

Neutron Characteristic and Related k_0 Parameters in TRIGA Mark II Research Reactor after Core Reconfiguration

A. Yavar^a, S. B. Sarmani^a, K. S. Khoo^a

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

Abstract: The thermal to epithermal neutron flux ratio f , epithermal neutron flux shape factor α , thermal neutron flux ϕ_{th} , and epithermal neutron flux ϕ_{epi} in the irradiation channels of the Malaysian Nuclear Agency research reactor were re-determined after a core reconfiguration to guarantee accuracy in applications of the k_0 -neutron activation analysis (k_0 -NAA) method. The f and α parameters were determined using bare bi-isotopic monitor and bare triple-monitor methods, respectively, and Au and Zr monitors were used in 30 rotary rack (RR) irradiation channels. It was found that f was between 14.82 and 24.67 and that α ranged from 0.0011 to 0.0672. Average values of ϕ_{th} and ϕ_{epi} were determined to be $(2.15 \pm 0.25) \times 10^{12}$ and $(1.20 \pm 0.20) \times 10^{11} \text{ cm}^{-2} \text{ s}^{-1}$, respectively. The results were compared to those of previous studies at the MNA reactor and those of similar reactors in other countries. The accuracy of the method was evaluated by analysing ERM-DB001 human hair reference materials. The results showed an adequate level of consistency.

Keywords: k_0 -NAA, Neutron flux, k_0 parameters, TRIGA Mark II research reactor.

Corresponding author:

Email: khoo@ukm.edu.my, Address: School of Applied Physics, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor, Malaysia

I. INTRODUCTION

The k_0 -NAA method introduced in the 1970s as a very useful and applicable method for multi-elemental analysis of biological, geological, environmental and high purity materials using reactor neutrons [1-14]. The k_0 -NAA method needs the accurate characterization of irradiation and counting facilities in addition to the use of composite nuclear constants known as k_0 -factors. The k_0 -factors, which are independent of irradiation and measurement conditions, are tabulated and published in literature as generally useful nuclear parameters [15, 16]. The k_0 -NAA method was successfully developed by the Høgdahl convention [17]. Its application is restricted to (n, γ) cross sections that follow the $1/v$ law in the thermal neutron energy region (i.e., up to ~ 1.5 eV). According to the Høgdahl convention, neutron flux spectrum parameters such as thermal to epithermal neutron flux ratio (f) and epithermal neutron flux shape factor (α) are necessary to determine the elemental concentrations in an unknown sample when using the k_0 -NAA method [10, 14]. In our work f and α were determined by the bare bi-isotopic monitor and bare triple monitor methods, respectively [1-14].

The Malaysian Nuclear Agency (MNA) research reactor, a TRIGA Mark II swimming-pool type reactor, was commissioned in 1982, and the most recent core reconfiguration was on 27 August 2013. As the f , α , ϕ_{th} and ϕ_{epi} parameters are dependent on the reactor configuration and irradiation positions, it is essential to re-determine these parameters before utilizing the k_0 -neutron activation analysis (k_0 -NAA) method [18-20]. These parameters were thus re-determined at 30 rotary rack (RR) irradiation channels of the MNA TRIGA Mark II reactor (Fig. 1), and the findings were compared with published values from earlier MNA reactor studies.

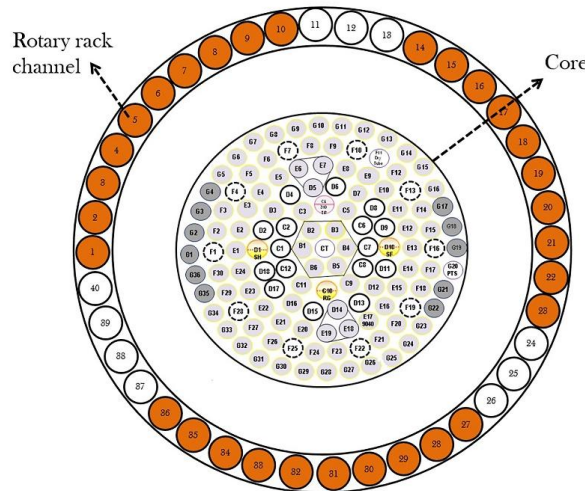


Fig. 1. Arrangement of the 40 rotary rack channels around the core of MNA research reactor (30 RR channels used in present study are marked).

II. Experimental Procedure

2.1. Efficiency calibration of HPGe detector

The γ - spectrometry measurements were implemented with an HPGe detector coupled with Canberra Accuspec multichannel analyzer (MCA); the computer code Gamma Acquisition Analysis was performed for peak area evaluation. Full energy peak efficiency calibration of the detector was carried out using ^{241}Am , ^{133}Ba , ^{109}Cd , ^{57}Co , ^{137}Cs and ^{60}Co multi-nuclide sources placed at the reference position 15.8 cm from the detector where true coincidence effects are negligible. Fig. 2 illustrates the peak detection efficiency of the HPGe detector plotted in logarithmic scale. The energy range was from 58.91 keV to 1332.58 keV [3, 20-22].

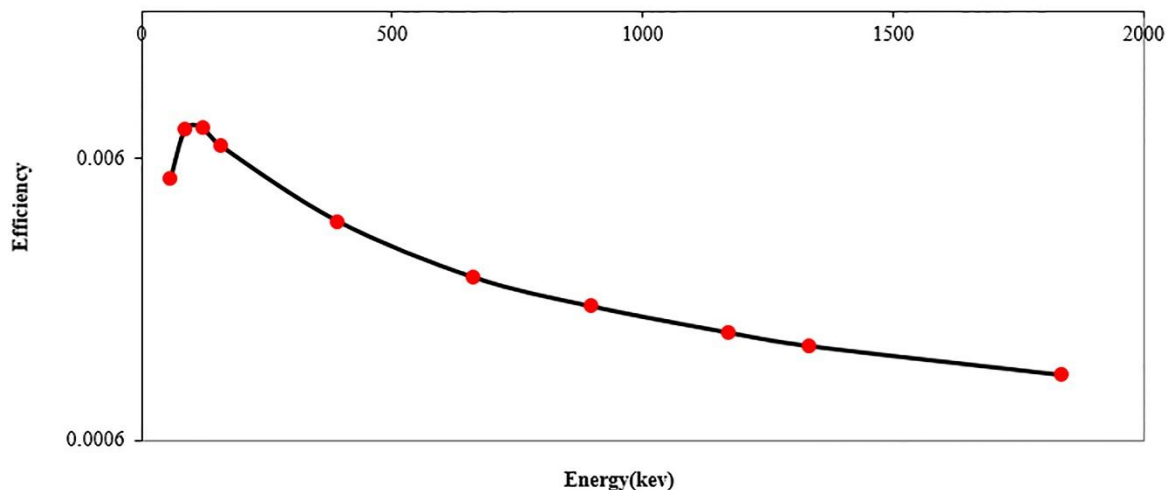


Fig. 2. Efficiency curve for the HPGe detector, “ref” = point geometry at 15.8 cm source detector distance.

2.2. Characterization of MNA research reactor spectrum

Au and Zr monitors were utilized to determine the f , α , ϕ_{th} and ϕ_{epi} parameters. The monitors were made of Al-0.1% Au alloy wire (IRMM-527a, diameter 1 mm, length 10 mm) and Zr foils (IRMM, 99.9%, 125 μm thick), respectively. The monitors were cut and carefully weighed so that the size range for Au monitors were 17.8 to 26.0 mg while for the Zr monitors the size range were 12.5 to 27.5 mg. The vials were chosen with 1 cm diameter and 1 cm length. The monitors were heat sealed inside the polyethylene vials and were packed in heat resistant plastic so that each vial included one Au monitor and one Zr one. In order to evaluation of results, ERM-DB001-human hair as certified reference materials (CRMs) were prepared. The CRMs were put in stove at 90 $^{\circ}\text{C}$ for 2 h to dehumidify, then the CRMs were carefully weighed as range were 100.1 to 110.2 mg, and heat sealed them in vials separately. Three vials involve one monitors vial, one CRMs vial and one blank vial (for

omit the background radiation) packed by two layers of heat resistance plastic. This package prepared for irradiation at 3 RR channels and 22 RR channels rest were used only for irradiation of the monitors. Allpacks were irradiated for 6 hours in thirtyRR irradiation channels (as marked in Fig.1) of the MNA research reactor. Since the half-lives of radionuclides of ^{198}Au and $^{97}\text{Zr}/^{97\text{m}}\text{Nb}$ are short, both monitors were counted for about 5 minutes after one day decay time. Also we counted CRMs after one day decay time for finding the short half-life radionuclides, after 7 days for medium half-life radionuclides and after 21 days for finding the long half-life radionuclides. The irradiated zirconium was counted for measurement of ^{95}Zr by 15 minutes counting time after 3 days decay time. Three gamma-lines were used in the estimation of f and α : 411.8 keV of ^{198}Au , 743.4 keV of $^{97}\text{Zr}/^{97\text{m}}\text{Nb}$ and the sum of the two peaks (724.2 and 756.7 keV) of ^{95}Zr [3, 20-22].

2.3. Result assessment

The accuracy of the analytical measurements was estimated via the z-score as follows:

$$z = \frac{|C_i - C_{ref,i}|}{\sqrt{\sigma_i^2 + \sigma_{ref,i}^2}} \tag{1}$$

where C_i is the concentration of element i in the sample; $C_{ref,i}$ is the concentration of the certified value for element i ; σ_i is uncertainty of the concentration of element i in the sample; $\sigma_{ref,i}$ is uncertainty of the certified respective consensus value for element i [23].

III. Results And Discussion

Table 1 presents the neutron flux parameters at the 30 RR irradiation channels of the MNA research reactor. The values of f and α were determined using the bare bi-isotopic monitor and bare triple-monitor methods, respectively.

TABLE 1. The results of f , α , ϕ_{th} and ϕ_{epi} at MNA research reactor (our work)

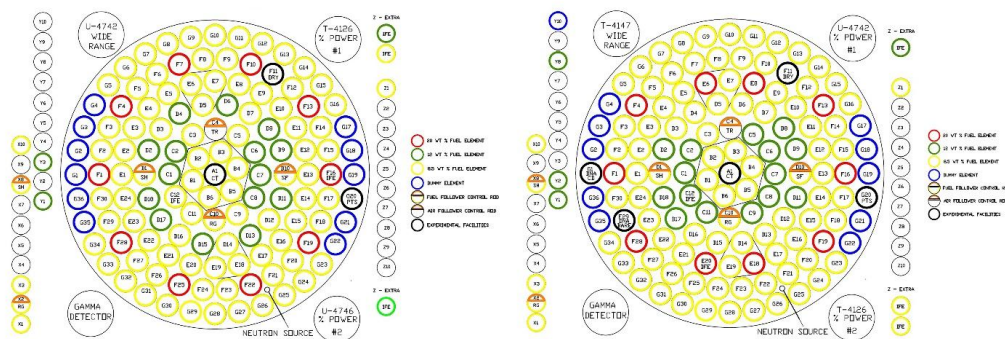
Channel number	f	α	ϕ_{th} (cm ² .s ⁻¹)	ϕ_{epi} (cm ² .s ⁻¹)
1.	24.67	-0.0160	1.98E+12	8.01E+10
2.	17.20	0.0426	2.04E+12	1.19E+11
3.	16.89	0.0440	2.04E+12	1.21E+11
4.	16.64	0.0495	2.00E+12	1.20E+11
5.	16.52	0.0465	2.05E+12	1.24E+11
6.	18.17	0.0175	2.08E+12	1.15E+11
7.	18.64	0.0337	2.12E+12	1.14E+11
8.	16.92	0.0443	1.95E+12	1.15E+11
9.	16.99	0.0478	2.05E+12	1.21E+11
10.	18.30	0.0392	2.09E+12	1.14E+11
11.	16.00	0.0544	2.56E+12	1.60E+11
12.	20.18	0.0133	2.42E+12	1.20E+11
13.	16.76	0.0457	2.55E+12	1.52E+11
14.	16.53	0.0468	2.49E+12	1.51E+11
15.	18.19	0.0335	2.60E+12	1.43E+11
16.	16.76	0.0672	2.46E+12	1.47E+11
17.	21.45	-0.0011	2.48E+12	1.15E+11
18.	14.82	0.0535	2.36E+12	1.59E+11
19.	18.15	0.0345	2.59E+12	1.43E+11
20.	21.96	-0.0042	2.37E+12	1.08E+11
21.	18.32	0.0408	1.94E+12	1.06E+11
22.	19.69	0.0297	1.97E+12	1.00E+11
23.	19.48	0.0336	1.99E+12	1.02E+11
24.	21.38	0.0090	1.82E+12	8.53E+10
25.	15.73	0.0548	1.85E+12	1.18E+11
26.	17.30	0.0515	1.93E+12	1.12E+11
27.	18.46	0.0418	1.96E+12	1.06E+11
28.	17.66	0.0545	1.85E+12	1.05E+11
29.	18.42	0.0430	2.00E+12	1.09E+11
30.	19.24	0.0391	1.98E+12	1.03E+11
Average	18.25	0.0364	2.15E+12	1.20E+11
Standard deviation	2.10	0.0163	2.55E+11	2.02E+10

The MNA research reactor core was reconfigured for the 11th time on 23 August 2001 to contain 111 fuel elements. The 12th core reconfiguration was carried out on 5 July 2006. After the 13th core reconfiguration on 20 April 2009, there were 112 fuel elements in the core. The 14th and 15th core reconfigurations with 111 fuel elements were carried out on 11 March 2013 and 27 August 2013, respectively. Previous studies by Wee

[24], Khoo [25], and Yavar[20] determined the neutron flux and related k_0 parameters of the MNA research reactor in 2003, 2005, and 2009, respectively (Table 2). The studies of Wee [24] and Khoo [25] were based on the same core configuration (the 11th), while the study of Yavar[20] was based on the 13th core reconfiguration. The differences in the average values of f shown in Table 2 are thus expected as the reactor configurations differed. In particular, the differences may be due to differences in the neutron flux distributions; in our study, since the reactor was rotating during the activation of monitors, the neutron flux distribution was homogeneous. Moreover, the variation in the f values may have arisen from differences in the shape of the RR container. If the longitudinal axis of the container is not precisely parallel to the longitudinal axis of the fuel elements, then there is some space between the container and inner surface of the irradiation tube [25]. In addition, as shown in Fig. 3, there were 111 fuel elements in the reactor core in our work (Fig. 3b), but there were 112 fuel elements in the previous study of Yavar [20]. A larger number of fuel elements in the core will increase the thermal neutron flux, which is in turn correlated with the f parameter.

TABLE 2. Average and standard deviation of f , α , ϕ_{th} and ϕ_{epi} in several TRIGA Mark II reactors on MNA research reactor [20, 24, 25].

Study by	Date of experiment	f	α	ϕ_{th} $\times 10^{12} \text{ (cm}^{-2} \cdot \text{s}^{-1})$	ϕ_{epi} $\times 10^{11} \text{ (cm}^{-2} \cdot \text{s}^{-1})$
Wee [24]	2003	17.2±0.9	0.016±0.005	2.29±0.09	1.33
Khoo[25]	2005	33.55±11.2	-0.087±0.046	2.03±0.27	0.61±0.45
Yavar[20]	2009	39.67±6.57	-0.102±0.033	2.06±0.02	0.52±0.09
Present study	2014	18.25±2.10	0.0364±0.0163	2.15±0.25	1.20±0.20



A: Core configuration of MNA research reactor on 11 March 2013 for 14th time. (Number of fuel elements in the core is 112)

B: Core configuration of MNA research reactor on 27 August 2013 for 15th time. (Number of fuel elements in the core is 111)

Fig. 3. Core configurations of the MNA research reactor as os (A) March 2013 and (B) August 2013

The average value of α for this study was found to be 0.0364 ± 0.0163 . The value of α depends on the reactor configuration and increases with increasing distance from the core. The negative values of α point out to a poor thermalisation and a higher thermalisation that would associate with a positive α value [3, 13]. We found that there was a wide variation in the thermalisation of the reactor, as α was negative for 3 channels (indicating poor thermalisation) and positive for 27 channels.

The average value of ϕ_{th} was found to be $(2.15 \pm 0.25) \times 10^{12} \text{ cm}^{-2} \text{ s}^{-1}$, while the average value of ϕ_{epi} was $(1.20 \pm 0.20) \times 10^{11} \text{ cm}^{-2} \text{ s}^{-1}$. The low standard deviations of ϕ_{th} and ϕ_{epi} are consistent with the homogeneous neutron flux. Our value of ϕ_{th} was in satisfactory consistent with those of previous studies at the MNA research reactor.

Table 2 shows that although the MNA research reactor had the same core configuration, there was some variation in the findings presented by Wee [24] and Khoo [25], i.e., the first and second studies. Fuel burn up will have a large effect on the parameter values, and it must also be noted that the positions of the control rods can have a significant impact. Furthermore, increasing the number of irradiation channels will improve accuracy. The first study was based on one RR channel with a non-rotary system (resulting in an inhomogeneous flux distribution), while the second study, also with a non-rotary system, used 20 RR channels to obtain more accurate results. The third study, that of Yavar [20] utilized 4 RR channels with a rotary system (resulting in a homogeneous flux distribution). In the present work, 30 RR channels with a rotary system were used to obtain adequately accurate results.

To check the experimental parameters obtained in this work, certified reference materials (CRMs) of ERM-DB001 human hair were analysed using 3 RR irradiation channels (Table 3). The elemental concentrations were calculated using the Høgdahl convention [6, 10, 14], and the Cu, Se, and Zn concentrations were in very good agreement with the certified values. In addition, the z-score measurement results in Table 3 validate the present work.

TABLE 3. Comparison of obtained Zn concentration by k_0 -INAA with the certified value in ERM-DB001-human hair

Element	This work (mg/kg)	Certified value (mg/kg)	z-score
Cu	46.58±2.58	33±4	2.85
Se	4.71±0.98	3.24±0.24	1.45
Zn	206.69±3.06	209±12	0.19

Based on the z-score (Eq. 1), the result is classified as anticipated if $z < 2$. If $2 < z < 3$, the quality of measurement is 'alarming', and $z > 3$ classifies the results as 'out of control' [20, 26]. Overall, our findings were validated ($z < 2$). The z-score for Cu was within the alarming range but acceptable.

IV. Conclusion

The core reconfiguration of the MNA research reactor in August 2013 necessitated the redetermination of the neutron flux and related k_0 parameters. This was achieved using Au and Zr monitors in 30 RR irradiation channels. We found that the fluctuation in the neutron flux in the irradiation channels was quite regular. Although there was a good level of consistency with the results of other studies of the MNA research reactor, some deviation was found. This deviation may have been caused by differences in the neutron flux distribution, the number of fuel elements in the core, and the positions of the control rods. The accuracy of the redetermined parameters was evaluated by analysing ERM-DB001 human hair, and the results showed an acceptable level of consistency.

Acknowledgments

The authors are grateful to Ministry of Education Malaysia for financial support (FRGS/2/2013/ST02/UKM/02/1).

References

- [1] De Corte, F., Speecke, A., Hoste, J., 1969. Reactor neutron activation analysis by a triple comparator method. *Journal of Radioanal. Nucl. Chem.* 3, 205.
- [2] De Corte, F., Simonits, A., DeWispelaere, A., Hoste, J., 1987. Accuracy and applicability of the k_0 -standardization method. *Journal of Radioanal. Nucl. Chem.* 113, 145.
- [3] De Corte, F., Simonits, A., Bellemans, F., Freitas, M.C., Jovanovic, S., Smodis, B., Erdtmann, G., Petri, H., De Wispelaere, A., 1993. Recent advances in the k_0 -standardization of neutron activation analysis: Extensions, applications, prospects. *Journal of Radioanal. Nucl. Chem.* 169, 125.
- [4] Simonits, A., De Corte, F., De Wispelaere, A., Hoste, J., 1987. Nuclear data measurements for zirconium isotopes used for activation analysis and neutron metrology. *Journal of Radioanal. Nucl. Chem.* 113, 187.
- [5] Simonits, A., De Corte, F., Hoste, J., 1975. Single-comparator methods in reactor neutron activation analysis. *Journal of Radioanal. Chem.* 24, 31.
- [6] Simonits, A., De Corte, F., Moens, L., Hoste, J., 1982. Status and recent developments in the k_0 -standardization method. *Journal of Radioanal. Nucl. Chem.* 72, 209.
- [7] De Corte, F., 1992. Problems and solutions in the standardization of reactor neutron activation analysis. *Journal of Radioanal. Nucl. Chem.* 160, 63.
- [8] De Corte, F., 1994. Neutron activation analysis: an old faithful to cherish. *ActaPhysicaHungarica* 75, 189.
- [9] De Corte, F., 2000. k_0 and comparator NAA: Influences and interactions. *Journal of Radioanal. Nucl. Chem.* 245, 157.
- [10] De Corte, F., 2001. The standardization of standardless NAA. *Journal of Radioanal. Nucl. Chem.* 248(1): 13.
- [11] De Corte, F., Hammami, K., Moens, L., Simonits, A., De Wispelaere, A., Hoste, J., 1981. The accuracy and precision of the experimental α -determination in the $1/E^{1+\alpha}$ epithermal reactor-neutron spectrum. *Journal of Radioanal. Nucl. Chem.* 62, 209.
- [12] De Corte, F., Moens, L., Jovanovic, S., Simonits, A., De Wispelaere, A., 1986. Applicability of the $1/E^{1+\alpha}$ epithermal spectrum representation and the effective resonance energy E_r in NAA. *Journal of Radioanal. Nucl. Chem.* 102, 37.
- [13] De Corte, F., Moens, L., Simonits, A., De Wispelaere, A., Hoste, J., 1980. Instantaneous α -determination without Cd-cover in the $1/E^{1+\alpha}$ epithermal neutron spectrum. *Journal of Radioanal. Nucl. Chem.* 58, 401.
- [14] Lin, X., Baumgärtner, F., Li, X., 1997. The program MULTINAA for various standardization methods in neutron activation analysis. *Journal of Radioanal. Nucl. Chem.* 215, 179.
- [15] De Corte, F., Simonits, A., 2003. Recommended nuclear data for use in the k_0 standardization of neutron activation analysis. *Atomic Data and Nuclear Data Tables* 85, 47.
- [16] Moens, L., De Corte, F., De Wispelaere, A., Hoste, J., Simonits, A., Elek, A., Szabo, E., 1984. k_0 -measurements and related nuclear data compilation for (n, γ) reactor neutron activation analysis. *Journal of Radioanal. Nucl. Chem.* 82, 385.
- [17] Høgdahl, O. T., 1962. Neutron absorption in pile neutron activation analysis, Technical Report. Reactor Division, Brookhaven National Laboratory (BNL), Upton, New York, USA.

- [18] De Wispelaere, A., De Corte, F., 2003. Recalibration of the irradiation facilities in the Thetis reactor, with an examination of the α versus E behavior in the keV neutron energy range. *Journal of Radioanal. Nucl. Chem.* 257, 519.
- [19] Mustra, C., Freitas, M., Almeida, S., 2003. Neutron flux and associated k₀ parameters in the RPI after the last configuration change. *Journal of Radioanal. Nucl. Chem.* 257, 539.
- [20] Yavar, A.R., Sarmani, S.B., Wood, A.K., Fadzil, S.M., Masood, Z. & Khoo, K.S. 2011. Neutron flux parameters for k₀-NAA method at the Malaysian Nuclear Agency research reactor after core reconfiguration. *Radiation Measurements Journal.* 46(2): 219-223.
- [21] Alghem, L., Ramdhane, M., Khaled, S., Akhal, T., 2006. The development and application of k₀-standardization method of neutron activation analysis at Es-Salam research reactor. *Nucl. Instru. Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment.* 556, 386.
- [22] Wasim, M., Zaidi, J., Arif, M., Fatima, I., 2008. Development and implementation of k₀-INAA standardization at PINSTECH. *Journal of Radioanal. Nucl. Chem.* 277, 525.
- [23] Yavar, A.R., Khalafi, H., Kasesaz, Y., Sarmani, S.B., Wood, A.K. & Khoo, K.S. 2012. Verification of MCNP simulation for neutron parameters measurement at Malaysian nuclear agency research reactor. *Applied Radiation and Isotopes.* 70(10): 2488-2493.
- [24] Wee, B.S., Dung, H.M., Wood, A.K., Salim, N.A., Elias, M.S., 2006. Testing the applicability of the k₀-NAA method at the MINT's TRIGA MARK II reactor. *Nucl. Instru. Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* 564, 716.
- [25] Khoo, K., Sarmani, S., Abugassa, I., 2007. Determination of thermal to epithermal neutron flux ratio (f), epithermal neutron flux shape factor (α) and comparator factor (F_c) in the Triga Mark II reactor, Malaysia. *Journal of Radioanal. Nucl. Chem.* 271, 419.
- [26] Bode, P., Dijk, CP., 1997. Operational management of results in INAA utilizing a versatile system of control charts. *Journal Radioanalytical and Nuclear Chemistry* 215(1), 87-94.