The set Au - Zr - Mo - Cr - Lu as multiple reactor flux monitor. The performance of ^{96}Zr and the analysis of inconsistencies

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Resumen

El uso simultáneo de Au, Zr, Mo, Cr y Lu, para la caracterización de facilidades de irradiación, con propósitos de análisis por activación neutrónica paramétrico, revela algunas inconsistencias sistemáticas en la determinación de los parámetros relevantes: flujos térmico y epitérmico, así como el exponente alfa (α), que describe el comportamiento no ideal del flujo neutrónico epitérmico. Se presenta un caso experimental que muestra que cuando los valores determinados son óptimos para la reacción ^{96}Zr(n,γ)^{97}Zr, pueden ser inaceptablemente discrepantes para las reacciones ^{98}Mo(n,γ)^{99}Mo y ^{50}Cr(n,γ)^{51}Cr y viceversa. En las condiciones empleadas en este trabajo, se ha encontrado que el par de reacciones ^{96}Zr(n,γ)^{97}Zr y ^{98}Mo(n,γ)^{99}Mo no produce una solución única para los valores de los flujos neutrónicos térmico y epitérmico, independientemente de los valores que se asignen al parámetro α.

Abstract

The simultaneous use of Au, Zr, Mo, Cr and Lu, for the characterization of irradiation facilities with the purpose of parametric neutron activation analysis, reveals some systematic inconsistencies in the determination of the values of the relevant parameters: thermal and epithermal fluxes, as well as the exponent alpha (α), which describes the non ideal behaviour of the epithermal neutron flux. An experimental case is presented, showing that when the determined values are optimal for the reaction ^{96}Zr(n,γ)^{97}Zr, can be unacceptably discrepant for the reactions ^{98}Mo(n,γ)^{99}Mo and ^{50}Cr(n,γ)^{51}Cr, and vice versa. Under the conditions of this work, it has been found that the pair of reactions ^{96}Zr(n,γ)^{97}Zr and ^{98}Mo(n,γ)^{99}Mo does not lead to a single solution for the values of the thermal and epithermal neutron fluxes, independently of the values given for the parameter α.

1. Introduction

The use of the sets Zr - Au and Au – Cr – Mo as bare multi-isotopic neutron flux monitors, for the characterization of irradiation facilities, with purposes of parametric neutron activation analysis, is frequently mentioned in literature (see for example [1-5]). It has been recently reported [5] that the sets Zr - Au and Au – Cr – Mo lead to considerably different values for the parameters *f* (thermal to epithermal flux ratio, ϕ_t/ϕ_e) and α, an exponent which describes the departure of the epithermal neutron flux from the ideal behaviour. In order to study this reported inconsistency, a careful determination of the parameters *f* and α was performed in one of the irradiation facilities of the RP-10 nuclear research reactor at IPEN.

2. Experimental

All the literature data were extracted from the following sources: atomic weights, reference [6]; isotopic abundances, thermal cross sections (σ_0), Q_0 = (resonance integral/cross-section ratios, I/σ_0), effective resonance energies, half lives, gamma intensities and k_0 values, reference [7].

The efficiency curve of the detector was initially determined at 210.7 mm, using standard sources of ^{133}Ba, ^{109}Cd, ^{57}Co, ^{137}Cs, ^{54}Mn, ^{65}Zn and ^{60}Co, from CANBERRA. Then, the activities of ^{165}Dy, ^{203}Hg, ^{51}Cr and ^{198}Au sources, prepared at the laboratory, were determined and used as secondary standards, together with the standard sources

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of ^{109}Cd, ^{57}Co, ^{137}Cs, ^{54}Mn and ^{65}Zn, to calculate the efficiency curve at 57.7 mm (^{133}Ba was not employed, in order to avoid coincidence summing effects). The obtained efficiency curve is shown in figure 1.

Monitors of Zr, Au, and Lu, and a mixed Au-Cr - Mo monitor [8] were weighed with a calibrated analytical balance, placed inside an aluminium capsule and irradiated together at the position A9Y2 of the RP10 nuclear reactor, for 8820 seconds, at a thermal power of 320 kW. After suitable decay times, the induced activities were measured by high resolution gamma spectrometry, using an HPGe detector with a relative efficiency of 15 %. The counting distance was kept at 57.7 mm (from the detector cover). The experiment was carried out three times with very similar results. The characteristics of the monitors and the counting data for one of the replicates are respectively shown in tables 1and 2.

Table 1: Irradiated monitors.

Element	*Weight (mg)*	*General description*
Zr	56.8	Foil, 0.1 mm thick, 99.9 % Zr.
Au_1	0.0125	12.5 mg of IRMM-530R certified reference material.
Au_2	0.00242	Au-Cr-Mo mixed monitor [8].
Cr	0.524	Au-Cr-Mo mixed monitor [8].
Mo	1.074	Au-Cr-Mo mixed monitor [8].
Lu	0.0072	Al-0.1% Lu wire, 1.00 mm diameter. From IRMM.

The method for determination of f and α is a modification of that developed by Arribere and Kestelman [9] for the simultaneous determination of thermal and epithermal fluxes. The specific activity of a monitor ***i*** evaluated at infinite irradiation and zero decay times can be expressed by

$$g(T)\sigma_i \phi_t + [I(\alpha)_i + \Delta I_i]\, \phi_e = A_i \qquad (1)$$

Where $\Delta I_i = 0.56\ g(T)\ \sigma_0$ is the resonance integral between E = μkT and the cadmium cut-off energy, and g(T) is the Westcott`s factor. The equation (1) is used to calculate, for each monitor, a series of values of the thermal neutron flux as a function of a series of arbitrarily given values for the epithermal neutron flux. In the Arribere and Kestelman's method an ideal behaviour is assumed for the epithermal flux, so that straight lines are obtained for every monitor. The values of the thermal and epithermal fluxes are calculated from the average of the values obtained at all the crossing points. The modification proposed in the present work is based on the following reasoning: provided that the input data of cross sections, resonance integrals and Wescott's factors are accurate, a right value of the α parameter should lead to a single crossing point for the curves corresponding to the reactions: ^{94}Zr(n,γ)^{95}Zr, ^{96}Zr(n,γ)^{97}Zr, ^{197}Au(n,γ)^{198}Au, ^{50}Cr(n,γ)^{51}Cr and ^{98}Mo(n,γ)^{99}Mo, thus implying that all the monitors should render the same values for the thermal and the epithermal neutron fluxes. If, in addition, the value of the Westcott's factor for ^{176}Lu is correct, the curve for the reaction ^{176}Lu(n,γ)^{177}Lu should also cross the other curves at the same point. Thus, α and T are adjusting parameters of the crossing point between all curves, fact that provides the basis for their calculation. In practice, it is expected that all the curves will not cross the others at exactly the same point, due to the presence of experimental uncertainties, as well as some degree of inconsistency in the input data.

Table 2: Counting data.

Isotope	*E (keV)*	*Net Peak Area*	*Normalised Specific Counting rate ($t_{irr} = \infty$ $t_d = 0$)*
^{95}Zr	724+756	74530	4.923×10^6
^{97}Zr	743	330854	3.225×10^6
^{198}Au_1	411.8	111234	6.541×10^{10}
^{198}Au_2	411.8	534846	6.561×10^{10}
^{51}Cr	320	59578	1.471×10^8
^{99}Mo	739	14387	3.98×10^{6} *
^{177}Lu	208	105181	1.189×10^{10}

*: Corrected by a true coincidence factor COI = 0.969.

3. Results and Discussion

The efficiency curve showed in figure 1 exhibits a good agreement between the standard and the prepared gamma sources, as well as between the fitting curve and the experimental points. Therefore, it can be accepted that the efficiency characterization of the HPGe detector is good enough for the purpose of the present study.

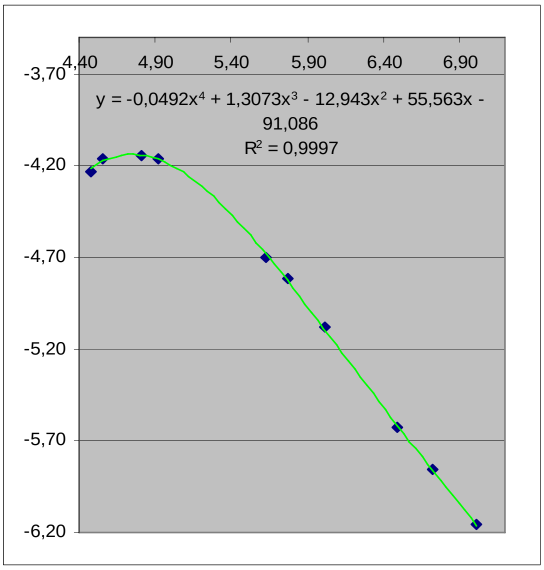

Figure 1: Efficiency curve for the used detector.

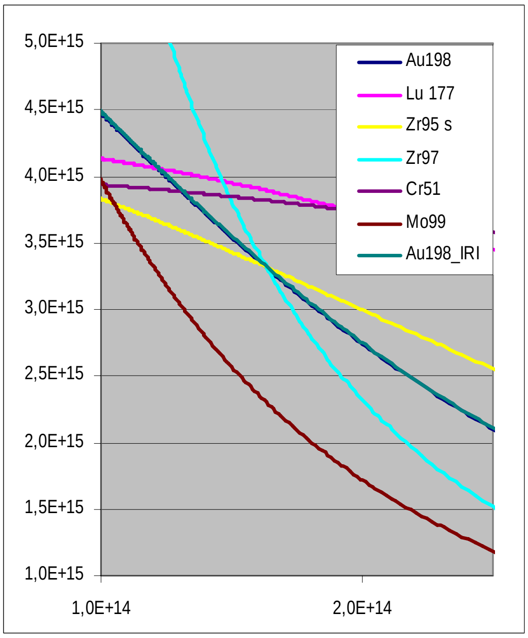

Figure 2: Thermal and epithermal neutron fluxes for α = -0.004, T = 305 K.

Figure 2 shows an example of the results obtained when the reactions ^{94}Zr(n,γ)^{95}Zr, ^{96}Zr(n,γ)^{97}Zr, ^{197}Au(n,γ)^{198}Au, often used for the simultaneous characterization of neutron fluxes and α, are taken as the leading reference for all the other monitors. The values of α and T are respectively setup as -0.004 (corresponding to the crossing point for the Au – Zr monitor) and 310 K (temperature of the moderator of the reactor). It can be seen that for the given value of α, the curves for the mentioned reactions cross together at a single point, which corresponds to $f = \phi_t / \phi_e$ = 3.34 x 10^{15} n m^{-2} s^{-1}/ 1.62 x 10^{14} n m^{-2} s^{-1} = 20.6. On the other hand, it is also apparent that this result is not consistent with the behaviour of Cr, Mo and Lu monitors.

The curve for the ^{176}Lu(n,γ)^{177}Lu reaction that would lead to an agreement of the Zr and Au monitors would be obtained at 354.9 K (82 C) neutron temperature. This temperature seems significantly high for a light water moderated nuclear reactor working at a moderator temperature of 310 K (37 C), with no strong absorbers of low energy neutrons in the vicinities of the irradiation facility. In addition, this would lead to a not acceptable solution for ^{50}Cr and ^{98}Mo monitors.

Alternatively, the value of α can be fixed as 0.112, which is the solution for the Au-Mo-Cr mixed monitor. This corresponds to f = 3.91 x 10^{15} n m^{-2} s^{-1} / 1.61 x 10^{14} n m^{-2} s^{-1} = 24.3 (Figure 3). The neutron temperature is in this case 313 K (40 C), which represents a good agreement between the ^{176}Lu monitor and the ^{50}Cr, ^{98}Mo ^{197}Au and ^{94}Zr ones, whereas the curve for the reaction ^{96}Zr(n,γ)^{97}Zr exhibits a pronounced departure.

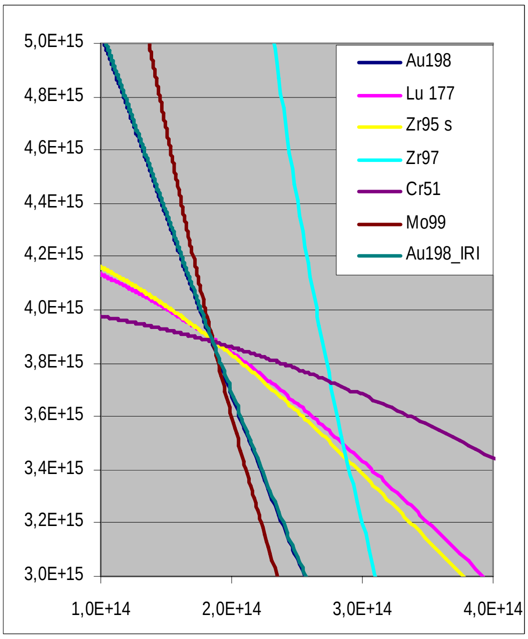

Figure 3: Thermal and epithermal neutron fluxes for α = 0.112, T = 313 K.

It is also observed that the curves for the reactions ^{96}Zr(n,γ)^{97}Zr and ^{98}Mo(n,γ)^{99}Mo do not cross each other, even for an interval as high as -0.4 < α < 0.4. This fact demonstrates a lack of consistency between these monitors. Since it is possible to find a good agreement between the ^{98}Mo monitor and the other ones, this inconsistency can not be attributed to eventual failures in the experimental conditions, which were carefully assessed. In addition, the nuclear and chemical data for the monitors ^{197}Au, ^{50}Cr, ^{98}Mo, ^{94}Zr and ^{176}Lu are reasonably consistent.

The determination of k_0 values from a set of experimental NAA data is a useful tool to check the overall quality of an experiment [11]. On this grounds, a good agreement between experimental and literature k_0 values is an indicator of the suitability of the experimental conditions and the nuclear and chemical input data.

The experimental k_0 values for the studied monitors, determined from the values of f = 24.3, α = 0.112, are shown in table 3. It is apparent that all the values obtained from the experimental data of the present work are in good agreement with the values reported in the literature.

Table 3: Experimental k_0 values from this work.

Monitor	*E (keV)*	*k_0 This work*	*k_0 Literature*
^{94}Zr	724+756	2.002×10^{-4}	2.000×10^{-4}
^{96}Zr	743	1.223×10^{-5}	1.237×10^{-5}
^{197}Au_IRI	411.8	1.003	1.000 (absolute)
^{50}Cr	320	2.60×10^{-3}	2.62×10^{-3}
^{98}Mo	739	8.32×10^{-5}	8.46×10^{-5}
^{176}Lu	208	7.19×10^{-2}	7.14×10^{-2}

The situation brings an old and quite difficult problem: the value of the ratio I/σ_0 for the ^{96}Zr. [10,12]. As it is quoted in reference [12], ^{96}Zr has the largest known Q_0 value, fact that implies high error propagation for Cd-ratios and Cd difference measurements at not too well thermalized irradiation facilities. The problem does not seem to be completely solved; the reported averages for the Q_0 value of ^{96}Zr (ref. [10]) range from 251.6 to 296.1. As described in references [10,12], the proposed values for the Q_0 value of ^{96}Zr historically covered a range going from 100 to 879.

It is interesting to note that a total consistency of the whole set of studied monitors would be reached, for a neutron temperature T = 313 K, if the value for the Q_0 value of ^{96}Zr is setup at 500, as it is indicated in figure 4 (The other two replicates, not shown here, require ^{96}Zr Q_0 values of 490 and 470).

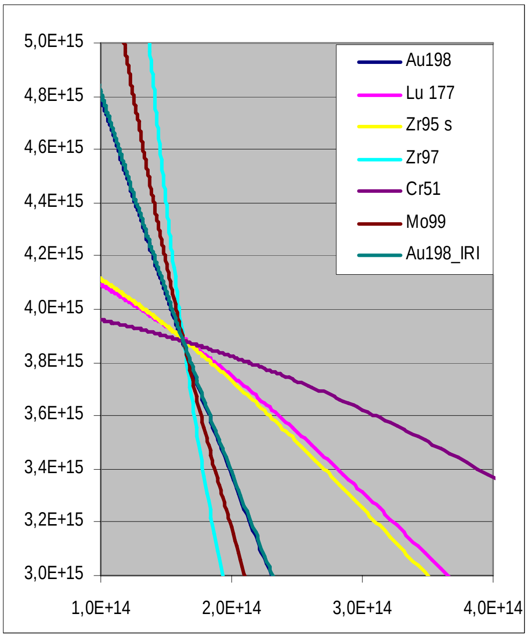

Figure 4: Thermal and epithermal neutron fluxes for α = 0.112, T = 313 K, Q_0 (^{96}Zr) = 500.

Of course, the choice of Q_0 (^{96}Zr) = 500 looks somewhat exotic at a first glance and can not be taken as a rigorous determination, but indicates that it is worthwhile to further study the problem in deep. In particular, a careful assessment of the consistency of the nuclear data for as many monitors as possible, which could be performed, applying the methodology followed in the present work, shed light on the eventual presence of any other problematic case.

4. Acknowledgment

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