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Elementary vs. Advanced techniques: A Comparative Approach to Examine Various Types of Blue Inks

Mandeep Kaur Purba[1,2], Shruti Gupta[1], Reeta Rani Gupta[3]

[1]*Amity Institute of Forensic Science, Amity University, Noida, Uttar Pradesh 201313, India*
[2] *Department of Forensic Science, Shree Guru Gobind Singh Tricentenary University, Gurugram, Haryana 122505, India*
[3]*Central Forensic Science Laboratory, Central Bureau of Investigation, New Delhi 110003, India*

[1]Mandeep.kaur0792@gmail.com, [2]sgupta@amity.edu, [3]reetacbi@gmail.com

Abstract *- Forensic document examiners' wide range of tasks incorporate the expertise to determine the origin of penmanship of the document; segregate among authentic, imitated or disguised penmanship; analyze components and characteristics of inks, papers, and different materials associated with documents. The present study focuses on the comparative aspects between various simplified techniques used for blue pen ink examination with advanced instrumental techniques such asSEM and SEM-EDX. The samples were analyzed primarily for different types of alterations, overlapping of ink, and composition of ink initially by using preliminary techniques followed by advanced instrumental techniques. The examination of ink led to unexpected and astonishing results. In spite of advancements in technology, the success rate of simplified techniques was found to be higher than 90% for all the parameters studied, that is, alterations, obliterations, and overlapping of ink, whereas advanced instrumental techniques did not ascertain to be of much use except for the determination of the composition of the ink. Even in today's advanced digital age, selected simplified techniques have proved to be reliable and gave easily reproducible results for the detection of alterations and overlapping of ink. Nonetheless, the combination of simplified and advanced instrumental techniques can also be used for the determination of the composition of the ink. This article provides an insight to the examiners to select the best suitable non-destructive technique/s and conduct the examination accordingly.*

Keywords - *Questioned documents; ink examination; hand lens, light sources; UV chamber; SEM-EDX*

I. INTRODUCTION

In the present scenario, the field of questioned documents is not only limited to the analysis of cases related to handwriting but also deals with the examination of paper and ink. Over the past years, the questioned document examiners have struggled with examination and identification of inks with respect to various documents[1]. Forensic fraud analysis has always played a major role in the determination of forgery[2]. Nowadays, the examination and identification of ink serve as a piece of essential evidence to establish the authenticity of questioned documents. There have been various instances where analysis of paper and ink provided a piece of strong corroboratory evidence and served to solve the case. One such famous case was of Hitler's forged diaries. The editor of the British Sunday Times started publishing Hitler's diaries which were supposed to be genuine, in 1983. Later, these diaries were sent for forensic analysis. The police and forensic examiners preferred analyzing paper and ink instead of handwriting which finally revealed that the diaries were forged. It was observed that the type of paper and ink used were not present during the reign of Hitler[3]. Thus, it helps in establishing the fact that ink serves as important evidence. Comparison of two or more overlapped inks in various cases of forgery can help us determine the company which manufactured the ink, its batch, and the date of manufacturing. In spite of being important corroboratory evidence, the information provided by ink is still limited. The major reason behind this limitation is the type of analytical technique used for analysis[4]. It is important to determine the most appropriate instrument which can be used for producing reliable, reproducible, and effective results. In the world of digitalization, handwritten documents such as contracts, wills, etc., are still used. Various measures are used to manipulate the data or signatures present on these written documents[5]. These measures include obliteration, additions, overwriting, and many more. Needless overwriting or overlapping of inks appear to be suspicious. Analysis of inks in such manipulated or tampered documents for determination of authenticity is the need of the hour. One such technique for the analysis of manipulated documents is paper spray mass spectrometry. It can be used for the analysis of pen inks of varied brands under ambient conditions and also provide a characterization of overlapped inks[6,7].

Reliable and reproducible examination of ink using various instrumental techniques has become crucial in the field of forensic science. In recent years, incidents of forgery using different ink pens have increased exponentially[8].

A. Composition of Ink

Different types of inks are available in the market, which vary in several aspects such as color, stability, solubility in water, and solvents and pigments used while

manufacturing. The major categories of inks available in the market are ballpoint ink, gel ink, and fountain ink. Each type of ink contains two basic components, colorants and vehicle. The color of the ink is due to the dye present in the colorant, whereas the media in which the dye is dissolved is known as a vehicle. Ballpoint inks are made up of a mixture of organic dyes and several glycol solvents. In some cases, the stability of the ink is improvised by adding carbon or graphite to it[9]. It also contains fatty acids such as oleic acid, resins, viscosity adjusters, and many other substances. In contrast to ballpoint inks, gel inks consist of insoluble inorganic colored pigments. Previously, fountain pen inks contained iron-gallotannate or aqueous solutions of synthetic dyes, but modern formulation consists of synthetic dyes. The color of the ink is due to the presence of such dyes, which oxidize and turn black with the passage of time. The solvent medium in ballpoint ink is oil-based, whereas it is water-based in gel and fountain pen ink, which makes the consistency of ballpoint ink thick as compared to gel and fountain inks. The pressure required to write using ballpoint ink pens is high in contrast to gel and fountain pen ink due to its potential of getting clumped. Certain pigments such as copper pthlocyanine are also added to these inks to provide stability and permanency[10].

Ink examination is not limited to the determination of composition and type of ink, and it also deals with the pressure applied while writing on a paper and the type of paper. A wide range of instruments is available which can be successfully used for the analysis of all these factors as well as to determine the presence of more than one ink on a document. Ultraviolet lamps are successful in determining the presence of secret inks [11]. Chromatography is one of the oldest and widely used techniques for the separation of components [12]. Chromatographic techniques have always been used for the differentiation of various types of dyes and pigments of scientific importance [13]. Techniques such as thin-layer chromatography, gas chromatography, ultra-performance liquid chromatography, and many more can be successfully used for the determination of composition, and the type of ink used [14,15]. Spectroscopic techniques determine the energy absorbed or released by any atom or molecule present in a substance. These techniques are of great relevance in forensic as well as other fields of science and technology[16-21]. Spectroscopic techniques such as Raman spectroscopy and micro-FTIR spectroscopy can also be successfully used for the determination of samples of the same type and color [22,23]. However, most of the techniques used are damaging in nature; therefore, it is crucial for a forensic investigator to analyze the evidence using non-destructive techniques for preserving the evidence for further analysis as per requirement. At present, the studies directed on overlapped inks utilizing non-destructive techniques are constrained, which decides the significant reason for conducting this research. It focuses on deciding the best technique which can be successfully used for determining the presence of overlapping additions or alterations along with the composition of overlapped inks. The technique should also be suitable for deciphering obliteration.

II. MATERIALS AND METHODS

A. Sample preparation

180 samples were prepared for examination with different combinations of blue ballpoint ink, gel ink, and fountain ink. Two distinct brands of the pen were selected from each category that is ballpoint inks (flair and Reynolds), gel inks (Rorito and Natraj), and fountain pen inks (Cello and Chelpak). The samples having alterations, additions and obliterations were prepared as shown in table1.

Table 1: Combination of blue pen inks

S.No.	Combination of blue inks	Ballpoint pen ink (Flair) (B1)	Gel ink 1 (Rorito) (G1)	Fountain pen ink 1 (Cello) (F1)
1.	**Ball point ink (Reynold) (B2)**	B1,B2	G1,B2	F1,B2
2.	**Gel ink 2 (Natraj) (G2)**	B1,G2	G1,G2	F1,G2
3.	**Fountain pen ink 2 (Chelpak) (F2)**	B1,F2	G1,F2	F1,F2

B. Procedure for examination

The examination was carried out using simplified techniques consisting of hand lens (15X), oblique, transmitted, and ultraviolet light sources, followed by advanced instrumental techniques consisting of SEM and SEM-EDX. The techniques used along with the procedure followed are described in table 2.

Table 2: Description of procedure of examination

S.No.	Technique used	Procedure followed
1.	Hand Lens (15X)	• The samples were kept on a plain smooth surface • The examination was conducted with a hand lens of 15X magnification.
2.	Oblique light source	• A standard white LED torch was kept at an angle of 45°. • The examination was conducted under the light at the specified angle.

3.	Transmitted light source	• For analysis, a setup was prepared by arranging a piece of glass on two pieces of cardboard. • The same white LED torch was kept under the glass in a manner that the light falls on the center of the glass. • The sample was kept on the glass for analysis with the transmitted light.
4.	Ultraviolet light chamber	• Analysis was conducted using Tanco Ultra Violet Fluorescence Cabin. • The samples were kept inside the chamber and were observed under UV light (254nm).
5.	Scanning Electron Microscope (SEM)	• Analysis was conducted using ZEISS SEM-EDX EVO-18. The attachment of EDX with SEM helps in the identification of the elements in the sample and its quantitative composition. • Prior to examination on SEM and SEM-EDX, the samples were coated with gold particles using Quorum sputter coater to make them more conducive and to prevent them from charring due to electron beam. • Each sample was kept in the sputter coater for a duration of 45 mins and then was placed on the studs for analysis.

C. Observations and Results

Hand lens has been the most elementary aid in the field of questioned documents. For being handy and easy to use, it is still the most preferred tool for examination. The present study shows that the hand lens of 15X magnification is successful in determining the presence of overlapping in about 93.3% of the cases but deciphering obliterations and alterations was only possible in 61.1% and 67.8% of the samples, respectively. Some of the samples are shown in figure 1.

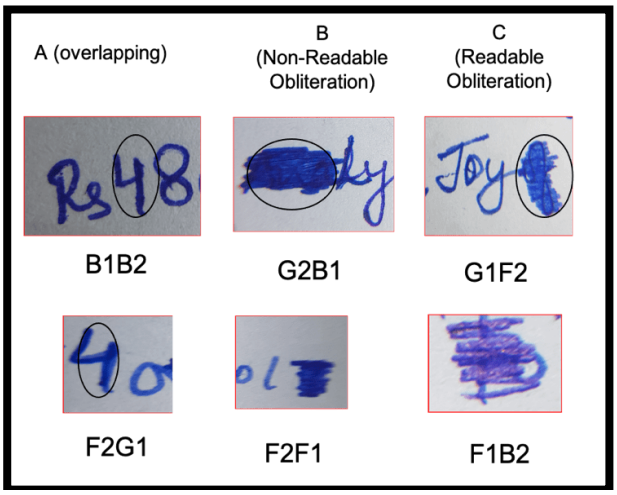


Figure 1: Observations of analysis using a Hand lens

The oblique light source is another easy-to-use method of examination. Focusing light at various angles has always been successful in the detection of alterations in the document. It continues to be an effective method of examination, according to the present study as well. It has been successfully used in determining the presence of overlapping in about 95.6% of the cases but deciphering obliterations and alterations was only possible in 66.7% and 70% of the samples, respectively. Some of the samples are shown in figure 2.

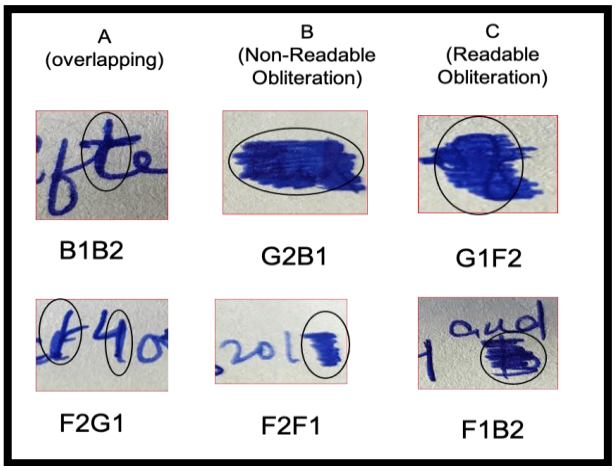


Figure 2:Observations of analysis using Oblique Light

Transmitted light sources have been and can still be used to identify slight variations in ink type. Regardless of the efficiency and skill of the forger, it can reveal the presence of two or more inks on a document. It was observed that the transmitted light source is successful in determining the presence of overlapping in about 91.7% of the cases but deciphering obliterations and alterations was only possible in about 77.8% and 82.8% of the samples, respectively. Some of the samples are shown in figure 3.

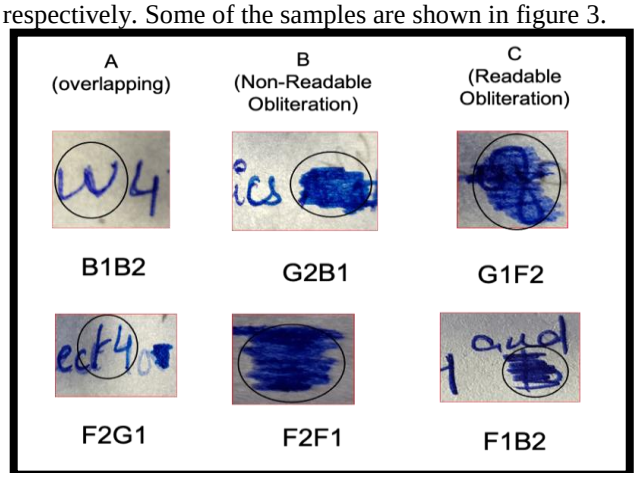


Figure 3:Observations of analysis using Transmitted light

The ultraviolet light chamber is successful in determining the presence of overlapping in about 93.9% of the cases but deciphering obliterations and alterations were only possible in 85.6% and 81.1% of the samples, respectively. Some of the samples are depicted in figure 4.

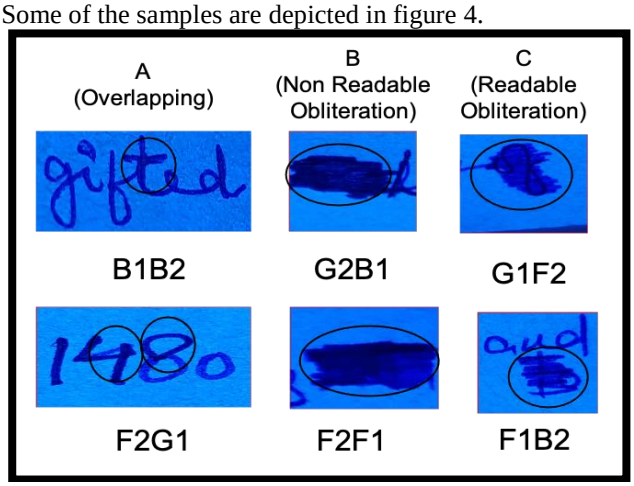
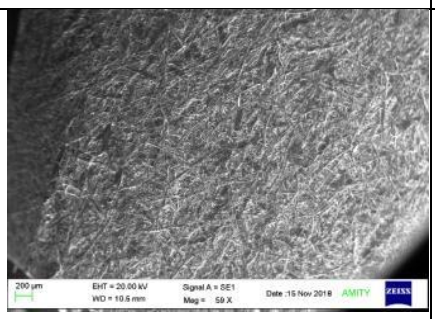
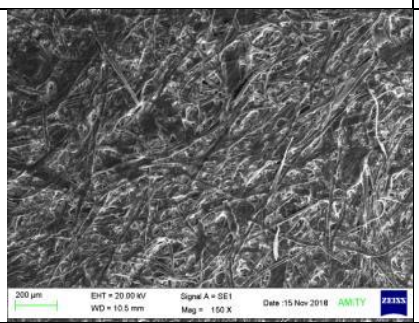
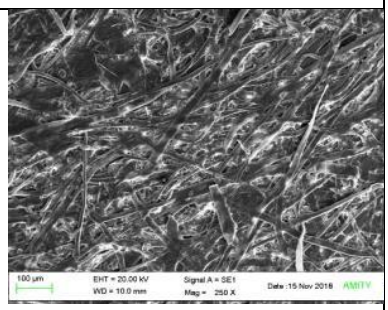
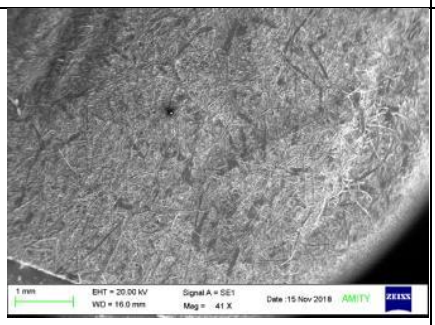
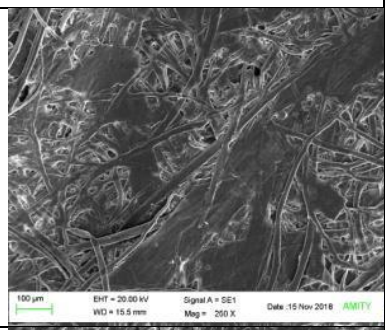
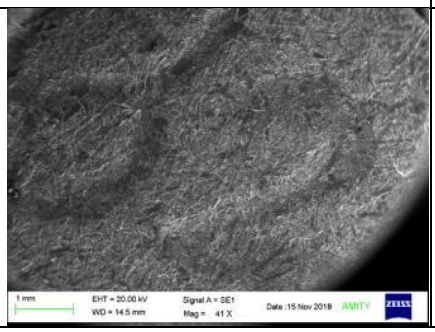
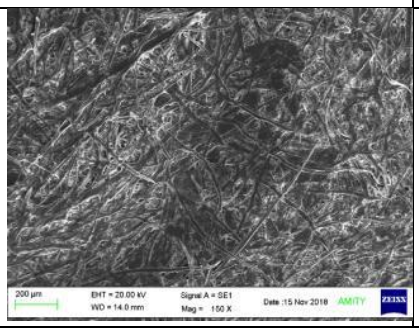
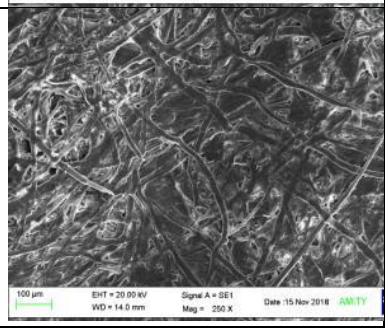
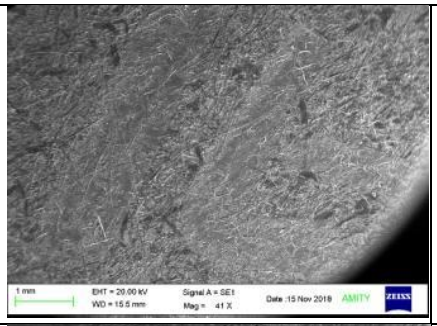
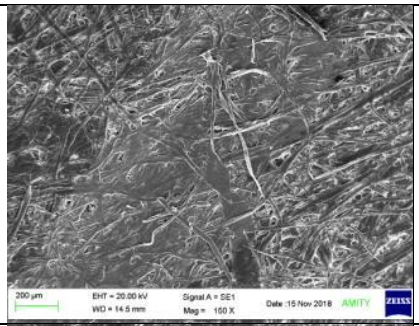
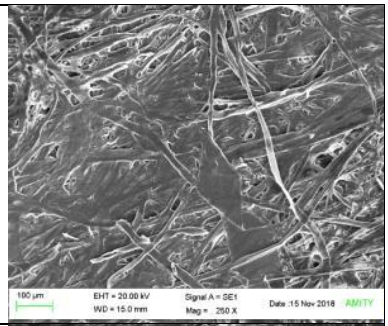
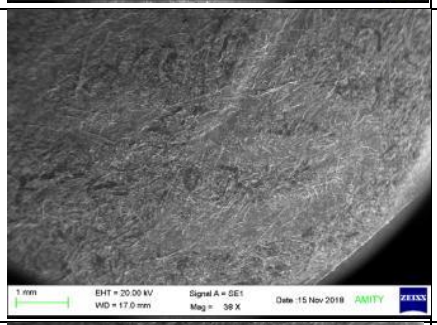
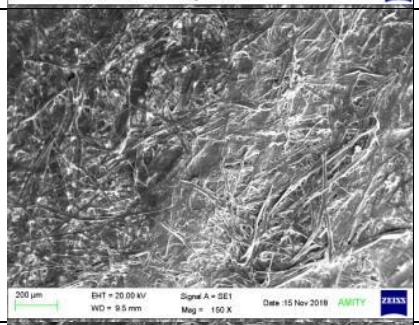
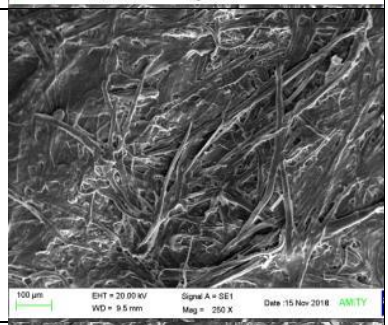
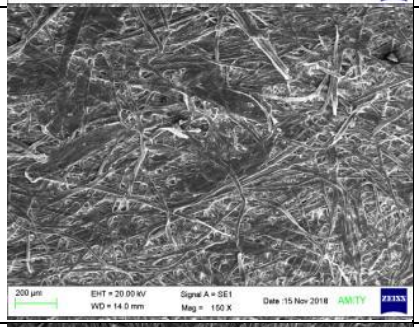
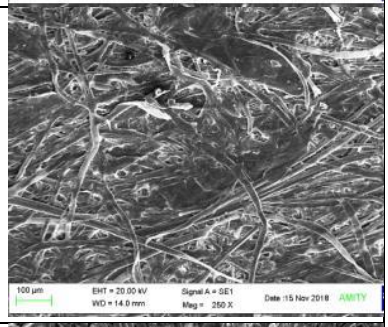
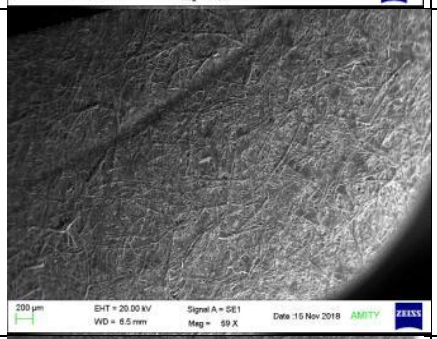
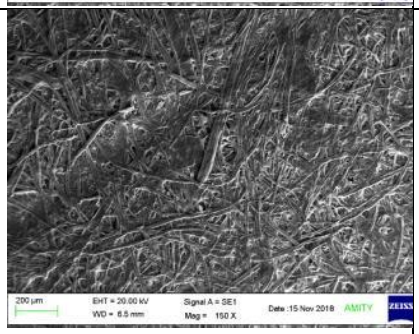
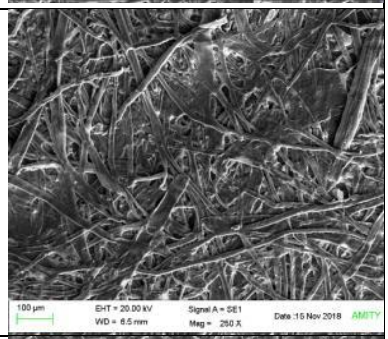
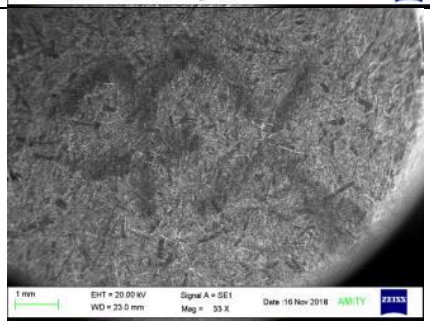
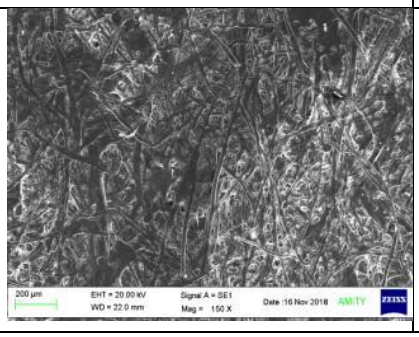
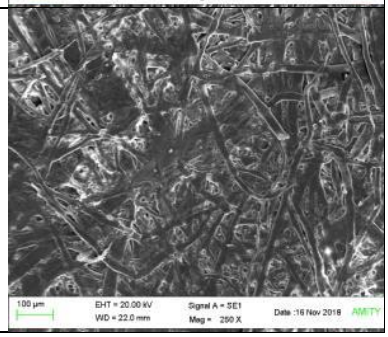


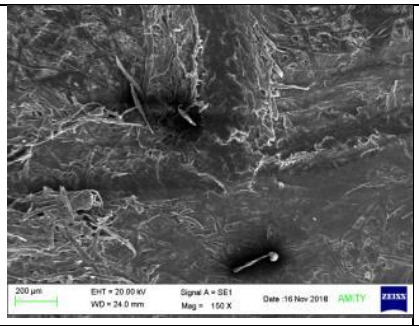
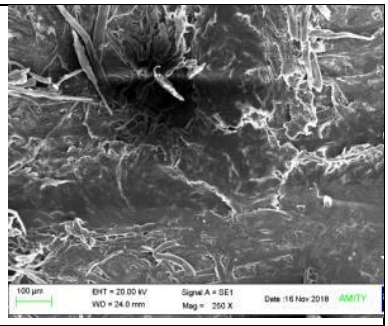
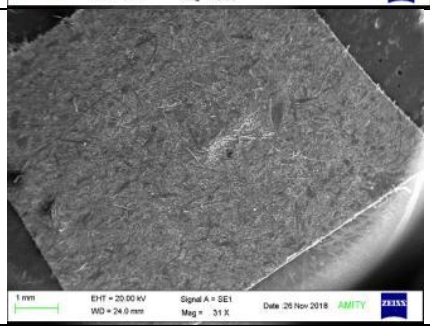
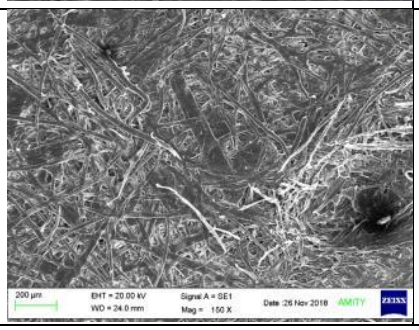
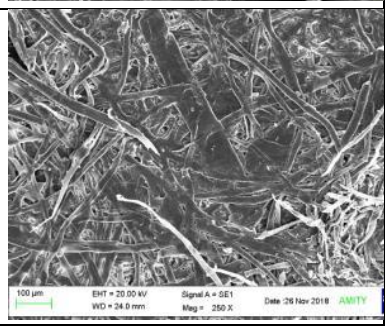
Figure 4: Observations of analysis using Ultraviolet light

SEM is successful only to detect the presence of written matter on paper but cannot be used for determining the overlap of inks. Obliteration and alterations could only be revealed in about 16% and 12% of the samples, respectively. Some of the observations are shown in table 3. It is a suitable tool for morphometrical studies of fibers and paper. It can reveal the interaction of pen and paper surfaces. The deposition of ink on paper can be seen with the help of this technique, but it only gives good results with ballpoint pen inks. Results with gel pen inks were moderately good, whereas it does not prove to be useful for fountain pen inks as writing with fountain pen inks is much smoother and does not require the application of a good amount of pressure to distort the fibers of the paper. SEM can only be used to detect the presence of written matter on paper but cannot be used for determining to the overlap of inks. Hence, SEM does not prove to be very successful for the analysis of overlapped inks or obliterations.

Table 3: Observations of analysis using SEM

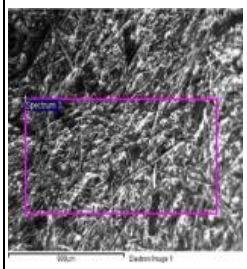
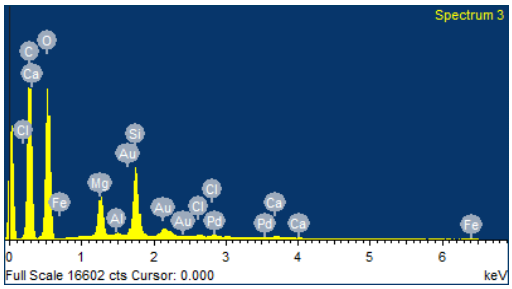
S.No	Sample	Low magnification (31-60x)	Medium magnification (120-160x)	High magnification (200x and above)
1.	Blank			
2.	B1			
3.	B2			

4.	G1			
5.	G2			
6.	F1			
7.	F2			
8.	B1B2			

9.	G1G2			
10.	F1F2			

SEM-EDX enables us to explore the chemical components and the corresponding elemental distribution of inks. It helps in carrying out qualitative (the type of elements) as well as quantitative (the percentage of the concentration of each element of the sample) analysis. This technique gave the elemental composition of the ink 80% accuracy. The addition of new elements as well as variation in concentration of certain elements in case of overlapped inks indicated that overlapping affects the elemental composition of the ink, which can be detected using SEM-EDX. The concentration might alter due to the presence of another element of overlapped ink, leading to the formation of chemical bonds which cannot be detected by SEM-EDX. Thereby, the presence of more than one ink can possibly be determined using SEM-EDX, as shown in table 4.

Table 4: Observations of analysis using SEM-EDX

<table>
<tr><th>S.No</th><th>Sample</th><th>Focused site</th><th>Spectrum</th><th>Elements and their %ages</th></tr>
<tr><td>1.</td><td>Blank</td><td>  </td><td> 
</td><td>
<table>
<tr><th>Element</th><th>Weight %</th><th>Atomic %</th></tr>
<tr><td>C K</td><td>34.90</td><td>44.00</td></tr>
<tr><td>O K</td><td>53.58</td><td>50.70</td></tr>
<tr><td>Mg K</td><td>3.45</td><td>2.15</td></tr>
<tr><td>Al K</td><td>0.15</td><td>0.09</td></tr>
<tr><td>Si K</td><td>4.90</td><td>2.64</td></tr>
<tr><td>Cl K</td><td>0.20</td><td>0.08</td></tr>
<tr><td>Ca K</td><td>0.16</td><td>0.06</td></tr>
<tr><td>Fe K</td><td>0.18</td><td>0.05</td></tr>
<tr><td>Pd L</td><td>0.55</td><td>0.08</td></tr>
<tr><td>Au M</td><td>1.93</td><td>0.15</td></tr>
<tr><td>Totals</td><td>100.00</td><td></td></tr>
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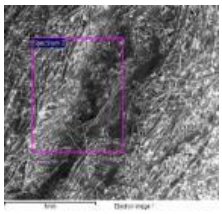
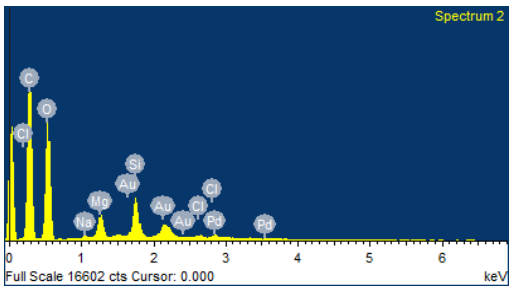
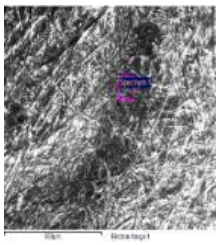
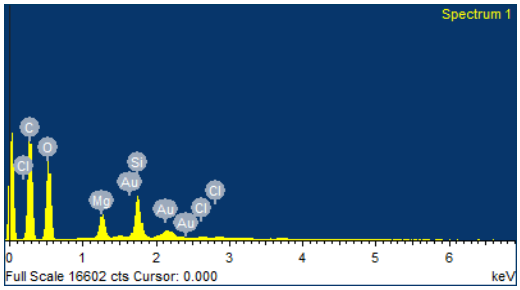
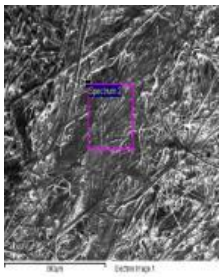
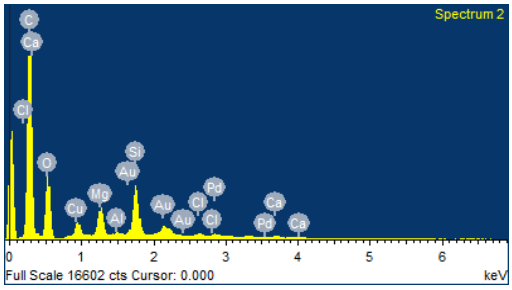
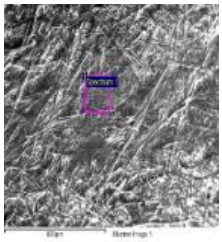
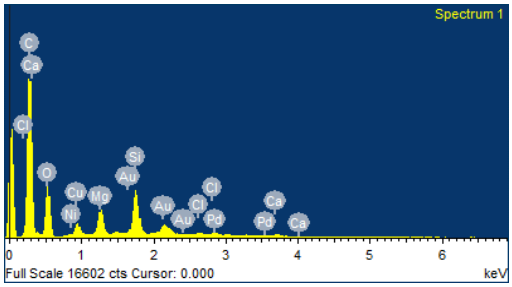
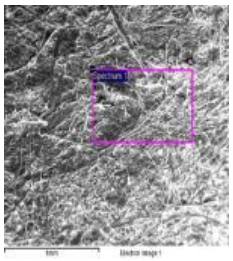
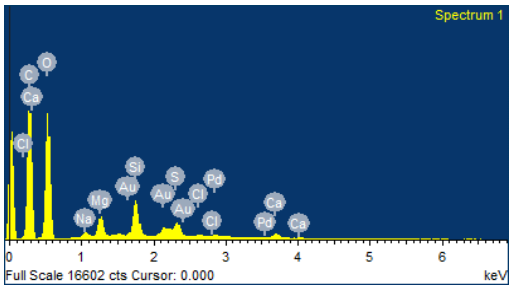
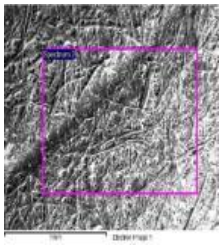
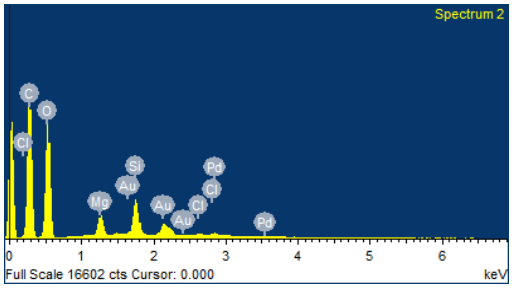
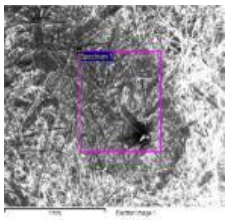
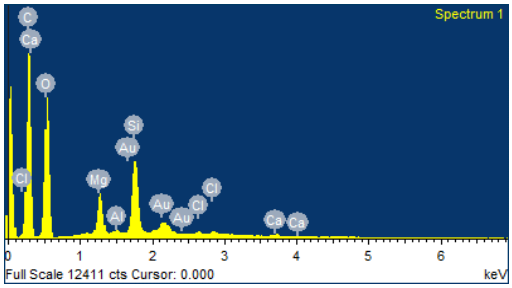
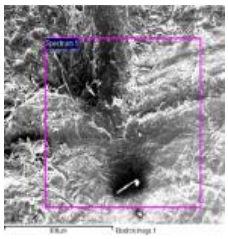
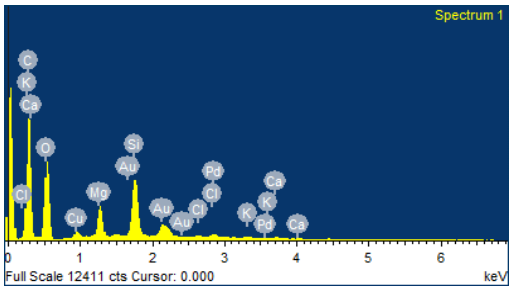
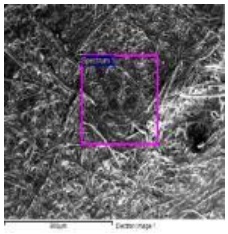
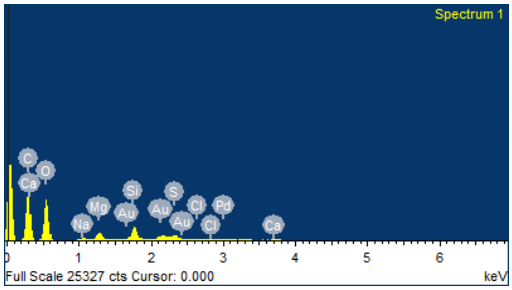
<tr><td>2.</td><td>B1</td><td>  </td><td> 
</td><td>
<table>
<tr><th>Element</th><th>Weight %</th><th>Atomic %</th></tr>
<tr><td>C K</td><td>40.71</td><td>50.45</td></tr>
<tr><td>O K</td><td>49.64</td><td>46.18</td></tr>
<tr><td>Na K</td><td>0.20</td><td>0.13</td></tr>
<tr><td>Mg K</td><td>2.01</td><td>1.23</td></tr>
<tr><td>Si K</td><td>2.88</td><td>1.52</td></tr>
<tr><td>Cl K</td><td>0.27</td><td>0.11</td></tr>
<tr><td>Pd L</td><td>0.88</td><td>0.12</td></tr>
<tr><td>Au M</td><td>3.42</td><td>0.26</td></tr>
</table>
</td></tr>
</table>

				Totals 100.00
3.	B2			Element: C K, O K, Mg K, Si K, Cl K, Au M, Totals Weight %: 48.13, 43.89, 2.33, 3.49, 0.21, 1.96, 100.00 Atomic %: 57.36, 39.27, 1.37, 1.78, 0.09, 0.14
4.	G1			Element: C K, O K, Mg K, Al K, Si K, Cl K, Ca K, Cu K, Pd L, Au M, Totals Weight %: 51.76, 34.53, 2.73, 0.20, 4.15, 0.30, 0.24, 2.90, 0.63, 2.56, 100.00 Atomic %: 63.24, 31.67, 1.65, 0.11, 2.17, 0.12, 0.09, 0.67, 0.09, 0.19
5.	G2			Element: C K, O K, Mg K, Si K, Cl K, Ca K, Ni K, Cu K, Pd L, Au M, Totals Weight %: 53.18, 32.89, 2.75, 4.12, 0.25, 0.19, 0.25, 2.66, 0.74, 2.98, 100.00 Atomic %: 64.89, 30.13, 1.66, 2.15, 0.10, 0.07, 0.06, 0.61, 0.10, 0.22
6.	F1			Element: C K, O K, Na K, Mg K, Si K, S K, Cl K, Ca K, Pd L, Au M, Totals Weight %: 35.47, 53.40, 0.65, 2.01, 2.96, 1.38, 0.23, 0.52, 0.61, 2.77, 100.00 Atomic %: 44.81, 50.66, 0.43, 1.25, 1.60, 0.65, 0.10, 0.20, 0.09, 0.21

7.	F2			<table><tr><td>Element</td><td>Weight %</td><td>Atomic %</td></tr><tr><td>C K</td><td>40.39</td><td>49.97</td></tr><tr><td>O K</td><td>50.52</td><td>46.92</td></tr><tr><td>Mg K</td><td>1.92</td><td>1.17</td></tr><tr><td>Si K</td><td>2.82</td><td>1.49</td></tr><tr><td>Cl K</td><td>0.18</td><td>0.08</td></tr><tr><td>Pd L</td><td>0.78</td><td>0.11</td></tr><tr><td>Au M</td><td>3.39</td><td>0.26</td></tr><tr><td>Totals</td><td>100.00</td><td></td></tr></table>
8.	B1B2			<table><tr><td>Element</td><td>Weight %</td><td>Atomic %</td></tr><tr><td>C K</td><td>48.91</td><td>57.99</td></tr><tr><td>O K</td><td>43.45</td><td>38.67</td></tr><tr><td>Mg K</td><td>2.26</td><td>1.33</td></tr><tr><td>Al K</td><td>0.12</td><td>0.07</td></tr><tr><td>Si K</td><td>3.34</td><td>1.70</td></tr><tr><td>Cl K</td><td>0.23</td><td>0.09</td></tr><tr><td>Ca K</td><td>0.14</td><td>0.05</td></tr><tr><td>Au M</td><td>1.54</td><td>0.11</td></tr><tr><td>Totals</td><td>100.00</td><td></td></tr></table>
9.	G1G2			<table><tr><td>Element</td><td>Weight %</td><td>Atomic %</td></tr><tr><td>C K</td><td>39.18</td><td>50.87</td></tr><tr><td>O K</td><td>43.53</td><td>42.43</td></tr><tr><td>Mg K</td><td>3.69</td><td>2.37</td></tr><tr><td>Si K</td><td>5.46</td><td>3.03</td></tr><tr><td>Cl K</td><td>0.33</td><td>0.15</td></tr><tr><td>K K</td><td>0.19</td><td>0.08</td></tr><tr><td>Ca K</td><td>0.21</td><td>0.08</td></tr><tr><td>Cu K</td><td>2.07</td><td>0.51</td></tr><tr><td>Pd L</td><td>1.01</td><td>0.15</td></tr><tr><td>Au M</td><td>4.33</td><td>0.34</td></tr><tr><td>Totals</td><td>100.00</td><td></td></tr></table>
10.	F1F2			<table><tr><td>Element</td><td>Weight %</td><td>Atomic %</td></tr><tr><td>C K</td><td>34.46</td><td>44.26</td></tr><tr><td>O K</td><td>52.76</td><td>50.86</td></tr><tr><td>Na K</td><td>0.80</td><td>0.54</td></tr><tr><td>Mg K</td><td>1.98</td><td>1.26</td></tr><tr><td>Si K</td><td>3.04</td><td>1.67</td></tr><tr><td>S K</td><td>1.44</td><td>0.69</td></tr><tr><td>Cl K</td><td>0.30</td><td>0.13</td></tr><tr><td>Ca K</td><td>0.39</td><td>0.15</td></tr><tr><td>Pd L</td><td>0.85</td><td>0.12</td></tr><tr><td>Au M</td><td>3.98</td><td>0.31</td></tr><tr><td>Totals</td><td>100.00</td><td></td></tr></table>

The number of samples identified using different instrumental techniques is depicted in graph 1. Along with it, table5 gives an overview of the success rate of the techniques used and provides researchers to select the appropriate technique.

Graph 1: Number of samples identified using different instrumental techniques

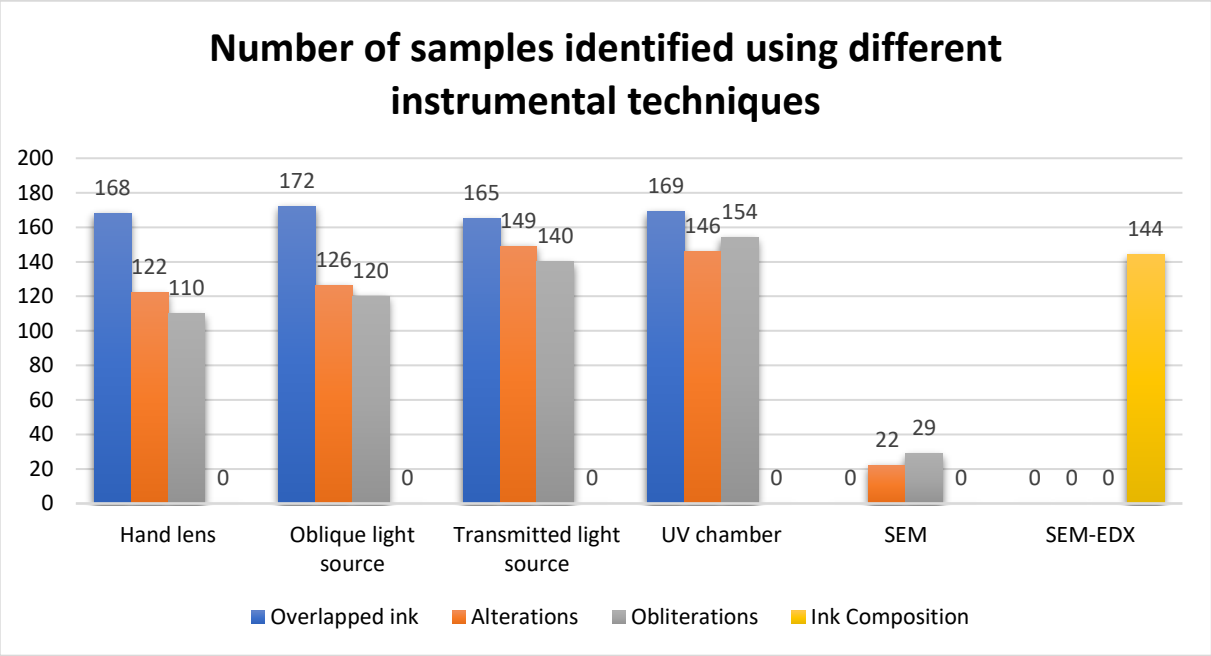


Table 5: Comparative success rate of non-destructive techniques

S.No.	Parameter	Hand Lens		Oblique light source		Transmitted light source		UV chamber		SEM		SEM-EDX	
		No. of samples identified	%	No of samples identified	%	No of samples identified	%	No of samples identified	%	No of samples identified	%	No of samples identified	%
1.	Overlapped ink (n= 180)	168	93.3%	172	95.6%	165	91.7%	169	93.9 %	0	0	0	0
2.	Alterations (n= 180)	122	67.8%	126	70%	149	82.8%	146	81.1 %	22	12 %	0	0
3.	Obliterations (n= 180)	110	61.1%	120	66.7%	140	77.8%	154	85.6 %	29	18 %	0	0
4.	Ink composition (n= 180)	0	0	0	0	0	0	0	0	0	0	144	80 %

III. CONCLUSION

Simplified techniques are less advanced but still prove to be better, convenient, and cost-effective for the determination of overlapping. Other alterations such as obliteration, additions, and overwriting can also be deciphered in the certain majority of the samples by using these techniques. On the other hand, advanced instrumental techniques are expensive, and the cost of analysis per sample is quite high. The availability of such high-end instruments is also limited. Moreover, SEM and SEM-EDX are not easy to use; proper training is required before conducting analysis on these instruments. Simplified techniques are easily available, and they are reliable and effective as per their performance is concerned, and moreover, they are affordable and convenient to use. Advanced techniques are more apt for the determination of the elemental composition of ink rather than detection of overlapping in inks is concerned. In spite of the development in advanced technology, simplified techniques have still proved to be extremely useful for analysis and giving conclusive results in this research. Nevertheless, a combination of simplified and

advanced techniques can also be used to aid the analysis. Therefore, with this research work, it can be inferred that simplified techniques are still successful and effective techniques that can provide fruitful and trustworthy results in these cases.

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