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Results of transmission measurements for ^{40}Ca at GELINA

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Abstract
Transmission measurements have been performed at the neutron time-of-flight facility GELINA to determine the total cross section for neutron induced reactions on ^{40}Ca. The experimental details, i.e. measurement conditions, sample characteristics, measurement procedures and experimental uncertainty components, together with the data reduction procedures are described. The experimental results including the full covariance information, based on the AGS-formalism, are reported following the latest recommendations of the International Nuclear Data Committee. This includes the information that is required to derive nuclear reaction model parameters together with their covariances by a least squares adjustment to experimental data.

RESULTS OF TRANSMISSION MEASUREMENTS

FOR ^{40}CA AT GELINA

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Executive summary

Transmission measurements have been performed at a 50 transmission station of the neutron time-of-flight facility GELINA to determine the total cross section for neutron induced reactions on ^{40}Ca. These measurements are part of a collaboration of the Joint Research Centre and the Oak Ridge National Laboratory (US) to improve nuclear data for nuclear criticality safety applications. They have been supported by the EUFRAT project.

This report provides the experimental details required to submit the data to the EXFOR data library which is maintained by the Nuclear Data Section of the IAEA and the Nuclear Energy Agency of the OECD. The experimental details, i.e. measurement conditions, sample characteristics, measurement procedures and experimental uncertainty components, together with the data reduction procedures are described. The experimental results including the full covariance information, based on the AGS-formalism, are reported following the latest recommendations of the International Nuclear Data Committee. This includes the information that is required to derive nuclear reaction model parameters together with their covariances by a least squares adjustment to experimental data.

Contents

1. Introduction

To study the resonance structure of neutron induced reaction cross sections, neutron spectroscopic measurements are required which determine with a high accuracy the reaction cross sections and energy of the neutron that interacts with the material under investigation. To cover a broad energy range such measurements are best carried out with a pulsed white neutron source, which is optimized for time-of-flight (TOF) measurements [1].

The TOF facility GELINA [2] has been designed and built for high-resolution cross section measurements in the resolved (RRR) and unresolved (URR) resonance region. It is a multi-user TOF facility, providing a white neutron source with a neutron energy range from 10 meV to 20 MeV. Up to 10 experiments can be performed simultaneously at measurement stations located between 10 m to 400 m from the neutron production target. The electron linear accelerator provides a pulsed electron beam with a maximum energy of 150 MeV, an average current of about 55 μA and a repetition rate ranging from 50 Hz to 800 Hz. A compression magnet reduces the width of the electron pulses to less than 2 ns [3]. The electron beam hits a mercury-cooled uranium target producing Bremsstrahlung and subsequently neutrons via photonuclear reactions [4]. Two water-filled beryllium containers mounted above and below the neutron production target are used to moderate the neutrons. By applying different neutron beam collimation conditions, experiments can use either a fast or a thermalized neutron spectrum. The neutron production rate is constantly monitored by BF3 proportional counters which are mounted in the ceiling of the target hall. The output of the monitors is used to normalize the time-of-flight spectra to the same neutron intensity. The measurement stations are equipped with air conditioning to reduce electronic drifts in the detection chains due to temperature changes.

In this report results of transmission measurements carried out at GELINA with a natural calcium sample are described. To reduce bias effects due to e.g. dead time and background, the measurement and data reduction procedures recommended in Ref. [1] have been followed. The main objective of this report is to provide the information that is required to evaluate the total cross section for ^{40}Ca in the resonance region and to extract nuclear reaction model parameters in a least squares adjustment to the data [1]. In the description of the data the recommendations of the International Nuclear Data Committee (INDC), resulting from a consultant's meeting organized by the Nuclear Data Section of the IAEA, are followed [5].

These measurements are part of a collaboration between the Joint Research Centre and the Oak Ridge National Laboratory (US) to improve nuclear data evaluations for neutron induced reactions that are important for nuclear criticality safety. Calcium is a concrete constituent and it is very frequently found in combination with uranium, either for the construction of nuclear power plants or in storage facilities for nuclear waste. Liquid radioactive waste is often solidified by mixing it with concrete. Calcium has strong neutron-absorbing properties that can affect the reactivity of systems with fissionable materials. The evaluated data for calcium found in the nuclear data libraries do not perform well in criticality calculations. In addition, a consistent set of covariance data is needed in support of sensitivity and uncertainty analyses.

2. Experimental conditions

The transmission experiments were performed at the 50 m measurement station of flight path 4 with the accelerator operating at 800 Hz. The flight path forms an angle of 9^{o} with the direction normal to the facet of the moderator viewing the flight path. The moderated neutron spectrum was used. A shadow bar made of Cu and Pb was placed close to the uranium target to reduce the intensity of the γ-ray flash and the fast neutron component. The sample and detector were placed in a acclimatised room to keep them at a constant temperature of 22 ^{o}C. A schematic view of the experimental set-up is shown in Fig. 1. The experimental conditions for each experiment, including the monitor counts and type of background and overlap filters used in the experiment, are specified in Appendix A based on a template provided in Ref. [5].

The neutrons scattered from the moderators are collimated into evacuated pipes of 50 cm diameter with annular collimators. A combination of Li-carbonate plus resin, Pb and Cu-collimators was used to reduce the neutron beam to a diameter of about 45 mm at the sample position. The sample was placed in an automatic sample changer at a distance of approximately 24 m from the neutron source. A ^{10}B overlap

filter, with an areal density of 8 10^{-3} at/b, was placed to absorb slow neutrons from previous bursts. The impact of the γ-ray flash was reduced by a 16 mm thick Pb filter. A set of Na, Co and W black resonance filters were mounted in an automatic filter changer close to the sample position to determine the background with the black resonance technique [1].

The neutron beam passing through the sample and filters was further collimated and detected by a 6.35 mm thick and 152.4 mm diameter NE912 Li-glass scintillator. The scintillator was connected through a boron-free quartz window to a 127 mm EMI 9823 KQB photomultiplier (PMT), which was placed outside the neutron beam and perpendicularly to its axis. The detector was placed at a distance of 49.34 m from the facet of the moderator viewing the flight path. The diameter of the neutron beam at the detector position was about 85 mm.

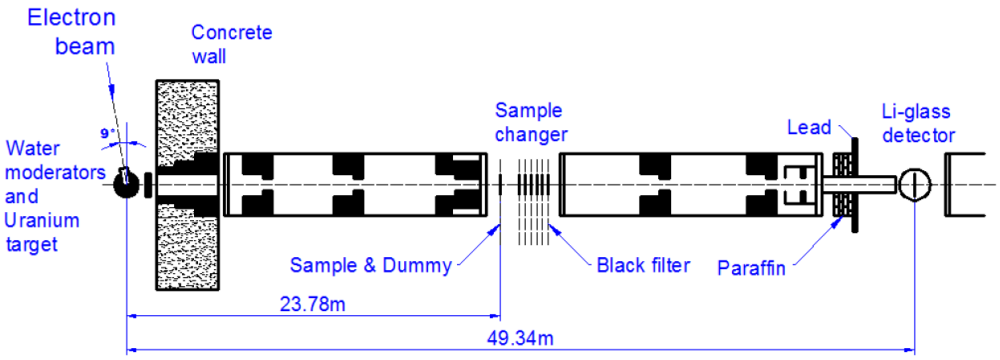

Figure 1 Schematic representation of the transmission set-up at the 50 m transmission station of GELINA.

The output signals of the PMT were connected to conventional analog electronics. The anode pulse of the PMT was fed into a constant fraction discriminator to create a fast logic signal which defines the time the neutron has been detected. The signal of the 9th dynode was shaped by a spectroscopic amplifier to determine the energy deposited by the ^{6}Li(n,t)α reaction in the detector. A module was included to produce a fixed dead time in the whole electronics chain directly after the detection of an event. This dead time t_d = 3305 ns was continuously monitored by recording the time interval between successive pulses. The time-of-flight (TOF) of the detected neutron was determined by the time difference between the stop signal (T_s) derived from the anode pulse of the PMT and the start signal (T_0), given at each electron burst. This time difference was measured with a multi-hit fast time coder with a 1 ns time resolution. The TOF and pulse height of a detected event were recorded in list mode data using a multi-parameter data acquisition system developed at the EC-JRC-IRMM [6]. Each measurement was subdivided in different cycles of about 900'' each.

Table 1. Characteristics of the samples used for the transmission measurements performed at GELINA. The uncertainties are standard uncertainties at 1 standard deviation. To calculate the areal density the Avogadro constant was taken as N_A = 6.0221367 10^{23} mol^{-1} and the atomic mass for ^{nat}Ca as m_a = 40.0780 g. The uncertainty on the areal density is dominated by the 0.2 % uncertainty on the area, which includes an uncertainty due to non-perfect circular shape of the samples.

Sample ID	Thickness	Diameter	Mass	Areal density
1	12.55 mm	60.00 mm	54.438 g	2.893 (0.006) 10^{-2} at/b
2	15.35 mm	59.95 mm	66.212 g	3.525 (0.007) 10^{-2} at/b
3	20.22 mm	59.98 mm	85.627g	4.554 (0.009) 10^{-2} at/b

To avoid background originating from oxygen and carbon in calcium carbonate samples, metallic calcium samples were chosen. Three metallic Ca discs were used to produce a sample with a thickness of ~ 5 cm and a total combined mass of 206.277 g. The characteristics of the discs are given in Table 1. They were encapsulated in a thin-walled aluminium container to prevent reactions with air. To compensate for the effect of the metallic can, data were taken with an Al empty container with similar characteristics as the sample container.

3. Data reduction

The experimental transmission T_{exp} as a function of the time-of-flight, denoted by t_m, was obtained from the ratio of the counts of a sample-in measurement C_{in} and a sample-out measurement C_{out}, after subtraction of the background contributions B_{in} and B_{out}, respectively [1]:

$$T_{exp}(t_m) = N\frac{C_{in}(t_m) - K\,B_{in}(t_m)}{C_{out}(t_m) - K\,B_{out}(t_m)} \quad , \tag{1}$$

where t_m denotes the measured time-of-flight. The TOF-spectra (C_{in}, B_{in}, C_{out}, B_{out}) in Eq. 1 were corrected for losses due to the dead time in the detector and the electronics chain. The factor K is introduced to account for the uncertainty due to the background.

The dead time correction was based on the formula of Moore [7]. This formula accounts for possible variations in the beam intensity. The dead time correction factors as a function of time-of-flight for the sample-in and sample-out data are plotted in Fig.2. The maximum dead time correction for $t_m > 10^4$ was less than 20%. It has been demonstrated in Refs. [1,8] that bias effects resulting from such corrections are negligible. Therefore, the uncertainties related to the dead time correction were not propagated.

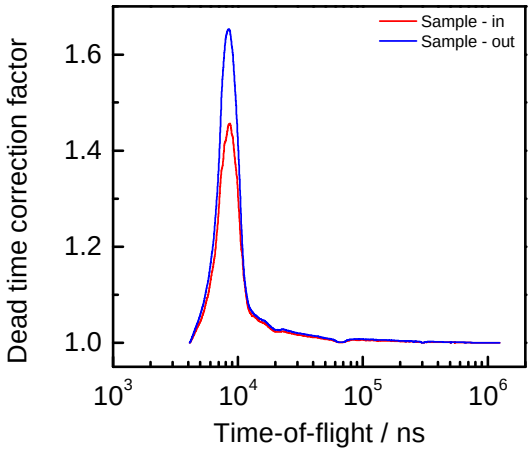

Figure 2 Dead time correction factor as a function of time-of-flight for both the sample-in and sample-out measurements.

All spectra were normalized to the same TOF-bin width structure and neutron beam intensity. The latter was derived from the response of the BF_3 beam monitors. To avoid systematic uncertainties due to slow variations of both the beam intensity and detector efficiency as a function of time, data were taken by alternating sample-in and sample-out measurements in cycles of about 900'' each. Such a procedure reduces the uncertainty on the normalization due to the beam intensity to less than 0.25 % [1]. This uncertainty was evaluated from the ratios of the counts in the ^{6}Li transmission detector and in the flux monitors. To account for this uncertainty the factor N = 1.0000 ± 0.0025 was introduced in Eq.1.

The background as a function of TOF was determined by an analytical expression applying the black resonance technique. The factor K = 1.00 ± 0.03 in Eq. 1 introduces a correlated uncertainty component accounting for systematic effects due to the background model. The background as a function of TOF was approximated by a sum of a constant and three exponentials [1]:

$$B(t_m) = b_0 + b_1 e^{-\lambda_1 t_m} + b_2 e^{-\lambda_2 t_m} + b_3 e^{-\lambda_3 (t_m + \tau_0)} \quad . \tag{2}$$

where τ_0 is related to the operating frequency of the accelerator, i.e. here τ_0 = 1.25 ms for the accelerator operating at 800 Hz. The time independent contribution b_0 is very small and can be estimated from measurements when the accelerator is not in operation. The first exponential accounts for the contribution due to the detection of 2.2 MeV γ-rays resulting from neutron capture in hydrogen that is present in the moderator. The second exponential originates predominantly from neutrons scattered inside the detector station. The last component is due to the detection of overlap neutrons. The free parameters in the analytical expression (b_0, b_1, b_2, λ_1, λ_2 and λ_3) were determined by a least squares fit to saturated resonance

dips observed in the TOF-spectra resulting from measurements with black resonance filters. The time dependence of the background was derived from dedicated measurements with S, Na, Cu, Co, W, Au and Ag black resonance filters in the beam. During the regular sample-in and sample-out runs Na, Co and W fixed filters were kept in the beam to continuously monitor the background at 2.85 keV, 132 eV and 20 eV, respectively, and to account for the dependence of the background level on the presence of the sample [1]. Examples of dead time corrected TOF-spectra together with the background contributions are shown in Figs. 3 and 4, respectively. The parameters of the analytical expression in Eq. 2 are given in Table 2.

The time-of-flight t_m of a neutron creating a signal in the neutron detector was determined by the time difference between the start signal (T_0) and the stop signal (T_s):

$$t_m = (T_s - T_0) + t_0 \quad , \tag{2}$$

with t_0 a time-offset which was determined by a measurement of the γ-ray flash. The flight path distance L = 49.345 (0.004) m, i.e. the distance between the centre of the moderator and the front face of the detector, was derived previously from result of transmission measurements on ^{238}U using the 6.673 ± 0.001 eV resonance of ^{238}U+n as a reference [9].

Table 2 Parameters for the analytical expressions of the background correction for the sample-in and sample-out measurements.

ID	$b_0/10^{-8}$ ns	$b_1/10^{-7}$ ns	$\lambda_1/10^{-5}$ ns^{-1}	$b_2/10^{-7}$ ns	$\lambda_2/10^{-6}$ ns^{-1}	$b_3/10^{-7}$ ns	$\lambda_3/10^{-6}$ ns^{-1}
C_{in}	1.25	3.58	2.94	0.209	1.35	5.47	2.70
C_{out}	1.41	4.61	2.94	0.275	1.35	7.41	2.70

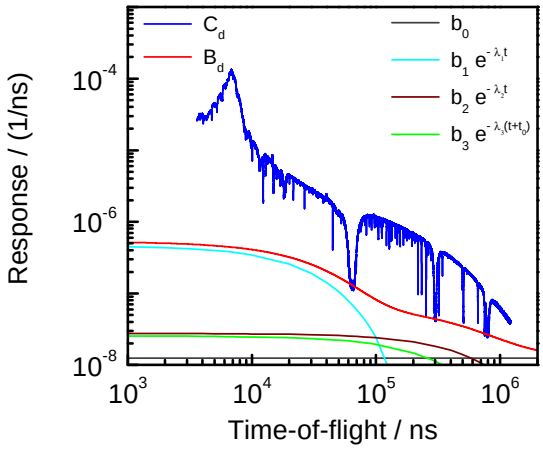

Figure 3 Time-of-flight spectrum without sample in the beam is compared with the total background and its components.

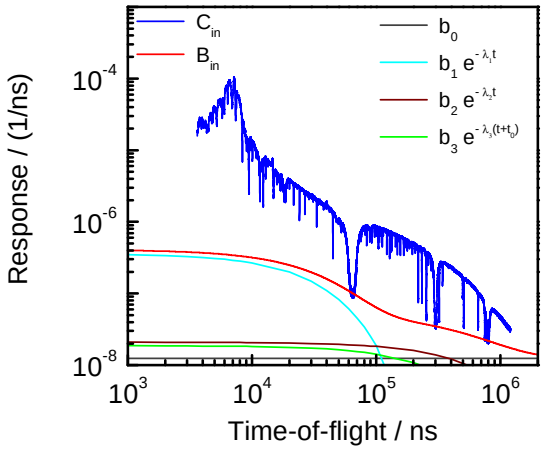

Figure 4 Time-of-flight spectrum with the dummy sample in the beam is compared with the total background and its components.

4. Results

The AGS (Analysis Of Geel Spectra) code [10], developed at the EC-JRC-IRMM, was used to derive the experimental transmission. The code is based on a compact formalism to propagate all uncertainties starting from uncorrelated uncertainties due to counting statistics. It stores the full covariance information after each operation in a concise, vectorized way. The AGS formalism results in a substantial reduction of data storage volume and provides a convenient structure to verify the various sources of uncertainties through each step of the data reduction process. The concept is recommended by the INDC [5] to prepare the experimental observables, including their full covariance information, for storage into the EXFOR data library [11,12]. The transmission for the measurements with the Ca sample and the dummy sample are both delivered to the EXFOR data base.

The experimental transmission resulting from the measurements on the 50 mm thick ^{nat}Ca sample is shown in Fig. 5. The format in which the numerical data is stored in the EXFOR data library is illustrated in Table 3. Fig. 5 compares the experimental transmission resulting from the experiments described in this work and the theoretical transmission using the resonance parameters recommended in the JEFF-3.2 library. This figure shows obvious inconsistencies between the present data and the recommended parameters: e.g. the interference profile in the 20 keV region shows that the parity assignment of the 20 keV resonance as an s-wave is not correct; below 50 keV the contribution of bound states and/or scattering radius is too large; and most of the resonance energies and neutron widths require an adjustment. Hence, a new evaluation of the parameters is required.

The data in Table 3 include the full covariance information based on the AGS concept. Applying the AGS concept described in Ref. 10 the covariance matrix V of the experimental transmission can be calculated by:

$$V = U_u + S(\eta)\, S(\eta)^T \quad , \tag{4}$$

where U_u is a diagonal matrix containing the contribution of all uncorrelated uncertainty components. The matrix S contains the contribution of the components η = {N,K} creating correlated components. The total uncertainty and the uncertainty due to uncorrelated components are reported, together with the contributions due to the normalization to the neutron beam intensity (N) and background model (K).

It is recommended that only the data between 200 eV and 2.5 keV and between 3 keV and 150 keV are used for a resonance shape analysis. The experimental details, which are required to perform an analysis of the data in terms of reaction model parameters, are summarized in Appendix A.

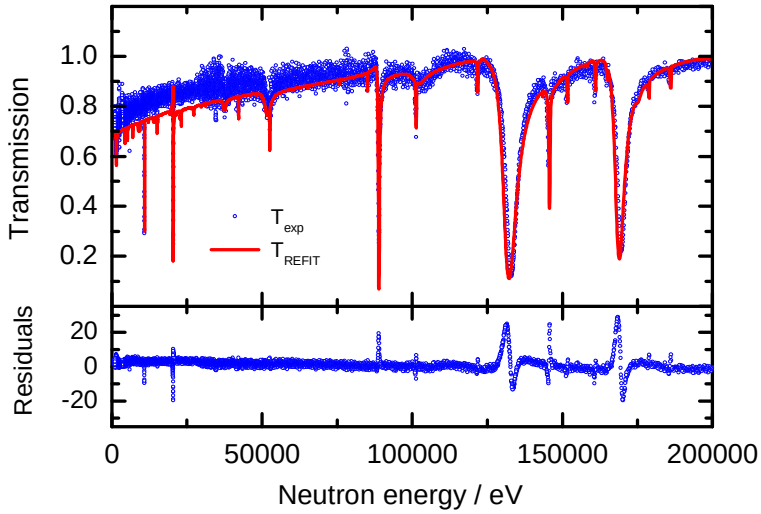

Figure 5 Comparison of the experimental transmission and the transmission derived from the resonance parameters in the JEFF-3.2 library. For the calculations the REFIT code [13] was used.

Table 3 Illustration of the data structure submitted to the EXFOR data library. The first column is the energy that is derived from the measured time-of-flight based on the relativistic equation and a flight path length L= 49.345 m. The second and third column provide the time-of-flight boundaries. The final experimental transmission together with the total uncertainty are given in column 4 and 5. The information to derive the full covariance matrix based on the AGS-formalism (Eq. 4) is given in columns 6, 7 and 8: the diagonal elements due to the uncorrelated uncertainty components are in column 6. The data required to account for the correlated components $S_{\{K,N\}}$ are given in columns 7 and 8. A high number of significant digits is given to ensure that the resulting covariance matrix can be inverted.

E/ eV	t_l / ns	t_h / ns	T_{exp}	u_t	AGS		
					u_u	S_K	S_N
198837.4	8000	8002	0.892	0.0103	0.0101	-0.0000288	0.00223
…	…	…	…	…	…	…	…
8.180	1247232	1247360	0.852	0.1313	0.1273	-0.0319624	0.00213
8.178	1247360	1247488	0	0	0	0	0
…	…	…	…	…	…	…	…
1.28	3149760	3149824	0	0	0	0	0

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Appendix A
SUMMARY OF EXPERIMENTAL DETAILS

A. EXPERIMENT DESCRIPTION

Main Reference		[1,2]
Facility	GELINA	[3]
Neutron production		
Neutron production beam	Electron	
Nominal average beam energy	100 MeV	
Nominal average beam current	55 μA	
Repetition rate (pulses per second)	800 Hz	
Pulse width	1 ns	
Primary neutron production target	Mercury cooled depleted uranium	
Target nominal neutron production intensity	$3.4\ 10^{13}\ s^{-1}$	
Moderator		
Primary neutron source position in moderator	Above and below uranium target	
Moderator material	2 water filled Be-containers around U-target	
Moderator dimensions (internal)	2 x (14.6 cm x 21 cm x 3.9 cm)	
Mass	H_2O	
Temperature (K)	Room temperature	
Moderator-room decoupler (Cd, B, …)	None	
Other experimental details		
Measurement type	Transmission	
Method (total energy, total absorption, …)	Good transmission geometry	[4]
Flight Path length (m) (moderator – detector: face to face distance)	L = 49.345 m	
Flight path direction	9^{o} with respect to normal of the moderator face viewing the flight path	
Neutron beam dimensions at sample position	45 mm in diameter	
Overlap suppression	^{10}B overlap filter (0.008 at/b)	
Other fixed beam filters	Na, Co, W, Pb (16 mm)	
Detector		
Type	Scintillator (NE912)	
Material	Li-glass	
Surface Dimensions	152.4 mm in diameter	
Thickness (mm)	6.35 mm in thick	
Detector(s) position relative to neutron beam	In the beam	
Sample		
Type (metal, powder, liquid, crystal)	Metal	
Chemical composition		
Areal number density (at/b)	^{nat}Ca: $(10.97 \pm 0.02) \times 10^{-2}$	
Isotopic composition (at%)	^{40}Ca (96.941), ^{42}Ca (0.647), ^{43}Ca (0.135), ^{44}Ca (2.086), ^{46}Ca (0.004), ^{48}Ca (0.187)	
Temperature	22 ^{o}C	
Sample mass (g)	(206.277 ± 0.002) g (see table 1)	
Geometrical shape (cylinder, sphere, …)	Cylinder	
Nominal surface dimension	60 mm diameter (see table 1)	
Nominal thickness (mm)	48 mm (see table 1)	
Containment description	Al canning	
Additional comment	Stack of 3 discs	

Data Reduction Procedure		[4,5]
Dead time correction	Done (< factor 1.2)	
Back ground subtraction	Black resonance technique	
Flux determination (reference reaction, …)	-	
Normalization	1.000 ± 0.0025	
Detector efficiency	-	
Self-shielding	-	
Time-of-flight binning	Zone length / bin width: 6240 / 2 ns 4096 / 4 ns 4096 / 8 ns 4096 / 16 ns 4096 / 32 ns 4096 / 64 ns 6144 / 128 ns 28672 / 64 ns	
Response function		
Initial pulse	Normal distribution, FWHM = 2 ns	
Target / moderator assembly	Numerical distribution from MC simulations	[6,7]
Detector	Analytical function defined in REFIT manual	[8]

B. DATA FORMAT

Column	Content	Unit	Comment
1	Energy	eV	Relativistic relation using a fixed FP length of 49.345 m
2	TOF_{min}	ns	Low TOF-bin boundary
3	TOF_{max}	ns	High TOF-bin boundary
4	T_{exp}		Transmission
5	Total Uncertainty		
6	Uncorrelated uncertainty		Uncorrelated uncertainty due to counting statistics
7	S_N-vector		Normalization (u_N/N = 0.25 %)
8	S_K-vector		Background model (u_K/K = 5 %)

Comments from the authors:
The AGS concept was used to derive the experimental transmission

$$T_{exp} = N\frac{C_{in} - K B_{in}}{C_{out} - K B_{out}}$$

and to propagate the uncorrelated uncertainties due to counting statistics and the uncertainty due to the normalization (u_N/N = 0.25 %) and background model (u_K/K = 3 %).
The quoted uncertainties are standard uncertainties at 1 standard deviation
The transmission of the measurements with the Ca sample and dummy Al-container are both given.

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