STATUS OF THE EURAD RESEARCH PROGRAM ACTIVITIES ON IMPROVING SOURCE TERM PREDICTIONS FOR SPENT NUCLEAR FUEL

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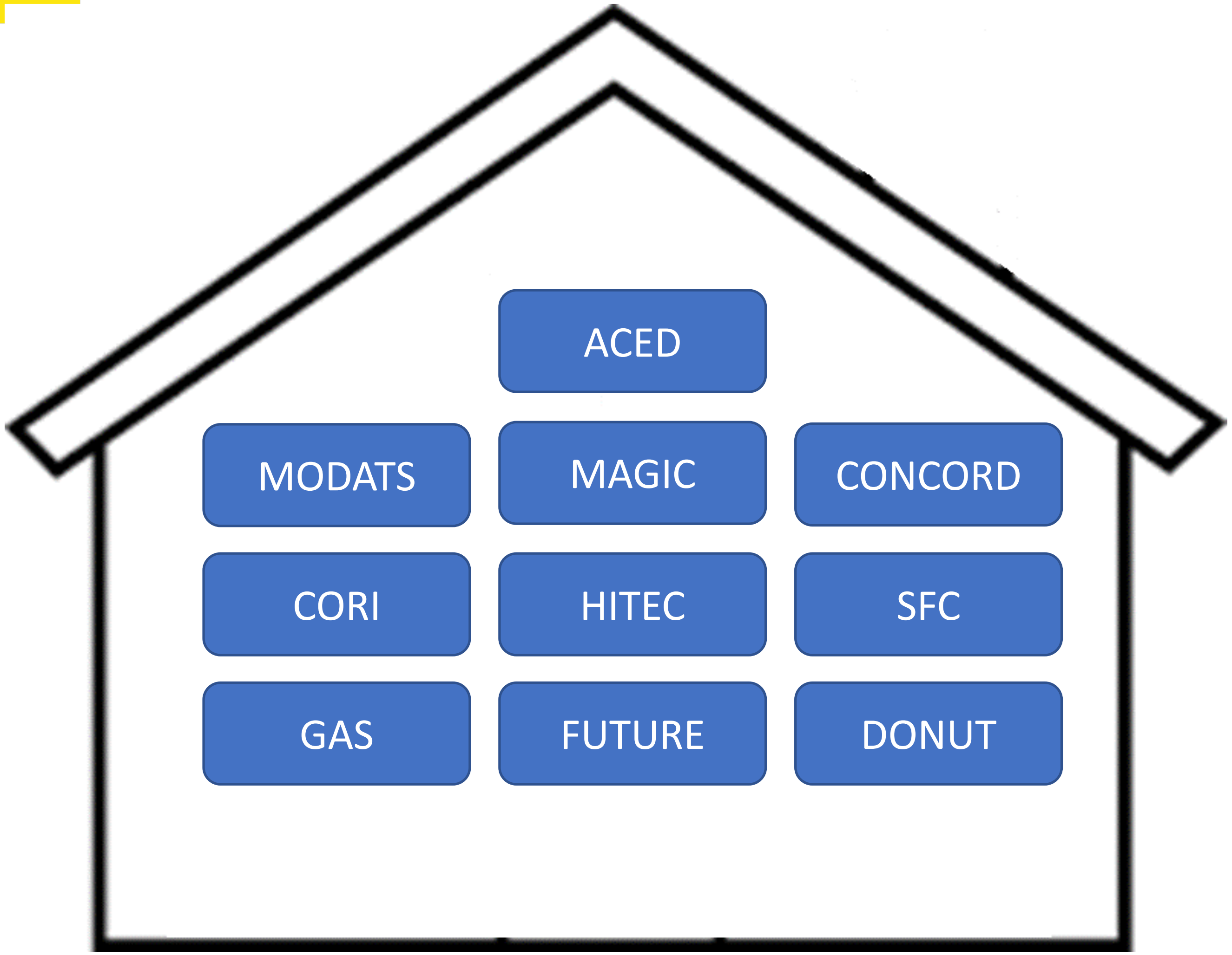
WHAT IS INSIDE THE EURAD R&D HOUSE?



European Joint Program on Radioactive Waste Management

ACED: Assessment of chemical evolution of ILW and HLW disposal cells

MODATS: Monitoring equipment and data treatment for safe repository operation and staged closure

MAGIC: Chemo-mechanical aging of cementitious materials

CONCORD: Container corrosion under disposal conditions

CORI: cement-organic-radionuclide interactions

HITEC: influence of temperature on clay-based material behaviour

SFC: spent fuel characterisation and evolution under disposal

GAS: mechanistic understanding of gas transport in clay materials

FUTURE: fundamental understanding of radionuclide retention

DONUT: development and improvement of numerical methods and tool for modelling couples processes

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THE SPENT FUEL CHARACTERISATION TASK (SFC): 4 ACTIVITIES

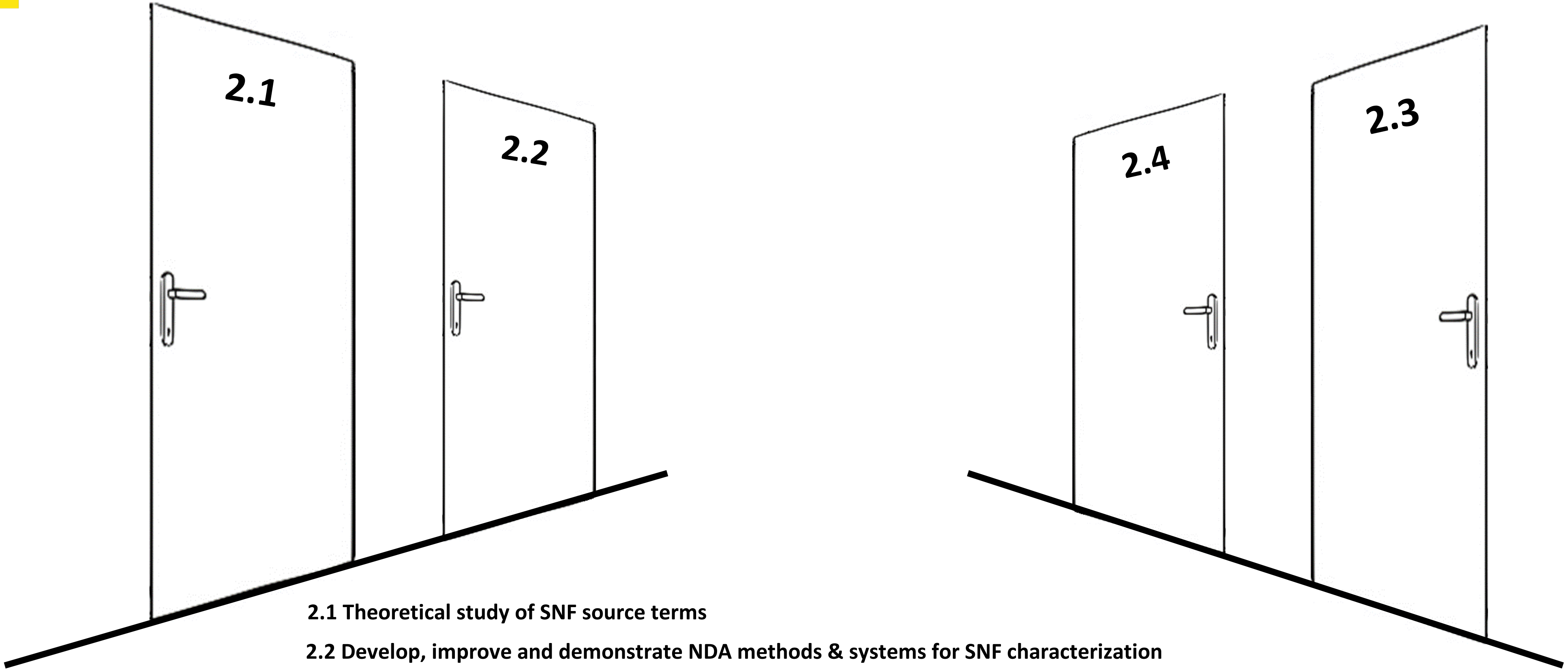
1 State-of-the-art and training activities

2 Fuel properties characterization and related uncertainty analysis

3 Behavior of nuclear fuel and cladding after discharge

4 Accident scenario and consequence analysis

THE SPENT FUEL CHARACTERISATION ACTIVITY 2



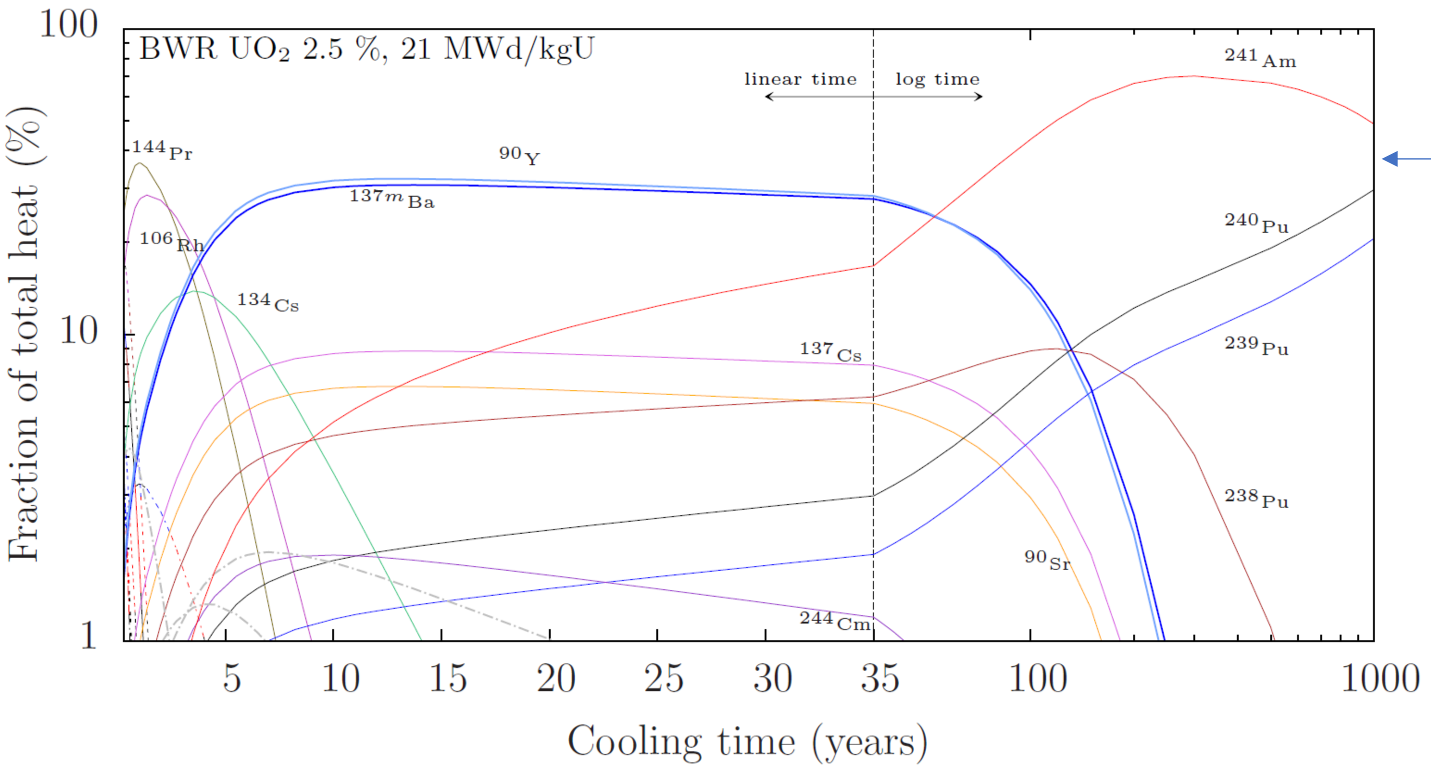
2.1 Theoretical study of SNF source terms

2.2 Develop, improve and demonstrate NDA methods & systems for SNF characterization

2.3 Determine the inventory of activation and fission products in cladding material

2.4 Define and verify procedures to determine the source terms of SNF assemblies with realistic confidence limits

WHICH NUCLIDES CONSTITUTE SPENT FUEL, WHICH ARE RELEVANT?



Relevance depends on safety parameter:

(a) decay heat
(b) neutron source term
(c) gamma source term
(d) other environmental factors

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HOW WELL DO WE KNOW THE NUCLIDE COMPOSITION?

- Nuclide vector at end of irradiation is usually determined with reactor simulation tools like CASMO/SIMULATE, SCALE, et al.

- These “tools” are based on experimental data: cross sections, fission yields, decay constants.

- Two types of validation:

  Integral test: measurement of decay heat, neutron and gamma dose (source terms).
  Single effects test: PIE measurement of nuclide concentrations.

- In the time window which allows observation of source terms codes can be fine-tuned to match observations. This is not possible for the large time frames for long term storage.

ASPECTS OF IRRADIATION HISTORY

- Initial fuel and cladding material composition and impurities comparatively well known.
- Power history: well known for fuel assembly average, less well known on a pellet-basis due to fuel assembly neighbour effects, local effects like fuel assembly and fuel rod bow, heat transfer regime status, fuel & moderator temperature uncertainties, pellet density changes due to cracking, relocation and densification.
- Neutron spectrum: also depends on all the above mentioned parameters and determines the balance of power generation between U and Pu and other actinides.
- Nuclide build-up & destruction by neutron capture and decay in shutdown periods is a non-linear process. Characterizing nuclide vectors as function of burnup only delivers 0-order results.
- Research on neutron capture cross sections, fission yields for higher actinides traditionally focussed on criticality safety and less so on back-end requirements.

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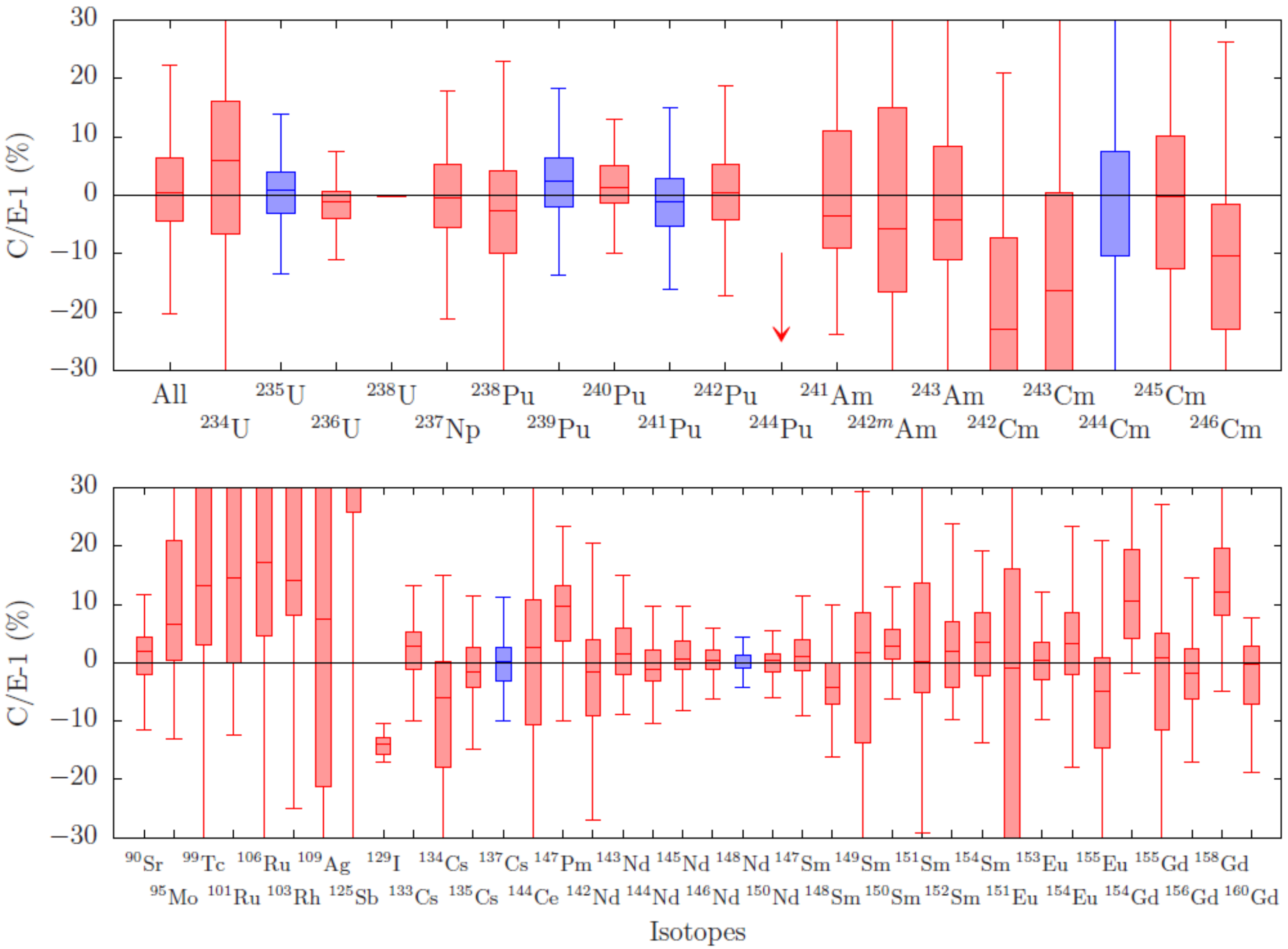
C/E COMPARISON OF MEASURED AND PREDICTED NUCLIDE CONCENTRATIONS



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C/E COMPARISON OF MEASURED AND PREDICTED NUCLIDE CONCENTRATIONS

- Observations: on average C (computed) and E (experimental) values agree roughly. However the spread is in many cases quite large. This means that for regulatory purposes there must be large margins for conservative reasons.

- What is the reason for the large spread?

- If differences are due to uncertainty of the microscopic data all C/E ratios are similarly affected and there would be a systematic bias and a small spread. For some isotopes like Cm242, Cm243, Mo95, Tc99,… this seems to be the case.

- If uncertainties of irradiation histories are important then C/E differences are expected to have stochastic character. However in many cases sensitivity analyses require implausibly large assumptions on reactor parameter variation.

- Uncertainties of radio-nuclide analysis. In many cases claimed to be negligible but questionable on some occasions.

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WHICH CONSEQUENCES DOES THE LARGE C/E SPREAD HAVE?

- Some regulators take the following standpoint:

- Assume X is a random variable representing the C/E value. A series of $N+1$ experiments is conducted and the sample mean m and sample variance s is determined. The task is to estimate how far away m is from the true mean value μ. It is agreed that X is distributed according to a Gaussian distribution with unknown μ and σ.

- Under the stated conditions it is well known that the Student-T distribution of degree N describes the random variable $t = \frac{m-\mu}{s\sqrt{n}}$. This means that for many hypothetical repetitions of the $N+1$ experiments the variable t is distributed like Student-T.

- Another less well known result is that given the first N experimental outcomes and given the sample mean m and sample variance s for the first N results the random variable $\tilde{t} = \frac{X_{n+1}-m}{s\sqrt{1+1/n}}$ is distributed like a Student-T with degree $N-1$.

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WHICH CONSEQUENCES DOES THE LARGE C/E SPREAD HAVE?

- The variable $\tilde{t}$ is a measure for the range in which the next observation of C/E will be located.
- The variable t is a measure for the range in which the true μ will be located.
- When assessing the predictive capability of codes it is natural to choose t because we want to determine if there is any bias in the code. Ideally $\mu = 1$. The confidence interval is $\left[m - c\frac{s}{\sqrt{n}}; m + c\frac{s}{\sqrt{n}}\right]$ (c: degree of confidence).
- Some regulators use $\tilde{t}$ as a proxy to conservatively estimate the range of future C/E values. A "future" C/E value is one from real, to be processed spent fuel assembly for final disposal. This means that after N PIE measurements, every fuel assembly which in the future is going to be disposed of is expected to have a theory-measurement uncertainty in the range $\left[m - c \cdot s\sqrt{\frac{n+1}{n}}; m + c \cdot s\sqrt{\frac{n+1}{n}}\right]$. Hence a large spread in PIE outcomes adds a large corresponding conservative margin or penalty.

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OBJECTIVE OF ACTIVITY 2.1 AND 2.4

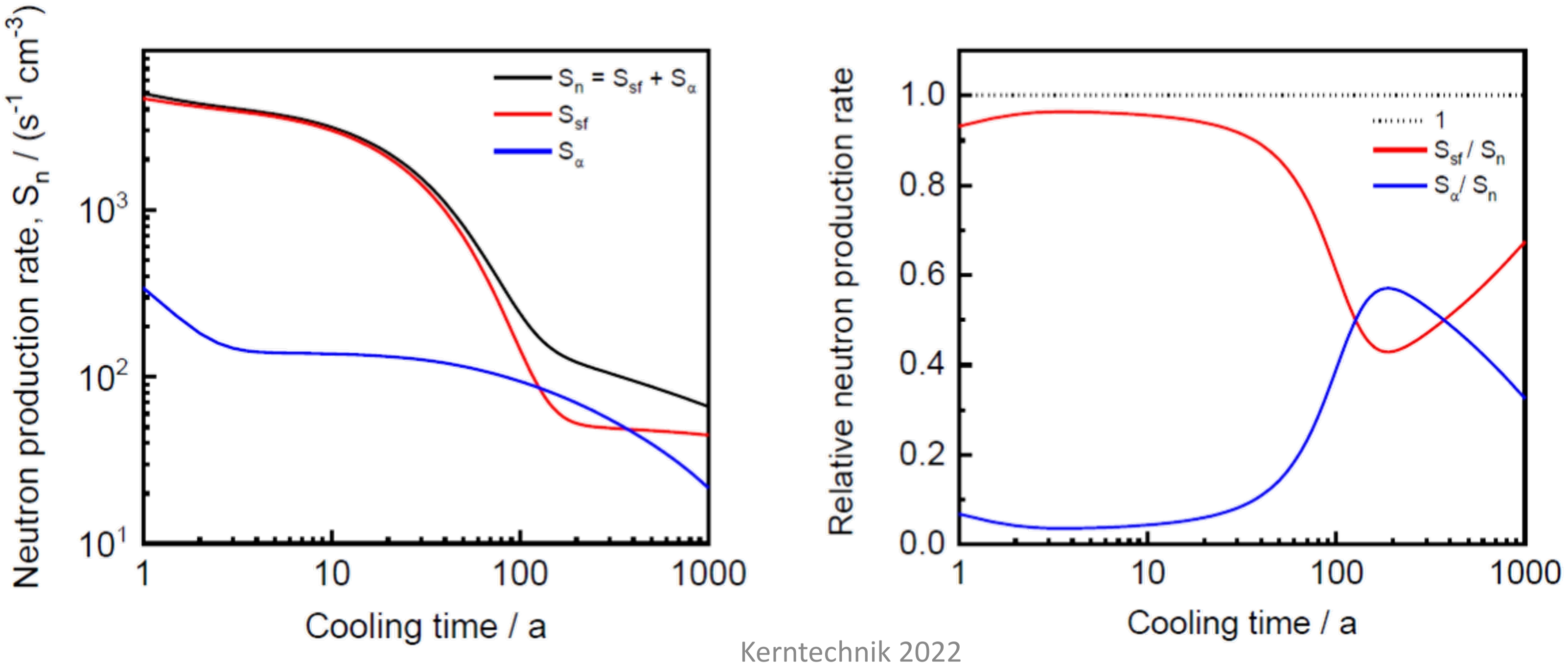
- Analysis of high-quality nuclide vector data: detailed knowledge of irradiation history and high confidence in experimental measurements. Disentangle microscopic data biases from uncertainties of irradiation history.

- SKB-50 dataset. Decay heat/neutron/gamma measurements on 50 fuel assemblies at CLAB. Continuation of a measurement series done in 2006.

- Some questions: based on biases and uncertainties from nuclide vector analysis, how well do source term measurements fall into predicted confidence intervals? Can the removal of biases in microscopic data also remove biases in source term C/E values? Does the spread of source term C/E values reflect the C/E nuclide vector spread or are there compensating effects? Are typical uncertainties of irradiation histories commensurate with C/E source term spreads?

NDA METHOD DEVELOPMENTS (2.3)

- Objective: measure non-destructively the neutron production rate of a SNF sample outside a hot cell under standard controlled area conditions with the neutron correlation method. In the work done so far, the goal was to derive the Cm244 inventory with a relative uncertainty of 2 % which is comparable to the performance of radiochemical analysis methods.
- Figure below: total and relative neutron production rate, PWR sample@50MWd/kg, 4.8w/o U235



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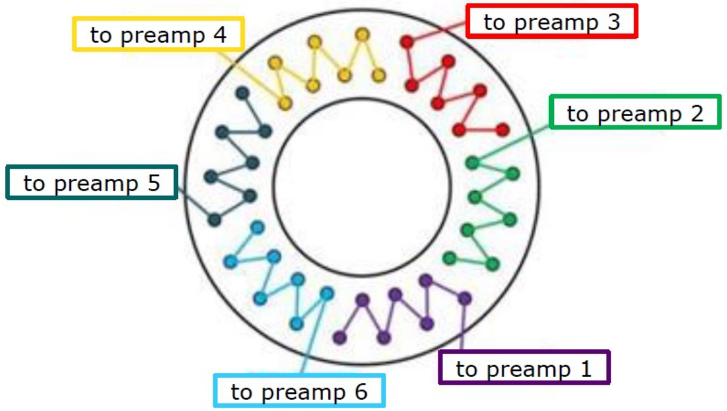
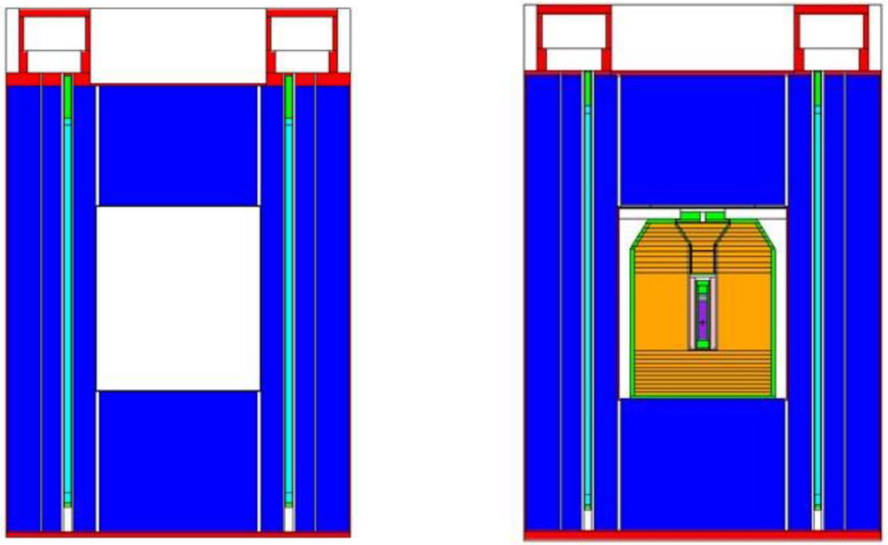
NDA METHOD DEVELOPMENTS (2.3)

- Neutron detection system is a transportable neutron well-counter based on an AWVV (Active Well Coincidence Counter) device.

- The detector consists of two concentric rings of 42 He3 proportional counters embedded in polyethylene that is used as a neutron moderator. They are divided into six groups of seven counters. Each group is connected to one hybrid charge sensitive preamplifier, discriminator and pulse shaper board.





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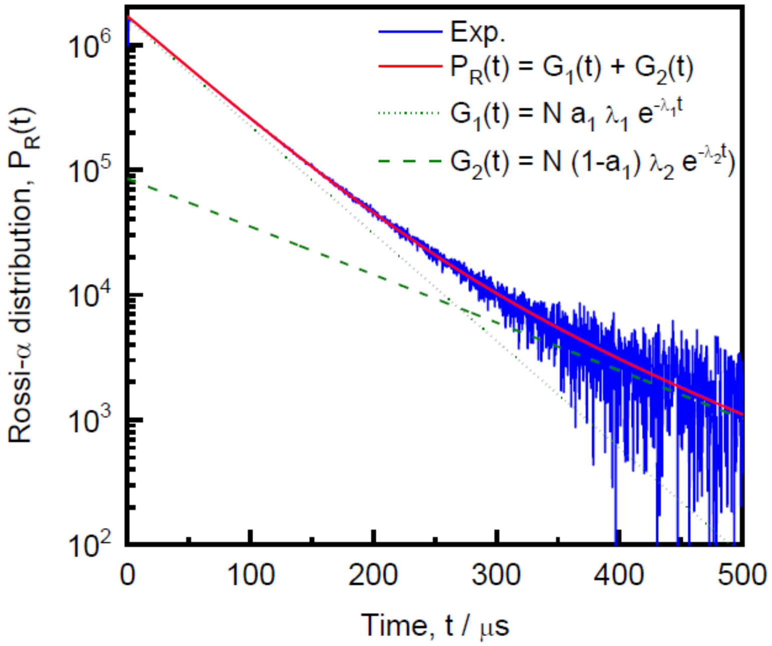
NDA METHOD DEVELOPMENTS (2.3)

- The measurement method is known as time interval analysis (TIA). It records the Rossi-α spectrum and determines the reals and totals count rate (accidental and real coincidence events). Together with other parameter like the normalized factorial moments for spontaneous and neutron induced fission the concentration of the spontaneous fission source can be calculated.

Uncertainty component, j	$\frac{u_{S_{sf},j}}{u_{S_{sf}}}$	$\frac{u_{\alpha,j}}{u_{\alpha}}$
Totals rate	< 0.01	0.06
Reals rate	0.18	0.18
Fraction of delayed neutrons	< 0.01	0.03
Detection efficiency for (sf) neutrons	0.46	0.23
Gate fraction	0.11	0.11
First order factorial moment for (sf)	0.24	0.25
Second order factorial moment for (sf)	0.83	0.83
Relative detection efficiencies	0.07	< 0.01
Neutron induced fission probabilities	0.06	0.04
Neutron induced capture probabilities	0.01	< 0.01
First order factorial moment for (n,f)	0.02	0.01
Second order factorial moment for (n,f)	0.02	0.02



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FISSION PRODUCTS IN CLADDING MATERIALS (2.2)

- For longer-term intermediate storage, the material properties of the fuel rod cladding materials are relevant.

- Cladding material impurities can come from the production process and from the contact of the pellets with the cladding during fabrication. Later cladding creep-down and pellet swell result in a periods of strong pellet-cladding surface contacts during which fission products and actinides can be implanted. Also, more volatile nuclides can be deposited from the pellet onto the cladding surface.

- For the experimental studies, two kinds of samples were prepared:(a) cladded fuel pellets of UO2 (50 GWd/tHM) irradiated in a pressurized water reactor (PWR); (b) a plenum cladding obtained from a UO2 fuel rod segment irradiated in a PWR. Various spectrometric methods are used to determine the exact material compositions.

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