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Effect of PWR Operational History on the Uncertainty of UNF’s Radionuclide Inventory

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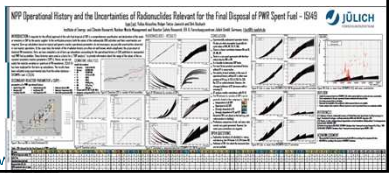
PKS Product Quality Control Office for Radioactive Waste

Outline

1. **Disposability of Radioactive Waste in Germany**
2. **Nuclide Characterization, Inventory Declaration, Numerical Tools**
3. **Direct Disposal and PQC of Spent Fuel**
4. **Fuel Operational History & SRP Analysis of BU & CT**
    - **Parameter of an operational history of PWR**
    - **Determination of the limits and dependencies between SRPs**
    - **Downtime analysis**
    - **Results of burn-up calculations and comparison with experimental data from SFCOMPO database**
5. **Summary & Outlook**

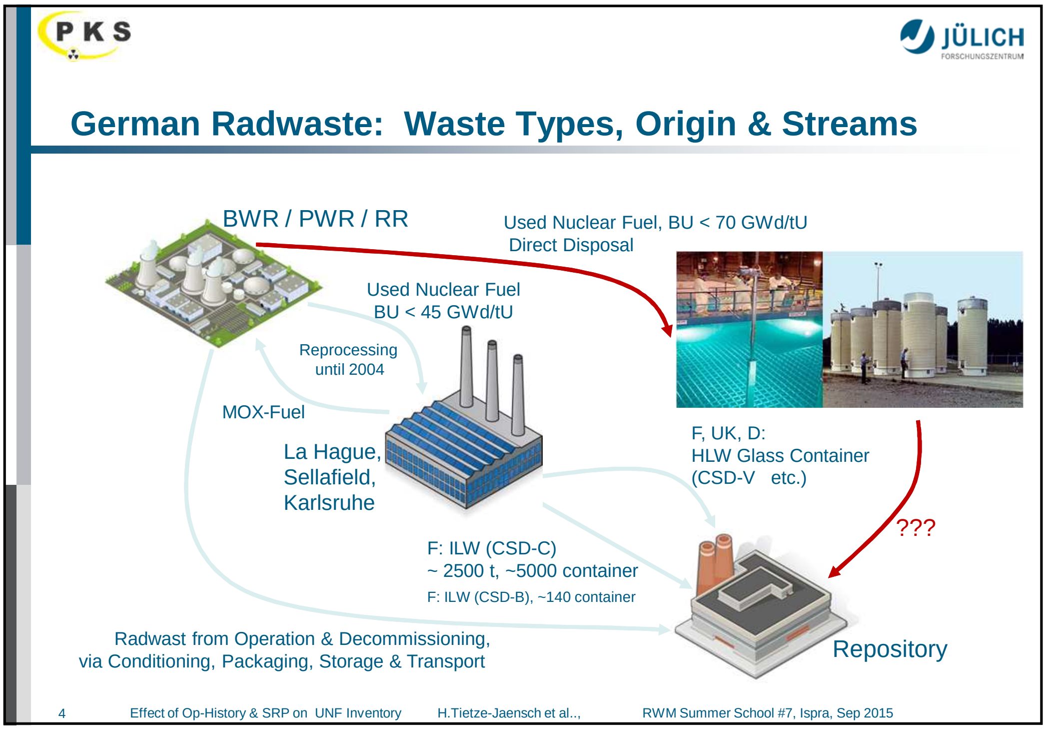
PQC: product quality control,
SRP: secondary reactor parameters,
BU: burn-up, CT: cooling time

Poster at WMS 2015:



Nuclear Waste: What to Do After Use?

Safe and Sustainable Disposal



German Radwaste: Waste Types, Origin & Streams

BWR / PWR / RR

Used Nuclear Fuel, BU < 70 GWd/tU
Direct Disposal

Used Nuclear Fuel
BU < 45 GWd/tU

Reprocessing
until 2004

MOX-Fuel

La Hague,
Sellafield,
Karlsruhe

F, UK, D:
HLW Glass Container
(CSD-V etc.)

???

F: ILW (CSD-C)
~ 2500 t, ~5000 container

F: ILW (CSD-B), ~140 container

Radwast from Operation & Decommissioning,
via Conditioning, Packaging, Storage & Transport

Repository

Sources of Radioactive Waste

1. **Operations**
    1. HLW: spent fuel element
        1. Reprocessing
        2. Direct disposal
    2. LLW / ILW: solid & liquid operational waste
        1. Rinsing & filter residues
        2. Activated or contaminated waste
2. **Decommissioning**
    1. HLW: spent fuel (cf. 1.1)
    2. Solid / liquid waste (large quantities)

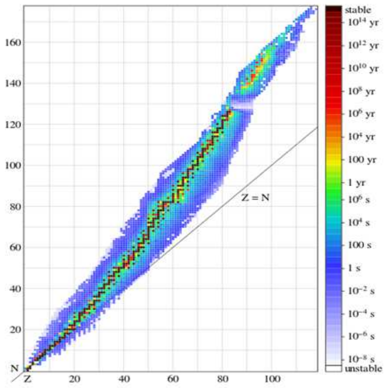
Professional

- Pre-Characterization
- Sorting
- Conditioning
- Full-Characterization
- Packing / Containers
- Documentation
- Transport
- Interim Storage
- Final Disposal

Product Quality Control

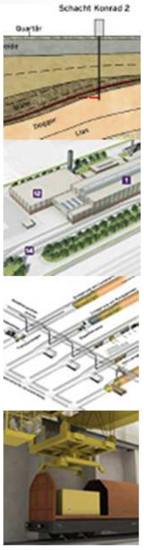
German HLW Repository Requirements

KONRAD (LLW / ILW) Repository (under construction):
Positive permissible nuclide and materials list, i.e. nothing else is permitted for disposal !



Permissible radio-nuclides:

- Defined & fixed list of ~230 declarable & limited RN-activities for whole repository & every approved container.
- Max. permissible masses of toxic material for the whole repository and for each container.
- Max. activity & toxic levels are limited also from safety (MCA) assessment: incidents / accidents that may affect host rock, heat load, criticality survey



Determination of a Standard Container Nuclear Inventory

Nucl. measurement

Photo: AREVA-NC, La Hague

Measured nuclides	Calculated nuclides (families)
	H-3, Kr-85, I-129, C-14
Co-60	Cl-36, Ca-41, Mn-54, Fe-55, Co-58, Ni-59, Ni-63, Mo-93
Cs-134 Cs-137	Ag-108m Ag-110m Cs-135
Sb-125	Se-79, Sb-124, Sn-126
Eu-154	(Ce+Pr)-144, Pm-147, Sm-151, Eu-152, Eu-155
	(Sr+Y)-90, Zr-93, Zr-95, Nb-94, Nb-95, Tc-99
	Ru-103, **(Rh+Ru)-106,** Pd-107
Cm-244	Ra-226, Pa-231, Np-237, Am-241, Am-242m, Am-243, Cm-242, Cm-243, Cm-245, Cm-246, Cm-247, Cm-248, Cf-249, Cf-251, Cf-252
U-235 U-238	Th-229, Th-230, Th-232, U-232, U-233, U-234, U-236
Pu-239 Pu-241	Ac-227, Pu-238, Pu-240, Pu-242, Pu-243, Pu-244

Motivation

Objectives

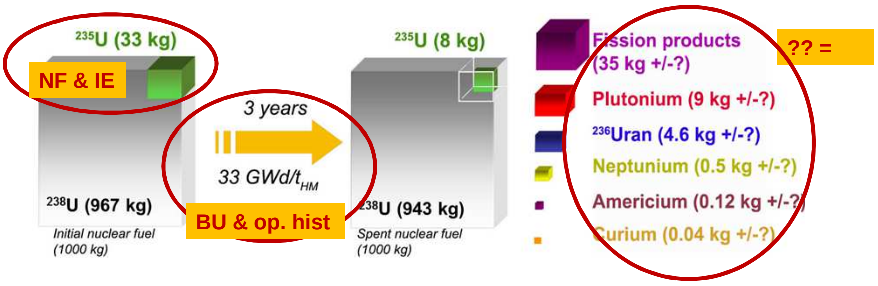
Knowledge of the composition of the radiological inventory of the radioactive waste

- for independent verification of the declared radwaste inventory by Product Quality Control
- for final disposal
- for long-term safety assessment

Challenges

- The radwaste may contain different fuel assemblies (FA) with several operational histories (e.g. reprocessing products)
- Comprehensive information about the operational history may not be fully available

Nuclear Inventory after Burn-Up: Operational History & SRP

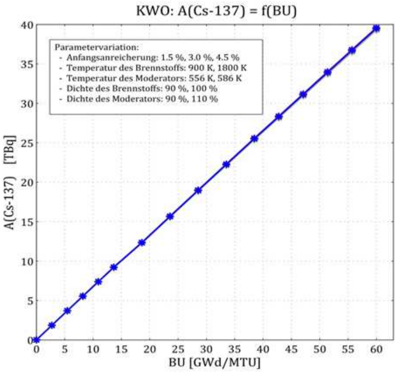


SRP

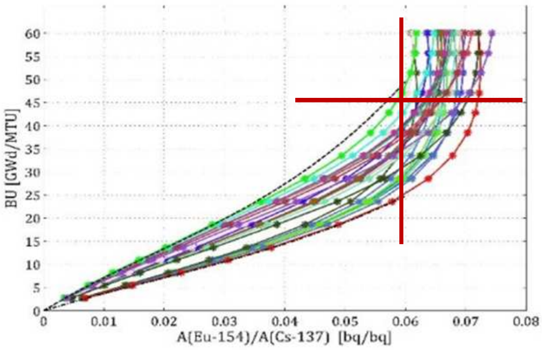
- Operational history of burn-up (BU)
  - Specific power (SP), downtime (DT), irradiation time (IT)
- Fuel properties
  - Initial enrichment (IE), density (FD), temperature (FT)
- Moderator properties
  - Density (MD), temperature (MT), boric acid concentration (BA)

Reactor Parameters (Fuel/Moderator) Influence BU Correlations

Cs-137 activity ~ burn-up (BU)
(direct FP, i.e. no secondary reactor parameter dependence)



Eu-154/Cs-134 correlation ~ burn-up (BU) is not valid for high-BU > 45 GWd/t$_{HM}$



From I. Fast et al., WM2013

Determination of the Limits and Dependencies Between the Secondary Reactor Parameters (SRP)

Objectives

- Analysis of the whole spectrum of the secondary reactor parameters
- Using of the existing data of real spent fuel
- Using of existing theoretical models (if available)
- Building a fuel assembly (FA) database including comprehensive SRP data
- Determining the nuclide vectors of SNF with representative mean values and corresponding minimum and maximum bandwidths
- Validation of the simulation results with experimental data
- Determining the uncertainties of PWR radionuclide inventory by calculated and experimental data

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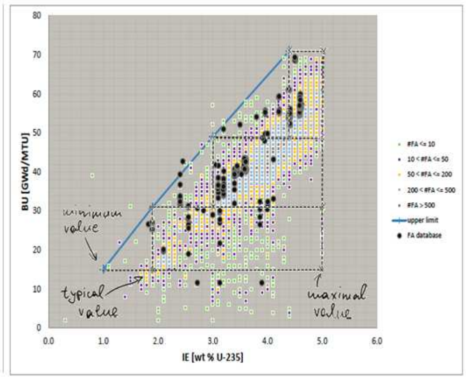
Downtime & SRP Analysis (BU vs. IE)



Ref.: J.M. Scaglione et. al., „Burnup Credit Approach Used in the Yucca Mountain License Application“, ORNL, 2010

Overview on fuel assembly (FA)

- FA database contains 339 FA
  69 FA supplied from literature studies
  270 FA from industry submission
- The FA data base comprises information of the SRP mean values for whole irradiation period and often also details of the SRP-values and the associated reactor operating cycle

SRP: secondary reactor parameters,
FA: fuel assembly

Results of BU-Calculations (SCALE 6.1) and Comparison with Experimental Data (SFCOMPO)

Nuclides to be declared for disposal in Germany:

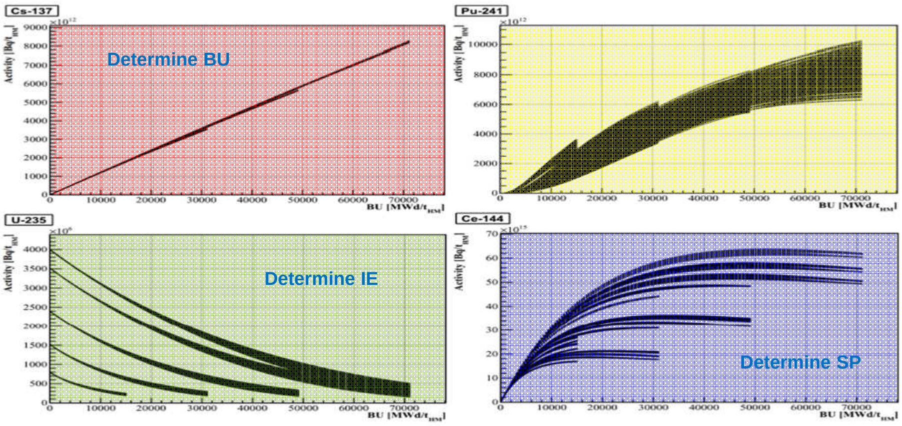
Ac-227, Ag-108m, Ag-110m, **Am-241**, **Am-242m**, **Am-243**, C-14, **Ce-144**, Cf-249, Cf-251, **Cm-242**, Cm-243, **Cm-244**, **Cm-245**, **Cm-246**, **Cm-247**, Cm-248, **Cs-134**, **Cs-135**, **Cs-137**, Eu-152, **Eu-154**, Eu-155, H-3, I-129, Kr-85, Mo-93, Nb-94, Nb-95, **Np-237**, Pa-231, Pd-107, Pm-147, Pr-144, **Pu-238**, **Pu-239**, **Pu-240**, **Pu-241**, **Pu-242**, Pu-243, Pu-244, Ra-226, Rh-106, Ru-103, **Ru-106**, Sb-124, **Sb-125**, **Se-79**, **Sm-151**, **Sn-126**, **Sr-90**, **Tc-99**, Th-229, Th-230, Th-232, **U-232**, U-233, **U-234**, **U-235**, U-236, **U-238**, Y-90, Zr-93, Zr-95

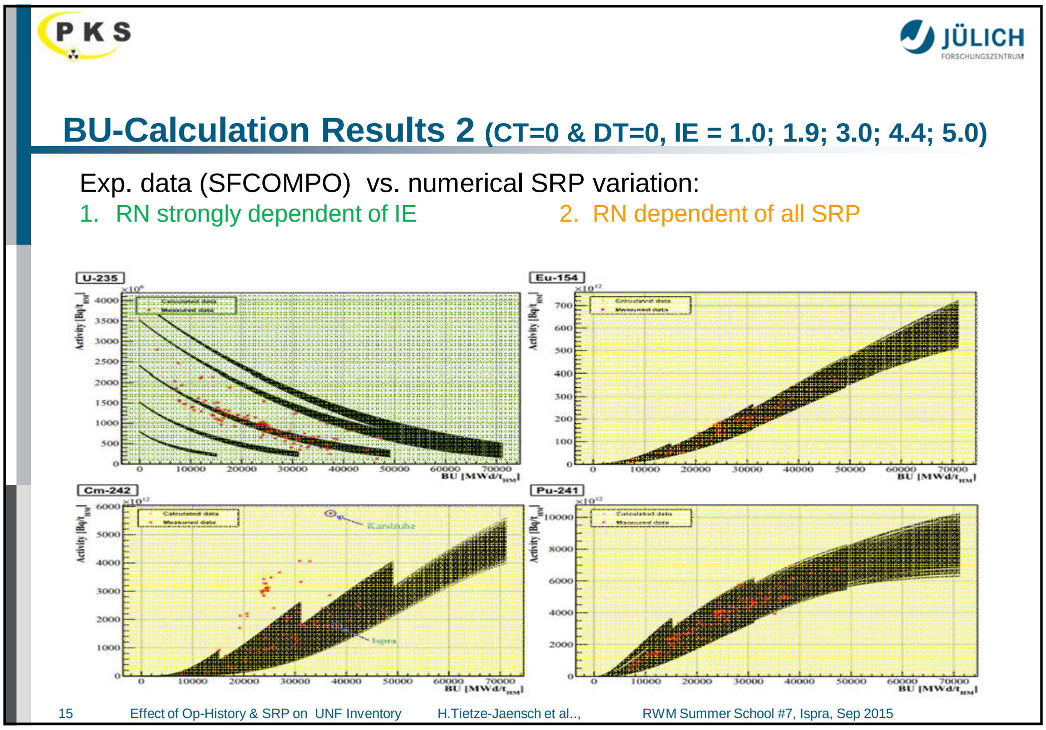
(in bold: SFCOMPO provides experimental data for comparison)

BU-Calculation Results 1 (CT=0 & DT=0, IE = 1.0; 1.9; 3.0; 4.4; 5.0)

Varying SRP → 4 groups of RN behaviour:

1. RN merely independent of all SRP
2. RN dependent of all SRP
3. RN strongly dependent of IE
4. RN strongly dependent of IE & SP

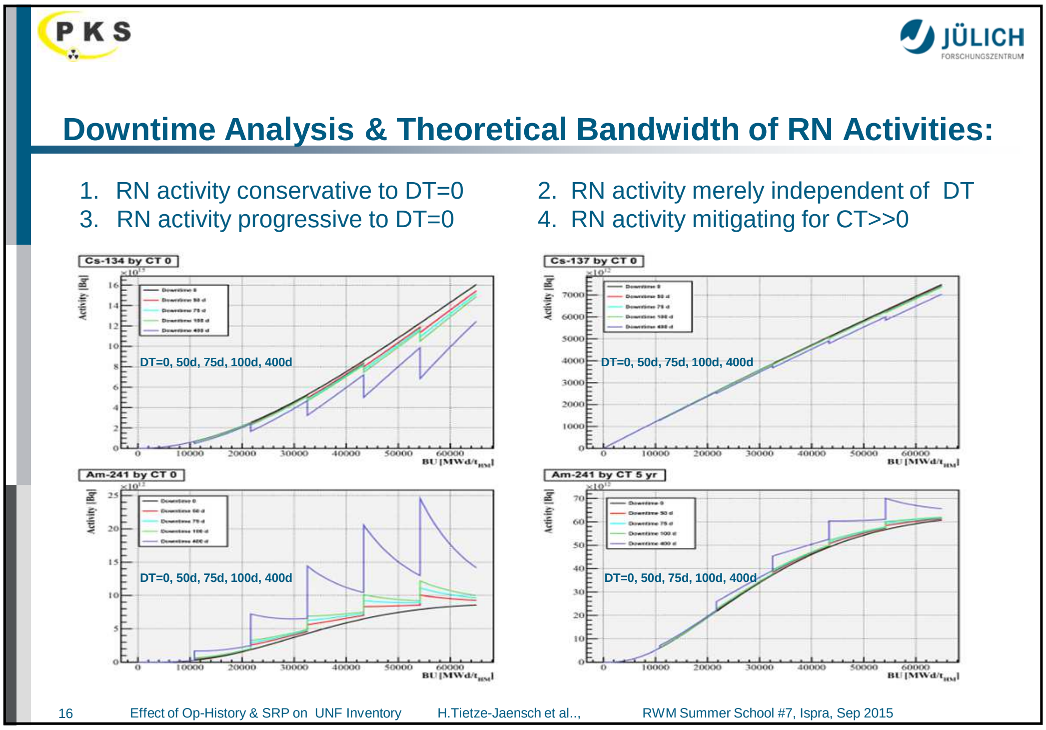




BU-Calculation Results 2 (CT=0 & DT=0, IE = 1.0; 1.9; 3.0; 4.4; 5.0)

Exp. data (SFCOMPO) vs. numerical SRP variation:

1. RN strongly dependent of IE
2. RN dependent of all SRP



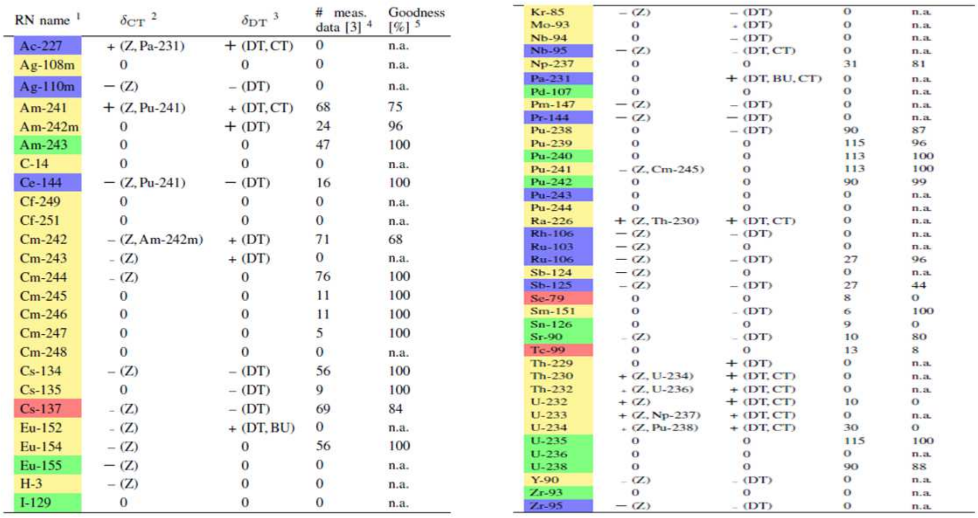
Downtime Analysis & Theoretical Bandwidth of RN Activities:

1. RN activity conservative to DT=0
2. RN activity merely independent of DT
3. RN activity progressive to DT=0
4. RN activity mitigating for CT>>0

Downtime Analysis, Model Description

- Geometry: FA 16×16 AFA 3G (Siemens)
- Burn-up: 65 GWd/tHM
- Specific power: 36 MW/tHM
- Initial Enrichment: 4.0 % wt U-235
- Fuel temperature: 900 K
- Moderator temperature: 580 K
- Fuel density: 10.408 g/cm3
- Moderator density: 0.73 g/cm3
- Boric acid concentration: 460 ppm
- 6 Cycles with DT: 0, 50, 75, 100 and 400 days
- Analysis for cooling time (CT): 0, 1, 3 and 5 years

Conclusion Table (cf. I. Fast et al., Animma 2015, Lisbon, April 2015)



RN name [1]	δ_{CT} [2]	δ_{DT} [3]	# meas. data [3] [4]	Goodness [%] [5]
Ac-227	+ (Z, Pa-231)	+ (DT, CT)	0	n.a.
Ag-108m	0	0	0	n.a.
Ag-110m	− (Z)	− (DT)	0	n.a.
Am-241	+ (Z, Pu-241)	+ (DT, CT)	68	75
Am-242m	0	+ (DT)	24	96
Am-243	0	0	47	100
C-14	0	0	0	n.a.
Ce-144	− (Z, Pu-241)	− (DT)	16	100
Cf-249	0	0	0	n.a.
Cf-251	0	0	0	n.a.
Cm-242	− (Z, Am-242m)	+ (DT)	71	68
Cm-243	− (Z)	+ (DT)	0	n.a.
Cm-244	− (Z)	0	76	100
Cm-245	0	0	11	100
Cm-246	0	0	11	100
Cm-247	0	0	5	100
Cm-248	0	0	0	n.a.
Cs-134	− (Z)	− (DT)	56	100
Cs-135	0	− (DT)	9	100
Cs-137	− (Z)	− (DT)	69	84
Eu-152	− (Z)	+ (DT, BU)	0	n.a.
Eu-154	− (Z)	0	56	100
Eu-155	− (Z)	0	0	n.a.
H-3	− (Z)	0	0	n.a.
I-129	0	0	0	n.a.
Kr-85	− (Z)	− (DT)	0	n.a.
Mo-93	0	+ (DT)	0	n.a.
Nb-94	0	− (DT)	0	n.a.
Nb-95	− (Z)	− (DT, CT)	0	n.a.
Np-237	0	0	31	81
Pa-231	0	+ (DT, BU, CT)	0	n.a.
Pd-107	0	0	0	n.a.
Pm-147	− (Z)	− (DT)	0	n.a.
Pr-144	− (Z)	− (DT)	0	n.a.
Pu-238	0	− (DT)	90	87
Pu-239	0	0	115	96
Pu-240	0	0	113	100
Pu-241	− (Z, Cm-245)	0	113	100
Pu-242	0	0	90	99
Pu-243	0	0	0	n.a.
Pu-244	0	0	0	n.a.
Ra-226	+ (Z, Th-230)	+ (DT, CT)	0	n.a.
Rh-106	− (Z)	− (DT)	0	n.a.
Ru-103	− (Z)	0	0	n.a.
Ru-106	− (Z)	− (DT)	27	96
Sb-124	− (Z)	0	0	n.a.
Sb-125	− (Z)	− (DT)	27	44
Se-79	0	0	8	0
Sm-151	0	− (DT)	6	100
Sn-126	0	0	9	0
Sr-90	− (Z)	− (DT)	10	80
Tc-99	0	0	13	8
Th-229	0	+ (DT)	0	n.a.
Th-230	+ (Z, U-234)	+ (DT, CT)	0	n.a.
Th-232	+ (Z, U-236)	+ (DT, CT)	0	n.a.
U-232	+ (Z)	+ (DT, CT)	10	0
U-233	+ (Z, Np-237)	+ (DT, CT)	0	n.a.
U-234	+ (Z, Pu-238)	+ (DT, CT)	30	0
U-235	0	0	115	100
U-236	0	0	0	n.a.
U-238	0	0	90	88
Y-90	− (Z)	− (DT)	0	n.a.
Zr-93	0	0	0	n.a.
Zr-95	− (Z)	− (DT)	0	n.a.

Explanations of Previous Table

1. The color describes the RN behavior to the variation of SRPs. There are four groups: RN independent of all SRP red, RN dependent of all SRP yellow, RN strongly dependent of IE green and RN strongly dependent of IE and SP blue.
2. CT is the correction factor of post-irradiation cooling time for measured data. This factor is a function of the own decay (Z) rate of that RN and applicable on the parent RN. The sign denotes positive or negative impact on the RN activity relatively to CT=0. The size of the sign shows the magnitude of this influence.
3. DT is the correction factor of down time for the theoretical bandwidth. The sign denotes positive or negative impact on the RN activity relatively to the theoretical bandwidth for DT=0. Again, the size of the sign shows the magnitude of this influence.
4. The number of individual radiochemical measurements of several samples from the SFCOMPO-1 database. Goodness provides the quality of agreement (in %) of calculated vs. measured data, i.e. number of measured data within the theoretical bandwidth; n.a.: not applicable.

Summary

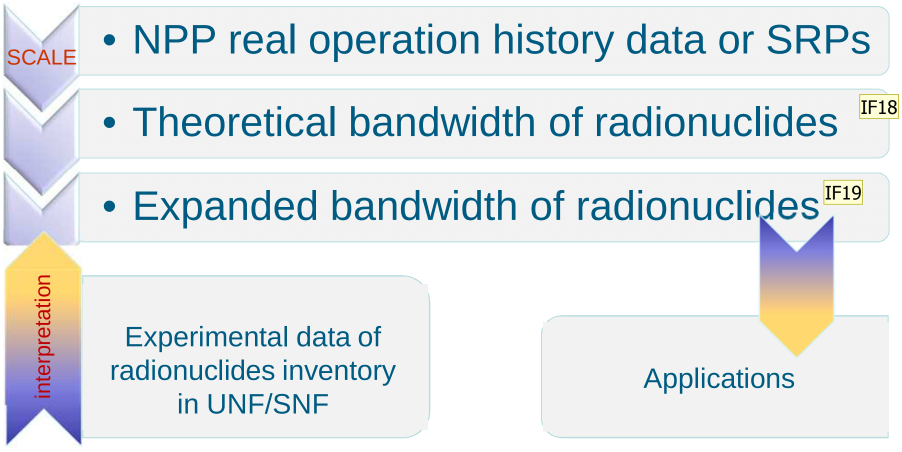
Summary

- Comparison of calculated and measured data reveals good agreement. However, for some cases correction is required.
- Missing measured data for high burn-up
- The nuclides are differently influenced by a variation of the operational history, revealing different behavior:
- Independent of all SRP: Se-79, Tc-99, Cs-137
- Dependence of all SRP: C-14, Am-241, Am-242, Pu-239, Pu-241 etc.
- Strong dependence of IE: Zr-93, I-129, Eu-155, U-235, Am-243m, etc.
- Strong dependence of IE & SP: Nb-95, Zr-95, Ru-106, Sb-126, Ce-144, Ac-227, etc.

Conclusion

1. Experimentally underpinned operation history FA data are often incomplete (especially for cycle averaged values of MD, MT, FD, FT, BA).
2. It is feasible to determine the SRP variability limits.
3. We have performed burn-up calculations for 64 RN relevant for the final disposal of radioactive waste. Activation products were not considered.
4. For most fission products the calculations assuming an operational history with DT=0 are either independent of DT or overestimated (exceptions: Eu-152, Mo-93).
5. Many actinides activities (operational history with DT=0) are either independent of DT (e.g. U-235, U-238, Pu-239, Np-237) or underestimated (e.g. Ac-227, Am-241, Cm-242, Ra-226).
6. DT analysis enables calculations with DT=0. In the case of activities systematically over- or underestimated, resp., the theoretical bandwidths must be corrected. IF17
7. Due to the variation of the SRP values the RN behavior can be generally divided into 4 groups.
8. Comparison of calculated and measured data reveals generally good agreement. However, for some RNs a comprehensive agreement between calculated and measured values requires corrections as specified in the table above.
9. The uncertainty of the radionuclide inventory of PWR spent fuel computed on the basis of real operational history data is less than one order of a magnitude of the actual activity of the relevant radionuclide.

Appendix: Numerical Methodology



- NPP real operation history data or SRPs
- Theoretical bandwidth of radionuclides IF18
- Expanded bandwidth of radionuclides IF19

Experimental data of radionuclides inventory in UNF/SNF

Applications

Slide 21

IF17 corrected

Ivan Fast, 8/23/2015

Slide 22

IF18 Ich würde hier einfach "Theoretical bandwidth ..." schreiben. Hier braucht man noch keine detaillierte Beschreibung.

Ivan Fast, 8/23/2015

IF19 Ich würde hier einfach "Expanded bandwidth of radionuclides" schreiben. es ist auch klar, das es die selbe Radionuklide gemeint wie bei theoretischer Bandbreite

Ivan Fast, 8/23/2015

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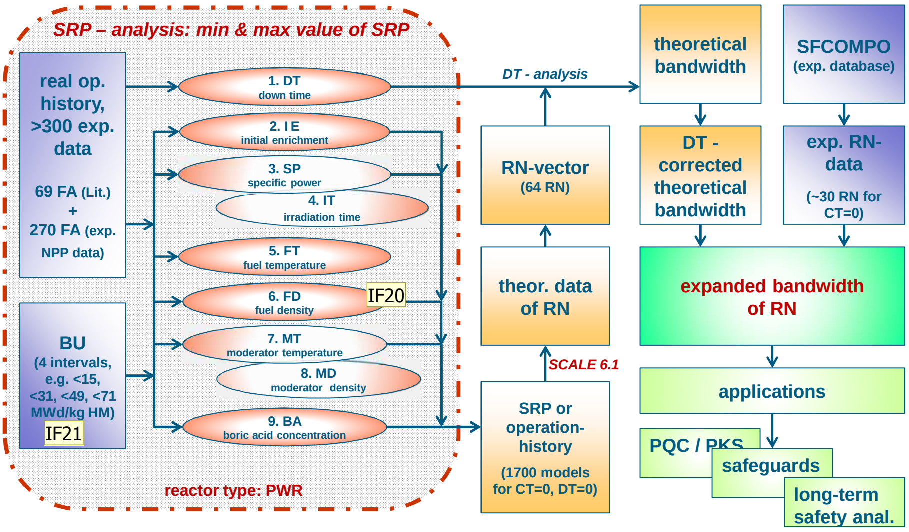
SRP – analysis: min & max value of SRP

real op. history, >300 exp. data

69 FA (Lit.)
+
270 FA (exp. NPP data)

BU
(4 intervals, e.g. <15, <31, <49, <71 MWd/kg HM)

IF21

1. DT down time
2. I E initial enrichment
3. SP specific power
4. IT irradiation time
5. FT fuel temperature
6. FD fuel density
7. MT moderator temperature
8. MD moderator density
9. BA boric acid concentration

IF20

reactor type: PWR

SRP or operation-history (1700 models for CT=0, DT=0)

SCALE 6.1

theor. data of RN

RN-vector (64 RN)

DT - analysis

theoretical bandwidth

SFCOMPO (exp. database)

DT - corrected theoretical bandwidth

exp. RN-data (~30 RN for CT=0)

expanded bandwidth of RN

applications

PQC / PKS

safeguards

long-term safety anal.

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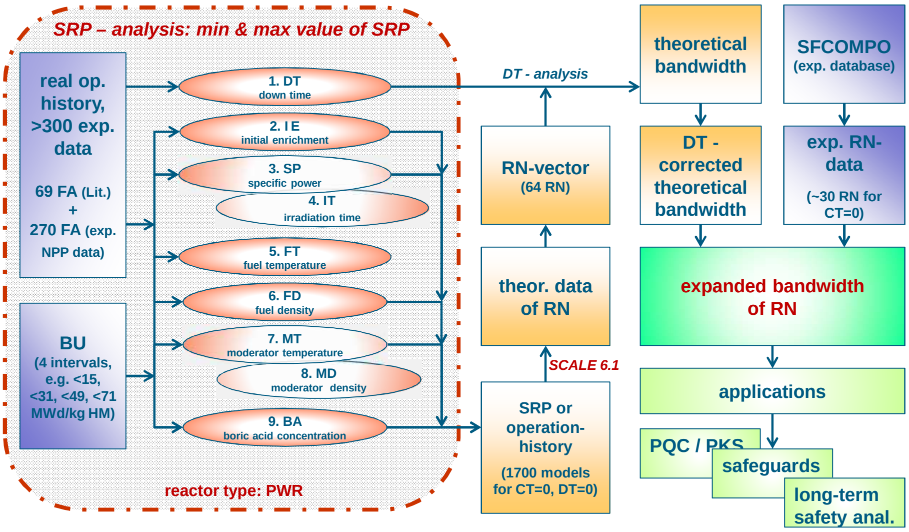
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exp. RN-data (~30 RN for CT=0)

expanded bandwidth of RN

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PQC / PKS

safeguards

long-term safety anal.

Slide 23

IF20 Fuel density würde ich auch klein schreiben wie alle andere SRP oder alle groß.
Ivan Fast, 8/23/2015

IF21 Hier würde ich in Klammern entweder nur "4 intervals" schreiben oder Intervalle richtig schreiben "0-15, 0-31, 0-49 und 0-71"
Ivan Fast, 8/23/2015

Applications

Applications

- PQC: product quality control of FA packages for direct disposal
- Nuclear forensic / safeguards
- Long-term safety assessment of UNF / SNF behaviour
- …

Acknowledgement to:

1. Wissenschaftlich Technische Ingenieurberatung GmbH (WTI), Jülich for FA data
2. NEA for SFCOMPO (2) access

Thank You for Your Attention!

Einlagerung von verfestigten schwachaktiven Abfällen

Folie 35

Schwachaktive Feststoffe in 200-l-Fässern

Flüssigkeiten in Beton oder Bitumen verfestigt

Lagerung im Salz

Erprobung seit 1967 in ASSE

Fässer abgekippt, mit Salz überdeckt

Bis Ende 1978 124 000 Fässer

Genehmigungsverfahren für Lagerung läuft

Of course we can do
a lot better than this

We must decide on the way forward
and not linger