Radiochemistry and Monte Carlo integrated approach to radiological characterization for nuclear facilities decommissioning

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Outline of the presentation

i. INTRODUCTION

- Radiological characterization for Decommissioning
- L-54M reactor

ii. SCOPE OF THE WORK

iii. STEP 1

- Why MCNP?
- Model development
- Model Verification

iv. STEP 2

- Activation reactions
- Impurities distributions

v. STEP 3

- Assessment of activation
- Model Validation

vi. CONCLUSIONS AND FUTURE WORKS

After nuclear reactor shut-down

… several possible strategies

1. Immediate dismantling → ***Decon***
2. Postponed dismantling → ***Safe storage***
3. ***Entombment***

Technical, economical, social and political factors influence the strategy choice

PURPOSES:

1. Restoration of **unrestricted re-use condition** (greenfield/brownfield);
2. Reduction of **costs** and financial risk;
3. Reduction of **radiological/contamination risk** to operators/public/environment;

4. Optimization of **material recycling**;
5. Better resources exploitation

MAIN OPERATIONS

1. **Preliminary radiological characterization**
2. **Decontamination**
3. **Dismantling** and **Confinement**

In-situ characterization

α + β/γ monitor

Dose rate

Portable γ-spectrometry

In-lab characterization

Sample preparation and radiochemical separation

γ-spectrometry

LSC – α/β

α-spectrometry

L-54M Reactor and Milano city

POLITECNICO di MILANO

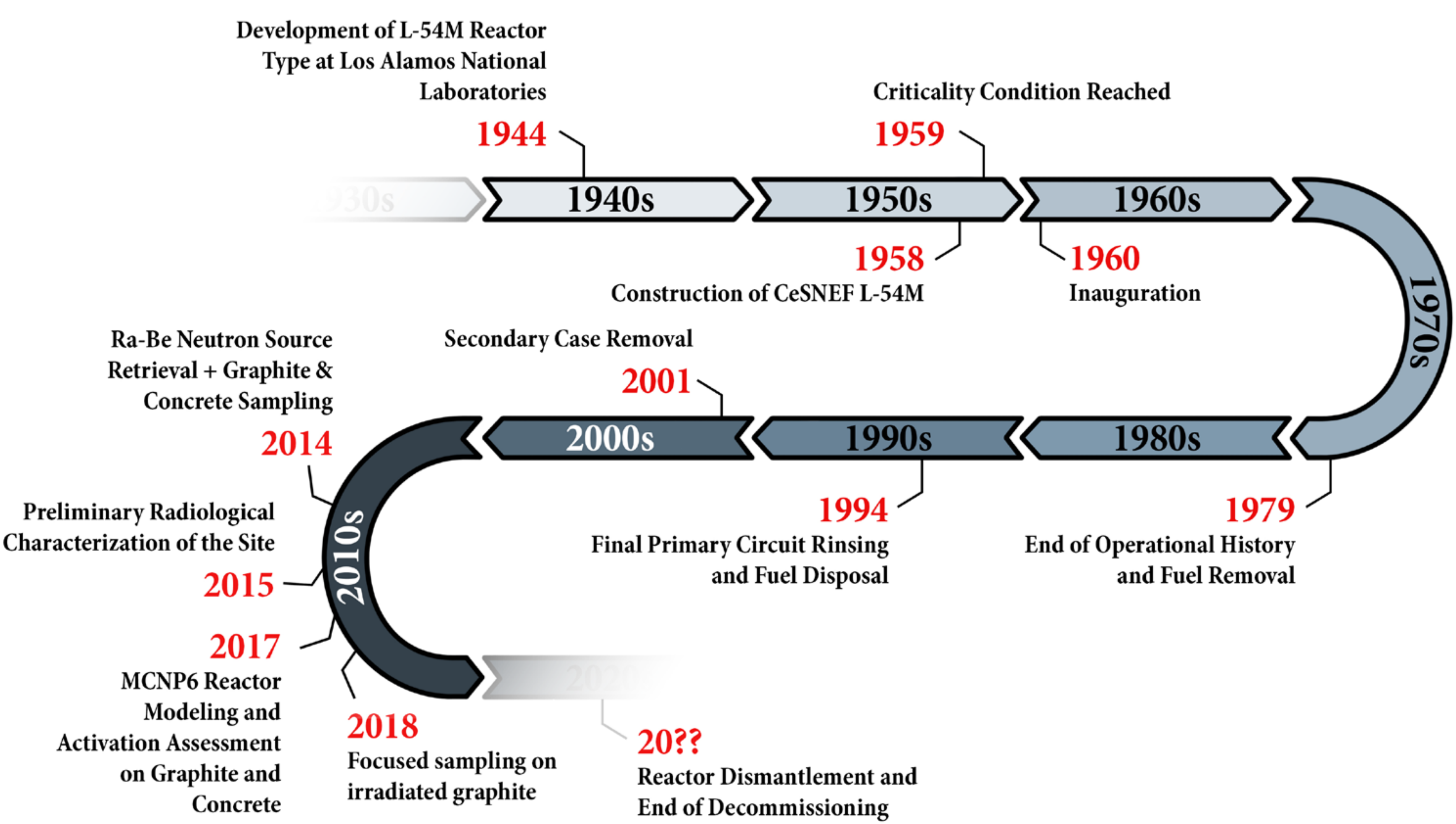
DEPARTMENT OF ENERGY

New B18 building with nuclear integrated labs

- Radiochemistry
- Radioprotection…

L-54M Reactor

L-54M Reactor history



Preliminary radiological characterization

SCOPE: 3D-computational assessment of neutron activated radionuclides distribution by means of **Monte Carlo N-Particles code** (MCNP6)

PURPOSE: cost savings and support to forthcoming characterization efforts

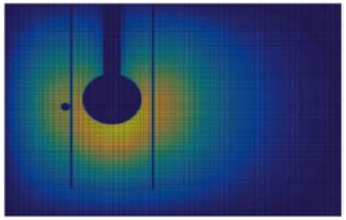
Input data → **1.** MCNP simulation of Nuclear Reactor

↕ Model **verification** with experimental values

- **Criticality**
- **Fluences**

Elemental composition → **2.** MCNP simulation of main nuclear reactions

- **Radionuclides of interest**
- **Main activation reactions**



3. MCNP simulation of graphite activation

↕ Model **validation** with experimental values

- **Activity concentrations** from radiological characterization

STEP 1: Why MCNP?

Input data

1. MCNP simulation of Nuclear Reactor

Model **verification** with experimental values

- **Criticality**
- **Fluences**

1. **MCNP** (**M**onte **C**arlo **N**-**P**article, version 6) transport code has been chosen to develop the Radiochemistry and Monte Carlo integrated approach
   - Well-established and validated;
   - Reliable;
   - Suitable to study thermal neutron diffusion and activation reaction rates;
   - Possible coupling with CAD

Los Alamos NATIONAL LABORATORY EST. 1943

T. Goorley et al. Nuclear Technology, 180(3):298 – 315, 2012

2. **ENDF** and **JEFF** cross-section libraries have been selected for the simulations

Retrieval of detailed and accurate data:

- **Geometry**: components dimensions and position;
- **Fuel**: volume, density, chemical composition;
- **Materials description**: density, chemical composition
- **Operational history**

Input data ←

From original vendor documents, blueprints, reports, previous master's thesis works…

1. MCNP simulation of Nuclear Reactor

↕

Model **verification** with experimental values

- **Criticality**
- **Fluences**

- Further analyses to bridge **knowledge gaps**
- Some uncertainties still holding: **sensitivity analysis**
- **Input script limitations** overlooked by substituting complicated geometries with simpler ones.

  → point-to-point **mass conservation** to guarantee best model accuracy

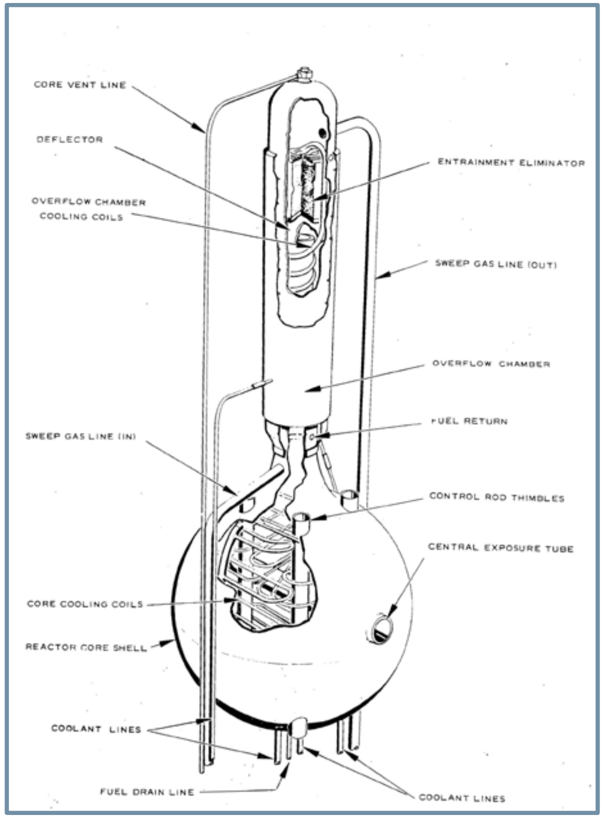
Example: L-54M model

Input data

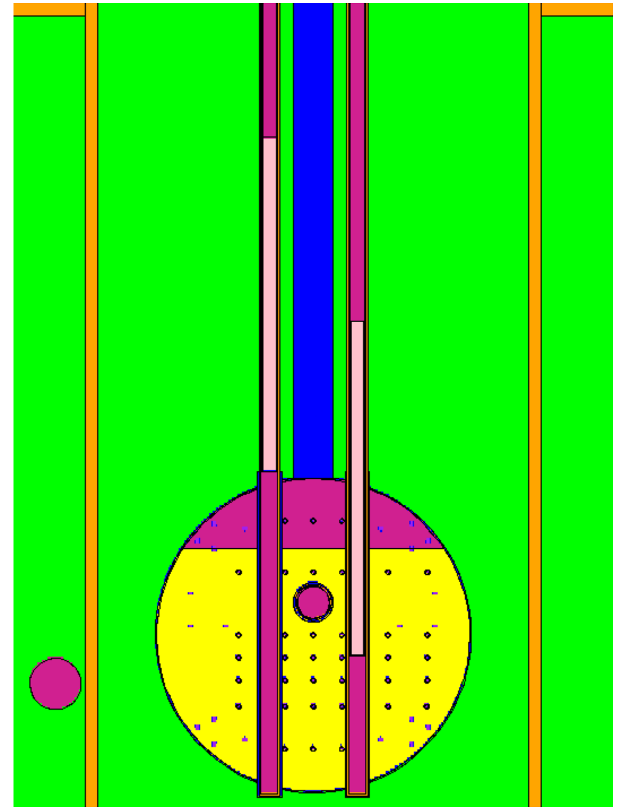
1. MCNP simulation of Nuclear Reactor

Model **verification** with experimental values

- **Criticality**
- **Fluences**



Reactor Core section



Implemented Core section

within IAEA collaborative research project on irradiated GRAphite Processing Approaches (GRAPA)

IAEA International Atomic Energy Agency Atoms for Peace · ROSATOM · ANDRA · SOGIN · NATIONAL NUCLEAR LABORATORY · MANCHESTER 1824 The University of Manchester · enresa

Input data

1. MCNP simulation of Nuclear Reactor

Model **verification** with experimental values

- **Criticality**
- **Fluences**

Criticality verification

KCODE card has been used for criticality simulations to determine neutron multiplication factor (k_{eff}) in a multiplying medium

Examples of criticality parameters:

- ***Subcritical reactivity*** → *control rods completely inserted*
- ***Supercritical reactivity*** → *control rods completely withdrawn*
- ***Total rod worth*** → *sum of Subcritical and Supercritical reactivity*
- ***Control rods inventory*** → *control rod inserted and others withdrawn*
- ***Control rod calibration*** → *control rod stepwisely moved from fully inserted to completely extracted position, with others withdrawn*

TMP card has been used with ***makxsf code*** to perform simulations at real working conditions (*i.e.* at non ambient temperature), thus considering temperature effect on cross sections and collision kinematics.

Fluences verification

F2 and ***F4 tallies*** have been used to determine the flux of particles with determined Energy crossing a surface or volume respectively.

F7 tally has been used to normalize the flux on the energy produced by fission events in the core region

$$\boldsymbol{\Phi_{norm}[cm^{-2}\cdot J^{-1}]} = \frac{F2\ or\ F4\ [cm^{-2}]}{F7\ [MeV\cdot g^{-1}]\cdot F7_{mass}\cdot (MeV\rightarrow J)} \xrightarrow{\cdot P[W]} \boldsymbol{\Phi_{norm}[cm^{-2}\cdot s^{-1}]}$$

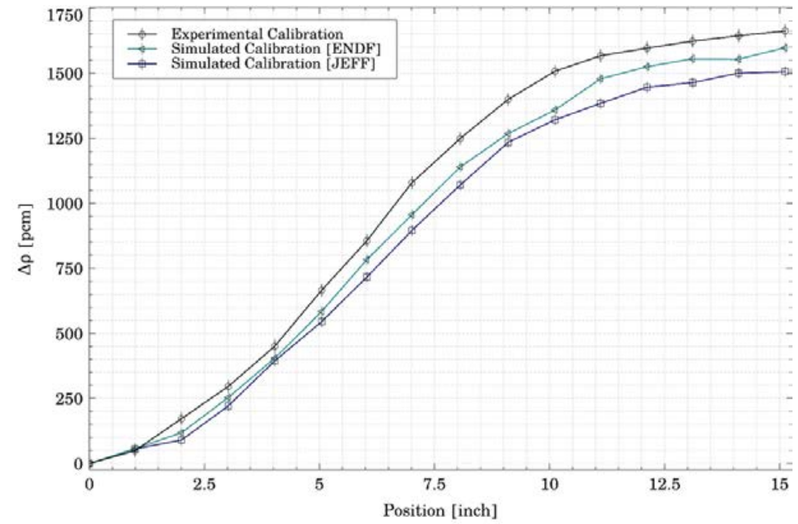
Input data

1. MCNP simulation of Nuclear Reactor

Model **verification** with experimental values

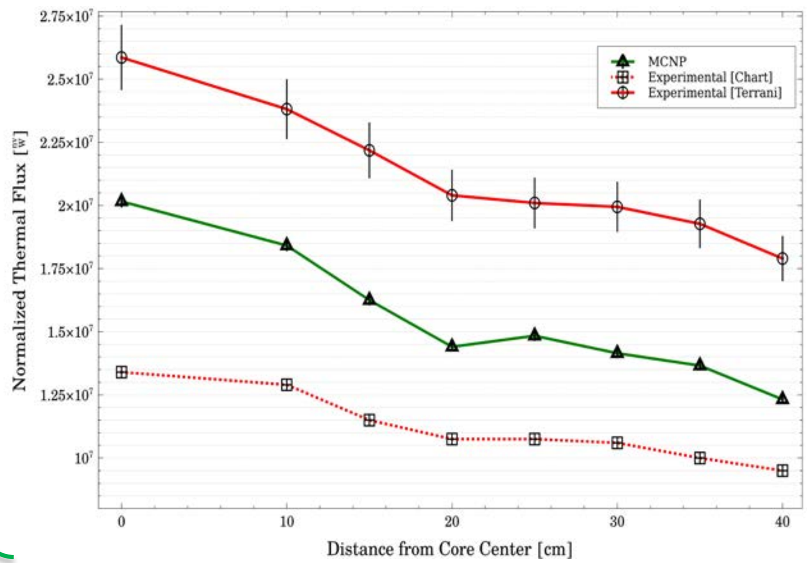
- **Criticality**
- **Fluences**

Example: L-54M model verification



Simulated vs Experimental control rod calibration

4th control rod simulated moving it from the fully inserted position to the fully extracted one with all other rods withdrawn.



Simulated vs Experimental thermal neutron flux in the central channel

The flux is normalized per energy deposited by fission events

STEP 2: Activation reactions

- **Elemental composition**

2. MCNP simulation of main nuclear reactions

- **Radionuclides of interest**
- **Main activation reactions**

Nuclide	Half-Life	Reactions
^{3}H	12.32 y	$^{6}Li\,(n,\alpha)\,^{3}H$ $^{7}Li\,(n,\alpha n)\,^{3}H$ $^{10}B\,(n,2\alpha)\,^{3}H$
^{14}C	5730 y	$^{13}C\,(n,\gamma)\,^{14}C$ $^{14}N\,(n,p)\,^{14}C$

$T_{1/2}$ coherent with reactor shut-down period

Main activation precursors to be included in the input file

- **Elemental composition**

2. MCNP simulation of main nuclear reactions

- **Radionuclides of interest**
- **Main activation reactions**

Example: L-54M simulated activation reactions

- **$T_{1/2}$ > 2y** since L-54M shut-down in 1979;
- **Scarce information in the literature about AGOT composition: Activation precursors** to be determined in virgin graphite

Nuclide	Half-Life	Reactions
^{3}H	12.32 y	^{6}Li (n,α) ^{3}H ^{7}Li $(n,\alpha n)$ ^{3}H ^{10}B $(n,2\alpha)$ ^{3}H
^{14}C	5730 y	^{13}C (n,γ) ^{14}C ^{14}N (n,p) ^{14}C
^{36}Cl	$3 \cdot 10^5$ y	^{35}Cl (n,γ) ^{36}Cl ^{39}K (n,α) ^{36}Cl ^{40}Ca $(n,\alpha p)$ ^{36}Cl
^{41}Ca	10^5 y	^{40}Ca (n,γ) ^{41}Ca
^{55}Fe	2.73 y	^{54}Fe (n,γ) ^{55}Fe ^{56}Fe $(n,2n)$ ^{55}Fe
^{59}Ni	$7.6 \cdot 10^4$ y	^{58}Ni (n,γ) ^{59}Ni ^{60}Ni $(n,2n)$ ^{59}Ni
^{60}Co	5.27 y	^{59}Co (n,γ) ^{60}Co

Nuclide	Half-Life	Reactions
^{59}Ni	$7.6 \cdot 10^4$ y	^{58}Ni (n,γ) ^{59}Ni ^{60}Ni $(n,2n)$ ^{59}Ni
^{60}Co	5.27 y	^{59}Co (n,γ) ^{60}Co
^{63}Ni	100 y	^{63}Cu (n,p) ^{63}Ni ^{62}Ni (n,γ) ^{63}Ni
^{93}Mo	4000 y	^{92}Mo (n,γ) ^{93}Mo
^{94}Nb	$2.03 \cdot 10^4$ y	^{93}Nb (n,γ) ^{94}Nb
^{99}Tc	$2.111 \cdot 10^5$ y	$^{98}\text{Mo}\,(n,\gamma)\,^{99}\text{Mo} \xrightarrow[2.74\text{ d}]{\beta^-} {}^{99m}\text{Tc} \xrightarrow[6.01\text{ h}]{\text{IT}} {}^{99}\text{Tc}$
^{108m}Ag	418 y	^{107}Ag (n,γ) ^{108m}Ag
^{152}Eu	13.54 y	^{151}Eu (n,γ) ^{152}Eu
^{154}Eu	8.59 y	^{153}Eu (n,γ) ^{154}Eu
^{233}U	$1.592 \cdot 10^5$ y	$^{232}\text{Th}\,(n,\gamma)\,^{233}\text{Th} \xrightarrow[21.8\text{ m}]{\beta^-} {}^{233}\text{Pa} \xrightarrow[26.97\text{ d}]{\beta^-} {}^{233}\text{U}$
^{239}Pu	24110 y	$^{238}\text{U}\,(n,\gamma)\,^{239}\text{U} \xrightarrow[23.5\text{ m}]{\beta^-} {}^{239}\text{Np} \xrightarrow[2.36\text{ d}]{\beta^-} {}^{239}\text{Pu}$
fission products	-	^{235}U (n,f); ^{238}U (n,f); ^{232}Th (n,f)

- **Elemental composition** → **Development of dedicated ICP-MS procedure**

i. **Oxidation** of graphite powder in a muffle at 650°C for 24 h;
ii. **Multistep digestion** of ashes by mixture of acids (HNO_3, HCl, HF);
iii. **Dilution** by 1% HNO_3;
iv. **ICP-MS** analysis.

2. MCNP simulation of main nuclear reactions

- **Radionuclides of interest**
- **Main activation reactions**

Element	ppm
Li	**0.0194** ± 2.7%
Al	**1.18** ± 2.9%
K	**1.22** ± 11%
Ca	**7.97** ± 9.8%
Ti	**0.622** ± 4.8%
V	**22.1** ± 1.1%
Fe	**2.81** ± 3.2%
Ni	**1.00** ± 1.3%
Cu	**0.208** ± 1.6%
Mo	**0.144** ± 1.3%
Ba	**0.0595** ± 1.9%

Element	ppb
B	***< LOQ*** *(13.9 ppb)*
Co	***9.3*** *± 1.5%*
Nb	***5.0*** *± 8.0%*
Ag	***< LOQ*** *(0.71 ppb)*
Cs	***0.47*** *± 4.8%*
La	***2.8*** *± 2.9%*
Ce	***3.2*** *± 4.6%*
Nd	***0.38*** *± 29%*
Eu	***0.55*** *± 4.0%*
Th	***886 ppt*** *± 5.0 %*
U	***66 ppt*** *± 33%*

Under definition collaboration to determine light and volatile elements (such as Li, B, N, Cl) by **Prompt-Gamma Neutron Activation Analysis** (PGNAA).

STEP 3: Assessment of Activation

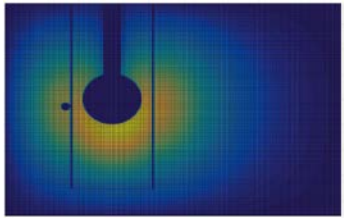
The **Reaction Rate** could be obtained by:

$$\boldsymbol{RR[atom \cdot cm^{-3} \cdot J^{-1}]} = C \cdot \iiint d^3r \int dE \cdot \boldsymbol{\Phi_{norm}(E,\vec{r})} \cdot \Sigma(\boldsymbol{E})$$

where:

- the **precursor nuclide density**, C[atom·barn^{-1}·cm^{-1}], is a normalization factor;
- the **cross-sections** of activation reaction , Σ[barn], is a response function.

Decay from production



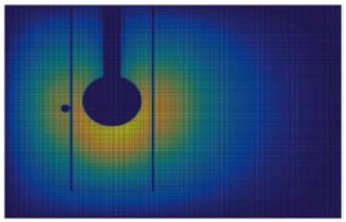
$$A[Bq/g] = \frac{\lambda\,[s^{-1}]}{\rho[g \cdot cm^{-3}]} \sum_{month} RR[atoms \cdot cm^{-3} \cdot J^{-1}] * Energy(i)[J] * e^{-\lambda \Delta t(i)}$$

Monthly energy production

3. MCNP simulation of graphite activation

Model **validation** with experimental values

- **Activity concentrations** from radiological characterization



3. MCNP simulation of graphite activation

Model **validation** with experimental values

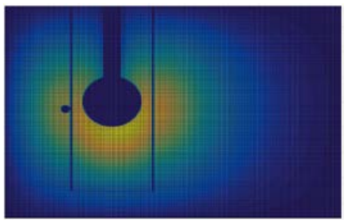
- **Activity concentrations** from radiological characterization

Example: L-54M pre-characterization

- Extraction of the graphite block;
- Drilling of graphite samples at several radial distances;

- **Gamma spectrometry**;
- Sample pre-treatment by **pyrolysis** and **^{3}H and ^{14}C trapping** forfollowing **Liquid Scintillation Counting** quantification

Radionuclide	0 cm	80 cm	120 cm
^{3}H [Bq·g^{-1}]	9170 ± 597	811 ± 54	51.5 ± 3.4
^{14}C [Bq·g^{-1}]	370 ± 25	37.5 ± 2.6	4.1 ± 0.3
^{152}Eu [Bq·g^{-1}]	253.9 ± 18.8	34.3 ± 2.3	4.1 ± 0.3



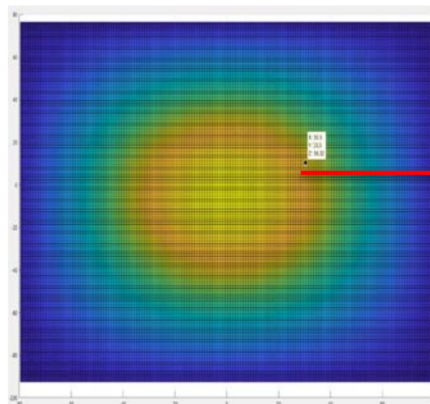
3. MCNP simulation of graphite activation

Model **validation** with experimental values

- **Activity concentrations** from radiological characterization

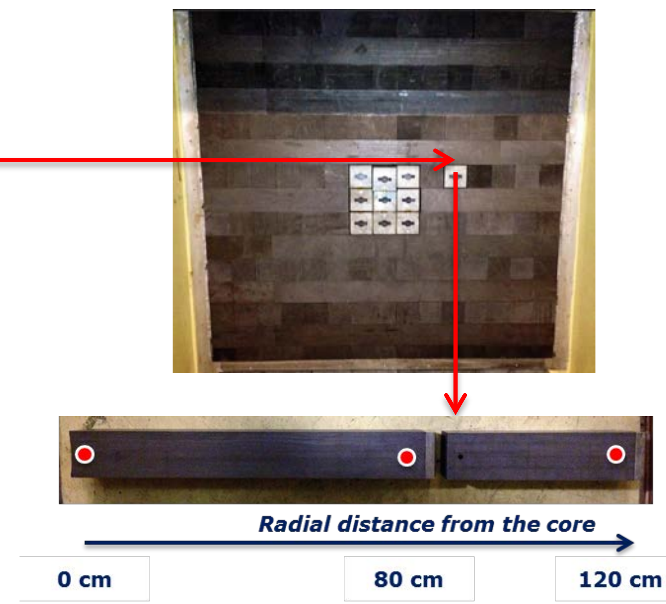
Example: L-54M model validation

^{152}Eu neutron activation (MCNP6)
(about 0.6 ppb Eu in virgin graphite)

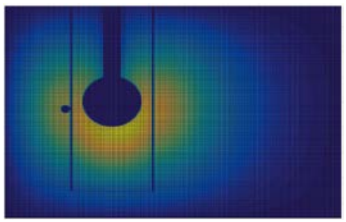


2D slice at 80 cm

Comparison with few **experimental data** available



^{152}Eu [Bq·g^{-1}]	0 cm	80 cm	120 cm
MCNP Simulation	207 ± 1	21.5 ± 0.2	1.8 ± 0.1
Experiment	253.9 ± 18.8	34.3 ± 2.3	4.1 ± 0.3



3. MCNP simulation of graphite activation

Model **validation** with experimental values

- **Activity concentrations** from radiological characterization

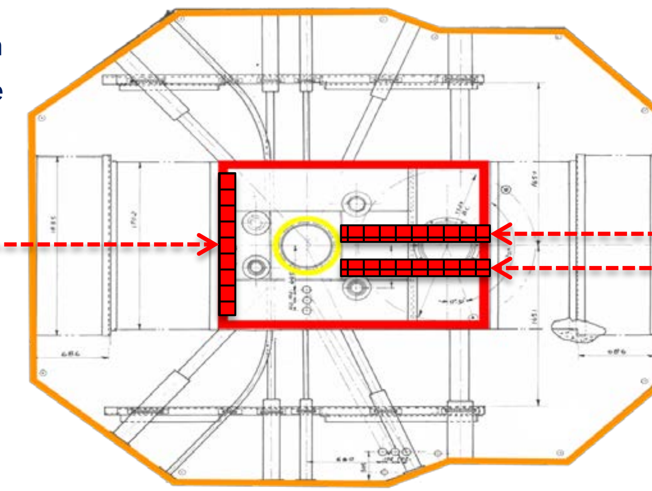
Example: further L-54M model validation

WEST side

- **7** samples on external graphite surface

EAST side

- **5** samples along 2 graphite rods



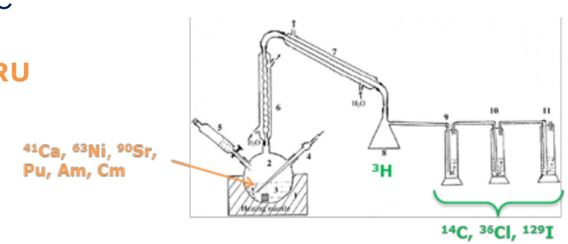
- **γ emitting radionuclides by HPGe**
- **Pure β and α emitting radionuclides**
  - **^{3}H, ^{14}C, ^{36}Cl, ^{129}I (volatiles)**

    Pyrolisis + radiochemical separation + LSC
  - **^{3}H, ^{14}C, ^{36}Cl, ^{129}I, ^{41}Ca, ^{63}Ni, ^{90}Sr, TRU**

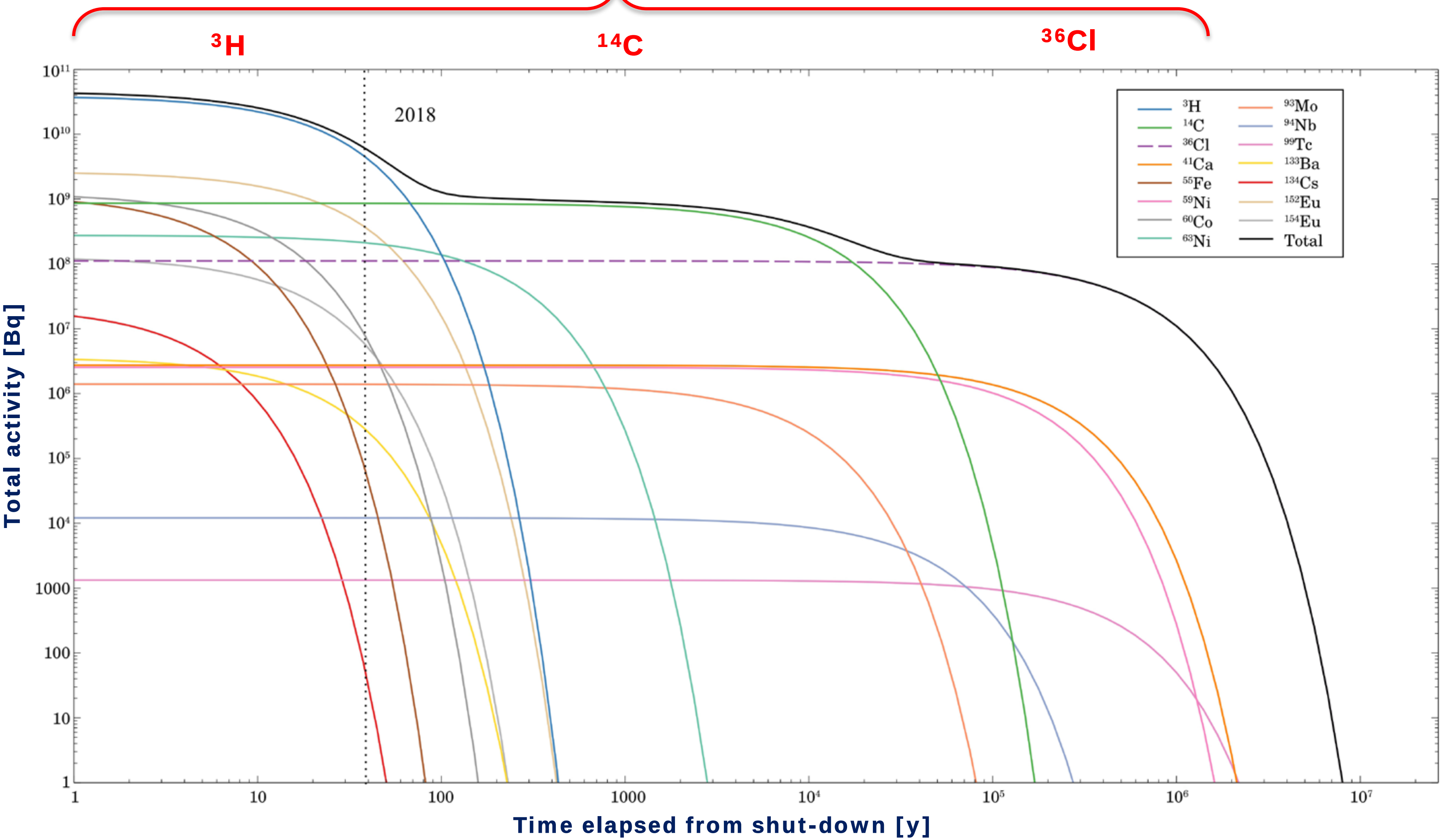
    Acid digestion +

    radiochemical separation +

    LSC/alpha spectrometry



Main contributors to Total Activity of L-54M graphite



3D-Model of Reactor Graphite Activation

1. The integrated computational approach prompted to be a **valid support to radiological characterization campaign**

   In fact, L-54M neutronic model

   1. **proficiently developed**,
   2. **verified** by comparing simulated and experimental criticality and fluences data;
   3. **validated** by comparing simulated and experimental radiometric data.

2. The approach is **general and applicable to other nuclear facilities**

Future works on L-54M reactor model

- **Light and volatile impurities** determination in virgin graphite by PGNAA and NAA.
- **Ultimate model validation** by additional radiometrical analyses on recently collected graphite samples: ^{3}H, ^{14}C, ^{36}Cl, ^{41}Ca, ^{63}Ni...

Any questions?

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