Egypt.J.Chem. **Vol. 62**, No. 6. pp. 1069 - 1091 (2019)

Egyptian Journal of Chemistry
http://ejchem.journals.ekb.eg/

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,

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Received 5/12/2018., Accepted 6/1/2019
DOI: 10.21608/EJCHEM.2019.6532.1549

lubricants, surfactants, corrosion inhibitors, sheer 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 toit›s 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.

Brand of printer	Model of brand	Color of ink
Hp LaserJet	2300dn	Black
Hp LaserJet	P1005	Black
HP LaserJet	1018	Black
LEXMARK	E350d	Black
EPSON aculaser	M 2000	Black

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^{o}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

Determination the sequence of intersecting heterogeneous strokes is taken place depending on the difference in composition of gel ink, toner and the next change of second ink due to the presence of the first ink [17,20]. The following physical characteristics are observed at the intersection point under the digital microscope:

Specular Reflection

The first physical characteristic observed under the digital microscope is a specular reflection which considered a mirror-like reflection from the surface of the intersection point. Generally, the specular reflection observed when the gel ink strokes crossed over the toner strokes[23].

Two types of the specular reflection are observed at intersection point depending on the brands and colors of gel pen ink. The first type, when the brands of gel pen listed in Table 4 crossed over the toner, a complete (full) specular reflection is showed up.

This is due to the luminescent materials present in the ink composition almost fill all parts and accumulate on toner stroke at intersection point as in Fig. 1.

The black and blue colors of all pen brands listed in Table 4 and red color of Uni-ball gel impact

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

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

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

Nature of paper surface	Brand of paper	Size of paper	Weight of paper (g/m^2)
non-coated	MultiOffice	A4	80
coated	Antalis	A4	80
smooth	Antalis	A4	80

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 brand scomposes 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 G388xincai	Black, red and blue
3ADIMOND	Black, red and blue

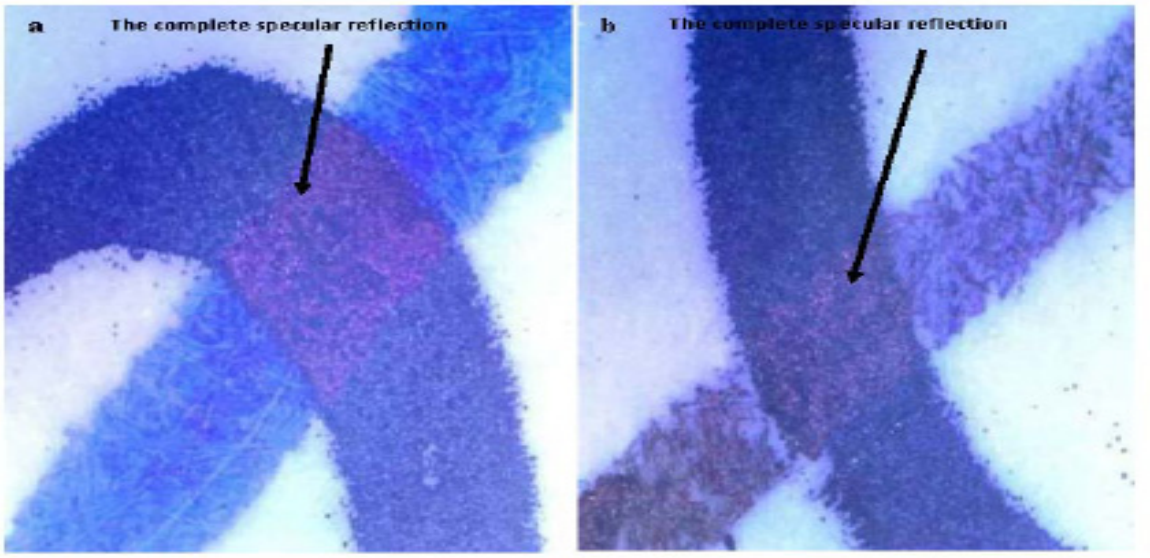

Fig. 1. The complete specular reflection when (a) blue Uni-ball gel and (b) black ROTO G-Plus inks crossed over the toner under the digital microscope

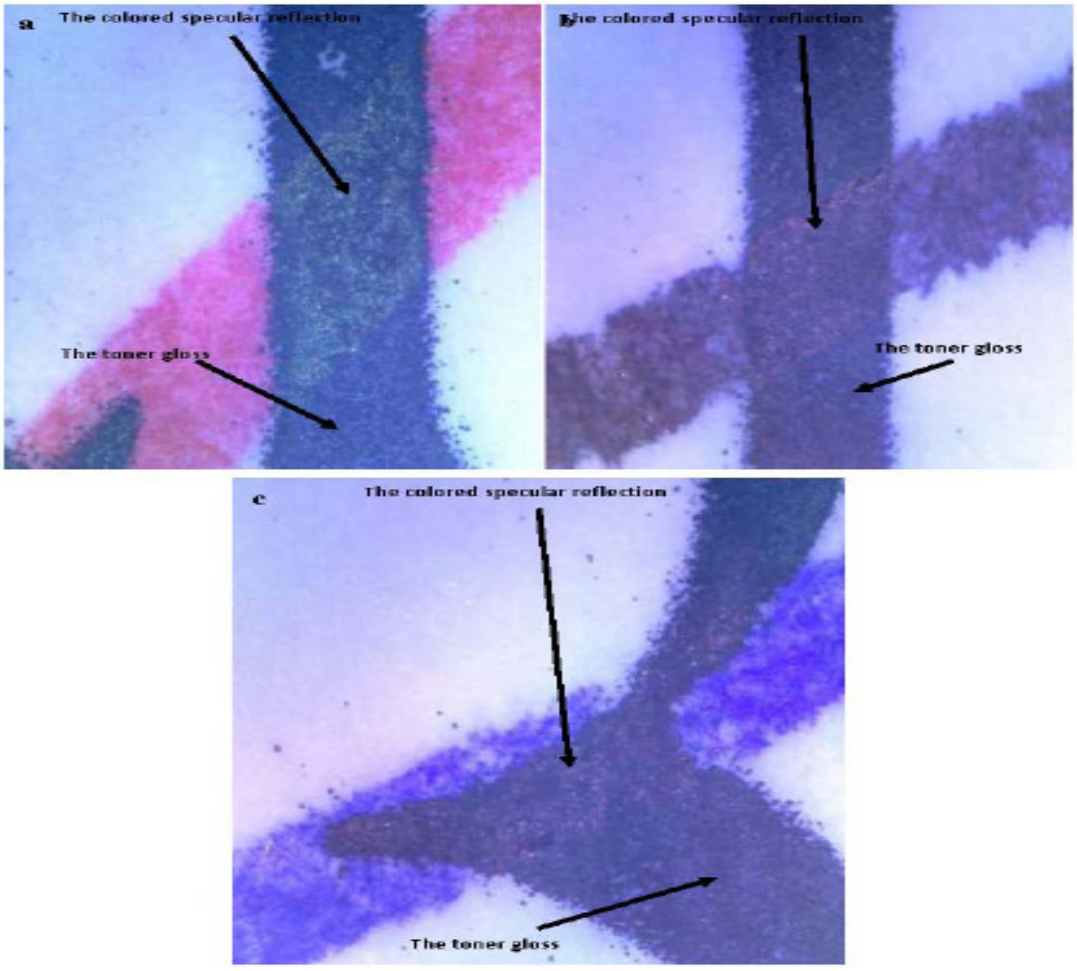

Fig. 2. The colored specular reflection when (a) red Uni-ball gel, (b) black LINC glycer and (c) blue LINC glycer inks crossed over the toner under the digital microscope

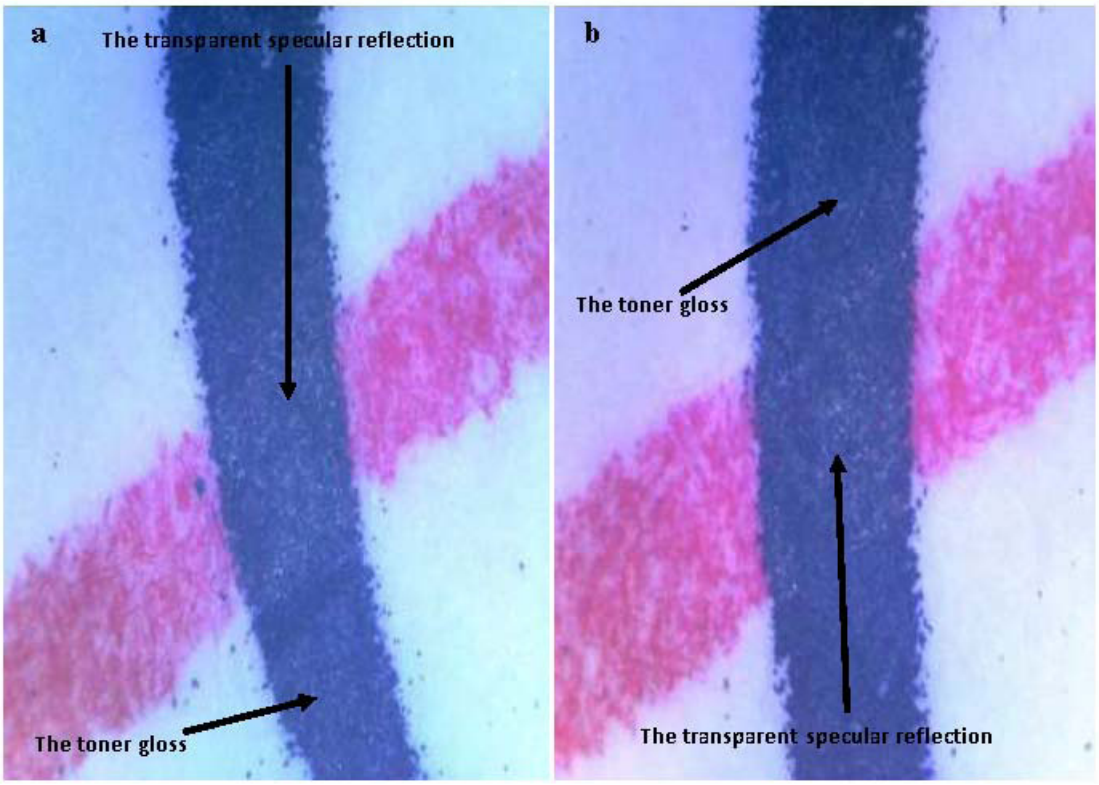

Fig. 3. The transparent specular reflection when red (a) ROTO G-Plus and (b) CLARO SIGMA inks crossed over the toner under the digital microscope.

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.

Ina 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

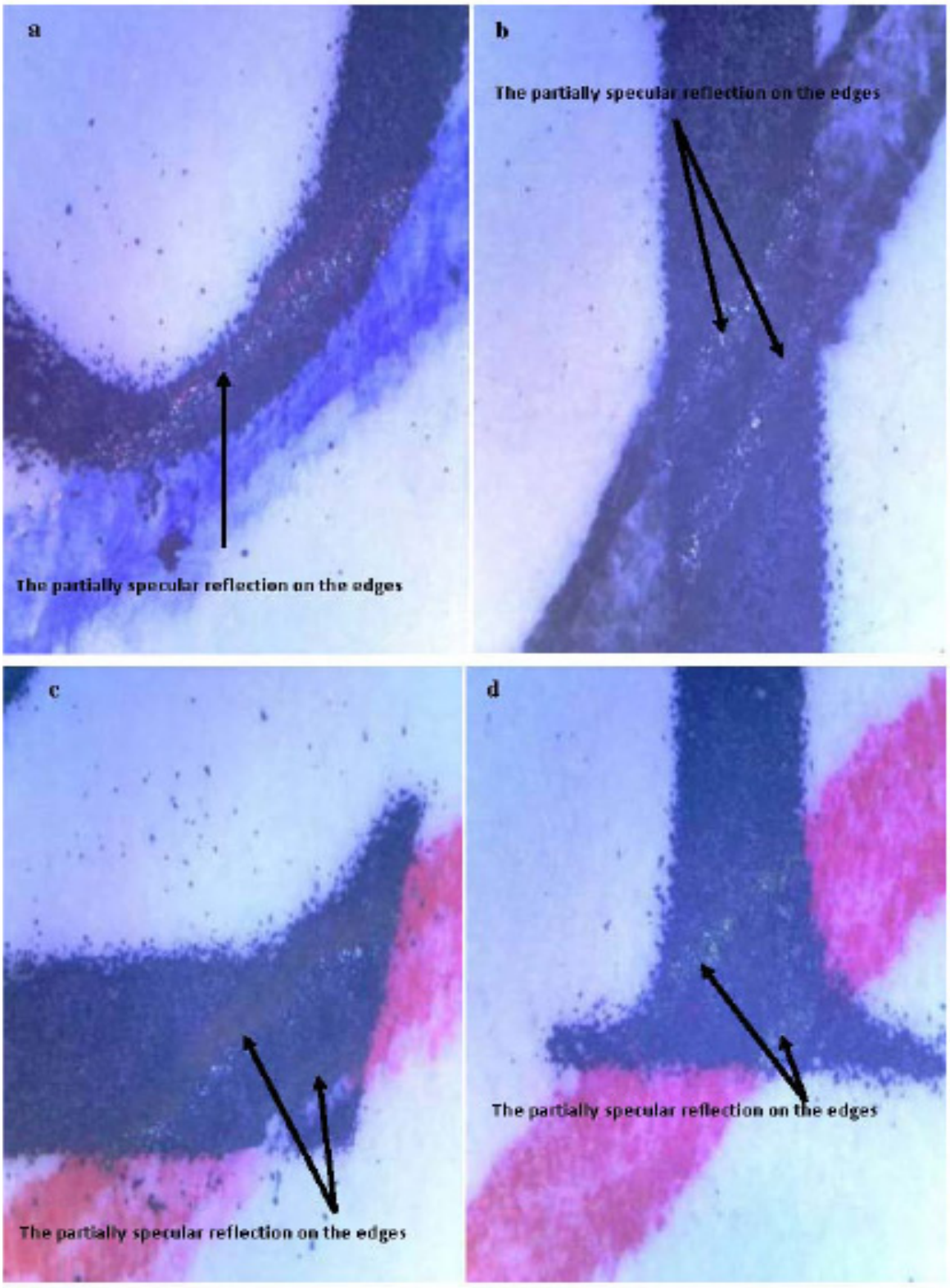

Fig. 4. The partially specular reflection when (a) blue BOIL G388 xin cai, (b) black MICRO, G-388 0.5 mm, (c) red 3A DIMOND and (d) red Pentel energel BL 107 inks crossed over the toner under the digital microscope

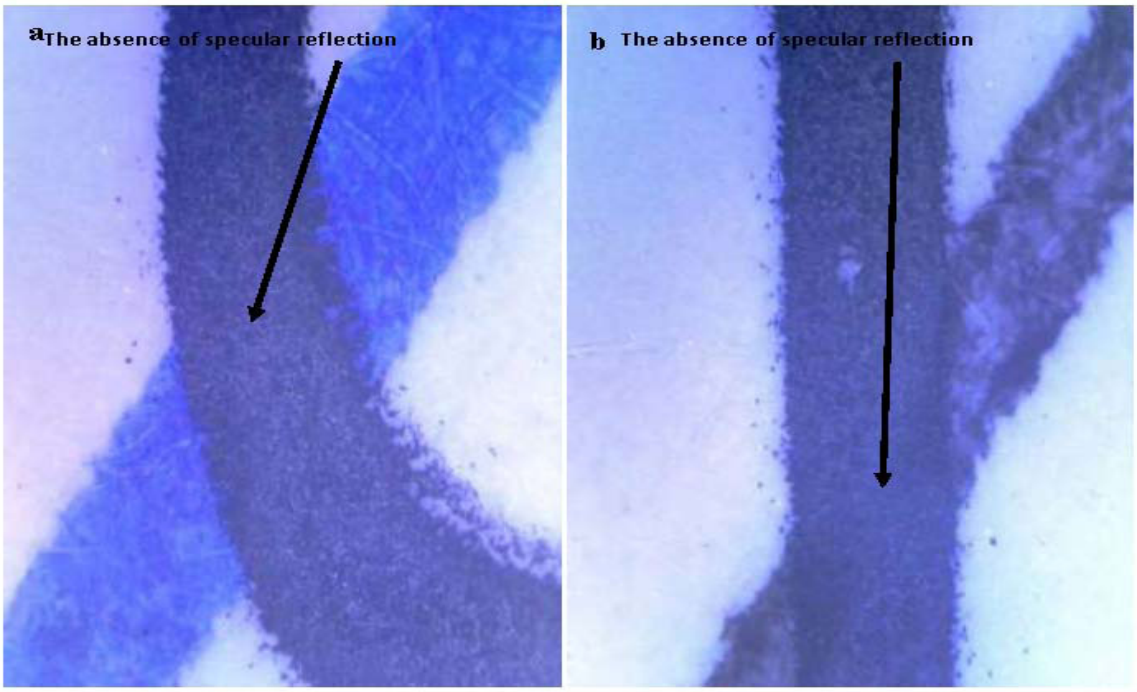

Fig. 5. The absence of specular reflection when the toner is over (a) blue Uni-ball Gel impact and (b) black MICRO, G-388 0.5 mm inks under the digital microscope.

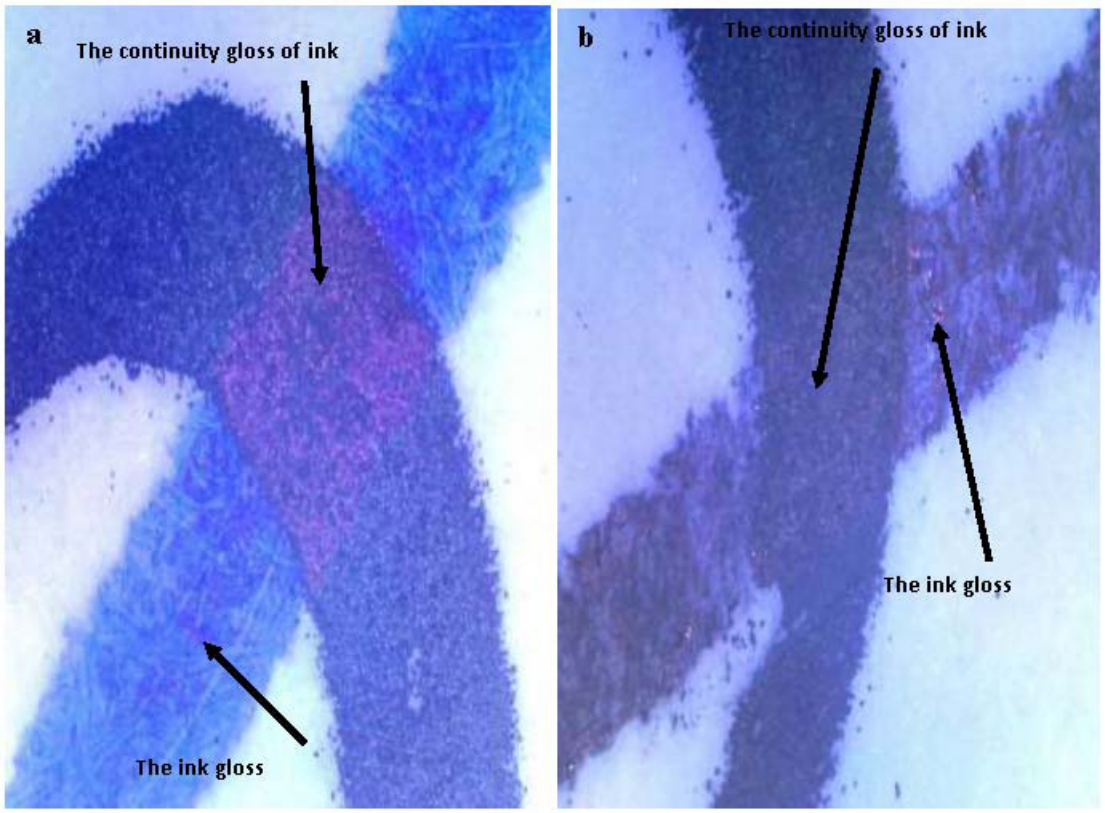

Fig. 6. The continuity gloss of ink when (a) blue Uni-ball Gel impact 1 mm and (b) black ROTO G-Plus crossed over the toner under the digital microscope.

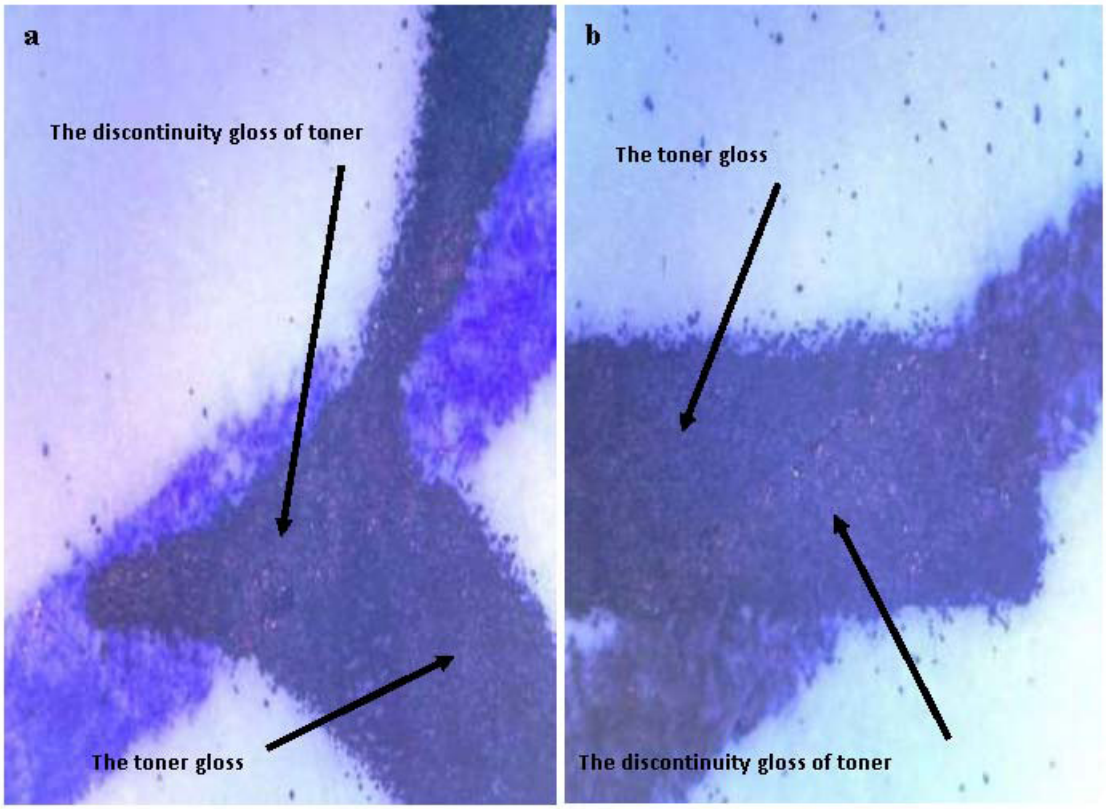

Fig. 7. The discontinuity gloss of toner when (a) blue LINCglycerand (b) black ROTO G-Plus inks crossed over the toner under the digital microscope

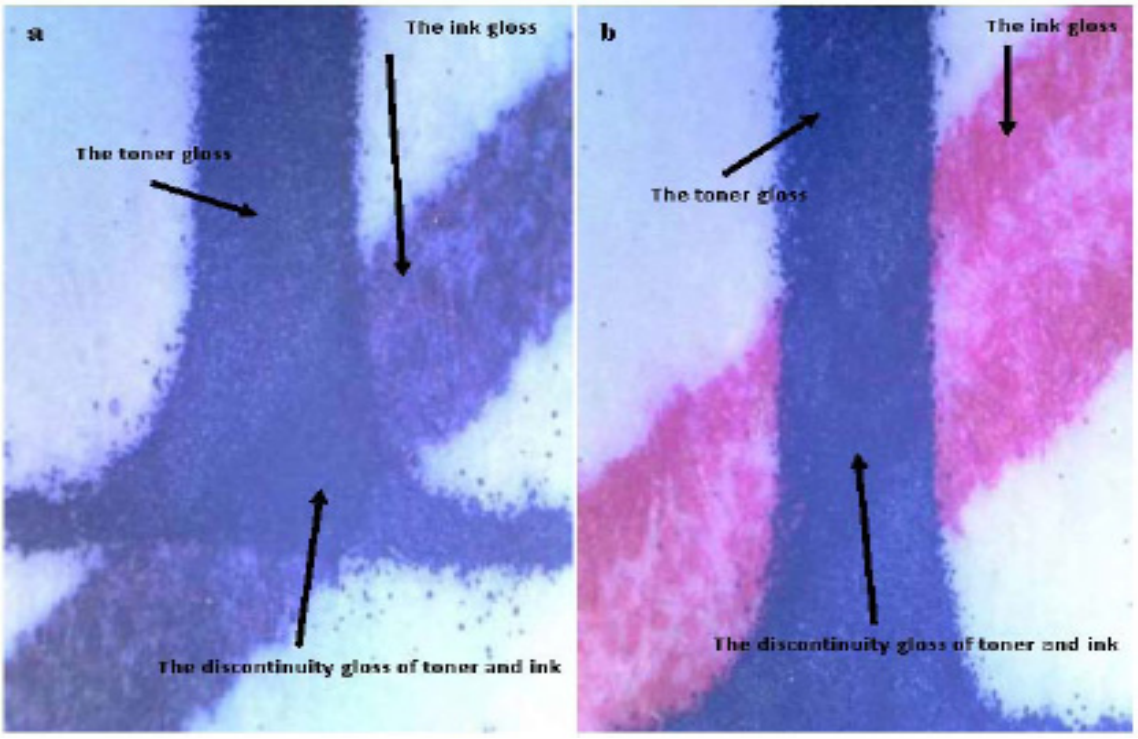

Fig. 8. The discontinuity gloss of toner and ink when the toner is over (a) black ROTO G-Plus and (b) red SAKRM.A.M (GL – 1220) under the digital microscope.

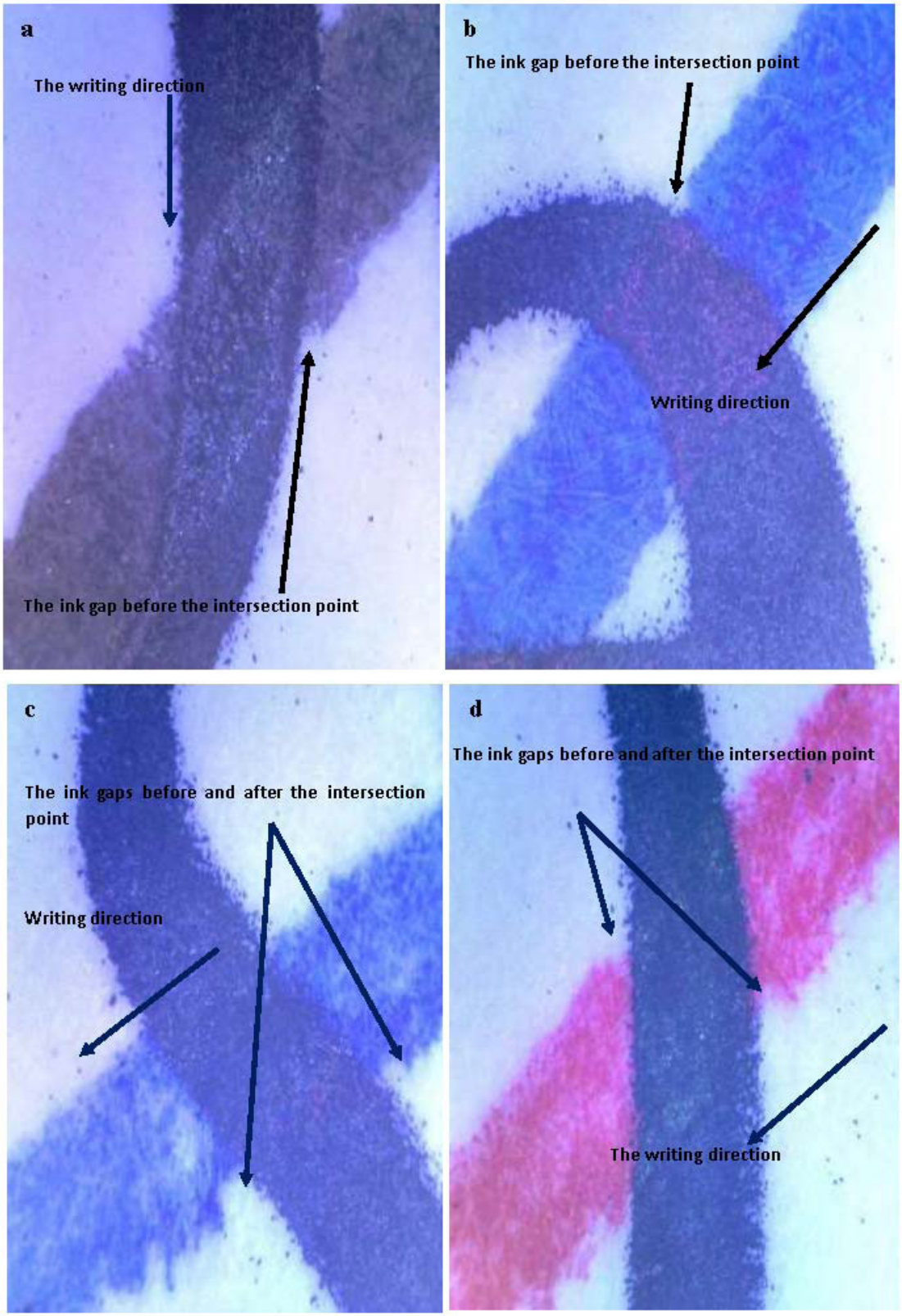

Fig. 9. The ink gaps when (a) black, (b)blue Uni-ball Gel impact, (c) blue and (d) red Pentelenergel BL 107 inks crossed over the toner under the digital microscope.

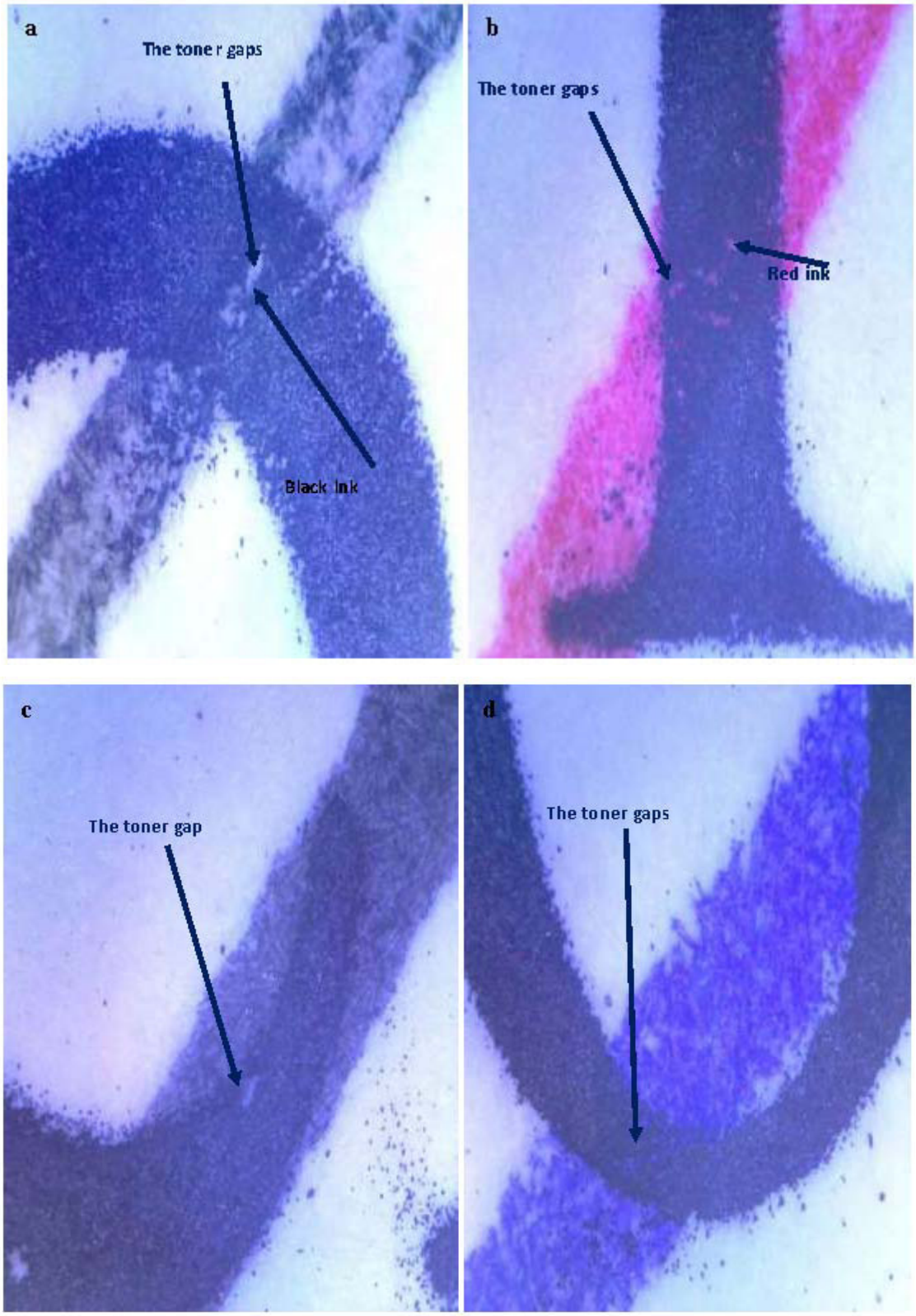

Fig. 10. The toner gaps when a toner is over (a) black 3ADIMOND, (b) red Pentelenergel BL 107, (c) black Uni-ball Gel impact and (d) blue CLARO SIGMA under the digital microscope.

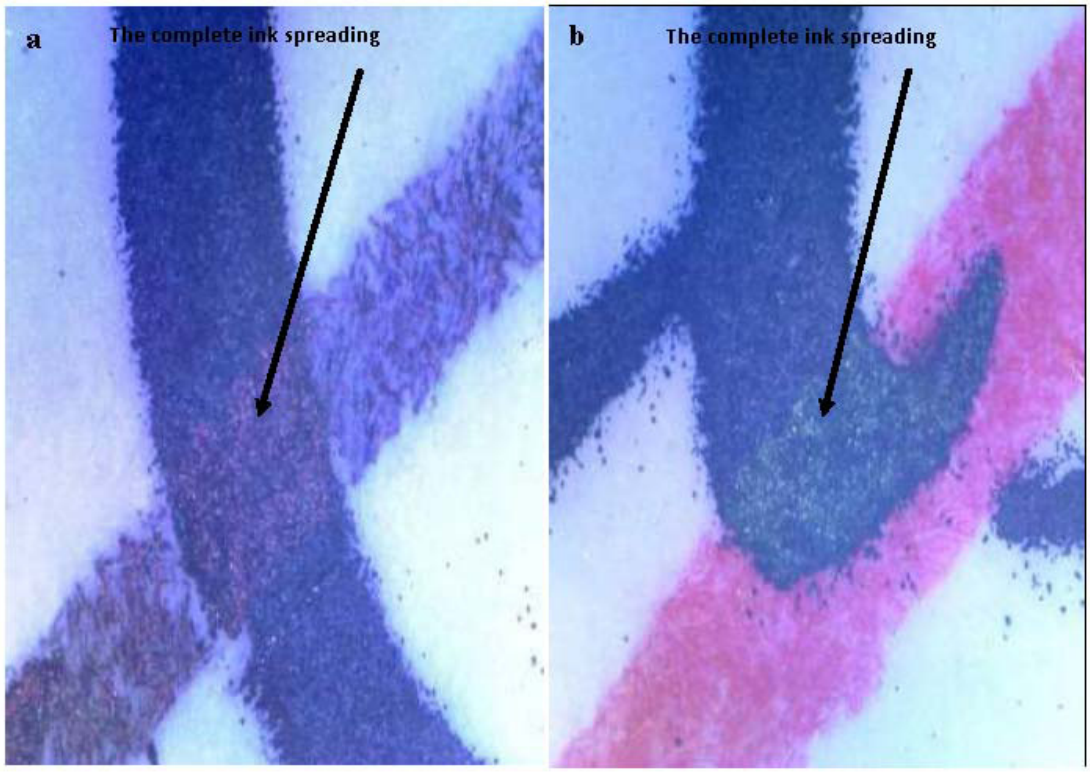

Fig. 11. The complete ink spreading when(a) black POP EURO and (b) red Uni-ball Gel impact crossed over the toner under the digital microscope

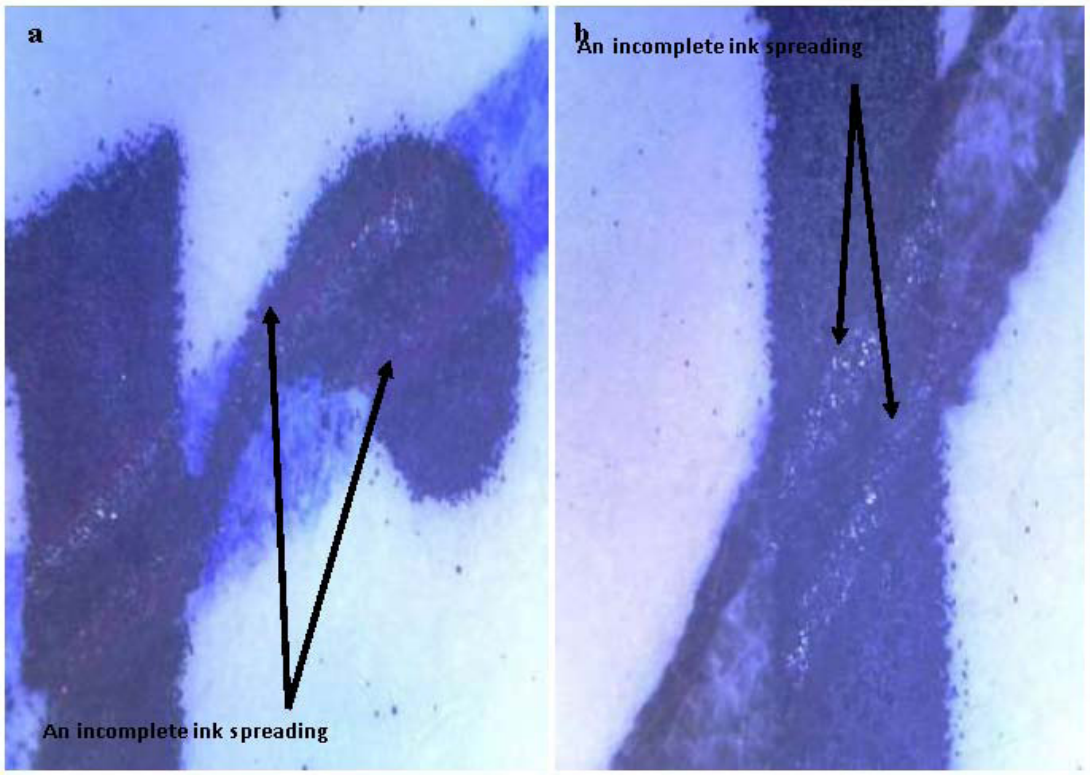

Fig. 12. An incomplete ink spreading when (a) blue BOIL G388xincai and (b) black MICRO G-388 inks crossed over the toner under the digital microscope.

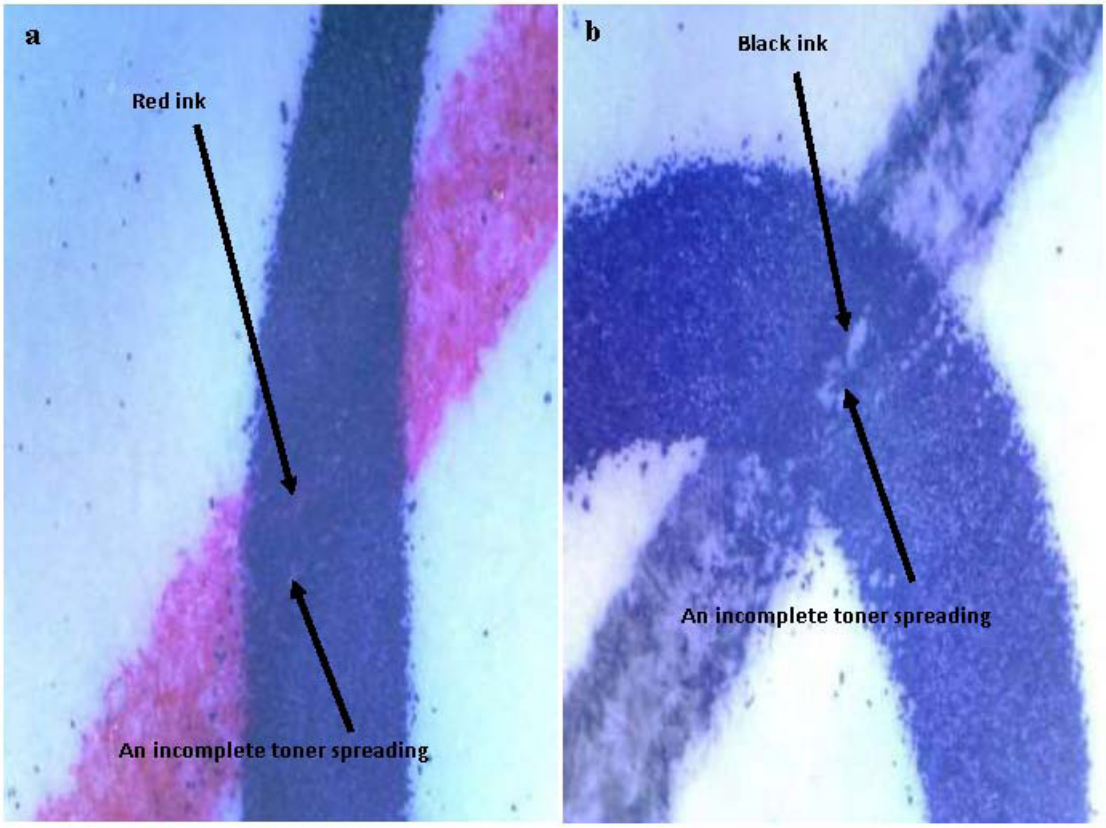

Fig. 13. An incomplete toner spreading when a toner is over (a) red POP EURO and (b) black 3A DIMOND inks under the digital microscope

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

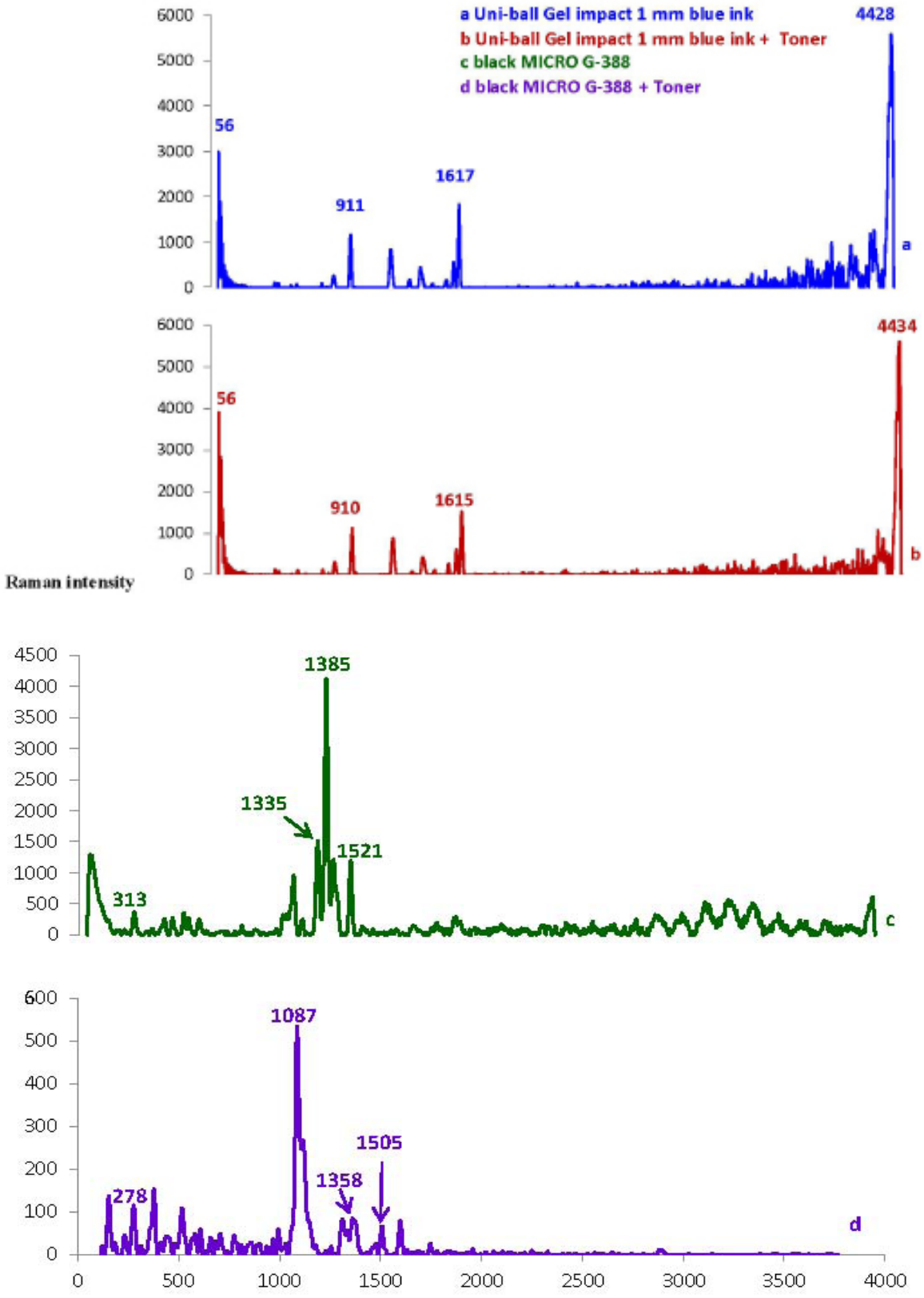

Fig. 14. Raman spectra of (a) Uni-ball, (b) Uni-ball crossed over the toner, (c) MICRO G-388 and (d) MICRO G-388 crossed over the toner.

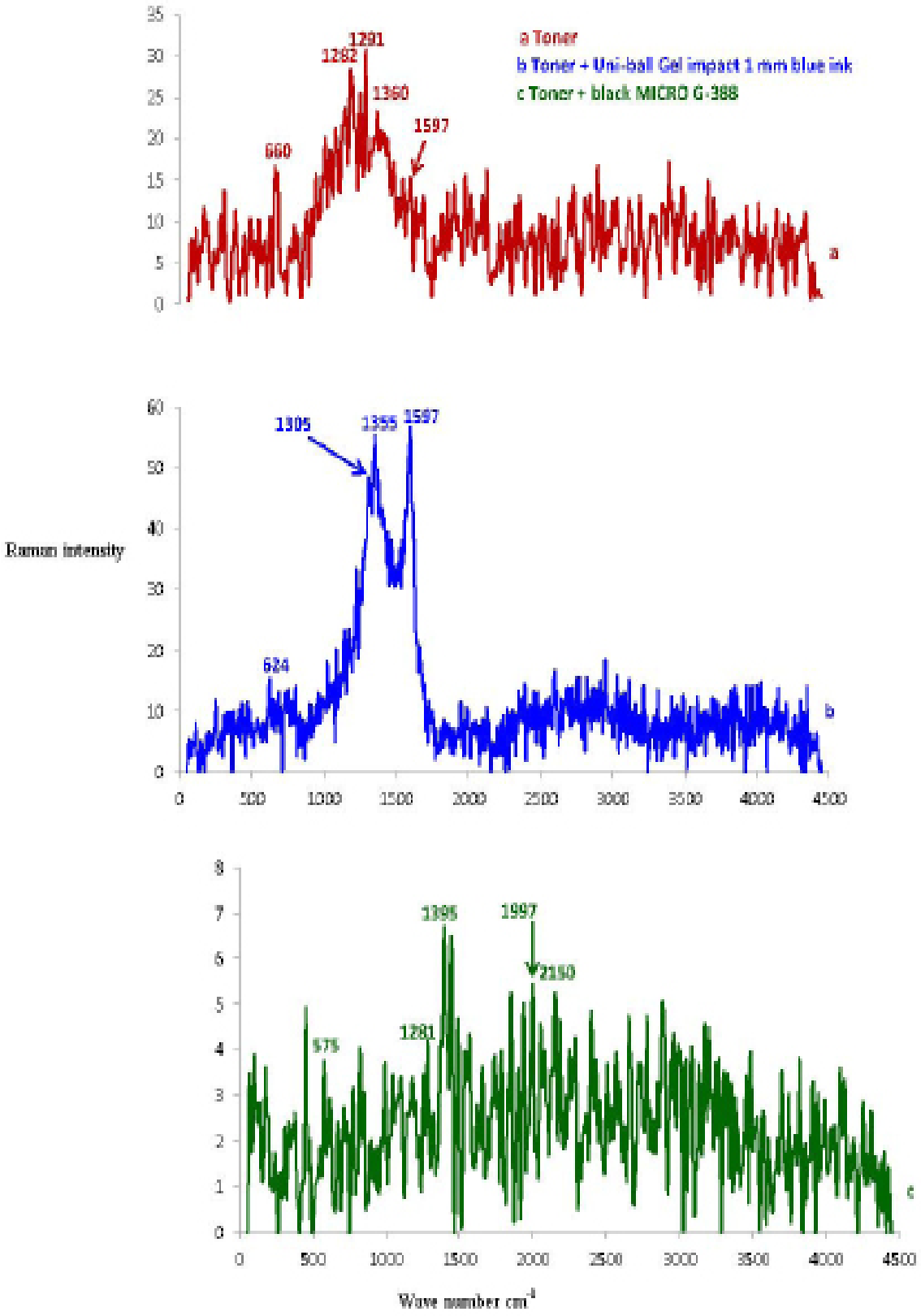

Fig. 15. Raman spectra of (a) toner, (b) toner crossed over Uni-ball and (c) toner crossed over MICRO G-388inks.

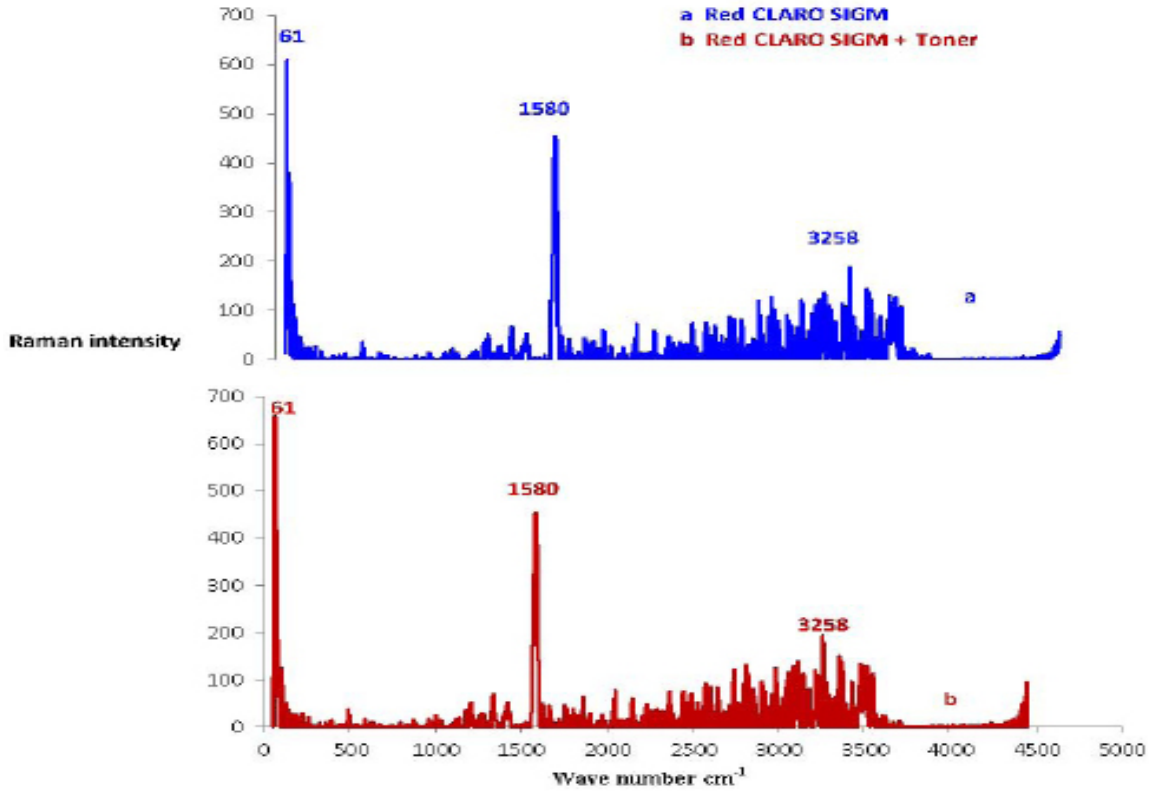

Fig. 16. Raman spectra of (a) red CLARO SIGMand (b) red CLARO SIGMink crossed over the toner

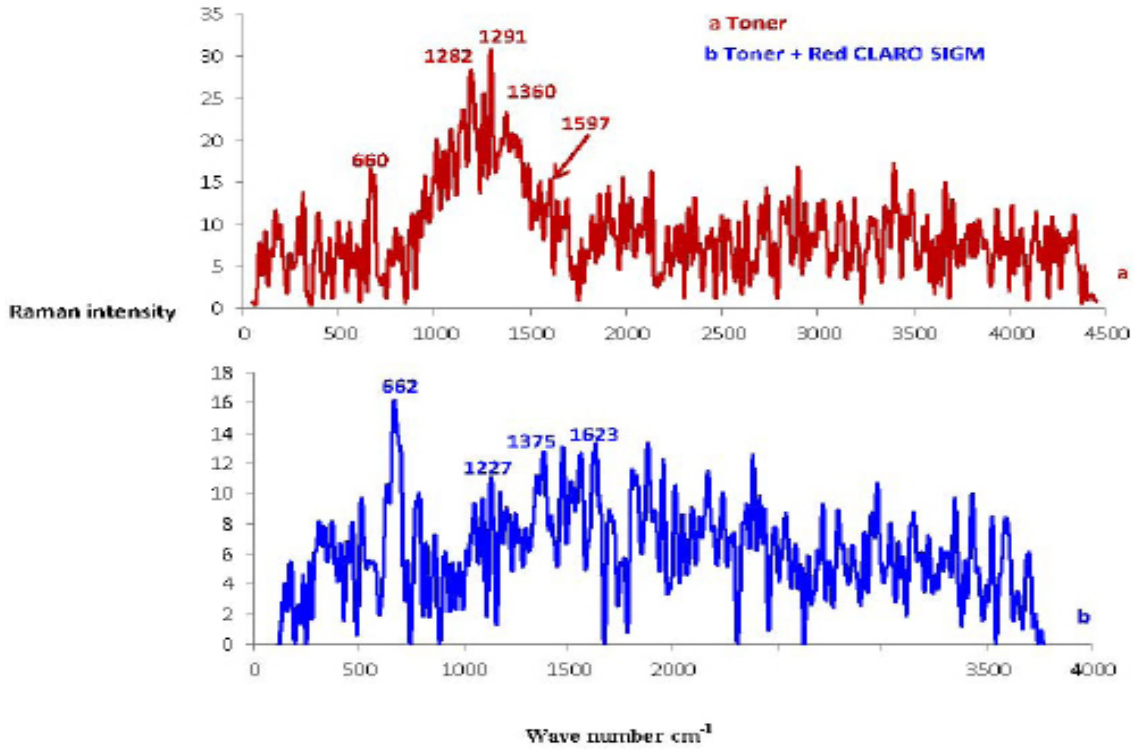

Fig. 17. Raman spectra of (a) toner and (b)the toner printed over red CLARO SIGM ink.

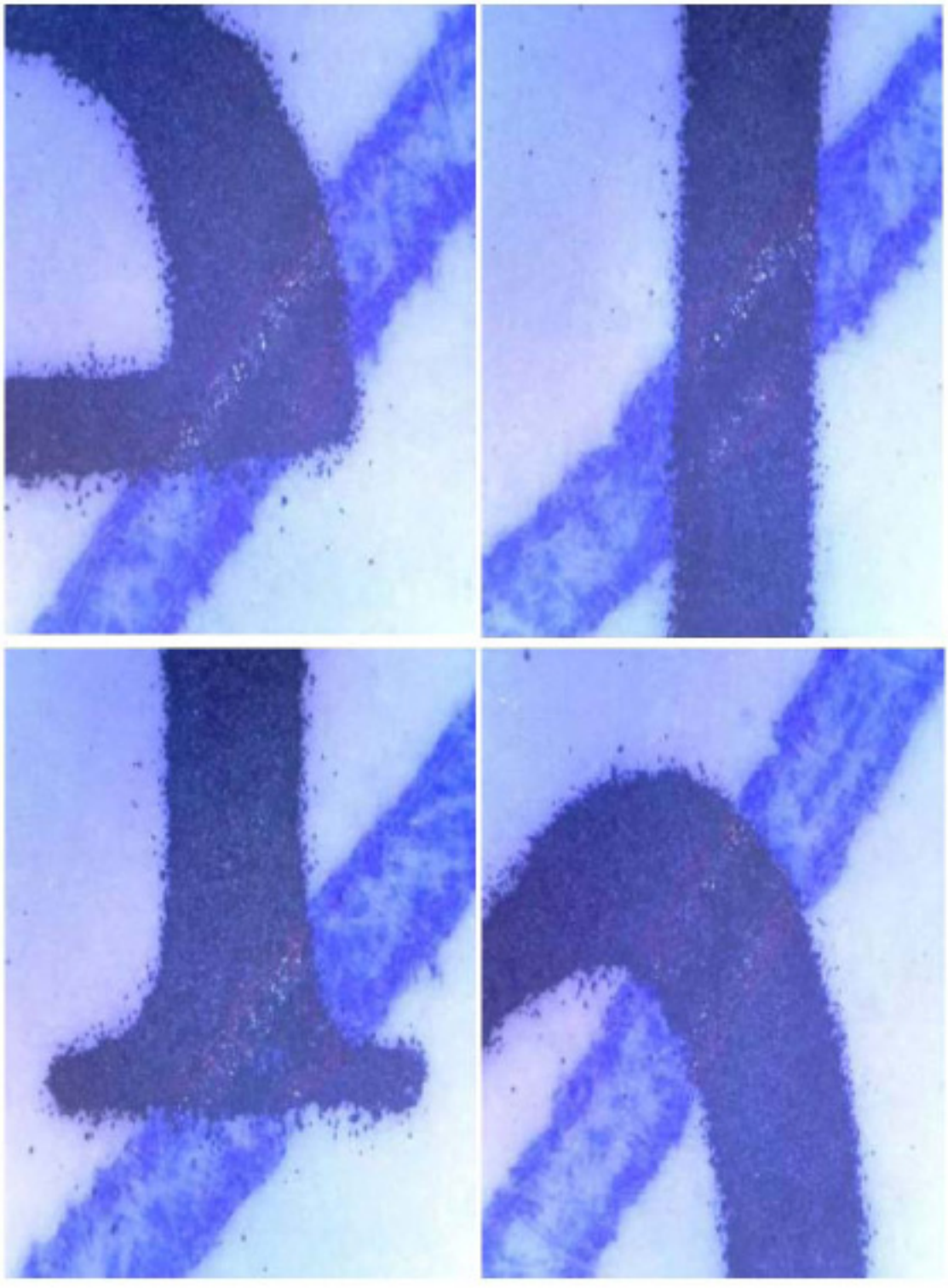

Fig. 18. The gloss of blue MICRO G-388 ink over a toner of (a) Xerox phaser 6700, (b) HP 2300, (c) HP 1018 and (d) Epson M 2000 under the digital microscope.

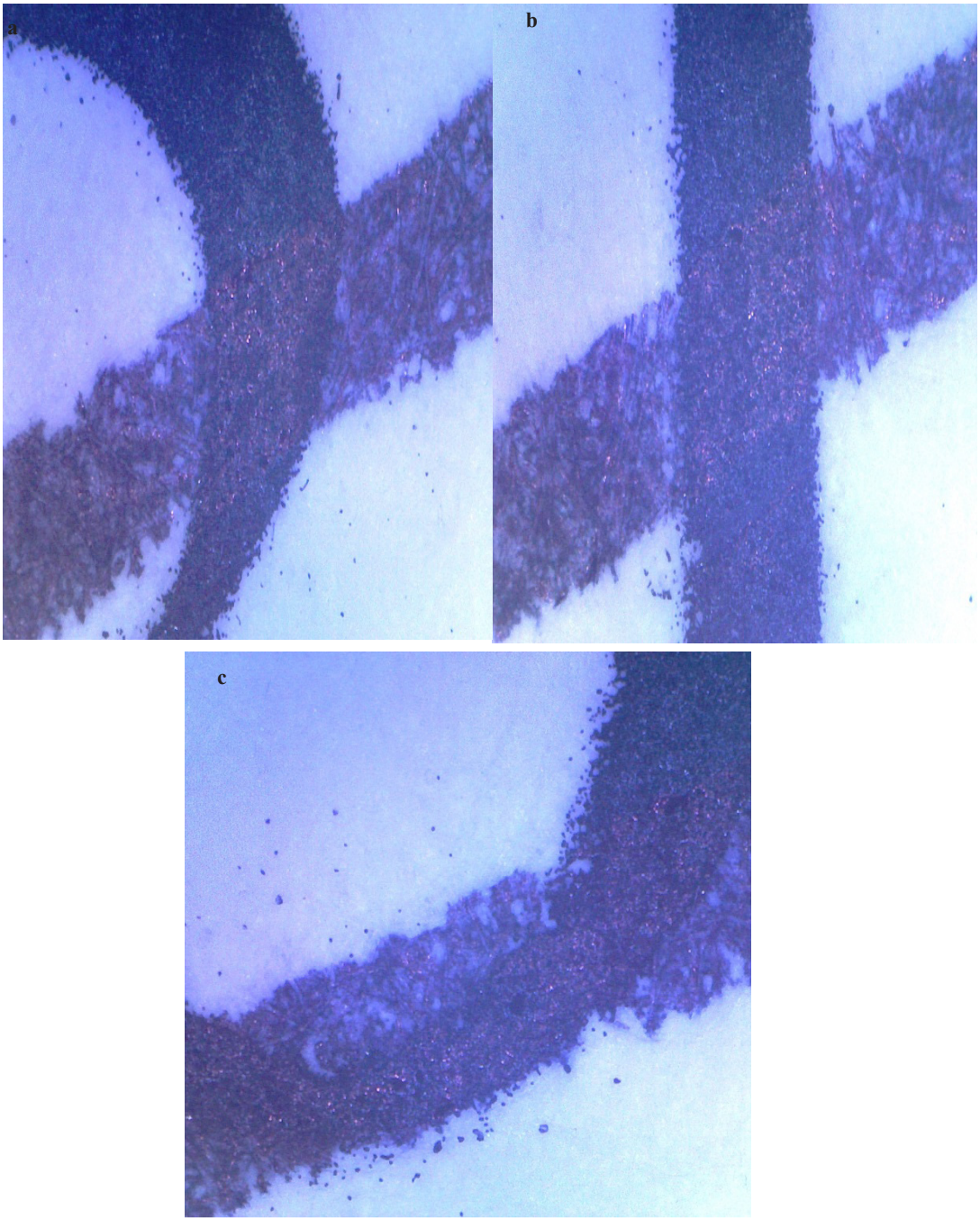

Fig. 19. The spreading of CLARO SIGMA ink over the toner on (a) coated,(b) non-coated and (c) smooth papers under the digital microscope.

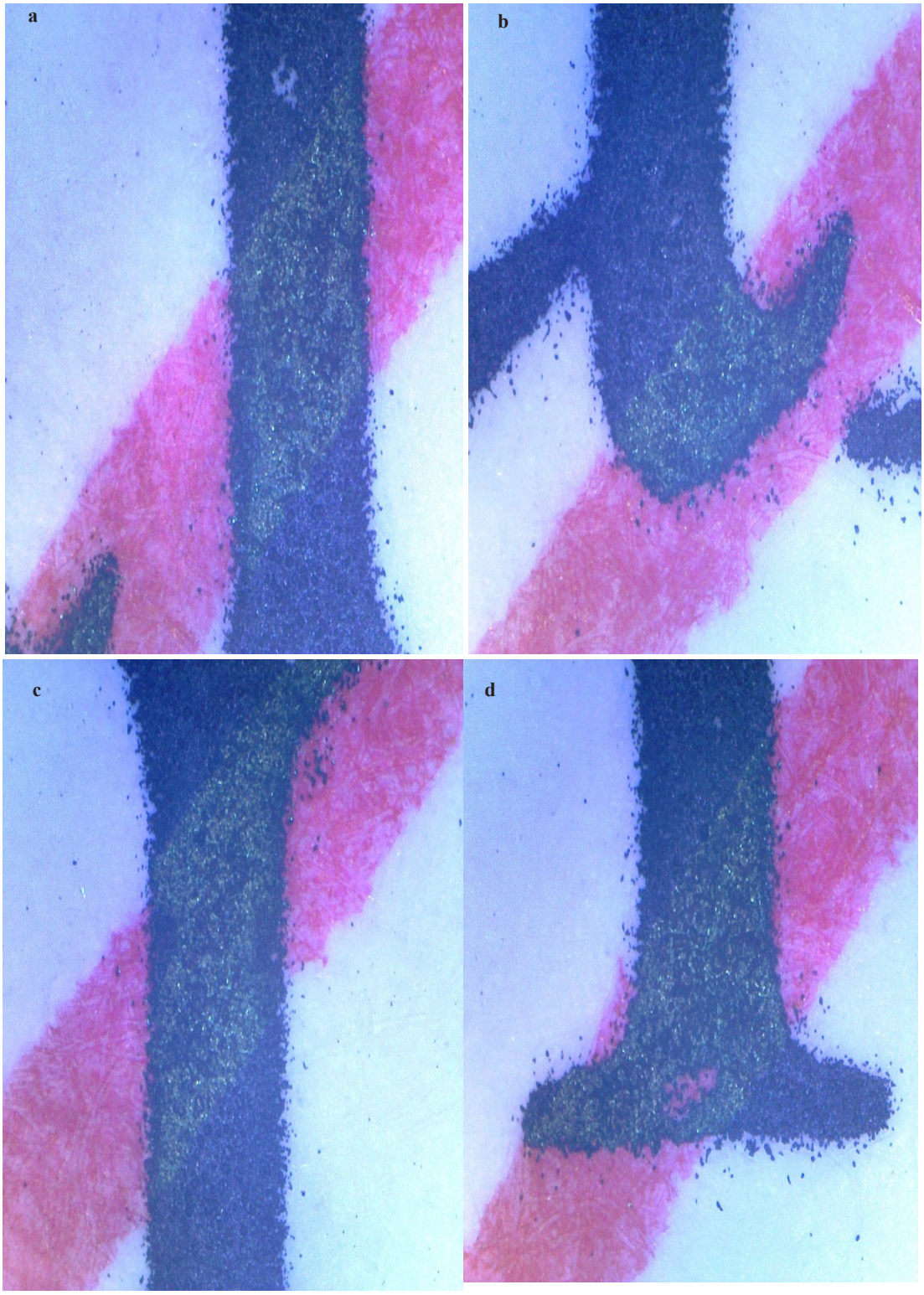

Fig. 20. The specular reflection when red Uni-ball Gel impact crossed over a toner after (a) 5 mins, (b) 2 hours, (c) 10 days and (d) 30 days under the digital microscope.

TABLE 6. Results of blind samples.

No of blind Samples	Order of strokes	Authors									
		Microscope	Raman	Microscope	Raman	Microscope	Raman	Microscope	Raman	Microscope	Raman
20	Toner over red ink	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct
20	Toner over black ink	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct
20	Toner over blue ink	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct
20	Black ink over toner	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct
20	Red ink over toner	5 Inconclusive 15 Correct	Correct	5 Inconclusive 15 Correct	Correct	5 Inconclusive 15 Correct	Correct	5 Inconclusive 15 Correct	Correct	5 Inconclusive 15 Correct	Correct
20	Blue ink over toner	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct	Correct

toner and ink appears from toner gaps as in Fig. 10 [30].

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

effect from the second stroke.

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

1. Kaur R., Saini K., Sood N.C. Determining the sequence of intersecting gel pen and laser printed strokes — A comparative study. *Sci Justice*. **49**, 286 (2009).

2. Verma N., Kumar R., Sharma V.Analysis of laser printer and photocopier toners by spectral properties and chemometrics. *Spectrochim Acta A Mol Biomol Spectrosc*. **196**, 40 (2018).

3. Liu Y.Z., Yu J., Xie M.X., Liu Y., Hana J., Jing T. Classification and dating of black gel pen ink by ion-pairing high-performance liquid chromatography. *Chromatogr Sep Tech*. 1135.57 (2006).

4. Wu Y., Zhou C.X., Yu J., Liu H.L., Xie M.X. Differentiation and dating of gel pen ink entries on paper by laser desorption ionization- and quadruple-time of flight mass spectrometry. *Dyes Pigm*. **94**. 525 (2012).

5. Williamson R., Djidrovska D., Ledic A., Brzica S., Antikj V., Hofer R., Almirall J. Characterization and identification of luminescent components in inks using various analytical techniques for the study of crossed-line intersections. *Forensic Chem.* **3**, 28 (2017).

6. Verônica A., da Silva G., Talhavini M., Zacca J.J., TrindadebB.R., Braga J.W.B. Discrimination of black pen inks on writing documents using visible reflectance spectroscopy and PLS-DA *J. Braz. Chem. Soc*. **25**, 1552 (2014).

7. da Silva V.A.G., Talhavini M., Peixoto I.C.F., Zacca J.J., Maldaner A.O., Braga J.W.B. Non-destructive identification of different types and brands of blue pen inks in cursive handwriting by visible spectroscopy and PLS-DA for forensic analysis. *Microchem.* **116,** 235 (2014).

8. Sharas I, Aboul El.Magd A.A., Moustafay M, Bakr A.A, Aziz I.A. Application of some physical techniques for forensic discrimination of printer toner. *Egypt J Chem*. **61**, 131(2018).

9. Ataeefard M., mohammadian F.N. Producing fluorescent digital printing ink: Investigating the effect of type and amount of coumarin derivative dyes on the quality of ink. *J Lumin*. **167**, 254 (2015).

10. Williamson R., Raeva A., Almirall J.R. Characterization of printing inks using DART-Q-

TOF-MS and Attenuated Total Reflectance (ATR) FTIR. *J Forensic Sci*. **61**, 706 (2016).

11. Assis A.C.A., Barbosa M.F., Nabais J.M.V., Custo´ dioA.F., Tropecelo P. Diamond cell Fourier transform infrared spectroscopy transmittance analysis of black toners on questioned documents. *Forensic Sci Int*. **214**, 59 (2012).
12. Kubiak I. The influence of the structure of useful signal on the efficacy of sensitive emission of laser printers. *Measure*. **119**, 63 (2018).
13. Palus J.Z., Kunicki M. Application of the micro-FTIR spectroscopy, Raman spectroscopy and XRF method examination of inks. *Forensic Sci Int.* **158**.172 (2006).
14. Gazy M.B., El-Zawawy W.K., Elsayed A.E., Eldebss T.M.A., Helal M.R., Rashed K. Application of Some Polymer latexes in Preventing Paper Documents Forgery. *Egypt J Chem*. **62**, 1 (2019).
15. Lee J., Nam Y.S, Min J., Lee K., Lee Y. TOF-SIMS Analysis of Red Color Inks of Writing and Printing Tools on Questioned Documents. *J Forensic Sci.* **6,** .822 (2016).
16. Brito L. R., Martins A.R., Braz A., Chaves A.B., Braga J.W., Fernanda M. Critical review and trends in forensic investigations of crossing ink lines. *Anal Chem*. **54**, 33 (2017).
17. 1Ozbek N., Braz A., Lo´ pezM.L., Ruiz C.G. A study to visualize and determine the sequencing of intersecting ink lines. *Forensic Sci Int*. **234**, 39(2014).
18. Li B. An Examination of the Sequence of Intersecting Lines using Microspectrophotometry. *J Forensic Sci*. **61,** 809-814 (2016).
19. FABIAÑSKA E., KUNICKI M. Raman spectroscopy as a new technique for determining the sequence of intersecting lines. *Prob Forensic Sci*. LIII, **60** (2003).
20. Vanco L., Kadlecíková M., Breza J., Belányiová E., Michniak P., Reháková M., Ceppan M. Raman mapping as a tool for discrimination of blue writing inks and their cross lines. *Vib.Spectrosc.* **79**, 11(2015).
21. Kaur R., Saini K., Sood N.C. Sequencing the intersections of printed strokes with writing instrument strokes using DocuCentre expert (PIA 6000-EDF). *Sci Justice.* **53**, 206 (2013).
22. Ozbek N., Braz A., Lo´ pezM.L., Ruiz C.G. A study to visualize and determine the sequencing of intersecting ink lines. *Forensic Sci Int*. **234**, 39 (2014).
23. Kaur R., Saini K., Sood N.C. A study for establishing the sequence of superimposed lines: Inkjet versus writing instruments. *Forensic Sci Int.* **193**, 20 (2009).
24. Kaur R., Saini K., Sood N.C. Application of Video Spectral Comparator (absorption spectra) for establishing the chronological order of intersecting printed strokes and writing pen strokes, *Sci Justice.* **53,** 212 (2013).
25. Liu R.H., Dai S.A., Chang F.J., Cheng W.T., Shih Y.F. Investigation on solubility of polymeric binder of xerographic toner and deinking by emulsion process. *J Taiwan Inst Cheme*., **40,** 90 (2009).
26. Lee C.W, Hsu D.J. Measurements of fine and ultrafine particles formation in photocopy centers in Taiwan.Atmos. *Environ.***41**, 6609 (2007).
27. Montani I., Mazzella W., Guichard M., Marquis R. Examination of heterogeneous crossing sequences between toner and rollerball pen strokes by digital microscopy and 3-D laser profilometry. *J Forensic Sci.* **57**, 1002 (2012).
28. Owoc M. Thermic transformations of writing materials. *Prob Forensic Sci*. **53**, 51 (2003).
29. Vaid B.A., Rana R.S., Dhawan C. Determination of sequence of strokes through reflection spectra. *Prob Forensic Sci,* **LXXXVII**, 193 (2011).
30. Bojko K., Roux C., Reedy B.J. An Examination of the sequence of intersecting lines using attenuated total reflectance–fourier transform infrared spectral imaging. *J Forensic Sci*. **53**.1467 (2008).

التطبيق الفيزيائي لطرق غير متلفة في تحديد تتابع تقاطع جرات احبار الطابعات الليزرية مع الاحبار الجيلاتينية

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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 الاقلام جيلاتينية الاحبار (التونر فوق الحبر والحبر فوق التونر) ودراسة بعض العوامل المؤثرة علي تحديد هذا التقاطع مثل ماركة وموديل الطابعات الليزرية, ماركة الاقلام جيلاتينية الاحبار, 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 الزمن. وقد نجح الميكروسكوب الرقمي في تحديد تتابع الجرات باستثناء بعض الالوان لبعض ماركات الاقلام جيلاتينية الاحبار. 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 الاقلام جيلاتينية الاحبار مع التونر بغض النظر عن الوانها. وعليه فان دمج الميكروسكوب الرقمي مع مطياف الرمن في تحديد تتابع تقاطع جرات التونر مع الاقلام جيلاتينية الاحبار هي طريقة غير متلفة، جديدة، واعدة في مجال فحص المستندات للتأكد من تعرضها للتزوير من عدمه.