Validation of nuclear cross section data by activation experiments

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Content

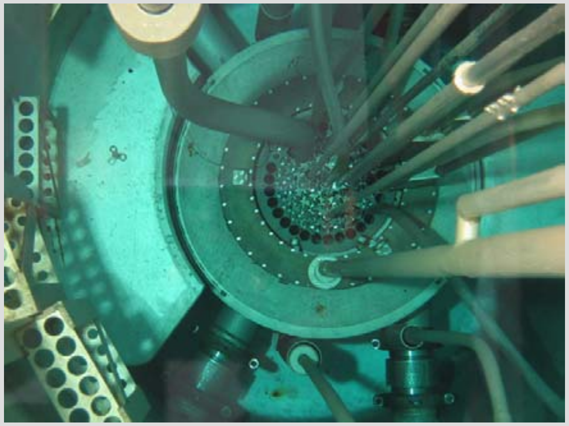
- TRIGA reactor and applications
- purpose of the project
- methods
- experiment
- calculations
- results

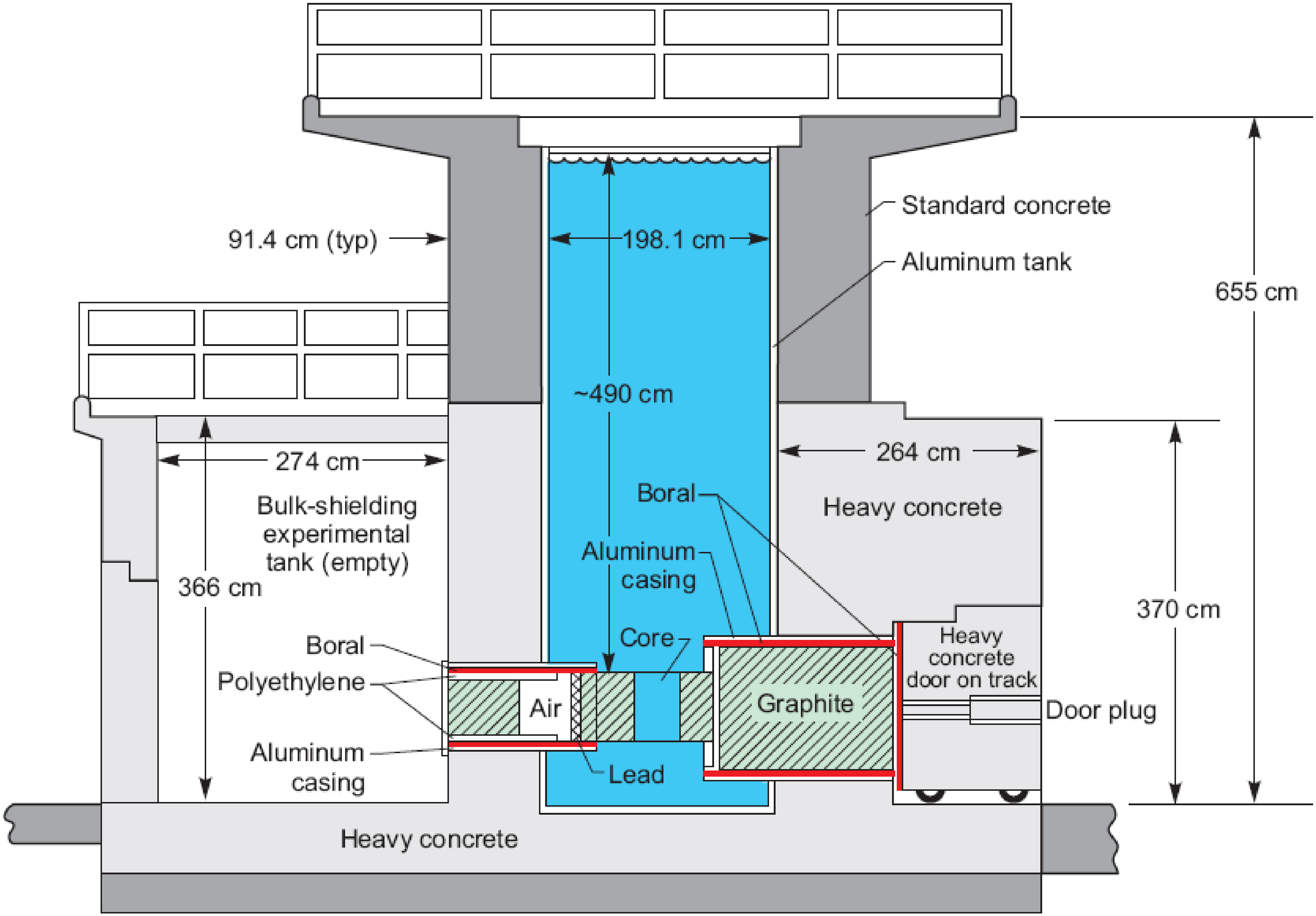
- Example:
  - Rhodium data validation

JSI TRIGA reactor

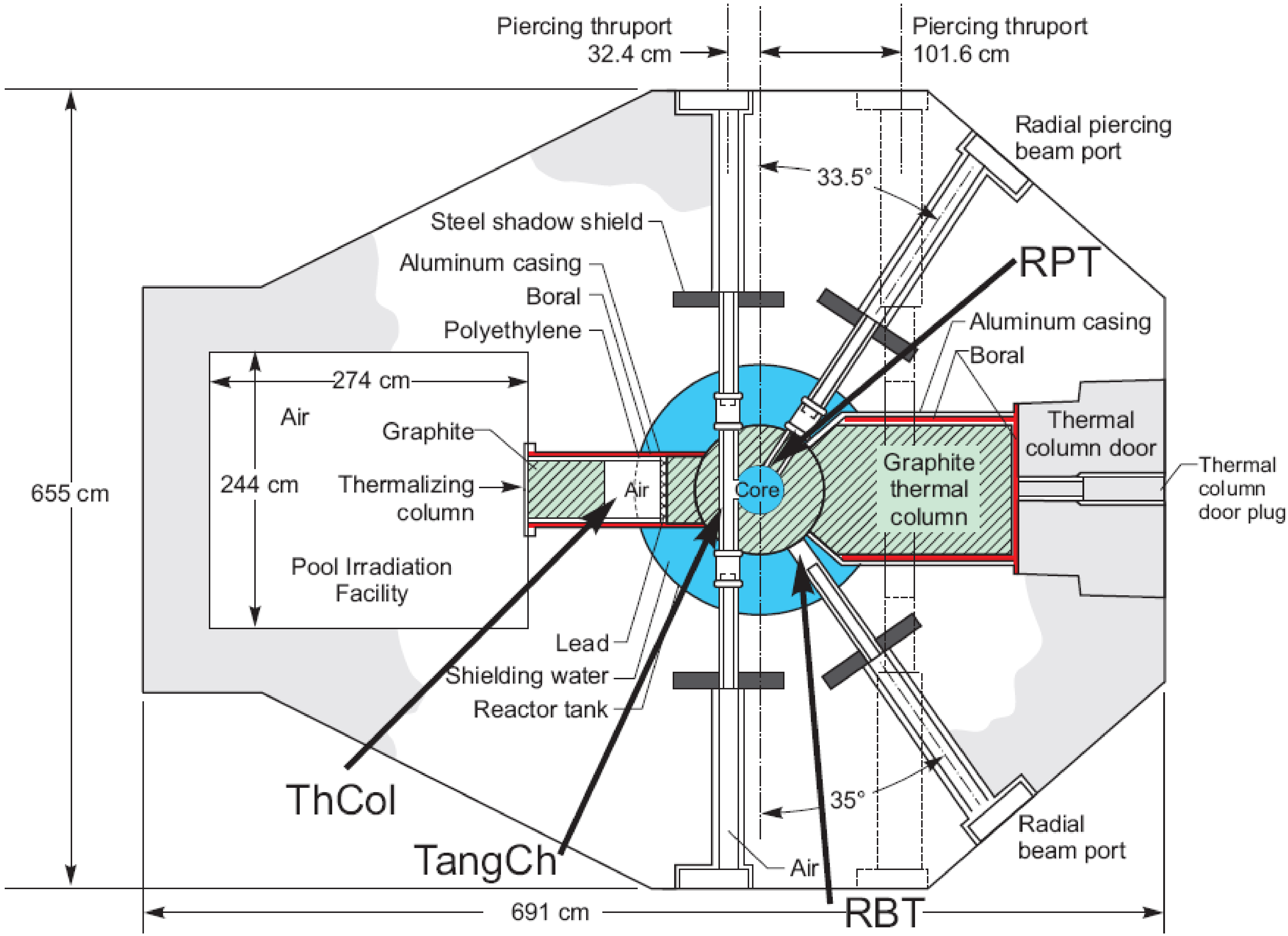
- research reactor used for:
  - **T**raining
  - **R**esearch
  - **I**sotope production
- manufactured by
  - **G**eneral **A**tomics
- main advantages:
  - simple design
  - inherently safe
  - easy to operate
  - reletively cheap

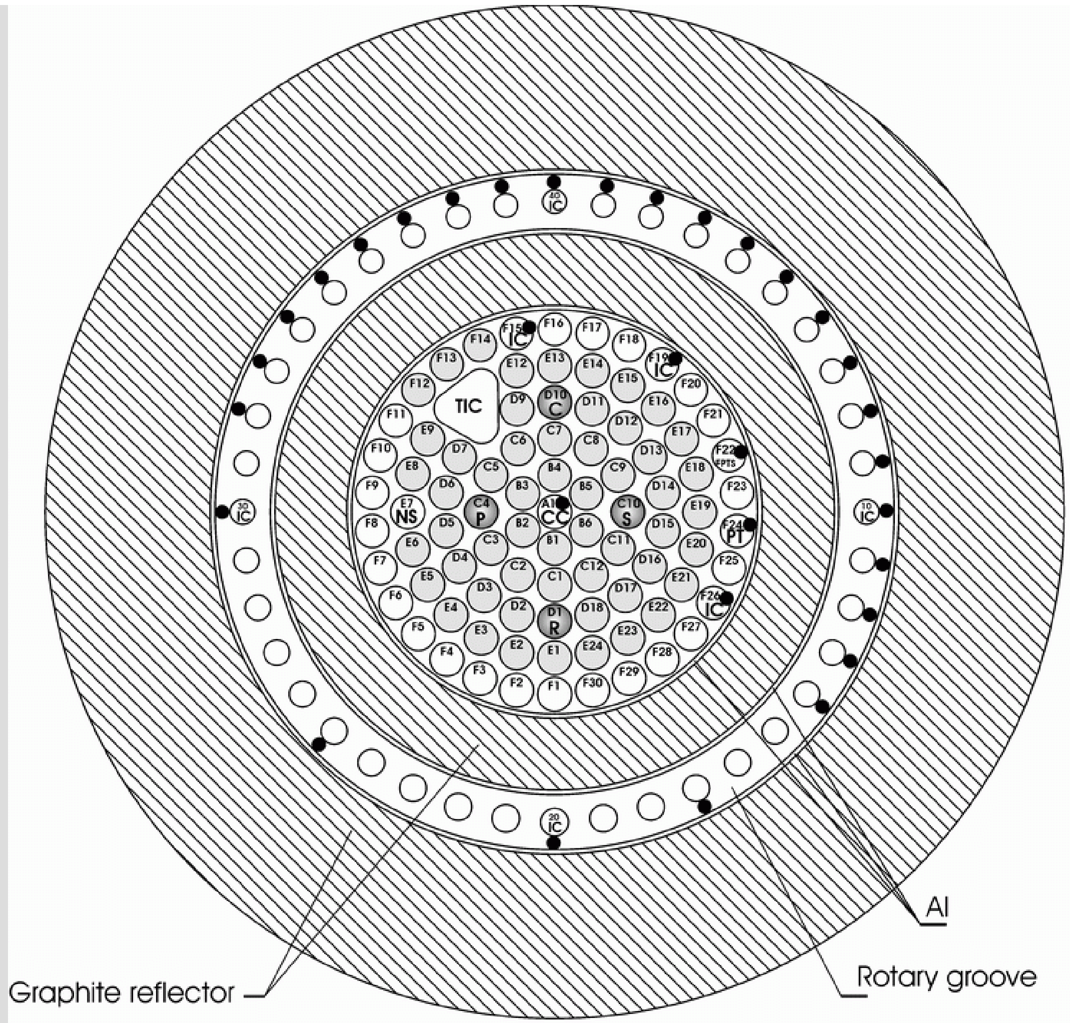
TRIGA Mark II at
Jozef Stefan Institute,
Ljubljana, Slovenia
(max. power ≈ 250 kW)





Standard concrete
Aluminum tank
91.4 cm (typ)
198.1 cm
655 cm
~490 cm
274 cm
264 cm
Bulk-shielding experimental tank (empty)
Heavy concrete
Boral
Aluminum casing
366 cm
370 cm
Core
Boral
Polyethylene
Air
Graphite
Heavy concrete door on track
Door plug
Aluminum casing
Lead
Heavy concrete


Piercing thruport
32.4 cm
Piercing thruport
101.6 cm
Radial piercing
beam port
33.5°
Steel shadow shield
Aluminum casing
Boral
Polyethylene
RPT
Aluminum casing
Boral
274 cm
Air
Graphite
244 cm
Thermalizing
column
Air
Core
Thermal
column door
Graphite
thermal
column
Thermal
column
door plug
655 cm
Pool Irradiation
Facility
Lead
Shielding water
Reactor tank
ThCol
TangCh
35°
Radial
beam port
Air
RBT
691 cm



- Fuel elements 20 % ^{235}U
- Control rods
- NS – Neutron source
- IC – Irradiation channels
- FPTS – Fast pneumatic transfer system
- PT – Pneumatic transport tube channel
- CC – Central irradiation channel
- TIC – Triangular irradiation channel

JSI TRIGA - applications

- source of neutrons
- neutron activation analysis (NAA)
- operator training
- neutron radiography
- isotope production
- testing of electronic components
- irradiation of particle detectors (CERN)

Neutron spectrum characterisation project

- Gain knowledge about neutronic properties of the reactor.
- Develop reactor analysis tools and methods.
- Characterise neutron spectrum of irradiation facilities.
- Improve the accuracy and reliability of NAA.
- **Resolve some discrepancies between nuclear constants for NAA and differential data.**

- Related to an IAEA coordinated research project.
- Related to bilateral collaboration with CEA.

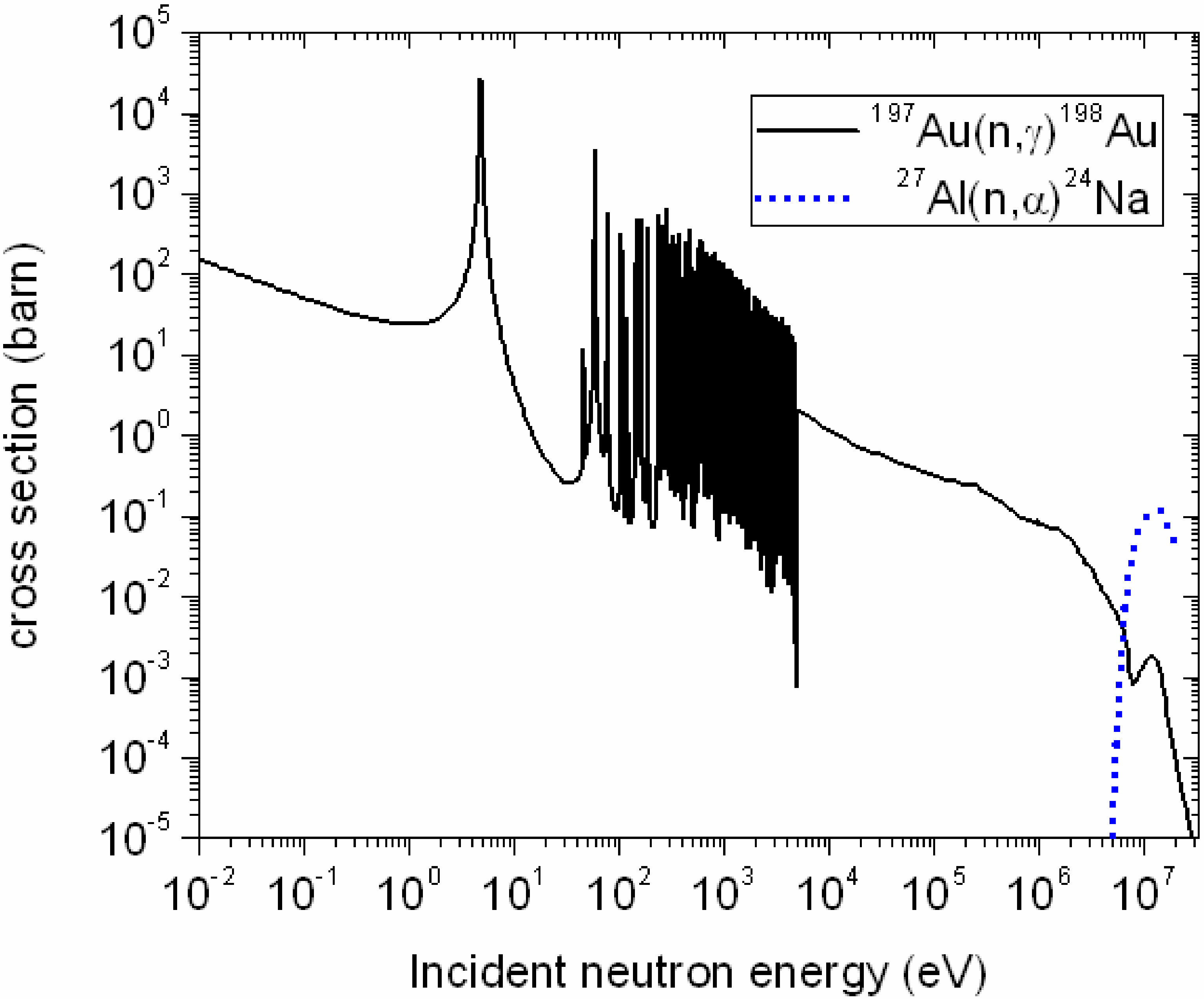
Methods

- Detailed computational model
- Series of experiments:
  - validation of computational model for spatial neutron flux distribution
  - neutron spectrum unfolding by combination of calculated spectra and multi-monitor foil activation
  - Validation of cross sections

Experiment 1

Validation of spatial neutron flux distribution

- disks of Al (99.9 w/o)-Au (0.1 w/o)
- 33 irradiation channels (6 in the core, 27 in the carousel facility in the reflector)
- irradiation for 73 min at 250 kW
- measured activity of $^{198}Au(n,\gamma)^{198}Au$ and $^{27}Al(n,\alpha)^{24}Na$



cross section (barn)
Incident neutron energy (eV)
$^{197}Au(n,\gamma)^{198}Au$
$^{27}Al(n,\alpha)^{24}Na$
10^{5} 10^{4} 10^{3} 10^{2} 10^{1} 10^{0} 10^{-1} 10^{-2} 10^{-3} 10^{-4} 10^{-5}
10^{-2} 10^{-1} 10^{0} 10^{1} 10^{2} 10^{3} 10^{4} 10^{5} 10^{6} 10^{7}

Experiment 2

Neutron spectrum unfolding

- 4 irradiation channels (1 core centre, 2 core periphery, 1 carrousel facility in the reflector)
- monitors
  - Al (99.9 w/o)-Au (0.1 w/o)
  - Ni (80.93 w/o)- Mo (15.16 w/o)-W (2.76 w/o)- Mn (0.41 w/o)- Au(0.29 w/o)
  - Zr (99.8 w/o), Zn (99.99 w/o), W(99.95 w/o), Fe
  - Al (99.55 w/o)-U (0.449 w/o)
  - Al (99.18 w/o)-Th (0.819 w/o) …
- reactions ...

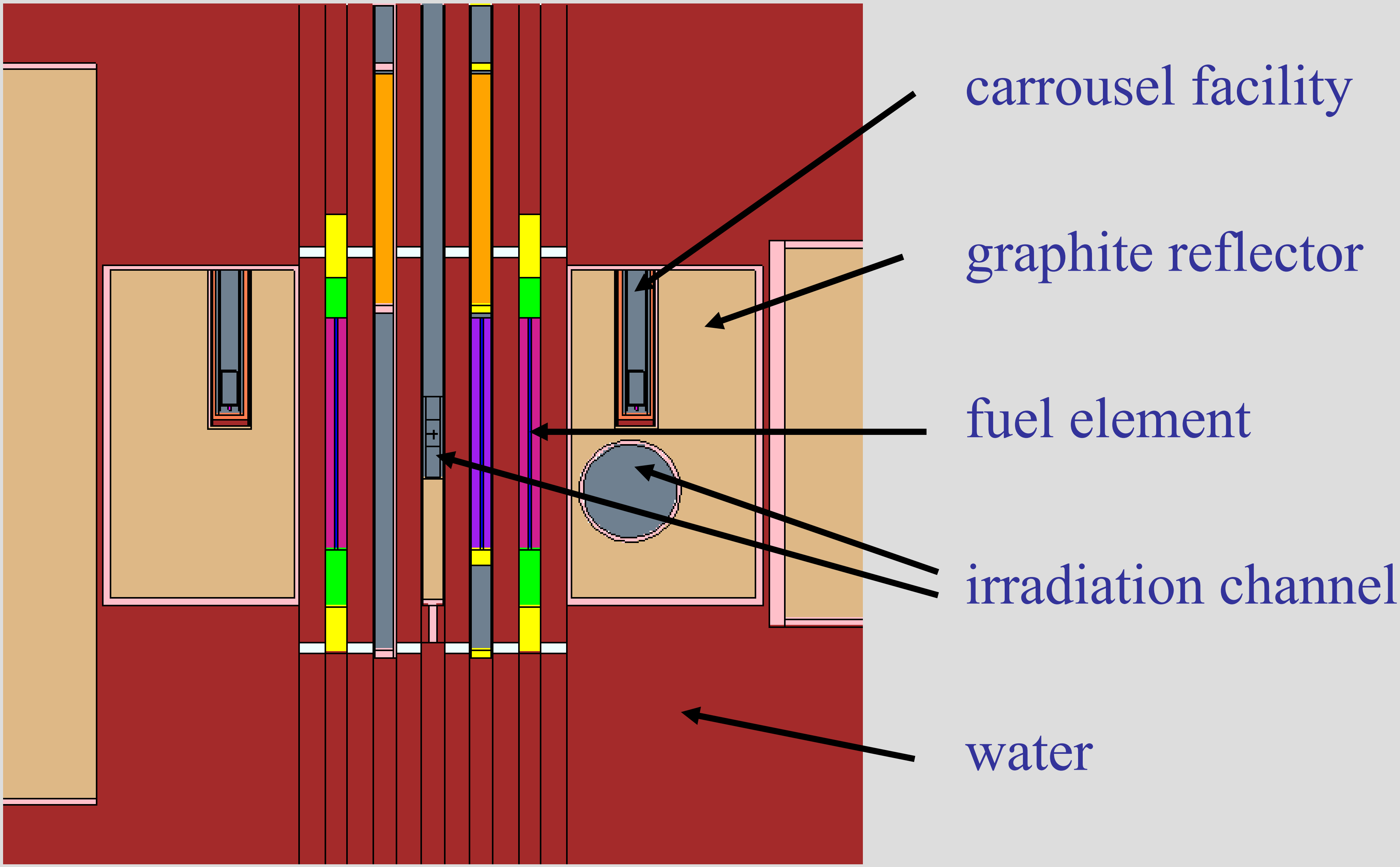
Experiment 2 (cont'd)

- reactions
  - ^{27}Al(n,α), ^{27}Al(n, γ), ^{197}Au(n,γ)
  - ^{58}Ni(n,p), ^{92}Mo(n,p), ^{64}Ni(n,γ), ^{98}Mo(n,γ), ^{100}Mo(n,γ), ^{55}Mn(n,γ), ^{186}W(n,γ), ^{198}Au(n,γ)
  - ^{90}Zr(n,p), ^{90}Zr(n,2n), ^{94}Zr(n, γ), ^{96}Zr(n, γ)
  - ^{66}Zn(n,p), ^{64}Zn(n,γ), ^{68}Zn(n,γ), ^{70}Zn(n,γ)
  - ^{238}U(n,γ), ^{232}Th(n,γ), ^{186}W(n,γ)

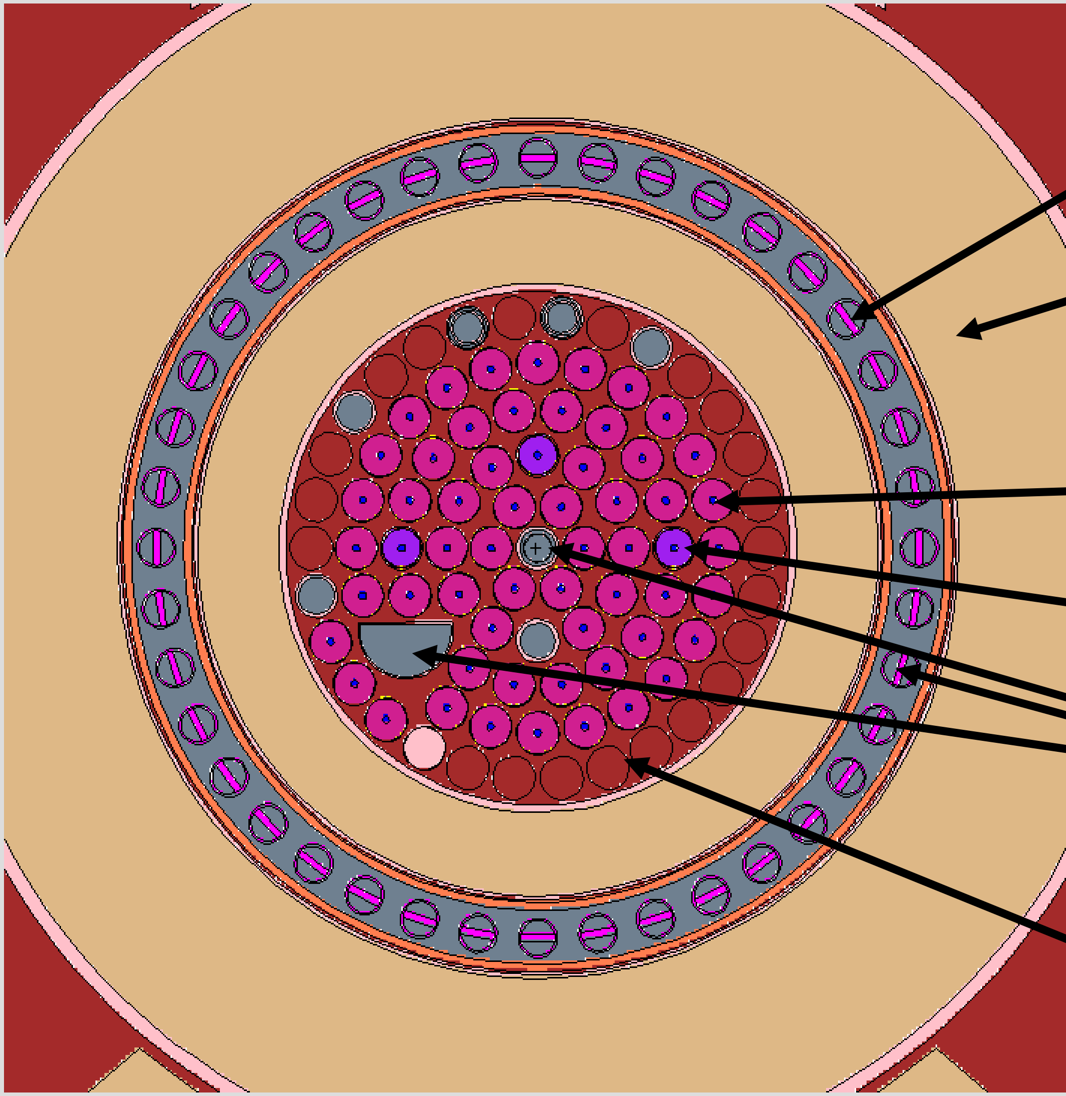
Computational model

- MCNP5 computer code
- detailed geometry model including structures in the reactor pool
- sensitivity study
  - structures around the core
  - temperature
  - fuel burnup, xenon
  - reflector density
  - impurities in the reflector

MCNP model – side view



MCNP model – top view



- carrousel facility
- graphite reflector
- fuel element
- control rod
- irradiation channels
- water

Status

Computational model development

- geometrical model completed
- sensitivity study completed

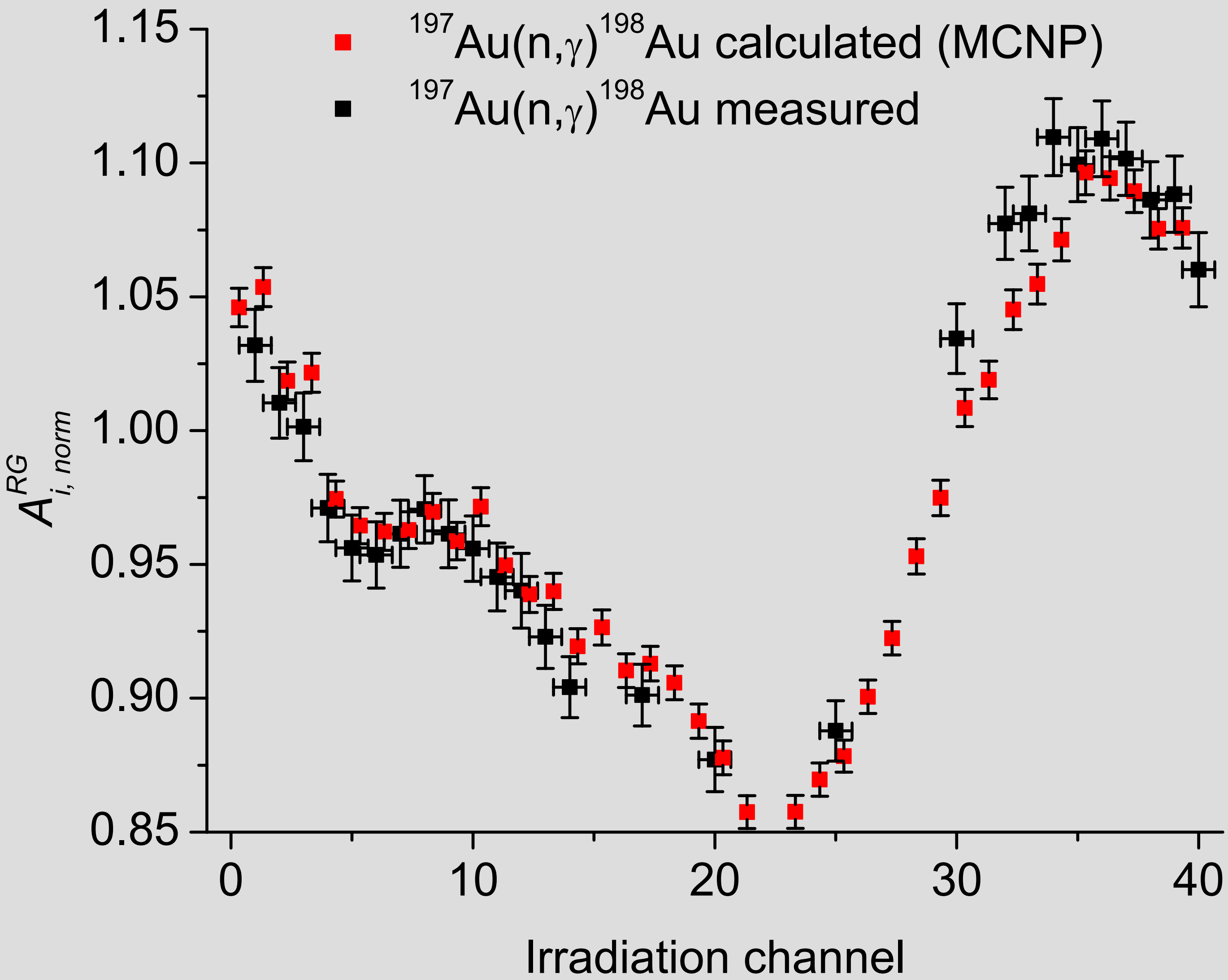
1st experiment

- measurement and analysis completed

2nd experiment

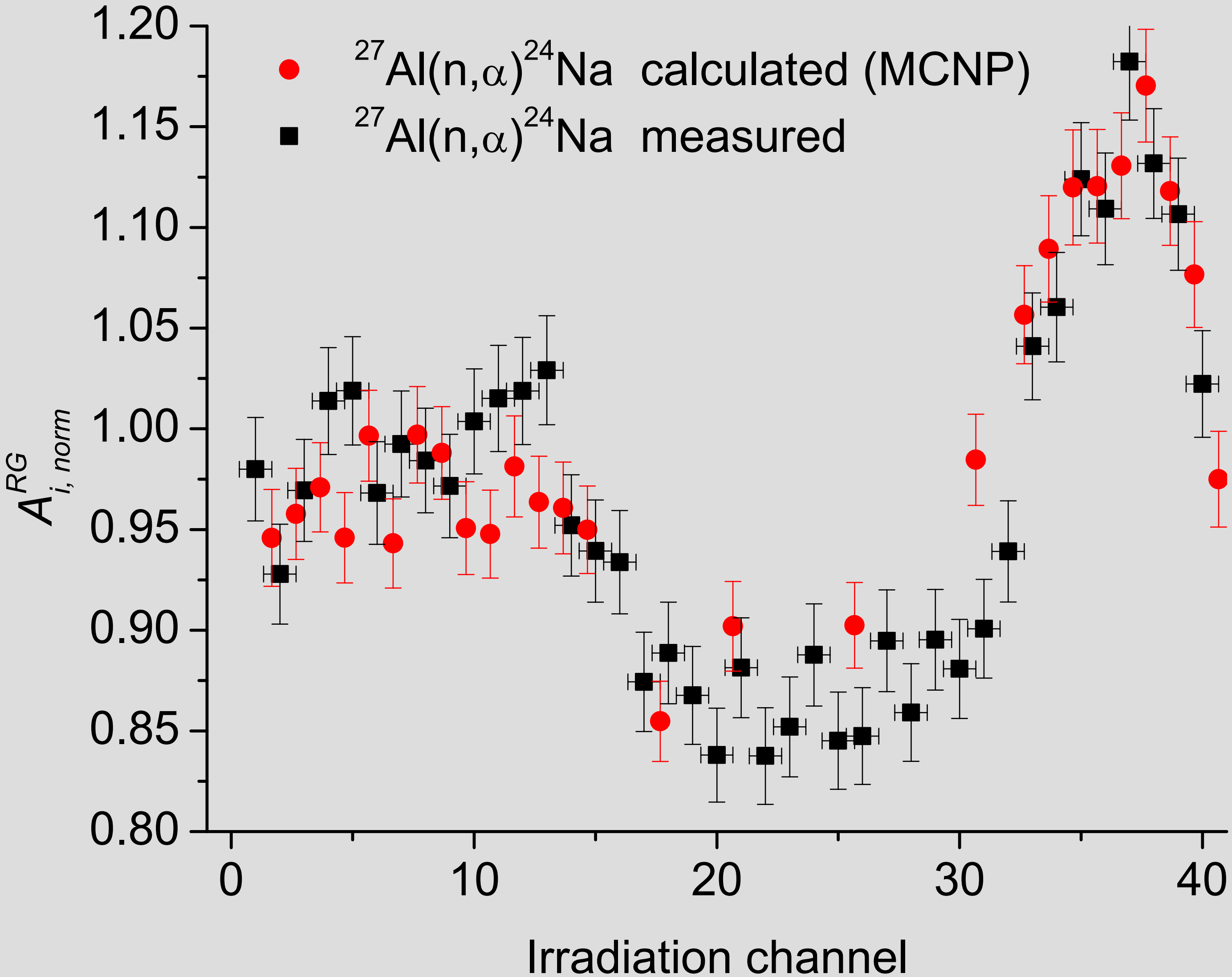
- measurement completed, analysis in progress

Results



$^{197}Au(n,\gamma)^{198}Au$ calculated (MCNP)
$^{197}Au(n,\gamma)^{198}Au$ measured
$A^{RG}_{i,\,norm}$
1.15
1.10
1.05
1.00
0.95
0.90
0.85
0
10
20
30
40
Irradiation channel

Results



$^{27}Al(n,\alpha)^{24}Na$ calculated (MCNP)
$^{27}Al(n,\alpha)^{24}Na$ measured
$A^{RG}_{i,\,norm}$
Irradiation channel
1.20
1.15
1.10
1.05
1.00
0.95
0.90
0.85
0.80
0
10
20
30
40

Results

C_a - Neutron flux attenuation factor in carrousel facilty relative to central channel (CC).

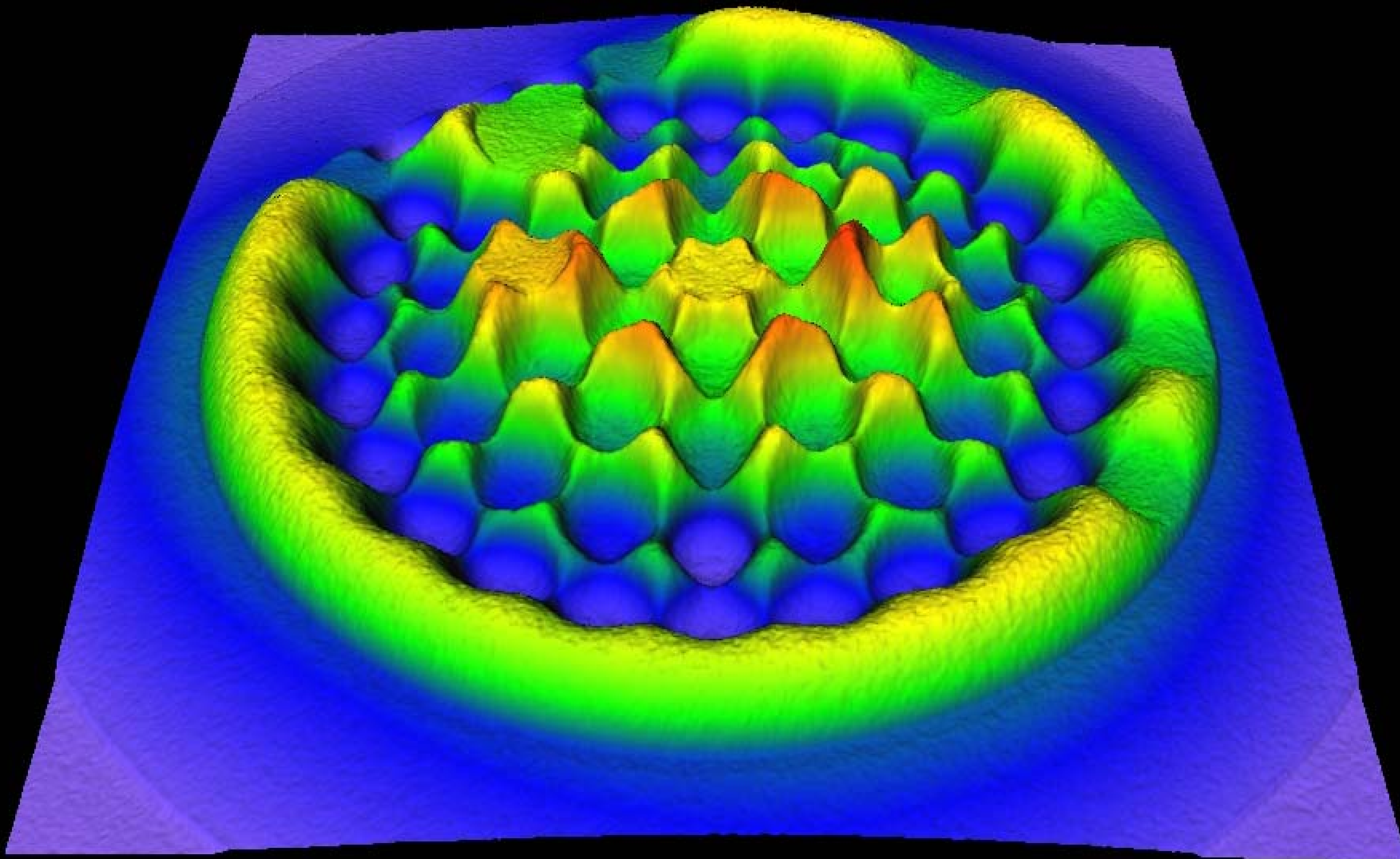
$$C_a = \frac{\bar{\mathrm{A}}_{\mathrm{carroussel}}}{\mathrm{A}_{\mathrm{central\ channel}}}$$

	fast flux $^{27}Al(n,\alpha)^{24}Na$	thermal flux $^{197}Au(n,\gamma)^{198}Au$
measured	0.02795 ± 0.00068	0.12868 ± 0.00190
calculated	0.02780 ± 0.00005	0.14228 ± 0.00003
rel. diff. [%]	-0.5 ± 2.4	10.6 ± 1.5

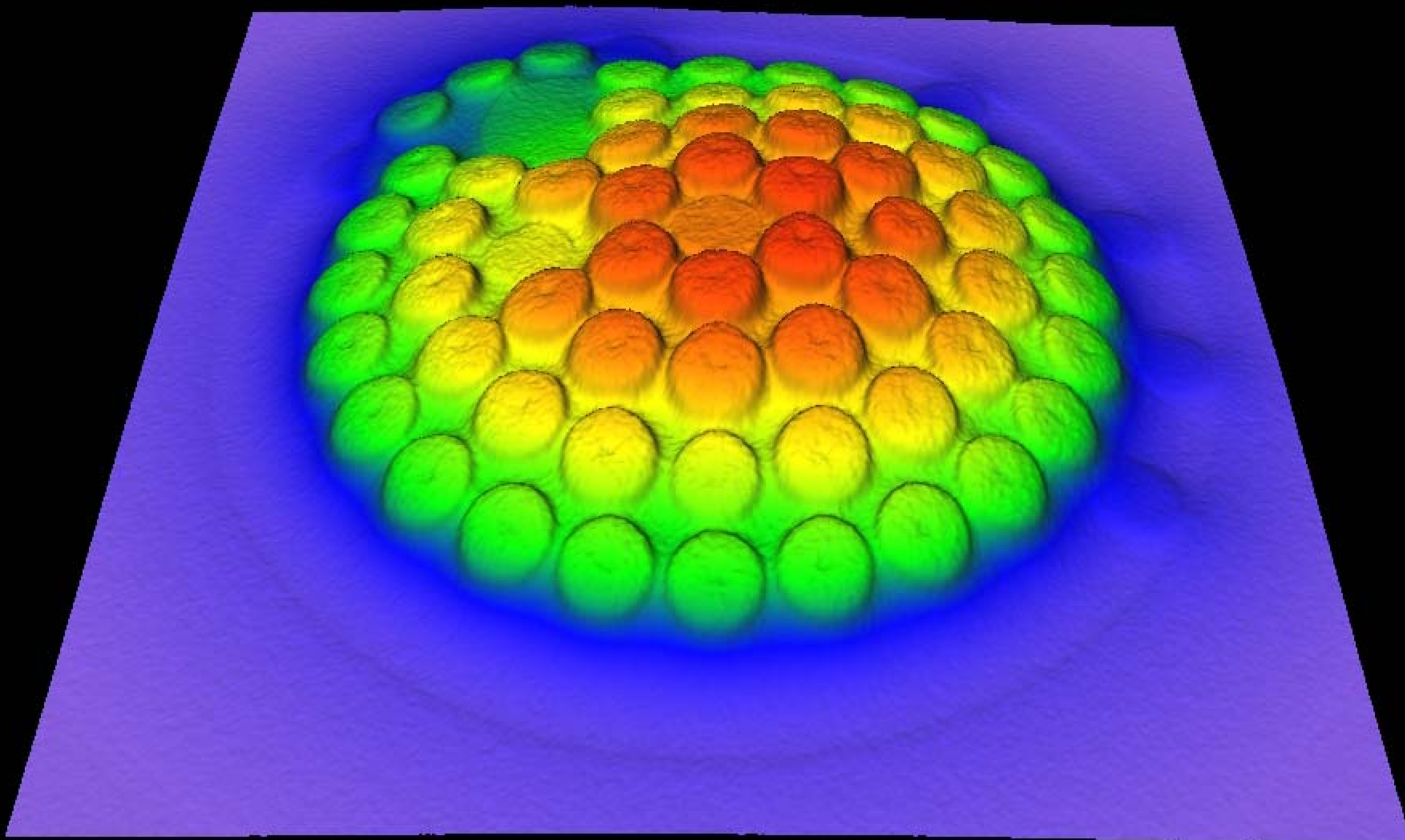
Conclusions on Experiment 1

- thermal flux in the carrousel facility drops by a factor of 7 compared to the central channel
- calculations underpredict the thermal flux attenuation in the reflector by approx. 10 %
- very good prediction of fast flux attenuation
- discrepancies between measurements do not exceed 5 % for thermal flux and 10 % for fast flux
- neutron spectra analysis is in progress – neutron self-shielding is investigated in detail

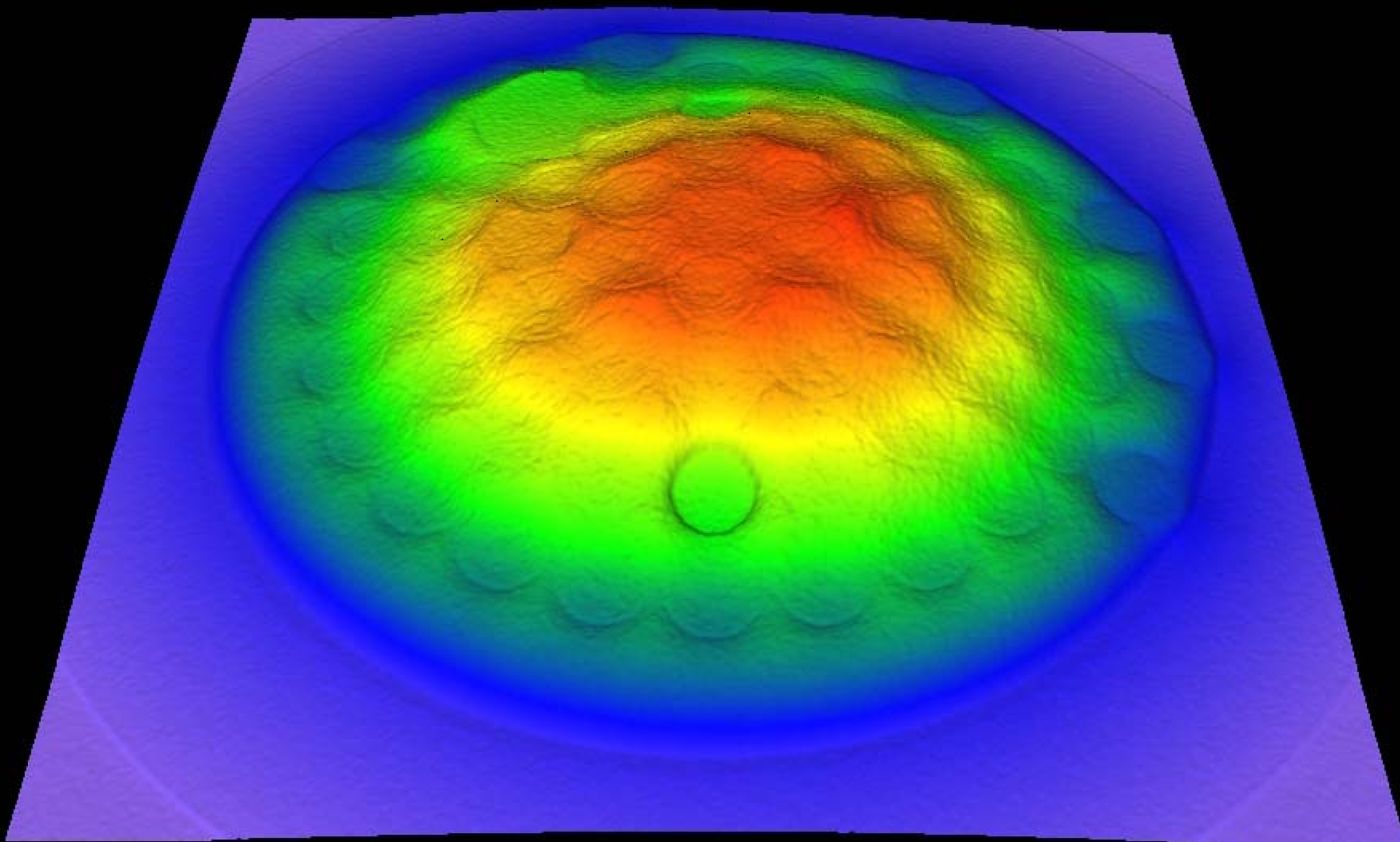
Thermal neutron flux



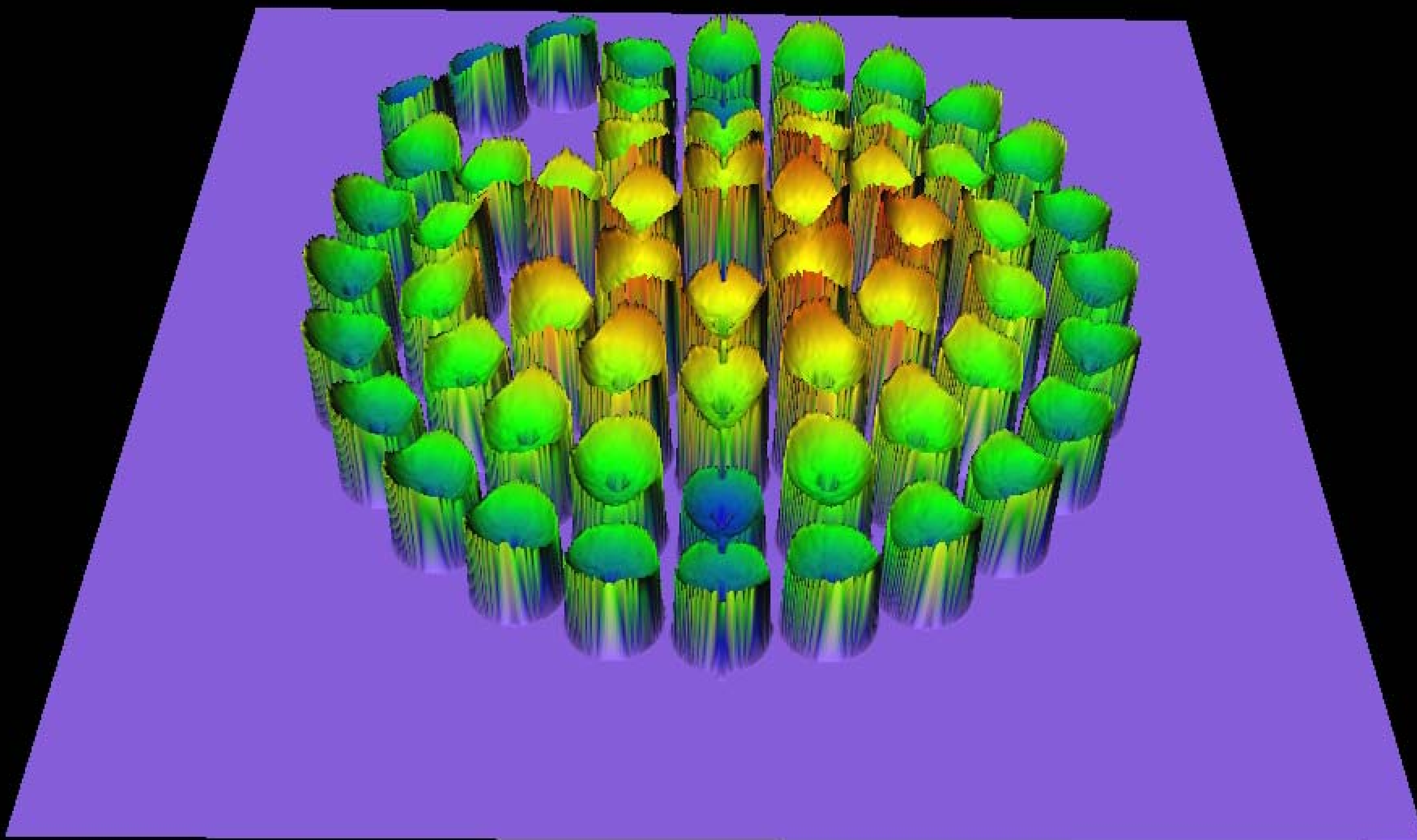
Intermediate neutron flux



Fast neutron flux



Power



Neutron spectrum characterisation

- Detailed spectrum in 640-groups by MC calculation
- Fit by smooth analytical function (up to 19 parameters)
- Modulation of smooth function to restore detailed shape
- Adjust parameters of analytical function to improve agreement with measured activities

Spectrum analytic function

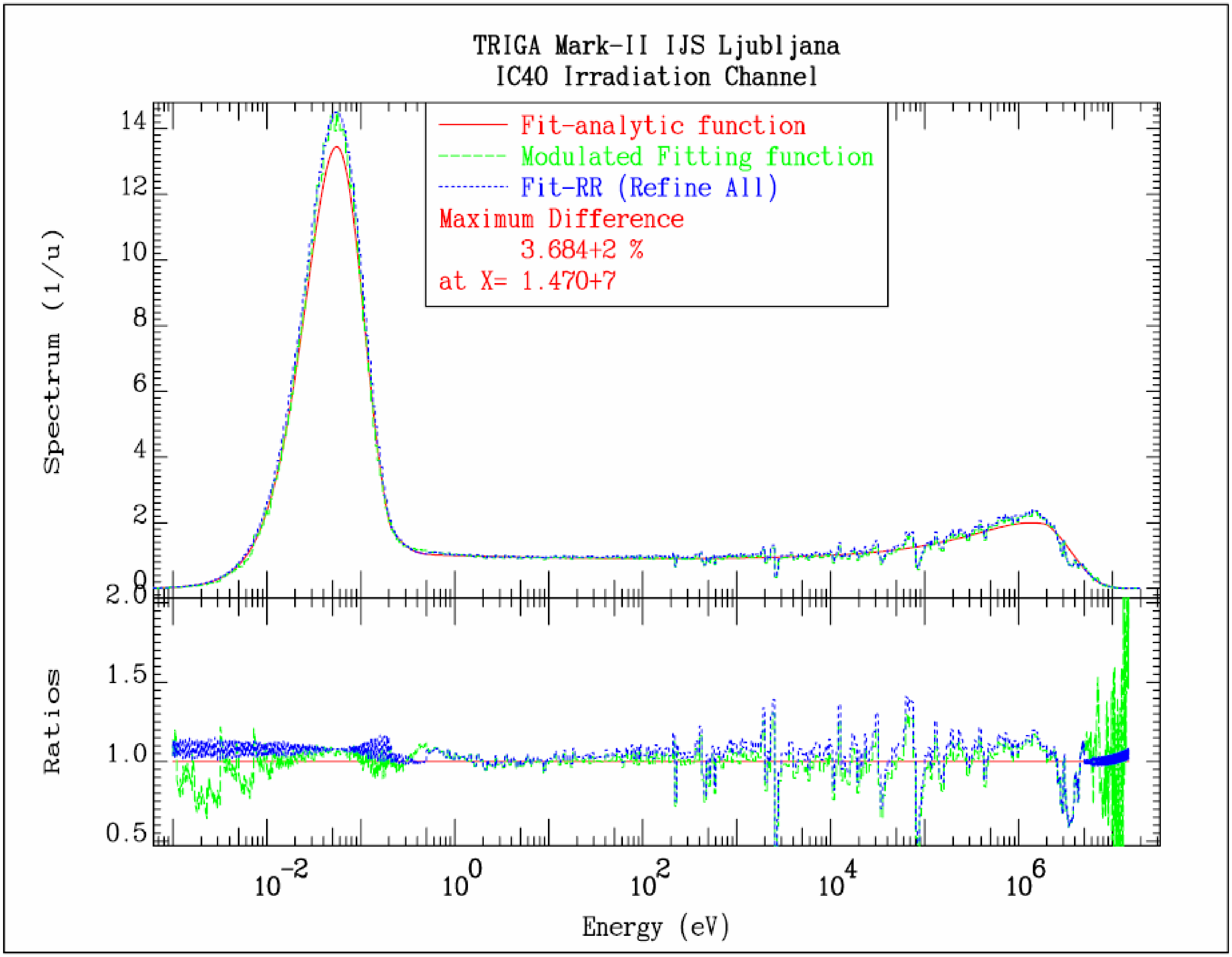
$$\psi_t = C_t\, E^n \left[e^{-E/kT} + C_{t1} e^{-E/kT_1} + C_{t2} e^{-E/kT_2} \right]$$

$$\psi_e = E^{-\left[1+\alpha_0+\alpha_1 \log E + \alpha_2 (\log E)^2\right]}$$

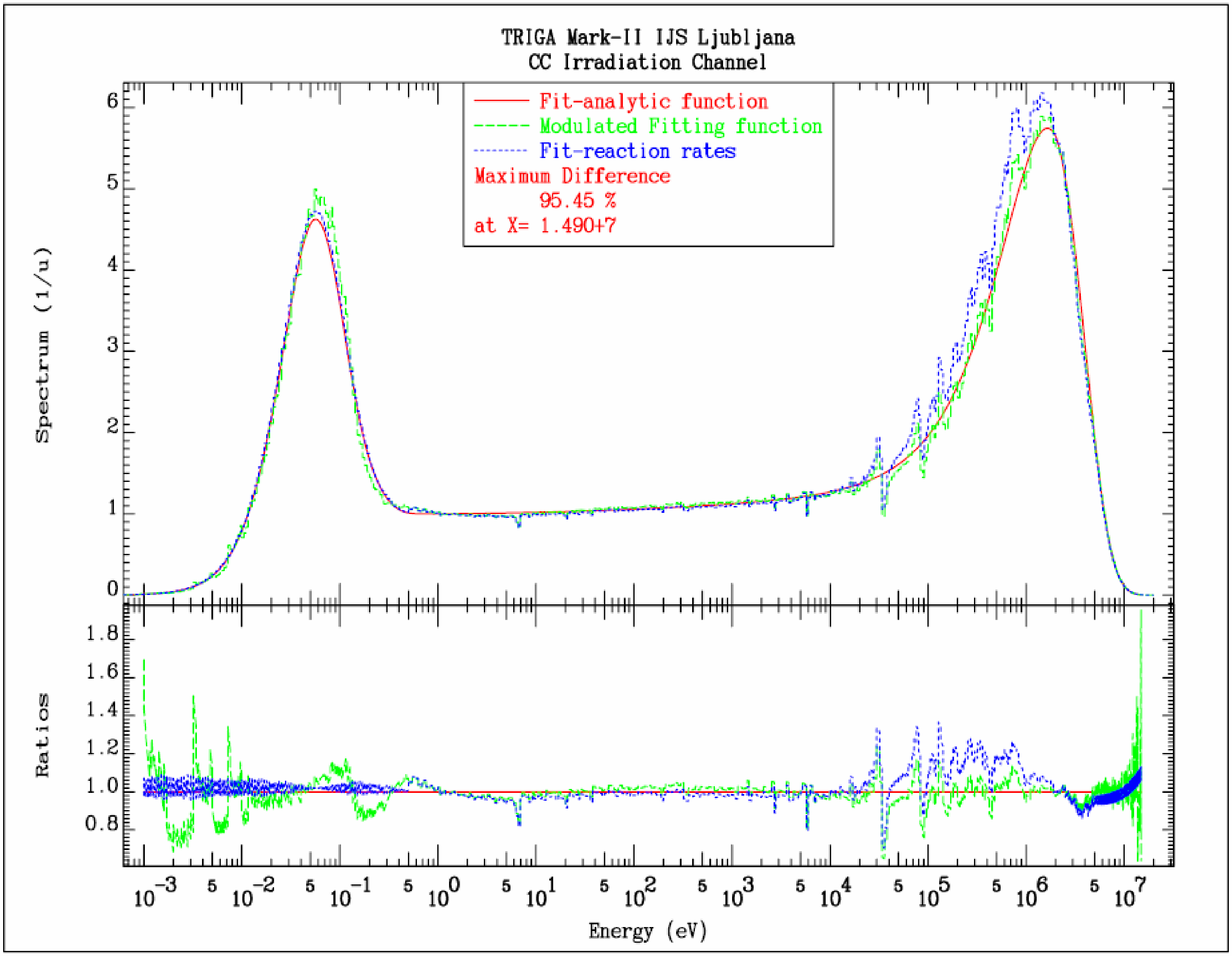
$$\psi_f = C_f e^{-E/W} \sinh\left(\sqrt{EW_b}\right) \cdot \frac{1}{E^{m_0+m_1 E}} \quad or$$

$$C_f \sqrt{E}\, e^{-E/E_T} \cdot \frac{1}{E^{m_0+m_1 E}}$$

$$\psi = K_t \psi_t + K_e \psi_e + K_f \psi_f$$



TRIGA Mark-II IJS Ljubljana
IC40 Irradiation Channel
Fit-analytic function
Modulated Fitting function
Fit-RR (Refine All)
Maximum Difference
3.684+2 %
at X= 1.470+7
Spectrum (1/u)
Ratios
Energy (eV)


TRIGA Mark-II IJS Ljubljana
CC Irradiation Channel
Fit-analytic function
Modulated Fitting function
Fit-reaction rates
Maximum Difference
95.45 %
at X= 1.490+7
Spectrum (1/u)
Ratios
Energy (eV)

Results – Central Channel (CC)

Comparison of measured and calculated react. Rates (prelim.)

Target		Ratio			Ratio	Dif[%]	Unc[%]
Al- 27g	(n,g)	Tot/Tot	Au-197g	(n,g)	9.040E-4	1.3	(2.8)
Al- 27g	(n,p)	Tot/Tot	Au-197g	(n,g)	1.697E-5	0.7	(2.8)
Al- 27g	(n,a)	Tot/Tot	Au-197g	(n,g)	3.232E-6	-0.3	(2.9)
Zr- 90g	(n,2n)	Tot/Tot	Au-197g	(n,g)	5.235E-7	-1.9	(3.0)
Mo-100g	(n,g)	Tot/Tot	Au-197g	(n,g)	2.672E-3	1.6	(3.8)
Mo- 98g	(n,g)	Tot/Tot	Au-197g	(n,g)	3.660E-3	-1.3	(2.9)
Zn- 64g	(n,p)	Tot/Tot	Au-197g	(n,g)	1.624E-4	-2.7	(3.0)
Al- 27g	(n,a)	Tot/Tot	Au-197g	(n,g)	3.232E-6	3.3	(2.9)
U -238g	(n,g)	Tot/Tot	Au-197g	(n,g)	0.1170	0.7	(2.8)
U -238g	(n,g)	Tot/Tot	Au-197g	(n,g)	0.1170	0.2	(2.8)
Al- 27g	(n,g)	Tot/Tot	Au-197g	(n,g)	9.040E-4	0.0	(3.0)
Al- 27g	(n,g)	Tot/Tot	Au-197g	(n,g)	9.040E-4	-3.9	(3.2)
Au-197g	(n,g)	Tot/Cad	Au-197g	(n,g)	1.6102	0.6	(2.8)
U -238g	(n,g)	Tot/Cad	U -238g	(n,g)	1.1268	0.1	(2.9)
U -238g	(n,g)	Tot/Cad	U -238g	(n,g)	1.1268	1.1	(2.9)
U -238g	(n,g)	Tot/Cad	U -238g	(n,g)	1.1268	0.2	(2.9)
U -238g	(n,g)	Tot/Cad	U -238g	(n,g)	1.1268	3.3	(2.9)

Results (Cont.)

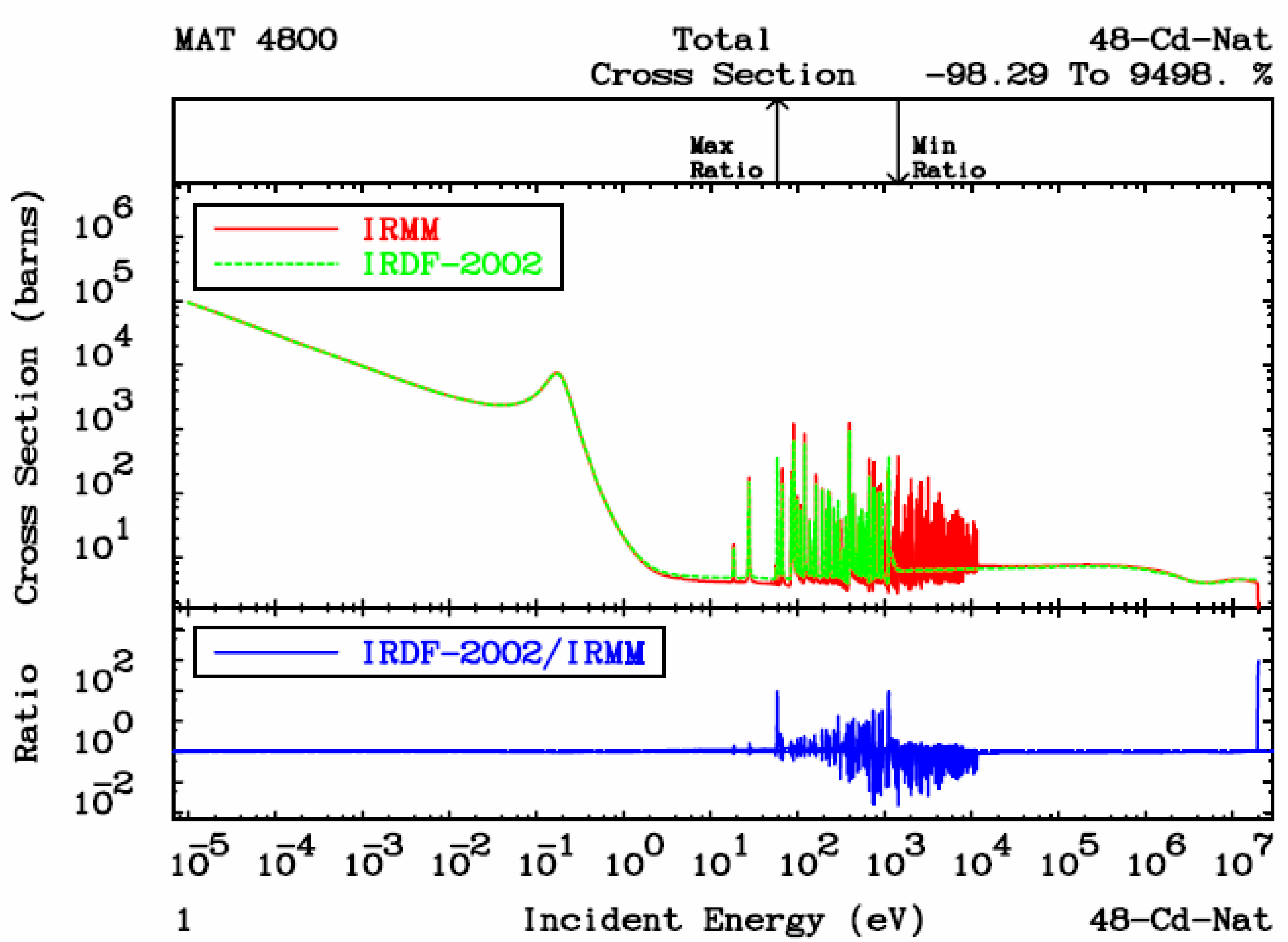
- More than 190 measured points accumulated (different gamma lines, samples, measuring campaigns)
- 17 measurements selected (reliability of cross sections, optimal measuring conditions, known problems, etc.).
- Selected measurements reflect precision and reproducibility.
- Similar analysis for IC40 channel.

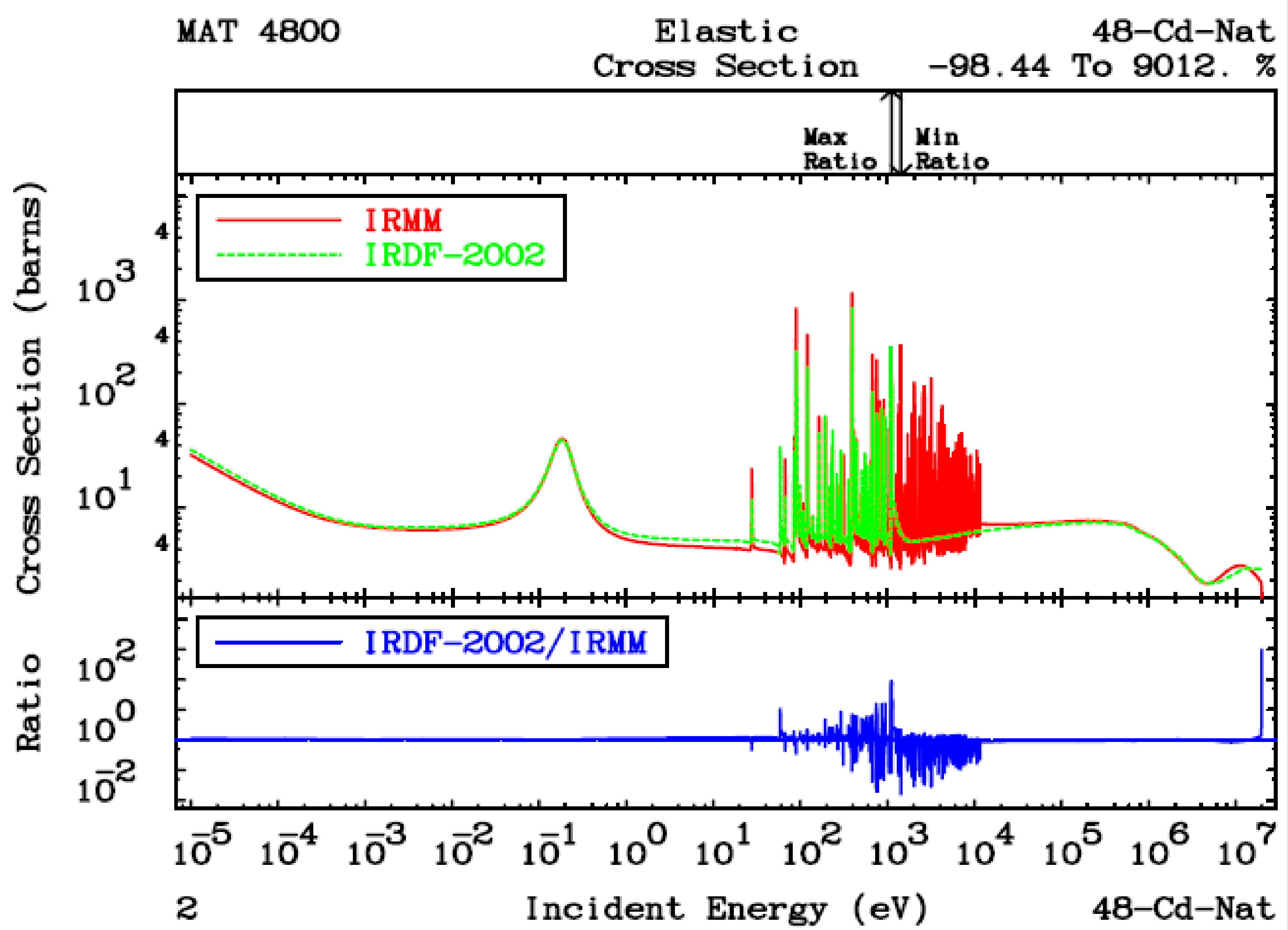
Data validation by activation experiments

- Data validation is the inverse of spectrum unfolding
- Reliable (trusted) data are used to adjust the spectrum
- Reliable spectrum can be used to adjust uncertain cross section data

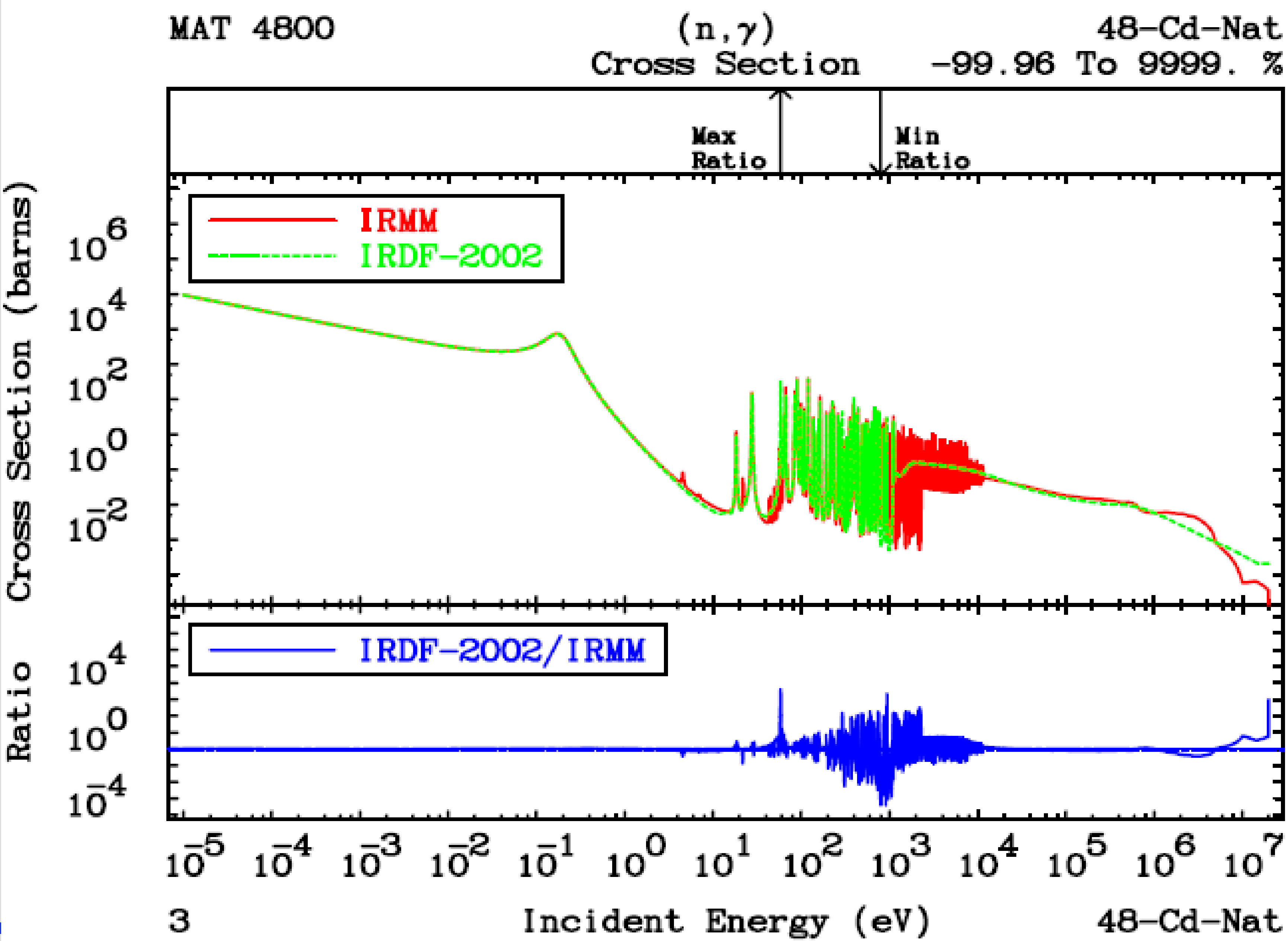
Cadmium data verification

- Evaluated isotopic data files were obtained from IRMM
- Cross sections for natural element were reconstructed from isotopic data
- Comparison with IRDF-2002 (=IRDF90)


MAT 4800
Total
Cross Section
48-Cd-Nat
-98.29 To 9498. %
Max Ratio
Min Ratio
IRMM
IRDF-2002
IRDF-2002/IRMM
Cross Section (barns)
Ratio
Incident Energy (eV)
1
48-Cd-Nat



MAT 4800
Elastic
Cross Section
48-Cd-Nat
-98.44 To 9012. %
Max Ratio
Min Ratio
IRMM
IRDF-2002
IRDF-2002/IRMM
Cross Section (barns)
Ratio
Incident Energy (eV)
2
48-Cd-Nat


MAT 4800
(n,γ)
Cross Section
48-Cd-Nat
-99.96 To 9999. %
Max Ratio
Min Ratio
IRMM
IRDF-2002
IRDF-2002/IRMM
Cross Section (barns)
Ratio
Incident Energy (eV)
3
48-Cd-Nat

Definitions

$$F_{cd} = \frac{\int_0^{\infty} t(E)\sigma(E)\varphi(E)\,dE}{\int_{E_{cd}}^{E_3} \sigma(E)\varphi(E)\,dE}$$

Cadmium factor

$$t(E) = e^{-N_{cd}\,d\,\sigma_{cd}(E)}$$

Flux attenuation due to Cd

$$N_{cd} = \frac{\rho_{cd} N_A}{M_{cd}}$$

Cd number density

Differences in F_{Cd} > 1%

	IRDF-90	IRMM	Difference [%]	
56-Ba-130g	0.862	0.998	15.8	Interference?
48-Cd-110g	0.751	0.782	4.1	
27-Co- 58g	1.520	1.481	-2.6	
48-Cd-114g	0.903	0.925	2.4	
56-Ba-140g	0.878	0.897	2.2	
46-Pd-107g	0.958	0.977	1.9	
34-Se- 74g	0.861	0.877	1.9	
34-Se- 78g	0.952	0.969	1.8	
90-Th-232g	0.976	0.992	1.6	First resonance
37-Rb- 86g	0.954	0.969	1.6	
96-Cm-242g	0.972	0.987	1.5	
54-Xe-132g	0.963	0.975	1.3	
48-Cd-112g	0.932	0.920	-1.2	
48-Cd-111g	0.892	0.903	1.2	

Current problems

Analysis of self-shielding factors:

- Paper published recently in NIM (Trkov et al: On the self-shielding factors in neutron activation analysis)
- Measurements on pure Rh samples of different thickness
- Verification of simple MATSSF approach
- Validation with detailed Monte Carlo calculations

Rhodium data validation

- Two samples of different thicknesses were obtained from IRMM.
- They were irradiated and measured.
- More samples of different thicknesses will be analysed to validate the self-shielding correction procedures.
- Consistency of differential data and integral measurements will be tested.

Steps

- Check consistency using existing data.
- Compare with differential data.
- Derive improved constants.

- Preliminary analysis completed.
- Further work is in progress.

Calculated concentrations

Sample	Irr.Ch.	Bare Conc.(%)	Cd-box Conc.(%)	
Rh1	PT	87.9	79.3	No corr.
0.006mm		91.3	90.4	MATSSF
		91.4	**90.9**	MCNP
	IC40	89.5	80.8	No corr.
		93.0	92.0	MATSSF
		93.1	**92.5**	MCNP
Rh2	PT	67.1	35.9	No corr.
0.112mm		90.3	96.1	MATSSF
		90.9	**92.3**	MCNP
	IC40	72.2	35.6	No corr.
		96.7	95.2	MATSSF
		97.4	**91.4**	MCNP

New Constants

Preliminary estimates:

$Q_0 = 6.37(12)$ -5.6 % from ref.

$k_0 = 6.68(8)\text{E-}2$ -3.5 % from ref.

Check E_r, F_{Cd}, g …

Check σ_0 and RI= $Q_0 . \sigma_0$ from differential data …

Note:

- Q_0 : no dependence on detector efficiency and gamma-ray emission probability P_g
- Conversion from k_0 to σ_0 requires assumptions about $P_{g.}$

Conclusion

- Combination of computational model and activation measurements works well for spectrum determination.
- Methodology for calculating self-shielding factors seems to work.
- Further measurements are in progress.
- **Integral activation measurements can be used for data validation.**