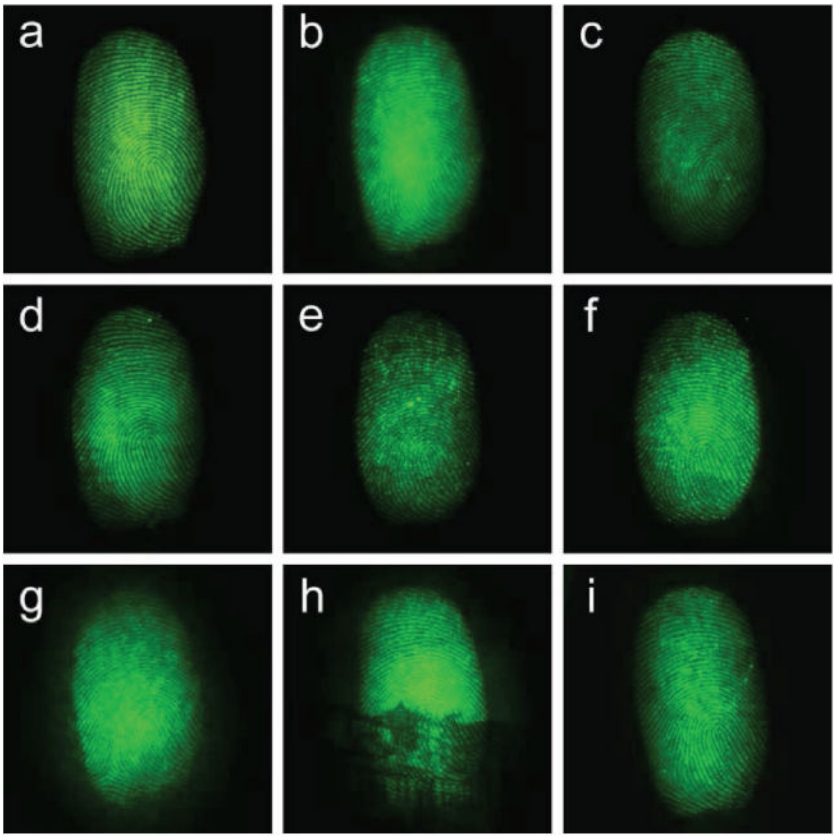


Figure 20.

Latent fingerprint development by using $NaYF_4$:Yb,Er UCNMs under the 980 nm NIR excitation. a–c) On the substrates with a single background color: (a) glass, (b) white ceramic tiles, and (c) black marbles. d–f) On the substrates with background color distraction: various marbles with different surface textures. g–i) On the substrates with background fluorescence interference: (g) note papers, (h) Chinese paper money, and (i) fluorescent plastic plates. Reproduced with permission.[74] Copyright 2015, American Chemical Society.

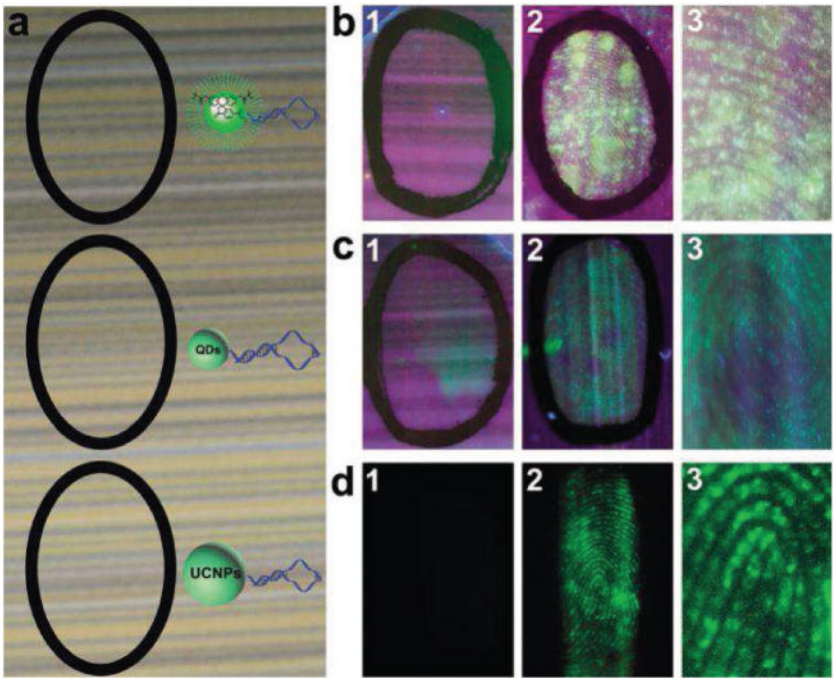


Figure 21.

a) Image of the marble with three latent fingerprints in the black circles, and the developers from top to bottom are FAM/LBA, CdTe/LBA, and $NaYF_4$:Yb,Er/LBA, respectively. b–d) Images of fingerprints treated by (b) FAM, (c) CdTe QDs, and (d) $NaYF_4$:Yb,Er UCNMs-based developers: from top to bottom, the images in row 1 are fingerprints treated by FAM, CdTe QDs, and $NaYF_4$:Yb,Er UCNMs, respectively; the images in row 2 are fingerprints treated by FAM/LBA, CdTe/LBA, and $NaYF_4$:Yb,Er/LBA, respectively; the images in row 3 are the corresponding magnified images of row 2. Reproduced with permission[38] Copyright 2014, John Wiley and Sons.

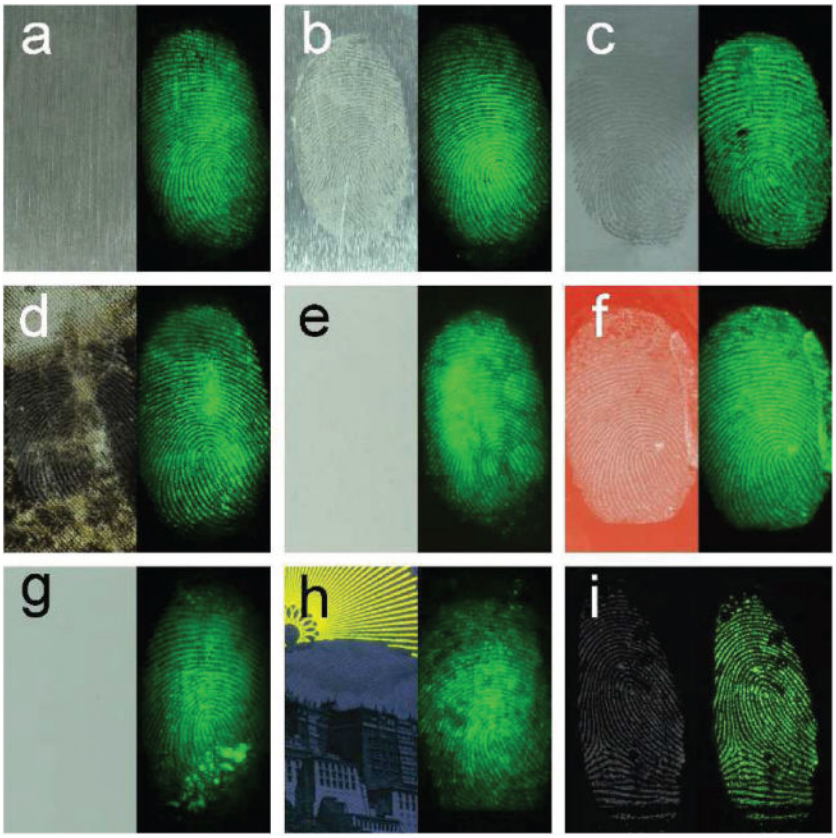
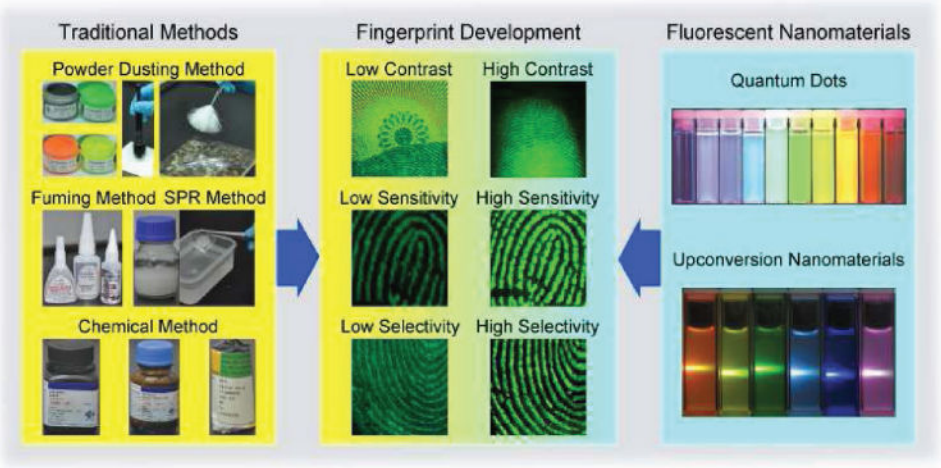


Figure 22.

Development of latent, fresh (2 h old) fingerprints by using a $NaYF_4$:Yb,Er UCNMs based suspension on a variety of substrates: a) stainless steel sheets, b) aluminum alloys sheets, c) aluminum foils, d) marbles, e) ceramic tiles, f) plastic cards, g) painted wood, and h) Chinese paper money. Development of latent, aged (1-year-old) fingerprints by using $NaYF_4$:Yb,Er UCNMs-based suspension on glass is shown in i). The left panels (except that in (h)) are images in a bright field without 980 nm irradiation; the left panel of (h) is the image under 254 nm UV excitation. Right panels show fluorescent images formed under 980 nm NIR excitation. Reproduced with permission.[32] Copyright 2016, Royal Society of Chemistry.



Scheme 1.

General idea of this review. Upper image in the right panel: Reproduced with permission.[47] Copyright 2001, Nature Publishing Group. Lower image at right panel: Reproduced with permission.[65] Copyright 2009, American Chemical Society. Images of the fingerprints in the middle pannel: Reproduced with permission.[74] Copyright 2015, American Chemical Society.