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Original Article

Forensic Analysis of Gel Pen Inks using Hyperspectral Imaging coupled with Chemometric Procedures

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التحليل الجنائي لأحبار الأقلام الهلامية باستخدام التصوير فوق الطيفي والمقترنة بإجراءات قياس كيميائية.

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Abstract

Forensic examination of inks on questioned documents has become an important practice and the law enforcement agencies rely heavily on these techniques during criminal investigations. Although nowadays there are a variety of methodologies focused on the analysis of inks, the combination of non-destructive nature of Hyperspectral Imaging (HSI) coupled with the multivariate chemometric technique has received little attention.

In this study, forty-five ($n = 45$) gel pen inks of three different colours i.e. blue, red and black of five different brands were analysed using HSI with the idea of classifying them according to the brand.

In terms of discriminating similar coloured components between samples, this was achievable only on the basis of the % reflectance spectra. However, arguably, there was sufficient evidence to suggest that it may be possible to discriminate the samples using chemometric of Principal Component Analysis (PCA). A 2-D score plot from PCA analysis was enough to characterise the samples into five clusters.

The synergy of complementary information provided by PCA narrows matching possibilities (in terms of classification) for forensic investigations involving ink analysis.

المستخلص

أصبح استخدام التحاليل الجنائية للأحبار على الوثائق المشكوك فيها أمرًا شائعًا، وتعتمد وكالات إنفاذ القانون بشكل كبير على هذه الأساليب أثناء الأنشطة الإجرامية. وعلى الرغم من أن هناك العديد من المنهجيات التي تركز على تحليل الأحبار، فإن الجمع بين الطبيعة غير المتلفة للتصوير فوق الطيفي (HSI) مقترنةً بالتقنية الكيميائية متعددة المتغيرات حصل على القليل من الاهتمام. في هذه الدراسة، تم تحليل خمسة وأربعين (٤٥) حبر هلامي من ثلاثة ألوان مختلفة، الأزرق والأحمر والأسود من خمس علامات تجارية مختلفة باستخدام HSI مع اعتماد التصنيف وفقاً للعلامة التجارية. وكان التمييز بين المكونات الملونة المتشابهة بين العينات قابل للتحقق فقط وفق النسبة المئوية لطيف الانعكاس. ومع ذلك، يمكن القول إن هناك أدلة كافية تشير إلى أنه قد يكون من الممكن التمييز بين العينات باستخدام القياس الكيميائي لتحليل المكونات الرئيسية Principal Component Analysis (PCA). وكانت نتيجة المقطع ثنائي الأبعاد لتحليل الـPCA كافية لتمييز العينات إلى خمس مجموعات.

وعليه إن تضافر المعلومات التكاملية التي يوفرها فحص PCA يقلل من احتمالات التطابق (من حيث التصنيف) في التحقيقات الفنية الجنائية الخاصة بتحليل الأحبار.

Keywords: *Forensic Science, Questioned Document, Hyperspectral Imaging, Principal Component Analysis, Gel Pen Inks.*

الكلمات المفتاحية: علم الأدلة الجنائية، الوثائق المشكوك بها، التصوير فوق الطيفي، تحليل المكونات الرئيسية، أحبار الأقلام الهلامية.

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1. Introduction

1.1 Forensic Ink Analysis

Historically, prior to the introduction and widespread distribution of ballpoint pens and the subsequent development of more contemporary writing instruments (gel pens, fountain pens and roller pens), ink analysis depended largely on a combination of physical and chemical examinations using lights of different wavelength and relatively simple micro-chemical spot tests [1]]. The physical examinations involved the use of infrared (IR) photography or Ultra Violet (UV) fluorescene photography (useful for detecting chemical erasures) [1]. Since IR and UV use light of very different wavelengths, certain classes and colours of ink would exhibit different behaviours under these lighting conditions, providing a means of differentiating inks on paper. Simple chemical spot tests involved adding, either insitu1 or by removal of a part of the ink line to a microscope slide, micro-droplets of chemical reagents such as 2% dilute sodium hydroxide solution [1]. Any resulting colour change was obeserved through a magnifying lens or microscope and used as a characteristic feature to determine a particular class of writing ink, e.g. fountain pen or ballpoint [1]. These kinds of tests were essentially non-destructive and required little or no specific expertise to perform [1]. However, they were not suitable for identifying specific ink formulations within a class and were therefore of limited evidential value [1].

In forensic questioned documents, analysis of inks have been carried out by the following techniques:

- Electrophoretic Techniques: Capillary Electrophoresis [2].
- Mass Spectrometric Techniques: Gas Chromatography-Mass Spectrometry (GCMS) [3], Inductively Coupled-Mass Spectrometry (ICP-MS) [4], Direct Analysis in Real Time (DART) [4] , Time of Flight-Secondary Ion Mass Spectrometry (ToF-SIMS) [5], and Laser Ablation-Inductively Coupled Plasma-Mass Spectrometry (LA-ICP-MS) [6].
- Other Spectroscopic Techniques: Microspectrophotometry (MSP) [7], Fourier Transform Infrared Spectrometry (FTIR) [8].

1.2 Key Issues

The term Filtered Light Examination (FLE) refers to the examination of inks under different lighting conditions, namely IR absorption/reflectance, IR luminescence/fluorescence and UV fluorescence. These examinations have been performed for several decades, but technological developments have meant that the use of IR and UV photography to perform such examinations have been replaced in most laboratories by specialist equipment such as the Video Spectral Comparator (VSC) produced by Foster and Freeman Ltd (UK), the latest version of which (VSC 8000/HS) incorporates both digital technology and Hyperspectral Imaging (HSI) capabilities [9]. In these cases, an image of the questioned documents placed on a stage inside the VSC is provided via a charge coupled device (CCD) onto either a self-contained visual display unit in older systems or onto a computer screen via a connected PC workstation in the latest versions [9]. The ink present on the document can be viewed under different lighting conditions using a system of in-built illumination sources, filters (i.e. coloured, dichroic, and interference filters), lenses and sensitive tubes [10].

The application of filtered light to the examination of writing inks, in particular ballpoint inks, is well documented [13-14]. However, the application of filtered light examination to gel ink samples has occurred more recently. Gernandt and Urlaub [13] analysed four Japanese brands of gel ink pen available In the United States (US) market using the the infrared (IR) and Ultra Violet (UV) functions of a Video Spectral Comparator (VSC-1). Three brands were available in black and blue, and one in red, all of which the authors claimed exhibited no UV fluorescence or IR fluorescence/ luminescence (IRL) when viewed under the VSC-1. Two brands were available in several colours and exhibited a range of fluorescence, though the authors highlighted these were specifically manufactured to have fluorescent properties. Wilson et al. [14] used a VSC 2000/HR to discriminate between 20 pigmented black gel and nine dye based black gel inks under visible and Near Infrared (NIR) reflectance (400- 1000 nm) and IRL conditions (400-580 nm band pass filter/610 nm cut off filter and 480 – 620 nm band pass filter/665 nm cut off filter).

Despite their modern approach, the authors still urged caution when considering their findings using direct visual examinations presumably due to its inherent subjective nature. Recently, Hyperspectral imaging (HSI) has a number of advantages as a technique for providing structural information about a sample that is of particular interest. In particular, a sample can be analysed in-situ, and analysis time is relatively less, in the range of a few seconds [9]. In VSC 6000/HS, it is possible to examine a document across a range of 400-1000 nm at incremental steps of between 1-20 nm in order to produce a digitally stored image cube [9]. An operator can playback the images as a real time movie to determine how two or more inks behave across the wavelength range, making it easier to visualise differences in their appearance under filtered light. In addition, the intensity of each pixel in each image is plotted as a function of wavelength, allowing the operator to produce a reflectance or fluorescene spectrum of the ink at any given wavelength in order to detect subtle spectral differences between chemically similar inks [9]. The potential for enhanced discrimination together with the non-destructive nature of this technique and minimal sample preparation means HSI has considerable benefits over existing non-destructive filtered light examination techniques.

To date, there is only one study investigating the use of HSI coupled with chemometrics applied to writing inks in the literature [15]. Reed et al. [9] investigated the use of HSI to examine forty-two different gel pen inks (blue and red inks) representing different manufacturers, as lines drawn on office paper. Reflectance spectra representing within ten measurements of the sample ink exhibited excellent reproducibility/repeatability and satisfactory signal when analysed over 400-1000 nm^{-1} wavelength range. Despite their modern approach, the authors still urged caution when considering their findings using direct visual examinations, presumably due to its inherent subjective nature. Indeed, the authors commented that the ability to apply a multivariate statistical methodology across the full reflectance spectral range suggested it may be useful for better discriminating between colours of gel ink. Again, Chlebda et al. [15] used HSI in combination with chemometric techniques (PCA and HCA) to discriminate 45 black gel pen inks available on the Polish market. The combined approach using these exploratory approaches (PCA and HCA) focussed on the different characteristics of the writing inks and was found to be highly effective for the discrimination of inks. The work was presented in an attempt to address the deficit in the literature with regards to the HSI of gel pen inks. No studies using HSI techniques involving gel pen inks (combination of blue, red and black colours) have been conducted, let alone using pattern recognition technique of PCA for characterisation and discrimination of the inks.

2. Materials and Methods

2.1 Sample Collection

Five different brands representing 45 gel pen inks taken from three countries (Malaysia, Japan, and Indonesia) were purchased from high street retailers and online suppliers. Brands were chosen to reflect a mixture of well-known stationary brands and some lesser-known and/or discount brands. Where possible, three pens of each brand for three colour groups blue, red and black were acquired, respectively (Table-1).

2.2 Instrumentation

HSI analysis was conducted using a VSC 6000/HS (Foster and Freeman, UK), a PC based document imaging system consisting of a main unit, PC system and bespoke VSC software.

The document is placed on the document platen within the main unit, and brought into view on the PC monitor via the video camera positioned directly above it. The video camera uses both a motorised zoom lens and a high magnification lens to give a maximum resolution of 2584 x 1292 pixels. Flood illumination for IR absorption conditions is provided by incandescent filament lamps, which produce white (visible) light and infrared radiation to provide an overall range of 400 – 1000 nm. White light emitting diodes (LED’s) are used to white balance the image, and long pass filters in front of the camera are used to view the image in the infrared. A halogen spot light (capable of producing 40–1000 nm) is used together with a continuously variable band pass filter placed in front of the light source in HSI mode. To control the wavelengths of illuminating

Table 1- *Sample of pen collected from three different countries.*

	Sample	Code	Country of Purchase
RED	Pilot G3	P	Malaysia
	G soft GS2 Gel Pen	GS	Malaysia
	Kilometrico	KM	Malaysia
	Stabilo	S	Malaysia
	Faber Castell Gel Rollerpoint	FC	Malaysia
BLUE	Partners Broad Gel	P1	Japan
	GStaples Sonix Gel	GS1	Japan
	Kilometrico R2	K1	Japan
	Staedtler RMG	S1	Japan
	G soft GS2 Gel Pen	FC1	Japan
BLACK	M& G R3 Retractable Gel Pen	P1	Indonesia
	Great Expression Gel	GS1	Indonesia
	Kilometrico	K1	Indonesia
	Staedtler R2	S1	Indonesia
	Faber Castell RX7	FC1	Indonesia

light, optical filters are placed into the path of the optical system. For IR absorption/ reflectance (IRR) examination, long pass filters are used to allow longer and shorter wavelengths of light rather than a specific cut-off wavelength value to illuminate the document, whilst band pass filters are used to allow a defined narrow range or band of wavelengths between a long and short cut-off wavelength value. The operation of the main unit is controlled using the VSC software, with each mode of operation associated with one of several working screens. For example, live and stored images of the document can be viewed via the main screen, which also controls the VSC settings, whilst spectral and chromaticity data can be viewed via the spectrum screen. Table-2 describes each of the viewing settings, their function in relation to the live camera image and whether they can be controlled automatically by the software.

The type of light source used is controlled by an illumination panel. The VSC 6000/HS incorporates Hyperspectral Imaging (HSI) technology, whereby it is possible to record images of a document using reflected light at specific well defined wavelengths across a spectral range of 400–1000 nm. Up to a maximum of 150 images can be taken and stored as an image cube, which can be replayed to view the wavelengths at which differences occur. A band pass filter step width between 5–10 nm can be selected to generate sufficient imagery data across the entire spectral range. Furthermore, spectral data from any given pixel in the image can be generated to compare any spectral differences within the document.

2.3 Instrumental Conditions

2.3.1 Development of Hyperspectral Imaging (HSI) Techniques

For HSI, the instrument was calibrated for the entire spectral range (400 – 1000 nm) permitted by the instrument, against a piece of A4 paper (Tesco, Malaysia). Preliminary experiments were conducted in terms of auto exposure (integration times 1.4 – 2.0 ms, iris (83%)) and auto focus modes (magnification 7.12) were switched on with the default brightness value (70) to focus on ink lines in order to produce well defined spectra.

Table 2- *Description of the viewing settings, their function in relation to the live camera image and software function.*

	Setting	Function	Auto- Controlled
Exposure	Brightness	Controls brightness of image across	Yes
	Gamma Correction	Can be applied to brighten darker areas within the image to reveal hidden details	-
	Iris	Controls the amount of light reaching the camera across a range of 0-100%	Yes (Auto Exposure)
	Integration Time	Controls the time permitted for the integration of the camera signal	
Focus	Magnification Zoom	Controls the optical zoom magnification	Yes (Auto Focus)

2.3.2 Spectral Acquisition

Spectral acqusition was obtained in reflectance mode. In order to perform a method of validation of this technique, six spectra were acquired from the same area of one blue, red and black gel ink, respectively, i.e repeatability and reproducibility analysis.

2.4 Data Analysis

PCA was conducted using the MINITAB® Version 16.2.3 statistical software (Minitab Incorporated, State College; USA).

3. Results and Discussion

3.1 Initial Discrimination of Reflectance Spectra

3.1.1 Red Inks

All five brand dye model combinations exhibited the same spectral pattern and were considered a single (same) spectral group (Figure-1). In this study, the reflectance spectra within red gel inks exhibited the same reflectance peaks (600-1000 nm). But upon closer inspection, the shape of the spectra for five brand combinations differed slightly from all others within the group (450-550 nm) (Figure-2). This uncertainty highlights the subjective nature of the interpretation of data from the HSI technique and demonstrates the need for a more definitive and reliable method of discrimination between such samples. However, in isolation, it is difficult to say with certainty that this subtle spectral difference is due to a real "chemical difference",

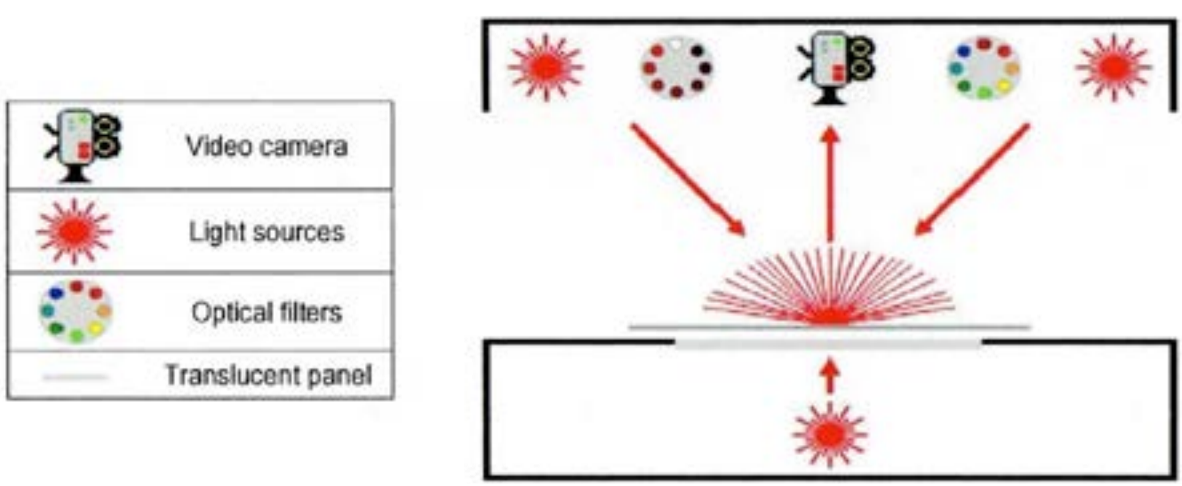

Figure 1- *Basic hardware component set up inside the VSC 6000/HS[14].*

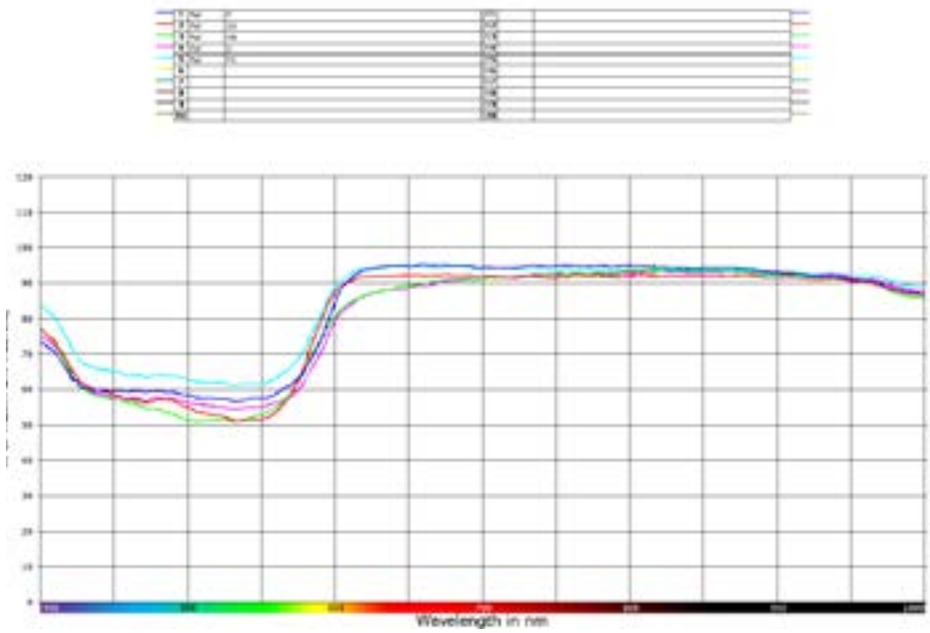

Figure 2- *A comparison of a set of five spectra from each of the red gel ink on paper highlighting a similar, yet distinguishable spectral pattern on the basis of differences in some peak position.*

and, therefore, in the author's opinion, cannot relied upon as evidence of within brand variation.

3.1.2 Blue Ink

The HSI data derived from the blue ink group displayed a wide variety of different spectral patterns suggesting a good level of discrimination between blue gel inks (Figure-3). In this study, the reflectance spectra within blue gel inks exhibited the same peaks (700-1000nm), but the shape of the spectra for five brand combinations (P1, GS1, K1, S1 and FC1) differed slightly from all others within the group (400-650nm). These spectral differences included a

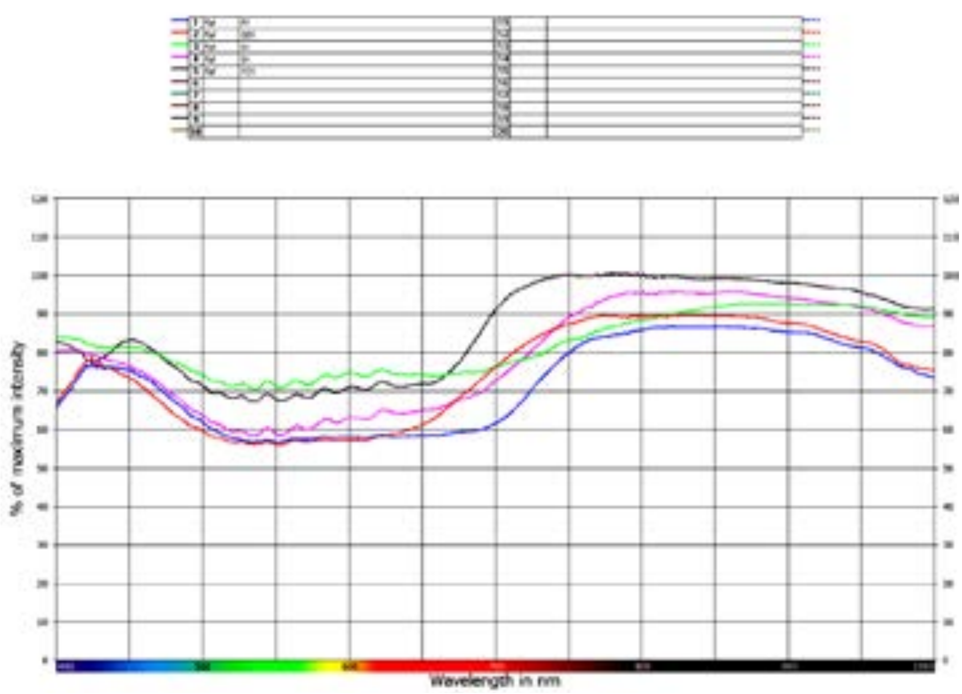

Figure 3- *A comparison of a set of five spectra from each of the blue gel ink on paper highlighting a similar, yet distinguishable spectral pattern on the basis of differences in some peak position.*

Table 3-*PPMC analysis for comparison of three ink samples (blue,red and black) analysed in triplicate analysis*.

	Pair-wise combination	r_1	r_2	r_3	r (average)
RED	P vs GS	0.8190	0.8190	0.8190	0.8190
	P vs KM	0.8254	0.8254	0.8259	0.8256
	P vs S	0.9249	0.9249	0.9249	0.9249
	P vs FC	0.7190	0.7191	0.7191	0.7191
	GS vs KM	0.7197	0.7197	0.7193	0.7196
	GS vs S	0.7391	0.7395	0.7397	0.7394
	GS vs FC	0.6710	0.6712	0.6717	0.6713
	KM vs S	0.6518	0.6716	0.6714	0.6615
	KM vs FC	0.6910	0.6910	0.6910	0.6910
	FC vs S	0.7190	0.7192	0.7193	0.7191
Black	P1 vs GS1	0.6567	0.6571	0.6751	0.6629
	P1 vs K1	0.7419	0.7419	0.7419	0.7419
	P1 vs S1	0.7165	0.7165	0.7168	0.7166
	P1 vs FC1	0.7219	0.7219	0.7219	0.7219
	GS1 vs K1	0.7619	0.7619	0.7619	0.7619
	GS1 vs S1	0.7417	0.7412	0.7411	0.7413
	GS1 vs FC1	0.7128	0.7128	0.7128	0.7128
	K1 vs S1	0.7191	0.7191	0.7191	0.7191
	K1 vs FC1	0.7005	0.7005	0.7004	0.7005
	S1 vs FC1	0.6211	0.6211	0.6210	0.6211
Blue	P1 vs GS1	0.6218	0.6218	0.6218	0.6218
	P1 vs GS1	0.6182	0.6182	0.6181	0.6181
	P1 vs K1	0.7318	0.7318	0.7318	0.7318
	P1 vs S1	0.7151	0.7151	0.7151	0.7151
	P1 vs FC1	0.7190	0.7190	0.7190	0.7190
	GS1 vs K1	0.8010	0.8010	0.8008	0.8009
	GS1 vs S1	0.8051	0.8051	0.8051	0.8051
	GS1 vs FC1	0.8110	0.8112	0.8112	0.8111
	K1 vs S1	0.8218	0.8217	0.8218	0..8217
	K1 vs FC1	0.8176	0.8174	0.8174	0.8175
	FC replicates	0.9255	0.9995	0.9997	0.9995
	K1 replicates	0.9990	0.9990	0.999	0.9999

PPMC; Pearson Product Moment Correlation, P; Pilot G3, KM; Kilometrico, S; Stabilo, FC; Faber Castel, P1;Partners Brand Gel, GS1; G Staples Sonix Gel, K1; Kilometrico R2, S1; Staedtler R2, FC1; Faber Castel Rx7 change in spectral shape in the 400-425nm-1 and 500-650 nm-1 regions, respectively. This provided further confirmation that these particular brand/model combinations were distinguishable by a real chemical difference in their composition.

3.1.3 Black Ink

The reflectance spectra of five black brand pens exhibited a characteristic large peak below 700 nm, and above that a comparatively featureless spectrum. Spectra from the S1, FC1, GS1, P1 and K1 combinations were almost "same", confirming that the pigment nature of the inks from solubility testing. However, a small reproducible well defined peak at 500-600 nm was observed in the spectra from pens FC1 and K1 (Figure-4) that was not as notable as in the spectra from pens P1, GS1 and S1 (Figure-4). The discrimination of the five pigment black inks combination (FC1, K1, S1, GS1 and P1) from all others was not possible using HSI alone. This suggests same in colour and additive composition may exist between different geographic locations, and these are not detectable by hyperspectral imaging. Indeed, this supports an earlier finding by Reed et al. (2014) that same black ink formulations tailored to end user of pigments may not be detectable by hyperspectral imaging techniques.

3.1.4 Pearson Product Moment Correlation (PPMC) Coefficients

Higher correlation values (greater than 0.80) obtained from the same brands of ink, indicate a strong relationship between them. The data obtained from PPMC analysis indicates that the differences observed in the correlation value of inks may come from the differences in chemical formulations of inks. Presence of a higher correlation value (0.9991 – 0.9999) between the same ink samples (in terms of replicates). Table-3 indicates PPMC analysis from three all ink samples.

3.2 Principal Component Analysis

The explanatory approach of PCA has been performed in this study to discriminate the inks. By comparing the score between the two principal components, a plot of PC1 vs PC2 can distinguish between the brands, respectively. Additionally, the feature shows that it is impossible (based on the VSC 6000/HS data alone) to say with certainty if these different classification groups are evidence that these

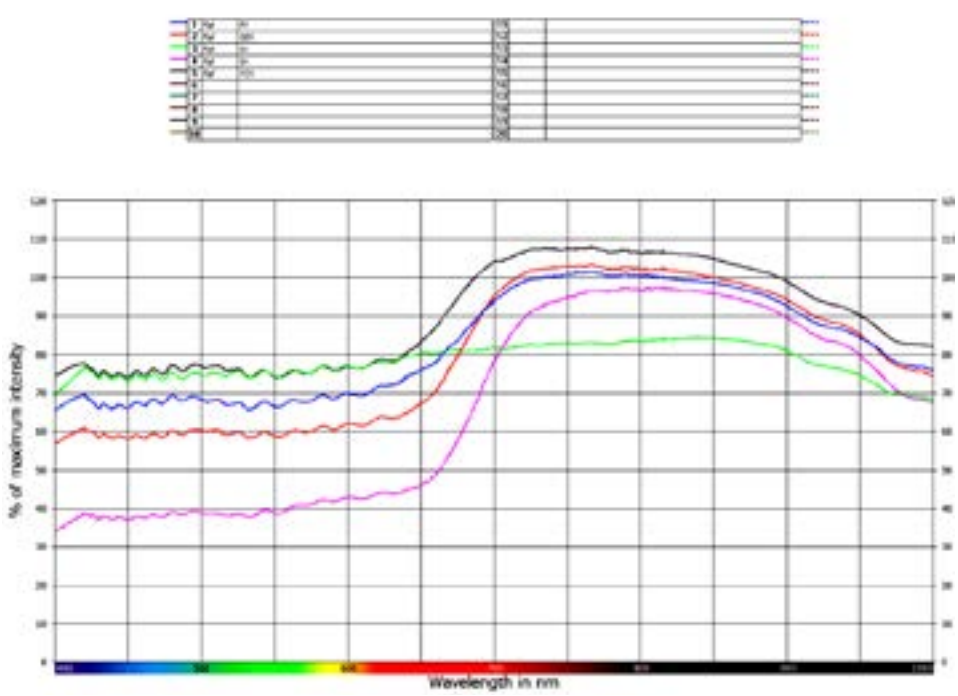

Figure 4- *A comparison of a set of five spectra from each of the black gel ink on paper highlighting a similar, yet distinguishable spectral pattern on the basis of differences in some peak position.*

samples can be discriminated on the basis of very subtle differences in the gel ink formulations, or whether they are as a result of the highly subjective nature of these techniques.

3.2.1 Red Inks

It can be clearly shown in Figure-5 that five groups have been produced using PCA, with all samples clearly distinguished from each other. Furthermore, within a group, the same samples show a higher level of similarity, suggesting less of a difference between their reflectance spectra (same sample). Interestingly, FC (light blue) and P (dark blue) which could arguably be divided into two same sub-group by having the same relative peak intensity (~450-550 cm^{-1}), could be distinguished using this explanatory approach of PCA. This is in contrast to the sub-classification of both inks (FC and P inks) observed in the reflectance data set, where PCA successfully identified two groups of high

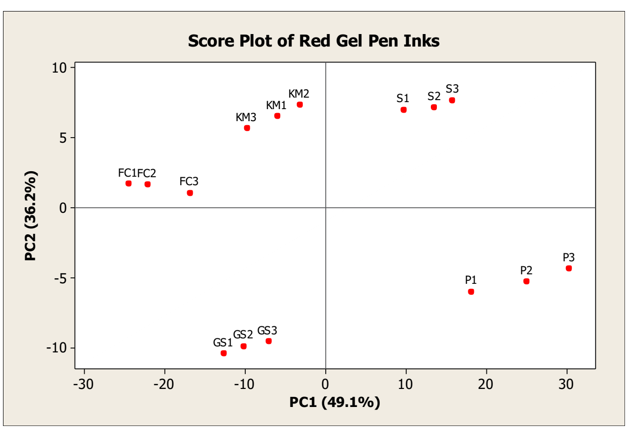

Figure 5- *2-D PCA score plot of red gel pen inks.*

similarity within the different cluster. Standardisation of the variables did not change the classification groupings, providing no further discriminatory value. Either way, it is believed that commercial competition has led to some manufactures (who lack the technical expertise and may be prohibited by patent issue rights) to use dyes in their own gel ink formulations rather than pigments. While pigments provide a range of added benefits, soluble dyes can be used in varied amounts to fine tune the hue and brightness of ink colour.

3.2.2 Blue Inks

The PCA shown in Figure-6 illustrates successful classification of the ink samples into five groups. Group 1 (P1) and Group 2 (GS1) ink samples are clearly distinguished, something which was not achieved for the same samples using direct visual inspection and PPMC analysis. Subtle differences in combined relative peak intensities of the wavenumber positions characterising the ink samples within GS samples and P samples can be observed. Furthermore, this is also in line with the sub-classification observed in the data set, where PCA successfully identified two groups of high similarity within the different cluster. Using the objective approach described, these three inks (K1, S1, and FC1) would then be classified as distinguishable from one another when in fact they are clearly distinguishable due to real difference in their chemical formulation. This is where the experience of an examiner is essential to the interpretation of reflectance spectra, and should not be underestimated despite its subjective nature. For this reason, it was decided to interpret comparisons of the reflectance spectra in this study using both the objective and subjective approach.

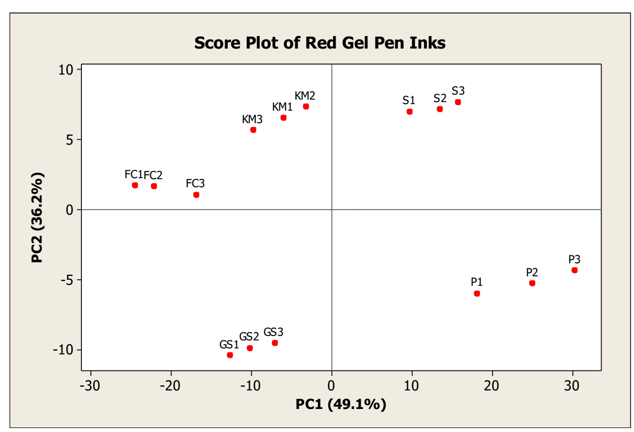

Figure 6- *2 –D PCA score plot of blue gel pen inks.*

3.2.3 Black Inks

Clustering into five groups by PCA, as demonstrated in Figure-7 , was in contradiction with the classifications made by visual comparison of reflectance spectra. Interestingly, all groups representing five ink samples in total are clearly discriminated from one another. These samples when analysed using direct visual represented a single group, that could only be distinguished into five subgroups on the basis of a relative peak intensity difference for a single peak, respectively. PPMC analysis of these two data sets could not find sufficient difference between them to classify them as separate groups. This demonstrates that multivariate analysis (PCA) at particular five ink samples was necessary in order to be able to confirm discrimination into separate groups.

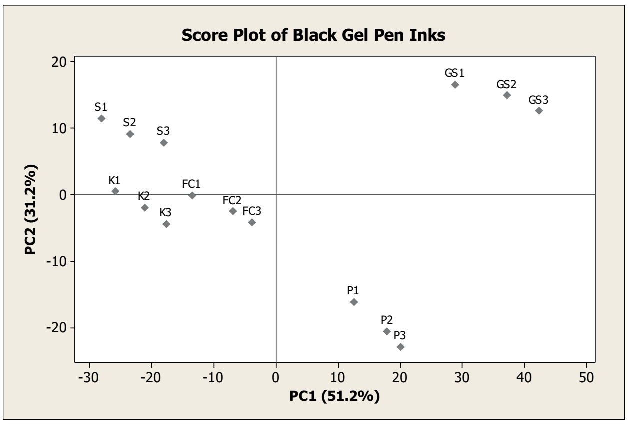

Figure 7- *2-D PCA score plot of black gel pen inks*.

4. Conclusion

The results obtained from HSI techniques showed that the discrimination of inks by visual comparison technique is more subjective. This suggests that it is therefore a more conclusive technique for the interpretation of evidence. The data gathered from HSI (reflectance spectra) coupled with chemometric techniques of PCA provides more objective conclusions to reveal ink differences. Further work has been conducted from this analysis i.e Linear Discriminant Analysis in order to predict the classification model from PCA analysis (16).

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Conflict of Interest

No.

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