Development of new radiochemical procedures for DTM radionuclides

SUB-TASK 4.5.3

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This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 945098.

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Context

- Measurement and characterization of radionuclides difficult to measure DTM
  - Characterized by their β or X-rays emissions alone,
  - Associated to their low energy emission (E<100keV) or low gamma intensity.
  - Difficulty to access to the standards calibrated with certificate to validate the process: rare, cost

Target	Reaction	DTM	Decay mode	
Li-6	(n,α)	H-3	β-	ACTIVATED PRODUCTED
C-13	(n,α)	C-14	β-	
Cl-35	(n,γ)	Cl-36	β-(β+, EC)	
Ca-40	(n,γ)	Ca-41	EC	
Fe-54	(n,γ)	Fe-55	EC	
Ni-58	(n,γ)	Ni-59	EC	
Ni-62	(n,γ)	Ni-63	β-	
Zr-92	(n,γ)	Zr-93	β-	
Mo-92	(n,γ)	Mo-93	EC	
Nb-93	(n,γ)	Nb-93m	IT	
Nb-93	(n,γ)	Nb-94	β-, γ	
Mo-98	(n,γ)	Tc-99	β-	
Sn-124	(n,γ)	Sb-125	β-, γ	

DTM	Production mode	Decay mode	
Sr-90	Fission	β-	FISSION PRODUCTS
Y-90	Fission, decay	β-	
Tc-99	Fission, activation	β-	
I-129	Fission	β-	
Cs-135	Fission	β-	

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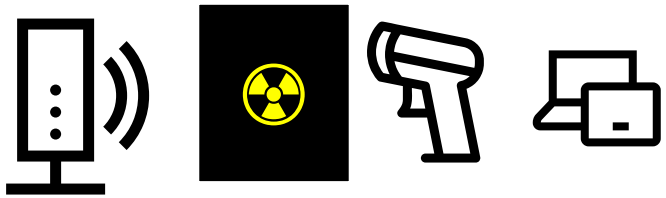
Taddei et al., 2015; Veres et al., 2022

Evaluation of activities

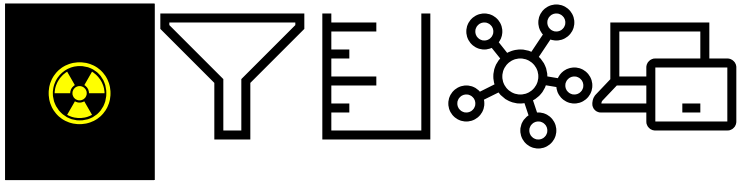
Non destructive method



- Scaling factor to determine the activity level via the ratio with the activitiy of the RN that is easy to determine (Co-60, Cs-137)
- Gamma or neutron measurements for qualitative controls without sampling

  – Discrepancy can exist between measured activities
    - Corrosion of steels,
    - Better knowledge of impurities in metallic materials

Destructive method



- To complete the non destructive method
- Important in particular when information is not accessible and available (chemical composition, irradiation conditions….) for the historical wastes

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IAEA (2009)

PREDIS

Objectives

VTT

Development of a radiochemical procedure for the optimal detection and measurement of Ni-59, Ni-63, Mo-93

CTU CZECH TECHNICAL UNIVERSITY IN PRAGUE

Development of simplified measurement procedure of Ni-59, Ni-63, Ca-41

IMT Atlantique
Bretagne-Pays de la Loire
École Mines-Télécom

Development of a radiochemical procedure for the optimal detection and measurement of Zr-93

- Targets:
  - Highly selective and efficient separation and purification
  - Development of sensitive to ultra-sensitive method of measurement, depending on the radionuclide

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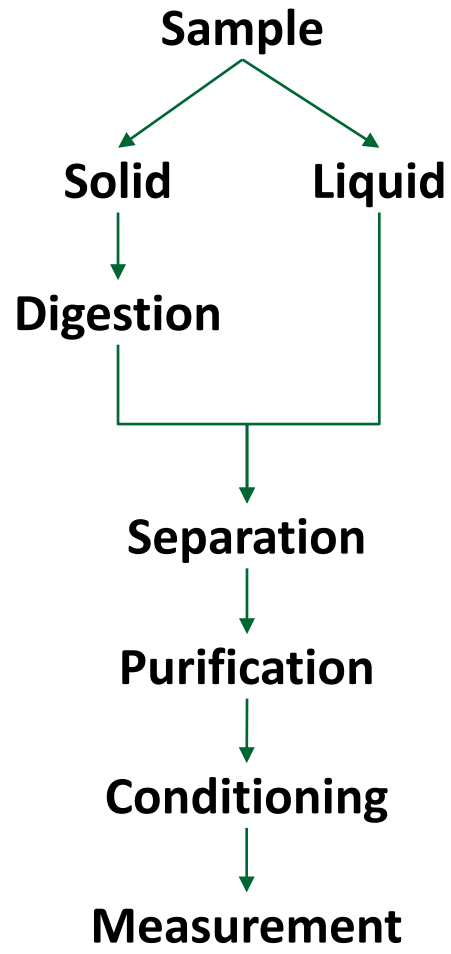
General methodology

- **Sample**
  - Synthetic: prepared in laboratory from chemical reagents
  - From solid containing the radionuclide source
  - Surrogate to simulate the radionuclide due to limited quantity or availability
- **Separation / Purification**
  - Purification by chromatographic resins to recover the radionuclide of interest
  - Electrodeposition
- **Conditioning**
  - Sample preparation for measurements
  - Liquid form or deposition on filters or plates
- **Measurement**
  - Selection of an adapted analytical technique
  - Optimization of detection efficiency



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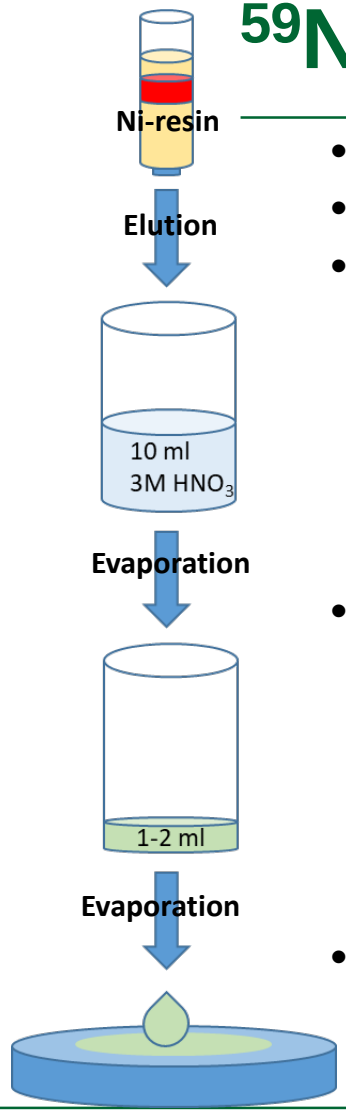
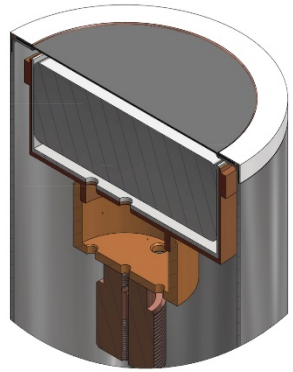
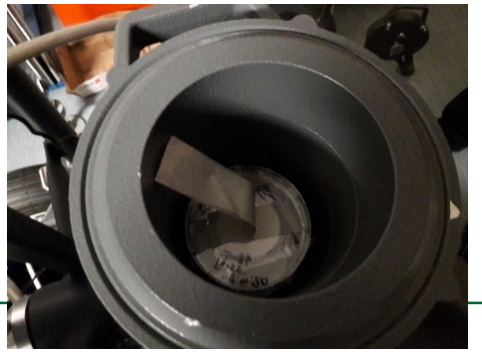
^{59}Ni and ^{63}Ni analysis in activated steel



- ^{63}Ni analysis method published [1]
- Study to detect ^{59}Ni using a broad energy gamma spectrometer (3 kev to 3 Mev)
- Preparation of a standard curve
  - ^{55}Fe used as surrogate
  - Evaporation on a filter
  - Good linearity between 20-1000 Bq (R^2 = 0,9991)
  - When samples are measured, new data points can be added in the std curve prior the measurement i.e. verification that the curve is still valid
- Interference of ^{60}Co contamination
  - Co and Ni chemically similar → ^{60}Co possible in purified Ni-fraction
  - ^{60}Co interference tested with 1000 Bq, 500 Bq and 100 Bq
    - Effect seen for ^{55}Fe activities below 250 Bq
  - Purity of Ni-fraction can be verified by gamma spectrometry prior to evaporation or during ^{59}Ni measurement as energy range covers also ^{60}Co
- Next step an intercomparison exercise and writing of a publication





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[1] LESKINEN ET AL. J Radioanal Nucl Chem **323** (2020) 399-413.

^{93}Mo (^{94}Nb and ^{93}Zr) analysis

Sequential radiochemical purification of ^{93}Mo (^{94}Nb and ^{93}Zr)

Dissolution

↓

Cation exchange
Retained: ^{54}Mn, ^{55}Fe, ^{60}Co, ^{59}Ni, ^{63}Ni etc
Through: ^{93}Zr, ^{125}Sb, ^{99}Tc, ^{93m}Nb, ^{94}Nb, ^{93}Mo etc

↓

TEVA
Elution of ^{93}Zr → ICP-MS
Elution of ^{93}Mo
Elution of ^{94}Nb → HPGe

↓

Alumina
Purification of ^{93}Mo

Detection of ^{93}Mo using ICP-MS

Elimination of interferences

NEXT

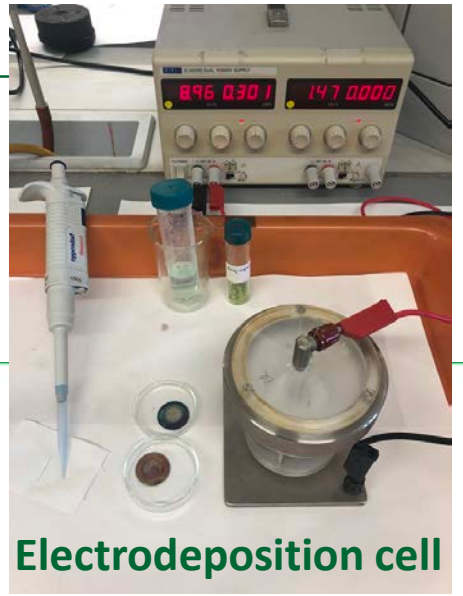
Production of ^{93}Mo with proton bombardment of Nb

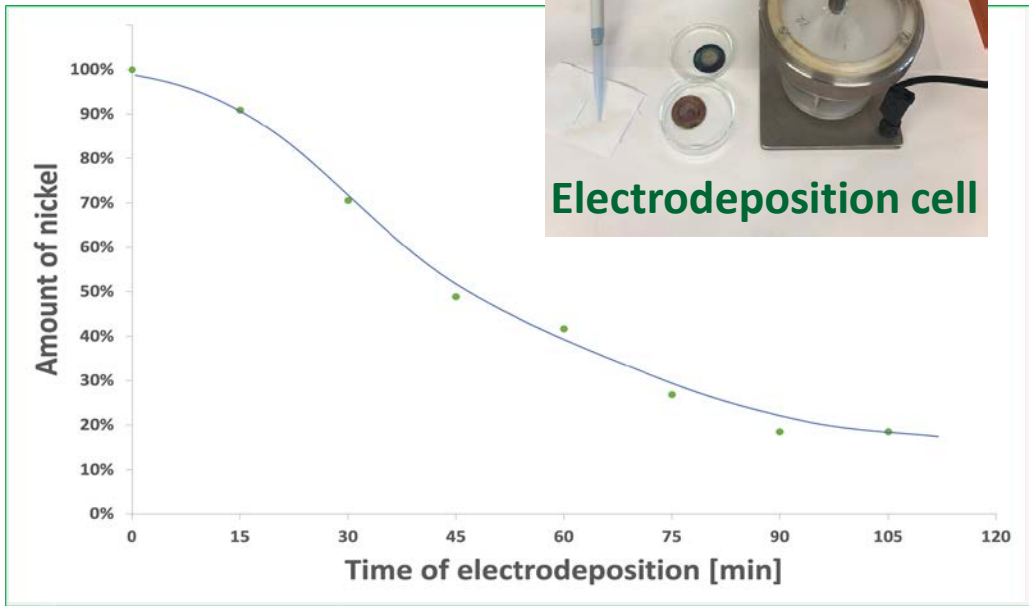
This project has received
research and innovation
020

PREDIS Development of measurement procedures for ^{59,63}Ni, ^{41}Ca

Ni isotopes:

- How to improve measurement of $^{59, 63}$Ni and simplify the whole procedure?
  1. Standard radiochemical separation using Ni-resin / DMG
  2. Electrodeposition of Ni on a thin stainless steel planchette
  3. Measure the planchette with the deposit towards detector in a calibrated geometry = ^{59}Ni
  4. Place the planchette into a LSC vial with the deposit towards cocktail volume = ^{63}Ni
- Calibration needed (geometry) and LSC windows has to be adjusted
- Electrodeposition = additional separation and concentration step against non-depositing RN, higher sample volume can be used, carrier possible.
- Yield up to 90% over 2 hours of electrodeposition in H_3BO_3 solution at adjusted pH, 1.5 Acm^{-2}





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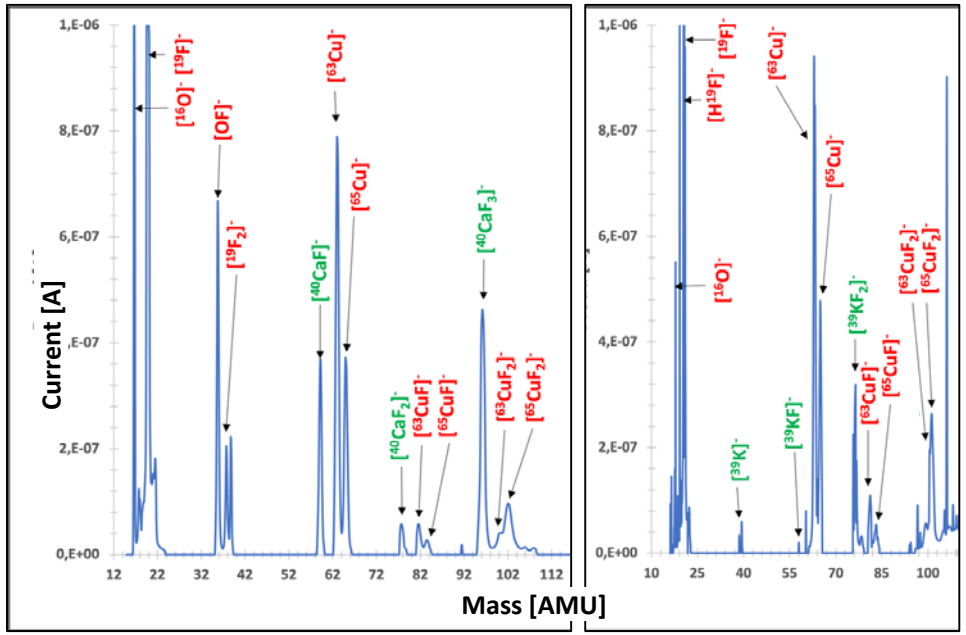
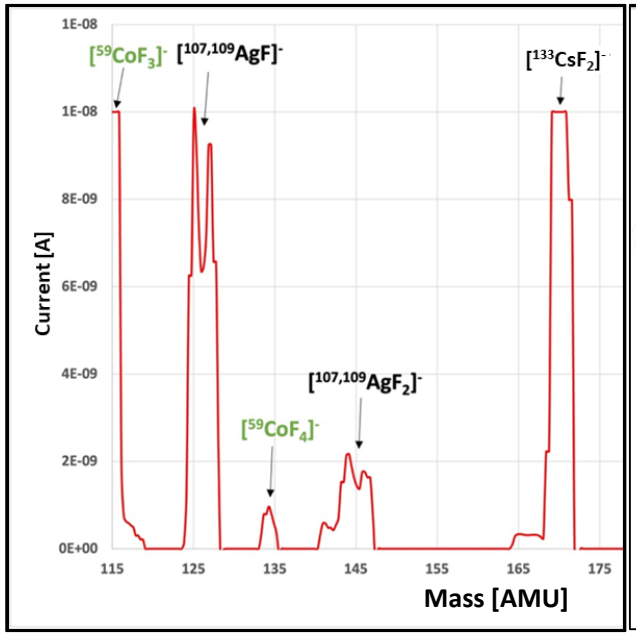
PREDIS Development of measurement procedures for ^{59,63}Ni, ^{41}Ca

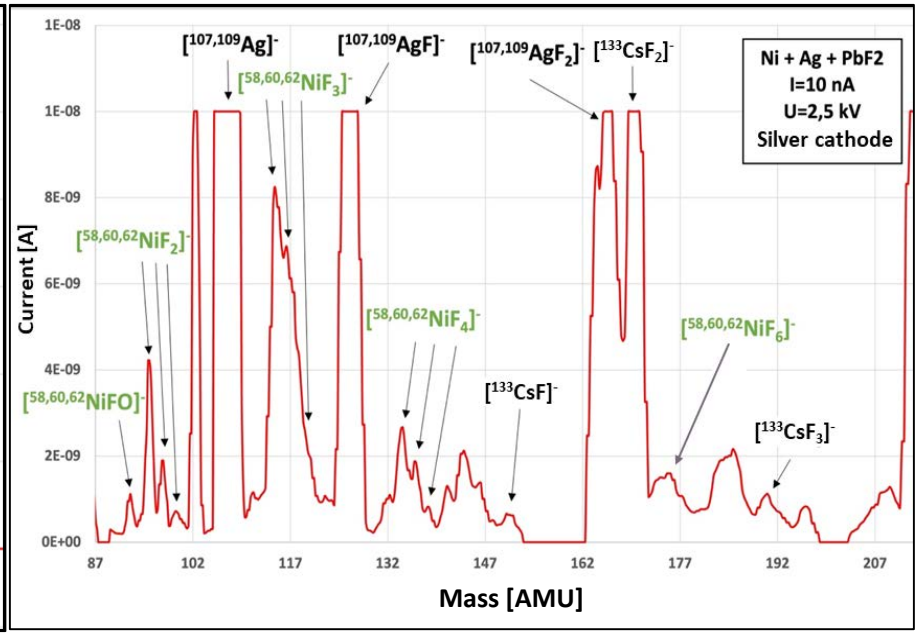
Testing of suppression of ^{59}Ni and ^{41}Ca isobars for AMS measurement:

- Ultrarace amounts, suppression of stable ^{41}K and ^{59}Co needed.
- Mass scans to prove/find heavy negative fluoride molecules $[MF_n]^-$ - mass spectra
- Measured at caesium sputter ion source in the form of metals/salts mixed with PbF_2

- MS spectra for: ^{40}Ca ^{39}K ^{59}Co ^{58,60,62}Ni







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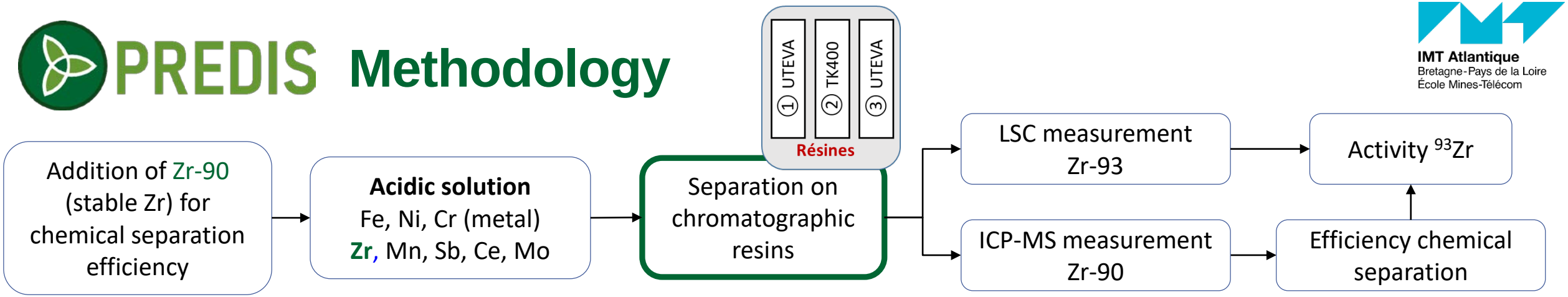
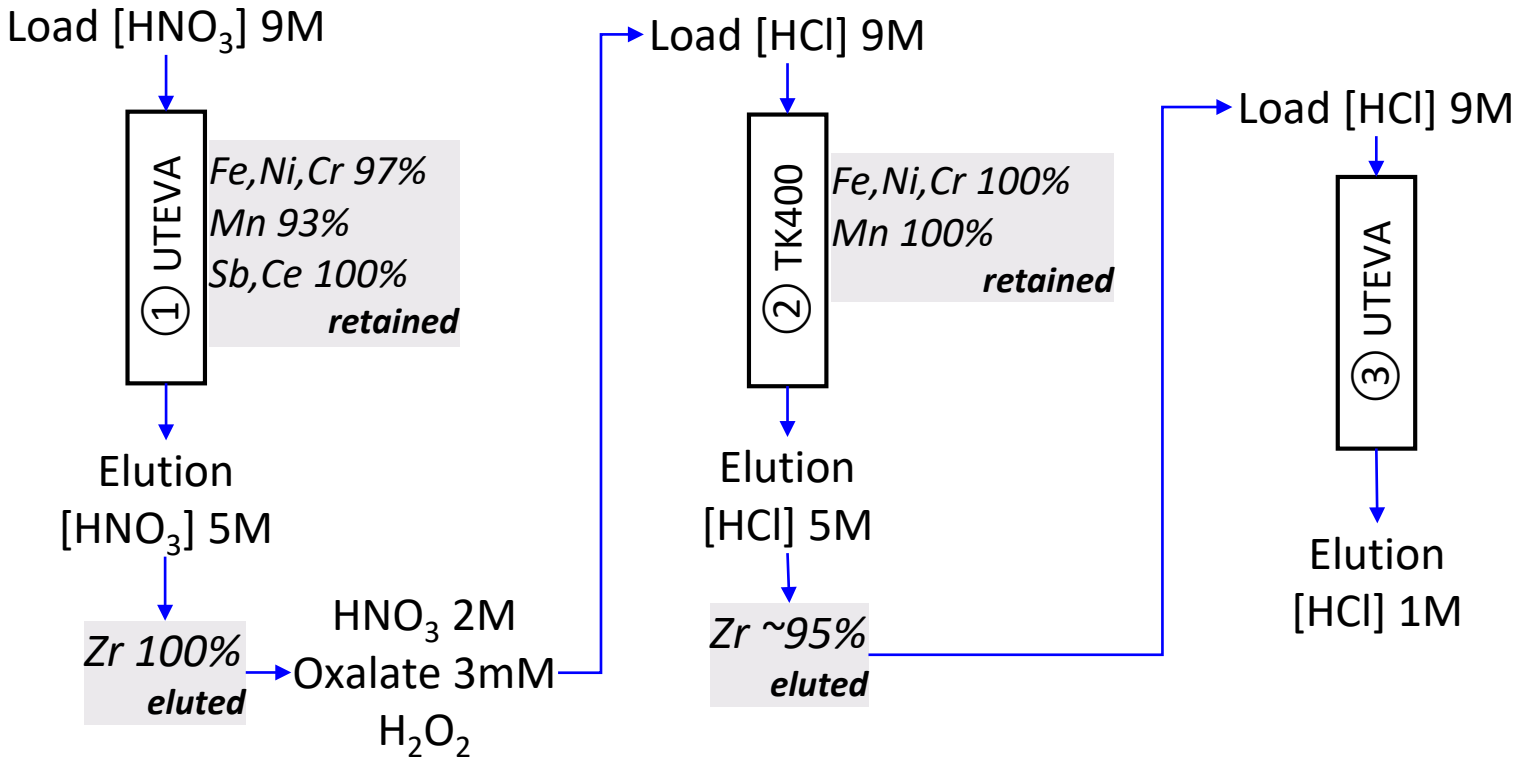
PREDIS Methodology





2g sample, 2h counting	background	Detection limit
HCl 1M / U.G. AB	0.07 cps	0.041 Bq/g
HCl 0.1M / U.G. AB	0.07 cps	0.039 Bq/g

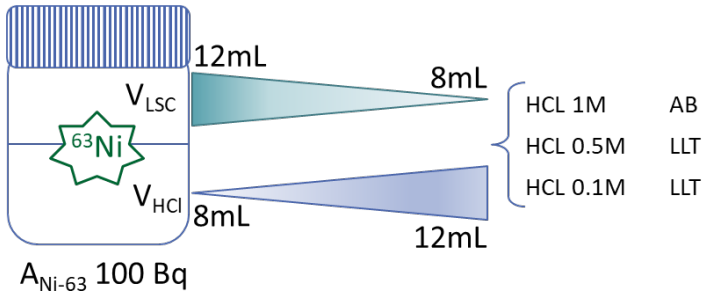
Collaboration TRISKEM

Zr-93 measurement

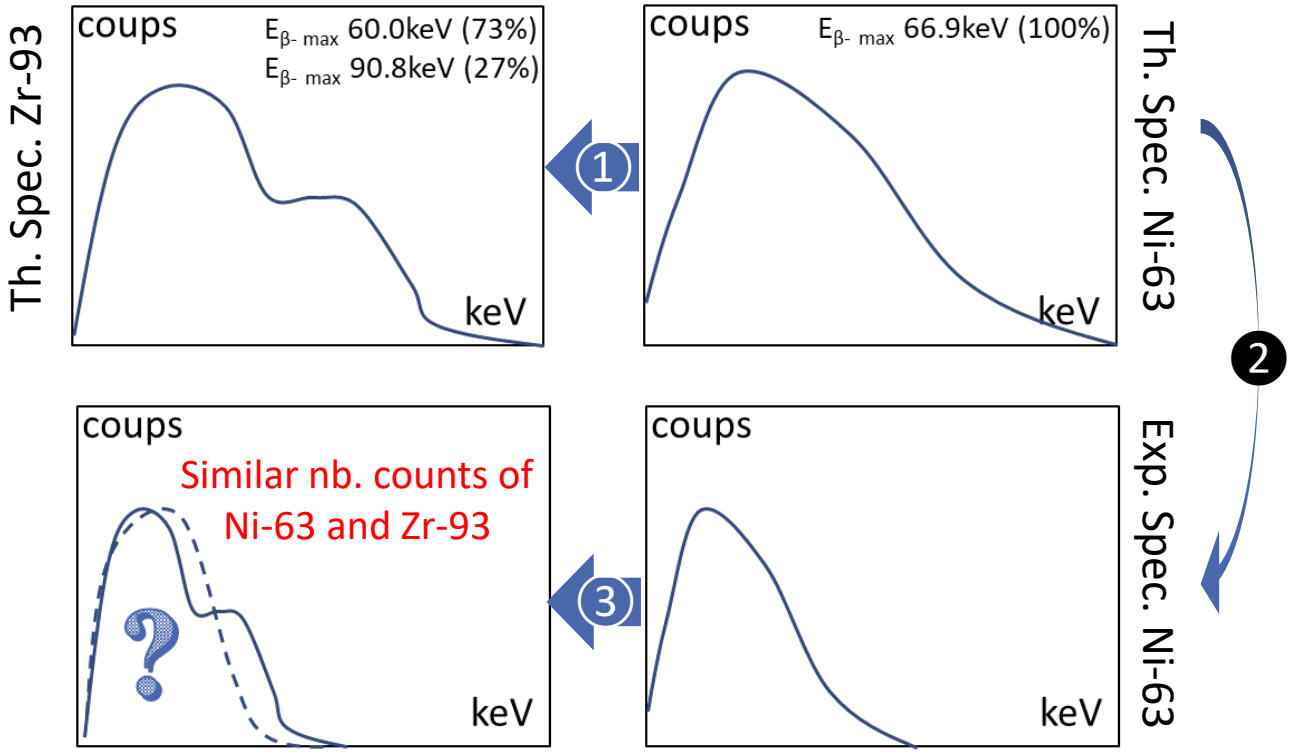
1/ limited supply of radioactive Zr-93 standard: challenging temporal projection

2/ use of Ni-63 as a surrogate

3/



4/ theoretical simulation of Ni-63 and Zr-93 spectra in the last elution condition for LSC analytical measurements conditions



Collaboration Ph. Cassette

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