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Some of the Physical and Chemical Characterizations Applied for the Laser Printers toner and Ballpoint Pen Inks to Determine the Sequence of their Intersections

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IN THE forensic document examination, the decision of the historical order of intersection strokes between the toner and ballpoint inks is paramount a challenge issue. Two kinds of intersections are produced on three different kinds of papersheets. The digital microscope and Raman spectroscopy are two non-destroy techniques used to decide, these sequences via cross lines. Four physical properties obtained at the crossing point under the digital microscope were, specular reflection, ink gloss, ink spreading and gap. The results of the digital microscope detect that the two kinds of crossing strokes except the red color of ink crossed over the toner. The results of Raman spectroscopy manifested that the crossing point similar to the pure ink executed later. The complementary techniques of Raman spectroscopy and the digital microscope were necessary to achieve a complete determination of the stroke's intersection sequences of the toner and ballpoint ink.

Keywords: Forensic document examination, Strokes intersection, Ballpoint inks, Tonerlaser printer, Raman spectroscopy, Digital Microscope.

Introduction

Nowadays, digital printing has widely spread for the formation of various documents using different printing techniques. The laser printer, technique was one of the most types of digital printing utilized for printing documents in the last years ascribed to the lowest cost, high quality, fast, easy to use [1]. In the last decade, the developments in the technology of pen ink compositions and characteristics have expanded swiftly. Although, the ballpoint pens are still very famous use and widespread kind of writing device in various documents[2-4]. In addition, laser printouts always used with ballpoint pen ink in daily routine, personal business and official work to form different documents like letters, wills, loan receipts, bank checks, medical certificates, working papers and contracts that can have significant financial implications.The dry ink utilized in the laser printers is called toner and it can be present in the form of powder particles or diffused in the liquid[5-11].

Modern ballpoint pen inks possess many materials to improve the characteristics of the ink. Clearly, the essential component in ink is the coloring substance as pigments, dyes or a mixture of them [3,4,12-16]. The intersection of heterogeneous strokes consisting of laser printout strokes with other writing devices as ballpoint pens must be investigated and studied as a great

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challenge in the forensic document examination [9,17,18].

In the earlier studies, two essential methods were employed to decide the order of crossing strokes, microscopic, and chemical analysis [19]. The microscopic methods investgate some characteristic of ink at the crossing points such as color, absorption, luminescence, and gloss. Therefore, it gives excllent facts in the case of two ink strokes of different structure and diffusing on the paper surface as ballpoint pen ink and the toner [17]. Although microscopic methods are still the first choice to solve the problem of crossing lines and they widely used because it's non-destructive, cheap, rapid and simple, but, they have some disadvantages as misleading and highly dependent on human interpretation [11]. Therefore, it is a maximum fair to apply microscopic methods as the first stage in strokes intersection and to combine with other chemical analysis methods such as Raman spectroscopy [7,20]. Raman spectroscopy is a tool with great significance in forensic checking and useful in the crossing lines due to it's soft, non-destroy (samples checked directly without preparation) and rapid analysis [21].

The purpose of this study is to detect all candidate brands of ballpoint pen ink and toner having the same behavior at the crossing lines and to compare between the observed data by using complementary techniques that consist of the digital microscope and Raman spectroscopy.

Materials and Methods

Specimen preparation

The samples prepared in the following way to detect the sequence of the intersecting strokes consist of the ballpoint pen ink and the toner. In the first arrangement of an intersection, the toner strokes printed first, afterward ink strokes were crossed them. In the second arrangement, the ink strokes are written first, then the toner strokes printed over them. The crossing of the toner and ballpoint pen ink is a moveable operation influenced by many chemical and physical variables that define how the ink and toner will spread and react with each other on the paper.

More than one factor may change the observed results, the first is the variation in printers brands and models, using more than one brand of the laser printer may effect on the interaction of intersecting strokes due to the variation in the chemical installation of the toner from brand to another one. Therefore, the printers of different and the same brand with different models listed in Table 1 used in this study.

The second factor is the variation in writing instrument brands and the ink color. Therefore, eleven different brands of ballpoint pen inks with different colors which commonly used in delay cases work were listed in Table 2.

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

The printer brands	Model of brand	The ink color
Hp LaserJet	2300dn	Black
Hp LaserJet	P1005	Black
HP LaserJet	1018	Black
LEXMARK	E350d	Black
EPSON Aculaser	M 2000	Black

TABLE 2. List of ballpoint pen brands used in this study.

The ballpoint pen brands	The writing instrument types	The ink color
STAEDTLER stick 430 M	Ballpoint pen	Black, red and blue
MY-TECH	Ballpoint pen	Black, red and blue
PRIMA FORMA	Ballpoint pen	Black, red and blue
BIC	Ballpoint pen	Black, red and blue
REBNOK dzire	Ballpoint pen	Black, red and blue
FABER CASTELL GRIP	Ballpoint pen	Black, red and blue
Uni-laknock MITSUBISHI	Ballpoint pen	Black, red and blue

The third factor is the paper surface properties which may affect how the two inks will distribute across the paper. To study the effect of the nature of paper surfaces, the samples of intersections were prepared on three different type'scoated, non-coated and smooth paper.

The later important factor is the time gap between the toner and ballpoint pen inks, which directly correlated to the ink drying processes. The second stroke was written or printed on the first stroke after the time gap is 5 mins, 2 hrs, 10 days, 20 days and 30 days, respectively. The resulting samples were left at the same time of the gap at room temperature and kept under similar climatic conditions before the examination and analysis process.

The digital microscope

The intersection specimens of toner and ballpoint pen inks were examined and captured with MDA1300 (China) digital microscope that has a magnification 20-200X. The investigation occurs by employ lightening system consists of eight flood light on the microscope head and directed in one direction. The digital microscope was fixed perpendicularly on the paper surface and zoom-in the meeting point. Zooming and lightening systems manifest distribution of dyes/ pigments in ballpoint pen ink and the toner gloss at the point of intersection. The images of investigation were observed and recorded directly on the computer monitor [22].

Raman spectroscopy

The measurment of Raman spectroscopy takes place by Senterra spectrometer connected to Bruker microscopes. Before the formation of Raman spectrum, the laser beam directed on the intersection point by 50× objective lens. A 50X magnification objective gained the diameter of laser spot nearly 0.2 µm. The laser wavelength is 785 nm applyied as a monochromatic light exporter 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 cooling of Charge Coupled Device (CCD) detector during the measurement happen at -65°C. When the beam of laser directed on the point of intersection, the scattered curve is generated after the reaction between the beam of laser and the ink and toner at the point of intersection [21,23].

Results and Discussion

Under the digital microscope

The determination sequences of intersecting strokes occurred depending on the differences in the composition of both the ballpoint pen inks and toner and the subsequent modification of a second ink due to the presence of the first ink [15,18]. The following physical characteristics are observed at the intersection point of the toner and ballpoint pen inks under the digital microscope:

The specular reflection

The first physical property studied under the digital microscope is the specular reflection which considered a mirror-like reflection from the surface at the intersection point. The bright specular reflection results from passing the tip of a soft-point ballpoint pen that may serve as a polisher of existing writing. The specular reflection observed at the intersection point when ballpoint pen ink stroke of all colors over the toner strokes. The pigments and/or dyes presented in the ink composition almost completely accumulate and fill all parts on the intersection point as in Fig. 1 (A) [24]. All brands of ballpoint pen inks (all colors) have the same manner for the absence of specular reflection when the toner passed over them as in Fig. 1(B). This is due to the fact that the toner made a thick and opaque layer which covered the specular reflection of inks[18,22].

The InkGloss

The second important physical property observed under the digital microscope is the ink and toner gloss. The toner has a gloss property due to the presence of wax in its composition [10,25–27] and ballpoint pen inks also have the same property due to the presence of luminescent substances in its composition [4]. The gloss of both toner and ballpoint pen ink discontinued when the toner intersects ink as in Fig. 2 (A). The discontinuity toner gloss that resulting from the solvent present in ink may interact with the photosensitive roller of laser printer causing a disturbance in the electrostatic process to prevent the glossy wax from fixing at the intersection point [28]. The discontinuity ink gloss because the toner fixed on paper surface as a black thick layer which covers and prevents the ink gloss from showing at the intersection point [18,22]. When the ink intersected toner, the toner gloss discontinued and the ink gloss is continued as in Fig. 2 (B). The toner gloss is discontinued due to the ink gloss block it at the intersection point. The ink gloss is continued because the toner black background helps the colors of ink to show its gloss and covers the toner gloss[28].

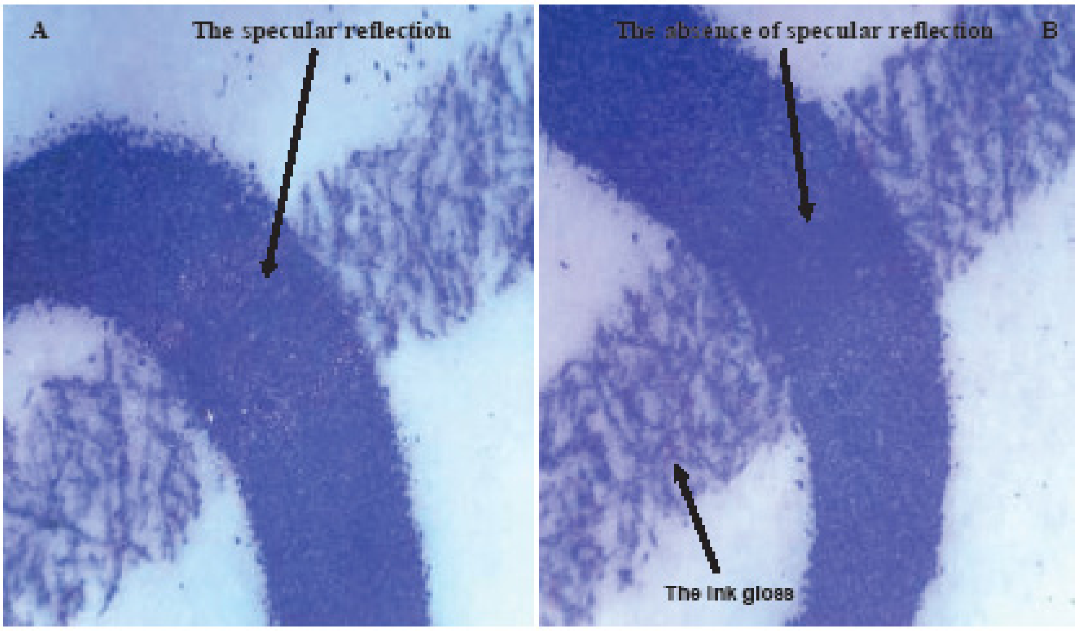

Fig. 1. (A) The presence of specular reflection when black BIC is crossed over the toner and (B) the absence of specular reflection when toner is printed over black BIC ink under the digital mi-croscope.

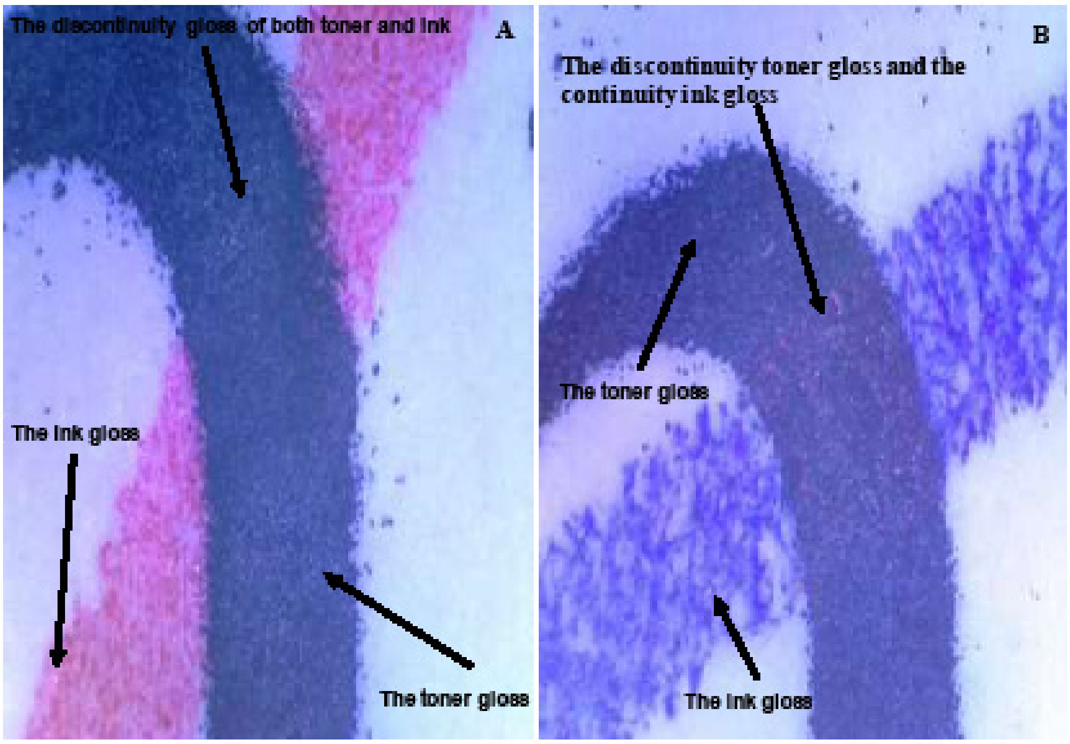

Fig. 2. (A) The discontinuity gloss of both toner and ink when the toner is printed over red STAEDTLER stick ink and (B) the discontinuity gloss of toner and continuity gloss of ink when blue BIC is crossed over toner under a digital microscope

The ink gap

Another important property is called the ink and toner gaps that arise in two types of the intersection. The ink gaps observed when the ink crossed the toner. The toner fixed on the paper surface at a higher level than the paper level. Therefore, during the writing process, the ink may be jumped before and/or after the toner to pass over it causing a gap in the ink as in Fig. (3 A)[24-27]. The toner gaps observed when toner crossed ink. This is due to the ink is written first and may be led to the disturbance in the electrostatic process due to interaction with the photosensitive roller of the laser printer or the change in the paper surface due to partially melt with the application of high temperature in a laser printer. Therefore, some toner gaps seem during the toner fixing at the intersection point and the ink color may appear throughout the toner gaps as in Fig. 3 (B) [28].

The ink spreading

The last physical property studied under the digital microscope is the ink and toner spreading. The ink spreading over toner takes place as in Fig. 4 (A). The complete ink spreading occurs because pigments and/or dyes present in an ink composition almost accumulate and fills all parts at the intersection point.The toner spreading over ink happens as in Fig. 4 (B). An incomplete toner spreading and the color of ink appear from toner gaps [26].

The net results of four physical properties studied under a digital microscope almost determine the sequence of intersecting strokes of ink and toner, particularly for blue and black ink colors. This is attributed to the black background of toner helps the blue and black colors of ink to be clear. The red color of pen brands listed in Table 3 over toner composes the yellow specular reflection which differs than the transparent toner gloss as in Fig. 5 (A). Therefore, the sequence of intersecting strokes easily to detect.

The red color of ballpoint pen brands listed in Table 4, over the toner, compose the transparent specular reflection which interferes with a gloss of toner as in Fig. 5 (B). Therefore, the

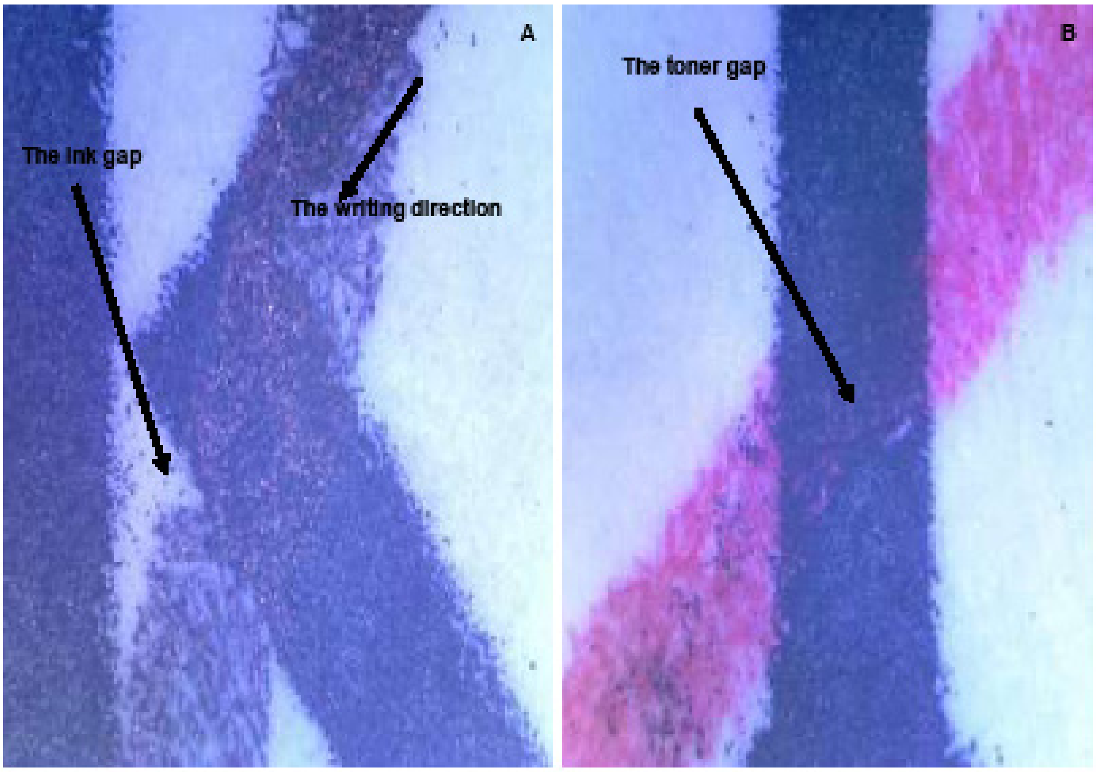

Fig. 3. (A) The ink gap after intersection point when black Uni-laknock MITSUBISHI is crossed over the toner (B) the toner gap at the intersection point when the toner is printed over red MY-TECH under a digital microscope.

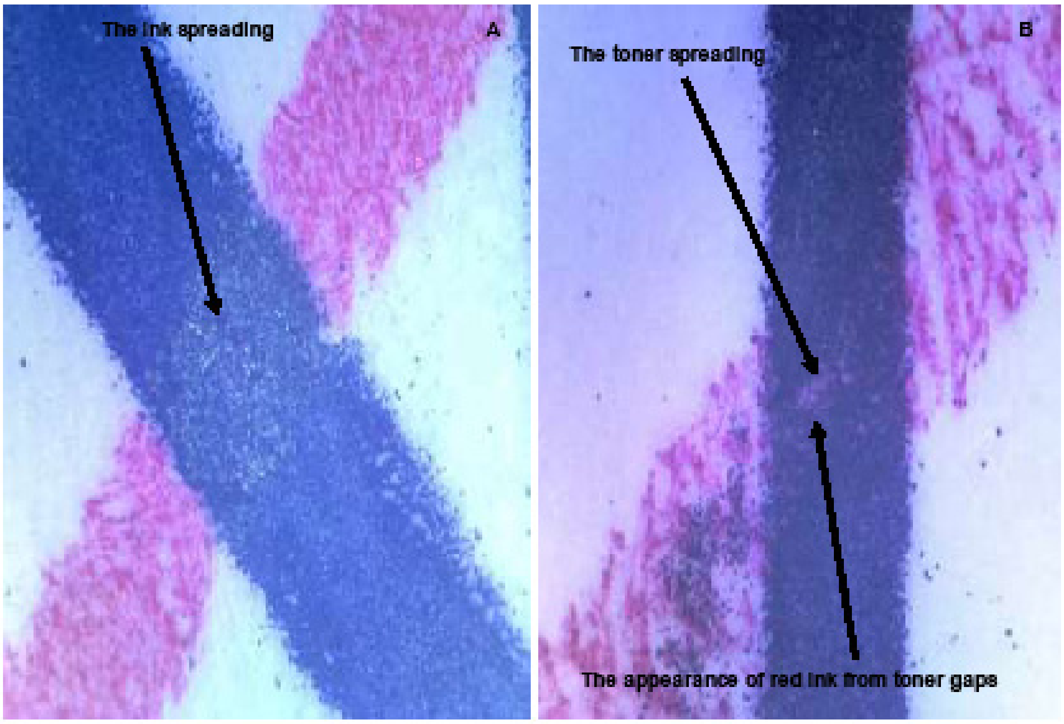

Fig. 4. (A) The red Uni-laknock spreading over a toner and (B) the toner spreading over the red PRIMA FORMA under a digital microscope.

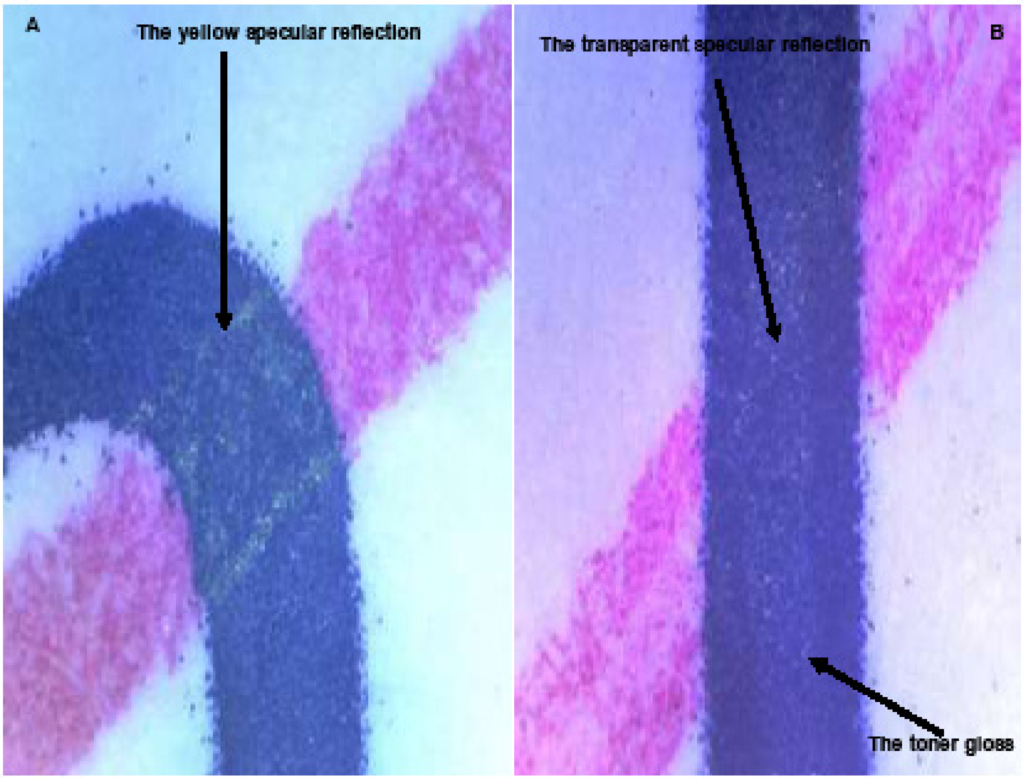

Fig. 5. (A) The yellow specular reflection when red FABER CASTELL over a toner and (B) the trans-parent specular reflection when red STAEDTLER stick ink over a toner under a digital micro-scope.

TABLE 3. List of ballpoint pen brands composes a yellow specular reflection.

The ballpoint pen brands	The writing instrument types	The ink color
FABER CASTELL GRIP	Ballpoint pen	Red
Uni-laknock MITSUBISHI	Ballpoint pen	Red

TABLE 4. List of ballpoint pen brands composes a transparent specular reflection.

The ballpoint pen brands	The writing instrument types	The ink color
STAEDTLER stick 430 M	Ballpoint pen	Red
MY-TECH	Ballpoint pen	Red
PRIMA FORMA	Ballpoint pen	Red
BIC	Ballpoint pen	Red
REBNOK dzire	Ballpoint pen	Red

determination of the sequence of the intersecting strokes becomes difficult to detect.

However, it mostly benefits to apply microscopic methods as the first step in strokes intersection and then combine with other non-destructive chemical analysis method as Raman spectroscopy that can give full detection to the sequence of the intersecting strokes [7,20].

Raman spectroscopy

Determination the sequence of the intersecting strokes by Raman spectroscopy occurs by comparison of the peaks, nature in the Raman spectrum recorded from two pure inks and the intersection point. The similarity of the spectrum shape and peak values in the Raman spectrum comes from an intersection point and one of them indicate that this ink executed later [19]. Two kinds of ballpoint pen ink (blue FABER CASTELL, red STAEDTLER stick) and laser toner are analyzed by Raman spectroscopy in two kinds of the intersections. In the first kind, when the ballpoint pen ink is written over the toner, the spectrum at the point of their intersections related to the spectrum of pure ink. The Raman spectra of pure ink and ink crossed over the toner almost have a high degree of similarity in the shape and value of the main peaks as in Fig. 6. This is may be because of the presance of pigments in ballpoint pen ink that are completely accumulated on the toner stroke and fill all toner gaps. Therefore, the Raman spectrum at the intersection point seems identical with the spectrum of pure ballpoint pen ink.

In the second kind, when the toner crossed over the ballpoint pen ink, it is clear that the spectra of toner and the toner printed over the ink are similar in the shape because of both consists of very narrow peaks. But, the main peaks of the two spectra differ in its wavenumbers and height as in Fig. 7.

This is due to the appearance some inks from toner gapsexsist on the top layer as in Fig. 4 (B), perhaps composes a new mixture of toner and ink which led to the formationof a new spectrum of some different peaks than pure toner [17]. As we have seenpreviously, Raman spectroscopy gives a good result in the case of blue color ink crossed toner. Also, used in determining the sequence ofred color of ink listed in Table 5 crossed over the toner which not detected by the digital microscope. Totally, Raman spectroscopy is a complementary technique with a digital microscope to determine the order of crossing lines resulting from all colors of ballpoint pen ink and the toner.

The changing of the brand and model of printer led to no variations on the results observed at the intersection point as in Fig. 8. This is may be related to the main constituents of the toner for all laser printers are similar to each other regardless of their brands and models. Also, the applied method for printing the toner on the paper surface in all used printers is one.

Furthermore, the variation in nature of the paper surface studied when blue Uni-laknock MITSUBISHI ink crossed over the printed toner as in Fig. 9. No changes in the observed results because the structure of the toner, ballpoint pen inks and used three paper surfaces that do not prevent the fixing of toner and absorption of the ballpoint pen ink on the surface of all paper and cellulose fibers.

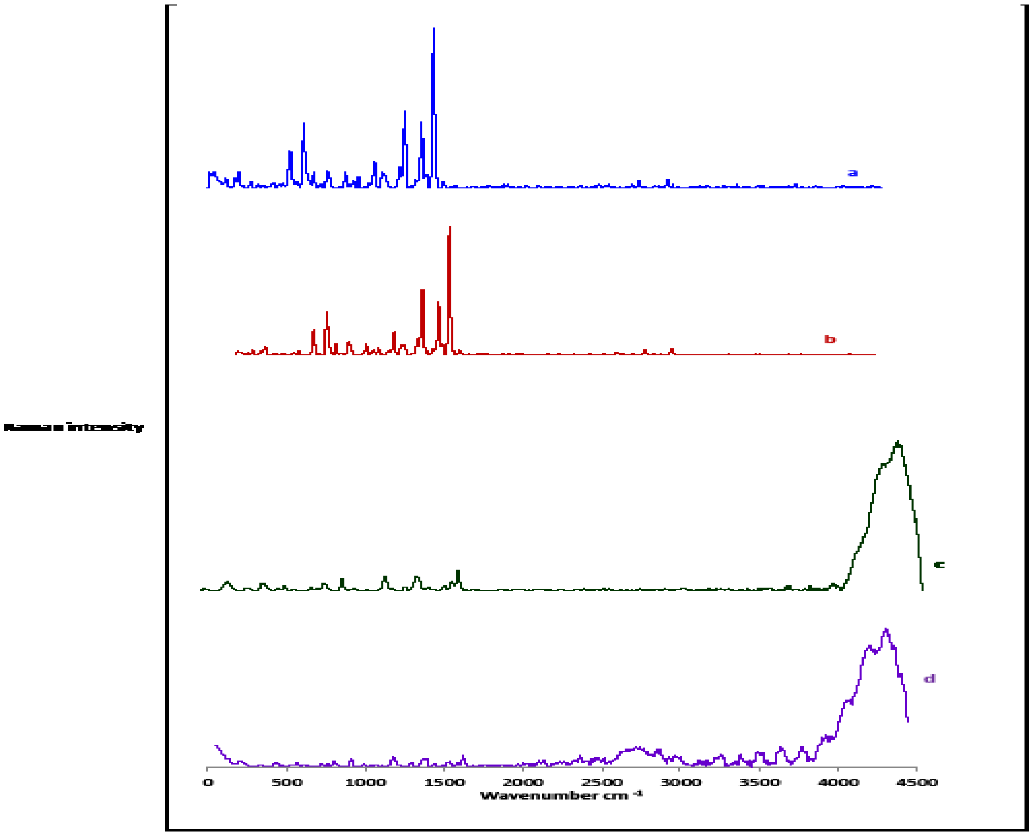

Fig. 6. Raman Spectra of (a) Red STAEDTLER, (b) Red STAEDTLER stick + toner, (c) Blue FABER CASTELL and (d) Blue FABER CASTELL + toner.

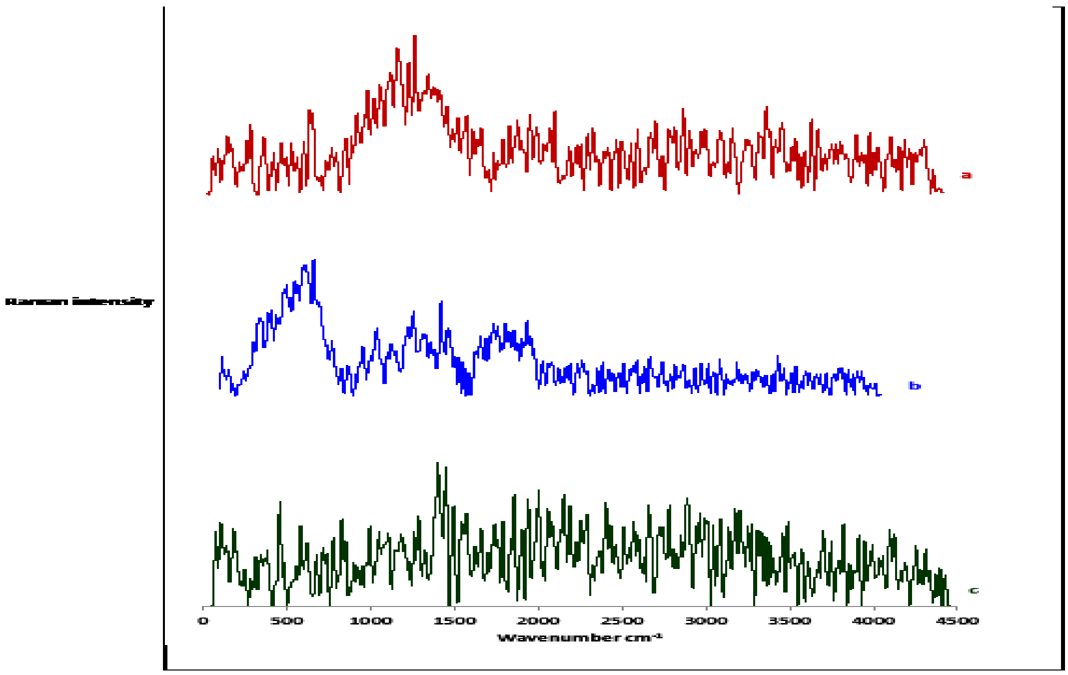

Fig. 7. Raman spectra of (a) toner, (b) toner + red STAEDTLER stick pen and (c) toner + blue FABER CASTELL..

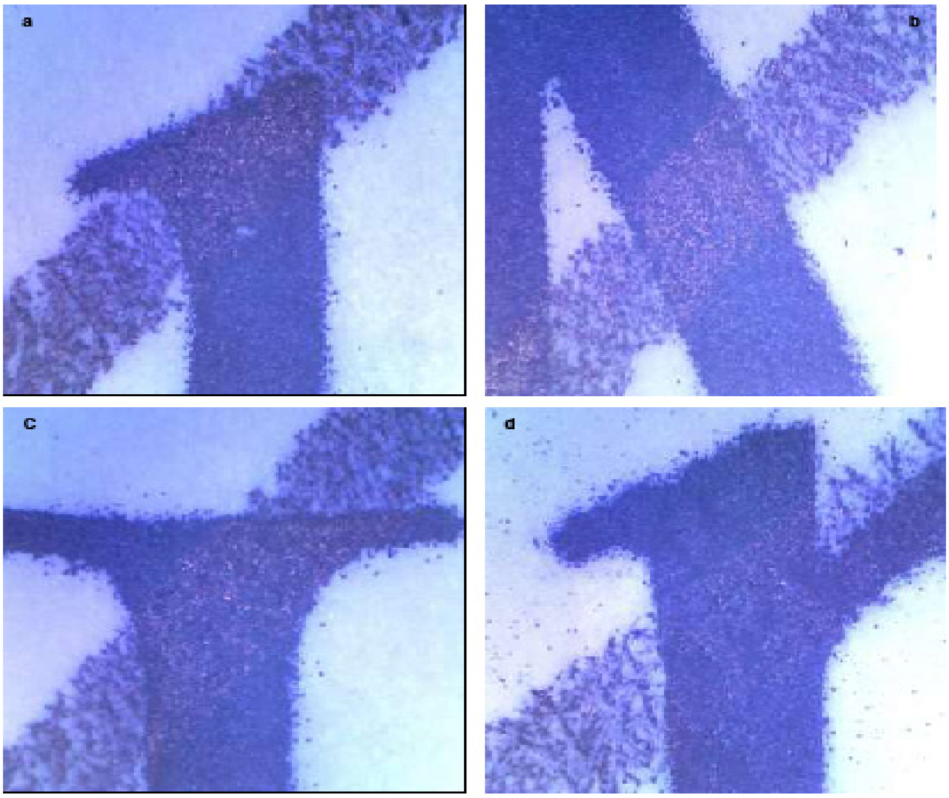

Fig. 8. Gloss of black Uni-laknock MITSUBISHI ink crossed over a toner of (a) Xerox phaser 6700, (b) HP 2300, (c) HP 1018 and (d) Epson M 2000 under the digital microscope.

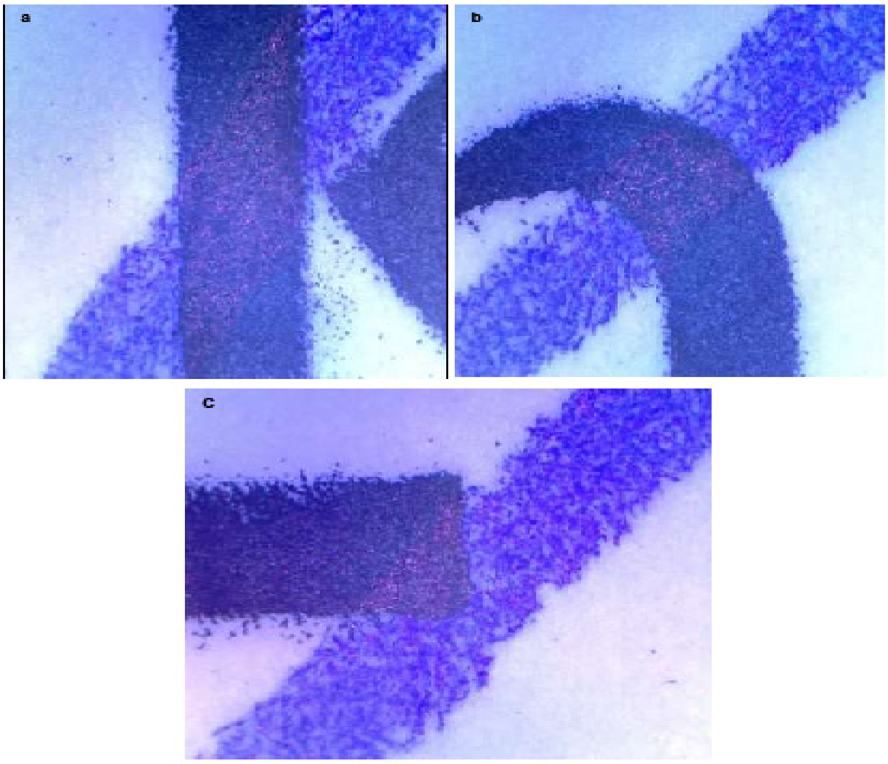

Fig. 9. The spreading of blue Uni-laknock MITSUBISHI ink crossed over toner on (a) coated, (b) non-coated and (c) smooth paper under the digital microscope.

Additionally, the five-time gaps between two strokes show that no effect recorded on the observed results as in Fig. 10. The five-time gaps are enough to the first stroke to fix and distribute normally on the paper surface without any effect from the second stroke.

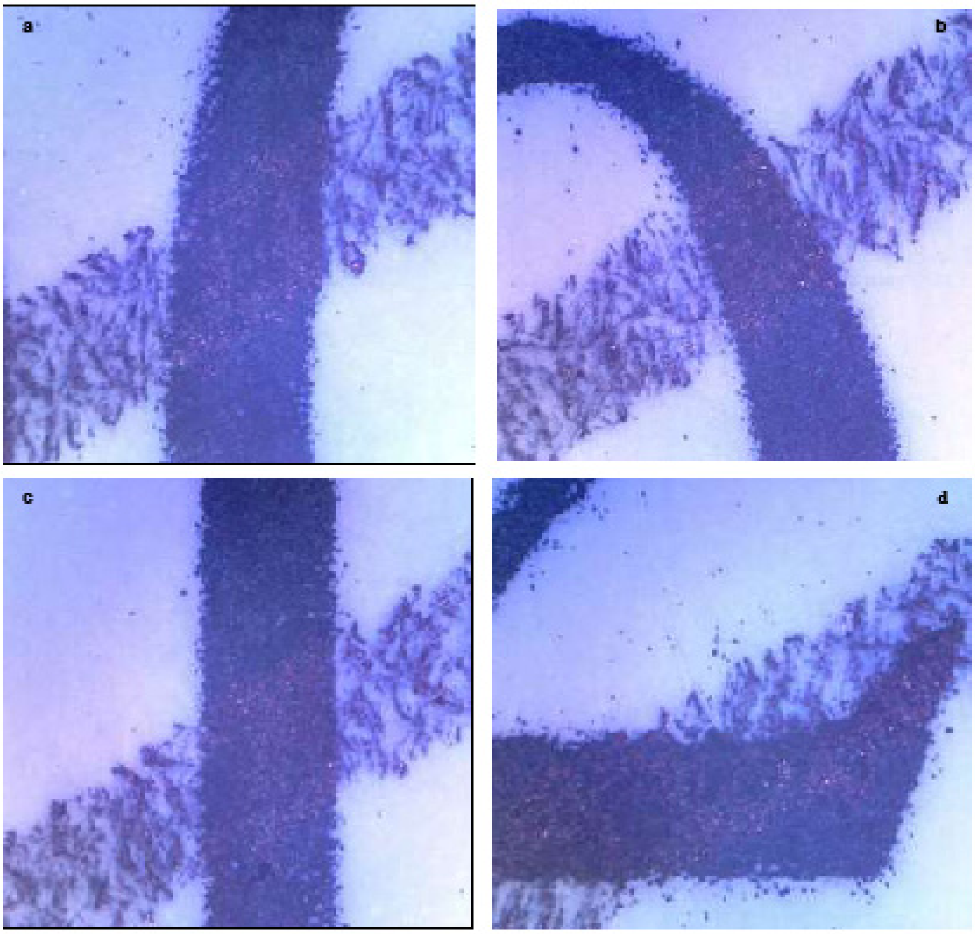

Fig. 10. The specular reflection when black PRIMA FORMA ink is crossed over a toner after (a) 5 mins, (b) 2 hours, (c) 10 days and (d) 30 days under digital microscope.

Blind testing

The volunteer person was prepared the unknowen samples observed from the crossing of the toner and ballpoint pen ink. The sequence of the toner and ink strokes was blind for some authors before the analysis and known to volunteer person only. Blind samples were examined by authors to detect the error rate perhaps present in using the application of a digital microscope and Raman spectroscopy. The results recorded by the authors were compared with the key of unknowen samples by volunteer person. Firstly, the data of Raman spectroscopy successful in detects the order of strokes intersection. Secondly, in the data of the digital microscope, all authors give right answers for the order of strokes intersection in all unknowen samples except in the case of five samples of red color crossed over the toner. All results observed by the authors are listed in Table 5. This blind testing' shows that the restrictions of using the digital microscope alone in detecting the order of ink crossed over the toner, especially for some brands of ballpoint pen red ink because of optical illusions and varied perceptions of a human. The combination of Raman curves which depend on the chemical differences between the composition of toner and ink with other method as the digital microscope has more efficiency and reach at one hundred percent in the detection of the order of strokes intersection. Therefore, these results are useful in the felid of forensic document examination.

TABLE 5. Listed the different results produced from the blind testing.

Authors										Order of strokes	No of blind Samples
Raman	**Microscope**	**Raman**	**Microscope**	**Raman**	**Microscope**	**Raman**	**Microscope**	**Raman**	**Microscope**		
Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Toner over red ink	10
Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Black ink over toner	10
Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Toner over black ink	10
Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Toner over blue ink	10
Correct	5 Inconclusive 5 Correct	Correct	5 Inconclusive 5 Correct	Correct	5 Inconclusive 5 Correct	Correct	5 Inconclusive 5 Correct	Correct	5 Inconclusive 5 Correct	Red ink over toner	10
Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Blue ink over toner	10

Conclusion

The specular reflection, gloss, gap, and spreading of inks, are four physical characterization that were studied by the digital microscope. The obtained results give more efficiency for the order of intersecting strokes, except in case of the red color of some brands of ballpoint pen inks. Also, the results observed from Raman spectroscopy at the intersection points were related to the stroke written or printed later regardless it's used color. Therefore, the utilizing of the digital microscope as a complementary technique was easily used to determine the chronological order of intersecting strokes. The time gap between ballpoint pen ink strokes and the toner strokes, the nature of the paper surface, brand and model of printer have no effect on the observed results.

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استخدام الخواص الفيزيائية والكيميائية لأحبار الطابعات الليزرية والاقلام الجافة في تعيين تتابع تقاطعهما

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يعتبر الفحص الشرعي للمستندات والوثائق من اقدم الفروع في مجال العلوم الطبية الشرعية. فالعلاقة المستندية الصحيحة تقوم علي قراءة اطراف هذا المستند لمتنه ثم التوقيع عليه بما يعني اقرارهم بما يحتويه هذا المستند. وقد يتعرض المستند لعملية تزوير بالإضافة او الاستبدال لبعض او كل ما ورد بمتنه استغلالا لوجود توقيع احد الاطراف عليه وقد يتقاطع ما تم اضافته او استبداله (البيانات الجديدة المزورة) لاحقا مع جرات التوقيع الموجودة سابقا وتكون الوسيلة الوحيدة لأستبيان التزوير هو تحديد التتابع بين الجرات اللاحقة والسابقة. في هذه الدراسة تم تحضير العينات التي تحتوي علي نوعي التقاطع للتونر مع الاحبار الجافة (التونر فوق الحبر والحبر فوق التونر) ودراسة بعض العوامل المؤثرة علي تحديد هذا التقاطع مثل ماركة وموديل الطابعات الليزرية ، ماركة الاحبار الجافة , نوع سطح الورق المحضر عليه العينات، المسافة الزمنية بين الجرة الاولي والثانية. وقد استخدم في فحص الجرات طريقتين غير متلفتين للمستند اثناء فحصه وهما الميكروسكوب الرقمي ومطياف الرمن. وقد نجح الميكروسكوب الرقمي في تحديد تتابع الجرات باستثناء بعض الالوان لبعض ماركات الاحبار الجافة. بناء عليه تم اللجوء للطريقة الأخرى التي يمكن من خلالها تحديد تتابع الجرات المتقاطعة التي فشل الميكروسكوب الرقمي في تحديدها مثل طريقة مطياف الرمن. وقد اعطي مطياف الرمن تحديد لتتابع كل جرات الاحبار الجافة مع التونر بغض النظر عن الوانها. وعليه فان دمج الميكروسكوب الرقمي مع مطياف الرمن في تحديد تتابع تقاطع جرات التونر مع الاحبار الجافة هي طريقة غير متلفة ، جديدة، واعدة في مجال فحص المستندات للتأكد من تعرضها للتزوير من عدمه.