Agenzia nazionale per le nuove tecnologie, l'energia e lo sviluppo economico sostenibile

Istituto Nazionale di Fisica Nucleare

Sezione di Bologna

Misura delle sezioni d'urto di cattura neutronica $^{155,157}Gd(n,\gamma)$ fra 10^{-2} e 10^{6} eV alla facility sperimentale n_TOF

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on benhalf of the n_TOF Collaboration

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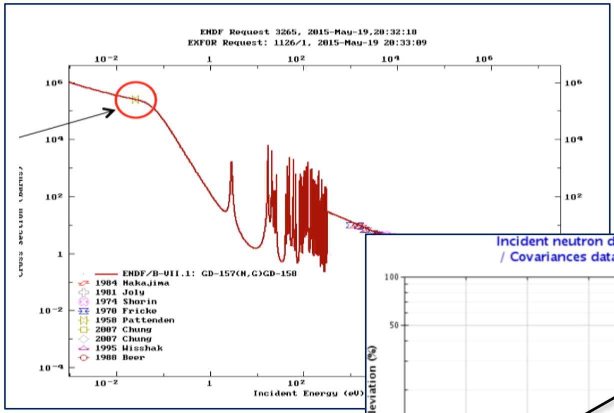
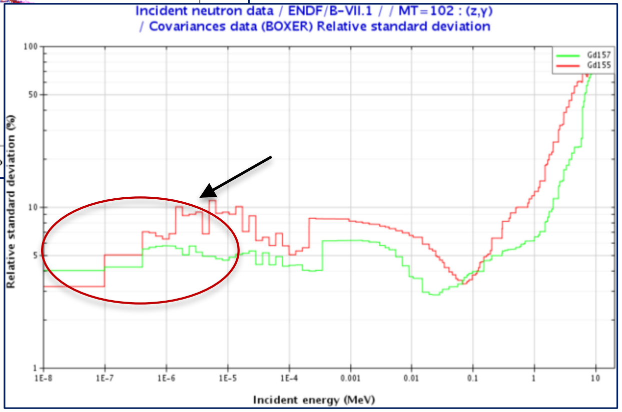
Scientific Motivation
(*Inconsistence of actual Gd data)*

- The **gadolinium odd isotopes σ(n,γ) thermal data present** sensitive (-16 ÷ 4%) deviation once compared to the ENDF/B-VII.1 nominal values **[1]**.
- **ENDF/B-VII.1 data uncertainty (i.e., 0.3%) cannot be considered a safe estimation** of the actual range values of the thermal Xs **[1]**.

Reference	Year	Thermal Xs (kb)	Deviation from ENDF
Pattenden	1958	264	+3.9%
Tattersall	1960	213	-16%
Moller	1960	254	=
Groshev et al.	1962	240	-5.5%
Sun et. al	2003	232	-8.7%
Leinweber	2006	226	-11%
Mughabghab (ENDF/B-VII.1)	2006	254 ± 0.3%	=
Choi et al.	2014	239	-5.9%





- **EXFOR database** presents only one experimental thermal point of ^{157}Gd(n,γ) published in 1958 with no uncertainty associated to it.
- ^{155,157}**Gd** present relatively high uncertainty values (5÷10%) in thermal range.

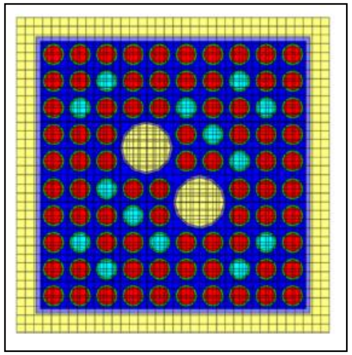
[1] F. Rocchi, *et al.*, EPJ Nuclear Science Technology 3, **21** (2017).

Scientific Motivation
(*ICSBEP comparison and S&U analysis*)

ICSBEP Conf.		Kref	ENDF/B-VII	JEFF-3.1	Leinweber et al.[2]	Improv.
HST-014	C2	1.0000	1.00996	1.01304	1.01903	No
	C3	1.0000	1.01827	1.01852	1.02636	No
LCT-035	C3	1.0000	0.99591	0.99556	0.99935	Yes
LCT-005	C2	1.0000	1.00029	1.00006	1.00466	No
	C3	1.0000	0.99907	1.00002	1.01651	No
	C4	1.0000	0.99721	0.99846	1.01602	No
	C6	1.0000	1.00684	1.00697	1.00962	No
	C7	1.0000	1.00191	1.00258	1.00846	No
	C8	1.0000	1.00163	1.00295	1.01213	No
	C9	1.0000	1.00257	1.00379	1.01459	No

- Analysis on a series of **International Criticality Safety Benchmark** confirmed the worsening of the latest Gd Xs data **[2]**.
- **The 74 LCT facilities** yield average values of *C/E-1* between *-578 pcm* (JEFF-3.1.1) and *-499 pcm* (JENDL-4.0).
- **The V. D. Marck works** concludes that part of the *C/E-1* differences of overall 2000 ICSBEP is due to Gd isotopes **[3]**.

- A **S&U analysis** showed that **^{155,157}Gd(n,γ)** give the largest contribution to Keff uncertainty after **^{235,238}U**.



Parameter	Data
FA; enr.	GE10x10-8; 4.12 wt%; HFP
Nr. of Gd pins	14 (enr. 5.0 wt%)
Boron content	0 ppm
Water density	0.45 g/cc
Xs MG library	**V7-238** (ENDF/B-VII.0)
Xs COV library	**44group7.0** (ENDF/B-VII.0)

Covariance matrix		Contribution to uncertainty	Rank (-)
Nuclide-reaction	Nuclide-reaction	Due to this marix (%Δk/k)	-
^{235}Unubar	^{235}Unubar	2.62E-01	1.00
^{238}U(n,γ)	^{238}U(n,γ)	2.11E-01	0.80
^{238}U(n,n')	^{238}U(n,n')	1.66E-01	0.63
^{235}U(n,γ)	^{235}U(n,γ)	1.47E-01	0.56
^{235}U(n,f)	^{235}U(n,f)	1.41E-01	0.54
^{235}Uchi	^{235}Uchi	1.33E-01	0.51
^{235}U(n,f)	^{235}U(n,γ)	1.18E-01	0.45
^{238}Unubar	^{238}Unubar	8.35E-02	0.32
^{157}Gd(n,γ)	^{157}Gd(n,γ)	6.72E-02	0.26
^{155}Gd(n,γ)	^{155}Gd(n,γ)	5.15E-02	0.20

[2] G. Leinweber *et al.*, Nucl. Sci. Eng. **154**, 03 (2006); **[3]** S. C. van der Marck, Nucl. Data Sheet **113**, (2012).

Data Analysis
(*Capture yield and Counting rate*)

- The Gd Xs were evaluated using the TOF technique through the measurement of the reaction yield:

$$Y(E_n)_x = N\frac{C_w(E_n) - B_w(E_n)}{\varepsilon_x \cdot \varphi_n(E_n)}$$

- N: normalized factor.
- C_w: weighted C_6D_6 counting rate.
- B_w: weighted background counting rate.
- ε_x: detection efficiency $f(\Omega, A, f_{BIF}, \Phi)$
- φ_n: neutron fluence.

C_w: Weighted C_6D_6 counting rate

- It was evaluated assuming the total-energy detection principle by means of Pulse Height Weight Technique (PHWT) that assure the proportionality between detection efficiency (DE) and y-ray energy:

$$\int_0^\infty R_d(E_d, E_\gamma) WF(E_d) dE_d = kE_\gamma$$

$$WF(E_d) = \sum_i (a_i \cdot E_{d,i})$$

- **WF definition:** is a function defined to assure the proportionality between detection response (DR) and γ-ray energy.
- **WF mathematical form:** expressed with a smooth functions of the deposited energy and a set of free parameters.

$$X^2 = \sum_i \left(kE_{\gamma j} - \int_{E_l}^\infty R_d(E_d, E_{\gamma j}) WF(E_d) dE_d\right)^2$$

$$C_w(T_n) = \int_0^\infty C(T_n, E_d) WF(E_d)\, dE_d$$

- **WF Parameters**: were obtained by a least square fit of a number of y-ray response and minimizing the ^2X.
- **Cw***:* using the evaluated WF, a weighted TOF spectrum proportional to the y-ray energy was obtained.

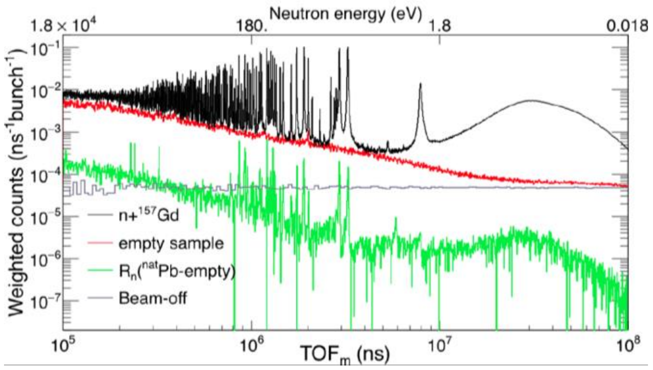
[6] J. Allison et al., Nucl. Instrum. Methods A **835**, 186 (2016)

Data Analysis
(*Background, Normalisation factor and BIF*)

B_w: Weighted background counting rate

- Dedicate measurements were performed in order to evaluate the various B_w component:

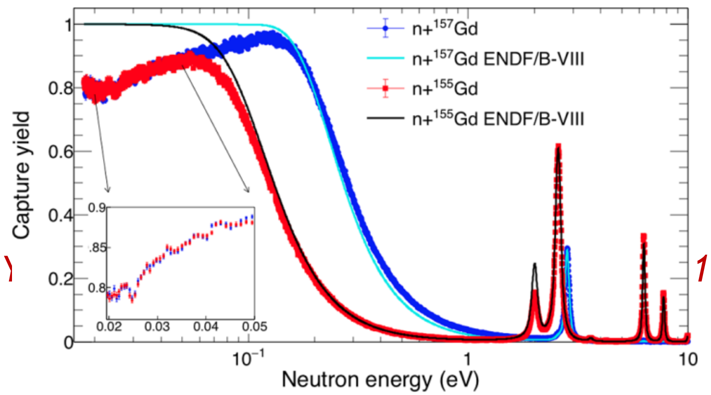
1. **Neutron beam interaction out the sample** → $[\boldsymbol{Empty\ Sample}]$
2. **Sample-scattered neutrons**→ $[({}^{nat}\boldsymbol{Pb} - \boldsymbol{Empty}) \cdot \boldsymbol{R_n};\ \boldsymbol{R_n} = \boldsymbol{Y^{sc}_{Gd}/Y^{sc}_{Pb}}]$
3. **γ-ray traveling in the beam** → $[\boldsymbol{Lead\ Sample}]$
4. **Time-independent background** → $[\boldsymbol{Beam - off}]$



N: Normalized factor

- Groups together the correction factors independent of neutron energy.
- Obtained with saturated resonance technique applied to the 4.9 eV resonance in $(n + {}^{197}Au)$.
- *N* was extracted with an uncertainty due to counting statistic of less than 0.1%.

BIF: Beam Interception factor



- The theorical capture yield for thick 155,157Gd sample is evaluated to be 1 below 0.07 and 0.1 eV $(n\sigma_{tot} \gg 1)$.
- Any departure of the measured capture yield from unit can be ascribed to a variation in the BIF of the sample.
- An experimental BIF was evaluated to correct the capture yields of the thin gadolinium and gold samples.

Data Analysis
(*Resonance Shape Analysis –*^{155}Gd)

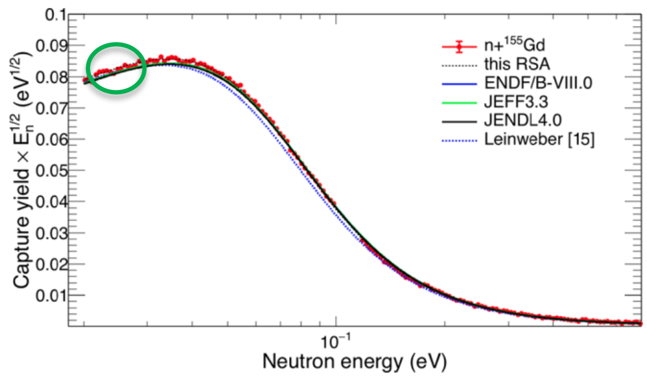
- **Code:** The RSA was performed with SAMMY code, using the Reich-Moore approximation. ENDF/B-VIII.0 resonance parameters (RP) and scattering radius (SR) were adopted as initial values of a fitting procedure.
- **Features:** The code includes corrections for experimental conditions such as: *Doppler broadening*, *self-shielding and multiple scattering*, *thermal motion of atoms* (free-gas model).

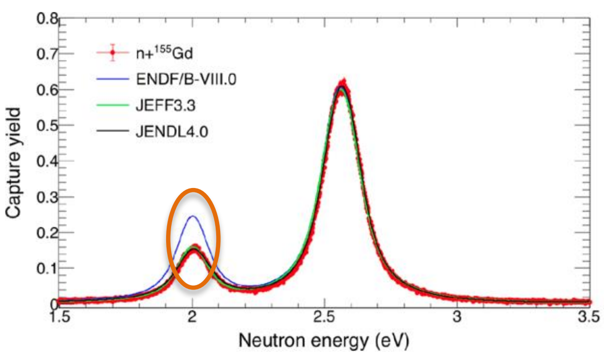
^{155}Gd

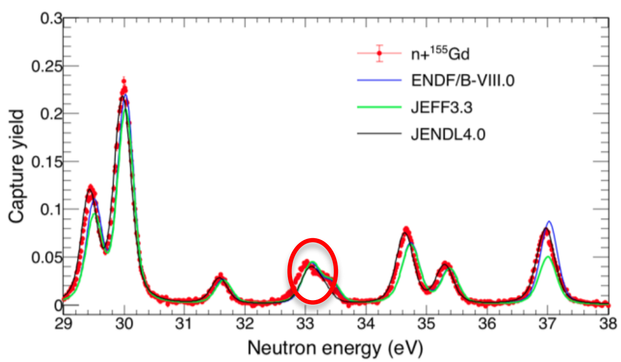
✓ *Thermal point:* is almost constant among libraries (60.735 ÷ 60.890 kb). A slightly higher Xs **62.2 ± 2.2 kb** was deduced. Wescott factor is 0.86.

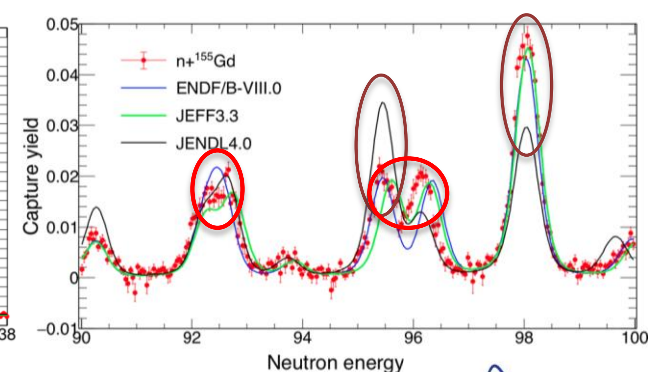
✓ *Resonance*: kapture kernel of the **2.0 eV resonance** is about 50% lower than the ENDF/B-VIII.0 value. Large deviations of **95.7 and 98.3 eV resonances** were also founded with respect to JENDL-4.0.

✓ *Structure*: the good energy resolution allowed to resolve some doublets such as the **resonance structure at 33, 92.5 or 96 eV**.





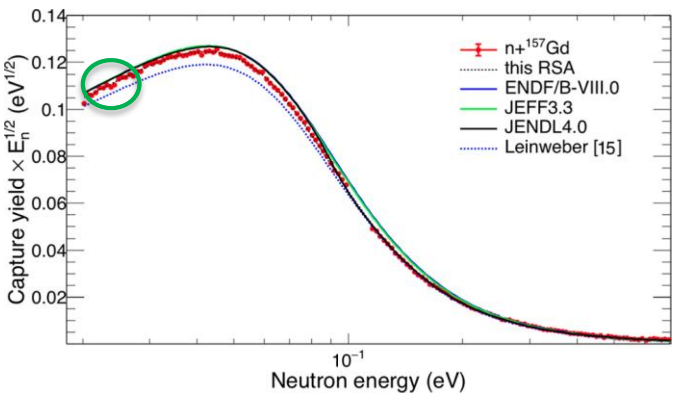




Data Analysis
(*Resonance Shape Analysis* – ^{157}Gd)

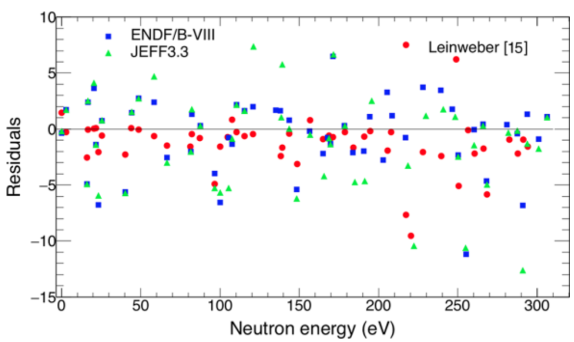
^{157}Gd

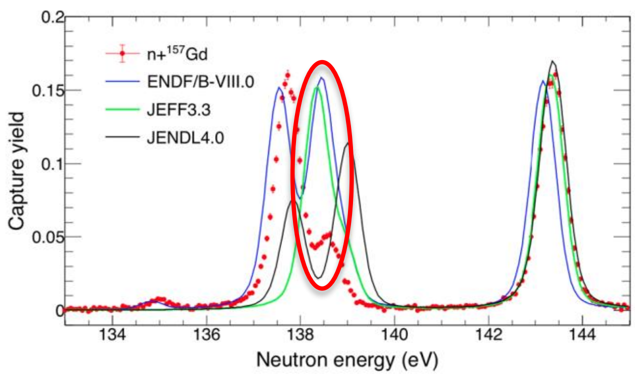
- ✓ ***Present thermal point***: is similar among libraries (253.2 ÷ 254.5 kb). Leinwwber *et al*. **[2]** deduced a smaller value (-12%).
- ✓ ***N_TOF thermal point:*** was estimated a value of **239.8 ± 8.4 kb**; this data settle between the two groups of evaluated values. WF is 0.89.
- ✓ ***Resonance***: According to ENDF/B-VIII.0 with **the exception of 139 eV value**. Only **one resonance was observed at 207.725 eV**.

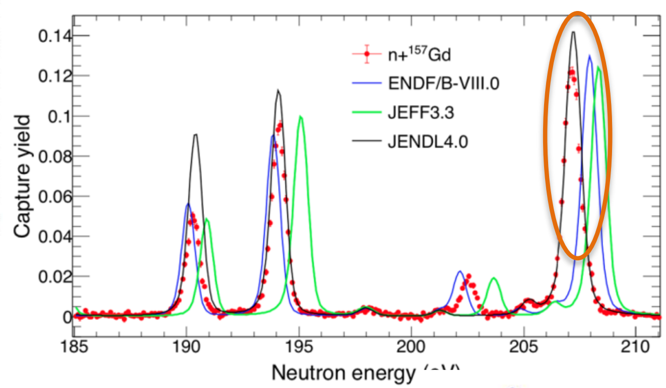


- ✓ **Kernel: *R**i* comparison reveals a average good agreement with ENDF/B-VIII.0 and JEFF-3.3 (Gaussian distribution, mean 0.98). On the contrary, JENDL-4.0 and **[2]** presents an average deviation of 13% (no Gaussian distribution).

$$R_i = \frac{k_\gamma^{n\_TOF} - k_\gamma^i}{j}$$
$$(i = eval.\ Xs;\ j = unc.)$$





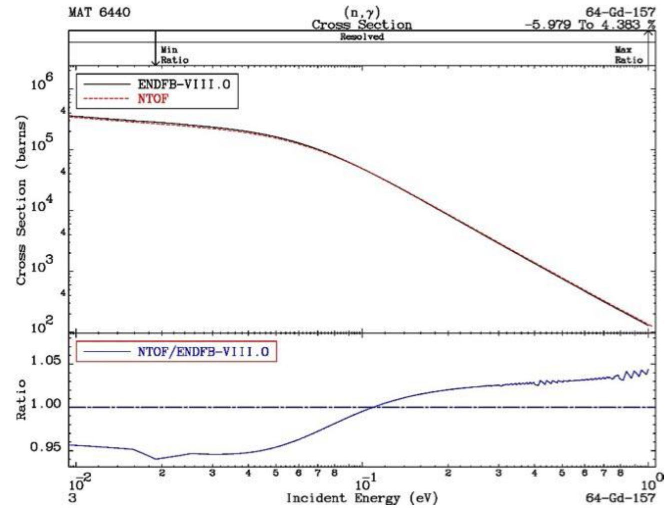


[2] G. Leinweber *et al*., Nucl. Sci. Eng. **154**, 261 (2006)

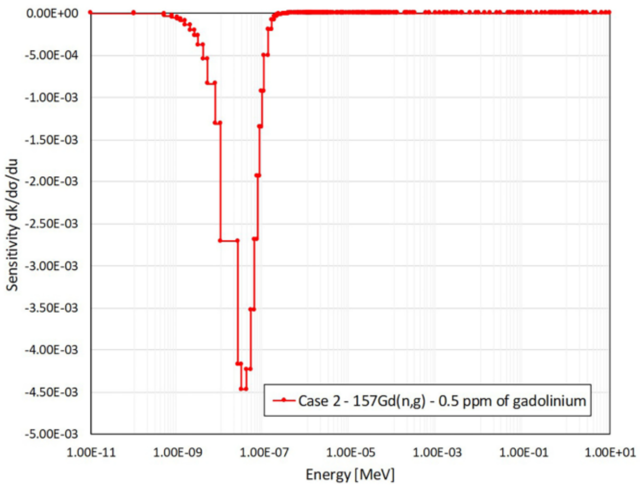
Impact of new results

Comparison between n_TOF and ENDF data

- ✓ New ENDF files were obtained updating the existing ENDF/B-VII evaluation with the new n_TOF data.
- ✓ ENDF n_TOF-based file for 157Gd (n,γ) was obtained with the COMPLOT module of the PREPRO2017 code suite **[10]**.
- ✓ ENDF/B-VII.0 was estimated on average -3.5% in thermal range (0.01÷0.1 eV) and about +2.5% in epithermal range (0.1÷1 eV) **[11]**.



Impact on a criticality thermal facility – ZED2



- ✓ ZED-2 is a criticality thermal-spectrum facility for which the bias on reactivity was already investigated.
- ✓ MCNP showed that 157Gd(n,γ) are overestimated in ENDF/B-VII.0 (254 kb) and underestimated in ENDF/B-VII.1β (226 kb).
- ✓ The criticality gain to attain the experimental criticality factor is about +22 pcm. The calculated theorical gain*,** is 6.1÷7.2 pcm higher **[8]**.

$$^{*}\Delta k_1 \approx k_{eff} \cdot S_1 \cdot \Delta\sigma_1/\sigma \ \ (1\ group\ theory) \ \Rightarrow \Delta k \approx +29.2\ pcm$$

$$^{**}\Delta k_2 \approx k_{eff} \cdot S_1 \cdot \Delta\sigma_1/\sigma + k_{eff} \cdot S_2 \cdot \Delta\sigma_2/\sigma \ (2\ group\ theory) \ \Rightarrow \Delta k \approx +28.1\ pcm$$

- ✓ n_TOF thermal point (239.8 kb) can be considered a better estimation.

[10] F. Rocchi *et al.*, Il nuovo Cimento **42 C** (2019) 140.
[11] F. Rocchi *et al.*, Ann. of Nuc. Ene. **132** (2019) 537-543.

Conclusions

- **^{155,157}Gd(n,γ)** despite their reactor safety-related importance, also stated by OECD/NEA that includes this isotopes in the High Priority Request List (HPRL), are presently know with an accuracy not full adequate to the nuclear industry purpose.

- **In 2016** new measurements have been performed at EAR1 hall of CERN n_TOF facility that has allow to obtain new experimental data with an uncertainty between 3.2 and 3.5%.

- **The data analysis** of the n_TOF results revealed some remarkable difference compared to the existing evaluated data in terms of resonance structure and kernel. ^{157}Gd thermal point value was found between ENDF/B-VII.0 and ENDF/B-VII.1 evaluations.

- **A comparison** between ENDF n_TOF related and existing ENDF data has revealed an average discrepancy between -3.5% and +2.5% in the 0.01 ÷ 10 eV energy range.

- **A preliminary validation** of the n_TOF ^{157}Gd data on a criticality thermal facility (ZED-2) showed that the new data can be considered a better estimation compared to the old ones.

- **New measurements** are currently underway at JRC GELINA facility to try to further reduce the thermal data uncertainty. In the future, other ICSBEP benchmarks will be used for additional validation of the new n_TOF data.

Grazie per l'attenzione!

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