9th INTERNATIONAL
CONFERENCE ON NUCLEAR AND
RADIOCHEMISTRY – NRC9

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A new plastic scintillation resin for single-step separation, concentration and measurement of ^{99}Tc

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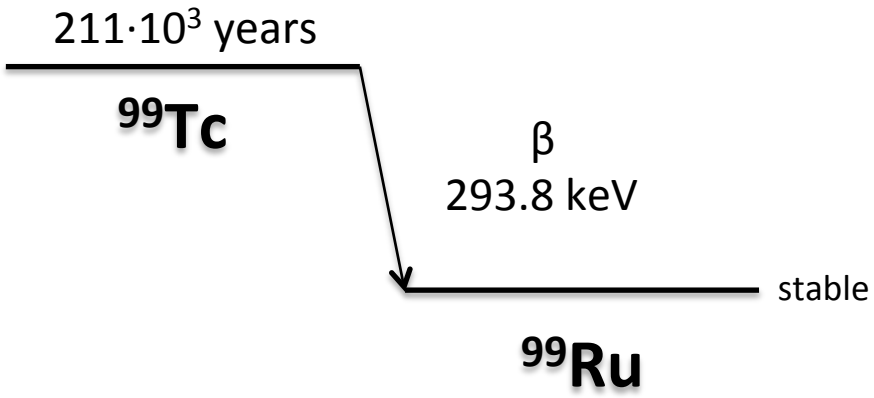


ORIGIN

- ^{99m}Tc used in nuclear medicine
- Fission product of ^{235}U and ^{239}Pu

CHEMISTRY

- Tc(VII): TcO_4^-
- Very high mobility

INTEREST

- Biological samples (hospitals)
- Sea water (reprocessing discharges)

ICP-MS ← ^{99}Tc analysis → Liquid Scintillation

Separation form isobaric interferences

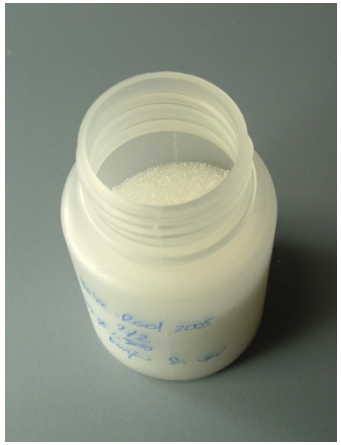
- Detection Limit
- Selectivity
- Simplicity
- Timing

Separation form beta-emmiters interferences

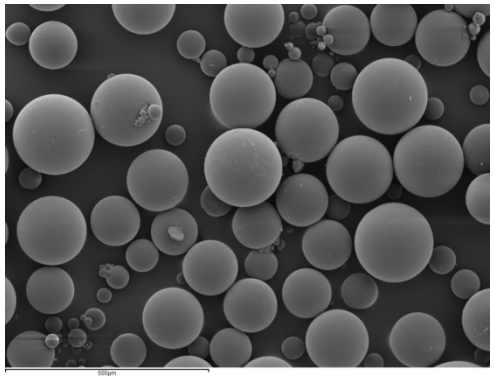
Separation + Measurement

Need for reducing operations and time without loosing selectivity and sensitivity

Plastic Scintillation microspheres (PSm) and Plastic Scintillation resins (PSresin)

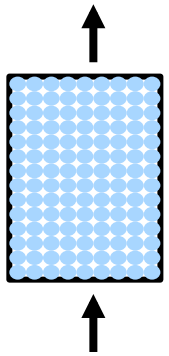


- Solid solution of fluorescence solutes (PPO, POPOP, pT, Bis-MSB,…) in a polymeric solvent (polyvinyltoluene or polystyrene) and other additives.
- Diameter: from few to hundreds of micrometers



- Sample preparation similar to LS
- Low efficiency for low-energy beta emitters
- Similar efficiency for high-energy beta emitters

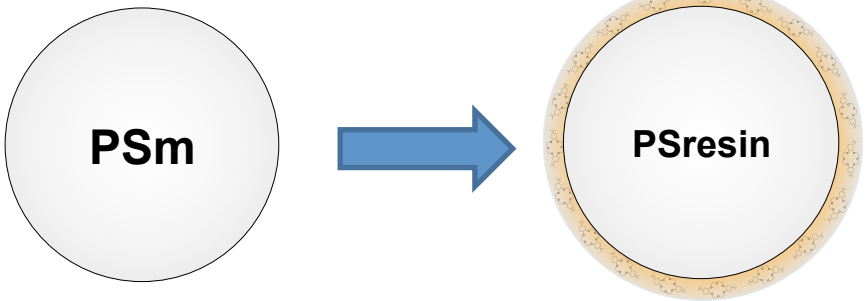
Uses and advantages of PSm

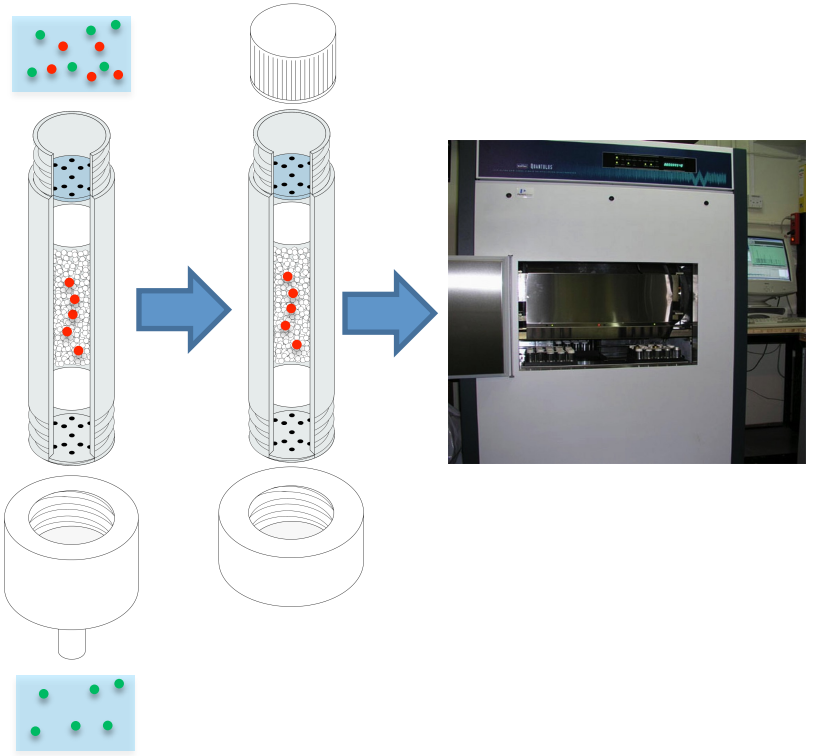


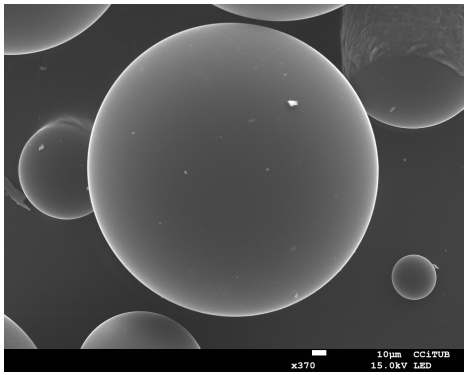
- No production of mixed waste.
- Reusability.
- Sample recovery after measurement.
- Salty samples can be used avoiding phase separation.
- Capability for real time and continuous monitoring.
- Absorption and measurement of ^{222}Rn
- Immobilization of selective extractants compounds onto the PSm surface.

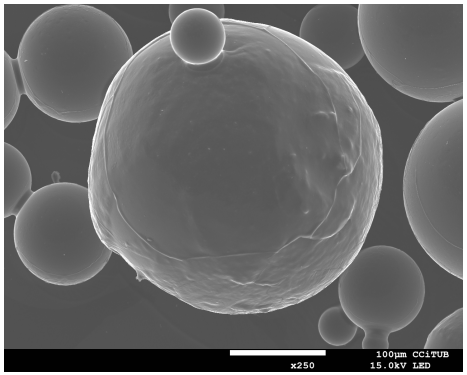
PSresin

Plastic Scintillation resins (PSresin)









Solid Phase Extraction and Scintillation Detection

A new plastic scintillation resin for single-step separation, concentration and measurement of ^{99}Tc

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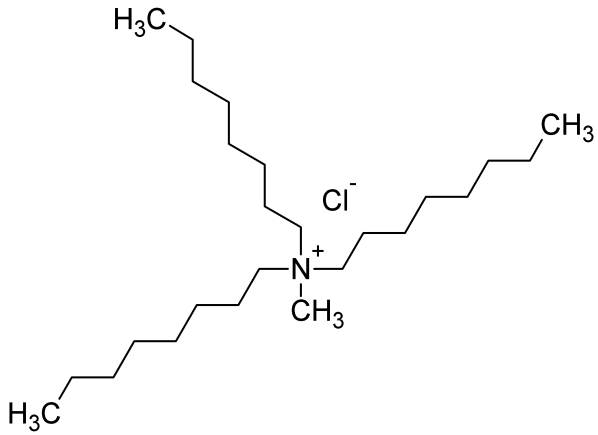
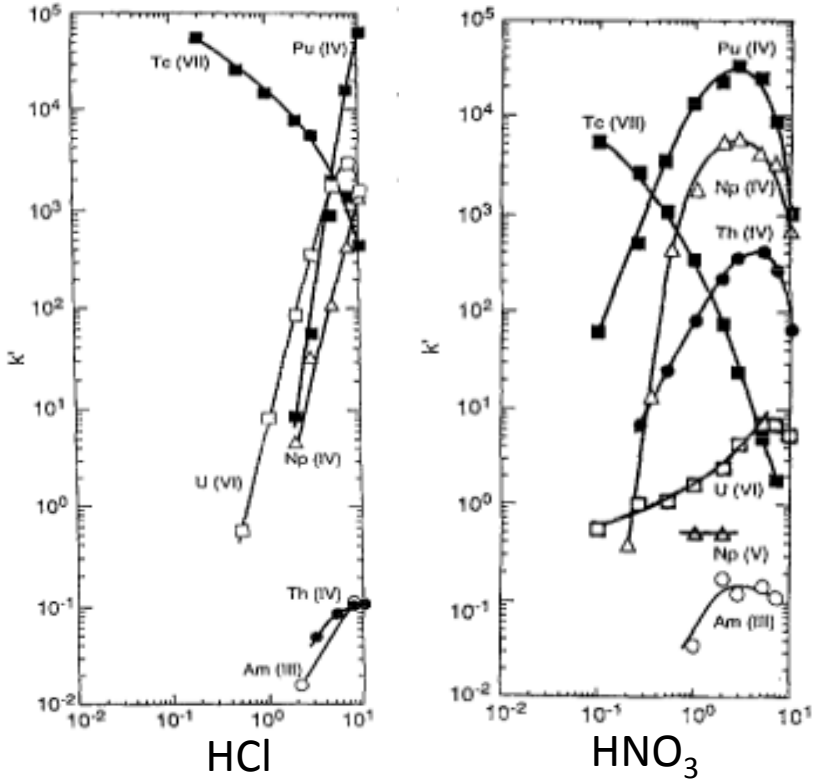
OBJECTIVE

- Development of analysis method for ^{99}Tc using a PSresin.
  - Development of the ^{99}Tc PSresin.
  - Study of the recovery and reproducibility.
  - Study of potential interferences.
  - Application to spiked sea-water and urine samples.

Development of the PSresin for ^{99}Tc

- Extractant: Aliquat·336



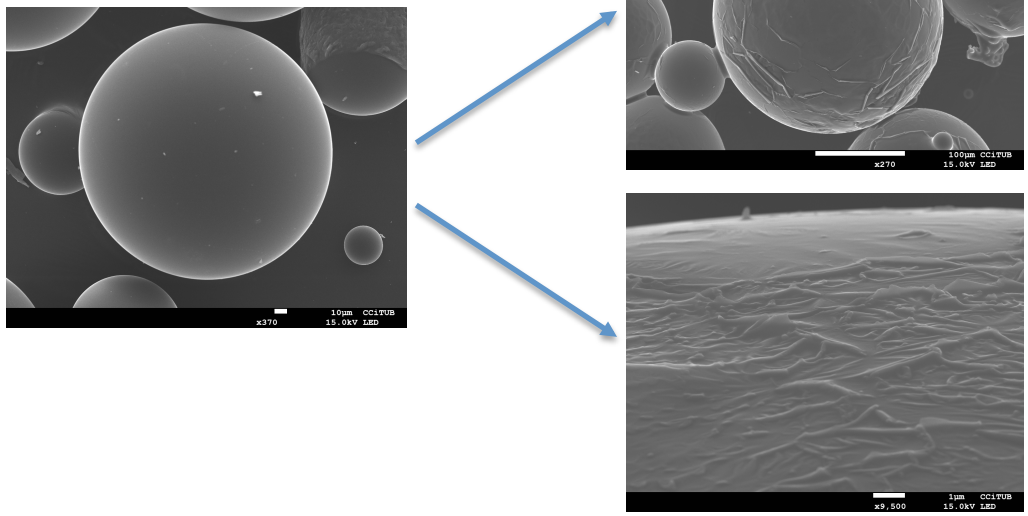


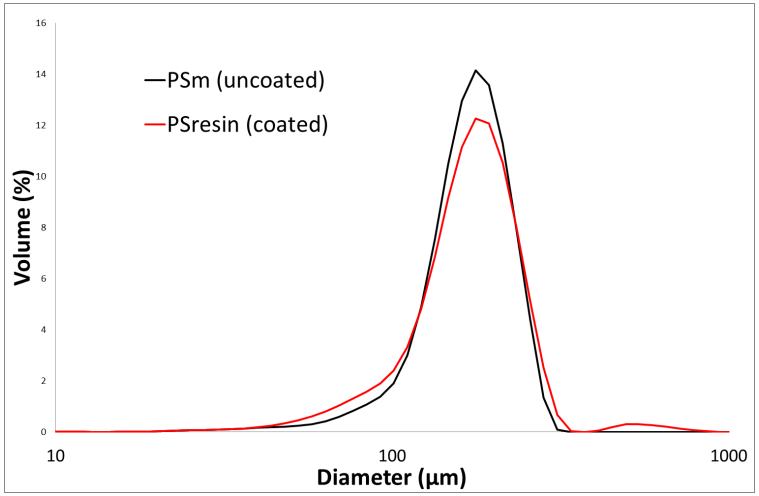
- Medium: HCl 0.1 M

E.P. Horwitz, M. L. Dietz et al. Analytica Chimica Acta, Volume 310, Issue 1, 1995, Pages 63-78,

Development of the PSresin for ^{99}Tc

- Extractant: Aliquat·336
- Immobilization: Impregnation through solvent evaporation at reduced pressure.





Satisfactory thin coating

Study of the reproducibility and recovery

- Four different batch of PSresin were prepared
- Analysis of three samples of ^{99}Tc with PSresin of each batch

- Sample: Standard Solution ^{99}Tc in 0.1 M HCl.
- Column: 3 g of PSresin in a 6 mL column-vial
- Separation conditions:
  1. Conditioning : 5 mL of 0.1 M HCl.
  2. Sample loading: 10 mL of sample
  3. Cleaning: 4*5 mL of deionized water.
- Flow: 0.5 mL min^{-1}
- Detector: Quantulus™

1. Column-Vial (^{99}Tc retained)
2. Wastes by LS (^{99}Tc non-retained)

Study of the reproducibility and recovery

Immobilisation batch	Blank (cpm)	Total efficiency (%)	SQP(E)	Recovery (%)
1	1.53 ± 0.09	71.9 ± 1.3	807 ± 5	> 98.2
2	1.69 ± 0.21	69.3 ± 1.5	811 ± 1	> 98.3
3	1.73 ± 0.22	68.3 ± 1.1	813 ± 2	> 98.4
4	1.56 ± 0.09	69.8 ± 1.9	809 ± 3	> 98.4

$$Total_{efficiency} = Recovery \cdot Counting_{efficiency}$$

- Reproducible results: background, total efficiency and quenching
- ^{99}Tc is non detected in wastes (quantitative recovery).
- Mean value of Counting efficiency: 70.0% ± 1.9%

Study of anionic interferences

Total efficiency (%)	SQP(E)
70.0 ± 1.9	810 ± 4

- SO_4^{2-}; NO_3^- at sea water level

Anion	Concentration (g L^{-1})	total efficiency (%)	SQP(E)	Recovery (%)
SO_4^{2-}	0,25	71.1 ± 2.9	809 ± 6	> 98.3
NO_3^-	0,05	71.3 ± 4.6	810 ± 8	> 98.3

- SO_4^{2-} and NO_3^- do not produce an interference in the ^{99}Tc analysis.

Study of anionic interferences

Total efficiency (%)	SQP(E)
70.0 ± 1.9	810 ± 4

- Cl^- (at sea water concentration levels and two lower concentration levels)

Anion	Concentration (g L^{-1})	total efficiency (%)	SQP(E)	Recovery (%)
Cl^-	1.2	67.4 ± 1.6	810 ± 14	> 98.4
Cl^-	11.2	67.5 ± 2.3	804 ± 1	> 98.4
Cl^-	29.2	67.8 ± 2.2	804 ± 4	> 98.3

- No ^{99}Tc is eluted (quantitative retention)
- A slight decrease in the counting efficiency is observed.
- Change in the retention pattern of ^{99}Tc on the column-vial.

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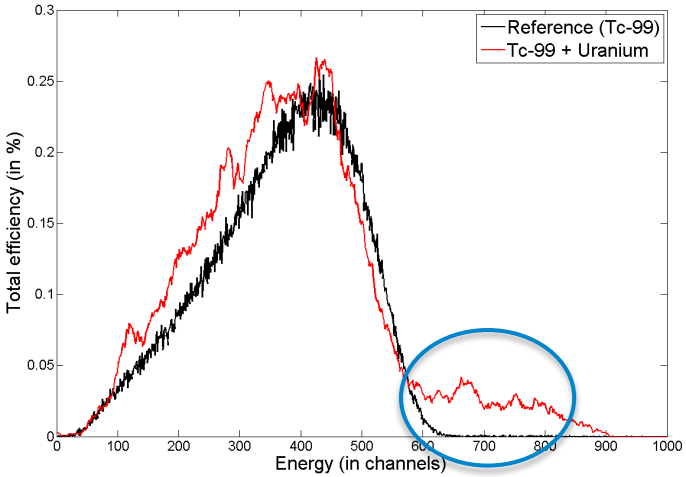
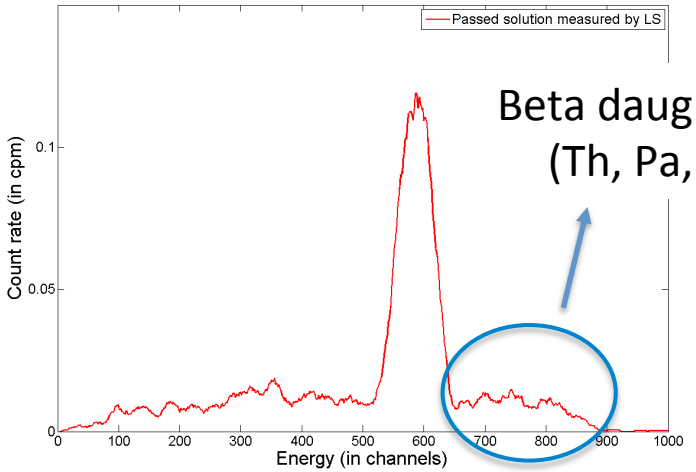
Study of radioactive interferences

Total efficiency (%)	SQP(E)
70.0 ± 1.9	810 ± 4

➢ Uranium (^{238}U + ^{234}U)

Rinse procedure	Total efficiency (%)	SQP(E)
water	82.5 ± 1.5	809 ± 8





Beta daughters
(Th, Pa,…)

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Study of radioactive interferences

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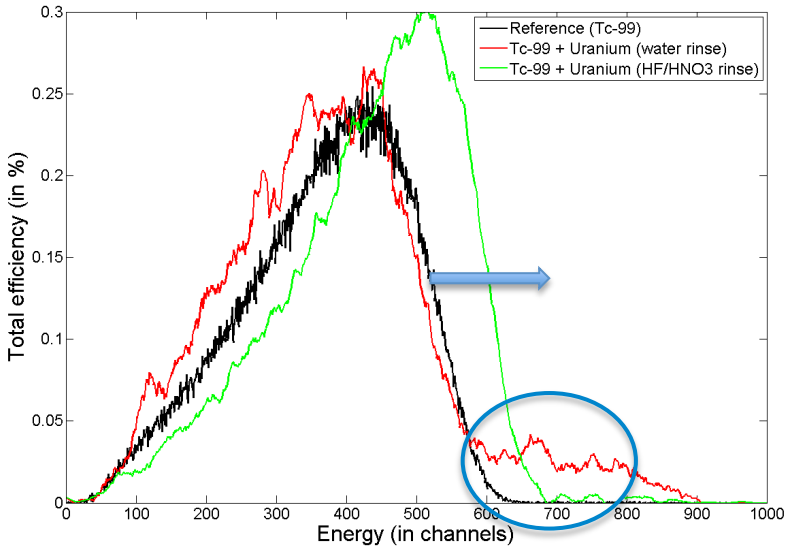
70.0 ± 1.9	810 ± 4

➢ Uranium (^{238}U + ^{234}U)

Rinse procedure	Total efficiency (%)	SQP(E)
water	82.5 ± 1.5	809 ± 8
0.1 M HF and 0.1 M HNO_3*	84.6 ± 1.4	832 ± 6

*Triskem International, Technetium-99 in Water, 2002. http://triskeminternational.com/full_eichrom_methods.asp.

Th removal



- Interference (Th) is removed
- Movement to high energies

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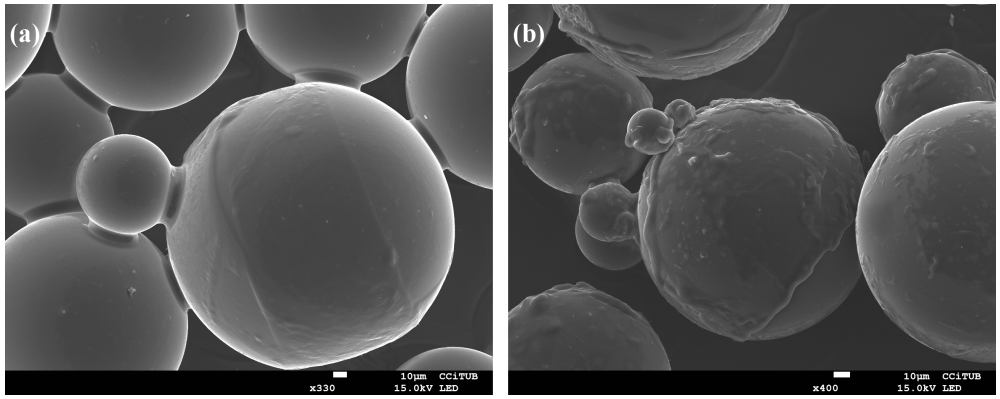

- Surface degradation
- Optics improvement

- ^{99}Tc can be analyzed in presence of U (and daughters) (HF/HNO_3 rinse)
- Calibration in the same conditions (counting efficiency: 84.6%)

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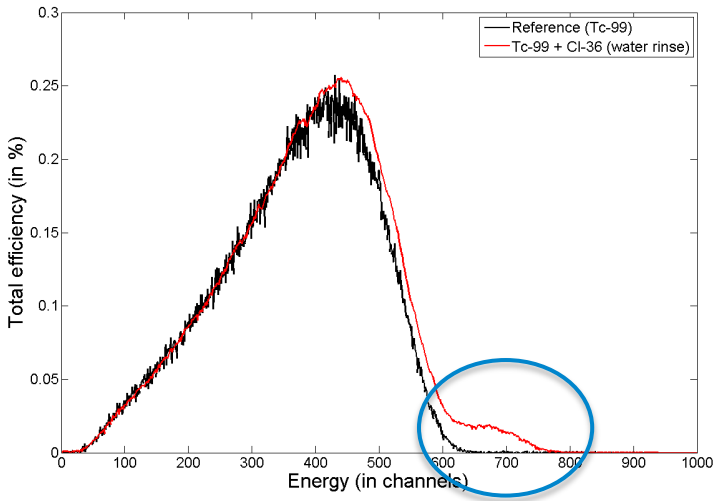
Study of radioactive interferences

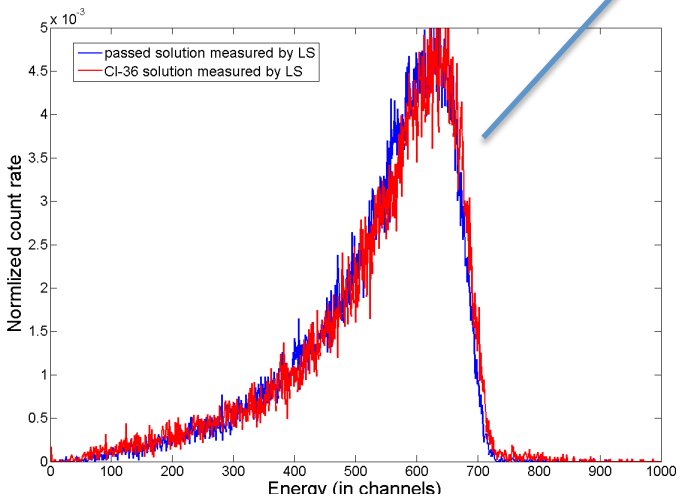
Total efficiency (%)	SQP(E)
70.0 ± 1.9	810 ± 4

➢ $^{36}Cl^-$

Rinse procedure	Total efficiency (%)	SQP(E)
water	75.4 ± 2.7	807 ± 6

Only Cl-36





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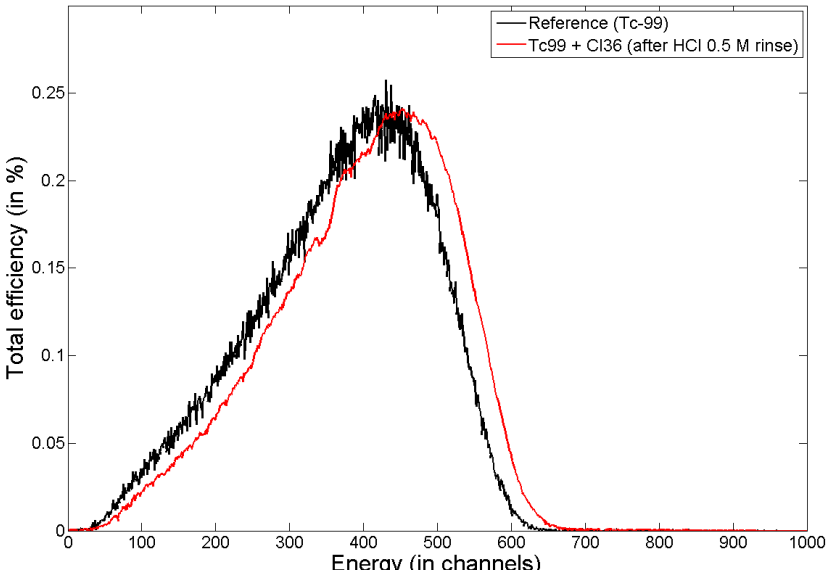
Study of radioactive interferences

Total efficiency (%)	SQP(E)
70.0 ± 1.9	810 ± 4

➢ $^{36}Cl^-$

Rinse procedure	Total efficiency (%)	SQP(E)
HCl 0.5 M	70.2 ± 2.3	800 ± 7



- Interference is removed.
- can be analyzed in presence of radionuclides in ionic form (e.g. $^{36}Cl^-$ with a HCl rinse).

Application to spiked samples.

Total efficiency (%)
70.0 ± 1.9

- Sea water: 10 mL of Mediterranea Sea water spiked with 4 Bq of ^{99}Tc
- Urine: 100 mL of urine spiked with 0,7 Bq of ^{99}Tc

Sample	Recovery (%)	Added Activity (Bq L^{-1})	Calculated activity (Bq L^{-1})	Deviation (%)
Seawater	> 98,2	407	390	-3.5
Seawater	> 98,2	407	427	5.2
Seawater	> 98,2	407	385	-4.5
Urine*	> 90,9	7.23	7.40	2.3
Urine*	> 91,5	7.77	7.12	-8.2

*T. International, Technetium-99 in Urine, 2004. http://triskem-international.com/full_eichrom_methods.asp.

- Quantification errors lower than 10%

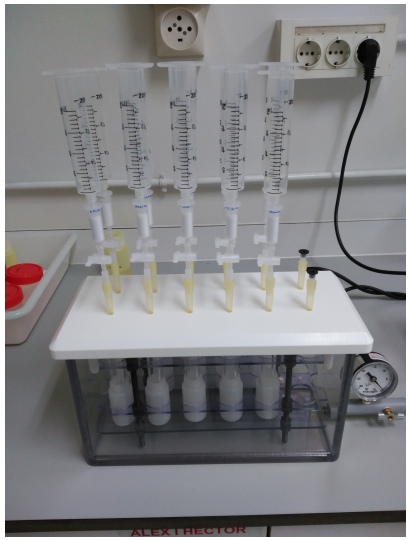
Conclusions

- A new procedure for ^{99}Tc activity determination in sea water and urine samples based on PSresins has been developed.
- PSresins used show adequate reproducibility within the same batch of production and between different batches of immobilisation.
- Quantitative retention of ^{99}Tc and detection efficiency of 70.0% ± 1.9%.
- Nitrate and sulphate have no detectable influence on the ^{99}Tc analysis with Psresins
- Chloride at seawater concentrations causes a very small decrease in total efficiency.
- Uranium and uranium daughters (Thorium) interferences can be removed by rinsing with 0.1 M HF and 0.1 M HNO_3.
- $^{36}Cl^-$ interference can be removed with 0.5 M HCl rinse.

On-going work

- Use of Solid Phase Extraction cartridges

- Use of vacuum box



- Application to nuclear waste samples

Acknowledgement

- Spanish *Ministerio de Economia y Competitividad* (MINECO) for financial support under CTM2014-02020

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Thank you for your attention

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