Nucl. Sci. and Tech., Vol.13, No. 1 (2023), pp. 29-36

Nuclear Science and Technology

Journal homepage: http://jnst.vn/index.php/nst

Self-characterization of irradiation facility using synthetic multi-element standard for determination of k0-factors of seven radionuclides of interest

Phonesavanh Lathdavong[1*], Ho Manh Dung[2], Ho Van Doanh[2], Tran Quang Thien[3], Truong Truong Son[4], Hoang Sy Minh Tuan[5]

[1]*Radiation and Nuclear Safety Office, Ministry of Education and Sports of Lao PDR*
[2]*Center for Nuclear Technologies, 217 Nguyen Trai Str., District 1, Ho Chi Minh City, 71014, Viet Nam*
[3]*Dalat Nuclear Research Institute, 01 Nguyen Tu Luc St., Dalat City, 67000, Viet Nam*
[4]*HCMC University of Education, 280 An Duong Vuong St., Dist. 5, HCMC 72711, Viet Nam*
[5]*Thu Dau Mot University, Tran Van On Street, Phu Hoa Ward, Binh Duong, Vietnam*
**Corresponding author's e-mail:lathdavong.psv@gmail.com*

Abstract: In this study, the k_0 factors of seven short and medium-lived radionuclides (i.e. ^{66}Cu, ^{52}V, ^{38}Cl, ^{134m}Cs, ^{128}I, ^{140}La, and ^{56}Mn) were experimentally determined at the Dalat nuclear research reactor (DNRR). The synthetic multi-element standard (SMELS) samples were irradiated at channel 7-1 of the DNRR. SMELS Type III contains Au and Zr, thus it allows the self-characterization of the irradiation facility (i.e. the direct determination of f and α with only this material). The k_0 factors of seven short and medium-lived radionuclides obtained in this work are in good agreement with the reference k_0 factors. The ratio values between the experiment and reference k_0 values were ^{66}Cu of 1.004, ^{52}V of 1.004, ^{38}Cl of 0.961 and 0.976 (1642.7 keV and 2167.4 keV, respectively), ^{134m}Cs of 0.831, ^{128}I of 1.022, ^{140}La of 0.936, 0.955 and 0.955 (328.8 keV, 487.0 keV and 1596.2 keV, respectively), and ^{56}Mn of 0.985 and 0.973 (846.8 keV and 1810.7 keV, respectively). The difference of k_0 values between the experiment and the reference for the above radionuclides were lower than 7%. The present result indicates that the k_0 factors can be determined by using the SMELS materials.

Keywords: *k_0 factor, neutron activation analysis, short-lived and medium-lived radionuclides.*

I. INTRODUCTION

The original 250-kW TRIGA Mark-II reactor, located at Dalat City, South of Viet Nam, was constructed and put into operation in March 1963. It was upgraded to a power of 500-kW in 1984 using the Russian VVR-M2 fuel type and renamed as Dalat nuclear research reactor (DNRR) [1]. The reactor has been used for neutron activation analysis (NAA), radioisotope production, nuclear educational training, and fundamental and applied research. NAA is one of the main applications at the DNRR for the determination of multiple elements via short-, medium- and long-lived radionuclides with the analyst capacity is up to 65 elements [2]. For short-lived radionuclides with the half-life is from 2 minutes to 2.9 hours and for medium-lived radionuclides with the half-life is from 10 hours to 3 days, the sample is often irradiated at the channel 7-1 by the semi-auto pneumatic transfer system (PTS) at the DNRR [3]. The irradiation time for short- and medium-lived samples is in the range of 45

https://doi.org/10.53747/nst.v13i1.402
Received 24 January 2023, accepted 07 September 2023

seconds to 20 minutes, and then the sample is measured by the gamma spectrometer coupled with HPGe detector GMX-30190 [4].

The k_0 standardization in NAA (named k_0-NAA) has been approved for multiple elements in the nuclear research reactor throughout the world [5], which is well established for its advantages, e.g., high sensitivity, high accuracy and low detection limits of elements in various of matrices and the k_0 standardization method is often used for multi-element analysis. k_0-NAA is based on the k_0 factors, which are a combination of nuclear data constants, e.g., atomic mass, isotopic abundance, the yield of gamma ray and thermal neutron cross-sections. However, Q_0 factors are related to the ratio of the resonance integral to the thermal neutron cross-section and the nuclear data determined by literature review and experiment [6, 7]. In k_0-NAA, more than 130 isotopes with the k_0 value are known with sufficient low uncertainty [8].

k_0-NAA is related to the simultaneous irradiation of a sample and a neutron flux monitor, such as gold (Au), and the use of a "k_0 factor" for the nuclear constant composition, which is often independent of irradiation and measurement conditions. To calculate the concentration of the element, k_0–NAA uses neutron spectral parameters, e.g., thermal to epithermal neutron flux ratio (f), the shape factor of epithermal neutron flux (α), and the detector absolute efficiency (ε). These parameters can be utilized depending on irradiation position of the nuclear research reactor and counting facility [5]. De Corte and Simonits indicated that other input parameters, for instance k_0 and Q_0 can be measured by the experiment in the literature review [9]. After several implementations of k_0–NAA in the NAA laboratory at DNRR [2], extensive innovation has arisen regarding the computer technology, evaluation programs, and laboratory instrumental improvements. Therefore, it has been significantly beneficial to re-validate this standardization technique in this laboratory.

In NAA method, the theory of k_0 is given by following equation as [10]:

$$k_0 = \frac{M^* \theta \gamma \sigma_0}{M \theta^* \gamma^* \sigma_0^*} \quad (1)$$

In Eq. (1), M is the atomic mass, θ is the abundance of isotope, γ is the absolute intensity, σ is the cross-section at 2200 m.s^{-1} of reaction (n, γ), "*" is the parameters of comparator (^{197}Au).

k_0 factor is determined experimentally by both bare irradiation method (without Cd-cover and with Cd cover), which can be calculated [11] by following Eq. (2):

$$k_{0,\mathrm{Au}}(a) = \frac{A_{\mathrm{sp,a}}}{A_{\mathrm{sp,Au}}} \times \frac{G_{\mathrm{th,Au}}.f + G_{\mathrm{e,Au}} Q_{0,\mathrm{Au}}(\alpha)}{G_{\mathrm{th,a}}.f + G_{\mathrm{e,a}} Q_{0,\mathrm{a}}(\alpha)} \times \frac{\varepsilon_{\mathrm{p,Au}}}{\varepsilon_{\mathrm{p,a}}} \quad (2)$$

In Eq. (2), where α is a shape parameter for epithermal neutron distribution; f is the thermal to epithermal neutron flux ratio, ε_p is the full energy peak detection efficiency, A_{sp} is the specific radioactive activity.

$Q_0(\alpha)$ is the ratio of resonance integral to cross-section, which is given by:

$$Q_0(\alpha) = \frac{Q_0 - 0.429}{\bar{E}_r^{\alpha}} + \frac{0.429}{(2\alpha+1)0.55^{\alpha}} \quad (3)$$

Where, $\bar{E}_{\mathrm{r}}$ is the effective resonance energy. In case of f ratio is sufficient in comparison with Q_0 value, the above Eq. (2) is presented in Eq. (4):

$$k_{0,\mathrm{Au}}(a) = \frac{A_{\mathrm{sp,a}}}{A_{\mathrm{sp,Au}}} \times \frac{G_{\mathrm{th,Au}}}{G_{\mathrm{th,a}}} \times \frac{\varepsilon_{\mathrm{p,Au}}}{\varepsilon_{\mathrm{p,a}}} \quad (4)$$

The Q_0 factor is determined experimentally by "Cd ratio" in following method [1] in Eq. (5):

$$Q_{0,a}(\alpha) = Q_{0,\mathrm{Au}}(\alpha) \times \frac{F_{\mathrm{Cd,Au}}.R_{\mathrm{Cd,Au}}-1}{F_{\mathrm{Cd,a}}.R_{\mathrm{Cd,a}}-1} \times \frac{G_{\mathrm{th,a}}}{G_{\mathrm{e,a}}} \times \frac{G_{\mathrm{e.Au}}}{G_{\mathrm{th,Au}}} \quad (5)$$

In Eq. (5), F_{cd} is the Cd transmission factor and R_{Cd} is the Cd ratio.

II. EXPERIMENTAL

A. Irradiation and measurement

The comparator, such as gold (Au), and SMELS Type I sample for determining the k_0 factors of all seven short- and medium-lived radionuclides (^{66}Cu, ^{52}V, ^{38}Cl, ^{134m}Cs, ^{128}I , ^{140}La, and ^{56}Mn), were prepared and packaged in a high purity polyethylene box for irradiation. The electronic balance was used to weight the comparator and SMELS samples, as shown in Table I. Then the comparator and SMELS sample were irradiated at channel 7-1 of DNRR. The irradiation time was 70 seconds at a thermal neutron flux of 3.80E+12 n/cm^2/s. The HPGe detector was used to measure gamma-rays emitted from the irradiated samples [9, 11, 12]. The efficiency of absolute full energy peak detection was calibrated by using a standard gamma-ray source of ^{152}Eu, and the efficiency curve at 51 mm to the detector surface is shown in Fig. 1. Regarding the k_0 factors determination, both the gold comparator and the SMELS Type I standard sample were measured at the same distance from the detector surface.

Table I. Sample preparation

No.	*Name*	*Type of sample*	*Sample ID*	*Mass (g)*
1	IRMM 530R Al-0.1%Au	Comparator	A0289	0.00411
2	SMELS Type I	Reference Material	SM21a	0.03185

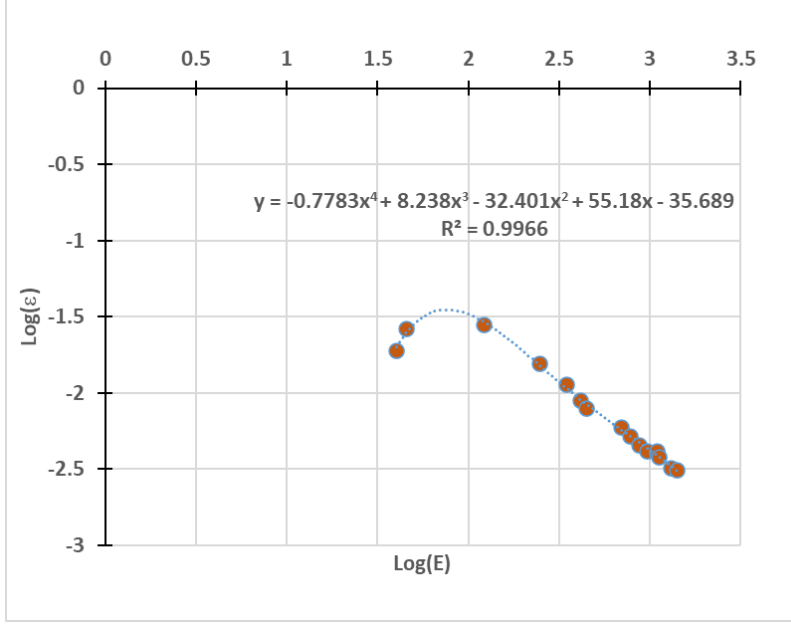

Fig. 1. GMX-30190 detector's efficiency curve at 51 mm to sample

The irradiation time was 70 s, the decay time was between 5-10 m for short-lived radionuclides, 1-2 h for medium-lived radionuclides and the counting time was kept at 300 s and 900 s for short- and medium-lived radionuclides, respectively. The conditions of the measurement and irradiation as well as the standard reference materials (SRMs) information are shown in Table II and Table III, respectively.

Table II. The irradiation, decay and counting times for determination of k_0 factor

Radionuclides	T_i (s)	T_d (s)	T_c (s)	Distance to Detector (mm)
^{66}Cu, ^{52}V	70 s (~30 mg)	5-10 m	300	51
^{198}Au, ^{38}Cl, ^{134m}Cs, ^{128}I , ^{140}La , ^{56}Mn		1-2 h	900	51

Table III. The standard reference materials (SRMs) and comparator information

Sample name	Element	Assigned Value ± U, mg/kg
SMELS Type I	Au	82.7 ± 1.7
	Cl	4330 ± 170
	Cs	897 ± 37
	Cu	3930 ± 120
	I	152 ± 5
	L a	265 ± 10
	Mn	113.9 ± 3.3
	V	39 ± 1.6
IRMM 530R Al-0.1%Au	Au	1003 2

B. Calibration of neutron spectra in channel 7-1

The Högdahl convention is often used to determine α and *f* values [11-13]. The bare double-comparator method was used to determine α-values by employing ^{197}Au and ^{96}Zr monitors [14, 15]. The α, *f*, and thermal neutron flux (ϕ) parameter are shown in Table IV.

Table IV. Neutron spectra parameters of irradiation channel

Parameter	Value	Uncertainty
ϕ_{th}	3.80E+12 n/cm^2/s	1.524E+11 n/cm^2/s
f	9.86	0.10
α	-0.0676	0.0006

For the irradiation condition at the DNRR, there are some recommended gamma energy (Eγ) peaks of the nuclear parameters and typically half-lives for k_0-NAA as shown in Table V.

Table V. Nuclear parameters condition for k_0-NAA

Element	X (n, x) Y	$T_{1/2}$	Eγ (keV)
Au	^{197}Au (n, γ) ^{198}Au	2.695 d	411.8
Cl	^{37}Cl (n, γ) ^{38}Cl	37.24 min	1642.7, 2167.4
Cs	^{133}Cs (n, γ) ^{134m}Cs	2.903 h	127.5
Cu	^{65}Cu (n, γ) ^{66}Cu	5.12 min	1039.2
I	^{127}I (n, γ) ^{128}I	24.99 min	442.9
La	^{139}La (n, γ) ^{140}La	1.678 d	328.8, 487.0, 596.2
Mn	^{55}Mn (n, γ) ^{56}Mn	2.579 h	846.8, 1810.7
V	^{51}V (n, γ) ^{52}V	3.75 min	1434.1

C. Calculations

For experimental determination of the k_0 factor with regards to the "bare monitor" [9], it is given by Eq. (2). Q_0 is the ratio of resonance integral (α correction factor) to 2200 m s^{-1} (n, γ) cross section, and is given [6, 16] as Eq. (3).

In Eq. (3), $\bar{E}$ is the effective resonance energy. Therefore, the experimental determination of k_0 factor requires the parameters e.g., α, ε, f and Q_0 [6, 11].

Both α and f are significant input parameters for calculating the k_0 factor and the concentration of elements of "1/ν" nuclides for the Högdahl convention. Thus, these parameters are dependent on the irradiation facilities and they should be determined for standardization purposes. The methods for determining the bare and cadmium ratios are used for α and f determination. However, it is also preferred for determination of intrinsic accuracy [5, 6, 9]. The method of cadmium ratio is often used for nuclear research reactors, in which the neutron spectrum remains unaffected for a long period of time. The well-known "non 1/ν" law is based on the (n, γ) reaction and requires additional neutron parameters, such as a modified spectral index and g-factor, as discussed elsewhere [13, 17].

III. RESULTS AND DISSCUSION

This study used the seven radionuclides of interest that were taken from k_0 reference [21]. The α, f, and the ϕ_{th} values from Table 4 were used to compute the k_0 factors. The α and f in this study are in good agreement with the literature values [16, 18, 19].

The k_0 factors were absolutely determined by the "bare monitor" method. Table VI shows the results of the experimental determination of k_0 factors of both short- and medium-lived radionuclides using DNRR as compared to the k_0 references.

Table VI. Experimental k_0, Au-factors of the short- and medium-lived radionuclides compared to the k_0 reference, using channel 7-1 facility at DNRR

Nuclides	E_γ (keV)	k_0-experiment	Uncertainty	k_0-Reference	Exp./Ref.
^{198}Au	411.8	-	-	1	-
^{52}V	1434.1	1.969E-01	1.237E-02	1.96E-01	1.004
^{66}Cu	1039.2	1.868E-03	1.156E-04	1.86E-03	1.004
^{38}Cl	1642.7	1.893E-03	1.197E-04	1.97E-03	0.961
^{38}Cl	2167.4	2.595E-03	1.635E-04	2.66E-03	0.976
^{134m}Cs	127.5	4.768E-03	3.049E-04	5.74E-03	0.831
^{128}I	442.9	1.185E-02	7.582E-04	1.16E-02	1.022
^{140}La	328.8	2.687E-02	1.679E-03	2.87E-02	0.936
^{140}La	487	6.080E-02	3.789E-03	6.37E-02	0.955
^{140}La	1596.2	1.279E-01	8.013E-03	1.34E-01	0.955
^{56}Mn	846.8	4.887E-01	3.028E-02	4.96E-01	0.985
^{56}Mn	1810.7	1.314E-01	8.120E-03	1.35E-01	0.973

The experimentally determined k_0 factors of 11 gamma-rays from seven radionuclides of both short- and medium-lived nuclides using the DNRR are given in Table 6, along with the recommended literature value [2, 6, 19, 20] and k_0 reference [21].

The k_0 factors reported in the Table VI show that ^{52}V at the energy of 1434.1 keV and ^{66}Cu at the energy of 1039.2 keV were determined for the short-lived radionuclides, and ^{128}I at the energy of 442.9 keV as medium-lived radionuclide was also determined and k_0 values of these radionuclides are in good agreement with the result of the k_0 reference, respectively. It is also taken into account that ^{52}V, ^{66}Cu, ^{128}I emit only a single gamma ray and there is no true coincidence effect. The ratios of experiment to reference were 1.004 for both isotopes ^{52}V and ^{66}Cu, and 1.022 for ^{128}I.

The medium-lived radionuclides ^{38}Cl and ^{56}Mn emit two gamma-rays with a high emission probability. The experimental results of the k_0 factors of ^{38}Cl at energies of 1642.7 keV and 2167.4 keV were 1.893E-03 and 2.595E-03, respectively. For ^{56}Mn at the energies of 846.8 and 1810.7, the k_0 factors were 4.887E-01 and 1.314E-01, respectively. The difference between the experimental results and reference values is lower than 3%. As a result, these experimental values matched the k_0 reference well [21].

For other radionuclides e.g., ^{140}La with three different energies of gamma-rays of 328.8 keV, 487 keV, and 1596.2 keV were used to calculate k_0 factors. The k_0 factors of ^{140}La are generally in good agreement compared to the reference values [21].

The ^{134m}Cs is a medium-lived radionuclide with an energy of 127.5 keV. Based on the result of the k_0 factor determination, the experimental value was compared to the k_0 reference, the ratio of Exp./Ref. is 0.831. The k_0 factor of ^{134m}Cs has a high difference with k_0 reference value because this energy is in low energy region (< 200 keV), in which some effects from Compton or low X-ray may influence the results. Thus, ^{134m}Cs does not appear to be good, due to low uncertainty.

IV. CONCLUSION

The self-characterization method of the irradiation facility based on the fast transfer pneumatic system at DNRR using a synthetic multi-element standard (SMELS) in this study demonstrated that the method is extremely useful for determining the k_0 factors of short- and medium-lived radionuclides. The irradiation of SMELS samples were performed at channel 7-1 of the DNRR. The k_0 factors of the seven radionuclides for both short- and medium-lived radionuclides including ^{66}Cu, ^{52}V, ^{198}Au, ^{38}Cl, ^{134m}Cs, ^{128}I , ^{140}La and ^{56}Mn with half-lives from 3.7 minutes to 2.9 hours were determined. The difference between the experimental k_0 results and reference k_0 values was within 7% for almost radioisotopes except for ^{134m}Cs, indicating that the k_0 factors of short- and medium-lived radionuclides of interest obtained in this study were very close to the reference k_0 values. The k_0 value for ^{134m}Cs biased significantly as compared with the reference value as its energy is in the low energy region in which the relative high interference of Compton effect on the efficiency curve contributes to the bias.

REFERENCES

[1]. H.M. Dung, Development of k0-based cyclic neutron activation analysis for short-lived radionuclides, Journal of Radioanalytical and Nuclear Chemistry, 291, 485-492, 2012.

[2]. H.M. Dung, P.D. Hien, The application and development of k0-standardization method of neutron activation analysis at Dalat research reactor, Journal of Radioanalytical and Nuclear Chemistry, 257, 643-647, 2003.

[3]. e.a. Doanh HV, A new rapid neutron activation analysis system at Dalat nuclear research reactor, Nuclear Science and Technology, 4, 82-91, 2014.

[4]. H. Doanh, C. Vu, T. Thien, P. Son, N. Sy, N. Giang, N. Dien, A new rapid neutron activation analysis system at Dalat nuclear research reactor, Nuclear Science and Technology, 4, 84-91, 2014.

[5]. R. Acharya, A. Nair, A. Reddy, S. Manohar, Validation of a neutron activation analysis method using k0-standardization, Applied Radiation and Isotopes, 57, 391-398, 2002.

[6]. F. De Corte, A. Simonits, Recommended nuclear data for use in the k0-standardization of neutron activation analysis, Atomic Data and Nuclear Data Tables, 85, 47-67, 2003.

[7]. G.L. Molnar, Z. Revay, L. Szentmiklosi, New perspectives for very short-lived neutron activation analysis, Journal of Radioanalytical and Nuclear Chemistry, pp. 157-163, 2003.

[8]. M. Kubešová, J. Kučera, Validation of k0-standardization method in neutron activation analysis—the use of Kayzero for Windows programme at the nuclear physics institute, Řež, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 622, 403-406, 2010.

[9]. F. De Corte, A. Simonits, k 0-Measurements and related nuclear data compilation for (n, γ) reactor neutron activation analysis: IIIb: tabulation, Journal of Radioanalytical and Nuclear Chemistry, 133, 43-130, 1989.

[10]. R. Acharya, J. Holzbecher, A. Chatt, Determination of k0-factors of short-lived nuclides and application of k0-NAA to selected trace element, Nuclear Instruments and Methods in Physics Research A, 680, 2012.

[11]. F. De Corte, The k0-standardization method: a move to the optimization of neutron activation analysis, Habilitation Thesis, Univ. of Gent, 1987.

[12]. R. Acharya, J. Holzbecher, A. Chatt, Determination of k0-factors of short-lived

nuclides and application of k0-NAA to selected trace elements, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 680, 1-5, 2012.

[13]. R. Acharya, A. Chatt, Characterization of the Dalhousie University SLOWPOKE-2 reactor for k0-NAA and application to medium-lived nuclides, Journal of Radioanalytical and Nuclear Chemistry, 257, 525-529, 2003.

[14]. H.M. Dung, P.D. Hien, The application and development of k0-standardization method of neutron activation analysis at Dalat research reactor, Journal of Radioanalytical and Nuclear Chemistry, 257, 2003.

[15]. H.M. Dung, Quality evaluation of the k0-standardized neutron activation analysis at the Dalat research reactor, Journal of Radioanalytical and Nuclear Chemistry, 309, 135-143, 2016.

[16]. R. Acharya, K. Swain, A. Kumar, N. Ajith, R. Verma, A. Reddy, Determination of k 0-factors and validation of k0-INAA for short-lived nuclides, Journal of Radioanalytical and Nuclear Chemistry, 286, 507-511, 2010.

[17]. F.D. Corte, A. Simonits, Recommended nuclear data for use in the k0 standardization of neutron activation analysis, Atomic Data and Nuclear Data Tables, 85, 2003.

[18]. H.M. Dung, Determination of k0 and Q0 factors for 5 short-lived radionuclides of interest in k0-NAA, Journal of Radioanalytical and Nuclear Chemistry, 2022.

[19]. M. Kubešová, k0-standardization in neutron activation analysis at LVR-15 reactor in Řež, 2012.

[20]. M.D. Ho, V.D. Ho, Q.T. Tran, N.S. Pham, V.C. Trinh, Q.D. Tran, T.S. Nguyen, M. Blaauw, The k0-standardized method for cyclic, epithermal and prompt gamma neutron activation analysis, 2017.

[21]. k_0-database 2020. (n.d.). Kayzero.com. Retrieved December 18, 2022, from http://www.kayzero.com/k0naa/k0naaorg/Nuclear_Data_SC/Entries/2020/8/24_Update_of_k0-database_I-128.html.