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Determination of uranium fission interference factors for INAA

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Abstract Instrumental neutron activation analysis (INAA) is a very suitable technique for the determination of several elements in different kinds of matrices. However, when the sample contains high uranium concentration this method presents interference problems of uranium fission products. The same radioisotopes used in INAA are formed in uranium fission. Among these radioisotopes are ^{141}Ce, ^{143}Ce, ^{140}La, ^{99}Mo, ^{147}Nd, ^{153}Sm and ^{95}Zr. The purpose of this study was to evaluate uranium fission interference factors to be used in the INAA of environmental and geological samples containing high levels of U. The obtained interference factors agreed with literature reported values. The results point to the viability of using these experimentally determined interference factors for the correction of uranium fission products.

Keywords Neutron activation analysis · Uranium fission product interferences · Correction factors

Introduction

Instrumental neutron activation analysis (INAA) is regarded as one of the well established analytical methods. It has been used in the determination of several elements in samples of interest from the geological, biological and environmental point of view.

However, in INAA, the determination of some lanthanide elements, molybdenum and zirconium is generally difficult in the presence of high uranium concentrations in the sample, as the same radioisotopes used in the determination of these elements are formed in uranium fission. The contributions of uranium interference depends on the concentrations of U and of elements to be determined as well as of irradiation conditions and decay time for counting.

In order to obtain reliable results for elements such as La, Ce, Nd, Sm, Mo and Zr in samples containing high level of U, it is necessary either to separate U from the sample before the irradiation or to carry out fission interference correction concerning the elements to be determined. The uranium fission interference factors have been determined and used in several studies over the years [1–11]. However these data obtained by irradiation in our IEA-R1 nuclear research reactor are very scarce.

The aim of this study was to evaluate uranium fission interference factors for ^{140}La, ^{141}Ce, ^{143}Ce, ^{147}Nd, ^{153}Sm, ^{99}Mo and ^{95}Zr radioisotopes commonly used in INAA of corresponding elements by irradiation at the IEA-R1 nuclear research reactor and to improve our analytical results.

The determination of these uranium interference factors is of great interest since the environmental samples constituted by plants and geological samples collected in uraniferous regions from the Brazilian territory contain relatively high levels of U.

Calculation of the interference factors

The interference factor F for a given isotope is defined as the ratio of the specific activity directly induced by neutron capture and the specific activity originated from uranium fission and it can be expressed by the following equation:

$$F = \frac{m_X}{m_U} \cdot \frac{A_U}{A_X} \quad (1)$$

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where m_X and m_U are the masses of the element of interest (X) and uranium, respectively; A_U and A_X are the activities originated from uranium fission product and from the activation of the element X, respectively.

The induced activity from a fission product and that induced directly by neutron capture of the element X can be expressed by Eqs. (2) and (3), respectively [12]:

$$A_U = \frac{m_U.a_U.N_0.f}{M_U}.\left(\phi_{th}.\sigma_{th}^U + \phi_{ep}.\sigma_{ep}^U\right).\left(1 - e^{-\lambda_X.t_{irrad}}\right) \quad (2)$$

$$A_X = \frac{m_X.a_X.N_0}{M_X}.\left(\phi_{th}.\sigma_{th}^X + \phi_{ep}.\sigma_{ep}^X\right).\left(1 - e^{-\lambda_X.t_{irrad}}\right) \quad (3)$$

where the indices X and U refer to element X and uranium, M is the atomic weight, a the isotopic abundance, N_0 Avogadro's number, f the cumulative fission yield, σ_{th}^U and σ_{ep}^U the fission cross sections for thermal and epithermal neutrons respectively, λ_X the decay constant of the radioisotope, ϕ_{th} and ϕ_{ep} are the thermal and epithermal neutron fluxes, respectively, σ_{th}^X and σ_{ep}^X the capture cross sections for thermal and epithermal neutron fluxes and t_{irrad} is the irradiation time.

Using Eqs. (2) and (3), the Eq. (1) can be written as follow:

$$F = \frac{M_X.a_U.f\left[\sigma_{th}^U + \left(\phi_{ep}/\phi_{th}\right).\sigma_{ep}^U\right]}{M_U.a_X\left[\sigma_{th}^X + \left(\phi_{ep}/\phi_{th}\right).\sigma_{ep}^X\right]} \quad (4)$$

By using Eq. (1), F can be determined experimentally by obtaining the ratio of the specific activities of the considered radioisotope measured in the standard of U and of element X, both irradiated and measured in the same experimental conditions. Moreover, F can be calculated using Eq. (4) and published data for the atomic masses, isotopic abundances and cross sections and reliable value for the thermal and epithermal neutron fluxes. These values of these parameters utilized in this study are presented in Table 1.

Experimental

Preparation of synthetic standard of elements

The certified standard solutions of elements provided by Spex Certiprep USA were first diluted in purified water for the preparation of diluted standard solutions of La, Ce, Nd, Sm, Mo, Zr and U. 50 μL of these diluted solutions were pipetted onto sheets of Whatman No. 40 filter paper. All the pipettes and volumetric flasks were calibrated before use. These filter sheets were dried at room temperature for about 24 h inside a desiccators, then placed into clean polyethylene bags and heat sealed. In these standards the quantities of each element, in μg (in parentheses) were the following: La (1.996), Ce (7.974), Nd (10.010), Sm (1.001), Mo (6.018), U (10.030) and Zr (100).

Determination of neutron fluxes

In order to obtain reproducible results all the irradiations of this study were performed in the same position of the IEA-R1 nuclear research reactor, coded 14b. For the neutron flux determinations, monitors of a gold–aluminum alloy (Certified Reference Material IRMM-530R with 0.1 % of gold) was used. Two samples of this monitor, weighing about 3.5 mg each, were cut, cleaned using isopropyl alcohol and then irradiated for 4 h. One monitor was irradiated bare and other inside a cadmium capsule in order to determine both the thermal and epithermal neutron fluxes. For this irradiation, these two monitors were placed in the same aluminum device (rabbit), but kept approximately 5 cm apart from each other to avoid flux depletion effects.

Irradiations

Firstly the irradiation of the flux monitors was carried out for 4 h, and then the synthetic standards of all the elements were irradiated together, in the same rabbit, for 8 h. The

Table 1 Nuclear parameters used in the calculation of uranium interference factors

Nuclear reactions	Isotopic abundance [13]	Atomic weight [14]	σ_{th} (b) [15]	σ_{ep} (b) [15]	Thermal fission yield [16]
$^{139}La(n,\gamma)^{140}La$	0.999088	138.905	9.04 ± 0.04	12.1 ± 0.6	0.06315 ± 0.00095
$^{140}Ce(n,\gamma)^{141}Ce$	0.88449	140.116	0.58 ± 0.02	0.54 ± 0.05	0.0586 ± 0.0015
$^{142}Ce(n,\gamma)^{143}Ce$	0.1108	140.116	0.97 ± 0.02	1.15 ± 0.05	0.0595[a]
$^{146}Nd(n,\gamma)^{147}Nd$	0.1719	144.24	1.41 ± 0.05	23.2 ± 0.5	0.02232 ± 0.0004
$^{152}Sm(n,\gamma)^{153}Sm$	0.267	150.36	206 ± 6	2,970 ± 100	0.001477 ± 0.000071
$^{98}Mo(n,\gamma)^{99}Mo$	0.2413	95.94	0.137 ± 0.005	6.9 ± 0.3	0.06132 ± 0.00092
$^{94}Zr(n,\gamma)^{95}Zr$	0.1738	91.224	0.0499 ± 0.0024	0.27 ± 0.03	0.06502 ± 0.00072
$^{235}U(n,f)$	0.0072	238.029	582	275	

[a] Glascock et al. [7]

Table 2 Uranium fission interference factors determined experimentally and theoretically and literature reported data

Radionuclide	Eγ(keV)	This study		Literature values		
		Experimental F	Theoretical F	Al-Jobori et al. [1][a]	Glascock et al. [7][b]	Tshiashala [2][c]
^{140}La	328.76	$(0.0021 \pm 0.0002)_{(e^{-0.35893t_d}-1)}$	0.0169 ± 0.0003	0.0018	–	0.025
^{140}La	487.021	$(0.0021 \pm 0.0002)_{(e^{0.35893t_d}-1)}$	0.0169 ± 0.0003	0.0018	$(0.0028 \pm 0.0002)_{e^{0.35893t_d}-1}$	0.025
^{140}La	815.77	$(0.0020 \pm 0.0001)_{(e^{0.35893t_d}-1)}$	0.0169 ± 0.0003	0.0018	–	0.025
^{140}La	1596.21	$(0.0021 \pm 0.0001)_{(e^{0.35893t_d}-1)}$	0.0169 ± 0.0003	0.0018	–	0.025
^{141}Ce	145.44	0.250 ± 0.006	0.281 ± 0.010	0.3	0.287 ± 0.008	0.282
^{143}Ce	293.27	1.227 ± 0.041	1.358 ± 0.019	1.38	1.35 ± 0.04	–
^{147}Nd	91.1	0.187 ± 0.018	0.204 ± 0.005	0.22	0.21 ± 0.01	0.226
^{153}Sm	103.18	0.0521 ± 0.0003	$(6.31 \pm 0.21)10^{-5}$	–	–	–
^{99}Mo	739.5	2.148 ± 0.160	2.135 ± 0.047	–	1.39 ± 0.05	2.15
^{95}Zr	724.2	9.683 ± 0.509	11.50 ± 0.33	7.3	–	10.88
^{95}Zr	756.73	9.718 ± 0.489	11.50 ± 0.33	7.3	11.3 ± 0.2	10.88

t_d is the decay time

[a] Experimental values

[b] Experimental values

[c] Calculated values

irradiations were carried out in the same day and in position 14b. The polyethylene bags containing the synthetic elements standards were individually wrapped up in aluminum foil, and then the whole set of standards was inserted together in a rabbit and irradiated shortly after the monitor. After about 4 days of decay time, the irradiated monitors and element standards were mounted in stainless steel planchets for gamma-ray measurements.

Activity measurements

The irradiated monitors and element standards were measured using a hyperpure Ge detector Model GC1930 coupled to a Digital Spectrum Processor DSA1000, both from Canberra. The resolution (FWHM) of the system was 0.90 keV for the 122 keV gamma-ray peak of ^{57}Co and 1.80 keV for the 1,332 keV gamma-ray of ^{60}Co. The measurements were carried out after two different decay times (~4 and 12 days) and the counting times from 3,600 to 36,000 s were used, based on the half-lives or activities of the radioisotopes considered. Gamma spectra were collected and processed using Canberra Genie 2000 Version 3.1 software.

Results and discussion

Six determinations were performed to obtain the mean value of cadmium ratio [17], and also the mean ratio of epithermal to thermal neutron flux, of 0.0172 ± 0.0010. The thermal and epithermal neutron fluxes obtained were $\phi_{th} = (4.63 \pm 0.81) \times 10^{12}$ s^{-1} cm^{-2} and $\phi_{ep} = (7.96 \pm 0.38) \times 10^{10}$ s^{-1} cm^{-2}.

Table 2 shows the uranium interference factors F determined experimentally using Eq. (1) and those calculated using the Eq. (4), together with reported values for comparison.

The experimental U interference factor F presented in this Table 2 are the mean values of six determinations and they presents good precision with relative standard deviation lower than 9.6 %. The theoretical F values are presented with standard deviations obtained by error propagation from the parameters utilized.

The interference factors determined in this study may be different when compared with literature values due to their dependence on the ratio of epithermal and thermal neutron fluxes, which may vary in different experimental setups. On the other hand, the experimental values obtained in this work are quite close to the ones calculated from literature data, and some small discrepancies may be due to simplifications made in these preliminary calculations (for instance self-shielding and flux depletion corrections, were not considered). These results point out the viability of using these interference factors determined experimentally for the correction of uranium fission products interference.

Conclusions

From the results obtained in this study we can conclude that the calculation of U interference factors special care is required since the interfering fission products are not

directly produced only by the fission of uranium, but from the product of the decay of fission products, or because of the problem of spectral interferences. The ^{140}La is a special case, because it is also formed from ^{140}Ba ($T_{1/2}$ = 12.75 days) fission product. Therefore the interference factor is generally given in function of decay time [1, 7, 9]. For 103.18 keV peak of ^{153}Sm, depending on the resolution of the detector there is spectral interference from the peak 103.7 keV of ^{239}Np that is formed by neutron capture of ^{238}U. The preliminary results obtained in this study encourage us the application of these interference factor in the analysis of actual samples.

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