The Physical Application of Non-destructive Techniques in Detection the Sequence of Intersecting Gel Ink and Printed Laser Toner Strokes

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DETERMINATION the sequence of intersecting strokes is great evidence in the field of the authentication of documents. Two types of the intersection utilized in preparing the samples of gel ink and toner. The effect of the brands and models of a laser printer, the brands, and colors of gel pen, the nature of the paper surface and the time gap were studied. The samples were examined by two nondestructive techniques, the digital microscope and Raman spectroscopy. The ink properties observed under the digital microscope succeed in determining the sequence of intersection except in case of some colors of some pen brands. Raman spectroscopy combines with a digital microscope to analyze the samples which indistinguishable by the digital microscope. The combination of a digital microscope and Raman spectroscopy is an excellent forensic base to full discrimination the sequence of intersecting strokes regardless of the brands or colors of gel ink.

Keywords: Forensic science, Strokes intersection, Toner laser printer, Gel pen, Cross lines, Raman spectroscopy, Non-destructive techniques.

Introduction

The determination of chronological arrangement of overlapping strokes occupies a large area from the field of forensic document examination. Especially, when utilizing the detection of a sequence of intersecting strokes in the authenticity of documents [1]. Recently, nothing is better than the digital printing, laser printer one of the most type digital printing applied in the production of documents in the last years because it’s fast, easy to use, low price and high quality [2]. Also, one of the most type spreading of writing instruments today is the gel pen due to its soft, rapid drying, good color, low-cost and environmentally friendly [3-7]. Accordingly, The value of detection the sequence of intersecting strokes increase with laser printed documents have signatures of gel pen as letters, wills, loan receipts, and contracts which expose permanently to forged because of the existence of significant financial implications in these documents [1].

The ink used in a laser printer is toner and can be presented in the form of powder particles or dispersed in a liquid. The toner is a mixture consists of organic polymeric resins (main component), pigments, wax and other organic and organometallic additives [8-12]. In general, the luminescent materials in gel ink were often insoluble organic and/or inorganic pigment which gives the inks its sheen colors with other ingredients as solvents(as ethylene glycol), resins,
lubricants, surfactants, corrosion inhibitors, shear thinning agents, emulsifying agents and polymerization agents [3,5,13-15].

Earlier studies showed that three main methods employed to determine the sequence of intersecting strokes, microscopic, spectroscopic and chemical analysis [16]. The microscopic methods are non-destructive, cheap, rapid simple and used to investigate some ink properties at intersection point such as the color, absorption, luminescence and the gloss. Therefore, it gives good results in the case of two strokes differ in its composition and distribution on paper. The microscopic methods are still the first choice to solve the problem of intersecting strokes and widely applied until now although the examination with these methods can be misleading and highly dependent on human interpretation [1,17,18]. Accordingly, forensic document examiners need to stratify other analysis technique as Raman spectroscopy to combine with the microscopic method as a digital microscopic for correct discrimination of intersecting strokes [16,19]. Raman spectroscopy is becoming a tool of major importance in forensic document examination and beneficial in detection the sequence of intersecting strokes due to its easy, non-destructive and fast analysis [20]. The aim of this study is the combination of two non-destructive techniques as a digital microscope and Raman spectroscopy for distinguishable the arrangement of intersecting strokes consist of gel ink and toner.

**Materials and Methods**

**Specimen Preparation**

The samples of intersection which consist of toner laser printer and gel ink were prepared in two types of arrangement. In the first type, the toner strokes printed first then the gel ink strokes crossed over them. In the second type, the gel ink strokes are written first then the toner strokes printed over them [21].

More than one factor perhaps effect on the intersection of gel ink and toner were studied. Seven brands and models of a laser printer as listed in Table 1 were utilized due to the variations in chemical composition of toner from brands and models to other [22].

Also, due to the impact of the gel ink color and the great alterations in its composition on determining the sequence of intersecting strokes, eleven different brands of a gel pen with different colors which commonly used in delay cases work as listed in Table 2 were applied in sample preparation [16].

In addition, the effect of nature of paper surface was investigated through prepared the samples of intersection on three different types of paper surface as listed in Table 3.

Lastly, to examine the influence of a time gap between the first and the second strokes on their intersection, the second stroke is written or printed on the first stroke after the time gap is 5 mins, 2 hrs, 10 days, 20 days and 30 days. The samples of intersection left for the same time gap at room temperature and kept under similar climatic conditions before the examination and analysis process.

**The Digital Microscope**

Detection the intersection of gel ink and toner depend on investigation the physical properties for both. The digital microscope examines the effect of intersection on some gel ink and toner properties as the specular reflection, the ink and toner gloss, gaps and spreading. The samples of intersection captured with MDA1300 (China) digital microscope which has magnification 20-200X. The lighting system of a digital microscope consists of eight lighting lamps of the annular light source on the head of a microscope and directed in the same direction. The digital microscope placed perpendicularly to the sheet of paper and zoom on the intersection point. Zooming and lighting system present in digital microscope show some ink properties as specular reflection, gloss, gap,

**TABLE 1. List of printer brands are used in this study.**

<table>
<thead>
<tr>
<th>Brand of printer</th>
<th>Model of brand</th>
<th>Color of ink</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hp LaserJet</td>
<td>2300dn</td>
<td>Black</td>
</tr>
<tr>
<td>Hp LaserJet</td>
<td>P1005</td>
<td>Black</td>
</tr>
<tr>
<td>HP LaserJet</td>
<td>1018</td>
<td>Black</td>
</tr>
<tr>
<td>LEXMARK</td>
<td>E350d</td>
<td>Black</td>
</tr>
<tr>
<td>EPSON aculaser</td>
<td>M 2000</td>
<td>Black</td>
</tr>
</tbody>
</table>

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and the distribution of ink at point intersections. The images observed and captured directly on the monitor of the computer [7, 17, 21].

**Raman Spectroscopy**

Raman spectrum was measured using Senterra spectrometer attached with Bruker microscopes. A 50X objective lens was used to concentrate the laser beam spot on the intersection point before beginning Raman analysis. A 50X magnification objective obtained a diameter laser spot of about 0.2 µm. Laser giving the 785 nm wavelength served as a source of the monochromatic light in this study. The numbers of sample scans were ten measurements performed at the intersection point with laser power 100 mW and integration time 1 sec to get the greatest permissible intensity of peaks. The selected points for measuring the spectrum were manually and randomly choice. The suitable temperature for cooling of CCD (Charge Coupled Device) detector during measurement is -65°C. When the laser beam powered on and directed at the intersection point, the interaction between the laser beam and intersection point takes place and the scattered curve is generated [19, 20].

**Results and Discussion**

*Under The Digital Microscope*

### TABLE 2. List of gel pen brands used in this study.

<table>
<thead>
<tr>
<th>Brand of writing instruments</th>
<th>Type of writing instruments</th>
<th>Color of ink</th>
</tr>
</thead>
<tbody>
<tr>
<td>MICRO, G-388, China, 0.5 mm</td>
<td>Rollerball Pen</td>
<td>Black, red and blue</td>
</tr>
<tr>
<td>Uni-ball gel impact 1 mm</td>
<td>Rollerball Pen</td>
<td>Black, red and blue</td>
</tr>
<tr>
<td>Pentelenergel BL 107</td>
<td>Rollerball Pen</td>
<td>Black, red and blue</td>
</tr>
<tr>
<td>Flair concorde</td>
<td>Rollerball Pen</td>
<td>Black, red and blue</td>
</tr>
<tr>
<td>SAKRM.A.M (GL – 1220)</td>
<td>Rollerball Pen</td>
<td>Black, red and blue</td>
</tr>
<tr>
<td>BOIL G388xincai</td>
<td>Rollerball Pen</td>
<td>Black, red and blue</td>
</tr>
<tr>
<td>3ADIMOND</td>
<td>Rollerball Pen</td>
<td>Black, red and blue</td>
</tr>
<tr>
<td>ROTO G-Plus</td>
<td>Rollerball Pen</td>
<td>Black, red and blue</td>
</tr>
<tr>
<td>CLARO SIGMA</td>
<td>Rollerball Pen</td>
<td>Black, red and blue</td>
</tr>
<tr>
<td>LINCglycer</td>
<td>Rollerball Pen</td>
<td>Black, red and blue</td>
</tr>
<tr>
<td>POP EURO</td>
<td>Rollerball Pen</td>
<td>Black, red and blue</td>
</tr>
</tbody>
</table>

### TABLE 3. List of different types of paper used in this study.

<table>
<thead>
<tr>
<th>Nature of paper surface</th>
<th>Brand of paper</th>
<th>Size of paper</th>
<th>Weight of paper (g/m²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>non-coated</td>
<td>MultiOffice</td>
<td>A4</td>
<td>80</td>
</tr>
<tr>
<td>coated</td>
<td>Antalis</td>
<td>A4</td>
<td>80</td>
</tr>
<tr>
<td>smooth</td>
<td>Antalis</td>
<td>A4</td>
<td>80</td>
</tr>
</tbody>
</table>
forming a colored specular reflection differs than the toner gloss. Therefore, the sequence of intersecting strokes easily distinguishable as seen in Fig. 2.

The red color of the remain pen brands listed in Table 4 forming a transparent specular reflection similar to the toner gloss and interference with it. Accordingly, the sequence of intersecting strokes indistinguishable and difficult to determinate as in Fig. 3 [24].

In the second type, when the brands of gel pen listed in Table 5 passed over the toner, an incomplete (partially) specular reflection appeared.

This is on account of the luminescent materials almost diffuse only on edges of the intersection point. The composition of ink in the above brands is a mixture of luminescent and non-luminescent materials and the non-luminescent materials have a masking effect which inhibits overall spreading of luminescence materials as in Fig. 4 [16].

On the other hand, when the toner printed over all brands of gel pen (all colors), occurs absence of the specular reflection. The thick and opaque powder layer of the toner covers the luminescence materials exist in ink and prevent the formation of specular reflection as in Fig. 5 [7,18,23].

Gloss of Ink

The second property observed under the digital microscope is the ink and toner gloss. The gel ink has glossy properties due to the presence of the luminescence materials in its composition [4] and the toner has the same properties due to the presence of wax in its composition [12,24-26].

In the first type of intersection when ink crossed over the toner, the ink gloss is continued and the toner gloss is discontinued. The ink gloss is continued because the black background of toner helps it to appear as in Fig. 6.

The toner gloss is discontinued because the luminescence materials present in gel ink composition covers the gloss of toner and prevents showing it at an intersection point as in Fig. 7.

In the second type of intersection when the toner is printed over the gel ink, the gloss of both ink and toner is discontinued as in Fig. 8.

The discontinuity gloss of ink because the black thick layer of toner block the luminescence materials present in the ink at the intersection point [27]. Also, the discontinuity gloss of toner because the ink written first penetrates and absorbed by cellulose fiber and interact with the photosensitive roller of the printer causing a

| TABLE 4. List of gel pen brands composes complete (full) specular reflection. |
|----------------------------------------|------------------------------|
| **Brands of gel pen**                  | **Color of ink**             |
| Uni-ball gel impact 1 mm               | Black, red and blue          |
| ROTO G-Plus                            | Black, red and blue          |
| CLARO SIGMA                            | Black, red and blue          |
| LINCglycer                             | Black, red and blue          |
| POP EURO                               | Black, red and blue          |

| TABLE 5. List of gel pen brands composes an incomplete (partially) specular reflection. |
|----------------------------------------|------------------------------|
| **Brands of gel pen**                  | **Color of ink**             |
| MICRO, G-388, China, 0.5 mm            | Black, red and blue          |
| Pentelenergel BL 107                   | Black, red and blue          |
| Flair concorde                         | Black, red and blue          |
| SAKRM.A.M (GL – 1220)                  | Red                          |
| BOIL G388xicnai                        | Black, red and blue          |
| 3ADIMOND                               | Black, red and blue          |
Fig. 1. The complete specular reflection when (a) blue Uni-ball gel and (b) black ROTO G-Plus inks crossed over the toner under the digital microscope.

Fig. 2. The colored specular reflection when (a) red Uni-ball gel, (b) black LINC glycer, and (c) blue LINC glycer inks crossed over the toner under the digital microscope.
disturbance in the electrostatic process to prevent the gloss of wax from fixing at the intersection point [7,18,23].

**Gap of Ink**

One of the most important properties observed under the digital microscope is a gap of gel ink and toner which depends on the type of fixing method of the first stroke. Firstly, the gel ink gaps take place when the ink intersects the toner. The toner fixed first on the paper surface has a higher level than the paper level, therefore the ink jump before and/or after the intersection point when written over toner as in Fig. 9 [12,24-26].

Secondly, the toner gaps occur when toner intersects the gel ink. The ink is written first led to a disturbance in the electrostatic process due to interacting with the photosensitive roller of the laser printer or causing a change in the surface of paper due to partially melt with the application of a high temperature in a laser printer. Therefore, the toner gaps appear at the intersection point during the printing process and the ink color shows up through the toner gaps as in Fig. 10 [27].

**Spreading of Ink**

The last physical characteristic observed under the digital microscope is the gel ink and toner spreading. The ink spreading over the toner in two ways, the first way, the complete ink spreading happen at the intersection point when the brands of gel pen listed in Table 4 crossed over the toner. The luminescence materials exist in the ink composition almost fill all parts at the intersection point and completely accumulate on the toner surface stroke as in Fig. 11.

In a second way, an incomplete ink spreading at the intersection point when the brands of gel pen listed in Table 5 crossed over the toner. The composition of ink in these brands are a mixture of luminescent and non-luminescent materials and the non-luminescent materials have a blocking effect inhibits overall spreading of luminescence materials as in Fig. 12.

An incomplete toner spreading over the gel ink occurs at intersection point as in Fig. 13.

The ink is written first led to a disturbance in the electrostatic process due to interacting with
Fig. 4. The partially specular reflection when (a) blue BOIL G388 xin cai, (b) black MICRO, G-388 0.5 mm, (c) red 3A DIMOND and (d) red Pentel energel BL 107 inks crossed over the toner under the digital microscope.
Fig. 5. The absence of specular reflection when the toner is over (a) blue Uni-ball Gel impact and (b) black MICRO, G-388 0.5 mm inks under the digital microscope.

Fig. 6. The continuity gloss of ink when (a) blue Uni-ball Gel impact 1 mm and (b) black ROTO G-Plus crossed over the toner under the digital microscope.

Fig. 7. The discontinuity gloss of toner when (a) blue LINCglycer and (b) black ROTO G-Plus inks crossed over the toner under the digital microscope.

Fig. 8. The discontinuity gloss of toner and ink when the toner is over (a) black ROTO G-Plus and (b) red SAKRM.A.M (GL – 1220) under the digital microscope.
Fig. 9. The ink gaps when (a) black, (b) blue Uni-ball Gel impact, (c) blue and (d) red Pentelenergel BL 107 inks crossed over the toner under the digital microscope.
Fig. 10. The toner gaps when a toner is over (a) black 3ADIMOND, (b) red Pentelenergel BL 107, (c) black Uni-ball Gel impact and (d) blue CLARO SIGMA under the digital microscope.
Fig. 11. The complete ink spreading when (a) black POP EURO and (b) red Uni-ball Gel impact crossed over the toner under the digital microscope.

Fig. 12. An incomplete ink spreading when (a) blue BOIL G388xincai and (b) black MICRO G-388 inks crossed over the toner under the digital microscope.

the photosensitive roller of the laser printer or causing a change in the surface of paper due to partially melt with the application of a high temperature in a laser printer [28].

Raman Spectroscopy

Raman spectroscopy used to determine the sequence of the intersecting strokes by comparison of the peaks nature in the scattered curve recorded from gel ink, toner and their intersection point. The similarity of the spectrum shape and peak values in the scattered curve comes from an intersection point and one of two intersected gel ink and toner indicate that this ink is executed later [19].

Toner and two brands of gel ink (Uni-ball gel impact 1 mm and MICRO G-388 black ink) are subjected to Raman analysis in two types of intersection. Firstly, when the gel ink crossed over the toner, the spectrum of their intersection point corresponded to the spectrum of pure gel ink and has a high degree of similarity in shape to it as in Fig. 14 [20].

This is due to in case of Uni-ball gel impact blue ink the luminescence materials completely accumulated on the surface of toner as in Fig. 1(a) [17,22,27]. While in the case of MICRO G-388 black ink although the similarity in shape between the spectra of ink and ink crossed over the toner as in Fig. 14, a little change in the value of peaks was present in the scattered curve. Beside the existence of luminescence materials on the edges of the intersection point, other non-luminescence materials compose a new mixture with other parts of the intersection point, therefore a new spectrum differs than the spectrum of pure gel ink is formed as in Fig. 4(b)[29].

Secondly, when the toner printed over the gel ink, the spectra of toner and toner over ink are similar in the shape because they consist of very narrow peaks but the main peaks of the two spectra differ in its wavenumber and height as in Fig. 15 [19,20,23]. The spectrum coming from the intersection point generated due to the formation of a new mixture result from the
Fig. 14. Raman spectra of (a) Uni-ball, (b) Uni-ball crossed over the toner, (c) MICRO G-388 and (d) MICRO G-388 crossed over the toner.

Fig. 15. Raman spectra of (a) toner, (b) toner crossed over Uni-ball and (c) toner crossed over MICRO G-388inks.

Fig. 16. Raman spectra of (a) red CLARO SIGM and (b) red CLARO SIGM ink crossed over the toner.

Fig. 17. Raman spectra of (a) toner and (b) the toner printed over red CLARO SIGM ink.

Fig. 18. The gloss of blue MICRO G-388 ink over a toner of (a) Xerox Phaser 6700, (b) HP 2300, (c) HP 1018 and (d) Epson M 2000 under the digital microscope.
Fig. 19. The spreading of CLARO SIGMA ink over the toner on (a) coated, (b) non-coated and (c) smooth papers under the digital microscope.
Fig. 20. The specular reflection when red Uni-ball Gel impact crossed over a toner after (a) 5 mins, (b) 2 hours, (c) 10 days and (d) 30 days under the digital microscope.
As we saw before, Raman spectroscopy gives good results in the case of blue and black gel ink crossed over the toner. Hence Raman spectroscopy can contribute in solving the problem of determining the sequence of the red color of pen brands listed in Table 4 (except in case of Uni-ball gel impact) crossed over the toner which indistinguishable by a digital microscope. Raman spectroscopy analyzed two intersection types of CLARO SIGM red color and toner. Firstly, when red ink crossed over the toner, the spectrum at the intersection point is similar to the spectrum of pure red ink as in Fig. 16.

Secondly, when the toner printed overred ink the spectrum at the intersection point is related and similar in shape to the spectrum of pure toner as in Fig. 17.

Totally, the combination of two non-destructive techniques consists of a digital microscope and Raman spectroscopy utilized as a scientific rule in discrimination the sequence of intersecting strokes resulting from the gel ink and toner regardless the colors of gel ink.

More than one factor studied to determine its effect on the intersection of gel ink and toner. The results of the first factor were studied show that the changing of brand and model of printer has no variations as observed in Fig. 18. This is may be because the main components of the toner of all laser printers are similar to each other regardless of their brands and models. Also, the applied technique used in the toner fixing on the paper surface in all laser printers is the same.

Also, no alterations during the investigation of samples due to the variation in the nature of the paper surface as in Fig. 19.

The composition of the gel ink, toner and the three types of paper surface were used not to prevent the absorption of gel ink and the fixing of toner regardless of the types of paper surface. Additionally, no effects take place due to the application of five-time gaps between the first and the second strokes as in Fig. 20. This is perhaps the five-time gaps are enough to the first stroke to absorbed or fixed and distributes normally on the paper surface without any
Blind Testing

The blind samples resulted from the intersection of toner and gel pen inks prepared by volunteer person. The sequence of toner and gel ink strokes was unknown for some authors before examination and known to volunteer person only. The blind samples examined by authors to detect the error rate present in using the combination of a digital microscope and Raman spectroscopy. The results recorded by authors compared with the key of blind samples prepared by a volunteer person. Firstly, with respect to Raman spectroscopy, it successful in determining the sequence of strokes intersection. Secondly, with respect to the digital microscope, all authors give correct answers for the sequence of intersecting strokes in all blind samples except in the case of five samples of red ink crossed over toner. All results obtained by authors are listed in Table 6.

This blind testing shows that the limitations of using the digital microscope alone to determine the sequence of red ink crossed over toner due to optical illusions and varied perceptions of a human. Therefore, the combination of Raman spectroscopy which producing curves based on chemical differences between gel ink and toner with a digital microscope has high advantages and arrived at one-hundred percent in the determination of a sequence of intersecting strokes. Accordingly, these results are useful in the field of forensic document examination.

Conclusion

Two nondestructive techniques, the digital microscope and Raman spectroscopy used to determine the chronological order of intersecting heterogeneous strokes consist of gel ink and toner. The four physical properties investigated under the digital microscope the specular reflection, the ink gloss, gaps and spreading give good results to the sequence of intersecting strokes, except in case of the red color of some pen brand. The results of Raman spectroscopy at intersection point related to the ink or toner written or printed later regardless its brands or colors. Therefore, the combination of the digital microscope with Raman spectroscopy easily determines the chronological order of intersecting strokes.

References

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THE PHYSICAL APPLICATION OF NON-DESTRUCTIVE TECHNIQUES FOR DETECTING CROSSOVER POINTS OF DOT WATERMARKS FOR LASER PRINTER INK AND GELATIN INK, AMR HANI ALI ALI 1, YASER MOHAMED MASTROFI 2, AHMED ALI ABDULAZIZ 2, ABDUL-WAHAB AHMED ABU MANDJ 1, SAMIA ABDELRAHMAN EL-SEIF 3, AHMED MOHAMED ABDULLAH ABDEL QADIR.

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The physical application of non-destructive techniques for detecting crossover points of dot watermarks for laser printer ink and gelatin ink, understanding the relationship between authentic and replicated documents in the field of forensic medicine and the circulation of authentic documents. Replication process includes reading the edges of this document, then endorsing it to signify their agreement on what is contained in this document. This process may be accompanied by a process of alteration or replacement of some or all of what is written in it as a means of acquiring a signature of a party on it. The existence of the signature can coincidentally interact with the newly inserted or replaced data (false data of the inserted or replaced data) later with the presence of existing signatures and can serve as the only means to determine the false. In this study, samples were prepared that contain two types of crossover point between the ink and gelatin ink and gelatin ink, and the study of some factors affecting the detection of this crossover, such as the laser printer mark, gelatin ink mark, type of paper on which the samples are prepared, the time interval between the first and second samples. In the case of the sample examination, two methods were used to determine the presence of the forgery, the first is the digital microscope, the second is the spectroscope. The digital microscope succeeded in determining the crossover points except for certain colors of certain gelatin ink marks. Therefore, the spectroscope was used to order the gelatin ink marks and the ink crossover points with the gelatin ink marks of any color. Therefore, combining the digital microscope with the spectroscope in determining the crossover points of gelatin and ink marks for gelatin ink is a new, promising method in the field of document examination to ensure its authenticity or its falsity.