

IRSN

INSTITUT
DE RADIOPROTECTION
ET DE SÛRETÉ NUCLÉAIRE

Enhancing nuclear safety

Radiological characterization of in-pile irradiated graphite

Metrological problem, mathematical method and physical modelisation

ETSON

EUROPEAN
TECHNICAL SAFETY
ORGANISATIONS
NETWORK

INTERNATIONAL WORKSHOP

Treatment processes for the sustainable management of radioactive waste including innovative techniques

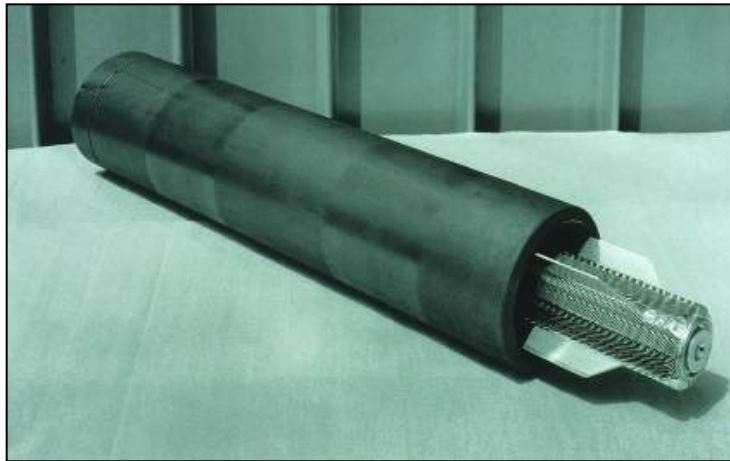
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4-6 JULY 2016
IAEA, Vienne (Austria)

- **UNGG reactor and graphite waste in France**
- **A metrological problem**
- **The mathematical reverse method proposed by EDF**
- **Modelisations performed at IRSN**
- **Perspectives**

UNGG reactor and graphite waste in France

Natural Uranium Graphite-moderated, Gas-cooled (air or CO₂)
20 years of operation



Fuel element bared by graphite sleeves

Credits @
ANDRA



Graphite stack of the moderator

Credits @
ANDRA

17000 t of irradiated graphite radwaste (6 reactors EDF 1963-1994)

A disposal design is under study in France, and needs radiochemical characterisations

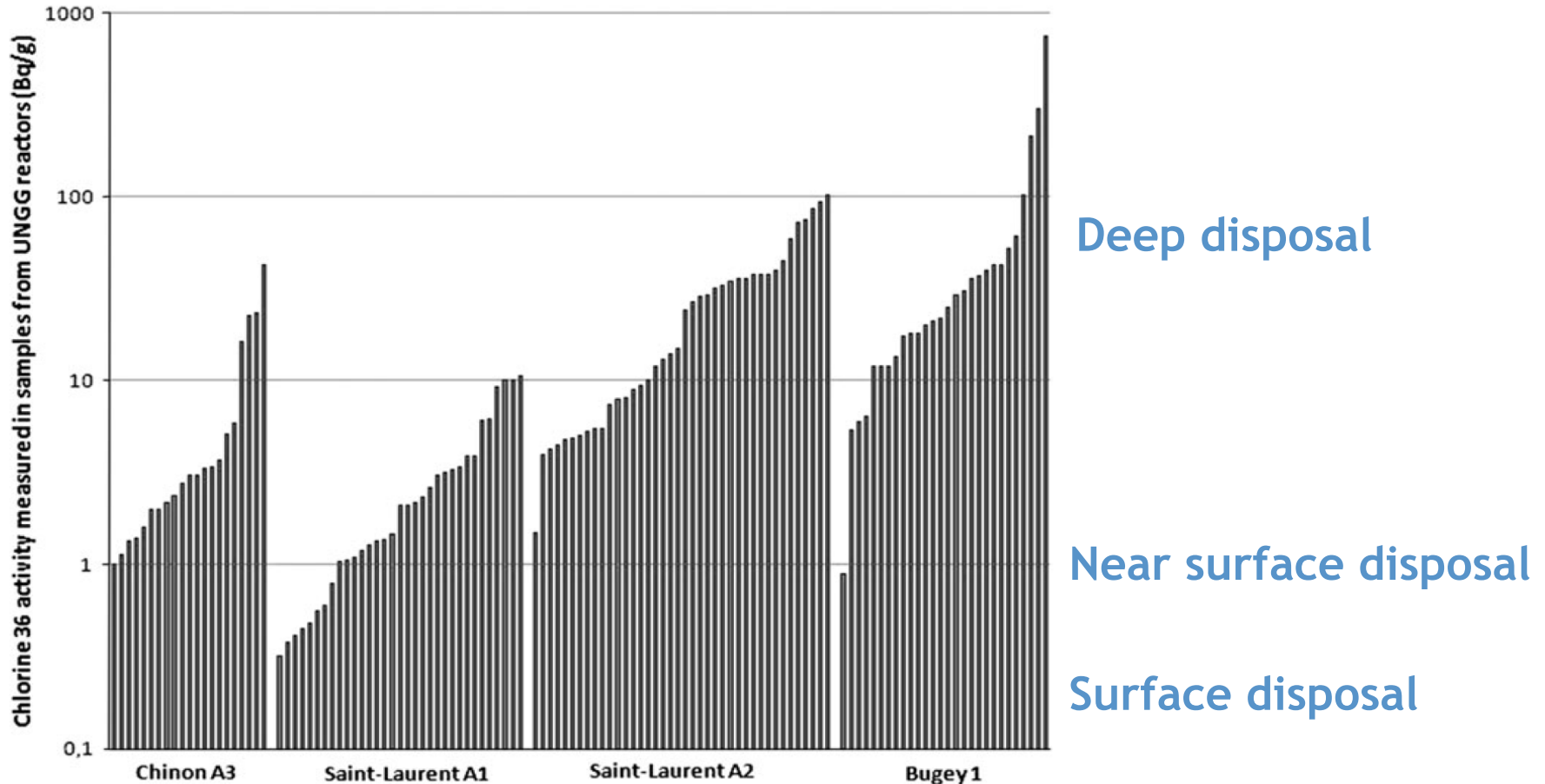
A metrological problem

- Activity measurements are highly dispersed for nearly every RN by 2-4 orders of magnitude

- Uncertainties do not correlate to:

- any in-pile physical parameter (neutron flux, temperature, gas flow...)
(<1 order of magnitude)
- nor the radiological measurement uncertainties (<1 order of magnitude)

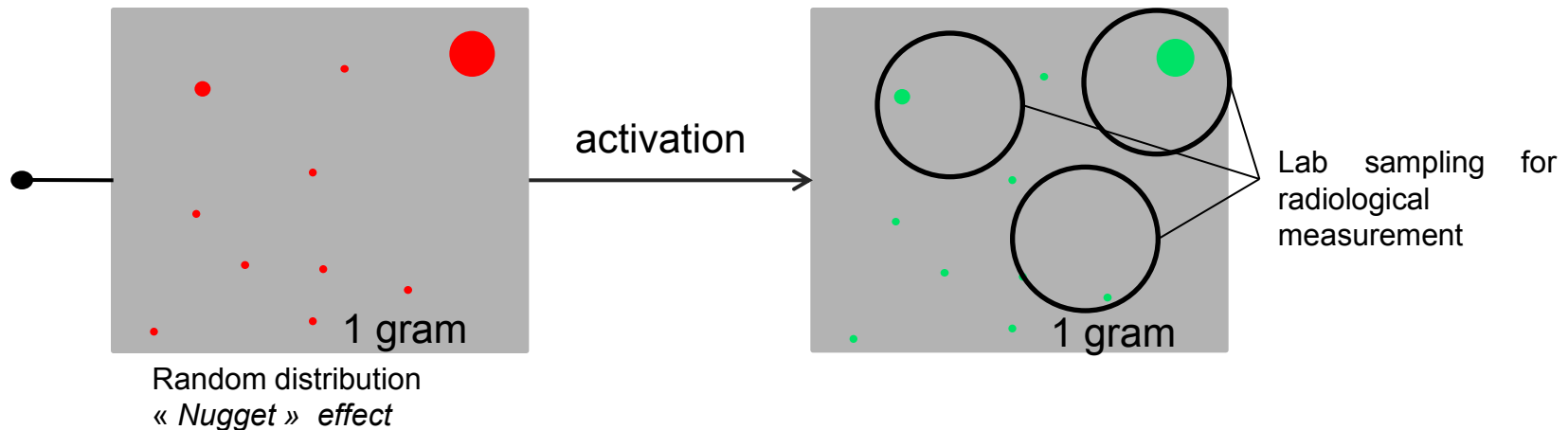
Illustration on ^{36}Cl



Disposal design?

The reason why

High purity (impurities are below 1 ppm) of graphite implies random distribution of impurities at small scale, with some *nugget effect*



Whatever dispersed, measurements reflect local reality, no one as to be eliminated

Combination of mathematical methods and physical studies

2014: A mathematical method has been proposed by EDF to handle uncertainties and help the disposal design

2015: It has been analysed by IRSN

Conclusion: the method is convenient in principles but it needs deeper studies to assess that the outcoming results are physically consistent.

2016: IRSN has started a research task on graphite irradiation modelisation

The reverse mathematical method

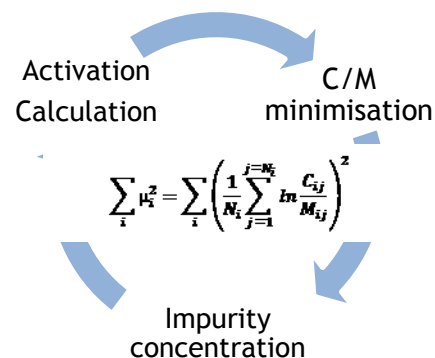
First step: identification/adjustment (reverse and iterative)

$$\begin{array}{c}
 \text{52 impurities} \\
 \xrightarrow{\hspace{1cm}} \\
 \left| \begin{array}{c} X_1 \\ \dots \\ X_k \\ \dots \\ X_{52} \end{array} \right|
 \end{array}
 \times
 \begin{array}{c}
 \left| \begin{array}{cccc}
 A_{1,1} & \dots & A_{1,k} & \dots & A_{1,52} \\
 \dots & & \dots & & \dots \\
 \dots & & \dots & & \dots \\
 A_{i,1} & \dots & A_{i,k} & \dots & A_{i,52} \\
 \dots & & \dots & & \dots \\
 \dots & & \dots & & \dots \\
 A_{144,1} & \dots & A_{144,k} & \dots & A_{144,52}
 \end{array} \right|
 \end{array}
 =
 \begin{array}{c}
 \xrightarrow{\hspace{1cm}} \\
 \left| \begin{array}{c}
 \text{144 RN} \\
 \text{RN}_1 \\
 \dots \\
 \dots \\
 \text{RN}_i \\
 \dots \\
 \dots \\
 \text{RN}_{144}
 \end{array} \right|
 \end{array}$$

Activation matrix

Identifies and quantifies impurity-radionuclide relation

Measure=1, ...,30



The iteration process C/M-1 is performed simultaneously on ~30 measurements

The reverse mathematical method

Second step: reactor scale activation calculation (direct)

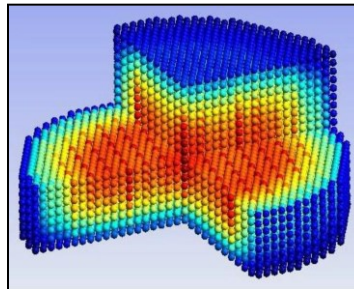
From step 1:

<Optimised impurity vector>
<Optimised activation matrix>

Reactor scale final
activation
calculation



Optimised
radiological
inventory



Neutronic distribution

IRSN analysis

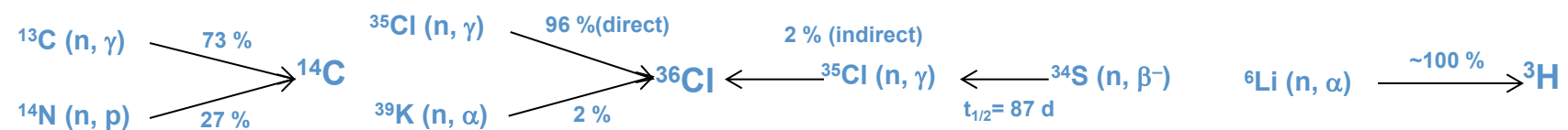
1- OPTIMISED IMPURITIES CONTENT

^{35}Cl (precursor of ^{36}Cl) is evaluated to 81 mg/t on Bugey 1 reactor

^{35}Cl (precursor of ^{36}Cl) is evaluated to 83 mg/t on Saint Laurent A2 reactor

This similarity is promising. Results to be benchmark (IRSN work with VESTA code)

2- OPTIMISED ACTIVATION MATRIX



These schemes are obtained at reactor scale at the end of ajustement process. They need to be compared from a reactor to another (further work) to assess that the method is not reactor-dependant.

IRSN analysis

Example of optimised activation matrix

Élément	ORIGINE EN POURCENTAGE DES 35 RN DEMANDES PAR ANDRA POUR L'EMPLEMENT DE BUGEY1 EN 2017																																					
	_H_3	BE_10	_C_14	CL_36	CO_60F	NI_59	NB_94F	NB_93M	CSI35F	EUJ54F	EUJ55	_I129	CSI37	_L234	_L235F	_L238	AM241	AM242M	BI247	CIV244	NP237	_L236	PL238	PL239	PL240	PL241	SR_90	ZR_93	TC_99F	MO_99F	_K_40	AG108M	CA_41	DY154	FR_81F			
LI	99																																					
C	1	64	73																																			
N	0	0	27																																			
CL	0	0	0	96	0																																	
CO	0				90	0																																
NI	0			0	9	100																																
NB	0						100	94																														
CS	0								97	0	0	0	0															0	0	0	0							
SM	0								0	20	23	0	0																									
EU	0								0	80	76	0	0																									
U	0	0	0	0	0	0	0	0	3	0	0	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	20	14	0							
TH	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0		
O	0	0	0																																			
FE	0			0	1	0																																
BA	0																																					
B	0	36	0																																			
K	0	0	0	2	0	0																																
CA	0	0	0	0	0	0																																
BR	0					0		0																														
ZR	0						0	0																					0	0	0	0						100
MO	0							6				0																	0	80	0	0						0
AG	0								0			0	0																0	0	85	100					0	
CD	0								0			0	0																0	0	0	0						0
DY	0								0	0	0		0																0	0	0	0						96
S	0	0	0	2																																		4
RU	0						0	0	0			0																	0	0	1	0						100
H	0																												0	0	0	0						0
BE	0	0	0																																			
NA	0	0	0	0																																		
AL	0	0	0	0																																		
SI	0	0	0	0																																		
SC	0			0	0	0																																
V	0			0	0	0																																
CR	0			0	0	0																																
MN	0			0	0	0																																
CU	0			0	0	0																																
ZN	0			0	0	0																																
MG	0	0	0	0																																		
AS	0				0	0		0	0																													
PD	0								0				0	0																								
IN	0								0			0	0																									
SN	0						0	0		0	0	0	0																									
SB	0								0	0	0	0	0																									
LA	0								0	0	0	0	0																									
CE	0								0	0	0	0	0																									
GD	0								0	0	0	0	0																									
HF	0																																					
TA	0																																					
W	0																																					
AU	0																																					
HG	0																																					

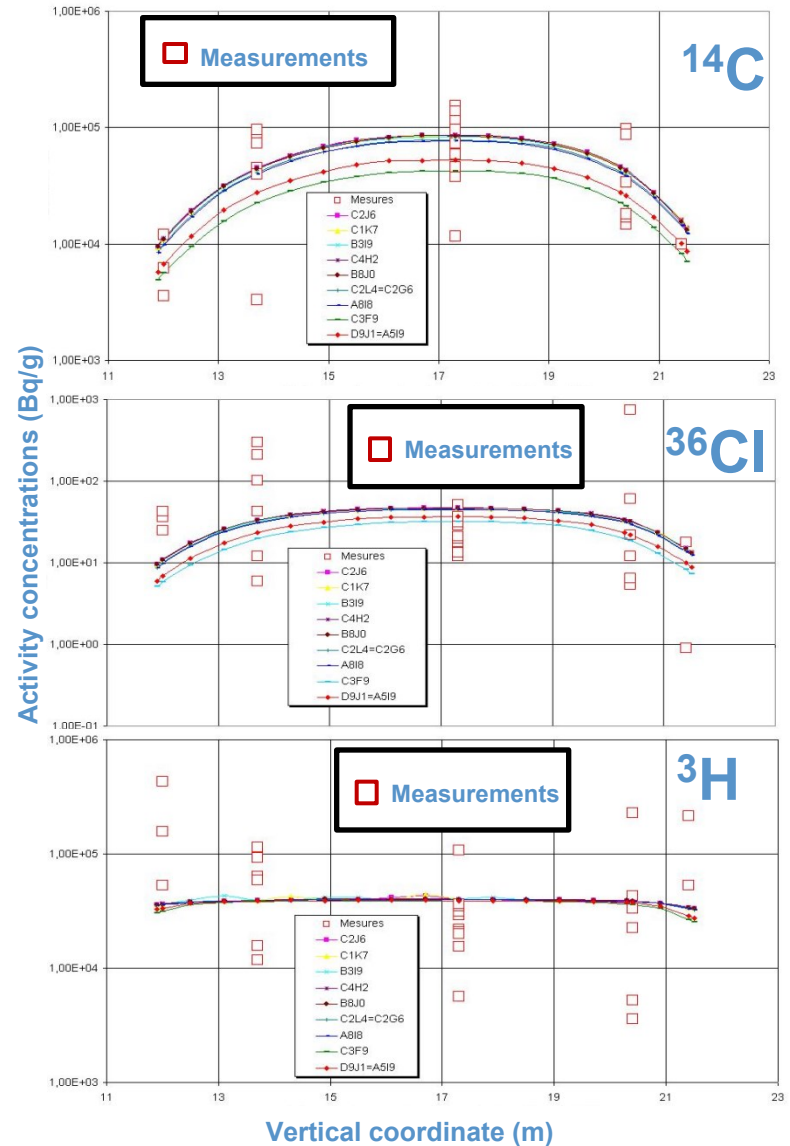
IRSN analysis

3- CALCULATED ACTIVITY CONCENTRATION PROFILE

IRSN has found out a correlation between the shape of neutron flux and the profile of calculated activity concentration taken into account the neutron capture cross section of the precursors:

- σ_{capt} of the $^{13}\text{C} (n, \gamma) ^{14}\text{C}$ reaction is 10^{-3} barn
- σ_{capt} of the $^{35}\text{Cl} (n, \gamma) ^{36}\text{Cl}$ reaction is 43 barns
- σ_{capt} of the $^6\text{Li} (n, \alpha) ^3\text{H}$ reaction is 940 barns

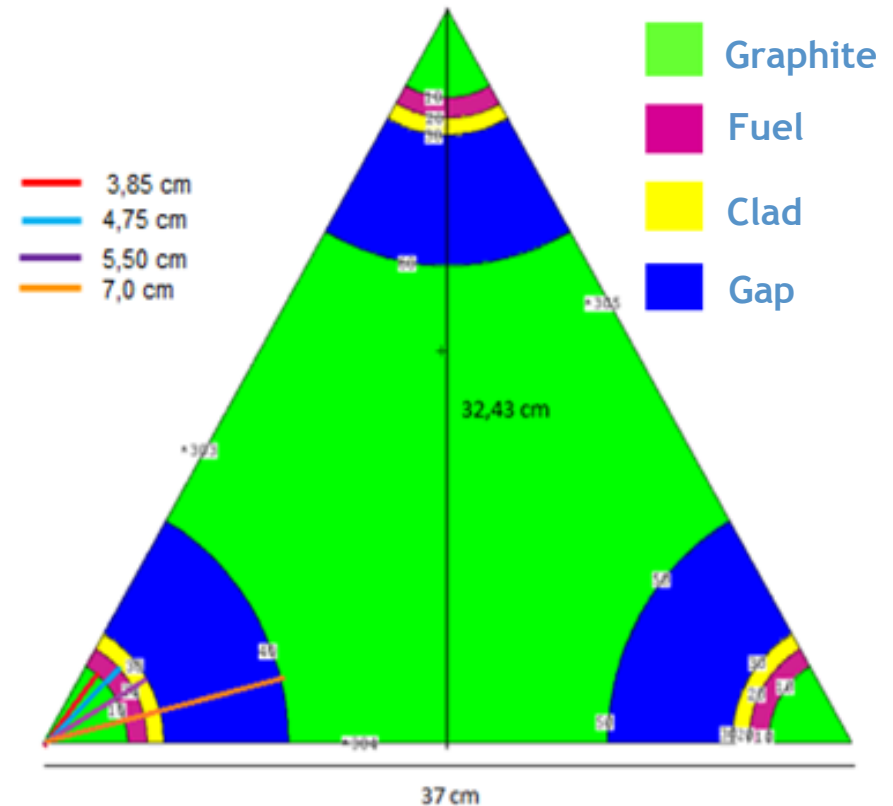
A very dependant to the neutron flux profile of ^{14}C activity concentration distribution, and a flat profile of ^3H (saturation) are very physical results regarding to σ_{capt}



IRSN research

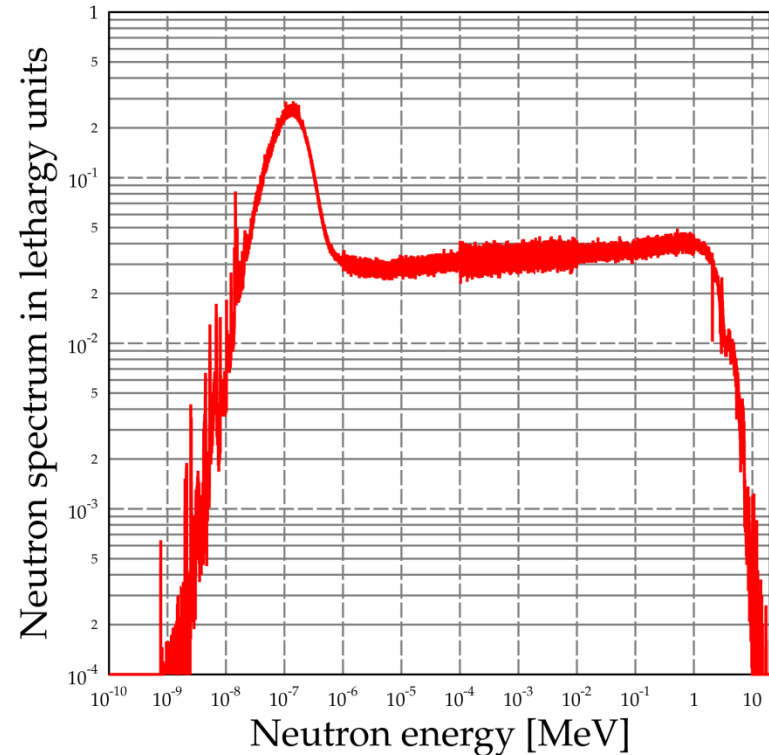
Use of VESTA code (MCNPX module + depletion module) to perform analytical studies on activation schemes

Fuel	^{238}U 99,3% ^{235}U 0,7%
Cooling fluid	CO_2
Graphite	^{12}C 98,88% ^{13}C 1,12%
Clad	Alliage Mg-Zr Mg 99,5% Zr 0,5%



IRSN research

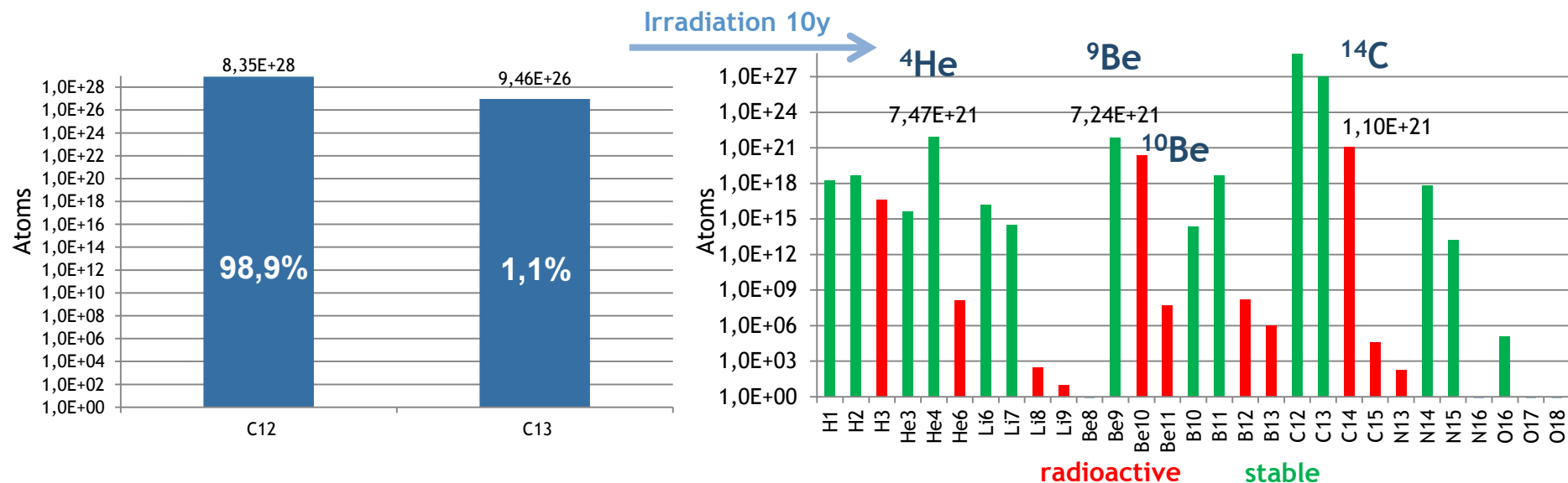
Calculated neutron energy spectrum (MCNP X module with 43 000 groups) for UNGG reactor



Test-1: $^{12}\text{C}+^{13}\text{C}$

<< Cooling >>=pure depletion, no neutronic flux

- First test: absolutely pure natural carbon irradiation (10 y) and cooling (10 000 y)
- 1m^3 (1685kg), $\phi=10^{13}\text{n/cm}^2/\text{s}$



Observations:

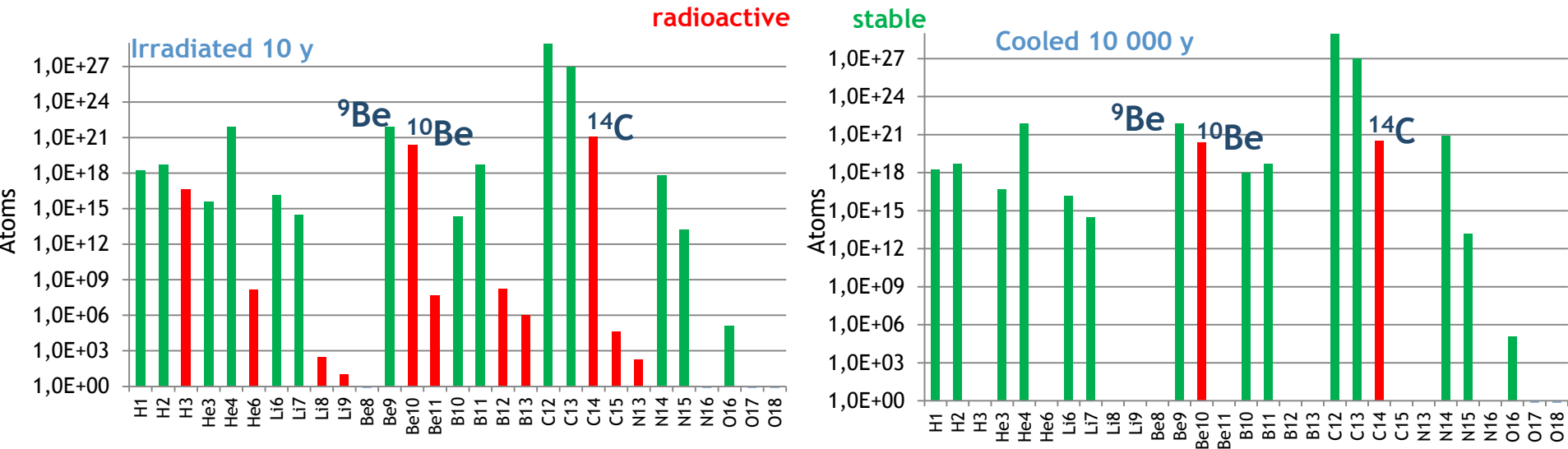
^{14}C is not the main product of $^{12}\text{C}+^{13}\text{C}$ activation, but stables ^4He and ^9Be .

^{14}C and ^{10}Be are the main radioactive products through activation process

Activation schemes are complex

Test-1: $^{12}\text{C}+^{13}\text{C}$

- Cooling phase



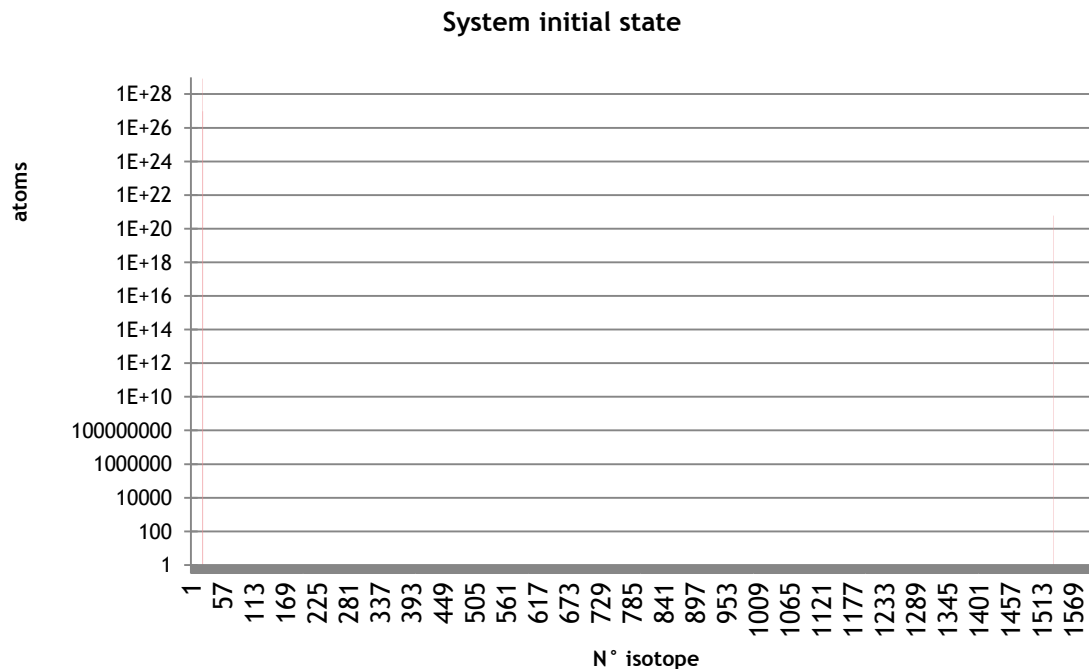
Observations:

At long term ^{14}C and ^{10}Be are the only radioactive RN coming from $^{12}\text{C}+^{13}\text{C}$ activation

Chemical aspects (stable isotopes): ^9Be is the main chemically active product overall. However, quantity created is low: 0,1 ppm

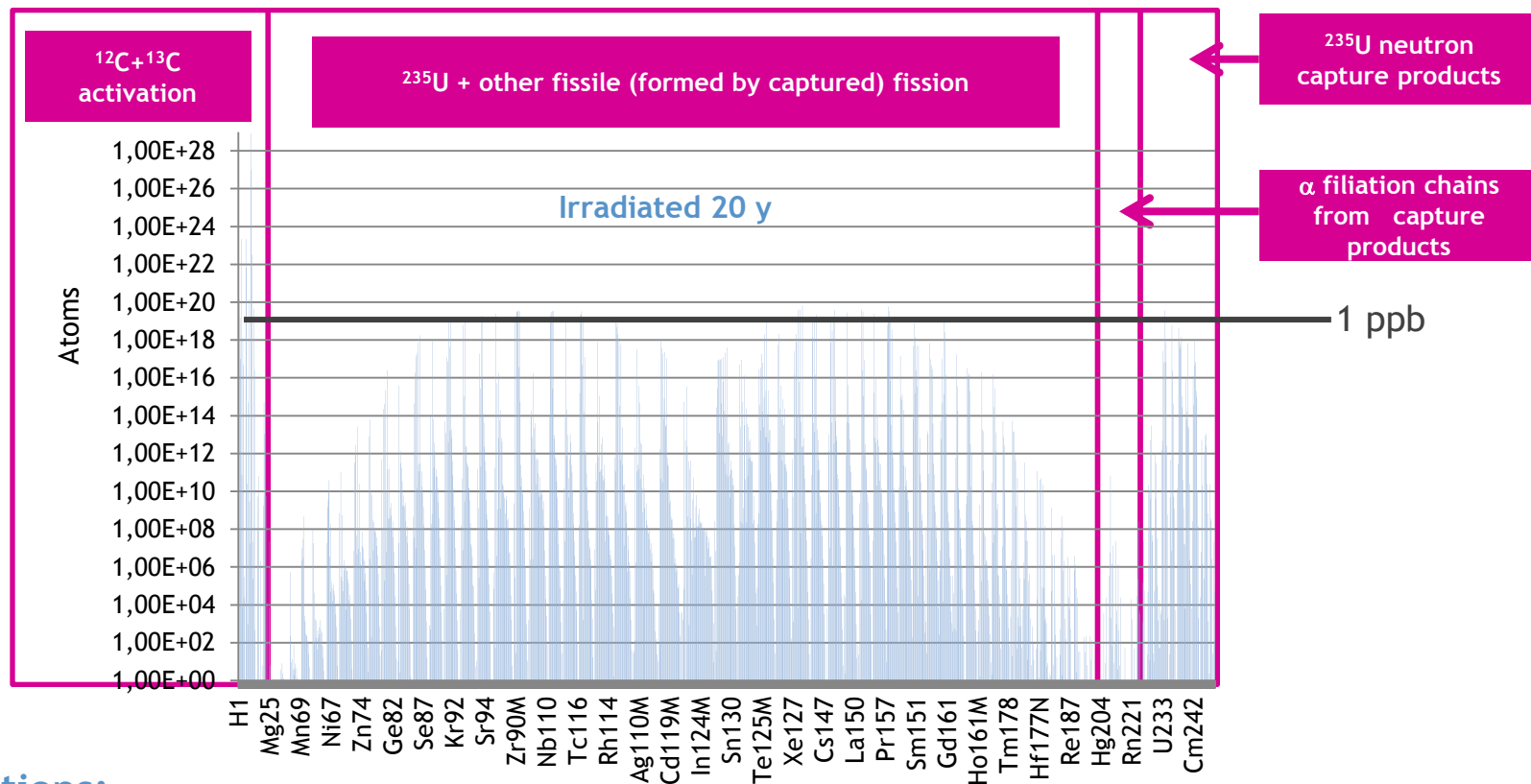
Test-2: $^{12}\text{C} + ^{13}\text{C} + \varepsilon \text{ } ^{235}\text{U}$

- **Second test: activation / fission**
- $1\text{m}^3(1685 \text{ kg})$, $\langle\phi\rangle \sim 10^{14}\text{n/cm}^2/\text{s}$, realistic in-pile irradiation sequences, 10^5y cooling
- pure nat. carbon+ 7,2 ppb ^{235}U (1 ppm nat. U, chemical detection limit hypothesis)



Test-2: $^{12}\text{C}+^{13}\text{C} + \varepsilon \text{ }^{235}\text{U}$

- Second test: activation / fission



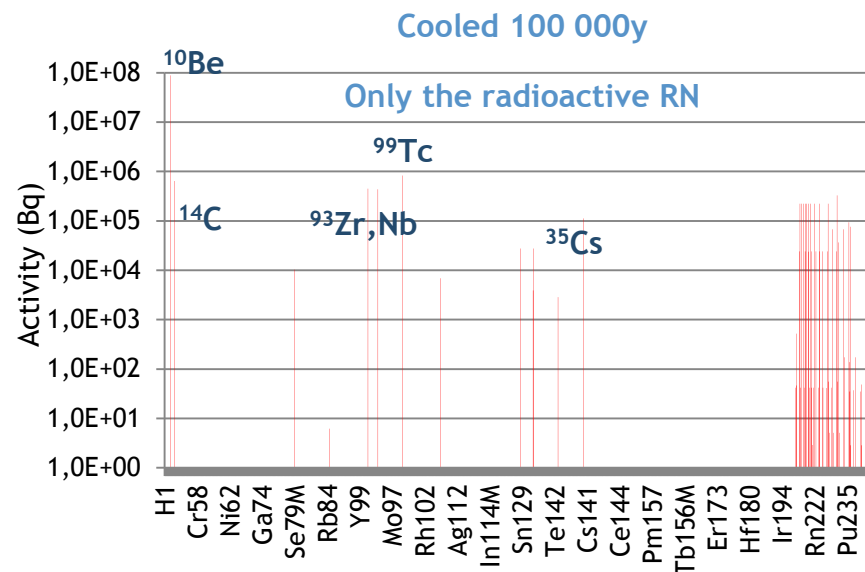
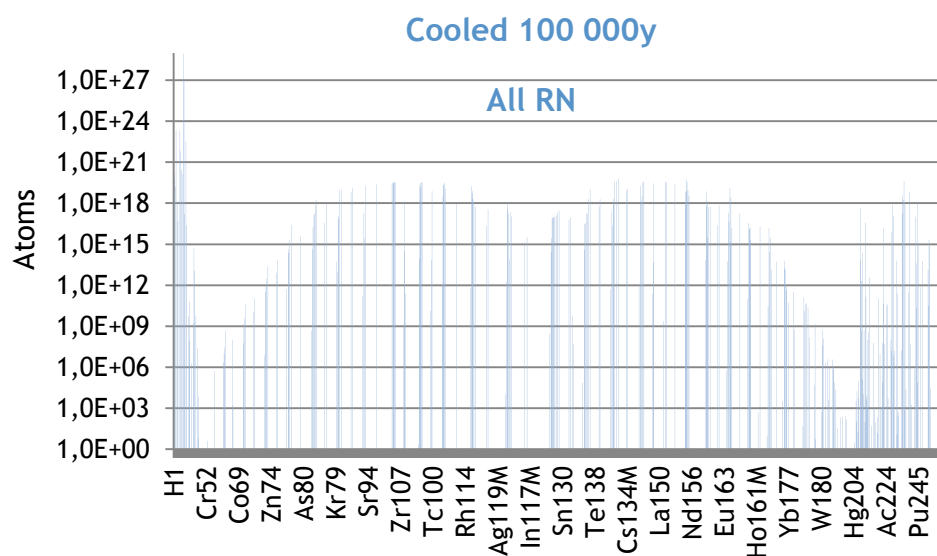
Observations:

High complexity: $3\text{RN} \rightarrow 1618 \text{RN}$, but decorrelation is OK

Max concentration of elements produced by fissions is $\sim 1\text{ppb}$ in graphite

Test-2: $^{12}\text{C} + ^{13}\text{C} + \varepsilon \text{ } ^{235}\text{U}$

- Second test: activation / fission



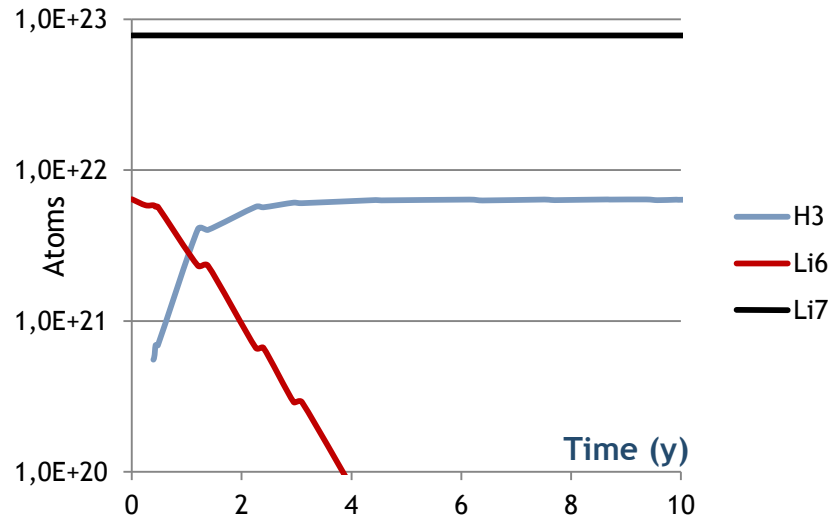
Observations:

^{10}Be remains the main radioactive component, ^9Be the main component (2,5 ppm)

Long term radiological inventory might not only be a matter of pure activation

Test-3: $^{12}\text{C}+^{13}\text{C}+ 1\text{ppm } (^6\text{Li}+^7\text{Li}) + \varepsilon \text{ } ^{235}\text{U}$

- Third test: kinetics of $^6\text{Li} (n, \alpha) ^3\text{H}$ reaction and saturation



Observations:

Saturation level of tritium is reached at 3 years of operation, a flat profile obtained par EDF clearly explained

Conclusions and further work

Preliminary conclusions

- ^{10}Be reveals to be the main holder of activity in irradiated high purity graphite at long term (100 000 y)
- ^9Be reveals to be the main stable element from by activation of carbon (2,5 ppm for 20 y of in pile irradiation)
- Other elements that can be created by fission are $\sim 1\text{ppb}$ for 7 ppb of fissile impurity

Further work

- Adding impurities step by step: Cl, Co, Ni, Sm, Eu...
- Building a realistic activation matrix and comparing to the optimised EDF one