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Archaeometrical studies of Greek and Roman silver coins

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Quantitative analyses of various silver coins from the first century BC, found on current Romanian territory (Thasian tetradrachmae, Apollonia and Dyrrachium drachmae, Roman republican denarii) were performed using PIXE (3 MeV external proton beam) and XRF (1.1 GBq ^{241}Am source). The elemental analysis provided evidence of a great variety of monetary alloys and helped Romanian archaeologists to classify the coins, in terms of their provenance, as originals, copies or imitations minted in different areas of the Balkan-Carpathian region.

Introduction

The composition analysis of archaeological objects requires simultaneously non-destructive (the objects are unique and precious), fast (large number of pieces have to be analyzed), versatile (samples with different geometry), sensitive (trace elements are often important) and multielemental methods. Particle-induced X-ray emission (PIXE) and X-ray fluorescence (XRF) are of special interest for archaeologists, because objects of almost any size can be quantitatively analyzed without damaging the samples and without any previous preparation. Romania has a lot of interesting archaeological sites: Greek on the Black Sea coast, Roman in Transylvania and Dobroudja, Byzantine on the Danube banks, where various coins, ceramics, glass and metal objects have been found. Coins (silver, copper, and bronze) are of particular interest. The purpose of our studies is to help Romanian archaeologists to identify coin provenance (workshops, technologies, mines) and explain commercial, military and political aspects.

The large number of silver coins from the first century BC found in the Balkan –Carpathian region have aroused a sharp interest among numismatic researchers.[1] The problem is to classify these coins – tetradrachmae of Thasos (Greek island near Thessaloniki), drachmae of Apollonia and Dyrrachium (ancient Greek commercial cities, now in Albania) and Roman republican denarii into originals, copies and imitations, in relation to their provenance. Among them, the Celtic Thasos tetradrachmae copies (good quality coins, closest to the originals), are well known but also the Barbarian imitations (absence or misspelling of the legend, disproportionate and simplified figures).[2–5] As far as the latter are concerned, it is very important for Romanian numismatics to identify the pieces struck by the Geto-Dacians (Thracian population from the current territory of Romania).[6,7] Barbarians (non-Greek population), i.e., Thracians, Dacians and Celts used to "produce" the same type of coins with Greek imported silver, or, as will be seen in this paper, using local sources of precious metals. Greek silver was probably obtained through salt trade, the Subcarpathian area being the only place in the Balkan region where salt was extracted in those times.[5] The dies used for coining were bought or stolen from the Greeks, while some local engravers manufactured their own dies.

The presence of a large number of silver coins from the first century BC, found on the same sites (different types of coins in the same trove) of the current territory of Romania, was probably due to extensive commercial contacts established by the Greeks (via Balkan mountains and low Danube) and the Romans (via Ilyria – actual Yugoslavia) with local Geto-Dacians. However, this monetization of the local economy is not completely supported by this finding, because all the coins were found as hidden treasures, probably set aside during the troubled times of Dacian king Burebista in the circumstances of the Roman civil war between Julius Caesar and Pompejus.

Visual examination, the first step of a numismatist's work, is not always sufficient to classify the coins. This is the reason why elemental analysis is required. This paper discusses results obtained with some batches of silver coins dating from the first century BC, belonging to Tarii Crisurilor Museum in Oradea and to the Bucharest National Institute of Archaeology. All analyzed coins were found in various places on the Romanian territory, being concentrated in the North-West of Transylvania and the South-East of Muntenia regions.

Experimental

To analyze the coins, two methods were used: External 3 MeV proton PIXE using Bucharest U-120 Cyclotron and energy dispersive XRF, employing a 1.1 GBq ^{241}Am source. Comparing these methods, we found that the PIXE method is faster and has a better sensitivity than the XRF method (3–5 ppm for Ti-Fe-Ni region), but entails a higher cost per analysis.

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The external beam technique makes also possible to examine objects of any size and shape without sampling. External 3 MeV proton PIXE was employed to determine elements with $20 \leq Z \leq 40$ and Au, Hg, Pb, Bi (using L rays) because of its high ionization cross section. For elements with $41 \leq Z \leq 60$ (Ag, Cd, In, Sn, Sb especially), XRF was used as a complementary technique, because of its higher sensitivity for higher Z elements. The method is simpler and cheaper than PIXE, even if a longer times of acquisition are required. The sensitivity in our case is 20–30 ppm (for Ag region). A drawback of the XRF method using a ^{241}Am source is the complicated spectrum of the source.

For external PIXE, we used a proton beam of approx. 3 MeV obtained from nominal energy protons of the 6.5 MeV cyclotron extracted through a 60 μm aluminum foil into air. The gamma-background from the Al foil can be neglected (Fig. 2). The beam passed through a couple of aluminum collimators in order to obtain a well-defined spot on the target (1.5–2 mm diameter). The distance between extraction foil and sample was 25 cm. The current intensity of the beam was 1–2 nA, maintained low to minimize the counting rate. Special holders were constructed for handling the samples. We tried to focus the beam spot only on flat external and bright regions, to avoid the irradiation of possible inclusions or deposits. X-rays were detected through reflection (90°), using a horizontal HPGe detector (5 mm thickness, 10 mm diameter), placed at 8.5 cm distance from the target. The detector resolution was 240 eV at Mn Kα line (5.9 keV). A conventional electronic chain (preamplifier and amplifier) and MCA (multichannel analyzer) were used to accumulate the spectra. To monitor the beam we used an air ionization chamber, consisting of a pair of insulated metallic plates, at 1 kV potential, which integrated the beam. XRF measurements were performed with a spectrometer consisting of a 1.1 GBq ^{241}Am annular gamma-source attached to a support that defines the angle of the incident photons and collimates the fluorescent X-rays on their path to a vertical Si(Li) detector (resolution: 220 eV at 5.9 keV).

Pure thick metal foils and alloys (Goodfellow) of known composition were used as analytical reference materials. These standards were irradiated (PIXE) or excited (XRF) under the same conditions as the samples to be analyzed. This approach has some drawbacks: because the standard and the sample are not identical, but only similar, enhancement and absorption effects in the real sample introduce some errors. Computer software designed in our laboratory was employed to determine the quantitative results.

The overall uncertainty for the PIXE method was 5% for major elements, 5–10% for minor elements and 15% for trace elements. (Major elements are those contributing 10% to overall composition, minor elements 0.1–10% and trace elements less than 0.1%, down to detection limits – see above). The errors are not only statistical. They also originate from the roughness of coin surface and from the chemical corrosion and/or wearing of the objects, altering the accuracy of the results.

In the case of silver matrix, the concentration values of trace elements are not relevant for the archaeologists.

Results and discussion

181 silver coins were analyzed in order to classify them into originals, copies and imitations. All coins had been struck between 60 and 48 BC, a very intense period of Roman civil wars. After completion of those analyses, the coins could be grouped into several categories, taking into account their chemical composition.

The results are presented in Tables 1, 2, 3, each table containing the compositions for the same type of coins. The values represent concentrations corresponding to those layers analyzed by XRF and PIXE. For these silver coins, excepting samples with higher copper content, the results are also relevant for bulk composition.[8]

Regarding the first group of Thasian tetradrachmae (16 pieces), they appear similar in composition to the so-called Celtic imitations, e.g., the pieces found in Slovenia and Austria, reported by SMIT et al.[3] They were high quality minted coins (Table 1).

The coins from the second group (12 pieces) appeared to be deliberately alloyed with copper (0.7–5%). The low concentration of copper may show that the alloying was done for hardening and not for debasement reasons. Traces of Hg could indicate the use of an amalgamation procedure for silver metallurgy, but additional evidence of this is necessary.

A "fingerprint" of the third group of Thasian tetradrachmae (8 pieces) is the bromine. The presence of this element in silver coins is mentioned in the literature only in one case, in which bromine is linked to marine spray, because the hoard was found near the seacoast.[9] In our study, the coins were found in a region, which is far away from the Black Sea coast (approx. 100 km). Taking into account the presence of bromine in silver ore from some Transylvania mines Rodna (embolite – Ag(Cl Br) and bromargirite – AgBr) and supposing an imperfect procedure of refinement (see also high Cu and Pb contents), these coins could be attributed to local Barbarian (Dacian) workshops. To confirm this hypothesis, further archaeological studies are necessary, e.g., an analysis of a silver jewelry definitely attributed to local craftsmen from this period.

Table 1. Composition of Thasian tetradrachmae

Group No.	Ag, %	Cu, %	Au, %	Pb, %	Br, %	Bi, %	Trace element
I	97–98	0.1–0.7	0.1–0.9	0.2–0.5	–	0.05–0.15	Fe, Sn, Hg
II	94–98	0.7–5	0.1–0.9	0.2–0.5	–	0.1...0.15	Fe, Sn, Hg
III	95–97	2–3.5	0.5–0.8	0.5–0.8	0.1–0.3	Traces	Fe, Sn, Hg

Table 2. Composition of Apollonia and Dyrrachium drachmae

Group No.	Ag, %	Cu, %	Au, %	Pb, %	Br, %	Sn, %	Fe, %	Trace element
I	96–98	0.5–2	0.2–0.8	0.2–0.7	–	Traces	Traces	Bi
II	78–92	4–20	0.2–0.8	2–4	–	Traces	Traces	Bi
III	95–97	0.5–1	0.7–1	0.5–1.0	0.1–0.2	Traces	Traces	Bi
IV	–	67–72	–	0.1–0.4	–	28–33	0.5–1.5	Sb
V (one coin)	–	7	–	0.1	–	90	3	Ni, Sb

Table 3. Composition of Roman denarii

Group No.	Ag, %	Cu, %	Au, %	Pb, %	Br, %	Sn, %	Fe, %	Trace element
I	96–98	0.2–1.6	0.4–1.5	0.2–1.5	–	Traces	0.3–0.7	Bi
II	94–97	0.1–0.4	0.6–1.2	0.1–0.3	0.2–3.3	Traces	0.2–04	Bi

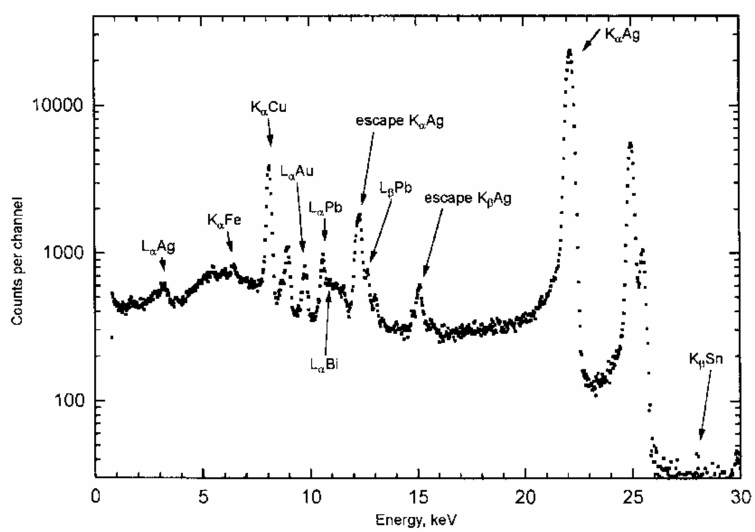

Fig. 1. X-ray spectrum induced by 3 MeV protons of an original Apollonia silver drachma (1st century B.C.)

As for the Apollonia and Dyrrachium drachmae, a similar situation can be retraced (Table 2). Because of the high silver content and the refined aspect of the coins belonging to the first group (32 pieces), we can assume that these drachmae are original ones minted of Macedonian silver (Fig. 1).

The percentage of copper in the second group of coins (48 pieces) is higher than in the previous case. We conclude that copper had been deliberately added to the alloy, reflecting a difficult economic situation during the civil wars.[8] These drachmae were struck in Apollonia, using Dyrrachium dies belonging to the MENISKOS magistrate.

The third group of drachmae (9 pieces) is similar in composition to a category of the tetradrachmae table (the second one): here is again a relatively high content of bromine. We can assume these coins too were minted in the current Romanian territory where silver is mixed with a relatively large amount of bromine in minerals.

Another important group (40 pieces) is that of bronze Dyrrachium drachmae, found in South-East of Romania. Some of these coins were covered with a thin (submicronic) layer of tin, which can be barely noticed by visual examination. The tin layer was strongly corroded. The Cu-Sn proportion was unbalanced, showing a frailty process because of the high content of Sn. Moreover, the absence of Zn results in a certain degree of porosity that can be noticed in some coins. The result of this alloying is a compound named "white bronze" which can easily be mistaken for silver. This was, probably, the intention of the manufacturer. This artifice was used in extreme situations, when silver resources were completely exhausted.

Only one sample differed in composition from the above one (it is listed at the end of Table 2). This Dyrrachium drachma bearing the inscription XENON was probably minted using Serbian tin.

A special case was that of the plated drachmae. The crust (0.3–0.5 mm thick) was made of high purity silver (95–97%). This crust was broken in some areas, revealing an inside core (0.2–0.3 mm) made of bronze (90–97% Cu, 3–10% Sn).

As a possible economic and political explanation for these last categories of coins, one can assume that during the Roman civil wars (1st century B.C.), the access to Macedonian silver mines was often interrupted. Therefore, the local administration started to mint coins just with a silver skin. After ingot exhaustion, the coins were only made of bronze, and the silver was reduced to a thin covering, sometimes even this covering was replaced with tin. These coins have a very elaborate aspect; we can conclude that they were struck using the

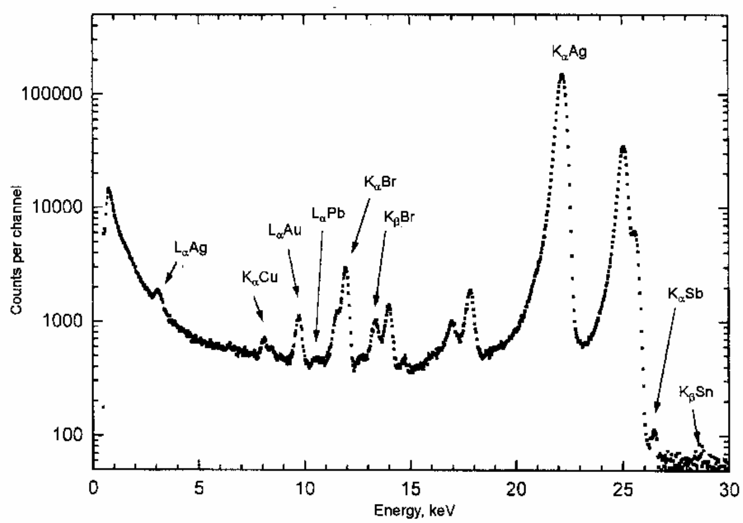

Fig. 2. XRF spectrum of a silver Roman denarius, a possible Dacian imitation

original dies. Romanian archaeologists suppose that these coins were minted particularly in Apollonia to be used as a stipend for Dacian king Burebista in his action as Pompejus' ally against Julius Caesar.

Regarding Roman denarii (Table 3), the coins can be grouped into two main categories. The first group (11 pieces) represents the originals (high silver content), probably coined in Ilyria.

The bromine content of the second group (4 pieces) led us to the conclusion that they were minted using local (Transylvania) ores (Fig. 2).

Conclusions

The elemental analysis reveals a large variation in the monetary alloys used to produce coins in the first century B.C. We can conclude that this work has demonstrated that the use of the PIXE and XRF analytical techniques combined with typical visual examination of figures and inscriptions could help archaeologists to distinguish pieces produced in different regions. The result was a classification of silver coins in terms of their provenance into originals, copies or imitations minted in different areas of Balkan-Carpathian region.

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