Determination of ^{140}La Fission Product Interference Factor for INAA

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Abstract. Instrumental Neutron Activation Analysis (INAA) is a technique widely used to determine the concentration of several elements in several kinds of matrices. However if the sample of interest has higher relative uranium concentration the obtained results can be interfered by the uranium fission products. One of these cases that is affected by interference due to U fission is the ^{140}La, because this radioisotope used in INAA for the determination of concentration the La is also produced by the $^{-}\beta$ of ^{140}Ba, an uranium fission product. The ^{140}La interference factor was studied in this work and a factor to describe its time dependence was obtained.

Keywords: Interference factor, neutron activation analysis, fission products.
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INTRODUCTION

INAA is an analytical method used to determine concentration of several elements in different kinds of matrices, such as geological, biological and environmental. This technique is established with a primary method that provides great results in determination of concentration of the elements at low concentrations. Among the advantages of this technique can listed precision, accuracy and the possibility of multi elemental analysis.

However, in the determination of some elements which sample has higher uranium concentration this technique can show interference problems due to uranium fission products. This problem occurs when the radioisotope used in the analysis is the same formed by the uranium fission, or by the decay of some other uranium fission product. The magnitude of this interference is proportional to the relative concentration of U and the element of interest, time of decay and irradiation conditions.

This problem can be solved making pre uranium separation of sample before the irradiation procedure, or determining the uranium fission interference factor. The determination of uranium fission interference factor has the advantage that once determinate it the analyst only needs to apply the correction to obtain the real concentration of the element of interest.

Among the radioisotopes used in INAA that suffers this kind of interference the ^{140}La is a special case, because this radioisotope is formed by β^{-} decay of fission product ^{140}Ba. Some authors have studied the interference for factor for lanthanum[1-6], however the uranium fission interference factor it is dependent on characteristics of the nuclear reactor facilities. In present work it was determined uranium fission interference factor for lanthanum in the IEA-R1 nuclear research reactor experimentally and theoretically. The obtained results were applied in analysis of certified reference material.

Uranium Fission Interference Factor for Lanthanum

The uranium fission interference factor is defined by the ratio of the specific activity of radioisotope formed by the uranium fission to specific activity of this radioisotope formed by neutron capture reaction.

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$$F^{La} = \frac{A_U / m_U}{A_{La} / m_{La}} = \frac{M_{La} \cdot a_U \cdot f[\sigma_{th}^{f} + (\phi_{ep} / \phi_{th}) \cdot \sigma_{ep}^{f}]}{M_U \cdot a_{La}[\sigma_{th}^{La} + (\phi_{ep} / \phi_{th}) \cdot \sigma_{ep}^{La}]} \quad (1)$$

Where A_U and A_{La} are activity of La formed by the uranium fission and activity of ^{140}La formed by the neutron capture reaction respectively; m_U and m_{La} are the masses of U and La; a_U and a_{La} are isotopic abundance of ^{235}U and ^{139}La; M_U and M_{La} are atomic masses of U and La; σ_{th}^{f} and σ_{ep}^{f} are thermal and epithermal fission cross sections for ^{235}U; ϕ_{th} and ϕ_{th} are thermal and epithermal neutron fluxes; λ_{La} is decay constants of ^{140}La and f is the cumulative fission yield.

To apply the first part of the equation 1 to obtain the uranium fission interference factor, standards samples of uranium and lanthanum were prepared with well known masses and then irradiated. This procedure is here called experimental. To use the second part of the equation 1, nuclear parameters as well epithermal to thermal neutron fluxes ratio, must be used. This way, though use experimental results, is called theoretical.

The ^{140}La is a special case because the most part of the interference due to uranium fission occurs by the β^- decay of ^{140}Ba produced by the uranium fission. The Figures 1 shows how the β^- decay of ^{140}Ba interferes:

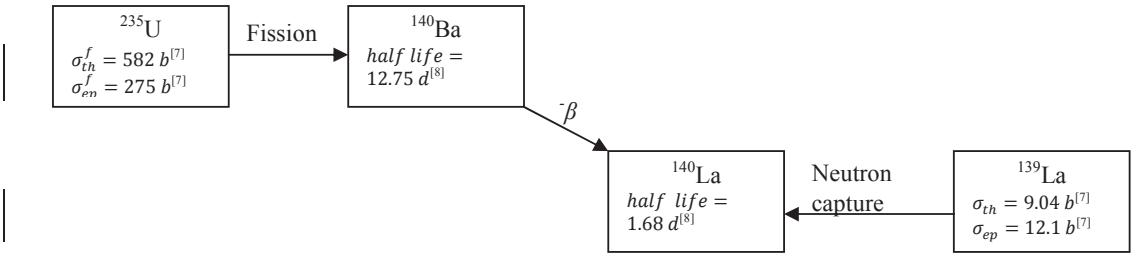

FIGURE 1. Formation of ^{140}La by β^- decay of ^{140}Ba and the formation of ^{140}La by neutron capture of ^{139}La.

Due to β^- decay of ^{140}Ba the activity specific of ^{140}La originally from uranium fission is dependent on decay time, consequentially the value of uranium fission interference factor for ^{140}La (F^{La}) is depending with decay time. Appling the radioactive equilibrium equation between ^{140}Ba and ^{140}La and using the definition of the uranium fission interference factor, the Equation 1 can be re-written as:

$$F^{La}(t) = \underbrace{\frac{\lambda_{Ba}}{\lambda_{La} - \lambda_{Ba}} \cdot \frac{N_{0(n,f)}^{Ba}}{N_{0(n,\gamma)}^{La}}}_{k} \cdot (e^{\lambda_{La} t_d} \cdot e^{-\lambda_{Ba} t_d} - 1) + \underbrace{\frac{N_{0(n,f)}^{La}}{N_{0(n,\gamma)}^{La}}}_{c} \quad (2)$$

Where t_d is decay time; $N_{0(n,f)}^{Ba}$, $N_{0(n,f)}^{La}$ and are number of atoms of the ^{140}Ba and ^{140}La formed by the fission of uranium, respectively and $N_{0(n,\gamma)}^{La}$ is numbers of atoms of ^{140}La formed by neutron capture.

Experimental Procedure

Preparation of Synthetic Standards

Were used certified standards solutions of U and La provide by Spex Certiprep USA, first these standards solutions were diluted and then pipetted 50 µl onto sheets of Whatman N_0 40. These pipetted standard were dried

inside a desiccator at room temperature and then the dried sheets were put into a desmineralized polyethylene bags. The masses of U and La were: La = (1996 ± 5) ηg and U = 1030 ηg.

Preparation of Monitor Fluxes

Was used the cadmium ratio technique to determine the epithermal to thermal neutron flux ratio[9]. The alloys of gold-aluminum (Certified Reference Material IRMM-530R with 0.1 % of gold) were cleaned using isopropyl alcohol and then they were weighed with masses about 3.5 mg each.

Irradiation and Counting of Standards and Fluxes Monitor

Were carried out 18 irradiations with standards of U and La to determine the uranium fission interference factor. The synthetic standards and monitor were irradiated in a position 14b shelf 3 in a IEA-R1 nuclear research reactor. The synthetic standards were irradiated inside a same device of irradiation called "rabbit" for 8 h. The activities were measured using the 1596 keV gamma ray recommended by IAEA[10] in different times of decay with range between 3 to 19 days, using a HPGe model GC1930 coupled a digital spectrum processor (DSA1000), both from Canberra, the acquisition times were about 9000 s for U and 3600 s for La.

The monitors of fluxes were irradiated in the same position of synthetic inside a same device of irradiation, however one was irradiated into a capsule of cadmium and the other without, but kept approximately 5 cm apart from each other to avoid neutron flux depletion. Were carried out 6 irradiations to determine the neutron flux. The system used to measure the activities was the same to the synthetic standards.

Results and Discussion

The monitoring of activities of uranium and lanthanum standards in different decay times shows that uranium fission interference factor varies strongly with the time decay, as can be seen in the Figure 2.

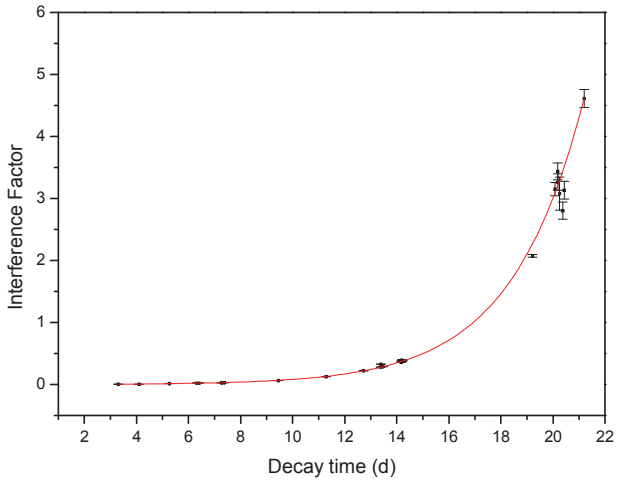

FIGURE 2. Variation of uranium fission interference factor for lanthanum as a function of decay time.

The epithermal and thermal neutron fluxes obtained by the mean of six irradiation were: $\phi_{ep} = (1.10 \pm 0.04) \times 10^{11}\, s^{-1} cm^{-2}$ and $\phi_{th} = (8.36 \pm 0.06) \times 10^{12}\, s^{-1} cm^{-2}$ respectively. Using these neutron flux values and nuclear parameters from literature it was calculated the "k" (here denominated k^{I}) and "c" constants presents in Equation 2. These values are presented in Table 2 together with obtained values by other authors.

TABLE (1). Values of "*k* "and "*c*" constants obtained in this work by nuclear parameters and epithermal to thermal neutron fluxes ratio and values obtained by other researches.

Reference	Constant	
	k^1	c
This Work	0.00236 ± 0.00001	(1.39 ± 0.001) x 10^{-6}
Glascock et al. (1986)[3]	0.0028 ± 0.0002	-
Al-Jobari et al. (1989)[2]	0.0018	-
Tshiashala (2005)[1]	0.025	-

Can be note that the "k^1" constant varies between different authors, it occurs because the uranium fission interference factor depends on the neutron fluxes ratio. The "*c*" constant represents the amount of ^{140}La produced directly by the uranium fission, this value is very low, in our case statistically consistent with zero and it is not found on the literature reported "*c*" constants values. The low value of "*c*" constant indicates that interference factor due to uranium fission is almost totally from the β^- decay of ^{140}Ba.

Considering the "c" constant in equation 2 equal to 0, was calculated the "*k*" constant using all measurements of uranium fission interference factor in different times decays (here denominated "k^2"). The value of "k^2" constant was obtained by weighted average of all irradiations. The obtained value was: $k^2 = (2.399 \pm 0.003) \times 10^{-3}$.

The "*k*" constant (here denominated "k^3") was obtained by the fitting of the graph present in Figure 2. The value of "k^3" constant was $k^3 = 0.00231 \pm 0.00002$ and it is very closely with the values obtained by the other ways, as can be seen in Table 2.

TABLE (2). Obtained values of "k" by three different methodologies.

Constant	Obtained Value
k^1	0.00236 ± 0.00001
k^2	0.00239 ± 0.00003
k^3	0.00231 ± 0.00002

The "*k*" value obtained in this work was applied to determine the real concentration of a Certified Reference Material (CRM) IAEA-SL-1 Lake Sediment. The Figure 2 shows the certified concentration of La and the obtained concentration in this study by the application of uranium fission interference factor.

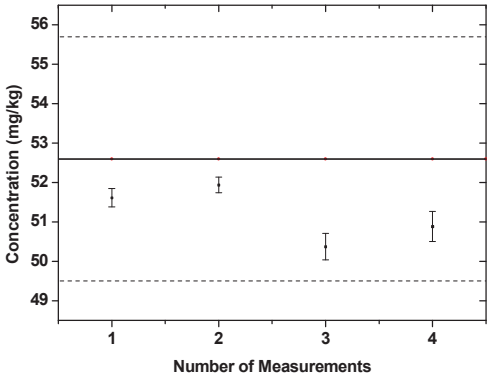

FIGURE 3. Certified concentration of La in a IAEA-SL-1 Lake Sediment Reference and the obtaine results using the uranium fission interference factor for ^{140}La.

The analisys of this reference material show us that the magnitude of correction is very low, it is because the certified concentration of U [4.02 ± 0.33 mg/kg] is very smaller than concentration of La [52.6 ± 3.1 mg/kg]. The magnitude of the correction depends on the ratio of U to La concentration.

CONCLUSIONS

The interferenc factor for ^{140}La varies strongly with the decay time, therefore the magnitude of the correction increase with the decay time. The interference due to uranium fission on the concentration determination of La is mostly due to $^{-}\beta$ decay of fission product ^{140}Ba.

Due to half life of ^{140}Ba (12.75 d) is bigger than half life of ^{140}La (1.68 d) is it impossible to wait a decay of ^{140}Ba, therefore is necessary to use the interference factor as function of decay time.

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