Characterisation and analysis of early Qur’an fragments at the Library of Congress

Yasmeen Khan
Senior Rare Book Conservator, Library of Congress, Washington.

Sophie Lewincamp
Paper Conservator, Australian War Memorial, Canberra.

Abstract
During the late 1820s, early 1920s, and 1990s, the Library of Congress African and Middle Eastern Division acquired a large collection of Arabic script calligraphy sheets, ranging from the 8th to the 17th centuries. More recently, a sub-group on parchment dated from the 8th through to the 13th century was identified as constituting part of the ‘treasures’ of the Library of Congress and, required greater bibliographic and security controls. An art historian was contracted to the conservation office to identify the calligraphy style and texts, and place them in their historical milieu. After the initial art historical research was done, the Conservation Division was asked to characterize the materials used to produce the now-identified Qur'an fragments.

The main aim of the project was to identify colourants used on the parchment sheets. Microscopic samples of pigment, ink and parchment were removed for analysis with a Scanning Electron Microscope (SEM) and X-ray microanalysis was used to determine the composition of the pigments. The links were observed under the microscope and with a Multi Spectral camera. Digital images of the parchments fragments were shot in various modes: under infrared (IR), false colour infrared 2 and ultra violet (UV) fluorescence. Observations were made of the sinking of links through the absorption of UV energy by metallic pigments, the fusing of iron oxide in IR2, and structure of the parchment through scattering of energy in UV fluorescence. Further analysis work continues on this project with the potential use of XRD and parchment DNA analysis. The information that has already been gathered has provided insights into differences in methods of calligraphy. The aim of the project has expanded as the Library of Congress hopes to create a baseline characterisation of the Qur’an parchment fragments and open an area of comparison for other fragments of similar date, script and region scattered in repositories around the LRS.

Introduction
During the late 1830s, early 1930s and 1990s, the Library of Congress’ African and Middle Eastern Division acquired a large collection of Arabic script calligraphy sheets. The collection included 355 calligraphy sheets, ranging from the 8th to the 17th centuries. Most, if not all the early sheets were acquired from the New York and Paris-based firm of Kirkor Minassian. The majority of these calligraphy sheets were written on paper with a smaller group of Qur’anic leaves executed on parchment. Through the value of individual sheets was acknowledged at the time of acquisition, little bibliographic work had been done on the collection. By the mid-seventies the early parchment Qur'an leaves or fragments had undergone conservation treatment and were tension mounted into mats by the Restoration Office conservators under the direction of Christopher Clarkson and Peter Waters.

In 2002, the Library of Congress (LC) decided to mount the Calligraphy Collection on their website and hired Dr. Christine Gruber, an Assistant Professor of Art History at Indiana University to do a bibliographic and codicological description of all the calligraphy sheets as a preliminary step. In her larger descriptive work on the collection, Dr. Gruber singled out some of the Qur’an parchment leaves as being amongst the earliest extant Islamic material in the Library of Congress as they dated from the 8th to 10th centuries. It is axiomatic in conservation that the better the understanding of the art historical aspects of an object, the more nuanced and contextualized the interpretation of the materials that comprise the artwork. When the African and Middle Eastern Division (AMED) expressed interest in discovering more about the pigments in their now well-described collection of Qur’an verses, the Conservation Division was able to develop a methodology to answer some of the curatorial concerns. AMED curators were interested in knowing if the media could be characterized so as to open an area of comparison with calligraphy sheets of similar date, script, and region within the collection, and in other cultural repositories. To this end, AMED was willing to allow microscopic sampling of pigments, ink and parchment from the original leaves. A representative sample group of eleven parchment leaves from the 8th through to the 12th centuries C.E. was chosen for this research project.

Most of the eleven parchment leaves are from the 9th to 10th century Qur’ans or collections of Qur’anic verses and have been individually identified according to the classification system developed by Francois Derco. Eight of the fragments are written in various iterations of the Kufic script. One sheet, dating from the 9th century is written in Hijazi 1 and has no original decorative elements. Two leaves (9 and 10 from left, 1/2 and apparently pertaining to the same manuscript are in Magribi script common to Morocco and Moorish Spain and have been dated to 1250-1350 C.E. Aside from the latter group, all of the calligraphy leaves pertain to the Middle East but have not been localized to a narrow geographical area.
The pigments

Background

The decoration on the parchment leaves varies. Some decorations are quite modest, showing only calligraphic text with red pigment dots as vowel markers and diacritical marks (leaf no. 9, 15, 16 & 17), while others are more elaborate with detailed illustrations in gold, red, green, and occasionally blue pigments (leaf no. 7, 10, 20 & 15.4.28).

Calligraphers and practitioners of the book arts in the Middle East during the Middle Ages appear to have had access to a rich palette at their fingertips. A few literary texts that specifically address artists’ materials from the Middle East, notably Persia and India, and dating from the 12th-13th century, have been translated into English. They enumerate a varied and extensive array of materials for the calligrapher and artist. The North African Zirid emir, Ibn Badis’ book on bookmaking and its allied arts, Kitab ‘under al-hattah wa ‘uddat dhawa‘ al-dibbi, written in the 10th century, provides earlier recipes for colourants.23 His list of raw materials used to make coloured inks and pigments is exhaustive and includes plants, insects and inorganic materials.

Table 1 Identification and background information of sample leaves.

<table>
<thead>
<tr>
<th>Leaf No.</th>
<th>Date C.E.</th>
<th>Script &amp; Style</th>
<th>Qur’anic Verse &amp; Sarab</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>9th-10th</td>
<td>Kufi (CII)</td>
<td>Complete surah Al-A’la</td>
</tr>
<tr>
<td>7</td>
<td>9th</td>
<td>Kufi (D I)</td>
<td>Verses 10-11, al-Furq</td>
</tr>
<tr>
<td>9</td>
<td>9th-10th</td>
<td>Kufi (D)</td>
<td>Verses 24-78, al-Hajj</td>
</tr>
<tr>
<td>10</td>
<td>9th</td>
<td>Kufi (D I)</td>
<td>Verses 85-88, al-Anam</td>
</tr>
<tr>
<td>13</td>
<td>9th-10th</td>
<td>Kufi (D)</td>
<td>Verses 19-31, al-Anam</td>
</tr>
<tr>
<td>16</td>
<td>9th</td>
<td>Kufi (D I)</td>
<td>Verses 68-75, al-Anam</td>
</tr>
<tr>
<td>17</td>
<td>8th</td>
<td>Hijaz 1</td>
<td>52-54, al-Saba’</td>
</tr>
<tr>
<td>18</td>
<td>1250-1300</td>
<td>Maghribi</td>
<td>130-140, al-Majdub</td>
</tr>
<tr>
<td>19 left</td>
<td>1250-1300</td>
<td>Maghribi</td>
<td>60-63, 142-144, al-Anam</td>
</tr>
<tr>
<td>20</td>
<td>10th</td>
<td>Kufi (New Style III)</td>
<td>60-80, al-Anam</td>
</tr>
<tr>
<td>154.28b</td>
<td>9th</td>
<td>Kufi (New Style III)</td>
<td>1-5, al-Haqq</td>
</tr>
</tbody>
</table>

The scribes and copyists during the early period created unadorned texts for example, the 8th century leaf, leaf number 17, written in the Hijazi style manifests a concern with clear transmission of the Qur’anic text rather than the ornamentation of the calligraphy. Gold is used for decoration, and to emphasize the divisions of the text in the various 9th to 10th century Kufi leaves. While the use of colour and style of decoration in the later Maghribi leaves (no. 18 & 19) are more elaborate and complex, with, for example, the use of two types of colourant on the sheet, one for paint and the other for ink. The red floral motiff on the Hijazi leaf 17, is a later addition, as are the illuminated verse markers on the later Kufi leaf 10.21 In general the pigment layer on all the Qur’anic fragments in this study is abraded and thin due to the condition and age of the leaves.

In contrast to the written sources, the palette of the calligraphers and artists that decorated the I.C. parchment leaves is very limited. The leaves have the basic colour palette as laid out by Ibn Badis including white; black, red, green, yellow and blue pigments are present. The list he gives of raw materials to make the colours is: sot, black; cinnabar, red lead or realgar, verdigris, orpiment and indigo or lapis lazuli.2 Some of the raw materials reflect the type of mining that was prevalent in the Near East, particularly in the region between the Persian Gulf and the Mediterranean. In the early Islamic period mercury ore (source of vermilion and cinnabar) was being used for the extraction of gold from its ore especially in Arabia, and also for the extraction of copper and silver.27 Orpiment deposits are found in mines where there are gold, lead and silver veins.25

Identification

Sophie Lewincamp prepared the micro-samples of the pigments from the Qur’An leaves. The stereo-microscope was used to locate suitable areas for sampling and to remove samples from the substrate with a scalpel. Due to the thinness of the pigment layer, parchment fibres were present in many of the pigment samples. Each sample was mounted for SEM-X-ray microanalysis on an aluminum pin mount with double-sided carbon tape. Photomicrographs were taken of the sample area before and after sampling at 40x and 66x magnification.

Frank Hengemihl analysed the samples using a Cambridge S200 scanning electron microscope (SEM) in combination with energy-dispersive x-ray microanalysis (EDS). The SEM was used to locate the sample and magnify it to 1000x, and Kevex Sigma 10 version 4.14 system software was used for microanalysis. The Kevex Quasar subprogram was used to analyse the x-ray spectra. Spectra collection conditions were a 100 second collection period with a deadtime of 35-40%. Spectra were compared to the reference collection of spectra of known pigments maintained by the Library of Congress’ Preservation Research and Testing Division. (Table 2)

Green Blue Pigments

Of the three blues on the parchment sheets, two from the later Maghribi leaves (no. 18 & 19) were painted with azurite, a basic copper (II) carbonate mineral with the formula Cu[Cu(CO3)](OH).28 (Table 2). Though azurite was used in the pre-Islamic Near East, Ibn Badis does not mention blue from azurite (or a green from malachite, the other basic copper (II) carbonate) in his treatise but rather indigo and lapis lazuli-based blues. Deroche identified the presence of azurite in several 14th century manuscripts from the Maghreb and Spain.29 Though azurite and malachite ores are found in the same mineral deposits and often aggregates of the minerals are found together, the green dots on the Maghribi leaves are not malachite.

The blue on Leaf no. 7 is ultramarine or lazurite from lapis lazuli. It is a sulphur-containing sodium aluminium silicate where the proportions of aluminium, silica, and oxygen are fixed while those of sodium and sulphur can vary.22 (Table 2) Lapis lazuli has been and continues to be mined in the Badakhshan region of Afghanistan as a semi-precious stone and for natural ultramarine.30 Recent finds at Sar-e-sole mine have yielded lazurite crystals of very high purity.31 Prior to the synthesis of synthetic ultramarine in the late 19th century Badakhshan was the sole source for the deep blue for luxury, illuminated medieval manuscripts.32

Figure 1 Leaf no. 19th, Maghribi script, 1250-1300 C.E.

Figure 2 Leaf no. 154.28b, Kufi (New Style III), 10th century. The chapter heading is written in gold ink and end with an elaborate parchment design.

Figure 3 Leaf no. 17th, Hijazi I Script, circa 8th century. Originally undecorated, the floral motif and interlinear inscriptions were added later.

Figure 4 Leaf no. 7, Kufi D I script, circa 9th century. Diacritical dots are painted in red, yellow, green and blue. The tenth verse marker is a painted gold medallion.

Figure 5 Spectra for azurite from the blue dots on Leaf no. 18.

PRELIMINARY CHARACTERIZATION OF THE LEAVES WAS UNDERTAKEN USING THREE METHODS:

1. SEM-EDS elemental analysis to help identify pigments.
2. Multiphoton imaging to observe the inks and the substrate.
3. Visual observation to characterize the parchment.
### Table 2: SEM-EDS pigment spectra and tentative identification based on spectra

<table>
<thead>
<tr>
<th>Leaf No.</th>
<th>Blue Spectra</th>
<th>Green Spectra</th>
<th>Red Spectra</th>
<th>Gold Spectra</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>N/A</td>
<td>O, S, AI, Cu, &amp; K</td>
<td>A copper green</td>
<td>Cinnaabar</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Hg, S, Ni, Cu, &amp; Ca</td>
<td></td>
<td>Gold: Au with trace amounts of Si, S, Ca &amp; Cu</td>
</tr>
<tr>
<td>7</td>
<td>Ni, Ca, O, S, AL, Na, trace amounts of Cu</td>
<td>Lazurite/Ultramarine</td>
<td>Hg &amp; S with trace amounts of Fe, Ca, O, K &amp; Cu</td>
<td>Cinnaabar</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Cu, O, Cl, Al, Si, K, &amp; Cu</td>
<td></td>
<td>Gold: Au with trace amounts of Si, As, Ca &amp; Fe</td>
</tr>
<tr>
<td>9</td>
<td>N/A</td>
<td>O, S, Ca &amp; Cu</td>
<td>A copper green</td>
<td>Iron oxide earth</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Red 1: Hg &amp; S</td>
<td></td>
<td>Gold: Au with trace amounts of Si, Fe &amp; Cu</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Red 2: O, S, Ca &amp; Fe</td>
<td>Note: Both show trace amounts of Na, Cu, Mg, and K</td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>N/A</td>
<td>Cu, O, Cl, K &amp; Cu</td>
<td>A copper green</td>
<td>Cinnaabar</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Hg &amp; S with trace amounts of Si, Ca, Al, Cu, Fe, Mg &amp; K</td>
<td></td>
<td>Gold: Au with trace amounts of Si, S &amp; Cu</td>
</tr>
<tr>
<td>15</td>
<td>N/A</td>
<td>Cu, Si, O, S &amp; K</td>
<td>A copper green</td>
<td>Cinnaabar</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Strong Hg &amp; S peaks with trace amounts of Si &amp; Fe</td>
<td></td>
<td>Gold: Au with trace amounts of Cu, Si &amp; Ca</td>
</tr>
<tr>
<td>16</td>
<td>N/A</td>
<td>N/A</td>
<td>Strong Hg &amp; S peaks with trace amounts of Si, Na, &amp; Al</td>
<td>Cinnaabar</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Gold: Au &amp; S with trace amounts of Si, Na, &amp; Cu</td>
</tr>
<tr>
<td>17</td>
<td>N/A</td>
<td>N/A</td>
<td>Hg, S, Cu, &amp; O with trace amounts of Na, Al, K &amp; Fe</td>
<td>Cinnaabar</td>
</tr>
<tr>
<td>18</td>
<td>Strong O &amp; Cu peaks with trace amounts of W, Al &amp; Ca</td>
<td>Azurite</td>
<td>O, Cu, Cu &amp; Na with trace amounts of Cl, Si, K, Al &amp; S</td>
<td>Cinnaabar</td>
</tr>
<tr>
<td></td>
<td></td>
<td>A copper green</td>
<td>O, Cu, Si &amp; Fe, with trace amounts of Na, Al, Cu, S, Cl &amp; Fe</td>
<td>Iron oxide earth</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Gold: Au with trace amounts of Si, S &amp; Al</td>
</tr>
<tr>
<td>19 left &amp; right</td>
<td>Strong O &amp; Cu with trace amounts of Ca, Si &amp; Na</td>
<td>Azurite</td>
<td>O, Cu, Cu &amp; Na with trace amounts of Cl, Si, K, &amp; Si</td>
<td>Cinnaabar</td>
</tr>
<tr>
<td></td>
<td></td>
<td>A copper green</td>
<td>O, Cu, Si &amp; Fe, with trace amounts of Na, Al, Cu, S, Cl &amp; Fe</td>
<td>Iron oxide earth</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Gold: Au with trace amounts of S, Si, Na, Mg, Ca</td>
</tr>
<tr>
<td>20</td>
<td>N/A</td>
<td>N/A</td>
<td>Red 1: Hg &amp; S</td>
<td>Cinnaabar</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Red 2: prop peaks for Hg, S, Fe &amp; O</td>
<td>Iron oxide earth</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Note: Both show trace amounts of Cu, Cl, Mg, &amp; K</td>
<td></td>
<td>Gold: Au with trace amounts of Cu, O, S, Ni, Cl, Mg &amp; Fe</td>
</tr>
<tr>
<td>154:28b</td>
<td>N/A</td>
<td>Strong Cu, followed by S &amp; Cl</td>
<td>A copper green</td>
<td>Cinnaabar</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Trace amounts of Ca, Si, O, K &amp; Mg</td>
<td>Strong Hg &amp; S followed by Si, O &amp; Cu</td>
<td>Iron oxide earth</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Note: Both show trace amounts of S, Si, Al &amp; Fe</td>
<td>Gold 1: Au</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Gold 2: Au with small Cu peak</td>
</tr>
</tbody>
</table>

**Figure 6** Spectra for lazurite from a blue dot on Leaf no. 7.

All of the green pigments sampled in this project displayed copper in their spectra but as there are many copper-based green pigments positive identifications with SEM was not possible. Initially the conservators assumed that where verdigris-type damage was visible then the pigment would most probably be verdigris or copper acider. Copper catalyses an oxidation reaction with the ornament and verdigris can burn through a substrate and or darken in colour. Where darkening of the green colour was observed on green cloisonnés, it was concluded that the pigments were probably verdigris. The same conclusion was drawn for the holes left by green painted dots on the Maghribi leaves (no. 18 & 19) by where there was an outline of green, gelatinized parchment.

**Figure 7** Spectra for copper green from the edges of a green dot on Leaf 18.

Subsequent x-ray diffraction analysis (XRD) did not show the presence of Cuin the former case and gave a very weak signal for the second sample. The pigment could not be identified. However, SEM-EDS at the Library showed a weak copper and chlorine signal. The green on these leaves had fallen out leaving a charred greenish ring from where the sample was taken. In his studies on green pigments in illuminated manuscripts, Rankin found that the more damaged and deteriorated the copper-based pigment was, the less copper was to be found in the actual colour sample. XRD of these samples showed the presence of calcite and quartz probably from the preparation of the parchment substrate.

### Red pigments

Two different red pigments were identified during this study: a variety of iron oxide containing earth pigments, and mercuric sulphide or cinnaabar (Table 2). The iron oxide pigments showed clear iron spectral peaks along with strong peaks for Ca, Si, and O. The proportions of the elements in X-rays of the spectra of the 10 red on the Maghribi leaves 18 and 19 are very similar and are probably the same pigment. Despite proximity to the cinnaabar mines in Spain, the only red pigment on these two leaves is an iron-oxide rich earth pigment.
Mercuric sulphide has a long history of use in Asia: the ore cinnaubar was mined in prehistoric times, and the Chinese have known how to make dry-process vermilion from mercury and sulphur since the 2nd century AD. A 10th-century Arabic text on pharmacology, Tuhfat al-abab, addresses the different methods of making vermilion and cinnaubar, respectively. Rich deposits of cinnaubar were to be found historically at the Almaden mines in Spain, in south western China and as mentioned before in Arabia. A later Chinese text from the 13th century, the Chao Hai huai, mentions cinnaubar as one of a list of the items imported from the Persian Gulf Island of Kish, a trading centre at the time.

Gold Pigments

The gold pigments samples are all very pure to fairly pure gold with small and varying amounts of trace elements such as iron, copper, and aluminium. Silica, calcium, sodium and magnesium are also visible but this is mostly probably due to contamination of the sample by the materials used to prepare the parchment (Table 2). The gold on leaf 10 was of poor quality compared to the other samples.

On leaves where two gold pigments are used, copper is present in the spectra for one sample. The addition of copper provides a warm tonal quality to the gold and extends the palette of the artist. In leaf no. 10, the cooler hue pure gold has been used to frame the calligraphy, while the verse markers are rendered with the copper-containing gold. In leaf no. 154:28 the gold ink used to calligraphy and illuminate the surah heading contains some copper while the floral verse markers are painted with the purest gold of all the samples in this study.

Yellow Pigment

Only parchment leaf no. 7 had a yellow pigment, a diactrical marl used dots painted with red, blue, green and pale yellow pigments, respectively. Sample spectra show silicon, arsenic, sulphur, calcium, and iron. However, they do not show the characteristic strong peaks for arsenic or sulphur associated with pure ore pigments: the only yellow inorganic pigment mentioned by the Radda.

Comparative spectra were taken for 'Gil Mash' a yellow clay pigment specimen acquired in Iran in 1935 and used in Persian miniatures painting at the time. It showed large iron peaks with traces of arsenic, sulphur, silicon, calcium, but no oxygen. The pigment on leaf 7 shows the same elements as Gil Mash but with more arsenic present and very little iron by comparison. It is probable that this pigment is either a naturally occurring ore with many impurities, or orpiment produced with the addition of a mineral extender. Further analysis with XRD could provide a more nuanced identification of the constituents of this pigment. The possibilities of the pigment being an ore is highly likely because of the many known orpiment and realgar mines throughout the Middle East in Iran, Iraq, the Caucasus, Turkestan and Anatolia.

The inks and black pigments

Ink

A few of the inks from leaves were sampled for SEM-EDS and all of them showed the presence of iron. Unfortunately, X-ray microanalysis could not identify the presence of carbon in the samples due to interference from the carbon SEM mounting tape.

The parchment leaves were viewed under the Art Innovations Artist camera for multi-spectral reflectography. In the infrared II or IIR2 mode (excitation band width of 90 to 120 nanometers) iron-gall inks typically do not absorb energy and appear transparent. Carbon black inks absorb strongly in the IIR2 range and appear black. However, in false colour infrared II or ICR2 mode the image is manipulated so that the faded iron gall ink appears red. Carbon black inks appear brown in ICR2. The main writing inks in all the parchment leaves were red in the ICR2 and appear to be iron-gall inks. However, in IIR2 mode only the inks on the later Maghribi leaves (no. 18 & 19 1/4) and leaves no. 20 and 154:28 were nearly transparent. The ink on all the other leaves of the sample group showed various degrees of transparency relative to each other.

To corroborate the multimodal imaging results, the inks were tested for Iron (II) ion, the stable iron ion found in the iron gall ink complex, as well as the unstable Iron (II) ion. The testing method using an indicator paper was developed by Netherlands Institute for Cultural Heritage. All the inks tested positive for Iron (II) ion with a few of them showing a minimal presence of the Iron (II) ion. The presence of the Iron (II) ions in an ink may be the result of excess iron in the ink formulation as well as the degradation of the ink complex over time. Typically iron-gall ink on parchment is more stable than on paper due to the alkalinity of the substrate. In addition the excess calcium in the parchment reacts with the acids in the ink, such as sulphuric acid to create calcium sulphate, a non-corrosive salt. The ink formulations appear to be well balanced: the iron (II) ions are bound into the iron-gall ink complex and there are fewer free Iron (II) ions present. However, heavily inked lines, especially horizontal lines, show some cracking associated with iron-gall ink degradation.

If all the inks are iron-gall inks why do they show different levels of transparency in the IIR2 range? Under normal inspection, many of the inks are muddy and appear to contain additives to bulk up the ink and promote opacity. The inks do not appear to contain carbon black or soot but possible organic material, because the ICR2 image does not show the strong absorbance characteristic of carbon black.
The calligrapher, Mohammed Zakariya, when viewing the leaves illustrated how the original calligrapher would have reworked each letter. Words in the more decorative and heavy Kufi script were reworked as soon as they were written to strengthen horizontals, apply serifs and thicken key areas of script. In the IR2 mode these areas appear darker. Test switches of a variety of layered iron gall ink viewed in the IR2 mode of the multispectral camera showed the same phenomena. In contrast, the Qur'an leaves written in the most free-hand script styles, namely Maghribi and New Style Kufi III are close to transparent, closely followed by the 8th century leaf written in Hijazi I.

Carbon-based pigments and inks
Gold-painted verse markers in Maghribi Qur'an leaves no. 18 and 19 lv have been outlined with an opaque black pigment. The presence of carbon could not be noted in the SEM/EDS spectra due to the SEM carbon mounting tape. The spectra show small amounts of iron and larger peaks for calcium and oxygen. Phosphorus is not present in the spectra and therefore bone material has been ruled out. However, there is a significant amount of carbon in the pigment as it absorbs in the IR2 range during multispectral imaging and appears dark brown in the FCR2. Until further study can be undertaken it is probable that the black pigments are some form of carbon black from lamp black or soot.

Other carbon pigments were recorded by IR2 reflectography. A word was reworked in carbon-based ink on leaf 11 while later annotations on leaves nos. 17 and 20 are also in similar ink. They all appear brown in FCR2. It is interesting to note that the black outlines on the illuminated verse markers all appear to use the same ink that the Qur'anic verses are written in, except for the Maghribi leaves no. 18 and 19 lv discussed above.

Parchment
To complete the initial characterization of the Qur'an leaves, a visual description of the substrate was undertaken using the microscope and the ultra-violet fluorescence mode of the Artie multispectral camera. Regardless of the age of the parchment, we hoped to find clues to manufacturing methods and animal species. Additional information was made available through SEM/EDS and XRD of parchment fibres.

Parchment is a writing material made from animal skin that has been soaked in lime, dehaired, scraped, and dried under tension. After further thinning, various surface treatments are used on parchment to prepare it for writing with ink. Nowadays, abrasives such as fine pumice, which is a volcanic glass comprised of varying silica (SiO2) depending on the source, are rubbed over the parchment by the calligrapher to create a uniform surface for the application of ink. Spectra for the Qur'an parchment leaves show silica quite consistently. XRD patterns for psyllium/silk, a magnesium aluminium phyllosilicate similar to the mildly abrasive Fuller's Earth, were detected on the Maghribi leaves.

Additional XRD patterns were identified for gypsum (CaSO4) and calcite (CaCO3) in leaves nos. 2, 9, 18 and 19 lv. Only gypsum was detected on leaves nos. 9 and 10. Both gypsum and calcite can form in parchment with prolonged exposure to air pollutants from burning fossil fuels, in particular atmospheric sulphur and nitrogen oxides.

Though it is often difficult to tell the species of animal that has been used to create a parchment, we assume that most of the medium-to-small parchments were produced from smaller domestic animals. According to Hesse and Wapnish, sheep and goats were found all over the Middle East and were raised by communities wishing to be self-sufficient, as well as for sale. They were a source of food, wool and leather; and were used as beasts of burden and for their motive power. Camels and deer skins were rarely used for parchment.

Parchment does not indicate the type of animal used in its making. The finest skins come from young or even foetal animals. The hair colour of the animal can affect the colour of the parchment itself in some species. The whitest parchments tend to come from animals with light colour hair whereas the animals with black, brown or variegated hair produce darker parchments. In the case of goats, haircolour does not reflect the pigmentation on the skin of the animal while in cowhides the pigmentation follows the colour of the animal's hair. The flesh side of a finished parchment is generally whiter and softer than the hair side. The hair side, however, is smoother and often displays scutelled traces of the hair follicles.

Hair and flesh sides were noted for each leaf. The hair side of parchment has a more compact structure and therefore the parchment will naturally curl or contract towards the hair side. Microscopic examination was used to locate hair follicles to identify both the hair side and the animal species. Finally, the ultra-violet fluorescence mode on the Artie camera was used to corroborate our findings from the other two methods. The granity nature of the prepared flesh side absorbs more and appears darker than the smooth hair side.

Three types of sheep were tentatively identified in the parchments of seven leaves. Leaves no. 1 and 17 were identified as a sheepskin parchment due to the presence of axilla formation. Axilla pattern on the right hand verso of leaf no. 17 (Hijazi I) led to a positive identification for sheep parchment. The axilla, which corresponds to the skin over the shoulder towards the forelegs of the sheep and is of variable thickness, has caused some of the creases in this parchment leaf. Leaf no. 20 shows brown circular lines along the right hand side that would correspond to lines that circle around the neck of the sheep. They are visible both on the recto and verso of the parchment despite the excessive thinning of the area by the parchment maker. The observation of clear primary and secondary hair fibre follicles in leaf no. 9 led to a positive identification for sheep hair. The few hair follicles visible in leaf no. 18 do not appear compact in structure and are similar to sheep follicle patterns. Leaves no. 19 lv has similar diameter in the primary and secondary hair follicles similar to primitive medium woolen sheep. Leaf no. 10 was very well prepared and had no hair follicles visible, however, the glazed surface appearance of the leaf has led to a tentative identification as sheepskin parchment. The high lanolin content of sheepskin can lead to a glazed appearance of the skin.

In general the parchment used in the leaves was not highly processed. The surfaces are not carefully prepared and uniform. The parchment is not always well-depilated. Leaves are cut from the extremities of the parchment (see above) as the caligraphers or bookbinders use as much of the skin as possible. Creases, blisters and cuts are retained. Leaf no. 15 lvv has not been prepared well by the parchment maker as a piece of membrane is still attached to the parchment. Leaf no. 15 has a marked scar on the bottom left hand side of the recto which could be a parchment maker's mark as unlike a natural blemish it does not manifest itself on the verso of the leaf.

Conclusion
Each phase of this study leads to further questions about the materials used to create the Qur'an leaves. At the conclusion of the first phase of the investigation, which was based on observations that could be made with the equipment the Library had available for use at the time, certain preconceptions of the conservators and curators were shaken. The parchment leaves showed a fairly consistent material and limited palette. All the leaves were written with iron gall inks. XRD of a limited sampling of the copper greens identified basic copper chlorides as the pigment of choice.

The study of the fragments will continue as there is no single method that can definitively identify all the constituents of any of the materials used in their production. Further areas of research will include completing the XRD analysis of the copper greens on the remaining leaves and polarizing light microscopy of the orpiment and lazurite for definitive
identification. A more in depth study of the inks shall be undertaken to identify the various constituents of the ink matrix using infrared fluorescence spectroscopy.

Notes


3. According to calligrapher Mohamed Zakeria, the spacing of the letters on the manuscript page does not accommodate the larger gold verse marker but rather a small dot similar to those visible in the rest of the leaf. (Private communication with Yasmeen Khan August 10th, 2009)


6. IBN BADIS, 29.


14. PORTER, 82.


21. SCOTT, 292.

22. SCOTT, 136.

23. SCOTT, 292.

24. BANIK, 95.

25. DEROCHÉ, F (1966). He found haematite on an 8th century Qur’an from Egypt or the Middle East, and two from 14th century Maghreb and Spain.

26. IBN BADIS, 25. Ibn Badis gives a recipe for making red ink from a piece of cinnabar.


28. WEST FITZGIBBON, 60.


33. IBN BADIS, 29.

34. Sample donated by the Freer and Arthur M. Sackler Galleries. Brought in Isfahan in 1893 by Katherine and Myron Beem Smith.

35. Fitzhugh states that the XRDs of a yellow pigment collected by Myron Smith (zarnikh-i zard) showed portlandite Ca(OH)$_2$ mixed with orpiment and that in nature they can be found in close proximity. WEST FITZGIBBON, E (1997). Orpiment and red/green Artists’ pigments: A Handbook of their history and characteristics, vol. 3. New York: National Gallery of America. p 63.

36. FITZGIBBON, E. Orpiment, p 54.


42. Under any ultra-violet lighting conditions gross planar inconsistencies in both paper and parchment are visible. It is an easy method of distinguishing grain and flesh sides of old parchment.


45. MORONY, xviii.

46. Parchment-making workshop discussion with Jesse Mayer, parchment maker and owner of Pergamena Handmade Parchment. (April 2002).

47. Parchment observations were done with the help and guidance of Jesse Munin, Senior Books Conservator at the Library of Congress.


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Authors’ Biography

Yasmeen R. Khan is a Senior Rare Book Conservator at the Library of Congress where she has since been 1996. She has an M.L.I.S with an Advanced Certificate in Conservation from the University of Texas at Austin and a R.A. In Middle Eastern Studies from Columbia University. Her areas of interest in conservation include the study of bookmaking in the Middle East and the development of conservation techniques to preserve manuscripts and miniature paintings.

Sophie Lewinckamp has been a Paper Conservator at Australian War Memorial since 2006. She received a Bachelor of Applied Science in Conservation of Cultural Materials from the University of Canberra. While undertaking this research she was the Harper-Inglish Conservation Fellow at the Library of Congress. Her areas of interest in conservation include parchment in particular of Middle Eastern origin and in disaster scenarios.