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Measurement of Average Thermal Neutron Flux for PGNAA Setup

Dalpat Meena[1], S. K. Gupta[1*], H. S. Palsania[1], Narendra Jakhar[1], Naresh Chejara[1], Pushpa Meena[2]

[1]Department of Physics, University of Rajasthan, Jaipur, India

[2]Department of Zoology, University of Rajasthan, Jaipur, India

ABSTRACT

The prompt gamma neutron activation analysis (PGNAA) is widely used for detection and quantification of elements in samples of interest such as - archaeological, biological, medical, environmental samples, etc. The detection limit of the PGNAA setup for an element depends upon the neutron flux available at sample. The aim of this study is two-fold. First, to determine the sensitivity of setup for different sample configuration and second, to determine the average neutron thermal flux at the sample of the PGNAA setup comprising a 5Ci Am-Be neutron source. The thermal neutron flux within water howitzer at different heights was measured using the gold foil activation technique with and without cadmium covers. The experimental results indicate that the sensitivity of the setup is higher for the aqueous form of the sample compared to the solid form of the sample. In 5L water howitzer of the setup, thermal and epi-thermal neutron fluxes gradually decrease with distances from the source. The measured average thermal neutron flux within 5 liter water howitzer is 3.8E+5 n/cm2 s.

Keywords: Thermal neutron flux, epithermal neutron flux, Isotopic Am-Be source, Foil activation method, Sensitivity

I. INTRODUCTION

In PGNAA technique, energy and intensity of prompt gamma emitted by the elements upon capture of either thermal neutron or an epithermal neutron are measured. The intensity of emitted prompt gamma highly depends upon the neutron flux [1,2]. PGNAA is widely used technique for elemental analysis using reactor due to availability of high thermal neutron flux. High cost and non-transportability limits the applications of reactor based PGNAA setup [3].

Nowadays, many configurations of radioisotope based PGNAA setups are reported in literature. The radio isotopic neutron sources are compact, transportable and relatively economical and have longer half-life. Radio isotropic neutron source based PGNAA setups are useful for in-situ and online multi-elemental analysis in a wide variety of applications. The radio-isotopic Am-Be neutron source is used in many research fields. The main advantage of this source is high stability of the produced fluxes.

Thermal neutron flux at sample volume is an important parameter for achieving lower detection limit for detection and quantification of elements in the sample of interest. The average thermal neutron flux at sample volume were measured using thin gold foils with and without cadmium cover using the monitor reaction ^{197}Au (n, γ) ^{198}Au.

II. METHODS AND MATERIALS

Due to electrical neutrality of neutrons, they are difficult to detect. When thin pure foils of elements are exposed to neutrons, the compound nucleus of elements is formed which subsequently decay and produce gamma rays. Number of gamma rays emitted is proportional to the neutron flux. The foil activation technique is the most convenient method for the measurement of neutron flux of nuclear reactors and radio-isotopic neutron source [4].

A. *Gold Monitoring Reaction*

Gold has one stable isotope, ^{197}Au and is considered the heaviest mono-isotopic element. 100 % abundance of $^{197}_{79}Au$ makes it most suitable candidate for neutron flux measurement using the foil activation method. The thermal neutron capture cross-section is high 98.7 barn. When a thermal neutron is captured by the ^{197}Au nucleus then compound nucleus $^{198}_{79}Au$ is formed. $^{198}_{79}Au$ nucleus is a β^- emitter with half-life 2.69517 days. $^{198}_{79}Au$ compound nucleus energy level diagram and decay schemes are shown in Fig. (1). During decay three gammas having energy 1087.7keV, 675.9keV and 411.8keV are emitted. Among them, the most intense 411.8keV gamma ray emitted through 98.99 % emission probability [5].

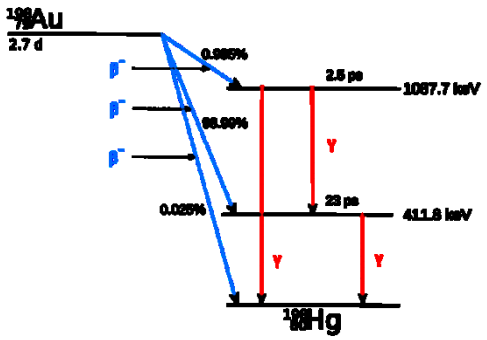

Figure 1: $^{198}_{79}Au$ energy level diagram and decay process [6]

B. Neutron flux by cadmium separation method

The conventional method for the measurement of the thermal flux using the foil activation technique is the cadmium separation method also called the cadmium difference. This involves irradiating a bare detector foil to obtain its activity and covering a similar foil with cadmium covers of thickness between 0.5-1mm and of 1.0cm diameter and irradiating it under the same experimental conditions to obtain its activity and then obtaining the thermal neutron flux from the two measurements. This means that any flux determination involves two measurements. The conventional method also assumes that the neutron flux is stable and remains constant during both measurements which might not be so in practice.

The specific count rate for bare gold foil (without cadmium covered) $A_{sp(bare)}$ *is* [7-9]-

$$A_{sp(bare)} = \frac{C_{net}\ /t_m}{S.D.C.w}$$

The specific count rate for cadmium covered gold foil $A_{sp(cd)}$ *is*:

$$A_{sp(cd)} = \frac{C_{net}\ /t_m}{S.D.C.w}$$

Where,

C_{net}= net (count) area of gamma peak of interest,

$S = \left(1 - e^{-\lambda t_i}\right)$

= saturation coefficient corrected to irradiation time (t_i),

$\lambda = \frac{ln2}{T_{1/2}}$; $T_{1/2}$ is half life,

$D = e^{-\lambda t_d}$

= delayed coefficient corrected to delayed time (t_d),

$C = \left(1 - e^{-\lambda t_m}\right)/\lambda t_m$

= measuring coefficient corrected to measurement time (t_m),

w = mass of gold foil in gram

Epi-thermal neutron flux will be -

$$\Phi_{epi} = \frac{A_{sp(Cd)}}{G_{epi}.I_0(\alpha).N_A.\theta.I_\gamma.\varepsilon_P/M}$$

Here;

ε_P = Absolute efficiency of detector at E_γ = 4.5x10^{-4}

I_γ = Intensity of gamma ray (411.8 keV) =0.95,

θ = Abundance of ^{197}Au isotope =1,

M = Atomic weight of ^{197}Au isotope,

N_A =Avogadro no= 6.023 x 10^{23} atom per mole

And, Thermal flux will be-

$$\Phi_{th} = \frac{G_{epi}}{G_{th}}\left(\frac{A_{sp(Bare)}}{A_{sp(Cd)}} - 1\right)Q_0.\Phi_{epi}$$

$$\Phi_{th} = \frac{(A_{bare} - A_{Cd})}{t_c.G_{th}.g.\varepsilon_\gamma.\sigma_0.N_{Au}}$$

Where

G_{th} and G_{epi} are self-shielding coefficients for thermal and epi-thermal neutrons, respectively (in fact, thin foils of gold are usually used, Gth ≈ Gepi =1);

σ_0 = Thermal cross section for ^{197}Au (n, γ) ^{198}Au reaction = 98.7(98.65±0.09) barn,

$Q_0 = \frac{I_0(\alpha)}{\sigma_0}$ is resonance integral to thermal cross section ratio of the target nucleus.

$$I_0(\alpha) = \int_0^\infty \frac{\sigma(E)dE}{E^{1+\alpha}}(1\ eV)^\alpha$$

is resonant integral for distribution of epi-thermal neutron fluxes without obeying the rule of $^1/_E$; here α is spectrum coefficient expressing spectrum deviation from rule of $^1/_E$ and it has value in range [-1,1].

$I_0(\alpha) = 1550 \pm 28$ barn, $\sigma_0 = 98.65 \pm 0.09$ barn; So $Q_0 = \frac{I_0(\alpha)}{\sigma_0} = 15.71 \pm 28$.

C. Experimental arrangement for gold foil activation

The setup consists of 5Ci ^{241}Am-Be neutron source, a source tank, a plastic container and a scale mounted with gold foil. The neutron source tank is a 5mm thick steel cylinder (diameter 36cm and height 45cm) filled with paraffin. Source tank has a central cylindrical cavity of diameter 8cm. Neutron source is placed inside cylindrical cavity of the source tank. Plastic container (18 cm x 21 cm) filled with 5L de-ionized water is kept above the cylindrical cavity of source tank. Distance from the neutron source to the bottom of the plastic container is 25cm. Five gold foils (thickness = 0.05mm and area = 1.0 cm^2) were fixed on the scale at distance 0cm (bottom of container), 5cm, 10cm,15cm and 20 cm. Scale is mounted along the central axis of the cylindrical plastic container. The geometrical arrangement of gold foils in water howitzer is shown in fig. 2.

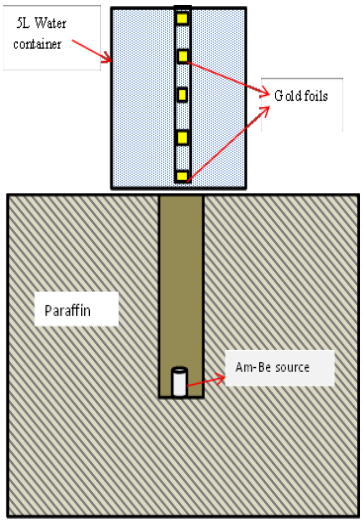

Figure 2 : Geometrical arrangement for neutron flux measurement

D. Gamma Spectroscopy Setup

The gamma spectroscopy system consists of a Canberra Coaxial HPGe semiconductor detector (GCD1518) and a Canberra digital spectrum analyzer (DSA1000) working with the acquisition software Genie 2000. The detector is operated under a high voltage of 2.5kV. The detector relative efficiency is 16.4 %, and FWHM is 1.63keV at 1332.5 keV (^{60}Co). Gamma ray spectrum of bare gold foil irradiated by neutron for two hours is shown in fig. (3).

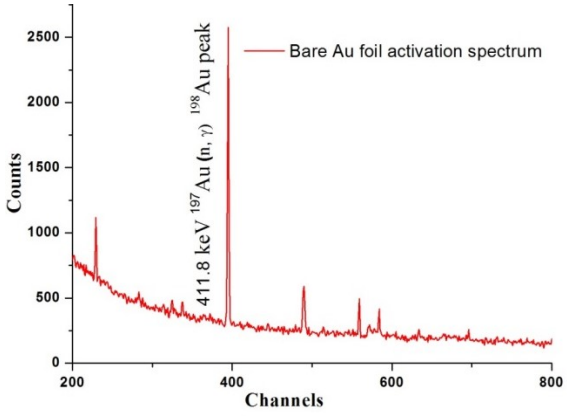

Figure 3. Au foil 2-hour gamma ray spectrum using HPGe detector

III. RESULTS AND DISCUSSION

A. Sensitivity of PGNAA setup

Sensitivity of PGNAA Setup is evaluated for three sample configurations. First, the sample is directly irradiated with a neutron beam. In a second case sample is kept in 5L water howitzer and in third configuration 5L aqueous solution of sodium chloride is irradiated with a neutron beam. 100g, 200g and 300g sodium chloride is used as sample. In each case prompt gamma spectra were recorded. ^{36}Cl prompt gamma peak (1164.84keV) for each case is shown in Fig. 4.

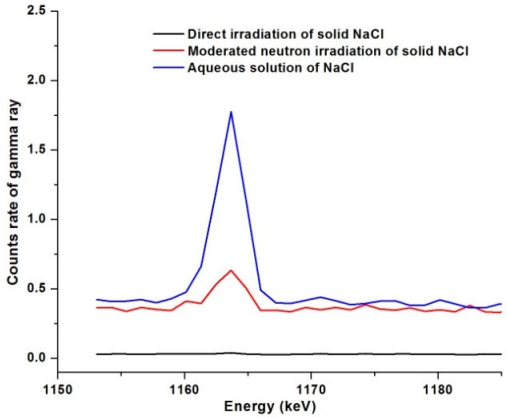

Figure 4: 1164.84 KeV ^{36}Cl prompt gamma peak in different sample configuration

The sensitivity S (counts. s^{-1}. g^{-1}) of the PGNAA setup for an element is defined as [10] -

$$S = \frac{N_A \theta \Phi \sigma_0 P(E_\gamma) \epsilon(E_\gamma)}{M}$$

Where,

S = sensitivity in counts. s^{-1}. g^{-1}

N_A=Avogadro's number

σ_0 = elemental thermal neutron absorption cross-section in cm^2

$P(E_\gamma)$ = Gamma yield, photon per neutron

θ = fractional abundance of isotope

ϕ = neutron flux in neutron.cm^{-2}. s^{-1}

$\varepsilon(E_\gamma)$ = detector photo peak efficiency at E_γ

M = atomic weight of element

Variation of sensitivity of chlorine with sodium chloride mass as shown in Fig. (5) indicates that the sensitivity of setup for detection of chlorine is maximum in case of aqueous solution. Decrease in sensitivity with mass of sodium chloride indicates increased self-absorption in the sample.

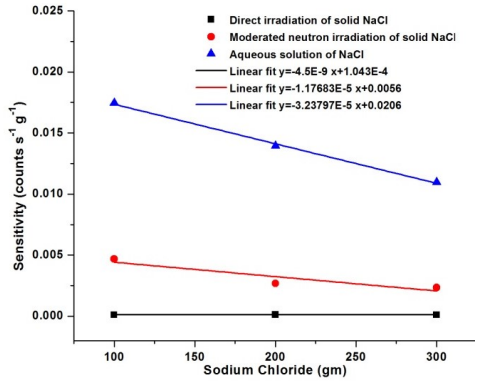

Figure 5 : Geometrical arrangement for neutron flux measurement

B. Measurement of Average Thermal Neutron Flux

Thermal neutron flux was measured for different distances from the bottom of the sample container using gold monitor reaction ^{197}Au (n, γ) with bare gold foil and gold foil covered with 0.5 mm thick cadmium foil. Gold foils were kept at different distances along central axis of sample container which is parallel to the neutron beam direction. Details of foil weight, activation time, cooling time and counting time without cadmium cover and with cadmium cover are summarized in table 1 and table 2 respectively.

Table 1 Gold foil activation detail for neutron flux measurement

Position (cm)	Weight (mg)	Activation time	Cooling time	Counting time
0	11	8 d	2 h	2 h
5	12	7 d 22 h	2 h	2 h
10	10	7 d 20 h	2 h	2 h
15	13	7 d 18 h	2 h	2 h
20	11	7 d 17 h	1 h	2 h

Table 2 Cd covered gold foil activation detail for neutron flux measurement

Position (cm)	Weight (mg)	Activation time	Cooling time	Counting time
0	11	13 d 3 h	2	2
5	12	13d 1 h	2	2
10	10	12 d 23 h	2	2
15	13	12d 21 h	2	2
20	11	12 d 19 h	1	2

Table 3 Thermal and epi-thermal neutron flux values for ^{241}Am-^{4}Be

Position (cm)	Epi-thermal Flux (n/cm^2 s^1)	Thermal Flux (n/cm^2 s^1)	Total Flux
0.5	5.79E+03	3.76 E+05	3.82E+05
5	2.45E+03	2.39 E+05	2.41E+05
10	1.65E+03	1.12E+05	1.14E+05
15	6.30E+02	4.21E+04	4.27E+04
20	3.25E+02	3.51E+03	3.84E+03

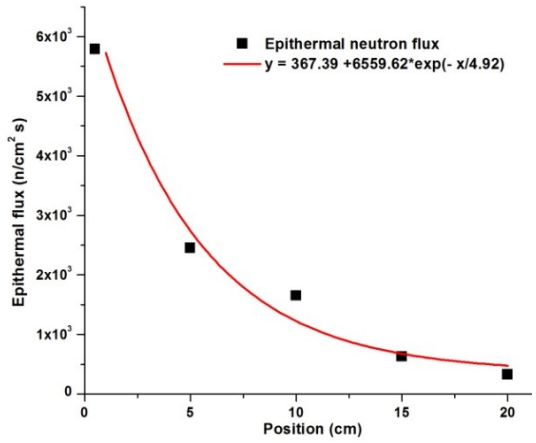

Figure 6: Distribution of epi-thermal neutron flux for ^{241}Am-Be source with position

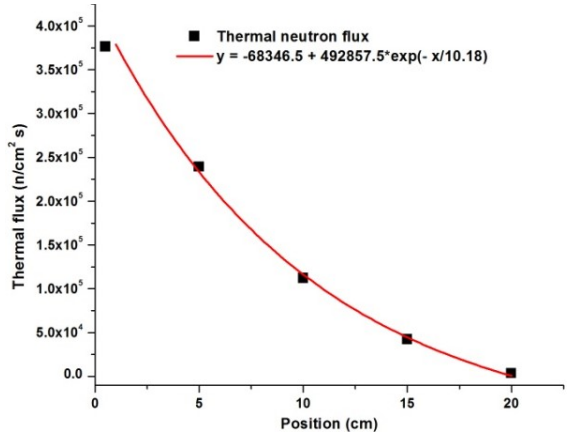

Figure 7: Distribution of thermal neutron flux for ^{241}Am-Be neutron with position

Measured and calculated thermal (ϕ_{th}), epithermal neutron (ϕ_{epi}) and total flux (ϕ_{tot}) decreases exponentially with distances from the bottom of the 5 L water howitzer as shown in Fig. (6) and (7). The results summarized in table 3 shows that neutron flux is maximum at the bottom of water howitzer. Calculated average thermal neutron flux within the 5L water howitzer is 3.8E+5 n/cm^2 s.

IV. CONCLUSION

The thermal neutron flux available at sample volume is sufficient for PGNAA studies. According to obtained results, PGNAA technique for elemental analysis comprising 5Ci Am-Be source is an efficient option when aqueous solutions of sample are used. Measured average thermal neutron flux within 5L water sample volumes is 3.8E5 n/cm^2 s.

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