Nuclear Instruments and Methods in Physics Research A 624 (2010) 180–183

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research A

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

Geometry independent indium activation analysis

Shikha Prasad [a,*], Natallia Pinchuk [b], Stephen E. Anderson [a], Ronald F. Fleming [a]

[a] Department of Nuclear Engineering and Radiological Sciences, University of Michigan, Ann Arbor, MI 48109, USA
[b] Massachusetts Institute of Technology, Cambridge, MA 02142, USA

ARTICLE INFO

Article history:
Received 22 February 2010
Received in revised form
30 July 2010
Accepted 3 August 2010
Available online 7 August 2010

Keywords:
Geometry independent
Indium activation
Cross-section measurement
Gamma detectors
Characterizing sources
Nonproliferation

ABSTRACT

A geometry independent method has been derived for calculating the ratio of the spectrum averaged cross-sections for the reactions ^{115}In(γ, γ′)^{115m}In and ^{113}In(γ, γ′)^{113m}In in the energy region of interest 300–500 keV. The technique is unique as it eliminates the need for a detailed detector efficiency formulation for varying source–detector geometries, thereby removing a source of systematic uncertainty. The validity of the technique is demonstrated experimentally by comparing the cross-section ratios for a geometrically diverse set of source–detector configurations. The average ratio value for seven independent measurements is 0.778 ± 0.0033 with a reduced χ^2 of 1.2, which is within 1.15% of the previously published value.

1. Introduction

Measurement of an isomeric activation cross-section ratio for two reactions using gamma ray spectrometry requires knowledge of detectors efficiencies at the two gamma ray energies. For small sample sizes or "point sources" it is possible to use point like gamma calibration as shown by Hawari [4] where the relationship between the logarithm of energy and the logarithm of efficiency for detection of a gamma ray can be approximated accurately by a low-order polynomial [2,5]. The cross-section measurement of large sample sizes is a more challenging problem and is the subject of this paper. The three gamma rays utilized in the study originate from three different decay processes, thus coincidence summing is not a concern. We proceed with the assumption that the Hawari's linear relationship model discussed above for point sources is also applicable to large samples irrespective of their geometry in the energy region of interest 300–500 keV. In effect, the following study minimizes the uncertainty in cross-sectional measurement of indium isotopes by eliminating explicit modeling of the intrinsic full energy peak efficiency. The validity of the approach is tested by showing that the measured cross-section is independent of geometry. The application of this technique is not limited to only cross-section measurements but can be extended to measurements of other crucial intrinsic properties of a sample without characterizing its geometry.

2. Theory

The isotopes of interest for the method are ^{113}In and ^{115}In. The three gamma energies utilized are 392 keV from ^{113m}In and 336 keV and 497 keV from ^{115m}In isotopes.

Table 1
Gammas from ^{113m}In (1.7 h) and ^{115m}In (4.5 h) [1].

$E\gamma$ (keV)	$I\gamma$ (%)	Decay mode	Isotope
391.690	64.2	IT	^{113m}In
336.240	45.83	IT	^{115m}In
497.358	0.047	β^-	^{115m}In

To demonstrate the concept the ratio of the spectrum averaged isomeric activation cross-sections $\langle\sigma\rangle_{113}/\langle\sigma\rangle_{115}$ is calculated. The net counts in each photopeak are used to calculate activity, A_0 as shown below [4].

$$A_0 = \frac{\lambda N e^{(\lambda t_1)}}{(1-e^{(-\lambda \Delta)})(1-e^{(-\lambda \tau)})} \tag{1a}$$

$$= \varepsilon \Gamma \frac{\alpha m N_A}{A} \langle\sigma\rangle \Phi \tag{1b}$$

ε is the detector efficiency, Γ the number of gamma rays of energy E per decay, α the isotopic abundance, m the mass of the sample, N_A the Avogadro's number, $\langle\sigma\rangle$ the spectrum averaged isomeric activation cross-section, Φ the photon flux, A the atomic weight,

* Corresponding author. Tel.: +1 516 225 7870.
E-mail address: shikhap@umich.edu (S. Prasad).

0168-9002/$ - see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.nima.2010.08.001

Δ the live time counting, λ the decay constant, τ the irradiation time, t_1 the time from end of irradiation to start of counting and N the net counts in gamma ray peak.

The ratio of natural abundances $\alpha_{115}/\alpha_{113}$ is 22.31. To obtain an expression for the ratio of the cross-sections, Eq. (1b) is evaluated using the 392 and 336 keV parameters as

$$\frac{\langle\sigma_{113}\rangle}{\langle\sigma_{115}\rangle}=\frac{\langle\tau\rangle A_{0_{392}}\varepsilon(336)\Gamma_\gamma(336)a_{115}}{\langle\tau\rangle A_{0_{336}}\varepsilon(392)\Gamma_\gamma(392)a_{113}}. \tag{2}$$

It is clear from Eq. (2) that the intrinsic full energy peak efficiency, ε is imperative to calculate the ratio, however, this parameter is a function of the gamma ray energy and is also dependent on the detector and source geometries. Therefore, the use of this parameter introduces another element of uncertainty. The purpose of the method discussed is to eliminate the need of explicitly modeling detector efficiencies for each source–detector geometry in the measurement of ^{113}In to ^{115}In activation cross-section ratio. This elimination would reduce the uncertainty introduced in the measurement: total statistical uncertainty would only be due to the uncertainty in counts.

To eliminate dependence on detector efficiencies in Eq. (2), the 497 keV gamma ray from ^{115m}In as described in Table 1 is utilized. For the ease of calculation and physical representation, the numerator and denominator are multiplied by τ to make τA_0 a dimensionless quantity which will be referred to as N_0. Here, A_0 is calculated using Eq. (1a). We can deduce that a generalized form of Eq. (2) calculated using the ^{115}In 336 keV gamma ray and the ^{115}In 497 keV gamma ray will simply be unity since both gamma rays come from the activation of ^{115}In and thus have the same microscopic cross-section and natural abundance. Thus, the following the relationship can be derived:

$$\frac{\varepsilon_{336}}{\varepsilon_{497}}=\frac{\Gamma_{497}N_0(336)}{\Gamma_{336}N_0(497)}. \tag{3}$$

Furthermore, from Knoll [2] and other sources [5] a power relationship between energy of the gamma ray and the detector intrinsic full energy peak efficiency is utilized. In the energy region of interest 300–500 keV, this relationship can be defined by a linear relationship between the natural logarithm of the two quantities the slope of which is negative. Equating the slopes we have

$$\frac{\ln\varepsilon_{336}-\ln\varepsilon_{497}}{\ln E_{336}-\ln E_{497}}=\frac{\ln\varepsilon_{336}-\ln\varepsilon_{392}}{\ln E_{336}-\ln E_{392}}. \tag{4}$$

Let $K=\ln(E_{336}/E_{392})/\ln(E_{336}/E_{497})$ which is 0.38996. Then, using Eqs. (3) and (4)

$$\frac{\varepsilon_{336}}{\varepsilon_{392}}=\left(\frac{\varepsilon_{336}}{\varepsilon_{497}}\right)^K=\left(\frac{\Gamma_{497}}{\Gamma_{336}}\right)^K\left(\frac{N_0(336)}{N_0(497)}\right)^K. \tag{5}$$

Substituting Eq. (5) and K in Eq. (2), $R_2\equiv\langle\sigma\rangle_{113}/\langle\sigma\rangle_{115}$ can be calculated as

$$R=\left(\frac{\Gamma_{497}}{\Gamma_{336}}\right)^K\left(\frac{\Gamma_{336}a_{115}}{\Gamma_{392}a_{113}}\right)\frac{N_0(392)}{N_0^K(497)N_0^{1-K}(336)}. \tag{6}$$

Propagating the uncertainty assuming Poisson statistics the uncertainty can be calculated by

$$\left(\frac{\sigma_R}{R}\right)^2=\left(\frac{\sigma_N}{N}\right)^2_{392}+K^2\left(\frac{\sigma_N}{N}\right)^2_{497}+(1-K)^2\left(\frac{\sigma_N}{N}\right)^2_{336}. \tag{7}$$

Following the above method a general conclusion in the energy region of interest 300 to 500 keV (or any other energy region where linear relationship between logarithm of full energy peak efficiency and logarithm of energy can be safely assumed) can be made: to calculate a full energy peak efficiency independent ratio of fundamental properties of two different isotopes (as described in Eq. (1)), a third gamma ray from one of the isotopes could be utilized. The above derivation is for a simple case with no cascade effects, for which a general formula for the ratios of the spectrum averaged cross-sections for two different isotopes A and B, with two characteristic gamma ray energies $E1$ and $E2$, respectively, and a third gamma ray energy $E3$ from isotope A is shown below:

$$R=\left(\frac{\Gamma_{E3}}{\Gamma_{E1}}\right)^K\left(\frac{\Gamma_{E1}a_A}{\Gamma_{E2}a_B}\right)\frac{N_0(E2)}{N_0^K(E3)N_0^{1-K}(E1)} \tag{8a}$$

where the general form of Eq. (5) as shown below is used to replace the intrinsic full energy peak efficiency values with measurable quantities N_0 which is τA_0 where A_0 is the activity for decay gamma ray

$$\frac{\varepsilon_{E1}}{\varepsilon_{E2}}=\left(\frac{\varepsilon_{E1}}{\varepsilon_{E3}}\right)^K=\left(\frac{\Gamma_{E3}}{\Gamma_{E1}}\right)^K\left(\frac{N_0(E1)}{N_0(E3)}\right)^K. \tag{8b}$$

3. Procedure and Data

For the purpose of experimental demonstration, ^{113}In and ^{115}In are required to be in their isomeric states. The ^{60}Co source at the Phoenix Laboratory, University of Michigan was used for this purpose. Samples of natural indium with varying geometries were irradiated using this 6.285 kCi ^{60}Co source. The isomeric activation of the two indium isotopes ^{113}In and ^{115}In is very weak with the ^{60}Co gamma rays, nearly 1 kg of sample had to be used in the experiments to obtain sufficient counts for good statistics. This is because there is a small but plausible probability that after Compton scattering the ^{60}Co gammas will have just the required energies to excite ^{113}In and ^{115}In to their isomeric states of 391.69 keV and 336.24 keV, respectively, above ground level [1].

The ^{60}Co source at the Phoenix Memorial Laboratory is stored in a 5.0 m (16.5 ft) deep pool of water, at a depth of 3.5 m (11.5 ft). The source consists of 9 stainless steel rods containing each 4–5 pellets of ^{60}Co. The rods are 1.25 cm in diameter and 30 cm in length. They are positioned in a 12.5 cm diameter cylindrical holder located on a platform. The indium targets were placed in the center well, which is located on the top cover, at the same axis as the source. The center well is cylindrical, with an inner diameter of 8 cm and a height of 33 cm. There were three unique indium targets and combination of such irradiated: indium raindrops with the total mass of 250.15 g, indium raindrops with the total mass of 269.1 g, and indium ingot with the total mass of 51.05 g. The indium raindrops were put in a pancake geometry container and a spherical container. The irradiation target shapes

Table 2
Irradiation, Decay and Counting times.

No.	Target shape	τ (s)	t_1 (s)	Δ (s)
1	In-bottle	95,400	329.76	72,000
2	In-pancake	114,300	303.98	72,000
3	In-bottle	130,320	1554.00	72,000
4	In-bottle+ingot	113,760	259.70	72,000
5	In-bottle+ingot	114,300	469.00	72,507
6	In pancake	113,760	498.00	91,680
7	In sphere	84,600	317.83	78,000