

Experimental determination of k_0 , Q_0 , \bar{E}_r factors and neutron cross-sections for 41 isotopes of interest in Neutron Activation Analysis

F. Farina Arboccò · P. Vermaercke ·
K. Smits · L. Sneyers · K. Strijckmans

Received: 30 July 2012 / Published online: 28 August 2012
© Akadémiai Kiadó, Budapest, Hungary 2012

Abstract The k_0 -literature has been reviewed every decade but some of its nuclear data is still more than 30 years old. Sometimes Q_0 values were adopted from the nuclear data at that time or were experimentally determined by only 1 laboratory. Other isotopes were listed with accurate pairs of (k_0 , Q_0) values but were also quoted as candidates for redetermination for different reasons (i.e. imprecise cadmium transmission factors, half-lives). In this work we aim at the experimental re-determination of k_0 and Q_0 values for 41 isotopes of analytical interest while introducing a methodology for simultaneous \bar{E}_r and Q_0 determination employing N irradiation channels. In order to satisfy the metrological level required, up to 12 repeats per standard were irradiated in up to 4 irradiation channels of the Belgian Reactor 1 (BR1, SCK•CEN) having a wide spread in neutron characteristics. Our relative percentile differences to the literature values were usually $\leq 10\%$ for Q_0 factors, $\geq 25\%$ for \bar{E}_r values and $\leq 4\%$ for k_0 values. Our precision and accuracy are discussed thoroughly.

Keywords Neutron Activation Analysis · k_0 factor · Q_0 factor · Effective resonance energy \bar{E}_r · Neutron cross-section · Resonance integral

Electronic supplementary material The online version of this article (doi:10.1007/s10967-012-2132-0) contains supplementary material, which is available to authorized users.

F. Farina Arboccò (✉) · K. Strijckmans
Department of Analytical Chemistry, Ghent University,
Krijgslaan 281 S12, 9000 Ghent, Belgium
e-mail: fulvio.farina@ugent.be

F. Farina Arboccò · P. Vermaercke · K. Smits · L. Sneyers
k0-INAA Laboratory, Studiecentrum voor Kernenergie - Centre
d'étude de l'énergie nucléaire (SCK•CEN), Belgian Nuclear
Research Centre, Boeretang 200, 2400 Mol, Belgium

Introduction

The k_0 -standardization of Neutron Activation Analysis (k_0 -NAA) provides a user-friendly framework for the mapping of reactor channels and the multi-elemental quantification of up to 70 elements, without the need of extensive standard preparation. Its success depends on nuclear constants known as k_0 and Q_0 factors. These factors were experimentally determined mostly during the eighties, by the joint collaboration of 2 specialized laboratories and with the occasional participation of a third one [1–6]. Since then, the method has been successfully applied and extended worldwide but the experimental redetermination of these factors has been carried out for a few cases only. Some of its current literature was either adopted from nuclear data at that time or is correlated to fundamental parameters that have changed since then (half-lives, decay branching fractions). Among others cases, a few factors were reported by only 1 laboratory or were quoted as candidates for redetermination [5].

In this work we aimed at the experimental determination of k_0 nuclear data for 41 radioisotopes of analytical interest through the cadmium subtraction technique employed over 3 irradiation channels of the BR1 reactor (SCK•CEN), having considerable spread in neutron characteristics. Occasionally, a 4th channel having a *pure thermal neutron spectrum* (the *Cavity*) was employed. A method for simultaneous Q_0 and \bar{E}_r determination through N irradiation channels is proposed here (the *α -vector method*; $N \geq 2$). The method was tested on 25 investigated isotopes and for $N \leq 3$. We refer to [1–6] for a full description of the k_0 -method. De Corte [7] contains a survey of the main aspects. The results from this work are compiled and provided as electronic supplementary materials (*Online Resources*).

Experimental

Isotopes under investigation and chosen convention

For all the determinations use was made of the standards shown in Table 1. These materials, mostly foils and wires, are especially suited because of the minimum preparation required, negligible chances of mass losses, homogeneity, stability and low uncertainty in the certified content. Furthermore, several of the diluted Al alloys or spiked paper filters can be regarded as free from strong *neutron self-shielding* effects during irradiations. The thermal (index *t*) and epithermal (index *e*) self-shielding correction factors (G_t and G_e ; both ≤ 1) were obtained through the *MATSSF* and the *Sigmoid* calculation methods [8–10].

The use of the much simpler Høgdahl convention [11] over the Westcott formalism [12] was chosen since from Holden [13] it can be seen that only the rhenium isotopes deviate slightly from the $1/v$ expected behavior. Their deviations are $<0.8\%$ at 60°C , while the temperature of our channels is estimated to be 30°C .

Q_0 determination

For Q_0 determination, the standards (index *s*) were positioned within a 2 cm distance to the nearest comparator (index *c*) along the axis of a cylindrical polyethylene container (*PE* rabbit) following a sandwich pattern. Up to 10 samples per rabbit were irradiated on each opportunity. Table 2 summarizes their neutron characteristics, the typical irradiation times and rabbit sizes employed. These channel parameters are in agreement with our latest reported values [14]. From that work, the choice of thin plastic bags (0.05 mm thick) for packing of the samples, over typical *PE* vials (1 mm thick), was clarified from the statistical significance observed when comparing the calibration curves obtained under both scenarios.

Replicate samples were positioned inside 1 mm thick cadmium covers (index *Cd*) and were irradiated on the same irradiation positions. These covers filter the thermal component from the total capture reaction rate. Hence, the ratio of the *induced specific gamma activity* (*A*) between a bare and its Cd-covered replicate is an indicator of the thermal-to-epithermal sensitivity attainable for a given isotope on an irradiation channel (*cadmium ratio*; $R_{Cd} = A/A_{Cd}$). In this work we define a *normalized cadmium ratio* (r_{Cd}) in a threefold way as:

$$r_{Cd} \equiv \frac{G_e}{G_t} (R_{Cd} F_{Cd} - 1) \equiv \frac{f}{Q_\alpha} \equiv \frac{\varphi_t \sigma_0}{\varphi_e I_\alpha}, \quad (1)$$

where F_{Cd} is the *cadmium transmission factor* for the isotope of interest, f is the ratio between the thermal and the

epithermal *conventional neutron flux* (φ ; in $\text{n cm}^{-2} \text{s}^{-1}$), σ_0 is the *thermal neutron cross-section* and I_α is the *resonance integral* (both in barns). The subscript α corresponds to a channel-specific dimensionless parameter necessary for an accurate mapping of the $\sim 1/E^{1+\alpha}$ distribution of epithermal neutrons observed in reactor channels. If $\alpha = 0$, the r_{Cd} factor is just inversely proportional to the Q_0 factor.

By writing eq. (1) for the comparator as well, is possible to avoid f :

$$Q_\alpha = \frac{r_{Cd,c}}{r_{Cd}} Q_{\alpha,c} \quad (2)$$

and shift the problem to the $Q_\alpha \rightarrow Q_0$ conversion. This is done by means of a theoretical expression that introduces the concept of a *hypothetical single-resonance energy* (or *effective resonance energy*; \bar{E}_r). This single-resonance would give the same resonance activation rate as all the actual resonances combined. The conversion function proposed in [15, 16] is:

$$Q_\alpha = (Q_0 - C_0) \bar{E}_r^{-\alpha} + C_\alpha \quad (3)$$

where $C_\alpha = C_0(2\alpha + 1)^{-1}(E_{Cd})^{-\alpha}$, $C_0 = 0.429$ and $E_{Cd} = 0.55$ eV is the cadmium cut-off energy for 1 mm Cd-covers. The α value of a channel can be found experimentally by bare and Cd-covered irradiations of other standards with well-known nuclear data, after computing the slope ($-\alpha$) on the resulting Log–Log plot of $(\bar{E}_{r,i}^{-\alpha}/r_{Cd,i})$ versus $\bar{E}_{r,i}$, when expressed for $i = 1, \dots, n$ isotopes. The parameter f is related to the intercept in this kind of plot [17].

Finally, the Q_0 problem is classically solved by writing eq. (3) for the standard and the comparator and their substitutions on eq. (2), assuming that the \bar{E}_r values are well-known. While this is obviously true for the chosen comparator, there is still doubt about the choice of \bar{E}_r values for certain isotopes, since these have been officially adopted from early calculations.

The α -vector method: simultaneous Q_0 and \bar{E}_r determination with N irradiation channels

The typical approach for \bar{E}_r determination is either: (a) to calculate it from weighted resonance data, assuming that it is independent of α (at least to a first order approximation) or, (b) to determine it experimentally from standard vs. comparator Cd-ratios obtained from 1 irradiation channel, assuming their Q_0 factors are well-known [18]. Clearly, the latter option would lead to a vicious circle when performing Q_0 determination.

The Q_0 factors obtained from largely different irradiation channels (spread in α values) can only be consistent if this simplified \bar{E}_r model is accurate irrespectively of the irradiation channel employed. By defining an experimental parameter Y_α for a given isotope and a given irradiation channel (index *j*) as:

Table 1 Materials employed in this work: provider, matrix, certified elemental content, average thickness (*D*) and their calculated self-shielding factors. The target (*T*) and formed isotopes (*FI*) and the reason for their investigation

Provider	Material matrix	Element	Weight %	<i>D</i> (mm)	<i>T</i>	Neutron self-shielding		FI	Investigated because
						G_t	G_e		
Fluka	High purity NaCl	Na	39.320	0.15	²³ Na	0.970	1.000	²⁴ Na	Q_0 adopted
Alfa Aesar	Liquid on paper		0.1			1.000			
Goodfellow	Foil	Mg	99.99+	0.1	²⁶ Mg	0.999	1.000	²⁷ Mg	Q_0 adopted
Goodfellow	Foil	Al	99.99+	0.015	²⁷ Al	0.999	1.000	²⁸ Al	Q_0 adopted
IRMM	Al alloys			0.1		0.999	0.996		
Fluka	High purity NaCl	Cl	60.680	0.15	³⁷ Cl	0.970	1.000	³⁸ Cl	Q_0 adopted
IRMM	Foil, Al alloy	Sc	1	0.1	⁴⁵ Sc	0.999	1.000	⁴⁶ Sc	Q_0 adopted; discrepant \bar{E}_r
CBNM	Foil	Ti	99.99+	0.1	⁵⁰ Ti	0.987	1.000	⁵¹ Ti	Q_0 adopted
CBNM	Foil	V	99.99+	0.1	⁵¹ V	0.988	1.000	⁵² V	Q_0 adopted
IRMM	Foil, Al alloy	Mn	1	0.1	⁵⁵ Mn	0.998	1.000	⁵⁶ Mn	Calibration, cross-section standard
IRMM	Wire, Al alloy	Fe	0.2	1	⁵⁸ Fe	0.999	1.000	⁵⁹ Fe	Calibration
Goodfellow	Foil		99.99	0.5		0.975			
			99.99+	0.25		0.984			
IRMM	Foil, Al alloy	Co	1	0.1	⁵⁹ Co	0.999	0.994	⁶⁰ Co	Calibration, cross-section standard
								^{60m} Co	
CBNM	Foil	Ni	99.99+	0.1	⁶⁴ Ni	0.986	1.000	⁶⁵ Ni	Q_0 adopted
IRMM	Foil, Al alloy	Cu	10	0.1	⁶³ Cu	0.997	0.998	⁶⁴ Cu	All Q_0 factors adopted
Goodfellow	Wire		99.99	1		0.980	0.790		
CBNM	Foil		99.99+	0.1		0.990	0.890		
					⁶⁵ Cu		0.998	⁶⁶ Cu	$F_{Cd} = 1.034$ not quoted in [6, 24]
Goodfellow	Foil	Zn	99.95	0.025	⁶⁴ Zn	0.999	1.000	⁶⁵ Zn	Calibration
					⁶⁸ Zn	0.999	1.000	^{9m} Zn	
IRMM	Wire, Al alloy	Ga	1	1	⁷¹ Ga	0.995	0.990	⁷² Ga	To provide additional k_0 values
Alfa Aesar	Liquid on paper	As	0.1	1.5	⁷⁵ As	0.999	0.998	⁷⁶ As	Q_0 adopted
NIST			1			0.996	0.978		
Alfa Aesar	Liquid on paper	Br	0.1	1.5	⁸¹ Br	0.999	0.991	⁸² Br	Calibration; efficiency control
	High purity KBrO ₃		47.847	0.15		0.979	0.640		
NIST	Liquid on paper	Rb	1	1.5	⁸⁵ Rb	0.998	0.997	⁸⁶ Rb	Redetermination desired
					⁸⁷ Rb		0.998	⁸⁸ Rb	
Dr. Hult	Foil	Y	99.3	0.1	⁸⁹ Y	0.998	0.982	^{90m} Y	Observed Q_0 discrepancy
Fluka	High purity Y ₂ O ₃		78.744	0.15		0.997	0.975		
Alfa Aesar	Foil	Zr	99.8	0.127	⁹⁴ Zr	0.999	0.977	⁹⁵ Zr	Calibration
								⁹⁵ Nb	Change in branching factors
IRMM	Foil, Al alloy	Nb	10	0.1	⁹³ Nb	0.999	0.998	^{94m} Nb	Calibration; short-lived control
CBNM	Foil		99.99+	0.022		0.999	0.985		
			99.99+	0.076		0.998	0.948		
Goodfellow	Foil	Mo	99.9	0.025	⁹⁸ Mo	0.998	0.975	⁹⁹ Mo	Calibration
								^{99m} Tc	
IRMM	Foil, Al alloy	Ag	1	0.1	¹⁰⁷ Ag	0.999	0.999	¹⁰⁸ Ag	Q_0 adopted
Goodfellow	Wire		99.9	1		0.812	0.582		
Sigma-Aldrich	High purity BaO	Ba	89.565	0.15	¹³⁰ Ba	0.992	0.991	¹³¹ Ba	Q_0 adopted; k_0 discrepancies in [27]

Table 1 continued

Provider	Material matrix	Element	Weight %	<i>D</i> (mm)	<i>T</i>	Neutron self-shielding		FI	Investigated because
						<i>G_t</i>	<i>G_e</i>		
IRMM	Foil, Al alloy	La	1	0.1	¹³² Ba		0.991	^{133m} Ba	<i>k</i> ₀ factors not recommended
					¹³⁴ Ba		1.000	^{135m} Ba	Not included in the <i>k</i> ₀ literature
					¹³⁸ Ba		0.975	¹³⁹ Ba	<i>Q</i> ₀ adopted
					¹³⁹ La	0.995	1.000	¹⁴⁰ La	<i>Q</i> ₀ determined on 1 laboratory
IRMM	Wire, Al alloy	Pr	1.25	1	¹⁴¹ Pr	0.995	0.990	¹⁴² Pr	<i>Q</i> ₀ adopted
IRMM	Wire, Al alloy	Sm	0.008	0.37	¹⁵² Sm	0.995	1.000	¹⁵³ Sm	Quoted with possible <i>F_{Cd}</i> < 1
IRMM	Ingot, Al alloy	Tb	1	3	¹⁵⁹ Tb	0.987	1.000	¹⁶⁰ Tb	Calibration; efficiency control
IRMM	Foil, Al alloy	Ho	5	0.1	¹⁶⁵ Ho	0.998	0.984	¹⁶⁶ Ho	<i>F_{Cd}</i> = 0.99 not quoted in [6, 24]
IRMM	Wire, Al alloy	Re	0.118	1	¹⁸⁵ Re	0.996	0.996	¹⁸⁶ Re	<i>F_{Cd}</i> = 0.98 not quoted in [6, 24]
					¹⁸⁷ Re		1.000	¹⁸⁸ Re	No <i>k</i> ₀ values for decay scheme IV/b
IRMM	Wire, Al alloy	W	0.01	0.37	¹⁸⁶ W	0.998	0.998	¹⁸⁷ W	<i>F_{Cd}</i> = 0.908 not quoted in [6, 24]
			0.2	0.5		0.996	0.996		Half-life changed
CBNM	Foil, Al alloy		1	0.1		0.995	0.995		
IRMM	Foil, Al alloy	Au	0.1	0.1	¹⁹⁷ Au	0.999	0.998	¹⁹⁸ Au	Ultimate comparator (IRMM-530R)

$$\begin{aligned}
 Y_{\alpha_j} &\equiv \ln\{(Q_0 - C_0)\bar{E}_r^{-\alpha_j}\} \\
 &\equiv \ln\left\{(r_{Cd,c}/r_{Cd})_j(Q_{0,c} - C_0)\bar{E}_{r,c}^{-\alpha_j}\right. \\
 &\quad \left.+ [(r_{Cd,c}/r_{Cd})_j - 1]C_{\alpha_j}\right\} \tag{4}
 \end{aligned}$$

it is possible to express the problem as a transcendental vector system:

$$\vec{Y}_{\vec{\alpha}} = b - \vec{\alpha}m \tag{5}$$

and to determine $\bar{E}_r = \exp(m)$ and $Q_0 = \exp(b) + C_0$ from the scalars *b* and *m* after solving the system through

substitution of the α -vector from the *N* channels. Alternatively, a graphical solution can be obtained from a linear regression on a *Y_α* versus α plot, since *b* corresponds to the intercept and *m* to the slope. The parameter *b* changes sign for $Q_0 < 1.429$, while *m* does so for $\bar{E}_r < 1$ eV. An interesting case is ⁴⁵Sc, since it is the best example of a $Q_0 \approx C_0$ situation.

For a few cases the effective resonance energy is still a function of α , so the expression $\bar{E}_{r,\alpha} = \bar{E}_r \exp(-p\alpha)$ should be used instead of \bar{E}_r . These *p* values are reported in [5] but not in the latest official compilation [6]. For ⁶⁴Zn, ⁹⁸Mo

Table 2 Neutron parameters (*f*, α) for the irradiation channels employed in this work (*Y4*, *S84*, *X26* and the *Cavity*) and their confidence intervals (2 σ). Radius (*r*) and length (*l*) of these channels and irradiation containers (in mm; in that order) and typical irradiation times (*t_{irr}*) applied

<i>Y4</i>		<i>S84</i>		<i>X26</i>		<i>Cavity</i>	
<i>f</i>	α	<i>f</i>	α	<i>f</i>	α	<i>f</i>	α
38.2 ± 1.0	0.066 ± 0.006	16.4 ± 0.8	-0.003 ± 0.010	95 ± 5	0.11 ± 0.01	70000	-
<i>r</i>	<i>l</i>	<i>r</i>	<i>l</i>	<i>r</i>	<i>l</i>	<i>r</i>	<i>l</i>
50	500	40	300	40	300	40	300
11.1	71	10	53	20	80	15	60
<i>t_{irr}</i> = 7 h		<i>t_{irr}</i> = 1 or 7 h		<i>t_{irr}</i> = 3 or 7 h		<i>t_{irr}</i> = 2 or 4 h	

and ^{187}Re , the p values are 1.17, 1.82 and 0.95 (respectively). In this case, the α -vector method would aim at solving the polynomial vector system:

$$\vec{Y}_{\vec{\alpha}} = b - m\vec{\alpha} + p|\vec{\alpha}|^2 \quad (6)$$

k_0 determination

From the ratio of A values between the standard and the comparator, a k_0 factor can be calculated assuming that f , α , Q_0 and \bar{E}_r are known. Alternatively, in terms of specific activities from bare and Cd-covered irradiations, a k_0 can be computed from:

$$k_{0,s,c} = \frac{(A - A_{Cd}/F_{Cd})_s G_{t,c}}{(A - A_{Cd}/F_{Cd})_c G_{t,s}} \quad (7)$$

This avoids the introduction of f , α , Q_0 and \bar{E}_r values and their uncertainties into the analytical result, which are of greater magnitude than the ones arising from counting statistics. For the experiments performed in the Cavity a k_0 factor was obtained from eq. (7) by setting $A_{Cd} = 0$, that is, the factor is just proportional to the ratio between activities and thermal self-shielding correction factors (no epithermal component). In this sense, the Cavity is a unique tool, but its low neutron flux limits our sensitivity ($7 \times 10^8 \text{ n cm}^{-2}/\text{s}$).

On the traceability, accuracy and precision of the determinations

When computing a Q_0 factor with aid of f estimated from a calibration curve, the correlation will be weighted in favor of the average neutron flux perceived by the calibration isotopes and to their nuclear data. A Q_0 can be computed in this way without the need of the comparator data at all (if it is not part of this calibration set). On the other hand, the introduction of the f parameter for Q_0 and/or k_0 determination is inevitable when the Cd-ratio was not determined, for instance, when adopting a Q_0 or when assuming that $f \gg Q_0$. Since the application of k_0 -NAA relies on the specific activities from a comparator that was co-irradiated next to the sample, it follows that its Cd-ratio would be a better indicator of the neutron flux in that specific region. This alternative bears the lowest uncertainty and maintains the correlation in favor of the comparator results and its nuclear data. In this work all of our reported k_0 values were determined experimentally through the use of Cd-subtraction (or ratio) technique. These values are therefore independent from the chosen f , α , Q_0 and \bar{E}_r values.

The precision of our measurement setup was tested by taking on average up to 20 measurements per standard on 5 calibrated source-detector distances (0, 3, 15, 20 and 27 cm) on 5 HPGe detectors equipped with LFC modules (for dead-

time, pulse pile-up corrections). The measurements aimed at $\leq 0.3\%$ uncertainty in counting statistics when possible. The photo-peaks deconvolutions were done with the software *HyperLab* [19]. For Q_0 determination the results from all detector positions available were employed while for k_0 determination only the results from the 2 farthest positions were considered. These positions pose negligible coincidence gamma (*COI*) and secondary unwanted effects (i.e. peak distortions, higher dead-times, pulse pile-up) as compared to closer ones. The *COI* and the efficiency calculations were performed by a software adaptation of the efficiency transfer code *SOLCOI* [20] that employs mass attenuation coefficients from the *NIST X-COM* online database [21] for any specific gamma energy of interest.

A computer code was developed in *Structured Query Language (SQL)* 2008 and *C# 4.0* programming languages, comprising the latest official k_0 -NAA literature and formulae. With aid of *SQL* constrains and parent/child data-table relationships it was possible to treat great amounts of data points while avoiding systematic errors from human input and data redundancy.

The uncertainty from the laboratory setup on a specific activity and/or Cd-ratio can be estimated from the individual uncertainty contributions listed in Table 3. A Cd-ratio has the uncertainty arising from the counting statistics of 2 samples but the *COI*, efficiency and elemental content contributions are cancelled by employing the same detectors and measurement positions (total of 1 %, 1s). The uncertainty contribution from F_{Cd} is usually low ($< 0.5\%$; except for ^{186}W , ^{185}Re , ^{65}Cu and ^{165}Ho), so the accuracy on a determined k_0 factor should not be worse than 4 % (at 2s). Thus, a k_0 factor was *recommended* when the standard deviation from 4–6 repeats per standard (per irradiation channel) was lower than this threshold value.

Results and discussion

The Online Resource 1 illustrates the linear fits on the Y_α versus α plots obtained for 25 investigated isotopes. The best α -vector was obtained after several iterations, aiming at the lowest sum of residuals on all the linear regressions from the calibration isotopes. This can be done, for instance; with the *Solver tool* provided in the software *Excel* [22]. In general, good agreement is observed between our linear fits (dashed black lines) and the expected values (dotted blue lines) when comparing biases in intercept (b) and slope (m) divergences. The extrapolated \bar{E}_r and Q_0 values and their uncertainty propagation are summarized in the *Online Resource 2*. A comparison is made with the official literature values from [6, 23, 24] and other sources [25, 26].

Only 11 isotopes reported a >10 % relative difference to the official Q_0 values. However 9 of them have adopted Q_0 values (20 % relative uncertainty; 2s) and are of very low magnitude. Under bare irradiation conditions, the accurate knowledge of a low- Q_0 isotope is not necessary (hence the reason for the adoption). Nowadays, irradiations under Cd are useful for avoiding spectral interferences from activation of unwanted nuclides such as ^{27}Al , ^{51}V , ^{23}Na . This is sometimes necessary for instance when performing trace analysis on pure Al and/or Ti matrices, vitamins, etc. Although the idea is to substantially reduce the activation of these unwanted nuclides, one can be still interested in accurately quantifying these elements for practical reasons (internal comparator/control), for which the accurate knowledge of the Q_0 factor is fundamental. Hence, we recommend our values over the adopted ones, especially for the As and Ba isotopes (high Q_0) since our findings are also in agreement with other authors [27]. The remaining discrepancies were for ^{87}Rb and ^{89}Y (11 and 41 %, respectively), but unfortunately these isotopes could only be tested on our sole irradiation channel suited for short-lived investigation (S84). Our Q_0 value for ^{87}Rb is not statistically significant at 3s level. For ^{89}Y , we tested first the same compound (pure yttrium oxide) employed in [5]. In virtue of the discrepancy, another matrix (pure yttrium foil) was tested, which confirmed our first results. The observed difference could not be explained in terms of self-shielding effects (negligible; see Table 1), F_{Cd} (equal to unity), decay scheme (I ; normal) and half-life (not changed).

Our Q_0 results can be weighted along the official ones if the same \bar{E}_r has been adopted (the classical method). Under an experimental approach, then (Q_0 , \bar{E}_r) values obtained from the α -vector method should be chosen instead. ^{94}Zr is quite a difficult case because of its high \bar{E}_r value. It has been re-determined several times, with results differing from author to author. Our Q_0 results are in better agreement with those reported in [4, 5, 28, 29].

Our relative differences to the official \bar{E}_r values were ≥ 25 % for 7 isotopes. This had a <4 % influence on the resulting Q_0 when alternating both inputs. In this respect, the choice of \bar{E}_r value does not play a significant role. However, 6 isotopes have adopted Q_0 factors so the accuracy of their \bar{E}_r values (adopted) has not been studied before. Several of our experimental \bar{E}_r values are in better agreement with reported values from others authors (i. e. for Mn, Zn, As, Sc and Mo isotopes).

The F_{Cd} factors are not quoted in the latest official references [6, 24] but can be found in [5, 23], from where we took $F_{Cd} = 0.98$ for ^{185}Re , 1.034 for ^{65}Cu , 0.99 for ^{165}Ho , 0.908 for ^{186}W and 0.991 for the comparator (^{197}Au). No discrepancies were observed in Q_0 factors for

these isotopes except for ^{185}Re (5 %). The 1.5 % lower half-life employed in the past [5] does not seem to be the reason for the disagreement.

The k_0 factors and thermal neutron cross-sections, derived with aid of data from [30–34] and the k_0 definition, are given in the *Online Resource 3*. In general, the percent relative differences between our k_0 factors and the official ones are usually ≤ 4 %, which is reasonable, since this is our worst expected precision (see Table 3) at 2s level. The results are satisfactory because less than 20 isotopes reported discrepancies >2 % (yet ≤ 4 %). For 13 isotopes the discrepancies are ≤ 1 %, which confirms the accuracy of our efficiency curves for most of the energy range dealt with. The k_0 -data for $^{134}\text{Ba}(n,\gamma)$ $^{135\text{m}}\text{Ba}$ and ^{188}Re (under decay scheme *IV/b*) are proposed here for the first time.

The only k_0 -factors with observed >5 % relative difference correspond to the ^{108}Ag , ^{88}Rb , ^{152}Sm and ^{131}Ba isotopes. The 434 keV line of ^{108}Ag seems biased in 6 % but the 633 keV line is in agreement with the literature instead. The k_0 standardization of ^{131}Ba has been a historical problem, worsened because of accidental typos or inaccurate Q_0 adoptions, as has been explained in [27]. Our Q_0 and k_0 factors for this reaction are in better agreement with [35]. The 511 keV k_0 factors for ^{64}Cu and ^{65}Zn are 10 % different and had always represented a spectrometric challenge. We aimed at ≤ 0.7 % uncertainty in counting statistics in order to facilitate the deconvolution while increasing the number of measurements to compensate. For ^{152}Sm , the 97.4 keV line should not be recommended as it is severely affected by the Compton from the more prominent 103.2 keV line.

Table 3 Maximum expected uncertainty on an induced gamma specific activity (A) and a cadmium ratio (R_{Cd}) at 1s confidence level. A cadmium ratio uncertainty corresponds to the quadratic summation of twice ($\times 2$) the uncertainty reported

% uncertainty (1s) from		Contributing to	
		A_{sp}	R_{Cd}
Reference efficiency	0.5	Yes	No
Efficiency transfer	0.8	Yes	No
COI corrections	0.6	Yes	No
Sample positioning	0.1	Yes	Yes
Weighting	0.1	Yes	Yes
Certified elemental content	0.5	Yes	No
Mass losses for spiked standard	0.5	Yes	Yes
Moisture content of compound	0.1	Yes	No
Pulse pile-ups	0.3	Yes	Yes
Timing	0.1	Yes	Yes
Half-life	0.2	Yes	Yes
Counting statistics	0.3	Yes	Yes
% Total uncertainty (1s)	1.4		0.7 ($\times 2$)

Although our results are from 1 independent laboratory, we had a clear advantage on several technological improvements and knowledge of nuclear data over researchers from 30 years ago. For instance, a typical ^{75}As spectrum is almost entirely made up of duplets, a limiting factor depending on the resolution of the detectors employed. We recommend the use of the 657.1 keV line over the others. For ^{95}Zr , the values are in agreement with [3–5, 28, 36] and the 765.8 keV factor for ^{95}Nb is only 1 % different when employing the official F_i values. Nonetheless, coherence with its mother nuclide (^{95}Zr) is not observed in terms of cross-sections. Instead, when the more recent and accurate F_i values from [32] are employed (where F_{24}/F_2F_3 is 5 % lower), the agreement is reached. We computed this k_0 factor for both cases but we recommend the use of the underlined one along the updated F_i values.

Conclusions

The validity of the Breit-Wigner expression and the \bar{E}_r concept was demonstrated here through the α -vector method. This method also offers a mechanism for studying the linearity of α and the generally assumed α -independence on the \bar{E}_r value. Apart from the half-lives and decay branching fractions, the \bar{E}_r values are the only values left that are still borrowed directly from the literature, so the accuracy of a theoretical \bar{E}_r value can be tested through this method. On overall, the experimental \bar{E}_r values from this work were ≥ 25 % different than the literature ones, but this had little influence on the Q_0 determination. Yet, since these are hypothetical (or effective) values calculated from approximate models and sometimes incomplete resonance data, we would like to encourage the adoption of our experimental values instead. This is because the calibration (f and α -determination) of a reactor irradiation channel does rely on the accurate knowledge of \bar{E}_r factors over the chosen energy range. The validation of a given \bar{E}_r though a high rank (and spread) α -vector aims at expanding the diversity of isotopes available for calibration (or comparator) purposes. We believe that our rank-3 α -vector satisfied these requirements but it would be better to construct (include) larger datasets from coordinated worldwide exercises.

When considering how our factors would influence a previous analytical result (elemental concentration), it is clear that differences in k_0 values will propagate entirely to it. The differences in Q_0 factors will propagate fully if $\alpha = 0$ and/or cadmium irradiations were performed ($f = 0$), but become negligible if f is sufficiently high ($f \gg Q_\alpha$) under bare irradiations. Our Q_0 factors are ≤ 10 % different and the

k_0 factors ≤ 4 %, on average. Our uncertainties are usually lower. New k_0 and Q_0 factors were proposed for ^{188}Re under the decay scheme IV/b and for $^{134}\text{Ba}(n,\gamma)^{135\text{m}}\text{Ba}$. Several recommended k_0 -factors are introduced for multi-gamma radioisotopes (i.e. ^{72}Ga , ^{140}La and ^{76}As).

After more than 30 years of development and applications of this technique, several improvements in instrumentation, software concepts, etc., we believe that more laboratories should embrace a general re-determination of all these factors or new ones (i.e. ^{192}Ir) when possible, as it is the only way to enrich the statistical pool if an accuracy better than 4–5 % at $2s$ is desired.

Acknowledgments This work would have not been possible without the technical support from Karen Contreras Nesbit. The corresponding author appreciates her unconditional help.

References

1. Simonits A, De Corte F, Hoste J (1975) J Radioanal Nucl Chem 24:31–46
2. Simonits A, Moens L, De Corte F, De Wispeleare A, Elek A, Hoste J (1980) J Radioanal Nucl Chem 60:461–516
3. Moens L, De Corte F, De Wispeleare A, Hoste J, Simonits A, Elek A, Szabo E (1984) J Radioanal Nucl Chem 82:385–482
4. De Corte F, Simonits A, De Wispeleare A, Elek A (1989) J Radioanal Nucl Chem 133:43–130
5. De Corte F (1987) In: The k_0 -standardization method: a move to the optimization of neutron activation analysis. Aggregate Thesis, Gent University, Belgium
6. De Corte F, Simonits A (2003) At Data Nucl Data Tables 85:47–67
7. De Corte F (2001) J Radioanal Nucl Chem 248:13
8. IAEA (2009) MATSSF Program. Austria. <http://www-nds.iaea.org/naa/matssf/>. Accessed 10 May 2011
9. Chilean C, St-Pierre J, Kennedy G (2008) J Radioanal Nucl Chem 278(3):745–749
10. Farina Arboccò F, Vermaercke P, Sneyers L, Strijckmans K (2012) J Radioanal Nucl Chem 291:529–534
11. Høgdahl OT (1965) In: Proceedings of the symposium on radiochemical methods of analysis (Salzburg, 1964) IAEA (Vienna), vol 1, p 23
12. De Corte F, Bellemans F, De Neve P, Simonits A (1994) J Radioanal Nucl Chem 179:93–103
13. Holden N (1999) Pure Appl Chem 71(12):2309–2315
14. Farina Arboccò F, Strijckmans K, Vermaercke P, Verheyen L, Sneyers L (2010) J Radioanal Nucl Chem 286(2):569–575
15. Moens L, De Corte F, Simonits A, De Wispeleare A, Hoste J (1979) J Radioanal Nucl Chem 52(2):379–387
16. De Corte F, Moens L, Jovanovic S, Simonits A, De Wispeleare A (1986) J Radioanal Nucl Chem 102:37–57
17. De Corte F, Sordo-El Hammami K, Moens L, Simonits A, De Wispeleare A, Hoste J (1981) J Radioanal Nucl Chem 62:209–255
18. Simonits A, Jovanovic S, De Corte F, Moens L, Hoste J (1984) J Radioanal Nucl Chem 82(1):169–179
19. Simonits A, Östör J, Kálvin S, Fazekas B (2003) J Radioanal Nucl Chem 257(3):589–595
20. KAYZERO/SOLCOI® (2003) User's Manual for reactor neutron activation analysis using the k_0 -standardization method, Ver. 5a, 10–120

21. NIST (2011) XCOM: Photon cross sections database. USA <http://www.nist.gov/pml/data/xcom/index.cfm>. Accessed 1 Dec 2011
22. Microsoft Office (2012) Introduction to optimization with the Excel Solver tool, USA. <http://office.microsoft.com/en-us/excel-help/introduction-to-optimization-with-the-excel-solver-tool-HA01124595.aspx>. Accessed 1 Feb 2012
23. IUPAC (2004) Compilation of k_0 and related data for NAA in the form of electronic database, USA. <http://iupac.org/web/ins/2001-075-1-500>. Accessed 1 Dec 2011
24. k_0 -International Scientific Committee (2012) Classic k_0 Database, International. http://www.kayzero.com/k0naa/k0naa/News/Artikelen/2012/3/25_The_IUPAC_databasa.html. Accessed 25 Mar 2012
25. National Nuclear Data Center - BNL (2012) NuDat 2.6: Nuclear Structure and Decay Data, USA. <http://www.nndc.bnl.gov/nudat2/>. Accessed 1 Feb 2012
26. Nuclear Data Center - JAEA (2012) JENDL 4.0: Japanese Evaluated Nuclear Data Library, Japan. <http://wwwndc.jaea.go.jp/jendl/j40/j40.html>. Accessed 3 Feb 2012
27. De Corte F (2010) Nucl Instrum Meth A 622:373–376
28. Simonits A, De Corte F, De Wispelaere A, Hoste J (1987) J Radioanal Nucl Chem 113:187–197
29. Smodis B, Trkov A, Jacimovic R (2003) J Radioanal Nucl Chem 257:481–487
30. Wieser ME, Coplen TB (2011) Pure Appl Chem 83(2):359–396
31. Berglund M, Wieser ME (2011) Pure Appl Chem 83(2):397–410
32. CEA - Laboratoire National Henri Becquerel (2012) Recommended Data. http://www.nucleide.org/DDEP_WG/DDEPdata.htm. Accessed 2 Feb 2012
33. Mughabghab SF (2006) Atlas of Neutron Resonances, 5th edn. Elsevier, The Netherlands
34. University of Delaware (2011) Uncertainty, precision and accuracy, USA. <http://www.udel.edu/pchem/C446/error.pdf>. Accessed 1 Dec 2011
35. Kennedy G, St-Pierre J (2006) Nucl Instrum Meth A 564: 669–674
36. Lin X, Gerstenberg H, Lieser von Gostomski Ch, Henkelmann R, Türler A, Rossbach M (2009) Appl Radiat Isot 67:2092–2096