Hyperspectral Database of Synthetic Historical Inks

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Abstract

The aim of this work is to provide the cultural heritage community with a comprehensive hyperspectral image database of handwritten laboratory samples, including various writing inks commonly found in historical documents. The database contains 195 samples registered in the VNIR (400-1000 nm) and SWIR (900-1700 nm) spectral ranges, along with complete information about the ink recipes (components and concentrations used for each ink and mixture), and their corresponding Ground Truth images. The database is now publicly available as part of a bigger database related to the Hyperdoc project and can be used to perform different tasks. We present here one example: the classification of iron gall vs non-iron gall inks.

Introduction

Non-destructive analysis of material cultural heritage is essential to preserve its integrity and value. Within the available tools, hyperspectral imaging is being increasingly used in this field to obtain high spectral and spatial resolution images in different spectral ranges, covering the visible, near infrared and short wavelength infrared [1]. These spectral images can then be used, among other applications, to monitor restoration interventions and cleaning processes [2], to study chromatic changes over time caused by degradation phenomena [3], to improve the legibility of deteriorated text [4], and to identify the materials used in polychrome surfaces such as drawings or paintings [5, 6], and historical documents, such as manuscripts [7, 8].

The analysis and identification of the inks present in manuscripts are an essential part of any codicological study and contribute to enhance their understanding, achieving a comprehensive historical and artistic contextualization. This is a fundamental step in planning any conservation intervention and developing long-term preservation and restoration strategies [1, 9]. To help with this task, a large database of samples that can be used to train models for classification of materials is highly encouraged. The use of different supports is necessary since the spectra of the support influence the spectra of inks.

In this context, different databases of real handwritten historical documents have been proposed in the past [10-24], most of them containing only RGB images to perform a binarization task. Only one multispectral database containing 10 handwritten documents with 8 bands in the VNIR range has been used for segmentation [25].

In this work, we aim to contribute to the cultural heritage community through the creation of a comprehensive hyperspectral image database with 39 handwritten laboratory samples in 5 different supports (papers and parchment), for a total of 195 samples, in two spectral ranges: visible and near infrared (VNIR), and short wavelength infrared (SWIR) [26]. The writing inks used to create the samples are commonly found in historical documents, including 8 recipes of metallo-gallate inks, 4 of sepia, 6 carbon-based inks, and 17 mixtures. In addition, 4 samples of constituent materials used in some of the inks were made. This database opens a wide range of possibilities for the study of historical inks, allowing the development and application of algorithms for material identification in manuscripts, or for segmentation of support and ink, and contributing to the automation of analysis processes.

Materials and Methods

Materials

The laboratory samples included a 1 x 1 cm square drawn with pencil and filled with ink, and two lines of text (as seen in Figure 1) written with brush (top) and fountain pen (bottom). This enables the study of how the spectrum changes depending on the amount of ink deposited, and different writing instruments can also be assessed. The inks were bound with Arabic gum.

Figure 1. False RGB images ([605, 535, 430] nm) of laboratory samples in cotton paper. (A) Full pages; (B) Extracted minicubes.

All the inks were applied on 5 different supports: 3 handcrafted papers from Paperlan® made of 100% linen fiber, 100% cotton fiber, and mixture of linen/cotton (50:50), handcrafted paper from Wanderings® made of 100% hemp fiber, and parchment. The supports were selected based on those found in historical documents [27].

Historically, the most prevalent inks include metallo-gallate inks, sepia, and carbon-based inks [28].

Metallo-gallate inks

Historic metallo-gallate inks consist primarily of two key components: a metal (predominantly ferrous sulfate), and a vegetable tanning agent, typically derived from oak apples in the form of gallotannin extracts. The nomenclature "iron gall inks" is assigned to this category of inks due to the significant presence of iron sulfate in their composition [29].

In this work, ancient recipes of iron gall ink were followed [30, 31]. In the preparation of this kind of ink, first, 23.53 g of oak apples (from Kremer Pigmente GmbH) were crushed (but not

pulverized), wrapped in a cotton cloth, and macerated in 400 ml of water under direct sunlight for 3 days. After that, the solution was filtered, obtaining the gallic acid.

Primarily, pulverized ferrous sulfate ($FeSO_4$-$7H_2O$) was used and added to gallic acid. We varied the ratio of gallic acid and ferrous sulfate obtaining 3 different inks. In addition, pulverized copper sulfate ($CuSO_4$) and zinc sulfate ($ZnSO_4$) were mixed separately with ferrous sulfate to create different inks. Powdered Arabic gum was added to all the recipes to obtain the inks.

Following Andalusian ink recipes [31] we made two variants of iron gall ink, including in both pomegranate juice, but with the addition of myrtle leaves in the gallic acid only in one of them.

In addition, a black pigment called "Atramentum" (from Kremer Pigmente GmbH) was used. It is produced from the reaction of tannic acid extracted from oak bark and iron salts, and has historically been referred to as "ink stone".

In total, 8 variants of pure metallo-gallate inks were produced.

Sepia

Sepia ink has a very dark brown color, almost black in full strength. It is obtained from the ink sac of the cuttlefish *Sepia officinalis* [32].

Two kinds of sepia ink were used in this database: the first one, extracted directly from the animal, and the second one, obtained from Kremer Pigmente GmbH in powder form. Both inks, the natural and the powdered one, were mixed with an Arabic gum solution in water. For the former one, three samples were made: pure ink extracted from the sac with some drops of water, and another two varying the concentration of the Arabic gum solution.

Carbon-based inks

The earliest writing ink is thought to have been a carbon-based ink derived from lamp-black or other form of charcoal. These inks are obtained by burning oil or other materials (such as fruit stones, bones, or wood) in controlled conditions with a limited supply of air and mixing them with a binder dissolved in a water-soluble medium [32]. They are very resistant compared with sepia and iron gall inks, being almost insensitive to acids, light, alkalis, and chemicals.

All the carbon-based inks (from Kremer Pigmente GmbH) were bound with an Arabic gum solution, and are listed below:

- Ivory black: it was originally obtained from charred ivory or horns, although nowadays it is based on bone black, being the best qualities of that ink. This is the most intense, deep black pigment.
- Bone black: this pigment is obtained by carbonization of bones, usually with temperatures over 400°C but not above 800°C.
- Lamp black: this pigment is obtained by collecting the condensed smoke produced from a flame fueled by mineral oil, tar, pitch, or resin within brick chambers.
- Grape seed black: for an extended period, different types of fruit stones have been charred and used as pigments. This pigment is obtained by carbonizing grape seeds, and it has a bluish tone.
- Cherry black: it is similar to grape seed black, but in this case cherry pits are charred, obtaining a warmer, brownish black.

- Bistre: it is a warm, deep transparent brown pigment, traditionally obtained from the soot of beech wood or other trees.

Mixed inks

The study of mixed inks has received little attention in both scholarly and material investigations, as their identification and acknowledgment of their significance are recent developments. Medieval Arabic ink recipes suggest that mixed inks held a significant role in the Islamic world [33, 34]. Mixtures of carbon ink and iron gall ink have been found since ancient times [9, 32] but detect and classify this kind of inks is still a challenge, especially when the identification is limited to non-invasive methods [33]. That is why we have included 85 samples (17 mixed inks in 5 supports) of different mixed inks in the database.

Sepia ink (the one obtained from the animal) was mixed with pure iron gall ink, lamp black, and bones black, in three different proportions: 25:75, 50:50, and 75:25.

In addition to sepia, iron gall ink was mixed with lamp black and bones black separately in the same three proportions: 25:75, 50:50, and 75:25.

Andalusian red earth (from Kremer Pigmente GmbH) was also mixed with iron gall ink and lamp black in a 50:50 ratio.

Constituents

Finally, 4 reference samples were made with constituent materials of different inks:

- Arabic gum (from Kremer) diluted at 20%.
- Pomegranate juice.
- Myrtle leaves infusion.
- Andalusian red earth (from Kremer), bound with an Arabic gum solution.

Methods

Spectral capture

Two hyperspectral cameras from Resonon Ltd. were used: the Pika L, which covers the VNIR spectral range from 400 to 1000 nm, with a spatial resolution of 900 pixels per line and a spectral resolution of 2.1 nm; and the Pika NIR, which covers the SWIR range from 900 to 1700 nm, with a spatial resolution of 640 pixels per line and a spectral resolution of 2.4 nm. Spectral binning and interpolation were carried out, obtaining a final sampling interval of 5 nm: 121 bands in VNIR and 161 bands in SWIR. Both cameras were used along with a linear stage and halogen illumination.

Database formation

All samples in the database are MAT-files that include: the cube (hypercube object), Metadata (struct) and wl (vector of wavelengths).

Once the samples in the 5 kinds of support were captured, the registration of the VNIR and SWIR ranges was done. To do that, the *Registration Estimator* app [35] included in MATLAB® software (R2023a version) was used. Automatic registration was performed by detecting, extracting, and matching features, using similarity or affine transformations, as appropriate.

The next step was the extraction of minicubes from the registered captures, reducing the processing time by decreasing the size of the files. That way, each minicube contained only support,

pencil (used to draw the square), and one specific ink. The sizes of the minicubes range from [508 x 181] to [273 x 120] pixels.

After that, the Ground Truth (GT) images were created using a semi-automatic procedure distinguishing between the three components of each minicube: the ink (white), the support (black), and the pencil (gray). First, the spectral band with the highest contrast in each VNIR minicube was selected using a metric based on the Signal-to-Noise Ratio [36]. Then, the ink skeleton was obtained using the MATLAB® function *bwskel* and grown until the pixel intensity was similar to the mean intensity at the edges of the strokes, detected with a Canny edge detector. Some manual corrections were made using GIMP software. These GTs can be used for both VNIR and SWIR minicubes.

Finally, a struct called 'Metadata' was included in all minicubes, containing information about the sample: number, name, ink components or recipe, kind of support, if it is varnished or not, if it is restored or not, if it is aged or not, date, height, width, number of bands; information about the capture: device, illumination, stage, reference white, and exposure time used; and, finally, some other important information as the GT, GT labels (if known), parent cube, coordinates within the parent cube, and a false RGB image of the minicube.

Classification task

A classification task is introduced as a possible application of the presented database, differentiating between iron gall inks and non-iron gall inks. In the former group we have included all the pure and mixed inks containing some amount of iron gall ink.

We have used the VNIR information of the samples applied on parchment and cotton-linen paper, supports commonly used in historical documents. The classification model used was a Bilayered Neural Network (NN), with two fully connected layers excluding the input and the final layer for classification. The selection of the model was based on a previous study [37] where a Fully Connected NN outperformed a One-dimensional Convolutional NN and a Support Vector Machine (SVM), as well as different spectral metrics, in a pigment classification task. The sizes of the layers were: 32 and 16 respectively, with the hyperbolic tangent as activation function, and softmax activation in the final layer [37].

The model was trained with 85000 spectra of iron gall ink, 85728 spectra of non-iron gall ink, 84000 spectra of support (including parchment and cotton-linen paper), and 21844 spectra of pencil. The ink spectra were extracted from the squared areas of the samples. To test the model, 72 full minicubes of ink in the VNIR range were used. A cross-validation with 5 folds was performed to evaluate the performance.

The *Classification Learner* app [38] included in MATLAB® software (R2023a version) was used to obtain the results with the pre-trained NN.

Results

Hyperspectral database

The hyperspectral database can be found here: [26]. It can be divided into 4 main groups, according to the composition of the inks: metallo-gallate, sepia, carbon-based, and mixed inks. In Table 1 the number of samples belonging to each group is specified, including the 5 supports.

Table 1. Hyperspectral Database: number of samples in the four main groups.

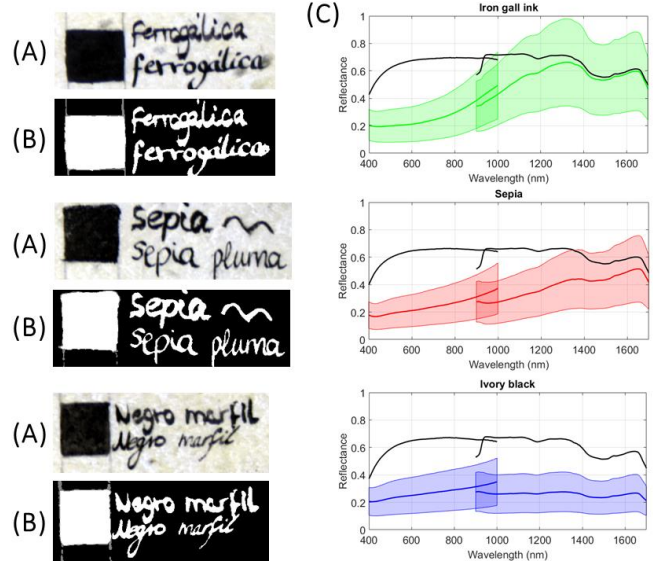| Group	# of samples
Metallo-gallate	40
Sepia	20
Carbon-based	30
Mixed	85

Figure 2. (A) False RGB images ([605, 535, 430] nm) of 3 minicubes: iron gall ink, sepia and ivory black, applied on parchment; (B) GTs of the minicubes; (C) Mean VNIR and SWIR spectra of pixels with support (black lines) and inks (green, red, and blue lines) with standard deviation.

In Figure 2.C, the mean spectra and standard deviation of three inks in the VNIR and SWIR ranges are presented. The spectra of iron gall ink, sepia, and ivory black have been extracted using the white pixels in the GTs (Figure 2.B). The mean spectrum of the support is also represented as a black line. In the VNIR range, the reflectance of the three inks exhibits a similar trend. However, as we approach the near infrared, the spectrum of iron gall ink deviates from the others, becoming increasingly similar to that of the support. This trend is particularly prominent in the SWIR range, especially from about 1300 nm onwards, where iron gall ink becomes nearly transparent. This transparency distinguishes iron gall ink from carbon-based inks like ivory black, which absorb a substantial portion of the infrared radiation incident on them. While sepia ink allows slightly more infrared radiation to pass through, it does not exhibit the near-total transparency characteristic of iron gall inks.

Classification

Table 2. Evaluation metrics (accuracy, precision, recall and F-score) for the four classes: iron gall ink (IGI), non-iron gall ink (NIGI), support (S) and pencil (P).

	IGI	NIGI	S	P
Accuracy	0.67	0.67	0.67	0.67
Precision	0.85	0.65	0.97	0.04
Recall	0.46	0.71	0.70	0.74
F-score	0.60	0.68	0.82	0.08

Figure 3. Classification maps of two minicubes: (A) sepia ink, and (B) mixture of iron gall ink and lamp black 50:50, both applied on cotton-linen paper.

As can be seen in Table 2, a 67% accuracy (proportion of correct predictions to the total number of input samples) is obtained using the Bilayered Neural Network. The lowest precision is achieved for the pencil class (4%). This is because a lot of support pixels have been misclassified as pencil, as can be seen in Figure 3.A, leading to an increased number of false positives for the pencil class. In contrast, the highest precision value (97%) is obtained for the support class, indicating that very few ink or pencil pixels are misidentified as support. Comparing both types of inks, a higher precision but a lower recall is observed for the iron gall ink class. A low recall value indicates a lower proportion of true positives to the actual total number of positive samples. So, an iron gall ink is more likely to be classified as non-iron gall rather than vice versa. When an iron gall ink is mixed with a non-iron gall ink, especially a carbon-based ink, the reflectance of the spectrum diminishes considerably, even with the addition of a minimal quantity of carbon-based ink. This makes the ink spectrum, despite containing a considerable amount of iron gall ink, to become more similar to the spectrum of carbon-based ink. This explains why most of the pixels in Figure 3.B, which shows the classification map for the minicube with an equal mixture of iron gall ink and lamp black (50:50), are classified as non-iron gall ink.

Conclusions

In this study, a comprehensive hyperspectral database of synthetic historical inks is presented [26]. 39 inks and components of inks have been applied on 5 types of supports, for a total number of 195 handwritten laboratory samples. All the samples were captured and registered in the VNIR and SWIR spectral ranges. The inks used for these samples are commonly found in historical documents, and include various formulations of metallo-gallate inks, sepia, carbon-based inks, and their mixtures.

A classification task has been conducted using a Bilayered Neural Network to distinguish between four classes: iron gall ink, non-iron gall ink, support, and pencil. A 67% of accuracy has been reached, and the pencil showed particularly low precision, being misclassified with the support. In addition, iron gall ink is more difficult to classify when it is mixed with other inks, especially carbon-based inks. Further efforts should be done to improve the overall accuracy, particularly with mixed inks and pencil. In the future, we will study how the type of fiber employed in paper preparation affects the results and interpretation of data. Additionally, we will explore the impact of using either paper (cellulosic support) or parchment (proteinic support) on the study outcomes.

Due to the versatility of the database, it can be used for different tasks related to hyperspectral image analysis and image processing in general. Machine learning and deep learning algorithms can be used to perform ink classification and binarization. In addition, it can be used to perform multivariate exploratory analysis in order to identify spectral features related to some kinds of inks or their components.

The database is now publicly available [26], making it a valuable resource for researchers and professionals in the cultural heritage field. This database is part of an extensive hyperspectral imaging database emerging from the Hyperdoc project, which will incorporate samples from real historical documents, including polychromatic samples. The specific composition of each ink will be provided along with the database, which will be made fully available progressively during 2024 at [26] and will include both real documents of historical interest and mock-up samples.

Acknowledgments

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