

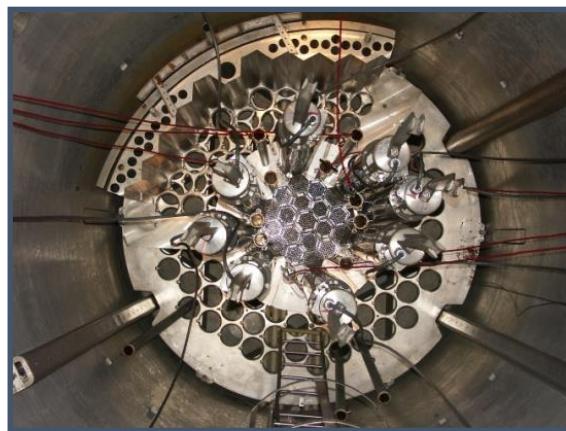
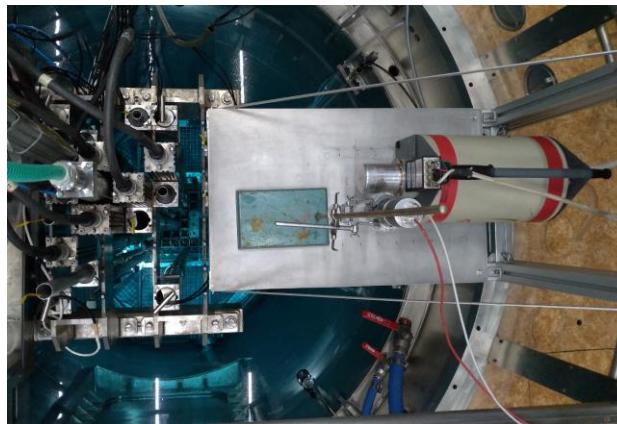
Rez contribution to inden: New copper broomstick and Teflon leakage experiments

Michal Kostal

INDEN 2024

Vienna, December 18th 2024

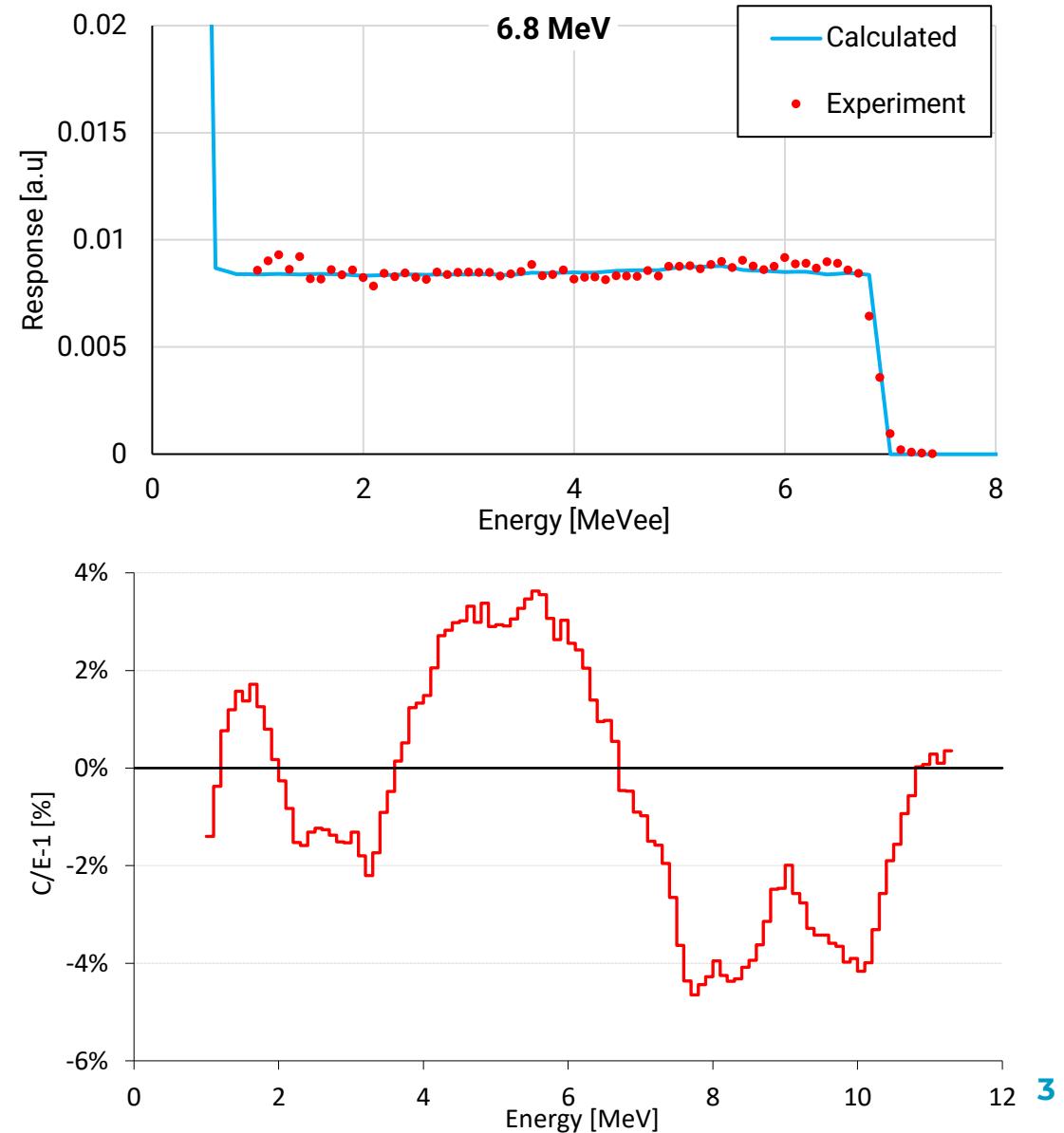
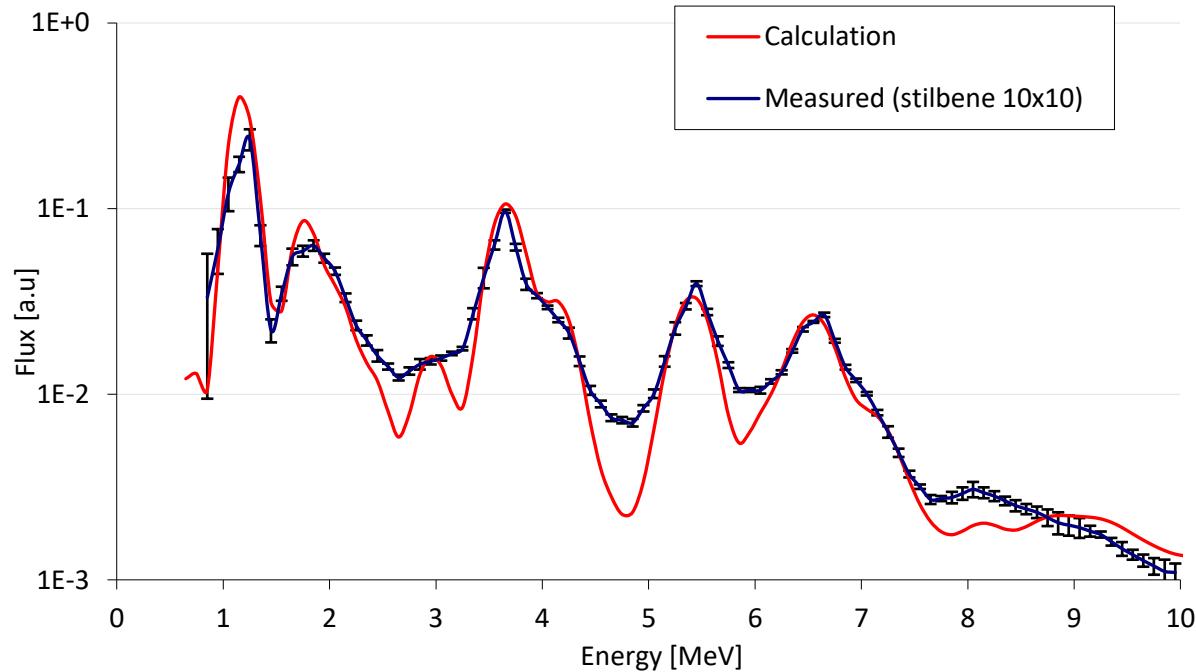
Research infrastructure used by CVR group in experiments



- **4 research reactors LR-0, LVR-15, VR-1 (university, CTU Prague), AKR-2 (TU Dresden)**
- **^{252}Cf neutron sources**
 - Material blocks for neutron leakage measurements:
 - spheres (Fe, Ni ... and others)
 - cubes (CF_2 , Al, stainless steel, Cu, graphite ...)
 - Spherical tank with aqueous solutions for prompt gamma leakage measurements
 - NaCl , MnSO_4 , FeSO_4 , , planned $\text{CO}(\text{NH}_2)_2$ (ad blue) , and many others
- **DT generators**
- **Well-described AmBe**
- **XRF analyzers**
- **(and many others)**

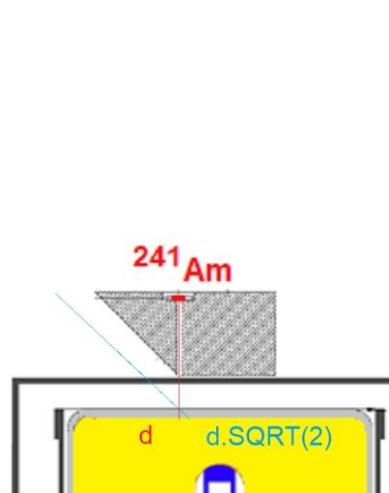
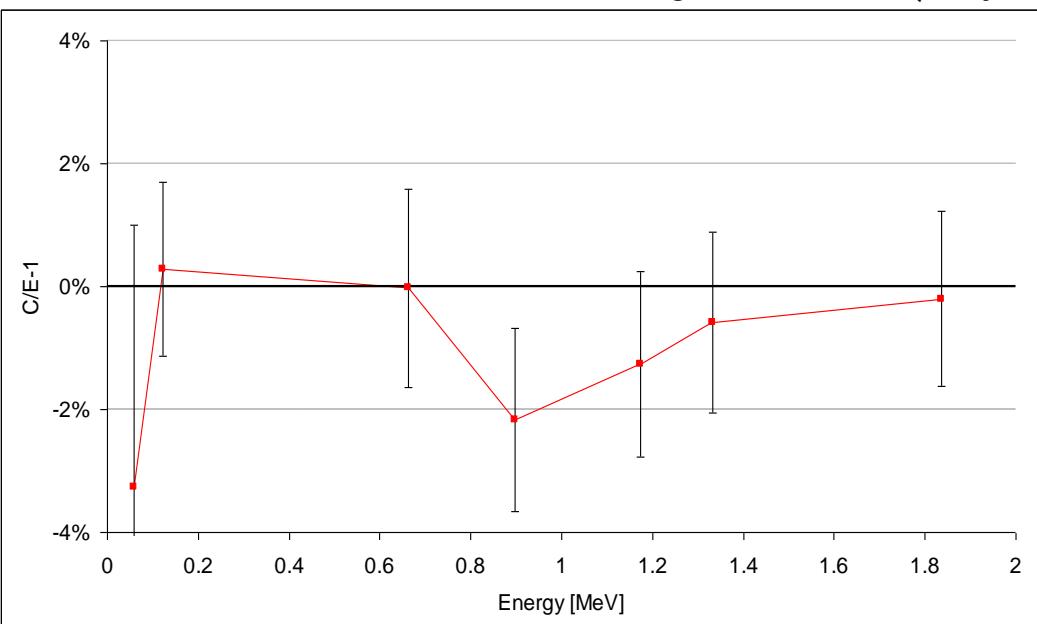
Neutron spectrometry

- Well-characterized spectrometer
- Validation in PTB
- Validation in ^{252}Cf
- Verified in Si filtered spectrum

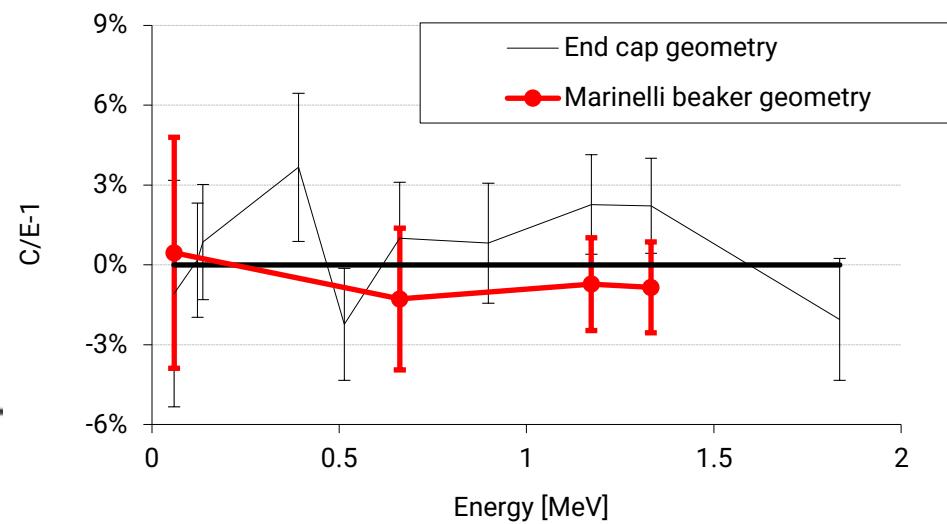
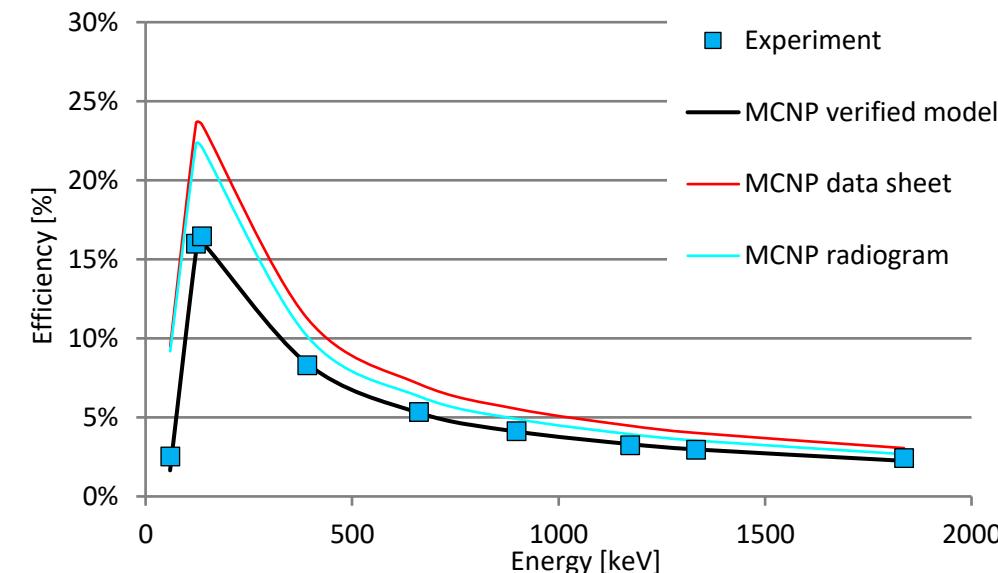


Gamma spectrometry - HPGe

- Most important is detector sensitivity
- Irradiated pins measurement
 - Mathematical model allows arbitrary axial power density profile
- Foil measurement
 - Mathematical model allows even large samples on detector cap
 - Determination of coincidence summing correction
- Gamma flux measurement
 - Model allows evaluation of gamma flux (only the

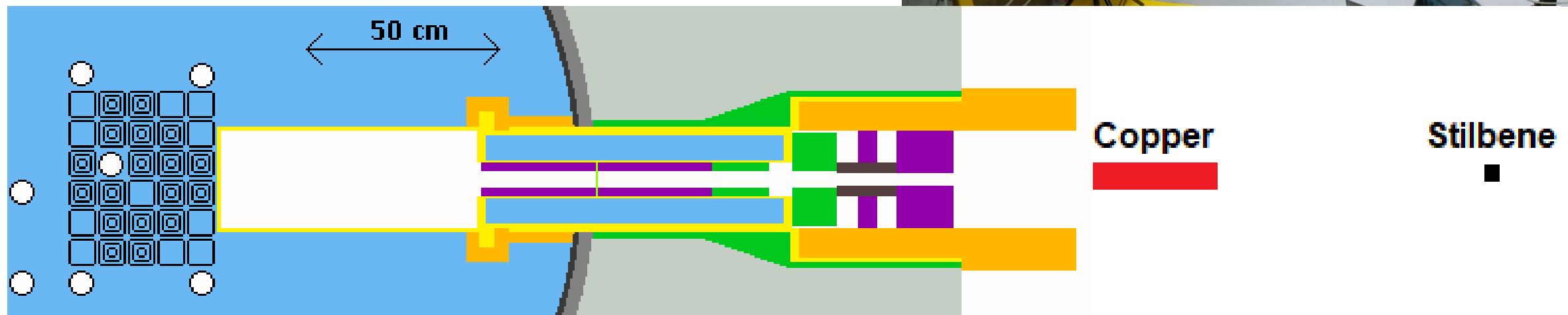


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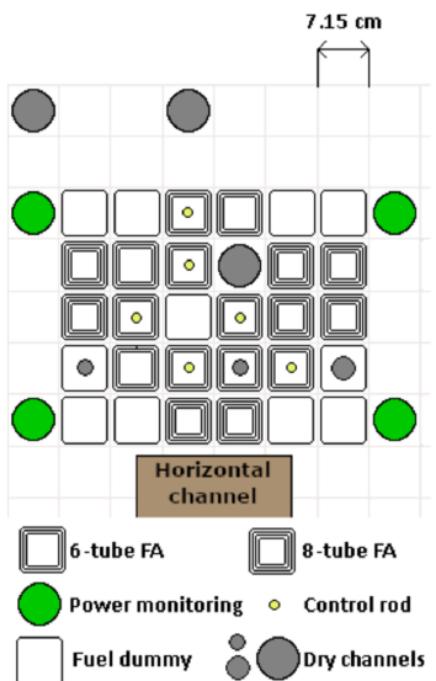
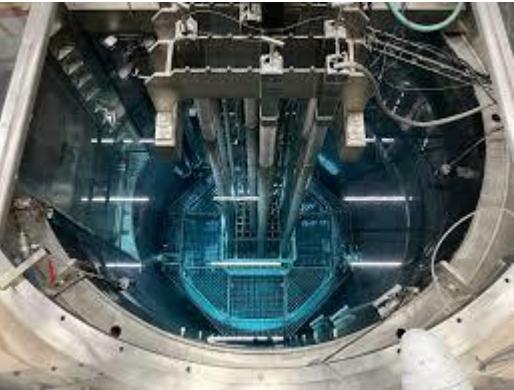
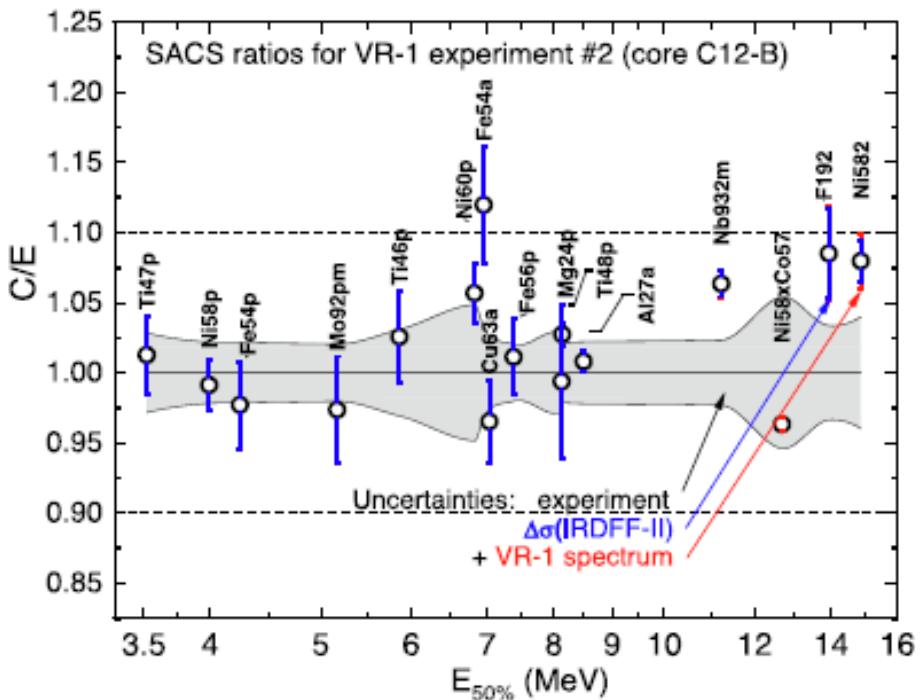
Broomstick experiment

- Integral quantities can usually be measured much more accurately than differential nuclear data,
 - Useful to use for tuning nuclear data evaluations to improve integral performance.
- Our experiment – measurement of spectra behind various thickness broomstick material
- The measured spectra are compared with calculation
- The experiment was realized in VR-1 reactor



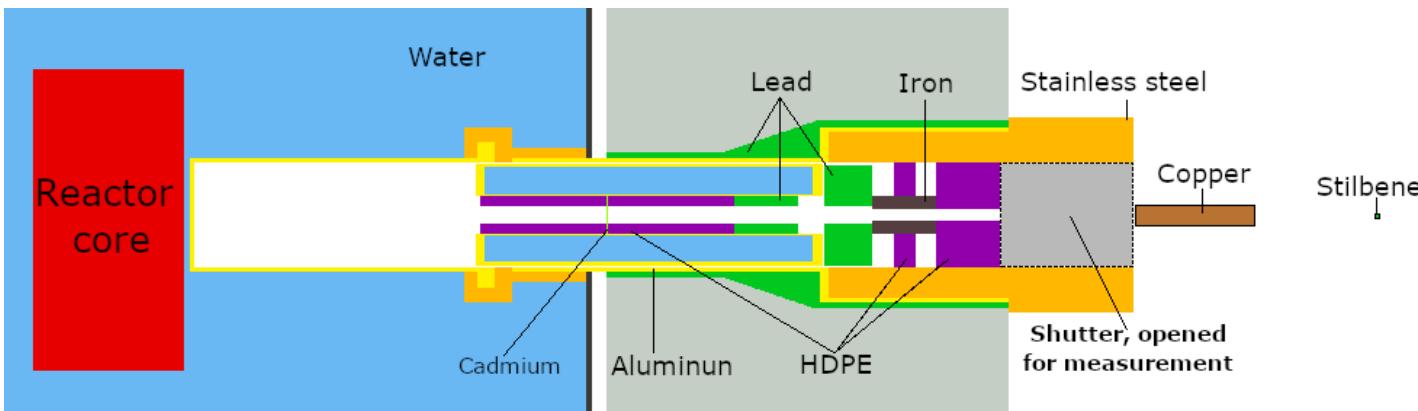
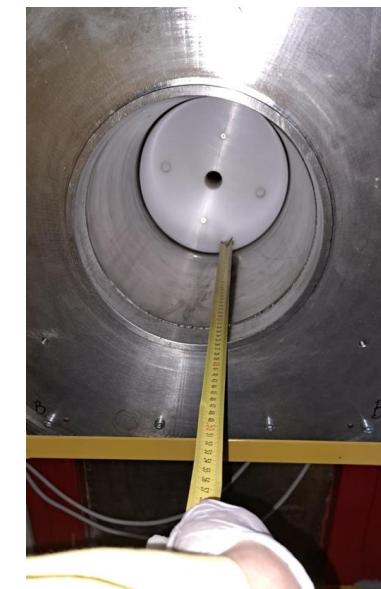
VR-1 reactor

- VR-1 university research reactor operated by CTU Prague
- Characterized neutron spectrum in radial channel
- Source spectrum known (large set of SACS measurements realized)



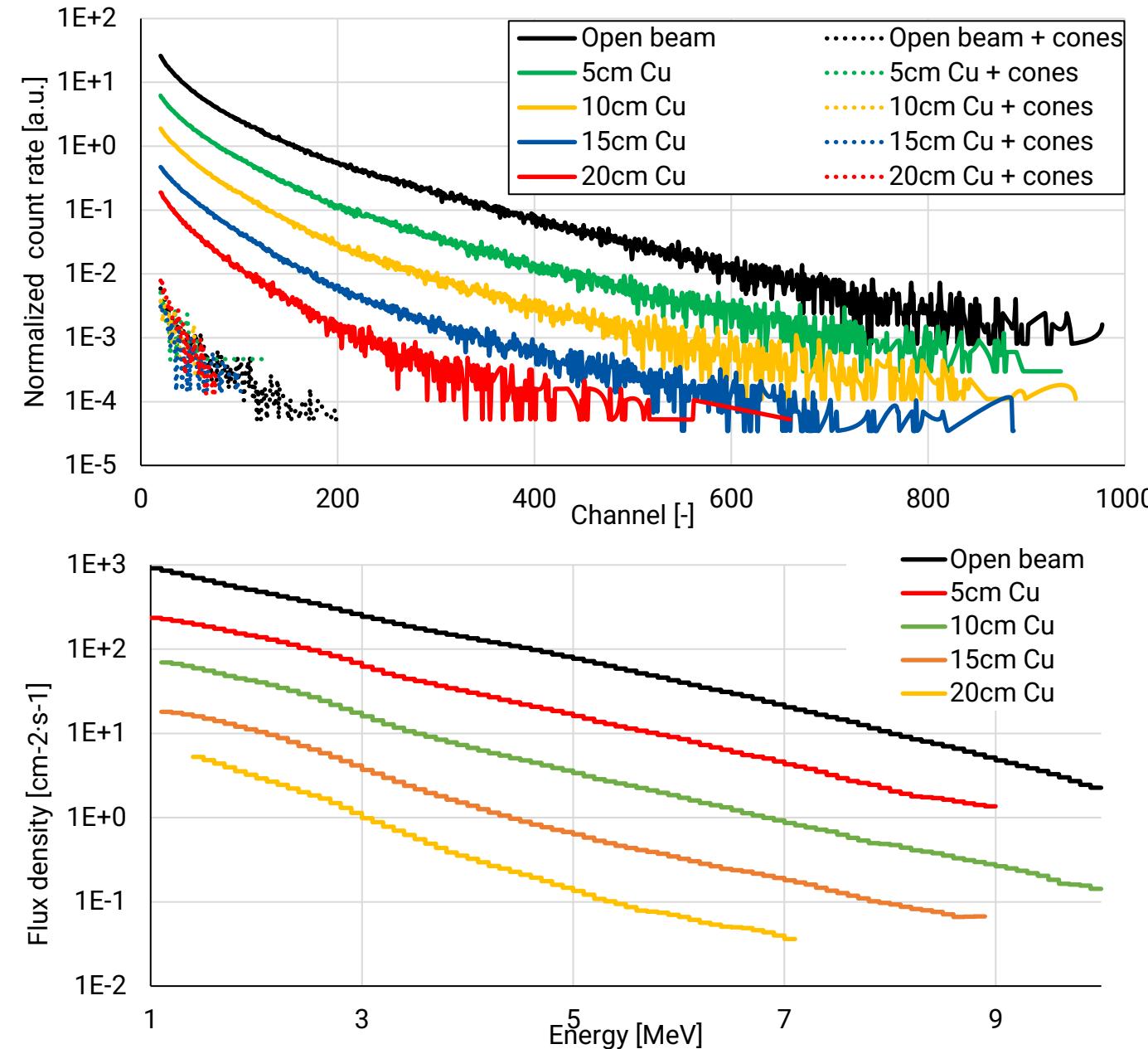
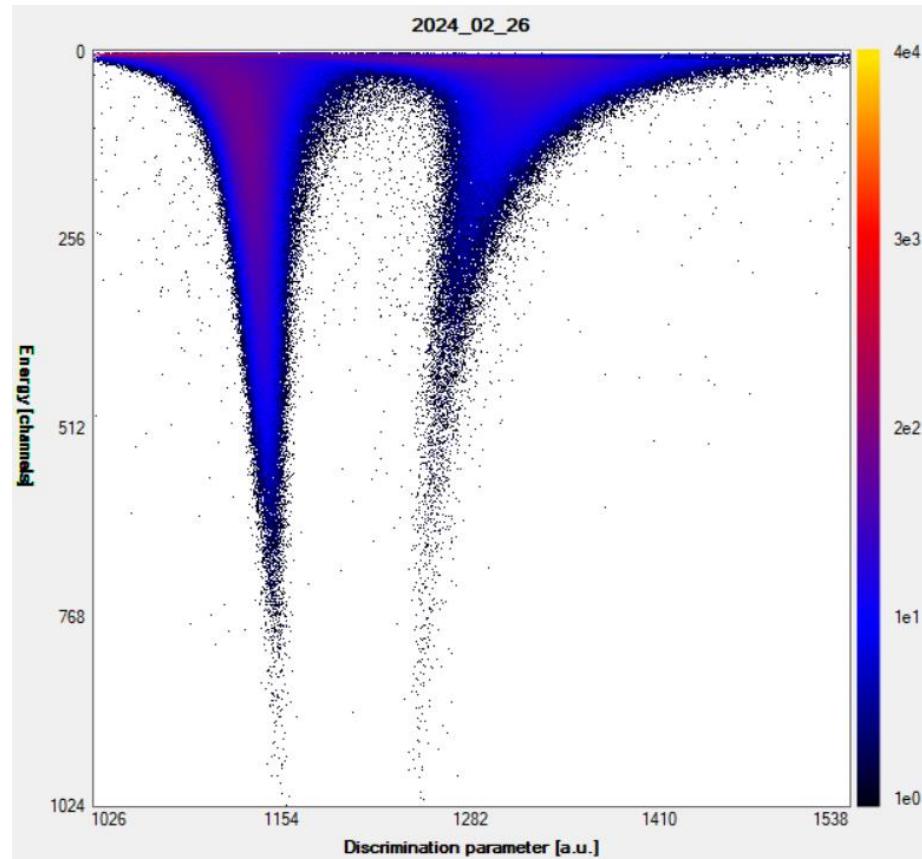
Development of pinhole beam

- Special insertion in VR-1 channel to collimate beam to diameter of 3 cm
- Large Pb block to suppress the gamma background
- Shutter to allow closing the beam without reactor shutdown



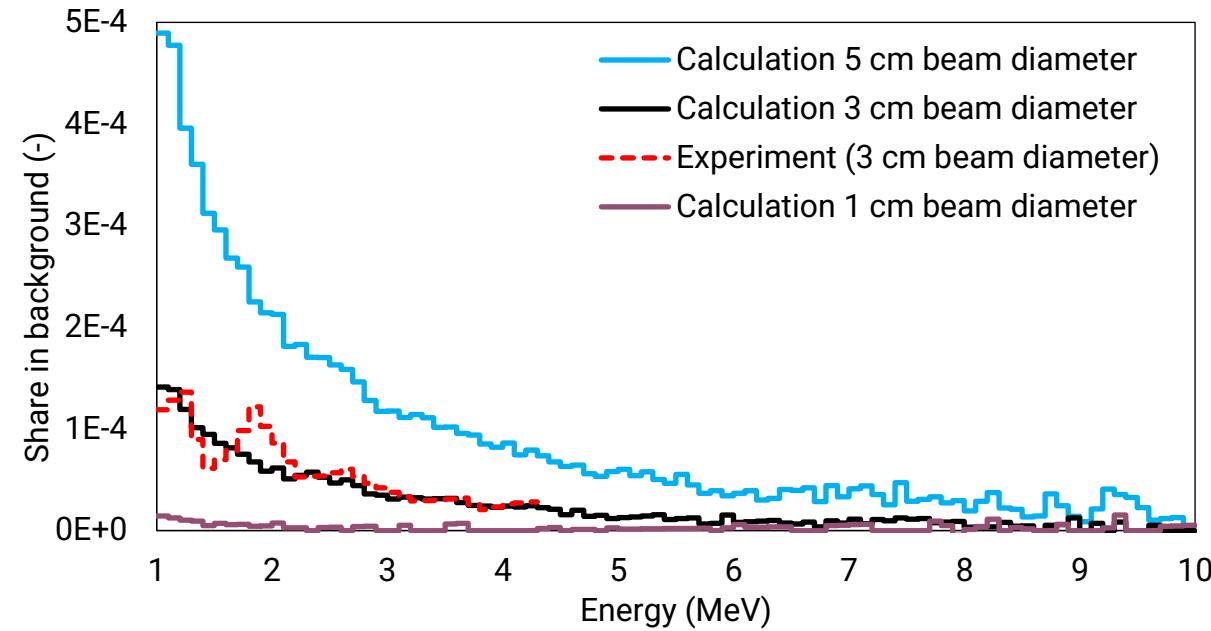
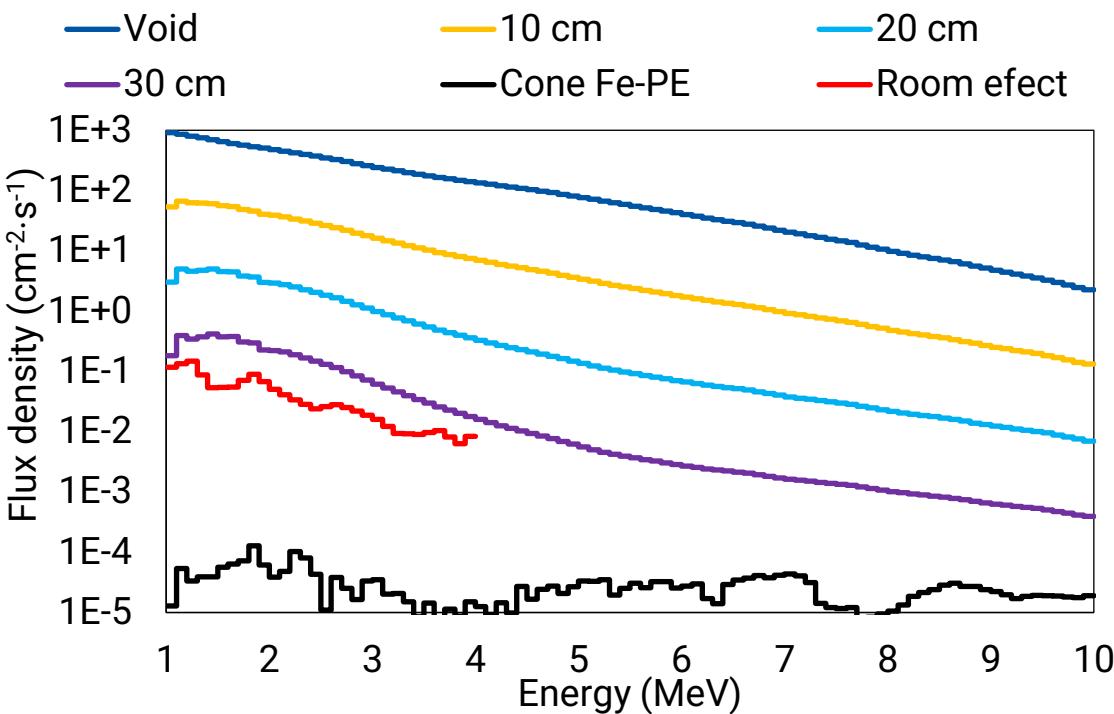
Spectra measurement

- Stilbene crystal 10 mm × 10 mm
- Good N/G separation
- Calibration using Cs (0.661 MeV) and H capture peak (2.23 MeV)



Background determination

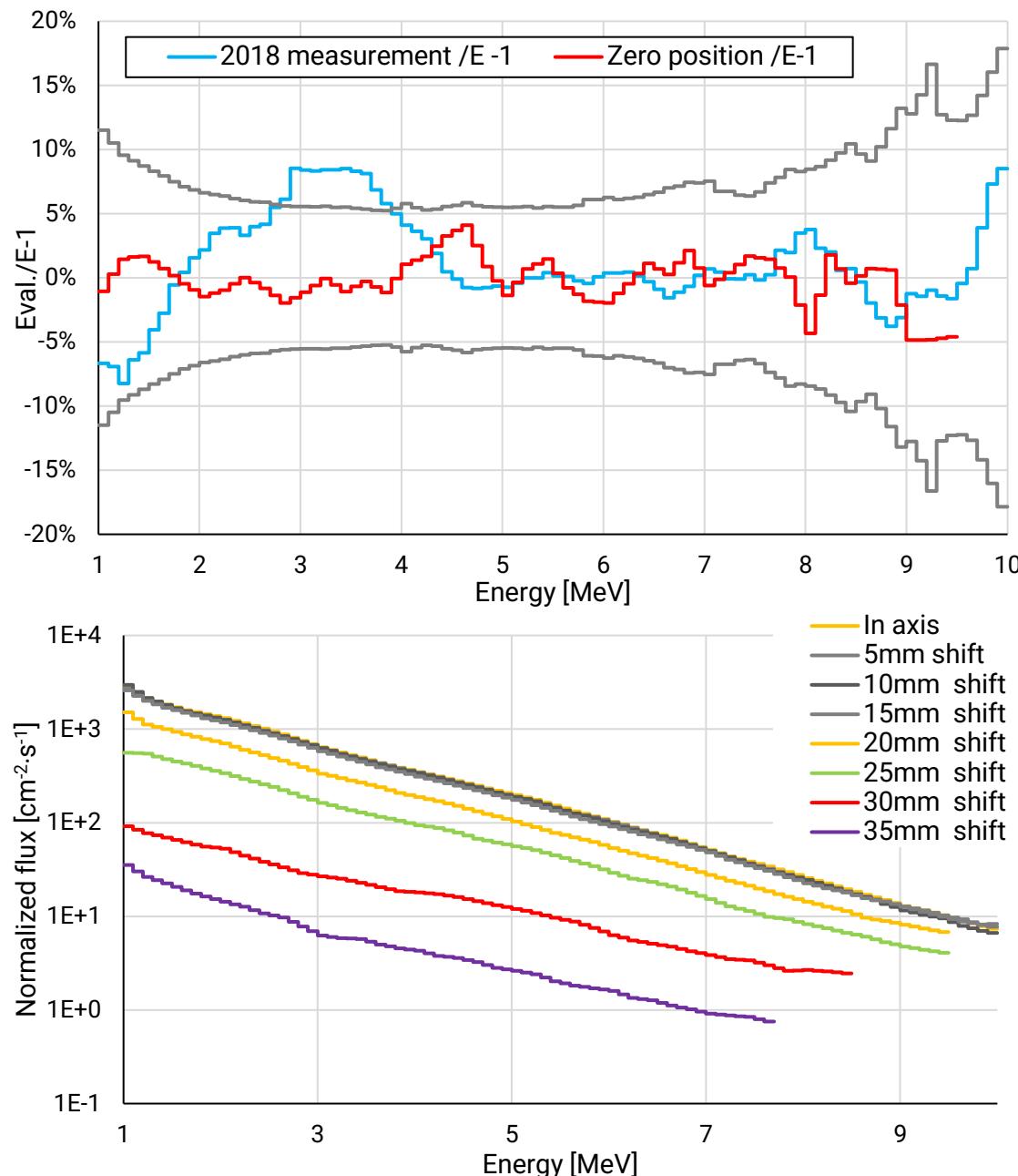
- Room effect determined using shielding cylinder
 - Iron + HDPE
- Its magnitude is affected by aperture diameter
- For blocks shorter than 20 cm Cu (attenuation ~ 1000) room effect is negligible



Beam characterization

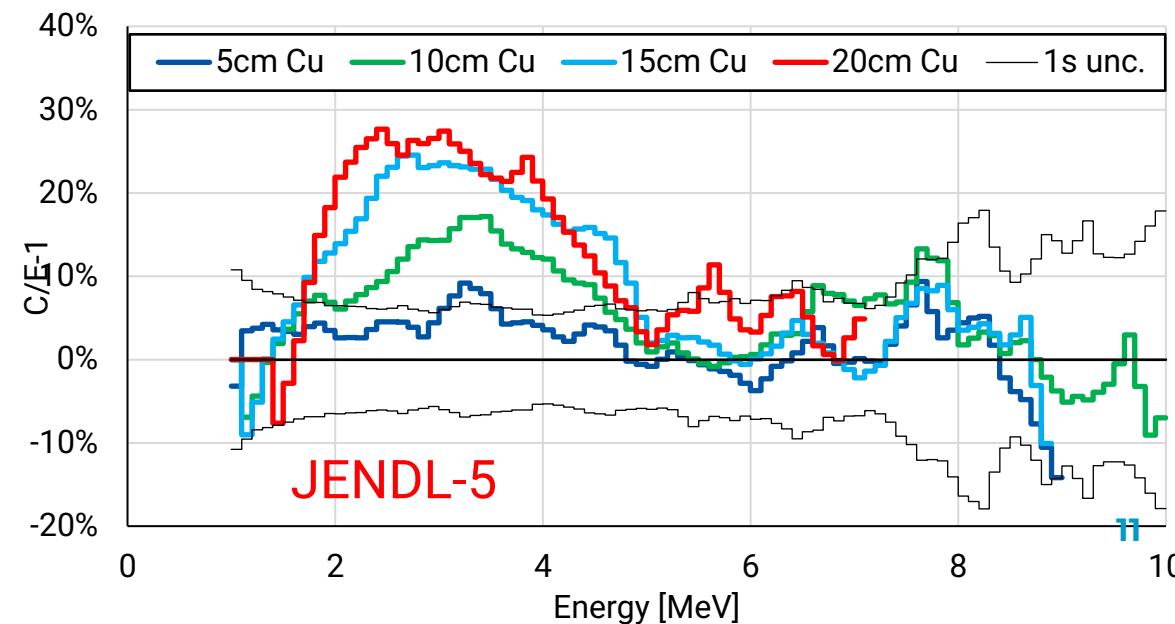
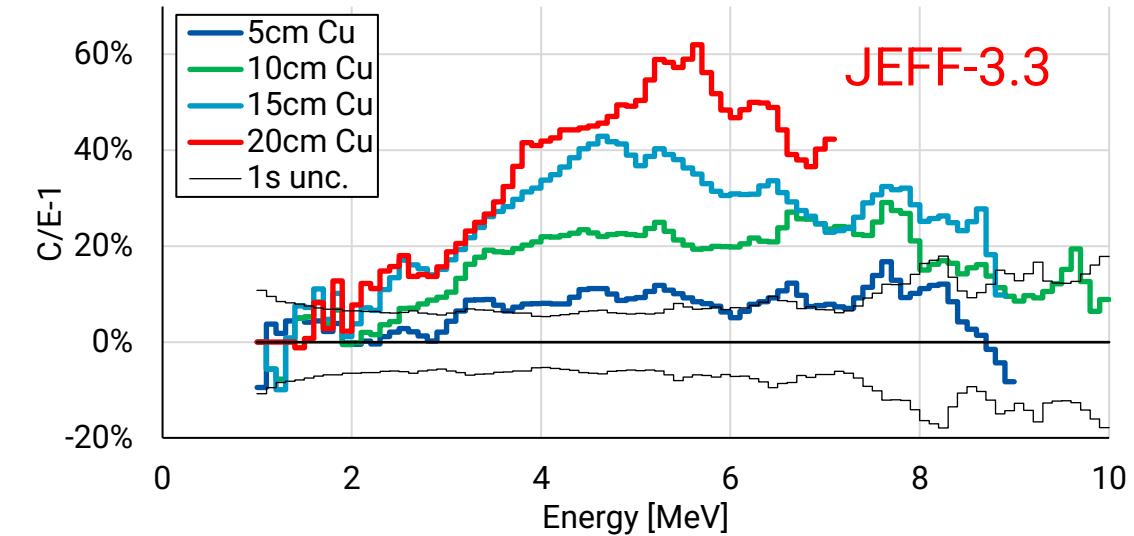
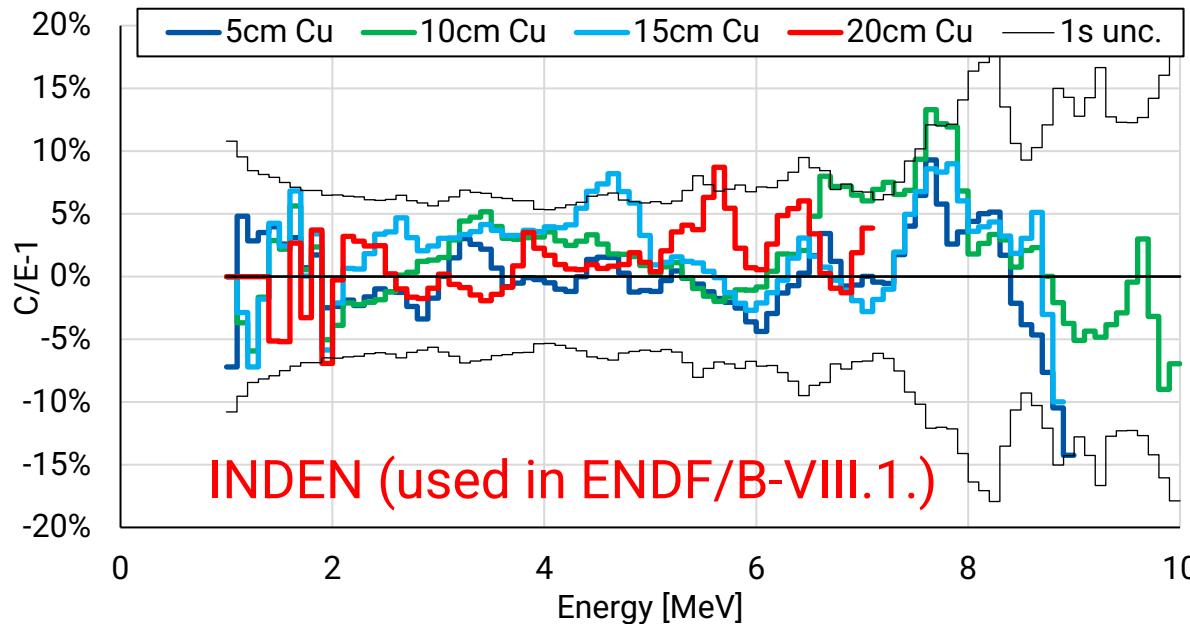
- The spectrum in beam is similar to spectrum in channel in previous measurement
- Spectra in output and measuring position are identical
 - Flux decrease (factor 2.612) implies that beam is relatively not divergent
- The beam is sharp (1% when moved by 35 mm corresponds to scatter from probe).

Misalignment [mm]	Rel. flux
0 (In axis)	1.000
5	0.965
10	0.963
15	0.905
20	0.533
25	0.265
30	0.044
35	0.011



Comparison with various evaluations

- Low uncertainty experiment (beam transmission)
 - Simple geometry
 - Good source description
- Discrepancies from simulation (if any) are on account of cross section evaluation
 - JEFF-3.3 highly discrepant
 - ENDF/B-VIII.1 GOOD
 - JENDL-5 problematic



Leakage from copper cube

Copper cube evaluated
($\geq 99.95\%$ Cu)

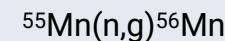
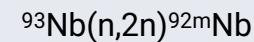
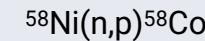
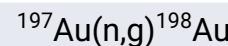


Leakage spectrum (1 - 10 MeV)

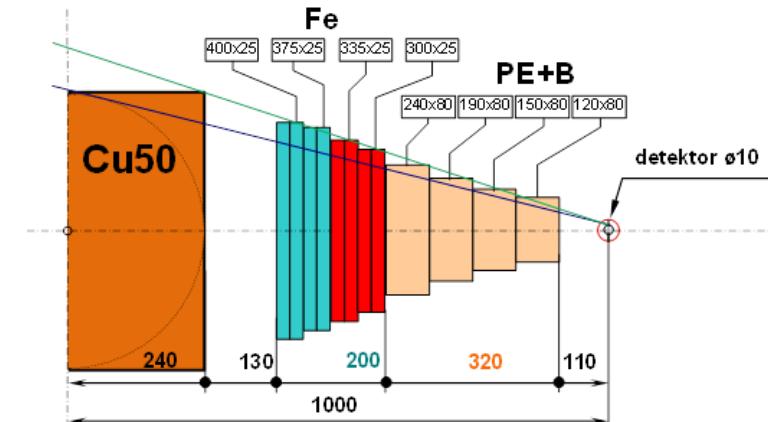
100 cm from center



Reaction rates of :



Background spectrum (room effect)

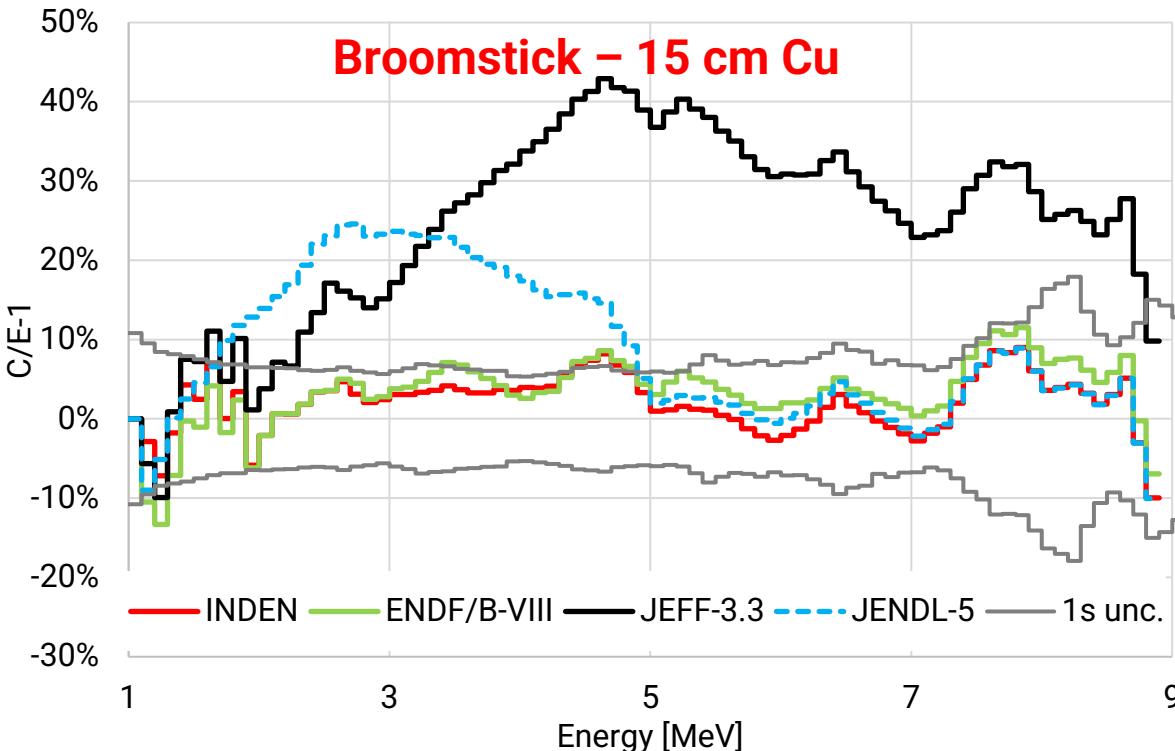


Cone 20cm Fe – 32cm PE

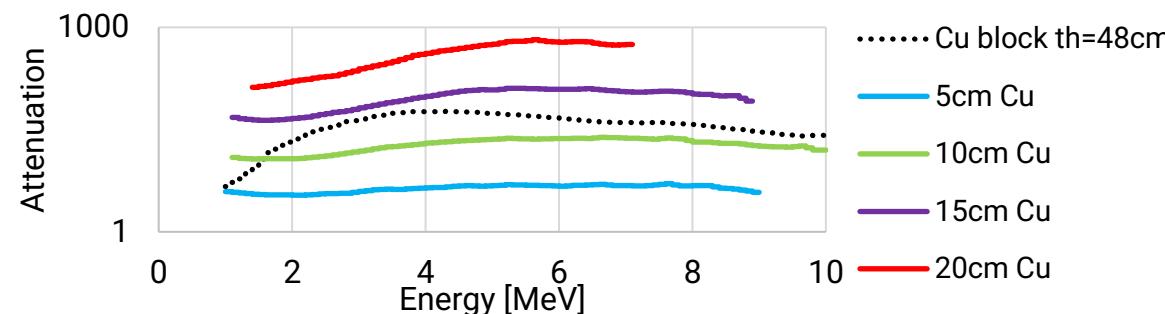
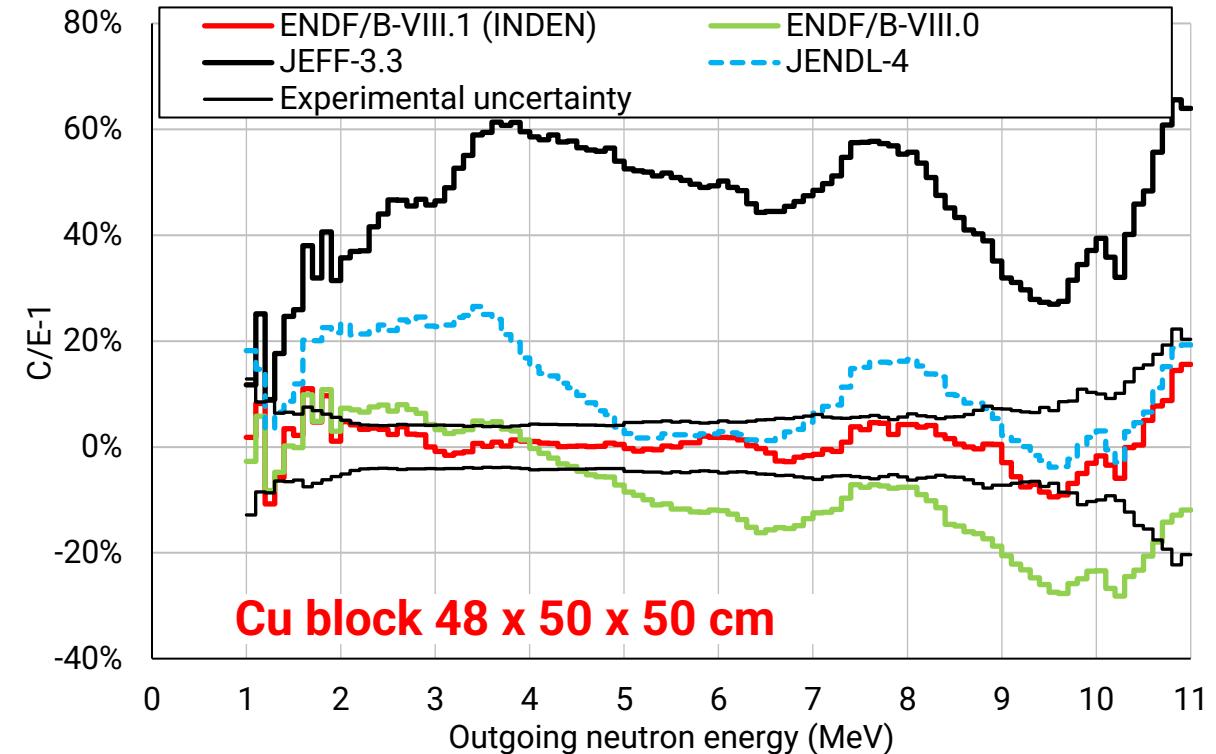


Comparison of broomstick and leakage

- Obtained results are in good consistence with previously measured neutron leakage from copper block

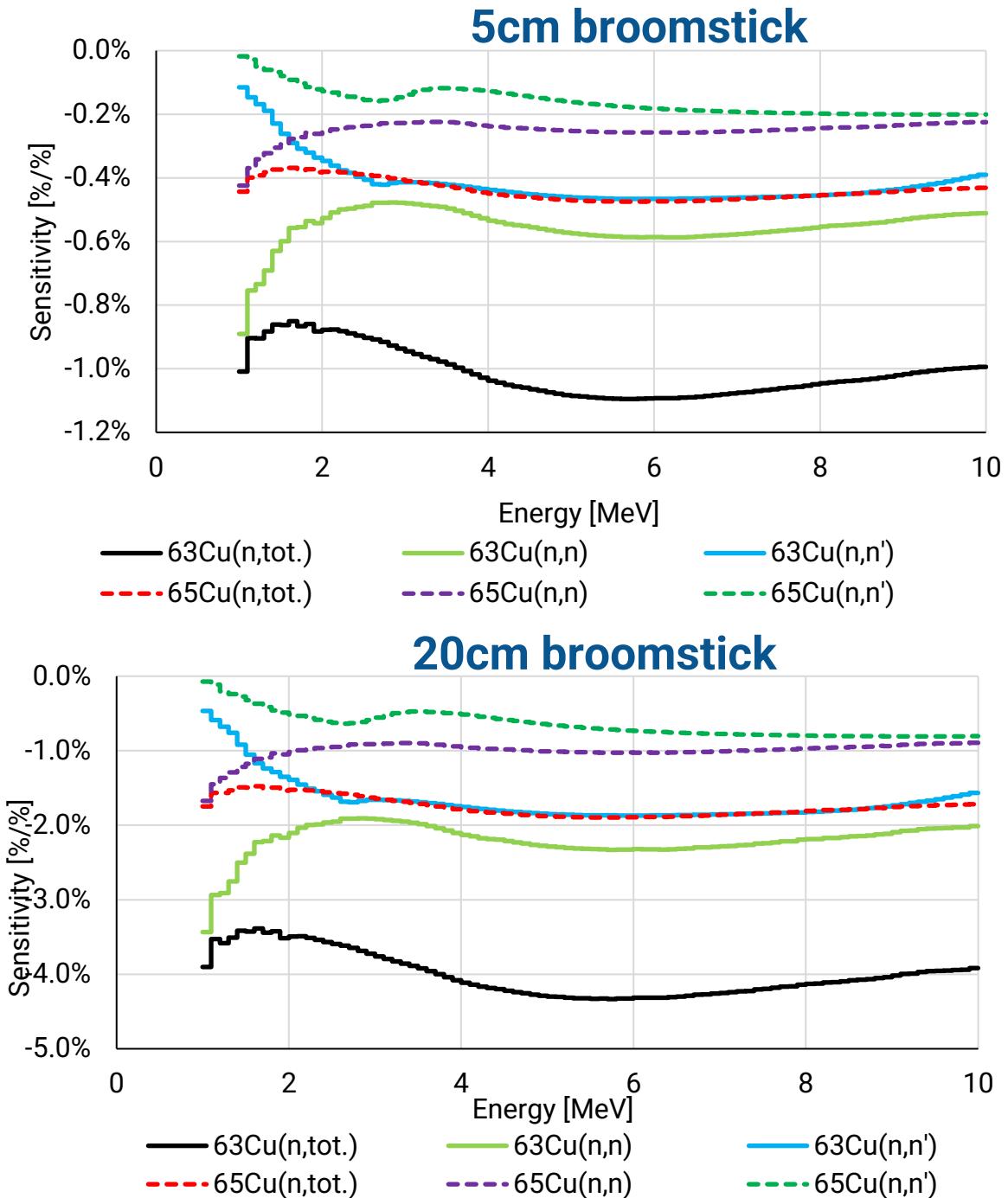
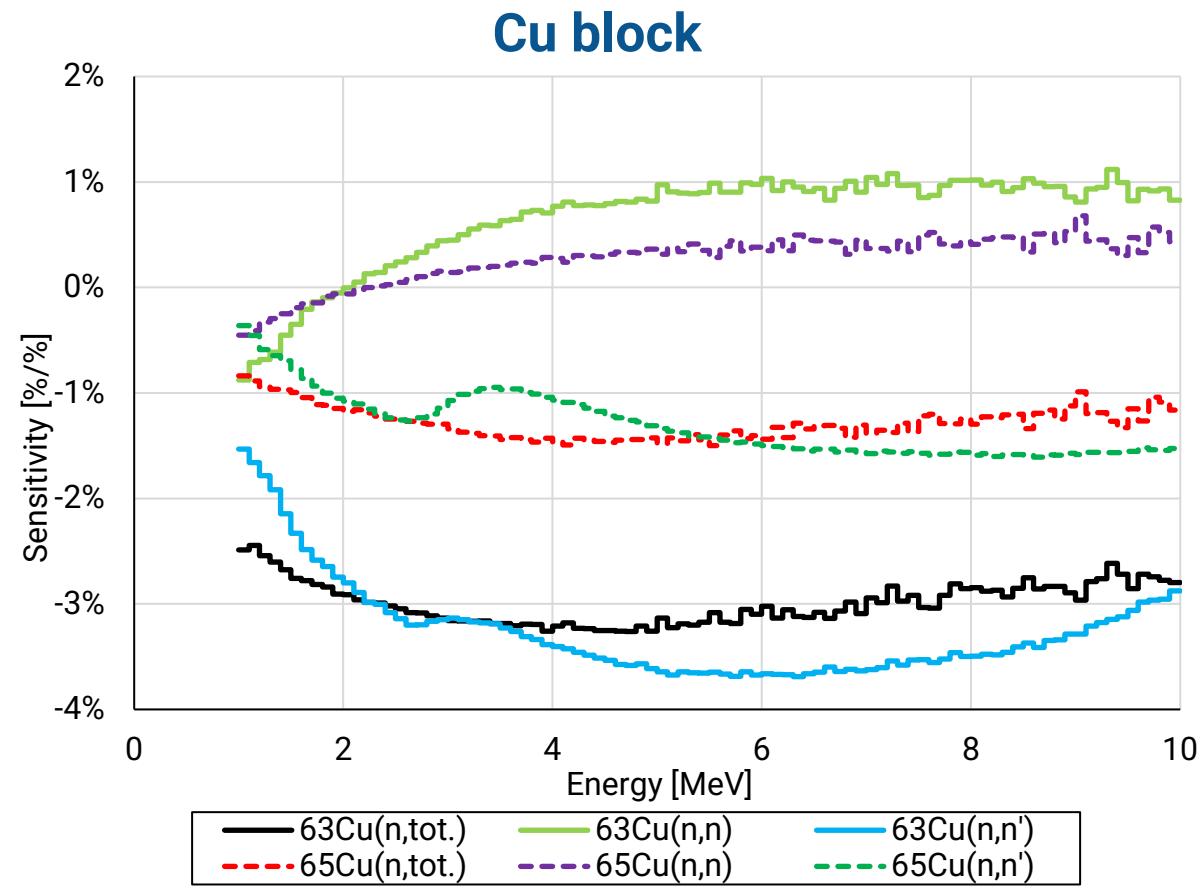


In broomstick more effective attenuation



Sensitivities

- Broomstick is excellent accompanying experiment in cross section validation because sensitivities are different from case of leakage from material block



New neutron leakage experiments

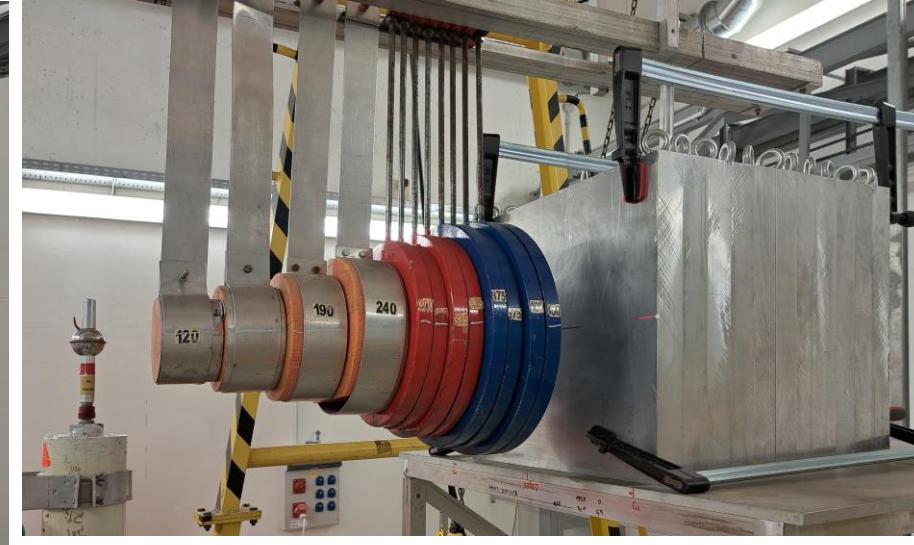
PTFE block
 $60 \times 50 \times 50$ cm



Perchloretylene D=50cm

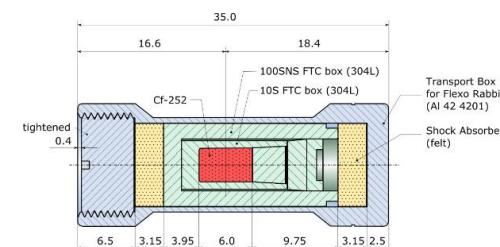


Aluminium block
 $55 \times 50 \times 50$ cm



Cones = 32cm PE + 20cm Fe

^{252}Cf in well defined cladding



Support for benchmarks - 2 m above the lab.floor (as usually)

Al leakage spectra

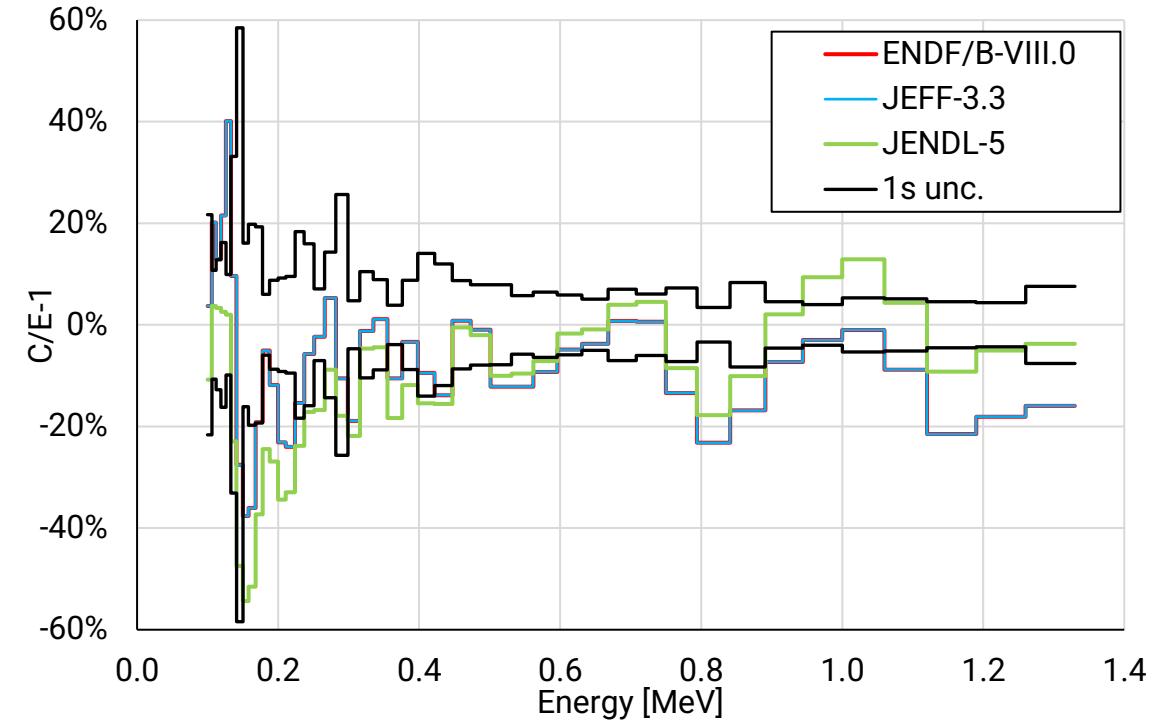
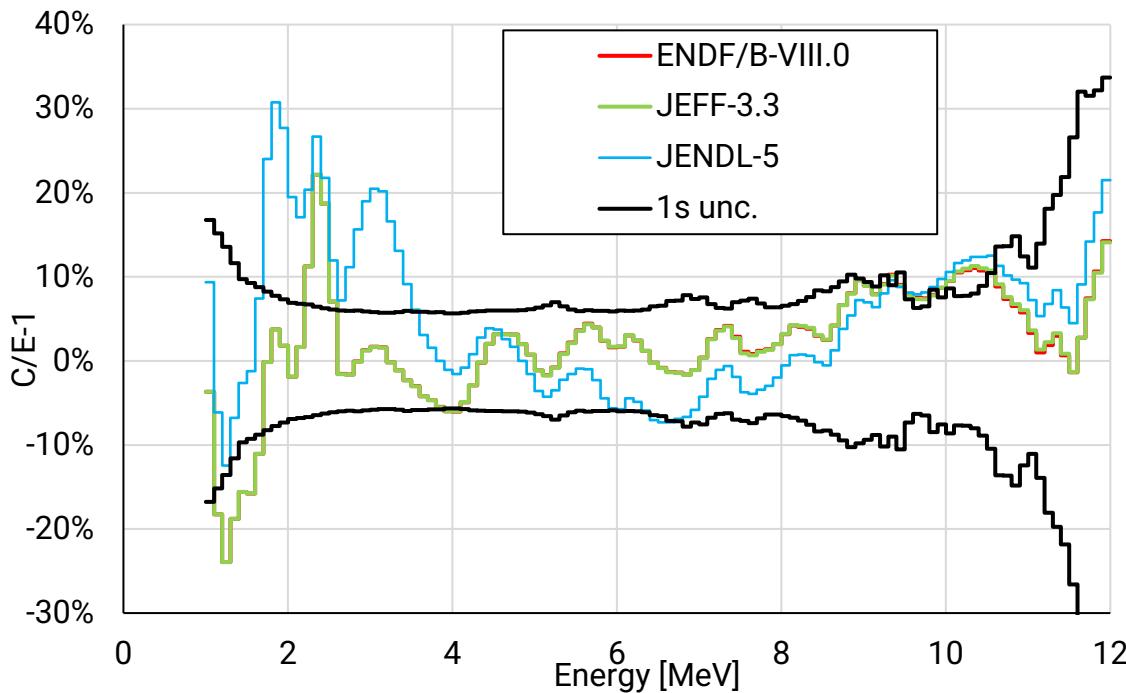
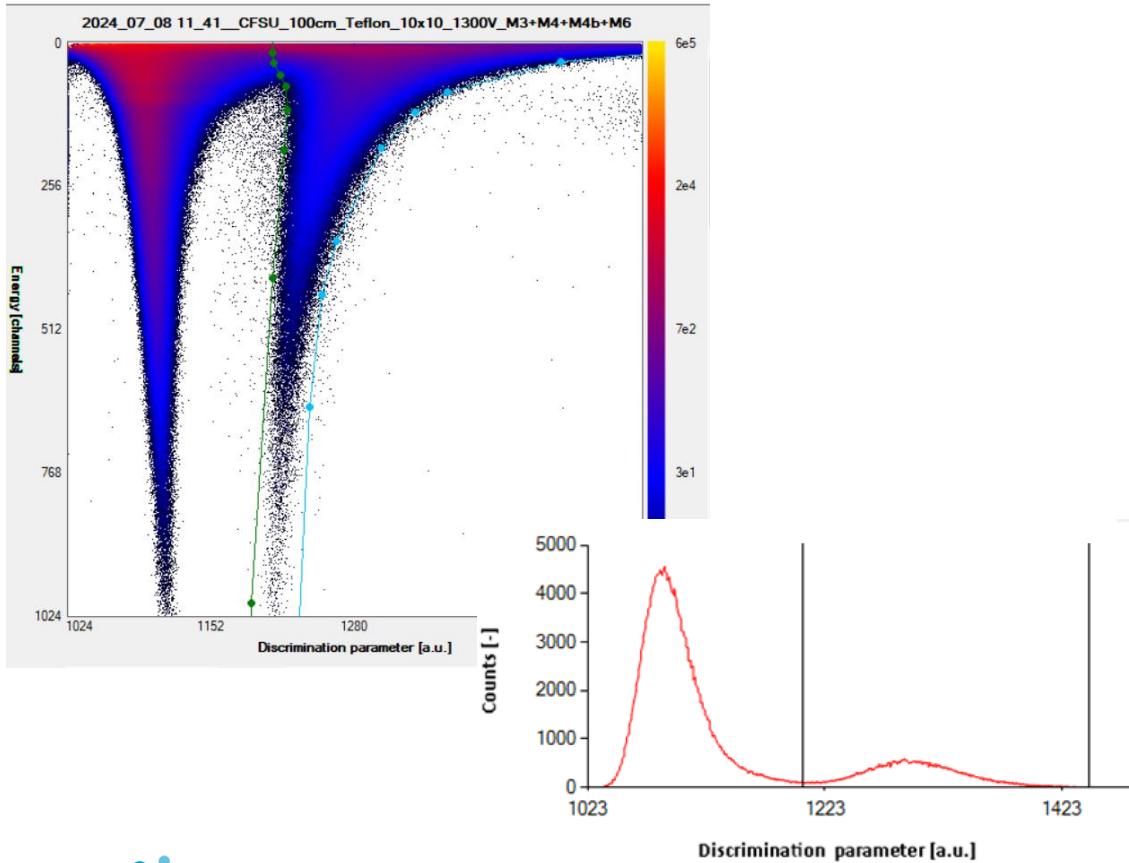


Plate position	Al thickness [cm]	115In(n,n')				58Ni(n,p)			
		ENDF/B-VIII.0 [%]	JEFF-3.3 [%]	JENDL-5 [%]	Unc. [%]	ENDF/B-VIII.0 [%]	JEFF-3.3 [%]	JENDL-5 [%]	Unc. [%]
6	1.18	-2	-2	-2.3	3.9	-9.4	-9.4	-9.6	12.2
7	6.12	-8.7	-8.7	-7.1	2.5	-7.2	-7.2	-6.5	4.3
8	11.06	-12.2	-12.2	-8.2	2.5	-8.7	-8.4	-6.6	2.9
9	16.01	-12.6	-12.6	-6.2	2.8	-11.9	-11.5	-10	2.6
10	20.95	-18.3	-18.3	-10.2	2.8	-8	-8	-2.7	2.4
11	25.91	-16.9	-16.9	-6.8	3.5				

Stilbene measurement of PTFE leakage flux

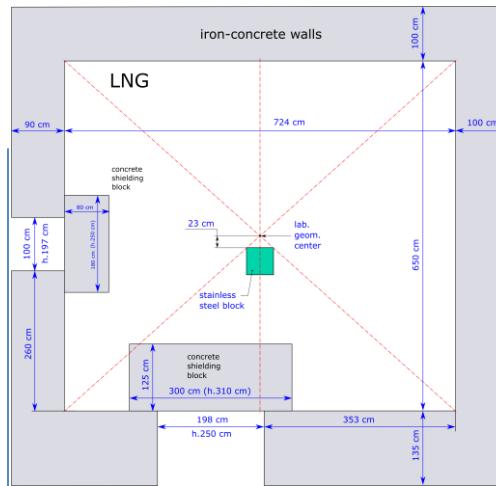
- Neutron leakage spectra in 1 m
- Stilbene detector 10x10 mm
- Not high gamma background (good PSD discrimination)



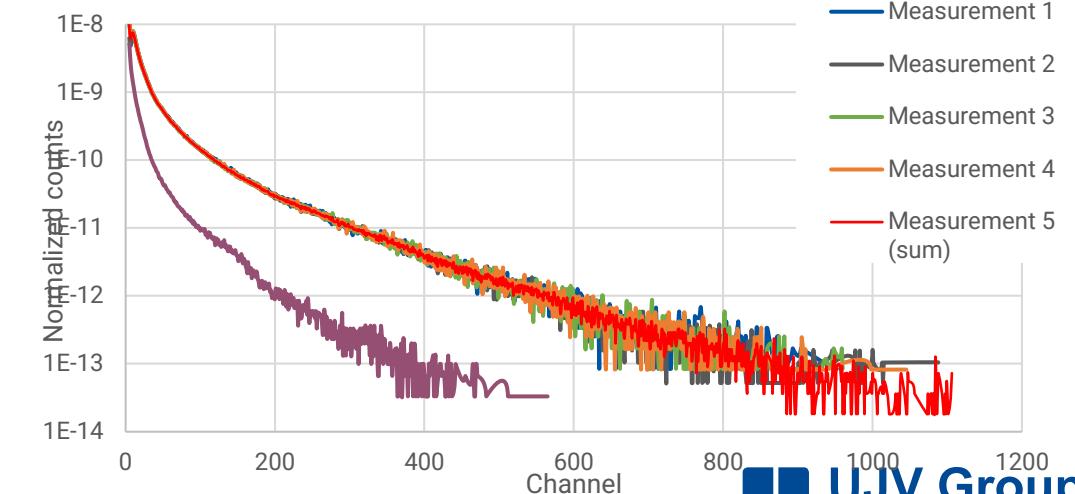
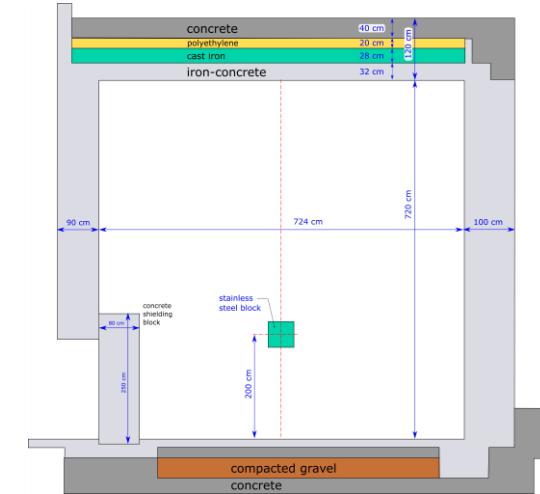
discrimination in 34 channel ~ 0.99 MeV

Room effect evaluation

Horizontal view

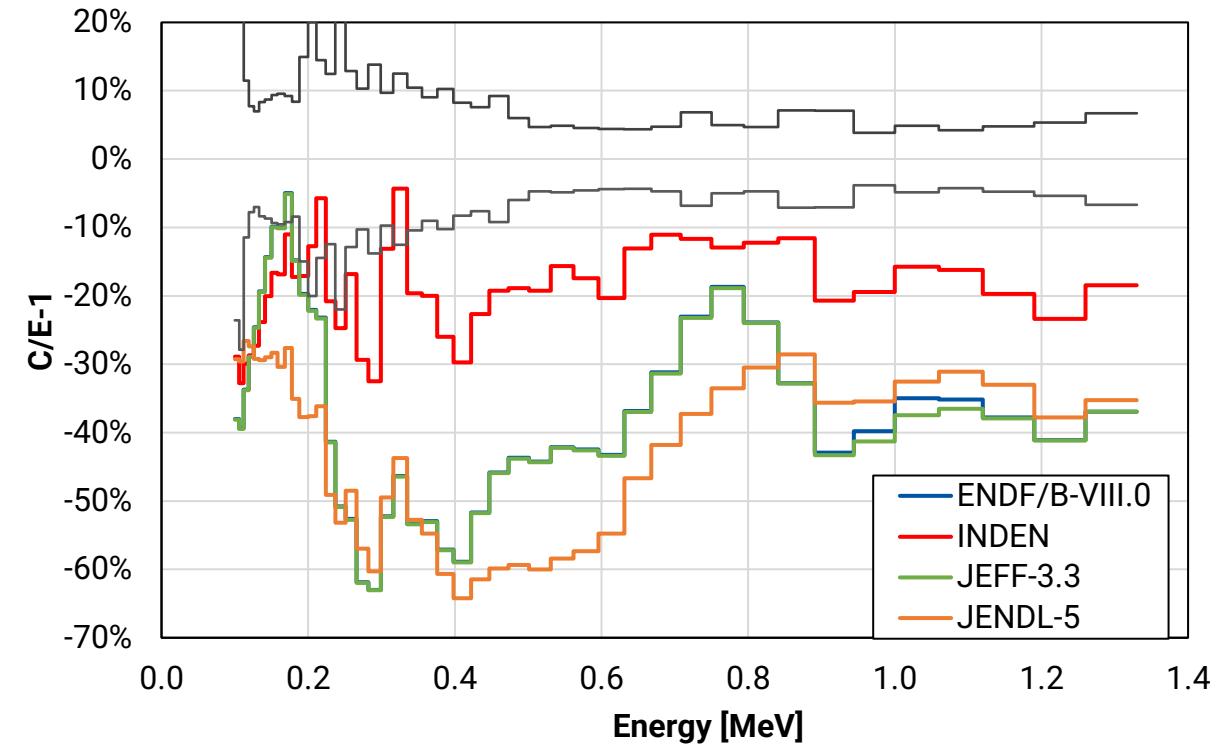
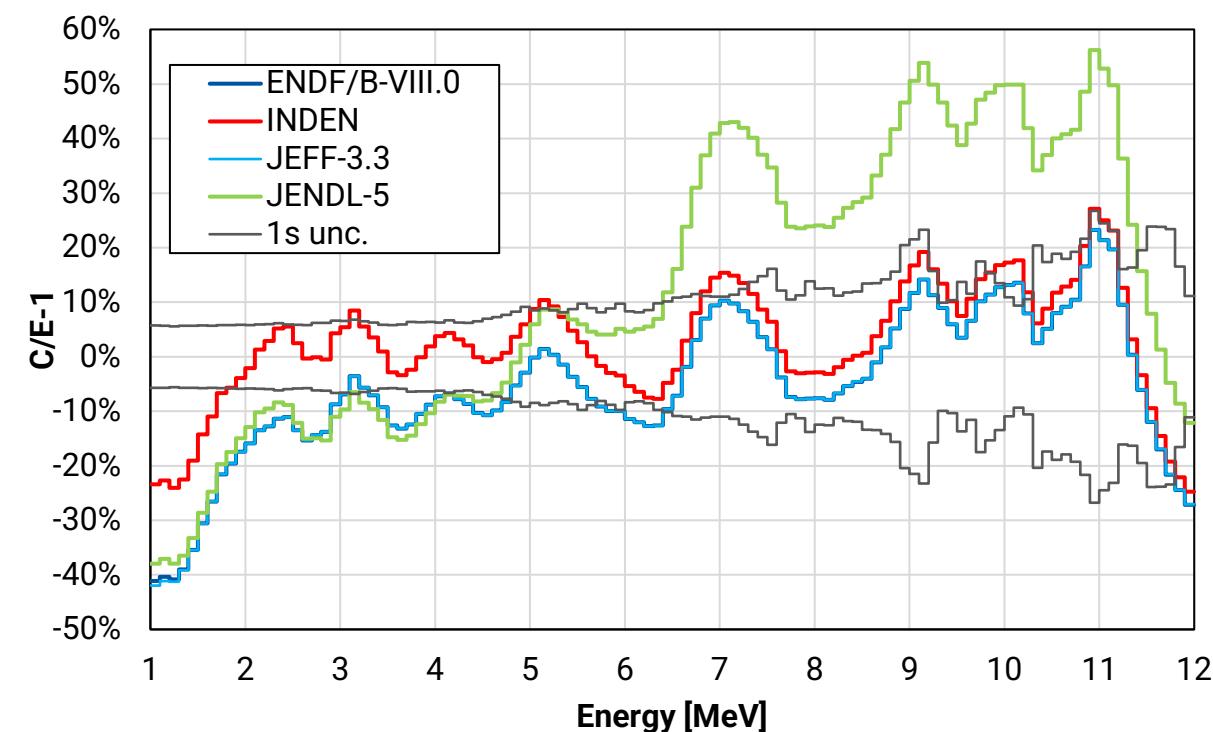


Vertical view



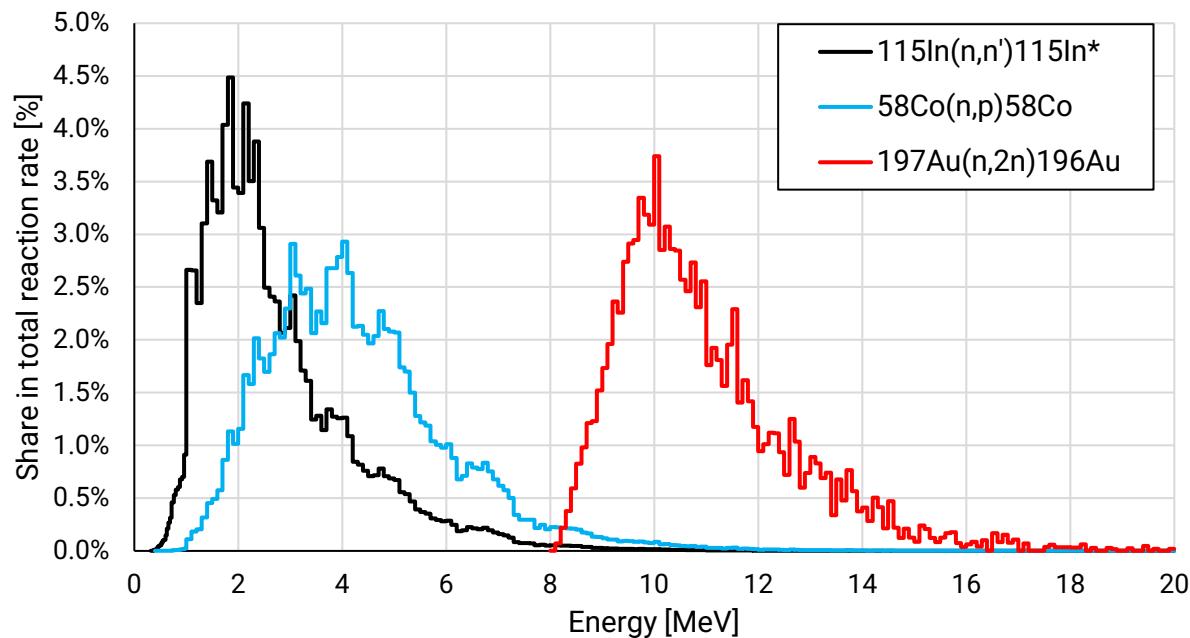
C/E comparison of measured spectra

- Best agreement in case of INDEN (ENDF/B-VIII.1.) however still some discrepancies are notable in region below 2 MeV



Foil results

- Independent monitors of neutron flux
- Even high flux monitored 5g Au foil, 2 month activation, 5d measurement => NPA ~ 1200 cts



Indium $^{115}\text{In}(n,n')$

Placement on plate No. back side	C_2F_4 thickness [cm]	ENDF/B -VIII.0	INDEN	JEFF-3.3	JENDL-5	u_r [%]
6	1.40	4.4	2.0	4.4	2.2	3.9
7	6.82	-4.8	-4.8	-4.8	-7.5	3.0
8	12.28	-5.2	-1.8	-5.3	-7.5	3.1
9	17.68	-14.4	-8.1	-14.5	-15.9	4.4
10	23.09	-24.6	-16.3	-24.6	-25.0	7.5
11	28.48	-26.7	-14.7	-26.8	-25.1	5.5

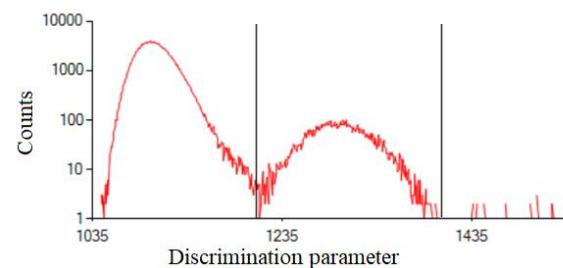
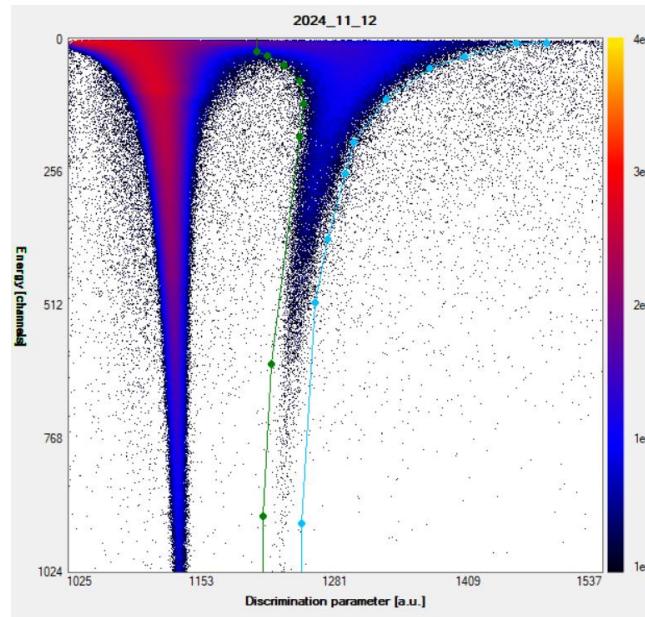
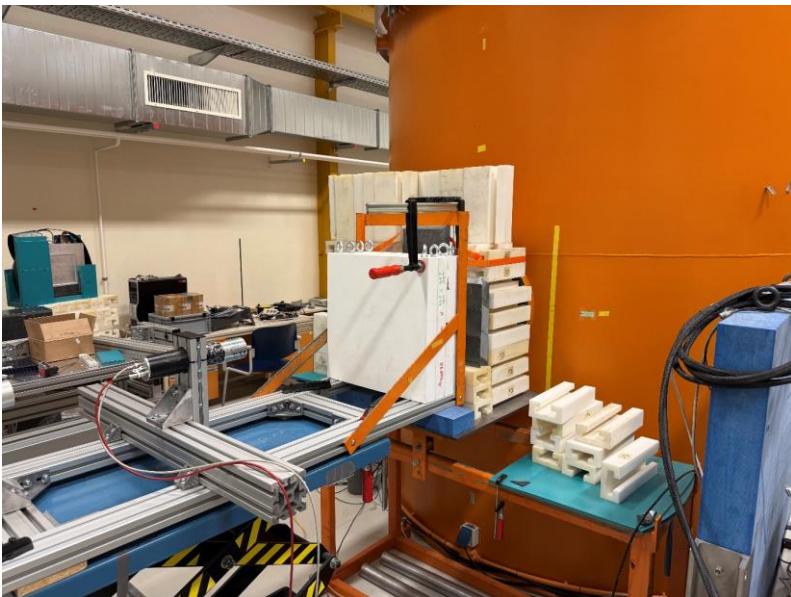
Nickel $^{58}\text{Ni}(n,p)$

Placement on plate No. back side	C_2F_4 thickness [cm]	ENDF/B -VIII.0	INDEN	JEFF-3.3	JENDL-5	u_r [%]
6	1.40	3.1	2.1	3.1	1.8	9.0
7	6.82	-2.0	-1.7	-2.1	-3.3	3.4
8	12.28	-5.6	-2.9	-5.6	-5.7	2.9
9	17.68	-7.8	-3.0	-7.8	-6.6	3.3
10	23.09	-12.3	-5.5	-12.3	-9.6	3.2
11	28.48	-9.8	0.5	-9.8	-4.0	3.5

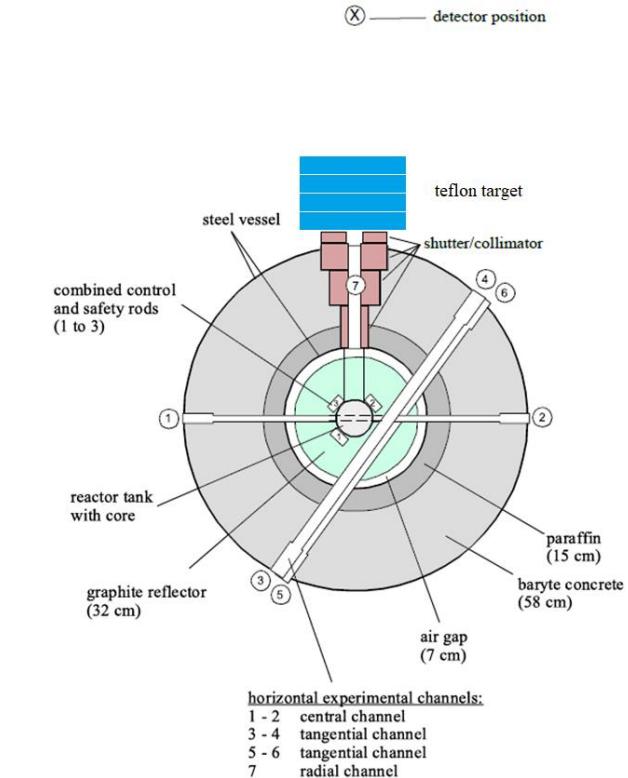
Gold $^{197}\text{Au}(n,2n)$

Placement on plate No. back side	C_2F_4 thickness [cm]	ENDF/B -VIII.0	INDEN	JEFF-3.3	JENDL-5	u_r [%]
10	23.09	-3.8	-2.6	-4.4	17.0	17.8

AKR-2 experiment

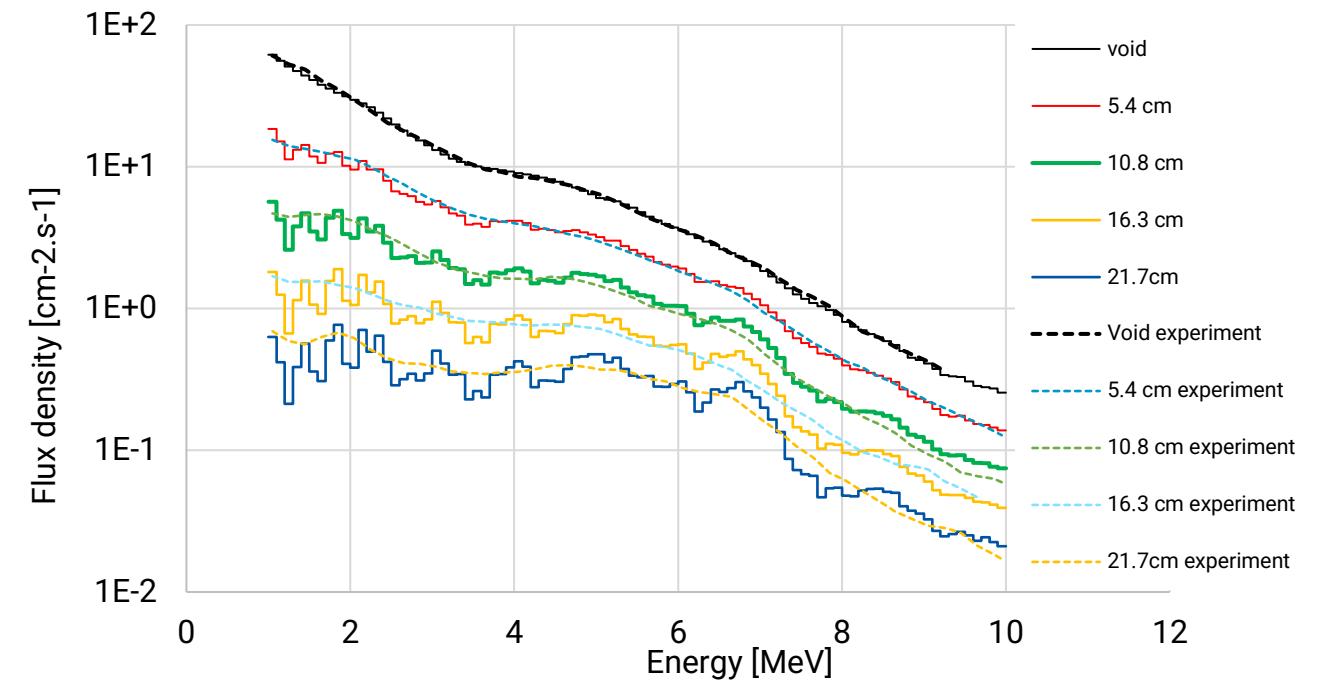
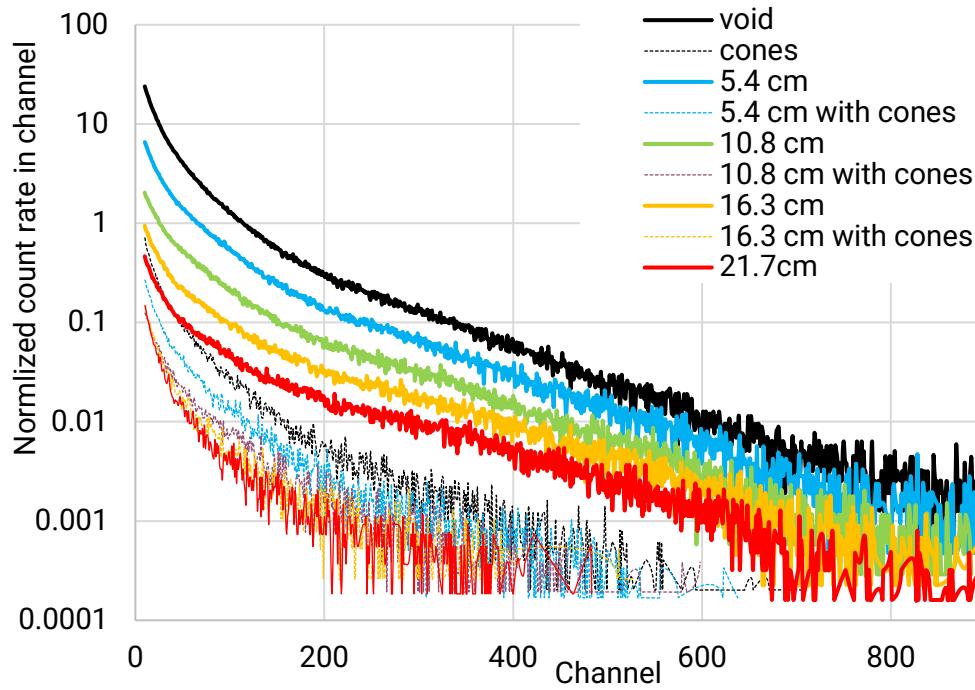


discrimination in 41 channel ~ 1.1 MeV



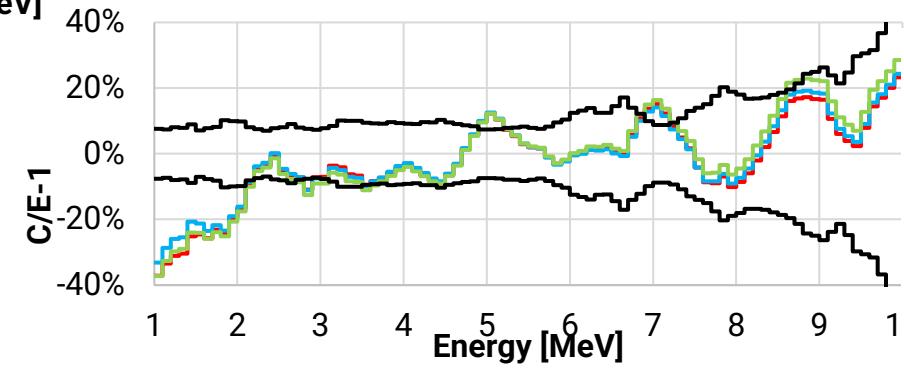
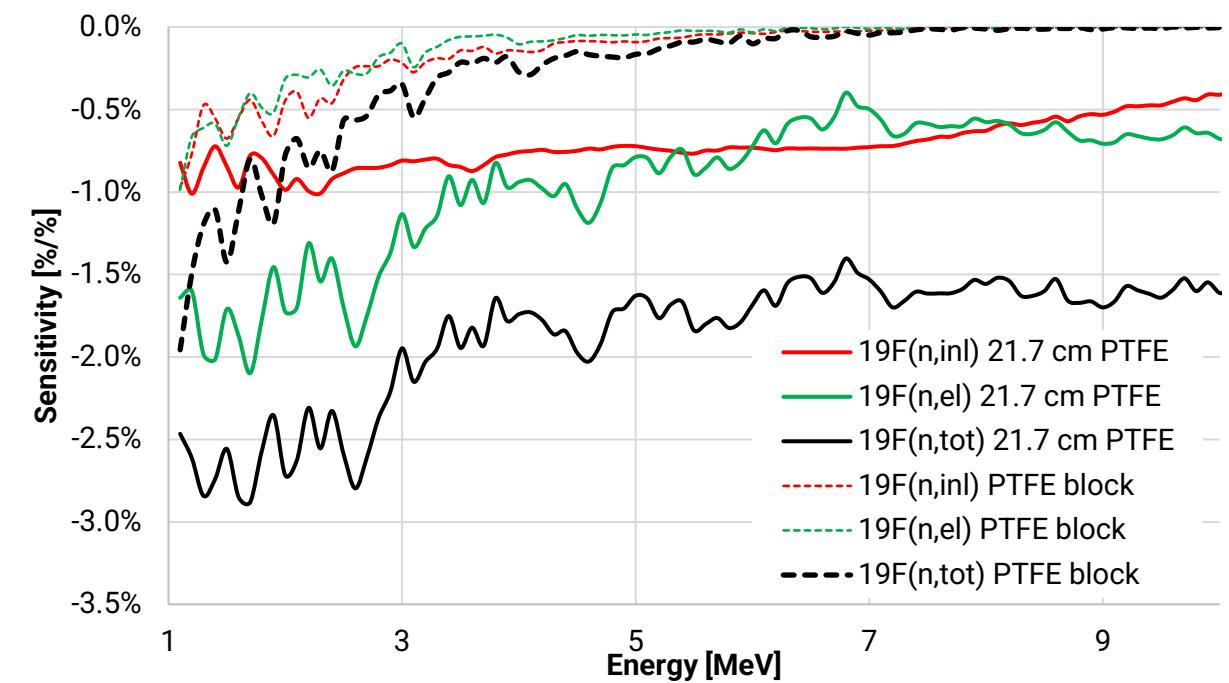
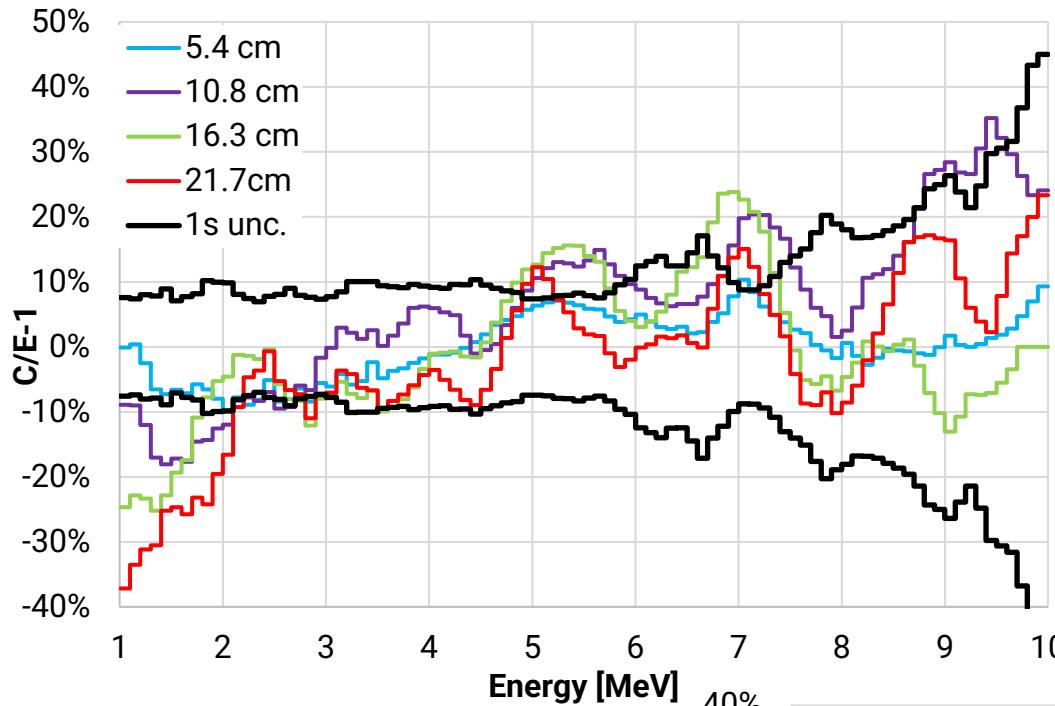
AKR-2 spectra

- Large beam diameter $\sim D=12$ cm in this experiment
- Due to geometry (holding table) + beam diameter ... notable background ..
- In this large beam diameter also elastic/inelastic play role . . . (but not so high as in leakage experiments)



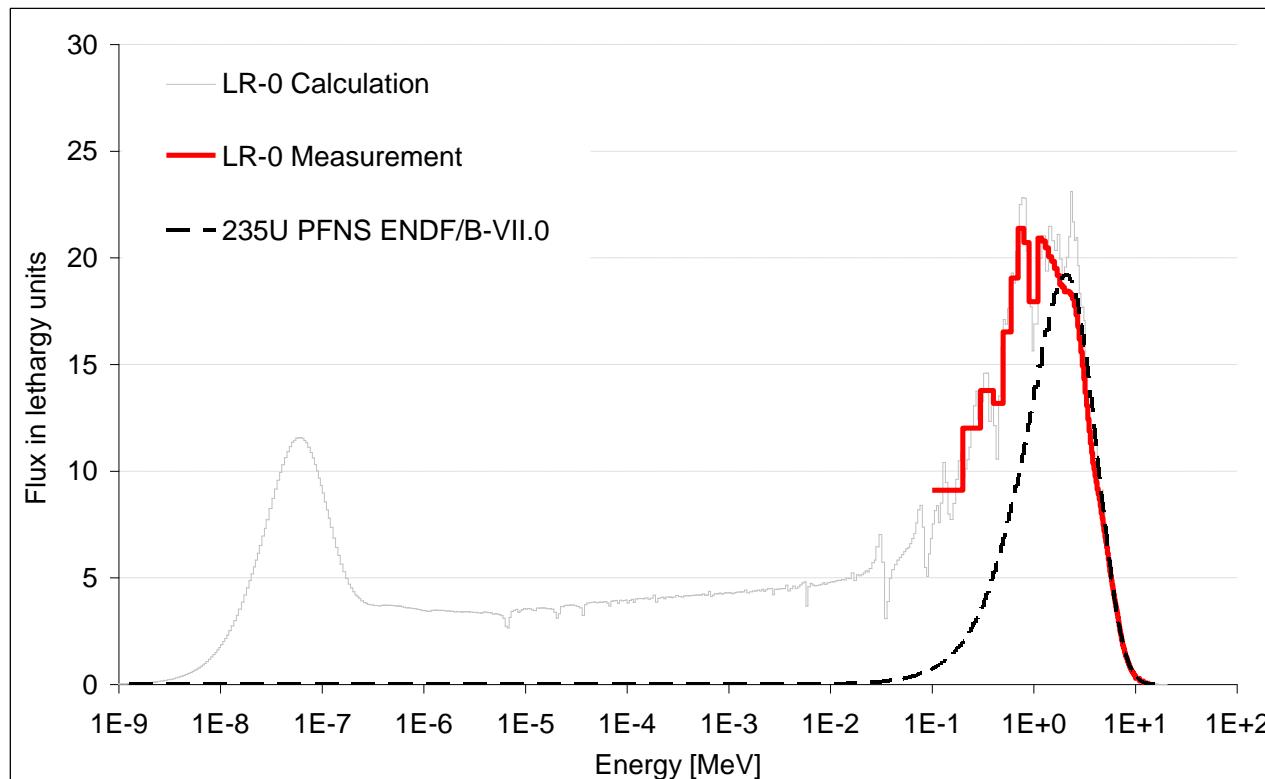
C/E of measured spectra

- Above 2 MeV good agreement below discrepancies similarly as in californium experiment
- Most likely issue in ^{19}F total cross section below 2 MeV



New experiments in Reference Neutron Benchmark Field

- Permanent reproducible neutron field
- Well defined spatial distribution of the neutron field
- Well defined neutron energy spectrum
- Characterized gamma spectrum

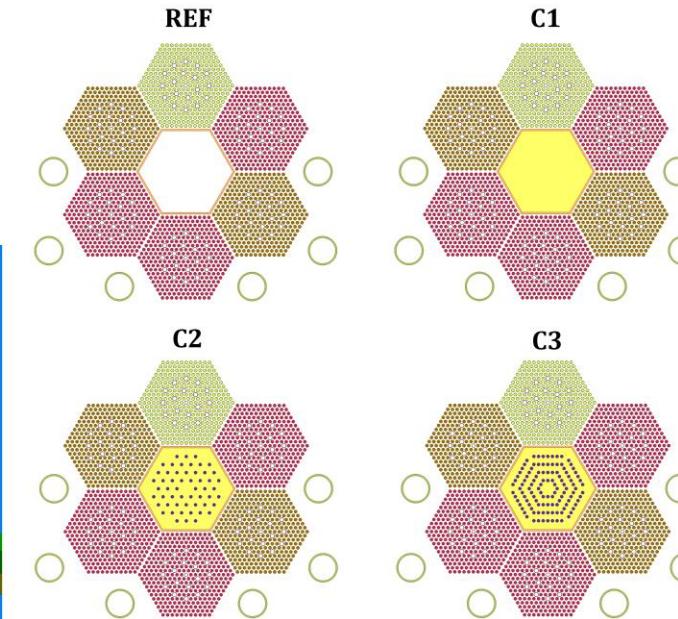
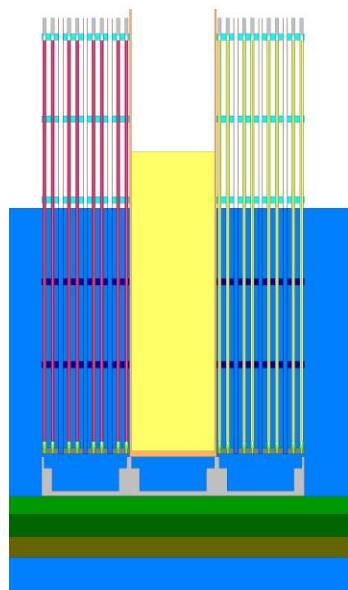
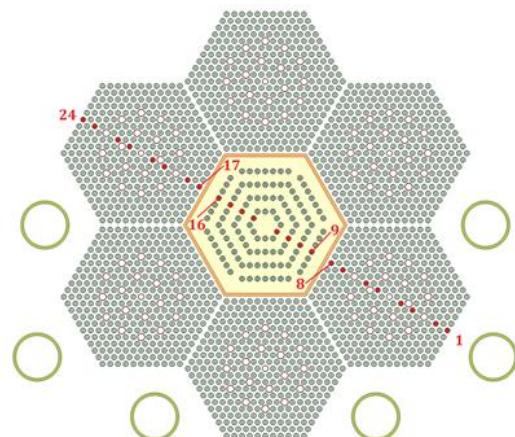


IRDFF-II: A New Neutron Metrology Library

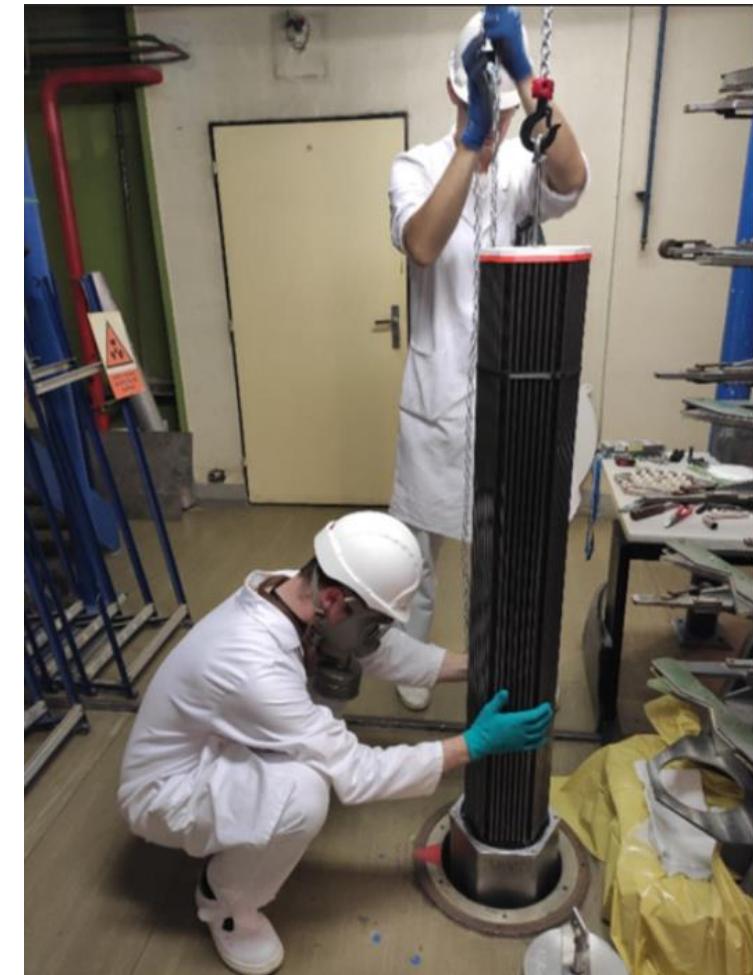
C. Reference Neutron Fields from Detailed Computational Models	54
1. SPR-III Fast Burst Reactor Central Cavity	54
2. ACRR Pool-type Reactor Central Cavity	55
3. ACRR Pool-type Reactor Pb-B ₄ C (LB44) Bucket	57
4. ACRR Pool-type Reactor Cd-Polyethylene Bucket	57
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6. ACRR Fueled Ring External Cavity (FREC-II)	58
7. Mol-BR1 Cavity ²³⁵ U Fission Spectrum	58
8. LR-0 Reactor at Řež	59
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Validation of chlorine cross sections

- Some molten salt reactor (MSR) concepts propose using molten chlorides
- New experiments realized in LR-0 special core in Rez
 - C_2Cl_4 (C is very well known => discrepancies are on account of Cl)
 - K_{eff}
 - Power density profile



REF – reference LR-0 core without C_2Cl_4 ,
C1 – case only with C_2Cl_4 ,
C2 – case with C_2Cl_4 and 54 fuel pins,
C3 – case with C_2Cl_4 and 114 fuel pins



Criticality results C2Cl4

- Critical parameter determined for selected arrangements depending on Cl content
 - Driven by water
 - Systematic uncertainties ~200pcm
- Large discrepancies in arrangements with chlorine
 - Significant underprediction

Modification	H_{cr} [mm]	Unc.	$d\rho/dH$ [pcm/cm]	Unc.
Reference core	538.75	0.81	285.67	11.64
Case 1	727.24	0.11	125.96	0.82
Case 2	709.84	0.08	130.03	0.68
Case 3	692.95	0.09	146.15	2.40

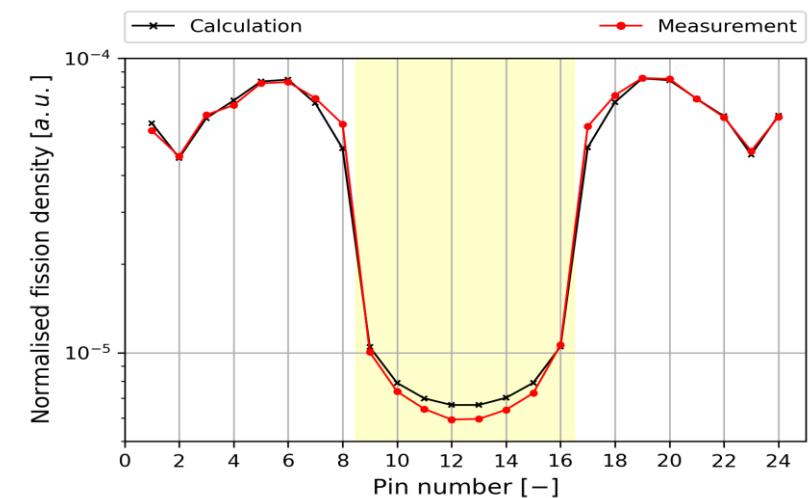
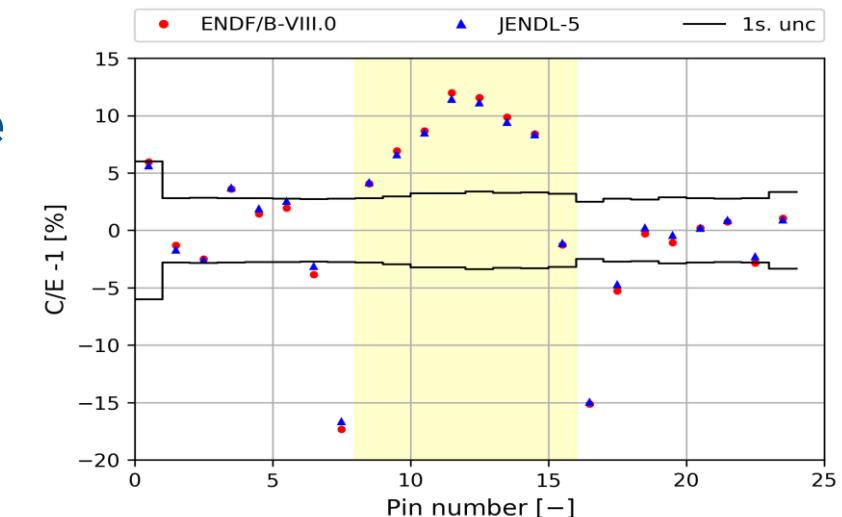
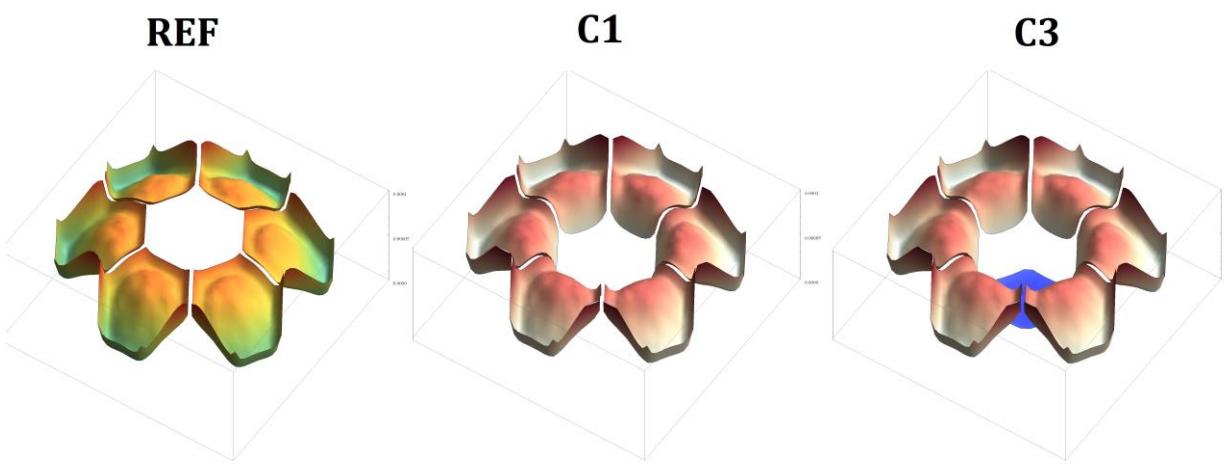
- Difference from reference case, core with ENDF/B-VIII.0

Library	Difference from reference case		
	C1	C2	C3
ENDF/B-VIII.0	-162	-168	-174
ENDF/B-VII.1	-166	-163	-175
JEFF-3.3.	-162	-170	-184
JENDL-5.	-38	-63	-90

Library	k_{eff} [-] ± 0.00004			
	REF case	C1	C2	C3
ENDF/B-VII.1	0.99990	0.99825	0.99826	0.99816
ENDF/B-VIII.0	0.99953	0.99791	0.99785	0.99779
JEFF-3.3.	1.00174	0.99957	0.99959	0.99961
JENDL-5.	1.00152	1.00067	1.00042	1.00023

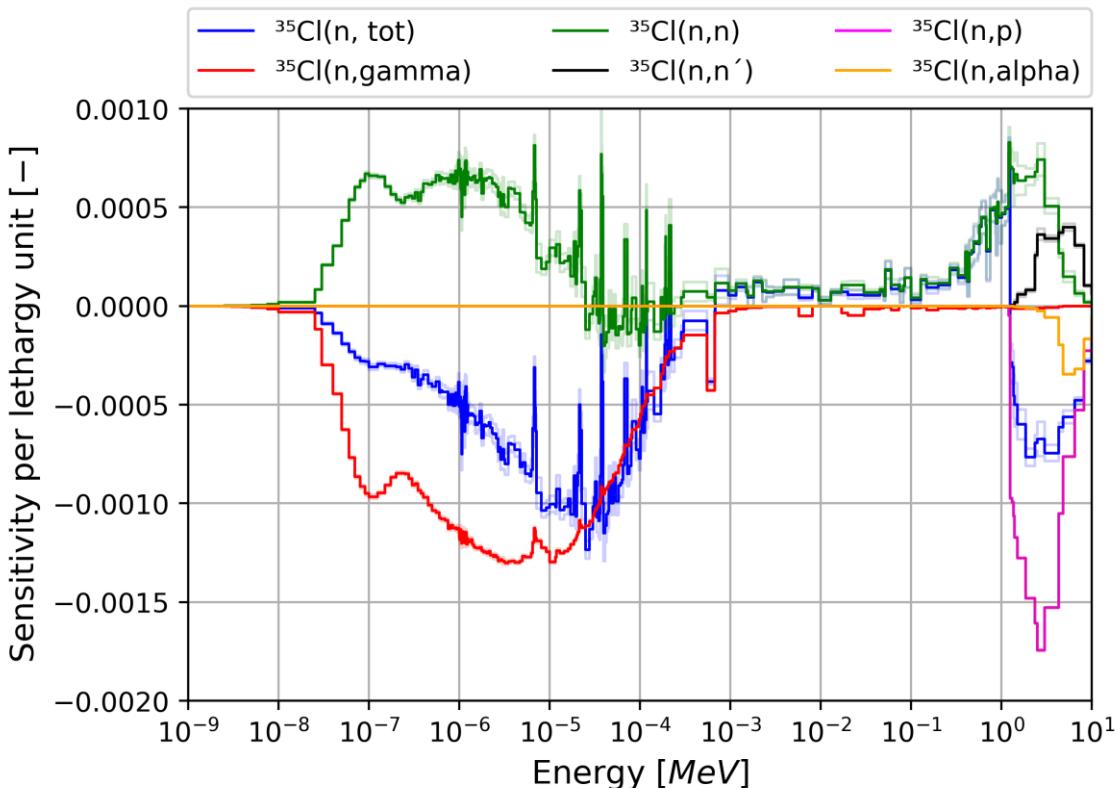
Power density profile

- Simulations are predicting significant effect on power profile
- The decrease is really observed
- ...
 - Systematic uncertainties ~ 200 pcm
- Large discrepancies in arrangements with chlorine
 - Significant underprediction



Sensitivity analysis

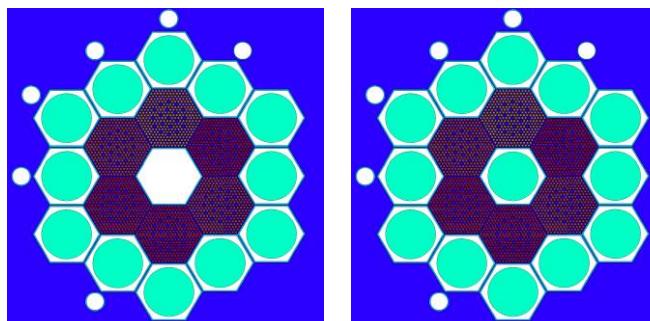
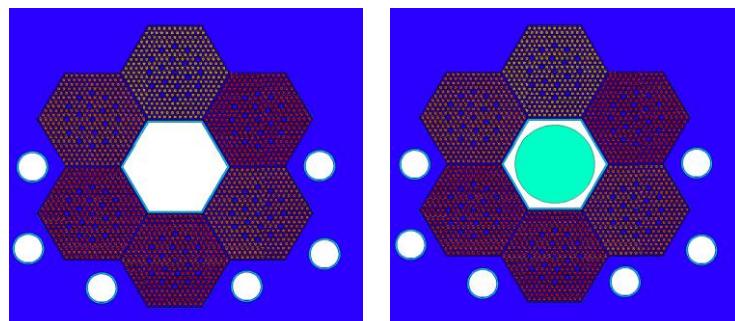
- This experiment is highly sensitive for $^{35}\text{Cl}(\text{n},\gamma)$ in low energy, $^{35}\text{Cl}(\text{n},\text{p})$ in high energy



Nuclide	Reaction	Sensitivity [pcm]	Std. Dev. [%]
^{35}Cl	(n, γ)	-915	0.12
^{35}Cl	(n,total)	-560	0.91
^{35}Cl	(n,n)	538	0.92
nat C	(n,total)	240	1.77
nat C	(n,n)	239	1.77
^{35}Cl	(n,p)	-202	0.07
^{37}Cl	(n,total)	75	2.88
^{37}Cl	(n,n)	67	3.18
^{35}Cl	(n,n')	48	1.41
^{35}Cl	(n,a)	-27	0.15
^{37}Cl	(n,n')	13	2.42

Construction materials in reactivity experiments

- The effect of inserted block is studied
- In past was tested steel (iron + 1.5% Mn)
- New experiment with copper block
 - Reference core 541.51 +/- 1.32 mm
 - Copper in center 611.42 +/- 0.12 mm
 - Iron in center 579.01 +/- 0.79 mm
 - Iron around 548.09 +/- 0.69 mm
 - Iron around + center 598.23 +/- 0.92 mm



	ENDF/B-VIII.0	ENDF/B-VII.1	JEFF-3.3
Reference case	0.99960	0.99989	1.00176
Iron in center	0.99808	0.99843	1.00001
Iron around	0.99651	0.99692	0.99946
Iron around + center	0.99520	0.99538	0.99785

$^{63}\text{Cu}(\text{n},\text{g})$

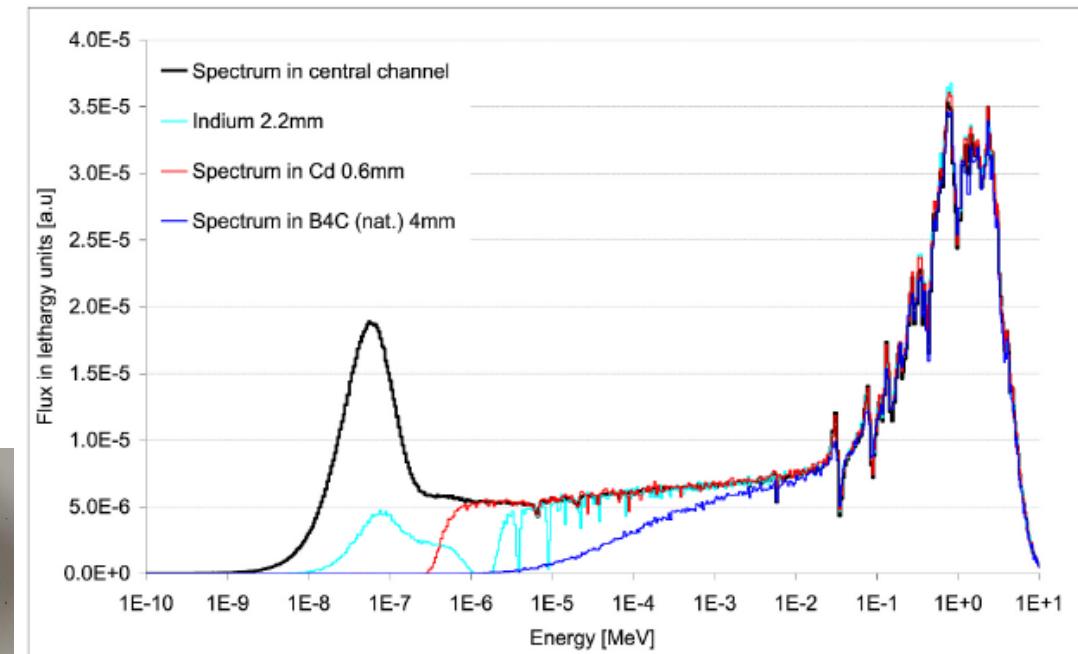
- Planned measurements with B_4C (^{10}B) filters (JSI collaboration), spring 2025
- In 2023 preliminary experiment with natural boron capsule (nearly equivalent to JSI one) ~ 2.364 g/cm^3 instead of JSI 2.36 g/cm^3

	Poured	Sintered
Density [g/cm ³]	1.197	2.36
Thickness [mm]	7.9	4
$^{55}\text{Mn}(\text{n},\text{g}) [\text{s}^{-1}]$	3.67E-29	3.75E-29

Table 5
Summary of C/E-1 for $^{63}\text{Cu}(\text{n},\text{g})^{64}\text{Cu}$ reaction in different neutron fields.

$^{63}\text{Cu}(\text{n},\text{g})^{64}\text{Cu}$	ENDF/B-VIII.0	JEFF-3.3	JENDL-5	IRDFF-II	Uncertainty
Void	20.2%	23.4%	20.9%	20.2%	4.5%
Cd filter	17.7%	28.1%	19.4%	17.8%	3.8%
B_4C filter	13.4%	26.4%	24.0%	13.9%	3.6%
$^{252}\text{Cf}(\text{s,f})$	-2.4%	-2.0%	-1.7%	-3.6%	3.4%

Schulc et al., Appl. Rad. and Isot. 215, (2025), 111586



Kostal et al., Appl. Rad. and Isot. 169 (2021) 109566



New benchmarking of reference core

During characterization of heterogenous critical assembly, in ch-1 of ICSBEP benchmark, the evaluators define material, geometrical, ... etc parameters. In case of fuel cladding use of producer certificate is welcomed. But is it reliable ??

Zr case

XRF analysis

El	m/m%	StdErr
--	-----	-----
Zr	96.84	0.47
Nb	1.63	0.08
Mo	0.80	0.17
Si	0.62	0.26
Cl	0.11	0.026

- Producer certificate

- Zr 98.97 %
- Nb 1 %
- Hf < 0.03 %

Mass spectrometry

	mean	unc
Hf	0.04	0.0032
Nb	1.10	0.088
Zr	97.70	7.816

NAA analysis

	449 ppm	50ppm
Hf	449 ppm	50ppm
Ta	2 ppm	0.2ppm
K	18 ppm	1ppm
Nb	1.25%	0.18%

Conclusions

Broomstick experiment realized at the VR-1 and it seems excellent tool for total cross section testing

There is a strong potential of AKR-2 for broomstick – discussion about use of C_2Cl_4 in future experiments is planned

The Cf benchmark experiment for Teflon and Aluminium will be submitted to ICSBEP

The new experiments realized in LR-0 reference field. The new benchmarking is ongoing ...

Thank you for attention

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XRF detail

