ANALIZĂ A UNEI PICTURI REALIZATĂ PE SUPORT METALIC PENTRU A STABILI APARTENENŢA
ANALYSIS OF A PAINTING MADE ON METAL SUPPORT IN ORDER TO ESTABLISH PATERNITY

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The work analyzes an artwork painted on a metal plate as a substrate in order to establish its authenticity. In the lower-right side is to be noticed a signature alongside a date (''Lecca 1866''), whilst on the reverse, in the upper-central part, there is another signature under which there is an inscription in Greek language (''Ο ΑΓΙΟΣ ΑΝΤΩΝΙΟΣ (phonetictranscription AGIOS ANTONIOS) 1866''. The signatures, dates and the inscription on the front and reverse sides of the paintings were added long after the original painting was carried out. The outcome is showing the work was repainted almost entirely on tin-plated iron sheet. The work includes a brief study on the work and activity of painter C. Lecca. The history of the use of metal supports in easel painting is also investigated. A complex metallographic analysis was performed on the metal support (i.e. optical microscopy and SEM-EDX). X-ray fluorescence spectrometry (XRF) analyzes were carried on the paint layer.

Lucrarea analizează o operă de artă pictată pe o placă de metal ca substrat în vederea stabilirii autenticităţii sale. În partea inferior-dreapta se remarcă o semnătură şi o datare (''Lecca 1866''), în timp ce pe verso, în partea superior-centrală apare o altă semnătură sub care se află o inscripție în limba greacă, datată ''Ο ΑΓΙΟΣ ΑΝΤΩΝΙΟΣ (transcriere fonetică AGIOS ANTONIOS) 1866''. Semnăturile, datările şi inscripția de pe fața şi verso-ul tabloului au fost adăugate la mult timp după realizarea picturii originale. S-a constatat ca lucrarea a fost repictată aproape în întregime pe o tablă de fier acoperită cu staniu. Lucrarea cuprinde un succint studiu referitor la opera şi activitatea pictorului C. Lecca. Este investigat şi istoricul utilizării suporturilor din metal în pictura de şevalet. Asupra suportului metalic s-a efectuat o analiză metalografică complexă (microscopie optică şi SEM-EDX). Analize de spectrometrie de fluorescență de raze X (XRF) au fost efectuate asupra stratului pictural.

Keywords:*metal support, iron sheet, SEM – EDX, XRF*

1. Introduction

Although in Western Europe metal supports for painting were common in the mid-sixteenth century, on the territory of our country (i.e. Romania), it began to be used since the period of the painter Constantin Lecca (1807-1887). At a certain moment he was considered an innovator in Romanian easel painting due to the unconventional painting materials used at that time: metal support (e.g. iron, copper sheet), oilcloth stuck on a stiff support material such as wood and ivory, etc.

Constantin Lecca painted a large number of icons [1, 2], both for Constantin Brâncoveanu's Mansion (the Prince of Vallachia during 1688-1714) and for ordinary people dwellings; he also worked on the restoration of a large number of icons [1], and in the case of large projects he collaborated with Misu Popp and Barbu Stanescu [3]. Religious paintings, sketches, lithographs, church paintings, historical compositions and portraits painted by Lecca show a multitude of clumsiness (reminiscent of the works of primitive painters), caused by ignoring the quality of drawing and plasticity / chromatics [1, 4]. However, Lecca's paintings had a pronounced influence in the transition from Byzantine religious painting to secular, modern painting dominated by portraiture, the painting appreciated in those times in Western Europe [1, 4 and 5].

Constantin Lecca was born in 1807 in a family of merchants from Brasov; he learned the basics of painting probably in Brașov [1], after which he studied painting at Buda (i.e. city of Budapest nowadays) between 1827-1833, with breaks of several months [6], then perfecting his knowledge in Vienna [5], Rome, Venice, Naples, Florence (where he painted reproductions / copies of second-hand paintings [2] from galleries and museums [6]), Berlin [1] and Paris [6]. The painter lived in the city of Craiova between 1833 and 1848 [6], where he worked as a teacher of drawing and calligraphy at the Central School, having Theodor Aman [4] as a student, then moved to Brasov, afterwards to Bucharest, teaching drawing and calligraphy to Saint Sava National College [1]. He retired from the artistic life in 1870, already suffering from Parkinson's disease and in 1887 he passed away [6].

It is also known that the artist Constantin Lecca introduced the metal support in the Romanian easel painting from the nineteenth century. He painted on the blackboard, and as proof, at the Church museum of Saint Nicholas

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in Brasov's neighborhood named Saddle (i.e. ȘcheiiBrașovului), three paintings with religious themes were discovered relatively recently (''The Last Supper'', ''Entering Jerusalem'' and ''Washing the feet'') painted by him. The paintings with religious themes were painted on iron sheet in oil colors, in the period 1846-1847, have an ellipsoidal shape and dimensions between 186 x 0.86 cm and 172 x 0.84 cm [6, 7]. The archives studied by Mihai Manolache in 2016 prove that some of the icons of the chapel and the women pavilion from the Church of Saint Nicholas in ȘcheiiBrașovului neighborhood were painted by Constantin Lecca between the years 1846 - 1847 [7]. One of the documents studied by Mihai Manolache was the Contract agreed by Constantin Lecca with the representatives of the Church of Saint Nicholas from ȘcheiiBrașovului on 22 September 1846, pledging for the execution of icons on various painting supports (e.g. iron sheet [8], copper plate, wood, oilcloth [8]), as well as the restoration of royal icons, respectively the repair of four large icons with silver [6, 7].

The work analyzes from a metallographic and microstructural point of view a possible work made by the painter Constantin Lecca having as religious theme the image of St. Anthony. Starting from the fact that on the front and reverse sides of the painting performed on metal support are signatures, dates, as well as an inscription, special attention was paid to both the methods of identifying the components of the painting support material and the painting layer, as well on studying the specialized painting literature.

2. Materials and Methods

For identifying the support material and the pigments from which the painting with religious theme was made, a complex metallographic analysis has been performed (i.e. optical and electron microscopy), X-ray fluorescence spectrometry (XRF) and ultraviolet reflectography.

XRF and optical metallographic analyzes were carried out at the Laboratory of Hydrometallurgy, Department of Engineering and Management of Obtaining Metallic Materials, Faculty of Materials Science and Engineering, University Politehnica of Bucharest - where there are interests in the field of art research since 2018 [9-11]. SEM-EDX analyzes were conducted at the National Research Center for Micro and Nanomaterials, Department of Science and Engineering of Oxide Materials and Nanomaterials, Faculty of Applied Chemistry and Materials Science, University Politehnica of Bucharest.

2.1. Ultraviolet reflectography

Ultraviolet reflectography consists in irradiating the examined object with UV radiation [12]. In this case the work was examined with fluorescent lamp with low pressure mercury vapor (5.0 mg) -TL-D 36W BLB 1SL / 25, with main application Blacklight Blue and the base of the fixing element G13 - which has a "inner capsule covered with a fluorescent powder that emits long-wavelength UV radiation to excite luminescence " [13].

2.2.Metallographic analysis of the support

Metallographic analyzes were performed using an optical microscope Reichert Univar. For highlighting the structure of the metallic support material of the painting, a sample of 1.5 mm^2 was taken from the left corner of the painting work.

The preparation of the sample was made by sanding and polishing on the Buehler Alpha type machine with vector head, as follows:

- Grinding was performed in four stages, using abrasive papers with granulations of 75, 45, 25, 15μm, having the following parameters: pressing force of 4N / mm^2; turntable speed of 300 rpm; grinding time of 3min / stage, water;
- Polishing was performed on textile support "Buehler-Microflok" in five stages, using abrasive material, diamond suspensions with granules of 9, 6, 3, 1μm, with the parameters: pressing force of 4N / mm^2; turntable speed of 200 rpm; grinding time of 3 min / stage;
- The last final polishing stage was performed on a "Buehler-Vibromet" vibration polishing device, using as abrasive aqueous colloidal silica suspension with a granulation of 0.02μm, for 6 hours.
- After the final polishing the sample was attacked with Nital 2% by immersion for 15 seconds.

2.3. Electron microscopy analysis (SEM)

The sample were analyzed by scanning electron microscopy using a Quanta Inspect F50, with a field emission gun (FEG) with 1.2 nm resolution and an Energy Dispersive X-ray Spectrometer (EDXS) having 133 eV resolutions at MnKα.

2.4. X - Ray fluorescence spectrometry (XRF)of composite artifacts

X-ray fluorescence spectrometry (XRF) was performed with the Oxford X-MET 5000 Handheld XRF Analyzer. Method's parameters: RX tube current 10 μA, high tube voltage 40 kV, XRF SiPIN detector, out time 10u s, analysis time 15 s.

Type of calibration used: 1-EC (Empirical Calibration) - Calibration with standards, where the matrix allowed; 2-FP (Fundamental Parameter) - General calibration without standards.

The spectrometer was previously checked

Fig. 1 - XRF analysis points on the front (a) and on the back (b) of the painting / *Puncte de analiză XRF pe partea din față (a) și pe partea din spate (b) a picturii.*

Fig. 2 - Overview image in direct/normal light (left) and in UV light (right) / *Imagine generală în lumină directă / normală (stânga) și în lumina UV (dreapta).*

at standards GBW 07236 and 316, Skyray instrument co., Ltd, TRH-801 for Pb in Painting and Fe, Cr, Ni, Cu in metal alloys respectively.

For this analysis, from which the elemental compositions were determined to find out the types of pigments used, this marked in the images in Figure 1. Of the 16 points of analysis13 were performed on the front of the artwork in areas that seem authentic and have their own network of cracks (1-11, 13, 14), while point of analysis 12 is in an area with repainting visible and without network of cracks (point 12-Table 1). Analysis points 15 and 16 were performed on the back of the artwork

3.Results and Discussions

3.1. Direct light and UV

Usually, the primary investigation of artworks consists in detailing their structure or morphology using various visualization or imaging techniques. The first examination of an object isperformed using the light reflected by it.

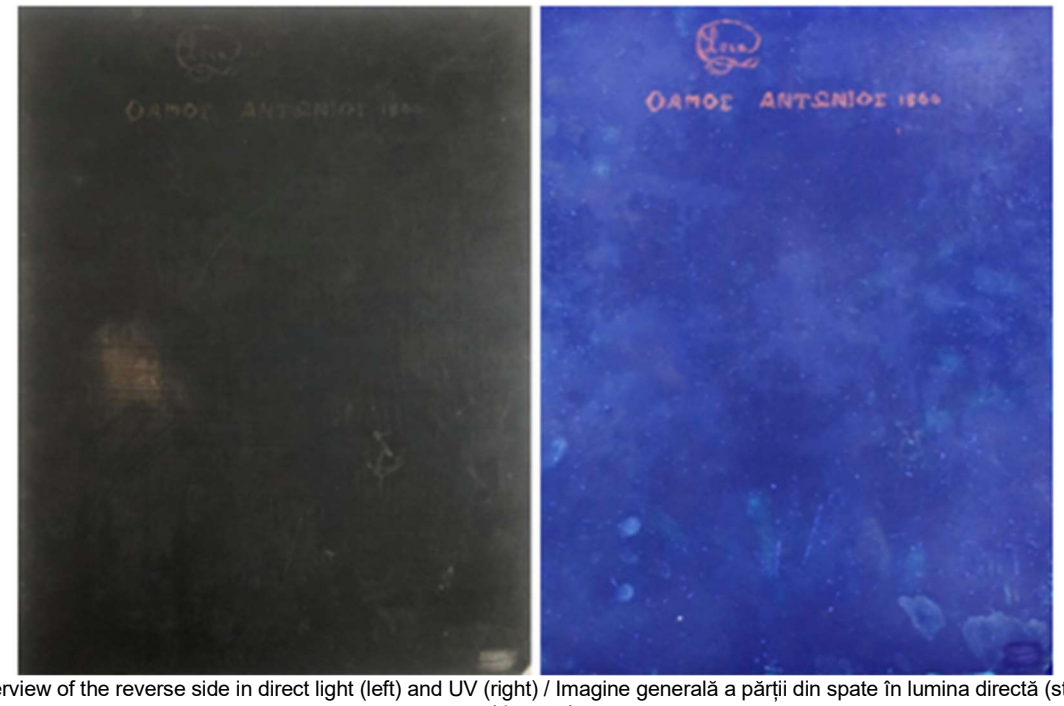


Fig. 3 - Overview of the reverse side in direct light (left) and UV (right) / Imagine generală a părții din spate în lumina directă (stânga) și UV (dreapta).

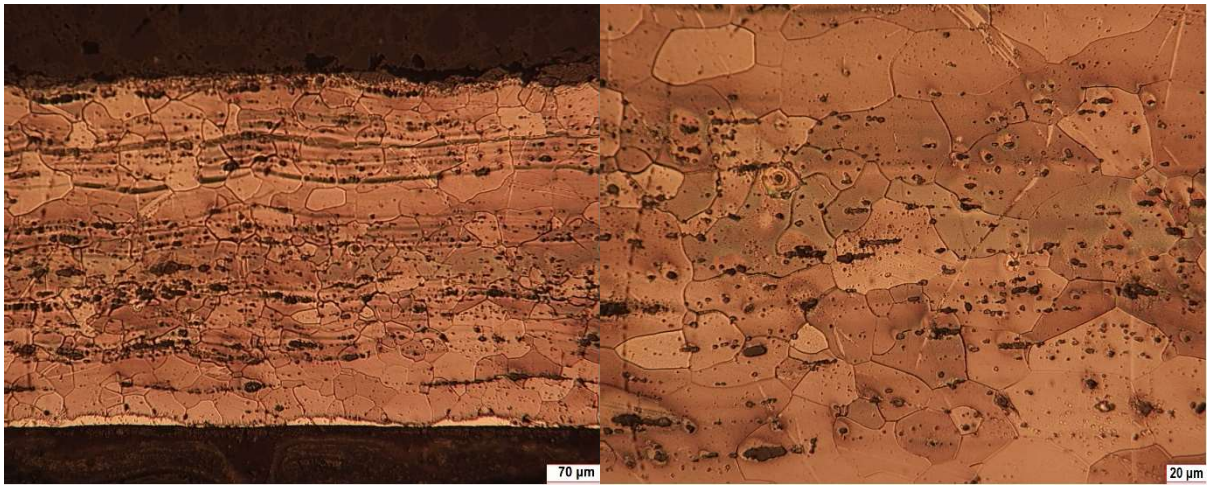


Fig. 4 - Metallographic structure of the painting support material highlighted by attack with the chemical reagent Nital 2% / *Structura metalografică a materialului suport al picturii evidențiată prin atacul cu reactivul chimic Nital 2%.*

Traditionally, visual observation uses light from the visible spectrum, but there is also the possibility of examining artworks using ultraviolet radiation to allow the observation of details that do not appear in the light of the visible spectrum [14].Figures2 and 3 show the images of the painting analyzed both in the visible spectrum and using ultraviolet light.

The signature and dating ‘’ Lecca 1866 ‘’ from the lower-right part of the painting, respectively the signature, inscription and dating ‘’Leca / Ο ΑΓΙΟΣ ΑΝΤΩΝΙΟΣ 1866 ‘’ from the upper-central part of the reverse, were added long after the finalization of the original painting, which is partially visible, especially on the open areas.

3.2. Metallographic analysis of the support

Following the metallographic analysis, it was found that the metal plate of artwork work is made of steel (iron and carbon) sheet. Iron was identified following XRF analysis (Table 1 XRF analyses of the 16 analyzed points -points 15 and 16).Figure 4 shows the optical microstructures of the metal material. The metal microstructure, observed by optical microscope, revealed that the steel has low carbon content.

The thickness of the layer deposited on the support material was also measured. Figure 5 shows the layer deposited on the support and the results of the measurements performed, the deposited layer having thicknesses between 7.60 and 16.70 µm. The samplehas a deposited, irregularlylayerthickness of approx. 12µm.

The layer whose thickness was assessed by light microscopy analysis was identified by EDX as tin.

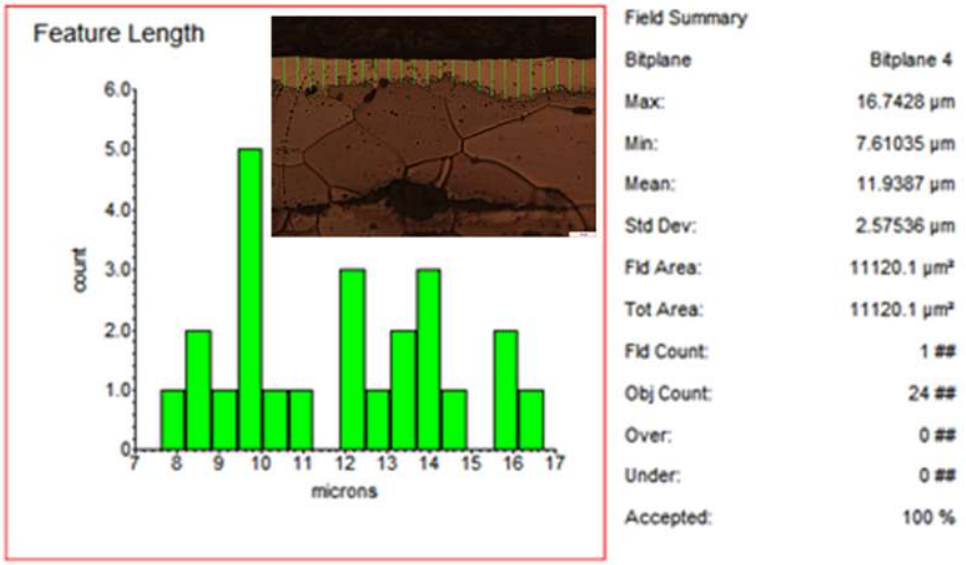


Fig. 5 - Layer thickness measurements (support material steel), calibration: 0.07610 µm / Pixel.
Măsurători ale grosimii stratului (material support - otel, calibrare: 0.07610 µm / Pixel.

3.3. Scanning Electron Microscopy analysis (SEM-EDX)

The electron microscopy analyzes were performed on the sample taken from the metal support on which the religious scene was painted. The results of SEM-EDX investigations are presented in Figures 6 to 10.Figure 6 shows the detailed aspect of the analyzed sample section, at different resolutions.

Figure 7 shows the area where the EDX analyzes were performed for the painting support material and for the layer deposited on it.

The SEM-EDX analysis of the samples taken from the metal supports showed that the support consisting of a metallic core of steel (iron and carbon), coated with a thin layer of tin on both sides (Fig. 7) The steel sheets were rolled and then immersed in a tank containing liquid tin to acquire the metal coating. The tin coating is very irregular and presents several fissures.

Some impurities were detected by SEM-EDX in the metallic core, such as silicon (Fig.9), which are elongated due to the rolling of steel (Fig. 4).

The analysis spectra for each analyzed area are shown in Figures 8 and 9. According to analysis points EDX1 and EDX2 the support is made of iron sheet, coated with tin against corrosion.

Sample investigation by SEM-EDX with the overall distribution of the elements per area analyzed is shown in Figure 10.

According to the SEM-EDX analysis for the selected micro-zone, the distribution of each element is observed, with Fe in high concentration, and Sn, Si, C and O.

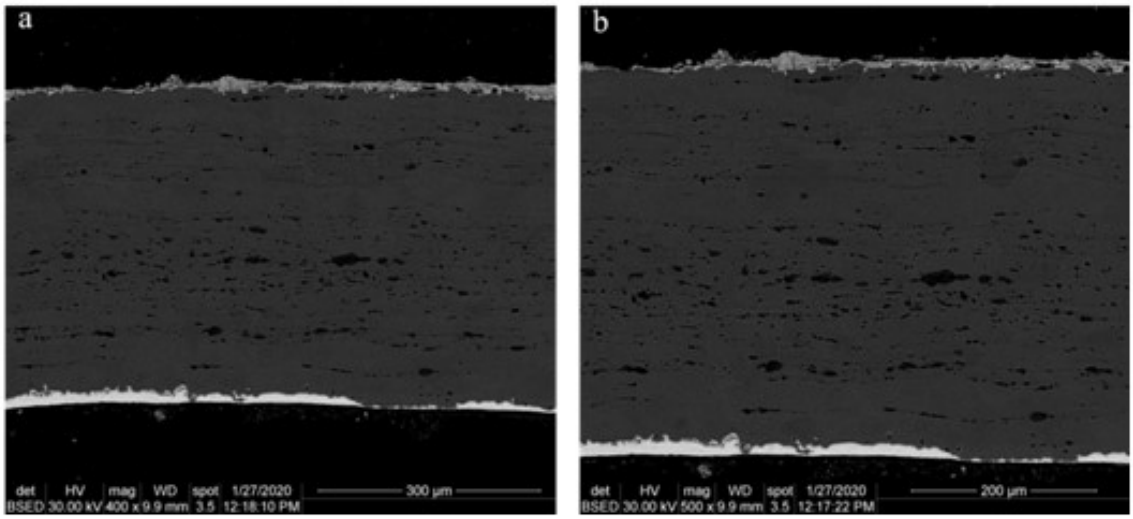


Fig. 6 - SEM images of the sample; a) resolution x400 and b) resolution x500 /
Imagini SEM ale probei; a) rezoluția x400 și b) rezoluția x500.

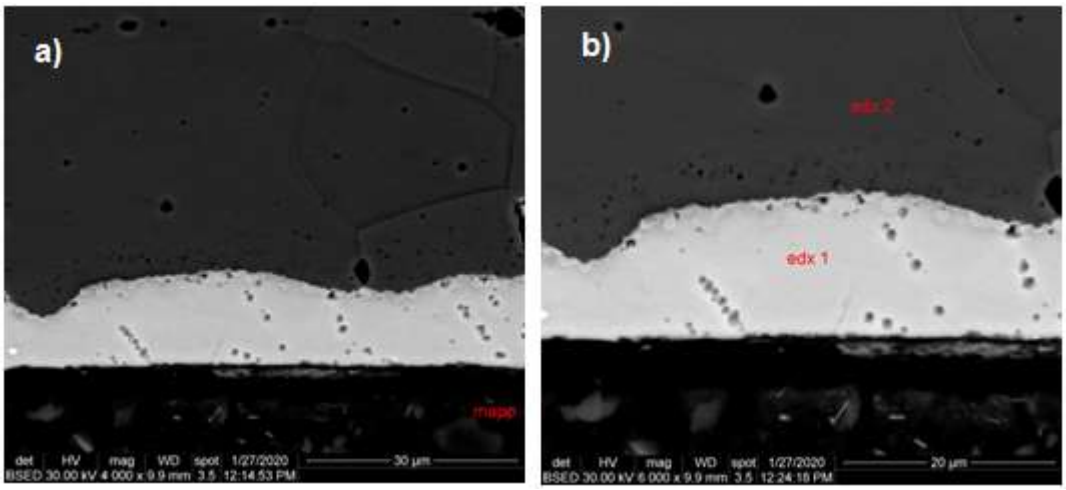


Fig. 7 - SEM image of the sample, resolution x 4.000 (a) and the EDX analysis points, resolution x 6.000 (b) / *Imaginea SEM a probei, rezoluția x 4.000 (a) și punctele de analiză EDX, rezoluția x 6.000 (b).*

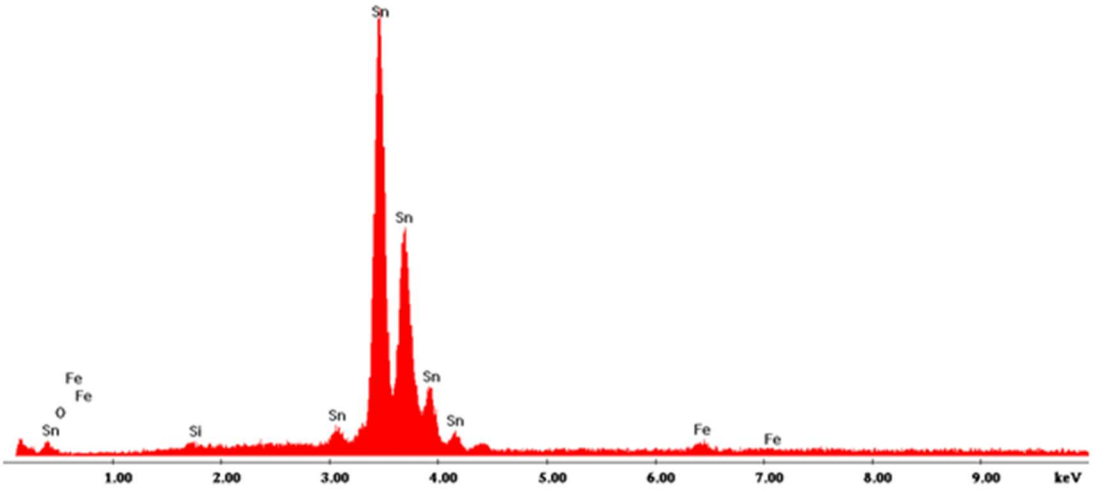


Fig. 8 - EDX1 – the main element is tin / *EDX1 - elementul principal estestaniul.*

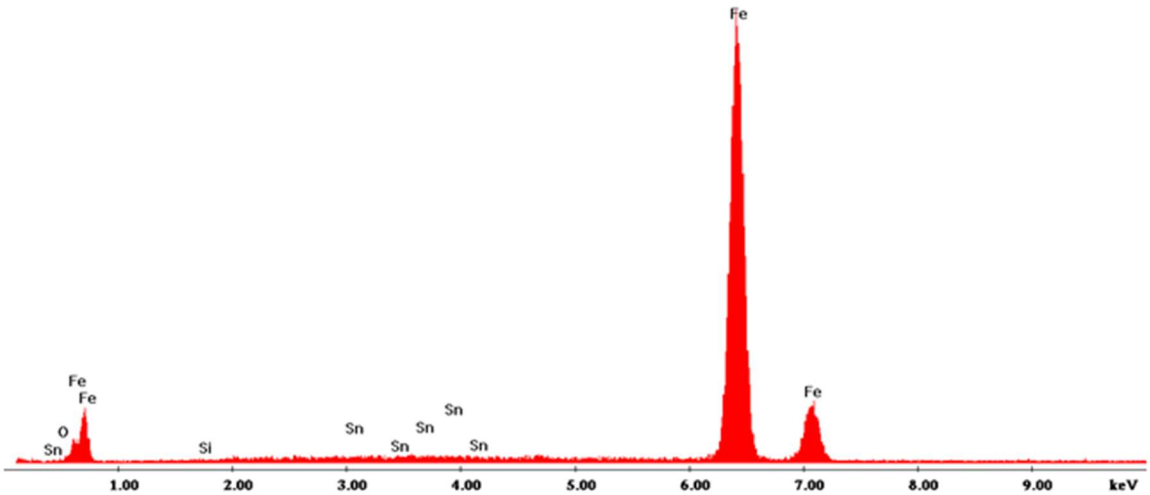


Fig. 9 - EDX2 – the main element is iron / EDX2 - elementul principaleste fierul.

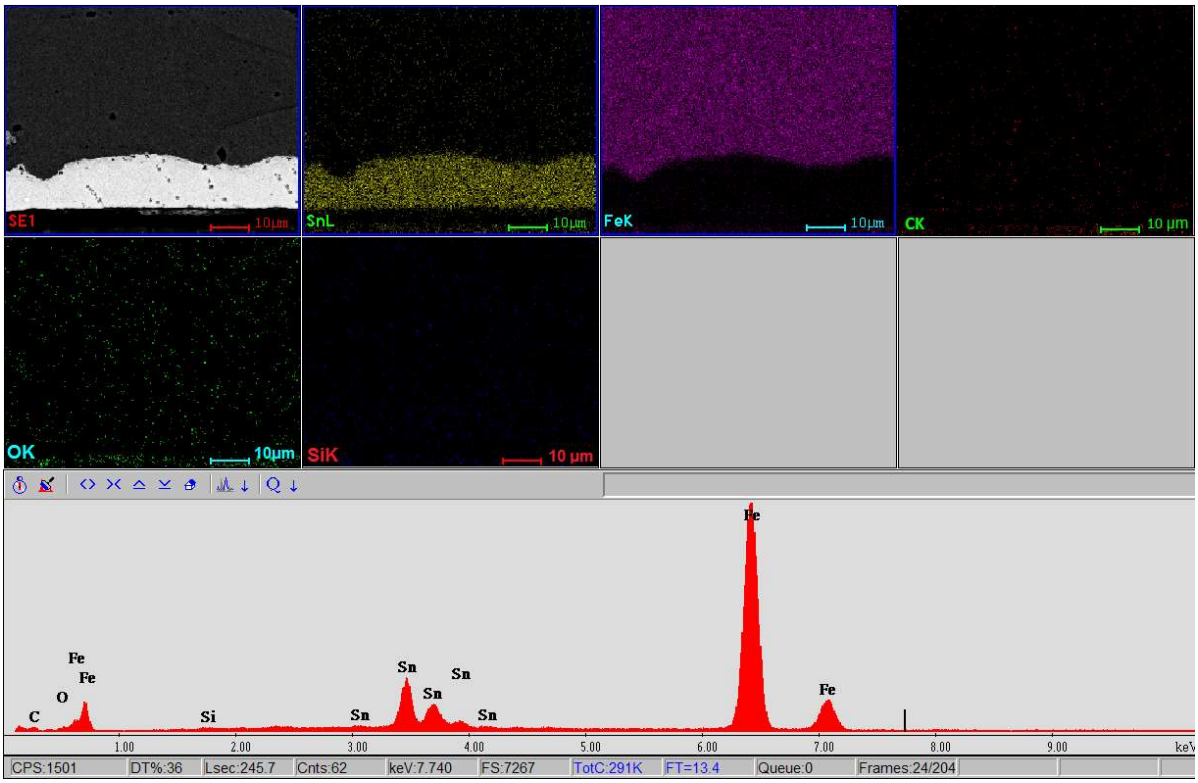


Fig. 10 - Overall distribution of the elements per area analyzed / *Distribuția generală a elementelor pe suprafața analizată.*

3.4. X - Ray fluorescence spectrometry (XRF)

X-ray fluorescence spectroscopy (XRF) is used both to identify the technology of fabrication of metal objects and to determine the inorganic pigments in the color film because many inorganic pigments can be characterized by the presence of a single detectable element or several [14].

The XRF analysis in order to determine the elemental compositions was performed on 16 points from which the chemical compositions were determined and are shown in Table 1.

3.4.1. Paint layers

Analysis points 1-14 were made on the front of the painting in order to identify / determine the inorganic pigments used by the artist. It is known that they were often used by painters due to their high coverage and stability to microclimate variations [15]. In general, for the analysis points, areas without repainting were chosen, except for the analysis point no. 12.

Although the identification of inorganic pigments is done efficiently based on the characteristic elements (specific color and constituent chemical elements), the data provided by XRF analysis can sometimes be nonspecific for several reasons: (a) the combination of several pigments to obtain a certain color ; (b) the chemical elements in the stratigraphy of the artwork that will influence the XRF spectrum, such as the application of lead white as a base layer; (c) majority elements found in completely different pigments that may hinder their correct determination / interpretation, as is the case with yellow-brown colors containing Fe [15].

Interpreting the XRF results is hampered due to the fact the artwork is painted on metal support, respectively tin-plated iron plate, meaning the metal support contains two major elements - iron and tin - which can also be found in certain pigments based on iron (sallow) and / or tin.

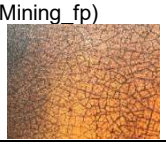
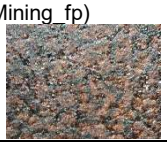
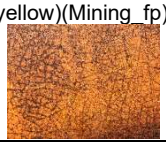
When choosing the exclusion of the main elements contained by the painting support - iron and tin - it can be seen that at the level of the paint layer two major components can be identified – lead (Pb) and calcium (Ca) - which can indicate the existence of white lead pigment, respectively bone black (charcoal) and/or asphalt.Lead has been identified in large quantities on all points analyzed at the level of the paint layer (even in the area of dark colors), reason for which it is possible that a very thin layer of lead white has been applied to the tin-plated sheet metal support a prióri the execution of the painting.

The limited chromaticity of the painting indicates a possible compositionof only sallow (e.g. iron ocher, iron brown, natural shade); carbon black and lead white (a higher amount of lead was identified in the lighter areas than on the darker surfaces).

Lead white [16] which is known as cerussa (in Latin), silver white, biacca, Krems' white or Kremnitz's white, is the most important lead pigment, chemically known as lead base carbonate ($PbCO_3$). This is one of the pigments used in antiquity, and

Table 1

XRF analyses of the 16 analyzed points / *Analizele XRF ale celor 16 puncte.*

XRF / areaanalysispoints / photodetailsfromtheanalysispointsarea	Key element(s) (%)				Name of possible pigment(s)
	Fe	Pb	Sn	Ca	
1.Front-side support(Mining_fp) 	24.0	5.3	4.5	4.3	Carbon black and/or Asphalt Lead white Sallow
2. Neck(Mining_fp) 	0.9	14.8	7.7	-	Lead white Yellow ochre
3.Forefront right(Mining_fp) 	0.3	31.1	6.3	-	Lead white Yellow ochre
4.Book down left (Mining_fp) 	1.2	10.7	3.7	-	Lead white Yellow ochre
5.Cuff (Mining_fp) 	2.2	10.9	4.0	-	Lead white Yellow ochre
6.Right down (black) (Mining_fp) 	17.5	9.9	6.5	3.6	Carbon black and/or Asphalt Lead white
7.Right background (sienna) (Mining_fp) 	1.3	10.1	6.4	0.8	Led yellow Carbon blackand/or Asphalt Lead white
8.Ear(Mining_fp) 	3.3	9.7	10.7	1.8	Lead white Yellow ochre and/or Led yellow
9.Hand(yellow)(Mining_fp) 	0.6	12.0	4.3	0.7	Yellow ochre and/or Led yellow Lead white
10.Cross (base)(Mining_fp) 	8.7	15.4	10.7	2.6	Yellow ochre and/or Led yellow Lead white
11.Shoulder(Mining_fp) 	7.1	14.4	8.1	1.0	Led yellow Carbon blackand/or Asphalt Lead white
12.Hair (repaint)(Mining_fp) 	5.3	9.8	8.6	8.7	Yellow ochre and/or Led yellow Lead white

13.Arm (cracks)(Mining_fp) 	15.4	5.4	9.3	4.3	Carbon black and/or Asphalt Lead white
14.Face downMining_fp 	273	16.0	5.2	2.8	Lead white Yellow ochre Carbon black
15. Suport – reverse(Alloy Mode) 	95.98	-	-	-	-
16.Suport - reverse(Alloy Mode) 	97.59	-	-	-	-

among the ancient writers who described the preparation of lead white from lead sheets and vinegar we can list Pliny, Vitruvius and C. Cennini. Bone charcoal (animal black) [16] is a pigment obtained from charcoal derived from incomplete bones burning in sealed containers.

3.4.2. The support material

The identification of the support material of the painting was made both on the basis of complex metallographic analysis (i.e. optical and electron microscopy) and by X-ray fluorescence spectrometry.

The metallographic analysis finds that the religious-themed artwork was painted on tin-plated iron sheet, and X-ray fluorescence spectrometry performed in both Alloy Mode and Mining_fp mode, highlighted iron as the main element. The values of the main element found - iron - identified in two points in Alloy Mode are between 95.98 and 97.59.

The technique of easel painting on metal support (especially on iron and copper sheet) has its origins in Europe [17]. Metal plates, especially those made of copper, began to be widely used by Dutchand German painters from the second half of the sixteenth century to the mid-seventeenth century, mostly for oil painting [18]. At the end of the eighteenth century, the tin-plated iron sheet material support was often used for mass religious painting, especially in the Spanish Colonies [19]. Metal surfaces were often protected against corrosion by coating with a thin layer of another metal [17]. In this particular case, the support is made of iron plate covered with tin. It cannot be excluded the artwork reached Romania at some point, and its precarious preservation state required all the interventions (repainting) in order to be capitalized / sold.

3.5. Signatures, dates and inscription

Due to the fact that he rarely signed and dated his artwork, it is difficult to determine the paternity / succession of Constantin Lecca's paintings [6]. In this particular case, the signature and the date “Lecca 1866” from the front lower-right part of the painting, respectively the signature, the inscription and the date “Leca / Ο ΑΓΙΟΣ ΑΝΤΩΝΙΟΣ 1866” from the upper-central part of the reverse, are not authentic; it do not have their own crackle networks, but have been added over areas with repainting, cracks or gaps, as shown in Figure 11 and Figure 12.

Fig. 11 - Inscription details on the painting back / *Detalii de înscriere pe spatele picturii.*

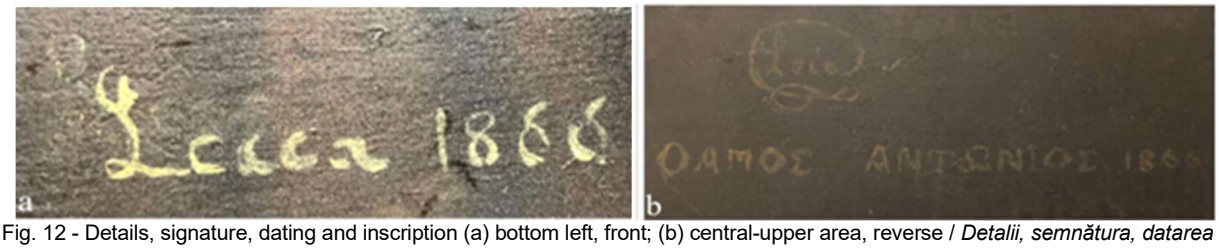


Fig. 12 - Details, signature, dating and inscription (a) bottom left, front; (b) central-upper area, reverse / *Detalii, semnătura, datarea și inscripția (a) partea stângă jos, față; (b) zona centrală-superioară, verso.*

During the period 1851-1870, Constantin Lecca painted in series [1], its artwork having religious themes being characterized by a naive way of execution, revealing a myriad of clumsiness, sometimes intertwined with valuable artistic elements [1], many of them making part of unknown art collections or being attributed to anonymous painters [6]. The precarious preservation state of the painting has been hiding by massive repaintings, and the details from the original paint layer are difficult to observe, especially on the lighter colored areas. The person who added the signature and the date "Lecca 1866" at the bottom right of the painting knew that after 1845 (after the period of its studies in Italy), ConstantinLecca signed his artworks with double "c" (i.e.‘’Lecca’’), and as for the signature on the reverse, it was probably inspired by the type of signature applied by the painter only on lithographs - more calligraphic, encircled and with several loops at the bottom, respectively with a single ‘’c’’- (‘’Leca’’). The inscription in Greek language - ‘’Ο ΑΓΙΟΣ ΑΝΤΩΝΙΟΣ (phonetic transcription AGIOS ANTONIOS)’’- specifies the character depicted in the painting, namely Saint Anthony.

4. Conclusions

The identification of the painting’s support material was based on a complex metallographic analysis (i.e. optical and electron microscopy), its outcome revealing that the religious theme artwork was painted on tin-plated iron sheet;

The interpretation of XRF analyzes applied to the paint layer was hampered by the fact that the artwork is painted on tin-plated iron sheet - two major elements: iron and tin - which can be found in many iron-based pigments (sallow) and / or tin. However, it is possible that the limited color palette used by the painter was made up of sallow, lead white and charcoal black.

The signature and the date on the front lower-right side, respectively the inscription and the date ‘’Leca / Ο ΑΓΙΟΣ ΑΝΤΩΝΙΟΣ 1866’’ from the upper-central part of the reverse, are not authentic. These were added long after the original painting was finalized (which is partially visible, especially on the lighter colored areas); it has been added over repainted areas, cracks or gaps, and do not have their own cracks.

It is difficult to determine the paternity of Constantin Lecca's paintings due to the fact that he rarely signed and dated his artworks, however it is known that he painted icons on various support materials, including iron sheet and that many of these paintings are part of unknown collections or attributed to anonymous painters.

The additional information obtained through XRF investigations supports the results of the metallographic analysis according to which the metal support is a tin plate coated with iron as the key elements Fe and Sn are found in all XRF analysis points performed. The role of tin is to protect the iron sheet against corrosion.

Regarding the identification of the pigments in the coat of paint based on the majority of chemical elements, it can be argued that the only element found in all XRF analysis points on the front of the painting and which is not part of the chemical structure of the support is lead. This can be explained by the fact that it is possible that on the front surface of the tin-plated iron sheet a preparative layer was applied - white lead - on which the drawing was made and which acts as a barrier between the metal support and the coat of paint.

The pigments indicated as probably part of the chemical structure of the coat of paint (Table 1 in the article) are not sufficiently relevant in this case, all the more so as the strong X-ray lines of the lead white in the preparation layer can cover the lines of weak X-rays of the pigments in the upper layers.

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