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Method for Analysis of Neutron Activation Measurements of Am-241 with Uncertainty Propagation

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ABSTRACT

With the aim of estimating thermal neutron capture cross section in ^{241}Am, samples containing ^{241}Am were irradiated in the central channel of the TRIGA reactor at the Jožef Stefan Institute (JSI), both with and without cadmium transmission filter. The neutron fluence was monitored by ^{59}Co(n,γ), ^{197}Au(n,γ) and ^{58}Ni(n,p) reactions. α-particle activity of ^{242}Cm, the decay product of the activation product ^{242g}Am, was measured alongside with γ-ray activities of activation products of the monitor reactions. Reaction rates were calculated from the measured detector count rates, using full uncertainty propagation. The final uncertainties are $\sim 1\%$. Due to use of two independent measurement techniques, the final reaction rate covariance matrix can be divided into two separate blocks, and the correlation coefficients assume values 0.4-0.8.

1 INTRODUCTION

^{241}Am is an important component of spent nuclear fuel due to its contribution to the decay heat. Due to its inherent γ-ray emission, time-of-flight (TOF) measurements of ^{241}Am capture cross section are difficult. Therefore, there is a potential to obtain a more accurate ^{241}Am thermal neutron capture cross section value using neutron activation measurements. This value may serve for normalisation of the energy dependent capture yields measured by TOF. Neutron activation analysis of ^{241}Am is comparatively complex. First, ^{241}Am cross section contains a resonance below Cd transmission filter cut-off energy ($\sim$ 0.55 eV) and another one overlapping with that energy. Second, the activation product is produced in both ground (^{242g}Am) and metastable (^{242m}Am) state, and the latter has a much longer half-life (141 years for ^{242m}Am vs. $\sim$ 16 h for ^{242g}Am). And finally, the decay scheme of activation products is relatively

complicated. γ-ray spectrometry of activation products is difficult due to low γ-ray energies and increased γ-ray background from e.g. ^{241}Am fission products.

Therefore, the only realistic option is α-particle spectrometry, originating from ^{242}Cm, which is a decay product of ^{242g,m}Am. Due to the much longer half-life of ^{242m}Am and a relatively small branching fraction for its production by neutron capture (~ 0.09), its contribution to the ^{242}Cm activity is very low in absolute terms and negligible compared to the contribution from ^{242g}Am for a few years after irradiation.

An experiment was performed at the JSI TRIGA reactor, where samples of ^{241}Am deposited from solution were irradiated with the ultimate goal to improve the ^{241}Am(n,γ)^{242g}Am thermal neutron cross section value, which is burdened with a relatively high combined weighted average uncertainty of $\sim 2\%$ [1, 2]. ^{242g}Am decay with half-life of 16.02(2) h [3] with probability (branching fraction) $82.7(3)\,\%$ [3] ($83.1\,\%$ according to DDEP [4]) to ^{242}Cm. The latter is a practically $100\,\%$ α-particle emitter with a half-life of 162.8(2) d [5].

For more accurate determination of the neutron fluence, ^{59}Co(n,γ), ^{197}Au(n,γ) and ^{58}Ni(n,p) reactions were chosen as monitor materials. Each measurement set consists of two irradiations, one with and one without a cadmium transmission filter with thickness 1.10 mm ± 0.05 mm. The activities of samples of monitor materials are measured by a γ-ray spectrometer.

The first step is to determine the specific reaction rates starting from the measured count rates from the α-particle spectrometer for ^{242}Cm and γ-ray spectrometer from ^{60}Co, ^{58}Ni and ^{198}Au. The specific ^{241}Am(n,γ)^{242g}Am reaction rate will be determined as a ratio to ^{241}Am activity, determined from the same α-particle spectrometric measurement as the ^{242}Cm activity. For other reactions, the specific reaction rates will be determined taking into account the nominal masses of the irradiated samples.

2 NEUTRON IRRADIATIONS AND ACTIVATION MEASUREMENTS

Irradiations were performed in the central irradiation channel (CC) of the JSI TRIGA reactor. Two sequential measurements were performed. The properties of the irradiated samples, irradiation and measurement conditions are given in Tables 1 and 2. During both irradiations, the core was in configuration No. 242.

Table 1: Irradiation conditions and properties of the samples. C_α represents the total α-particle count rate within measurement time $t_{m,\alpha}$, whereas t_{irr} is the irradiation time at constant power. The values in round brackets represent the absolute uncentainties, corresponding to the last digit(s).

Irradiation	1 (Cd cover)	2 (no Cd)
Cd cover	yes	no
t_{irr}	36000(1) s	36000(1) s
$C_\gamma(^{241}\text{Am})$	102409(699)	249883(632)
$t_{m,\gamma}(^{241}\text{Am})$	527735(1) s	768468(1) s
$C_\alpha(^{241}\text{Am})$	3342055(1828)	1038473(1019)
$t_{m,\alpha}(^{241}\text{Am})$	344003(1) s	177615(1) s
$m(^{59}\text{Co})$	4.364(52) μg	4.725(57) μg
$m(^{58}\text{Ni})$	178.2(2) mg	192.6(11) mg
$m(^{197}\text{Au})$	5.787(35) μg	5.747(34) μg

The γ-ray detector efficiencies η were determined by means of the MMCNP-6.1.1 [6] and a computational model of the source-detector configuration using electron/photon/relaxation

Table 2: Parameters of the spectrometric measurements, including cooling times t_c between end of irradiation and start of measurement.

Irradiation	E_γ	1 (Cd cover)	2 (no Cd)
Cd cover		yes	no
t_{irr}		36000(1) s	36000(1) s
t_c(Am)		418595(1) s	1015660(1) s
t_m(Am)		344003(1) s	177615(1) s
$C(^{242}\text{Cm})$		61224(247)	120057(346)
η(Cm)		$1.433(27) \times 10^{-3}$	
t_c(Co)		782393(1) s	426439(1) s
t_m(Co)		179680(1) s	243805(1) s
$C(^{60}\text{Co})$	1173 keV	34164(222)	255213(703)
	1332 keV	31775(204)	238274(1016)
t_c(Au)		758557(1) s	672264(1) s
t_m(Au)		10800(1) s	10800(1) s
$C(^{198}\text{Au})$	411.8 keV	2136909(5193)	4367877(2125)
t_c(Ni)		577917(1) s	1099245(1) s
t_m(Ni)		312037(1) s	10800(1) s
$C(^{58}\text{Co})$	810.8 keV	70638964(8457)	2652964(1637)

data library eprdata12 [7]. The corresponding covariances were estimated by variation of key geometrical parameters within their uncertainties (a detailed study is available in Ref. [8]) and are given in Table 3. The main sources of uncertainty were the detector dead layer, source to detector distance and a fitted normalisation constant. Uncertainties due to photo-atomic nuclear data and detector response function were considered negligible.

Table 3: γ-ray detector efficiencies at energies 411.8 keV, 810.8 keV, 1173 keV and 1332 keV and components of the corresponding covariance matrix.

E_γ	η	411.8 keV	810.8 keV	1173 keV	1332 keV
411.8 keV	1.514×10^{-3}	1.43×10^{-10}	7.51×10^{-11}	5.83×10^{-11}	5.30×10^{-11}
810.8 keV	9.446×10^{-4}	7.51×10^{-11}	5.01×10^{-11}	3.49×10^{-11}	3.16×10^{-11}
1173 keV	7.392×10^{-4}	5.83×10^{-11}	3.49×10^{-11}	3.06×10^{-11}	2.79×10^{-11}
1332 keV	6.775×10^{-4}	5.30×10^{-11}	2.79×10^{-11}	2.79×10^{-11}	2.59×10^{-11}

3 CORRESPONDENCE BETWEEN REACTION RATES AND COUNT RATES

3.1 ^{241}Am(n,γ)^{242g}Am

Neglecting the contribution from ^{242m}Am, the following is valid during irradiation:

$$\frac{\mathrm{d}N_{Amg}}{\mathrm{d}t} = R_{Am,g} - \lambda_{Amg} N_{Amg}, \qquad \frac{\mathrm{d}N_{Cm}}{\mathrm{d}t} = k_{\beta^{(-)}} \lambda_{Amg} N_{Amg} - \lambda_{Cm} N_{Cm}, \tag{1}$$

where the indices Amg and Cm refer to ^{242g}Am and ^{242}Cm, respectively, $R_{Am,g}$ is the ^{241}Am(n,γ)^{242g}Am reaction rate and $k_{\beta^{(-)}}$ is the branching fraction for β^- decay of ^{242}Cm.

Assuming constant $R_{Am,g}$ and zero initial condition for both nuclides, after irradiation time t_{irr} and cooling time t_c, the expected number of α-particle detector counts C_{Cm}, originating from ^{242}Cm decay, within a measurement time t_m equals:

$$C_{Cm} = \frac{\eta_{Cm} k_{\beta^{(-)}} R_{Am,g}}{\lambda_{Amg} - \lambda_{Cm}} \times \left[\frac{\lambda_{Amg}}{\lambda_{Cm}} \left(1 - \mathrm{e}^{-\lambda_{Cm} t_{irr}}\right) \mathrm{e}^{-\lambda_{Cm} t} \left(1 - \mathrm{e}^{-\lambda_{Cm} t_m}\right) + \frac{\lambda_{Cm}}{\lambda_{Amg}} \left(1 - \mathrm{e}^{-\lambda_{Amg} t_{irr}}\right) \mathrm{e}^{-\lambda_{Amg} t} \left(1 - \mathrm{e}^{-\lambda_{Amg} t_m}\right) \right], \tag{2}$$

where η_{Cm} is the detection efficiency for the α-particles originating from ^{242}Cm decay and α-particle emission probability of 1 was assumed for the ^{242}Cm decay. Eq. (2) represents a general solution for the count rate of ^{242}Cm decays, assuming a constant ^{241}Am(n,γ)^{242g}Am reaction rate and cooling time short compared to ^{242m}Am half-life.

Assuming that the cooling time is long compared to ^{242g}Am half-life (i.e. $t_c \gg t_{1/2}(^{242g}\mathrm{Am})$):

$$C_{Cm} \simeq \frac{\eta_{Cm} k_{\beta^{(-)}} R_{Am,g}}{\lambda_{Amg} - \lambda_{Cm}} \frac{\lambda_{Amg}}{\lambda_{Cm}} \left(1 - \mathrm{e}^{-\lambda_{Cm} t_{irr}}\right) \mathrm{e}^{-\lambda_{Cm} t_c} \left(1 - \mathrm{e}^{-\lambda_{Cm} t_m}\right). \tag{3}$$

The number of detected α-particles, originating from ^{241}Am decay, equals:

$$C_{Am241} = N_{Am241} \lambda_{Am241} t_m, \tag{4}$$

assuming that the irradiation, cooling and measurement times are all small compared to the half-life of ^{241}Am (432.7 years), and negligible depletion of ^{241}Am during irradiation.

Finally, the specific ^{241}Am(n,γ)^{242g}Am reaction rate can be expressed as:

$$r_g \simeq \frac{C_{Cm}}{C_{Am241}} \frac{\lambda_{Am241} \lambda_{Cm} (\lambda_{Amg} - \lambda_{Cm})}{\lambda_{Amg}} \frac{\eta_{Am}}{\eta_{Cm}} \frac{t_m}{k_{\beta^{(-)}}} \left[\left(1 - \mathrm{e}^{-\lambda_{Cm} t_{irr}}\right) \mathrm{e}^{-\lambda_{Cm} t_c} \left(1 - \mathrm{e}^{-\lambda_{Cm} t_m}\right) \right]^{-1}. \tag{5}$$

3.2 ^{59}Co(n,γ)^{60}Co

Assuming constant ^{59}Co(n,γ)^{60}Co reaction rate R_{Co} during irradiation, a short irradiation time in comparison with ^{60}Co half-life, no ^{60}Co removal due to neutron capture, and a comparatively short measurement time t_m:

$$C_{Co,j} \simeq \eta_{Co,j} P_{\gamma,Co,j} \lambda_{Co} R_{Co} t_{irr} t_m \mathrm{e}^{-\lambda_{Co} t_c}, \tag{6}$$

where the index j refers to γ-rays of a characteristic energy, $\eta_{Co,j}$ is the γ-ray detector efficiency at the corresponding γ-ray energy and $P_{\gamma,Co,j}$ is the emission probability for the corresponding γ-ray energy.

Thus, for each characteristic γ-ray energy, an estimate for the specific ^{59}Co(n,γ)^{60}Co reaction rate is obtained:

$$r_{Co,j} = \frac{R_{Co,j}}{N_{Co59}} = \frac{C_{Co,j}}{\eta_{Co,j} P_{\gamma,Co,j} \lambda_{Co} N_{Co59} t_{irr} t_m} \mathrm{e}^{\lambda_{Co} t_c}, \tag{7}$$

The final estimate for R_{Co} is then a weighted average over all characteristic γ-ray energies. For ^{60}Co, characteristic γ-rays with energies of 1173.228(3) keV and 1332.492(4) keV and emission probabilities of 0.9985(3) and 0.999826(6) [9], respectively, were used.

3.3 ^{197}Au(n,γ)^{198}Au

Assuming constant ^{197}Au(n,γ)^{198}Au reaction rate R_{Au} during irradiation, no ^{198}Au removal due to neutron capture, and a comparatively short measurement time t_m:

$$C_{Au,j} \simeq \eta_{Au,j} P_{\gamma,Au,j} R_{Au} t_m \left(1 - \mathrm{e}^{-\lambda_{Au} t_{irr}}\right) \mathrm{e}^{-\lambda_{Au} t_c}. \tag{8}$$

Thus, for each characteristic γ-ray energy, an estimate for the specific ^{197}Au(n,γ)^{198}Au reaction rate is obtained:

$$r_{Au,j} = \frac{R_{Au,j}}{N_{Au197}} = \frac{C_{Au,j}}{\eta_{Au,j} P_{\gamma,Au,j} N_{Au197} t_m} \mathrm{e}^{\lambda_{Au} t_c} \left(1 - \mathrm{e}^{-\lambda_{Au} t_{irr}}\right)^{-1}, \tag{9}$$

The final estimate for R_{Au} is then a weighted average over all characteristic γ-ray energies. For ^{198}Au, the characteristic γ-ray with energy of 411.80205(17) keV and emission probability of 0.9562 [10] were used.

3.4 ^{58}Ni(n,p)^{58}Co

Assuming constant ^{58}Ni(n,p)^{58}Co reaction rate R_{Ni} during irradiation, no ^{58}Co removal due to neutron capture, and a comparatively short measurement time t_m:

$$C_{Cop,j} \simeq \eta_{Cop,j} P_{\gamma,Cop,j} R_{Ni} t_m \left(1 - \mathrm{e}^{-\lambda_{Cop} t_{irr}}\right) \mathrm{e}^{-\lambda_{Cop} t_c}. \tag{10}$$

Thus, for each characteristic γ-ray energy, an estimate for the specific ^{58}Ni(n,p)^{58}Co reaction rate is obtained:

$$r_{Ni,j} = \frac{R_{Ni,j}}{N_{Ni58}} = \frac{C_{Cop,j}}{\eta_{Cop,j} P_{\gamma,Cop,j} N_{Ni58} t_m} \mathrm{e}^{\lambda_{Cop} t_c} \left(1 - \mathrm{e}^{-\lambda_{Cop} t_{irr}}\right)^{-1}, \tag{11}$$

The final estimate for R_{Ni} is then a weighted average over all characteristic γ-ray energies. For ^{58}Co, the characteristic γ-ray with energy of 810.7593(20) keV and emission probability of 0.99450 [11] were used.

4 UNCERTAINTY PROPAGATION

For uncertainty propagation it is important to note that the components of uncertainty, common to different specific reaction rates, are: detector efficiencies η (for α-particles and γ-ray separately), branching fraction for the decay of ^{242g}Am, (some) γ-ray emission probabilites.

Other sources of uncertainty, such as e.g. the detector count rates, samples masses and activities, are in principle independent for each measurement and can be treated as such.

Some sources of uncertainty may be neglected: all decay constants, all times (irradiation, cooling, measurement), (some) γ-ray emission probabilites, ^{242}Cm α-particle emission probabilities with energies above the highest ^{241}Am α-particle emission energy 5545 keV.

4.1 ^{241}Am(n,γ)^{242g}Am

The parameters in Eq. (5) with non-negligible uncertainties are: C_{Cm}, C_{Am241}, η_{Am}, η_{Cm} and $k_{\beta(-)}$. $k_{\beta(-)} = 0.832(6)$ was adopted from the JEFF-3.1.1 [12] nuclear data library. If the ratio of α-particle detection efficiencies is treated as a single parameter $k_\eta = \eta_{Am}/\eta_{Cm} \sim 1$,

the relative uncertainty $\delta r_g/r_g$ of the ^{241}Am(n,γ)^{242g}Am reaction rate can be expressed as sum of squares of the relative uncertainties of the individual components:

$$\frac{\delta r_g}{r_g} = \sqrt{\left(\frac{\delta C_{Cm}}{C_{Cm}}\right)^2 + \left(\frac{\delta C_{Am241}}{C_{Am241}}\right)^2 + \frac{4(\delta T_{Cm})^2}{C_{Cm}C_{Am241}} + (\delta k_\eta)^2 + \left(\frac{\delta k_{\beta(-)}}{k_{\beta(-)}}\right)^2}, \qquad (12)$$

which are assumed to be uncorrelated, except C_{Cm} and C_{Am241}, for which the expression from Ref. [13] was used. Here, δT_{Cm} denotes the uncertainty in the overlap count rate between ^{241}Am and ^{242}Cm ($\delta T_{Cm} = 110$, $\delta T_{Cm,Cd} = 93$).

For separate ^{241}Am irradiations / ^{242}Cm measurements, e.g. with and without Cd cover, the common uncertainty originates from k_η and $k_{\beta(-)}$, whereas the uncertainty originating from the measured count rates is uncorrelated by nature:

$$\frac{\mathrm{cov}(r_g, r_{g,Cd})}{r_g r_{g,Cd}} = \mathrm{cov}\left(\frac{k_\eta}{k_{\beta(-)}}, \frac{k_\eta}{k_{\beta(-)}}\right) \Bigg/ \left(\frac{k_\eta}{k_{\beta(-)}}\right)^2 = (\delta k_\eta)^2 + \left(\frac{\delta k_{\beta(-)}}{k_{\beta(-)}}\right)^2 \qquad (13)$$

4.2 ^{59}Co(n,γ)^{60}Co

The parameters in Eq. (7) with non-negligible uncertainties are: $C_{Co,1}$, $C_{Co,2}$, $\eta_{Co,1}$, $\eta_{Co,2}$ and N_{Co59}. The relative uncertainty $\delta r_{Co,j}/r_{Co,j}$ of the ^{59}Co(n,γ)^{60}Co reaction rate, derived from each of the two prominent γ-ray peaks can be expressed as sum of squares of the relative uncertainties of the individual components:

$$\frac{\delta r_{Co,j}}{r_{Co,j}} = \sqrt{\left(\frac{\delta C_{Co,j}}{C_{Co,j}}\right)^2 + \left(\frac{\delta \eta_{Co,j}}{\eta_{Co,j}}\right)^2 + \left(\frac{\delta N_{Co59}}{N_{Co59}}\right)^2}. \qquad (14)$$

From the count rates, corresponding to both peaks, a weighted averages is calculated:

$$r_{Co} = \left(\frac{r_{Co,1}}{(\delta r_{Co,1})^2} + \frac{r_{Co,2}}{(\delta r_{Co,2})^2}\right)\left(\frac{1}{(\delta r_{Co,1})^2} + \frac{1}{(\delta r_{Co,2})^2}\right)^{-1}. \qquad (15)$$

This weighted average is unbiased and ensures numerical stability, however it does not yield lowest combined uncertainty:

$$\frac{\delta r_{Co}}{r_{Co}} = \sqrt{(\delta r_{Co,1})^2 + (\delta r_{Co,2})^2 + 2\mathrm{cov}(r_{Co,1}, r_{Co,2})} \Bigg/ \left(r_{Co,1}\frac{\delta r_{Co,2}}{\delta r_{Co,1}} + r_{Co,2}\frac{\delta r_{Co,1}}{\delta r_{Co,2}}\right), \qquad (16)$$

$$\mathrm{cov}(r_{Co,1}, r_{Co,2}) = K_{Co}^2 \frac{C_{Co,1}C_{Co,2}}{P_{\gamma,Co,1}P_{\gamma,Co,2}} \frac{\mathrm{cov}(\eta_{Co,1}, \eta_{Co,2})}{\eta_{Co,1}^2 \eta_{Co,2}^2}, \qquad K_{Co} = \frac{\mathrm{e}^{\lambda_{Co} t_c}}{\lambda_{Co} N_{Co59} t_{irr} t_m}.$$

For separate ^{59}Co irradiations / ^{60}Co measurements, e.g. with and without Cd cover, the common uncertainty originates from $\eta_{Co,j}$, whereas the uncertainties originating from the measured count rates and sample ^{59}Co masses are uncorrelated by nature:

$$\frac{\mathrm{cov}(r_{Co}, r_{Co,Cd})}{r_{Co} r_{Co,Cd}} = \qquad (17)$$

$$= \frac{\frac{r_{Co1}}{(\delta r_{Co1})^2}\frac{r_{CoCd1}}{(\delta r_{CoCd1})^2}\left(\frac{\delta \eta_{Co1}}{\eta_{Co1}}\right)^2 + \frac{r_{Co2}}{(\delta r_{Co2})^2}\frac{r_{CoCd2}}{(\delta r_{CoCd2})^2}\left(\frac{\delta \eta_{Co2}}{\eta_{Co2}}\right)^2 + \left[\frac{r_{Co1}}{(\delta r_{Co1})^2}\frac{r_{CoCd2}}{(\delta r_{CoCd2})^2} + \frac{r_{Co2}}{(\delta r_{Co2})^2}\frac{r_{CoCd1}}{(\delta r_{CoCd1})^2}\right]\frac{\mathrm{cov}(\eta_{Co1}, \eta_{Co2})}{\eta_{Co1}\eta_{Co2}}}{\left(\frac{r_{Co1}}{(\delta r_{Co1})^2} + \frac{r_{Co2}}{(\delta r_{Co2})^2}\right)\left(\frac{r_{CoCd1}}{(\delta r_{CoCd1})^2} + \frac{r_{CoCd2}}{(\delta r_{CoCd2})^2}\right)}$$

4.3 ^{197}Au(n,γ)^{198}Au and ^{58}Ni(n,p)^{58}Co

The parameters with non-negligible uncertainties are: C_x, η_x and N_x, where x denotes Au or Ni. The relative uncertainty $\delta r_x/r_x$ can be expressed as sum of squares of the relative uncertainties of the individual components:

$$\frac{\delta r_x}{r_x} = \sqrt{\left(\frac{\delta C_x}{C_x}\right)^2 + \left(\frac{\delta \eta_x}{\eta_x}\right)^2 + \left(\frac{\delta N_x}{N_x}\right)^2}. \tag{18}$$

For separate irradiations of x, e.g. with and without Cd cover, the common uncertainty originates from η_x, whereas the uncertainties originating from the measured count rates and sample masses are uncorrelated by nature:

$$\frac{\mathrm{cov}(r_x, r_{x,Cd})}{r_x r_{x,Cd}} = \left(\frac{\delta \eta_x}{\eta_x}\right)^2. \tag{19}$$

Since ^{58}Ni(n,p) is a threshold reaction, no noticable difference between the measurements with and without Cd cover is expected. These two measurements can serve as an additional neutron fluence monitor, taking into account the correction factor for the Cd transmission function.

4.4 Cross-material covariances

Fhe following cross-terms are valid for all combinations with and without Cd cover:

$$\frac{\mathrm{cov}(r_{Au}, r_{Ni})}{r_{Au} r_{x,Ni}} = \eta_{Au}\eta_{Ni}\mathrm{cov}\left(\frac{1}{\eta_{Au}}, \frac{1}{\eta_{Ni}}\right) = \frac{\mathrm{cov}(\eta_{Au}, \eta_{Ni})}{\eta_{Au}\eta_{Ni}}, \tag{20}$$

$$\frac{\mathrm{cov}(r_{Co}, r_x)}{r_{Co} r_x} = \frac{\frac{r_{Co1}}{(\delta r_{Co1})^2}\frac{\mathrm{cov}(\eta_{Co1}, \eta_x)}{\eta_{Co1}\eta_x} + \frac{r_{Co2}}{(\delta r_{Co2})^2}\frac{\mathrm{cov}(\eta_{Co2}, \eta_x)}{\eta_{Co2}\eta_x}}{\frac{r_{Co1}}{(\delta r_{Co1})^2} + \frac{r_{Co2}}{(\delta r_{Co2})^2}}. \tag{21}$$

4.5 Summary – final reaction rate vector and covariance matrix

The final reaction rate covariance matrix can be divided into two uncorrelated blocks. The first block consists of ^{241}Am(n,γ)^{242g}Am reaction rates (Table 4), and the second block of all other reaction rates (Table 5), which are correlated via the γ-ray detector efficiency.

Table 4: ^{241}Am(n,γ)^{242g}Am reaction rates and corresponding correlation matrix components.

sample	r / s^{-1}	Am	Am(Cd)
Am	$4.185(33) \times 10^{-9}$	1	0.798
Am(Cd)	$6.465(54) \times 10^{-10}$	0.798	1

The relative reaction rates are highest for ^{241}Am(n,γ), then ^{197}Au(n,γ) and ^{59}Co(n,γ), which is in accordance with the neutron spectrum averaged cross sections for these reactions. As expected, the reaction rates under Cd filter are significantly lower for these non-threshold reactions. For ^{58}Ni(n,p), the reaction rates are several orders of magnitude lower due to much lower cross section and contribution from only fast neutrons. Due to the latter, the relative difference between the cases with and without Cd cover is $< 10\%$. The final uncertainties are of the order of 1%, which is within the acceptable limits. The correlations are highest for both measurements with samples containing Am due to the use of the same α-particle spetrometer and nuclear data, whereas the contributions of the independent component, i.e. the detector count rates, to the final uncertainty, is smaller than for other samples.

Table 5: ^{197}Au(n,γ), ^{58}Ni(n,p) and ^{59}Co(n,γ) reaction rates and corresponding correlation matrix components.

sample	r / s^{-1}	Au	Au(Cd)	Ni	Ni(Cd)	Co	Co(Cd)
Au	$1.163(11) \times 10^{-9}$	1	0.616	0.563	0.701	0.450	0.430
Au(Cd)	$7.306(75) \times 10^{-10}$	0.616	1	0.543	0.677	0.434	0.415
Ni	$3.638(34) \times 10^{-14}$	0.563	0.543	1	0.785	0.451	0.431
Ni(Cd)	$3.416(26) \times 10^{-14}$	0.701	0.677	0.785	1	0.562	0.537
Co	$1.944(23) \times 10^{-10}$	0.450	0.434	0.451	0.562	1	0.393
Co(Cd)	$3.823(47) \times 10^{-11}$	0.430	0.415	0.431	0.537	0.393	1

5 CONCLUSIONS

Reaction rates were calculated starting from the measured detector count rates, using full uncertainty propagation. Notable uncertainty components include, sample masses, branching fraction for β^- decay of ^{242g}Am and detector efficiencies. The latter two induce correlations between samples, whereas other uncertainty components can be treated as independent. The final uncertainties range from 0.8% to 1.2%. The final reaction rate covariance matrix can be divided into two separate blocks, one for samples containing americium, and one for all other samples. The correlation coefficients within a single block typically range between 0.4 and 0.8, which indicates that the detector efficiency and branching fraction for β^- decay of ^{242g}Am are dominant sources of uncertainty, and that the reaction rates cannot be treated separately.

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