Decay heat calculations and safety analyses for ARAMIS-A molten salt reactor

Jad HALWANI – PhD Seminar: 05/04/2025

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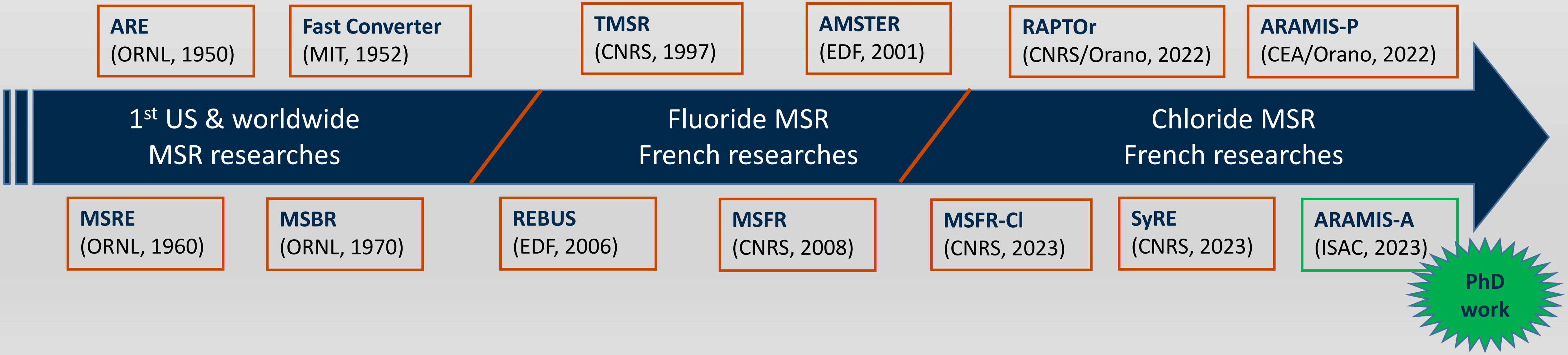
Supervisors:

- Lydie Giot (SUBATECH-Co-Director)
- Axel Laureau (LPSC)
- Elsa Merle (LPSC-Co-Director)

- **What is a Molten Salt Reactor (MSR)?**
  - Gen IV fission concept
  - Fuel is dissolved in a salt (fluoride / chloride)
  - Burner (U, Pu, Actinides) or breeder (Th/U or U/Pu)
  - Thermal or fast neutrons
- Fission products accumulation in fuel salt
  -> **Chemical treatments required**

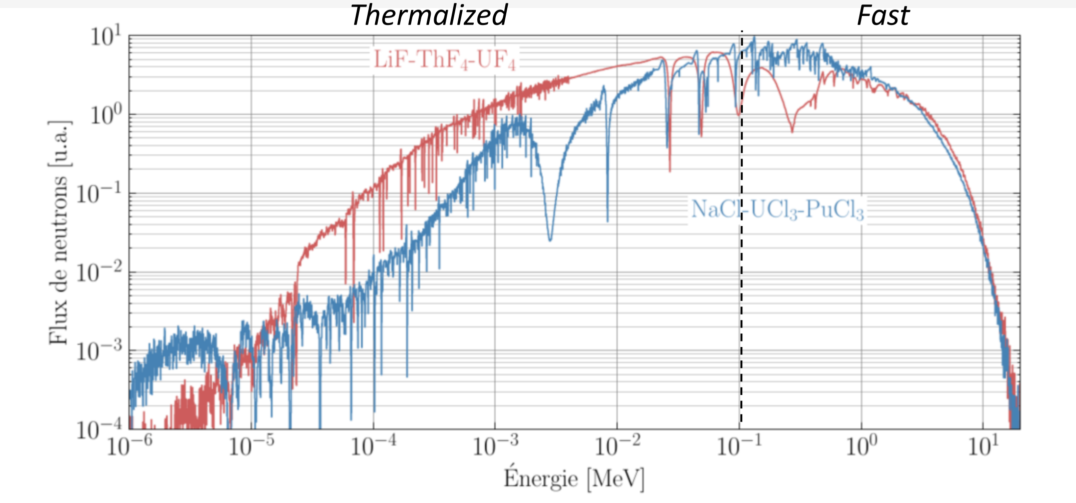
Potential benefits	Key challenges
Actinides transmutation	Salt chemistry
Inherent safety behaviour	Materials and technologies qualification
Flexibility	Safety in operation (design & paradigm)

Summary of MSR potential benefits and key challenges [GIF4]

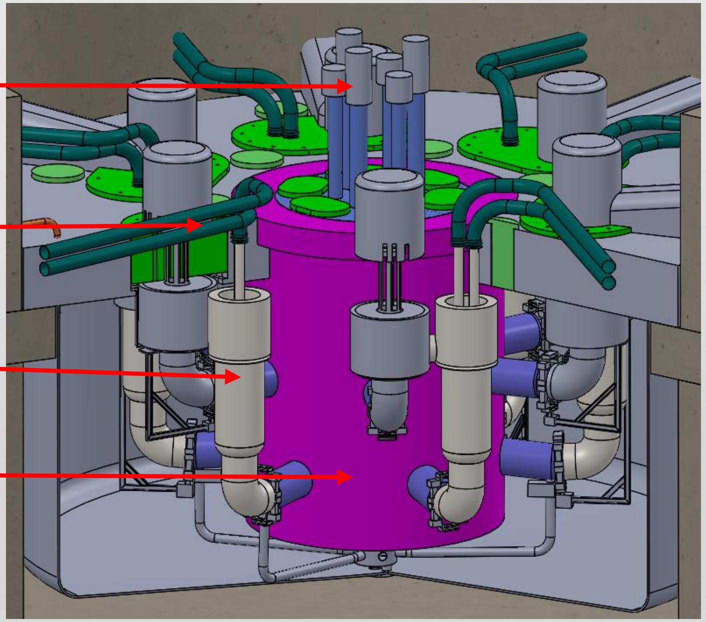


- **Why studying chloride MSR?**
  - Better Pu and minor actinides (Np, Am, Cm, …) solubility in chlorides than fluorides salt
  - Harder neutron spectrum in chlorides than fluorides salt
  - Higher fission cross-sections for Pu and minor actinides

-> Promising way to burn minor actinides like Am
(main radioactivity and decay heat contributor over a century)



Neutron spectra in fluorides (red) and chlorides (blue) [LMes2023]



Illustrative schematic view

- **ARAMIS-A concept**
  - 300 MW_{th} fast chloride molten salt reactor
  - Fuel salt: $NaCl\text{-}MgCl_2\text{-}(Pu\text{-}Am)Cl_3$
  - Am burning target: 50 kg/TWh_e (ADS maximum performance)
  - Periodic fuel salt reprocessing to control reactivity
  - Hypothesis: continuous gaseous and metallic fission products (FP) removal
    -> 3 source terms !
- **ISAC project**
  - French collaboration between CEA, CNRS, EDF, Framatome & Orano
  - Topics studied: design, maintenance, salt chemistry, materials integrity, nuclear scenarios, **decay heat**, **safety**, …

[LMes2023] L. Mesthiviers, *Capacité de conversion des transuraniens en RSF,* PhD Thesis, Grenoble-Alpes University, LPSC (2023)
[VPas2022] V. Pascal, *Molten Salt Reactor Technology: Opportunities of molten salt fuel for actinides management*. Proceedings of a Technical Meeting, IAEA TECDOC Series. Presented at the Technical Meeting (TM) on Considerations for the Back End of the Fuel Cycle of Small Modular Reactors, International Atomic Energy Agency, Vienna, Austria (2022)

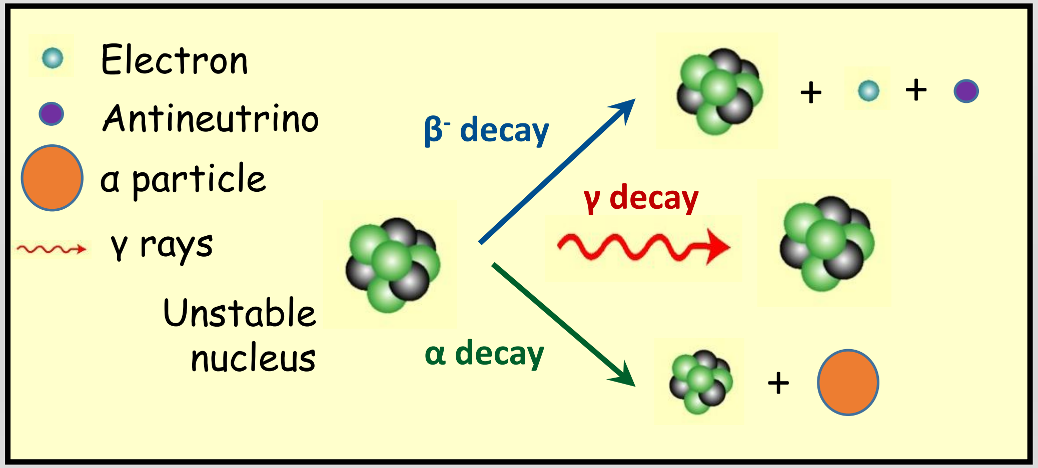
- **What is decay heat (DH)?**
  - Heat produced by nuclear power plant after shutdown
  - **Calculated by summing all *i* nucleus contributions to DH**

$$P_{res}(t) = \sum_i (E_{i,LP} + E_{i,EM} + E_{i,HP}).\lambda_i.N_i(t)$$

- $N_i(t)$: atoms number
- λ_i : decay constant
- $E_{i,LP}$: light particles mean decay energy (mostly β^- decays)
- $E_{i,EM}$: electromagnectic mean decay energy (mostly γ decays)
- $E_{i,HP}$: heavy particles mean decay energy (mostly α decays)

Concept	Maximum DH (% nominal power)
Pressurized Water Reactor (PWR) [MBro2013]	6%
Sodium-cooled Fast Reactor (SFR) [ACal2020]	7%
Molten Salt Fast Reactor (MSFR) [MBro2013]	5.5%

DH ordres of magnitude



-> Inventories have to be calculated during and after reactor operation

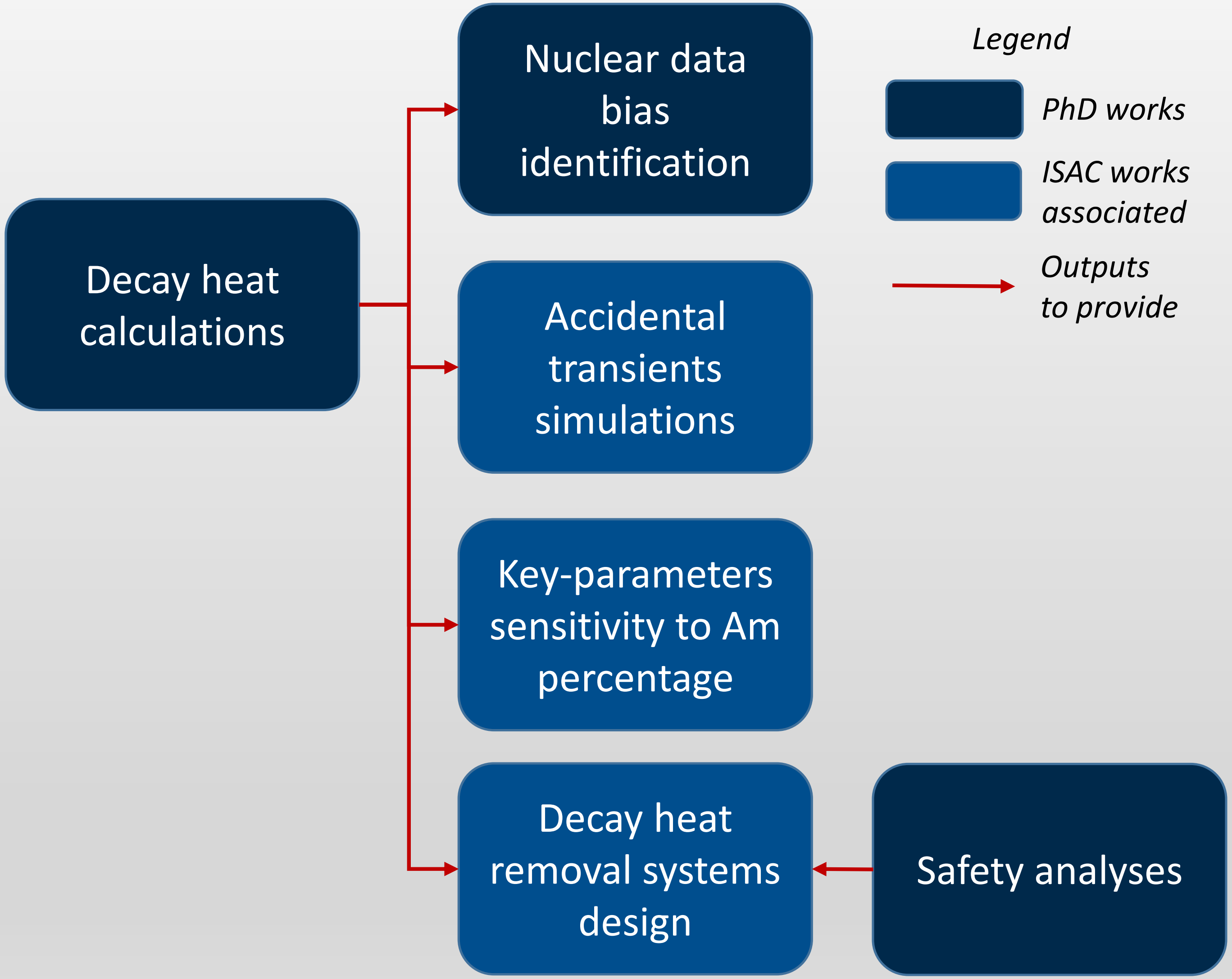
Cross sections, decay data and mean decay energies uncertainties and bias can have a significant impact on DH

-> Biased mean decay energies (Germanium energy efficiency)

[MBro2013] M. Brovchenko, *Études préliminaires de sûreté du réacteur à sels fondus MSFR*, PhD Thesis, Grenoble-Alpes University, LPSC (2013)
[ACal2020] A. Calame et al., *Analysis of a decay heat experiment in Superphenix for the experimental validation of the DARWIN3 package*, In Proceedings of Best Estimate Plus Uncertainty International Conference (BEPU 2020)

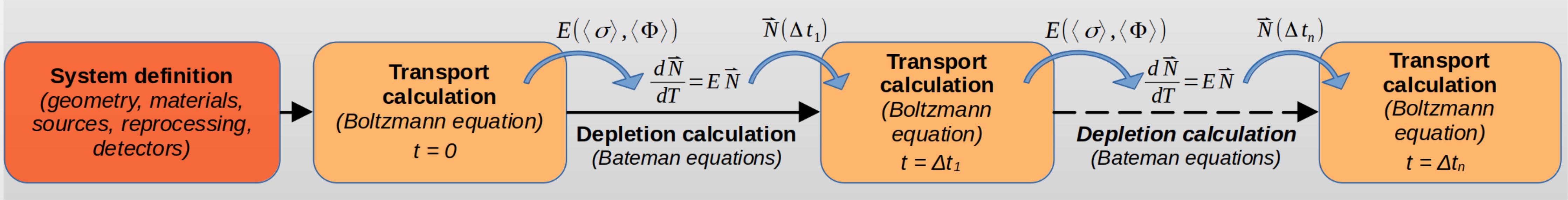
- **What are the motivations of this PhD?**
  - Decay heat calculations and safety analyses done for fluoride MSFR [MBro2013] [DGer2019]
  - Decay heat calculations done for 3 GW_{th} chloride MSFR reactivity-controlled by continuous reprocessing [HPit2023]

-> Not any decay heat calculations and safety analyses performed for a small fast chloride MSR reactivity-controlled by discontinuous reprocessing



[MBro2013] M. Brovchenko, *Études préliminaires de sûreté du réacteur à sels fondus MSFR*, PhD Thesis, Grenoble-Alpes University, LPSC (2013)
[DGer2019] D. Gérardin, *Développement de méthodes et d’outils numériques pour l’étude de la sûreté du réacteur à sels fondus MSFR*, PhD Thesis, Grenoble-Alpes University, LPSC (2018)
[HPit2023] H. Pitois, Design and optimisation of a Molten Salt Fast Reactor using chloride salts and uranium cycle, PhD Thesis, Grenoble-Alpes University, LPSC (2023)

- **How inventories are calculated?**



Transport and depletion equations coupling

- Transport (Boltzmann) and depletion (Bateman) equations are solved by SERPENT2 [Lep2015]
- Transport equations are solved with probabilistic (Monte-Carlo) approach
- Depletion equations are solved with CRAM method (order 16)

$$\frac{dN_i}{dt}(t) = \underbrace{-\lambda_i.N_i(t) + \sum_{j\to i}\lambda_j.N_j(t)}_{\text{Terms related to decays}} \underbrace{- N_i(t).\sigma_i.\phi + \sum_{l} N_l(t).\sigma_{l\to i}.\phi}_{\text{Terms related to reactions}} \underbrace{- \sum_k \lambda_{ext,k}.N_i(t) + \sum_p \lambda_{inj,p}.N_i(t)}_{\text{Terms related to reprocessors (MSR specificity)}}$$

Terms related to decays | **Terms related to reactions** | **Terms related to reprocessors (MSR specificity)**

Depletion equation for MSR

At CNRS MSR team, transport and depletion codes used: REM/MCNP (MSR reference code), OpenMC & SERPENT2

- **None of them can simulate reactivity control by periodic batch reprocessing**

-> A new interface code, coupled with an existing one, is required to simulate new physical processes

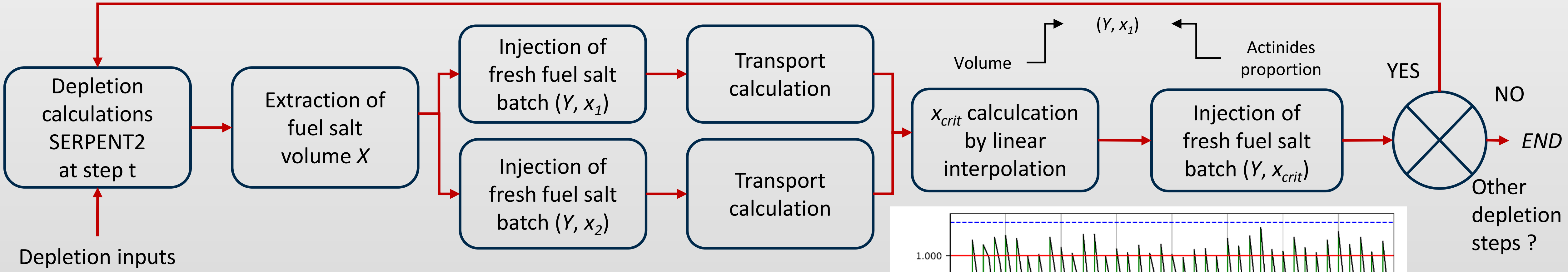
CEREIS logo

[Lep2015] J. Leppänen et al., The Serpent Monte Carlo code: Status, development and applications in 2013, Ann. Nucl. Energy, 82 (2015) 142-150

- **What is CEREIS (*Contrôleur en Evolution d'un Réacteur par Extraction et Injection de Sel*)?**
  - Python interface code between transport and depletion code (here SERPENT2) and control modules (here of reactivity)
  - Development started with Alexis Bodinier inthernship works
  - **Hypothesis: k_{eff} increase is linear proportional to actinides proportion injected**

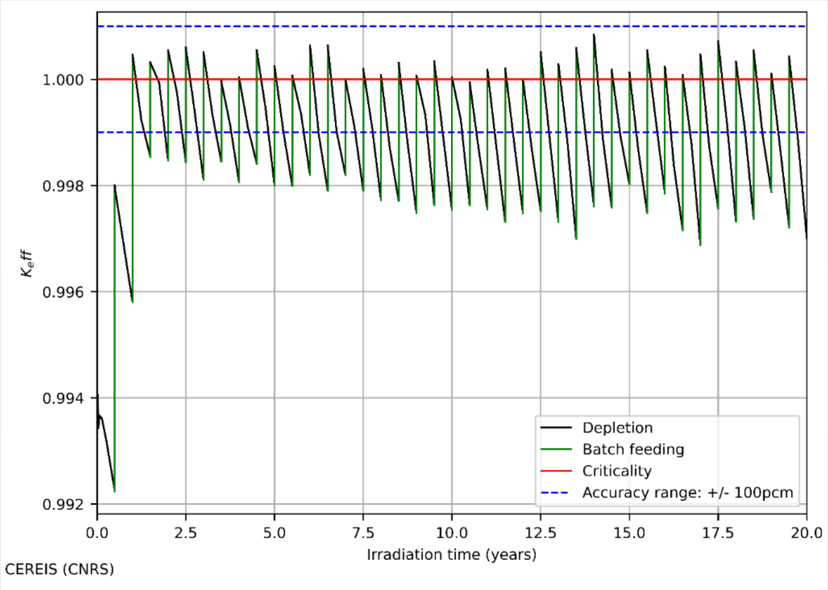
$$k_{eff} = \frac{Neutrons\ produced\ by\ fission}{Neutrons\ lost\ by\ absorption\ and\ leakage}$$

Target: k_{eff} = 1 (crititcality)

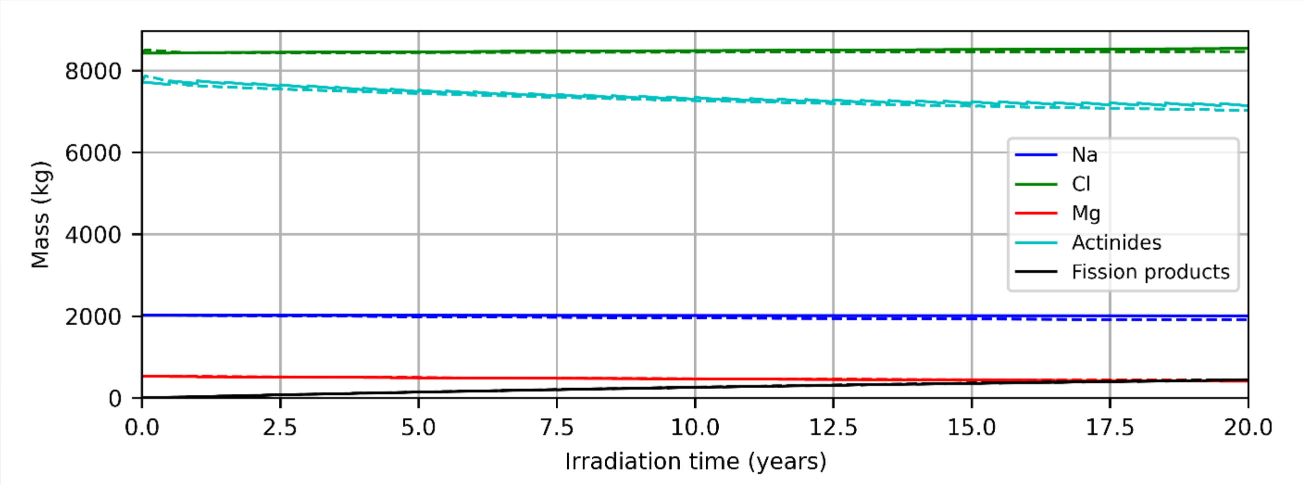


- **Approach precise (+/- 100 pcm) but time-consuming**
  -> Not suited for operator controls
- **Alternative approaches under-development**
  - **k_{eff} approximation (4 factor-formula + leakage estimation)**
  - **Sensitivity factor ($\Delta\%(Pu\text{-}Am)Cl_3/\Delta k_{eff}$)**

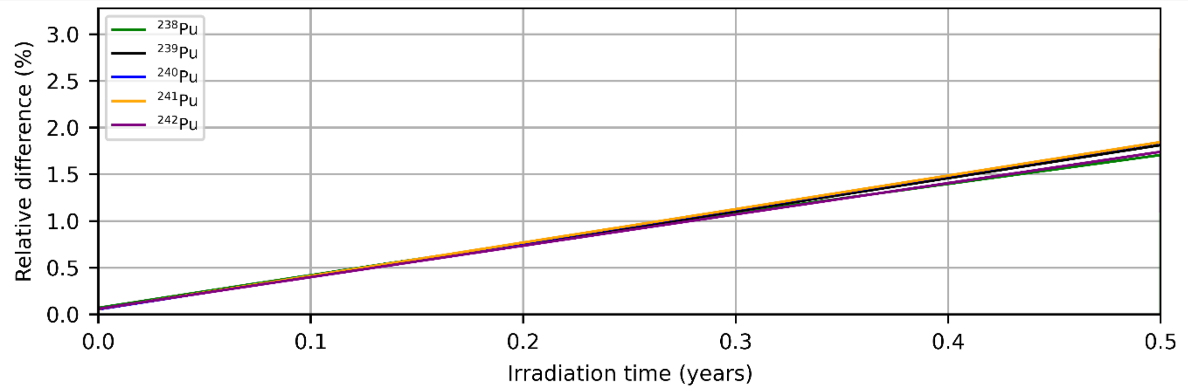
Poster at MIMOSA Summer School (July 2024)



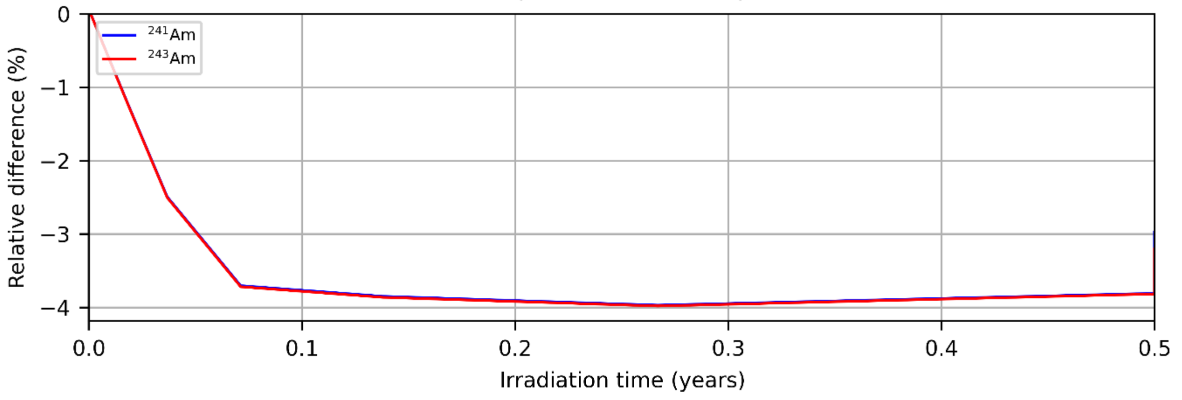
k_{eff} evolution of ARAMIS-A



Global inventories evolution (CEREIS with continuous lines and REM with dashed lines)



CEREIS/REM discrepancies on ^{239}Pu and ^{240}Pu



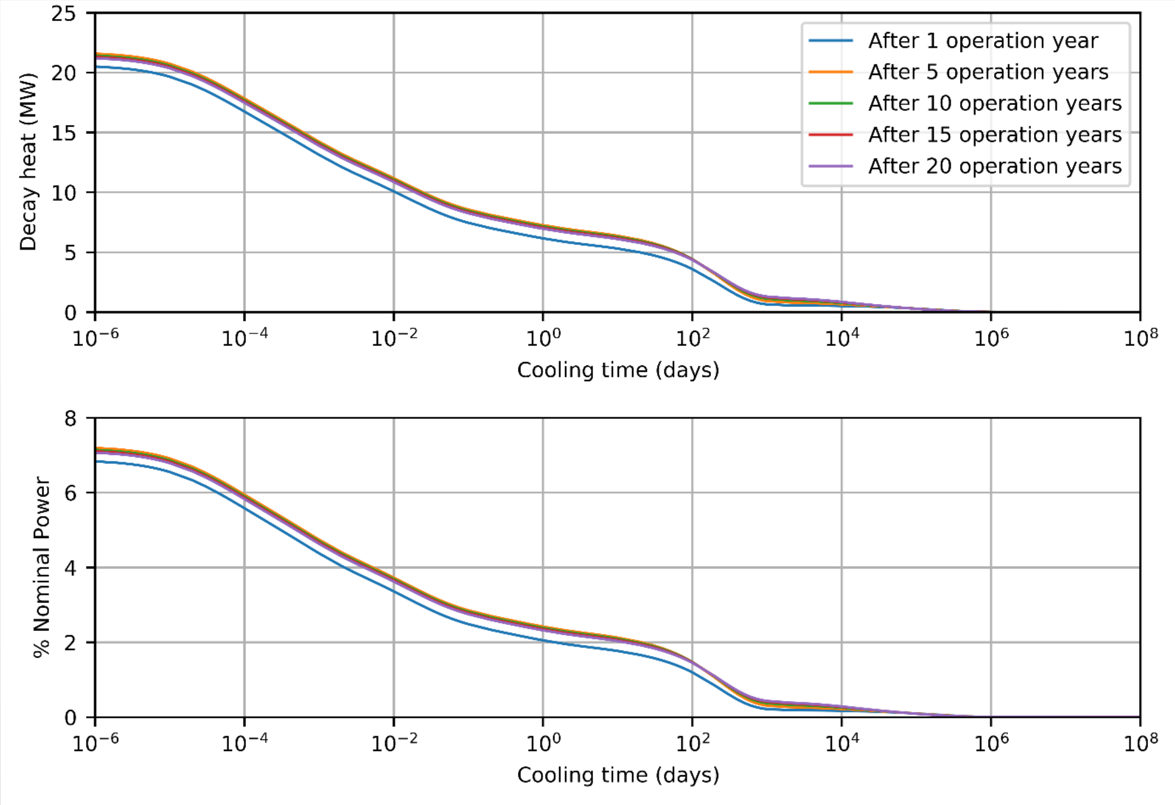
CEREIS/REM discrepancies on ^{241}Am and ^{243}Am

Impact of modelling differences under analysis

- Reactivity control process
- Multiplication factor estimator used for reactivity control
- Feeding volume and composition

After 20 operation years, decay heat maximum of 21.32 MW (7.11 % of nominal power)

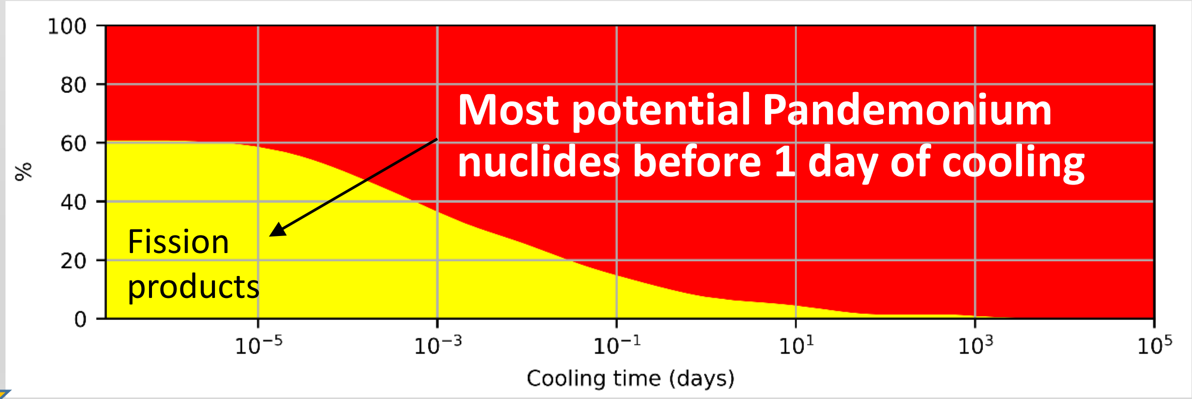
- 15.43 MW from depleted fuel
- 1.83 MW from extracted gaseous FP
- 4.06 MW from extracted metallic FP



Decay heat from depleted fuel and extracted fission products

Significant decay heat increase between 1 and 5 operation years

- **Consequence of mid-life actinides accumulation: ^{238}Pu, ^{242}Cm and ^{244}Cm -> Investigation in progress**

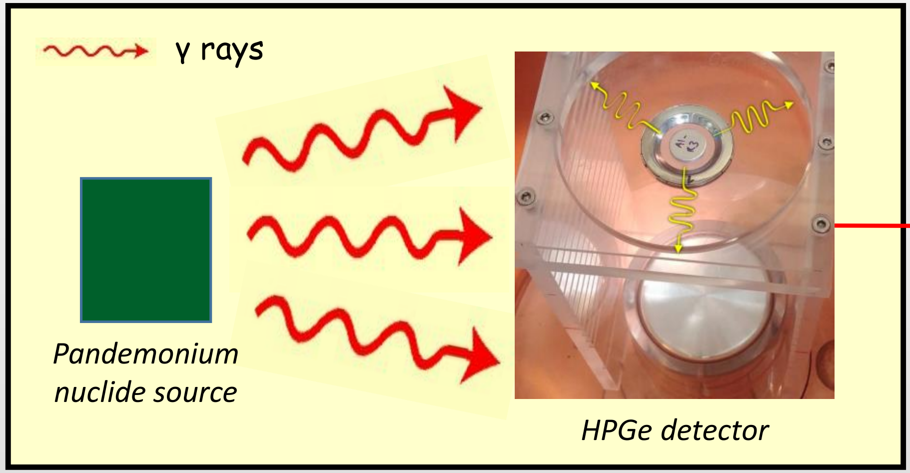


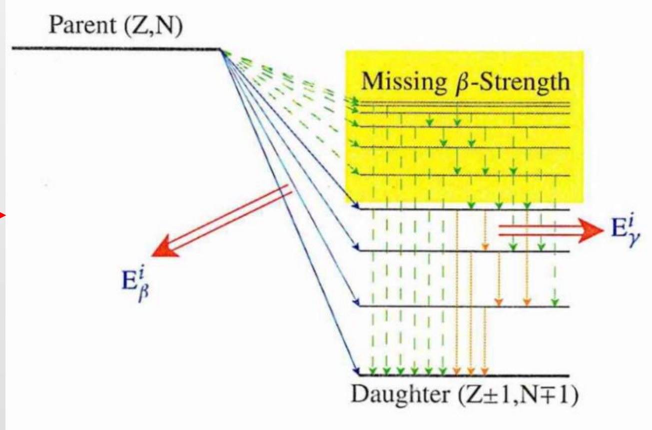
Decay heat repartition between actinides (in red) and fission products (in yellow) in depleted fuel

ISAC technical note (Septembre 2024)

Proceeding submitted to ICAPP 2025

- **What is the Pandemonium effect?**
  - To calculate $E_{i,LP}$ and $E_{i,EM}$, β- decay scheme has to be determined
  - β- decay scheme is deduced from γ intensities coming from excitation levels measured with high-resolution γ dectector, like Hyper Pure Germanium (HPGe)

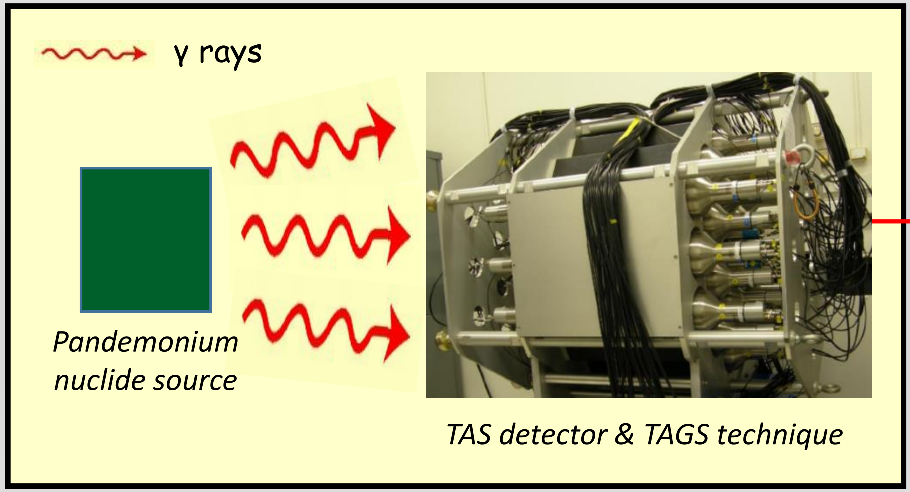


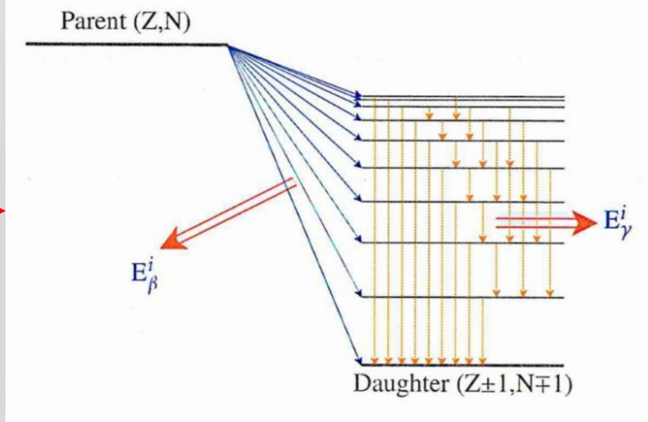


Biased β- decay scheme

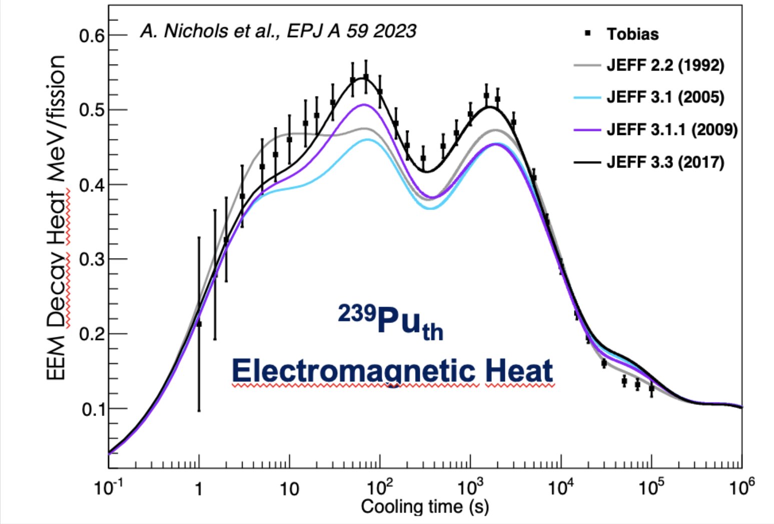
-> β- feeding at high excitation levels is not detected and is wrongly assigned to β- feeding at lower excitation levels

-> $E_{i,LP}$ (β- decays) is overestimated & $E_{i,EM}$ (γ decays) is underestimated





Real β- decay scheme

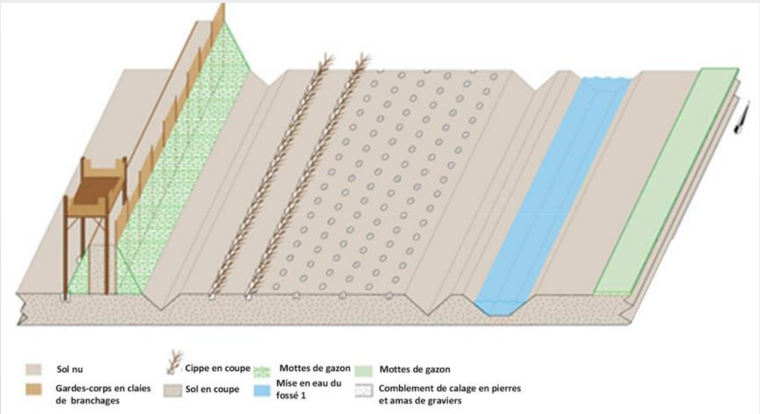


$E_{i,EM}$ comparaison for ^{239}Pu thermal fission pulse [ANic2023]

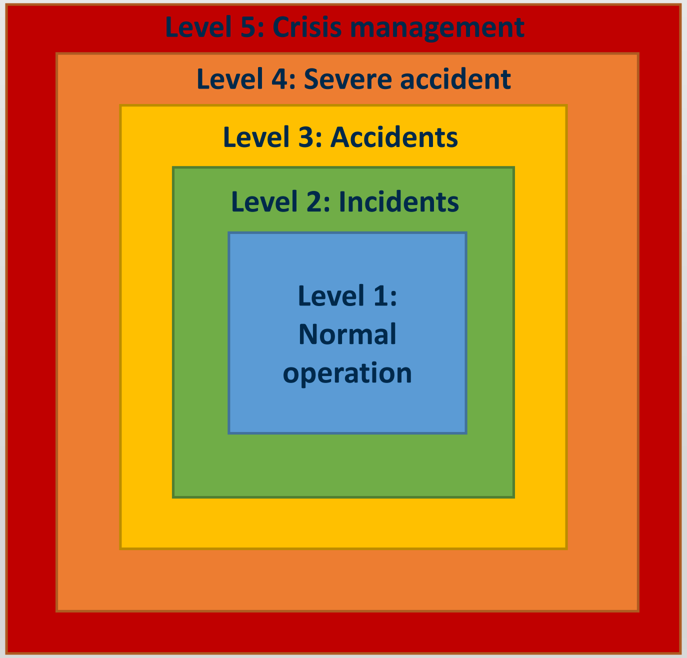
[SJRice2014] S.J Rice *Decay Heat M easurements of Fission Fragments ^{86}Br, ^{91}Rb & ^{94}Sr Using Total Absorption Gamma-ray Spectroscopy*, PhD Thesis, Surrey University (2014)
[ANic2023] A. Nichols et al. - *Improving fission-product decay data for reactor applications: part I—decay heat* – Eur. Phys. J. A (2023) 59:78 https://doi.org/10.1140/epja/s10050-023-00969-x

Before switching to safety, say « Hi » to Teddy

- **What is nuclear safety?**
  - Radionuclides are contained by physical **confinement barriers**
  - Safety provisions to protect these barriers have to ensure **3 functions at any operation condition and at any time:**
    - Reactivity control (as much neutrons produced by fission than lost by absorption or leakage)
    - Heat removal (from fission and decay heat)
    - Radionuclides confinement
  - Safety provisions are positionned according to **defence in depth principle**



Defence in depth military application for Alesia battle [Vid2010]



Defence in depth schematic view

Level 1: Normal operation
Monitoring and control provisions to stay there

Level 2: Incidents
Small impact failure
Monitoring and control provisions to return to level 1

Level 3: Accidents
High impact failure
Protection provisions to reach safety state

Level 4: Severe accident
Several high impact failures (melting core for PWR, not identified yet for MSR)
Protection provisions to reach safety state and mitigate consequences

Level 5: Crisis management
How to avoid to « RUN FOR YOUR LIFE »?
Public plans to protect people and environement

What provisions are required for ARAMIS-A?

-> Focus on decay heat removal function

[ASN2017] ASN/IRSN, *Guide n'°22 : Conception des réacteurs à eau sous pression*, 2017
[FBer2018] F. Bertrand, *Méthodologies d'études de sûreté et applications pour la pré-conception de différents types de réacteurs nucléaires de quatrième génération*, HDR, Grenoble Alpes University, CEA (2018)
[Vid2010] J. Vidal et C. Petit, *L'eau sur le site d'Alésia : la contrainte hydrogéologique lors du siège de 52 av. J.-C*, Revue Archélologique de l'Est, Tome 59-1 | 2010 : Fasc. 1 - n° 181

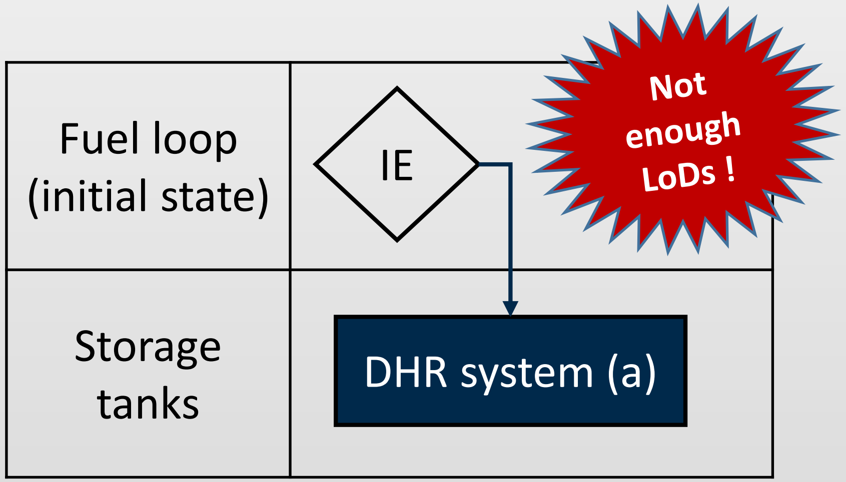
- **How to conduct safety analyses?**
  - Collaborative workshops involving conceptual design and safety specialists among CEA, CNRS, EDF, Framatome and Orano
  - Through 4 workshops, **Lines of Defence method** has been applied to identify decay heat removal (DHR) systems requirements
- **What is the Lines of Defence method?**
  - Ensure that every abnormal evolution of the reactor into a severe accident (contact between fuel salt and last confinement barrier) is always prevented by a minimum set of homogenous (in number and quality) safety provisions called *lines of defence* (LoD)

  -> If not, make a feedback to design
  - 2 types of lines of defence:
    - Strong lines (a): Provisions with a failure rate between 10^{-3} to 10^{-4} by year and use (active and passive systems with redundancies)
    - Medium lines (b): Provisions with a failure rate between 10^{-1} to 10^{-2} by year and use (active systems without redundancies, inherent safety behavior, operator actions)
    - 2b lines are equal to 1a
  - An incidental or accidental initiating event (IE) is the starting point of an abnormal evolution
  - Lines of defence minimum number is dependant to IE freqency

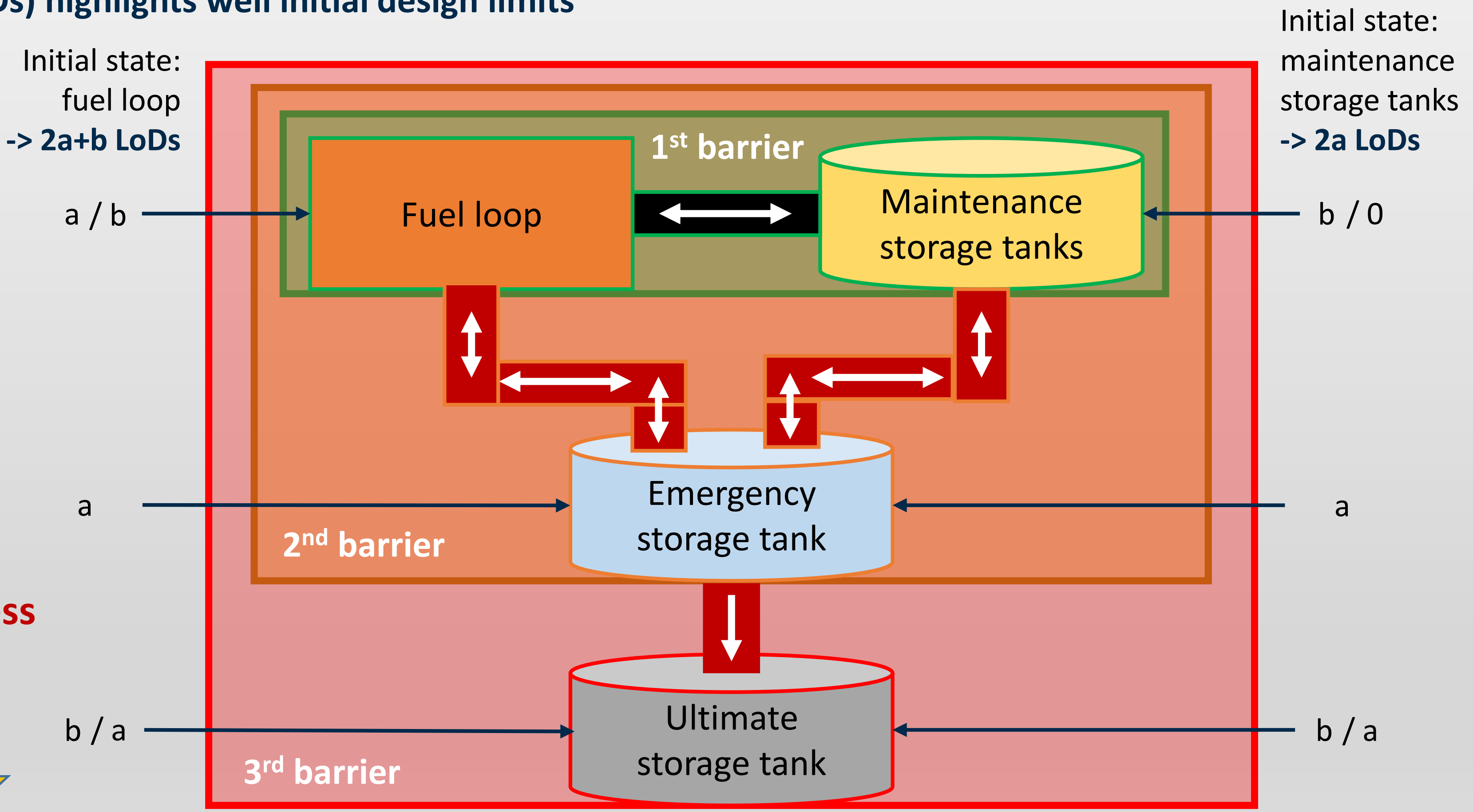
Initiating event category	Initiating event frequency (per year)	Number of LoDs required
Incidental	$> 10^{-2}$	2a+b
Accidental	$< 10^{-2}$	2a
Hypothetical accident	$< 10^{-4}$	a+b

LoDs number needed per initiating event category

- **What are LoD application conclusions?**
  - LoD method applied to several IE: loss of main heat sink (LOHS), loss of liquid fuel (LOLF), reactivity anomalies accident (RAA), loss of fuel flow (LOFF), overcooling (OVC) and total loss of power (TLOP)
  - **Loss of liquid fuel (accidental IE = 2a LoDs) highlights well initial design limits**



LoD application to fuel loop break with initial design



Proposed fuel salt locations configuration with DHR systems and preliminary confinement barrier definition

- **Confinement barrier definition in progress**
- **What about radioactive gases ?**

Proceeding submitted to ICAPP 2025

Decay heat calculations

- CEREIS interface code with reactivity control modules developped
- 1st inventories and decay heat calculations on ARAMIS-A
- Coming:
    - Discrepancies analysis with REM code
    - Decay heat parametric studies
    - Pandemonium nuclides identification
    - Internship: Implementation and comparison of valencies calculation methods (Sylvain Le Coupannec & Nathan Le Bourdonnec)

Safety analyses

- LDD method applied to several initiating events
- Configuration proposed for fuel salt locations and decay heat removal systems
- Coming:
    - Deepening of confinement barriers definition
    - Analyse extension to radioactive gases confinement and decay heat removal

UNE PEPITE

Sens Critique

Coming in August 2025

UN BANGER

Les Cahier du cinéma

SURPRENANT

Télérama

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framatome

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orano

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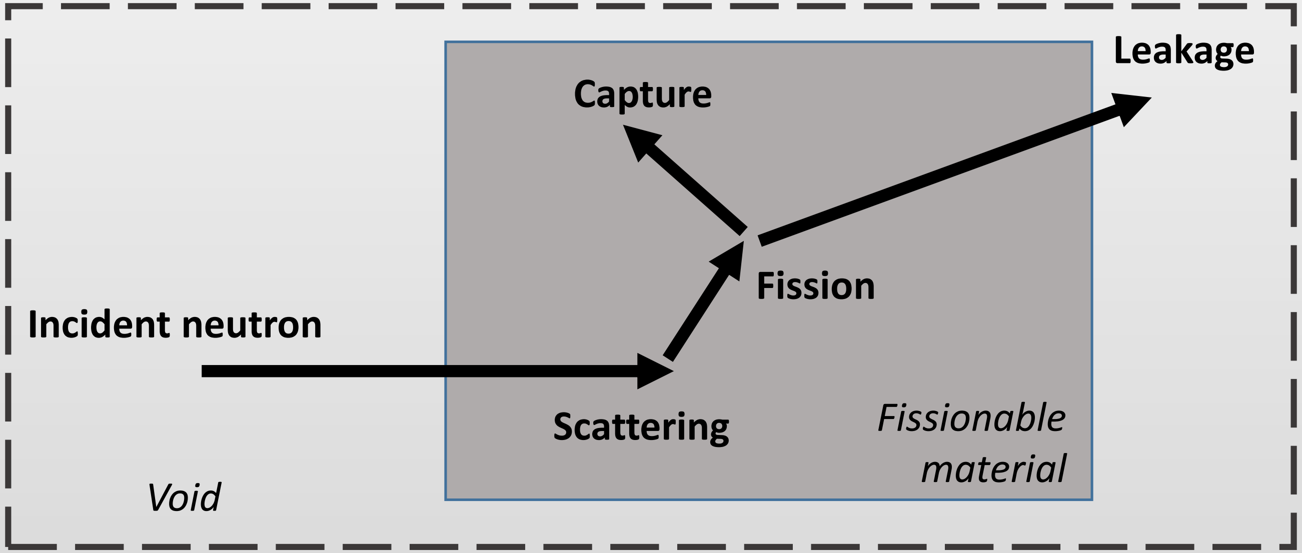
Thank you for your attention !

Appendix

- **How does Monte-Carlo approach works?**



Monte-Carlo simulation example with 1 incident neutron

- Reaction probabilities P_i (cross-sections) are computed by transport code before process
- N incidents neutrons are simulated multiples times
  -> Reduce k_{eff} estimator uncertainty
  -> Stabilise Shannon entropy

Appendix

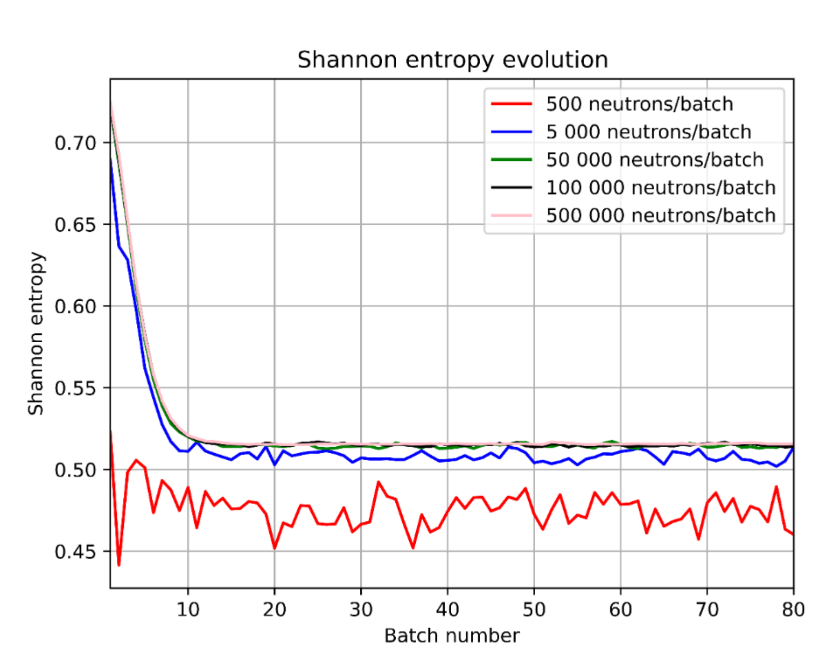
- **What is the Shannon entropy?**

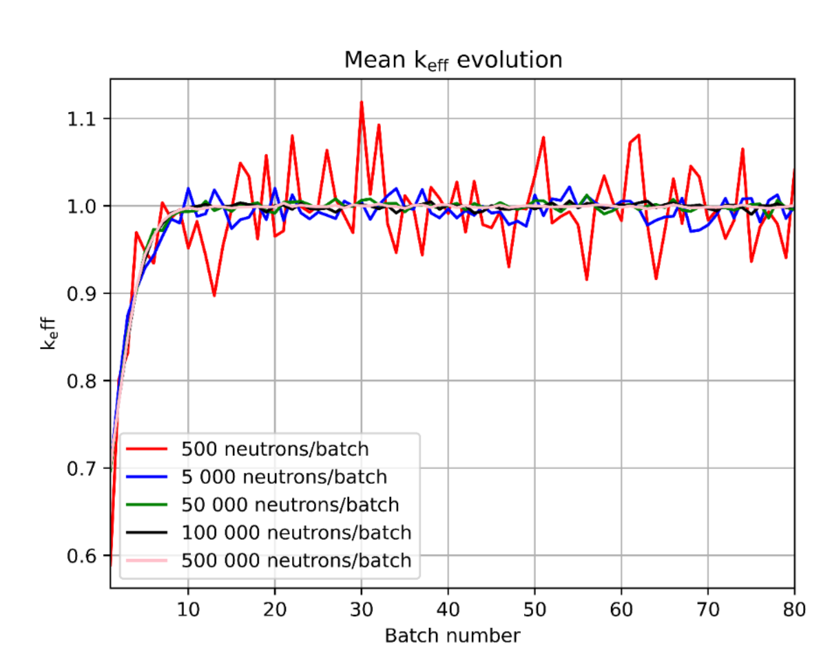
$$H(X) = -E[log_2 P(X)] = -\sum_{i=1}^{N} P_i . log_2(P_i)$$

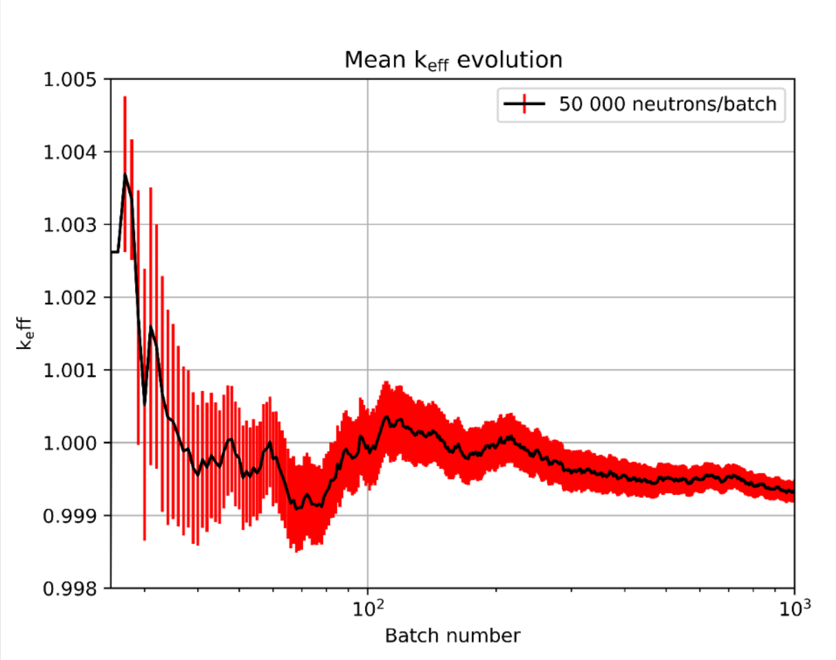
with X the discontinuous random variable describing N neutrons

- To quantify uncertainty level of neutron source distribution
- In Monte-Carlo calculations, neutron flux and reaction cross-sections are averaged over multiples calculations (batches) involving each N neutrons

-> Every batch must have similar Shannon entropy

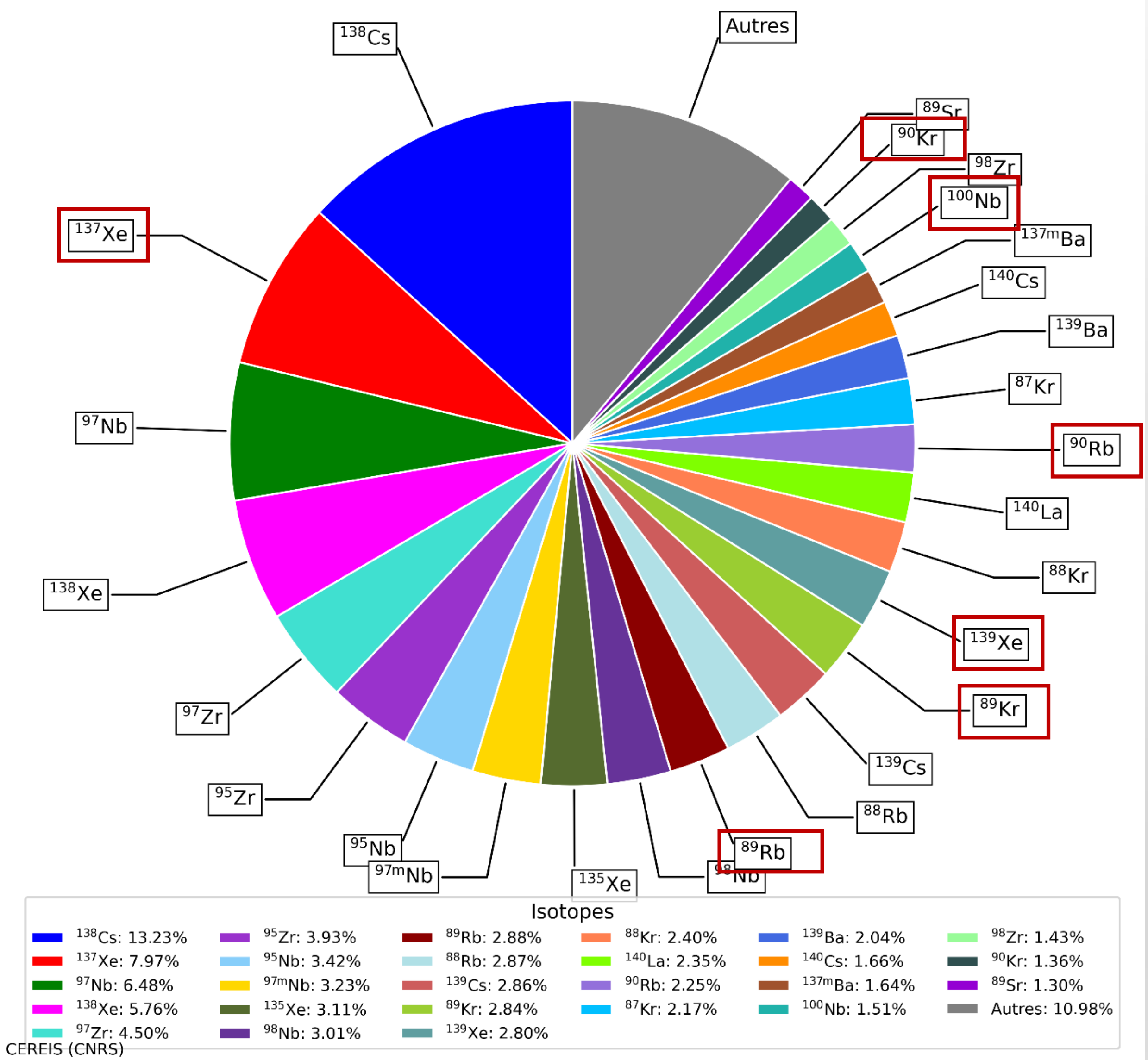






Parameters considered: 50 000 neutrons, 25 inactive cycles and 1000 active cycles

Appendix



Decay heat main contributors (> 0.2% of total DH) in extracted gaseous PF after 20 operations years and 1 second of cooling

- **At least 6 main contributors to DH are already identified and published Pandemonium nuclides**
- **They contribute to 13.64% of total DH !**
- **Are there other ones ?**

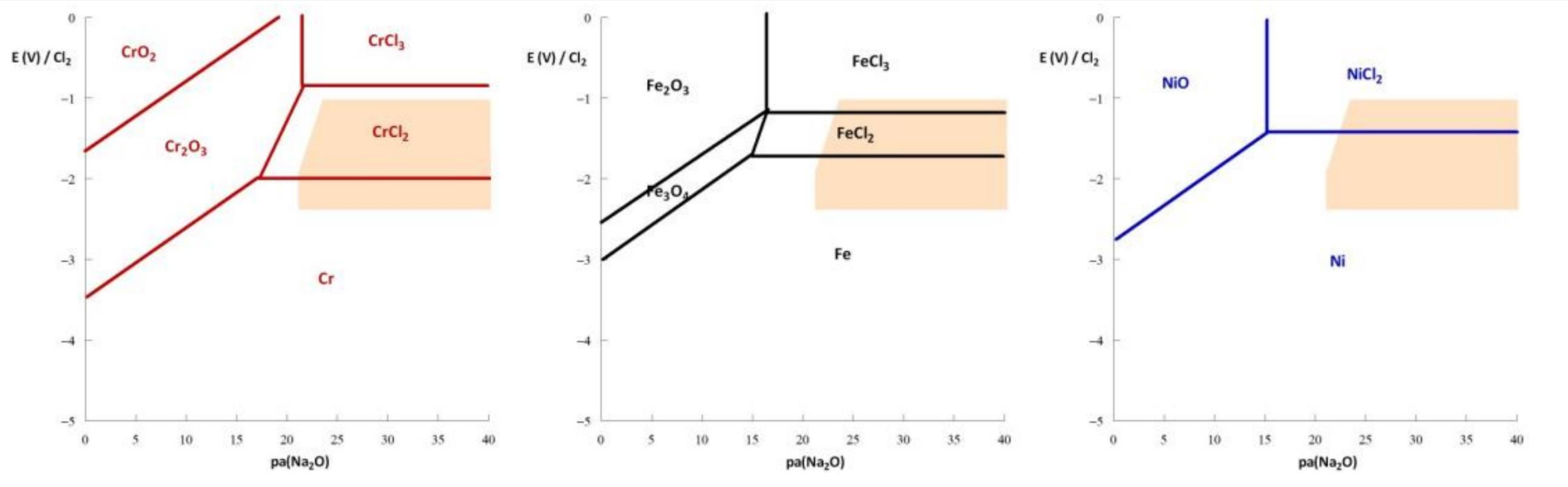
-> Among main decay heat contributors, apply criteria to identify potential Pandemonium nodes

-> Identifed nodes will be input for potential future TAGS measures

Appendix

Why calculating valencies?

- The valency of a chemical element is the maximal number of chemical bonds it can forms
- Valency is dependant of fuel salt RedOx potential
  - -> Valencies of some elements, from fuel salt or structural materials, could change
  - -> New chemical species could be formed **-> safety issues (corrosion, precipitation, gaseous species)**
- In current depletion calculations, no prediction of the chemical species formed in the fuel salt
  - **-> This internship is a "pioneer" work to implement chemical predictions modules into CEREIS**



Thermodynamic diagrams of Cr (left),and Fe (middle) and (right) in NaCl-based molten salt at salt at 600 °C.
The orange domain corresponds to the stability domain of the NaCl-UCl3-PuCl3 (80-10-10 mol%) mixture [SDel2024]

[SDel2024] S. Delpech et al., *Corrosion Mitigation in Molten Salt Environments*, Materials 2024, 17, 581. https://doi.org/10.3390/ma17030581