Characterization of nuclear material by Neutron Resonance Transmission Analysis (NRTA)

UCANS V

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P. Schillebeeckx

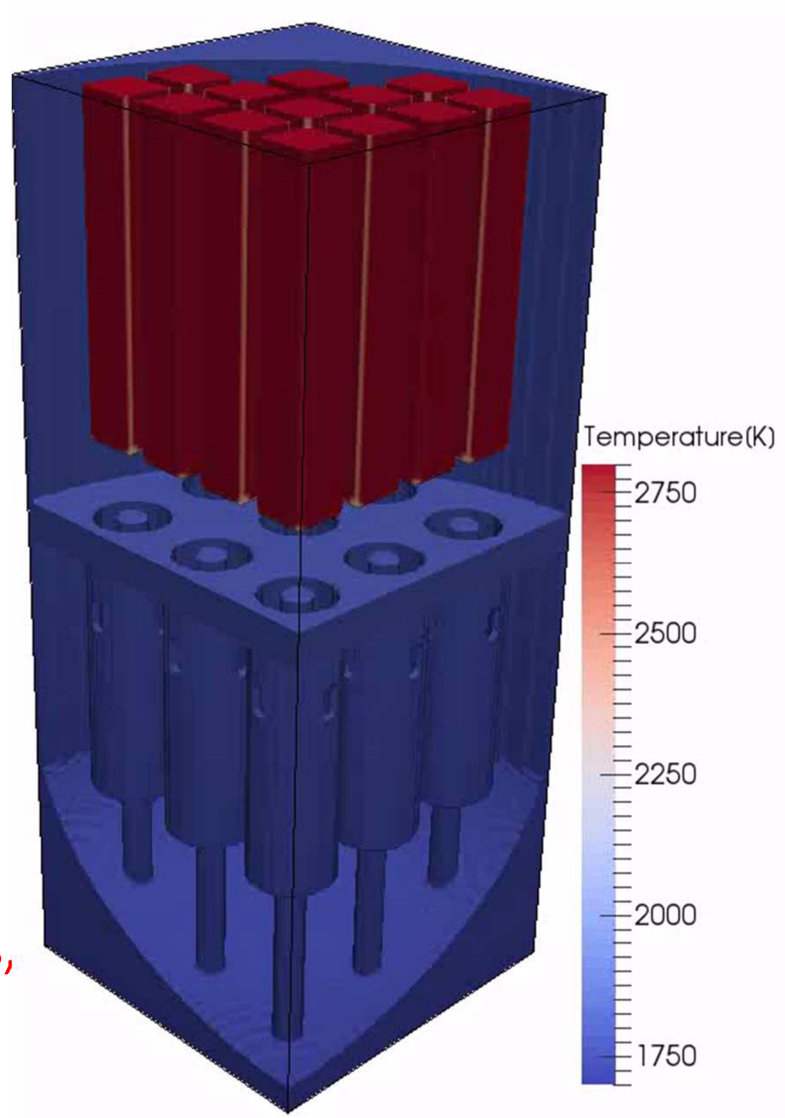
EC – JRC (Geel)

Standards for Nuclear Safety, Security and Safeguards (SN3S)

Fukushima accident

Earthquake followed by a Tsunami (15 m)

- **core meltdown (units 1,2,3)**
- production of hydrogen followed by an explosion



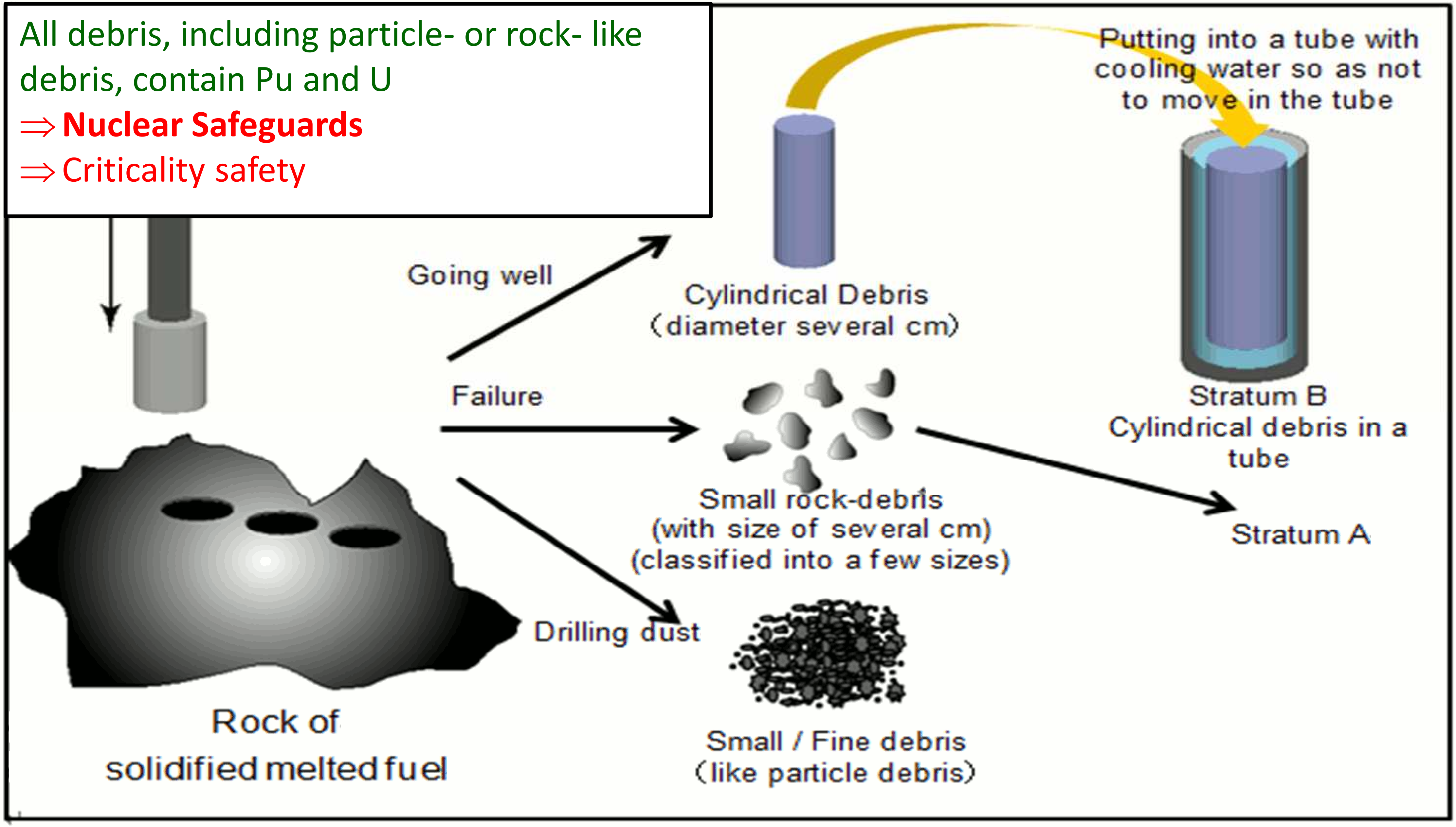
Melted fuel:
Complex mixture of materials in fuel and control/safety rods, i.e. U, Pu, fission products, structural materials and neutron absorbers (^{10}B)

Removal of melted fuel
huge amounts of debris will be produced

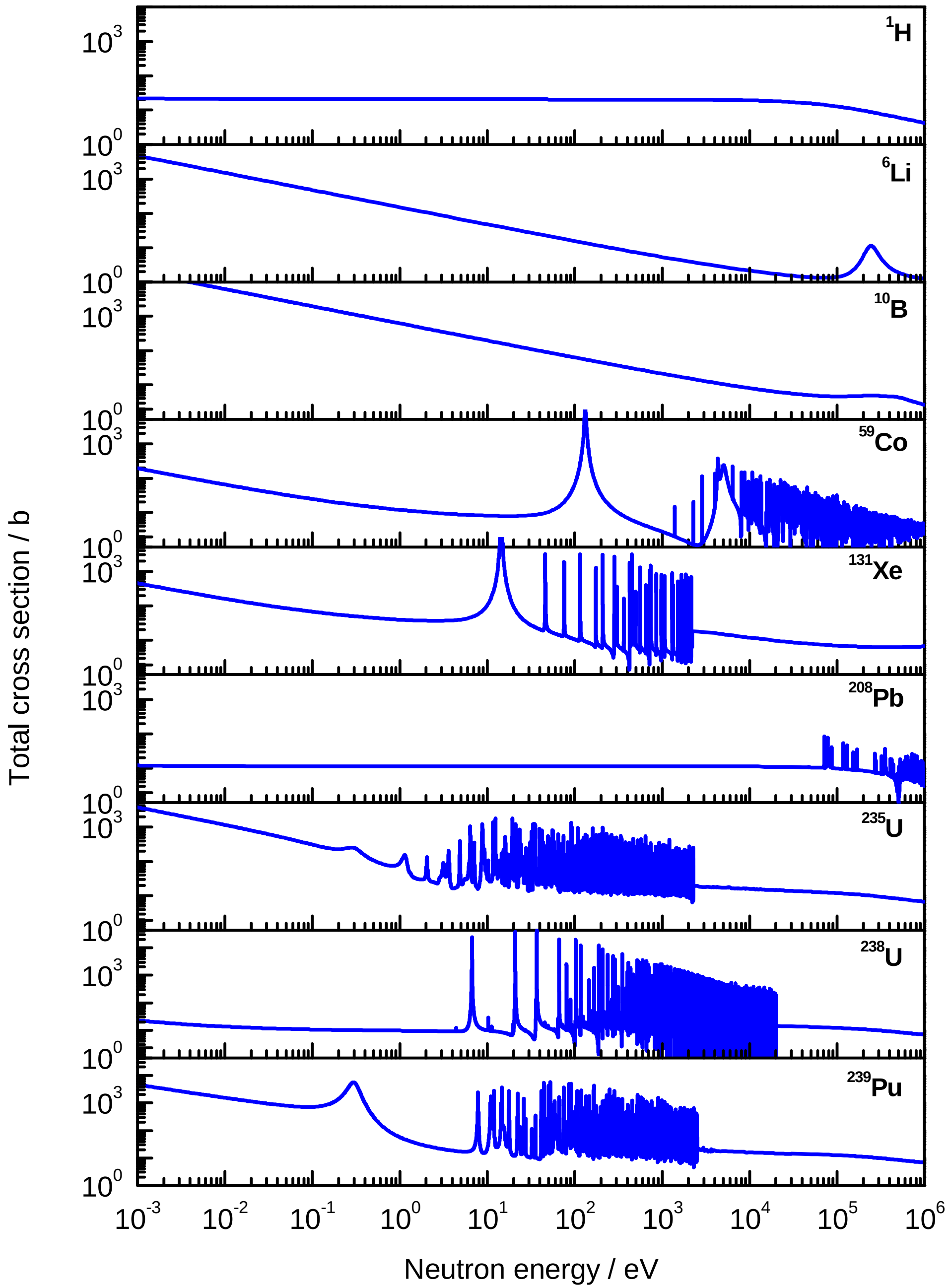
All debris, including particle- or rock- like debris, contain Pu and U

⇒ **Nuclear Safeguards**

⇒ Criticality safety



Neutron resonance transmission analysis NRTA

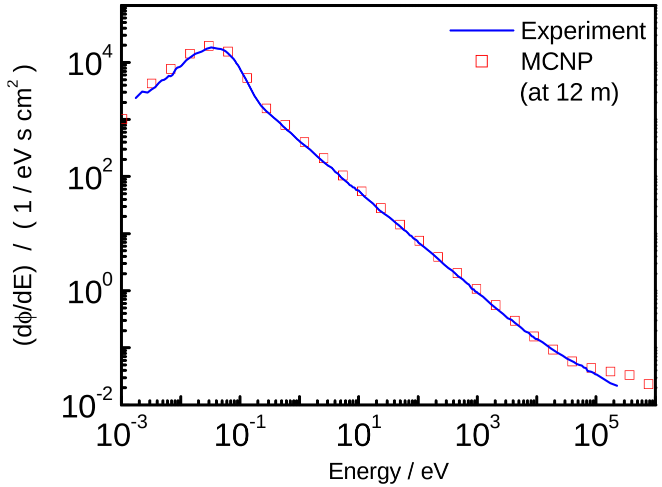


- Resonances appear at energies, which are specific for each nuclide
- Position and amplitude of resonances can be used as fingerprints to
  - identify and quantify nuclides
  - elemental & isotopic composition
- NRTA developed at the JRC - Geel
  - Non-Destructive Analysis (NDA)
  - sensitive to almost all nuclides (except light)
  - no sample preparation required
  - **requirements: TOF-measurements at a white neutron source**

Schillebeeckx et al., JRC Science and Policy Report, EUR 26848 EN

TOF – facility GELINA

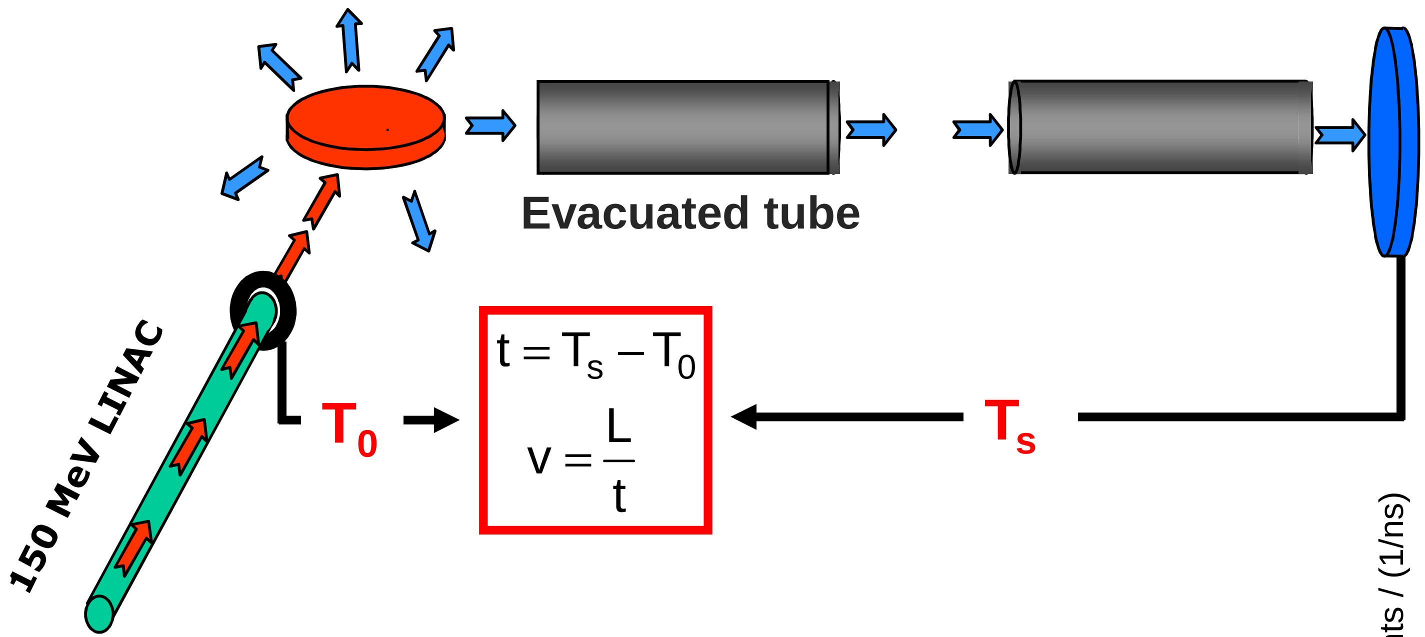
- Pulsed white neutron source (10 meV – 20 MeV)

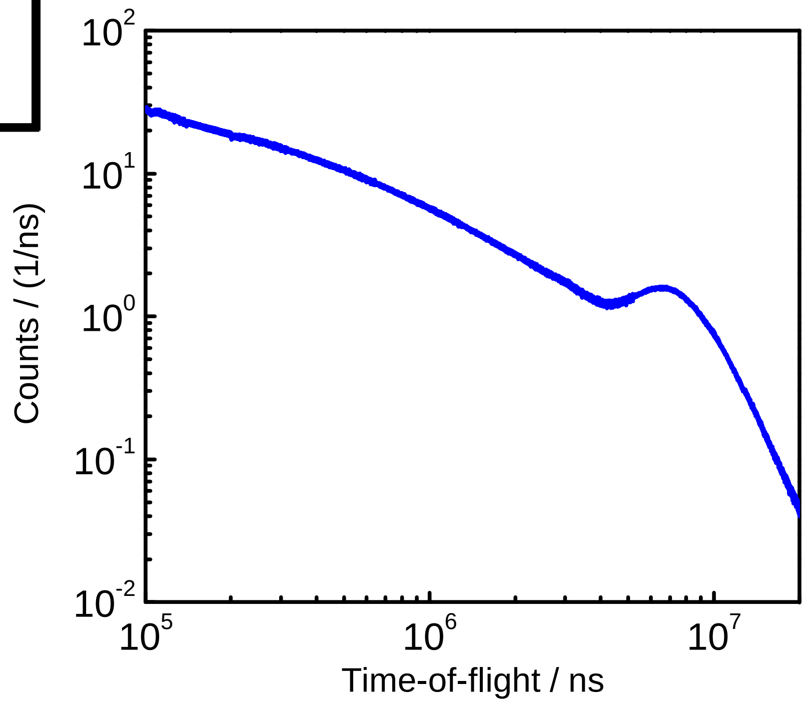


- Neutron energy : time – of – flight (TOF)
- Multi-user facility: 10 flight paths (10 m – 400 m)
- Measurement stations with special equipment:
  - Total cross section measurements
  - Partial cross section measurements

GELINA TOF-Facility: High resolution cross-section measurements

Neutron detector



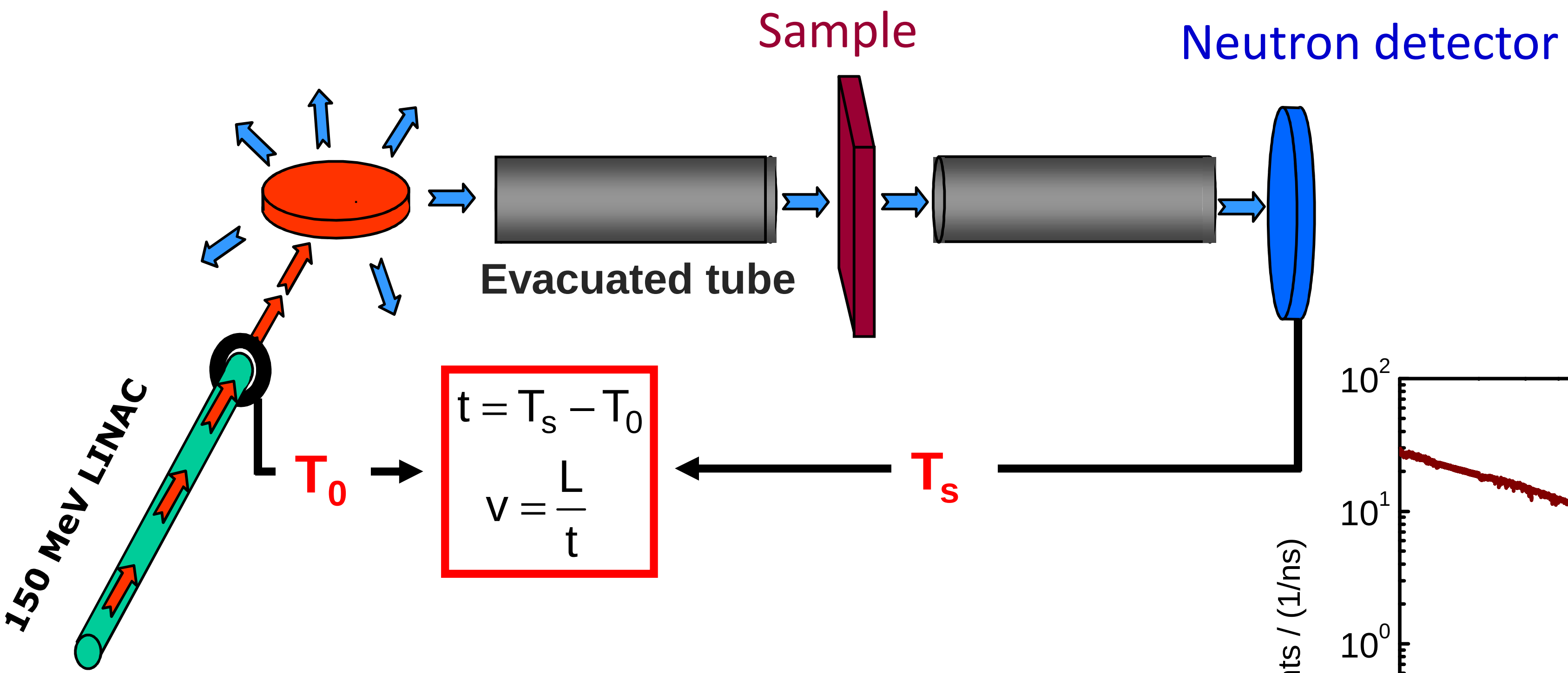


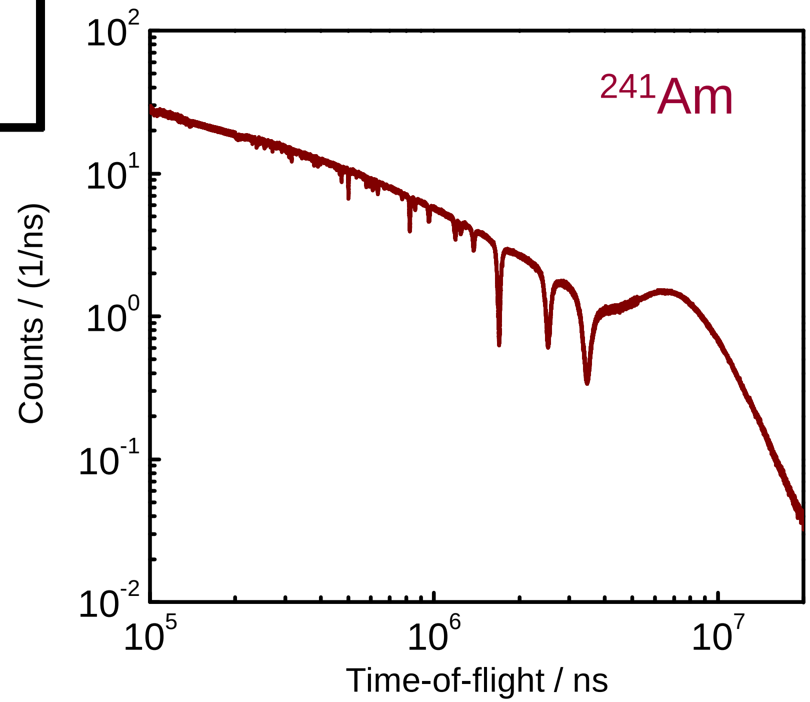
L : distance between target and detector

t : time-of-flight

Schillebeeckx et al. ,Nucl. Data Sheets, 113 3054 (2012)

GELINA TOF-Facility:
High resolution cross-section measurements





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Total cross section
Homogeneous sample

Transmission : fraction of neutron beam traversing without any interaction the sample

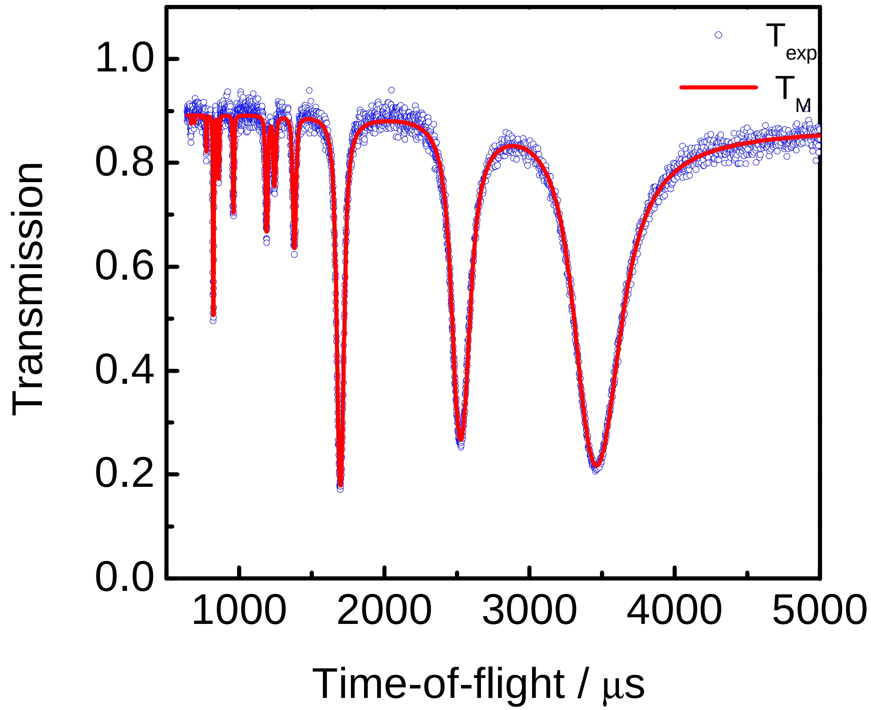
$$T_{exp} = \frac{C_{in}}{C_{out}} \qquad \frac{u_{T_{exp}}}{T_{exp}} < 0.3\%$$

$$\chi^2(RP) = \left(T_{exp} - T_M\right)^T V_{T_{exp}}^{-1} \left(T_{exp} - T_M\right)$$

- Detection efficiency cancels
- Incoming neutron flux cancels

⇒ **absolute measurement**

⇒ **no calibration measurement required**



$$T_M(t) = \int R(t, E)\, e^{-n\, \sigma_{tot}(E)}\, dE$$

R(t,E) : response of TOF-spectrometer

σ_{tot} : total cross section

n : areal number density
total number of atoms per unit area

σ_{tot} : most accurate cross section (uncertainty ≤ 1.0 %)
use of well-characterized sample

Schillebeeckx et al. ,Nucl. Data Sheets, 113 3054 (2012)

NRTA
Homogeneous sample

Transmission : fraction of neutron beam traversing without any interaction the sample

$$T_{exp} = \frac{C_{in}}{C_{out}} \qquad \frac{u_{T_{exp}}}{T_{exp}} < 0.3\%$$

- Detection efficiency cancels
- Incoming neutron flux cancels

$\Rightarrow$ **absolute measurement**
$\Rightarrow$ **no calibration measurement required**

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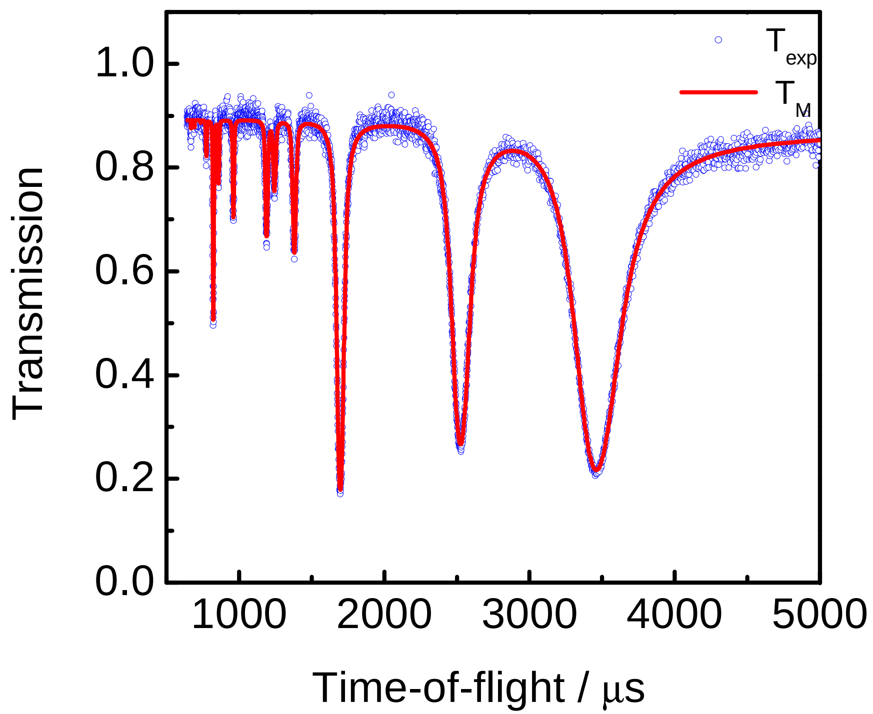


σ_{tot} **: most accurate cross section (uncertainty ≤ 1.0 %)**
NRTA : most accurate NDA technique

Schillebeeckx et al., JRC Science and Policy Report, EUR 26848 EN

Characterization of debris of melted fuel NRTA challenges

Characterization of debris of melted fuel by NRTA

Target value: uncertainty on Pu and U content ≤ 2%

Challenges due to the material characteristics:

- Inhomogeneity of the samples: due to diversity in shape and size of the particle like debris samples (particle-like or rock-like, granularity)
- Impact of impurities: structural material and strong neutron absorbers, e.g. ^{10}B (control rods and borated water)
- Complex transmission spectra due to fission products

Solutions have been studied and validated by measurements at GELINA as part of a JRC/JAEA collaboration

Schillebeeckx et al., ESARDA Bulletin 50, 9 (2013)

Workshop on Neutron Resonance Densitometry

Joint EURATOM / JAEA workshop

4 – 5 March 2015

JRC-IRMM, Geel, Belgium

Objective of the workshop

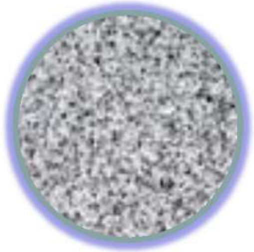
- Report on progress made
- Demonstration of NRTA performance

50 participants

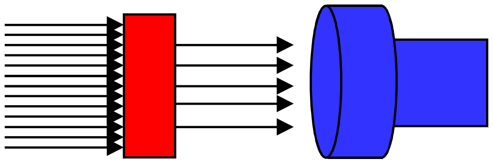
- Extensive delegation from Japan
  (JAEA, Univ. Kyoto & Nagoya)
- DG-ENER, JRC
- EU Member states
- Participants from IAEA, US (DOE, LANL, LLNL, ORNL)

Impact of particle size distribution

Transmission is a non-linear function of n



- Homogeneous sample : $T = e^{-n\,\sigma_{tot}}$
- Heterogeneous sample : $\langle T \rangle = \langle e^{-n\,\sigma_{tot}} \rangle \neq e^{-\langle n \rangle\,\sigma_{tot}}$



<n> is the quantity of interest

⇒ Dedicated model for debris samples is required to avoid bias effects

Validation of different models by stochastic calculations (MC simulations)

⇒ LP – model (Levermore, Pomraning et al., J. Math. Phys. 27, 2526, (1986))

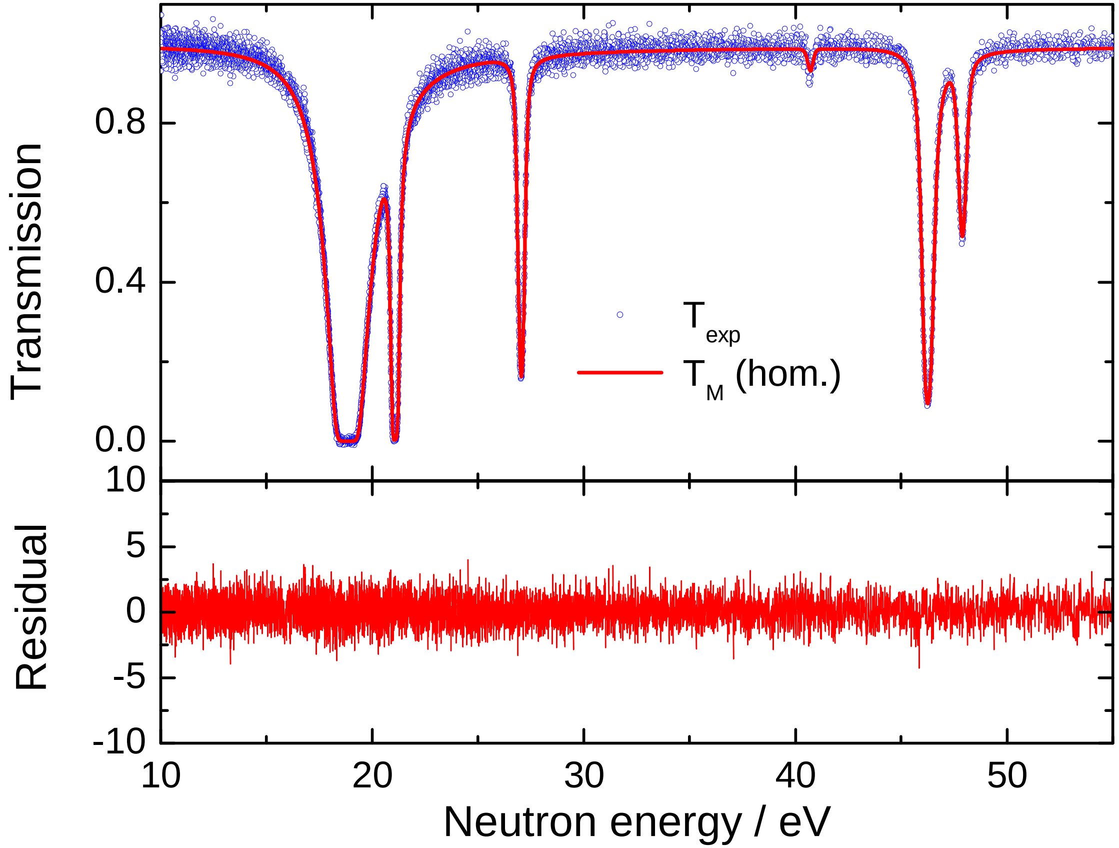
- Widely used for other problems dealing with radiation transport through stochastic media, e.g. scattering of sunlight in clouds
- Starts from microscopic properties of the sample such as grain size
- Applicable for powder samples
- Validated by experiments at GELINA

Becker et al., EPJ Plus 129, 58 (2014)

Experimental validation of LP-model at GELINA

Declared : $n_W = 9.38\ 10^{-4}$ at/barn

T_M (hom.) : $n_W = 9.36\ 10^{-4}$ at/barn

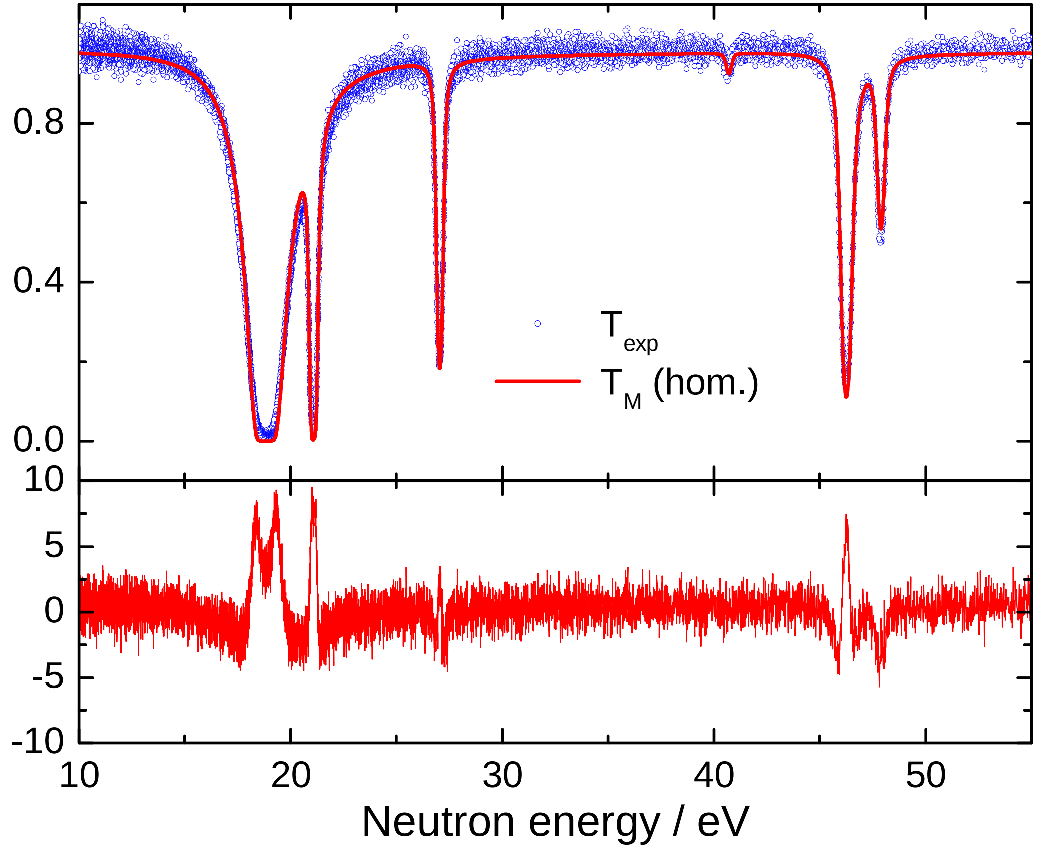


natW-metal disc

⇒ bias < 1%

Declared : $n_W = 1.03\ 10^{-5}$ at/barn

T_M (hom.) : $n_W = 0.85\ 10^{-5}$ at/barn



natW-powder mixed with natS-powder

Homogeneous model ⇒ bias > 15%

Becker et al., EPJ Plus 129, 58 (2014)

Experimental validation of LP-model at GELINA

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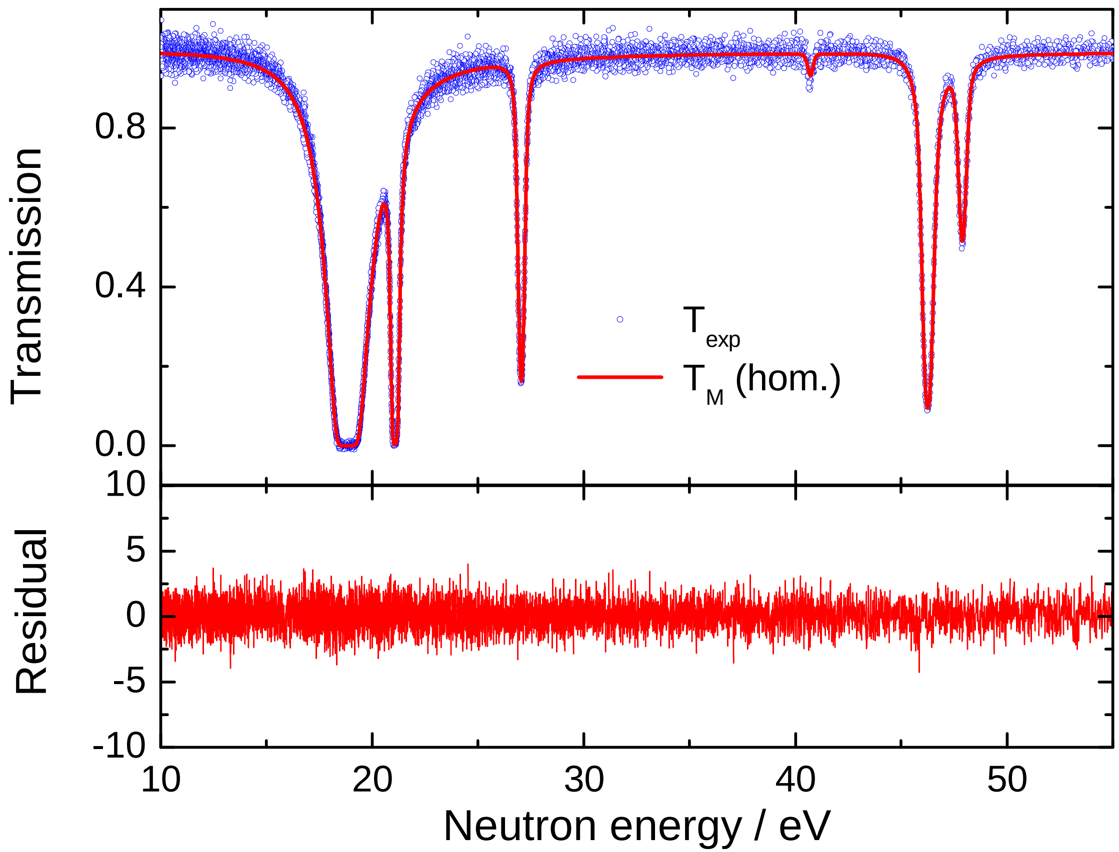
T_M (hom.) : $n_W = 9.36\ 10^{-4}$ at/barn

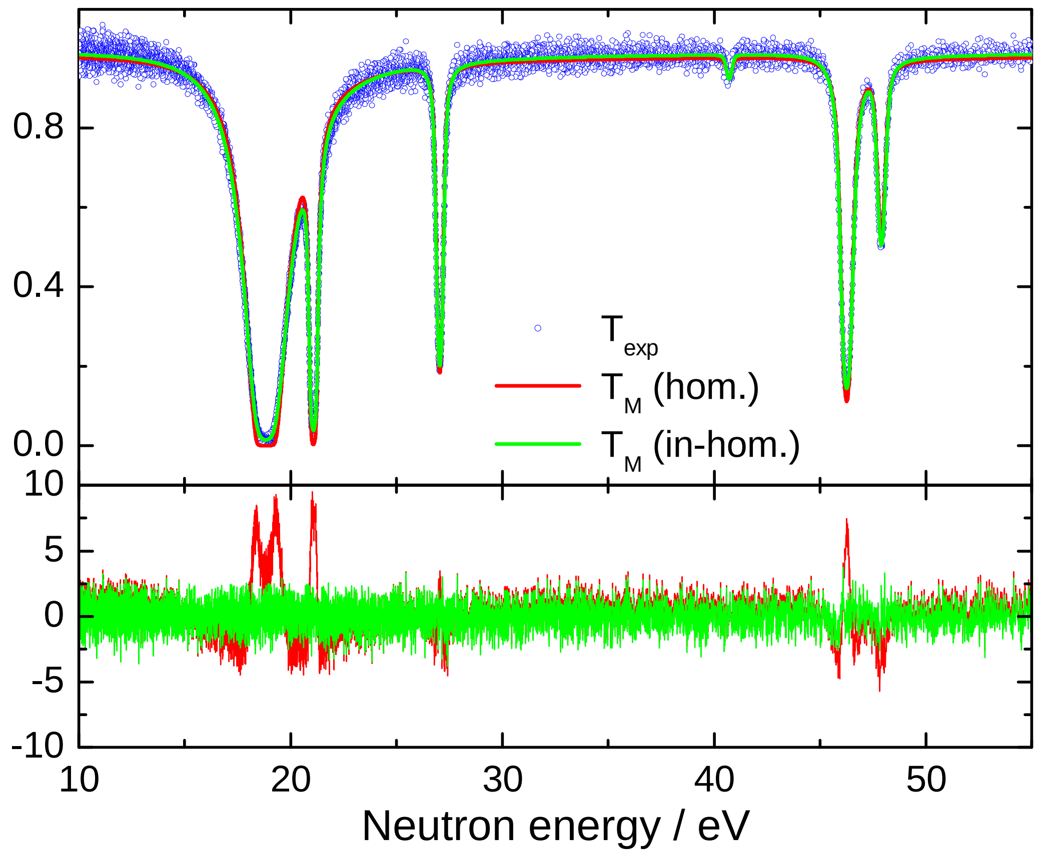


^{nat}W-metal disc

$\Rightarrow$ bias < 1%

Declared : $n_W = 1.03\ 10^{-5}$ at/barn

T_M (inhom.) : $n_W = 1.05\ 10^{-5}$ at/barn

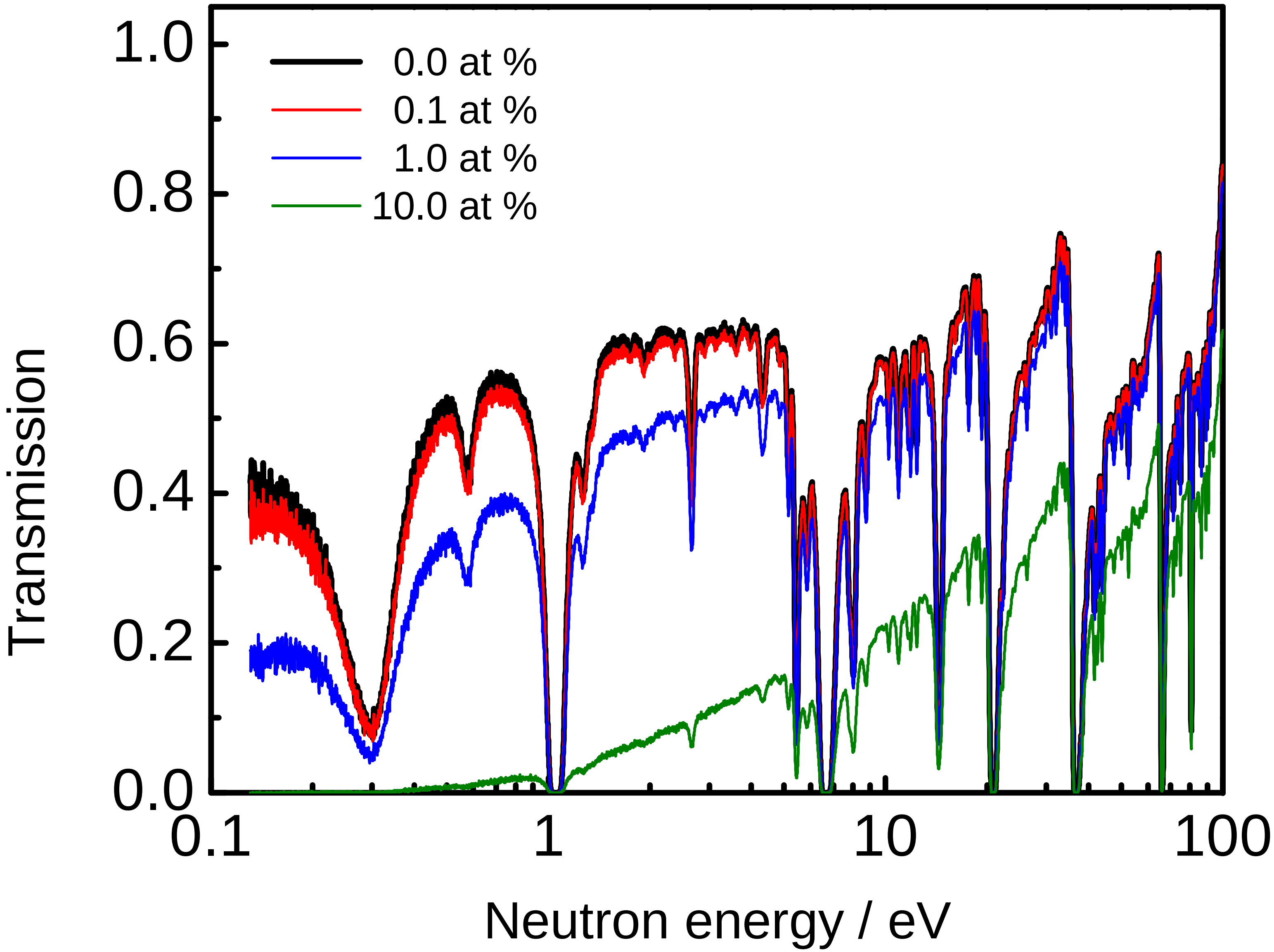


^{nat}W-powder mixed with ^{nat}S-powder

LP - model $\Rightarrow$ bias $\leq$ 2 %

Becker et al., EPJ Plus 129, 58 (2014)

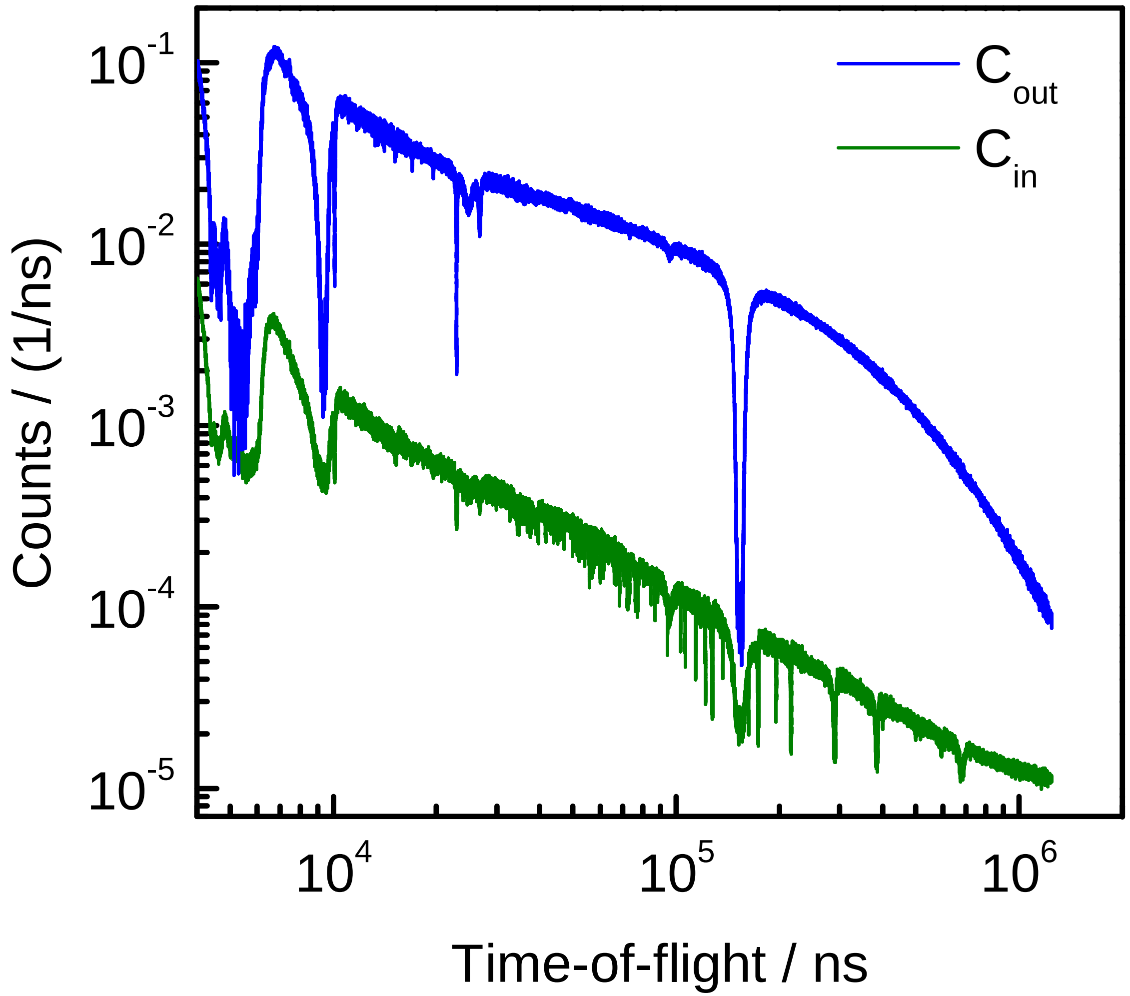
Impact of impurities e.g. ^{10}B



Experimental validation of final model at GELINA

U_3O_8 reference sample
EC NRM 171

Strong impact
of matrix material



Schillebeeckx et al., JRC Science and Policy Report, EUR 26848 EN

Experimental validation of final model at GELINA

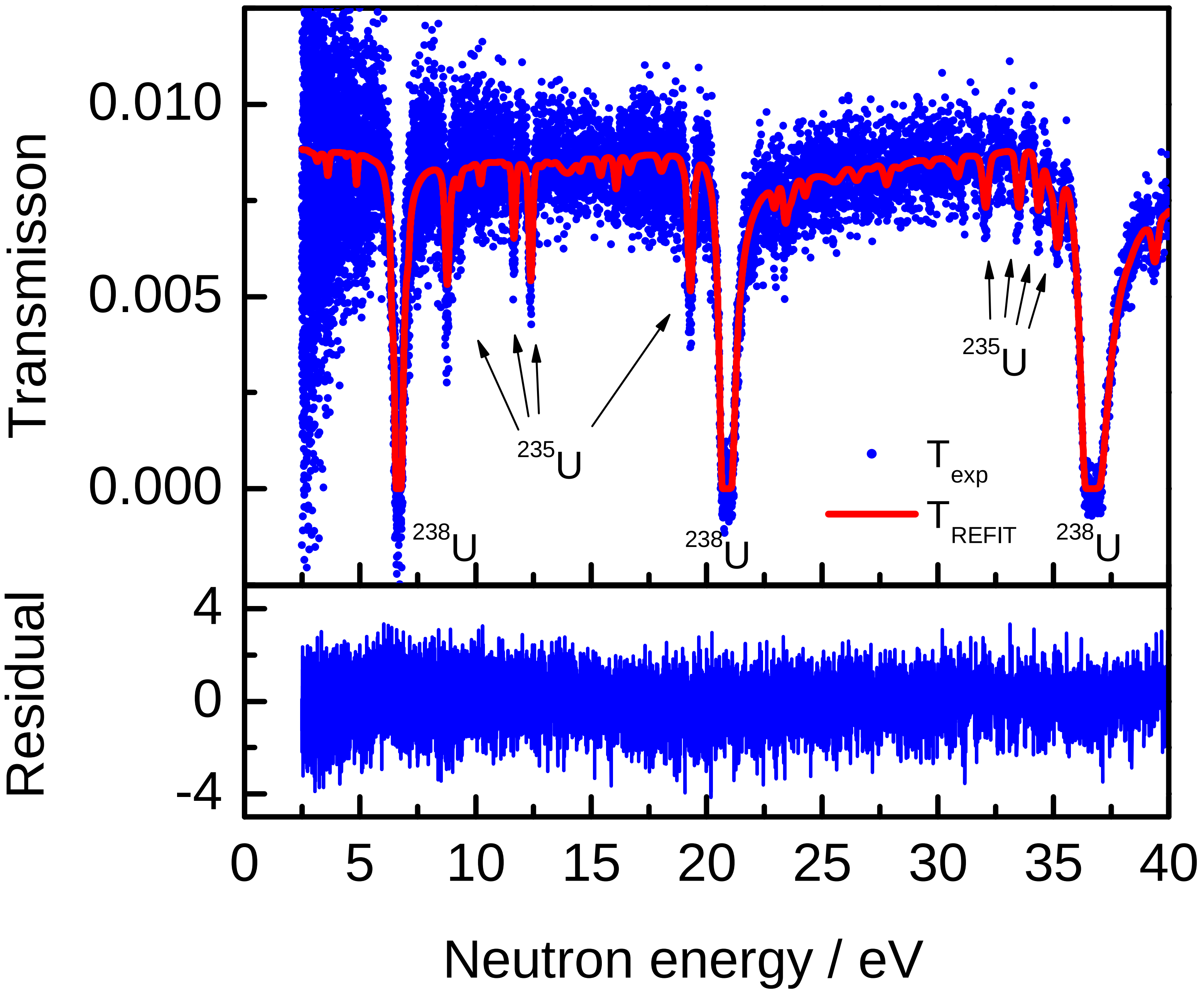
U_3O_8 reference sample
EC NRM 171

Beam attenuation
due to matrix
~ 99%

Fit for areal density

+

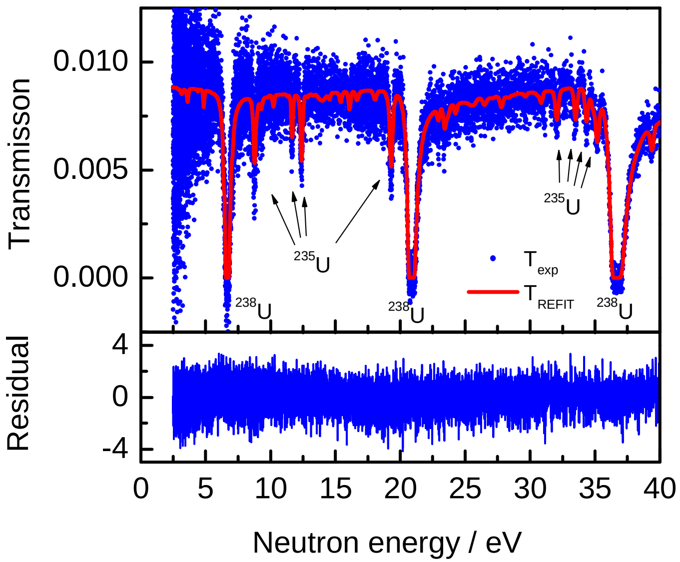
$$n_X\ \sigma_{tot,X}(E) = a_X + \frac{b_X}{\sqrt{E}}$$



Experimental validation of final model at GELINA

U_3O_8 reference sample
EC NRM 171

U-isotope	Areal number density (at/b)		Ratio
	Declaration	NRTA	
^{235}U	(5.0326 ± 0.0080) x 10^{-4}	(5.063 ± 0.09) x 10^{-4}	**1.006**
^{238}U	(1.0628 ± 0.0015) x 10^{-2}	(1.062 ± 0.01) x 10^{-2}	**0.999**



$\Rightarrow$ bias < 1.0 %

Schillebeeckx et al., JRC Science and Policy Report, EUR 26848 EN

Demonstration experiment at GELINA selection of samples

Samples
- 18 different samples
- 8 different elements

Black box: 8 slots

➔ **B, Mn, Co, Cu, Nb, Rh, W, Au samples with different thicknesses**

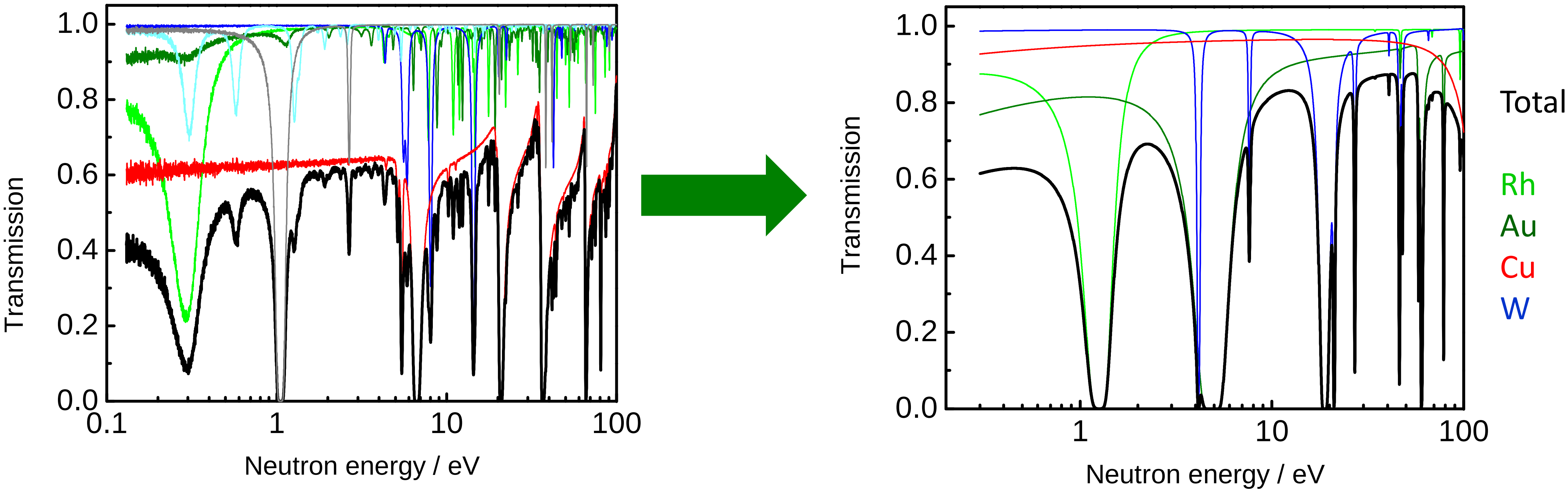
Selection of samples by DG-ENER, IAEA and DOE representatives

Demonstration experiment at GELINA complex transmission

Experiments with radioactive material containing U and Pu are not possible

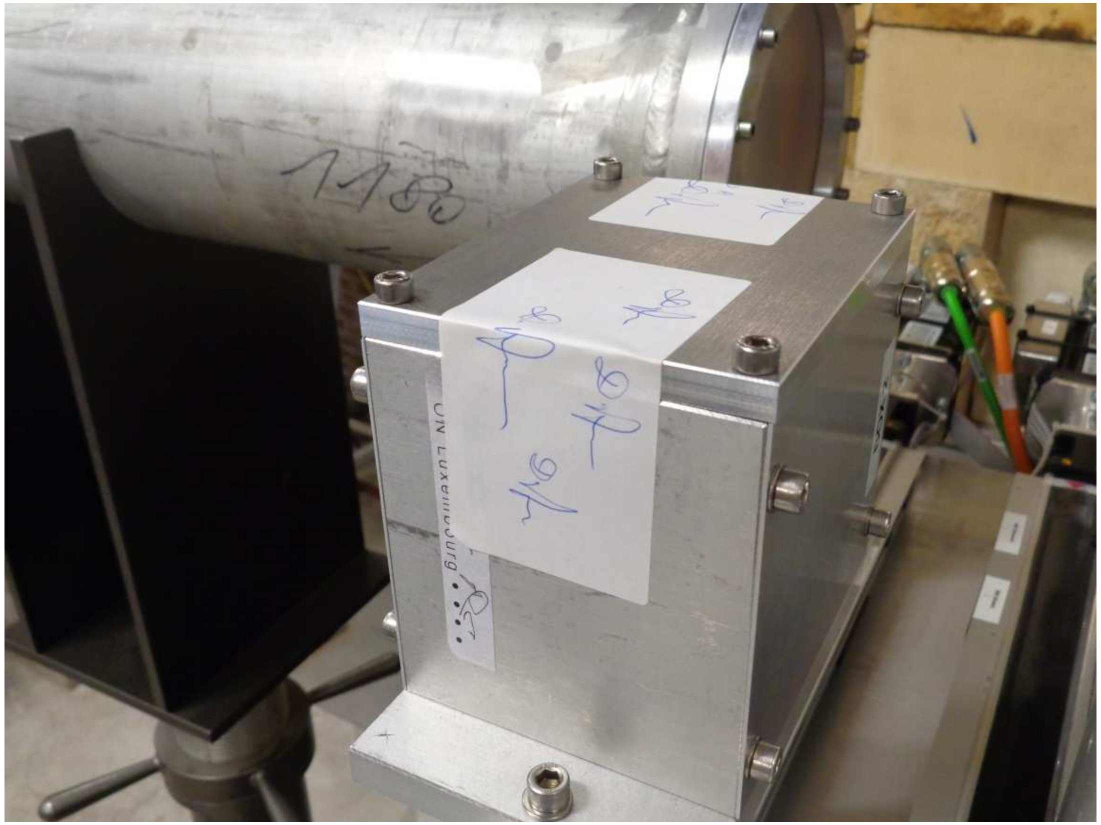
→ produce similar complex spectrum using

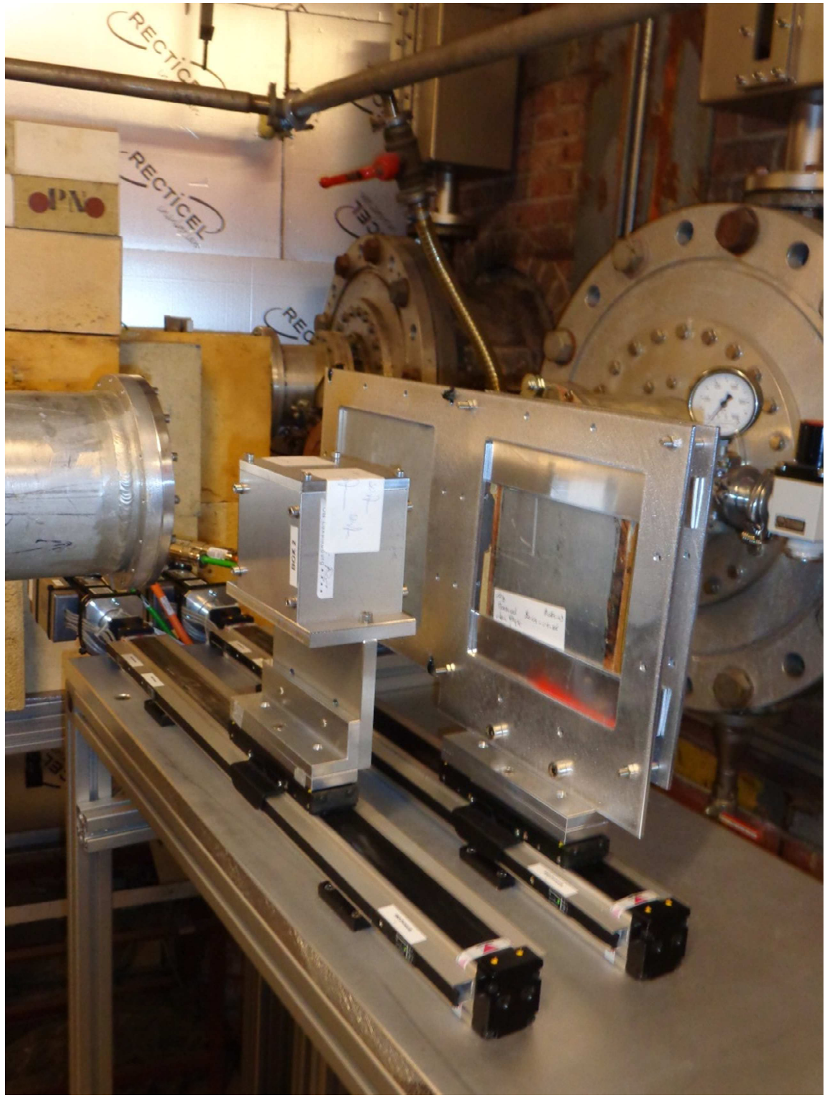
- elements with resonances in the low energy region
- elements with reliable resonance parameters
- samples that are not radioactive and do not contain nuclear material



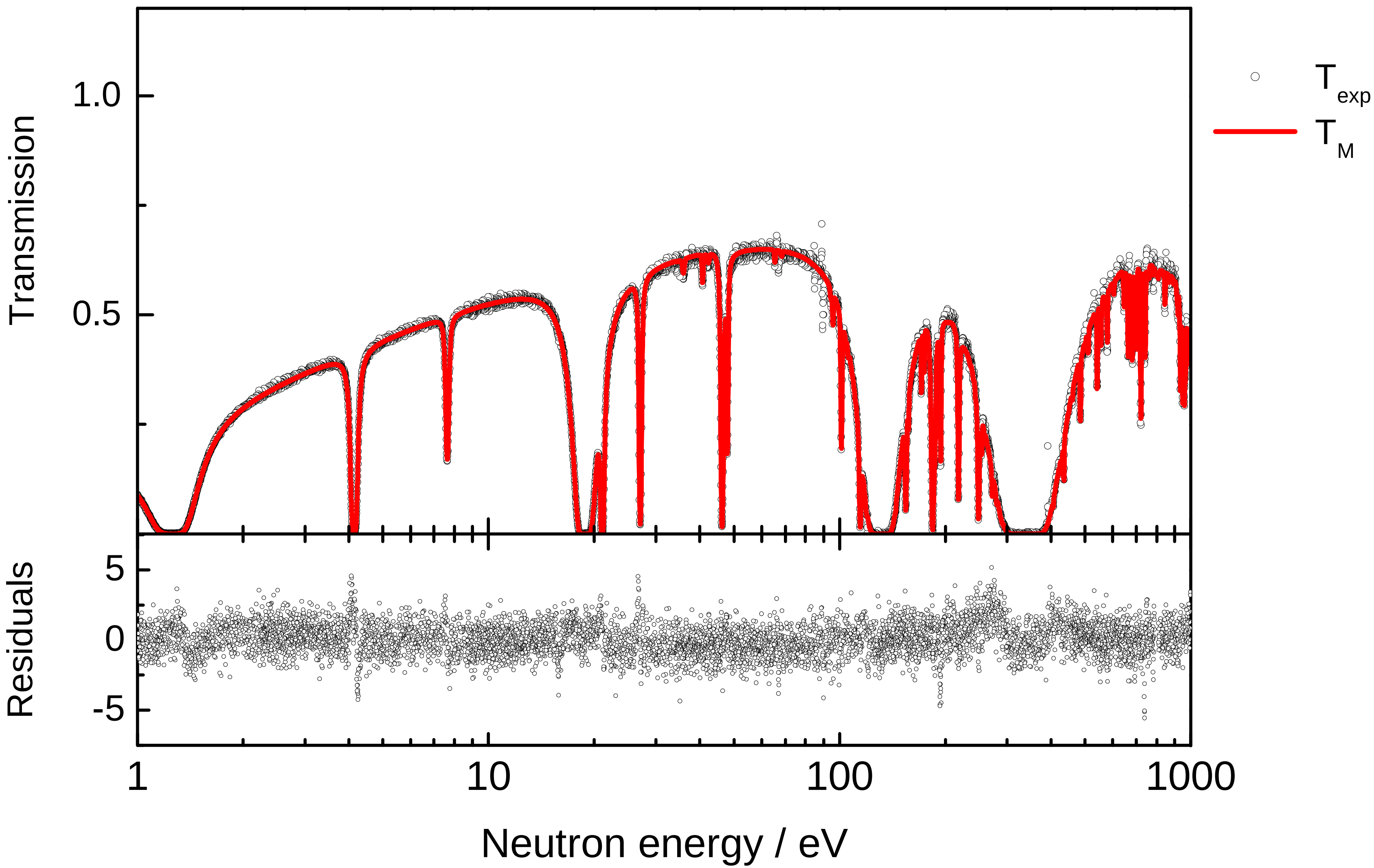
Demonstration experiment at GELINA start of the experiment

NRTA station at 10 m





Demonstration experiment at GELINA
Results



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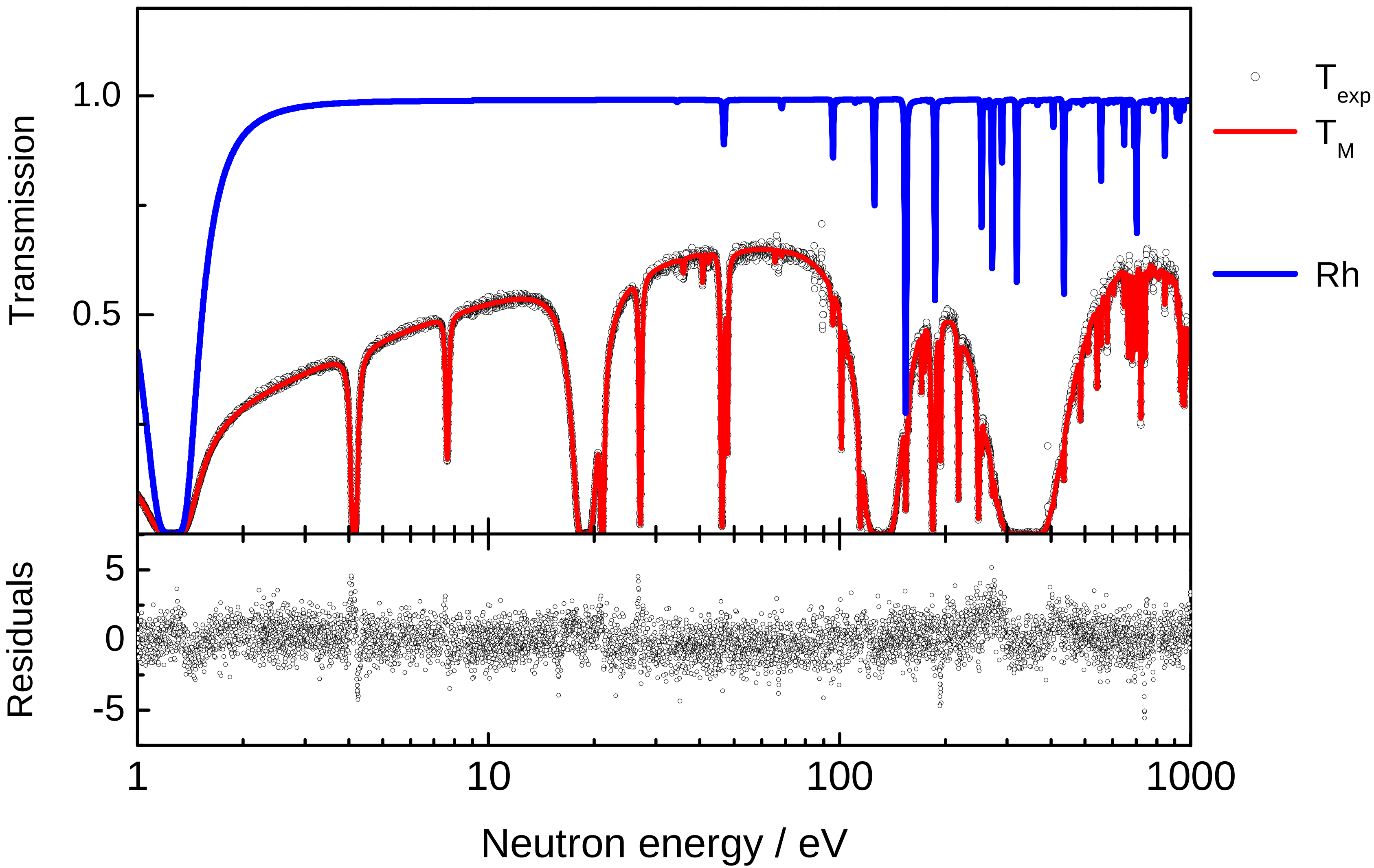

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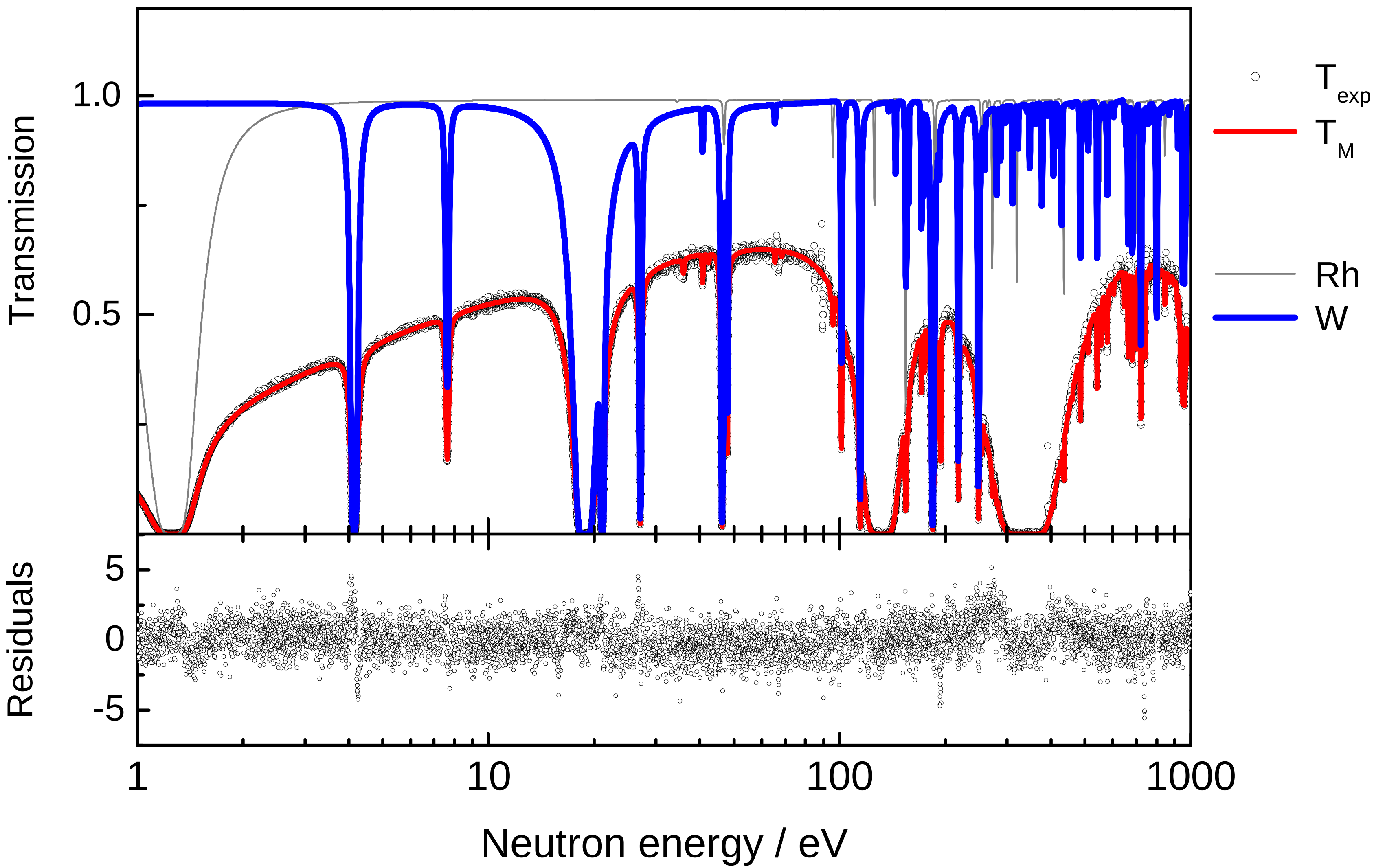

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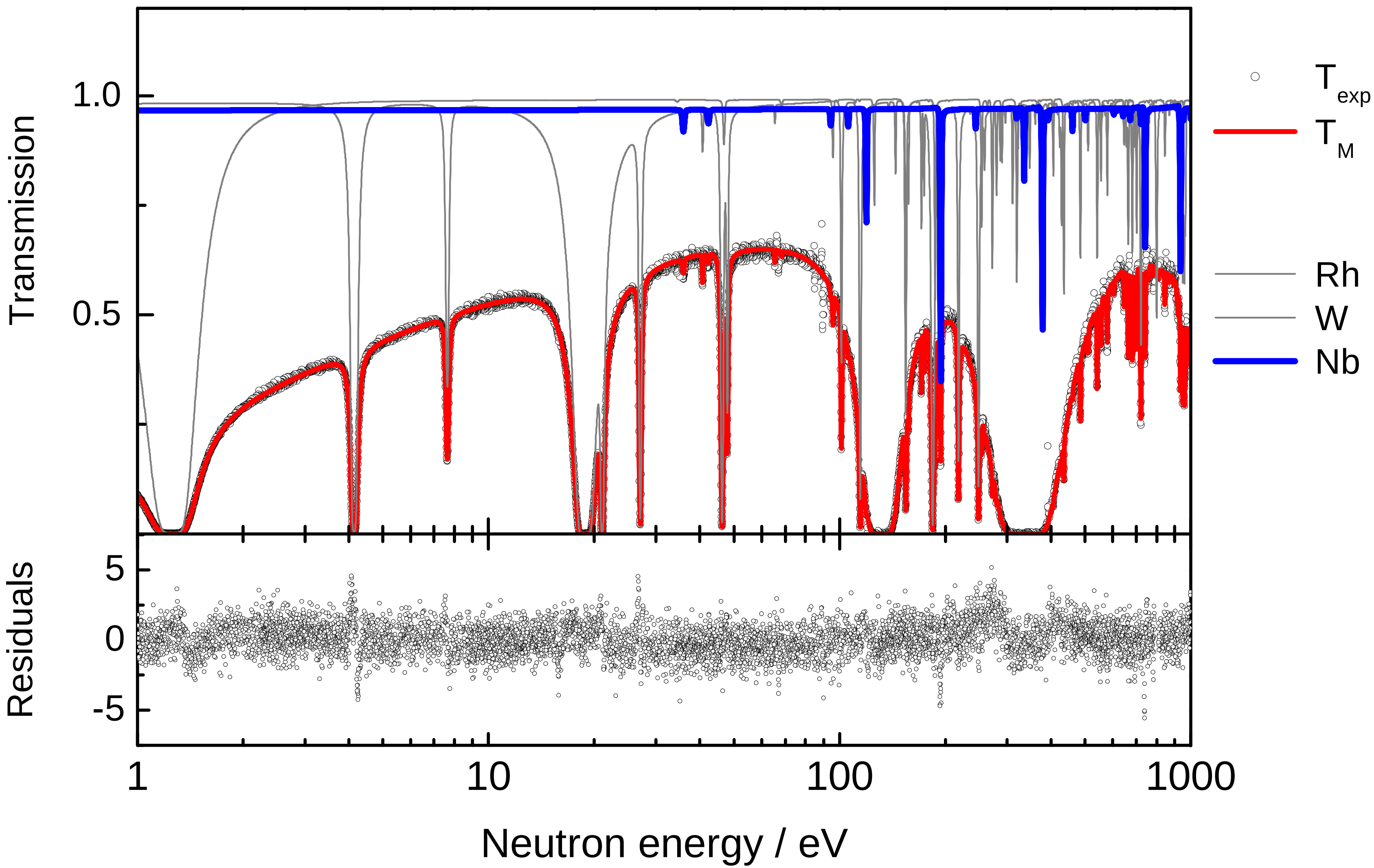

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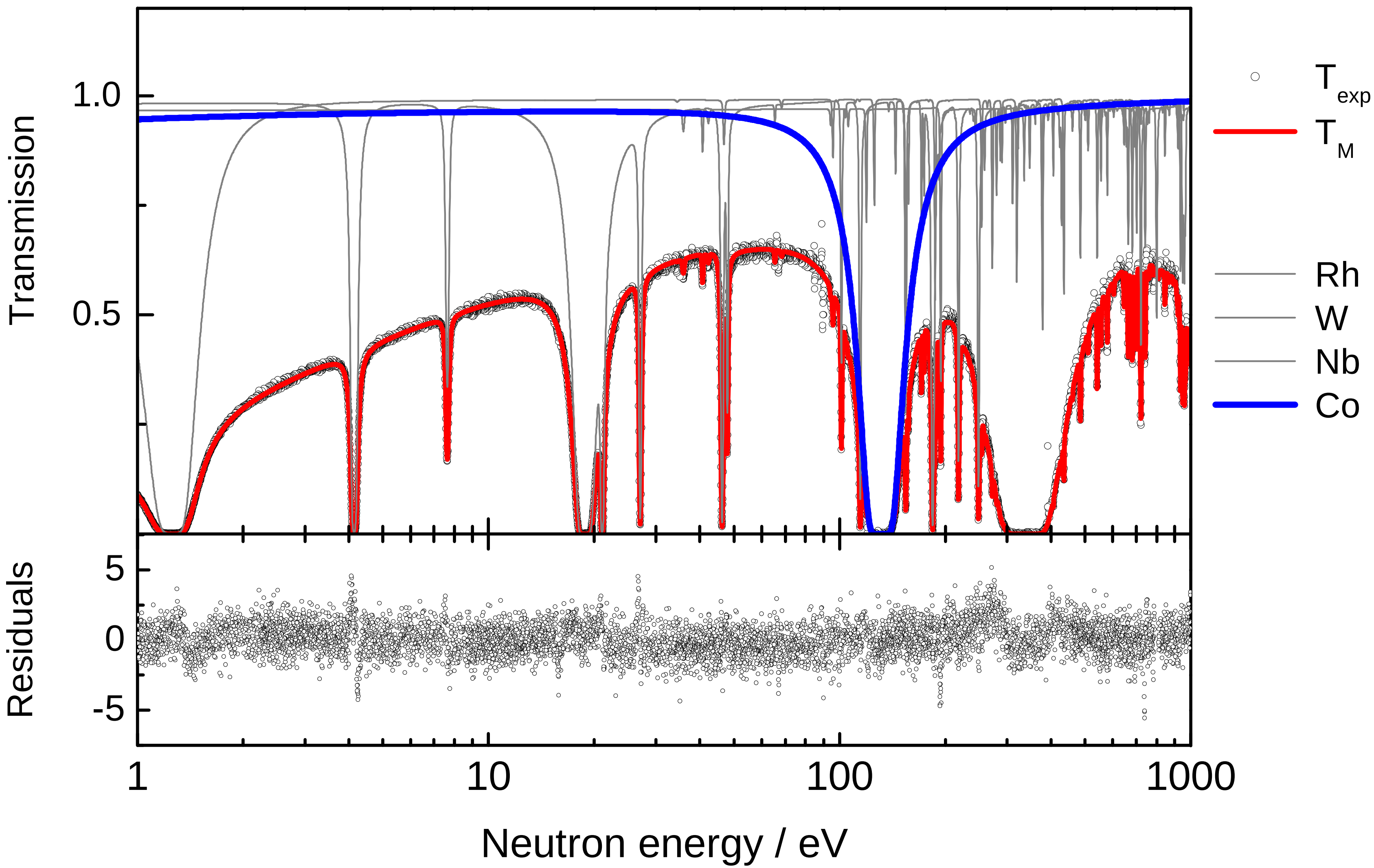

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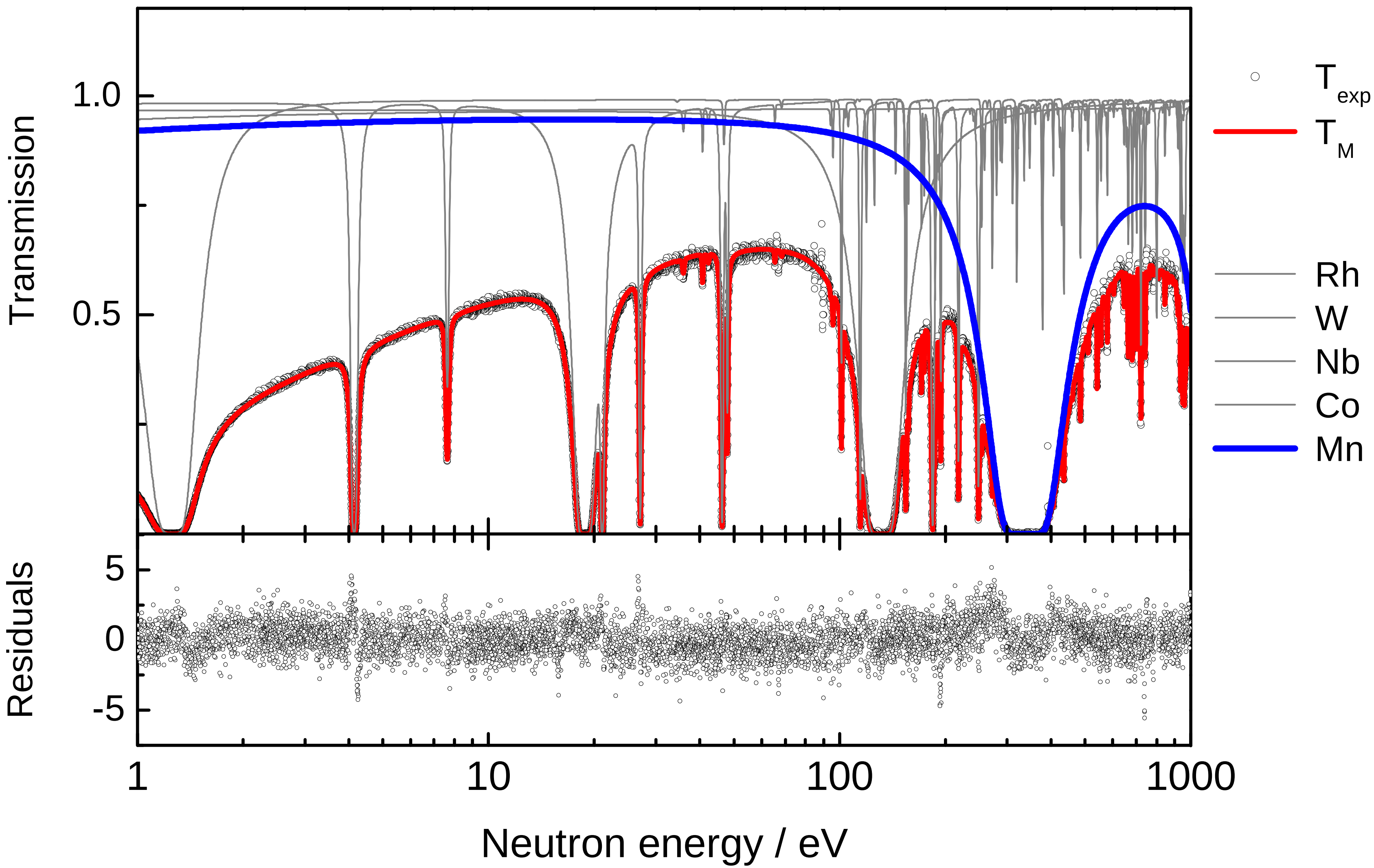
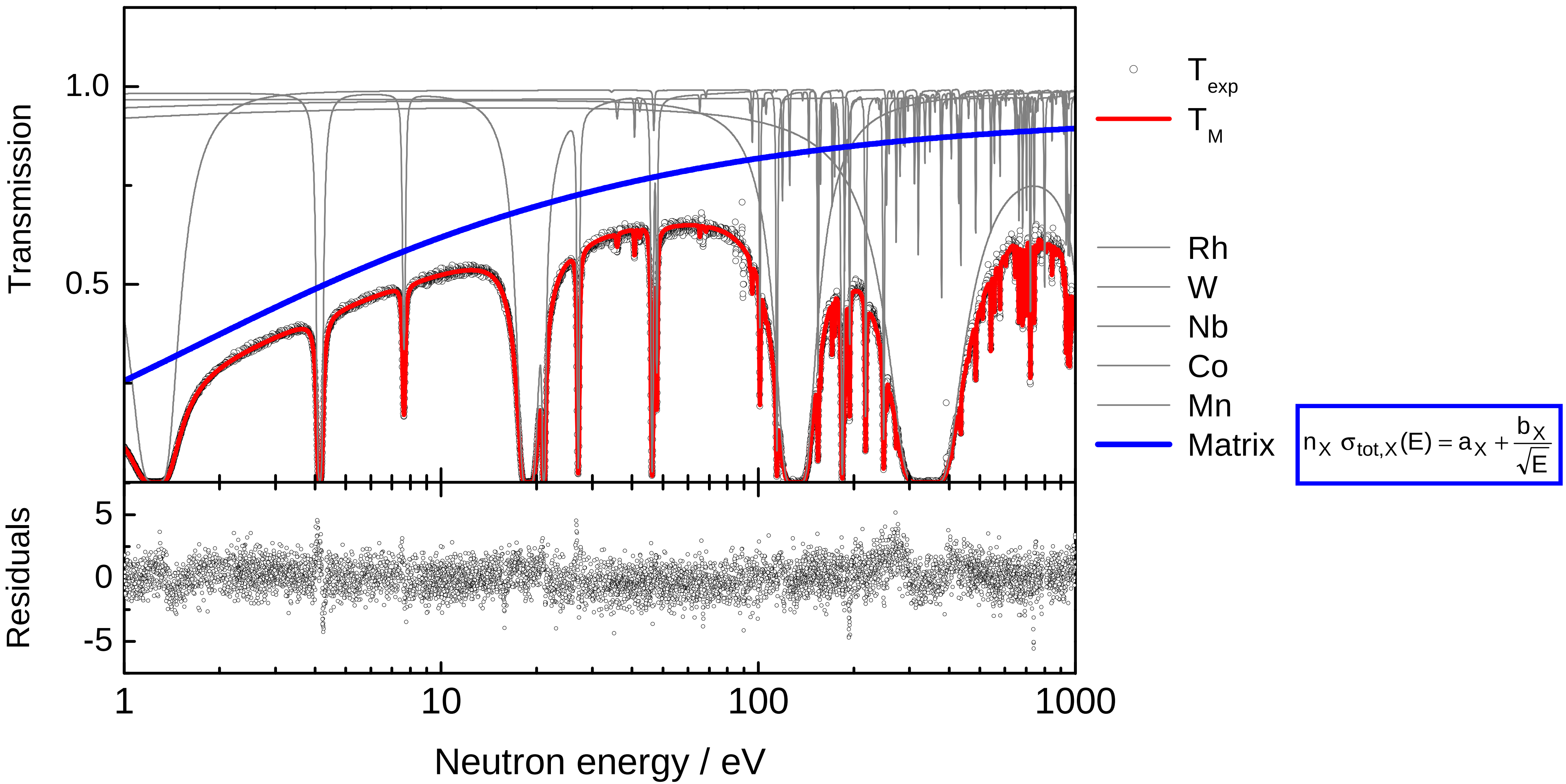
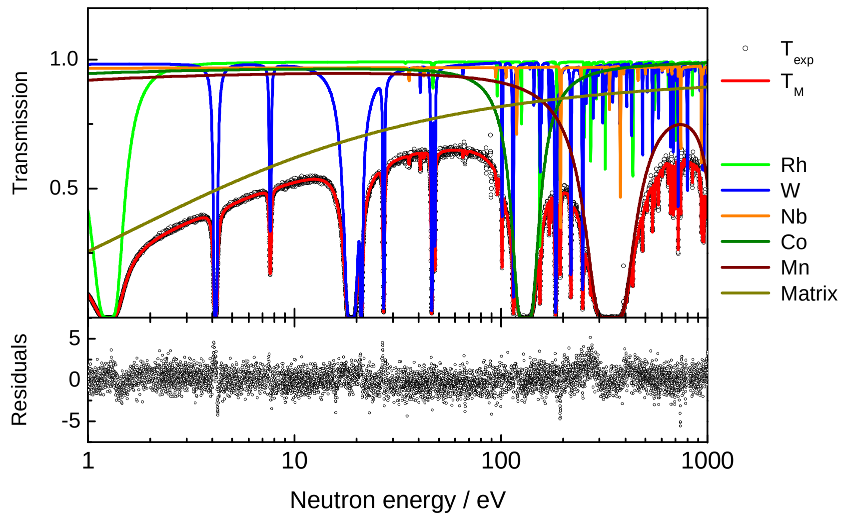


Demonstration experiment at GELINA Results



Demonstration experiment at GELINA comparison with reference data

Element	Areal number density (at/barn)		n_{Ref}/n_{NRTA}
	n_{Ref}	n_{NRTA}	
Mn	1.901 x 10^{-2}	(1.928 ± 0.003) x 10^{-2}	1.014 ± 0.002
Co	4.583 x 10^{-3}	(4.509 ± 0.015) x 10^{-3}	0.984 ± 0.003
Cu	0	0	
Nb	5.485 x 10^{-3}	(5.382 ± 0.010) x 10^{-3}	0.981 ± 0.002
Rh	1.856 x 10^{-3}	(1.891 ± 0.003) x 10^{-3}	1.019 ± 0.002
W	2.269 x 10^{-3}	(2.250 ± 0.002) x 10^{-3}	0.992 ± 0.001
Au	0	0	



Summary

Neutron Resonance Transmission Analysis (NRTA)

- Non-destructive analysis method (NDA)
- Based on well-established methods for cross section measurements
- Applicable for high radioactive nuclear material
- No sample preparation required
- Sensitive to almost all nuclides (except light nuclides)
- Absolute method, no calibration requirements
- Models to apply the method on particle size debris, including strong neutron absorbers, have been validated at GELINA
- Accurate method (bias effects < 2%, depends only on nuclear data)

Acknowledgements

The JRC / JAEA NRD collaboration

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G. Alaerts, D. Vendelbo and R. Wynants

Research and training options at Neutron Facilities of JRC Geel

EUFRAT – European facility for nuclear reaction and decay data measurements

Transnational Access of external users to JRC-IRMM nuclear facilities

https://ec.europa.eu/jrc/en/eufrat

GENTLE – Graduate and Executive Nuclear Training and Lifelong Education

Student Research Experience

http://gentleproject.eu/

Thank you for your attention