# Flux calculation in LSNAA using an $^{241}$Am–Be source

**R. Khelifi,[1]* P. Bode,[2] A. Amokrane[3]**

[1] *Departement de Physique, Université Saad DAHLAB, BP:270, 09400 Blida, Algerie*
[2] *Interfaculty Reactor Institute, Mekelweg 15, 2629 JB, Delft, The Netherlands*
[3] *Faculté de Physique, BP:32 El Alia, Bab Ezzouar, Alger, Algerie*

The CITATION code based on neutron diffusion theory is used for flux calculation inside voluminous sample in prompt gamma activation analysis with an isotopic neutron source ($^{241}$Am–Be). The code used the specific parameters related to energy spectrum source, irradiation system materials (shielding, reflector, etc.), geometry and elemental composition of the sample. The flux distribution (thermal and fast) was calculated on three-dimensional geometry for the system: source, air, and polyethylene and water cylindrical sample of 125 liters. The thermal flux was calculated in series of points inside the sample, and agreed with the results obtained by measurements with good statistical uncertainty. The maximum thermal flux was measured at distance of 4.1 cm and calculated at 4.3 cm by the CITATION code. Beyond a depth of 7.2 cm, the ratio of thermal flux to fast flux increases up to twice and allows us the optimization of the detection system in the scope of in-situ PGNAA.

## Introduction

Large samples neutron activation analysis (LSNAA) has proved to have great potential of investigation for various applications in a wide range of analytical field including archeological, waste characterization geological, environmental and industrial studies, using isotopic neutron sources and high resolution gamma-ray spectroscopy.[1,2] For quantitative LSNAA, the neutron flux distribution inside the samples during irradiation must be known for an absolute elemental determination. Measurement of the neutron flux inside the sample with flux monitors became in concurrence with the wish for universal applicability. However, the use of this technique on routine basis requires standards or calibrating means of the facility system. To solve this problem, a number of calculation models have been developed to simulate the experimental setup.[3–6]

The goal of this paper is to calculate the neutrons flux in large water samples on simple irradiation setup without resorting to the long duration of experimental tests and the use of powerful codes (i.e., Monte Carlo: MCNP, etc.) with an acceptable result.

The present work describes the estimation of the yield emission ($\chi(E)$) from the $^{241}$Am–Be neutron spectra, which are used by the input file of the diffusion code CITATION.[7] In the input data file, we mentioned the $^{241}$Am–Be neutron spectra parameters, sample with known composition, so all parameters; geometry as well as neutron parameters were known. The code was used to calculate the distribution of neutrons over five energy groups where the analytical solution must be scaled by the source strength.

The paper deals also with the benchmarking of thermal flux distribution obtained by code against data measured with an irradiation assembly for PGNAA using an $^{241}$Am–Be neutron source. This model has been tested successfully with measurement of thermal flux distribution by indium foil activation technique in voluminous water sample. The knowledge of neutrons flux produced by the neutron source of Am–Be to the inside of these voluminous samples allows us to estimate the optimization of the experimental device and the volume of the sample to interrogate as well as the use of the PGNAA in routine.

## Method

*Estimation of $\chi(E)$ parameter from the $^{241}$Am–Be energy spectrum*

Recent measurements have been published which provide the spectra of the neutron source.[8] The five groups' energy used for the $^{241}$Am–Be source is shown in Table 1. The first four groups energy band are fast neutrons and the last group is set equal to zero. The neutron energy spectrum, gives the differential distribution of neutron fluency, $\chi(E)$, where $E$ is the neutron energy.

*Table 1.* Five energy groups' boundaries

| Group No. | Neutron energy spectrum, eV | Normalized |
|---|---|---|
| 1 | $(6.065–20)\cdot10^6$ | 0.2772 |
| 2 | $(2.019–6.065)\cdot10^6$ | 0.5027 |
| 3 | $(1.11–2.019)\cdot10^6$ | 0.1905 |
| 4 | $(0.5–1.111)\cdot10^6$ | 0.0036 |
| 5 | $(0–0.5)\cdot10^6$ | 0 |

* E-mail: Khelifi@iri.tudelft.nl

From the graph:

$$\chi(E) = \Delta E \cdot Norm \ (\text{MeV})^{-1}$$

where $\Delta E$ are the group parameter bins and *Norm* is the corresponding value from the spectrum.

Measured or computed neutron spectra are in most cases originally obtained as group fluencies in energy bins and a standard energy grid is used. Even bin is described by its lower boundary, $E_i$, and its upper boundary, $E_{i+1}$. Twenty five energy bins are used with $E_1 = 19.64$ MeV and $E_{26} = 0.5$ MeV.

The group fluency in the *i*th energy bin is given by:

$$\varphi_i = \int_{E_i}^{E_{i+1}} dE \ \chi(E)$$

However, from Reference 9 it is understood that the values representing neutron spectra, $\chi(E)$, in tables and figures are group fluencies divided by the lethargy interval (i.e., $\ln E_{i+1} 1 - \ln E_i$) and are normalized:

$$\phi = \sum_{i=1}^{I} \varphi_i$$

where $I$ is selected as 25 and the value of $\chi(E)$ is by definition 1/MeV.

However, in order to taking into account lower energy than 0.5 MeV, the following distribution is assumed from the graph:

$$\chi(E) = C \cdot E^{1/2} \text{ for } E \leq 0.5 \text{ MeV}$$

From the spectrum:

$$\chi(E_0) = \chi(0.5 \text{ MeV}) = 7.6$$

Then:

$$C = 0.01075 \cdot 10^{-6} \ (\text{eV})^{-1}$$

For $I = 26$ to 50:

$$\int_{E_i}^{E_{i+1}} dE \ \chi(E) = C \int_{E_i}^{E_{i+1}} E^{1/2} \, dE = 0.38$$

The results obtained for the normalized spectrum are given in Table 1.

### Experimental setup

The arrangement of the detection system for PGNAA measurement is illustrated in Fig. 1. The setup consists of a 37 Gbq (1 Ci) [241]Am–Be neutron source which produce $2.2 \cdot 10^6$ s$^{-1}$ and located inside a cubic paraffin block in irradiation position.

The sample is a 0.125 m$^3$ volume of water in polyethylene tank with 0.3 cm wall thickness and 49 cm of diameter. The water also plays a very essential role in shielding against neutrons and 4.43 MeV $\gamma$-rays energy emitted from the neutron source.

The three dimensional calculations using the code CITATION were performed for a sample consisting completely of water according to Fig. 1. The code solves the equation of multi-groups diffusion and gives flux values at desired energy and positions.

## Results and discussion

### Flux calculations

The flux distribution was calculated for five groups' energy, fast and thermal energy over the depth along the central axis of the water sample. The curves obtained are shown in Fig. 2 according the geometry: source, air, polyethylene and water. Within the source–air distance of 4.1 cm, only fast neutrons are present compared to ~100 n·cm$^{-2}$·s$^{-1}$ thermal flux obtained by fast neutron interaction inside the shielding of the source container.

Inside 0.3 cm of polyethylene and the water, the fast neutron flux decreases drastically and in the opposite thermal flux occurs and their number increase until a maximum value.

In Fig. 3, thermal flux is plotted across the width of the water at various depth ($x_0$, y, 0), so, the dimension of the sample can be optimized for best sensitivity of analysis (low background, high flux and efficiency) and by taking into account radioprotection consideration.

### Flux comparison

In order to validate calculations, thermal neutron flux versus depth along central axis of the water sample was measured by indium foil activation technique using average thermal capture cross section of 70 b for the $^{115}$In(n,$\gamma$)$^{116m}$In reaction. For this purpose 1 g of high purity indium foil was irradiated during 3 hours at various radial positions in demineralized water (Fig. 1). The induced activity was measured during 2 hours with an Ortec HPGe detector, with relative efficiency of 20% and an analyzer Model 916 of 4096-channels in a very low background counting station.
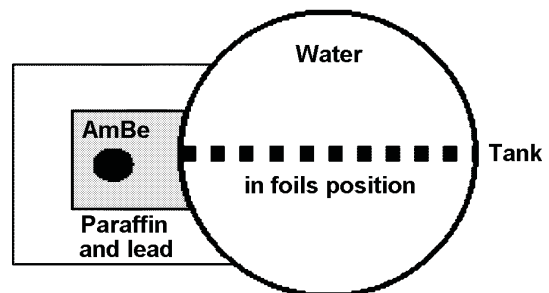


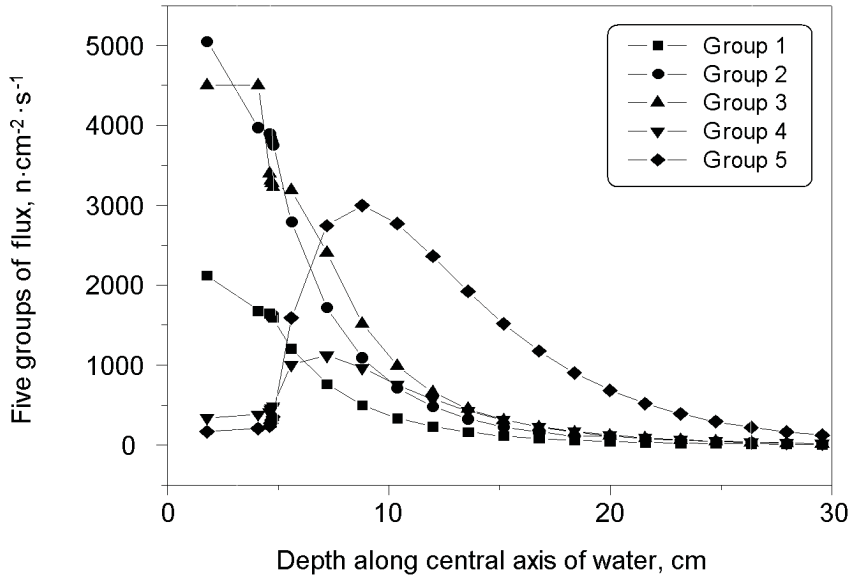*Fig. 1.* Experimental setup for thermal flux measurements

*Fig. 2.* Thermal and not thermal flux variation versus depth along the central axis of water
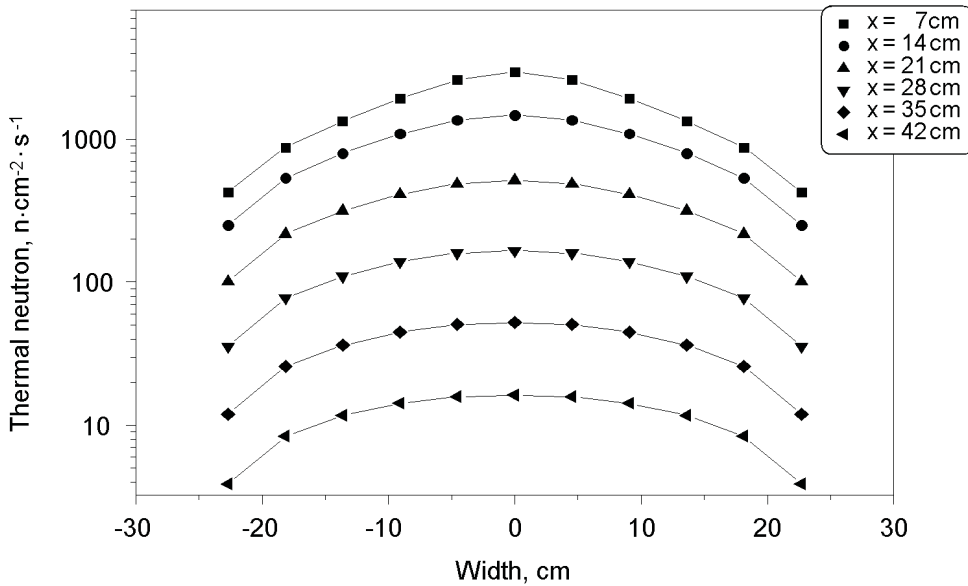


*Fig. 3.* Thermal flux across the width of the water at various depth (x, y, 0)

Because of very high resonance, it was not a pure thermal activation with the indium foil and that is why the epithermal contribution was considered. For this, the indium foil was encapsulated inside a cadmium cover and a second set of measurement was carried out in the same conditions. This experimental procedure permits the measurements of the thermal flux distribution, which is plotted in Fig. 4 and to compare them with the theoretical results. The calculated values for thermal flux distribution curve were adjusted with a pulse fit. It appears a good agreement in the shape of the curves and practically not significant variation from the measured and calculated values of flux.
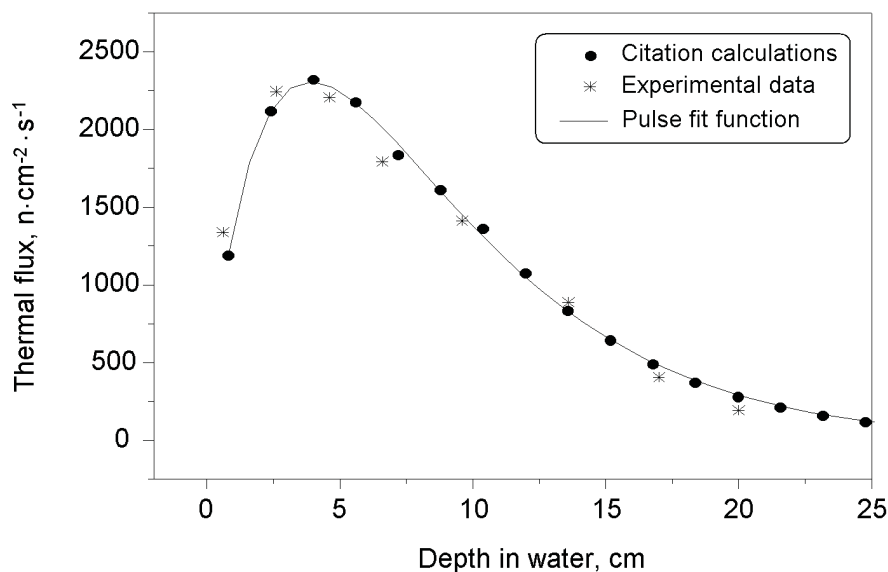
*Fig. 4.* Relative thermal neutron fluency versus depth along the central axis of water

The highest value of the thermal neutrons flux was $2.3 \cdot 10^3$ n·cm$^{-2}$·s$^{-1}$, which corresponds to a depth of 4.3 cm along the central axis of water while the maximum measured value was 4.1 cm. So, the best position of irradiation is located around 2 to 5 cm inside the water tank. Beyond 20 cm, the flux decrease was $10^2$ n·cm$^{-2}$·s$^{-1}$.

## Conclusions

The calculations with CITATION code shows that the flux values obtained correspond perfectly to the measured flux values. The knowledge of thermal flux distribution inside the voluminous water sample permits the calibration of PGNAA for some light elements in liquid sample as routine method.

These calculations allowed us the knowledge of the optimal value of flux then, the best sensitivity for small sample analysis by INAA using this simple experimental setup. It appears from the results that the experimental setup can be optimized by reducing the actual volume of water to 80 liters.

## References

1. P. Bode, R. M. W. Overwater, J. Radioanal. Nucl. Chem., 167 (1993) 169.
2. R. Khelifi et al., Intern. J. Appl. Radiation Isotopes, 51 (1999) 9.
3. N. M. Spyrou et al., J. Radioanal. Nucl. Chem., 259 (2004) 287.
4. C. Chung, T. C. Tseng, Nucl. Instr. Meth., A267 (1988) 223.
5. C. H. Chao, C. Chung, Nucl. Instr. Meth., A299 (1990) 651.
6. R. M. W. Overwater, P. Bode, J. J. M. Degoeij, Nucl. Instr. Meth., A324 (1993) 209.
7. T. B. Fowler et al., Nuclear Reactor Core Analysis Code: CITATION, ORNL-TM-2496, Revision 2, July 1971.
8. ISO 8529-1, Reference Neutron Radiations – Part 1: Characteristics and Methods of Production, International Organization of Standardization, 8529(1), 2002.
9. International Atomic Energy Agency, Compendium of Neutron Spectra and Detector Responses for Radiation Protection Purposes, Supplement to Technical Reports Series No. 318, IAEA, Vienna, 2001.