



From invisible to visible: a concise review on conjugated polymer materials in latent fingerprint analysis

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Abstract

Over the past decades, significant scientific and technological advancements have been made in the field of forensic science, particularly in the advancement of fingerprint technology. Latent fingerprints (LFPs) are considered to be the most crucial evidence found at crime scenes, necessitating extensive research efforts for their advancement. Conversely, conjugated polymers are extremely adaptable substances that are utilized in many domains like photonics, bio-imaging, optoelectronics, nanomedicine, and more. Conjugated polymers, especially conjugated polymer nanoparticles and polymer dots, exhibit remarkable photophysical and chemical features that contribute to their high resolution, enhanced selectivity, and heightened sensitivity in the development of LFPs. This provides a detailed overview of the production of LFPs utilizing conjugated polymers, as well as the imaging techniques employed for their visualization. Particular attention is given to conjugated polymer nanoparticles and polymer dots. In addition, the most often employed techniques for producing conjugated polymer-based nanoparticles have also been examined. Additionally, it encompasses the historical background of LFP identification and offers a valuable understanding of the diverse conventional approaches investigated for their advancement. This article offers a comprehensive examination of conjugated polymers as contrasting agents for imaging LFPs on different surfaces.

Keywords Latent fingerprints · Conjugated polymer nanoparticles · Polymer dots · Forensic science · Imaging

Introduction

Fingerprints are widely regarded as a highly valuable kind of physical evidence in the context of identification and criminal inquiry. The phenomenon has remained consistent since the inception of humanity. Fingerprints have significant evidentiary value owing to their distinctiveness and unchangeability in facilitating personal identification [1]. Dactylography, the study of fingerprints as a method of identification, is a crucial scientific field in criminal investigations. On the other hand, dermatoglyphics refers to the scientific study of the frictional ridge pattern present on the fingers, palms of

hand, toes and soles of feet [2]. A fingerprint refers to the imprint inscribed by the dermal layer located on the inner surface of the digits. Friction ridge skin refers to the skin on the inside surface of fingers, and fingerprints are formed by the presence of little ridges on the hands of each individual [3]. The friction skin is located on the digits and is distinct to each individual. The accumulation of secretions, such as sweat and oil, from the exposed area of the skin results in the formation of fingerprints. It is important to note that no two individuals possess an identical fingerprint pattern. Fingerprints are often regarded as highly effective tools in various domains such as unique identification, forensic investigations, and biometric password verification, owing to the distinctiveness of their ridge patterns. Typically, crime scenes commonly exhibit three distinct categories of fingerprints, namely latent fingerprints (LFPs), visible fingermarks, and impression marks. Among these, LFPs are imperceptible to the unaided eye and require specific procedures for accurate examination. Fingerprint patterns are typically categorised into three distinct levels, as depicted in Fig. 1. Level 1 refers to the arrangement created by the movement of ridges, and

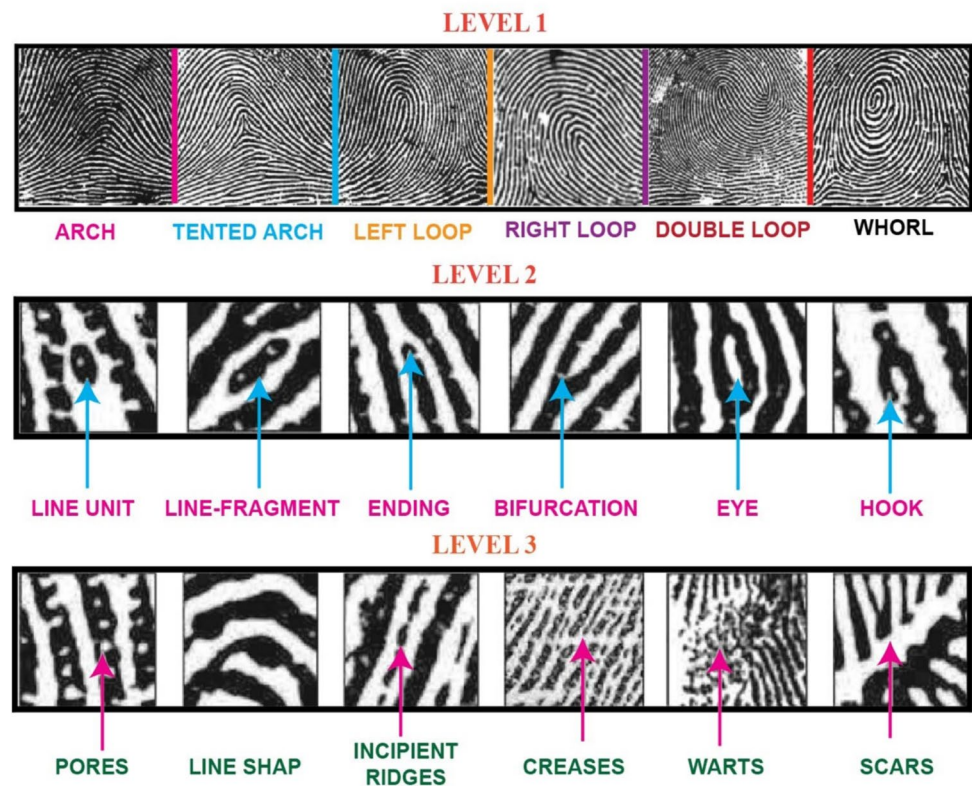
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Fig. 1 Fingerprint features at levels 1, 2 and 3 (adapted with permission from ref. 4)



three common patterns are commonly employed to delineate marks: whorl, loop, and arch. Level 2 encompasses the characteristics that emerge as a result of disturbances in the ridge flow, encompassing ridge terminations and bifurcation, wherein a solitary ridge divides into two separate branches. Additional characteristics can be described by the amalgamation of ridge terminations and bifurcations. Specialists rely on Level 2 details as the primary means of identification. Level 3 has characteristics linked to friction ridges, which may also be present in fingerprints. These traits can be utilised in conjunction with first and second level indicators to deduce the individual's identification [4, 5]. These characteristics may encompass pores, ridge edge forms, and interruptions within the ridge. This category may also encompass permanent scars and wrinkles that are present within the mark.

Fingerprints are widely recognised as the primary form of evidence in criminal investigations. These fingerprints can be broadly classified into two categories: patent or visible fingerprints, and latent fingerprints. Visible fingerprints are observable in the absence of any specific intervention and might manifest as positive, negative, or indented. The latent fingerprint is widely recognised as the predominant type of fingerprint evidence due to its inherent presence but imperceptible nature. The latent fingermarks, which are formed by the ridge of the finger, consist of an intricate combination of natural fluids and environmental pollutants. The skin

secretes three types of glands: sudoriferous eccrine glands, apocrine glands, and sebaceous glands [6].

The composition of sweat consists of water (> 98%), minerals (0.5%), and organic molecules (0.5%). Eccrine sweat is composed of several components such as protein, urea, amino acids, uric acid, lactic acid, carbohydrates, creatine, and choline. On the other hand, sebaceous sweat is composed of glycerides, fatty acids, wax esters, squalene, and sterol esters [7]. To visualise latent fingerprints, it is necessary to employ physical or chemical reagents as they are not perceptible to the unaided human eye. Physical approach for creating latent finger impressions requires interacting with impression deposits, while the chemical method involves developing latent fingerprints by a chemical reaction between the developers and the components of sweats [8]. The "Powder dusting" technique is a commonly employed physical approach for the development of latent fingerprints on nonporous surfaces. The process of powder dusting involves the attachment of fingerprint powder particles to the oily or watery components present in fingerprint deposits. There are four distinct categories of fingerprint powders, regular, luminous, metallic, and thermoplastic [7]. The formation of LFPs involves the utilisation of many chemical processes, such as the iodine fuming process, ninhydrin method, silver-nitrate method, and cyanoacrylate method, which are selected based on the specific characteristics of the surfaces involved. With time, there has been a notable

progression in both technology and criminal actions. Consequently, conventional methodologies have demonstrated diminished efficacy in the identification of historical fingerprints, as well as in terms of their durability and differentiation. Hence, it is crucial to identify cost-effective and eco-friendly fluorescent reagents to develop latent fingerprints.

Conjugated polymers, when utilised as fluorescent materials, have several notable advantages in comparison to other small molecular fluorophores which include their ease of synthesis, cost effective, good emission capabilities, and resistance to photobleaching. Additionally, conjugated polymers demonstrate favourable mechanical stability and processibility [9]. The utilisation of nanoparticles, polymer nanoparticles, polymer dots, and nanoparticle electrolytes has demonstrated significant potential in enhancing the sensitivity and contrast of latent fingerprints. This is attributed to their distinctive characteristics, such as high fluorescence intensity, large Stokes shift, and strong photochemical stability [10]. Conjugated polymer nanoparticles (CPNs) have gained prominence as versatile materials at the nanoscale, exhibiting significant promise in the fields of biosensors, imaging, and optoelectronics. This is primarily due to their tailored emission characteristics, compatibility with biological systems, and reduced toxicity when compared to inorganic nanoparticles, rendering them preferred materials.

Over the past decade a lot of research has been done in the field of LFP analysis and every year the count of articles being published is increasing enormously as depicted in Chart 1. The present review provides a concise overview of the significance of LFP imaging, the historical background of fingerprint identification, conventional techniques employed in the creation of latent fingerprints and their related constraints, as well as contemporary approaches to CPN synthesis. Moreover, this review comprehensively addresses the advancement of latent fingerprints using CPNs, encompassing polymer dots, and concludes by presenting a future outlook for LFP imaging.

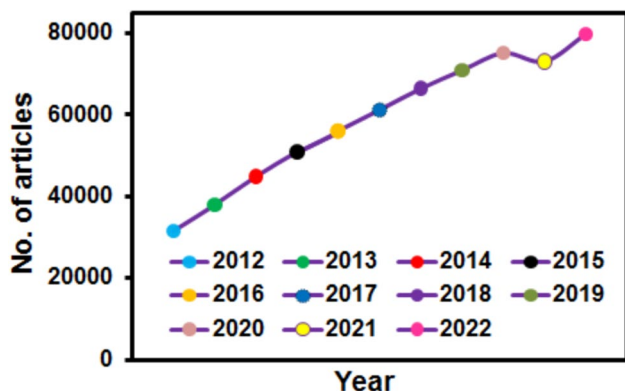


Chart 1 Graphical representation of the number of articles being published yearly over the past decade in the field of LFP analysis

History of fingerprint identification

Fingerprint identification possesses a significant historical background that may be traced back to ancient Babylon, wherein fingerprints were employed on clay tablets as a means of facilitating commercial transactions. Fingerprints are widely regarded as the most ancient forms of friction ridge skin imprints that have been documented thus far. In Babylon, fingerprints were detected on clay tabs, seals, and ceramics that contained written deeds dating back to the second millennium BC [11–13]. During the reign of King Hammurabi (1792–1750) BC in Babylon, law enforcement agents utilised these fingerprints to apprehend individuals [14]. Fingerprints have been observed on the walls of Egyptian tombs, as well as on potteries from ancient Greece and China. Additionally, they have been detected on bricks and tiles recovered in ancient Babylon and Rome [13]. Hand and foot prints were gathered and utilised as evidence throughout the Chinese Dynasty. In the year 650, the Chinese historian Kia Kung Yen and the Arab trader Abu Zayd Hasan observed fingerprints being used as a method of verification [15, 16]. Rashed-al-Din Hamadani (1247–1318), a Persian physician, makes reference to the Chinese tradition of fingerprint identification and observes that empirical evidence indicates the absence of identical fingerprints across individuals [17]. The detailed description of friction ridge skin was initially provided by Dr. Nehemiah Grew in 1684 [18]. The anatomy of the human body, including the skin and the papillary ridges of the fingers, was published by Gorarrd Bidloo [19]. In 1687, Marcello Malphigi documented in his book that the friction ridge of the skin possesses distinct characteristics in terms of its function, form, and structure [20]. Dr. Purkinge categorised fingerprint patterns into nine distinct groups in 1823 and assigned each category a specific name [21]. Subsequently, it facilitated the development of the Henry Classification System [22]. In 1877, Sir William Harschel in India initiated the utilisation of fingerprints for the purpose of registering inmates and executing contracts and deeds [23]. A subsequent establishment of a fingerprint bureau took place in Kolkata. Azizul Haque and Hem Chandra Bose, two Indian fingerprint experts, were the main contributors to the development of a fingerprint categorization system called the Henry classification system, named after their supervisor [24, 25]. The initial publication on fingerprints was authored by Henry Faulds in 1880 and was included in the scientific journal 'Nature'. Faulds emphasised the significance of the friction ridge skin for the purpose of individualization, particularly in its application as evidence [26]. Subsequently, Sir Francis Galton conducted additional research on the subject and demonstrated that the probability of a false positive

fingerprint was approximately 1 in 64 billion individuals [27]. Juan Vucetich, a prominent fingerprint researcher, was employed by the Central Police Department in Argentina. He initiated an investigation into the fingerprints of offenders and devised his own system for categorising them. In 1892, the person established the inaugural fingerprint bureau globally, employing fingerprints as a means of identifying criminals within the judicial system. Consequently, Argentina achieved the distinction of being the inaugural nation to exclusively depend on fingerprints as a means of individualization [28]. Currently, it is widely employed on a global scale to identify individuals in the context of legal disputes and inquiries within the courtroom. In 1962, Chatterjee postulated that the integration of ridge edges with other friction ridges would contribute to the development of a unique characteristic known as edgeoscopy. The author successfully categorised the shapes of friction edges into eight distinct varieties, namely straight, pocket, angle, table, peak, concave, and convex. The permanence and distinctiveness of sweat pore positions and ridge edge morphologies have been identified as factors that aid in the identification of individuals.

Traditional methods of LFP development

Scientists have employed many conventional techniques, such as the powder dusting method, iodine fuming, silver nitrate method, and 1,2 indanedione (1,2 IND), to create latent fingerprints in recent decades. Nevertheless, even conventional approaches possess certain limitations. Due to their notable attributes of high contrast, selectivity, and sensitivity, efficient techniques have been devised for the advancement of latent fingerprints.

Powder dusting method

The powder dusting technique is a time-honoured method for developing fingerprints and remains widely employed worldwide. Adsorption of powder particles onto moisture and greasy fingerprint components is observed in this approach. This method is utilised for surfaces that lack porosity [29]. The process of powder dusting involves the application of powder onto fingerprint remnants through brushing. Various types of powders are employed in the process of developing latent fingerprints, encompassing ordinary powders such as Black and White Powder, metallic powders like Aluminium Powder and Magnetic Black Powder, as well as fluorescent powders like Greenecent and Pinkecent Fluorescent fingerprint powder [30]. The composition of normal powders typically exhibits variation however it typically consists of a binder and a colour ingredient. The pigment material facilitates surface contrast and enables visualisation,

whereas the binder material enhances the adhesion to the fingerprint components in a maximal and selective manner [29]. Several pigment substances include carbon black, talc, kaolin, aluminium, and metal flakes. On the other hand, iron powder, maize starch, and gum Arabic are examples of effective binders [30]. Metallic powders consist of several metals, including but not limited to iron, aluminium, copper, bronze, and numerous others, which are present in their composition. The application of these powders is mostly done with a magnetic brush due to their predominantly magnetic properties. One notable benefit of employing this technique is the absence of brushing, hence reducing the likelihood of fingerprint damage. Fluorescent powders are comprised of organic chemicals that possess fluorescent or phosphorescent characteristics upon exposure to ultraviolet (UV) or laser emissions. Examples of such substances are violet crystal and coumarin. These powders are employed for surfaces that exhibit reflectivity or multipolarity, hence potentially resulting in contrast issues when conventional powders are utilised [6, 31].

Iodine fuming method

For millennia, the iodine technique has been employed. Previously, it was thought that a chemical reaction takes place between iodine and fingerprint compounds. However, recent research indicates that physical adsorption occurs. When heated, the iodine crystals undergo sublimation, resulting in the formation of a purple vapour. The vapour subsequently forms a connection with the lipidic chemicals present in the fingerprint, a phenomenon that is thought to take place via non-covalent intermolecular interactions, such as van der Waals forces. Consequently, the acquisition of yellow-brownish fingerprints is observed. Nevertheless, the colour is not enduring and gradually vanishes over time [7, 32].

Ninhydrin (NH) method

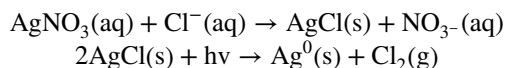
This approach was developed in the middle of the 1800s and is still one of the most often used methods for creating LFPs on a variety of porous materials, including paper, cardboard, and plasterboard. Using this technique, the amino acids on the fingerprint residues react with NH to produce Ruhemann's purple product, which reveals the LFPs [33–35].

Amino acids are the right targets for creating old fingerprints because they get impregnated into the surface of porous substrates like paper during the production of LFP residues and stay there for a longer amount of time [36]. The interaction between NH and the fingerprints needs to occur under high humidity (50–80%) and an optimal pH range of 4.5–5.5 for this to emerge. Notably, to prevent Ruhemann's purple from deteriorating when exposed to light and oxygen, the treated LFPs should be stored in a cool, dark

environment. Furthermore, while heat and steam treatment is frequently performed to increase the NH reaction rate, a temperature rise might result in background discolouration.

Silver Nitrate method

Silver Nitrate is an achromatic, translucent, cylindrical crystal that transforms into a shade of grey or greyish black when exposed to light. The process creates hidden imprints on both absorbent and non-absorbent substances. The utilisation of silver nitrate for the formation of LFPs is a time-honoured method, employed since 1891, albeit its usage has diminished in contemporary times. The methodology employed in this study is founded upon the fundamental assumption that the interaction between silver nitrate and the chloride anion found inside the fingerprint residue occurs. The development progresses through two distinct stages. The precipitation reaction of silver chloride (AgCl) is initiated by the reaction described in the references.



Subsequently, upon exposure to ambient light, namely ultraviolet (UV) radiation, the silver chloride undergoes a series of oxidation–reduction reactions, ultimately resulting in the formation of solid silver. The dark grey colour of elemental silver enables the visualisation of fingerprints [30, 32]. This reagent exhibits favourable performance on porous surfaces due to two primary factors. One notable observation is that the rate of precipitation reaction surpasses that of dissolution. The second observation pertains to the retention of AgCl at the interstices of the surface where the fingerprint residues were previously absorbed, which can be attributed to its insolubility.

Cyanoacrylate fuming method

Super glue fuming technique is the term used to describe this process. It was developed in the latter half of the twentieth century and is used to leave fingerprint residues on any nonporous surface. By introducing vaporised cyanoacrylate ester monomers into the LFPs, this approach forms a durable white polymer that covers the fingerprint residues through polymerization in the presence of initiators such as water, acid, alkali, etc. [33, 34]. By encouraging the volatilization of the cyanoacrylate ester monomer by a variety of acceleration methods, including heat, chemical, and vacuum acceleration, the fuming period can be decreased. However, because of the white matrix that appears in LFPs created with this method, there is not as much contrast, which makes LFP visualisation difficult.

An easy and effective way to create LFPs on nonporous surfaces is via superglue fuming. However, this method's broad use is limited by serious health risks, such as skin and eye damage caused by its vapours [37, 38].

2-Indanedione (1, 2-IND) method

The compound known as 1, 2-indanedione was initially synthesised in 1997 and is primarily employed in the visualization of latent fingerprints on porous surfaces. The reaction between 1, 2-IND and amino acids yields a pale pink product that exhibits intense luminescence when exposed to light with a wavelength ranging from 480 to 560 nm [39]. The reaction mechanism of 1, 2-IND exhibits a high degree of similarity to the DFO mechanism. In conclusion, a chemical reaction occurs between a 1, 2-IND molecule and another molecule, leading to the formation of Joullié's Pink (JP). JP is an azomethine ylide characterised by enhanced conjugation extension, a pale pink hue, and fluorescence [39, 40]. One limitation of this approach is the potential for colour and luminosity degradation to manifest within a few days. Nevertheless, research has demonstrated that the inclusion of zinc or cadmium salts can enhance colour and luminescence, while also extending the duration of these effects through a complexation reaction between the metal and JP.

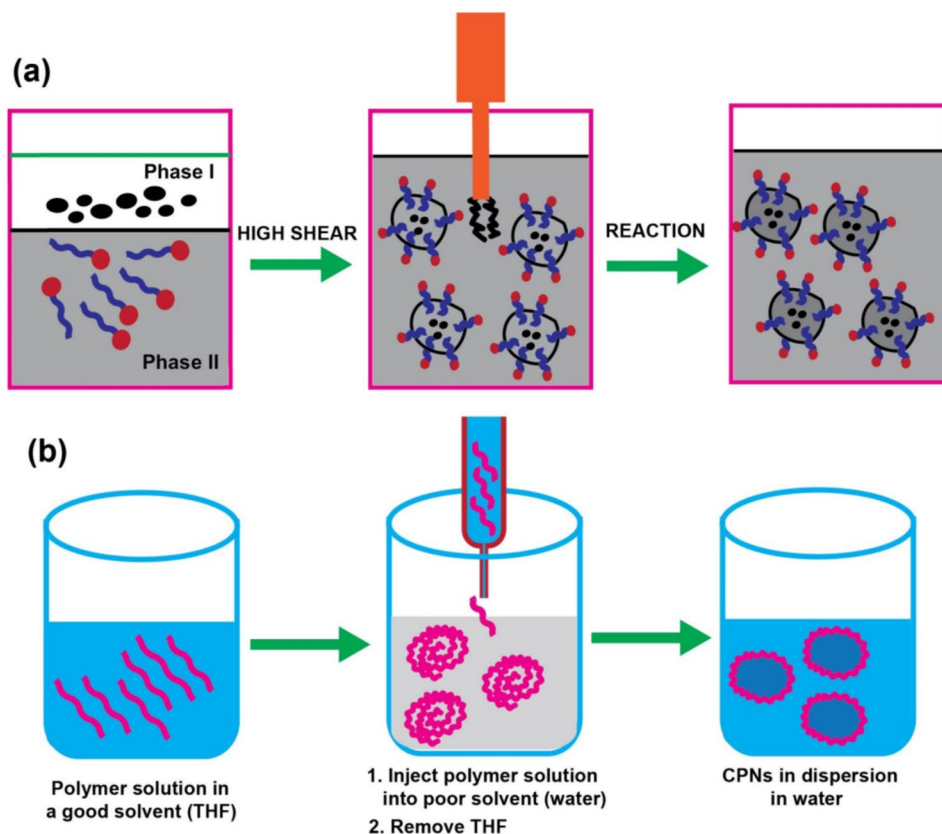
Methods to prepare CPNS

The primary methods employed for the synthesis of conjugated polymer nanoparticles (CPNs) include mini-emulsion and reprecipitation techniques [41]. In this part, a concise overview of both preparation procedures will be provided.

Miniemulsion

It is a widely used technique for preparing CPNs. This approach [42–44] involves dissolving a conjugated polymer in an organic solvent and subsequently injecting it into an aqueous solution that contains a suitable surfactant. The resultant mixture is subjected to ultrasonication, leading to the formation of a stable miniemulsion consisting of polymer solution in the form of minute droplets, as depicted in Fig. 2a. Subsequently, the organic solvent is evaporated to achieve the dispersion of CPNs in an aqueous solution. The particle size within the range of 30–500 nm can be achieved, with the specific value being contingent upon the concentration of the polymer solution. In this particular scenario, the droplets that are generated may experience destabilisation through the processes of Ostwald ripening and flocculation. The prevention of flocculation can be achieved through the incorporation of appropriate surfactants, while the reduction of Ostwald ripening

Fig. 2 Methods of CPNs preparation (a) Miniemulsion and (b) Reprecipitation method (inspired from ref. 41. Copyright 2017, RSC)



can be achieved by introducing a hydrophobic agent. This substance facilitates the generation of osmotic pressure within the droplet, hence impeding its diffusion into the surrounding aqueous medium.

Reprecipitation

It involves dissolving [45–49] units of the conjugated polymer in a suitable solvent, such as an organic solvent like THF. Subsequently, the resulting polymer solution is transferred into a less suitable solvent, such as water, which exhibits miscibility with the aforementioned solvent. The resulting mixture is subjected to strong sonication to generate nanoparticles. Subsequently, the organic solvent is eliminated, resulting in the dispersion of nanoparticles in water, as depicted in Fig. 2b. The primary factor contributing to the generation of nanoparticles in this particular scenario is the hydrophobic phenomenon, wherein the polymer chain endeavours to evade interaction with water upon immersion in the aqueous medium. Consequently, the polymer chains undergo folding, resulting in the production of spherical structures. In this scenario, no additional substances such as surfactants or hydrophobes are included. This technique successfully generates nanoparticles from a single polymer chain, with a size range of approximately 5–10 nm.

Development of latent fingerprints (LFPS) using conjugated polymer (CP) materials

Over the past few decades, a wide range of contrast agents and imaging techniques have been employed to visualise LFPs. Several factors significantly impact the resolution and sensitivity of LFP imaging when employing conventional contrast agents [50–56]. In the field of high-resolution fingerprint visualisation, numerous advanced fluorescent techniques and contrast agents have been investigated. These include small molecules, quantum dots, rare earth upconversion nanomaterials, silane-modified conjugated oligomers, AIE aggregates, and metal–organic frameworks (MOFs), among others. Fluorescent contrast agents demonstrate a higher level of efficacy compared to non-fluorescent reagents, mostly attributed to their exceptional sensitivity and ability to effectively generate fingerprints on diverse coloured surfaces. Despite the effectiveness of these fluorescent probes in visualising LFPs under certain conditions, some of them still experience significant problems such as photoblinking, photobleaching, and toxicity. Hence, it is imperative to acknowledge and rectify the aforementioned constraints associated with these probes.

Due to their exceptional physical and chemical properties, conjugated polymers have garnered considerable attention in various fields of chemical, material, and biological science.

These properties include strong fluorescence emission, high quantum yield, resistance to photobleaching, easy synthesis, and low toxicity, among others. In addition, the optoelectronic properties of CP, along with its delocalized backbone structure, contribute to its advantageous high absorption cross-section and efficient intra-/inter chain excitation transfer. Consequently, CP exhibits exceptional sensitivity, a low detection limit, and high spatiotemporal resolution, making it highly suitable for applications in biosensing and imaging. CP materials are currently engaged in perpetual competition with both established technology and emerging materials to detect fingerprint impressions on diverse surfaces. This is primarily attributed to their exceptional light-amplifying and light-harvesting characteristics. CP possesses several advantageous qualities that make it the preferred material for the efficient creation of LFP images. These properties include ease of operation, high selectivity and sensitivity, little fingerprint contamination, high resolution, and increased contrast on various substrates. CP enables the generation of high-quality LFPs with level 3 features, hence offering distinguishing information for individual identification. The preservation of material evidence, such as fingerprint-containing articles, at crime scenes holds significant importance within the field of forensic science. CP can effectively and non-destructively visualise and maintain the latent fingerprint for an extended period. In recent studies, researchers have investigated the potential of π -conjugated polymer nanomaterials, including polymer nanoparticles and polymer dots (PDots), for latent fingerprint imaging. These materials have garnered attention due to their notable characteristics, such as high single-particle fluorescence, high absorption coefficient, ease of synthesis, and changeable emission wavelength. Polydots (PDots) are a distinct classification of Conjugated Polymer Nanoparticles (CPNs) characterised by a size smaller than 30 nm. Additionally, to get a high level of single-particle brightness, it is recommended that PDots contain a minimum of 50% conjugated polymers. Furthermore, the interior of the PDots must exhibit hydrophobic properties. There exist three essential factors that serve to distinguish PDots from CPN. The preparation process plays a crucial role in determining the size and optical characteristics of conjugated polymer nanoparticles. In addition, the desirability of conjugated polymer nanoparticles for latent fingerprint generation lies in their customisable size, changeable optical characteristics, facile manufacturing, and varied surface functionalization.

LFP imaging via conjugated polymer nanoparticles

Poly-(p-phenylenevinylene) (PPV) nanoparticles were synthesised by Fan and coworkers [57] using a modified Wessling method. This method involves the thermal elimination of sulfonium groups and the formation of a C=C bond

on the backbone with the addition of the catalyst TEA57 and the surfactant SDS (to prevent the precipitation of PPV aggregates from aqueous solution) (Fig. 3a). PPV-NPs exhibit an emission of 500 nm and a shoulder at 530 nm upon excitation at 365 nm (Fig. 3b). To create LFPs, a variety of adhesive tape kinds as well as additional substrates including cover glass and aluminium foil were employed. After being submerged in the developing solution for five to ten minutes, the substrate was removed, cleaned with distilled water, and allowed to dry. The images were acquired under a UV lamp (365 nm) using a digital camera. It was found that treating the fingerprints with different organic and aqueous solutions either before or after development did not affect the developing solution's exceptional efficiency in imaging both young and old visible fingerprints. Furthermore, taking advantage of the high magnification photographs more particular details might be acquired for the identification of an individual (Fig. 3c). The mechanism for the selectivity and high resolution of the developing solution towards the fingermarks is suggested to be the hydrophobic interaction between PPV and the oily component in the finger secretion.

Fan et al. [58] successfully combined CPNs with the superglue fuming process to create bright LFPs. A range of CP nanoparticles PPV-N were documented, each with different N values (0, 1, 2, 3, and 4), which corresponded to different substitution reaction times (0, 12, 24, 48, and 72 h) accordingly. The PPV-NPs were synthesised in an aqueous solution through the substitution of sulfonium with methoxy groups, followed by the heat removal of sulfonium groups and the formation of C=C bonds on the main chain. This process was carried out in the presence of the anionic surfactant SDS and TEA catalyst (Fig. 4). The process of LFP development was conducted on a range of surfaces, including glass sheets, aluminium foil, and others, encompassing a two-step procedure. Initially, the fingerprints were subjected to in situ fumigation using superglue ethyl-2-cyanoacrylate ester, a potent adhesive. This procedure was conducted using a self-constructed apparatus that operates in a manner analogous to the specialised chamber employed in the police system for the fuming process. Furthermore, the fingerprints were subsequently stained utilising the PPV-N NPs solution, which is regarded as the primary stage in the advancement of LFPs through this technique. In order to facilitate comparison, fumed fingerprints were divided into two sections: one section was labelled with PPV-0 NPs, while the other section was labelled with R6G. It was demonstrated that the images produced by PPV-0 are clearly visible, but the other half produced by R6G is less readable. The study's findings indicate that the utilisation of the superglue fuming process, in conjunction with the introduction of the fluorescent PPV-N NP solution, is a viable and efficient approach for the development of LFPs.

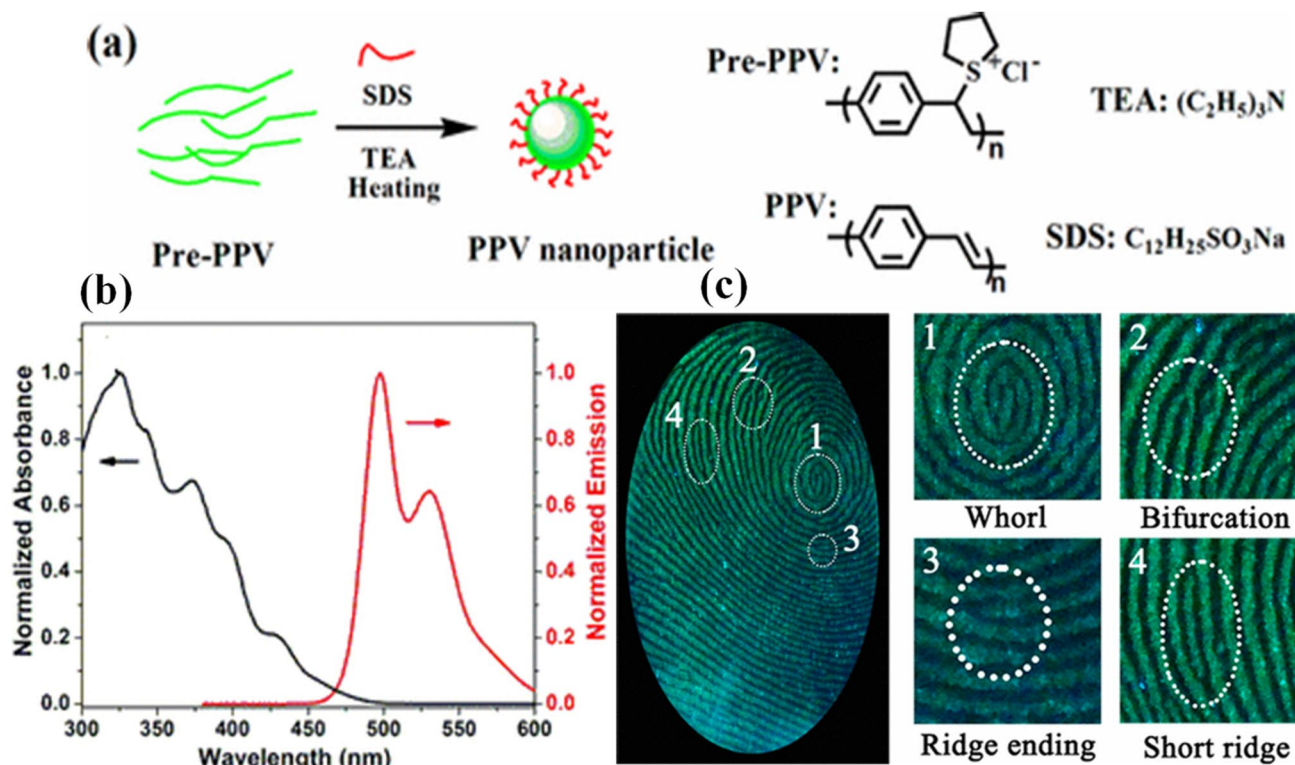


Fig. 3 **a** The (p-phenylenevinylene) (PPV) nanoparticle production and chemical structures used are depicted schematically. **b** The Normalized emission and absorption spectra of the developing solution with excitation occurring at a wavelength of 365 nm. **c** Fluorescence

images depicting the development of fingerprints, along with magnified images that exhibit 1–3 level details (adapted with permission from ref. 57. Copyright 2017, ACS)

Li-Juan Fan and group [59] synthesised amino functionalized poly (p-phenylene vinylene) branched polyethyleneimine nanoparticle (PPV-brPEI NPs) using an aqueous colloidal solution as the development reagent. The study revealed that brPEI molecules exhibited dual functionality, facilitating the heat removal of PPV precursors and engaging in the self-assembly process to generate nanoparticles. The successful development of latent sebaceous fingerprints (LSFPs) and latent blood fingerprints (LBFPs) on various nonporous substrates was achieved by utilising a cotton pad saturated with PPV-brPEI NPs. The aforementioned approach demonstrated a high level of sensitivity and efficacy in detecting aged, polluted, and mouldy fingerprints. The developed photos were digitally magnified to accurately identify level 1, 2, and 3 details. The process elucidates that the interplay between PPV-brPEI and sebum constituents plays a role in the formation of latent sebaceous fingerprints, while the interaction between PPV-brPEI and proteins present in the bloodstream contributes to the formation of latent blood fingerprints. Overall, Li-Juan Fan and coworkers presented a straightforward, environmentally sustainable, and economically efficient approach that holds significant promise for future use in real-world criminal investigations (Fig. 5).

Latent fingerprint development via conjugated polymer dots

Oxetane-functionalized polymer dots, or Ox-Pdots, were synthesised by Wu et al. [60] to visualise LFPs using covalent patterning. To create blue, green, and red-emitting polymers, respectively, fluorene monomer functionalized with oxetane groups was first synthesised. Fluorene, (4,7-dibromobenzo[c]-1,2,5-thiadiazole, and 4,7-bis(2-bromo-5-thienyl)-2,1,3-benzothiadiazole) were then copolymerized with these monomers. To achieve good colloidal stability, poly-styrene-comaleic anhydride (PSMA) was used as a functional polymer in the nanoprecipitation process to prepare Ox-Pdots of the appropriate CPs. Strong resolution imaging of LFPs is achieved by the aforementioned Pdots due to their strong fluorescence, substantial Stokes shift, and good quantum yields. Various surfaces, including an iron spoon, a glass microscope slide, aluminium foil, and a plastic Petri dish, were used to create LFPs. Every fingerprint was first surrounded by a hydrophobic barrier. A Pdot solution was then applied to the marks, and they were let to sit at room temperature for one to five minutes. The sample above was exposed to a 3 weight percent aqueous solution of triarylsulfonium hexafluorophosphate salts (a

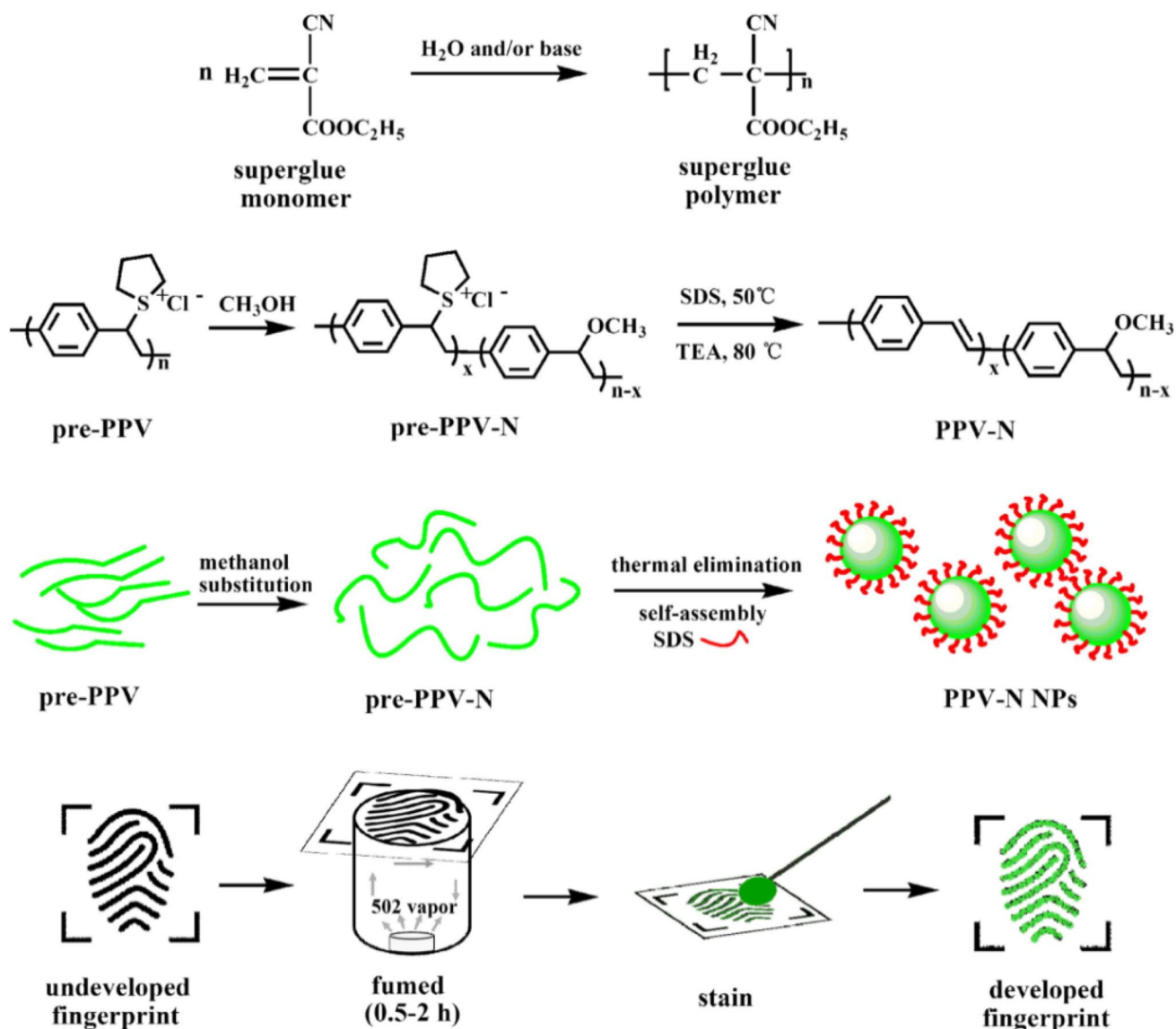


Fig. 4 The diagram illustrates the process of generating *poly*(p-phenylene vinylene) nanoparticles (PPV-NPs) and then developing fingerprints using superglue fuming, followed by staining with PPV-NPs

in an aqueous colloidal solution (adapted with permission from ref. 58. Copyright 2018, Elsevier Inc)

photoinitiator) under UV lamp irradiation for 10 s at room temperature to photo-crosslink, which produced a 3-D intermolecular polymer network (Fig. 4). It was found that Ox-Pdots, which produce far better and more consistent image quality, yield greater resolution than Oxetane-free Pdots. It was hypothesised that fast and high-quality imaging of the LFPs is caused by high frequency ether groups on the Ox-Pdots particularly interacting with fatty residues of the fingerprint ridges (Fig. 6).

Chan et al. [61] developed NIR fluorescent semiconducting polymers PF-BT-DBT by the process of Suzuki polymerization. The nanoprecipitation method was employed to prepare polymer dots (Pdots) of the PF-BT-TQ polymer that

was embedded with ninhydrin (Fig. 7a). This was achieved by blending the Pdots with carboxyl-functionalized polystyrene (PS-PEG-COOH), thiol-terminal polystyrene (PS-SH), and polyoxyethylene nonylphenylether (CO-520) to customise the functional groups of the resulting Pdots. Both polymers exhibited a significant Stokes shift of around 200 nm and focused on UV–vis absorption throughout the wavelength range of 360–500 nm. PF-BTDBT Pdots exhibited an emission peak at a wavelength of 655 nm. Conversely, PF-BT-TQ Pdots had emission maxima at a wavelength of 680 nm, with quantum yields of 0.29 and 0.32, respectively.

The detection of LFPs was conducted on a variety of substrates, including porous and nonporous materials such as

Fig. 5 a Schematic diagrams illustrating the process of preparing poly(p-phenylenevinylene) branched polyethyleneimine nanoparticles (PPV-brPEI NPs) and the subsequent fingerprint development.

Fluorescent photos and regional images were digitally magnified to capture the intricate details of sebum (a) and blood (b) fingerprints that had grown on steel. These images were captured using 365 nm UV light (adapted with permission from ref. 59. Copyright 2023, Elsevier Inc)

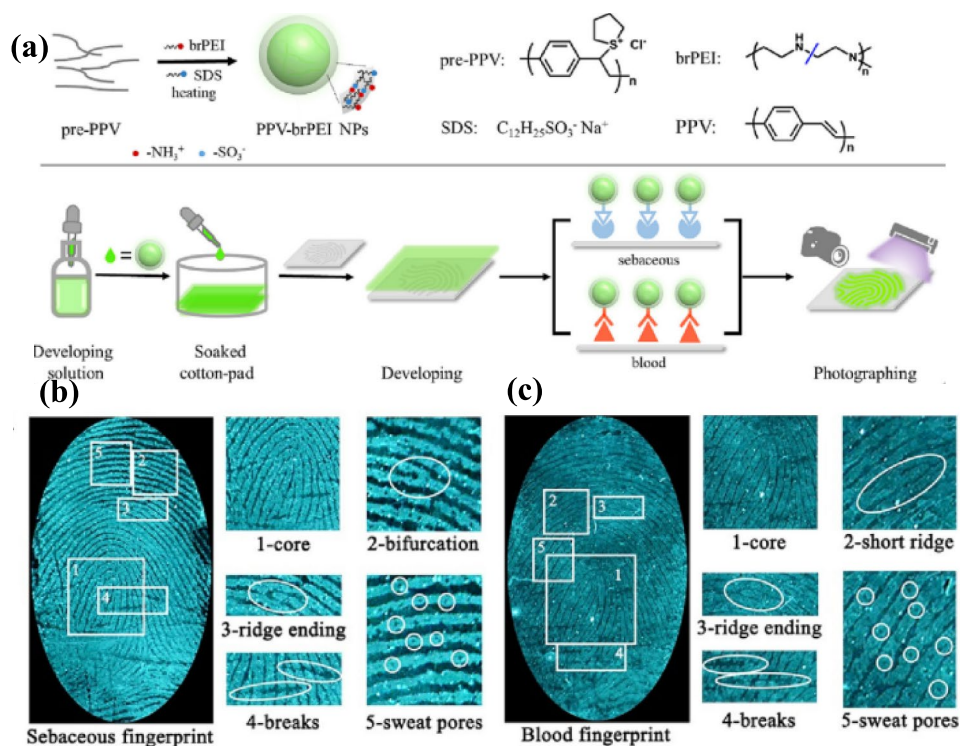
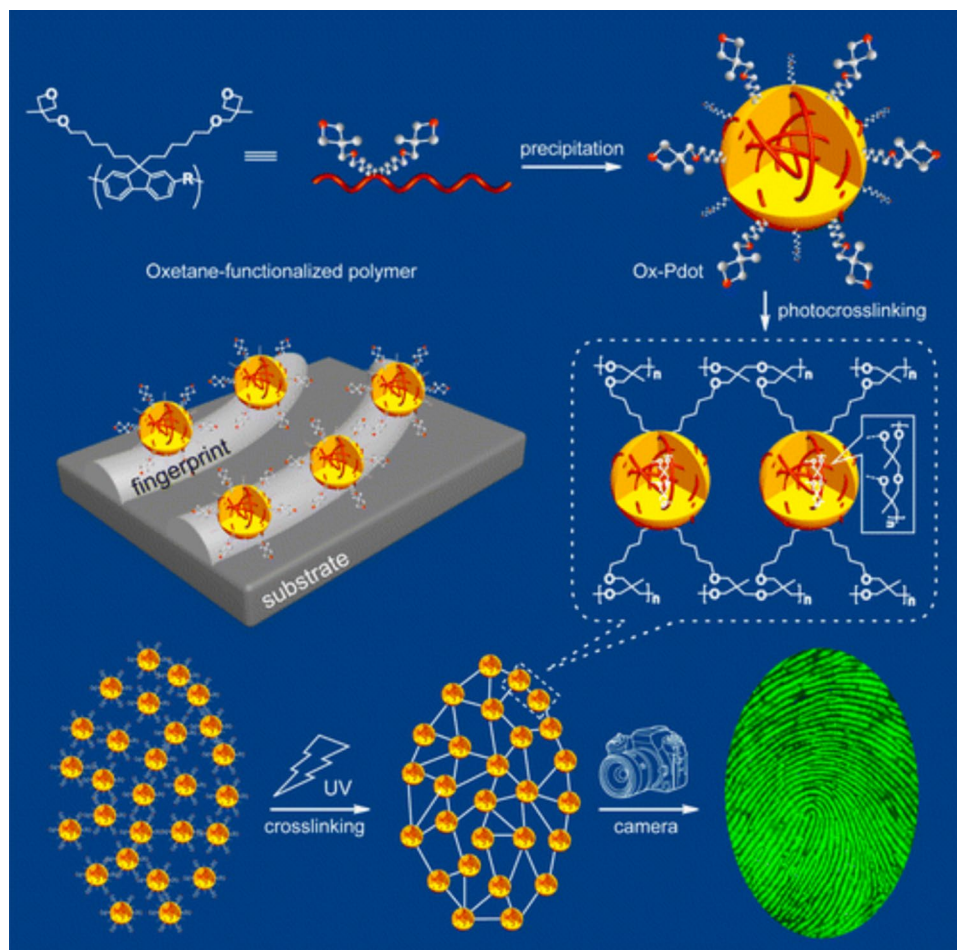


Fig. 6 Graphical representation and chemical arrangement of LFPs using photo-cross-linked Ox-Pdots (adapted with permission from ref. 60. Copyright 2015, ACS)



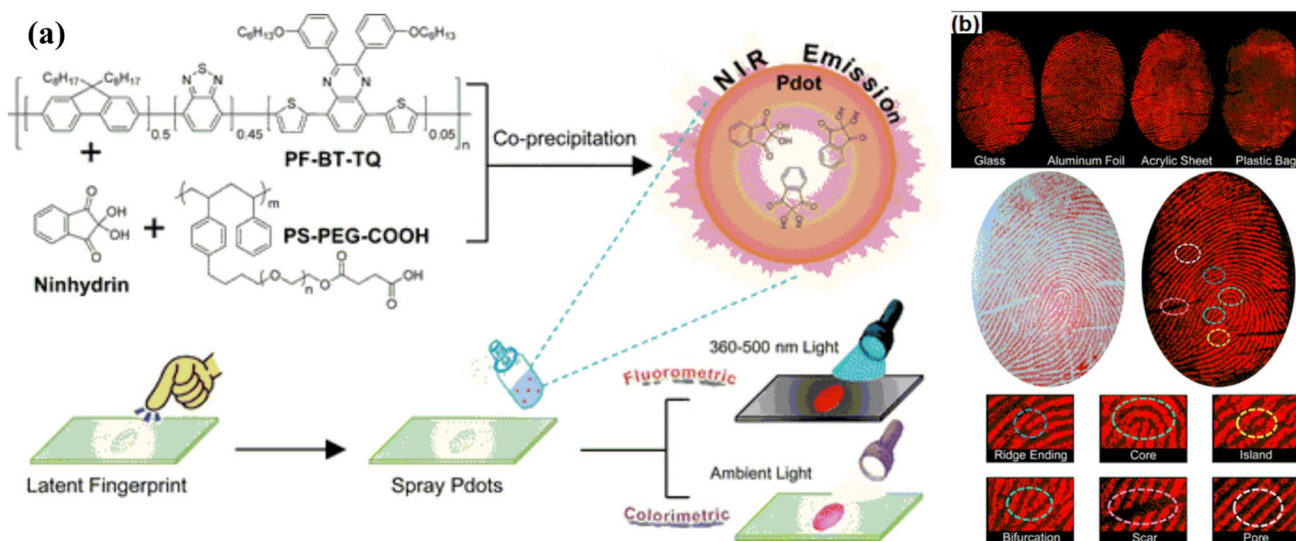


Fig. 7 **a** Semiconducting polymer polyfluorene-2,1,3-benzothiadiazole-5,8-bis(thiophen-2-yl)-2,3-bis(3-(hexyloxy)phenyl)quinoxaline (PF-BT-TQ) carboxyl-functionalized polystyrene PS-PEG-COOH, and ninhydrin dyes. **b** High-resolution images of latent fingerprints

paper, glass, aluminium foil, plastic bags, and acrylic sheets. The process of creating fingerprint impressions on different surfaces involved initially rubbing the fingertips across the nose and forehead, followed by applying gentle pressure to the surface. A volume of 0.2–0.5 mL of Pdot solution was applied directly onto the fingerprint of nonporous surfaces and allowed to dry at ambient temperature. A spray bottle containing Pdot solution and a 5–20% (v/v) acetone solution were used to apply the solution onto porous surfaces. The substrates were then dried in an oven at a temperature of 60 °C for 10 min. The LFPs that were created were captured using a digital camera in both UV and ambient light conditions. Figure 7b displays fingerprint pictures created with NH-embedded PF-BT-TQ Pdots on various surfaces. The naked eye was able to easily perceive the purple colour of the generated prints on white, green, and red paper, but not on blue paper. Conversely, well-developed pictures can be readily observed when exposed to 450 nm light with minimal interference. Moreover, this new technique enables the easy recognition of details at levels 1, 2, and 3 with great resolution and sensitivity, so showing its viability for LFP development. The excellent resolution of the forming images is related to the interactions between the components contained in fingerprints and the Pdots, which involve both electrophilic and hydrophobic interactions.

Conclusion

This review aims to present an overview of the significance of CPs for LFP imaging (Table 1) and also provides current information on different techniques and methods

were obtained using Pdots on various materials such as glass, aluminium foil, acrylic sheets, and plastic bags exhibiting level 1–3 details (adapted with permission from ref. 61. Copyright 2016, ACS)

used to create latent fingerprints. These methods include traditional approaches such as powder dusting, iodine fuming, and the silver nitrate method. However, it is important to note that these traditional methods have certain limitations. The unique optical properties of conjugated polymer materials, particularly conjugated polymer nanoparticles and polymer dots, make them highly promising for the construction of latent fingerprints. These traits include high quantum yield, emission tuneable characteristics, photo-stability, and a significant Stokes shift. Consequently, advanced imaging techniques can be used to provide high-resolution images of LFPs, surpassing the capabilities of conventional approaches. One notable benefit lies in the heightened sensitivity exhibited by these conjugated polymeric materials towards specific chemical interactions. This characteristic enables them to selectively bind to various constituents of latent fingerprints, including lipids, proteins, and amino acids. Consequently, this leads to the production of more pronounced and well-defined fingerprints, even when applied to difficult surfaces such as plastic and metals. This technique facilitates the identification of an individual by visualising intricate features at 1–3 levels, such as the arrangement of pores and the shape of ridge ends. Moreover, their diminutive dimensions provide enhanced infiltration into permeable surfaces, hence enhancing the identification of concealed imprints on substrates such as paper or cardboard. Furthermore, the stability and resistance to photobleaching exhibited by these materials render them well-suited for extended periods of storage and subsequent investigation.

Table 1 Comparative analysis of conjugated polymers for LFP imaging

Publication	Material	Substrate	Ref
<i>Chem. Commun.</i> , 2009, 2112–2114	poly[1-phenyl-2-(p-trimethylsilyl) phenylacetylene] (PTMSDPA) membrane	steel, glass, plastic	[62]
<i>Macromol. Rapid Commun.</i> , 2013, 34, 102–108	PPV-NPs with superglue ethyl-2-cyanoacrylate ester	glass sheet, aluminium foil	[63]
<i>Nat. Commun.</i> , 2014, 5, 3736	polydiacetylenes ((PDAs)	PET films	[64]
<i>Chem. Commun.</i> , 2015, 51, 13634–13637	conjugated polyelectrolyte dots (CPEDs) and surfactants	glass slide	[65]
<i>ACS Appl. Mater. Interfaces</i> , 2015, 7, 14477–14484	oxetane-functionalized polymer dots (Ox-Pdots)	glass slide, aluminum foil, iron spoon, plastic	[60]
<i>Adv. Funct. Mater.</i> , 2016, 26, 498–506	polydiacetylenes ((PDAs)	paper	[56]
<i>ACS Appl. Mater. Interfaces</i> , 2016, 8, 24025–24029	sulfonated poly(diphenylacetylene) (SPDPA)	paper, glass, plastic	[66]
<i>ACS Appl. Mater. Interfaces</i> , 2016, 8, 6245–6251	magnetically active polydiacetylenes (MNP-PDAs)	steel, PET film, glass, paper	[67]
<i>Anal. Chem.</i> , 2016, 88, 11616–11623	polymer dots (Pdots) of PF-BT-DBT	paper, glass, aluminum foil, plastic bags, acrylic sheets	[61]
<i>ACS Appl. Mater. Interfaces</i> , 2017, 9, 37501–37508	AIEE-active polyfluorene based conjugated polyelectrolyte	aluminum foil, glass slide, coins, adhesive tape	[68]
<i>ACS Appl. Mater. Interfaces</i> , 2017, 9, 4908–4915	poly-(p-phenylenevinylene) (PPV) nanoparticles	adhesive tapes, aluminium foil and cover glass	[57]
<i>Opt. Mater.</i> , 2018, 86, 79–86	Pdots of p-phenylenediamine	stainless steel, adhesive tape, marble, CD painted wood	[69]
<i>J. Colloid Interface Sci.</i> , 2018, 528, 200–207	poly-(p-phenylenevinylene) (PPV) nanoparticles	aluminium foil and cover glass	[58]
<i>ACS Applied Materials & Interfaces</i> 2021, 13, 27419–27429	poly[p-(phenylene ethylene)-alt-(thienylene ethynylene)] (PPETE-NMe ³⁺)	Glass, coin aluminum foil, ceramic, plastic	[70]
<i>Journal of Colloid and Interface Science</i> 2023, 642, 658–668	poly (p-phenylene vinylene) branched poly-ethyleneimine nanoparticle (PPV-brPEI NPs)	aluminum foil, steel and ceramic	[59]
<i>ACS Applied Polymer Materials</i> 2024, 6, 5957–5969	Poly(p-(tetraphenylethene)-alt-(phenylene ethylene)) (PTPEPE-IDA)	glass, plastic aluminum, steel	[71]

Despite the first successful advancements, there is a need to improve the utilisation of CP-based applications in the field of forensic research, such as LFP imaging. Obtaining top-notch LFP development with level 3 capabilities and real-time virtual screening using the existing database continues to be a significant obstacle. In order to achieve improved contrast and selectivity, it is necessary to create such polymers that have an appropriate hydrophilic and hydrophilic contact with the fingerprint components. In addition, the incorporation of AIE moieties into the polymer backbone, in conjunction with other donor acceptor moieties, has been demonstrated to significantly improve the imaging quality, even in the aggregated form. This enhancement leads to a notable increase in contrast for the created layer-free polymers (LFPs). Designing near-infrared (NIR) emissive carbon dots (CPs) is a promising method for developing light-fast polymer (LFP) technology. This strategy enables fingerprint imaging with minimal light scattering, low interference absorption, and minimal autofluorescence, leading to high-resolution fingerprint images. Incorporating

narrow band gap moieties into the conjugated backbone enables the acquisition of NIR emissive CPs.

Declarations

Competing interests The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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