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Effects of the behaviour of the proton-induced isotopes production on the analysis of ancient alloys

G. Pappalardo [a,b,*], A. Esposito [a,b], G.A. Cirrone [a,b], G. Cuttone [a], S. Garraffo [c,d], L. Pappalardo [a,c], F. Rizzo [a,b], F.P. Romano [a,c], S. Russo [a]

[a] *INFN, Laboratori Nazionali del Sud, Via S.Sofia 62, I95123 Catania, Italy*
[b] *Dipartimento di Fisica e Astronomia, Università di Catania, Via S.Sofia 64, I95123 Catania, Italy*
[c] *CNR, Sezione di Catania dell'Istituto per i Beni Archeologici e Monumentali, Via Sangiuliano 262, I95124 Catania, Italy*
[d] *CNR, Istituto delle Tecnologie Applicate ai Beni Culturali, Montelibretti, I00100 Roma, Italy*

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Abstract

The present work concerns non-destructive ion beam analysis carried out by means of proton-beam-induced nuclear reactions in the 10–30 MeV energy range. We focused our attention on the yield distribution of the isotopes produced in the internal region of a metal alloy. This distribution, which defines the analytical region, displays, at incident energy of about 20 MeV in a bronze based alloy, a bell-shaped curve centred at about 600 μm with an average width of about 400 μm. By changing the incident proton energy it is possible to displace the above region in the interior of the metal body. It should be pointed out that if we neglect to take into account the correct behaviour of the isotope yield distribution in samples with surface inhomogeneities we can obtain erroneous analytical results. We describe some experiments based on proton activation analysis (PAA) carried out at the INFN-Laboratori Nazionali del Sud (LNS) in Catania where we verified the bell-shape of the radioactive isotope distribution. The resulting formulas are given for quantitative analysis.

It is straightforward that, by choosing the appropriate incident proton energy, it is possible to confine the whole analytical region to the interior of the body, thus eliminating any influence of surface effects. We have called this particular approach deep proton activation analysis (DPAA). Examples of its use on archaeological artefacts are given.

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Keywords: IBA; Energetic protons; Cross section behaviour effect; Core analysis; PAA

1. Introduction

In recent years, strong evidence has been accumulated to show that in ancient alloys the content of heavy metals, like Ag, Pb and Sn, exhibits strong surface inhomogeneity [1–6]. The inhomogeneous thickness extends from the surface to the interior ranging from a few microns to about 200 μm [1,2,4,7–10]. In this case, near surface non destructive techniques (like PIXE or XRF) cannot be used to obtain realistic information on the bulk alloy composition [2–5]. As an example, Fig. 1 shows the tin concentration in 26 Greek coins from the Syracuse mint obtained using the superficial XRF method (^{109}Cd and ^{241}Am radioactive sources were used) compared, in the case of two of the coins, with the tin concentration obtained by measuring the alloy powder drilled up to a depth of 800 μm. Results highlight the strong tin enrichment of the surface (up to a factor of 5) [5].

In some cases ion beam analysis (IBA) carried out by means of nuclear reactions induced by energetic protons (some tens of MeV) has been taken into consideration to obtain reliable information on the composition of the

* Corresponding author. Address: INFN, Laboratori Nazionali del Sud, Via S.Sofia 62, I95123 Catania, Italy. Tel.: +39 095 542287; fax: +39 095 7141815.
E-mail address: pappalardo@lns.infn.it (G. Pappalardo).

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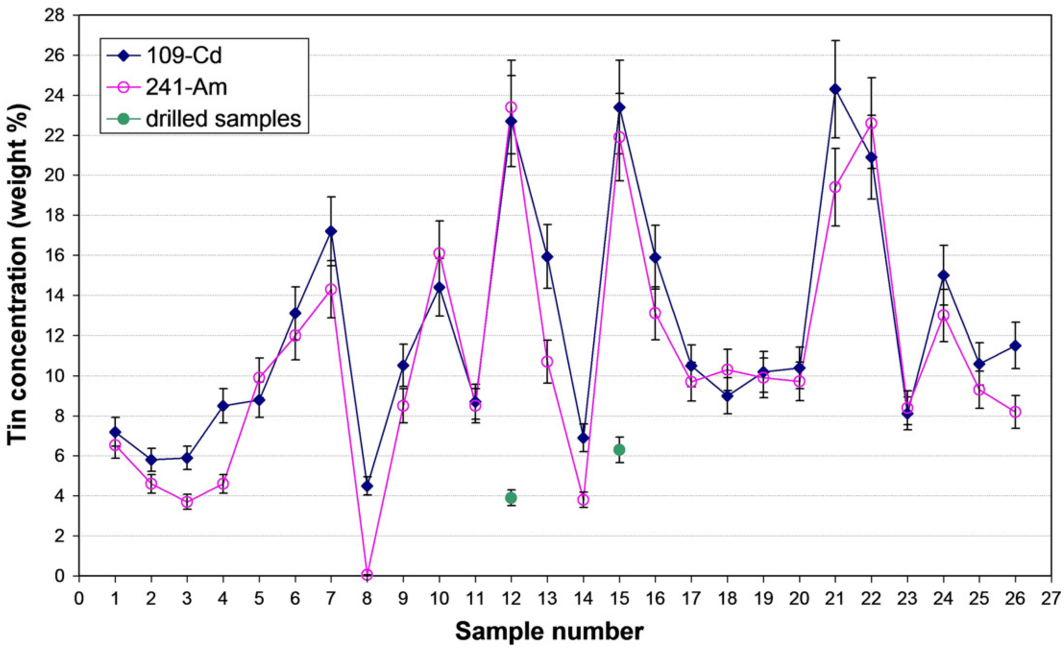

Fig. 1. Comparison between tin concentration XRF measurements obtained using a ^{241}Am (open circles) and a ^{109}Cd (diamonds) X-ray sources and those obtained by measuring the powder from samples 12 and 15 drilled up to a depth of 800 μm (full circles) [5].

metal alloy. However, when using this analytical method, much care has to be devoted in order to correctly take into account the *proton induced production yield in the various layers* of the alloy under analysis. This production yield is directly related to the behaviour of the energy excitation function of the selected nuclear reaction. If this effect is not considered, analytical errors can be introduced in presence of the above discussed inhomogeneities.

In the second section of the present work we analyse and discuss the above effect.

In the third section we show that this effect can be used to restrict the analytical region to only the interior part of the alloy, thus eliminating any influence of the surface layers.

2. Effects of nuclear excitation function behaviour on IBA with energetic protons

In a proton-induced nuclear reaction with incident energy in the 10–30 MeV region the cross section for the production of a given isotope is not constant with energy, but it displays a "bell shaped" trend centred in proximity to the energy corresponding to the Coulomb barrier height and slowly decreasing both at lower and higher energies. A typical example is shown in Fig. 2 concerning the experimental cross section of the ^{130}Te(p,n) ^{131}I reaction [11]. In general, systematics shows that the maximum of the cross section lies at about $E_{inc} = 10$ MeV for proton-induced reactions in medium mass elements. It follows that, with a given incident energy, due to the slowing down of the protons inside the metal alloy sample, the production of isotopes drastically changes in the various layers at different depths.

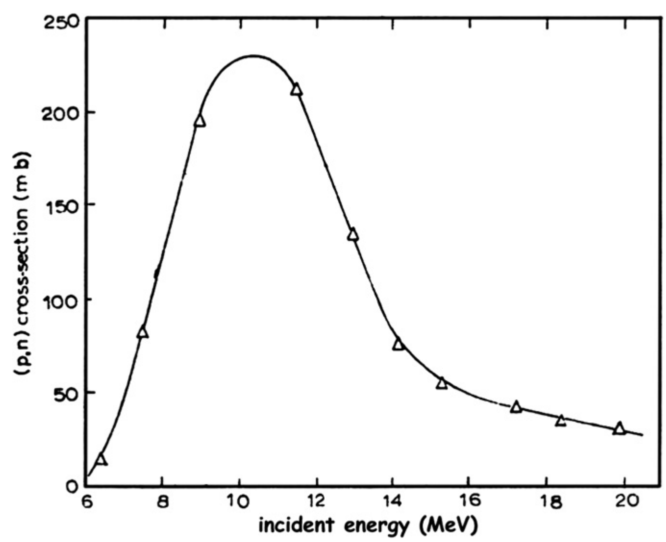

Fig. 2. Cross section of the reaction ^{130}Te(p,n)^{131}I as a function of proton incident energy [11].

To verify this prediction we performed some measurements using the proton beam provided by the 14 MV Tandem accelerator at LNS in Catania. In our experiment, we simulated the bronze alloy by building a sample with 10 stacks made up of Cu ($t = 100$ μm), Sn ($t = 14$ μm), Ag ($t = 10$ μm), Pb ($t = 10$ μm), Zn ($t = 10$ μm), Ni ($t = 1.5$ μm) and Fe ($t = 1.5$ μm) obtaining a total thickness $t = 1470$ μm; the weight percentages of the corresponding composition were 68% Cu, 8% Sn, 8% Ag, 9% Pb, 5% Zn and about 1% Ni and Fe. We used the proton activation analysis (PAA) [12] method to deduce the content of the elements at various depths in the sample. The above sample was irradiated by a $E_p = 19.2$ MeV, $I = 10$ nA proton beam