

Task 2: Fuel properties characterisation and related uncertainty analysis

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Marc Verwerft	SCK•CEN

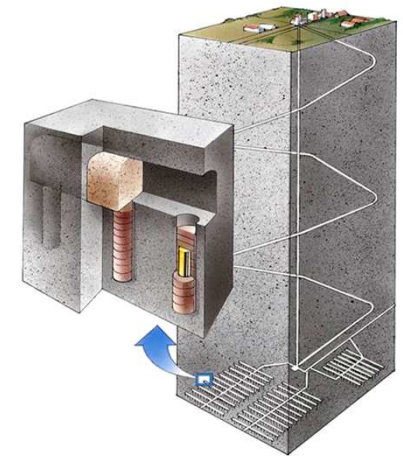
Partners:

CIEMAT, CPST, CTU (SURAO), ENRESA, ENUSA, JRC, JSI, KIT,
LEI, NAGRA, PEL, PSI, SCK•CEN, SKB, SSTC NRS, TUS, VTT, UU

Spent Nuclear Fuel (SNF) intermediate storage or final disposal

A **safe, secure, economic** and **ecological** transport, storage and final disposal requires that **SNF is characterised** for the main source terms of interest:

- Decay heat
- Neutron emission
- γ -ray emission
- Reactivity (burnup credit)
nuclides with high neutron absorption cross section)
- Fissile material (Safeguards)
i.e. ^{235}U , ^{239}Pu
- Specific long-lived radionuclides (Long term safety)
e.g. ^{14}C , ^{79}Se , ^{94}Nb , ^{99}Tc , ^{129}I , ^{226}Ra



Neutron emission by SNF

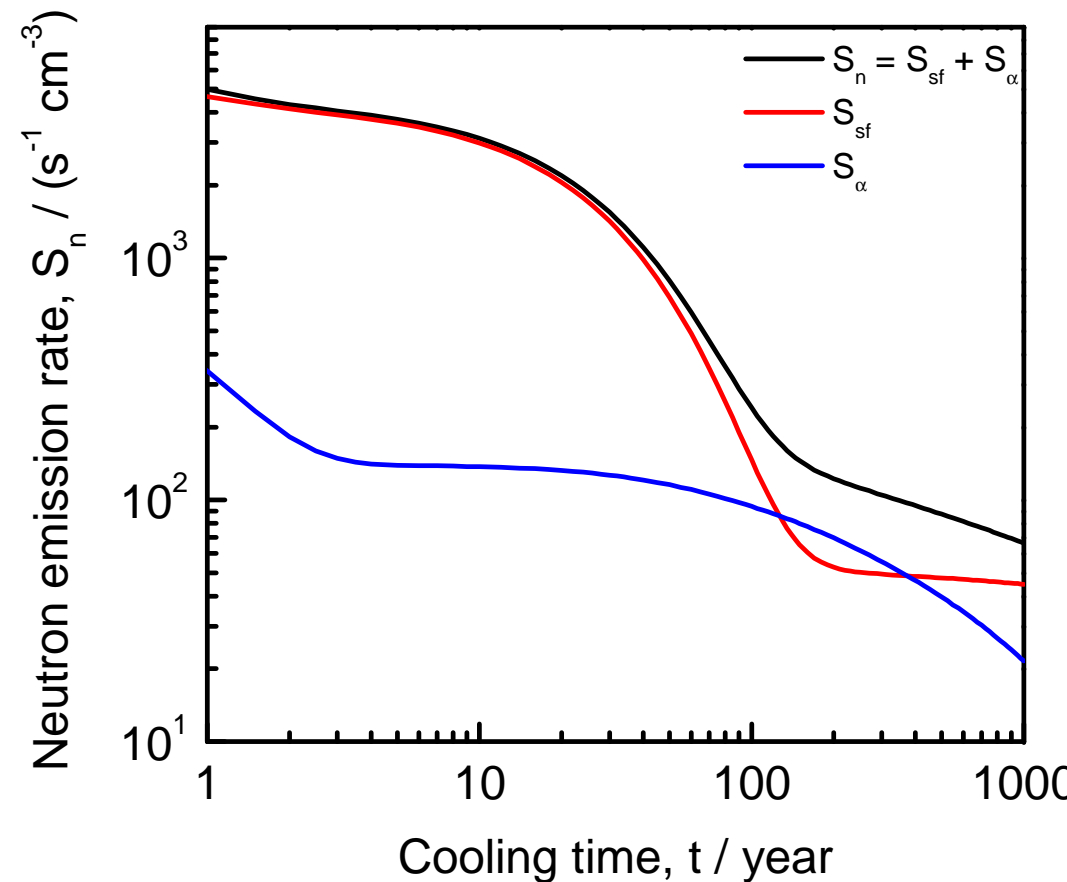
$$S_n(t) = \sum_k S_{n,k}(t)$$

- $S_{n,k}(t)$: contribution of radionuclide k
- $S_{n,k}(t) = (s_{sf,k} + s_{\alpha n,k}) N_k(t)$
 - $N_k(t)$: number of nuclei of nuclide k at time t
 - $s_{sf,k}$: specific neutron emission rate of nuclide k due to sf
 - $s_{\alpha,k}$: specific neutron emission rate of nuclide k due to (α,n) reactions

PWR UO₂ pellet (5 g)

²³⁵U/U = 4.8 %

burnup = 44 GWd/t



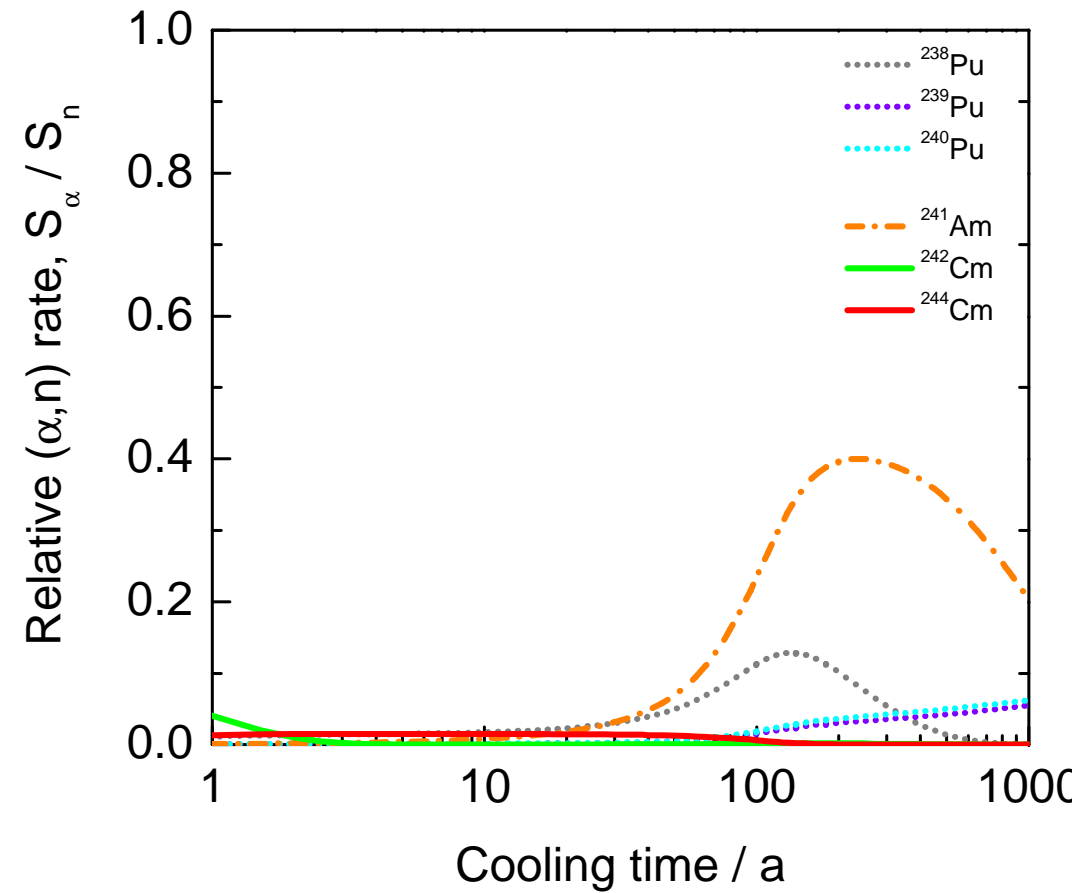
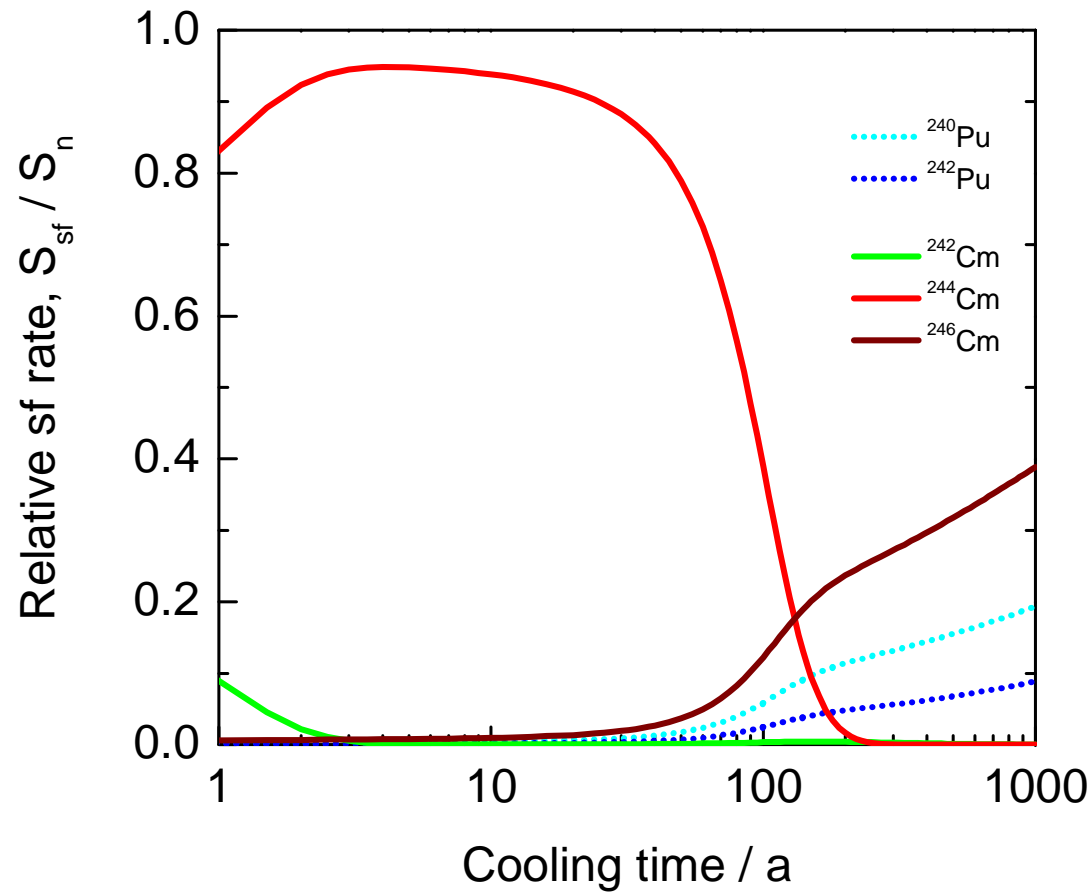
Neutron emission by SNF

$$S_n(t) = \sum_k (s_{sf,k} + s_{\alpha n,k}) N_k(t)$$

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Gamma-ray emission by SNF

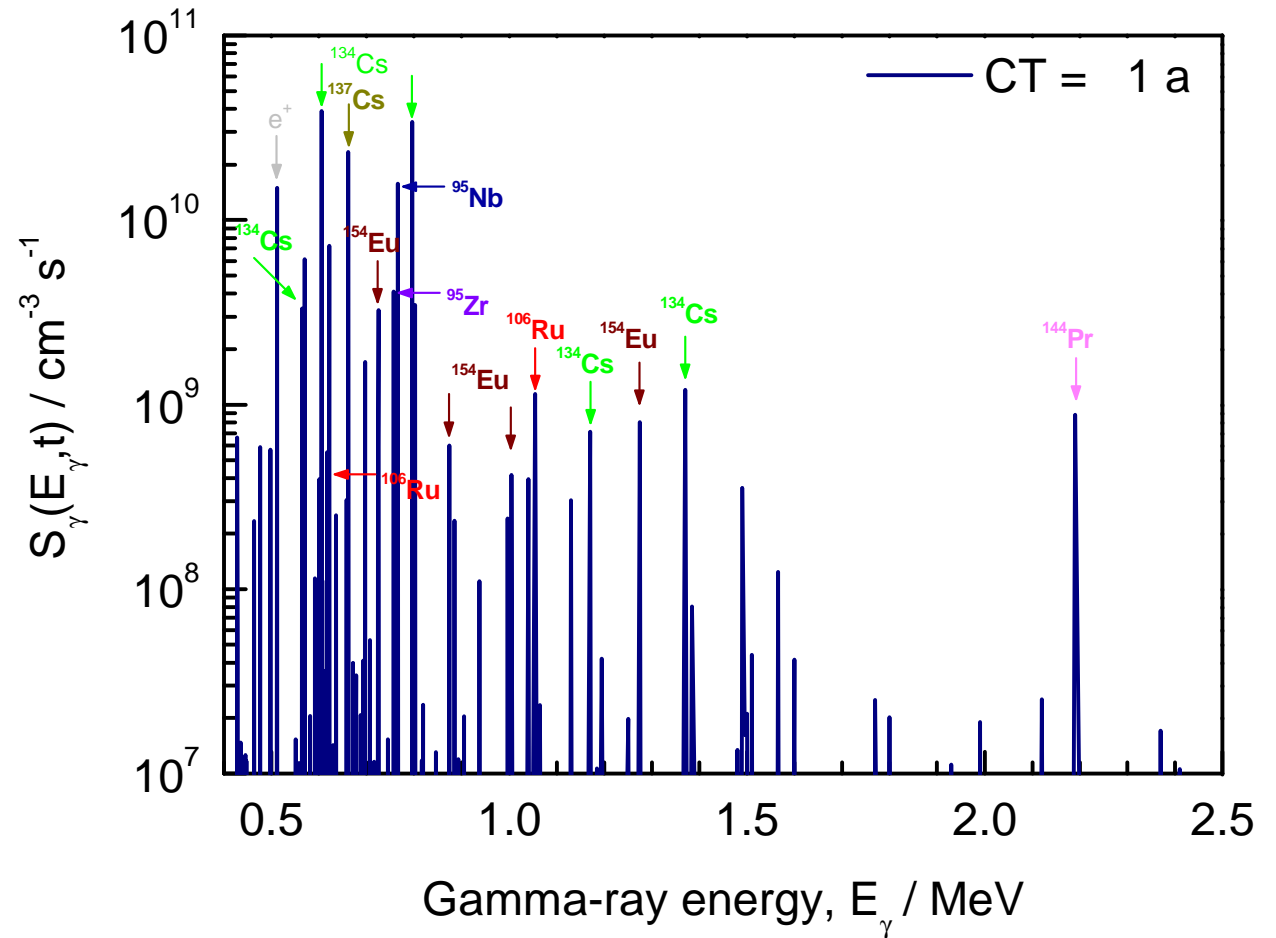
$$S_{\gamma}(t) = \sum_k S_{\gamma,k}(E_{\gamma}, t)$$

⁹⁵ Nb	35.0 d
⁹⁵ Zr	64.0 d
¹⁴⁴ Ce/ ¹⁴⁴ Pr	284.9 d
¹⁰⁶ Ru/ ¹⁰⁶ Rh	1.02 a
¹³⁴ Cs	2.06 a
¹⁵⁴ Eu	8.8 a
¹³⁷ Cs/ ^{137m} Ba	30.0 a

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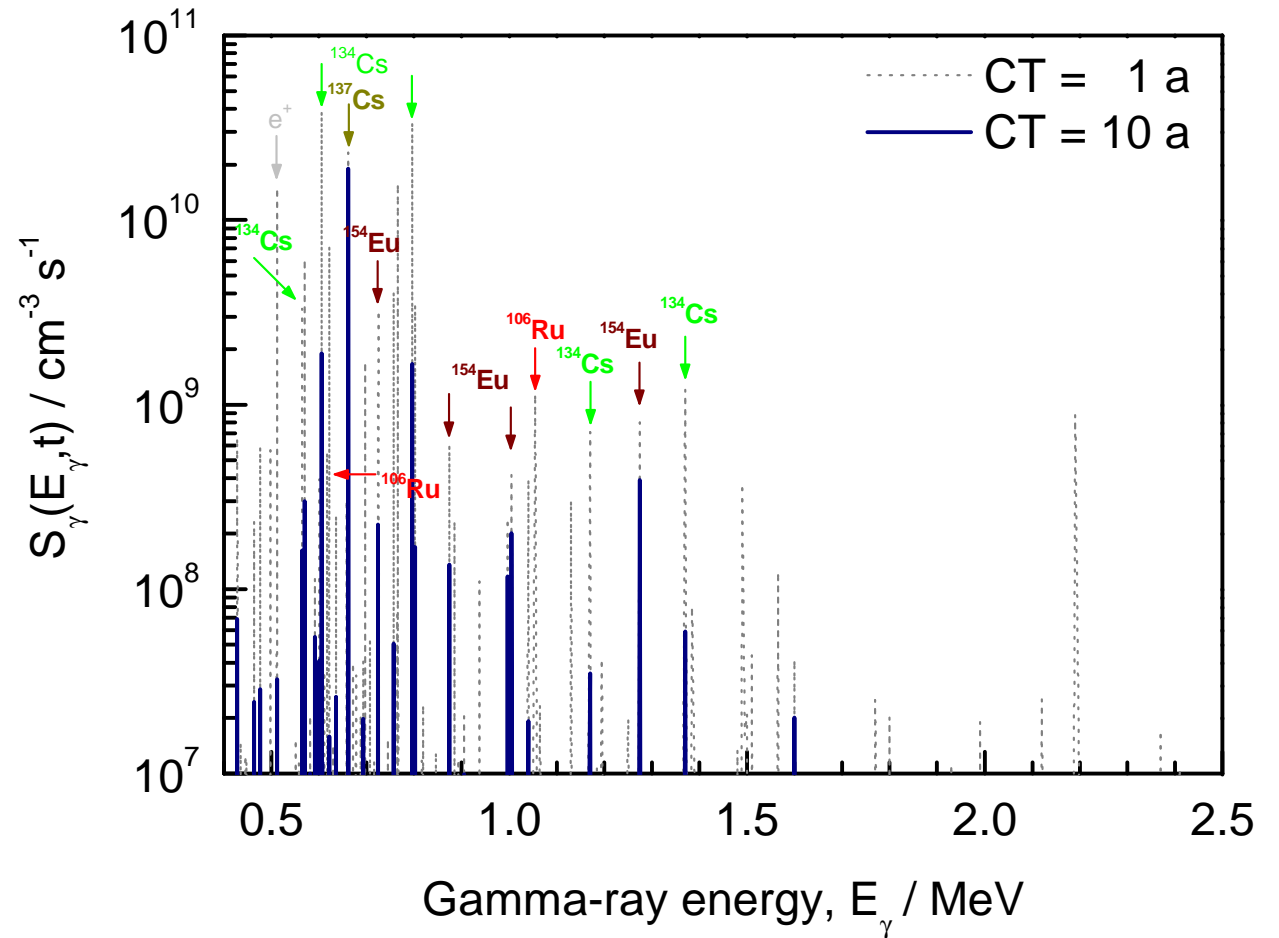
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^{154}Eu	8.8 a
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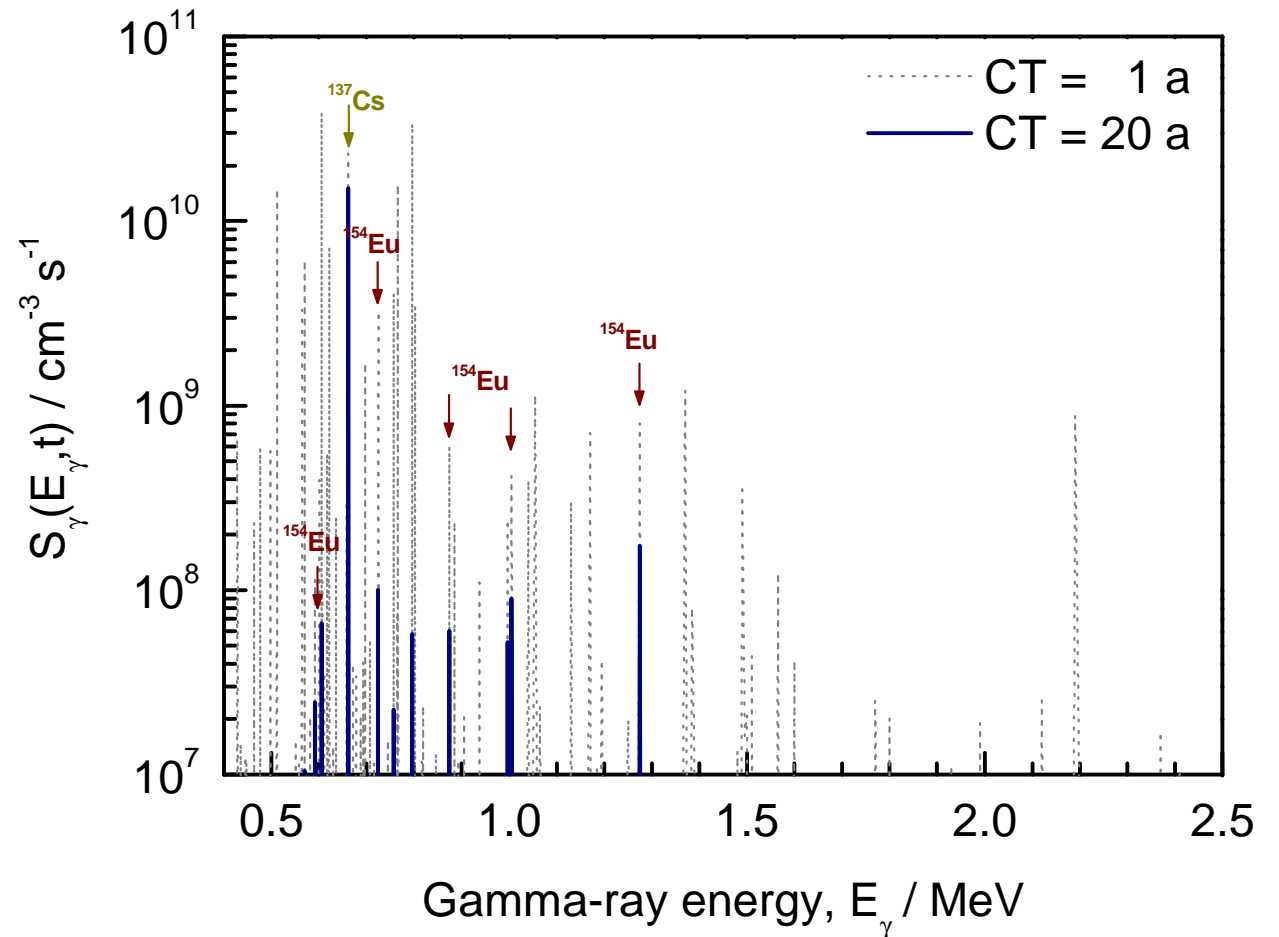
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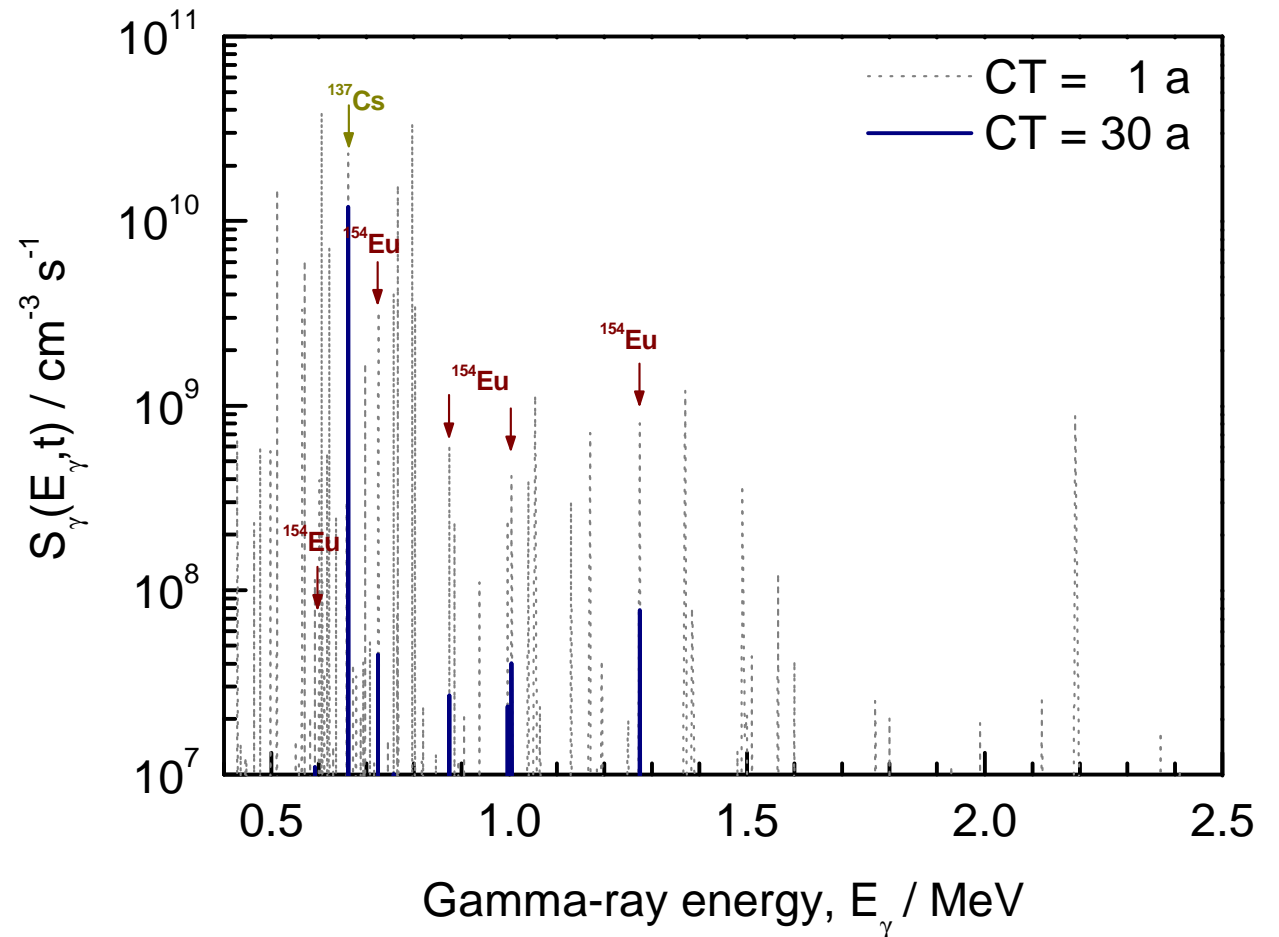
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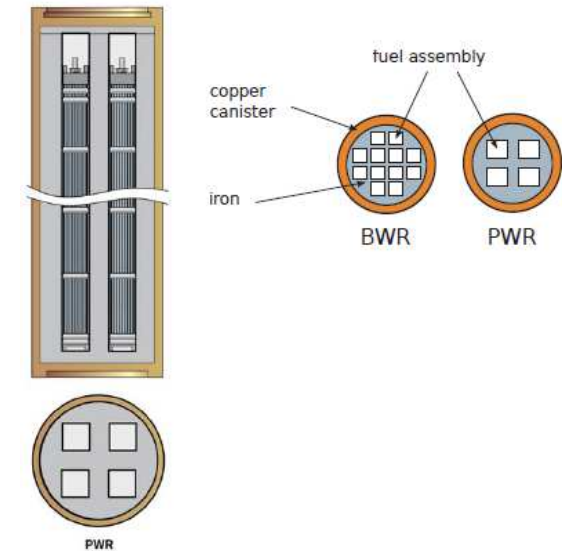
burnup = 44 GWd/t



Characterisation of SNF

Main source terms of interest:

- Decay heat : H
- Neutron emission : S_n
- γ -ray emission : S_γ
- Reactivity : ^{235}U , ^{239}Pu , ^{241}Am , Fission Products (BUC)
- Fissile material : ^{235}U , ^{239}Pu
- Long-term safety : e.g. ^{14}C , ^{79}Se , ^{94}Nb , ^{99}Tc , ^{129}I , ^{226}Ra



Contributions of nuclides with different characteristics

Difficult to be measured directly, in particular during industrial operation

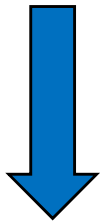
e.g. decay heat by calorimetry at CLAB: accurate but long measurement times

⇒ Estimated by **theoretical calculations** using a burnup code

$(N_k(t_0), k = 1, \dots, n)$: by theoretical calculations

Coupled neutron transport – nuclide depletion/creation calculation

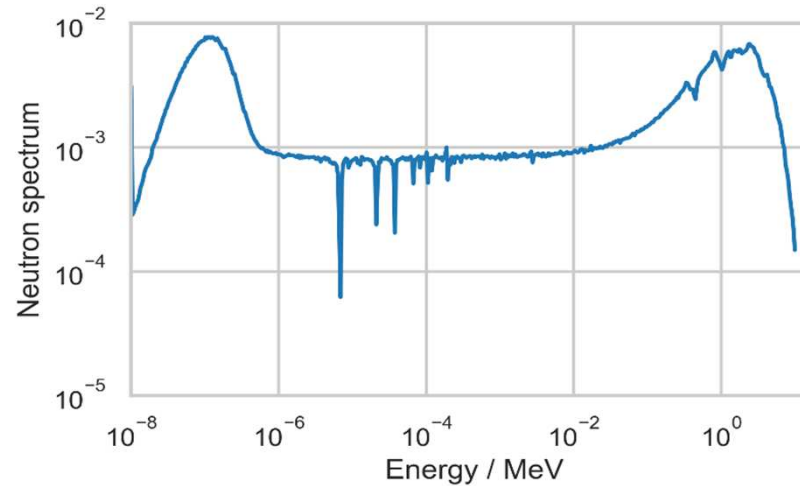
Neutron transport



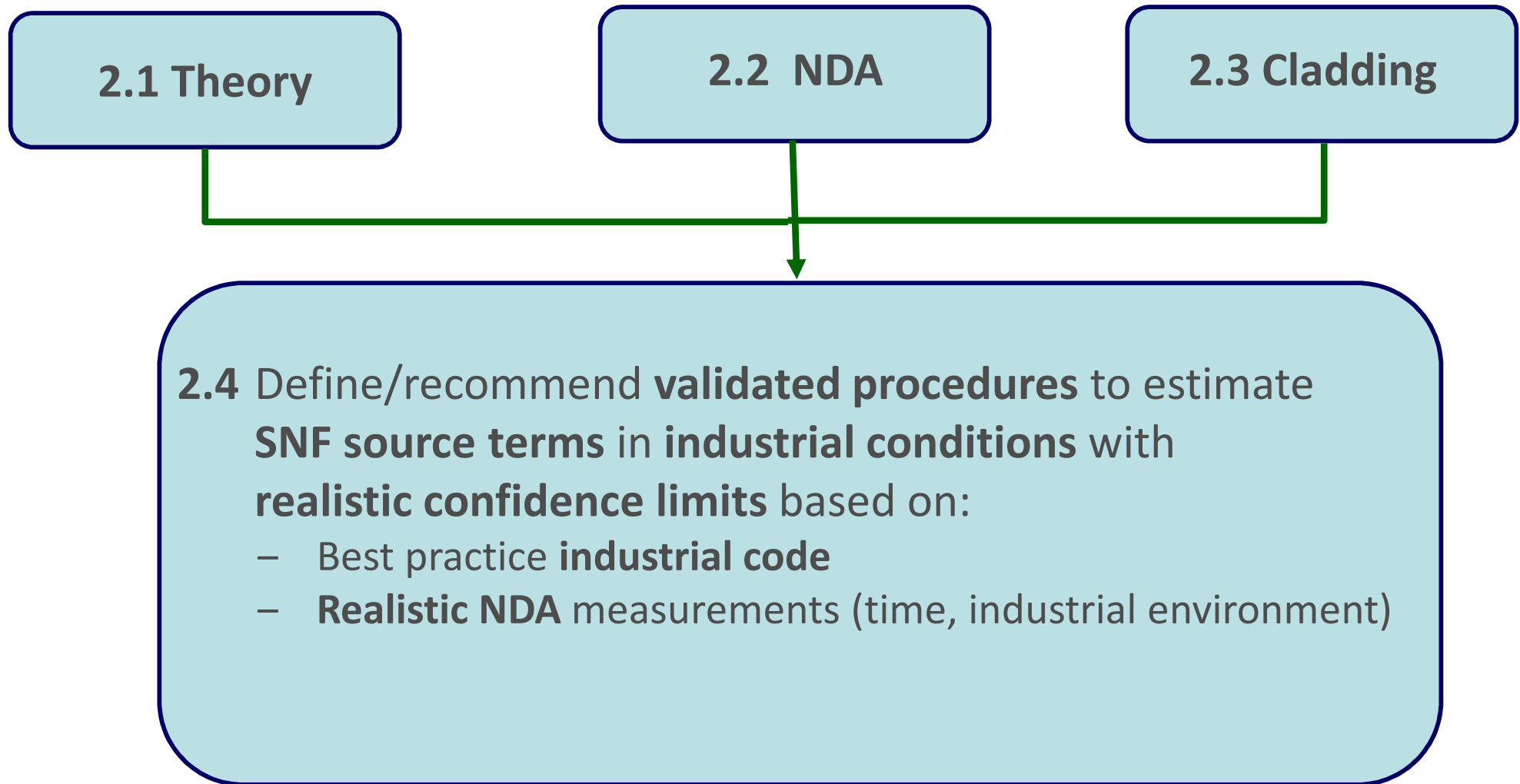
Bateman equation

$$\frac{dN_k}{dt} = Y N_f \sigma_f \varphi + \sum_i \lambda_i N_i + \sum_j \sigma_j N_j \varphi - (\lambda_k + \sigma_{k,a} \varphi) N_k$$

Update nuclide vector



SFC – Task 2 (structure)



SFC- Task 2: Code comparison

- CIEMAT : EVOLCODE (MCNP)
- JRC Geel : SERPENT, SCALE
- JSI : SCALE, DRAGON
- KIT : MCNP-CINDER
- NAGRA : SCALE
- PSI : CASMO
- SCK CEN : ALEPH-2 (MCNP)
- VTT : SERPENT

Sensitivity and Uncertainty (S/U) analysis

- Input data:

- **Nuclear Data (ND)**

- Cross sections (neutron interactions)
- Fission yields
- Neutron emission probabilities
- Decay data

- **Fuel History (FH)**

- Fuel properties (design, composition)
e.g. Initial enrichment (IE)
- Reactor operation and irradiation conditions
e.g. Burnup (BU)
- Cooling time (CT)

BurnUp (BU):

time integrated power per mass of initial fuel (MWd/kg)

∝ total number of fission x energy per fission event

- Computational

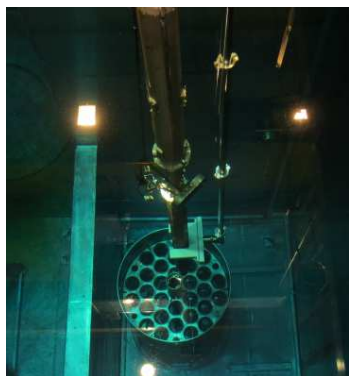
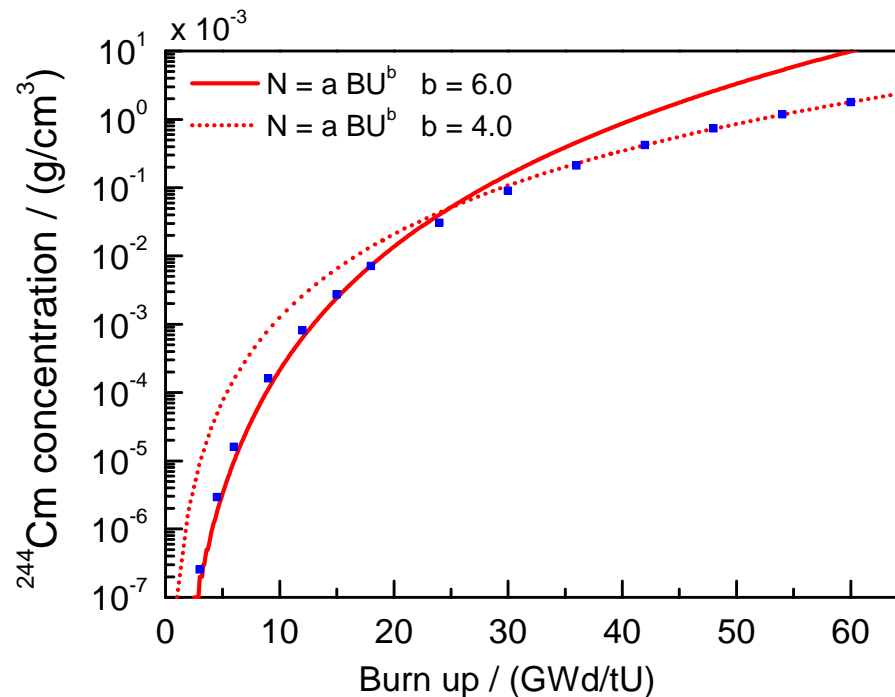
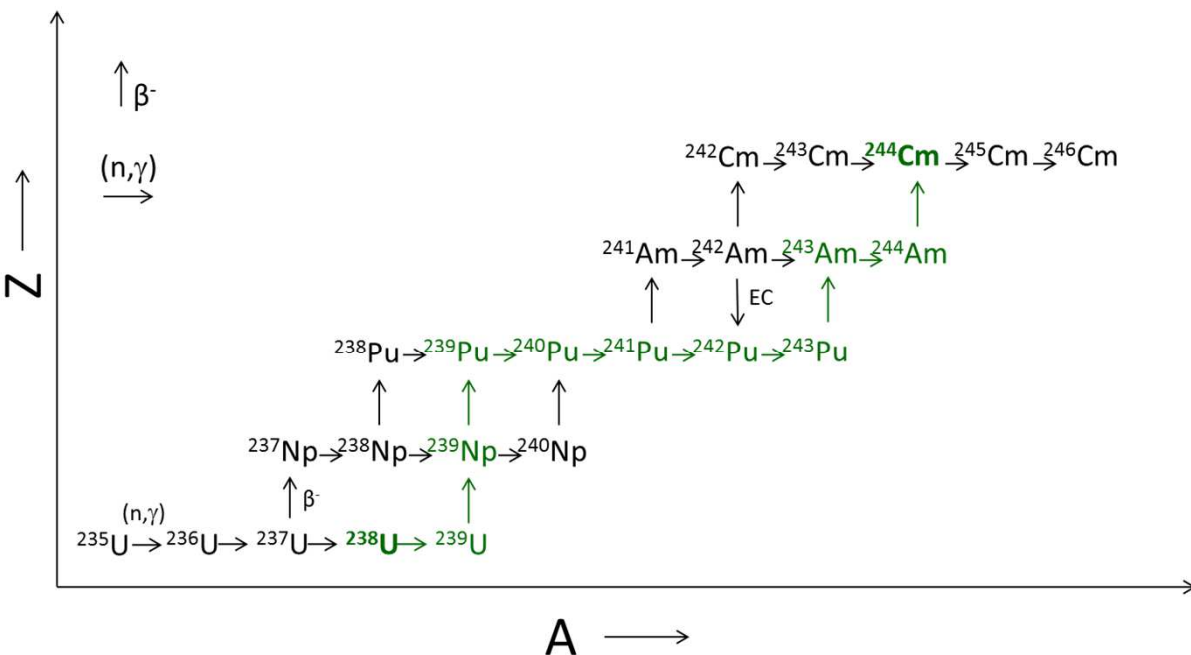
- Method: stochastic/deterministic
- Model (2D/3D, boundary conditions, ...)
- Numerical approximations (depletion time steps, depletion zones, ...)

S/U analysis: ^{244}Cm production

$$\frac{dN_k}{dt} = Y N_f \sigma_f \varphi + \sum_i \lambda_i N_i + \sum_j \sigma_j N_j \varphi - (\lambda_k + \sigma_{k,a} \varphi) N_k$$

$$N = aBU^b$$

$$b = 6 \text{ for } BU < 20 \text{ GWd/t}$$



⇒ ^{244}Cm : burnup indicator

⇒ Neutron emission rate : burnup indicator

S/U analysis: estimation of ^{244}Cm inventory

$$\frac{dN_k}{dt} = Y N_f \sigma_f \varphi + \sum_i \lambda_i N_i + \sum_j \sigma_j N_j \varphi - (\lambda_k + \sigma_{k,a} \varphi) N_k$$

Ref.	Library	Reactor	Fuel	BU	IE	^{244}Cm
				GWd/t	wt%	
Rochman	ENDF/B-VII.0	PWR	UO ₂	10	4.1	18.7 %
	ENDF/B-VII.0	PWR	UO ₂	20	4.1	16.9 %
	ENDF/B-VII.0	PWR	UO ₂	30	4.1	15.5 %
	ENDF/B-VII.0	PWR	UO ₂	40	4.1	14.1 %
Zwermann	SCALE-6.1	PWR	UO ₂	40	4.1	8.5 %
Leary	ENDF/B-VII.1	PWR	UO ₂	54	3.4	9.6 %
Rochman	ENDF/B-VII.1	PWR	UO ₂	54	3.4	9.1 %
Rochman	ENDF/B-VII.1	PWR	UO ₂	40	4.1	9.7 %

^{244}Cm inventory prediction production

- uncertainty due to nuclear data : ~10%
- due to $^{242}\text{Pu}(n,\gamma)$, $^{243}\text{Am}(n,\gamma)$

⇒ Systematic study for key nuclides

⇒ Improve nuclear data

- Input HPRL NEA/OECD

- Input to SANDA (DG-RTD)

<https://cordis.europa.eu/project/id/847552>
Supplying Accurate Nuclear Data for energy and non-energy Applications

Innovative NDA methods/systems for SNF characterisation

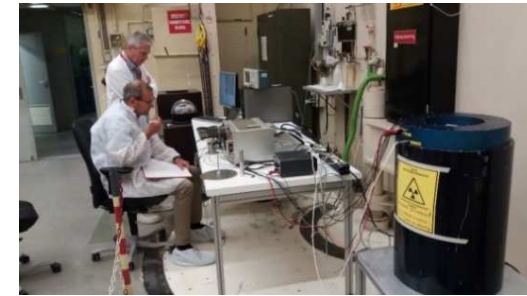
- **Innovative NDA** methods to characterise **pin segments**
 - **Validate codes** (alternative to radiochemical analysis)
 - Production of a reference pellet
- **NDA** methods to characterise **fuel assemblies (SKB-50)**
 - Improve theoretical source term predictions during industrial routine operation
Improve fuel history data: e.g. BU
 - Validate codes
- **Study new detectors**
 - CLYC, *CVD*

NDA methods to characterise pin segments

- **Neutron emission rate of a SNF pin segment**

(collaboration SCK CEN – JRC Geel, Ispra)

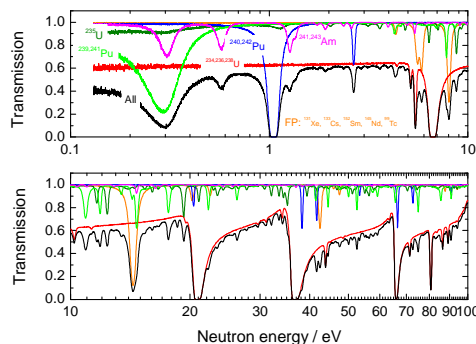
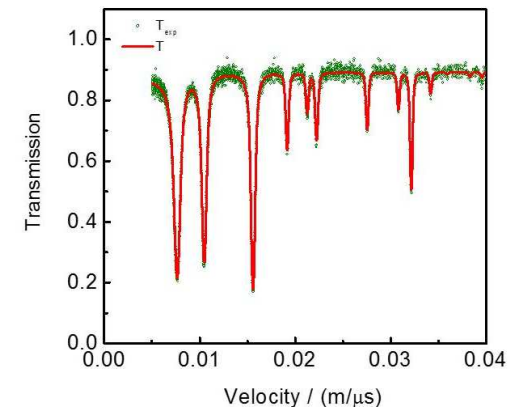
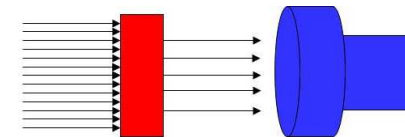
- Non-destructive method to determine ^{244}Cm content
- Measurements in conventional controlled area conditions
Hage's point model, JRC Ispra (B. Pedersen)



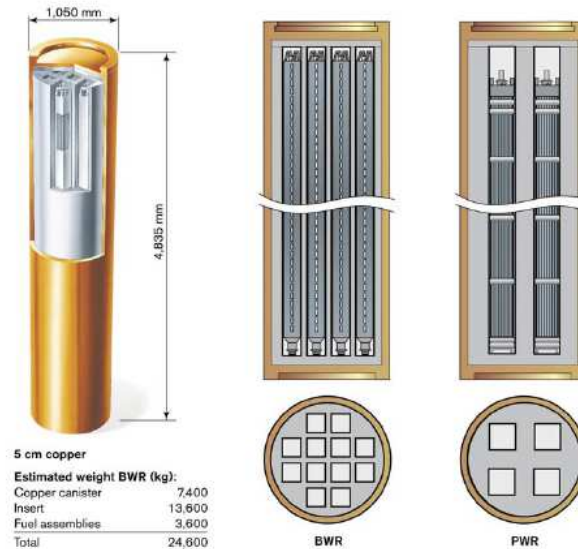
- **Nuclide vector of SNF pin segment by NRTA**

(collaboration SCK CEN – JRC Geel)

- **Non-destructive**; no chemical analysis
- **Absolute** measurement (no calibration)
- Measurements at **GELINA** facility of JRC Geel



Experiments at CLAB: SKB-50



Installed systems

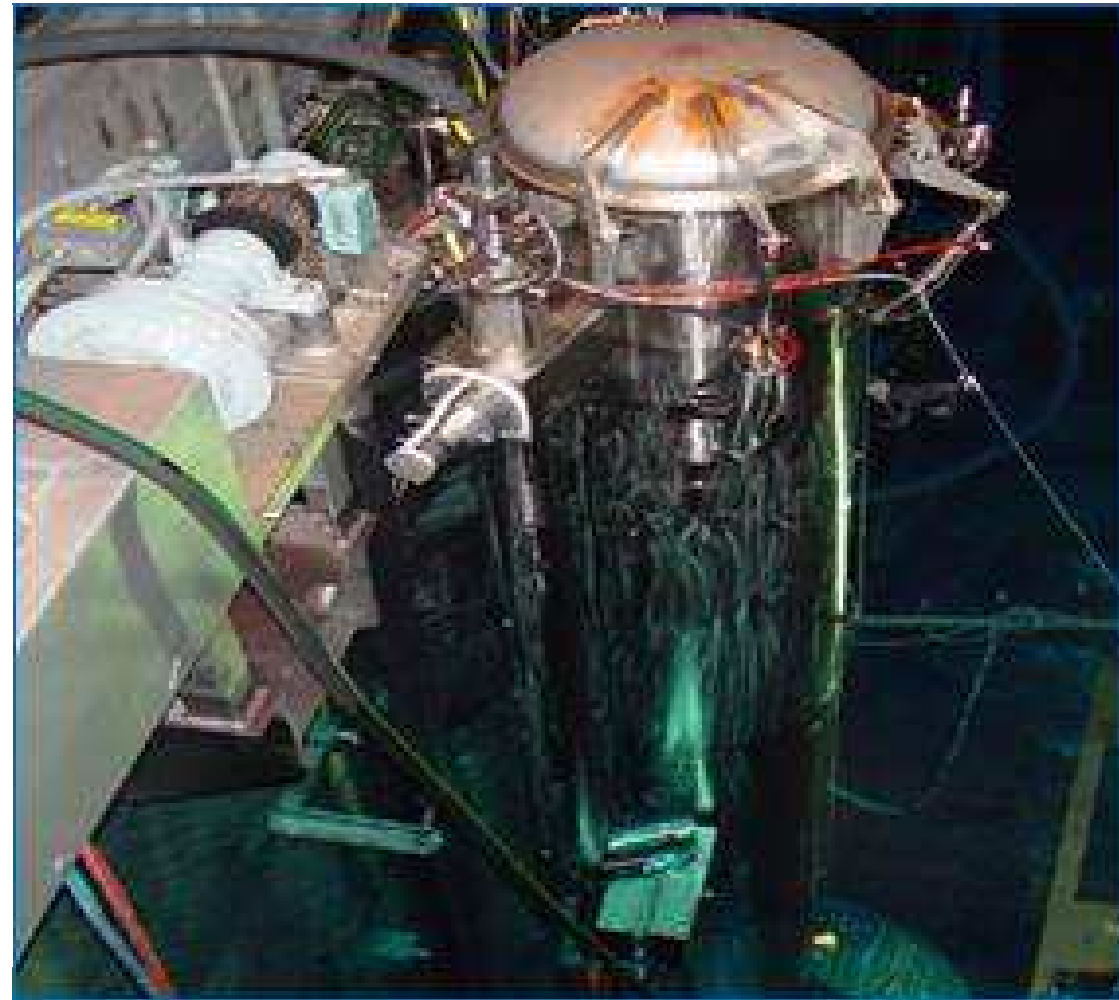
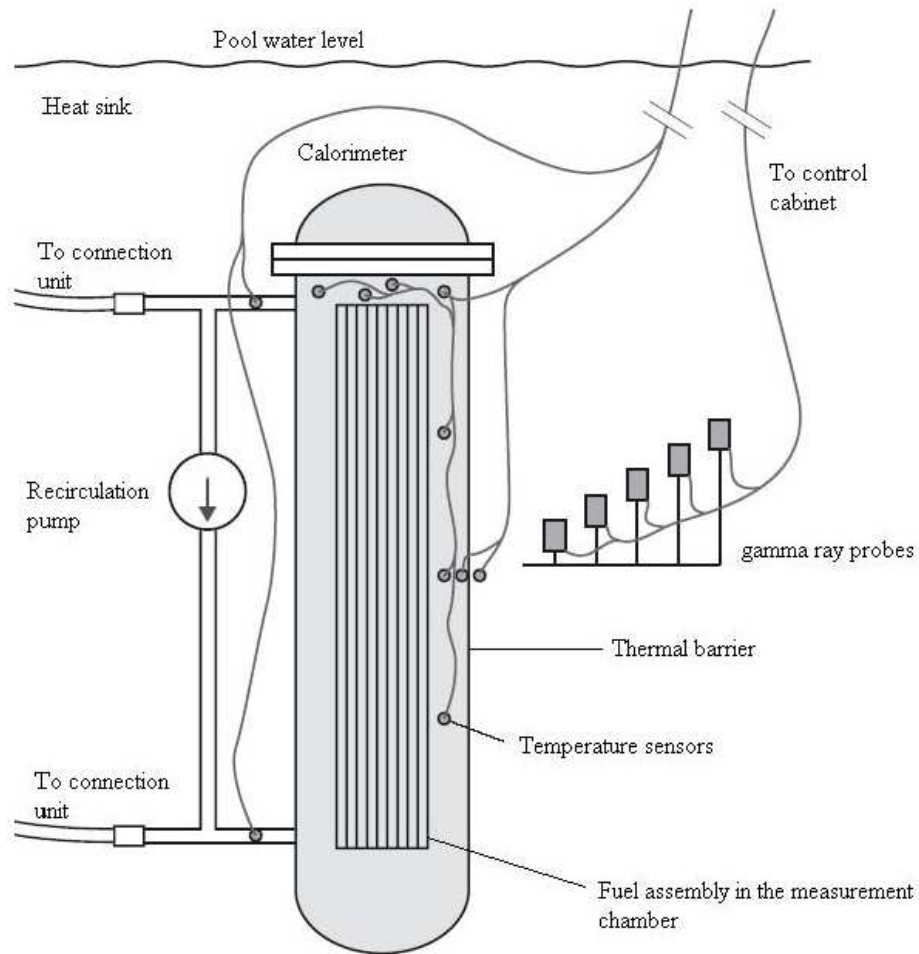
- Calorimeter
- Gamma-ray spectroscopic scanner

Testing of advanced systems (LANL)

- Differential Die-Away Self-Interrogation (DDSI)
- Differential Die-Away (DDA)



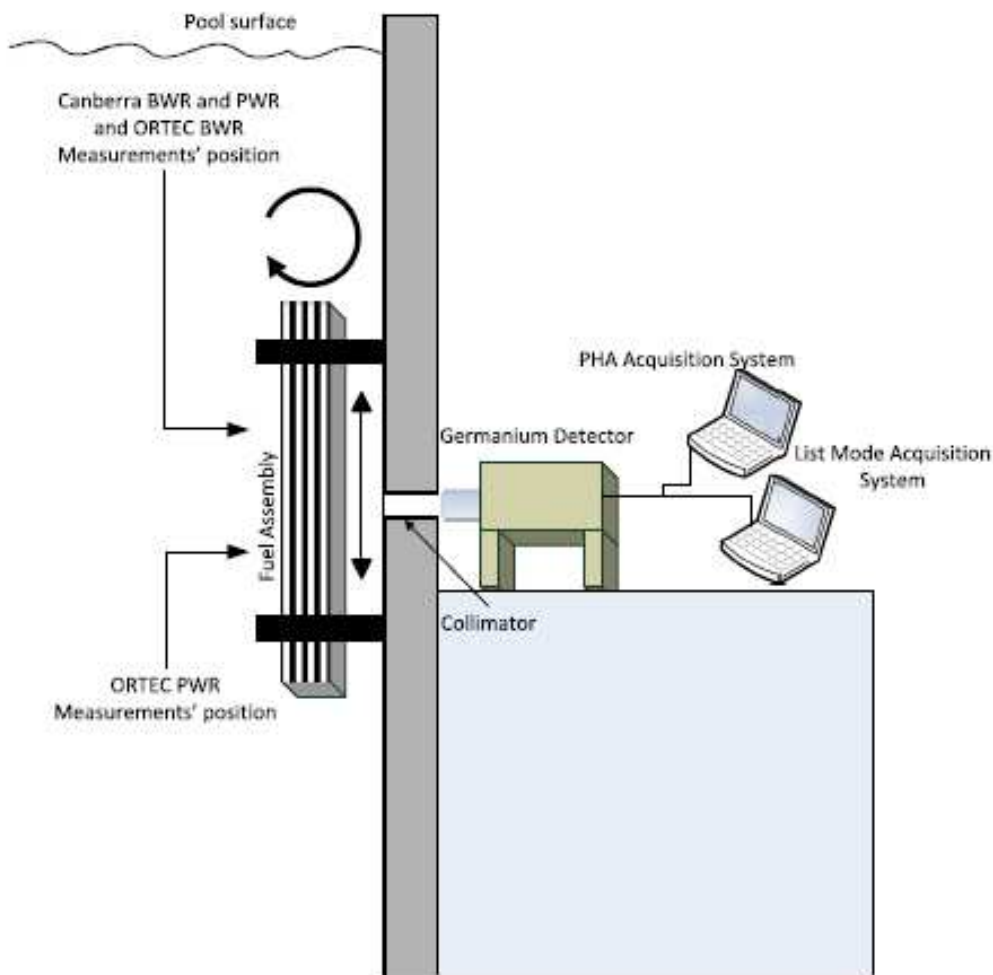
Calorimeter at CLAB



⇒ Target value uncertainty $\leq 2\%$

⇒ Reference instrument for decay heat of SNF

Gamma-ray spectroscopic scanning system at CLAB



Vaccaro et al. NIMA 830 (2016) 325

^{134}Cs , ^{137}Cs , ^{154}Eu

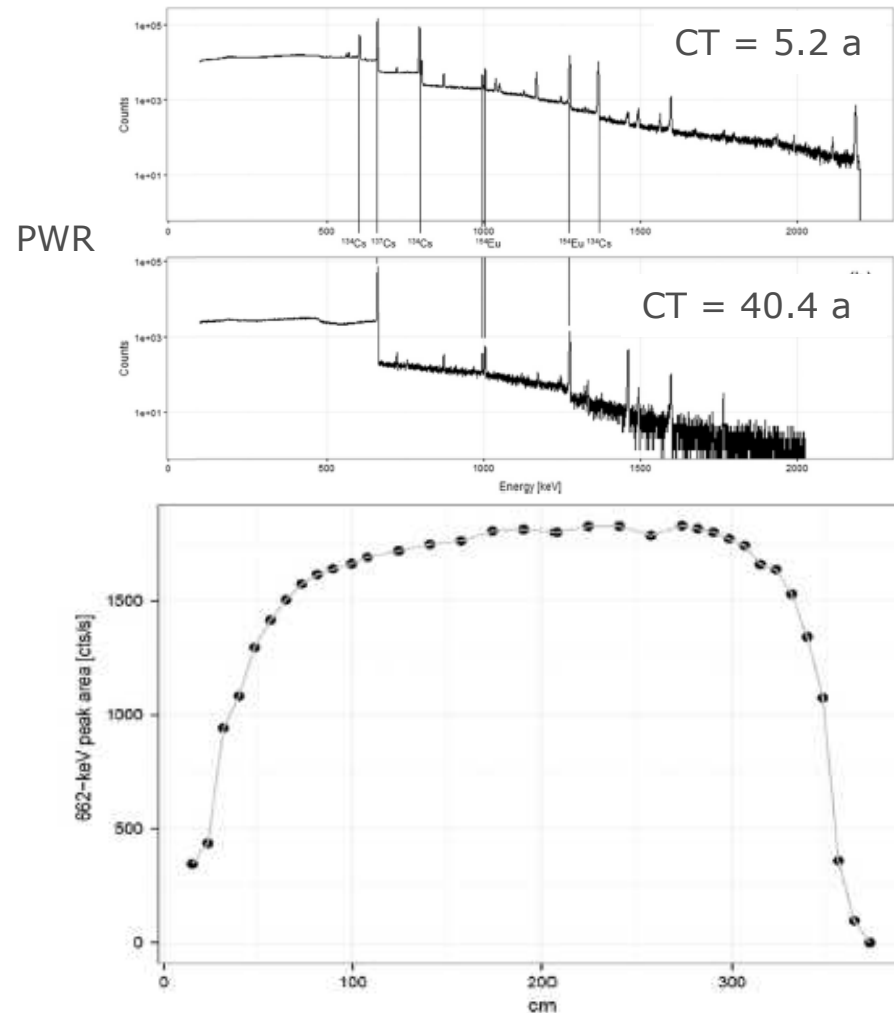
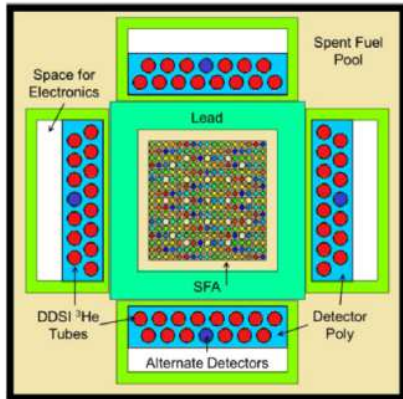


Fig. 8. Count rate for the 662-keV ^{137}Cs net peak area as a function of axial location along BWR9. These data were measured with the ORTEC GMX detector and Canberra Lynx MCA from the 45° corner. The axial location is specified as downward along the uranium containing portion of the fuel (The indicated absolute positions are accurate to ± 10 cm, the error bars are much smaller than the data points).

Fuel assemblies: DDSI and DDA at CLAB (LANL development, NGSi)

- Differential Die Away Self-Interrogation (DDSI, passive)

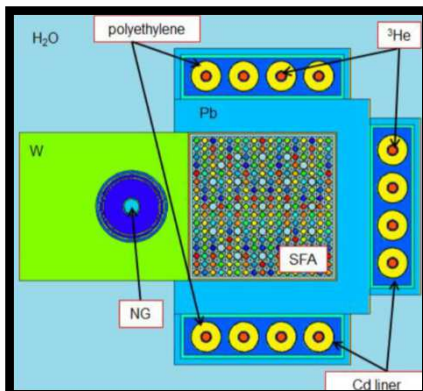
A.C. Trahan, LA-UR_16_20026 Kaplan et al., NIMA 764 (2014) 347 - 351



⇒ Optimise data analysis procedures
for source term determination
(not only safeguards)

- Differential Die Away (DDA, active)

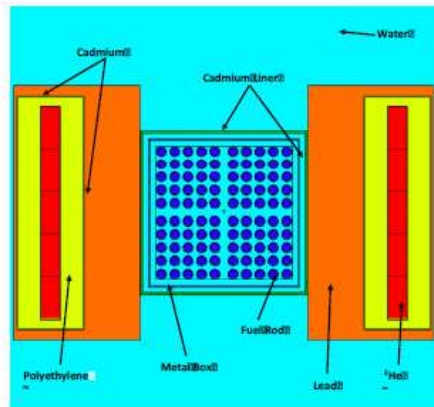
V. Henzl, LANL-UR-123025



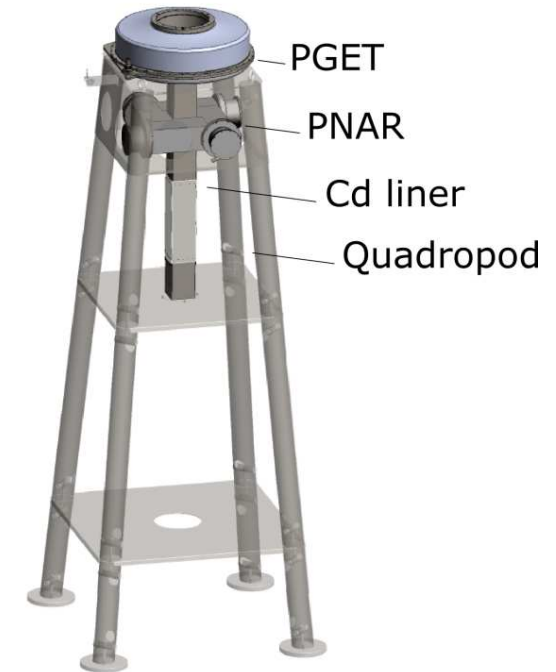
Fuel assemblies: Finland

- **Passive Neutron Albedo Reactivity (PNAR, NGS)**

LANL development, Tobin et al., NIMA 897 (2018) 32 - 37

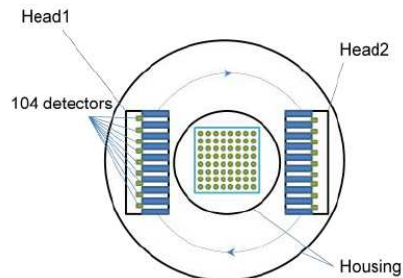


Tobin et al., ESARDA bulletin 56 (2018) 12 - 18



- **Passive Gamma Ray Emission Tomography (PGET)**

IAEA development, Honkamaa et al., Symp. Int. Safeguards, IAEA Vienna 2014



EURAD-SFC (4 years project)

Define/recommend **validated procedures** to estimate **SNF source terms** (including fissile material) in **industrial conditions** with **realistic confidence limits** based on:

- Best practice **industrial code**
- **Realistic NDA** measurements (time, industrial environment)

- EURAD project (SNF characterisation)
 - <https://www.ejp-eurad.eu/about-eurad>
 - NUGENIA, <http://nugenia.org/call-for-mobility-grants-open/>
- ARIEL
support for open access, scientific visits, training early researchers, ...
 - www.ariel-h2020.eu
- JRC Geel open access
 - <https://ec.europa.eu/jrc/en/research-facility/open-access>