

OVERVIEW OF INAA METHOD AND ITS APPLICATION IN MALAYSIA

A.R.Yavar¹, S. B. Sarmani², H. Khalafi³, A. K. Wood⁴, K. S. Khoo^{1,*}

¹School of Applied Physics, Faculty of Science and Technology, Universiti Kebangsaan Malaysia (UKM), 43600 Bangi, Selangor, Malaysia.

²Chemistry Programme, School of Chemical Sciences & Food Technology, Faculty Science and Technology, Universiti Kebangsaan Malaysia (UKM), 43600 Bangi, Selangor, Malaysia.

³Nuclear Science and Technology Research Institute (NSTRI), Atomic energy organization of Iran, North Kargar Avenue, P. O. Box: 14155-1339, 14374, Tehran, Iran.

⁴Analytical Chemistry Application Group, Industrial Technology Division, Malaysian Nuclear Agency (MNA), Bangi, 43000 Kajang, Selangor, Malaysia.

*e-mail: khoo@ukm.my

ABSTRACT

Present work shows the development of nuclear technology in Malaysia and highlights its applications that have been developed by using the instrumental neutron activation analysis (INAA) method. In addition, present study exhibits a comprehensive review of INAA for calculation of neutron flux parameters and concentration of elements. The INAA is a powerful method to analyse the sample which identifies qualitative and quantitative of elements present in a sample. The INAA is a working instrument with advantages of experimental simplicity, high accuracy, excellent flexibility with respect to irradiation and counting conditions, and suitability for computerization. In INAA, sample is irradiated and measured directly. In practical, INAA is based on an absolute, relative and single-comparator standardisation method. The INAA has been developed since 1982 when the TRIGA Mark II reactor of Malaysia has commissioned. The absolute method was less utilised, the relative method has been used since 1982, and the k_0 -INAA method is derived from single-comparator standardization method has been developed since 1996 in Malaysia. The relative method, because of its advantages, such as high accuracy, easy for using, has many applications in Malaysia. Currently, local universities and Malaysian Nuclear Agency (MNA) research reactor use INAA method in Malaysia.

ABSTRAK

Kajian ini menunjukkan pembangunan teknologi nuklear di Malaysia dan menyerlahkan dengan aplikasi yang telah dibangunkan dengan menggunakan kaedah analisis pengaktifan neutron (INAA). Di samping itu, kajian semasa menunjukkan semakan yang komprehensif terhadap penggunaan teknik INAA dalam pengiraan parameter berkaitan fluks neutron dan kepekatan unsur. INAA ini adalah satu kaedah yang berkuasa untuk menganalisis sampel dalam mengenal pasti unsur-unsur yang hadir dalam sampel kuantitatif dan kualitatif. INAA itu adalah satu instrumen bekerja dengan kelebihan eksperimen bersifat sederhana, berteknologi, fleksibiliti yang cemerlang berkaitan dengan sinaran dan

keadaan pembilangan dan kesesuaian untuk pengkomputeran. Dalam teknik INAA, sampel disinari dan diukur secara langsung. Secara praktikal, INAA adalah berdasarkan satu kaedah penyeragaman mutlak, dan menggunakan teknik perbandingan-satu secara relatif. INAA telah digunakan sejak tahun 1982 sejak reaktor TRIGA Mark II Malaysia ditauliahkan. Kaedah mutlak kurang digunakan, manakala kaedah relatif digunakan sejak 1982, dengan kaedah k₀ diterbitkan daripada kaedah perbandingan-satu secara relatif. Keadah relatif ini, dengan kelebihanannya seperti kejituan tinggi dan mudah digunakan, mempunyai paling banyak penggunaannya di Malaysia. Pada masa ini, reaktor penyelidikan di Agensi Nuklear Malaysia dan universiti tempatan di Malaysia menggunakan teknik INAA.

Keywords: Instrumental Neutron Activation Analysis (INAA), k₀-INAA method, Absolute method, Relative method

INTRODUCTION

Neutron Activation Analysis (NAA) was introduced by Georg von Hevesy and Hilde Levi as a method for the quantitative determination of element concentrations an early as 1936, in which neutrons are used to activate nuclei in the sample. All atomic nuclei in the sample have a probability of capturing a neutron. This probability is expressed in units of area and called the neutron capture cross section (σ). The neutron flux is expressed as intensity per unit area per unit time ($n/cm^2/sec$). Nuclei with the same number of protons but different numbers of neutrons are isotopes of each other, i.e. belongs to the same element. The fraction of nuclei of a certain element that have a particular number of neutrons is the isotopic abundance (θ). After capturing a neutron, the nucleus may have become unstable, i.e. radioactive. The level of induced radioactivity depends on the number of generated unstable nuclei and the half life of the radionuclide. When decaying at a later time to a stable form, it may emit γ - rays, each with a particular probability called the absolute gamma intensity (γ). The emitted γ -rays can be detected with a semiconductor detector with a very high energy resolution. In the resulting γ -ray spectrum, the energy of each detected peaks indicates what element presents in the sample, and the areas of detected peaks allow for the quantitative determination of amount of the element present in the sample. The INAA with main advantages as compared to some other trace element analysis techniques are as: INAA is non-destructive, i.e. the sample need not be dissolved and the probability of loss or contamination is therefore low; INAA is nuclear, i.e. the method is independent of the chemical and physical state of the sample; INAA is insensitive to low-Z element, i.e. other elements in low-Z matrices can be determined with high sensitivities; INAA is linear, i.e. after calibration at only one concentration level, the technique is accurate no matter whether ppm's or percentages close to 100% are to be determined. In practical, INAA is based on an absolute, relative or single-comparator standardization method [1-12].

In the beginning, the Ra-Be mixtures were used as neutron source. The long-lived radium (Ra-226) radioactive isotope will decay by emitting an alpha particle and this particle is just like helium (He) nucleus with 2 protons and 2 neutrons. When mix this source with a sample of light isotope such as beryllium (${}^9\text{Be}$) and the following reaction will occur:



Later, with the development of the nuclear reactor, a powerful neutron source has become more available, yielding substantially larger neutron fluxes and therefore providing the feasibility of determination of lower concentrations or the analysis of smaller samples. The Geiger-Muller (GM) counter was used for counting beta and γ -rays. However, there was no energy resolution available. Therefore, all elements were identified based on half-lives and chemical separations carried out after the irradiation. This technique is called radiochemical NAA

(RNAA). Later, with the development of NaI scintillation detectors introduced the possibility of measuring a γ -ray spectrum with an energy resolution of 4 % at 1 MeV. Then, improvement was made when Ge(Li) semiconductor material was applied for analysis with 0.1 % resolution at 1 MeV. The advantage of these detectors is that the chemical separation steps are skipped. The INAA then was introduced.

The absolute method

The application of the absolute neutron activation method goes back more than 50 years, that the first systematic methodological investigation was reported by GIRARDI et al. during the time of scintillation detectors. In the absolute standardization method, the physical parameters determining an elemental characteristic, e.g. σ , θ , ν and M (atomic mass) are taken from literature. For many (n, ν) reactions of interest, these parameters are not precisely known. Because they were determined by independent methods, their imprecision will add up when calculating the amount of elements, leading to large systematic errors of more than 100% in some cases. The use of the absolute method presents several advantages over the relative method on the basis of speed, cheapness, versatility and ease of automation. It also presents the possibility of multi-element analysis in one single irradiation. But its disadvantages are as: it has been long recognized that the nuclear data, ν -ray emission probabilities and neutron fluxes are the major sources of errors in the absolute method. Indeed they are and they have to be known with reasonable precision. This method will work well in very stable reactors where flux changes are negligible, otherwise continuous flux monitoring is essential. The efficiency, essential coincidence and attenuation corrections and geometry may reduce the achievable precision particularly when extended sources are to be counted [4, 5, 7, 9, 13]. The absolute method calculates the ρ_a concentration (g/g) of elements as:

$$\rho_a = 1.661 \cdot 10^{-24} \cdot \left(\frac{N_p}{W.S.D.C.t_c} \right) \cdot \left(\frac{M}{\theta.\gamma.R.\varepsilon_p} \right) \quad (2)$$

which N_p is measured gamma net peak area (counts); t_c is counting time; S is saturation factor; $S = 1 - e^{-\lambda t_i}$, with t_i irradiation time and $\lambda = \frac{\ln 2}{T_{1/2}}$ with $T_{1/2}$ half life; D is decay factor; $D = e^{-\lambda t_d}$, with t_d decay time; C is counting factor; $C = (1 - e^{-\lambda t_c}) / \lambda t_c$, correcting for decay during counting; W is mass of irradiated element (g); θ is Isotope abundance (fraction); ε_p is Full-energy peak detection efficiency; and R is reaction rate.

Also the absolute method is utilised for calculation of thermal to fast neutron flux ratio (f_{fast}) and fast neutron flux (ϕ_{fast}) using the reaction of “1” = $^{58}\text{Fe}(n, \nu)^{59}\text{Fe}$ and the reaction of “2” = $^{96}\text{Zr}(n, \nu)^{97}\text{Zr}/^{97m}\text{Nb}$ as follows [4, 5, 7, 9, 13]:

$$f_{fast} = \frac{A_{SP,2}}{A_{SP,1}} \cdot \frac{\left(\frac{\theta.\gamma.\sigma}{M} \right)_1}{\left(\frac{\theta.\gamma.\sigma}{M} \right)_2} \cdot \frac{f}{f + Q_{0,2}(\alpha)} \cdot \frac{\varepsilon_{p,1}}{\varepsilon_{p,2}} \quad (3)$$

$$\phi_{fast} = \frac{\phi_{th}}{f_{fast}} \quad (4)$$

where the specific count rate ($s^{-1}g^{-1}$) is defined:

$$A_{SP} = \frac{N_p / t_C}{SDCW} \quad (5)$$

$Q_0(\alpha)$ is obtained as:

$$Q_0(\alpha) = \frac{Q_0 - 0.429}{E_r^\alpha} + \frac{0.429}{(2\alpha + 1) \cdot (0.55)^\alpha} \quad (6)$$

\overline{E}_r is effective resonance energy in eV; $Q_0 = I_0/\sigma_0$ with I_0 is the resonance integral for the (n, γ) reaction and σ_0 is the thermal neutron cross section (2200 ms⁻¹); M is atomic mass (g.mol⁻¹); α is expression for the deviation of the epithermal neutron distribution from 1/E shape, approximated by a 1/E^{1+ α} dependence.

The ϕ_{th} is the thermal neutron flux (cm²s⁻¹); the thermal neutron flux (ϕ_{th}) and epithermal neutron flux (ϕ_{epi}) are calculated as follows:

$$\phi_{th} = \frac{f \cdot A_{sp,Au} \cdot 3.47}{(f + Q_{0,Au}(\alpha)) \cdot \varepsilon_{p,Au}} \quad (7)$$

$$\phi_{epi} = \frac{\phi_{th}}{f} \quad (8)$$

The absolute method is utilised in the fast neutron activation analysis (FNAA). The most important applications of FNAA are the analysis of oxygen content in a wide variety of matrices including metals, geologic materials, coal, liquid fuels, ceramic materials, petroleum derivatives and fractions and chemical reaction products. The determination of nitrogen in biological materials, including nitrogen as a measure of protein content as well as nitrogen determination in fertilizers, explosives, and polymers is also important applications. Other elements that are routinely analyzed by FNAA include Ag, Al, Au, Si, P, F, Cu, Mg, Mn, Fe, Zn, As, and Sn [4, 5, 7, 14-21].

The relative method

In the relative standardization method, the unknown sample is irradiated together with a calibration sample containing a known amount of the element of interest. The calibration sample or standard is measured under the same conditions as the sample. The ratio of the net areas of the photo peaks corresponding to the element of interest in the two measured spectra is used to calculate the concentration. Advantages of this method are as: In this procedure, all parameters except the half life of the radionuclide of interest cancel out and therefore are of no consequence. This standardization method is still being regarded as one of method which has the highest accuracy of NAA. It eliminates many errors such as those due to flux parameters, nuclear data, decay scheme, efficiency, self-shielding, coincidence summing. Disadvantages of this method are as: It is not suited for multi-element analysis. It is impossible to put individual standard for all 70 detectable elements that might be present in the sample in the same place as the sample during irradiation. It is also virtually impossible to produce a multi- element standard containing known amounts of all these elements with sufficient accuracy, homogeneity and stability. Sometimes, certified reference materials are used as multi-element standards. This is a dangerous practice, because reference materials are not primary standards certified concentration often are imprecise, sometimes even inaccurate.

The ρ_r concentration (g/g) of elements in sample is obtained by measurment of sample and comparator (*) as follows:

$$\rho_r = \frac{\left(\frac{N_p}{W.S.D.C.t_c} \right) \cdot \varepsilon_p^*}{\left(\frac{N_p}{W.S.D.C.t_c} \right)^* \cdot \varepsilon_p} \quad (9)$$

In ideal case the ratios S*/S and $\varepsilon^*/\varepsilon$ are equal to unity [3, 7-9].

The single comparator standardization method

The single comparator standardization method makes multi-element analysis with INAA feasible. Assuming stability in time of all relevant experimental conditions, standards for all elements are irradiated each in turn with the chosen single comparator element. Once the sensitivity for all elements relative to the comparator element is known, this comparator element can be used in routine measurements instead of separate standard for each element.

The original single comparator method is expressed in the definition of the k-factors, which are experimentally determined by irradiation of a standard and a single-comparator:

$$k_c(s) = \frac{M_c \gamma_s \theta_s \sigma_{0,s}}{M_s \gamma_c \theta_c \sigma_{0,c}} \cdot \frac{f + Q_{0,s}(\alpha)}{f + Q_{0,c}(\alpha)} \cdot \frac{\varepsilon_{p,s}}{\varepsilon_{p,c}} \quad (10)$$

Where, c and s denote for comparator and standard elements, respectively. These k-factors, obtained from direct measurements, are usually much more precise than independent physical parameters obtained from literature data in the absolute standardization method. On the other hand, the measured k-factors are valid only for a specific detector, counting geometry and irradiation facility, and remain valid only as long as the neutron flux parameters of the irradiation facility remain stable. The standardization methods have usefully contributed in many application fields of NAA, however, they have also been prompted by the inconveniences in application, i.e. in the relative standardization, the experimental workload, the impossibility to quantify unexpected elements and the unsuitable for multi-element analysis; in the absolute standardization, the inaccuracy and inconsistency of the nuclear data; and in the single-comparator standardization, the inflexibility with respect to varying irradiation and counting conditions. The advantages of this method are as: Although it presents similar advantages to that of absolute methods, the problem of flux variations is removed when using the comparator method. Moreover, the flux-ratio, efficiency, k_0 , etc. may be determined precisely hence reducing contributions to the total uncertainty. The disadvantages of this method are as: The problem of choosing suitable comparator elements for multi-element analysis may not be easy as far as nuclear data and decay scheme parameters are concerned. In the every day practice of NAA, counting at small source-to-detector distances for extended sources is common, problems of correction for coincidences and attenuation may involve tedious calibration procedure, experimental corrections and a complicated computer calculation [1-7, 9-11].

The k_0 -INAA method

Through the years, many efforts have been spent to overcome the disadvantages of the above mentioned standardization methods. Generally, the required aspects for a new standardization protocol are: (i) experimental simplicity; (ii) high accuracy; (iii) excellent flexibility (with respect to the irradiation and counting conditions); and (iv) suitability for computerization. These are the reasons so that the k_0 -standardization method for INAA (k_0 -INAA), one of the remarkable developments of INAA, launched in the mid-70s. It is not a theory describing a physical phenomenon, but a protocol for calibration procedures. It has been developed as an absolute standardization where the unreliable nuclear data are replaced by accurate experimentally determined compound nuclear constants, so called k_0 -factors, or as a single-comparator standardization which is made flexible with respect to varying characteristics of the k_0 -factors, the independence with irradiation and measurement conditions is done, k_0 -factor is expressed as:

$$k_{0,c}(s) = \frac{M_c \gamma_s \theta_s \sigma_{0,s}}{M_s \gamma_c \theta_c \sigma_{0,c}} \quad (12)$$

which can be tabulated and published in literature as a generally useful parameter. Then, by converting $k_{0,m}(s) = k_{0,c}(s) / k_{0,c}(m)$, the irradiated sample with monitor m, the analytic concentration can be obtained [22]. The k_0 -method was formulated in the Høgdahl convention and Westcott-formalism. The parameters such as thermal to epithermal neutron flux ratio (f) and epithermal neutron flux shape factor (α) are determined by the Høgdahl convention. Since the applicability of Høgdahl convention is restricted to (n, γ) reactions for which

Westcott's g-factor is equal to unity, this convention excludes handling of "non-1/v" (n, v) reactions of nuclides (e.g, ¹⁷⁶Lu, ¹⁵¹Eu etc.) with Westcott's g≠1. For the k₀-NAA to be generally applicable for all nuclides, the Westcott-formalism is adopted and parameters such as the modified spectral index $r(\alpha)\sqrt{T_n/T_0}$, the Westcott $g_{Lu}(T_n)$ factor and the absolute neutron temperature T_n are determined besides α and f [1-11, 23-34].

Parameters of the Høgdahl convention

The α factor

The α factor can be determined from three method: Cd-ratio, Cd-covered, and bare irradiation methods as described below [1, 9, 11, 35-37]:

"Cd-covered multi-monitor" method

A set of N monitors is irradiated simultaneously under Cd-covered and subsequently counted on a Ge-detector, α can be obtained as the slope - α of the straight line when plotting:

$$\log \frac{(\bar{E}_{r,i})^{-\alpha} (A_{sp,i})_{Cd}}{k_{o,Au}(i) \cdot \epsilon_{p,i} \cdot F_{Cd,i} \cdot Q_{o,i}(\alpha) \cdot G_{e,i}} \text{ versus } \log \bar{E}_{r,i} \quad (13)$$

where i denotes isotope 1, 2 ...N, F_{Cd} is Cd-transmission factor and G_e is epithermal Neutron Self-shielding Correction. The left hand term of Eq. (11) is itself a function of α , and thus an iterative procedure should be applied. aslo α can be solved as follows:

$$\alpha + \frac{\sum_{i=1}^N \left\{ \left[\log \bar{E}_{r,i} - \frac{\sum_{i=1}^N \log \bar{E}_{r,i}}{N} \right] \left[\log T_i - \frac{\sum_{i=1}^N \log T_i}{N} \right] \right\}}{\sum_{i=1}^N \left[\log \bar{E}_{r,i} - \frac{\sum_{i=1}^N \log \bar{E}_{r,i}}{N} \right]} = 0 \quad (14)$$

with,

$$T_i = \frac{(\bar{E}_{r,i})^{-\alpha} (A_{sp,i})_{Cd}}{k_{o,Au}(i) \cdot \epsilon_{p,i} \cdot F_{Cd,i} \cdot Q_{o,i}(\alpha) \cdot G_{e,i}} \quad (15)$$

when a irradiation of Au and Zr monitors is made under Cd-cover, i.e. in Eqs. (11), (12) & (13), N=3 and a specific case of the "Cd-covered multi-monitor" method would be so- called the "Cd-covered triple monitor" method.

"Cd-ratio for multi-monitor" method

A set of N monitors is irradiated with and without Cd-cover, and the induced activities are measured on a Ge detector. The α can be obtained as the slope - α of the straight line when plotting:

$$\log \frac{(\bar{E}_{r,i})^{-\alpha}}{(F_{Cd,i} \cdot R_{Cd,i} - 1) \cdot Q_{o,i}(\alpha) \cdot G_{e,i} / G_{th,i}} \text{ versus } \log \bar{E}_{r,i} \quad (16)$$

where i denotes isotope 1, 2... N and G_{th} is correction factor for thermal neutron self-shielding.

As in the "Cd-covered multi-monitors" method, α can be solved from Eq. (12) with:

$$T_i = \frac{(\bar{E}_{r,i})^{-\alpha}}{(F_{Cd,i} \cdot R_{Cd,i} - 1) \cdot Q_{o,i}(\alpha) \cdot G_{e,i} / G_{th,i}} \quad (17)$$

In this method, the use of monitors with very high Q_0 -value should be avoided.

“Bare multi-monitor” method:

A set of N monitors, together with a ‘reference’ monitor isotope, are irradiated without Cd-cover, thereafter the induced activities are measured on a Ge-detector. In this method, there is a possibility to make use of the ‘bare triple monitor’ method, by using Eqs of below:

$$(a-b)Q_{0,1}(\alpha)G_{e,1}/G_{th,1} - aQ_{0,2}(\alpha)G_{e,2}/G_{th,2} + bQ_{0,3}(\alpha)G_{e,3}/G_{th,3} = 0 \quad (18)$$

with,

$$a = \left\{ 1 - \frac{A_{sp,2}}{A_{sp,1}} \cdot \frac{k_{0,Au}(1)}{k_{0,Au}(2)} \cdot \frac{\varepsilon_{p,1}}{\varepsilon_{p,2}} \right\}^{-1}; b = \left\{ 1 - \frac{A_{sp,3}}{A_{sp,1}} \cdot \frac{k_{0,Au}(1)}{k_{0,Au}(3)} \cdot \frac{\varepsilon_{p,1}}{\varepsilon_{p,3}} \right\}^{-1} \quad (19)$$

which it come with irradiation Au and Zr monitors under reactions of “1” = $^{96}\text{Zr}(n, \gamma)^{97}\text{Zr}/^{97m}\text{Nb}$; “2” = $^{94}\text{Zr}(n, \gamma)^{95}\text{Zr}$; “3” = $^{197}\text{Au}(n, \gamma)^{198}\text{Au}$.

Methods for determination of f-factor

The parameter f, the ratio of the thermal to epithermal neutron flux can be determined by the “Cd-ratio” method or “bare bi-isotopic monitor” method. Cd-ratio method using Eq. (18) as follows [1, 9, 11, 35-37]:

$$f = (F_{Cd}R_{Cd} - 1)G_e Q_0(\alpha) / G_{th} \quad (20)$$

In Eq. (18), the monitor used is an element which is irradiated subsequently with and without Cd-cover. A gold or cobalt monitor is suitable for this requirement. It is obviously that the α -value must be inputted to calculate the $Q_0(\alpha)$ parameter.

A proven technique for in-situ f-determination is the “bare bi-isotopic monitor” method using zirconium monitor with reactions of “1” = $^{96}\text{Zr}(n, \gamma)^{97}\text{Zr}/^{97m}\text{Nb}$; “2” = $^{94}\text{Zr}(n, \gamma)^{95}\text{Zr}$.

$$f = \frac{G_{e,1} \frac{k_{0,Au}(1)}{k_{0,Au}(2)} \cdot \frac{\varepsilon_{p,1}}{\varepsilon_{p,2}} \cdot Q_{0,1}(\alpha) - G_{e,2} \frac{A_{sp,1}}{A_{sp,2}} \cdot Q_{0,2}(\alpha)}{G_{th,2} \frac{A_{sp,1}}{A_{sp,2}} - G_{th,1} \frac{k_{0,Au}(1)}{k_{0,Au}(2)} \cdot \frac{\varepsilon_{p,1}}{\varepsilon_{p,2}}} \quad (21)$$

The “bare bi-isotopic monitor” is a convenient method for f-determination.

The parameters of Westcott-formalism

The $S_{0,Lu}(\alpha)$ factor

The value of $S_{0,Lu}(\alpha)$ determine using the following expression:

$$S_0(\alpha) = \frac{S_0}{E_r^\alpha} \times (1eV)^\alpha \quad (22)$$

where S_0 is the corresponding quantity for an ideal 1/E epithermal neutron flux distribution [3, 7, 9].

The modified spectral index $r(\alpha)\sqrt{T_n/T_0}$

The modified spectral index $r(\alpha)\sqrt{T_n/T_0}$ is a measure for the epithermal to total neutron density ratio. It must be considered as one single parameter, and as such it can be experimentally determined from the “bare bi-isotopic monitor” method using Zr monitor and reactions of “1” = $^{96}\text{Zr}(n, \gamma)^{97}\text{Zr}/^{97\text{m}}\text{Nb}$; “2” = $^{94}\text{Zr}(n, \gamma)^{95}\text{Zr}$:

$$r(\alpha)\sqrt{T_n/T_0} = \frac{G_{th,2} \cdot \frac{k_{0,Au}(1)}{k_{0,Au}(2)} \cdot \frac{\varepsilon_{p,1}}{\varepsilon_{p,2}} \cdot g_1(T_n) - G_{th,1} \cdot \frac{A_{SP,1}}{A_{SP,2}} \cdot g_2(T_n)}{G_{r,2} \cdot \frac{A_{SP,1}}{A_{SP,2}} \cdot S_{0,2}(\alpha) - G_{r,1} \cdot \frac{k_{0,Au}(1)}{k_{0,Au}(2)} \cdot \frac{\varepsilon_{p,1}}{\varepsilon_{p,2}} \cdot S_{0,1}(\alpha)} \quad (23)$$

where G_r is correction factor for resonance neutron self-shielding and $g_{Lu}(T_n)$ is the Westcott g-factor at a neutron temperature T_n . Also $r(\alpha)\sqrt{T_n/T_0}$ can be obtained from the ‘Cd-ratio’ method:

$$r(\alpha)\sqrt{T_n/T_0} = \frac{G_{th} \cdot g(T_n)}{R_{Cd} \cdot F_{Cd} \left[\frac{g(T_n) \cdot (1eV)^\alpha}{K \cdot (1+2\alpha) \cdot E_{Cd}^\alpha} - \frac{2}{\sqrt{\pi}} W'(\alpha) + G_r \cdot S_0(\alpha) \right] - G_r \cdot S_0(\alpha)} \quad (24)$$

which $W'(\alpha) = W'(\bar{E}_r)^{-\alpha} (1eV)^\alpha$

The W' is known value for each nuclide. $K = \frac{1}{4} \sqrt{\frac{\pi E_{Cd}}{E_0}}$ with $E_0 = 0.0253 \text{ eV}$ [3, 7, 9].

The Westcott $g_{Lu}(T_n)$ factor

The $g_{Lu}(T_n)$ factor and T_n evaluate by bare irradiation of “non 1/v” Lu target (reaction of “4” = $^{176}\text{Lu}(n, \gamma)^{177}\text{Lu}$) and pure 1/v target (one of the reactions of “1” = $^{96}\text{Zr}(n, \gamma)^{97}\text{Zr}/^{97\text{m}}\text{Nb}$; “2” = $^{94}\text{Zr}(n, \gamma)^{95}\text{Zr}$; “3” = $^{197}\text{Au}(n, \gamma)^{198}\text{Au}$) by following expression:

$$g_{Lu}(T_n) = \left[\frac{\left[\frac{A_{SP}}{k_{0,Au} \cdot \varepsilon_p} \right]_{Lu}}{\left[\frac{A_{SP}}{k_{0,Au} \cdot \varepsilon_p} \right]_{1/v}} \cdot \left(g_{1/v}(T_n) + r(\alpha)\sqrt{T_n/T_0} \cdot s_{0,1/v}(\alpha) \right) \right] - r(\alpha)\sqrt{T_n/T_0} \cdot s_{0,Lu}(\alpha) \quad (25)$$

The corresponding $r(\alpha)\sqrt{T_n/T_0}$ values use to calculate the $g_{Lu}(T_n)$ factor. The T_n value obtains using the literature values of $g_{Lu}(T_n)$ vs. T_n [3, 38].

Concentration calculation in Høgdahl convention and Westcott formalism

The concentration of an element in a sample by Høgdahl convention is calculated as:

$$\rho_H = \frac{\left(\frac{N_p/t_C}{SDCW} \right)_a}{A_{SP,m}} \cdot \frac{1}{k_{0,m}(a)} \cdot \frac{G_{th,m} \cdot f + G_{e,m} \cdot Q_{0,m}(\alpha)}{G_{th,a} \cdot f + G_{e,a} \cdot Q_{0,a}(\alpha)} \cdot \frac{\varepsilon_{p,m}}{\varepsilon_{p,a}} \quad (26)$$

where ρ_H is concentration of analyst a (in g/g); m is irradiated neutron fluence rate monitor and W is sample mass (in grams). The actual equation used in the Westcott formalism for ρ_W concentration (in g/g) calculation is:

$$\rho_W = \frac{\left(\frac{N_p/t_C}{SDCW} \right)_a}{A_{SP,m}} \cdot \frac{1}{k_{0,m}(a)} \cdot \frac{g_{Au}(T_n) + r(\alpha)\sqrt{T_n/T_0} \cdot S_{0,Au}(\alpha)}{g_a(T_n) + r(\alpha)\sqrt{T_n/T_0} \cdot S_{0,a}(\alpha)} \cdot \frac{\varepsilon_{p,m}}{\varepsilon_{p,a}} \quad (27)$$

The Eq. 25 is utilised for concentration calculation of the Westcott elements such as $^{176}\text{Lu}(n, \nu)^{177}\text{Lu}$, $^{151}\text{Eu}(n, \nu)^{152}\text{Eu}$, $^{151}\text{Eu}(n, \nu)^{152\text{m}}\text{Eu}$, $^{153}\text{Eu}(n, \nu)^{154}\text{Eu}$, $^{164}\text{Dy}(n, \nu)^{165\text{m}}\text{Dy}$, $^{164}\text{Dy}(n, \nu)^{165}\text{Dy}$, $^{168}\text{Yb}(n, \nu)^{169}$ and $^{175}\text{Lu}(n, \nu)^{176\text{m}}\text{Lu}$ [3, 5, 7, 9].

Applications of INAA in Malaysia

The TRIGA Mark II reactor of the Malaysian Nuclear Agency was commissioned in 1982. This reactor uses light-water as moderator, coolant and reflector. The fuel assembly consists of an alloy of uranium U-235 and zirconium hydride (U-ZrH). Several experimental facilities are available in the MNA research reactor. For activation analysis and isotope production, a rotary specimen rack is located around the top portion of the core and inside the reflector. The rotary specimen rack assembly consists of ring-shaped, seal-welded aluminium housing containing an aluminium rack mounted on special bearings. The rotary rack (RR) supports 40 evenly spaced tubular aluminium containers that serve as receptacles for the specimen containers. Each receptacle has an inside diameter of 3.17 cm and height of 27.4 cm and can hold two specimen containers. At Present most of reactor operation time has been utilised for samples irradiation related to the INAA application. Majority of the samples are from MNA analytical chemistry laboratory, and the rest of the samples are from local universities [39].

As shown in Fig 1. only one study by absolute method [40], forty five studies by relative method [41-85], and nineteen studies by k_0 -INAA [86-104] were carried out in Malaysia. It indicates the relative method, because of its advantages as easy for using, had the most application in Malaysia.

As shown in Fig. 2, most of application of INAA in Malaysia is in environmental field (29 papers), 17 papers in nutritional epidemiological studies, 7 papers in nuclear data studies, 4 papers in quality assurance of analysis and reference materials studies, 3 papers in industrial materials analysis, 3 papers in geology and geochemistry studies, 2 papers in archaeological studies, and 0 paper in forensic studies [40-104].

In order to utilising of INAA method as entirely, determination of parameters f , α , f_{fast} , ϕ_{th} , ϕ_{epi} , ϕ_{fast} , $S_0(\alpha)$, $r(\alpha)\sqrt{T_n/T_0}$, $g_{Lu}(T_n)$ and T_n are necessary. The parameters of f and α determined for first time by Abugassa et al. [86] in Malaysia. The f and α are useful for trace elements by k_0 -INAA method base on Høgdahl convention. The parameters of ϕ_{th} and ϕ_{epi} calculated by Wee et al. [98] for first time in Malaysia. The ϕ_{th} and ϕ_{epi} evaluate distribution of neutron flux in reactor. The parameters of f_{fast} and ϕ_{fast} determined by Yavar et al. [101] for first time in Malaysia. The parameters of f_{fast} and ϕ_{fast} are useful for FNAA applications. In order to using the FNAA, special facilities need to install in MNA research reactor. The parameters of $S_0(\alpha)$, $r(\alpha)\sqrt{T_n/T_0}$, $g_{Lu}(T_n)$ and T_n were determined by Yavar et al. [104] for first time in Malaysia. The Westcott parameters of $S_0(\alpha)$, $r(\alpha)\sqrt{T_n/T_0}$, $g_{Lu}(T_n)$ and T_n use to trace elements that handle “non-1/ ν ” (n, ν) reactions of nuclides (e.g, ^{176}Lu , ^{151}Eu) with Westcott’s $g \neq 1$.

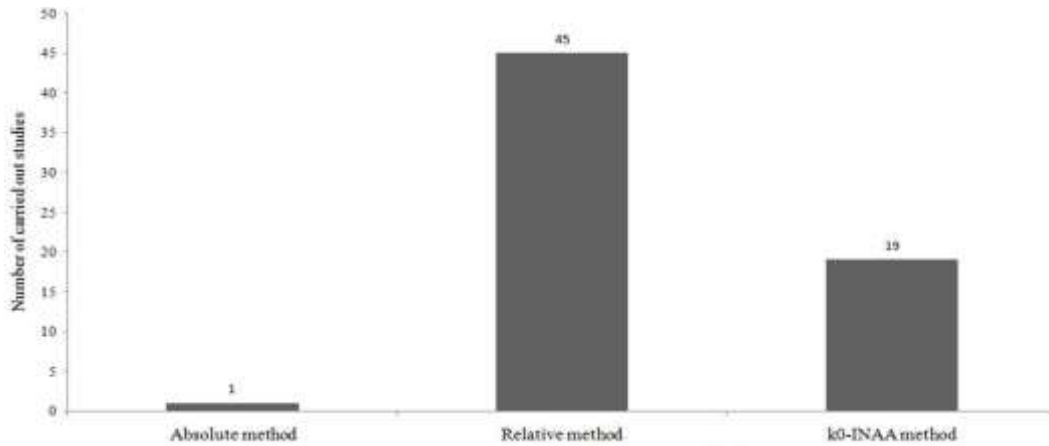


Figure 1. Number of publications related to absolute, relative and k₀-NAA methods in Malaysia since 1982.

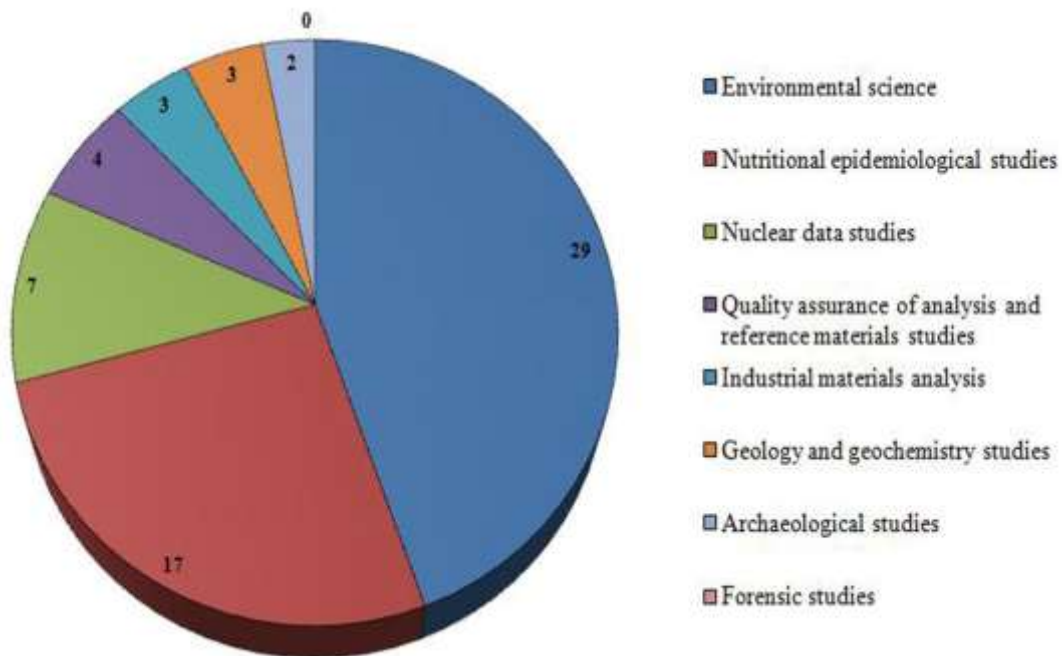


Figure 2. Number of publications related to INAA applications in Malaysia

CONCLUSION

The INAA method has utilised as a powerful instrument for determination of elemental concentration in Malaysia since 1982. The INAA was developed as absolute, relative and k₀-INAA methods, respectively. The INAA utilises to trace elements present in geological, environmental, and biological samples. The absolute method is useful for FNAA application. After installation of FNAA facilities in Malaysian Nuclear Agency (MNA) research reactor, FNAA applications will be used in Malaysia. The k₀-INAA method based on Høgdahl convention and Westcott-formalism has developed for determination of neutron flux parameters at MNA research reactor and trace elements increasingly since 1996 in Malaysia. The relative method by advantage of experimental simplicity has the most application in Malaysia.

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