Photon activation analysis of metallurgical tap smelting slag using a high energy medical accelerator: Further studies

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Activation analysis of a metallurgical tap smelting slag sample using a high energy medical accelerator: Further studies

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Abstract   The potential of large sample activation analysis for non-destructive determination of manganese in a metallurgical tap smelting slag sample of a mass of 236 g, as a whole, using an 18 MV medical linear accelerator was investigated. Manganese concentration was determined by employing both the photon and the neutron activation reaction in ^{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 in the accelerator head. The induced activity in the sample was determined using a germanium detector based gamma spectrometry system. A relative quantification method was applied based on the simultaneous irradiation of the sample and standard manganese foils. Full Energy Peak Efficiency as a function of photon energy was calculated using the efficiency transfer method and a Monte Carlo model of the sample and detector configuration. The results of the analysis showed that the manganese concentration of the sample was 39.5 % w/w. A good agreement between the applied photon and neutron activation analysis methods was observed.

Keywords   photon activation analysis, metallurgical tap smelting slag, medical accelerator

INTRODUCTION

The potential of large sample photon activation analysis for nondestructive multi-elemental analysis of a metallurgical tap smelting slag using a 23 MV medical linear accelerator as a photon source has been presented [1]. In the current work, the feasibility of activation analysis for non-destructive determination of manganese in a metallurgical tap smelting slag sample using an 18 MV medical linear accelerator, is investigated. Manganese was determined by employing both the photon and the neutron activation reactions in ^{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 foils was employed. The detector Full Energy Peak Efficiency (FEPE) for the slag 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, National Centre for Scientific Research Demokritos (Fig. 1). It was an ancient metallurgical slag obtained 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 measured with the water displacement method due to its irregular shape and found to be 56.5 ± 0.5 cm^3.
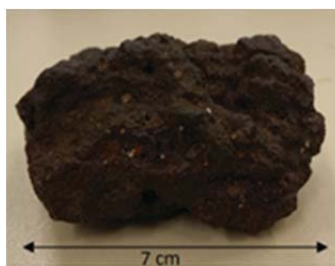
Fig. 1. Iron-age metallurgical slag sample

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, given in a total time of 90 min. The target to sample distance was 100 cm and the beam area at the sample level was 15×15 cm^2. 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. The foils and the large sample were both included in the beam and were irradiated simultaneously.

The nuclear reactions used for analysis and their characteristics are shown in Table 1. The linear accelerators used in radiotherapy with bremsstrahlung energy over 10 MeV generate undesired neutron contamination in the therapeutic beam and the treatment room. Therefore, during irradiation the sample was also exposed to a secondary neutron field [2], resulting in neutron induced reactions with the sample nuclei. The ^{55}Mn photo-activation cross section and neutron capture cross-section were taken from References [4], [5], respectively. The threshold energy for the (γ,n) reaction is 10.23 MeV.

Measurements of the emitted gamma rays were performed using a germanium detector based spectrometry system of 85% relative efficiency and FWHM of 1.82 keV at 1.332.5 keV. This system consists of a shielded, coaxial germanium detector (EG & G ORTEC), a digital signal acquisition and data acquisition system (DSPEC™) and a support bracket for sample positioning during measurement. Spectrum analysis was performed using the Gamma-Vision™ 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).

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

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 on the basis of the FEPE measured for a reference point source and Monte Carlo estimation of the FEPE for the voluminous sample at the corresponding gamma ray energy. Simulations were performed using the MCNP6 code. The detector model was developed by modifying a validated model of the germanium detector [6]. 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 sources as a function of photon energy is given in Fig. 2. The data shown in Fig. 3 enable evaluation of the detector efficiency for the slag sample at the photon energy of interest.
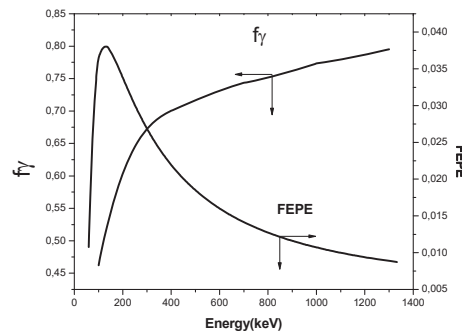
Fig. 2. FEPE for point source and MCNP derived large sample to point efficiency transfer factor (f_γ) with photon energy for the counting geometry.

RESULTS AND DISCUSSION

The results of the analysis are shown 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, very 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. Nevertheless, the uncertainties in the photo-activation technique could be significantly improved if higher activation fluence was employed. This can be achieved by reducing the

source to sample distance, which in the present experiment was 100 cm.

Advantages of the technique are the improved representativeness of results, as 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.

Nuclear reaction	Product half-life	Photon energy (keV)	Yield (%)	Detector efficiency ($\times 10^{-3}$)	m_{Mn} (gr)	C_{Mn} (% w/w)
$^{55}Mn(n,\gamma)^{56}Mn$	2.58 h	846.8	98.9	9.7	93.8 $\pm$ 6.1	39.7 $\pm$ 2.6
$^{55}Mn(\gamma,n)^{54}Mn$	312.3 d	834.8	100.0	9.8	93.2 $\pm$ 37.2	39.5 $\pm$ 15.8

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

Acknowledgements

References

[1] I. E. Stamatelatos et al., J.Radioanal.Nucl.Chem. 309, p. 165 (2016)

[2] H.R. Vega-Carrillo et al., App. Radiat. Isotop. 71, p. 75 (2012)

[3] C. Segebade et al., J. Radioanal. Nucl. Chem. 312, p. 443 (2017)

[4] IAEA, Handbook on photonuclear data for applications. Cross sections and spectra, IAEA-TECDOC-1178, Vienna, 2000

[5] BNL, Nuclear National Data Center, Evaluated Nuclear Data File (ENDF/B-VII), https://doi.org/10.1016/j.nds.2011.11.002

[6] W.G. McKinney et al., MCNPX overview, Proceedings of the 2006 HSSW, FNAL, IL, LA-UR-06-6206 (2006)

[7] T. Vasilopoulou et al., J. Radioanal. Nucl. Chem. 303, p. 853 (2015)