Nuclear Instruments and Methods in Physics Research A 622 (2010) 460–463

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research A

journal homepage: www.elsevier.com/locate/nima

Large sample NAA work at BARC: Methodology and applications

R. Acharya [a],*, K.K. Swain [b], K. Sudarshan [a], R. Tripathi [a], P.K. Pujari [a], A.V.R. Reddy [b]

[a] *Radiochemistry Division, Bhabha Atomic Research Centre, Trombay, Mumbai 400 085, India*

[b] *Analytical Chemistry Division, Bhabha Atomic Research Centre, Trombay, Mumbai 400 085, India*

ARTICLE INFO

Available online 17 February 2010

Keywords:
Large sample NAA
Internal monostandard method
In situ detection efficiency
Non-standard geometry
Pottery
Uranium ore
Stainless steel

ABSTRACT

Large sample neutron activation analysis (LSNAA) was carried out using thermal column facility of Apsara reactor at Bhabha Atomic Research Centre, Mumbai, India. The k_0-based internal monostandard NAA (IM-NAA) using in situ detection efficiency was used to analyze large and non-standard geometry samples of clay pottery, uranium ore and stainless steel. Elemental concentration ratios with respect to Na as a monostandard were used in the study of pottery and ore samples. For stainless steel sample of SS 304L, the absolute concentrations were calculated from concentration ratios by mass balance approach since all the major elements (Fe, Cr, Ni and Mn) were amenable to NAA. Applications of LSNAA in the above-mentioned three different areas are described in this paper.

© 2010 Elsevier B.V. All rights reserved.

1. Introduction

Large sample analysis is often advantageous for obtaining better analytical representativeness instead of replicate sub-sample analysis. It is more so for the samples that are not so homogeneous at lower level [1,2]. Analysis of large sample is feasible by neutron activation analysis (NAA). However, it is necessary to take care of the neutron flux perturbation, if any during neutron irradiation, and γ-ray attenuation in the large samples during measurements. The k_0 method of NAA has been used to analyze large size samples, where elaborate procedures for accounting the neutron self-absorption/flux perturbation during sample irradiation in a reactor and γ-ray self-attenuation for determining the efficiency of the detector have been addressed [3,4]. Detection efficiency calibration is carried out using effective solid angle concept and γ-ray transmission methods using standard multi γ-ray sources.

Simpler methods for the analysis of large samples under varying geometrical conditions would greatly enhance the applicability of NAA for variety of samples like high-purity alloys and metals, archeological objects and samples of biological, geological and environmental origin. A k_0-based internal monostandard method for determination of elements in samples of non-standard geometry by prompt gamma ray neutron activation analysis (PGNAA) was proposed by Sueki et al. [5]. They have utilized γ-rays from the monostandard for the calibration of relative detection efficiency. A k_0-based internal monostandard NAA (IM-NAA) using in situ relative detection efficiency [2,6], developed in our lab, is useful to analyze large and non-standard geometry samples. In this method, an element present in the sample was used as the monostandard, which takes care of neutron flux perturbation, if any, inside the sample. Usually, a major or a minor element present in the sample is chosen as a monostandard, as a better homogeneous distribution of this element in the sample is expected compared to the trace element distribution. However, a trace element present in the sample can also be chosen as a monostandard if any of its isotopes has suitable nuclear properties like high isotopic abundance and (n,γ) cross-section, with favorable half-life and high γ-ray abundance for its activation product.

The in situ relative detection efficiency was obtained using γ-rays of two or more radionuclides produced in neutron activation that have γ-rays covering the energy range of interest. The method was validated using IAEA reference materials in the mass range from 50 mg to 5 g [6] as well as by determining added impurities in 0.5 kg silica and 0.5 L water [2]. The method was applied for the compositional analysis of irregular shape and size samples of nuclear fuel cladding materials [2,7–9] namely Zircaloys 2 and 4, stainless steels (SS 316M and D9 alloy) and 1S aluminium. In the cases of zircaloy and stainless steel, composition analysis was carried out by standard-less approach since all major and/or minor elements were amenable to NAA. The IM-NAA method was applied to wheat grains in the mass range from 50 mg to 1 kg to arrive at the representative sample size. It was found from elemental concentration ratios with respect to Na that the mass ratios are constant for samples having mass 1 g and above indicating that 1 g is the minimum representative sample size [9]. This method was also applied to small and large size powdered coal samples, where the representative sample size was determined as 1 g [10].

---

* Corresponding author. Tel.: +91 22 2559 4089; fax: +91 22 2550 5151.
E-mail address: racharya@barc.gov.in (R. Acharya).

0168-9002/$ - see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.nima.2010.02.056

In the present work, applicability of IM-NAA to large and non-standard geometry samples of ancient and new clay pottery, uranium ore and stainless steel (SS 304L) has been explored. Elemental concentration ratios with respect to Na as a mono-standard were used for the pottery and uranium ore samples. In the case of SS 304L, Fe was used as the internal monostandard and its analysis was carried out by a mass balance approach.

2. Experimental

Samples of three ancient potteries and one new pottery (mass range 15–25 g), two different uranium ores (about 55 g) and one SS 304L (21 g) were irradiated in the thermal column of Apsara reactor for 7 h. For comparison of LSNAA results of one of the uranium ores, four small size homogenized samples (100 mg) were also irradiated in E8 position of Apsara reactor. The corresponding thermal neutron fluxes are about 10^8 and $10^{12}\,\mathrm{cm^{-2}\,s^{-1}}$. For neutron flux characterization in thermal column (TC), indium (10 mg) was irradiated with and without cadmium cover (0.8 mm thick) for 6 h at the nearest position to the core. Samples were counted using a 40% relative efficiency HPGe detector coupled to an 8k channel analyzer that has spectrum analysis software PHAST [11].

The ratio of mass (m) of an element (x) to mass of the internal monostandard element (y) in the sample by the k_0-based IM-INAA method is given by the following expression [2]:

$$\frac{m_x}{m_y}=\frac{((SDC)(f+Q_0(\alpha)))_y}{((SDC)(f+Q_0(\alpha)))_x}\frac{P_{Ax}}{P_{Ay}}\frac{(\varepsilon_\gamma)_y}{(\varepsilon_\gamma)_x}\frac{k_{0,Au}(y)}{k_{0,Au}(x)} \qquad (1)$$

where P_A is the net peak area under the gamma peak of interest, S the saturation factor, D the decay factor, C the counting factor used for correcting the decay during counting period, f the subcadmium to epithermal neutron flux ratio, α the epithermal neutron flux shape factor, $Q_0(\alpha)$ the ratio of the resonance integral (I_0)-to-thermal neutron cross-section (σ_0) corrected for α, the $k_{0,Au}$ the literature $k_{0,Au}$-factors [12] and ε the in situ relative detection efficiency in the present case [2,6]. The in situ relative efficiency is obtained by the following expression:

$$\ln\varepsilon_\gamma=k_j+\sum_{i=0}^{m}a_i(\ln E_\gamma)^i \qquad (2)$$

where a_is are the coefficients of the polynomial of order m and k_j is a constant characteristic of the jth nuclide. In the calculations a second-order polynomial (m=2) was used. As f-value is of the order of 10^3 in a highly thermalized irradiation position like in thermal column, Eq. (1) is simplified to Eq. (3) as given below.

$$\frac{m_x}{m_y}=\frac{(SDC)_y}{(SDC)_x}\frac{P_{Ax}}{P_{Ay}}\frac{\varepsilon_y}{\varepsilon_x}\frac{k_{0,Au}(y)}{k_{0,Au}(x)} \qquad (3)$$

For the samples in which all the major and the minor elements are amenable to NAA, the absolute concentrations can be determined using a mass balance procedure. This procedure is used in those cases where the major and minor elements account for nearly 100% of the sample mass. The sum of relative elemental concentration ratios with respect to an internal monostandard (y) can be written as

$$\sum_{i=1}^{n}m_i/m_y=W/m_y \qquad (4)$$

where $\sum_{i=1}^{n}m_i=W$, where W is the mass of the sample. Finally, the wt.% of the element is obtained by

$$m_i(\%)=\frac{m_i/m_y}{W/m_y}\times 100 \qquad (5)$$

Details of calculations by IM-NAA method using mass balance procedure can be found in our earlier publications [8,9].

3. Results and discussion

3.1. Characterization of irradiation sites

The f-value of thermal column was found to be $(6.0\pm0.4)\times10^3$ ($>99.9\%$ thermal neutron component) and the corresponding thermal equivalent neutron flux is $1.2\times10^8\,\mathrm{cm^{-2}\,s^{-1}}$ [10]. The cadmium ratio method using multi-monitors (Au, Zr, Zn, Mo and In) and Au was used for determination of α and f in E8 position of Apsara reactor and the corresponding values are 0.035 ± 0.003 and 50.0 ± 1.5 [13], respectively.

3.2. Application of IM-NAA to potteries

Chemical analysis is an important tool for provenance study of archaeological artifacts like pottery, brick and stone. In the present work, samples of three ancient potteries and a new pottery were analyzed for provenance study using elemental concentration ratios. Concentrations of 16 elements namely Na, K, Sc, Cr, Fe, Co, Zn, Ga, As, Br, La, Ce, Sm, Eu, Yb and Th, were determined in small and large samples. A typical in situ relative detection plot for large size pottery sample (23 g) is given in Fig. 1, which covers the energy range of 122–1811 keV.

The elemental concentration ratios of 8 elements with respect to Na and La/Ce values are given in Table 1. The uncertainties on the concentration ratios are 2–7%, which are due to counting statistics only. From the La/Ce values, it was observed that these four pottery samples broadly fall into two major groups: group I (OP1, OP2 and OP3) and group II (NP1), though La/Ce value of OP3 (0.20) is slightly different from OP1 and OP2 (0.17). The elemental

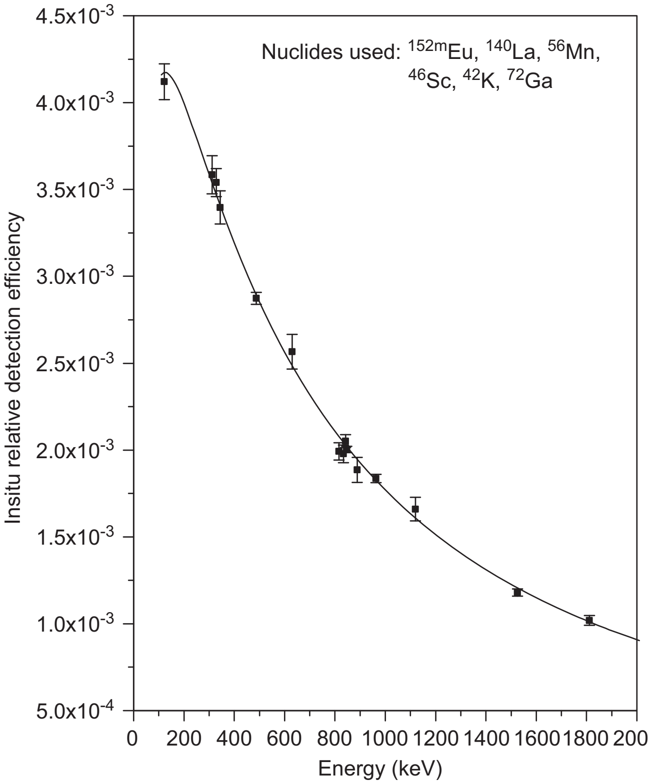

Fig. 1. In situ relative detection efficiency for large size pottery sample irradiated using thermal column of Apsara reactor.

Download English Version:

https://daneshyari.com/en/article/1826972

Download Persian Version:

https://daneshyari.com/article/1826972

Daneshyari.com