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PRELIMINARY MEASUREMENTS OF k_0 VALUES FOR W-186

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ABSTRACT

There are various methods of neutron activation analysis, one of these is the k_0 Method for quantitative reactor Neutron Activation Analysis (NAA). The k_0-NAA procedure is nowadays widely used in numerous laboratories performing NAA all over the world. Among these reactions, $^{186}W(n, \gamma)^{187}W$ can be considered important because it can be used for a W concentration measurements. The irradiations were performed at position 24A, near the core of the IEA-R1 4.5 MW swimming-pool nuclear research reactor of the Instituto de Pesquisas Energéticas e Nucleares (IPEN-CNEN/SP – Nuclear and Energy Research Institute), in São Paulo, Brazil. Two irradiations were carried out in sequence, using two sets of samples: the first with a cadmium cover around the samples and the second without, in a total of three data sets with and without Cd cover performed in 2014 and 2015. The activity measurements were carried out in an HPGe gamma-ray spectrometer. Standard sources of ^{152}Eu, ^{133}Ba, ^{60}Co and ^{137}Cs supplied by the IAEA 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 preliminary values of k_0 for $^{186}W(n, \gamma)^{187}W$ reaction for the gamma transition energy of 479.53 keV was $3.17 \times 10^{-2}(5)$, for 618.77 keV was $9.08 \times 10^{-3}(15)$ and for 685.77 keV was $3.88 \times 10^{-2}(6)$. These preliminary values for k_0 have been compared with the literature.

1. INTRODUCTION

Neutron activation analysis (NAA) has been applied in many fields of science. The NAA is a well-known technique for determining multi-element concentrations in different materials. On k_0 method case, the sample is irradiated together with a comparator (usually Au) and from the ratio between the sample and comparator activities, the element concentrations can be derived [1].

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 [2]. This constant k_0 is independent of irradiation and measuring conditions. In order to achieve good results, there is a continuing need for improving the accuracy of k_0 parameters for several neutron capture reactions [1].

Interest in the subject of nuclear data uncertainties began to emerge in the early of 1970's in response to a need within the reactor physics community for rigorous methods of data manipulation, particularly as applicable to nuclear data evaluation, to the analysis of data

from nuclear benchmark experiments, and to reactor sensitivity studies. From these beginnings, applications of covariance analysis have expanded to other areas of nuclear research [3].

The methodology of covariance analysis is already a well-known statistical procedure and used by internationally renowned institutions [4,5,6] for the analysis of neutron cross section in the thermal and resonance regions, however, this rigorous statistical method was not used to treat the uncertainties of k_0 for the reaction $^{186}W(n, \gamma)^{187}W$. In fact, there are only so few works in literature using the methodology of covariance analysis in order to determinate the uncertainties of k_0 [7,8].

The paper aims to give new insights into determination of k_0 using the methodology of covariance analysis to treat the uncertainties of k_0 for the reaction $^{186}W(n, \gamma)^{187}W$.

2. MATERIALS AND METHODS

2.1. k_0 equation

The parameter k_0 can be obtained by the following relationship [9]:

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

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 [7,15]. All these parameters were included in the covariance analysis. The parameter with largest contribution to the overall uncertainty was the peak efficiency obtained by least squares fitting with standard sources.

The thermal neutron self-shielding factor was evaluated by MATSSF code, a program recommended by IAEA (International Atomic Energy Agency) [10], and the cadmium factor was calculated by the average transmission in the cadmium cover, applying cross section data from ENDF/B-VII [11], considering variation in the cadmium thickness due to isotropic neutron flux.

The following equation was applied [12]:

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

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 [11]. 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]:

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

2.2 Gamma-ray detection efficiency curve

The peak efficiency $\varepsilon_p(E)$ [13] 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. 6:

$$\varepsilon_p(E) = \frac{S_p(E)}{I_\gamma\, At}.f_c \quad (6)$$

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 [3]:

$$Y = Y(a_1, a_2, a_3, ..., a_n) \tag{7}$$

The variance of *Y* is given by:

$$\sigma_Y^2 \cong \sum_{\nu=1}^{n} \frac{\partial Y}{\partial a_\nu} \sum_{\lambda=1}^{n} \frac{\partial Y}{\partial a_\lambda} \langle (a_1 - a_{0,\nu})(a_\lambda - a_{0,\lambda}) \rangle \tag{8}$$

The partial derivatives in Eq.8 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:

$$\mathrm{cov}(a_\nu, a_\lambda) = \langle (a_\nu - a_{0,\nu})(a_\lambda - a_{0,\lambda}) \rangle = \sum_{k=1}^{m} \rho_{\nu,\lambda,k} \sigma_{\nu,k} \sigma_{\lambda,k} \tag{9}$$

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

When a_ν is independent of a_λ the covariance is zero.

2.4. Weighted average of k_0

The preliminary result for k_0 was determined from the weighted average of the values obtained in the irradiations carried out in 2014 and 2015 by the Eq.10. The uncertainty of the preliminary k_0 was determined by Eq. 11.

$$\overline{k_0} = \frac{\left(\frac{k_{0_1}}{\sigma_1^2} + \frac{k_{0_2}}{\sigma_2^2} \right)}{\left(\frac{1}{\sigma_1^2} + \frac{1}{\sigma_2^2} \right)} \tag{10}$$

$$\sigma_{\overline{k_0}} = \sqrt{\frac{1}{\left(\frac{1}{\sigma_1^2} + \frac{1}{\sigma_2^2} \right)}} \tag{11}$$

Where k_{0_1} is the value of k_0 obtained in the irradiation carried out in 2014 and σ_1 is its uncertainty and k_{0_2} is the value of k_0 obtained in the irradiation carried out in 2015 and σ_2 is its uncertainty.

2.5. 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, 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 [14], 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 [15].

The selected targets were ^{197}Au (0.10% Al alloy) and W (99.96%), 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 3 (^{186}W) to 10 (^{197}Au (0.10% Al)) 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, , in a total of three data sets with and without Cd cover performed in 2014 and 2015. 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.

Starting 24 hours after the end of irradiation the activity of the samples was measured.

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. A maximum around 121 keV can be noticed.

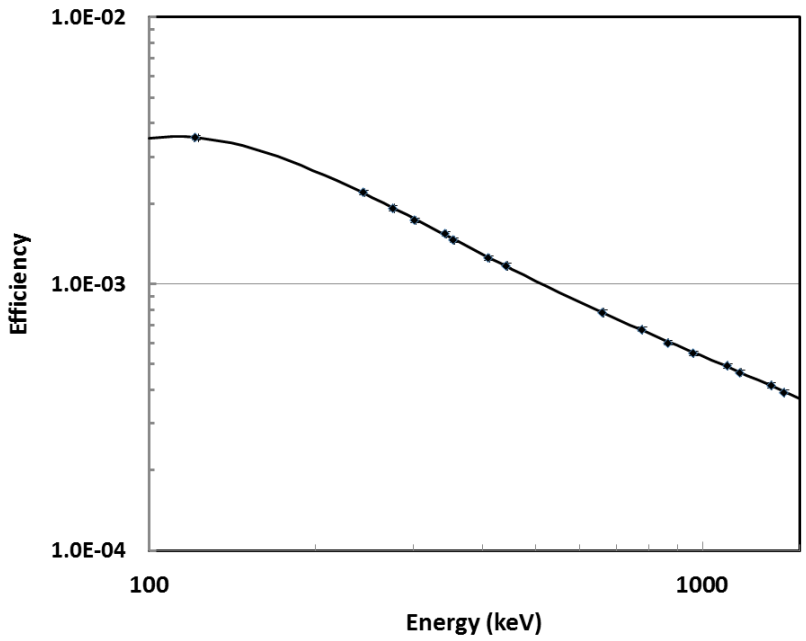

Figure 1: Experimental peak efficiency as a function of the gamma-ray energy. The energy interval was 121–1408 keV, corresponding to the energies of the IAEA standard sources. The solid line corresponds to polynomial fitting in log-log scale.

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} was obtained with the Eq. 5 and G_{th} was evaluated by MATSSF code [10]. 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 corresponds to the uncertainty in the last digits.

Target	F_{Cd}	G_{th}
^{197}Au	0.9999 (1)	1.0000 (0)
^{186}W	0.9919(11)	0.9589(82)

3.3 k_0 for ^{186}W(n, γ)^{187}W reaction

The k_0 results obtained in 2014 and 2015 are presented in Table 2 and 3, respectively. The number inside brackets corresponds to the uncertainty in the last digits.

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: Results of k_0 for the $^{186}W(n, \gamma)^{187}W$ reaction obtained in 2014. The number in parenthesis corresponds to the uncertainty in the last digits.

Target	Product	Energy (keV)	k_0 (Present work)	k_0 Literature [13]
^{186}W	^{187}W	479.53	$3.11(8)\times10^{-2}$	$2.97(3)\times10^{-2}$
		618.77	$8.77(21)\times10^{-3}$	$8.65(4)\times10^{-3}$
		685.77	$3.73(8)\times10^{-2}$	$3.71(2)\times10^{-2}$

Table 3: Results of k_0 for the $^{186}W(n, \gamma)^{187}W$ reaction obtained in 2015. The number in parenthesis corresponds to the uncertainty in the last digits.

Target	Product	Energy (keV)	k_0 (Present work)	k_0 Literature [13]
^{186}W	^{187}W	479.53	$3.23(7)\times10^{-2}$	$2.97(3)\times10^{-2}$
		618.77	$9.16(20)\times10^{-3}$	$8.65(4)\times10^{-3}$
		685.77	$3.94(8)\times10^{-2}$	$3.71(2)\times10^{-2}$

The preliminary result of k_0 to the ^{186}W and its uncertainty at the energies of 479.53; 618.77 and 685.77 (Table 4) were determined according Eq.10 and 11, respectively, from the values obtained in the irradiations carried out in 2014 and 2015 (Table 2 and 3).

Table 4: Preliminary results of k_0 for the $^{186}W(n, \gamma)^{187}W$. The number in parenthesis corresponds to the uncertainty in the last digits.

Target	Product	Energy (keV)	k_0 (Present work)	k_0 Literature [13]
^{186}W	^{187}W	479.53	$3.17(5)\times10^{-2}$	$2.97(3)\times10^{-2}$
		618.77	$9.08(15)\times10^{-3}$	$8.65(4)\times10^{-3}$
		685.77	$3.88(6)\times10^{-2}$	$3.71(2)\times10^{-2}$

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 preliminary result of k_0 to the ^{186}W and its uncertainty at the energies of 479.53; 618.77 and

685.77 were determined. The present preliminary results do not agree with the literature, within the estimated uncertainties. The causes of the differences are being investigated.

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REFERENCES

1. IAEA-TECDOC-1215, *Use of research reactors for neutron activation analysis.* Report of an Advisory Group meeting held in Vienna, 22–26 June (1998).
2. F. de Corte, "The Standardization of Standardless NAA," *Journal of Radioanalytical and Nuclear Chemistry*, **Vol. 248**, pp.13-20 (2001).
3. D. L. Smith, *Probability, statistics and data uncertainties in Nuclear Science and Technology.* Series: Neutron Physics and Nuclear Data in Science and Technology, American Nuclear Society (1991).
4. M. B. Chadwick *et al.*, "ENDF/B-VII.1 nuclear data for science and technology: Cross sections, covariances, fission product yields and decay data," *Nucl. Data Sheets*, **Vol. 112**, no.12, pp. 2887-2996 (2011).
5. P. Robouch, G. Arana, M. Eguskiza, and N. Etxebarria, "Uncertainty Budget for k0-NAA," *J. Radioanal. Nucl. Chem.*, **Vol. 245**, no. 1, pp. 195–197 (2000).
6. T. Nakagawa *et al.*, "Japanese Evaluated Nuclear Data Library Version 3 Revision-2: JENDL-3.2," *J. Nucl. Sci. Technol.*, **Vol. 32**, no. 12, pp. 1259–1271 (1995).
7. M. S. Dias *et al.*, "Measurements of k_0 and Q_0 values for $^{64}Zn(n,\gamma)^{65}Zn$ and $^{68}Zn(n,\gamma)^{69m}Zn$ reactions with covariance analysis," *Appl. Radiat. Isot.*, **Vol. 69**, no. 7, pp. 960–964 (2011).
8. L. F. Barros *et al.*, "Determination of k_0 *for* $^{63}Cu(n,\gamma)^{64}Cu$ reaction with covariance analysis," *Proceeding of INAC 2015- International Nuclear Atlantic Conference*, São Paulo, SP, Brazil, October 4-9, ISBN: 978-85-99141-06-9 (2015).
9. F. de Corte, *The k0-Standardization Method. A Move to the Optimisation of Neutron Activation Analysis.* Ryksuniversiteit Gent, Faculteit Van de Wetenschappen. 464 p. (1986).
10. "MATSSF Program - Details and Instructions," https://www-nds.iaea.org/naa/matssf/ (2016).
11. ENDF/B-VII. Cross Section Library, http://www.nndc.bnl.gov (2010).
12. I. Kodeli, A. Trkov, "Validation of the IRDF-2002 dosimetry library." *Nucl. Instrum. Methods Phys. Res. A*, **Vol. 577**, pp. 664–681 (2007).
13. G. F. Knoll, *Radiation Detection and Measurement*. 4th Edition, John Wiley & Sons (2010).
14. M.S. Dias. LINFIT: a code for linear least square fit with covariance analysis. Internal Report, IPEN-CNEN/SP (1999).
15. M. S. Dias, V. Cardoso, M. F. Koskinas, I. M.Yamazaki, "Determination of the neutron spectrum shape parameter α in k_0 NAA methodology using covariance analysis." *Appl. Radiat. Isot.*, **Vol. 68**, pp. 592 (2010).

16. F. de Corte, A. Simonits, "Recommended nuclear data for use in the k_0 standardization of neutron activation analysis." *At. Data Nucl. Data Tables,* **Vol. 85**, pp. 47–67 (2003).