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Validation of ^{59}Co and ^{93}Nb Activation Cross Sections in a Quasi-Mono Energetic Neutron Spectrum (<35 MeV) Including Irradiation, Measurement and Computational Analysis

Final Report on Task 4.2 of the
Grant Agreement F4E-2010-F4E-GRT-056 (ES-AC), Action 2,
on
"Nuclear Data Experiments and Techniques"

M. Majerle[1], P. Bém[1], J. Novák[1], E. Šimečková[1], S. P. Simakov[2,3], U. Fischer[2]

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April 2016

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Produced by the IAEA in Austria
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Abstract

The quasi mono-energetic neutron source of NPI/Řež based on the p+Li/C reaction was used to irradiate Nb and Co materials with neutrons in the energy range 20 to 35 MeV. The obtained activities were analysed, the related activation cross-sections were extracted with a modified version of the SAND-II code and compared to the values from the EAF-2010 activation data base. A detailed computational study of the setup, experimental and computational procedure was performed to determine the spectral flux at the sample positions and to identify possible sources of uncertainties. Most of the experimental data points obtained as part of this task confirm the validity of the EAF-2010 data. A few reaction cross-sections of the EAF-2010 activation data base, however, need to be revised.

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Disclaimer

The work leading to this publication has been funded partially by Fusion for Energy under the Grant Agreement F4E-2010-GRT-56.02. This publication reflects the views only of the author, and Fusion for Energy cannot be held responsible for any use which may be made of the information contained therein.

0. ABSTRACT

The quasi mono-energetic neutron source of NPI/Řež based on the p+Li/C reaction was used to irradiate Nb and Co materials with neutrons in the energy range 20 to 35 MeV. The obtained activities were analysed, the related activation cross-sections were extracted with a modified version of the SAND-II code and compared to the values from the EAF-2010 activation data base. A detailed computational study of the setup, experimental and computational procedure was performed to determine the spectral flux at the sample positions and to identify possible sources of uncertainties. Most of the experimental data points obtained as part of this task confirm the validity of the EAF-2010 data. A few reaction cross-sections of the EAF-2010 activation data base, however, need to be revised.

1. INTRODUCTION

The materials ^{93}Nb and ^{59}Co are important neutron monitors for devices where neutrons with higher energies are present (fusion, fast reactors, ADS). The EAF-2010 activation database includes a complete set of evaluated cross-sections scaled to experimental data. However, experimental data with cross-sections above 20 MeV are rare, and most database cross-sections above this energy cannot be validated against relevant experimentally measured points.

The p+Li/C facility at the NPI/Řež produces quasi-monoenergetic neutrons up to the energy 35 MeV which were used to measure the missing cross-sections. Seven neutron energies in the range between 20 and 35 MeV were used. Nb and Co materials in the form of thin foils were activated with neutrons and the subsequent activities were measured.

In the computational pre-analysis the optimization of the set-up for the activation experiments was performed and, after completion of the measurements, the cross-sections were extracted, sources of uncertainties were identified and the obtained cross-sections were checked against the EAF-2010 database.

The work performed in Task 4.2 of F4E-GRT-056, Action 2, is a continuation of the program underway at NPI/Řež and KIT/INR for measuring and analysis of the neutron activation cross-sections above 20 MeV for reactions which were selected for the determination of energy distribution and for the monitoring the neutron fluence at accelerator driven facilities such as International Fusion Material Irradiation Facility (IFMIF) [2, 3]. In the previous years the activation cross sections were obtained for ^{209}Bi(n,3-5n) and ^{197}Au(n,2-4n) reactions [4].

The details of the present analysis and experimental work were presented and discussed at several relevant Meetings and Workshops [5, 6].

2. MAIN PARAMETERS OF THE NPI/ŘEŽ EXPERIMENT ON Nb AND Co ACTIVATION

The experimental facility has been described in details elsewhere [1], here the main parameters of the arrangement and procedure are mentioned. A proton beam directed to a thin Li foil (2 mm) backed with thick C beam stopper (1 cm) is used to produce quasi-monoenergetic neutrons at the NPI.

The neutron irradiation of Nb and Co has been performed at the NPI cyclotron at 7 incident proton energies, see Tables 1 and 2 for further details. The resolution (FWHM) and uncertainty of the incident proton energy were measured to be 1.5% [1].

The foils made from pure metallic Nb and Co had a disc shape with diameter 15 mm and thickness 0.5 mm and 0.25 mm respectively. In every experimental run two foils were simultaneously irradiated at distances of 48-49 and 86-87 mm from the neutron source (beginning of the Li foil) to check the consistency of the measurements. Foils were at all experiments arranged with other activation foils in the same order: 0.05 mm Au, 0.15 mm Cu (present only at energies 27.633, 32.56, and 37.411 MeV), 0.25 mm Co, and 0.5 mm Nb. with the same order in both distances.

As it was shown in [6, 7], the ratio of specific activities measured at two distances varies as a function of the proton energy is lower than the ratio calculated on the basis of target-foil distances and exhibits the increasing trend with the energy. This effect is well understood and is mainly due to the different shape of the neutron spectra at two distances (closer distance = larger spatial angle = softer spectrum). The samples were irradiated over the time period of about 20 hours. The irradiation time profile was recorded by measuring the proton current at the target assembly on pulse by pulse scale and the correction for beam instabilities was included in the calculation of the reaction rates.

Twelve radioactive products were detected in the Nb and Co foils (six in Nb, six in Co) by the offline γ-spectroscopy employing two calibrated HPGe detectors with efficiencies of 23% and 50% and an energy resolution FWHM = 1.8 keV at 1.3 MeV. The decay gamma-ray spectra were measured during cooling period from minutes up to 100 days. The activities of the specific isotopes at the end of the irradiation were calculated using tabulated decay half-lives and gamma intensities from the LUND database [8]. The results are given in Tables 1 and 2. The uncertainties given in the Tables include the uncertainty of the gamma peak fitting and the uncertainty of the HPGe detector calibration (ca. 2%). Further systematic uncertainties of the reaction rates are discussed in detail in the sections 6.

Date	Ep[MeV]	^{92m}Nb(10.15d)		^{91m}Nb(60.86d)		^{90}Nb(14.60h)	
		48 mm	86 mm	48 mm	86 mm	48 mm	86 mm
8.8.2008	19.838	81.11±3%	27.93±3%				
17.5.2008	25.126	83.67±3%	28.67±3%	2.257±4%	0.7518±3%		
18.9.2009	27.633	83.82±3%	28.17±3%	3.576±4%	1.189±5%		
14.6.2008	30.075	98.31±4%	33.48±3%	4.556±4%	1.609±3%		
17.4.2009	32.56	112.7±4%	38.87±4%	4.546±4%	1.612±3%	2.178±6%	0.8051±4%
13.6.2008	35.023	143.4±4%	46.47±3%	4.125±4%	1.572±4%	86.59±4%	31.59±3%
29.5.2009	37.441	172.6±3%	50.93±4%	4.175±4%	1.358±5%	353.5±6%	114.8±3%

Date	Ep[MeV]	^{91m}Y(49.71m)		^{90m}Y(3.19h)		^{88}Y(106.65d)	
		48 mm	86 mm	48 mm	86 mm	48 mm	86 mm
8.8.2008	19.838			75.44±3%	26.38±3%		
17.5.2008	25.126			96.67±5%	34.9±3%		
18.9.2009	27.633	16.93±20%	4.822±29%	122.3±4%	37.49±3%	0.03885±6%	0.01467±8%
14.6.2008	30.075	35.73±13%	10.36±15%	140.9±4%	44.45±3%	0.2209±3%	0.08566±3%
17.4.2009	32.56	57.72±10%	18.49±12%	154.9±4%	49.09±3%	0.5972±3%	0.2209±3%
13.6.2008	35.023	86.33±7%	27.37±15%	187.3±4%	56.24±3%	0.9511±3%	0.355±3%
29.5.2009	37.441	119.2±10%	37.73±18%	214.2±5%	53.74±3%	1.189±4%	0.423±4%

Table 1: Experimental activities in the Nb foils. The activities are expressed in [Bq/kg/ C]. The uncertainties shown include the uncertainties of the gamma peak fitting and ca. 2% uncertainty of the HPGe detector calibration.

Date	Ep[MeV]	^{60}Co(5.2714y)		$^{58m+g}$Co(70.86d)		^{57}Co(271.79d)	
		48 mm	86 mm	48 mm	86 mm	48 mm	86 mm
8.8.2008	19.838	0.167 ± 3%	0.0526 ± 3%	31.2 ± 3%	10.5 ± 3%		
17.5.2008	25.126	0.194 ±3%	0.0637 ± 3%	50.5 ± 3%	17.6 ± 3%	0.658 ± 3%	0.234 ± 3%
18.9.2009	27.633		0.0711 ±3%		16.9 ±3%	3.16 ±3%	0.967 ±3%
14.6.2008	30.075	0.275 ±3%	0.0929 ±3%		18.3 ±3%	5.22 ±3%	1.64 ±3%
17.4.2009	32.56		0.0956 ±3%		17.9 ±3%	6.48 ±3%	1.98 ±3%
13.6.2008	35.023	0.365 ±3%	0.120 ±3%		20.6 ±3%	7.11 ±3%	2.130 ±3%
29.5.2009	37.441		0.122 ±4%		22.07 ±4%	7.03 ±3%	2.10 ±3%

Date	Ep[MeV]	^{56}Co(77.27d)		^{59}Fe(44.503d)		^{56}Mn(2.5785h)		^{54}Mn(312.3d)	
		48 mm	86 mm	48 mm	86 mm	48 mm	86 mm	48 mm	86 mm
8.8.2008	19.838			4.06±3%	1.34±3%	807±3%	268±3%		
17.5.2008	25.126			4.93±3%	1.65±3%	780±4%	261±3%		
18.9.2009	27.633			5.33±3%	1.87±3%	829±4%	282±3%		
14.6.2008	30.075			6.62±3%	2.38±3%	992±4%	338±3%		0.0463±5%
17.4.2009	32.56			8.09±3%	2.77±3%	1201±4%	405±3%	0.4615±4%	0.164±3%
3.6.2008	35.023			10.3±3%	3.41±3%	1570±4%	505±3%	0.8403±4%	0.304±4%
29.5.2009	37.441		0.023 ±18%	12.1±3%	3.73±3%	1882±4%	585±3%	1.048±4%	0.355±3%

Table 2: Experimental activities in the Co foils. The activities are expressed in [Bq/kg/ C]. The uncertainties shown include the uncertainties of the gamma peak fitting and ca. 2% uncertainty of the HPGe detector calibration.

3. MCNPX SIMULATION OF THE p-Li NEUTRON SOURCE AND VALIDATION AGAINST THE CYRIC EXPERIMENT

To overcome the previously observed effect of different neutron spectral shapes in two sample positions and study its effect on measured activities [6a], a detailed computational investigation of all relevant arrangement details and their influence on the shape of the spectra was carried out.

The Monte-Carlo particle transport code MCNPX [9] was employed in the present analysis to simulate the proton interactions with the target. For the precise representation of the neutron generation in the target materials, the Los-Alamos evaluations for proton induced cross-sections on ^{7}Li and ^{12}C, LA-150h [10], were used. For other nuclides the relevant cross-sections were represented by the FENDL 2.1 [11] library.

This approach and the nuclear data were checked against the neutron differential yields measured at the CYRIC laboratory (Tohoku university) [12] with a 20-40 MeV proton beam. The experiment has employed a target similar to the NPI target consisting of a 2 mm thick Li foil, 12 mm thick carbon beam stopper and an aluminium case as shown in Figure 1.

The geometry of the CYRIC target setup was accurately modelled in MCNPX and simulations with the LA-150h and FENDL 2.1 cross-section data were performed. A detailed study comparing the simulated and experimental values was done. The conclusions from further similar studies [7] were confirmed, and some new facts were discovered. The comparisons of the spectra for proton energies 20, 25, 30, 35, and 40 MeV can be seen in Figure 2.

In these comparisons special attention has to be put to the following facts:

- The experimental data extend above the border of Ep-1.88MeV (threshold for $p+^{7}Li \rightarrow n+^{7}Be$). This can be accounted for either by the FWHM and energy definition of the proton beam at CYRIC or by the accuracy of the TOF detection system.
- The number of neutrons in the quasi-monoenergetic peak: The increase of forward directed neutrons with energy differs for the CYRIC experiments and corresponding MCNPX simulations (up to 20% for the areas under the quasi-monoenergetic peak). From other experimental data it is difficult to estimate which data are correct [23, 24].
- The simulated data in general slightly underestimate the emission of neutrons below the quasi-monoenergetic peak. Reaction rates for reactions with thresholds around 10 MeV are affected significantly by these neutrons at proton beam energies above 20 MeV and the extracted cross-sections might not be accurate.

From the comparisons in Figure 2 it can be concluded that MCNPX manages to some extent to describe the neutron spectrum and thus can be used as a prediction tool. But it is also obvious that a detailed analysis of the uncertainty of the cross-sections results based on the quality of the neutron field prediction has to be performed. Such analysis will be described in more details in the following sections. However, in general the mentioned uncertainties depend on the cross-section curve and are between 10-50%.

Figure 2 also depicts experimental [12] and calculated results for the pure carbon target bombarded by protons. The comparison with Li/C target shows that the yield of neutrons produced in the carbon beam stopper becomes comparable with that from the $^{7}Li(p,xn)$ reaction only at energies much lower than the quasi-monoenergetic peak (Q=18.1 MeV). For the activation analysis this means that the neutrons originating from the C beam stopper generally do not affect the reactions rates having a threshold above 10 MeV. However, the comparison of the reaction rates calculated with the fields from Li/C and C targets shows that some low threshold reactions at energies above 25 MeV can be substantially influenced by the neutrons from the C beam dump (see Table 3). Since the accuracy of the MCNPX prediction of the neutrons produced by C target only is essentially worse than for neutrons from Li/C (Figure 2), and it is therefore not clear how to extrapolate the lower parts of the neutron spectra to distances 48 and 86 mm, the results where most of the activity was produced by neutrons from the C beam dump should not be regarded as very reliable ones.

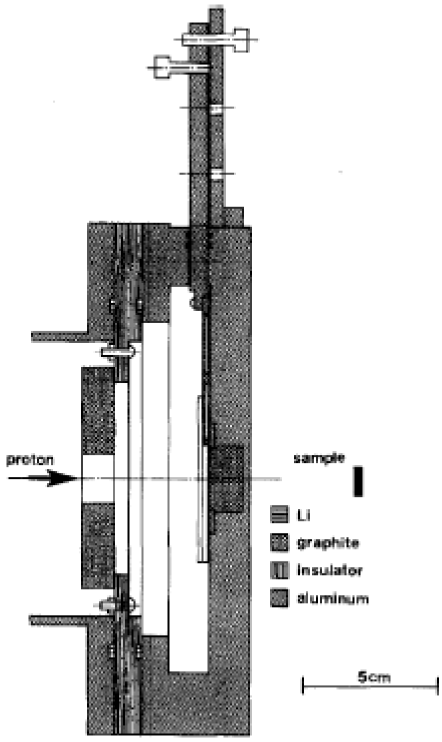

Figure 1: ^{7}Li target set-up at CYRIC laboratory [12].

E_p[MeV]	^{92m}Nb	^{91m}Nb	^{90}Nb	^{91m}Y	^{90m}Y	^{88}Y	^{57}Co	^{56}Co	^{59}Fe	^{56}Mn	^{54}Mn
20	1%	0%		0%	2%	0%			2%	2%	0%
25	2%	0%		0%	2%	0%	0%		3%	3%	0%
30	13%	2%		1%	19%	0%	1%		28%	23%	0%
35	41%	2%	0%	1%	42%	0%	1%	0%	48%	55%	0%
40	68%	9%	2%	4%	57%	3%	6%	1%	72%	75%	3%

Table 3: Differences in activities produced by C spectra only and by spectra from complete Li/C setup.

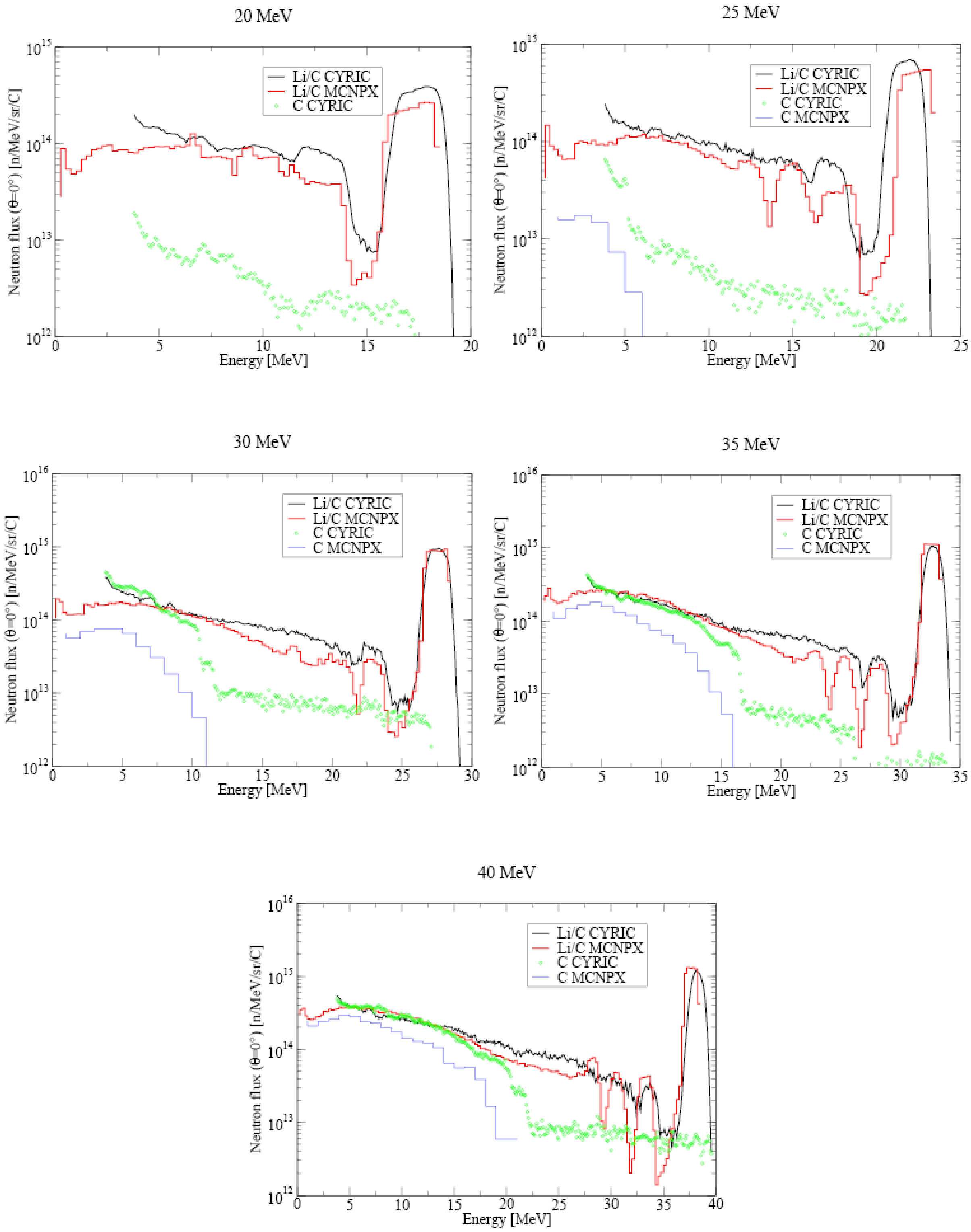

Figure 2: Energy differential neutron yields: measured for Li/C and bare C targets and calculated by MCNPX/LA150h.

4. NEUTRON SPECTRA FOR NPI/ŘEŽ EXPERIMENT

4.1 MCNPX simulation of neutron spectra

For the Monte Carlo calculations, the target setup at NPI/Řež was represented by the geometrical model as seen in Figure 3, which includes a 40 mm (∅) x 2 mm thick lithium foil, a 20 mm (∅) x 10 mm thick carbon beam stopper, a 6 mm thick alcohol coolant, a steel target corpus and an aluminium holder for the activation foils. The proton beam was approximated with a Gaussian profile (3 mm FWHM) and with well defined energy (0 MeV FWHM). The neutron spectra averaged at the positions of the foils at 48 and 86 mm were calculated (MCNPX, with LA-150h and FENDL 2.1 libraries) for all proton energies. In Figure 4, the simulated neutron spectra grouped to 0.25 MeV bins are shown for the distances 48 and 86 mm.

4.2 Inter- and extra-polation of spectra from Uwamino CYRIC results

The spectra measured at the CYRIC facility were binned to 0.25 MeV bins and interpolated to the energies which were used at NPI irradiations of Nb and Co. A simple interpolation procedure was used: the spectrum below and the spectrum above the interpolated energy were translated to the interpolated energy and weight summed. The weights were determined on the basis of the energy difference from the measured to the interpolated energy as $w_1=(E_i-E_1)/(E_2-E_1)$, and $w_2=(E_2-E_i)/(E_2-E_1)$. In our case, the interpolated energies are centered between the energies of the CYRIC spectra (27.633, 32.56), and the weights were taken to be 0.5 (the spectra were translated and averaged). For other energies, the closest CYRIC spectra were taken without changes.

To extrapolate the spectra of the Cyric arrangement (12 meters from the Li target) to the sample positions of the present experiment (48 and 86 mm from the Li target), the factors obtained by MCNPX simulations were used. The spectra simulated for the NPI setup at distances of 48 and 86 mm and the spectra simulated for the Cyric arrangement (the spatial angle of 2°) were compared and spectral ratios were extracted. The simulations show that the ratios are more or less constant above 5 MeV. This constant includes the spatial angles covered by the foils at 48 and 86 mm, the integral over the angular distribution of neutrons (analytical form factors from Uwamino and Schery for the positions 48 and 86 mm are 0.85 and 0.92, respectively), the decrease of the energy with the angle, and other details. Below 5 MeV, the scattered neutrons/neutrons produced in C start to influence the neutron field, as seen in Figure 5. For the extrapolation of the Uwamino spectral data to the present arrangement, the ratios obtained from MCNPX simulations averaged over 2 MeV were used. The obtained spectra are shown in Figure 6.

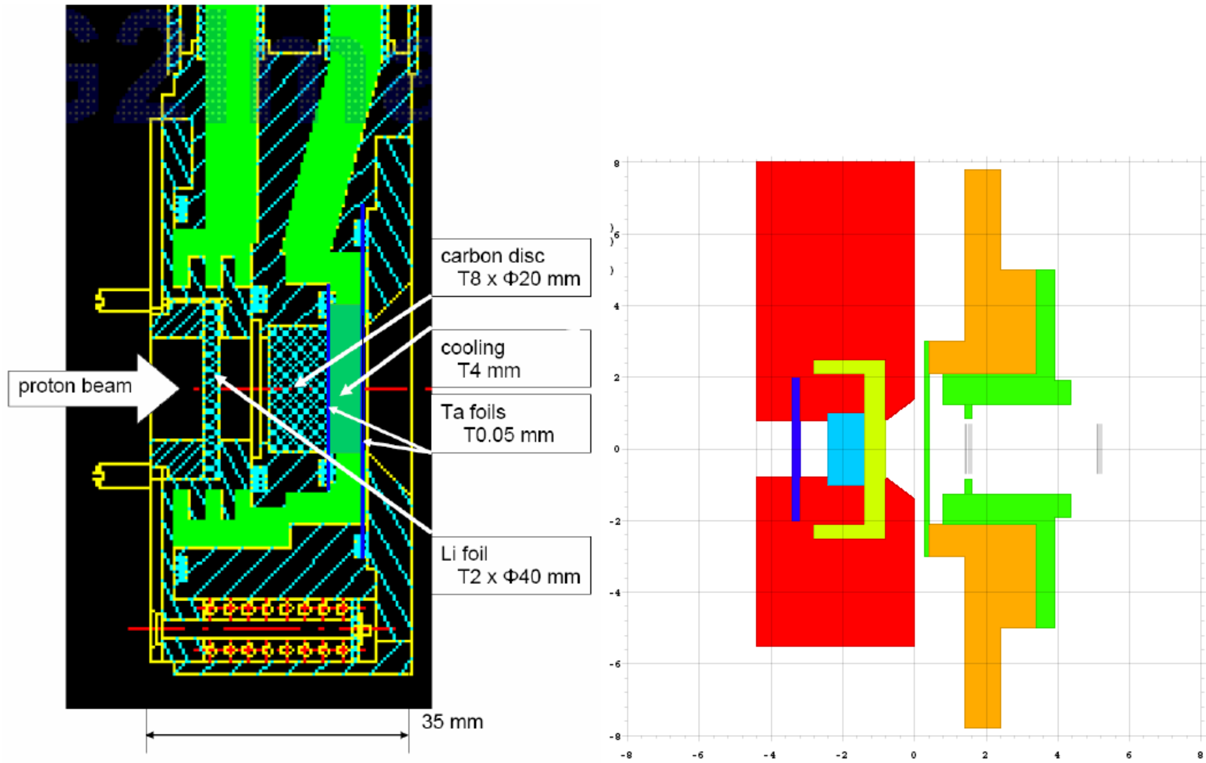

Figure 3: Li target setup at the NPI laboratory and the corresponding MCNPX model.

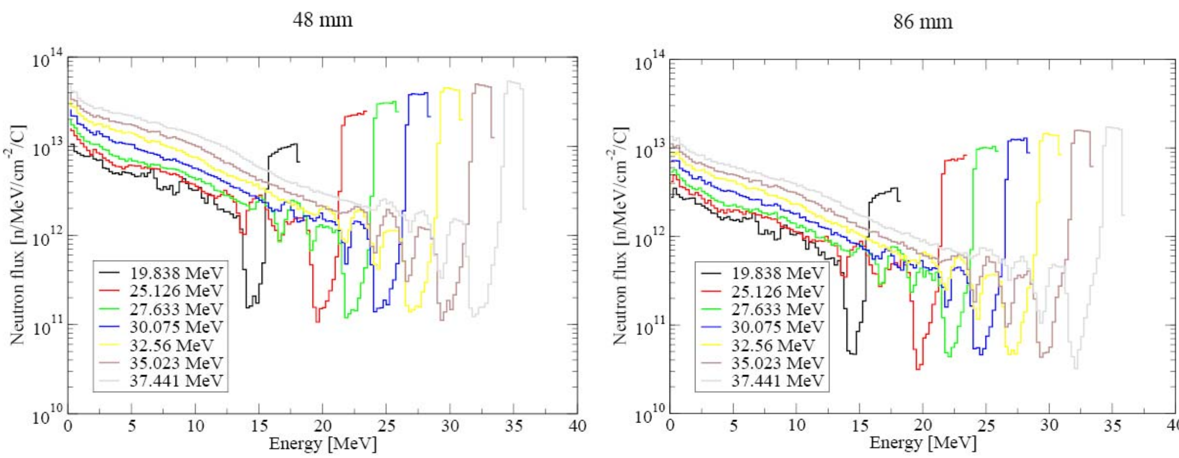

Figure 4: MCNPX calculated spectra averaged over the position of the activation foils at 48 and 86 mm from the target front.

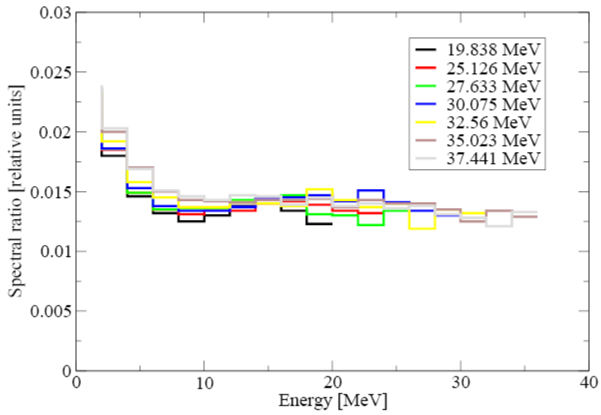

Figure 5: Spectral ratios between the spectra taken under the spatial angle 2° and the spectra in the foil (diameter 14 mm) 86 mm from the target.

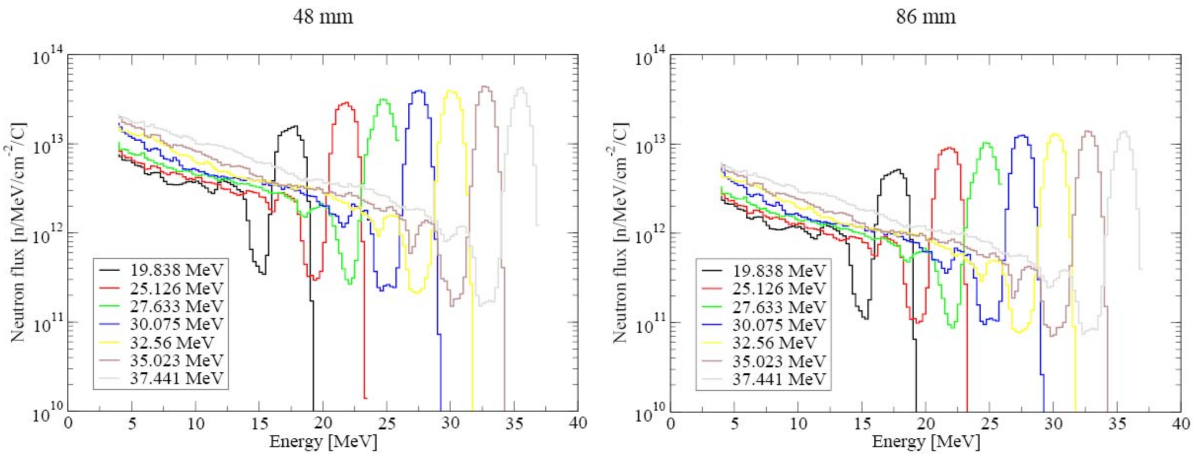

Figure 6: Neutron spectra obtained by modification of spectra from CYRIC experimental facility at the place of activation foils at 48 and 86 mm from the target front.

5. EXTRACTION OF ^{93}Nb AND ^{59}Co CROSS-SECTION CURVES WITH THE MODIFIED SAND-II CODE

5.1 Analysis of existing experimental data for ^{93}Nb and ^{59}Co cross-sections

The EXFOR database was searched for existing experimental cross-section data of neutron reactions on ^{93}Nb and ^{59}Co. The general conclusions for most materials can be applied also to these two isotopes:

- There are few cross-section measurements for higher order reactions, eg. (n,3n), (n,4n) ..
- There are few measurements above 20 MeV.

As an example, available experimental data together with the EAF evaluation is plotted for ^{92}Nb in Figure 7.

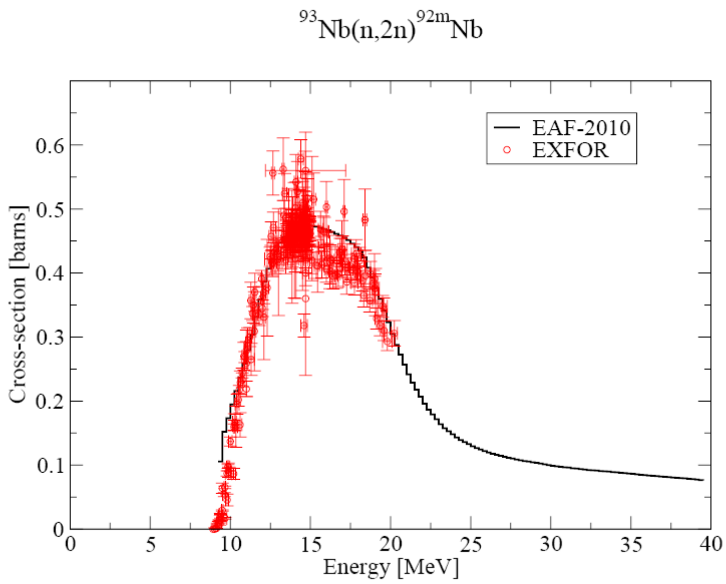

Figure 7: Available experimental data (EXFOR) with the EAF evaluation for the reaction ^{93}Nb(n,2n)^{92m}Nb.

5.2 Extraction procedure

The Li/C source neutron spectra calculated at the locations of the Nb and Co samples indicate that mono-energetic peaks account for 30-50% of the total flux, the rest being the low energy neutrons from the Li breakdown reaction and scattering on the target assembly.

To derive the activation cross-sections curves in such a complex neutron field a modified version [2,3] of the SAND-II [17] code was used. This code is traditionally used for the neutron spectrum adjustment. In the present case the procedure was reversed: the neutron spectra in the foil were supposed to be known and fixed, whereas the activation cross-section curve was allowed to vary to get the C/E ratio close to unity at all proton energies. In such a way the cross-section curves for the ^{93}Nb(n,*) and ^{59}Co(n,*) reactions were adjusted to the reaction product activities at the distances of 48.5 and 86.5 mm. These distances were used in the calculations, since the foils were placed ca 0.5 mm from the front wall of the foil holder (paper envelope, other foils in front of Nb and Co).

The EAF-2010 [14,15] evaluated data processed with PREPRO [16] tools and grouped in 0.25 MeV bins were used as input cross-sections for the SAND-II procedure. In the case of contributions of several reactions to the produced isotope, the corresponding cross-sections were extracted and summed. For the neutron spectra, both the spectra measured by Uwamino (extrapolated to our energies) and the spectra calculated with MCNPX were used.

The obtained cross-section curve exhibits a specific behaviour (see e. g. Figure 8):

- There are two different pairs of curves, one for cross-sections extracted with extrapolated Uwamino spectra and another one for cross-sections extrapolated with MCNPX simulated spectra. The differences are up to 30% (see Table 4). The differences between results extracted from different foil positions (48.5 mm and 86.5 mm) are minimal.

- The cross-section curve is in general non-physical, there are several regions showing a discontinuous increase or decrease of the cross-section. The width of these regions corresponds to the width of the quasi-monoenergetic peaks.
- The SAND-II code modifies the cross-section curve below the lowest quasi-mono-energetic peak.

The final experimental data for the measured cross-sections were obtained from the produced curves by taking the average value in the regions determined by the quasi-mono-energetic peaks (see Section 7). Another analysis of the measured cross-sections is presented in [18,19]. In those works, another method of the cross-section extraction and slightly different neutron spectra were used. However, the results of both methods are close to each other.

5.3 ^{93}Nb cross-sections

The (n,2n) reaction on ^{93}Nb results in the population of several states in ^{92}Nb nuclei, long lived are the ground state (denoted as g), and the first (denoted as m) isomeric state. The isomeric state m has a shorter half-life (10.15 d) and was reliably measured. The cross-section for the ^{93}Nb(n,2n)^{92m}Nb reaction is shown in Figure 8 together with evaluated data from EAF-2010 and IRDF-2002 [21]. With the exception of the measurements at the lowest and the highest energies of the proton beam, the present measurements agree with EAF-2010/IRDF-2002 evaluated data.

In the case of the (n,3n) reaction on ^{93}Nb, the decay of the first isomeric state with the half-life 60.86 d was measured. The cross-section for ^{93}Nb(n,3n)^{91m}Nb reaction is also displayed in Figure 8 together with evaluated data from EAF-2010. The overestimation of the EAF-2010 evaluation is significant. The disagreement can be explained with the database uncertainties of the ^{91m}Nb gamma lines. The database intensities of the most visible gamma line is 2.9%(LUND) and 2.02% (ENSDF [22]). The values calculated with the intensity from ENSDF are ca 1.5 times higher and agree within uncertainties with the EAF evaluation. A similar behaviour is exhibited by the measured values for the cross-section of the ^{93}Nb(n,*)^{91m}Y reaction (the sum of the (n,n2p), (n,^{3}He), and (n,pd) reaction cross-sections), see Figure 9. In this case, the gamma intensities from LUND and ENSDF databases agree.

The cross-sections for other reactions that could be measured on ^{93}Nb were in agreement with the EAF-2010. The cross-section for the reaction ^{93}Nb(n,4n)^{90}Nb is shown in Figure 9. The cross-section for the reaction ^{93}Nb(n,*)^{90m}Y includes the reactions (n,n^{3}He), (n,α), (n,pt), (n,npd), and (n,2n2p), the cross-section for the reaction ^{93}Nb(n,*)^{88}Y is the sum of (n,2nα), (n,3n^{3}He), (n,ndt), and (n,4n2p) cross-sections. Figure 10 shows both obtained cross-section curves.

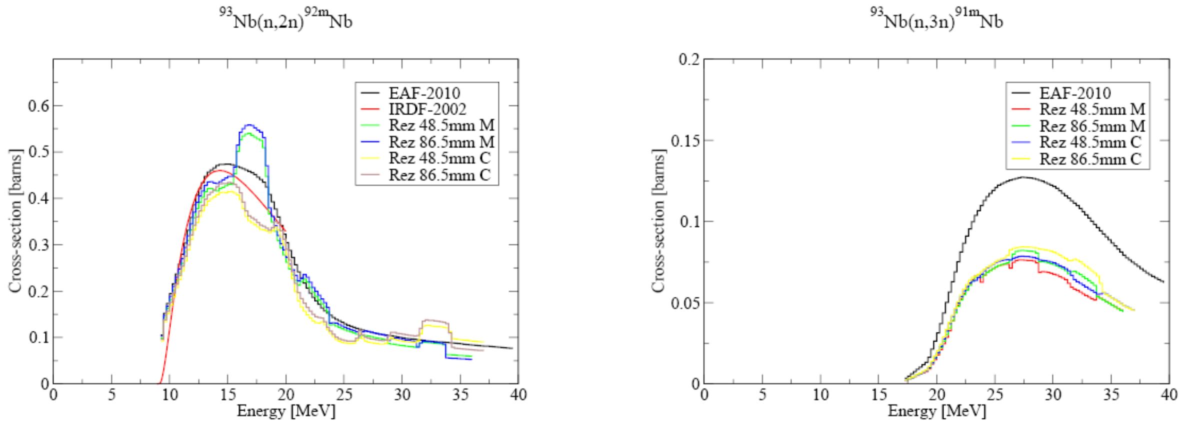

Figure 8: The cross-section curves for the ^{93}Nb(n,2n)^{92m}Nb and ^{93}Nb(n,3n)^{91m}Nb reactions extracted with SAND-II and comparison with evaluated data of EAF-2010 and IRDF-2002. Cross-sections marked with "M" are extracted with MCNPX spectra, those with "C" are extracted with CYRIC extrapolated spectra.

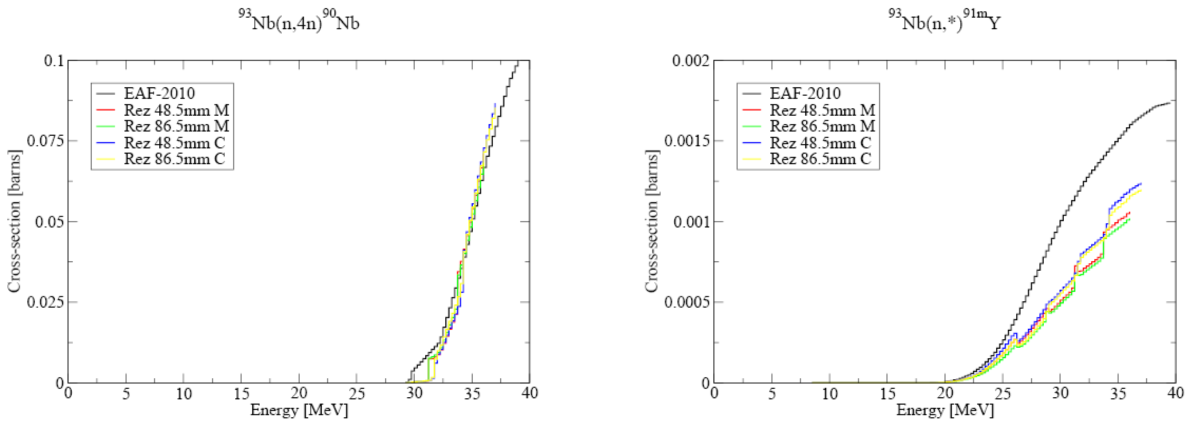

Figure 9: The cross-section curves for the ^{93}Nb(n,4n)^{90}Nb and ^{93}Nb(n,*)^{91m}Y reactions extracted with SAND-II and comparison with evaluated data of EAF-2010 and IRDF-2002. Cross-sections marked with "M" are extracted with MCNPX spectra, those with "C" are extracted with CYRIC extrapolated spectra.

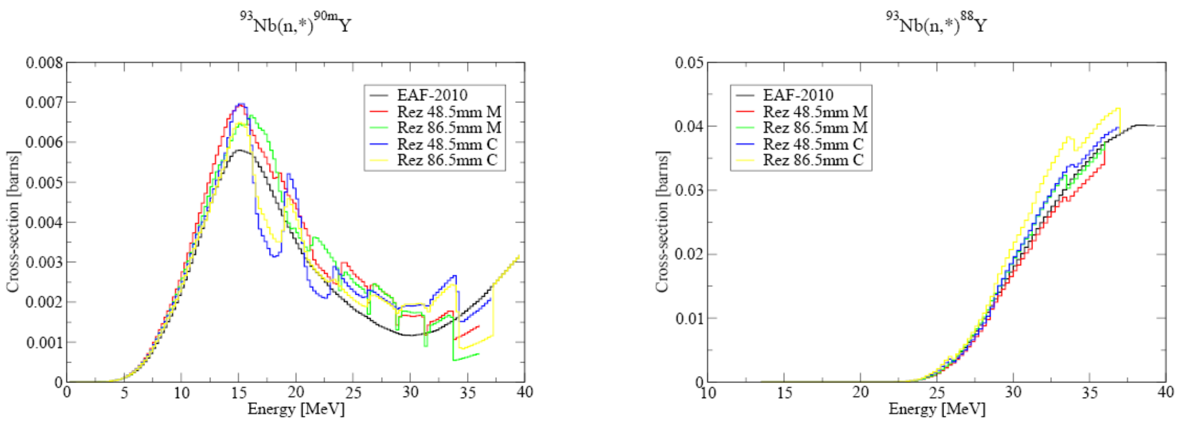

Figure 10: The cross-section curves for the ^{93}Nb(n,*)^{90m}Y and ^{93}Nb(n,*)^{88}Y reactions extracted with SAND-II and comparison with evaluated data of EAF-2010 and IRDF-2002. Cross-sections marked with "M" are extracted with MCNPX spectra, those with "C" are extracted with CYRIC extrapolated spectra.

5.4 ^{59}Co cross-sections

In the reaction $^{59}Co(n,2n)^{58}Co$ two isotopes of ^{58}Co with different decay times are produced. The sum of cross-sections and their comparison to EAF-2010 and IRDF-2002 databases is shown in Figure 11 together with the cross-sections for the $^{59}Co(n,3n)^{57}Co$ reaction. The data obtained for $^{59}Co(n,3n)^{57}Co$ reaction show a slight underestimation of the EAF-2010 evaluation.

Only two data points measured for the reaction $^{59}Co(n,4n)^{56}Co$ show an overestimation of the EAF-2010 evaluation. The data measured for the reaction $^{59}Co(n,p)^{59}Fe$ are in good agreement with EAF-2010. Significant disagreement are seen only at the lowest (MCNPX spectra) and highest (extrapolated Uwamino spectra) proton energies, Figure 12.

The cross-section for $^{59}Co(n,*)^{56}Mn$ is the sum of the (n,n^{3}He), (n,α), (n,pt), (n,npd), and (n,2n2p) cross-sections and $^{59}Co(n,*)^{54}Mn$ reaction includes (n,2nα), (n,3n^{3}He), (n,ndt), and (n,4n2p) reaction channels. In the comparison with EAF-2010, (Figure 13) it can be seen that the values for $^{59}Co(n,*)^{56}Mn$ agree with EAF-2010 evaluation and the measured values for $^{59}Co(n,*)^{54}Mn$ are underestimated by the EAF-2010.

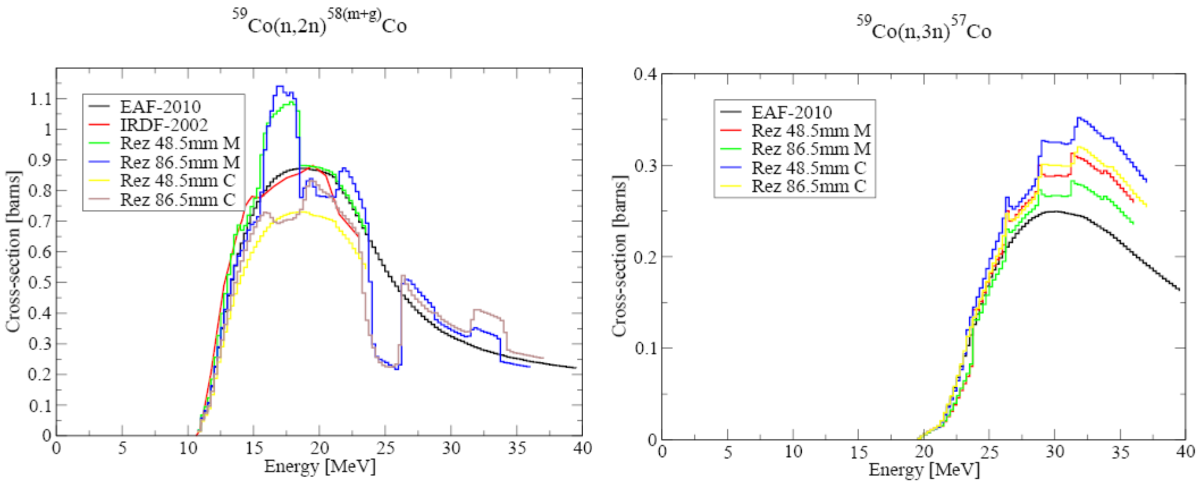

Figure 11: The cross-section curves for the $^{59}Co(n,2n)^{58(m+g)}Co$ and $^{59}Co(n,3n)^{57}Co$ reactions extracted with SAND-II and comparison with evaluated data of EAF-2010 and IRDF-2002. Cross-sections marked with "M" are extracted with MCNPX spectra, those with "C" are extracted with CYRIC extrapolated spectra.

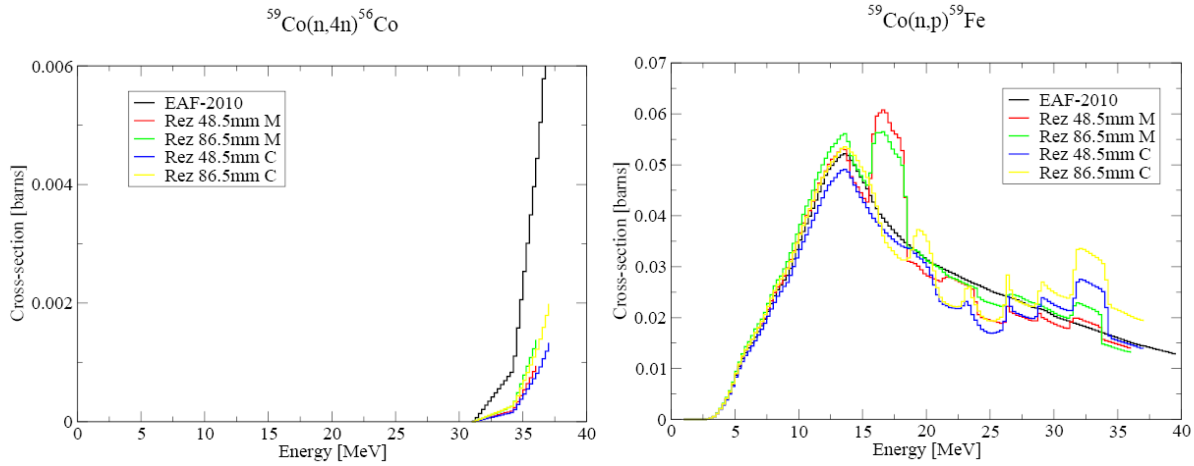

Figure 12: The cross-section curves for the $^{59}Co(n,4n)^{56}Co$ and $^{59}Co(n,p)^{59}Fe$ reactions extracted with SAND-II and comparison with evaluated data of EAF-2010 and IRDF-2002. Cross-sections marked with "M" are extracted with MCNPX spectra, those with "C" are extracted with CYRIC extrapolated spectra.

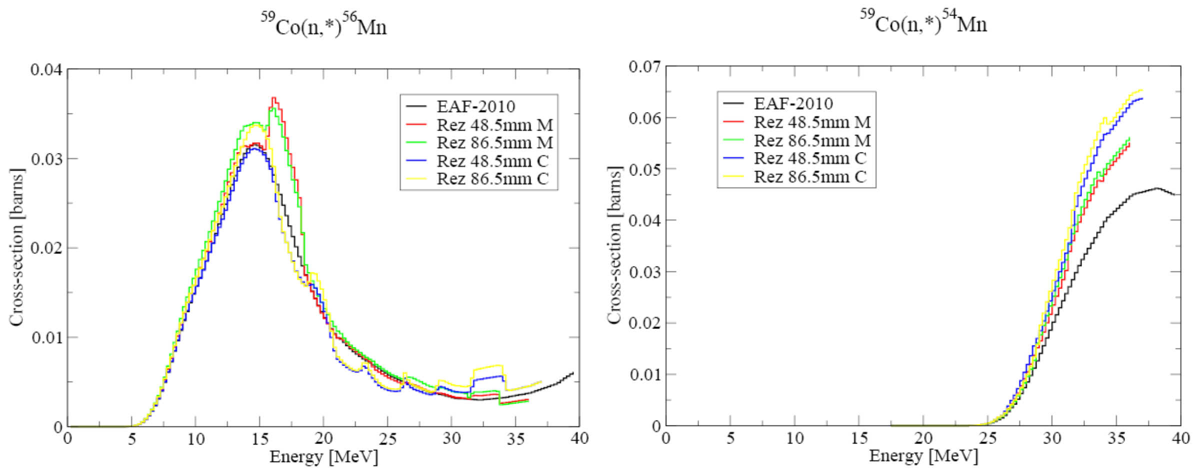

Figure 13: The cross-section curves for the $^{59}Co(n,*)^{56}Mn$ and $^{59}Co(n,*)^{54}Mn$ reactions extracted with SAND-II and comparison with evaluated data of EAF-2010 and IRDF-2002. Cross-sections marked with "M" are extracted with MCNPX spectra, those with "C" are extracted with CYRIC extrapolated spectra.

6. QUALITY ASSURANCE ANALYSIS

Before averaging the cross-section curves from the previous step, a detailed quality assurance analysis was performed. The goal was to reasonably set the uncertainties of the final results. For easier understanding, the uncertainties are divided in several categories.

6.1 Statistical uncertainties

Statistical uncertainties arise for the analysis of the gamma peaks and in the Monte Carlo simulations. The uncertainties of the gamma peak analysis are in the order of 1-2% (in some cases up to tens of percents), accurate values are given in Tables 1 and 2 (together with ca. 2% uncertainties of the detector efficiency). The number of histories used in the Monte Carlo simulations was always sufficient to keep the statistical uncertainties of the results below 1-2% (if the main part of the production is by low energy neutron background), usually below 0.5% (production by quasi monoenergetic neutrons). In comparison with other uncertainties the statistical uncertainties are the smallest contribution in most cases.

6.2 Systematic uncertainties

Systematic uncertainties cover a broad range of inaccuracies which are always present in the experimental work. During the irradiation, these are the parameters of the proton beam (energetic distribution, profile), the measurement of the total proton flux, the exact placement of the activation samples in relation to the target (accuracy 1 mm to target, <1 mm in relation to other activation samples),.. After the irradiation, the activity of the foils is measured by spectroscopy methods, and other systematic uncertainties because of positioning, calibration, etc. arise. These uncertainties apply to the experimentally measured reaction rates and are in the order of 10-15%.

6.2.1 Systematic uncertanties at irradiation

During the experiments, several parameters were controlled only up to certain accuracy. The impact of these parameters was studied with MCNPX simulations. The simulations of reaction rates with these parameters within the parameter accuracy were compared. The conclusions on the parameter impact were drawn on the basis of the calculated differences in reaction rates.

The energetic distribution of the cyclotron proton beam has approximately a Gaussian distribution with a FWHM of 200-300 keV (1.5%). To study if this affects our results, the neutron spectra were simulated with a mono-energetic proton beam and a proton beam with 300 keV FWHM. The reaction rates were calculated with cross-sections from EAF-2010. The differences were in most cases below 0.5%. This shows that the impact of beam distribution on energetic scale is negligible.

The displacements of the beam on the target has an observable impact on the results: a displacement of 3 mm from the target center affects the reaction rates (reaction rates decrease) up to 5%. Since the displacements are well controlled in our experiments (< 1 mm), we consider that the inaccuracy originating from this kind of displacement is negligible.

The spatial beam profile has limited impact. The difference in the activity the with a point beam and a Gaussian beam with 3 mm FWHM can be maximally 2.5%. The studies of the irradiation spots on Li foils after the irradiation show that beams in all

experiments had a well defined shape (Gaussian with 3 mm FWHM). Thus the uncertainties originating from the beam profile can be neglected.
The displacement of the samples in relation to the target impacts significantly the reaction rates: a displacement of 0.5 mm further from the target means 2% lower reaction rates at 48 mm and 1% lower reaction rates at 86 mm.

The decrease of the foil activation because of shielding of the neutrons by other foils was also studied. Two simulations were performed, one with the positions for foils left empty and the other one with the foils at the positions, at the energies 19.838 MeV and 37.441 MeV (to cover the whole energy range of our experiments). The reaction rates with the foils in positions are decreased in relation to the reaction rates without foils. This decrease is slightly higher for lower beam energies. For the foils placed at the position 48 mm, this decrease is negligible (0.3-0.5%). For the foils at the position 86 mm it is 2-3%.

At each irradiation a new Li target was used. The thickness of the targets is guaranteed to be the same within 5%. The simulations show that the target thickness in the range of these 5% would affect linearly the reaction rates (thickness ~ neutrons ~ 1/RR). Our reaction rates are normalized to the measurement of the proton current and not on the direct measurement of the neutron yield from the target. The uncertainty of the reaction rates due to the target thickness uncertainty is 5%.

The target construction block is designed as an alcohol-cooled Faraday cup in order to measure the proton beam current passing through the Li-foil target ended by the carbon disc. The tests of the beam charge monitor operation the comparative measurements are carried out repeatedly and results are found to be consistent within 5% of accuracy.

Summing up all important contributions of uncertainties - positioning of the foils, Li target thickness, current measurement - the total uncertainty originating from the irradiation part is 10-15%.

6.2.2 Systematic uncertanties at activity measurements

The gamma activities of irradiated samples are investigated using gamma-spectrometry sets based on the High-Purity Germanium detectors, relevant spectrometry electronics and data analysis software. The energy range of our apparatus is 30 keV - 3 MeV. Beside standard spectroscopy corrections (detector efficiency, decay during the irradiation, measurement dead time, cascade coefficients,..), the corrections for self-absorption of gammas in the samples, and geometrical factors for non-pointlike samples were calculated if necessary. The main source of uncertainties originates from the detector efficiency curve, the uncertainty estimate is ca. 2%. The positioning of the samples was secured in the way that the uncertainties were below 1%.

6.3 Cross-section extraction uncertainties

Further uncertainties arise from the method applied for the extraction of cross-sections from the reaction rates. The activity (reaction rate) of the irradiated sample can be calculated as:

$$RR = C \cdot \int_{0}^{E_p} \Phi(E)\sigma(E)dE,$$

where C is a normalization constant, $\Phi(E)$ is the neutron flux, and $\sigma(E)$ is the cross-section for the reaction producing the considered isotope (the sum of all possible reactions). In order to reverse this calculation and to obtain the approximation of the cross-section from the measured reaction rate, different methods can be applied. These methods can give results that significantly differ from each other. In this work the method of the SAND-II code was used, in [18,19] another method ("recommended" by IAEA [20]) is used and gives similar results.

One of the input parameters of these methods is the neutron spectrum $\Phi(E)$ which is not well known. Either the spectra from the MCNPX simulations or the CYRIC extrapolation can be used. The calculated spectra cannot reproduce the experimental CYRIC spectra with an accuracy better than 10-15% but include several construction details of the NPI/Řež facility which become important close to the target. By comparing the cross-section curves obtained with MCNPX and extrapolated CYRIC spectra (Figures 8-13), the reader can get a picture of the differences. It is also possible to compare the reaction rates calculated with MCNPX and extrapolated CYRIC spectra, as seen in Table 4. The numbers from this Table (in most cases up to 15%) were added to statistical (1-2%) and systematic uncertainties (10-15%) to obtain the total uncertainty of extracted cross-section values.

The neutron spectra measured at the CYRIC facility indicate that the beam energies given by the authors are accurate up to 200-300 keV. With simulations of reaction rates for proton energies 300 keV apart, it was found that the reaction rates change for ca. 5%. These extra 5% impact only the cross-sections extracted with the use of extrapolated Uwamino spectra.

Ep[MeV]	^{92m}Nb	^{91m}Nb	^{90}Nb	^{91m}Y	^{90m}Y	^{88}Y	^{58}Co	^{57}Co	^{56}Co	^{59}Fe	^{56}Mn	^{54}Mn
19.838	31%	57%		68%	28%		33%			26%	26%	
25.126	24%	-3%		-43%	21%		22%	-46%		16%	20%	
27.633	17%	-4%		-17%	15%	-35%	12%	-10%		10%	17%	-47%
30.075	13%	-3%		-5%	12%	-6%	12%	-5%		6%	12%	-5%
32.56	5%	-10%	-5%	-13%	4%	-12%	7%	-12%	89%	-2%	3%	-12%
35.023	-3%	-10%	-2%	-14%	-5%	-13%	2%	-13%	-2%	-9%	-7%	-14%
37.441	-1%	-8%	-2%	-12%	-3%	-12%	4%	-13%	14%	-8%	-4%	-14%

Table 4: Differences in activities produced by MCNPX and extrapolated CYRIC spectra.

7. FINAL RESULTS AND COMPARISON TO EAF-2010 DATA

The experimental cross-section results were extracted from the curves produced by the SAND-II code. In these curves, the regions determined by the mono-energetic peaks are clearly seen, and the cross-sections in these regions were determined by the equations:

$$\langle\sigma\rangle = \frac{\int \sigma dE}{\int dE}, \langle E\rangle = \frac{\int E dE}{\int dE},$$

where <σ> is the derived cross-section at the mean energy in the region <E>. The mean energy is set in the center of the energy interval.

Four values were extracted for each energy (distances 48.5 and 86.5 mm, MCNPX and Uwamino spectra) and averaged to obtain the final value. The widths of the regions (1-2 MeV) are used as the uncertainties of the results on the energy scale. The uncertainties on the cross-section scale are quadratic sums of uncertainties originating from the irradiation (10-15%), activity measurement (few percents), and cross-section extraction procedure (up to 15% + extra 5% for Uwamino spectra). The results for all isotopes are shown in Figures 14-19.

With some exceptions, the obtained experimental results are in general in some agreement with the EAF-2010 data. In the case of ^{93}Nb, the experimental points for reactions (n,2n) and (n,4n) reactions agree very well with the EAF-2010, while the points for reaction (n,3n) show a significant discrepancy (~2x). Partially this can be explained with the uncertainty in the intensity of ^{91m}Nb gamma line as mentioned in section 5.3. A similar disagreement is observed for the reaction ^{93}Nb(n,*)^{91m}Y. There is, however, no indication that the disagreement comes from the experimental procedure. The small disagreement of the experimental data points and the EAF-2010 curve for the reaction ^{93}Nb(n,*)^{90m}Y, on the other hand, is considered more incidental since a significant part of the activity is produced by neutrons originating from the C beam dump, which are modelled with worse accuracy.

In the case of ^{59}Co, the important experimental points which show discrepancy from EAF-2010 are those for the reactions ^{59}Co(n,3n)^{57}Co and ^{59}Co(n,*)^{54}Mn. The reason for these discrepancies traced to the experimental procedure. It is thus assumed that EAF-2010 underestimates the cross-sections for these reactions. Other occasional disagreements for other reactions (e. g. n,2n cross-section at 25 MeV) can be due both to deficient experimental or evaluated data.

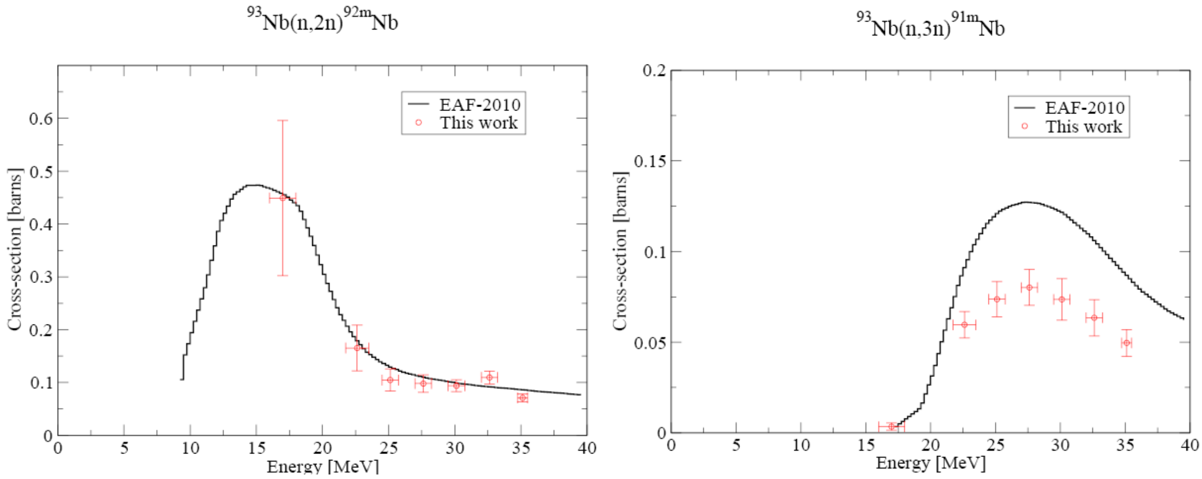

Figure 14: Comparisons of activation cross-sections extracted from NPI/Řež experimental data and evaluated EAF-2010 data.

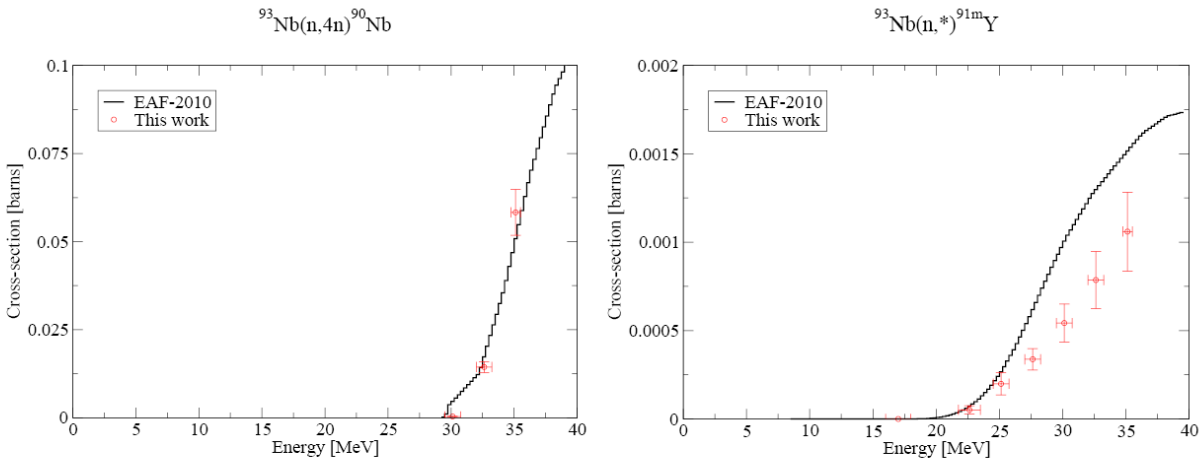

Figure 15: T Comparisons of activation cross-sections extracted from NPI/Řež experimental data and evaluated EAF-2010 data.

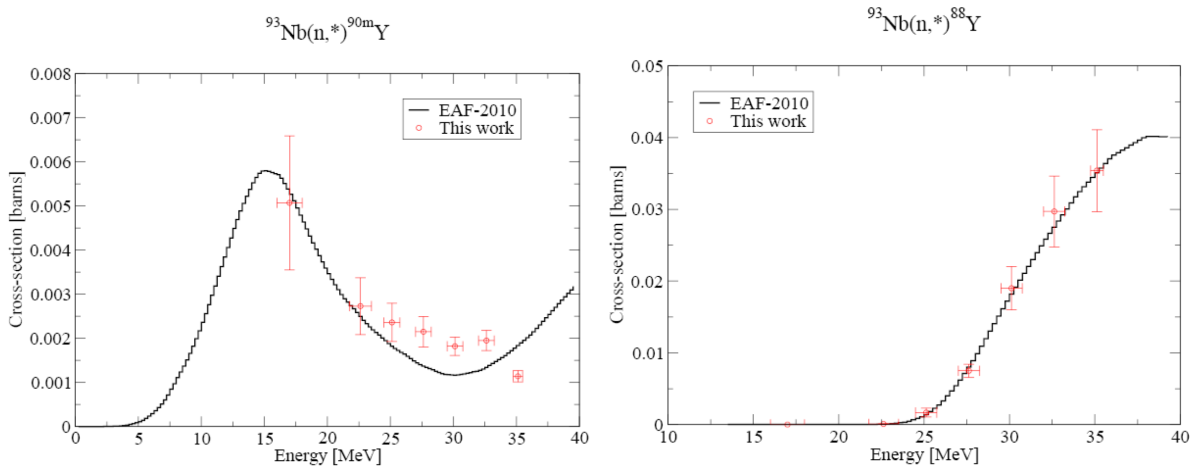

Figure 16: Comparisons of activation cross-sections extracted from NPI/Řež experimental data and evaluated EAF-2010 data.

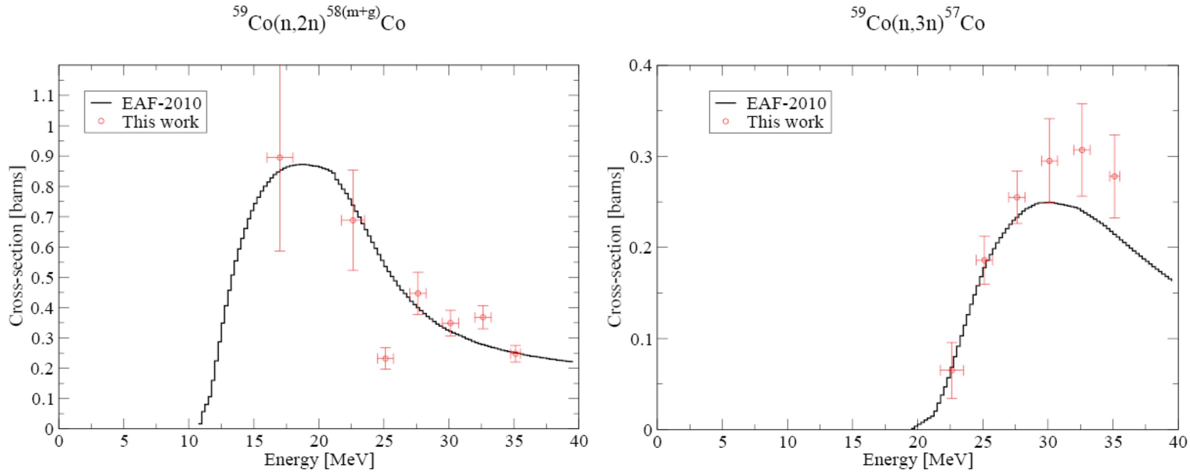

Figure 17: Comparisons of activation cross-sections extracted from NPI/Řež experimental data and evaluated EAF-2010 data.

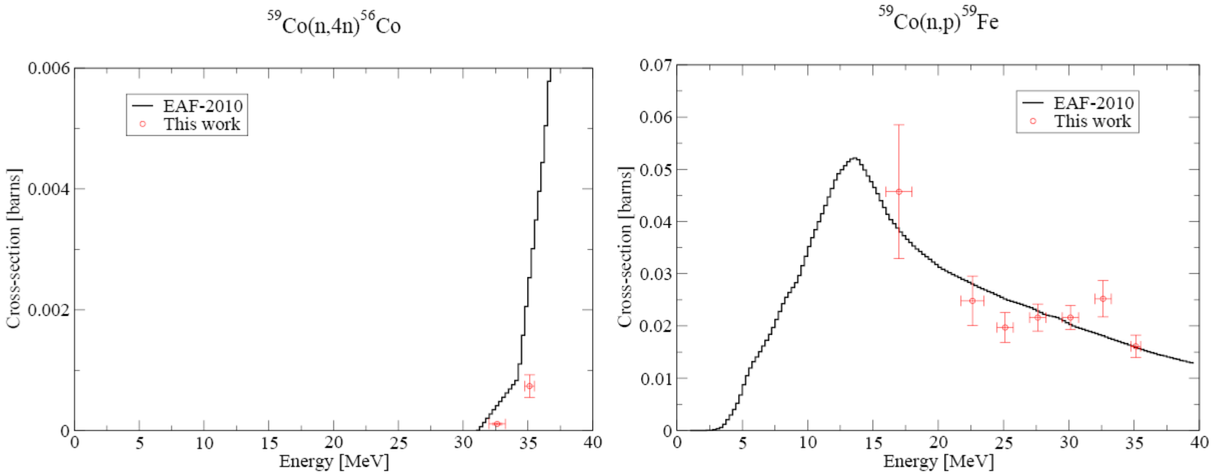

Figure 18: Comparisons of activation cross-sections extracted from NPI/Řež experimental data and evaluated EAF-2010 data.

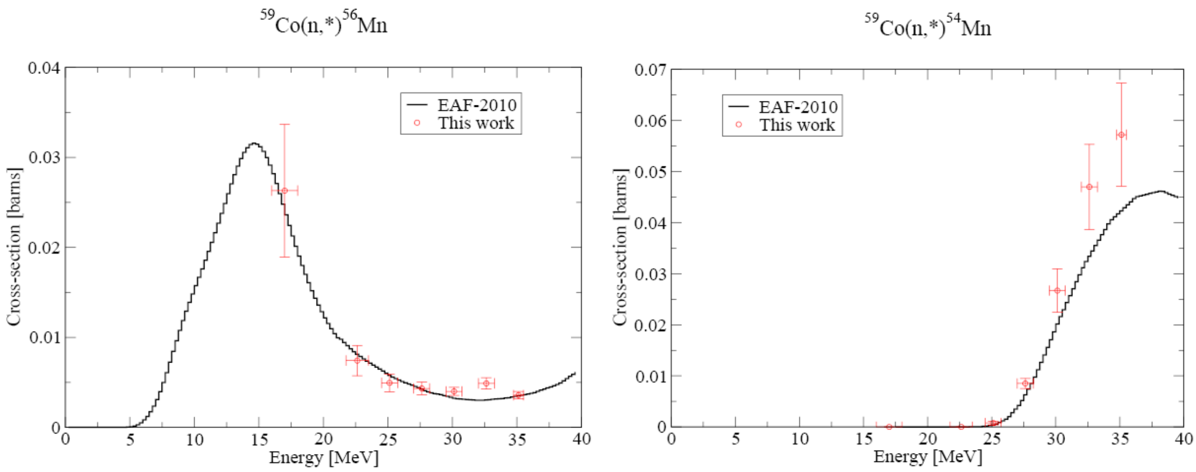

Figure 19: Comparisons of activation cross-sections extracted from NPI/Řež experimental data and evaluated EAF-2010 data.

8. CONCLUSION

Task 4.2 of F4E-2010-F4E-GRT-056 (ES-AC), Action 2, was completed successfully. In the pre-analysis part of the task, a detailed investigation of the p+Li/C neutron source was performed. Systematic discrepancies in the results of previous tasks were uncovered and resolved. In the experimental part, seven irradiations of Nb and Co samples were performed with quasi-mono-energetic neutrons in energy the range 20-37.5 MeV, and the produced activities were measured. The post-analysis included the extraction of the cross-sections from measured activities using an ad-hoc modified version of the SAND-II code, and subsequent comparisons of the extracted cross-sections with EAF-2010 data. Most of the experimental data points obtained as part of this task confirm the validity of the EAF-2010 data. A few reaction cross-sections of the EAF-2010 activation data base, however, need to be revised.

9. ACKNOWLEDGEMENT

This work was supported by Fusion for Energy under the grant contract no. F4E-2010-GRT-056-02 (ES-AC). The views and opinions expressed herein reflect only the author's views. Fusion for Energy is not liable for any use that may be made of the information contained therein

10. INTELLECTUAL PROPERTY RIGHT (IPR) STATEMENT

Experimental cross-section data were generated for neutron induced reactions on ^{59}Co and ^{93}Nb in the range between 20 and 35 MeV using a ^{7}Li(p,xn) quasi-mono-energetic neutron source and an ad-hoc modified version of the SAND-II code for extracting the cross-sections from measured reaction rates.

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