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N. Togia, T. Vasilopoulou, E. Filippaki, M. I. Savva, P. Georgolopoulou, T. J. Mertzimekis, Y. Basiakos, I. E. Stamatelatos

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Improved calibration for non destructive determination of manganese in metallurgical tap smelting slag using a high energy medical accelerator

N. Togia^{1,2}, T. Vasilopoulou¹, E. Fillipaki³, M.I. Savva¹, P. Georgolopoulou⁴,
T.J. Mertzimekis², Y. Basiakos³ and I.E. Stamatelatos^{1*}

¹ *Institute of Nuclear and Radiological Sciences, Energy, Technology & Safety, NCSR "Demokritos"*

² *Department of Physics, National and Kapodistrian University of Athens*

³ *Laboratory of Archaeometry, Institute of Nanoscience and Nanotechnology, NCSR "Demokritos"*

⁴ *Department of Medical Physics, General Anticancer-Oncologic Hospital of Athens 'Saint Savvas'*

Abstract An improved calibration method for the non-destructive determination of the manganese content in large metallurgical tap smelting slag samples using photon activation analysis is presented. A metallurgical tap smelting slag of archaeological interest from the Psaphaki site, Laconia, was irradiated under the mixed photon and parasitic neutron field produced by an 18 MV medical accelerator. The sample was 236 g in mass. Manganese was determined by the photon and neutron induced reactions $^{55}\text{Mn}(\gamma, n)^{54}\text{Mn}$ and $^{55}\text{Mn}(n, \gamma)^{56}\text{Mn}$, measuring the 834.8 keV ^{54}Mn and 846.8 keV ^{56}Mn photopeaks, respectively. Gamma spectra from the large sample and calibration foils were acquired using a HPGe detector of 85% relative efficiency. The Efficiency Transfer method was applied to determine the Full Energy Peak Efficiency (FEPE) of the large sample. The results of the analysis showed that the manganese content in the slag was $(39.5 \pm 15.8) \% \text{ w/w}$ and $(39.7 \pm 2.6) \% \text{ w/w}$ measuring the ^{56}Mn photopeak and the ^{54}Mn photopeak, respectively. Therefore, a good agreement between the photon and neutron activation analysis techniques was observed. Advantages of the technique include improved representativeness of results, elimination of reference samples, irradiation versatility and convenience, as well as reduced contamination probability since a minimal sample preparation was required for the analysis. Moreover, the introduction of the relative calibration technique removes the dependency on parameters that are usually evaluated with larger uncertainties, such as detector efficiency, increasing the quality and reliability of the procedure.

Keywords Photon activation analysis, archaeometallurgy

INTRODUCTION

The potential of large sample photon activation analysis for multi-element, non-destructive elemental analysis of metallurgical tap smelting slag using a 23 MV medical linear accelerator as photon source was studied [1]. However, the recent decommissioning of the 23 MV accelerator used in the study necessitated the need to examine whether such experiments can be performed using an accelerator of a lower nominal energy.

In the present work the feasibility to perform large sample photon activation analysis for the determination of manganese in metallurgical tap smelting slag samples of archaeological interest using an 18 MV medical accelerator was investigated. Manganese was determined by

employing both the photon and the neutron activation reactions of ^{55}Mn . Photon activation was performed using the primary bremsstrahlung beam of the accelerator, while neutron activation was performed employing the secondary neutron fluence produced mainly by (γ, n) reactions in the high Z materials in the accelerator head. A relative quantification method based on simultaneous irradiation of the sample and standard manganese foils was employed. The detector Full Energy Peak Efficiency (FEPE) for the large sample was calculated using the efficiency transfer method and a Monte Carlo model of the sample-detector configuration.

EXPERIMENTAL

The sample was provided from the archaeometry laboratory of the Institute of Nanoscience and Nanotechnology, NCSR Demokritos. It was an ancient metallurgical slag from the area of Psaphaki, Neapolis where the chief “iron-age” Laconia workings were situated. The mass of the sample was 236 ± 1 gr. The volume of the sample was 56.5 ± 0.5 cm³.

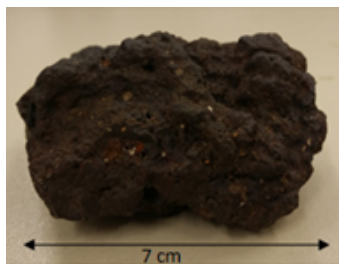


Fig. 1 Metallurgical slag sample

The sample was irradiated using an ELEKTA SYNERGY 18 MV accelerator at Saint Savvas Hospital, Athens. The irradiation was performed for a total of 30K Monitor Units. The target to sample distance was 100 cm and the beam area at the sample level was 15×15 cm². Manganese foils of high purity (> 99.99 %) were used as standards for the analysis. The foils were in the form of disks with thickness of 0.5 mm and diameter of 18 mm. During irradiation the sample was exposed to the parasitic neutron field due to (γ, n) reactions in the accelerator head materials [2], resulting in neutron induced reactions with the sample nuclei. The nuclear reactions employed in the analysis are shown in Table 1.

Gamma measurements were performed using a germanium detector based spectrometry system of 85% relative efficiency and FWHM of 1.82 keV at 1332.5 keV. This system consists of a shielded, coaxial germanium detector (EG & G ORTEC), a digital signal and data acquisition system (DSPECTM) and a support bracket for sample positioning during measurement. Spectrum analysis was performed using the Gamma-VisionTM software. The detector was calibrated with respect to energy and full energy peak efficiency using a set of standard point sources.

Quantification was performed using the relative calibration technique [3] using manganese foils as standards (Eq. 1).

$$\frac{m_{sample}}{m_{foil}} = \frac{A_{0,sample}}{A_{0,foil}} = \frac{N_{sample}}{N_{foil}} \frac{t_{c,foil}}{t_{c,sample}} \frac{\varepsilon_{\gamma,foil}}{\varepsilon_{\gamma,sample}} \frac{e^{-\lambda t_{cool,foil}}}{e^{-\lambda t_{cool,sample}}} \quad (\text{Eq. 1})$$

where, A_0 is the activity at the end of irradiation, N is the number of net counts at the photo-peak, t_{cool} is the cooling time, t_c is the counting time, ε_γ is the FEPE at the photon energy of interest for the large sample and the foil, respectively.

SIMULATIONS

FEPE for the voluminous sample was evaluated using the efficiency transfer method. FEPE was measured for a reference point source and Monte Carlo techniques were applied for the estimation of the FEPE for the voluminous sample at the corresponding gamma ray energies. Simulations were performed using the MCNP6 code [4]. The detector model was developed by modifying a validated model of the germanium detector [5]. Pulse height tally was used to predict the detector's response in terms of energy deposited in the active volume of the crystal in the specified energy bin and to estimate FEPE of the detector. The experimentally determined FEPE for the point source and the MCNP calculated efficiency transfer factor between the point and the volume source as a function of photon energy is given in Fig. 2. This data enables the evaluation of the detector efficiency for the slag sample at the photon energy of interest.

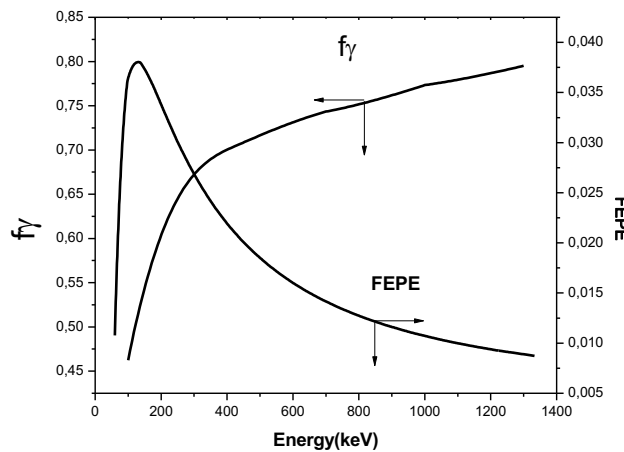


Fig. 2 FEPE for point source and MCNP derived large sample to point efficiency transfer factor (f_γ) as a function of photon energy for the counting geometry

RESULTS AND DISCUSSION

The results of the analysis are summarized in Table 1. Although the energy of the photon beam was significantly lower than the optimum for photon activation analysis, manganese concentration could still be determined by the (γ, n) reaction. Moreover, a good agreement

between the results of the photo-activation analysis technique using the primary accelerator beam and the neutron capture reaction using the secondary neutron fluence was observed. It is noted that the uncertainties in the determination of Mn using the photon activation technique were high, of the order of 40%. Nevertheless, the uncertainty in the photo-activation technique could be significantly improved if higher activation fluence was employed, for example by reducing the source to sample distance, which was 100 cm in the present experiment.

Table 1 Nuclear data used and determined-Mn mass (m) and concentration (C)

Nuclear reaction	Product half-life	Photon energy (keV)	Yield (%)	m _{Mn} (gr)	C _{Mn} (% w/w)
$^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$	2.58 h	846.8	98.9	93.8 ± 6.1	39.7 ± 2.6
$^{55}\text{Mn}(\gamma,n)^{54}\text{Mn}$	312.3 d	834.8	100.0	93.2 ± 37.2	39.5 ± 15.8

The advantages of the technique include improved representativeness of results, since the analysis of larger mass samples is facilitated, irradiation versatility and convenience due to the design and wide availability of the radiation source, as well as reduced sample contamination probability since a minimal sample preparation is required for the analysis. Moreover, the introduction of the relative calibration technique removes the dependency on parameters that are usually evaluated with larger uncertainties, such as detector efficiency, increasing the quality and reliability of the procedure.

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