High-precision measurement of the ^{238}U(n,γ) cross-section at the n_TOF facility, CERN

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Abstract

A new experiment to measure the ^{238}U(n,γ) cross-section has been carried out at the CERN n_TOF facility within the ANDES project. Two complementary detection systems have been used: two low neutron sensitivity C_6D_6 detectors and the Total Absorption Calorimeter (TAC) comprising 40 BaF_2 scintillators. High quality data has been obtained up to 10 keV with the TAC and up to a higher neutron energy unresolved region with the C_6D_6. All the steps in the measurements and the subsequent data analysis are dealt with in depth, aiming for overall 2-3% accuracy per detection system.

Introduction

New concepts for nuclear systems are being explored to improve the sustainability of nuclear energy that appears in the EU SET-Plan (EC, n.d.) as an unavoidable component in the mix for energy generation. Despite many previous measurements, challenges still exist within the present level of basic nuclear data knowledge, as shown in the case of ^{238}U. The OECD/NEA High Priority Request List and WPEC SG26 report (OECD/NEA, n.d., 2008) state the need for a reduction in the uncertainty of the ^{238}U(n,γ) cross-section to 1-2% in the neutron energy range between 22 eV and 25 keV. Measurements have been performed at both the n_TOF (Guerrero, *et al.*, 2013) facility and the GELINA (Ene, *et al.*, 2010) facility, using two contrasting detection systems to reduce any systematics from a single measurement. The measurements performed at the n_TOF facility will be presented in this contribution.

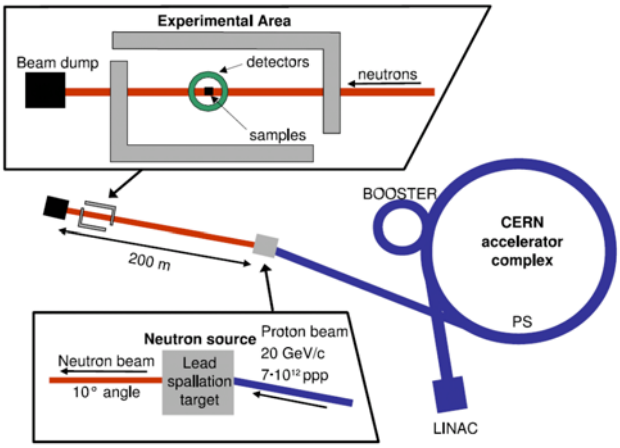
Experimental set-up

The n_TOF Facility at CERN

At the n_TOF facility (Guerrero, *et al.*, 2013) neutrons are generated in spallation reactions by a pulsed 20 GeV/c proton beam impinging on a lead block (see Figure 1), which is surrounded by 5 cm of water and borated water which serves as a coolant and as a moderator of the originally fast neutron spectrum. The resulting white neutron beam ranges from thermal energies to 1 GeV with a nearly isolethargic flux dependence up to 100 keV. The neutrons travel through an evacuated beam line to the experimental area at a distance of 185 m from the spallation target, where the sample and the detectors are placed.

Figure 1: Layout of the n_TOF facility within the CERN accelerator complex

20 GeV/c protons are sent to the n_TOF lead spallation target in bunches of 7×10^{12} protons. The experimental hall is located near the end of the 200 m long neutron beam line.



The n_TOF experimental programme includes measurements of fission cross-sections performed with parallel plate avalanche counters (PPAC) and MicroMegas (MGAS) detectors, and of capture cross-sections studied either with total energy detectors C_6D_6 or with the TAC. An overview of the n_TOF facility and the various measuring devices is given in Guerrero, *et al.* (2013). One of the main features of the experimental set-up at n_TOF is the fully digitised data acquisition system, which is described in detail in Abbondanno, *et al.* (2005).

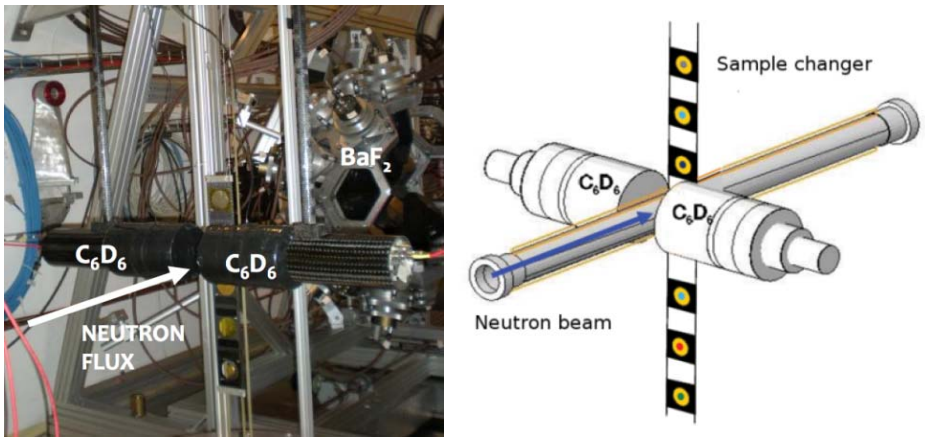
Samples

The ^{238}U sample was provided by the EC-JRC-IRMM, where an isotopic analysis was undertaken in 1984. It is an extremely pure, 6.125(2) grams approximately rectangular (53.90×30.30 mm^2) sample which contains <1 ppm of ^{234}U, ~11 ppm ^{235}U and <1 ppm ^{236}U. It is a wide sample, which with perfect alignment covers 97% of the n_TOF neutron beam. The sample was encased in ~60 microns of aluminium foil and ~75 microns of Kapton, so as to comply with CERN's radioprotection regulations. As well as measuring ^{238}U, it was necessary to perform some auxiliary measurements to accurately characterise any backgrounds that were present. For the TAC measurement, these samples were ^{197}Au, no sample or canning present and ^{nat}C. For the C_6D_6 measurement these samples were no sample present, Al + Kapton (sample canning), ^{nat}C and ^{nat}Pb. For the C_6D_6 detection set-up ^{197}Au, ^{nat}Fe and ^{nat}Ag samples were measured to validate the analysis procedure. Further details of the sample properties can be found in Schillebeeckx, *et al.* (n.d.).

C_6D_6 set-up

The measurement was carried out using two C_6D_6 liquid scintillators, placed opposite each other at 90° with respect to the beam, 9 cm upstream from the sample: one commercial BICRON and one custom made [Forschungszentrum Karlsruhe (FZK)] (Plag, *et al.*, 2003). Both the detectors and the geometry are optimised to have a very low sensitivity to scattered neutrons and a very low solid angle in order to apply the total energy detection method, which requires that the total detection efficiency for a capture event turns out to be proportional only to the total energy released in the event itself. To achieve this proportionality, only one γ-ray per cascade is detected and the pulse height weighting technique (PHWT) (Abbondanno, *et al.*, 2004) is exploited, which utilises the assumption that it is always possible to find a function of the pulse height W(E) that, once convoluted with the pulse distribution from capture events, gives as a result the total radiative energy.

Figure 2: The C_6D_6 detection system in the experimental area (left) and modelled for simulations (right)

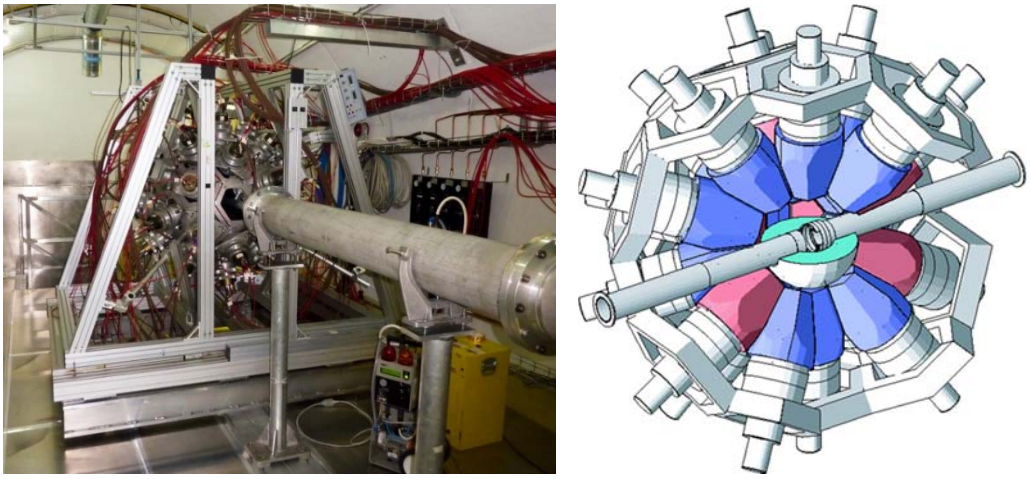


TAC set-up

The Total Absorption Calorimeter (TAC) (Guerrero, *et al.*, 2009) is a segmented 4π detector array made of 40 BaF_2 crystals specifically designed for measuring neutron capture cross-section measurements at the n_TOF facility. The large solid angle coverage and high efficiency for detecting gamma rays means the TAC can reach an almost 100% efficiency for detecting capture cascades. It has been used in many previous measurements to accurately measure capture cross-sections from small mass actinides (Massimi, *et al.*, 2010; Guerrero, *et al.*, 2012a) and recently has been shown to work in combination with a fission detector to measure fission and capture cross-sections simultaneously using a tagging technique (Guerrero, *et al.*, 2012b). The TAC features excellent time resolution (~0.6 ns signal rise time), reasonable energy resolution (~12% at 1.8 MeV) and strong background rejection capabilities using analysis conditions in the crystal multiplicity (m_{cr}) and the deposited energy (E_{sum}).

To reduce the background induced from neutrons scattered at the sample, each crystal is surrounded by a ^{10}B loaded (16% in mass) carbon fibre capsule and a borated polyethylene neutron absorber surrounds the sample. The neutron absorber material was carefully selected to minimise the background due to scattered neutrons using Monte Carlo simulation software, Geant4 (Agostinelli, *et al.*, 2003). A detailed description of the TAC can be found in Guerrero, *et al.* (2009).

Figure 3: The TAC detection system in the experimental area (left) and modelled for simulations (right)



Data analysis: RAW data to capture yield

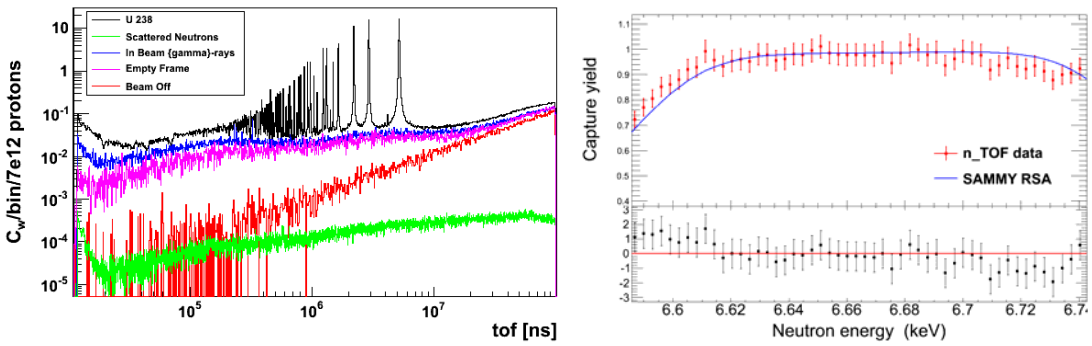
For both measurements the raw TOF spectra were converted into neutron energy by the use of well-known ^{238}U resonance energies.

C_6D_6 analysis

The required proportionality between the γ-ray efficiency and total radiative energy released in a capture event is achieved with the PHWT. To calculate the weighting functions, the precise knowledge of the detector response as a function of energy is required, which is obtained from very detailed Monte Carlos simulations using Geant4 (Agostinelli, *et al.*, 2003). The detectors were calibrated in energy by three calibration sources (^{137}Cs, ^{88}Y and Am-Be). Furthermore, the stability of the neutron flux and detector gain was monitored to ensure only wholly accurate data was taken. In Figure 4 the time of flight spectra are shown for ^{238}U and the main backgrounds. It is seen that the uranium radioactivity dominates for long flight times, i.e. at low neutron energies, while for energies above 100 keV, the main background contribution is due to in-beam γ-rays. The background related to scattered neutrons is negligible as expected for the C_6D_6 detection set-up.

Figure 4: Weighted counts for the ^{238}U(n,γ) reaction with all the main background components (left); the first saturated resonance of ^{238}U fitted with SAMMY (right)

Left figure key: ^{238}U reaction – black, natural radioactivity – red,
sample independent – magenta, in-beam γ-rays – blue, scattered neutrons – green



The observable in-neutron capture cross-section measurements is the capture yield $Y_{(n,\gamma)}$ i.e. the number of capture reactions occurred per incident neutron, which is defined from measureable quantities as:

$$Y_{(n,\gamma)}(E_n) = \frac{C_W(E_n) - B_W(E_n)}{N_{BIF} \cdot \varepsilon_{n,\gamma} \cdot \varphi_n(E_n)} \quad (1)$$

where $C_W(E_n)$ and $B_W(E_n)$ are the total and background counts per pulse, weighted by the same weighting functions, N_{BIF} (beam interception factor) is the fraction of the beam intercepted by the sample, $\varepsilon_{n,\gamma}$ is the efficiency for detecting capture cascades and $\varphi_n(E_n)$ is the neutron flux per pulse. The neutron flux at n_TOF has been well characterised using different detection systems and reactions of known cross-sections, as discussed in Guerrero, *et al.* (2013). The values of N_{BIF} and $\varepsilon_{n,\gamma}$ are difficult to calculate accurately and usually dominate the overall uncertainty of capture cross-section measurements. In order to avoid this, in this particular measurement the thickness of the ^{238}U sample has been chosen in such a way that in the first three resonances all the neutrons arriving at the sample undergo at least one interaction, thus being saturated at their peaks. Moreover, for the first resonance the neutron width is much smaller compared with the radiation width. Under this condition, $Y_{(n,\gamma)}$ becomes constant and close to one, therefore the product $N_{BIF}\varepsilon_{n,\gamma}$ is determined by comparing the measured yield with the expected one at the saturated region of the first resonance. The resonance analysis code SAMMY (Larson, 2008) has been used to calculate the expected capture yield and the fit can be seen in Figure 4. It has been shown that this saturated resonance method can be used to determine the normalisation with an uncertainty of less than 1%, as discussed in Schillebeeckx, *et al.* (2012). Further details of this analysis can be found in Mingrone, *et al.* (2014).

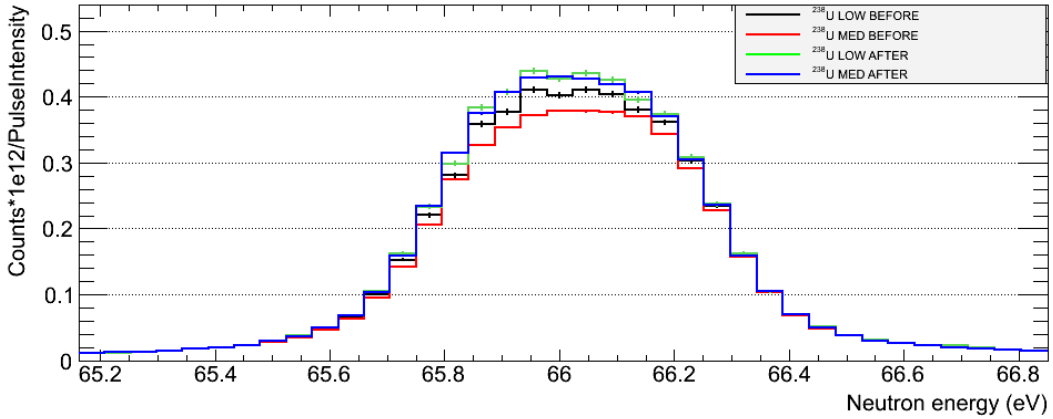
TAC analysis

The TAC was calibrated in energy with three γ-ray sources (^{137}Cs, ^{88}Y and Am-Be). Furthermore, the two simultaneous γ-rays of ^{88}Y were used to calibrate the 40 crystals in time, which allows a coincidence window of 20 ns to be used. One of the advantages of the TAC is that the signal to background ratio can be optimised by putting a constraint on the multiplicity and total deposited energy. For this measurement, the optimal conditions were found to be $m_{cr} \geq 2$ and $2.5 < E_{sum} < 5.75$ MeV. This specifically allows the rejection of the 478 keV and 2.2 MeV γ-rays originating from the ^{10}B(n,$\alpha\gamma$) and ^{1}H(n,γ) reactions respectively.

Similar backgrounds are present within the TAC as the C_6D_6, however some additional effects must be taken into account. Firstly, the TAC has higher neutron sensitivity, therefore the background originating from neutrons scattered by the sample must be taken into account. This background is particularly problematic, as it follows the same resonant structure as the ^{238}U(n,γ) cross-section. This is done using the measurement of a ^{nat}C sample, which can be considered a pure neutron scatterer. In the TAC deposited energy spectra, one assumes any counts above the neutron separation energy of ^{238}U (S_n ~ 4.9 MeV) are from scattered neutrons. By scaling the experimental counts in this region to that of the carbon spectra, one retrieves an estimation of the neutron scattering background. Further information can be found in (Guerrero, *et al.*, 2009).

Due to the TAC's high efficiency and the large sample size, dead time and pile-up effects influence the results. To correct for this, a novel dead time correction method has been applied (Mendoza, *et al.*, n.d.; Guerrero, *et al.*, n.d.). To validate the correction, two different beam intensities were taken (low ~ $0.6.10^{12}$ protons per pulse and med ~ $1.1.10^{12}$ protons per pulse) which subsequently gave different counting rates. As is shown in Figure 5, before the correction is applied counts are being lost due to dead time and pile-up when a higher count rate is present, however after application the two data sets agree within 1%. Furthermore, the asymmetrical shape of the 66.02 eV resonance due to

Figure 5: Illustration of the effect of dead time in the TAC data



pile-up changes after application of the correction. The capture yield is subsequently calculated following the same procedure as for the C_6D_6. Further details of this analysis can be found in Wright, *et al.* (2014).

Results and uncertainties

The following plots show the capture yields from each detection system in different neutron energy regions.

Figure 6: Experimental capture yield Y_{exp} of a ^{238}U sample with an areal density of 9.56 10^{-4} at/b resulting from measurements at n_TOF with the C_6D_6 detectors

Experimental data are compared with the corresponding theoretical yield Y_{cal} derived with SAMMY using the parameters of Derrien, *et al.* (2005).

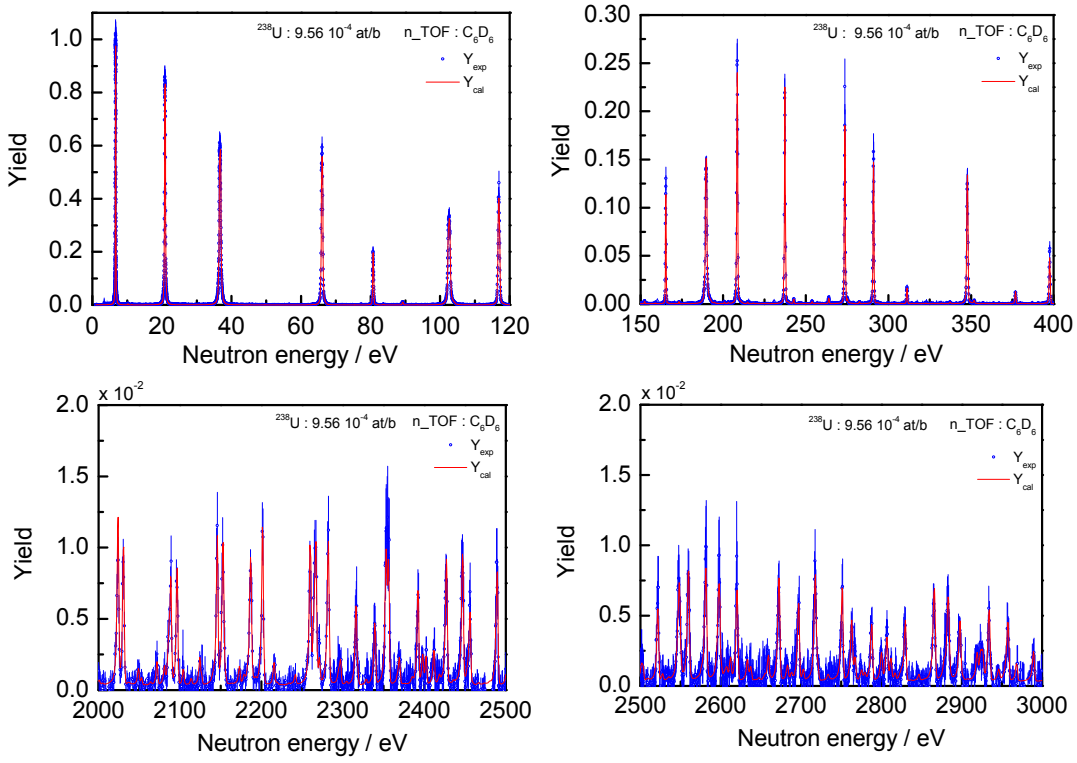
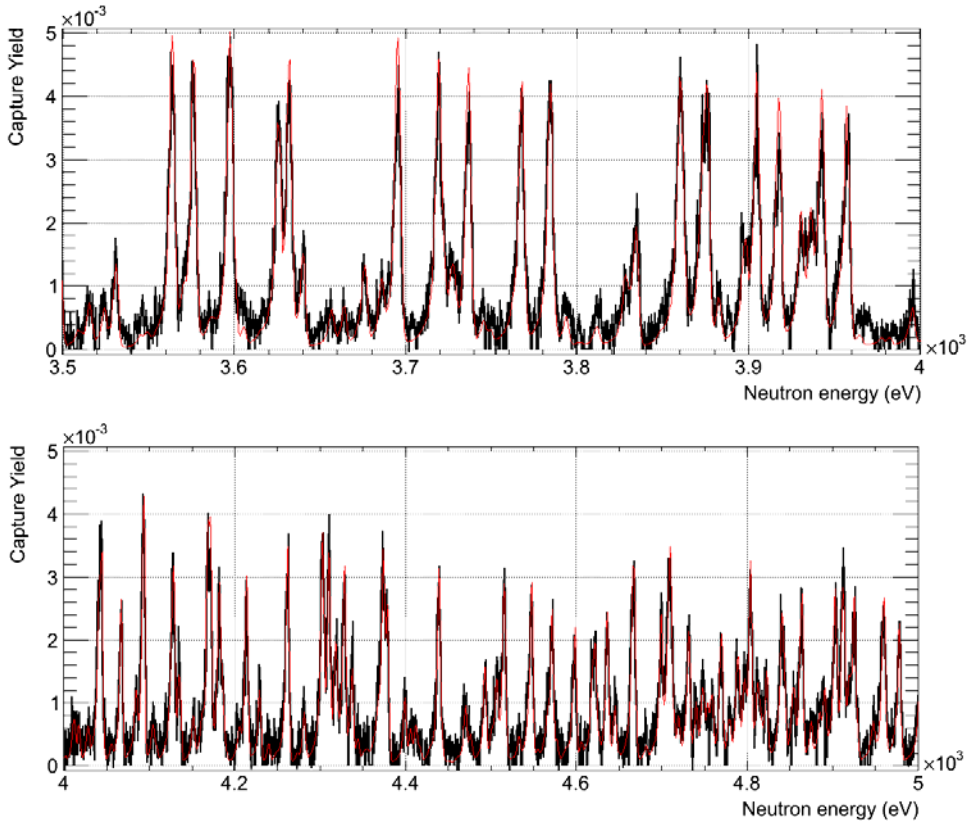


Figure 7: Experimental capture yield Y_{exp} of a ^{238}U sample with an areal density of 9.56 10^{-4} at/b resulting from measurements at n_TOF with the TAC

Experimental data are compared with the corresponding theoretical yield Y_{cal} derived with SAMMY using the parameters of Derrien, *et al.* (2005)



The two data sets complement each other, as better statistics are obtained with the TAC but with an upper limit in neutron energy of 10 keV. As above this energy range, the TAC is saturated by the so-called γ-flash and the cross-section can be analysed as unresolved through the C_6D_6 detection system, allowing the statistical error to be kept to a minimum. For a detailed description of the uncertainties within the two measurements, see Schillebeeckx, *et al.* (n.d.). It is believed that by taking care of all the sources of systematic error in the best way, the final uncertainty of the yield will be of the order 2-3% for each measurement. This is on the limit of what is possible from a time-of-flight cross-section measurement facility.

Summary, conclusions and outlook

In summary, a new measurement of the ^{238}U(n,γ) cross-section has been successfully completed at the n_TOF facility, CERN. Two separate capture yields have been extracted which allow an analysis of resolved resonances up to 10 keV, and an unresolved cross-section analysis above this. All backgrounds have been dealt with utmost care, and the resulting uncertainties have been minimised and quantified. An initial comparison with the current evaluations suggest that a good agreement is seen, but there are significant differences of individual resonances therefore this new data set should prove extremely useful in any future ^{238}U(n,γ) evaluation. A detailed resonance analysis evaluation will be performed in the immediate future, where any true differences from the evaluations will be quantified.

Acknowledgements

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