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PARAMETERISATION OF FISSION NEUTRON SPECTRA (TRIGA REACTOR) FOR NEUTRON ACTIVATION WITHOUT THE USED OF STANDARD

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Neutron activation analysis (NAA) is a sensitive and reliable technique for both qualitative and quantitative analysis of wide range of elements in various types of samples. The method involves the irradiation of samples and the detection of gamma energies emitted from the isotopes formed from the process of neutron capture. Most NAA were done by comparison method, which is found to have high errors due to the differences in the matrix composition of sample as well as comparator. The purpose of this study is to demonstrate an alternative technique of activation analysis based on absolute gamma ray measurements and the direct calculation of elemental concentrations from reaction rates equation of neutron capture process. The neutron spectrum parameters (f and α), thermal and epithermal neutron flux (φth, φ_{epi},) at a specific irradiation position of the rotary rack 1-MW TRIGA reactor at Malaysia Nuclear Agency were determined. The accuracy and precision of this technique were verified by analyzing two certified reference materials provided by

IAEA, Soil-1 and Soil-7. The experimental results obtained for both the materials were found to be in good agreement with the certified values. Signified by the average z-score for the concentration values were below 2 for most elements. In conclusion, the proposed technique can be applied for many of our future activation analyses with high accuracy without having to rely on the availability of comparator standards.

7.1 OVERVIEW

Neutron activation analysis (NAA) is among the most sensitive tools used in identification and quantitative elemental analysis. NAA is a rapidly spreading analyzing technique applied extensively in the field of geological and biological studies. Basically, a sample is irradiated by thermal neutron and becomes radioactive. Then the activated nucleus decays according with a unique half life by emitting gamma quanta with specific energies into a more stable configuration. The intensity of the characteristic gamma ray lines in the spectrum, which is proportional to the elemental concentration, were measured, and used for quantitative identification of the element of interest.

The quantitative measurement of NAA has been done through comparative method; usually carried out by comparing the intensity of the gamma ray line in the spectrum of an irradiated unknown sample with that gamma ray line from known chemical standard. In this method, no detail information of the neutron flux, φ at the reactor irradiation site or of the nuclear data for the isotope concerned is required. The drawback of this method includes the difficulty in maintaining the stability of chemical standard which will contributes to additional uncertainty. Moreover, it is too cumbersome in the case with a large number of elements in one sample have to be determined because these processes are time consuming.

This work presents an absolute method by directly determining concentration in unknown samples. This method demands accurate and precise nuclear data for the isotope concerned such as atomic weight, cross section and isotopic abundance. The absolute NAA method also requires determination of parameters experimentally[1] such as photopeak efficiency of the γ detector (εγ), deviation of the epithermal flux distribution from the ideal 1/E distribution, (α), thermal to epithermal neutron flux ratio, (*f*), thermal and epithermal neutron flux (φ_{th}, φ_{epi}).

7.2 THEORETICAL CONCEPTS

In absolute method, the mass of an element, m (μg/g) derived from the activity of its isotope can be calculated from a neutron activation analysis experiment without a standard [2] using the following equation:

$$m = \frac{N_p M}{N_A \theta P_\gamma R \varepsilon_\gamma SDC}$$

where Np is the net peak area and R is the reaction rate per target nuclei, S, D, and C are the correction factors for saturation during irradiation, decay between irradiation and γ counting, and decay during counting, respectively. M is the atomic weight, N_A is Avogadro's number, θ is the natural isotopic abundance, Pγ is the absolute γ-ray emission probability, εγ is the photopeak efficiency of the γ detector and m is the mass of the target element (μg/g).

The reaction rate per target nuclei, R, of a sample irradiated by reactor neutrons can be described according to the HOGDHAL convention in terms of neutron flux and nuclear data parameters [3]. The neutron spectrum in a thermal reactor is the sum of thermal and epithermal neutrons. In the practical situation, the modified reaction rate including $1/E^{1+\alpha}$ neutron spectrum can be written as

$$R = R_{th} + R_e = \varphi_{th}\,\sigma_o + \varphi_e\, I_{o\,(\alpha)}$$

where $R_{th} = (\varphi th\ \sigma_o)$ is the reaction rate induced by pure thermal neutrons and $R_e = (\varphi_e\ I_o\ (\alpha))$ is the reaction rate induced by epithermal neutrons. φ_{th} is the thermal neutron flux and φ_e is the epithermal neutron flux. σ_o is the thermal neutron capture cross section at 2200 ms^{-1} and $I_o(\alpha)$ is the resonance integral for a $1/E^{1+\alpha}$ neutron spectrum. Parameter α measures the non-ideality of the epithermal neutron flux distribution from the ideal 1/E neutron spectrum distribution.

7.2.1 DETERMINATION OF α AND f

The deviation of the epithermal flux distribution, α, is fundamental as the correction of resonance integrals I_o, i.e., the conversion of I_o values to $I_o(\alpha)$ values for its use in actual $1/E^{1+\alpha}$ epithermal neutron spectrum. The parameter α can be determined by cadmium ratio multimonitor method, cadmium covered multi monitor methods or bare multimonitor methods [4]. In this work, the parameter α was determined using cadmium ratio multimonitor method which improves the estimates of the uncertainty.

Experimental determination of α, using the cadmium ratio method involves irradiating two or more monitors summarized in Table 1with and without cadmium cover and subsequently measuring on a high purity HPGe detector with known detection efficiency. If all the monitors have σ (*v*) ~1/ dependence up to ~1.5 eV, α can be obtained as slope (-α) of the straight line [5] by plotting graph of Equation (7.3):

$$\log \frac{(Er_{,i})^{-\alpha}}{(F_{cd,i} R_{cd,i}^{-1}) Q_{o,i(\alpha)}} \quad \text{versus} \quad \log \text{Er,i,} \tag{7.3}$$

where *i* denotes isotope1, 2,..., N.

$E_{r,i}$ - isotope effective resonance energy.

R_{cd} - ratio of the specific count rates of the samples irradiated without and with a cadmium cover, respectively.

F_{cd} - correction factor for Cd- transmission of epithermal neutron

with $Q_{0,\,i\,(\alpha)}$ is the ratio of resonance integral to cross section at 2200 ms^{-1} neutron velocity considered the correction parameter and shown in Equation (7.6):

$$Q_{o,i\,(\alpha)} = \frac{Q_{o,i} - 0.429}{(E_{r,i})^{\alpha}} + \frac{0.429}{(2\alpha + 1)(0.55)^{\alpha}}$$

The parameter *f*, ratio of the thermal-to-epithermal neutron flux can determine using Cd-ratio measurement from gold monitor with cadmium cover [4] as in Equation (7.5).

$$f = (F_{cd}\, R_{cd} - 1)\, Q_{o\,(\alpha)}$$

Table 7.1. Monitors and relevant nuclear data require in the work. Data for Au, Co, Zn, Zr and Mo were taken from compile data by DE CORTE [6]

Monitor	*Er, eV*	*Qo*	*Half life*	*E, keV*	*Fcd*
Au-197	5.65	15.71±	2.695	411.8	0.991
Co-59	± 7.1	1.8	± 0.1d	1173.2	1
Zr-94	136	1.993	5.271	756.7	1
Zr-96	± 5.1	± 2.7	± 0.02y	657.9	1
Zn-64	6260	4.61±	64.03	1115.5	1
Mo-98	± 4.0	2.0	± 0.01d	140.5	1
	338	231 ±	72.1 ±		
	± 2.1	1.5	1.0min		
	2560	1.908	244.0		
	± 10	± 4.9	± 0.08d		
	241	53.1±	6.02 ±		
	± 20	6.3	0.3h		

7.3 EXPERIMENTAL RESULTS

7.3.1 CALIBRATION OF GAMMA RAY SPECTROMETER

In this experiment a HPGe detector, Canberra GC3018 detector with Genie 2000 software was used to measure the energies and intensities of gamma rays from samples. The efficiency calibrations were performed by using gamma-ray standard point sources namely Ba-133, Cs-137, Co-60, Am-241, Eu-152 and Na-22. The standard sources were counted independently at 2cm and 10cm geometries from the end cap of the detector.

7.3.2 CHARACTERIZATION OF REACTOR NEUTRON SPECTRUM PARAMETERS (φ_{th}, φ_{epi}, f, and α)

The TRIGA Mark II reactor of Malaysia Nuclear Agency (MNA) is a pool type reactor, with a thermal power capacity of 1 Megawatt, an average neutron flux of 1.2×10^{12} $cm^{-2}s^{-1}$. Light water is used as moderator to dissipate heat, high purity graphite as reflector and Uranium-235 as nuclear fuel. All the irradiation for the work has been performed in the rotary rack housed within the graphite reflector which contains of 40 locations for sample irradiation.

Thermal, epithermal neutron fluxes, as well as α and f were determined in irradiation position 27 of rotary rack reactor by applying the cadmium ratio method. The following set of monitors was used: Al-Au (IRMM, Belgium), purity: 0.1124% Au, pure: 99.9845%, diam. 0.0508 mm; Zr, purity: 99.7329%; Zn, purity: 99.9872%, thickness: 0.254mm; Co, purity 99.9133% and Mo, 99.945, diam. 0.0508mm. The monitors were roughly cut into equal pieces and rolled spirally. The weighing of the monitors was carefully carried out using an electrical balance.

Two set of this five monitors were prepared. One set was irradiated without cadmium cover and another set for irradiation under a cadmium cover (1 mm thickness) for undergoing epithermal irradiations. For bare irradiation, the 5 monitors were packed together in a standard polyethylene vial. The irradiation time was one hour in stationery mode at maximum thermal power of 750kW.

After one day of cooling, all the monitors were counted using the calibrated HPGe detector with counting time of 5-60 minutes in order to obtain suitable counts. The thermal and epithermal neutron fluxes (φ_{th}, φ_{epi}) and flux ratio, f was determined using gold monitor with the similar procedure.

7.3.3 ANALYSIS OF RESULTS

In order to evaluate the accuracy and precision of the absolute method, this technique was applied to two certified reference materials (CRM): IAEA Soil-1 and Soil-7. The samples thus prepared were enclosed and sealed in polyethylene vials. Approximately 30 mg of each CRM samples were irradiated for one hour in stationery mode at irradiation position 27 of rotary rack facilities. After irradiation, the samples were cooled for 1 and 3 days, respectively; then, their activities were measured.

7.4 RESULTS AND DISCUSSION

The resulting full energy peak detection efficiency of both geometries was plotted against gamma ray energy in logarithmic scale represented in Figures 7.1 and 7.2, respectively. For an ideal calibration source and procedure, the efficiency curve revealed a smooth function of energy at spectral range from 59.88 to 1332.51 keV. The measured efficiencies were fitted to appropriate equation proposed by Ahmad and Gray [2], which allowed the interpolations at the particularly energies of interest.

The result of reactor neutron parameter (f, α, φ_{th}, φ_{epi}) at irradiation position 27 of rotary rack TRIGA reactor is reported in Table 7.2. The α measurements were performed using Au, Co, Zn, Zr and Mo. The measured f value shows that irradiation position at 27 is well thermalized with about 95% thermal neutrons. The negative flux deviation parameter, α predisposes the TRIGA reactor as an “under-moderated” reactor type.

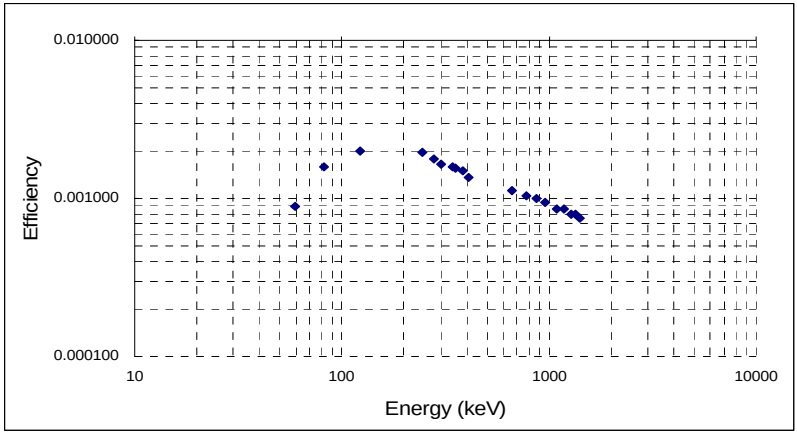

Figure 7.1. Efficiency curve at a distance of 12cm from the Canberra GC3018 detector.

Therefore, the facility is under safe condition for reactor physics experiments and ideal for NAA application. Neutron fluxes were determined from reaction rates of Au with and without cadmium cover. The result of reactor neutron parameters (f, α, φ_{th}, φ_{epi}) is required in the calculation of reaction rates for any irradiated element.

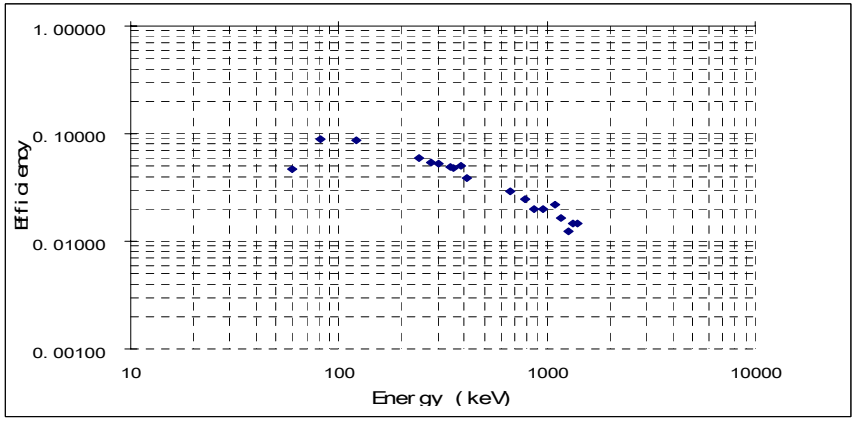

Figure 7.2. Efficiency curve at a distance of 2cm from the Canberra GC3018 detector.

Table 7.2. Results for *f*, α, thermal and epithermal neutron fluxes at irradiation site 27 of rotary rack TRIGA reactor obtained by Cd ratio multi- monitor method.

Reactor Irradiation Position	*f*	α	φ_{th} $10^{12}cm^{-2}s^{-1}$	φ_{epi} $10^{10}cm^{-2}s^{-1}$
27	20.13 ±0.78	-0.0464 ±0.0048	2.4475 ±0.0991	12.1597 ±0.9635

The results of the analyses are presented in Tables 7.3 and 7.4 for IAEA Soil-1 and Soil-7, respectively. The elements concentrations were determined by measuring the reaction rates of the product radionuclide from Equations (7.1) and (7.2). The deviation between experiment and certified values issued by International Atomic Energy Agency is expressed in the ratio between experimental values and certified values. The results obtained agree reasonably well with the certified values. The accuracy of the results was statistically evaluated using z-score. Based on NAA absolute method, most of the analytical results showed z-score within $0 < |Z| < 2$ and hence are accepted with precision. The accuracy of the analytical result for each element in Soil-1 and Soil -7 depends obviously on uncertainties of the involved nuclear properties and thus vary from element to element. However, the accuracy of the absolute method adopted in the analysis of the soil-1 and soil-7 are as good as that of the comparative method.

Table 7.3. Comparison of concentration element in IAEA Soil-1 result obtained from absolute method to certified value at irradiation position 27

Elements	*Certified Value*		*Irradiation Position 27*			
	µg/ g	1σ(%)	µg/ g	1σ(%)	Exp/cert.	|Z-score|
As	27.5	11	22.1	14	0.80	1.29
La	52.6	6	46.9	8	0.89	1.13
Mn	3460	5	3151.4	7	0.91	1.18
Na	1720	6	1893.8	6	1.10	1.17
Sc	17.3	6	17.9	32	1.03	0.10
Sm	9.25	6	9.7	16	1.05	0.30
Ga	24.0	22	25.6	16	1.06	0.24
K	14500	15	12406.1	8	0.86	0.89
Hf	4.2	14	3.1	39	0.74	0.81
Ba	639	8	482.0	39	0.75	0.81
Cr	104	9	113.6	17	1.09	0.44
Sb	1.31	9	1.1	22	0.80	1.01
Th	14	7	12.8	13	0.91	0.61
Co	19.8	8	20.8	17	1.05	0.26
Ta	1.58	37	1.7	60	1.09	0.12
Zn	223	5	235.1	33	1.05	0.16
Yb	3.42	19	3.0	38	0.88	0.30
U	4.02	8	4.1	28	1.03	0.09
Cs	7.0	13	8.0	84	1.14	0.15

Table 7.4. Comparison of concentration element in IAEA Soil-7 obtained from absolute method to certified value at irradiation position 27

Elements	*Certified Value*		*Irradiation Position 27*			
	µg/ g	1σ(%)	µg/ g	1σ(%)	Exp/cert.	|Z-score|
As	13.4	6	11.3	19	0.84	0.91
La	28	4	26.9	29	0.96	0.14
Mn	631	4	645.1	11	1.02	0.19
Na	2400	4	2651.2	6	1.10	1.39
Sc	8.3	13	6.7	9	0.80	1.34
Sm	5.1	7	6.3	20	1.24	0.93
Ga	10	20	11.3	25	1.13	0.38
K	12100	6	12236.7	8	1.01	0.11
Hf	5.1	7	4.4	27	0.86	0.59
Ba	159	20	180.3	83	1.13	0.14
Sb	1.7	12	2.0	96	1.16	0.15
Ce	61	11	60.4	25	0.98	0.04
Th	8.2	13	6.6	18	0.80	0.99
Fe	25700	2	21423.4	16	0.83	1.23
Co	8.9	10	11.3	23	1.27	0.88
Yb	2.4	15	3.0	50	1.23	0.40
U	2.6	21	2.5	31	0.95	0.13
Tb	0.6	33	0.5	76	0.86	0.18
Cs	5.4	14	4.9	33	0.91	0.26
Ca	163000	5	183959.2	19	1.13	0.59

7.5 CONCLUSION

As a summary, the use of absolute counting has been shown to yield good agreement between the calculated concentration and the certified values. This method had been implemented successfully and provides the accuracy which may overcome the conventional comparative method. Presently, the nuclear data are much more accurate and reliable, which should rekindle a new interest applied in this absolute technique. The expected outcome

from this research is a viable approach of doing NAA which no longer rely on the use of multi standard materials that can be subjected to many errors. Moreover, this technique is a potential method that can be effectively used on routine basis for the NAA analysis especially for a large numbers of samples.

7.6 REFERENCES

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