FROM RESEARCH TO INDUSTRY

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APPLICATION OF LSC FOR ^{3}H, ^{14}C, ^{36}Cl, ^{41}Ca AND ^{63}Ni DETERMINATION IN VARIOUS MATRIX FROM NUCLEAR WASTE

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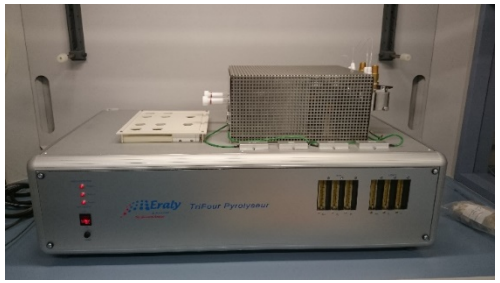
OUTLINE

Context of the study

Beta emitters isolation

^{3}H, ^{14}C in graphite and concrete
^{36}Cl, ^{63}Ni, ^{41}Ca



Matrices validation and / or proficiency test results

Conclusion and prospects

CONTEXT

- The National Radioactive Waste Management Agency (**ANDRA)** is in charge of the long-term management of radioactive waste in France.
  - Controlling the different types of wastes is essential for the waste management.

Various types of Nuclear wastes

Radionuclide	Acceptance limit ($Bq.g^{-1}$) for LLW
^{3}H	2×10^{5}
^{60}Co	**1.3×10^{8}**
^{137}Cs	3.3×10^{5}
^{14}C	9.2×10^{4}
^{36}Cl	**5**
^{63}Ni	**3.2×10^{6}**
^{55}Fe	**6.1×10^{9}**
Σ alpha-emitters	3.7×10^{3}

^{3}H : Half life 12,3 years
^{14}C : Half life 5700 years
^{36}Cl : Half life 3,02 10^{5} years
^{63}Ni : Half life 98,7 years
^{41}Ca : Half life 1 10^{5} years

For surface disposal :

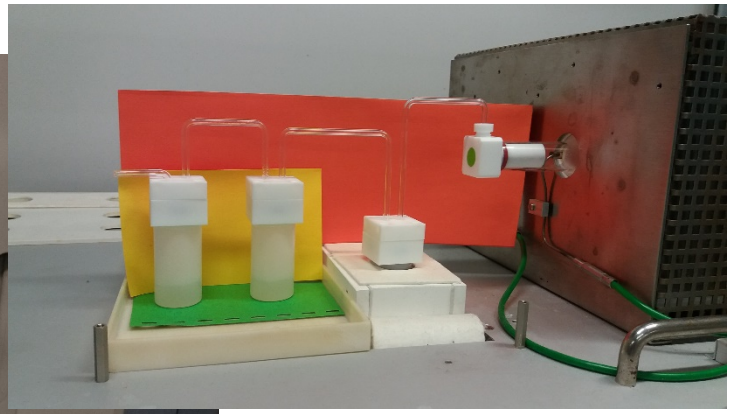
Maximum activity limit : depends of the RN

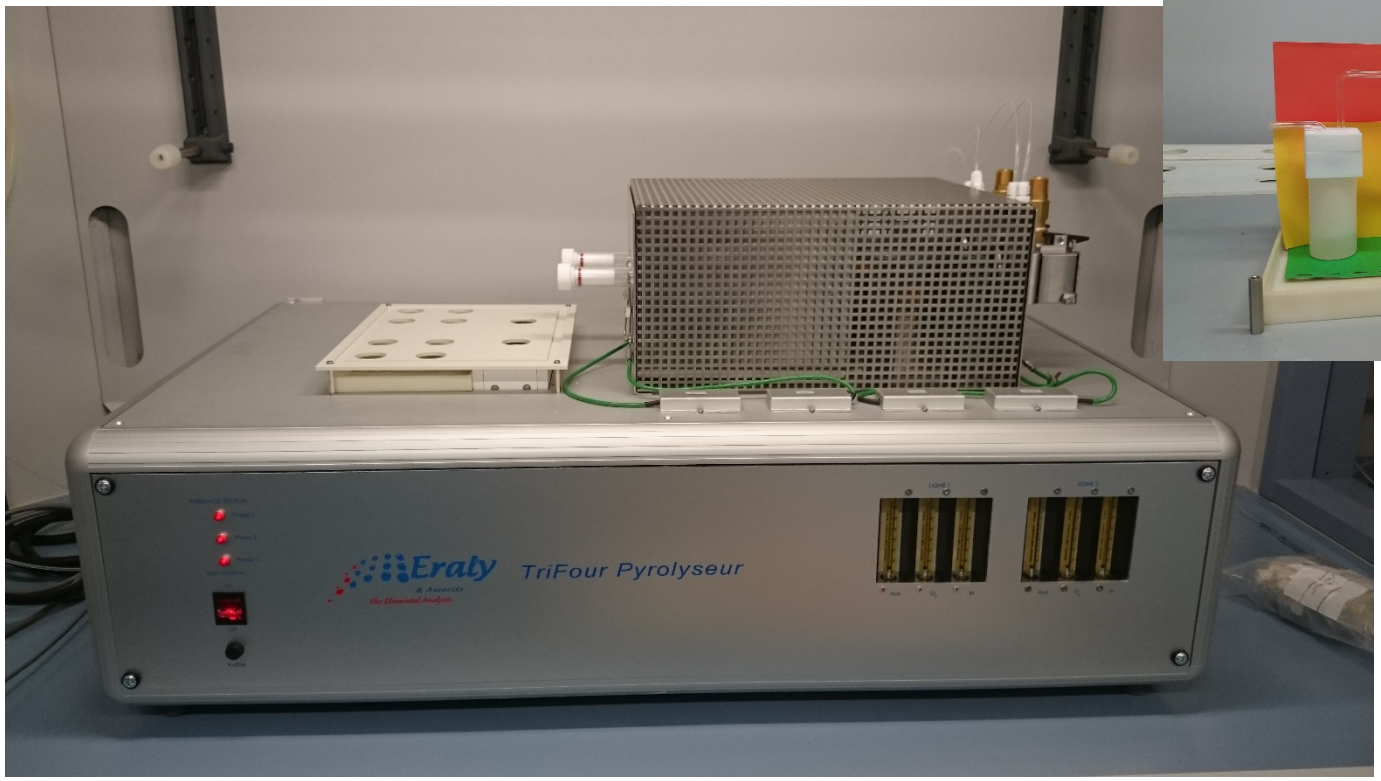
BETA EMITTERS SEPARATION : $^{3}H/^{14}C$

Beta emitters measurement : **Liquid Scintillation Counting**

→ Most widely used for isolated RN

RN need to be isolated first from the entire matrix





Pyrolysis method for ^{3}H and ^{14}C in all matrix

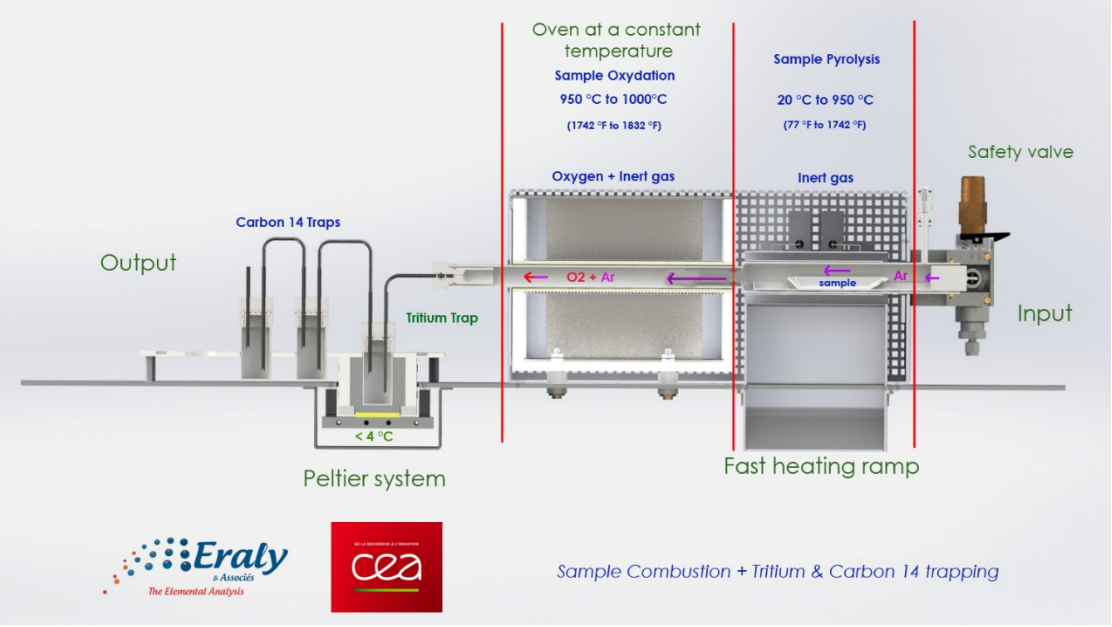
^{36}Cl in graphite

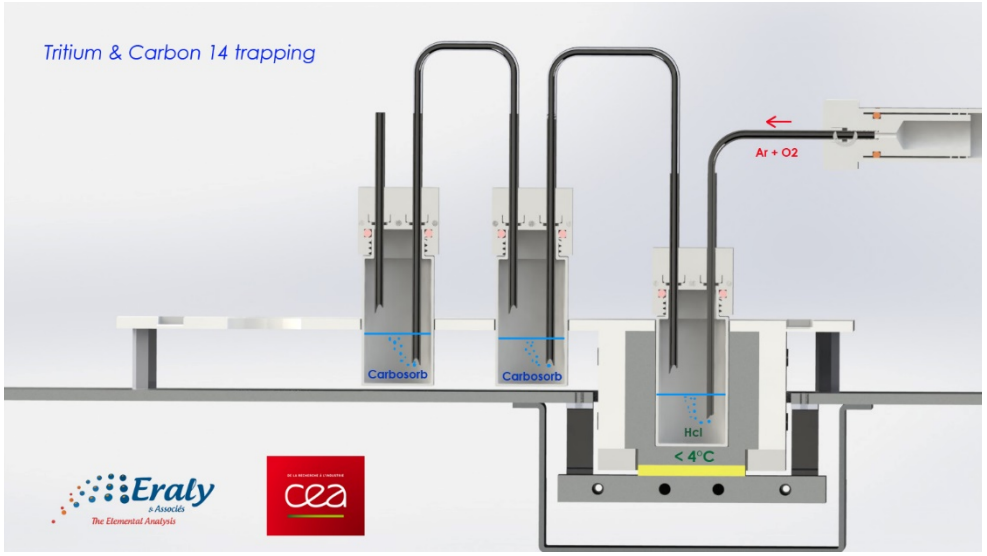
COMBUSTION METHOD FOR $^{3}H/^{14}C$ IN GRAPHITE

For graphite :

T° rising from amb to 950°C
in 20 min
Then 1H at 950 °C
Under O_2

COMBUSTION





^{3}H trapping : HCl 0.2M (2 traps)
^{14}C trapping : carbosorb (2 traps)

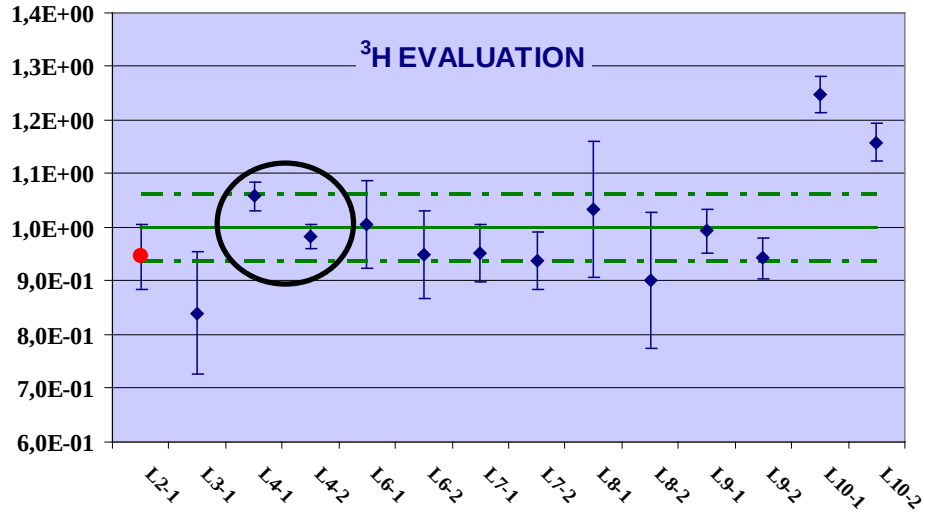
Sample boat empty at the end

→ Extraction yield : 100%

CARBOWASTE RESULTS FOR $^{3}H/^{14}C$ IN GRAPHITE

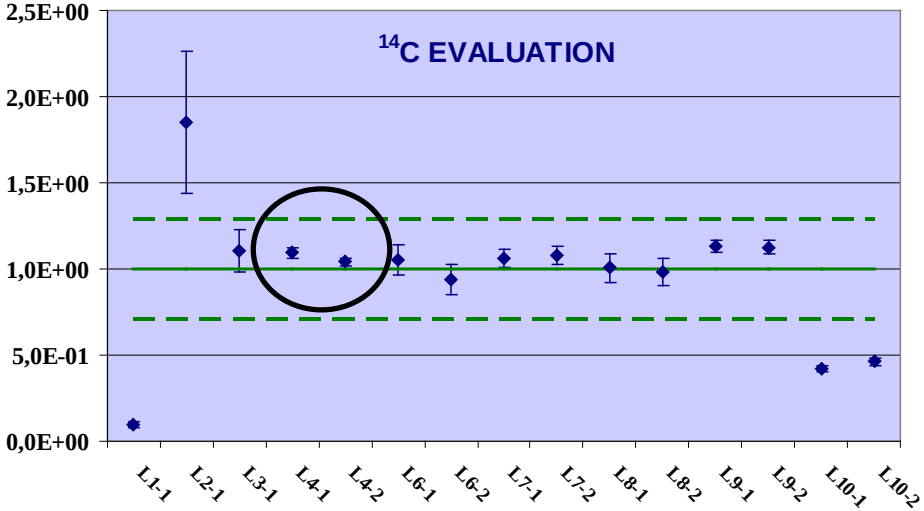
Carbowaste : 5 g of irradiated graphite segmented 2 + 2 (+ 1) with 2 determinations

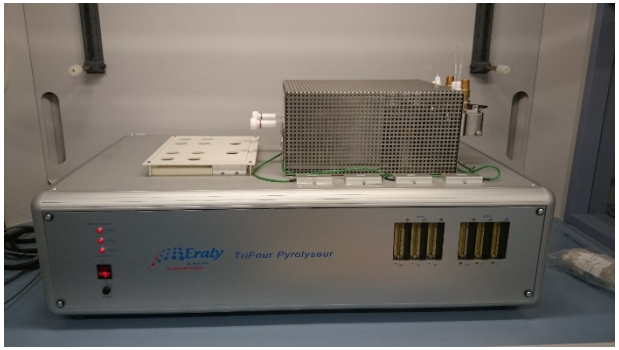
Addition of ^{3}H activity obtained in the 2 first traps, more than 95% in trap 1
Addition of ^{14}C activity obtained in the 2 last traps, more than 95% in trap 3



^{3}H activity : 83,320 Bq/g ± 5%

^{14}C activity : 21,310 Bq/g ± 5%





Very good agreement

MATRIX VALIDATION : CONCRETE

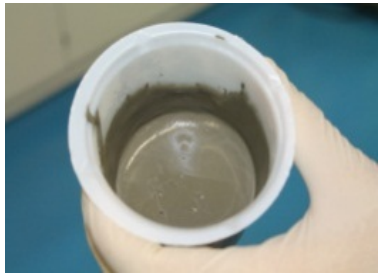
Methodology :

Use of labelled H-3 and C-14 concrete

optimization of temperature rising program for maximum tritium recovery by comparison with calculation from concrete preparation

Preparation of labelled concrete :

Use of H-3 glucose and C-14 glucose in H_2O



Cement added with water and mixed

activity check deposited in vessel

Drying of concrete for at least 3 weeks

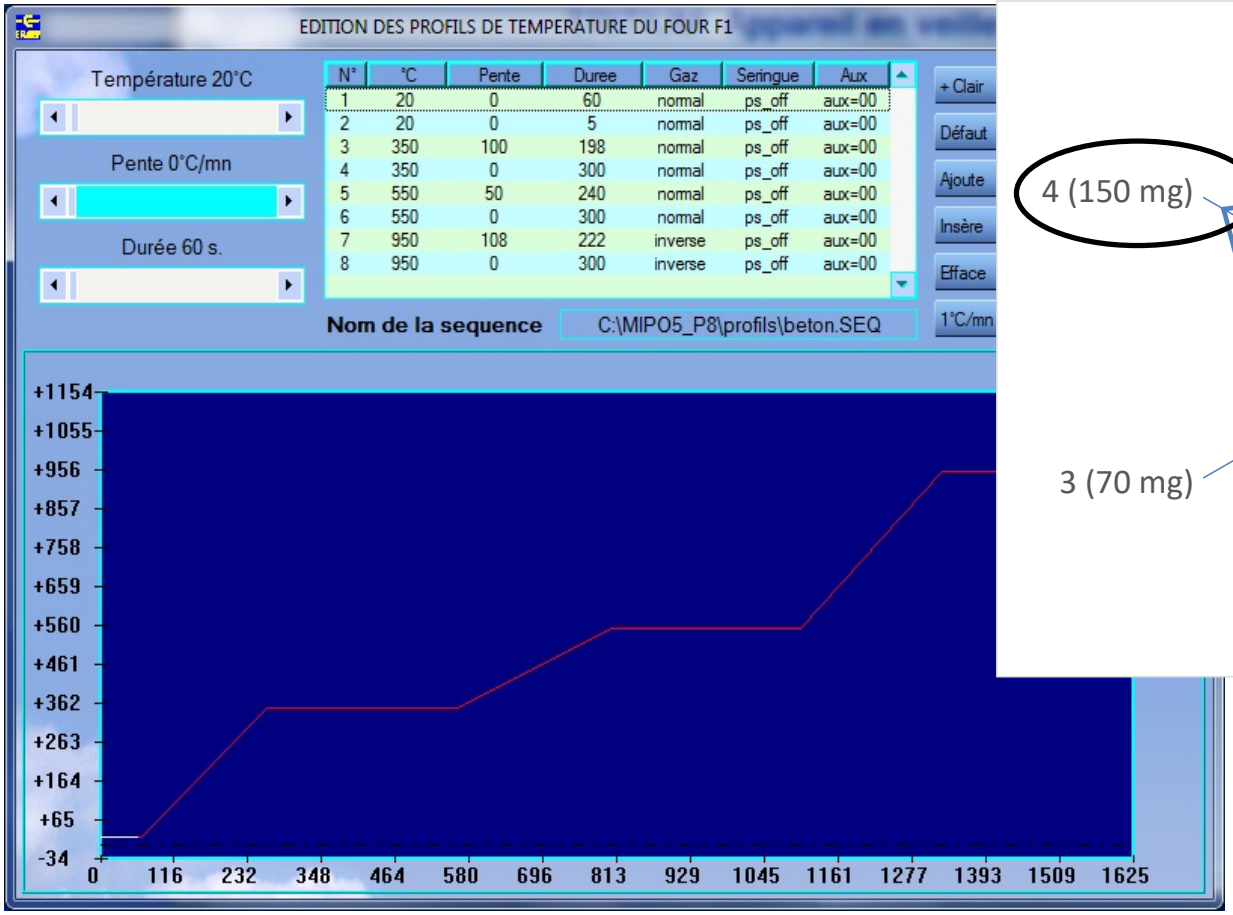
Weighing of concrete and calculation of H-3 and C-14 activity

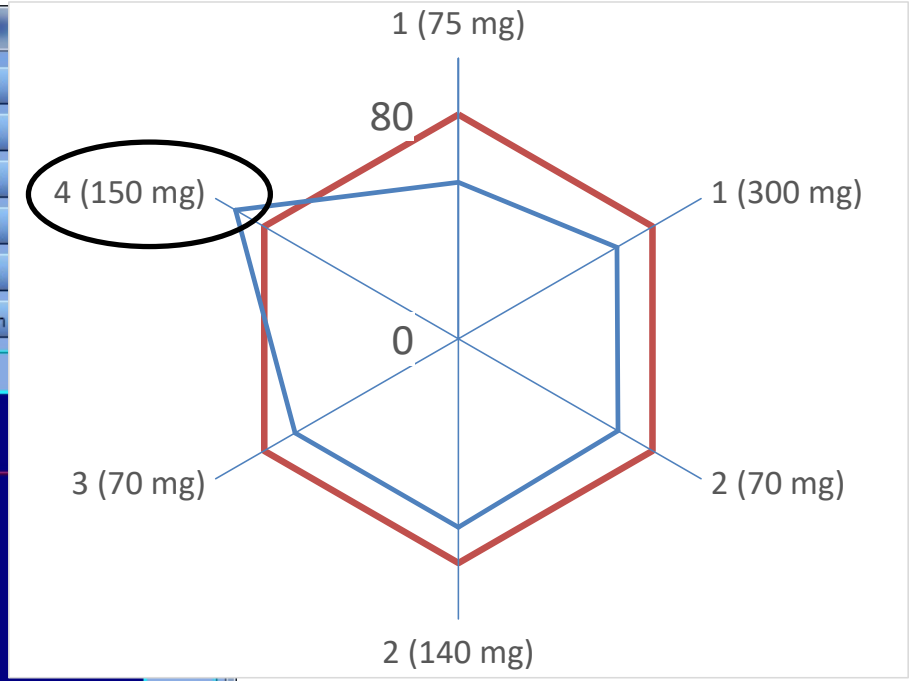
concrete Grinding to obtain powder

MATRIX VALIDATION : CONCRETE

To optimize temperature program, use of fine powder (< 250 µm obtained after sieving)

Different temperature program and gas flow tested :



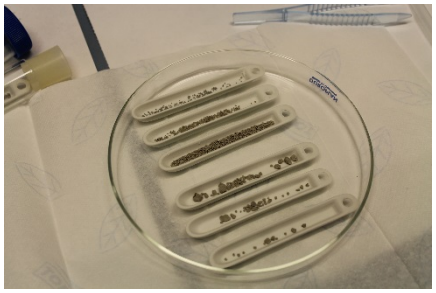


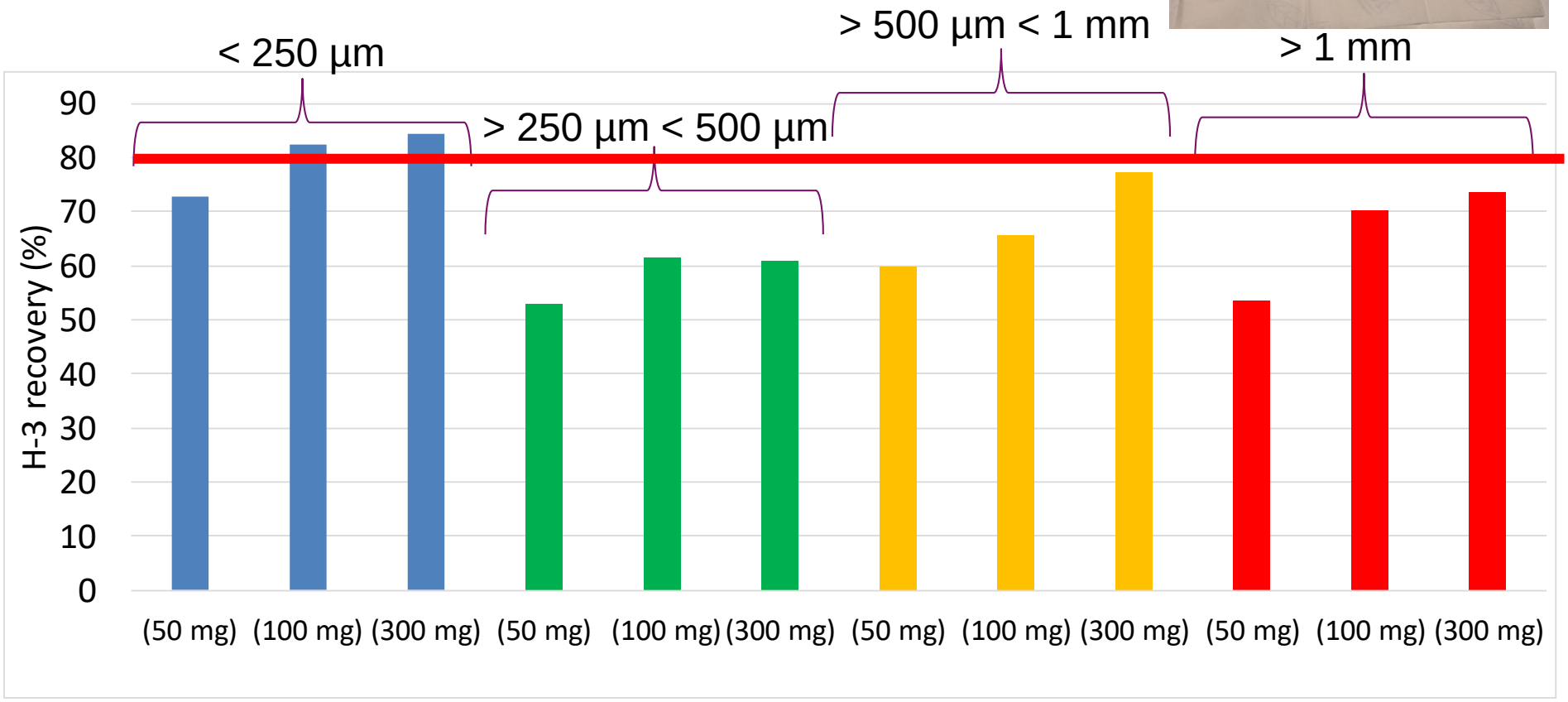
Only program 4 for recovery > 80%

MATRIX VALIDATION : CONCRETE

Different factors has been studied :

- particle sizes
- amounts





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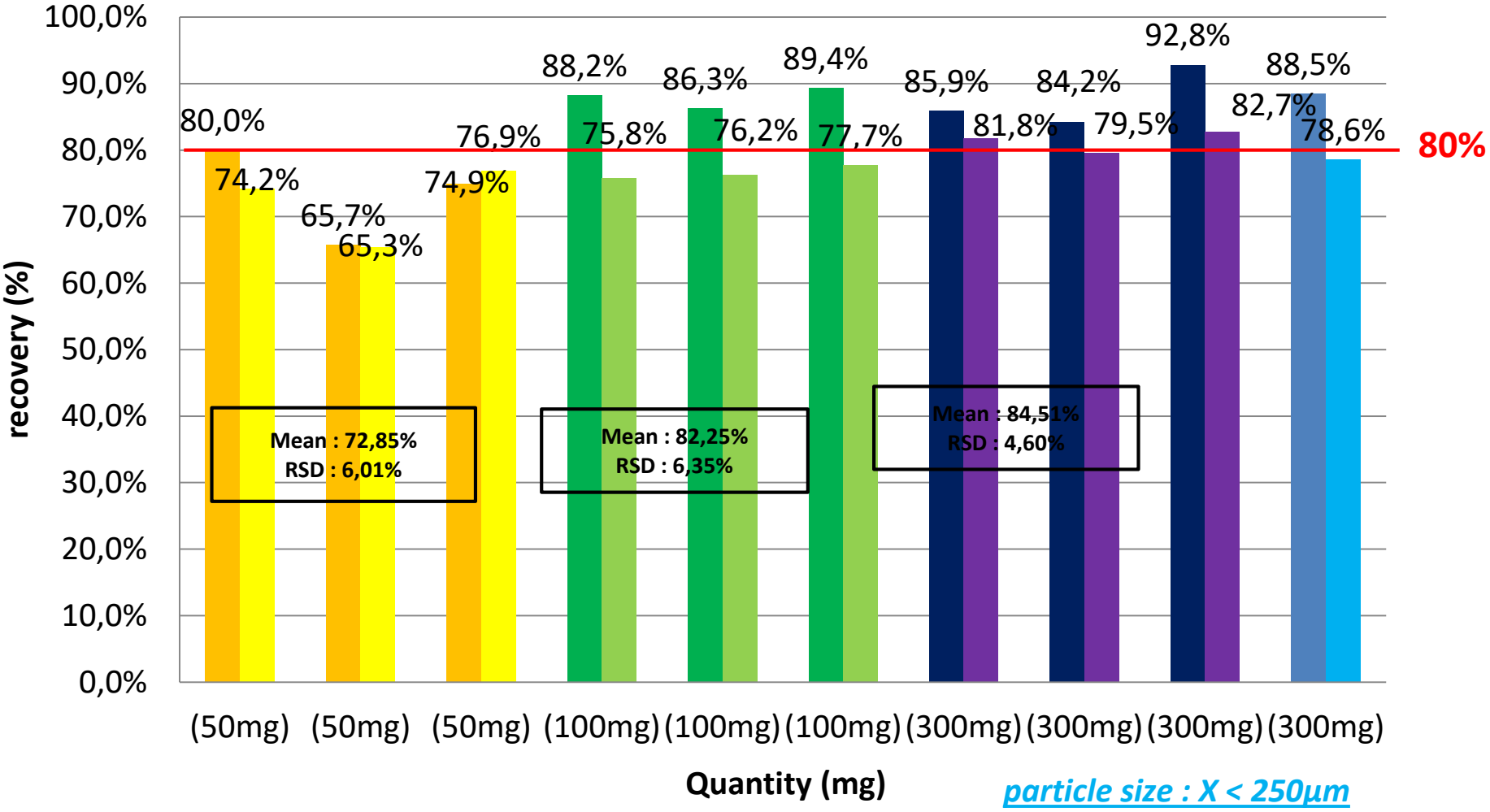


BETA EMITTERS SEPARATION : ^{63}Ni

Sample dissolution (acid digestion)

Ni, Co career

Nitroso 2 naphtol citrate

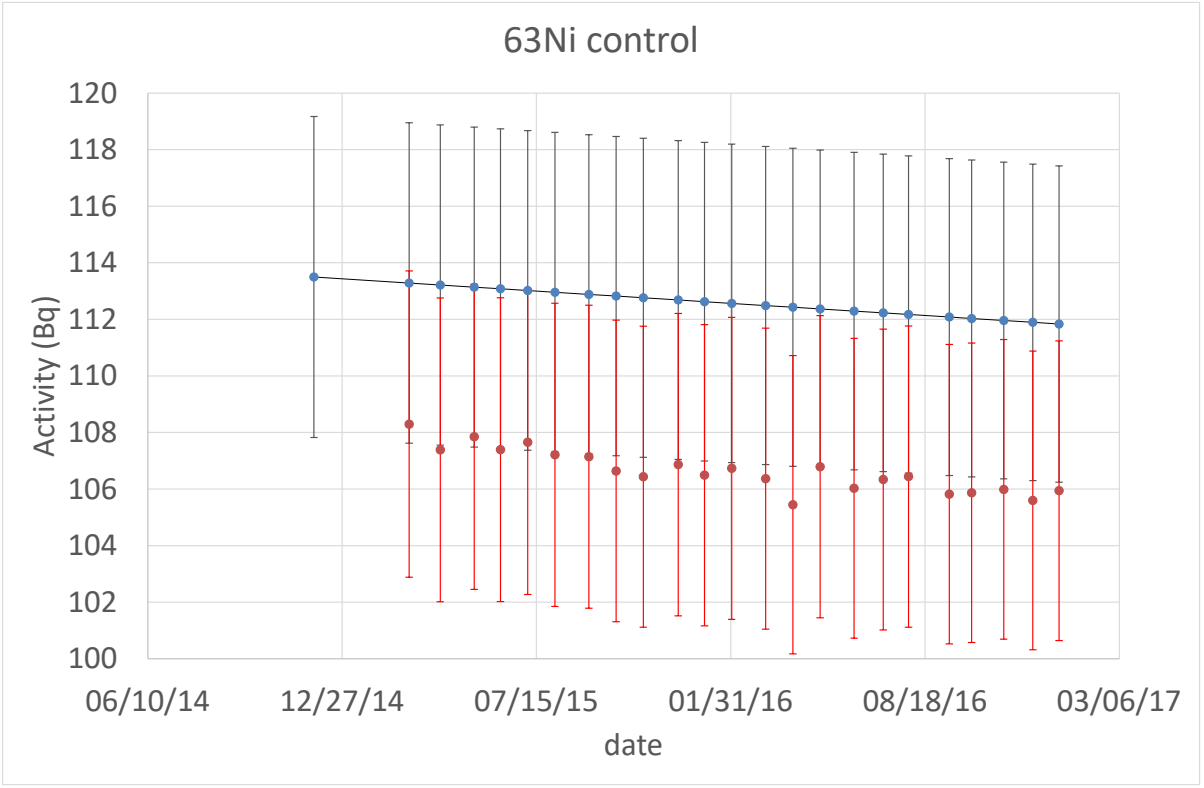
Liq-Liq extraction for Co

Liq-Liq extraction For Ni-DMG

ICP-AES Ni

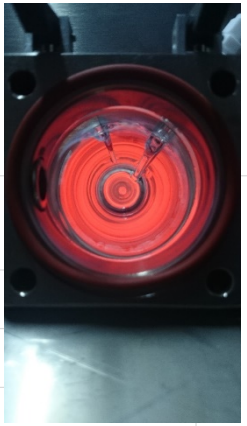
LSC ^{63}Ni

Quality check every month of the quenching curve
With certified activity sealed source



BETA EMITTERS SEPARATION : ^{36}Cl

Quality check every month
With certified activity sealed source



Graphite

950° C

Trapping in carbonate/H_2O_2 solution

NaCl carrier

Precipitation/dissolution of AgCl

Reduction of Ag and elimination

Addition of chlorine to octadecene

Purification on silica column

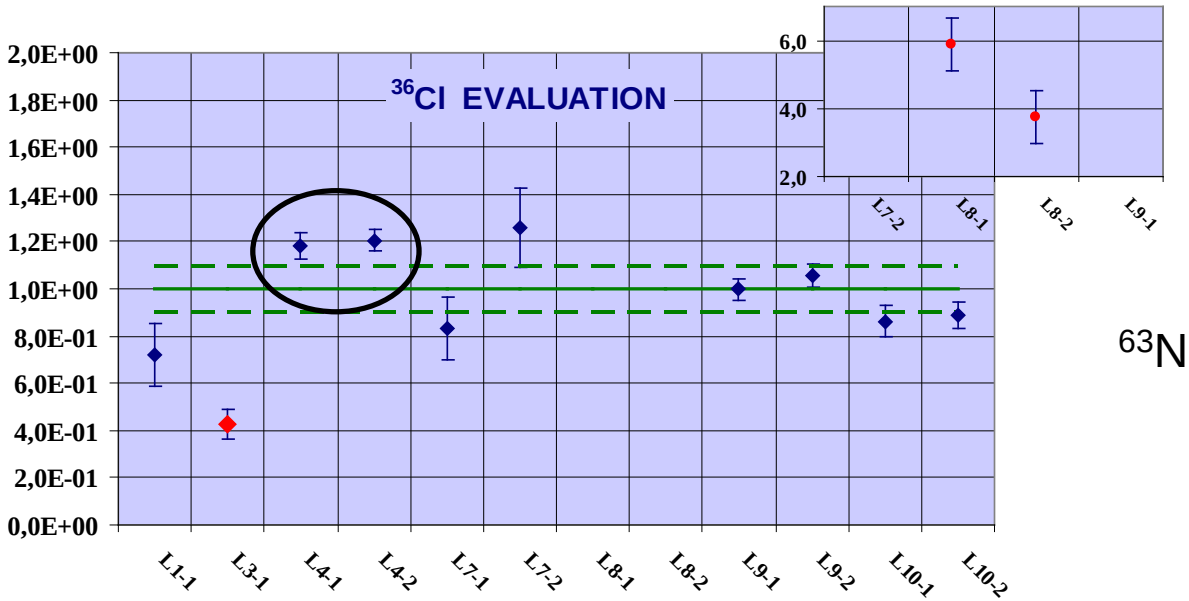
Organic molecule decomposition

HNO_3 1M

Measurement of ^{36}Cl by LSC

IC
Quantum yield

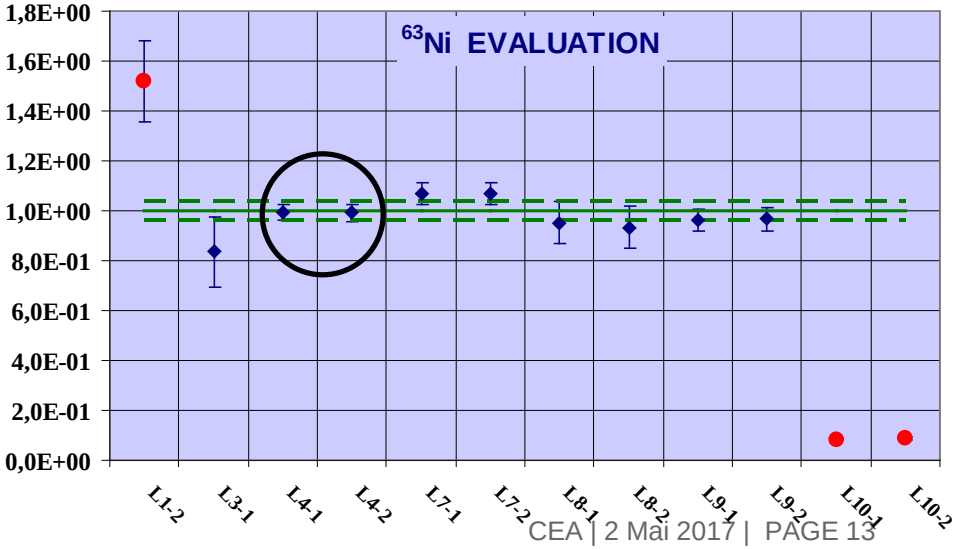
CARBOWASTE RESULTS FOR ^{36}Cl AND ^{63}Ni IN GRAPHITE



^{36}Cl activity : 27 Bq/g ± 10%

^{63}Ni activity : 113,460 Bq/g ± 10%

Very good agreement

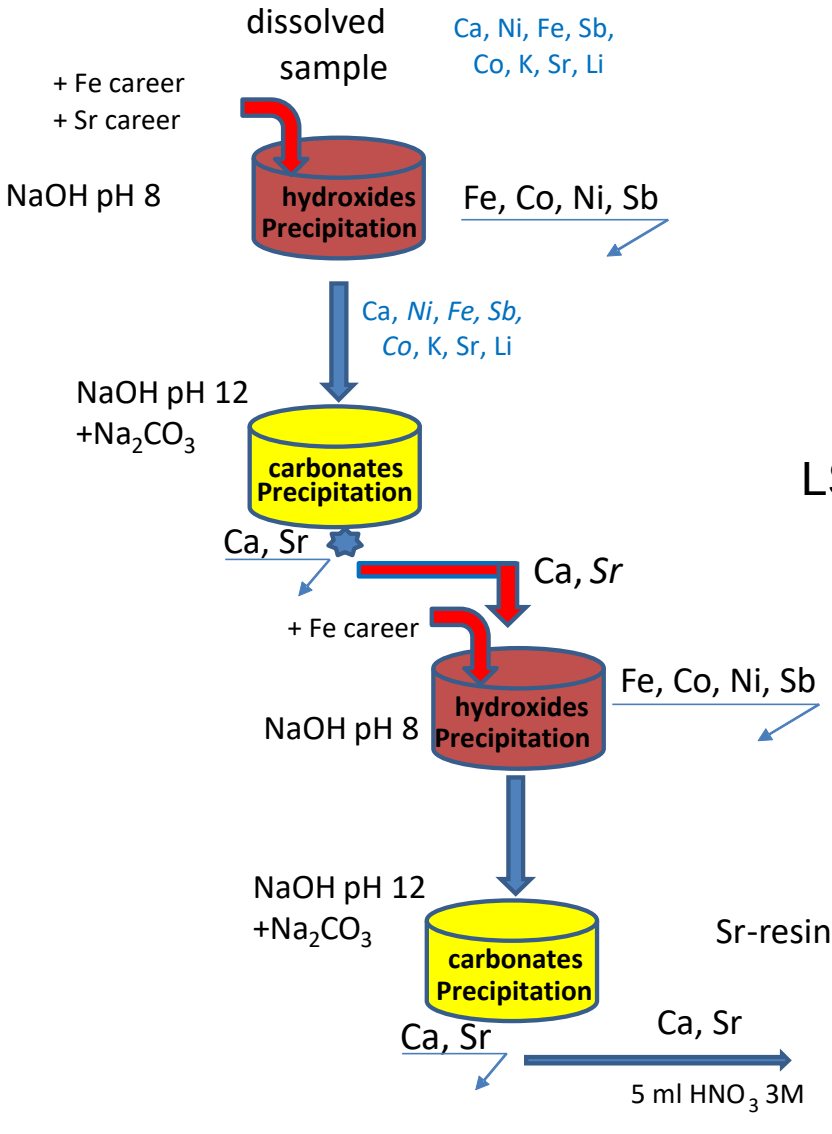


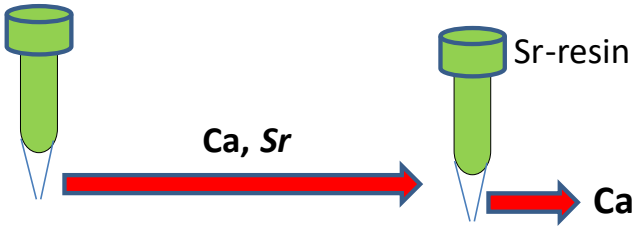
Inter comparison on real matrix :

Concrete, metals, graphite, …

→ **too rare**

BETA EMITTERS SEPARATION : ^{41}Ca





Dissolution step :

→ Depends of the matrix

Concrete, metals : **acid digestion**

LSC measurement :

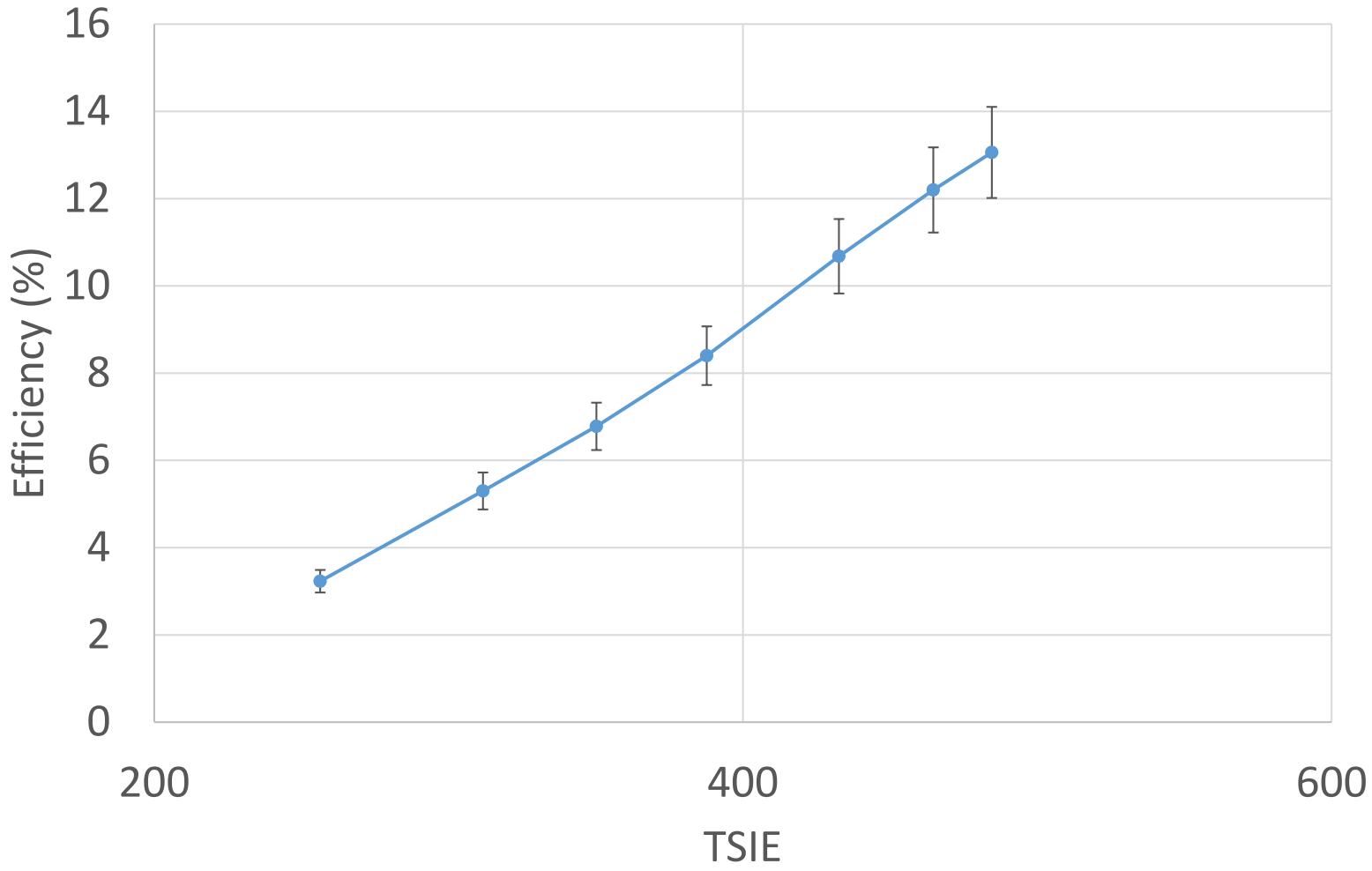
difficult for **low** contents
due to poor efficiency of ^{41}Ca

→ Counting time high

If Sr high content

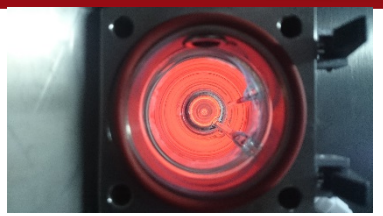
Inspired from X. Hou, Radiochimi. Acta, 93, 611, (2005)

^{41}Ca QUENCHING CURVE ON TRICARB 2900TR



ERM-AE701/1 IRMM standard ^{41}Ca/^{40}Ca 1,0114 10^{-6}

CONCLUSION



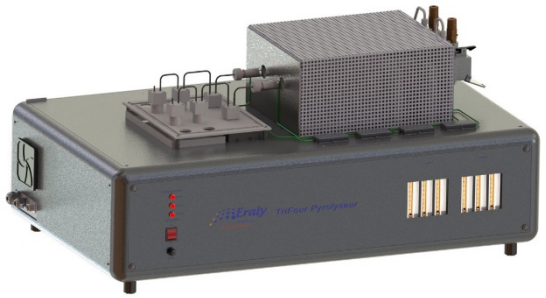
High importance for beta emitters measurement in nuclear waste

LSC is the reference method

^{3}H and ^{14}C extraction yield by pyrolysis has been validated for various matrices :

Concrete : 85% (^{3}H) 90% (^{14}C)

graphite : 100% for ^{3}H and ^{14}C



Associated rising temperature program

And gas flow values

^{63}Ni and ^{36}Cl radiochemistry efficiency proven with carbowaste results (graphite)

Need of inter comparison test on real samples (concrete, metals,…..)

^{41}Ca radiochemistry elaborated and quenching curve on TRICARB 2900 realized

Work in progress for ^{3}H measurement in another matrix : metals, resins, oils

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