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Original Article

Determination of k_0 factors of short-lived nuclides ^{46m}Sc and ^{110}Ag for the k_0-NAA

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ABSTRACT

The k_0-standardization neutron activation analysis method has successfully determined the mass fraction of elements of interest using around a hundred analytical radionuclides. However, several very short-lived nuclides with half-life less than 100 s have not been used at Dalat research reactor. One of the reasons is that the values of k_0-factors of these nuclides are significantly different. Therefore, this work focused on re-determination and evaluation of k_0-factors of very short-lived nuclides ^{110}Ag ($T_{1/2} = 24$ s) and ^{46m}Sc ($T_{1/2} = 18.75$ s). The results of determination of the short-lived nuclides revealed that k_0-factor of ^{110}Ag is significantly difference between the existing data and the obtained results in this work. The evaluation of the k_0-factors was done by using the obtained results for application of k_0-NAA for NIST-1566b and NIST-2711A standard reference materials.

1. Introduction

The k_0-NAA standardization was a reliable and sensitive method that applied for determination of the quality and quantity of elemental concentration in the various samples. The accuracy of the k_0-NAA method depends directly on the k_0 factor [1]. Recently, the nuclear data of k_0 factor has been updated for 2003, 2012 and 2015 [2]. However, there are some data for short-lived nuclides that have not been updated or were measured by the only one laboratory. Thereby it does not have much credibility. The k_0 standardization in neutron activation analysis has been developed and applied to the Dalat nuclear research reactor (DNRR) of Ag and Sc elements.

While the determination results of the k_0 factor for most short-lived nuclides exhibit similarity in the literature, existing data in references show significant differences in the k0 factor of ^{110}Ag [3–5] as can be seen in Table 1. Therefore, before using k_0 factor of these short-lived nuclides, re-determination and re-valuation of k_0 factors for ^{110}Ag and ^{46m}Sc are necessary for recent applications and updating database on our k0-DALAT sofware (see Table 2).

1.1. The definition of k_0 factor

In the k_0 standardization of NAA, the concentration of an analyte "a" is obtained by the equation [1,6]:

$$\rho_a = \frac{(N_P/Wt_mSDC)_a}{(N_P/wt_mSDC)_{Au}} \times \frac{1}{k_{0,Au}(a)} \times \frac{f + Q_{0,Au}(\alpha)}{f + Q_{0,a}(\alpha)} \times \frac{\varepsilon_{P,Au}}{\varepsilon_{P,a}}$$

Where "Au" refer to the co-irradiated gold monitor ^{197}Au(n,γ)^{198}Au, $E_\gamma = 411.8$ keV.

$k_{0,Au}(a)$ – k_0 factor of monitor "m" (commonly Au) for analyte "a".

k_0 factor is defined $k_{0,Au}(a) = \frac{M_{Au}\theta_a\sigma_{0,a}I_{\gamma,a}}{M_a\theta_{Au}\sigma_{0,Au}I_{\gamma,Au}}$ where M is the atomic weight, θ is the isotopic abundance, σ_0 is the 2200 ms^{-1} (n,γ) cross-section, and γ is the emission probability. Although the k_0 factors are defined versus Au (co-irradiated with the sample to be analyzed), the gold monitor can be replaced by any monitor "m" for which a $k_{0,Au}$ factor is available, because $k_{0,m}$ (a) = $k_{0,Au}$ (a)/$k_{0,Au}$ (m).

The k_0 factor is experimentally measured according to the following equation:

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Table 1
The k_0 factor in the literatures.

Nuclide	$T_{1/2}$ (s)	E (keV)	γ (%)	Literature-$k_{0,Au}$ (%RSD)
^{110}Ag	24.56	657.8	4.5	0.0374 (2.0) (P Z Hien, 1991) [5] 0.0306 (0.4) (Van Lierde S, 1999) [3] 0.03627 (1.7) (Szentmikiosi L, 2006) [7] 0.0352 (1.7) (Acharya R, 2012) [4]

Table 2
The results of neutron spectrum parameters for calculation of k_0 factor [8].

Irradiation position	ϕ_{th} (n/cm^2/s)	α	f
13-2 channel	(4.2 ± 0.1) x 10^{12}	−0.038 ± 0.006	10.7 ± 2.4
Thermal column	(1.24 ± 0.03) x 10^{11}	0.092 ± 0.035	195 ± 4

$$k_{0,Au} = \frac{(N_P/wt_mSDC)_a}{(N_P/wt_mSDC)_{Au}} \times \frac{f + Q_{0,Au}(\alpha)}{f + Q_{0,a}(\alpha)} \times \frac{\varepsilon_{P,Au}}{\varepsilon_{P,a}}$$

Where N_p is the net number of counts in the full energy peak (corrected for pulse losses), w is mass of element, t_m is measuring time, $S = 1 - \exp(-\lambda t_i)$, λ is the decay constant, t_i is the irradiation time, $D = \exp(-\lambda t_d)$, $C = (1 - \exp(-\lambda t_m))/\lambda t_m$, f is the thermal to epithermal neutron flux ratio, $Q_0 = I_0/\sigma_0$ (resonance integral to 2200 ms^{-1} cross-section ratio), α is the measure for epithermal neutron flux distribution, approximated by $1/E^{1+\alpha}$ dependence and ε_p is the full-energy peak detection efficiency.

2. Experimental

The experiment was carried out by using a short irradiation and rapid measurement system, connected to irradiation position of vertical channel 13-2 with the thermal neutron flux of $(4.2 \pm 0.3) \times 10^{12}$ cm^{-2} s^{-1} and horizontal thermal column with the thermal neutron flux of $(1.2 \pm 0.1) \times 10^{11}$ cm^{-2} s^{-1}. The neutron spectrum parameters of f and α at two irradiation positions were determined and shown in Table 1. The transfer time of sample from irradiation position to detector (T_{trans}) was 3.165 ± 0.002 s (including both T_{trans} and the time required to start the detector). Timing information for both irradiation and counting would be instantly delivered to the workstation. Characterizations of this system has been reported elsewhere and thus will not be described here [7]. The operating principle of the PTS 13-2/TC is as shown in Fig. 1. The sample is loaded in loading sample (LS1) unit through sample changer (SE1) unit. Then the sample is transferred to irradiation position by fresh air from valve 11. At the end of irradiation time, the sample is ejected by fresh air from valve 12 or valve 13. Next, sample passes relief unit (RU) to separation unit (SU) before going to counting chamber (CC). Sensor 4 is used to initiate the measurement. After the measurement, the sample is taken out through D4. V11, V12, V13, V14, V15 and V16 are the clean air valves to push the sample.

Table 3
The fitting coefficients of efficiency curves for calculation of k_0 factor.

Distance	a_0	a_1	a_2	a_3	a_4
5 cm	−22,993	34,146	−19,822	5,008	−0,475
10 cm	−8,297	9,502	−4,943	1,059	−0,085
15 cm	−15,632	21,836	−12,964	3,352	−0,328
18 cm	−23,032	32,912	−19,232	4,915	−0,473

In the experimental determination of k_0 factor, the Ag monitors were measured at 5 cm from GMX-4076 detector and the Sc monitors were measured at 10 cm. The fitting coefficients of efficiency curves equation for GMX-4076 detector were used for calculation k_0 factor as shown in Table 3:

$$log\ \varepsilon_p = a_0 + a_1 \log E + a_2 (\log E)^2 + a_3 (\log E)^3 + a_4 (\log E)^4$$

To determine k_0 factors, the Ag, Sc and Au monitors in Table 4 were irradiated and measured with repetition of 3 times for each monitor. The experimental parameters were presented in Table 4. The Sc monitors were measured at 10 cm. The Ag monitors were measured at 5 cm. Au monitors were measured at 5 cm and 10 cm. The irradiation time and decay time for all measurements were corrected in 0.01 s (see Table 5).

NIST-1566b and NIST-2711A standard reference materials (SRM) were prepared as can be seen in Table 6. The samples were used for evaluation of k_0 factors obtained in this work.

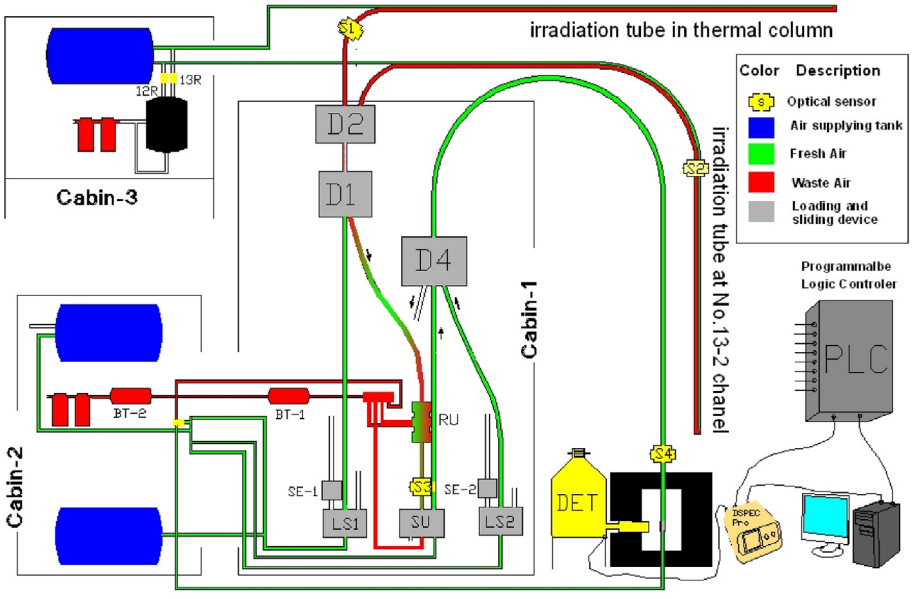

Fig. 1. Diagram of the auto-pneumatic transfer system installed at DNRR.

Table 4
The information of the monitors in the experiment.

Monitor (Shaping)	Producer	Radius (mm)	Thickness/Length (mm)	Weight (mg)	Density (g/cm^3)	G_{th}	G_e
Al-0.1%Au (Foil)	IRMM-530RC	5.44	0.100	6.268	2.7	1.00	1.00
99.9%Sc (Foil)	Shield werx	2.21	0.127	1.448	3.0	0.9994	1.00
1%Ag-Al (Wire)	Potash & Chem	1.00	1.397	2.960	2.7	1.00	0.9995

Table 5
The irradiation, decay and measurement time for the k_0 factor determination.

Nuclide	t_i (s)	t_d (s)	t_m (s)	Distance (mm)
^{46m}Sc	10	69	10	10 cm
^{110}Ag	20	4	30	5 cm

3. Results and discussion

Table 7 presents the experimental measurement results of k_0 factors for ^{110}Ag and ^{46m}Sc nuclides. The k_0 factor of ^{110}Ag nuclide determined is 0.0368 ± 0.0012. The k_0 factor value of ^{110}Ag in this work is entirely consistent with our earlier research on Dalat research reactor [5] with deviation of 1.6% and in line with the measurement result of other literatures such as Szentmikiosi in 2006 [9] and Acharya in 2012 [4]. Howeve, there is a significantly difference of 20% with Van Lierde in 1999 [3].

The k_0 factor of ^{46m}Sc nuclide determined in this study is 0.220 ± 0.005. This value is in good agreement with the results of other authors (Szentmikiosi in 2006 [9]; Acharya R in 2010 [10] and 2012 [4]). There are some reasons for that: (1) ^{46m}Sc emits only one gamma-ray with high emission probability; (2) Sc monitor have the purity of 99.9%, not mixed with Al. Therefore, it is insignificantly affacted by Compton background from Al; (3) Sc is light element used in its purity form, it absorbs insignificantly gamma-rays. The efficiency is also not much affected by the monitor geometry.

The truth value of k_0 factor obtained in this work for ^{110}Ag and ^{46m}Sc have been assessed by analyzing the NIST-1566b and NIST-2711A reference samples. The concentration of Ag in NIST-1566b and Sc in NIST-2711A were calculated by k_0-NAA method and shown in Table 8. The data indicate that an agreement between measured values and certified values was acceptable in regarding to the deviation of the mentioned two values below 11% for Ag and Sc in both recommended k_0 values and values from this work.

Table 8
The concentration result of Ag and Sc in SRM using recent k_0 value and k_0 value obtained from this work.

Ele.	SRM	Nuclide	k_0 factor	$\rho_{ex} \pm \Delta\rho_{ex}$	$\rho_{ce} \pm \Delta\rho_{ce}$	ρ_{ex}/ρ_{ce}
Results of Ag and Sc concentration using recently existing k_0 value [2]						
Ag	NIST-1566b	^{110}Ag	0.0306	0.72 ± 0.15	0.666 ± 0.13	1.08
Sc	NIST-2711a	^{46m}Sc	0.226	8.19 ± 0.70	8.50 ± 0.10	0.96
Results of Ag and Sc concentration using k_0 value from this work						
Ag	NIST-1566b	^{110}Ag	0.0368	0.59 ± 0.13	0.666 ± 0.13	0.89
Sc	NIST-2711a	^{46m}Sc	0.220	8.42 ± 0.72	8.50 ± 0.10	0.99

ρ_{ex} and **$\Delta\rho_{ce}$** are the experimental concentration and uncertainty obtained in this work.
ρ_{ce} and **$\Delta\rho_{ce}$** are the certificate concentration and uncertainty of standard reference material.

The ρ_{ex}/ρ_{ce} ratio for Ag is 1.08 in case of calculating from k_0 recommended value of 0.0306 while the ρ_{ex}/ρ_{ce} ratio for Ag is 0.89 in case of calculating from k_0 obtained in this work is 0.0368. This result shows that our result is in contrast to the result using recommended value.

The ρ_{ex}/ρ_{ce} ratio for Sc is 0.96 in case of calculating from k_0 recommended value of 0.226 while the ρ_{ex}/ρ_{ce} ratio for Ag is 0.99 in case of calculating from k_0 obtained in this work is 0.220. This result shows that k_0 value of 0.220 provides result of elemental concentration of Sc that are comparable to existing k_0 value.

4. Conclusions

Technical procedure for determination of k_0 factor was established for short-lived nuclides using well-thermal neutron irradiation position in Dalat research reactor. The facility for experimental procedure was calibrated carefully for k_0 factor in this work and further measurements. The k_0 factor of ^{110}Ag and ^{46m}Sc were determined in this work. The k_0 factor of ^{46m}Sc is entirely consistent with the literatures. However, the k_0 factor of ^{110}Ag

Table 6
The sample preparation parameters for NIST-1566b SRM and NIST-2711A

No.	Sample/Monitor	ID sample	Height of sample (mm)	Weight of sample (mg)
1	Al-0,1%Au	Au1113	1	4,750
2	Blank vial	BLK-2	10	200
3	NIST-1566b	OT22	10	201,81
4	Al-0,1%Au	Au1119	1	4,020
5	Al-0,1%Au	Au1228	1	6,41
6	NIST-2711a	MO204g	4	136,42

Table 7
The result of k_0 factor of this study and comparison with the other literatures.

Nucl.	Energy	This work, $k_{0,Au}$ (%RSD)	Literature-$k_{0,Au}$ (%RSD)	Bias (%)
^{110}Ag	657.8 keV	0.0368 (3.2)	0.0374(2.0) (P Z Hien, 1991)	−1.6
			0.0306 (0.4) (Van Lierde S, 1999) [3]	20.0
			0.03627 (1.7) (Szentmikiosi L, 2006)	1.4
			0.0352 (1.7) (Acharya R, 2012) [4]	4.5
^{46m}Sc	142.5 keV	0.220 (2.2)	0.225 (2.4) (Szentmikiosi L, 2006)	−2.0
			0.223 (5.4) (Acharya R, 2010)	−1.1
			0.222 (2.5) (Acharya R, 2012)	−0.7

determined in this work is significantly difference with recommended value. The truth value of k_0 factor has been assessd by the analysis of the elemental concentration of interest in a certified reference material in regarding to the deviation of the mentioned two values below 11%. Therefore, the k_0 factor for ^{110}Ag obtained in this work has been considered to update on the database/library of the k0-DALAT software.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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