



First results on radiometric dating of metals by alpha spectrometry☆

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ABSTRACT

A new TSAC (Thin-Source Alpha Particle-Counting spectrometry) setup of high geometrical efficiency and low intrinsic background was built to measure the alpha particles coming from the ²³⁸U and ²³²Th decay chain radionuclides with concentration as down to five ppm level. It was designed to test the ²²⁶Ra/²³⁸U method, which is the only direct metal dating method proposed to date. Micro-PIXE, micro-RBS and SEM-EDS were used as complementary analytical techniques in order to search for heterogeneities and/or impurities that could bias the TSAC data. Ores, resulting slags and metallic profile from two recent smelting experiments that reproduced ancient techniques and two ancient ores were studied to determine how radionuclides fractionation occurs during smelting. With this TSAC setup the background signal was extremely low, but comparable to the signal from the measured metals. Preliminary results show no evidence of a preferential depletion of ²²⁶Ra in ancient metal with respect to the other radionuclides.

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1. Introduction

Archaeo-metallurgical research focuses in the reconstruction of early trade routes in terms of location of metal sources (ore deposits) and in the study of metallic artifacts and metallurgical remains through various analytical and examination techniques. Important information can be derived from these studies, like ore provenance, contact between different ancient societies and their technological development.

It's not an overstatement to say that the Holy Grail of archaeo-metallurgists is to achieve a method that can directly give the date of the manufacture of a metallic artifact.

Until now, the dating of metallurgical processes is made indirectly, under favorable conditions, by [1]: a) thermoluminescence (TL) and optical stimulated luminescence (OSL) of burnt soil in kilns and crucibles, or of remaining core material in the metal artifact and slag products [2]; b) radiocarbon (¹⁴C) applied on carbon found in trapped slag inclusions (mainly from artifacts); c) an association with other archaeological artifacts found in the same archaeological context; and d) some aging or degradation specificities that may differentiate an old from a new object (e.g. intergranular corrosion is a slower corrosion process).

In 2005 [1] I. Liritzis proposed a radiometric dating method for metals that are produced from ores (native metals excluded), based on the isotopic fractionation that may occur during ore smelting. This fractionation disrupts the secular equilibrium in the ²³⁸U series. In this sense, ²²⁶Ra, a radionuclide that occurs naturally in the decay chain of ²³⁸U with a half-life of 1602 years, tends to be withdrawn by slag products away of the melted metal, as a result of its low specific gravity, 5–6 g/cm³, and its lower melting point (700 °C) when compared to some metals, as for example copper (8.9 g/cm³; 1083 °C).

As a consequence, in a metal, ²²⁶Ra would appear from the decay of its parent, ²³⁸Th (half-life, $t_{1/2} = 80000$ years). Assuming that initially in the metal ²²⁶Ra is not present, then at time t , the ratio between the activities of ²²⁶Ra and ²³⁸Th is given by Eq. (1) [1]

$$\frac{A_{226Ra}}{A_{238Th}} = \frac{\lambda_{238Th}}{\lambda_{226Ra} - \lambda_{238Th}} \left[1 - e^{-(\lambda_{226Ra} - \lambda_{238Th})t} \right], \quad (1)$$

where λ_{226Ra} and λ_{238Th} are the decay constants ($\lambda = \ln(2)/t_{1/2}$) of ²²⁶Ra and ²³⁸Th, respectively. As $\lambda_{226Ra} \gg \lambda_{238Th}$ ($4.33 \times 10^{-4} \text{ yr}^{-1} \gg 8.66 \times 10^{-6} \text{ yr}^{-1}$) the ratio of activities tend to the value $\lambda_{226Ra} / (\lambda_{226Ra} - \lambda_{238Th}) \approx 1$, and the secular equilibrium is attained after around 5000 years. This period is comparable to the elapsed time since the onset of metallurgy in some areas of the globe. Therefore, this growth of ²²⁶Ra back to secular equilibrium with its parent ²³⁸Th is the basis of this method. This model, known as the ²²⁶Ra/²³⁸Th Liritzis method,

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