

**FORENSIC EXAMINATION OF INK ON DIFFERENT  
SURFACES BY ATR-FTIR MICROSCOPY**

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OF THE REQUIREMENTS FOR THE DEGREE  
OF MASTER OF SCIENCE (FORENSIC SCIENCE)  
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Thesis  
entitled  
**FORENSIC EXAMINATION OF INK ON DIFFERENT  
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**FORENSIC EXAMINATION OF INK ON DIFFERENT SURFACES BY ATR-FTIR  
MICROSCOPY**

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**ABSTRACT**

In the examination of questioned documents on which letters or numbers may have been added, it is generally assumed that an ink written on paper gives the same infrared spectrum regardless of the type of paper. The technique of Attenuated Total Reflectance – Fourier Transform Infrared Spectroscopy (ATR-FTIR) has been applied to the comparison of four common blue ballpoint inks on 41 different common paper surfaces in an attempt to distinguish between the inks and between the paper surfaces. It has been found that the inks could be classified into four groups according to the general shapes and details in the fingerprint regions of their ATR-FTIR spectra. The paper surfaces themselves could also be similarly classified into four groups.

Therefore, a document examiner has to be extremely careful before giving an opinion on whether or not a document has been forged and chemical analyses may be needed for confirmation.

**KEY WORDS: INFRARED SPECTRUM / BLUE BALLPOINT PEN INK / ATR-  
FTIR TECHNIQUE / PAPER SURFACES / FORGED DOCUMENT**

56 pages

การตรวจสอบหมึกปากกาลูกลื่นบนพื้นผิวต่างๆในเชิงนิติวิทยาศาสตร์ ด้วยวิธี เอทีอาร์-เอฟทีไออาร์ สเปกโทรสโกปี

FORENSIC EXAMINATION OF INK ON DIFFERENT SURFACES BY ATR-FTIR MICROSCOPY

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บทคัดย่อ

ในการตรวจพิสูจน์เอกสารที่มีข้อสงสัยว่าจะถูกปลอมแปลงโดยการต่อเติมตัวเลขหรือตัวอักษร โดยทั่วไปมักจะสันนิษฐานว่า หมึกชนิดเดียวกันจะให้อินฟราเรดสเปกตรัมที่เหมือนกันไม่ว่าจะเป็นกระดาษชนิดใด เทคนิคเอทีอาร์-เอฟทีไออาร์ สเปกโทรสโกปี ถูกนำมาใช้ในการเปรียบเทียบหมึกสีน้ำเงินจากปากกาลูกลื่น 4 ชนิด บนพื้นผิวกระดาษที่แตกต่างกัน 41 พื้นผิว โดยสามารถแบ่งกลุ่มหมึกตามรูปร่างทั่วไปและรายละเอียดในช่วงที่มีความเด่นชัดเฉพาะของเอทีอาร์-เอฟทีไออาร์ สเปกตรัม ได้เป็น 4 กลุ่ม ส่วนพื้นผิวนั้นก็สามารถแบ่งกลุ่มได้เป็น 4 กลุ่มเช่นเดียวกัน

ผลการวิจัยแสดงว่า ผู้ตรวจพิสูจน์ต้องพึงระวังอย่างมากในการให้ความเห็นในเอกสารที่ถูกปลอมแปลง และการวิเคราะห์ทางเคมีอาจเป็นสิ่งจำเป็นในการยืนยันผลการตรวจพิสูจน์

56 หน้า

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## **CHAPTER I**

### **INRODUCTION AND OBJECTIVES**

Documents are unavoidable evidence in forensic caseworks because they are common in daily life. An original or official paper is relied upon as the basis, proof, or support of anything else within its most extended sense, including any writing, book, or other instrument conveying information in the case; any material substance on which the thoughts of men are represented by any species of conventional mark or symbol. Most documents are made from paper and completed in ink which is written by writing instruments such as pencil, ball-point pen, fountain pen, gel pen, or other substances which can be used for the same purpose. Nowadays, forged documents are a crucial problem in forensic science. Items are usually added to or erased from the original document which cannot be observed by the naked eye. However, they are significantly different in chemical composition when examined laboratory testing. The forgery specialists use lighting, spectrograph equipment, and the like to determine whether a document or parts of document have been erased, changed, or otherwise doctored. There are several techniques for ink analysis including Thin Layer Chromatography (TLC), High Performance Liquid Chromatography (HPLC), X-Ray Microanalysis, and Fourier Transform Infrared Spectroscopy (FTIR). But some of these techniques are destructive and require sample preparation. In general, forged documents should be firstly examined by non-destructive methods because they may be required for other tests in the forensic identification process.

Recently Attenuated Total Reflectance Fourier Transform Infrared microspectroscopy (ATR-FTIR), which is based on measuring changes that occur in a totally internal reflected infrared beam when the beam comes into contact with a sample, was introduced to Forensic Document Examination (FDE). Because ATR-FTIR is a non-destructive method and can penetrate the sample surface to approximately a 0.5 -2 microns depth, it can be used to analyze ink trace on a sample

surface. In this study, ink samples from four blue ball point pens written on 41 surfaces commonly found in the local market will be analyzed.

One problem which arises in document examination is that it is generally believed that a certain ink written on paper gives the same IR spectrum regardless of the type of paper. Ink comparison is, therefore, based on this assumption. If the assumption is wrong, however, many of the past results of document examinations may have been false.

This study, therefore, aims to verify, or falsify, the above assumption and to see whether the surfaces of various kinds of paper have any effect on the IR spectra of ballpoint inks. This was done by using ATR-FTIR spectroscopy to differentiate different inks on the same surfaces and distinguish between ATR-FTIR spectra of the same ink on different surfaces.

This investigation did not attempt to assign peaks in the IR spectra to vibrations of functional groups or to chemical bond types. Spectra were compared by peak positions, focusing on the fingerprint region (below  $1,400\text{ cm}^{-1}$ ).

## **CHAPTER II**

### **LITERATURE REVIEW**

#### **2.1 Composition of Paper**

Paper is made from cellulose and water, adding some filler to increase other properties. Materials which are used in paper making process can be divided into two groups:

*Fibrous raw materials:* wood pulp fibers (softwood and hardwood). The chemical components of fibers are cellulose, hemicelluloses, lignin and extractives.

*Non-fibrous raw materials* are separated in two groups:

I Functional additives increase the efficiency of existent or add new properties to paper.

II Chemical processing aids to help functional additives work better (Britt, 1970).

Paper can break up into several types, depending upon the purpose to use. Additionally, different manufacture processing, raw materials, chemical and paper machines have affected to the paper characteristics. Examples of paper names that were called according to the normally using are Newsprint, Bond paper, Wood Free paper, Kraft paper, Cover paper, Drawing paper, etc (Richard and Reed, 1984).

#### **2.2 Composition of Ball Point Pen Ink**

Ball point ink is frequently encountered in questioned document examination because it very popular and widely used. The ink of ball point pen is viscous and insoluble in water. The three main components are solvents, dyes & pigments, and resins & polymers.

Ball point ink was made with oil-based solvents: mineral oil, linseed oil and ricinoleic acid. After 1950, the oil-based solvents were changed to glycol based because of the common use of ethylene glycol as a dye solvent. The commonly used solvents are phenoxyethanol, phenoxyethoxyethanol, dipropylene glycol, oleic acid, benzyl alcohol, 2-pyrrolidone, butylenes glycol, etc.

Dyes are aromatic coloring agents which are usually soluble in organic solvents. The color of dyes is a result of their absorption of a portion of visible electromagnetic (EM) radiation. Ballpoint inks contained soluble dyes which dissolved in solvents as ethylene glycol such as Victoria Blue, rhodamine B and 6G, Methyl Violet and tetramethyl-pararosanilines. In 1960s, copper phthalocyanines, which were metalized dyes, were used in the ink industry.

Pigments are not similar to dyes. Pigments consist of natural or synthetic, organic or inorganic coloring agents. In addition, they are produced as quite insoluble finely divided particulate powders poised in a liquid solvents. Some compounds can be both pigments and dyes, depending on their form.

Resins are synthetic substances of high molecular weights that can be solids or amorphous semi-solids, which are soluble in organic solvents but not in water. They are used to adjust the viscosity of ballpoint ink. If resins dry, they will build a chemical bond between ink and paper (Crawford, 2003 and Ezcuura *et al.*, 2010).

**Table 2-1: Examples of Modern Ballpoint Ink Formulations***Example 1*

<b>Blue</b>	<b>%</b>
A-2 Resin Solution	35.9
Oleic Acid	11.3
Carbitol Solvent	12.5
1,3 Butylene Glycol	11.6
Victoria Blue Base	5.4
Victoria Blue Base BGO	8.6
GG (dye materials)	9.1
Methyl Violet 2B Base	2.6
Polyvinylpyrrolidone (PVP) K30	1.6
Yellow Tx	1.4
	100.0

*Example 2*

<b>Blue</b>	<b>%</b>
A-3 Resin Solution	55.5
Glyceryl Monoricinoleate	35.3
Oleic Acid	3.7
Methyl Violet 2B Base	3.7
Victoria Blue B Base	1.8
	100.0

**Table 2-1: Examples of Modern Ballpoint Ink Formulations***Example 3*

<b>Blue</b>	<b>%</b>
Hexylene Glycol	25.0
50% MHP/C90	15.5
Victoria Pure Blue BGO Ex	16.6
MBSN	16.6
Oleic Acid	4.9
Diethylene Glycol	7.5
Phenyl Methyl Carbinol	1.5
Acetopheone	0.5
Glyceryl Monoricinoleate	3.2
Carbitol	5.4
Methyl Violet 2B Base	1.1
Black NJ (Dye material)	1.2
GRL	1.0
	100.0

**2.3 Forensic Examination of Ink**

Because there was mentioned about a individuality of ink that had not sufficient enough to identify source of instrument (Neu mann, 2010) Thus, there are many techniques to analyze ball point inks including Thin Layer Chromatography (TLC), High Performance Thin Layer Chromatography (HPTLC), Raman Spectroscopy, visible microspectrometry, High Performance Chromatography, Gas Chromatography-Mass Spectrometry (GCMS) and Fourier Transform Infrared Spectroscopy (FTIR) (Tappolet, 1983; Totty *et al.*, 1985; Claybourn and Ansell, 2000 and Bugler *et al.*, 2005).

Thin Layer Chromatography (TLC) is widely used for ink comparison and analysis, because no visualization technique is needed and it is rapid, inexpensive and minimally destructive. In 2008, Djozan et al. (Djozan et al., 2008) proposed a new method for discrimination of blue ballpoint inks and comparison of writing inks on a document which is Thin Layer Chromatography and image analysis (TLC-IA). This method was recommended by the Research Center of Criminal Police of Iran. It combines the specific image software to analyze the chromatogram of TLC after scanning. The new method was simple and easy and needed no expensive instrument. However, TLC was utilized only for examination colorant substances in inks, not for other chemical components.

Due to the fact, the TLC method is not available for all forensic document examination and it's destructive in nature. So, several studies were focused on FTIR which is a non-destructive. In 2004, Rasool and co-workers (Rasool *et al.*, 2004) studied the use of FTIR microscopy to examine the ink in inkjet printer cartridges. They indicated that this technique was rapid and eliminated sample preparation but it may be not sensitive to identify minute artifacts to identify batch to batch differences in similarly colored inks. Beside ink sample, in the same year, Fourier transform infrared (FITR) combine with Raman spectroscopy (RS) was used to analyze a 40 market green spay paints. Based on the composition of the paint, 12 main groups were distinguished by FTIR. Later the pigment content was determined by RS using two laser sources which are Argon ion and Helium-Neon. For this study, the authors mentioned that combination of FTIR and RS has been the important technique for paint analysis (Buzzini and Massonnet, 2004).

Raman spectroscopy can applied individually to analyze blue gel pen ink. In 2005, Mazzella and Buzzini used Raman spectroscopy to analyze 55 blue gel pen inks available in present using two wavelength laser sources which are Argon ion laser (514.5 nm) and NIR diode laser (830 nm). They stated that using at least two laser sources given more advantage in analytical process. The result showed that when used these combined laser sources, 36 of 55 of blue gel pen inks which different brands and models were discriminated with the RS power was 0.68. In addition, they separated these 55 gel pen inks into two classes based on their solubility in methanol. One class

was dye-based gel ink and another was pigment-based gel ink (Mazzella and Buzzini, 2005).

Laser Desorption Ionization Mass Spectrometry (LDI-MS) supplement with High-performance Thin-Layer Chromatography (HPTLC) has been used for differentiation of blue ball point pen inks (Weyermann *et al.*, 2007). LDI-MS is non-destructive and rapid method that would be use tool for forensic ink examination. However, it should be note that this method can be affected by ageing of ink dyes.

Four years later, in 2008 Valerio *and team* reported that the combination between FTIR Spectroscopy and TLC or UV-vis spectroscopy were very useful for forensic document examiner since FTIR can analyzed other chemical components in ink such as resins and solvents. In addition, Attenuated Total Reflectance Fourier Transform Infrared micro-spectroscopy (ATR-FTIR) is a novel technique which is non-destructive, simple and does not require any sample preparation (Valerio *et al.*, 2008). Since the contact area between penetrator and surface sample is less than 2 microns, the specific area on the surface can be selectively analyzed. These advantages provide differentiation of ink trace because unique spectral features are related to chemical composition, brand, and color of an individual ink (Ekgasit *et al.*, 2007). However, detection with FTIR technique, solvent of ink can be influence the result, but higher in the first 1-2 months after the inks was deposit into paper (Weyermann *et al.*, 2007).

## **CHAPTER III**

### **MATERIALS AND METHODS**

#### **Theoretical Background**

Attenuated Total Reflectance Fourier Transform Infrared Spectroscopy (ATR-FTIR) has a strong evanescent field which protrudes only 0.5-2 microns and is generated under total internal reflection and its unique exponential decay characteristic for probing the molecular information at the surface and interface. The depth of an ATR spectrum depends on the experimental condition and material characteristics. The attenuated energy from evanescent field is passed back to the IR beam and to the detector. After that, the system generates an infrared spectrum and shows on the computer display. The requirements to use this technique are:

- The sample must be in direct contact with the ATR crystal
- The refractive index of the crystal must be significantly greater than that of the sample or else internal reflectance will not occur - the light will be transmitted rather than internally reflected in the crystal.

#### **3.1 Sample Preparation**

##### **3.1.1 Ink Samples**

Four blue ball point pens were used as the source for ink samples. These four pens are different in 2 brands as follows:

- 045 Reynolds 0.8 mm FINE CARBURE (ink 1)
- Horse Hand Crystal N500 (ink 2)
- Horse-Ball Pen H-501 (ink 3)
- Horse Broad Point 1.0 mm (ink4)

### 3.1.2 Surface Samples

Forty-one surface samples used in this study are described in Table 3.1. All were surfaces usually encountered in daily life, divided into 17 glossy paper surfaces and 24 non-glossy paper surfaces.

**Table 3-1:** Details of the 41 surface samples

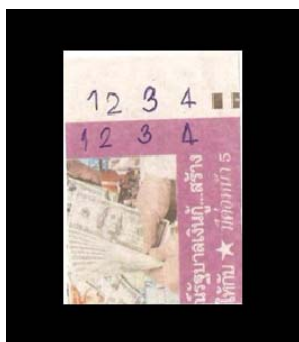
Number	Surfaces Details	Type of surface	
		Glossy	Non-Glossy
S01	Double A A4 paper		/
S02	Hospital hand bill	/	
S03	Food packaging material with foil	/	
S04	Medicine box container	/	
S05	Thai Military Bank deposit slip		/
S06	Thai Military Bank withdrawal slip		/
S07	Supermarket hand bill	/	
S08	Thanachart Bank transaction record		/
S09	Yellow post-it		/
S10	Cash sales bill		/
S11	Bualuang Bank inquiry deposit slip with stamp		/
S12	Printed label on plastic envelope		/
S13	Thairat newspaper separated to be plain and pink color area(S13w,S13p)		/
S14	White plastic envelope	/	
S15	Colorless plastic envelope	/	
S16	Outside of Kraft envelope		/
S17	Inside of Kraft envelope		/
S18	Calendar		/
S19	Cash sale roll paper		/
S20	Bualuang Bank ATM slip		/

**Table 3-1:** Details of the 41 surface samples

Number	Surfaces Details	Type of surface	
		Glossy	Non-Glossy
S21	Back side of BAY Sales flip flap slip customer copy		/
S22	Notebook		/
S23	Gift wrap	/	
S24	Back side of gift wrap		/
S25	Telephone paper book		/
S26	Thai airways boarding pass		/
S27	Coated food packaging material	/	
S28	Back side of Coated food packaging material		/
S29	Color ink jet printing paper	/	
S30	Normal page of the Seventeen magazine	/	
S31	Glossy card paper	/	
S32	Electricity bill (receipt)		/
S33	Cover page of the Seventeen magazine	/	
S34	White plain roll tissue paper		/
S35	Normal page of Honda magazine	/	
S36	Outside cover page of Honda magazine	/	
S37	Inside cover page of Honda magazine	/	
S38	Outside of Mister Donuts box	/	
S39	Inside of Mister Donuts box		/
S40	Color lab photographic paper	/	

### 3.2. Sample Analysis

The numbers 1, 2, 3 and 4 were written on each piece of paper (S01-S41) by using all the 4 blue point pens (Figure 3-1) and left it at least two months before analyzed. The written inks were then analyzed by FT-IR spectroscopy (Hyperion Series ATR FT-IR Microscope, BRUKER). In addition, blank samples were not written on by any pens were also analyzed. After this was completed, ATR-FTIR spectra were examined following by showed on experimental flow chart (Figure 3-2).



**Figure 3-1:** An example of surface sample used in this study. This sample is a piece of newspaper that was divided into 2 surfaces; one is white plain surface and another is colored surface (pink). The numbers 1, 2, 3 and 4 were written by using 045 Reynolds 0.8 mm FINE CARBURE, Horse Hand Crystal N500, Horse-Ball Pen H-501 and Horse Broad Point 1.0 mm, respectively.

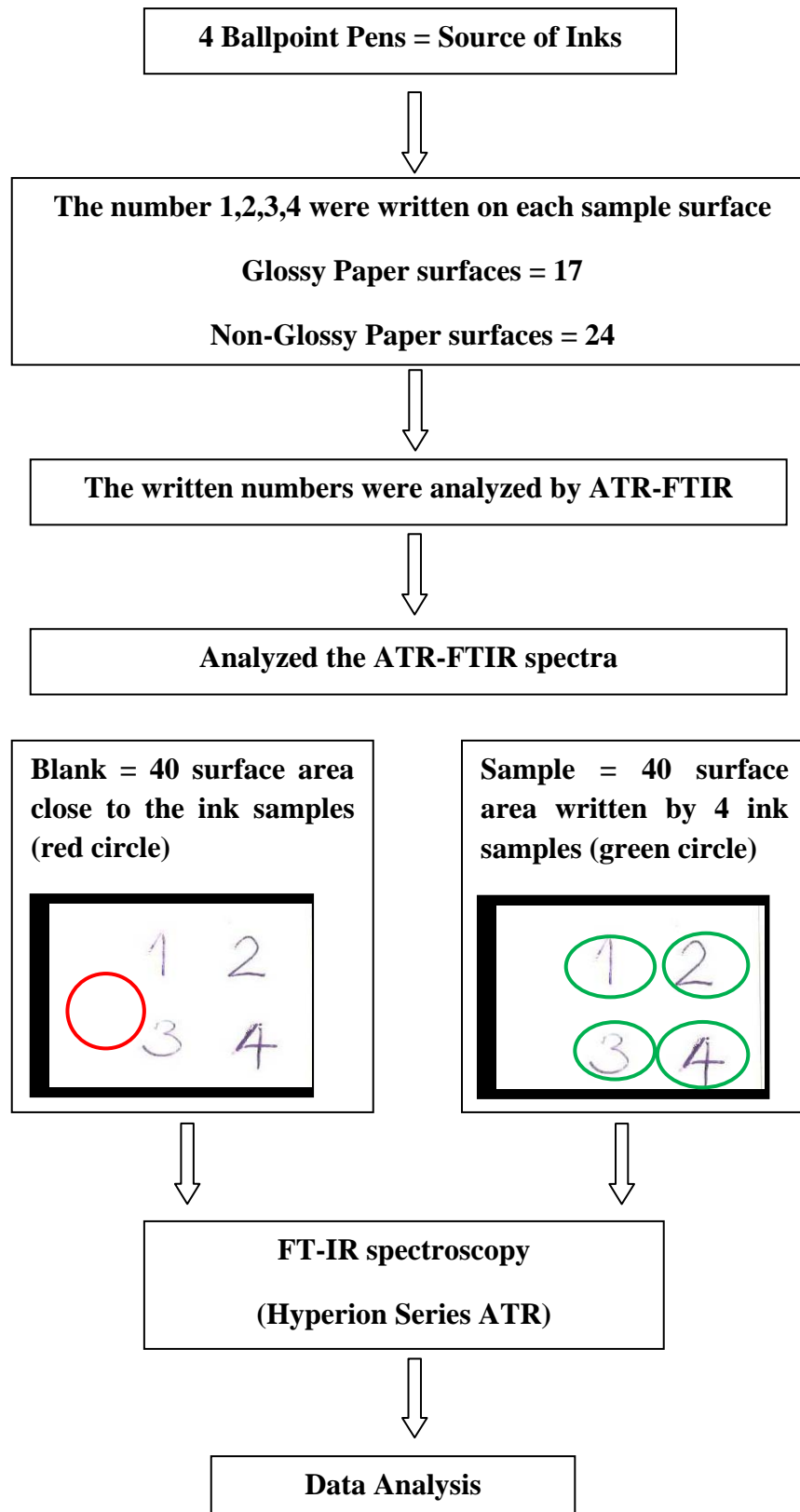


Figure 3-2: Flow chart of experiment.

## **CHAPTER IV**

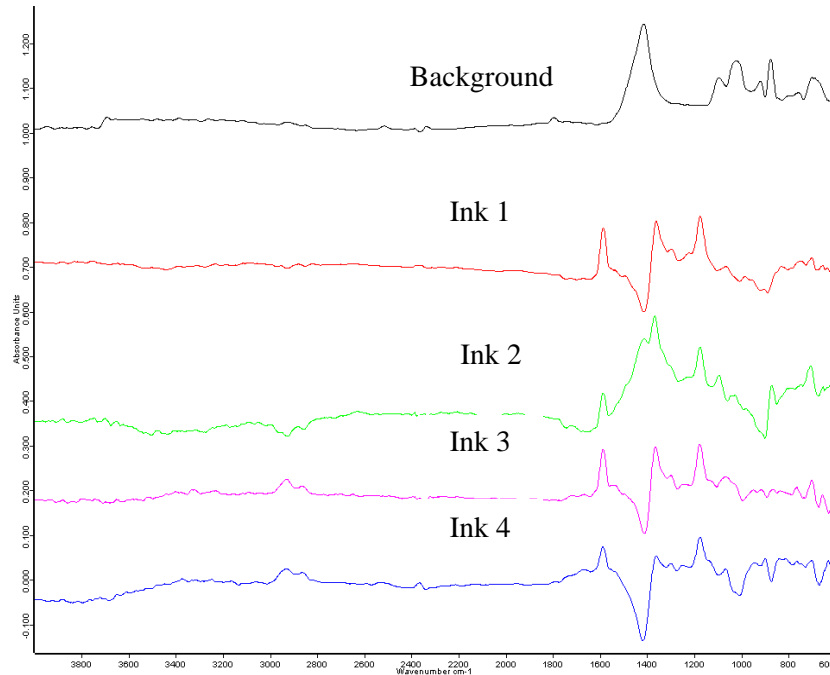
### **RESULTS**

#### **4.1 Inks Spectra on the Same Surface**

ATR-FTIR spectra of all blue ballpoint pen inks written on each sample surfaces.

##### **4.1.1 Group A**

In this group, one of the inks had a spectrum clearly different from the other three. A spectrum will be Ink 1, Ink 2, Ink 3 or Ink 4 depended on the sample surfaces. Examples are given in Figure 4-1 when the sample surface was the inside of a Mister Donut box for which only Ink2 (green line) gave a spectrum different from the other three. For other surfaces the distinguishing spectrum might belong be any one of these inks. The surfaces were food packaging material with foil, medicine box container and gift wrap, yellow post-it, Bualuang Bank ATM slip, telephone paper book and the inside of Mister Donut box.

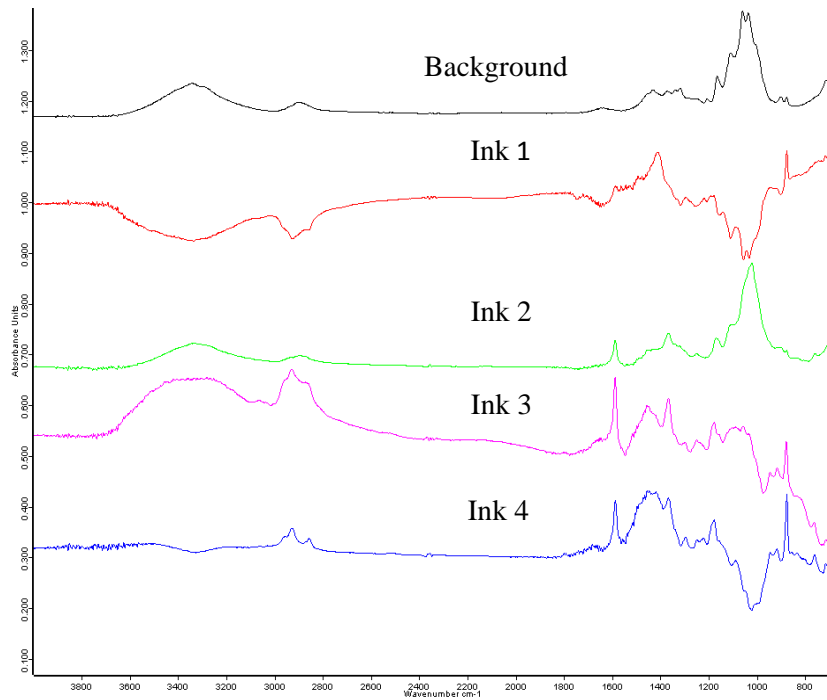


**Figure 4-1:** The ATR-FTIR spectra of inks which written on the inside of Mister Donuts box (Group A).

From seven sample surfaces in this group, three of them were glossy paper (food packaging material with foil, medicine box and gift wrap) other four sample surfaces were non-glossy (yellow post-it, Bualuang Bank ATM slip, Contacts book paper and inside of Mister Donuts box).

#### 4.1.2 Group B

In this group, two inks exhibited spectra of significantly different shapes but the spectra of the other two inks were essentially the same. Figure 4-2 shows examples for inks written on pieces of Double A paper for which Ink1 and Ink2 (red and green spectra) are of different shapes from the other two. The surfaces used were Double A A4 paper, hospital band bill, printed label on plastic envelope, colorless plastic envelope, calendar, cash sale roll paper, normal page of the Seventeen Magazine, glossary card paper, the outside cover page of Honda Magazine, inside cover page of Honda magazine and the outside of Mister Donuts box.



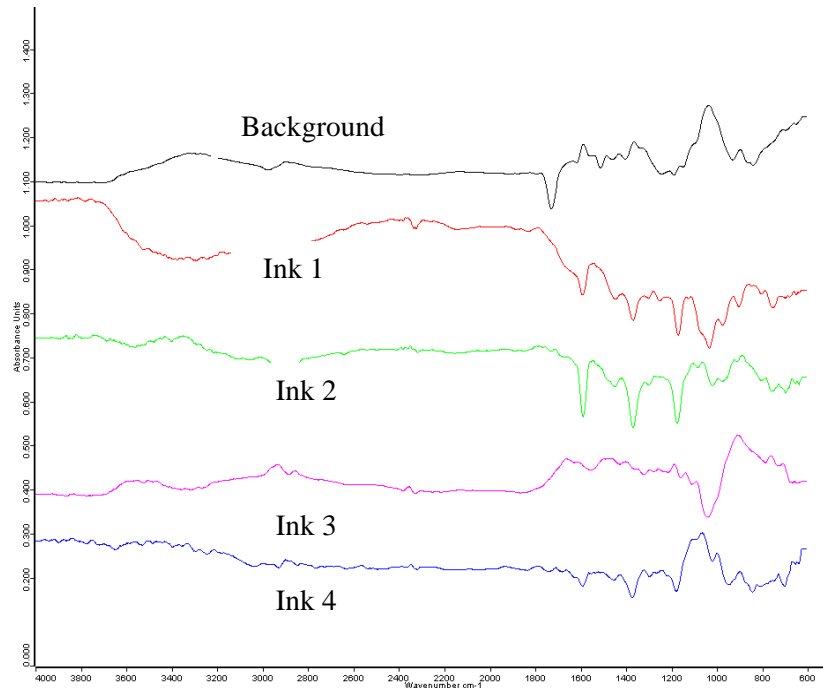
**Figure 4-2:** The ATR-FTIR spectra of inks written on Double A A4 paper (Group B).

ATR-FTIR spectra of two inks are definitely different shapes from the other two which are the same on each the 12 sample surfaces of out the forty-one surfaces. This group includes seven of the seventeen glossy surfaces (hospital band bill, colorless plastic envelope, normal page of the seventeen magazine, glossary card paper, outside cover page of Honda magazine, inside cover page of Honda magazine and outside of Mister Donuts box) and five of the twenty-four non-glossy surfaces (Double A A4 paper, printed label on plastic envelope, calendar, cash sale roll paper and notebook).

#### 4.1.3 Group C

For surfaces in this group, all the four inks exhibited significantly different spectra and the inks were distinguishable. From each other Figure 4-3 gives spectra of inks written on white plain tissue paper. The other sample surfaces in this group were Supermarket hand bill, color ink jet printing paper, normal page of Honda Magazine, color lab photographic paper, Thai Military Bank deposit slip, Thai Military Bank withdrawal slip, Thanachart Bank transaction record, cash sales bill,

Bualuang Bank inquiry deposit slip with stamp, Thairath newspaper plain and pink color areas, electricity bill (receipt) and white plain tissue paper.



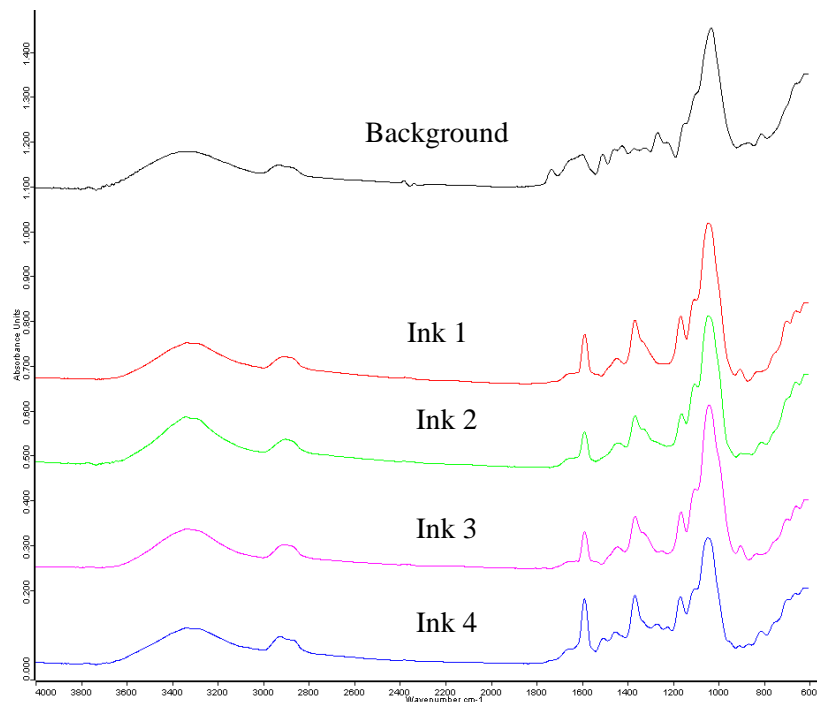
**Figure 4-3:** The ATR-FTIR spectra of inks written on white plain roll tissue paper (Group C).

ATR-FTIR spectra of four inks are significantly different pattern on thirteen of all forty-one sample surfaces. The result showed four of all seventeen glossy paper sample surfaces (Supermarket hand bill, Color ink jet printing paper, Normal page of Honda magazine and Color lab photographic paper) and eight of all twenty-four non-glossy paper sample surfaces (Thai Military Bank deposit slip, Thai Military Bank withdrawal slip, Thanachart Bank transaction record, Cash sales bill, Bualuang Bank inquiry deposit slip with stamp, Thairath newspaper, Electricity bill (receipt) and White plain roll tissue paper).

#### 4.1.4 Group D

In this group, the surfaces used were white plastic envelope, coated food packaging material, cover page of the Seventeen Magazine, the outside of Kraft envelope, inside of Kraft envelope, Back side of gift wrap, Thai airways boarding

pass, the back side of Coated food packaging material and the back side of BAY Sales flip flap slip customer copy. On these surfaces, all the inks gave spectra of essentially the same shape. Figure 4-4 shows examples that none of the spectra could be distinguished from each other on the inside of Kraft envelope.



**Figure 4-4:** The ATR-FTIR spectra of inks written on the inside of Kraft envelope (Group D).

From nine of forty-one samples, three samples are glossy paper that was 17.66% of all seventeen glossy samples (White plastic envelope, coated food packaging material and Cover page of the Seventeen magazines). Others 6 samples are non glossy sample surfaces that is 25% of all twenty-four non glossy paper sample surfaces (Outside of Kraft envelope, Inside of Kraft envelope, Back side of gift wrap, Thai airways boarding pass, Back side of Coated food packaging material and Back side of BAY Sales flip flap slip customer copy).

Table 4-1 showed the number in each sample group: Group A consisted of three glossy sample surfaces and four non-glossy sample surfaces, Group B comprised seven glossy sample surfaces and five non-glossy sample surfaces, Group C contented

four glossy sample surfaces and nine non-glossy sample surface and Group D was composed of three glossy sample surfaces and six non-glossy sample surfaces, respectively.

**Table 4-1** showed the number in each sample group.

<b>Differentiated Spectra</b>	<b>Number</b>		
	<b>Glossy Sample Surface</b>	<b>Non-Glossy Sample Surface</b>	<b>Total</b>
<b>Group A</b>	3	4	7
<b>Group B</b>	7	5	12
<b>Group C</b>	4	9	12
<b>Group D</b>	3	6	9
<b>Total</b>	<b>17</b>	<b>24</b>	<b>41</b>

## 4.2 Inks Spectra on the Different Surfaces

The ATR-FTIR spectra of blue ballpoint pen inks on the different surfaces will be described.

### 4.2.1 Ink 1 Group

This group represents surfaces on which ink 1 gave spectra of the same shape. There are:

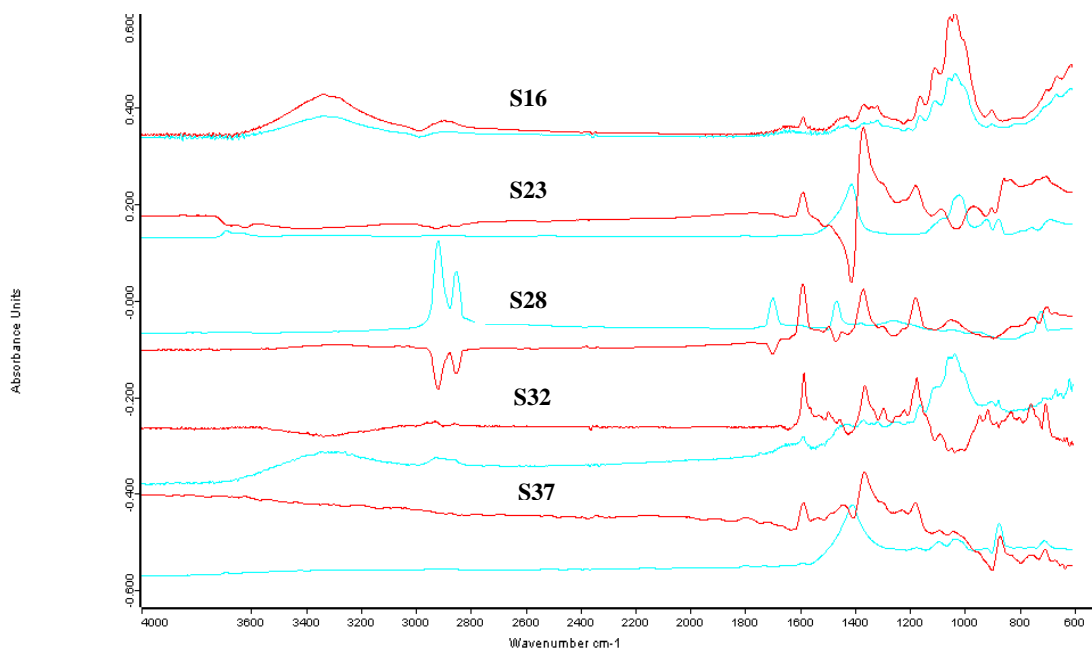
#### *Glossy Surfaces*

- Hospital band bill
- Medicine box
- White plastic envelope
- Glossy card paper
- Cover page of the Seventeen magazine
- Color lab photographic paper
- Food packaging material with foil
- Colorless plastic envelope
- Gift wrap
- Coated food packaging material
- Normal page of the Seventeen magazine
- Inside cover page of Honda magazine

*Non-Glossy Surfaces*

- Thanachart Bank transaction record
- Bualuang Bank inquiry deposit slip
- Outside of Kraft envelope
- Cash sale roll paper
- Bualuang Bank ATM slip
- Back side of BAY Sales flip flap slip customer copy
- Notebook
- Calendar
- Inside of Mister Donuts box
- Thai Military Bank deposit slip
- Yellow post-it
- Thairath newspaper plain color area
- Inside of Kraft envelope
- Back side of gift wrap
- Back side of Coated food packaging material
- Thai airways boarding pass
- Contacts book paper
- Electricity bill (receipt)

Ink 1 written on the 31 surfaces gave spectra which were indistinguishable. (Figure 4-5).



**Figure 4-5:** The ATR-FTIR spectra of ink 1 which were written on the outside of Kraft envelope(S16), gift wrap(S23), back side of Coated food packaging material (S28), electricity bill (S32), inside cover page of Honda magazine (S37) with background spectra(light blue line).

### 4.2.2 Ink 2 Group

This group represents surfaces on which ink 2 gave spectra of the same shape. The surfaces include:

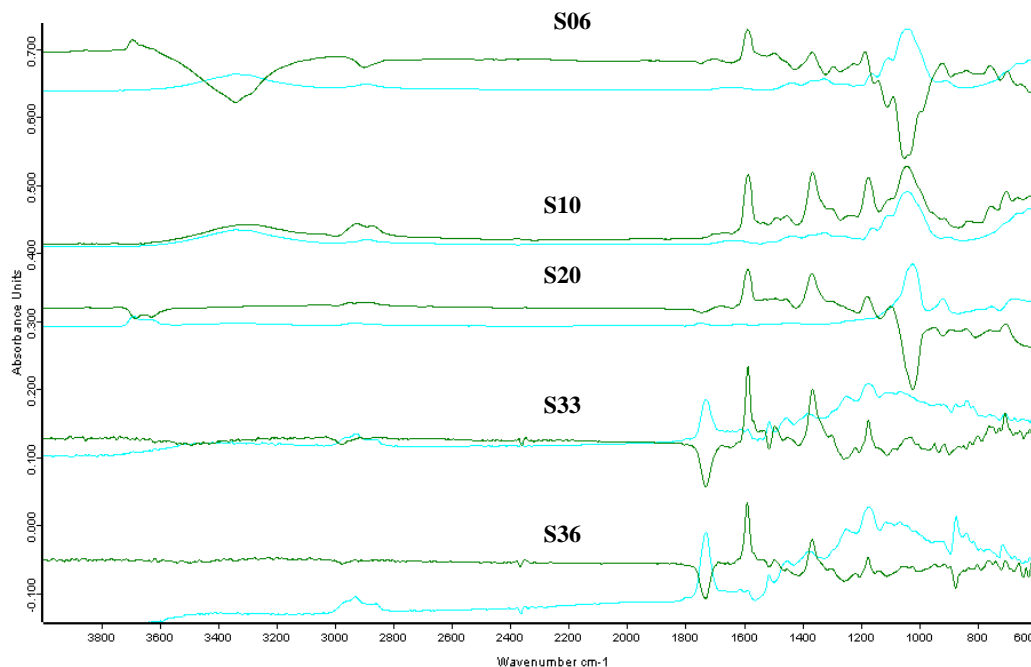
#### *Glossy Surfaces*

- Hospital band bill
- Food packaging material with foil
- White plastic envelope
- Cover page of the Seventeen magazine
- Outside cover page of Honda magazine
- Outside of Mister Donuts container box
- Medicine box
- Supermarket hand bill
- Gift wrap
- Coated food packaging material
- Colorless plastic envelope
- Normal page of Honda magazine

#### *Non-Glossy Surfaces*

- Thanachart Bank transaction record
- Thai Military Bank deposit slip
- Thai Military Bank withdrawal slip
- Outside of Kraft envelope
- Cash sale roll paper
- Bualuang Bank ATM slip
- Back side of BAY Sales flip flap slip customer copy
- Notebook
- Calendar
- Double A A4 paper
- Yellow post-it
- Cash sale bill
- Inside of Kraft envelope
- Back side of gift wrap
- Thai airways boarding pass
- Back side of Coated food packaging material
- Thai airways boarding pass
- Contacts book paper
- Inside of Mister Donuts container box

**Figure 4-6** shows ATR-FTIR spectra of ink 2 on some surfaces in this group.



**Figure 4-6:** The ATR-FTIR spectra of ink2 which were written on Thai Military Bank withdrawal slip (S06), Cash sales bill (S10), Bualuang Bank ATM slip (S20), Cover page of the Seventeen magazine (S33), Outside cover page of Honda magazine (S36) with each background spectrum (light blue line).

### 4.2.3 Ink 3 Group

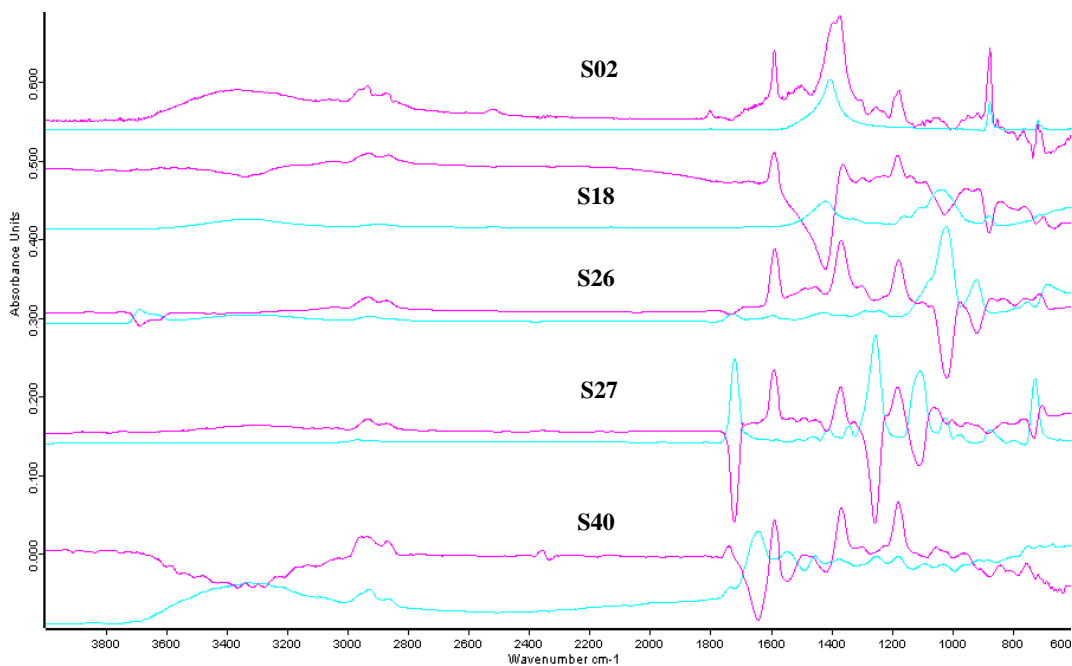
Included in this group are surfaces on which ink 3 gave spectra of the same shape. They are:

#### *Glossy Surfaces*

- Hospital band bill
- Food packaging material with foil
- White plastic envelope
- Cover page of the Seventeen magazine
- Outside cover page of Honda magazine
- Outside of Mister Donuts container box
- Medicine box
- Supermarket hand bill
- Gift wrap
- Coated food packaging material
- Colorless plastic envelope
- Normal page of Honda magazine

*Non-Glossy Surfaces*

- Thanachart Bank transaction record
- Thai Military Bank deposit slip
- Thai Military Bank withdrawal slip
- Outside of Kraft envelope
- Cash sale roll paper
- Bualuang Bank ATM slip
- Back side of BAY Sales flip flap slip customer copy
- Notebook
- Calendar
- Double A A4 paper
- Yellow post-it
- Cash sale bill
- Inside of Kraft envelope
- Back side of gift wrap
- Thai airways boarding pass
- Back side of Coated food packaging material
- Thai airways boarding pass
- Contacts book paper
- Inside of Mister Donuts box



**Figure 4-7:** The ATR-FTIR spectra of ink3 which were written on Hospital hand bill (S02), Calendar (S18), Thai airways boarding pass (S26), Coated food packaging material (S27), Color lab photographic paper (S40) with each background spectrum , light blue line.

**Figure 4-7** shows ATR-FTIR spectra of some surfaces in this group.

#### 4.2.4 Ink 4 Group

Included in this group are surfaces on which ink 4 exhibited ATR-FTIR spectra of essentially the same shape.

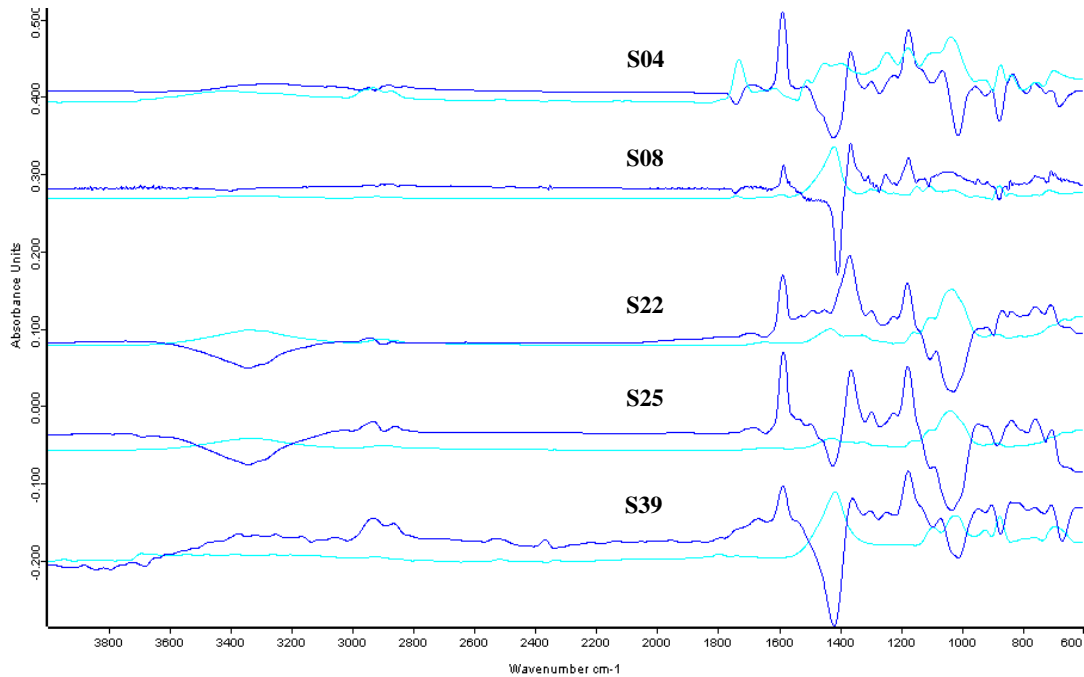
##### *Glossy Surfaces*

- Hospital band bill
- Food packaging material with foil
- White plastic envelope
- Cover page of the Seventeen magazine
- Normal page of the Seventeen magazine
- Glossy card paper
- Outside cover page of Honda magazine
- Medicine box container
- Supermarket hand bill
- Gift wrap
- Coated food packaging material
- Normal page of Honda magazine
- Outside of Mister Donuts container box
- Color ink jet printing paper
- Color lab photographic paper

##### *Non-Glossy Surfaces*

- Printed label on plastic envelope
- Thai Military Bank deposit slip
- Thairath newspaper pink color area
- Outside of Kraft envelope
- Cash sale roll paper
- Bualuang Bank ATM slip
- Bualuang Bank inquiry deposit slip with stamp
- Back side of BAY Sales flip flap slip customer copy
- Calendar
- Inside of Mister Donuts container box
- Thai Military Bank withdrawal slip
- Notebook
- Yellow post-it
- Thanachart Bank transaction record
- Inside of Kraft envelope
- Back side of gift wrap
- Thai airways boarding pass
- Back side of Coated food packaging material
- Contacts book paper
- Thairath newspaper plain color area
- Electricity bill (receipt)

Figure 4-8 shows ATR-FTIR spectra of ink 4 on some surfaces. Moreover, Table 4-3 shows the number of ink 1, ink 2, ink 3 and ink 4 group which were written on the different surfaces.



**Figure 4-8:** The ATR-FTIR spectra of ink4 which were written on Hospital hand bill (S04), Calendar (S08), Thai airways boarding pass (S22), Coated food packaging material (S25), Color lab photographic paper (S39) with each background spectrum (light blue line).

**Table 4-2** showed the number of each ink group on the different surfaces.

Source of Ink	Glossy Sample Surface	Non-Glossy Sample Surface	Total
Ink 1	12/17	18/24	30/41
Ink 2	12/17	18/24	30/41
Ink 3	11/17	20/24	31/41
Ink 4	15/17	21/24	36/41

## **CHAPTER V**

### **DISCUSSION AND CONCLUSION**

#### **5.1 The ATR-FTIR spectra of 11 blue ballpoint pen inks that were written on the same surfaces.**

In this study, four ink samples written on forty-one of glossy and non-glossy sample surfaces were examined by ATR-FTIR spectroscopy. The resulting spectra can be divided into four groups depending on the spectral pattern of each ink sample. The result in Table 4-1 suggests that for glossy sample surfaces, two types of ink can be clearly differentiated while the ATR-FTIR spectra of all ink samples on non-glossy sample surfaces could all also show that there are 9 sample surfaces on which the 4 ink samples have the same ATR-FTIR spectra are the same. The result suggests for most surfaces (~78%), at least one type of ink from the others. This study indicates that the ATR-FTIR technique can be applied to both glossy and non-glossy paper (Table 4-1) and different surfaces.

#### **5.2 The ATR-FTIR spectra of each type of blue ballpoint pen ink that was written on the different surfaces.**

The result in Table 4-2 suggests that the spectra pattern of ink 4 were totally matched on thirty-six from forty-one sample surfaces, included fifteen from seventeen and twenty-one from twenty-four glossy and non-glossy sample surfaces. On the other hand, the best average results were obtained for each ink on non-glossy surfaces (~80%).

The coating layer of the glossy surface may have affected the analysis and caused this finding (Lindblom, 2006). From the result, it can be concluded that the ATR-FTIR technique can be used to analyze fraudulent documents written in blue ballpoint pens on the same surface and the different surfaces.

### **5.3 Conclusion**

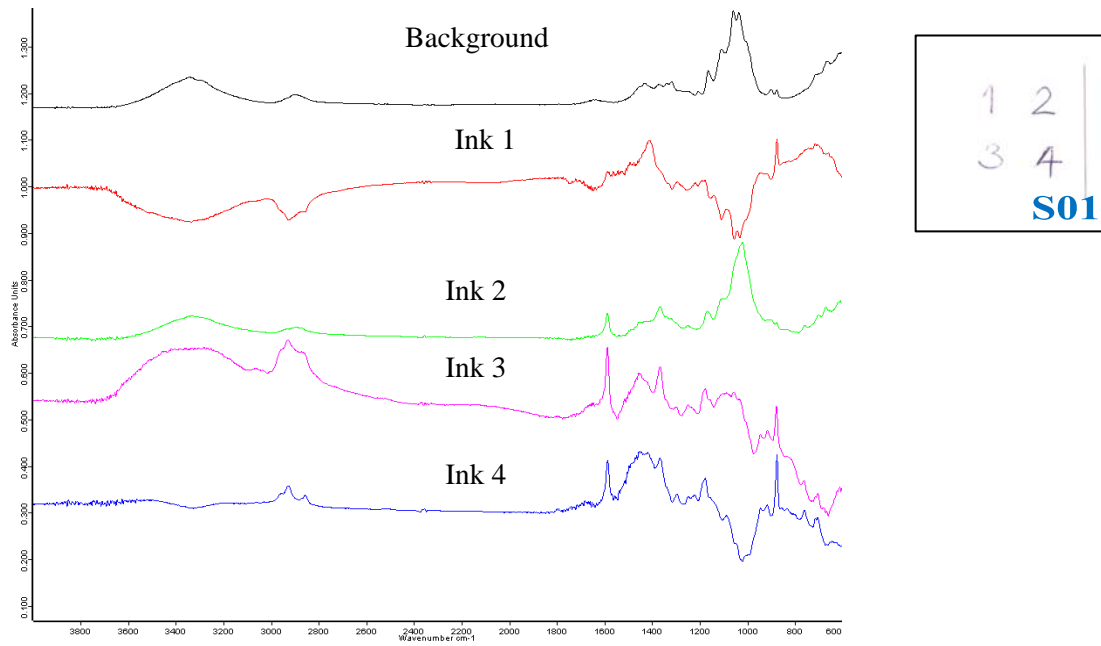
It is clear that paper surfaces do affect FTIR spectra of inks although it is not yet clear why. Document examiners have to be careful before giving their opinions whether a document is forged. It may be impossible at this stage to judge this solely from FTIR spectra. However, one can at least place an ink in a group with similar characteristics.

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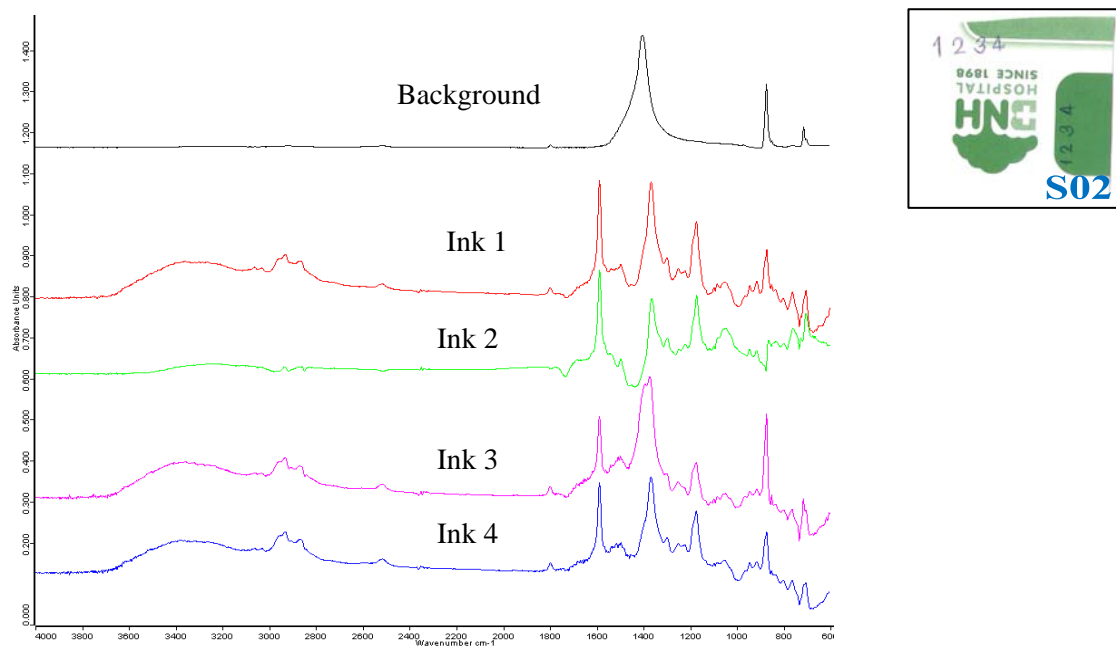
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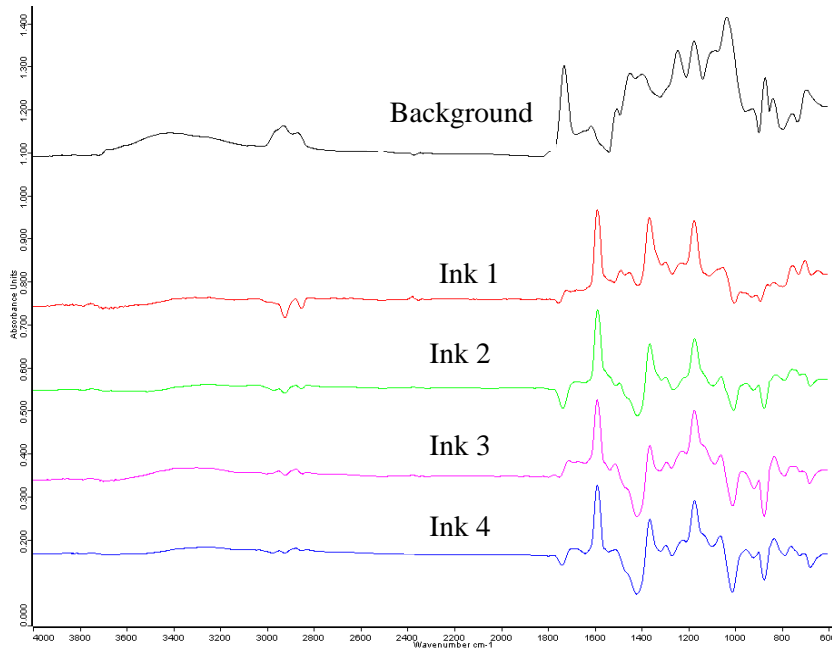
## **APPENDIX**



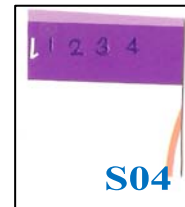
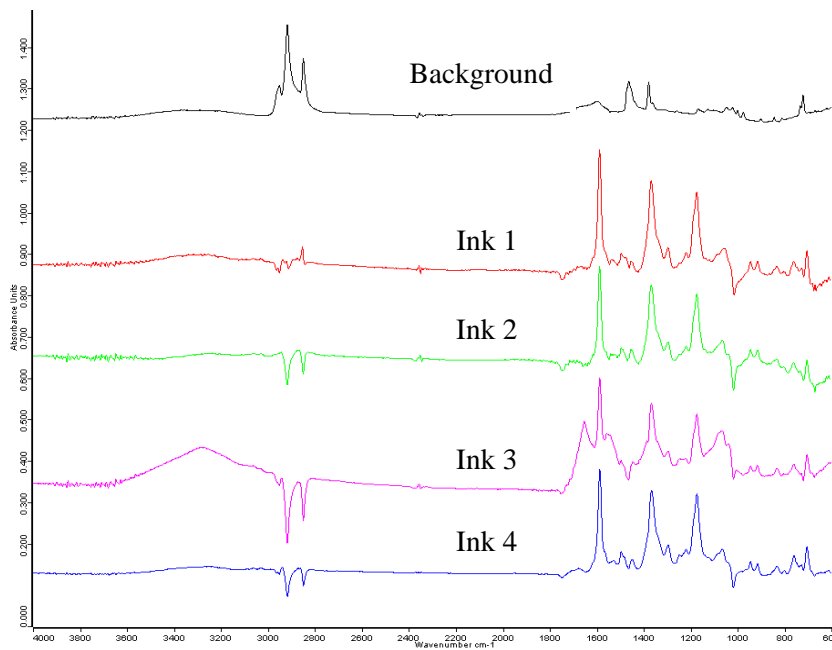
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Double A A4 paper (S01).



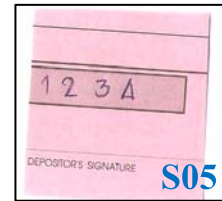
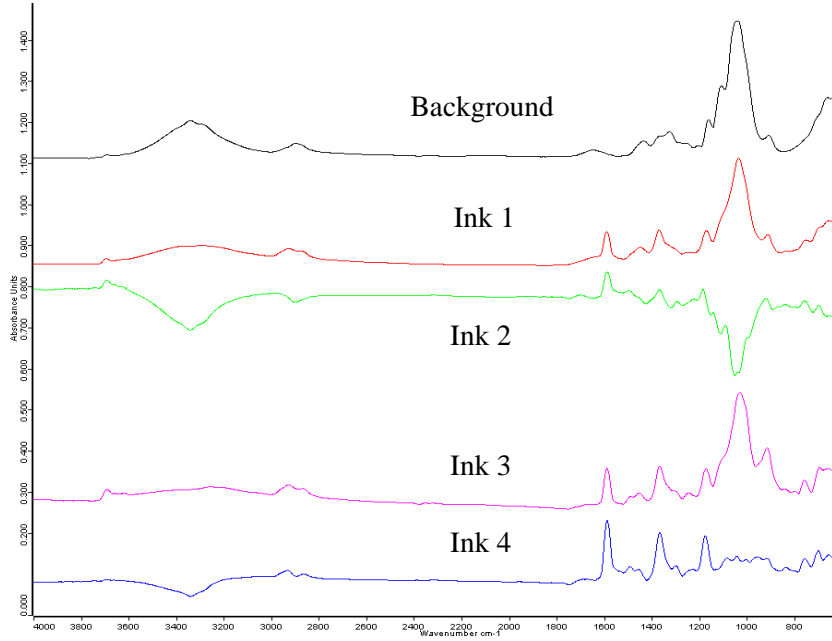
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Hospital hand bill (S02).



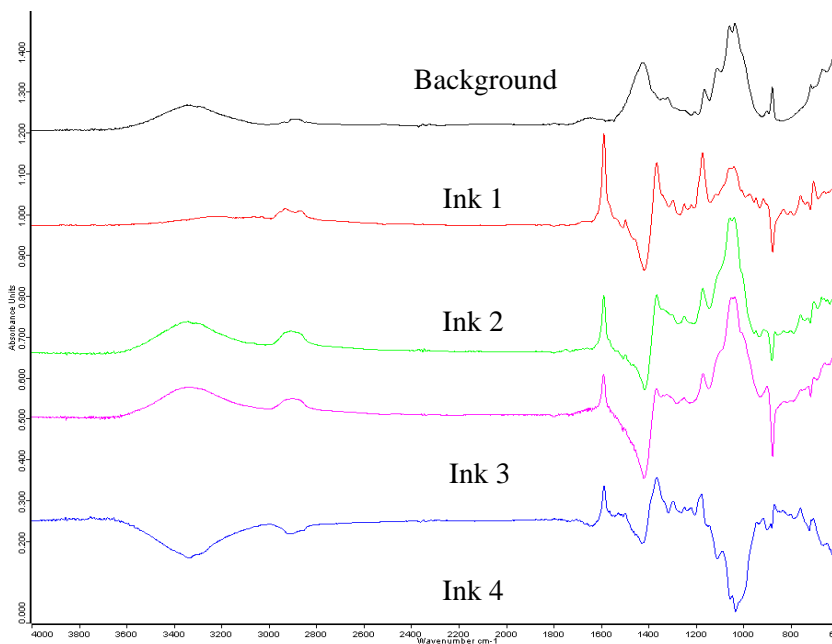
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Food packaging material with foil (S03).



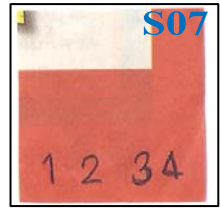
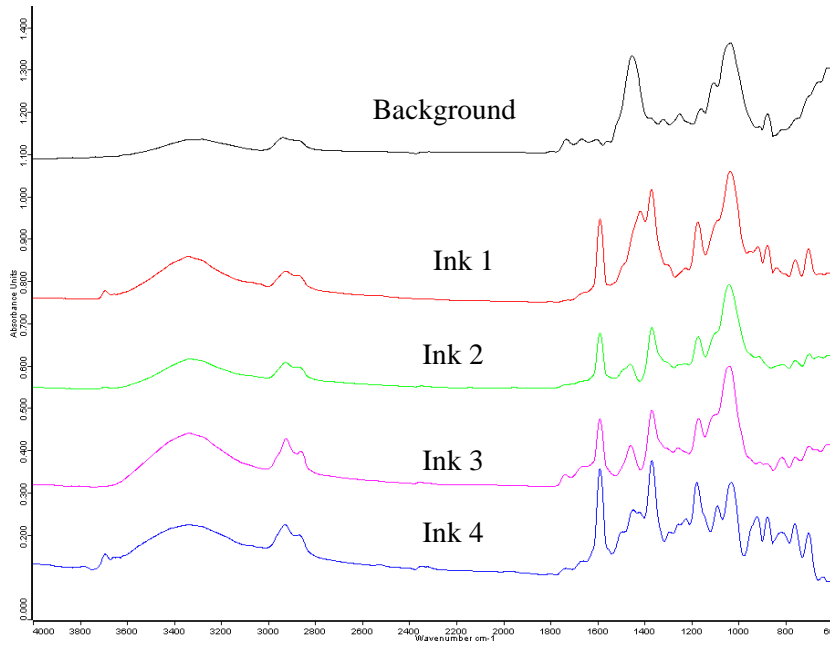
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Medicine box container (S04).



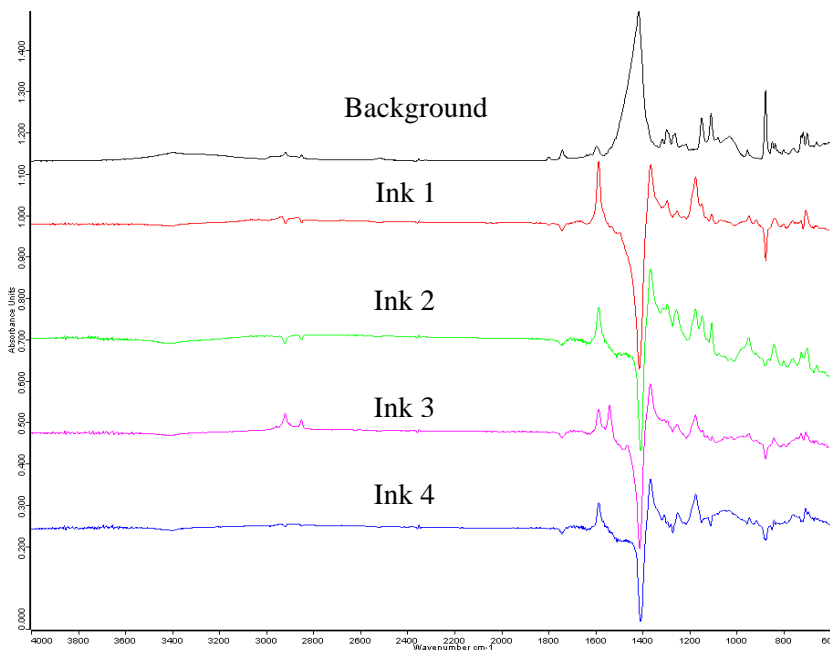
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Thai Military Bank deposit slip (S05).



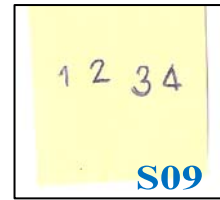
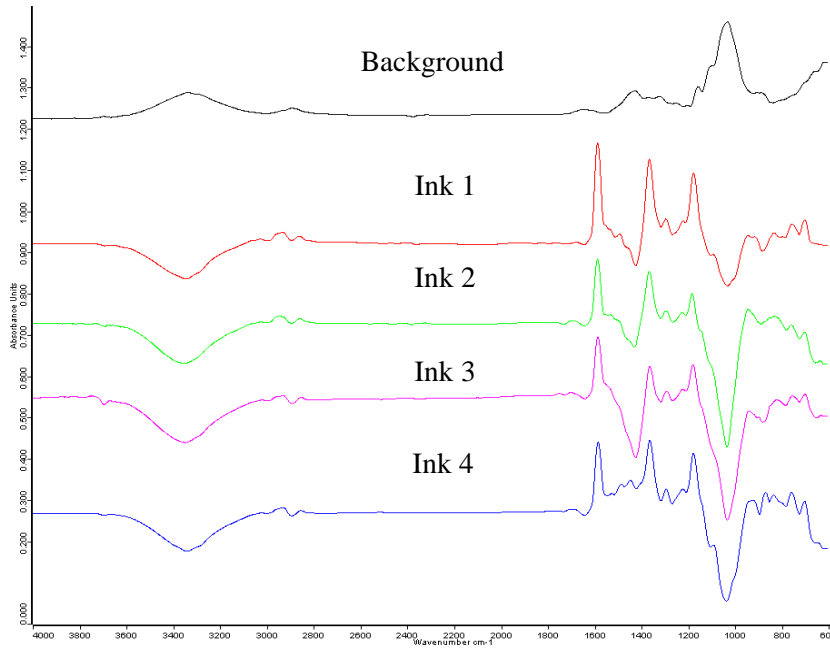
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Thai Military Bank withdrawal slip (S06).



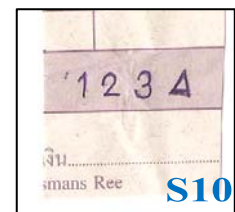
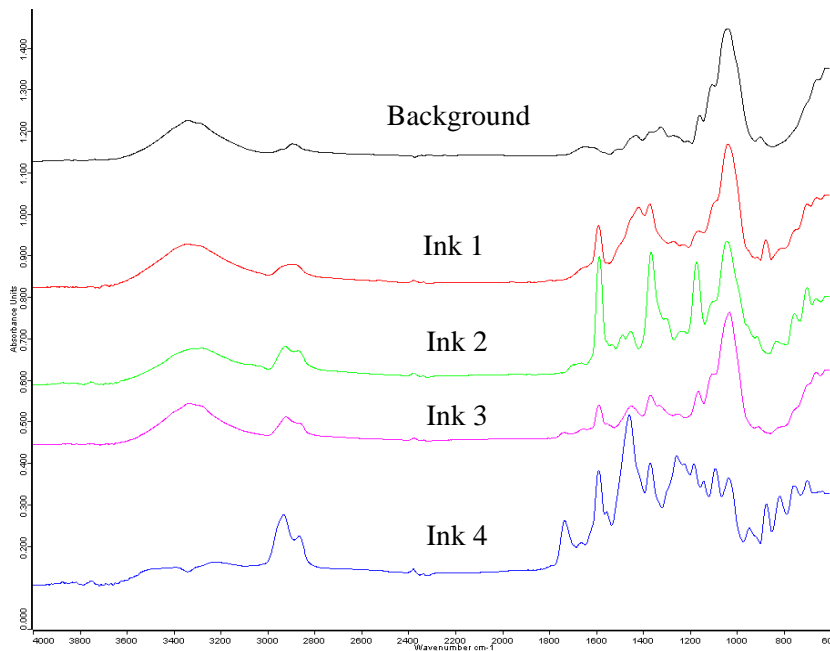
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Supermarket hand bill (S07).



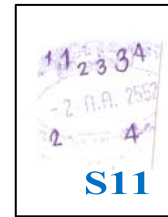
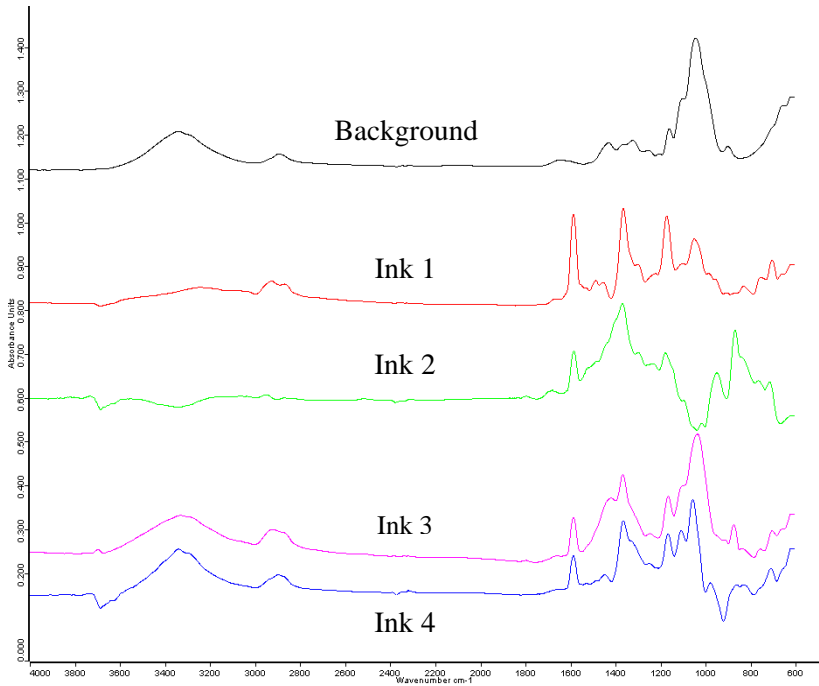
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Thanachart Bank transaction record (S08).



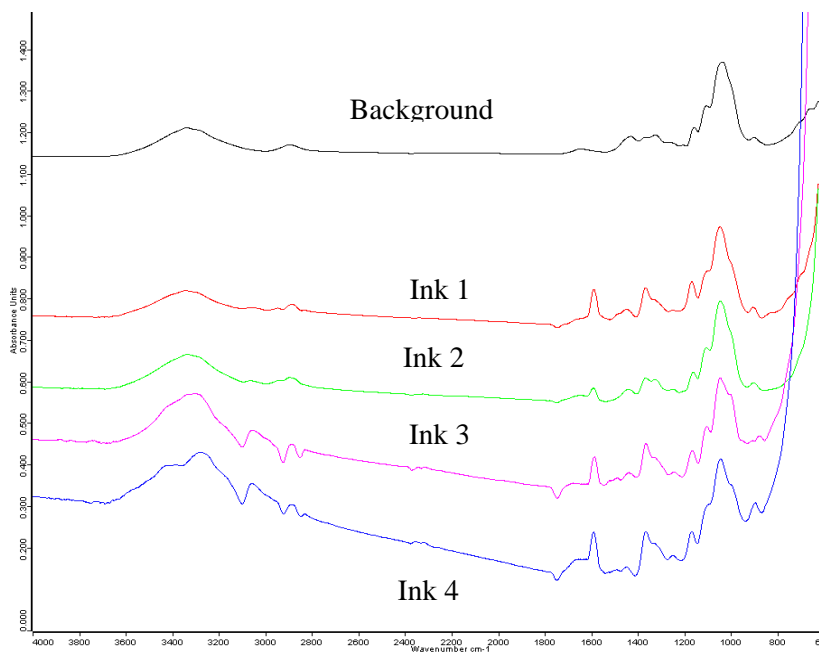
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Yellow Post-it (S09)



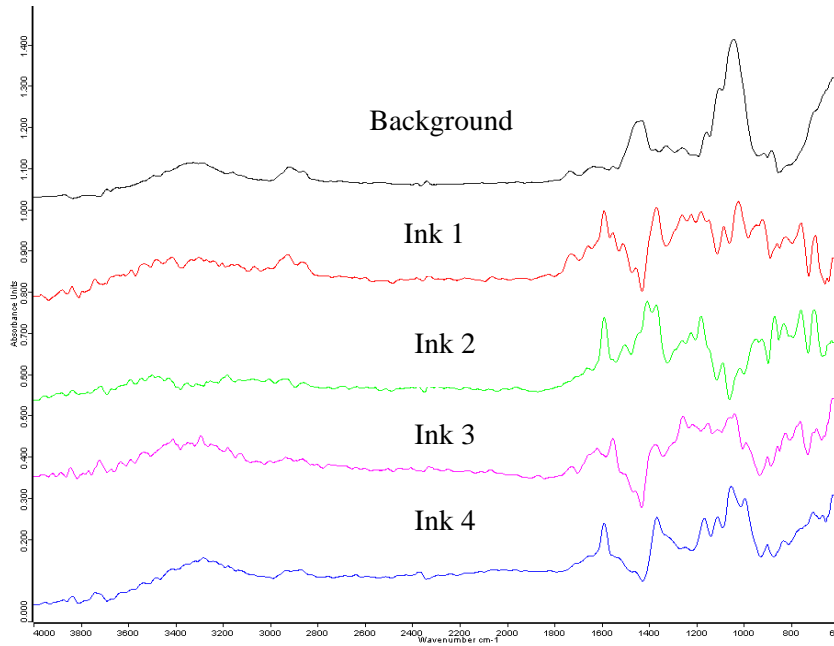
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Cash sales bill (S10)



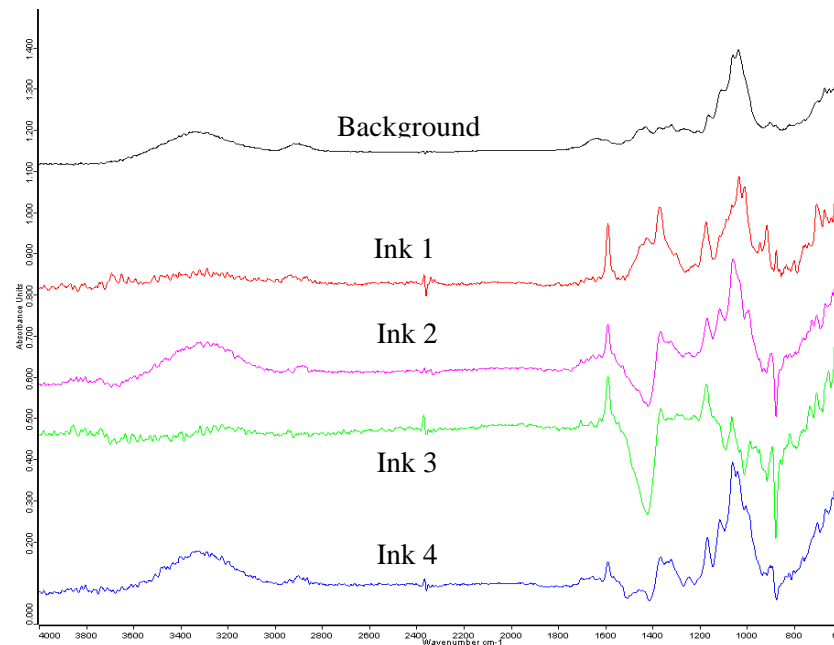
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Bualuang Bank inquiry deposit slip with stamp (S11).



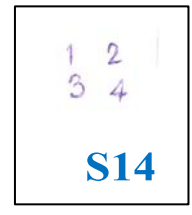
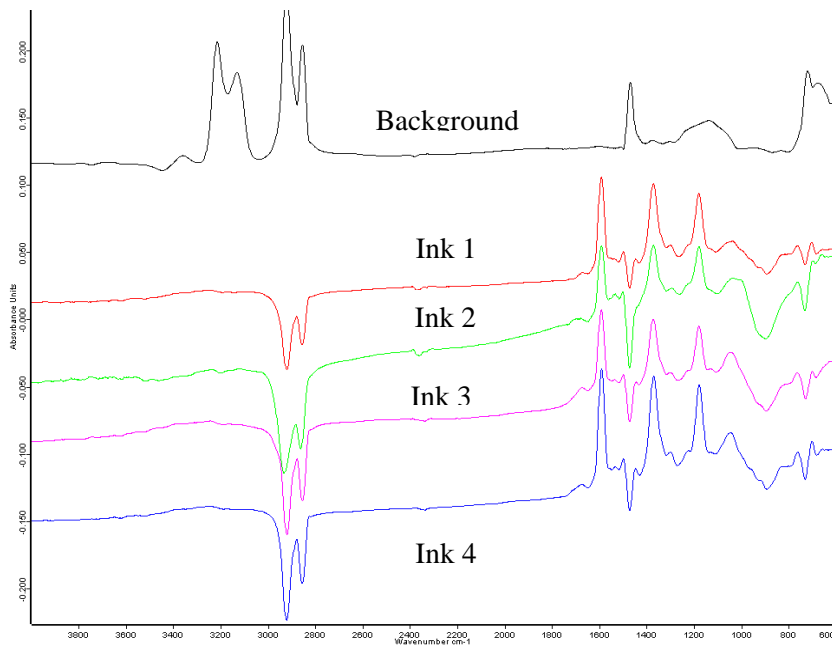
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Printed label on Plastic Envelope (S12)



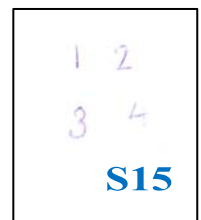
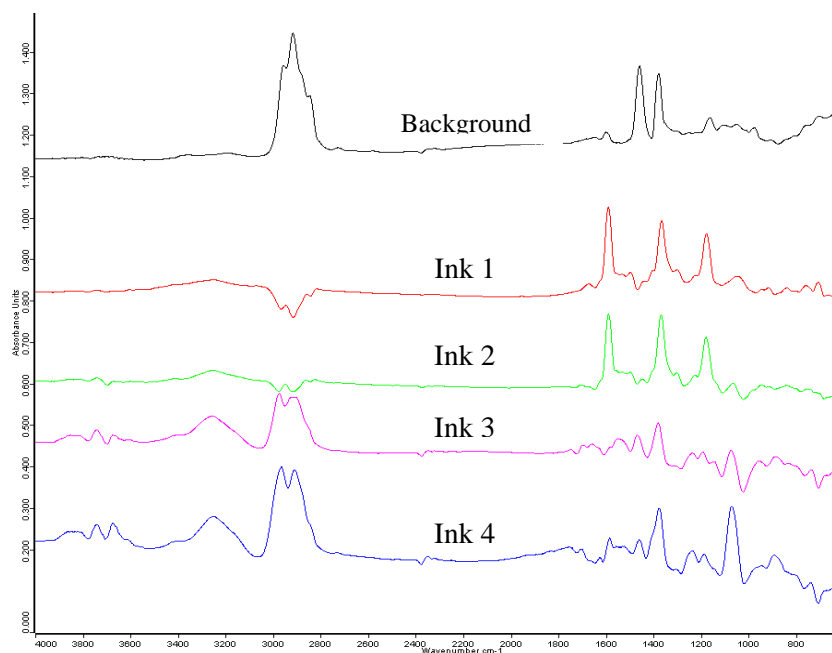
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Thairat Newspaper Plain color area (S13W)



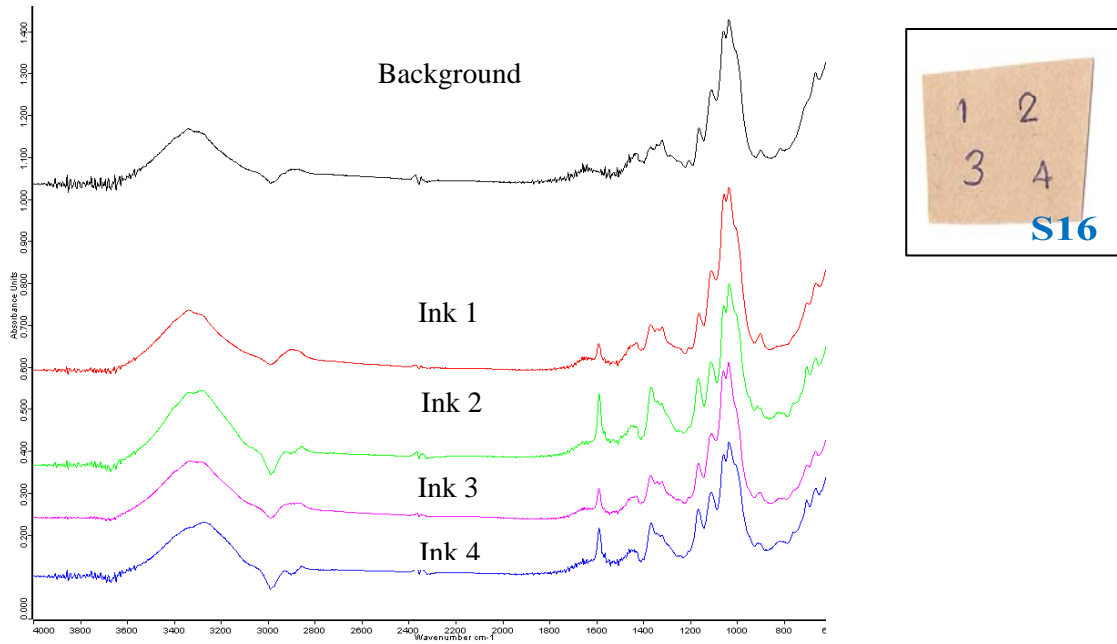
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Thairat newspaper pink color area (S13P)



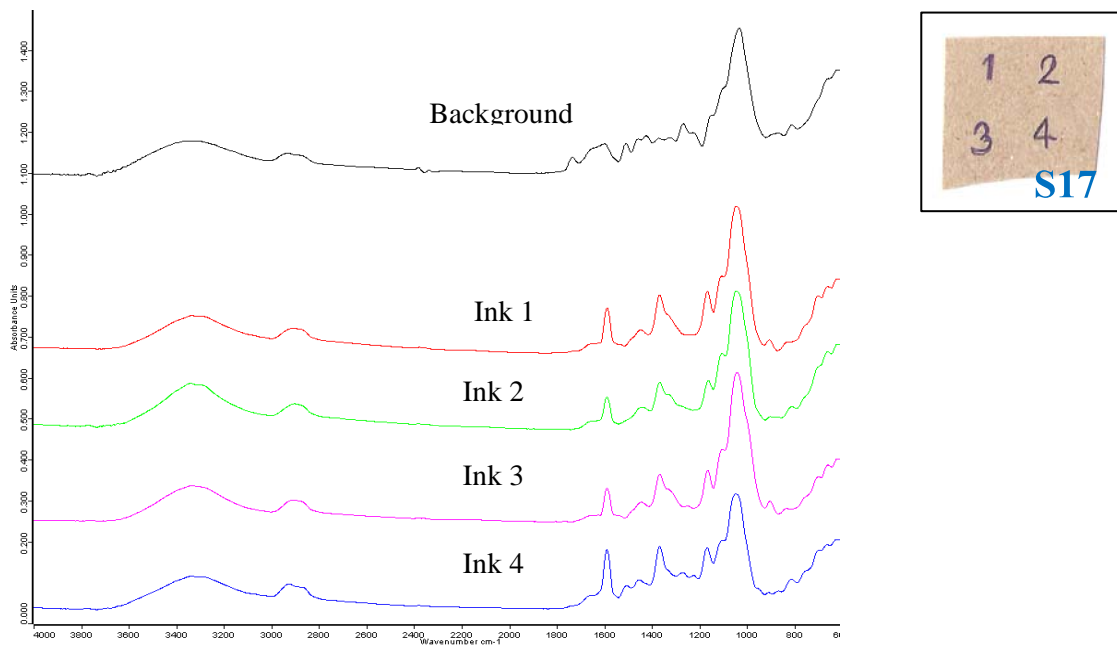
ATR-FTIR spectra of ink 1, 2, 3 and 4 on White plastic envelope (S14)



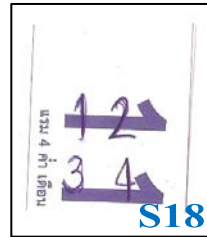
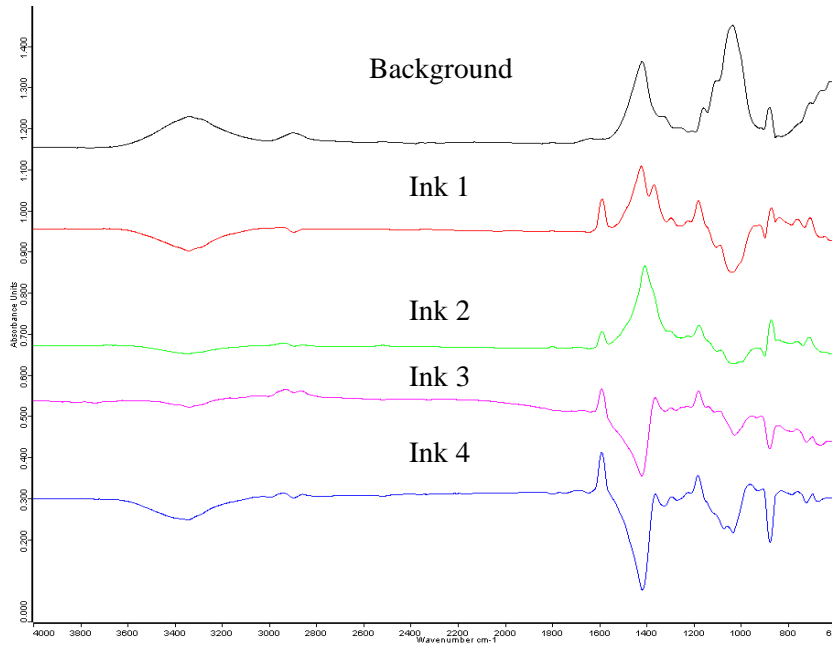
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Colorless plastic envelope (S15)



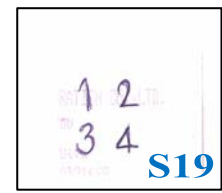
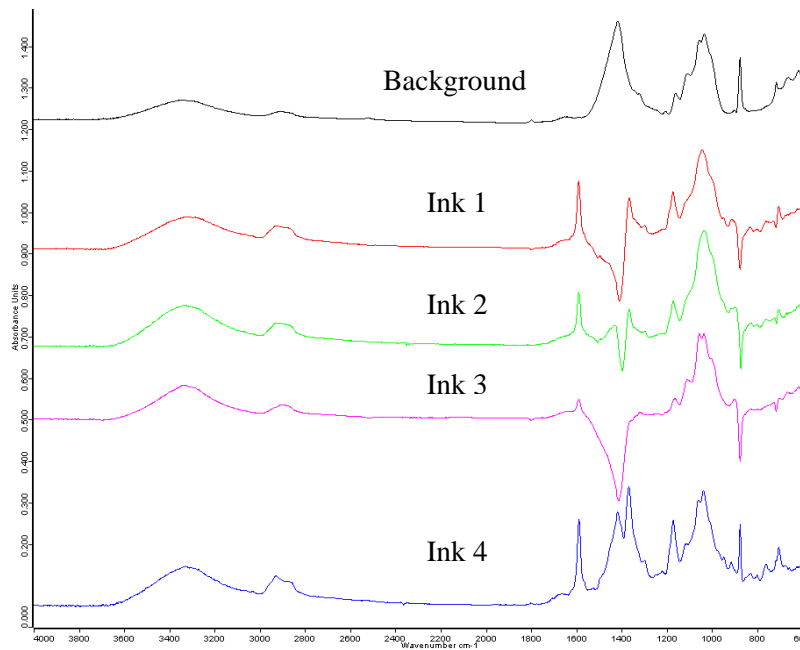
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Outside of Kraft envelope (S16)



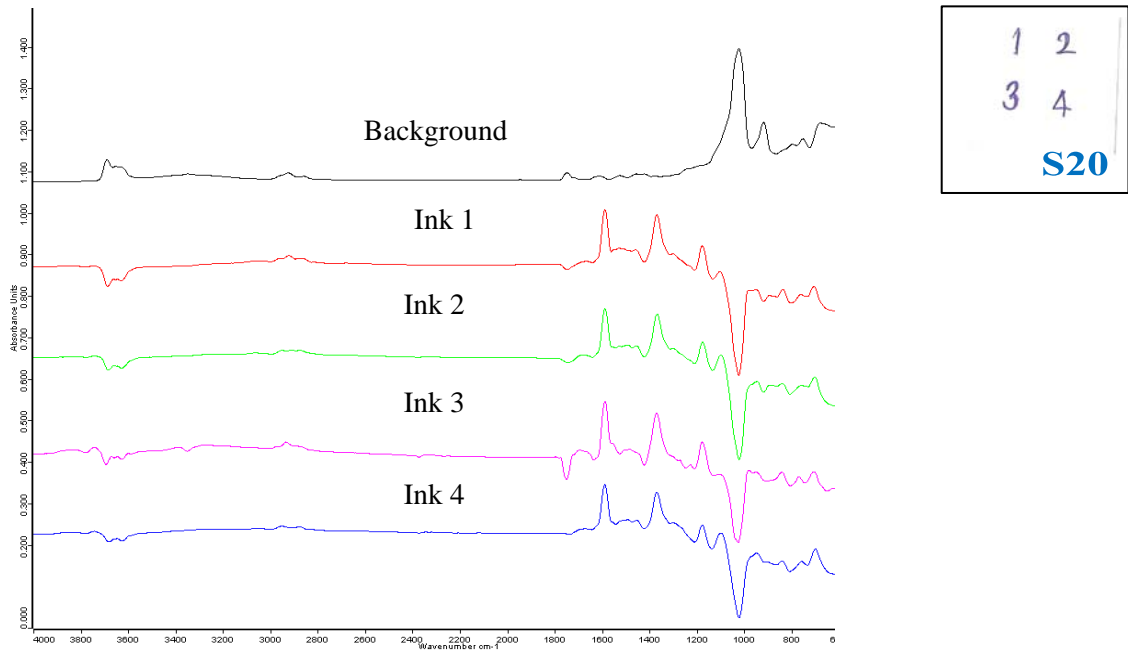
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Inside of Kraft envelope (S17)



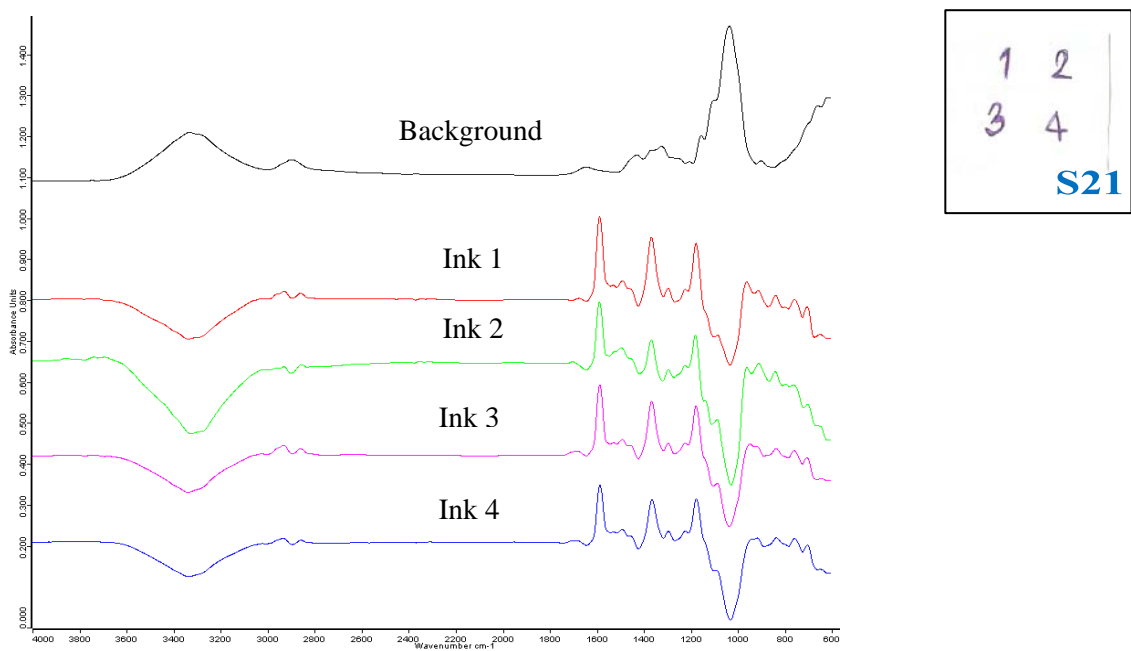
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Calendar (S18)



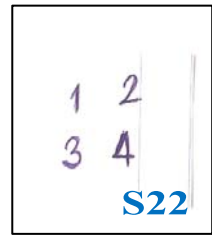
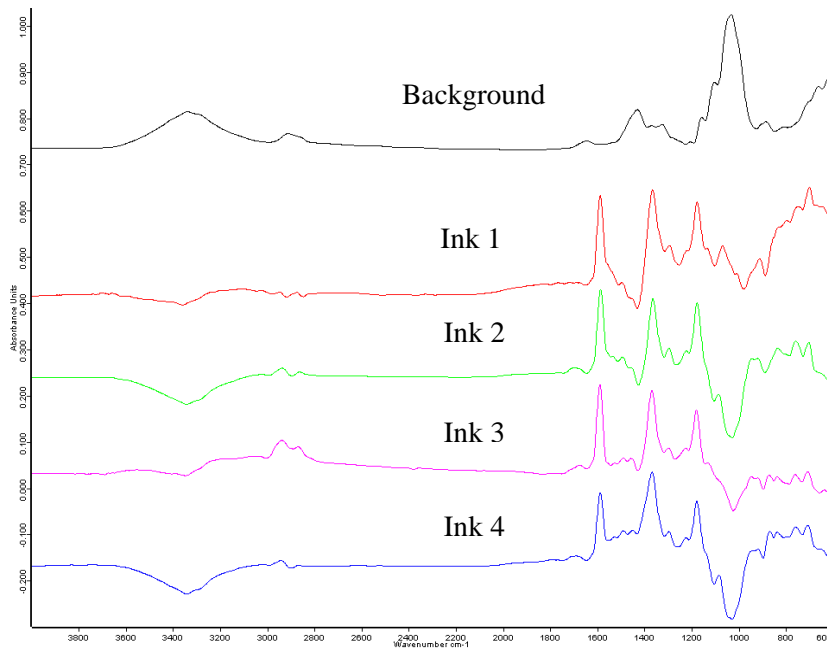
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Cash sale roll paper (S19).



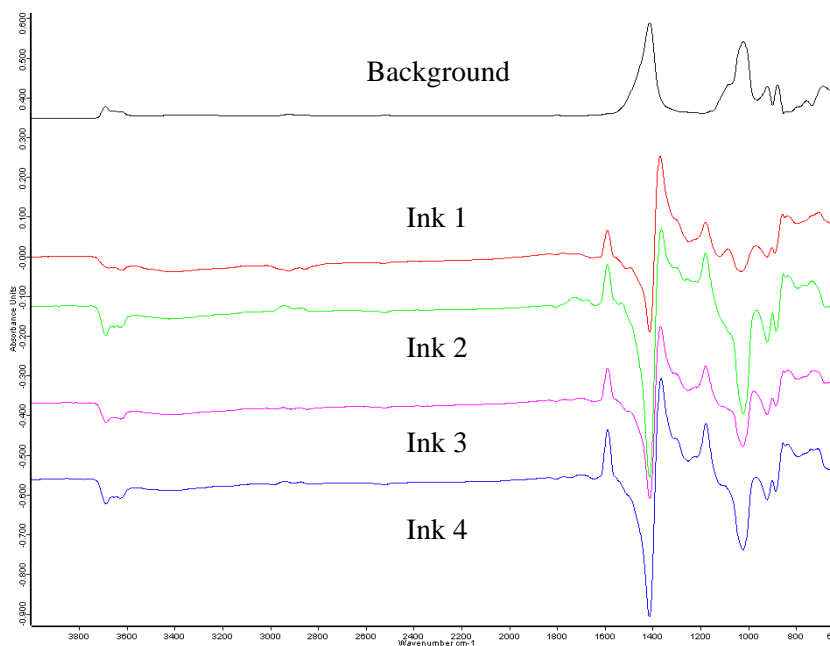
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Bualuang Bank ATM slip (S20).



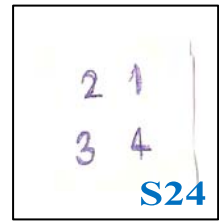
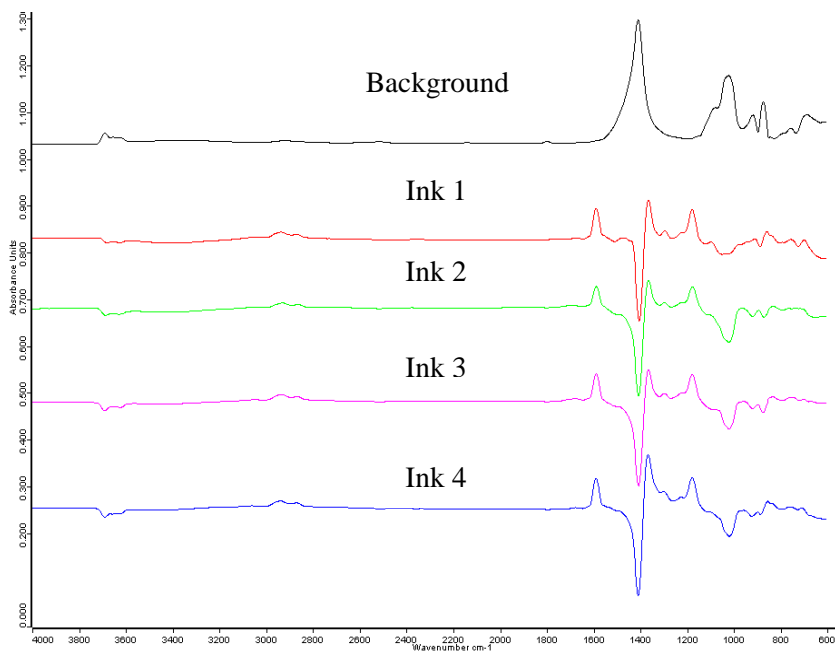
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Back side of BAY Sales flip flap slip customer copy (S21).



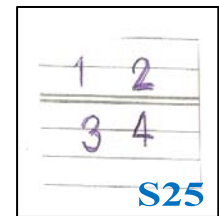
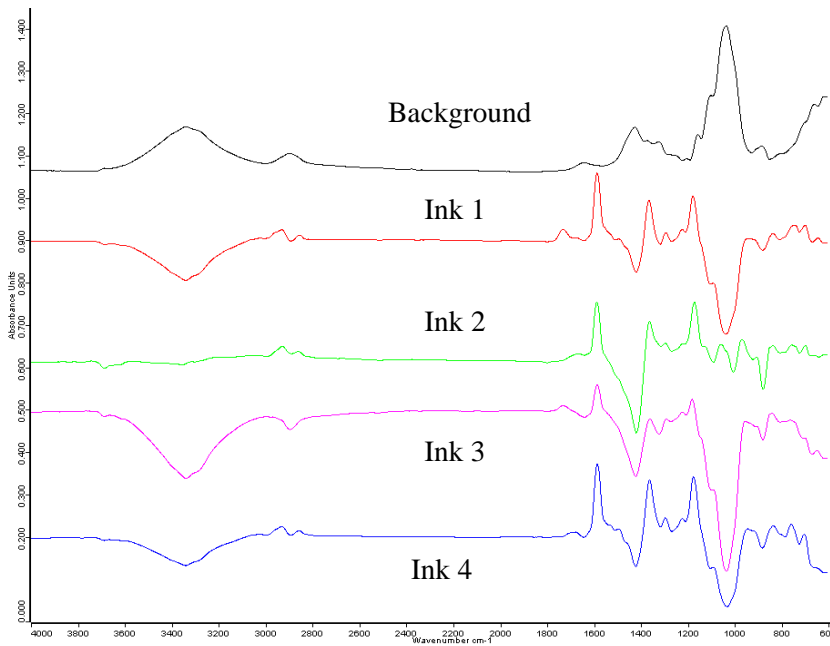
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Notebook (S22).



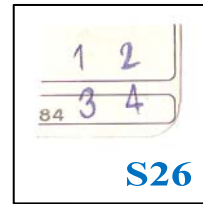
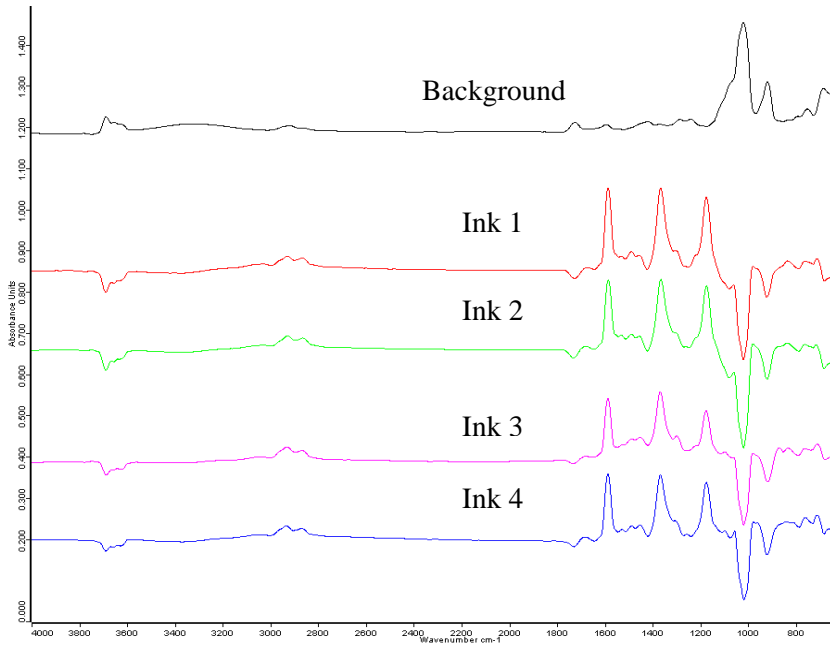
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Gift wrap (S23).



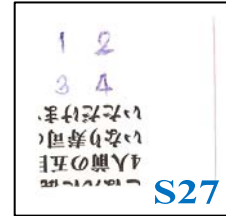
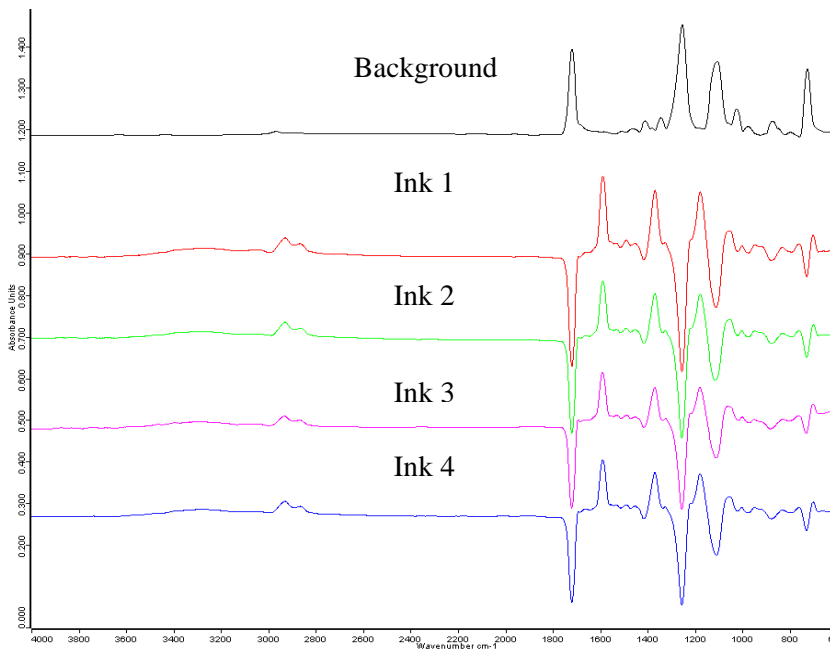
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Back side of gift wrap (S24).



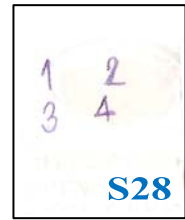
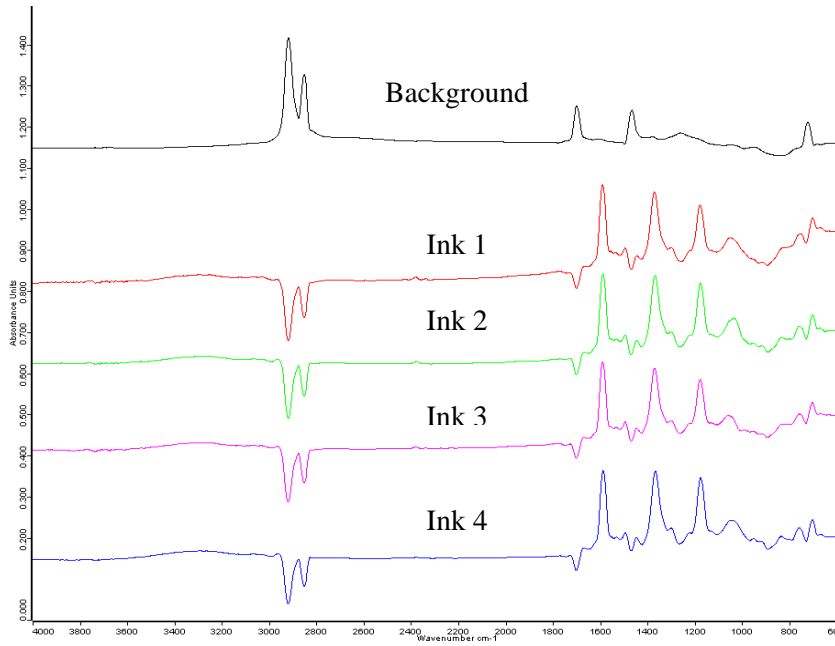
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Contacts Paper book (S25).



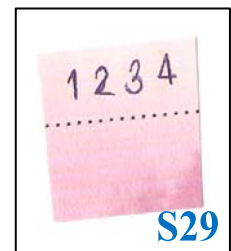
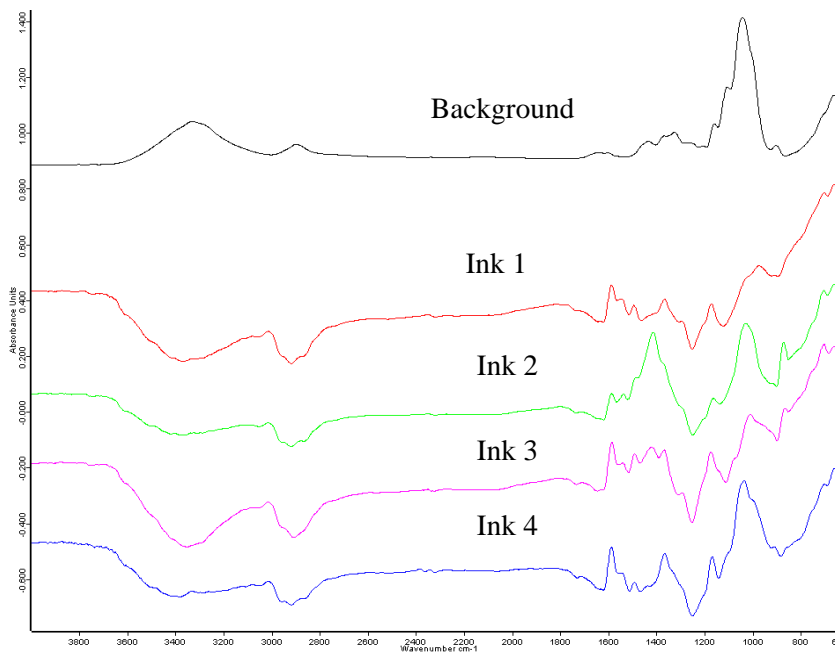
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Thai airways boarding pass (S26).



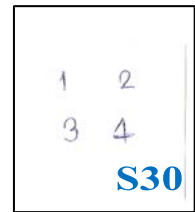
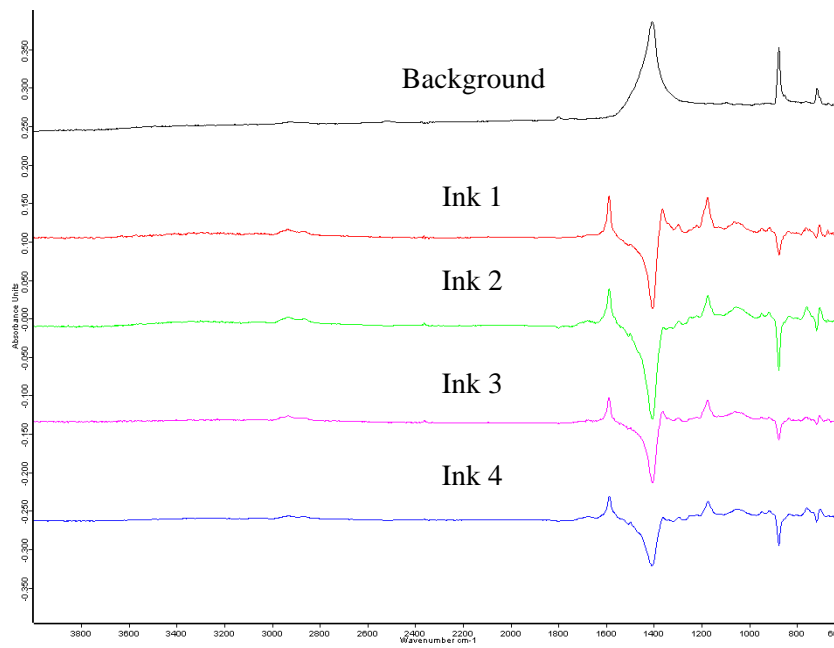
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Coated Food packaging material (S27)



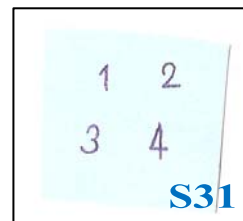
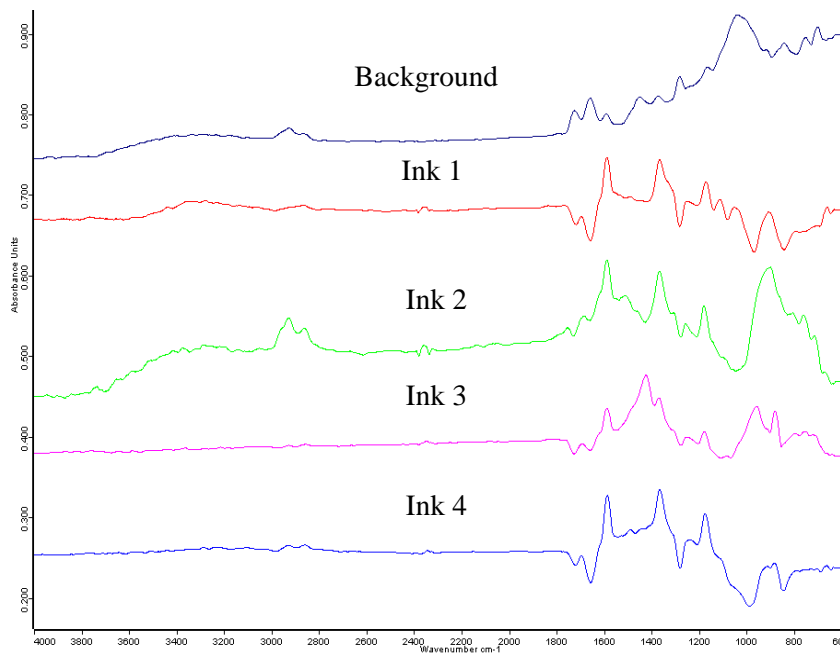
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Back side of Coated Food packaging material (S28)



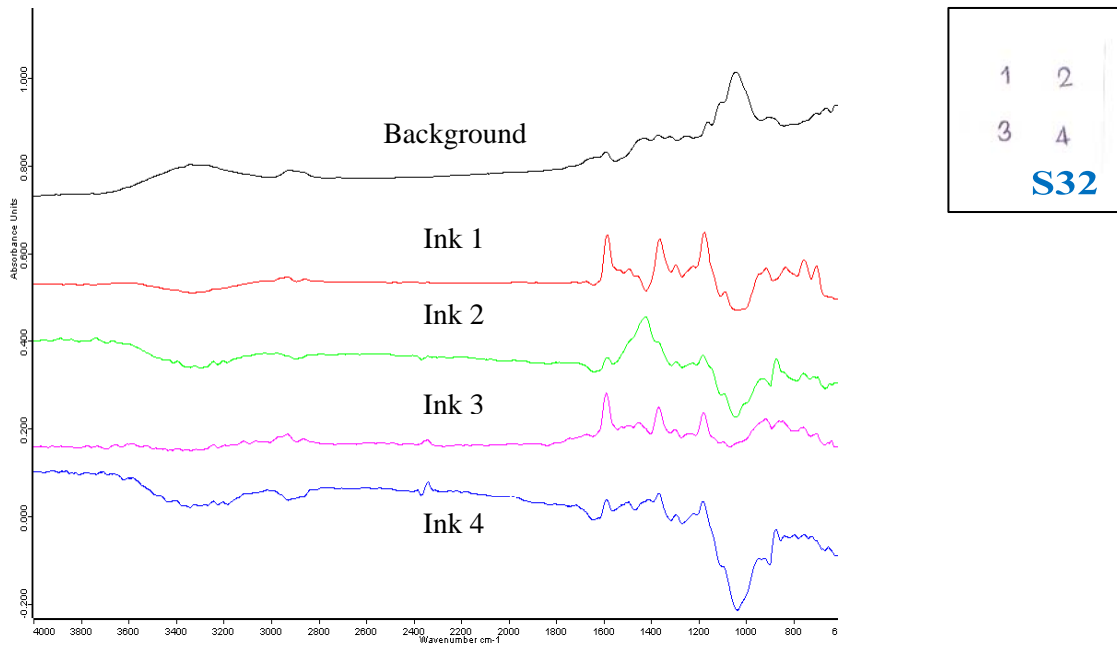
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Color ink jet printing paper (S29).



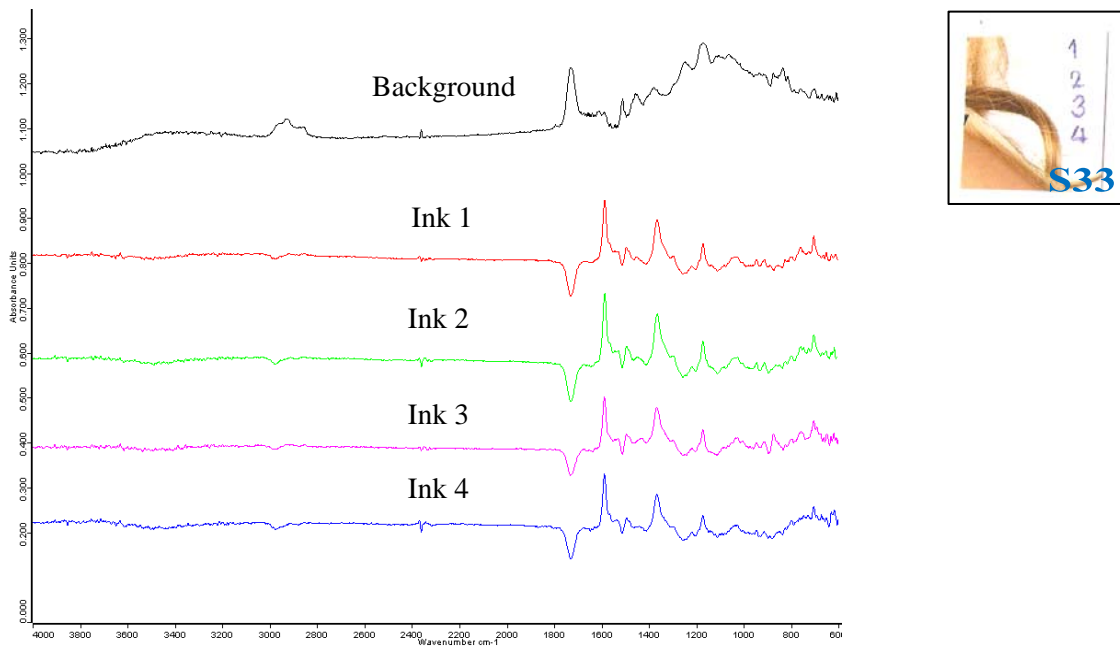
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Normal page of the Seventeen magazine(S30).



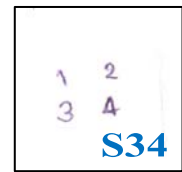
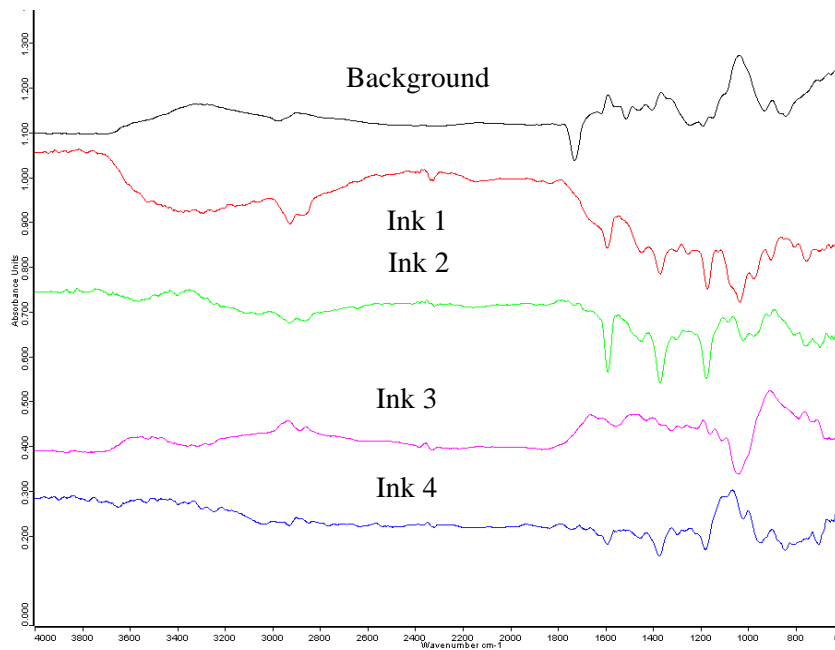
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Glossy card paper (S31)



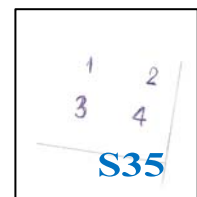
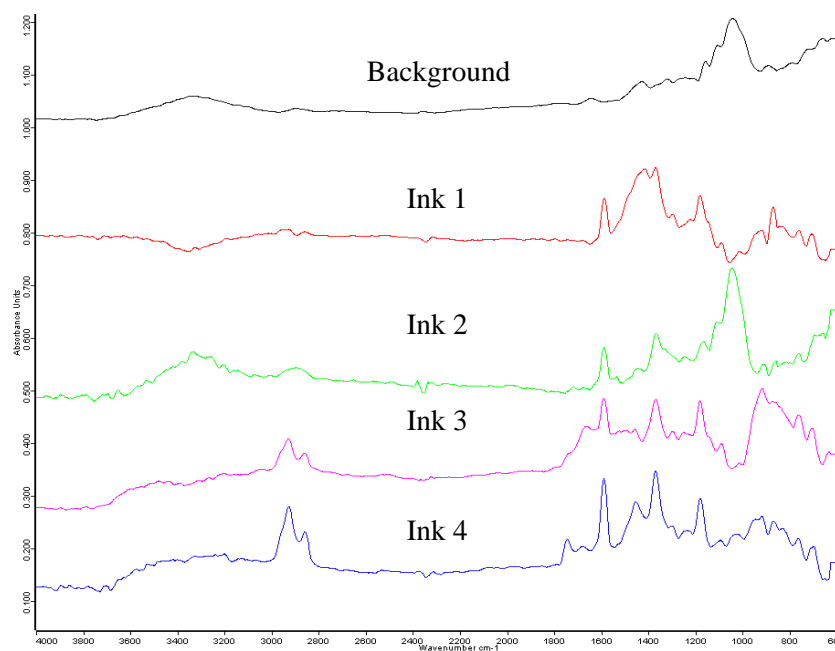
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Electricity bill (receipt) (S32).



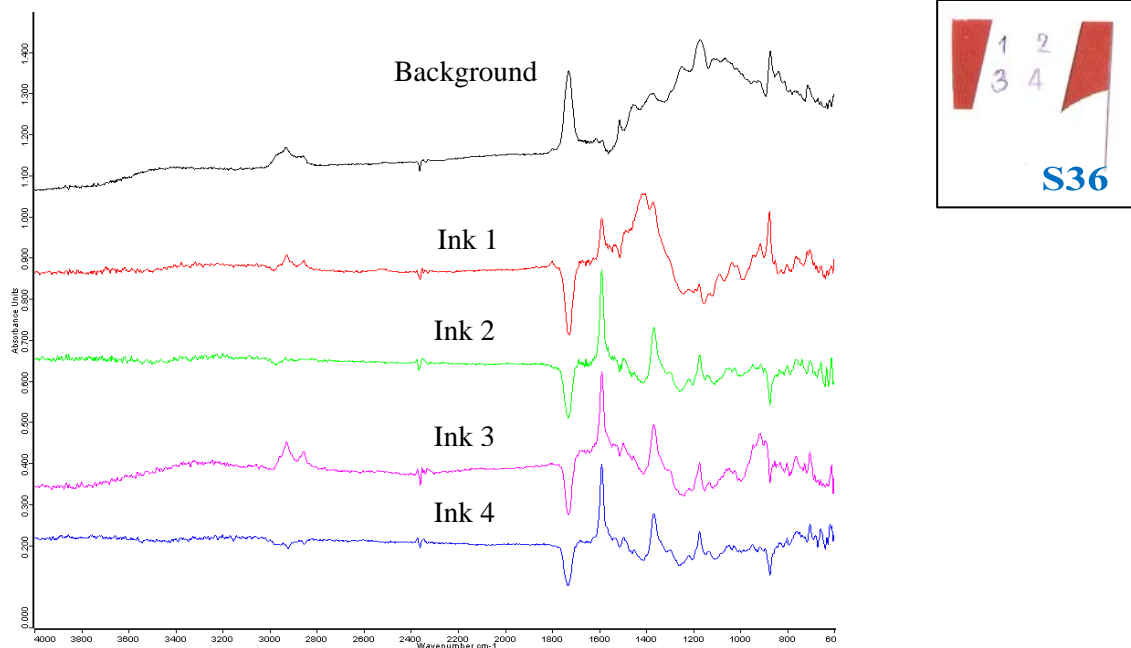
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Cover page of the Seventeen magazine (S33).



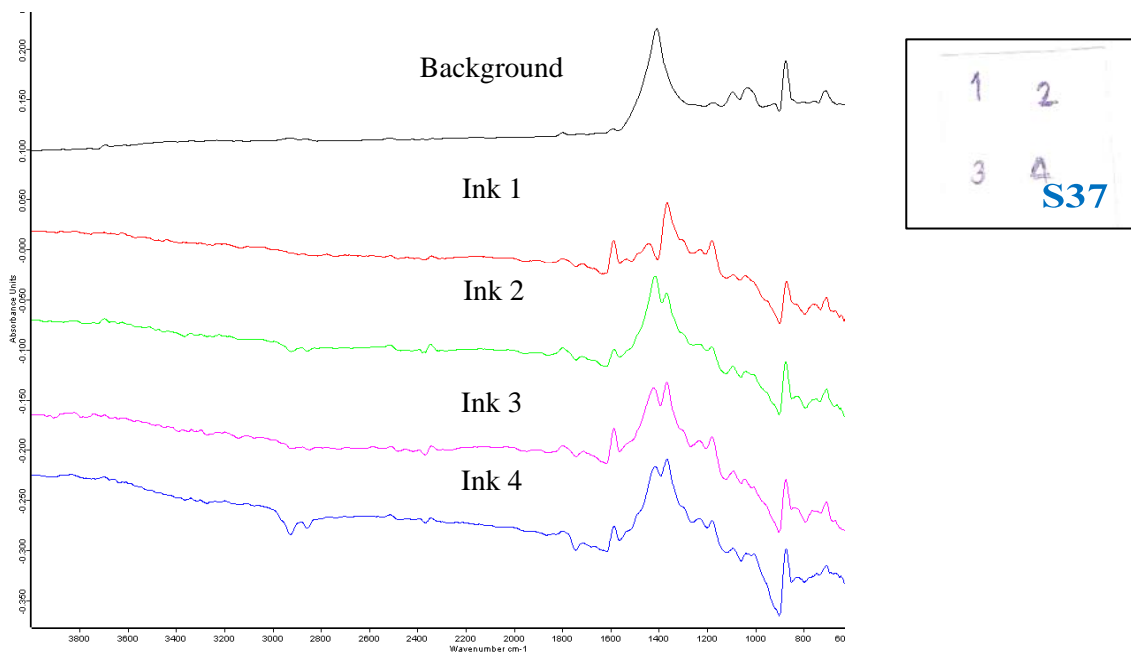
ATR-FTIR spectra of ink 1, 2, 3 and 4 on White plain roll tissue paper (S34).



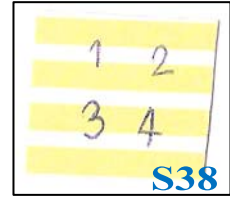
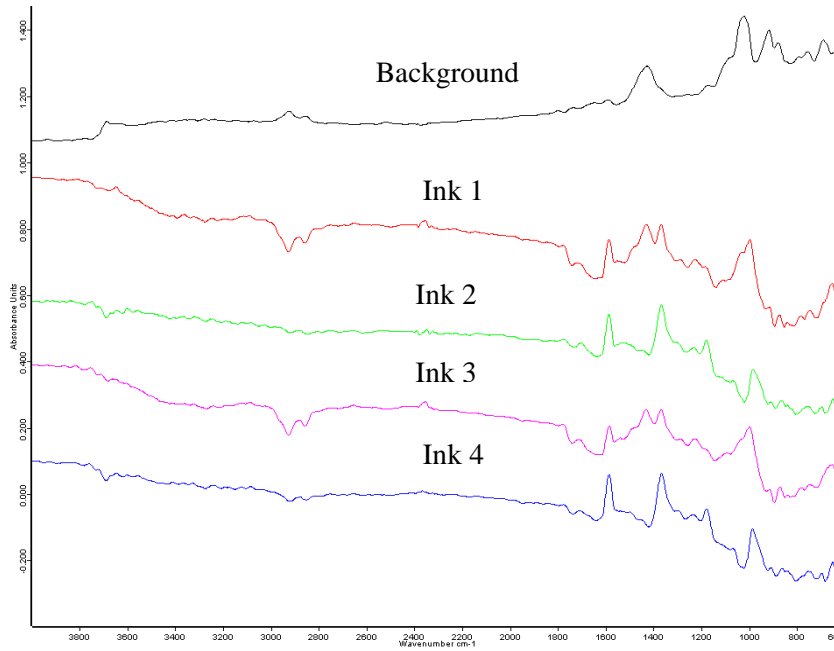
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Normal page of Honda magazine (S35).



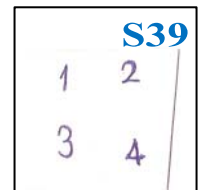
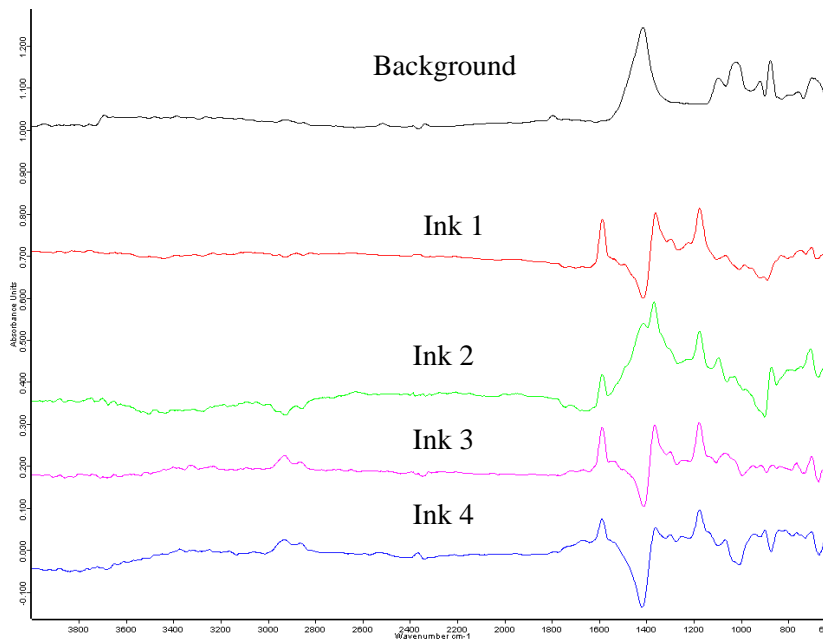
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Outside cover page of Honda magazine (S36).



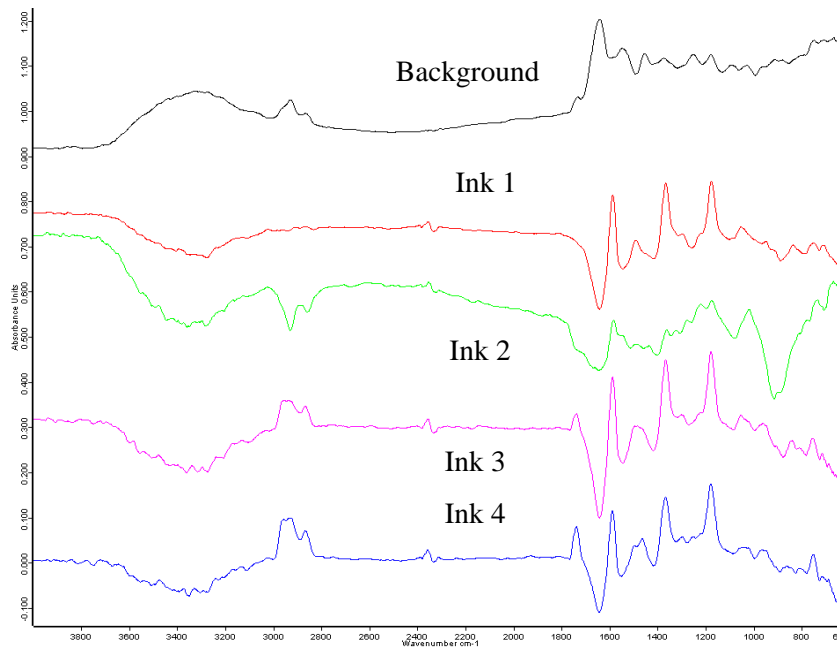
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Inside cover page of Honda magazine (S37).



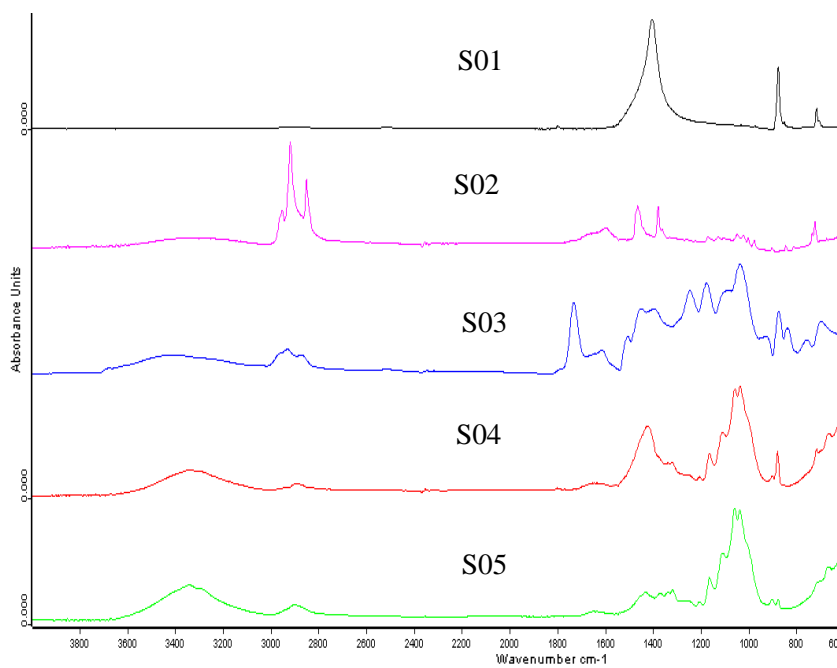
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Outside of Mister Donuts box (S38).



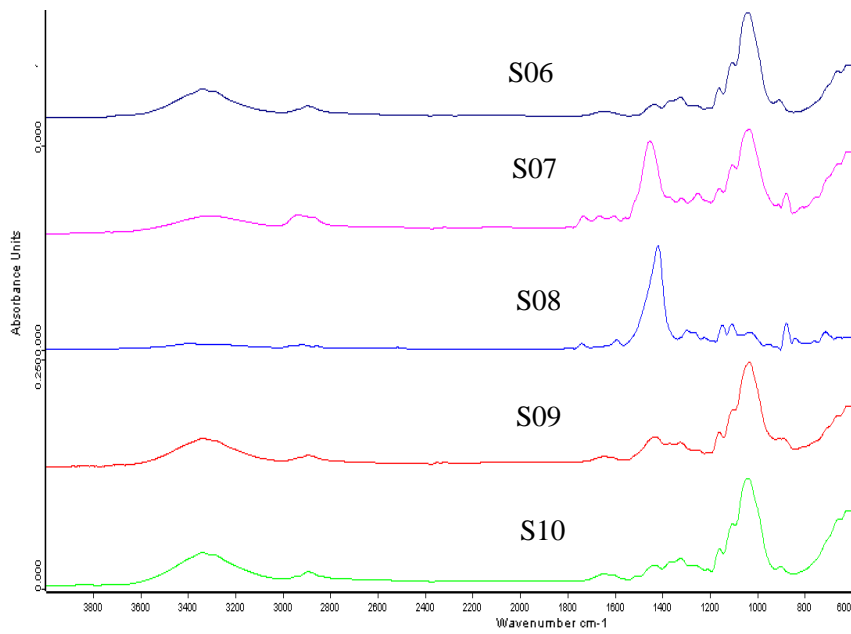
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Inside of Mister Donuts box (S39).



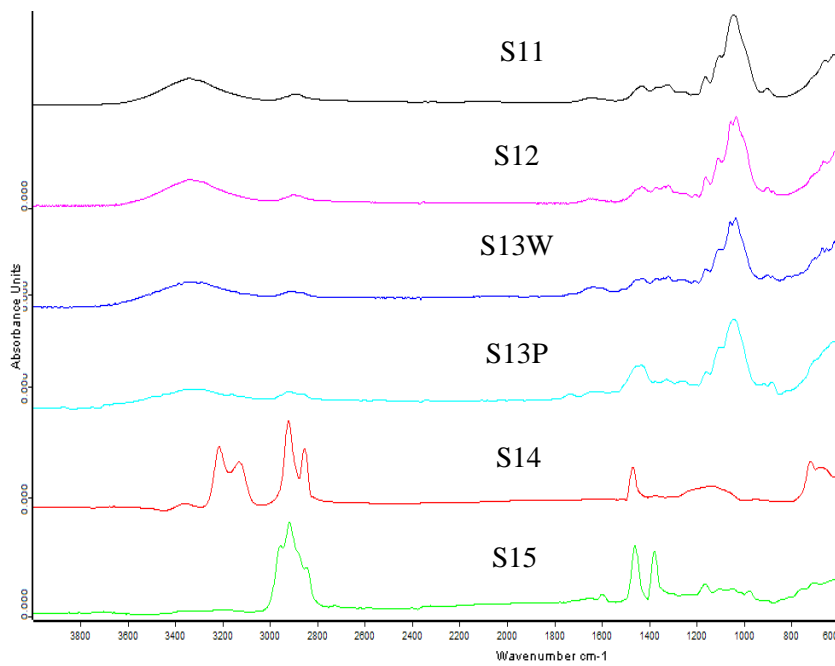
ATR-FTIR spectra of ink 1, 2, 3 and 4 on Color lab Photographic paper(S40).



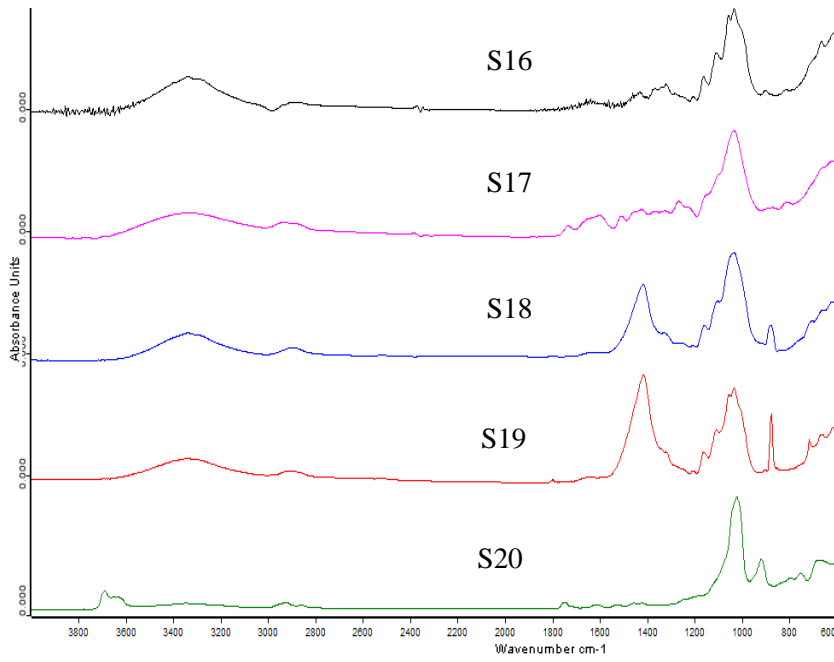
ATR-FTIR spectra of background surface number S01-S05



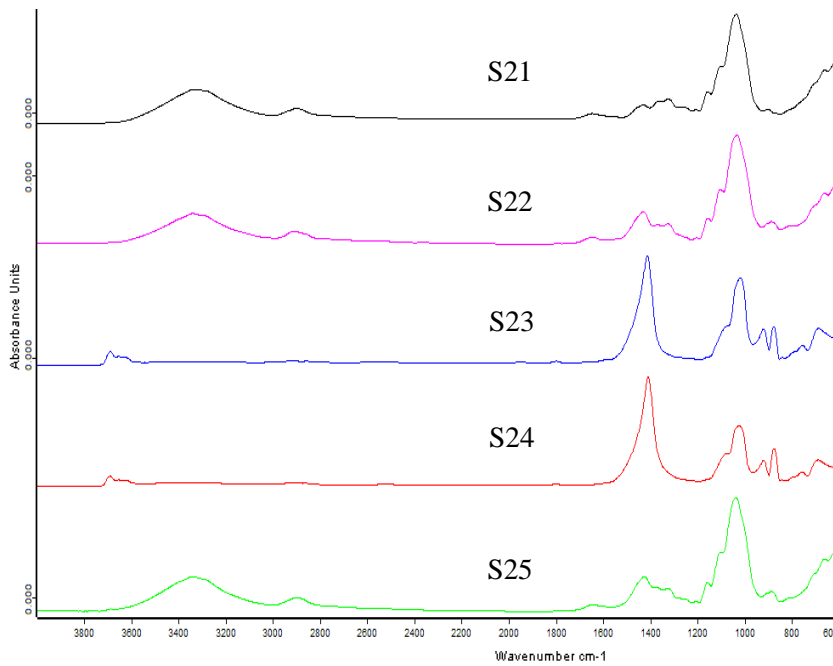
ATR-FTIR spectra of background surface number S06-S10



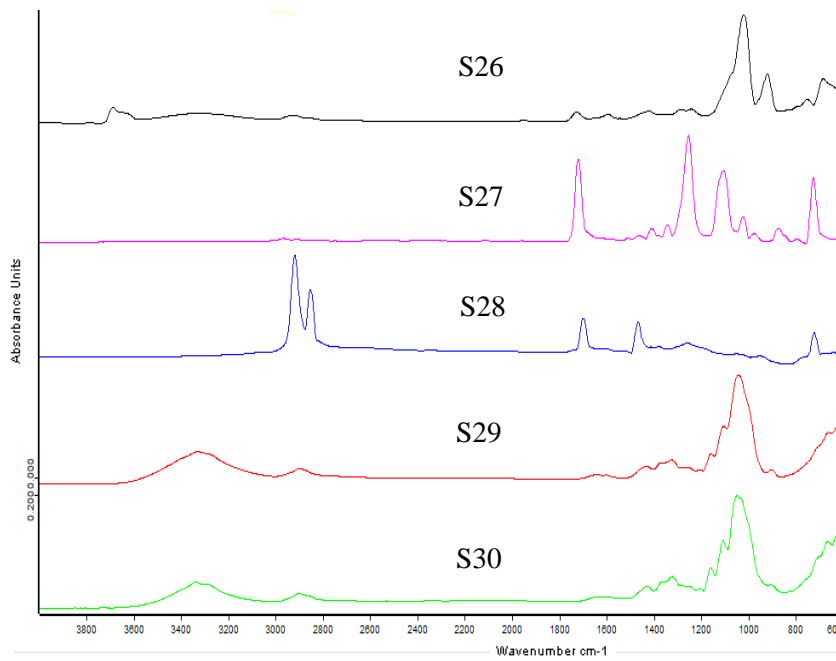
ATR-FTIR spectra of background surface number S11-S15



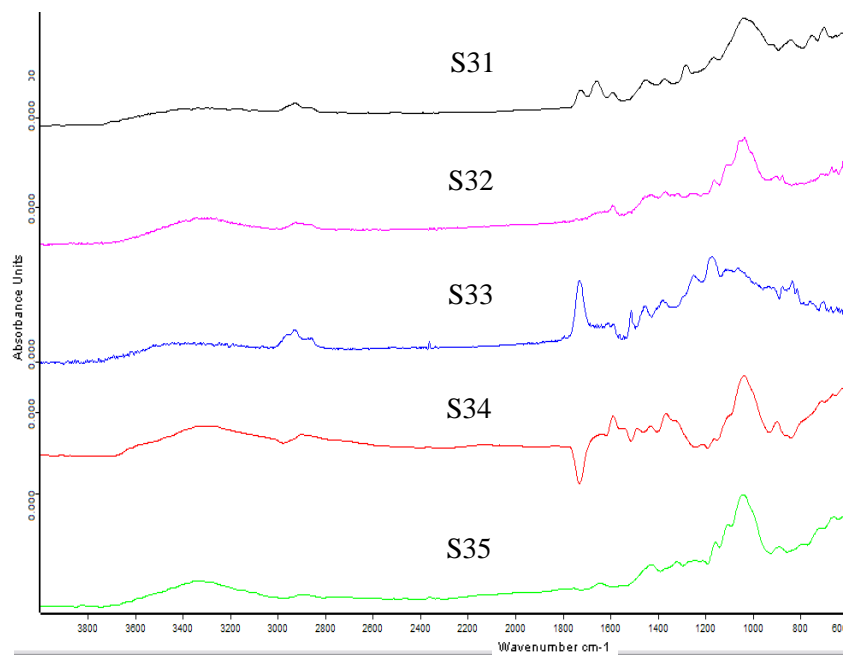
ATR-FTIR spectra of background surface number S16-S20



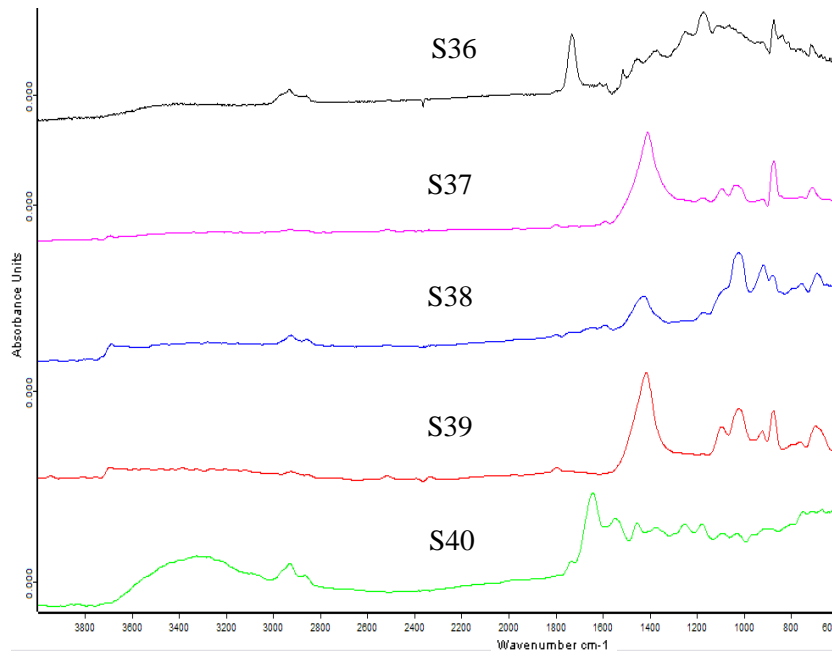
ATR-FTIR spectra of background surface number S21-S25



ATR-FTIR spectra of background surface number S26-S30



ATR-FTIR spectra of background surface number S31-S35



ATR-FTIR spectra of background surface number S36-S40

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