2015 International Nuclear Atlantic Conference - INAC 2015
São Paulo, SP, Brazil, October 4-9, 2015
ASSOCIAÇÃO BRASILEIRA DE ENERGIA NUCLEAR - ABEN
ISBN: **978-85-99141-06-9**

DETERMINATION OF k_0 FOR ^{63}Cu(n,γ)^{64}Cu REACTION WITH COVARIANCE ANALYSIS

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ABSTRACT

Nowadays the k_0 Method is one of the most used procedures on Neutron Activation Analysis (NAA). For an element of interest, the parameter k_0 can be used to determine its mass concentration. The recommended nuclear data has been investigated, and the measurement of this parameter for ^{63}Cu(n,γ)^{64}Cu reaction was motivated by some discrepancies that were observed in the literature. The irradiations were performed near the core of the IEA-R1 4.5 MW nuclear research reactor of the Nuclear and Energy Research Institute – IPEN-CNEN/SP, in São Paulo, Brazil. Two irradiations were carried out in sequence, using two sets of samples: the first one with a cadmium cover around the samples and the second one without it. The activity measurements were carried out in a previously calibrated HPGe gamma-ray spectrometer. Standard sources of ^{152}Eu, ^{133}Ba, ^{60}Co and ^{137}Cs supplied by the IAEA with gamma transitions ranging from 121 keV to 1408 keV were used in order to obtain the HPGe gamma-ray peak efficiency as a function of the energy. The covariance matrix methodology was applied to all uncertainties involved. The resulting value of k_0 for ^{63}Cu(n,γ)^{64}Cu reaction for the gamma transition energy of the formed isotope ^{64}Cu 1345.77 keV was 4.99x10^{-4} (78).This final value for k_0 has been compared with the literature.

1. INTRODUCTION

The knowledge of chemical element concentrations in samples is very important in various fields of science. It is relevant to mention the concentration of chemicals in foods, environmental studies, in biological samples, geological samples, metal alloys, ceramics archaeological, among others. Among others, the Neutron Activation Analysis (NAA) is a well-known technique for determining multi-element concentrations in different materials. As part of this technique, the k_0 Method is nowadays widely used by laboratories performing NAA all over the world, and applied to many fields of science [1].

Copper has been one of the earliest metals used by man. Some copper properties are responsible for its intense use nowadays, such as: abundance, attractive color, ease of working and resistance to corrosion [2].

The modern civilization is heavily dependent on copper and its derived products are used both in industry and in agriculture. As a consequence, serious problems of contamination to the environment may occur, particularly in rivers, lakes and seas. Besides, copper ores can be found in large deposits, relatively close to the surface, and be extracted by amenable opencast mining methods [2]. For these reasons, it is very important to know accurately the copper concentration in environmental samples. An important way to find the copper concentration measurement is by means of NAA.

The NAA technique can be applied in two different ways: in the analysis technique by Instrumental Neutron Activation that uses the comparative method, in which a standard element is irradiated together with the sample of interest, and in the k_0 method case, in which the sample is irradiated together with a comparator (usually Au), and from the ratio between the sample and comparator activities, the element concentration can be derived [3]. The k_0 method has some advantages over the comparative method that usually is laborious, expensive and time-consuming. Since its introduction, the k_0 methodology and its protocols have grown from a mere theoretical concept to a fully operational tool. There are estimates that k_0 Method is in operation today in more than 50 industrial laboratories, universities and government around the world [4].

Despite the efforts of several laboratories in activity, some parameters related to Method k_0 still require a more exact investigation: some are considered discrepant cases, considering the different results found in the literature, some data is still missing. Therefore improvement of this method is of great importance. In order to achieve good results, there is a continuing need for improving the accuracy of k_0 parameter for several neutron capture reactions [1]. This fact motivated the present work which is focused on the measurement of k_0 value for the $^{63}Cu(n,\gamma)^{64}Cu$ reaction, with the purpose of improving the existing data catalogues.

2. MATERIALS AND METHODS

2.1. k_0 equation

The parameter k_0 can be obtained by the following relationship [3]:

$$k_{0,i} = \frac{A_{sp,i} - \dfrac{(A_{sp,i})_{Cd}}{F_{Cd,i}}}{A_{sp,c} - \dfrac{(A_{sp,c})_{Cd}}{F_{Cd,c}}} \times \frac{G_{th,c}}{G_{th,i}} \times \frac{\varepsilon_c}{\varepsilon_i} \qquad (1)$$

where $k_{0,i}$ is the k_0 factor of sample i with respect to the comparator (Au); $(A_{sp,i})_{Cd}$ and $A_{sp,i}$ are the gamma-ray total energy absorption peak area of the reaction products, obtained by HPGe gamma-ray spectrometry measurements, with and without cadmium cover, respectively. These values were corrected for saturation, decay, cascade summing, geometry, measuring time and mass; ε_c and ε_p are the peak efficiencies for the comparator and target nuclei, respectively. The $G_{th,c}$ and $G_{th,i}$ are the thermal neutron self-shielding factors, for comparator and sample, respectively.

The cadmium factor F_{Cd} is calculated by the average transmission in the cadmium cover, applying cross section data from ENDF/B-VII [5] and considering variation in the cadmium thickness due to isotropic neutron flux.

The following equation was applied [6]:

$$F_{Cd} = \frac{\int_0^{\infty} t(E)\sigma(E)\phi(E)dE}{\int_{E_{Cd}}^{E_3} \sigma(E)\phi(E)dE} \quad (2)$$

In the present work this equation has been approximated by:

$$F_{Cd} = \frac{\sum_i t(E_i)\sigma(E_i)\phi(E_i)\Delta E_i}{\sum_i \sigma(E_i)\phi(E_i)\Delta E_i} \quad (3)$$

The transmission factor $t(E_i)$ is the given by:

$$t(E_i) = e^{-N_{Cd} \cdot d \cdot \sigma_{Cd}(E_i)} \quad (4)$$

In this equation, N_{Cd} is the number density of cadmium atoms, d is the crossing distance inside the cadmium layer and $\sigma_{Cd}(E_i)$ and $\sigma(E_i)$ are the cadmium and sample absorption cross sections, respectively, taken from ENDF/B-VII [5]. The neutron spectrum $\phi(E_i)$ was assumed to follow the 1/E law. E_{Cd} and E_3 are the cadmium cut off energy and the upper energy limit, assumed to be 0.55eV and 2 MeV, respectively. ΔE_i corresponds to the i-th energy bin from the cadmium cross section library. The sample cross section value was interpolated to match the same energy found in the cadmium cross section table.

In order to account for isotropic neutron incidence, the cadmium factors given by Eq. 3 have been averaged with respect to the solid angle Ω_i covered by the cadmium box, according to the following expression [7]:

$$\overline{F_{Cd}} = \frac{\sum_k F_{Cd,\Omega_k} \Delta\Omega_k}{\sum_k \Delta\Omega_k} \quad (5)$$

The thermal neutron self-shielding factor G_{th} has been determined as follows [8]:

$$G_{th} = \frac{1}{1 + \left(\frac{z}{1{,}029}\right)^{1{,}009}} \quad (6)$$

with

$$z = x\Sigma_t \left(\frac{\Sigma_a}{\Sigma_t}\right)^k \quad (7)$$

Where k=0.85±0.05, Σ_t and Σ_a are the total and absorption macroscopic cross sections averaged over the thermal neutron spectrum at room temperature, respectively, and x is the typical dimension of the sample for a given geometry ($x=t$ foil thickness; $x=R$ wire or sphere radius; $x= rh/(r+h)$, r and h: radius and height of the cylinder) [8].

2.2 Gamma-ray detection efficiency curve

The peak efficiency $\varepsilon_p(E)$ [9] corresponds to the ratio between the number of events recorded in the total absorption peak, and the number of photons emitted by the source being represented by the Eq. 8:

$$\varepsilon_p(E) = \frac{S_p(E)}{I_\gamma \, At} . f_c \tag{8}$$

where $S_p(E)$ is the area under the total absorption peak for the energy range considered, I_γ is the gamma emission probability per decay, A is the source activity, t is the measuring time, f_c are correction factors for dead time, detection geometry, radioactive decay, source self-attenuation and cascade summing.

2.3 Covariance matrix methodology

The covariance matrix methodology is necessary for rigorous statistical analysis and was applied to all uncertainties involved. A series expansion of a multi-parametric function may be given by [10]:

$$Y = Y(a_1, a_2, a_3, ..., a_n) \tag{9}$$

The variance of Y is given by:

$$\sigma_Y^2 \cong \sum_{\nu=1}^{n} \frac{\partial Y}{\partial a_\nu} \sum_{\lambda=1}^{n} \frac{\partial Y}{\partial a_\lambda} \langle (a_1 - a_{0,\nu})(a_\lambda - a_{0,\lambda}) \rangle \tag{10}$$

The partial derivatives in Eq.10 are calculated at $a=a_0$, where a_0 is the expectancy value of a. The covariance of a_ν with respect to a_λ is called $cov(a_\nu, a_\lambda)$ and usually has a non-zero value. The $cov(a_\nu, a_\lambda)$ is given by:

$$\mathrm{cov}(a_\nu, a_\lambda) = \langle (a_\nu - a_{0,\nu})(a_\lambda - a_{0,\lambda}) \rangle = \sum_{k=1}^{m} \rho_{\nu,\lambda,k} \sigma_{\nu,k} \sigma_{\lambda,k} \tag{11}$$

Where $k = 1, ..., m$ is the partial uncertainty index.

When a_ν is independent of a_λ the covariance is zero.

2.4. Sample preparation, irradiation and measurement

A Hyper Pure Germanium (HPGe) detector was used in this work, CANBERRA, GX020 model, cylindrical geometry with efficiency relative 20% for energy 1332.5 keV of ^{60}Co. The energy resolution obtained experimentally was 2.15 keV. The associated electronic system comprises a pre-amplifier and high voltage filter incorporated into the cryostat, a INTERCHNIQUE amplifier, model 724, a ORTEC multichannel analyzer, ACE model with 8192 channels and a microcomputer PC compatible.

The HPGe gamma-ray peak efficiency curve was obtained making use of ^{60}Co, ^{133}Ba, ^{137}Cs and ^{152}Eu standard sources supplied by the IAEA (International Atomic Energy Agency), considering 15 data points in the energy range from 121 and 1408 keV. The distance from the radioactive source to HPGe detector front face was approximately 17.9 cm in order to minimize cascade summing corrections.

The efficiency was adjusted as a function of the gamma-ray energy by a polynomial in log-log scale [11], applying the least square method with covariance matrix. This method can provide information on the correlation between each pair of data points and between each pair of fitted coefficients [12].

The selected targets were ^{197}Au (0.10% Al alloy) and Cu (99.9%), activated by (n,γ) reaction. The samples were sealed in polyethylene envelopes. The targets were placed inside an aluminum rabbit 7.0 cm long, 2.1 cm in diameter and 0.05 cm thick wall, encapsulated by an aluminum sheet and attached to an aluminum rectangular plate centered within the rabbit. The masses ranged from 1 (^{63}Cu) to 15 (^{59}Co) mg, with an uncertainty of ±20 μg.

Two irradiations were carried out in sequence using two sets of samples: the first without a cadmium cover around the samples and the second with a cadmium cover around the samples. Each set of samples was irradiated for 1 hour. The irradiations were performed near the core of the IEA-R1 4.5 MW nuclear research reactor of the Nuclear and Energy Research Institute – IPEN-CNEN/SP, in São Paulo, Brazil. At the selected irradiation position, the thermal neutron flux was around 2.1×10^{13} $n.cm^{-2}.s^{-1}$.

The activity measurements were carried out in an HPGe gamma-ray spectrometer. The irradiated wires (samples) were positioned within the detector at a distance of about 17.9 cm from the sensitive crystal. In the measurement procedure, the copper samples were encapsulated by an "aluminum sandwich" formed by two aluminum discs with 3 cm diameter and 0.2 cm thick. This was done to prevent the beta minus decay of cooper interfere with the measurement of gamma rays.

Starting 24 hours after the end of irradiation the activity of the samples was measured. The counting times of copper samples with and without cadmium were 10^3 and 1.1×10^4s, respectively. The counting times of gold samples with and without cadmium were 4×10^3 and 10^3 s, respectively.

3. RESULTS AND DISCUSSION

3.1 HPGe efficiency curve

The behavior of the experimental peak efficiency as a function of the gamma-ray energy for the HPGe spectrometer is presented in Fig. 1. In this case, the covered gamma-ray energy range of the IAEA standards was between 121 and 1408 keV. It can be noted a maximum value around 121 keV.

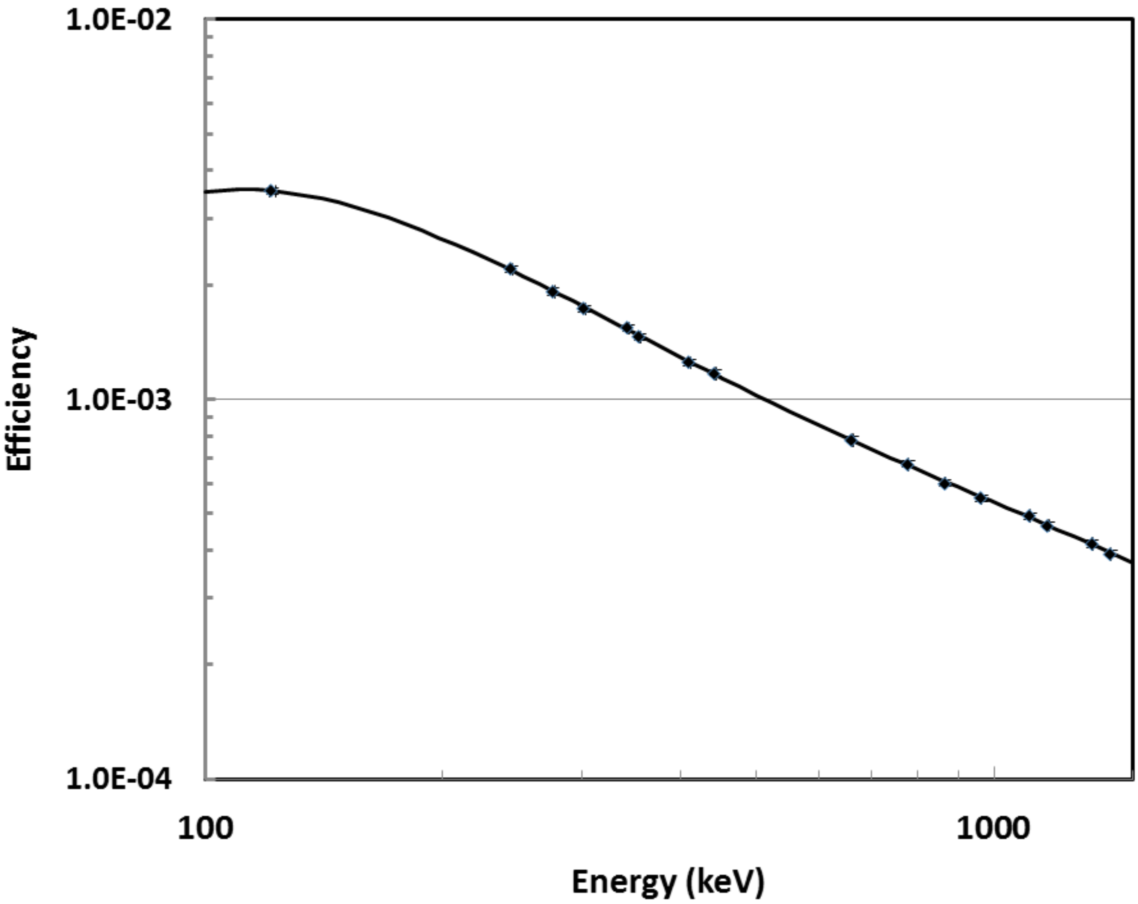

Figure 1: Experimental peak efficiency as a function of the gamma-ray energy. The energy intervals were 121–1408 keV, corresponding to energies of the IAEA standard sources. The solid line corresponds to polynomial fitting in log-log scale.

3.2 F_{Cd} and G_{th}

The cadmium factor F_{Cd} and the thermal neutron self-shielding factor G_{th} were obtained for the targets as shown at Table 1. The number inside brackets corresponds to the uncertainty in the last digits (one standard deviation).

The F_{Cd} and G_{th} were obtained with the Eq. 5 and Eq.6, respectively. Considering the uncertainties in the neutron cross sections, in the Monte Carlo modelling and in the sample thickness, the overall uncertainty was estimated to be around 20% of the correction, in both cases, for F_{Cd} and for G_{th}.

Table 1: Cadmium factor F_{Cd} and thermal neutron self-shielding factor G_{th} for the targets. The number in parenthesis correspond to the uncertainty in the last digits.

Target	F_{Cd}	G_{th}
^{197}Au	0.9999 (1)	1.0000 (0)
^{63}Cu	0.9456(34)	0.9896 (21)

3.3 k_0 for ^{63}Cu(n,γ)^{64}Cu reaction

The k_0 result is presented in Table 2. The number inside brackets corresponds to the uncertainty in the last digits. For ^{63}Cu the k_0 value of experimental result of this present work agrees with De Corte and Simonits (2003) [13].
The uncertainty in k_0 was obtained applying the covariance matrix methodology. This rigorous treatment was used taking into account all partial errors involved and their mutual correlations (Eq.1).

Table 2: Result obtained of k_0 for the ^{63}Cu(n,γ)^{64}Cu reaction. The number in parenthesis correspond to the uncertainty in the last digits.

Target	Product	Energy (keV)	k_0 (Present work)	k_0 Literature [13]
^{63}Cu	^{64}Cu	1345.77	4.982(68)x10^{-04}	4.980(90)x10^{-04}

4. CONCLUSIONS

The present work applied covariance analysis for k_0 measurement. A rigorous treatment was used taking into account all partial errors involved and their mutual correlations. The present result agrees with the literature, within the estimated uncertainties.

ACKNOWLEDGMENTS

The authors acknowledge the help from IEA-R1 reactor staff and the CNEN financial support.

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