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A simple non-solid multimonitor HAV-1 for the systematic evaluation of reactor neutron flux parameters for k_0 neutron activation analysis is presented. Solution 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, the use of thin foils or wire of the must 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 expanded. 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 determinate using a Lu monitor, based on the reaction $^{176}Lu(n,\gamma)^{177}Lu$ [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 propose the thermal and epithermal self shielding effect in the irradiated monitor must should be taken into account. It 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 solution is to use dilute Al-based alloys in the form of thin wires or foils. In the last few years, such a "combined" monitor is begin 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.

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

MONITOR PREPARATION AND CERTIFICATION.

HAV-1 multimonitor was prepared, taken into account all recommended accuracy norms for standard preparation [9], as follows:

High pure for analysis (HPA) compounds (Table 1) were dissolved in 50 ml volumetric (except Au in 100 mL) with HNO_3 and aqua regia (Riedel-deHaen acids) and well homogenized. Portions of each solution were mixed in a 50 mL volumetric. Relevant nuclear data are shown in Table 2. The elemental concentrations were estimated for A_{ep} error less than 1%, using the NAA formula for T_i=20 min, T_d= 24 h, T_m= 30 min and $\Phi=1.10^{13}$ $n.cm^{-2}s^{-1}$. The final solution is a HAV-1 multi-standard. Aliquot of 0.01 mL of HAV-1 (JUSTOR micropipet) was finally deposit in a filter paper (Ø = 2cm).

Table 1

Estimated concentration values of monitors present in HAV-1

Element	Reactive	T. Mark	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	$CoCl_2 \cdot 6H2O$	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 %

Recommended α-monitors as Mo and Ag were excluded, because they can form an insoluble compounds with the rest of the present reactives, and the necessary homogeneity can not to be obtain.

Table 2

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

Monitor	Nuclide	E_r, eV	Q_0	F_{Cd}	γ-line, keV	s_0	$g(T_n)$	k_0	$T_{½}$
^{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	$2,7710^{-3}$	2.355 d
^{232}Th	^{233}Pa	54.4	11.53	1	311.9	12.53	1	$2,520^{-2}$	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	$5,99.10^{-5}$	115.1 d
^{96}Zr	^{97}Zr	338	248.0	1	743.6	279	1	$1,30.10^{-5}$	16.74 h
^{64}Zn	^{65}Zn	2560	1.908	1	1115.5	1.669	1	$5,63.10^{-3}$	244.0 d
^{94}Zr	^{95}Zr	6260	5.05	1	(724.2 + 756.7)	5.21	1	$2,09.10^{-4}$	64.03 d
^{176}Lu	^{177}Lu	0.158	-	-	112.9	1.67	1.746*	$7,14.10^{-2}$ '	6.71 d

Notes:

* T_n=20 °C, g_{Au}(100 °C)=1.011, g_{Lu}(100 °C)=2.344

' - theoretical calculated via Høgdahl convention

The HAV-1 elemental concentration was determinate by relative method of NAA using:

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], and,

b.-) High 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).

So as to obtain good statistic, the irradiation were repeated 5 times. Gamma activities were measured in a HPGe (ORTEC, 1.8 keV for 1332 keV line 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.

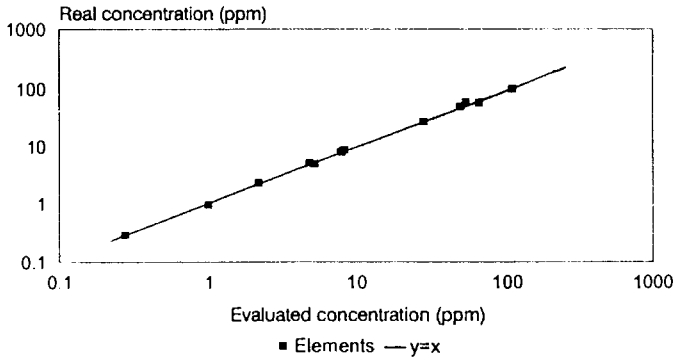

Figure 1.- IAEA-SOIL-7 relative neutron activation analysis using the reference material SL-1

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

Table 3

Concentration values of HAV-1 by NAA (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

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 Figure 2), were irradiated in the mentioned reactor channels (Φ_{CC}= 3.71x10^{13} n.cm^{-2}s^{-1}, Φ_{PT}= 1.37x10^{13} n.cm^{-2}s

[1], Φ_{SIFCA}= 0.92x10^{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 the ORTEC mentioned detector.

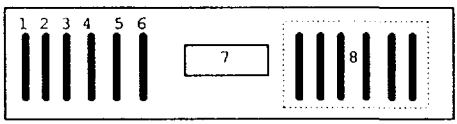

Figure 2.- HAV-1 irradiation container.

(1- HAV-1, 2-Au, 3- Zr, 4-Co, 5-Zn, 6-Sn, 7- filter paper, 8- Cd-covered monitors)

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

$$\alpha + \frac{\sum_{1}^{N}\left\{\left[\log(E_{r,i}) - \frac{\sum_{1}^{N}\log(E_{r,i})}{N}\right]\left[\log(T_i) - \frac{\sum_{1}^{N}\log(T_i)}{N}\right]\right\}}{\sum_{1}^{N}\left[\log(E_{r,i}) - \frac{\sum_{1}^{N}\log(E_{r,i})}{N}\right]^2} = 0$$

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

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

with A_{sp} = (N_p/t_m)/SDCw, where N_p - measured net peak area, t_m - counting time, S - saturation factor, D - decay factor, C - counting factor and w - sample mass (in grams). $k_{0,Au}$ - tabulated k_0-factors and G_e is the correction factors for epithermal neutron self-shielding.

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

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

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

Neutron temperature (T_n) was experimentally determinated, using the "absolute" method introduced by De Corte et al. [6] for the Wescott's factor $g(T_n)$ measurement, based in the Lu coirradiating with a "1/v" ($g(T_n)=1$) monitor,

$$g_{Lu}(T_n) = \left[\frac{A_{sp,Lu}\ G_{th,Lu}}{k_{0,Au}(Lu)\varepsilon_{p,Lu}} \Big/ \frac{A_{sp,1/v}}{k_{0,Au}(1/v)\varepsilon_{p,1/v}}\right]\left[G_{th,1/v}\cdot g_{1/v}(T_n) + G_{r,1/v}\cdot r(\alpha)\sqrt{T_n/T_0}\cdot s_{0,1/v}(\alpha)\right] -$$

$$G_{r,Lu}\cdot r(\alpha)\sqrt{T_n/T_0}\cdot s_{0,Lu}(\alpha)/G_{th,Lu}$$

where $r(\alpha)\sqrt{T_n/T_0}$ is a modified spectral index, which is as 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 [14].

Table 4

α and f-values determinated 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

The difference between the results obtained with HAV-1 and foils monitors is only in the uncertainties. The large HAV-1 uncertainties respect to the foil's is comprehensible. HAV-1 Co, Zn and Zr concentrations are some order less than the HP foil concentrations. In this case, according to the used irradiation regimes, it is introduced a large gamma activity experimental error (for mentioned elements) in HAV-1 spectra, 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's α, and are satisfactory for NAA needs [3]. The last, and the good coincidence obtained for α and f values, are an evidence of the HAV-1 quality as α and f multimonitor for Triga type nuclear reactors.

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 $g_{Lu}(Tn)$ Wescott's coefficient was calculated as $g_{Lu}(T_n) = aT_n + b$, where a = 0.00745 and b = 1.5965 were calculated for $g_{Lu}(20\ ^{\circ}C) = 1.746$ and $g_{Lu}(100^{\circ}C) = 2.344$ 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 expecting cooling water temperature in the reactor neighborhood.

Table 5

Neutron temperature ($^{\circ}C$) 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

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

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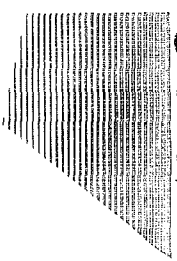

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