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Material Characterisation of 19–20th Century Manuscripts from Northern Thailand

Materialanalyse von Nordthailändischen Manuskripten aus dem 19.-20. Jahrhundert

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Abstract: Material analysis was carried out on four manuscripts from Northern Thailand, which included two palm-leaf manuscripts and two paper manuscripts. The two palm-leaf manuscripts MS 6 and MS 7 were found to have been written in the traditional method, wherein text was incised on the surface of the leaves, and then soot applied to the surfaces, confirmed by the identification of carbon ink. MS 7 additionally showed the presence of trace levels of iron in the ink, either from soot paste or from the stylus used for inscribing. The paper manuscript MS 3 was written in iron-gall ink, with sections written using a methyl-violet based ink while MS 4 was written in carbon ink. The paper used in the case of MS 3 was found to be machine made, while *khoi* fibres (*Streblus asper*) were used for making the paper used for MS 4. A combination of traditional and modern pigments, like molybdenum orange, was used for decorating the edges and cover. The results improved our understanding of these manuscripts in particular, and also provided us with insights about the rapid adoption of modern

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materials and their incorporation into the production of written artefacts from Northern Thailand in the late 19th and early 20th centuries.

Keywords: Lan Na manuscripts; material analysis; paper; palm-leaf manuscript; ink analysis; pigment analysis

Zusammenfassung: Vier nordthailändische Manuskripte wurden untersucht; dabei handelt es sich um zwei Palmblattmanuskripte und zwei Papiermanuskripte. Die beiden Palmblattmanuskripte MS 6 und MS 7 wurden nach der traditionellen Methode geschrieben, bei der die Oberfläche der Palmblätter eingeritzt und dann mit Ruß eingerieben wird, wie durch die Identifikation von Kohlenstofftinte bestätigt wurde. In MS 7 zeigte die Tinte die Anwesenheit von geringen Mengen Eisenionen, entweder aus dem Ruß oder vom Griffel, der für die Beschriftung benutzt wurde. Das Papiermanuskript MS 3 wurde mit Eisengallustinte geschrieben, einige Passagen mit einer Tinte auf Methylb-laubasis. Demgegenüber wurde MS4 mit Kohlenstofftinte geschrieben. Das Papier, das für MS3 verwendet wurde, wurde maschinell hergestellt, während bei MS4 *khoi* Fasern (*Streblus asper*) verwendet wurden. Eine Kombination von traditionellen und modernen Pigmenten, z. B. Molybdenumorange, wurde für die Dekoration der Ecken und des Bucheinbands eingesetzt. Die Ergebnisse verbessern unser Verständnis von diesen Manuskripten, erlauben uns aber auch einen Einblick in die schnelle Adaption von modernen Materialien und ihrer Verwendung in der Herstellung von schriftlichen Artefakten aus Nordthailand im späten 19. und frühen 20. Jahrhundert.

Schlüsselwörter: Lan Na Manuskripte; Materialanalyse; Papier; Palmblatt manuskripte; Tintenanalyse; Pigmentenanalyse

1 Introduction

Northern Thailand (Lan Na) finds its roots of cultural identity in the kingdom of Lan Na (1296–1775_{CE}), distinct from that of the Kingdom of Siam, located in Central Thailand, from which the national culture of present-day Thailand descends. The Golden Age of Lan Na (circa 1440–1525 _{CE}), was characterised by territorial expansion, political consolidation, and a thriving Buddhist culture. One of the most remarkable features of Lan Na's cultural identity is its flourishing manuscript culture, characterised by the Lan Na Dhamma script. Developed from the Old Mon script used in the region centuries earlier, Lan Na Dhamma gradually became the main script used in the monastic context for notating both Pali, the religious language in the Theravada Buddhist tradition, and the vernacular Tai Yuan language. This script, initially used for the recording of religious texts, soon replaced the Fak Kham script and the Thai Nithet scripts to emerge as the single 'national script' (Grabowsky 2008, 2011; Veidlinger 2006).

The manuscript culture of the Lan Na in the Dhamma script is characterised by the use of palm-leaves as the preferred writing support, but paper was also introduced at a later time (Helman-Ważny et al. 2020). The earliest palm-leaf manuscript written in the Dhamma script can be dated to around the 15th century CE, while the earliest dated paper manuscript found in Northern Thailand is an astrological treatise (Th: *horasat*) from the Mae Hông Sòn province and was written in 1818 CE; both types of support were likely used earlier than these first dated manuscripts. Palm-leaf manuscripts are in the form of *pothi*, a stack of folios in landscape format that are flipped upward to be read. The folios are usually strung together by means of a thread that runs through the holes pierced into the leaves, normally two, and are often kept between two wooden boards, also secured by the same thread. They can also be further protected by wrapping the manuscripts with cloths (Ciotti 2023). Palm-leaf manuscripts were more widespread than paper manuscripts and preferred for the recording of religious texts. Paper manuscripts were usually in the form of leporellos, but stab-stitched manuscripts were also frequent (Helman-Ważny et al. 2020). They were used more often for secular texts, perhaps hinting that the selection of the writing material depended on the content and the context of production.

The writing techniques and consequently the inks used also depended on the writing support. In the case of palm-leaf manuscripts, the text was incised on the surface with a stylus, before a black paste, traditionally made of soot and resin oil, was applied. After the surface was cleaned, the paste remained in the inscribed traces, highlighting the text (Ciotti 2023). Paper manuscripts were instead simply written with ink pens or quills. Additional pigments, mostly red and gold colours, were used to decorate the pages, the margins, and the covers of the manuscripts. Although the advent of the printing press and industrial paper towards the end of 19th century led to a decrease in the production of palm-leaf manuscripts, the overall manuscript production in the Dhamma script continues to this day, with different writing supports and book forms coexisting.

Several works focussing on the material and techniques employed in the production of palm-leaf manuscripts have been published in the recent years, primarily with a focus on the Indian subcontinent (Rachman 2018; Sharma et al. 2018; Singh and Sharma 2020; Yu et al. 2023). However, studies on the material aspects of manuscripts from Southeast Asia and particularly Northern Thailand are very limited (Burgio et al. 1999; Eremin et al. 2008; Huang 2006; Helman-Ważny et al. 2020, 2021). Therefore, the identification of their materials and production techniques is of paramount importance to their preservation. In fact, the composition of the inks and pigments as well as the material aspects of the writing supports greatly affect the selection of the most effective treatments and conservation strategies. In this work, we are sharing the results of a pilot study carried out on four manuscripts from Northern Thailand,

dating to the 19th and 20th centuries CE. The manuscripts, characterised by different writing forms, writing supports, and writing substances, were examined with non-destructive or micro-destructive methods with the aim of better understanding Lan Na manuscript culture and thus how to better preserve their manuscripts. The relatively recent origin of these manuscripts also helps us in investigating the impact that synthetic pigments and modern technologies had on the local production of written artefacts.

2 Materials and Methods

2.1 Manuscripts Investigated

The four manuscripts (Figure 1) employed for this study are part of a larger group of written artefacts from Southeast Asia that were loaned to the Centre for the Study of Manuscript Cultures for scientific investigation. Three of them (referred to here as MS 3, MS 4, and MS 6) belong to Singkha Wannasai's library in Lamphun, a province in Northern Thailand. It is noteworthy that Singkha Wannasai (1920–1980 CE) was a prominent scholar of Lan Na manuscripts and one of the pioneering lecturers on Lan Na language and literature at Chiang Mai University in the 1970s (Grabowsky 2019, xiii–xv). His private collection, which later became the main part of Singkha Wannasai's library, consists of several Lan Na manuscripts acquired from different provinces in Northern Thailand. MS 7 belongs to Dr. Anant Lualertworakul (born 1968 CE), a former lecturer of Thai etymology, epigraphy, and Lan Na manuscripts at the Faculty of Arts within Chulalongkorn University in Bangkok, who acquired it from a laywoman in Lampang province (Northern Thailand). The selected manuscripts originate from Northern Thailand and are dated or datable *ante quem* to the 19th and 20th centuries, thus making for a convenient corpus for investigating the interactions between modern technologies and traditional manuscript culture in Lan Na.

Thai MS 3 (Figure 1a): The manuscript measures 27.5 × 23.5 cm and consists of 33 paper folios, each approximately 0.15 mm in thickness. Examination of the paper with the naked eye did not show any identifiable sieve structure. In conjunction with the date of the manuscript and unified characteristics within paper sheets, this suggests that the paper might be industrially produced. The folios were bound together by two staples in the left-side vertical edge, mimicking the stab-stitched form which is normally obtained by sewing the folios with a thread. In addition, what appears to be a split pin is found in the centre of the same edge, inserted from the back. The folios consist of a combination of bifolios and single folios: three quires can be clearly recognised (fols 1–7, possibly with one folio missing at the front; fols 15–22;

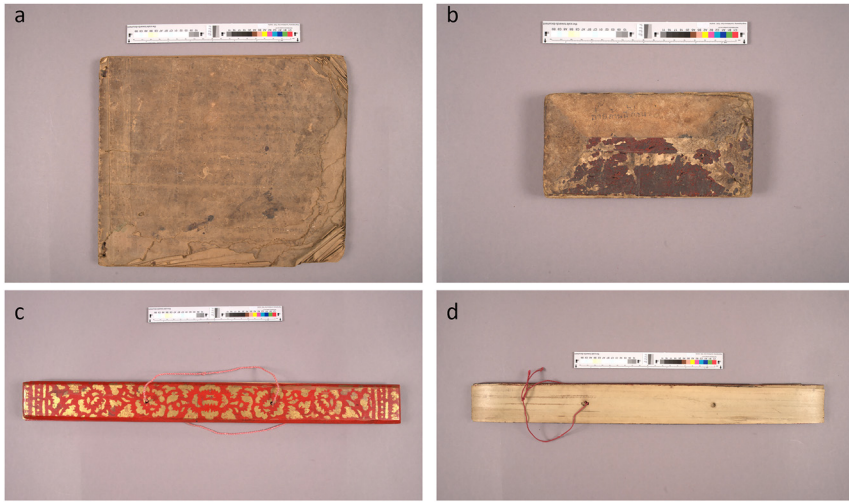


Figure 1: The four manuscripts (a) Thai MS 3; (b) Thai MS 4; (c) Thai MS 6; and (d) Thai MS 7.

fols 27–33, possibly with a folio missing at the end), and the holes of overcast stitching suggest that fols 11–14 were also tied together. The manuscript was mainly written in black ink in Dhamma script and organised in 18 lines of *scriptio continua* per page. Some of the pages have been ruled with pencil. On the top left corner of each page, there is pagination written in Dhamma script, appearing until page 34 (fol. 16v), although one folio is likely missing between fols 10 and 11, as the pagination is lacking numbers 21 and 22. This multiple-text manuscript, written by a single scribe, comprises three texts: a *jātaka* tale titled *Indakumāra* (fols 1r–15v), a general didactic text (fols 16r–22r), and the tale of *Nang Pathawi* (fols 23r–33v). The writing on the front and back covers is so faint that it is barely readable. Structuring paratext addressed to readers is sometimes found, written in black ink by the same hand as the main text, in central Thai script, although the language is Tai Yuan vernacular (Supplementary Material, MS 3 Annotation A–D). Furthermore, the scribe left several colophons using black and blue inks, and regular and coloured pencils. On fol. 15v, corresponding to the end of the first text, there are two colophons and two annotations dating to 1929 CE, written in different scripts, with different media, within the span of a few days (Supplementary Material, MS 3 Colophon A). On fol. 22r, corresponding to the end of the second text, there are two colophons, dating to 1934 CE, indicating that the second text had been written five years later than the first part. Between the two colophons there is an ambiguous *mantra* verse written in blue pencil that is possibly a magical spell and a short instruction of recitation. This combined annotation does not seem to have any connection with the main text and

the colophons and might have been added later in a blank space (Supplementary Material, MS 3 Colophon B). On fol. 33v, the colophon corresponding to the end of the third text can be found. Unfortunately, this text is only partially legible and could not be dated.

Thai MS 4 (Figure 1b): This paper manuscript is in leporello form. It measures 19.5 × 9.5 cm and is made of six connected sheets of paper for a total of 120 panels or folded pages. ‘Folded page’ is the term normally used in Thai disciplines to describe and number the folded parts of the writing support and follows the order of reading of the text, from front cover to back cover on the recto and from back cover to front cover on the verso. This means that fol.1v is not on the back of fol.1r. To avoid confusion, we use the term ‘panel’ to refer to the folded portion of the paper sheet, including both recto and verso. The writing support, consisting of two types of woven paper made in a floating mould, presented with different thicknesses. No sieve structure was observed with a light box. Type 1 corresponds to the majority of the paper used in the manuscript and has a thickness of 0.27 mm–0.33 mm. The thinner paper type (type 2), 0.17 mm–0.21 mm thick, was used twice to insert or replace small parts of the text, for a total of three panels. The sheets of paper had been connected by pasting the extremities together. For the two inserted sheets, one of the extremities had been sewn with a green thread. In some cases, the folds had been damaged and then repaired with self-adhesive tape. The front and back leporello covers are made of a thick (approx. 40 mm), folded, double-layered card-like paper, coated with a red layer. This multiple-text manuscript comprises two texts: *Kammaṭṭhāna* (meditation), consisting of nine parts, and medical recipes. The main texts were written with different hues of black ink. The texts were written in Dhamma and Central Thai scripts and organised in five to eight lines of *scriptio continua* per page, following blind ruling, except for the portion of text on the inserted sheets, where the ruling was made with a greyish writing medium. There are two colophons referring to the two main texts, partially written with pencils, that mention different years of writing: 1855 CE for the part on the recto and 1851 CE for the verso side of the manuscript (Supplementary Material, MS 4 Colophons A–B), showing that the main texts were not copied on a single instance. Full-page illustrations sketched in black ink, and additional texts and notes (in Dhamma script) in black, blue inks, and pencil, are found throughout the manuscript, and were possibly added later in the remaining blank pages of the manuscript.

Thai MS 6 (Figure 1c): This palm-leaf manuscript measures 55.7 × 5.0 cm and consists of 15 folios, made of the leaves of the Talipot palm (*Corypha umbraculifera*), whose margins were gilded with red and gold pigments. A pair of wooden covers protects the manuscript: they measure 55.5 × 5.5 cm, and are painted in red, with decorative floral patterns in gold on the margins. The manuscript is bound by a red thread that passes through the pair of aligned holes of the covers and the leaves and wraps around the bundle. The writing on each leaf runs for five lines of *scriptio*

continua from left to right with some space left on the second, third and fourth lines around the pair of holes. The text was inscribed with a stylus and the incisions filled with black paste. It contains a text titled *Kammavācā* or ‘formulas for higher ordinations’. The colophon was written in the Dhamma script on the first leaf (fol. 1r), stating that the *Kammavācā* manuscript was donated to an unnamed monastery by a laywoman in 1970 CE (Supplementary Material, MS 6). On the top left corner of the same leaf, there is a Pali text that reads “*idaṃ saṅghassa kathinadussaṃ*” (‘This is [the text] of the *Kathin* for the monastic community’). The *Kathin* is a religious ceremony in which laypeople offer monks robes and other alms-gifts after the end of the Buddhist Lenten period, and is not directly related to the main text of this manuscript (Premchit and Doré 1992). This note suggests that this leaf may have been the last folio of another bundle of palm-leaves bearing the *Kathin* text. As this folio was mostly empty, it is possible that the scribe of Thai MS 6 recycled this leaf by incorporating it to the volume hosting the *Kammavācā* and wrote its colophon there.

Thai MS 7 (Figure 1d): This palm-leaf manuscript measures 48.0 × 4.5 cm and is composed of 18 folios, also made using leaves of the Talipot palm (*Corypha umbraculifera*), whose margins were gilded with red and gold colours. Unlike Thai MS 6, it does not have a cover. The manuscript was written in Dhamma script and organised in five lines of *scriptio continua* with space interrupting the second, third and fourth lines to provide space for the red thread that binds the manuscript. The text goes left to right and was inscribed with a stylus and the incisions filled with black paste. The manuscript contains a text titled *Maleyya* or ‘a story of Venerable Maleyya’ (known in Central Thai as Phra Malai). The first folio of this manuscript (1r), displaying in the middle of the recto only the word “jūjaka” in Dhamma script and in pencil, was likely added by mistake. In fact, the holes for the thread do not align with the rest of the bundle. In addition, “jūjaka” is the title of one of the 13 episodes of Vessantara Jātaka. Since both stories of Maleyya and Vessantara Jātaka are chanted in an annual religious ceremony, this folio was perhaps displaced from its original bundle and added to this one. According to the colophon, the manuscript was copied in 1903 CE (CS 1265) from an exemplar called *Wing wòn dòn klang Chiang mai* (Supplementary Material, MS 7). The scribe was a novice named *Abhijaya*, who copied the manuscript in assistance to a senior monk named *Nandā* at Thong Man Tai monastery.

2.2 Analytical Methods

In order to analyse the black writing media, we followed the now well-established protocol developed by the Centre for the Study of Manuscript Cultures (CSMC, University of Hamburg) and Bundesanstalt für Materialforschung und -prüfung (BAM) for the characterization of black inks (Colini et al. 2021; Rabin et al. 2012). The protocol

consists of three steps: an initial screening to identify the type of ink, performed using a handheld Dino-Lite USB microscope AD413T-I2V under white light (visible), ultraviolet (395 nm), and near infrared (940 nm) radiation, followed by Infrared Reflectography beyond 1,500 nm to confirm the presence of carbon and an in-depth analysis using spectroscopic methods, such as X-ray fluorescence (XRF) and Raman spectroscopy. The red and gold paint used to decorate the manuscripts was analysed with XRF, Raman spectroscopy and VIS-NIR spectrophotometry. Writing supports of palm-leaf and paper manuscripts were analysed using Diffused Reflectance Fourier Transform Infrared spectroscopy (DRIFT-FTIR). The papers of Thai MS 3 and 4 were also sampled and subjected to additional investigation with optical and digital microscopy. The analysis was performed on multiple points per folio, spanning around 5 to 6 folios throughout the manuscript.

2.2.1 Digital Microscopy

Micrographs of the black inks were obtained using a handheld Dino-Lite USB digital microscope (AD413T-I2V) under white light (visible), ultraviolet (395 nm), and near infrared (940 nm) radiation. The behaviour of the inks at these wavelengths is used to classify them based on their typology (Mrusek et al. 1995). Carbon inks remain unchanged under the whole spectrum of infrared radiation, while plant-based inks appear completely transparent under the 940 nm illumination. Iron-gall inks exhibit a decrease in opacity at 940 nm, turning completely transparent only at higher wavelengths (around 1200–1500 nm). Additionally, ultraviolet radiation can also hint towards the presence of metal ions and tannins in the ink. Metal ions absorb UV radiation, while tannins can act as fluorescence quenchers, enhancing the contrast between the fluorescing writing support and the ink (Fuster López et al. 2020; Soares, Mateus, and de Freitas 2007; Zhang et al. 2005).

2.2.2 Infrared Reflectography (IRR)

To verify the presence of carbon, the APOLLO Infrared Reflectography Imaging System (IRR) from OPUS Instruments was used in combination with a Long Wave Pass Filter (LWP1510, range 1510–1700 nm) with two 20 W halogen lamps as light sources. The filter allowed us to limit the range of infrared radiation to higher wavelengths – specifically the region wherein iron-gall inks are transparent.

2.2.3 X-Ray Fluorescence Spectroscopy (XRF)

X-ray fluorescence spectroscopy provides us with an easy, non-destructive method of determining the elemental composition of a wide array of inorganic materials.

However, XRF spectroscopy cannot be used for the detection of organic materials. Three different spectrometers were employed for the elemental analysis of inks and pigments. The dimensions of the area of interest, the total number of measurements per manuscript and the duration of a single measurement influenced the choice of instrument. Spot analysis was carried out using Bruker/XG Lab Elio with a Rh X-ray tube, a 25 mm² large-area silicon drift detector (SDD) with an interaction spot of 1 mm. The measurements were conducted at 40 kV voltage, 80 μ A current, with a measurement time of 120 s per spot. Line scans were acquired with Bruker ARTAX with a Mo X-ray tube, polycapillary X-ray optics, an electrothermally cooled Xflash SDD detector, and an interaction spot of 100 μ m. The measurements were conducted at 50 kV voltage, 600 μ A current, and with a measurement time of 30 s per spot. Spatial maps were obtained using Bruker M6 Jetstream with a Rh X-ray tube, polycapillary X-ray optics, a 50 mm² Xflash SDD detector, and an adjustable measuring spot ranging from 50 to 650 μ m. The measurements were conducted at 50 kV voltage and 600 μ A current, with a spot size of 50 μ m, an acquisition time 30 ms per spot and a step size of 50 μ m.

2.2.4 Raman Spectroscopy

Spot analysis of carbon inks and red and gold pigments was carried out using a Renishaw inVia Raman spectrometer equipped with a 100 mW 532 nm laser and a 300 mW 785 nm laser and with a 100 \times objective. Most analyses were conducted with the green laser, with 10–50 accumulations of 1–10 s at \sim 1–2 mW. The 785 nm laser was employed to further characterise the red pigments, with 10–50 accumulations of 1–10 s at \sim 2–6 mW.

2.2.5 VIS-NIR Spectrophotometry

The nature of the red pigments was also investigated using Malvern-Panalytical Analytical Spectral Devices (ASD) LabSpec 4 Hi-Res spectrophotometer, measuring the reflectance spectrum in the range 350–2500 nm with a scanning time of 100 ms. The measurement was carried out using a small diameter reflectance probe in combination with a probe holder, with a spot size of 2 mm. A white standard was used as reference. The first derivative of the spectrum was used for the characterisation of the pigments.

2.2.6 Fourier Transform Infrared Spectroscopy (FT-IR)

The writing supports were analysed using the portable Agilent 4100 ExoScan FT-IR spectrometer using Diffuse reflectance (DRIFTS) mode with a measuring spot of

1 cm, spectral range 650–4000 cm^{-1} , 256 scans per measurement and a resolution of 4 cm^{-1} .

2.2.7 Optical Microscopy

The two paper manuscripts were subjected to additional investigation to discern paper and pulp characteristics and fibre composition of the paper writing supports. Paper characteristics were examined with a Keyence VHX-500 digital microscope, while fibre sample examination was conducted using an Olympus BX51 Transmitted/Reflected light microscope, with an affixed Olympus UC30 camera for the acquisition of micrographs via the Olympus Stream software suite. The samples were examined under a range of magnifications, from 20 \times to 200 \times , under both white and polarised light.

3 Results

3.1 Thai MS 3

Thai MS 3 was penned predominantly with black ink, with a few sections written in pencil and blue ink (e.g., fol. 15v; Figure 2a). The digital microscopy images of the black and blue ink showed a decrease in opacity under near-infrared radiation (Figure 2b). XRF maps and line-scan measurements of the black ink indicated the presence of iron and calcium in the ink, suggesting the use of iron-gall ink in the manuscript (Figure 2c). No characteristic elements were detected when examining the blue ink, suggesting a blue organic dye. Methyl violet dye was identified by Raman spectroscopy (Figure 2d) thanks to its characteristic peaks at 1634 cm^{-1} , 1429 cm^{-1} , 1386 cm^{-1} , 910 cm^{-1} , 809 cm^{-1} , 481 cm^{-1} , and 446 cm^{-1} (Borba, Honorato, and Juan 2015; Zaffino et al. 2017).

The writing support was subjected to both microscopic investigation and characterisation using FTIR spectroscopy. The FTIR spectra (Figure 2e) showed peaks corresponding to cellulose (3434 cm^{-1} , 2900 cm^{-1} , 1430 cm^{-1} , 1374 cm^{-1} , and 1030 cm^{-1}), starch (1000 cm^{-1} and 989 cm^{-1}), and carbonate (871 cm^{-1} and 1410 cm^{-1}), suggesting that the paper had been treated with calcium carbonate and sized with starch (Calvini and Gorassini 2002).

Library records accompanying the book indicated the paper to have been made from *khoi* (*Streblus asper*) or another type of mulberry fibre. However, under optical microscopy, fibres with window-like pits and frequent swellings were found in abundance (Figure 3b and c), alongside with fibres with thin lumens that exhibited reticulate perforations (Figure 3d), which indicated that they were fibres of softwood

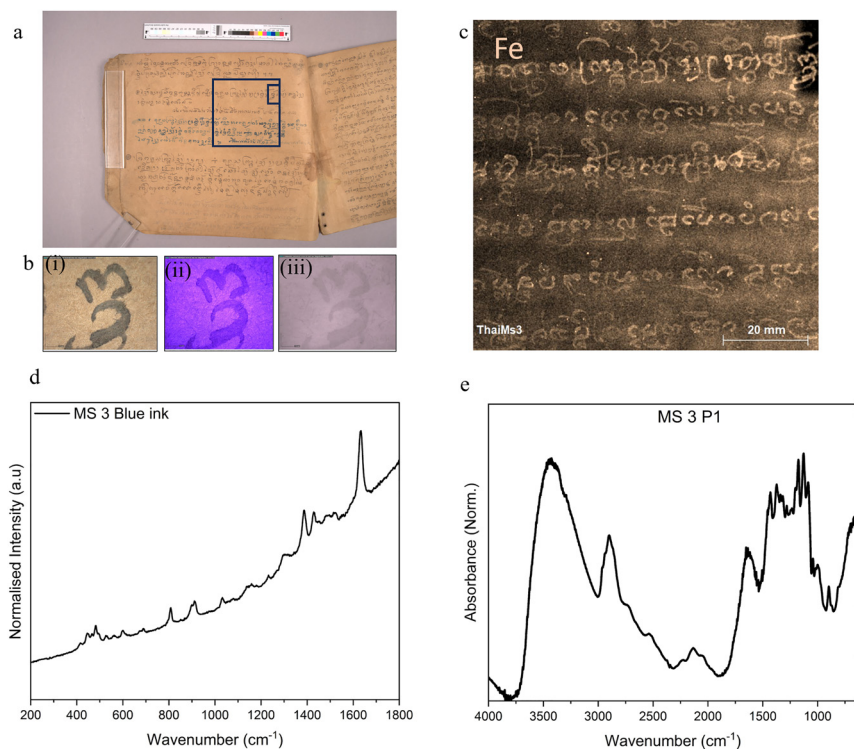


Figure 2: Thai MS 3 (a) fol. 15v, indicating the regions analysed; (b) Digital microscopy images under (i) visible (ii) ultraviolet and (iii) infrared illumination; (c) XRF spatial map indicating the presence of Fe in the black ink, including contributions from the ink on the other side of the folio; (d) Raman spectrum of the blue ink; (e) DRIFTS-FTIR spectrum of the paper.

pulp. Additionally, a few singular smooth fibres were found, likely artificial impurities from the industrial papermaking process. No fibres of *khoi* or mulberry could be found. Examination of the surface with a digital microscope showed a regular topographical pattern that pointed toward an industrial origin (Figure 3a).

3.2 Thai MS 4

Visual examination of the leporello manuscript Thai MS 4 suggested the presence of two black inks employed for the main texts (fols 13r–14r; Figure 4a). They bear different shades of colour, one paler than the other. The more recent additions on the manuscript, the notes in pencil and blue pen, as well as the illustrations, were not

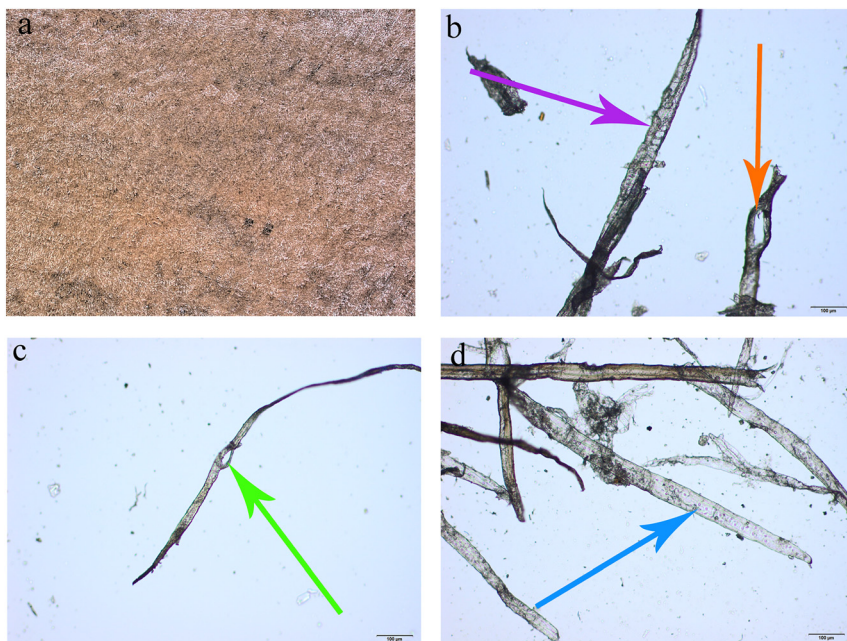


Figure 3: Optical microscopy result for Thai MS 3 (a) digital micrograph showing a regular pattern on the surface of MS 3; (b) and (c) window-like pits and frequent swellings; (d) wide fibres with thin lumens, exhibiting reticulate perforations.

considered for the analysis. According to the results of the protocol, both inks are carbon-based, as they did not lose opacity in NIR radiation (Figure 4b and c) and clearly showed the peaks of carbon at 1365 cm^{-1} and 1601 cm^{-1} in their Raman spectra (Figure 4d). No additional trace elements were detected with XRF spectroscopy.

The FTIR spectra of the writing supports of MS 4 indicated the use of two different papers. Peaks corresponding to kaolin at 3691 cm^{-1} and 3919 cm^{-1} were present in the spectra of type 1 paper, the thicker paper that was predominantly used in the manuscript, while kaolin spectra were absent in type 2 paper, the thinner paper used for the inserted sheets (Figure 4e, with type 1 represented by spectrum P4 and type 2 by spectrum P9). Peaks corresponding to cellulose (3434 cm^{-1} , 2900 cm^{-1} , 1430 cm^{-1} , 1374 cm^{-1} , and 1030 cm^{-1}), starch (1000 cm^{-1} and 989 cm^{-1}) and carbonate (871 cm^{-1} and 1410 cm^{-1}) were common for both papers. Lignin peaks were not observed.

Under digital microscopy, the papers appeared uneven, presenting swirls and accumulations of fibres consistent with woven paper made in a floating mould

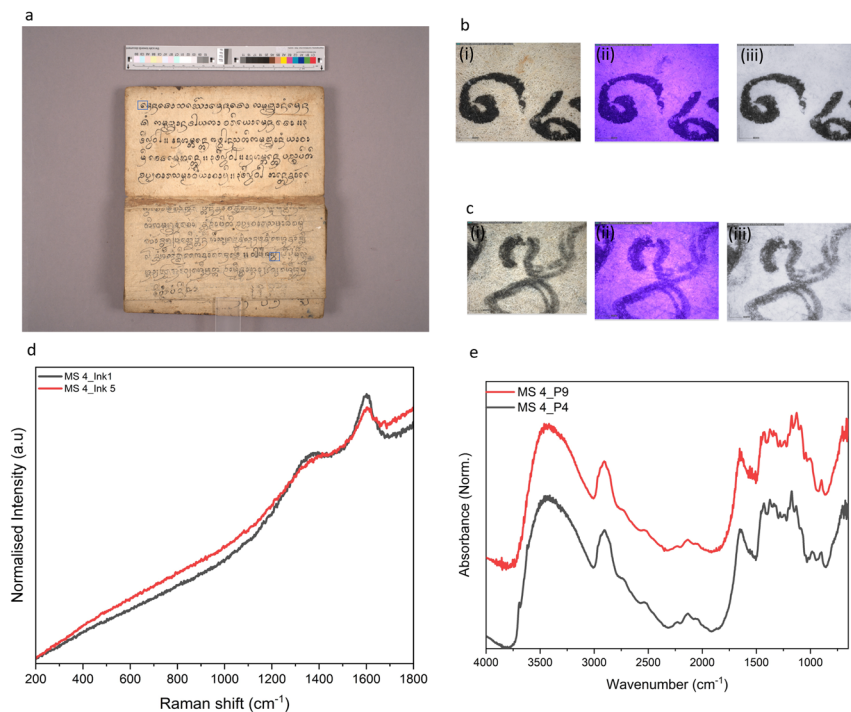


Figure 4: Thai MS 4 (a) fols 13r–14r with the indication of the areas analysed; (b) and (c) Digital microscopy images of the inks with different hue under (i) visible (ii) ultraviolet and (iii) infrared illumination; (d) Raman spectra of the two corresponding inks: ink 1 (used in Type 1 paper) and ink 2 (used in Type 2 paper) ; (e) DRIFT-FTIR spectra of the two paper types: P4 (Type 1) and P9 (Type 2).

(Figure 5a). They seemed to have undergone only a low degree of beating: on some folios, large fragments of plant material are still visible, and under microscopy, thick bundles of unseparated fibres occurred (Figure 5b). Under optical microscopic observation, the fibres in both MS 4 paper types were long and narrow, with transparent membranes, i.e., primary fibre walls, enveloping the fibres (Figure 5c). The fibres presented an abundance of phytoliths, occasional broadening, and frequent cross markings (Figure 5d). In this case, the library records stating that the paper was made of mulberry were confirmed by microscopic observation, as the dominant fibres present in both the writing supports were *khoi* (*S. asper*) fibres – although a few cotton fibres were also found. The presence of cotton was particularly noticeable in Type 2, adding another parameter for the differentiation of the two papers. The difference between the two types of paper in MS 4 appeared to be, beyond the presence of kaolin as a filler in Type 1 and the thicknesses, a matter of the proportion of mulberry to cotton.

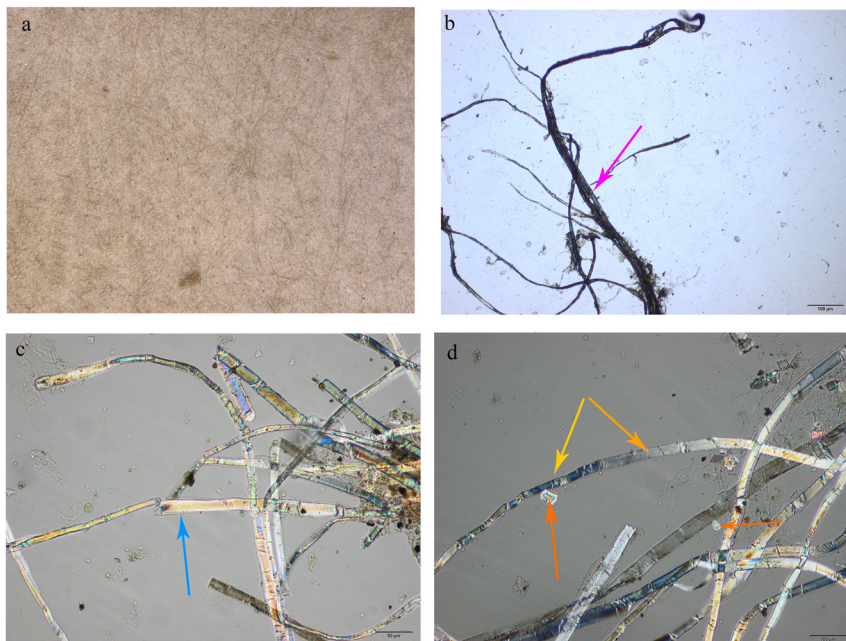


Figure 5: Optical microscopy results for Thai MS 4 (a) uneven paper surface with swirls and accumulations of fibres, typical of woven paper made in a floating mould; (b) thick bundle of unseparated fibres, indicating a low degree of beating (c) long and narrow fibres exhibiting loose transparent membranes; (d) phytoliths, fibre broadening, and frequent cross markings, typical of *khol* fibres.

Examination of the red pigment on the leporello cover using XRF spectroscopy indicated the presence of a significant amount of mercury, pointing to the use of vermilion. This was confirmed by the characteristic peaks at 253 cm^{-1} , 287 cm^{-1} , and 343 cm^{-1} in the Raman spectrum (Figure 6a). Examination of the cover using FTIR spectroscopy (Figure 6b) showed peaks at 3433 cm^{-1} , 2930 cm^{-1} , and 252 cm^{-1} that can be attributed to O–H and C–H stretching vibrations respectively. The peaks at around 1700 cm^{-1} , 1560 cm^{-1} , 1460 cm^{-1} , 1245 cm^{-1} , and 1070 cm^{-1} are indicative of lacquer, possibly urushiol, a type of oriental lacquer (Hao et al. 2017; Narita et al. 2023; Okahisa, Narita, and Yoshimura et al. 2019).

3.3 Thai MS 6

The ink of this palm-leaf manuscript (Figure 7a) was identified as carbon ink based on results from digital microscopy (Figure 7b) and Raman spectroscopy (Figure 7c).

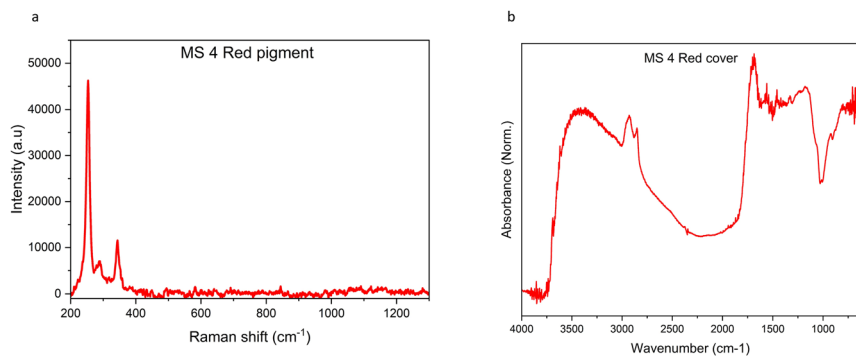


Figure 6: Analysis of the red pigment present in the cover of Thai MS 4 (a) Raman spectrum; (b) DRIFTS-FTIR spectrum.

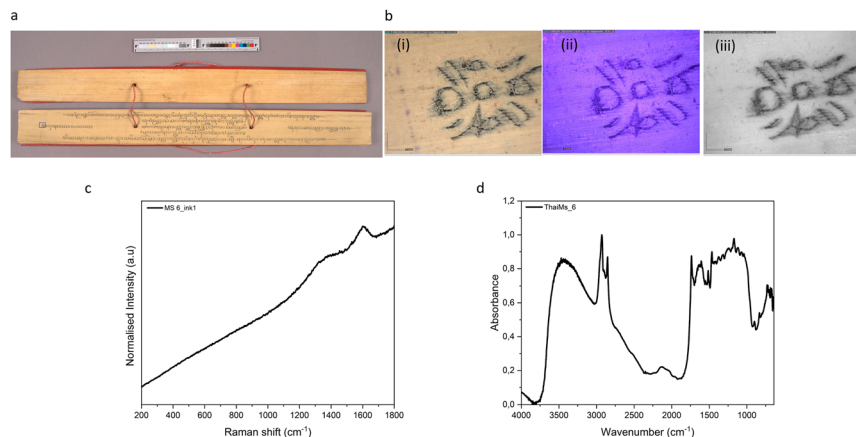


Figure 7: Thai MS 6 (a) fol. 2r with the indication of the area analysed; (b) Digital microscopy images under (i) visible (ii) ultraviolet and (iii) infrared illumination; (c) Raman spectra of the ink; (d) DRIFTS-FTIR spectrum of the palm leaf.

The FTIR spectrum of the palm leaf (Figure 7d) showed peaks at 2926 cm^{-1} and 2853 cm^{-1} corresponding to the stretching vibrations of the CH_2 group. The peak around 1738 cm^{-1} corresponds to the $\text{C}=\text{O}$ group and can be attributed to both pectin and hemicellulose. The peaks at 1600 cm^{-1} , 1514 cm^{-1} , 1175 cm^{-1} , and 833 cm^{-1} could be attributed to lignin, while peaks at around 1650 cm^{-1} and 940 cm^{-1} are characteristic of pectin. Peaks corresponding to cellulose were observed at 3403 cm^{-1} , 2917 cm^{-1} , 1377 cm^{-1} , and 1026 cm^{-1} (Boeriu et al. 2004; Chu et al. 2023; Geminiani et al. 2022).

This manuscript additionally shows decoration in red and gold colour on the margins of the palm leaves and on the wooden covers. The XRF investigation of the red pigment on the wooden covers (Figure 8a) indicated the presence of lead, chromium, and traces of molybdenum. Raman spectroscopy (Figure 8b) confirmed the use of molybdenum orange, featuring characteristic peaks at 824 cm^{-1} and 1179 cm^{-1} (Chua et al. 2016). The golden colour on the cover was determined to be brass, as clearly indicated by the increase in the amounts of Cu and Zn in the XRF spectra. In contrast, the golden decoration in the edges is characterised by the presence of gold (Au) (Figure 8c), while the red pigment showed no difference to unpainted areas in elemental composition when examined using XRF, indicating the use of an organic dye. Reflectance measurement of the red pigment suggests the use

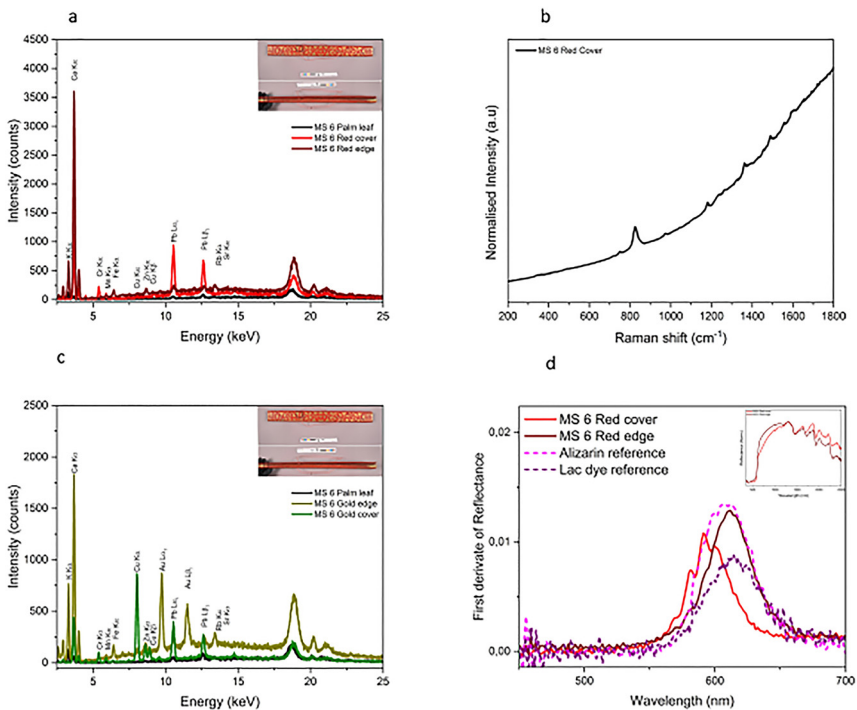


Figure 8: Thai MS 6 (a) XRF spectra of the support and the red pigments present in the decoration on the wooden cover and the edges; (b) Raman spectrum of the red pigment on the cover; (c) XRF spectra of the support and the gold pigments present in the decoration on the wooden cover and the edges; (d) Reflectance measurement (inset) and first derivative plot of reflectance of the red pigments and references (IRUG database), suggesting the use of either Lac dye or Alizarin for the red in the edge.

of lac dye or alizarin (Figure 8d). The difference in the red and golden pigments used for decorating the covers and the margins suggests that they were produced in separate manufacturing contexts.

3.4 Thai MS 7

The ink of this palm-leaf manuscript (Figure 9a) showed no significant change in opacity under infrared radiation (Figure 9b). XRF analysis of the ink indicated the presence of trace amounts of iron (Figure 9c), although both Raman spectroscopy and IRR (Figure 10a) confirmed the ink to be carbon-based.

The FTIR spectrum of the palm leaf showed peaks at 2930 cm^{-1} and 2854 cm^{-1} corresponding to the stretching vibrations of the CH_2 group while the peaks at 1600 cm^{-1} , 1520 cm^{-1} , 1208 cm^{-1} , 1179 cm^{-1} , and 812 cm^{-1} could be attributed to lignin. Peaks corresponding to cellulose were also observed at 3422 cm^{-1} , 2904 cm^{-1} , 1425 cm^{-1} , 1377 cm^{-1} , and 1047 cm^{-1} ; the peak at around 1736 cm^{-1} was associated to the presence of hemicellulose and pectin, and the peak at 1660 cm^{-1} was attributed to pectin (Figure 9d).

While this palm-leaf manuscript does not have an accompanying wooden cover, its margins are decorated with red and gold. The XRF results of both the red and gold pigments showed the presence of barium, suggesting an undercoating composed of barium sulphate. The use of barium sulphate in the pigment was further confirmed by the peaks at 1187 cm^{-1} , 1086 cm^{-1} , and 985 cm^{-1} in the FTIR spectra (Correa et al. 2023). Lead was detected with XRF in the red pigment (Figure 10b). However further attempts to characterise the red pigment using Raman spectroscopy did not yield any results due to a high degree of fluorescence similar to Thai MS 6. gold (Au) was detected with XRF in the golden decoration on the margins (Figure 10c).

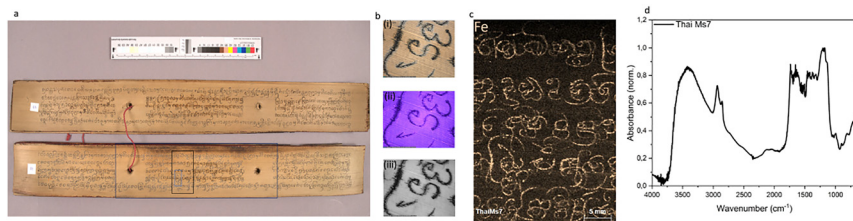


Figure 9: Thai MS 7 (a) fol. 6v–7r with the indication of the areas analysed; (b) Digital microscopy images under (i) visible (ii) ultraviolet and (iii) infrared illumination; (c) XRF spatial map indicating the presence of Fe in the ink (relative intensity); (d) DRIFTS-FTIR spectra of the palm leaf.

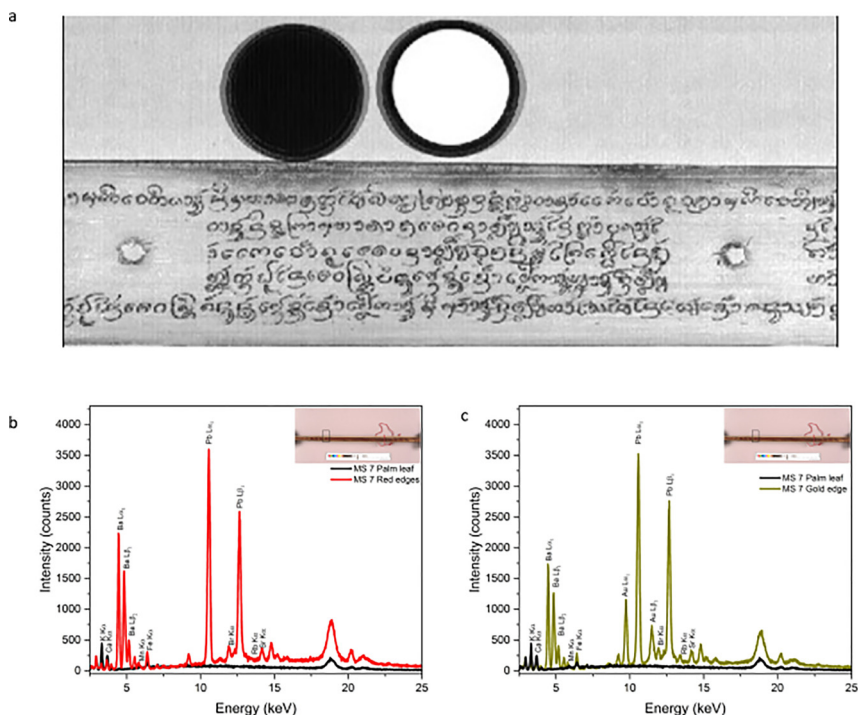


Figure 10: Thai MS 7 (a) IRR image of a section of the palm leaf; (b) XRF spectra of the support and the red pigment in the margins of the manuscript; (c) XRF spectra of the support and the gold pigment in the margins of the manuscript.

4 Discussion

The results of our investigations showed that a variety of writing materials were used to produce these manuscripts (Table 1). As expected, both palm-leaf manuscripts were inked with a soot paste, as indicated by the detection of carbon. However, we observed the presence of iron in the writing of Thai MS 7, suggesting either that this element was part of the carbonaceous paste, in the form of impurity, or that an iron stylus had been employed to inscribe the text.

The case of the two paper manuscripts was more complex, as they had been written with multiple media. The difference in the appearance of the inks used for the main texts of Thai MS 4 could not be ascribed to different compositions, since all the areas analysed show the use of pure carbon inks, without any trace of other elements. The manuscript, however, was copied on at least two separate occasions, four years apart. As carbon inks are usually produced in sticks, it is possible that the

Table 1: Summary of the results.

Manuscript	Writing supports	Inks	Pigments
Thai MS 3	Industrially-made softwood pulp paper	Black: Iron gall ink Blue: Methyl violet ink	–
Thai MS 4	Handmade bast paper from <i>khoi</i> (<i>Streblus asper</i>) with minor additions of cotton	Carbon ink	Cover Red: Vermillion
Thai MS 6	<i>Corypha umbraculifera</i> (Talipot palm)	Carbon ink	Cover Red: Molybdenum Orange Cover Gold: Cu-Zn Margins Red: Organic dye (Alizarin/ lac dye) Margins Gold: Gold (Au)
Thai MS 7	<i>Corypha umbraculifera</i> (Talipot palm)	Carbon ink with iron	Margins Red: Lead oxide Margins Gold: Gold (Au) Barium sulphate base

same ink stick was used, diluted, to write during the years, and that the hue variation is simply due to a different concentration of the pigment. Thai MS 3 was written in iron-gall ink, pencil, and methyl violet ink. First synthesised towards the end of the 19th century, aniline-based dyes were widely used to produce synthetic inks (Mitchell 1922). This included methyl violet, which was employed for fountain pen inks and which continues to be an important component of modern ballpoint pen inks (Brunelle and Crawford 2003). Analytical investigations by Cesaratto et al. (2018) on the introduction of synthetic dyes in Japanese woodblock prints suggest that methyl violet was commercially available by 1875 CE in Asia. Since the colophon of Thai MS 3 dates it to 1929 CE (Supplementary Material, MS 3 Colophon A1 fol. 15r), it is likely that the text had been written with a fountain pen using a methyl violet-based ink.

The pigments used in the manuscripts also range from traditional to synthetic modern materials. While the cover of the leporello manuscript (MS 4) was most likely decorated with a vermilion paint lacquered with urushiol, a traditional technique widely employed in Asia, the presence of the industrial pigment molybdenum orange in the wooden covers of Thai MS 6 is surprising. Also known as molybdate red, this colour was patented in Germany by Lederle in 1930 and commercialised in the following years in the US, Germany, and Belgium. According to research on the history of modern artists' synthetic pigments conducted by De Keijzer (1990; 2002), the pigment was of no real importance, at least in the European artistic community. However, it was identified by Chua et al. (2016) as having been among the pigments used by the Japanese artist Haku Maki (1924–2000) in 1970–71. We do not know the

exact production date of Thai MS 6, but the colophon indicates that it had been donated in 1970, and the wooden cover might be contemporary to the donation. Interestingly, the golden pigment used in the cover was identified as brass, a cheaper material compared to the real gold used to decorate the margins of the same manuscript. The red pigment of the margin decoration is an organic dye, identified as lac dye or alizarin. The different materials used in the decoration of the covers and the margins of the manuscript suggest that they were not painted in the same context, and it is possible that the wooden covers were produced in series using cheaper synthetic materials. The pigments identified in Thai MS 7 – pure gold and, likely, red lead – seem to confirm that traditional materials were preferred for the decoration of the margins of palm-leaf manuscripts.

Concerning the writing supports, the palm leaves of both manuscripts showed very similar results upon examination using FTIR spectroscopy. However, the peaks corresponding to pectin and hemicellulose in the spectra of MS 7 have a lower absorption intensity compared to the corresponding peaks in MS 6. The lower intensity was likely a result of the degradation of hemicellulose and pectin as observed by Chu et al. 2023. It is possible that this degradation could have been induced by a different storage environment, with MS 7 likely preserved in a more humid setting, resulting in a relatively faster degradation of hemicellulose and pectin. It is more likely, however, that this difference originates from a distinct manufacturing process. The production process for palm leaves often involves boiling them in water together with other plants or plant extracts such as tamarinds, thereby influencing its chemical and mechanical properties, including the amount of polysaccharides. It is possible that the leaves used for these two manuscripts were boiled for different lengths of time, or with a different mixture of materials, resulting in relatively different amounts of pectin and hemicellulose left in the leaves. Independently to the cause of this minimal discrepancy, we can conclude that in both cases the palm leaves were processed according to the conventional method, as we could not identify any unusual substances having been employed in their manufacture. Overall, aside from the use of modern pigments in the cover of MS 6, the materials employed for the two palm-leaf manuscripts conforms with technological descriptions of manuscript making in the broad palm-leaf manuscript culture from Southeast Asia as recorded in literature.

The two paper manuscripts, instead, showed that the Lan Na adopted contemporarily available but then-nascent technologies, such as industrial papermaking, and modern fountain pen, in a remarkably short interval and integrated them into their traditional book forms and manuscript culture.

MS 3 was predominantly written in iron-gall ink, a technology that had been available, at least in Europe, for some 14 centuries. Methyl violet, as has been discussed above, was a relatively recent invention at the time of the manuscript's

production, but would have been easily manufactured or transported to Lan Na. The paper writing support of MS 3, however, was made from industrially-produced wood pulp, commercially available only by 1867 CE, and even then, only from a few scattered papermakers in the West (Hunter 1978). This allows about 62 years for the technology to have travelled from the West to Lan Na, a strikingly brief timeframe, especially given the long and robust history of hand papermaking in Thailand. We assume that the paper used here was imported, although it is also possible that papermaking machinery had already been imported or built in Siam at the time of production. MS 3 is also interestingly bound. Folios 11–14 were overcast, but the thread was removed before the manuscript was bound in the form we see today. It uses two integrated metal-based systems to keep the folios together: the manuscript was stapled twice in the left-side vertical edge and a split pin, now damaged, was inserted from the back in the centre of the same edge. These elements are placed in a way that is reminiscent of the traditional stab-stitched form, obtained with a thread, although this sewing is normally found on the head edge instead of the left edge. Additionally, the folios are folded to form bifolios and then sewn together at the edge opposite of the fold, as in Chinese stabbed-sewing, while in the case of MS 3 they are either single leaves or bifolios folded into quires, in the Western fashion. In any case, the concurrence of quasi-traditional over stitching with staples and a split pin in the same manuscript is a striking and fascinating snapshot of the complex interplay between traditional Lan Na manuscript culture and incipient technologies in 1930s Thailand.

MS 4 is remarkable for indicating an even more rapid adoption interval. While the leporello form is traditional, the paper type marked type 1, constituting the preponderance of the paper in the manuscript, contained kaolin (Helman-Ważny et al. 2020). Hunter (1978) wrote that China clay, or kaolin, was first used as a paper filler circa 1807 CE. The colophons of MS 4 date it *ante quem* to 1851 CE, meaning that the technology of kaolin as paper filler took at most about 44 years to travel to Siam. There is little doubt as to the locality of the paper fibers in this case – it was positively identified as *khoi*, a species that grows only in Southeast Asia and which has been used for papermaking in Thailand since the late 17th century (Khantayanuwong et al. 2021).

5 Conclusions

The analysis of these four manuscripts offered us some interesting results and insights into contemporary production processes and materials. The two religious texts take the form of palm-leaf *pothi* manuscripts, inscribed in the traditional way by incising text on the leaves and then filling the incisions with carbon-based paste. One of the two paper manuscripts has a leporello form, frequently employed in previous

centuries, while the other has a modified version of the stab-stitched form, also common in northern Thailand, with two staples partially substituting the stitching. Intriguingly, modern materials and technology feature prominently in these production processes, often very soon after their introduction. That a centuries-old manuscript culture should so quickly adopt and integrate modern materials is not only a display of the adaptability of Lan Na manuscript culture, but also the sheer ability of technology to inflect or even outright change manuscript production processes that had remained largely static for significant amounts of time. These results also expand our knowledge of the materials and techniques that were used in Southeast Asia, and specifically in Northern Thailand, thereby helping scholars and conservators alike in understanding the materials and developing strategies for the preservation and conservation of these manuscripts.

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