

Background

Neutron activation analysis (NAA) is a physical technique used for the absolute measurement of the concentration of substances in solids and liquids. The method uses neutron irradiation which is commonly realised using a nuclear reactor in order to activate (make radioactive) different isotopes of the elements present in the sample. The radionuclides produced in this way emit gamma-rays that are characteristic of the elements present in the sample. Using gamma-ray spectrometry these radionuclides can then be identified and quantified, and hence their concentration in the sample can be determined. Although NAA is a straightforward method it requires a sound control of the many physical parameters involved to obtain accurate results and to guarantee a set accuracy in routine analysis.

The accuracy of NAA depends on the specific measurement method used. One can perform NAA in a relative way by co-irradiating a known standard and the unknown sample in the same conditions and by comparing the ratio of gamma-rays they emit. Relative NAA has limited applicability since it requires reference standards with a comparable composition as the unknown. A more generally applicable method is the k_0 -NAA method. In the k_0 -NAA method all measurements are relative to the element Au resulting in ^{198}Au when irradiated. The k_0 -NAA method further relies on the fact that the neutron energy spectrum produced in a given position in the reactor can be parameterised with two parameters:

- α : the shape factor of the epithermal neutron flux, indicating the deviation of the epithermal neutron spectrum from the ideal $1/E$ shape approximated by a $1/E^{1+\alpha}$ distribution, with E the neutron energy;
- f : the thermal-to-epithermal neutron flux ratio.

The parameters f and α are characteristic for the irradiation facility (reactor and irradiation channels) and may change or fluctuate in time according to the irradiation conditions. The way elements activate (become radioactive) when interacting with neutrons is determined by the nuclear cross sections. The cross sections depend on the neutron energy E and also these physical properties are parameterised in the k_0 -NAA method resulting in the so-called k_0 -factors for each element and for the different isotopes of an element. The quantification of an element then involves the use of a ratio of k_0 -factors in which one k_0 -factor is the one for ^{198}Au . In practice, ratios of k_0 -factors accounting for the energy spectrum of the neutrons (f , α) are used in an analysis. These ratios are called the comparator factor F_C .

It is clear that the quality assurance of the k_0 -NAA method requires the control of the parameters f and α for each irradiation of samples. This is generally realised by using several flux monitors e.g. a set of Zr and Au monitors which are co-irradiated with the samples to analyse. However, this approach is quite labour intensive and recently we started investigating an approach based on co-irradiation of Synthetic Multi-Element Standards (SMELS) for this purpose. SMELS contain different elements in known concentrations, and three types of materials exist: Type I (elements forming short-lived radionuclides), Type II (elements forming medium lived radionuclides) and Type III (elements forming long lived radionuclides). Au was added to all three SMELS types and Zr in type III.

Objectives

The objectives are to optimise the ratio of quality of analysis to workload by appropriate quality control methods to determine and monitor the irradiation parameters (f , α) using SMELS and to set up a Quality Control (QC) system for the complete measurement and analysis process for k_0 -NAA.

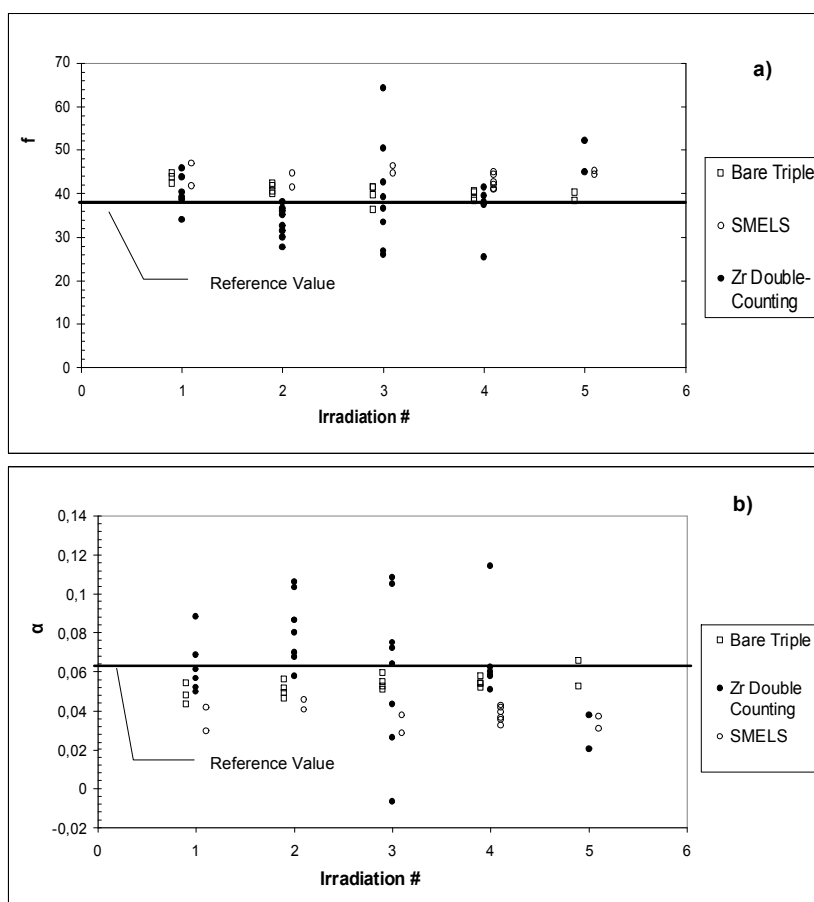
Principal results

Different well known methods exist for the determination of the neutron flux parameters f and α in channel Y4 of the BR1 reactor. Among these we used the following three methods as reference methods for comparison with the SMELS methods:

- The “Bare Triple-Monitor”-method which involves the quantification of ^{197}Au – ^{94}Zr – ^{96}Zr from a Au-Zr monitor;
- The “Zirconium Double-Counting”-method which involves the quantification of ^{197}Au - ^{95}Zr and $^{97}\text{Zr}/^{97\text{m}}\text{Nb}$ by two gamma-ray spectrometry measurements optimising counting statistics
- The “Cadmium ratio for multi monitor”-method. which involves two irradiations, one with and one without Cd-shielding, of a large set of single element monitors (foils).

The figures (a) and (b) below compare measurement data of f respectively α obtained by analysis using the different methods discussed above, using the “Cadmium ratio for multi monitor”-method as the reference. From these graphs, the precision and accuracy of the methods can be determined. The following conclusions have been drawn from the experiments:

- Comparison of the results obtained by the different methods show that the parameters f and α are clearly correlated and that the couples (f, α) for the different methods can be fitted with a unique relation.
- A small bias is observed for the different methods compared to the reference.
- Separate monitoring in QC of f and α as single parameters shows not to be a good approach. Indeed, limited precision is obtained due to the correlation.
- Monitoring the quantity $f \times F_C$ and the deviations from an empirical curve expressing $f(\alpha)$ seem the best solution for QC monitoring purpose.
- The measurements prove that the variability of f and α are well under control (constant) for irradiations in BR1.
- QC of f and α can be based on the use of Zr and Au in SMELS as monitor. Moreover, the SMELS monitor also provides a more direct and fast control by observing the deviations of the measured concentrations of the different elements with respect to the reference concentrations. As it does not involve the preparation of separate Au/Zr foils and extra measurements this is a fast method.



Variation of f and α for different irradiations (multiple monitors per irradiation) with the three techniques involved.

Future work

It will be investigated whether SMELS can be used as a "single monitor irradiation" for the determination of f and α using the “Cadmium ratio for multi monitor”, thus avoiding the need of multiple evaluations.

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Main reference

Using synthetic multi-element standards (SMELS) for calibration and quality control of the irradiation facilities in the BR1 reactor; P. Vermaercke, P. Robouch, L. Sneyers, F. De Corte, J. Radioanal. Chem. (to be published in 2007)