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Novel production method for traceable surface sources by aluminium functionalisation

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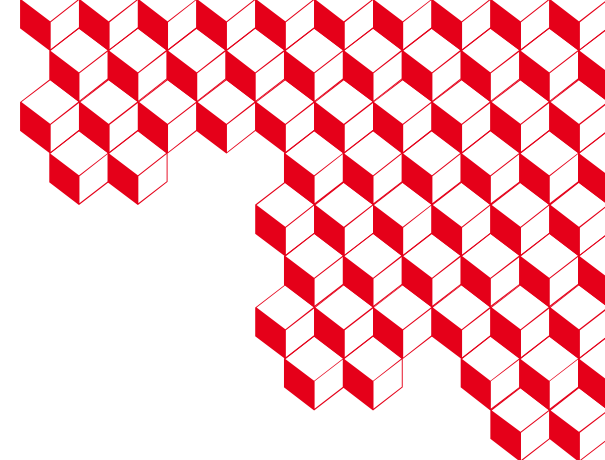
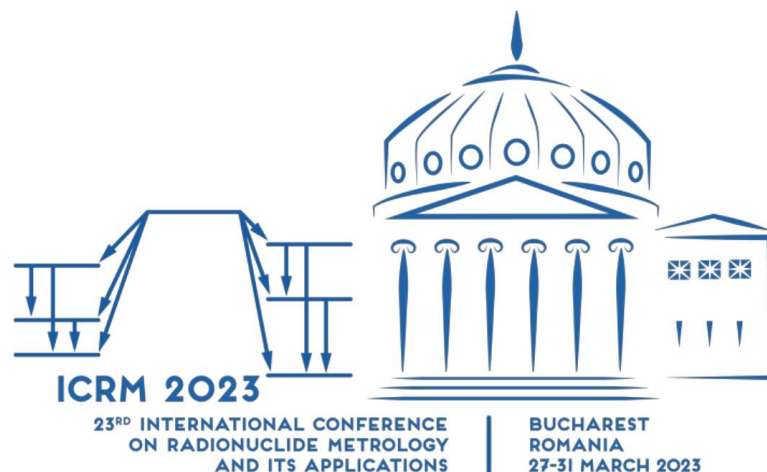
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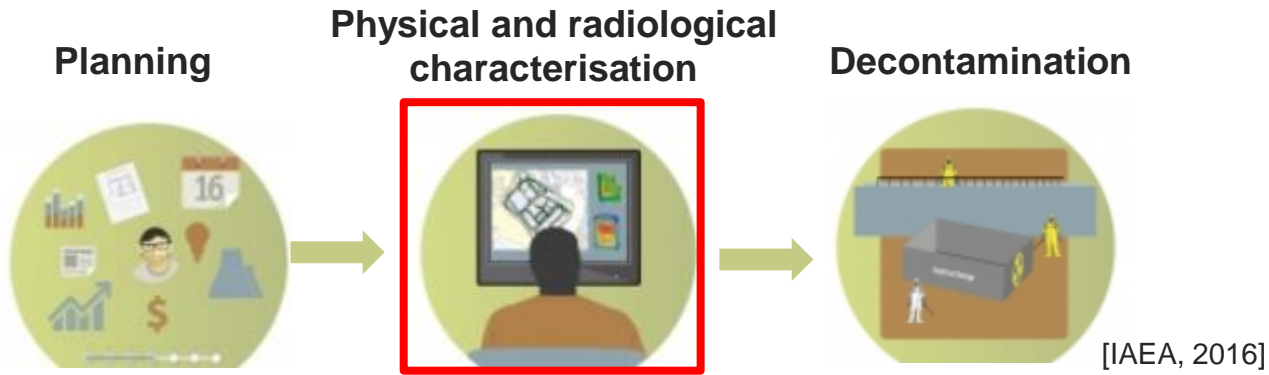
Novel production method for traceable surface sources by aluminium functionalisation

Dilan Tüzün, V. Lourenço, L. Chambon, Y. Kergadallan

Université Paris-Saclay, CEA, List, LNE-LNHB, Palaiseau, France

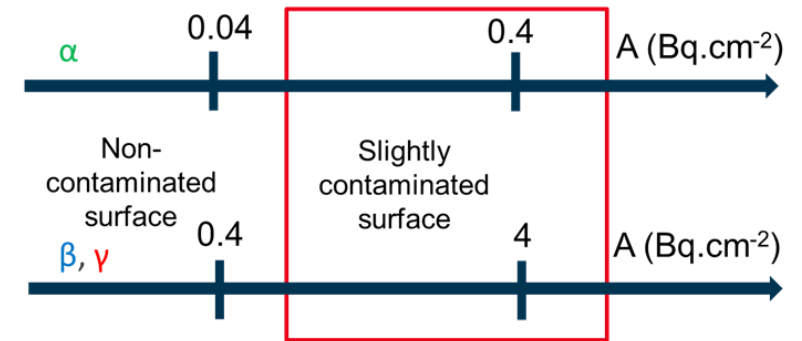
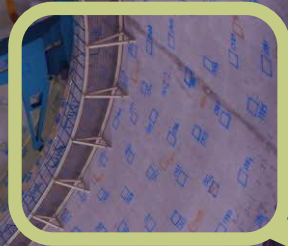


Aim: Measuring radioactivity on complex surfaces



Challenges:

- Discrimination of non-contaminated surface from low-level contaminated surface
- Very large areas



- Non-flat surfaces

Aim: Measuring radioactivity on complex surfaces



Complex shapes

Needs:

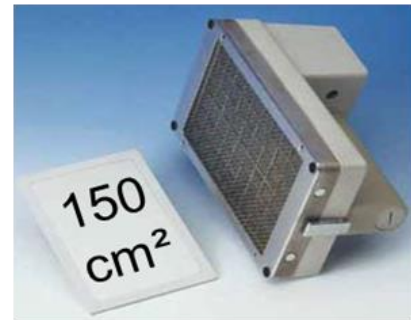
- Reliable contamination measurements
- Calibration of measurement systems

Aim:

- Traceable activity and emitted flux
- Large surface areas
- Non-contaminating surface (use on-site)
- Complex shapes



Commercial source

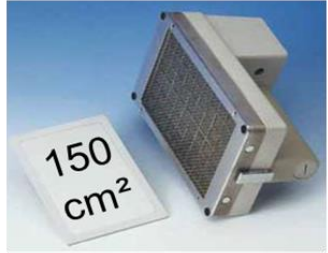


Provide metrologically traceable standard surface sources of various shapes

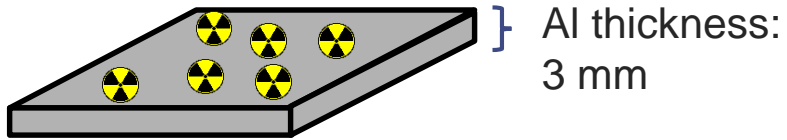
Our approach: chemical attachment



Commercial source



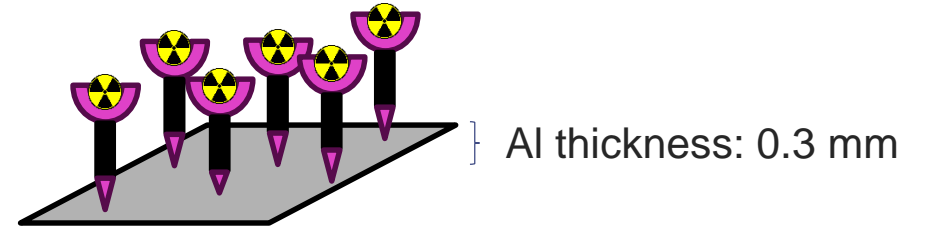
Commercial sources :
Radionuclide dried + protected



Adsorption / Physisorption $E_{\text{bonding}} \sim \text{kJ/mol}$



Chemical bonds radionuclide-surface



Chemisorption $E_{\text{bonding}} > 100 \text{ kJ/mol}$

Use of **functionalisation**

- Strong chemical bonds (non-contaminating sources, to check)
→ **No cover layer**
- Thin sources → **Minimise self-absorption** of low penetration radiation (α , β)

Chosen substrates:

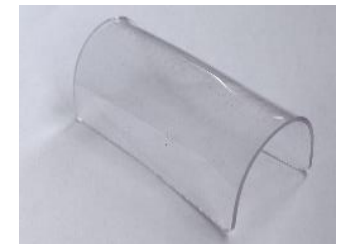
- **Aluminium foil**
- Polymeric surfaces



Radionuclide

Surface functionalisation

Substrate



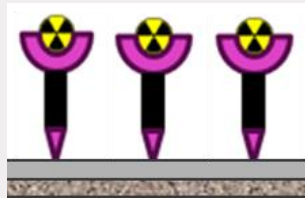
Emitted particles



Outline

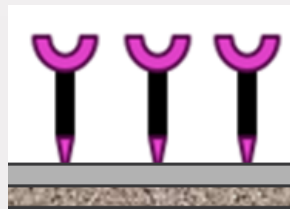
3) Radionuclide Binding

- Attach the radionuclide ^{152}Eu and ^{241}Am



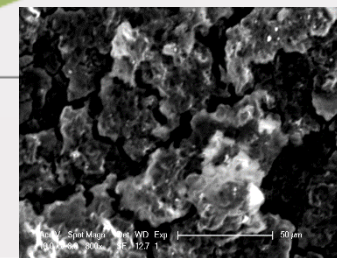
1) Functionalisation

- Attach functions to the surface
- 3 main methods



2) Chemical characterization

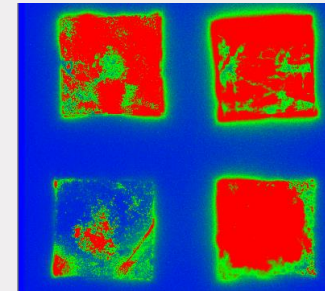
- SEM analyses (topology of the surface)
- Infrared Spectroscopy (Chemical bonds)



4) Radiological Characterization

- Uniformity
- Traceability
- Non-contamination

Uniform, traceable, large area surface sources ?



Functions on the surface



Manganese oxide/hydroxide

Manganese oxide layer

Ra^{2+} , UO_2^{2+}

- Reduction of Mn(VII)
- Precipitate as an oxide layer
- Not polymeric
- Brownish colored layer

(Ren et al, 2020)



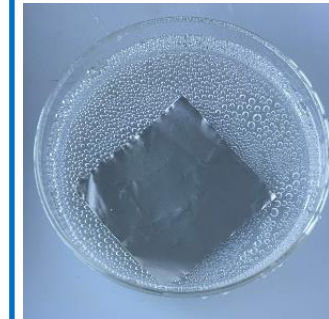
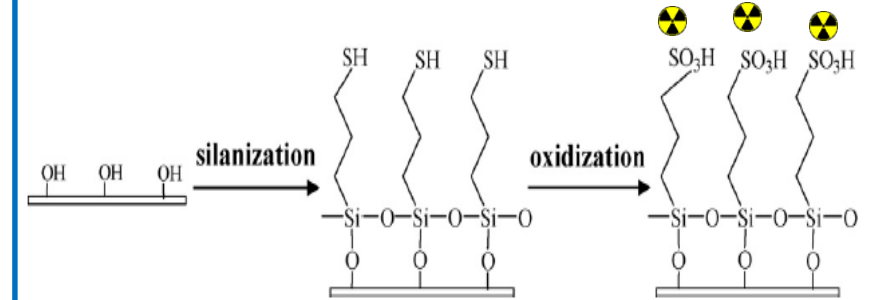
Chromium oxide/hydroxide

Chromium oxide layer

Ra^{2+} , UO_2^{2+}

- Reduction of Cr(VI)
- Inorganic polymer
- Soft-gel like mixture formed
- Yellow to dark brown color

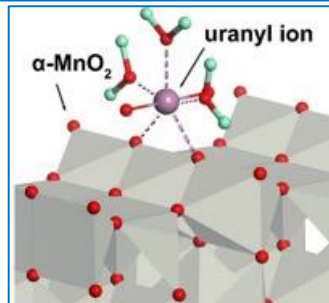
(Pommiers et. al. 2014)



Sulfonation Treatment

Eu^{3+} , Am^{3+}

- Greyish color
- (Gao et.al. 2017, Oudah et.a., 2018)

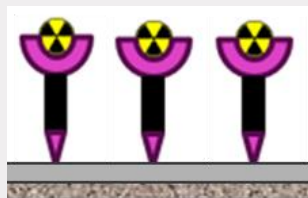


Act as cation exchangers to bind radioactive element

1st step

3) Radionuclide Binding

- Attach the radionuclide ^{152}Eu and ^{241}Am



1) Functionalisation

- Attach functions to the surface
- 3 main methods



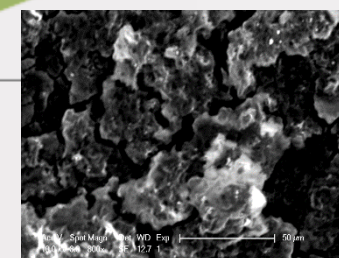
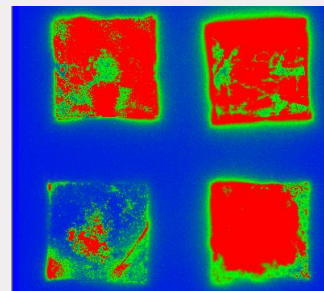
4) Radiological Characterization

- Uniformity
- Traceability
- Non-contamination

Uniform,
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area surface
sources ?

2) Chemical characterization

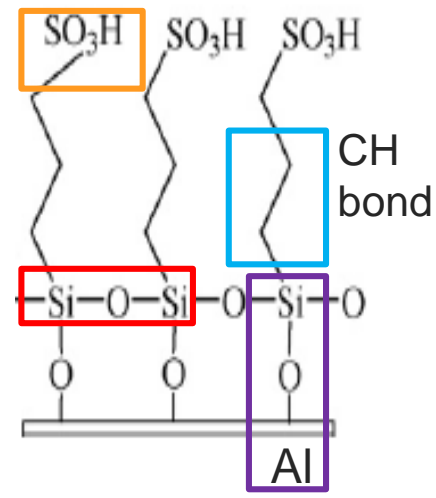
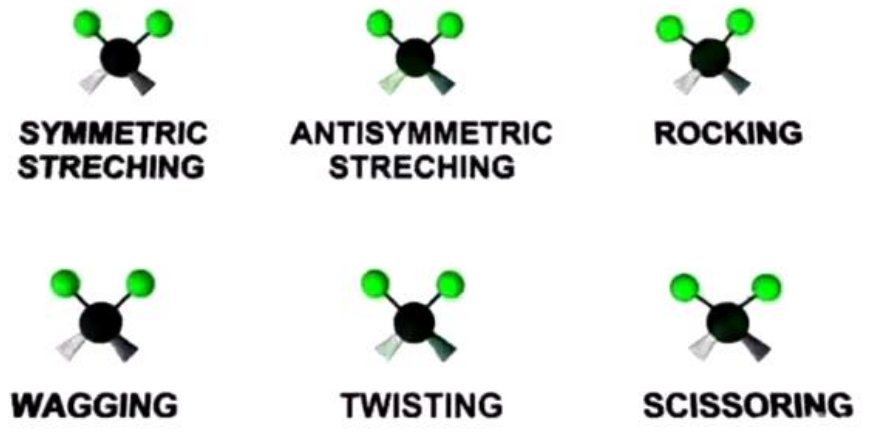
- SEM analyses (topology of the surface)
- Infrared Spectroscopy (Chemical bonds)



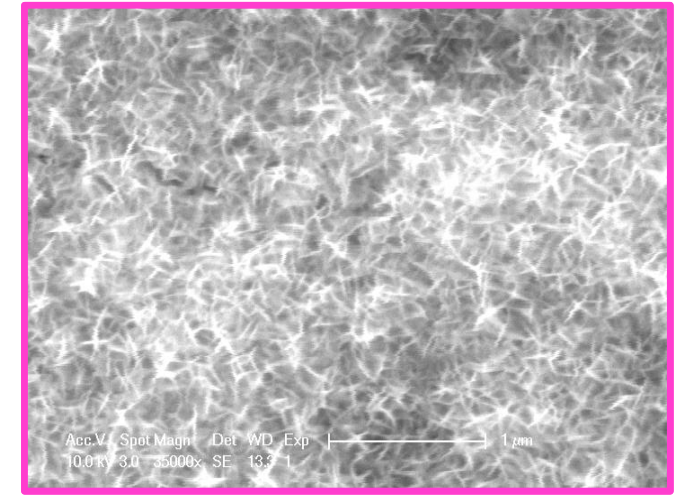
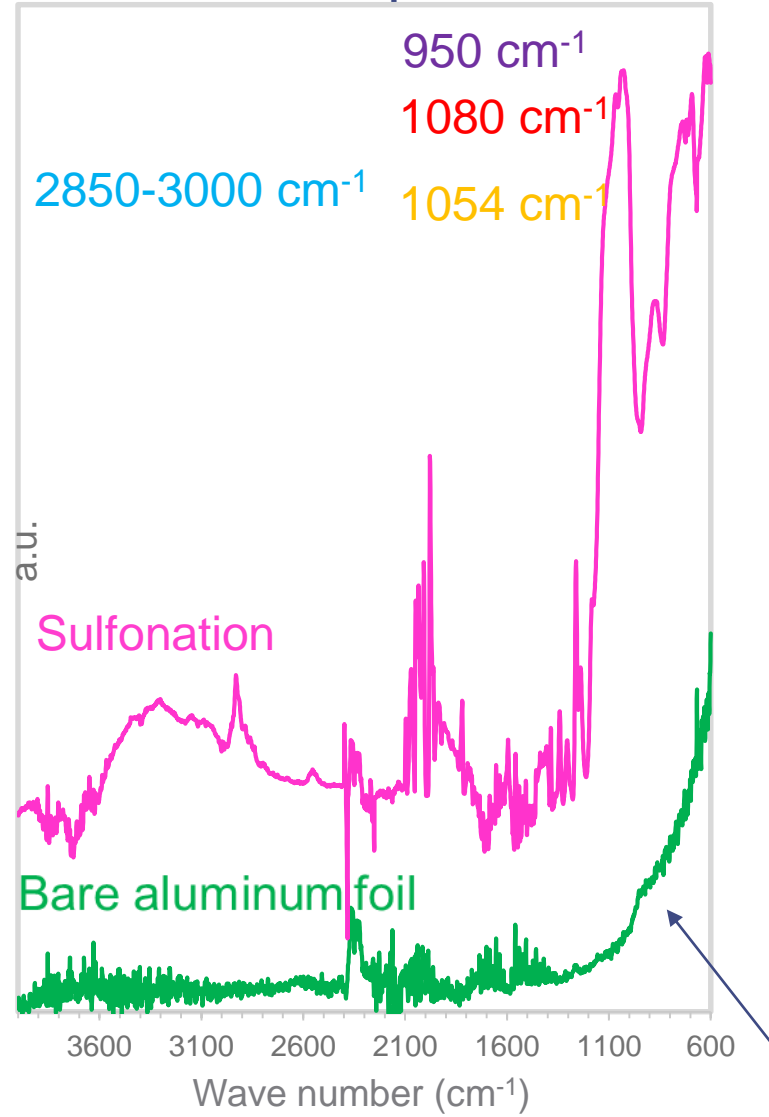
How effective is functionalisation?



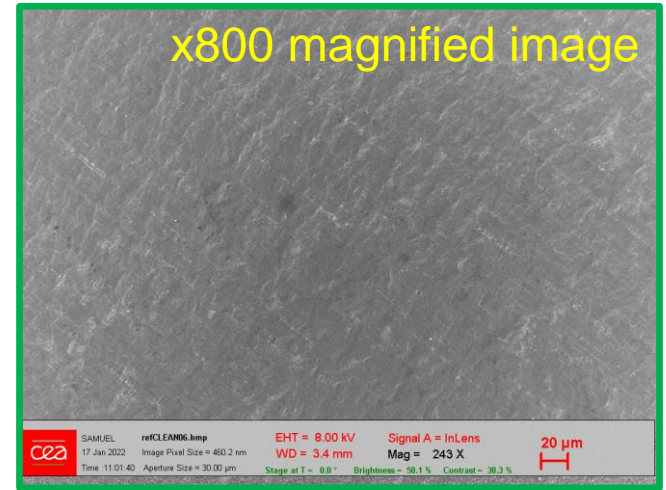
3D animation of vibrations in infrared spectroscopy



Infrared spectra



Magnification: x800 x35000

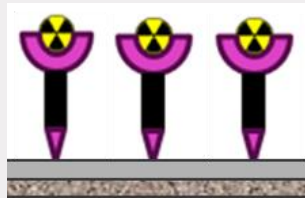


Al-OH bonds

2nd step

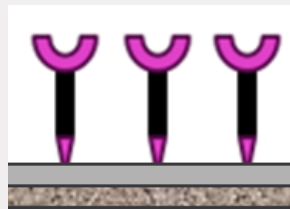
3) Radionuclide Binding

- Attach the radionuclide ^{152}Eu and ^{241}Am



1) Functionalisation

- Attach functions to the surface
- 3 main methods



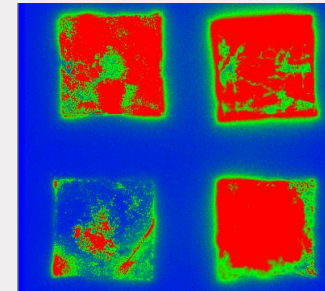
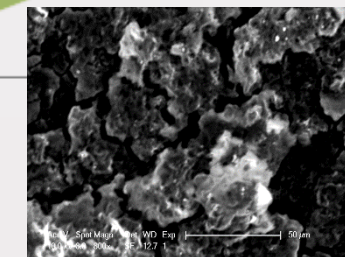
4) Radiological Characterization

- Uniformity
- Traceability
- Non-contamination

Uniform,
traceable, large
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sources ?

2) Chemical characterization

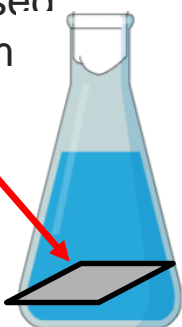
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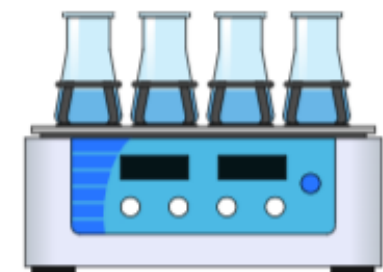
Radionuclide binding experiment

Functionalised aluminium

25 cm²

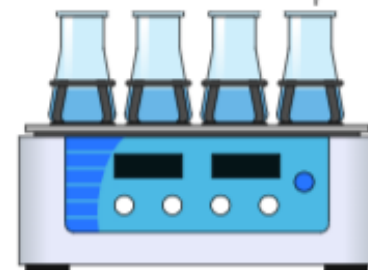


Radionuclide
(¹⁵²Eu or ²⁴¹Am) medium
Condition: pH 2



Under agitation 4 hours

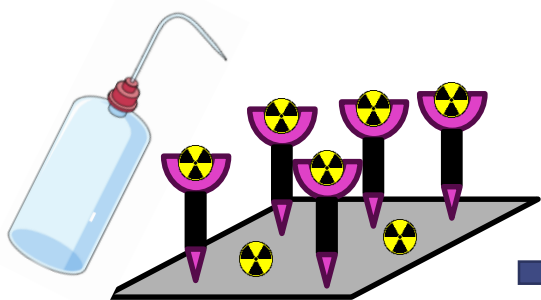
1 mL from the
radionuclide
solution



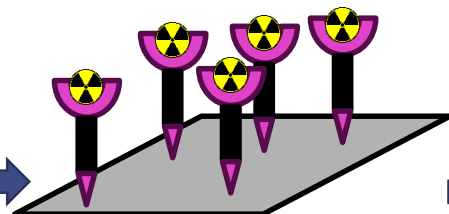
Sampling from the
radionuclide medium



Weighing the radioactive
source



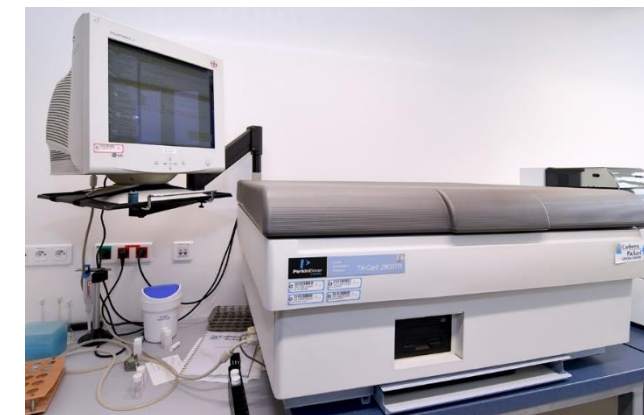
Rinsed with water



Radioactive sample

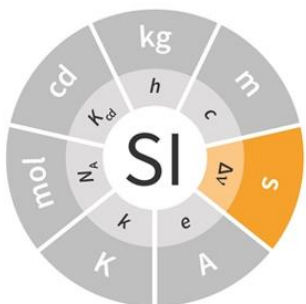


Digital autoradiography with imaging plate



Liquid scintillation counter

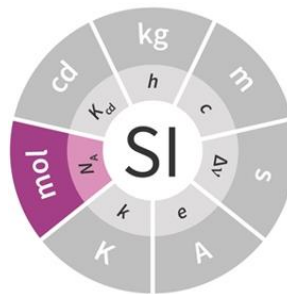
Experimental conditions



Activity (Bq = s⁻¹)

$$n = \frac{A \cdot T_{1/2}}{N_A \cdot \ln 2}$$

Amount of substance (mol)



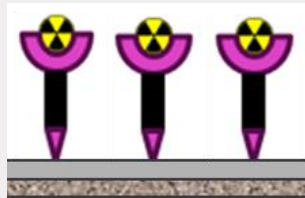
	²⁴¹ Am	¹⁵² Eu
Half-life (<i>t</i> _{1/2})	432.2 y	13.54 y
Emitter type	α, γ	β, γ

Samples	Radionuclide	Stable/Active element ratio	Total amount of element in solution (picomol)
Sulfonation	¹⁵² Eu	~ 57	~ 0.4
Chromium oxide layer	¹⁵² Eu	~ 57	~ 0.4
Manganese oxide layer	¹⁵² Eu	~ 57	~ 0.4
Manganese oxide layer	²⁴¹ Am	No carrier	~ 0.5

3rd step

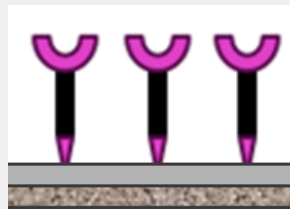
3) Radionuclide Binding

- Attach the radionuclide ^{152}Eu and ^{241}Am



1) Functionalisation

- Attach functions to the surface
- 3 main methods



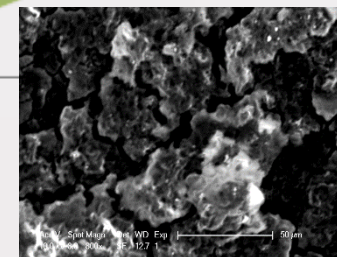
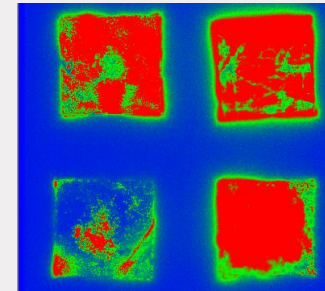
4) Radiological Characterization

- Uniformity
- Traceability
- Non-contamination

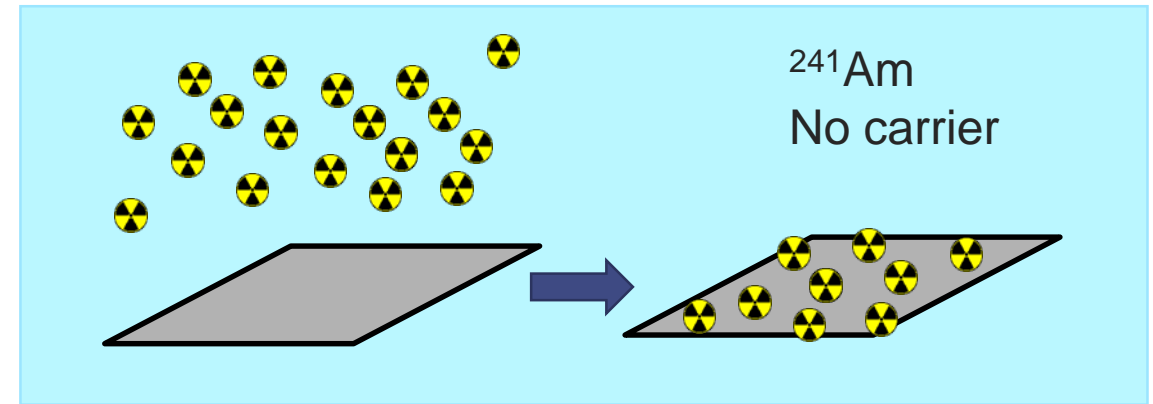
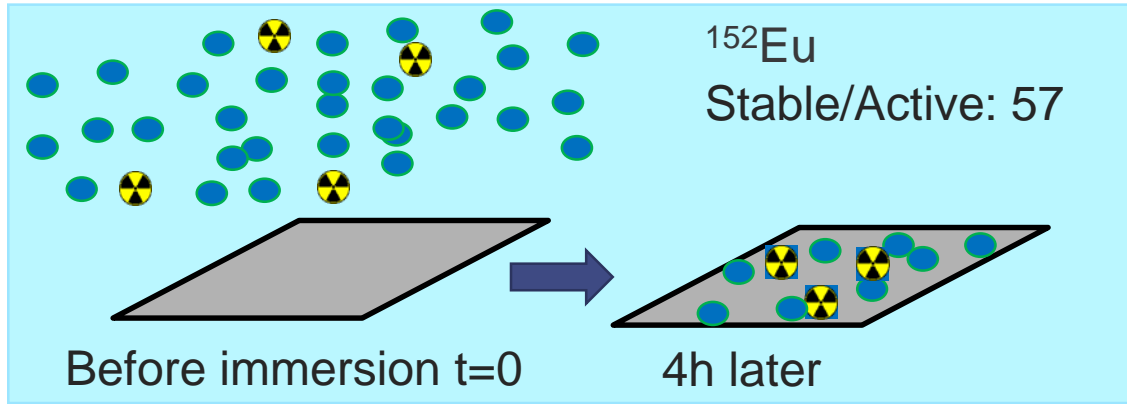
Uniform, traceable, large area surface sources ?

2) Chemical characterization

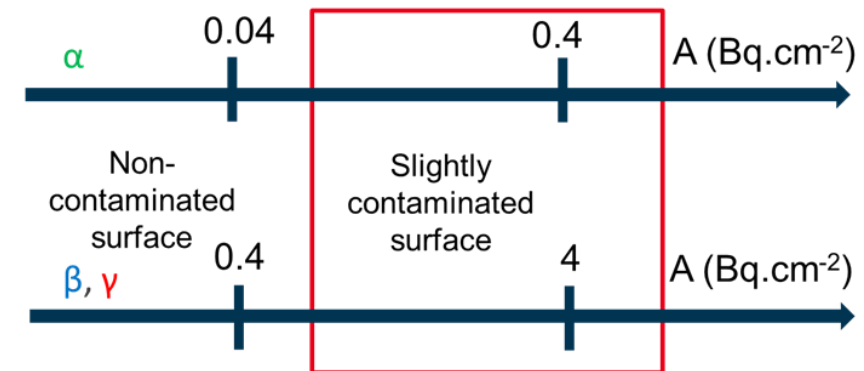
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Activity of the sources



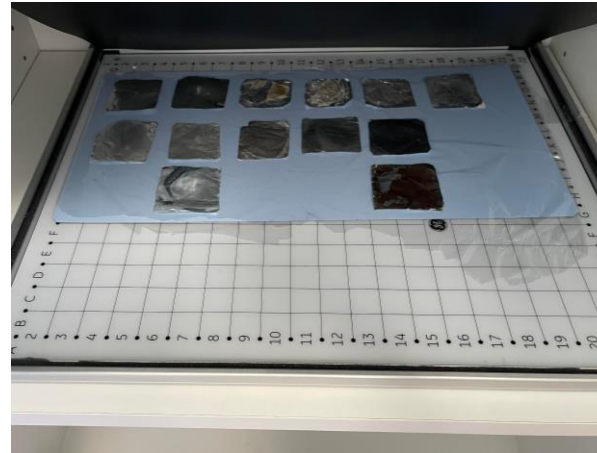
Samples	RN	% LSC activity decrease	Max nber of moles on the surface (picomol)	Max activity bound ($\text{Bq}\cdot\text{cm}^{-2}$)
Sulfonation	^{152}Eu	$(15 \pm 1)\%$	~ 0.07	3.5 ± 0.3
Cr oxide	^{152}Eu	$(19 \pm 1)\%$	~ 0.07	5.0 ± 0.5
Mn oxide	^{152}Eu	$(8.0 \pm 0.5)\%$	~ 0.03	2.0 ± 0.2
Mn oxide	^{241}Am	$(70 \pm 5)\%$	~ 0.4	18.5 ± 2



Autoradiography imaging

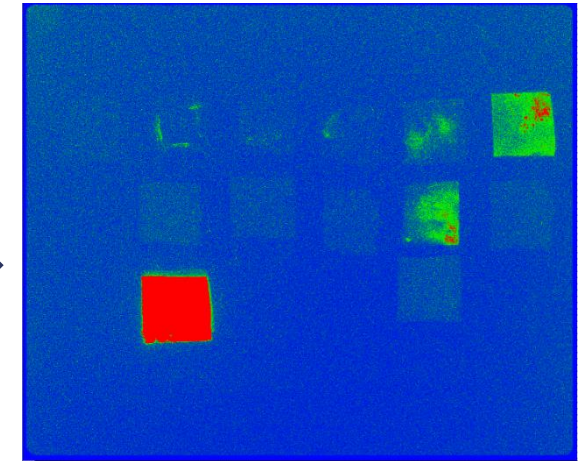


Digital autoradiography
(Amersham™ Typhoon™)



Sources on the screen

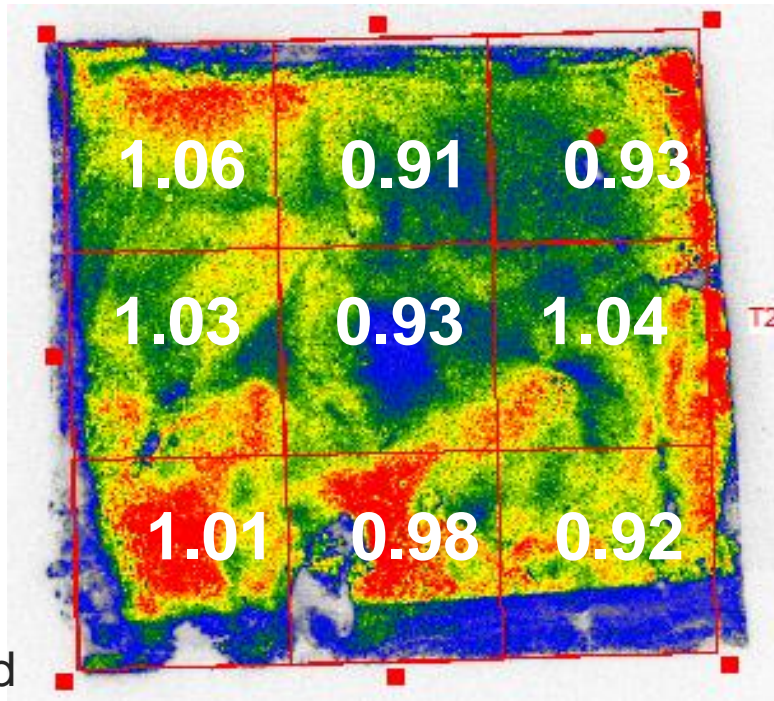
- Close contact with screen for exposure



Autoradiography images

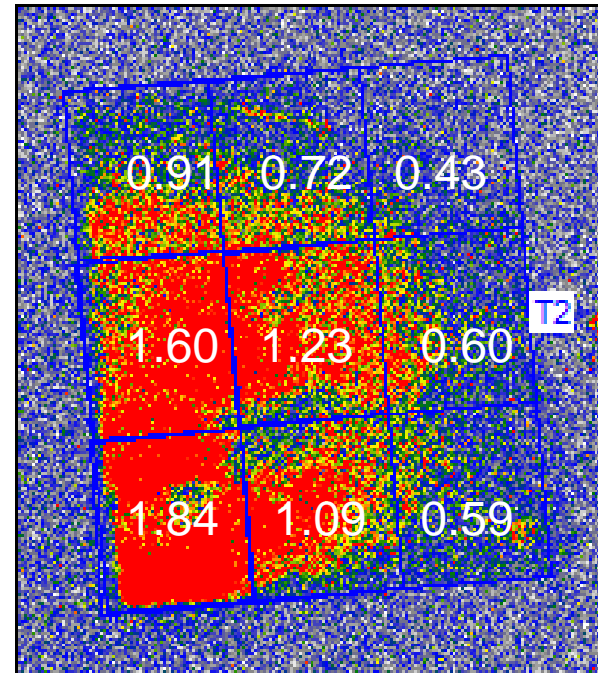
Autoradiography imaging

^{241}Am source
18 Bq/cm²



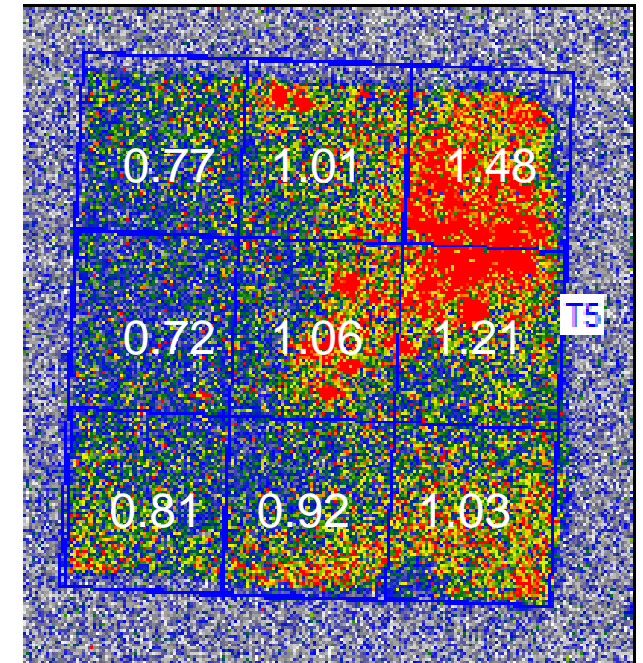
Manganese oxide layer
Uniformity (standard dev.): **95%**

^{152}Eu source
5 Bq/cm²



Chromium oxide layer
Uniformity: ~50%

^{152}Eu source
2 Bq/cm²

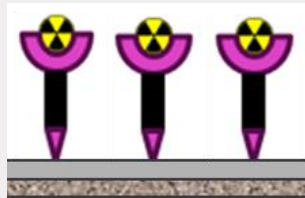


Sulfonation
Uniformity: ~80%

Last step

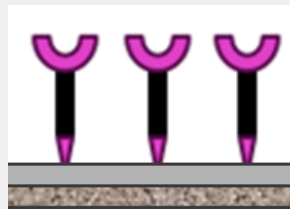
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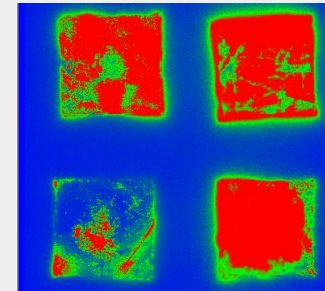


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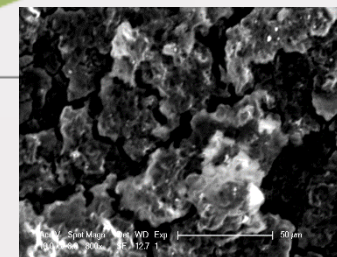
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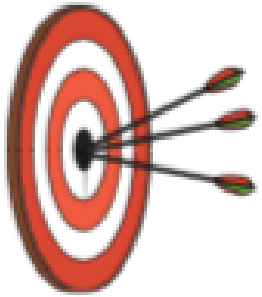
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Uniform,
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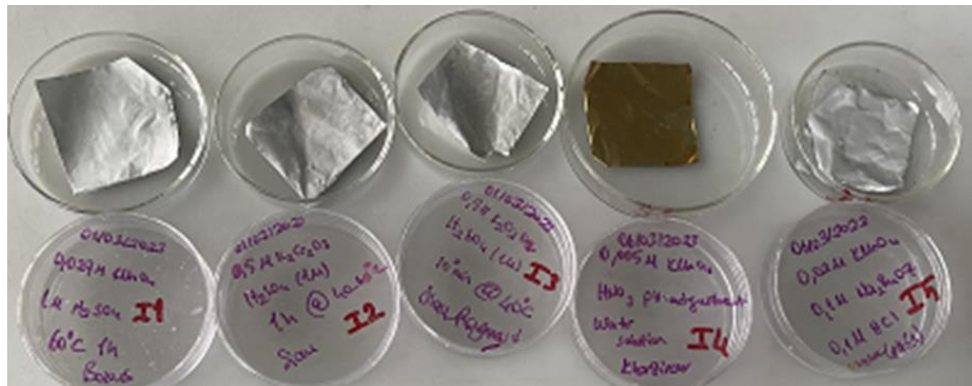


Conclusions & future work



ACHIEVEMENTS

- 3 different source production methods
- Surface activities from 2-20 Bq/cm²:
tunable
- ²⁴¹Am and ¹⁵²Eu sources
- Some uniform sources (>90%)



IMPROVEMENTS

- Uniformity: by using 0.5 mm Al foils
- ²⁴⁴Cm binding (α)
- Non-contamination tests to be performed

