Optimum conditions and application of one-step fluorescent cyanoacrylate fuming method for fingermark development based on PolyCyano UV

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ABSTRACT

Cyanoacrylate fluorescent staining has become a common method for developing latent fingermarks on light-coloured or white objects. The method requires two steps and skilled operation, and the handling process has the risk of damaging the ridge details or even biological samples. To take full advantage of the high-sensitivity fluorescence, recent research efforts on fluorescent cyanoacrylate have aimed to avoid rinsing after staining, add fluorescence to the cyanoacrylate, increase the fluorescence intensity and broaden the fluorescence spectral range. PolyCyano UV is a novel product that can be used to overcome the disadvantages associated with progressing to one-step fuming to directly develop fingermarks. To explore the optimal development conditions and application effects of PolyCyano UV, thermogravimetric differential thermal analysis, fluorescence spectroscopy and control variable analysis are used to determine and analyse the best conditions for using the reagent, including temperature, fluorescence excitation band, relative humidity and fuming concentration. The temperature range of the one-step fuming development method is 212.14 °C–275.16 °C, the wavelength range of the excitation light source is 235–580 nm, the relative humidity is 60%–80%, the concentration of the fume is 1–3 g/m3 and the specific value is related to the surface properties of the object and the aged time of the fingermarks. Additionally, fluorescence spectroscopy shows that the excitation wavelength range of the light source is 235–580 nm. Based on the best fuming condition, many common objects were selected to proceed to the application experiments, which allowed for a comparison with the traditional visualisation method, namely the Cyanobloom reagent + dyeing two-step development method. The comparison showed that latent fingermarks on most non-porous smooth surfaces, especially on light-coloured or white objects, were developed successfully. In addition, a comparative study with the conventional cyanoacrylate glue–fluorescent staining experiments showed that the effect of dyeing afterwards with Rhodamine 6 G is better than the one-step method. The effect of dyeing with BBD is basically the same as that of the one-step method.

KEY POINTS:
• One-step fluorescent cyanoacrylate fuming can be a promising alternative for fingermarks developing based on PolyCyano UV in forensic science.
• The optimum application conditions of PolyCyano UV will provide guidance for fingermark examiners.
• The one-step fuming method based on PolyCyano UV has many advantages over the secondary dyeing method.

Introduction

The cyanoacrylate fuming method is commonly used to develop latent fingermarks on the surface of non-porous smooth objects. At present, the basic principle of cyanoacrylate to develop fingermarks is defined as follows: after cyanoacrylate is volatilised, the water and amino acids (mainly -OH) in the fingermark residues will undergo anionic polymerisation, and then a kind of polymer will form on the ridge residues to develop the fingermark in white [1]. However, when fingermarks are detected on the white or milky background object surface at a crime scene, the white ridges of the fingermarks developed by this method have very weak contrast with the white background, which means that it is not easy to observe them and take photos for the record. To solve this problem, the commonly used method is fluorescent
dyeing, which enhances the white lines of the fingermarks obtained by fuming.

In recent years, forensic scientists have systematically summarised the most commonly used fluorescent dyes in various countries around the world, such as Ardrox, Basic Yellow 40, MBD, Rhodamine 6G and RAM [2]. Ardrox is a fluorescent liquid dye, which works well with lasers, forensic light sources and UV lamps [3]. Basic Yellow 40 is an excellent dye stain for cyanoacrylate, and it fluoresces under blue light [4]; MBD is fluorescent when exposed to a forensic light source between 415 nm and 535 nm, which makes it ideal for use with a wide variety of forensic light sources; and Rhodamine 6G produces fluorescence with the use of lasers and forensic light sources [5]. RAM is a combination of Rhodamine 6G, Ardrox and MBD [6]. The secondary fluorescence enhancement also has the following shortcomings: (i) the use of fluorescent dyes usually introduces organic solvents, which can easily cause partial dissolution or damage of the cyanoacrylate polymer, and then damage the ridges and their minutiae; (ii) the use of some organic solvents will change the nature of the object surface, and they will destroy biological samples such as exfoliated cells; (iii) some staining methods require rinsing and the degree of rinsing has a serious impact on the fluorescence effect, so there is a certain risk associated with rinsing, where improper rinsing will easily rinse fingermarks off; (iv) the dyeing process is a secondary treatment, which is time-consuming and cumbersome, and is not easy to popularise in practical application; (v) fluorescent dyes are usually toxic and improper handling, which refers to directly touching the dye, can be detrimental to health [7].

To overcome the shortcomings of secondary fluorescent dyeing, Weaver DE and Clary EJ at the Scientific Crime Laboratory in Alaska, USA, conducted research on the one-step fluorescent cyanoacrylate fingermark development method, and believed that the magenta styrene fluorescent dye would polymerise with cyanoacrylates after heating [8]. Under the excitation of an argon-ion laser, through an orange-red filter, pink fluorescent fingermarks could be observed [8]. A fluorescent cyanoacrylate copolymer has been successfully developed, which is called CN Yellow [9]. Subsequently, Weaver et al. [10] conducted a study on the excitation spectrum range of CN Yellow and expanded its excitation range to 530 nm, but there has been no successful application case. Sari et al. [11] did a lot of research on cyanoacrylate reagents to show latent fingermarks. It took 4 years to change the structure of cyanoacrylate reagents and introduce functional groups that could form fluorescent polymers with residues of latent fingermarks, but so far, no progress has been made.

Hahn and Ramotowski [12] from the US have briefly introduced a new type of fluorescent cyanoacrylate powder that can display latent fingermarks in one step. They named this type of cyanoacrylate UV powder PolyCyano UV powder, but it has not been subjected to in-depth application research and the optimal development conditions have not been determined. Chadwick et al. [13, 14] conducted a preliminary study and evaluation on the process and fuming conditions of PolyCyano UV powder. However, our research group believes that further research is needed. PolyCyano UV powder is a new type of latent fingerprint fuming reagent in powder form, which integrates cyanoacrylates and fluorescent dyes, and was launched by British company Foster + Freeman [15, 16]. Farrugia et al. [17] conducted an evaluation of the Lumicyano™ cyanoacrylate fuming process for fingermarks developing on plastic carrier bags, and the result showed that the Lumicyano™ appears to be suitable for this task and can help to save lab space and time as it does not require dyeing or drying procedures, and it is therefore same as PolyCyano with the same characteristic as one-step fuming. In addition, the FBI Laboratory developed a new product called Cyano-Shot™, and tested the efficacy of using Lumicyano™ products in conjunction with Cyano-Shot™ products. This product consists of a cyanoacrylate solution, an activator fluid and a canister with a wick containing activator crystals. When the Cyano-Shot™ activation components are combined, an acid-base reaction occurs that generates heat and leads to vapourisation of the Cyano-Shot™ solution, and then the fingermarks can be developed under forensic light sources without RAM application [18].

Our research group had conducted application research on the development method with PolyCyano UV and the effect of the reagent according to the experimental operation methods we have mastered [19, 20]. On this basis, we further used relevant instruments and equipment to investigate and summarise the application conditions, to obtain the best visualisation scheme, with the aim to analyse the properties of PolyCyano UV reagents and optimise its application conditions. This research aims to promote a one-step fluorescent fingerprint development method that can quickly and systematically increase the display rate of fingermarks in cases, enrich the development technology of fingermarks, and play a better role in criminal technology for investigating and solving cases, prosecution and trial.

Materials and methods

Materials and equipment

The equipment used was a Q600 SDT thermogravimetric-differential scanning calorimetry
integrated thermal analyser (TA Company, New Castle, DE, USA); SolidSpec-3700 ultraviolet-visible near-infrared spectrophotometer (Shimadzu Corporation, Kyoto, Japan); Fluorolog\textsuperscript{\textcircled{R}}-Tau-3 fluorescence spectrometer (Jobin Yvon Company, Longjumeau, France); MVC3000 handprint fuming cabinet, DCS-4 handprint photographing and inspection system (Foster + Freeman Company, Evesham, UK).

The chemicals used were PolyCyano UV (powder, 95%); Cyanobloom (colloid, 95%, Foster + Freeman Company, Shanghai, China); Rhodamine 6 G (98%); BBD (98.53%, MCE Company); anhydrous ethanol; acetone (analytical grade, Beijing Chemical Plant, Beijing, China); and all water was deionised water.

Preparing experimental samples

A transparent plastic bag, a white ceramic plate, a transparent glass and a metal can were selected as experimental objects since fingermarks can be commonly found on these objects in crime scenes. Nine volunteers (six males, three females) were chosen to prepare the samples. To ensure the reproducibility of the research programme in the laboratory, the volunteers were asked to wash and dry their hands first, and then wore PVC gloves for 5 min before each imprinting to ensure sufficient sweat. Each volunteer chose a finger to press continuously and successively on an object until the amount of sweat was extremely weak. Fingermarks were directly printed and aged for 1, 3, 7, 15 and 30 d.

Preparation of fluorescent dyeing liquor

Preparation of Rhodamine 6 G dyeing liquor: 0.1 g of Rhodamine 6 G powder was weighed and dissolved in 200 mL of absolute ethanol with stirring using a glass rod at room temperature until it was completely dissolved. This resulted in a 0.5 g/L Rhodamine 6 G dyeing liquor.

Preparation of BBD dyeing liquor: 0.5 g of BBD solid was weighed and dissolved in 100 mL of acetone at room temperature and stirred until it was completely dissolved, which resulted in a 5 g/L BBD stock solution. The stock solution was stored in a refrigerator. Then 10 mL of the BBD stock solution was taken and mixed with 90 mL of absolute ethanol mixing well, resulting in a 0.5 g/L BBD dyeing solution.

Measurement of the best fuming temperature

To accurately determine the temperature required for the experiment, thermogravimetric and differential scan calorimetry analyses were performed on the PolyCyano UV powder. After starting the instrument and computer, the parameters of the analyser were set according to the needs of the experiment: measuring temperature range, 0°C–600°C; heating speed, 10°C/min; and atmosphere, static air. Then the heating was started, stopped after the experiment and recorded after the temperature curve was stabilised.

Determination of the best excitation light source

It is necessary to measure the excitation and emission spectra so that the best excitation light source and filter could be selected to observe and record the fluorescence effect after the fingermark was displayed, which will reveal the fluorescence performance of PolyCyano UV. The specific operations were as follows: (i) obtain the UV-visible absorption spectrum by pre-scanning the fluorescent powder to find the position of its maximum absorption wavelength; (ii) screen the excitation wavelength from the vicinity of strong absorption by first scanning the emission spectrum to find the emission peak position, and then monitor the emission peak to obtain the excitation spectrum; (iii) determine the best excitation wavelength from the excitation spectrum, and then use the best excitation wavelength to excite the dye.

Selection of the best relative humidity and concentration

The experimental method used was the controlled variable method, and the research was carried out based on the optimal fuming temperature determined by a thermogravimetric and differential scan calorimetry analyser. The temperature was 230°C, with a powder dosage of 0.6 g and the fumigation time of 15 min. The relative humidity value of the MVC3000 fuming cabinet was set at 50%, 60%, 65%, 70%, 75%, 80%, 85%, and 90% for fuming in stages and the visualisation effect was compared under different conditions. The humidity value was selected by the corresponding optimal relative humidity value of 60%, 75% and 80% according to the time left by the fingerprint, and the fuming time was set to 15 min. Powders with concentrations of 0.5 g/m\textsuperscript{3}, 1 g/m\textsuperscript{3}, 2 g/m\textsuperscript{3}, 3 g/m\textsuperscript{3}, and 4 g/m\textsuperscript{3} were chosen to carry out the fuming experiment, and comparative analysis was made. The volume of the MVC3000 fuming cabinet used was 0.7 m\textsuperscript{3}.

Comparison with Cyanobloom + dyeing two-step development method

This section mainly focuses on comparison of the PolyCyano UV one-step fluorescence fuming and
Cyanobloom reagent + dyeing two-step fingermark visualisation effect. First, the prepared fingermark samples were divided into two halves evenly to ensure that the residues of fingermarks were equally distributed between the two halves. Second, the two methods were carried out. The PolyCyano UV one-step fluorescent fuming method was performed according to the best development conditions obtained from the above research. The MVC3000 fingermark fuming cabinet was used directly to set the relevant parameters for fingermark visualisation. The Cyanobloom reagent + dyeing two-step development method was conducted in the MVC3000 fuming cabinet first, then Rhodamine 6 G was used as the dye, to observe and photograph under 530-nm green light. Blue light of 450 nm was used to observe and take pictures after BBD dyeing. Finally, a comparative analysis was made based on the experimental effects of the two methods.

**Results and discussion**

**Measurement of the best fuming temperature**

According to the experimental data recorded by the thermogravimetric and differential scan calorimetry analyser, the graph in Figure 1 was obtained after processing.

As shown in Figure 1, during the endothermic process of the powder transforming from solid to gas, the sample only had one peak in the measured temperature range, which indicated that the powder state only underwent one transition, and the transition temperature was 212.14 °C. When the temperature was approximately 160 °C, sublimation began and the mass proportion gradually decreased. At 275.16 °C, the mass of the sample decreased by 98.97% and was almost completely sublimed into a gas. Therefore, to ensure the full progress of the fingermark fuming process, the proper fuming temperature should be selected within the range of 212.14 °C–275.16 °C.

**Determination of the best excitation light source**

As shown in Figure 2, the strongest absorption peak of the PolyCyano UV powder was measured by the ultraviolet-visible near-infrared spectrophotometer at around 360 nm. After obtaining the experimental data, the Origin software (V8.0725, OriginPro 8, OriginLab Corporation, Northampton, MA, USA) was also used to draw the spectrum.

According to the above experimental method, the strong absorption wavelength at 360 nm was selected as the best excitation wavelength to find the position of the emission peak. There were two emission peaks found, namely at 405 nm and 480 nm. Two excitation spectra were obtained using these two peaks as the excitation monitoring wavelengths. The monitoring wavelength range was adjusted again according to the incomplete curve shape of the obtained excitation spectra. When the wavelengths of 550 nm and 600 nm were scanned, the curve shape of the excitation spectrum was complete. The peak was obvious, and there was almost no absorption when the wavelength was greater than 600 nm. So the excitation spectrum and emission spectrum were obtained, as shown in Figure 3.

According to the spectrogram, the powder had a wide excitation spectrum range, which was in the...
broad wavelength range of 235–580 nm. When the wavelength of the excitation light was near 530 nm, the fluorescence intensity was the highest, and small emission peaks near the wavelengths of 470 nm and 365 nm were also observed. The sample could be excited in the range of ultraviolet, violet, blue, yellow, and green light after fuming. The Stokes shift was large, and the shift value was 45 nm. The fluorescence intensity was high and the interference of background fluorescence could be eliminated. It is also convenient to select a filter, so that a fingerprint with good fluorescence effect can be obtained.

Selection of the best relative humidity and concentration

For most non-porous objects, the optimal relative humidity when using PolyCyano UV for fuming varied with the aged time of the fingerprint. We carried out a grading evaluation of the effect of fingerprint developing, and the specific evaluation standard definitions are shown in Table 1.

As shown in Figure 4, for fingerprints aged for 1 d and 3 d, the developing effect was the best when the relative humidity reached 60%. The best effect for fingerprints aged for 7 d was at 70%. For fingerprints aged for 15 d, the best effect was at 80%. For fingerprints aged for 30 d, when the relative humidity was 85%, the developing effect was the best.

When fuming, the temperature and the humidity of the environment, water absorption of the sample material, and the aged time of the fingerprints should be fully considered. The best relative humidity should be selected according to the actual situation. The relative humidity of the fuming should not be too large, and water condensation in the fuming cabinet should generally be avoided since even a small change in the ambient temperature in the fuming cabinet may be enough to cause condensation of water vapour on the surface area with a lower temperature. The condensation will not only hinder the relative humidity to be reached during the humidification cycle, but most importantly, it will affect the quality of the development of fingerprints.

Under the same conditions of temperature, humidity and fuming time, the requirements of the PolyCyano UV powder concentration of the latent fingerprints with different aged times were also different. As shown in Figure 5, when the aged time of the fingerprints were 1 d, 3 d and 7 d, and the concentration of the PolyCyano UV powder was 1 g/m³, the fuming effect of the latent fingerprints

| Score | Definition of evaluation criteria                                                                 |
|-------|---------------------------------------------------------------------------------------------------|
| 4     | Fingerprint was fully developed and showed more than four-fifths of continuous ridge lines, the ridge detail disclosed being sufficient for identification. |
| 3     | Fingerprint was fully developed and showed more than two-thirds of continuous ridge lines, the ridge detail disclosed being sufficient for identification. |
| 2     | Fingerprint was partially developed and showed more than one-third of continuous ridge lines, the ridge detail disclosed being just sufficient for identification. |
| 1     | Fingerprint was partially developed, and a general imprint could be found, but it showed no obvious ridge detail or less than one-third of continuous ridge lines. |
| 0     | Fingerprint had no development.                                                                   |
on the surface of most objects was developed the best. When the aged time was 15 d and the powder concentration was 2 g/m³, the effect was the best. The effect was the best when the aged time was 30 d and the powder concentration was 3 g/m³.

For latent fingermarks with different aged times on different objects, when the concentration reached the optimal value, the effect decreased as the powder concentration increased. Therefore, the principle of powder dosage was the same as that of ordinary superglue or cyanoacrylate reagent. The principle of using it was that less was better than more, with a small amount added at a time. According to the appearance of latent fingermarks on the surface of different objects, the concentration of fuming is not only related to the aged time of the fingermarks, but also closely relates to the properties of the surface of the object. If the properties of the surface of the object itself have a strong ability to absorb product after PolyCyano UV powder sublimation, the required fuming concentration will correspondingly be lower.

**Comparison analysis of the effect of the two-step development method with Cyanobloom + dyeing**

**Comparison of the effect of using white light directly after fuming**

Fingermarks were observed and pictures were taken directly under white light or sunlight when the fuming was complete. Experiments showed that the
two fuming methods with ethyl cyanoacrylate as the main component had a good developing effect on the fingermarks left on the surface of non-porous objects, and the visualisation effect of the two methods was basically the same, with only small differences. As shown in Figure 6, the PolyCyano UV powder had no advantage on the latent fingermarks after fuming directly. In addition, the PolyCyano UV powder had a good effect for fingermarks on the surface of dark, non-porous objects, but the direct visualisation effect of the latents on the surface of light-coloured, white or complex objects with complex background colours was not obvious. At this point, it was the same as Cyanobloom fumed.

**Figure 5.** Fingermark developing effect under different concentration conditions of different aged times for four different objects (A. transparent plastic bag; B. white ceramic plate; C. transparent glass; D. metal cans). The horizontal axis represents the fingermark aged time, the vertical axis represents the fingerprint appearance effect score, and the different colours of bars represent different concentrations of fuming.

**Comparison of effects of one-step fumigation and secondary dyeing**

The PolyCyano UV reagent can be used in a one-step fluorescence fuming method, which can develop fingermarks on the surface of non-permeable smooth objects. Direct excitation of a light source with a wavelength of 450 nm can produce a good fluorescence effect. To directly develop the fingermarks on the surface of the object in light-coloured, white, transparent and complex background, the fluorescence intensity produced was generally slightly weaker than the intensity of Rhodamine 6 G staining and BBD. However, there were some differences in the developing effects of the two methods of one-step development and secondary dyeing owing
to the difference in the aged time of the latent fingermarks on the surface. As shown in Figure 7, the effect of the secondary dyeing with Rhodamine 6 G was better. As shown in Figure 8, the effect of dyeing with BBD was basically the same as that of the one-step fuming.

Conclusions

Through experimental exploration and preliminary research on the four aspects of the fuming temperature, fluorescence band, relative humidity and concentration of PolyCyano UV, it is shown that the best temperature range for powder fumigation is within the range of 212.14 °C–275.16 °C. The best fluorescence excitation light source are wavelengths of 360 nm, 450 nm and 530 nm, the best relative humidity is 60%–80% and the best fuming concentration is 1–3 g/m³. The specific values are related to the surface properties of the object and the aged time of the fingermarks.

PolyCyano UV has a good developing effect of the latent fingermarks on the surface of most non-porous smooth objects and fingermarks with different aged time, especially for the latent fingermarks on objects with light colours, white, transparent and complex background colours. Although there are some differences in the developing effect of the two methods owing to the difference of aged time of the latent fingermarks on the surface, in general, the effect of the secondary dyeing with Rhodamine 6 G is better. For one-step fuming, the effect of dyeing with BBD is basically the same.

The use of the powder can be used as a common method to show latent fingermarks on the surface of non-porous smooth objects, especially suitable

![Figure 6](image1.png)

Figure 6. Comparison of the fuming effect of PolyCyano UV (left) and Cyanobloom (right) on different objects with white light (A. metal cans; B. transparent plastic bag; C. yellow tap; D. transparent glass). The fuming temperature of the PolyCyano UV is 230 °C, with 80% humidity and concentration of 1 g/m³, and the fuming time is 15 min; the fuming temperature of the Cyanobloom is 120 °C, the humidity is 80% and the concentration is 1 g/m³, and the fuming time is 20 min. The aged time of fingermarks is 15 d.

![Figure 7](image2.png)

Figure 7. Comparison of the one-step fuming method with Polycyano UV and two-step method with Cyanobloom reagent + Rhodamine 6 G. Four left halves are obtained from one-step fuming from four different objects (A. transparent plastic bag; B. white plastic bag; C. transparent glass; D. metal can) and right halves are from the two-step method. The fuming temperature of the Polycyano UV is 230 °C, with 80% humidity and concentration of 1 g/m³; the fuming time is 15 min, and the excitation light source is 450-nm blue light. The fuming temperature of Cyanobloom is 120 °C, the humidity is 80%, the concentration is 1 g/m³, the fuming time is 20 min, and the excitation light source is 530-nm green light. The aged time of fingermarks is 15 d.
for developing latent fingerprints on light-coloured, white, transparent and complex background colours of a non-porous or semi-permeable object surface. The developing effect is slightly weaker than the secondary dyeing, but more importantly, the use of the powder for one-step fuming does not introduce organic solvents and will not cause the dissolution or damage of the cyanoacrylate polymer, which will ensure the completeness of the ridges and minutiae. It also avoids the possibility that the fingerprint ridges will be washed off arising from improper rinsing after dyeing. This method is therefore suitable for popularisation.

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**Authors’ contributions**

Kang Li designed and was responsible for this research. Kang Li, Shuo Li and Jun Yang conducted the experiment and prepared the preliminary data. Kang Li and Shuo Li analysed the data. Kang Li wrote the draft manuscript. Kang Li reviewed and revised the manuscript. All authors read and approved the final manuscript.

**Compliance with ethical standards**

The study was approved by the Institutional Review Board of Zhejiang Police College and the Graduate School of People’s Public Security University of China for use of human subjects and followed their policies. Written informed consent was obtained from all individual participants included in the study.

**Disclosure statement**

No potential conflict of interest was reported by the authors.

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