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DETECTOR EFFICIENCY DETERMINATION APPLIED TO THE k_0-NAA METHOD

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ABSTRACT

The IAEA-k_0 software offers an easy and quick way to calculate elemental concentration in samples when the k_0-NAA method is used; among other options, the software offers the possibility of including a correction for situations when the sample analysis requires the use of some absorber (for instance, a sample holder) that was not present when the detection efficiency calibration was performed. In the present study the mathematical absorber correction included in the IAEA-k_0 software was tested by analyzing the same sample measurement twice, first using calibration spectra obtained by placing the calibration sources over the same sample holder that is used when counting the samples (thus not requiring the use of the absorber correction in the software) and, also, using a second set of calibration spectra obtained by directly placing the calibration sources at the same position where the sample was counted and enabling the absorber correction in the IAEA-k_0 software. The concentration results obtained were then compared, and the mathematical correction proved to give reliable results.

1. INTRODUCTION

Neutron Activation Analysis (NAA) is an analytical technique consisting in the irradiation of a sample of unknown elemental composition in a neutron flux and the composition of this sample is then obtained by measuring the gamma-ray activity induced in the sample. In general, if a given mass of some element (m_e) is subjected to a thermal neutron flux ϕ_{th} for a period of time (t_i), the activity of a specific gamma transition induced in the sample due to the irradiation of this element is [1]:

$$Activity = \frac{A_p}{\varepsilon(E_\gamma)} = \frac{m_e \cdot N_A \cdot \phi_{th} \cdot \sigma \cdot I_\gamma \cdot F_I \cdot \left(1 - e^{-\lambda \cdot t_i}\right) \cdot \left(1 - e^{-\lambda \cdot t_c}\right) \cdot e^{-\lambda \cdot t_e}}{M_A \cdot \lambda} \quad (1)$$

where σ is the target isotope's thermal neutron cross section, $\varepsilon(E\gamma)$ the detector efficiency for the energy of that gamma-ray, A_p is the observed count rate of the peak related to the gamma transition, N_A is Avogadro's constant, $I\gamma$ is the absolute intensity of the gamma ray in that decay, F_I is the isotopic fraction of the target isotope, M_A is its atomic mass, λ is the decay constant of the radionuclide produced and t_e and t_c are the waiting and counting times, respectively.

The concentration of any element in the irradiated sample can, then, be quantitatively determined if all the parameters in Eq. 1 are known - the neutron flux (ϕ_{th}) can also be obtained by means of Eq.1 by irradiating a flux monitor (i.e., a sample with well-known composition – usually a gold foil) together with the sample, and this method is known as *absolute NAA*. Nevertheless, the propagation of the uncertainties from all the parameters involved in the determination of both the neutron flux and of the sample concentration usually leads to large uncertainties, so alternative NAA methods have been proposed, such as the comparative NAA [2] or, more recently, the k_0-NAA [3].

k_0-based instrumental neutron activation analysis is a method which combines the simplicity of the absolute method with the accuracy of the comparative one. This method provides the determination of almost all elements whose gamma-ray peaks are present in the gamma spectrum. The concentration of elements is calculated using one element, usually gold, as neutron flux monitor and then using literature values for the element-specific parameter k_0, eliminating the need of elementary standards [4]; the concentration, then, is given by:

$$\rho_a = \frac{A_{sp,a}}{A_{sp,Au}} \cdot \frac{\varepsilon_{p,Au}}{\varepsilon_{p,a}} \cdot \frac{1}{k_{0,Au}(a)} \cdot \frac{G_{th,Au} \cdot f + G_{e,Au} \cdot Q_{0,Au}(\alpha)}{G_{th,a} \cdot f_i + G_{e,a} \cdot Q_{0,,a}(\alpha)} \qquad (2)$$

where subscript “a” is for analyte and Au is for gold and it refers to $^{197}Au(n,\gamma)^{198}Au$ (E_γ = 411,8 keV and $k_{0,\,Au}$ = 1), A_{sp} is the specific count rate, ε_p is the full peak efficiency, $k_{0,Au}$ is the k_0 factor of an element related to that of the gold, G_{th} is the correction factor for thermal neutron self-shielding, G_e is the correction factor for epithermal neutron self-shielding, f is the thermal-to-epithermal neutron flux ratio and $Q_0(\alpha)$ is the ratio of resonance integral I_0 to thermal neutron cross-section σ_0 in the $1/E^{1+\alpha}$ representation).

On the other hand, this method requires the knowledge of gamma detection efficiency of the counting system for the energies of both the gamma ray from the radioisotope to be determined and the 411,8 keV gamma-ray from the decay of ^{198}Au. Moreover, as the standard gamma sources used in detector calibration are usually encapsulated into polyethylene, the efficiency calibration is often performed without the presence of any absorber between the source and the detector, while the samples are placed upon some container, usually a stainless-steel disc, thus requiring that some sort of efficiency correction be applied to the calculation.

In the case of the present study, the analysis of gamma-ray spectra and the calculation of concentration are performed by specially-developed software, the k_0-IAEA [5]. This software also allows correcting the influence of the sample container. The aim of this work was, then, to evaluate this correction by making two different efficiency calibrations: one without the stainless-steel disc (thus demanding the use of the efficiency correction in the analysis) and the other with the sample container, so that the calibration sources are used in the exact same geometry as the samples and the efficiency correction is not needed.

2. EXPERIMENTAL PROCEDURE

Initially, the efficiency of the gamma-ray detection system was assessed by measuring calibrated standard gamma-ray sources of ^{152}Eu and ^{137}Cs. The detector system used was a

planar 20%-HPGe detector with a beryllium window, coupled to a regular analogical nuclear spectroscopy electronic system and to an 8192-channel MCA analyzer; the source-detector distance was fixed at 30.5 cm. This determination was performed twice, once simply placing the radioactive sources in the desired position and the other using a 0.1 mm thick stainless-steel source container (Fig.1) under the calibration sources.

Figure 1. A view of the stainless-steel sample holder used in the measurements. Scale of the caliper rule in centimeters.

Once the two calibrations were performed, a 149.1 mg test sample was weighted, placed inside a previously cleaned polyethylene bag and irradiated together with a thin 44 mg Au wire in the IEA-R1 research reactor for 16h under an estimated thermal neutron flux of $7.8 \times 10^{12} cm^{-2} s^{-1}$. After two weeks of decay, the sample was then counted in the previously-calibrated detector system with the same source-detector distance at which the calibrations were performed; in order to perform the counting of the sample, the use of a sample container as the one shown in Fig.1 is mandatory; the Au flux monitor was also counted.

The spectra obtained in the calibration where the sample container was used were then used, together with the spectra obtained counting the sample and the Au monitor, as input for the IAEA-k0 software, which performed the calculations to determine the elemental concentration of the sample using Eq.2, where the flux parameters f and α were obtained

from a previous characterization measurement performed at the same irradiation position [6] as: f = 35 and α= 0.05). Again, the same procedure was repeated using the calibration spectra obtained without the sample container and enabling the absorbed correction procedure available in the IAEA-k0 software; the results obtained in the latter measurement were compared to the ones obtained using a sample holder at all times in order to verify the accuracy and reliability of this absorber correction.

3. RESULTS AND DISCUSSION

The calibration curves obtained for the two efficiency calibration measurements are shown in Fig.2.

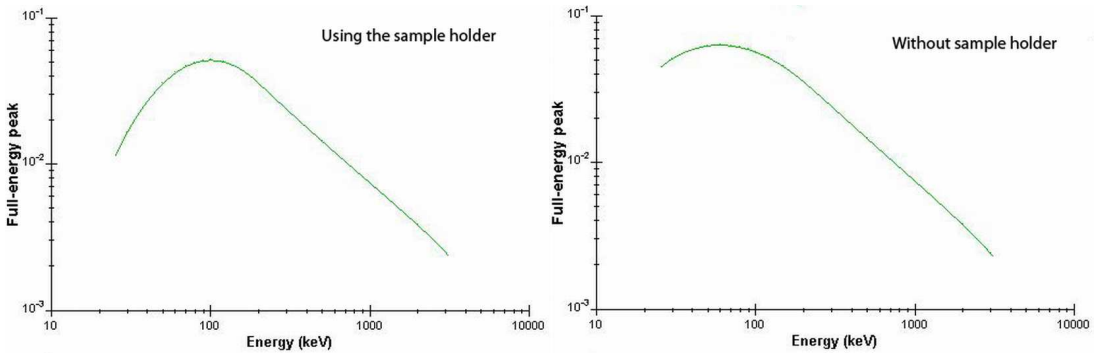

Figure 2. Calibration curves obtained with (left) and without (right) the use of a sample holder.

From all the results obtained in the measurements, the results for Ce and Sm were selected because they rely on low-energy gamma-rays, where the absorber correction should be more relevant; the results for Br and K were also selected because they had the lowest uncertainty among all results, so they could work as a good benchmark, too; these results are shown in Table 2, where the z-score was calculated assuming the measurement that didn't involve the absorber correction as the true value. These results show that, while the difference was larger for Sm (as expected, as the Sm calculation relies on the lowest energy gamma-rays, at 70 and 103 keV), the result obtained using the mathematical correction was still acceptable, with a z-score only slightly over 1; for the other three elements the results of both measurements were in excellent agreement, thus showing that the mathematical absorbed correction used in the IAEA-k_0 software can be safely used when the calibration can't be performed physically using the same absorbers that will be present when the samples are counted.

Table 1. Elements and gamma-ray energies used in this work [7].

Element	Energy (keV)	Transition Intensity (%)
Ce-141	145.44	100.00
Sm-153	69.66	20.00
	103.17	100.00
Br-82	554.34	80.00
	776.50	100.00
K-42	1524.70	100.00

Table 2. Concentration results obtained for Ce, Sm, Br and K with and without the use of the mathematical absorber correction.

Element	Concentration (mg/kg)		z-score
	Using the absorber correction (mg/kg)	Without the absorber correction (mg/kg)	
Ce	13.8 ± 1.1	13.4 ± 1.2	-0.24
Sm	1.19 ± 0.05	1.27 ± 0.05	1.13
Br	13.3 ± 0.5	13.1 ± 0.5	-0.32
K	32320 ± 1584	31240 ± 1687	-0.47

As for the uncertainties introduced by the absorber correction, Table 3 shows the relative standard deviations given by the IAEA-k_0 software in both cases, where it is clear that the results obtained using the absorber correction had consistently lower uncertainties; this is an indication that the software doesn't take into consideration the uncertainties arising from this correction, so some care should be taken with the uncertainties obtained when using the correction, especially at lower energies, where its influence is larger.

Table 3. Comparison of the relative standard deviations of the concentration values obtained with and without the mathematical absorber correction.

Element	Relative Standard Deviation (%)	
	Using the absorber correction	Without the absorber correction
Ce	8.2	9.3
Sm	3.9	4.1
Br	3.5	3.7
K	4.9	5.4

3. CONCLUSIONS

The tests performed here show that the absorber correction used in the IAEA-k_0 software delivers reliable results and can be safely used when it's not possible to perform the calibration using the same absorbers that will be present in the sample measurements; on the other hand, the software doesn't seem to do any uncertainty estimation or propagation for these corrections, so some care is advised when the experimental uncertainties are too low (which was not the case in the present measurement).

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