Determination of TRIGA 2000 reactor parameters for NAA absolute methods

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Abstract. Neutron Activation Analysis (NAA) is a nuclear analytical technique for elemental analysis that has been widely applied in various fields such as environment, industry, health and food. It has high specificity and sensitivity for the analysis of elements with very low concentrations up to the picogram order. In Indonesia, NAA technique was developed in BATAN that has a research reactor as a neutron source. Relative method of NAA that uses the multi-elements standard as a comparison is widely used in Indonesia. At present, BATAN is developing an absolute method k0-NAA that has several advantages such as faster and more economics due to the reduction of multi-elements standard dependence. BATAN k0 Utility (BAkUL) beta version software has also been developed for data processing of the absolute methods. k0-NAA requires reactor parameters (a and f) as well as detector parameters for data analysis. The parameter f is the ratio of the thermal neutron flux to the epithermal whereas a is the epithermal spectrum deviation parameter from the 1/E distribution. The aim of this study is to measure parameters a and f using the triple bare monitor method by irradiating simultaneously Au and Zr without Cd cover. Measurement of reactor TRIGA 2000 parameters (a and f) were carried out at the irradiation positions CT1-1 and G17-2, and the results for a and f parameters at the CT1-1 irradiation position were -0.100 and 8.22 respectively, while at the G17-2 position was -0.087 and 13.8. Each irradiation position has different a and f values so it is necessary to determine the reactor parameters for other irradiation positions. The measurement results a and f can be used for the next samples irradiation and as a database to support the application of the k0-NAA method in Center for Applied Nuclear Science and Technology (PSTNT) BATAN.

1. Introduction

Activation analysis is a method for the determination of elements based upon the conversion of stable nuclei to other, mostly radioactive nuclei via nuclear reactions, and measurement of the reaction products. In neutron activation analysis (NAA) the nuclear reactions occur via bombardding the material to be analyzed with neutrons. The reaction products to be measured are either the radiation, released nearly instantaneously upon neutron capture; or, if the resulting new nuclei are radioactive, the induced radioactivity by which they decay [1]. NAA is very useful as sensitive analytical technique for performing both qualitative and quantitative multi-elemental analysis of major, minor and traces components in variety of terrestrial samples and extra-terrestrial materials [2]. From the early 1960s, NAA was a quickly developing technique at the first generation of research reactors. Since then, NAA has become one of the most reliable methods for trace-element analysis [3]. In addition, because of its accuracy and reliability, NAA is generally recognized as the "referee method" of choice when new procedures are being developed or when other methods yield results that do not agree [2].
Generally, there are three techniques for elemental calculation using NAA which is relative, absolute and k0. Relative method calculates the element content where the induced activity of the unknown sample is compared to that of a standard of known concentration, irradiated and counted under identical conditions. The absolute method calculates the element content with the help of the neutron activation equation from nuclear constants of the nuclides involved and by additional physical parameters of the activation and measurement processes. Hence, the relative method is the simplest when only a small number of the matrix elements are to be determined. Therefore, in handling a large number of samples of different origins; biological, environmental, geological, etc., where, the use of relative method in its classical form makes tedious and laborious due to the time consuming invested in preparation, irradiation, measurements and recounting these standards spectra. In order to eliminate many of such difficulties many efforts have been done by researchers [4].

The k0 method was first introduced by Simonits in 1975, then developed and refined by Frans de Corte in 1987. k0-NAA is a single-comparator standardization method, in particular using 197Au, allowing accurate elemental analysis of any material, without the need to recalibrate for all elements when analysing new matrices or using new detectors [5]. This technique developed rapidly, especially for the European regions, pioneered by Hungary, Germany and the Netherlands. This k0-NAA technique has the advantage such as no sample contamination and no blank subtraction is needed (blank is negligible) [6]. In the Asian region, China is one of the most developed countries in developing the k0 method, followed by Vietnam. Quantitative determination is based on the equation (1):

\[ \rho_x = \frac{\left( \frac{N_p}{t_m} \right)}{A_{p,m}} \cdot \frac{1}{k_{o,m}(\alpha)} \cdot \frac{G_{f,k,m} \cdot f + G_{e,m} \cdot Q_{o,a}(\alpha)}{G_{f,v,a} \cdot f + G_{e,v,a} \cdot Q_{o,a}(\alpha)} \cdot \frac{e_{p,m}}{e_{p,a}} \cdot 10^6 \]  

(1)

The notations a and m respectively state the analyte and the flux monitor, and \( \rho_x \) is the concentration of analyte in mg/kg. Other notations are explained as follows:

- \( N_p \): the number of counts collected at the peak of full-energy, after being corrected for the lost pulse (including dead time detector and coincidence effect)
- \( t_m \): measurement time interval (seconds)
- \( S \): irradiation saturation factor = 1 – exp \((-\lambda t_{irr})\)
- \( D \): decay factor = exp \((-\lambda t_d)\)
- \( C \): counting factor (measurement) = \(\frac{1 - \exp(-\lambda t_m)}{\lambda t_m}\)
- \( A_{p} \): Specific activity \((\text{sec}^{-1} \text{g}^{-1})\)
- \( e_p \): detection efficiency from full energy peaks, including corrections for attenuation \(\gamma\)
- \( G_e \): epithermal self-shielding factor
- \( G_{th} \): thermal self-shielding factor
- \( Q_{o} \): the ratio of resonance integrals to thermal cross-sections (=\(I_0/S_0\))
- \( r \): flux monitor that has a known-value of \(Q_o\) and \(E_r\)

Equation (1) shows the reactor parameters \(f\) and \(\alpha\) which are characteristic of a research nuclear reactor. In elemental analysis using the k0-AAN method, reactor parameters such as \(f\) and \(\alpha\) are the important parameters of the analysis results. Therefore the determination of this parameters is essential. The parameter \(f\) is the ratio of the thermal neutron flux to the epithermal whereas \(\alpha\) is the epithermal spectrum deviation parameter from the 1 / \(E\) distribution [7]. In general, there are three methods for determining the \(\alpha\) parameter such as multi monitor method with Cd cover, Cd ratio for multiple monitors and triple bare monitor method.

Method 1 is used for epithermal irradiation facilities in the epithermal neutron activation analysis (ENAA) method while method 2 is used to monitor the parameter \(\alpha\) especially if the neutron flux characteristics are always stable at all times. The most suitable method applied for the determination of the TRIGA 2000 reactor parameter is the triple bare monitor. In this method, two monitors and one "reference" monitor isotopes are irradiated simultaneously without Cd cover.
If all monitors have a cross-section $\alpha(\nu) \sim \nu$ to an energy of 1.5 eV, the $\alpha$ parameter is obtained from the slope of the linear line to the plot:

$$\log \left[ \frac{E_{r,1}}{E_{r,2}} \right]^\alpha A_i \text{ versus } E_{r,1}, \text{ with:}$$

$$A_i = \frac{A_{\nu,1}(t)}{k_{0,1}(t)} \frac{Q_{0,1}}{G_{\nu,1}} - \frac{A_{\nu,2}(t)}{k_{0,2}(t)} \frac{Q_{0,2}}{G_{\nu,2}}$$

(2)

In this study, 1 = $^{198}$Au ($Q_{\nu,1}$=15.71(1.8), $E_r$=5.65(7.1) eV), 2 = $^{97}$Zr ($Q_{\nu,2}$=5.05(2.0), $E_r$=6260(4.0) eV) and 3 = $^{95}$Zr atau $^{97}$Nb ($Q_{\nu,3}$=248(1.5), $E_r$=338(2.1) eV).

Parameter $f$ measurement was performed using the bare bi-isotope method, by simultaneously irradiating two isotopes and a comparator without Cd cover. Parameter $f$ was determined using the equation (3):

$$f = \frac{k_{0,c(1)}}{k_{0,c(2)}} \frac{\varepsilon_{p,1}}{\varepsilon_{p,2}} \frac{Q_0}{Q_{0,1}} - \frac{k_{0,c(1)}}{k_{0,c(2)}} \frac{\varepsilon_{p,2}}{\varepsilon_{p,1}} \frac{A_{sp,1}}{A_{sp,2}}$$

(3)

2. Material and methods

In this study, several equipment and material was utilized such as Gamma Spectrometer (Canberra Industries Inc., Meriden, Connecticut, USA) with high resolution of High Purity Germanium (HPGe) detector GR2519 (25% relative efficiency, manufactured by Canberra Industries Inc., Meriden, Connecticut, USA) and multi-channel analyzer (MCA); analytical balance (AG 245 Mettler Toledo Ltd., Melbourne, Australia), Al-0.1%Au wire (IRMM 530R), Zr plate and Polyethylene vials with hinged cap (0.3 mL, Cole Palmer).

2.1. Preparation

Al-0.1%Au wire (IRMM 530R) and Zr plate were cleaned using ethanol (Merck), then were cut and weighed for about 0.0015 – 0.0018 and 0.0013 – 0.0023 gram, respectively. The wire and plate were placed in the 0.3 mL polyethylene vial then sealed by heating.

2.2. Irradiation and Counting

Irradiation of Au-Al wire and Zr plate were carried out for 3 days in TRIGA 2000 reactor, Bandung. After decay for a day then counted using Gamma Spectrometer for ~300 and ~1500 seconds to obtain $^{198}$Zr (T$^{1/2}$ = 16.74(0.1) hours, $E_r$ = 507.7 keV) and 198Au (T$^{1/2}$ = 2.695(0.1) days, $E_r$ = 411.8 keV). The data of $^{97}$Zr 95Zr (T$^{1/2}$ = 64.03(0.01) days, $E_r$ = 756.7 keV) was collected by counting Zr plate for ~4000 sec, after 2 weeks decay. Gamma spectrum was observed using Genie 2000 software version 3.1 (Canberra Industries Inc., Meriden, Connecticut, USA).

2.3. Determination of $\alpha$ and $f$ parameter

Triple bare monitor method was applied for $\alpha$ parameter measurement. In this method, monitor and the reference monitor isotope (Au and Zr) were irradiated simultaneously without Cd cover. Specific activity of isotope $^{198}$Au (Asp,$^{198}$Au), $^{97}$Zr (Asp,$^{97}$Zr) and isotope $^{95}$Zr (Asp,$^{95}$Zr) or $^{97}$Nb (Asp,$^{97}$Nb) was measured for all gamma energy and then plotting the linear graph $\log \left[ \frac{E_{r,1}}{E_{r,2}} \right]^\alpha A_i$ versus $E_{r,1}$ using software BATAN k0 Utility (BAkUL) v.1.0 developed by Putranto Ilham Yazid, where $\alpha$ value was calculated using equation (2). The $\alpha$ parameter was obtained from slope of the linear graph. Parameter
f measurement was performed using the bare bi-isotope method, by simultaneously irradiating two isotopes and a comparator without Cd cover.

3. Result and Discussions

Every irradiation position in the TRIGA 2000 research reactor under normal conditions has a greater number of thermal neutrons than epithermal neutrons [8]. This affects the number of atoms that can be activated due to reaction with thermal and epithermal neutrons so that the statistical fluctuations in the formation of isotopes induced by thermal neutrons will overlap with those induced by epithermal neutrons, for this reason calibration of reactor parameters including $f$ and $\alpha$ [9]. For the measurement of $\alpha$ and $f$ parameters, some flux monitor material used in the form of pieces or metal alloy wires with varying $\tilde{E}$ which represents the range of low to high energy to determine the stability of alpha values in all epithermal neutron energy regions at an irradiated position [10][11]. Table 1 displays a list of relevant flux monitors and nuclear data used in this study.

| Monitor                  | E$_r$, eV | Q$_0$        | k$_0$ | Ge | Gt | T1/2 |
|--------------------------|-----------|--------------|-------|----|----|------|
| -$^{197}$Au(n,\gamma)$^{198}$Au | 5.65 ± 0.40 | 15.71 ± 0.28; F$_{cd}$=0.991 | 1 | 1 | 1 | 2.695 d |
| -$^{95}$Zr(n,\gamma)$^{95}$Zr | 6260 ± 5.05 ± 0.42 | 1.10E-04 | 0.9 | 1 | 64.03 d |
| -$^{96}$Zr(n,\gamma)$^{97}$Zr | 330 ± 248 ± 1.5 | F$_{cd}$=1 | 6.79E-07 | 0.9 | 1 | 16.74 h |

Source: www.kayzero.com/k0nna/k0nnaorg/Nuclear_Data_SC

In this study, Au-Al wire and Zr plates with weights ranging from 0.0015 - 0.0018 and 0.0013 - 0.0023 grams were irradiated for 3 days at the TRIGA 2000 reactor, respectively. Irradiation was carried out at CT1-1 and G17-2 positions on the reactor core, as shown in Figure 1 and 2. After one day decay, Au-Al wire and Zr plate were counted using gamma spectrometer equipped with HPGe detector for ~300 and ~1500 seconds, to obtain counting data of isotope -$^{97}$Zr (T$_{1/2}$ = 16.74(0.1) hours, E = 507.7 keV) and -$^{198}$Au (T$_{1/2}$ = 2.695(0.1) hours, E = 411.8 keV). The data of isotope -$^{95}$Zr (T$_{1/2}$ = 64.03(0.01) days, E = 756.7 keV) was obtained by counting Zr plate for ~4000 seconds, after 2 weeks decay. The analysis of Gamma spectrum from counting process as shown in Figure 3, 4 and 5 was performed using software Genie 2000.
Figure 1. Configuration of irradiation position

Figure 2. Irradiation position at CT1-1 and G17-2

Figure 3. Spectrum of Au

Figure 4. Spectrum of Zr

Figure 5. Spectrum of Zr

Data analysis of Zr plate and Au-Al wire after 3 days irradiation in TRIGA 2000 reactor including counting data, specific activity for each isotope also α and f parameter data of TRIGA 2000 reactor. Specific activity of each isotope as presented in Table 2, and parameter α dan f was calculated using software Batan kO Utility v.1.0.

Table 2. Specific activity of Au and Zr at 2 irradiation position.

| Isotope | T½       | Energy (keV) | Asp (sec⁻¹ g⁻¹) | Posisi G17-2 | Posisi CT1-1 |
|---------|----------|--------------|-----------------|--------------|--------------|
| 198Au   | 2.695(0.1) days | 411.8        | 1.74 x 10⁹      | 5.25 x 10⁶   |
| 97Zr    | 16.74(0.1) hours | 507.7        | 1.12 x 10⁶      | 7.07 x 10⁴   |
The reactor parameters $\alpha$ were determined by the triple bare monitor method, by simultaneously irradiating the monitor with a reference isotope monitor (in this activity Au and Zr were used) without Cd cover while the $f$ parameter was determined using the bare bi-isotope method. The $\alpha$ parameter is the epithermal spectrum deviation parameter from the $1/E$ distribution. According to equation 2, $\alpha$ parameters could be calculated using the following formula:

$$(a-b)Q_{01}(\alpha)\frac{G_{e1}}{G_{th1}} - aQ_{03}(\alpha)\frac{G_{e2}}{G_{e2}} + bQ_{03}\frac{G_{e3}}{G_{e3}} = 0$$

with  

$$a = \left(1 - \frac{A_{xp2}}{A_{xp1}} \frac{k_{0,Au}(1)}{k_{0,Au}(2)} \frac{\varepsilon_{p1}}{\varepsilon_{p2}}\right)^{-1}$$  

and  

$$b = \left(1 - \frac{A_{xp3}}{A_{xp1}} \frac{k_{0,Au}(1)}{k_{0,Au}(3)} \frac{\varepsilon_{p1}}{\varepsilon_{p3}}\right)^{-1}$$  

(4)

The curve for determining the $\alpha$ parameters generated by the BATAN k0 Utility (BAkul) v.1.0 program is shown in Figure 6 and Figure 7. The TRIGA 2000 reactor alpha values were obtained at G17 and CT1 positions respectively -0.087 and -0.100.

4. Conclusions

The TRIGA 2000 reactor parameter $\alpha$ and $f$ measurements were carried out at the CT1-1 and G17-2 irradiation positions. The values of $\alpha$ and $f$ of the TRIGA 2000 reactor at the CT1-1 irradiation position were -0.100 and 8.22, respectively, while the G17-2 positions were -0.087 and 13.8. Each irradiation hole has different $\alpha$ and $f$ values, so it is necessary to determine the reactor parameters for the other irradiation positions. $\alpha$ and $f$ measurement results can be used for irradiation of the sample and can be used as a database to support the implementation of k0-NAA method in PSTNT.

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