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RESEARCH ARTICLE
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DETERMINATION OF BALLPOINT AND GEL PEN INKS USING UV VIS SPECTROSCOPY

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Abstract
This study investigates methods to estimate the relative age of ink in questioned documents using thin-layer chromatography (TLC) and UV-visible spectroscopy. Ink samples from ballpoint and gel pens were extracted from simulated entries on A4 paper and aged naturally under controlled conditions. TLC analysis measured retention factor (Rf) values, while UV-visible spectroscopy recorded absorption spectra to track ink changes over time. Results revealed that ink aging, influenced by solvent evaporation and dye degradation, can be effectively monitored using these methods. The proposed approach offers a simple and reliable way to estimate ink age, supporting forensic document analysis.

Keywords: Ink aging, thin-layer chromatography, UV-visible spectroscopy, forensic analysis, questioned documents.

1. Introduction

The majority of dating problems are difficult; therefore, every factor that might have some context on the matter must be taken into consideration. Evidence must be sought from the paper itself, any printed/typewritten matter thereon, as well as from any handwriting or other markings that are present; even the design of a paper fastener or the nature of any trace of adhesive present may very occasionally be taken into consideration. In the examination of the paper, the fiber make-up, the nature of the filler, the size, and even the exact nature of the dyestuff used for improving its color all have to be taken into consideration. In marked contrast, with but few exceptions, where contemporary documents are concerned, it is rare for the examination of the paper to yield any significant data from which its age may be deduced. The history of paper-making helps explain the methods used to determine the relative and absolute age of ink in questioned documents. Early researchers identified a method for estimating the relative age of ink by comparing the solvent content between questioned and known dated samples. For absolute age estimation, the changing ratio of solvent components in heated versus unheated samples is analyzed, even in the absence of known samples. These methods have been found to be accurate and reliable. Yuanyuan Xu et al.

researched the relative and absolute age of roller and gel ink using gas chromatography (GC) and UV-visible spectroscopy methods [1]. In 1987, Cantu and Prough developed the Solvent Extraction Technique to measure the relative age of ink [2]. They noted that for the method to work effectively, the compared inks must have the same formula and be applied to the same type of paper—or at least paper of the same manufacture under identical storage conditions. Mitchell proposed a method for extracting iron-gall ink from paper using a reagent for further analysis [3]. He suggested that ink that has been on paper for a year or more reacts slowly with oxalic acid. If it has been on the paper for six to ten years, the reaction is significantly delayed, which can be used to estimate the relative age of a document. Soderman and O'Connel were the first to describe the accelerated aging of ink for age determination [4]. They proposed that inks that have been exposed to radiation are more difficult to dissolve in water compared to non-exposed inks, indicating that the ink is older. However, if both inks dissolve similarly, the test is inconclusive. Early ballpoint pen inks contained solvents such as castor oil and mineral oil, with glycols being introduced in the 1950s. Today, common solvents in this ink group include phenoxyethanol, di propylene glycol, phthalic anhydride, oleic acid, benzyl alcohol, and butylene glycol.

Gel inks, popular for their smooth writing qualities, have been widely used since 1984, particularly in blue ink formulations containing pigments like Carbazole violet, Acid blue 9, and Phthalocyanine blue. In the context of ink aging, it is helpful to differentiate between closed and open systems. A closed system refers to ink contained in the writing instrument's reservoir, while an open system refers to ink that has been deposited on paper and exposed to environmental conditions. In 2005, Andrasko and Kunicki studied ink aging within ballpoint pen chambers [5]. They found no noticeable aging effects in inks within the chamber of a regularly used pen. However, they did observe significant aging near the tip of pens that had been used for years, with the evaporation of volatile compounds (notably phenoxyethanol) and degradation of the dye mixture. For an open system, two profiles are defined: static and dynamic. The static profile refers to the unchanging characteristics of the ink, offering a reliable means to compare inks and establish differences in their composition over time. The static approach can also help identify the source of ink by matching its profile to an ink library, which can be used to detect anachronisms and potential fraud. In contrast, the dynamic profile considers the aging processes that occur after the ink is applied to paper, such as colorant degradation, solvent evaporation, and resin hardening.

Relative age refers to determining which of two inks—having the same formula and applied to the same paper—was used first. This is a dynamic concept where inks of different ages are compared. The conditions for relative age determination include identical ink formulas, paper type, storage conditions, and ink quantity. On the other hand, absolute age refers to determining the ink's age without needing to identify its formula or compare it to other inks. Aginsky's operational approach introduced absolute age estimation methods, which can be helpful even without knowing the specific ink formula.This study aims to establish a simple and understandable method for estimating a document's relative age, using UV-visible spectroscopy as a practical tool.

2. Methodology

2.2.1. Sample preparation

Preparation of simulated samples was done on A4 size 80gsm paper; straight lines were drawn on paper for preparing the ink sample. The ink listed in Table 1 is used for examination purposes; the pens were purchased from the local market of Ahmedabad, India. Further, for natural aging, drawn ink entries are stored in darkness at the controlled condition of room temperature, which is dated every month. For examination purposes, ink was not directly taken from the pen; it was extracted from paper because, in genuine situations, FDE gets forged paper and has to examine ink from the written document. Therefore, the changes of the ink with time in the open system were studied by cutting 2cm in length samples of entries and then extracting the ink in methanol.

Table 1 types of pen with brand name consider in the study

Ballpoint pen	Ballpoint gel pen
Pierre Cardin	Cello max writer
Cello Gripper	2k Add gel
Cello jet ace	Montex active gel
Classmate to ball	Add gel achiever

2.2.2 Instrumentation

Two methods were employed to determine the relative age of documents: thin-layer chromatography for the preliminary examination of ink constituents and UV-visible spectrophotometry for the determination of absorbance at specific λ max.

2.2.3 TLC analysis

For Thin-layer chromatographic analysis, Merck TLC Plate Silica Gel 60 F254 was used with the following parameters.
Solvent system - Ethyl Acetate (14): Methanol(7): Water(6)
Stationary Phase: - Silica Gel G
Solvent: - Methanol
As the ink itself has color pigments, there is no need for spraying reagents.

2.2.3.1 Procedure

1. Small pieces of size 5X7cm are cut from Merck TLC Plate Silica Gel 60 F254.
2. Afterward, the ink spots were kept at one end of the plates using capillaries, maintaining a proper distance to prevent the samples from being intermixed.
3. Then, the plates are slowly and carefully placed in the solvent system.
4. The solvent is allowed to run up to 70% of the distance of the plate taken out and allowed to dry.
5. Measurements of the distances traveled by solvent and solute are noted down.

2.2.4 UV-VISIBLE SPECTROPHOTOMETER:

Ultraviolet spectroscopic analysis was done on Jasco V-570 UV/VIS spectrophotometer with thermo-electric controller and Pbs power supply. The following are the specifications-

1. Wavelength: 190-2500nm
2. Bandwidth Selectable:0.1-40nm
3. Stray Light: Less than 0.03%(0.5% at 1690nm)

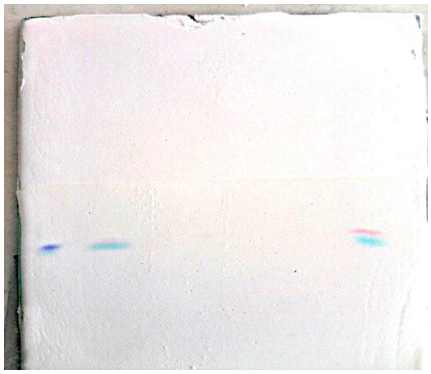
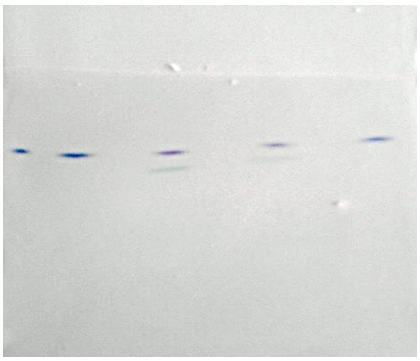
2.2.4.1 Procedure:

1. Extracted ink samples in 1ml methanol are allowed to stand for 20 minutes to maximize the amount of all available volatile components of the ink.
2. The solution is extracted secondarily with 0.9mL DMF for 20 minutes to maximize the amount of all available dye components of the ink.
3. UV-Vis analyses the solvent of extraction, and absorption spectra of the colored extracts obtained are recorded. The graphs obtained are overlayed and analyzed.

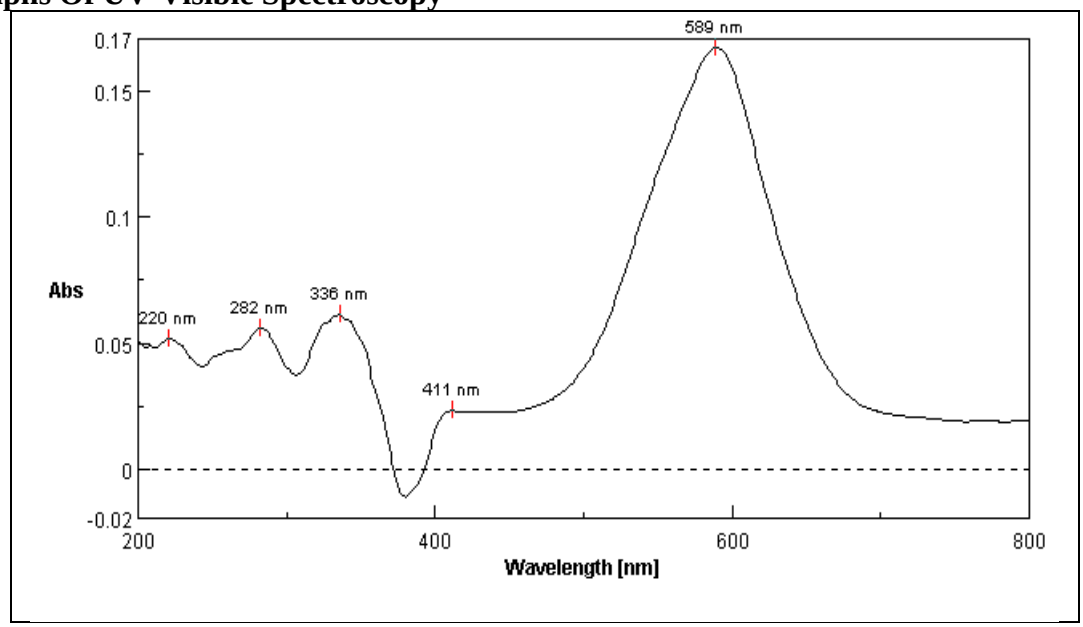
3. Results and Discussion

Differences in retention factor (R_f)values of the same type of pen in blue ink within a period were observed as follows: R_f value = Distance traveled by solute / Distance traveled by the solvent.

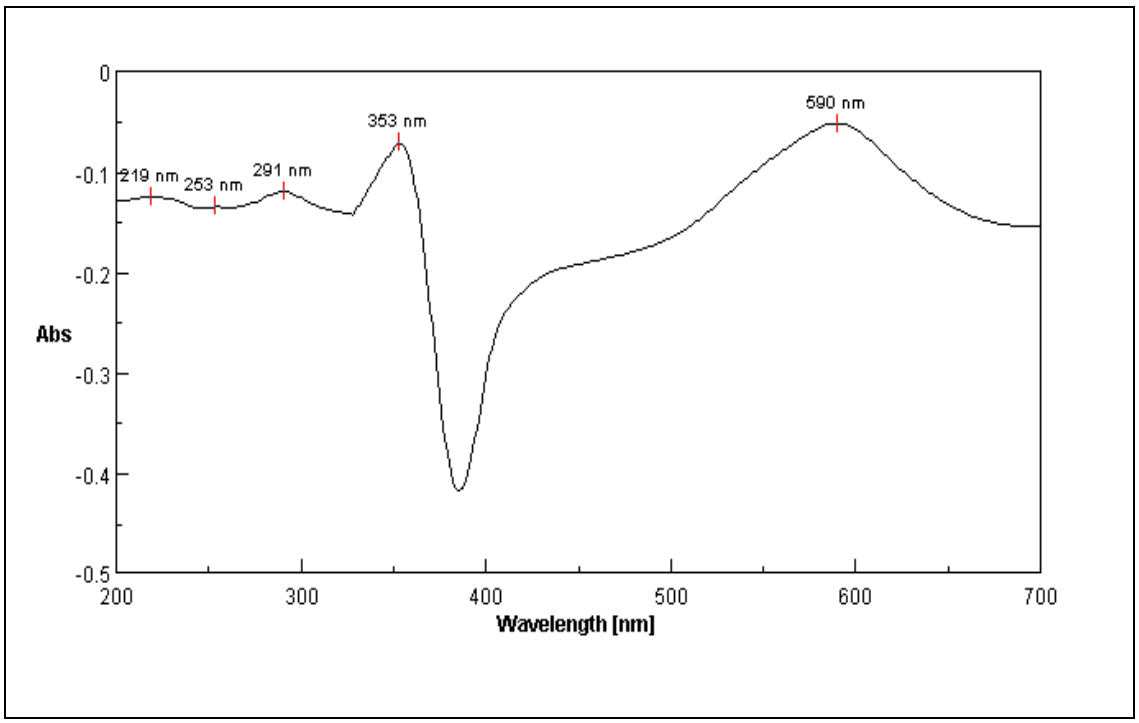
Table 2 TLC of different inks

GEL PENS From left to right		**BALL PENS** From left to right	
			
1)	2K Add Gel	1)	Pierre Cardin
2)	Add Gel Achiever	2)	Cello Gripper
3)	Classmate Octane	3)	Cello Jet-Ace
4)	Flair Osmium Gel	4)	Cello Maxwriter
5)	Montex Activa Gel	5)	Classmate UVO Ball

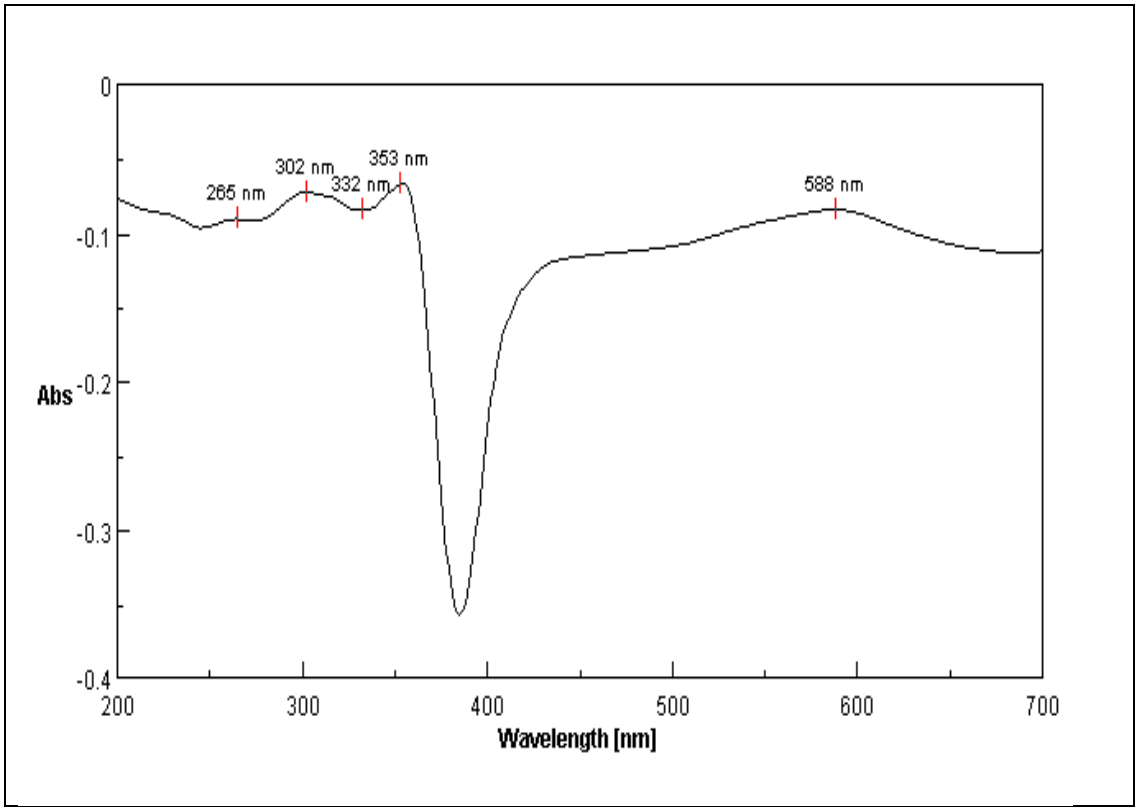
3.1 Graphs Of UV-Visible Spectroscopy



Graph 1: Showing UV-Visible Spectra of Pierre Cardin after one hour in open system



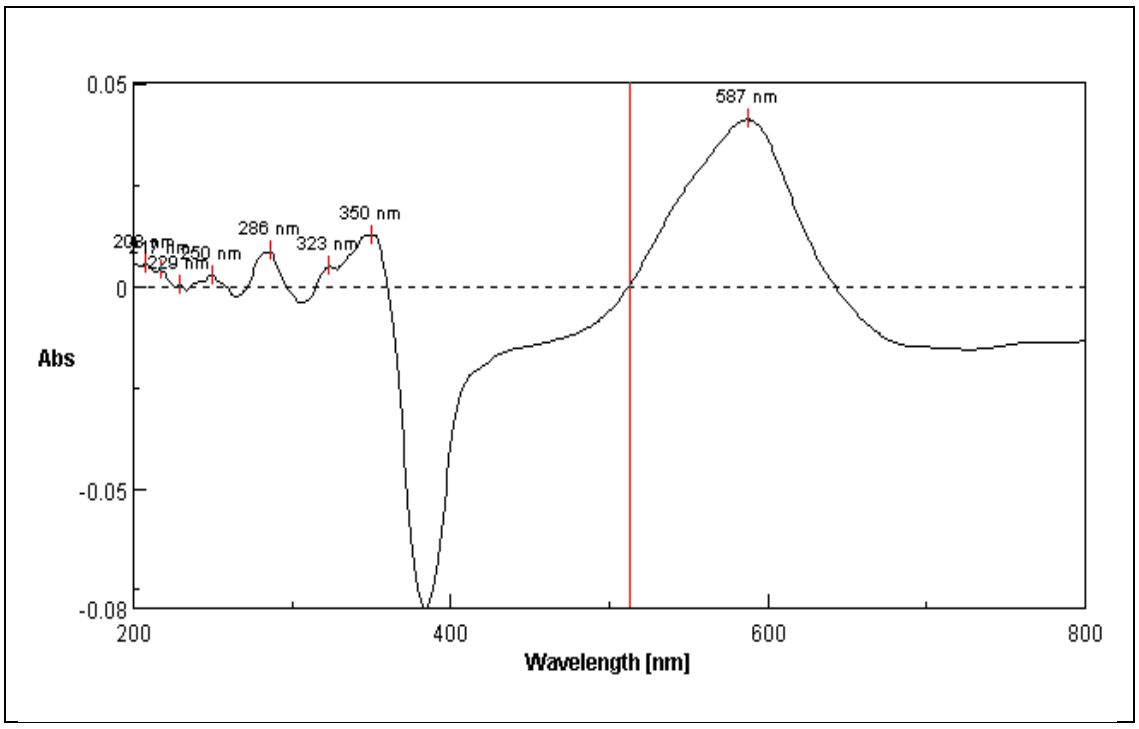
Graph 2: Showing UV-visible spectra of Pierre Cardin after one month in an open system



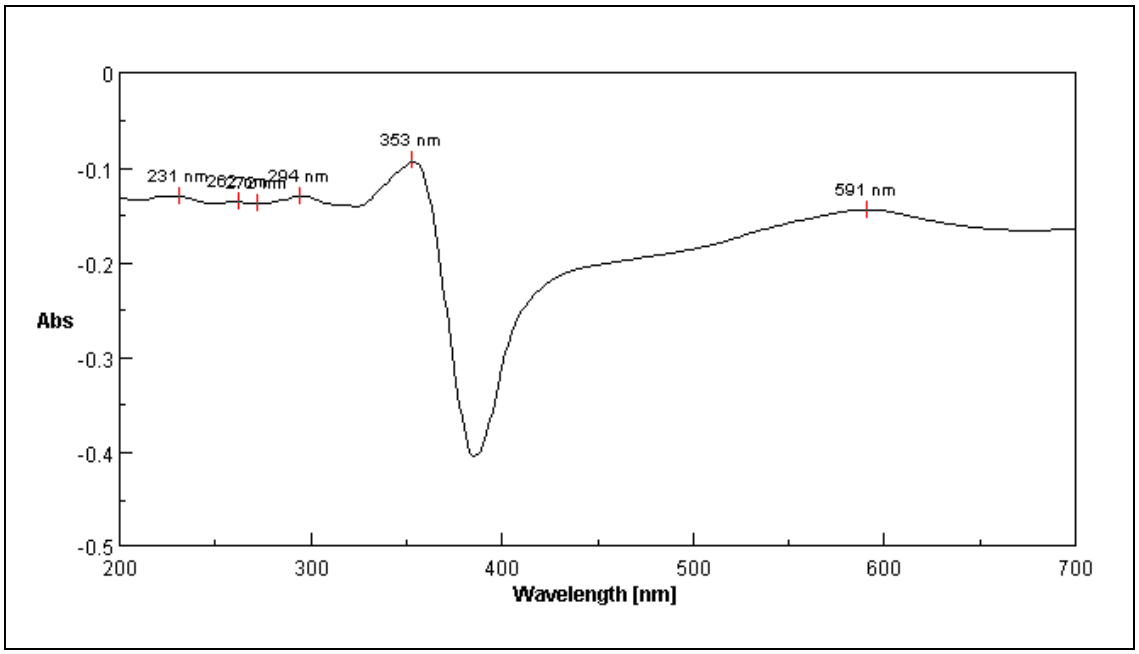
Graph 3: Showing UV-visible spectra of Pierre Cardin after two months in an open system.

FOR TLC:

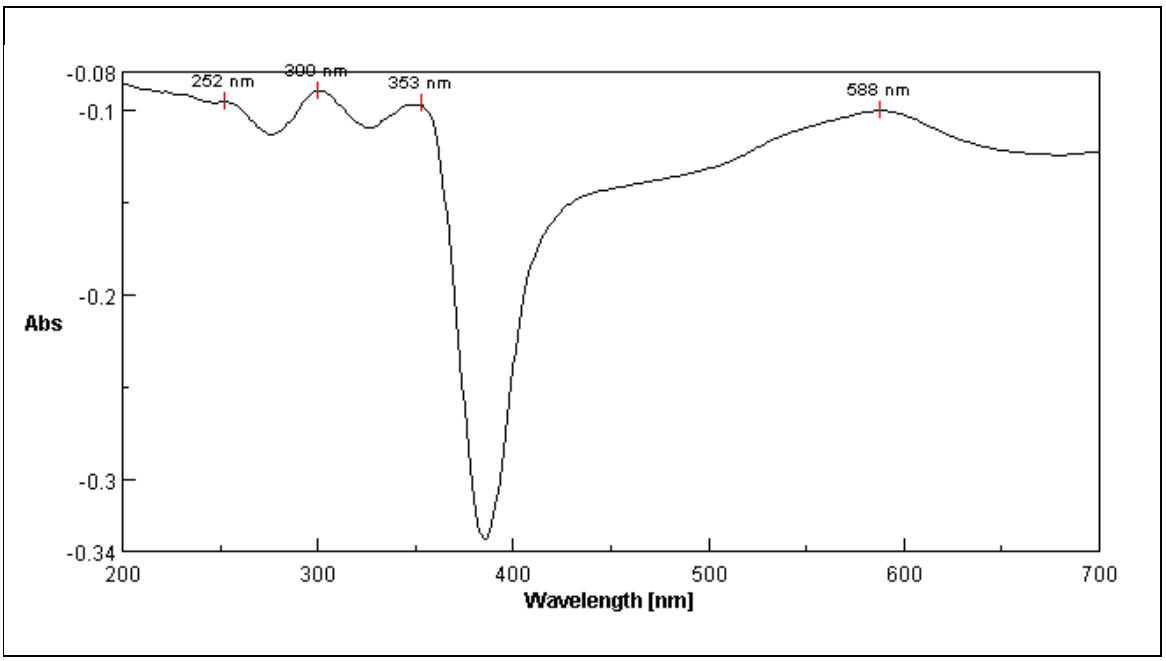
NAME OF PEN	COLOUR OF SPOT	Rf VALUE
Pierre Cardin	**Blue**	**0.7636**



Graph 4: UV-visible spectra of Cello Gripper after one hour in an open system.



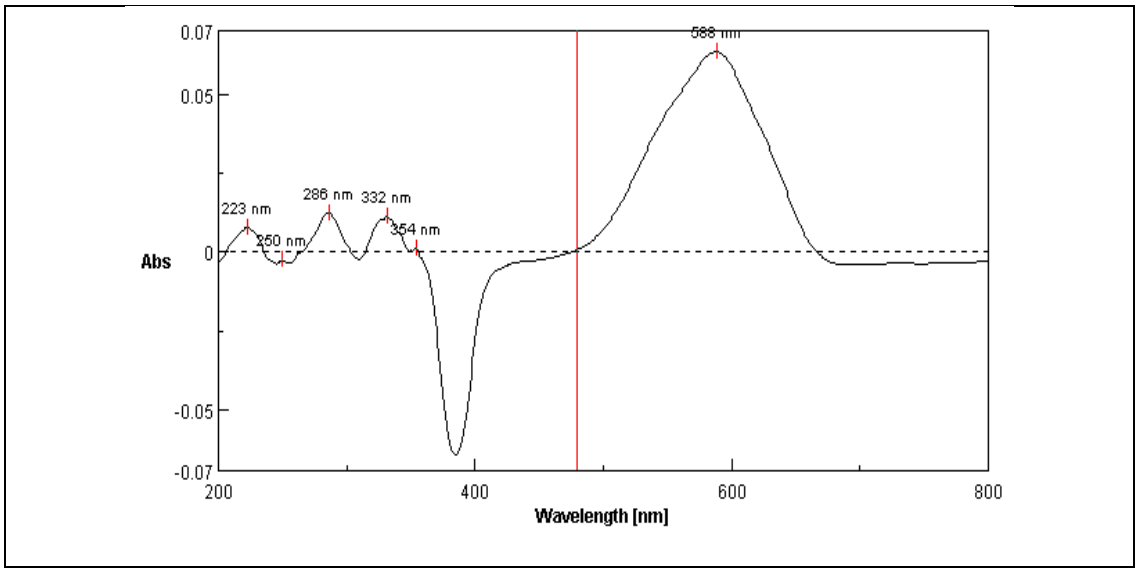
Graph 5: UV-visible spectra of Cello Gripper after one month in an open system.



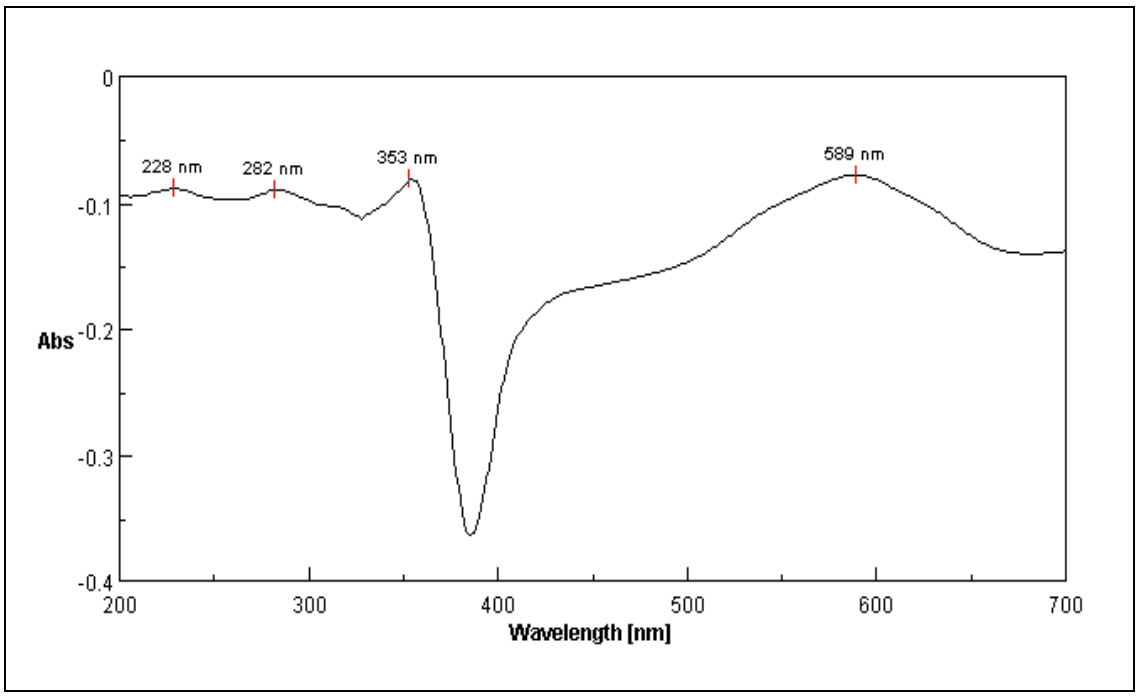
Graph 6: Showing UV-visible spectra of Cello Gripper after two months in the open system

FOR TLC:

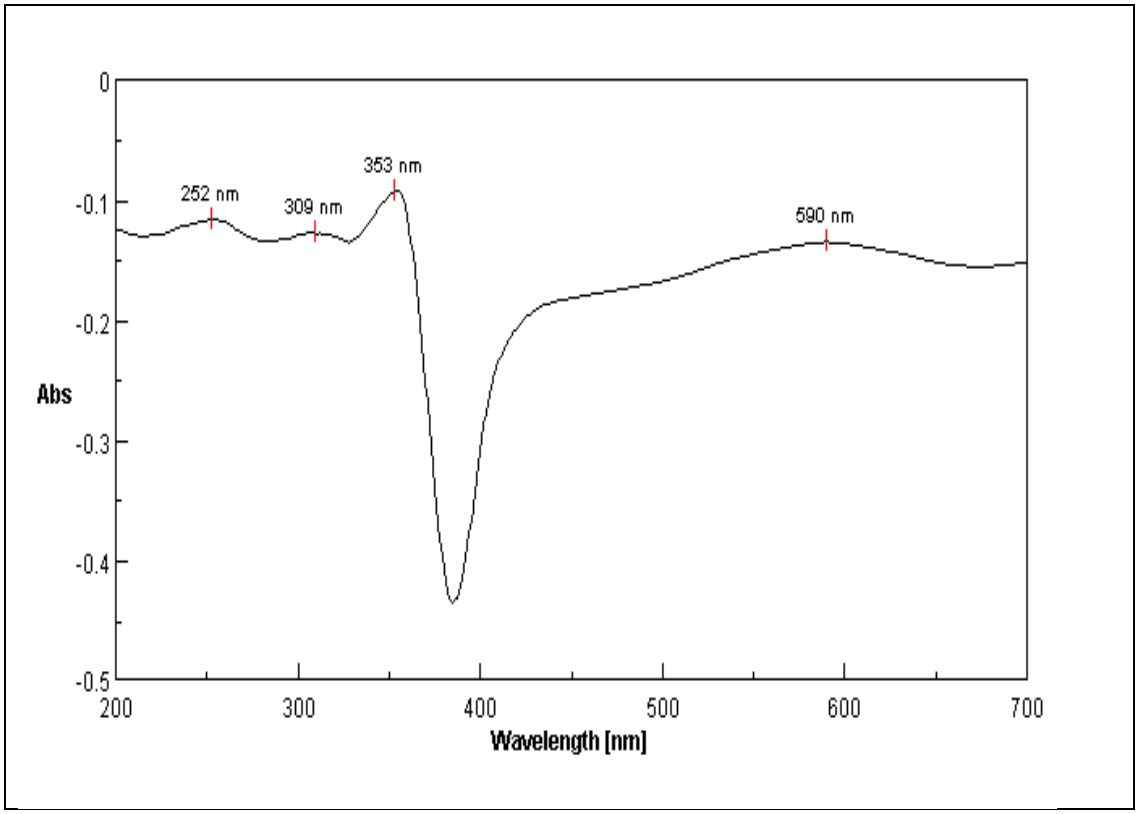
NAME OF PEN	COLOUR OF SPOT	Rf VALUE
Cello Gripper	**Blue**	**0.7636**



Graph 7: UV-visible spectra of Cello Jet-Ace after one hour in an open system.



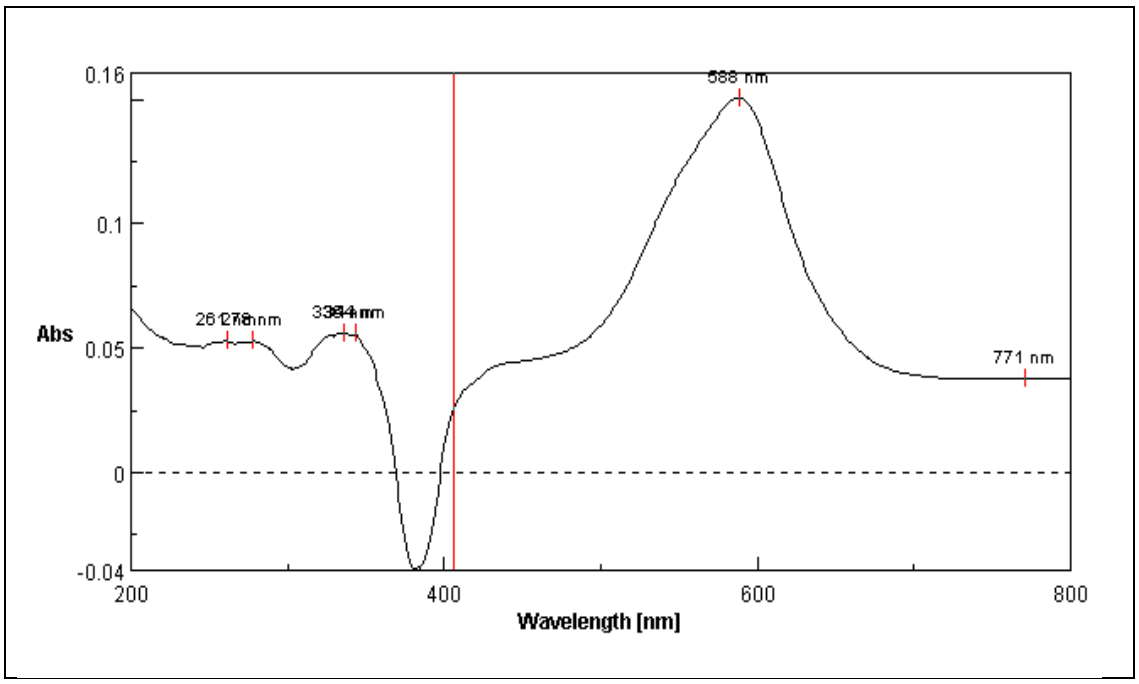
Graph 8: UV-visible spectra of Cello Jet-Ace after one month in an open system.



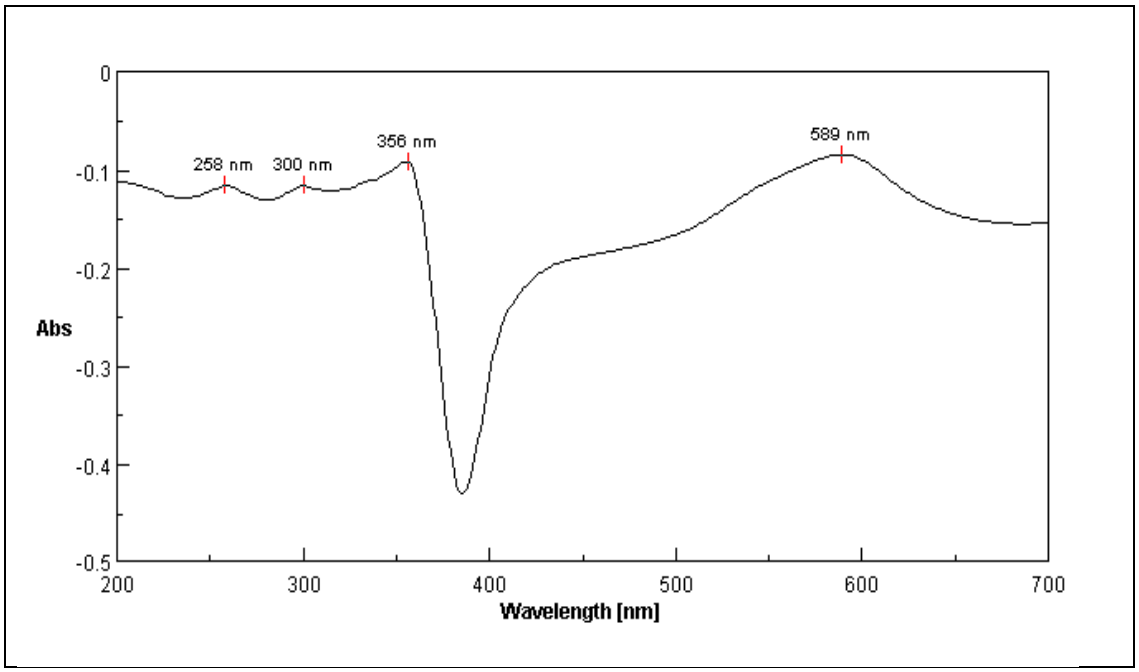
Graph 9:Showing UV-Visible spectra of Cello Jet-Ace after two months in an open system.

FOR TLC:

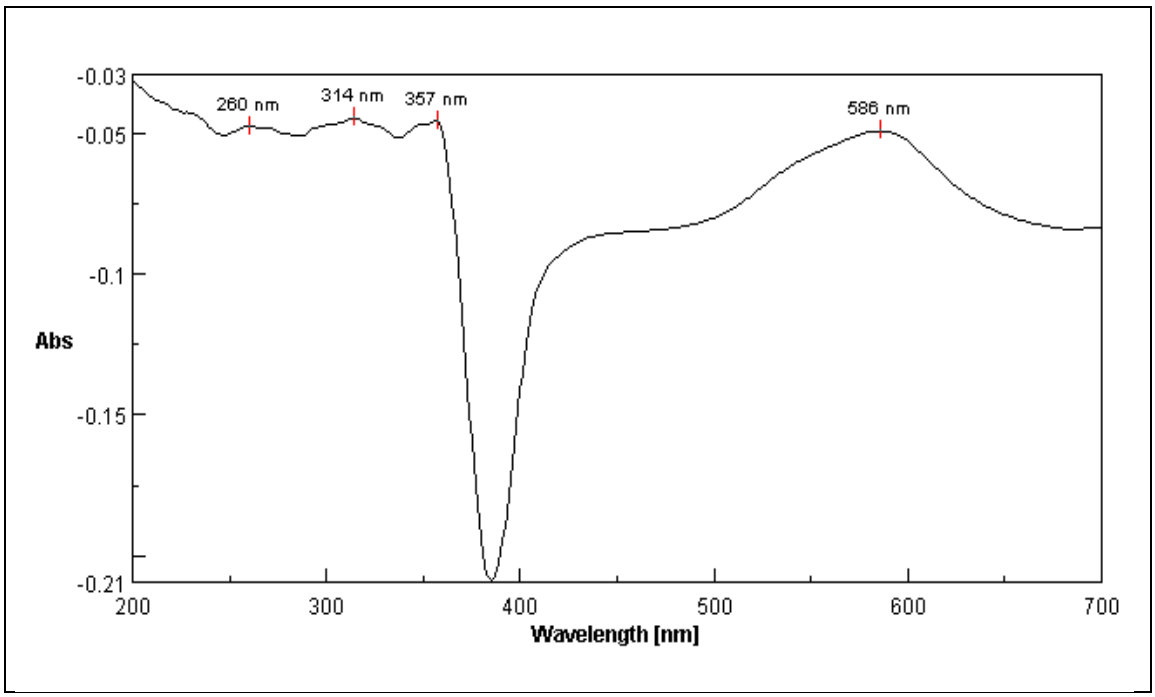
NAME OF PEN	COLOUR OF SPOT	Rf VALUE
Cello jet ace	Purple Blue	0.7727 0.7181



Graph 10:Showing UV-visible spectra of Classmate UVO Ball after one hour in an open system.



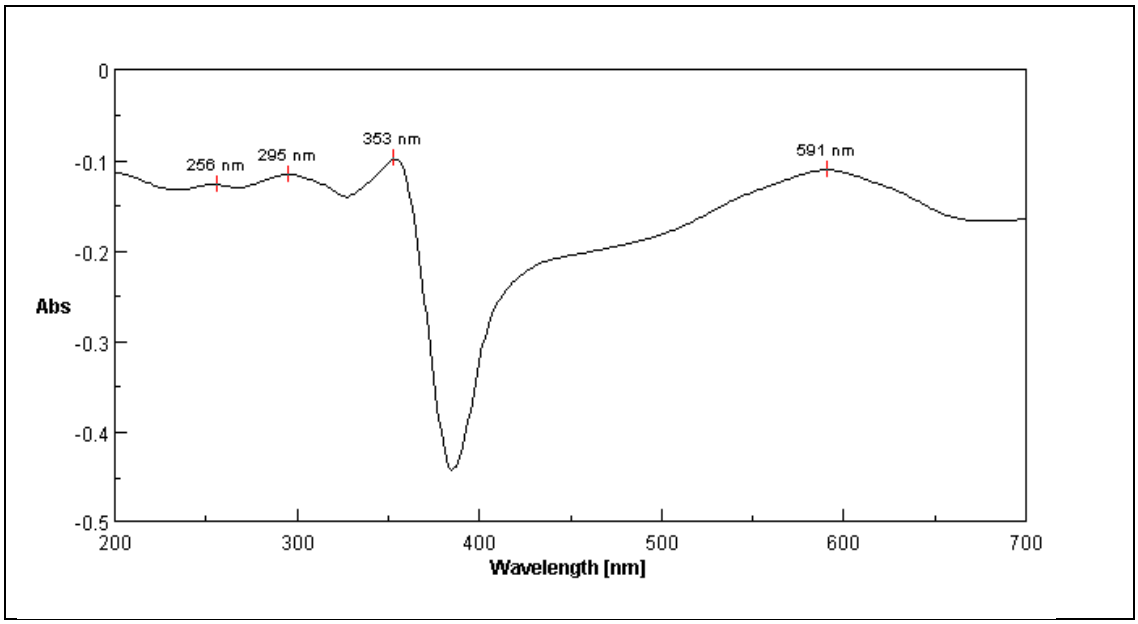
Graph 11:Showing UV-Visible spectra of Classmate UVO Ball after one month in an open system.



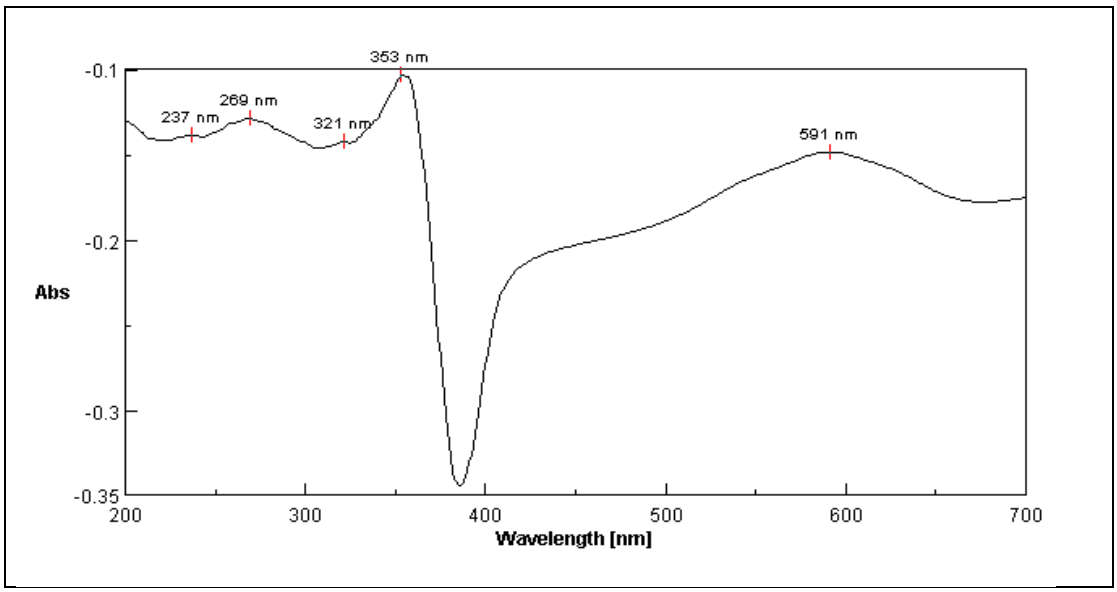
Graph 12:Showing UV-Visible spectra of Classmate UVO Ball after two months in an open system

FOR TLC:

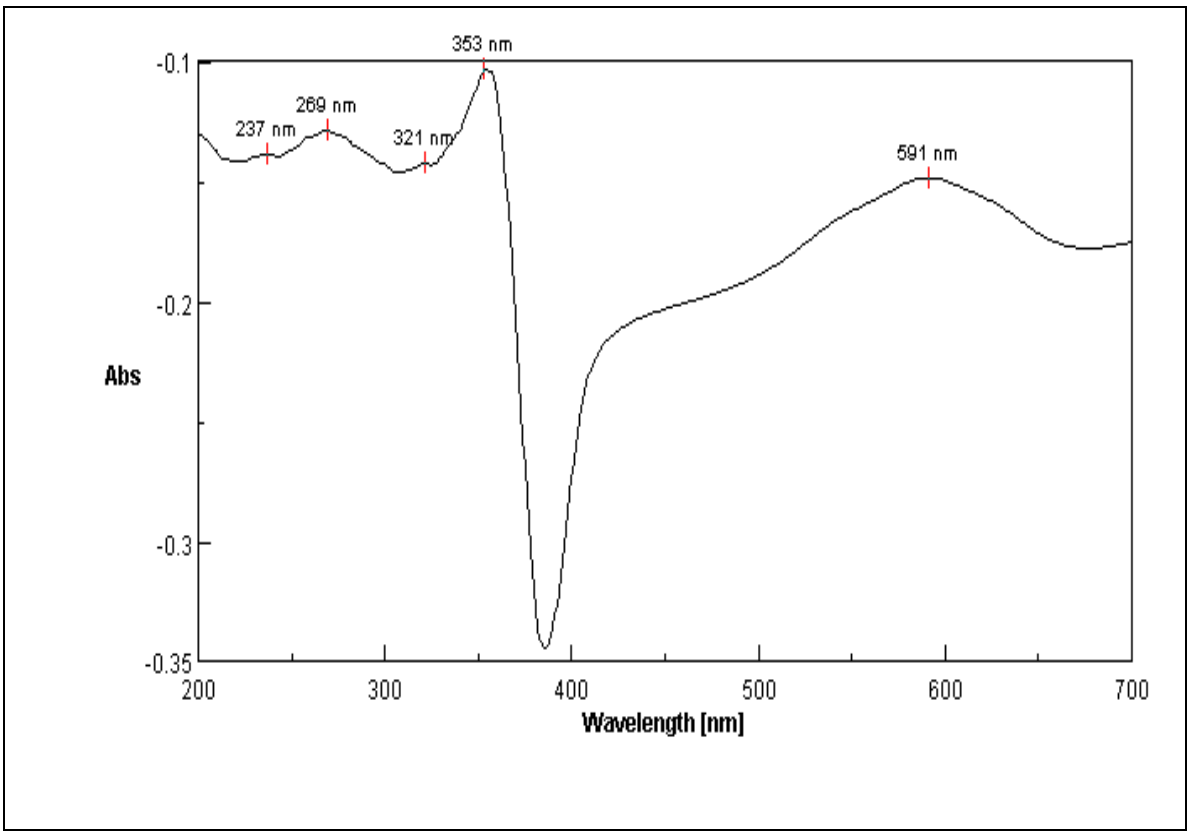
NAME OF PEN	COLOUR OF SPOT	Rf VALUE
Classmate uvo ball	Purple	0.81



Graph 13:Showing UV-Visible spectra of Cello Maxwriter after one hour in an open system.



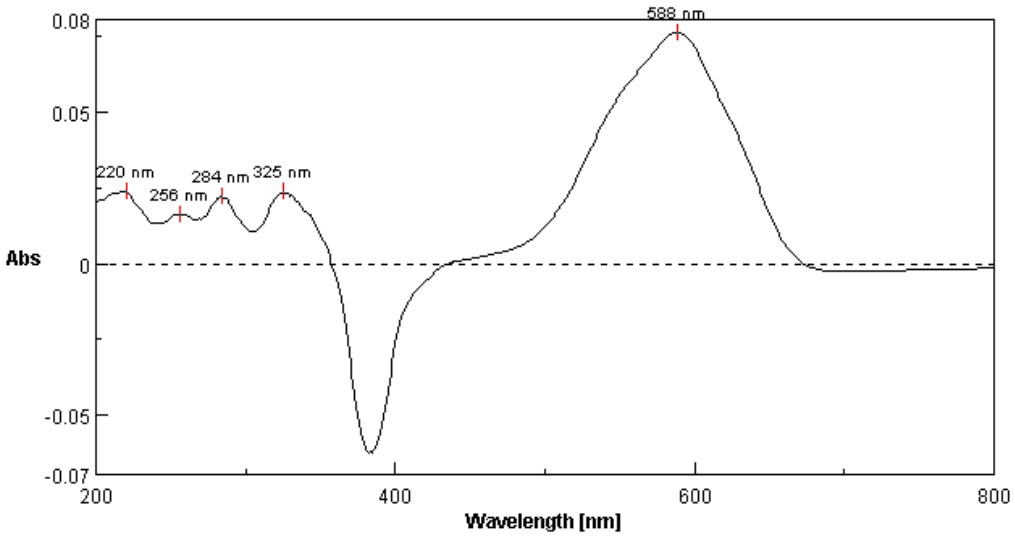
Graph 14:Showing UV-Visible spectra of Cello Maxwriter after one month in an open system.



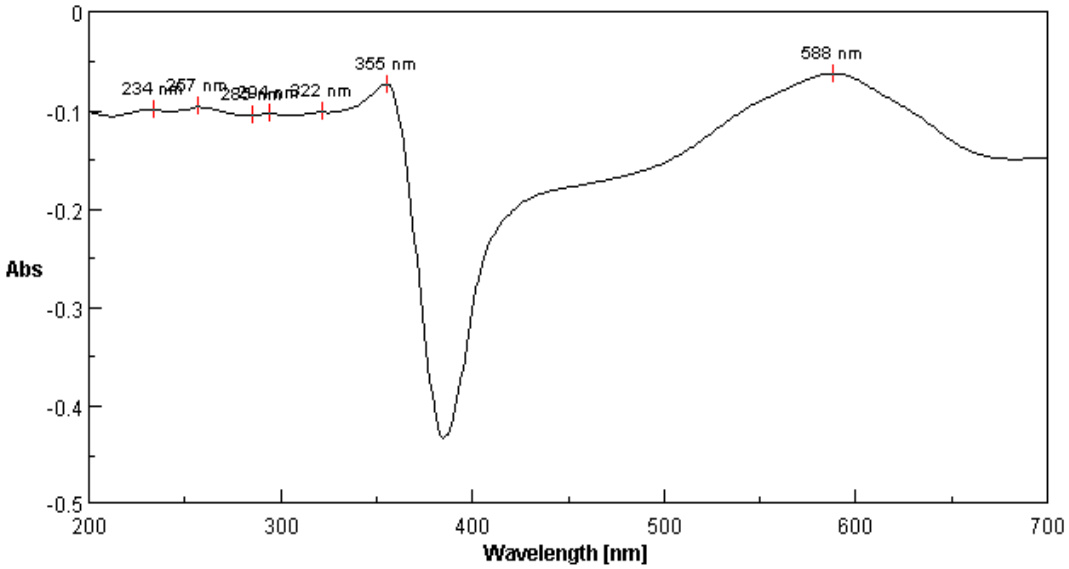
Graph 15:Showing UV-Visible spectra of Cello Maxwriter after two month in open system.

FOR TLC:

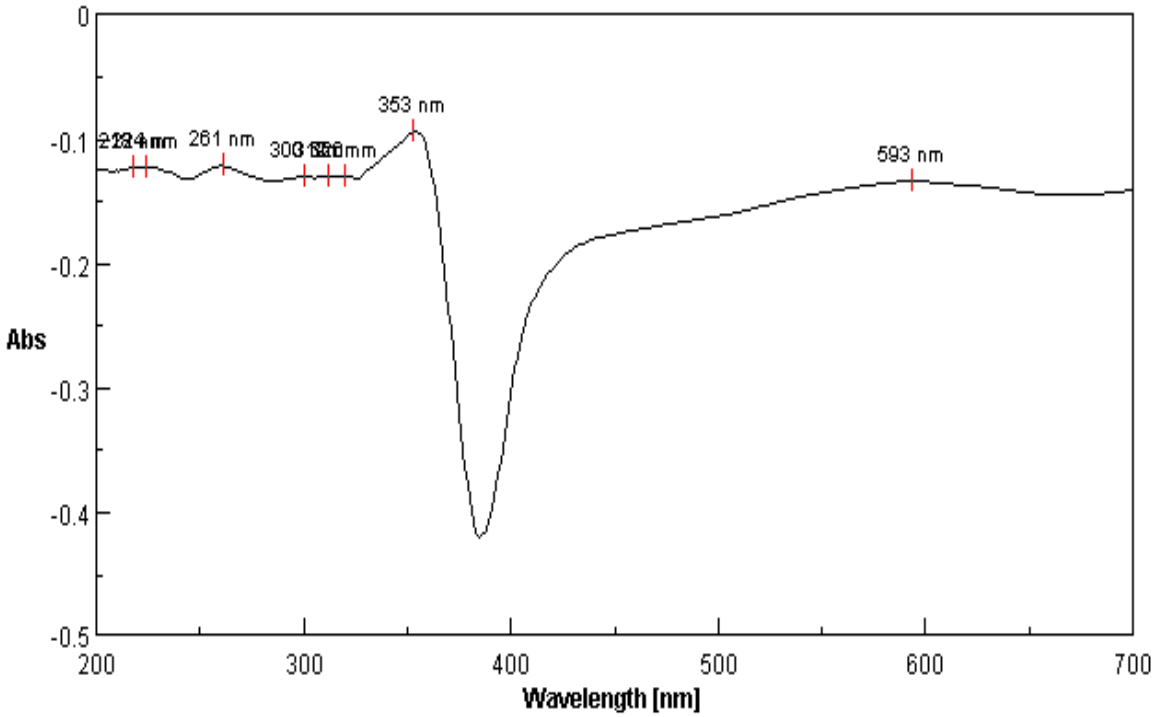
NAME OF PEN	COLOUR OF SPOT	Rf VALUE
Cello max writer	Purple Blue	0.7909 0.7454



Graph 16: Showing UV-Visible spectra of 2K Add Gel after one hour in open system



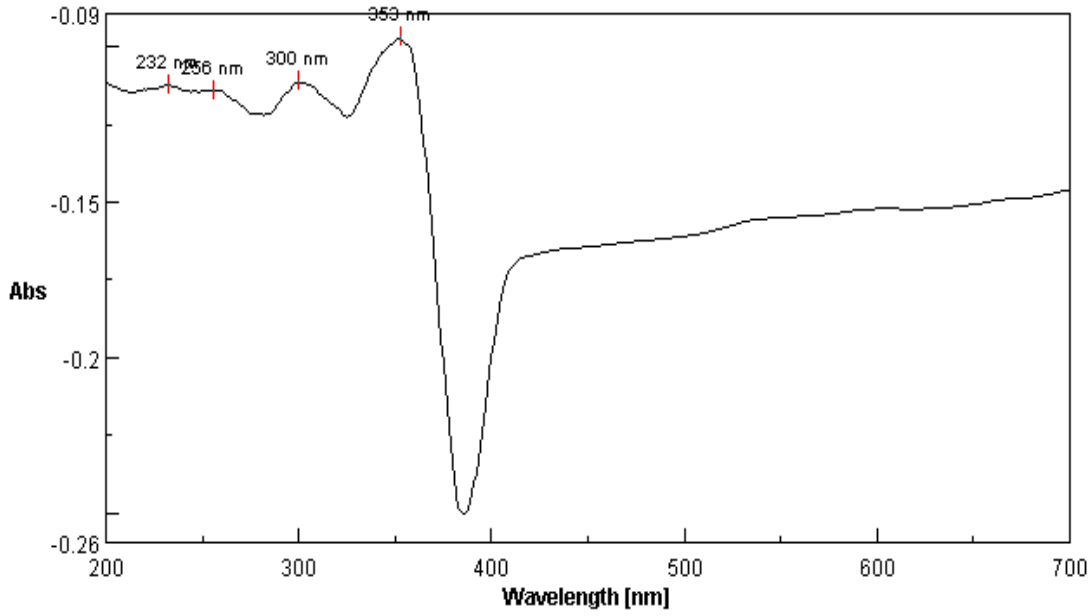
Graph 17: Showing UV-Visible spectra of 2K Add Gel after one month in open system.



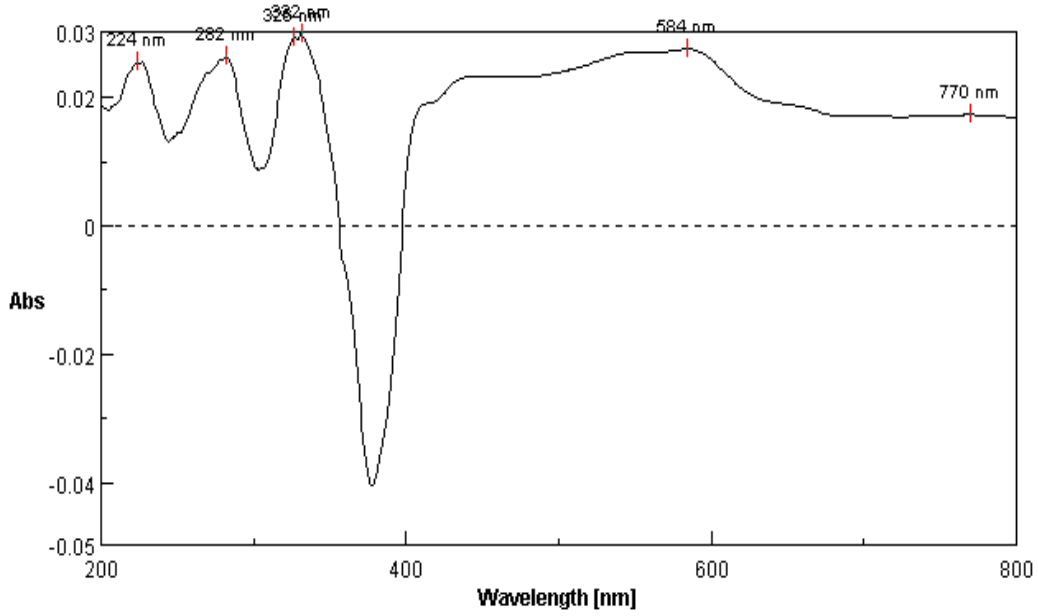
Graph 18:Showing UV-Visible spectra of 2K Add Gel after two month in open system.

FOR TLC:

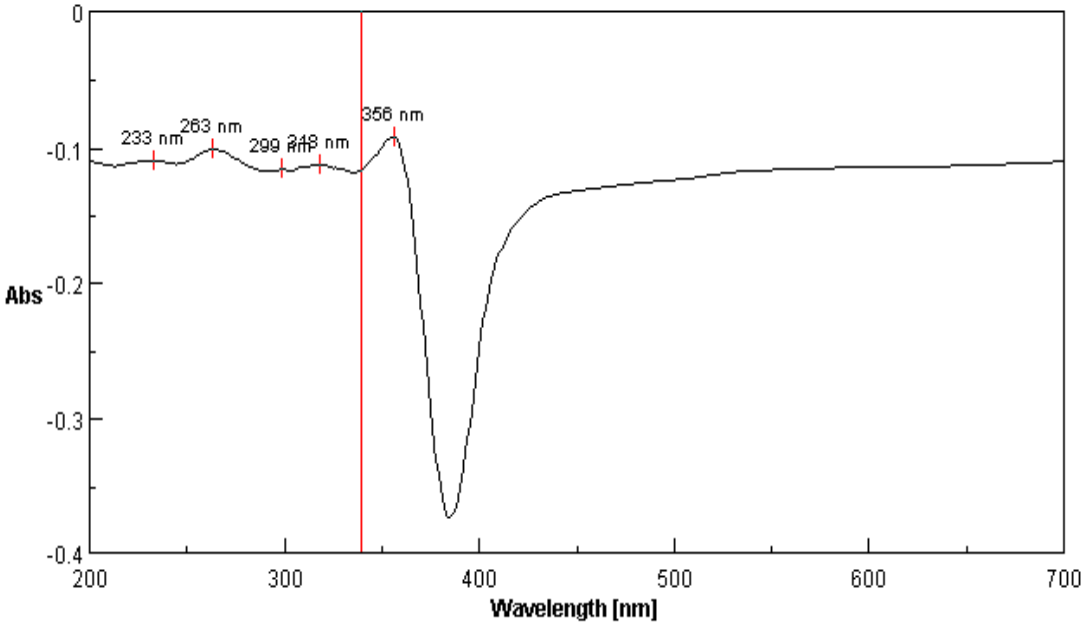
NAME OF PEN	COLOUR OF SPOT	Rf VALUE
2k Add gel	Purple Blue	0.7964 0.7876



Graph 19:Showing UV-Visible spectra of Flair Osmium Gel after one month open system.



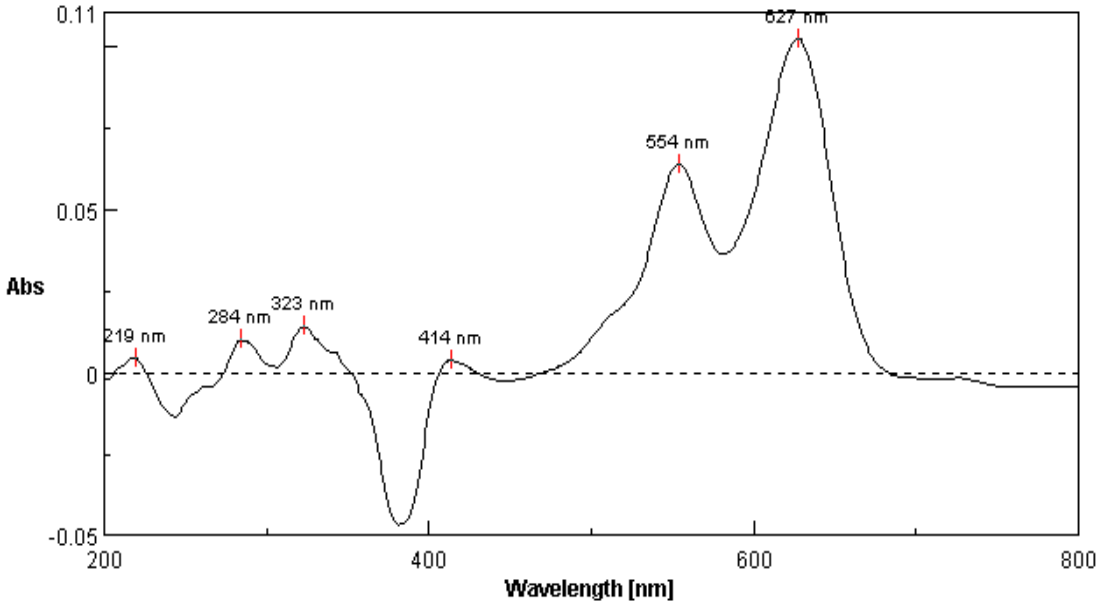
Graph 20: Showing UV-Visible spectra of Flair Osmium Gel after one hour open system.



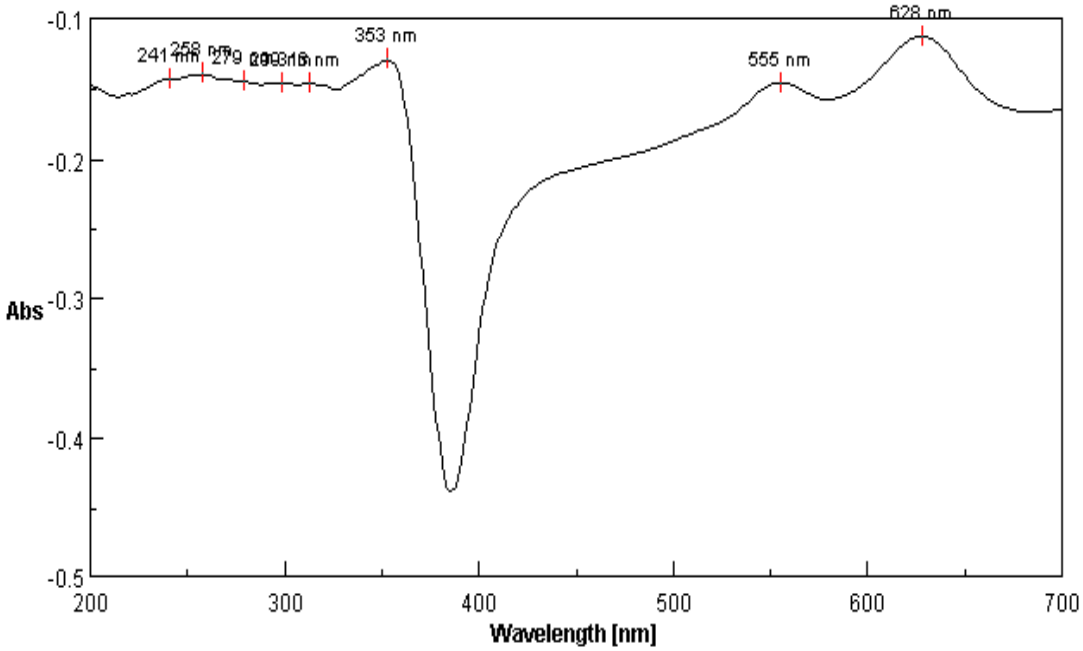
Graph 21:Showing UV-Visible spectra of Flair Osmium Gel after two month open system.

FOR TLC:

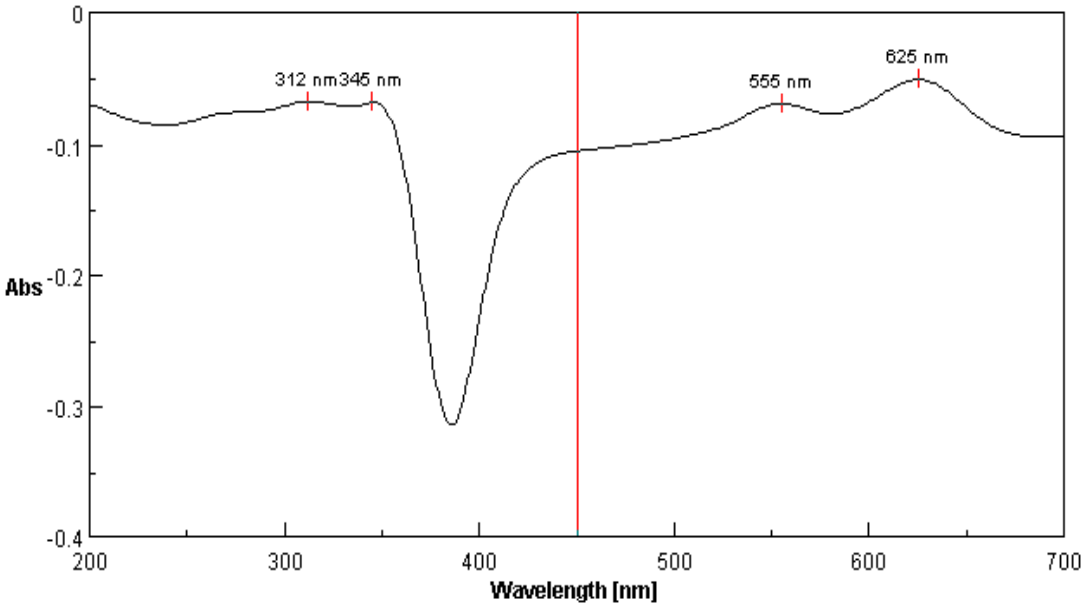
No TLC run was observed for Flair Osmium Gel pen ink.



Graph 22:Showing UV-Visible spectra of Montex Activa Gel after one hour open system



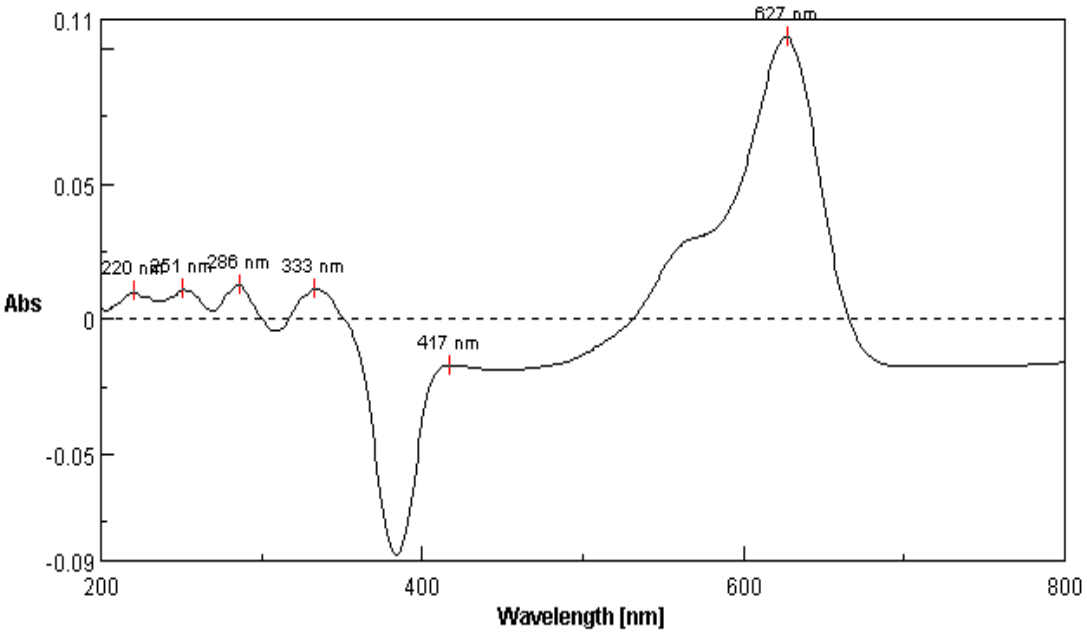
Graph 23:Showing UV-Visible spectra of Montex Activa Gel after one month open system.



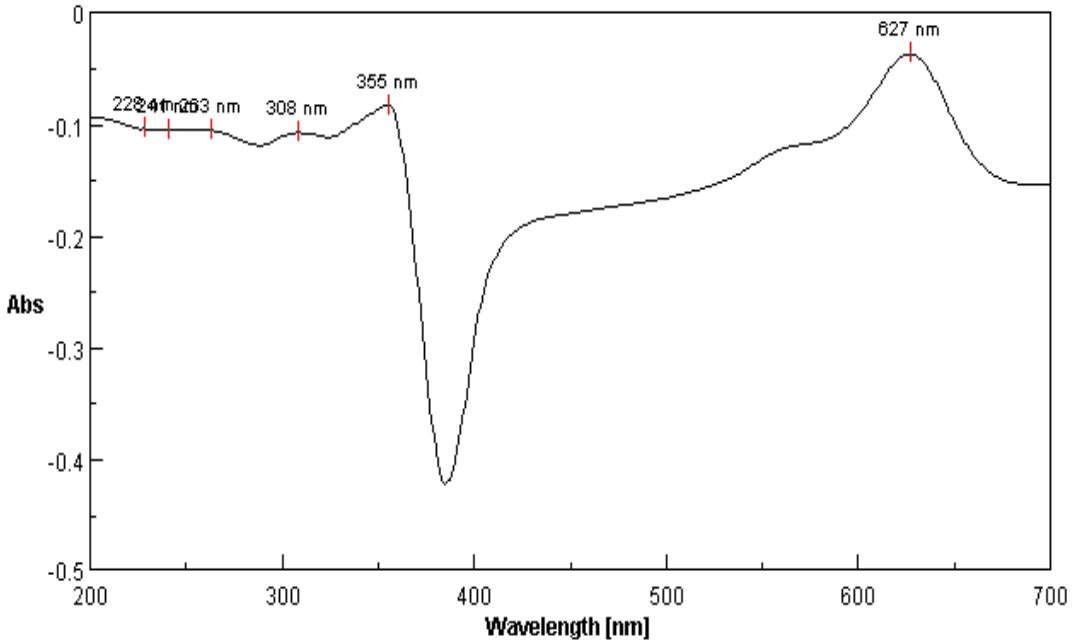
Graph 24:Showing UV-Visible spectra of Montex Activa Gel after two month open system.

FOR TLC:

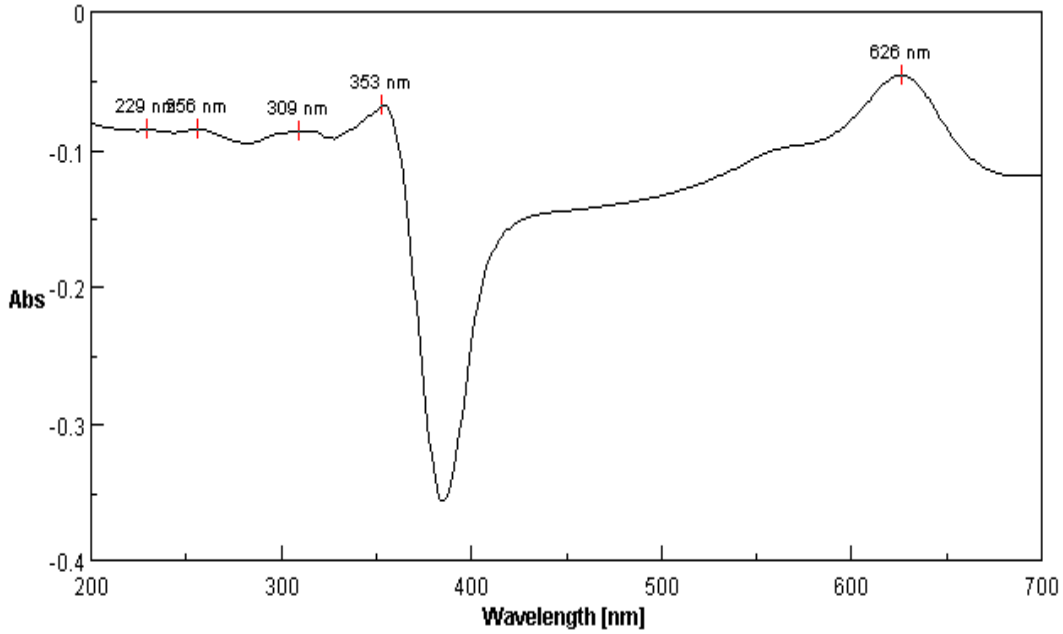
NAME OF PEN	COLOUR OF SPOT	Rf VALUE
Montex activa gel	Red Blue	0.8230 0.7876



Graph 25:Showing UV-Visible spectra of Add Gel Achiever after one hour open system



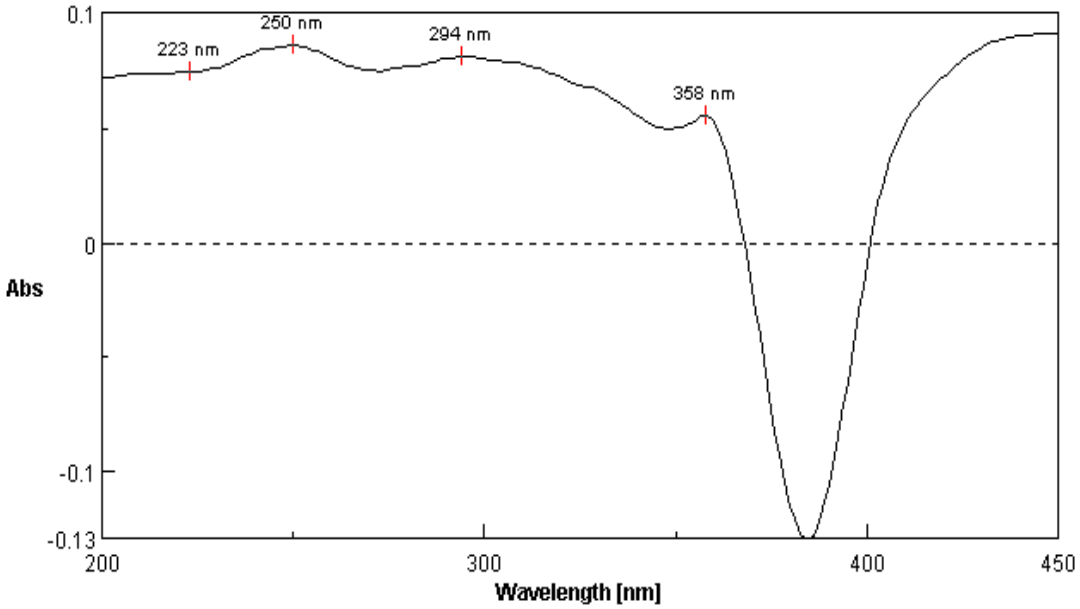
Graph 26: Showing UV-Visible spectra of Add Gel Achiever after one month



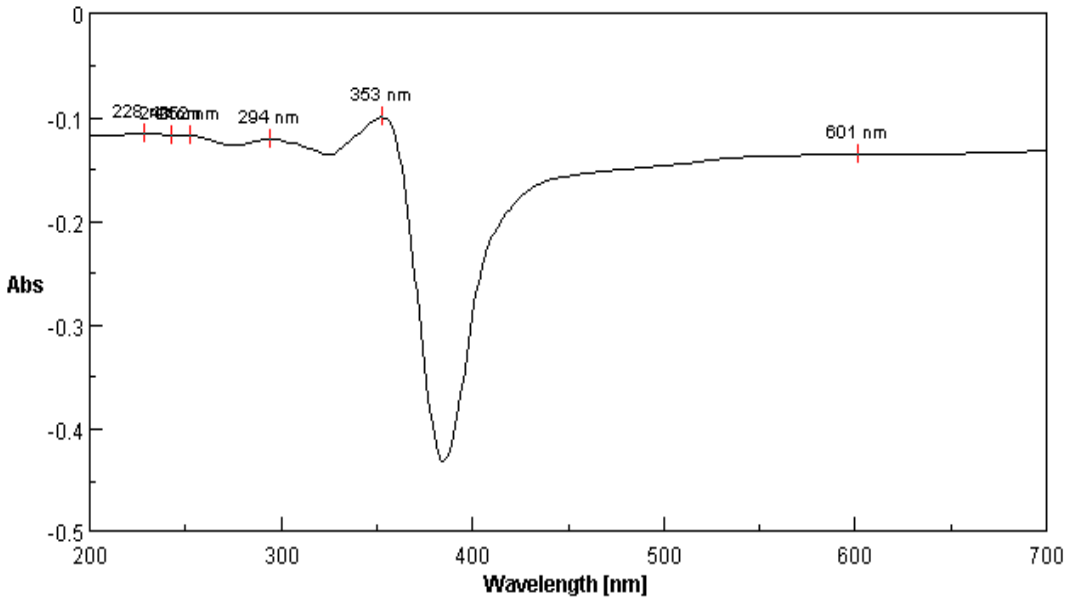
Graph 27Showing UV-Visible spectra of Add Gel Achiever after two month

FOR TLC:

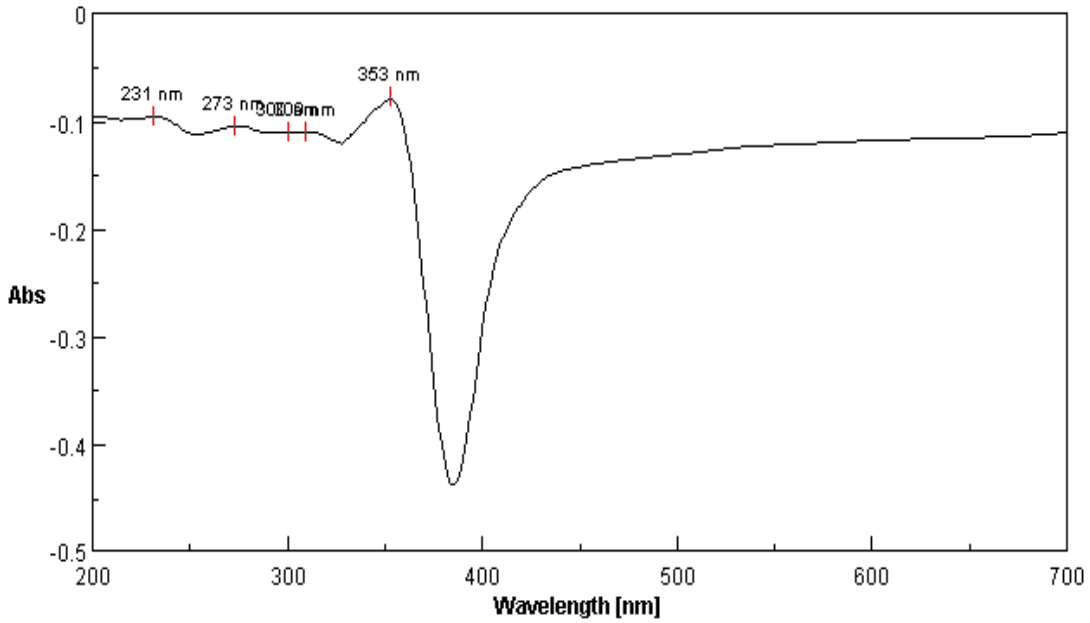
NAME OF PEN	COLOUR OF SPOT	Rf VALUE
Add gel achiever	Purple Blue	0.8053 0.7876



Graph 28: Showing UV-Visible spectra of Classmate Octane after one hour.....open system.



Graph 29: Showing UV-Visible spectra of Classmate Octane after one month.open system.

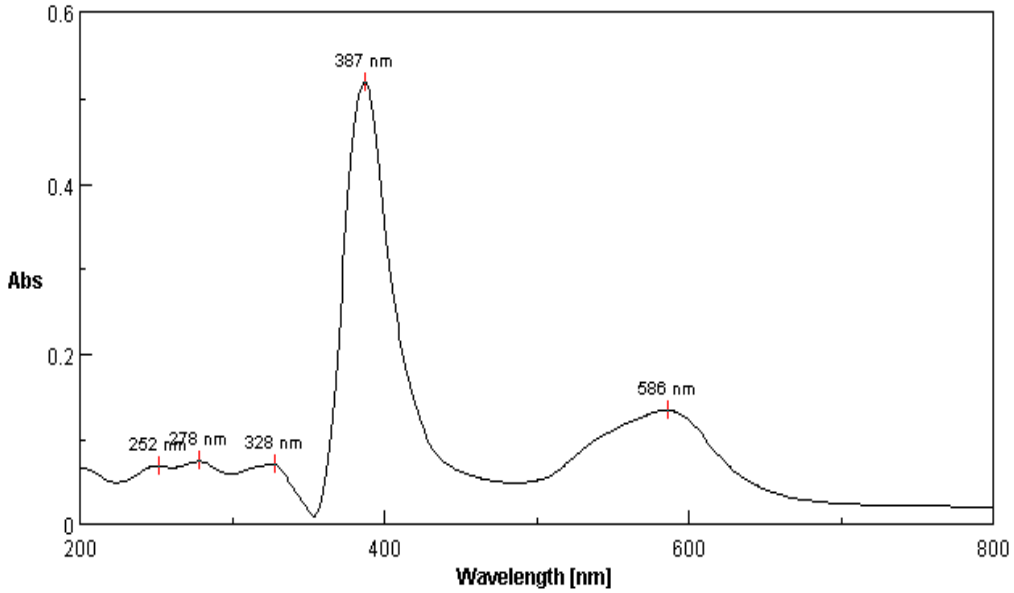


Graph 30: Showing UV-Visible spectra of Classmate Octane after two month..open system.

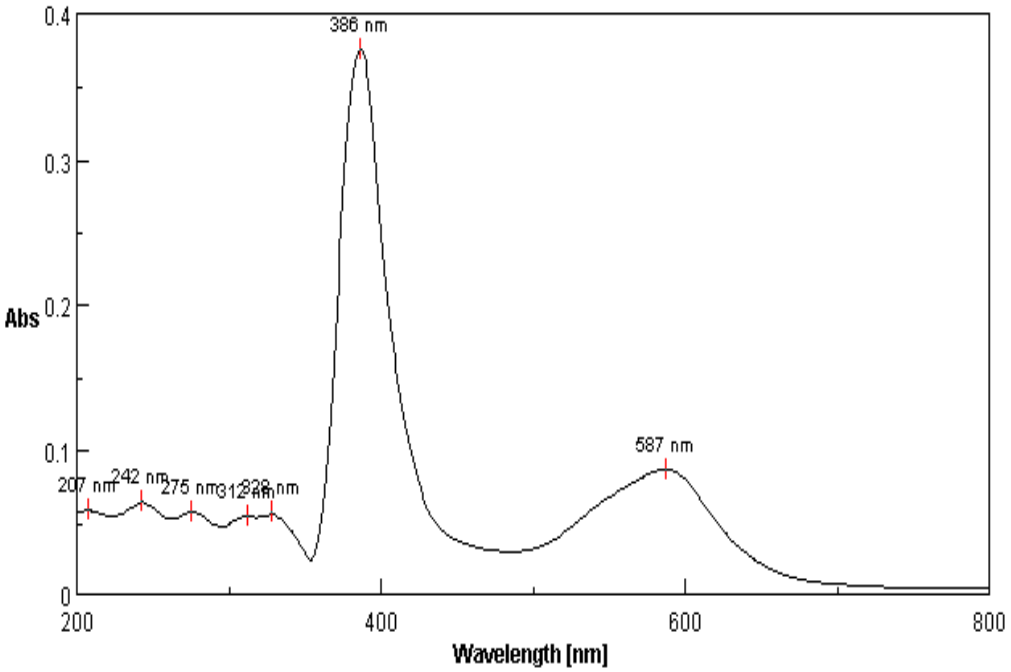
FOR TLC:

No TLC run was observed for Classmate Octane pen ink.

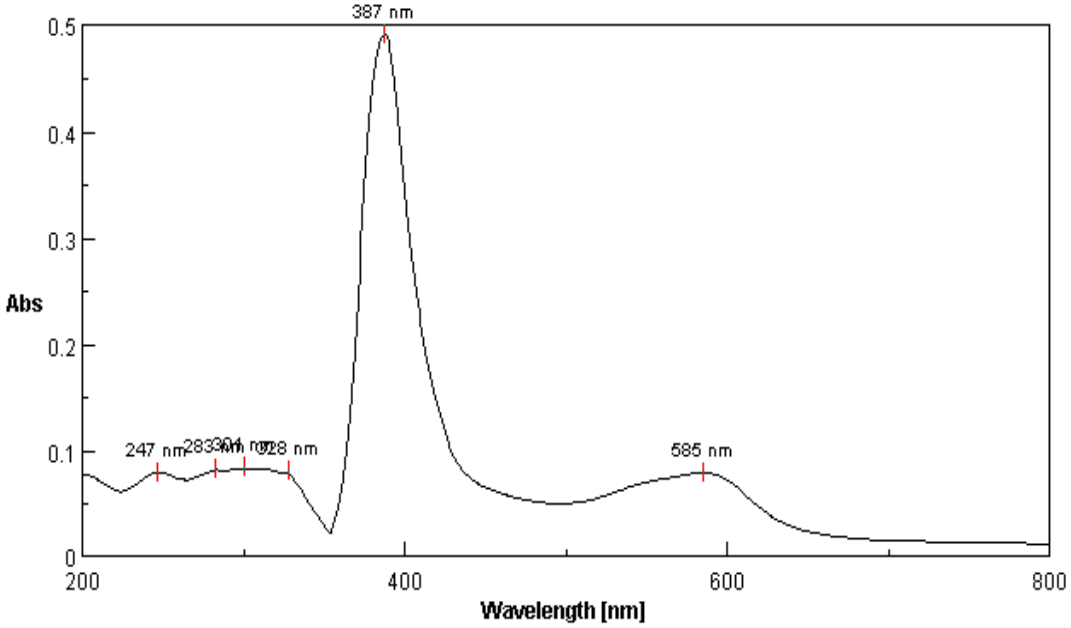
NATURAL SAMPLES



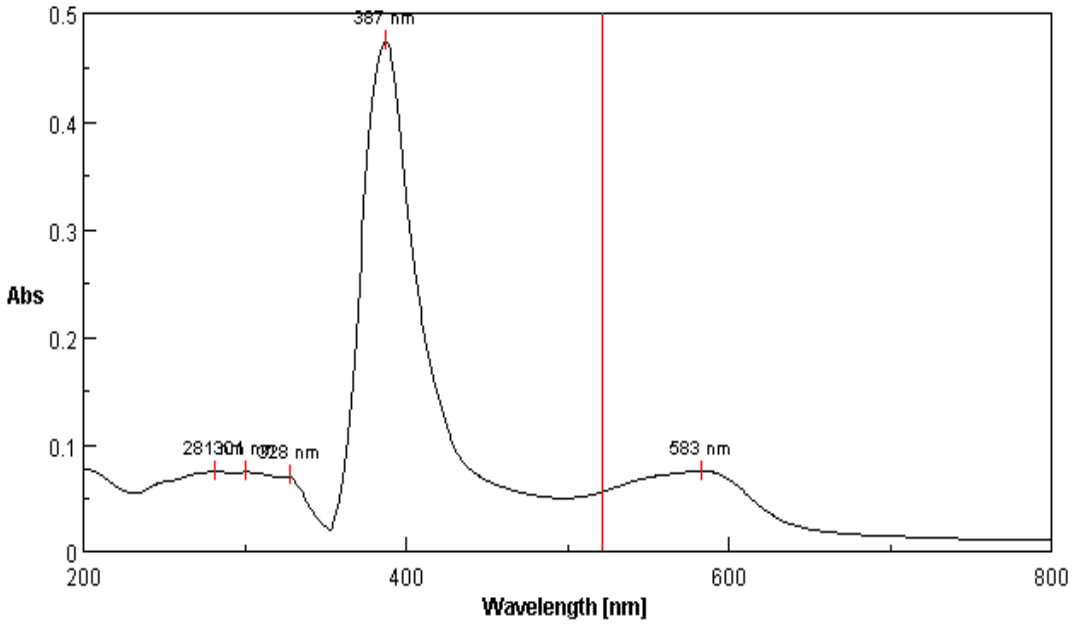
Graph 31Showing UV-Visible spectra of a Natural Sample of the year 2010.



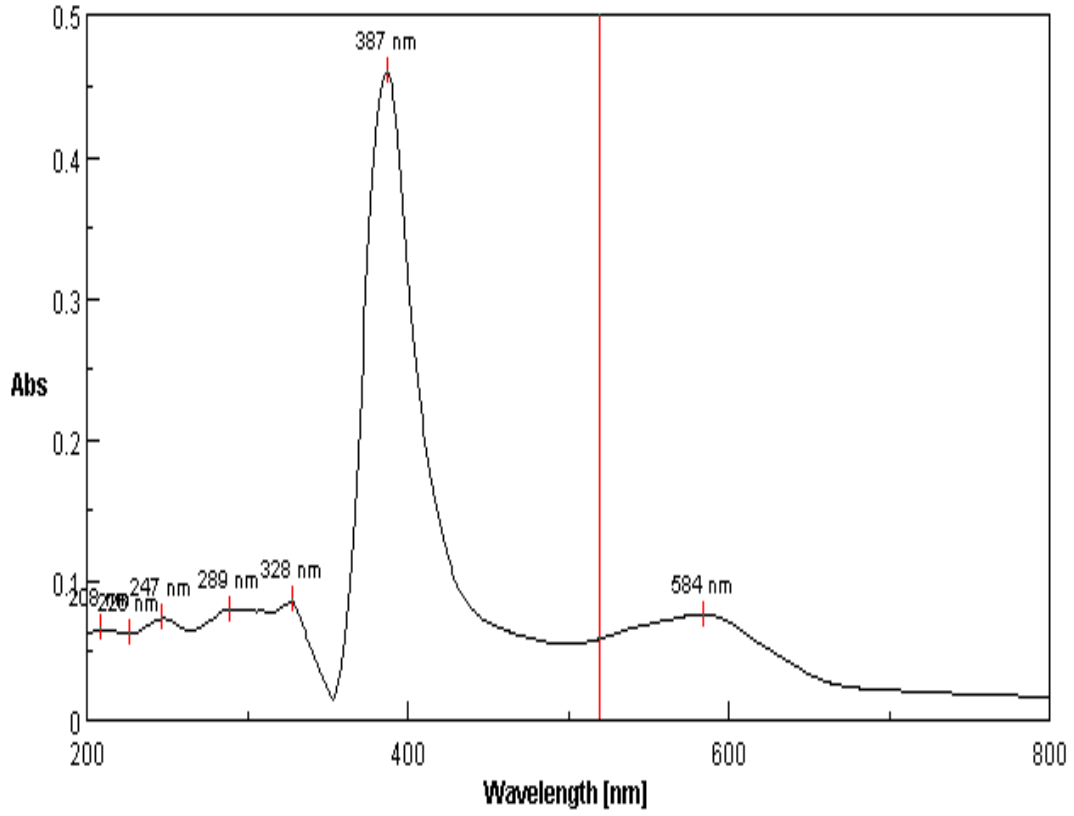
Graph 32: Showing UV-Visible spectra of a Natural Sample of the year 2009.



Graph 33: Showing UV-Visible spectra of a Natural Sample of the year 2008.

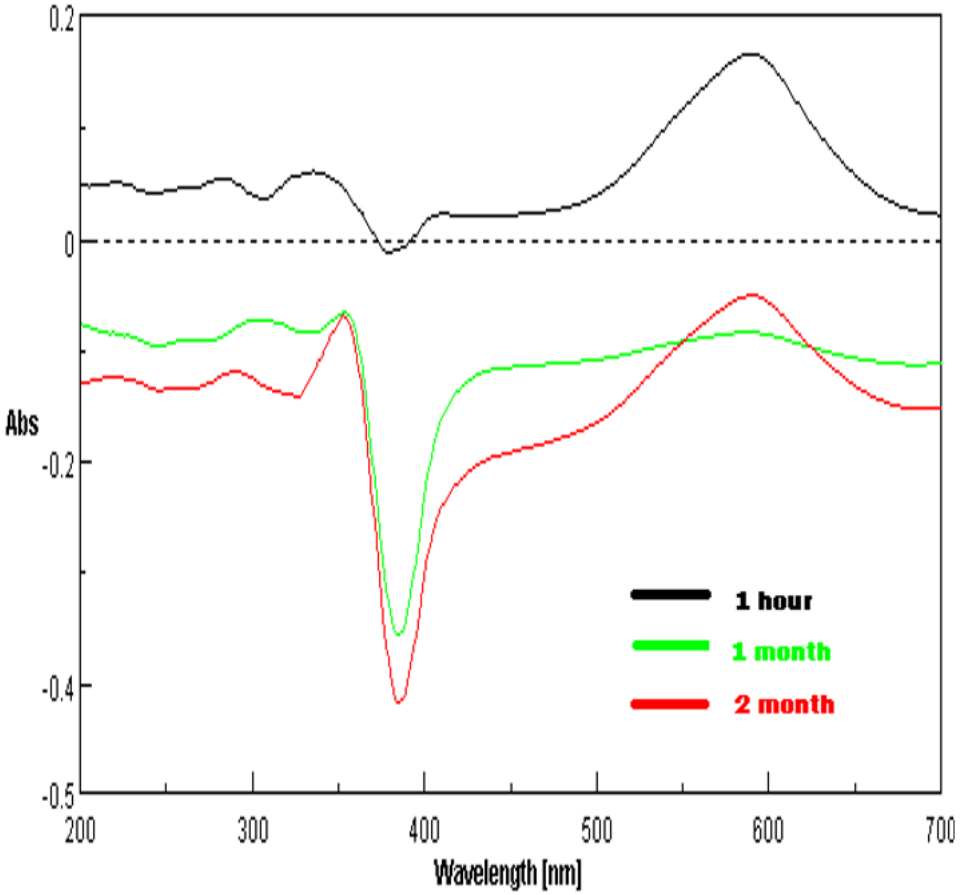


Graph 34: Showing UV-Visible spectra of a Natural Sample of the year 2007.



Graph 35: Showing UV-Visible spectra of a Natural Sample of the year 2006.

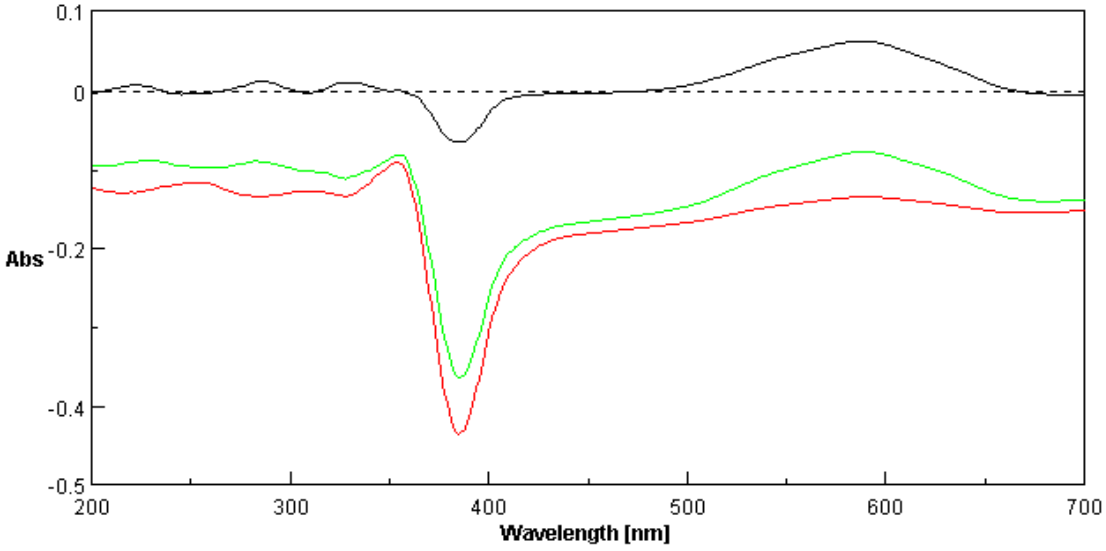
COMPARISON GRAPHS



Graph 36: Showing UV-Visible comparison spectral graph of Pierre Cardin with different time intervals.

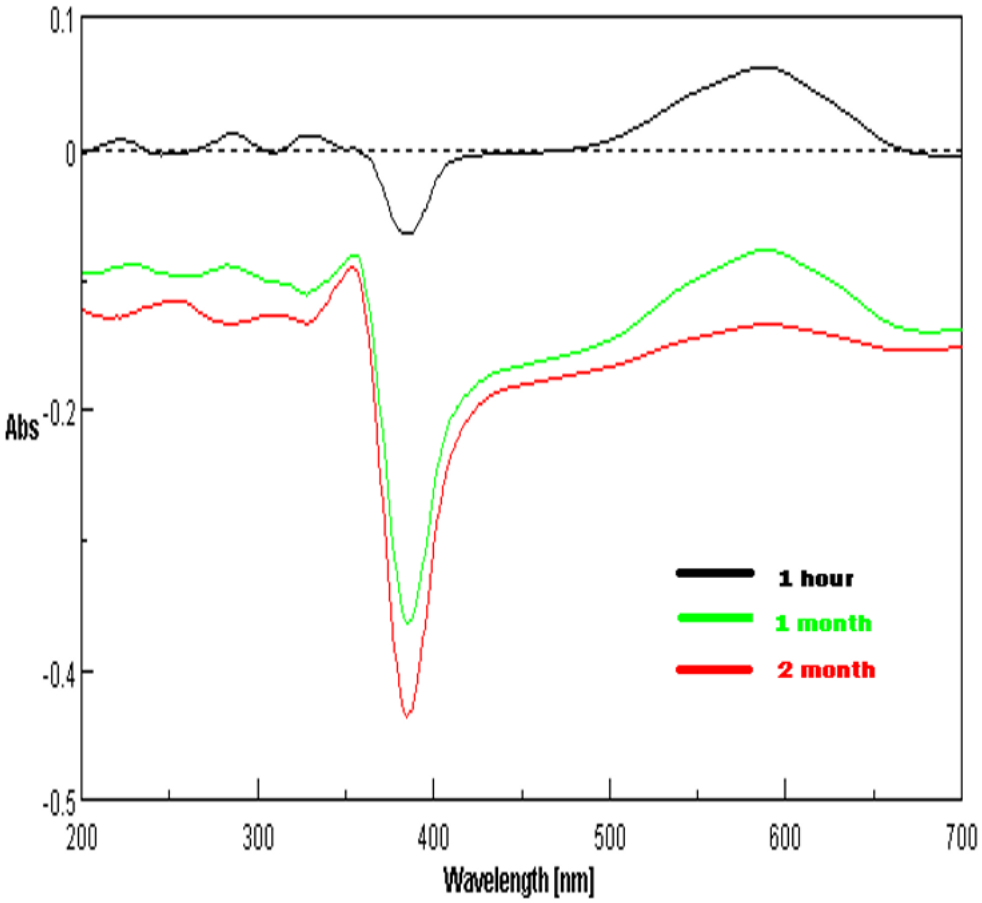
The absorbance observed for samples after one hour, one month, and two months was 0.05, -0.13, and -0.07, respectively.

Wavelength peaks found are 590, 589, and 588 - which show the blue shift due to the color of the ink; 220 – which shows the presence of alkanes in the ink sample; 353, 332, 336, 302 shows the presence of nitrites or ketones.



Graph 37: Showing UV-Visible comparison spectral graph of Cello Gripper with different time intervals.

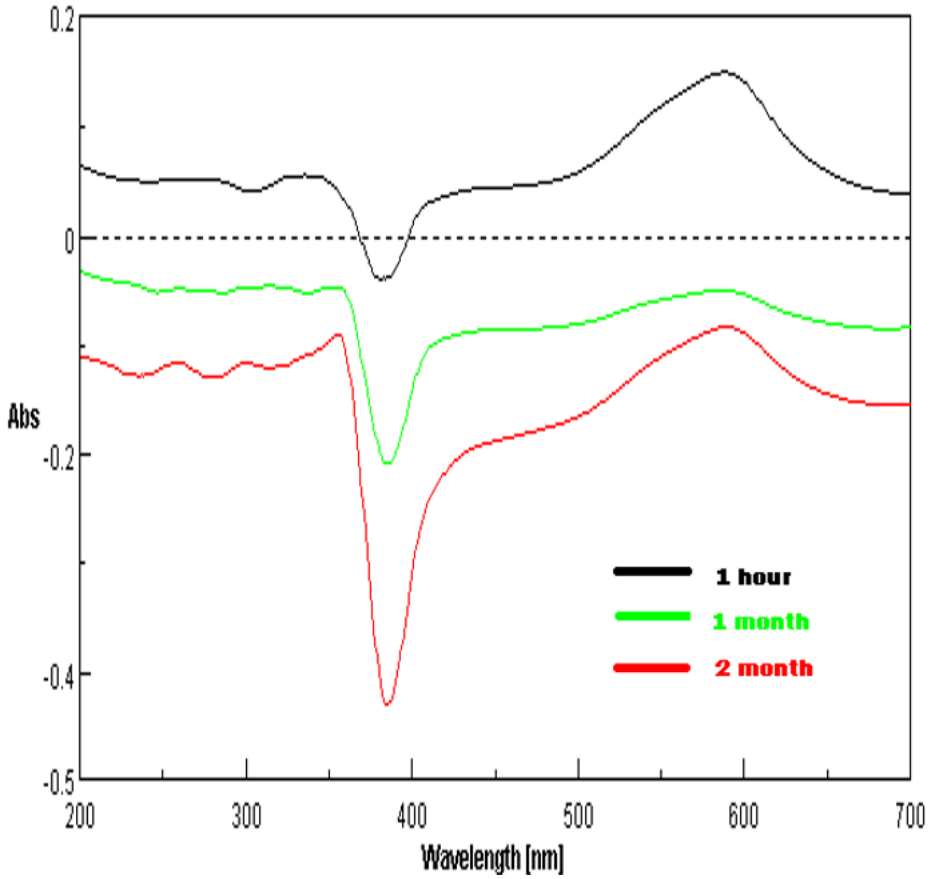
Absorbance observed is 0.001, -0.13, and -0.08 for samples after one hour, one month, and two months respectively.Wavelength peaks found are 587, 588, 591 - shows the blue shift due to the color of ink; 250 – shows the presence of alcohol (isobutanol or propanol) or halocarbons (1, 2-dichloromethane) or alkanes. In the ink sample; 350 – indicates the presence of ketones (5-methylexanone) or aromatic hydrocarbons (1, 2-dichlorobenzene, or 1, 2, 4-trichlorobenzene); 320- indicates the presence of nitrites; 294 – shows that azo may be present in the sample.



Graph 38: Showing UV-Visible comparison spectral graph of Cello Jet-Ace with different time intervals.

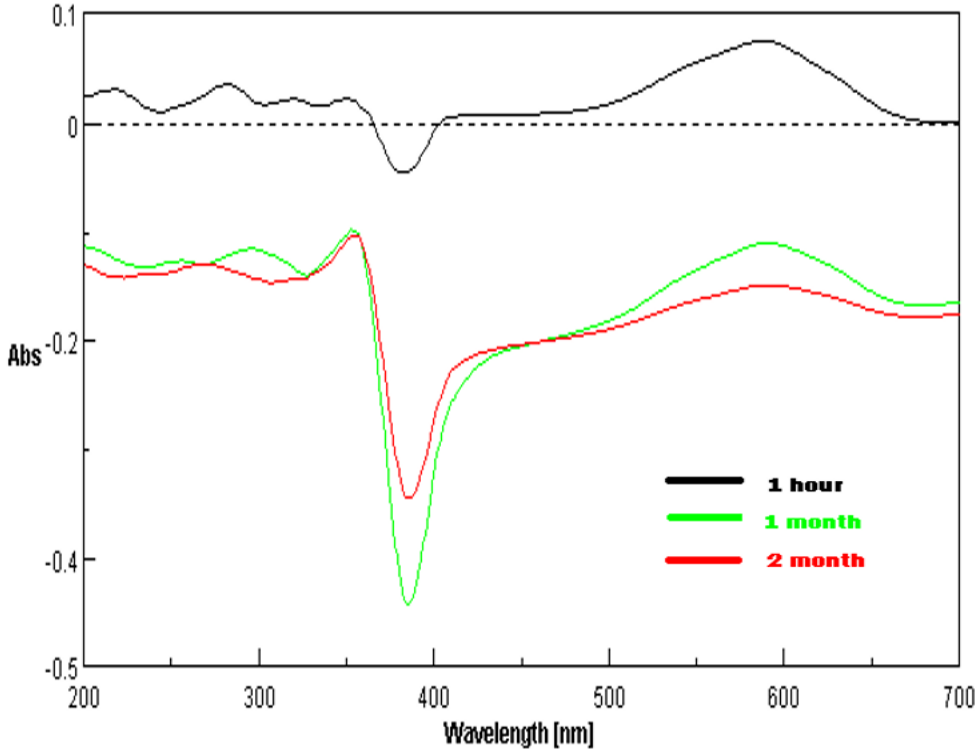
Absorbance observed is 0.01, -0.80, and -0.12 for samples after one hour, one month, and two months respectively.Wavelength peaks found are 539, 588, 590 - shows the blue shift due to the color of ink; 250 – shows the presence of alcohol (isobutanol or propanol) or halocarbons (1,2-dichloromethane) or alkanes. In the ink sample; 353, 354, 332, 309 – shows the presence of nitrites or ketones.

The absorbance observed for samples after one hour, one month, and two months was 0.75, -0.11, and -0.03, respectively.



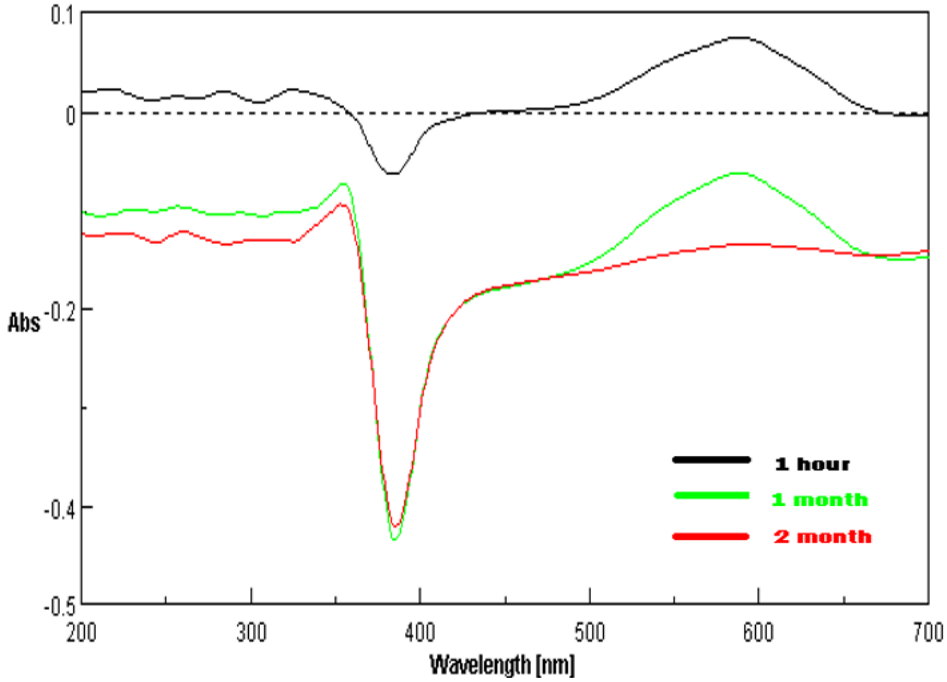
Graph 39 : Showing UV-Visible comparison spectral graph of Classmate UVO Ball with different time intervals.

Wavelength peaks found are 586, 588, and 589 - showing the blue shift due to the color of ink; 260 indicates the presence of alkenes; 300 showing the presence of nitroso or NN-dimethylformamide or ethers (1, 2-dimethoxyethane); 356, 357, 314, 304 – shows the presence of nitrites or ketones.



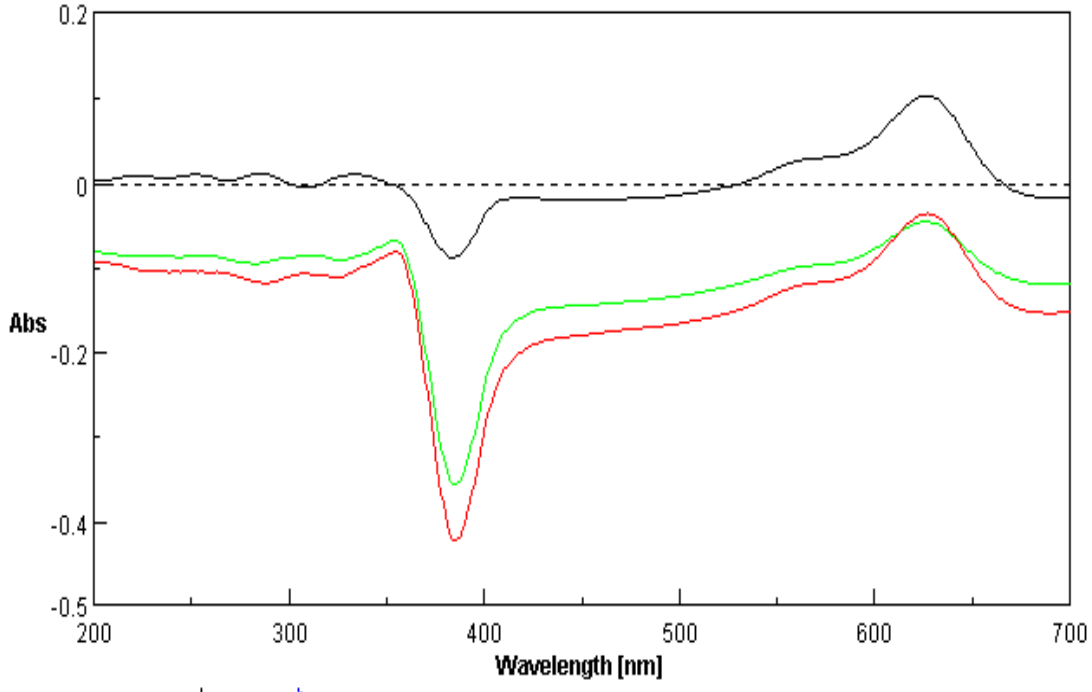
Graph 40: Showing UV-Visible comparison spectral graph of Cello Maxwriter with different time intervals.

The absorbance observed for samples after one hour, one month, and two months was 0.025, -0.11, and -0.13, respectively.



Graph 41 Showing UV-Visible spectra comparison spectral graph of 2K Add Gel with different time intervals.

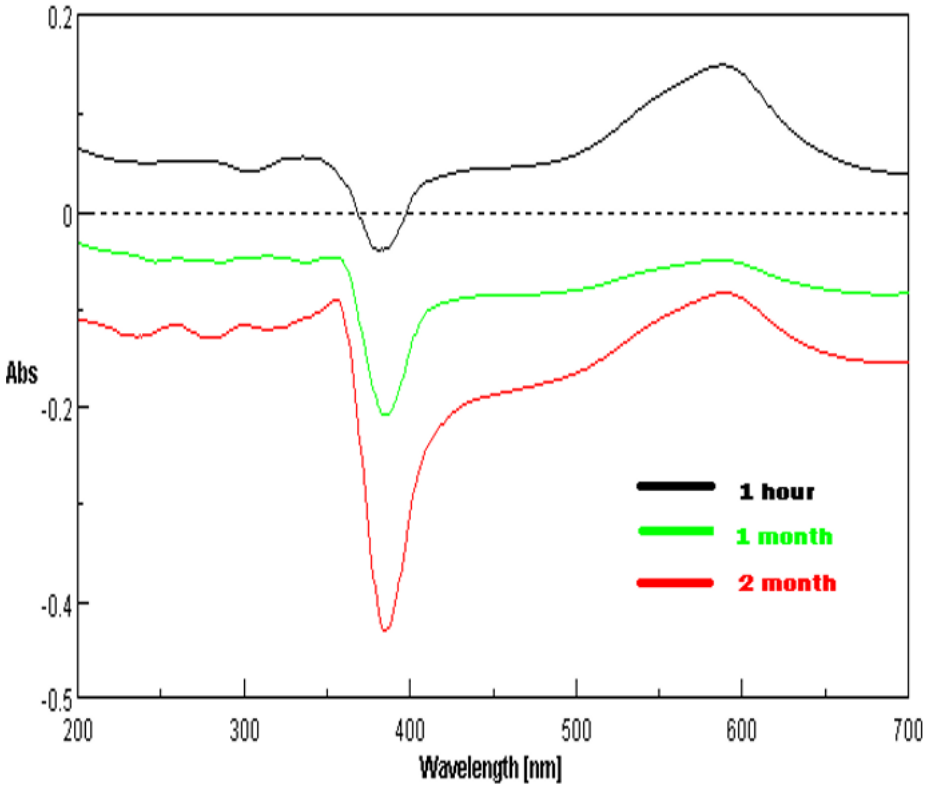
The wavelength peaks found are 554, 555, and 625. 554 shows the blue shift due to the color of ink; 320 indicates the presence of halocarbon (tetrachloroethylene); 295 points out the presence of azo; and 321 and 353 show the presence of nitrites or ketones.
The absorbance observed for samples after one hour, one month, and two months was 0.024, -0.10, and -0.12, respectively.



Graph 42: Showing UV-Visible comparison spectral graph of Add Gel Achiever with different time intervals.

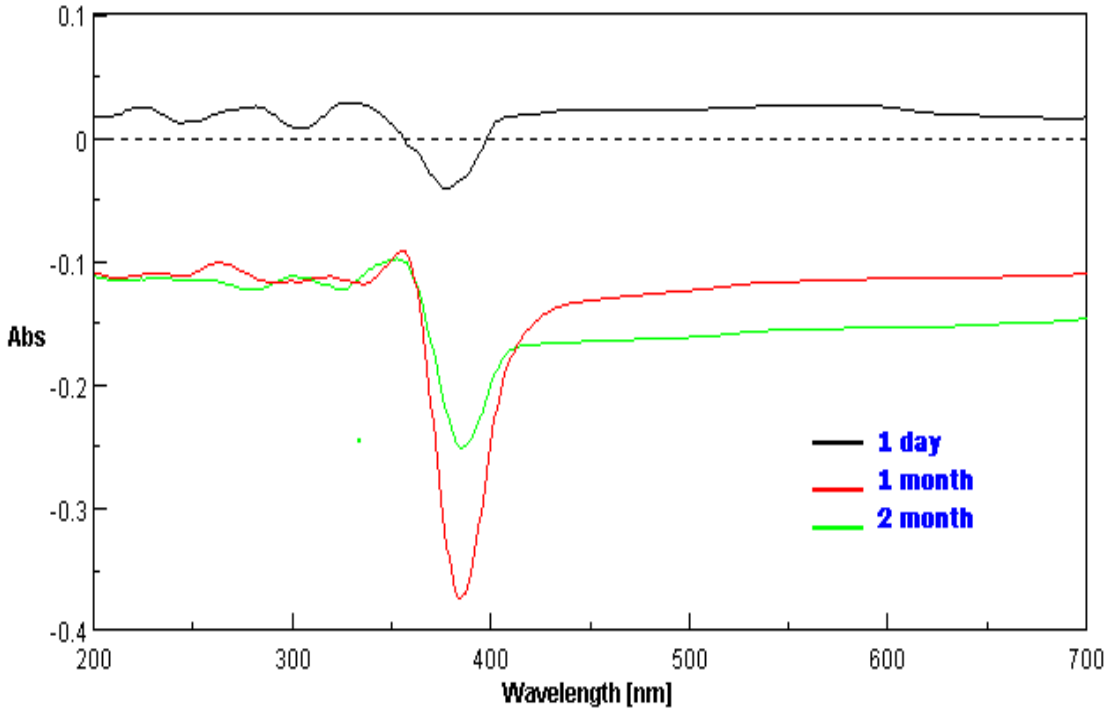
Wavelength peaks found are 588, 593 - shows the blue shift due to the color of ink; 220-indicates the presence of alkanes (cyclopentane) or enimine or naphthalene; 205-shows the presence of carboxyl or ester; 280-shows the presence of ether (tetrahydrofuran or 2-ethoxyethanol) or phenol; 325- shows the presence of *o*-xylene; 353,355 – shows the presence of nitrites or ketones.

Absorbance observed is 0.001, -0.08, and -0.09 for samples after one hour, one month, and two months, respectively.
Wavelength peaks found are 626, 627 - shows the blue shift due to the color of ink;417- Points out the presence of nitromethane or halocarbons(trichloroethylene) ;
280-points out the presence of esters(ethyl acetate) or ethers(tetrahydrofuran or 2-ethoxyethanol); 333, 308, 309 – shows the presence of nitrites or ketones.



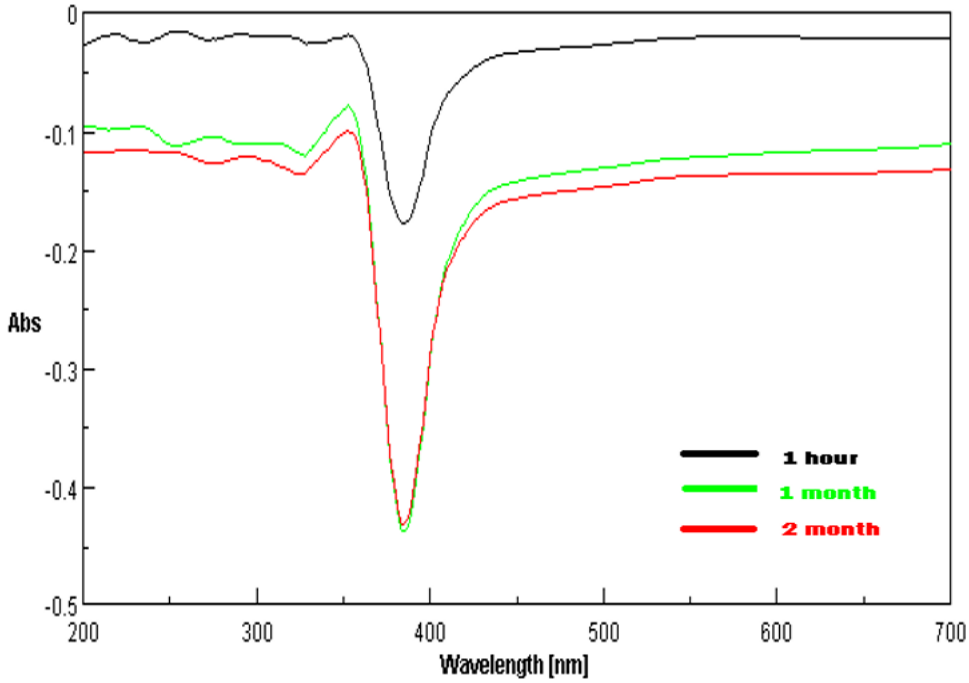
Graph 43 : Showing UV-Visible comparison spectral graph of Montex Activa Gel with different time intervals.

The absorbance observed for samples after one hour, one month, and two months is 0.001, -0.15, and -0.70, respectively. Wavelength peaks found are 554, 555, 628, and 625. 554 shows the blue shift due to the color of ink; 345 indicates the presence of ketones(butane-2-one); 414 indicates the presence of nitromethane or halocarbons(trichloroethylene); and 323, 312, and 353 show the presence of nitrites or ketones.



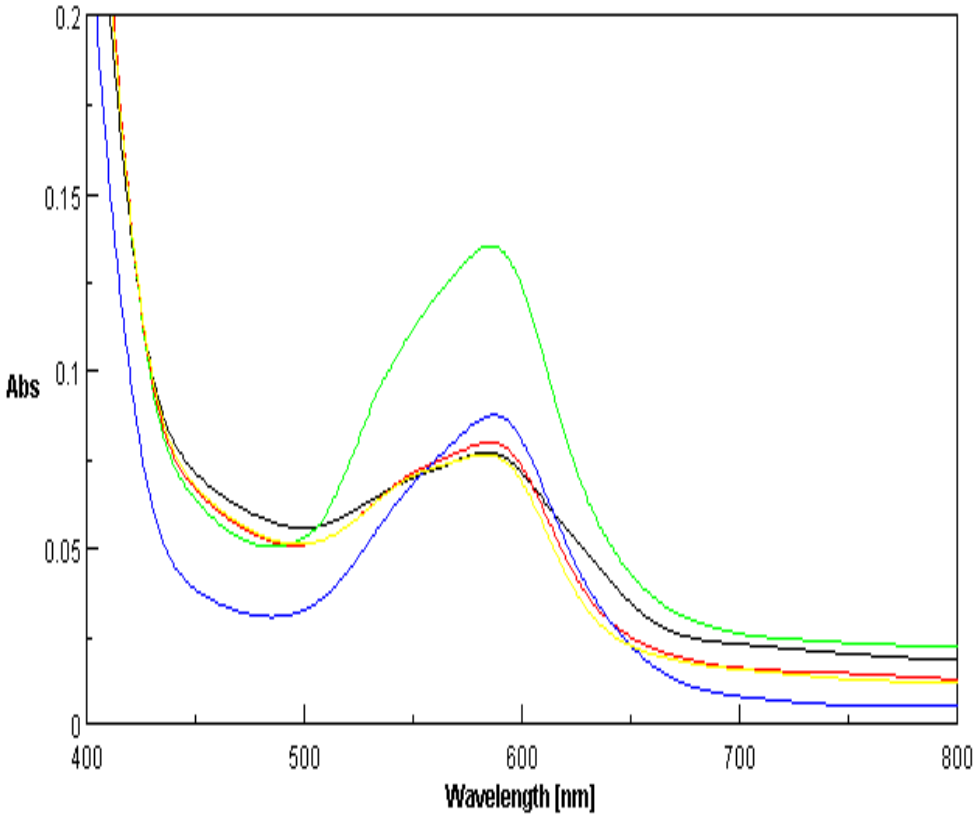
Graph 44: Showing UV-Visible comparison spectral graph of Flair Osmium Gel with different time intervals.

The absorbance observed for samples after one hour, one month, and two months was 0.019, -0.093, and -0.10, respectively.
Wavelength peaks found are 584, 594 - shows the blue shift due to the color of ink;300-shows the presence of nitroso or NN-dimethylformamide or ethers (1,2-dimethoxyethane); 299-show the presence of azo; 359,356 – shows the presence of nitrites or ketones.



Graph 45: Showing UV-Visible comparison spectral graph of Classmate Octane with different time intervals.

The absorbance observed for samples after one hour, one month, and two months is -0.015, -0.12, and -0.14, respectively. The wavelength peaks found are 591, 601, and 659. 591 shows the blue shift due to the color of the ink; 294 indicates the presence of azo; and 353 shows the presence of nitrites or ketones.



Graph 46: Showing UV-Visible comparison spectral graph of natural samples of year 2006,2007,2008,2009 and 2010.

NATURAL SAMPLES COMPARISON GRAPH

The absorbance observed for samples after one hour, one month, and two months was 0.127, 0.027, 0.026, 0.025, and 0.025, respectively. The wavelength peaks found are 583, 587, 588, and 589. The latter shows the blue shift due to the color of ink; 220 indicates the presence of alkanes (cyclopentane), enimine, or naphthalene; and 304, 312,328 shows the presence of nitrites or ketones.

1. Montex Activa Gel pen ink has two spots of red and blue color, indicating the presence of two different dyes.
2. Likewise, two spots of purple and blue color are found in 2k Add Gel, Add Gel Achiever, Cello Jet Ace, and Cello Maxwriter pen inks, pointing out that more than one dye or color pigment is used.
3. Due to the degradation of the colorant's hardening, polymerization of the resins, and evaporation of solvents, the ink samples show differences in their peak values in UV-visible spectroscopy.
4. The graphs of UV-visible spectroscopy show a gradual decrease in the absorbance of the entire prepared sample at 15-day, 1-month, and 2-month intervals.
5. Similar observations of the decrease in the absorbance of the natural samples from 2010, 2009, 2008, 2007, and 2006, respectively, are observed.
6. Since ink aging is a natural phenomenon, it lowers the R-ratio, i.e., the ratio of ink extraction in a given duration of time, which can also be observed in the graphs.

CONCLUSION

The study highlights the use of simple analytical techniques, particularly UV-visible spectroscopy,

for ink dating in forensic document examination. Before 1950, ink analysis relied on filter photography and chemical spot tests, while modern methods included IR spectroscopy, microspectrophotometry, and TLC. Though TLC is popular, advanced techniques like GC are now explored for volatile compound analysis in ink. The study shows a gradual decrease in ink absorbance with age due to dye degradation and evaporation of volatile compounds, while λmax remains stable. UV-visible spectroscopy proves efficient for determining ink age and plays a crucial role in criminal cases involving questioned documents. Additionally, ink analysis can reveal the pen's brand, the economic status of the accused, alterations or obliterations, and changes in ink color over time.

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