



Rapid Communication

NIR–NIR fluorescence: A new genre of fingerprint visualisation techniques

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ABSTRACT

A preliminary study reveals that finely divided cuprorivaite powder may be used to efficiently develop and subsequently image latent fingerprints across a range of highly patterned, coloured non-porous and semi-porous substrates using near infrared illumination and imaging. Problematic multi-coloured backgrounds provide very little interference under the illumination conditions used, and invoked fluorescence observed, when using this material. This is the first reported example of a NIR–NIR fluorophore for use within latent fingerprint visualisation and offers the potential for application at the scene and in the laboratory.

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1. Introduction

Forensic fingerprint powders remain a crucial and effective tool in the development of latent fingerprints across a range of substrate types. Despite being one of the oldest and most widely used fingerprint visualisation techniques, [1–3] fingerprint dusting powders offer significant versatility in terms of their colour, method of application and ability to, for the most part, not interfere with subsequent fingerprint processing using chemical-based reagents. Fingerprint powders are exceptionally useful in their capacity to be deployed rapidly both at the crime scene and in the laboratory, whilst taking up very little space in the practitioners' arsenal. The art of powdering surfaces to develop latent fingerprints has changed little over recent years. Real advances have come in the form of their incorporation within magnetic matrices, fluorescence characteristics designed to allow greater contrast between fingerprint and background and the development of more 'specific' reagents which may have some sort of affinity for a particular constituent within the latent residue. [4–6]

The most important consideration when developing latent fingerprints using traditional powders is selecting a powder that provides the maximum contrast in relation to the background surface to which it is being applied. [7] Unfortunately, in the

complex environment that surrounds us, most surfaces are not of uniform colour or pattern, meaning that the choice of fingerprint powder can become extremely limited; on a black and white substrate, for example, a white or black powder would only clearly develop ridge detail that could be readily visualised on the contrasting colour to which it is applied. In an effort to overcome such barriers, research groups and manufacturers have sought to develop luminescent powders whose fluorescence can be viewed upon excitation with an appropriate light source and blocking filter (often worn in the form of goggles). [7,8] The advantage of such powders is that the (multi)coloured, or patterned, nature of the background becomes slightly less significant owing to the independent fluorescence of the powder. Therefore, it is hoped that developed fingerprints fluoresce independently to the surface they are being viewed on, thereby reducing the variable-contrast issue that would arise if non-fluorescent powders were used [9].

The majority of commercially available luminescent fingerprint powders work on the notion that their optical properties enable them to fluoresce somewhere within the visible part of the spectrum, that is 400–700 nm, usually upon excitation with ultraviolet (UV) light (such as those manufactured and/or distributed by Lightning Powder Company, Tetra Scene of Crime and Sirchie, for example). Although inherently this serves as a useful property of the bulk material itself, a major limitation and drawback arises when the powders are applied to unusually coloured, multi-coloured and/or patterned backgrounds whose intrinsic formulations (inks, binders, coatings, etc.) also cause them to fluoresce within the same area of

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the spectrum. This creates a lack of distinguishability between the fluorescence of the developed latent print and the background [9]. In addition, historical limitations of such powders at the crime scene have concerned the requirement for specific equipment needed to visualise the results; this is largely overcome in the modern era with the abundance of commercially available handheld forensic light sources and fluorescence filters.

The proposed use of lanthanide-based 'anti-stokes' upconverter powders has gone some way towards alleviating background interference issues typically encountered across substrate types [10–13]. The anti-stokes emission occurs when high energy (~6 W) Infrared radiation is directed over fingermarks that have been dusted with an anti-stokes powder, thereby inducing a blue-shift fluorescence into the visible spectrum (typically green). Often, background suppression may be improved, but as the imaging still resides within the visible spectral window, substrate artefacts naturally occur within the resulting image and these may still prove problematic. Moreover, the laser intensity Infrared radiation required for anti-stokes emission means that a field-portable instrument is largely impractical owing to health and safety issues that surround this method of detection.

Recently, our research group reported the development of a novel, naturally occurring, non-toxic infrared (IR) fluorescent fingerprint powder that is suitable for use on both non-porous and semi-porous substrates that are typically considered extremely difficult to treat [9]. The fundamental benefit of imaging in the IR is that while many substrates fluoresce under UV illumination, less do so under higher wavelength regions of the visible spectrum. In addition, many inks used in security documentation (banknotes, passports, etc.) tend to become transparent in the IR [14] and so a contrasting fluorescent material on its surface can yield high contrast to the background. The material developed within our group, *fpNatural 1*TM, has the unique advantage of fluorescing in the NIR upon blue or red illumination and with appropriate camera filters, intense NIR fluorescence may be easily captured with minimal background interference.

However, illuminating with a visible wavelength can still, in some instances, allow background artefacts to leak through/fluoresce in the lower wavelength region of the NIR. Although this has been found to rarely detract from the distinguishability of the developed fingermarks, [9] advances are always sought to optimise efficiency, ease of fingerprint recovery and imaging capability. In addition, the use of narrowband visible illumination commonly requires the evidence to be shrouded (blacked-out) to allow only that specific wavelength to illuminate the sample to ensure maximum fluorescence contrast to be observed and for some circumstances of end-user operation this may not always be possible.

Cuprorivaite is a mixed calcium/copper tetra silicate, $\text{CaCuSi}_4\text{O}_{10}$, which was discovered as the naturally occurring mineral, by Minguzzi [15] within the Vesuvio lava in 1938 and confirmed initial findings from Pompeii by Davy in 1815 [16]. Since then, synthetic efforts to prepare cuprorivaite took the form of high temperature solid-state reactions in which, typically, calcium carbonate, malachite, silica and a soda flux are heated together at temperatures between 850 and 1000 °C [17–19]. The resulting crystalline material is blue in colour owing to the presence of square planar Cu^{2+} linked by SiO_4 tetrahedra in an extended layered framework containing alkali earth ions. The material itself is extremely robust and is chemically and physically stable at temperatures up to 1000 °C. It has also been shown to be resistant to acidic exposure [16,18,20,21]. These properties make cuprorivaite an ideal material for use within the pigment industry as it remains versatile enough to be applied as a paint and robust enough to stand the test of time and environmental stresses.

Importantly, however, cultural heritage investigations over the past 15 years have led to the intriguing discovery that when

illuminated within the visible spectral window (500–700 nm, $\lambda_{\text{exc}} = 637$ nm) cuprorivaite exhibits an intense broad emission in the near-infrared region ($\lambda_{\text{max}} = 910$ nm). Accorsi and co-workers conducted photophysical investigations of cuprorivaite in which they concluded that three different electronic transitions are attributable to Cu^{2+} ions which are expected to be the only photoluminescent component within the compound [22].

The majority of luminescence investigations conducted on cuprorivaite have concentrated on illuminating the samples as close to the primary excitation maxima, 637 nm (i.e. a red light source), as possible. It was noted, however, that cuprorivaite also has a secondary excitation absorption band between 700 and 800 nm, with a peak maxima close to 780 nm. This falls into the near-infrared (NIR) region of the spectrum, which is an area that has been little exploited within fingerprint forensics, but an area that we have already shown to offer real advantages over conventional techniques used to develop latent fingermarks.

Herein, we report the development of a novel, non-toxic fingerprint powder which has the unique ability to act as a NIR–NIR fluorophore, thereby suppressing nearly all background interference by fluorescing at longer wavelengths. The material may be imaged under typical domestic lighting conditions, given its impressive high-quantum yield and 'invisible' excitation wavelength outside of the visible spectrum, thereby reducing the need for a 'blackout' environment. The application of tailored light sources and portable instrumentation are illustrated, and the use of this combination discussed for crime scene utilisation.

2. Methods and materials

All latent fingermarks deposited during these investigations were taken from a range of 10 donors (six male, four female, all aged 25–50 years) at varying times across the day (9am–5pm) so as to create a 'natural' working portfolio of latent deposits. Approximately 300 fingermarks were deposited in total throughout this preliminary investigation. Unless otherwise stated, fingermarks were deposited as 'natural' marks and not groomed or loaded in any way with sebaceous or eccrine content. Typical aging of fingermarks prior to development was 1–14 days. The split depletion trials discussed, *vide infra*, consisted of 10 natural fingermarks being acquired from two donors using polymer banknotes as the test substrate. The marks were planted as depletions across the divide of identical notes. All samples were stored under atmospheric room temperature conditions (*ca.* 18 °C) in sealed plastic containers.

A range of substrates were selected with a variation in porosity, surface treatment, colour and pattern. Typical examples included metals (untreated, painted, plastic coated, aerosol cans), glossy papers (magazines, packaging, boxes), glass, plastics, wrapping paper, gift bags, banknotes (polymer and paper). Some samples, where mentioned, were retrieved from recycling collection points and treated 'as received', thereby developing pre-existing true latents.

The NIR–NIR fluorescent fingerprint powder developed during this study was a modified variant of cuprorivaite, $\text{CaCuSi}_4\text{O}_{10}$ (L. Cornelissen & Son). Powders were milled initially using a Fritsch Pulverisette planetary ball mill to establish a mean particle size of approximately 9 μm , as determined through use of a Malvern Masterseizer 2000 Particle Size Analyser (volume weighted mean = 9.3 μm , $d_{50} = 6.3$ μm , $d_{90} = 21.1$ μm) and a Leica VZ105 C Digital Video Microscope fitted with an AVT Stingray F201 C camera, under white light illumination at 350x magnification. Latent fingermarks were developed using a Tetra Scene of Crime fibreglass zephyr fingerprint brush with typical rotational application, ensuring the developed marks were gradually built up.

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