REACTOR NEUTRON ACTIVATION ANALYSIS
BY A TRIPLE COMPARATOR METHOD

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The single comparator method has been extended to a triple comparator method, using ^{60}Co, ^{114m}In and ^{198}Au. In this technique, the k-ratios of the elements to be analyzed, now determined against the three comparators, are corrected for each new ratio of thermal to epithermal reactor neutron flux. These flux ratios are calculated from the absolute activities of the three comparators. The thermal neutron activation cross-section and the resonance integral for the reaction ^{113}In(n,γ)^{114m}In have been determined.

In neutron activation analysis, especially when determining a great number of elements in one sample, the treatment of the standards, *i.e.* preparation, dissolution, counting, etc., is very cumbersome and time-consuming. Furthermore, in many cases it is difficult or even impossible to irradiate standards and samples together in one capsule. For these reasons some authors[1-6] used a single comparator method by evaluating beforehand the ratios of the specific photopeak activities of the isotopes investigated to the specific photopeak activity of a comparator, both measured in well-defined experimental conditions. These ratios are defined as k-values. This method, which has been critically evaluated by Girardi et al.,[7] has the disadvantage of being strictly valid only for one set of reactor conditions, namely for one ratio of thermal to epithermal neutron flux, Φ_{th}/Φ_{epi}. Although the authors state that the applicability of the procedure can be increased by using a flux monitor with an appropriate value of I/σ_{th} (I = resonance integral, σ_{th} = thermal activation cross section), no correction procedure is given for variations of Φ_{th}/Φ_{epi}.

For the determination of Φ_{th}/Φ_{epi} in each new irradiation site, different ways can be followed. The flux ratio can readily be calculated from the cadmium ratio R_{Cd} of an isotope, for instance, by irradiating gold with and without Cd-cover. However, this technique has the disadvantage that large flux depressions are caused in the neighbourhood of the cadmium foil. For this reason, and also because it

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is not allowed in some reactors to irradiate this metal, we chose a method by which Φ_{th}/Φ_{epi} can be determined by irradiation and absolute counting of three comparators. Though a dual comparator method suffices for the calculation of Φ_{th}/Φ_{epi} and next for the correction of the k-values, we used in this work a triple comparator method with ^{60}Co, ^{114m}In and ^{198}Au in order to cover a wide range of resonance energies. Indeed, as a result of this choice, switching from one reactor spectrum to another is always possible.

Restrictions of the single comparator method

When irradiating an element in a reactor, the specific photopeak counting rate, A_{sp}, of the radioisotope formed by (n, γ) reaction, is given by the relation

$$A_{sp} = \frac{\Phi_r \, \widehat{\sigma} \, \Theta \, N_A \, SE \, \alpha \, D}{M} \tag{1}$$

where Φ_r = total reactor neutron flux, contributing to the (n, γ) reaction;

$\widehat{\sigma}$ = effective activation cross-section;
Θ = isotopic abundance of the target nuclide;
N_A = Avogadro's number;
S = saturation factor = $1 - e^{-\lambda t}$; t = irradiation time; λ = decay constant;
E = efficiency of the detector for the γ-ray measured;
α = γ-ray abundance in the decay scheme of the radioisotope;
D = decay factor = $e^{-\lambda T}$; T = decay time;
M = atomic weight of the irradiated element.

In view of the fact that both thermal and epithermal neutrons contribute to the (n, γ) reaction, Φ_r and $\widehat{\sigma}$ can be written as

$$\Phi_r = \Phi_{th} + \Phi_{epi} \tag{2}$$

where Φ_{th} = thermal neutron flux;
Φ_{epi} = epithermal neutron flux
and

$$\widehat{\sigma} = \sigma_{th} \frac{\Phi_{th}}{\Phi_r} + I \frac{\Phi_{epi}}{\Phi_r} \tag{3}$$

where σ_{th} = thermal activation cross section;
I = resonance integral.
Equation (1) becomes

$$A_{sp} = \frac{\Theta N_A \, SE \, \alpha \, D}{M} \, (\sigma_{th} \Phi_{th} + I \Phi_{epi}) \tag{4}$$

The importance of Φ_{th}/Φ_{epi}, and of I/σ_{th}, when using a comparator method, can be calculated as follows. Assuming constant irradiation and counting conditions for an element and the comparator, Eq. (4) becomes:

$$A_{sp1} = C\,(\sigma_{th}\,\Phi_{th_1} + I\,\Phi_{epi_1}) \tag{5}$$

$$A_{sp1}^{*} = C^{*}\,(\sigma_{th}^{*}\,\Phi_{th_1} + I^{*}\,\Phi_{epi_1}) \tag{6}$$

where the asterisk (*) refers to the comparator and index 1 to an irradiation in a well-fixed reactor neutron spectrum.

One can then write from Equations (5) and (6):

$$k_1 = \frac{A_{sp1}}{A_{sp1}^{*}} = \frac{C}{C^{*}} \frac{\sigma_{th}\,\dfrac{\Phi_{th1}}{\Phi_{epi1}} + I}{\sigma_{th}^{*}\,\dfrac{\Phi_{th1}}{\Phi_{epi1}} + 1^{*}} \tag{7}$$

A similar relation holds for an irradiation of the same elements in a second reactor neutron spectrum (index 2):

$$k_2 = \frac{A_{sp2}}{A_{sp2}^{*}} = \frac{C}{C^{*}} \frac{\sigma_{th}\,\dfrac{\Phi_{th2}}{\Phi_{epi2}} + I}{\sigma_{th}^{*}\,\dfrac{\Phi_{th2}}{\Phi_{epi2}} + I^{*}} \tag{8}$$

where C and C^{*} remain unchanged.

The condition for the validity of the single comparator method is that k_1 equals k_2 and:

$$\frac{\dfrac{\Phi_{th1}}{\Phi_{epi1}} + \dfrac{I}{\sigma_{th}}}{\dfrac{\Phi_{th1}}{\Phi_{epi1}} + \dfrac{I^{*}}{\sigma_{th}^{*}}} = \frac{\dfrac{\Phi_{th2}}{\Phi_{epi2}} + \dfrac{I}{\sigma_{th}}}{\dfrac{\Phi_{th2}}{\Phi_{epi2}} + \dfrac{I^{*}}{\sigma_{th}^{*}}} \tag{9}$$

Equation (9) will be fulfilled in two separate cases, namely:

1. If the ratio of thermal to epithermal flux in the two irradiation places is not equal, $\dfrac{\Phi_{th1}}{\Phi_{epi1}} \neq \dfrac{\Phi_{th2}}{\Phi_{epi2}}$, it should be necessary that $\dfrac{I}{\sigma_{th}} = \dfrac{I^{*}}{\sigma_{th}^{*}}$.

2. If the ratio of the resonance integral to the thermal activation cross-section for the two elements is not equal, $\dfrac{I}{\sigma_{th}} \neq \dfrac{I^{*}}{\sigma_{th}^{*}}$, it should be necessary that $\dfrac{\Phi_{th1}}{\Phi_{epi2}} = = \dfrac{\Phi_{th2}}{\Phi_{epi2}}$.

As in practical work, especially when comparing a great number of elements using one comparator, the first case will never be obtainable, the single comparator method will be restricted by the second condition, *i.e.* by the equality of the flux ratios.

Principle of the triple comparator method

As already mentioned, the specific photopeak activity of an isotope, formed by (n, γ) reaction, is given by the relation:

$$A_{sp} = C \left(\sigma_{th} \Phi_{th} + I \Phi_{epi} \right) \tag{5}$$

When irradiating and counting three comparators, for which all parameters of the C-factor are known, a system of three equations with two unknowns (Φ_{th} and Φ_{epi}) arises, provided that the σ_{th} and I values are given. By considering three times a combination of two equations, three sets of Φ_{th} and Φ_{epi}, and next of Φ_{th}/Φ_{epi}, can be calculated. In further calculations, the average value of the three flux ratios thus found may be used. After determining the k_s values of the elements under investigation against the three comparators for one standard value of $(\Phi_{th})_s/(\Phi_{epi})_s$ in a reference reactor position, new values of k_x for irradiation at any other flux ratio $(\Phi_{th})_x/(\Phi_{epi})_x$ can be computed.

In the standard irradiation place (index s), one may write for the k_s value of an element *versus* a comparator:

$$k_s = \frac{\Theta\, N_A\, S_s\, E\, \alpha\, D_s\, M^*}{\Theta^*\, N_A\, S_s^*\, E^*\, \alpha^*\, D_s^*\, M} \cdot \frac{\sigma_{th}\, \dfrac{(\Phi_{th})_s}{(\Phi_{epi})_s} + I}{\sigma_{th}^*\, \dfrac{(\Phi_{th})_s}{(\Phi_{epi})_s} + I^*} \tag{10}$$

For the k_x value at any other irradiation place (index x), the same relation holds:

$$k_x = \frac{\Theta\, N_A\, S_x\, E\, \alpha\, D_x\, M^*}{\Theta^*\, N_A\, S_x^*\, E^*\, \alpha^*\, D_x^*\, M} \cdot \frac{\sigma_{th}\, \dfrac{(\Phi_{th})_x}{(\Phi_{epi})_x} + I}{\sigma_{th}^*\, \dfrac{(\Phi_{th})_x}{(\Phi_{epi})_x} + I^*} \tag{11}$$

Thus

$$k_x = k_s\, \frac{S_x\, D_x\, S_s^*\, D_s^*}{S_s\, D_s\, S_x^*\, D_x^*} \cdot \frac{\left[\dfrac{(\Phi_{th})_x}{(\Phi_{epi})_x} + \dfrac{I}{\sigma_{th}} \right] \left[\dfrac{(\Phi_{th})_s}{(\Phi_{epi})_s} + \dfrac{I^*}{\sigma_{th}^*} \right]}{\left[\dfrac{(\Phi_{th})_x}{(\Phi_{epi})_x} + \dfrac{I^*}{\sigma_{th}^*} \right] \left[\dfrac{(\Phi_{th})_s}{(\Phi_{epi})_s} + \dfrac{I}{\sigma_{th}} \right]} \tag{12}$$

For reason of accuracy, the three comparators should fulfil the following requirements:

1. Sufficient spreading of the I/σ_{th} values is desirable, so that the mathematical operations do not produce senseless results.

2. An accurate determination of the E α-values [= the counting efficiency for the γ-ray(s) measured] is, of course, also required. Liquid scintillation, 4π-counting, or the method described by HEATH[8] may be applied. The measurement parameters may be tied up arbitrarily, but should be kept rigorously constant.

3. The half-lives of the isotopes, used as the comparator, should be large enough to avoid complicated corrections for the k-values after irradiation. This has the additional advantage that the saturation and decay factors can be calculated as a linear function of time, when relatively short irradiation times are used and when no large time intervals after irradiation are considered.

4. The thermal activation cross-section and the resonance integral of the comparator elements should be known with satisfactory precision. As these have not been known for one of the comparators used in this work (^{114m}In), σ_{th} and I of this isotope were determined separately. The σ_{th} and I values of the other comparators, ^{60}Co and ^{198}Au, could be taken from the literature.

5. As already mentioned, the comparators must be chosen such that they cover a wide field of resonance energies.

6. From the chemical point of view, it is desirable that the elements or compounds used as comparators should allow convenient handling before as well as after irradiation.

With all these requirements in mind, we selected cobalt, indium and gold as the comparators, which fulfil most of the above-mentioned conditions. The ^{239}U isotope, which would be very convenient with respect to its high I/σ_{th} value (= 103), was not used because of interfering fission products activities (especially when irradiating in a less moderated reactor neutron spectrum).

Experimental

Irradiation

All irradiations were performed for a period of exactly 3 h in the Thetis reactor of the Institute for Nuclear Sciences at Ghent. The reactor was operating at a constant power of 15 kW. In this work, use was made of two irradiation positions (mentioned hereafter as position 1 and 2). The thermal and epithermal neutron fluxes of these sites were determined by the usual Co and Au flux monitors, irradiated with and without Cd-cover. The results of these experiments are summarized in Table 1.

Counting techniques

After irradiation, the elements or compounds were dissolved and measured in 20-ml glass tubes on a 25-ml well-type NaI(Tl) scintillation counter coupled

4   *J. Radioanal. Chem. 3 (1969)*

Table 1

Thermal and epithermal neutron fluxes in two irradiation positions of the Thetis Reactor

Irra-diation site	Flux monitor	Φ_{th} n · cm^{-2} · sec^{-1}	Φ_{epi} n · cm^{-2} · sec^{-1}	Φ_{th}/Φ_{epi}
1	Co	$7.46 \cdot 10^{10}$	$3.16 \cdot 10^{9}$	23.6
	Au	$7.37 \cdot 10^{10}$	$3.10 \cdot 10^{9}$	23.8
	Average value	$7.42 \cdot 10^{10}$	$3.13 \cdot 10^{9}$	23.7
2	Co	$1.39 \cdot 10^{10}$	$1.20 \cdot 10^{8}$	116
	Au	$1.36 \cdot 10^{10}$	$1.12 \cdot 10^{8}$	121
	Average value	$1.38 \cdot 10^{10}$	$1.16 \cdot 10^{8}$	119

to a 400-channel spectrometer. The purity of the isotopes was in some cases checked by means of a Ge(Li) detector, coupled to a 4096-channel spectrometer.

For the determination of the detector efficiency, use was made of the liquid scintillation method.

Nuclear data

The values of σ_{th}[9], I[10] and I/σ_{th} for the comparators and other isotopes used in this work, are given in Table 2. For the resonance integrals, the average values of the most recent determinations were used. It should also be noted that I is defined according to the HØGDAHL convention.[11]

Table 2

σ_{th}; I and I/σ_{th} values

Isotope formed by (n, γ)	σ_{th}, barn	I, barn	I/σ_{th}
Comparators			
^{60}Co	37	75	2.03
^{198}Au	98.8	1550	15.7
^{114m}In	9.45*	258*	27.3*
^{46}Sc	23	10.2	0.44
^{64}Cu	4.5	4.2	0.93
^{24}Na	0.53	0.517	0.98
^{72}Ga	5.2	21.6	4.15
^{76}As	4.5	40.3	8.96
^{122}Sb	6.06	143	23.6

* Values determined in this work.

Counting efficiency

The counting efficiency of the detector [$E\alpha$ factors from Eq. (4)] can readily be determined for the comparator isotopes used in this work by liquid scintillation. Indeed, the three isotopes ^{60}Co, ^{198}Au and ^{114m}In emit a nearly single β^--radiation.[12]

The energy of the β^-, used for the liquid scintillation counting, the γ-energies of the photopeaks selected for the measurement of A_{sp}, and the experimentally determined $E\alpha$ values, corrected for the abundance of the β^- ray in the decay scheme, are listed in Table 3.

The low counting efficiency for the 192 keV γ-ray of ^{114m}In is due to internal conversion (e/γ internal conversion factor = 5).[12]

Table 3

Counting efficiency of the comparator isotopes

Compara-tor isotope	β^--energy (keV)	γ-energy (keV)	$E\alpha$
^{60}Co	312	1173—1332	0.175
^{198}Au	960	412	0.300
^{114m}In	1986	192	0.0960

Determination of σ_{th} and I for the reaction ^{113}In (n, γ) ^{114m}In

For the activation cross section of the 49 d isomer of ^{114}In, the literature lists different values in poor agreement with each other. Whereas in the compilation of HUGHES et al.[13] a value of 56 b is given, more recent determinations yielded values of 8.1 b[14] and 7.5 b[15]. As the 49 d isomer, ^{114m}In, is formed not only by direct activation of ^{113}In, but also by internal transition of the 42 msec ^{114m}In (Fig. 1), it is obvious that the given cross sections refer to the sum of the cross sections for the two isomers, ^{114m}In and ^{114m}In. For the resonance integral, which will be also a sum value, only one result is reported as being 1050 b[16].

As the accurate knowledge of σ_{th} and I is necessary for the use of In as a comparator, both values were determined.

Samples containing about one milligram of spectroscopically pure In_2O_3, mixed with one gram of spec. pure Al_2O_3 were irradiated with and without a 0.75 mm cadmium foil in the above mentioned irradiation positions 1 and 2 of the Thetis reactor. After dissolution of the In_2O_3 in HCl and filtering off the Al_2O_3, the measurements were started after a one week cooling period to allow the decay of the 54 min ^{116m}In and of the residual 15 h ^{24}Na-activity, the latter being formed by an (n, α) reaction on ^{27}Al. Using the neutron fluxes and the counting efficiency for ^{114m}In, as determined before, σ_{th} and I could be computed. The results are given in Table 4.
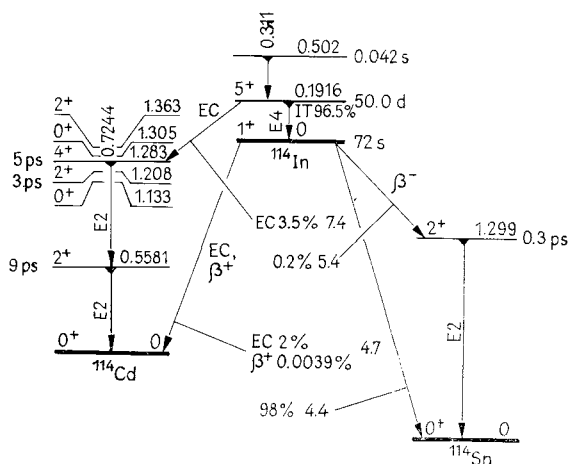
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Fig. 1. Decay scheme of ^{114}In (See ref.[12])

Table 4

σ_{th}, I and I/σ_{th} for the reaction ^{113}In(n, γ) ^{114m}In

Irradiation position	σ_{th}, barn	I, barn	I/σ_{th}
	9.19	251	27.3
1	9.03	245	27.1
	9.71	293	30.2
	10.01	251	25.1
2	9.16	262	28.6
	9.60	246	25.6
Average value	9.45	258	27.3
Standard deviation	0.38 (4.0%)	18 (7.0%)	1.9 (7.0%)

Results

Determination of $\dfrac{\Phi_{th}}{\Phi_{epi}}$ by the triple comparator method

The gold comparator was irradiated after evaporating a solution, containing 10.52 μg of gold in a small plastic tube. Indium was irradiated as a mixture of In$_2$O$_3$ and Al$_2$O$_3$, as described before. The cobalt comparator was irradiated as a Co–Al wire, containing 2% cobalt. Care was taken to fill the capsules each

time in the same way with the comparators close together, so that errors due to flux gradients were avoided. The isotopes were counted after cooling the short lived radioisotopes (^{116m}In, ^{24}Na).

After counting, the calculations of Φ_{th}, Φ_{epi} and Φ_{th}/Φ_{epi} were performed as described before. The results, obtained from the coupling of ^{60}Co $-$ ^{114m}In, ^{60}Co $-$ ^{198}Au and ^{114m}In $-$ ^{198}Au, are summarized in Tables 5 and 6, respectively, for the irradiation positions 1 and 2.

Table 5

Determination of Φ_{th}/Φ_{epi} for irradiation position 1

Combination	$\Phi_{th\,1}$ n · cm^{-2} · sec^{-1}	$\Phi_{epi\,1}$ n · cm^{-2} · sec^{-1}	$\Phi_{th\,1}/\Phi_{epi\,1}$
^{60}Co $-$ ^{114m}In	$7.25 \cdot 10^{10}$	$3.22 \cdot 10^9$	22.5
^{60}Co $-$ ^{198}Au	$7.24 \cdot 10^{10}$	$3.30 \cdot 10^9$	21.9
^{114m}In $-$ ^{198}Au	$7.51 \cdot 10^{10}$	$3.12 \cdot 10^9$	24.1
Average value	$7.33 \cdot 10^{10}$	$3.21 \cdot 10^9$	22.8

Table 6

Determination of Φ_{th}/Φ_{epi} for irradiation position 2

Combination	$\Phi_{th\,2}$ n · cm^{-2} · sec^{-1}	$\Phi_{epi\,2}$ n · cm^{-2} · sec^{-1}	$\Phi_{th\,2}/\Phi_{epi\,2}$
^{60}Co $-$ ^{114m}In	$1.33 \cdot 10^{10}$	$1.20 \cdot 10^8$	111
^{60}Co $-$ ^{198}Au	$1.32 \cdot 10^{10}$	$1.41 \cdot 10^8$	94
^{114m}In $-$ ^{198}Au	$1.44 \cdot 10^{10}$	$1.18 \cdot 10^8$	122
Average value	$1.36 \cdot 10^{10}$	$1.26 \cdot 10^8$	109

Reasonable agreement has been found between these values and those obtained by cobalt and gold flux monitoring with and without cadmium, as described earlier. However, it is obvious from Tables 5 and 6 that the triple comparator method, with respect to the epithermal flux, gives better results for irradiation position 1. This is due to the fact that in position 2 the contribution of the epithermal flux in Eq. (5) is very low so that the mathematical operations produce less accurate results.

Conversion of the k-values

The k_1-values, experimentally determined for a few isotopes in irradiation position 1, were converted with the aid of Eq. (12) to the k_2-values in irradiation position 2, having determined the flux ratios for both places. The calculated k_2-values

were then compared to the experimentally measured k_2-values. As can be seen from Table 7, the calculated and experimental values are in good agreement. The isotopes used for this purpose were: ^{24}Na, ^{64}Cu, ^{72}Ga, ^{76}As, ^{122}Sb. The measurements were carried out two days after irradiation.

Table 7

Conversion of the k-values

Comparator	Isotope	k_1 experimental value	k_2 calculated value	k_2 experimental value
^{60}Co	^{24}Na	5.69	5.89	5.73
	^{46}Sc	24.9	26.2	27.1
	^{64}Cu	7.06	7.31	7.55
	^{72}Ga	18.1	17.0	16.9
	^{76}As	33.6	27.9	28.5
	^{122}Sb	40.3	25.8	25.3
^{114m}In	^{24}Na	24.2	41.1	39.1
	^{46}Sc	105.5	182.6	184.9
	^{64}Cu	29.9	50.9	51.5
	^{72}Ga	76.6	118.2	115.4
	^{76}As	142.6	194.7	194.6
	^{122}Sb	171.1	179.7	172.8
^{198}Au	^{24}Na	$6.21 \cdot 10^{-3}$	$8.87 \cdot 10^{-3}$	$8.41 \cdot 10^{-3}$
	^{46}Sc	$2.71 \cdot 10^{-2}$	$3.94 \cdot 10^{-2}$	$3.95 \cdot 10^{-2}$
	^{64}Cu	$7.70 \cdot 10^{-3}$	$1.10 \cdot 10^{-2}$	$1.10 \cdot 10^{-2}$
	^{72}Ga	$1.97 \cdot 10^{-2}$	$2.55 \cdot 10^{-2}$	$2.47 \cdot 10^{-2}$
	^{76}As	$3.67 \cdot 10^{-2}$	$4.21 \cdot 10^{-2}$	$4.17 \cdot 10^{-2}$
	^{122}Sb	$4.40 \cdot 10^{-2}$	$3.88 \cdot 10^{-2}$	$3.70 \cdot 10^{-2}$

Conclusion

As can be seen from Tables 5 and 6, the triple comparator method using ^{60}Co, ^{114m}In and ^{198}Au, allows the calculation of the Φ_{th}/Φ_{epi} flux ratios, in good agreement with the values obtained by the usual Co and Au flux monitoring. Furthermore, it is obvious from Table 7 that the calculated k_2-values do not differ significantly from the experimental values. For these reasons, the triple comparator method offers the possibility of a multi-element analysis without irradiating all of the standards, or without being limited by the constancy of the reactor neutron spectrum. An application of this method will be published in due course.

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