Journal of Radioanalytical and Nuclear Chemistry, Vol. 220, No. 1 (1997) 99–102

HAV-1: A multipurpose multimonitor for reactor neutron flux characterization

O. Diaz Rizo,[1] I. Alvarez,[1] E. Herrera,[1] L. Lima,[1] J. Torres,[1] M. V. Manso,[2] M. C. Lopez,[3] M. Ixquiac,[1] D. De La Rosa[1]

[1]*Institute for Nuclear Sciences and Technology, Ave. Salvador Allende, esq. Luaces, Habana, Cuba*
[2]*Isotopes Center, Autopista Nacional, km 3½, Pedro Pí, Habana, Cuba*
[3]*Nuclear National Research Institute, Carr. México-Toluca, km 36½ Salazar, México D. F., Mexico*

(Received January 27, 1997)

A simple non-solid multimonitor HAV-1 for the systematic evaluation of reactor neutron flux parameters for k_0 neutron activation analysis is presented. Solutions of Au, Zr, Co, Zn, Sn, U and Th (deposited in filter paper) are used to study the parameters α and f. Dissolved Lu is used to neutron temperature (T_n) determination, according to the Wescott's formalism. A multipurpose multimonitor HAV-1 preparation, certification and evaluation is presented.

Introduction

Spectrum characterization in the reactor irradiation site is required to the k_0-standardization of NAA.[1] Traditionally, thin foils or wire of the most suitable monitors for determination of the parameter α, representing the non-ideal $1/E^{1+\alpha}$ epithermal neutron flux distribution (as Au, Co, Zr, Zn, etc.)[2–5] and the thermal-to-epithermal neutron flux ratio – f (Zr, Au, etc.)[3–5] is uzed. If one has to make the formalism where the Maxwellian neutron temperature (T_n) plays a role (such as Wescott's), the latter can be determined using a Lu monitor, based on the $^{176}Lu(n,\gamma)^{177}Lu$ reaction.[6,7]

The accuracy of these f, α and T_n determinations was significantly improved by the careful redetermination of the relevant nuclear constants (k_0, Q_0).[6,7] For this purpose the thermal and epithermal self-shielding effect in the irradiated monitor should be taken into account. This is specially a problem for Au, Co and Lu with their high (n,γ) cross sections and resonance integral.

As traditionally done,[2] a convenient way is to use dilute Al-based alloys in the form of thin wires or foils. In the last few years, such a "combined" monitor has been developed, for example, a Zr-based dilute alloy of Au and Lu[6] or Al-based dilute alloy of Au, Co and Lu,[8] thus enabling the measurement of all above mentioned relevant neutron flux parameters via the irradiation and γ-spectrometric counting of one single foil or wire.

Such a multipurpose multimonitor is being developed in the Institute for Nuclear Sciences and Technology (NST), in Havana. HAV-1 is a non-solid monitor deposited an filter paper, where the major of the recommended monitors[3] and Lu are included. Monitor preparation, certification and evaluation is presented in this paper.

Experimental

Monitor preparation and certification

HAV-1 multimonitor was prepared, taken into account all recommended accuracy norms for standard preparation.[9] High pure for analysis (HPA) compounds (Table 1) were dissolved in 50 ml (except Au in 100 ml) HNO_3 and aqua regia (Riedel–deHaen acids) and well homogenized. Portions of each solution were mixed in a 50 ml flask. Relevant nuclear data are shown in Table 2. The elemental concentrations were estimated for an A_{sp} error of less than 1%, using the NAA formula for $t_i = 20$ min, $t_d = 24$ h, $t_m = 30$ min and thermal neutron flux $\Phi = 1 \cdot 10^{13}$ n · cm^{-2} · s^{-1}. The final solution is a HAV-1 multi-standard. Aliquots of 0.01 ml of HAV-1 (JUSTOR micropipet) were finally deposited on a filter paper ($\varnothing = 2$ cm).

Recommended α-monitors as Mo and Ag were excluded, because they can form an insoluble compound with the rest of the present reagents and the necessary homogeneity can not be obtained.

The HAV-1 elemental concentration was determined by a NAA relative method of:

a) IAEA-SL-1 reference material[10] ($t_i = 25$ h, $t_d = 7$ d, $t_m = 30$ min) in the fixed irradiation system (SIFCA) of the Triga Mark III reactor of the National Nuclear Research Institute, Salazar, Mexico.[11]

b) Highly Pure for Analysis foils (Goodfellow Metals) in the three characteristic irradiation channels of the Triga Mark III reactor, i.e., in the "central channel (CC)" ($t_i = 5$ min, $t_d = 24$ h, $t_m = 30$ min), in the "pneumatic transfer tube channel (PT)" ($t_i = 5$ min, $t_d = 24$ h, $t_m =$ = 30 min), and in the SIFCA system ($t_i = 20$ min, $t_d = 24$ h, $t_m = 30$ min).

For a good statistic, the irradiation was repeated 5 times. Gamma-activities were measured in a HPGe (ORTEC, 1.8 keV for 1332 keV lin of ^{60}Co) detector. The spectra were processed using the Spectrum Analyzer system.[12] Figure 1 shows the evaluation of the SL-1 standard in the quantitative analysis of some elements in the reference material IAEA SOIL-7,[13] for the same HAV-1 irradiation conditions.

Elemental concentrations of the different monitors presented in HAV-1 and the corresponding precertification are reported in Table 3.

0236–5731/97/USD 17.00

Elsevier Science B.V., Amsterdam
Akadémiai Kiadó, Budapest

Table 1. Estimated concentration values of monitors present in HAV-1

Element	Reagent	Make	Solvent	Concentration
Zr	$ZrOCl_2 \cdot 8H_2O$	Reachim	HNO_3	9.9999%
Lu	Lu_2O_3	Fluka	HNO_3	100.83 μg/g
Th	$Th(NO_3)_4 \cdot 6H_2O$	JMC	HNO_3	1000.67 μg/g
Zn	ZnO	JMC	HNO_3/HCl	9.998%
Co	$CoCl_2 \cdot 6H_2O$	JMC	HNO_3	1001 μg/g
U	U_3O_8	JMC	HNO_3	499.75 μg/g
Au	Au	Goodfellow	HNO_3/HCl	10.06 μg/g
Sn	Sn	Reachim	HNO_3/HCl	9.945%

Table 2. Nuclear data for the nuclides chosen as monitors[6,7]

Monitor	Nuclide	E_r, eV	Q_0	F_{Cd}	Gamma-line, keV	s_0	$g(T_n)$	k_0	$T_{1/2}$
^{197}Au	^{198}Au	5.65	15.7	0.991	411.8	17.24	1.007*	1.00	2.695 d
^{238}U	^{239}Np	16.9	103.4	1	208	116.2	1	$2.77 \cdot 10^{-3}$	2.355 d
^{232}Th	^{233}Pa	54.4	11.53	1	311.9	12.53	1	$2.5 \cdot 10^{-2}$	27.0 d
^{59}Co	^{60}Co	136	1.990	1	1173.0	1.765	1	1.32	5.271 y
^{112}Sn	^{113m}Sn	107.3	48.4	1	391.7	–	1	$5.99 \cdot 10^{-5}$	115.1 d
^{96}Zr	^{97}Zr	338	248.0	1	743.6	279	1	$1.30 \cdot 10^{-5}$	16.74 h
^{64}Zn	^{65}Zn	2560	1.908	1	1115.5	1.669	1	$5.63 \cdot 10^{-3}$	244.0 d
^{94}Zr	^{95}Zr	6260	5.05	1	(724.2 + 756.7)	5.21	1	$2.09 \cdot 10^{-4}$	64.03 d
^{176}Lu	^{177}Lu	0.158	–	–	112.9	1.67	1.746*	$7.14 \cdot 10^{-2}$**	6.71 d

*T_n = 20 °C, g_{Au}(100 °C) = 1.011, g_{Lu}(100 °C) = 2.344.
**Theoretical calculated via Høgdahl convention.

Table 3. Concentration values of HAV-1 by NAA (in μg/g)

Element	SOIL-7	Metallic monitors	Main
Zr (%)	–	9.54 ± 0.47	9.5 ± 0.5
Lu	99.8 ± 4.5	–	99.8 ± 4.5
Th	1020 ± 100	–	1020 ± 100
Zn (%)	9.95 ± 0.69	9.73 ± 0.85	9.8 ± 1.2
Co	942 ± 103	963 ± 164	953 ± 190
U	499 ± 50	–	499 ± 50
Au	–	11.8 ± 0.7	11.8 ± 0.7
Sn (%)	–	9.87 ± 0.45	9.9 ± 0.5

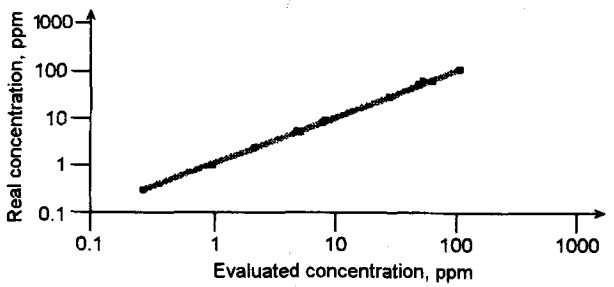

Fig. 1. IAEA SOIL-7 relative neutron activation analysis using the reference material SL-1. The straight line represent the function Real Concentration = Evaluated Concentration, ■ elements

Evaluation of HAV-1 as a multipurpose monitor

For evaluation of HAV-1 as α, f and T_n monitor, five containers with – bare and Cd-covered – HAV-1 samples and Au, Zr, Co, Zn and Sn monitor foils (see Fig. 2), were irradiated in the mentioned reactor channels (Φ_{CC} = $= 3.71 \cdot 10^{13}$ n · cm^{-2} · s^{-1}, $\Phi_{PT} = 1.37 \cdot 10^{13}$ n · cm^{-2} · s^{-1}, $\Phi_{SIFCA} = 0.92 \cdot 10^{13}$ n · cm^{-2} · s^{-1}). The irradiation times were 5, 5 and 20 minutes, respectively. In all cases decaying and measuring times were 20–24 hours and 30 minutes. The spectra were measured in an ORTEC detector.

The epithermal spectrum shape factor α was obtained as the slope (– α) of the straight line when plotting log (T_i) versus log ($E_{r,i}$/1 eV), which is similar to the solving of the implicit equation (1 eV omitted):

$$\alpha + \frac{\sum_i^N \left[\log(E_{r,i}) - \frac{\sum_i^N \log(E_{r,i})}{N}\right]\left[\log(T_i) - \frac{\sum_1^N \log(T_i)}{N}\right]}{\sum_1^N \left[\log(E_{r,i}) - \frac{\sum_1^N \log(E_{r,i})}{N}\right]^2} = 0$$

where N is the number of coirradiated α-monitors, $E_{r,i}$ – the average resonance energy of the monitor i and T_i was given by the "Cd-covered multi-monitor method"[14] as

$$T_i = \frac{(E_{r,i})^{-\alpha}(A_{sp,i})_{Cd}}{k_{0Au}(i)\,\varepsilon_{p,i}\,F_{Cd,i}\,Q_{0,i}(\alpha)\,G_{e,i}}$$

with $A_{sp} = (N_p/t_m)SDCw$, where N_p is the measured net peak area, t_m is the counting time, S is the saturation factor, D is the decay factor, C is the counting factor and w is the sample mass (g). $k_{0,Au}$ are tabulated k_0-factors, $\varepsilon_{p,i}$ is the detector efficiency for used γ-line, $F_{Cd,i}$ is the Cd-transmition factor and G_e is the correction factors for epithermal neutron self-shielding.

The thermal-to-epithermal neutron flux ratio (f) was determined by the "bare bi-isotopic monitor" method using Zr[3] as

$$f = \frac{G_{e,1}\,\frac{k_{0,Au}(1)\varepsilon_{p,1}}{k_{0,Au}(2)\varepsilon_{p,1}}\,Q_{0,1}(\alpha) - G_{e,2}\,\frac{A_{sp,1}}{A_{sp,2}}\,Q_{0,2}(\alpha)}{G_{th,2}\,\frac{A_{sp,1}}{A_{sp,2}} - G_{th,1}\,\frac{k_{0,Au}(1)\,\varepsilon_{p,1}}{k_{0,Au}(2)\,\varepsilon_{p,1}}}$$

were $1 = {}^{97}Zr/{}^{97m}Nb$ (743 keV), $2 = {}^{95}Zr$ (724.2 + + 756.7 keV), G_{th} is the correction factors for thermal neutron self-shielding and $\varepsilon_{p,1} = \varepsilon_{p,2}$. Due to the single-decayed γ-lines, it is allowed to position the Zr monitor as close as possible to the detector cap.

The neutron temperature (T_n) was determined experimentally, using the "absolute" method introduced by DE CORTE et al.[6] for the Wescott's factor $g(T_n)$ measurement, based on the Lu co-irradiating with a "1/v" ($g(T_n) = 1$) monitor,

$$g_{Lu}(T_n) = \left[\frac{\frac{A_{sp,Lu}/G_{th,Lu}}{k_{0,Au}(Lu)\,\varepsilon_{p,Lu}}}{\frac{A_{sp,1/v}}{k_{0,Au}(1/v)\,\varepsilon_{p,1/v}}}\right][G_{th,1/v}\cdot g_{1/v}(T_n) + G_{e,1/v}\cdot$$

$$r(\alpha)\sqrt{T_n/T_0}\cdot s_{0,1/v}(\alpha)] - G_{e,Lu}\cdot r(\alpha)\sqrt{T_n/T_0}\cdot s_{0,Lu}(\alpha)/G_{thLu}$$

where $r(\alpha)\sqrt{T_n/T_0}$ is a modified spectral index, which is obtained from experiments and $s_0(\alpha)$ is a measure for the epithermal to thermal (n,γ) cross section. The above equation yields $g_{Lu}(T_n)$, and, from tables of $g_{Lu}(T_n)$ versus T_n. Finally T_n is obtained.

Average α and f values obtained for each irradiation position are shown in Table 4. The uncertainties were calculated according to the error propagation study given in Reference 14.

The difference between the results obtained with HAV-1 and foil monitors is in the uncertainties. The large HAV-1 uncertainties in respect to the foils is comprehensible. HAV-1 Co, Zn and Zr concentrations are some orders of magnitude lower than the HP foil concentrations. In this case, according to the used irradiation regimes, a large gamma activity experimental error (for mentioned elements) is introduced in HAV-1 spectra, in respect to the foil spectra. This effect is bigger for the Cd-covered irradiated samples.

On the other hand, the relatively high (10–30%) relative uncertainties are acceptable for lower absolute α, is and are satisfactory for NAA needs.[3] The good coincidence obtained for α and f values, is an evidence of the HAV-1 quality as α and f multimonitor for Triga type nuclear reactors.

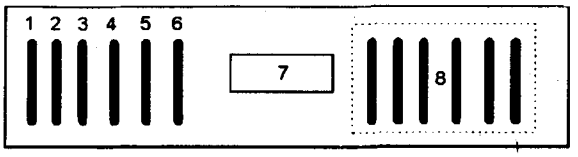

Fig. 2. HAV-1 irradiation container: 1 – HAV-1, 2 – Au, 3 – Zr, 4 – Co, 5 – Zn, 6 – Sn, 7 – filter paper, 8 – Cd-covered monitors

Table 4. α and f-values determined in the irradiation positions of the Triga Mark III reactor

Channel	α (HAV-1)	α (foils)	f (HAV-1)	f (foils)
CC	–0.13 ± 0.02	–0.13 ± 0.01	27 ± 4	29 ± 2
PT	–0.12 ± 0.02	–0.14 ± 0.01	43 ± 4	40 ± 2
SIFCA	–0.07 ± 0.02	–0.08 ± 0.02	64 ± 4	64 ± 3

Table 5. Neutron temperature (°C) in the irradiation channels of Triga Mark III reactor as obtained from "absolute" T_n-monitoring with Lu

1/v-monitor	CC	PT	SIFCA
Au	79.0 ± 4.3	57.2 ± 4.3	32.7 ± 2.5
U	85.6 ± 5.1	61.3 ± 4.3	30.1 ± 3.3
Co	84.4 ± 5.0	61.0 ± 4.4	35.4 ± 3.1
Zn	85.2 ± 4.7	62.7 ± 4.9	41.4 ± 5.1
Average	83.7 ± 9.5	60.6 ± 8.9	34.9 ± 7.3

Results of the T_n-determination in the three irradiation channels of the Triga Mark III reactor, using, Au, U, Co and Zn from HAV-1 as 1/v-monitors, are shown in Table 5. T_n dependence of $g_{Lu}(T_n)$ Wescott's coefficient was calculated as $g_{Lu}(T_n) = aT_n + b$, where $a = 0.00745$ and $b = 1.5965$ were calculated for $g_{Lu}(20\ °C) = 1.746$ and $g_{Lu}(100\ °C) = 2.344$ as recently published.[7] For the SIFCA system, which is positioned far away from the reactor core (5 cm), the T_n-value found is practically equal to the expected cooling water temperature in the reactor neighborhood.

In Table 5 only random uncertainties are shown. The systematic uncertainties, mainly arising from those on nuclear data of the Lu-monitor and experimental α-determination error with HAV-1 monitor, amount to 30% (CC), 40% (PT) and 50% (SIFCA). This is not unacceptably high, since the $g(T_n)$ versus T_n curves for other reactions which have to be corrected for a "non-1/v" behavior (for example, $^{151}Eu(n,\gamma)^{152m}Eu$)[7] shows a much flatter slope.

Conclusions

Taking into account the simple and non expensive preparation, good coincidence for α and f evaluation as compared to metal monitors, acceptable values obtained for T_n and the possibility to measure these parameters in a short time period (one day vs. few days with conventional monitors), the use of HAV-1, or similar multipurpose multimonitor is recommendable for regular evaluation of the reactor conditions for k_0 NAA.

*

The authors are grateful to the Consejo Nacional de Ciencia y Técnica (CONACyT) and National Nuclear Research Institute from Mexico for the partial financial support of this research. Gratitude is also to the NNRI reactor group for its help during the experiments.

References

1. F. DE CORTE, A. SIMONITS, A. DE WISPELAERE, J. HOSTE, J. Radioanal. Nucl. Chem., 113 (1987) 145.
2. Neutron Flux Measurements, IAEA, Techn. Repts. Ser. No. 107, IAEA, Vienna, 1970.
3. F. DE CORTE, Ph. D. Thesis, Univ. Gent, 1987.
4. A. SIMONITS, F. DE CORTE, J. HOSTE, J. Radioanal. Chem., 31 (1976) 467.
5. F. DE CORTE, A. SIMONITS, Vade Mecum for k_0-users, DSM Research, Gelen (NL) R29/11492, December, 1994.
6. F. DE CORTE, A. SIMONITS, F. BELLEMANS, M. C. FREITAS, S. JOVANOVIC, B. SMODIS, G. ERDTMANN, H. PETRI, A. DE WISPELAERE, J. Radioanal. Nucl. Chem., 169 (1993) 125.
7. F. DE CORTE, F. BELLEMANS, P. DE NEVE, A. SIMONITS, J. Radioanal. Nucl. Chem., 179 (1994) 93.
8. F. DE CORTE, K. MASUMOTO, A. DE WISPELAERE, F. BELLEMANS, J. Radioanal. Nucl. Chem., 192 (1995) 91.
9. G. GREENBERG, J. Radioanal. Nucl. Chem., 179 (1994) 131.
10. R. DYBCZYNSKI, O. SUSCHNY, Report IAEA/RL/64, Vienna, 1979.
11. L. C. PAREDES, Multipurpose Research Reactors, IAEA-STI/PUB/762, Vienna, 1988, p. 27.
12. E. HERRERA, A. PERDOMO, C. GARCIA, Spectrum Analyzer, INST, Havana, Cuba, 1995.
13. L. PSZONICKI et al., Report IAEA/RL/112, Vienna, 1984.
14. F. DE CORTE, K. SORDO-EL HAMMAMI, L. MOENS, A. SIMONITS, A. DE WISPELAERE, J. HOSTE, J. Radioanal. Chem., 62 (1981) 209.