

ARCHAOMETRIC STUDIES ON EGYPTIAN OBJECTS FROM THE “MARIA AND DR. GEORGE SEVEREANU” COLLECTION

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Cuvinte cheie: *amuletă, scarabeu, colier, mumie, Egipt dinastic, artefacte egiptene, analize arheometrice, colecția Severeanu.*

Abstract: *This study is a first attempt to investigate some Egyptian objects from the “Maria and Dr. George Severeanu” Collection within Bucharest Municipality Museum. The key piece, the one that triggered the present study, is represented by a mummy’s hand (palm) with bandage remnants and a fragment of an amulet firmly attached. The opinions of some specialists from Romania and the United Kingdom led to slightly divergent conclusions, which triggered the archaeometric investigations. Other objects from this collection were used for comparison: three scarabs of different sizes and a necklace with amulet representing the god Thoth. The investigations consisted of EDXRF, pXRF, SEM-EDX, FTIR, FTRaman, radiocarbon dating analyses. The results and interpretations of multidisciplinary investigations led to a series of conclusions that will eventually be presented to specialists, namely: the necklace was formed during the dynastic Egypt from pieces belonging to different periods, the scarabs are also from different periods, and the mummy hand with the attached amulet are from a later period of dynastic Egypt than the one in which they were originally classified. The bandage, initially associated with a later period than the human tissue, will be re-dated using an original pre-treatment variant, for the final validation of the radiocarbon data.*

Rezumat: *Articolul de față este o primă încercare de analiză a unor obiecte egiptene din colecția „Maria și Dr. George Severeanu”, parte din Muzeul Municipiului București. Piesa cheie, cea care a declanșat prezentul studiu, este reprezentată de o mână de mumie (palmă) cu resturi de bandaj și un fragment de amuletă atașat. Părerile unor specialiști din România și din Marea Britanie au dus la concluzii ușor divergente, ceea ce a condus la începerea investigațiilor arheometrice. Pentru comparație s-au folosit și alte obiecte din această colecție: trei scarabei de diferite mărimi și un colier din mărgelile tubulare cu amuletă reprezentându-l pe zeul Toth. Investigațiile au constatat în analize EDXRF, XRF, SEM-EDX, FTIR, FTRaman și*

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datare radiocarbon. Rezultatele și interpretările analizelor multidisciplinare au condus la o serie de concluzii care în final vor fi prezentate specialiștilor, și anume: colierul a fost format în perioada Egiptului dinastic din piese aparținând unor perioade diferite, scarabeii sunt și ei din perioade diferite, iar mâna de mumie și amuleta atașată sunt dintr-o perioadă mai târzie a Egiptului dinastic decât cea în care au fost inițial încadrate. Bandajul, asociat inițial unei perioade mai târzii decât țesutul uman, va fi redatat folosind o variantă originală de pretratere, pentru verificarea finală a datelor radiocarbon.

Introduction

The investigation was triggered by the discovery in 2017 in the attic of the main building of the museum, during the restoration and modernization of the permanent exhibition, of a mummy's hand (Pl. I.1a-b) without associated documents or inventory sheet. It was most likely purchased by the Severeanu family in the first decades of the 20th century for their collection of Egyptian curiosities. In order to actualize an inventory sheet and display it in the showcase, some information on the certified provenance was needed. During the first stage of the investigations, the hand was taken to a medical centre, for X-ray radiography (Pl. I.1c). Thus, the hidden details could be observed without acting invasively on the object. In general, all archaeometric analyses were performed by non-invasive or minimally invasive techniques, except for radiocarbon dating or FTIR analyzes.

This first set of information was presented, together with the photo details (Pl. I.1-2) to Egyptologists from the United Kingdom (Janet Picton, Petrie Museum, London) and Romania (Miron Ciho, Faculty of History, University of Bucharest.).

According to Janet Picton expertise, despite the lack of information, however, the presence of a blue faience amulet, on which hieroglyphs are inscribed, places it during the dynastic period of Egypt. The only indication regarding its dating is given by the amulet's partially preserved text, which reads "The King's wife", most probably Tiye, who lived between c. 1398 and 1338 BC (the wife of Amenhotep III, the 18th Dynasty, the New Kingdom).

A relative chronology was issued by comparing it with another piece from the Metropolitan Museum of Art, London: "*Definitely hmt nsw... = King's wife, and most probably hmt nsw Tiye (guessing from the top of the two reeds) = King's wife Tiye. Wife of Amenhotep III. The ring would have been a festival gift to the deceased. Here's a similar cartouche ring of the queen in the Metropolitan Museum of Art (11.215.83. from Malqata House 2, 1911-12 excavations).*" (Janet Picton)

According to Miron Ciho's observations on the amulet, it seemed to be „*a later or contemporary imitation. However, a scarab near the fingers is unusual, but it does not seem to have been used as a seal and would have remained on the finger. Eventually an anthropologist can tell if it is something modern or older... What matters from the historical perspective can be found out by determining the composition of the glaze.*"

The inconsistencies that arose regarding the correct classification in a certain period of dynastic Egypt could be summarized by the following questions:

- (i) Is the amulet manufactured between 1398 and 1338 BC or immediately after?
- (ii) Is it a later copy?
- (iii) Is it an imitation from the period of dynastic Egypt?
- (iv) Is it a modern / contemporary imitation or forgery?

Materials and methods

This study invites you to (re)discover the “Maria and Dr. George Severeanu” Collection in the Museum from Henri Coandă Street No 26 Bucharest, and various objects from the Egyptian collection: a mummy’s hand for which archaeometric investigations provided new data about the amulet, human tissue and bandage; pieces of a necklace: a few beads, pendant, cord; various amulets in the form of a scarab (Tab. 1, Pl. I-II)¹.

| MMB Inventory No. | Description | Dimensions |
|-------------------|--|--|
| FN 93 | The left palm of a mummy’s hand with a blue amulet (Pl. I.1-2) | length=20 cm; width=6,5 cm; weight=97 g |
| 19489 | Necklace made of blue tubular beads on a linen cord and blue amulet representing the god Thoth (beads can be analysed individually) (Pl. II.3) | beads length =4 cm amulet length =7 cm amulet width=4 cm |
| 19713 | Little blue-green glazed soapstone scarab (Pl. II.4) | length=1 cm; width=0,7 cm |
| 19715 | Medium blue-green glazed soapstone scarab (Pl. II.5) | length=1,5 cm; width=1 cm |
| 19714 | Large blue faience scarab (Pl. II.6) | length=7 cm; width=4,5 cm |

Table 1. General description of the Egyptian objects included in this study

Archaeometry was used to give justified answers. First, the amulet was analysed by EDXRF at the MMB and then by pXRF and FTIRaman at IFIN-HH. The slightly inconsistent and inconclusive results in this stage of the study led to widening the number of Egyptian objects analysed from the Severeanu Museum’s collection and of the analytical methods.

Thus, they were investigated at the museum, using a portable XRF / FTIRaman device: the amulet on the mummy’s hand and a series of three scarabs of different sizes, together with the beads and the pendant of a necklace (Pl. I-II). Due to the analysis / methods’ limitations, namely the surface depth profile, and due to the specificity of the outer layer of the investigated objects (glazes), the results were not in line with expectations.

For this reason, all the objects were taken to Măgurele research platform where XRF (at IFIN-HH) and SEM-EDX (at INCDFM) investigations were continued. Subsequently, as secondary set of analyses to supplement the information, FTIR (analysis of yarns from the mummy’s bandage) at IFIN-HH and MNIR, and radiocarbon dating at IFIN-HH of human tissue, mummy bandage and textile fibers from the necklace were used.

EDXRF

The first geochemical characteristics of the hand amulet were determined using a portable (fourth generation Genius) X-ray fluorescence spectrometer. This instrument comprises an X-ray

1. Opreș *et alii* 2019.

tube with an Ag anode with maximum voltage and current of 50 kV and 100 mA respectively. The system includes an ANTEK Silicon Drift Detector with big window, 25 mm² active area and a resolution higher than 139 eV. The chemical data was acquired with a beam of 6 mm diameter on the sample, having a penetration of 100 µm in depth. The calibration curve of the spectrometer was set on “Mineral mode”. The compositional data was gathered with a voltage of 45 kV, a current of 60 mA and a measuring time of 100 seconds. The detected concentrations were of minor and trace elements that ranged from K to Pb, while the major elements specific to faience composition (as Si and Al) were not measured due to the technical limitations of the EDXRF spectrometer.

pXRF

pXRF measurements; IFIN-HH: were made using a portable XRF, Bruker Tracer S1 Titan. The spectrometer uses a Rh tube to generate an X-ray beam with an energy of 40 kV and a current of 30 µA. A collimator with a diameter of 3 mm was used to reduce as much as possible the beam that reaches the target. The device cover placed above the samples was not used during the recording of the spectra. The recorded spectra were analyzed using device-specific software, Bruker Instrument Tools. The operating mode was “Mineral Mode”.

SEM-EDX

For scanning electron microscopy and elemental analysis, a TESCAN LYRA 3XMU electron microscope and a Bruker Quantax 200 detector were used at INCDFM. The acceleration voltages were 15 kV and the size was correlated with the area of interest of the object to be analysed.

ATR-FTIR and FTRaman

ATR-FTIR and FTRaman measurements; IFIN-HH: the spectra were recorded on a Vertex 70 Bruker FTIR / FTRaman spectrometer with solid rock interferometer, equipped with Helios micro DRIFT, Raman module (RAM II), and Raman probe (RAMPROBE) for non-destructive analysis of solid and liquid samples. All FTIR measurements were performed in a diamond crystal ATR-FTIR cell, at room temperature. FTIR spectra were recorded using 32 scans with a resolution of 4 cm⁻¹ in the spectral region 4000 - 400 cm⁻¹. The FT-Raman module was equipped with NIR LASER excitation source with a wavelength of 1064 nm (Nd: YAG) and adjustable power between 1 and 500 mW, ultrapure liquid-cooled germanium semiconductor detector. All measurements were performed using 64-1064 scans, laser power 1-500 mW and 4 cm⁻¹ resolution. The FT-Raman configuration offers a spectral range of 3600 - 50 cm⁻¹. The spectra were imported and processed in the OPUS software application version 6.0, provided by the equipment manufacturer.

XRF and Raman Portable Spectrometer is an innovative portable spectrometer designed to perform elementary and molecular analyses combined *in situ*, fast and non-destructive, using complementary EDXRF and RAMAN techniques. EDXRF Large area silicon radiation detector (SDD): active area of 25 mm². Raman High performance thermoelectric cooling fibre optic spectrometer: Raman analysis range is 120-3650 cm⁻¹, resolution type 5-6 cm⁻¹, 1064 nm excitation laser, thermo-electric cooling for laser temperature control, optical power up to 500 mW.

FTIR

FTIR measurements; MNIR: a Tensor 27 spectrometer (Bruker Optics) was used for analysis. The working parameters used were: wavelength range 4000 - 400 cm⁻¹, 32 scans,

resolution 4 cm⁻¹. Prior to the analysis, a background was recorded on a KBr pellet (no sample). Spectrum acquisition and data processing were performed with specialized software, Opus 7.0.

Radiocarbon dating

For radiocarbon dating, linen fibres were collected from the bandage and human tissue was taken from a less visible part inside the palm. The pre-treatment took place according to recommendations from the literature². The pre-treated tissue was graphitized and the elemental carbon was subsequently dated by the AMS method at IFIN-HH³. Graphitization was performed on the EA: AGE-3 system (ETHZ, Ch). Measurements were made at the Cockcroft-Walton Tandetron 1 MV accelerator (HVEE, The Netherlands). $\delta^{13}\text{C}$, [‰] as a general process parameter and CRA \pm SD value, [BP years] were obtained using BATS 4.0 program⁴. Calibration was performed using OxCal Online program and IntCal 13 / IntCal 20 calibration curves.

Results

EDXRF analysis of the mummy's hand amulet

According to the observations made by Ingrid Poll, former chemist at the Bucharest Municipality Museum:

“The object is blue - turquoise and has the appearance of an Egyptian faience. As the piece cannot be detached from the mummified hand, stratigraphic observations cannot be made to specify whether it is a ceramic faience covering, whether it is a coloured faience in the whole mass or whether it is steatite covered with a layer of faience. What has been confirmed is that the colouring agent is copper-based. The absence of potassium suggests the use of sodium / natron as a flux, along with calcium and copper completing the composition of blue frits known as “Egyptian blue”. The presence of tin and traces of lead in the determined composition suggests the use of bronze as a source of copper for colouring the mixture.”

FTRaman investigations

FTRaman analysis of the amulet resulted in a spectrum (Pl. IV.9) that could not be associated with similar spectra obtained in the laboratory for certified specimens of blue chalcedony from South Africa (Pl. IV.10) or lapis lazuli from Afghanistan (Pl. IV.11). The two specimens were analysed in the hypothesis of an amulet made of a solid semi-precious stone (chalcedony) or starting from a natural pigment widely used in the 18th Dynasty (lapis lazuli). Nor could the comparison with spectra and literature data on some natural pigments such as lazurite, azurite, or blue pigment (known as Egyptian Blue) could provide a clear association with a pigment, and thus its identification in databases⁵.

The subsequent analysis of the amulet at the museum, in association with pieces from the collection, could not provide additional information. Therefore, the objects were brought to Măgurele, and the analyses were resumed using complementary techniques.

pXRF and SEM-EDX investigations

The collection analysed by pXRF and SEM-EDX methods includes seven distinct

2. Quarta *et alii* 2013; Richardin *et alii* 2017a; Richardin *et alii* 2017b.

3. Stan-Sion *et alii* 2015; Sava *et alii* 2019.

4. Wacker *et alii* 2010

5. Smith, Clark 2004; Schmidt *et alii* 2009

objects, without historical and archaeological connection between them, except the pendant and the beads in the necklace (where the association of the moment of their last reunion in the necklace could be proven by radiocarbon dating of the cord): one amulet attached to a hand of an Egyptian mummy – Hand Amulet; one small scarab – Little scarab; one medium scarab - Medium scarab; one large scarab - Large scarab; one necklace, of which were analysed: one pendant and two different beads, without undoing the necklace – Thoth Amulet; Faience bead 1; Faience bead 2 (Table 2).

The glaze on the Egyptian artefacts from the Severeanu collection was coloured by using copper based pigments. It was not possible to identify either by pXRF technique or by the complementary SEM-EDX, any association with raw materials or traces of bronze reuse (absence of tin).

At first sight, Bi-plot of the first and second principal components resulting from PCA (Principal Components Analysis, Minitab 17.0 Program) (Pl. III.7) run on chemical concentrations (major, minor and trace elements in ppm) revealed that:

- Measurements made on the same sample are well correlated and this demonstrates both the relevance of the measurements and the good homogeneity of the glaze;

- Variable distribution of the items' chemical concentrations indicates a distinct composition of the glaze that can be related both to different raw material sources and to specific recipes of mixing the glaze ingredients (site specific or dynastic specific recipes);

- The similarity observed between the Hand Amulet, Thoth Amulet and Faience Bead 1 can be a clue for a common site production, the same geological area for the raw materials used or same recipe / period of manufacturing.

SEM-EDX analyses were performed only on Little, Medium and Large scarabs; the analysis of other items in the collection was not safe. They must be introduced into the *vacuum* chamber of equipment. The results and associated SEM images are presented in Tables 3-5. The SEM images are not yet fully understood and interpreted up to now.

The complementarity of these two methods stands out when assessing the results obtained. Although there are differences in the type of analysis technique, detection / element limits, or data processing software, EDX brings important additional information to this study. In principle, this new information shows the presence of light elements, their chemical combinations and, in the particular case of these surfaces, the presence / absence of sodium. Its correct quantification provides clues to the low amount of percentages in XRF experimental data, but also to the recipes used in the past⁶.

6. De Viguerie *et alii* 2009; Okkelberg 2011.

| Object | Measured on | Si | S | Ca | Ti | Mn | Fe | Ni | Cu | Zn | As | Sr | Rb | Th | Pb | Y |
|----------------|----------------|-------|------|-------|-----|----|------|------|--------|------|-------|-----|----|-----|------|------|
| Hand Amulet | Smoothed P1 | 29579 | 468 | 856 | 59 | 13 | 515 | 140 | 38290 | 268 | 251 | 63 | 18 | 8 | 29 | nd |
| Hand Amulet | Smoothed P2 | 34596 | 537 | 757 | 24 | 18 | 378 | 110 | 33292 | 249 | 234 | 63 | 18 | 8 | 29 | nd |
| Hand Amulet | Smoothed P3 | 31058 | 642 | 1011 | 61 | 16 | 577 | 68 | 32747 | 240 | 242 | 70 | 19 | 7 | 33 | nd |
| Hand Amulet | Smoothed P4 | 34563 | 975 | 1661 | 71 | 15 | 767 | nd | 20730 | 177 | 200 | 59 | 14 | nd | 27 | nd |
| Hand Amulet | Smoothed P5 | 36497 | 613 | 1292 | 52 | 20 | 535 | 117 | 39628 | 299 | 229 | 58 | 16 | 6 | 30 | nd |
| Hand Amulet | Smoothed P6 | 34808 | 555 | 809 | 35 | 18 | 415 | 107 | 35208 | 285 | 242 | 58 | 16 | nd | 30 | nd |
| Hand Amulet | Inscription P1 | 23150 | 505 | 852 | 31 | 12 | 422 | 77 | 29358 | 190 | 259 | 62 | 14 | 6 | 27 | nd |
| Hand Amulet | Inscription P2 | 26853 | 440 | 779 | 46 | 13 | 424 | 92 | 33344 | 247 | 242 | 65 | 19 | 7 | 30 | nd |
| Hand Amulet | Inscription P3 | 31589 | 644 | 1081 | 60 | 12 | 578 | 91 | 31247 | 222 | 267 | 71 | 18 | 7 | 35 | nd |
| Hand Amulet | Inscription P4 | 30277 | 544 | 1097 | 36 | 15 | 567 | 94 | 37759 | 283 | 257 | 67 | 16 | 7 | 30 | nd |
| Little scarab | Front1 | 27550 | 692 | 4245 | 133 | 30 | 5751 | 1022 | 76739 | 533 | 2103 | nd | nd | 6 | 30 | nd |
| Little scarab | Back1 | 18938 | 824 | 2623 | 72 | 25 | 4934 | 834 | 27184 | 258 | 1602 | nd | nd | nd | 27 | nd |
| Medium scarab | Front2 | 19368 | 8436 | 2160 | nd | 30 | 3998 | 1698 | 98379 | 565 | 22307 | 36 | 8 | 169 | 1807 | 502 |
| Medium scarab | Back2 | 26669 | 7591 | 1124 | nd | 34 | 1151 | 1181 | 189131 | 1031 | 65863 | 98 | 10 | 457 | 2898 | 1287 |
| Large scarab | Front3 | 25756 | 712 | 6816 | 47 | 17 | 1173 | 398 | 219499 | 2339 | 5078 | 630 | nd | 48 | 356 | 202 |
| Large scarab | Back3 | 25892 | 1390 | 7517 | 63 | 19 | 1376 | 405 | 238588 | 2463 | 5081 | 636 | nd | 40 | 331 | 203 |
| Toth Amulet | Toth P1 | 40253 | 1781 | 1608 | 23 | 11 | 559 | 77 | 45960 | 299 | 2665 | 52 | nd | 32 | 532 | 117 |
| Toth Amulet | Toth P2 | 37560 | 1568 | 1303 | 54 | 21 | 512 | 58 | 35336 | 227 | 1887 | 31 | nd | 21 | 340 | 84 |
| Faience bead 1 | Bead 1 | 35575 | 647 | 740 | nd | 13 | 188 | nd | 16029 | 138 | 477 | 29 | nd | nd | 71 | nd |
| Faience bead 2 | Bead 2 | 29268 | 2721 | 11663 | 84 | 14 | 1174 | 213 | 96907 | 594 | 4800 | 382 | nd | 63 | 710 | nd |

Table 2. The concentrations of the chemical elements detected in the Egyptian faience surfaces (glaze), analysed by XRF (ppm) (nd=not detected)

The main inconvenient of XRF and SEM-EDX data comparison is due to the fact that the PCA analysis found similarities only between the Hand Amulet, Thoth Amulet and Faience bead 1, but not with any of the scarabs. Thus, the preliminary interpretations on the composition in SEM-EDX cannot be extrapolated to the Hand Amulet - the central object of the compositional analysis researches.

However, if we compare XRF data with EDX data for scarabs, we notice the contribution of light elements and their combinations to the increase of the determined overall mass percentage. By default, it is a proof that the values up to 15%, usually obtained by XRF, are due to the nature of the surface. For glazes, the compounds / oxides of light elements are concentrated in the first layers, both due to the production technology and due to the different densities of the oxides. In principle, the basic source for sodium is natron⁷.

7. Bezur, Casadio 2012; Abadir 2014.

Natron is a flux remaining in the upper layers of the scarab or amulet and cannot be detected in XRF, and in the FTIR spectra. In FTIR, it most likely causes a “diffuse background” of sodium silicate that overlaps the spectrum of the majority of silicon dioxide, being able to lead to the allure of the spectrum obtained at IFIN-HH (Pl. IV.9)⁸. The distinctive elements given by the pigment become a minority. Deconvolution of the spectrum and recording at different wavelengths could greatly improve the results. SEM-EDX technique remains more useful, highlighting the combinations of sodium, when heavy elements and their combinations are moved to the inner layers, harder to detect.

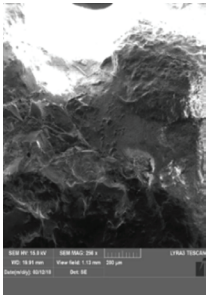
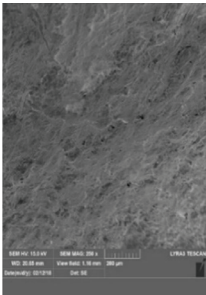
| 19713 | Glazed area | Unglazed area | SEM images | |
|--------------------------------|--------------|---------------|---|--|
| | % norm | % norm | | |
| Na ₂ O | 6.28 | 6.44 |  |  |
| MgO | 2.15 | 1.92 | | |
| Al ₂ O ₃ | 1.44 | 2.32 | | |
| SiO ₂ | 13.55 | 0.68 | | |
| K ₂ O | 0.30 | 0.25 | | |
| CaO | 0.63 | 0.14 | | |
| TiO ₂ | 0.17 | 0.01 | | |
| CuO | 0.01 | 0.01 | | |
| Rb ₂ O | 13.77 | 1.70 | | |
| PbO | 0.45 | 0.76 | | |
| Total | 38.78 | 14.22 | | |

Table 3. EDX results, expressed as normalized mass percent and SEM images for Little scarab

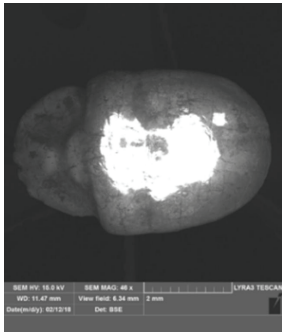
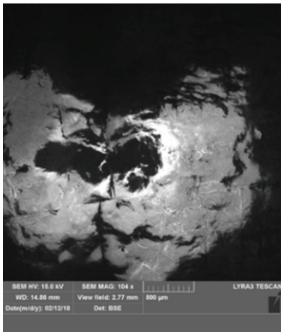
| 19713 | Rear area | Glazed area | SEM images | |
|--------------------------------|--------------|--------------|---|--|
| | % norm | % norm | | |
| Na ₂ O | 7.54 | 7.36 |  |  |
| MgO | 2.74 | 3.63 | | |
| Al ₂ O ₃ | nd | nd | | |
| SiO ₂ | 17.17 | 23.32 | | |
| SO ₂ | 1.60 | 1.42 | | |
| Fe ₂ O ₃ | 1.72 | 3.25 | | |
| K ₂ O | nd | nd | | |
| CaO | nd | 0.57 | | |
| TiO ₂ | nd | nd | | |
| CuO | nd | 1.56 | | |
| Rb ₂ O | nd | nd | | |
| PbO | nd | nd | | |
| Total | 30.78 | 41.13 | | |

Table 4. EDX results, expressed as normalized mass percent and SEM images for Medium scarab

8. Bio-Rad 2017.

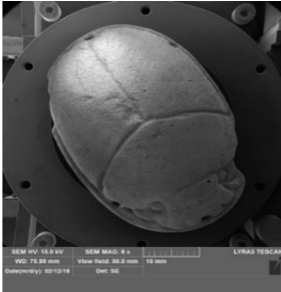
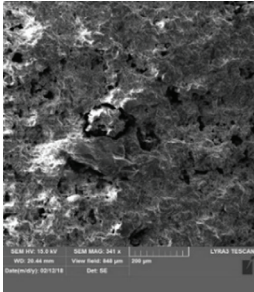
| 19714 | Rear area | Glazed area | SEM images | | | |
|--------------------------------|--------------|--------------|---|--|------------------|--------------------|
| | % norm | % norm | | | | |
| Na ₂ O | 4.22 | 4.24 |  |  | | |
| MgO | 1.63 | 2.05 | | | | |
| Al ₂ O ₃ | 2.92 | 2.76 | | | | |
| SiO ₂ | 47.20 | 48.72 | | | | |
| K ₂ O | 0.23 | 0.34 | | | | |
| CaO | 2.16 | 2.74 | | | | |
| TiO ₂ | 1.16 | 1.02 | | | | |
| CuO | 10.12 | 10.56 | | | | |
| Rb ₂ O | 1.17 | 1.21 | | | | |
| PbO | 2.75 | 5.53 | | | | |
| Total | 73.60 | 79.21 | | | Rear area | Glazed area |

Table 5. EDX results, expressed as normalized mass percent and SEM images for Large scarab

The need to find out an algorithm for correlation between XRF and EDX data

The algorithm built in Excel 2016 started from the following input parameters:

1. XRF experimental results; mass composition per determined element, ppm;
2. Preliminary results; transforming concentrations [ppm] into mass percentages per element [%], and then into mass percentages per element oxide [%];
3. Intermediate results; calculation of correlation coefficients between measurement points, using Correl function of Excel 2016 program; reaching homogeneity criteria;
4. Intermediate results; calculation of the relative average composition of the pigment “Egyptian Blue” (Egyptian Blue) (EB) in the mass of the amulet, determined from the surface to the penetration depth of the beam.

Egyptian Blue; Theoretical calculation: (CaCuSi₄O₁₀), M = 375.958; 376 g / mol; 10.66% Ca; 16.90% Cu; 29.88% Si; 42.56% O or reported as mass percentages per element oxide: 14.91% CaO; 21.16% CuO; 63.93% SiO₂. Theoretical ratios between oxides: SiO₂ / CuO = 3.02; CuO / CaO = 1.42.

The experimental ratios between oxides of the pigment, supposed to be contained in the material of the Hand Amulet, and established *de facto* as being Egyptian faience, met the general classification criteria, and the normalized sum of CaO + CuO + SiO₂ (1.56 + 36.47 + 61.97 = 100%) provide a pigment purity of 96.41% by reporting 128970.3 ppm to 133770 ppm, the total sum of elements oxides.

We considered that SiO₂, CuO and CaO are involved only in the Egyptian Blue pigment and that they cannot exist in other individual compounds or in combinations with other determined elements in faience. Compared to the theoretical values of the ratios between oxides: SiO₂ / CuO = 3.02; CuO / CaO = 1.42, the experimental ratios are different, respectively SiO₂ / CuO = 1.70; CuO / CaO = 23.38. If SiO₂ / CuO ratio difference can be explained by comparison with literature data, the second is very different from the theoretical value but also

from the experimental values in the literature, expressed in mass percentages⁹. For example, for the pigment measured on the Bust of Nefertiti, Berlin, Germany (New Kingdom), c. 1340 BC (period very close to the estimates of the Hand Amulet' manufacturing, according to Janet Picton, see above), the ratios are $\text{SiO}_2 / \text{CuO} = 1.73$; $\text{CuO} / \text{CaO} = 1.73$ (given that the purity of the pigment was $\sim 100\%$ and the applied pigment was measured directly, not as pigment in faience!).

As a first approximation, we could say that the $\text{SiO}_2 / \text{CuO} = 1.70$ ratio would fall within the limits of the literature interval (average = 2.35, compared to theoretical 3.02; closest to 1.73), while the CuO / CaO ratio = 23.38 is far from these limits (average = 1.48, compared to 1.42 theoretical; very different from 1.73 associated with the $\text{SiO}_2 / \text{CuO}$ ratio for the same object), even if the ratios are expressed to the mixtures as faience and not to the pure pigment. Basically, the limiting element would become calcium, respectively CaO which dictates the amounts of SiO_2 and CuO actually involved in EB, and finally, the percentage of EB in the mixture on the surface of the amulet.

All this leads up to 13.50% total oxides of the elements, a value actually determined by the XRF method. The question arises how can the rest of up to 100% be highlighted experimentally? The XRF method could not provide an answer to this question. The study of literature and databases suggests that compounds, mixtures of compounds and phase separations appear in the upper layers, leading to a complex and stratified distribution depending on depth profile and analyses to be applied. For compensation, other objects from the set of the Severeanu Museum were measured under similar conditions using XRF and then SEM-EDX methods.

For the Hand Amulet, another 6 points were measured on the deeper blue areas of the amulet surface and another 4 points on the deeply incised areas, using XRF method. Although a number of interesting new data were obtained (Table 2), they did not significantly change the above conclusions.

If we report the mass percentage of copper oxide in Hand Amulet, to the sum of all oxides identified by XRF, for a purity of 96.41% and a total amount of 13.37%, then the effective mass percentage of CuO dispersed in faience glaze is actually 4.70%. According to literature data¹⁰ for similar situations encountered throughout all dynastic periods, it is an indicator (within the limits of propagated measurement errors) of Dynasties 11-12 or Dynasties 21-25, and less of Late 18th. As Dynasty 11-12 is much too early in terms of chrono-typological similarities and text, then the variant of Dynasty 21-25 could be taken into consideration, supporting Miron Cihă's hypothesis.

Thus, the framing of the amulet according to the $\text{SiO}_2 / \text{CuO}$ ratio places it in the 18th Dynasty, but in the conditions in which the comparison is made with pigments applied by painting. If the framing is done according to the mass percentage of CuO determined by XRF, namely 4.15, then the framing could be in Dynasties 13-17, also early in relation to the object. If the framing is done by the mass percentage of CuO in the whole mass of the glaze, then it is 4.70 and can be associated with Dynasties 21-25.

9. Berke 2007.

10. Kaczmarczyk, Hedges 1983; Vandiver 1983.

In other words, a selection criterion must be applied that takes into account the actual composition of the whole mass of material, and not the surface composition, which is also incompletely determined. Thus, the comparison with different materials, belonging to different historical periods will be much more practical, having many values to compare, not only one.

However, these compositions are experimentally obtained by invasive analysis, and complete destruction of the samples. This option cannot be taken into account in the case of the objects from the Severeanu collection. Under these conditions, the cumulative standardization of XRF and SEM-EDX results, using a calculation algorithm can be a good solution that can match the classification in recipes databases.

Thus, it is possible to normalize of oxides contributions toward values comparable to those in the literature. The statistical analysis of the experimental data and literature data may be a solution. In this study, only the values for the three scarabs could be normalized and compared. It is illustrated by calculating correlation factors using Excel 2016 under Windows 10, as parameter control (Pl. III.8).

The total amount of oxides becomes 72.88% for Little scarab, 73.80% for Medium scarab and 95.10% for Large scarab. Moreover, the percentages of copper oxide become: 6.50% for Little scarab, 18.00% for Medium scarab and 28.67% for Large scarab, the values being in the wider range usually found in the literature.

This original part, concerning how to build-up such an algorithm will not be detailed here. It focusses on correlating XRF and EDX data and finding increments to populate XRF data with EDX data.

Hand Amulet; Supplemental information from optical microscopy images of bandage

A small fragment of the bandage, detached from an area inside the palm, was investigated by electron microscopy (Pl. V.12-13). At a scale of 1:200X, the nature of the fibre was highlighted, proving the use of linen.

Microscopic observation and documentation of the untreated mummy bandage sample; MNIR: were made using a Nikon SMZ 1000 stereomicroscope coupled with a Nikon D3100 Kit 18-55mm DSLR camera. A Nikon Eclipse LV 100 microscope to which the same photo camera was coupled, was used to identify the fibre based on morphological characteristics.

Hand Amulet; Supplemental information from FTIR spectra of untreated linen fibres

FTIR spectra of linen fibres, before pre-treatments for radiocarbon dating, were performed at IFIN-HH and MNIR.

IFIN-HH: spectrum was directly compared to those of untreated silk fibres, fragments extracted from osteological material to be radiocarbon dated, unearthed during archaeological surveys at Hurezi Monastery Romania, and with certified flax fibres (Pl. V.14-15).

MNIR: similar infrared spectrum (FTIR) of mummy textile fibres was compared to the spectrum of flax in the IRUG database (www.irug.org) (ICB00058) and with the spectrum of a vegetable resin in the same database (INR00049), highlighting the characteristic signals. A Tensor 27 spectrometer (Bruker Optics) was used for analysis. The working parameters were: wavelength range 4000 - 400 cm^{-1} , 32 scans, resolution 4 cm^{-1} . A background was measured against KBr pellet. Spectrum acquisition and data processing were performed using specialized software, Opus 7.0.

Supplemental information from optical microscopy images of the bandage and from FTIR spectra outlined the nature of fibres, their relatively poor degree of preservation, and the presence of contaminants, especially a vegetal resin, very hard to remove in the subsequent pre-

treatments for radiocarbon dating.

Determining the nature of the resin and recording the spectrum of pre-treated fibres before graphitization (see radiocarbon dating section) will give a comprehensive image concerning the success of pre-treatment and the efficiency of decontamination.

Hand Amulet; Supplemental information from radiocarbon dating

Investigations of the three scarabs have established that their surfaces are the result of different techniques of manufacturing, according to different recipes, and most likely come from different dynastic periods. Although for these three pieces it was possible to make advanced correlations with literature data, none of them could be associated with the Hand Amulet. According to the typological classification, it belongs either to Late 18th, or to some later dynasties, a good correlation being found with the Dynasties 21-25. Due to the fact that these dynasties are historically recorded, they may provide some information on the most probable date when it was manufactured.

However, the correct framing of the amulet and the determination of its antiquity provide only a relative date on the moment of its use in the mummification process. It could be visibly older, contemporary or even newer than the mummy itself. The intersection of the most probable intervals for both the amulet and the date of death will determine the mutual support of the estimated ages.

If for the amulet the analyses EDXRF, XRF, SEM-EDX and FT-Raman could converge for a certain period, in the case of human tissue important are the anthropological analysis, the analysis of the embalming technique and the radiocarbon dating.

The radiocarbon result offers as calibrated interval for $s = 2$ level of confidence, 756 (95.4%) 415 calBC which roughly corresponds to Dynasties 23-27 (including 1st Persian dynasty)¹¹.

By superposing the intervals for the amulet and human tissue, a period corresponding to Dynasties 23-25 emerges.

The first attempt to date linen fibres after pre-treatments according to literature recommendation gives a calibrated interval of 398 (95.4%) 206 calBC. The reason for this discordance with human tissue may be a consequence of the limits of pre-treatments for textile fibres or a later (re) embalming procedures using new linen textiles or resins. The unusual place of the amulet, noted by Miron Ciho, may explain a late reconsideration of the mummy's body or of classical dynastic Egypt.

This discrepancy is the reason why the dating of the mummy's bandage will be resumed, using an original pre-treatment variant that proved to be effective in other cases where persistent contaminations were recorded.

| Lab. ID | Collection No. | Material | Date BP | SD | Calibrated intervals for 2s, calBC |
|----------------|----------------|--------------|---------|----|------------------------------------|
| RoAMS:623.78 | FN93 | Human tissue | 2458 | 32 | 756 (95.4%) 415 |
| RoAMS:622.78 | FN93 | Linen fibre | 2255 | 35 | 398 (95.4%) 206 |
| RoAms: 1774.78 | 19489 | Cord fibre | 2645 | 44 | 902 (95.4%) 773 |

Table. 6. Radiocarbon dates of the organic Egyptian artefacts from the "Maria and dr. George Severeanu" collection

11. Shaw 2003.

Necklace; Supplemental information from radiocarbon dating

From XRF information it resulted that Thoth Amulet and the two beads in the necklace probably belong to different historical periods. Moreover, PCA analysis of the data suggests similarities between the Hand Amulet, Thoth Amulet and Faience bead 1 (Pl. III.7), which could place them in close historical intervals.

In this context, the dating of the necklace cord becomes important because its age would show the moment when the amulet and the beads formed the current necklace from the Severeanu collection.

The Thoth Amulet was evaluated typologically as being made ca. 1000 - 700 BC. Radiocarbon dating of cord consisted of the same steps of pre-treatment, graphitization, AMS measurement, calculations and calibration as above, and provided for confidence level $s = 2$, the range 902 (95.4%) 773 calBCE (Table 6).

The inclusion in the expected historical interval of 1000–700 BC has several implications: it proves the efficacy of the laboratory strategy; confirms the date of making the necklace, moment associated with the manufacture of the pendant and some beads, others being probably recovered from older pieces; by association, it strengthens the hypothesis that the manufacture of the Hand Amulet was done in a more recent historical period than initially expected.

Related to the conclusions regarding the mummy's hand, it can also be deduced that: it is very probable that the amulet production date and the time of the person's death were close, falling in a later period than Late 18th Dynasty, as initially was presumed; the bandage must be re-dated, and if the new radiocarbon date approaches that of the human tissue, together they will be corroborated and restrict the historical interval given by the absolute chronology, as well as they will also support the approximate moment of the manufacture of the Hand Amulet. If the differences remain as in Table 6, then the result suggests a reuse of the amulet, arranged in an unusual position compared to the mortuary practices of classical dynastic Egypt, as suggested by Miron Cihă.

Conclusions

In an attempt to associate the conclusions of the archaeometric study with the initial estimates of the specialists, we could advance the following ideas:

- The three scarabs, both in terms of appearance, manufacturing technology, and typological classification, belong to different historical periods of dynastic Egypt;

- The pendant and the beads of the necklace also belong to different periods, but the cord absolute chronology proved the moment of assemblage of the current necklace as being related to the manufacture of the amulet and some beads in the necklace; other beads may be associated with older periods or other manufacturing workshops; thus its formation corresponds to the style adopted during the Dynasties 21-25¹²;

- The mummified tissue places death and embalming in the Dynasties 23-27, while the amulet placed on the finger, in association with some pieces in the necklace and the data in the literature, suggests the Dynasties 21-25 as the period of manufacture¹³;

12. Shaw 2003.

13. Shaw 2003.

- The mummy's bandage, made of linen, offers a more recent radiocarbon date, placing the (re) embalming in the Late Period or Ptolemaic Period; in this case, re-dating of linen fibres becomes required.

If we analyse the placement of the necklace and the mummy's hand from the Severeanu collection in large historical intervals, we can say that they belong to the dynastic Egypt, Third Intermediate Period. The resumption of the radiocarbon dating of the bandage, although secondary in relation to the strategy of characterizing the faience objects coloured with Egyptian Blue, becomes important in clarifying some aspects related to the piece in the collection that generated the study and its original progress: the mummy's hand. A calibrated date placed in the range obtained for the human tissue will reconfirm the mummy's age. A date that will not be different from the first result will confirm it and shows an interesting aspect of the rituals of Ancient Egypt late periods.

Investigations on Egyptian faience pieces from the Severeanu collection proved the limitations of investigators' experience and the used analyses. However, the analytical methods were developed, adapted and combined, by finding original models of interpretation in the context of literature data. The combination of specialists' presumptions with the interpretations of the experimental results offered by the archaeometric study refuted / confirmed or ranked the first ones.

Indirectly associated analysis methods (FTIRaman, FTIR, optical / electron microscopy, radiocarbon dating) ensure the convergence of the two approaches (specialized and archaeometric / contact chronology and absolute chronology). The questions related to the samples were largely answered, thus justifying the need for the archaeometric study.

Experimental research will continue with the completion of the re-dating of the linen bandage for which an original method of purification was developed using a combination of solvents not used so far in the literature.

The archaeometric study of the selected Egyptian artefacts from the "Maria and dr. George Severeanu" collection showed some similarities but mainly diversity among them, being indicative for a tangled provenance network, specific for the Egyptian artefacts market developed in Europe during the 19th century and at the beginning of the 20th century.

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Bibliography / Bibliografie

Abadir 2014: M.I. Abadir, *Experimental Study of the Egyptian blue degradation by the copper chloride cancer*, Bachelor Degree in Chemical Engineering, 2014.

Berke 2007: H. Berke, *The invention of blue and purple pigments in ancient times*, Chemical Society Review, 2007, 36, p. 15-30.

Bezur, Casadio 2012: A. Bezur, F. Casadio, *The analysis of porcelain using handheld and portable X-ray*

fluorescence spectrometers, in Aaron N. Shugar, Jennifer L. Mass (Eds.) *Handheld XRF for Art and Archaeology*, Leuven University Press, 2012, p. 249-311.

Bio-Rad; *FTIR spectrum of Natron*, 2017.

Kaczmarczyk, Hedges 1983: A. Kaczmarczyk, R. E. M. Hedges, *Ancient Egyptian faience: an analytical survey of Egyptian faience from predynastic to Roman times*, Warminster: Aris and Phillips Ltd., 1983.

Okkelberg 2011: E.I. Okkelberg, *Exploring ancient Egyptian faience with nanotechnology: compositional mappings, microstructure analysis, and modern applications*, PhD Thesis, 2011.

Opriş et alii 2019: V. Opriş, A. Majuru, D. Pîrvulescu, D.A. Mirea, C.A. Simion, *Silent collectors, noisy collections, Tracing the provenance of artifacts from the Severeanu archaeological collection (early 20th century, Bucharest)*, 25th Annual Meeting of the European Association of Archaeologists, Bern, september 6, 2019 (oral presentation).

Quarta et alii 2013: G. Quarta, S. Malgora, M. D'Elia, V. Gaballo, E. Braione, L. Maruccio, C. Corvaglia, L. Calcagnile, *The strange case of the Ankhphakered mummy: results of AMS 14C dating*, Proceedings of the 21st International Radiocarbon Conference, A. J. T. Jull & C. Hatté Eds., Radiocarbon, 2013, 55, (2-3), p. 1403-1408.

Richardin et alii 2017a: P. Richardin, A. Perraud, J. Hertzog, K. Madriga, D. Berthet, *Radiocarbon dating of a series of the heads of Egyptian mummies from the Musée des Confluences, Lyon (France)*, Radiocarbon, 2017, 59 (2), p. 609-619.

Richardin et alii 2017b: P. Richardin, S. Porcier, S. Ikram, G. Louarn, D. Berthet, *Cats, crocodiles, cattle, and more: initial steps toward establishing a chronology of ancient Egyptian animal mummies*, Radiocarbon, 2017, 59 (2), p. 595-607.

Sava et alii 2019: T.B. Sava, C.A. Simion, O. Gâza, I.M. Stanciu, D.G. Păceşilă, G.O. Sava, L. Wacker, B. Ştefan, V.D. Moşu, D.G. Ghiţă, Al. Vasiliu, *Status Report on the Sample Preparation Laboratory for Radiocarbon Dating at the New Bucharest Roams Center*, Radiocarbon, 2019, 61(2), p. 649 – 658.

Smith, Clark 2004: G.D. Smith, J.H.R. Clark, *Raman microscopy in archaeological science*, Journal of Archaeological Science, 2004, 31, p. 1137-1160.

Schmidt et alii 2009: C.M. Schmidt, M.S. Walton, K. Trentelman, *Characterization of Lapis Lazuli Pigments Using a Multitechnique Analytical Approach: Implications for Identification and Geological Provenancing*, Analytical Chemistry, 2009, 81, p. 8513-8518.

Shaw 2003: I. Shaw, *The Oxford History of Ancient Egypt* (Paperback, 2003).

Stan-Sion et alii 2015: C. Stan-Sion, M. Enachescu, A.R. Petre, C.A. Simion, C.I. Calinescu, D.G. Ghita, *A new and compact system at the AMS laboratory in Bucharest*, Nuclear Instruments & Methods in Physics Research Section B - Beam Interactions with Materials and Atom, 2015, dx.doi.org/10.1016/j.nimb.2015.02.059.

De Vignerie et alii 2009: L. de Vignerie, A. Duran, A. Bouquillon, V.A. Solé, J. Castaing, P. Walter, *Quantitative X-ray fluorescence analysis of an Egyptian faience pendant and comparison with PIXE*, Analytical and Bioanalytical Chemistry, 2009, 395 (7), p. 2219-2225.

Vandiver 1983: Vandiver P. B., *Ancient Egyptian Faience, Appendix A: The Manufacture of Faience*, in A. Kaczmarczyk and R. E. M. Hedges (Eds.) *Ancient Egyptian faience: an analytical survey of Egyptian faience from predynastic to Roman times*, Aris and Phillips Ltd., Warminster, England, p. A1-A144.

Wacker et alii 2010: L. Wacker, M. Christl, H.-A. Synal, *Bats: A new tool for AMS data reduction*, Nuclear Instruments & Methods in Physics Research Section B - Beam Interactions with Materials and Atom, 2010, 268, p. 976-979.

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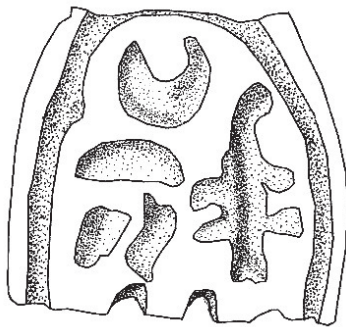
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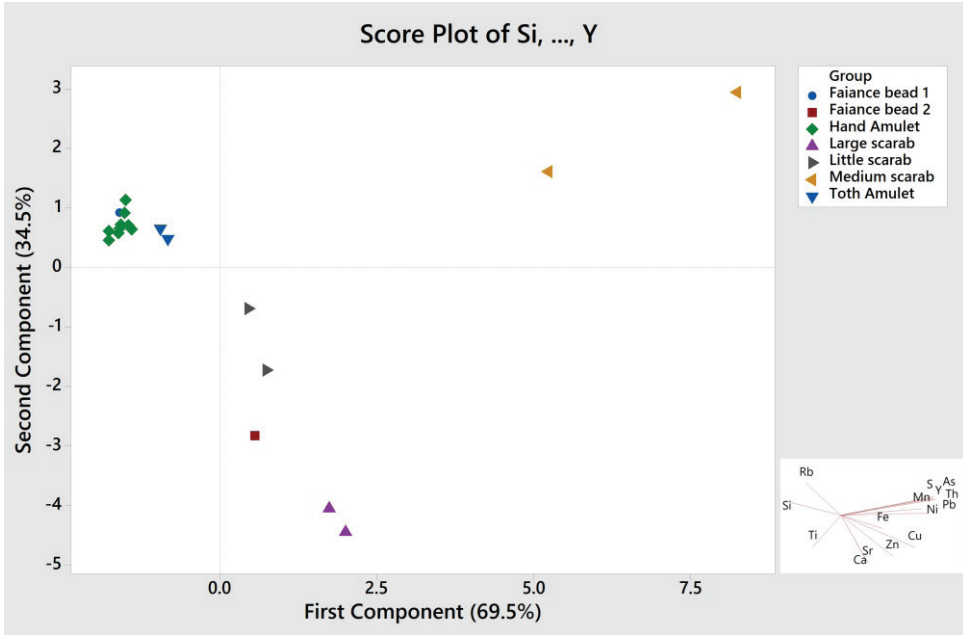


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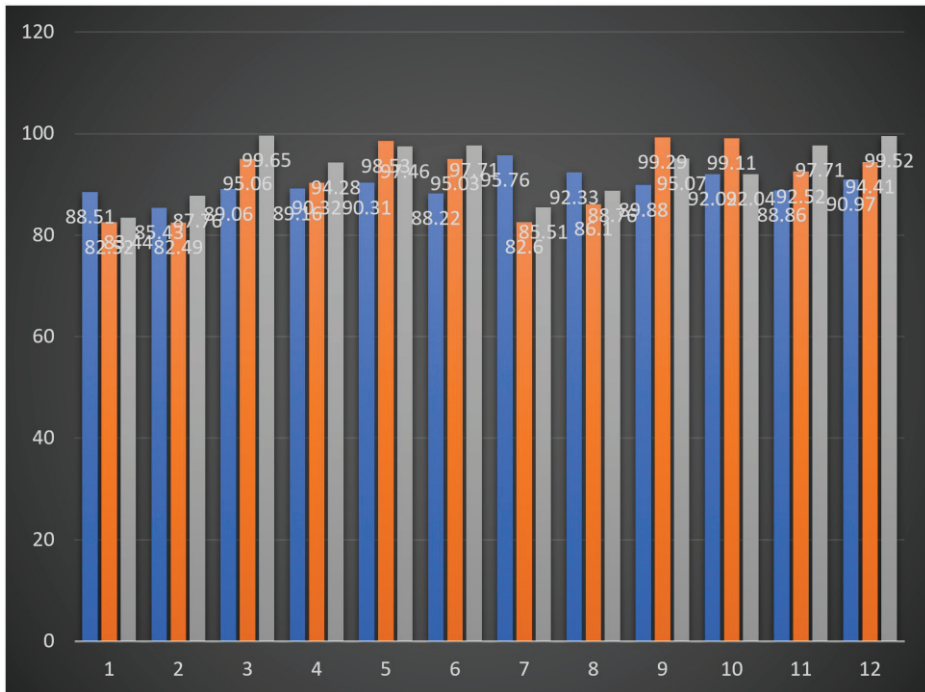
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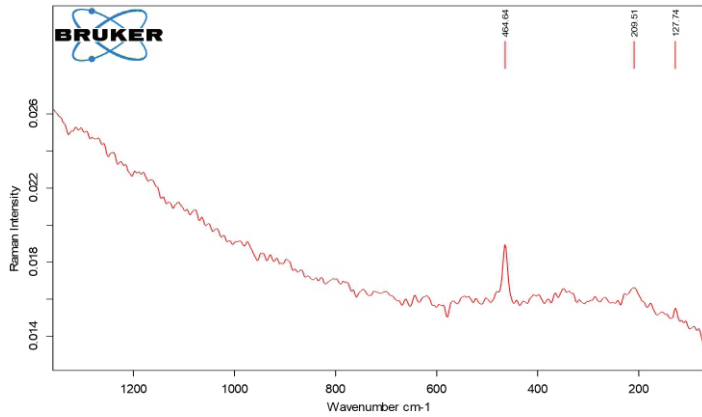


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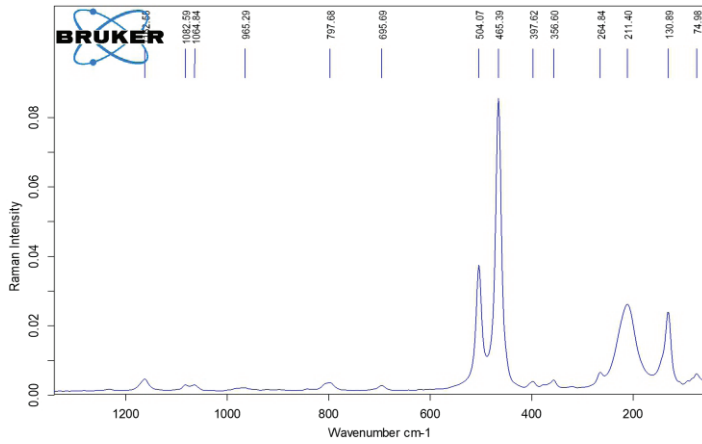


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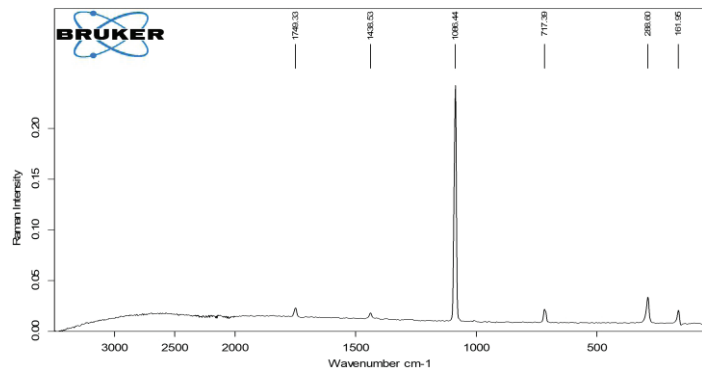
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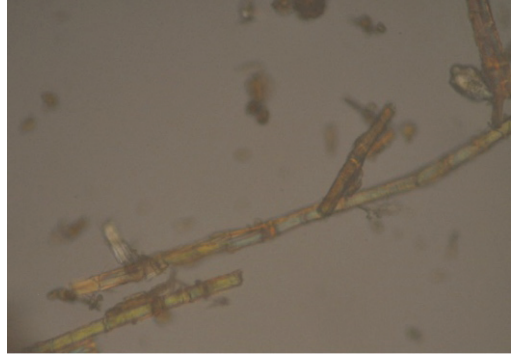


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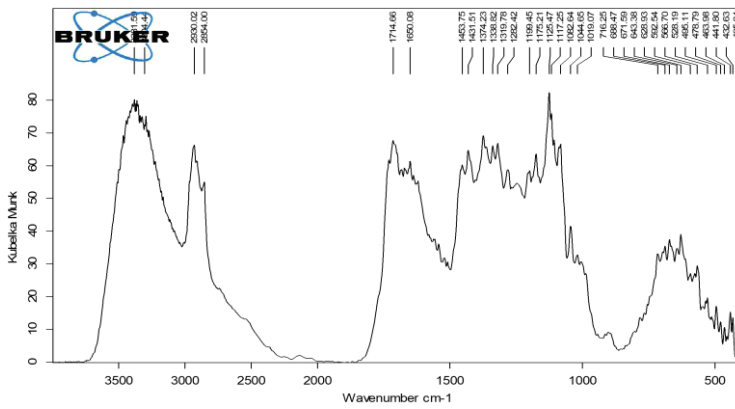
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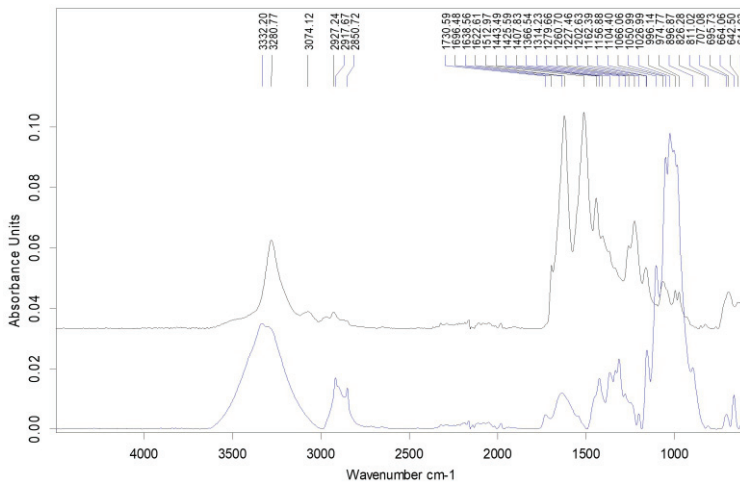
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