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Forensic Examination and Identification of Writing Inks on Documents

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Abstract:

Ink analysis plays a significant role in questioned document examination, a discipline of forensic science. Writing inks based on their chemical substances such as solvents, resins, colorants as dyes or pigments, may be distinguished by applying two step methodology including non-destructive or visual examination and destructive or chemical analysis. Both approaches involve identification and comparison techniques by contributing certain advantages to the investigation such as Thin Layer Chromatography has been marked for a long time. However, forensic scientists have the availability of evolutionary methods that led to the use of less destructive techniques including spectroscopy that minimizes the scope of previous separation techniques. This review paper delivers an outline of techniques/methods including spectroscopic and chromatographic development with possible advantages or drawbacks. The acquired results should be evaluated accurately by paying more consideration as providing reliable findings in the court of law is certainly of central significance.

Keywords: Writing Inks, Visual Examination, Chemical Techniques, Instrumental Analysis, Dating Inks.

1. Introduction

Documents are essential part of our lives that accompany us in a number of ways from birth to death certificates. It records everything we do throughout life in the form of letters, personal diaries, receipts, wills, identity documents, bank checks, insurance agreements, promissory notes, loans, leases, bonds/deals, tax returns and testimonies etc. [1,2]. Forensic examination of these documents is important as their validity is often questioned during legal proceedings. Crimes committed with documents are more prevalent with greater influence on society and comprise billions of dollars as compared to any other violent crimes. For example media and news reporters/journalists report homicides, sexual assaults, kidnappings/abduction, fires, blasts, and other violent acts [1]. The current progress in the process of digitalization has established significant modifications in way of exchanging information and replaced traditional means of documents. Nevertheless, there are many cases where conventional methods of transactions or agreements are still practiced involving ink application [2]. Therefore, forensic examination of ink on

writing documents either a typed, printed or handwritten is being applied in a number of civil and criminal investigations.

Forensic examination of ink is a principal piece of investigative and legal cases related to forgeries, counterfeits, ransom notes or threat letter etc. [3]. Forgery involves fabrication and falsification of documents that causes alterations or modifications to an original documents and creates a false document from scratch of existing sample respectively [2]. These fabricated and falsified items include writing materials such as writing ink and paper, laser printer and photocopy toners, typewriter and printer inks, and correction fluids. Writing inks are the most valuable of these materials and are a mixture of composite components that provide preferred appearance, thickness and drying dynamics, and other properties based on different writing instruments [1,2]. Such as fountain pen inks are water based, ballpoint pen inks are based on dyes and organic solvents giving them oil consistency, and rollerball pens use gel inks based on water vehicles with dispersed pigments. However, all of these writing inks irrespective of instrument types, work as crime tools [2].

In questioned document, the main purpose of ink analysis is to isolate or identify the source of ink used on writing documents, comparing two or more ink entries and dating process of ink entry. Here the source could be a particular brand or class of concern writing tool or specific manufacturer batch/year of questioned ink entry [2,3]. The source identification process of an ink sample can be obtained by comparing it directly against specific/known writing or writing instrument involved. For some intelligence based cases, information related to ink manufacturer is collected by comparing ink samples to an ink library/database or control samples. The result interpretation of these comparisons, is influenced by the similar characteristics of inks available in the market [3,4]. Dating of ink has progressed since its first development and now not only it is possible to determine the first manufacturer date of an ink sample but also determines the date when an ink was written on a document in questioned [1,3].

Two step identification process is applied that can be accomplished by conducting preliminary examination prior to chemical analysis that involves optical and visual examination of ink based on its composition like solvents, colorants, additives or resins. The optical and visual properties of an ink sample may be evaluated by examining its responses towards UV, VIS or IR spectrum. However, detailed chemical analysis is required as preliminary examination is typically not enough to answer the queries from judiciary panel. Chemical examination/analysis consists of destructive and non-destructive methods, of which non-destructive methods from a forensic perspective are more often performed as the given evidence sample remains intact and available for further characterization. It is not possible to identify all the components of ink sample thoroughly due to screening characteristics of forensic examination of ink evidence. The results of physiochemical examination are compared for both suspect and reference ink samples instead of identifying individual component. According to Neumann and Genessay, it is not always feasible to link association between ink and suspected manufacturers of writing instruments as the same ink procedures during their production are supplied to many different manufacturers. Thus, leaving a substantial impact on methodical practice, more specifically method that estimates the evidential assessment of

findings. Other factors include unstable entry of ink composition and ink degradation deposited on paper as a result of aging processes triggered by environmental conditions [2].

Ink examination is routinely used by local law enforcement agencies for criminal investigations as well as by lawyers in private sector to date disputed documents relevant to civil cases. For example, altered wills, disputed patents, medical negligence, divorces, tax and stock frauds, insurance schemes, copyright case, and a wide range of other contractual disputes etc. [1,3]. A Society of Forensic Ink Analysts, the SOFIA, was established in August, 1997 in the state of Virginia for the progress of forensic science discipline and methods in identification, comparison and dating of writing inks on questioned documents. The purpose of this organization is to set standards for executing these examinations associated with criminal and civil investigations. The establishment of this society led to better understanding of paper effects on writing ink and dating ink methods, and validation of these methods among the ink chemists and private examiners [1].

2. Ink Examination

Forensic ink analysis is considered as a specified part of forensic questioned document examination as it covers much of the documents involved in number of analysis categories. Therefore, resulting in particular instances where ink analysis is performed distinctly from questioned documents. Such as dealing with murder and criminal cases where threats have been written on walls instead of documents/paper. The inks from these writings are compared with writing evidences provided as known source markers of respective case [1]. Sometimes during an ink examination, it becomes required to identify a particular writing instrument/pen and differentiate between the two inks applied on one document if they are same. Additional changes either long passages or a single digit in the form of writing can prominently alter or modify the sense of words or numbers regarded as money. Sufficient amount of ink is available during manufacture and development process of inks and pens for quality control tests. Whereas, in a forensic document examination ink analysis is performed on a minute quantity of ink wrote on a paper [5]. Therefore, when it comes to the handling of forensic ink evidence, it is preferred that the examiner must be familiar with the field and not to damage the document in questioned [1,5]. Field awareness and advance training will reduce the problems that may arise from semi-destructive property of many inks during analysis process [1].

To analyze and investigate ink evidence, the examiner should identify possible source of ink sample and its general chemical composition. Ink is a notable liquid or paste with different colors developed for writing, printing and drawing purposes. Anciently ink has originated and influenced from many civilization, contributing to our society for the spread of writing and printing knowledge [1]. Writing inks are composed of many functional types of ingredients commonly organic compounds. The key ingredients of inks are dyes or pigments or coloring agents, the device or vehicle to apply ink and the additives that regulate pH, polymerization and viscosity, and to avoid pen blockage. Other components included are solvents, lubricates, resins, biocides, corrosion inhibitors, surfactants, buffers and emulsifying agents formulated from a range of natural and artificial products either organic or in organic. Inks and their chemical composition can be classified based on basis of pen types including liquid,

aqueous, paste or powder. Other basis of classification is it's end use that includes writing and printing inks as two main classes [1,6]. Some of the inks with related substance encountered in forensic examination include ballpoint pen ink, roller ball pen ink, fiber tip pen ink, marker ink, fountain pen ink, gel pen ink, porous tip pen ink, plastic tip pen ink, pencil, rubber stamp ink, ink jet printer ink, letterpress ink, typewriter ink, and copier toner [1].

Other aspects of ink examination involve coordination with forensic handwriting comparisons for the purpose of identifying or eliminating a writer and coordination with latent print examination. Writer elimination or identification is most revealing as it involves analysis of original or an unaltered questioned document. Therefore, ink samples should not be removed from a document before any required handwriting comparison. For example, there is consequence of a conflict if ink sample is removed from the line intersections as these sites have greater quantity of ink with sequence of application regarding handwriting analysis. As a result, this determination becomes impossible because of ink removal from such positions. Similarly, heavy ink deposition site is gooping on an ink line of ballpoint as result of debris or blots on the tip of pen, which indicates the pen movement direction. This aspect of examination can be hindered if these sites are removed prior to handwriting comparison.

Another concern related to questioned document is preservation of its readability. During ink analysis the examiners who work for civil litigation, defense or criminal cases, if remove large section of the line from written samples, they may compromise not only the handwriting comparison but also make the entries unreadable on a document. Doing so will leave photographic record as only means of evidence. Consequently, in order to assure the evidentiary value of a document, the analyst should seek advice from a forensic document examiner and case attorney prior to ink sampling if handwriting comparison is required. Furthermore, to process a document chemically for latent prints, ink sample should be removed to avoid contamination with developing reagents of latent prints such as ninhydrin and ink dissolution on the page by reagent solvents [1]. Therefore, techniques those give more information from the ink using either visual or nondestructive ways are applied at first and then those involving samples removal from the documents or destructive/chemical methods [5] as described below.

3. Preliminary Or Visual Examination / Non-destructive Methods

Preliminary examinations of ink evidence are those methods that can be performed quite quickly prior to chemical/instrumental analysis. These methods are nondestructive in nature as there is no removal of ink from evidence sample thus maintaining integrity of questioned document. Furthermore, preliminary methods do not require any costly instrumental procedures and provide result analysis without causing any serious issues [2] as follows;

3.1. Pen Line Microscopy

Human eye is reflected as a dominant scientific tool with proficiency to find out the facts from inspection of ink line on a paper. Addition of a microscope with low power magnification may give vital information from the appearance of a written line [5,13]. This information can include general chemical composition of an

ink, useful methods of ink identification and differentiation, and assessment of two samples either with same origin or same pen. Normally stereo microscopy up to 60X magnification with reflected illumination reveals the type of writing instrument from pen line features [1,5&10]. Following microscopic features of pen inks with related substances can be used to identify their sources;

- **Ballpoint Pen Ink:** ballpoint ink line provides a clear sign of its origin under a magnification of 20-50X. It has a unique pasty texture and distinct glossy appearance due to its partial absorption into the paper. There are many cases where apparent imperfection or dirty ball casing cause striations with removal of ink as ball rotates in casing [5,10].

  Striations: are fine un-inked bands within the line of ink that spread from inside to outside of the line curve in the path of pen movement [1] as shown in following figure;

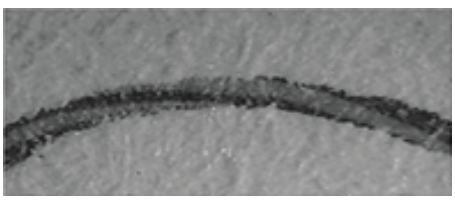

Figure 1: Ballpoint Striation 40x [1].

Writing Grooves: While writing with ballpoint, extra pressure is required during writing that can cause indentations or furrow as writing grooves in the paper as in following figure;

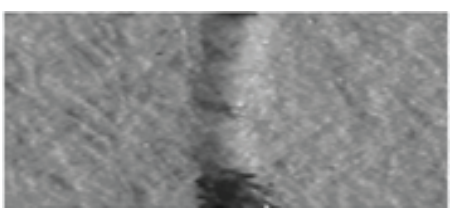

Figure 2: Writing groove 40x [1].

Gooping: Ballpoint has the tendency to deposit extra amount of ink in the line as the pen turns a corner, resulting in gooping as shown in following figure;

Figure 3: Ballpoint gooping 40x [1].

Fiber Diffusion: However, ballpoint ink does not show fiber diffusion or shading as compared to water based ink or wet inks that color the paper in a narrow line by absorbing into the paper fiber. In fiber diffusion, wet color absorbed in the paper does not show any layer but it evenly colors the surface area [5,10].

- **Roller Ball Pen Ink:** writing with roller ball pen ink dose not form striations or gooping but there may be sign of fiber diffusion. There is presence of more shallow writing groove as compared to that of ballpoint.
- **Fiber Tipped Pen Ink:** the stylus of fiber tipped pen as compared to ballpoint pen is made up of compressed fibers that are not hard enough in texture to form writing grooves. Fibers forming stylus bundle may cause striation within ink line due to splitting or forming fine lines with the main ink line because of usage fray as shown in figure 4. There may be presence of fiber diffusion as the ink used is water based [1,18].

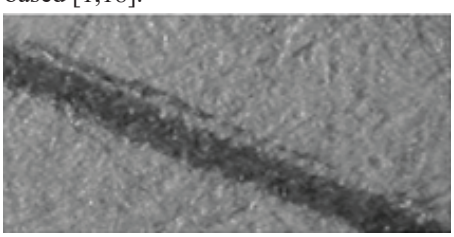

Figure 4: Fiber-tip pen striation/fiber bundle split 40x [1].

- **Felt Tipped Pen Ink:** similar to fiber tipped pen, the stylus is incapable of forming writing grooves as it is made up of soft or compressed material. It exhibits fiber diffusion as ink used is either water based or organic solvents as shown in following figure;

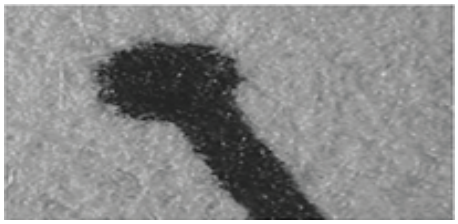

Figure 5: Fiber diffusion/felt-tip pen 40x [1].

- **Fountain Pen Ink:** this uses nib point thus producing dual nib marks or pathways based on the tip roundness, nib hardness or pressure exerted. It normally uses water based ink that results in fiber diffusion formation. Stroke shading with up and down pathways is a unique feature of the hard tipped fountain pens as shown in figure 6. There is considerable variation in the ink line near the pen pressure point where the nibs separate. With horizontal movement, pen line narrows followed by darkening of the line at the end because of the back flow of ink.

Figure 6: Fountain pen nib tracks/shading 40x[1].

- **Porous Tip Pen Ink:** it forms writing grooves due to hard feature of stylus with open tip and fiber diffusion using water based ink.
- **Gel Pen Ink:** gel pen similar to roller ball or ballpoint pen, uses a ball and casing. There is no gooping or striation formation in the ink line but is stable and dark in color. Most of the writers lower the pressure point due to smooth writing of the pen that intern reduces groove production. However, applying heavy pressure without any pen defect, sometimes may leave light inked portion at the center of the ink line like that of fountain pen. Gel pen ink can be effortlessly notable from fountain pen ink by showing single ball pathway or track under microscopic examination as shown in following figure;

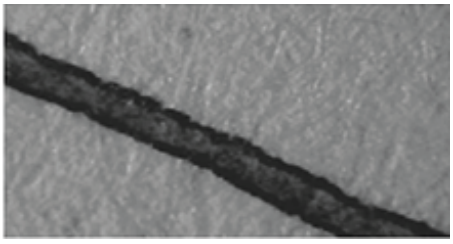

Figure 7: Gel-pen ink line (40x)

3.2. Microscopy of Non Pen Inks and Relevant Materials

Inks used to write other than pen are of graphic images or fonts that can be distinguished by microscopic features and usage on documents is a link to their origin such as followings;

- Typewriter Ink: ink used in cloth ribbon typewriter is deposited on the paper by impression device such as type font or pin of dot matrix printer. The ink can be distinguished microscopically as the carbon ribbon gives impressions with dull and less defined edges whereas the ink impressions from dot matrix are sharped. This can be useful for the examiner to

detect the ribbon that produced such impressions as shown in following figure;

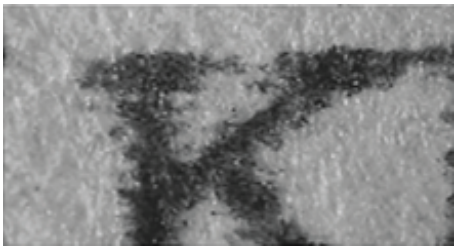

Figure 8. Cloth ribbon typewriter impression 40x [1].

- Rubber Stamp Ink: this type of ink usually red or black deposited by a rubber or polymer plastic printing surface on a document, can be recognized by faulty alignment of numbers, signature or notation. The sign of indentation is not likely and the surface area may be inked unconsciously.
- Offset Printing Ink: offset ink can be in the form of any color either high quality printing material or mass produced. The absence of toner spatter and sharp edges, distinguish it microscopically from a copier or printer origin device as shown in figure 9 where lines are produced by evenly spaced dots.

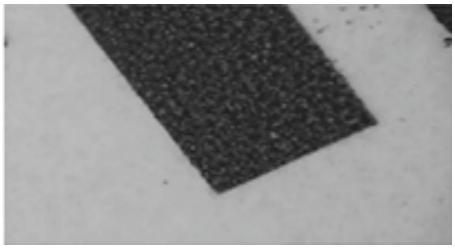

Figure 9. Offset printing 40x [1].

- Inkjet Printer Ink: this type of ink can be produced as a black or multicolored images onto a paper in the form of microscopically visible irregular shaped dots. There can be seen some color spatters around the image area and fiber diffusion on uncoated paper. Handwriting images similar to original handwritings performed with liquid ink pen, can be printed from graphic files by using high resolution printers based on fiber diffusion.

Figure 10. Inkjet printer ink 40x [1].

- Laser or Copier Printer Toner: resin coated and heat fused pigments are used to form an image on paper surface. Liquid toner colors the paper fiber following fiber diffusion characteristics whereas dry tone formulates unconnected granules. Color printers and copiers may show microscopic striation on large solid colored areas.
- Dye Pack: it is a dye or stain that in combination with exploding device is used for marking the stolen currency. Normally it is noticed on suspected currency as red ink spatter.
- Pencil: pencil and crayons are non-ink writing instruments that can leave distinctive microscopic traces in the form of solid deposits on the paper. Graphite and crayons are distinct from other materials because of their shiny appearance and waxy look, respectively. Impression from carbon paper may cause problems but it produces regular shading from the edges by lowering the pressure away from the center of the line, which distincts them from sharp edges directly made on the paper [1,5].

3.3. Color Assessment of Ink

The color difference is the main source between a questioned ink entry and a known writing on a paper and is one of the initials steps to be performed after determining the two ink samples were produced either with the same or different writing instrument. The three main elements of color assessment of ink are the ink sample, light source and observer [1]. The colors of dye and pigment that we view are either actual reflection of color materials or they are color additives which are produced by adding various reflected colors [1,5]. Such materials absorb light at specific wavelengths as they have combinations of atoms in their structure known as chromophors that exhibit the property of absorbing range of wavelengths. For example, some components of white light are absorbed such as green, blue and yellow, while the red is not absorbed so it will reflect as red color. In the same way, any object will be appeared as black if all of its spectrum is absorbed [1].

The combination of reflected wavelengths can be mixed by the eye that leads to identical appearance of the reflected light source with two different absorption pattern. For example, green color can be produced by reflection of two different dye source as green or mixture of yellow and blue dyes. Consequently, two inks with similar look but with different dyes and absorption pattern is not an indication of same origin or source. So methods are developed to distinguish these differences. The simplest method involves the use of any color light other than white on both inks. The differences will be detected if those portions of colored light reflected with different appearance to the eye [10,12].

Another method uses dichroic filter to detect differences in the spectrum by combining two colored glass or gelatin filters bonded together such as red and green filters. Light must pass through both filters and is partly absorbed based of absorption spectrum of two combined filters. At certain wavelengths, light transmitted will be consisting of small windows. A particular color will be visible if it is reflected with same wavelength as that of window. An absorption difference will be detected at this wavelength of two inks with similar appearance. Special equipment is used to determine absorption curve. Micro spectrophotometer is used to overcome the problem of small availability of material in ink lines. This is not used as common method due to expense and complexity [10,15].

3.4. Ultraviolet and Infrared Radiation

Ultraviolet and infrared radiations are invisible but these are absorbed similar to visible light. Absorption and reflectance of most black and blue inks can be examined as they absorb ultraviolet. On the other hand, most of the inks with similar color, absorb visible light and some differ with infrared absorption [1,5] as shown in figure 11. Infrared absorption depends on the chromophors and absorption range may vary from visible light to near infrared range. This variation will not affect the color of the ink as infrared invisible. Hence, an ink may continue to absorb radiation from the red to the infrared or will pass through it or will reflect as invisible or transparent light. Infrared radiation ranges wavelengths from 700nm to 1000nm where it is used to detect document examination and extends beyond this range where it becomes source of identifying chemical compounds [5,8].

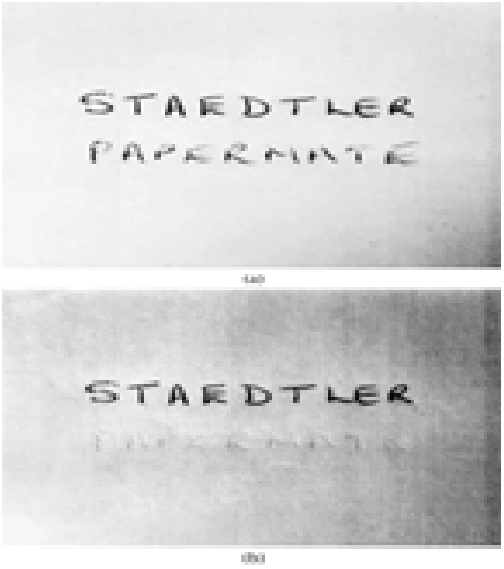

Figure 11: Two black inks of different manufacture photographed in (a) normal light and (b) infrared radiation, showing the difference of absorption in the infrared region [5].

Infrared is detected by using photography, emulsions or electronic means like photoelectric cells on display unit. Photography method uses infrared sensitive film and appropriate filters that are placed over the lens to allow only suitable wavelengths of radiation to pass through it. This method is used to obtain good quality of photographs which later on improved by introducing image converters and infrared sensitive tubes with suitable filters and light sources [1,8]. These mechanisms were followed by Foster and Freeman Ltd to make a purpose built apparatus Video Spectral Comparator (VSC) combined with filters, light sources, visual display unit, sensitive tubes and lenses. VSC enables to carry out a wide range of examinations under an ideal condition. VSC was further modified and other manufacturers developed similar apparatuses such as Projectina AG of CH-9435 Heerburgg made the Docucenter. It was combined with enhanced image techniques, improved light sources and video printers that facilitates instant recording of the screen of the monitor [8,14].

VSC and similar devices detect the differences between the inks with components absorbable in infrared range at a certain wavelength where one ink disappears and the other remains visible. Care must be taken as a thicker or more intense line of the same ink under similar conditions may show more prominently than the weaker one [14]. In addition to ink comparisons, infrared also detects graphite that constitutes to a large portion of pencil lead, and is absorbable in all ranges of infrared and visible spectrum. For instance, graphite is easily detected in a simulated signature where an ink if written over the signature made by the pencil line, is transparent in all spectrum of infrared radiation. [1,5].

3.4.1.Ultraviolet Fluorescence

Questioned document materials such as paper, glues, sealing waxes and adhesive tapes produce specific ultraviolet fluorescence that is excited in invisible region by ultraviolet radiation and is used as a source of identifying similarities or differences [15]. Chemicals or solvents applied to papers when dried apparently leave no trace but cause changes to the fluorescence that can be observed under ultraviolet radiation. Thus, erased writing can be revealed under ultraviolet examination. In the same way, inks can be distinguished from one another if having similar appearance such as red inks under ultraviolet radiation fluoresce. Inks differences have been reported through the production of fluorescence from lower wavelength to higher wavelength of ultraviolet region [5]. Ultraviolet fluorescence was more useful in past as evidence from older ink formulations were more detectable by this

method. But today's dye based inks are more detectable by infrared luminescence. However, visibility of some modern inks are projected only to ultraviolet radiation. Some inks are invisible when used to write on various documents such as signatures for security reasons but these inks are rarely involved in laboratory examination [8.11].

3.4.2.Infrared Luminescence

Infrared luminescence refers to emissions by papers, inks and erased ink leftovers in the visible infrared region in longer wavelength as shown in figure 13. Like ultraviolet radiation, infrared luminescence also uses photographic or electronic devices for direct vision [1,5]. To have a detectable result, high intensity of luminescence is excited by providing suitable sources such as quartz iodine or tungsten filament or xenon arc lamps combined with copper sulfate saturated solution or glass or gelatin filters. These allow passing of the green-blue exciting radiations as illumination but prevent falling of infrared from the source on the document. During this illumination, both the green-blue light and infrared luminescence emission will be reflected by the document so another filter is required to reduce exciting light coming from the detecting source of luminescence [7].

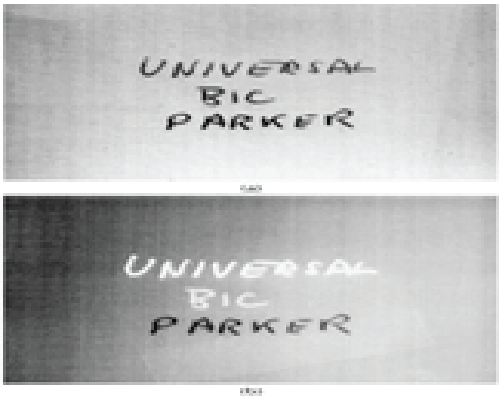

Figure 13: Three blue inks of different manufacture photographed in (a) normal light and (b) conditions suitable for the excitation and detection of infrared luminescence [5].

Photography or video camera like VSC with range of filters are used to detect the luminescence from different inks with recorded results on a video printer. Laser light is used as an extension of green-blue light to excite the luminescence radiation. As it is intense and monochromatic so does not require to filter out unwanted radiation. Therefore, visible luminescence closer to exciting light wavelength can be detected by cutting out the laser wavelength through filters. [9,16].

Other effects of luminescence under laser radiation include the observation of ink components printed from one page onto the adjacent page that has been in contact. The printed part of the ink is not obvious but its effect can be detected under laser light. Similarly, transparent plastics especially designed for wallets have the tendency to absorb and show impressions from ink traces or paints of details on the credit cards that have been in contact. Furthermore, fluorescent traces of vehicle index number written on a hand and then washed off, have been detected hours later under laser light. It is also possible to detect finger prints if they are contaminated with fluorescent materials [1,5&21].

3.4.3. Ink Erasures and Obliterations

Possible traces of invisible inks on or below the surface whose color components have been removed, when illuminated with visible light may fluoresce or luminesce. Detection methods are sensitive and can be affected by weak fluorescence or greater luminescence of paper than ink traces. Therefore, erased area must be observed in all possible wavelengths

as it is not always evident that what components of ink penetrate onto the surface more deeply when drying on the paper [5,9]. Infrared luminescence can be useful in detection of typewriter ribbon, stamp and pad ink erasures but not possible in some cases where all inks will not give a luminescence trace. [7,9].

In case of obliteration, original writing can be detected by examining the obliterated entries either of similar or different ink color under infrared radiation only when the original writing absorbs the infrared radiation. This technique will be of no use if both ink entries react to infrared radiation. However, this problem can be solved if the original ink absorbs infrared luminescence and will be visible under the non-luminescent obliterating ink. On contrary, if the obliterating ink fluoresce and the original ink absorbs infrared luminescence by reducing the luminescence of the covering ink so a dark area equivalent to obliterated area will be apparent [1]as shown in figure 14. It is required to control the lighting and filtering conditions.

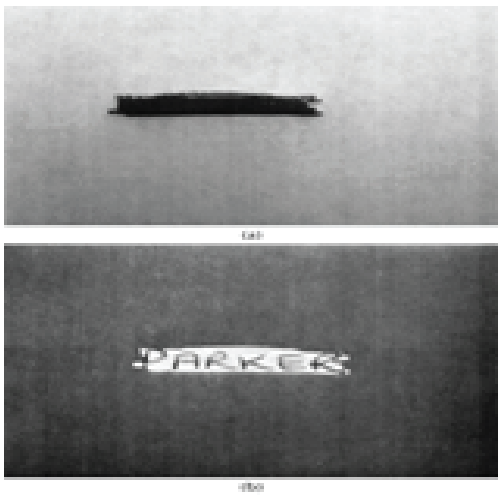

Figure 14: (a) An obliteration of one blue ink with another, photographed in normal light. (b) The same photographed in conditions suitable for the detection of infrared luminescence. Although the obliterating ink luminesces strongly, the non-luminescing ink absorbs the luminescence [5].

In addition to infrared absorption, microscopic examination is effective in heavily obliterated writings. Choice of suitable filters can reduce the invisibility if the color of obliterating ink is different. A photograph of greatest contrast can reduce the obliteration by providing high contrast for original ink with background rather than obliterating ink. An obliterated entry can be identified in cases where the original writing is left uncovered with sufficient evidence. Similarly, an enlarged photograph taken with favorable conditions can enable it to identify those obliterated lines that are faded out but left with uncovered portion of the original entry [1,5].

3.5. Digital Imaging Software

This technique was reported by William Bodziak in 2000, being used to differentiate writing ink. It was equipped with a desktop computer using digital imaging software Adobe PhotoShop 5.0 and a conventional flatbed scanner. A questioned writing ink and known black ballpoint inks were color scanned. Adjustment function was used for creating color differentiation of different black inks on the window screen of the software. From the settings of original image, positive or negative adjustments of Hue, Saturation and lightness were made. This procedure with suggested settings as Hue = 0, Saturation = +90 and lightness = -7 by Bodziak was tested and produced distinct results from the black ballpoint inks differentiation. However, this technique was not developed for ink differentiation but is effective [1].

4. Destructive/Chemical Methods

Additional information can be obtained from chemical or destructive methods but involve removal of ink or other writing materials from the paper. Therefore, before applying any chemical test to the document, a permanent record of the test entries should be made in the form of high quality photograph or with substitute of photocopy. Chemical methods such as chromatography can best discriminate the variation in ink components dried on the paper by. This technique is based on principle of separating a mixture's components and identifying or comparing them with others. It follows a stationary phase that absorbs small amount of entry material placed onto it and a mobile phase that passes through the stationary phase. The speed of material traveling through stationary phase depends on its chemical composition. Therefore, the method is ideal for mixture separation and identification based on different travel rates. The medium of stationary phase can be a solid, liquid and gas or liquid of mobile phase. Chromatography involves two methods for the examination of ink components [1,5] as following;

4.1. Thin-layer Chromatography (TLC)

Thin-layer chromatography is generally used for the comparison and identification of inks. It is distinguished from other chromatographic techniques on the bases of its stationary phase which is supported on glass, plastic or aluminum surface such as Merck TLC plates with coated silica gel but without fluorescent indicator. There are different varieties of thin layer chromatography including adsorption, continuous, stepwise, partition, two dimensional, radial, centrifugal and forced flow [7,20]. Of these, adsorption has been employed as principle method that separates ink components by adsorption interaction of stationary and mobile phase. The non-volatile components of ink have different migration rates that cause their separation into bands [1] as shown in following figure 15;

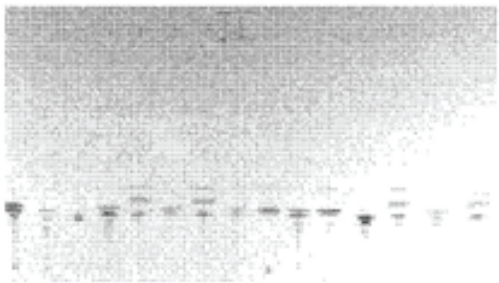

Figure 15: Blue ballpoint inks- thin layer chromatograms bands [1].

Based on the nature of the product, TLC enables immediate comparison of writing inks on the plate by separating both visible dyes and invisible organic components, whereas other organic compounds require post visualization treatments when analyzed by TLC [24,25]. Routinely four parameters visual, ultraviolet radiation, infrared luminescence and infrared reflectance or absorption are exhibited by some invisible ink components in TLC that may add further features to the identification. Such components with dim IR luminescence TLC bands can be detected by the use of video spectral analysis. UV absorbing components are revealed by short UV wavelength, while visible UV fluorescence is excited by long UV wavelength preferably using high intensity commercial long UV wavelength lamps other than standard long UV wavelength lamps. Additional description of the components can be yielded by spectrophotometric analysis of dye spots, while quantitative analysis can be obtained by carrying out densitometry [1,5].

4.2. TLC Densitometry

After identification of inks by TLC, the

relative concentrations of dyes present in these inks can be measured by scanning the TLC plate in the scanning densitometer. Blue and black inks dyes are scanned by spectrometer type densitometer at 585 nm. Whereas, all spots on TLC plates can be seen in shades of black by video densitometer that does not require wavelength settings. However, wavelength with maximum absorption by the densitometer is determined and used for scanning of other colored inks [28,30]. Relative concentrations of the dyes are compared and if found with no significant differences, then the results from preliminary and chemical tests justify a conclusion based on scientific certainty that all inks have same formulation [1].

4.3. High-Performance Liquid Chromatography (HPLC)

Similar to TLC where ink solution on a plate is evaporated as spots, HPLC uses high pressure to force the ink solution in a glass tube through a column with absorbent material that separates the components based on their different passage rates and physical properties [5,7]. The pressure is run until all the dyes have passed through the column and are detected by their merging to a device that measures either their color or absorption at specific wavelengths [5]. Quantitative analysis of each component can be obtained by proportionality of absorption to quantity of material. A graph is obtained representing the results with peaks showing presence of each component and proportions. HPLC determines more accurate proportion of the major components of the dye solution than that of TLC. However, the apparatus required for HPLC is costlier than TLC [29].

HPLC equipped with multi-wavelength detector has been used by Tabbett et al. that distinguished 108 blue non-ballpoint pen inks out of 113. TLC as compared to HPLC, separated only 17 groups from this set of ink. However, TLC when combined with densitometry enabled the authors to discriminate all these non-ballpoint inks. On the other hand, UV has been effective in distinguishing these inks as compared to visible spectrum because ink vehicles showed UV absorbance properties. Two such non-ballpoint blue inks were distinguished by using HPLC with multi-wavelength detection as shown in following HPLC chromatograms [1];

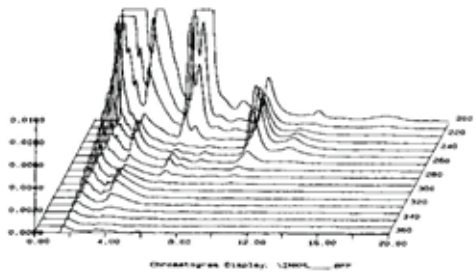

Figure 16.1: HPLC chromatogram of non-ballpoint ink 1 [1].

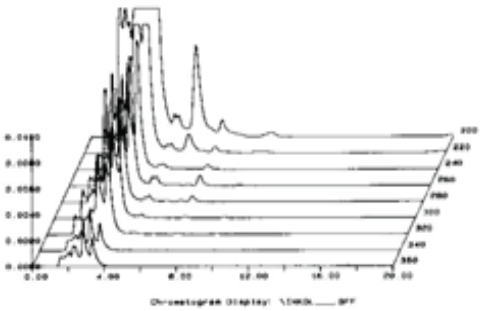

Figure 16.2: HPLC chromatogram of non-ballpoint ink 2 [1].

4.4. Other Chemical Tests

Other chemical methods for ink examination may be of followings:

- Gas Chromatography (GC) and Gas Chromatography-Mass Spectrometry (GC-MS) are handy particularly to measure the volatile components of ink. However, these methods are narrowed to inks that are not older than one year because the volatile components of written ink lines are detectable only within a duration of 6 to 12 months [5,19,21&22].
- Capillary Electrophoresis (CE) or Micellar electrokinetic capillary chromatography is a recently developed method used for comparison and separation of dyes present in all kinds of writing and ink jet inks with exceptional results [17]. It has higher resolving power with prominently small sample sizes and better results than that of TLC and HPLC [1,5&7].
- In some cases, two inks can be distinguished by their solubility. The dyes of a written line with application of a solvent drop, may drain into the nearby area on the paper. In general, most of the ballpoint inks have similar formulation that react in similar way to any solvent application. However, other inks can be easily distinguished by the application of a simple water solubility test that is time saving as well as does not cause further change to the document.
- It is rare to find some invisible iron based inks that convert their colorless compounds into colored compounds by further chemical application. It can be possible in two ways: first is the exposure of document to thiocyanic acid fumes set by potassium thiocyanic and dilute sulfuric acid mixture that convert the colorless iron salt to red brown ferric thiocyanate; second is the application of potassium ferrocyanide to the paper surface containing iron ink traces that on combining turns dark blue by forming ferrocyanide [5].

5. Instrumental Analysis of Inks

Chemical methods discussed above generally provide adequate writing ink comparison but sometimes we may come across the cases where further instrumental analysis of inks with diverse discrimination is required as followings;

5.1. Spectroscopy

Spectroscopy determines electromagnetic radiation and measures its wavelength interaction with substances resulting into absorption, emission or transmittance charts. It may follow non-destructive or semi-destructive procedures with small fiber removal from ink coated paper.

5.1.1.Color Space & Visible Spectrophotometry

Visible light micro-spectrophotometry is a fast and precise source of color sampling, storing and comparing. This method when combined with computer software, transforms reflectance data into color space coordinates by producing reflectance spectrums. Ballpoint inks have been analyzed through this technique by L. Keith Kerr using chromatic data examination with increase of 43% IR video spectral discrimination and 8.0% visible range. During this, colors are placed at a chromaticity point where they coordinate with color space diagram and consider as matches if they fall within in this point. Chromaticity point was created from data generated through recording of reflectance spectra from 10 different points within the ink stroke. This technique signifies as one of the non-destructive source of ink

examination [1,16].

5.1.2. Ultraviolet Visible Near Infrared (UV-VIS-NIR) Micro-spectrophotometry & Micro-spectrofluorimetry

Ballpoint pen inks or dyes and toner particles of color copier can be compared by performing UV-VIS-NIR micro-spectrophotometry and spectrofluorimetry directly on the document as non-destructive means. However, destruction can be reduced in transmitted mode by using single inked paper fiber. According to Seipp, ink can be discriminated in both transmitted and reflected ways but encountering some problems during reflectance [1,8] as follow;

- Multiple sampling is required for differentiating microscopic surface characteristics.
- UV recording was limited to not less than 380nm while using glass objectives.
- When using fiber optics that transmit UV, support sample radioactivity event and prohibited UV shortwave usage.

Micro-spectrophotometry when compared to TLC or HPLC cannot discriminate ballpoint inks efficiently as it gives spectra based on whole ink analysis, whereas chromatographic methods are based on separation of ink components. Micro-spectrophotometry limitations in the UV region is remarkable by differentiating various ballpoint inks being indistinguishable in other spectral region such as spectra of blue ballpoint inks [1,11&16] shown in following figure;

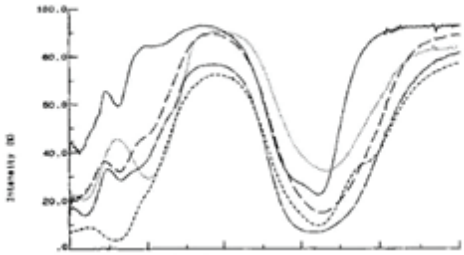

Figure 17: UV/VIS Spectrum of Blue ballpoint inks [1].

5.2. Fourier Transform Infrared Spectroscopy (FTIR)

Where color sample is measured by visible spectroscopy, its molecular features are revealed by infrared spectroscopy. Based on substance capability to absorb infrared radiation, FTIR provides absorbance fingerprint through IR beam and substance molecular bond interaction with equal vibrational frequencies. Absorption occurring at certain wavelengths, matches to specific functional groups and is measured through FTIR high resolution method that replaced older IR techniques. FTIR creates two source beams by using a beam splitter and a moving mirror to change the path difference between the beams producing intrusion pattern. Computer software then converts the time domain to frequency domain by using mathematical process of Fourier transform. FTIR as compared to dispersive instruments, gives greater throughput with higher resolution and involves numerous sampling techniques combined with different instruments for ink analysis [1,33&34] as following;

5.2.1 .Attenuated Total Reflectance (ATR)

ATR uses optically compact crystal that is pressed against the sample and absorbs IR beam by rotating it into momentary waves. Energy from these waves, is absorbed by the sample in its specific functional group regions and waves exit the crystal from the opposite end. The IR absorbance spectrum is generated from attenuated/reduced interference beam being analyzed by instrument's detector. ATR sampling method has been useful specifically in ink analysis with minute or no destruction to

the document because of shallow penetration of beam into the paper thus reducing paper interference. Penetration depth varies with respect to different types of crystal material and reduces with increase in reflections. Nicolet Continuum Microscope ZnSe ATR has been used for the analysis of indistinguishable ballpoint pen inks such as blue ballpoint inks [1,33] shown in following figures;

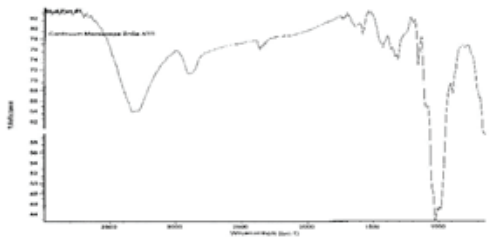

Figure 18.1: FTIR spectrum of blue ballpoint ink 1 [1].

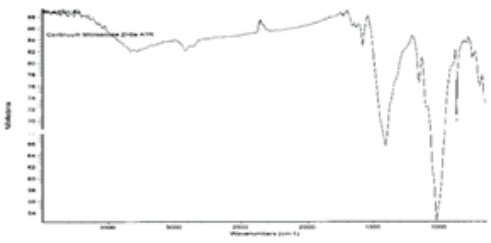

Figure 18.2: FTIR spectrum of blue ballpoint ink 2 [1].

5.2.2. Diffuse Reflectance Infrared Spectroscopy (DRIFTS)

This sampling method measures diffused IR reflected light absorption by preparing ink samples in a non-absorbing medium that increases penetration depth into the sample by reducing spectral reflectance. Thus, increasing internal reflection of components with increased transmission rate. Nicolet 20SXC FTIR spectrometer with DRIFTS has been used by Rena Merrill and Edward Bartick to differentiate visibly indistinguishable ballpoint pen inks and matched their spectra with library standards. Similarly, William & Mazzella in 1990 used DRIFTS for differentiating photocopier toners removed from the documents. They examined 'fingerprint' in 2100-700cm^{-1} IR range and based on IR spectra, categorized 149 toners out of 152 into 36 sets. Both ninhydrin used for latent print and toner age, did not affect the IR spectra. [1,34].

5.3. Raman Spectroscopy

Raman is the effect of wavelength change in scattered light as resulted radiation of certain substances and named for Sir Chandrasekhara Venkata Raman of India after 1930 Nobel Laureate. An intense source of radiation is required for one photon excitation to cause this change; thus laser is used as monochromatic radiation source to measure this change at angle of 90º. It results into Elastic or Rayleigh scattering, if the scattered light's wavelength does not go through radiation changes. However, if the scattered radiation wavelength changes, it results into inelastic scattering as stokes with longer wavelength or anti-stokes with shorter wavelength than that of source radiations. Raman spectroscopy commonly uses stokes changes as they are more intense with magnitude of 4000 cm^{-1}.

Both Raman and FTIR are non-destructive means of analysis but FTIR is provided with better sensitivity without any influence of specimen's fluorescence. Whereas, Raman with weak signals is concealed by sample fluorescence. However, Raman spectroscopy leads with clear peaks and gives enhanced sampling resolution of 1 micron as compared to IR absorption with 10 microns if incorporated with microscope. Raman spectroscopy usually does not follow sample preparation and gives results without paper interference. Raman spectra libraries of

desired substances including inks have been compiled by some laboratories. For example, 12 black ballpoint inks analysis was reported by Lyter in 2000 using a Foster & Freeman Foram 685 Raman spectrometer with laser power of 1.0 and compared the results with TLC for effective casework. These 12 black ballpoint ink samples based on Raman spectra were divided into six different groups including featureless group with two ink samples, and fluorescence exhibited group with two more ink samples as shown in figure 19.1-19.6. Raman spectroscopy being non-destructive, has the capability of differentiating two similar ink formulations but not the same ink formulations. Whereas, TLC being semi-destructive differentiated all 12 ink formulations [1,36].

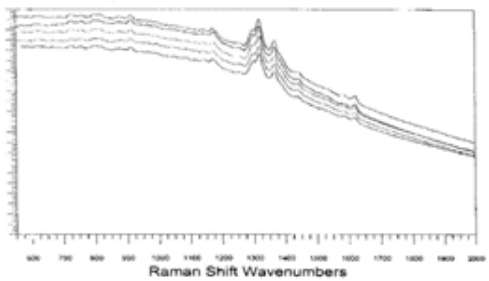

Figure 19.1: Black ballpoint 1-unique spectrum [1].

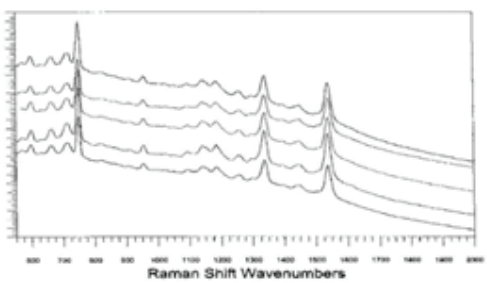

Figure 19.2: Black ballpoint 2-unique spectrum [1].

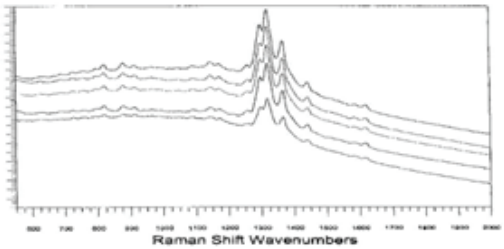

Figure 19.3: Black ballpoint 3-unique spectrum [1].

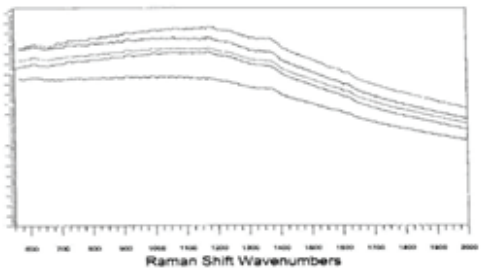

Figure 19.4: Black ballpoint 3-small features [1].

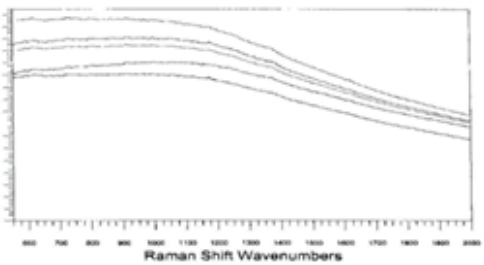

Figure 19.5: Black ballpoint 4-featureless [1].

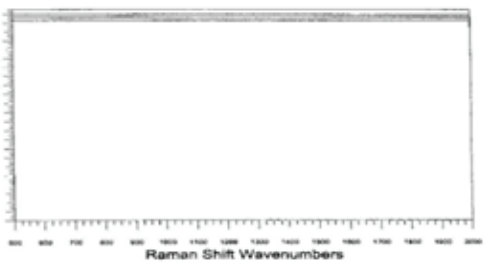

Figure 19.6: Black ballpoint 4-fluorescence [1].

6. Ink Dating

Aging of ink was determined in 1920s when chloride or sulfate ions of iron gallotannate inks along with paper fibers were verified with aging features. Early researches from past to present about dating ink have been conducted in the forensic ink examination.

6.1. The Aging Process

Inks comprise of numerous substances such as dyes or pigments as colorants, carrier solvent as ink vehicle, and fatty acids and resins work as drying and viscosity agents of the ink. All of these substances may exhibit aging properties with the passage of time causing considerable changes in the ink and can be as simple like evaporation of volatile substances and complex like oxidation of substance or resins hardening on the paper surface. Ink extraction to a weak solvent and residues of this solvent in the ink are two common aging properties. Resin hardening and solvent evaporation contributes to fresh ink aging for a period of one year. The main factor of ink aging over a year old, is the hardening of resins when examined during ink dating. Most of the research methods involve ink extraction to weak solvents [1,16].

Dating of questioned writing inks has a great demand in the criminal and civil litigations because of the growing capabilities and services of this knowledge throughout the world. Advancements have been made since the development of the first dating ink technique in 1968 and have been passed and accepted as routine in U.S. courts. Israeli, Australian, Hong Kong and Japanese courts also have accepted the testimony of such advanced methods. Therefore, it is reliable to say that these methods have been testified 1000 times by the government and private dating ink chemists [31].

6.2. Method of First Production Date

After the identification of ink, the first date of ink production with specific formulation can be verified from the manufacturer of that specific ink formulation. If the production and marketing date taken from manufacturer is not till the document preparation date, then document was not written on a claimed date. The questioned entry must have been written after first production date of ink and dated back.

6.3. Ink Tag Method

The first ink tag was added to ballpoint inks in 1970 by the Formulabs manufacturer and its use was dropped by June, 1994. Ink tags are chemicals that exhibit fluorescence properties and can be identified when reacted to ultraviolet light. Therefore, ink tag identification leads to determining actual year of ink production. TLC is used to detect and identify the ink dating tags in the same way as other questioned inks. The tags if present can be viewed using ultraviolet light and their relative concentrations can be compared with that of standard tags. However, no further information can be provided about tags as these are considered as Formulabs branded information [5,32].

6.4. Ballpoint Inks Relative Aging

Inks can be distinguished by detecting their colorants and solubility in specific solvents which is proportional to the time duration being on the paper. Solubility test is possible by assessing the quantity of extracted color in a time period during dissolving the dried ballpoint inks. Dissolution rate is determined

by taking solution samples with intervals of one or two minutes after introducing the dried ink into the solvents. If the rates of two samples of the same ink on the same document are different, say one is faster than the other, so this sample dried earlier and has been for a shorter time on the document. However, this phenomenon is not for different ink samples or a different surface so will mislead the results. Therefore, this test is only useful for writing lines of two similar inks on the same document or inks aged from few weeks to about nine months [5].

6.5. Dating of Inks

The exact time of writing ink cannot be determined by relative aging of ink nor other ink analysis but if the ink and any writing made in a certain date. This principle is applicable when the disputed dates have extended the new type of ink such as ink from ballpoint pens. Keeping in view various formulations of different inks and manufacturers, complete records and support of ink producers are required for obtaining well know information. Such collection of ink formulations is built up in the United States by the Laboratory of Alcohol, Tobacco and Firearms. The same laboratory in collaboration with ink producers in the United States, has arranged the tag method for the indication of manufacture year. These tags/chemicals even in small proportion can be detected by particularly designed analytical techniques. The presence of such chemicals also can distinguish the two inks from each other that require such comparison [1,5].

7. Guide to the Best Techniques for Ink Comparison

Any technique described above can potentially identified two inks with different chemical formulations as long as the proper procedure and technique's limitations are followed. Some methods can have greater percentage of ink differentiation than others but all non-destructive methods available should be tried first by flowing thumb rule. However, at current the high performance thin-layer chromatography (HPTLC) is believed one of the best semi-destructive techniques with combination of simplicity, economy and performance. The two techniques, HPLC and capillary electrophoresis are considered more selective than HPTLC but both have higher initial equipment investment and do not perform side by side sample analysis.

Different techniques are combined to provide evidentiary results for identical ink formulations by measuring different chemical factors. It is of evidentiary significance if rarer pen matches in formulations. However, only matching two ink formulations chemically does not prove that the two written entries were made with the same pen. But ink formulation can be associated with the pen if it is unique such as the DNA Pen. This is the product from DNA Technologies, Inc. which is not typically encountered in casework. The written lines produced by this pen can be matched with it by forensic DNA testing as the ink used in the pen is combined with owner's DNA [1].

8. Ink Libraries

Ink standards required for identifying ink formulation can be obtained from ink manufacturers by accessing inclusive collection with information such as ink manufacturer, ink formulation number and first manufacture date of particular ink formulation. A complete ink library should have numerous thousand ink standards by being updated annually. Up to date standards should be obtained from different manufacturers from

different regions of the country preferably different countries and ask for a difference if encountered with similar ink standards. Manufacturer companies are supplemented to obtain the standards of pens being purchased in retail stores. It is important to inquire the pen companies about the ink information that they use in their pens as most of the pen companies do not manufacture their own ink product rather they purchase from other ink suppliers e.g. Dokumental or National Ink.

The ink standards in an ink library are deposited on plain white photocopy paper as a whole written page if the ink standard is in the pen and as a smear onto the paper by using cotton swab if the standard is in the form of bottle or tube liquid ink. The deposited written ink standards are completely dried prior to storing in a file of ink library and are provided with limited exposure to light so that to reduce ink's fading. Ink standards storage is based on ink types and colors that will simplify the recovery of an accurate ink standard during comparison of questioned ink. In ink library, every ink standard should be evaluated by TLC with different concentrations from weak to high on the same TLC plate that is stored suitable size of envelops for future examination [1].

9. Conclusion

Each suggested techniques may provide valuable information if applied appropriately and incorporated with other reliable methods. Preliminary visual examination combined chemical methods is commonly recommended by many forensic experts for ink analysis. The ink chemists should be aware of certain factors that may influence the written entries such as aging processes and technique's limitations. Thus, the volatile components that evaporate as aging entries should not be the only piece of information when performing comparison or identification process. Similarly, caution is required when performing dye based examination as there may be possibility of dye degradation in both qualitative and quantitative explanation of the resulted data. Furthermore, ignoring aging mechanisms may lead to unexpected false negative outcomes during comparison of writing materials with varying chemical profiles. Therefore, research should be focused on validating current techniques and introducing standard procedures assisting reliable interpretation of results rather than developing new analytical methods.

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