Pristine Graphic Carbon Nitride Quantum Dots for the Visualized Detection of Latent Fingerprints

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Abstract
User-friendly fingerprint powders, namely efficient, low-cost and nontoxic ones, are always desirable for the latent fingerprints (LFPs) developments. Here, we described the use of pristine graphic carbon nitride quantum dots (g-C$_3$N$_4$ QDs) as a new kind of user-friendly fingerprint powders. The g-C$_3$N$_4$ QDs could be facilely prepared from urea and sodium citrate precursors through the low temperature solid-phase reaction. Due to their good optical properties and selective interactions with secretion residuals, the g-C$_3$N$_4$ QDs powders were exploited to develop the LFPs on different substrates by the powder dusting technique. The LFPs images on plastic bag exhibited high ridges and furrows contrast ratio, where the level 1~3 details of LFPs were easily identified. This work indicates that the g-C$_3$N$_4$ QDs powders have good performance on LFPs visualizations and will find some applications for forensic investigations.

Keywords: Graphic carbon nitride quantum dots, Low temperature solid-phase reaction, Latent fingerprint, Visualized detection
Introduction

If one’s hand has been in contact with surface of a substrate, the transfer of a complex mixture (i.e. the natural skin secretions and exogenous residues on the finger) from the skin to the surface occurred and the fingerprint would be created simultaneously. The fingerprint pattern composes of the ridge and furrow impressions on the surface. Thus, fingerprints on the substrate not only reflect the characteristics of the papillary ridges and furrows of fingertips, but also provide additional information on human metabolism and some stuffs you have touched. These ridge and furrow characteristics on the fingerprint are unique to each individual and remain unchanged throughout the entire human lifetime. Undoubtedly, fingerprint has become one of the most powerful evidences in personal identification, which has been widely used for forensic investigations. In addition, the analysis of the complex mixture on the fingerprint is applicable in clinical diagnosis, drug abuse and explosive residue detection. Nonetheless, most of the fingerprints left on the substrate surface are invisible to naked eyes due to the poor optical contrast. These latent fingerprints (LFPs) need additional physical or chemical treatments to enable their visualization on the substrate. Since 19th century, a continuous effort has been made to develop LFPs for easier detections. In this regard, various developing techniques for the LFPs detection have been established based on physical, chemical and biological process, including the powder dusting, fluorescence staining, chemical fuming and biomolecule recognition. Among these techniques, powders dusting is convenient and widely used in the fingerprint detections on diverse substrate surfaces. In this technique, fingerprint powders play a key role that determine the quality of fingerprint pattern. Commercial fingerprint powders, such as metallic powders, magnetic powders, and luminescent
powders, have been used a lot in the detection of LFPs. But there are still some challenges that we might encounter in the powders dusting, for instance strong background interference, low optical contrast, poor selectivity and potential toxicity. Consequently, there is still of great interest to exploit a new fingerprint powder for developing high quality of LFPs.

The quantum dots (QDs) are another promising fingerprint powders that frequently studied in the past years. Generally, QDs can be defined as certain type of semiconductor materials with a nanoscale (2–20 nm) size in three dimensions formed by the group II–VI binary compounds. Owning to the extraordinary optical properties of QDs, including strong luminescence, high quantum yields, tunable emission wavelength and long luminescence lifetime, CdS, CdSe and CdTe QDs have been prepared for developing LFPs with high sensitivity and excellent contrast against the background. However, despite of their good performance in the LFPs development, the practical use of cadmium contained QDs has raised more and more concerns over the highly toxic cadmium ion. Even though QDs can be prepared by coating various materials to reduce the direct exposure of cadmium-contained QDs, further improve biological compatibility and selectivity, the complex synthesis process will raise the cost and does not entirely solve the toxicity problem. Recently, the graphic carbon nitride quantum dots (g-C₃N₄ QDs), as the subcategory of QDs, share very similar optical properties, thus, have attracted great attention in the applications of photovoltaic device, photo-catalysis, chemical/bio-sensing and bio-imaging. Besides, there are some other advantages over the traditional QDs, including the easy preparation, good chemical stability, excellent biocompatibility, low cost, and nontoxicity. These superiorities will allow addressing the aforementioned problems. Generally, the preparations of g-C₃N₄ QDs can be achieved in two routes: the top-down route and the
bottom-up route.\textsuperscript{20} The former route involves firstly the preparation of large-size bulk g-C\textsubscript{3}N\textsubscript{4} nanostructure, then following by breaking down and fragmentation of the g-C\textsubscript{3}N\textsubscript{4} precursor to uniform and smaller g-C\textsubscript{3}N\textsubscript{4} QDs. But this preparation route is usually performed under some harsh conditions, such as high temperature (>400 °C), use of strong acid, base or oxidant, intensive ultra-sonication and so on.\textsuperscript{25-28} In the bottom-up route, small molecule precursors composed of carbon precursor and nitrogen precursor undergo a series of chemical reactions, including condensation, decomposition and carbonization, to form the g-C\textsubscript{3}N\textsubscript{4} QDs.\textsuperscript{29-32}

Like other luminophores, the g-C\textsubscript{3}N\textsubscript{4} QDs may suffer from the aggregate caused quenching (ACQ) in the dry or aggregate states. Further modification or doping of g-C\textsubscript{3}N\textsubscript{4} structures has been often applied to avoid the ACQ effects on the g-C\textsubscript{3}N\textsubscript{4} QDs.\textsuperscript{22} Only the g-C\textsubscript{3}N\textsubscript{4} QDs powders with strong photoluminescence (PL) emission can be used in the LFPs development. The first study on the use of g-C\textsubscript{3}N\textsubscript{4} QDs as fingerprint powders was reported by Song et.al,\textsuperscript{33} in which g-C\textsubscript{3}N\textsubscript{4} doped with phenyl groups was prepared by the copolymerization of urea and trimesic acid at 500 °C. Fingerprint imaging was obtained after dusting the LFPs on a glass surface with phenyl doped g-C\textsubscript{3}N\textsubscript{4} powders. In another work of Prabakaran,\textsuperscript{34} the bulk g-C\textsubscript{3}N\textsubscript{4} precursors prepared from melamine were further treated with the ultra-sonication to produce porous g-C\textsubscript{3}N\textsubscript{4} QDs. Then the obtained porous g-C\textsubscript{3}N\textsubscript{4} powders were mixed with commercial silica gels to prepare the fingerprint powders. Indeed, the two reports have demonstrated the earliest studies of LFPs development using the g-C\textsubscript{3}N\textsubscript{4} materials. But the preparation of these g-C\textsubscript{3}N\textsubscript{4} fingerprint powders either requires relative high temperature or extra post-modification. Herein, we report the direct use of g-C\textsubscript{3}N\textsubscript{4} QDs for the LFPs development without any modifications or doping. The pristine g-C\textsubscript{3}N\textsubscript{4} QDs were facilely prepared from urea and sodium citrate small molecule precursors through the
low temperature solid-phase reaction, also known as the bottom-up route. The produced g-C$_3$N$_4$ QDs exhibit intense blue PL emission with the maximum emission wavelength at 452 nm. Furthermore, the g-C$_3$N$_4$ QDs powders were exploited to develop LFPs on aluminum foil, plastic bag and paper. High quality LFP image was obtained on the plastic bag substrate with high sensitivity and selectivity.

**Experimental**

*Chemicals and materials*

All chemicals used in this study were analytical grade and used as received without further purification. Urea and sodium citrate were purchased from Aladdin Reagent Co., Ltd (Shanghai, China). Sodium hydroxyl and anhydrous ethanol were purchased from Sinopharm Chemical Reagent Company (Shanghai, China). Dialysis tubes (molecular weight cut-off, 3500 Da) were bought from E-bioeasy Co. Ltd (Shanghai, China). UV lamp (multi-wavelengths excitation), forensic brush and commercial fluorescent magnetic powders were obtained from Beijing Huaxing Co. Ltd. Deionized water (DI) was used as solvent in the experiment.

*Synthesis of the g-C$_3$N$_4$ QDs*

The g-C$_3$N$_4$ QDs were prepared according to literature procedures with minor modifications.\textsuperscript{31} Briefly, 0.256 g (4.26 mmol) of urea and 0.148 g (5.03 mmol) of sodium citrate were mixed thoroughly by grinding in an agate mortar. The resultant mixtures were transferred into a 25 mL Teflon-lined stainless steel autoclave and heated in a drying oven at 180 °C for 1 h. The as-prepared light-yellow powders were dissolved in DI water, following by the purification of the solution against DI water in a
dialysis tube for 24 h. The purified solution was filtered with microporous membrane (0.22 μm) to remove the large particles. Finally, the g-C$_3$N$_4$ QDs powders were collected after the freeze-drying.

*Application of the g-C$_3$N$_4$ QDs in LFPs development*

All the fingerprint samples were prepared from the same volunteers. Firstly, the fingers of the volunteers were washed with soap and dried in air. Then, LFPs from the volunteers were collected by gently wiping their fingertips over forehead or nose region and pressing them on different substrates, including glass slide, aluminium foil, plastic and paper. The prepared g-C$_3$N$_4$ QDs powders were cautiously dusted onto the substrate surface. Excessive powders were removed upon a light brushing action with a forensic brush. Finally, LFPs on the substrate was imaged by illuminating under a UV lamp (tunable excitation wavelength of 320 ~ 400 nm). The images of LFPs were recorded using a commercial digital camera (Canon EOS 500D).

**Results and Discussion**

*Preparation and characterization of the g-C$_3$N$_4$ QDs*

The g-C$_3$N$_4$ QDs were prepared from urea and sodium citrate as the nitrogen and carbon sources through the low temperature solid-phase method. After mixing the precursors completely by grinding, urea would easily decompose to isocyanic acid and NH$_3$ upon the heating at 180 °C, resulting high pressure in autoclave. This could facilitate, in turn, the decomposition of sodium citrate.$^{35,36}$ As a result, the g-C$_3$N$_4$ QDs were formed by the chemical reaction among the solid reactants without any solvent in this method. In Fig.1a, transmission electron microscopy (TEM) image showed that the
as-prepared g-C₃N₄ QDs were monodisperse nanoparticles, which the size distribution ranges from 1 to 9 nm and the average diameter is about 5 nm (Fig. 1b). The insert in Fig. 1a is the high-resolution TEM image of a single g-C₃N₄ QDs with high crystallinity. The lattice spacing of g-C₃N₄ QDs is 0.34 nm in consistent with the (002) crystal plane of graphite phase carbon nitride.³⁷ Powder X-ray diffraction (XRD) analysis further demonstrated the lattice structure the g-C₃N₄ QDs. As shown in Fig.1c, the strongest sharp diffraction peak at 27.4° (002) could be assigned to the interlayer d-spacing of 0.34 nm between the planar carbon nitride sheets. A small diffraction peak at 13.1° (001) corresponds to (tri)-s-triazine unit of the g-C₃N₄.³⁵,³⁸ X-ray photoelectron spectroscopy (XPS) measurement was performed to study the elemental composition and valence distribution. In Fig. 1d, the peaks of C 1s, N 1s and O 1s were observed in the XPS spectrum at 285.0, 400.0 and 532.3 eV, in good agreement with that of the graphite carbon nitride in the previous report.³⁹ The high-resolution XPS spectra of C 1s (Fig. S1a) produces four peaks at 284.7 eV for sp² C-C, 286.6 eV for C-O, 288.1 eV for sp² N-C=N, and 289.3 eV for O-C=O.³¹ The N 1s XPS spectra (Fig. S1b) could be divided into three components at 398.8 eV, 399.6 eV and 400.4 eV, which are corresponding to C=N-C, C-N-C and N-(C)₃.⁴⁰ The O 1s XPS spectra (Fig. S1c) confirmed the oxygen-containing groups that the peaks at 530.9 eV and 531.6 eV could be assigned to C=O and C-O-C/C-OH, respectively.⁴¹ The mass percents of the three elements were calculated to be 53.4%, 16.8%, and 29.8%. Accordingly, the atomic ratio of C:N:O is 3.8:1:1.6 for the g-C₃N₄ QDs. The functional groups of the g-C₃N₄ QDs were investigated by Fourier transform infrared spectroscopy (FTIR) as presented in Fig. S2. Three strong bands center at 1406, 1590 and 3422 cm⁻¹ were attributed to the stretching of C≡N in heterocycles, the asymmetric stretching of C=O in the carboxylate groups and the stretching of N-H or O-H, respectively.³¹,⁴²,⁴³ Another two small peaks at
606 and 747 cm\(^{-1}\) were due to out of plane bending vibrations and deformation vibrations of (tri)-s-triazine units.\(^{34,44}\)

**Optical properties of the g-C\(_3\)N\(_4\) QDs**

The light absorption and emission properties of the g-C\(_3\)N\(_4\) QDs were studied by the UV-vis and PL spectroscopy. In Fig. 2a, the UV-vis spectra demonstrated the absorption increased rapidly from 520 nm to 250 nm with formations of two characteristic shoulder at 300 and 379 nm.\(^{37}\) The absorption peak at 300 nm could be considered π–π* transition of the aromatic ring, while the other one at 379 nm was related to n–π* transition of the C=N bond.\(^{45}\) The broad absorption range indicates that the PL emission of g-C\(_3\)N\(_4\) QDs can be triggered by multiple excitation wavelengths. It was shown in Fig S3 that the maximum emission wavelength remained unchanged with different excitation wavelengths. The strongest emission at 452 nm of was obtained upon excitation at 320 nm in the PL spectra. Moreover, light yellow color of g-C\(_3\)N\(_4\) QDs powders was seen in under white light (Fig. 2b), while bright blue color was observed under a 365 nm UV lamp (Fig. 2c). These results have confirmed that g-C\(_3\)N\(_4\) QDs exhibit good optical properties, making them a potential candidate for LFPs developments.

**Application of the g-C\(_3\)N\(_4\) QDs to LFPs developments**

To explore the feasibility of the g-C\(_3\)N\(_4\) QDs for LFPs developments, different substrates were used to prepared LFPs samples. Generally, the secretions left on LFPs refer to the trace amounts of sweat (i.e. inorganic salts, water, amino acids, urea, proteins, and glucose) and sebum (i.e. fatty acids, triglycerides, wax esters, squalene, and cholesterol) from skin glands.\(^{46}\) The g-C\(_3\)N\(_4\) QDs consisting of diverse of functional
groups, such as amine, hydroxyl and carboxyl, offering the good binding sites toward the targets and form easily negatively charged states upon the deprotonation. The negatively charged property of the fingerprint powders could make them more stable for the storage and induce the electrostatic interaction, which was found to be beneficial for the LFPs developments.\textsuperscript{47,48} As shown in Fig S4, the zeta potential of the g-C\textsubscript{3}N\textsubscript{4} QDs in water was measured to be -26.7 mV, while the zeta potential for the mixture of the g-C\textsubscript{3}N\textsubscript{4} QDs and the skin secretions increased to -4.41 mV. Such zeta potential change verified the strong electrostatic interactions between the g-C\textsubscript{3}N\textsubscript{4} QDs and the skin secretions. When dusting these g-C\textsubscript{3}N\textsubscript{4} QDs powders on LFPs samples, immobilization of g-C\textsubscript{3}N\textsubscript{4} QDs powders will take place on the papillary ridges due to the physical absorption between g-C\textsubscript{3}N\textsubscript{4} QDs and the sebum compounds, the chemical binding with metal ions, urea or amine acids and the electrostatic interactions with the positively charged species in the sweat residues. The LFPs images were developed by the g-C\textsubscript{3}N\textsubscript{4} QDs powders on glass slide, aluminum foil, plastic bag and paper. As shown in Fig. 3, the LFPs images were captured by a digital camera after the UV lamp illumination with the set excitation wavelength. The PL emission color of g-C\textsubscript{3}N\textsubscript{4} QDs differs on these substrates, which might be related to the texture, color, and light reflection difference in each substrate.\textsuperscript{49} These images depicted the whole bright papillary ridge patterns of LFPs on the aluminum, plastic and paper substrates. These results suggest that the g-C\textsubscript{3}N\textsubscript{4} QDs can used to develop LFPs on both non- and semi-porous substrates. However, a low optical contrast LFPs image was displayed on the glass slide, where the serious background interference, weak PL intensity and bad ridge continuity were recorded.

As for the forensic investigations, level 1 details about the core and delta can be easily identified from the papillary ridge pattern of LFPs, which is useful for the
classification and indexing of LFPs. But these macro details are not distinctive enough for a personal identification. There are still many the minutiae points and pore details in the LFPs pattern, including level 2 (ridges endings, enclosure, bifurcations and hook) and level 3 (sweat pore, island, scar and line shape) types. Because these minutiae details are stable, unique and informative, so further discriminating them is more accurate and convincing for the LFPs detection. Obviously, the LFPs image on plastic bag substrate exhibited higher resolved ridge pattern than those on the other substrates. It did provide more identified details of LFPs as shown in Fig.4. The representative details of level 1-3, such as double loop whorl, bifurcation, enclosure, termination, scar and sweat pore, could be found in the LFPs pattern. The intensity profiles of the LFPs image were also measured by Image J software to study the contrast ratio between the ridges and furrows. In Fig. 5, the immobilized g-C₃N₄ QDs powders produced a large variation of PL intensities along the arrow line on the LFPs images with an average contrast ratio of 1.7, revealing the selective interactions between the secretion compounds and g-C₃N₄ QDs powders. The LFPs image on the aluminum foil could provide some level 1and 2 details for identification, and had a better contrast ratio up to 2.3, but with bad ridge continuity around LFPs edge (Fig. S5a and S5c). In addition, the LFPs image the on the paper substrate showed poor ridges and furrows contrast (the average contrast ratio is 1.1), making LFPs details difficult to fully identify (Fig. S5b and S5c). This could be attributed to the absorption of a small amount of g-C₃N₄ QDs powders in the furrows by the semi-porous paper surface. Therefore, the LFPs image developed on the plastic bag was selected for further investigations.

The LFPs image developed by powder dusting technique highly depends on the interactions between the secretion residues and the fingerprint powders. The stability of either the secretion residues or the fingerprint powders influence the visualization of
LFPs. On one hand, the amount of the secretion residues on LFPs will gradually decrease with time. This can weaken the selective interactions with the fingerprint powders. On the other hand, low PL intensity may be caused by the possible decomposition and contamination of the fingerprint powders after long term storage. All these problems could increase the difficulty for the visualization of an old LFPs. For that reason, we have employed the g-C₃N₄ QDs powders for the visualization of LFPs aged for different times. As shown in Fig. S6a, the best LFPs image was obtained by the powder dusting on the fresh fingerprint. Even for those LFPs at age of 5 days and 30 days (Fig. S6b-c), the LFPs could be still visualized with clear ridge details. Besides, the g-C₃N₄ QDs powders stored in a vial for 6 months without any special protection were used for LFPs visualizations. It was found that the LPFs image exhibited the poor ridges and furrows contrast in the core zone, where the level 1 details of the core could not be identified (Fig. S7). But the level 2 details out of the core zone were still observed in ridge pattern, such as bifurcation, enclosure, termination and hook. These results demonstrated that the g-C₃N₄ QDs powders were robust and stable enough for the visualization of LFPs on plastic bag.

Comparison with the commercial fluorescent magnetic powders

The commercial fluorescent magnetic powders were used to develop LFPs on plastic bag as a comparison. In order to better compare the performance of the two fingerprint powders, we developed two LFPs using the same finger and prepared the LFPs according to the following procedures: 1) wash thoroughly the finger with soap; 2) dry the finger in air; 3) wipe the finger over forehead to collect the skin secretions; 4) press the finger on the plastic bag. One of LFPs was developed by dusting g-C₃N₄ QDs powders, while the other one was treated by dusting the commercial fluorescent
magnetic powders. After the visualization upon the lumination of a UV lamp, both digital images of LFPs were recorded and cut into two halves. One half of each image was combined to a matched LFPs image and compared. The comparison of LFPs images in Fig. 6 showed both the g-C$_3$N$_4$ QDs powders and commercial fluorescent magnetic powders worked well and gave good ridges pattern. However, the g-C$_3$N$_4$ QDs powders could display a better ridges and furrows contrast than the commercial fluorescent magnetic powders did. It could be seen clearly that some commercial fluorescent magnetic powders were co-existed in both ridges and furrows in the right half image, leading to unclear ridges details. Indeed, because these commercial fluorescent magnetic powders are prepared with magnetic powder, alumina powder, fluorescent dye and graphite powder, the physical absorption can be the main interaction between the inorganic components and secretion residuals in the LFPs development, which make the immobilization of commercial fluorescent magnetic powders on LFPs less selective. Thus, the g-C$_3$N$_4$ QDs powders offered the better performance in LFPs development due to their strong interactions with the secretion residuals on the ridges.

**Conclusions**

In summary, we have demonstrated that the pristine g-C$_3$N$_4$ QDs, facilely prepared from urea and sodium citrate through the low temperature solid-phase reaction, could be used as the fingerprint powders. The g-C$_3$N$_4$ QDs powders showed good PL emission under the excitation with UV light and reserved diverse functional groups. The g-C$_3$N$_4$ QDs powders were applied to the LFPs development on different substrates. The LFPs image developed on plastic bag exhibited good sensitivity and selectivity, in which the level 1–3 details of LFPs were found. Furthermore, the g-C$_3$N$_4$ QDs powders were robust and
stable for the interactions with the secretion residuals, making it more competent for LFPs development than the commercial green fluorescence powders. Finally, it was noteworthy that both urea and sodium citrate precursors were low-cost and abundance materials and no sophisticated method was involved in the preparation process. All these characteristic of the g-C₃N₄ QDs powders highlight their potential use in forensic investigations.

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Figure Captions

Scheme 1  a) The preparation process of the g-C₃N₄ QDs, b) The illustration of procedures for the visualized detection of LFPs.
Fig. 1 (a) TEM image, (b) QD size distribution, (c) XRD and (d) XPS spectra of the g-C$_3$N$_4$ QDs.

Fig. 2 a) The UV-vis and fluorescent spectra of the g-C$_3$N$_4$ QDs dispersion solution, the photographs of the g-C$_3$N$_4$ QDs powders under b) the white light and c) a 365 nm UV lamp.

Fig. 3 LFPs images developed with g-C$_3$N$_4$ QDs powders on different substrates: a) glass slide, a) aluminum foil, c) plastic bag, d) paper upon excitation at 365 nm using a UV lamp.
Fig. 4 The LFPs image on the plastic bag with level 1-3 details including whorl, bifurcation, enclosure, termination, scar and sweat pore.

Fig. 5 A representative PL intensity profile of the ridges and forrows along the arrow line in the LFPs image of Fig. 4.
Fig. 6 Comparison of the LFPs images on plastic bag developed by the g-C$_3$N$_4$ QDs powders (left half) and the commercial fluorescent magnetic powders (right half) under the illumination of a UV lamp.