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RESEARCH ARTICLE

Integration of ATR-FTIR Spectroscopy and Machine Learning for Age Prediction Model of Black Gel Pen Inks

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Abstract Ink analysis provides a crucial function in forensic document examination for authentication, forgery detection, and ink dating. Today, black gel inks are widely used in both legal and non-legal documents, however they are difficult to analyse due to their pigment formulations, which undergo subtle chemical changes during ageing. Destructive techniques such as Thin Layer Chromatography (TLC) and High-Performance Liquid Chromatography (HPLC) can discriminate ink but are unsuitable for evidentiary purposes. Non-destructive Attenuated Total Reflectance-Fourier Transform infrared (ATR-FTIR) spectroscopy offers an alternative, yet spectral similarities among black gel inks necessitate advanced computational models to facilitate discrimination, classification and age prediction. In this study, predictive ageing models for black gel inks was developed by integrating ATR-FTIR spectroscopy with machine learning (ML). Thirty black gel inks from 23 brands were analysed. Ink lines made using the black gel ink samples were aged for twelve months under three different environmental conditions, and their infrared (IR) spectral data were recorded monthly over the period of 12 months. For age prediction, four classifiers namely Discriminant Analysis (DA), Support Vector Machine (SVM), k-Nearest Neighbour (kNN), and Decision Tree (DT) were trained on full mid-IR, fingerprint region, and PCA (Principal Component Analysis)-derived datasets. Performances of the classifiers were evaluated using accuracy, precision, recall, F1-score, ROC (Receiver Operating Characteristics), and Area Under the Curve (AUC). For age prediction, DA achieved the best accuracy (81.5%) with PCA features, outperforming SVM (76.1%), kNN (48.8%), and DT (40.2%). ROC-AUC values exceeded 90% across all classes. This study demonstrates that ATR-FTIR spectroscopy integrated with machine learning provides a reliable, non-destructive framework for black gel ink classification and age prediction, addressing limitations of destructive methods and strengthening forensic document analysis.

Keywords: Ink ageing, ATR-FTIR spectroscopy, black gel inks, forensic document examination, machine learning, non-destructive analysis.

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Introduction

Forensic document examination ocassiobally relies on ink analysis for verifying authenticity, authorship, forgery, and temporal aspects of documents. Writing inks, comprise of complex mixtures of solvents, dyes, pigments, resins, and additives, each imparting distinct chemical composition that enables discrimination. Of the writing inks, black gel inks are especially problematic due to their pigment-rich and water-based gel matrices which provides stability however contributed to minimal spectral variations over time hence, challenging ageing studies.

Immediately after ink is deposited on paper, it undergoes both chemical and physical changes, including solvent evaporation, resin polymerisation, and dye degradation [1]. In forensic document examination, ink age refers to the time elapsed since the ink was first deposited on the paper surface. Forensic document examiners often face significant challenges to estimate the age of ink, as the type of ink used, its chemical composition, and the conditions under which the document has been preserved are frequently unknown.

Non-destructive vibrational spectroscopy such as ATR-FTIR spectroscopy has increasingly being preferred in forensic analysis of inks because it preserves evidence integrity and facilitate rapid data acquisition. It has gained recognition as a reliable non-destructive technique for ink analysis, as its principal advantage lies in obtaining spectra directly from document surfaces without or with minimal sample preparation, thereby ensuring evidence preservation and integrity [2,3]. Several studies have demonstrated its sensitivity to determining the functional groups, enabling the differentiation of complex ink formulations [4,5]. However, spectral overlap among inks from different brands often reduces discrimination power and complicates data interpretation [2]. To address this limitation, chemometrics methods such as PCA and LDA have been employed to decipher latent spectral patterns and improve the separation of spectrally similar inks [5]. Nevertheless, even in the presence of spectral overlap and reduced discrimination power, ATR-FTIR spctroscopy remains a preferred technique in forensic document analysis for the reasons previously mentioned [3].

Predicting the age of inks is one of the most challenging tasks in forensic document analysis. Traditional chemical approaches often unsuccessful because solvent evaporation and dye degradation stabilize after only a few months. As a result, researchers have therefore reframed age estimation as a multiclass classification problem, treating each month of ageing as a separate class whereby dimensionality reduction has played an important role in this approach. Ortiz-Herrero *et al*. [5] combined PCA with hierarchical cluster analysis (HCA) to predict the age of inks up to two years, with error rates below 25%.

Machine learning (ML) has significantly impacted the field of spectroscopy by enhancing the analysis and interpretation of complex spectral datasets. The integration of ML in spectroscopic data analysis enables automated and real-time decision-making without human intervention, as it can provide feature-importance mapping and achieve high classification accuracy, thereby improving interpretability and analytical precision [6]. For instance, ML has been employed in addition to chemometrics techniques to capture non-linear spectral variations associated with ink ageing using spectral data that cannot be adequately modelled using conventional linear chemometric approaches alone [7]. These developments highlight the role of ML in strengthening forensic ink analysis by enabling robust data handling and deeper insight into complex compositional and ageing-related spectral changes.

Additionally, ML also shows a significant promise in predicting the age of ink, particularly when framed as a multiclass classification. Comparative studies of various classifiers have demonstrated that DA and SVM outperformed other classifiers in ink age prediction tasks. For example, SVM achieved the highest accuracy of 96.0% in classifying pen ink samples in a study using mass spectrometry data [6]. Similarly, López-Baldomero *et al*. [8] in their study claimed that DA and SVM models have shown superior performance in hyperspectral imaging- based ink classification, with SVM surpassing 90% accuracy.

Most studies involving inks to date have focused on ink classification, rather than age estimation. Addressing this gap, the present study integrates ATR-FTIR spectroscopy with ML to train and evaluate DA, SVM, kNN, and DT models for black gel inks age prediction, aiming to compare their performances in terms of accuracy, robustness, reliability and ultimately establish a predictive framework applicable in forensic contexts.

All four classifiers in this study provided distinct advantages for modelling infrared (IR) spectral data in ink age prediction. DA reduces high -dimensional spectral data to lower dimensionality, enhancing class separation while reducing within-class variance, and is computationally efficient for extensive datasets, however susceptible to noise [9,10]. SVM transform spectra into higher-dimensional space through kernel functions, effectively addressing intricate, non-linear age-related spectrum variations, despite requiring more computational demands [8,11] whereby kNN classifies according to spectral similarity, identifying slow aging patterns without strict data distribution assumptions; nonetheless, it necessitates feature extraction to eliminate insignificant variables [9]. DT employ a rule-based hierarchical framework to segment data according to the most relevant aspects, identifying spectral patterns associated with aging without requiring preprocessing, and integrating both numerical and categorical variables [12,13].

This study aims to develop and validate non-destructive models for the age prediction of black gel inks by integrating infrared (IR) spectroscopic data with machine-learning (ML) techniques. Specifically, the performance of multiple ML classifiers is systematically evaluated and compared to identify the most reliable and robust predictive model, while also examining the influence of different environmental ageing conditions on ink ageing behaviour.

Materials and Methods

In this study, thirty (30) black gel inks representing 23 brands (Table 1) were purchased from local sources and coded for ease of identification. Ink sample from each pen was prepared by drawing 5 cm strokes/lines on white A4 paper, air-dried for five minutes, and analysed in seven replicates per sample to ensure reproducibility. The samples were aged under three controlled environments: (i) direct exposure to light and air, (ii) the “sandwich effect” by placing the samples between plastic protective sleeves in a folder, and (iii) drawer storage with minimal light and airflow. Spectra were collected monthly over a 12-month period. In total, the three ageing conditions generated 7,560 IR spectra. ATR- FTIR measurements were performed using a LUMOS (Bruker, Germany) micro-FTIR interfaced with OPUS 8.8.2 software and equipped with a germanium (Ge) crystal sampling stage, covering the entire range of mid-IR from 4000 to 600 cm^{-1} at 4 cm^{-1} resolution, with 64 background scans. Figure 1 shows representative image of an ink stroke/line captured using the camera integrated in the LUMOS micro-FTIR while Figure 2 shows the typical seven replicates sampling locations on an ink line.

Table 1. Detailed information and corresponding IDs of the black gel ink samples used in this study

No	Brand	Model	Code ID
1	M&NISUN	606	A
2	Faber Castell	True Gel	B
3	Faber Castell	Eco Gel	C
4	Pilot	Super Gel	D
5	Pilot	Wingel	E
6	M&G	AGP02372-R3	F
7	M&G	AGP13271-Office	G
8	Pentel	BL110-Energel	H
9	NMANAN	MiWi Get	I
10	Zhi Xin	Diamond	J
11	Zhi Xin	Lovein Test Good	K
12	Zhi Xin	POS G-518	L
13	BIC	GLI X - Fine	M
14	Faster	SP-F-072	N
15	Monami	Jeller Pen 502	O
16	Monami	Jell Line	P
17	Paper Mate	Gel 300	Q
18	Unicorn	TGP-812C	R
19	CHOSCH	CS-G169	S
20	U-Fine	UC-Q8	T
21	Digno	Trinok Gel	U
22	Buncho	Fine Tech	V
23	Buncho	Jellie	W
24	TYNO	GP100	X
25	BEIFA	GA102601	Y
26	Mr DIY	Gel-067	Z
27	Zui Xua	CS801	A’
28	Test 2	GP-300	B’
29	MUJI	5110	C’
30	Stabilo	Palette	D’

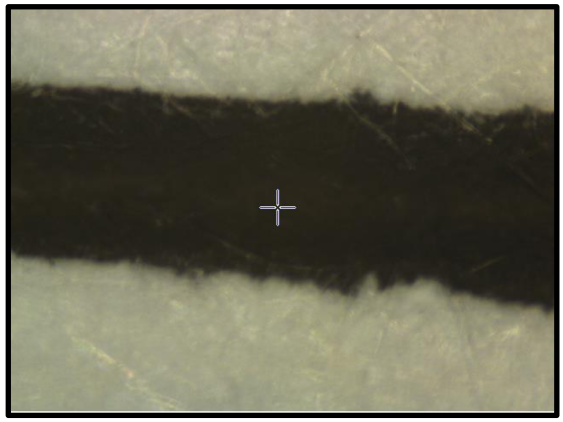

Figure 1. Representative image of an ink stroke/line captured using the camera integrated in the LUMOS micro-FTIR

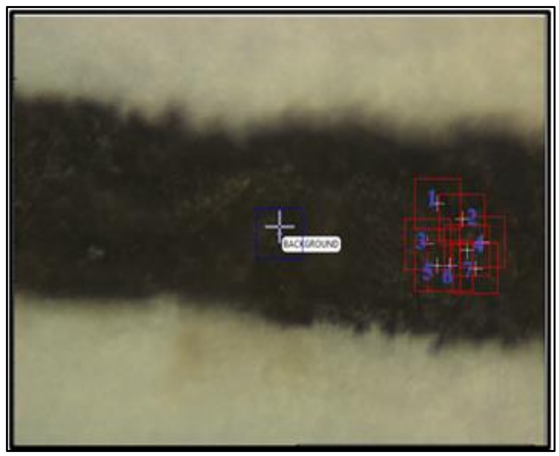

Figure 2. Typical seven replicates sampling locations on an ink sample/line

Prior to ML modelling, an initial data screening process was performed to ensure dataset suitability and to assess the contribution of different spectral representations. All IR spectra were first visually inspected for artefacts, baseline instability, and acquisition anomalies, with no spectra excluded as no systematic errors were observed. Three input datasets were then prepared for analysis: (i) raw IR spectra (4000–600 cm^{-1}), (ii) fingerprint region spectra (1500–600 cm^{-1}), and (iii) PCA-extracted features derived from the fingerprint region. Principal component analysis (PCA), performed using Minitab version 17 (Minitab, Inc.), was applied as a feature-extraction technique to reduce data dimensionality while retaining the most informative spectral features. The selection of principal components was guided by eigenvalues, scree plot analysis, and cumulative explained variance. In parallel, the raw mid-IR and fingerprint datasets were directly used as inputs to the machine-learning algorithms without feature compression, enabling comparison between feature-extracted and non-extracted datasets and evaluation of their effectiveness in supporting accurate ink age prediction.

Four ML classifiers, namely DA, SVM, kNN, and DT were evaluated for predicting ink age. All classifiers were trained using MATLAB 2024a (MathWorks, USA) software and their performances were compared across the three datasets described previously. DA demonstrated superior performance due to its effectiveness in modelling linear decision boundaries and accommodating inter-class variability. In contrast, kNN and DT were more susceptible to overfitting, as the high dimensionality and complexity of FTIR data diminished the discriminative meaning of distance-based measures [12] and creating complex rules that can fit with training dataset but failed to generalize the new data [13] respectively. SVM on the other hand, optimised decision boundary that maximises class separation by defining an optimal hyperplane known as support vectors, which enhances generalisation performance in high-dimensional spectral datasets.

Performance metrics, including accuracy (Eq. 1), precision (Eq. 2), recall (Eq. 3), F1-score (Eq. 4), receiver operating characteristic (ROC) curves, and the area under the curve (AUC), were employed to evaluate the predictive and classification performance of the developed ML classifiers. Accuracy quantifies the overall proportion of correctly classified samples, precision denotes the proportion of predicted positive samples that are correctly identified, and recall represents the proportion of actual positive samples correctly detected by the classifier. The F1-score, defined as the harmonic mean of precision and recall, provides a balanced assessment of classification performance. The ROC curve illustrates the trade-off between sensitivity and specificity across varying classification thresholds, while the AUC summarises the model's overall discriminative capability. The confusion matrix (Table 2) consists of true positives (TP), false positives (FP), true negatives (TN), and false negatives (FN), reflecting correctly and incorrectly classified samples across the two classes. Accuracy, precision, and recall were computed from the confusion matrix using the confusionmat function in the MATLAB. These metrics were extracted from the classifier identified as the best-performing model based on the highest overall classification accuracy among all evaluated models.

Accuracy = (TP + TN) / (TP + TN + FP + FN) Eq. 1
Precision = TP / (TP + FP) Eq. 2
Recall = TP / (TP + FN) Eq. 3
F1 = 2 x (Precision x Recall) / (Precision + Recall) Eq. 4

Table 2. The confusion matrix

	Predicted (Positive)	Predicted (Negative)
Actual (Positive)	TP	FN
Actual (Negative)	FP	TN

Results and Discussion

FTIR Spectra for Ageing Trends

The selected ATR-FTIR spectra from Buncho black gel ink (Sample V) which had been subjected to three different ageing conditions showed minimal observable chemical changes during the 12 -month experimental period. Representative spectra for each ageing condition (Figures 3-5) display subtle decline in specific functional group intensities. The most pronounced and reproducible change was the gradual decrease in absorbance within the 1033–1007 cm⁻¹ region (black circle), corresponding to C–OH stretching vibrations associated with alcohol as a solvent in black gel ink [14]. This decline suggests progressive solvent evaporation, particularly in samples exposed to light and air (Condition I) (Figure 3). This decrement is consistent with the findings reported by Said and Ismail [15], who observed a decrease in absorbance intensity within the same frequency region after 90 days of ageing.

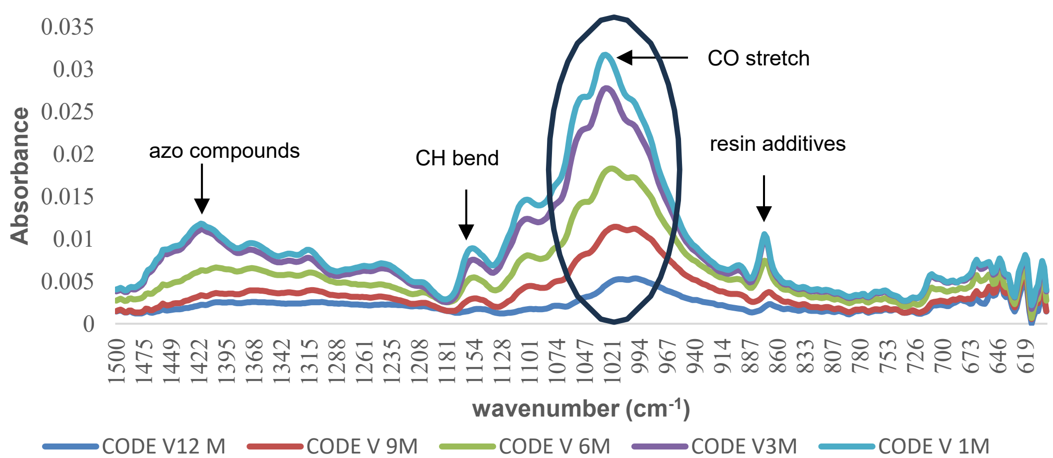

Figure 3. Representative IR spectra of black gel ink (Sample V) under Condition I over the 1, 3, 6, 9, and 12 months ageing period

Under Condition II ("sandwich effect") (Figure 4), the inks were enclosed between protective sleeves, slowing solvent evaporation and potentially inducing interactions with plasticizers in the storage medium. In this instance, it is possible that solvents trapped within the sleeves and undergo chemical interactions with ink components, thereby influencing the observed spectral behaviour [16]. Peaks in this condition displayed comparatively higher intensities in early months however gradually diminished.

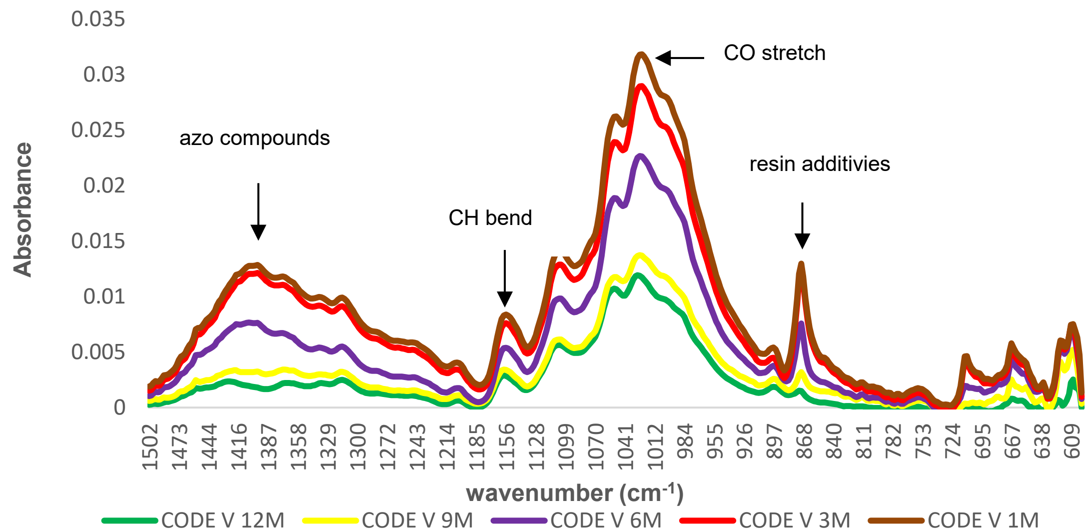

Figure 4. Representative IR spectra of black gel ink (Sample V) under Condition II over the 1, 3, 6, 9, and 12 months of the ageing period

Condition III (drawer storage) (Figure 5) shows controlled degradation, where reduced exposure to light and oxygen may have limited oxidative changes. Nevertheless, the intensity of absorption bands associated with additives around 865–872 cm⁻¹ region decreased under this condition, which may indicate resin hardening, a common phenomenon observed during ink ageing [17,18].

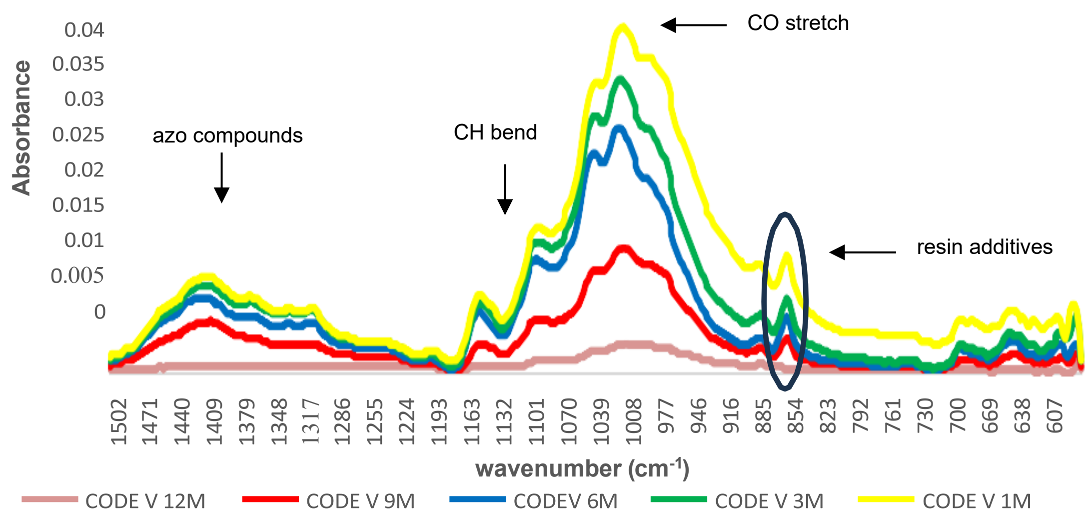

Figure 5. Representative IR spectra of black gel ink (Sample V) under Condition III over the 1, 3, 6, 9, and 12 months of the ageing period

No significant new peaks or losses occurred throughout the 12-month period, and all 30 ink samples exhibited spectral similarity, suggesting common formulations and mainly reflected physical changes such as solvent evaporation, resin hardening or degradation rather than the formation of new compounds in chemical changes. Despite these spectral observations, the overall similarity of the IR profiles across dye-based and pigment-based inks suggests that ageing differences are subtle and not easily distinguished by visual assessment.

Principal Component Analysis (PCA)

The PCA model (Figure 6) explains 72.9% of the total variance (PC1 = 49.2%, PC2 = 23.7%), indicating that the score plot represents the dominant spectral variability of the dataset. Despite the presence of two tightly packed clusters (red and blue circles) are evident, however, these clusters do not correspond to different ageing or storage conditions. Instead, samples from all three ageing environments are convoluted within both clusters, indicating no discernible condition-dependent. An outlier (yellow circle) was detected and is attributed to isolated spectral variability of ageing trend. This behaviour is consistent with the minimal chemical changes observed in the ATR-FTIR spectra over the 12-month period, confirming that the dominant sources of variance arise from inherent spectral similarity and subtle ageing-related fluctuations. Although more rapid ageing was anticipated under greater exposure to light and oxygen (Condition I) and slower ageing under minimal exposure (Condition II and Condition III), no measurable condition-specific differences were apparent. Similar trends are consistently observed across all remaining samples; therefore, environmental ageing conditions were not included as discriminative variables in subsequent ink ageing models. The clustering behaviour observed for Sample V is consistent with findings reported in previous study by Sauzier *et al*. [16] who demonstrated that inks stored in different environment or protected conditions exhibit minimal spectral changes over extended ageing periods.

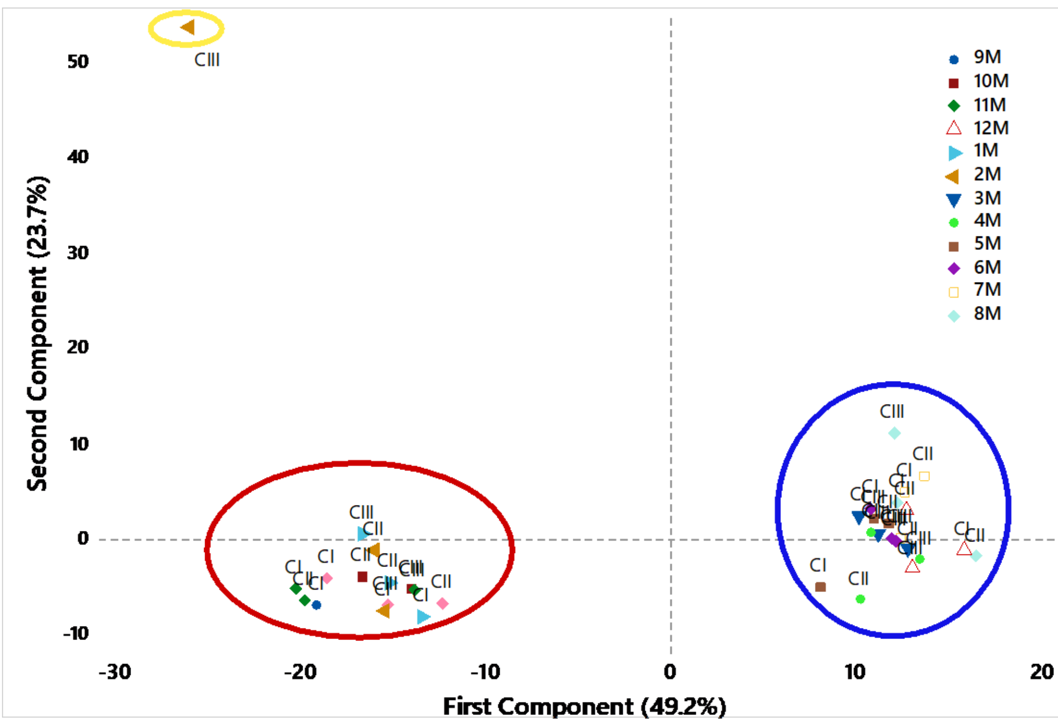

Figure 6. PCA score plot for Sample V during 12 months of ageing under three storage conditions demonstrates clustering with minimal separation across conditions.

Comparison of Machine Learning (ML) Classifiers

In this study, four ML classifiers namely DA, SVM, kNN, and DT for ink age prediction model were developed using the IR spectra acquired. The used of IR spectra for quantifying the ageing period is justified following the study conducted by Merriman et. al [19], in which IR spectroscopy was successfully used to quantify additive concentrations in rubber. Initial dataset on raw mid-IR spectra indicated modest accuracies (DA: 68%, SVM: 61.9%), with kNN and DT underperforming (<40%)

as illustrated in Figure 7. When restricted only to the fingerprint regions, accuracy further declined, reflecting the difficulty of extracting meaningful ageing features from limited spectral ranges.

However, incorporating PCA feature extraction dataset markedly improved the performances across the four classifiers. DA achieved the highest accuracy (81.5%), followed by SVM (76.1%), while kNN (48.8%) and DT (40.2%) remained comparatively weak. These results signify that dimensionality reduction enhances classifier robustness by filtering noise and highlighting informative IR spectral features.

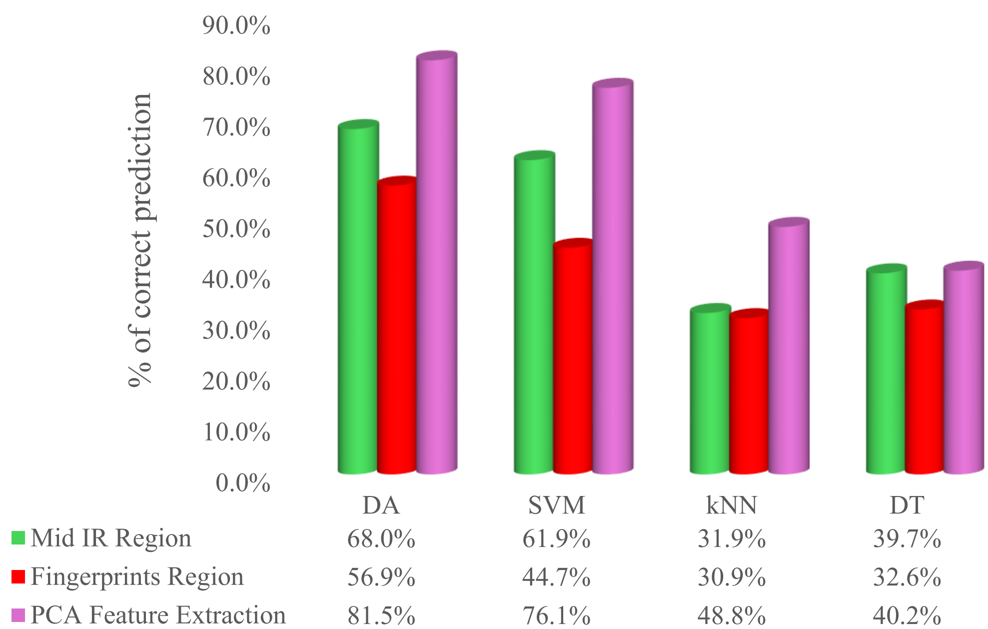

Figure 7. Comparison of classifiers (DA, SVM, kNN, DT) for ink age prediction using mid -IR, fingerprint, and PCA feature-extracted datasets highlights DA as most accurate (81.5%) after PCA feature extraction

Performance Metrics

For performance evaluation, four parameters were assessed for DA predictive model, the best-performing classifier, to determine its suitability for forensic ink ageing applications. These parameters include accuracy, precision, recall, F1 score. An accuracy (Figure 8) shows the excellent classification performances, with early-stage samples (Month 1) and late-stage samples (Months 11–12) achieving accuracies exceeding 99%. However, reduced accuracies (93–95%) in Months 3–6 indicated greater spectral overlap, complicating class separation. Accuracy comparisons showed near-perfect classification at Month 1 and late months, while intermediate stages were less reliable.

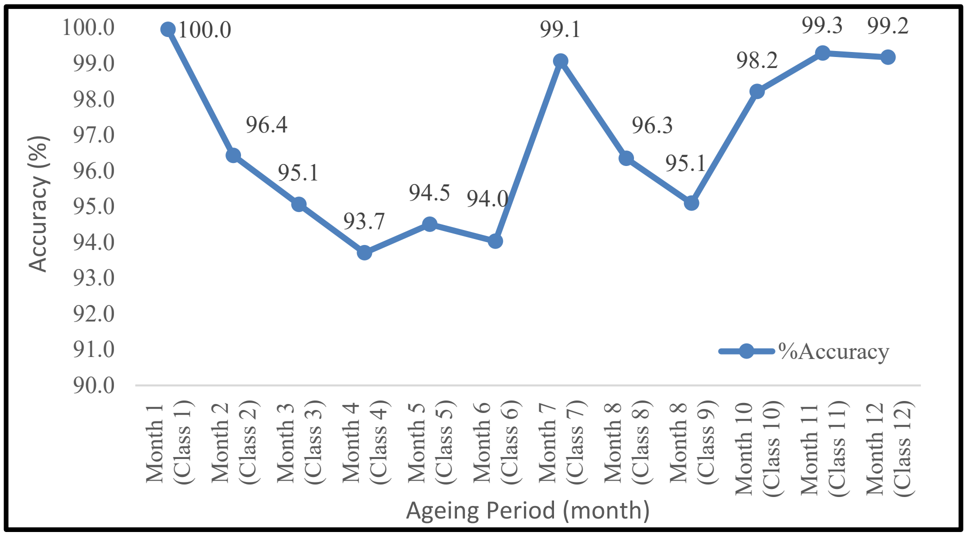

Figure 8. Accuracy comparison curve of DA across 12 classes.

Month 1 achieved precision of 100% (Figure 9), but subsequent months dropped below 70% due to overlapping features that increased false positives. The poorest performances occurred in month 2 and month 8, both around 59%, where samples were frequently misclassified as other ink ages. This suggests that inks from these classes lacked sufficiently distinct spectral or chemical attributes, making their profiles comparable to other aged samples and therefore more prone to misclassification.

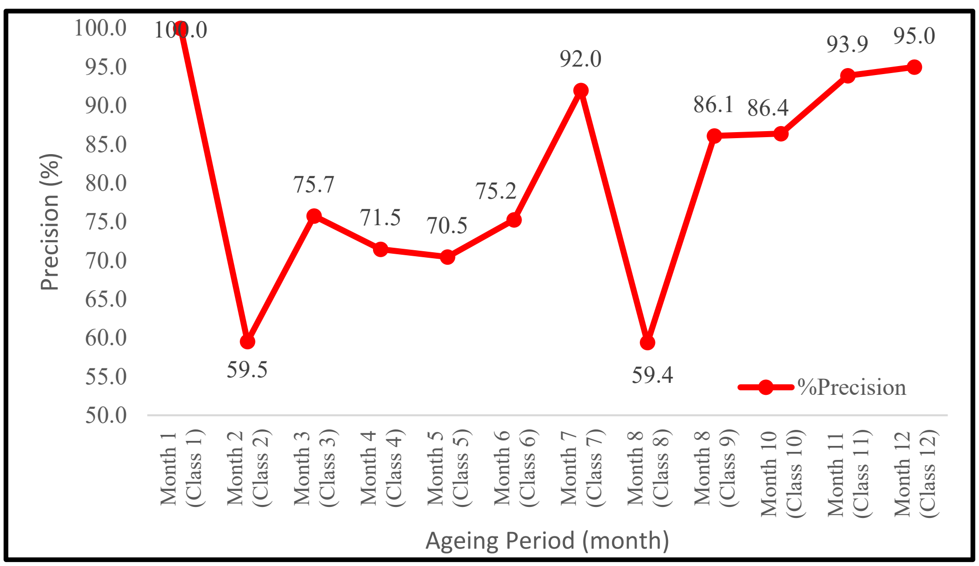

Figure 9. Precision curve of DA classifier across 12 ageing classes

Recall values (Figure 10) remained relatively strong (>90%) across most classes, although precision recall imbalance was evident in Months 2 and 8. Month 2 exhibits an outstanding recall of 96.70%; however, its low precision value indicates that, while it accurately identifies most of the samples, it also misclassifies several other samples as such, resulting in a significant number of false positives.

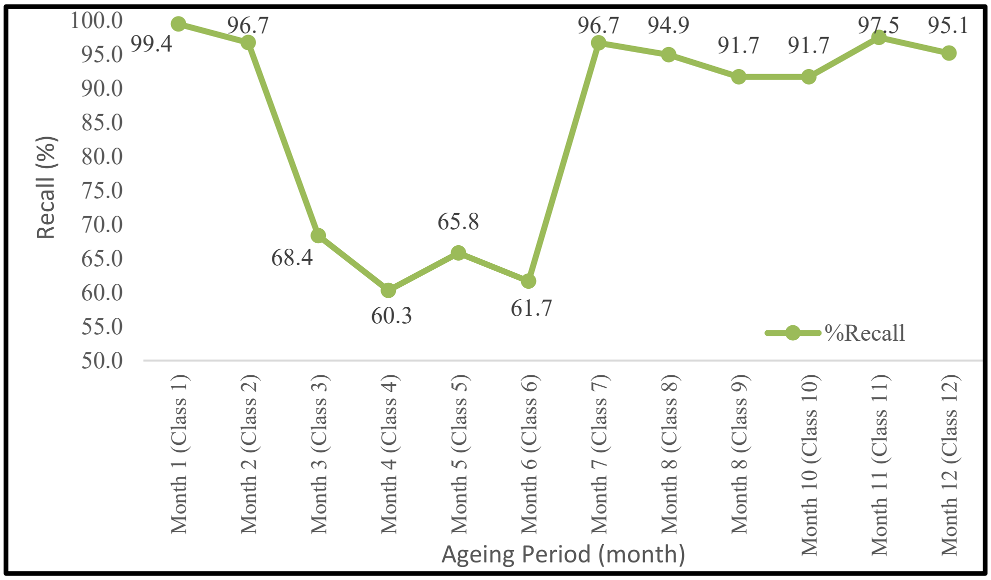

Figure 10. Recall curve of DA classifier across 12 classes

The harmonic F1-scores (Figure 11) mirrored this performance, with highest scores at early and late stages and moderate scores in the middle ageing range. Month 1 achieves the highest F1 score of 99.7%, demonstrating outstanding precision and recall. Month 7 to month 12 except for month 8 also exhibit robust performance, with F1 scores surpassing 88%, highlighting the DA persistent ability to balance precision and recall during this period.

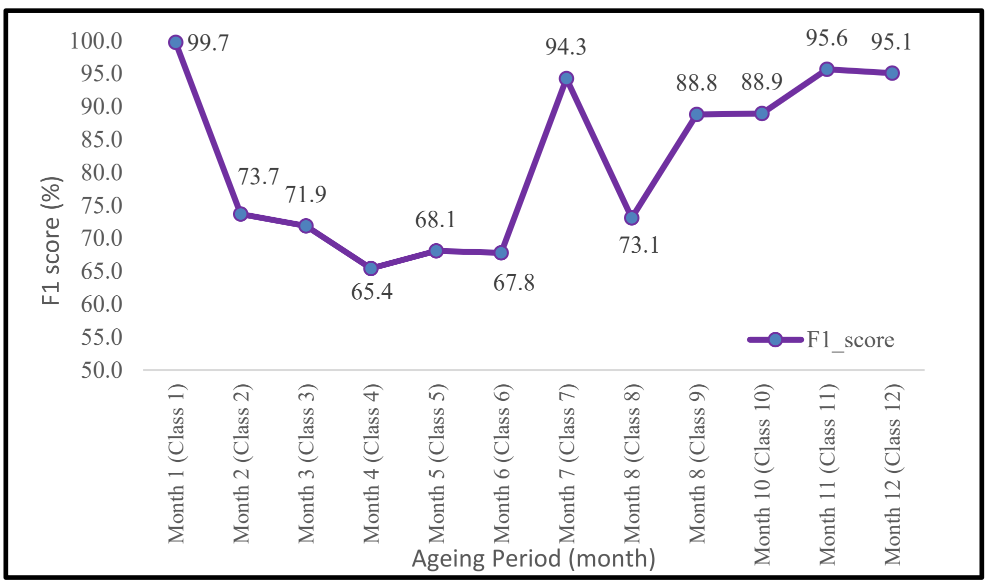

Figure 11. F1-score curve for DA classifier across 12 classes

ROC curves (Figure 12) confirmed DA's reliability, with AUC values consistently above 90% for all classes, and 100% for Month 1. These results reinforce that FTIR-based DA modelling is highly effective at distinguishing early and late ink deposits but less precise for intermediate months, where degradation is gradual and less chemically distinct.

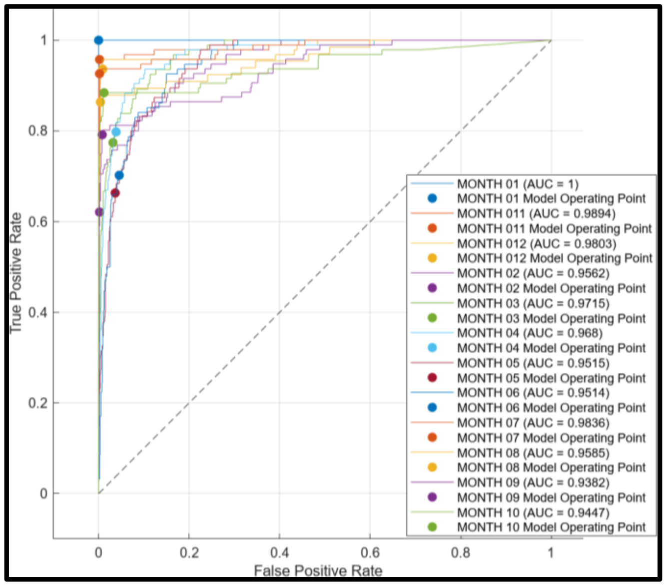

Figure 12. ROC curves and AUC values for DA classifier across 12 classes consistently >90% with perfect 100% for Month 1, confirming strong predictive power.

The results of this study align with and extend recent advances in non -destructive forensic ink analysis. Sugawara [20] reported 97.6% accuracy using DA for black marker inks, while Silva *et al*. [21] achieved 100% classification of blue inks with LDA. Gautam *et al*. [14] also demonstrated 100% classification accuracy for black ball and gel pen inks using PCA-FTIR analysis. Although the present study achieved a slightly lower maximum accuracy (81.5%), this is attributed to the inherent complexity of black gel inks, which often contain pigment-based formulations that exhibit minimal spectral variation during early ageing.

The novelty of this work lies in integrating ATR-FTIR spectroscopy with ML for black gel inks, a category less frequently addressed in the literature. While other study [16] has largely focused on blue inks, this study demonstrates that black gel inks present unique analytical challenges due to their pigment formulations and slower degradation. The successful application of DA with PCA feature extraction dataset highlights a reliable pathway for forensic ink dating, particularly when destructive techniques such as GC-MS or HPLC are impractical. This integration acts as the foundation for broader applications in questioned document examination.

Conclusions

This study demonstrates that integrating ATR-FTIR spectroscopy with machine learning provides a robust non-destructive approach for the classification and age prediction of black gel inks. Among the evaluated models, Discriminant Analysis (DA) applied to PCA-extracted features dataset achieved the best performance, with an accuracy of 81.5% and AUC values consistently exceeding 90%. The results indicate that early and late ageing stages are more readily distinguishable, whereas intermediate ageing periods remain challenging due to overlapping degradation processes. Overall, the proposed methodology reduces reliance on destructive testing and offers a reproducible analytical framework for forensic questioned document examination involving black gel inks.

Conflicts of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

Acknowledgments

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