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Natural Dyes in Embroideries of Byzantine Tradition, the Collection of Embroidered Aëres and Epitaphioi in the National Museum of Art of Romania

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Abstract: The medieval textiles collection of the National Museum of Art of Romania (MNAR) has been in place since 1865 and nowadays preserves about 1000 medieval and pre-modern weavings and embroideries. These extremely valuable objects, dated between the 14th and the 19th centuries, are mainly religious embroidered garments and veils with special significance in the Byzantine li-turgy. Ecclesiastical embroideries of Byzantine tradition are characterized by a complex technique: metallic threads with a silk core, metallic wires and coloured silk threads are couched over padding on layers of silk and cellulosic supports so as to create relief through light reflection. The silk sup-ports and the sewing threads are coloured, mainly in red, blue, green and yellow hues, and analytical investigations of the dyes used in embroideries preserved in the MNAR, in the Putna and Sucevița Monasteries, have been released in previous studies by the corresponding author. The present work continues the approach with research into dyes in about 25 *aëres* and *epitaphioi* from the MNAR collection. Considering their privileged function in the liturgical ritual, these luxurious pieces embroidered with silver, gilded silver or coloured silk threads and decorated with pearls, sequins or semi-precious stones are the most faithful description of the stylistic and technological evolution of the art of post-Byzantine embroidery in the Romanian provinces. The data resulting from the present research will improve the knowledge regarding this topic. Dye analysis was performed by liquid chromatography with diode array detection, while fibres were characterized by infrared spectroscopy (with attenuated total reflectance) and optical microscopy. The biological sources identified—carminic acid-based dyes, redwood, dyer's broom, weld, indigo-based dyes—-will be discussed in correspondence with their use in the embroidery technique: support, lining and embroidery threads, together with other sources previously reported on Byzantine embroideries in Romanian collections, and in similar objects preserved at Holy Mount Athos.

Keywords: natural dyes; liturgical embroideries of Byzantine tradition; *epitaphios*; *aër*; liquid chromatography; identification; Romania

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1. Introduction

Public and ecclesiastical museums in Romania preserve valuable medieval and pre-modern textile collections, perfectly integrated within the European luxury textiles, especially the Byzantine and post-Byzantine. Medieval embroideries, and especially the liturgical vestments and veils, are the most representative. These objects belong to the treas-uries of the Orthodox or Catholic churches that have been active over time in the three historical Romanian provinces, Moldavia, Wallachia and Transylvania. Liturgical

embroideries in the Byzantine style particularly enjoyed great appreciation in the Romanian ter-ritories and set up a special chapter in the history of medieval and pre-modern South-eastern European art.

The large number of embroidered liturgical objects in Byzantine or post-Byzantine workshops perfectly document the interest of the Romanian medieval cultural elites in the Byzantine imperial art. The same enthusiasm was equally evident in all the other artistic fields: architecture, painting, sculpture and illuminated manuscripts. The geographical position of the three historical Romanian provinces allowed direct cultural exchanges with Byzantium during the Middle Ages, which is nowadays richly documented by archaeological research. However, towards the end of the Middle Ages, the political history of the provinces took different forms of attachment to the Byzantine artistic heritage. The principality of Transylvania, which had been in the sights of the newly established Hungarian state in the Pannonian Plain since the 10th century, was much more receptive to Western European art, which was intensively promoted through the Catholic Church and the papacy. Moldavia and Wallachia remained constantly within the sphere of influence of the Byzantine Empire, especially after the state organization of the two provinces, which had been under the spiritual authority of the Patriarchate of Constantinople since the mid-14th century [1].

The National Museum of Art of Romania (MNAR) preserves the largest and most representative collection of medieval textiles in Romania. Created in and developed since 1865, it was at that time, the first public collection of Byzantine and post-Byzantine art in the world [1]. Some of these objects, mainly those dated from the 15th and 16th centuries, were studied in terms of materials and techniques in the early 2000s, the years which preceded the re-opening of the Romanian Medieval Art Gallery [2]. Other liturgical embroideries from the same period, or later, preserved in MNAR or in monasteries in the northeastern part of Romania were also subject of research, with reference to the metallic threads and dyes [3–7].

Liturgical textiles of Byzantine tradition have two different supports, with distinct roles: one to provide resistance and another one as a base for building the relief [8]. The former is composed of a cellulosic support (never visible) (Figure 1), which is not coloured, and a silk support with an aesthetic role, which is totally or partially visible [8]. In order to provide a contrast with the precious metallic embroidery, as well as to give it importance, the silk support was coloured according to the Church canons: purple-red in most cases, with blue, yellow and brown also accepted [8]. Embroidery is made with silk threads—visible or wrapped up in precious metallic bands and wires—-or just with metallic threads, to result in the so-called "needle painting" [9]. The scenes and decoration cover most of the silk support, which in some cases becomes almost completely masked [8] (Figure 1).

Natural dyes' identification, together with attribution of the most probable biological sources used, represents an essential output in the studies of historical textiles. Data regarding the dyes' places of origin, information on their first commerce and about trade routes, as well as guild regulations represent valuable instruments to document textile production and place a studied object in time and place [10,11]. The identification of dyes in historical textiles is nowadays possible mainly by liquid chromatography with diode array detection (LC-DAD). Since its first use in 1985, the above-mentioned configuration was considered for many years as the standard method for dye analysis [12–16]. Later on, the development of mass spectrometers (MS), with increased sensitivity and lower detection limits, mainly used in LC-DAD-MS layouts, proved to be extremely useful in biolog-ical sources' detection [17–19]. Results are optimized when these structures are exploited together with dedicated in-house built databases. Moreover, when applied in tandem configurations, mass spectrometers confirmed their efficiency to characterize unknown marker compounds, with consequences in the accurate attribution of the biological sources used, and better contextualization of the studied textiles [20–22]. The interest in optimizing the information acquired from the tiny samples available and discerning between sources

containing the same aglycones but different glycosides also led to the development of mild extraction methods, as alternatives to the classical acid hydrolysis [23–26].



Figure 1. Images to illustrate the technique of liturgical embroideries worked in the Byzantine tradition. (**a**) Left, front side: detail of embroidery on satin where the metallic thread, which runs only on the obverse, is visible. (**b**) Right, reverse side: detail of the embroidery where the cellulosic support and the attachment points of the metal threads are visible.

With all these resources available, a vast amount of knowledge was acquired regarding the dyes used in various textiles from archaeological and historical contexts [27–29]. With reference to liturgical embroideries worked in the Byzantine tradition, the studies performed on textiles in Romanian collections evidenced that for the purple-red silk satin embroidery supports in objects dating from the second half of the 15th century, the combination of lac dye (*Kerria lacca*) and madder (*Rubia tinctorum* L.) was used for the warp, and redwood (*Caesalpinia* species, as, for example, *C. sappan*—now reclassified as *Biancaea sappan* or *C. echinata*—now *Paubrasilia echinata*) for the invisible weft. Kermes (*Kermes vermilio*) was responsible for the colour in the warp in a few samples, which correspond to very precious objects [4]. Starting from the last decades of the 16th century, warps in purple-red silk supports were dyed with Mexican cochineal (*Dactylopius coccus*) [4]. For the rare cases when blue silk satin supports were used in 16th century embroideries, indigo dyes were detected. A larger palette of colours and dyes, used individually or in combination, was identified in the embroidery threads in the studies performed on liturgical textiles in Romanian collections [2–5].

The present study aims to contribute to the existing knowledge on liturgical embroideries illustrating the theme of "Lamentation over Dead Christ" and dated between the 14th and the 19th centuries, using dye research. The objects had an important role in the Eastern Church ritual and this gives them the greatest artistic, historical and material value from the whole collection of liturgical embroideries of Byzantine tradition preserved in Romania. *Epitaphioi* and *aëres* belong to the same group of liturgical textiles but have different functions during the Orthodox religious service. *Epitaphioi* are mainly used on Good Friday when they are brought into the Church to symbolize Descending from the Cross and Lamentation and are then returned to the Altar and laid on the Table until Re-surrection (Figure 2). *Aëres* are used to cover the vessels and are then worn on the priest's shoulders during the ceremony of the presentation of the "holy gifts" [1,30] (Figure 3). The objects studied are dated over a period of about 500 years, with most of them from the 17th to the 19th century and are all conserved in the National Museum of Art of Romania (MNAR).



(**a**)



(**b**)

Figure 2. *Cont.*



(**c**)



(**d**)

Figure 2. *Cont.*



(**e**)

Figure 2. Examples of *epitaphioi* preserved in the National Museum of Art of Romania (MNAR): (**a**) *Epitaphios* dated 1437, Moldavia (inv. 15827_B182); (**b**) *Epitaphios* dated 1661–1662, Constantinople? (inv. 10718_B111); (**c**) *Epitaphios* dated 1679–1680, Wallachia (inv. 10683_B76); (**d**) *Epitaphios* dated 1752, Wien (inv. 15833_B188); (**e**) *Epitaphios* dated 19th century, Russian (inv. 10695_B88).



Figure 3. *Aër* dated ~1700, Greek? (inv. 10668_B61), worked in the Byzantine tradition, preserved in the National Museum of Art of Romania (MNAR).

2. Materials and Methods

2.1. Objects, Samples and Sampling Strategy

Twenty-four liturgical textiles were studied, all defined as *epitaphios* or *aër*. According to the technique, most of the objects (21/24) are embroideries in the Byzantine tradition, while two have the scene painted on a silk support (Figure 4), and in one other, it is represented in the Gobelin needlepoint technique. The textiles are dated between the 14th and the 19th century and are documented as Byzantine, Moldavian, Wallachian, Greek, Russian, of Viennese origin or from Constantinople (present-day Istanbul) (see Table 1). Fifty-seven samples were available for analysis. According to the sampling strategy, the specimens' withdrawal was limited to the degraded areas, as the objects reverse sides were not accessible because they were covered by linings. As a consequence, most of the samples (32/57) are from the textiles' supports, with samples being taken either from the main scene or from the frame, for all but four objects. For the other objects, samples are from embroidery threads (17/57) or from the original linings (8/57). One sample (1/57) comes from an ornamental silk cord frame. The present approach, which aims to document materials in the MNAR collection of liturgical textiles is very different compared with the previous ones performed in the same research group [2–5], where studies were correlated with restoration and more sampling areas were accessible.



Figure 4. *Epitaphios* dated 19th century, Greek (inv. 10869_Ț148), painted on silk support, preserved in the National Museum of Art of Romania (MNAR).

Table 1. Information about the liturgical textiles studied: inventory number, date, technique, brief description and workshop.

Object, Inventory Number	Date	Technique	Brief Description	Workshop
Epitaphios, inv. 15826_B181	1395–1396	embroidery	embroidery on blue satin support, frame with embroided inscription on blue support	Byzantine
Epitaphios, inv. 15827_B182	1437	embroidery	embroidery on red satin support, frame with embroided inscription on blue support	Moldavia
Epitaphios, inv. 15829_B184	1556	embroidery	embroidery on red satin support, frames with embroided inscription and decoration on red silk support	Moldavia
Epitaphios, inv. 15832_B187	1600–1601	embroidery	embroidery on red satin support, frame with embroided medalions on blue satin support	Russian
Epitaphios, inv. 11048_Ț327	1613	painting on silk support	painting on red satin support over a blue satin support, blue cotton lining (observed in the degraded areas), beige satin frame with traces of painting	Moldavia
Epitaphios, inv. 15830_B185	1628	embroidery	embroidery on dark green satin support, frame with embroided inscription on the same support, red lining	Moldavia
Epitaphios, inv. 15831_B186	1638	embroidery	embroidery on red satin support, two frames with embroided inscription and decoration respectively, both on green satin support, and a green velvet frame	Moldavia
Epitaphios, inv. 15845_B200	~1639	embroidery	embroidery on red satin support, frame with embroided decoration on the same red support; blue lining	Moldavia
Epitaphios, inv. 10718_B111	1661–1662	embroidery	embroidery and embroided frames on red satin support, embroided insriptions on green velvet frame (with interventions in some areas)	Constan-tinople?
Epitaphios, inv. 10683_B76	1679–1680	embroidery	embroidery on red satin support, frames with embroided inscription and medallions on the same support, green velvet frame	Wallachia
Epitaphios, inv. 16154_B288	1680–1681	embroidery	embroidery on red satin support, frames with embroided inscription and medallions on the same support, green velvet frame (not embroided); red satin lining	Wallachia
Aër, inv. 10668_B61	~1700	embroidery	embroidery on red—violet velvet, frame with embroided decoration on yellow satin applied on the velvet, pale blue cord frame	Greek?
Aër, inv. 10676_B69	end 17th c.	embroidery	embroidery on yellow satin support	Wallachia
Epitaphios, inv. 15835_B190	1751	embroidery	embroidery on brown satin support, frames with embroided medalions and inscription on the same support; yellow-green silk lining	Wallachia
Epitaphios, inv. 15833_B188	1752	embroidery	embroidery on green velvet support, decorative embroided frame on the same support	Wien
Epitaphios, inv. 15834_B189	1770	embroidery	embroidery and decorative embroided frame on velvet support	Wien

Table 1. *Cont.*

Object, Inventory Number	Date	Technique	Brief Description	Workshop
Epitaphios, inv. 10686_B79	1791	embroidery	embroidery on red velvet support, frames with embroided decoration, frame with tassels in the corners on the same velvet support; red lining (new?)	Wien
Epitaphios, inv. 15837_B192	end 18th c.	embroidery	embroidery on red satin support, frames with decoration and embroided inscription on the same red support and one more frame, everything mounted on a new blue satin support; red cotton intervention on the silk support	Moldavia?
Epitaphios, inv. 15836_B191	1823	embroidery	embroidery on pale blue support, frame with embroided inscription on pale blue support, red velvet decorated frame; fringes frame with tassels in the corners	Wien
Fragment of embroidery with "Lamentation", inv. 10673_B66	19th c.	embroidery	embroidery on satin support	Wien
Epitaphios, inv. 10695_B88	19th c.	embroidery	embroidery on red—violet velvet support, frames with inscription and decorations on the same support, yellow fringes frame	Russian
Epitaphios, inv. 10869_Ţ148	19th c.	painting on silk support	painting on red satin support centered to leave a (red satin) frame, yellow fringes frame	Greek
Epitaphios, inv. 11047_Ţ326	19th c.	gobelin needle-point technique	gobelin, frame with embroided inscription on ochre cotton support, yellow fringes frame	Romanian
Epitaphios, inv. 15825_B80	19th c.	embroidery and painting	painted central scene with embroided decorations on red—violet velvet support in the corners, frame with inscription on velvet support, yellow fringes frame	Russian

2.2. Sample Documentation and Preparation for Dye Analysis

Samples about 0.5–1 cm long were first observed under the digital microscope at ~50× magnification and non-destructively analysed by attenuated total reflectance infrared spectroscopy (FTIR-ATR) for fiber identification. Yarns of about 0.5 cm (~3 mg) were cut from the original samples, when larger specimens were available, and placed in Eppendorf vials for sample preparation.

Dyes were extracted from the yarns by acid hydrolysis. This was an assumed decision although the limits of this method, caused by the decomposition of glycosides to their parent aglycones, in the case of flavonoid dyes, were known [23–26]. The reason for choos-ing acid hydrolysis was the existing database, which contains information about natural dyes, used as standards or extracted from standard dyed yarns by acid hydrolysis. An amount of 200 µL of a mixture containing 37% HCl, CH_3OH and H_2O in a ratio of 2:1:1 ($v/v/v$) was added to each yarn, followed by incubation at 100 °C for 10 min. Samples were then evaporated to dryness in a vacuum desiccator. Each sample was re-dissolved in a 100 µL solution of CH_3OH/H_2O 1:1 (v/v) and centrifuged at 12,000 rpm for 10 min. The supernatants were transferred to chromatography vials and injected into the LC system. When the presence of indigo-based dyes was suspected, as in the cases of visually blue, green and black samples, a second extraction in 100 µL dimethyl sulfoxide (DMSO) was made, with the samples kept at 80 °C for 10 min. The two solutions were analysed separately.

2.3. Fiber Identification

Fiber documentation and image collection were made with a DinoLite digital microscope, model AM4113ZT. Further fiber investigation was made by infrared spectroscopy (FTIR-ATR), where a Bruker Optics Alpha spectrometer equipped with a Platinum ATR single reflection diamond ATR module was used. Spectra were acquired in the 4000–400 cm^{-1} domain, with a resolution of 4 cm^{-1}. Spectra collection and data processing were made with a dedicated software, Opus 7.0. Where FTIR indicated that cellulosic fibers were used, samples were also observed by optical microscopy for the clear identification of cotton.

2.4. Dye Analysis by Liquid Chromatography. Instrumentation and Parameters

All the samples were analysed by liquid chromatography with UV–Vis (diode array) detection (LC-DAD) on System 1. Some of the samples where carminic acid was identified were also examined on a similar configuration, further referred to as System 2, at the Royal Institute for Cultural Heritage (KIK/IRPA), in Brussels. These analyses were aimed at the attribution of the carminic acid-based insects down to the species level. More information about this will be given in Section 3.1., Supports, Red.

2.4.1. System 1 (LC-DAD)

An Agilent 1260 Infinity II series liquid chromatograph (Agilent Technology, USA) consisting of a quaternary pump (G7129A), a standard autosampler (G7111B), a column thermostat (G7116A) and a multi-channel diode array detector (G7115A) was used for dye analysis. OpenLAB CDS software was used for the chromatographic system control, data acquisition and processing. A Zorbax C18 column, of 150 mm length, 4.6 mm i.d. and 5 µm particle size, thermostated at 40 °C, was used. The mobile phase consisted of a mixture of aqueous 0.2% (*v*/*v*) formic acid (solvent A) and methanol/acetonitrile (1:1, *v*/*v* as solvent B). Gradient elution was applied by using the following profile: at 0 min, 15% solvent B; from min 0 to 5, linear increase to 25% solvent B; from min 5 to 10, constant at 55% solvent B; from min 10 to 16, linear increase to 100% solvent B; from min 16 to 18, constant at 100% solvent B; and step jump at 15% solvent B, with 5 min re-equilibration period between runs (post-time). The flow rate was set at 0.8 mL/min and the injected volume of the sample was 10 µL. The UV–Vis spectra were acquired in the range from 200 to 900 nm, with a simultaneous monitoring at five wavelengths (255, 275, 295, 420 and 490 nm), having a frequency of 0.03 min and a resolution of 2 nm.

2.4.2. System 2 (LC-DAD, with a Specific Method Constructed for Species Attribution of Carminic Acid-Based Insects)

High-performance liquid chromatography with diode array detection (DAD) was performed with an ACQUITY Arc HPLC system (Waters Chromatography n.v.), with a column heater/cooler and a quaternary solvent manager, a 2998 PDA with a low-dispersion flowcell detection system and Empower3 data handling software. The solvents used were (A) methanol (for HPLC > 99.8%), (B) a mixture of 1/9 (*v*/*v*) methanol/Milli-Q water and (C) 0.5% phosphoric acid (85 wt% p.a.). The analysis was performed at a flow rate of 1.2 mL/min with the following gradient: isocratic state 23A/67B/10C for 0–3 min, linear gradient to 90A/0B/10C between 3 and 29 min and isocratic 23A/67B/10C from 30 to 35 min. For the stationary phase, a temperature-controlled column of LiChrosorb RP-18 with a 125 mm x 4 mm diameter end cap with 5 µm particle size and 100 Å pore diameter was used. Calculation of the relative percentage of anthraquinone dyes was made after integration of the areas in the chromatograms registered at 255, 275, 290, 420 and 500 nm. More information regarding the methodology used is given below (see Section 3.1. Supports, Red) and an in-depth description of the method can be read in an earlier publication [31].

2.4.3. Dye Attribution—Databases

Dyes were attributed based on their retention and UV–Vis data, according to information collected on standards, dyes and dyed yarns. MS data were also considered for

identification when the samples were investigated by LC-DAD-MS. As described in previous publications [32,33], biological source attribution was based on data collected on yarns dyed in the laboratory, by following traditional dyeing recipes. Analytical data (retention and UV–Vis spectra) and the corresponding biological sources of the natural dyes discussed in the present study are given in Table 2.

Table 2. Dyes identified in the present study, their biological sources and identification criteria: retention and UV–Vis maximum.

Dye	Abb.	Biological Source Common and Latin Name(s)	Retention (min.)	UV-Vis Data (nm)
Alizarin	al	Madder (*Rubia tinctorum* L.) Synthetic alizarin (1869)	15.4	202; 248; 278; 430
Anthrapurpurin (1,2,7-trihydroxyanthraquinone)	anthra	Synthetic alizarin (1869)	13.6	274; 338; 428
Apigenin	ap	Dyer's broom (*Genista tinctoria* L.) Weld (*Reseda luteola* L.)	13.9	210; 268; 336
Dehydro-brazilein product (also called Type B)	bra′	Redwood based dye (*Caesalpinia* species, see text)	9.2	238; 260; 322; 384; 452
Carminic acid	ca	Carminic acid-based dye Mexican cochineal (*Dactylopius coccus* C.) Armenian carmine scale insect (*Porphyrophora hameli*) Polish carmine scale insect (*Porphyrophora polonica*) (see text)	8.5	226; 276; 310; 494
Chrysoeriol	chry	Weld (*Reseda luteola* L.)	14.2	206; 268; 348
Ellagic acid	ea	Tannins	10.3	254; 366
Flavopurpurin (1,2,6-trihydroxyanthraquinone)	flavo	Synthetic alizarin (1869)	13.8	272; 406
Genistein	ge	Dyer's broom (*Genista tinctoria* L.)	13.5	208; 260
Haematein derivative (also called Type H)	hae′	Logwood (*Haematoxylun campechianum* L.)	7.9	244; 276; 330; 456
Indigotin	ind	Indigo-based dye (see text) Woad (*Isatis tinctoria)* or Indigo (*Indigofera* sp.)	16.2	238; 285; 330; 610
Luteolin	lu	Dyer's broom (*Genista tinctoria* L.) Sawwort (*Serratula tinctoria* L.) Weld (*Reseda luteola* L.)	13.0	208; 254; 266; 348
Urolithin C	srw	Redwood based dye (*Caesalpinia* species, see text)	10.8	238; 306; 336

2.5. Elemental Analysis

In two cases, when the presence of iron needed to be confirmed, the samples were investigated by X-ray fluorescence spectrometry (XRF). Elemental analysis was performed with a portable XRF spectrometer Bruker S1 TITAN Model 600, with the following specifications: rhodium (Rh) tube, silicon drift chamber detector (SDD) and 5 mm spot size. The system used was air-path, with an elemental range $Z > 12$ (Mg). Energy was set at 40 keV.

3. Results

Results will be discussed according to the visual colours of the yarns (Table 3) and the function of the fibers (support, lining, embroidery thread (Table 4)) and will be correlated with similar data obtained in previous studies of liturgical embroideries in the Byzantine tradition, in Romanian collections and beyond.

Table 3. Samples (textile, colour and description of the type of thread), dyes identified and their biological sources. Objects are listed in chronological order. For more information regarding the objects, see Table 1.

Epitaphios, inv. 15826_B181 (1395–1396)						
1.	15826_P1	blue	silk	support, warp	ind	Indigo based dye
2.	15826_P2	blue	silk	support, weft	ind	indigo based dye
Epitaphios, inv. 15827_B182 (1437)						
3.	15827_P1	blue	silk	embroidery thread	ind	indigo based dye
4.	15827_P3	red	silk	embroidery thread	ca	carminic acid based dye
Epitaphios, inv. 15829_B184 (1556)						
5.	15829_P1	red	silk	support, warp	ca	carminic acid based dye
Epitaphios inv. 15832_B187 (1600–1601)						
6.	15832_P2	blue	silk	support (frame)	ind	indigo based dye
Epitaphios, inv. 11048_Ț327 (1613)						
7.	11048_P1	yellow	silk	red support, weft	urolithin C, ea	redwood based dye and tannins
8.	11048_P2	red	silk	red support, warp	ca	carminic acid based dye
9.	11048_P3	pale blue	silk	blue support, weft (frame)	ind	indigo based dye
10.	11048_P5	blue	cotton	lining, weft	ind	indigo based dye
Epitaphios, inv. 15830_B185 (1628)						
11.	15830_P1	brown-green	silk	support, weft	ind	Indigo based dye (in a dyeing combination with a yellow dye (under detection limit)
12.	15830_P2	red	silk	lining	ca	carminic acid based dye
13.	15830_P3	dark blue	silk	support, warp	ind	indigo based dye
Epitaphios, inv. 15831_B186 (1638)						
14.	15831_P1	red	silk	red support, warp	ca,	carminic acid based dye
15.	15831_P2	green	silk	green satin support, weft	lu, ge, ap, ind	dyer's broom and indigo based dye
Epitaphios, inv. 15845_B200 (~1639)						
16.	15845_P1	yellow	silk	support, weft	urolithin C	redwood based dye
17.	15845_P2	blue	silk	lining	ind	indigo based dye

Table 3. *Cont.*

Epitaphios, inv. 10718_B111 (1661–1662)						
18.	10718_P1	yellow	silk	red support, weft	urolithin C	redwood type
19.	10718_P2	red	silk	red support, warp	ca, ea	carminic acid based dye and tannins
Epitaphios from Cotroceni, inv. 10683_B76 (1679–1680)						
20.	10683_P1	yellow	silk	red support, weft	urolithin C	redwood based dye
21.	10683_P2	red	silk	red support, warp	ca	carminic acid based dye
Epitaphios from Tismana, inv. 16154_B288 (1680–1681)						
22.	16154_P2	yellow	silk	red support, weft	urolithin C	redwood based dye
23.	16154_P3	yellow	silk	lining, weft	urolithin C	redwood based dye
24.	16154_P4	red	silk	lining, warp	ca	carminic acid based dye
25.	16154_P5	green	silk	green velvet plush (frame)	lu, ge, ap, ind	dyer's broom and indigoid based dye
Aër, inv. 10668_B61 (~1700)						
26.	10668_P1	yellow-pink	silk	yellow satin support (frame)	urolithin C and bra' (type B)	redwood based dye
27.	10668_P3	pale blue	silk	blue cord	ind	indigo based dye
Aër, inv. 10676_B69 (end 17th c.)						
28.	10676_P1	yellow	silk	support	lu, ge, ap	dyer's broom
Epitaphios, inv. 15835_B190 (1751)						
29.	15835_P1	yellow-green	silk	embroidery thread (silk core in metallic thread)	lu, ge, ap	dyer's broom
30.	15835_P2	brown	silk	support, warp	ea	tannins
31.	15835_P3	yellow-green	silk	lining	lu, ap, chry	weld
Epitaphios, inv. 15833_B188 (1752)						
32.	15833_P1	green	silk	velvet	lu, ap, chry, ea ind	weld and tannins indigo based dye
33.	15833_P2	yellow	silk	embroidery thread (silk core in metallic thread)	lu, ge, ap	dyer's broom
Epitaphios, inv. 15834_B189 (1770)						
34.	15834_P1 galben	yellow	silk	embroidery thread (silk core in metallic thread)	lu, ge, ap,	dyer's broom
35.	15834_P2	red	silk	velvet, warp	Bra' (type B) urolithin C	redwood type
36.	15834_P3	red	silk	velvet, plush	urolithin C, bra' (type B)	redwood type
37.	15834_P5	green	silk	embroidery thread	lu, ge, ap, ind	dyer's broom and indigo based dye
38.	15834_P6	green	silk	embroidery thread	lu, ge, ap, ind	dyer's broom and indigo based dye

Table 3. *Cont.*

Epitaphios, inv. 10686_B79 (1791)						
39.	10686_P2	pale yellow	silk	embroidery thread (silk core in metallic thread)	lu, ge, ap	dyer's broom
40.	10686_P4	green	silk	embroidery thread	lu, ind	luteolin based dye and indigo based dye
41.	10686_P5	red	silk	velvet lining (new?)	ca, ea	carminic acid based dye and tannins KIK/ IRPA: Mexican Cochineal and tannins
Epitaphios, inv. 15837_B192 (end 18th c.)						
42.	15837_P2	red	silk	support (frame with inscription), warp	ca	carminic acid based dye
43.	15837_P3	red	cotton	support (new?) (central area)	al, anthra, flavo	synthetic alizarin (1869)
Epitaphios, inv. 15836_B191 (1823)						
44.	15836_P1	black	silk	embroidery thread (with metallic thread)	ea	tannins
45.	15836_P2	green	silk	embroidery thread	lu, ge, ap, ea	dyer's broom
46.	15836_P4	yellow-pink	silk	velvet, plush (support, frame)	urolithin C, bra' (type B)	redwood based dye
Fragment of embroidery with " Lamentation ", inv. 10673_B66 (19th c.)						
47.	10673_P1	green	silk	embroidery thread	lu, ge, ap	dyer's broom
Epitaphios, inv. 10695_B88 (19th c.)						
48.	10695_P1	red	silk	velvet, warp	ca, ea	carminic acid based dye and tannins KIK/ IRPA: Mexican Cochineal and tannins
49.	10695_P2	red	silk	velvet, plush	ca, ea	carminic acid based dye and tannins
50.	10695_P3	blue	cotton	lining, warp	Prussian Blue *	Prussian Blue * (1704)
Epitaphios, inv. 10869_Ţ148 (19th c.)						
51.	10869_P1	yellow	silk	support, weft	urolithin C	redwood based dye
52.	10869_P2	red	silk	support, warp	ca, ea	carminic acid based dye and tannins
Epitaphios, inv. 11047_Ţ326 (19th c.)						
53.	11047_P2	red	silk	embroidery thread	ca, ea	carminic acid based dye and tannins
54.	11047_P4	green	silk	embroidery thread	lu, ap, chry, ind	weld and indigo based dye

Table 3. *Cont.*

55.	11047_P5	pale violet	silk	embroidery thread	hae′ (type H) bra′ (type B) urolithin C	redwood based dye logwood based dye
56.	11047_P6	grey	silk	embroidery thread	ind	indigo based dye
Epitaphios, inv. 15825_B80 (19th c.)						
57.	15825_P1	red	silk	velvet	ca	carminic acid based dye

* Note: Prussian blue was revealed by FTIR-ATR (specific CN signal at 2083 cm^{-1}) and confirmed by the presence of Fe in the XRF spectrum.

Table 4. Objects (listed in chronological order) and summary of results, presented separately for the three fibre functions: support, lining, embroidery thread.

Object, Inventory Number/ Technique	Date	Workshop	Results		
			Support	Lining	Embroidery Threads
Epitaphios, inv. 15826_B181, embroidery	**1395–1396**	Byzantine	Indigo based dye (satin)		
Epitaphios, inv. 15827_B182, embroidery	**1437**	Moldavia			Indigo based dye Carminic acid dye
Epitaphios, inv. 15829_B184, embroidery	**1556**	Moldavia	Carminic acid based dye (satin)		
Epitaphios, inv. 15832_B187, embroidery	**1600–1601**	Russian	Indigo based dye blue (satin) (frame)		
Epitaphios, inv. 11048_Ț327, painting on silk support	**1613**	Moldavia	Carminic acid based dye (satin) Indigo based dye (satin)	Indigo based dye (silk)	
Epitaphios, inv. 15830_B185, embroidery	**1628**	Moldavia	Indigo based dye (satin)	Carminic acid based dye (silk)	
Epitaphios, inv. 15831_B186, embroidery	**1638**	Moldavia	Carminic acid based dye (satin) Indigo based dye and dyer's broom (satin) (frame)		
Epitaphios, inv. 15845_B200, embroidery	**~1639**	Moldavia	Redwood based dye (weft) (satin)	Indigo based dye (silk)	
Epitaphios, inv. 10718_B111, embroidery	**1661–1662**	Constan-tinople?	Carminic acid based dye (satin)		
Epitaphios, inv. 10683_B76, embroidery	**1679–1680**	Wallachia	Carminic acid based dye (satin)		
Epitaphios, inv. 16154_B288, embroidery	**1680–1681**	Wallachia	Redwood based dye (weft) (satin) Indigo based dye and dyers broom (satin) (frame)	Carminic acid based dye (silk)	
Aër, inv. 10668_B61, embroidery	**~1700**	Greek?	redwood based dye (satin) (frame) Indigo based dye blue cord		
Aër, inv. 10676_B69, embroidery	**end 17th c.**	Wallachia	dyers broom (satin)		

Table 4. *Cont.*

Object, Inventory Number/ Technique	Date	Workshop	Results		
			Support	Lining	Embroidery Threads
Epitaphios, inv. 15835_B190, embroidery	**1751**	Wallachia	tannins (satin)	weld (silk)	dyers broom
Epitaphios, inv. 15833_B188, embroidery	**1752**	Wien	Indigo based dye (velvet)		dyers broom
Epitaphios, inv. 15834_B189, embroidery	**1770**	Wien	redwood type (velvet)		dyers broom dyers broom and indigo based dyers broom and indigo based
Epitaphios, inv. 10686_B79, embroidery	**1791**	Wien		Mexican cochineal silk and tannins (silk)	dyers broom luteolin based dye and indigo based
Epitaphios, inv. 15837_B192, embroidery	**end 18th c.**	Moldavia?	Carminic acid based dye (satin) Synthetic alizarin support central area (intervention)		
Epitaphios, inv. 15836_B191, embroidery	**1823**	Wien	Redwood based (velvet)		tannins dyers broom
Fragment of embroidery "Lamentation", inv. 10673_B66, embroidery	**19th c.**	Wien			dyers broom
Epitaphios, inv. 10695_B88, embroidery	**19th c.**	Russian	Carminic acid based dye (velvet)	Prussian blue (cotton)	
Epitaphios, inv. 10869_Ț148, painting on silk support	**19th c.**	Greek	Carminic acid based dye red		
Epitaphios, inv. 11047_Ț326, gobelin	**19th c.**	Romanian			Carminic acid based and tannins weld and indigo based Redwood based and logwood based dyes indigo based
Epitaphios, inv. 15825_B80, embroidery and painting	**19th c.**	Russian	Carminic acid based dye red (velvet) (frame)		

3.1. Supports

In total, 32 samples from the supports in 20 liturgical textiles were available, which means they were from all but four of the textiles under discussion. In the present publication, the term "support" is used in a larger sense to include un-embroidered frames.

3.1.1. Red

Due to the high liturgical symbolism, red is the most common support in Christian Medieval Europe, regardless of the production area. About half of the supports in the *epitaphioi* and aëres studied (14/20) are red and the material was characterized as satin in most cases (10/14) and velvet in the remaining cases (4/14). With reference to textiles in the present study, red satin support corresponds to liturgical textiles worked in the Byzantine technique dated from 1556 to the end of the 18th century and red velvet supports are correlated with similar objects from 1770 to the 19th century. This perfectly aligns with the scientific literature which states that in the embroideries of Byzantine tradition, satin supports are replaced by velvet starting from the 16th–17th centuries [8]. Red satin was also used as the support in two painted *epitaphioi*, dated 1613 and the 19th century, respectively. The poor conservation status of *epitaphios* inv. 11048 (dated 1613, painted), enabled a detailed investigation of the support. Microscopic observation of both sides of

the red satin revealed that it was woven with a red warp and a yellow, or more correctly yellow-pink, weft (Figure 5). Carminic acid was the main dye component identified for the red warp, according to retention and the UV–Vis spectrum (λmax = 226; 276; 310; 494 nm). Carminic acid is the main dye component in several species of insects, with *Porphyrophora* species from the Old World and *Dactylopius coccus* (Mexican cochineal) from the New World as the main representatives. Identification down to the species level is sometimes possible based on the calculation of the relative amount between the minor compounds, dcII (the 2-C-glucoside of flavokermesic acid), kermesic and flavokermesic acids, with re-ference to carminic acid [34–37]. For the seven samples in this study where carminic acid was identified in red satin warps, no minor components were detected, which made any precise attribution of the species impossible.

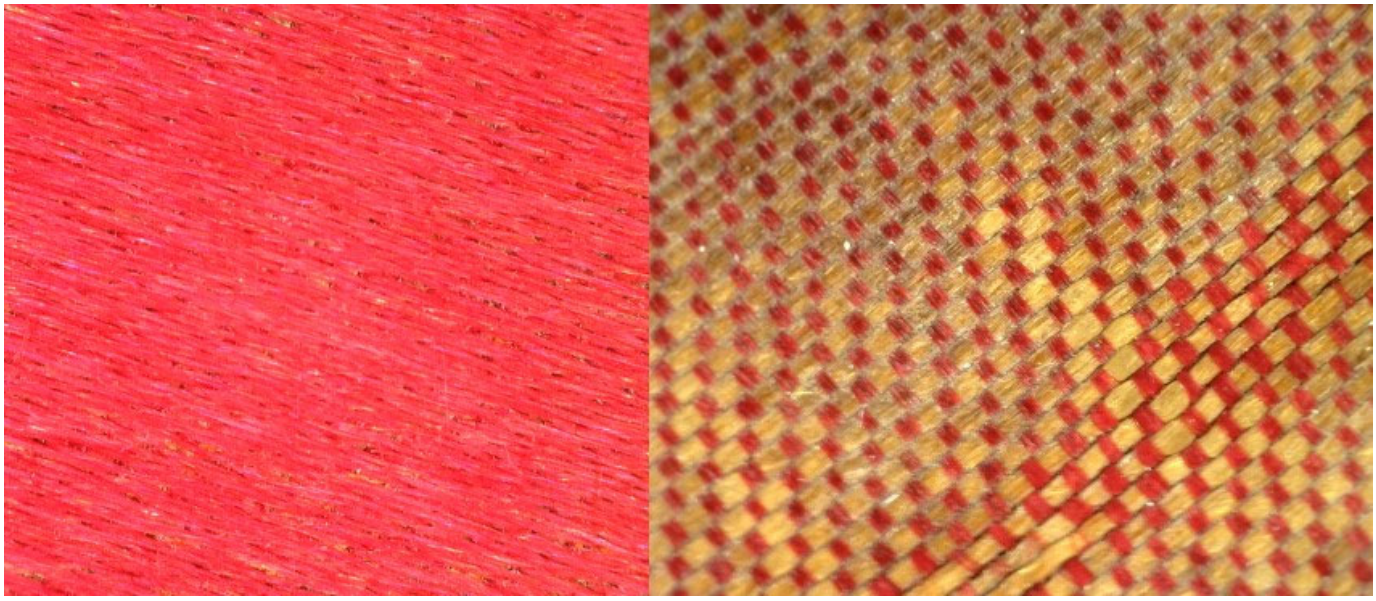


Figure 5. Image (~50x) to illustrate the weaving structure in the red satin support in *Epitaphios* inv. 11048. (**Left**) Front side where only the red warp dyed with carminic acid-based dye is visible. (**Right**) Back side where both the red warp and the redwood dyed yellow weft could be observed.

Urolithin C, whose structure was only recently elucidated [38], was identified in the corresponding yellow-pink wefts, as well as in wefts from satin supports where no sample was available from the warp (inv. 15845). Its presence was revealed by retention and the UV–Vis spectrum (λmax = 238; 306; 336 nm) and suggests a soluble redwood species was used for dyeing. The above-mentioned dye component, also called "srw" or "type C" [39,40], was considered by the dye expert community as representative of the recognition of the soluble redwood dyeings for many years before its structure was decoded. Soluble redwoods contain brazilin as the main dye component, which oxidizes to brazilein, and transforms through acid hydrolysis to a brazilein derivative, coded "bra' (bra prime)" or "type B" (λmax = 238; 260; 322; 384; 452 nm) [38], characterized as a dehydro-brazilein product [17]. Analysis performed on historical objects may not reveal the brazilein deri-vative due to its severe degradation, and attributions of redwood dyeings are in many cases based on the detection of urolithin C, thus a marker compound, which remains vis-ible in historical samples, even if the dehydro-brazilein product is gone [40]. It should be supposed that the original colour of the yellow-pink redwood dyed yarns was red.

In contrast to the samples discussed above when only urolithin C was present in yellow or yellow-pink yarns, in others described as red, the brazilein derivative bra' (also called type B) was detected. These samples come from the warp and plush in velvet supports in *epitaphios* inv. 15834 (dated 1770). Brazilein derivative was also evidenced together with

urolithin C in a yellow-pink sample from the satin in *epitaphios* inv. 10668 (dated ~1700) and from the velvet plush in *epitaphios* inv. 15836 (dated 1823). Figure 6 illustrates the difference in the analytical results of redwood dyed red and yellow yarns. While urolithin C (238; 306; 336 nm) is present in both cases, detection of the dehydro-brazilein product (bra') (238; 260; 322; 384; 452) is associated with the red colour of the sample.

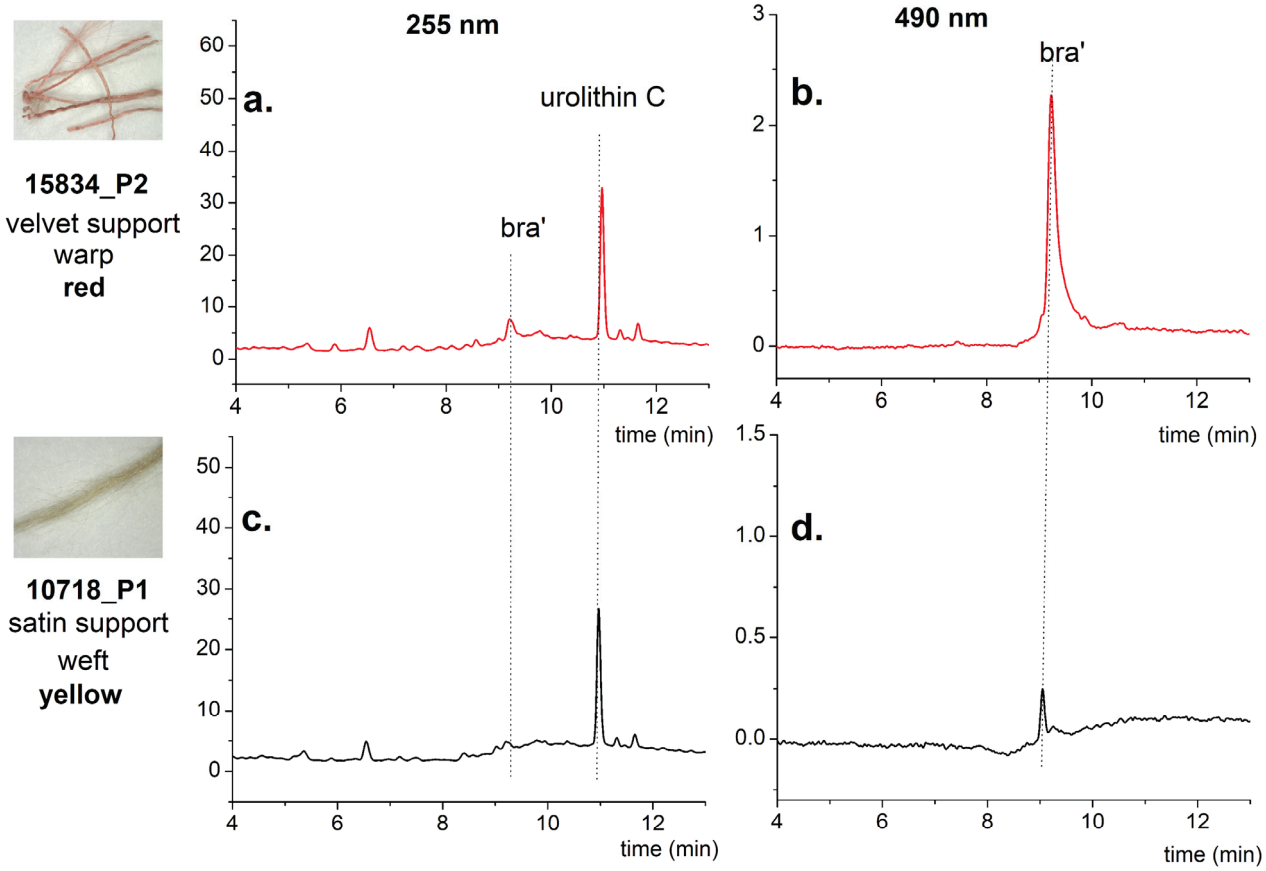


Figure 6. Comparative chromatograms of samples 15834_P2 (velvet support, warp), red ((**a**,**b**) top images) and 10718_P1 (satin support, weft) yellow ((**c**,**d**) bottom images). While in both samples urolithin C (238; 306; 336 nm) is present, detection of the dehydro-brazilein product (bra') (238; 260; 322; 384; 452) in 15834_P2 should be associated with the red colour of the sample. Details of the region between 4 and 13 min. Left: 255 nm; right: 490 nm.

Three samples from red velvet supports in two 19th century *epitaphioi* were available, and carminic acid was identified in all cases, for warp and plush (10695_P1 and P2; 15825_P1). Analysis performed on sample 10695_P1 (warp) at the Royal Institute for Cultural Heritage in Brussels (KIK/IRPA), where a procedure to discriminate between the carminic acid insect dyes was set up [37], revealed the presence of minor components dcII (the 2-C-glucoside of flavokermesic acid), and kermesic and flavokermesic acids. Furthermore, based on calculation of the ratio between the amounts of flavokermesic acid C glycoside and the total percentage of flavokermesic and kermesic acids, relative to carminic acid, it was proved that *Dactylopius coccus* (Mexican cochineal) was used for dyeing (Figure 7). More details regarding the procedure used was given in an earlier publication [37].

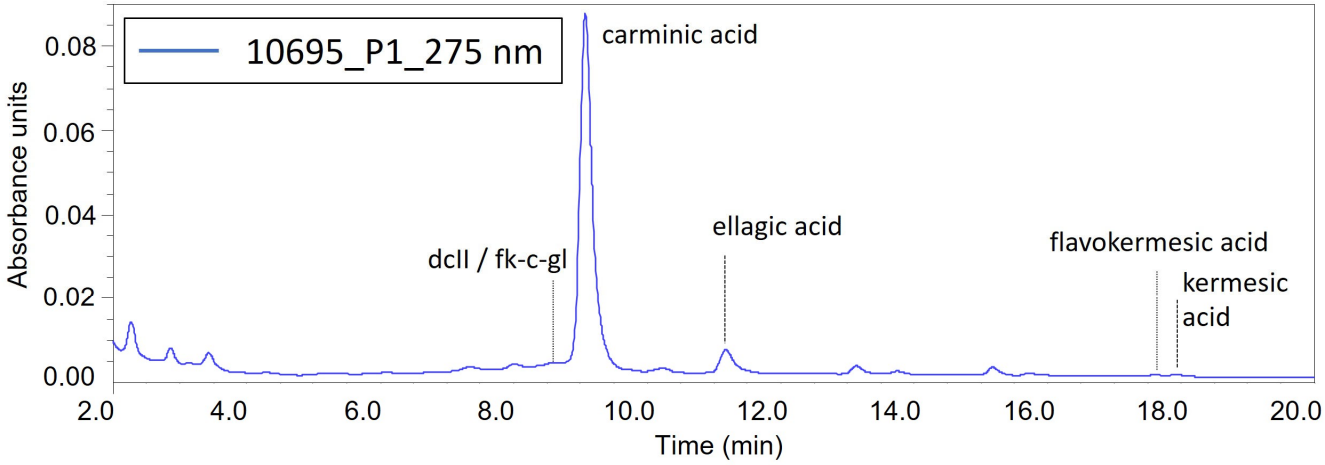


Figure 7. Chromatogram (registered at 275 nm) to support identification of Mexican cochineal in sample 10695_P1 based on the detection of carminic acid and the minor components dcII (flavokermesic acid C glycoside), flavokermesic and kermesic acids.

As already mentioned, red satin was the most common support for *epitaphioi* worked in the Byzantine tradition. In the present study, it was observed in embroidered liturgical textiles dated from the 15th to the 18th century, from various workshops: Moldavian, Wallachian, Russian and from Constantinople as well as in painted *epitaphios* from Moldavian (17th century) and Greek workshops (19th century). In previous studies which referred to Byzantine liturgical embroideries from the Romanian provinces Moldavia and Wallachia, the combination of lac dye and madder was revealed for the weft in the 15th and the 16th century satin supports, while the preference for carminic acid-based dyes was observed from the late 16th century onwards. The small amounts of samples in the present approach made any attribution down to the species level impossible. However, it may be supposed that Mexican cochineal was used in these textiles, as it has been described as having replaced all the other insect dyes in European textiles from about 1570 [36]. This hypothesis is also supported by analysis performed on the support (warp) in a Byzantine liturgical embroidery preserved in Putna Monastery, dated to the late 16th century, where Mexican cochineal was identified in the presence of tannins [3].

One sample, visually red in colour, was available from the central area of *epitaphios* inv. 15837 (dated end 18th century), from a zone which according to visual and microscopic observations represents a later intervention. Analysis performed on the cotton red yarn evidenced the presence of alizarin (λmax = 202; 248; 278; 430 nm), anthrapurpurin (1,2,7-trihydroxyanthraquinone) (λmax = 274; 338; 428) and flavopurpurin (1,2,6-trihydroxyanthraquinone) (λmax = 272; 406), which suggests the use of synthetic alizarin. As the synthetic dye became available only in 1869, it becomes clear that the *epitaphios* was repaired and the intervention took place after the date mentioned.

3.1.2. Blue and Green

Samples were available from five blue or green silk satin and velvet supports, including frames in the respective colours. As in the case of red, satin was observed in objects dated from 1395-96 to 1638 and velvet in those dated later, more exactly, 1680 and 1752. For the blue satin supports, when samples from both weft and warp were analysed, indigotin was identified in both, as, for example, in *epitaphios* inv. 15826 from Cozia Monastery (dated 1395-96). Indigotin was detected based on retention associated with the UV–Vis spectrum (λmax = 238; 285; 330; 610 nm) and suggests that indigo-based dyes were used for dyeing. There are several species of plants which contain precursors of indigotin. *Isatis tinctoria* (woad), which was cultivated in Europe for a long time, and *Indigofera* species (indigo), which have a long history of use in Europe [10], would be the most probable candidates. However, with the analytical instrumentation and existing knowledge, it is not possible

to distinguish between the various indigo-containing plants, nor between natural and synthetic indigo [41,42]. From the theoretical perspective, both woad and indigo could have been used.

When embroidery satin supports with green hues were intended, a combination of indigo-based and yellow flavonoid dyes was used for the weft, as for *epitaphios* inv. 15831 (dated 1638). Luteolin, genistein and apigenin were evidenced based on retention and UV-Vis spectra (λmax = 208; 254; 266; 348 nm, λmax = 208; 260 nm and λmax = 210; 268; 336 nm) in the acid hydrolysed extract of 15831_P2, and indigotin in the di-methyl-sulfoxide extracted solution, which suggests that *Genista tinctoria* (dyer's broom) was used for dyeing. The same biological source was evidenced in the green velvet frame and pile threads (plush) in *epitaphios* inv. 16154 (dated 1680–1681). The yellow dye source was combined with indigo dyes to achieve green. For the green velvet support in *epitaphios* inv. 15833 (dated 1752), luteolin and apigenin were identified in the presence of chrysoeriol (λmax = 206; 268; 348 nm), which suggests the use of *Reseda luteola* (weld) (Figure 8).

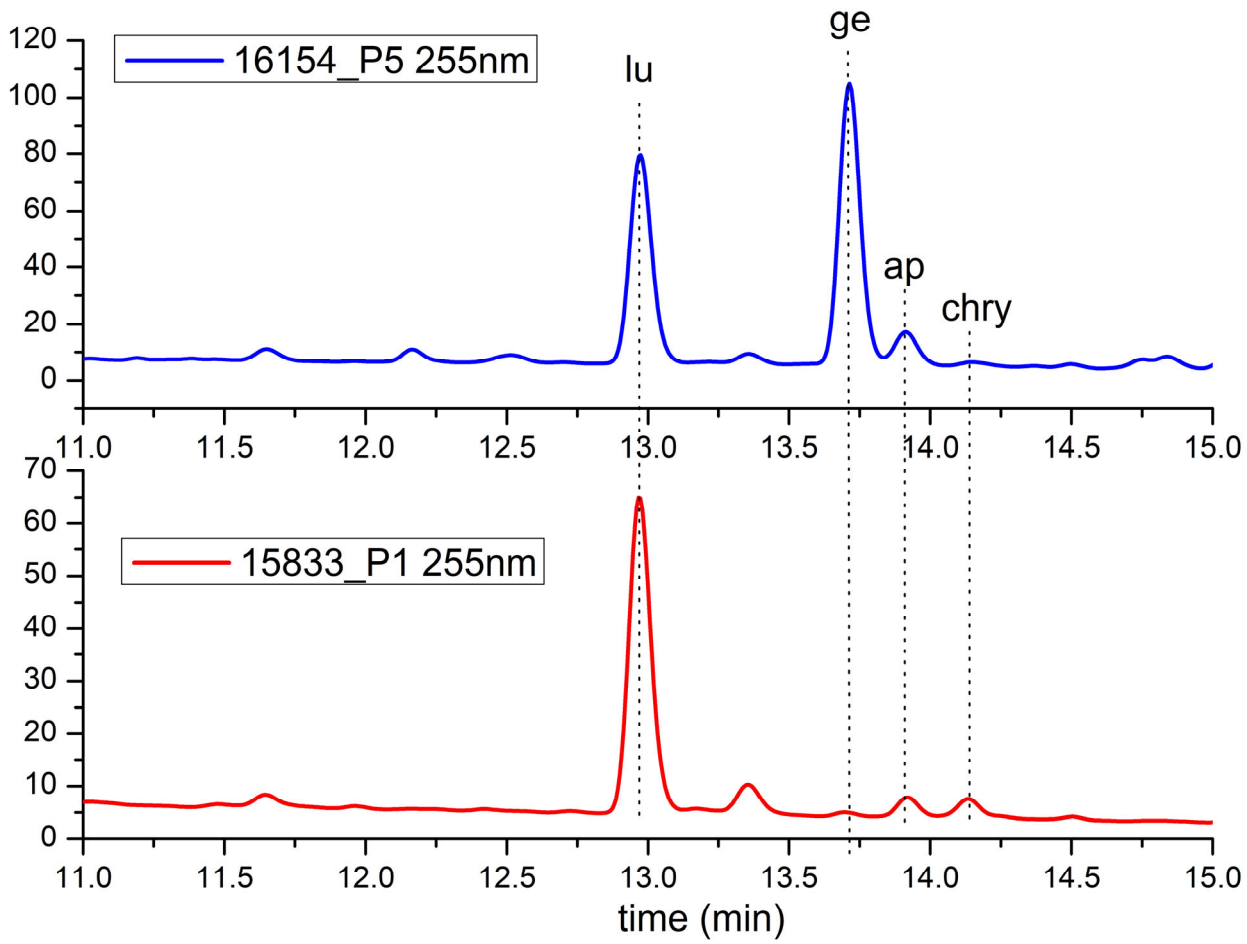


Figure 8. Comparative chromatograms (registered at 255 nm) of samples 16154_P5 (velvet plush) and 15833_P1 (velvet) to suggest identification of dyer's broom (luteolin, genistein, apigenin) and weld (luteolin, apigenin, chrysoeriol). Detail from 11 to 15 min.

3.1.3. Yellow

The three dyes representative of dyer's broom were identified in a yellow satin support used as a decorative frame in *aër* inv. 10676 (dated to the 17th century), the only support in this colour in the actual group of objects.

3.1.4. Brown

A brown silk sample from the warp in the embroidery satin support in *epitaphios* inv. 15835, dated 1751, was selected for analysis. The presence of ellagic acid (λmax = 254; 366 nm) suggested that tannins were used for dyeing. Tannins are substances of vegetal origin, very common in the plant world, which give a brown-black hue in the presence of iron salts. The use of iron was confirmed by elemental analysis performed non-destructively by XRF (see Section 2.5.). Although very colourfast dyes are produced by tannin dyeing in the presence of iron, the mordant gradually corrodes and degrades the fibers [10]. This process was also observed for *epitaphios* inv. 15835.

3.2. Linings

Eight samples from linings in seven objects were available for analysis, with all but two characterized as silk.

3.2.1. Red

Carminic acid was detected in the red satin in two linings, from *epitaphios* inv. 15830 (dated 1628) and inv. 16154 (dated 1680–1681) (weft), as well as in the red velvet in *epitaphios* inv. 10686 (dated 1791). Analysis performed on the latter, at the Royal Institute for Cultural Heritage in Brussels, evidenced the presence of minor components dcII (the 2-C-glucoside of flavokermesic acid), and kermesic and flavokermesic acids, and proved that Mexican cochineal was used for dyeing.

Urolithin C was identified in a yellow silk sample from the weft in the lining of *epitaphios* inv. 16154 (dated 1680–1681), which suggests the use of soluble redwood. As carminic acid-based dyes were identified for the warp (see text above) in the same weaving, it is clear that the satin lining is similar to that used in the red embroidery supports discussed in Section 3.1. Furthermore, it should be noted that the satin lining is similar to the support in *epitaphios* inv. 10683, documented in the museum archives as donated by the same prince to a different monastery, and dated 1679–1680.

3.2.2. Blue

Indigo-based dyes were detected in a blue silk lining in *epitaphios* inv. 15845 (dated ~ 1639). As explained earlier, when blue embroidery supports were discussed, either woad or indigo could have been used for dyeing.

Two samples from blue cotton linings were available for analysis. Indigo-based dyes were detected in the painted *epitaphios* inv. 11048 (dated 1613) and the Prussian blue pigment in *epitaphios* inv. 10695 (dated 19th century). Prussian blue was identified based on the specific carbon–nitrogen bond at 2083 cm^{-1} in the attenuated total reflectance infrared spectroscopy (FTIR-ATR) and confirmed by the detection of iron in the elemental analysis by X-ray fluorescence spectrometry (XRF) (Figure 9). Prussian Blue, a pigment with the chemical formula $Fe_4[Fe(CN)_6]_3$, was discovered in the early 18th century; although mainly used as pigment, it became a popular alternative to indigo in textile dyeing in the 18th and 19th centuries [43]. With reference to textiles in Romanian collections, this statement was proved by its identification, in dyeing combinations with Mexican cochineal or yellow dye sources, in black wool threads used in the decoration of 19th century traditional shirts or sheepskin coats from Sibiu, Transylvania [44,45].

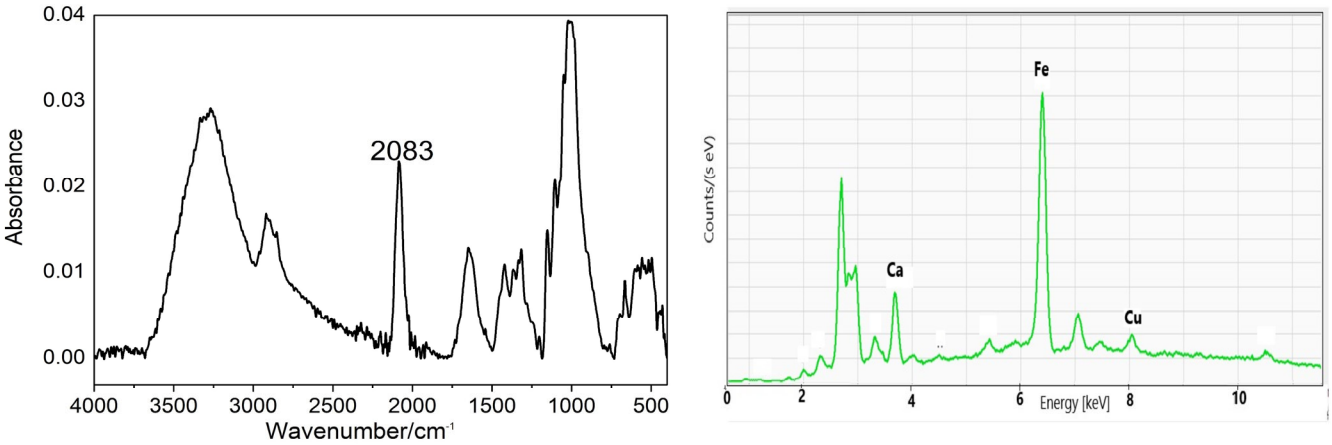


Figure 9. Image to support identification of Prussian Blue ($Fe_4[Fe(CN)_6]_3$) on cotton, in *Epitaphios* inv. 10695 (19th century). (**Left**) FTIR-ATR image where the specific carbon–nitrogen bond at 2083 cm^{-1} could be observed. (**Right**) XRF image where the presence of iron is confirmed.

3.2.3. Yellow

For the only yellow-green silk lining in *epitaphios* inv. 15835 (dated 1751), luteolin, apigenin and chrysoeriol were identified, which suggests the use of weld.

3.3. Embroidery Threads

Sixteen samples of embroidery threads were available from eight *epitaphioi*, one dated 1437 and the others from the 18th and 19th century.

3.3.1. Red and Violet

Two red samples representing embroidery threads in two different *epitaphioi* were studied. Carminic acid-based dyes were detected in both cases, the first dated 1437 (*epitaphios* inv. 15827) and the other 19th century (*epitaphios* inv. 15825). In none of the cases was it possible to establish the species of the insect used. However, it may be supposed that for the object dated 1437, a date which is before the arrival of the Mexican insects in Europe, either the Armenian or the Polish carmine scale insects were used.

In one case only were the dye components in redwood (urolithin C and brazilein derivative) and the hematein derivative coded hae' (also called Type H) detected. For the latter, identification was made based on retention time and UV–Vis data (λmax = 244; 276; 330; 456 nm), which were compared with information available in the literature [17,46]. The compounds were evidenced in a visually pale violet embroidery thread from the only *epitaphios* in the present study, which is worked in the Gobelin needlepoint technique (inv. 11047, dated 19th century). Logwood was identified in previous studies in liturgical embroideries in the Byzantine tradition from Romanian collections, dated to the 18th and 19th centuries [47], as well as in similar 19th century textiles preserved at the Holy Mount Athos [17]. In some of these detections, logwood was also determined in dyeing combination with redwood type. The identification of logwood in 18th and 19th century European textiles is not surprising considering that the biological source is native from South America [10].

3.3.2. Blue, Green and Yellow

Indigo-based dyes were identified in five cases, twice in blue and three times in green samples. In all the cases when a green hue was intended, indigoid dyes were detected together with dyer's broom. The flavonoid dye source was also responsible for the colour in four embroidery threads with a yellow colour. When these samples represented silk core in metallic threads, the objects were all dated to the 18th century, more specifically, between

1751 and 1791. While the earliest of these objects, *epitaphios* inv. 15835, is from Wallachia, all the others are attributed, for stylistic reasons, to Viennese workshops.

4. Discussion and Chronology

Carminic acid-based dyes represent the most frequent source of red. They were identified in the warp, which gives the colour, in the red satin, in objects dated from 1556 to the end of the 18th century. Satin was used as a support for embroidery or painting, and as a lining. Carminic acid was also detected in velvet supports in 19th century *epitaphioi* and in a red velvet lining in an embroidery dated 1791. Carminic acid-based dyes were also the source of red in embroidery threads in *epitaphioi* which are dated 1437, and from the 18th and 19th centuries. In just two cases, both from velvets, one used as a support in a 19th century *epitaphios* and the other the lining of an embroidery dated 1791, was it possible to establish that Mexican cochineal was used as a dye source (see Section 3.1. Supports, Red and Section 3.2. Lining, Red). In the case of the latter, the identification of Mexican cochineal indicates that it is more likely that the velvet is the original lining and not a later addition, as suggested by visual observations.

The identification of expensive insect dyes in the liturgical objects perfectly corresponds to the high value of the objects. The hypotheses stated earlier, about the use of Mexican cochineal in the red satin supports (Section 3.1. Supports, Red) in objects dated after ~1570, could be extended for the velvet, for linings and embroidery threads. The two successful identifications of Mexican cochineal tend to support these hypotheses. Moreover, the Mexican insect was also attested in previous studies on liturgical embroideries in Romanian collections [4,47]. The insect was also recognized in similar liturgical objects worked in the Byzantine tradition, from the same period, conserved in monasteries of the Holy Mount Athos [17]. As Mexican cochineal was a frequently used dye source, available everywhere in Europe, it becomes impossible to draw any conclusions regarding the origin of the materials in the *epitaphioi* studied.

Redwood dyes, known for their poor lightfastness, were detected in the (invisible) warps of the satin used in the *epitaphioi* supports and linings, as well as in velvets. The colour of the threads varies from yellow-pink to red, depending on the amount of dehydrobrazilein product present, and a perfect correlation was observed between the colour of the thread and the relative amounts of the two components in the chromatograms. While most of the detections are in the materials used as supports, redwood dyes were also identified, in a dyeing combination with logwood, in a pale violet embroidery thread in the 19th century *epitaphios* worked in the Gobelin needlepoint technique. The presence of logwood explains the violet colour of the fiber. Redwood and logwood dyes were also present in similar liturgical objects from the Holy Mount Athos, but in the cited work, no mention regarding the function of the samples was given [17].

Indigo-based dyes were identified in the blue satin embroidery supports, in silk and cotton linings and in embroidery threads. They were also present in green satin and velvet supports and in embroidery threads, where they were used together with weld and dyer's broom. Either woad or indigo could have been used as dye sources. Indigo-based dyes were also present in liturgical objects from the Holy Mount Athos [17] and were the main source of blue on protein and cellulosic fibers in all the categories of textiles in Europe.

Prussian Blue, a pigment which was introduced in the early 1700s, was identified in a blue cotton lining in a 19th century *epitaphios* attributed as Russian. It was never identified in liturgical textiles from Romanian collections before, nor in similar objects from other regions within the Orthodox space, to the best of our knowledge. However, as already mentioned in Section 3.2. Linings, Blue, it was evidenced in 19th to 20th century traditional textiles from Romanian collections [44].

Weld and dyer's broom are the two flavonoid dye sources identified in the present study. Weld was present in three cases only, in a silk satin lining and in a velvet support—both in *epitaphioi* dated to the 18th century, of Wallachian and Viennese origin, respectively, and in a green embroidery thread in a 19th century *epitaphios* worked in the Gobelin

needlepoint technique. Dyer's broom was the most frequently used source of yellow in the present study. It was identified in the satin and velvet supports, one time each in 17th century *epitaphioi* from Moldavia and Wallachia, and in embroidery threads in a large number of objects, also from the 17th century onwards. It was also present in an embroidery from Wallachia and in several samples in *epitaphioi* of Viennese origin, mostly as an individual dye source but also in combination with indigo-based dyes. The two yellow dye sources were also detected in liturgical embroideries in Romanian collections previously studied, as well as in similar objects preserved in monasteries from the Holy Mount Athos [17]. It should be noted that in the above-mentioned studies, another flavonoid-based source of yellow, young fustic, was also equally detected as a dye source in textiles dated between the 16th and the 20th centuries. Other yellow dye sources were also detected in liturgical textiles from Romanian collections [4].

Tannins may be used as dye sources, either to achieve black in the presence of iron salts [11], or for ochre yellow when combined with alum. They may also be used for silk weighting, a treatment applied to degummed silk in order to increase its weight. In the present study, tannins were used as a dye, in the presence of iron salts, in a brown velvet support and an embroidery thread in the same colour. The first detection was in a Wallachian *epitaphios*, dated 1752, while the latter was in a liturgical textile, dated 1823, of Viennese origin. The use of tannins in liturgical embroideries was also evidenced in previous studies, both for textiles in Romanian collections and from the Holy Mount Athos [2–4,6,17].

Chronology on the use of dyes in liturgical embroideries worked in the Byzantine tradition

The results of the present study, where most of the objects were dated from the 17th to the 19th centuries, are complementary with previous information achieved in the same research group for liturgical embroideries in Romanian collections, mainly dated to the 15th and 16th centuries [2–6]. Table 5 presents an overview of the dyes detected for each of the fibers and their function discussed in the present study: supports (satin and velvet), lining and embroidery threads.

Red is the main colour for supports, and lac dye, madder, Polish carmine and kermes were used in the 15th and 16th centuries, while carminic acid-based dyes (sometimes identified as Mexican cochineal) came from the 16th century onwards. As far as the American insect is concerned, the earliest date of use may be around 1570. It should be noted that the above-mentioned sources are used for the visible warp, while redwood dyes are mainly used for the wefts. Velvets are used as embroidery supports after the 18th century, and red is always achieved with carminic acid insect dyes. Earlier velvets dyed with lac dye and madder (or yellow and blue sources dyer's broom and indigo-based dyes) were also observed in liturgical embroideries but they were not really used as supports, but as frames or base for embroidered inscriptions on satin. Blue is the second most frequently used colour for supports after red, and it is always based on indigo dyes. Yellow and brown satin supports are less common and they are dyed with dyer's broom, safflower yellow (occasionally) and, rarely, tannins. Redwood velvet supports were also observed from the 18th century onwards (Table 5a).

The earliest surviving linings date from the 16th century, and are mainly in blue, silk or cotton, dyed with indigo. Red, yellow and green linings, dated from the 17th century onwards, were also preserved and the same dye sources discussed for the supports were observed. Weld was also detected in yellow 18th century satin linings, while a Prussian blue cotton one was noticed in a 19th century embroidery (Table 5b).

A larger palette of colour sources, as compared with supports and linings, was observed for the embroidery threads. Madder, lac dye, redwood, carminic acid-based (*Porhyrophora* species) and safflower were used for red in the 15th century, while carminic acid-based dye (probably Mexican cochineal) remains the only choice from the 17th century onwards. Several yellow dye sources, which were not detected in the present study, were revealed with different frequencies for yellow and green embroidery threads: young fustic,

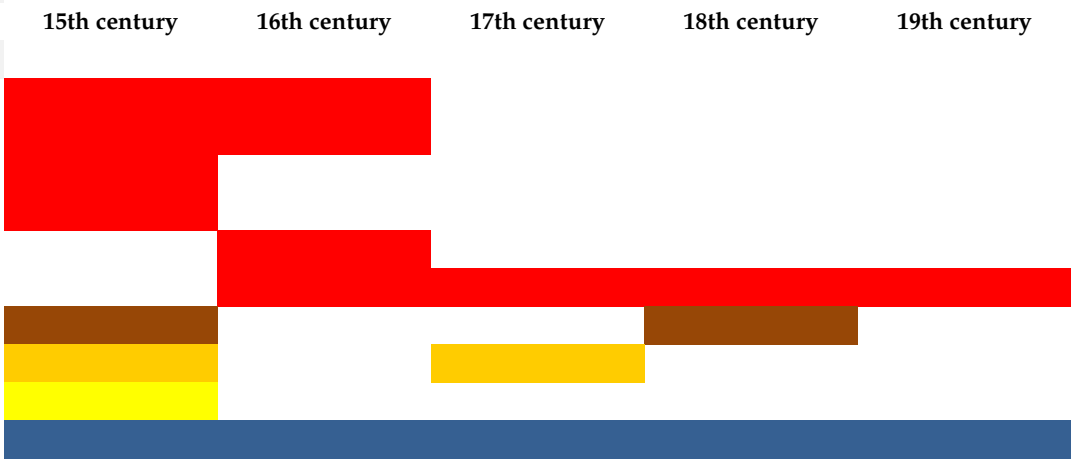
bastard hemp, emodin-based dyes and berries. Indigo-based dyes were used for blue and green embroidery threads (Table 5c).

Table 5. Chronology on the use of dyes in liturgical embroideries worked in Byzantine tradition: (**a**) supports (satin and velvet); (**b**) lining; (**a**,**c**) embroidery threads.

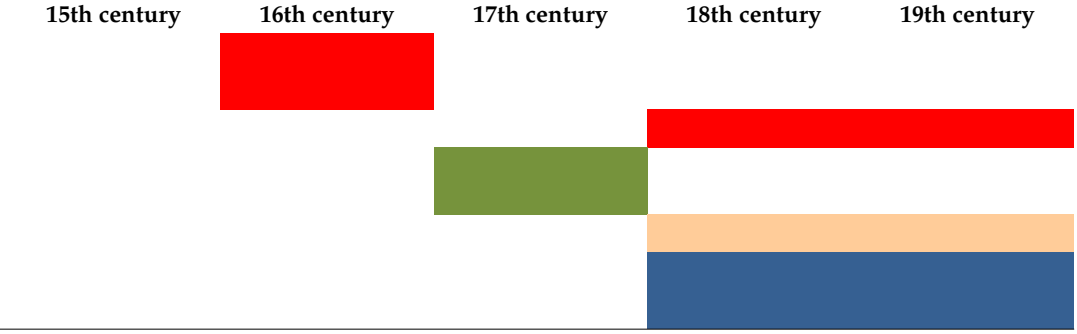
a.

SUPPORTS

SATIN



VELVET



b.

LINING

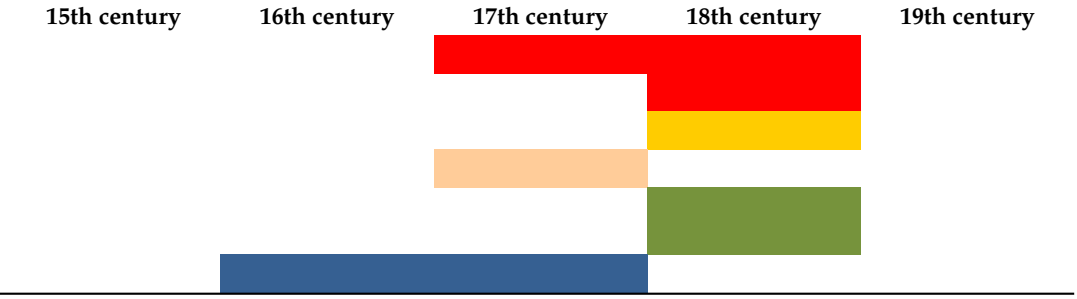
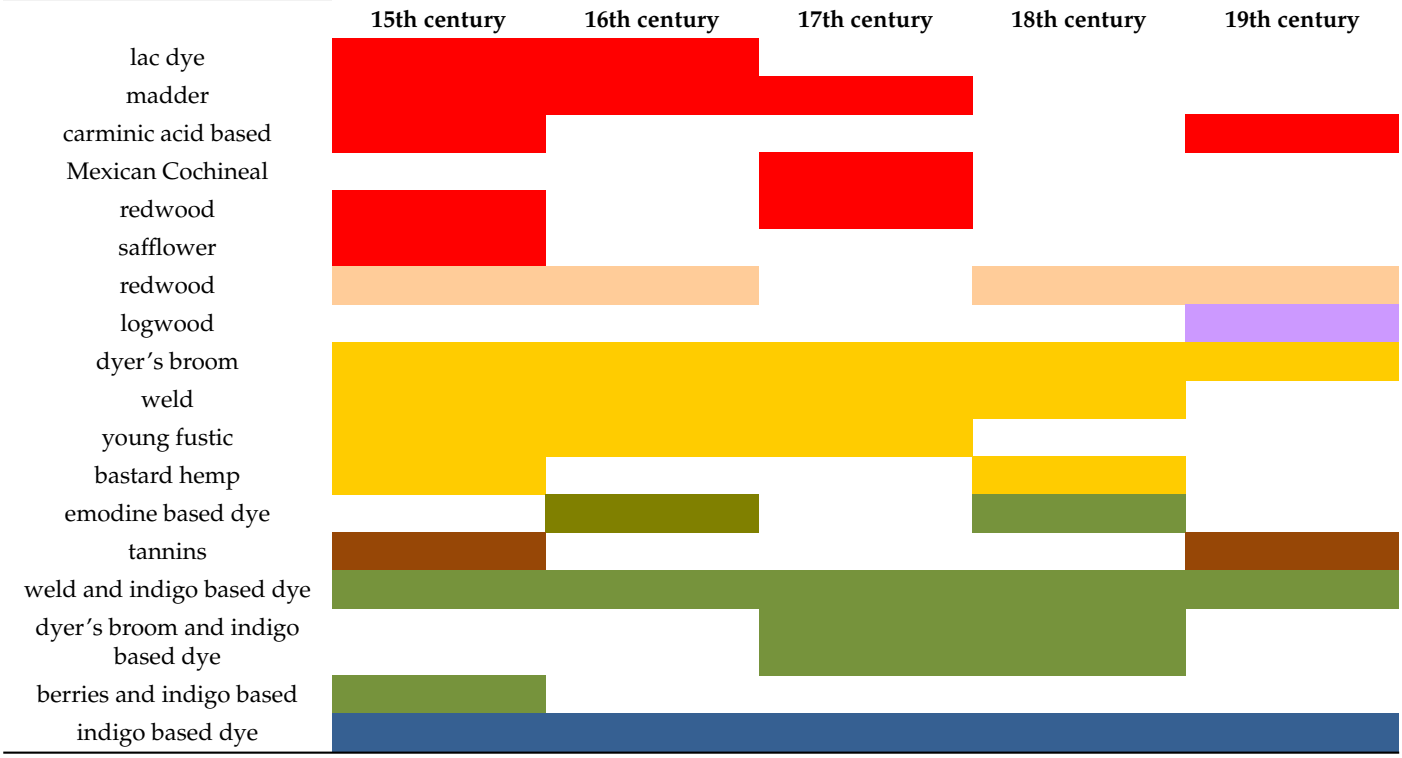


Table 5. *Cont.*

c.

EMBROIDERY THREADS



5. Conclusions

A large number of objects with similar liturgical function, dated over a period of 500 years, were studied in terms of dye analysis, for the first time with reference to Romanian collections. The objects are attributed to Byzantine, Moldavian, Wallachian, Greek, Russian, Constantinople and Viennese workshops.

Most of the samples are from the supports and, of these, the most common were red satin, with a warp dyed with a carminic acid containing dye and a soluble redwood weft. The material was also used as lining. The above-mentioned sources, as well as the others identified in the supports, lining and as embroidery threads were discussed together with those identified in previous studies, and chronologies on the use of dyes were proposed for each of the three functions of the fibres.

The biological sources identified in the few samples accessible for each object do not differ with the geographical areas of provenance of the objects studied. Consequently, it became impossible to establish any criteria to be used for attribution purposes. However, it was observed that Moldavian supports are always red, redwood-type dyed velvet was common for the supports in *epitaphioi* of Viennese origin and all the embroidery threads from Viennese liturgical textiles studied are dyed with dyer's broom.

The results obtained are extremely promising and valuable information was acquired, despite the limited number of samples available for each object. With reference to the whole Orthodox jurisdiction, liturgical embroideries of Byzantine tradition are little studied, and even less with reference to the materials and techniques used, which includes the dye sources. In-depth studies of liturgical embroideries in Romanian collections and beyond, correlated with conservation and a well-defined sampling strategy, would be worthwhile in order to better comprehend and appreciate these extremely valuable testimonies of world heritage.

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