Geophysical Research Abstracts,
Vol. 11, EGU2009-257-2, 2009
EGU General Assembly 2009

Micro-PIXE and micro-SR-XRF studies for Romanian archaeological gold identification

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For gold, trace elements are more significant for provenancing archaeological artifacts than the main components: Platinum Group Elements (PGE), Sn, Te, Sb, Hg, Pb, but also high melting point elements, such as Ta and Nb. Several small fragments of native Carpathian gold were studied using micro-PIXE technique at the AGLAE accelerator, Louvre Museum and at the Legnaro AN2000 microbeam facility, and using SR-XRF at BESSY synchrotron. The goal of the study was to identify the trace elements, especially Sn, Sb and Te. At BESSY, the SR-XRF measurements were performed in air by using a 34 keV beam to excite the characteristic X-lines in Sn-Sb-Te region. We found Sn to be present in placers from Valea Arieşului and Valea Pianului, Sb in primary gold from Zlatna, Ruda-Brad, Valea Morii, Runculeţ-Straja and Pb in primary gold from Brădişor-Brad, Zlatna, Runculeţ-Straja, Valea Morii, Muşariu-Brad. Ten native gold nuggets and several fragments of objects coming from Visigothic Pietroasa "The Golden Brood Hen with Its Chickens" hoard were analyzed using micro-PIXE technique at the AGLAE accelerator, Louvre Museum, Paris and at the Legnaro AN2000 microbeam facility. We found Te in primary gold from Brădişor-Brad, Muşariu-Brad (different samples from BESSY analyzed ones), and Roşia Montană, Sb in primary gold at Bucium-Izbiţa. For Pietroasa hoard, we found Sn in the Oenochoe cup and small fibula, indicating that alluvial gold - probably from Anatolia (Pactolus river) - was used. We also detected Ta inclusions in the large fibula, indicating that Ural Mountains (the only region where Ta and Au minerals are together) gold was (at least partially) used. A spectacular application to nine Dacian gold bracelets (belonging to National History Museum of Romania) authentication is presented. These bracelets look like spirals and are based on the same artistic idea, are centred around the same theme. The number of spiral varies from 6 to 8. When uncoiled, some bracelets measure 2.30 m and others even 2.80 m. 4 of them weigh 1 kg each. At each end, the bracelets are decorated with 7 palm-leaf like ornaments. There are no two identical bracelets. The plate is continued with a so-called "protoma", a decorative element which looks like the head of an animal (a wolf, a snake or a dog). The Dacian bracelets were measured using XRF technique (Am-241 and Pu-238 sources and a portable Mo X-Ray tube used to control the homogeneity of the alloy for each bracelet). Compositions (Au-Ag-Cu) very similar to Brad region native gold (primary and placers), but different form bracelet to bracelet, were obtained. Differences in homogeneity, especially Cu content, for each bracelet were observed. Traces of Sn and Sb were also detected. Our conclusion: native gold (mainly alluvial – placers) from Brad region, primitive metallurgy (no refined gold).