THE HISTORY OF BLUE IN THE LITURGICAL CODICES OF EARLY ALCOBAÇA AS TOLD BY THE MATERIAL ANALYSES

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Abstract
Mediaeval blue colour has long been the subject of several histories and stories concerning its meanings and sources, especially the use of ultramarine and the introduction/transition to azurite in the art history painting's production. Being the most expensive blue pigment during the mediaeval to renascence period, ultramarine is said to have been selectively used for the most important representations of the painting compositions. In contrast, azurite – a less expensive blue pigment – was used for the less important blue paint representations. The analysis of eleven illuminated Liturgical Codices from the 12th-14th centuries produced in the Alcobaça scriptorium allowed fingerprinting the introduction of azurite in the most important Portuguese scriptorium, not just in terms of the period it occurred in, but on how it started to be used. The holistic approach followed in this study, combining the results gathered from the liturgical analysis of the manuscripts with the in-situ non-invasive analysis of the illuminations (h-EDXRF, UV-Vis-NIR-FORS and Hyperspectral Imaging analysis), allowed screening the use of ultramarine and azurite along circa two centuries of the activity of the scriptorium. Within this, it was possible to identify what appears to be the manuscript of transition at Alcobaça scriptorium concerning the introduction of azurite – the Alc. 167, produced during 1197-1198. This is the ancient manuscript from those which have reached our days, where azurite was first identified, in a remarkable way of use: not in the composition of secondary elements (such as titles or small capital letters) – as referred to in the bibliography of azurite use in Art History – but in the production of the most prominent illuminated initials of the manuscripts, combined with ultramarine blue.

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On the use of colour in mediaeval illuminated manuscripts

Colour plays a crucial role in the interpretation of an artwork. In mediaeval illuminations, the use – or absence – of colour might reflect a special meaning that must be considered. Also, the choice of a specific colour for composing the illumination was not a frivolous choice, not just because of the costs involved in the use of some of the colour sources available at the time, but also because, at the time, linked to colours were meanings that would help on the transmission of the message behind the image. Of all the colours in the mediaeval palette, red, green, and blue were the most frequently used colours to produce mediaeval illuminated manuscripts. For this, understanding its use and meaning strongly contributes to a more accurate reading of the message behind an illuminated manuscript.

A. "So God created Man in His own image" (Genesis 1, 27)

The symbolism of red dates back to the Beginning of Time. According to the Hebrew religious tradition, the first man, Adam, was fashioned from red clay. Thus, the original Hebrew word *adama* means land, man or red. In Latin, *Adamus* translates into "man of red earth".[1] In classical Arabic, red was commonly ascribed to ardour, intensity, and violence.[2] Over time, red symbolises strength, life, and passion for different cultural and traditional backgrounds.

The use of red colour pigments dates back to the pre-historic period when man commonly used red ochres (pigments) to produce red hues, which naturally occur disseminated through rock or earth.[3] Due to its abundance in Nature, it was – and is – the cheapest among all red pigments. Therefore, red ochre pigments were the most frequently used source of red pigments by painters throughout the ages, although not commonly used in manuscript production. It was only with cinnabar[4] (red (II) mercury sulphide) that magnificent bright red hues were achieved, making it known as the "Red King".[5] Plinius (Roman, 1st century) refers to its high price and frequent adulteration with red lead[6] (a cheaper pigment). Until the 11th century, it "remained as costly to cover the page with vermilion as gold".[7] Therefore, the extent of use of this pigment reflected the financial resources of a scriptorium. Aside from the economic aspect of cinnabar/vermilion, the medieval documentary sources never

[1] Anne Varichon, *Colors – what they mean and how to make them* (New York: Abrams Books, 2006), 85.
[2] Varichon, *Colors – what they mean and how to make them*, 112.
[3] Nicholas Eastaugh, Valentina Walsh, Tracey Chaplin and Ruth Siddall, *The Pigment Compendium: A Dictionary of Historical Pigments* (Burlington: Elsevier Butterworth-Heinemann, 2004), 200.
[4] Red mercury (II) sulphide has two sources, one natural and the other artificial. As a natural material, the red mercury (II) sulphide may occur as the red-coloured mineral (cinnabar); as a synthetic pigment, it takes the name of vermilion. See also F. Brunello, [*De Arte illuminandi* and other Treatises on Mediaeval Illumination] (Vicenza: Neri Pozza, 1975); R.J. Gettens et al., "Vermilion and Cinnabar," in *Artists' Pigments: A Handbook of their History and Characteristics*, Vol. 2, ed. A. Roy (Washington and London: National Gallery of Arts and Archetype Publications, 1993), 159-182.
[5] Pillip Ball, *Bright Earth: The invention of Colour* (London: Penguin Books, 2001), 76.
[6] A lead oxide, with chemical formula Pb_3O_4.
[7] Ball, *Bright Earth: The invention of Colour*, 87.

hint at any symbolic purposes associated with those pigments, suggesting that their use was more related to its bright red hue than to some special meanings.

B. "The Lord is my shepherd, I lack nothing. He makes me lie down in green pastures, He leads me beside quiet waters" (Psalm23)

Green has a more substantial commitment to its coeval socio-cultural context than red. Medieval Christendom commonly associated green with instability, unpredictable changes or anything ephemeral.[8] In the feudal world, it was on a *green meadow* where the Lords used to face legal duels. For this reason, green becomes the colour of hazard, game, destiny, luck, money, and chance.[9] Despite this "negative" connotation, other cultures would give other significance to this colour. Green carried an overwhelmingly positive connotation in the Arabic-Islamic civilisation until it became the characteristic colour of the Islamic tradition. According to Arabic-Islamic tradition, when the archangel Gabriel appeared to the Prophet Mohamed, the Prophet was dressed in green, and the archangel's wings were green as well.[10] Moreover, the Prophet's successors wear green turbans, and in Paradise, Allah welcomes the souls of martyrs who fly to him in the form of green birds.[11] Moreover, God made plants green and the sky blue for the inhabitants of the Arabian Desert to make things easier visually for His creatures because these two colours were both so beneficial to visualise.[12]

C. "The garden had hangings of white and blue linen, fastened with cords of white linen and purple material to silver rings on marble pillars" (Est 1, 6)

Of all the three colours mentioned in this section (red, green, and blue), blue is the colour with the most significant change in its symbolism throughout history and art history. The Romans ascribed blue to the enemy: the Insular Celts used to dye their skin with blue paint before going to war, providing them with a ghostly appearance, which terrified the soldiers of the Roman Empire.[13] In Christianised societies, the increased devotion to the Virgin Mary and the fact that the Virgin Mary's mantle began to be painted in blue strongly contributed to the inversion of blue's paradigm. The reason for this change has several justifications from the authors who write about colour, its history, and symbolism.[14] Blue is the colour of the sky, and therefore, it has been ascribed to heaven. For some authors, blue became the colour of

[8] Michel Pastoureau, *Vert-histoire d'une couleur* (Paris: Éditions du Seuil, 2013), 9.
[9] Pastoureau, *Vert-histoire d'une couleur*, 99.
[10] Varichon, *Colors – what they mean and how to make them*, 204.
[11] Varichon, *Colors – what they mean and how to make them,* 204; M. Pastoureau, *Vert-histoire d'une couleur*, 56.
[12] Varichon, *Colors – what they mean and how to make them,* 204.
[13] "All the Britons, indeed, dye themselves with *vitrum*, which produces a blue colour, and makes their appearance in battle more terrible." (*Omnes vero se Britanni vitro inficiunt, quod caeruleum efficit colorem*), Caesar, *De Bello Gallico*, Book V, 14. Translation by M. Van der Veen, A.R. Hall and J. May, "Woad and the Britons painted blue," *Oxford Journal of Archaeology* 12 (1993): 367-371 and Varichon, *Colors – what they mean and how to make them,* 275.
[14] Michel Pastoureau, *Bleu-histoire d'une couleur* (Paris: Éditions du Seuil, 2020), 43-47.

mourning in mediaeval times; by wearing blue, one testifies to his assurance that the deceased has gone to heaven. For this reason, the Virgin Mary's mantle began to be painted in blue, a sign of Her close relationship with Heaven and the Divine.[15] In earlier Byzantine art (*ca.* 330-730), the artists represented the Virgin Mary with a Tyrian purple mantle (at the time, the most expensive dye, destined for imperial productions, becoming thus known as "royal purple").[16] In 12th-13th century Europe, ultramarine blue was the costliest pigment due to its geographic provenance and the complicated process of obtaining the ultramarine blue pigment from the mineral lapis lazuli. Lapis lazuli stones (the mineral from which ultramarine was extracted) came from overseas sources (=*ultra maris*), namely Afghanistan. Combined, the two facts limited ultramarine blue to the most important representations of the Catholic Church: the Virgin Mary's mantle and Jesus Christ's clothes.[17] As people grew accustomed to relating blue with the Virgin Mary, this colour grew in the Catholic Church as the most characteristic of Heaven and the Divine. Aiming to fill the ranks of the society of the elect (the Augustinian "City of God"), the nobility promptly adopted the blue as their preferred tone. From this period onward, the blue evoked royalty, nobility, fidelity, and peace.

However, not all views agree on the sanctity of blue. The Arab-Islamic culture used blue to represent Nature, cold and the sky; at the same time, this was the colour of the sinful ones for them. The Koran characterises the guilty excluded from paradise at the Last Judgement as "blue". Moreover, during the mediaeval period, blue was the obligatory colour worn by Jews and Christians living in Islamic territory.

The use of ultramarine as the blue source of mediaeval illuminations and, above all, the extent of its use was an unquestionable financial investment for a scriptorium. To afford its use, it was common to restrict this pigment to the most important and emblematic manuscripts, as this was the most expensive artistic material of the mediaeval palette.

On the use of colour in Portuguese early mediaeval illuminated manuscripts

Colour production in mediaeval illuminations was a complex process. High-quality coloured paints required the finest materials, the most exact formulations and mastery of drawing and painting techniques. For this, the financial resources of a monastery and the knowledge about material processing and paint formulations were of utmost importance. By studying the materials and technology underlying its production, new specificities can be found, namely those related to the use of specific formulations or the sharing of knowledge between the production centres (the religious monasteries) and other cultures. Previous studies on the materials used to produce early Portuguese mediaeval illuminations from the three main Portuguese mediaeval scriptoria - the Alçobaça, the Lorvão and the Santa Cruz de Coimbra

[15] Varichon, *Colors – what they mean and how to make them*, 179-180; P. Ball, *Bright Earth*, 239.
[16] Lloyd B. Jensen, "Royal Purple of Tyre," *Journal of Near Eastern Studies* 22 (1963): 104-118.
[17] Victoria Finlay, *Color: A natural history of the palette* (New York: Random House Trade Paperbacks, 2004), 292-293.

scriptoria - revealed the use of the most important materials used in the European production of 12th-13th century mediaeval illuminations:[18] lead white was the white pigment used *par excellence* as pure paint, but also for highlighting other colours (such as blue and deep organic reds) and for lightening colours. Orpiment was occasionally used as pure paint, and in one notable occurrence, it was found mixed with indigo to produce dark green colours. Turmeric lake (extracted from the roots of *Curcuma longa*) was notably identified as the organic yellow source used in Alc. 433 – a *Lectionary according to the Cistercian rite* - produced in the early days of the activity of the scriptorium of Alcobaça.[19] Red lead is present as pure paint or applied below other colours to produce volume effects. Vermilion red is widely present in the letterings as in red paints. Lac dye was applied as a single reddish colour for shading red lead and mixed with vermilion to produce dark reds. Ultramarine was used as a pure pigment to produce blue paints, but it was mixed with lead white for lightened shades or indigo for darkened shades. Indigo is widely present as a pure paint or mixed with ultramarine for darkening blue shades. A deep-glassy saturated green copper proteinate is widely present as the green colour source. Black was produced by using vegetable and/or animal carbon black pigments.

A. On the use of colour in early mediaeval illuminated manuscripts from the Alcobaça scriptorium

Together with the materials used to produce the illuminated capital letters, both the colour schemes used in the illuminations and the extent of use of each colour can provide interesting information regarding the *fingerprint* in producing illuminations of a scriptorium.[20] To this, the colour schemes and the extent of use of the colours present in 116 letterings and initials letters of three representatives' early illuminated manuscripts produced in the scriptorium of Alcobaça - the three volumes of the *Legendarium* (BNP, Alc. 419, BNP, Alc.420 and BNP, Alc. 421) were analysed.

Colour contrast in the selected illuminated initials is mainly achieved through the contrast between the colours of the painted backgrounds (such as bright red, deep

18 Maria João Melo et al., "The colour of medieval Portuguese illumination: An interdisciplinary approach," *Revista de História da Arte* 1 W (2011), 153-173, and Catarina Miguel et al., "Scientific Study of Cistercian Illuminated Manuscripts: Techniques, Aesthetics and Religion," in *Manuscripts in the Making: Art and Science*, Vol. 2, eds. S. Panayotova and P. Ricciardi (London and Turnhout: Harvey Miller and Brepols, 2018), 134-145.

19 Catarina Miguel et al., "On the Life of a *scriptorium*: Unveiling the Interdisciplinary Study of the MS Alc. 433 Manuscript and its Unique Chronological Record of the Work at the Portuguese Scriptorium of Alcobaça," in *Drugs & Colors In History* (Valencia: Tirant Lo Blanch editors), *in press*.

20 For the analysis of the use of each colour a "pure" colours palette was produced with the most common colour paints found in Portuguese illuminations, enabling the association of a "molecular palette" to each colour paint. The CIELAB parameters of the paints were measured and used to build a CIELAB colour palette that was used as a calibration matrix on a custom-designed algorithm. The mapping of the regions occupied by the colours in each jpeg image was obtained using an in-house built MATLAB function (MATLAB 7.11.0 R2010b) developed by Jorge Sarraguça and João A. Lopes from Requimte-FFUP/Portugal. For more details, see C. Miguel, *Le vert et le rouge: A study on the materials, techniques and meaning of the green and red colours in medieval Portuguese illuminations* (Lisbon: Universidade Nova de Lisboa, 2012), 178-179.

organic red, blue and deep green colour) and the colour present on the initial's ornamentation (Fig. 1). The most common colour schemes were found to be ultramarine-based backgrounds with deep organic red ornamentations; deep-organic red backgrounds with deep-green ornamentations; deep-organic red backgrounds with lapis-lazuli ornamentations; and vermilion-based backgrounds with deep-green ornamentations (Fig. 1).

Fig. 1. Examples of the most common colour schemes found in the Liturgical Codices of early Alcobaça scriptorium. From left to right: BNP, Alc. 418, folio 235v; BNP, Alc. 418, folio 192, BNP, Alc. 418, folio 213 and BNP, Alc. 418, folio 195v.

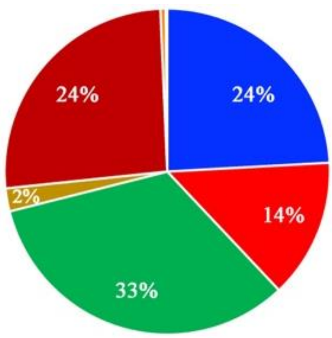

Fig. 2. Colour analysis for the hagiographies from the Portuguese scriptorium of Alcobaça, *i.e.*, BNP, Alc. 419, BNP, Alc. 420 and BNP, Alc. 421. The percentage presented corresponds to the area of each colour paint used in the selected manuscripts. Minor rates related to rose and yellow colour (*circa* 1% of the painted areas of the illuminations) in Alcobaça are not presented in the figure. Green copper proteinate paints are represented by •, vermilion paints by •, lapis-lazuli paints by •, lac-dye paints by •, and brownish-organic paints by •.

The analysis of the extent of use of the areas of the main colours present in the three volumes of the *Legendarium* (BNP, Alc. 419, BNP, Alc. 420 and BNP, Alc. 421) was quantified. Based on the material characterisation of each colour paint, *bright red*, *deep organic red*, orange, blue, green, rose, and yellow were linked to each molecular colour source previously analysed, *i.e. bright reds* were characterised as vermilion, *deep organic reds* as lac dye, oranges as red lead, blues as ultramarine, greens as copper proteinate, roses and yellows to organic dye sources.[21] Considering the importance of red and blue colours on the mediaeval *palette* already mentioned in this chapter, it is interesting to note a more significant extent of the use of deep-green copper proteinate colour (33%) in the three volumes of the *Legendarium* from Alcobaça scriptorium at the expense of red hue (vermilion, 14%; and *deep organic red*, 24%) (Fig. 2).[22]

Deep-green shades consisted entirely of copper proteinate and were used as extensively as ultramarine, suggesting the similar importance of these two colours in the Alcobaça scriptorium (Fig. 2). Somehow, the meaning of spiritual colour is shared between two colours and two cultures once for the Christian culture it was linked to the ultramarine blue colour, whereas for the Islamic culture was linked to the green colour.[23]

Deep organic red (lac dye) reveals a more conspicuous concentration than the bright red vermilion. This marks the first fingerprinting trait of the Alcobaça *atelier* that favoured this organic material, contrary to what was found for the other two Portuguese scriptoria of Santa Cruz de Coimbra and Lorvão.[24] Considering that the amount of ultramarine use is *ca.* 24% of the painted area under analysis, the progression of the use of lac dye together with ultramarine (two of the costliest colour sources at the time) in Alcobaça might well be assigned to an improvement in the financial resources available in the scriptorium of Alcobaça at the time. The wide presence of lac dye in the manuscripts produced at the Alcobaça scriptorium may also give evidence of further facts. Extracted from the secretion of the insect *Kerria lacca* present across India, Southeast Asia and Southern China, lac dye was among the most expensive red dyes used in mediaeval times, with a profound use in the Mediterranean regions under Islamic influence.[25] Its trading was assured mainly by the Jewish and Arabs, who, during the 12th century, controlled the trade routes across these territories. In this sense, being lac dye such an expensive dye at the time and considering the extensive use of vermilion in the initial letters and titles throughout the three manuscripts (reflecting the availability of vermilion in the scriptorium), the use of this colour might have been intentional, suggesting once more the effect of the Islamic culture in Alcobaça scriptorium.

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21 Melo et al., "The colour of medieval Portuguese illumination.".

22 Miguel et al., "Scientific Study of Cistercian Illuminated Manuscripts: Techniques, Aesthetics and Religion," 134-145.

23 As already mentioned in section I of this chapter.

24 Miguel, *Le vert et le rouge*, 80-83.

25 Jo Kirby, "Paints, pigments, dyes" *in* T. Glick, S. J. Livesey and F. Wallis (editors). *Medieval Science, Technology, and Medicine – an Encyclopedia* (New York: Routledge, 2005), 379-383.

B. On the use of blue colour in early mediaeval illuminated manuscripts from the Alcobaça scriptorium

The research into the colour distribution in the illuminated manuscripts from the Alcobaça scriptorium reflects well the trend in the use of blue colour in the earliest period of its activity. During this period, blue colours had their source of either inorganic or organic origin. In early produced manuscripts, ultramarine was the pigment used to produce the inorganic blue paints that would later be replaced by azurite – a cheaper and easier-to-be-found inorganic pigment. Organic indigo was often used in the Alcobaça scriptorium to produce darker to blue-greyish hues (Fig. 3).[26]

Fig. 3. Inorganic (ultramarine and azurite) and organic (indigo) blues sources and their use in early illuminated manuscripts from the Alcobaça scriptorium (from left to right: ultramarine, azurite and indigo; from top to bottom: raw colour source, magnified image of the paint and detail of its application).

- Ultramarine blue

Extracted from lapis lazuli, ultramarine was the blue *par excellence* in mediaeval art. It was the most prestigious and expensive of all pigments, both for its brilliance, flawless and optical qualities, but also because it was difficult to obtain (both of the geographical rarity to mine it and the labour required for its preparation).[27] The most important lapis lazuli deposits are placed in the Kokcha River valley in Afghanistan, where it has been mined for more than 6000 years for jewellery production, as an ornamental stone and a pigment.[28] During the 13th century, Marco Polo describes in his reports the main source for lapis lazuli to be placed in Badakhshan (now

[26] Miguel, *Le vert et le rouge*, 36.
[27] Eastaugh et al., *The Pigment Compendium*, 217-218, and Kirby, "Paints, pigments, dyes.", 379-383.
[28] Eastaugh et al., *The Pigment Compendium*, 217.

Afghanistan).[29] Besides lazurite - the aluminosilicate blue source ($Na_{6-10}Al_6Si_6O_{24}S_{2-4}$) of lapis lazuli stones - the mineral contains white streaks of calcite ($CaCO_3$) and goldish-look-like agglomerates of iron pyrites (FeS_2).[30] For very high-quality minerals (meaning minerals with a high content of lazurite), it could be carefully ground and directly used as a pigment. However, grinding produces a greyish-pale blue pigment, as it mechanically breaks the aluminosilicate cage of lazurite, resulting in the freedom of the S_3^{2-} groups that are present in its lattices structure, which is responsible for the deep-intense blue colour of lazurite (note that the charge transfer transition inside the S_3^{2-} has its maximum of absorption at ca. 600 nm, with a visual observation reflected in the deep blue colour range).[31] Several procedures could be followed to reach high-pure ultramarine pigment. In this regard, D.V. Thompson refers to some mediaeval procedures for lazurite extraction from lapis lazuli stone during the 13th-14th century, involving the mixture of the powdered lapis lazuli to waxy and resinous bodies with water or mixing it with a paste of wax, oil, and resin, followed by kneading this mixture underwater (or under ash water) until the blue comes out in it.[32] In his *Il Libro dell'Arte* (14th-15th century), Cennino Cennini describes its purification with a detailed procedure involving the mixture of powdered lapis lazuli to a mixture of pine rosin and mastic gum to produce a *plastic* that should be *handworked* for several days with the hands greased with linseed oil, followed by kneading this *plastic* under a strong ash water mixture, to where the purified lazurite would be extracted. [33]

- *Azurite*

Azurite is a blue basic copper carbonate ($2CuCO_3.Cu(OH)_2$). It is usually associated in Nature with malachite ($CuCO_3.Cu(OH)_2$), the basic green copper carbonate that is even more abundant in Nature, and with other copper-rich minerals, such as cuprite (Cu_2O), tenorite (CuO), limonite ($FeO.OH.nH_2O$), chrysocolla ($(Cu,Al)_2H_2Si_2O_5(OH)_4.nH_2O$), or occasionally, with iron-rich phases like limonite ($FeO.OH.nH_2O$).[34] Azurite is often found in places where copper is mined; in Portugal, it could be extracted from the copper mines in Aljustrel, which, during the 12th century, were still under Muslim control.[35]

Several methods were available in the Middle Ages to prepare azurite pigment. The simplest one consisted of grinding the mineral, followed by several cycles of water decantation to separate the desired quality of pigment. The higher the number of those cycles, the more only the coarsest fraction of azurite grains would be selected

[29] Ball, *The invention of Colour*, 267.

[30] Eastaugh et al., *The Pigment Compendium*, 217.

[31] Joyce Plesters, "Ultramarine Blue, Natural and Artificial," in *Artists' Pigments: A Handbook of their History and Characteristics*, Vol. 2, ed. A. Roy (New York: Oxford University Press, 1993), 37-54.

[32] Daniel V. Thompson, *The materials and techniques of mediaevel painting* (New York: Dover Publications, 1956), 77, 147.

[33] Daniel V. Thompson, *The Craftsman's Handbook 'Il libro dell'Arte' by Cennino d'Andrea Cennini* (New York: Dover Publications, 1960), 37-38.

[34] Eastaugh et al., *The Pigment Compendium*, 33.

[35] Olivia Remie Constable, *Mediaeval Iberia: Readings from Christian, Muslim and Jewish sources,* 2nd edition (Philadelphia: University of Pennsylvania Press, 2012), 186.

for use as a pigment. This fact would have several implications: the largest grains of azurite give the most perfect, deep blue; the smallest grains yield a dull, pale blue. However, using the most significant fraction of grain required some skill from the illuminators, as the coarsest pigment, the feel-like sandy and complex to apply in pictorial techniques it becomes. Therefore, the illuminator had to establish the best balance between the desired optical effects and the quality of the grain he wished to reach. Other more complex recipes could be used, such as the one described by Cennino Cennino, in which the mineral was mixed with honey and lye (ash water) and just afterwards ground in a porphyry.[36] Likewise to ultramarine, also azurite grinding was a sensitive process: coarsely ground azurite produces dark blue; fine grindings make lighter-greyish tones (note that d-d electronic transitions of copper have its maximum absorption band at ca. 640 nm, with a visual observation reflected in the deep blue colour range; the coarser the grains, the more forbidden the d-d electronic transitions of copper, the lighter-greyish is the resulting pigment).[37] With ageing, azurite might become greenish, primarily due to its alteration into malachite.[38] Although considered the best blue *for all ordinary purposes*, its lower cost when compared to the costs involved with the acquisition of ultramarine pigment, made azurite often to be replaced ultramarine in mediaeval paintings or used to prepare underpaints of the more precious ultramarine blue paints.[39]

- *Indigo*

Indigo is the blue colourant extracted from the *Indigofera* species. The first indication of this colourant is by Vitruvius in the 1st century BC, who mentioned it in his *De architectura* (Book VII, Chap. 14).[40] Centuries later, Discorides refers to it in his work *De Materia Medica* as "the dye that is called indigo comes from India. It forms a spontaneous exudation on a type of reed growing tree" (Book 5, Chap. 107, 1st century).[41] Indigo was more used by the Greeks and Romans as a pigment than as a dye.[42] It has its origin in India, and in this period, its trading costs were reflected in the high prices that indigo could reach.[43] There are references that in the Mediaeval period, it was extracted from *Indigofera tinctoria* and traded from Persia to Muslim Iberia, and from there, distributed to other European territories.[44] In mediaeval manuscripts, it was extensively used as a pigment, mainly extracted from *Indigofera*

[36] Thompson, *The Craftsman's Handbook Il libro dell'Arte' by Cennino d'Andrea Cennini*, 35.
[37] Rutherford J. Gettens and Elisabeth West FitzHugh, "Azurite and Blue Verditer" in *Artists' Pigments: A Handbook of their History and Characteristics*, Vol. 2, ed. A. Roy (New York: Oxford University Press, 1993), 23-35, and Paula Ricciardi et al., "Use of Visible and Infrared Reflectance and Luminescence Imaging Spectroscopy to Study Illuminated Manuscripts: Pigment Identification and Visualization of Underdrawings," *Proceedings of SPIE: The International Society for Optical Engineering* 7391 (2009), 739106.
[38] Eastaugh et al., *The Pigment Compendium*, 33.
[39] Thompson, *The Craftsman's Handbook 'Il libro dell'Arte' by Cennino d'Andrea Cennini*, 134.
[40] Helmut Schweppe, "Indigo and Woad," in *Artists' Pigments: A Handbook of their History and Characteristics*, Vol. 3, ed. E.W. FitzHugh (New York: Oxford University Press,1997), 81-107.
[41] Schweppe, "Indigo and Woad."
[42] Schweppe, "Indigo and Woad."
[43] Jenny Balfour-Paul, *Indigo* (London: British Museum Press, 2000), 11.
[44] Maria João Melo, "History of natural dyes in the ancient Mediterranean world," in *Handbook of Natural Colorants*, eds. T. Bechtold and R. Mussak (Chichester: John Wiley & Sons, 2009), 3-18.

tinctoria leaves in alkaline water, to induce the process of fermentation.[45] Ancient recipes commonly add urine, ash, or slaked lime. The soaked leaves were beaten to a pulp and stirred vigorously to oxidise the mass until the indigo formed a precipitate, which was then dried and often formed into cakes.[46]

Mediaeval treatises on mediaeval painting mention using Indigo for shadings and to paint grey areas.[47]

C. Analysis of the chronological use of blue colour sources in early mediaeval illuminated manuscripts from the Alcobaça scriptorium

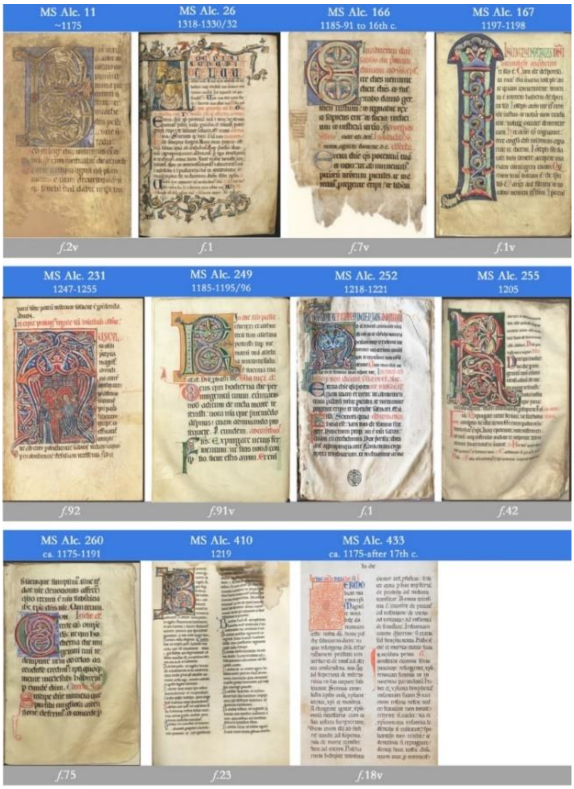

Fig. 4. Manuscripts selected for the chronological use of blue colour sources in early mediaeval illuminated manuscripts from the Alcobaça scriptorium. The dates provided follow the results of the liturgical studies by Catarina Fernandes Barreira published in this book © National Library of Portugal, Lisbon.

45 Melo, "History of natural dyes in the ancient Mediterranean world."

46 Eastaugh et al., *The Pigment Compendium*, 194.

47 *Schedula diversarum atrium* (twelfth century) mentions this material in Chapters 14 and 16; another treatise devoted specifically to illumination, *De Arte illuminandi* (fourteenth century), mentions it in Chapter 9. See also F. Brunello, [*De Arte illuminandi and other Treatises on Mediaeval Illumination*] (Vicenza: Neri Pozza, 1975).

The study of the chronological use of blue colour sources in the Alcobaça scriptorium here presented adopted a holistic approach based on documental analysis, liturgical analysis, and technical-scientific inspection. On the one hand, the documentary-liturgical process relied on the information in the manuscripts regarding both agreements reached in the previous Cistercian General Chapter and Historical events. On the other hand, the material characterisation focused on the analysis of the blue paints present in both illuminated capital letters and blue colour letterings.

For this, a set of eleven illuminated Liturgical Codices from the 12th-14th century produced in the Alcobaça scriptorium were analysed: Alc. 11, 26, 166, 167, 231, 249, 252, 255, 260, 410 and 433. For most of the codices, it was possible to date, from the liturgical point of view, the primitive nucleus and the successive additions that were being integrated in the form of folios or notebooks.[48] Following this, in-situ, non-invasive analytical techniques were used to obtain elemental (h-EDXRF), molecular (UV-Vis-NIR FORS) and chemical imaging (Hyperspectral Imaging) data of their blue-colour paints (Fig. 4).

- Blue colour sources in the Liturgical Codices of early Alcobaça scriptorium

The blue colour characterisation in the Liturgical Codices of early Alcobaça scriptorium (late 12th century) had its starting point with the material analysis of the BNP, Alc. 11 – a Psalter-hymnal which, according to the liturgical research, might have been produced at the very early begging of the activity of the Alcobaça *scriptorium*. [49] As one of the oldest manuscripts produced in Alcobaça that has reached our days, the manuscript Alc. 11 is a unique testimony of the materials and techniques available during the early activity of this scriptorium. The manuscript Alc. 11 opens with a full-blue background-painted capital letter in f. 2v (Fig. 4). UV-Vis FORS[50] analysis of the blue paint allowed for the identification of a reflectance band at 475 nm, a shoulder at 755 nm and a maximum absorption band at ca. 600 nm attributed to the charge transfer between the sulphur atoms of the trisulphur anion S_3^{2-} present in the lattice of the aluminium-silicate complex of ultramarine blue pigment (Fig. 5).[51]

Elemental h-EDXRF[52] confirmed the tentative attribution of UV-Vis FORS by identifying the presence, as primary elements, of aluminium (Al), silicon (Si) and

[48] On the methods and results of liturgical studies and accurate dating of the manuscripts from the Alcobaça *corpus*, please see the article by Catarina Fernandes Barreira in this book.

[49] Conceição Casanova et al., "Narrating codex history: the case study of a psalter-hymnal from Alcobaça Monastery, Portugal," *Journal of Medieval Iberian Studies* 14 (2022), 127-141.

[50] Fiber-optic reflectance spectroscopy in the ultraviolet and visible range (UV-Vis FORS) was performed using a LR1-T v.2 compact spectrometer (ASEQ Instruments) with a spectral range of 300–1000 nm, a spectral resolution < 1 nm (with 50 µm slit) and a spot size of 12 mm2 (3 mm × 4mm). Measurements were taken using the ASEQ CheckTR software. Calibration was achieved using Whatman filter paper. Samples were analysed at an exposure of 100–200 ms, five scans. To ensure the statistical reproducibility of the acquired data, three blue paints along the manuscript were analyses (ff. 2v, 93r and 144r), each in three different areas measured three times each.

[51] Maurizio Aceto et al., "Characterisation of colourants on illuminated manuscripts by portable fibre optic UV-visible-NIR reflectance spectrophotometry," *Analytical Methods*, 6 (2014): 1488-1500.

[52] Elemental characterization of blue paints was performed with a Tracer III-SD® Fluorescence handheld EDXRF spectrometer (Bruker), equipped with a 10 mm XFlash® SDD, a Peltier cooled

sulphur (S), arising from the aluminosilicate matrix of lazurite ($Na_{6-10}Al_6Si_6O_{24}S_{2-4}$). Aside from these elements, calcium and iron were also identified, which might be related to the presence of calcite ($CaCO_3$) and iron pyrite (FeS_2), two minerals often present as secondary phases of lapis lazuli stones.[53]

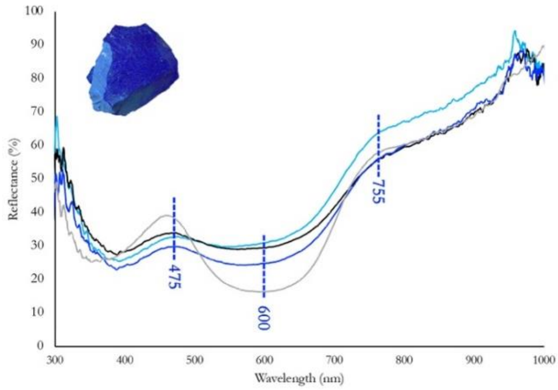

Fig. 5. Representative UV-Vis FORS spectra of blue paints present along the BNP, Alc. 11: a lighter blue is present in the background of the illuminated capital letter in folio2v (*blue spectrum*) and the blue lettering in folio 93 (*cyan spectrum*), and a darker blue is present in the blue lettering in folio 144 (*black spectrum*); and of historically accurate reconstruction of an ultramarine paint (*grey spectrum*).

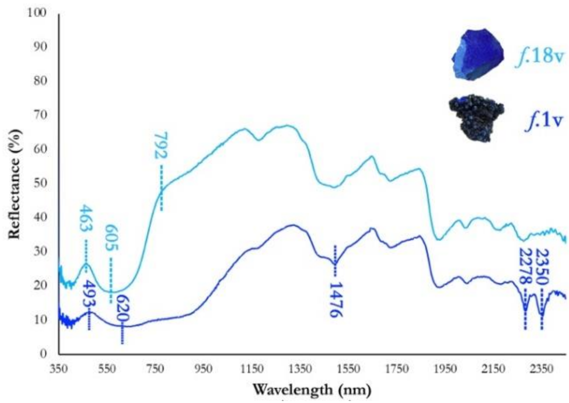

Fig. 6. Representative UV-Vis-NIR FORS spectra of blue paints in the MS Alc. 433: *light blue*, UV-Vis-NIR FORS spectrum of the ultramarine blue paint in folio18v; *dark b*lue, UV-Vis-NIR FORS spectrum of the azurite blue paint in folio1v

detector with a resolution of 145 eV (FWHM for Mn-Kα) at 100,000 cps, and an Rh target. Analyses were made at 40 kV and 11 μA with no filter, an acquisition time of 30 s and a 3 mm x 4 mm spot size. The instrument was positioned approximately 2–3 mm away from the surface under analysis. EDXRF spectra were acquired from three contiguous spots to evaluate the reproducibility of the results. All h-EDXRF spectra were acquired with the S1PXRF software and analysed with the ARTAX software.

53 Eastaugh et al., *The Pigment Compendium*, 217-218.

The second manuscript selected for this study was the second volume of the *Leccionarium* for the Divine Office produced in the Alcobaça *scriptorium*, the manuscript Alc. 433. Produced between 1201 and 1250, according to the National Library catalogue,[54] liturgical studies by Catarina Fernandes Barreira,[55] already mentioned in this text and presented in this publication, allowed the conclusion that the present physical arrangement of the BNP, Alc. 433 does not correspond to a chronological organisation but to a non-organized compilation of texts produced in five different periods:

1. An *initial core*, which was produced in Alcobaça during its earliest years of activity (around 1175) (folio 18v to folio 227v);
2. A *first addition*, made after 1175 (most probably between 1176-1180; folio 15 to folio 17);
3. A *second addition*, made around 1292 (folio 228 to folio 231);
4. A *third addition* dating after 1318 (folio 1 to folio 14v);
5. A *fourth* and last *addition* dates back to the 17th century (folio 232 to folio 236).[56]

UV-Vis-NIR FORS[57] analysis of the blue paints present throughout the five sections of the manuscript Alc. 433 allowed identifying the characteristic spectral fingerprints of ultramarine in the blue colour paints of the two earliest sections of the manuscript (*initial core* and *first addition*), namely those related to the charge-transfer absorption bands of the sulphur polyatomic ions present in the lattice of the aluminium-silicate cage of lazurite minerals (reflectance band at 463 nm, the shoulder at 792 nm and the maximum absorption band at ca. 605 nm) (Fig. 6). The blue paints present in the remaining additions (the *second addition* around 1292 and the following two additions) were painted with azurite, identified by its characteristic UV-Vis-NIR spectral fingerprints, namely the reflectance band with a maximum at 493 nm, a maximum absorption band at ca. 620 nm related to the d-d electronic transitions of copper, and the vibrational absorption bands at 1476 nm [2ν(OH)] and at 2278 and

[54] Information available at (Accessed: January 23rd 2023): https://catalogo.bnportugal.gov.pt/ipac20/ipac.jsp?profile=bn&source=~!bnp&view=subscriptionsummary&uri=full=3100024~!1862802~!2&ri=1&aspect=subtab13&menu=search&ipp=20&spp=20&staffonly=&term=lus%C3%83%C2%ADadas&index=.TW&uindex=&aspect=subtab13&menu=search&ri=1.

[55] On the results of liturgical studies and accurate dating of the MSS from the Alcobaça *corpus*, please see the article by C. Fernandes Barreira in this book.

[56] Shatila Fitri, "The Study of the Biographical Trajectory of a Portuguese twelfth Century Illuminated Manuscript: the *Leccionarium* MS Alc. 433 from the Alcobaça Collection held by the Biblioteca Nacional de Portugal,".

[57] UV-Vis-NIR spectra were collected with an i-Spec® 25 portable spectrometer (B&WTek) combined with a handheld reflectance probe of trifurcated fibre optic bundle series integrated with a 5W tungsten halogen source, with a focal aperture of 5mm and extended InGaAs array sensor, in the 400- 2500 nm spectral range. Analyses were performed with the iSpec4 software. For Detector 1 (BRC711U- 512, spectral range of 345.6 nm-1061.3 nm), the integration time was 95 ms, 25x accumulations, no smoothing; for Detector 2 (BTC261P-512-OEM61, spectral range of 883.0 nm- 1718.0 nm), the integration time was 250 µs, 50x accumulations, no smoothing (high-sensitivity detector mode); for Detector 3 (BTC263E-256- OEM61, spectral range of 1482.4 nm-2654.9 nm), the integration time was 332 µs, 100x accumulations, no smoothing (high-sensitivity detector mode).

2350 nm [(ν+δ)(OH) and 3ν(CO_3)] related to the hydroxyl and carbonyl groups of azurite ($2CuCO_3.Cu(OH)_2$) (Fig. 6).[58]

Elemental h-EDXRF analyses supported these initial attributions, identifying, as primary elements, the presence of aluminium (Al), silicon (Si), and sulphur (S) for the ultramarine-based paints; copper (Cu), iron (Fe) and calcium (Ca) in the analyses of azurite-based paints corroborating the presence of the mineral azurite ($2CuCO_3.Cu(OH)_2$). Iron may be related to the presence of pyrite (FeS_2) and calcium to calcite ($CaCO_3$), also commonly associated with azurite. [59]

In this sense, somewhere between the first and the second *addition* of the MS Alc. 433 (i.e., after 1175 and up to around 1292), azurite replaced ultramarine as a pigment to produce blue paints consistently up to the *fourth addition* in the 17th century (Fig. 7).

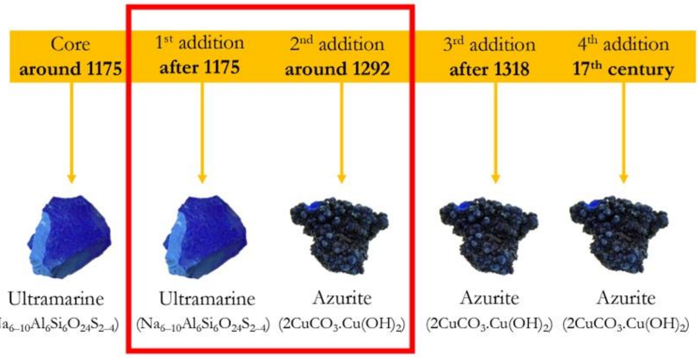

Fig. 7. Timeline for the use of inorganic blue pigments in the BNP, Alc. 433.

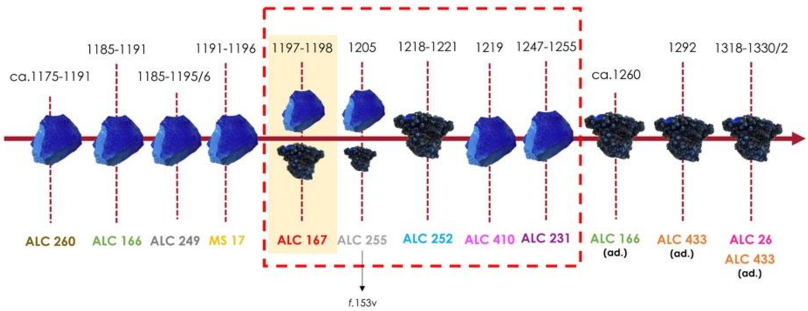

Fig. 8. Timeline for the use of inorganic blue pigments in the eleven selected Liturgical manuscripts (core texts and additions – signed as "*ad.*"), considering the accurate dates determined by the liturgical analysis and the blue pigments' sources identified by the material characterisation.

[58] Kathryn A. Dooley et al., "Mapping of egg yolk and animal skin glue paint binders in Early Renaissance paintings using near infrared reflectance imaging spectroscopy," *Analyst* 138 (2013): 4838.
[59] Eastaugh et al., *The Pigment Compendium*, 33-34.

The analysis of the manuscript Alc. 433 yielded an essential piece of information: the transition between ultramarine and azurite occurred between the late 12th century and the late 13th century (after 1175 up to ca. 1292). Aiming to narrow this transition window to a more specific period, other manuscripts were studied using the same holistic approach. For this, a combination of liturgical studies-material characterisation was extended to the remaining codices of the set of eleven illuminated Liturgical Codices under analysis in this chapter (Fig. 4), of which liturgical study places its date of production between ca.1175-1191 (BNP, Alc. 11) and 1318/1330-32 (BNP, Alc. 26), the material analysis highlighted the consistent use of ultramarine from ca.1175-1191 (Alc. 11) to 1196 (BNP, Alc. 167), and of azurite from ca. 1260 (with the last addition in the Alc. 166) to 1318/1330-32 (Alc. 26) (Fig. 8). In-between this period (a range of ca. 58 years) ultramarine and azurite are used combined in the scriptorium. It is just after ca. 1260, with the last addition to the Alc. 166, that azurite appears to replace ultramarine to produce blue colour paints (both in the two last additions of Alc. 433, as in the entire Alc. 26) (Fig. 8).

From the five manuscripts highlighted in red in the timeline for the use of inorganic blue pigments in the selected Liturgical manuscripts (Fig. 8), a note for the Alc. 252, on which azurite appears only in one illuminated capital letter (the one present in folio 153v), in what seems to be an ancient conservation/restoration procedure. The materials identified through its material characterisation agree with those found to be in use in the scriptorium during the late 13th century and onwards.

The following two manuscripts (BNP, Alc. 410 and BNP, Alc. 231) reveal an interruption of the emerging trend toward a complete replacement of ultramarine with azurite in both the letterings and the illuminated capital letters (Fig. 8).

Finally, the BNP, Alc. 167 – an *Evangeliarium* according to the Cistercian rite – provided the most decisive information for the present study. This manuscript marks the first phase of introducing azurite on the desks of Alcobaça *illuminators*, who endeavoured in what seems to be an experimental attempt in the scriptorium.

The abovementioned approach combining UV-Vis-NIR FORS with h-EDXRF[60] and Hyperspectral Imaging data unveiled the presence of azurite combined with ultramarine across the entire manuscript, not only for producing dichromatic initials (Fig. 9) but also to produce full-illuminated capital letters (Fig. 10).

Important to stress is the fact that ultramarine is the sole pigment used for blue internal letterings throughout the manuscript (*litterae notabilis*).[61] In initials of a different hierarchical order (*opening initials*, *chapter initials* or *paragraph initials*), azurite appears as supplementary to the later textual portions of the Alc. 167. The reason for the augmentation of the colour palette with azurite may be something other than an economic order.

[60] UV-Vis-NIR FORS allowed the identification of the characteristic spectral features for the presence of lapis lazuli and azurite, already mentioned in this text. Elemental h-EDXRF analyses corroborated the presence of lapis lazuli by the identification, as major elements, of sodium (Na), aluminium (Al), silicon (Si), sulphur (S); whereas azurite was corroborated with the identification, as major elements, of copper (Cu), iron (Fe) and calcium (Ca).

[61] An enlarged letter within a textual portion that marked the beginning of a new sentence of a passage.

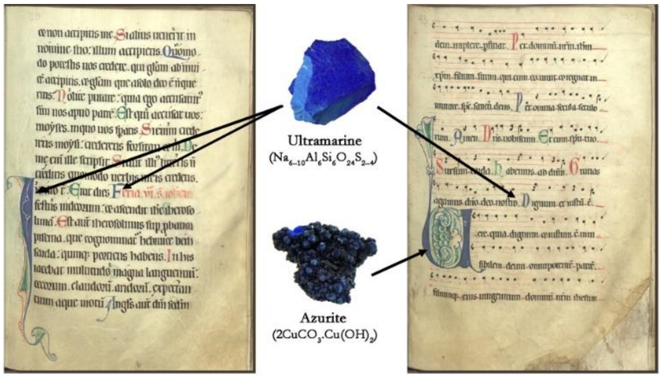

Fig. 9. Two remarkable uses of azurite and ultramarine in the BNP, Alc. 167: *left*, the use of ultramarine as the blue colour source to produce the blue paints of both the initial and of the letterings in folio 20; *right*, the use of azurite as the blue colour source to produce the blue paint of the initial, whereas ultramarine was used to produce the blue paint of the letterings in folio 83.

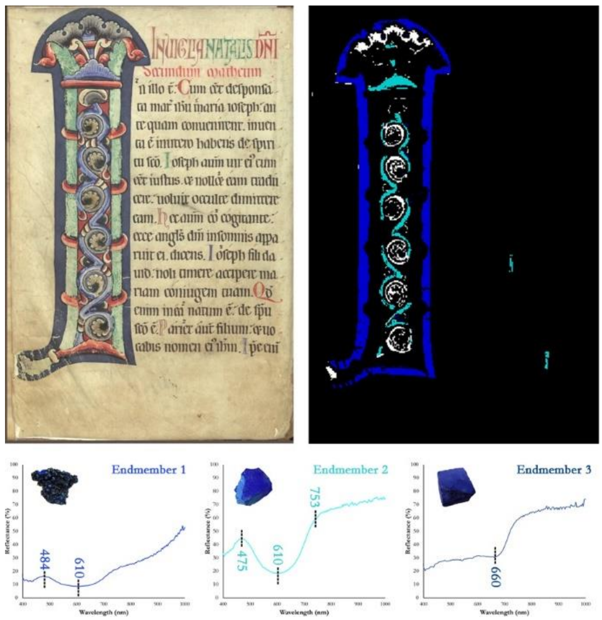

Fig. 10. Distribution maps obtained with HSI data for Alc. 167, folio 1v. Top left: visible image; top right, HSI map (the solid blue areas are calculated with SAM pixel-matching algorithm, with the following values of cosine angle (tolerance): *endmember 1* (dark blue) 0.090 rad; *endmember 2* (cyan) 0.100 rad, *endmember 3* (grey-white) 0.085 rad. Bottom: spectra of *endmembers 1*, *2* and *3*, where azurite, ultramarine and indigo were identified, respectively.[62]

[62] Azurite and ultramarine were identified based on the characteristic spectral features already presented in this paper. Indigo was identified based on the absorption band at ca. 680 nm related to the $\pi \rightarrow \pi^*$

If azurite had been introduced due to a lack of availability of ultramarine in the scriptorium or to save resources, it would have been more likely that these secondary motifs would have been painted with azurite - a cheaper pigment than ultramarine. Instead, azurite is used contextually to ultramarine and to produce letters of a higher hierarchy–compared with Fig. 9 (folio 83), where azurite is used for the paragraph initial. In contrast, ultramarine is the pigment of the *litterae notabilis*.

Regarding the major capital letters, azurite produced a trinary-blue colour scheme with ultramarine and indigo (Fig. 10).

This same feature of combining ultramarine and azurite was found for other full-elaborated capital letters present in this manuscript, suggesting that the illuminator followed this combination to produce two different shades not as a resource approach but as a stylistic choice. When combined with indigo (as seen in Fig. 10), the illuminator reached three different pure-colour hues ranging from *blue-greyish* to *bright-dark blue*, with an extraordinary visual impact.

Conclusion

The holistic approach followed in this work, i.e. combining the results gathered from the liturgical analysis of the manuscripts with the in-situ non-invasive analysis of the illuminations, proved to be fundamental for establishing the chronological use of blue colour sources in the Portuguese medieval scriptorium of Alcobaça. Identifying the introduction of azurite in the Alcobaça scriptorium back to 1197-1198 places its use back before the introduction of the French university manuscripts at Alcobaça *armarium* around 1260, when azurite was thought to have been introduced into the scriptorium. If so, that would have agreed with what was found by P. Ricciardi et al. in a study of French and Italian illuminated manuscripts held by the Fitzwilliam Museum, which identified the use of azurite in manuscripts from the mid-13th century onwards.[63] In fact, with the present work, it is proved that not only azurite was introduced in the Alcobaça scriptorium much earlier (ca. 60 years), but also it was introduced contextually to the use of ultramarine or combined with indigo to produce letters of a higher hierarchy, reaching pure-colour hues ranging from *blue-greyish* to *bright-dark blue*, with an extraordinary visual impact. It is important to stress that this introduction occurred during a period of territorial expansion of the Alcobaça Monastery, with the purchase of a considerable area of land, denoting the economic capacity of the Monastery at the time.[64] In this sense, the azurite introduction should

electronic transitions of the molecule orbitals of indigo structure. For a deeper description of the phenomenon, see Marco de Leona et al., "Identification of the Pre-Columbian Pigment Maya Blue on Works of Art by Noninvasive UV-Vis and Raman Spectroscopic Techniques," *Journal of the American Institute for Conservation* 43 (2004): 39-54.

[63] Paula Ricciardi et al., "*It's not easy being green*: a spectroscopic study of green pigments used in illuminated manuscripts," *Anal. Methods* 5 (2013): 3819-3824.

[64] Saul Gomes, "Revisitação de um velho tema: a fundação do Mosteiro de Alcobaça," in *Espaços, Territórios, Paisagens. Actas do Colóquio Internacional, Mosteiro de Alcobaça, Portugal, 16 – 20 Junho de 1998*. Lisboa: IPPAR, 1(2000), 27-72; and Saul Gomes, "Entre memória e História. Os primeiros tempos do Mosteiro de Alcobaça," in *Revista de História da Sociedade e da Cultura* 2 (2002), 187-256.

not have been for financial purposes but for artistic intention. What is certain now is that the innovative use of materials and stylistic visual effects at Alcobaça in its early decades of activity reflect the richness of this scriptorium, not just in terms of its resources but in the high-skill ability of its illuminators. A richness that stands Alcobaça out from the *common* Cistercian scriptoria, placing it into the group of the most outstanding Cistercian scriptoria regarding materials advancements and stylistic creativity.

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The Portuguese Foundation for Science and Technology (FCT) with National Funds under the research project "Cistercian Horizons Studying and Characterizing a Medieval Scriptorium and its Production: Alcobaça, dialogues between local identities and liturgical uniformity" (PTDC/ART-HIS/29522/2017) the Norma Transitória (DL 57/ 2016/CP1453/CT0070 and DL 57/2016/CP1372/ CT0012) and the HERCULES Laboratory (UIDP/04449/2020 and UIDB/04449 /2020) supported the research project.

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