University of Évora

ARCHMAT

(Erasmus Mundus Master in Archaeological Materials Science)

Mestrado em Arqueologia e ambiente (Erasmus Mundus – Archmat)

The Study of Biographical Trajectory of Portuguese 12th Century Illuminated Manuscript: LECCIONARIUM ALC. 433 from Alcobaça Collection held by The Biblioteca Nacional de Portugal

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This thesis has been defended on 7 November 2020, in University of Évora, Portugal with the juries:

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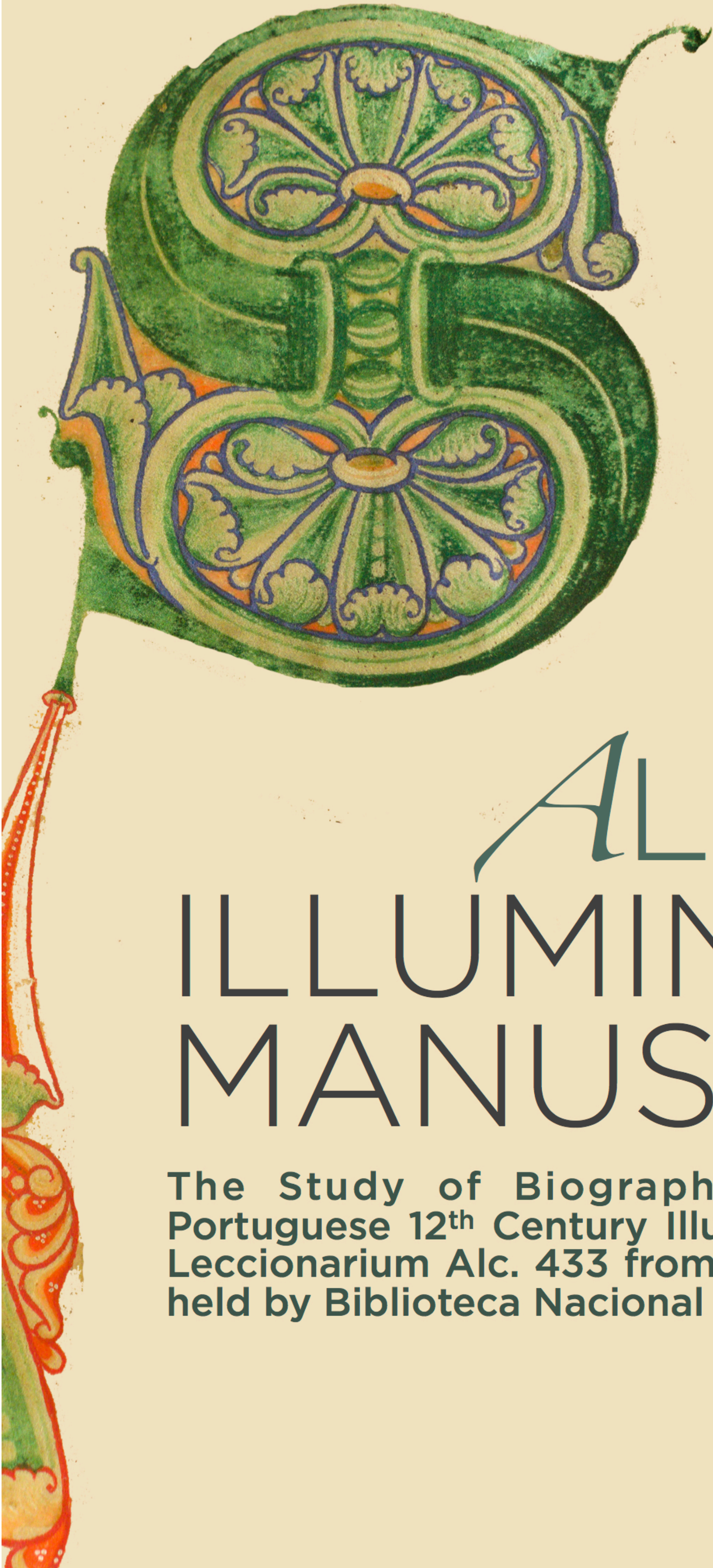

"This research is supervised by Catarina Miguel, Ph.D; with Catarina Barreira, Ph.D, and Conceição Casanova, Ph.D."

Master's degree obtained through this research is funded under the Erasmus Mundus Programme: Archaeological Materials Science

ALC. 433 ILLUMINATED MANUSCRIPT

The Study of Biographical Trajectory of Portuguese 12th Century Illuminated Manuscript: Leccionarium Alc. 433 from Alcobaça Collection held by Biblioteca Nacional Portugal

SHATILA FITRI

ACKNOWLEDGMENTS

First of all, I am wholeheartedly grateful to Education, Audiovisual, and Culture Executive Agency (EACEA) of the European Commission, as well as with the Erasmus Mundus Consortium for my selection and funding to participate in this master programme. At the same time, I am thankful for the support of HERCULES Laboratory (Herança Cultural, Estudos e Salvaguarda and to the University of Évora for the availability of the equipment needed for the execution of this research.

Special thanks to the Biblioteca Nacional de Portugal for the availability and permission access to its valuable illuminated manuscripts collection for this research.

My deepest gratitude is expressed to Prof. Catarina Miguel, Prof. Catarina Fernandes Barreira and Prof. Conceição Casanova for their guidance and supervision during the research and also to the research project team of Cistercian Horizons (PTDC/ART-HIS/29522/2017), also Silvia Bottura who willingly guided me and inspired me through her passion on science, art, and culture.

I thank Prof. Nicola Schiavon as the coordinator of the consortium of ARCHMAT, coordinator in Aristotle University of Thessaloniki: Prof. Evangelia Varella, coordinator in University of Rome Sapienza, Prof. Donatella Magri, and all of the professors in three universities who taught and shared many knowledges and experiences during the my studies in Europe.

My sincere gratitude for my parents who let me to get a higher education 12.672 km away from home, and my brother Ahmad Ahsani Taqwim who always be there whenever I need him within seven hours different of time. Also, for my best friend Fakhriyya Huseynli with whom I travelled and studied in Europe, and my flatmate, Giulia Lombardo with whom I shared pandemic lockdown for 5 months who constantly supported each other- this thesis would be impossible without you.

Above all, thanks to Allah سُبْحَانَهُ وَتَعَالَى who never stop blesses me with many things in my life, and Prophet Muhammad صَلَّى اللهُ عَلَيْهِ وَسَلَّمَ who always encourage me to seek knowledge, as the first verse of the Holy Quran, *"Read, Read in the name of Your God."*

ABSTRACT

The Study of Biographical Trajectory of Portuguese 12th Century Illuminated Manuscript: LECCIONARIUM ALC. 433 from Alcobaça Collection held by The Biblioteca Nacional de Portugal

This thesis presents an interdisciplinary approach to a 12th century illuminated manuscript, a *Leccionarium* (Alc. 433), produced in Alcobaça Monastery which currently is being preserved at Biblioteca Nacional de Portugal in Lisbon.

The aim of the work was to trace the biography of this illuminated manuscript, through the liturgical studies and to obtain the chronological timeline of the use of materials in Alcobaça *scriptorium*, through centuries. The representative *folia* of Alc. 433 were characterised with h-XRF, UV-Vis-NIR-FORS, and hyperspectral images.

The result indicates that Alc. 433 was produced in the last quarter of the 12th century, followed by addition of some *folia* in 13th, 14th, and the beginning of 17th century. Materials identification revealed the use of different pigments in different periods: vermilion and minium (red), copper proteinate (bottle green), yellow lake pigments, azurite and lapis lazuli (blue). The PCA study of yellow lake dye reproduction indicates the use of turmeric yellow lake pigment in the initial core. Furthermore, the analysis of iron gall ink also shows that the initial core of Alc. 433 contains the similar ratios of elements with Alc. 11 (primitive manuscript of Alcobaça) thus proved the Alc. 433 was also the produced in the earliest period of the active year of Alcobaça *scriptorium*.

Keyword: Portuguese Medieaval Manuscript; FORS; h-XRF; hyperspectral images; yellow lake dye

RESUMO

O Estudo da Trajetória Biográfica do Manuscrito Português Iluminado do Século XII: Leccionarium Alc. 433 da Coleção de Alcobaça Detida pela Biblioteca Nacional de Portugal

Esta tese apresenta uma abordagem interdisciplinar de um manuscrito iluminado do século XII, um Leccionarium (Alc. 433), produzido no Mosteiro de Alcobaça e que se encontra preservado na Biblioteca Nacional de Portugal em Lisboa.

O objetivo deste trabalho foi traçar a biografia deste manuscrito iluminado, através do seu estudo litúrgico e obter a linha cronológica da utilização de materiais no scriptorium de Alcobaça, ao longo dos séculos. Os fólios mais representativos do Alc. 433 foram caracterizados h-XRF, UV-Vis-NIR-FORS e imagens hiperespectrais.

O resultado indica que o Alc. 433 foi produzido no último quarto do século XII, e que foi enriquecido com a adição de fólios e/ou cadernos nos séculos XIII, XIV e inícios do século XVII. A identificação dos materiais revelou o uso de diferentes pigmentos em diferentes períodos: vermelhão e minium (vermelho), proteinato de cobre (verde garrafa), pigmento lago amarelo, azurite e lápis-lazúli (azul). O estudo PCA da reprodução do corante lago amarelo indica o uso de açafrão no núcleo inicial do pigmento. Além disso, a análise das tintas de escrita evidenciou uma analogia de composição da tinta ferrogálica utilizada no núcleo inicial do manuscrito Alc.433 e da tinta ferrogálica utilizada no texto do Alc.11 (outro manuscrito produzido nos primeiros anos do scriptorium de Alcobaça) o que comprovou assim que também o Alc. 433 foi produzido na mesma época, isto é, em torno de 1175.

Palavras-chave: Manuscrito Medieaval Português; FORS; h-XRF; imagens hiperespectrais; corante lago amarelo

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LIST OF ABBREVIATION

BNP	Biblioteca Nacional de Portugal
FORS	Fibre optic reflectance spectra
h-XRF	Handheld X-ray fluorescence spectroscopy
HIS	Hyperspectral Images Spectroscopy
UV-Vis-NIR	Ultraviolet-visible-near infrared

IN TEXT REFERENCES OF ILLUMINATED MANUSCRIPT COLLECTIONS

In-text Reference	Title of the Manuscript	Location	Online Access
Alc. 433	*Liturgia e ritual. Leccionátio.*	Biblioteca Nacional Portugal	http://purl.pt/25154
Alc. 11	*Saltério Segundo o rito cisterciense*	Biblioteca Nacional Portugal	http://purl.pt/24657/3/#/1
Alc.360	Homiliae in Leviticum, Numerum, Josue, et Judices	Biblioteca Nacional Portugal	http://purl.pt/24810

NOTES TO IMAGES

All images from Alc. 433, and Alc. 360 Anon are © Biblioteca Nacional Portugal

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All handheld digital microscopic images, HIS images, technical photography of Alc. 433 are © Laboratório HERCULES and Biblioteca Nacional Portugal

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LIST OF TABLES

This thesis is parted into four sections according to its purpose. The beginning of each part is decorated by Batik (UNESCO 4.COM 13.44), medieval Indonesian wax-dye textile illumination technique, personally designed by the author of this dissertation. All motives are inspired from the pattern used in Alc. 433.

Part I

THE QUEST

Historical Biography of Alc. 433

Chapter I: Introduction

1.1. Devising New Approach

"Quos libros non liceat habere diverse. X. Missale, epistolare, textus, collectaneum, graduale, antiphonarium, regulates, psalterium, lectionarium, kalendarium, ubique uniformiter habeantur " (Canivez 1933: vol. 1, 13 and Waddell 2002: 513)[1]

As a direct source of history, a manuscript can inform us about past civilisation explicitly from the content of the text, illuminated decoration and production techniques. However, cultural heritage research on characterising manuscripts has been developed rapidly due to advance methods on scientific investigation using in-situ and non-invasive techniques. This is very essential to the manuscripts due to its preservation. In the past two decades, hundreds of medieval books have been researched scientifically. The collections of medieval illuminated manuscripts from several *scriptoria* in Europe were investigated and proceed towards new studies of its materials, such as pigment, binder, parchment/paper and writing ink alongside the bookbinding technique. Furthermore, materials investigation usually complements the historical analysis because several causes, such as culture, economy and politics, contributed to the *scriptoria* and the production of the manuscript. In Portugal itself, numerous notable *scriptoria* spread along the country which were active under the influence of religious Orders, such as Cistercian Order. One of them is Alcobaça *scriptorium*- one of most productive Portuguese Cistercian *scriptoria*. The Alcobaça Monastery produced approximately around 467 manuscripts (Barreira, 2017) that embodies normative texts, Bibles, a great variety of liturgical books (for Mass and Divine Office), books pertaining Liturgy, Rules of Saint Benedict, texts from the Church fathers, among other spiritual readings essential to the monastery (Barreira et al, 2019). It must be highlighted that the liturgical manuscripts present important information about liturgical practices, celebrations, and festivities which lead the essential development of the worship practices and how it flourished. Moreover, liturgical studies present a unique and valuable information for complementing scientific analysis on illuminated manuscripts. Despite Cistercian liturgy evolved dynamically through centuries, the Cistercian kept the tradition of uniformity, by trying to keep its main formula and prayers as an effort to preserve the Liturgy as Saint Bernard imagined. When a Cistercian monastery found a new monastery or mother house, then the exact same liturgy from the monastery would be copied as a template or model context for the new affiliated monastery, although there were some liturgical variations, depending on the local context of each monastery.

The annual meeting of Cistercian monasteries – the General Chapter, held in Cîteaux- established the *Statuta* as the official written testimonies, containing information about various aspects of Cistercian life and monasteries, namely concerning liturgy (new feasts introduced in the Cistercian Calendar and *Sanctoral*). Depending on the typology of the liturgical book and given the length of the texts, some liturgical books are organised in three

[1] This statute *(Statuta)* informs us that each Cistercian monastery should have a uniform set of liturgical books in order to function, this is, to practice the Cistercian liturgy in a uniform manner.

or more volumes. By following a multidisciplinary approach based on both liturgical studies and materials characterisation, a new approach to the study of the chronological bibliography of a liturgical book was accessed.

Between hundreds of illuminated manuscripts produced in Alcobaça *scriptorium,* Alc. 433 leaves obvious signs through its *folia*. It is evident that some *folia* were the initial core, while some others were added centuries later. This envisioned the possibility of further and advance research connecting this manuscript through its trajectory from one period to another, with the history of Alcobaça. Thus, liturgical analysis and materials characterisation were chosen to acquire this purpose.

The materials used to produce Alc. 433 were analysed in order to investigate the period of production and changes over time. The primary structure of the bookbinding, that is sewing and supported materials if not changed overtime will contribute to set the date of the manuscript production as whole. As stated in the first paragraph of this chapter, this research is also presented within multidisciplinary studies. The important approach to trace the biography of the respective Alc.433 revealed the complex chronology of the manuscript production in Alcobaça in the matter of materials, liturgical context, and bookbinding. Finally, the studies will broaden the research approach within Portuguese illuminated manuscript of Alcobaça's *scriptorium*.

1.2. Illuminated Manuscript Time Tracing

Despite the fragility of the materials, illuminated manuscripts are one of most valuable artefacts that arrived to our days and are substantial to be studied. The number and variety of illuminated manuscripts that survived to our days are a reflect of its valuable meaning in medieval and renaissance time that guaranteeing their preservations during centuries. Also because of the previous reason, illuminated manuscripts are not like other artefacts. They were rarely exposed to the external elements, social upheaval, and unsympathetic restoration- unlike paintings, frescoes, or stained glass, that makes its materials are mostly in superior conditions (Panayotova & Ricciardi, 2016). Thus, the illuminated manuscripts studies and researches are always enticing yet challenging. Many illuminated manuscript studies in cultural heritage approach objectives such as provenance, uses and circulation, and dating. The studies of illuminated manuscripts are always multidisciplinary studies since it concerns codicological, textual, palaeographical, and art historical examinations of structures, contents, scripts, iconography, and style along with the uses within specific communities and individuals involved within the manuscripts (Panayotova & Ricciardi, 2016).

Dating the illuminated manuscript sometimes can be a complement purpose of studies, due to its explicit context of writing that we can obtain the information of the time of its production directly. For example, the illuminated manuscript The History of Outremer[2], written by William of Tyre which explicitly written in the Crusades time in 12th century (Folda, 1973). However, this research expands beyond distinguishing the time of manuscript production. It

[2] *The History of Outremer* is the famous illuminated manuscript about the history of Crusades. William of Tyre was an archbishop in the Kingdom of Jerusalem (Edbury et al, 1991). He wrote several illuminated manuscripts that explicitly telling the story of The Crusades, such as *Historia Ierosolimitana.*

is a complex yet informative new approach to study step by step about the manuscript: the core model, the addition, the materials, the liturgical context, to establish the chronological idea of this intricate manuscript. It is a beguiling topic with several questions regarding the purpose of the research, since as far as the author searched, the study of biography of a Portuguese illuminated manuscript produced in Alcobaça *scriptorium* had not yet been done. Several research about investigating the transformation of styles and materials may have been conducted, for instance liturgical manuscript which was specifically dated to be produced in one century (Barreira et al, 2016)[3] or the study of the origin, uses and circulation of liturgical codices (Bragança, 1984; Rêpas and Barreira, 2016; Barreira 2017; Barreira 2017), colour tracing on illuminated manuscript which linked to the spread of the several Cistercian monasteries (Castro et al, 2014)[4], or palaeographical studies on historical context (Ferreira, 2013)[5]. By referencing these researches and their methods of time tracing, it was very optimistic that the result of this research would contribute to the chronological trajectory of the methods, techniques, and materials used in Alcobaça *scriptorium*, regarding the Alc. 433 has 5 different additions within 5 different centuries. This study resumed up the chronology of the materials used in the *scriptorium* which, of course, beneficial for upcoming research and increased our knowledge about the medieval Portuguese *scriptoria*. *Figure 1* spotlights the scheme of the

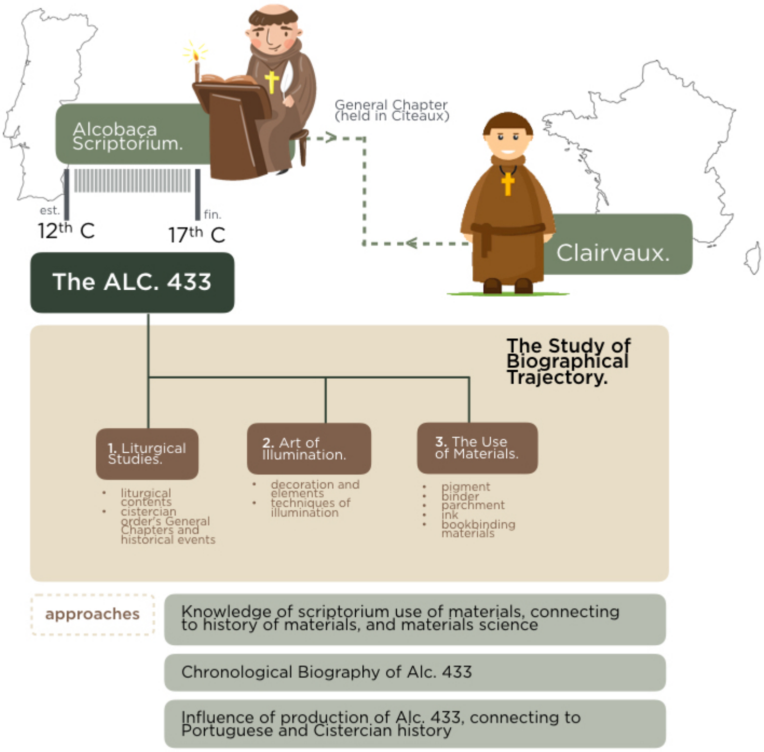

Figure 1: Scheme of Devising the new approach of the Alc. 433.

[3] The study presents 14th century winter breviary Alc. 54 produced in Alcobaça scriptorium that compiled the liturgical study and pigment characterisation. The transformation in style was introduced during second half of the century showing the scriptorium was opened and influence by outside world. The result of this research shows the presence of several pigments used during the Alcobaça scriptorium productive time in 14th century that also linked to this research, such as lapis lazuli (blue pigment) that commonly known to be used in Alcobaça as mentioned to be "liturgical" colour.

[4] The research of "The Secret Behind the Colour of the Book of Birds" traced the distribution of colours in several Portuguese Cistercian monasteries, including its trajectory to Clairvaux (the centre of Cistercian Order), also the influence of other cultures such as Arabic and Jewish trade that might affected the pigment source. The investigation was performed in manuscript contextualisation and the chemical characterisations. The result shows the connection of the colour distribution, including the influence of the trade during the period (12th century).

[5] The palaeography study of this paper "Dating a fragment: a Cistercian litany and its historical context" was investigated only by palaeographic data from the respective manuscript, in order to production year of the manuscript.

process of studies as how result and investigation will complement each other, and approaches that were obtained from this research.

1.3. Experimental Design

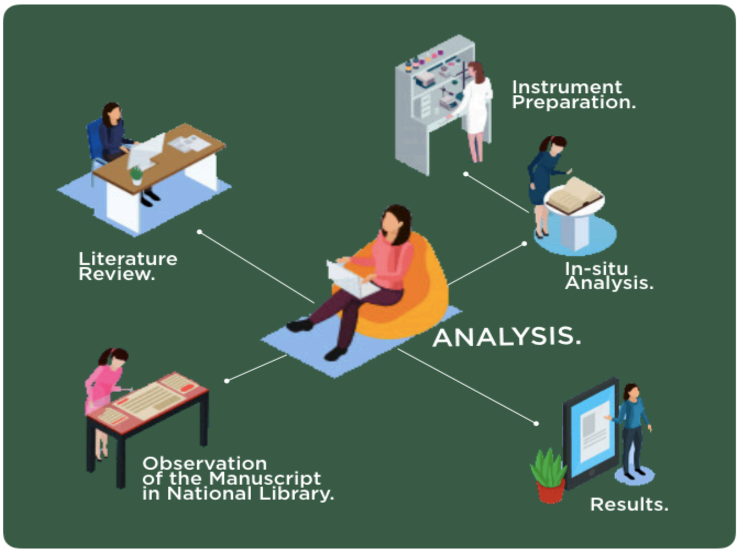

Figure 2 : Flowchart of analysis process containing multidisciplinary investigations

The first step of this research was conducted by the liturgical study of the manuscript and its context, as shown in *Figure 2*. This study corroborated and/or contextualised with liturgy practiced in the Monastery of Alcobaça. From this content, it was obvious to distinguish and determine the period of production due to the typical ceremonies and festive that Portuguese Cistercian conducted in medieval time, for instance different format of hours, and masses (Kerr, 2016). Even though the Liturgical uniformity was a must under the Cistercian order, each manuscript documents the evolution of the Liturgy and a "contamination" of the local context when confronted with the *Statuta*. The record of events, new formulas or prayers, and marginal commentaries were essential to assume the period of time within liturgical context. From different *folia* and quires which are assumed to be added in different years, we could see the changes of styles during centuries of Alcobaça *scriptorium.*

As stated before that the characterisation used are non-invasive and in-situ, it is important to stress that both techniques are highly suitable and recommended for illuminated manuscripts characterisation since it requires no sample, fast, portable, and sufficient. The most common used for illuminated manuscript is usually spectrophotometry, such as the technique used in this research: UV-Vis-NIR FORS. This technique has been used for pigment analysis especially for paintings- in books or other media. Easy to use, UV-Vis-NIR FORS also exhibits the spectra in three regions where we can efficiently observe the molecular vibrations and overtones which led to identification of the molecules. This method was complemented with Hyperspectral Image where we can plot certain spots and obtained the spectra of UV-Vis absorption. This method enabled the mapping of pigments' spatial distribution on the basis of their characteristic visible and near infrared absorption spectral features (Melessanaki et al, 2001). However, both of them are restricted to be sensitive to organic materials. Therefore, another technique which is more sensitive to inorganic materials (to characterise inorganic pigment, specifically) need to be used as well, such as XRF. The h-XRF is very essential for illuminated manuscript characterisation due to its portable instrument. It is necessary to distinguish the key elements in order to obtain an accuracy of the identification. And compare to other inorganic characterisation, h-XRF presents faster and effective technique and data interpretation. In order to obtain the accuracy of the numbers of data, the method of PCA (Principal Component Analysis) and ratio comparison were used. All characterisations complemented each other, because one or two techniques are not enough to determine the result. This multidisciplinary analysis resulted the establishment of the chronological trajectory of the production of Alc. 433. The entire process of the research is illustrated in *Figure 2*.

The research is parted in three parts as seen in *Figure 3*. Firstly, the Historical Biographical Analysis of the Alc. 433 that contains the reviews and literature studies as well as the comparison of the content, artistic and historical elements of the manuscripts. This part mostly focused on the liturgical studies of the content of Alc. 433 and on its context, including tracing back the history of the monastery, and the decoration of the *folia*.

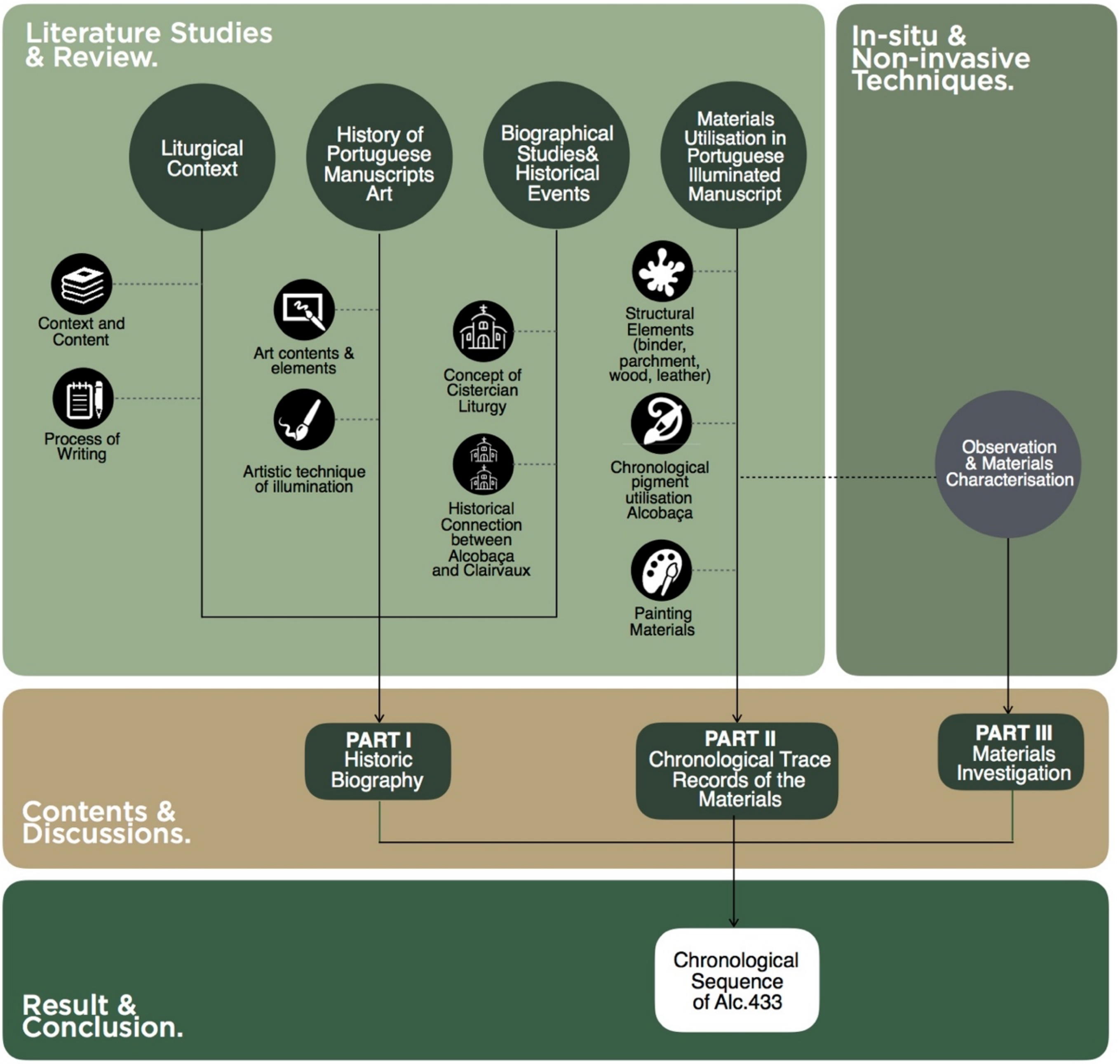

Figure 3 : Research design.

In the second part: Chronological track of the materials resumed up the use of materials in Alcobaça *scriptorium* connected to Alc. 433 (*Figure 3*). The tracking of materials used to produce illuminated manuscript and the colour treatise of medieval pigments were also studied to trace the influence in Alc. 433 (in broader scale- manuscripts produced in Alcobaça *scriptorium*) with Clairvaux or outside Cistercian Context/or Cistercian *scriptoria*.

In order to collaborate the liturgical studies, the observation and materials characterisations are restricted only to several folia (*Figure 4*) as the representatives of *folia* of each chapters, as well as presenting each different period of production.

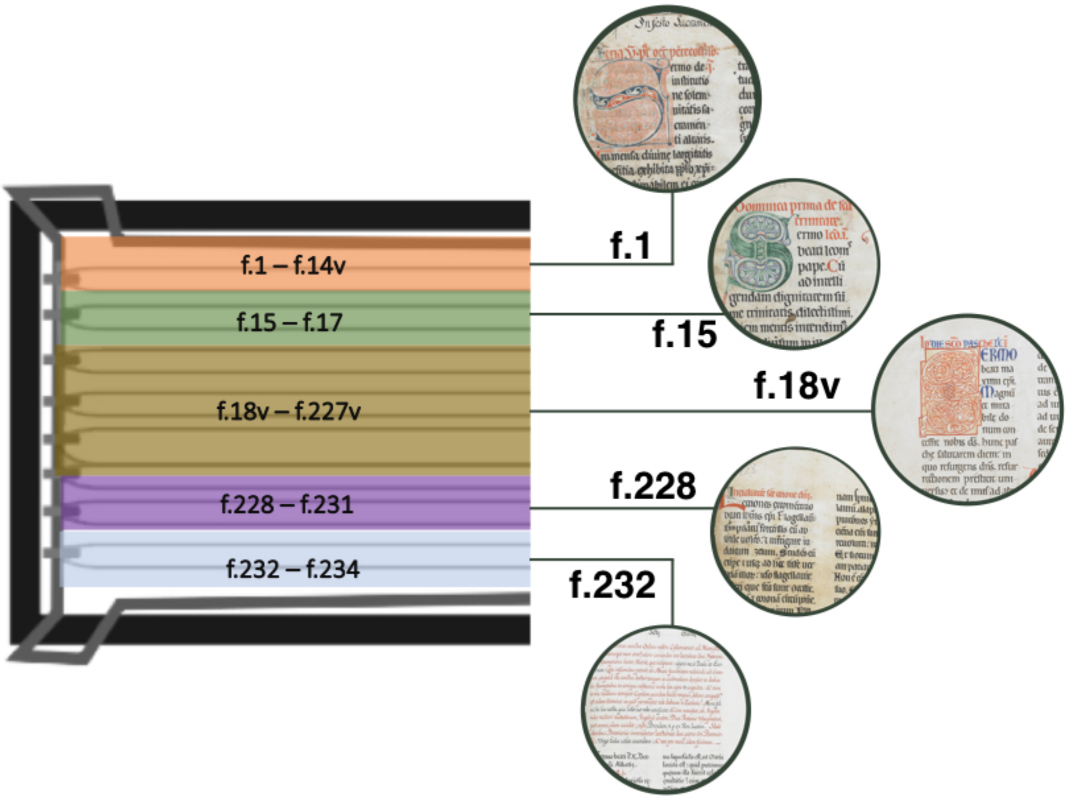

Figure 4 : The folia that were characterised and the insights of each folio that shows different initials and decorations.

Chapter II: Biographical Trajectory of the Alc. 433

2.1. Alcobaça *Scriptorium* and Its Importance in Portuguese Medieval History

2.2.1. Portuguese Cistercian monasteries and Clairvaux

> "*Quinque de causis addiscit homo: ut sciatur scire, ut vendat, ut aedificet, ut aedificetur. Ut sciat, curiositas est; ut sciatur scire, vanitas; ut vendat, simonia; ut aedificet, charitas; ut aedificetur, humilitas." (St. Bernard of Clairvaux)*[6]

Portuguese history cannot be separated from the influence of Cistercian Order. Portugal and some Cistercian Monasteries are part of the European Charter of Cistercian Monasteries, mentioned in 2010 Council of Europe Cultural Itinerary (Martins & Carlos, 2016). The Cistercian itself was introduced in Portugal in the second half of 12th Century, deployed its centre in Clairvaux, France. Between 1129 and 1153, St. Bernard of Clairvaux proclaimed the Cistercian monasticism throughout Europe, including Iberian Peninsula (Rodrigues et al, 2017).

Derived from the word *Cistercian* in French, the Cistercians, also known as white monks, named after the village of Cîteaux, where the monastic order was formed in the late 10th century to the beginning of the 11th century, it was then largely spread throughout Europe, including Iberian Peninsula (Brittannica, 2008). Bridging the Cistercian order from Cîteaux to Portugal, of course, the monks carried the specific policies and rules of monastic life.

During the 12th to the end of the 13th century, around twenty-four Cistercian monasteries (male and female) were founded in Portugal: Tarouca, Tiago de Sever, Alcobaça, Salzedas, Sta. Maria das Júnias, S. Cristóvão de Lafões, Sta. Maria de Aguiar, Maceira-Dão, Fiães, Sta. Maria de Seiça, Bouro, S. Pedro das Águias, Tamarães, Serra da Estrela, S. Paulo de Frades e Ermelo (male); Lorvão, Arouca, Celas, Cós, Bouças, Cástris, Almoster e Odivelas (female) (Sousa et al, 2016). However, this research focused to the Alcobaça monastery and its biographical history within its *scriptorium.*

Figure 5 : The satellite image of Alcobaca monastery captured from Google Earth, showing the topography of the location.

As the 53th affiliation of Clairvaux (Cordonnier, 2009), Alcobaça monastery followed the rules of the Cistercian uniformity. The monastery construction was deployed the topography of its origin: the Clairvaux Monastery (Martins, 2018), which fulfilled the requirement of Cistercian topography that must contain of isolation,

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[6] Quotation from St. Bernard of Clairvaux within the Sententiae 19, states: *"There are five reason why men study. To know, so it be known that he knows to sell, to build, to edify himself. To know curiosity, to be known is vanity, to sell is simony, to build is charity, and to edify yourself is humility."*

water, and stone (Jorge, 1994). It was a good example of ideal plan of unity and simplicity of the ideal monastic life (Martins & Carlos, 2016). The observation through Google Earth in *Figure 5* shows that the location of the monastery is alongside the *Rio* (river) Baça and Alcoa. As stated in Official UNESCO Website, the monastery was declared as World Heritage site in 1989. The construction of the monastic precinct of Alcobaça took many centuries: the monastery started in 1178 (Gusmão ,1992; Gomes, 2002), the church was consecrated in 1252 (Gomes, 2000; Gomes, 2002), two aisles of the cloister (church and Chapter house) were built with the patronage of King. D. Dinis, in the beginning of the 14th century; the *conversos* or lay brothers cloister aisle was probably finished in the 15th century.

2.2.2. Alcobaça *Scriptorium*

The major importance of Alcobaça monastery for Portuguese history was not just about religious Order, but also its contribution for the Portuguese culture. As other medieval monasteries, Alcobaça played an important role in the transmission and cultural preservation of texts. It is common in the culture of European Middle Ages that monasteries produced and copied important religious documents and texts which were saved as precious archives. Thus, it is not surprising that Alcobaça monastery acted as an important cultural centre where the production and copy of valuable documents as the source of religion practices, or other administrative documents were expected to be produced and well-preserved. Fortunately, documentation was very essential in Cistercian Order, which led the Alcobaça monastery to be one of most celebrated and well-known Cistercian monasteries where many codices and administrative documents survived.

Compare to other Cistercian monasteries in France, Alcobaça *scriptorium* can be an excellent case-study (Barreira, 2017) due to impressive survival of books and documents and many of them conserved with several pristine elements. Presumed to begin the production of manuscripts in the last quarter of 12th century, the *scriptorium* was active until early 18th century (Barreira, 2017). According to the database on the *Biblioteca Nacional Portugal* (BNP), the liturgical manuscripts produced are from various types: collectars, psalters, missals, breviaries, lectionaries, a processional, an evangeliary collector, a pontifical and book of hours. Being preserved in BNP, these illuminated manuscripts are coded *Alc.* for the abbreviation of Alcobaça following with the code number.

Concerning Alcobaça *scriptorium*, it must be considered that the constitution of such a library was a long and complex process, and that was not limited to the copying and preservation of books, but was organised around a Cistercian community with common interests, namely cultural interest. It was a long process because the production and/or acquisition of books depended on several factors and was done at the service of the monks, their liturgical needs and readings (collective or individual). In most of the cases, a monastic library was made by the monks, for their own use and the all community, which provided the existence of a *scriptorium* (Nascimento, 2018). Its functioning presupposed within mastery of skills linked to the production of the book, well organisation in all stages, since the preparation of the parchment to the binding, as well as the financial capacity to acquire the respective materials (Barreira 2020). For further recognition of Alcobaça importance, the monastery and its *scriptorium* essential aspect were mentioned in several historical documents. For instance, an Italian humanist, Poggio Bracciolini (1380-1459) mentioned Alcobaça in his letter[7], asking his friend to send him a copy of *integrum* manuscript of the *Noctes Atticae* of Gelilo (Bettini, 2008). This shows the significance of Alcobaça *scriptorium* of preserving important medieval manuscript. And despite all social, political and economic crises, the monks were the main responsible for the care and preservation of the library and the documentation, till the extinction of religious orders in 1834, when the codices went to the BNP.

2.2. Liturgical Study of the Alc. 433

> "*Fida memorie custos est scriptura; hec enim antiqua inouat, noua confirmat, confirmata, ne in posterum notitite temportum diuturnitate obliouioni trandantur respresentat"* (ANTT, Alcobaca, Particulares, maço 1 doc 13)[8]

Liturgy is the centre of the monastic life performed in ceremony rituals throughout the day and it is believed that liturgy is the medium through which Bible and patristic tradition are received, in order to achieve the understanding to glorify God (Reilly, 2019). Liturgical manuscripts are generally divided into the following parts: Calendar (concerning the "civil year", from January to December), the Proper of time (*temporale*)[9] that concerns the feast of Christ, the Proper of saints (*santorale*), that includes Virgin Mary feasts[10] and the Common of saints. This makes liturgical manuscripts meticulously suitable to trace their historical biography.

The Alc. 433 is a parchment codex that measures 460 x 329 mm, (written space 346 x 225 mm) with a total 235 *folia* (text in 2 columns, 27 lines). The *folia* are not visibly numbered. The texts were written in Latin with Protogothic script. Even though the core of the Alc. 433 was assumed to be produced around 1201 and 1250, it is known that some *folia* of this manuscript were added later in different periods due to obvious difference of the context of the liturgy, as well as the illumination. In *Table 1*, the liturgical content listed only for the *folia* that were characterised within this research, regarding its different period of production.

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[7] The letter of Poggio Bracciolini mentioning Alcobaça was the *Lettere II-Epistolarum libri*, where he also mentioned some of his Portuguese circles with direct access to Alcobaça *scriptorium* (Bettini, 2008).

[8] "Treacherous writing is the guardian of memory; for these ancient intuit away, and new he confirms, confirmed, must be handed over to oblivion, not even in the represent of the long duration of time," taken from a charter date in 1176, written in Alcobaça (Nascimento, 1992)

[9] According to Oxford dictionary, Proper of time or *Temporale*, is a part of liturgical offices that varied according to the calendar which consists of movable feast, such as Easter (different Sunday every year), Ascension, and Pentecost (Rose, 2016).

[10] Reverse with the *Santorale* or Proper of Saints, which is fixed every year. The *Temporale* had been recorded to be written in Iberian illuminated manuscripts since 7th century (Zapke, 2007).

Table 1 : The Liturgical Content of the Alc. 433

Foliation	Liturgical Content	Assumed Period	Note
f.1 ***(Added to the beginning of the codex)***	***Corpus Christi* (Office lessons).** *Feria quinta post octavas pentecostes. Lectio prima Sermo de institutione solemne sacrament altaris. Immensa divinac largitatis benefitia….*	Quires added to this codex in early 14th Century (between 1318-1350)	The *Corpus Christi* feast was authorised by the General Chapter in September 1318[11].
f.15 ***(Second addition only with 3 folia)***	***Festa SS. Trinitate* (Trinity Sunday Lessons).** *Dominica prima de sancta trinitate, lectio prima. Sermo beati leonis pape. Cum ad intelligenda dignitatem summe trinitatis….*	Folia added after 1175	The feast was instituted and authorised by the General Chapter in September 1175[12].
f.18v ***(Initial core of the codex)***	***Temporale* (the Temporal or Proper of Time)** *In die sancto pasche. Lectio prima. Sermo beati maximi episcopi. Magnum et mirebile donum….*	Written around 1175 (the manuscript model was written before 1175)	No information available that could provide an accurate date.
f.228 ***(addition)***	***Exaltatio Sanctae Coronae Domini* (Office Lesson of the Exaltation of the Holy Cross)** *In exaltation sanctae coronae domini. Lectiones ex commentario beati Iohannis episcopi….*	Quire added around 1292	This office was authorised by the General Chapter for the French context in 1240, and authorised for the whole Christendom in 1292[13]
f.232 ***(Quire with 3 folia and another quire with 2 folia)***	**Sermo in Assumptione Primus** (Sermon of Saint Bernard Abbot) *Sermo beati P.N. Bernardi Abbatis, lectio prima. Virgo hodie gloriosa caelos ascendens….*	Last addition, around 17th Century	Because of a mention of the brother Brandão (a monk from Alcobaca who wrote the Lusitanian Monarchy in the beginning of 17th Century)

The initial core of Alc. 433 began with f.18v where the *Temporale* or Proper of Time starts with the lessons concerning Easter Sunday (regarding the succession of liturgical time, the first volume of the Lectionary begins with Advent, and stopped before Easter, which is where this volume begins with Easter day celebrations). It is very difficult

[11] Canivez, Statuta, III p. 338

[12] Canivez, Statuta, I p. 82

[13] Canivez, Statuta III p. 258-259

to date this primitive core of the codex with precision. Thus, we can assume that this folio was produced in the Alcobaça scriptorium during its primitive period of establishment around 1175 (Barreira, 2017).

The first addition that this codex starts with f.15, which was added after 1175. This was assumed by the liturgical content containing the *Festa SS. Trinitate* or Trinity Sunday Lessons which was authorised in General Chapter 1175 (Barreira, 2018).

The second addition begins with f.228 which containing the Office lessons of the Exaltation of the Holy Cross, which was authorised in 1240 in French context and 1292 in whole Christendom. This *folio* might have been added soon after 1292 right after Alcobaça *scriptorium* received the texts and formulas for its celebration. The September 14th is when this feast is celebrated - Exaltation of the Holy Cross - dedicated rites of the Holy Sepulchre in 4th century Jerusalem (Baldovin, 2002). In western medieval, the texts explaining the feast was more common to the later medieval Cross piety particularly through Bernard de Clairvaux, focusing the suffering of the human Christ (Van Tongeren, 2000).

Afterwards, the third addition received by this manuscript, starts with f.1 that was likely added between 1318 and 1350, since it contains of the feast of *Corpus Christi* which was authorised in the General Chapter 1318. The feast of *Corpus Christi* celebrated annually on Thursday after Trinity Sunday (Davidson, 2011). In medieval English manuscripts, this feast also recorded post-1318, such as a Franciscan missal with English decoration (Rubin, 1986) which explained this feast spread in Western medieval after the General Chapter 1318 was authorised- including in Alcobaça

The last addition to the codex begins with the f.232. The *folio* was assumed to be aggregated to this *leccionarium* during 17th century due to the mentioning of Frei António Brandão[14] (1584-1637), a Cistercian monk and chronicle who wrote *Monarchia Lusitana*[15] (Barreira & Repas, 2016) on its first four volumes (Gloël, 2017). This supports the presumed idea that the manuscript's bookbinding was done in 17th century which presented in detail in the next chapter. Nevertheless, Brandão depicted the importance of Alcobaça monastery and specifically the *scriptorium* where valuable manuscripts were produced from, within the long history of Portugal.

[14] Frei Antonio Brandão was a native from Alcobaça, and a monk in Alcobaça monastery, mastered in philosophy and theology, and graduated from University of Coimbra. Not only wrote the *Monarquia Lusitania*, he also wrote another historical and poetic works. At the age of 73, he died in Alcobaça on 27 November 1637 (This biography was taken from an old archive without author name, entitled *Archivo Popular: Leituras de Instrucçao e Regreio, Semanario Pintoresco* Volume VI, dated 1842, the book is stamped within the Library of University of California, Berkeley).

[15] Monarchia Lusytana is books within 8 volumes, started in 1597 by a Portuguese chronicle Frei Bernando de Brito (the first and second part), continued by António Brandão (3rd and 4th part), Francisco Brandão (5th and 6th part), Rafael de Jesus (7th part), and Frei Manuel dos Santos (8th part). The books reflected the Portuguese kingdom, its history for over a century (Fernandes, 2007).

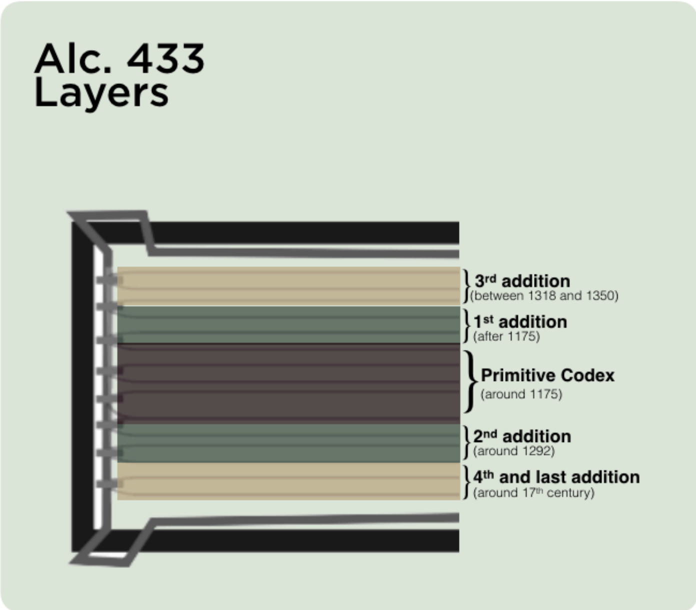

Figure 6 : Scheme of the layers of the Alc. 433 according to the period of the addition.

Based on the liturgical contents of different additions to its core of the codex, it becomes clear that the Alc. 433 received additions over centuries, up to the beginning of 17th century. The clear timeline of the addition is shown in *Figure 6.* It can be observed that the Alc. 433 had a very long time of liturgical use by the monks: copied in the last quarter of the 12th century, shortly after the foundation of Alcobaça monastery, it received liturgical offices as they were being authorised by the General Chapter, which were aggregated during five centuries of intensive use and received its last addition in the 17th century. The complete timeline of the addition and the General Chapter contextualisation can be seen in *Figure 7*.

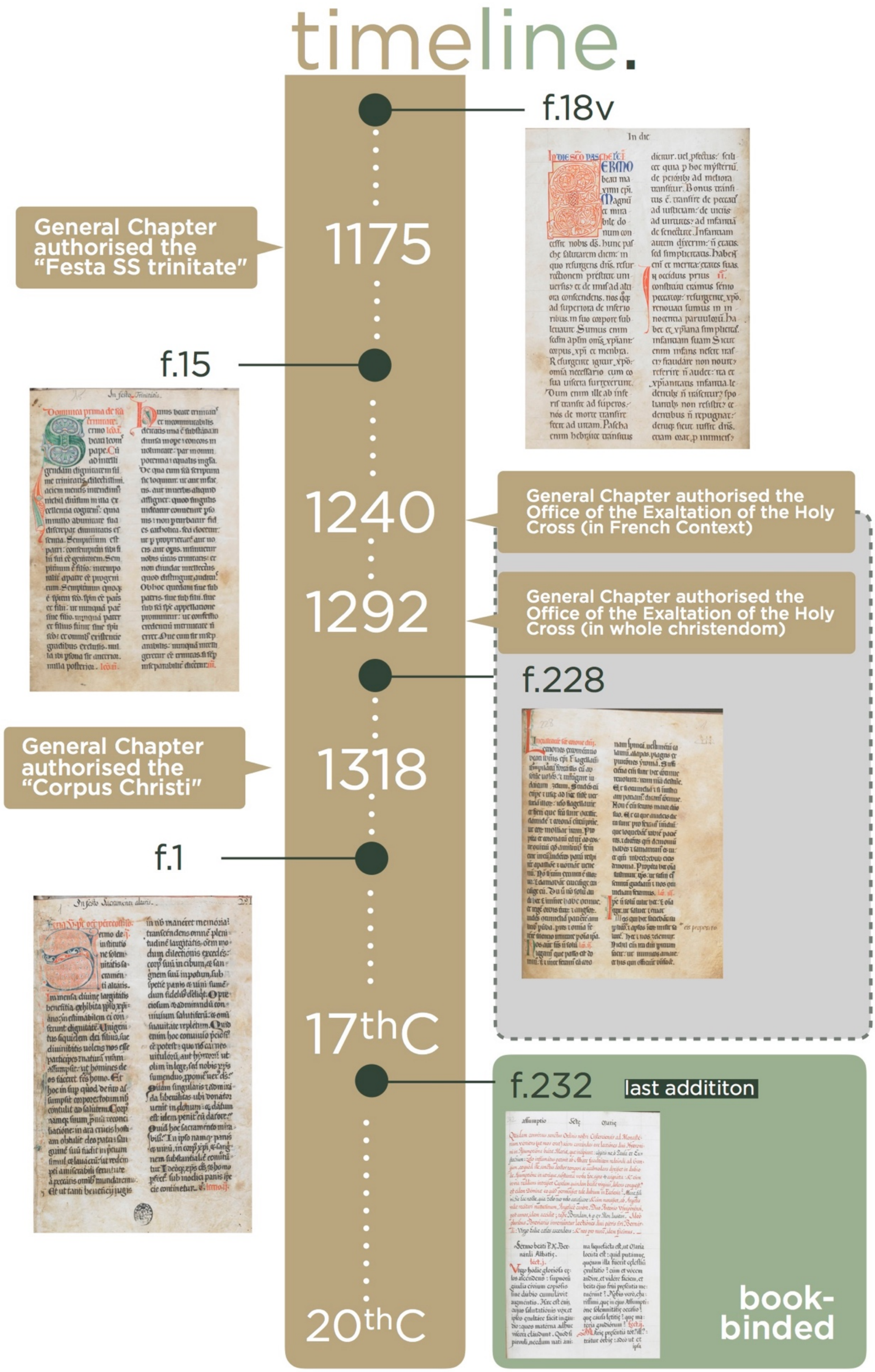

Figure 7 : Timeline of addition of Alc. 433 alongside with the authorisation of the feasts.

2.3. Bookbinding Studies

Figure 8 : The Alc. 433 © C. Casanova.

Bookbinding studies of a manuscript has a significant role in order to give us some clues about the manuscript production period, as well as alteration suffered by the manuscript along its existence. In fact, bookbinding is also important to study the archaeology of the book, to carefully observe the book layer by layer, understanding the sequence steps of the binding process. If pigment production connects to chemistry, then bookbinding studies is linked to the craftmanship due to its importance of requirement of mechanical functionality and materials.

Several bookbinding methods were used across centuries with different materials and techniques. The most important characteristic of a bookbinding that needs to be observed are the text block, and sewing structure, supports and lacing technique, board shapes and covering materials methods.

According to the Szirmai who divided bookbinding methods in several periods, the earliest method of bookbinding in western medieval manuscript is Carolingian binding, around 8th-12th century and it is earliest sewing characterised in Western bookbinding tradition. The sewing materials sourced from vegetable origin, and was frequently used in France and Italy (Szirmai, 2017). The following tradition of bookbinding is Romanesque binding, around 11th until the end of 14th century were leather thongs replace cords of vegetable origin.

As mentioned previously about the uniformity of the Cistercian liturgical books, it may appear the same for the bookbinding. The bookbinding of Romanesque Cistercian manuscript between 12-13th century is described to be stripped down, sturdy, undecorated, and partly made of scrap materials that meant to signal their origin as books produced within newly reformed community that preferred utility and simplicity over superfluous features, supported by the use of heavy unadorned tanned leather and seal skin that prevented any form of surface decoration (Turner, 2019). Usually in Alcobaça *scriptorium*, strong and heavy bindings were commonly found, yet they were mastered by its handcraft, with precise needle-work and detailed wood and leather work (Casanova, 2020).

Gothic binding followed in period of early 14th-16th century (Szirmai, 2017). The bookbinding looked more sophisticated as the trend of manuscript making were widespread around medieval Europe, as illuminated manuscript began to be the trend among the monasteries that led to the more fashioned embellishment resulting the gothic binding that rich with decoration on the cover (Szirmai, 2017).

Decorations and structure shape lead us to propose that Alc. 433 was assumed to be bind around 17th century. The bookbinding structure are sewn on 5 raised cords, which are laced in heavy wood boards covered by a plain brown leather, decorated with blind tools using a geometric pattern (*Figure 9*), such as central diamond inserted in a

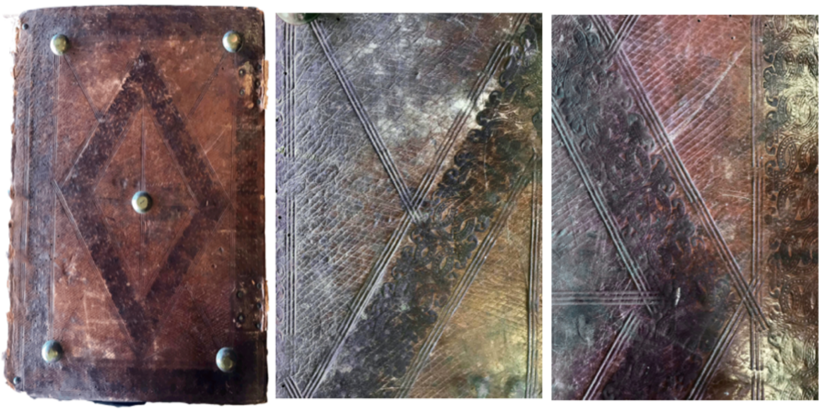

Figure 9 : The front cover of Alc 433 and details of decoration and embellishment on the front cover. © C. Casanova.

larger rectangle which is very common in Portuguese 15th- 17th century binding (Seixas, 2011)[16]. End-bands are very simple, with round shape of a single thread on cord, and the spine is double reinforced with linen straps and parchment. It shows round metal bosses and squares for protection and two fastenings for text-block holding. The binding looks deteriorated, especially in the spine of the book (*Figure 10*).

According to Seixas' doctoral dissertation, similar cover design with the geometric pattern, can be categorised as 'neo-Manueliene' bookbinding style, rather than gothic (Seixas, 2011). Further research could have been done for its leather materials characterisation (binding and endleaves) and analysis to complement the liturgical study and archaeological of the book[17].

Figure 10 : The spine of Alc. 433 shows the deteriorated leather. © C. Casanova

[16] Seixas identified the bookbinding of several illuminated manuscript which has similar cover decorations like Alc. 433, such as EM 20 (Museu Municipal Da Figueira da Foz) bind in 17th century, EM 25 (Museu Municipal da Figueira da Foz) bind in 16th century; EM 26 (Palácio Ducal/Vila Viçosa), bind in 16th century; EM 27 (Palácio Ducal/Vila Viçosa), bind in 17th century,and EM 28 (Palácio Ducal/Vila Viçosa), bind in 16th century (Seixas, 2011).

[17] Author was supposed to characterise the bookbinding and analyse it, but due to the COVID-19 global pandemic, the access was restricted, therefore the Alc. 433 bookbinding was analysed within reviews and reference studies. Hopefully, further research will complete this work.

Chapter III: The Illumination of Alc. 433

3.1. Illumination Contents of the Alc. 433

> "*We take special delight in vision because light and colour have singular beauty, exceeding that of other things that are conveyed to our senses. But it is not simply the beauty and delight furnished by light, colour, and vision that recommend them to us, they also possess epistemic of superiority.*" (Roger Bacon, 1240)[18]

The obvious appearance of illuminated book, compared to other manuscripts, is the illumination. When we turn a page, it always comes embroidered with bright shapes and subtle line with the various decorations, some also filled by illustrations. This relates to illuminated manuscripts' function of symbolising the illumination of the new textual literacy alongside religious devotion and materials culture. Also, this expresses that illuminated manuscript is respectively the richest source for identification of art materials and its technique, because the contents within the illumination were indeed to illuminate those the readers that sometimes the manuscripts were also called by 'display books' in order to describe the colours, decorations, appearances of each page.

In most illuminated manuscripts, the illumination contents generally consist of initial letters, quotations, *marginalia*, headings or *incipit* and closings or *explicit*, *bas-de-page*[19], emblem[20], headpiece[21], tailpiece[22], and miniature illustration. Some also decorated within chrysography. These contents are various according to each manuscript, depends on the various factors such as: period of production, the *scriptorium* and the social environment that influence the activity of the production (such as politics, religious order, and trade), and the type of illuminated manuscripts itself. In liturgical manuscripts, these contents may be found due to its function to illuminate the readers through the words of God present in the context of the liturgy. In the case of Cistercian illuminated manuscripts, the rule of the Order held a significant role on how to trend its contents. However, for Alcobaça's manuscripts, these contents often appeal within various type of decorations, colours, characters, and styles- revealing the essence and meaning behind the art. For example, in *De Avibus* or the Book of Birds produced during 1180-1190, magnificent illustrations of birds were used across the manuscript, which embodies the complete and best example for medieval

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[18] This quote was written in a medieval English illuminated manuscript by celebrated English philosopher, Roger Bacon. The illuminated manuscript was written in 12th century, contains of philosophical works and fragments which are today preserved in The British Library. The quote was taken from a part called *Opus Maius* part 5: *Perspectiva* devoted to light, colour, and vision (Bacon & Linberg, 1996) which philosophically suits the general purpose of illumination.

[19] Scenes or images at the bottom of the page usually unframed that may or may not refer to the text or another image on the page, with French meaning of bottom of the page, and commonly found in gothic illumination (Brown, 2018)

[20] A pictorial allegory or symbolic representation, often companied by a motto to identify a person, family or nation (Brown, 2018).

[21] A panel of ornament, sometimes incorporating a rubric or heading, stands at the beginning of the text (Brown, 2018).

[22] A panel of ornament sometimes containing a rubric or colophon which stands the end of the text (Brown, 2018).

iconographic studies within illuminated manuscripts (Castro et al, 2014). Unfortunately, the Alc. 433 does not contain illustrations nor historiated initials due to its function within religious practices. Nevertheless, it does not reduce its substantial liturgical studies since Alc. 433 was one of most valuable primitive manuscripts produced by Alcobaça *scriptorium*, and its function as liturgical manuscripts produced and added in different periods.

3.1.1. Initial Letters

Immense size, decorative, colourful, shady, illustrated, and noticeable initial letters might be the most well-known content of illuminated manuscripts as it always marks the beginning of a text, of a section, of an important paragraph, or the beginning of a liturgical formula of a prayer in a profound connection with the text. The significant shape and size of initial letters are not only for the art purpose, but also holds other intentions: level of significance. The initial letters have their hierarchical system within the content of the liturgy, the bigger size, the more motifs, the more significant the level. Therefore, based on this hierarchical system, initial letters are divided into major and minor initials. The major initials, known as *litterae horissales* (Brown, 2018), were decorated with delicate geometric and foliate motifs. In reverse, the minor initials, or *litterae notabiliares,* served as an adjunct to punctuation (Brown, 2018). Sometimes, the minor initials consist of several grades. In other words, the firsts are the capital letters of each chapter, and the seconds are enlarged letters to begin or highlight a new paragraph within the same chapter.

From the perspective of decoration, initial letters are divided into historiated and decorated initials. Historiated initials are the initials presenting identifiable scene or figures relating to the text, meanwhile the decorated initials are composed of non-figural and non-zoomorphic decorative elements (Brown, 2018). The Alc. 433 are full with decorated initials, but do not contain historiated initials. The decorated initials are major and minor initials (with various size) painted in red, green, and blue.

Folio 18v (estimated around 1175)

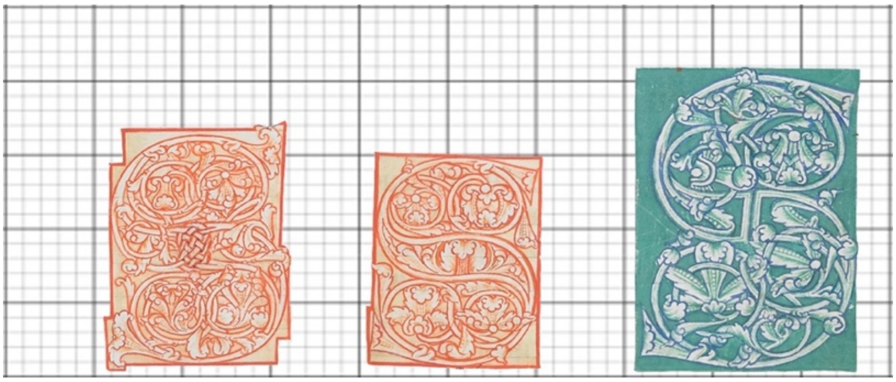

Figure 11 : Comparison between initial letters (from left) f.18v, f.115, and f.56 which shows the different size on 1 mm grid. © Biblioteca Nacional de Portugal.

The initial letter S (*Figure 11*, left) was painted in red pigment, and inserted with a light-yellow background. The letter was formed within a kind of stem of a plant, which makes it look foliated. The decorative S shows the details of white vine-stem border (*bianchi girari*), which spread from Italy in 15th Century despite the fact that it is found on Italian manuscripts from 12th Century (Brown, 2018), so it matches the possibility that this style was a trend of initial letter used in 12th century Alcobaça (Benulic, 2012).

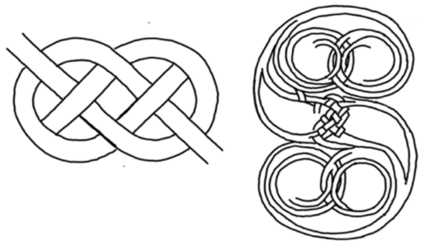

Figure 12 : Basic interlacement detail (left) and the initial letter of S sketched from f.18v without foliated details (right).

The stem was left blank on the parchment. Inside the curves of S, Interlacement band is seen (*Figure 12, right*), which was chiefly from byzantine and Romanesque period as well as old Frankish (Meyer, 1957). In *Figure 12*, the comparison between basic interlacement and the infoliated initial letter shows the clear narrow interlacement within the S curves.

Similar initial letters also appeared in f.56 and f.115. The one in f.56 is painted in solid green within the frame with similar white vine-stem border, meanwhile the initial in f.115 looks more alike painted in the same red and shady yellowish within the frame. The comparison of the size of these initials is shown in *Figure 11*.

Folio 15 (estimated after 1175

Figure 13 : The initial letter S in f.15: the detail initial letter (above) and the initial letter within the text. © Biblioteca Nacional de Portugal

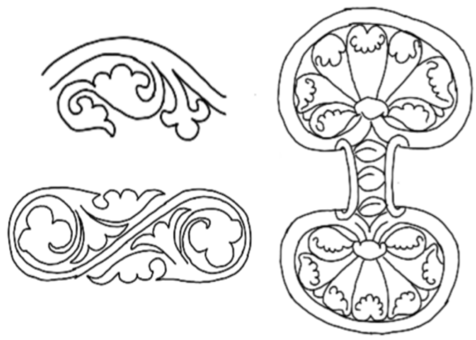

Figure 14 : The basic undulate ornaments (left) and the ornaments inside the S curve of the initial letters on f.15.

The initial letter present in f.15 is decorated by green and orangish red colour with undulate ornament (*Figure 13*). The S shape looks clearer rather than the initial in f.18v. We can see the undulate ornaments within the S curves. Fingerprinted from the Romanesque period, its undulate ornament is evidently schematised in *Figure 1*4.

The extension of the initial letter, or *cadel*, a calligraphic extension to ascending or descending strokes of letters, usually on the first or the last lines of a page (Brown, 2018). As shown clearly in the initial letter in f.15, the *cadel* was extended until the eight line from upper texts (*Figure 13*).

Figure 15 : The variety of initial letters S from Alc. 421 which was produced in 12th -13th century. © Biblioteca Nacional de Portugal

The shape of undulate ornament of the initial letter looks so similar with the initial letter from Alc. 421 (*Figure 15*) which was estimated to be produced in 12th – 13th century (Muralha et al, 2012). All the S letters have *cadel* in the corner of left side, and a short extended pointed shape on the top corner side. The foliated undulate ornaments look very similar, especially the green one. This generates the possibility that there was likely a layout model and format for the design of the shapes and ornaments for every letter in this period.

Figure 16 : The initial letters of (from left) f.134, f.151 and f.16 of Alc. 433. © Biblioteca Nacional de Portugal

Similar S initial letters found in Alc. 433 (*Figure 16*) in f.16, f.151, and f.134 which are in the same chapter. The foliated undulate ornaments are present but it was painted in simpler way without any colour gradation that appeared in the S body letter in *folio* (even though it is also possible that the gradation of green is caused by deterioration).

The S letter present in f.16 looks similar with the initial on f.15, as both of them appeared with *cadel*. The comparison between both (*Figure 17*) shows the S body of S on f.15 is twice larger than on f.16. This means the hierarchy of the initial letter in f.15 is higher than in f.16 (*litterae horissales).* It is possible that f.15 and f.16 were produced in the same year.

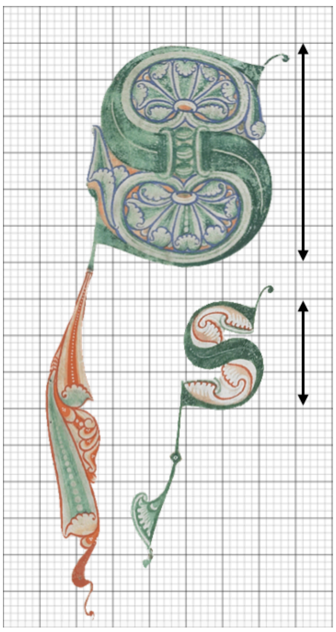

Figure 17 : The comparation size between initial letters from f.15 (above) and f.16 (below) on a 1mm grid. © Biblioteca Nacional de Portugal

Similar letters like this were found in other *folia* which assumed to be added in the same period (*Figure 18*). The ornaments were also similar and painted with red and green. The variety of the letters can be seen on *Figure 17*. According to the ornamental decoration and the comparation with Alc. 421 (*Figure 15*), it is likely that the f.15 was added in 12th-13th century- complementing the liturgical content, to be exact after 1175.

Figure 18 : The variety of the letters with similar decorations and ornaments, from left: C (f.16v); Q(f.15v); N(f.15v); C(f.16v); and S (f.15). © Biblioteca Nacional de Portugal

<u>*Folio* 228 (estimated after 1240 or after 1292)</u>

Figure 19 : Initial letter of f.228 (left), the minor letters of "N" (centre) and "I" (right), also from f.228. © Biblioteca Nacional de Portugal.

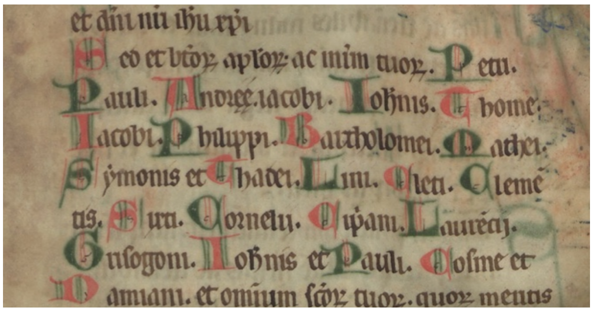

Figure 20 : Alc. 250 produced around 13th century, f.128 shows variations of initial letters that looks similar with Alc. 433 f.228. © Biblioteca Nacional de Portugal.

The major initial letter in f.228 is a red L (*Figure 19*, left) drawn in the left corner of the page. It almost does not look like any other initial letter which is decorated distinctly in Alc. 433. The L letter is only surrounded by a simple filigree: delicate, conventional designs, usually in gold, on a flat coloured surface, in overall patterns of curling vines, branches, and sprigs and/or leaves; used as a background to miniatures and initials and on band borders and miniature frames (Scott, 1996). In this *folio,* the filigrees are painted by green and red.

The minor letters of N and I (*Figure 19*, centre and right) also have the same type of decoration of the L. Unfortunately, no similar initial letter was found within the other *folia* of Alc. 433. Furthermore, Alc. 250 which was produced during the 13th century (Barreira, 2016) presents the similar decorations of the initial letters (*Figure 20*). This supports the presumption that f.228 was also produced within this period.

<u>*Folio* 1 (estimated after 1318)</u>

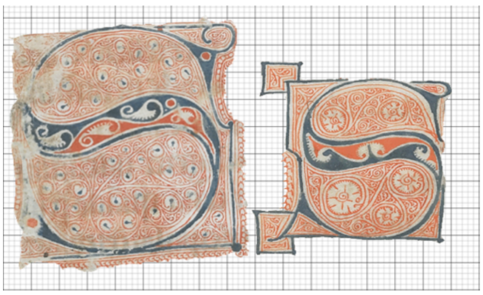

Figure 21 : The comparison of initial letters on f.1 (left) and f.10 (right) of Alc. 433, within 1mm grid. © Biblioteca Nacional de Portugal.

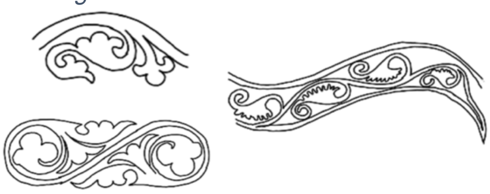

Figure 22 : The basic undulated ornaments (left) and the undulated ornament within the body of the S of initial letter on f.1 (right).

Another S initial letter was painted in f.1 (*Figure 21*, left) with a different decoration of the ones present in previous *folia*. The S is painted with a deep blue, ornamented with a foliated undulate band inside the S body which is left blank (or another probability to be painted by white). The undulated band can be seen clearly in *Figure 22*, compared with the basic undulated band.

The body letter is surrounded within the complex red pen-work decoration, rich with *rinceaux* ornaments. Derived from Old French of rain, was well-known to be decorating the French illuminated manuscripts dated from 13th century (Burin, 1985), which complements the liturgical content that estimated this *folio* was added after 1318. The ornaments

Figure 23 : Initial letter of "L" in Alc. 433 f.5v. © Bibilioteca Nacional de Portugal.

Figure 24 : Initial letter of "U" in Alc. 250 f.126, produced in 13th century. © Biblioteca Nacional de Portugal.

of the S body form a fretwork[23] knot because it features repeated openwork rectilinear geometric pattern (Brown, 2018). The fretwork was already present on European medieval manuscripts mostly in those produced during 8th and 11th century (Wagner, 2011). The initial letter actually has the *cadel* straight down until the bottom of the page, but unfortunately it is deteriorated leaving only the little trace. The body and fretwork were also deteriorated on the left side.

Many similar initial letters are found in Alc. 433 with the same ideas of body letter surrounded by pen-work decoration. Most of them are minor initial letters, but an identical initial letter presents in f.10 (*Figure 21*). The shape of S, and the variety of colours are the same, even though the band within the S body is simpler than in f.1. However, the red pen-work decoration surrounding the body letter has significant different size forming the floral band-alike (it looks like floral band but not precisely floral or flower-shaped band). In f.1, the ornaments are uniformed, repeated at the same proportion bended together, but in f.10, the ornaments are not uniformed nor repeated, thus it cannot be defined as fret-work. The comparison size also shows that initial letter in f1 has the higher hierarchy than in f.10, as seen in the comparison in *Figure 21*.

Similar style and fretwork were found on f.5v of Alc. 433 as well, within the letter "L" (*Figure 23*), showing that f.5v could have been produced in the same period as f.1, as well as being produced under the same Chapter. Interestingly, a very similar design, ornament, and colour was present on Alc. 250 in f.126 (*Figure 24*) which was presumed to be produced within 13th century (Barreira, 2016). This similar initial letter complements the estimation of the production of f.1 (after 1318).

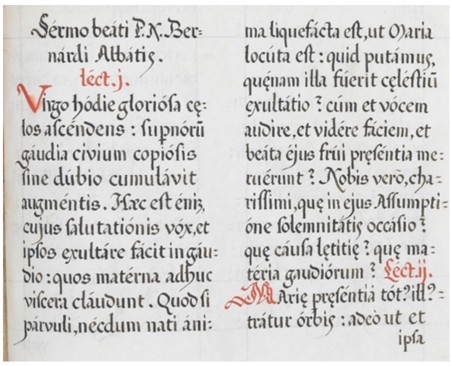

Figure 25 : The beginning of the chapter of f.232 that shows no decorated initial letter. © Biblioteca Nacional de Portugal.

<u>*Folio* 232 (estimated around 17th Century)</u>

The page in this *folio* is dissimilar with the previous others. The is no decorated initial letters which makes it really plain. The capital letter was only painted by written red-ink without any ornament (*Figure 25*).

[23] Fretwork, a knot or ornament that consists of two lifts, or small fillets, variously interlaced or interwoven, and running at parallel distances equal to their breadth, every turn of which and intersection must be at right angles, they were used by the ancients on flat members as the faces of the Corona or leaves of cornices (Hitch & Austen, 1734)

3.1.2. *Incipit* and *Explicit*

The heading or *incipit* can actually be found in every *folio* analysed in this work, painted in red, most of them are written next to the initial letters. The incipit marks the beginning of each chapter, or highlight the importance of the sentence(s).

Figure 26 : The incipit in f.18v. © Biblioteca Nacional de Portugal.

In f.18v, the incipit written *In die sancto pasche. Lectio prima. Sermo beati… (Figure 26)* marks the beginning of the lection and the time of Easter. This complements the liturgical study that explaining the context of this *folio* is about "The Proper of time". The *incipit* is painted in red and blue alternately, except the word *pasche* which painted by both blue and red. The letters were painted with simple decorations of line ornaments.

Figure 27 : The incipit of f.15 © Biblioteca Nacional de Portugal.

Unlike in f.18v, the *incipit* in f.15 (*Figure 27*) follows a simpler style. It was only painted in red with no other decoration or ornament. Written *Dominica prima de sancta trinitate, lectio prima,* it marks the beginning of "Sunday's Holy Trinity Lesson", as explained in the previous liturgical analyses (See Chapter II). The same style *incipit* also appeared in f.228. The difference was in the beginning of the sentence, the initial letter "I" was painted with green (*Figure 28*) and followed by red.

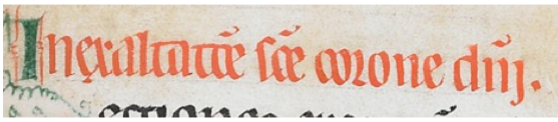

Figure 28 : The incipit of f.228. © Biblioteca Nacional de Portugal.

Written *In exaltation sanctae coronae domini*, it marks the beginning of the "Office Lesson of the Exaltation of the Holy Cross". This is similar with the *incipit* in f.1 (*Figure 29*) which starts with the initial letter painted in different colour (blue). The colour is deteriorated but from the following word, we can read *Feria quinta post octavas pentecostes* marking the beginning of the Office Lessons.

Figure 29 : The incipit of f.1 with the initial letter and enlarged view. © Biblioteca Nacional de Portugal

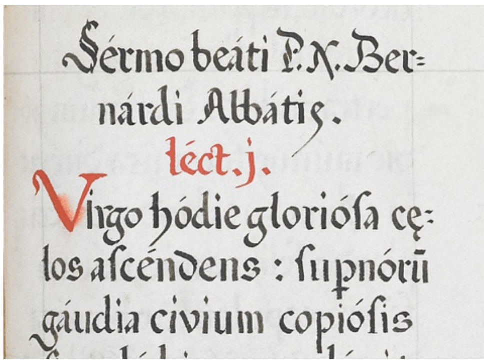

Figure 30 : The incipit of f.232. © Biblioteca Nacional de Portugal.

The last one, the f. 232, (*Figure 30*) presents a different *incipit,* written with iron gall ink, likewise the remain paragraphs. It can be categorised as incipit because the writing position is above the first paragraph and the initial letter.

Observing the *incipit* in those five different *folia*, it is possible to see that the trend of red *incipit* since the end of 12th century was consistently used until the 13th century, as seen in f.15, f.228, and f.1.

Moreover, in f.228 and f.1, the *incipits* start with different colour, meanwhile the older incipit was more colourful.

The *explicit* which marks the closing of each chapter can only be found in f.14v, f.17, and f.234, as the closing of different chapters from different *folia* categorised in this research regarding the trend of the decoration and the using of colour. However, this does not mean that the *explicits* were written in the same year or period. There always lies the possibility of its addition in different times, even though the styles used were similar. This is could be the influence of the uniformity concept of liturgical source in Cistercian order and its consistency through centuries.

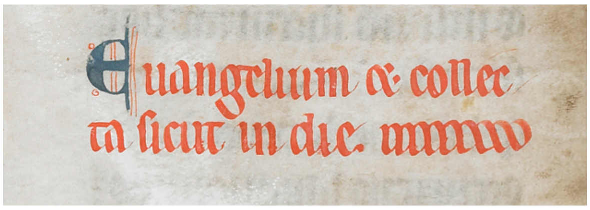

Figure 31 : The explicit in f.14v. © Biblioteca Nacional de Portugal.

The first *explicit* is in the f.14v (*Figure 31*). If we observe from the order of the page and foliation, it is supposed to be the end of f.1 which consists of the "Office Lesson of *Corpus Christi*". The template of the decoration also matches with the *incipit* in f.1 which begins with different colour capital letter.

Figure 32 : The explicit in f.17. © Biblioteca Nacional de Portugal.

The next explicit in f.17 (*Figure 32*) is the end of the chapter started with f.15. The *explicit* in this *folio* is quite unique. The geometrical patterns were decorated above the *explicit* to mark the end of the chapter. Painted by green, red, and yellow, two blocks of different patterns end the chapter that begins from the f.15 (painted by bold bottle green). The *incipit* of the f.15 also matches with f.17 which written with red pigment only.

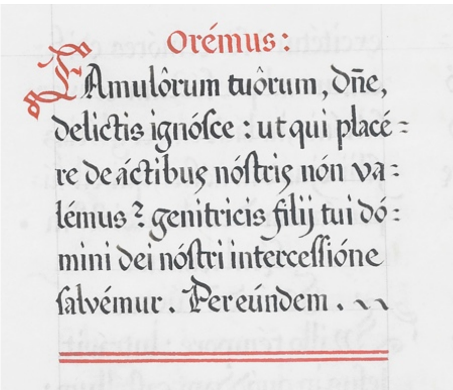

Figure 33 : The explicit of f.234. © Biblioteca Nacional de Portugal.

The last *explicit* was found in f.234 (*Figure 33*), marking the end of the chapter started with f.232. As the *incipit* in f.232, the explicit was also written in simple text, without any decoration. It is ended by two red lines. As already mentioned before, this chapter is evidently different. Everything looks simpler without any meticulous details, ornaments, or colours. This comparison complements the possibility that f.232 was produced within 17th century- the latest within the Alc. 433, together with the bookbinding.

3.2. Illumination Technique

In European middle ages, painting methods were various, according to the artist style and its period. The use of the materials of the painting media is a contributing factor to create an aesthetic yet endless painting that could pass through centuries. Unlike in any other grounds, book painting in monastic context has some specificities because in medieval time books were created and made within the influence of a monastery for specific purposes, thus only particular illuminators who had access to do it. It is known that illuminators learned illumination apprenticeship since they were children over several years, followed by monastic *scriptorium* training and years of process of knowledge acquisition (Panayotova & Paola, 2016). Specifically purpose as religious texts, such as liturgical manuscripts (because it was linked to the way of living of a specific community or religious order), it was also a form of dedicating soul and mind to God.

It is obvious that illuminated manuscripts are not *illuminated* manuscripts at all without its illuminations, that is why the technique of creating the illumination through ornaments and figures is probably one of the mysteries that researchers have tried to figured out. Certain monasteries within certain periods must have their own methods and techniques to make a complicated and detailed illustrations in initial letters or other figures. Regarding that illuminated manuscript has been precious medieval artefacts, the knowledge of the illumination technique must have been preserved for centuries. It is also known that certain religious Orders had their own preserved methods that passed from one monastery to another to keep their authenticity and uniformity- especially in the Cistercian Order. That is why, in this research, the historical reconstruction is significant to find out the best way how to do the illumination.

Known as detailed piece of art, illumination is not easy to be (re-)produced. In European medieval art, the sketch models are known to be used and to be copied as it was accepted by the monastic rules (Panayotova & Paola, 2016). The Göttingen Model Book (GMB) and Berlin Model Book described how the 15th century German illumination was produced. It describes step by step how a foliage scroll was created including its sketching and colouring process (Scott et al, 2001). The sequence step of the illumination is pretty straightforward yet very detailed painted: raw sketching, outlining, shadowing, and highlighting. Although it is very detail yet promising stages to make illumination, it is stated that the model book was not suggested to be a model book for illuminators (Calkins, 1978).

The best approach to study the illumination process in sequence step is to analyse from the unfinished initials that sometimes can be found in one of several illuminated manuscripts. By making comparison between one and another similar illumination, it is possible to analyse the steps of drawing and painting. Because it is complicated to make sure the exact sequence of illumination procedure, this is the best way and the closest approach to study the steps alongside historical reconstruction of with experimental illumination procedures. Thus, we understand not only the stages, but also experience the detail work of the illuminators as well as manage to compare different hypothesis of the procedures. For example, the similar illuminations present in Book of Birds from different scriptoria in Portugal,

Figure 34 : The unfinished illumination from (clockwise): Lorvão 5, (f.16); Sta Cruz 34, (f.32); MS 117 (f.139); Alc. 238 (f.206v). © Arquivo Nacional da Torre de Tombo; © Bibiloteca Pública Municipal do Porto; © Biblioteca Nacional de Portugal; © Bibliothèque Nationale de Troyes.

compared to the scriptorium in Clairvaux (*Figure 34*) show the unfinished painted figure of birds that clearly spotlight the step of how illumination technique was produced.

Another method was used in earlier 8th Century using backlight tracing methods, used for the Lindisfarne Gospel which was produced in the Monastery of Lindisfarne[24] (Backhouse, 1981). It is said that the drawing sketch was traced on the glass with light behind the model sketch. This method was believed to be used after the appearance of trace marks under oblique light. Furthermore, this method was also described by Cennino Cennini in *Il Libro dell'arte* in 15th century which marked its utilisation in this period[25].

3.2.1. The Process of Illumination Technique in Alcobaça Scriptorium

As previously mentioned, the initial ornaments were produced with strong influences from French, Iberian and Italian ornaments, as same as the illumination technique. The book of birds or *De Avibus*, produced in 12th century Portuguese monasteries (Alcobaça, Lorvão, and Santa Cruz) shows its connection with the MS. 117 from Clairvaux which also produced around 12th century (Miguel, 2012), as seen in *Figure 34*. Whereas its drawing and techniques bridges the connection between the Cistercian Portuguese with the Clairvaux, as well as in liturgical

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[24] Lindisframe Gospel is a 8th century Anglo Saxon/Insular (Brown & Clark, 2004) codex with intricate illumination of the letters of its incipits, the so-called 'carpet' pages, the evangelist portraits, the elegant arcades of its canon tables (Watson, 2017)

[25] Cennino Cennini describe the backlight method in his book (Il Libro dell'Arte), chapter 8 about The Method of Drawing quoting "*In what manner you should begin to draw with a style, and with what light*", also *"… to acquire command of hand in using the style, begin to draw with it from a copy as freely as you can, and so lightly that you can scarcely see what you have begun to do, deepening your strokes little by little, and going over them repeatedly to make the shadows. Where you would make it darkest go over it many times; and, on the contrary, make but few touches on the lights. And you must be guided by the light of the sun and the light of your eye and your hand; and without these three things you can do nothing properly. Contrive always when you draw that the light is softened, and that the sun strikes on your left hand…" followed by on Chapter 9 entitled "How to arrange the light and give chiaroscuro and proper relief to your figures" with explanation "…be sure to give relief to your figures or design according to the arrangement of the windows which you find in these places, which have to give you light, and thus accommodating yourself to the light on which side so ever it may be, give the proper lights and shadows."*

study, it is assumed that the influence of illumination technique is as strong as the liturgical content. Nevertheless, it is complicated to study and get the exact result of how illumination was produced in Alcobaça *scriptorium*.

Additionally, some manuscripts were left with unfinished initials such as the initials in Alc. 360 that was produced in the 12th century (Cavero et al, 2016). Comparison of finished and unfinished illuminations from the same manuscript (f. 55v, f.113v, and f.123v), as seen *in Figure 35*, clearly shows the possible step of the production.

Figure 35 : The comparison between unfinished initials (left and middle: f.55v and f.113v) with finished initial in f.47v, from Alc. 360, produced around 12th century. © Biblioteca Nacional de Portugal.

in Alc. 360 on f.55v and f.113v (*Figure 35*, left and middle), it can be seen clearly the trace of the sketch of the initials, also unfinished sketch of the border framing the "h" body letter, sketched by lead-point. Comparing the finished initial in f.47v (Figure 35, right) it is obvious that the frame of the letter and the detail decoration were painted later. Using this study by comparing between finished and unfinished initials, and historical reconstruction undergoing several possibilities of steps, such as: (1) Hard-point sketching, (2) lighter colouring, (3) darker colouring, (4) text framing, (5) shading, (6) highlighting, (7) detailed ornament drawing. This hypothesised sequence of the illumination sequence was applied to the historical reconstruction of the Alc. 433 of f.18v in this research because it was produced in the same *scriptorium* as well as the same period: the last quarter of the 12th century, when the Alcobaça *scriptorium* just began to produce manuscripts. However, it is unlikely to guess how the illuminator sketch the hard-point on the parchment. Several methods have been conducted referencing some manuscript production in 15th century Frankish manuscripts such as stencil lettering and the use of model book as in 15th German illuminated manuscript. As there is no reference on how to do the precise sequence of illumination production in Alcobaça, the experiment of the historical reconstruction revealed promising ideas to the new approach of the illumination process.

3.2.2. Historical Reconstruction of Alc. 433: f.18v.

The f.18v was chosen to be historically reconstructed considers to the factor that the Chapter includes f.18v is in the primitive set of *folia*, as well as the initial core of the manuscript, thus studying the illumination technique of f.18v opened the opportunity to explore the illumination technique in Alcobaça. The drawing and painting technqiues are good example to study the illumination technique of Alcobaça scriptorium in its beginning of the active year (second half of the 12th century). Moreover, the initial of f.18v does not only painted with red, but also yellow, which was assumed to be yellow lake pigments (organic dye)[26]; therefore, this historical reconstruction completed the purpose of the characterisation as well.

The historical reconstruction of the drawing procedure of f.18v followed several possibilities of methods which was referenced from previous studies (Stijnman, 2000; Cannon, 2011; Adcock, 2002; Nascimento, 1989; Leturque, 2013; Cardon, 1999; Calkins, 1989; Kennedy, 1986; Antoine, 2007; Correia et al, 2014); as the scheme in *Figure 36*. In total, 4 methods were reconstructed using transfer and freehand copy methods.

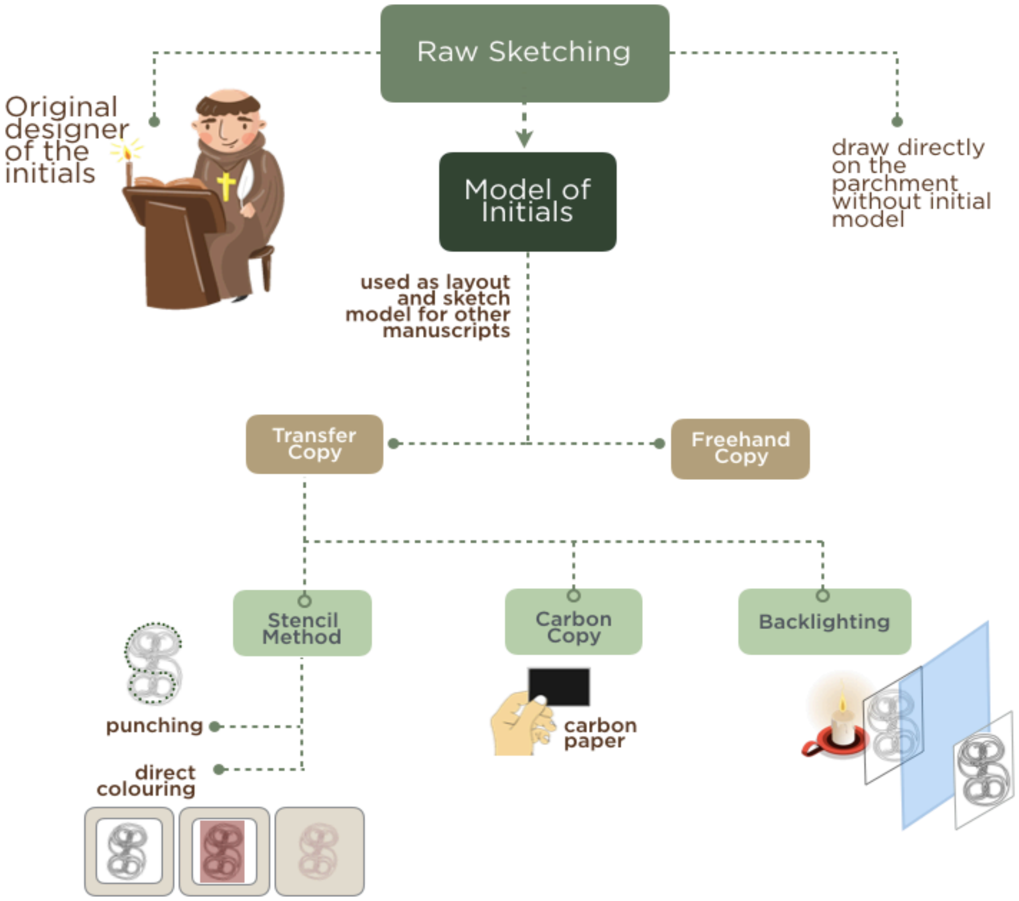

Figure 36 : Scheme of several assumed illumination techniques of Alc. 433 f.18v.

[26] According to the result of UV-Vis-NIR FORS, Hyperspectral Image, and h-XRF analysis presented in the next chapter.

Regarding the historical reconstruction, two major steps were followed: copying or transferring, and painting[27]. The transferred models were copied from the original manuscript, printed in the original scale, thus all the copies made are in the same size as the original Alc. 433. The initials were then copied within 4 methods: stencil (punching and cutting), backlighting, and freehand method. Each transferred copy is coloured using lapis lazuli (blue), organic lake dye synthesised in laboratory (yellow), and vermilion (red)[28].

3.2.2.1. Transferring

Transferring was the first step of the historical reconstruction that allowed to copy the original model that was then molested into colouring. For this, a printed copy of the capital letter presented in f.18v with the same scale of that are present in the original manuscript was used[29].

<u>Stencil Methods</u>

Stencilling is a tracing method, majorly used in this reconstruction due to its specific and detailed accuracy. This method is known to be used since the 11th century in illuminated manuscripts (Heagney, 2004). Stencilling is made by piercing the design that cut out from a sheet (paper) which will be transferred to a surface by passing ink or pigment through the stencil openings (Kindel, 2003). In illuminated manuscript, the stencil technique appeared as under-drawing in the result of manuscript investigations. It can be known by the sign of dot of carbon and trace of metal-point (lead, silver, or tin) that mostly lay under the pigment (Turner et al, 2019).

The transferring trials in the historical reconstruction were constructed using different methods, some resulting in very clear images of initial letters. The stencil methods using cutting (*Figure 37*) and punching (*Figure 38*), were combined, resulting in a clear transfer into the paper. The procedure of the stencil methods is very simple,

Figure 38 : Stencil method model by cutting.

Figure 37 : Stencil method models by punching.

[27] Detail steps of the historical reconstruction is presented in Appendix V.

[28] The pigments of the colour paints (red, blue, and yellow) were chosen according to the result of the characterisation present in next chapter. The method of colour reproduction is explain in Appendix, referenced from Medieval Treatise.

[29] The same scale is used in order to make historical reconstruction as close as possible, with consideration of size of the ornaments of initial letter that will influence the preferable tracing methods.

as pressing charcoal powder to each hole and left the paper under the model traced with the charcoal. Then, the dots were linked by using a lead-point. Transfer by cutting was applied by directly colouring a paper in the model and leaving the pigment on the model opening. The direct colouring trial of cutting stencil using commercial watercolour also resulted in a clear transfer.

<u>Backlight Drawing</u>

The backlight model was produced by setting up a light lamp under the glass with the original model cut on a specified area (*Figure 39*). The original model was the printed copy of the initial in f.18v in the same scale of original Alc. 433. The light from the lamp passing through the original model was, then, traced on the paper using a lead-point.

The result of backlight drawing was quite clear in the body of the S (*Figure 40*). However, the details of the ornaments inside the letter must be added after the backlight tracing process done.

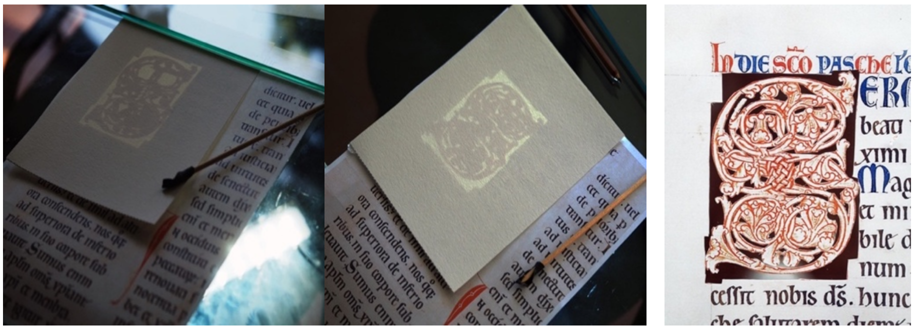

Figure 39 : The set up of the backlight drawing (left) and the light passing through the original model, ready to be traced (middle), and original model which was cut in the specific area to let the light pass through.

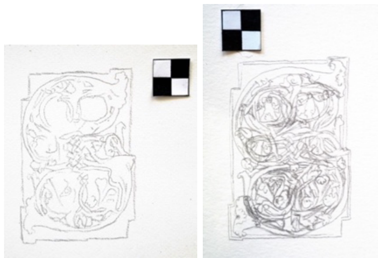

Figure 40 : The result of transferring using backlight method: without (left) and with detail ornament added (right)

<u>Carbon and Lead Copy</u>

Figure 41 : The result of tracing by carbon (left) and lead (right) copy.

The carbon and lead copy method were undergone by greasing charcoal and lead on trial paper and parchment, followed by tracing the original model using both approaches. Even though the charcoal worked and resulted a very intense and clear copy of the initials (*Figure 41,* left), the lead left vague trace which cannot be used as illumination mode (*Figure 41*, right). On parchment, the carbon powder was dispersed and could not easily removed, hence this transfer method was not likely suitable to be used.

Freehand Copy

As an illuminator that might have trained for years, it is expected that initials were also drawn directly with freehand by the illuminator. Judging from the initial models that looked similar (Chapter III: 3.1.1), if the illuminator drew it directly, it is likely that he still used the model and scaling calculation, such as distance between the body S letter and between the ornaments. Judging as a 21st century point of view[30] with slight experience of drawing, freehand drawing on initial is not relatively difficult. Within years of experience, it is possible that illuminators drew the initials in freehand style and even develop the ornaments freely as they wished that could possibly lead as the signature of their work or of the *scriptorium*.

Despite it is called by freehand, to obtain the same size and the precise ornament, the scaling and calculation of distance was necessary. For this, the frame was firstly sketched following by distance calculation of S letter, then drawn carefully in details. The result may not exactly the same as in carbon copy, however, it still usable and possibly to be used in medieval time. The result of the sketch of freehand transfer can be seen in *Figure 42*.

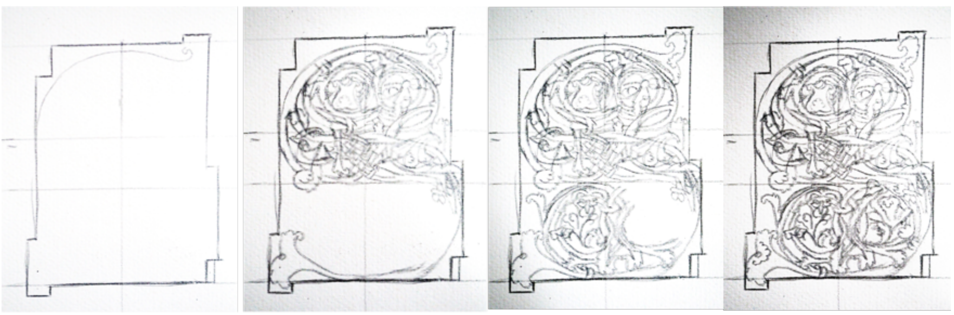

Figure 42 : Sequence of sketch of the initial from f.18v of Alc. 433 by freehand copy

3.2.2.2. Painting

After the illumination sketch on parchment[31] , colour paints were applied using the pigments and paints as close as the real manuscript. From the result of characterisation of the original paints, the red was painted with vermilion, the blue with lapis lazuli, and the ink was written with iron gall ink[32]. Yellow was found as background colour of the initial letter. From the materials characterisations on yellow paints found in f.18v, it is likely that the yellow was not made with inorganic yellow- but rather with an organic lake pigment. For this, recipe for producing

[30] Author (Shatila Fitri) is a freelance anime drawer (modern Japanese cartoon)- drawing portraits by looking at the real portrait photo. Freehand copy drawing with an original model may appear relatively easy for her point of view as 21st century drawer, that it will be very reasonable if the illuminators would sketch the initial with freehand copy drawing.

[31] The historical reconstruction was tried on papers before finally applied to parchments. The sequence step is explained on the Appendix.

[32] The characterisation of the pigments and ink are presented in Chapter V

colour was researched in medieval colour treatises, such as De Arte Illuminandi (14th century) and The Bolognese manuscript (15th century) where recipes to produce saffron, turmeric, and weld were found to be the most suitable to produce yellow lake pigments[33].

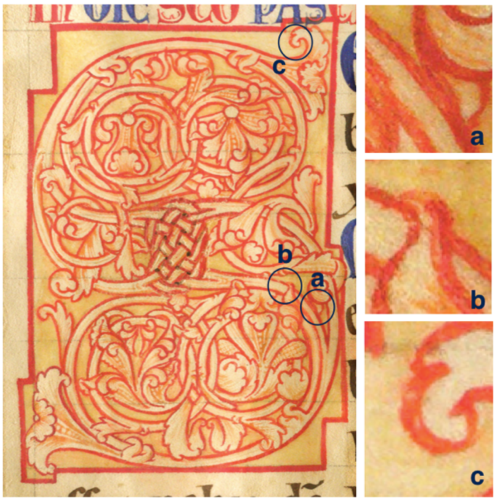

Figure 43 : Several spots in the initial letter of f.18v where the red paints were detached, leaving underlayer yellow lake pigment. © Biblioteca Nacional de Portugal.

The historical reconstruction results were painted with different yellow lake pigments for each side to observe the closest possible yellow lake pigment used. Yellow was painted first using a brush, then followed by the red on the initial with wooden stick, so it will appear consistent through the sketch, and finally red and blue paint were applied in *incipit* using male peacock quill[34]. The sequence steps of painting followed the observation towards the real manuscript, such as the trace of lead-point as well as magnified images. Nevertheless, there was some doubts about which was painted first- yellow or red. Based on painting attempts and on the observation of several magnified images, it can be seen that red colour on several points of were detached from the support, leaving visible the underlayer yellow lake pigments in different defined section at the initial letter (*Figure 43*), so that it was decided to paint the yellow, then the red.

Since yellow paints were produced from turmeric, saffron, and weld, in order to observe the difference, three of them were painted in the same manuscript (*Figure 44)*. Therefore, it can be observed that some yellows were paler and/or more intense than the other.

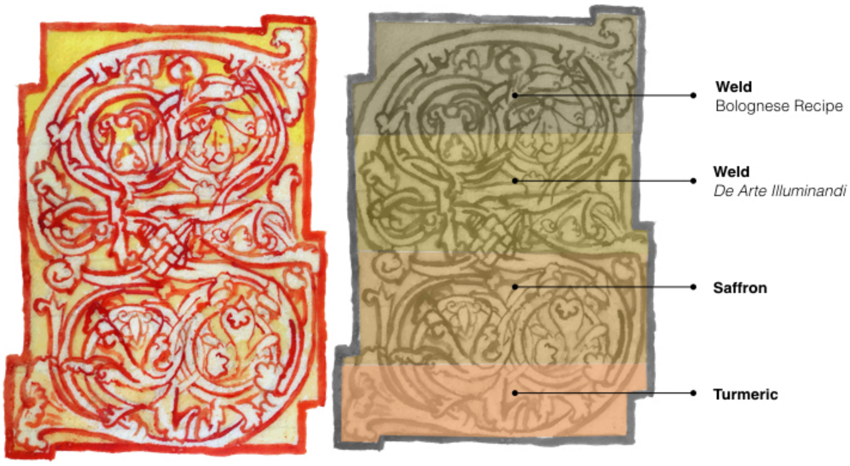

Figure 44 : The painted initial of historical reconstraction (left) and the scheme of yellow lake pigments painted in different areas (right).

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[33] The recipes are explained in Appendix IV.

[34] The specification of the red and blue paints is presented in Appendix V.

3.2.2.3. Writing Text

Figure 45 : The intense black appearance of the written text using iron gall ink.

The texts were written using male peacock quills[35] and iron gall inks[36], as referenced by previous researches on historical reconstruction of illuminated manuscripts. The nib was cut quite thin for producing small letters (Johnston, 1945). The writing ink was very bright when it was written on the parchment, and it transformed to be darker after around 30 minutes. The black colour appeared very intense day by day. As stated before, the writing process was done before the painting, therefore, the colour of the iron gall ink already oxidised well and turned into intense black. The result of the writing intense appearance is presented in *Figure 45*.

The texts were written right after the tracing and before the painting, so that the iron gall ink appeared more intense or well-oxidised. This step was taken also by the observation on the unfinished initial letters that was presented in Chapter III: 3.2.1 (see *Figure 35*) that obviously prove that the initial was painted after the text.

3.2.2.4. The Result of Historical Reconstruction

After carefully well sketched, from 4 methods of tracing, it is found that the stencil method was the most convenient, fastest, and suitable method used for f.18v of Alc. 433. However, freehand tracing method was also reasonable, yet the comparison between both highlighted that stencil tracing method resulted more accurate and precise model of the initial letter, particularly the details of the ornament. The comparison of the final result of both is presented in *Figure 46*.

Figure 46 : Comparison of final result of the historical reconstruction: sketching by freehad tracing method (left) and stencil tracing method (right).

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[35] The use of quill is based on “The Illumination Manual of Missals and Manuscripts”, known to be favoured in Western Europe, during 12th until 16th century, rather than the use of steel and iron stick which was stated to be ‘inconvenient’ to use for writing the text and calligraphy (Bradley, 1879).

[36] Iron gall ink has been used since the 4th century by scribes to write manuscripts (Aceto & Cala, 2017), and it was produced using gallnuts, homemade wine, an iron salt, and Arabic gum (Merrifield, 1999).

Part II

THE COMPENDIUM

Historical Records of the Materials of Alc. 433

Chapter IV: Materials

4.1. Pigments of Illumination

> *"There is no difficulty in seeing how other colours are probably composed. But he who should attempt to test the truth of this by experiment, would forget the difference of the human and divine nature. God only is able to compound and resolve substances; such experiments are impossible to man.*
>
> *These are the elements of necessity which the Creator received in the world of generation when he made the all-sufficient and perfect creature, using the secondary causes as his ministers, but himself fashioning the good in all things. For there are two sorts of causes, the one divine, the other necessary; and we should seek to discover the divine above all, and, for their sake, the necessary, because without them the higher cannot be attained by us." (Plato, cited from Timeaus)*[37]

Regarding the purpose and content of illumination, colour is the most essential material used in illuminated manuscript, and the matter of the light of the text as the word of "illumination" literally connects to light[38] (scientifically colour is the only "light" visible to human eyes). Plato, one of most celebrated ancient Greek philosophers, in his notable dialogue *Timeaus*, mentioned the connection of the process of sight resulting different colour. Additionally, the famous Aristotelian colour theory, *Perspectiva*[39], which led to the Neoplatonic metaphysics and aesthetic within medieval epistemology, which makes colour indispensable for human cognition and brought within the domain of the new discipline of optics (Panayotova, 2016). Linked to the illumination, colour is used to create the impression of the magnificent illustration on the manuscript, that appeared as it is being shined by light, symbolising the spiritual devotion. Note that medieval illuminated manuscripts were produced under specific orientations concerning the churches and religious orders, and other orientation, such as Alc. 433 produced in the Alcobaça scriptorium within Alcobaça monastery of Portuguese Cistercian Order. At the same time, the medieval churches, namely cathedrals and monasteries were the centre of intellectualism, culture and religion, on which these illuminations appeared as the way to illuminate. Most importantly, the illumination and the colour make the illuminated manuscripts classified as art (using modern criteria), and not merely just text.

In medieval world, everything in art must embodies a meaningful significance, including colour. Each colour rarely represented itself as explicitly, but rather to its philosophical or symbolic meaning. Somehow, it also encodes complex message of the artist and illuminator. The colour perception might not only reveal the aesthetic purposes, but also the moral, religious, and social paradigms (Panayotova, 2016). The colour meaning in religious matter is closely allied to the liturgical usage.

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[37] *Timeaus* or Τίμαιος in greek, is Plato's dialog, written around 360 BC, containing the philosophical thinking of correlation between nature and divine

[38] According to Oxford Dictionary, illumination is described as light, derived from Latin of verb *'illuminare'*

[39] *Perspectiva* is emerged from 13th century from commentaries on Aristotle's works on natural philosophy (Panayatova, 2016)

Colour also led into the impression and exclusiveness, knowing that the materials of the colours (pigments and/or lake pigments), were considered precious, some were often rare, expensive and unreachable by common people. In fact, several pigments are still considered highly precious until today. This reflects not only to the capability of the trade in medieval period, but also to the relation of the monasteries and/or the churches with outside world, or even the kingdom or religious orders in international scale. This means the availability of pigments in each period was influence by the situation of the country or kingdom itself. If the politics condition did not allow the possible international commerce, the availability of certain rare pigments must have been threatened.

Scientific analysis of illuminated manuscript can be done, also because of the existence of colour, through specific techniques. In materials science, colour can be identified and characterised by several techniques, leading to the provenance of the pigments. Not only that, the colour characterisation links to the production technique of pigment. The method of pigment making, of course, is certainly not a simple matter. The medieval pigment production is a complicated process. The *Mappae Clavicula*[40], one of oldest pigment handbook which produced in the ninth century (Finlay, 2002), shows complicated mixtures of recipes for producing colours for illuminated manuscripts. The pigment production was considered an alchemy work at that time.

Regarding limited knowledge of science in medieval time, pigments were mostly produced from natural sources such as plants, bugs, or minerals which were processed in certain ways following ancient recipes, sometimes modified according to the pigment maker creativity to obtain desirable colour. The addition of certain unusual materials such as earwax, could be a method to improve the mechanical properties of the paints (Thompson, 1956). This is why colour bonds art and science, and why each other cannot be separated. Both studies- in colour science or history of art- includes multidisciplinary knowledges of each other. In fact, the study of colour and science for centuries led the new era of pigment production. The pocket colour palette that we can easily buy anywhere today is actually a product of centuries of research.

Illumination was created within *Ingenium, Intellectus* and *Ratio* (Thompson, 1956), that is why each medieval manuscript was decorated within carefully-designed styles and painted by meticulously produced pigments from its *scriptorium*. Each *scriptorium* has pigment trends that used in several periods. The pattern of the usage of the pigments can be studied from several manuscripts within different recipes for its production. Various kind of pigments were used for medieval manuscripts. Each colour has its own recipes. The composition of the pigment is not only important to be studied, but also the colour binder is essential because it might affect differently in final colour appearance of the paint. Binder, or binding, also serves to hold the pigment on the media, and to give certain effect of the colour (Thompson, 1956). The colour might be darkened, lightened, or glistened which makes different effect to the illumination. Despite limited source of scientific knowledge during medieval time, the colour

[40] *Mappae Clavicula* is a handbook of recipes of colours, known as alchemical procedures includes 200 titles for metallic makings, arranged by seven metals. Believed to be 4th century translation in Latin from Greek, it includes 95 recipes of volumes, at least 13 manuscripts. The second nucleus consists of recipe books known as *Compositiones lucenses* (CLT) which is devoted to various handicrafts such as glass colourings, skin dyes, painting pigments, metal inks, parchment dyes, clays, and mortars (Brun, 2017).

theory on composition, making, and mixing were noted in numerous recipe books during centuries, as seen in Table 2.

Table 2 : Medieval Pigment Recipes

Title	**Estimated year/period of production**	**Author**	**Important Notes**
Mappae Clavicula	9-10th century, expanded to 12th century	Unknown	Estimated to be originally written in Alexandria, Egypt
De Clarea	11th century	Unknown	
Schedula diversarum artisum	1100 and 1120	Theophilus	Technical data of arts and crafts in northern and central Europe
Breviloquium diversarum artium		Suggested to be Theophilus, although it is doubtful	
Livro de como se façen as cores	1262	Abraham ben Judah ibn Hayyim	Written in Portuguese, using Hebrew character
De coloribus naturalia excripta et collecta	13th-14th century		
Liber de coloribus illuminatorum sive pictorum	14th century	Unknown French author	
Manuscripts of Jehan Le Begue	1431	Only the preface and glossary were written by Jehan Le begue	
De coloribus faciendis	14th century	Pietro de Sancto Audemaro	
De diversis coloribus	1398-1411	Alcherius and Antonio di Compendio	
Experimenta de coloribus	1409-1410	Alcherius	Recipe I-88, compiled from book borrowed from Italian monk, Dionisio, and recipe 100-116 copied from a book of Giovanni a Modena, a painter
Miscellaneous Recipes	1431	Le Begue	Written in French
Tabula de vocabulis sinonimis		Le Begue	Written in Latin
Il Libro dell'arte	1390s	Cennino Cennini	Written in Italian
Segretti per colori	15th century	Unknown	Known as Bolognese Manuscript, written in Italian and Latin
The Strasbourg manuscript	15th century	Heinrich von Lübeck and Andreas von Colmar	
De arte illuminandi	14th century	Unknown	
Göttingen Model Book	15th century	Unknown	Written in old German
Ricepte daffare piu colori	1462	Ambrugio di Ser Pietro da Siena	

From several previous research, Portuguese medieval illumination colours- on which are included the Alcobaça manuscripts, are commonly produced by several basic colours such as red, blue, green, and yellow (Miguel, 2012). Portuguese monastic world shared between São Mamede do Lorvão, Santa Cruz de Coimbra, and Santa Maria de Alcobaça produced Romanesque illuminations. However, as a central of artistic protagonist, from these three *scriptoria*, Alcobaça *scriptorium* presented more consistent collections of colour palette (Melo et al, 2011). Alcobaça colour palette in the end of the 12th century was influenced by the local context (Melo et al, 2011). The list of the pigments of several common used colours in Portuguese medieval illumination that has been characterised from Alcobaça's manuscripts are listed on Table 3.

As mentioned before, the pigment investigation of Alc. 433 occurred across 5 different *folia* (representative of the 5 different periods and production), on which several different colours are present: red, blue, green and yellow. Since these *folia* were added in different years, it is hypothesised that same colour could have been produced with different pigments.

Table 3 : The Characterised Pigments of Alcobaça Manuscripts

Colour	Pigment Used	Manuscripts	Period of Production
Red	Vermillion	Alc. 238, Alc 249, Alc. 347, Alc. 360, Alc. 405, Alc. 412, Alc. 419, Alc. 426, Alc. 427, Alc. 433, Alc. 446 (Miguel, 2012) Alc. 247 (Cavero et al, 2016) Alc. 54 (Barreira et al, 2016)	12-13th Century Around 1200 14th Century
	Red Lead or Minium	Alc. 249, Alc. 358, Alc. 360, Alc. 402, Alc. 410, Alc. 419, Alc. 427, Alc. 433, Alc. 446 (Miguel, 2012) Alc. 247 (Cavero et al, 2016)	12-13th Century Around 1200
	Dark lac dye	Alc. 247 (Cavero et al, 2016) Alc. 419 (Castro et al, 2016)	Around 1200 12-13th Century
Blue	Lazurite or Lapis Lazuli	Alc. 238, Alc 249, Alc. 347, Alc. 360, Alc. 402, Alc. 405, Alc. 410, Alc. 412, Alc. 419, Alc. 421, Alc. 426, Alc. 427, Alc. 433, Alc. 446 (Miguel, 2012) Alc. 247 (Cavero et al, 2016)	12-13th Century Around 1200

	Indigo	Alc. 249, Alc. 427 (Miguel, 2012)	12-13th Century
	Azurite	Alc. 433 (Miguel, 2012) Alc. 54 (Barreira et al, 2016)	12-13th Century 14th Century
Yellow	Orpiment	Alc. 249, Alc. 410 (Miguel, 2012)	12-13th Century
	Ochre	Alc. 54 (Miguel, 2016)	14th Century
Green	Bottle Green or Copper Proteinate	Alc. 249 (Miguel, 2012) Alc. 247 (Cavero et al, 2016) Alc. 418 (Miguel, 2018)	12-13th Century Around 1200

4.1.1. Red

> *"That if you ground it every day for twenty years, the colour would still become finer and more handsome." (Cennino Cennini, on The Character of the Red called Vermilion Chapter XL on his book: Il Libro dell Arte)*[41]

It is known that red has been used in art since ancient time- one of the oldest proofs is the prehistoric cave painting in Altamira, Spain, dated between 22000 - 15000 BC, painted by red ochre (Álvarez-Fernández, 2010). Its abundance in nature with high iron content makes it as the earliest colour used in art. In western medieval illuminated manuscript, red is almost always present. Particularly, with the fact that high quality of cinnabar as red pigment has been widely known to be produced in Almadèn, Spain (Miguel et al, 2014), it could have been easy to afford red as most used colour paint. Furthermore, previous research proved that some samples from Portuguese illuminated manuscripts from Sta. Cruz and Alcobaça *scriptorium* were possibly painted red paints produced with minerals cinnabar, not by artificial cinnabar (Miguel et al, 2014).

[41] *Il Libro dell Arte* or The Craftman's Handbook is Cennino Cennini's guide book on recipes of colour making, written in circa 1400 Century and has been source of the studies of European paintings (Burns, 2011).

According to legend, red was considered as the illumination itself as it is the definite alchemical colour of the ingredients (mercury and sulphur) related to the metals' theoretical twin principles (based of Francis Bacon[42]) led to the Philosopher's stone- the red elixir, as known as transforming materials (Gage, 1999). In illuminated manuscripts, as well as medieval painting, red evokes flower's petals, Christ's Wounds, lover's lips, dawn sky, flames of hell, and the power of the Holy spirit (Panayotova, 2016).

In western medieval illumination, the red pigment was known to be popularised after the 8th Century after the establishment of *Compositiones ad tingenda musiva pellles et alia*, recipes for colouring mosaics (Thompson, 1956). Even though the existence of this handbook means that the use of red pigment in western illumination already started before 8th century. The Romans was known to extract the red pigments (cinnabar) from Almadén, Spain (Melniciuc-Puicā, 2013), and as mentioned before, the red paints used in some Portuguese manuscripts was produced from Almadén as well (Miguel et al, 2014).

As a very attentive-looking colour, red is commonly used for incipit, explicit, and initial letters. The pigment used may be sourced from organic and inorganic materials (minerals). The pigment production was referenced according to several medieval colour recipe books.

4.1.1.1. Vermilion (Mercury Sulfide)

Vermilion[43] was characterised in several Alcobaça's illuminated manuscripts as mentioned previously. Vermilion red is an important colour for European medieval colour and displays very intense red with good conservation condition (Matos & Alfonso, 2014). Interestingly, vermilion costed as high as gold until 11th century which made this pigment was restricted to be used in *scriptorium* (Miguel, 2012).

The production of vermilion as red colour for painting (and respectively illumination) in Western illumination is first known from 8th century the *Compositiones ad tingenda*[44] which describes the recipe of vermilion by heating mercury with sulphur (Kroustalis & Bruquetas, 2014). Most known recipe of vermillion in medieval treatises is the wet process written in the Judeo-Portuguese book on the art of illumination, attributed to the 14th century (Miguel et al, 2014; Melo & Miguel, 2010).

[42] Francis Bacon (1561-1621) was a natural renaissance philosopher. He is well-known for his treaties on empiricist natural philosophy (The Advancement of Learning, Novum Organum Scientiarum)

[43] Vermilion is the standard name given to the red pigment based on artificially made mercuric sulfide. Mineralogists and crystallographer have given the common red crystalline name cinnabar, which is reserved for the natural mineral and its specific crystal structure. Three kinds of mercuric sulfide pigment: (1) natural, finely ground cinnabar; (2) synthetic, made by dry process- commonly called by vermillion; and (3) synthetic, made by wet process- also called by vermillion (Gettens et al, 1972).

[44] Lucca, Biblioteca Capitolare, Cod. 490, f.223v

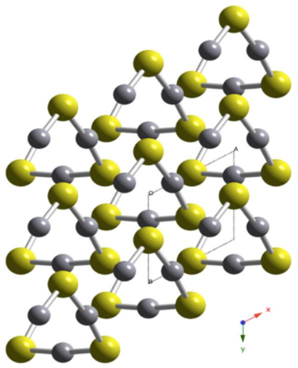

Figure 47 : Crystal structure of alpha polymorph of mercury sulfide, found in nature as mineral cinnabar; grey corresponds to mercury (Hg) and yellow to sulphur (S). (Pöttgen et al, 2016).

Based on the crystalline form of mercury sulphide, vermilion can be distinguished as α-HgS (Gettens et al, 1972) that has octahedral coordination (Miguel et al, 2014) as illustrated in *Figure 47*. Blackening of HgS may occur due to natural degradation phenomena (direct sunlight, humidity, or mechanical stress), that has been observed on raw HgS materials as well as on vermilion paints in artworks as a result of continuous process of amorphisation which started at the crystal surface in contact with surroundings rather than a congruent transformation to metacinnabar (Melniciuc-Puică, 2013).

In illuminated manuscript, vermilion red can be characterised by several in-situ techniques such as h-XRF, Hyperspectral Images, and UV-Vis-NIR FORS. Not only safe, fast, quick, and portable, the using of these techniques relatively result in comparable outcomes with almost no invasiveness to the manuscripts.

Vermilion gives a vary of colour of red-sheds, from dark into reddish-orangish hues, which can be characterised in spectrophotometry. Brighter red hue is absorbed strongly at wavelength about 560 nm (Feller, 1967). These spectra can be characterised through FORS and hyperspectral image very clearly, also with different binder to study the influence of binders to the hues of vermilion.

Through h-XRF, Hg is the key element which will be distinguished on $L\alpha_1$= 9.989 keV ; $L\beta_1$ = 11.824 keV; and Sulphur on $K\alpha_1$= 2.309 keV; $K\beta_1$= 2.465 keV (based on Bruker's h-XRF Elemental Database).

4.1.1.2. Minium (Red Lead)

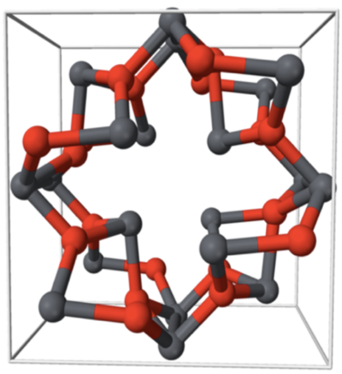

Figure 48 : Figure 58 Tetragonal unit cell of lead (II, IV) oxide (Pb3O4)which grey corresponds to lead and read to oxygen.
© Ben, Chemistry Department of University of Bristol.

Minium appeared in the late 10th Cenutry in medieval western Europe which believed to be sourced from the texts of Greek and Roman book of decoration (Melniciuc-Puică, 2013). As same as vermilion, minium is an inorganic red pigment that abundantly founded in illuminated manuscript. The colour is bright, intense in orange hues, making very suitable in illumination art. Minium pigment is Pb_3O_4 (*Figure 48*) that the manufacturing process was described originally in first century BC by Vitruvius in his manuscript *De Architectura* (Aze, et al, 2008), although in middle ages, the production of minium was obtained from lead oxide heating resulting different overtones of colour (Melniciuc-Puică, 2013). In Portuguese Illuminated Manuscript, minium was also widely used to provide intense oranges. Like vermilion, minium can be identified through elemental characterisation of with Pb as the key element, which can be distinguished on $L\alpha_1$= 10.551 keV; $L\beta_1$ = 12.614 keV; (based on Bruker's h-XRF Elemental Database) as well as the inflection point in FORS at appoxiametely 560 nm (Aceto, 2014).

4.1.2. Blue

> "*Saphirus tante virtuis est quod gemma gemmarum vocatur, colorem. Colorem habet firmament quando sine nube est." (The Aberdeen Bestiary, folio 101v)*[45]

Often described as blue sapphire, blue is associated to divine that from 12th Century was considered to be most suitable colour for the Virgin Mary (Panayotova et al, 2016) and was used as the medieval Christian counterpart of the ancient pagan purple (Jacobs & Jacobs, 1958). Unlike red, blue is very rare to be found in nature, thus blue was relatively precious colour and synthetic blue was significantly produced across art history. The first synthetic blue was the Egyptian blue, which assumed to be produced around 2600 BC (Collins et al, 2004). It was appeared on 8th century Godescalc Gospels as well as manuscript in 10th-12th century Medieval England and disappeared from artists' palettes during early middle ages (Ricciardi & Beers, 2016). The quest of blue continued from time to time allowing people to extract the colour from minerals, such as lapis lazuli (ultramarine), and blue indigo. However, blues throughout medieval period were varied in availability and popularity throughout periods and regions (Panayotova & Ricciardi, 2016).

As seen in nature, blue was used significantly to paint the sky and as mentioned before- Virgin Mary, that usually painted on her robe (Jacobs & Jacobs, 1958; Pastoreau, 2016). In medieval ages which combined to the early Christian symbolism, blue symbolised the breastplate and the heavenly Jerusalem with magical properties of stones given in the more popular pagan lapidaries, the blue colour of sapphire, as the colour of heaven that contemplate the heavenly kingdom (Jacobs & Jacobs, 1958; Pastoreau, 2016).

Among several Alcobaça illuminated manuscripts that have been characterised, it is known that blue pigments were painted by with lapiz lazuli, azurite, and indigo (as mentioned in Table 4).

4.1.2.1. Lapis Lazuli (Ultramarine)

Believed to be one of most expensive pigments, lapis lazuli has been well-known as the precious and rare pigments ever trade. The nickname 'Ultramarine'[46] is literally across the sea, intended to its original provenance in Afghanistan which could cost as much as gold (Bucklow, 2016). Lapis lazuli was transported for centuries, carried west by Arab merchants who bought them in India and transported it first by sea to Hormuz, in Persia, then by long chain of caravans, to the Levantine commercial ports of Mediterranian: from Baghdad to Alexandria, Cyprus, or

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[45] *Sapphire is of such virtue that it is called the gemstone of gemstones. In colour, it is like the sky when it is cloudless.*

[46] The term of ultramarine was originally used adjectively for example Azzurro oltramarino, nevertheless it serves to distinguish the genuine lapis lazuli (Plesters, 1966). Lapis lazui itself originally is genitive form of medieval latin *lazulum* taken from Arabic lazaward لازورد that etymologically used as the colour itself until present day including Portuguese and Spanish *azul* (Senning, 2006).

Rhodes, and finally were sold to Italian maritime merchants of Amalfi, Pisa, Genoa, and Venice- who eventually carried them west and north to Europe (Delamare & Guineau, 2000).

Since lapis lazuli blue pigment was extracted from natural lapis lazuli, it always contains non-blue materials such as calcite and iron pyrite (Thompson, 1956). There was no record of lapis lazuli in colour recipes before early 13th century, nevertheless close to 14th century, Cennino Cennini wrote about it in his book (Thompson, 1956).

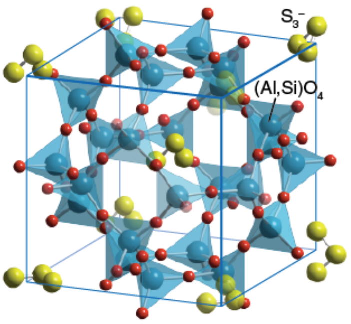

Figure 49 : Depiction of ultramarine. © ChemTube 3D

Lapis lazuli's most important mineral is lazurite, a complex sulphur-containing sodium aluminium silicate $(Na,Ca)_8(AlSiO_4)_6)S,SO_4,Cl)_{1-2}$ with the blue S_3^- radical ion as chromophore, incorporated in sodalite cage structure of $[Al6Si_6O_{24}]^{6-}$ (Berke, 2007) as seen in *Figure 49*.

The blue of lapis lazuli pigment used in illuminations can be characterised through h-XRF, with Na, Al, Si, and S as key elements. UV-Vis-NIR FORS analysis allows to identify its characteristic reflectance band at 475 nm, a shoulder at 755 nm and the maximum absorption band at ca. 600 nm due to the change transfer between the sulphur atoms of S_3^- present in the lattice at the aluminium silicate complex, however the pigment does not present any characteristic absorption bands in the NIR region (Vetter & Schreiner, 2014).

4.1.2.2. Azurite

Azurite[47] is a copper-based mineral, which was called "Armenian Stone" when Armenian and Spain were once the source of supply (Thompson, 1956). Azurite may have been used as paint pigment as early as Egyptian Blue, and it is one of the most important blue in Medieval European paintings (Vink, 1986). Even though, lapis lazuli has been known to be one of the most precious and it was indeed the well-spread blue pigment in medieval Europe, as medieval painting improved, the blue palette used by painters and illuminators was replaced. Azurite then widely started to be recognised as major source of blue (Anderson, 1981) especially with its blue hues that close to greenish blue with its exceptional quality, also more affordable than lapis lazuli (Thompson, 1956). However, the choice between lapis lazuli and azurite was meaningful according to its hierarchy of decoration and/or symbolic values, or sometimes they were combined in the same blue area in an expedient manner, with ultramarine painted in thin layers over azurite (Ricciardi & Beers, 2016).

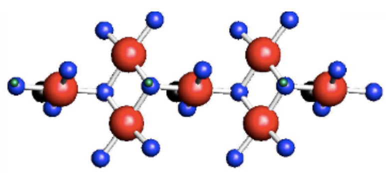

Figure 50 : Azurite structure which red corresponds to Cu, black to C, blue to O and green to H (Rule et al, 2011).

Azurite $(Cu_3(CO_3)_2(OH)_2$ is a monoclinic crystal (Anthony, 1990) as depicted on *Figure 50*. Chemically, azurite is destroyed by heat breaking the carbon dioxide and water- forming copper (II) oxide (CuO) powder or cause

[47] The word "Azure" belonged originally to lapis lazuli, that sometimes azurite was called by citramarine azur to differ it with lapis lazuli since azurite was found in Europe (East France and Hungary)- unlike lapis lazuli (Thompson, 1956).

conversion to malachite ($CuCO_3.Cu(OH)_2$) and paratacamite/atacamite ($Cu_2Cl(OH)_3$) (Coccato et al, 2017) Cu is the key element for the h-XRF characterisation, appeared on $K\alpha_1 = 8.046$ keV ; $K\beta_1 = 8.904$ keV; (based on Bruker's XRF Elemental Database). With the UV-Vis-NIR characterisation, azurite exhibits the maximum absorption around 640 nm (Aceto, 2014) and specific absorpsion bands in NIR region around 1493, 2284, and 2350 nm (Ricciardi & Pallipurath, 2016).

4.1.3. Green

> "*Grene colour is the most liking to the sight for cmynge togyderes of fiery parts and of earth. For briythnesse of fuyre that is in grene is pleseth the sight." (Bartholomeus)*[48]

Green is mostly used to paint plants and foliated ornaments in most illuminated manuscript. Unlike other colours, green is remarkably constant in terms its symbolic resonance*: viriditas* or greenness signalled new life, the Resurrection (Jackson, 2016; Pastoreau, 2019). In Portuguese illuminated manuscript, it appeared remarkably as in initial letters and some foliated ornaments in figures and can be distinguished easily. Its appearances were not as many as red and blue- and somehow that what makes green is more substantially 'seen'.

From the point of view of chemical analysis, green can be considered as a difficult colour. Illuminators used both synthetic and natural pigments, organic and inorganic, as well as mixtures of yellow and blue, to obtain various shades of green, and some presented significant deterioration issues (Ricciardi et al, 2013).

4.1.3.1. Verdigris: The Bottle Green (Copper Proteinate)

Bottle green appeared distinctively in Portuguese illuminated manuscript. Its intense and green colour is remarkable- somehow more than blue and red. Bottle green is a Verdigris[49] based pigment- that has been known as ancient and used in enormous extent in medieval times. In illuminated manuscripts, Verdigris was often used in initial letters, alternating with vermillion letters, or letters with vermillion and azurite. Verdigris was formed by result of corrosion of copper when exposed to air, moisture, and acid- it is unstable (Ricciardi & Beers, 2016). As the acetate form, Verdigris was made from a plate of copper exposed to hot vapour of hot vinegar or urine[50] in a closed

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[48] Interpretation of colour of Pseudo-Aristotle's Bartholomesus in John of Trevisa's version (Gage, 1999).

[49] The term "Verdigris" describes a wide range of synthetic copper-based greens of variable compositions (Ricciardi & Beers, 2016)

[50] Urine is mentioned in recipes for verdigris preparation, stated that the principal of urine are urea, uric acid, and creatinine, along with various amounts of sodium, potassium, calcium, magnesium, chloride, bicarbonate, sulphate, and phosphate ions. This

environment (Gilbert et al, 2003). The instability might result in the change of colour during ages, with the contact with air, or even with the parchment. A case was studied that Verdigris began to bleed the parchment of 8th Century Northumbrian Gospels and 13th century English Hours (Ricciardi & Beers, 2016). Cennino described Verdigris as "very lovely to the eye, but it does not last" (Cennini et al, 1932). Verdigris tends to become darker as it is oxidised and it mislead art historians (Thompson, 1956). To avoid this problem, some green synthetic pigments (still based on copper) were explored. Various recipes were begun to be developed, such as copper chloride (by coating the copper with honey and salt), copper phosphate (by coating the copper by soap), and copper proteinate (verdigris was reacted with egg yolk, resulting copper complex) (Gilbert et al, 2003). Bottle green or *verde garrafa*, has been researched meticulously to differ it with other Verdigris and other green pigments. Previous study of bottle green was conducted within the micro-samples of the bottle green paints taken from Alcobaça and Clairvaux manuscripts, showed its characteristics from alteration of the methyl and methylene groups of the protein binder and loss of cohesion of the paint (Miguel et al, 2018).

As a copper-based pigment, of course, the Cu acts as the key element for the h-XRF characterisation which exhibited at $K\alpha_1$= 8.046 keV, $K\beta_1$ = 8.904 keV; (based on Bruker's h-XRF Elemental Database). The characterisation of bottle green can be obtained by FTIR[51] and Uv-Vis-NIR FORS through the specific band of carbonate which can be quite complicated to analyse. The result can be complemented by hyperspectral image. To obtain a very accurate result, elemental analysis by sampling using Liquid-Gas Chromatography can be obtained- however, it is likely to be very complicated and difficult to obtain a sample from illuminated manuscript due to its precious value and possibility to damage the manuscript as well.

4.1.3.2. Malachite

Malachite ($CuCO_3.Cu(OH)_2$) is naturally occurring basic copper yields turquoise green and frequently used in 15th century France (Ricciardi & Beers, 2016). Native from azurite, malachite appears as beautiful green mineral that sometimes called by blue green[52] or *verde azzuro*- explaining its origin, as described in Cennini's Il Libro dell Arte (Thompson, 1956). Malachite was used since antiquity in Egypt on tomb paintings and although it is used in medieval paintings, it is rarely used and characterised within illuminated manuscript (Gettens & Fitzbugh, 1974). Similar to azurite, the characterisation can be performed through h-XRF with Cu as key element. However, to differ

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complex mixture will be capable of dissolving both verdigris and corroding copper to some extent, and is much more active as an ingredient that water alone, resulting in a very complex organic and inorganic salt (Scott et al, 2001).

[51] Characterisation by FTIR is not used in this research, however, FTIR can be used to characterise the molecular bonds of the copper-based pigments, to distinguish the molecules bonds to the copper; so that it is likely to be more accurate in identifying which copper-based pigment used.

[52] Malachite was also called by mountain green, gold solder, or chrysocolla, and *azurinum conversum in viridem colorem* (Blue in which has turned into green) (Johnson, 1956).

azurite and malachite, it is important to characterise within UV-Vis-NIR FORS to see the molecular bonds of carbonate and O-H, as well as in Hyperspectral image.

4.1.4. Yellow Lake Pigments

Apart from gold, of which appearance in illuminated manuscript is famously depicted in the word ‘illuminate’, yellow pigments also had significant uses in medieval illuminated manuscript, for example imitating the appearance of gold (Thompson, 1956). However, yellow was used to paint accordingly without overshadowing gold. Inorganic yellow pigments were used by illuminators, such as orpiment (As_2S_3), ochre (Fe_2O_A), or realgar (As_4S_4) (Thompson, 1956). Interestingly, organic lake pigments have also been used, usually sourced from local plants. Organic yellow pigments- commonly called by yellow lake pigments- were known to be extracted from *Reseda luteola*, *Crocus sativus* flower (Ricciardi & Beers, 2016), buckthorn, *Rhus cotinus* (Thompson, 1956), turmeric, onion skins (Bechtold & Mussak, 2009). However, yellow lake pigments are complicated to be analysed with in-situ and non-invasive techniques to have a very accurate result (Clarke, 2011). UV-Vis-NIR FORS characterisation and comparison by using a consistent database built up with historically accurate reproductions of references for colourants, binders, and colour paints will be very useful to validate the lake pigments identification and can be complemented by following a chemometric approach (Nabais et al, 2018).

The colour source- the chromophore, is extracted from its source, then precipitated into lake pigment to be, then, painted on the parchment. This process may include some procedures with specific purposes such as the addition of metallic ions known as mordants, which also can play an important role in the appearance of final colour (Melo, 2009).

4.1.4.1. Weld

Reseda luteola is the most used plant as the source of luteolin[53] and apigenin (Favaro, et al, 2007) and is considered as the most stable yellow, thus it was widely used for dyeing (Melo, 2009). From medieval period to the 19th century, weld supplied the textile industry and was cultivated in a large scale (Cardon, 2009). Metal ions were used as mordants to fix and stabilise the colour on the fibre; the metal salt was also obtained from different salt (Favaro et al, 2007). For weld, the use of alum mordant produces a yellow-orange colour, and copper to produce greenish yellow. However, the addition of chalk or hydrated alumina to precipitate the dye source was used for paintings and illuminated manuscripts (Favaro et al, 2007). In medieval recipes and colour treatises, weld earliest known recipe was written on *De art illuminandi* (14th century) which specifically mentions a yellow colorant prepared with *erba dei tintori* (dyers’ weed) (Pasqualetti, 2015).

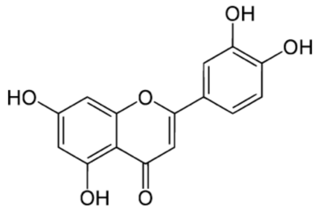

Figure 51 : Luteolin structure

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[53] Luteolin is a chromophore which appear in plants as glycosides. Luteolin is flavonoid yellow that contains of the flavonols, known as antioxidant properties (Melo, 2009).

4.1.4.2. Saffron

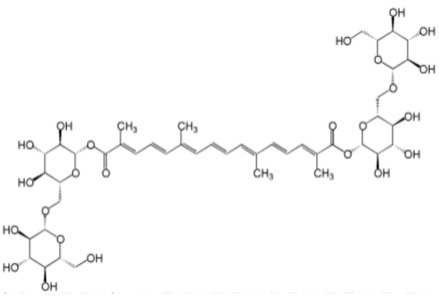

Figure 52 : Crocin structure

Saffron[54] is often found to be used as spice and colouring agent from ancient times as well as dye for wool and cotton since antiquity (Mousavi & Bathaie, 2011). In its composition, saffron contains crocetin and its glycosdic derivatives (the crocins[55]) as chromophore, which produces the yellow colour of carotenoids (Bathaie & Mousavi, 2010). In medieval Europe, saffron was imported. The highest value was that from Persia, around the 10th century, however it was eventually growing in Spain and Italy around 13th century (Bucklow, 2016), that it could have possibly grown in Portugal as well. The yellow colour that saffron produced has been widely known to imitate gold. Saffron was mentioned in 15th century Sloane MS 1698 and 14th century manuscript in Montpellier, however, its lack of durability makes saffron somehow excluded from illuminators palette (Johnson, 1956).

4.1.4.3. Turmeric

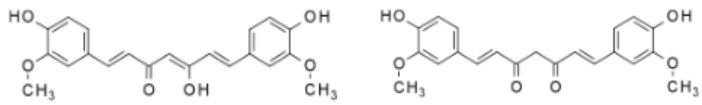

Figure 53 : Structures of curcumin in enol form (left) and keto form (right).

As saffron, turmeric has been widely used for colouring agents across Asia and Middle East. The component that responsible for the yellow pigment is curcumin [1,7-bis-(4-hydroxy-3-methoxyphenyl)-1,6-heptadien-3,5-dione] and 2 curcumnoids, demethoxycurcumin, and bisdemethoxycurcumin (Kanhathaisong et al, 2011). As same as saffron, the problem of turmeric is its poor pastness in light, that can be avoid by metal complex formation or mordanting (Kanhathaisong et al, 2011). In illumination manuscript, the use of turmeric is sometimes referred as the yellow colour source in Islamic manuscripts production (Aceto, et al, 2018).

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[54] Saffron is derived from the Arab word of yellow, a name reflecting the high concentration of carotenoid pigment present (Melnyk, et al, 2010)

[55] The yellow crocins include mono- and diglycosyl esters of the dicarboxylic acid, crocetin. The nonionic crocin-1, or α -crocin ($C_{44}H_{64}O_{24}$; molecular weight, 977), is large, hydrophilic and constitutes the major reddish yellow crocin component of saffronCrocin-1 is readily soluble in warm water and produces a yellow-orange solution, meanwhile Crocetin ($C_{20}H_{24}O_4$; molecular weight 328) is another carotenoid that is isolated from saffron; it is one of two principal chemicals responsible for the red saffron colour (Bathaie et al, 2014).

4.1.5. Black Writing Ink: Iron Gall Inks

Although black is not delightful to the eyes as other 'visible' colour to create illumination, black has the most important role in illuminated manuscript: the written words. Historical writing with quill appears as calligraphic writing with variability of trace width caused by physical properties of the writing instrument that can be studied through quantitative palaeography of the ink trace (Brink et al, 2012). In medieval times, three kinds of black inks could be used: suspension of carbon, suspension of a black organic salt of iron mixed with other salts in solution (Thompson, 1956), or known as iron gall inks, or a mixture of both these inks. Numerous varieties of recipes of iron gall inks have been passed down, but they share the common colour-forming of gallic acid tannins and iron ions (Kolar et al, 2003).

The main inorganic compound of iron gall ink, vitriol, was obtained from different mines and by various techniques. This iron sulphate source was contaminated by other metals, such as copper (Cu), aluminium (Al), zinc (Zn), manganese (Mn) and lead (Pb), which do not contribute to its colour formation (Hahn et al, 2004). For this, the contamination of these elements in the preparation of the inks was consistent.

To study the influence of the different composition of iron gall inks, its characterisation plays significant roles. Determining the elemental composition of iron gall ink can be inferred on the use of different composition (Tiburio et al, 2020) and its changes during centuries, using score plots to separate the signals from the various component (Catelli et al, 2017). By observing different ratios of Fe h-XRF counts to other elements such as Cu, Zn, and Mn, we expect to see the different clusters according to the different periods of production of different *folia*.

4.1.6. Binder and Extender

Pigments are not complete without a binder to make the colour more intensely appear, and easily bond to the parchment. Pigment and binder are mixed together to be applied on the parchment, and when the binder becomes dried, the pigment is safely hold to the parchment (Thompson, 1956). Several proteineceans binders were used for medieval illumination production, such as parchment glue, egg white (glair), egg yolk, or even earwax (Thompson, 1956). Some factors such as viscosity, media, transparency and quantity (Thompson, 1956) may be consideration of illuminators to obtain different hues, glossiness, and intensity of the colour.

Identifying the binder used with the pigment is one of the greatest challenges when it comes to the in-situ and non-invasive characterisation of the binder used to produce illumination. However, in-situ methods like UV-Vis-NIR FORS, with comparison of the spectra between real manuscript and historical reconstruction (colour reproduction), if it is done meticulously as accurate as possible may result significant accuracy.

4.1.6.1. Parchment Glue

As mentioned by Cennini, parchment glue was used in medieval Europe for paintings and illumination. Parchment glue is very sufficient for parchment illumination since it has the compatibility of 'repair' the parchment that works as filling since parchment is prone to degradation (Wouters, 2000), thus when an illuminator uses parchment glue as binder to the pigment, the very same collagen of the parchment will act as binder as well as filling to the parchment. As for this advantage, parchment glue was (and it is still) also used as coating material for conservation purposes.

4.1.6.2. Egg White or Glair

Clarified egg white, or glair, was also used for painting and manuscript illumination. It was the basic binding media for medieval illumination. The anonymous author of 11th century tract of Berne quoted it as *De clarea*, wrote the preparation of egg white as binder in different preparation (Thompson, 1956). Egg white consists of water and proteins (hydrophobic and hydrophilic amino acids) and strong beating denaturised and settled the protein (Kroustallis, 2011). Compare to egg yolk, glair was used and mentioned more often in colour treatises. Several manuscript of colour recipes such as *Livro de como se fazen as cores*, *De coloribus faciendis*, *Bolognese manuscript*, and *De coloribus naturalia* (Kroustallis, 2011) mentioned it.

4.1.6.3. Egg Yolk

The reference for using egg yolk (*vitello ovi*) as binder dates back to the early end on 11th century, on Bavarian Gospels, where it was assumed to be mixed with vermilion, and it is known that the use of egg yolk affects the red lead to have warm hues- yellow glow and high gloss (Panayotova & Ricciardi, 2016). The use of egg yolk had been evolved widely in medieval Europe, especially for the illumination of liturgical books (Panayotova & Ricciardi). Anonymus author of 12th century *Hi sunt omnes colores* also mentioned of the use of egg yolk as binder for illumination together with egg white and cherry gum (Kroustallis, 2011). The use of the egg yolk is very simple as well- it is only needed to be diluted in the water. However, the yellow of egg yolk may affect the colour.

The use of egg yolk as binder is known to result nearly permanent, durable, and almost waterproof (Thompson, 1956). Even though when the pigment is mixed with the egg yolk may appears dense and sticky resulting magnificent intense colour.

4.1.6.4. Other Additives Mixed with the Binder

Other additives that used as binder media also written in medieval colour treatises, such as honey, sugar, or earwax to obtain more adhesive binding media. Honey and sugar, for example, can avoid binders to be brittle, or earwax to avoid annoying bubles (Kroustallis, 2011).

4.1.6.5. Chalk and Gypsum

Chalk is relatively found as extender in a paint composition mixed with other pigment to improve the mechanical properties of the paints, increasing its plasticity and its covering effect (Thompson, 1956). Besides chalk, there are references for the use of lead white as extender, usually concerning organic lake pigments (Moura et al, 2007; Coccato et al, 2017). Additionally, in the matter of using chalk for lake pigments extender, the dye solution of lake pigments could be filtered over a chalk or gypsum stone as mentioned in the Judeo Portuguese illuminators manuscript for producing materials for illumination (Melo et al, 2018). Not only for organic lake pigments, the use of chalk was considered as catalyst as well as in the case of vermilion in Portuguese Lorvão manuscript (Miguel et al, 2009).

Chalk was also used to prepare the parchment for manuscript, known to be undergone in medieval period in mainly northern and southern Europe (Coccato et al, 2017). In Alcobaça's manuscripts, chalk and gypsum were characterised as extender and used as surface ground preparation, as the historical accurate reproduction of Alcobaça Beatus conducted (Miguel, 2012).

4.2. Structural Elements: Bookbinding

4.2.1. Parchment

Hamlet. "Is not parchment made of sheepskins?"
Horatio. "Ay, my lord, and of calves' skins too."
(Hamlet's Shakespeare, 1599-1602)

Parchment is the main materials support used as writing support in medieval Europe before paper- which is relatively cheaper and easier to produce. Basically, parchment is a membranous substrate produced from animal's skin (Bicchieri et al, 2011). Usually made from calf, sheep, or goat skin, parchment analysis requires DNA analysis or proteomics in order to figure out the origin of the animals (van der Werf et al, 2017).

4.2.2. Covering Leather

Medieval manuscript was sewn and band into a to leather, major covering materials- except for bind bookbinding, to protect the parchments. The leather was sometimes made from vegetable-tanned calf, chamois wool sheep, or alum-tawed pigskin (Szirmai, 2017). However, to distinct each leather, usually the authorities catalogued them by colour- such as for 18th century binding, it is described by *cum tabulis nudis, sine tabulis, or in carta pecudina* (Szirmai, 2017). Furthermore, specifically for gothic bindings, the covering leather was embellished and decorated by stamps and gold.

Part III

THE INVESTIGATION

Materials Investigations of Alc. 433

Chapter V: Materials Characterisation

5.1. Alc. 433 Materials Characterisation

"Et ipsa scientia potestas est." [56]
(Francis Bacon, from Meditationes Sacrae)

The characterisation of the drawing technique (lead point trace) of the illumination technique and of the materials used to produce the colour paints of Alc. 433, such as pigments and binders, and the writing ink started by a general observation (photography and digital microscopy), followed by elemental analysis (h-XRF) and molecular analysis (hyperspectral image and UV-Vis-NIR FORS). The misson to the BNP for the analysis of Alc. 433 was performed by Prof. Catarina Miguel, Silvia Bottura and Manuel Plácido[57]. All raw data were interpreted, analysed and discussed by the author.

5.1.1. Colour Paints Analysis

As stated in Chapter I, four *folia* were chosen as representative of the *folia* of the four different periods of production identified towards the liturgical studies: f.1; f.15; f18v, and f.228 (*Figure 54*).

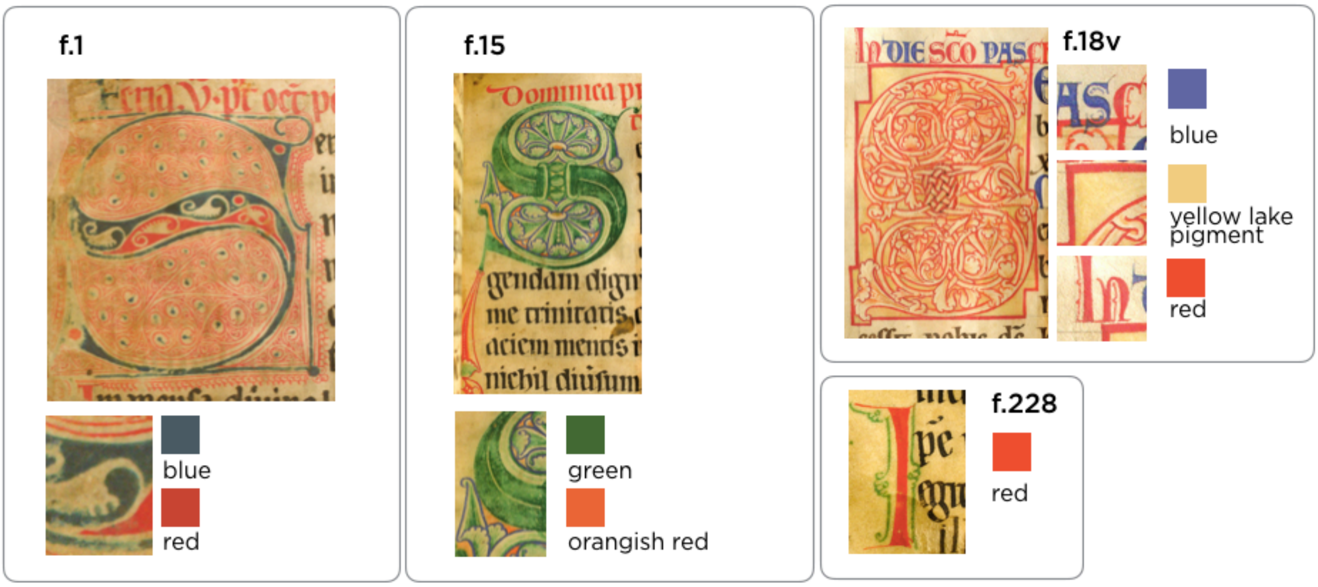


Figure 54 : Characterised colours from each folio of Alc. 433

[56] "Knowledge is Power"

[57] The author was meant to analyse in BNP as well, but due to the COVID-19 Pandemic Restriction, the plan had to be postponed, following the rules of the authorities of library and Portuguese Government.

5.1.1.1. Blue: Lapis Lazuli or Azurite?

Figure 55 : Alc. 433 f.1 (left) and f.18v (right) © Silvia Bottura - Biblioteca Nacional de Portugal.

<u>General Observation</u>

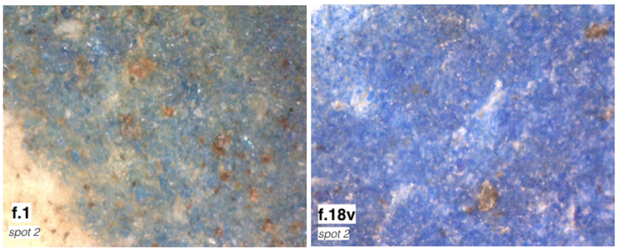


Figure 56 : Magnified images of the blues of f.1 from spot 1 (left) and f.18v from spot 12 with magnification 430x, indicated in Appendix II.

From the selected *folia* representative of 4 different periods of production, blue paints are found in f.1 (ornament) and f.18v (incipit), as shown in *Figure 55*. From the direct investigation of both blue paints, f.18v blue paints look glossier and brighter than the f.1 blue paint. The crack is obvious on f.18v blue, yet it slightly looks less deteriorated than f.1 (*Figure 56*). Under magnified observation, the blue paint present in f.1 presented higher heterogeneity, with blue paint dispersed in a matrix of brownish and white particles. Regarding the blue paint present in f.18v, the intensity of the blue colour is considerably higher than distribution, when compared with the blue paint of f.1 (*Figure 56*).

Materials Characterisation

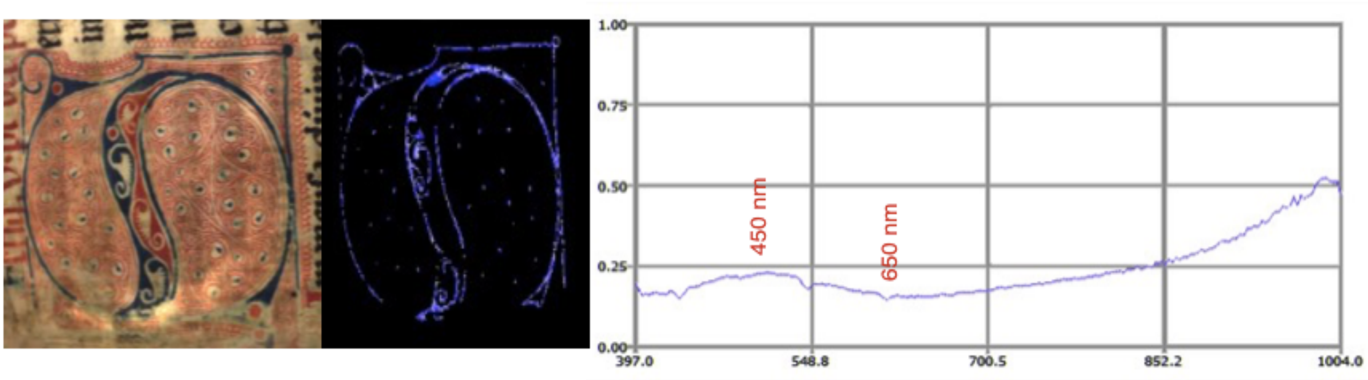


Figure 57 : Left, region of interest (ROI) of f.1 analysed by hyperspectral analysis; centre, map of distribution of the UV-Vis spectra of the blue paint present in the ROI; right, UV-Vis spectrum of the blue paint, where it is identified the characteristic bands

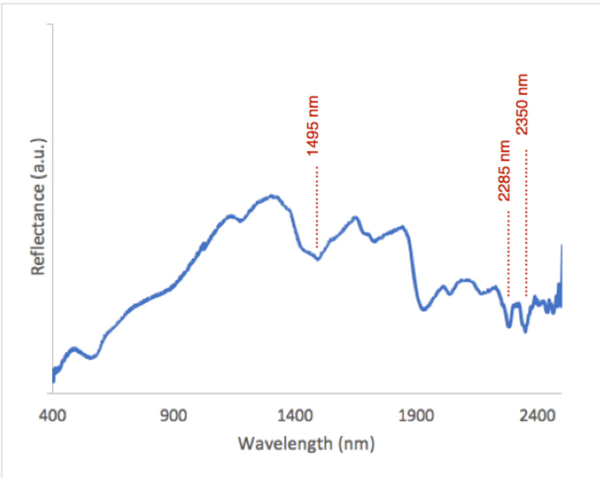


Figure 58 : UV-Vis-NIR spectra of f.1 blue paint.

UV-Vis FORS analysis of the blue paint of the initial letter in f.1 allowed to identify the characteristic spectral profile of azurite ($2CuCO_3.Cu(OH)_2$), namely its characteristic reflectance band with a maximum at 450 nm and a maximum absorption band at ca. 640 nm related to the d- d transitions of copper (*Figure 57*). UV-Vis-NIR FORS analysis of this blue paint allowed to identify, besides the characteristic profile of azurite in the UV-Vis region previously presents, two specific vibrational overtones of the hydroxyl groups (-OH) of azurite at 1495, 2285, and 2350 nm are present (Ricciardi et al, 2009), shown in *Figure 58*.

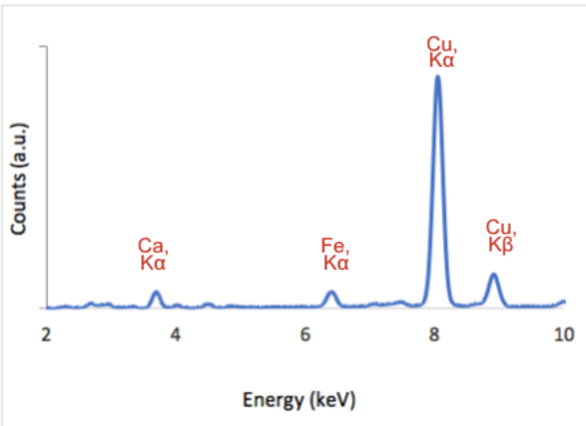


Figure 59 : h-XRF spectrum of f.1 blue paint.

h-XRF elemental point analysis revealed the presence, as major elements, of copper (Cu), iron (Fe) and calcium (Ca) (*Figure 59*). The presence of copper corroborates the presence of azurite, whereas iron might be related to the presence of pyrite (FeS_2) and calcium to the presence of calcite ($CaCO_3$), commonly associated to the mineral azurite (Eastaugh, 2014).

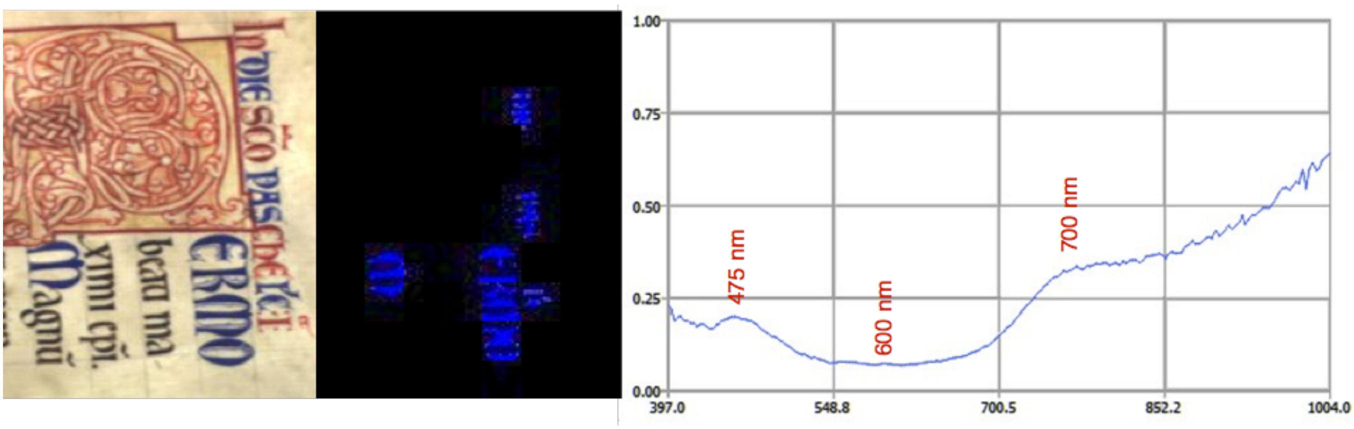


Figure 60 : Left, region of interest (ROI) of f.18v analysed by hyperspectral analysis; centre, map of distribution of the UV-Vis spectra of the blue paint present in the ROI; right, UV-Vis spectrum of the blue paint, where it is identified the characteristic bands.

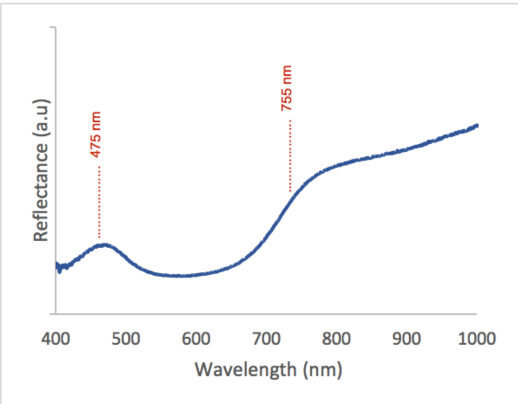


Figure 61 : UV-Vis-NIR FORS spectra of f.18v blue paint.

Regarding the blue paints present in the *incipit* of f.18v, FORS analysis allowed to identify the characteristic UV–Vis spectrum of lapis-lazuli ($Na_{6-10}Al_6Si_6O_{24}S_{2-4}$), namely the reflectance band at 475 nm, the shoulder at 755 nm and the maximum absorption band at ca. 600 nm (*Figure 60 and 61*) due charge transfer transition inside S_3^{2-} group that is present in the lattice of the complex aluminosilicate (Bacci et al, 2009).

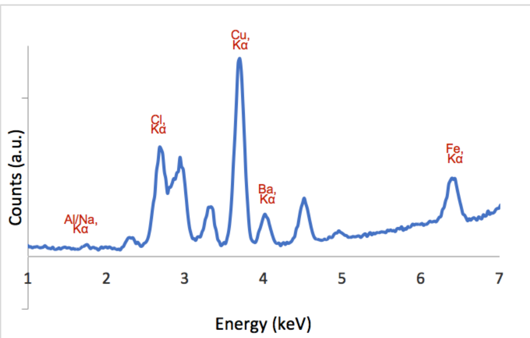


Figure 62 : h-XRF spectrum of f.18v blue paint.

h-EDXRF elemental point analysis of the blue paint used for the incipit in f.18v revealed the presence, as major elements, sodium (Na), aluminium (Al), silicon (Si), sulphur (S), chlorine (Cl) and barium (Ba), often present in lapis lazuli minerals (*Figure 62*). Likewise to azurite, calcium (Ca) and iron (Fe) might be related to the presence of pyrite and calcite, which can be present as minor impurities. Worth to stress that Ca may also present due to $CaCO_3$ that used to prepare parchment (Fachechi & Bracci, 2019) or that could be extender in the paint formulation.

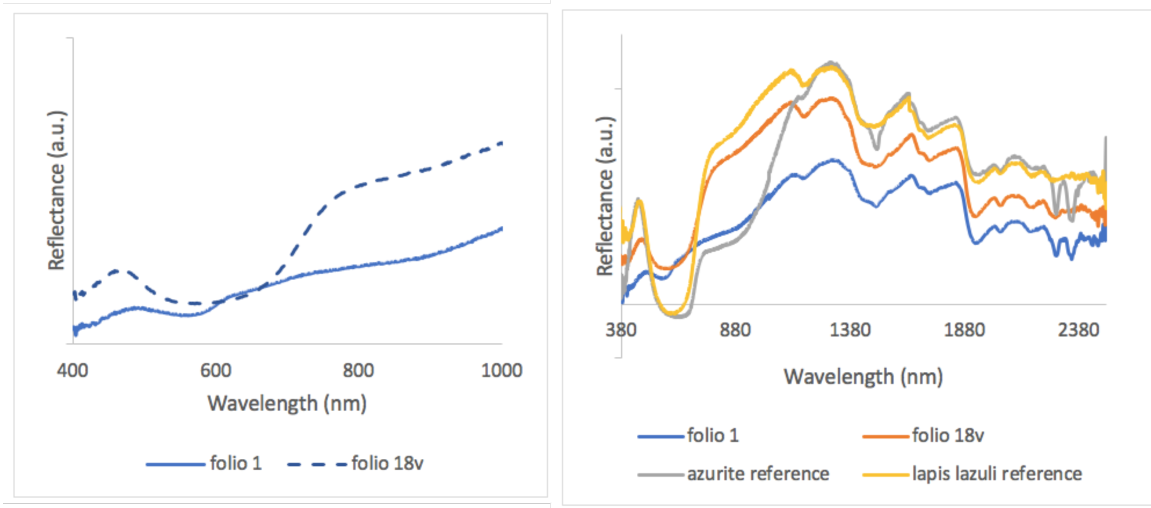


Figure 63 : The comparison between blue paints of f.1 and f.18: UV-Vis-NIR FORS spectra (left); with the spectra of azurite and lapis lazuli reference (right).

As for the comparation between f.1 and f.18v, both UV-Vis-NIR FORS spectra show different spectral profile (*Figure 63*) revealing the obvious and clear distinction of differences between the blue paints of f.1 and f.18v. In order to complement the result from previous characterisation, the f.1 and f.18v blue UV-Vis-NIR spectra are compared with the reference of azurite and lapis lazuli (*Figure 63*).

5.1.1.2. Red: Vermillion

<u>General Observation</u>

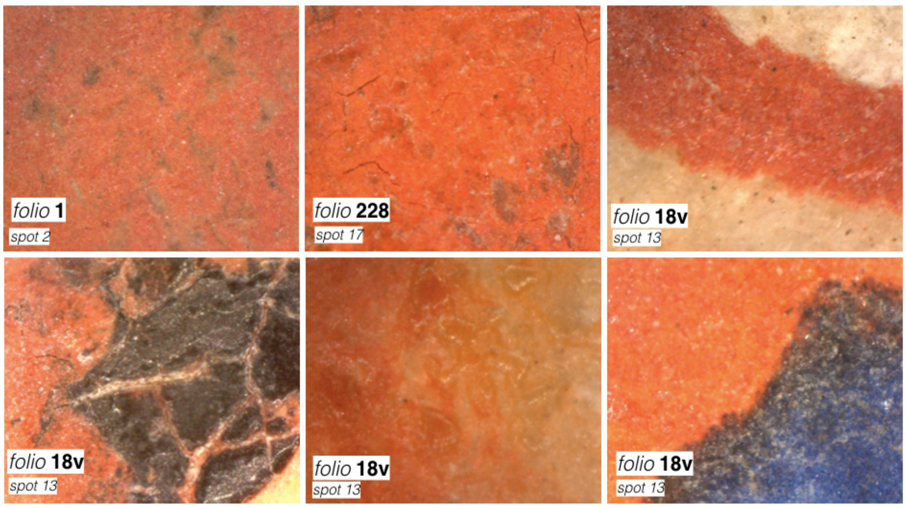


Figure 64 : Magnified images of the red paints of f.1, f.18v, and f.228 with magnification 430x, indicated in Appendix II.

Red paints are found in f.1 (initial letter), f.18v (initial letter and *incipit*), and f.228 (initial letter). Under magnified observation (*Figure 64*), all reds look very intense, dense and opaque without significant crack to the parchment. The pigment covers the parchment very well, even in some parts of f.1 where the paints are cracked, no parchment appeared. In f.18v, the red painted near to the ink looks cracked only in the ink area (*Figure 64, spot 13*). The next image of f.18v shows pale red, unlike any other. This leads to two possibilities: it is painted in yellow paint, or it has faded. Nevertheless, it has to be considered the fact that in f.18v, the yellow was painted meticulously among the ornaments of the initials that both paints may be overlapped one another.

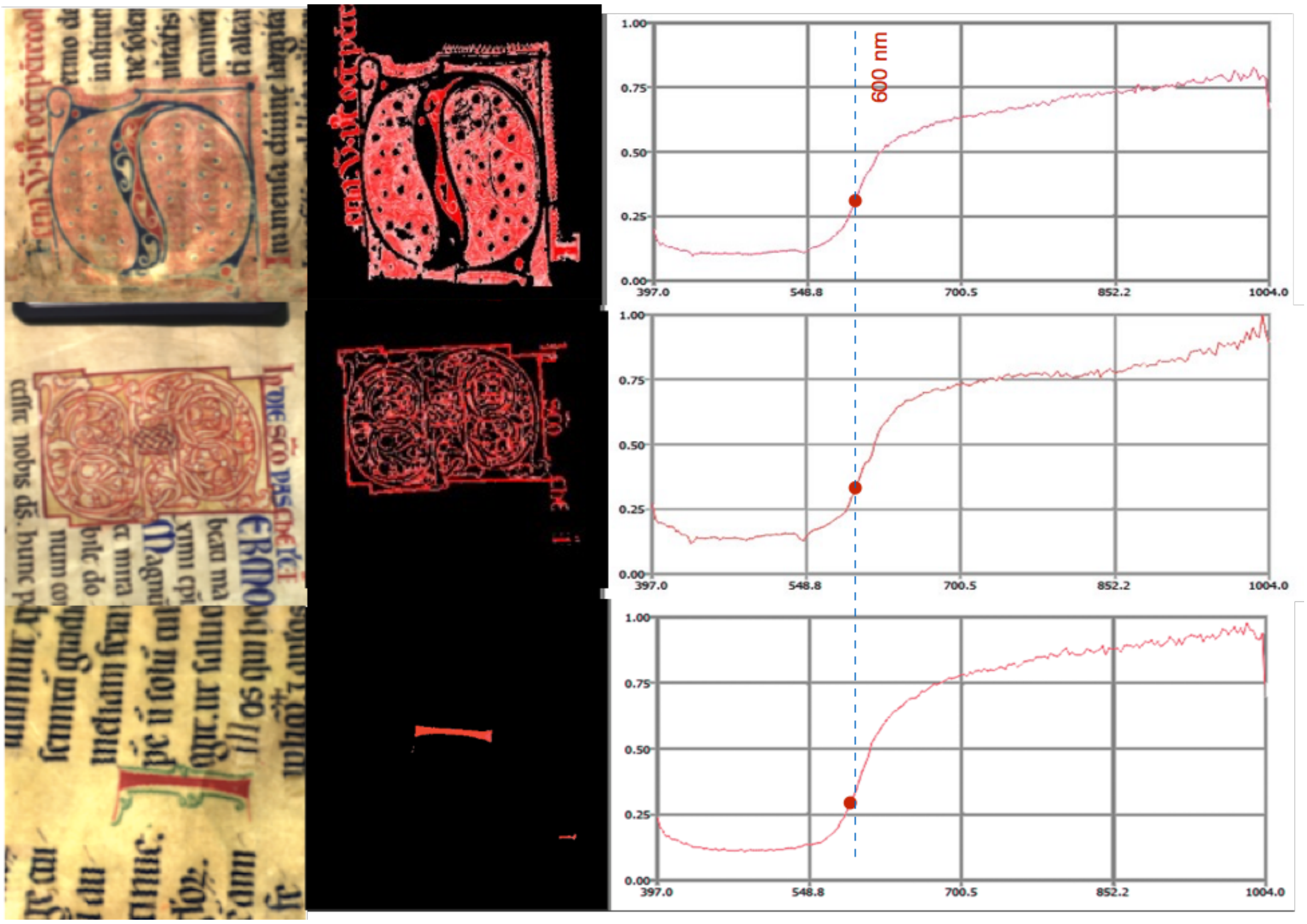


Figure 65 : Left, region of interest (ROI) of analysed by hyperspectral analysis; centre, map of distribution of the UV-Vis spectra of the red paint present in the ROI; right, UV-Vis spectrum of the red paint, where it is identified the characteristic bands of red vermillion; from above to below: f.1; f.18v; f.228.

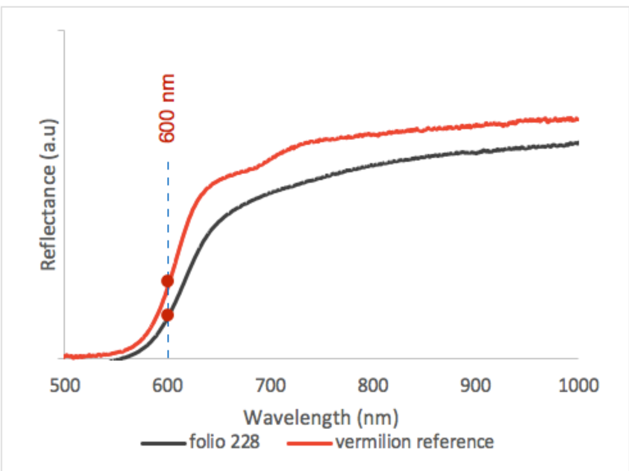
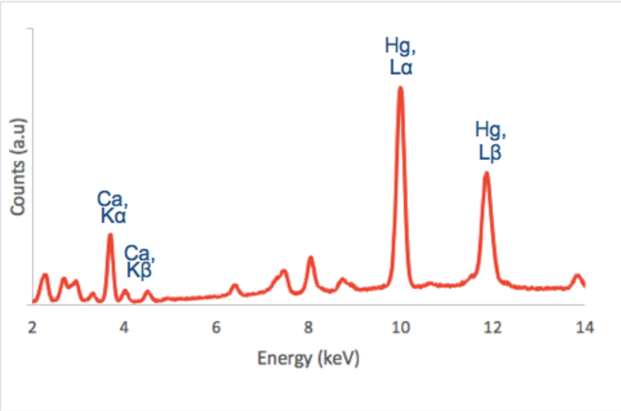


Figure 66 : The comparison between UV-Vis FORS spectra of red f.228 and vermilion reference.

<u>Materials Characterisation</u>

UV-Vis FORS analysis of this red paint allowed to identify the characteristic spectral profile of vermilion (HgS) in f.1, f.18v, and f.228 (*Figure 65*), where the spectral profiles for all *folia* show similar behaviour between 550 – 700 nm region. The inflection points are reached at the same wavelength around as the characteristic of vermilion: 600 nm (Aceto et al, 2014) as marked with a straight line through all spectra, on *Figure 65*. Additionally, the comparation of UV-Vis FORS spectra of f.228 and vermilion reference (*Figure 66*) shows similar spectral profile with the inflection point around 600 nm; and maximum peak around 590-605 nm, as the characteristic of vermilion (Aceto et al, 2014).



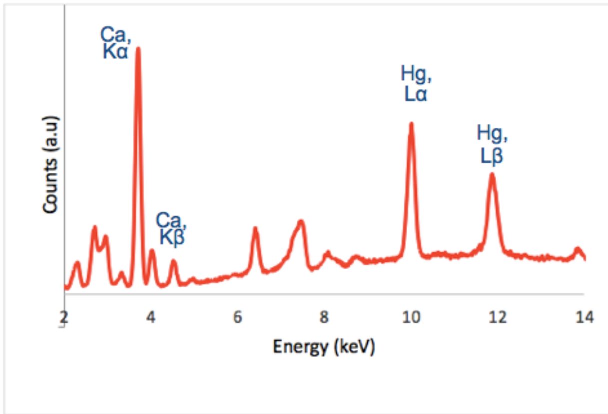


Figure 67 : h-XRF spectra of red paints of f.228 (left) and f.1 (right).

h-XRF elemental point analysis of the red paints of f.228 and f.1 exhibit the presence of the key element of vermilion: Hg (*Figure 67*). Both has significant peaks at Lα = 9.989 keV and Lβ = 11.824 keV – the characteristics of Hg. However, on red f.1, we can observe clearly intense peak on Kα = 3.692 keV and Kβ 4.013 keV that corresponds to Ca. This led to the possibility of the high amount of likely calcium carbonate as a filler (Miguel et al, 2009), or CaO as white pigment additive to the vermilion (Ricciardi et al, 2016). The presence of calcium on red pigment are almost ubiquitous in illuminated manuscript (Ricciardi et al, 2016). Although it is not as significant as on f.1, the peak that related to Ca can be found also on red f.228 spectrum (*Figure 67*, left).

5.1.1.3. Orangish Red: Minium or Vermilion?

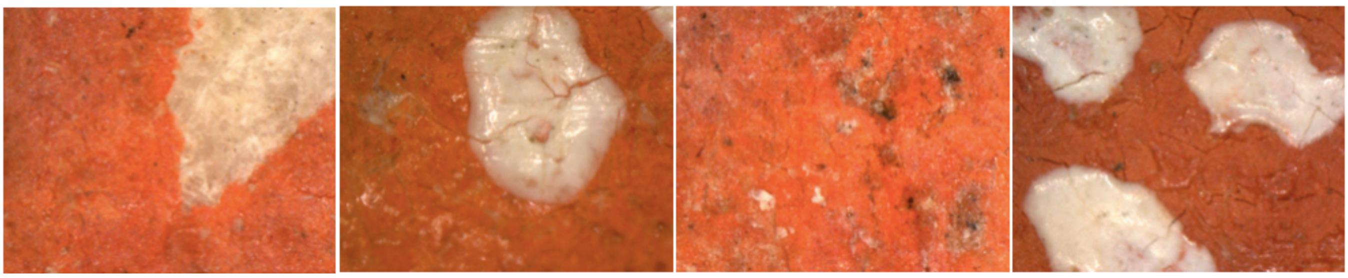


Figure 68 : Magnified images of the orangish red of f.15 with magnification 430x, indicated in Appendix II.

<u>General Observation</u>

The red paint observed in f.15 is quite different from the ones present in f.18v, f.1, and f.228. The colour appeared to be bright red- near to orange, as we describe it: orangish red. Magnified images of the orangish red of f.15 (*Figure 68*) shows clear hue of orange with a very dense and intense bright colour.

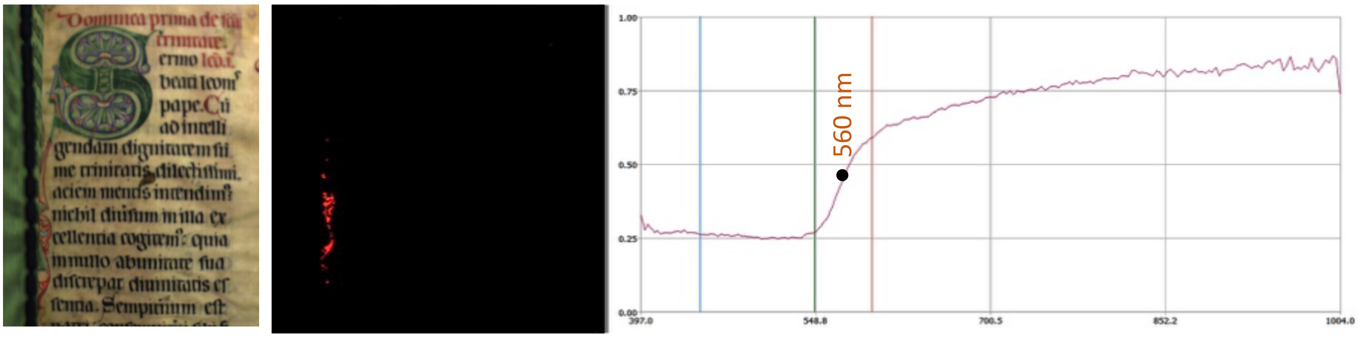


Figure 69 : Left, region of interest (ROI) of analysed f.15 by hyperspectral analysis; centre, map of distribution of the UV-Vis spectra of the orangish red paint present in the ROI; right, UV-Vis spectrum of the red paint, where it is identified the characteristic bands of minium.

<u>Materials Characterisation</u>

UV-Vis FORS analysis identifies the characteristic spectral profile of minium on f.15 (*Figure 69*) where the spectral profile exhibits the inflection point around 560 nm that correlate to the inflection point characteristic of minium (Pb_3O_4) (Aceto et al, 2014). Compare to UV-Vis FORS spectra of red paints, the spectra of orangish red in f.15 has narrower curve which we can assume that both are not the same pigment.

Although the result of XRF characterisation (*Figure 70*) shows significant peak of the key element of minium (Pb) at $L\alpha$ = 10.551 keV and $L\beta$ = 12.614 keV, it becomes complicated to distinguish since the spectra also shows the significant amount of the key element of vermilion (Hg) at $L\alpha$ = 9.989 keV and $L\beta$ = 11.824 keV. Some possibilities may occur, such as: (1) Pb might corresponds to the addition of lead white ($2PbCO_3.Pb(OH)_2$), so that the bright red vermillion looks orangish red; or (2) the presence of vermilion mixed with minium. However, the addition of white lead to vermilion gives maximum absorbance at 560 nm (Gettens et al, 1972); and white lead was likely used to be mixed with vermilion for centuries (Gettens et al, 1967). Nevertheless, Plinius referred to the addition of minium to vermilion for economic purposes (Nichols, 2016). The possibility of mixture of both pigments is likely to happen. Cennino Cennini also mentioned that the mixture of both- vermilion and minium can achieve special decoration effect (Edwards, et al, 1999). Further research is needed in order to identify the pigment by several characterisation such as Raman spectroscopy. Furthermore, if it is indeed a mixture between both pigments, the quantity of minium and vermilion can be analysed quantitatively combined with THZ-TDS (terahertz time domain spectroscopy) and LFRS (Low-Frequency Raman Spectroscopy) (Kleits & Korter, 2019).

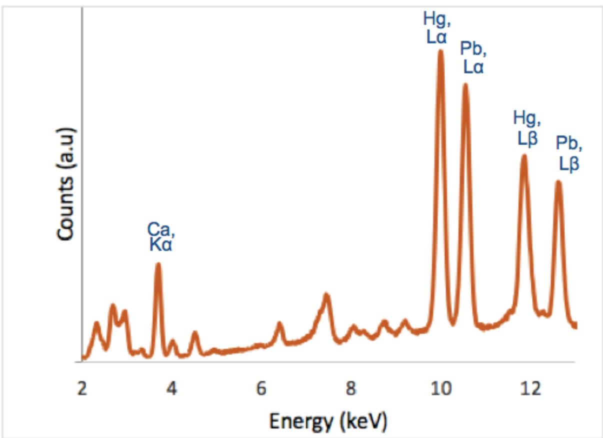


Figure 70 : h-XRF spectrum of orangish red present in f.15.

5.1.1.4. Bottle Green: which Copper Green?

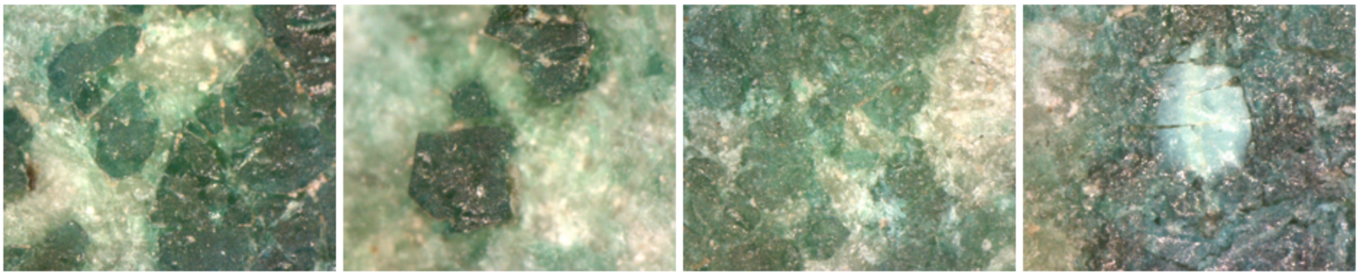


Figure 71 : Magnified images of the green paint of f.1 from spot 6 with magnification 430x, indicated in Appendix II.

<u>General Observation</u>

Magnificent intense bottle green paint was identified on the initial of f.15. The observation through digital microscopy (*Figure 71*) in the green paint of S body letter of f.15 shows the deep green, glassy fractured appearance. The green tones are not uniformed, some appeared to be very intense and dark, some very light.

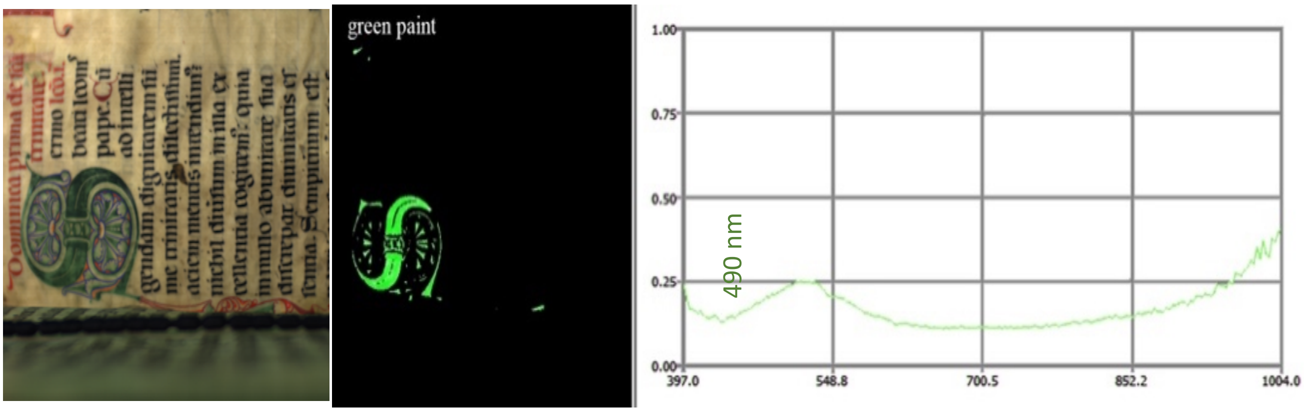


Figure 72 : Left, region of interest (ROI) of f.15 analysed by hyperspectral analysis; centre, map of distribution of the UV-Vis spectra of the red paint present in the ROI; right, UV-Vis spectrum of the red paint, where it is identified the characteristic bands.

<u>Materials Characterisation</u>

The UV-Vis FORS spectra present a maximum absorbance around 490 nm (*Figure 72*) showing a similarity to the characteristic of Verdigris (green copper or $Cu_2(CH_3CO)_4.2H_2O$) which presents a maximum absorbance around the region of 498 nm (Kühn, 1970).

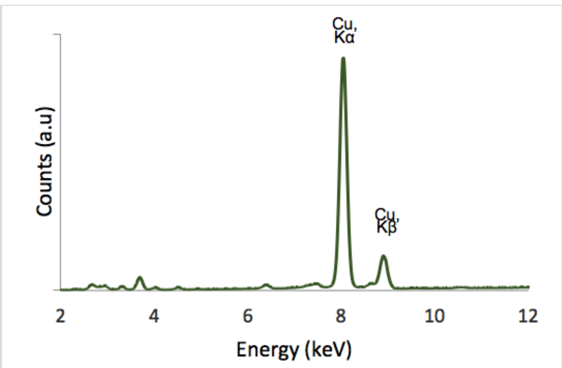


Figure 73 : h-XRF spectrum of f.15 green paint.

h-XRF spectrum of f.15 green exhibits significant peak at Kα = 8.046 keV and Kβ = 8.904 kV that corresponds to Cu (*Figure 73*). Both, UV-Vis-FORS and h-XRF characterisation shows that the green paint in f.15 has the characteristics of Verdigris. However, as stated on previous chapter, Verdigris has various green hues and compositions, thus this characterisation cannot justify which Verdigris was used for to produce the green paint. We may propose copper proteinate, but it is also possible to be painted by malachite ($CuCO_3.Cu(OH)_2$) because through XRF we can only confirm the existence of Cu. Further characterisation is needed to optimising the result and justify which Verdigris the pigment is. Through micro-FTIR and Raman spectroscopy, identifying the molecular bonds will complement the result. Previous study on bottle green pigments of Alcobaça manuscripts shows that bottle green (copper proteinate) alterations revealed a relationship between the copper proteinate complexation, the alterations of the methyl and the methylene groups of the protein binder and the loss of cohesion of the paints, which can be observed through micro-FTIR (Miguel, et al, 2018).

5.1.1.5. Yellow: Which Lake Pigment?

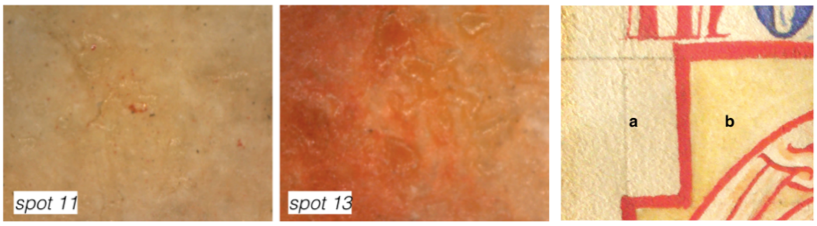


Figure 74 : Magnified Images of f.18v yellow on Spot no. 11 (all yellow) and 13 (near to red paint (or possibly mixed or covered up by red paint), depicted on Appendix II, and zoomed in inset of f.18v to differ the blank parchment (point a) and yellow paint (point b).

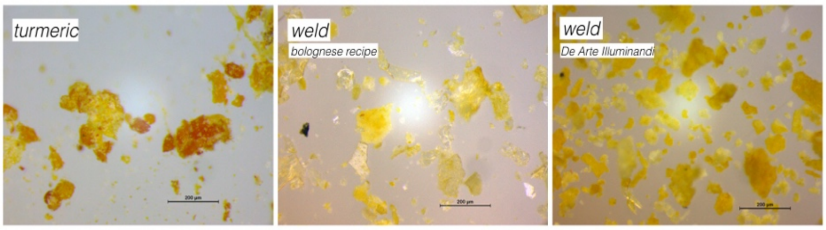


Figure 75 : Stereo Microscopy Images of Yellow Lake Pigments Reproduction.

<u>General Observation</u>

Yellow paint is found in f.18v. Magnified images evidenced the glossy pale yellow for a full painted yellow part, and a yellow coral at spot no. 13 *(Figure 74).* Unlike strong and bright inorganic yellow pigments, the f.18v yellow looks faded and pale that it was proposed to be synthesised from organic yellow dyes or yellow lake pigment. Inorganic yellow pigments were usually present in Alcobaça manuscripts, as previously stated in Chapter IV (the manuscripts are listed in Table 3). In fact, this was the first time that yellow lake pigment was characterised from Alcobaça manuscript.

To support and corroborate the analysis, the colour reproduction[58] of several yellow lake dyes was undergone: saffron, turmeric, weld (following the Bolognese recipe), and weld (following the recipe written in the De Arte Illuminandi[59]). The yellow lake pigment reproduction results show different hues of yellow as seen from stereo

[58] The reproductions of yellow lake dyes are based on medieval recipes, as explained and depicted in Appendix III.

[59] The Weld lake pigments were reproduced within two different recipes: Bolognese and De Arte Illuminandi, depicted in Appendix III. The major differences are the addition of lead white to the pigments. All recipes were reproduced with different binders: parchment glue, egg white, egg yolk, and Arabic gum.

microscope[60] (*Figure 75*). Turmeric lake pigments appeared to be yellow orange, meanwhile the welds are bright yellow. The weld produced following Bolognese recipe contains of lead white, while another weld does not.

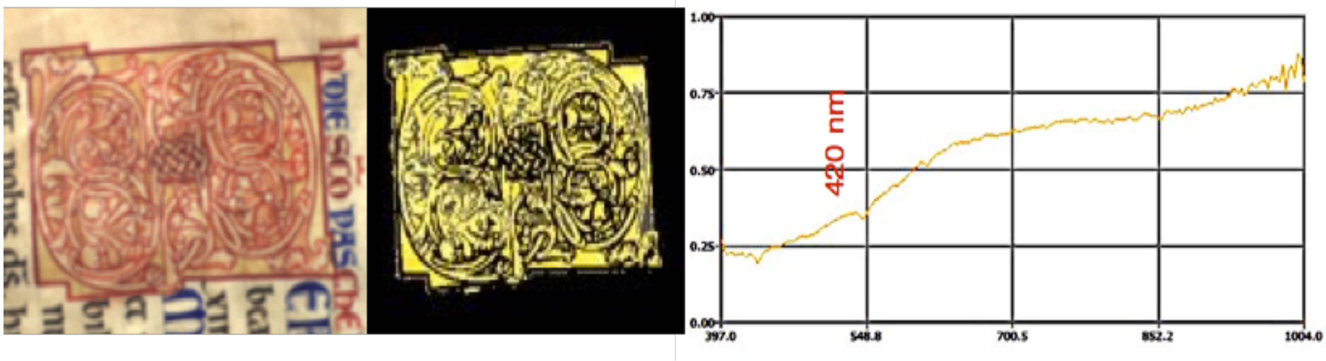


Figure 76 : Left, region of interest (ROI) of f.18v analysed by hyperspectral analysis; centre, map of distribution of the UV-Vis spectra of the yellow paint present in the ROI; right, UV-Vis spectrum of the yellow paint, where it is identified the characteristic bands of yellow lake pigment.

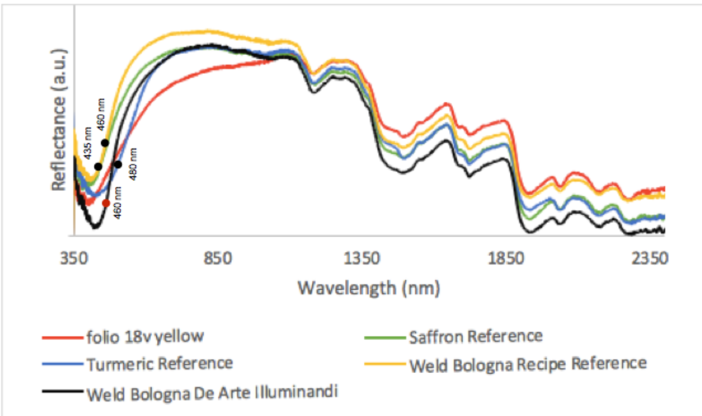


Figure 77 : UV-Vis-NIR FORS spectra of historical reconstruction of yellow lake pigments and of the yellow paint present in f.18v.

Materials Characterisation

UV-Vis FORS spectrum from hyperspectral analysis (*Figure 76*) shows a maximum absorbance in around 420 nm, close to the absorbance of yellow lake pigments (Aceto, et al, 2014). Nevertheless, this is still not an accurate enough result to be interpreted. From UV-Vis-NIR FORS analysis (*Figure 77*) of yellow paint reproduction shows similar spectral profile, close to yellow paint of f.18v, in all regions. Inflection points of each lake pigments can be distinguished clearly, with turmeric's inflection point placed on higher wavelength (500 nm) than the inflection points of saffron and weld.

On h-XRF spectrum (*Figure 78*), the highest peak corresponds to the presence of Ca. However, the question remains if all yellow lake pigments have the same spectral profiles on FORS that cannot specifically distinguish, which lake pigment is likely to be used?

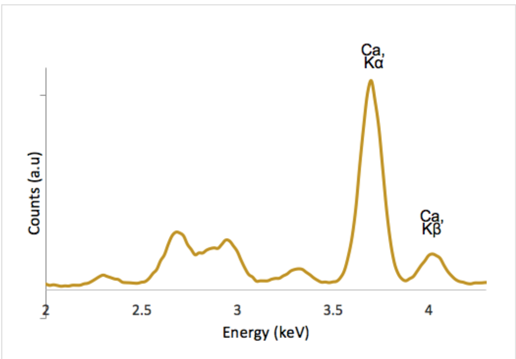


Figure 78 : h-XRF spectrum of f.18v yellow paint.

[60] The yellow saffron was not observed within microscope because it was directly grounded and mixed within the binder to produce the colour.

Chemometric analysis has been started to be widely used in cultural heritage in order to recognise and identify specific samples. PCA, or Principal Component Analysis, is powerful data-mining technique that reduces data and provides more interpretable representation (Navas et al, 2010), to principal component (Pastor, 2012). In this case, PCA is applied to the FORS data of all historical accurate reconstructions of yellow lake pigments on which the historical spectra of f.18v were projected, in Visible region (400-700nm) and NIR region (720-2090 nm) with consideration of discrimination found between the three pigments; and molecular and vibrational overtones found in NIR region.

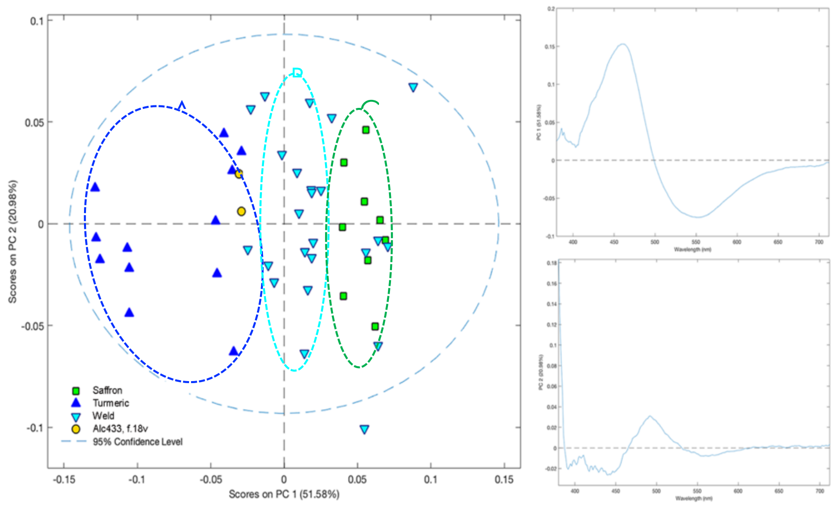


Figure 79 : Left, scores plot of PCA analysis calibrated with 51 FORS spectra of historical accurate reconstructions of turmeric (cluster A), weld (cluster B) and saffron (cluster C), on which were projected the two FORS spectra of organic yellow paints from f.18v.

The analysis of the scores plot and of the loadings (*Figure 79*) evidenced that PC1 discriminates the spectra based on the principal of inflection points of FORS spectra (400-700 nm). The loading plot indicated that contributions from the higher wavelengths are associated with a negative PC1. As for PC2, the determination is due to shift of the inflections and the change of slope, with PC2 for higher wavelength, and negative PC2 for lower wavelength.

The projection of PCA scores of the yellow lake pigments cluster closer to turmeric lake pigment (*Figure 79*). However, this result still needs to be complemented by other characterisations that may require microsample such as molecular in-situ spectroscopic techniques (such as spectrofluorometry) and Liquid Chromatography. Nevertheless, the use of turmeric is likely possible and could be interesting result if we take account to Aceto et al, stated that turmeric lake pigment is typical used in Islamic illumination (Aceto et al, 2014), which can be a very promising further research in the future.

5.1.2. Ink Analysis

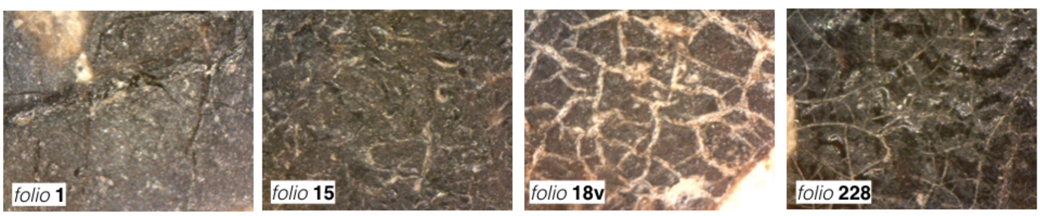


Figure 80 : Digital microscopy images of iron gall ink of Alc. 433.

<u>General Observation</u>

Iron gall ink was used writing ink in most of Alcobaça's manuscripts, as well as in Alc. 433. As what we can see clearly in *Figure 80*, each ink shows different surface heterogeneities. The assumed earliest writing ink present in f.18v, shows more cracked black layers of inks than other *folia*, as we can see the parchment under the writing ink. Under magnification, f.1 and f.15 present the more homogeneous surface; f.228 present a cracked surface, but it is in f.18v where the ink crack reaches a higher effect, with some losses of the writing ink over the parchment (*Figure 80).* However, more than just general observation and how it appears, iron gall ink may show some findings through its composition.

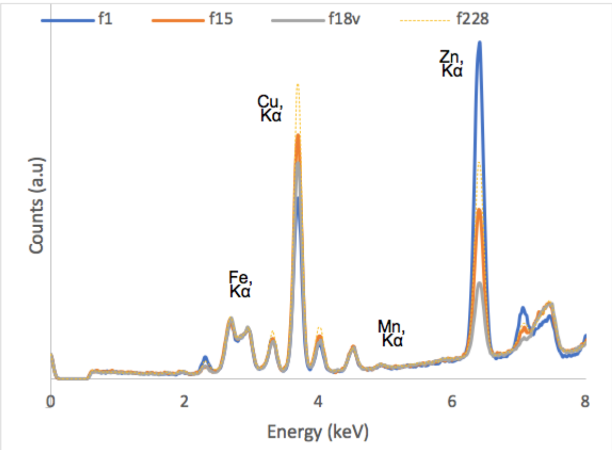


Figure 81 : h-XRF spectrum of iron gall inks from different folia of Alc. 433.

<u>Materials characterization</u>

h-XRF analysis of the writing inks present in f.1, f.15, f.18v and f.228 revealed a similar elemental composition, with Cu, Zn, Fe and Mn present in all paints, although in different composition ratios (*Figure 81*). As already mentioned in the introduction of this work, the use of iron sulphates for producing iron-gall inks was commonly linked to the presence of other metal ions, such as copper, zinc and manganese (all present in the mineral vitriol used as iron-sulphate source to produce these inks). As a mineral, it is expectable that different origins of vitriol present different chemical composition (reflected into different XRF Fe: Cu: Zn: Mn counts-ratios), which means, iron gall inks produced in different periods are expected to present different elemental compositions (Tiburcio et al, 2020). In this sense, to evaluate the use of different iron-gall inks composition along time in the Alc.433 and evaluate its link to each period of production of the manuscript already proposed according to the liturgical studies (Chapter II), a strontium[61] normalised Fe:Zn, Fe:Mn and Fe:Cu XRF counts-ratios of the writing inks present in the selected folia was analysed (*Figure 82*).

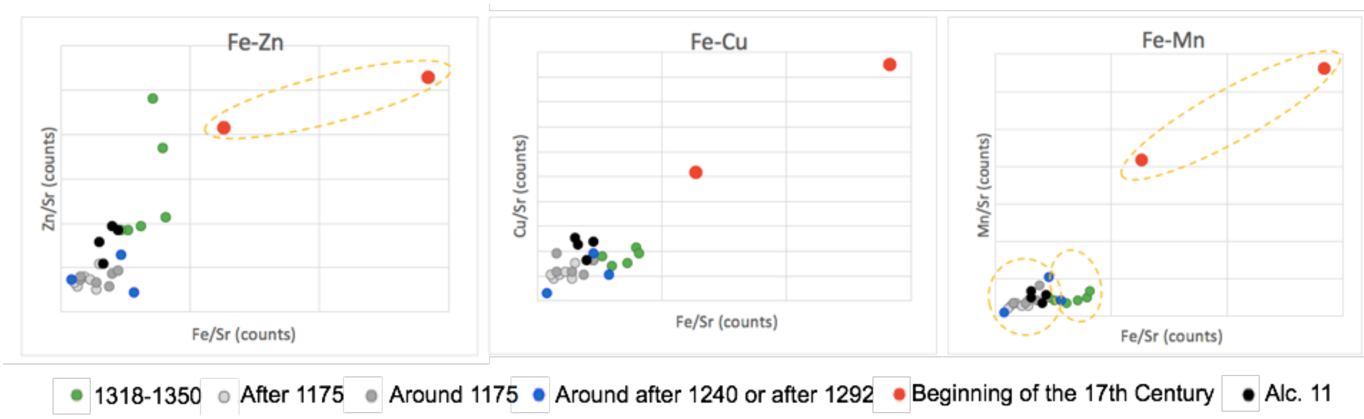


Figure 82 : XRF strontium normalised of Fe-Zn, Fe-Cu, and Fe-Mn of Alc. 433 and Alc. 11.

[61] Different normalizations were tested to ensure the influence of the distance from spot of analysis (argon normalization), the influence of the angle between the equipment and the spot of analysis (rhodium normalization) and of the inks' thickness (strontium normalization). Strontium normalization presented the best accuracy and consistency - reflecting the low influence of the inks' thickness over the EDXRF results (strontium is present as a minor element on the calcium carbonate used on the parchment production.

The analysis of the XRF strontium normalised Fe-Zn counts-ratios evidenced three main clusters, corresponding to the three periods of liturgical additions: one cluster corresponding to the *folia* linked to the around/after 1175 and around1240 or after 1292 (*Figure 82*, light-dark grey circles), on which iron and zinc are present in lower concentrations; a cluster more enriched in zinc present in *folia* linked to the 1318-1350 period (*Figure 82*, green circles); and a third cluster more enriched in zinc and iron, present in *folia* linked to the beginning of the 17th century (*Figure 82*, red circles); Interesting to notice from these results was an increment along time of the iron-zinc content to produce the iron-gall inks used in the written-texts, suggesting the use of different metallic-sulphate sources (three different vitriol sources) along time in this *scriptorium*. As for the strontium normalised Fe-Cu and Fe-Mn counts-ratios, two clusters are observed: one corresponding to the results of the earliest period of production (*Figure 82*, light-dark grey and green circles), and a second corresponding to the latest period of production – the beginning of 17th century (*Figure 82*, red circles). Even though the rations-counts were only clustered into 2 groups, the periods of the production interpreted from these data are still complementing the result of the liturgical studies and supports the use of different vitriol sources for producing the writing-inks. To evaluate the similarity of the writing-inks compositions at the beginning of the activity of the Alcobaça *scriptorium*, a caparison between the strontium normalized Fe:Zn, Fe:Mn and Fe:Cu XRF counts-ratios of the writing inks of Alc.433 and of Alc.11[62] was performed (*Figure 82*, black circles). Produced between 1185-1191, is mentioned in the *Statuta* of the foundation of the abbeys (Barreira, 2017) which means that this manuscript was directly written and noted to be produced in the late 12th century. Moreover, alongside with Coletário-ritual, the Alc. 11 contextual and palaeographic studies are the primitive product of Alcobaça. In this sense, this comparison with Alc.433 strengths the liturgical results and the writing-inks results, which dates the first corpus of Alc.433 back to the beginning of Alcobaça's *scriptorium* activity, as the XRF strontium normalised counts-ration of Alc. 11 ink clustered into the cluster of the Alc. 433 earliest analysed set (*Figure 82*).

[62] The analysis of Alc.11 were performed in the framework of the Cister.Hor project and of the Master Thesis of Samuel Arrojado (Arrojado, et al, 2019)

Table 4 : Summary of materials characterisation results based on digital microscopy, hyperspectral images, XRF and FORS (elements with asterisk sign corresponds to the key element of the pigments).

Colour	Spot	FORS & HIS Assignment	h-XRF Results	Pigment Identified
Red	2, 13, 16, 18, 21	Vermilion	Hg*, Cu, Ca	Vermilion (HgS)
Blue	12	Lapis Lazuli	Si*, Ca	Lapis Lazuli $((Na,Ca)_8(AlSiO_4)_6)S,SO_4,Cl)_{1-2}$
Blue	1	Azurite	Cu*, Ca	Azurite $(Cu_3(CO_3)_2(OH)_2$
Green	6	Verdigris (unspecified)	Cu*	Copper Carbonate
Yellow	11, 13	Yellow Lake Pigment (unspecified, possibly weld or turmeric)	Ca	Turmeric (based on chemometric analysis)
Orangish Red	7	Minium	Hg*, Pb*	Possibly vermilion (HgS) with lead-white or minium (Pb_3O_4)
Writing ink	3, 4,5, 8, 9, 10, 14, 15, 17, 19, 20, 21, 22	Iron Gall Ink	Fe*, Zn*, Cu*, Mn*	Iron Gall Ink

5.2. Drawing Technique Analysis

Figure 83 : The initial letter of Alc. 433 f18v with the zoomed-in lead-point traces. © Biblioteca Nacional de Portugal

The observation under magnification of the capital letter of f.18v allowed to identify the presence of some points and lines in the middle of letter, suggesting the use of "guiding-supports" to draw the red-paint interlaced (*Figure 83*). XRF analysis of these "guiding-supports" allowed to identify the presence of lead, suggesting the use of a lead-point as a tool (*Figure 84*). Interesting to notice that the grey line beneath the red line of body S is noticeable, however, the same line is not seen underneath the red ornaments. Based on the references studies of illumination techniques explained in previous chapter, it is likely that the stencil technique was used during this period, nevertheless the evidences for the use of a trace lead-point clearly seen with naked eyes in the body of this capital letter, points for the use of a free-hand approach, making use of "guiding-lines and points" drawn with a lead point to trace the guide-lines of the drawing of the capital letter.

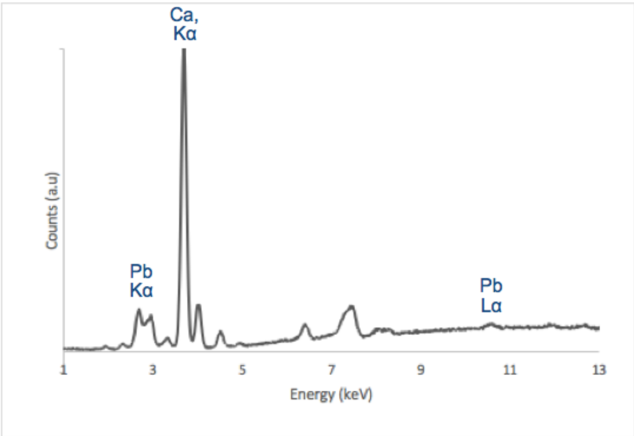


Figure 84 : h-XRF spectrum of lead-point trace from f.15v of Alc. 433.

Part IV
THE GRAIL
Final Remark and Future Research

Chapter VI: The Final Remarks

"Scientiae enim ea natura est, et use intelligi non possit, nisi ab illis qui sunt scientia praediti"[63] *(Francis Bacon, from Meditationes Sacrae)*

As people said, *every quest has its grail*- the grail of this research was to find the answer of the main question stated before: the biography of Alc. 433. Through multidisciplinary research spotlighting each *folio* that was assumed to be produced within different period, liturgical studies (contextualisation and content of the liturgy and ornament analysis) combined with materials investigation (general observation and materials characterisation), allowerd reach to the conclusion of the biography of Alc. 433.

6.1. Pigments Time to Time

It has been known that each *scriptorium* of a monastery has their own pigment palette time to time. As time flew centuries to centuries, the palette has changed and adapted, too. Some colour hues were developed, yet some pigments might be gone and replaced by other pigments that were more affordable reachable, stable, and less harmful, following the environment of *scriptoria*.

First, the colour of Virgin Mary: Blue. The beginning of Alcobaça establishment in the late 12th century, we focused on the precious lapis lazuli pigment that had been characterised within f.18v on the *incipit*. This is obviously an expected result because we certainly knew that lapis lazuli used in Alcobaça during this period through several previous studies on different manuscripts produced in this century. However, judging from the next folio that assumed to be produced during 14th century, f.1, the azurite replaced the precious lapis lazuli- as it is predicted, since azurite was more affordable.

Second, the most popular and important colour: Red. Although some papers stated that the use of minium was firstly came up before vermilion, it was evidently clear that vermilion was used within the 12th century, until the 14th century; judging from the red characterised on f.1, f.18v, and f.228. As vermilion sources were abundant in Europe, it is likely possible that vermilion was kept being used all the period of the Alcobaça production. Although minium was appeared on f15 which was painted for orangish red which the colour was seen orangish than vermilion. It is possible that minium was used to paint specific orangish red hues rather than bright red colour.

Third: the mysterious bottle green which appeared on f.15- assumed to be produced on early 13th century. Since blue and red are basic colour palette, the appearance of bottle green was rather distinctive. It is no strange

63 "For science is of that nature, as none can understand it to be, but such in a good measure have attained it" (Translated by Thomas Hobbes, in his later work: De Corpore, 1655).

that this colour actually found in this period, where the illuminated manuscript became trend on the medieval Europe.

Finally, the only organic pigments characterised in selected *folia*: the yellow lake pigment. At the beginning, we hypothesised that the pale yellow appeared on the f18v was weld lake pigments after slightly observe the FORS spectra, but the result of chemometric analysis was surprising yet promising. The turmeric is likely to be used within this period. Logically thinking, if we focus on 12th century, the uses of turmeric which was widely well-known to be used as herbs was not uncommon at all. As stated on the previous research by Aceto et al, if the turmeric was indeed commonly used in Islamic illumination, then turmeric was not surprising at all to be found on Alc.433. As we know, culture and political environment influenced the trade of the pigments as well. Eventually it is assumed that the yellow paints characterised on f.18v was painted with turmeric lake pigment.

In conclusion, the result of the pigment characterisation alongside with the liturgy studies can be seen clearly on Figure 103 as pigment timeline of Alc. 433.

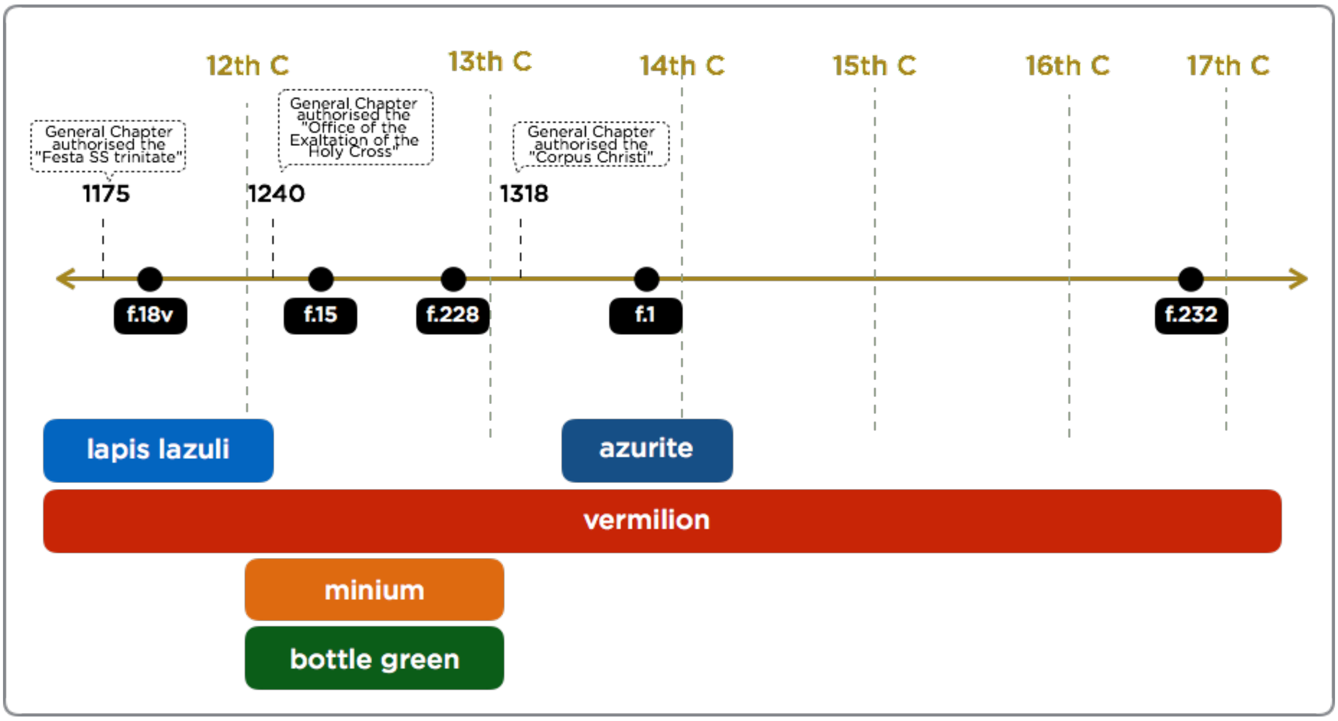


Figure 85 : The timeline of the Alc. 433 and the pigments used that were characterised in this research.

6.2. The Alc. 433 Biography: The Complete Story

This research was meant to deliver the story of Alc. 433 which will develop the biographical story of Alcobaça scriptorium since its establishment until the end of the active year. The final result of this idea is expected to eventually reveal the complete and detail history of Alcobaça *scriptorium* and its illumination details- that lead to the history of Portuguese Cistercian Order- indirectly to the Portugal history. It is a very optimistic project, promising result, and immense contribution for this field.

This thesis has described the multidisciplinary studies across science and humanities to reveal the complete story beyond the Alc. 433 through its representative *folia*. From the liturgical studies, we have seen that the *folia* chronologically added and successfully traced through its liturgical context from well-recorded *Statuta* of Cistercian Order. Furthermore, the scientific analysis of the Alc. 433 materials complemented the result proficiently well.

The Alc. 433 story is presumably begun on the as the initial core- The *Temporale*, started on f.18v- with the initial painted by enchanting intense red vermillion; the red ornament filled by presumably turmeric yellow, and completed by incipit written using lapis lazuli blue. The next chapter was added: the *Festa SS Trinitate*, for the celebration of The Holy Cross, started by the f.15- with the initials painted by bottle green decorated by orangish minium. The Office Lesson, then, added later on 13th century, more precisely after 1292; marked by the f228 where vermillion was apparently still used to paint the initial letter. On the next addition- after 1318 the *Corpus Christi*, we figured out that the lapis lazuli blue was replaced by azurite blue- darker hue of blue, alongside with red vermillion. Finally, with the evident of bookbinding and no illumination exist on the last chapter addition started by f.232, the Alc. 433 was finished to be produced on the beginning of 17th century.

This research has tried to show the trajectory of Alc. 433 which had been undergone within multidisciplinary embracing materials science, history of art, and liturgical studies, that it is believed to improve the state of art of the production of illuminated manuscripts in Alcobaça, that hopefully will be enhanced in future research.

6.3. Future Research

It is indeed simple and short to describe briefly the story of the result of this thesis, written in previous paragraph. The result is indeed promising and complementing each other from different aspect, however it is far from perfect.

This thesis was undergone between March-September 2020 when the COVID-19 world pandemic hit Europe that affected the work of this research as well. For the safety of the researchers, we could not investigate the Alc. 433 further during this period to characterise the bookbinding, detail examination of each *folio* using other techniques such as Raman microscopy and chromatography if micro-sampling is possible to do. Other than that, the result of this thesis is clearly need to be researched more to obtain even more accurate and promising result.

The liturgical studies will be complemented even better if the characterisations of the materials are complete with the bookbinding analysis. Some colours of the *folia* can be characterised in order to collaborate and support the result within the same the chapter. Another important development of this research is the yellow lake pigments. It is needed to be researched more as well as to be compared with another Alcobaça manuscripts that probably painted by yellow lake pigments. The research also can be explored to the palaeography and the complete archaeology of the Alc. 433; linked to the history of the *scriptorium* as well as the monastery itself. By then, it is highly optimistic that the objective of the complete vision of the purposes of the project will be revealed as full body *Holy Grail* of the Medieval Portuguese illuminated manuscript.

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Appendix I: Instruments of the Characterisation

Fibre Optic Reflectance Spectroscopy (FORS)

The FORS analysis of the Alc. 433 and references materials were performed by Lr1-T v.2 compact spectrometer (ASEQ instruments) with 50 μm slit; with spectral region in UV-Vis-NIR: 300-1000 nm. Data was processed through ASEQ Check TR Software, resulting spectra from each characterised material, then, processed using Microsoft Excel.

UV-Vis-NIR analysis were performed using a BWTEK i-Spec 25portable spectrometer in combination with a handheld reflectance probe of trifurcated fiber optic bundle series integrated with a 5W tungsten halogen source with a focal aperture of 5mm and extended InGaAs array sensor, was used for analysis at Escoural Cave. The spectrometer measures across the UV-Vis-NIR range from 400- 2500 nm, and the analysis is compiled through the supporting iSpec4 program.

After initial setup of the i-Spec 25 FORS with a portable field laptop, the 3 detectors (Detector 1: BRC711U-512 [345.6nm-1061.3nm]; Detector 2: BTC261P-512-OEM61 [883.0NM- 1718.0NM]; Detector 3: BTC263E-256-OEM61 [1482.4NM-2654.9NM]) were set to the previously optimized settings of Detector 1: integration time 95ms, average 25, smoothing none; Detector 2: integration time 250µs, average 50, smoothing none, detector mode of high sensitivity; Detector 3: integration time 332µs, average 100, smoothing none, detector mode of high sensitivity.

As for yellow pigment, an unsupervised approach using Principal Component Analysis (PCA) of FORS spectra restricted to the visible region (400-700 nm) and to the NIR region (720-2090 nm) was performed using MATLAB (version R2016a) and PLS toolbox (version 8.2.1) from Eigenvector Research Inc. FORS spectra of historically accurate paint reconstructions of yellow lake pigments were pre-processed with Standard Normal Variate (SNV) for scaling the spectra (weighted normalization), followed by Mean Centre for removing mean offset from each variable, and a Savitzky-Golay smoothing filter with 15-point window size, second-order polynomial and first derivative. The PCA model was calibrated with 51 FORS spectra of historically accurate reconstructions of saffron, turmeric and weld paints produced with proteinaceous (parchment glue, egg white and egg yolk) and polysaccharides binders (gum Arabic) on which two FORS spectra of yellow paints from Alc.433, f.18v were projected.

Handheld X-Ray Fluorescence (h-XRF) Spectrometer

The XRF analysis was performed using Bruker Tracer III-SD handheld XRF Spectrometer Bruker, equipped by a 10 mm^2 Xflash SSD, a peltier cooled detector with a typical resolution of 145 eV at 1--,000 cps, a Rh target and a maximum voltage of 40 kV. Analyses were made at 40 kV and 12.5 μA, with an Al/Ti lter and an acquisition time of 90 seconds.The instrument was set up on a tripod and positioned approximately 2–3 mm away from the surface under analysis. A total of 150 XRF spectra were collected using the S1PXRF software and analysed using the ARTAX software.

Hyperspectral Camera

The Hyperspectral Image Characterisation was performed by SPECIM Hyperspectral camera (400-1000 nm) alongside lamp to artificially illuminate the object. The camera was used in default recording method (DRM) with simultaneous or custom white reference collection. After focusing to the object, approving integration time, the spectral data is recorded, followed by calibration of white, and image was captured with the hyperspectral wavelength. All data were imported to Specim IQ studio to be processed.

Digital Microscopy

Magnified images were processed by Dino-Lite digital microscope with 430x magnification. Data was processed through DinoCapture 2.0 Software.

Stereo Microscopy

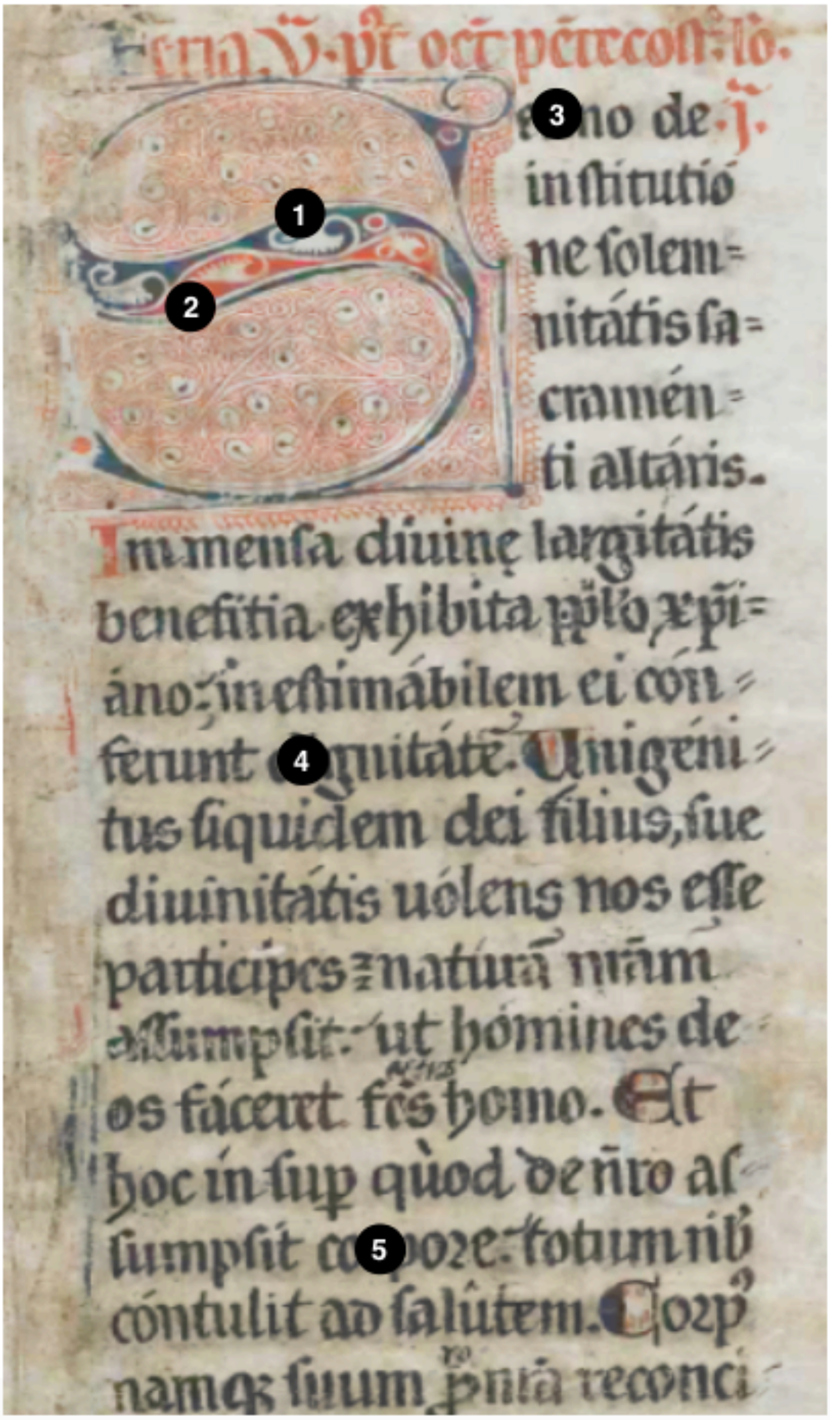
Magnified images of yellow pigment were taken under LEICA M205C stereomicroscope (zoom range 7.8x-160x) equipped with LEICA DFC295 camera.

Technical Photography

The technical photography was undergone using Nikon D3100 14.2MPx digital single reflex (DSLR) camera with a 10.0-55.0 mm f/3.5-5.6 lens (for Alc. 433); and Olympus E-M10 Mark III Kt 1442 Mirrorless camera with M. Zuiko Digital lens 45mm f1.8 (for historical reconstruction and yellow lake pigments reproduction.

Appendix II: Characterisation Spots

Folio 1



Legends

1 - Blue

Hyperspectral Image, FORS, XRF, digital microscopy

2 - Red

Hyperspectral Image, FORS, digital microscopy

3 - Iron Gall Ink

XRF, digital microscopy

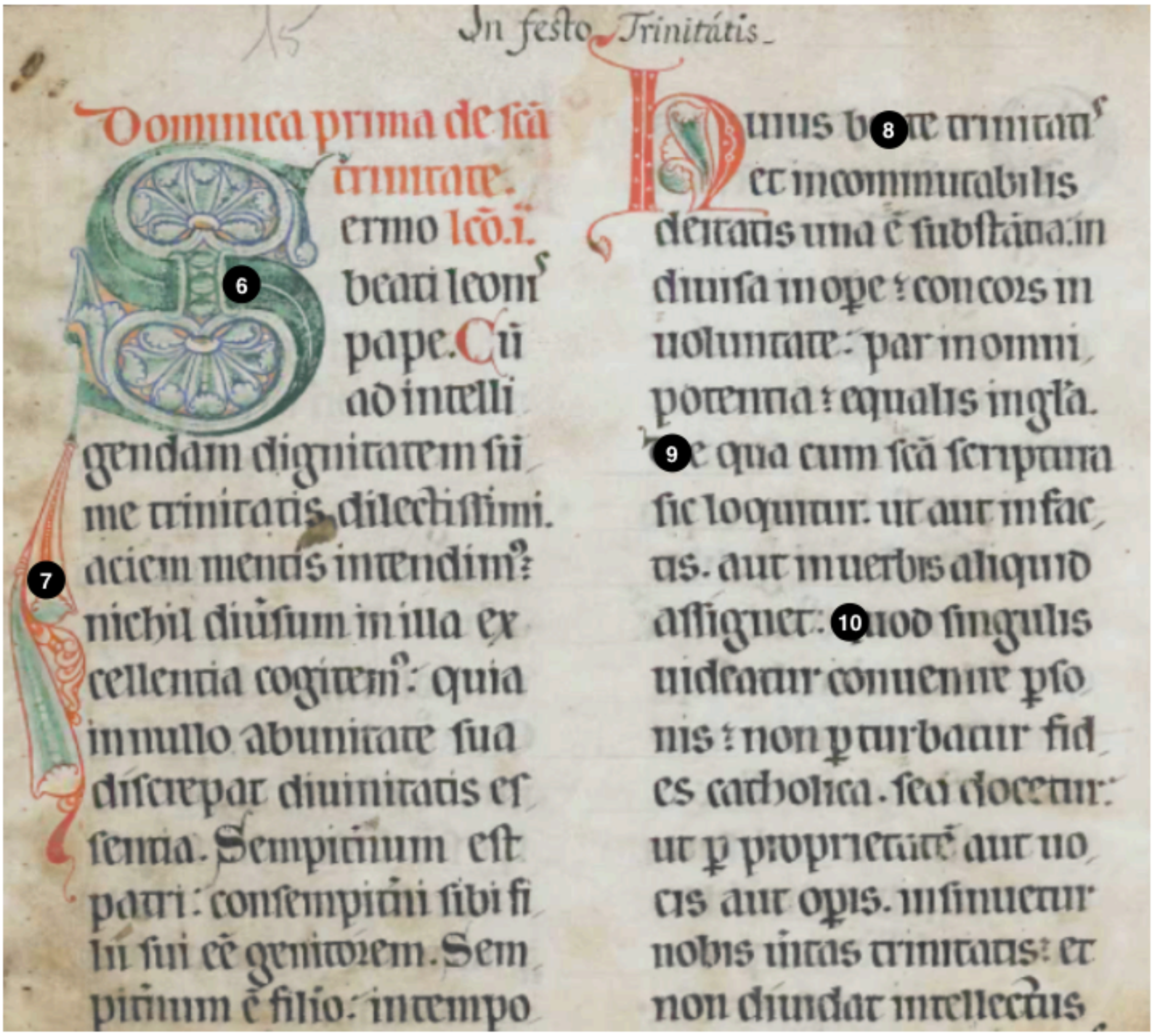
4 - Iron Gall Ink

XRF, digital microscopy

5 - Iron Gall Ink

XRF, digital microscopy

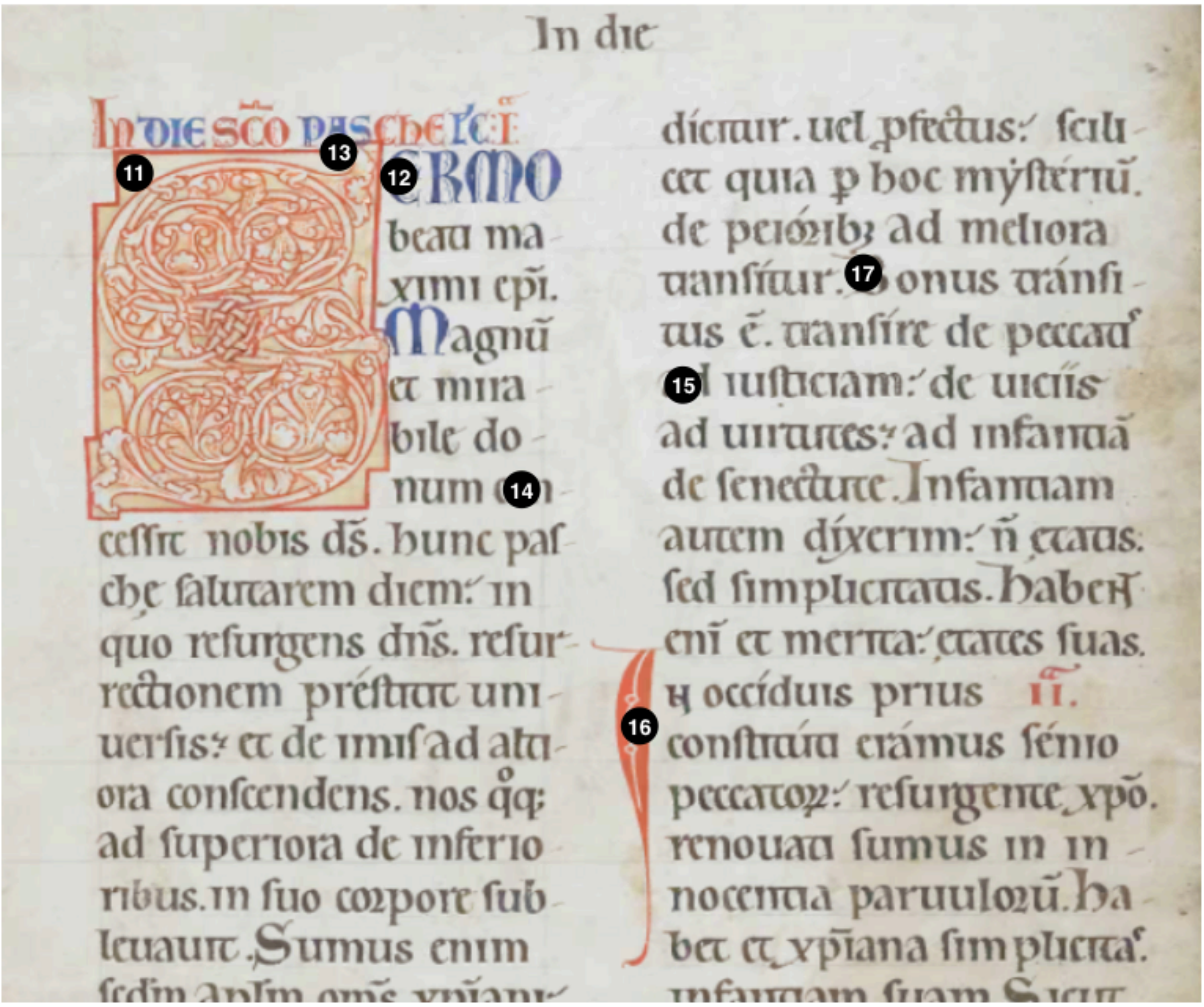
Folio 15



Legends

6 - Green
Hyperspectral Image, XRF, digital microscopy
7 - Red
Hyperspectral Image, XRF, digital microscopy
8 - Iron Gall Ink
XRF, digital microscopy
9 - Iron Gall Ink
XRF, digital microscopy
10 - Iron Gall Ink
XRF, digital microscopy

Folio 18v



Legends

11 - Yellow
Hyperspectral Image, FORS, XRF, digital microscopy

12 - Blue
FORS, XRF, digital microscopy

13 -Red
digital microscopy

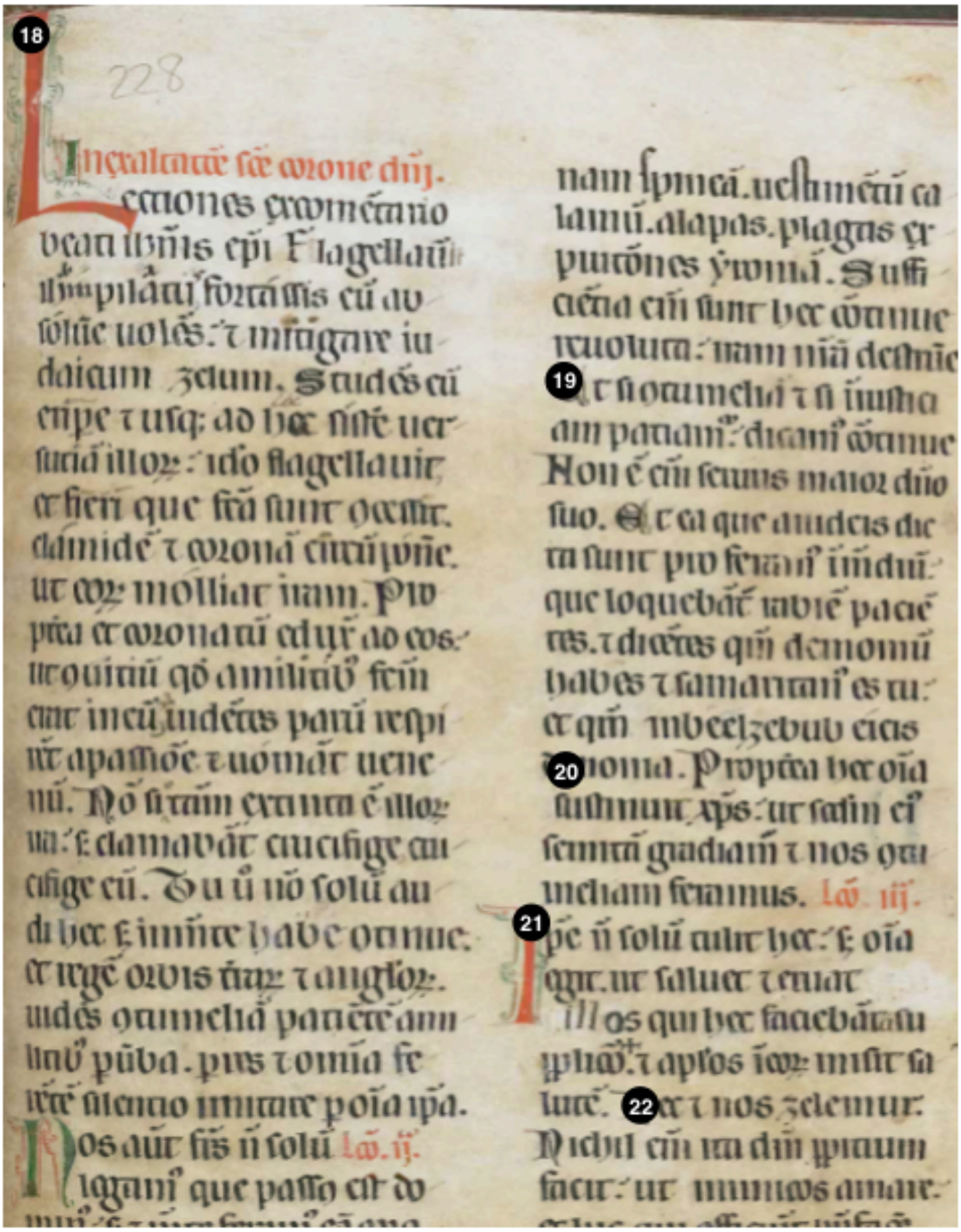
14 - Iron Gall Ink
XRF, digital microscopy

15 - Iron Gall Ink
XRF, digital microscopy

16 - Red
XRF, digital microscopy

17 - Iron Gall Ink
XRF, digital microscopy

Folio 228



Legends

17 - Red

Hyperspectral Image, FORS,

XRF, digital microscopy

18 - Iron Gall Ink

XRF, digital microscopy

19 - Iron Gall Ink

XRF, digital microscopy

20 - Red

Hyperspectral Image, FORS,

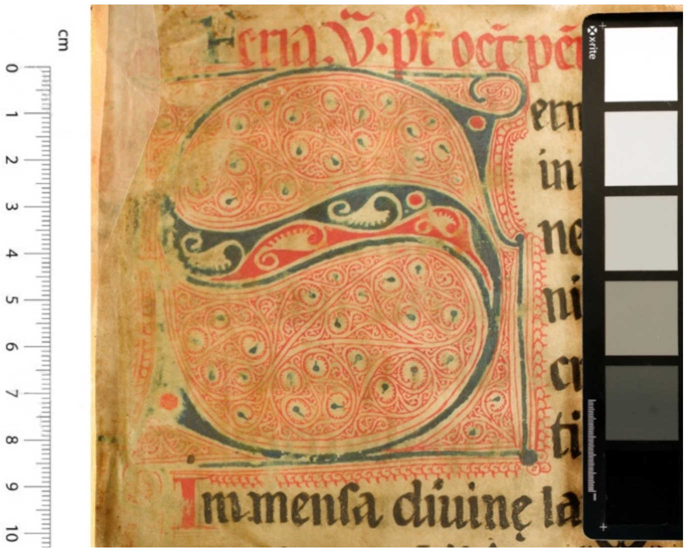
XRF, digital microscopy

21 - Iron Gall Ink

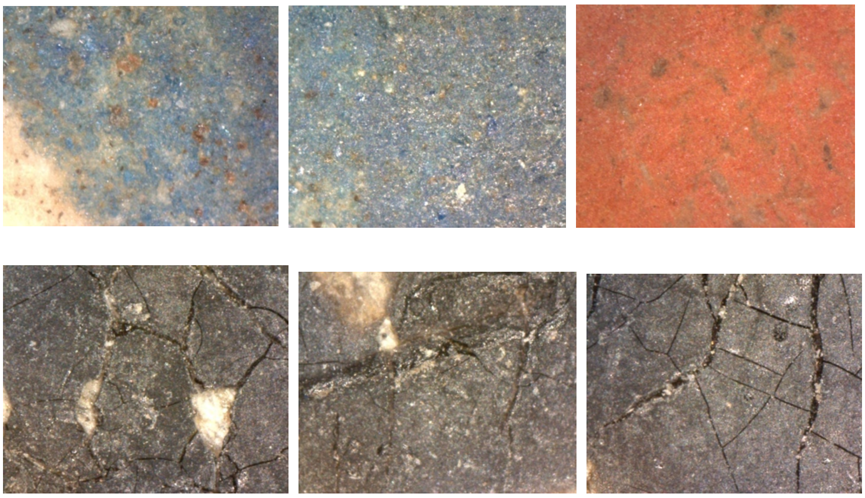
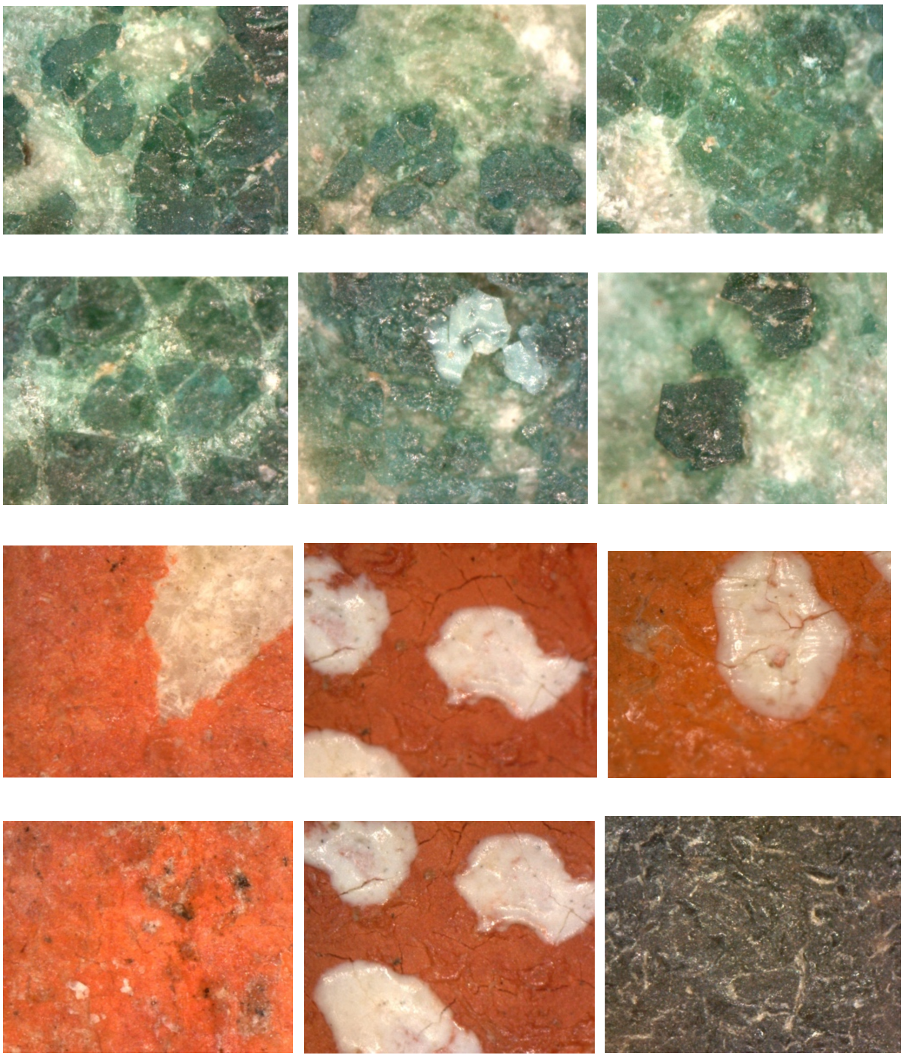
XRF, digital microscopy

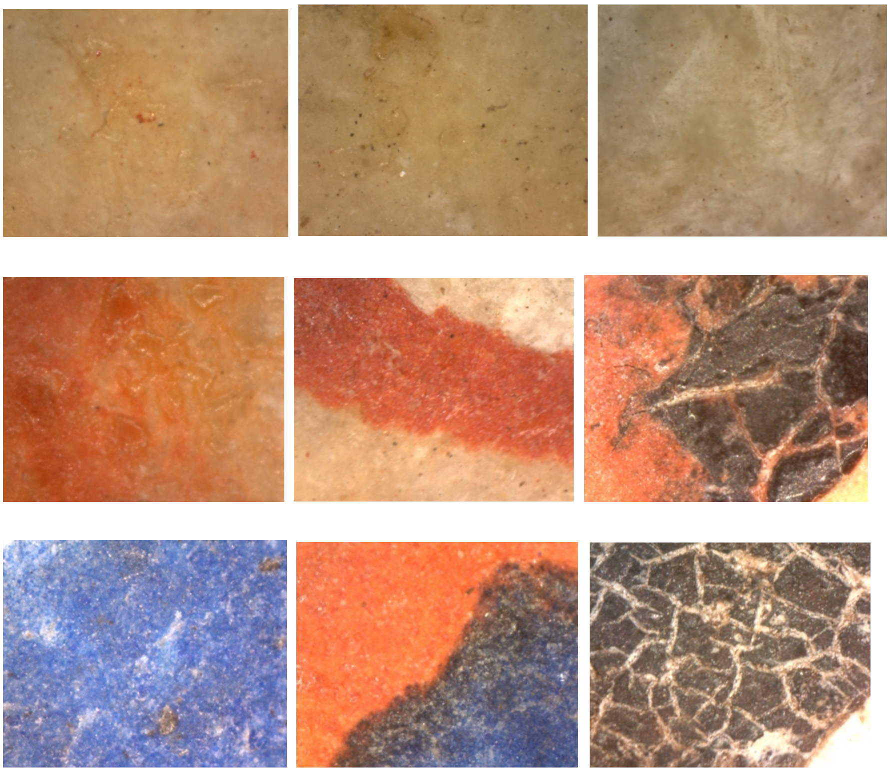
Appendix III: Characterisation Result

Technical Photogtaphy



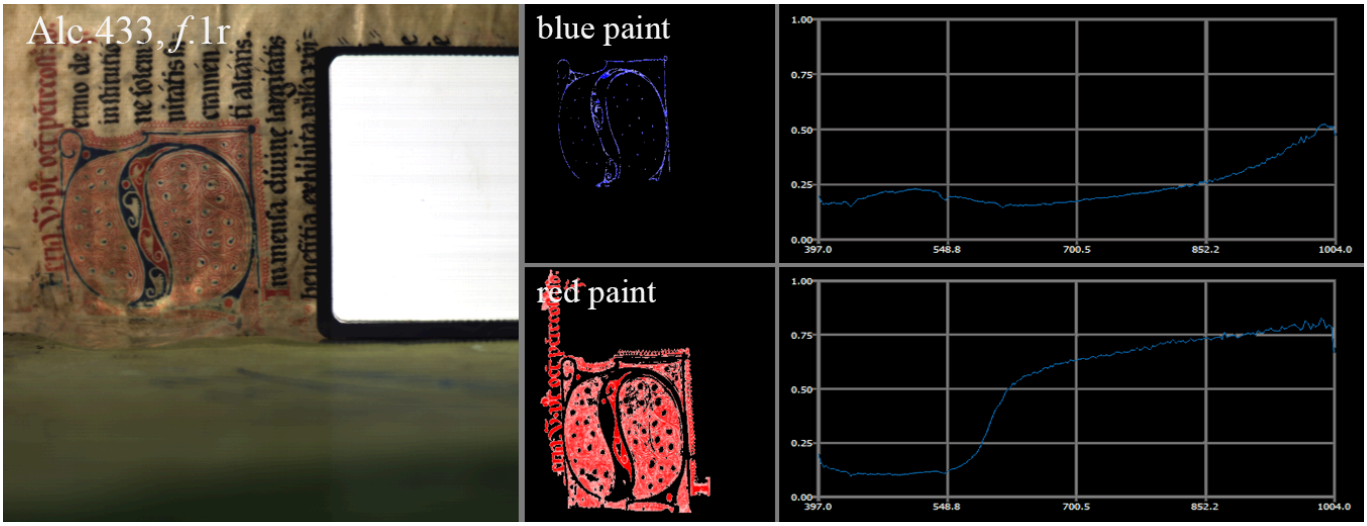
Digital Microscopy

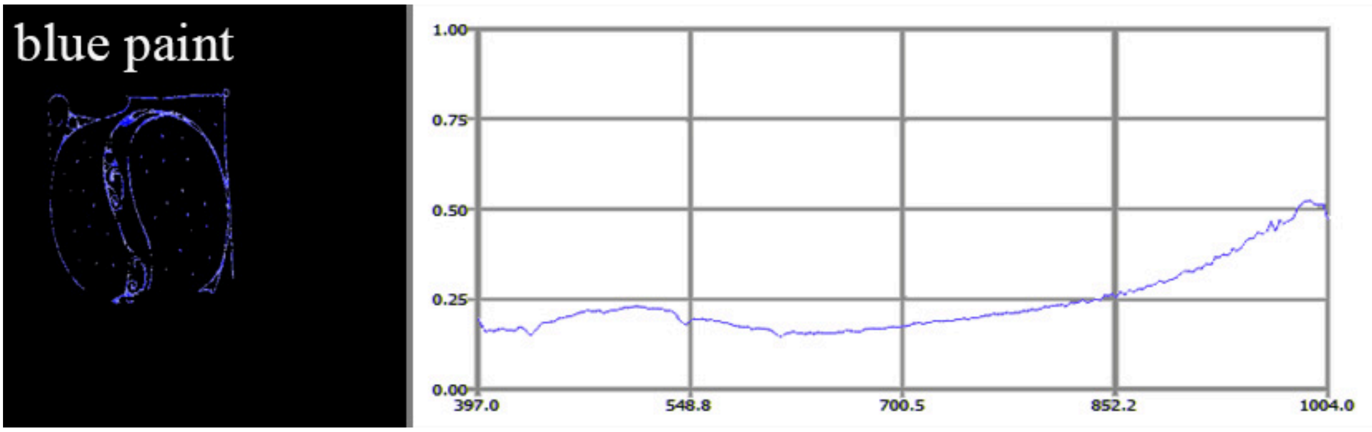
Folio	Images
f.1	
f.15	

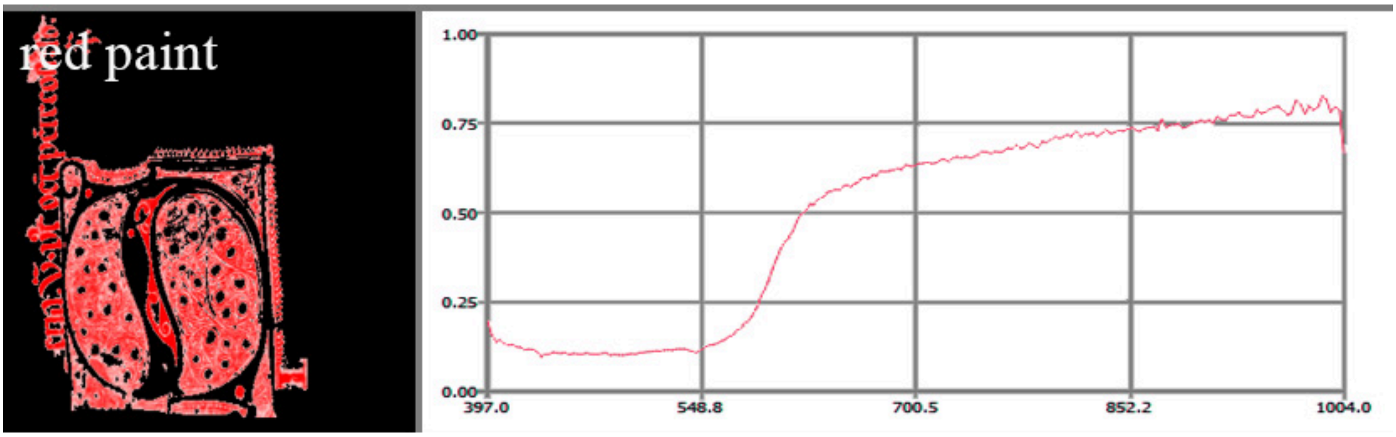
f. 18v	
f.228	

Hyperspectral Images

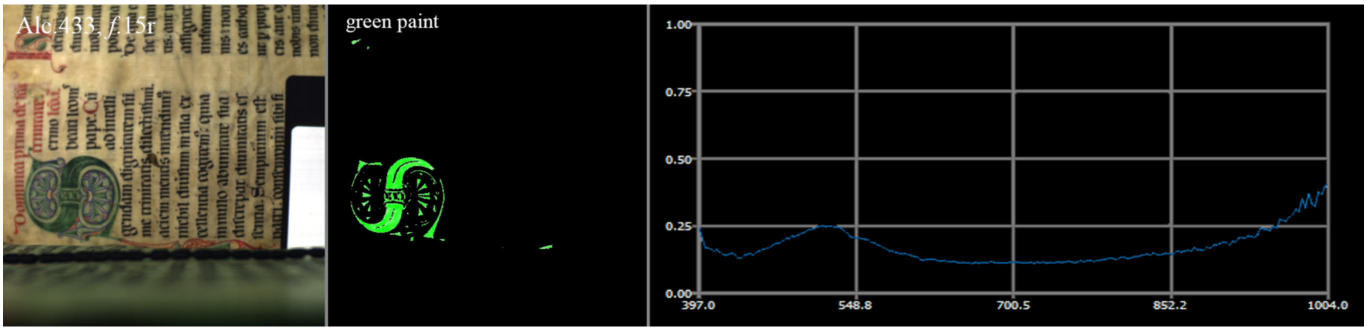
Folio 1

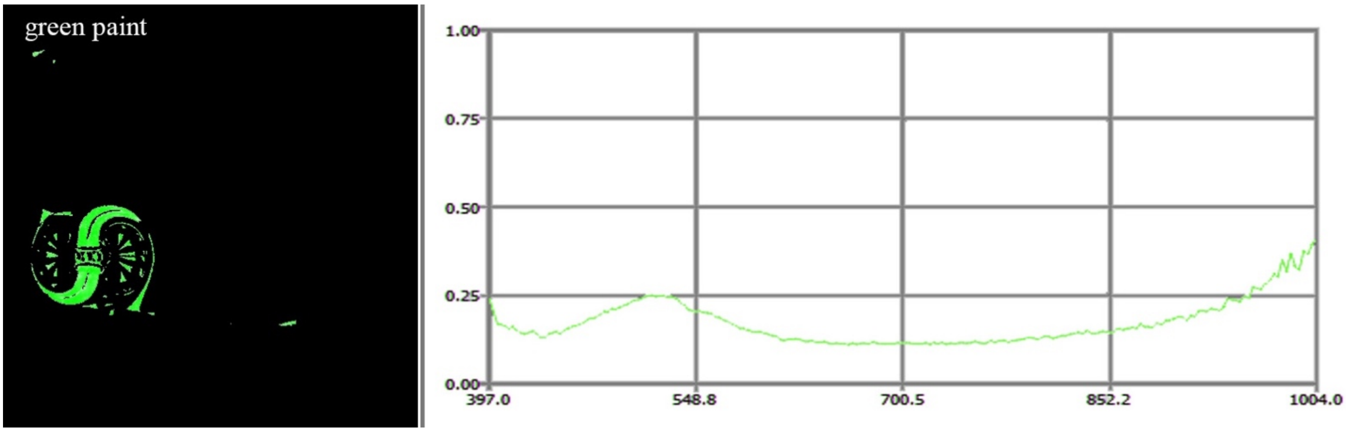


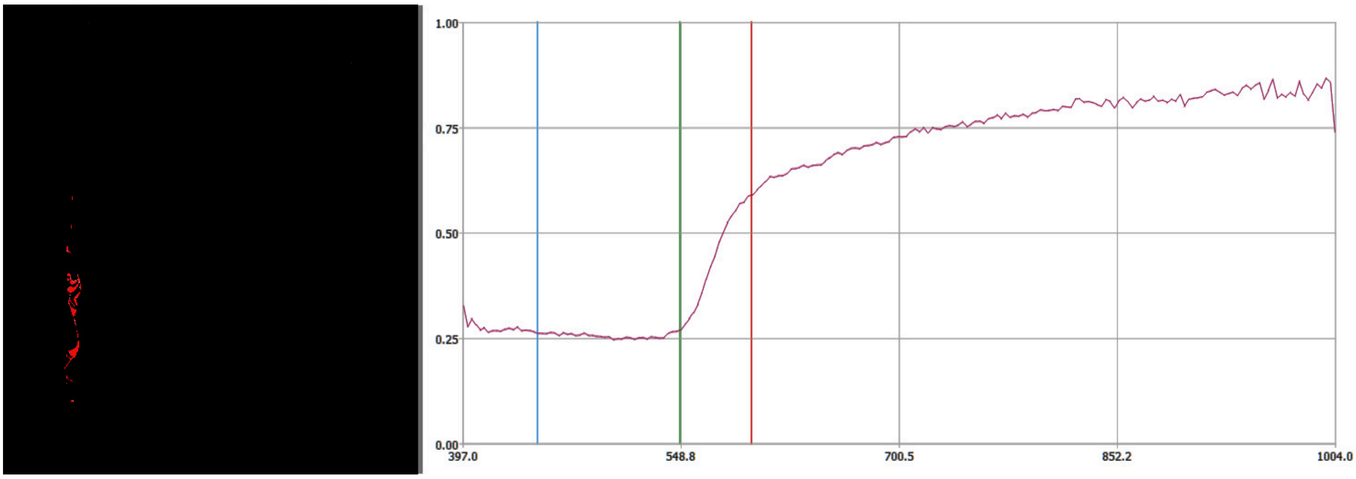




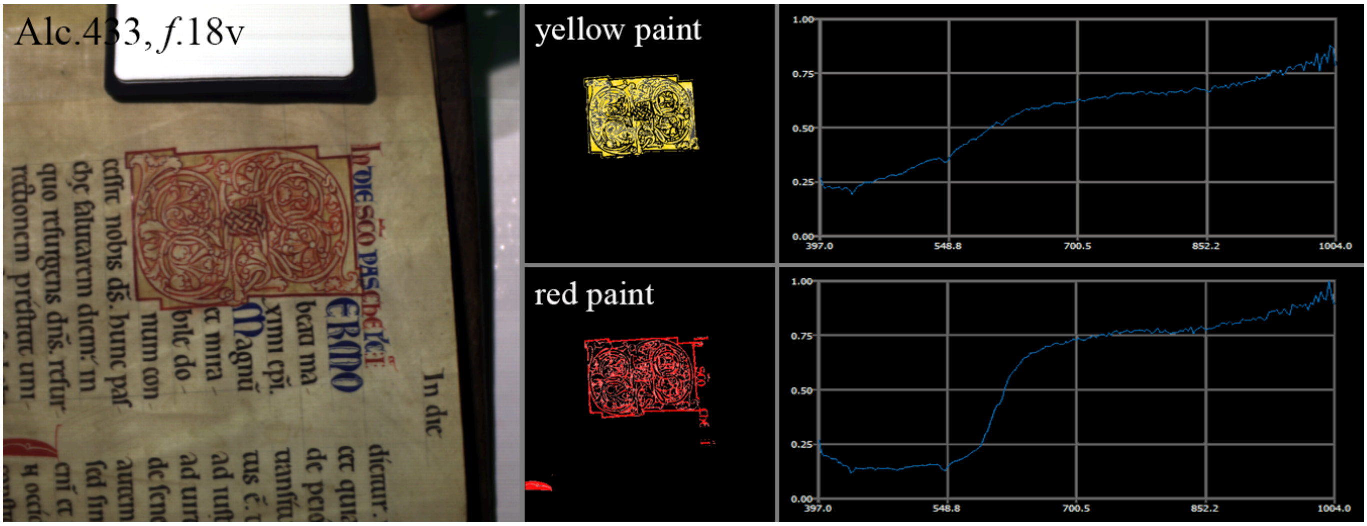
Folio 15

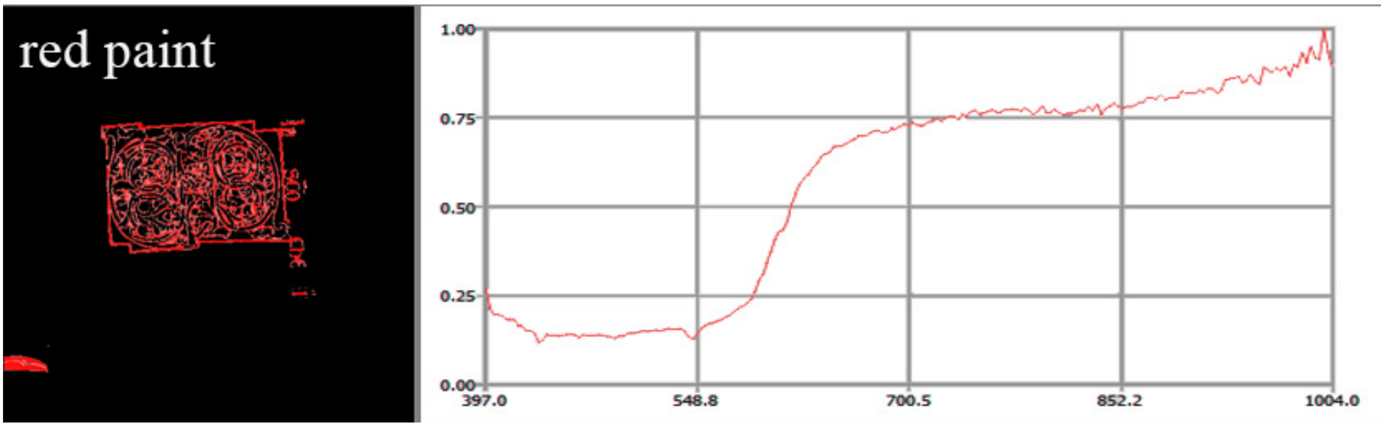

Alc.433, f.15r
green paint
1.00
0.75
0.50
0.25
0.00
397.0
548.8
700.5
852.2
1004.0

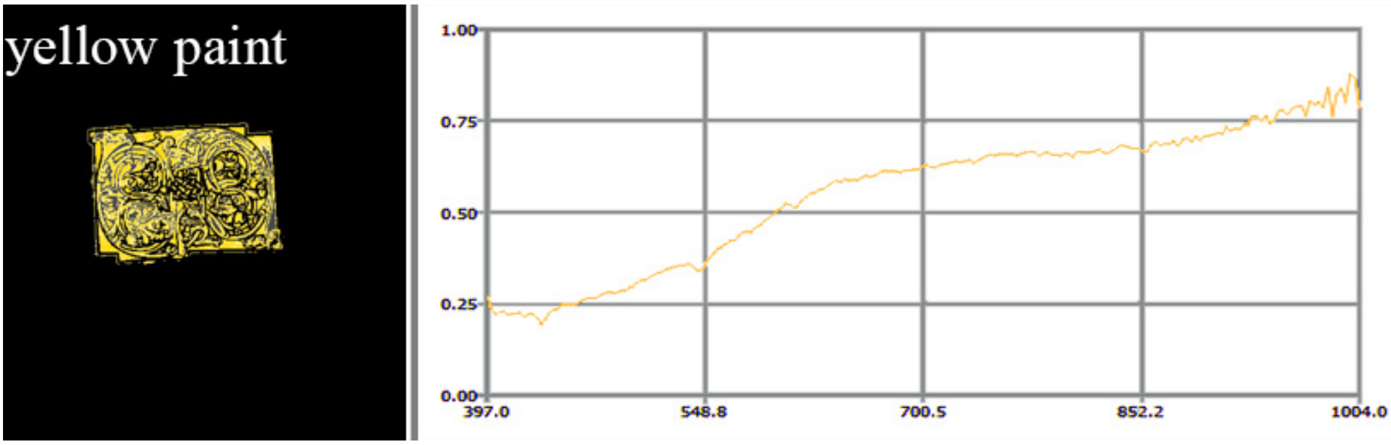

green paint
1.00
0.75
0.50
0.25
0.00
397.0
548.8
700.5
852.2
1004.0


1.00
0.75
0.50
0.25
0.00
397.0
548.8
700.5
852.2
1004.0

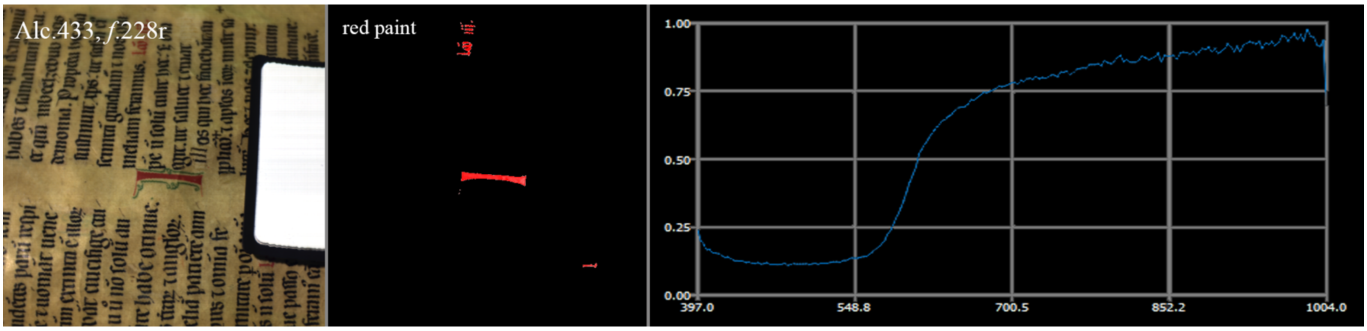
Folio 18v

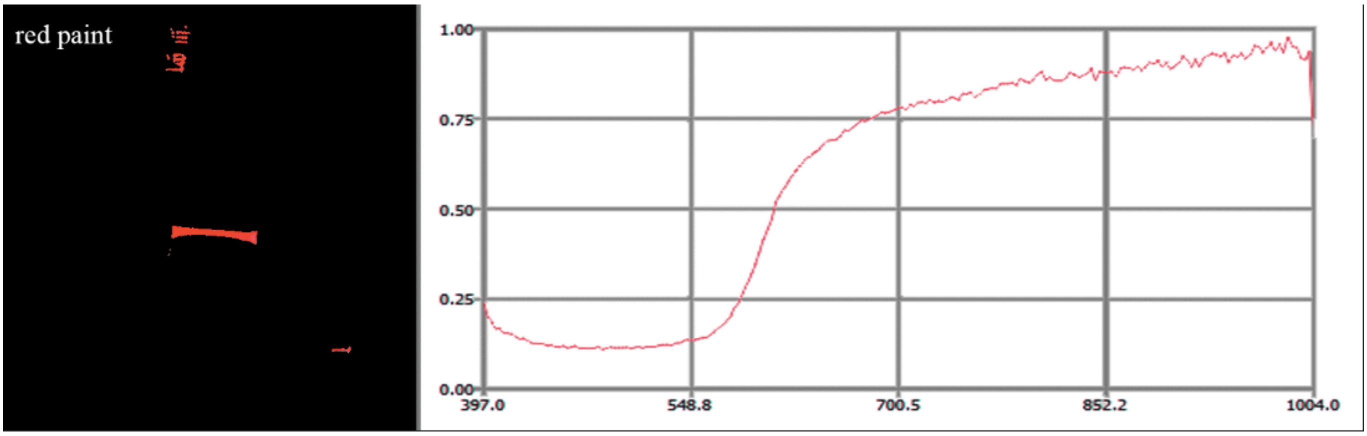

yellow paint
red paint
1.00
0.75
0.50
0.25
0.00
397.0
548.8
700.5
852.2
1004.0


red paint
1.00
0.75
0.50
0.25
0.00
397.0
548.8
700.5
852.2
1004.0

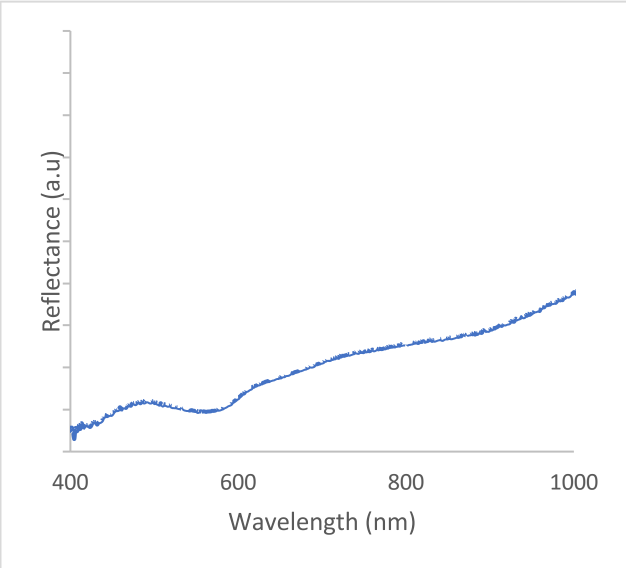

yellow paint
1.00
0.75
0.50
0.25
0.00
397.0
548.8
700.5
852.2
1004.0

Folio 228

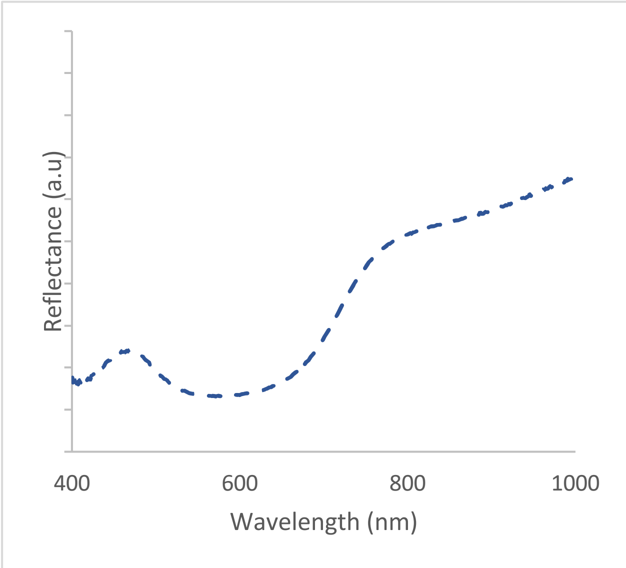

Alc.433, f.228r
red paint
1.00
0.75
0.50
0.25
0.00
397.0
548.8
700.5
852.2
1004.0


red paint
1.00
0.75
0.50
0.25
0.00
397.0
548.8
700.5
852.2
1004.0

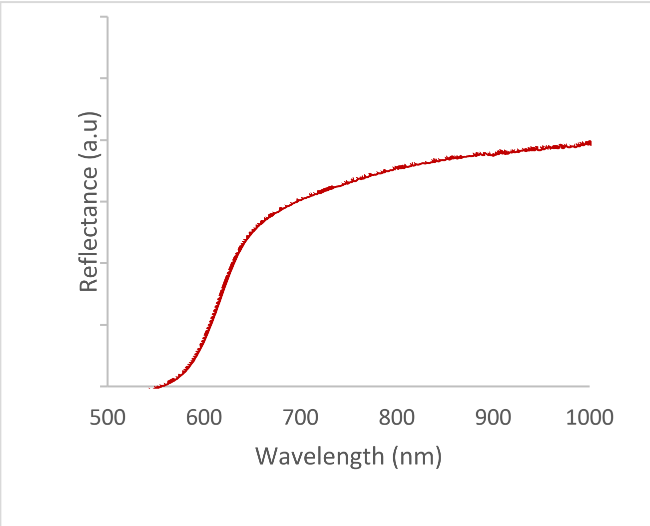
UV-Vis-NIR FORS (Alc. 433)



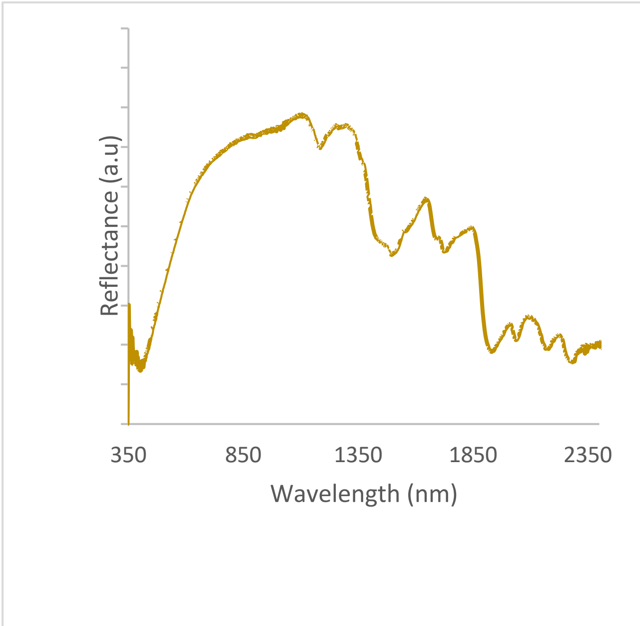
f.1 (blue)



f.18v (blue)

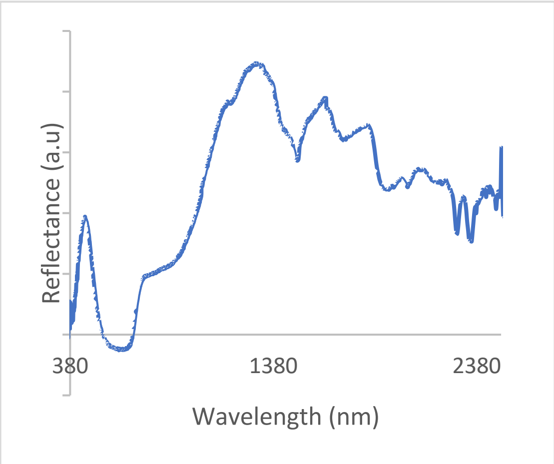


f.228 (red)

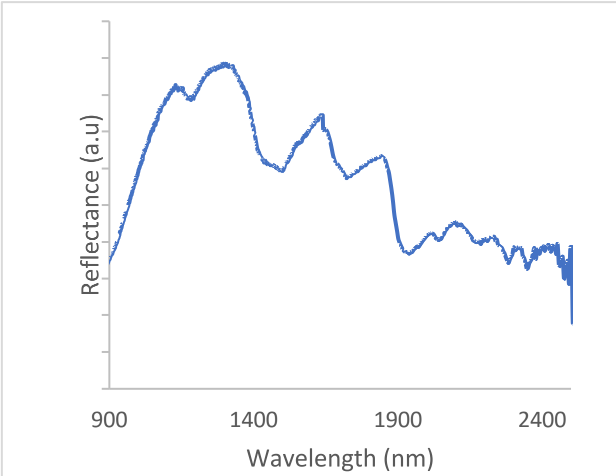


f.18v (yellow)

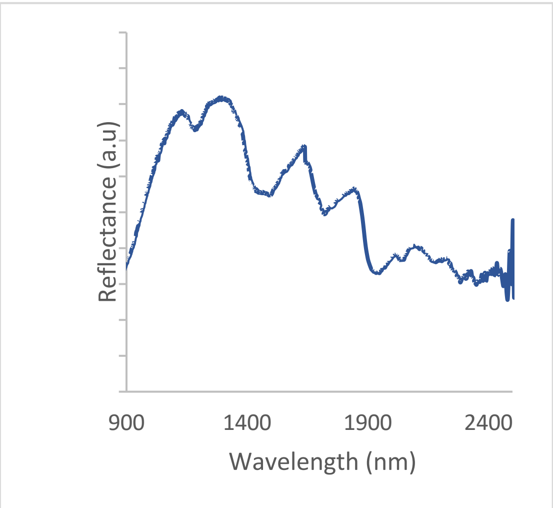
UV-Vis-NIR FORS (Reference)



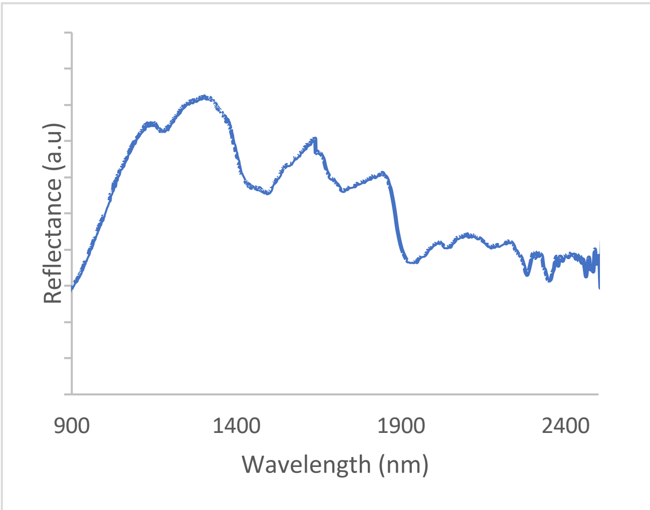
Azurite (parchment glue)



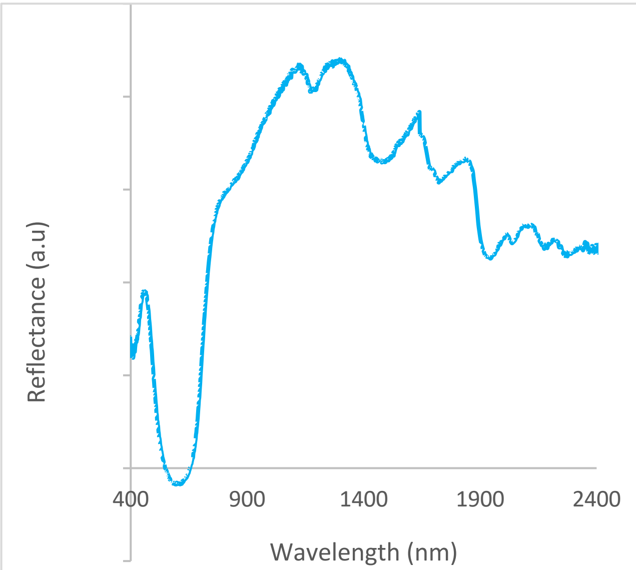
Azurite (egg white)



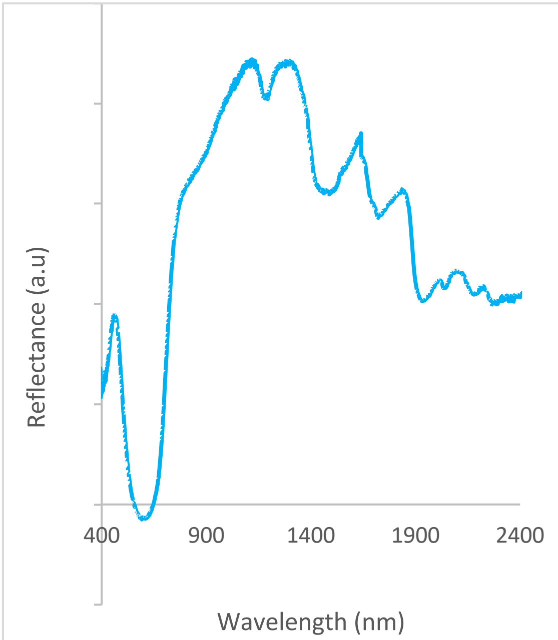
Azurite (egg yolk)



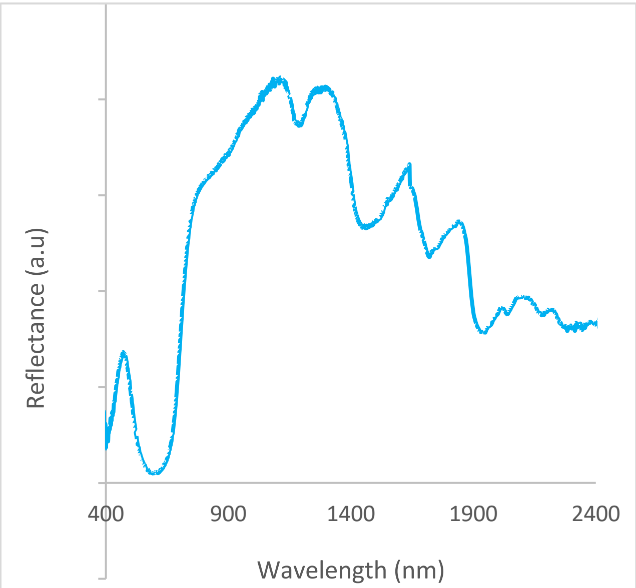
Azurite (arabic gum)



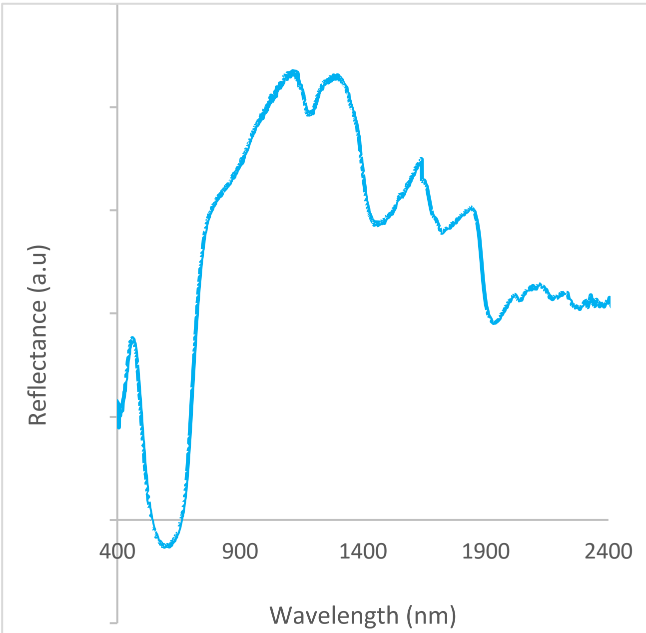
Lapis Lazuli (parchment glue)



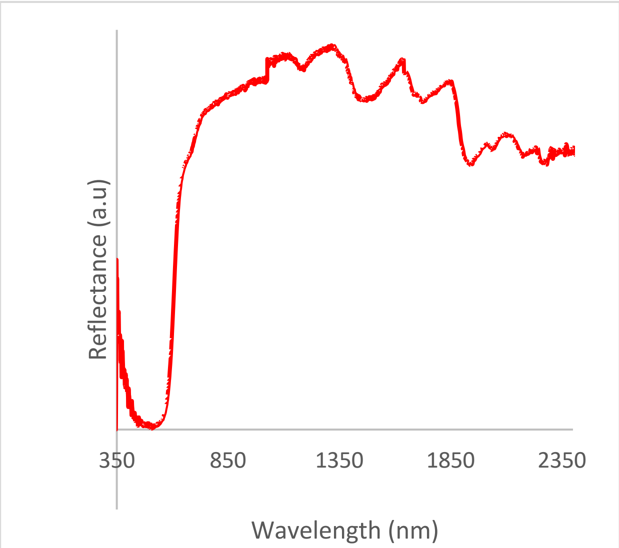
Lapis Lazuli (egg white)



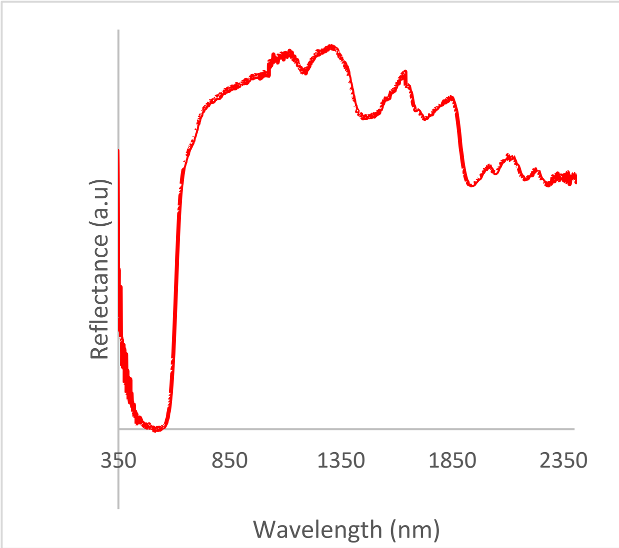
Lapis Lazuli (egg yolk)



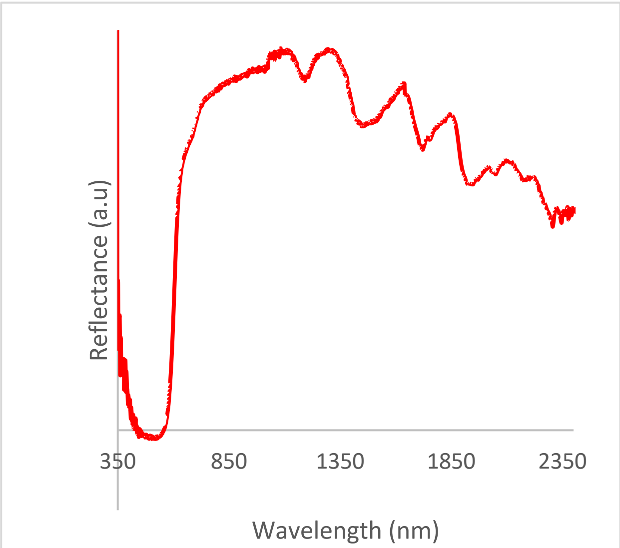
Lapis Lazuli (Arabic gum)



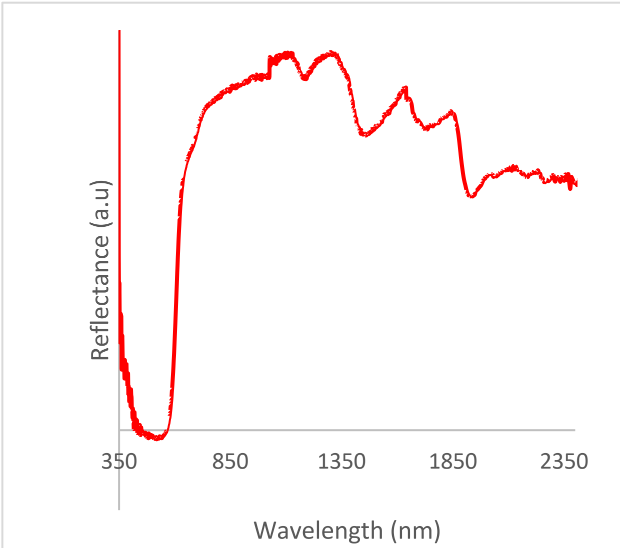
Vermillion (parchment glue)



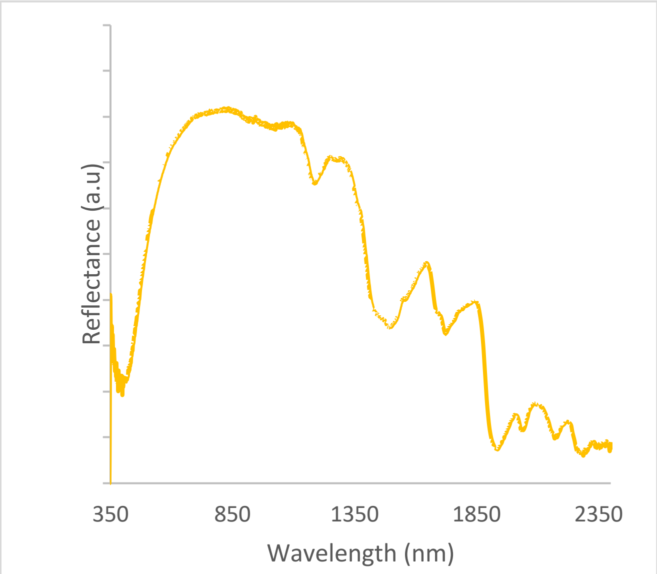
Vermillion (egg white)



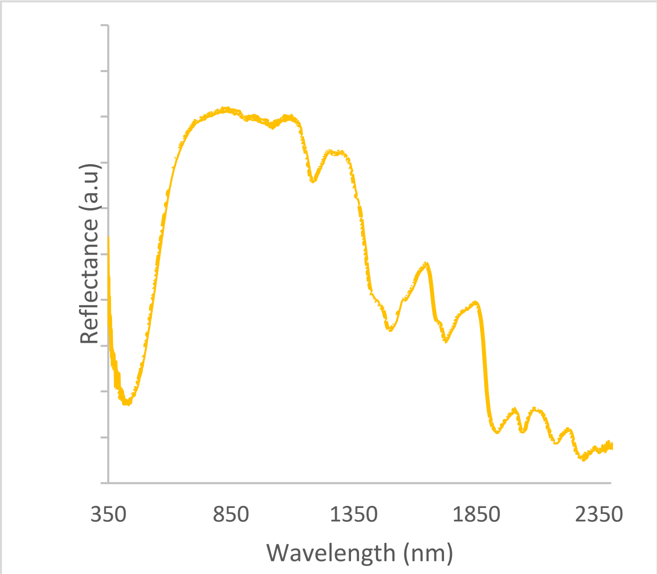
Vermillion (egg yolk)



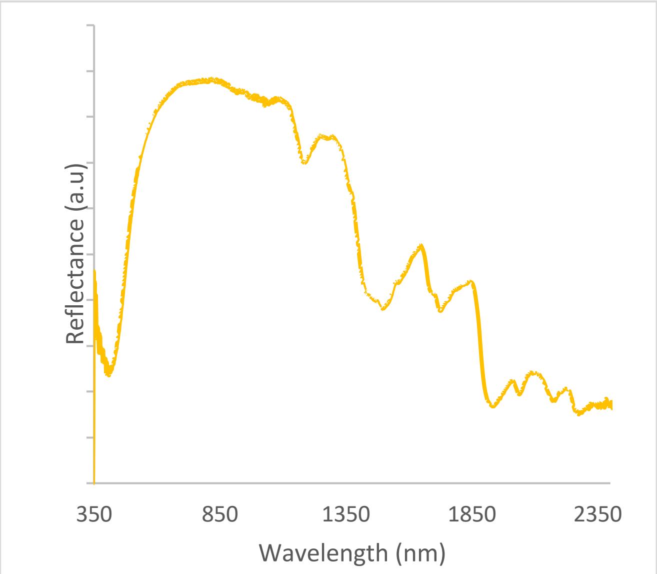
Vermillion (Arabic gum)



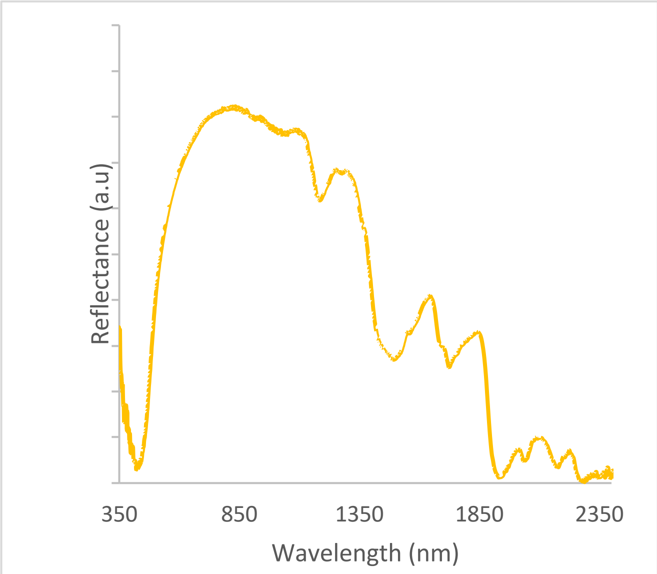
Saffron Yellow



Turmeric Yellow

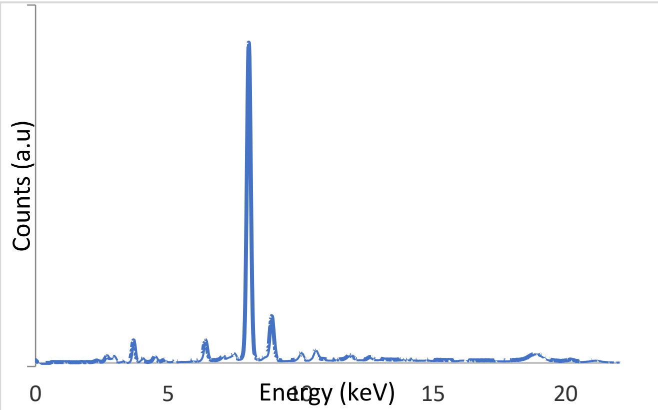


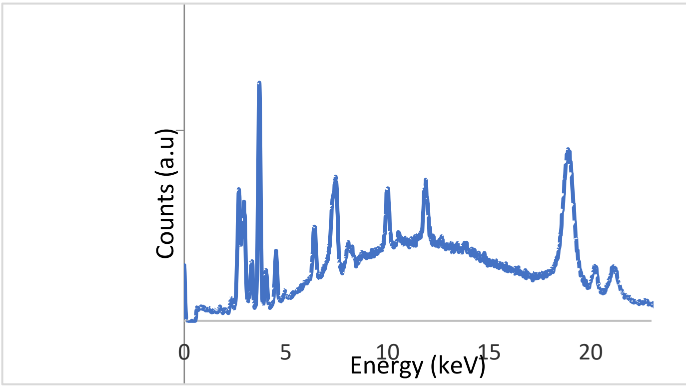
Weld Bolognese Recipe



Weld De Arte Illuminandi Recipe

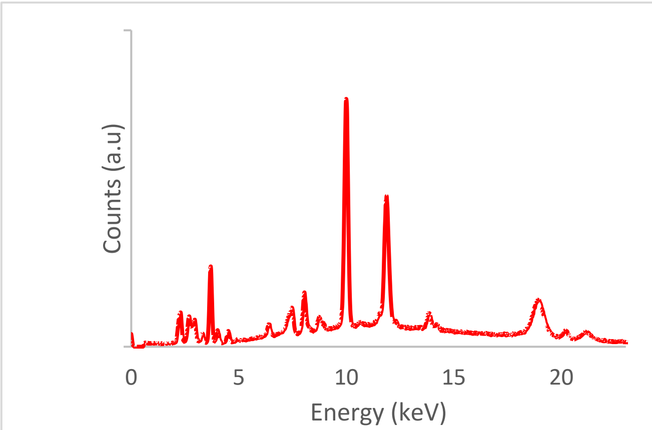
h-XRF Spectra (Alc. 433)

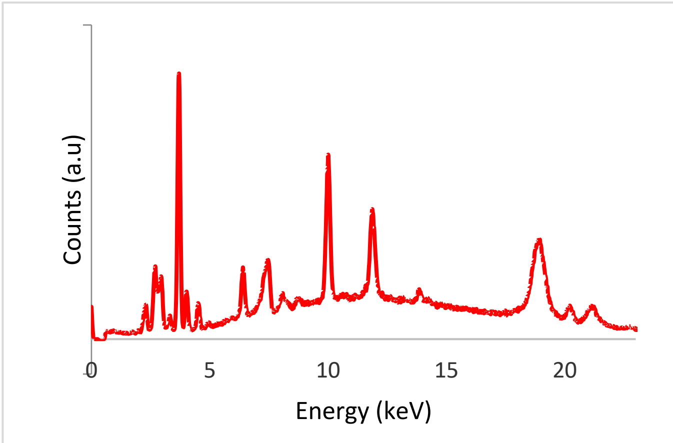




f.18v (blue)

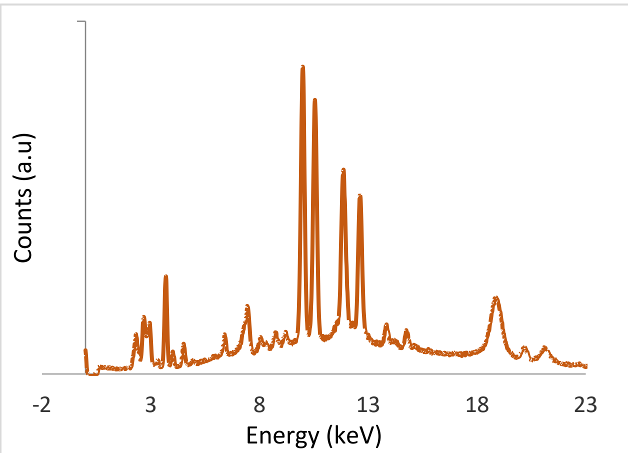
f.1 (blue)

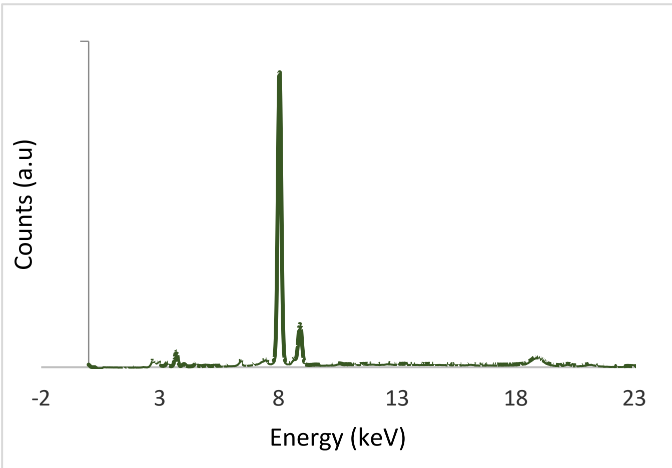




f.228 (red)

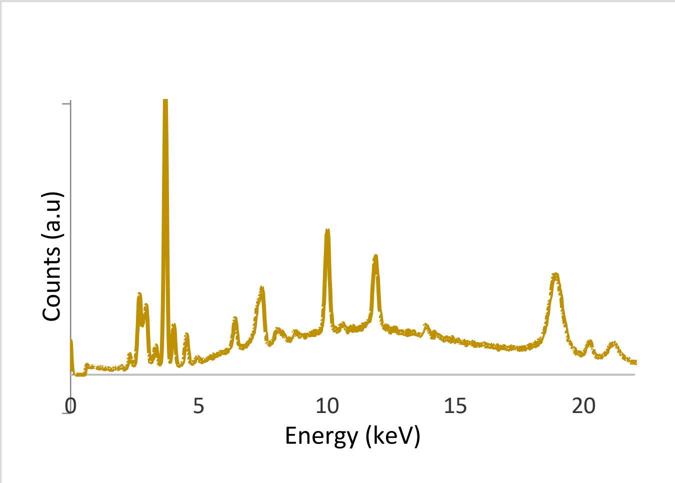
f.1 (red)



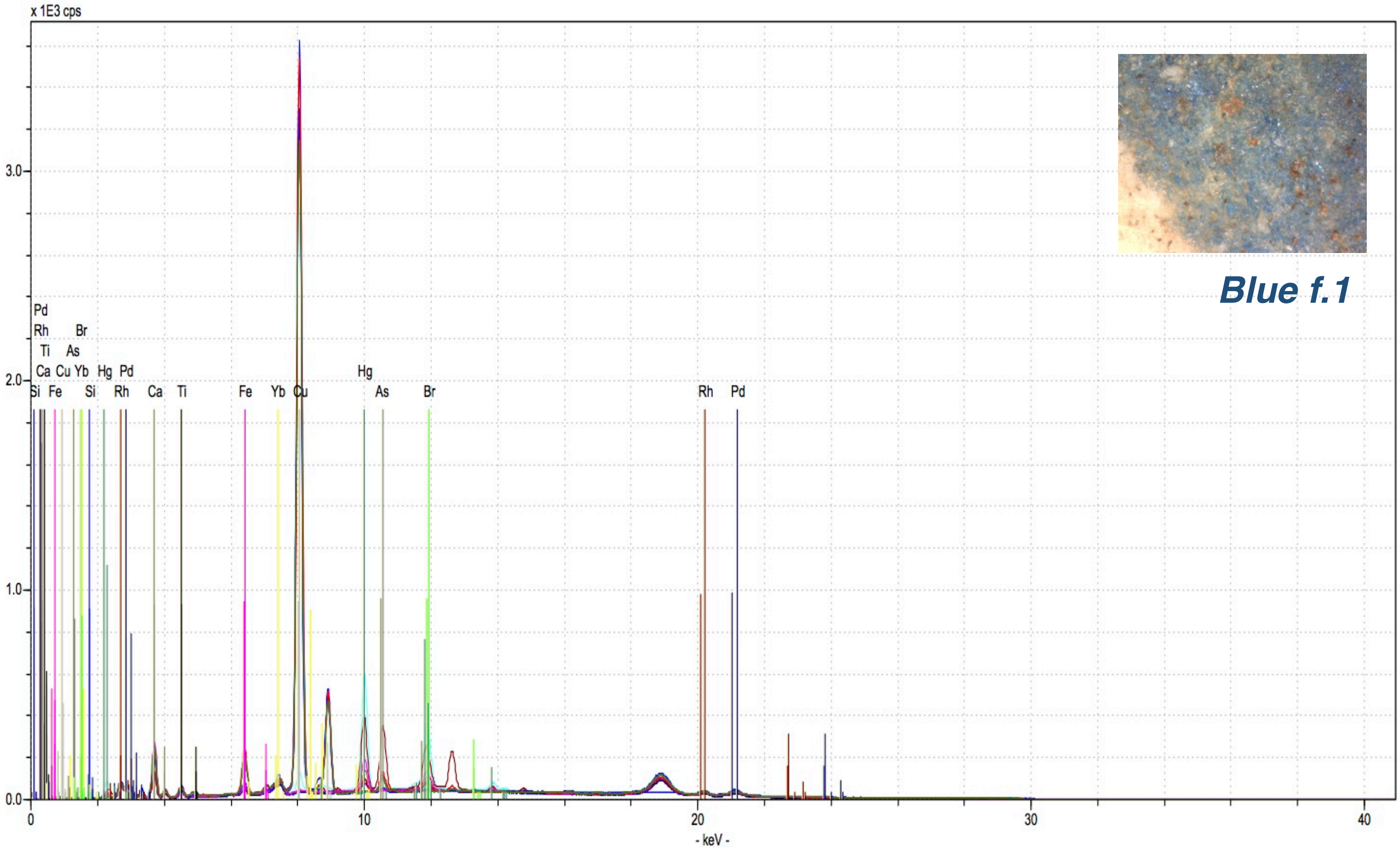


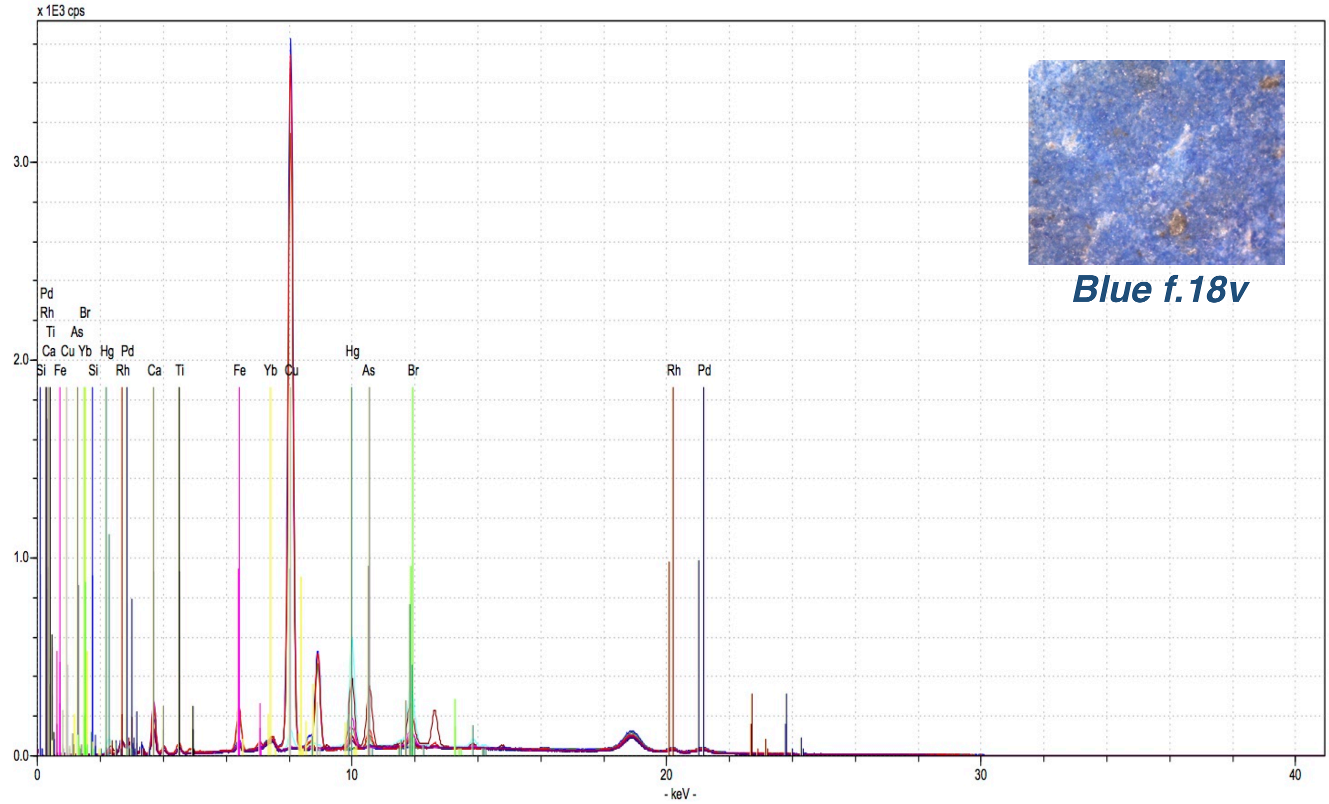
f.15 (orangish red)

f.15 (green)

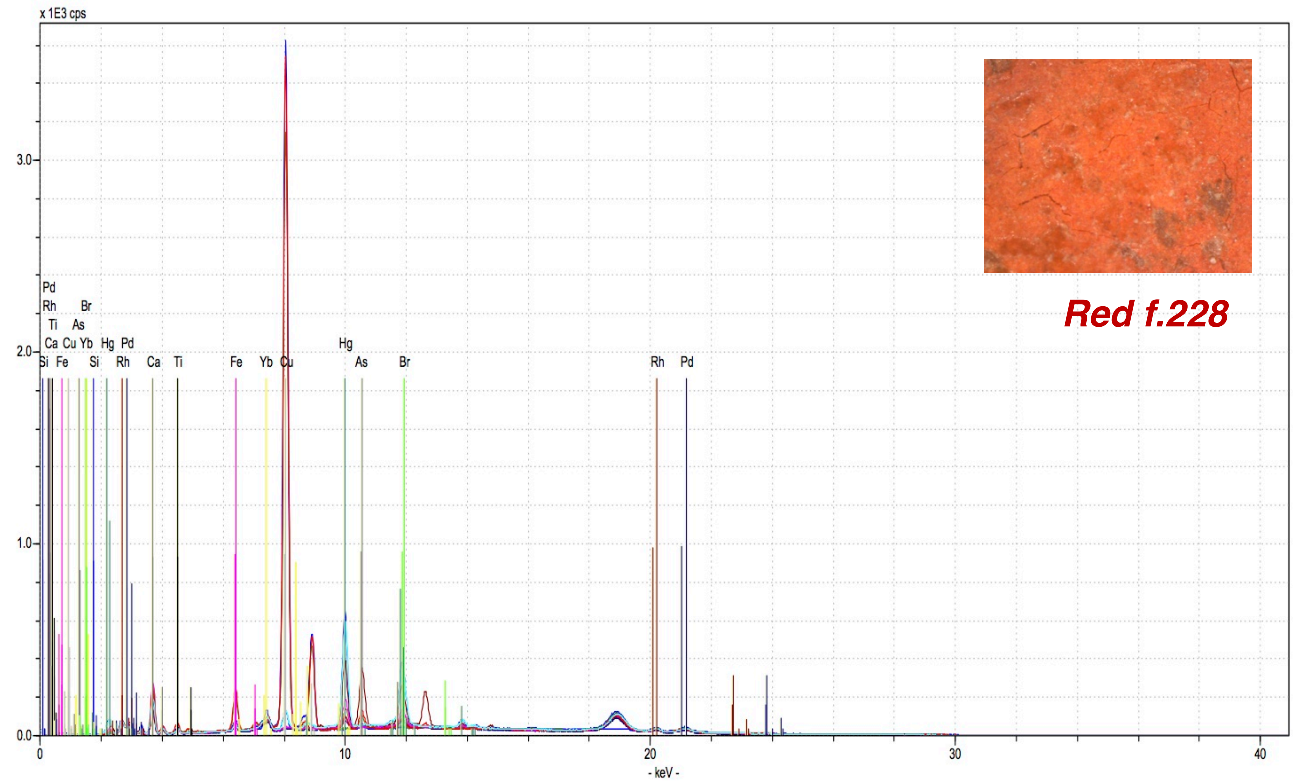

Counts (a.u)
0
5
10
15
20
Energy (keV)

f.18v (yellow)

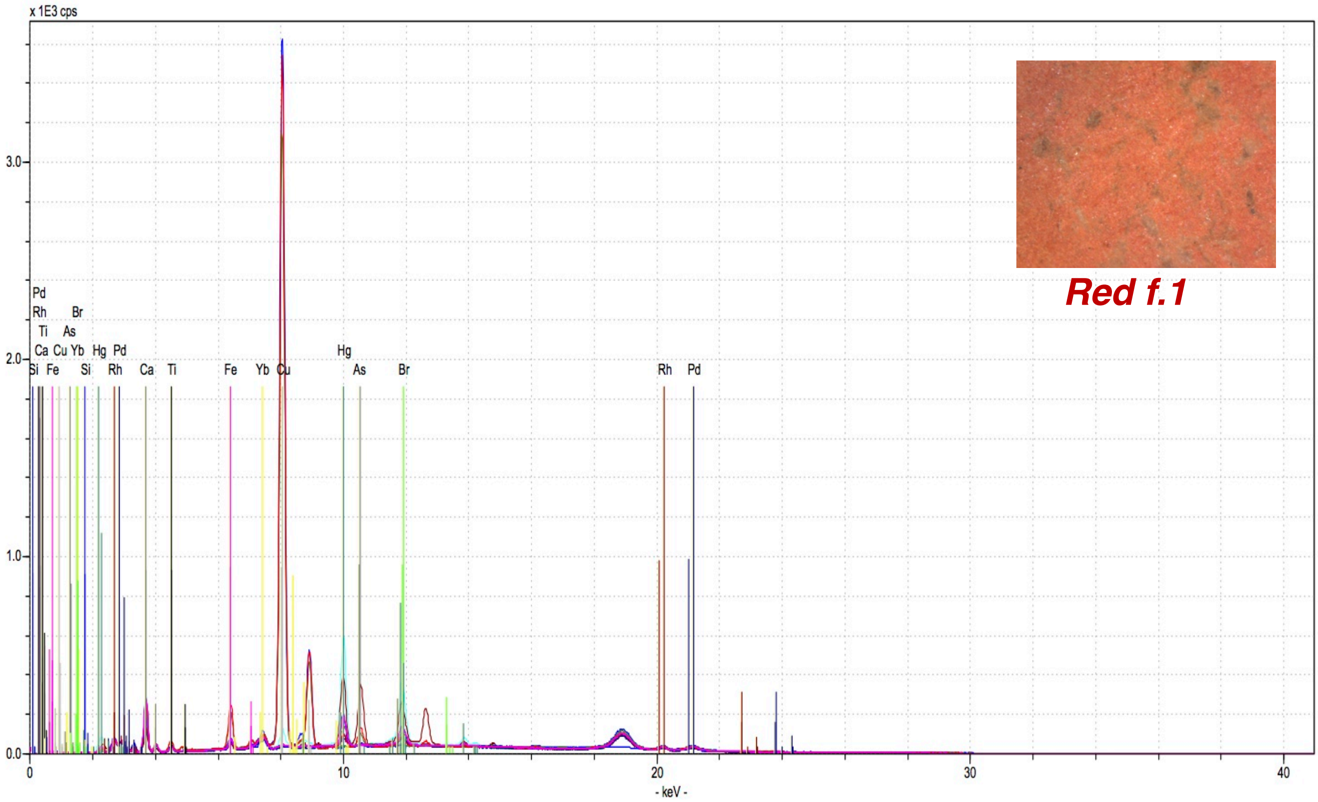

x 1E3 cps
3.0
2.0
1.0
0.0
Pd
Rh
Ti
Ca
Si
Fe
Cu
Yb
Br
As
Hg
Si
Pd
Rh
Ca
Ti
Fe
Yb
Cu
Hg
As
Br
Rh
Pd
0
10
20
30
40
- keV -
Blue f.1

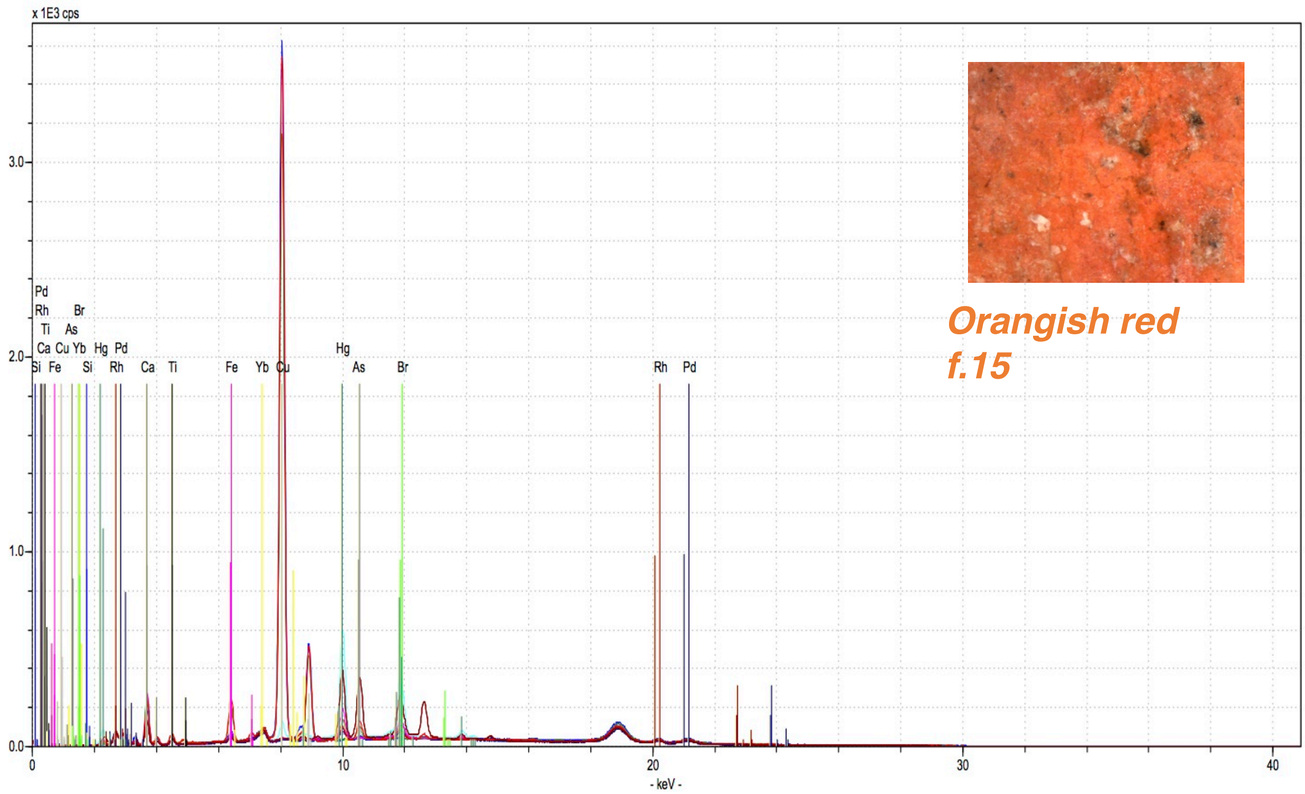


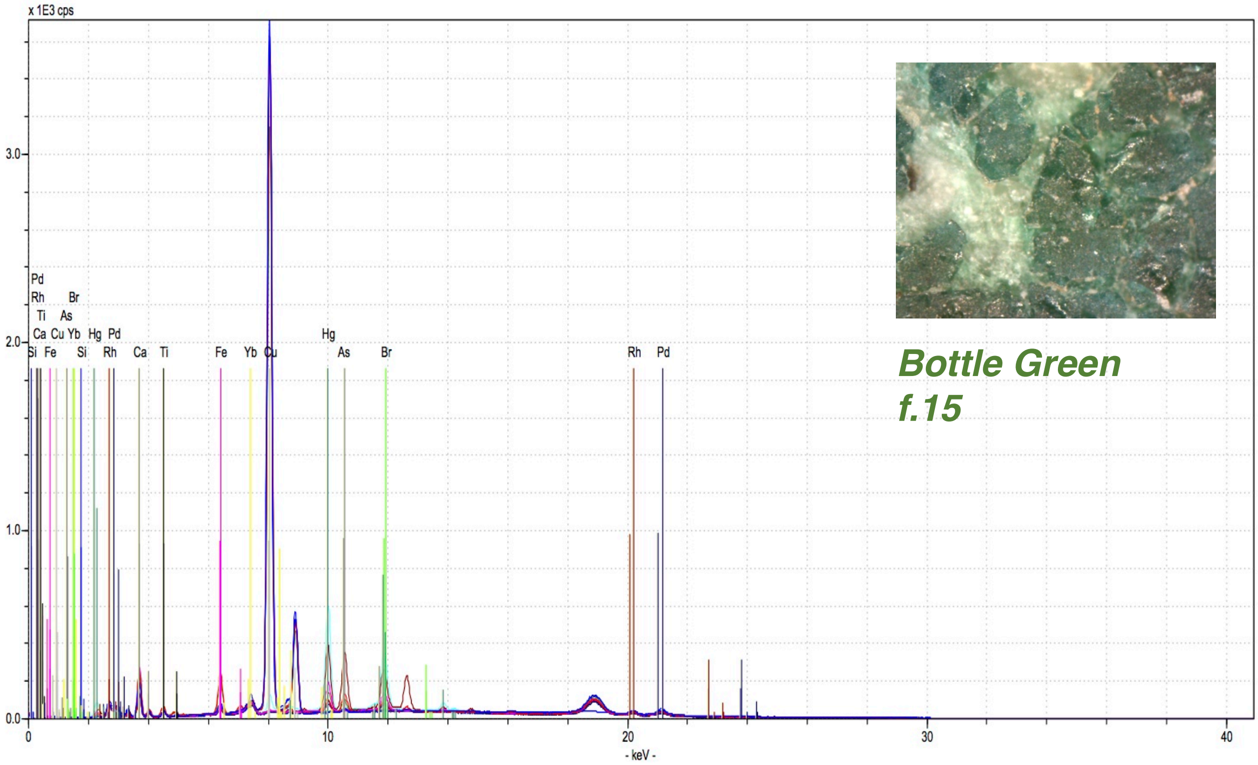
Blue f.18v



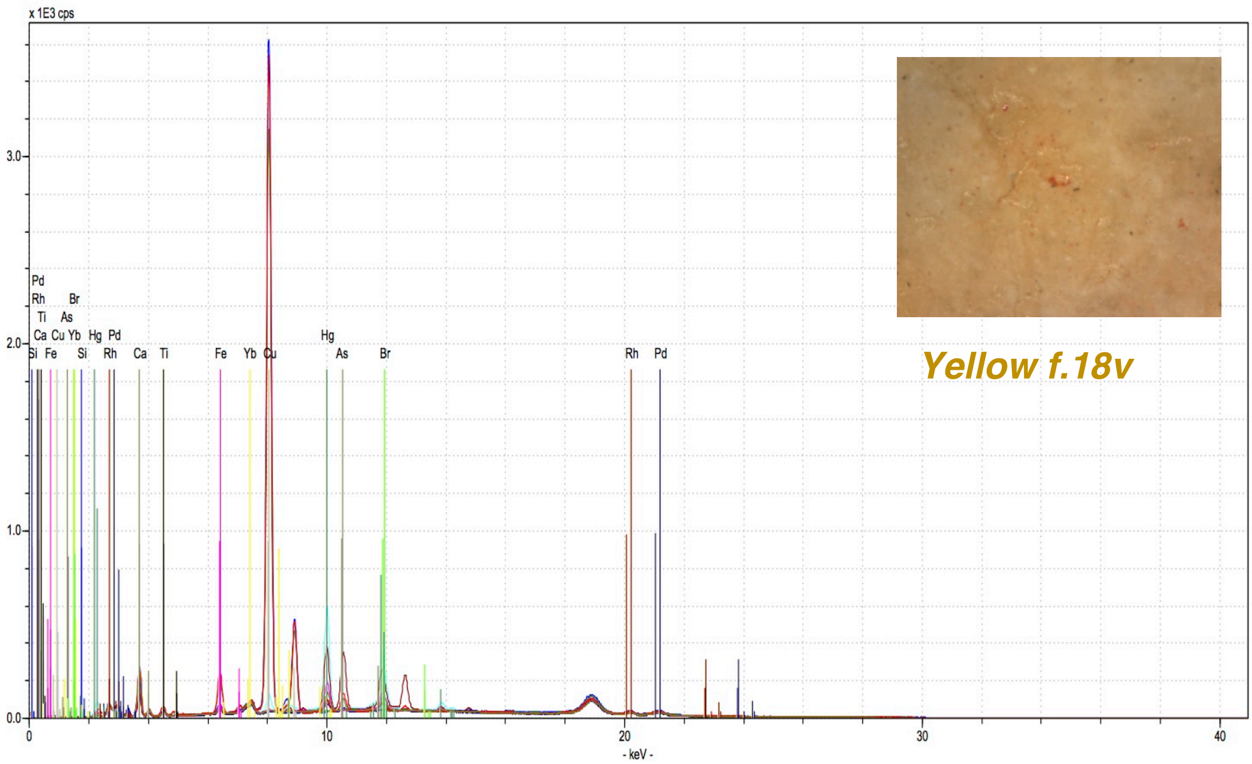
Red f.228


x 1E3 cps
Red f.1
Pd
Rh
Ti
Ca
Si
Cu
Fe
Br
As
Yb
Si
Hg
Pd
Rh
Ca
Ti
Fe
Yb
Cu
Hg
As
Br
Rh
Pd
3.0
2.0
1.0
0.0
0
10
20
30
40
- keV -


x 1E3 cps
Orangish red f.15
Pd
Rh
Ti
Ca
Si
Cu
Fe
Br
As
Yb
Si
Hg
Pd
Rh
Ca
Ti
Fe
Yb
Cu
Hg
As
Br
Rh
Pd
3.0
2.0
1.0
0.0
0
10
20
30
40
- keV -



Bottle Green f.15



Yellow f.18v

Appendix IV: Yellow Lake Pigment Reproduction

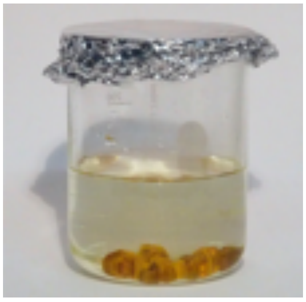
Turmeric Lake Pigment

From *De arte illuminandi* (Naples, Nat. Lib., Ms XII.E.27), Chapter 6, *De glauco.*

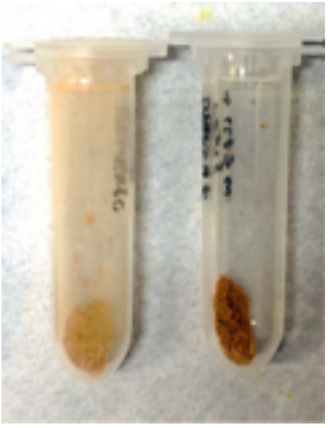
Original Recipe

*"Artificial yellow is made in many ways: first, as mentioned above, from the root of turmeric; or madder grass, also called dye weld / rocket. This pigment is prepared as follows: take 1 oz. turmeric roots, well cut and minced finely with a knife, then pour 0.5 pt. pure water in a glazed, fire-resistant vessel and add **0.125 oz. of potash alum**. Soak the roots in immersion and let them soften over a day and a night until they turn bright yellow. Add 1 oz. of thoroughly ground **lead white**, mix the liquor with a wooden stick as long as it heats on the fire, always stirring with the stick so that the solution will not overflow as it froats. Subsequently, [put a cloth to cover another] non-glazed earthenware vessel and filter the solution through the cloth. Let [the solution] stay [on the cloth] in this way, then remove [more] water carefully [by mixing the plant remains], subsequently put the [filtered] liquid aside for your work. [Otherwise, arzica] is similarly prepared with another herb called dyer's weld / rocket. Take accordingly a handful of this herb and mince it duly with a knife and put it in ordinary water, or in sufficiently concentrated lye, ensuring that the water or lye far exceeds the herb in quantity; boil for a while, then, if there is more than a handful of herb, add 1.5 oz. more of finely ground **lead white**. But before adding [this ingredient], however, grind finely 1 oz. of potash alum, add to the mentioned vessel filled with the liquor of that herb and dissolve in the solution; once dissolved, sprinkle the **lead white** powder over it, stirring constantly with the stick until all those substances form an homogeneous solution. After doing so, [filter the solution in the same way as before,] by placing a cloth on the top of a vessel of unglazed earthenware, let the solution sit, then throw away the water. Pour later additional pure water [and let the solution settle again]. When a [good amount of precipitate will have formed on the bottom of the vessel] throw away the water and let [the formed powder] dry and keep aside. Alternatively, **lead white** can be similarly incorporated with saffron. And note that if the colour [of your pigment] is mild, you can repeat the process. If, [on the other way around], it becomes excessively coloured, you can add more **lead white**"*

Observations

Steps	Observation
120 mL of purified water is added by potash alum, and heated for 30 minutes within 80° C	$m_{alum(s)}$ = 0.2291g pH of alum solution = 4 Alum solution is colourless Potash alum is acidic solution with vicinity pH=4.3 with main hydrated species of Al^{3+} The heating is a catalytic process to fasten the chemical reaction.
Place ***Curcuma longa*** and leave the solution for 24 hours	The solution turned into clear yellow in instance. After 21 hours, the yellow is very intense and bright pH of curcuma-alum solution = 4-5 5 minutes:  21 hours:  Curcumin, chemically called by diarylheptanoid [2], has natural phenol which corresponds to its appearance of

	bright yellow (acted as the chromophore, detected within 270 nm on UV characterisation). However, in different pH, curcumin will appear as different colours. Theoretically, in acidic solution it appears as brownish and reddish. Potash alum works as mordant to turn the dyarylheptanoid as complex compound.
Curcuma-alum solution is added by lead white, and being heated for 30 minutes within 70° C	$m_{lead\ white(s)}$ = 2.0005 g The solution turned into solid bright yellow and denser than before. pH of final pigment solution = 7  Lead white is used to create deeper and colour of the pigment.
The final pigment solution is filtered and desiccated, resulting the pigment powder of turmeric.	The first filtration did not work. Filtrate was still containing of yellow pigment that visible with naked eyes. Both, filtrate solution and solid residue were re-filtered 3 times,finally resulting pigment powder of turmeric as desired.

	
Result	The pigment in two flasks have different tone of yellow. One is lighter and another one is darker. The lighter one is assumed because the appearance of lead white. 

Weld Lake Pigment (Bolognese Recipe)

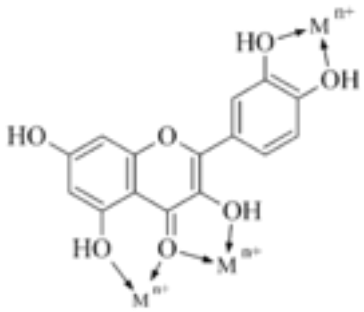
From *Bologna manuscript* (Bologna University Library, Ms 2861), Chapter 7.16, *Per fare l'arzica bella e buona*

Original Recipe

"Take one oz. of fresh weld which is also used by dyers, and mince it finely, then place it in either a glazed vessel or a tin vessel, then pour enough water to covers the herb so prepared and boil the solution until the herb is reduced to half of its original volume. Should too much liquid fade away in the process, keep on pouring much new water to let the solution keep on boiling, not more. Then add 2 oz. of finely ground travertine powder; otherwise, [you can use] 2 oz. of lead white and 0.5 oz. of very fine potash alum. Add immediately such materials to the mentioned vessel, before the water gets cold and little by little – However, keep on stirring the solution, then take it out of the fire; when it gets almost cold, you can drain the water. Then take a new brick with a hole in the centre, place the [precipitate of the solution in it, which is your] arzica. Let [this compound] rest inside for some time, then place it on a well-clean board to dry. [So your pigment] is done."

Observations

Steps	Observation
126 mL of purified water is added by ***Reseda luteola*** (weld), and heated until reached half of the volume within 80° C	m_{weld} = 2.0081 g pH of weld solution = 7 The weld solution is bright brownish clear Before (left) and after (right) heating: 
Potash alum and lead white are added immediately to weld solution during the stirring	$m_{lead\ white}$ = 0.2505 g $m_{potash\ alum}$ = 2.0017 g The solution turned into bright and intense yellow pH of weld solution = 5

	The solution is more acidic caused by the addition of potash alum (with pH theoretically around 3-4). The solution appeared with more intense yellow colour because the forming of complex (chelation). Alum is the metallic mordant to form flavonoid metallic complex. Possible complex (with $M^{n+}=Al^{3+}$): 
The final pigment solution is filtered and desiccated, resulting the pigment powder of weld.	The first filtration did not work. Filtrate was still containing of yellow pigment that visible with naked eyes. Both, filtrate solution and solid residue were re-filtered,finally resulting pigment powder of weld as desired.
Result	The pigment in bright yellow and it is assumed because of lead white. 

Weld Lake Pigment (De Arte Iluminandi Recipe)

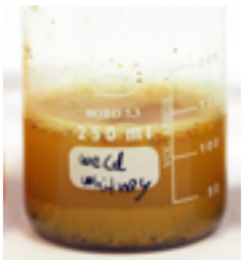
From: Jacobs, W. (2016). *The fingerprinting of the materials used in Portuguese illuminated manuscripts: origin, production and specificities of the 16th century Antiphonary paints from the Biblioteca Pública de Évora.* Évora- Thessaloniki-Rome: Universidade de Évora-Aristotle University of Thessaloniki-"La Sapienza" University of Rome, p. 29 - *Weld and Buckthorn Lake Pigments*;

Kirby, J. et al. (2014). *Natural Colourants for Dyeing and Lake Pigments: Practical Recipes and their Historical Sources*. London: Archetype.

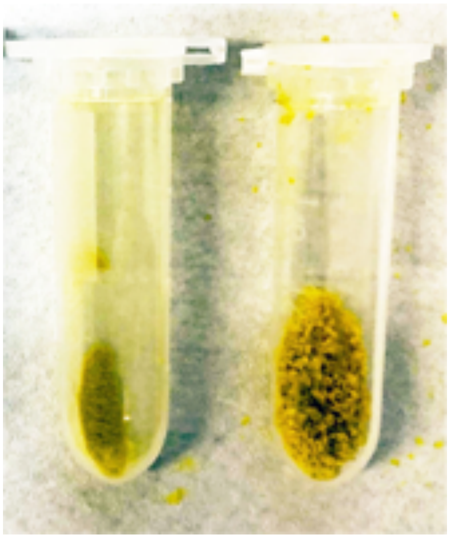
Original Recipe[64]

"It is also made in a similar way out of that dyiers' weed." Take this and cut it up ne with a knife, and put it into plain water or fairly strong lye," and have the water or lye stand over the weed in good measure. Boil it hard for a while; then, if there is one handful of the plant, put in an ounce and a half of white lead, well worked up. But, before you put in the white lead, work up one ounce of rock alum thoroughly, and put it into this dish with the decoction of the plant, and get it dissolved. And when it is dissolved, add the white lead gradually, constantly stirring it with a stick until all those things are thoroughly incorporated. And then strain it through a linen cloth into an earthenware porringer, red but not glazed, and let it se le. And pour o the water, and again put in some clear plain water. And when the substance se les pour o the water; and let it dry, and put it away."

Observations

Steps	Observation
120 mL of purified water is added by ***Reseda luteola*** (weld) and being settled for 24 hours	$m_{weld\ (s)}$ = 2.0024 g The weld solution is bright brownish clear, and turned into intense yellowish clear after 21 hours
Weld solution is heated for 30 minutes within 80° C	pH of weld solution = 6 the colour looks more intense and darker than before 

[64]The recipe used was the original recipe combined by other references above considering the result and the purpose of comparation between the use of lead white and without lead white.

The weld solution is added by potassium carbonate	m $_{potassium\ carbonate\ (s)}$ = 2.0005 g pH = 10 Potassium carbonate is a basic solution with pH around 10. The potassium solution is used to dissolve the pigment, since the pigment does not dissolve in water.
Alum solution is made by heating for 30 minutes within 80° C, and being added gradually to weld solution	m $_{alum\ (s)}$ = 2.0008 g pH of alum solution = 3 pH of alum + weld solution = 8 Alum acts as mordant to precipitate the weld pigment and chelating the pigment as complex
Result	The pigment in darker yellow than weld in bologna recipe, and assumed because of the lack of lead white. 

Appendix V: Historical Reconstruction of f.18v

Pigments, Binders, and Ink

- Ultramarine Blue Pigment Powder (from Winsor & Newton)
- Vermilion Red Pigment Powder (from Kremer Pigmente)
- Yellow Lake Pigments (reproduced following the historical recipes)
- White Egg
  Following the recipe in *De Clarea* (11th – 12th century) [Thompson 1932, p.19]:

  "So when you are ready to prepare glair, you separate the white of the egg from the yolk; and placing the white on a platter you beat that white of egg strongly, unintermittently, with a little stick described above, until it is converted, as it were, into a water-froth, or into the likeness of snow, and sticks to the platter, and looses the power of running or shifting in any direction, even of you turn it bottom-side-up, that is, the bottom of the platter on the top and the glair underneath. (...) After the white is whipped, put the platter in a quiet and clean place, slanting it a little, so that the glair-liquid mail distil from the froth. (...) Now when the glair liquid has distilled, and you have cleaned the shell of the egg, pour the same liquid into the shell."

  Reproduction: Egg white was beaten with a fork and allowed to stand for 6 hours. The deposited serum was used as binder. Eggs were acquired in a local market.

- Yellow Egg (already prepared as binder)
  Following the recipe in *De Clarea* (11th – 12th century) [Thompson 1932, p.71]:

  "Put the yolk of an egg, separated from the white, upon a platter, and adding water, so that the yolk may loose its thickness in the water, and become lighter, you beat it with the little whisk almost like the other glair, until it is all whipped."

  Reproduction: Egg yolk was diluted in water (1:1, v/v) after withdrawal from its sheath. Eggs were acquired in a local market.
- Arabic Gum (alredy prepared as binder)
- Parchment Glue

  Following the recipe in *O Livro de Como se fazem as cores* (Portuguese, attributed to the 13th -14th century), Chp.40 [Strolovitch 2010, p.235]:

  "If you wish to make glue, take two pieces of parchment and wash them very well, and then place them in a new pot, and heat them vigorously until they are well boiled. And once the first water has disappeared, add more water into it, and as soon as you wish to try [it], take a little of it and place it in your palm. And place one hand with the other, and if your hands grip, it must be that your glue is well made."

  Reproduction:
  For producing parchment glue, *circa* 3.5g of calf parchment (cut into small squares of 0.5cm side) was boiled in 50mL of water (80-90oC) for 4 hours. After that, water was allowed to evaporate, until a sticky consistency was obtained. Calf parchment was acquired from *Museé du Parchemin* (Rouillon, France).

- Iron Gall Ink

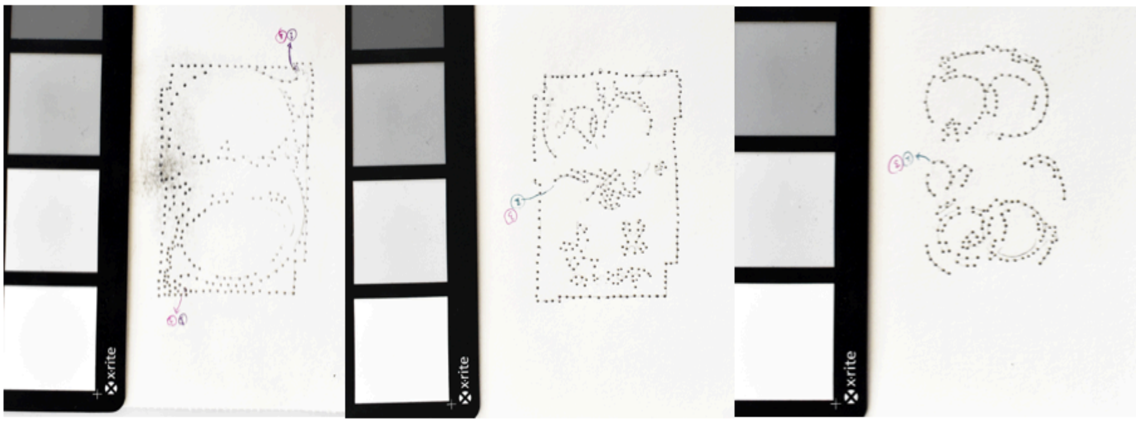
  Iron-gall ink was reproduced using gallnuts (Zecchi, Italy), a homemade wine, an iron salt (iron sulphate - Aldrich, Germany) and Arabic gum (Zecchi, Italy), following the recipe described in the Padova manuscript [Merrifield 1999, p.676].

Sequence Steps

1. All parchments are on the *recto* side, and marked to the measurement of the distances between writing ink, incipit, and initial letter space (according to the real scale)
2. Carefully write the text part with iron gall ink in each parchment, leave approxiametly 2-3 hours to let the ink oxidised and dried completely
3. Sketch and paint the initial letter according to the chosen tracing technique on the parchment
4. Write the *incipit* texts

Initial Letter Stencil Tracing Methods

1. 1:1 Scale Initial letter of f.18v is punched following the line of (1) S frame (2) body S and (3) middle body S. Each in 3 different art papers.



2. Using lead-point stick, mark the punch one by one with parchment under the *punched* art paper
3. Sketch the initial letter following the mark of the punched dots using lead-point stick
4. Carefully paint the initial letter in order: yellow, red frame, red body S, and red ornaments

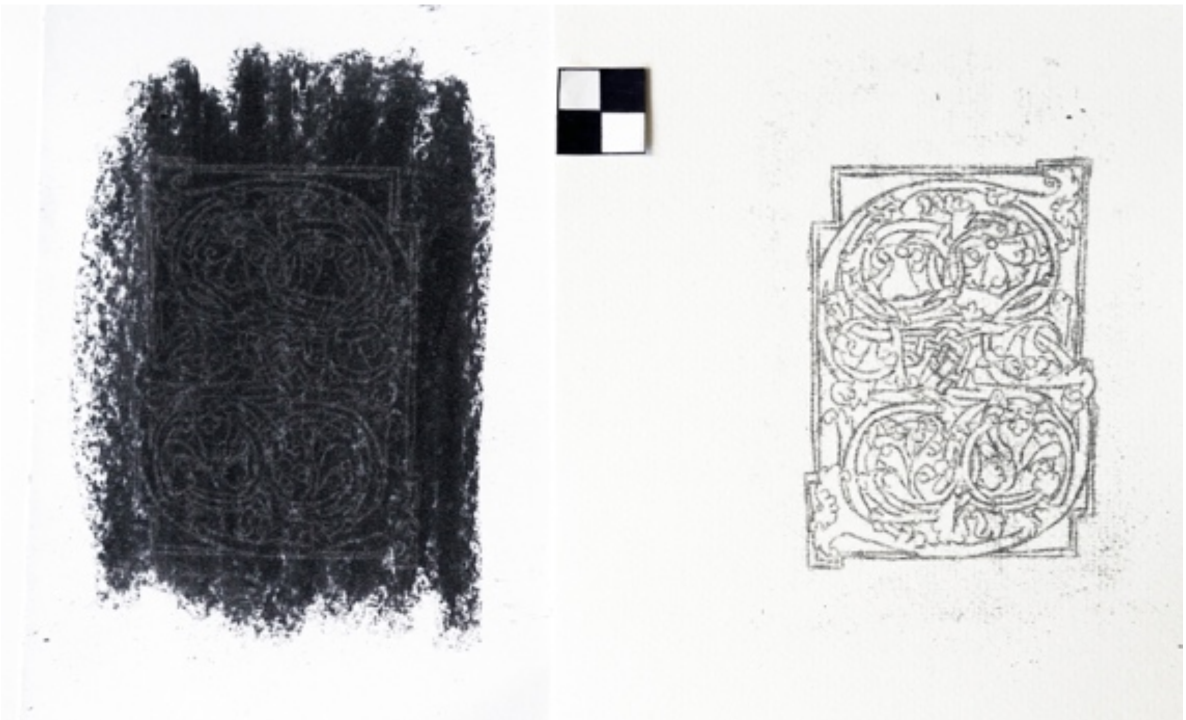
Initial Letter Freehand Methods

1. Measure the distance between the S frame, body S, and the ornaments on the 1:1 Scale Initial Letter
2. Mark the distance using lead-point stick on the parchment and sketch it meticulously
3. Carefully paint the initial letter in order: yellow, red frame, red body S, and red ornaments

Intitial Letter Carbon Copy Methods

1. Cover the text part of the parchment
2. Spread carbon powder on the art paper; face it on the parchment side

3. Put the 1:1 Scale initial letter; and trace carefully



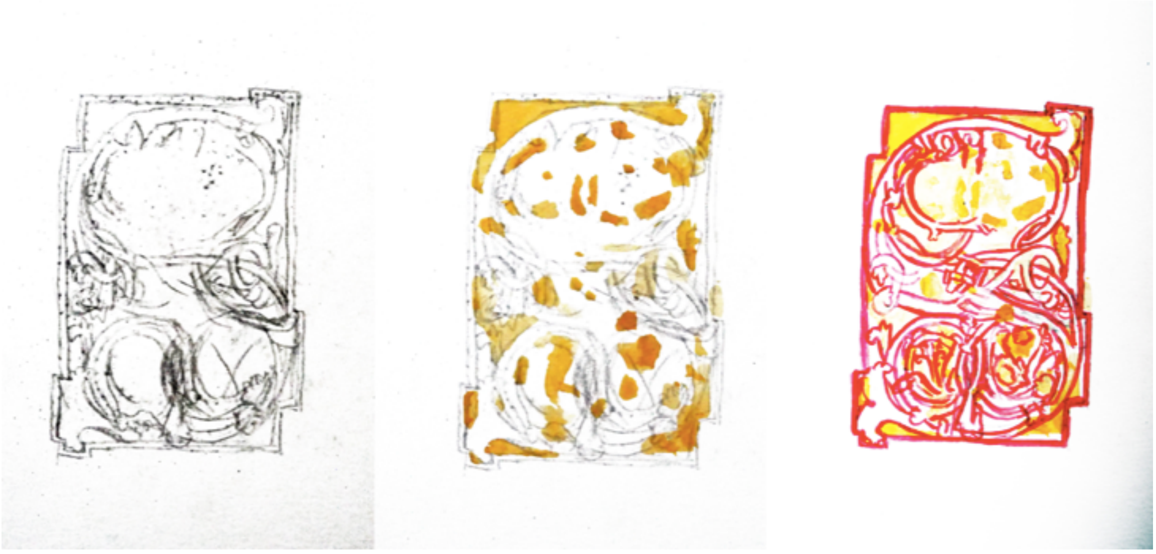
4. Take the art paper carefully
5. Carefully paint the initial letter in order: yellow, red frame, red body S, and red ornaments

Intitial Letter Backlight Methods

1. Place the desk lamp beneath the glass desk and manage the room as dark as possible
2. Put the cut 1:1 Scale of original letter on the glass desk; right on the direction of the light of the lamp

3. Put the parchment right on the 1:1 Scale
4. The shade of the light will be perfectly seen and sketch carefully; not to move the lamp or the drawing set up.

5. Put the cut body S art paper on the parchment; start paint meticulously the yellow



6. Paint the red in order: red frame, red body S, and red ornaments

Biography of the Author

Shatila Jihadiyah Fitri, usually known with her mother's family name: *Shatila Algaff*, was born in Bangkalan, a small town in the island of Madura, East Java, Indonesia. During her studies in Chemistry Department of Institute of Sepuluh Nopember Surabaya, she figured out the branch of chemistry she treasures: materials chemistry. Since then, she started to be keen to focus in this branch. During her bachelor studies, she was awarded with Outstanding Student Scholarship from Research and Higher Education Ministry of Republic of Indonesia (2013-2014), and General Electric Foundation Scholarship and Future Leaders Scholarship award from International Education Foundation, based in New York (2014-2016). After her bachelor graduation, she was offered in several scholarships and master programmes focusing in Materials Science and Engineering in several countries in Europe, nevertheless, she chose to be a part of Erasmus Mundus Archaeological Materials Science (ARCHMAT), following her big passion in history- especially Middle Ages. Thanks to the influence of her father, who has a background in Islamic Education, she grew up with the story of Sultan Saladin and King Richard that led her to be keen to study about the Middle Age, thus, this research which present the valuable manuscript from middle age of Europe- is one of her biggest dream that she finally obtained.

As a young and dedicated Indonesian, Shatila pledged to contribute to her beloved country through the field she loves: cultural heritage and its preservation through proper education. In 2015, alongside her colleagues, she co-founded *Bangkalan Bisa*, a non-governmental foundation for Bangkalan (her hometown) to facilitate Bangkalan youths and people, especially in education.

Shatila still has several checklists and quests for her own personal life purposes and dedication for the Republic of Indonesia. Hopefully, the lists will be all checked, soon.