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## Novel production method for traceable surface sources by aluminium functionalisation

Dilan Tuzun, V. Lourenço, Lucille Chambon, Yann Kergadallan

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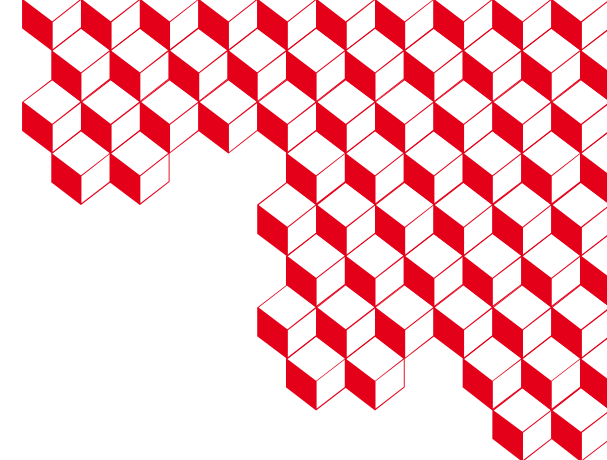
Submitted on 6 Nov 2023

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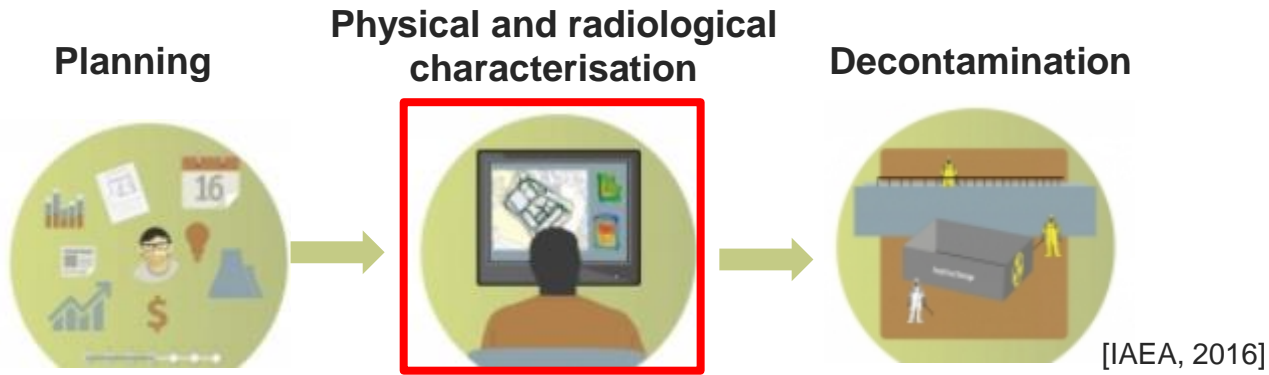
# Novel production method for traceable surface sources by aluminium functionalisation

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

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

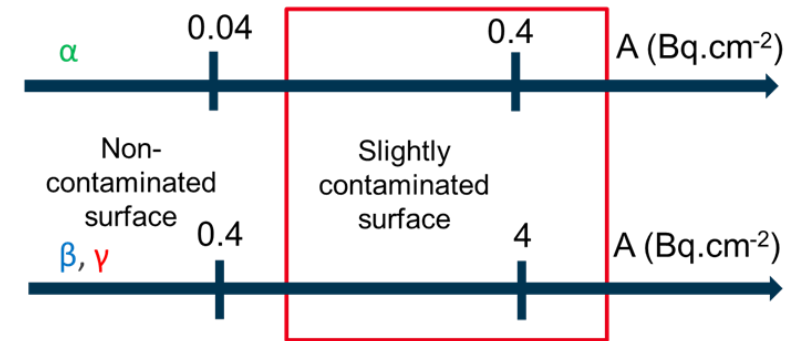


# Aim: Measuring radioactivity on complex surfaces



## 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:

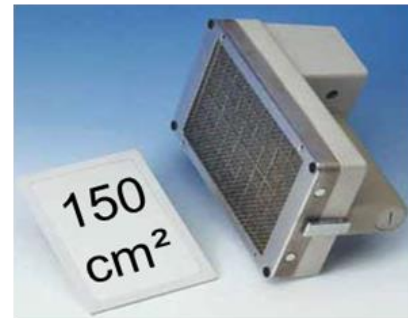
- Reliable contamination measurements
- Calibration of measurement systems

## Aim:

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



Commercial source

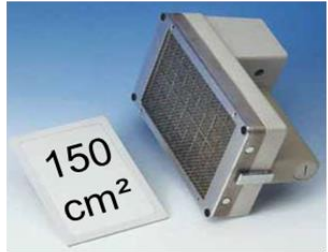


Provide metrologically traceable standard surface sources of various shapes

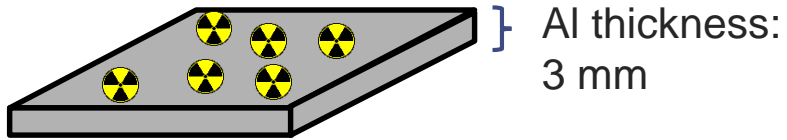
# Our approach: chemical attachment



Commercial source



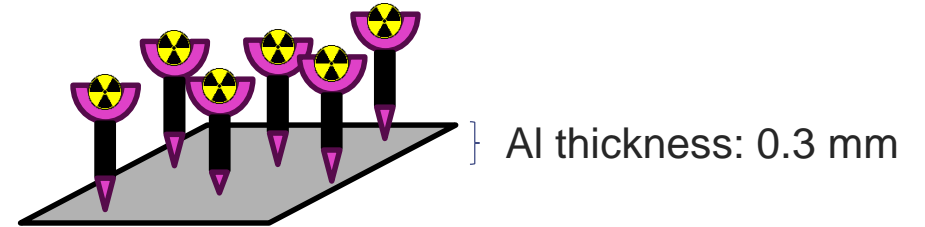
Commercial sources :  
Radionuclide dried + protected



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 ( $\alpha$ ,  $\beta$ )

## Chosen substrates:

- **Aluminium foil**
- Polymeric surfaces



Radionuclide

Surface functionalisation

Substrate



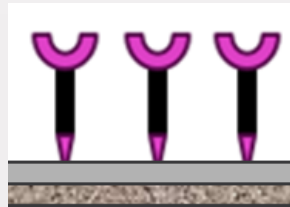
Emitted particles



# Outline

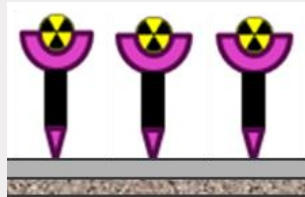
## 1) Functionalisation

- Attach functions to the surface
- 3 main methods



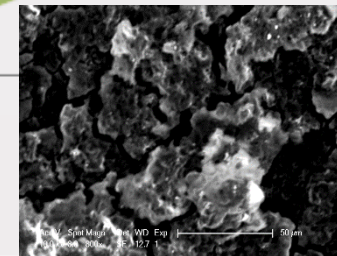
## 3) Radionuclide Binding

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



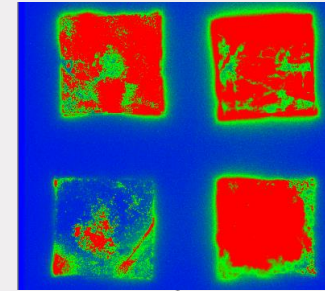
## 2) Chemical characterization

- 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)



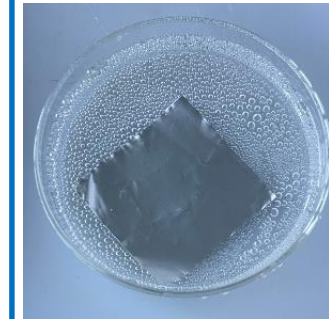
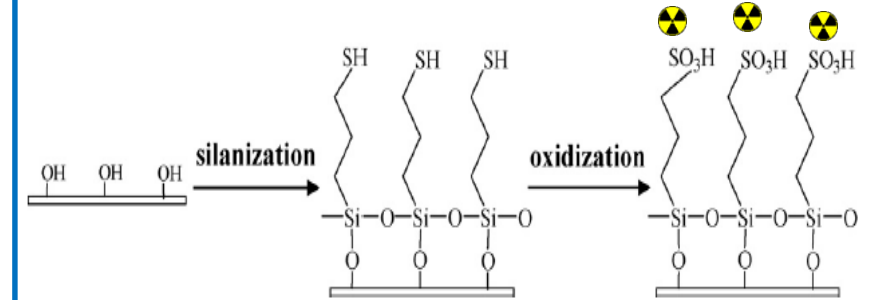
Chromium oxide/hydroxide

## Chromium oxide layer

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

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

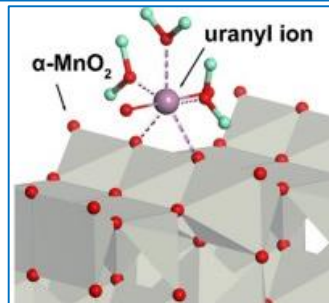
(Pommiers et. al. 2014)



## Sulfonation Treatment

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

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



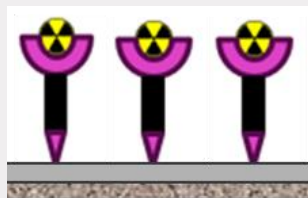
Act as cation exchangers to bind radioactive element



# 1<sup>st</sup> step

## 3) Radionuclide Binding

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



## 1) Functionalisation

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- 3 main methods



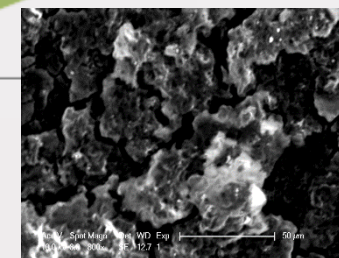
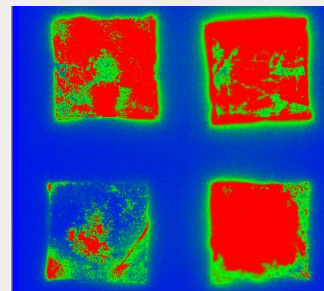
## 4) Radiological Characterization

- Uniformity
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Uniform,  
traceable, large  
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sources ?

## 2) Chemical characterization

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

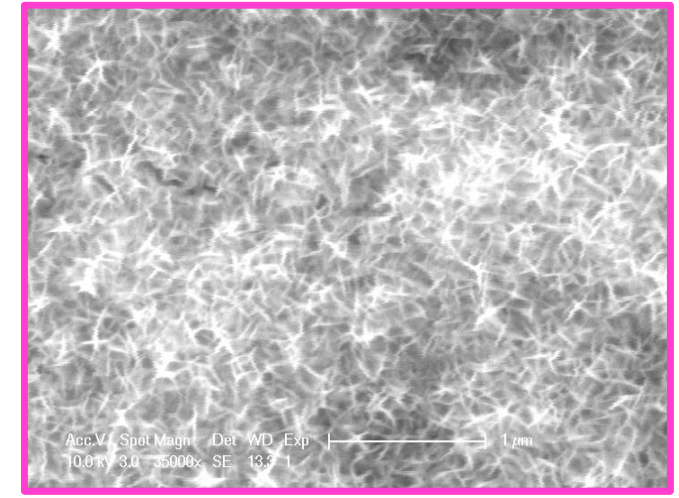
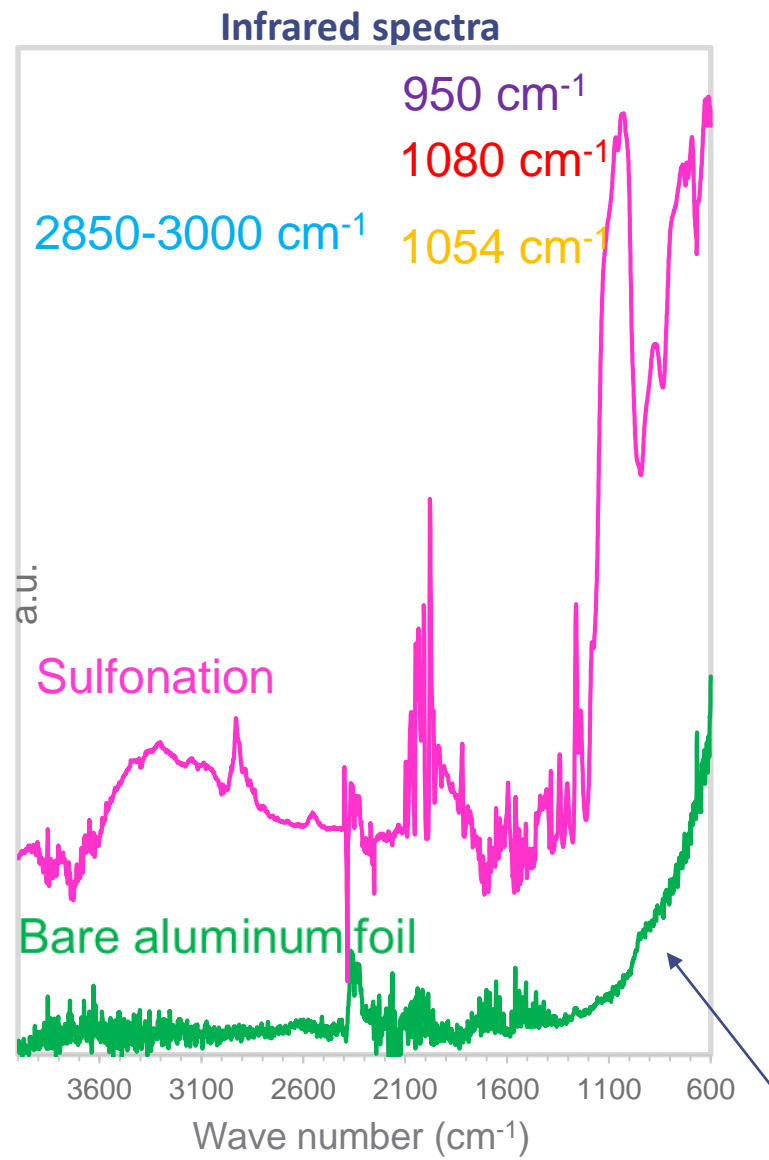
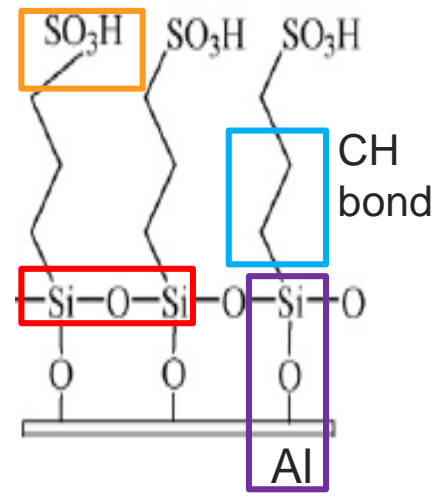
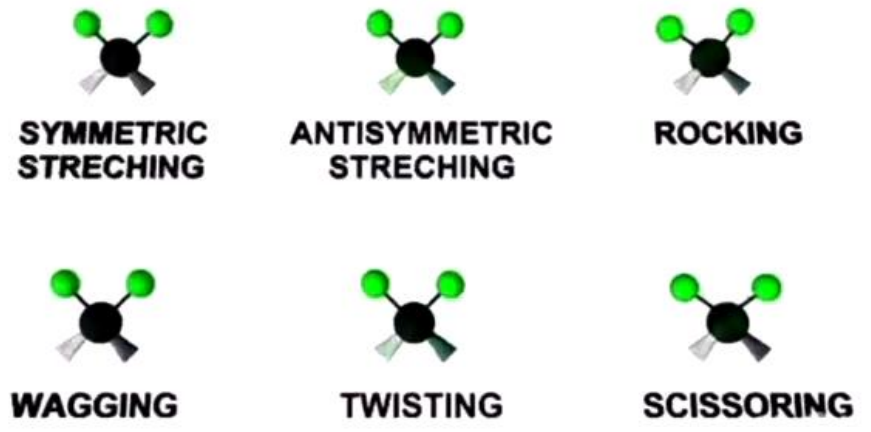




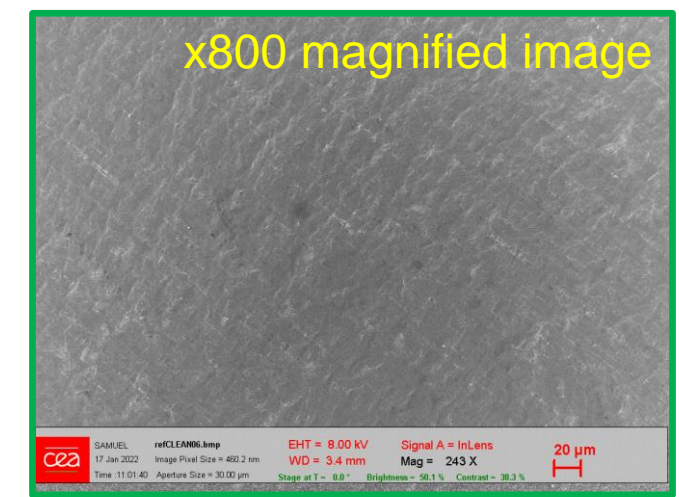
# How effective is functionalisation?



3D animation of vibrations in infrared spectroscopy



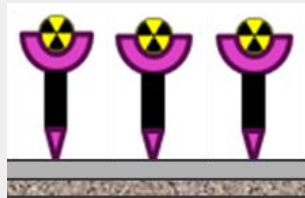
Magnification: x800 x35000



# 2<sup>nd</sup> step

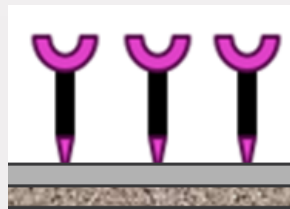
## 3) Radionuclide Binding

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



## 1) Functionalisation

- Attach functions to the surface
- 3 main methods



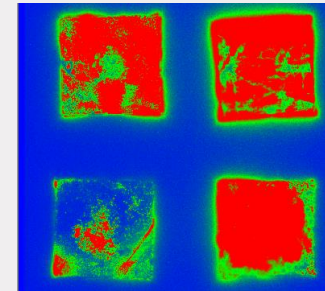
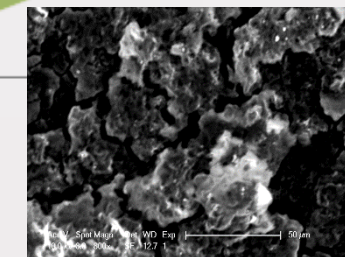
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Uniform,  
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## 2) Chemical characterization

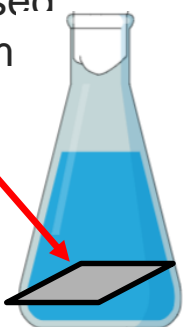
- SEM analyses (topology of the surface)
- Infrared Spectroscopy (Chemical bonds)



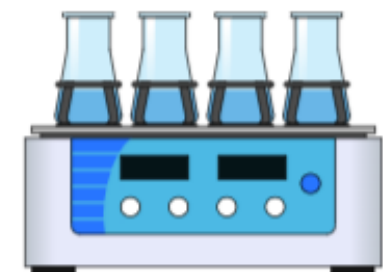
# Radionuclide binding experiment

Functionalised aluminium

25 cm<sup>2</sup>

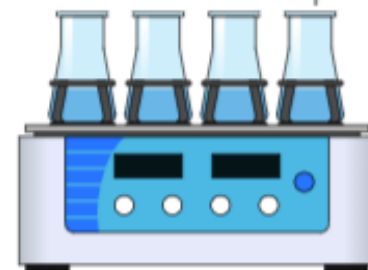


Radionuclide (<sup>152</sup>Eu or <sup>241</sup>Am) medium  
Condition: pH 2



Under agitation 4 hours

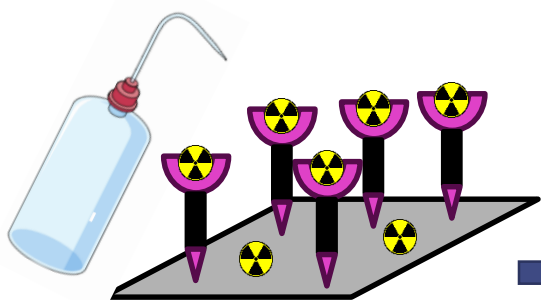
1 mL from the radionuclide solution



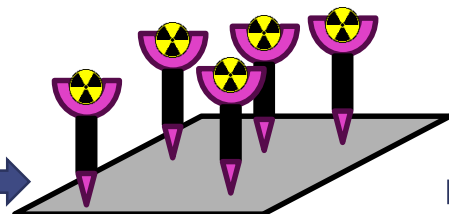
Sampling from the radionuclide medium



Weighing the radioactive source



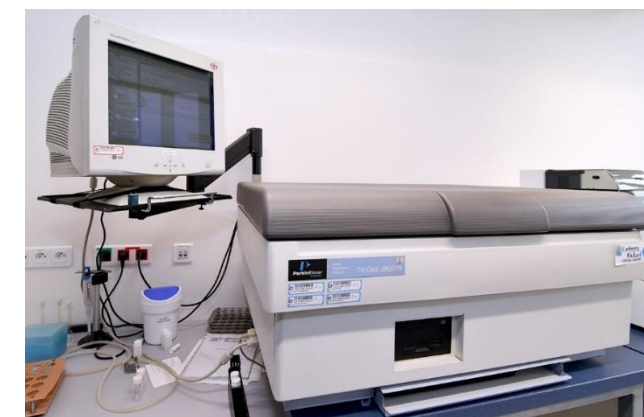
Rinsed with water



Radioactive sample

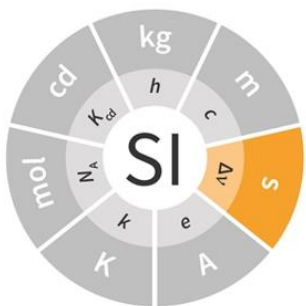


Digital autoradiography with imaging plate



Liquid scintillation counter

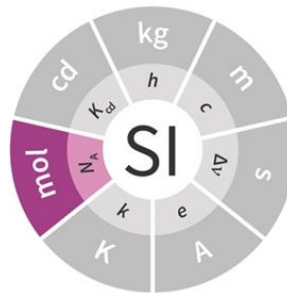
# Experimental conditions



Activity (Bq = s<sup>-1</sup>)

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

Amount of substance (mol)



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

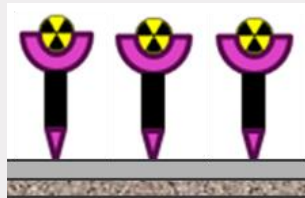
Samples	Radionuclide	Stable/Active element ratio	Total amount of element in solution (picomol)
Sulfonation	<sup>152</sup> Eu	~ 57	~ 0.4
Chromium oxide layer	<sup>152</sup> Eu	~ 57	~ 0.4
Manganese oxide layer	<sup>152</sup> Eu	~ 57	~ 0.4
Manganese oxide layer	<sup>241</sup> Am	No carrier	~ 0.5



# 3<sup>rd</sup> step

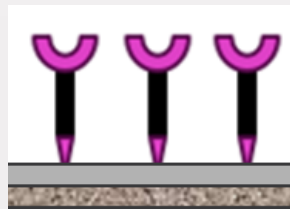
## 3) Radionuclide Binding

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



## 1) Functionalisation

- Attach functions to the surface
- 3 main methods



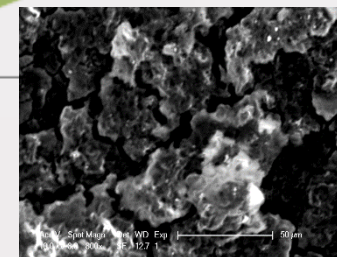
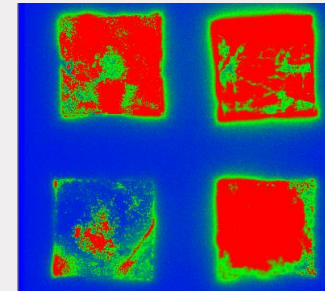
## 4) Radiological Characterization

- Uniformity
- Traceability
- Non-contamination

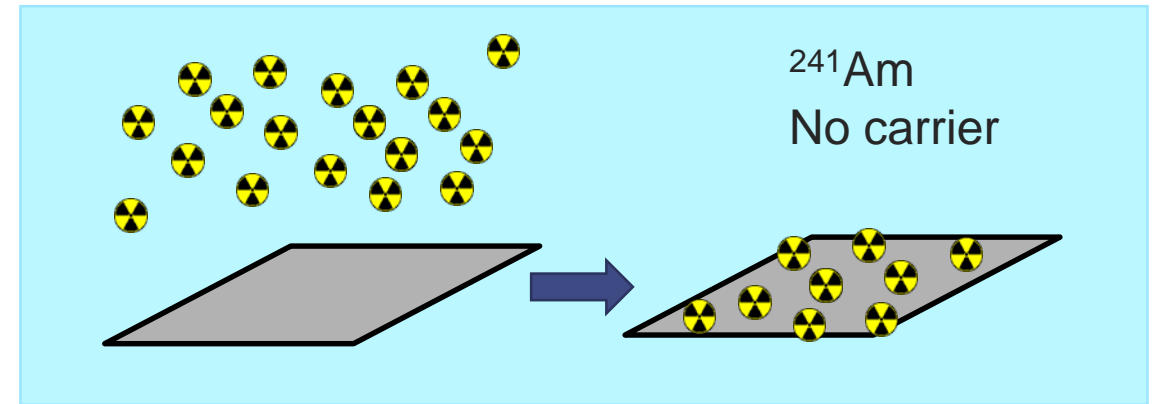
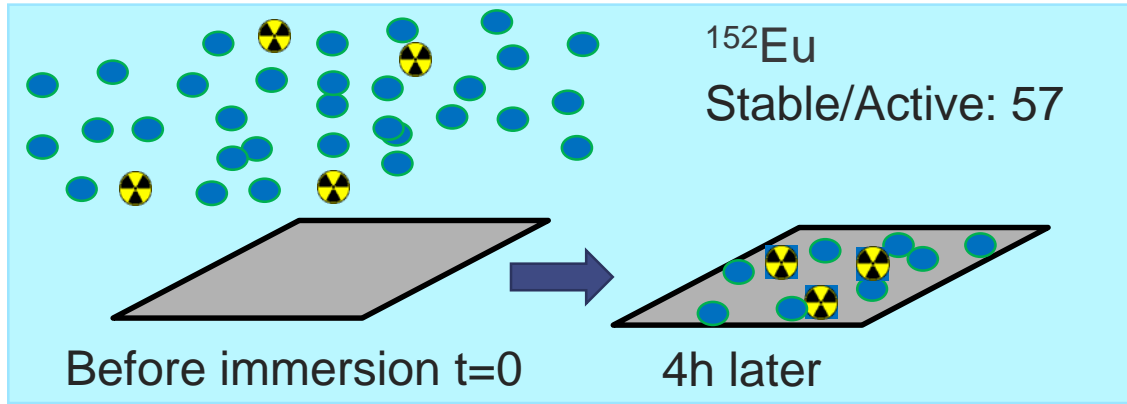
Uniform, traceable, large area surface sources ?

## 2) Chemical characterization

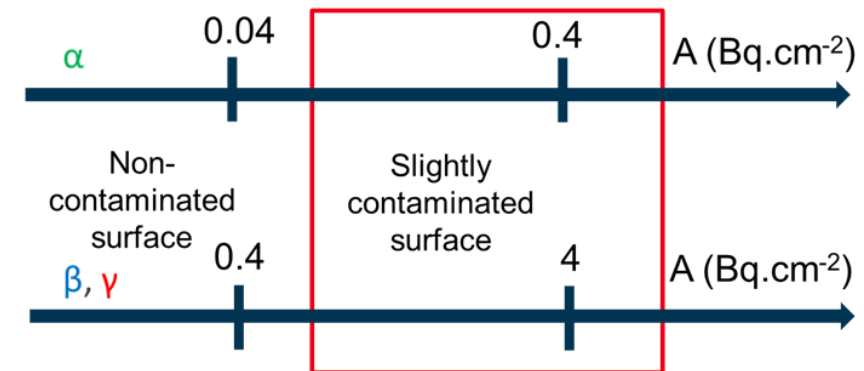
- SEM analyses (topology of the surface)
- Infrared Spectroscopy (Chemical bonds)



# Activity of the sources



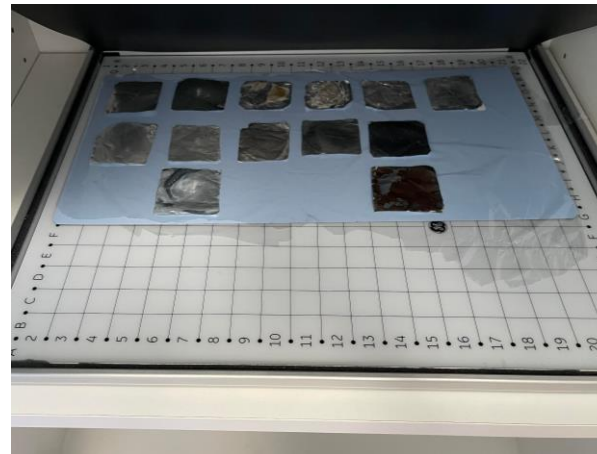
Samples	RN	% LSC activity decrease	Max nber of moles on the surface (picomol)	Max activity bound ( $\text{Bq}\cdot\text{cm}^{-2}$ )
Sulfonation	$^{152}\text{Eu}$	$(15 \pm 1)\%$	$\sim 0.07$	$3.5 \pm 0.3$
Cr oxide	$^{152}\text{Eu}$	$(19 \pm 1)\%$	$\sim 0.07$	$5.0 \pm 0.5$
Mn oxide	$^{152}\text{Eu}$	$(8.0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



# Autoradiography imaging

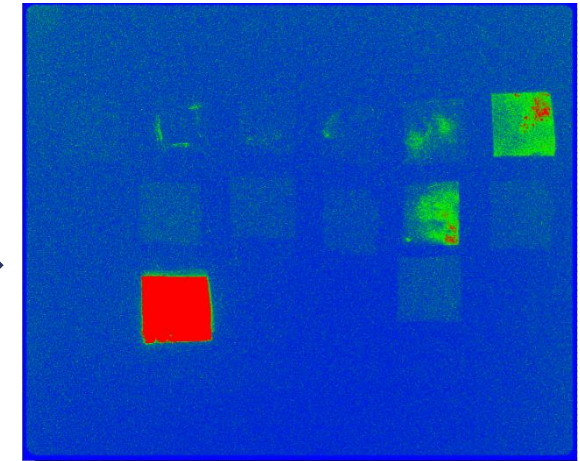


Digital autoradiography  
(Amersham™ Typhoon™)



Sources on the screen

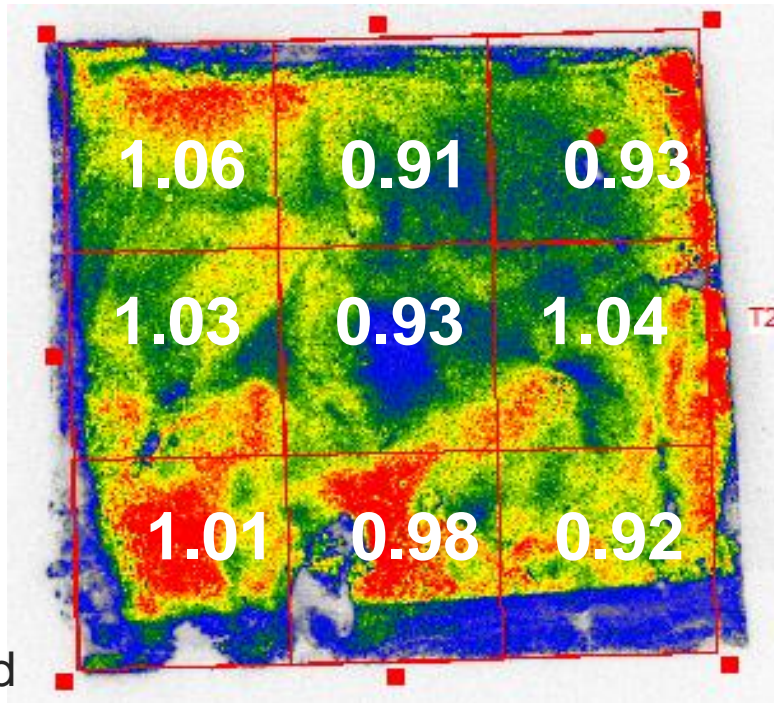
- Close contact with screen for exposure



Autoradiography images

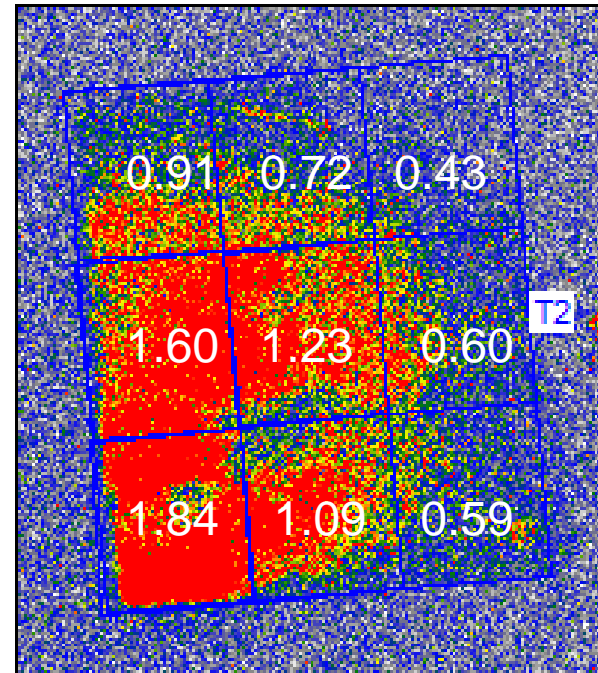
# Autoradiography imaging

$^{241}\text{Am}$  source  
18 Bq/cm<sup>2</sup>



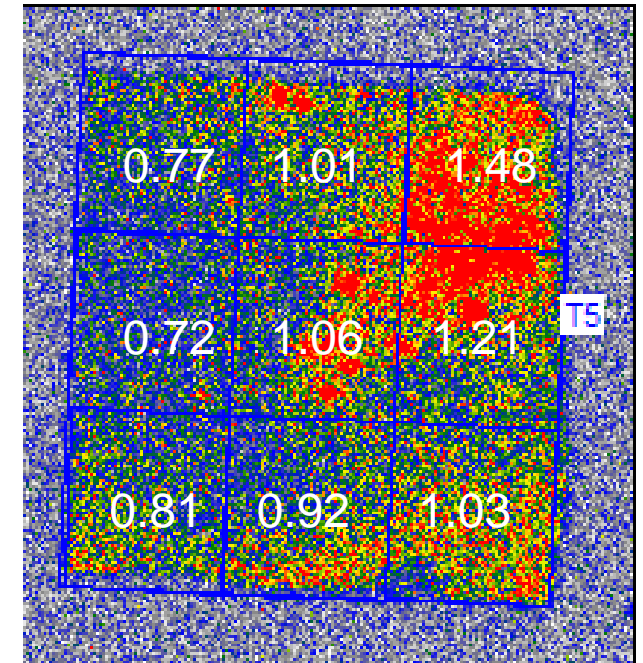
Manganese oxide layer  
Uniformity (standard dev.): **95%**

$^{152}\text{Eu}$  source  
5 Bq/cm<sup>2</sup>



Chromium oxide layer  
Uniformity: ~50%

$^{152}\text{Eu}$  source  
2 Bq/cm<sup>2</sup>



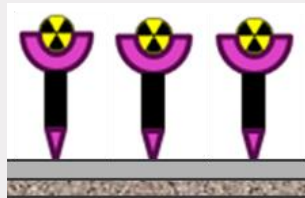
Sulfonation  
Uniformity: ~80%



# Last step

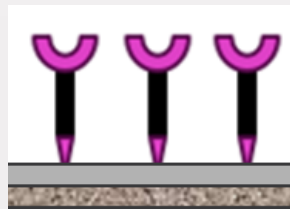
## 3) Radionuclide Binding

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



## 1) Functionalisation

- Attach functions to the surface
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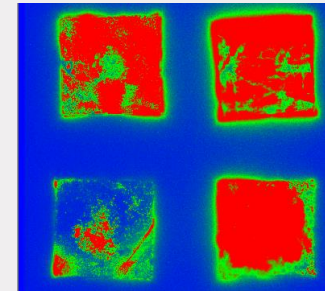


## 4) Radiological Characterization

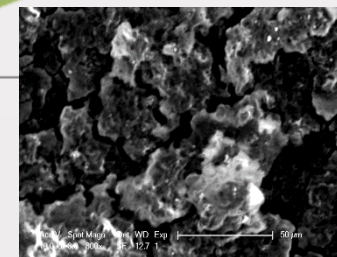
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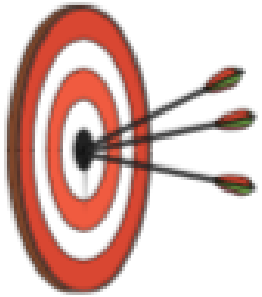
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Uniform,  
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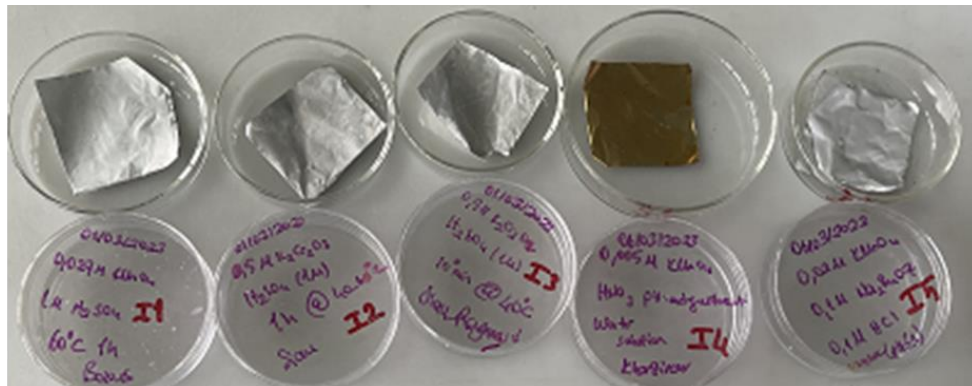


# Conclusions & future work



## ACHIEVEMENTS

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



## IMPROVEMENTS

- Uniformity: by using 0.5 mm Al foils
- <sup>244</sup>Cm binding ( $\alpha$ )
- Non-contamination tests to be performed

