



Developing Fingermarks on Circulated Scottish Banknotes

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Executive summary

Polymer banknotes entered circulation in the UK from 2015 to 2022. These are designed to last twoand-a-half times longer than their paper or cotton predecessors, and some are now reaching their end-of-life. The type of substrate material affects the fingermark development technique or sequence that will reveal the most and best quality marks, therefore as banks switch to polymer notes, there has been a focus of forensic research on assessing the optimum visualisation technique. Most of this work has necessarily been done on mint notes, supplied by the issuing bank. However, the surface of a banknote changes as it is handled, and this affects the way fingermarks and development processes behave at the surface.

This work examines mint, laboratory handled, and circulated £5 and £10 polymer banknotes, studies the performance of fingermark visualisation processes and relates this to the degradation of the surface structure. A range of studies covered 3296 fingermarks, aged 8 to 56 days, on Bank of England, Bank of Scotland, Royal Bank of Scotland, and Clydesdale Bank polymer notes.

A total of 1856 fingermarks on mint and handled £5 banknotes from the four different issuing banks were visualised with Vacuum Metal Deposition (VMD), Cyanoacrylate Fuming (CAF) and, on Clydesdale Bank notes, fluorescent powder.

1440 fingermarks on mint and circulated £10 polymer banknotes from Scottish banks were visualised with VMD, Power Suspension (PS), one-step CAF technique, Lumicyano[™], as well as sequences of these techniques, including visualisation with reflected infra-red (IR) illumination.

As a single technique, VMD was significantly more effective than other processes in developing fingermarks on handled or older circulated banknotes, across all the banks studied, although effectiveness varied with issuer. For example, on handled Bank of England notes 45% of marks showed ridge detail with VMD development and 28% with CAF; for circulated Royal Bank of Scotland 2016 issue notes success rates were 49% with VMD and 25% with Lumicyano. Although for the 2021 issue, which are less degraded, Lumicyano produced better quality marks, with 59% development compared to 56 % for VMD.

The surface structure of the banknotes was studied to examine the texture at a macro and micro level. Circulation of banknotes causes degradation of the surface structure, with different features observable by eye and with simple light microscopy, such as folds and cracks, loss of intaglio and pearlescent surfaces, and contamination. At a higher resolution the formation of a micro-cracked surface structure can be seen in the handled and circulated notes. The wettability of the surface is also increased. These micro features can lead to the trapping of powder, or contaminants, increasing

quantity of development agent in fingermark background between the ridges, decreasing contrast and decreasing performance of powder-based fingermark development techniques. For example, circulation of Bank of Scotland notes reduced the number of marks developed with PS showing ridge detail from 63% to 29%. These same features can restrict the migration of components of the fingermark, lessening fingermarks degrading through spread of material and thus reducing potential formation of empty prints, so that VMD development is not adversely affected to the same extent by the surface degradation, showing a reduction from 92% to 70% with circulation.

The separate areas of the banknote surface, such as opaque, transparent and pearlescent regions, as well as the raised text, complicate aging of the note and fingermark development. Some fingermarks were partially visualised due to crossing different regions of the banknote, and a sequence of techniques is beneficial. Lumicyano followed by VMD, or VMD followed by powder suspension, give improved results over single techniques; for example, the second technique increasing observed marks from 25% to 57% and 49% to 77% respectively on Royal Bank of Scotland 2016 issue notes. Reflected IR imaging enables suppression of background features and improves visualisation, regardless of technique or banknote issue or condition, for example on 56-day-old fingermarks on circulated Bank of Scotland note IR imaging increases VMD developed marks with visible ridge detail from 31% to 61%. IR imaging is also beneficial for sequences of techniques, on Clydesdale Bank notes for example increasing VMD + PS developed marks from 66% to 75%.

Different designs and manufacture leading to different ageing behaviour and distribution of material and textures across the banknote, suggest that studies on one issuer such as Bank of England are not necessarily directly applicable to notes issued by Scottish banks. The condition of the banknote needs to be considered when deciding on the optimum fingermark visualisation technique(s).

Recommendations

- 1. Performance of a fingermark visualisation technique on a mint banknote is not an adequate guide to performance on a circulated note. The handling or circulation of banknotes causes changes in the surface structure of the note that affect the behaviour of fingermarks and development processes. These include wettability and texture at a macro and microscale.
- 2. Recent issues of banknotes may have lower degradation; issue date or sequence of serial number may be useful as an initial screening of degradation, although notes of the same issuing bank and issue date will have different levels of damage.
- 3. Low powered ("inspection") light microscopy of the surface allows initial assessment of the degradation of the banknote which may be used to inform the choice of fingermark development technique; however, this is effectively a proxy measure for the micro-degradation of the surface which adversely affects the powder-based methods.
- 4. Vacuum metal deposition (VMD) as opposed to powders, powder suspension or cyanoacrylate is optimum as a single technique on older circulated or handled notes. One-step cyanoacrylate fuming such as Lumicyano, may be more effective on less degraded banknotes. A sequence of techniques helps improve the overall development of the mark. Using sequences either as Lumicyano followed by VMD or VMD followed by powder suspension (with photographing at each step), further significantly improves the fingermarks obtained.
- 5. Reflected Infra-Red (IR) imaging regardless of the development method, reduces the background interference and can significantly improves mark quality.

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1 Introduction

In the field of forensic fingermark detection the development of fingermarks on polymer banknotes is evolving, as the number and types of circulating polymer banknotes increase. Launching polymer banknotes in 1988, The Reserve Bank of Australia released a new series of polymer banknotes during 2016-2020 with new designs and enhanced security features. The Bank of Canada launched a new series of polymer notes over 2011-2013, with additional issues of commemorative notes in 2015-2018. The UK had an extended process of switching from 'paper' to polymer banknotes with staggered release during 2015-2022 with examples of both types in circulation in 2015-2022. Unusually, in the UK in addition to the Central Bank (The Bank of England) some commercial banks are authorised to issue banknotes. In Scotland circulating notes are primarily issued by four banks, which have been converting to polymer notes over this period with a staggered release of a new issue; designs and release dates vary with denomination, issue and issuing bank. Polymer banknotes are designed to last two-and-a-half times longer than previous materials, and may degrade over a period of approximately five years, dependent on circulation and use [1-3].

Fingermarks (or latent fingerprints) and their visualisation are strongly affected by the donor, the ageing of the mark, and the substrate [4-10]. The choice of methodology for visualising the deposited mark is informed by the nature of the substrate, usually classified as porous, semi-porous or non-porous. "Paper" banknotes are porous and techniques such as amino acid stains, for example ninhydrin, may be optimal. Polymers are non-porous (or in some cases, semi-porous) and further consideration of the substrate is needed. In the case of polymer banknotes the substrate includes the type of the polymer itself, the pigmentation, lacquering, intaglio (raised) printing and surface texture, as well as some additional security features. Therefore, the polymer banknote surface provides a new set of challenges, and the behaviour of deposited fingermarks and development processes may not always be transferable between different issuing banks, or different designs of notes. The importance of surface properties for fingermark development quality was highlighted in a study of counterfeit paper US dollar bills [11] which were tested for fingermarks using the standard techniques used for paper banknotes; however, on the counterfeit sample notes the standard techniques were unsuccessful due to the effects of the differing surface. Other studies have shown variation in efficacy of development, and in the optimum choice of technique, among different surfaces within a single classification such as light polymer, thermal paper or adhesive tape [4,5,8,9,10,12,13].

1.1 Surface texture

The effect of polymer substrate texture on a fingermark and its subsequent development has been discussed by Jones et al [5]. The research investigated three different polymeric surfaces, classified as "smooth, light coloured" leading to the same recommended treatment processes through reference to the UK Home Office Centre for Applied Science and Technology (CAST) manual [6]. Atomic Force Microscopy (AFM) images of the virgin surfaces were used to assess the texture, and Scanning Electron Microscopy (SEM) images of the surface with developed fingermarks showed how the surface characteristics affected the mark visualisation. In addition to the numerical magnitude of the surface roughness, classified for example with R_a values, it was seen that the shape of the features, characterised by kurtosis and skewness, as well 'lay' of the surface, which is the direction of linear features such as ridges and scratches, had a significant effect on the development of fingermarks. This may be due to the migration of deposited fingermark material, which is affected by substrate

texture and surface energy [7] or by interaction with the developing agent for example trapping applied powders [5]. This links to the topography of polymer banknotes due to the variability of the surface including the polymer, security features and intaglio printing.

1.2 Polymer and additives

Studies into the influence of polymer type on the quality of marks developed using gold/zinc Vacuum Metal Deposition (VMD) varied the quantity of gold and precursor treatments [8,9]. Five different polymers were examined and there was no single optimal condition that was effective across all of the polymers used, or even for two substrates with the same polymer base layer. Bacon et al. [10] showed variability in fingermark development on light coloured polymers related to differences in type and local concentration of surface and sub-surface pigment, with proximity of titania based pigmentation increasing adherence of fingerprint development powder to the surface in that region, tentatively linked to surface energy.

In addition to variation in development efficacy between issuing banks, these findings could elucidate differences in fingermark behaviour or development of fingermarks across a banknote. This may be due to the difference between a clear or opaque section of a note, integration of surface and sub-surface security features, or two printed areas with different structures caused by the pigmentation.

1.3 Developing fingermarks on polymer banknotes

Various techniques, including vacuum metal deposition (VMD), cyanoacrylate fuming (CAF), powders and powder suspensions, have been found to work with differing degrees of success on several polymer banknotes [14-21].

Studies on the detection of fingermarks on Canadian polymer banknotes [15,16] focused on determining the most suitable sequential process for the development of latent fingermarks. The work recommended a combination of CAF, VMD and dye staining; VMD has been shown to enhance CAF development through nanodecoration improving contrast [22], as well as increase fingermark detection.

Other studies have investigated the use of copper VMD as well as IR and near-IR active powders to remove background interference from polymer banknotes [8,18,19]. An extensive study by CAST provided an initial insight for the development of latent fingermarks on the Bank of England £5 polymer banknote [20] although only examined mint, uncirculated notes; an iron oxide based powder suspension is now routinely used. In spite of the tendency of textured surfaces with negative skewness to capture powders thus reducing fingermark contrast [5], fluorescent powders have been shown to be effective on other complex surfaces, despite texture, background and environmental considerations [23]. Work investigating the enhancement of marks on mint £5 and £10 polymer notes from Clydesdale Bank and Royal Bank of Scotland found that a CAF process of PolyCyano UV followed by black magnetic powder was the most effective series of processes and this is now being implemented on these notes in Scotland [17], further work has used circulated notes in a laboratory setting [21].

Crucially, the majority of these studies so far are on mint, or uncirculated notes. Work on polymer banknotes in Israel showed that the recommended protocols for enhancing marks only enhanced 4

latent marks on 224 circulated banknotes [14]. Work on polymer structure shows light mechanical action can damage the surface and alter the surface roughness [24]. A surface roughness increase can negatively affect the efficacy of fingermark development processes, particularly powder-based processes [5], and influences the spreading of the fingermark over the surface [7].

This work therefore studies mint, handled, and circulated notes with a range of development techniques and relates the performance to the surface structure of the banknote.

1.4 Experimental Studies

Study 1 examines new and handled £5 notes from Clydesdale Bank, comparing the development of fingermarks on mint and degraded notes, using techniques of VMD and Green Fluorescent Magnetic Powder (GFMP).

Study 2 examines new and handled £5 notes from four UK banks that have been converting to polymer notes with a new issue over a period of time, comparing the development of fingermarks on notes across banks, using techniques of VMD and CAF.

Study 3 examines mint and circulated £10 notes from Bank of Scotland, to compare the visualisation of fingermarks on mint and degraded notes, using techniques VMD, Powder suspension (PS) and reflected infra-red (IR) imaging.

Study 4 examines circulated £10 notes from Scottish banks, to compare the visualisation of fingermarks on mint and degraded notes, using techniques of Lumicyano, VMD, PS in sequence, including IR imaging.

Across all studies the degradation of the banknotes is examined and related to the development efficacy to the surface properties.

2 Methodology

2.1 Banknotes

2.1.1 Mint Notes

As-issued banknotes were supplied by, or sourced from, Clydesdale Bank (CB), Royal Bank of Scotland (RBS), Bank of Scotland (BoS) and Bank of England (BoE) at or close to time of first issue. Notes were used as supplied ("mint") No additional cleaning method was conducted; mint banknotes may have some cashier handling, although extra care was requested this was not monitored.

2.1.2 Handled Notes

Banknotes were subjected to a light manual handling regime of approximately 2 minutes as a proxy for standard circulation of folding and crumpling ("handled"). Additional notes were subjected to extended repeated manual handling to mimic end-of-life state, with five repeats of the light handling process and storage on user in between process iterations.

2.1.3 Circulated notes

Banknotes were retrieved from general retail circulation over an extended period ("circulated"). Circulated notes were separated by bank of issue and nominal issue date, with Clydesdale Bank and Bank of Scotland each having one issue entering circulation in September or October 2017, respectively, and Royal Bank of Scotland with notes dated 2016 ("RBS16") entering circulation October 2017 and dated 2021 ("RBS21") printed in May 2021. Mint and circulated notes were kept separate and were stored, stacked, away from light, until required. Studies on circulated notes were conducted from November 2021 to June 2022, such that CB, BoS, and RBS16 banknotes were in use for approximately four years and RBS21 banknotes for approximately nine months. The reduction in cash use during covid restrictions may influence interpretation of these periods.

2.2 Surface analysis

2.2.1 Visual inspection – Light Microscopy (LM)

£10 Banknotes were examined by low power light microscopy (LM) using a Leica MX16 microscope and Brunel Laboratories camera. Each banknote was visually scanned over under low magnification (x1.8 magnification) and any area of interest feature imaged at varying magnification (x1.8 - x40) as appropriate for size, location, and orientation. This process allowed for the identification of several macroscale degradation features including linear such as cracks (long/short and shallow/deep), scratches (deep/shallow), creasing, and non-linear such as apparent bubbling, contaminants, ink wear, ink transfer, edge or corner wear, wear on Spark (pearlescent) number/denomination/symbol.

2.2.2 Surface texture- Laser Scanning Confocal Microscopy (LSCM) and Atomic Force Microscopy (AFM)

Laser Scanning Confocal Microscopy (LSCM, Keyence VK-X200) was utilised to image five areas of the mint and the end-of-life £5 banknotes. The LSCM has a minimal step of 10 nm between two adjacent focal plane scans which determine the Z-resolution.

Atomic Force Microscopy (AFM, JPK Nanowizard III) was used to examine the surface of mint, handled and circulated banknotes. Silicon probes of resonant frequency approximately 300 kHz were used in intermittent (tapping) mode, in air. AFM examination was conducted on regions of

banknotes each representing a single surface type (such as opaque, pearlescent or transparent polymer). Images of 100 μ m x100 μ m consisting of 512 scan lines of 512 points were collected at 0.1 Hz. Images were processed and analysed with JPK SPM 4.4.41 (JPK Instruments AG) and Gwyddion [25,26] for feature measurement and texture parameters. Five areas from each region were collected, mean values and standard deviations are reported.

2.2.3 Wettability – Optical Contact Angle (OCA)

Optical contact angle (OCA) measurements on sessile water droplets were conducted using a FTA1000B (First Ten Angstroms, Inc.) tensiometer. Droplet size was approximately 0.5 µl, using a 20 gauge needle, leading to a minimum droplet diameter of approximately 0.9 mm. Where banknotes were used for both AFM and OCA, the AFM was completed prior to OCA examination, each region of the note was further prepared if necessary to ensure a level region without surrounding obstructions. For each region of a banknote five droplets were examined. Droplets that spread to the edge of the region were discounted. Analysis of drop shape was conducted with FTA32 Video software (First Ten Angstroms, Inc.) images of the droplet after contact with the surface were taken at an initial interval of 0.100 s with an interval multiplier for successive images of 1.01 until the droplet was clearly stable, or up to 300 images (approximately 185 s). Optical contact angle was measured from the 2D image of the droplet, based on the image contrast. The mean value of the contact angle from ten consecutive measurements of each droplet were made once the droplet had stabilised on the surface; a manual, linear baseline was applied, and non-spherical drop shape assumed.

The size of the fingermarks (~1 cm) is needfully different from that of areas of examination for surface structure by LSCM (~1 mm), OCA (~1 mm) or AFM (~100 μ m). A fingermark may cover a range of different substrate surfaces which may affect the overall quality of developed ridges.

2.3 Fingermark study 1 Clydesdale Bank £5 comparison of VMD and GFMP development

Prior to deposition of fingermarks, twenty Clydesdale Bank notes were subjected to a light manual handling regime ("handled") by one person, as detailed in section 2.1.2. A further twenty were used as supplied ("mint").

A total of 320 natural fingermarks (those where the sebaceous secretions were not artificially increased through a grooming process) were deposited by 80 donation events by 76 individual donors over a two-day period. No instructions were given to donors except to request that they had not washed their hands for a period of 30 minutes prior to donation, aligned with the Home Office guidelines [27] and as used in other research for natural latent prints [4,5,13,22]. However, donor compliance was not monitored. Donors were limited to one donation per day. Banknotes were not physically cut or marked for deposition of fingermarks but were virtually divided into eight octants, in two rows of four, for ease of deposition and reference. The front (obverse) of the notes only was used. Donors deposited four marks, one from each of left and right index and middle fingers, onto individual octants of the supplied banknotes each donor placing latent marks onto two octants of a mint note and the matching octants of a handled note.

Notes were stored, lightly covered, in ambient conditions for a period of 11 - 13 days. Notes were developed with VMD (section 2.7.1) or GFMP (section 2.7.2).

2.4 Fingermark study 2 Interbank comparison of VMD and CAF treatment on £5 notes

Twenty-four notes from each of Clydesdale Bank, Royal Bank of Scotland (RBS), Bank of Scotland (BoS) and Bank of England (BoE) were treated as supplied ("mint") and a similar set were subjected to a light manual handling regime ("handled") by two persons, as detailed in section 2.1.2.

A total of 1536 fingermarks were used for this study. An approximately even mix of male and female donors, between the ages of 20-60 were asked to deposit natural fingermarks after having not washed their hands for at least 30 minutes prior to donating marks. To ensure an even distribution of secretions across the hands, donors were requested to rub their hands together and to then deposit marks using firm even pressure. Each collection of donated marks used sixteen donors, and sixteen notes (one of each condition from each bank for each development technique). The front (obverse) of the notes only was used. Each donor was attributed a specific area of handled or mint notes during each donation day, and a different finger was used between each note and each donor per donation day. This meant that each note would contain marks from eight different donors and eight different fingers, which were comparable among the eight different notes. The position on the note and finger used on each note was changed for each donor over a 10-week period used to build up the sample set.

All banknotes with fingermark deposits were stored for eight days between donations and processing days. After donations, notes were lightly covered, deposit side-up and stored at room temperature before being processed. Half of the set of notes was treated with VMD (section 2.7.1) and half with cyanoacrylate fuming and basic yellow 40 dye (CAF-BY40), as detailed in section 2.7.3.

2.5 Fingermark study 3 Comparison of mint and circulated Bank of Scotland £10 notes

This study compares performance of development processes on mint and circulated notes, and uses 80 banknotes, 18 donors, 800 fingermarks and 1200 visualisations, using VMD (section 2.7.1) and PS (section 2.7.4) development techniques. Ten mint and ten circulated Bank of Scotland notes were used for each development technique and aging period. The obverse (front) of the notes were used, ten marks were deposited.

Fingermark donors were scheduled to individually attend the laboratory for fingermark depositions on mint and circulated Bank of Scotland banknotes. To collect a natural mark, donors were instructed to avoid washing their hands for at least one hour prior to attending the deposition sessions [27], although compliance was not monitored. A donor deposited 10 marks, in two rows of five, on the obverse (front) side of a mint banknote with one hand and, similarly, on a circulated banknote with their other hand. Prior to deposition, the fingermark donors were instructed to rub their hands together for a few seconds to evenly distribute the naturally occurring secretions on the surfaces of their hands. Five marks were deposited to either the top or bottom row of the banknote with the assigned hand using each finger in an order unique to each donor. A waiting period of 5-10 minutes was observed and the process was repeated with the bottom or top row, during which the donors were asked to briefly rub their hands together to redistribute the secretions on their hands.

Banknotes were kept, unstacked, flat in lightly closed cardboard boxes in the dark, for an ageing period of 14 or 56 days.

2.6 Fingermark study 4 Interbank comparison of circulated £10 notes with sequences of techniques

This study compares development sequences on circulated notes, and uses 32 banknotes, 17 donors, 640 fingermarks and 1920 visualisations, using VMD-PS and Lumicyano-VMD development technique sequences (section 2.7). Ten Clydesdale bank notes and 20 RBS notes, ten from each issue (2016, 2021) were used. Both sides (obverse and reverse) of the notes were used, ten marks were deposited on each side.

Fingermark donors were scheduled to individually attend the laboratory on two occasions for fingermark depositions on the front and back of circulated polymer banknotes. The donors were instructed to avoid washing their hands for at least one hour prior to attending the deposition sessions, as before. When attending deposition sessions, the donors were instructed to rub their hands together for a few seconds to evenly distribute the naturally occurring secretions on the surfaces of their hands. Each donor deposited a two-mark depletion on the front of each of ten different banknotes (from a range of issues) with a random finger, a different finger for each banknote. The process was repeated with different donors, with this side of each banknote receiving ten deposits, made up of first and second depletions from five different donors. The donors were then asked to return 2-3 hours later, again avoiding washing their hands within an hour of returning. They were instructed to rub their hands together to redistribute the residue on their hands and the deposition process was repeated for the reverse (back) side of each banknote. A single BoS note for each process was similarly prepared for comparison. Notes with deposited marks were then stored, as before, for two weeks prior to first development.

2.7 Development and visualisation of fingermarks

2.7.1 Gold-zinc Vacuum Metal Deposition

After ageing for the assigned time period, the samples for VMD were securely attached to a flat work tray using magnets, accompanied by a small paper sample with a control fingermark deposited on it. The tray was inverted and placed on the top rack of the VMD chamber (West Technology VMD360). 3mm of 0.25mm diameter gold wire (Alfa Aesar, 99.999% purity) was placed into the relevant molybdenum boat. For each development, the zinc quantity was checked and independent zinc embedding runs, without banknote samples, were carried out as required. The chamber was evacuated to a maximum pressure of 2.4x10⁻⁴ bar. The boat was heated to ensure complete thermal evaporation of the gold. Subsequent evaporation of the appropriate amount of zinc was judged, including using the paper sample as a positive control, and until the banknotes were judged fully developed [6]. The chamber was vented back to atmospheric pressure and the developed samples removed, the reverse side subsequently developed if required, and transferred for storage.

2.7.2 Magnetic powder

Green fluorescent magnetic powder (GFMP, CSI Equipment UK) was applied for study 1 according to standard powdering methods. Marks were illuminated with white light and no viewing filter and UV / Blue light (340-413nm / 400-469nm) with a viewing filter (415nm / 476nm); angle of view and lighting configuration was optimised for each mark. Some trial notes were developed with red fluorescent magnetic powder, with some successful marks but the visualisation with green was preferred by the grader; however, this may be related to physiology rather than technique performance, as seen in other comparison studies [23].

2.7.3 CAF-BY40

For cyanoacrylate fuming a CA30 chamber (Air Science) was used with 2 g of cyanoacrylate (Orapi 601). The humidity of the chamber was set to 80 % and after the chamber had reached this level of humidity, notes remained inside for a total of 45 minutes. After each run in the chamber the boat was measured to ensure that at least 90 % of the cyanoacrylate had evaporated. Once banknotes had been processed using CAF, they were dipped in a solution of Basic Yellow 40 dye (BY40), which was prepared by dissolving 2g BY40 (Sirchie, LV507) in 1L of ethanol (Fisher CAS64-17-5) for a period of approximately 15 seconds. Each note was then individually rinsed with water and left to dry.

CAF-BY40 developed fingermarks were viewed using a Mason Vactron Quaser 2000/30 and illuminated with blue light or blue-green (400-469 nm or 400-519 nm) with a viewing filter (476 nm or 529 nm respectively); angle of view and lighting configuration were optimised for each mark, and captured using the Integrated Rapid Imaging System (IRIS, Media Cybernetics) with a Redlake Megaplus 4-2i camera.

2.7.4 Powder Suspension

As the main component of the most commonly used powder suspension (PS), Triton X 100, is no longer approved for use due to environmental concerns, a commercial premixed powder suspension, Wet Powder[™] Black (Kjell Carlsson Innovation AB) was used throughout this study. Initial trials with the formula described by Downham *et. al.* [4] using iron-oxide powder and 10% Tween 20 also showed effective development, although the differences among powder suspension formulations, powder batch variation and specificity required limited use of alternative processes in this study [4,13].

Each sample was held securely in the middle of one end of the banknote and not subjected to prewetting. The sample was gently brushed with Wet Powder[™] using a soft-haired brush. Once the entire obverse of the banknote was covered, the reverse was treated in a similar way if relevant. The powder suspension was allowed to sit for 10-15 seconds before being rinsed thoroughly under a running tap. The samples were then laid flat on absorbent material on an open bench in the laboratory for air drying. Once dry, all samples were placed into lidded cardboard boxes for storage, as before.

2.7.5 Lumicyano[™]

Banknotes were prepared for Lumicyano[™] (Crime Scene Technology) fuming by cutting short, thin slits in one end of the notes to enable vertical suspension from metal hooks in a pre-cleaned CA30 chamber (Air Science). Approximately 2g of Lumicyano solution with 8% Lumicyano powder was

thoroughly mixed and placed onto the hotplate set to 123°C and chamber set to 80% relative humidity. Banknotes were treated at these conditions for 45 mins, followed by an automatic 15 mins purge cycle. Samples were stored as before.

Fluorescence imaging for Lumicyano-developed marks was conducted with a wavelength of 468-526 nm (Mason Vactron Quaser 2000/30) and a viewing filter of 529 nm on the day of development.

2.7.6 Photography and reflected infra-red (IR) imaging

For studies 1 and 2 to facilitate comparison with fluorescent techniques, VMD developed marks were and captured using the Integrated Rapid Imaging System (IRIS, Media Cybernetics) with a Redlake Megaplus 4-2i camera.

For studies 3 and 4 White light photography was conducted with a Nikon D5100 with fixed multi angle illumination. All samples were photographed within 14 days post development. Selected samples were further photographed at shorter subject distance, or imaged with low power light microscopy, as detailed in section 2.2.1, post development.

Infra-red reflection photography was subsequently conducted using a modified Nikon D7100, with illumination by Superlite M05 Infrared (Lumatec GmbH) LED system, supplying up to 1500 mW with output centred at approximately 875nm and bandwidth (FWHM) 40nm. Illumination position and power were optimised for each image.

2.8 Grading fingermarks

A range of different fingermark grading scales are in use to assess fingermark development techniques [28]. All grading of developed fingermarks on all samples was carried out according to the IFRG recommended Home Office scale (also called "CAST" or "Bandey") for fingermark grading [28,29] of 0 (no evidence of a print) to 4 (over 2/3 of mark with good ridge detail), table 1. The boundary between sufficient and insufficient for identification is likely to be within grade 2, where some marks will have appropriate levels of detail, but other marks may have limited ridge detail or have insufficient minutiae evident [23]. Marks were graded on visualisation of ridge detail; marks with the same grade may have differing strength of contrast between mark and background; contrast and brightness are adjusted in the image, but no additional processing has been applied. Graders are consistent within each study.

Where development or visualisation techniques are applied in sequence, the fingermarks are assessed after each process, and the best grade in the sequence is awarded, which may be different from the grade for the quality of the mark after the final process. Throughout the report we discuss levels of marks with "ridge detail", meaning graded 2, 3 or 4, and marks that are "identifiable" based on a grading of 3 or 4, although some grade 2 marks will also be sufficient to be identifiable.

CAST Grade	Descriptor
0	No evidence of a mark
1	Evidence of a mark, no ridge detail
2	Ridge detail covering less than 1/3 print area
2	Ridge detail covering more than 1/3 print area but less
5	than 2/3 print area
4	Ridge detail covering more than 2/3 print area

Table 1. Grading of fingermarks by amount of ridge detail present, using the Home Office or CASTscale [29].

As fingermark grading is a classification, rather than a continuous or quasi-continuous measurement, taking an average grade is not mathematically robust. It is more appropriate to consider the number of marks in each grade, or the number that are, for example, at least a grade 3. We therefore use stacked bar charts to present the results, and Wilcoxon and Kruskal-Wallis tests for significance, as detailed elsewhere [30] instead of the equivalent t-test and Anova. For techniques in sequence a Wilcoxon signed rank test is applied, correlations use Spearman's rho [31,32]. The open source R software [33] and Jamovi [34] were utilised to carry out these significance tests.

3 Model handling studies

3.1 Surface degradation of the banknotes with handling

The Clydesdale £5 banknotes have been examined by Laser Scanning Confocal Microscopy (LSCM). Five small, relatively uniform areas were selected for LSCM examination for texture analysis, collectively representing the principal areas of the banknote. Surface parameters are shown in table 2. Roughness increased on handled notes on all areas (table 2), for example on £5 logo (opaque coloured polymer) from R_a of 0.9 µm to 6.3 µm and transparent area from R_a of 0.2 µm to 2.8 µm. Kurtosis, which reflects the sharpness of features, and skewness, which reflects predominance of peaks or troughs, moved closer to uniformity with handling of notes.

Table 2. Surface texture parameter values of mint and heavily handled Clydesdale £5 banknoteassessed by Laser Scanning Confocal Microscope over different regions of the banknote.

	R _a / μm		Skewness		Kurtosis	
	Mint	Handled	Mint	Handled	Mint	Handled
Opaque colour polymer	0.9 ±0.1	6.3 ±3.9	0.3	-0.3	6.1	2.7
Intaglio Printing	2.8 ±1.7	6.4 ±2.9	0.7	-0.4	6.6	3.7
Pearlescent	0.5 ±0.1	2.8 ±1.2	-0.9	-0.1	15.7	5.6
White Print	1.0 ±0.1	3.1 ±1.4	0.3	0.2	4.8	3.1
Transparent	0.20 ±0.05	2.8 ±0.7	-44.5	0.2	3210	6.9

The LSCM examination of the heavily handled Clydesdale £5 banknote revealed the emergence of features such as cracking, up to 5 μ m in depth, segmenting of the surface through a connected network of cracks on areas of coloured polymer, and scratch marks on transparent area as shown in figure 1.



Figure 1. Surface structures of heavily handled Clydesdale Bank £5 note studied by LCSM (a-c) mint and (d-f) handled note. (a) image and (b) corresponding height map of opaque area, with intaglio features in bottom right. (c) intaglio shading from portraiture. Height map scale is 25μ m. Degradation with handling in (d) and (f) opaque polymer, and (e) showing removal of intaglio features, with degradation of polymer.

Intaglio printing, contributing to some text as well as shading of portraiture at a macro level, consists of raised areas approximately $15 \pm 4 \mu m$ in height on the mint note. This is reduced or completely removed in places with handling of the notes. The principal degradation method appears to be at the adhesion between the printing and the polymer substrate, rather than the cohesion of the printing. The material breaks off, rather than gradually eroding.

The cracked network effect observed on opaque polymer regions on handled notes contributes the large increase in R_a roughness from 0.9 μ m to 6.3 μ m. This also explains the high levels of variation observed in the roughness measurement (± 3.9 μ m) as the surface is composed of large cracks (up to 5 μ m wide) that separate areas of relatively flat, undamaged polymer in this region of the degraded notes.

Prominent in the transparent region of the mint note are 'pin prick' holes which contribute to the high kurtosis and high negative skewness in this material. As the note is handled, scratches and creases appear and pin prick holes are filled or distorted, which is reflected in the reduction of texture parameters kurtosis and skewness to levels similar to those in other areas.

The pearlescent area (in the shape of a map of Scotland in the Clydesdale notes) initially comprises of flakes of reflectant material embedded at angles within a smooth polymer with $R_a = 0.5 \mu m$. This is degraded principally by the removal of polymer material covering the reflectant flakes, the roughness increases to $R_a = 2.8 \mu m$.

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Lightly handled notes imaged within the AFM show the initiation of the degradation, proceeding in a similar way with the initiation of surface cracking, and some heavily cracked areas. Figure 2 shows (a) mint and (b, c) handled opaque regions of Bank of Scotland notes. The mint note shows no cracking, whereas all regions of the handled note observed some surface cracking, with some areas such as figure 2 (b) a few short cracks initiating, to figure 2 (c) where the surface cracking becomes the dominant feature.



Figure 2. Atomic force microscopy (AFM) height map images of 100x100µm areas of Bank of Scotland £5 (a) mint and (b,c) lightly handled banknotes, showing development of surface cracking.

3.2 Fingermark study 1 Clydesdale Bank comparison of VMD and GFMP development

The quality of detected fingermarks developed from Clydesdale £5 banknotes in each of the handling states and development methods, is summarised in figure 3, example images of fingermarks are shown in figure 4. The data is presented as the Home Office scale grades 0-4 a description of which can be found in Table 1. Figure 3 outlines the grades given to detected fingermarks on both the mint and handled Clydesdale banknotes with each development method (VMD and GFMP). This shows that VMD is a more effective technique, this process developed 55 % and 65 % of fingermarks to grades 3 or 4, an identifiable standard, on handled and mint notes respectively, whilst GFMP developed 1 % and 39 % of marks to an identifiable standard on handled and mint notes respectively (p<0.0001). These results also clearly show that the handling process has a detrimental effect on the subsequent enhancement of fingermarks; however, this effect is greater for GFMP than it is for VMD. This is expected to be due to the GFMP becoming trapped and building up in the cracks and scratches, produced through the handling of the notes which can be seen in Figure 1, similar to that observed in a study on texture effects on fingermark development on polymers [5].



Figure 3. CAST grades of fingermarks developed on Clydesdale £5 banknotes, study 1, showing GFMP and VMD development on handled and mint banknotes.



Figure 4. Example fingermarks developed on Clydesdale Bank £5 notes, study 1, showing VMD developed marks (a) empty mark on mint note opaque area; (b) high quality mark on mint note, with some obstruction from intaglio printing; (c) high quality mark across handled region of opaque polymer; (d) partial mark on mint note showing some ridge detail, and substantial background obstruction; (e) high quality mark across different materials; (f) GFMP developed with some background obstruction.

The variation in quality of detected fingermarks between the regions of the mint banknotes is consistent with the wide variability in the surface texture and material. Each of the eight regions of the banknote used for each fingermark deposition is not uniform but contains more than one of the five main materials/surfaces examined by LSCM. Higher grades of fingermarks are more likely in regions corresponding to three of four corners, and areas of lower roughness and more uniformity. Multiple components, such as transparent and pearlescent areas, may lead to lower development efficacy. The intaglio printing is also likely to have reduced the efficacy of development, this raised text is at a sufficient height to cause interference with the deposition of the fingermarks as well as development processes. The surface material, chemistry and energy will also have effects on the

behaviour of the fingermark deposit and the development process. Figure 4d, e show differential quality of development of areas of individual fingermarks dependent on substrate surface.

The variation in development quality may not be only due to the different substrate characteristics, but also due to the donor variation of the fingermarks deposited. Inter and intra donor variations on equivalent areas of mint banknotes result in some high quality marks, but also empty prints, for example figure 4a, which is consistent with other studies of fingermark development on polymers [14,35]. However, the number of donors should allow this variability to be averaged out over the study, therefore mitigating the impact of this variable.

Banknote handling affects the features of the banknote in a number of ways, that will have different balance in each of the regions. The handling reduces intaglio (raised) features; and fills 'pin prick' holes, and introduces surface scratching in transparent areas; and introduces surface cracking in the opaque polymer, segmenting of the surface. All these features can affect spreading of fingermark components and the development quality. It is known from previous studies [5,7] that scratches, narrow valleys, narrow ridges or cracks limit fingermark components spreading when parallel to the fingermark ridges but increase the spreading when they are on an angle towards them. The degradation seen on the opaque polymer causes segmentation of the surface at a scale of approximately 20 μ m, see figure 1. This is below the ridge width (~100 μ m) and will therefore likely improve the retention of the fingermark ridges in their deposited position and limit any spreading of the mark, good quality marks on handled notes in regions of opaque polymer can be seen in figure 4c,e. VMD development has been suggested to be affected by layers of deposited material 3 nm thick [36] and fingermarks have been shown to exhibit migrating layers of material of approximately 3-4nm, spreading tens of microns from the fingermark ridge [7]. This cracking may therefore act to stabilise or improve the VMD development on these regions of the handled notes.

3.3 Fingermark study 2 Interbank comparison of VMD and CAF treatment on £5 notes

In study 2, the efficacy of VMD and cyanoacrylate fuming was assessed on mint and handled banknotes issued by four different banks. These have different designs, leading to different surface features in each octant and different proportions of coverage by these variable features. Example images of developed fingermarks are shown in figure 5 and 6 for VMD and CAF respectively.



Figure 5. Example fingermarks on £5 banknotes developed with VMD on interbank study 2: (a,b) RBS, (c,d) BoE, (e) Clydesdale, (f) BoS. (a,b) Show high quality marks across opaque polymer with reduced visualisation on transparent region and boundary, on mint and handled notes respectively. (c) Mark on mint note well visualised on opaque region reduced clarity on foil and transparent areas. (d) Primarily empty print with some ridge detail on edge. (e) high quality development and visualisation of mark across heavily intaglio printed area although some distortion of ridges. (f) Handled note with high background interference with development.



Figure 6. Example fingermarks on £5 banknotes developed with CAF on interbank study 2: (a,b,c) BoE, (d,e) BoS (f) RBS. (a) handled and (b,c) mint notes sharing variability of visualisation of ridge detail across different regions of the substrate. (d) small amount of ridge detail developed. Other areas (e,f) show excellent ridge detail development, although visualisation is affected by background pattern in some places (f).

Collated results presented in Figure 7a show the performance of the techniques in developing fingermarks on mint banknotes. It is clear that the techniques are relatively comparable in terms of effectiveness and quality of fingermark development, and there are no significant differences between VMD and CAF development for any issuing bank, the effect size is very small for RBS, BoE and BoS and small (in favour of VMD) for Clydesdale. The effectiveness of each technique varied among banks (p<0.05) for example, on notes from RBS approximately 30% of marks were enhanced to grade 3 or 4, an identifiable standard, with either technique. On Clydesdale 50% with VMD and 41% with CAF were developed to this standard.

The effect on fingermark development of the stressing of notes to simulate circulation can clearly be seen in figure 7a and b by the decrease in grades achieved by fingermarks developed by both techniques.



Development Method and Issuing Bank

Figure 7. CAST grades of fingermarks developed on £5 banknotes on (a) mint and (b) handled notes with VMD and CAF development across all banks.

Figure 7b shows that VMD is more effective than CAF for enhancing latent fingermarks on the handled polymer notes, irrespective of bank of origin (Clydesdale, BoS, RBS p<0.001; BoE p<0.05). The effectiveness of the methods did vary between the banks, however, with approximately 15% and 18% respectively of marks developed to an identifiable standard using VMD on notes issued by the Bank of England and Clydesdale Bank. In comparison, fewer than 5% of marks on notes from both the Royal Bank of Scotland and Bank of Scotland were developed to a grade 3 or 4 standard with VMD, although 19% and 17% respectively showed some ridge detail. This is still significantly more effective than CAF on the same type of notes where 1% and 0% of marks for RBS and BoS respectively were enhanced to identifiable standard, and 6% and 1% showed some ridge detail. The handling of the notes results in degradation of the substrate which adversely affects the fingermark

The variation between banks may be related to differences in the manufacture and design of the notes, as the designs have different distributions of intaglio printing, transparent regions and pearlescent features. As was observed on the Clydesdale notes, the various regions comprised of a combination of different features, such as transparent, opaque or reflective areas, as well as intaglio printing. These differences in region composition are also apparent for banknotes produced by the other issuing banks. For example with VMD development, in figure 5a on mint RBS note, and corresponding area on handled note (figure 5b) fingermarks show clear ridge detail on opaque polymer region (left) but poorer on transparent, pearlescent and interface regions. Similar effects are shown on a BoE note (figure 5c). This suggests an additional technique in sequence could be beneficial in improving the visualisation on (parts of) fingermarks on substrate areas not well visualised with VMD. Donor effects also contribute to mark variation; figure 5d shows a predominantly empty print on opaque polymer with some ridge detail at edge. Areas of high variation in background structure can still exhibit marks, Figure 5e shows a high-quality mark, although with some distortion, on a heavy intaglio region. Figure 5f shows ridge detail but heavily obscured by handling and substrate background, suggesting post processing or additional visualisation process may be beneficial, as will be explored in studies 3 and 4, section 4.2, 4.3.

Figure 6a shows a handled BoE note and Figure 6b the corresponding region on mint note showing variation in development with substrate surface. Figure 6c shows excellent ridge detail and contrast with CAF on transparent and metallic region but reduced on opaque polymer region. This suggests, along with the VMD results, that a combination in sequence of CAF and the VMD may be appropriate. Partial marks are present in some cases, such as figure 6d, although high quality ridge detail is also observed e.g. figure 6e,f although background staining and pattern interference affect contrast, suggesting a one-step approach such as Lumicyano or PolycyanoUV may produce better visualised marks in some cases, as in section 4.3.

3.4 Comparison of handled note studies

Both studies show a decrease in effectiveness of fingermark development techniques with handling of banknotes, however, this decrease is less pronounced for development by VMD. The scratches and cracks observed in the banknotes by LCSM on extensively handled Clydesdale Bank notes, and by AFM on lightly handled banknotes across banks, will reduce the potential for the fingermark components to spread, potentially favourable for VMD development effectiveness. These same features will also adversely affect the GFMP development efficacy due to the trapping of the powder

visualisation.

particles within the cracks and scratches which were observed in the earlier study [5], this is evident in the reduction in numbers of grade 3 or 4 marks from 39 % to 1 % seen with GFMP development on banknotes that have been handled. Surface features such as these have also been shown to cause background development with CAF processes [10]. Reasons for low quality of mark visualisation in some cases, e.g. figure 5f, 6f, may be background interference in the visualisation processes (enhanced by BY40 dyeing for example, or requirement for incident light wavelength that enhances intensity of security or material features) and this may be rectified by approaches that use infra-red or anti-stokes development techniques, or post-hoc image processing [19,20]. However, in many cases the background trapping of development agent within surface structures, has reduced the contrast. Other marks exhibit reduced development (as opposed to visualisation) of the ridge detail, e.g. figure 5d, 6d, which may be related to the action of the fingermark components on the substrate surface, which if residue is (still) present in a ridge pattern, may require an alternative process or sequence of techniques.

There was a significant drop in the efficacy of the VMD technique in developing fingermarks on Clydesdale notes between studies 1 and 2. This is approximately 15% on mint notes and 20% on handled notes. This difference is likely due to a range of factors which includes: differences in banknote material or structure with issue date, donor secretion composition [35,37], duration and environmental differences such as humidity during storage, variation in the VMD operator and differences in the grader of developed marks [38]. The trends are the same in both studies, with VMD effective on mint and handled banknotes.

4 Circulated banknotes studies

4.1 Condition of mint and circulated banknotes

Example light microscopy images of circulated notes are show in Figure 8. Linear features are evident, such as scratches and cracking (figure 8 a,c) and other damage and contamination is also observed (figure 8 b,d). Cracking appears similar to that observed on handled banknotes, but overall less severe or extensive than the damage to £5 notes handled to mimic end-of-life [39]. The number of observed defects on mint and circulated banknotes is shown in figure 9. For study 3 the Bank of Scotland mint notes occasionally show a maximum of one scratch or other linear feature, with circulated notes have a maximum of 46. The study 4 data demonstrates that the extent of damage to the notes observed through light microscopy is a function of issuing bank and date of issue, though these are conflated. Clydesdale banknotes exhibit the most degradation, and were the first issued (entering circulation in September 2017); these have a mean of 40, and maximum of 77, linear features per banknote, similar to that of the RBS notes entering circulation the following month, compared with a mean of 18 and maximum of 31 for the RBS 2021 issue.



Figure 8. Example light microscopy images of circulated Bank of Scotland £10 banknotes, showing different types of damage. (a) and (c) show linear damage, arrows indicate multiple features within images, 1) light cracking 2) deep cracking 3) heavy scratches 4) light scratches. b) Contamination and d) pattern damage are also shown. Approximate magnifications a) 16x, b,c) 20x, d) 12x.



Figure 9. Areas of damage on £10 banknotes observed by light microscopy, by bank, issue and condition. Grey boxes indicate number of linear features such as cracks and scratches on each note, black boxes total number of areas of damage observed (including edge wear, contamination and ink transfer).

Surface area normalised to the projected area, RsA, derived from AFM scans of 100µm x 100µm areas, is shown in figure 10. This allows comparison of ten banknotes across different banks and issues, examining an area of opaque polymer without intaglio printing or iridescent features. This method of expressing a roughness factor, of the "actual surface" as a percentage of the "projected" or "geometric surface" reflects the fundamental behaviour of a liquid on the surface [40]. The lowest possible value of this parameter is 100 which represents a perfectly flat surface. The Bank of Scotland mint note has the lowest roughness factor 100.8 ± 0.3 this will represent the as supplied surface structure. The circulated notes have a higher roughness factor, caused by damage or degradation of the note, although the extent of this and the size of the variation of the measured areas of one note varies across the notes studied. Circulated Bank of Scotland note D (BoSD) with R_{SA} = 101.0 ± 0.1 and Royal Bank of Scotland 2021 issue (RBS21) 101.2 ± 0.4 are not significantly different from the mint note, whereas other circulated BoS notes are significantly higher roughness, BoSB has a roughness factor of 102.0 ± 0.4 and BoSC 101.8 ± 1.0 and Clydesdale Bank (CBB) 101.9 ± 0.6 . Although the date of issue will give an initial indication of the degree of degradation from the circulation of the note, such as between the RBS notes issued in 2016 and 2021, there is significant variability across the different notes from the BoS issue entering circulation in October 2017, with the texture of surfaces of some notes close to that of a mint note in this area.



Figure 10. Surface roughness factor, R_{SA} of the banknote surface from atomic force microscopy measurements of opaque polymer region of individual £10 banknotes.

As well as roughness factor derived from surface area, other aspects of the substrate, such as the shape and directionality of features are also important [5,7] in the behaviour of fingermarks and their development. The AFM images, examples of which are shown in figures 11 and 12, show the development of a microscale cracking (annotated "1") on the circulated banknotes, similar to that observed in handled banknotes, figure 2 (b,c) and does not appear on the mint note (Figure 11 a). The extent and severity of this microcracking of the surface varies from area to area and between circulated notes. Visible microcracking develops on the opaque polymer, initially 50 nm to 200 nm in depth and approximately 0.5 μ m across, a few 10s of micrometres long (Figure 11, c, e); as notes are further circulated cracks extend, deepen and widen, Figure 11 (b) shows a circulated RBS16 note with microcracks up to 500 nm deep developed, the surface becoming fragmented and beginning to appear like handled notes [39]. Other defects also develop with circulation and examples are shown in figure 11, such as micro holes, indicated 2, and macro scratches, indicated 3, which extend across the whole image and will be those visible by light microscopy as linear defects.

The spark metallic or pearlescent region of the bank notes also suffers from the development of microcracking in the surface, as shown in figure 12 (a, b) which may lead to the observed macro scale degradation of this region. The transparent region of the polymer is overall smoother than other regions, although even on the mint note does exhibit some pinhole defects as shown in figure 12 (c) and by LSCM in section 3.1. This region does not degrade through the development of microcracking, but macro scale scratches develop across the surface with circulation, these features are also visible with light microscopy and are apparent on the handled notes.



Figure 11. Atomic force microscopy height images of 100x100 μ m areas of £10 banknotes, (a) mint and (c,e,f) circulated Bank of Scotland notes, circulated (b) Royal Bank of Scotland 2016 issue and (d) Clydesdale Bank issue. With (1) microcracking (2) micro scale holes and (3) macro scratches. Height colour scale of 2 μ m on all images.



Figure 12. Atomic force microscopy height images of 100x100 μ m areas of (a,c) mint and (b,d) circulated Bank of Scotland £10 notes, (a,b) Spark region with height colour scale of 2 μ m (c,d) transparent polymer region with height colour scale of 1 μ m.

Contact angle measurements show circulation of the banknotes has an effect on the wettability of the surface, as detailed in figure 13. The Mint Bank of Scotland note, with an OCA of 90.4 \pm 1.7 $^{\circ}$ showed the smallest range of contact angle over the sample. The recently introduced RBS21 and one Bank of Scotland circulated note (BoSB) show similar contact angles, though with a greater variation in the sample, no significant difference from the mint notes. Other circulated banknotes have reduced contact angles, for example 76.7 \pm 5.2 °, 82.1 \pm 4.7 ° and 71.7 \pm 12.7 ° for circulated notes from Bank of Scotland, Clydesdale Bank, and Royal Bank of Scotland, respectively. The reduced contact angle shows water-based liquids spread to a greater extent on the surface, and the increased variation over the sample highlights the inhomogeneity of the surface degradation and contamination. Although the recently introduced RBS21 note shows no significant difference from the mint Bank of Scotland note in terms of surface roughness and wettability, the other circulated banknotes may show an increase in surface roughness factor, or a decrease in contact angle, or both, in comparison to the mint note. This reflects the variability of degradation and contamination with circulation across a note and between notes, as well as the different analysis size of the techniques (two orders of magnitude in area). The presence of defects may also promote transition between Cassie state contact, where air is trapped in the textured surface beneath a droplet, and Wenzel state contact where the texture is completely filled with liquid [40-42] which affects spreading over the surface and adherence of the liquid to the substrate.



Figure 13. Wettability of the banknote surface from optical contact angle measurements of opaque polymer region of individual £10 banknotes.

4.2 Fingermark study 3 Comparison of mint and circulated £10 Bank of Scotland notes.

Across both ages of fingermark and all visualisation processes, the circulation of banknotes adversely affects the effectiveness of the visualisation, as shown in Figure 14. For two-week aged marks VMD shows ridge detail on 92% of mint bank notes, and produces 38% identifiable marks; by contrast on circulated notes, 12% of marks are identifiable and 70% exhibit some ridge detail. For powder suspension the effect of circulation of notes is even greater, 18% of marks are identifiable with 63% showing ridge detail on mint notes, reducing to 2% and 29% respectively on circulated notes. Similar effects are shown on the handled £5 notes, figure 3. For eight-week aged marks there is still a strong effect of circulation reducing the effectiveness of the visualisation. For powder suspension with infrared imaging, 35% of eight-week aged marks on mint notes were identifiable, and 90% showed ridge detail, on circulated notes this reduced to 4% and 59% respectively. Figure 15 shows example 8 week old fingermarks developed with (a) PS and (c) VMD, the corresponding reflected IR image (b, d) shows substantial additional ridge detail. The infrared visualisation has a clear beneficial effect here, allowing the reduction of the background and improved visualisation of the fingermarks, although there is no significant difference on the eight-week aged marks between the performance of powder suspension and VMD (6% identifiable and 61% ridge detail).



Figure 14. Breakdown of fingermark grades on mint and circulated Bank of Scotland £10 notes, study 3, showing performance of technique or sequence of techniques.



Figure 15. Fingermarks aged for 8 weeks on Bank of Scotland £10 notes, showing development with (a) PS and (c) VMD and visualised with white light, and the corresponding visualisation of the same marks with reflected IR light (b,d).

4.3 Fingermark study 4 Interbank comparison of development processes on circulated £10 notes.

Across all banks and issues, on circulated notes, VMD developed ridge detail on 60% of marks, compared to Lumicyano which developed 40% of marks with ridge detail. A breakdown of the fingermark grades can be seen in figure 16. Applying a second technique in sequence can improve development, although in some cases can be deleterious to marks that have already been developed. However, photographing or assessing the visualisation of the fingermark after each stage in the sequence allows the best development within this sequence of processes to be found. VMD followed by powder suspension enables 72% of marks to be developed with ridge detail; with the powder suspension method improving the development of 16% of marks, 8% improved by two or more grades. Lumicyano followed by VMD enables 59% of marks to be developed with ridge detail; with the VMD improving the development of 23% of marks, 16% improved by two or more grades. In each sequence 27% of marks were reduced in quality of ridge detail by application of the second process.



Figure 16. Breakdown of fingermark grades by technique or sequence of techniques, across all banks on circulated £10 notes.

Adding IR imaging allows the reduction of the background and offers some further improvement in the visualisation of the fingermark ridge structure. Using IR on VMD+PS further improves 23% of marks with 10% improved by two grades. IR also improves the Lumi+VMD sequence with 13% of marks improving, 10% by two grades. Overall VMD+PS+IR allows 80% of marks to be visualised with

ridge detail, 11% at grade 3 or 4; Lumi+VMD+IR allows 67% of marks to be visualised with ridge detail, 15% at grade 3 or 4.



Figure 17. Breakdown of fingermark grades by bank and issue date, showing performance of technique or sequence of techniques, on circulated £10 notes.

However, there are significant differences in performance of the techniques by bank and issue, as shown in figure 17. Lumicyano develops 59% of marks with ridge detail on RBS 2021 issue, but this is reduced to 40% on Clydesdale and 25% on RBS 2016 issue. VMD is less affected by bank or issue, with 56% of marks developed with ridge detail on RBS21, 57% on Clydesdale and 49% on RBS16. As a single technique VMD is significantly better than Lumicyano on the 2016/2017 notes that have been circulated for longer. Applying VMD in sequence following Lumicyano significantly improves the fingermark visualisation, RBS16 and Clydesdale with Lumi+VMD improve to 57% of marks with ridge detail, and RBS21 improves to 63%; which are also better than VMD as a single technique. Figure 18 shows the addition of VMD on a mark already well-developed with Lumicyano, and VMD developing significant ridge detail on a mark which showed no development with Lumicyano. A previous study [22] showed VMD development improves contrast of marks developed areas of fingermarks with no polymerised cyanoacrylate. The techniques work in a complementary manner, the need for this is also highlighted in the study of laboratory handled notes [39], related to the different materials of the banknote substrate, and is the goal of a technique sequence. Powder

suspension used in sequence following VMD significantly improves fingermark visualisation, improving from 49 % to 77 % of marks with ridge detail on RBS16, for example, though this process is less affected by bank or issue, figure 19 shows the sequences of VMD, PS, IR imaging on examples from Clydesdale Bank and RBS21, with PS adding extra ridge detail to the VMD developed mark in places, and IR allowing a different visualisation of the ridge detail.

Overall, VMD+PS+IR sequence allowed the most marks to be developed with ridge detail, on Clydesdale, RBS16 and RBS21 with 75 %, 81 %, 81% respectively compared to 64%, 64%, 72% respectively for Lumi+VMD+IR. However, on RBS banknotes the Lumicyano sequence produced a greater number of grade 3 and 4 marks.



Figure 18. Fingermarks on RBS £10 banknotes developed with Lumicyano (a,d) showing an identifiable mark (d) and an area of note with no indication of a mark following the Lumicyano process (a). The equivalent areas are shown in (b,e) respectively with a subsequent VMD process, the Lumi+VMD sequence showing ridge detail in both cases. IR imaging (c,f) in this instance does not add substantial benefit.



Figure 19. Fingermarks on Clydesdale Bank (a,b,c) and RBS21 (d,e,f) £10 banknotes. Developed with VMD (a,d) showing a potentially identifiable mark (a) and an area of note with little indication of a mark following the VMD process (d). The equivalent areas are shown in (b,e) respectively with a subsequent PS process, the VMD+PS sequence showing substantial ridge detail in both cases. IR imaging (c,f) allows a different way of visualising the marks.

4.4 Effects of condition on development

Reduced development on mint and circulated notes by VMD is in some cases related to variation of the banknote surface associated with the different material regions, for example marks obscured by intaglio printing, or crossing from opaque polymer to transparent polymer as shown in Figure 20 (a,b).



Figure 20. Fingermarks on Bank of Scotland £10 notes, (a,c,d,e) VMD on circulated notes (b) VMD on mint note and (f) PS on circulated note, where faint ridge detail is visible in the top right.

As in this image, in a number of cases on both mint and circulated notes, the transparent polymer and immediate surrounding area showed reduced (or no) background development with VMD, reducing contrast. However, marks were still developed on this area on some banknotes, as shown in Figure 20 c, although this may have been deposited by a user of the banknote during circulation. Other VMD developed fingermarks on circulated notes that show a lower quality than an equivalent on mint notes show variation over the mark that cannot be linked completely to the variation in the material of underlying substrate region, such as shown in Figure 20 (d); these may be affected by the microscale degradation and contamination changes to texture and surface energy. Other marks were near complete on the circulated notes, although in some cases were affected by the macroscale linear features of circulation disrupting the ridge pattern Figure 20 (e). This effect is also seen when considering surface topography as an aid to fingermark ridge migration and stabilisation [5,7]. Lumicyano development is strongly affected by the circulation of the banknote, performing well on recently issued 2021 notes, but significantly more poorly than VMD on circulated notes issued in 2016 (Figure 17). This is similar to the development of fingermarks on handled notes with CAF, section 3.3. Earlier work [43,44] has shown cyanoacrylate developing along linear features in the substrate, reducing contrast between ridge and background. For powder suspension development the linear features cause some powder trapping, Figure 21 (e), as seen in earlier studies [5], however, the principal differences observed between mint and circulated notes with PS development is the level of background staining on the circulated notes, which acts to reduce contrast. Figure 20 (f) shows a macro image of a PS developed mark with high background staining, figure 21 shows LM images of PS developed notes, (a,b) show development on mint notes with ridge detail, (c,d) show corresponding areas on circulated notes where PS has stained the whole area. This is likely to be linked to the microscale changes to the surface structure and energy, including microscale cracks. The LM shows degradation of the ink serial number and some apparent roughening of the surface texture under adherent powder, although AFM (figures 11, 12) shows this more clearly, as well as the scratching on the transparent polymer region (Figure 21 f, 12).

The opaque polymer surface of the mint banknotes, as shown by AFM, varies smoothly in height by up to 2μ m over the 100 μ m analysis length, this is unlikely to be detrimental to fingermark deposition or development [5]. However, microcracking develops with circulation, becomes more extensive, and develops into deeper cracks and a fragmented surface. This is of a scale and feature shape that is likely to be detrimental to the development of fingermarks with powder-based processes [5,39,44] causing the background staining observed. The relative orientation of these features with respect to a deposited fingermark ridge may also affect the behaviour of the fingermark on the surface, potentially acting as a channel or a barrier to mark spreading [5,7].

The spreading of the fingermark ridges will be affected by the surface energy of the substrates, the circulation of the notes has increased the hydrophilicity of the surface, indicated by the decrease in OCA, increasing the spreading of water-based liquids. This potentially widens the ridges, reducing the contrast of the fingermarks [7,45]. As the contact angle drops below 90° this leads to a tendency for a deposit to spread, and be governed by the volume of material deposited, rather than the initial shape of the deposit. The change in contact angle may also affect the quantity and composition of the residue transferred to the substrate as it is touched [45] as well as the behaviour of the mark with time from deposition including spreading [7] and crystallisation [45]. However, as the fingermarks age the spread area starts to degrade reducing to approximately 40% integrity from 16 to 61 days [7] which may influence differences in performance of development techniques between 2 week and 8 week fingermark ageing in the current study.

Linear features are observed which may have an impact on development [5,39,43,44] and fingermark migration [7,43,44]. The number of linear degradation features on the banknotes (such as scratches) is moderately to strongly negatively correlated [31,32] with the number and the quality of developed fingermarks on the banknote, with either VMD+PS+IR or Lumi+VMD+IR sequences. The overall amount of damage on a banknote, used here, may not always be the best indicator of potential effectiveness of fingermark development, as this is one component of the changes in the note

surface due to circulation. The distribution of the damage across the note will affect the deposited fingermarks and development techniques differently. The quality of development is also influenced by the variation between and within donors, depleted marks, and surface inhomogeneity. However, the condition of the banknote observed through the presence of linear defects may be useful as a proxy measure for the overall degradation, which includes macro and micro scale cracking and increase in hydrophilicity.



Figure 21. Light microscopy of PS developed Bank of Scotland £10 notes, (a,b) mint notes with ridge detail, (c,d) corresponding areas of opaque polymer and Spark printing on circulated note, showing material degradation and coverage of powder, (e) powder trapped in macroscale cracking as well as background staining, (f) transparent polymer region showing surface scratches.

5 Conclusions

Circulation of polymer banknotes causes degradation of the surface structure, with different features at the macroscale which can be seen with low powered light microscopy, such as scratches, cracking, and loss of intaglio printing. At the microscale circulated notes develop a cracking leading to segmentation of the surface, observed by AFM and as seen in the laboratory model of handling. Circulated notes exhibit a higher wettability than mint notes, as observed through a lowering of the optical contact angle. The extent of the degradation varies between different notes, and is affected by the circulation period of the note, with recent issue RBS 2021 having little change to wettability, and fewer areas of macro damage, but still exhibiting microcracking in some areas. The earliest notes entering circulation, Clydesdale Bank, have most macro damage, although notes of the same issuing bank and issue date will have different levels of damage or degradation.

Mint notes have lower damage and better performance of fingermark development processes. Performance of a fingermark visualisation technique on a mint note is not a good guide to performance on an older circulated note. The features induced by circulation affect the deposition [45] and movement [5,7,43,44] of the fingermark and interact with the development technique [5,43,44] such that powder-based and cyanoacrylate-based methods are adversely affected, through trapping of powders or initiation of polymerisation [39,43,44]; VMD is less affected by the material degradation. For example, on circulated Bank of Scotland notes, PS develops 29% of marks with ridge detail, VMD 70%; on Royal Bank of Scotland 2021 issue Lumicyano develops 59% of fingermarks with ridge detail, VMD 56%, but on the 2016 issue Lumicyano develops 25% of marks with ridge detail, VMD 49%.

This suggests that VMD, as opposed to powder, powder suspension or cyanoacrylate processes is optimum as a single technique on circulated notes. However, surface structure varies across the notes, with individual development techniques having varying effectiveness across materials [39]. A sequence of techniques helps improve the overall development of the mark. Using sequences either as Lumicyano followed by VMD or VMD followed by powder suspension, further significantly improves the fingermarks obtained. Background texture, images and structures help obscure developed marks [21,39], visualisation with reflected IR, regardless of the development method, reduces the background interference and significantly improves mark quality.

The number of macroscale linear degradation features (such as scratches and cracks) visible through light microscopy is moderately to strongly correlated with the number and quality of fingermarks developed on the banknotes, the greater number of linear-type defects [5] observable by LM the fewer fingermarks are recovered. Light microscopy of the surface allows initial assessment of the degradation of the banknote which may be used to inform the choice of fingermark development technique; however, this is effectively a proxy measure for the micro-degradation of the surface which adversely affects the powder-based and cyanoacrylate methods.

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Ancillary

This work has been approved by the Abertay University research ethics committee approval numbers EMS4230 and EMS4689.

Publications

This work is peer-reviewed and contributes to the following articles:

B.J. Jones, J.W. Cammidge, C. Evans, G. Scott, P.B. Sherriffs, F. Breen, P.M.B. Andersen, K.T. Popov, J. O'Hara, (2022). "Degradation of polymer banknotes through handling, and effect on fingermark visualisation." *Science and Justice*, *62*(5), 644-656. <u>https://doi.org/10.1016/j.scijus.2022.08.007</u>

P. M. B. Andersen, H. Gulbrandsen, Z.M. Davies, K.T. Popov, B. J. Jones, (2024) "Circulation of polymer banknotes, degradation and fingermark visualisation." Under review.

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Acronyms

AFM	Atomic Force Microscopy. Technique to measure surface texture at a micro scale.
BoS	Bank of Scotland.
BoSA, BoSB, BoSC, BosD, BosE	Individual circulated Bank of Scotland notes for surface degradation studies.
BoSMint	Individual mint Bank of Scotland notes for surface degradation studies.
BY40	Basic Yellow 40 (dye).
CAF	Cyanoacrylate fuming.
CAST	Centre for Applied Science and Technology. Formerly part of the UK Home Office, now absorbed into Dstl.
CBA, CBB	Individual circulated Clydesdale Bank notes for surface degradation studies.
Dstl	Defence Science and Technology Laboratory.
GFMP	Green fluorescent magnetic powder.
IFRG	International fingerprint research group.
IR	Infra-red (light).
IRIS	Integrated Rapid Imaging System.
LM	Light microscopy. Here, low power light microscopy to visualise degradation of banknote surface
LSCM	Laser Scanning Confocal Microscopy. A method to measure 3D surfaces at a range of scales.
Lumi+VMD+IR	Lumicyano [™] followed by gold-zinc Vacuum Metal Deposition (sequential treatment) and visualised under infrared light.
Lumi+VMD	Lumicyano [™] followed by gold-zinc Vacuum Metal Deposition (sequential treatment) and visualised under white light.
Lumi	Lumicyano [™] - a one-step luminescent cyanoacrylate fuming product.
OCA	Optical Contact Angle. A measure of surface wettability, a component of calculation of surface energy. The lower the OCA the more water spreads over a surface.
PS	Powder Suspension.
R _a	Average roughness.

RBS	Royal Bank of Scotland.
RBS16	Royal Bank of Scotland banknote(s) with issue date December 2016 and which entered circulation in October 2017.
RBS21	Royal Bank of Scotland banknote(s) with issue date January 2021, which were printed in May 2021.
R _{SA}	Roughness factor, actual surface area as a percentage of geometric or projected surface. Minimum value of 100 representing a perfectly flat surface.
SEM	Scanning Electron Microscopy.
VMD+PS+IR	Gold-zinc Vacuum Metal Deposition followed by black Powder Suspension (sequential treatment) and visualised under infrared light.
VMD+PS	Gold-zinc Vacuum Metal Deposition followed by black Powder Suspension (sequential treatment) and visualised under white light.
VMD	Vacuum Metal Deposition. Fingermark development process.