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MEASUREMENT OF SPECTRUM-AVERAGED REACTION CROSSSECTIONS ON TIN ISOTOPES

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JULY 2024

Measurement of spectrum-averaged reaction cross-sections on tin isotopes

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1 INTRODUCTION

1.1 MOTIVATION

Nuclear data are fundamental quantities that are required in the design and evaluation of nuclear technologies and systems. Detailed simulation of these systems, from simple detectors to advanced reactor concepts, is a robust and cost-effective methodology for determining operational and safety parameters. The quality and reliability of these simulations depends on the quality of the nuclear data.

For the EURATOM call for Nuclear Fission and Radiation Protection (NFRP-2018), a nuclear data project was proposed called SANDA, Solving Challenges in Nuclear Data for the Safety of European Nuclear facilities. As part of the SANDA project NPL proposed to measure the spectrum-average cross section (SACS) of ^{117}Sn(n,n')^{117m}Sn and ^{60}Ni(n,p)^{60}Co. The need for such measurements arises because of known shortcomings in, for example, the data in the reactor dosimetry file IRDFF (see IAEA reports INDC(NDS)-0616 and INDC(NDS)-0639, and the High Priority Request List under Special Purpose Quantities).

1.2 NUCLEAR CAPABILITIES AT NPL

The Nuclear Metrology Group at the UK National Physical Laboratory has expertise and facilities across a wide range technical areas important for this project.
Neutron metrology facilities are used to produce well-characterised neutrons fields in a large low-scatter experimental area. These standardised fields are realised using a 3.5 MV Van de Graaff accelerator and a range of radionuclide sources.
Radiochemistry expertise and facilities are available to prepare the activated samples and enable accurate measurement.
A suite of well-characterised gamma spectrometers, calibrated using standardised radioisotope solutions, is available for use.
Expertise in Monte Carlo simulations using MCNP is available.

1.3 THE NICKLE REACTION

The cross section for the ^{60}Ni(n,p)^{60}Co reaction peaks at approximately 170 mb at 13 MeV (ENDF/B-VIII.0). When the cross section is folded with the ^{252}Cf spectrum (spectrum of W. Mannhart in 71 group structure, taken from ENDF/B-VIII.0) a determination of the spectrum-average cross section (SACS) gives approximately 2.7 mb. Calculating the saturated activity of a typical ^{60}Ni sample using the calculated SACS, the number of atoms in the sample, the emission rate of the ^{252}Cf neutron source and half-life of the daughter product (^{60}Co, ~5.3 years) results in ~12 Bq. After a realistic irradiation of 10 days the activity of the sample is ~0.04 Bq. This is too low to be measured with the required accuracy using the proposed method and ^{252}Cf sources available at present at NPL.

1.4 THE TIN REACTION

The cross section for the ^{117}Sn(n,n')^{117m}Sn reaction peaks at approximately 750 mb at 9 MeV (The European Activation File EAF-2010). When the cross section is folded with the ^{252}Cf spectrum (spectrum of W. Manhart in 71 group structure, taken from ENDF/B-VIII.0) a determination of the spectrum-average cross section (SACS) gives approximately 214 mb. Calculating the saturated activity of a typical tin sample using the calculated SACS, the number of atoms in the sample, the emission rate of the ^{252}Cf neutron source and half life of the daughter product (^{117m}Sn, ~14 days) results in ~116 Bq. After a realistic irradiation of 10 days the activity of the sample is ~45 Bq. Measuring a sample with this activity in a well characterised high purity Ge (HPGe) detector at NPL (158 keV gamma intensity of 86% and

detector efficiency of 0.43%) gives a statistical uncertainty of better than 0.5% after a 3-day acquisition period.

This measurement is complicated by natural tin consisting of several isotopes (see Table 1)

Table 1: Isotopic abundances of stable tin isotopes

Isotope	Abundance (%)
$_{112}$Sn	0.970
$_{114}$Sn	0.66
$_{115}$Sn	0.34
$_{116}$Sn	14.5
$_{117}$Sn	7.68
$_{118}$Sn	24.2
$_{119}$Sn	8.59
$_{120}$Sn	32.6
$_{122}$Sn	4.63
$_{124}$Sn	5.79

The reaction of interest, ^{117}Sn(n,n')^{117m}Sn, produces the meta-stable state of ^{117}Sn state. However, this state is also produced by the ^{116}Sn(n,γ)^{117m}Sn and the ^{118}Sn(n,2n)^{117m}Sn reactions. The SACS for these two reactions when folded with a ^{252}Cf spectrum are 2.6 mb and 0.98 mb respectively. Combining the spectrum averaged cross sections with the isotopic abundances it is expected (using EAF-2010 and assuming the ratio of the cross sections are correct) that ~96.4% of the ^{117m}Sn activation will come from the ^{117}Sn(n,n') reaction and 2.2% and 1.4% from the ^{116}Sn(n,γ) and ^{118}Sn(n,2n).

All of the SACS in section 1.3 and 1.4 were performed per isotope, i.e. the SACS for ^{117}Sn(n,n')^{117m}Sn is the SACS per ^{117}Sn isotope. To convert to the SACS per ^{nat}Sn these numbers can be multiplied by the atomic abundance (see in Table 1 for tin isotopes) and summed. So, the SACS of ^{nat}Sn(x,x)^{117m}Sn is approximately, (214 mb x 7.68%) + (2.6 mb x 14.5%) + (0.98 mb x 24.2%) = 17 mb.

2 EXPERIMENTS

The experiments performed can be split into two campaigns.

2.1 CAMPAIGN 1

The first campaign consisted of irradiating 4 natural tin foils, 15 mm in diameter, 1 mm thickness, with neutrons from a well characterised ^{252}Cf source. The foils were placed ~9 cm from the centre of the source and irradiated for a period of 18 days. The neutron fluence rate incident on the foils was ~4.2x10^{4} neutrons.cm^{-2}.s^{-1}, and the integrated neutron fluence was 7.1x10^{10} neutrons.cm^{-2}.

The initial intent was to count the decay radiation in a well-characterised and shielded HPGe detector system, but this was unavailable at the time of the experiment. A shielded NaI scintillator detector was used to count the decay radiation; however, this system was uncharacterised before this experimental work. The detector crystal was cylindrical, 102 mm diameter and 102 mm height coupled to a photomultiplier. The system utilised analogue electronics with the detector connected to a Canberra 2007B preamplifier an Ortec 57A amplifier and an Ortec EASY-MCA. The detector system was characterised for energy and efficiency using 5 calibration point sources of varying activity, namely: ^{241}Am, ^{133}Ba, ^{22}Na, ^{137}Cs

and ^{60}Co. These emitted gamma rays covering energies of 60 keV to 1332 keV. The sources were mounted approximately 12 cm from the centre of the front face of the NaI detector front window. GEANT4 simulations of the system were performed to determine the shape of the detector efficiency curve, which was scaled to fit the experimental data. The peak of interest for the decay of ^{177m}Sn has a gamma energy of 158.56 keV and at this energy the absolute detector efficiency is 2.43 %, for a point source at ~12 cm.

Due to the low activity of the tin foils it was decided to mount them directly on the front face of the detector. This decision improved the counting statistics but necessitates a large geometry correction factor to transfer the measured efficiency of point sources at ~12 cm to extended sources (1 mm thick disks) situated on the detector face. The foils were simulated on the front face of the detector to determine the detection efficiency. At the energy of interest, the absolute efficiency is 22.75 %, for an extended disk on the front face. This correction factor (almost 10) is significant and any uncertainty in this value will linearly impact the result. The shape of the efficiency curve was also different as the low energy gamma rays were less likely to emerge from the thick foil than the point sources. Considerable extra effort to characterise the detector system was required to reduce the uncertainties of the measurement. As such it was decided that this experiment would not be able to meet the required uncertainty of 2-5 %.

2.2 CAMPAIGN 2

A significant source of uncertainty during the first campaign arose from the distance between the source and the activation foil. The foils were mounted ~9 cm from the source with an uncertainty of ~0.2 cm, resulting in an uncertainty in the neutron fluence of >4 %. To remove this uncertainty the second experiment was designed to encapsulate the source in a layer of tin. This results in the uncertainty of the emission rate of the neutron source being the dominant uncertainty in the neutron fluence. Source emission rates are measured at NPL using the manganese bath technique[1] and have a typical standard uncertainty of ~0.5 %.

2.2.1 Irradiation

A source cup was designed and manufactured at NPL to hold a ^{252}Cf source in modified X35 capsule with enough space to wrap the source in a nominal 0.125 mm layer of natural tin. The source cup included a lid allowing tin to be placed on top as well as below the source. This is due to the tin being a soft metal requiring the cup lid to hold it in-place. This arrangement increased the neutron rate as all emitted neutrons were encountering activation foil .

The tin foil, nominally 0.125 mm thick, was cut using a metal punch and scalpel to the correct shape and fit around the inner edge of the two halves of the source holder. The source holder was mounted into the centre of the low scatter area of the Chadwick building at NPL[2]. The foil was irradiated for ~11.8 days with a total neutron emission of 1.557×10^{13} neutrons. This is an increase of over 380 times more neutrons than during campaign 1.

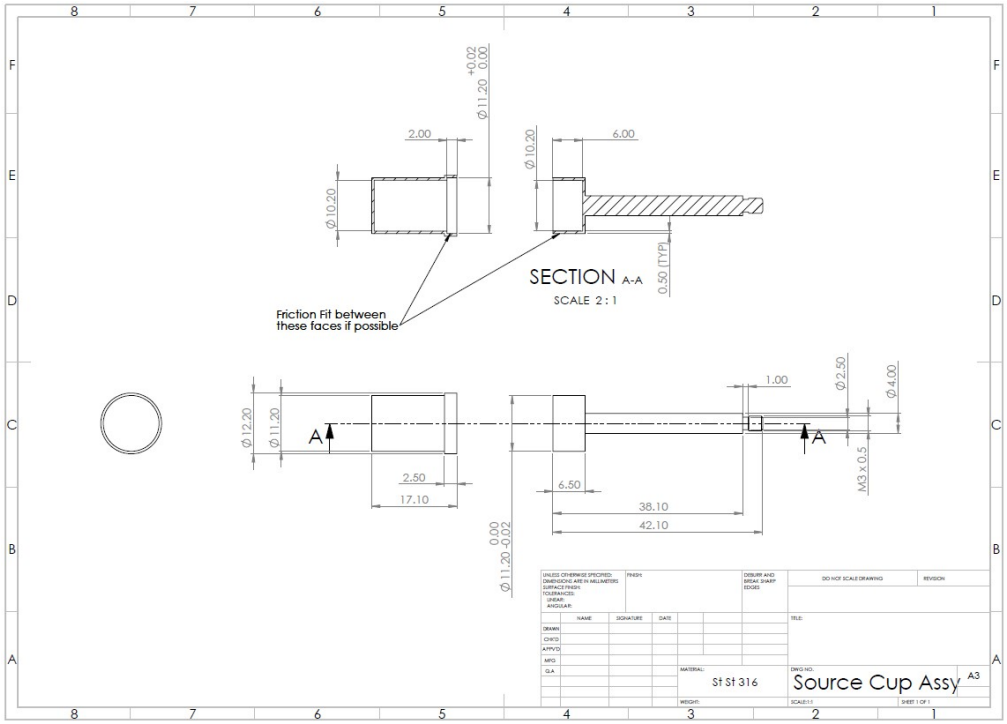

Figure 1: Engineering drawing of the source cup designed to hold the foil and source.

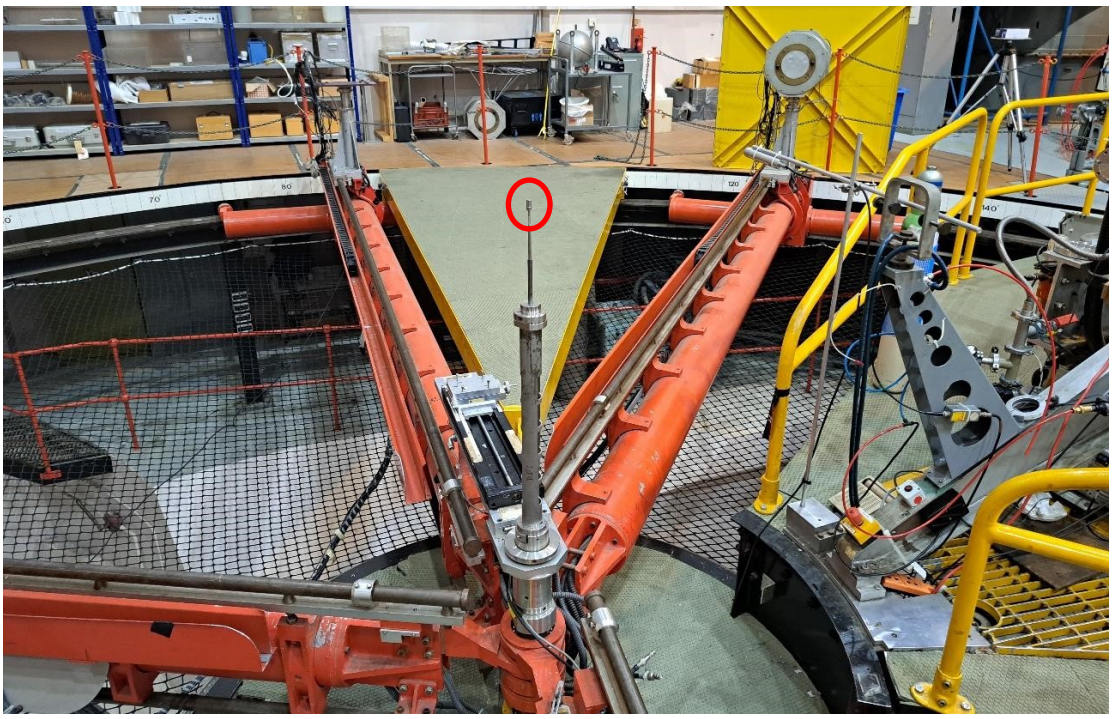

Figure 2: Photograph of the low-scatter area in the Chadwick building at the NPL. The source is held in the source cup holder, circled in red.

2.2.2 Sample preparation

The irradiated foil (approximately 0.72 g) was dissolved to near-dryness by placing in a glass beaker with 10 mL of 6M hydrochloric acid which was located on a warming plate. It was then re-dissolved in 3 mL of 0.1M hydrochloric acid and transferred into a 5 mL ISO glass ampoule which was flame sealed. The transfer from the glass beaker to the ampoule was achieved using a dolly bottle and this process was 99.4 % efficient. All radiochemistry was performed within the radiochemistry suite at NPL and utilised 7-figure balances which are calibrated and

traceable to national standards. Check-weight measurements were performed before the measurement for quality assurance.

Figure 3: Photographs from the radiochemistry suite at NPL. The left picture shows the tin foil dissolving in 10 mL of 6M hydrochloric acid on a warming plate. The right picture shows the 5 mL ISO glass ampoule.

2.2.3 Gamma spectrometry

The activity of the ampoule was determined using a p-type coaxial HPGe gamma detector housed within a large lead shield. The ampoule was measured in a well-characterised geometry using a jig at 15 cm from the front face of the detector. The system is a secondary standard and is calibrated using a mixed radionuclide source which is traceable to primary standards of radioactivity.

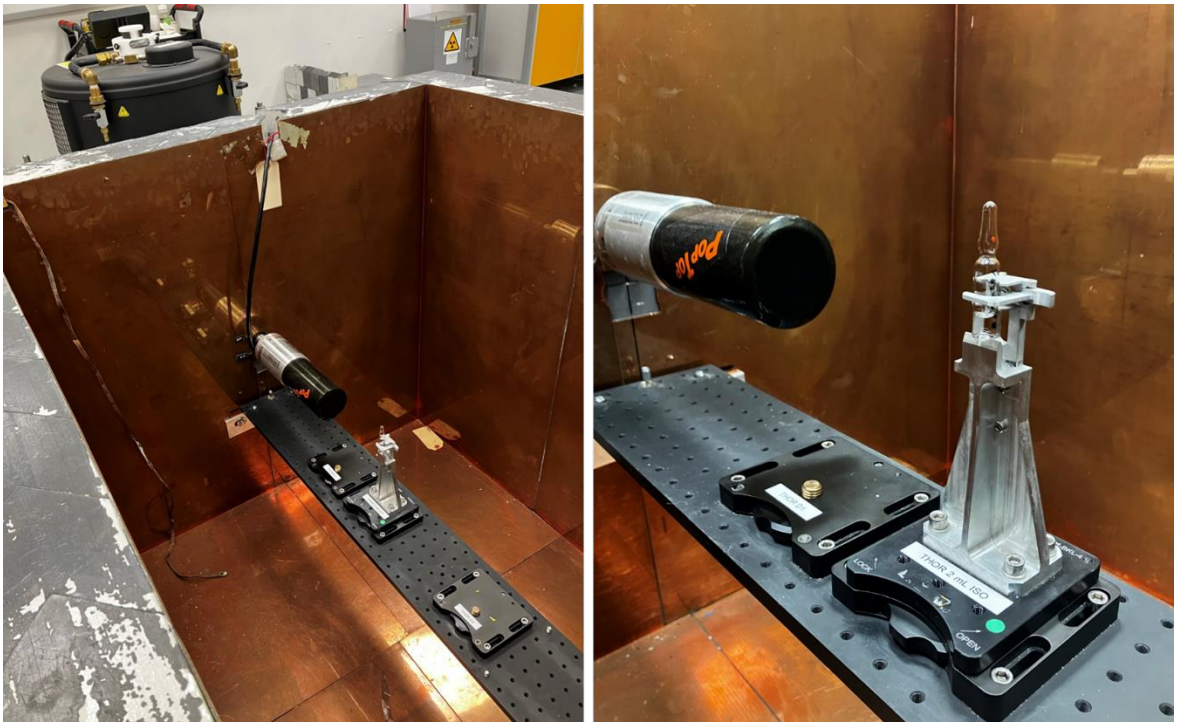

Figure 4: Photograph of HPGe gamma spectrometer within a lead shield. The right picture shows the glass ampule in the measurement location.

The results from the gamma spectrometry give an activity per gram of material of 16.70 ± 0.24 Bq/g. The total activity of the ampoule was 68.69 ± 0.99 Bq. These activities were decay corrected to a reference time of 20/02/2024 12:00:00 UTC.

2.2.4 Determining the measured SACS

The saturated activity was determined using the following equations[3],

$$A_\infty = \frac{\lambda(C-B)}{\in (1-e^{-\lambda t_0}) \cdot e^{\lambda t_0} \cdot (e^{-\lambda t_1} - e^{-\lambda t_2})}$$

where,

A_∞ is the saturated activity,
C is the number of counts,
B is the number of background counts,
ϵ is the detector efficiency, λ is the decay constant for ^{117m}Sn, t_0 is the end of the irradiation, t_1 is the start of the measurement and t_2 is the end of the measurement.

See Figure 5 for illustration (from reference 3). Since the activity at the refence time is,

$$A^{(t_{ref})} = \frac{C-B}{\in}$$

Then,

$$A_\infty = A(t_{ref}) \frac{\lambda}{(1-e^{-\lambda t_0}) \cdot e^{\lambda t_0} \cdot (e^{-\lambda t_1} - e^{-\lambda t_2})}$$

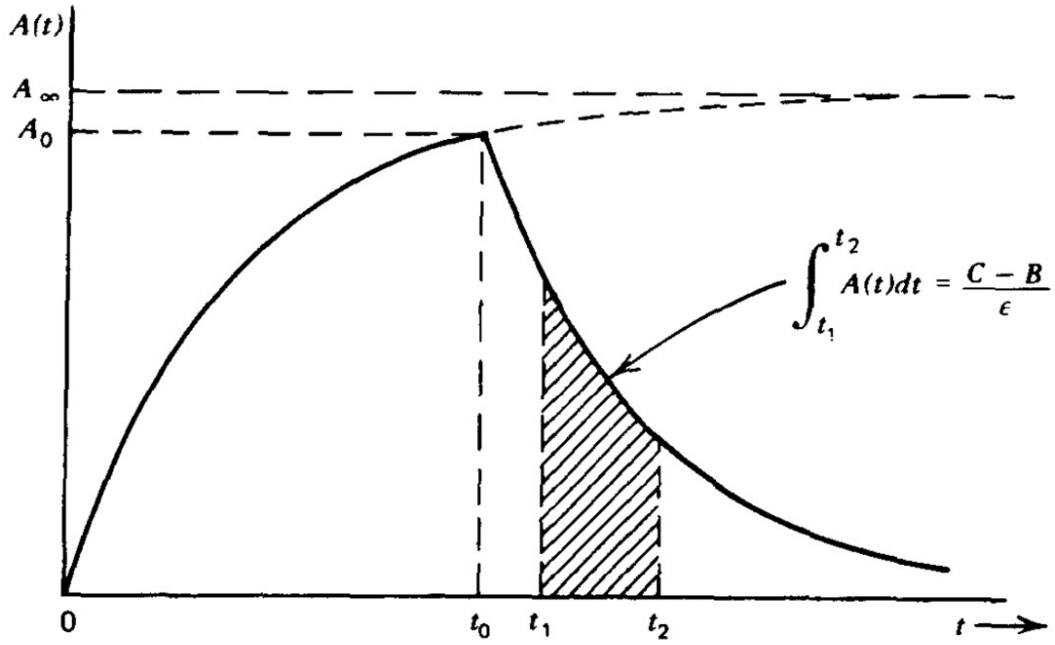

Figure 5: Illustration of the growth and decay of a material under constant neutron bombardment between t=0 and t=t0.

The saturated activity was calculated as 156.1 ± 2.3 Bq. The spectrum averaged cross section is then simply the production rate of ^{117m}Sn (which is the same as the saturated activity), divided by the neutron rate incident on the foil and the number of natural tin atoms within the foil per cm^2. The incident neutron rate is the same as the average emission rate over the irradiation which was (1.529 ± 0.0083) x 10^7 neutrons per second. The number of natural tin atoms per cm^2 was (4.68 ± 0.14) x 10^{20}. The measured spectrum averaged cross section (SACS) per natural tin atom was 21.82 ± 0.72 mb.

2.2.5 MCNP simulations

To compare this to the predicted value the experiment was simulated using MCNP6.1[4] to determine the neutron spectrum that was incident on the activation foil. The laboratory at NPL, including the low-scatter area, were included in the model. The source, source-holder and foil were also included, see Figure 6 for an illustration of the MCNP geometry. The simulation was run for greater than 3 x 10^9 neutrons and an F4 tally was used to record the average flux within the foil volume. The energy was binned using a 709 group structure that has 50 tally bins per energy decade, equally spaced in the logarithm of the energy between 10^{-5} eV and 10 MeV, and thereafter bins with appropriately chosen equally-spaced boundaries in energy up to 1 GeV. The start spectrum used was a Watt spectrum shown below:

$$N(E) = Ce^{E/a}\sinh(bE)^{1/2}$$

where *E* is the neutron energy, C is a normalisation constant, and a and b are the parameters for the Watt spectrum. For ^{252}Cf the a and b parameters used were 1.025 and 2.926 respectively. The output from these simulations are shown in Figure 7. The free in air spectrum (red line) show the source-term as described by the Watt spectrum. The laboratory model spectrum (blue line) shows the flux averaged over the activation foil collected using an F4 tally. The down-scattered component of the field is present, but it is a small fraction of the total field. This is due to the proximity of the foil to the source and large low-scatter environment.

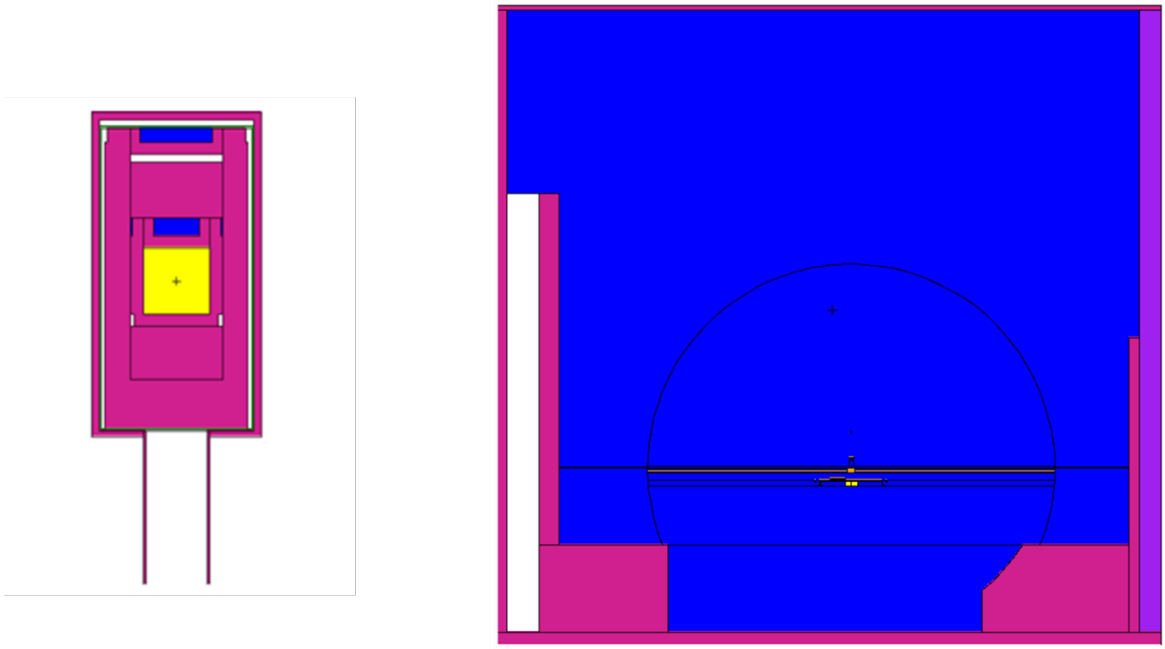

Figure 6: Illustration of the MCNP model geometry of the NPL facility used to simulate the experiments. The left picture shows a zoomed slice on the source, source-cup and activation foil. The ^{252}Cf active material is shown in yellow. The right picture shows a slice of the whole experimental area with source location at the centre of the circle.

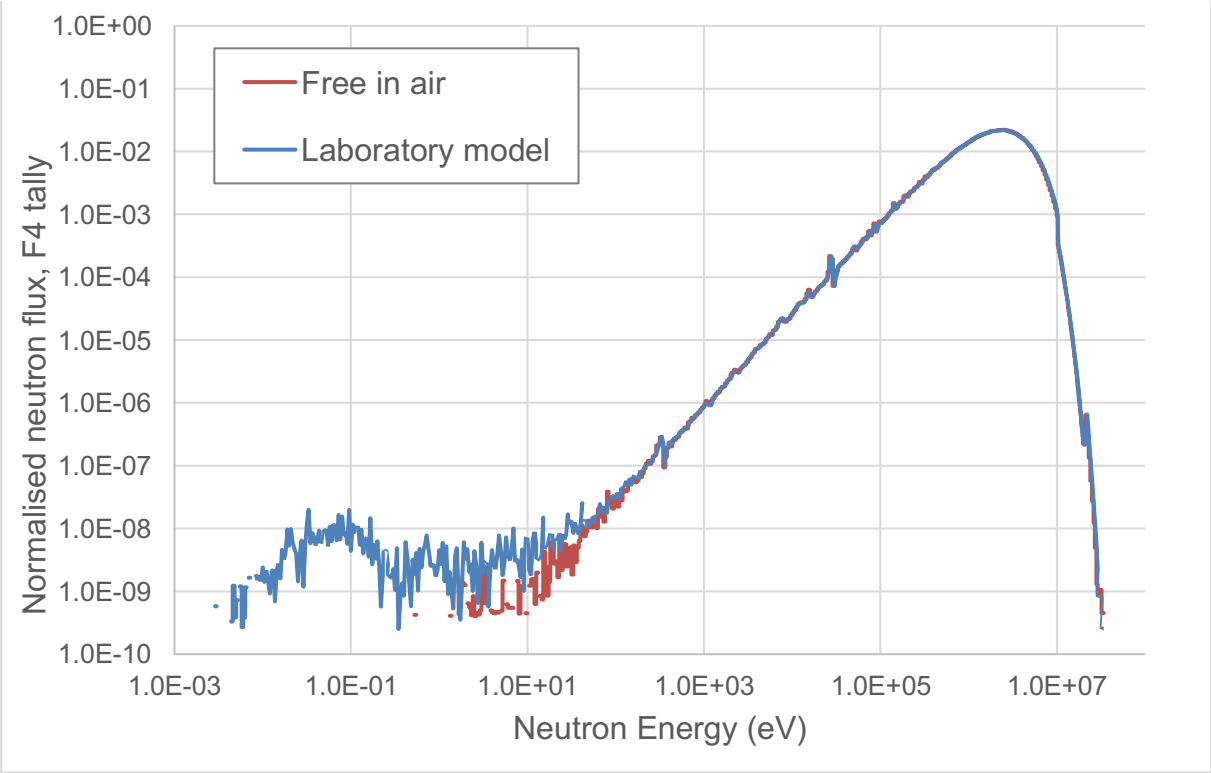

Figure 7: Results of the MCNP simulations. The Free in air simulation shows the source term, i.e. the Watt spectrum. The Laboratory model simulation includes the realistic experimental setup and shows the down-scattered component of the neutron field.

2.2.6 Determining the theoretical SACS

The SACS can simply be calculated by folding the simulated neutron spectrum with the cross-section for each reaction channel, taking data from evaluated nuclear data libraries. The nuclear data was taken from the EAF-2010[5] and JEFF3.0/A[6] libraries as they had evaluated cross sections for each of the three reactions of interest. The data was output using the JANIS4.0[7] software package in the same 709 group structure described above. The SACS for each reaction were multiplied by the atomic abundance for each isotope to compare with the measurement.

Table 2: Results for the Spectrum Averaged Cross Section (SACS) from folding the MCNP simulations with evaluated nuclear data libraries. The three reactions shown populate the ^{117m}Sn state. The sum of the SACS per natural tin is shown at the bottom of the relevant columns.

			SACS (mb)			
			EAF-2010		JEFF-3.0/A	
Isotope	Abundance	Reaction	^{11x}Sn	^{nat}Sn	^{11x}Sn	^{nat}Sn
^{116}Sn	14.50%	^{116}Sn(n,γ)	2.47	0.36	2.47	0.36
^{117}Sn	7.68%	^{117}Sn(n,n')	231.80	17.80	189.53	14.56
^{118}Sn	24.20%	^{118}Sn(n,2n)	1.21	0.29	1.08	0.26
				18.45		15.18

The results from the folding are shown in Table 2. They are lower than the measured SACS of 21.82 ± 0.72 mb. The experimental data and evaluated cross sections used in the folding are shown in Figure 8, Figure 9 and Figure 10.

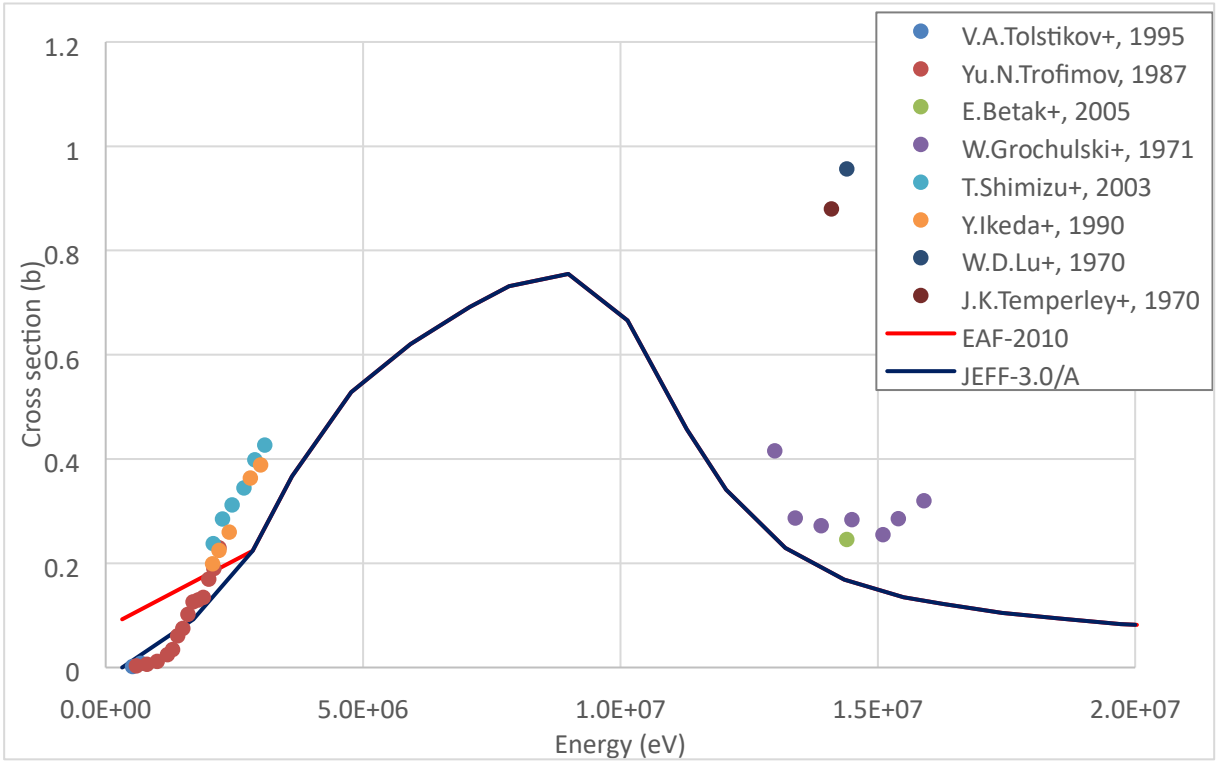

Figure 8: Experimental data for $^{117}Sn(n,n')^{117m}Sn$ from EXFOR shown with evaluations from EAF-2010 and JEFF-3.0/A

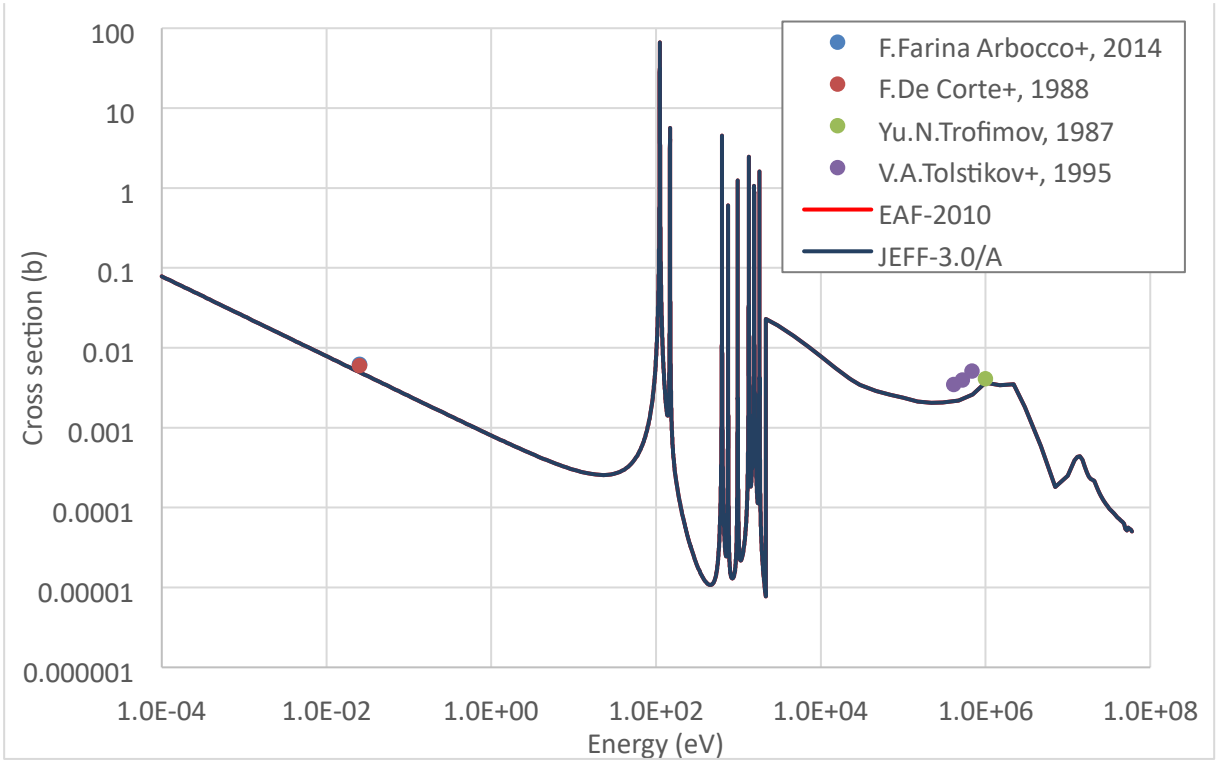

Figure 9: Experimental data for $^{116}Sn(n,\gamma)^{117m}Sn$ from EXFOR shown with evaluations from EAF-2010 and JEFF-3.0/A. The EAF-2010 and JEF-3.0/A evaluations are the same so the curves overlap.

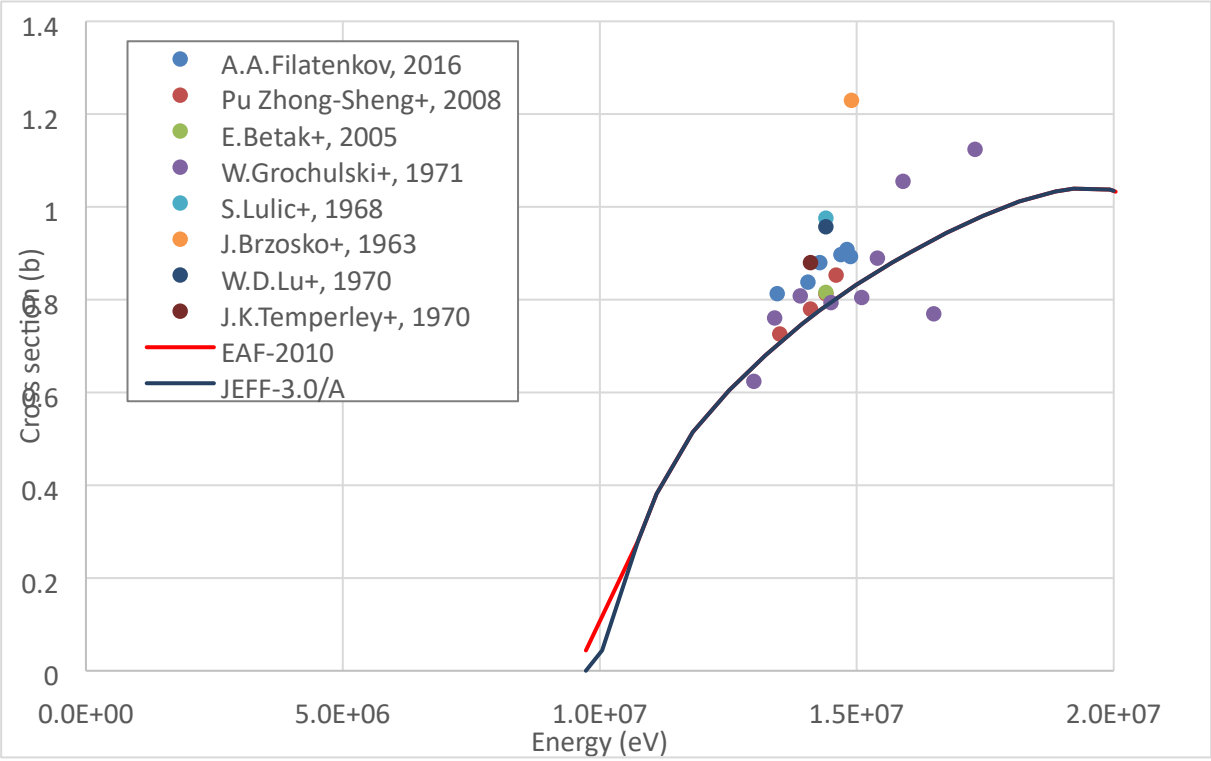

Figure 10: Experimental data for ^{118}Sn(n,2n)^{117m}Sn from EXFOR shown with evaluations from EAF-2010 and JEFF-3.0/A

It can be seen that for the ^{117}Sn(n,n')^{117m}Sn (in Figure 8) that the evaluated cross sections are lower than the data across the majority of the energy range. This is also the case for the ^{116}Sn(n,γ)^{117m}Sn (in Figure 9). If we assume that these evaluated cross sections are correct relative to each other, then we can simply scale the evaluated cross sections using the ratio of the measured SACS to the simulated SACS and compare with the data. The scale factors are 1.1824 and 1.4378 for the EAF-2010 and JEFF-3.0./A libraries respectively. The scaled evaluated data are shown in Figure 11, Figure 12 and Figure 13.

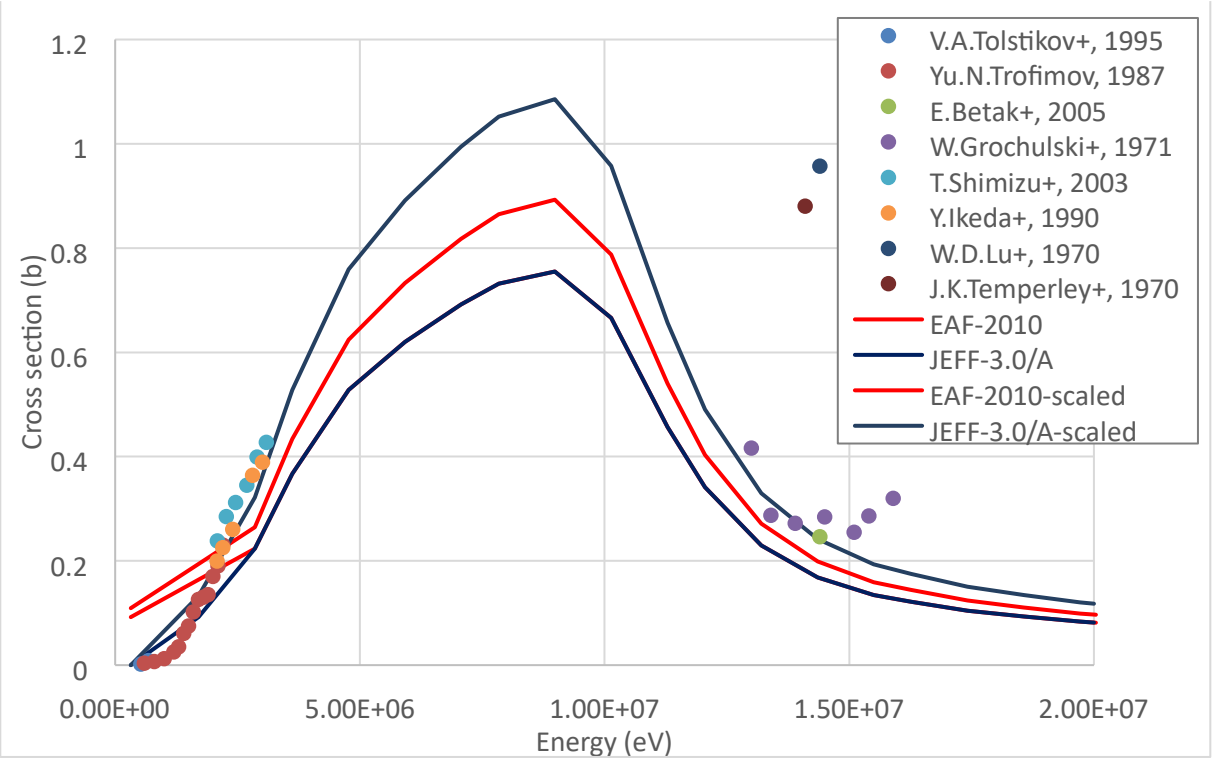

Figure 11: Experimental data for ^{117}Sn(n,n') ^{117m}Sn from EXFOR shown with evaluations from EAF-2010 and JEFF-3.0/A, and scaled versions of the same.

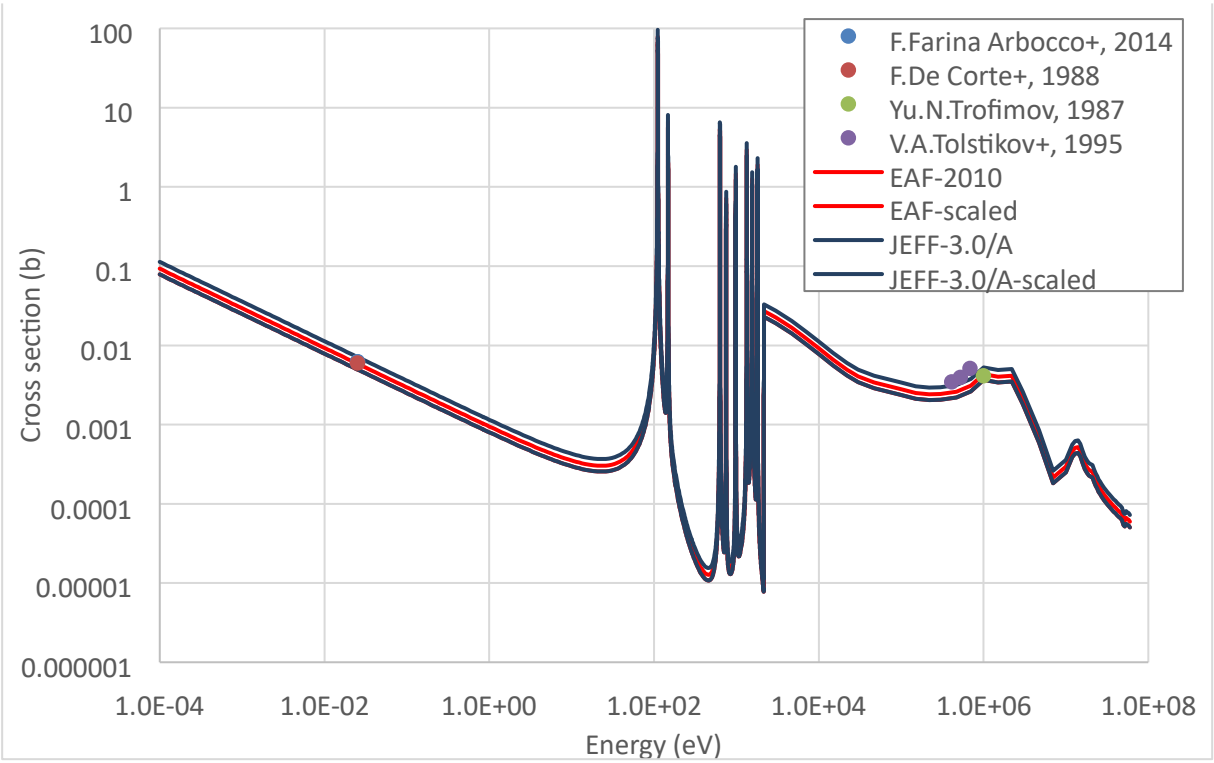

Figure 12: Experimental data for ^{116}Sn(n,γ)^{117m}Sn from EXFOR shown with evaluations from EAF-2010 and JEFF-3.0/A, and scaled versions of the same.

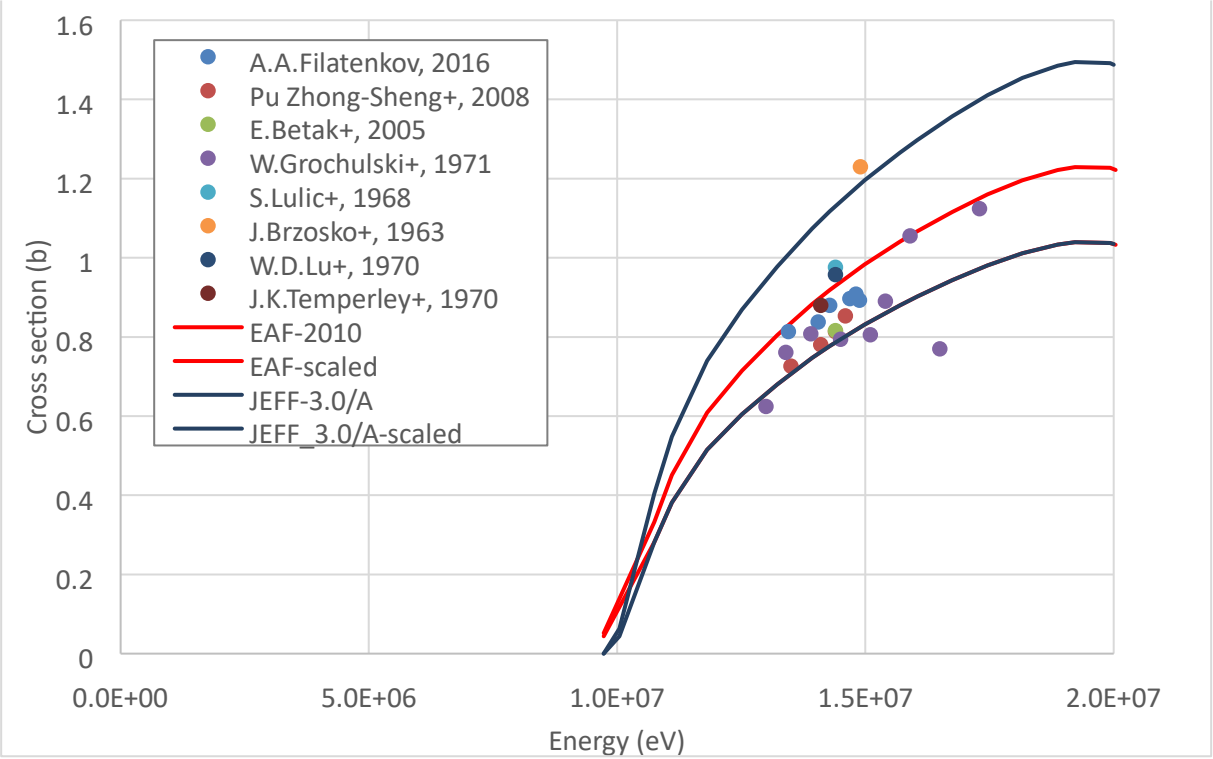

Figure 13: Experimental data for ^{118}Sn(n,2n)^{117m}Sn from EXFOR shown with evaluations from EAF-2010 and JEFF-3.0/A, and scaled versions of the same.

From this simple scaling it is clear the data qualitatively agrees better with the scaled evaluations for the ^{117}Sn(n,n')^{117m}Sn and ^{116}Sn(n,γ)^{117m}Sn. This is particularly the case for the scaled JEFF-3.0/A for the ^{117}Sn(n,n')^{117m}Sn reaction where the low energy section, up to approximately 3 MeV, reproduces the shape of the data better than the EAF-2010 evaluation. This scaling is less successful for the ^{118}Sn(n,2n)^{117m}Sn reaction channel with the scaled evaluations, particularly the scaled JEFF-3.0/A evaluation, appearing to be high compared to the data.

Another approach is to rescale each evaluation to the available data by performing a least squares fit of the data to the evaluation curve by assuming a linear interpolation between the evaluated points and scaling. This method results in a different scale factor for each reaction channel and was performed for the JEFF-3.0/A evaluation.

Table 3: Results for the Spectrum Averaged Cross Section (SACS) from folding the MCNP simulations with evaluated nuclear data libraries that were scaled using a least squares fit to the data. The three reactions shown populate the ^{117m}Sn state. The sum of the SACS per natural tin is shown at the bottom of the relevant columns. These results are for the JEFF3.0/A evaluation.

			SACS (mb)	
Isotope	Reaction	Scale	$_{11x}$Sn	$_{nat}$Sn
$_{116}$Sn	^{116}Sn(n,γ)	1.29	3.19	0.46
$_{117}$Sn	^{117}Sn(n,n')	1.63	309.60	23.78
$_{118}$Sn	^{118}Sn(n,2n)	1.11	1.20	0.29
				24.53

The result using this method is in better agreement than the SACS using the un-scaled evaluated cross section. For the JEFF-3.0/A evaluation the least squared scaled SACS is approximately 12% high compared with the measurement and the un-scaled SACS is approximately 30% low. However the shape of the evaluated cross section for the ^{117}Sn(n,n')^{117m}Sn reaction still does not reproduce the low-energy data and a re-evaluation, particularly below 3 MeV, is required (see Figure 14).

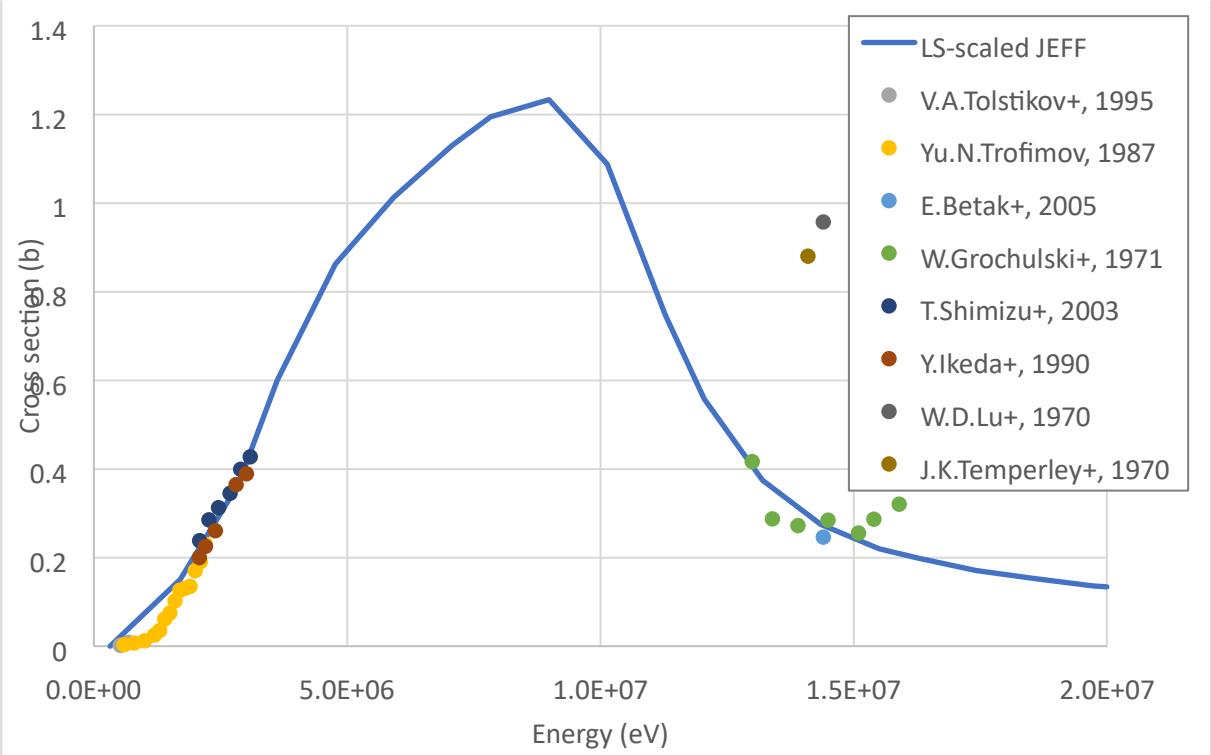

Figure 14: Experimental data for ^{117}Sn(n,n')^{117m}Sn from EXFOR shown with evaluations from JEFF-3.0/A scaled to the data using a least squares method.

2.2.7 Uncertainties

The uncertainties reported above are standard combined uncertainties for each stage of the measurement. This means they are quoted at *k=1* giving a level of confidence of approximately 68 %. These standard uncertainties can be multiplied by a coverage factor, *k=2*, providing a level of confidence of approximately 95%. There is a separate uncertainty budget for each stage of the measurement: irradiation, sample preparation, and gamma spectrometry. These budgets are available on request. These uncertainties were then combined to provide an uncertainty in the SACS.

Throughout this report, the uncertainty evaluation has been carried out in accordance with the JCGM 'Guide to the expression of uncertainty in measurement' (GUM).

3 CONCLUSION

The spectrum averaged cross section (SACS) for the production of ^{117m}Sn from ^{nat}Sn was measured at the National Physical Laboratory. A ^{252}Cf radionuclide source was used to activate the tin sample. This sample was dissolved and sealed in a 5 mL ISO ampoule, using radiochemistry techniques. The activity was then measured in standard gamma-spectrometry measurement geometry using a shielded HPGe detector. The measured spectrum averaged cross section (SACS) per natural tin atom was 21.8 ± 1.4 (6.6%) mb. This uncertainty is based on standard uncertainties multiplied by a coverage factor, *k*=2, providing a level of confidence of approximately 95%.

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MCNP simulations were performed to determine the neutron spectrum incident on the sample. Using this in conjunction with evaluated data libraries the SACS can be calculated and compared to the experimental result. A new evaluation to better reproduce the data would be beneficial. New differential measurements between 3 and 7 MeV would also help to determine the shape of the cross-section in any new evaluation.

4 ACKNOWLEDGEMENTS

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