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A DEMAND FOR CONSISTENCY OF NUCLEAR DATA RELATED TO REACTOR NEUTRON ACTIVATION ANALYSIS

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Abstract : A plea is made for the utmost alertness to aspects of consistency when evaluating 2200 ms^{-1}(n,γ) cross-sections [σ_0], which are (or should be) reported together with data for the isotopic abundances [θ], the absolute gamma-intensities [γ] and the half-lives [T]. Indeed, θ, γ and T serve as input (and should thus be quoted by the experimentalists) in σ_0-determinations according to the activation method, upon which evaluations are largely based. In the extreme, it can be argued that the consistency of these data, and not their accuracy, is the matter of primary concern in absolutely standardized reactor neutron activation analysis, where selected literature data for σ_0, θ, γ and T have to be combined.

(cross section; isotopic abundance; gamma-intensity; half-life; consistency; traceability; activation; n,gamma; evaluation)

Introduction

In absolutely standardized (parametric) reactor neutron activation analysis (NAA) involving gamma-spectrometry, the most crucial input parameters are the molar mass (M), the isotopic abundance (θ), the 2200 ms^{-1}(n,γ) cross-section (σ_0), the absolute gamma-intensity (γ) and the half-life (T), which are figuring in the expression for concentration calculation as :

$$\text{concentration} \propto \frac{1}{SDC}\frac{M}{\theta\gamma\sigma_0} \qquad (1)$$

with :

$S = 1-\exp(-\lambda t_{irr})$; t_{irr} = irradiation time ;

$D = \exp(-\lambda t_d)$; t_d = decay time ;

$C = [1-\exp(-\lambda t_m)]/\lambda t_m$; t_m = measuring time ;

$\lambda = (\ln 2)/T$; T = half-life.

For half-lives much larger than t_{irr}, t_d and t_m, Eq.(1) becomes to a good approximation :

$$\text{concentration} \propto T\frac{M}{\theta\gamma\sigma_0} \qquad (2)$$

As to both accuracy and traceability of NAA-results it is thus essential that evaluators of σ_0-values consider the correlation between all parameters involved. Indeed, many experimental σ_0's (the basis of evaluations) originate from determinations according to the activation method with natural targets and gamma-spectrometry, where M, θ, γ and T appear in the relevant expression as :

$$\sigma_0 \propto \frac{1}{SDC}\frac{M}{\theta\gamma} \qquad (3)$$

or, for T >> t_{irr}, t_d and t_m :

$$\sigma_0 \propto T\frac{M}{\theta\gamma} \qquad (4)$$

The present paper reveals some of the observations made during a study of NAA-standardization methods /1/.

A selection of problematic cases

Probably the best known example of preserving the consistency concerns the θ and σ_0 values for Fe-58(n,γ)Fe-59, as shown in Table 1 for a number of compilation works. The sudden change of the accepted θ-value from 0.3-0.31% to 0.28% (following the experimental results of James et al. /10/ and Schmidt et al. /11/, and the subsequent evaluation of IUPAC-SAIC /12/) was accompanied by a shift of σ_0 from 1.15-1.16 b to 1.28 b, thus reflecting the constancy of θ x σ_0. The renormalized σ_0-value was also adopted in the recent IAEA-Handbook on Nuclear Activation Data /9/, quoting in the same compilation, however, the obsolete θ-value. Thus, when selecting this σ_0-θ set, an error of ≃ 12% will be committed.

Table 1. The case Fe-58(n,γ)Fe-59

COMPILATION	σ_0,barn	θ,%	σ_0 x θ
BNL-325(1973)/2/	1.15	0.31	0.356_5
NUKLIDK.(1974/3/	1.15	0.31	0.456_5
CH.NUCL.(1977)/4/	1.16	0.3	0.348
NUKLIDK.(1981)/5/	1.15	0.3	0.345
MUGHABG.(1981)/6/	1.28	0.28	0.358
CH.NUCL.(1984)/7/	1.28	0.28	0.358
NNDC COMP.(1985)/8/	1.28	0.28	0.358
IAEA273(1987)/9/	1.28	0.31	0.397

It is appreciated that evaluators are often confronted with a difficult task, since not all experimentalists specified the input data when reporting their σ_0-results, thus making a (re)normalization difficult. This is for instance the case for Sn-124(n,γ)Sn-125m (Table 2). Obviously, σ_0-evaluation was based on the experimental results of three authors, who did not quote, however, the input θ-value. When realizing that θ-data reported in literature range from 5.64 to 6.11% /16/, it is clear that combination of σ_0 = 0.13 b with the nowadays accepted θ = 5.79% can lead to significant errors.

For many cases, however, normalization of experimental values effectively could be carried out. The example of Os-184(n,γ)Os-185 shows that this was not always done (Table 3). In recent evaluations, σ_0 = 3000 b is quoted, together with θ = 0.02% as proposed by IUPAC/SAIC /16/. Clearly, this σ_0-value is the round figure of the experimental result obtained by Kim et al. /17/, who introduced, however, the then accepted θ = 0.018%, which is still (consistently) given by IAEA /9/.

Table 2. The case Sn-124(n,γ)Sn-125m

EXPERIMENTAL	σ_0,barn	θ,%	
MANGAL(1963)/13/	0.125	?	literat. data : from 5.64 to 6.11%
TILBURY(1968)/14/	0.13	?	
GLEASON(1977)/15/	0.135	?	
UNWEIGHTED MEAN	0.130±0.005		
COMPILATION	**σ_0,barn**	**θ,%**	
NUKLIDK.(1981)/5/	0.13	5.6	
MUGHABGH.(1981)/6/	0.130±0.005	5.6	
CH.NUCL.(1984)/7/	0.13	5.6	
NNDC COMP.(1985)/8/	0.130±0.005	5.79	
IAEA273(1987)/9/	0.130±0.005	5.8	

Table 3. The case Os-184(n,γ)Os-185

EXPERIMENTAL	σ_0,barn	θ,%	σ_0 x θ
KIM(1968)/17/	3005±122	0.018	54.1
COMPILATION	**σ_0,barn**	**θ,%**	**σ_0 x θ**
NUKLIDK.(1981)/5/	3000	0.02	60.0
MUGHABGH.(1984)/18/	3000±150	0.02	
CH.NUCL.(1984)/7/	3000	0.02	
NNDC COMP.(1985)/8/	3000±150	0.02	
IAEA273(1987)/9/	3005±122	0.018	54.1
"CORRECT"	2705	0.02	54.1

Thus, if the result of Kim et al. is adopted, the "correct" (renormalized) set σ_0 = 2705 b/θ = 0.02% should have been quoted, whereas the inconsistent combination σ_0 = 3000 b/θ = 0.02% is leading to an error of ≃ 11%.

Most σ_0-compilations do not give any information on the gamma-intensity data. This might lead to problematic situations, as shown in Table 4 for Ba-138(n,γ)Ba-139. In recent evaluations on decay data, the quoted gamma-intensity for the Ba-139 165.9 keV line varies from 17% /19/ over 22.0% /20/ to 23.8% /21/, the latter being the experimental result of Gehrke /22/. Thus, combination of γ=17%

Table 4. The case Ba-138(n,γ)Ba-139

EXPERIMENTAL	σ_0,barn	γ_{166},%	
KRAMER(1965)/23/	0.360±0.036	22.4	
COMPILATION	**σ_0,barn**	**γ_{166},%**	
NUKLIDK.(1981)/5/	0.35	-	recent evaluat.: from 17 to 23.8%
MUGHABGH.(1981)/6/	0.360±0.036	-	
CH.NUCL.(1984)/7/	0.4	-	
NNDC COMP.(1985)/8/	0.360±0.036	-	
IAEA273(1987)/9/	0.360±0.036	22	

or γ=23.8% with the evaluated σ_0=0.360 (± 0.036)b /6,8/ will lead to significant discrepancies of ∿ 24% or ∿6%, respectively, with the experimental result of Kramer et al. /23/ (whereon the evaluations are based), who introduced γ=22.4%. The only σ_0-compilation giving (round) figures for the gamma-intensities is IAEA /9/, and in this case the correct γ=22% is quoted. This observation cannot be generalized, however, as demonstrated below.

For Ni-64(n,γ)Ni-65, IAEA /9/ reports σ_0=1.58 (± 0.04)b, θ=0.95% and γ(1482 keV) = 23% (round figure of the current 23.5% /24/), as shown in Table 5. This σ_0-value is clearly adopted from the work of Gryntakis /25/, who introduced θ=1.16% and γ=24.6%. Thus, the IAEA-set leads to a discrepancy of ∿24%; after a double normalization, σ_0=2.06 b should have been reported. In fact, according to the present state-of-the-art, the "correct" combination (based on Gryntakis) should read as : σ_0= 2.11 b, θ=0.91% /16/ and γ=23.5%.

Table 5. The case Ni-64(n,γ)Ni-65

EXPERIMENTAL	σ_0,barn	θ,%	γ_{1482} %	σ_0xθxγ
GRYNTAKIS(1978)/25/	1.58±0.04	1.16	24.6	45.1
COMPILATION	**σ_0,barn**	**θ,%**	**γ_{1482} %**	**σ_0xθxγ**
IAEA273(1987)/9/	1.58±0.04	0.95	23	34.5
"CORRECT"	2.11	0.91	23.5	45.1

Again for Ni-64(n,γ)Ni-65, the basis of σ_0-evaluation in other compilation works is not perfectly clear, except for the 1981 KFK-Nuklidkarte /5/, as seen in Table 6. There, σ_0=1.49b and θ= 0.91% are quoted. Obviously, this σ_0 is based on the results of Gleason /26/, with no mention of θ, and of Ryves /27/, who introduced θ=1.08%, thus revealing again a problematic situation with respect to consistency.

Table 6. The case Ni-64(n,γ)Ni-65

EXPERIMENTAL	σ_0,barn	θ,%	γ_{1482},%
RYVES(1970)/27/	1.49±0.03	1.08	[β-γ]
GLEASON(1975)/26/	1.49	?	?
COMPILATION	**σ_0,barn**	**θ,%**	**γ_{1482},%**
NUKLIDK.(1981)/5/	1.49	0.91	-

As mentioned in the Introduction, straightforward conclusions can be drawn with respect to the half-life as well, if T >> t_{irr}, t_d and t_m. This is the case for Eu-153(n,γ)Eu-154[m+g]. As shown in Table 7, IAEA /9/ quotes σ_0=603(± 23)b and T=8.5 y. This σ_0-value clearly originates from the result of Kim et al. /28/, who introduced the then accepted (but now obsolete) T=16 y value, thus leading to a discrepancy in σ_0/T-ratios of ∿88%. According to the present state-of-the-art, the "correct" combination (based on Kim) should be : σ_0=323b, T=8.561y /29/.

Tabel 7. The case Eu-153(n,γ)Eu-154[m+g]

EXPERIMENTAL	σ_0,barn	T	σ_0/T
KIM(1975)/28/	603$\pm$23	16y	37.7
COMPILATION	σ_0,barn	T	σ_0/T
IAEA273(1987)/9/	603$\pm$23	8.5y	70.9
"CORRECT"	323	8.561y	37.7

In fact, Table 8 reveals that, after normalization for T and γ (whenever possible), all "activation method"-results for Eu-153(n,γ)Eu-154[m+g] are reasonably consistent, exception made for the somewhat high value of Sims et al. /30/. For comparison, the data quoted in recent compilations are shown as well.

Table 8. The case Eu-153(n,γ)Eu-154[m+g]

EXPERIMENTAL	σ_0,barn	
	PUBLISHED	NORMALIZED FOR T=8.561y {YOSHIZAWA(1985) /29/}
SIMS(1967)/30/	639	372
KIM(1975)/28/	603	323
LUCAS(1977)/31/	325	324
HEFT(1979)/32/	295	318
DECORTE(1988)/33/	307	307
COMPILATION	σ_0,barn	T
NUKLIDK.(1981)/5/	390	8.8y
MUGHABGH.(1984)/18/	312	8.5y
CH.NUCL.(1984)/7/	350	8.5y
NNDC COMP.(1985)/8/	390	8.8y
IAEA273(1987)/9/	603	8.5y

Conclusion

From the examples given in the present work, it is clear that the consistency of evaluated nuclear data reported in compilations is capable of improvement. In the extreme, it can be argued that the consistency of the nuclear data, and not their accuracy, is the matter of primary concern in NAA. This conclusion has far-reaching consequences, since ut led to the concept of the k_0-standardization in NAA, the k_0-factor being nothing else than $(\theta\gamma\sigma_0/M)/(\theta_{Au}\gamma_{Au}\sigma_{0,Au}/M_{Au})$ - as accurately determined according to the activation method with Au-197(n,γ)Au-198 as the ultimate comparator /34/.

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