

Modelling and Simulation

On the Fission Interference Correction and its Dependence on the Epithermal to Thermal Neutron Flux Ratio in NAA of Molybdenum

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It is well known that the presence of uranium in samples interferes with the determination of a number of elements by neutron activation analysis (NAA), namely molybdenum and zirconium, in particular due to the identity of some ^{235}U fission products and radionuclides formed by radiative capture of neutrons on those elements. The relation to be applied for correcting the fission interference is $[X]_{\text{true}} = [X]_{\text{app}} - F_X \cdot [U]$, where $[X]_{\text{true}}$ and $[X]_{\text{app}}$ denote the true and the apparent concentration of the element X in the sample, respectively, $[U]$ is the uranium concentration in the same sample and F_X (ppm of X per ppm of U, for example) is the fission interference factor concerning the element X. The extent to which the interference may affect the accuracy of the determination depends in particular on the ratio of the concentrations of the element to be determined and of the uranium in the sample, as well as on the neutron irradiation conditions.

Molybdenum is one of the elements which requires the most attention concerning the correction of the fission interference, for several reasons: molybdenum and uranium are both present, to more or less extent, in a very large number of samples, specially in those from geological and biological materials; the accurate determination of the concentration of molybdenum in geological samples may be very important in studying the processes taking place during the formation of rocks, in elucidating the petrogenesis of granitoids and ore deposits, etc.; and it is frequently necessary to determine the molybdenum concentration at low (sub ppm) levels in many biological materials, and, although the analysts often neglect *a priori* their uranium content, the fact is that the uranium concentration in these materials can be significant. In such instances the problem of correcting for the fission interference becomes particularly crucial, because molybdenum is an essential or toxic element for living organisms, thus of importance in biomedical sciences, nutrition studies, etc..

Experimental values of F_{Mo} found in the literature are very much scattered covering an interval from 1.04 (Wu *et al.*, 1997) to 1.86 (Martinho *et al.*, 1986). Danko and Dybczynski (1997) conclude that F_{Mo} varies in the range 0.7–1.5, but the concentrations in the biological samples which were analysed appear to support a different range of values (0.1 to 1.1). W. Tian (1987) proposes a neutron spectrum-independent “compound nuclear constant”, Ik_0 , for the fission interference correction in reactor NAA by the k_0 -method, but from his data it appears that the neutron spectrum-dependent data for F_{Mo} which were obtained range from 0.80 to 2.16. Several authors consider the scatter in the F_{Mo} data as being due to different sample irradiation conditions, particularly concerning the relative importance of the epithermal component of the neutron spectrum at the irradiation facility used. However, to our knowledge, no study has yet been published where such a correlation might have been established clearly in order to provide a satisfactory basis to explain the scatter in the experimental data.

Consequently, the present work aims at: (1) obtaining experimental F_{Mo} data under controlled irradiation conditions concerning the knowledge of the epithermal to thermal neutron flux ratio Φ_{epi}/Φ_0 , widening the interval of values as much as the Portuguese Research Reactor makes it possible; (2) analysing as widely as possible the experimental F_{Mo} data obtained by the various

authors, namely those who have described the irradiation conditions used as concerns Φ_{epi}/Φ_0 ; (3) establishing a simple calculation model which may describe the dependence of F_{Mo} on Φ_{epi}/Φ_0 in an adequate way, to be validated by the available experimental data; and (4) clearly indicating the basic recommendations to take into account in order to obtain with high accuracy the concentration of molybdenum in samples containing uranium, namely biological samples.

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Determination of the ^{235}U Abundance in Uranium by Neutron Activation on the Basis of the Molybdenum Fission Interference

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Abstract

Practical experience reveals that one finds in the market compounds, metallic alloys and other uranium materials where this element is depleted in ^{235}U . This happens frequently and is due to the production of enriched uranium which is the counterpart. For certain experiments involving the utilization of the neutron activation analysis (NAA) technique, one needs to use uranium of a high degree of nuclear purity, but with the natural abundance in ^{235}U or with a known isotopic composition. In the present market conditions, there may therefore be an interest in knowing whether the uranium which is present in a given material is natural or depleted uranium. On the other hand, an expert in NAA may be asked to determine the ^{235}U abundance in a given uranium sample, either a natural, depleted or slightly enriched one.

The ^{235}U abundance can be determined using several analytical techniques, such as isotope dilution mass spectrometry and alpha radiation spectrometry, for example. NAA can also be used on the basis of a ^{235}U certified standard. The purpose of the present work is to suggest a method for determining the ^{235}U abundance through a particular application of the NAA technique, on the basis of the fission interference concerning molybdenum, which does not require ^{235}U certified standards.

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Production of radionuclides to medical applications:

- Calculation of specific activity of ^{64}Cu , ^{89}Sr , ^{90}Y , $^{117\text{m}}\text{Sn}$, ^{142}Pr , ^{153}Sm , ^{159}Gd , ^{165}Dy , ^{166}Ho , ^{170}Tm , ^{177}Lu , and ^{186}Re
- Study of the generator $^{188}\text{W} \rightarrow ^{188}\text{Re}$

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As sources of neutrons, nuclear reactors are used to produce radionuclides (via neutron activation of appropriate target materials) to be applied in medical research, care or treatment. In this work the specific activity of some radionuclides (^{64}Cu , ^{89}Sr , ^{90}Y , $^{117\text{m}}\text{Sn}$, ^{142}Pr , ^{153}Sm , ^{159}Gd , ^{165}Dy , ^{166}Ho , ^{170}Tm , ^{177}Lu , and ^{186}Re) has been estimated considering the irradiation conditions available at the Portuguese Research Reactor (RPI), and a general theoretical study concerning the production of the generator $^{188}\text{W} \rightarrow ^{188}\text{Re}$ has been performed.

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NAA of Zirconium in Uraniferous Samples and the Fission Interference Correction

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Abstract

The present work is similar to a previous one [1], but it concerns the neutron activation analysis (NAA) of zirconium (instead molybdenum) in the presence of uranium, and the corresponding fission interference factor, F_{Zr} . This work aims at: (1) analysing as widely as possible the experimental F_{Zr} data obtained by the various authors, namely those who have described the irradiation conditions used as concerns the epithermal-to-thermal neutron flux ratio, Φ_{epi}/Φ_0 ; (2) establishing a simple calculation model which may describe the dependence of F_{Zr} on Φ_{epi}/Φ_0 in an adequate way, to be validated by the available experimental data (and to provide a satisfactory basis to explain the scatter found in the experimental F_{Zr} data); and (3) clearly indicating the basic recommendations to take into account in order to obtain with high accuracy the concentration of zirconium in samples containing uranium, namely geological samples.

[1] E. Martinho, M.C. Freitas, "On the fission interference correction and its dependence on the epithermal to thermal neutron flux ratio in NAA of molybdenum", *Biological Trace Element Research* (in press)

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Monte Carlo Calculation of Fast Neutron Spectra Inside a Lead Hollow Cylinder

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Abstract

Experiments involving irradiation of samples in research nuclear reactors, specially those for studies of changes of properties induced by fast neutrons in materials, require the knowledge of the absorbed dose in order to establish dose-effect correlations.

This work presents the results of a simulation study carried out in order to enhance the fast neutron spectra in a sample irradiated in a thermal neutron reactor. The irradiation device consists of a lead hollow cylinder where an aluminium sample holder containing the sample to be irradiated is placed. Calculations were performed considering that the space between the fission neutron source and the irradiation device was filled either by water or beryllium+water, and the thickness of the lead wall of the irradiation device was varied up to 10 cm. The fast neutron spectrum in the sample was calculated, and the average fast neutron energy, $\langle E \rangle$, the total neutron fluence, $\phi(E > 0.1 \text{ MeV})$, the fast neutron fluence, $\Phi(E > 1 \text{ MeV})$, the conversion factor, C , and the gain factor of the fast neutron flux, G , were determined.

The lead container surrounding the sample induces an appreciable enhancement on the fast neutron spectra. The gain of the fast neutron flux varies between 2 and 4.5 when the lead thickness varies between 2 and 10 cm.

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