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Nuclear data requirements for an accurate estimation of the neutron production rate of spent nuclear fuel

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# Introduction

- Spent Fuel Characterisation
  - Pool storage
  - Transport
  - Interim storage
  - Repository systems





- **Observables:** DH, n and  $\gamma$  emission, reactivity, fissile material, long-lived radionuclides
- Isotopic content determined with **fuel depletion codes**







## Radiochemical analyses (pellet average)

- Medium to high BU UO<sub>2</sub> and MOX fuels (30-60 GWd/t<sub>HM</sub>)
- PWR (commercial) & BWR (prototype) reactors

## Local investigations (radial profile)

• SIMS & EPMA

### Representative and validated data

- Code validation (core reactivity, transport, storage)
- Source term studies (back-end)

• ...

# **Radiochemical analysis**



#### **Detailed databook**



RCA results										
BU	Analysis date	Nuclide Inventory	BU							
indicator		Nx∕N∪	MWd/kg							
<sup>137</sup> Cs	21/10/2013	2.539 (55) x 10 <sup>-3</sup>	52.6 (11)							
<sup>143+144</sup> Nd	05/02/2014	5.701 (60) x 10 <sup>-3</sup>	53.95 (56)							
<sup>145+146</sup> Nd	05/02/2014	3.643 (38) x 10 <sup>-3</sup>	53.05 (56)							
<sup>148</sup> Nd	05/02/2014	0.974 (21) x 10 <sup>-3</sup>	53.3 (12)							
<sup>150</sup> Nd	05/02/2014	0.463 (21) x 10 <sup>-3</sup>	52.2 (23)							
		Avera	ge: 52.78 (37)							

#### Operator-based BU: 54.30 MWd/kg

**Source**: P. Schillebeeckx *et al. An absolute measurement of the neutron production rate of a spent nuclear fuel sample used for depletion code validation.* Front. Energy Res. 11 (2023) 1162367

## **Neutron measurements**

- Provide experimental data to validate depletion codes
  - <sup>244</sup>Cm inventory: reduced uncertainty compared to radiochemical analysis
- Experimental method to verify declared burnup
  - Extremely sensitive to BU
  - Axial BU profiles
- Important for the validation of any characterisation scheme based on depletion calculations
  - Support nuclear safeguards verification by e.g. FORK, DDSI, PNAR measurements
  - Support nuclear criticality safety procedures relying on loading curves
  - Support decay heat estimates based on depletion calculations

## **Neutron measurements**





Source: P. Schillebeeckx. Neutrons as a signature for the characterization of spent nuclear fuel. EURAD Annual Event 2021

#### **SNF segment sample: characteristics**

Parameter	Value
Segment length	52.01 (4) mm
Segment weight	42.616 (1) g
Cladding weight	6.71 (4) g
Net fuel weight	35.91 (4) g

 $S_{sf} = 678 (12) s^{-1}g^{-1}$  $S_{\alpha n} / S_{sf} = 0.039 (18)$ 









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# **Depletion calculations**

- ALEPH2, SCALE and Serpent 2.2.0
  - Calculations normalized to <sup>148</sup>Nd inventory
- Nuclear data libraries:
  - ENDF/B-VII.0, ENDF/B-VII.1 and ENDF/B-VIII.0
  - JEFF-3.1.2, JEFF-3.3, JEFF-4T1 and JEFF-4T2
  - JENDL-4.0u and JENDL-5
  - Recommended SFY from Santi and Miller (2008)\* and DD from DDEP

\*Source: P. Santi and M. Miller. Reevaluation of prompt neutron emission multiplicity distributions for spontaneous fission. Nucl. Sci. Eng. 160 (2008) 190–199

## <sup>137</sup>Cs, <sup>244</sup>Cm and BU

Code	Library	<sup>137</sup> Cs	<sup>137</sup> Cs	<sup>244</sup> Cm	BU	<sup>137</sup> Cs	<sup>244</sup> Cm	BU
		$N_{\rm X}/N_{\rm U} \times 10^{-3}$	C/E	$N_{\rm X}/N_{\rm U} \ge 10^{-5}$	MWd/kg			
ALEPH2	JEFF-3.3	2.225	0.982	6.290	53.25	0.990	0.981	0.983
SCALE	ENDF/B-VII.0	2.241	0.990	6.380	54.01	 0.997	0.995	0.997
Serpent2	ENDF/B-VII.0	2.285	1.009	6.633	54.37	1.016	1.035	1.004
	ENDF/B-VII.1	2.274	1.004	6.710	54.39	1.012	1.047	1.004
	ENDF/B-VIII.0	2.274	1.004	6.701	54.38	1.012	1.045	1.004
	JEFF-3.1.2	2.248	0.993	6.110	54.18	1.000	0.953	1.000
	JEFF-3.3	2.225	0.982	6.354	53.37	0.990	0.991	0.985
	JEFF-3.3 (1)	2.290	1.011	7.149	55.24	1.019	1.115	1.020
	JEFF-3.3 (2)	2.249	0.993	6.644	54.21	1.000	1.037	1.001
	JEFF-3.3 (3)	2.246	0.992	6.599	54.12	0.999	1.029	0.999
	JEFF-4T1	2.248	0.993	6.410	54.16	1	1	1
	JENDL-4.0u	2.301	1.016	7.009	55.07	1.024	1.093	1.017
	JENDL-5.0	2.253	0.995	7.194	54.97	1.002	1.122	1.015

(1)  $\sigma(n,\gamma) = 0$  for <sup>147</sup>Nd (2)  $\sigma(n,\gamma)$  for <sup>147</sup>Nd from JENDL-4.0u (3)  $\sigma(n,\gamma)$  for <sup>147</sup>Nd from JEFF-4T1

## <sup>137</sup>Cs, <sup>244</sup>Cm and BU

Code	Library	<sup>137</sup> Cs	<sup>137</sup> Cs	<sup>244</sup> Cm	BU	<sup>137</sup> Cs	<sup>244</sup> Cm	BU
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 $σ(n, γ) = 0 \text{ for } {}^{147}\text{Nd}$   $σ(n, γ) \text{ for } {}^{147}\text{Nd} \text{ from JENDL-4.0u}$   $σ(n, γ) \text{ for } {}^{147}\text{Nd} \text{ from JEFF-4T1}$ 

(1)

(2)

(3)

S<sub>sf</sub>

Code	Library	Ssf/ (1/sg)	Ssf/ (1/sg)	C/E	C/E			
		LIB	REC	LIB	REC	LIB/REC		
ALEPH2	JEFF-3.3	640.1	642.4	0.944	0.947	0.996		
SCALE	ENDF/B-VII.0	653	652.1	0.963	0.962	1.001	-	
Serpent2	ENDF/B-VII.0	683.7	678.1	1.008	1.000	1.008	-	
	ENDF/B-VII.1	689.4	685.3	1.017	1.011	1.006		
	ENDF/B-VIII.0	688.5	684.3	1.015	1.009	1.006		
	JEFF-3.1.2	632.5	623.8	0.933	0.920	1.014		
	JEFF-3.3	656.8	648.9	0.969	0.957	1.012		
	JEFF-3.3 (1)	739.4	730.5	1.091	1.077	1.012	(1)	σ(n,γ) =
	JEFF-3.3 (2)	686.8	678.6	1.013	1.001	1.012	(2)	σ(n,γ) fo
	JEFF-3.3 (3)	682.2	673.9	1.006	0.994	1.012	(3)	σ(n,γ) fo
	JEFF-4T1	662.6	654.6	0.977	0.965	1.012		
	JEFF-4T2	676.4	668.3	0.998	0.986	1.012		
	JENDL-4.0u (4)		715.9		1.056	0.000	(4)	No data
	JENDL-5.0	738.4	733.9	1.089	1.082	1.006		

 $S_{sf} = 678 (12) s^{-1} g^{-1}$ 

 $σ(n,γ) = 0 \text{ for } {}^{147}\text{Nd}$   $σ(n,γ) \text{ for } {}^{147}\text{Nd} \text{ from JENDL-4.0u}$  $σ(n,γ) \text{ for } {}^{147}\text{Nd} \text{ from JEFF-4T1}$ 

) No data available to calculate S<sub>sf</sub>

S<sub>sf</sub>

-							_
Code	Library	Ssf/ (1/sg)	Ssf/ (1/sg)	C/E	C/E		
		LIB	REC	LIB	REC	LIB/REC	
ALEPH2	JEFF-3.3	640.1	642.4	0.944	0.947	0.996	
SCALE	ENDF/B-VII.0	653	652.1	0.963	0.962	1.001	
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 $σ(n,γ) = 0 \text{ for } {}^{147}\text{Nd}$   $σ(n,γ) \text{ for } {}^{147}\text{Nd} \text{ from JENDL-4.0u}$  $σ(n,γ) \text{ for } {}^{147}\text{Nd} \text{ from JEFF-4T1}$ 

No data available to calculate S<sub>sf</sub>

S<sub>sf</sub>

Code	Library	Ssf/ (1/sg)	Ssf/ (1/sg)	C/E	C/E			<b>C</b> -
		LIB	REC	LIB	REC	LIB/REC		Ca
ALEPH2	JEFF-3.3	640.1	642.4	0.944	0.947	0.996		۵ <sub>sf</sub>
SCALE	ENDF/B-VII.0	653	652.1	0.963	0.962	1.001		
Serpent2	ENDF/B-VII.0	683.7	678.1	1.008	1.000	1.008		
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	JEFF-3.3	656.8	648.9	0.969	0.957	1.012		
	JEFF-3.3 (1)	739.4	730.5	1.091	1.077	1.012	(1)	σ(n,
	JEFF-3.3 (2)	686.8	678.6	1.013	1.001	1.012	(2)	σ(n,
	JEFF-3.3 (3)	682.2	673.9	1.006	0.994	1.012	(3)	σ(n,
	JEFF-4T1	662.6	654.6	0.977	0.965	1.012		
	JEFF-4T2	676.4	668.3	0.998	0.986	1.012		
	JENDL-4.0u (4)		715.9		1.056	0.000	(4)	No
	JENDL-5.0	738.4	733.9	1.089	1.082	1.006		

Source: P. Schillebeeckx et al. Task 2.2 Neutron measurements on REGAL sample. EURAD Task 2 Workshop, KIT, Karlsruhe (Germany), May 10-12,2023

## $S_{sf} = 678 (12) s^{-1} g^{-1}$

Calculated from RCA with REC:  $S_{sf} = 699 (28) s^{-1} g^{-1}$ 

 $σ(n, γ) = 0 \text{ for } {}^{147}\text{Nd}$   $σ(n, γ) \text{ for } {}^{147}\text{Nd} \text{ from JENDL-4.0u}$   $σ(n, γ) \text{ for } {}^{147}\text{Nd} \text{ from JEFF-4T1}$ 

4) No data available to calculate S<sub>sf</sub>

## Conclusions

- Recommended decay and neutron emission data not always adopted in evaluated data libraries
- <sup>147</sup>Nd(n,γ) cross section in JEFF-3.3 and ENDF/B-VIII.0 are too high (important for normalisation of PIE data)
- Fission yields for <sup>148</sup>Nd in JENDL-5.0 are too low
- <sup>242</sup>Pu(n,γ) and <sup>243</sup>Am(n,γ) cross sections require a re-evaluation (use of available experimental data)

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