An Examination of the Sequence of Intersecting Lines using Attenuated Total Reflectance – Fourier Transform Infrared Spectral Imaging*

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ABSTRACT: In this study, the potential of attenuated total reflectance – Fourier transform infrared (ATR-FTIR) spectral imaging as a technique to determine the sequence of line crossings was examined. The technique was successful in determining the sequence of heterogeneous line intersections produced using ballpoint pens and laser printers. By imaging at characteristic frequencies, it was possible to form spectral images showing the spatial distribution of the materials. By examining the spectral images from the inks, it was possible to determine whether the ink was above or below the toner. In blind testing, ATR-FTIR spectral imaging results were directly compared to those obtained by eight experienced forensic document examiners using methods regularly employed in casework. ATR-FTIR spectral imaging was shown to achieve a 100% success rate in the blind tests, whereas some incorrect sequence determinations were made by the forensic document examiners when using traditional techniques. The technique was unable to image ink-jet printing, gel pens, roller ball pens and felt-tip pens, and was also unable to determine the sequence of intersecting ballpoint pen lines.

KEYWORDS*:* forensic science, questioned documents, spectral imaging, intersecting lines, line crossings, infrared, FTIR, ATR ballpoint ink, toner

Forensic document examiners are often called upon to determine the sequence of intersecting lines. An example of where it may be important to determine the order of strokes could be in a document such as a will or contract. The addition of an extra clause or paragraph could dramatically alter the terms of the agreement. If the added text overlaps, or intersects, with the signature on the document, then the alteration may be detected by examining the order of the strokes.

There are two main types of line intersections, homogeneous and heterogeneous. A homogeneous line intersection is defined as one where the line crossings have been produced using the same type of writing instrument or printing device, for example line crossings produced using two ballpoint pens. A heterogeneous line intersection is one where two different types of writing instruments and/or printing devices have been used to produce the line crossings, such as a laser printer and a ballpoint pen, or even a ballpoint pen and a gel pen (1). In this research, both types of line crossings are examined.

A number of techniques are available to examine intersecting line samples, and the choice of technique depends on a number of factors such as the writing/printing tools used to produce the intersecting lines, and whether a non-destructive technique is required over a destructive one. A comprehensive review of the techniques available is given by Poulin (1). Each of the available techniques has various advantages and disadvantages. For forensic analysis, a non-destructive technique is always preferred over a destructive one, and therefore destructive techniques, such as the scraping technique (2), where part of the toner material can be scraped away to reveal whether the ink deposit is below or above the toner, may not be suitable in many cases.

Many of the techniques used to examine line crossings are based on optical examination and observation of physical characteristics. These have the advantage of being non-destructive, but they do suffer some drawbacks. One of the biggest disadvantages of many techniques currently used is that they are subject to human interpretation. Optical microscopy is the most widely used technique in determining the sequence of intersecting lines (3). The drawback of this technique is that optical illusions can occur; for example, a heavier or darker pen stroke may appear to be on top of a lighter pen stroke, regardless of the actual order of strokes (4, 5). It has also been reported that with line intersections, 'there is a tendency to see what one perceives to be the correct sequence' (2). It may be that two document examiners examine the same intersection, and come to different conclusions regarding the order of strokes, as demonstrated later in this paper.

The ideal technique for determining the sequence of intersecting lines is one which is non-destructive, and can provide an objective method of analysis for a variety of material configurations encountered. Infrared spectroscopy is an objective, non-destructive technique, and is widely accepted for the analysis of many types of forensic evidence. FTIR spectroscopy has been used to analyse various materials encountered when examining intersecting lines, such as paper (6), ballpoint pen ink (7), and photocopy and printer toners (8-12). Even though FTIR spectroscopy has been used to analyse many of the materials commonly encountered in intersecting line samples, to date it has not been used to determine the sequence of strokes. Previously, to obtain both spectral and spatial information, a time-consuming process called infrared mapping had to be employed. This involved obtaining numerous infrared spectra, one at each point on a grid across the sample, through the use of a motorised stage. In the past decade or so, significant improvements in technology have led to the development of FTIR imaging, a powerful technique that can simultaneously obtain both spectral and spatial information in rapid collection times (13, 14).

The theory of FTIR imaging has been covered extensively elsewhere (14), so only a brief overview will be given here. The technique employs a Focal Plane Array (FPA) detector, with (typically) 64 x 64 discrete detectors (or pixels) laid out in a grid pattern. This detector simultaneously collects 4096 spectra in a single image, therefore allowing spectral and spatial information to be obtained in a short period of time. The image data collected can be thought of as a three-dimensional datacube, with vertical (x) and horizontal (y) spatial dimensions, and a spectral frequency domain (z) (15, 16). In other words, at every point (x, y) on the sample, an infrared spectrum is collected (z). It is possible to view an infrared spectrum at a particular point on the sample, and identify the chemical components present, or 'spectral images' can be formed by mapping the intensity of a particular parameter (such as spectral intensity at a chosen frequency) across the sample. A false colour map is produced by assigning a colour to each pixel according to its intensity at the chosen frequency, and the scale used in this research ranges from high-intensity red to low-intensity blue. By imaging at particular frequencies corresponding to the vibrations of chemical functional groups, the spatial distribution of different chemical components in the sample can be seen. It is hoped in this research that by choosing functional groups specific to the materials commonly encountered in intersecting line samples e.g. paper, ink and toner, spectral images showing their spatial distribution can be formed, and that they will provide information as to the order in which the line crossings were applied.

FTIR imaging has been shown to be successful for a number of forensic applications, including fingerprint visualisation (15), analysing multilayered paint chips (16), bicomponent fibre analysis (17) and visualising and identifying various materials including drugs and fibres either directly on a finger, or contained in a latent fingerprint (18, 19). As can be seen with the examples above, there are a number of ways in which infrared imaging can be used for forensic applications, including i) the pure visualisation of components (e.g. detecting and enhancing latent fingerprints), and ii) visualisation and identification of chemical components (e.g. multilayered paint chips and bicomponent fibre analysis). Determining the sequence of intersecting lines falls into the first category.

In this paper, the authors examine the use of infrared spectral imaging as an objective, non-destructive method for determining the sequence of intersecting lines. Both homogeneous and heterogeneous line crossings produced using a range of writing/printing materials are examined. The techniques attempted in this work are infrared reflectance microspectroscopic imaging and “macro” ATR (attenuated total reflectance) infrared imaging (20, 21). Since ATR is a surface-preferenced technique, and has been used successfully in conventional (non-imaging) analysis of inks and toner on paper (10, 22), ATR imaging holds the greater promise for sequencing of line crossings on paper.

Materials and Methods

Sampling

A variety of writing/printing materials was used to produce homogeneous and heterogeneous intersecting line samples including black ballpoint pens, gel pens, roller ball pens, felt-tip pens, and laser and ink-jet printers. The materials tested are listed in Tables 1-4. The line crossings sequences were known for all samples examined, apart from the blind sample set. All intersections, apart from the blind samples, were drawn on white EXP800 Laser/Copy Paper. For ease of analysis, an area roughly 2 x 2 cm in size, containing the line intersection to be imaged, was cut out of the document. It is possible, however, to manoeuvre an A4 page so that the intersection does not have to be cut from the document to be imaged.

Infrared Spectral Imaging

The intersections were examined using a Digilab Stingray® FT-IR imaging system, which is comprised of an FTS7000 FTIR spectrometer, coupled to an UMA 600 infrared microscope fitted with a Lancer 64 x 64 mercury cadmium telluride (MCT) focal plane array (FPA) detector. The system also has the optional Digilab LS large sample accessory, which allows for imaging of macrosamples. Images collected using this accessory had a spatial resolution of 44 µm, and a spectral resolution of 8 cm^{-1}. Images and spectra were collected and processed using Digilab Win-IR Pro 3.4 Software. The spectral range collected was 900 – 4000 cm^{-1}, with 900 cm^{-1} being the low energy cut-off of the FPA detector.

Feasibility Study and Optimisation of the Sampling Method

An initial feasibility study was conducted to determine the capabilities of the technique and also to optimise the sampling parameters. The feasibility study focused on heterogeneous intersections produced using four black ballpoint pens and two laser and ink-jet printers (see Table 1). The samples were imaged using two different sampling methods, these being reflection analysis using the microscope, and ATR analysis using a zinc selenide ATR crystal in the large sampling accessory.

For reflection analysis, the samples were imaged using the "expanded field of view" mode on the microscope in which each individual image tile is approximately 700 µm x 700 µm. It is possible using a motorized stage to stitch together or "mosaic" several image tiles to form a larger image of up to 2 x 2 cm. To obtain a large enough image size to examine an intersection, four (2 x 2) image tiles were collected, giving a total area imaged of ~ 1.4 x 1.4 mm. The infrared spectra within each image tile were collected at 8 cm^{-1} resolution, using up to 1024 co-added scans. Background images were collected from infrared-reflective metal oxide-coated glass slides (Kevley Technologies).

For ATR imaging, due to the size of the intersections, it was necessary to use a macro ATR crystal in the large sampling accessory. A FastIR horizontal / single bounce ATR accessory (Harrick Scientific) was used, which allowed for images roughly 3.0 x 2.3 mm in size to be collected. Initially a zinc selenide (ZnSe) ATR crystal was used, however results obtained (see Results and Discussion) led the authors to also investigate the use of a germanium (Ge) ATR crystal. For both ATR crystals, infrared spectra were collected at 8 cm^{-1} resolution, using 256 co-added scans. Background images were collected from the vacant ATR crystal surfaces.

Attempts were made to image both homogeneous and heterogeneous ink-ink intersections produced using the various different writing pens listed in Table 2. This testing was conducted using both the ZnSe and Ge ATR crystals in the large sampling accessory, under the parameters previously described.

Validation Study and Blind Testing

Validation studies were conducted using the optimal imaging method, i.e. using a ZnSe ATR crystal in the large sampling accessory. Five black ballpoint pens and five laser printers were used to produce known intersections where the ballpoint pen ink was variously applied before and after printing (see Table 3 for details).

Tests were conducted to determine whether ageing of the samples, and the pressure applied when producing the intersecting lines, affected the accuracy and quality of the results obtained. Ballpoint pen and laser printer line crossings were prepared for the ageing experiments using three black ballpoint pens (Sanford Suregrip, Staedtler and Uni S-AS) and two laser printers (Epson AcuLaser C1900 and Minolta Magicolour 2200). The samples were aged up to 12 months, and were imaged at various time intervals along the way. These ages were 0 days, 1 day, 3 days, 1 week, 2 weeks, 1 month, 3 months, 6 months and 12 months. The samples were stored in the dark in a drawer at room temperature. The same set of samples was imaged at each age, along with replicate samples of the same age that had not previously been imaged (in case the original samples had been affected by being clamped repeatedly onto the ATR crystal). Pressure tests were also conducted on intersecting black ballpoint pen and laser printing lines, which were prepared using two black ballpoint pens (Papermate Kilometrico and Sanford Suregrip) and an Epson AcuLaser C1900 printer. A SpectraTech Contact Alert system was used to monitor the pressure used when applying the ballpoint pen ink lines, both before and after printing. The line crossings were prepared using a pressure indicator of 4, 7 and 10 (arbitrary scale) when applying the ballpoint pen ink lines.

Blind tests were performed, in which the order of strokes was unknown prior to imaging. Four volunteers prepared twenty-one blind samples using various ballpoint pens and laser printers, and were asked to sign their name, neatly print a passage of writing and write a paragraph in their normal style of writing, either before or after printing. This protocol was used to ensure there was a variety of pen pressures and angles within the samples through the different styles of writing (see Table 4 for sample details). The results obtained using infrared spectral imaging were directly compared with those obtained by eight experienced forensic document examiners, who used methods they would use in casework (see Table 5 for details). The forensic document examiners were asked to independently examine the same set of blind samples and come to one of the following findings – ink over toner, ink under toner or inconclusive.

The intersecting line samples were photographed under white light illumination using a Video Spectral Comparator (VSC) 2000 (Foster and Freeman, UK) or a Sony DSC-W1 Cyber-shot digital camera and a Leica MS5 microscope with a Leica CLS50E light source.

Results and Discussion

Feasibility Study

An initial feasibility study was conducted to examine the potential of infrared spectral imaging for determining the sequence of intersecting lines. As mentioned previously, there are a number of ways the infrared spectral imaging data can be viewed. Infrared spectra can be extracted from specific points on the sample to potentially identify individual components, and data can also be viewed as a series of images, produced by mapping the spectral intensity at a given frequency across the sample. By choosing an infrared band specific to a given material, it is possible to view the spatial distribution of the chosen material across the sample. In order to successfully determine the sequence of intersecting lines, the following three criteria need to be fulfilled:

- the various writing/printing materials (as printed on the paper) must yield infrared spectra that can be resolved from the paper background;
- each writing/printing material must have at least one characteristic infrared band that allows it to be independently imaged; and
- there must be a consistent pattern of results for the two possible line crossing situations (i.e. material A on material B or vice versa); this means that there must be no physical mixing of the two materials and that it must be possible to detect the spectrum of the uppermost material without interference from the other material in at least one of the two situations mentioned.

Intersections produced using ballpoint pens, laser printers and ink-jet printers were initially examined using two different infrared sampling methods. With reflection analysis, as mentioned previously, it is possible to image larger areas (up to 2 x 2 cm) by stitching together or “mosaicking” multiple image tiles collected using an automated stage. Reflection imaging using up to 1024 scans at 8 cm^{-1} resolution was attempted, and while it was possible to image a laser-printed toner character, the ballpoint pen ink and ink-jet printer spectra seemed to be swamped by the paper’s spectral contribution (from cellulose and inorganic constituents within the paper), and so it was not possible to image any of the ballpoint pen inks or ink-jet printers tested. Another disadvantage of reflection imaging is that the time needed to scan an entire 12 point toner character (an image area of ~ 1.4 x 1.4mm) took over an hour and a half.

It was therefore necessary to try another sampling method. Imaging with an ATR crystal was thought to be suitable since the IR penetrates a short distance into the sample from the surface.It was hypothesized that the infrared spectra collected would contain less contribution from the paper background and allow for the ballpoint ink and ink-jet printing spectra to be seen. The theory of ATR analysis is covered elsewhere (23), therefore only a brief explanation will be given here as to why it is considered a surface-preferenced technique. Under the right conditions, the IR beam undergoes an internal reflection within the ATR crystal. Even though the IR beam is completely reflected at the internal interface, part of the radiation’s electrical field penetrates a small distance into the sample material; this penetrating field is often referred to as the evanescent wave. The distance the evanescent wave penetrates into the sample depends on a number of factors, including the refractive index of the ATR crystal material and of the sample.

ATR infrared imaging, using the ZnSe ATR crystal in the large sampling accessory, was found to successfully image laser toner and ballpoint ink crossings. Around six minutes was required to collect a 3.0 x 2.3 mm image (with 256 co-added scans at 8 cm^{-1} resolution). By choosing an infrared band present only in either the toner spectrum or the ink spectrum, separate spectral images showing the spatial distribution of these materials could be obtained. An example of the spectral images formed can be seen in Fig. 1. Figure 1a shows the visible light image of a line crossing produced with a black Staedtler ballpoint pen and an Epson AcuLaser printer. Figures 1b and 1c show the infrared spectra of the toner and ink materials, respectively. Figure 1d shows the infrared image of the toner material, and has been formed using the integrated peak intensity under 1724 cm^{-1}, which has previously been assigned as a carbonyl band in some major resins typically found in toner, such as polystyrene-co-acrylate or epoxy plus acrylate (8). Figure 1e is an infrared image of the ink material, which has been formed using the integrated peak intensity under 1584 cm^{-1}. The peak at 1584 cm^{-1} is characteristic of the ballpoint pen ink, and has previously been assigned to a skeletal vibration of triarylmethane dye and the C=C stretch vibration of epoxy resin (7). In this work, the actual identification of the peaks used to form the images is not vital; it is only necessary that the peaks are characteristic of the ink or toner materials and allow for them to be independently imaged, so that their respective distributions within the sample can be viewed.

The third criterion for the infrared spectral imaging technique to be successful in determining the sequence of intersecting lines was that a consistent pattern needed to be observed when one material was lying above or below the other material. The preliminary results indicated that examining the toner spectral image was not particularly useful in determining the sequence of intersecting lines. No consistent changes or breaks were seen in the toner image at the point of intersection to indicate whether the ink line was present above or below the toner. However, when examining the ink spectral image, a consistent trend was observed. When the ink was underneath the toner, there was a gap in the ink image where the toner had passed over the ink (see Fig. 1). When ink was present over toner, no break was seen in the ink line where it crossed over the toner line (see Fig. 2). This can be explained by noting that the toner material absorbs strongly in the infrared, and blocks the infrared light from penetrating through to the ink material, causing a gap in the image where the ink lies under the toner. When the ink lies on top of the toner, the ink spectrum is stronger, as there is less spectral contribution from the cellulose in the paper. In this case, no gap is seen in the ink line, and generally the image is stronger at the point of intersection (i.e. closer to a red colour (high intensity) in the image than green-blue (lower intensity)). The toner material generally gives a strong spectrum, and most of the time could be seen in the toner infrared images regardless of whether the ink lay above or below the toner. It therefore does not appear to help in determining the sequence order of the strokes.

Overall the preliminary study indicated great potential for infrared spectral imaging in determining the sequence of intersecting ballpoint pen ink and laser printer toner lines. However, infrared spectral imaging was unable to image either brand of ink-jet printer ink on paper, with the weak ink-jet spectrum being swamped by strong paper spectral interference. Therefore it was necessary to look at further sampling methods to see if any improvement could be made to the technique to make it more sensitive to ink-jet printing and/or less sensitive to the underlying paper. One approach was to investigate the use of a different ATR crystal material. The higher the refractive index of the ATR crystal, the shorter the distance the evanescent wave penetrates into the sample. Therefore germanium, which has a refractive index of 4.0, gives a lower depth of penetration into the sample than the zinc selenide crystal, which has a refractive index of 2.4. The expected downside from using a crystal with a higher refractive index is that the overall signal from the sample would be reduced, but it was hoped that using the germanium crystal would reduce the spectral contribution from the paper substrate relative to that of the ink, and thus enable the imaging of ink-jet printed characters on the paper surface.

Attempts were made to image the feasibility sample set using a germanium ATR crystal in place of the ZnSe crystal. Unfortunately, it was still not possible to image either brand of ink-jet printing using the germanium ATR crystal. This is possibly due to the mechanism of printing. With laser printing, the toner material, which consists of fine black particles, is fused to the paper surface through the action of heat or pressure. However, with ink-jet printing, the liquid ink droplets are simply sprayed onto the paper, and absorb into it (24, 25).

Overall the results obtained with the ZnSe ATR crystal were far superior to those obtained using the Ge ATR crystal, including for the laser printed samples. This is demonstrated in Fig. 3, where the differences in results between the two types of ATR crystals are depicted. The Ge ATR crystal struggled to image the toner and ink materials, whereas the ZnSe ATR gave very good images of these materials. A further disadvantage of using the Ge ATR crystal was that its sensitivity was so poor that it was difficult to see if the line intersection was clamped down onto the correct location on the ATR crystal prior to imaging. With the ZnSe ATR crystal, it was possible to push the sample onto the crystal surface using a finger, and by viewing the 'real time' infrared window (which shows the integrated intensity response of the detector in real time), instantly tell whether the line intersection of interest was in the correct position on the crystal for imaging (see Fig. 4b). However with the Ge ATR crystal, no image could be seen in the 'real time' infrared window, so there was a lengthy trial and error process to get the intersection onto the correct position on the crystal (see Fig. 4c).

Ink-ink Intersections

Attempts were made using both the germanium and zinc selenide ATR crystals to image various ink-ink intersections. Four different types of writing pens were tested, these being ballpoint pens, gel pens, roller ball pens and felt-tip pens. Unfortunately, the technique failed to give infrared spectra of the ink from gel pens, roller ball pens and felt-tip pens on paper. It appeared that as for the ink-jet printing, the spectra were swamped by the paper spectrum. The only pen type that could be imaged was the ballpoints. As with the ink-jet printing, this can be explained by the different ways in which these inks deposit onto the paper surface. Ballpoint pen ink is a viscous paste-like material that smears onto the surface of the paper, and is only partly absorbed into the paper, whereas the other types of pens, such as the felt-tip and roller ball pens, contain water-based inks that are more readily absorbed into the paper material (26, 27).

Line intersections produced using a number of different ballpoint pens were examined, and even though spectra of the inks could be obtained, the peaks allowing the formation of spectral images were common to all types of ballpoint pens encountered. The major ink peaks observed in the spectra occurred at 1584, 1360 and 1176 cm^{-1}, and are characteristic of triarylmethane dyes (7). Any minor differences in the ballpoint pen ink spectra that may exist between the different pen brands were swamped by the paper spectral contribution. This meant that, even though it was possible to form images showing the spatial distribution of the ink lines, these images gave no information as to which line lay on top of the other, as can be seen in Fig. 5. In a previous study by Wang et al, over 100 blue ballpoint pens were analysed by FTIR spectroscopy (7). They found two major categories of ink spectra, as shown in Fig. 6. One of the categories is the typical ballpoint pen ink spectrum that was encountered in this study, where the major peaks were due to the triarylmethane dyes (1584, 1360 and 1176 cm^{-1}). However, the other type of spectrum was quite different, with the major peaks occurring at 1730 and 1285 cm^{-1}. It is possible that infrared images of a line crossing produced using the two different types of ballpoint ink might be used to determine the order of the strokes, if the inks do not mix significantly at the intersection. Unfortunately in this study only one of these two types of ballpoint pen spectrum was encountered, and so this hypothesis could not be tested.

Validation Study

In the validation study, over 100 intersections produced using five brands of ballpoint pens and five laser printers were examined. Similar results were obtained to those found in the feasibility study, and the technique could accurately identify the sequence of all of the line crossings. A consistent pattern was observed in the ink spectral images, in that if the ink was above the toner material, no gap was seen in the ink line at the point of intersection, but if the ink was below the toner material, then a gap could be seen. The peaks used to form the spectral images were the same across the different brands of writing/printing materials, i.e. 1584 cm^{-1} for the ballpoint pen inks and 1724 cm^{-1} for the toner materials.

In the validation study it was observed that the spectral images were horizontally stretched in comparison with the visible light images such that the sample area imaged was 3.0 x 2.3 mm. This is due to the elliptical imaging area formed on the crystal surface when the circular beam of light hits it at 45^o. A detailed explanation of the phenomenon can be found in Chan and Kazarian (20). It would be possible to convert the image back to an un-stretched version using image processing software, but this was unnecessary for this study. For presentation at court it may be worthwhile un-stretching the image, or otherwise explaining why it appears slightly different to the visible image so as to avoid any confusion.

The aged samples were successfully imaged up to the age of 12 months, and the ink spectral images could still be used to correctly determine the order of strokes. At the age of 12 months, the general pattern of a gap in the ink image at the point of intersection if the ink was underneath the toner and no gap when the ink was lying on top of the toner could still be seen. A study by Wang et al. found that when ballpoint pen ink samples were artificially aged (through heating to 50-150 ^{0}C and exposure to UV irradiation), the intensity of the 1584 cm^{-1} peak decreased (7). In this study, no consistent trend in the intensity of the peak at 1584 cm^{-1} over time was observed, and this peak could still be used to form images showing the spatial distribution of the ink materials aged 12 months.

Pressure testing was conducted to determine whether the pressure used when applying the ballpoint pen ink, either before or after laser printing, affected the accuracy of the technique in determining the correct sequence of strokes. The pen pressure applied ranged from heavy to light. The results showed that, even with the varied pen pressure, there was still a consistent pattern in the ink images which could be used to accurately identify whether the ink was above (no gap at the point of intersection) or below (with a gap at the point of intersection) the laser printing. A notable finding was the effect the pen pressure had on the toner images produced. As stated previously, there was no consistent pattern in the toner images to indicate whether the ink was above or below the toner. More often than not, no gap was seen where the ink was lying on the top of the toner, leading to the earlier hypothesis that this was due to the strong toner spectrum produced. However, with the heaviest pen pressure (pressure indicator of 10), there was a gap in the toner image at the point of intersection with the ink (see Fig. 7d), at the medium pen pressure there was a small gap seen at the point of intersection (see Fig 7e), and at the weakest pen pressure, there was no gap seen in the toner image (see Fig. 7f). Observation of the toner spectral image may therefore help in some cases to reinforce the determination of the order of strokes; however the main focus should remain on the more reliable pattern seen in the ink spectral images, i.e. whether or not there is a gap in the ink line at the point of intersection. It is also important to examine the infrared spectra around the point of intersection to confirm whether toner or ink peaks are present, in addition to viewing the infrared images.

Blind Testing

Twenty one blind tests, where the order of strokes was unknown prior to examination, were conducted. The results obtained using the infrared spectral imaging technique were directly compared with those obtained by eight experienced forensic document examiners, using techniques they would employ in casework. The infrared spectral imaging technique was the most successful in determining the correct sequence of the line crossings, with a 100% success rate. The results obtained by the document examiners can be seen in Table 6; the majority of the document examiners got most sequences correct or made an inconclusive determination. However, there were two samples (BS1 and BS16) for which an incorrect determination was made by some of the document examiners. The samples where there were differing opinions among the document examiners were more difficult intersections, where the ink line was fairly heavy underneath the toner character (BS16) or where there was a fairly light ink line over the toner (BS1). This emphasises the need for a technique which does not rely on visual interpretation, as optical illusions can occur. The infrared imaging results for BS16 are shown in Fig. 8. This was one of the samples for which two document examiners incorrectly assigned the order of strokes, and two document examiners came to inconclusive results. By examining the ink spectral image (Fig. 8c) it is very clear from the gaps seen at the points of intersection that the ink was under the toner. This example highlights the effectiveness of the imaging technique, and the advantages of producing images based on chemical differences, rather than relying on visual observations, which can be subject to optical illusions and varied human perceptions.

For the majority of the blind samples, the sequence order of the strokes was fairly apparent upon visual examination, and in those cases the use of the more advanced technique of infrared spectral imaging may not be warranted. However, for more borderline intersections, or ones where an inconclusive determination is made, it would be worthwhile to examine the intersection using infrared spectral imaging, where available, to ensure that no errors are made in determining the sequence of line crossings. A further advantage of infrared spectral imaging is that images are formed that can clearly convey the results to a layperson, such as a jury member.

Conclusions

Overall, the infrared spectral imaging technique, using a ZnSe ATR crystal in the large sampling accessory, was found to be very successful in determining the sequence of intersecting ballpoint pen and laser printer lines. The chief advantages of the technique are that it is an objective method of analysis, and relies on chemical differences in the samples rather than simply visual differences that are subject to human interpretation. The technique is relatively fast, taking only six minutes to analyse a sample, and is non-destructive as long as the document can be manoeuvred so that the line crossing of interest is placed in contact with the ATR crystal. The results are also displayed in a way that makes them more easily understood by a layperson, such as a jury member.

The disadvantages of the technique include its inability to image a number of writing/printing materials on paper, including ink-jet printing, and various pens such as gel pens, roller ball pens and felt-tip pens. The instrumental requirements are more demanding than infrared microscopic imaging: it is necessary to have the optional large sampling accessory, and also a large enough focal plane array detector to collect an image of a line intersection. Many forensic laboratories do not have the infrared imaging instrumentation, but as more companies enter the market, the cost is starting to drop. Instrument companies are now marketing infrared microscopes that can be upgraded with the addition of an imaging detector, a much less expensive purchase than a complete new instrument.

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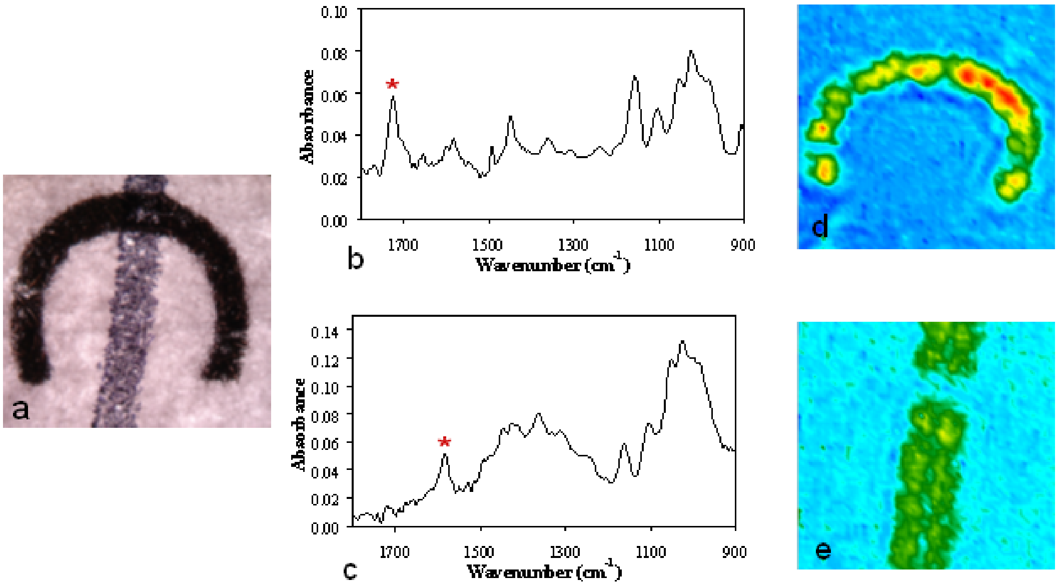

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b
c
d
e
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0.04
0.06
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0.10
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1700
1500
1300
1100
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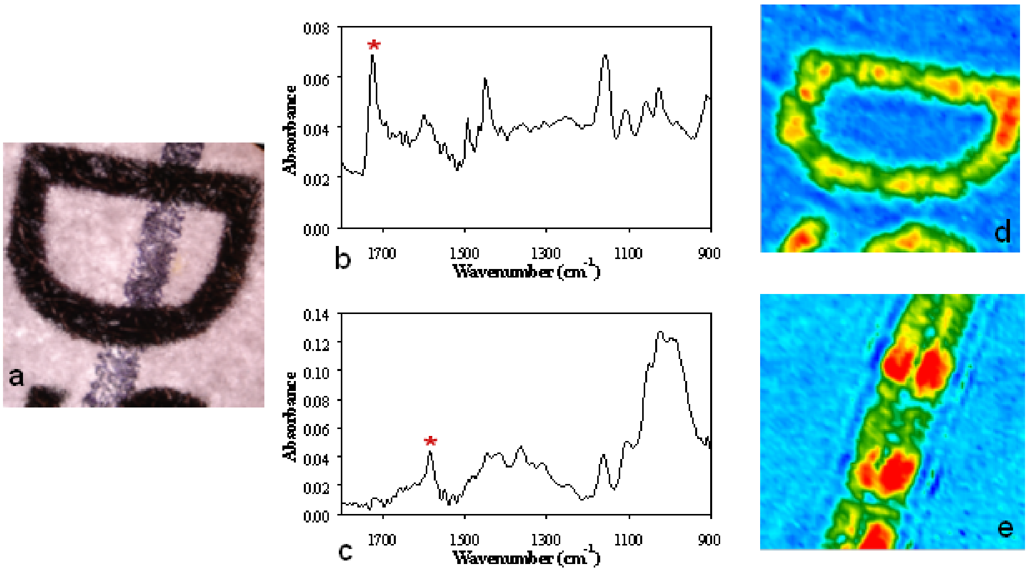
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Absorbance
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Wavenumber (cm-1)
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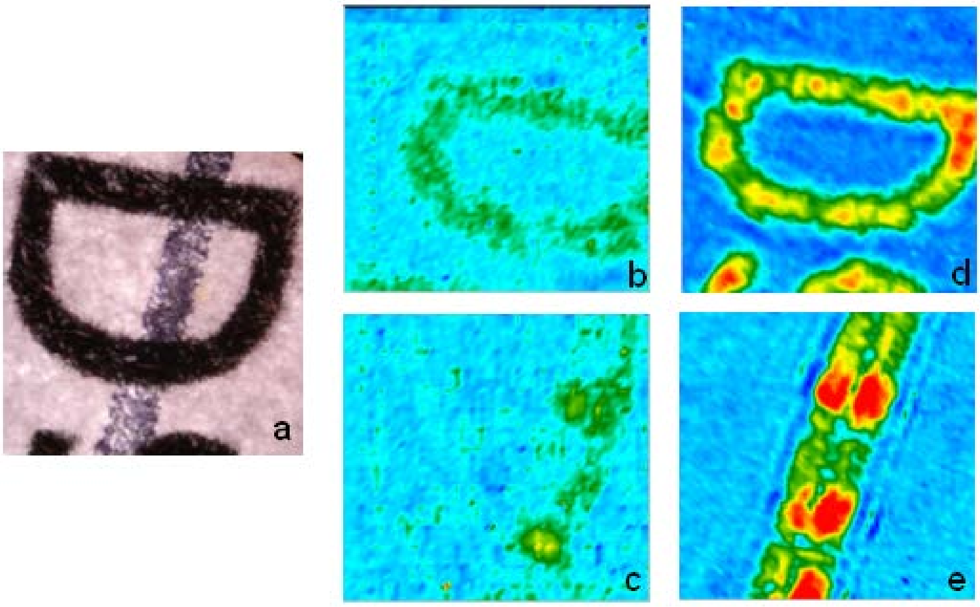
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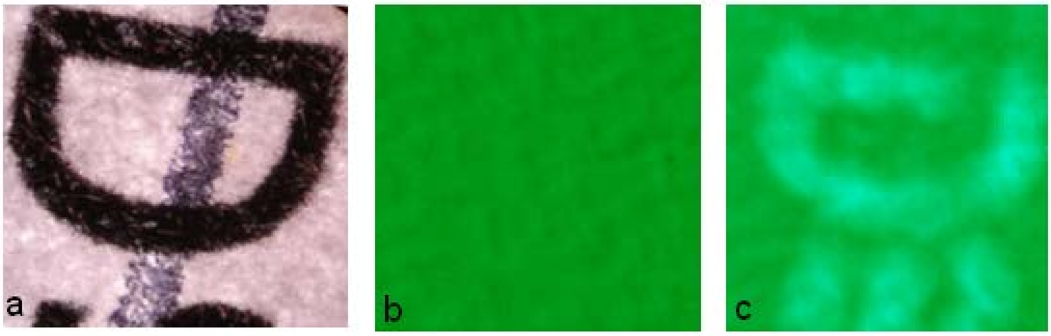

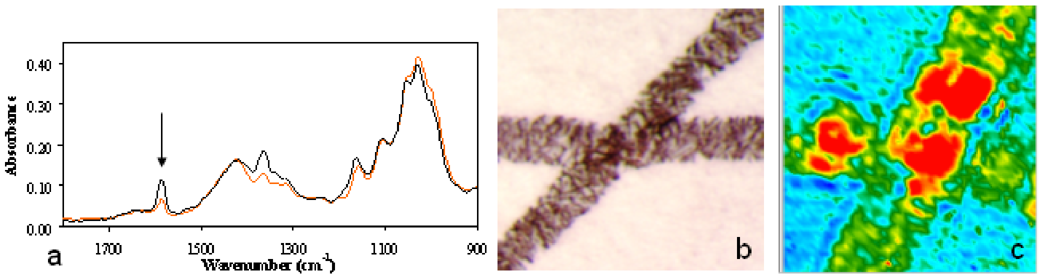
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Absorbance
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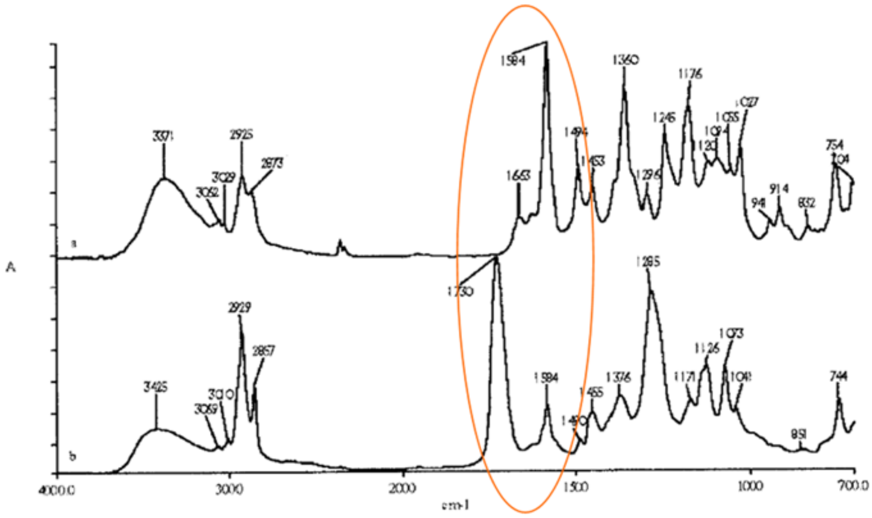
Wavenumber (cm-1)
a
b
c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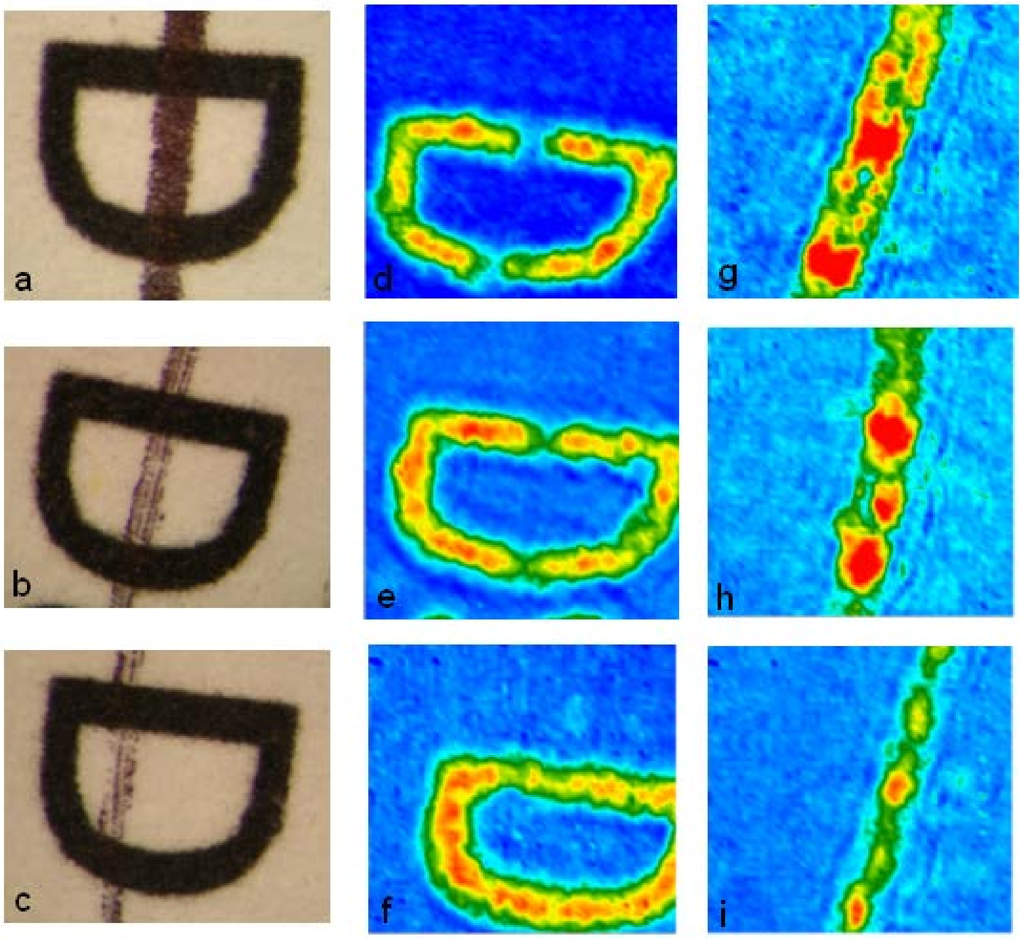
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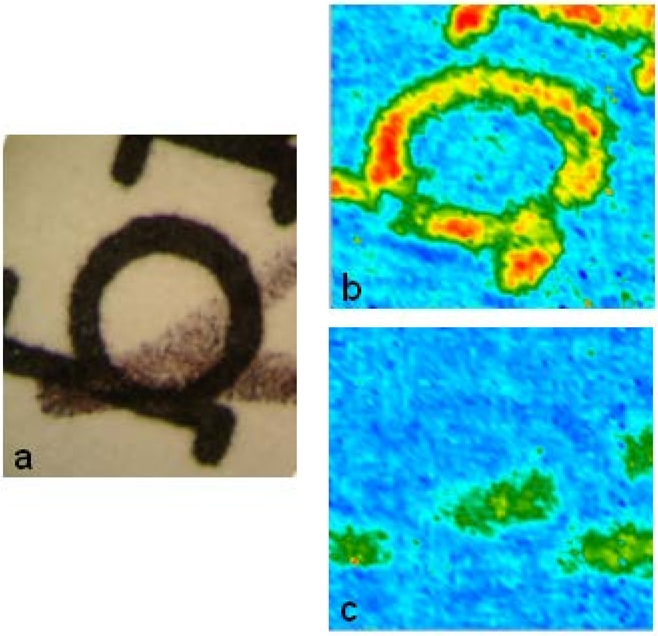
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b
c

TABLE 1 - *Feasibility study samples.*

Black Ballpoint Pens	Laser Printers	Ink-jet Printers
Papermate Kilometrico (M)	Epson AcuLaser C1900	Epson Stylus Photo 700
Uni SA-S (M)	HP LaserJet 1000	Canon Bubble-Jet BJC200
Staedtler Stick 430 (M)		
Sanford Suregrip (M)		

TABLE 2 - *Ink-ink intersection samples.*

Ballpoint Pens	**Gel Pens**	**Roller Ball Pens**	**Felt-tip Pens**
Sanford Suregrip (M)	BIC Intensity Clic	Staedtler cool roller	Artline 220 Superfine
Uni SA-S (M)	Hybrid Gel Grip DX	Uni-ball Micro	Pilot drawing ink (pigment ink)
Reynolds (F)	Zebra Sarasa	Bic Exact-Tip Roller	Staedtler triplus fineliner
Biro (F)	Pentel Energel (liquid gel ink)		
Staedtler Stick 430 (M)			
Bic Cristal (M)			
Ohto Gripper (F)			
Papermate Kilometrico (M)			

TABLE 3 - *Validation study samples.*

Ballpoint Pens	**Laser Printers**
Biro (F)	Epson C1900 AcuLaser
Pentel BKL10	Minolta Magicolour 2200
Artline (M)	HP LaserJet 1000
Sanford Suregrip (M)	Ricoh Aficio 1060 RPCS
Reynolds (F)	Kyocera FS-1000

TABLE 4 - *Blind sample details.*

Sample ID	Ballpoint pen type	Laser printer type	Paper type
BS1	Zebra	Canon LBP-810	EXP
BS2	Biro	Canon LBP-810	EXP
BS3	Sanford Suregrip	Canon LBP-810	EXP
BS4	Sanford Suregrip	Canon LBP-810	EXP
BS5	Schmidt	Apple Laserwriter 16/600PS	EXP
BS6	Pilot BP-S	Epson C1900 Aculaser	HP
BS7	Pilot BP-S	Epson C1900 Aculaser	HP
BS8	Uni SA-S	Xerox Docucolour 3535	Unknown
BS9	Schmidt	Ricoh Aficio 1060 RPCS	Canon
BS10	Papermate	Konika	PPC
BS11	Ohto	Konika	PPC
BS12	Bic	HP Laserjet	Xerox
BS13	Schmidt	HP Laserjet	Xerox
BS14	Reynolds	HP Laserjet	Xerox
BS15	Sanford Suregrip	Panasonic KX-P7305	HP
BS16	Sanford Suregrip	Panasonic KX-P7305	HP
BS17	Zebra	Canon LBP-810	Reflex
BS18	Biro	Ricoh Aficio 1060 RPCS	Reflex
BS19	Staedtler Stick	Minolta magicolour 6100 DeskLaser	EXP
BS20	Staedtler Stick	Lexmark T522	EXP
BS21	Staedtler Stick	Brother MFC-8840D	EXP

TABLE 5 - *Techniques used by forensic document examiners to examine blind samples.*

Forensic Document Examiner (FDE)	**Testing performed**
FDE 1	Visual observation through stereomicroscope & ring light illumination (looking for 'bronzing' of the ink)
FDE 2	Incident lighting at 100x mag. Polarising microscope using reflected light.
FDE 3	Light stereomicroscope.
FDE 4	All intersections examined using polarizing microscope with reflected light, which gave colour contrast between ink / toner.
FDE 5	Visual inspection using magnification. VSC2000 – infrared, ultraviolet examinations.
FDE 6	Oblique and vertical lighting, spectral reflectance.
FDE 7	All binocular microscope (x6.5 – 40), F.O. lighting – specular reflectance.
FDE 8	Microscope

TABLE 6 - *Blind testing results.*

Blind Sample	Correct Order	IR Imaging	FDE 1	FDE 2	FDE 3	FDE 4	FDE 5	FDE 6	FDE 7	FDE 8
BS1	Ink over toner	✓	✓	✓	✗	✓	Inconclusive	Inconclusive	Inconclusive	Inconclusive
BS2	Ink over toner	✓	✓	✓	✓	✓	Inconclusive	✓	✓	✓
BS3	Ink under toner	✓	✓	✓	✓	✓	Inconclusive	✓	✓	✓
BS4	Ink under toner	✓	✓	✓	✓	✓	Inconclusive	✓	✓	✓
BS5	Ink over toner	✓	✓	✓	✓	✓	Inconclusive	✓	✓	✓
BS6	Ink under toner	✓	✓	✓	✓	✓	Inconclusive	✓	✓	✓
BS7	Ink under toner	✓	✓	✓	✓	✓	Inconclusive	✓	✓	✓
BS8	Ink over toner	✓	✓	✓	✓	✓	Inconclusive	✓	✓	✓
BS9	Ink under toner	✓	✓	✓	Inconclusive	✓	Inconclusive	✓	Inconclusive	✓
BS10	Ink over toner	✓	✓	✓	✓	✓	✓	✓	Inconclusive	Inconclusive
BS11	Ink over toner	✓	Inconclusive	✓	✓	✓	✓	✓	✓	Inconclusive
BS12	Ink over toner	✓	✓	✓	✓	✓	Inconclusive	✓	✓	✓
BS13	Ink over	✓	✓	✓	✓	✓	Inconclusive	✓	✓	✓

	toner									
BS14	Ink over toner	✓	✓	✓	✓	✓	Inconclusive	✓	✓	✓
BS15	Ink under toner	✓	✓	✓	✓	✓	Inconclusive	✓	✓	✓
BS16	Ink under toner	✓	✓	×	Inconclusive	×	Inconclusive	✓	✓	✓
BS17	Ink under toner	✓	✓	✓	✓	✓	Inconclusive	Inconclusive	Inconclusive	Inconclusive
BS18	Ink over toner	✓	✓	✓	✓	✓	Inconclusive	✓	✓	✓
BS19	Ink over toner	✓	Inconclusive	✓	Inconclusive	✓	Inconclusive	Inconclusive	Inconclusive	Inconclusive
BS20	Ink over toner	✓	✓	✓	✓	✓	Inconclusive	✓	✓	✓
BS21	Ink over toner	✓	✓	✓	✓	✓	Inconclusive	✓	✓	✓

LIST OF FIGURES

FIG. 1 – *Example of ballpoint pen ink under laser printing showing a) visible image of intersection; b and c) infrared spectra of laser printing and ballpoint pen ink; d and e) infrared spectral images of laser printing and ballpoint pen ink formed by imaging at 1724 cm*$^{-1}$ *and 1584 cm*$^{-1}$.

FIG. 2 – *Example of ballpoint pen ink over laser printing showing a) visible image of intersection; b and c) infrared spectra of laser printing and ballpoint pen ink; d and e) infrared spectral images of laser printing and ballpoint pen ink formed by imaging at 1724 cm*$^{-1}$ *and 1584 cm*$^{-1}$.

FIG. 3 – *Comparison of the quality of infrared spectral images obtained when using different ATR crystals. Visible image of ink over toner shown in (a); (b-c) shows the toner and ink spectral images obtained using a Germanium ATR crystal; (d-e) shows the toner and ink spectral images obtained when using a Zinc Selenide ATR crystal.*

FIG. 4 – *'Live infrared window display' of line intersection (a) produced when using the (b) Germanium and (c) Zinc Selenide ATR crystal.*

FIG. 5 – *Example of ink-ink intersection showing (a) visible image of two intersecting ballpoint pen lines; (b) infrared spectra of two ballpoint pens and (c) infrared spectral image of ink lines formed using the integrated peak intensity under 1584 cm*$^{-1}$.

FIG. 6 – *Infrared spectra of two main types of ballpoint pen ink found in Wang et al. (2001) study (7) (Reprinted, with permission, from the Journal of Forensic Sciences, Vol. 46, Issue 5, copyright ASTM International, 100 Barr Harbor Drive, West Conshohocken, PA 19428).*

FIG. 7 – *Example showing results obtained when different pen pressures used. (a-c) shows the visible light images of Sanford ink over Epson toner using pressure indicators of 10, 7 and 4 respectively; (d-f) the respective toner spectral images formed using the integrated peak intensity under 1724 cm^{-1}; (g-i) the respective ink spectral images formed using the integrated peak intensity under 1584 cm^{-1}.*

FIG. 8 – *Infrared spectral image results for blind sample 16, with the visible light image shown in (a), the toner spectral image formed using the peak intensity at 1724 cm^{-1} in (b) and the ink spectral image formed using the peak intensity at 1584 cm^{-1} in (c).*