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Measurement of k_0 values for europium, lutetium and iridium at FRM II with a very well thermalized neutron spectrum

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Abstract
The k_0 values of 6 non-1/*v* nuclides (^{152}Eu, ^{152m}Eu, ^{154}Eu, ^{177}Lu, ^{192}Ir and ^{194}Ir) were determined using the extended Høgdahl formalism at the research reactor FRM II with very high *f* values. Standards were irradiated in 4 channels at different local temperatures between 40 °C and 55 °C measured using temperature sensitive irreversible labels. A good agreement with the recommended values was found for ^{152}Eu, ^{154}Eu and ^{177}Lu using the original $g(T_n)$ factors by Gryntakis, however, the k_0 values for ^{152m}Eu in this work were 7% higher. New k_0 values were also determined using the $g(T_n)$ factors by Van Sluijs. Differences up to 6% were found for Eu isotopes compared with the recommended values. The recommended k_0 values for ^{192}Ir and ^{194}Ir could be confirmed using $g = 1$. The theoretical k_0 values for ^{177}Lu were calculated using new nuclear data. They are up to 6% less than the recommended values. The present k_0 values determined in this work showed a similar trend. The influence of different $g(T_n)$ factors on the determination of the k_0 values was investigated.

Keywords k_0 values · k_0 NAA · *g* factors · Non-1/*v* nuclides · Westcott convention · Extended høgdahl formalism · Neutron temperature · Neutron activation analysis

Introduction

The k_0 NAA method was first developed with the Høgdahl convention, which treated the activation with both thermal and epithermal neutrons as a temperature independent static process describing very well the (n,γ)reactions of most nuclides with cross sections varying as 1/*v* (*v* is the neutron velocity) in the thermal neutron energy region [1, 2]. For a few so-called non-1/*v* nuclides, a modification using the Westcott convention [3] with consideration of the influence of the neutron temperature on the cross sections was introduced in the 1990s [4, 5], while the definition of the k_0 factor including the cross section at a neutron velocity of 2200 m s^{-1} (corresponding to a neutron temperature T_n at 20 °C) remained unchanged. The Westcott $g(T_n)$ factors were used for the correction of the cross section at any temperature.

For the determination of the k_0 values for the non-1/*v* nuclides, the local neutron temperature at the irradiation position must first be determined. As a temperature monitor, Lutetium was suggested since the beginning, because the thermal neutron capture cross section of the reaction ^{176}Lu(n,γ)^{177}Lu varies very strongly with the neutron temperature. In this case, the recommended k_0 values of ^{177}Lu were calculated using atomic and nuclear data, including the cross section at the neutron velocity of 2200 m s^{-1}, from the 1980s [2]. For the experimental determination of the ^{177}Lu k_0 value, the neutron temperature has to be measured in an independent way, e.g. by using temperature readings of the reactor moderator measured by thermocouples [6]. At FRM II, a method using irreversible thermometer labels was developed to measure the temperature at the irradiation positions in situ [7] and applied to determine the k_0 values of some non-1/*v* nuclides in this work.

The Westcott $g(T_n)$ factors can be calculated from the neutron cross section $\sigma(E)$ values and have been updated several times since the 1960s [3, 8–11]. The latest update of the $g(T_n)$ factors was the calculations of Van Sluijs et al.

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published in 2015 [11]. However, the k_0 values for most of the non-1/*v* nuclides were determined in the 1980s with the $g(T_n)$ factors of the 1970s, above all with the $g(T_n)$ factors published by Gryntakis and Kim in 1975 [8]. Due to the different input databases of σ(*E*), e.g. ENDF/B or the European Activation File EAF and different algorithms, significant discrepancies of $g(T_n)$ factors can be found for Lu and other non-1/*v* nuclides. These lead to different neutron temperatures determined by using the Lu standard and different k_0 values finally.

The here mentioned three elements Lu, Eu and Ir are very important elements in multi element analysis, especially in the geosciences and cosmochemistry. With k_0 NAA, Ir was usually determined by measuring the short-lived isotope ^{194}Ir although its sensitivity is lower than that of the long-lived ^{192}Ir. Based on the calculation of the $g(T_n)$ factors in the 1970s, the ^{191}Ir(n,γ)^{192}Ir reaction was considered as non-1/*v* and no k_0 values had been measured for ^{192}Ir until 2014 [12, 13]. However, new calculations of $g(T_n)$ factors seem to support the temperature independence of the ^{191}Ir(n,γ)^{192}Ir reaction [10, 11].

Also in 2014, an extended Høgdahl formalism retaining the classical parameters (*f*, α, Q_0) was developed to replace the spectral index and s_0 factor of the Westcott convention for the non-1/*v* isotopes [14]. Cimpan and Kennedy recently determined the k_0 and Q_0 values of Lu and Eu using this new formalism and the $g(T_n)$ factors published by Holden [9] at a reactor with significant proportion of epi-thermal neutron flux [15]. However, they found large discrepancies between their results and the recommended values for Eu.

The research reactor FRM II with very well thermalized neutron flux can minimize the activation with the epi-thermal neutrons, so that the uncertainties of the determination can be reduced significantly. In the present work the k_0 values of Lu, Eu and Ir were determined; however, due to the very high *f* values, a determination of Q_0 values was not possible.

Conventionally, the $g(T_n)$ factors are associated with the target nuclides of the (n,γ) reactions, whereas the k_0 values are associated with the product nuclides because they refer to a specific emitted gamma-ray. Similarly, non-1/*v* nuclides mean the target nuclides, but are also used to describe the product nuclides in k_0 NAA. We follow these conventions in this paper.

Theory

For the determination of non-1/*v* nuclides with NAA, the k_0 method was modified with the Westcott formalism. If the neutron temperature is measured, the mass fraction ρ of an unknown element can be determined by the following formula neglecting the neutron self-shielding [5]:

$$\rho = \frac{\left(\frac{N_p}{SDCWt_m}\right)}{\left(\frac{N_p}{SDCwt_m}\right)^*} \frac{1}{k_0} \frac{g(T_n)^* + r(\alpha)\sqrt{T_n/T_0}s_0^*(\alpha)}{g(T_n) + r(\alpha)\sqrt{T_n/T_0}s_0(\alpha)} \frac{\varepsilon_p^*}{\varepsilon_p} \quad (1)$$

where (*) is the co-irradiated comparator, N_p is the measured peak area, $S = 1—exp\ (-\lambda t_{irr})$ with the decay constant λ and the irradiation time t_{irr}, $D = exp\ (-\lambda t_d)$ with the decay time t_d, $C = [1—exp\ (-\lambda t_m)]\ /\ \lambda t_m$ with the measuring time t_m; *W* and *w* are sample and monitor mass, $g(T_n)$ is the Westcott's *g* factor, T_n is the neutron temperature, *r* is the modified spectral index, s_0 is the modified reduced resonance integral to thermal cross section ratio, α is the epithermal spectrum shape parameter, ε_p is the full-energy peak efficiency. For a pure Maxwellian neutron spectrum, the epithermal index *r* is equal to 0 [3].

A simplified extended version of the modified Høgdahl convention was introduced recently to replace the classical parameters of Westcott formalism with the conventional k_0 parameters [14]

$$\rho = \frac{\left(\frac{N_p}{SDCWt_m}\right)}{\left(\frac{N_p}{SDCwt_m}\right)^*} \frac{1}{k_0} \frac{1 + Q_0^*(\alpha)/f}{g(T_n) + Q_0(\alpha)/f} \frac{\varepsilon_p^*}{\varepsilon_p} \quad (2)$$

where ^{197}Au (*) is used as the comparator and treated as a 1/*v*-nuclide, *f* is the thermal to epi-thermal flux ratio, Q_0 is the cross section ratio (resonance integral divided by cross section at $v = 2200$ m s^{-1}). The Q_0 values of some non-1/*v* nuclides were determined by using this new modified version [15]. Table 1 shows the ranges of Q_0 values found in the literature [12–18]. For a modern reactor with a very well thermalized neutron spectrum, i.e. a very high *f* value, the ratio of $Q_0(\alpha)\ /\ f$ is less than 0.3% for most nuclides [19]. Therefore, the above equations can be simplified further by replacing the activation term with just the $g(T_n)$ factor for all non-1/*v* nuclides:

$$\rho = \frac{\left(\frac{N_p}{SDCWt_m}\right)}{\left(\frac{N_p}{SDCwt_m}\right)^*} \frac{1}{k_0} \frac{1}{g(T_n)} \frac{\varepsilon_p^*}{\varepsilon_p} \quad (3)$$

Table 1 literature values of Q_0 [12–18]

Reaction	Q_0 literature
^{151}Eu(*n*,γ)^{152m}Eu	0.1–1.2
^{151}Eu(*n*,γ)^{152}Eu	0.5–1.36
^{153}Eu(*n*,γ)^{154}Eu	3.95–5.66
^{176}Lu(*n*,γ)^{177}Lu	1.67–3.59
^{191}Ir(*n*,γ)^{192}Ir	3.47–3.94
^{193}Ir(*n*,γ)^{194}Ir	12.0–13.4

Note that this simplified formula of the extended Høgdahl convention still has one difference from that of the Westcott formalism. The Westcott formula has $g(T_n)$ of the monitor in the numerator. With $g(T_n)$ of $^{197}Au(n,\gamma)^{198}Au$ approximately 1.007 at typical research reactor moderator temperatures [4], there is a 0.7% difference between the two formulae.

This formula can be applied to determine the k_0 values, if standards with known concentration are used:

$$k_0 = \frac{\left(\frac{N_p}{SDCW\rho t_m}\right)}{\left(\frac{N_p}{SDCwt_m}\right)^*} \frac{1}{g(T_n)} \frac{\varepsilon_p^*}{\varepsilon_p} \quad (4)$$

In this expression, the normally relevant parameters such as f, $Q_0(\alpha)$, $\overline{E}_r$(effective resonance energy) or $s_0(\alpha)$ have no influence on the calculation, which makes the determination of the k_0 values simpler and potentially more accurate.

It can be seen in Eq. 4 that the k_0 determination depends directly on the $g(T_n)$ factor used. Figures 1–5 show the Westcott $g(T_n)$ factors for Lu, Eu and Ir calculated by different authors [3, 8–11] in the temperature range between 20 °C and 100 °C, which is the operating temperature range at most research reactors. The neutron cross section data $\sigma(E)$ from the ENDF database used for these $g(T_n)$ calculations for ^{176}Lu and ^{151}Eu over a period of more than 50 years evolved over time, as shown in Fig. 6. The curves show also why ^{176}Lu is so sensitive on the thermal neutron temperature and why the $g(T_n)$ factor of ^{151}Eu has a negative slope.

The $g(T_n)$ factors can be fitted with a polynomial regression of 1st or 2nd order. In recent works, Van Sluijs et al. proposed fitting with a third-degree polynomial [11, 20]. However, the difference is less than 0.1% within the temperature range considered here for most nuclides. Even for ^{152}Eu, whose $g(T_n)$ factors show a slight curvature, the maximum deviation can reach only 0.2% in the region between 60 °C and 70 °C. In order to simplify the calculations, linear approximations were used in this work (Eq. 5) and the coefficients are given in Table 2.

$$g(T_n) = a_0 + a_1 T_n/1000 \quad (5)$$

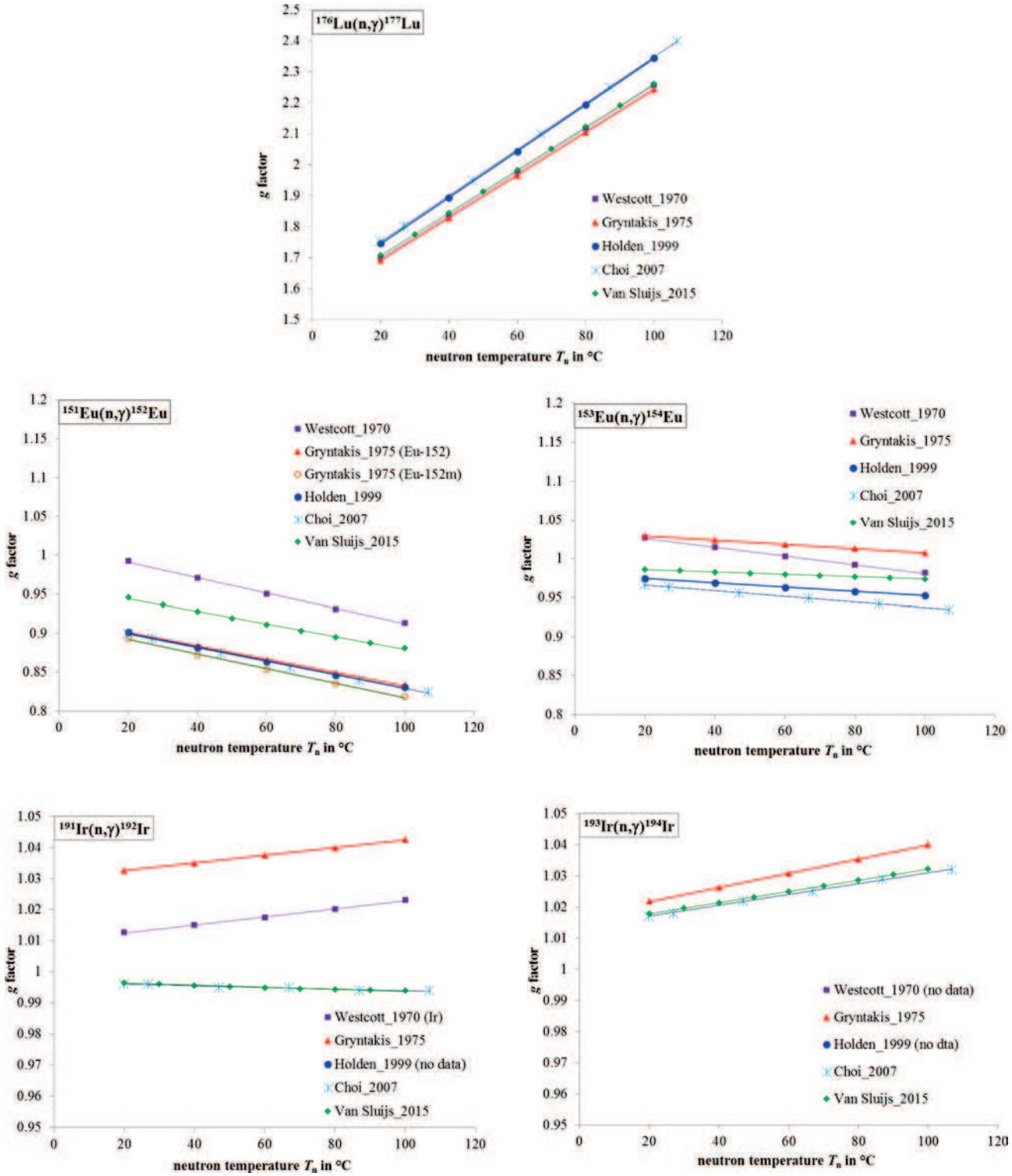
For Lu, the $g(T_n)$ factor lines in Fig. 1 are close to parallel and can be divided into two groups. The $g(T_n)$ factors of Holden (1999) and Choi and Trkov (2007) are quite close to each other, although they used different input databases of ENDF/B-V and EAF-99, but they are about 4% higher than the other three $g(T_n)$ data sets. This would lead to different results of the k_0 determination or temperature determination (up to 10 °C), if Lu is used as the temperature monitor [7]. Holden used in his 1999 paper the ENDF/B-VI data base for all nuclides except for ^{176}Lu. His g factors for ^{176}Lu were still based on the older ENDF/B-V database. He mentioned the fact that databases evolve and thus lead to changes in any data like $g(T_n)$ derived from them. The changes in the neutron cross sections $\sigma(E)$ for ^{176}Lu since ENDF/B-V, see Fig. 6, were the reason for the differences in the more recently calculated $g(T_n)$ factors seen in Fig. 1. Surprisingly, the $g(T_n)$ factors calculated by Van Sluijs et al. in 2015 using the new evaluated nuclear data of BNDF/B-VII.1 came back to the old factors calculated in the 1970s.

Large differences could be also found between the $g(T_n)$ factors for ^{151}Eu. Here, the $g(T_n)$ factors by Van Sluijs et al. are significantly different from the others, up to 5% because the evaluated cross section data was changed after ENDF/B-VI. Gryntakis and Kim calculated the $g(T_n)$ factors separately for the activation products ^{152}Eu, ^{152m}Eu with different isomer yield ratios. However, the difference is about 1.5% between them. Other authors neglected this tiny difference because they based their calculations on evaluated data that did not contain this detailed information. For ^{153}Eu, the influence of the temperature on the $g(T_n)$ factors is not strong: all lines are quite flat. Therefore, ^{153}Eu was usually treated as a 1/v nuclide. But the difference between the $g(T_n)$ factors is more than 6%.

^{191}Ir was always treated as a non-1/v nuclide in the past. Obviously, the $g(T_n)$ factor by Gryntakis and Kim with a deviation up to 4% from unity had a big influence on this decision at that time, when the k_0 method was launched in the 1980s. However, Holden considered both isotopes of Ir as 1/v nuclides and did not calculate the $g(T_n)$ factors for them. Westcott did not give the $g(T_n)$ factors for both isotopes of Ir explicitly and seemingly gave the factors for the element Ir. Choi and Trkov and Van Sluijs et al. calculated the factors separately [10, 11]; however, their results for ^{191}Ir are very close to unity. On the contrary, the $g(T_n)$ factors for ^{193}Ir show greater deviation from unity.

Experimental

Lu, Eu and Ir standards for the determination of the k_0 values were prepared using 1000 mg L^{-1} (±0.5%) certified ICP standard solutions CertiPUR® manufactured by Merck. A 200 µL aliquot of each standard was pipetted onto 0.1 mm thick round filter paper (Ø 16 mm) directly on a balance during weighing. The filter papers were enclosed in PE bags after drying. The certified element concentrations were (989 ± 5) µg g^{-1} for Lu, (986 ± 4) µg g^{-1} for Eu and (970 ± 5) µg g^{-1} for Ir. Au–Al foils (IRMM-530R, 0.1003 ± 0.0012%) as monitors and Lu–Al standard foils (IRMM-sp96091, 0.100 ± 0.002* % Lu, *: estimated) as reference standard were punched to disk shapes with the



Figs. 1–5 g(T_n) factors for the non-1/*v* isotopes of Lu, Eu and Ir from different data sources [8–11]

same diameter as the filter papers. Both foils had a thickness of 0.1 mm.

For the in-situ determination of local temperature, irreversible thermometer labels from the company RS with a measuring range between 40 °C and 82 °C in different steps of 3 °C to 5 °C were chosen. The labels indicate the temperature by changing from white/grey to black permanently. The sensitivity tolerance and the response time are ± 1 °C

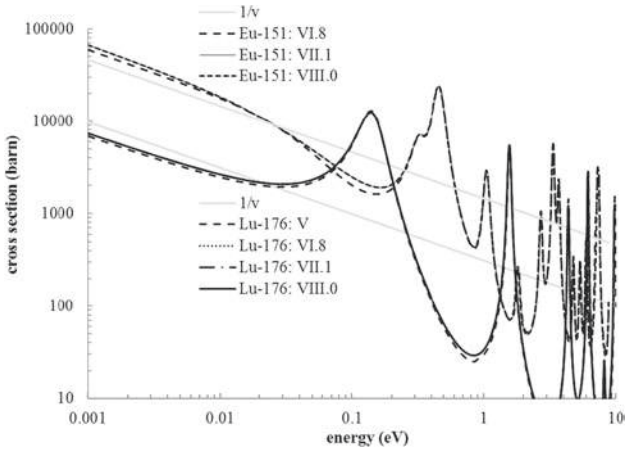


Fig. 6 neutron cross section for ^{176}Lu and ^{151}Eu in the ENDF database

and 1 min, respectively [21]. Standards, comparators and thermometer labels had almost exactly the same geometry and were packed side by side together in the irradiation capsules, so that the influence of the flux gradients could be minimized effectively.

The research reactor FRM II has a compact core with highly enriched uranium located in the centre of the moderator tank filled with heavy water. The prepared standards were irradiated in four positions with different neutron fluxes from 3.5E12 to 4.4E13 cm^{-2} s^{-1} and f values from 3500 to 6500. The irradiation durations varied from 10 min to 1 h.

All standards including the Au comparators were measured at 25 cm on two HPGe detectors with relative efficiencies of 27–34%. The gamma counting was performed using Genie2000 (Mirion) spectroscopy software. The uncertainties of counting statistics were less than 0.5% for most gamma lines and the spectrometer dead time was always below 1% except for the ^{152m}Eu measurements. Thus, the pulse pile-up effect was well within the correction capabilities of the spectrometer and led to negligible error. The first measurements were carried out at the same or the next day after the irradiation to determine the short-lived nuclides ^{194}Ir and ^{177}Lu and after three days for ^{152m}Eu to reduce the count rate. The long-lived nuclides were measured at least two weeks later.

The full-energy peak efficiencies at 25 cm on the detectors were calibrated by using a multiple radionuclide standard solution QCY48 from Eckert & Ziegler, which was pipetted on a filter paper with a diameter of 16 mm. The uncertainties of standards used for the calibration were 2.3% to 3.5%. All standard samples, monitors used for the k_0 determination and the QCY standard had exactly the same geometry and were measured at the same distance, so any systematic errors could be minimized, because for each detector only one efficiency curve was used and it was not necessary to do the calculation for the solid angles.

Results

The temperature measurements with the thermometer labels showed a local temperature range from ca. 40 °C to 55 °C (41.5 ± 1.5 °C, 41.5 ± 1.5 °C, 47.5 ± 1.5 °C and 54 ± 3 °C) in the irradiation positions (3 rabbit channels RPA2, 3, 5 and 1 fishing position JBE70, respectively) chosen for the k_0 determination. These results were in accordance with the temperatures monitored by the reactor control system at different places in the reactor. A detailed description of the in situ temperature measurement can be found in another publication [7]. The $g(T_n)$ factors were calculated according

Table 2 The coefficients of the linear approximation used for the calculation of $g(T_n)$ factors in this work

Nuclide year	Coefficient	Westcott [3] 1970	Gryntakis [8] 1975	Holden [9] 1999	Choi [10] 2007	Sluijs [11] 2015
Data library		BNL-325	diverse	ENDF/B-VI.4	EAF-99	ENDF/B-VII.1
^{176}Lu	a_0	1.560	1.552	1.594*	1.602	1.569
	a_1	6.966	6.892	7.488*	7.451	6.910
^{151}Eu	a_0	1.011	0.9180	0.9177	0.9158	0.9604
(^{152}Eu)	a_1	−0.9875	−0.8575	−0.8835	−0.8664	−0.8107
^{151}Eu	a_0		0.9110			
(^{152m}Eu)	a_1		−0.9410			
^{153}Eu	a_0	1.037	1.034	0.9794	0.9727	0.9886
	a_1	−0.5610	−0.2700	−0.2725	−0.3537	−0.1485
^{191}Ir	a_0	1.010	1.030		0.9965	0.9970
	a_1	0.1290	0.1235		−0.0252	−0.0328
^{193}Ir	a_0	1.010	1.017		1.014	1.014
	a_1	0.1290	0.2270		0.1748	0.1800

* ENDF/B-V.1

to Eq. 5 using the parameters given in Table 2 and the measured local temperatures of each irradiation.

The k_0 values shown in Table 3 were determined using Eq. 4 with different selected $g(T_n)$ factors for a well-aimed comparison with the literature values [4, 15, 16, 18]. The values are averages from at least 8 measurements. The $g(T_n)$ factors by Gryntakis and Kim were used to determine the k_0 values for ^{152}Eu in the early time of k_0 NAA [2, 4]. Holden's data were mentioned in the literature [16] and used for the determination of the k_0 of Lu and Eu in 2016 [15]. This work is the first to use the $g(T_n)$ of Van Sluijs et al. to determine k_0 values. For ^{154}Eu, ^{192}Ir and ^{194}Ir, the k_0 values were also determined with $g=1$, as was done in the previous published works.

For convenience, in Table 3 and in the following text, abbreviations, e.g. Holden k_0 values, will be used to refer to the k_0 values calculated with Holden's $g(T_n)$ factors and so on.

Comparison with earlier k_0 factor determinations

Cimpan and Kennedy measured k_0 values for Lu and Eu in 2016 [15]; these data were used in the comparison next to earlier recommended data. The measurements presented

Table 3 k_0 values obtained using different $g(T_n)$ factors from Gryntakis [8], Holden [9], Van Sluijs [11] and $g=1$ in this work compared with literature values [4, 16, 18] and recalculated data from [15]

	Energy (keV)	k_0, this work					k_0, recommended		k_0, [15] recalculated		
		Gryntakis	Holden	Van Sluijs	$g=1$	s,%	(tentative)	s,%	Gryntakis	Van Sluijs	s,%
^{152}Eu	121.8	12.1	12.2	11.5		1.7	12.8	0.8	13.3	12.5	3
	244.7	3.40	3.43	3.24		1.7	3.44	0.3	3.49	3.27	2
	344.3	11.7	11.8	11.1		1.7	11.9	0.9	12.3	11.51	2
	443.9	1.41	1.42	1.34		1.7	1.39	1.2	1.48	1.39	2
	778.9	5.73	5.76	5.45		1.7	5.70	0.8	6.06	5.68	2
	867.4	1.87	1.89	1.78		1.8	1.88	0.9	1.99	1.86	2
	964.1	6.48	6.52	6.17		1.8	6.46	0.4	6.89	6.46	2
	1085.9	4.49	4.52	4.27		1.9	4.57	0.4	4.73	4.44	2
	1112.1	5.97	6.01	5.68		2.0	6.06	0.8	6.40	6.00	2
	1408.0	9.35	9.41	8.90		2.3	9.36	0.6	9.67	9.06	2
^{152m}Eu	121.8	1.64	1.61	1.56		1.8	(1.48)	–	1.81	1.68	3
	344.3	0.534	0.520	0.508		1.9	(0.498)	–	0.585	0.543	2
	841.6	3.23	3.17	3.07		1.8	(3.02)	–	3.51	3.26	2
	963.4	2.67	2.62	2.54		1.8	(2.49)	–	2.88	2.67	2
^{154}Eu	248.0	0.155	0.164	0.161	0.158	1.8	(0.155)	–	0.151[a]	0.156[b]	2
	591.8	0.110	0.117	0.115	0.112	2.3	0.108	1.5	0.115[a]	0.119[b]	2
	723.3	0.446	0.471	0.464	0.455	1.8	0.446	1.5	0.471[a]	0.488[b]	2
	756.9	0.104	0.110	0.109	0.106	2.6	(0.108)	–	0.119[a]	0.124[b]	2
	873.3	0.272	0.287	0.283	0.277	2.0	0.272	1.4	0.293[a]	0.304[b]	2
	996.4	0.234	0.248	0.244	0.239	2.0	(0.230)	–	0.239[a]	0.248[b]	2
	1274.4	0.790	0.835	0.822	0.806	2.0	0.777	1.1	0.813[a]	0.843[b]	2
^{177}Lu	112.9	0.0398	0.0383	0.0394		3.1	0.0415	–	0.0421[c]	0.0446[d]	4
	208.4	0.0706	0.0680	0.0699		2.6	0.0714	–	0.0716[c]	0.0758[d]	3
^{192}Ir	296.0	1.09	–	1.14	1.13	1.7	1.14	1.2			
	308.5	1.13	–	1.18	1.17	1.7	1.18	1.2			
	316.5	3.14	–	3.27	3.25	1.7	3.26	1.2			
	468.1	1.78		1.85	1.84	1.7	1.87	1.2			
^{194}Ir	293.5	0.0199	–	0.0201	0.0204	1.8	0.0204	1.1			
	328.4	0.103	–	0.104	0.106	1.7	0.105	1.0			
	645.1	0.00924	–	0.00929	0.00948	1.8	0.00960	1.1			
	938.7	0.00455	–	0.00458	0.00467	2.0	0.00481	1.7			
	1150.8	0.00462	–	0.00465	0.00474	1.9	0.00476	1.1			

[a]Calculated with $g(T_n)=1$ and $Q_0=5.1$, [b]Calculated with $g(T_n)=1$ and $Q_0=3.95$, [c]Calculated with Holden $g(T_n)$ factor and $Q_0=3.2$, [d]Calculated with Van Sluijs $g(T_n)$ factor and $Q_E=2.84$ at the mean T_n of 36 °C

in [15] required significant corrections for epithermal neutrons ($f=18$ and $f=52$) and the neutron temperatures in [15], averaging 35 °C for ^{152}Eu, 30 °C for ^{152m}Eu and 36 °C for ^{177}Lu, were derived from the estimated temperature of the moderator surrounding the samples. In Table 3 the k_0 values measured in [15] have been recalculated using updated data. For ^{152}Eu, ^{152m}Eu, ^{154}Eu and ^{177}Lu the detection efficiencies needed for the calculation of the k_0 values were improved with new estimates of gamma attenuation in the samples. This reduced the k_0 values by 2.4% at 121.8 keV and by 0.9% at 1408 keV. For ^{152}Eu and ^{152m}Eu the k_0 values were recalculated with the Gryntakis $g(T_n)$ factors for better comparison with the values from [4], while the ^{177}Lu k_0 values are calculated with the Holden $g(T_n)$ factors and the measured Q_0 value of 3.2 as they were in [15]. For ^{154}Eu the k_0 values were calculated with $g(T_n)=1$ as was done previously [2, 15] and the correction for epithermal activation was carried out using the Q_0 value of 5.1 measured in [15] and also with the Q_0 value of 3.95 and effective resonance energy of 8.0 eV calculated recently by Van Sluijs [17]. For ^{152}Eu, ^{152m}Eu, and ^{177}Lu the k_0 values were calculated a second time using the parameters $g(T_n)$, $Q_E(\alpha,T_n)$ and $dQ(\alpha)$ of Van Sluijs in the recently published [20] extended version of the modified Høgdahl convention. The formula from [20] used to calculate these k_0 values corrects the non-1/v nuclide for epithermal neutron activation using a value of $Q_E(\alpha,T_n)$ calculated from tables of measured $\sigma(E)$ rather than a measured value of Q_0

$$k_0 = \frac{\left(\frac{N_p}{SDCW\rho t_m}\right)}{\left(\frac{N_p}{SDCwt_m}\right)^*} \frac{f + Q_0(\alpha)^*}{g(T_n)(f - dQ(\alpha)) + Q_E(\alpha, T_n)} \frac{\varepsilon_p^*}{\varepsilon_p} \tag{6}$$

where (*) designates the ^{197}Au comparator which is treated as a 1/v-nuclide.

Uncertainties

The uncertainty of the activity calculation includes the uncertainties of net peak areas N_p (< 0.5% for the main gamma lines) and sample weights W (< 0.1%). These give a statistical uncertainty of maximum 0.5% for the calculation of the k_0 values using Eq. 4. All other uncertainties related to the irradiation and the gamma counting such as the uncertainties of the time factors were not taken into account.

Furthermore, the main contributions for the systematic uncertainty are the concentration ρ of the standards (1.2% for Au and 0.5% for the rest) and the efficiency ratio between the gamma-energy of the comparator ^{198}Au at 411.8 keV and the gamma-energies, whose k_0 value should be determined. If the same efficiency curve is used, the efficiencies are correlated, and therefore the systematic errors of the efficiency calibration can be reduced significantly. According the approach of Smodiš and Bucar [22] the uncertainty of the efficiency ratio can be estimated to be lower than 1% for the gamma-lines with energy near the monitor energy of 411.8 keV and 1.8% for the higher energies up to 1408 keV, which is the highest gamma-energy concerned in this work.

In addition, the contribution of the g factor in Eq. 4 to the total uncertainty is dependent on the uncertainty of the temperature determined by using the thermometer labels and the slope of the chosen $g(T_n)$ function. Due to the discrete levels on the thermometer labels, the uncertainty of the temperature could be estimated as the half of the detection steps. That was 1.5 °C for the temperature range below 49 °C and 2.5 °C for the higher range [21] and created an uncertainty of the g factor less than 1% for ^{177}Lu and less than 0.2% for the other nuclides. The total uncertainty for the k_0 value can be calculated with a simple propagation formula:

$$\frac{\Delta k_0}{k_{0(x)}} = \sqrt{\left(\frac{\Delta\rho}{\rho}\right)^2_{\mathrm{Au}} + \left(\frac{\Delta\rho}{\rho}\right)^2_x + \left(\frac{\Delta N_p}{N_p}\right)^2_{\mathrm{Au}} + \left(\frac{\Delta N_p}{N_p}\right)^2_x + \left(\frac{\Delta\varepsilon_p}{\varepsilon_p}\right)^2 + \left(\frac{\Delta g}{g}\right)^2} \tag{7}$$

In the extended Høgdahl formalism (Eq. 2) the $g(T_n)$ factor of Au is set to 1. Actually, depending on the selection of the different data sources, the $g(T_n)$ factor of Au varies between 1.005 and 1.007 in the temperature range in this work. For a reactor with small f, this tiny deviation is much smaller than the contribution of the activation with epi-thermal neutrons (Q_0/f). However, for reactors with large f, the part of the epi-thermal neutron reaction is negligible, and the small deviation of the $g(T_n)$ factor of Au can be transferred to the k_0 determination proportionally. Therefore, all k_0 values in Table 3 should be theoretically adjusted upwards by about 0.5% if they are to be compared to previously measured values such as the ^{152}Eu and ^{152m}Eu k_0 values presented in [4] which were calculated using the Westcott formalism. However, in the extended version of the modified Høgdahl convention [14, 20] this factor of 1.005 is not used.

Discussion

The starting point for the k_0 determination for non-1/v nuclides is the measurement of the neutron temperature. According to Eq. 5 and Table 2, the whole temperature range of about 15 K determined in this work can create only a small change of the g factors. It is about 5% for ^{176}Lu, 1.5% for ^{151}Eu and less than 1% for the rest. That means that even a large error in the temperature determination, say 3 K, has a small influence on the measured k_0 values compared to the discrepancies between the different $g(T_n)$ factors.

Looking first at the results in Table 3 for ^{152}Eu, the Gryntakis k_0 values determined in this work have good agreement, usually within 2%, with the recommended values. This good agreement can be understood easily because the same

$g(T_n)$ factors were used for the calculation of the recommended values measured in the 1980s [2, 4], and it appears to confirm that the two sets of measurements are accurate. Only the value for the 121.8 keV gamma-ray is about 5% lower, possibly due to the interference with the 123 keV line of ^{154}Eu. The Van Sluijs k_0 values are generally about 5% lower, evidently because of the 5% higher Van Sluijs $g(T_n)$ factors.

All gamma lines of ^{152m}Eu suffer interference from ^{152}Eu. The contributions of the long-lived isotope ^{152}Eu to the net peak areas of the interfered gamma lines of ^{152m}Eu were corrected carefully. But it can still be found that Gryntakis and Holden k_0 values for ^{152m}Eu are 7% and 5% higher than the tentative values in the literature [4, 16, 18], respectively. It is postulated here that the ^{152m}Eu k_0 values of the present work are an improvement over the values of [4, 16] because of improved counting loss corrections with the modern gamma-ray spectrometers used.

^{154}Eu was always treated as a 1/v nuclide in the k_0 databases. The k_0 values of the present work in Table 3 calculated with $g = 1$, except for the weak 756.9 keV line, are all 1% to 4% greater than the recommended values, suggesting a 2% or 3% systematic difference. Any of the k_0 values for the 756.9 keV gamma-ray may be up to 10% too high due to the possible presence of interfering gamma-rays at 756 keV, including the 756.1 keV gamma-ray from ^{152}Eu. If the k_0 values of the present work for the stronger high-energy gamma-rays are compared to those measured in [15] corrected for epithermal neutrons using the measured Q_0 value of 5.1, the values of [15] are on average 2.6% higher. If the comparison is done with the values of [15] corrected for epithermal neutrons using the Q_0 value of 3.95 calculated in [17] from $\sigma(E)$, then the average difference rises to 6.3%. This change from 2.6% difference to 6.3% illustrates the uncertainty in correcting for epithermal neutrons, which can be eliminated by using a very well thermalized neutron spectrum as was done in this work.

For the highly non-1/v nuclides ^{152}Eu, ^{152m}Eu, and ^{177}Lu the k_0 values of the present work were compared to those of [15]. Only the more interference-free high-energy gamma lines were considered, and the comparison was done with the k_0 values calculated with the Van Sluijs [17] $g(T_n)$ factors. On average, the k_0 values of [15] were found to be 4.0% higher for ^{152}Eu, 5.7% higher for ^{152m}Eu, and 8.4% higher for the 208.4 keV gamma-ray of ^{177}Lu. For ^{152}Eu the average difference rises from 4.0% to 5.6%, if the comparison is made with Gryntakis k_0 values because in this case the Gryntakis k_0 values of [15] were corrected for epithermal activation using the measured Q_0 value of 0.3 rather than the Q_E value of 1.48 calculated by Van Sluijs [20].

The reasons for these differences are unknown but they suggest that k_0 NAA with non-1/v nuclides may continue to have uncertainty of 4% to 8% at different research reactors until further measurements are performed. The irradiation channels at the FRM II reactor are surrounded by heavy water (or light water in one position) while those at the SLOWPOKE reactor of [15] are surrounded by beryllium or light water and beryllium. Does a beryllium moderator change the shape of the thermal neutron spectrum so that the available $g(T_n)$ calculations are no longer valid?

The Lu standards made with the ICP-standard solution and the LuAl-alloy standards showed the same results in this work. The new k_0 values determined with $g(T_n)$ factors by Gryntakis and Van Sluijs are 2% lower than the recommended values for the 208 keV, but 4–5% lower for the 112 keV line. The Holden's k_0 values are much lower. The reason for lower k_0 values at the lower energy 112 keV might be the uncertainty of the efficiency calibration in this region. The calibration curve on the left side of the turning point has generally an extreme curvature and the nuclides in the QCY standards used for the calibration according to the certification have larger uncertainty, up to 3.5%, in this region than the nuclides with higher gamma energy.

Until now, the recommended k_0 values for ^{177}Lu have remained unchanged since they were calculated in the 1980s [2]. By definition, the k_0 value (relative to Au) is given as follows:

$$k_{0,Au} = \frac{\left(\frac{\theta\sigma_0 e_\gamma}{M}\right)}{\left(\frac{\theta\sigma_0 e_\gamma}{M}\right)_{\mathrm{Au}}} \tag{8}$$

where θ is the fractional isotopic abundance, σ_0 is the (n,γ) cross-section at neutron velocity of 2200 m s^{-1}, $e\gamma$ is the absolute gamma-intensity, M is the molar mass of the target element.

New evaluations show the absolute gamma intensity $e\gamma$ 5.6% lower for the main gamma line at 208 keV and 3% lower for the second gamma line at 112.9 keV compared to the old values used for the calculation of the recommended k_0 value [23, 24]. These new $e\gamma$ values have progressively been accepted in many databases worldwide [25–27]. New k_0 values for ^{177}Lu were calculated in this work with the new data shown in Table 4. They are lower than before and, for the 208 keV gamma-ray, the new calculated value is 5.7% lower than the old recommended value and 3.7% lower than the Van Sluijs k_0 value measured in the present work. These large variations in measured and calculated k_0 values for ^{177}Lu indicate a need for a new evaluation of the recommended k_0 values and new measurements at other research reactors.

For a long time ^{192}Ir was considered as a non-1/v nuclide. However, in the current version of the k_0 database [18], updated for ^{192}Ir and ^{194}Ir in 2015, the new k_0 values of ^{192}Ir are recommended after an evaluation of measurements at three laboratories in 2014 [12, 13] where it was assumed

Table 4 Different nuclear data used to calculate the k_0 values of ^{177}Lu

	M (g/mol)	θ (%)	σ_0 (barn)	$E\gamma$ (keV)	$e\gamma$ (old) (%) [2]	$E\gamma$ (new) (%) [23]	k_0, rec.* [16]	k_0 (new cal.)*
^{197}Au	196.97	100	98.66	411.8	95.62	–	1	–
^{176}Lu	174.97	2.59	2100	112.9	6.4(4)	6.20(7)	0.0415(26)	0.0402(5)
				208.4	11.0(4)	10.38(7)	0.0714(26)	0.0673(5)

*rec.: recommended values, new cal.: new calculated values in this work, $E\gamma$: gamma energy

that $g(T_n)$ was equal to unity and no temperature correction was performed. The new calculation by Van Sluijs et al. [11] confirmed this assumption that $g(T_n)$ factors for ^{192}Ir are very close to 1. The k_0 values of ^{192}Ir determined in this work have good agreement with the recommended values, whether they are calculated with these latest $g(T_n)$ factors or with $g(T_n)$ equal to 1. For ^{194}Ir, the k_0 values of the present work, calculated with the latest $g(T_n)$ factors or with $g(T_n)$ equal to 1, are also in good agreement with the recommended values.

As shown in this work, the determination of the k_0 values is dependent on the choice of the $g(T_n)$ factors; the k_0 value obtained depends on which $g(T_n)$ function was used. However, for the determination of concentrations with k_0 NAA, the product of k_0 and $g(T_n)$ plays the key role, see Eq. 3. Thus it doesn't matter which k_0 values are used as long as the same $g(T_n)$ function is used for the determination of a concentration as was used when the k_0 value was determined. Then the absolute magnitude of the $g(T_n)$ factor cancels and there is only a small temperature correction using $g(T_n)$. Even though different $g(T_n)$ calculations may differ by up to 5%, the uncertainty of k_0 NAA measurements due to the temperature correction with a consistent $g(T_n)$ will usually be less than 1%.

Conclusions

The present work confirmed the recommended k_0 values for ^{152}Eu, ^{154}Eu and ^{177}Lu using the $g(T_n)$ factors by Gryntakis and Kim. For ^{152m}Eu, the k_0 values of the present work are about 7% higher than the tentative values of the recommended k_0 database, possibly due to improved counting loss corrections with modern gamma-ray spectrometers; the values measured in [15] are even higher. The ^{152m}Eu k_0 values should be measured again at other laboratories.

The present results for ^{192}Ir and ^{194}Ir are consistent with the new recommended k_0 values of 2015 and they confirm that the both Ir isotopes can be treated as 1/v nuclides and no corrections with $g(T_n)$ factors are necessary.

The k_0 values of ^{177}Lu were calculated using new nuclear data. They are 3–6% less than the recommended values calculated in the 1980s. The measured k_0 values of the present work showed the same trend. Perhaps an update of the recommended k_0 values for ^{177}Lu should be considered.

The present work deals with the determination of k_0 factors in the ideal case of a highly thermalised neutron flux where the epithermal contribution is negligible. For practical use in not so well-thermalised conditions, the determination, evaluation and recommendation of "epithermal-related" data such as $\overline{E}_r$, Q_0 or s_0 should be an additional task to be undertaken.

In this work the situation of the various calculated $g(T_n)$ factors has been clarified. The $g(T_n)$ factors calculated by Van Sluijs et al. using more recent $\sigma(E)$ data are likely more accurate than the old factors. It is therefore recommended that they be used for future k_0 NAA work with non-1/v nuclides with the extended Høgdahl formalism, but they must be used with k_0 values adjusted for these same $g(T_n)$ factors. It has been shown that the influence of the neutron temperature measurement on the k_0 determination for all non-1/v nuclides is rather small; the estimation of neutron temperatures using thermometer labels as in this work or by moderator thermocouple readings as in [15] are of sufficient accuracy. More important is the choice of suitable calculated $g(T_n)$ factors. Different $g(T_n)$ factors lead to different k_0 values.

Finally, there is a general recommendation: the k_0 values for non-1/v nuclides should be used in k_0 NAA only in combination with the $g(T_n)$ factors which were used for the determination of these k_0 values.

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