# INVESTIGATION OF GENUINE AND FAKE MALAYA HISTORICAL BANKNOTES USING VIDEO SPECTRAL COMPARATOR AND CHEMOMETRIC TECHNIQUES

by

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Dissertation submitted in partial fulfilment of the
requirements for the degree
Of Bachelor of Science (Honours) Forensic Science

June 2016

# ACKNOWLEDGEMENT

My foremost gratitude goes to my supervisor Dr. Wan Nur Syuhaila Mat Desa who constantly and patiently gave ideas, guide, taught, support and enlighten me on not only on my research but as a forensic officer. Secondly is to my co-supervisor Dr. Dzulkiflee Ismail, who never gave up in trying and guide me in performing statistical analysis. I personally would like to thank them for all the effort that they had put day and night in helping me to complete this dissertation. Thank you for being great advisors for me.

My deepest thanks go to the Questioned Department Section of Maktab Polis Diraja Malaysia Kuala Lumpur (MPDRMKL), Cheras headed by Supt. Nor Azman B. Mohd Noor who allowed me to use Video Spectral Comparator (VSC). My gratitude is also dedicated to the friendly and kind MPDRMKL officers for their cooperation and hospitality, Insp. Nurul Hayati Bt. Mat Husin and En. Mohd Tarmizee B. Mohammad who taught me on procedures of using VSC and patiently answer all my queries on banknote authentication; as well as to En. Rizal B. Harifin who had helped in retrieving valuable data from VSC. I would also like to say my thanks to En. Baharuddin, staff of Forensic Laboratory of Universiti Sains Malaysia who helped me with procedure of using Leica Stereomicroscope.

I wish to extend this gratitude and appreciation to Dr. Nur Haslindawaty Abd Rashid who had work hard as course coordinator of GTF400 and to all lecturers, staff and friends that had earnestly taught and guided me throughout these four years in Forensic Programme. This acknowledgement is also especially dedicated to my parents, En. Rosli Abu Hasan and Dr. Roslina Abdul Latif and family for their endless support and encouragement for all these years. Thank you very much.

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# LIST OF ABBREVIATION

| | |
|---|---|
| CIE | *Commission Internationale de l'Eclairage* |
| MPDRMKL | Maktab Polis Diraja Malaysia Kuala Lumpur |
| NFSTC | National Forensic Science Technology Center® |
| nm | Nanometer |
| OVD | Partial Least Square Discriminant Analysis |
| PCA | Principal Component Analysis |
| PPMC | Pearson Product Moment Correlation |
| RSD | Relative standard deviation |
| SPSS | Statistical Package for the Social Sciences |
| SUNY | State University of New York |
| VSC | Video Spectral Comparator |

# ABSTRAK

Penyiasatan bagi menentukan ketulenan duit merupakan suatu subjek menarik dalam pemeriksaan dokumen forensik. Kehadiran matawang palsu dalam pasaran memberi kesan negatif pada ekonomi sesebuah negara. Matawang lama merupakan artifak yang dikategorikan sebagai barang terkumpul dan mudah dipalsukan dengan perkembangan teknologi percetakan pada hari ini. Penyelidikan ini membantu mengurang dan mencegah jenayah pemalsuan matawang melalui kajian perbezaan matawang lama yang asli dan palsu menggunakan *VSC* dan teknik-teknik *chemometric*. Dalam kajian ini, matawang Malaya iaitu Duit Pisang dari zaman Jepun digunakan. Sampel duit simulasi yang menyerupai Duit Pisang telah dihasilkan menggunakan empat teknik percetakan yang berbeza iaitu *HP Laser*, *Compaq* jet dakwat, fotokopi Ricoh dan percetakan CDH. Kedua-dua sampel dibandingkan menggunakan *Leica Stereomicroscope* dan *VSC* melalui pelbagai sumber cahaya. Pemerhatian menggunakan stereomikroskop menunjukkan kualiti percetakan yang baik bagi sampel asli berbanding sampel palsu. Setiap sampel menunjukkan reaksi yang berbeza apabila terdedah pada sumber cahaya yang berbeza. Data kromatisiti (ukuran warna) yang dihasilkan oleh *VSC* dianalisa menggunakan *PPMC* dan *PCA*. Hasil analisa warna dakwat menggunakan *PPMC*, menunjukkan hubung kait yang tinggi antara sampel simulasi dari percetakan CDH, *HP Laser* dan fotokopi Ricoh dengan sampel asli. Hasil *PCA* menunjukkan sampel yang dicetak oleh CDH, berkelompok dekat dengan sampel asli. *PCA* juga menunjukkan kelompok sampel yang baik dengan kebanyakan sampel berkumpul berdasarkan teknik percetakan mereka. Kesimpulannya, matawang asli dan palsu boleh dibezakan menggunakan *VSC* dan teknik-teknik *chemometric*, justeru menyumbang kepada pengenalpastian jenayah pemalsuan terutama dalam pengendalian artifak matawang lama.

# ABSTRACT

Investigation on banknote authentication is an interesting subject in forensic document examination. The existence of counterfeit banknote in line with their circulation in the market could badly impact the economy of a country. Historical banknotes which are artefacts and considered as collectible items are prone to counterfeiting with advanced printing technology today. Thus, this study aims to help in reducing and prevent fraud crime through discrimination of genuine and fake historical banknotes by VSC and chemometric techniques. In this study, old Malaya money which was Banana notes since the Japanese era were used. Simulated samples were produced using four different printing techniques namely HP Laser, Compaq inkjet, Ricoh photocopy and commercial CDH printing. Both genuine and simulated samples were compared using Leica stereomicroscope and VSC under different light sources. Observation under stereomicroscope showed a better printing quality produced on the genuine samples as compared to all simulated samples. Under different light sources by VSC, all samples showed different reaction towards different light sources. PPMC and PCA were conducted on chromaticity data (colour measurement) generated by VSC during spectral analyse. For PPMC, CDH printing, HP Laser and Ricoh photocopy produced ink colour of high correlation with the ink colour of genuine samples. PCA results revealed that banknotes printed at commercial printing house, CDH were clustered in a closer vicinity to genuine notes. Additionally, PCA also showed good clustering of samples with most of the banknotes are grouped according to their printing methods. In conclusion, genuine and fake historical banknotes could be discriminated using VSC and chemometric techniques, contributing positively to detect fraud crime, especially when dealing with this kind of artefact.

# CHAPTER ONE

# INTRODUCTION

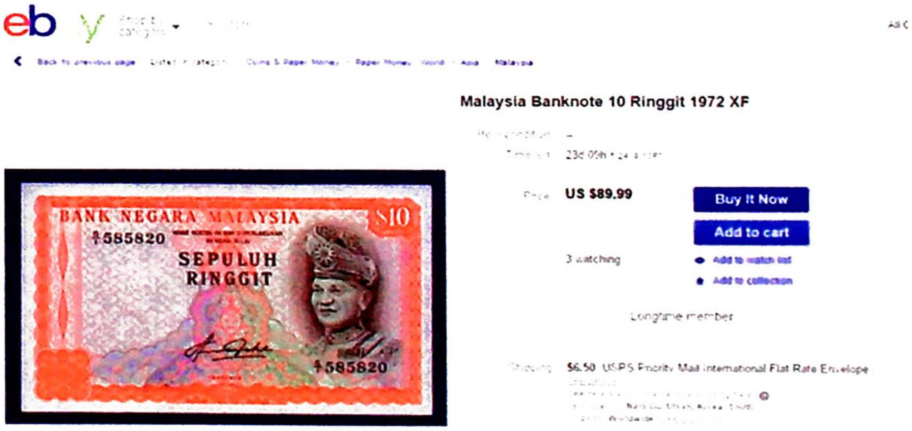
## 1.1 Background of study

Currency counterfeit started its journey ever since coins and paper moneys were introduced replacing the barter system. In European countries for example, there have been a rapid growth in fake euro banknotes since the launch of the currency in 2002 (Choi *et al.*, 2010). According to the European Central Bank (ECB), the fake euro notes seized by authorities in the second half of 2009 was 8% higher than in the first six months (Choi *et al.*, 2010). In China, the counterfeiting rate of high-denomination bills (100 yuan) has jumped to more than 22.3% in 2008 compared to previous year (Choi *et al.*, 2010).

In the past, banknote issuer have resorted to print warnings on their currency to deter potential counterfeits which appeared on Chinese currency notes of the early Ming dynasty (1368 - 1644) but this did not deter counterfeiters for their illegal activities (Hardwick *et al.*, 2001). With the advance of technology in this millennium, a reasonable high quality photographic copier can produce realistic counterfeit banknotes (ThermoScientific, 2013). With respect to currency forgery, according to Prime and Solomon (2010), 'The perfect forgery has never been detected. The forger does not require to reproduce actual note but only a simulation that can be passed at least once' (Mohamad *et al.*, 2014). Levels of counterfeiting, forgery and product diversion keep on increasing globally (Hardwick *et al.*, 2001).

Originally, currency was counterfeited with the intention to destabilise economy of an enemy. In one instances, during World War II in 1942, 'Operation Bernhard' was conducted where the Germans attempt to destabilise the British by having the inmates to counterfeit the 5, 10, 20 and 50 British pound banknotes (Heij, 2010). Nowadays, historical banknotes can be considered a cultural heritage with a huge historical and social value (Imperio *et al.*, 2015). Thus, being considered as an artefact, the price could cost up to millions and billions. With lack of security features in past banknotes, undoubtedly historical banknotes are easy to duplicate therefore hard to distinguish between genuine or fake copies of banknotes.

Similarly, in Malaysia, historical banknotes were sold at high price depending on the age, period and condition of the item. Popular site for online trading such as mudah.com or ebay Malaysia advertise historical banknotes with values up to hundreds or even thousands and sold in other currency as shown in Figure 1.1 until Figure 1.4. Regardless of the source where the notes are obtained, vital questions are still to be answered, is this genuine and is it worth the price?



Malaysia Banknote 10 Ringgit 1972 XF

23d 05h

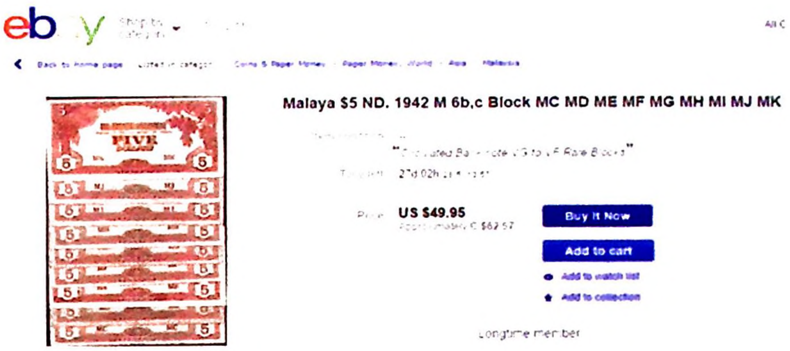
US $89.99

3 watching

Longtime member

$6.50 USPS Priority Mail International Flat Rate Envelope

Figure 1.1: A 1972 Malaysian banknote worth 10 Ringgit listed in ebay website adapted from Jongpaly (2016).



Malaya $5 ND. 1942 M 6b,c Block MC MD ME MF MG MH MI MJ MK

US $49.95

Figure 1.2: 9 banknotes of different block worth 5 Dollars listed in ebay website adapted from Darwishbanknotes (2016).

**Duit jepun**
RM 100
6 Apr 09 50
Cheras

**JIM Duit Lama Jepun Malaya 1000 Dollars**
RM 138
20 Apr 12 49
Batu

**Duit Lama Zaman Jepun Malaya 100 dollars 1942**
RM 180
18 Apr 13 49
Batu

**Duit kertas lama zaman jepun**
RM 500
9 Mar 23 25
Jalan Ampang

**Duit jepun lama**
RM 199 000
29 Apr 19 12
Cheras

Figure 1.3: Various old Malaya-Japanese banknotes sold in mudah.my at high prices adapted from mudah.my (2016).

$999.99

$188.00

$109.00

$172.00

$225.00

$400.00

Figure 1.4: Various other old Malaya banknotes sold in ebay at high prices adapted from ebay (2016).

## 1.2 Problem statement

In present time, modern currency is guarded with sophisticated safety features. Nonetheless, problems still persist although fake modern notes only comprises of a small portion of genuine notes in circulation (Choi *et al.*, 2010). Bigger problems arise when dealing with historical banknotes because they do not or have very few security features, thus the discrimination between fake and genuine historical notes become harder. Because of their historic values, these banknotes have become one of the collectible items that sometimes are valued to thousands of ringgit. Counterfeiters see this as opportunity to gain profit by selling fake currency, an act considered as act of fraud and unlawful. Nowadays, problem on counterfeiting modern banknotes to circulate in the system that affects economy, selling counterfeit historical banknotes is no different in affecting the economy of a person.

For counterfeiter, it was reported to be easier for them to create banknotes that are of good quality visually than to produce a counterfeit note with identical chemical composition of ink or paper to the original (Sonnex *et al.*, 2014). Thus, it is practically very difficult to distinguish genuine or counterfeit banknotes with naked eyes. Consequently, it is of great interest by the police or forensic document examiner to interrogate chemical composition in investigating authenticity of a document evidence (Sonnex *et al.*, 2014). The need for advanced chemical analysis to support forensic case work has become more important due to the complexity and sophistication of fake items (ThermoScientific, 2013).

## 1.3 Scope of study

The focus in this research is to distinguish genuine and fake historical banknotes based on ink colour of the banknotes. Dating of paper, although is another method useful for such study, is not considered due to resources limitation. Non-destructive approach is taken in this study considering the values of genuine notes as it is important to preserve the genuine banknotes. Additionally, non-destructive methods are preferred in forensic investigation so that the samples can be utilised as criminal evidence and further investigation (Kao *et al.*, 2013). Therefore, analytical methods, including microscope, Video Spectral Comparator (VSC) or infrared spectroscopy are preferred over wet chemical analysis. Applying these techniques on historical banknotes enable the original condition to be maintained and also preserve the notes. Comparison of ink by colour measurement will be conducted to determine authenticity status of the sample. Simple statistical work is applied on the data for objective comparison.

## 1.4 Objective of study

### 1.4.1 General objective

To discriminate genuine and fake historical banknotes by visual observation and colour quantitation.

### 1.4.2 Specific objectives

1) To compare the physical and visual characteristics of genuine and fake historical banknotes under different lighting sources using VSC 6000.
2) To study the feasibility of using colour measurement to distinguish printed inks on genuine and fake historical banknotes.

## 1.5 Research question

Based on this research topic, there are a few questions that are projected. How to discriminate between genuine and fake historical banknotes? What are the characteristics of genuine and fake historical banknotes under different light sources? Some inks could show different reaction to different types of light exposures. The third research question is how to differentiate ink colour? Different ink has different colour tone and shade. Hence, this may help to identify the characteristics based on colour quantitation along with other physical and visual examination.

## 1.6 Significance of study

This study is to help distinguishing genuine and fake historical banknotes particularly involving notes with minimal or no security features. It is hope that such work could contribute positively to fraud crime.

# CHAPTER TWO

# LITERATURE REVIEW

## 2.1 History of Malaya Banana banknotes

After the attack on Pearl Harbour on 7 December 1941, Japanese army invaded and occupied the whole South East Asia by May 1942. The Imperial Japan introduced their own currency to finance the Second World War occupation of South East Asia. Following the first financial principle of occupation, the occupied country should bear the cost where Japan did not have the financial options available in the far east countries, namely Burma, Thailand, Malaya (including Singapore), Thailand, Indochina and Philippines. With the downturn of export economies, Japanese tried to raise their resources by introducing new taxes but resulted in little profit. Thus, money creation happened as an effort for the Japanese to transfer resources back to themselves by printing the required amount of currency as military script in countries administered by military governments which were Malaya, Indonesia, Burma and Philippines (Huff and Majima, 2015).

Printed with 'appropriate' illustrations, such as banana plants for Malaya and Indonesia, pagodas for Burma and two lettered printing blocks, these scripts were only legally used in occupied territories only. Following that, Occupation currencies of Japan attracted derisive names, namely 'Banana money' in Malaya and Indonesia, as well as 'Mickey Mouse' in Philippines (Huff and Majima, 2015). It was stated that initial issue of this historical banknote was serially numbered with control letters and safety features such as watermark and security thread. However, during that moment of Japan rule, hyperinflation occurred and the banana notes were printed at large quantities without any security features and overtime, serial numbers also disappeared from the notes (Ho, 2016). With initial intention to fund civil administration and purchase supplies for the military, the Japanese began to print more, resulting with 30 times greater active circulation than before the war (Friedman, 2004). This currency deteriorated as war continued and had no longer values after the surrender of Imperial Japan in 1945 (Huff and Majima, 2015).

## 2.2 Ink and printing

History of ink is dated back in centuries from ancient Egypt and China. Black writing inks were first developed before 2500BC using natural and environmental sources for ink which composed of lampblack or coloured minerals dispersed in water-soluble natural gum (Kunjappu, 2014). From its uses to write and draw, ink was developed for printing purposes which was invented by the Chinese about 3000 years later using a mixture of coloured earth, soot, plant matter and gums. By 1440, Johannes Guttenberg invented the first printing press with movable type where the ink was made of soot mixed with linseed oil or varnish (Wansbrough, 1998).

There are two types of ink which are soluble ink and pigments. Soluble ink or dye is ink that is soluble into substrate once applied that is commonly used for writing while pigments are fine particles dispersed in the solvent used in printing (Kunjappu, 2014). For ink printing, the chemical compositions and physical properties differ depending on the requirement of different printing process (Nie *et al.*, 1998). Printing ink generally composed of pigments, resins, solvents and binders (Wansbrough, 1998).

- Pigments: Gives colour of the ink which can be organic or inorganic pigments. It can also affect physical properties like opacity, resistance, gloss and abrasiveness. Common examples of in pigments are inorganic white titanium oxide ($TiO_2$), organic red lithol (C.I. 26670[3]), organic orange pyrazolone (C.I. 21110) and inorganic black carbon black (Wansbrough, 1998).
- Resins: Primary binders that bind all ink composition together to form a film and bind ink to paper. It also contributes gloss and resistance of ink from heat, chemicals and water. Examples are epoxides, hydrocarbons, acrylics, ketones, maleics and cellulose derivatives (Wansbrough, 1998).
- Solvents: To keep ink in liquid form when applying on printing plate. Once the ink is printed, the solvent must separate from the body of ink to allow image to dry and bind to surface. Different type of solvents can have different evaporation rate depending on type of printing process for example isopropanol (82.5°C), ethyl acetate (77.0°C) and n-propyl acetate (101.6°C) (Wansbrough, 1998).
- Additives: Added to alter final properties of the ink. Common types are butyrolactone, butoxyethanol, cyclohexanone and aromatic distillates (Wansbrough, 1998).

According to Kunjappu (2014), the principle of printing can be illustrated by simple pad operation. The operation explains how liquid ink is used to wet the pad and pressed onto a substrate producing an impression. However, there are many printing processes available that differ from its ink composition, type of printing result and how the printer work, more specifically, how the ink distribution rollers are arranged in the printing press. Letterpress, flexography, lithography, screen printing, gravure printing and intaglio are major classes of printing processes (Kunjappu, 2014). In questioned document examination, each of these printing process are identified by their general characteristics of letters printed whether it is flat ink or raised ink, squash and halo or patterned printed. An article from Wansbrough (1998) describes all the printing process in Table 2.1.

Table 2.1: Descriptions of printing processes (Wansbrough, 1998).

| Printing process | Descriptions |
|---|---|
| Letterpress | A 'relief' printing process where it uses plate of raised image area. This image area will be inked prior to printing on a substrate. Generally, it results in flat ink, squash and halo, straight edges and indentation. |
| Flexography | A similar process to letterpress where it uses plate of raised image area. The difference is that letterpress uses a metal plate while flexography uses rubber plate. Generally, it results in flat ink, squash and halo, straight edges and no indentation. |
| Lithography | It uses the concept of oil and water which do not mix with each other. Area of image is inked with oil-based ink while non-image area is wetted with water. Generally, it results in flat ink, straight edges and no presence of squash and halo. |
| Screen printing | This produces image by allowing ink to pass through the image instead of transferring it. Generally, it results in flat ink and grid pattern. |
| Gravure printing | It converts image into thousands of tiny inkwells recessed into surface of the printing plate. Generally, it results in flat ink, honeycomb or zig zag edges. |
| Intaglio | Produces image by using etched or engraved steel. Generally, it results in raised ink and straight edges. |

## 2.3 Principle of questioned document examination

The discipline of forensic document examination is frequently associated with white-collar crimes, such as check fraud. However, in practice, this area of forensic science can be used in a wide array of cases from medical malpractice to art forgeries to homicides (NFSTC, 2013). Analysis covers on signature or handwriting correspondence, bank checks, security documents, ink examinations, currency or even machine-generated documents (NFSTC, 2013). Examination is performed to evaluate the evidence of obliterations, alterations or erasures that presented on the document. Similarly to any other forensic analysis, it aims to determine whether a questioned document come from the same source, by determining authenticity and authorship or detecting fraudulent of the document (Silva *et al.*, 2014). In ink examination, it involves the examination on differences in ink or substances on the basis of physical characteristics, chemical composition and optical properties (Lacalamita, 2013).

Currently available analysis are ranged from traditional methods that are time consuming and destructive to quick, easy and non-destructive method. A study has outlined the chronology of methods exist in identification and characterising of inks for over 40 years from 1950s to the 1990s (Chen *et al.*, 2002). Chemical analysis through chromatography so far was the best method in separation and subsequent comparison of ink components (Chen *et al.*, 2002). Being simple and quick, Thin Layer Chromatography (TLC) and High Performance Liquid Chromatography (HPLC) were also carried by a number of researchers (Chen *et al.*, 2002). Depending on different chromatographic conditions, partitioning between mobile phase and stationary phase was used to determine the retardation factor (Rf) of a compound (Sharma *et al.*, 2014).

## 2.4 Non-destructive analysis for questioned document

Before 1950s, inks on questioned documents were primarily observed and examined by photography, using filters to enhance different contrasts between different inks (Bauer, 1966). Later, incorporation of light for ink examination was established in 1978 using luminescence technique introduced by Hardcastle and Hall (1978). The theory of the technique is based on promotion of dye molecules to a higher energy state by absorption of a photon of energy (Chen *et al.*, 2002). Infrared luminescence was found to be effective non-destructive method introduced to differentiate ink (Sensi and

Cantu, 1982). However, it did not provide ink composition as performed by HPLC and Gas Chromatography (GC) (Chen *et al.*, 2002).

Non-destructive is preferable because it can perform analysis straight on the questioned document without any sample preparation needed allow a sample to remain intact. This allows preservation of its integrity throughout the process. It has been reported that instruments like Raman spectroscopy, infrared spectroscopy and mass spectrometry, combined with chemometric analysis provide a new level of ink examination (Silva *et al.*, 2014).

### 2.4.1 Stereomicroscope

Stereomicroscope is also known as dissecting microscope. It is an optical microscope that allows observation with two separate eyepieces. These two optical paths provide different angles to the left and right eyes resulting in a three-dimensional visualisation. Stereomicroscope is different from standard compound microscope by having relatively large depth of field, longer working distance and a relatively low magnifications, typically 10x – 40x. Principally, it uses incident light which is reflected from the object before entering the lens. Some may also incorporate transmitted light in the system. With the advantage of use of stereomicroscope, it is able to give direct views of objects due to its large field of view (Kreindler, 2012).

Leica MZ16 is a fluorescence stereomicroscope with 16:1 manual zoom with motorised focus being optional, 100% apochromatic optic system and built-in double iris diaphragmme for adjustment of depth of field. According to Leica Microsystems, it is a high-tech stereomicroscopes with vision availability to see 0.6-micron structures and save 80% more time with motorised zoom and automatic measurement. Provided with magnification of 2x Planapo objective 230x and resolution of 840 Lp/mm, it claims to save a great deal of time since there is no need to switch to a light microscope with interference contrast for accurate identification of samples (Microsystems, 2002).

Besides that, stereomicroscope can also be incorporated with cold light sources to provide incident light on samples. Cold light source was invented by Dr. Karl Storz in 1960. It is a standalone unit that generates light through the use of powerful lamps or LEDs (Endoscope, 2016). KL Series is an usual illumination for stereomicroscope applications. KL 1500 LCD for example is the main used light in the field of 150 watt halogen cold light sources. With constant colour temperature of halogen lamp and intensity control, it helps in optimising the lifetime of the halogen lamp and provides homogenous light output (SCHOTT, 2015).

### 2.4.2 Video Spectral Comparator (VSC)

Video Spectral Comparator is an imaging instrument incorporated with radiation filtered at various wavelengths that allows documents to be inspected for ink differentiation, visualise hidden security features and detect any obliteration or alteration (NFSTC, 2013). Functioning through visual observations and basic principles of light, it observes responses of ink in ultraviolet (UV), violet and infrared (IR) regions of electromagnetic spectrum.

VSC 6000 is an improved multi-functional workstation for questioned document examiners with a high resolution imaging system. It is composed of two components which are a desktop for software running and imaging system that features a FireWire 5

Mpixel CCD colour camera with zoom magnification up to 130x (*approx.*) (Foster and Freeman, 2012). This device is packed with various illumination and filtering devices to reveal differences in IR description, transmittance and luminescence and long wave UV excitation of visible luminescence (fluorescence) of ink. This instrument facilitates image processing and enhancement of questioned documents examination. Incorporation of intelligent software enables automatical setting for change compatible combinations of illumination and imaging (Foster and Freeman, 2012).

VSC 6000 can also perform spectral investigation similar to spectrometers via microspectrometer that is of real-time grating for recording absorption, reflectance, transmittance and fluorescence spectra (Foster and Freeman, 2012). However, it was pointed out in a study that VSC was not calibrated as an analytical instrument, and thus, the results are indicative instead of definite. VSC was unable to discriminate colour mixture of pigment and dyes for individual pigment identification (Lera *et al.*, 2013). Despite that, the highlight of this instrument is its capability of providing quantitative measures of colours, or colorimetry analysis based on the CIE (*Commission Internationale de l'Eclairage*) colour standards (Allen and Lera, 2013). It measures reflectance data to compute colorimetry parameters defined by CIE system (Caswell, 2013).

## 2.5 Light, colour and colorimetry

Colorimetry is the science and technology used to quantify and describe physically the human colour perception (Herenden *et al.*, 2011). The concept of colorimetric analysis is based on determination of concentration of a substance by measuring relative absorption of light with respect to a known concentration of the substance. The best colour comparator device is actually human eyes which are very sensitive. It only requires a few quanta of light for the detection of a signal. Even so, the human eyes are unable to measure light (Ghosal *et al.*, 2011). Thus, the emergence of colorimetric analysis in analytical approach as shown by VSC is beneficial to aid human in measuring colour density based on numbers of coordinate in chromaticity.

Human sense of sight is guided by light and light is actually a form of electromagnetic radiation as shown in Figure 2.1. Depending on the composition of specimen and combination of excitation wavelengths, there are three things that can occur which are:

- Reflection: Whole of the light or a part of it can be reflected off the specimen making it appear white or lighter.
- Absorption: All or most of the light can be absorbed by the specimen making it appears black or darker.
- Transmission: Light can be transmitted through the specimen.

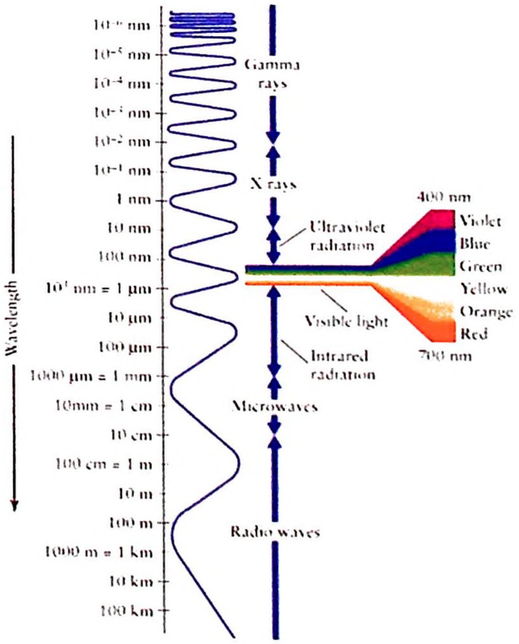


Figure 2.1: Wavelength adapted from Freedman and Kaufmann (2011).

Nowadays, instruments like VSC, Raman and infrared spectrometry had incorporated light source to differentiate object. Based on spectrophotometry principle, colour intensity is a measure of the concentration of material present in the matter, whereas matter absorbs and transmit only certain type and range of light. Thus, spectral analysis could acts as a 'fingerprint' of a compound (SUNY, 2005). The visible part for humans is in the wavelength range of 380 nm (violet) to 780 nm (red) which each frequency value corresponds to a distinct colour.

Colours are affected by the light of surrounding or the nature of the surface object as human perceived due to selective absorption of certain wavelengths of visible light and transmittance of the remaining wavelengths (SUNY, 2005). Due to the different and unique human brain and eyes, colours sometimes are interpreted differently due to perceptual factor. So, a system to give a certain value to colour has been established.

In 1931, CIE or translated as International Comission of Lighting helps to standardise the definition of colours by characterising colours of the light perceive by human brain. There are few colour spaces introduced by CIE over years for better reflect of human visual perception of colour like CIE 1960 and CIE 1976 (Herenden *et al.*, 2011). This colour system is presented as colour spaces that is described as chromaticity diagram shown in Figure 2.2.

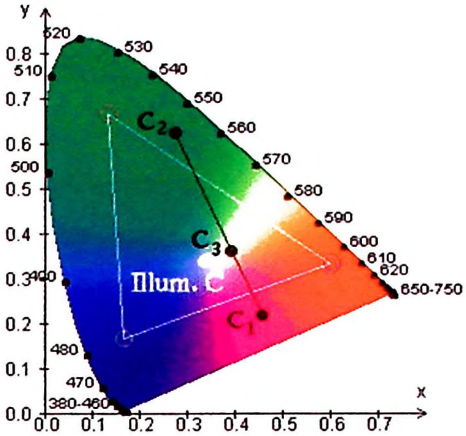


Figure 2.2: Chromaticity diagram adapted from Vishnevsky (2007).

The numerical measure of colour presented in chromaticity coordinates are as follows (Korifi *et al.*, 2013):

- X, Y and Z represent the tri-stimulus values
- x and y represent the coordinates in the chromaticity diagram (when the 1931 x,y option is selected)
- u and v represent the coordinates in the chromaticity diagram (when the 1960 u,v option is selected)
- L* represents the luminosity of the sample ranging from 0 (black) to 100 (white)
- a* and b* represent the colour. a*: negative values indicate green while positive values indicate magenta. b*: negative values indicate blue and positive values indicate yellow

## 2.6 Chemometric

Immense amount of data produced by modern instruments for example analytical instruments such as, are handled by multivariate analysis to analyse and convert data variation into meaningful information (Matero *et al.*, 2013; Workman. *et* *al.*, 1996). By definition, multivariate analysis is the analysis of multiple variables in a single relationship or set of relationships (Hair *et al.*, 2010). In chemistry, chemometric is considered as one of the common multivariate analysis applied for analysis of chemical data.

Wold (1995) pointed out that, "the art of extracting chemically relevant information from data produced in chemical experiments is given the name of 'chemometrics' in analogy of biometrics and econometrics. Chemometric like other 'metrics', is heavily dependent on the use of different kinds of mathematical models (Wold, 1995). This task demands knowledge of statistics, numerical analysis, operation analysis and more and in all, applied mathematics. However, as in all applied branches of science, the difficult and interesting problems are defined by the applications. In chemometrics, the main issue is to structure the chemical problem to a form that can be expressed as a mathematical relation. Therefore, chemometrics must not be separated from chemistry, or even be allowed to become a separate branch of chemistry. It must remain as integral part of all areas of chemistry." (Wold, 1995)

As highlighted by Wold, the convention of chemometrics originates in chemistry related to the use of computers for working statistical and mathematical methods. It was first established in early 1970s with one of the initial aims was to make complicated mathematical methods practicable (Otto, 2007). Chemometric uses mathematical and statistical methods to design or select optimal measurement procedures and experiments, and to provide maximum chemical information by analysing chemical data (Tauler, 2014).

In relation to forensic science disciplines, the use of multivariate analysis is also common as demonstrated in many research literatures (Almeida *et al.*, 2013; Baerncopf *et al.*, 2010; Desa *et al.*, 2010; Hupp *et al.*, 2008; Mehmedic *et al.*, 2010; Mohamad *et al.*, 2014; Willard *et al.*, 2012). Many previous works applied multivariate analysis for sample and individual characterization, including ink analysis, questioned document analysis, drugs analysis and fire debris, etc. Figure 2.3 showed algorithms of multivariate analysis.

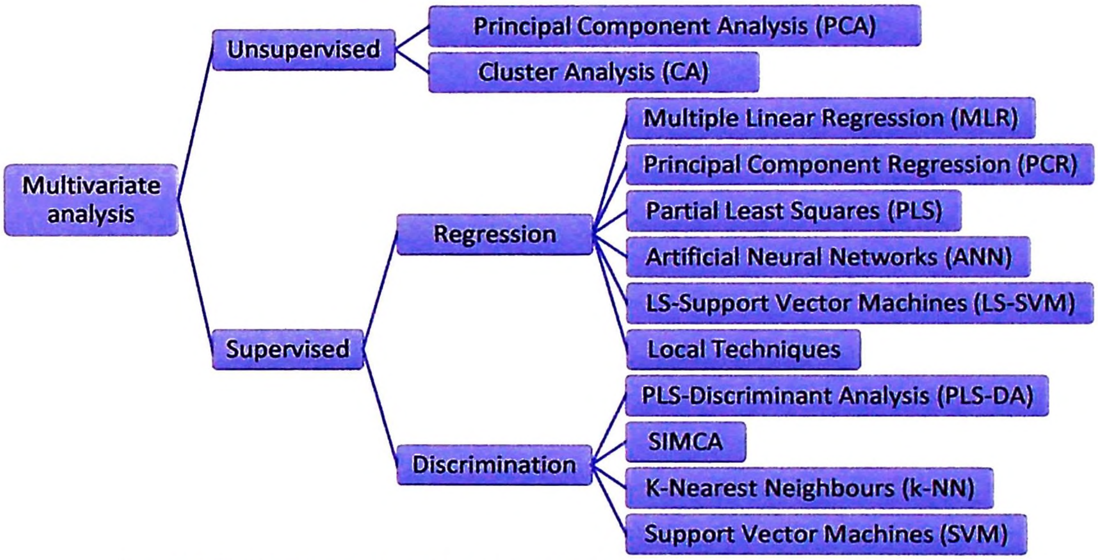


Figure 2.3: Algorithms of multivariate analysis adapted from Pierna *et al.* (2015).

### 2.6.1 Pearson Product Moment Correlation (PPMC)

Pearson Product Moment Correlation (PPMC) is a multivariate analysis that focuses on assessing correlation between two variables that quantifies the strength and direction of such relationship. The correlation coefficient is represent as rho (ρ) of Greek letter for a population and letter "r" for a sample (Andale, 2012). The idea of this relationship is that, if there is a change in one variable of correlated variables, the other would also change either in the same or reverse direction. Thus, PPMC result always noted with $r$ value that range between -1 to 1.

1 (strong, positive correlation); 0 (no correlation); -1 (weak, negative correlation)

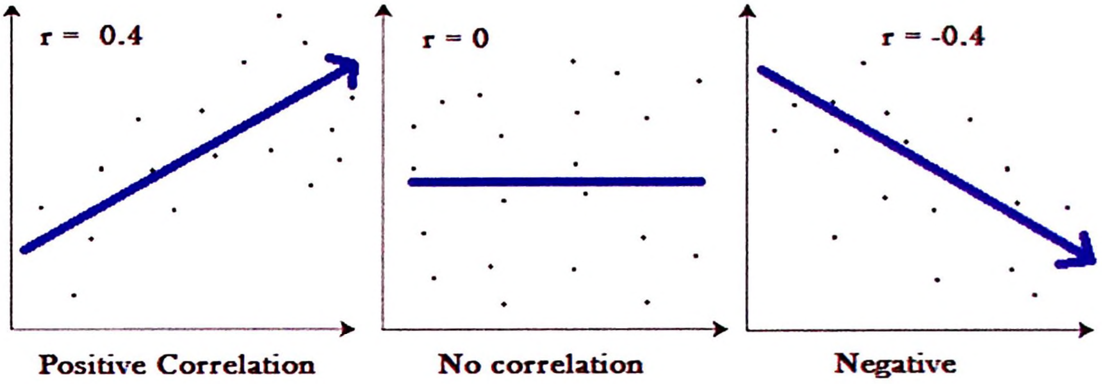


Figure 2.4: Pearson correlation with corresponding "*r*" value adapted from Andale (2012).

Based on Figure 2.4, the data points are shown by its distribution around the line of best fit (blue line) which is based on strength of the relationship between variables. When there is no correlation between two variables where r=0, the scattering of data points would be far from the line of best fit and vice versa (Andale, 2012).

PPMC has its drawbacks that should be noted. It is stated that *r* only measures linear relationship, and thus, *r* may not help if it is not a straight line (Mathematics Enhancement Programme, 1995). PPMC was used in different field to find correlation between studied variables. One of it was identification of evaporated ignitable liquid residues by comparing with their corresponding liquid standard (Prather *et al.*, 2012).

### 2.6.2 Principal Component Analysis (PCA)

Principal Component Analysis (PCA) is an unsupervised multivariate analysis that applied to observe clusters of samples which lead to discrimination of samples. The clustering depends on similarity of variables measured between the samples. If the samples are of similar characteristics, they would cluster together indicating similar origin and vice versa. It is the most widely used multivariate technique in chemometrics study. The idea is based on two principals which are (i) number of significant Principal Components (PCs) scores and (ii) characteristics of PC that related to the samples and variables. PCA usually presented in a scree plot with PCs against each other (Brereton, 2007) as in Figure 2.5.

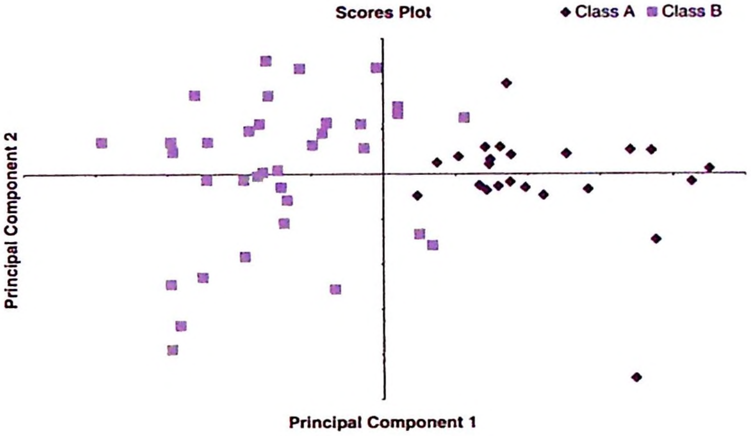


Figure 2.5: Scree plot presentation of PCA result adapted from Brereton (2007)

PCA aims to reduce dimensionality of data but still retain a maximum amount of variance in the data set (Gemperline, 2006). This analysis requires for data to be mean-centered and this was done by calculating the average for each variable which then subtracted from each data point. This is to make sure the normalisation over the entire data set and maximum variation from the origin (Baerncopf *et al.*, 2010; Hupp *et al.*, 2008).

### 2.6.3 PPMC and PCA in forensic analysis

In forensic analysis, multivariate analysis methods proved to be useful in interpreting outcomes of Gas Chromatography/Mass Spectrometry (GC/MS) and provide identification of samples by data distribution using certain analysis. In a study, both PPMC and PCA were used in individualising ignitable liquid in fire investigation. Six ignitable liquid residues (petrol, diesel, fresh paraffin lamp, adhesive remover, torch fuel and paint thinner) in the presence of matrix interference and burned at different conditions were identified. All of the residues were reported to be able to trace to their original products (Viitala and Hyyppä, 2013).

A similar work was also performed by Baerncopf *et al.* (2010). They characterise five diesel samples by manipulating the effect of temperature. Statistical analysis was incorporated due to difficulties in visually assessing results of Total Ion Chromatogram (TIC) and Extracted Ion Profiles (EIP). They aimed to associate ignitable liquid residues (ILR) to neat ignitable liquids and also to individualise the ILR from matrix interferences. PPMC and PCA displayed consistent in the association and discrimination of diesel samples. PPMC reported a higher average coefficients between replicates of same diesel than those between different diesel samples while PCA was proven to figure the class of unknown ILR by comparing it to reference collection. These methods were found to be useful in discrimination for aromatic profile but not for alkane profile due to similar alkane content among the samples (Baerncopf *et al.*, 2010).

In 2008, Hupp *et al.* also used GC/MS to analyse diesel fuel samples and the chosen ions were tested with PPMC and PCA. PPMC result showed that diesel samples from the same brand had higher correlation and samples from different brand demonstrated lower correlation. They also revealed that PCA result was complimented to PPMC result, proven by close clustering of diesel samples in PCA which had high correlation in PPMC while the ones that are from different clusters had lower correlation (Hupp *et al.*, 2008).

In different analysis by The University of Mississippi, they inquired the potency of cannabis preparation concentrations of $\Delta^9$-tetrahyrocannabinol ($\Delta^9$-THC) on all drugs seized during 1993-2008. From GC analysis, the results were furthered with statistical analysis using PPMC. Mean and standard deviation of sample concentrations were calculated between the correlated data set, by years and sample types, and for domestic and non-domestic samples. It showed an increase in the mean of $\Delta^9$-THC from 3.4% to 8.8% during the study period (Mehmedic *et al.*, 2010).

The most recent work done using these particular multivariate analysis was by Willard *et al.* (2012) in discriminating *Salvia* species based on the extraction of active compound *Salvia divinorum*. Apart from visual assessment of chromatograms generated by GC/MS they carried out PPMC on PCA scores to provide numerical assessment of discrimination. PPMC was also in aim to foresee its advantages and disadvantages for forensic applications. The outcome was that PCA showed clear distinction of *S. divinorum* from other *Salvia* species by the close position of replicated species. PPMC meanwhile showed strong correlation among replicates of each species. They concluded that, PPMC was useful in indicating absolute level of discrimination and useful in forensic purposes (Willard *et al.*, 2012).

## 2.8 Fake banknotes studies

In past years, banknotes authentications were evaluated using various instruments and methods like infrared spectroscopy, Raman spectroscopy, mass spectroscopy, Artificial Neural Network (ANN) and Full-field Optical Coherence Tomography (FF-OCT). An analysis using fibre structure of paper to determine authenticity of documents were conducted by Takalo *et al.* (2014) where stamps and banknotes were analysed under transmitted light.

Authentication determination using spectroscopy either infrared spectroscopy or Raman spectroscopy were carried out by Vila *et al.* (2006), Badovinac *et al.* (2010), Pal and Pratihari (2012), Guedes *et al.* (2013), Almeida *et al.* (2013), Sonnex *et al.* (2014) and Imperio *et al.* (2015). Vila and colleagues (2006) analysed euro notes of different countries from Spain, Germany, France, Italy with fake banknotes provided by the authority worth €50 and €100 using Fourier Transform Infrared (FTIR) spectrometer with Attenuated Total Reflectance (ATR) mode microscope. Comparison were made between spectra recorded on blue colour of European flag, red letters of The European Central Bank (green letters in €100), shiny hologram and white area of the banknote (Vila *et al.*, 2006).

Other than banknote authenticity study, Badovinac *et al.* (2010) performed micro-Raman and X-ray fluorescence (XRF) analysis on historical postage stamps of Hungary in 1918. Comparison was made between pigments used on original banknote with pigments on trial prints, suggesting some differences between inks of original and fake overprints of the postage stamps. This also applied on the banknote one Lira that showed difference in ink pigment composition between original and trial prints notes.