

DE LA RECHERCHE À L'INDUSTRIE



# Improved MOX fuel calculations using new Pu-239, Am-241 and Pu-240 evaluations

Wonder 2012 | G. Noguere, O. Bouland, D. Bernard, P. Leconte, P. Blaise, Y. Pennelieu, J.F. Vidal,  
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# Context

- Extensive experimental programs, carried out in the zero-power research reactor EOLE of the CEA Cadarache, were designed to study advanced LWR and BWR core configurations of MOX fuel assemblies.
- The parameter of interest in this work is the residual reactivity.
- New reactivity calculations using the TRIPOLI-4 Monte-Carlo Code with the JEFF-3.1.1 nuclear data library have confirmed the systematic overestimation of the  $k_{\text{eff}}$ .

# Integral experiments

MH1.2 (EOLE, Cadarache)	PWR-MOx mixed core	4 years
BASALA-Hot (EOLE, Cadarache)	BWR-MOx BWR-MOx	12 years 13 years
MISTRAL-2 (EOLE, Cadarache)	PWR-MOx	8 years
MISTRAL-3 (EOLE, Cadarache)	PWR-MOx	9 years
MISTRAL-4 (EOLE, Cadarache)	MOx-REF MOx-AIC MOx-Hf MOx-B4C	10 years 10 years 10 years 10 years
FUBILA-Hot (EOLE, Cadarache)	BWR-REF (EPICURE) BWR-NORM (EPICURE) BWR-70% Void (EPICURE) BWR-10×10 (EPICURE) BWR-UGD (EPICURE)	1 years (16-17 years) 1 years (16-17 years) 1 years (16-17 years) 1 years (16-17 years) 1 years (16-17 years)
ERASME (EOLE, Cadarache)	ERASME/R, HCPWR ERASME/S	
OSMOSE (MINERVE, Cadarache)	Oscillation measurements (R1U02 and R1MOX lattices )	
Post Irradiated Experiments (PIE)	GRAVELINE (ALIX-HTC), GUNDREMMINGEN, DAMPIERRE, ...	

# Am-241 evaluation

## New evaluation of the Resolved Resonance Range ( $E < 150$ eV)

- IRMM/CEA Collaboration (PhD Thesis C. Sage, sample preparation, ...)
- Neutron Resonance Shape Analysis with REFIT/CONRAD
- Resonance Parameter Covariance Matrix generated by marginalization

## ECIS/TALYS description of the « continuum » ( $150$ eV $< E < 20$ MeV)

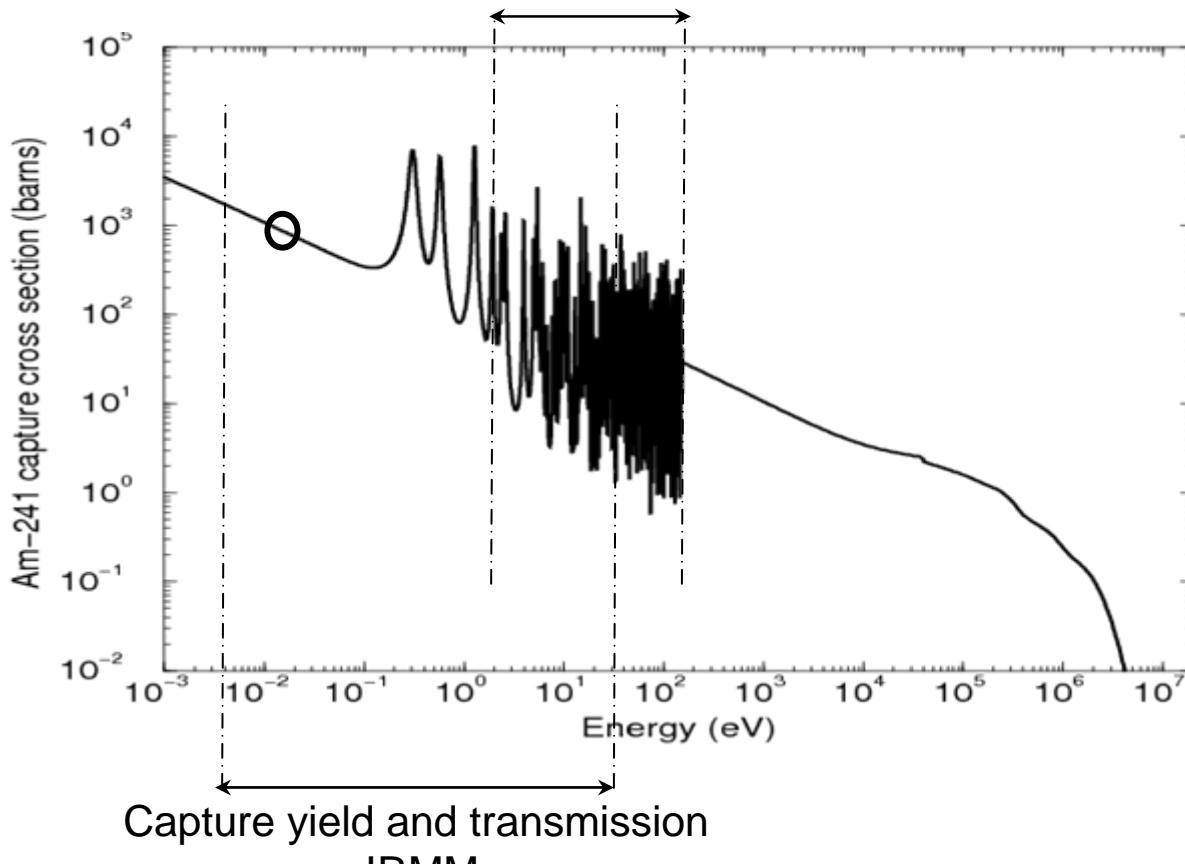
- Collaboration with the Bucarest University (PhD Thesis C. Morariu)
- Collaboration with Los Alamos National Lab. (« head band states »)

## Integral benchmarks

- MELUSINE (Grenobles)
- MINERVE, EOLE, MASURCA (Cadarache)
- PHENIX (Marcoule)
- Monte Carlo code TRIPOLI-4
- Deterministic codes APOLLO-2.8 and ERANOS-2

# Am-241 evaluation

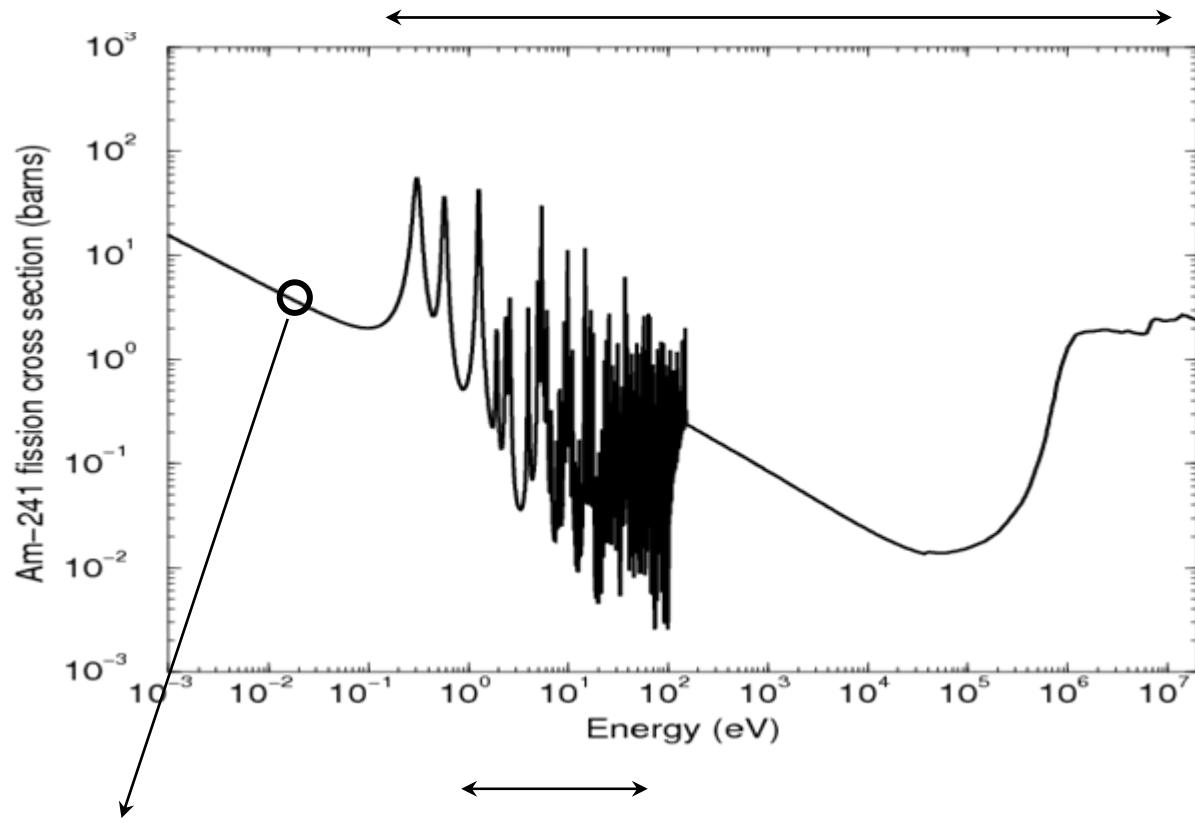
Capture yield from G. Vanpraet, IRMM, 1985  
Transmission from H. Derrien, Saclay, 1975



See talk of C. Lampoudis

# Am-241 evaluation

J.W.T. Dabbs, ORELA, 1983



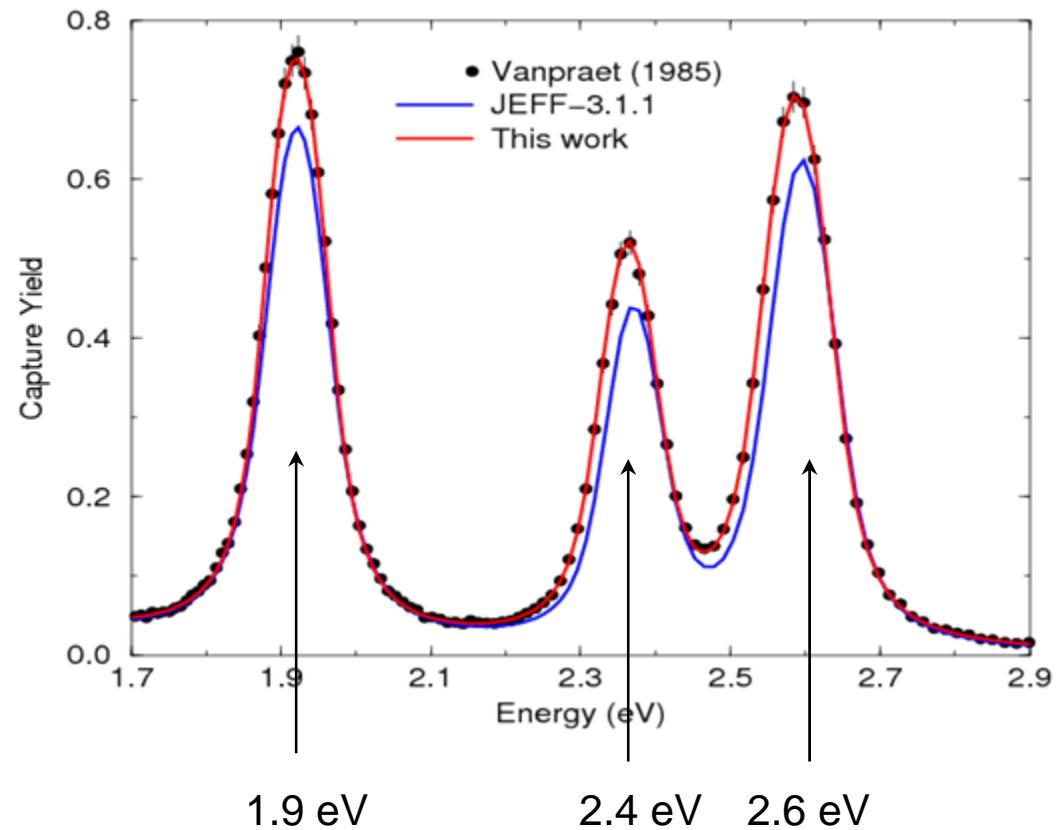
Alain Letourneau  
MINI-INCA (ILL)

Derrien  
Saclay  
1975

# Am-241 evaluation

## Capture cross section (C6D6) : G. Vanpraet et al. (1985)

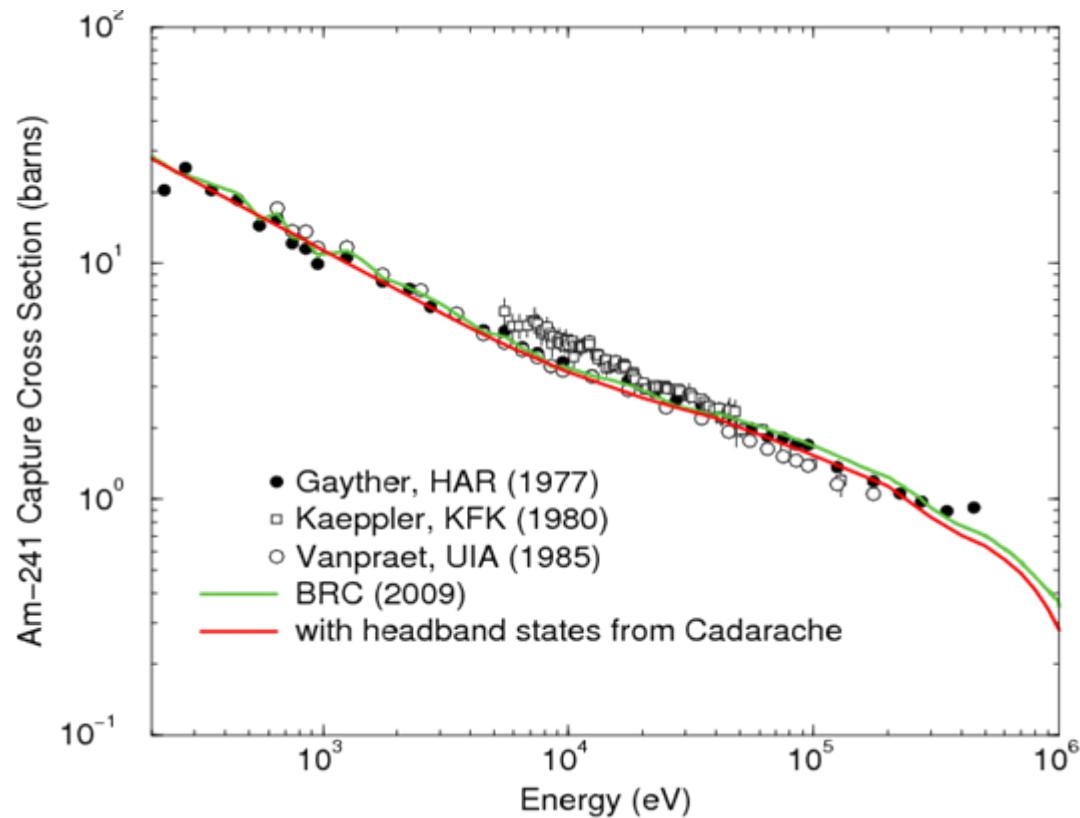
Re-normalization of the capture Yield measured by Vanpraet by using the transmission data measured at the IRMM



# Am-241 evaluation

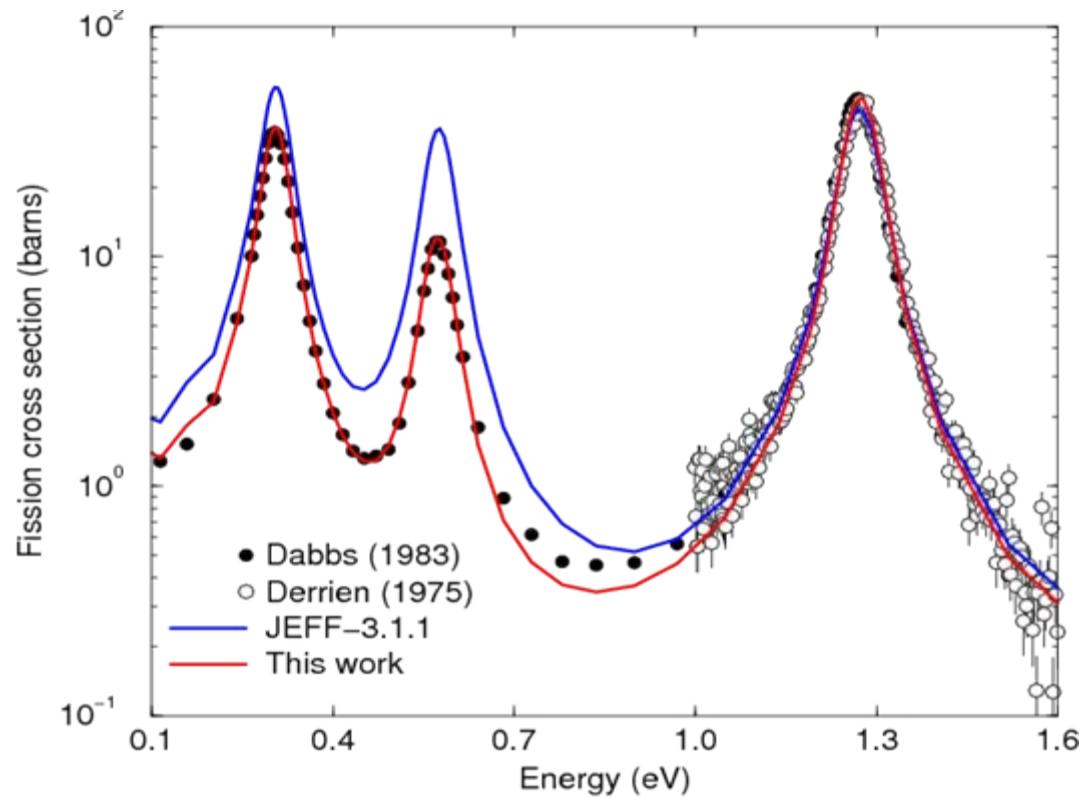
## Capture cross section :

ECIS/TALYS calculations based on the Optical Model Parameters of Soukhovitskii et al,  
Phys. Rev. C72, 024604 (2005) ⇒ see talk of J.M. Quesada Molina



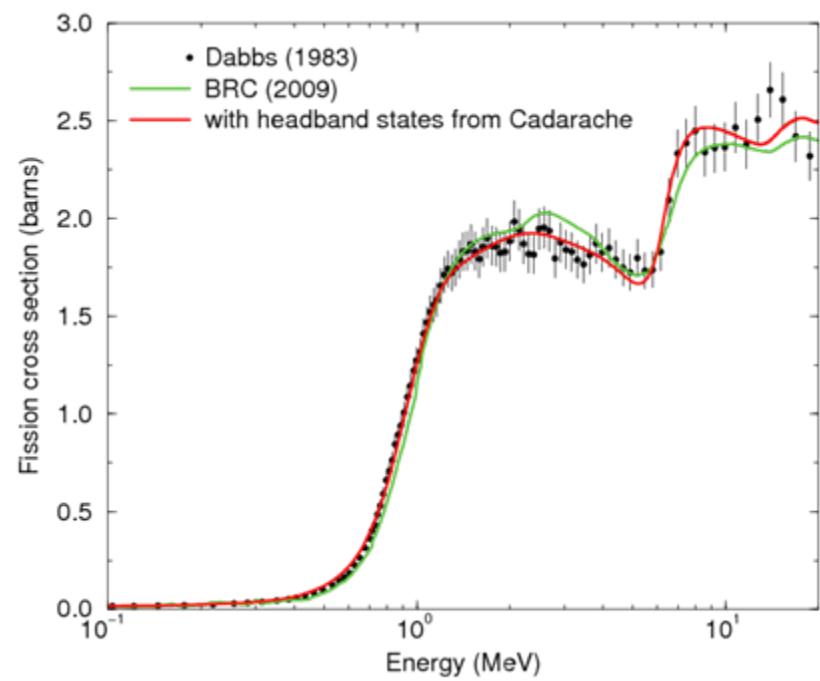
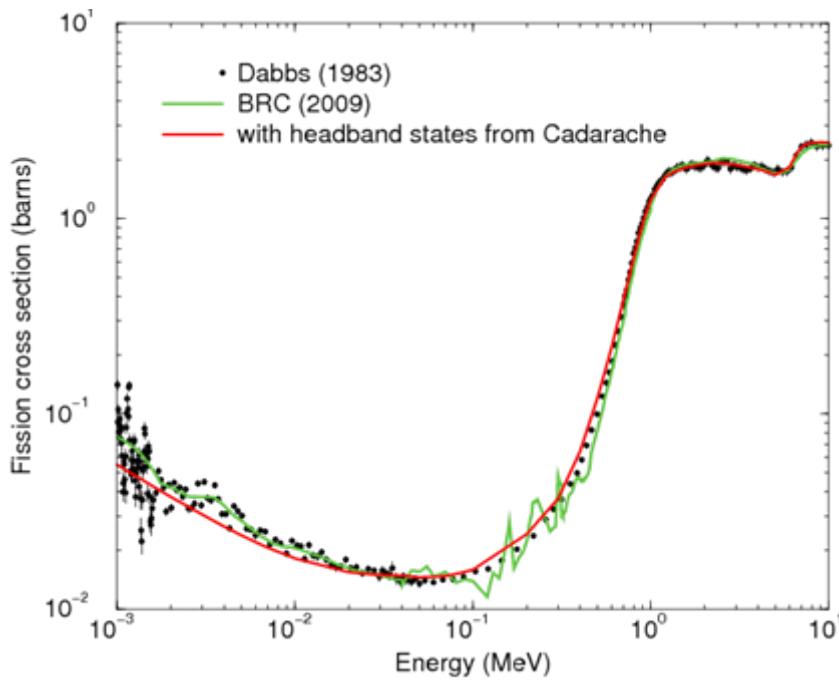
# Am-241 evaluation

Fission cross section : Dabbs (ORELA, 1983) and Derrien (Saclay, 1975)



# Am-241 evaluation

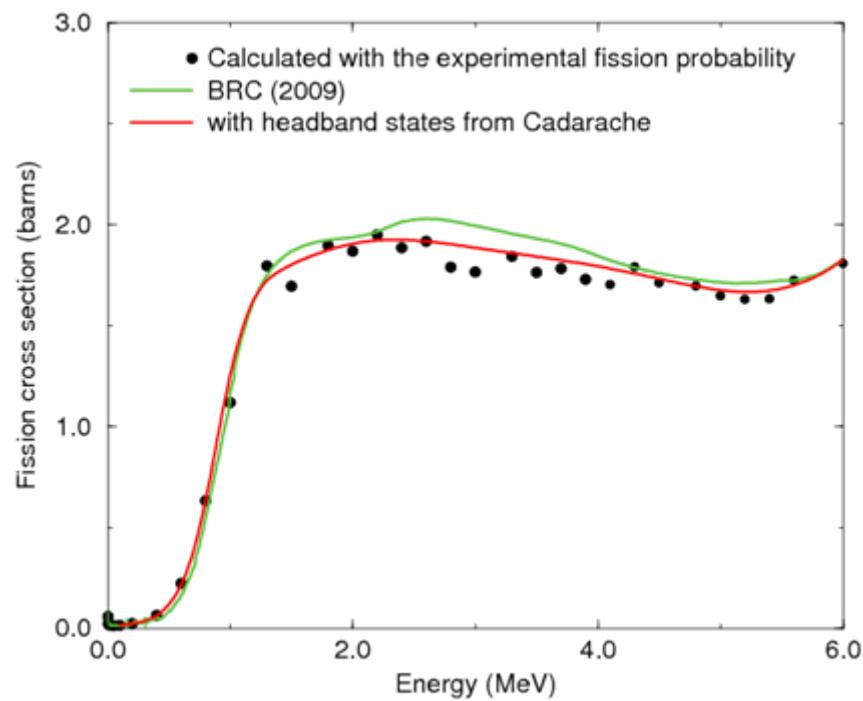
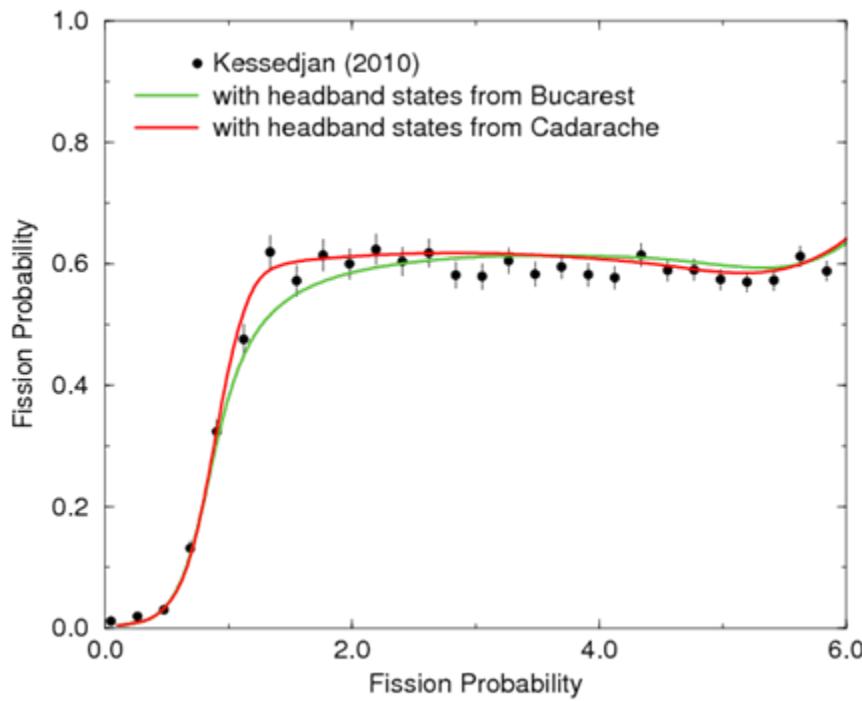
**Fission cross section : ECIS/TALYS calculations**  
Head-band states based on the work of Olivier Bouland (LANL/CEA collaboration)



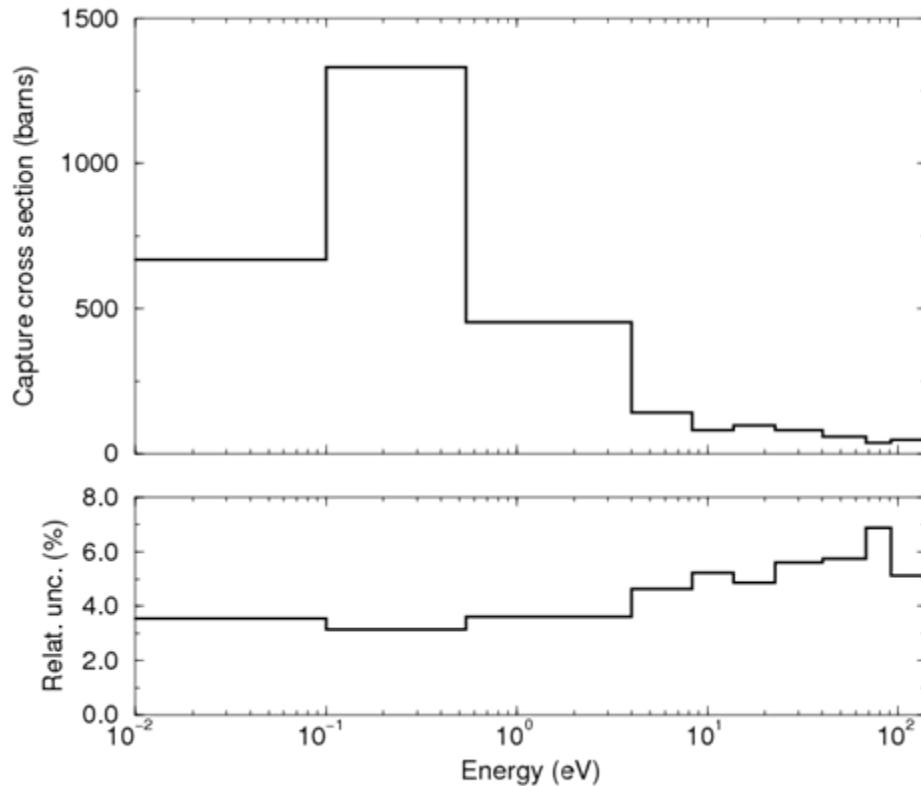
# Am-241 evaluation

**Fission Probability** : surrogate-reaction method, CENBG

See talk of Beatriz Jurado



# Am-241 evaluation



**Thermal Capture Xs (+15%)**

JEFF-3.1.1 = 647 b

New =  **$746 \pm 24$  b**

**Capture resonance integral (+20%)**

JEFF-3.1.1 : 1526 b

New :  **$1825 \pm 65$  b**

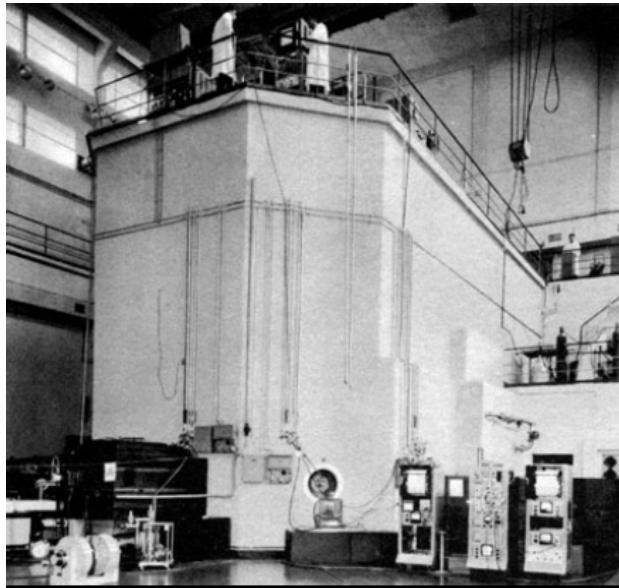
**Fission resonance integral (-19%)**

JEFF-3.1.1 : 17.3 b

New :  **$14.5 \pm 1.0$  b**

# Am-241 evaluation

## ICARE-S program (1986)



Irradiation of two Am-241 samples in MELUSINE (Grenoble, France)

This experiment contained rods with samples of almost pure actinide isotopes separated into small quantities.

The variables of interest for the interpretation of ICARE are the sample composition at the beginning and at the end of the irradiation period (6 months)

The **Integral Data Assimilation** technique implemented in the CONRAD code was used to extract the capture resonance integral from the ICARE-S experiment

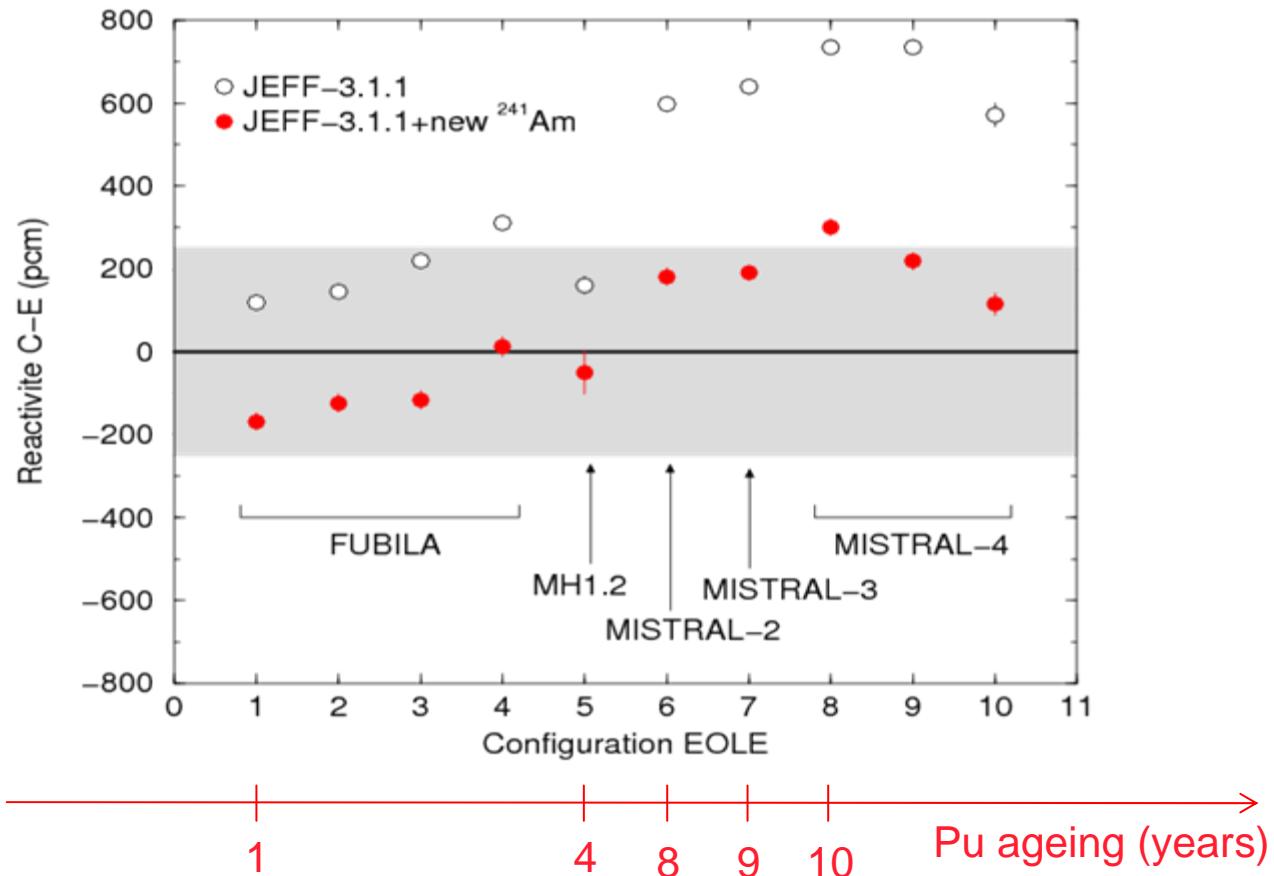
$$I_0 \text{ (JEFF-311)} = 1526 \text{ b}$$

$$I_0 \text{ (new)} = 1825 \pm 65 \text{ b}$$

$$\mathbf{I_0 \text{ (ICARE-S)} = 1895 \pm 90 \text{ b}}$$

# Am-241 evaluation

## zero-power research reactor EOLE (Cadarache)



# Pu-239 evaluation

## New evaluation of the Resolved Resonance Range ( $E < 2.5$ keV)

- ORNL/NEA/CEA Collaboration (Luiz Leal)
- Neutron Resonance Shape Analysis with SAMMY/CONRAD
- Resonance Parameter Covariance Matrix generated by marginalization
- Investigation of the  $(n,\gamma f)$  reaction
- Phenomenological description of the neutron multiplicity  $\nu_p$

## Description of the « continuum » ( $2.5$ keV $< E < 30$ MeV)

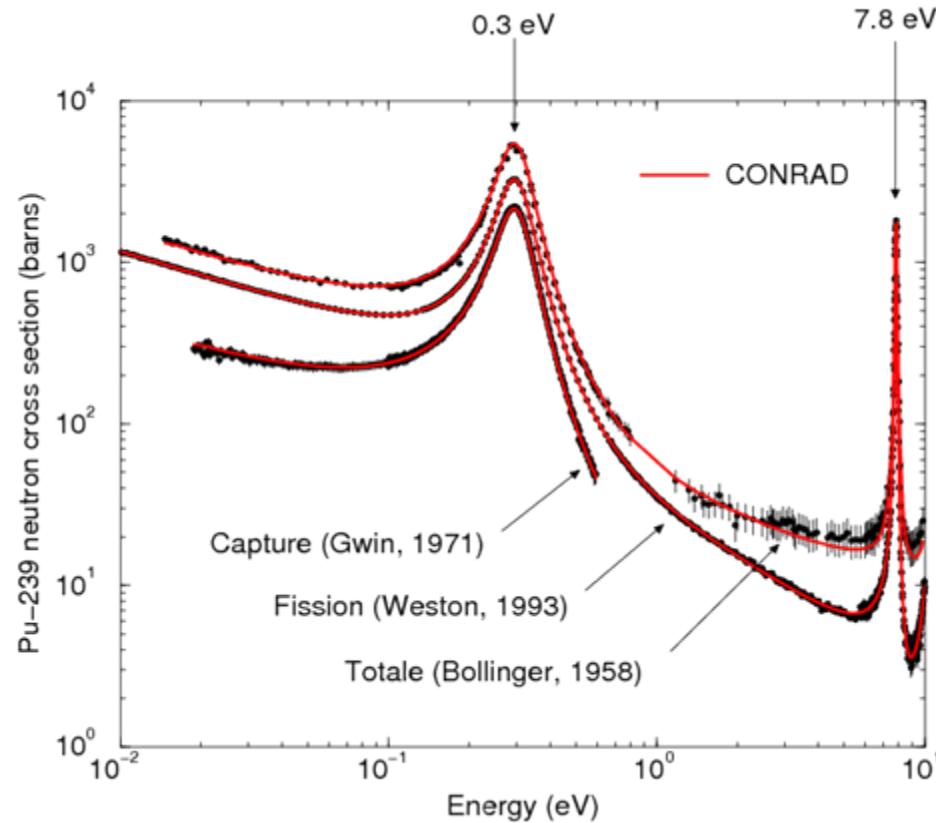
- CEA/Dam of Bruyère-Le-Châtel  $\Rightarrow$  [See talk of Yannick Penelaiou](#)
- LANL/CEA Collaboration  $\Rightarrow$  [See talk of Olivier Bouland](#)

## Integral benchmarks

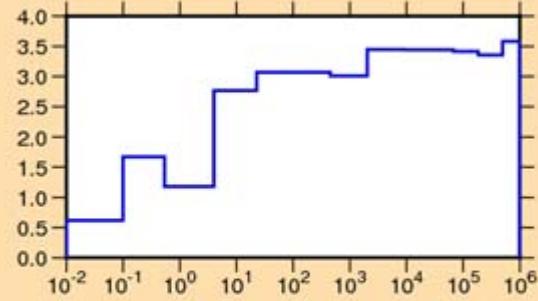
- ICSBEP (Pu-Sol-Therm) and EOLE (Cadarache)
- Monte Carlo code TRIPOLI-4
- Deterministic code APOLLO-2.8

# Pu-239 evaluation

**WPEC/SG34, Co-ordinator : C. De Saint Jean**  
**« Coordinated evaluation of Pu-239 in the resonance region »**  
See talk of A.C. (Skip) Kahler

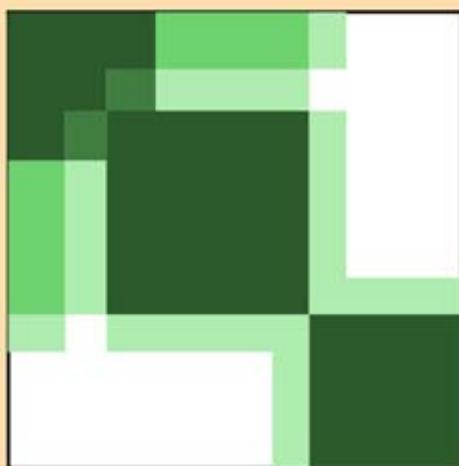


$\Delta\sigma/\sigma$  vs. E for (n,f)

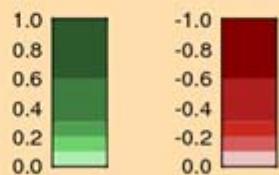


Ordinate scales are % relative standard deviation and barns.

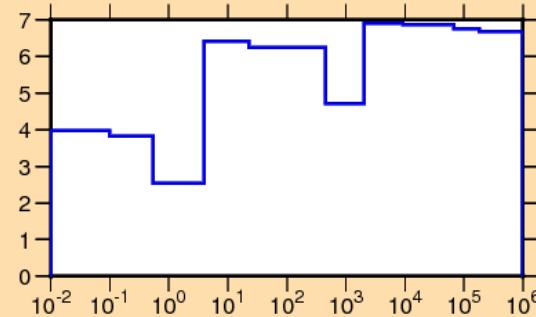
Abscissa scales are energy (eV).



Correlation Matrix

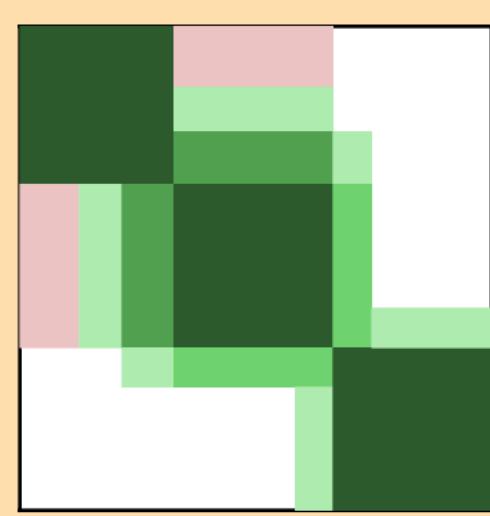


$\Delta\sigma/\sigma$  vs. E for (n, $\gamma$ )

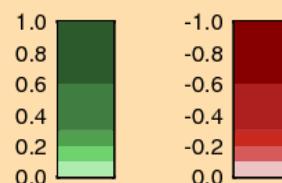


Ordinate scales are % relative standard deviation and barns.

Abscissa scales are energy (eV).



Correlation Matrix



# Pu-239 evaluation

## New investigation of the two-step ( $n,\gamma f$ ) process

- 1959** : unpublished estimation of the  $\Gamma_{\gamma f}$  width for the ( $n,\gamma f$ ) reaction by E. Lynn
- 1965** : On the slow neutron, gamma-fission reaction, E. Lynn, Phys. Lett. 18
- 1967** : Evaluation des données neutroniques pour le Pu-239, G. LeCoq, PhD thesis
- 1973** : Etudes des sections efficaces de réaction des neutrons de resonance avec Pu239, H. Derrien, PhD thesis
- 1974** : Etude des neutrons et des rayons gamma émis lors de la fission induite dans 235U et 239Pu par neutrons lents: mise en évidence de la réaction ( $n,\gamma f$ ), D. Shackleton, PhD thesis
- 1980** : The double-humped fission barrier, S. Bjornholm and E. Lynn, Rev. Mod. Phys. 52
- 1988** : **Evaluation of  $v_p$  for Pu-239, E. Fort et al. Nucl. Sci. Eng. 99**

# Pu-239 evaluation

The evaluation of the neutron multiplicity  $\nu_p$  takes into account

- 2 opened fission channels for  $J^\pi=0^+ \Rightarrow \Gamma_{f1}(0^+)$  and  $\Gamma_{f2}(0^+) \Rightarrow \Gamma_f(0^+)$
- 1 opened fission channels for  $J^\pi=1^+ \Rightarrow \Gamma_f(1^+)$
- J-dependent width for the  $(n,\gamma f)$  reaction  $\Rightarrow \Gamma_{\gamma f}(0^+)$  and  $\Gamma_{\gamma f}(1^+)$

## Phenomenological description

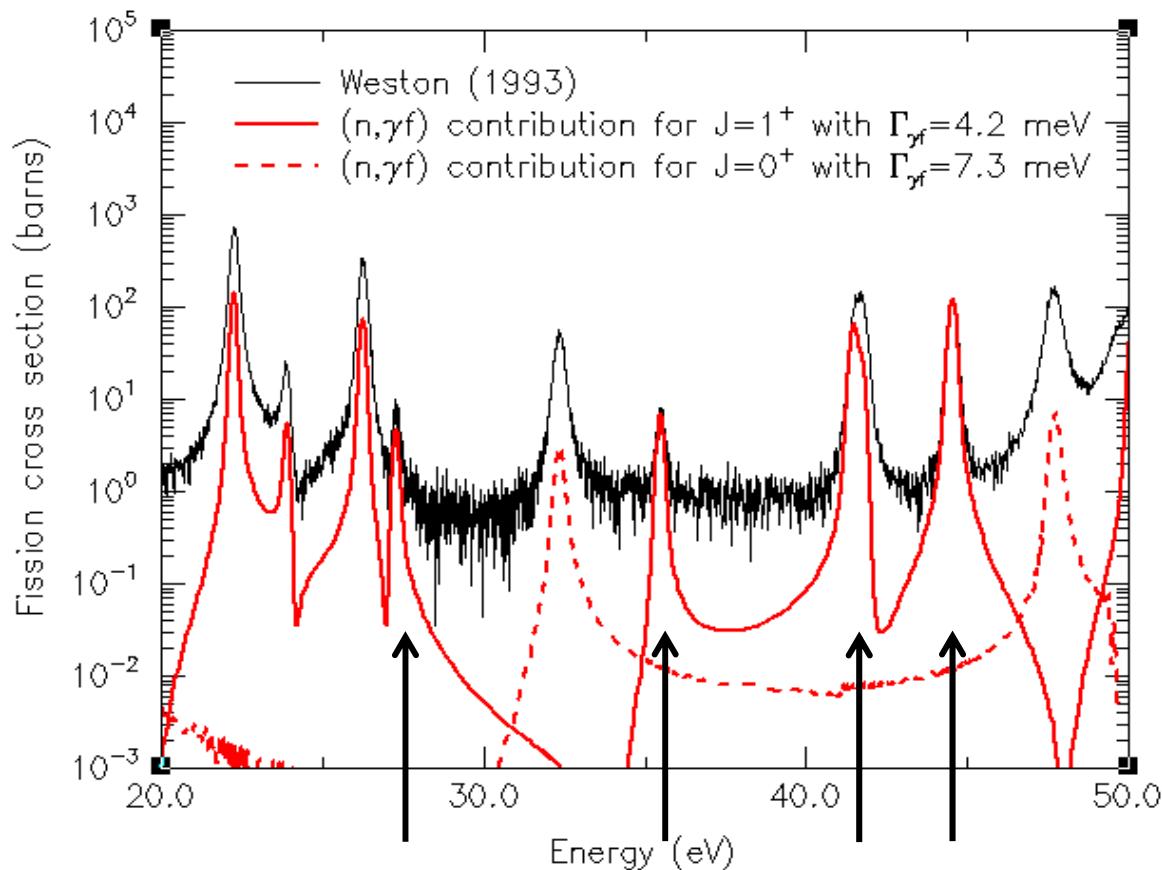
$$\nu_p(E) \approx \sum_{i=1}^4 \nu_i P_i(E)$$

Where

$$P_i(E) = \frac{\sigma_{fi}(E)}{\sigma_f(E) + \sigma_{\gamma f}(E)}$$

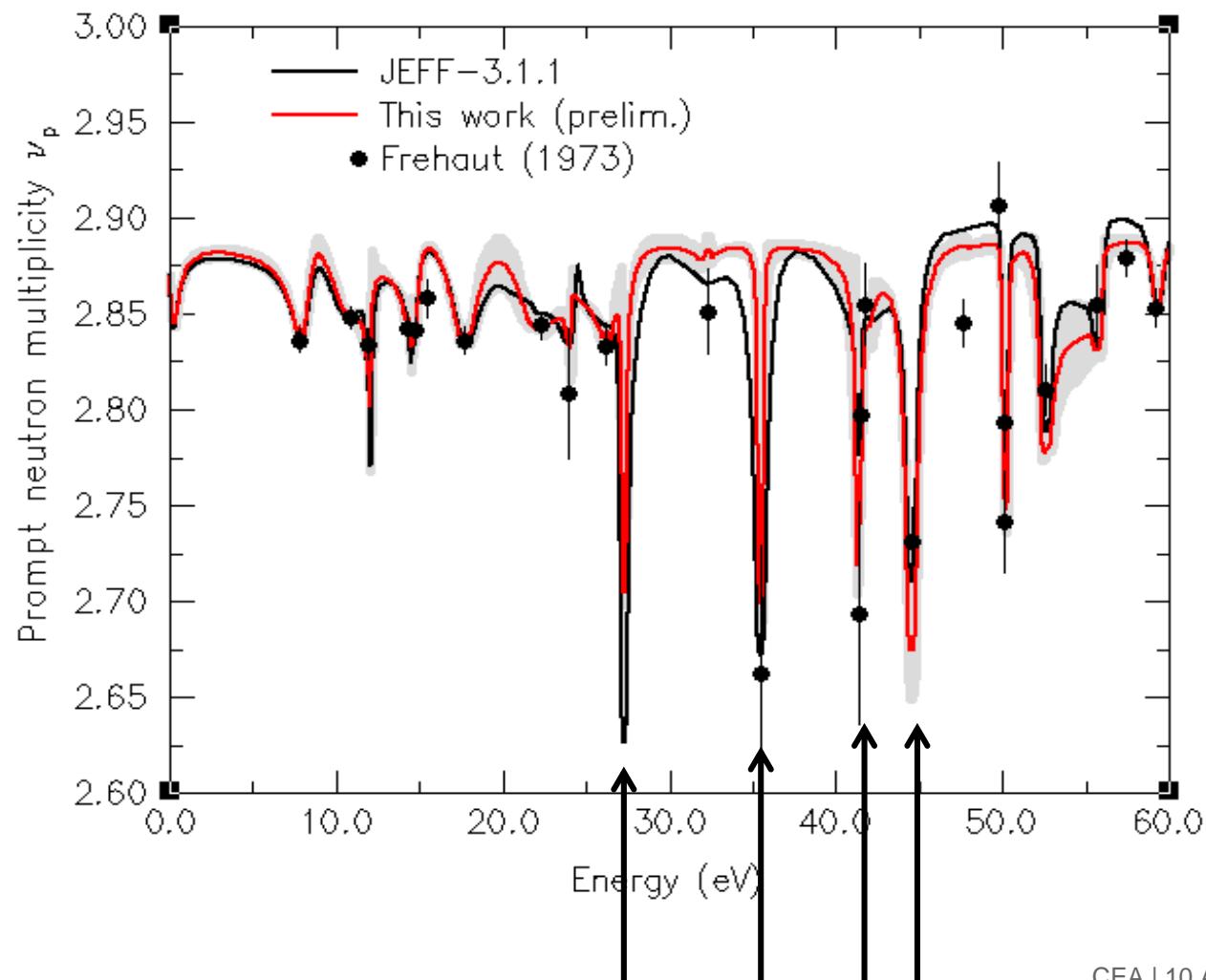
# Pu-239 evaluation

For  $J^\pi=1^+$ , the smallest resonances are due to the  $(n,\gamma f)$  process



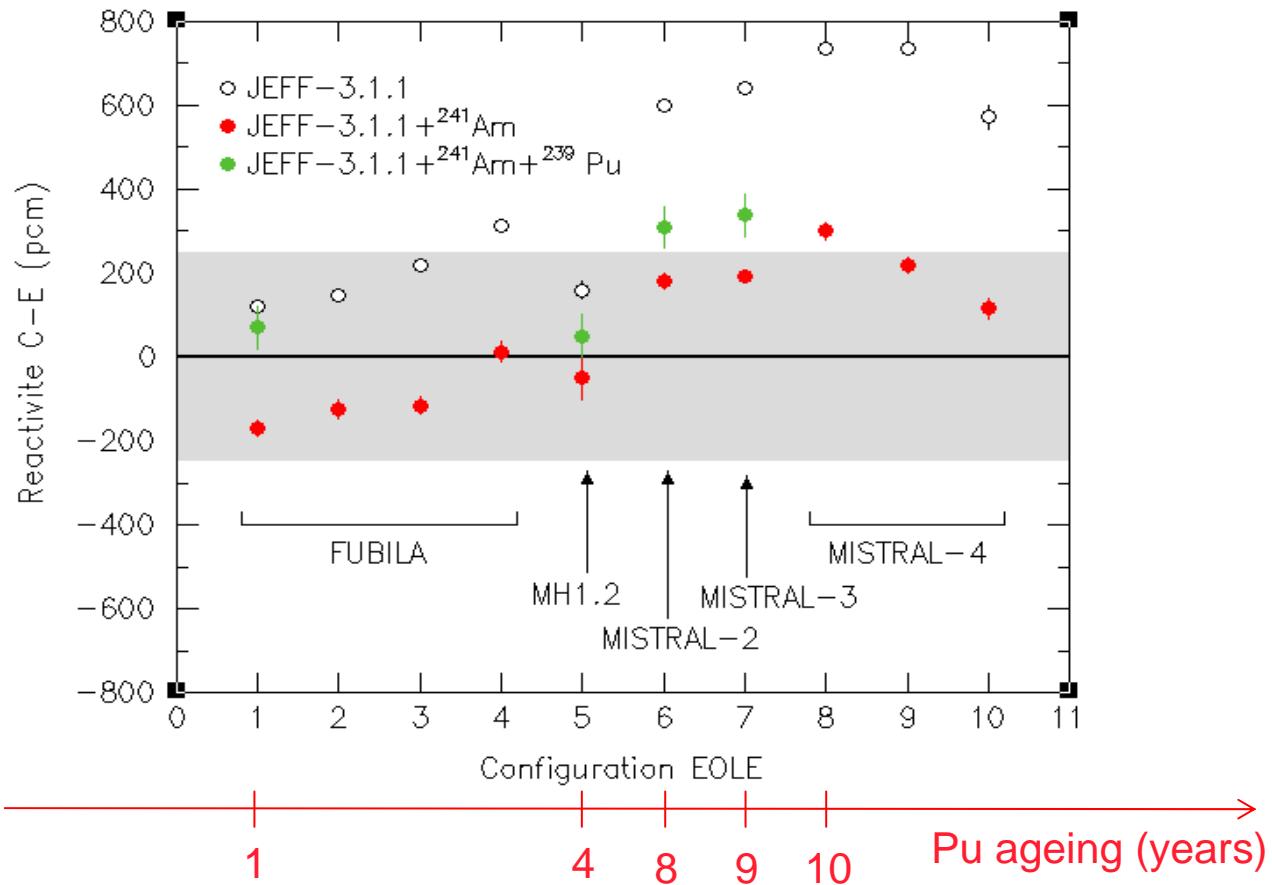
# Pu-239 evaluation

The contribution of the  $(n,\gamma f)$  process can be observed for resonances with  $J^\pi=1^+$



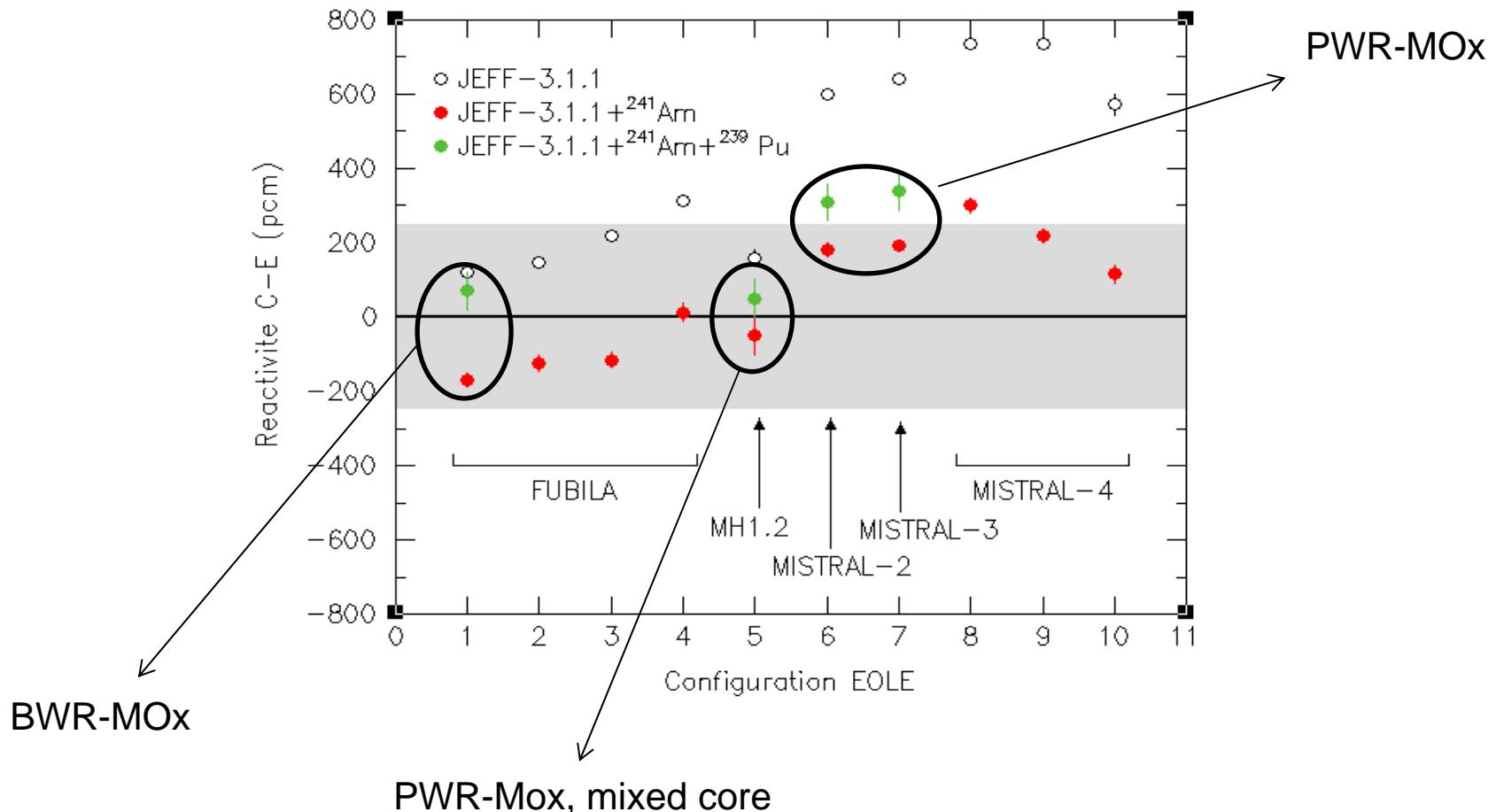
# Pu-239 evaluation

## zero-power research reactor EOLE (Cadarache)



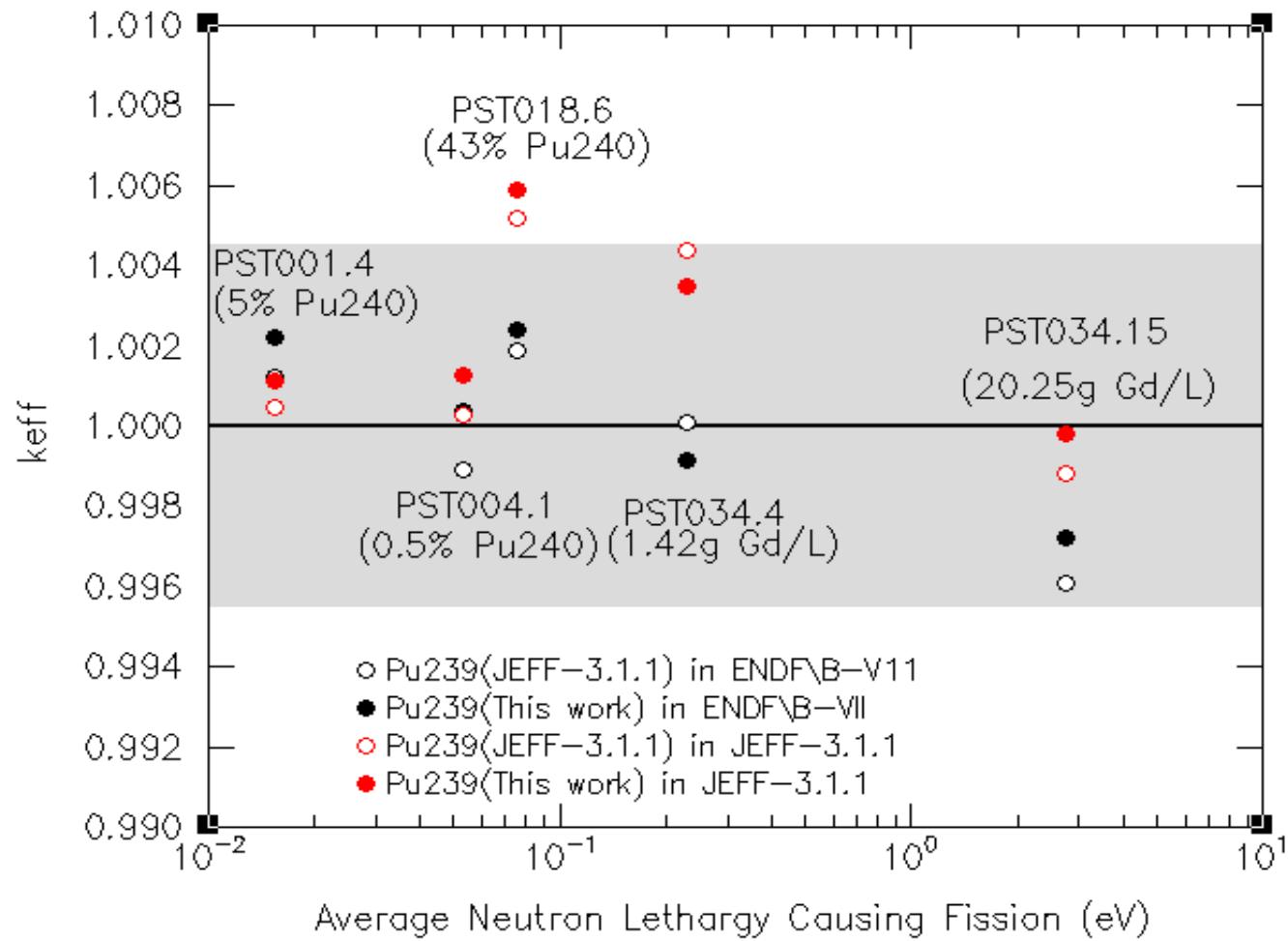
# Pu-239 evaluation

## zero-power research reactor EOLE (Cadarache)



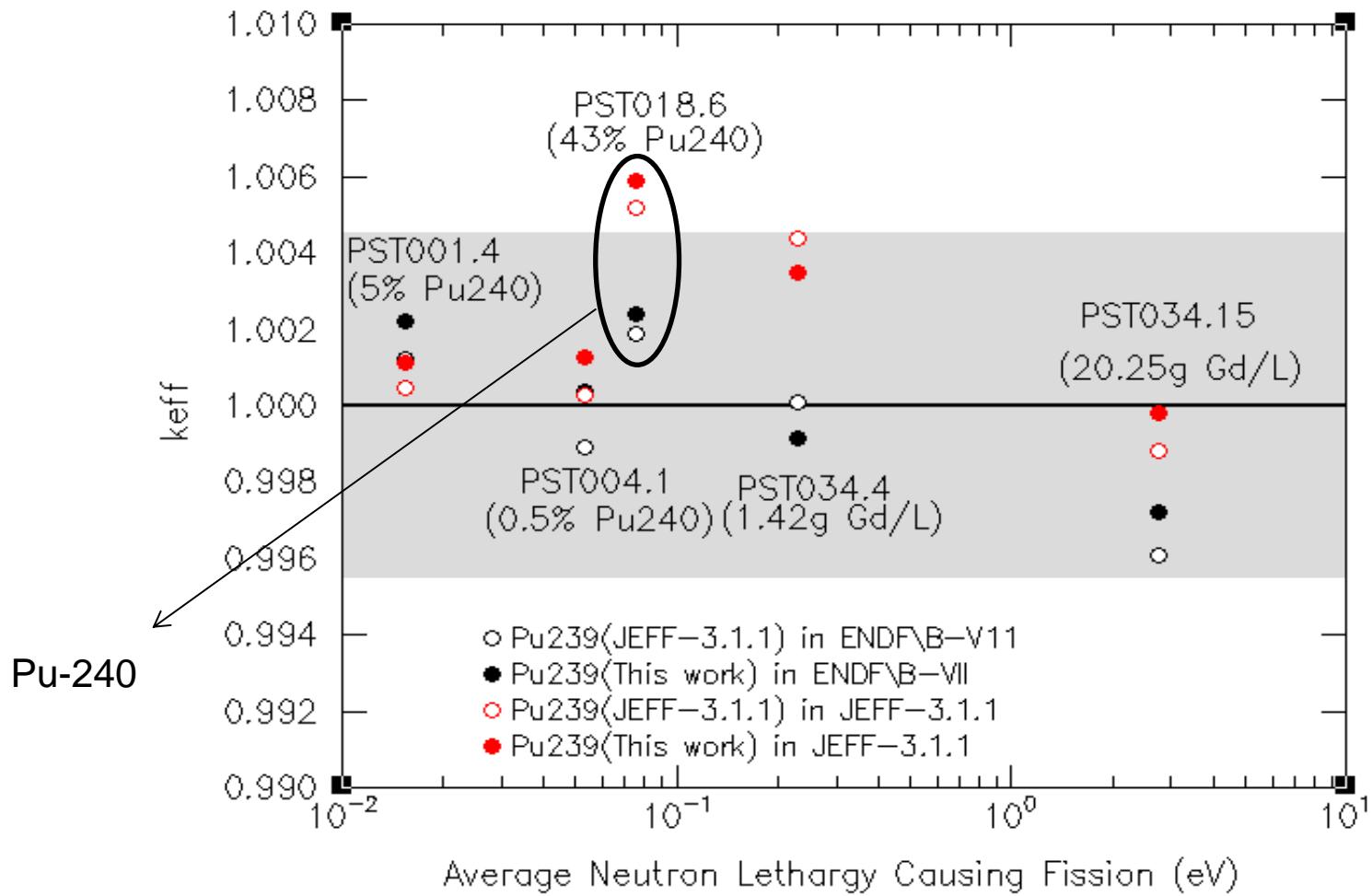
# Pu-239 evaluation

## ICSBEP benchmark results : JEFF-3.11 Vs. ENDF\B-VII



# Pu-239 evaluation

## ICSBEP benchmark results : JEFF-3.11 Vs. ENDF\B-VII



# Pu-240 evaluation

A new evaluation of Pu-240 is in progress (Luiz Leal, ORNL)

The present work can give some recommendations on the radiation width of the first resonance at 1 eV

Author	Reference	Radiation width $\Gamma\gamma$
O. Bouland	NSE 127, 105 (1997)	$29.14 \pm 0.6$ meV (2%)
S.F. Mughabghab	Atlas of Neutron Resonances, 2006	$29.14 \pm 0.6$ meV (2%)
JEFF-3.1.1		29.15 meV
A. Meister	Private communication	$30.50 \pm 0.31$ meV (1%) $30.39 \pm 0.13$ meV (0.4%)

→ Cristal Latice Model

Free Gas Model ←

# Pu-240 evaluation

Impact of the Pu-240  
evaluation of ENDF\B-VII

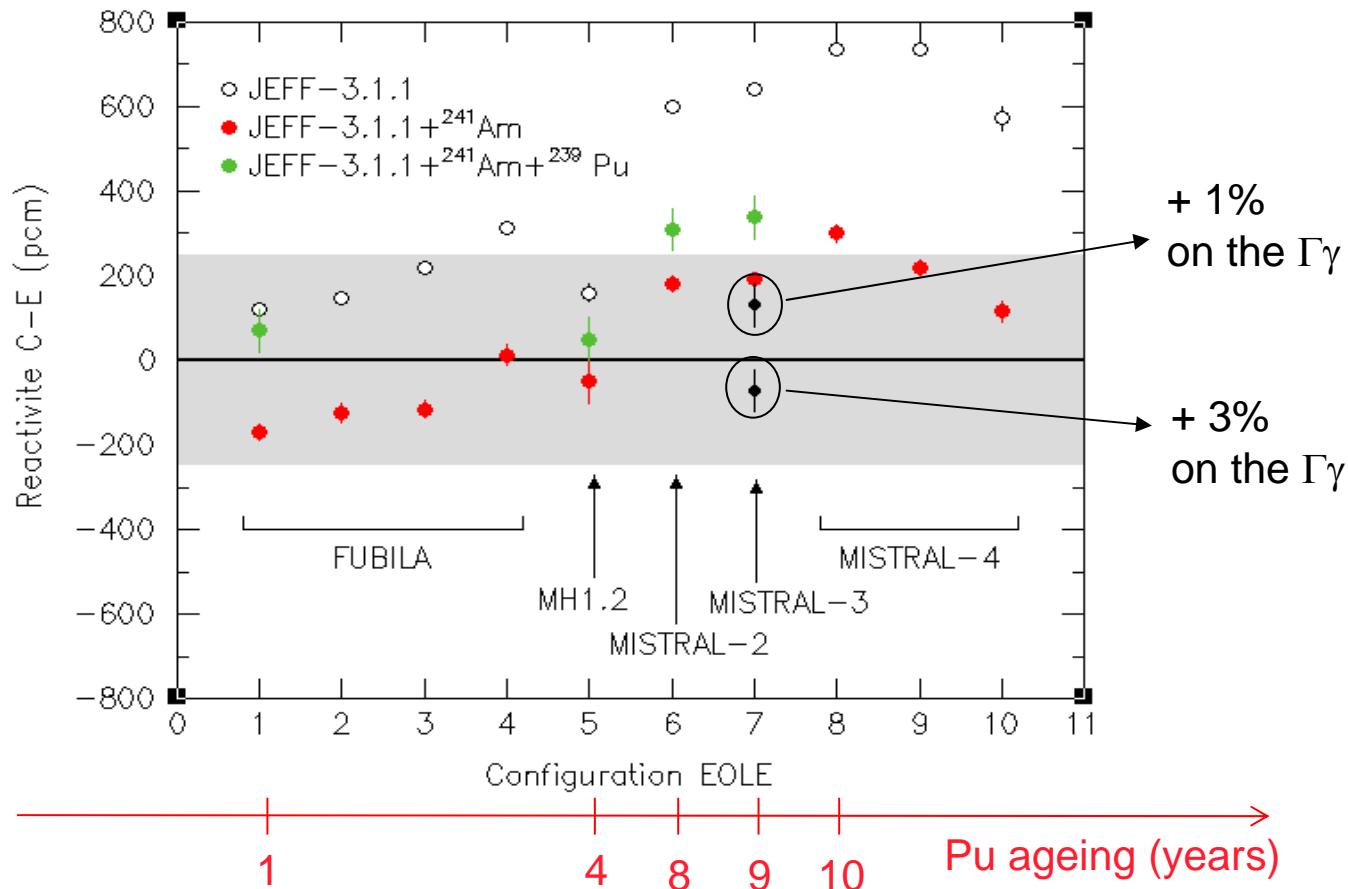
Direct perturbation of the  
radiation width of JEFF-3.1.1  
(first resonance at 1 eV)

The diagram consists of two arrows. One arrow originates from the text "Impact of the Pu-240 evaluation of ENDF\B-VII" and points to the second column of the table. The other arrow originates from the text "Direct perturbation of the radiation width of JEFF-3.1.1 (first resonance at 1 eV)" and points to the third and fourth columns of the table.

Benchmark	ENDF\B-VII	+ 3 %	+ 1 %
FUBILA-REF	-190±50 pcm		
MISTRAL-2	-390±50 pcm		
MISTRAL-3	-340±50 pcm	-410±50 pcm	
PST-18.6	-420±50 pcm	-420±50 pcm	-230±50 pcm

# Pu-240 evaluation

## zero-power research reactor EOLE (Cadarache)



# Conclusions

- Significant improvements of the  $k_{\text{eff}}$  results by using the new Am-241 et Pu-239 evaluation
- The remaining overestimation (~200 pcm) could be explained by Pu-240
- New Time Of Flight measurements of the totale cross section of Pu-239 and Pu-240 (77K, 300 K and  $T>300$  K) are needed
- Use of the Cristal Lattice Model for the Neutron Resonance Shape Analysis (phonon spectrum is needed)