



Physical Distinguishable of Heterogeneous Overlapping Resulting from Stamp-pad and Laser Printing Inks

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DETEECTING the arrangement of intersecting stamp-pad ink and the toner of laser printing strokes is an excellent method of examining the documents originality. In this study, different six brands of stamp-pad inks, toner of five laser printers of various brands and models, three types of paper surfaces, two kinds of seal materials and three methods of seal manufactures were utilized to produce the samples of heterogeneous intersecting strokes. A combination of the microscopic method as a digital microscope and analytical instrumental technique as Raman spectroscopy were performed to determine the arrangement of intersecting strokes. The digital microscope succeeds in the detection of the arrangement of intersecting strokes of some stamp-pad inks and fails in detection the others depending on their brands and colors through notation four physical properties at intersection point the specular reflection, the ink and toner glosses, gaps and spreading. The Raman spectroscopy determined the ink or toner stamped or printed later by comparison the spectra of pure ink, toner, and their intersections and succeeds in detection all samples through the spectral data such as peak location and peak intensity. The combination of a digital microscope and Raman spectroscopy which applied successfully discriminates the arrangement of intersecting strokes of toner and all brands of stamp-pad inks regardless of their brands and color.

Keywords: Forensic discrimination, Strokes intersection, Stamp-pad inks, Toner laser printer, Seal inks.

Introduction

The problem of determining the sequence of intersecting strokes always considered a great aim for forensic document examiners[1-5]. An investigation of the sequence of intersecting strokes is a sufficient method for detecting the authentication and validation of the documents. In order to adopt the legal form, the signature of the parties involved or their representatives must be signed on the document[6-9].

With respect to some documents have a signature such as checks, ratified contracts,

receipts, identity documents, office paperwork, invoices and agreements, a signature perhaps is not enough and the document is often required to stamped after the person reads and agrees on the content of the document[10]. The documents created by printing technology and have stamps are growing and increased significantly nowadays. Therefore, in the authentication of the documents, the seal impression will appear over the printed text of the document[11].

For criminal purposes, the forgery or alteration documents often took place by adding

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a new printing text on the empty section of the paper or on empty whole papers that have original signatures and/or seal. The inclusion of an added paragraph could dramatically convert the clauses of the agreement[12]. This new sort of documents forgery is becoming more common. If the extra text intersects with the seal or signature on the document, then the modification perhaps determined by investigating the sequence of intersecting strokes[13].

Informer researches, the intersecting of stamp-pad ink and toner laser printer strokes was determined and many techniques are available to examine them. A comprehensive survey of the available techniques shows that the techniques have various destructive and nondestructive. For forensic document examination, a nondestructive method is often desirable over a one. A lot of techniques applied to investigate strokes intersection are depended on visual examination and noticing of physical characteristics of the ink[14]. Although, optical methods are still, until recently, the commonly used and first choice in determining the sequence of intersecting strokes. But, its results alone are unsatisfying as forensic evidence in court because it depends highly on the skill levels and experiences of the examiner. Especially, in the case of stamp-pad ink over toner because it passes through the different layers present because of its high penetrability[15]. Furthermore, the using of optical methods as digital microscope may be cannot determine the sequence of intersecting strokes successfully. Accordingly, in addition to the application of optical methods, other nondestructive techniques such as Raman spectroscopy is used which beneficial for the identification of organic compounds, involving glossy components[16]. In this study, utilizing a digital microscope in combination with Raman spectroscopy to detects the sequence of stroke intersections that are produced by stamp-pad ink and toner laser printer.

Materials and Methods

Specimen preparation

Two types of overlapping were used to prepare samples of intersecting strokes. In the first intersection type, toner printed first then stamp-pad ink crossed over it. In the second intersection type, stamp-pad ink stamped first then the toner printed over it. All ink samples stamped using seals under the same conditions and by the same *Egypt.J.Chem.* **62**, No. 8 (2019)

person to overcome the variations in the normal pressure[10].

The intersecting of stamp-pad ink and toner is a dynamic process affected by many physical and chemical variables which determine how the ink and toner will distribute and interact with each other on the paper.

Accordingly, more than one factor was studied to determine their effect on the observed results. The first factor was the alteration in the brand and model of the printers as listed in Table 1 because the chemical composition of the toner may differ from brand to other[12].

The second factor was brands of stamp-pad ink and their colors. Recently, the speed developments in the composition of stamp-pad ink from brand to others also perhaps effect on the obtained results[17]. Therefore, seven brands of stamp-pad ink listed in Table 2 with different colors available in Egyptian markets and commonly used in delay cases work were used in the samples preparation[18,19].

The third factor is paper surface properties which may be affected by how the stamp-pad ink and toner distribute over the paper. To study the effect of nature paper surface, the intersection samples were prepared on three different types of paper as listed in Table 3[13].

The fourth factor was the time gap between application of stamp-pad ink and toner which directly related to the ink and toner drying process. The second stroke stamped or printed on the first stroke after the time gap is 5 mins, 2 hrs, 10 days, 20 days and 30 days[2,7].

The last factor has studied the type of materials and manufacture methods of the seal. More than one materials and manufacture method can be used in the production of seals. For example, copper, zinc and rubber used as materials, and hand, chemical and laser engraving used as manufacture method. The hand carving involves producing a metal seal by hand engraving using a hard metal hand machine has a sharp head. Hand engraving is time-consuming and requires high-level hand skills. In both chemical and laser engraving a seal design is carried out by image computer software's[20]. Then in chemical engraving the design printed on a film, placed on a photosensitive compound, exposed to the light, and etched using a solvent to create the desired seal pattern. While in laser engraving

a seal design produced by computer-aided laser engraving machine. Therefore, the interaction between the seal materials and paper surface perhaps effect on the amount of ink and clearness of seal impression on paper. Accordingly, the intersection samples were stamped using seals of different materials and engraving methods as listed in Table 4[21].

All prepared samples were left for the same period of time gap at room temperature and keep under similar climatic conditions before the examination and analysis process.

The digital microscope

The intersection specimens of toner and stamp-pad ink were examined and captured with MDA1300 (China) digital microscope that has a magnification 20-200X. The lighting system of the digital microscope consists of eight lighting lamps of the annular light source located on the head of a microscope and directed in the same direction. The

digital microscope was placed perpendicularly to a sheet paper and zoom-on intersection point. Zooming and lighting systems in a digital microscope show distribution of dyes/pigments in stamp-pad ink and gloss in toner at point intersections. The images were observed and captured directly on the computer monitor [2].

Raman spectroscopy

Raman spectrum was measured using the Senterra spectrometer attached with Bruker microscopes. A 50X objective lens was used to concentrate the laser beam spot on the intersection point before 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[22]. 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

TABLE 1. List of the printer brands and models.

The printers brand	The models of the brands	The toner 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 the stamp-pad ink brands.

The brands of stamp-pad ink	The ink color
Faber Castell	Black, red and blue
flower	Black, red and blue
Horse	Black, red and blue
Huhua	Black and blue
Mac	Black and red
RADEX metallica	Red and blue

TABLE 3. List of different nature paper surface.

The nature of the paper surface	The paper brands	The paper size	The paper weight (g/m^2)
non-coated	MultiOffice	A4	80
coated	Antalis	A4	80
smooth	Antalis	A4	80

TABLE 4. List of materials and manufacture methods of seals.

The types of seal martial	The types of seal manufacture
Rubber	Chemical engraving
Metal	
Rubber	Laser engraving
Metal	Hand engraving

of peaks. The selected points for measuring the spectrum were manually and randomly choice[23]. 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 [24].

Results and Discussion

Under the digital microscope

Determination the sequence of intersecting strokes consist of stamp-pad inks and toner is taken place depending on the difference in the composition between stamp-pad ink and toner, furthermore, modification the later ink due to the existence of the first ink [10]. The investigation by the digital microscope concentrate on some visual properties exist in both stamp-pad ink and toner and the effect of their intersection on these properties, therefore the following physical characteristics were studied under the digital microscope[13]:

Specular reflection

The first physical property studied under the digital microscope is the specular reflection which considered mirror-like reflections from the surface at the intersection point. The specular reflection observed only when brands of stamp-pad ink as listed in Table 5 stamped over toner strokes[19].

The existence of specular reflection because the colorant particles (pigments and /or dyes) present in stamp-pad inks accumulate over toner and fills some parts at intersection point as in Fig. (1a-1d). Accordingly, the sequence of intersecting strokes become easier to detect [25].

The absence of specular reflection has taken place when brands of stamp-pad ink listed in Table 6 stamped after toner strokes as in Fig. (2a-2b).

This is due to the composition of ink brands listed in Table 6 is a mixture of colorants and non-colorants components and the non-colorant components perhaps have a masking effect which inhibits the colorants components to shows its specular reflection or all colorant components soluble in the ink solvent, therefore evaporated or/and absorbed in cellulose fiber of paper. Furthermore, the specular reflection of ink similar to the toner glosses accordingly, the sequence of intersecting strokes become difficult to detect[10].

All brands of stamp-pad ink (all colors) have the same manner for the absence of specular reflection when toner printed after them as in Fig. (3a–3d). The laser printer forms an opaque and thick layer of toner powder covers the specular reflection of the colorant particles present in stamp-pad inks and hide it[13].

Gloss of ink

One of the greatest physical properties investigated under the digital microscope is the ink and toner gloss. The toner has glossy properties due to the presence of wax in its composition [26,27] and stamp-pad inks also have the same properties due to the presence of colorants particles in its composition [25]. Therefore, the gloss continuity or discontinuity of ink and toner can be used as an indicator in determining the ink or toner stamped or printed later.

When brands of stamp-pad ink listed in Table 5 crossed toner, the gloss of both ink and toner is continued. The continuity of ink gloss appears only at some parts and the continuity of toner gloss shows on the other parts at intersection point as in Fig. (4a-4d). A part of colorant particles present in stamp ink accumulates on the toner surface and the other penetrate it to absorb by cellulose fiber of paper[22].

TABLE 5. List of stamp-pad ink brands has a specular reflection.

The brands of stamp-pad ink	The ink color
Faber Castell	Black and blue
flower	Black, red and blue
Horse	Red and blue
Huhua	Black and blue
Mac	Black
RADEX metallica	Blue

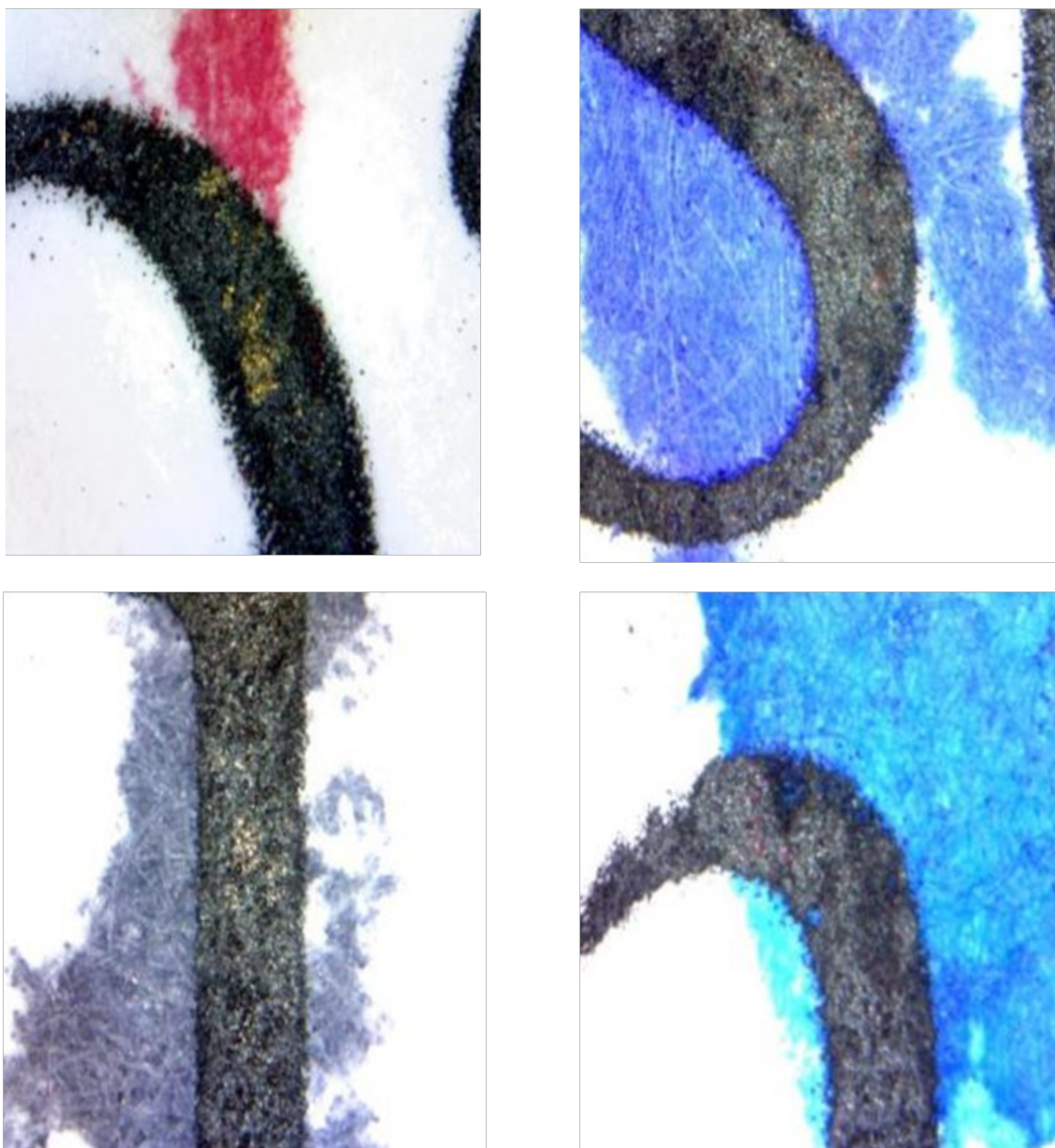


Fig. 1. The specular reflection when (a) red flower, (b) blue Horse, (c) black Huhua and (d) blue RADEX metallica stamp-pad inks are over a toner under a digital microscope.

TABLE 6. List of stamp-pad ink brands hasn't specular reflection.

The brands of stamp-pad ink	The ink color
Faber Castell	Red
Horse	Black
Mac	Red
RADEX metallica	Red

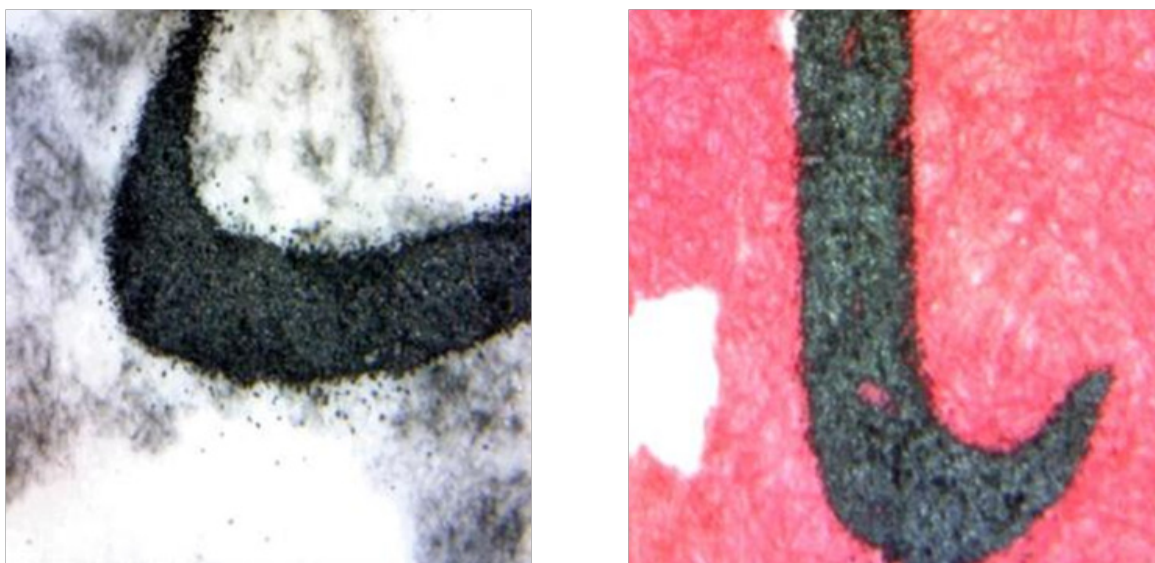


Fig. 2. The absence of specular reflection when (a) black Horse and (b) red RADEX metallica stamp-pad inks are over a toner under a digital microscope.

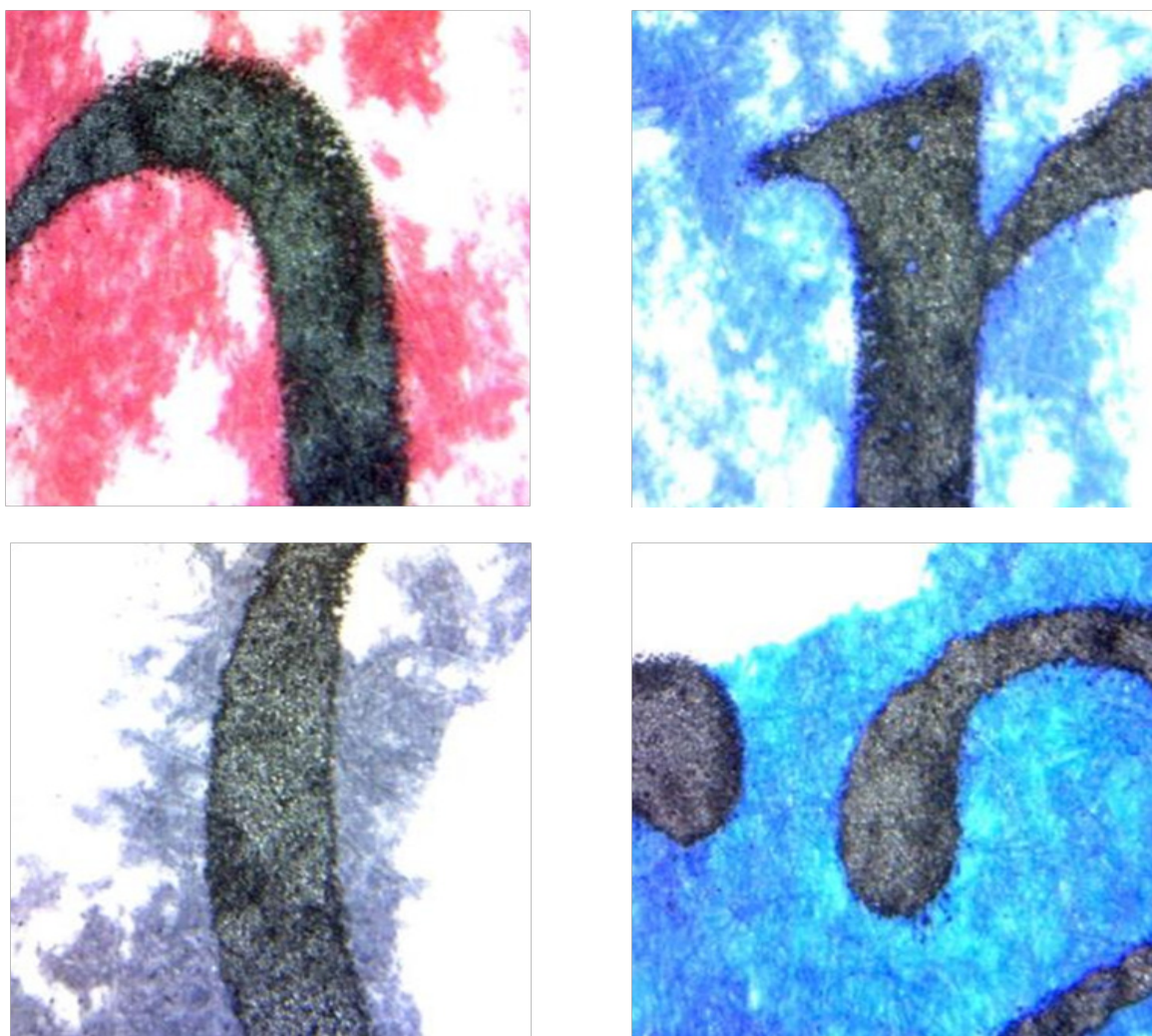


Fig. 3. The absence of specular reflection when toner is printed over (a) red flower, (b) blue Horse, (c) black Huhua and (d) blue RADEX metallica inks under a digital microscope.

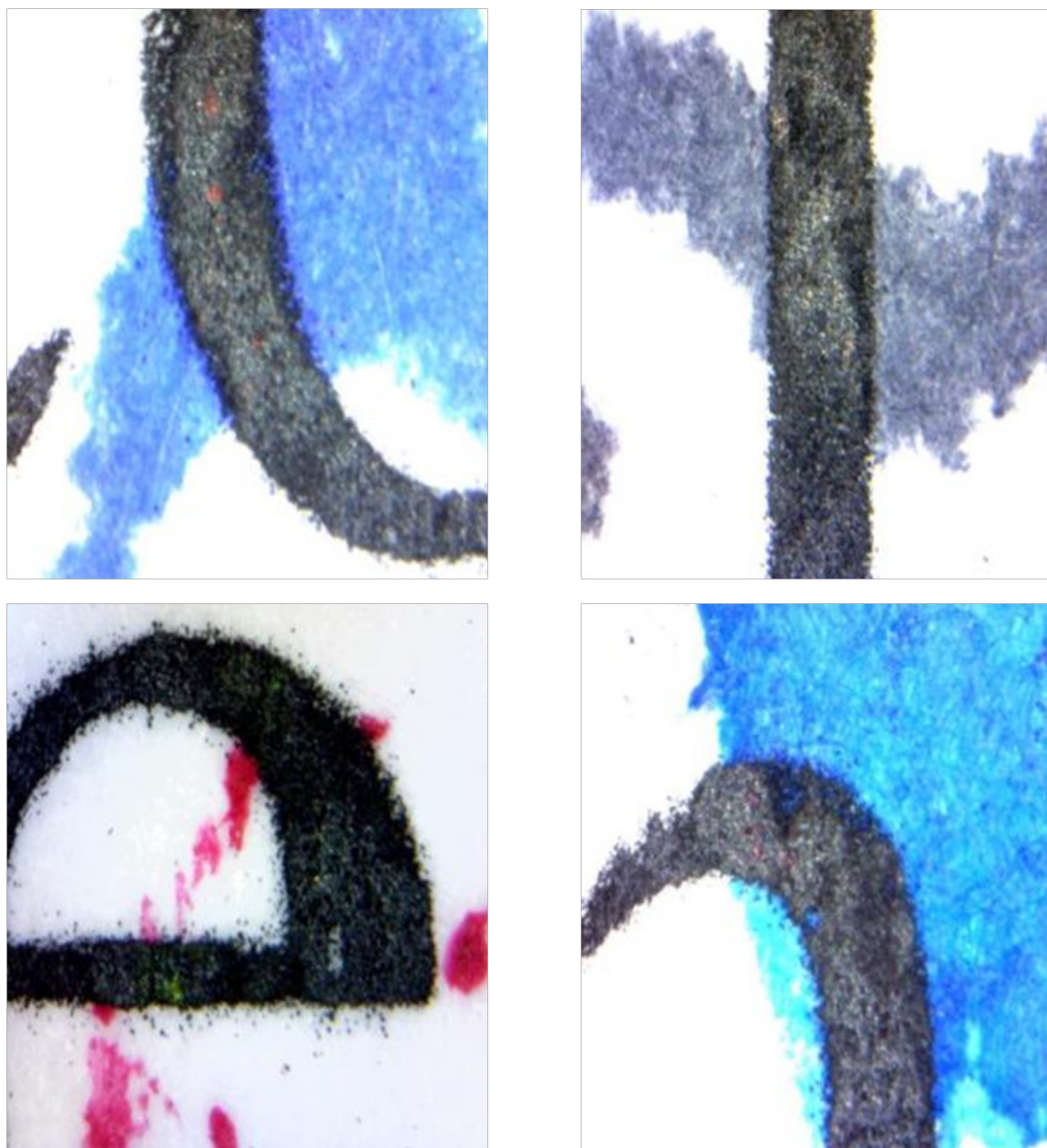


Fig. 4. The continuity gloss of toner and ink when (a) blue Faber Castell, (b) black Huhua, (c) red Horse and (d) blue RADEX metallica are over the toner under a digital microscope.

On the other hand, when toner printed after brands of stamp-pad ink listed in Table 5 and after or before brands of stamp-pad ink listed in Table 6 the toner gloss is continued and ink gloss is discounted [10]. With respect to ink listed in Table 5 and listed in Table 6 especially in case toner printed after ink, the toner gloss is continued and ink gloss is discounted because the toner fixed on paper surface as a glossy thick layer which covers the ink gloss at the intersection point as in Fig. (5a-5d) [13].

In the case of toner printed before brands of ink listed in Table 6, the toner gloss continuity and ink gloss discontinuity because the colorant particles exist in ink stamped later may be full penetrate holes of top toner layer and absorbed by cellulose fiber of paper or the toner printed first has block effect to the colorant particles in ink as in Fig. (6a-6b).

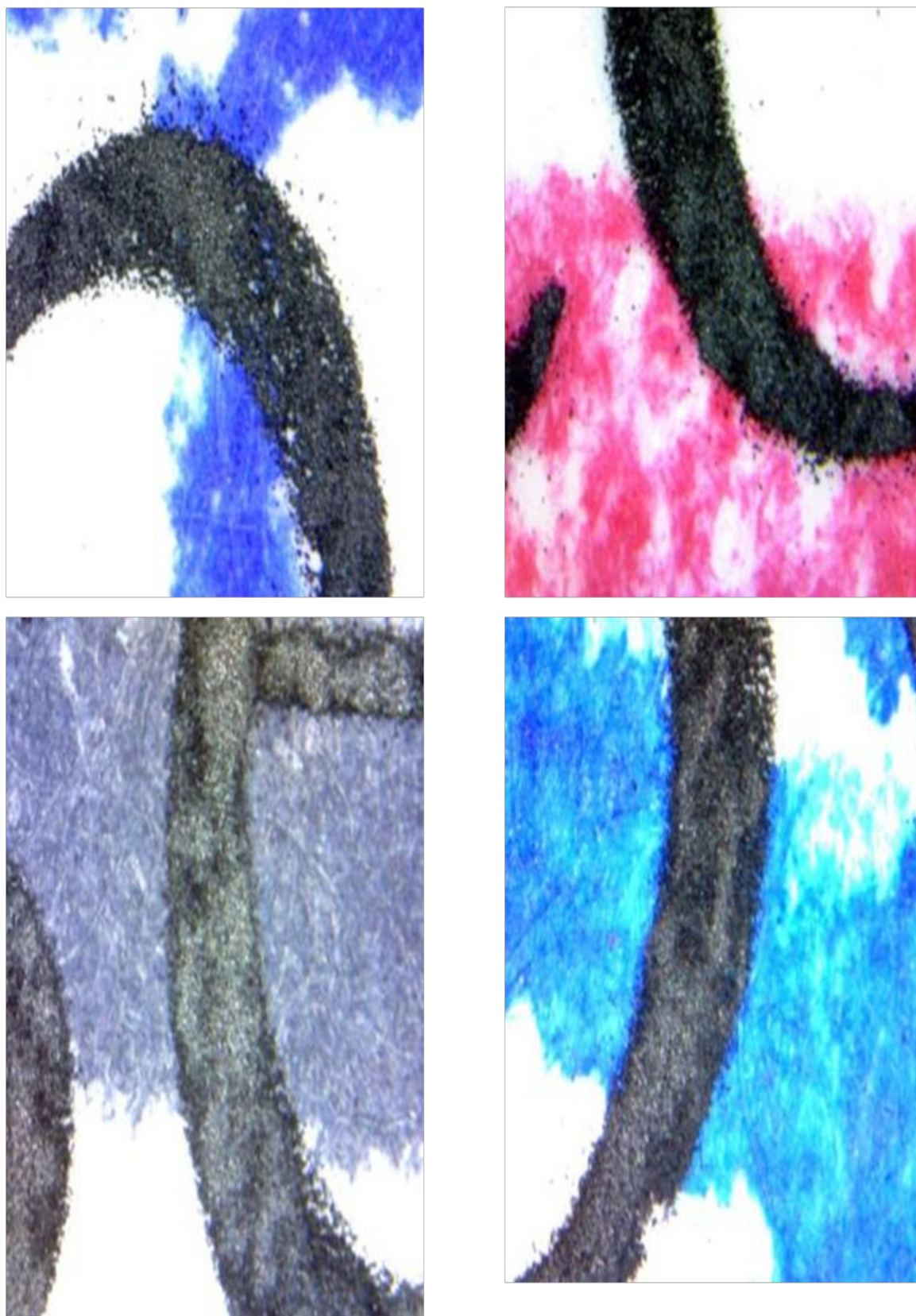


Fig. 5. The continuity gloss of toner and discontinuity ink gloss when the toner is printed over (a) blue flower, (b) red Horse, (c) black Mac and (d) blue RADEX metallica inks under a digital microscope.



Fig. 6. The continuity gloss of toner and discontinuity ink gloss when (a) red Faber Castell and (b) black Horse inks are over a toner under a digital microscope.

Another important property investigated under a digital microscope is the ink and toner gaps which arise in two intersection types. The ink gaps formed when the stamp-pad ink intersects toner. The toner fixed on the paper surface forming a higher layer than the paper layer. Therefore, during the stamping process, the ink stamped on two levels (paper and toner) accordingly, the ink gaps appear before and/or after the intersection point as in Fig. (7a–7b) [19].

The toner gaps observed when toner intersects stamp-pad ink. This is due to the stamp-pad ink stamped first, sometimes led to a disturbance in the electrostatic process due to interacting with the photosensitive roller of the laser printer. There is another effect causing a change in the surface of paper due to partially melt with the application of a high temperature in a laser printer. Accordingly, some toner holes appear during the fixing of toner at the intersection point and color of stamp-pad ink shows through the toner holes as in Fig. (8a–8d) [28].

Spreading of ink

The last physical property studied under a digital microscope is the ink and toner spreading. The stamp-pad ink spread over toner in two ways.

The first way when the brands of ink listed in Table 5 stamped over toner, the spreading of colorant particles exists in the ink composition of these brands differs than the toner gloss. However, the ink spreading distinguishable at the intersection point and the sequence of the intersecting strokes become easier to detect as in Fig. (9a–9d).

The second way when the ink brands listed in Table 6 crossed over toner, the spreading of colorant particles present in the ink composition of these brands similar to the toner gloss. Therefore, the ink spreading indistinguishable at the intersection point and the sequence of the intersecting strokes become more difficult to detect as in Fig. (10a–10b).

On the other hand, an incomplete toner spreading overall ink brands due to the existence of toner holes and the ink color of the lower layer appears from these holes at intersection point as in Fig. (11a–11b). The ink stamped first, almost led to a disturbance in the electrostatic process due to interacting with the photosensitive roller of the laser printer or the change in the surface of paper due to partially melt with the application of a high temperature in a laser printer. Therefore, incomplete toner spreading over ink takes place.

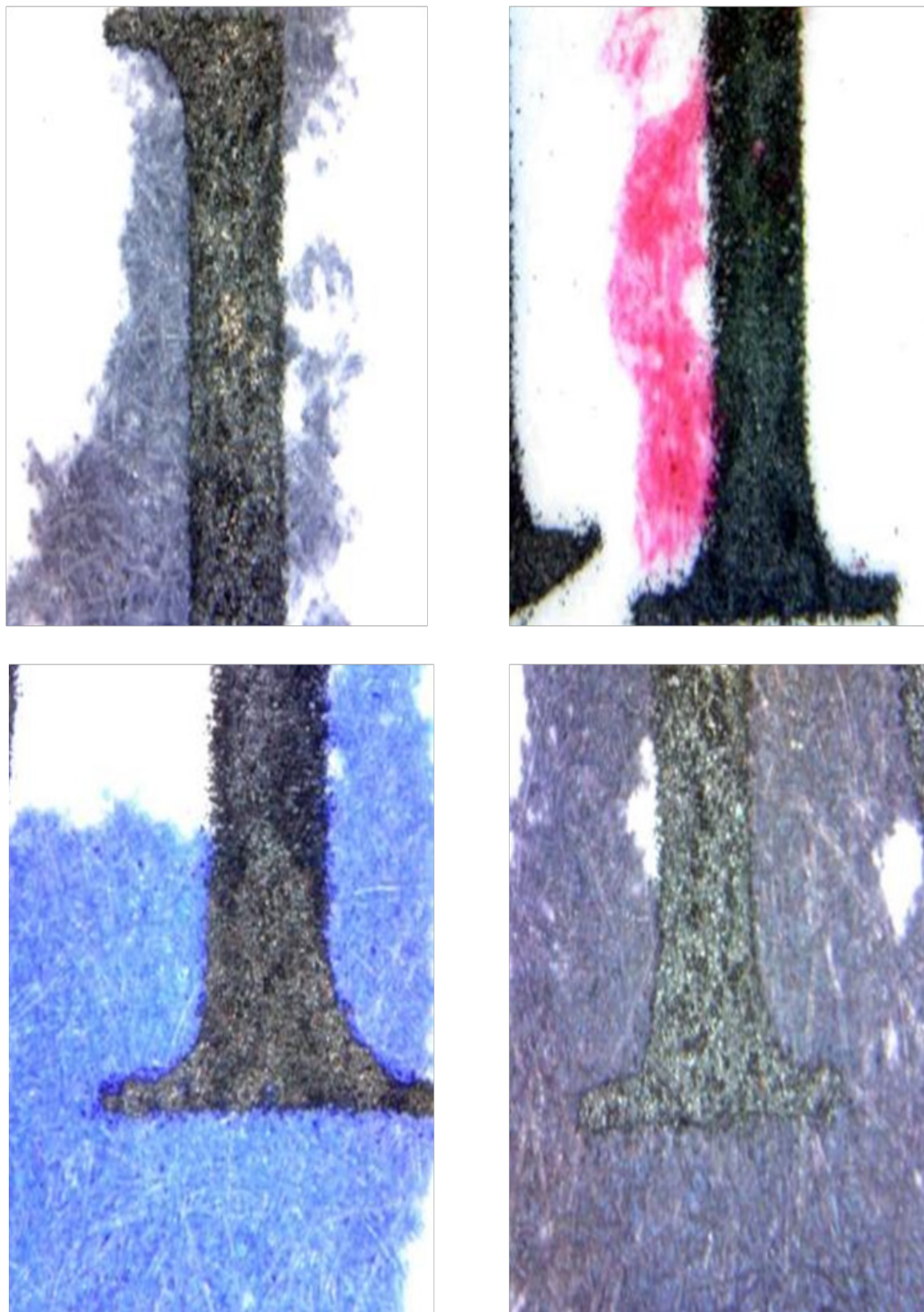


Fig. 7. The ink gaps when (a) black Huhua, (b) red flower, (c) blue Horse and (d) black Faber Castell inks are over a toner under a digital microscope.

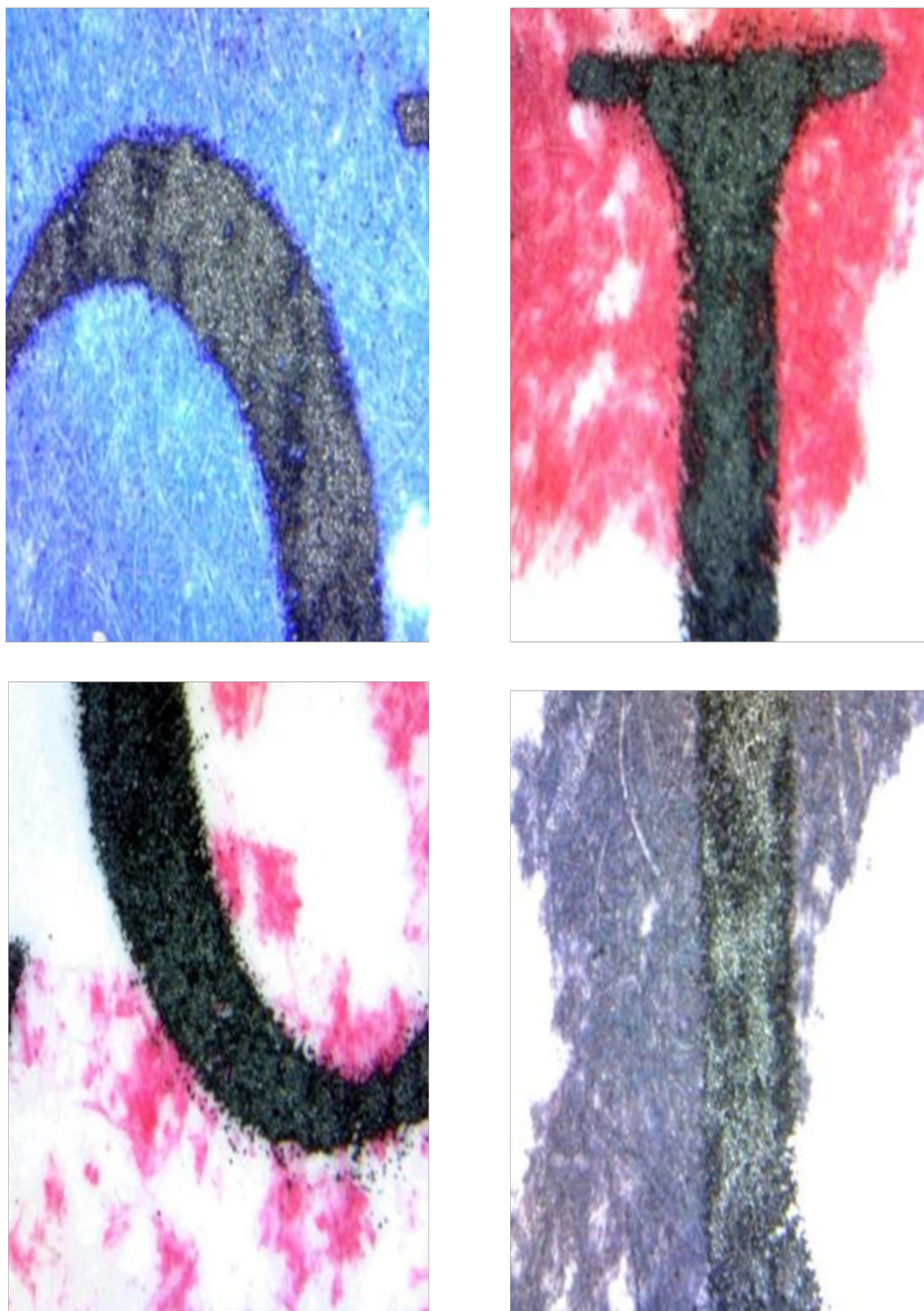


Fig. 8. The toner gaps when toner is printed over (a) blue Huhua, (b) red flower, (c) red Horse and (d) black Faber Castell inks under a digital microscope.

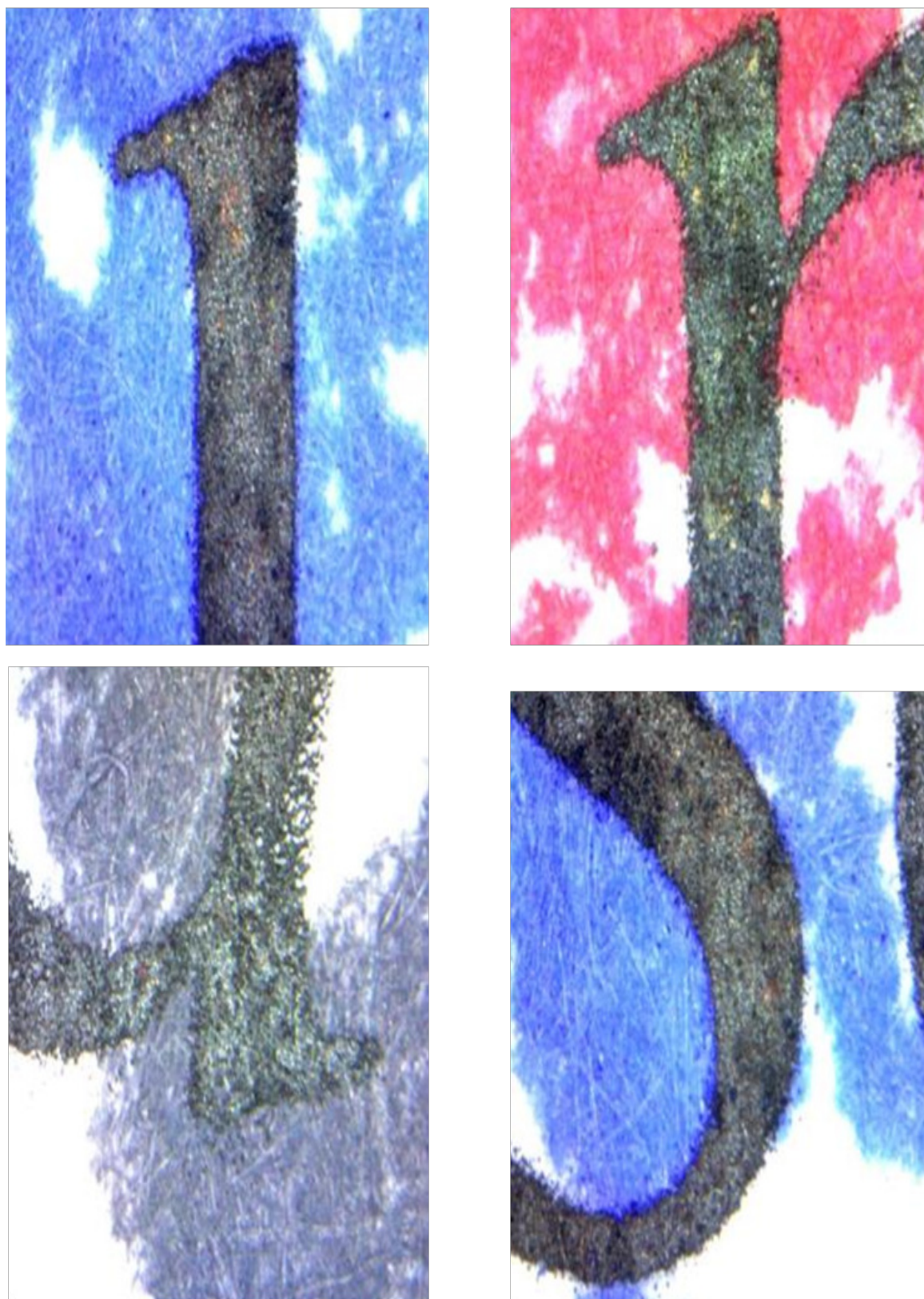


Fig. 9. The spreading of (a) blue Huhua, (b) red flower, (c) black Faber Castell and (d) blue Horse inks over a toner under a digital microscope.

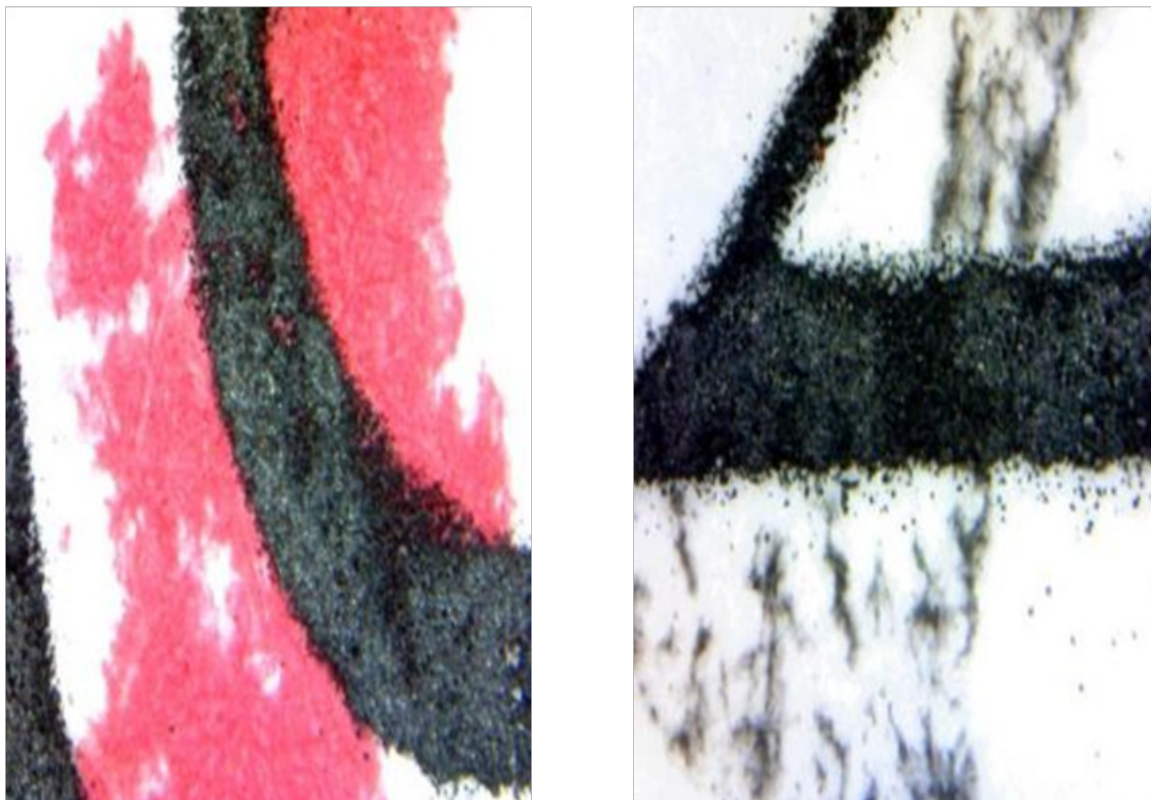


Fig. 10. The spreading of (a) red Faber Castell and (b) black Horse inks over toner under a digital microscope.

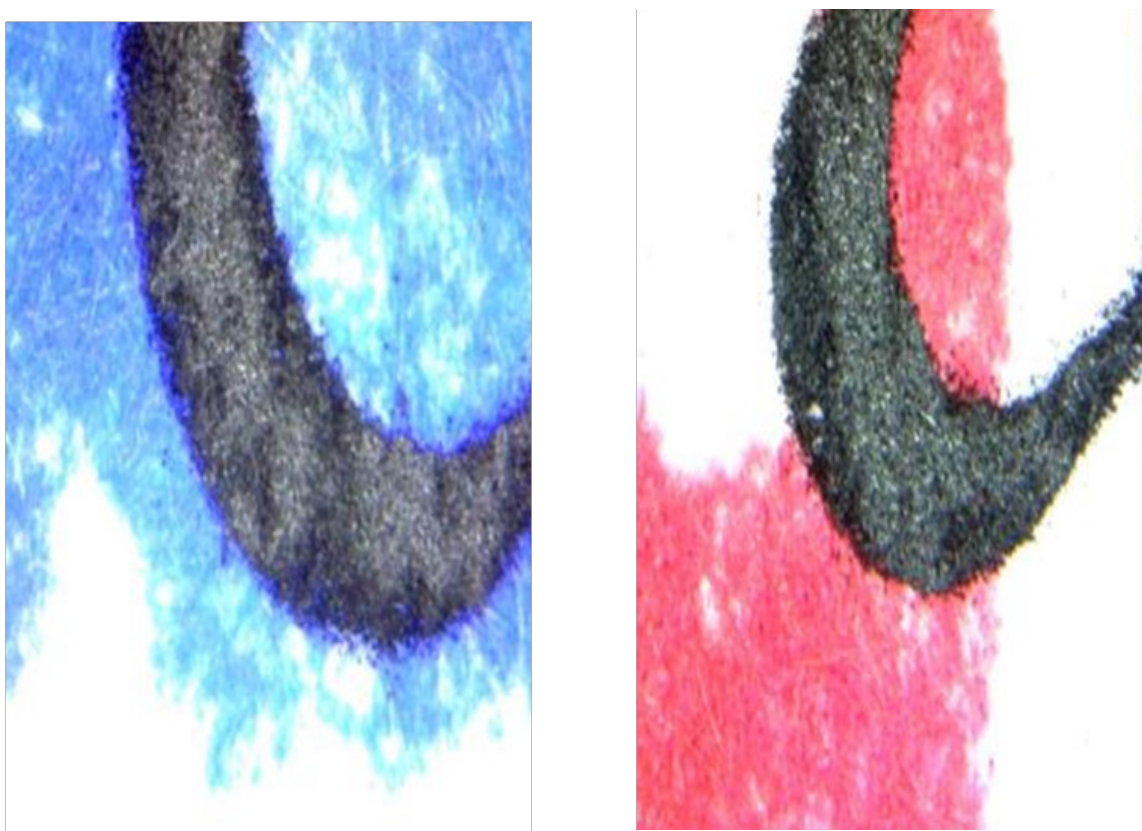


Fig. 11. The incomplete toner spreading over (a) blue flower and (b) red Mac ink under a digital microscope.

The net results of four physical properties studied under a digital microscope almost determine the sequence of intersecting strokes of stamp-pad ink and toner only for brands listed in Table 5. In brands of the ink listed in Table 6 almost no differences between the manners of two intersection types, therefore the sequence of intersection strokes fails to determine as in Fig. (2a-2b, 5a-5d)[23].

Accordingly, it is a great benefit to apply a digital microscopic as the first step in detection the sequence of intersecting strokes by the combination with other non-destructive chemical analysis method such as Raman spectroscopy which gives results can be complete the results of digital microscopic [24].

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 pure stamp-pad 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 inks indicate that this ink executed later.

The toner and two kinds of stamp-pad ink (Mac red ink, Huhua blue ink) are subjected to Raman examination in two types of intersection. Firstly, when the stamp-pad ink impressed over toner, the spectrum at the intersections point corresponded to the spectrum of pure ink. The Raman spectra of pure ink and ink impressed over toner almost have a high degree of similarity in the shape and value of the main peaks as in Fig. 12.

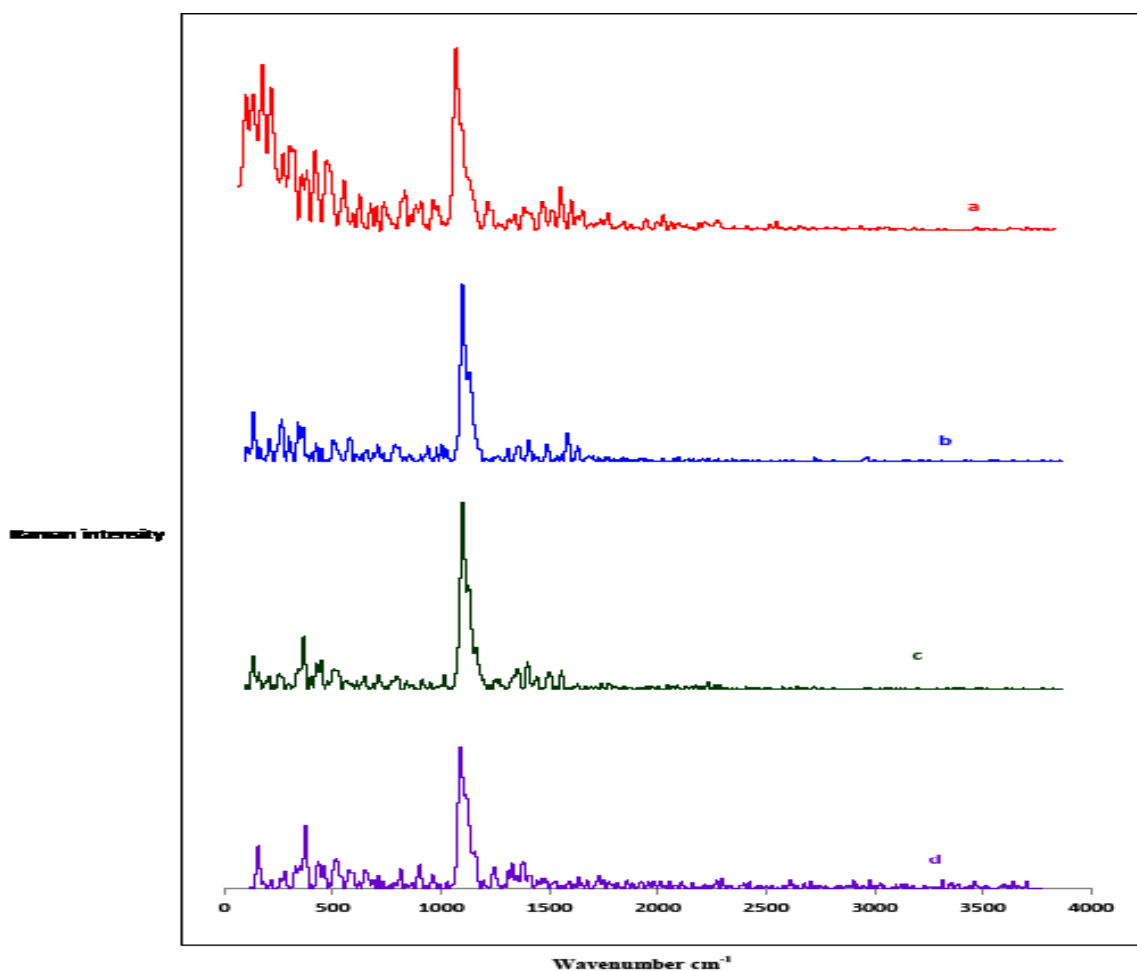


Fig. 12. Raman spectra of (a) red Mac, (b) red Mac (chemical engraving) + toner, (c) blue Faber Castell and (d) blue Faber Castell + toner.

In case of ink crossed over the toner, when the solvents and other organic additives present in ink composition are evaporated, the colorant materials (dyes and/ or pigments) are totally maintained at the intersection point. Additionally, when the toner printed over the ink the resin present in toner accumulate at the intersection point. Therefore, the Raman spectrum generated from the intersection point indicated to either the existence of dyes in ink or resin in the toner.

This is may be due to the colorant and non-colorant matters exist in the ink composition completely accumulated on the surface of toner

at the intersection point. Therefore, the Raman spectrum at intersection point seems identical with the spectrum of pure stamp-pad ink.

Secondly, when toner printed overstamp-pad ink, it is clear that the spectra of toner and toner over ink similar in the shape which consists of very narrow peaks. Also, three spectra have printer ink of characteristic peaks at 660-680 and 2900-2920 cm^{-1} which related to polystyrene/acrylate the main component in the toner of a laser printer. But, the main peaks of the two spectra differ in its wave numbers and height as in Fig. 13.

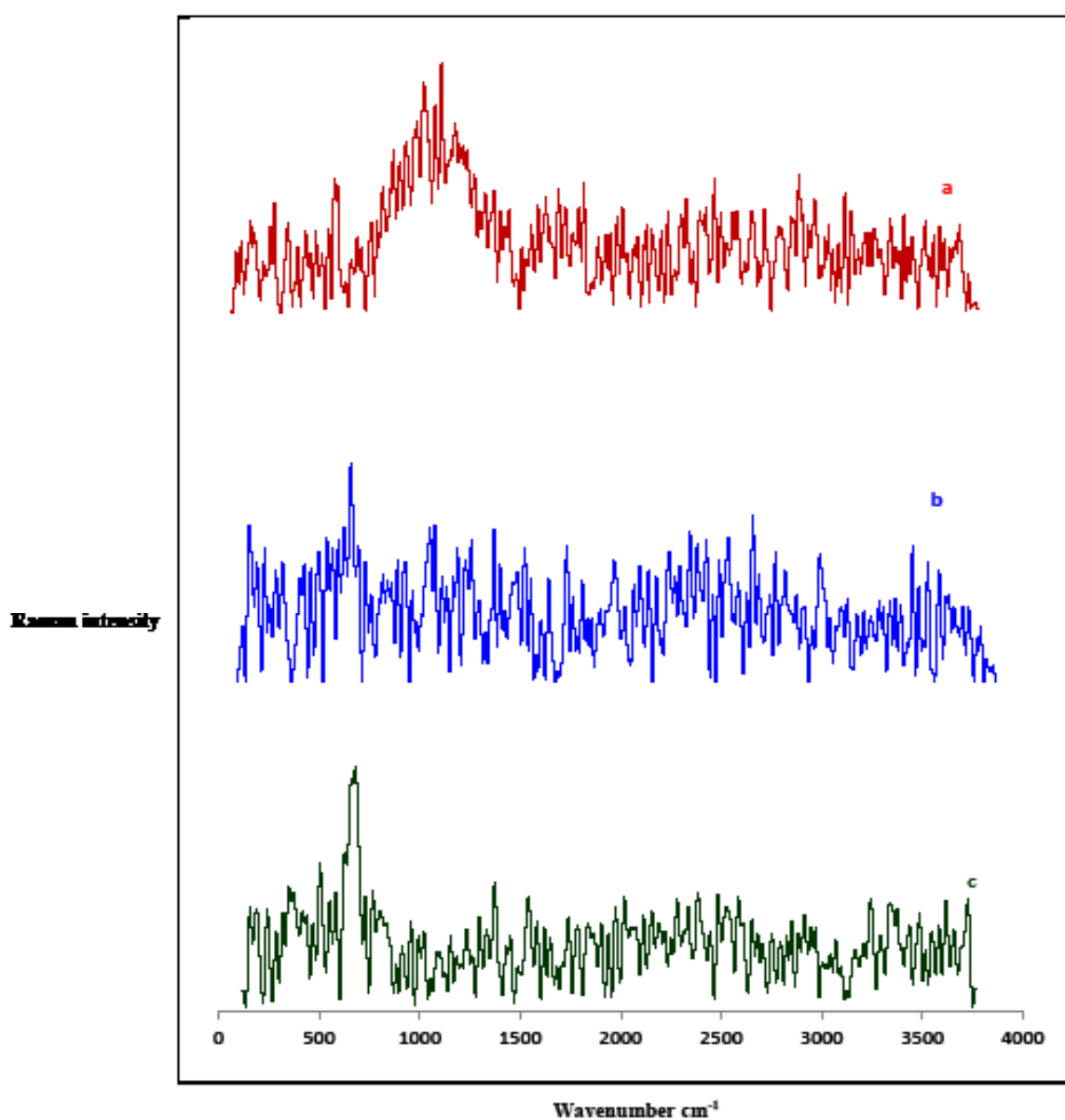


Fig. 13. Raman spectra of (a) toner, (b) toner + red Mac (hand engraving) and (c) toner + blue Huhua.

This is because of the appearance of ink from toner gaps present on the top layer of toner as in Figs. (8a-8d), perhaps composes a new mixture of toner and ink which formed a new spectrum of some peaks differs than pure toner peaks[22].

As we seen before, Raman spectroscopy gives a good result in case the ink brands listed in Table 5 crossed over toner. Also, used in determining the sequence of ink brands listed in Table 6 crossed over toner which not detected by the digital microscope. Totally, Raman spectroscopy is a complementary technique with a digital microscope to detect the sequence of

intersecting strokes resulting from toner and all brands of stamp-pad ink listed in Tables (5-6)[23].

On the other hand, the results for the first factor was studied shows that the changing of the brand and model of printer led to no variations on the results observed at the intersection point as in Figs. (14a-14d). This is may be due to the main constituents of the toner for all laser printers are similar to each other regardless of their brands and models. Furthermore, the applied technique for fixing the toner on the paper surface in all used printers is the same[24].

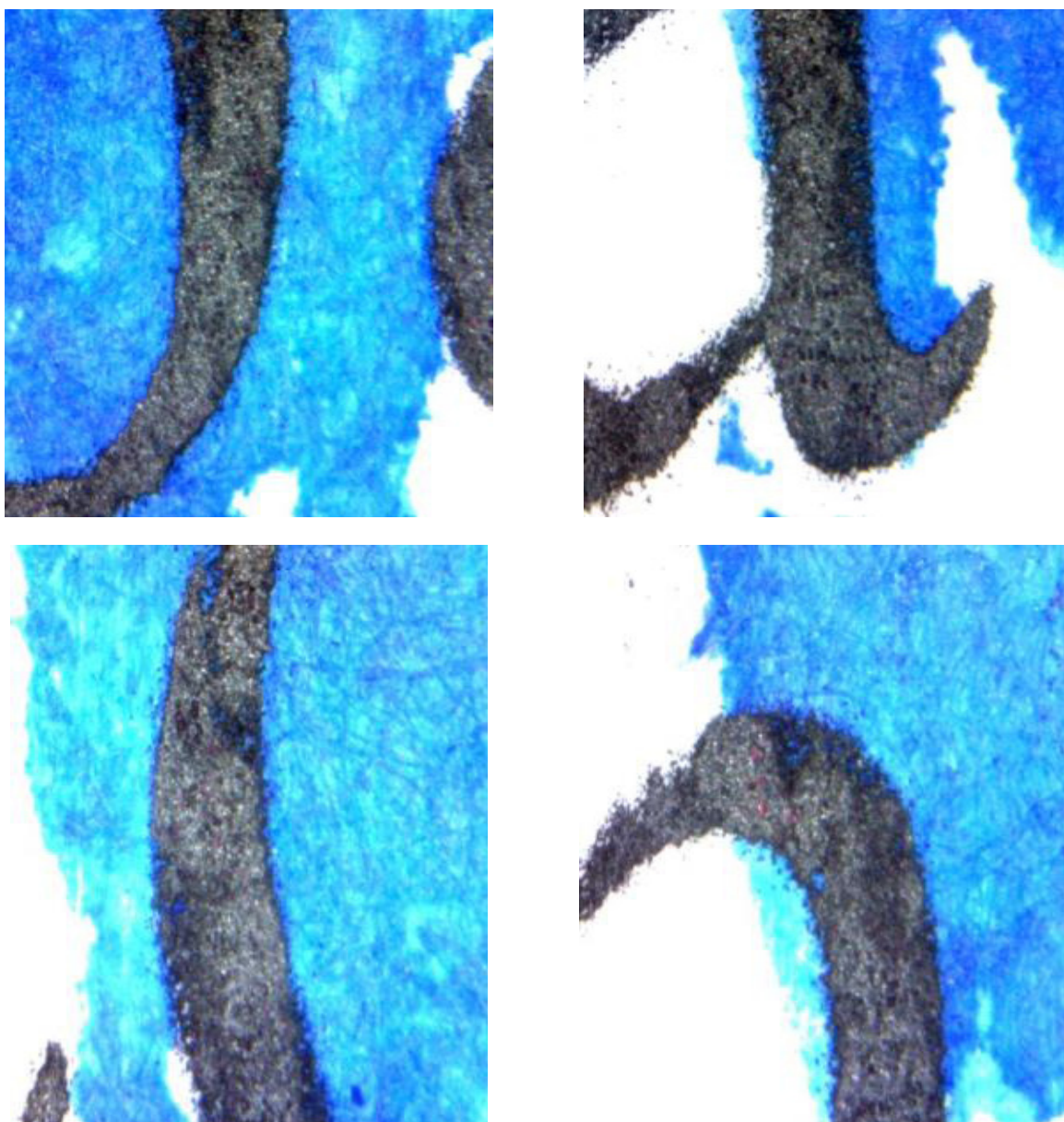


Fig. 14. The ink spreading of blue RADEX metallica ink over a toner of (a) Xerox phaser 6700, (b) HP 2300, (c) HP 1018 and (d) Epson M 2000 under the digital microscope

Also, when the variable nature of the paper surface studied when blue Uni-laknock MITSUBISHI ink over the toner, no changes in the observed results as in Figs. (15a–15c) because the composition of the toner, stamp-pad inks and used three papers surfaces that do not prevent the fixing of toner and absorption of stamp-pad inks on the surface of all paper and cellulose fibers.

Additionally, the five-time gaps between the first and the second strokes show that no effect recorded on the observed results as in Figs. (16a-16d). This is perhaps due to 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.

Furthermore, the variations in the type of materials and manufacture methods of seal have no changes in the results. All materials and manufacture methods of seal used in our study almost have the same manner for the amount of ink stamped on paper regardless the type of its surface and the existence of toner before ink as in Figs. (17a-17d).

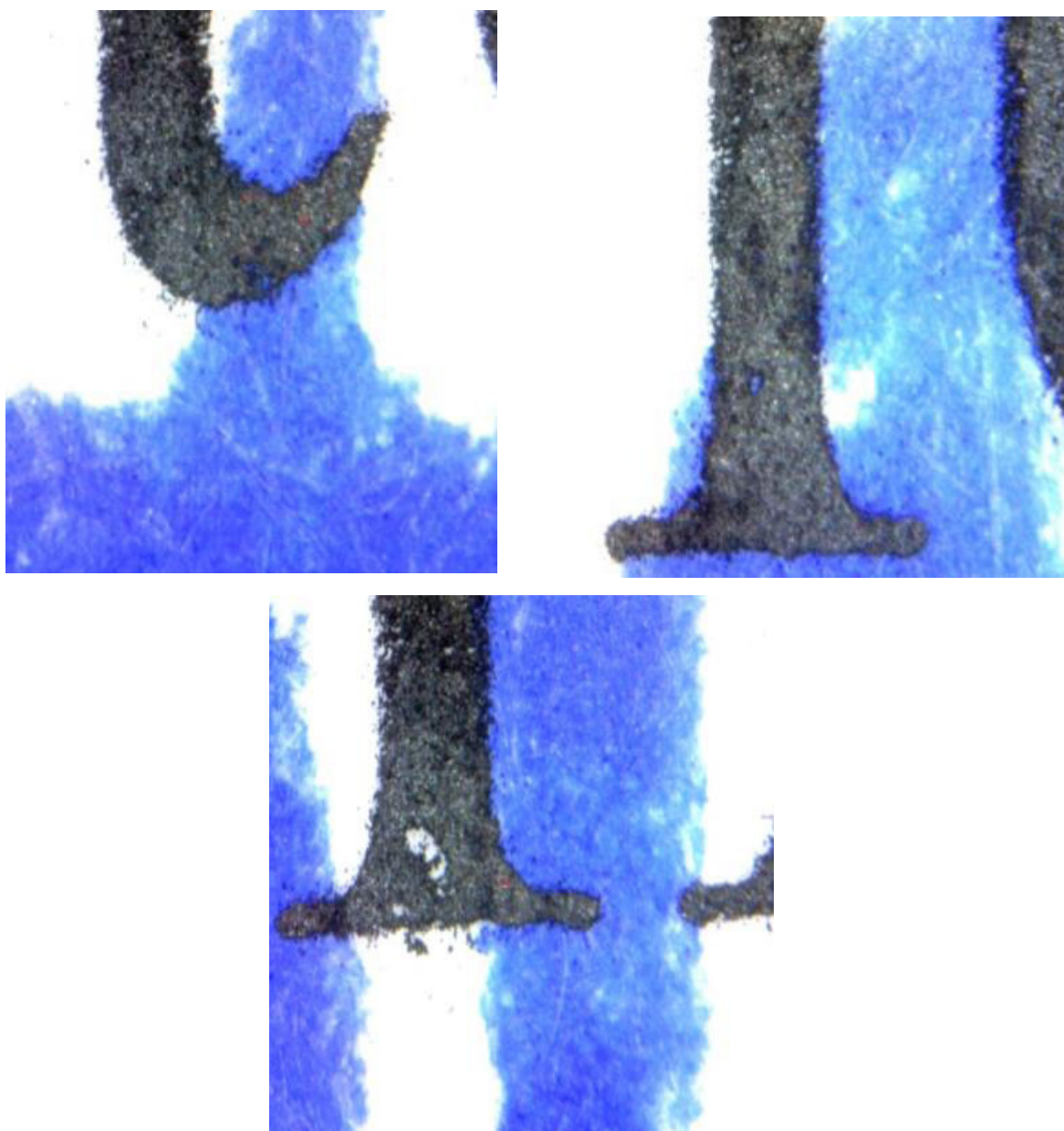


Fig. 15. The specular reflection when blue Faber Castell ink over the toner on (a) coated, (b) non-coated and (c) smooth papers under the digital microscope.

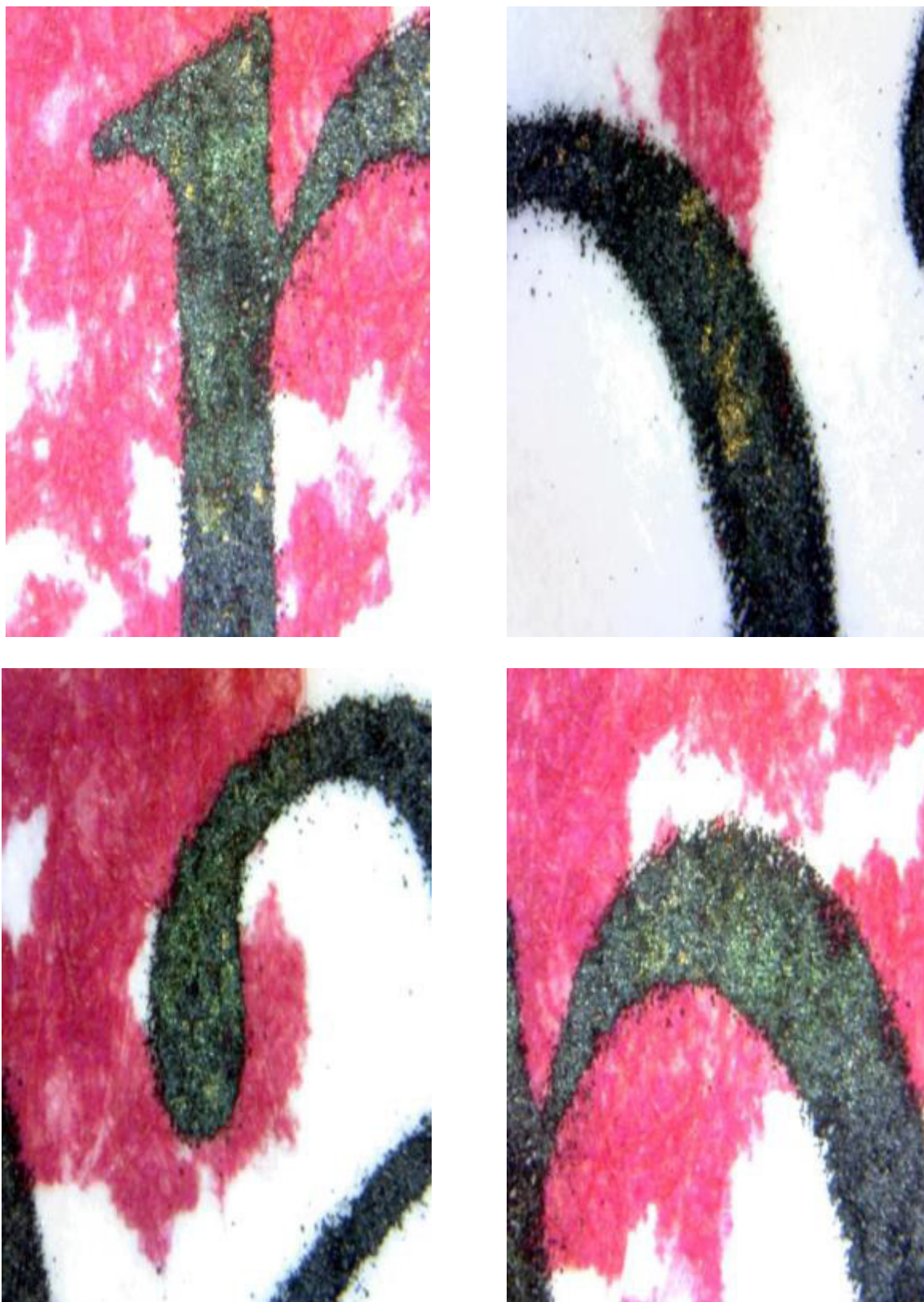


Fig. 16. The continuity gloss of toner and ink when red flower ink is over a toner after (a) 5 mins, (b) 2 hours, (c) 10 days and (d) 30 days under the digital microscope.

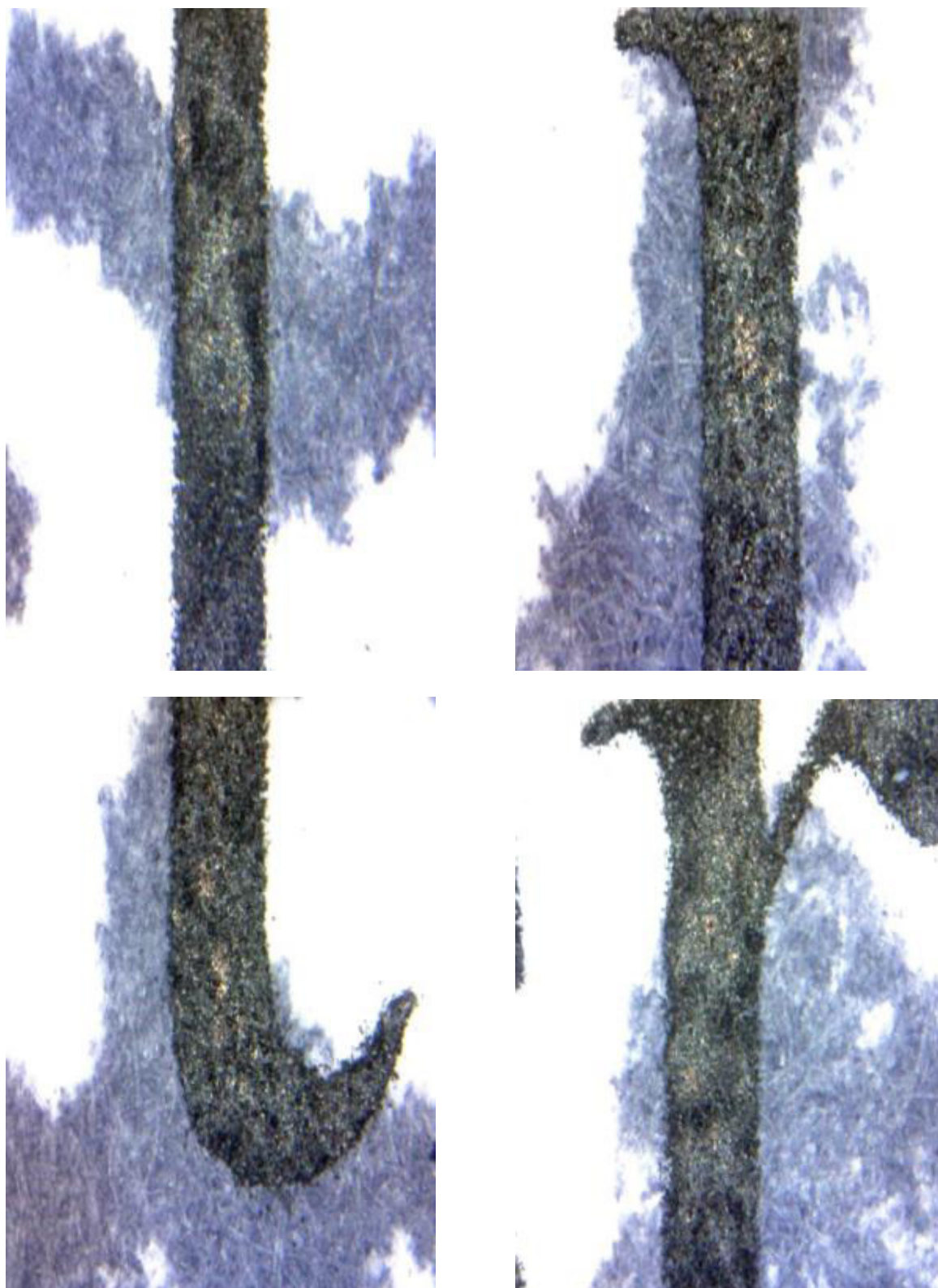


Fig. 17. The ink spreading when black Huhua ink stamped by the metal seal of (a) chemical engraving, (b) hand engraving, rubber seal of (c) laser engraving and (d) chemical engraving over a toner under the digital microscope.

Blind Testing

The outcomes of blind samples observed from the intersection of the toner and stamp-pad inks pen inks were prepared by volunteer person. The sequences of the toner and ink strokes were unknown for some authors before examination and known to volunteer person only. Blind samples were examined by authors to detect the error rate and may be present in using the combination of a digital microscope and Raman spectroscopy. The results recorded by authors were compared with the key of blind samples prepared by volunteer person. Firstly, with respect to Raman spectroscopy, it successful in determines the sequence of strokes intersection. Secondly, with respect to the digital microscope, all authors give correct answers for a sequence of strokes intersection in all blind samples except in case of five samples of red ink over toner, four samples of black ink over toner and seven samples of toner over ink. All results obtained by authors are listed in Table 6.

These blind testing' shows that the limitations of using the digital microscope alone in determining the sequence of ink over toner especially for some brands of stamp-pad red ink due to optical illusions and varied perceptions of a human. The combination of Raman spectroscopy which producing curves based on chemical differences between toner and ink with a digital microscope has high advantages and arrived at one-hundred percent in the determination of a sequence of strokes intersection. Therefore, these results are useful in the felid of forensic document examination.

Conclusion

The samples investigated were consisting of two types of intersection resulting from toner and stamp-pad inks. More than one factor perhaps effect on the determination of the sequence of intersecting strokes was studied. Two nondestructive methods; digital microscope and Raman spectroscopy were used in the samples investigated. Four physical characteristics are investigated under the digital microscope. The results of Raman spectroscopy detect the sequence of intersecting strokes regardless of the colors or pen brands.

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التمييز الفيزيائي للتقاطع غير المتجانس الناتج من احبار الاختام واحبار الطابعات الليزرية

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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 لمتته 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 تتابع بين الجرات اللاحقة والسابقة. في هذه الدراسة تم تحضير العينات التي تحتوي علي نوعي التقاطع للتونر مع احبار الاختام (التونر فوق الحبر والحبر فوق التونر) ودراسة بعض العوامل المؤثرة علي تحديد هذا التقاطع مثل ماركة وموديل الطابعات الليزرية، ماركة احبار الاختام، نوع سطح الورق المحضر عليه العينات، المسافة الزمنية بين الجرة الاولى 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 احبار الاختام. 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 احبار الاختام مع التونر بغض النظر عن الوانها. وعليه فان دمج الميكروسكوب الرقمي مع مطياف الرمن في تحديد تتابع تقاطع جرات التونر مع احبار الاختام هي طريقة غير متلفة، جديدة، واعدة في مجال فحص المستندات للتأكد من تعرضها للتزوير من عدمه.