Preparation and characterization of actinide targets for superheavy element production

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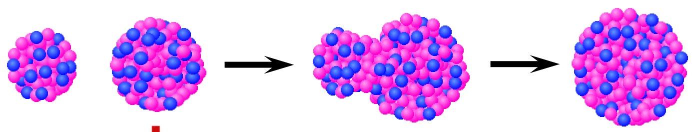
- Target production by Molecular Plating
- Target characterization
- Recovery and purification of target material
- Alternative target production techniques

SHE production with actinide targets



- E114 ⇒ $^{244}Pu(^{48}Ca,xn)$
- E115 ⇒ $^{243}Am(^{48}Ca,xn)$
- E116 ⇒ $^{248}Cm(^{48}Ca,xn)$
- E117 ⇒ $^{249}Bk(^{48}Ca,xn)$
- E119 ⇒ $^{249}Bk(^{50}Ti,xn)$
- E120 ⇒ $^{248}Cm(^{54}Cr,xn)$
- E120 ⇒ $^{249}Cf(^{50}Ti,xn)$

Target thickness:…...500 µg/cm^2

Target production technique:

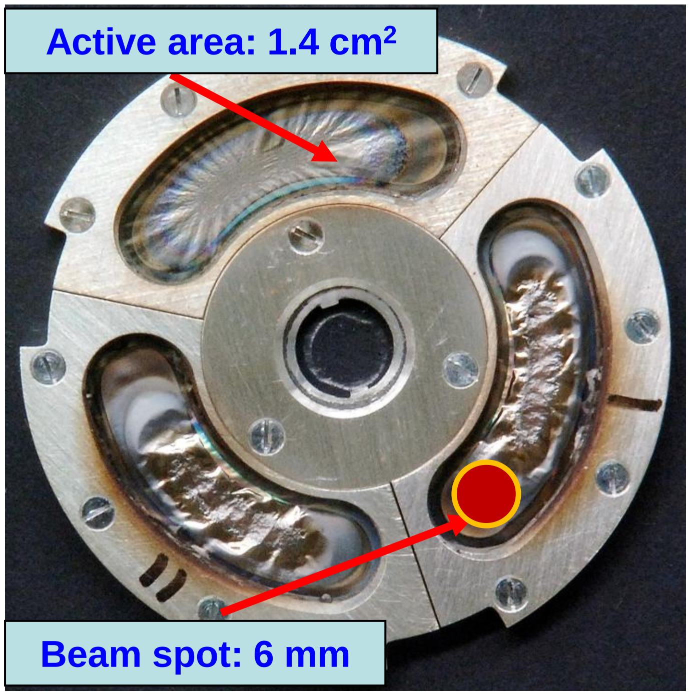
- Chemical purification prior to deposition (if required)
- Recovery and chemical purification of used target material (sooner or later……)
- Small and simple set-up
- Components easy to replace to avoid cross-contamination
- High deposition yield

Electrochemical deposition from organic media:

Molecular Plating

Rotating *TASCA* target wheels for high beam intensities

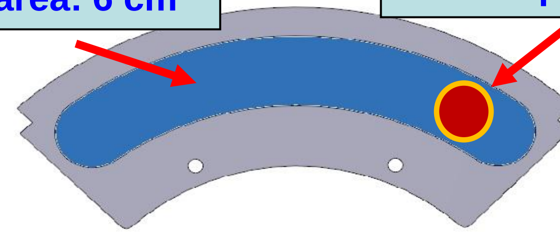
Arc-shaped target geometry → **New Design: 2010**

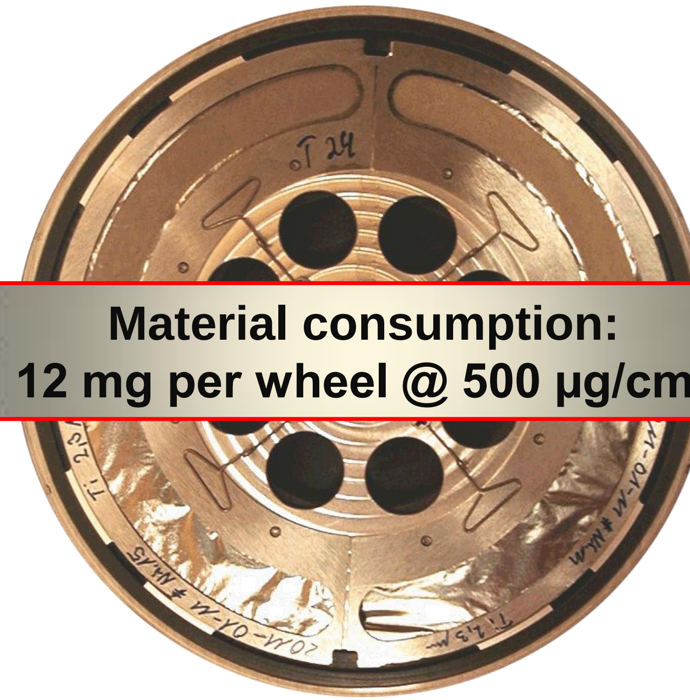


Material consumption:
2 mg per wheel @ 500 µg/cm²

Used in the production of element 114

Ch.E. Düllmann et al., Phys. Rev. Lett. 104 (2010) 252701





Used in search for element 120

To be used in the production of element 115

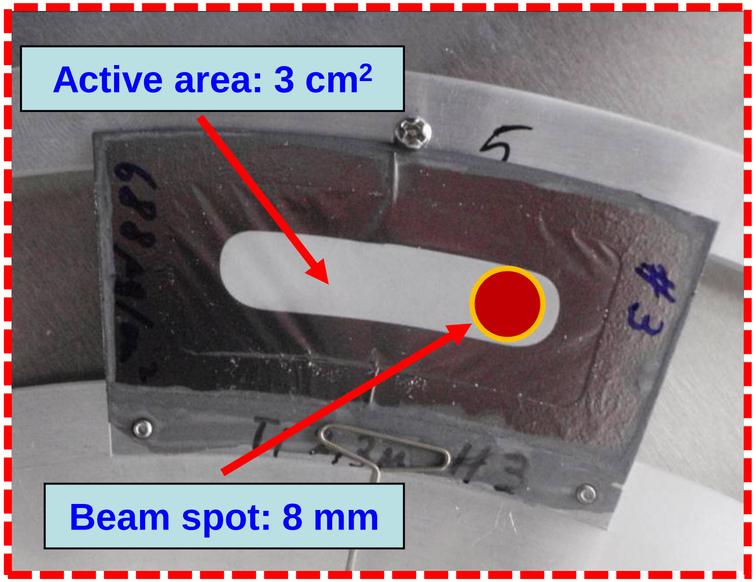
Rotating target wheel for SHIP

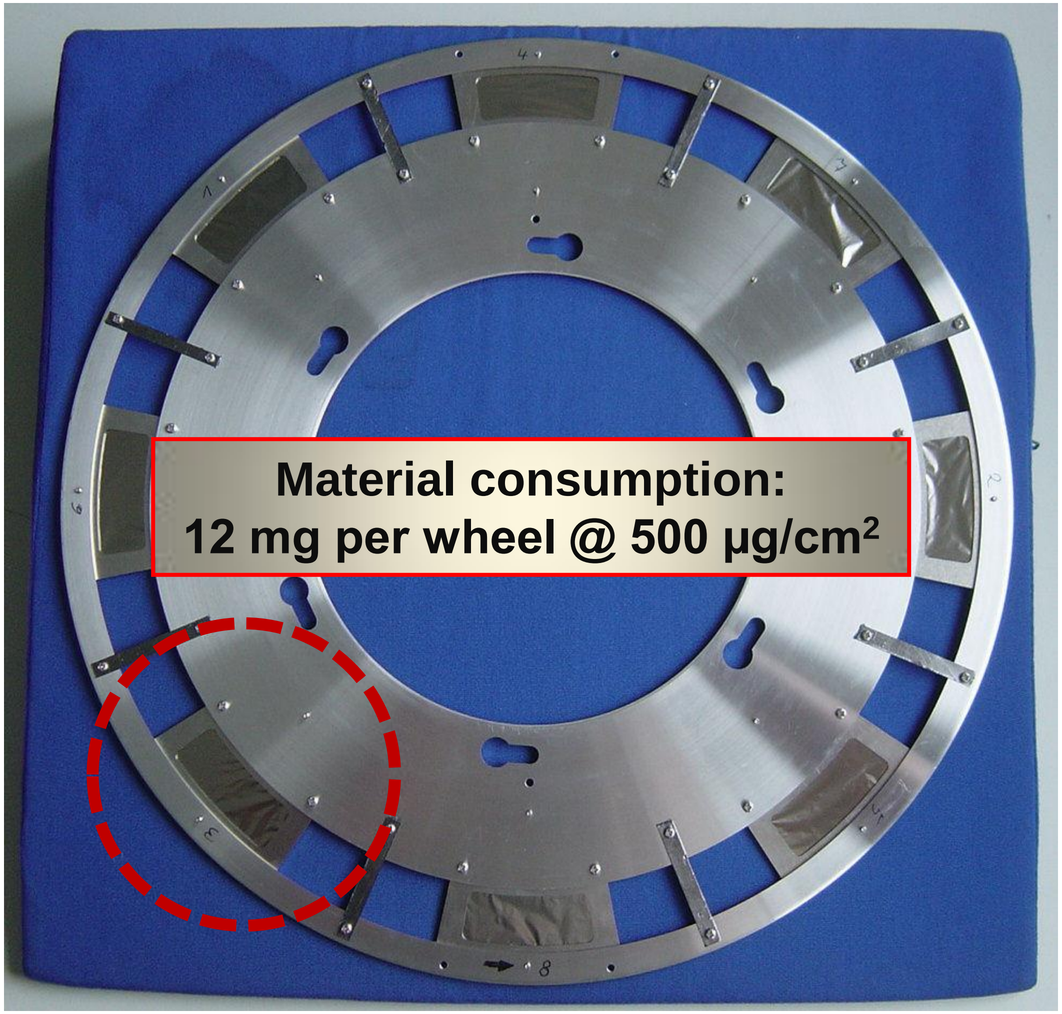
GSI target laboratory:

- 2 µm Ti-foils produced by cold rolling
- Foils are glued onto Al-frame

SHIP target wheel:

- Target area: 3 cm^2
- 8 targets per wheel



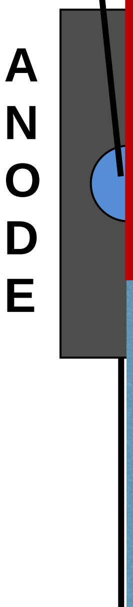


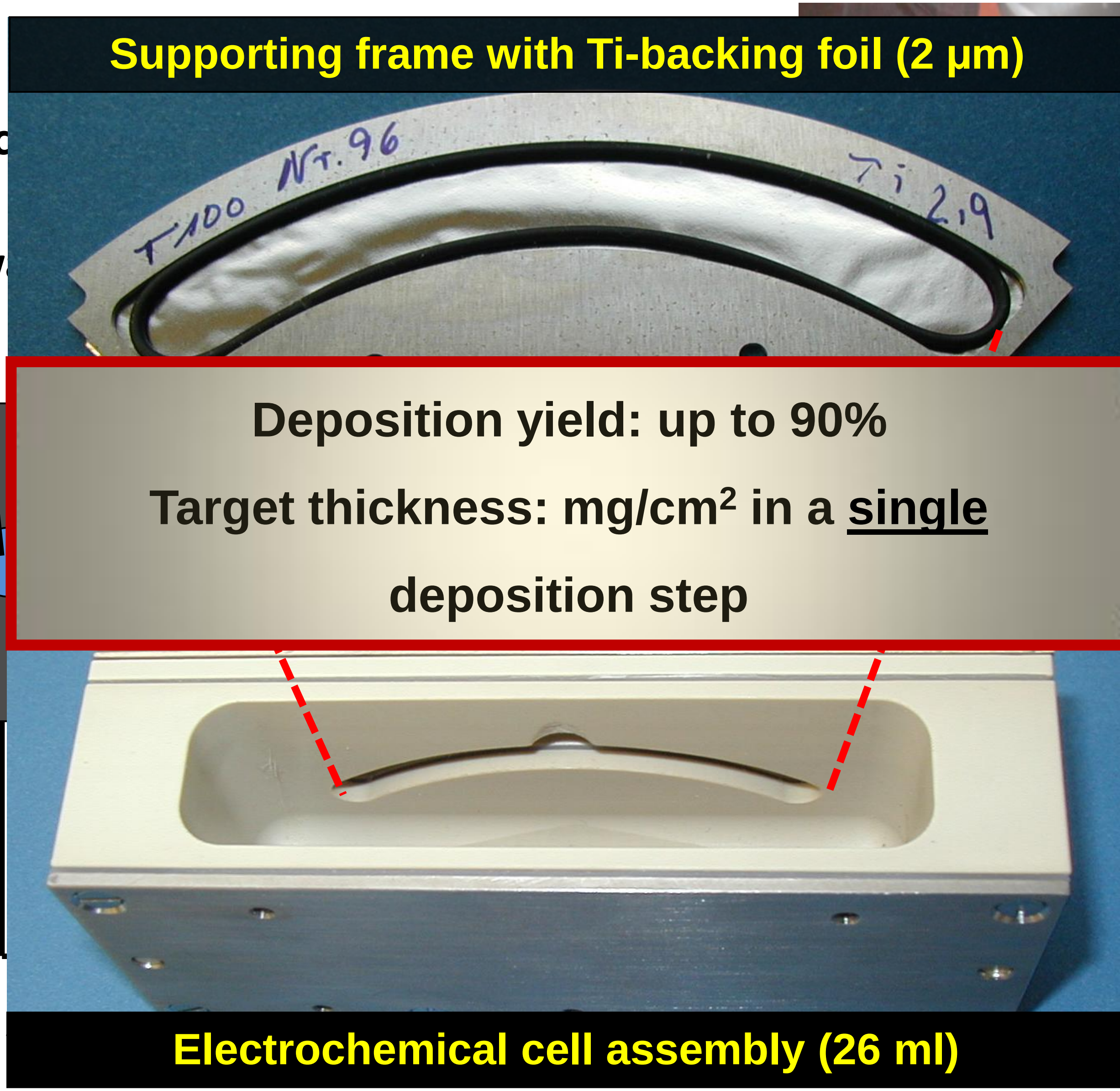
Used in the production of element 116
Used in search of element 120

Actinide deposition by Molecular Plating

Ultrasonic

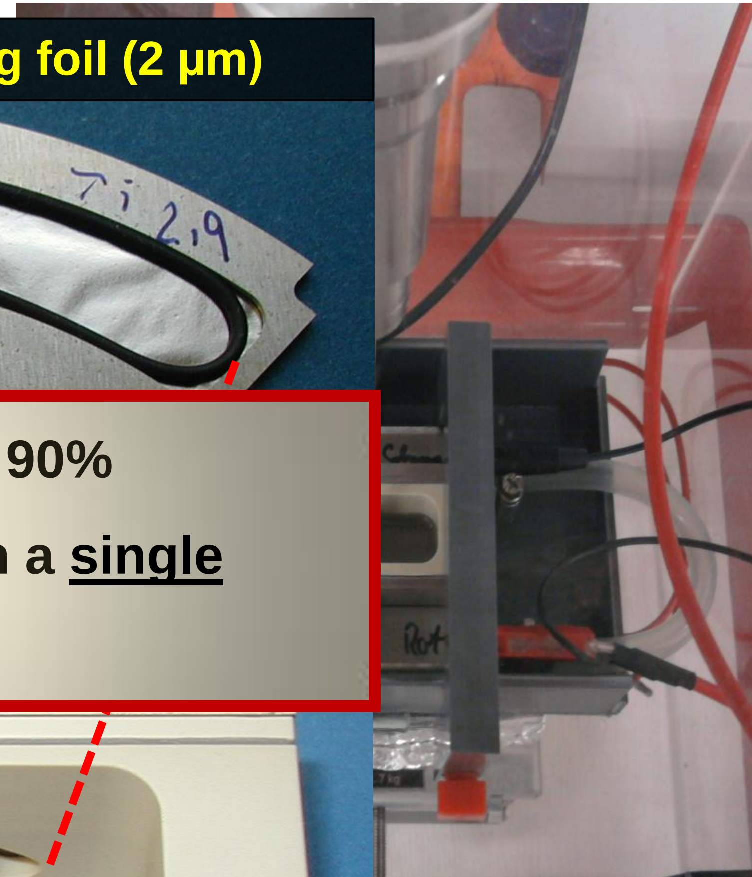
Cooling w





Deposition yield: up to 90%

Target thickness: mg/cm² in a <u>single</u> deposition step



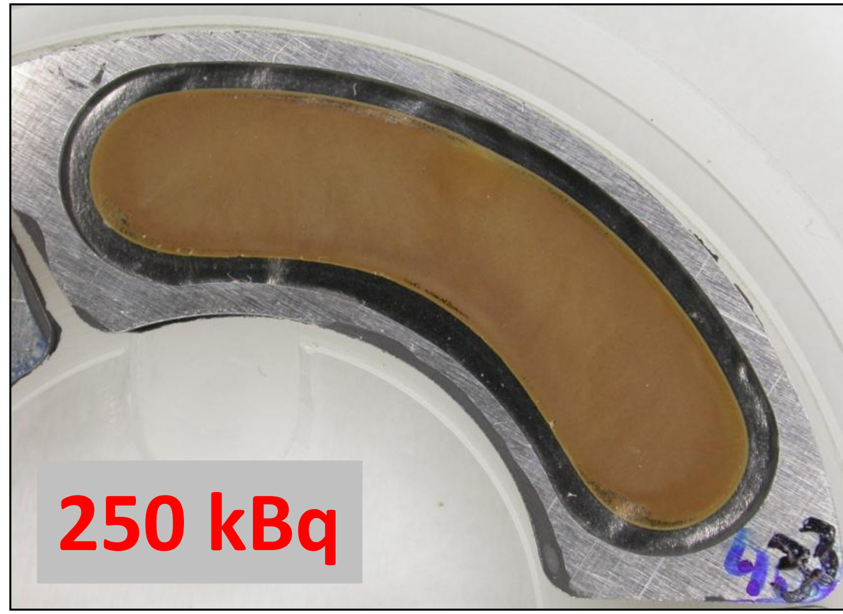
me:

Cell design according to H. Haba [RIKEN], TASCA05, Oslo, Oct. 2005

Deposition of Pu and Cm by Molecular Plating

Deposition conditions for Pu:

Solvent……………..Isobutanol
Plating time……….6 h
Voltage……………..150 – 200 V
Current density…..1.4 mA/cm^2
Stirring at………….1200 U/min
Temperature……….15 C



Deposition conditions for Cm:

Solvent……………..Isobutanol
Plating time……….3 h
Voltage……………..150 – 300 V
Current density…..0.7 mA/cm^2
Stirring at………….1200 U/min
Temperature……….15 C

Deposition of Pu and Cm by Molecular Plating

Pu	Thickness [µg/cm²]
^{244}Pu	401
^{244}Pu	502
^{244}Pu	490
^{244}Pu	390
^{244}Pu	472
^{244}Pu	673
^{244}Pu	724
^{244}Pu	790
^{244}Pu	785
^{239}Pu	337
^{239}Pu	420
^{239}Pu	367
^{239}Pu	350
^{239}Pu	115
^{239}Pu	340
^{239}Pu	425

Deposition of Pu:

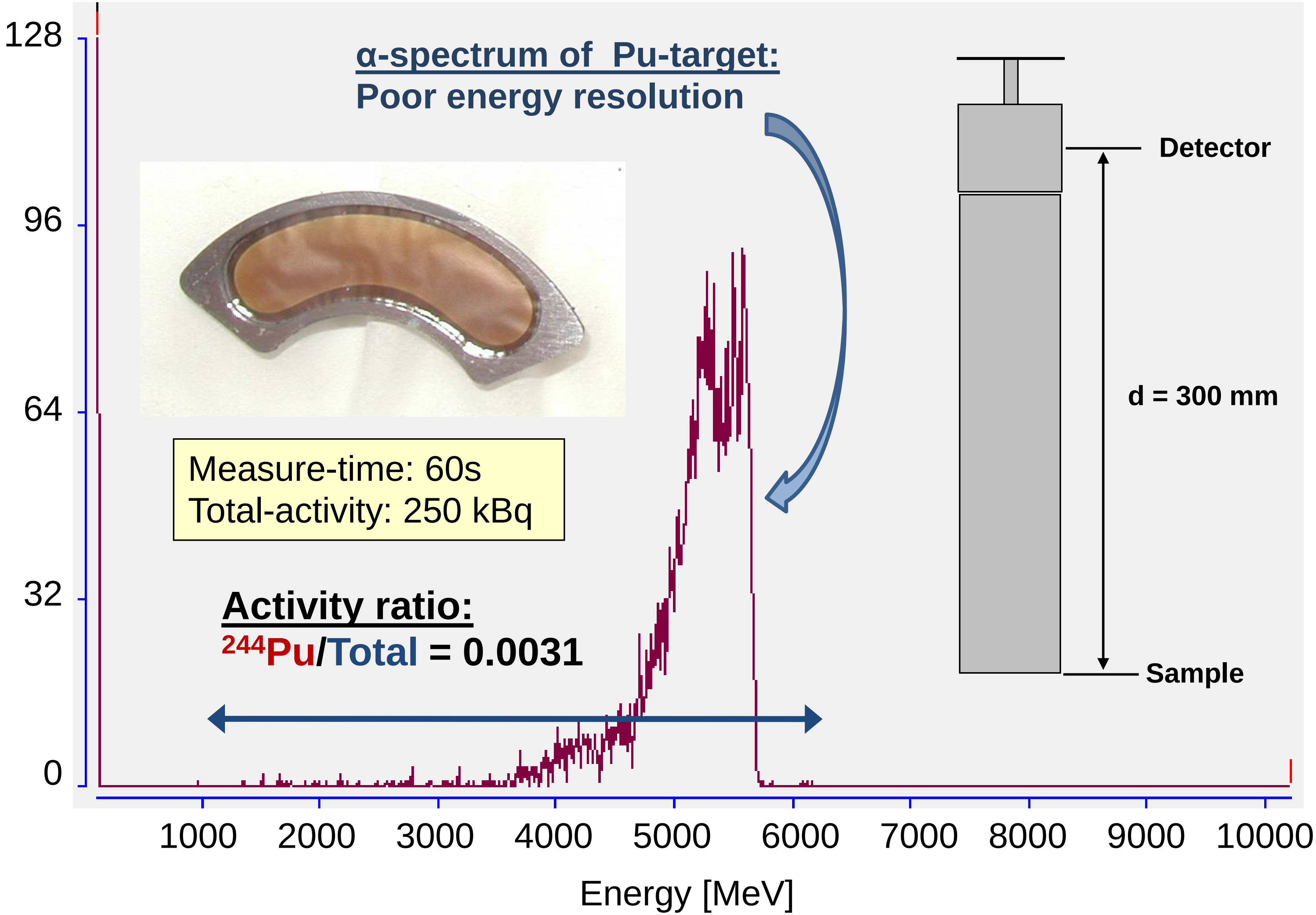
- yield 60-90%
- Test runs with ^{239}Pu
- 800 µg/cm² possible
- Cell made of Teflon
  ⇒ strong Pu-adsorption at PEEK-surface!
- Yield determination by α-spectroscopy and NAA

Deposition of Cm:

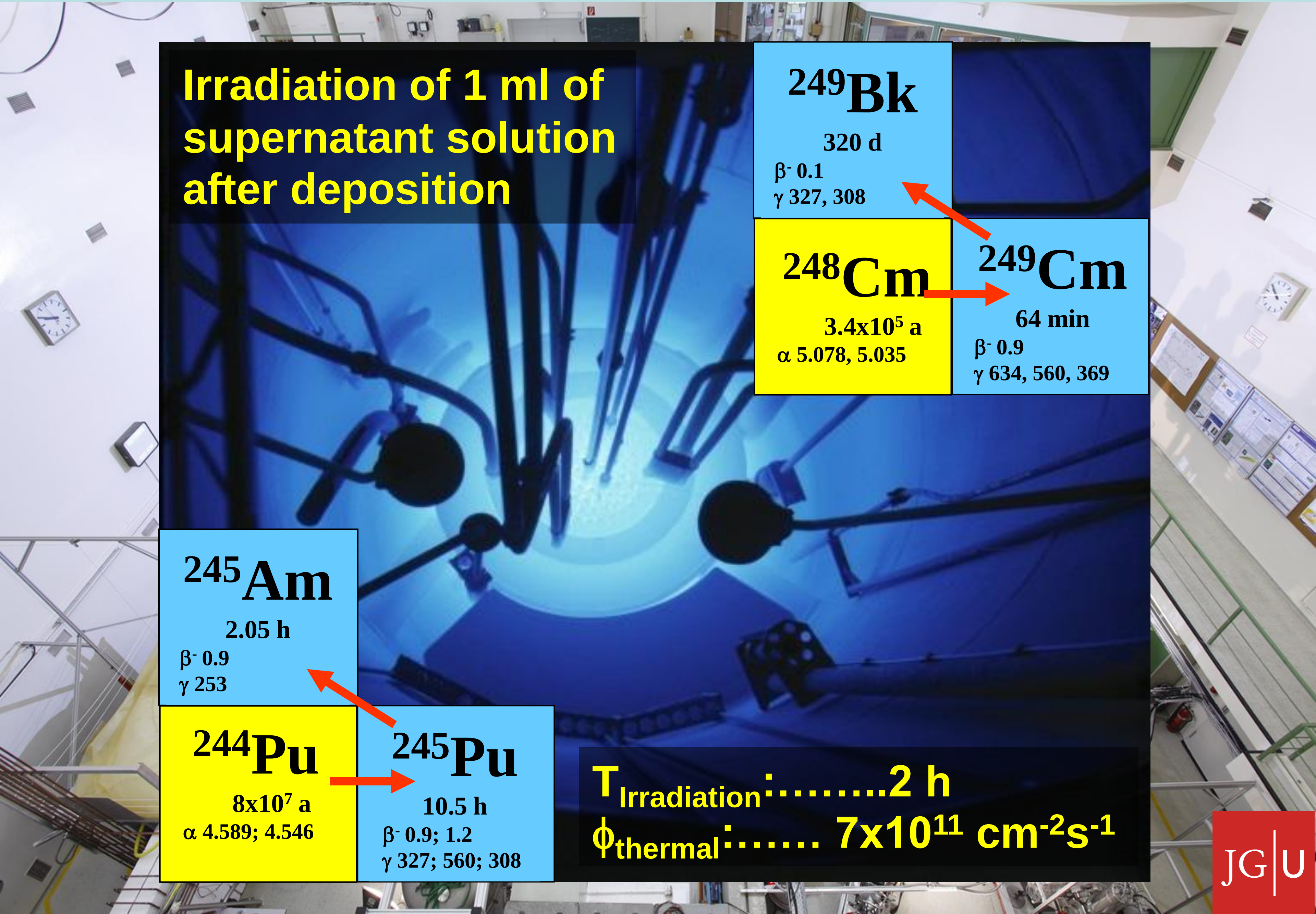
- Yield > 90%
- Cell made of PEEK
- Yield determination by α-spectroscopy and NAA
- Two production runs in 2010 and 2011

Cm	Thickness [µg/cm²]
^{248}Cm	454
^{248}Cm	465
^{248}Cm	458
^{248}Cm	457
^{248}Cm	462
^{248}Cm	463
^{248}Cm	463
^{248}Cm	460
^{248}Cm	371
^{248}Cm	373
^{248}Cm	376
^{248}Cm	376
^{248}Cm	375
^{248}Cm	373
^{248}Cm	375
^{248}Cm	371

Direct yield measurement: α-particle spectroscopy



Indirect yield measurement: NAA @ TRIGA Mainz



Irradiation of 1 ml of supernatant solution after deposition

^{249}Bk	320 d	β^- 0.1; γ 327, 308
^{248}Cm	3.4×10^5 a	α 5.078, 5.035
^{249}Cm	64 min	β^- 0.9; γ 634, 560, 369
^{245}Am	2.05 h	β^- 0.9; γ 253
^{244}Pu	8×10^7 a	α 4.589; 4.546
^{245}Pu	10.5 h	β^- 0.9; 1.2; γ 327; 560; 308

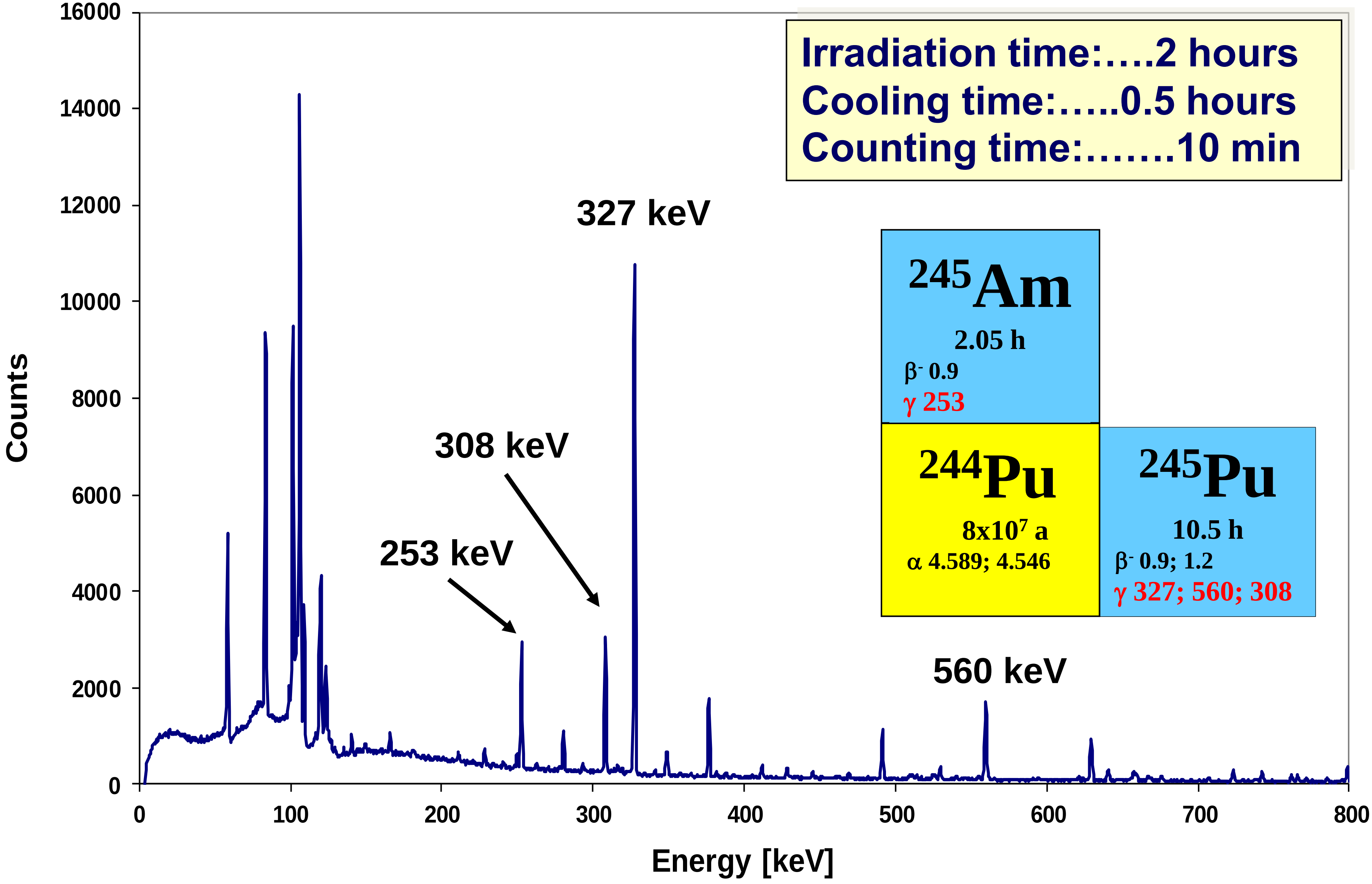
^{248}Cm → ^{249}Cm → ^{249}Bk

^{244}Pu → ^{245}Pu → ^{245}Am

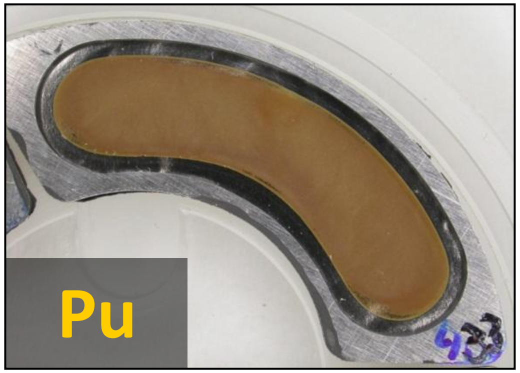
$T_{Irradiation}$:……..2 h

$\phi_{thermal}$:…… 7×10^{11} cm^{-2}s^{-1}

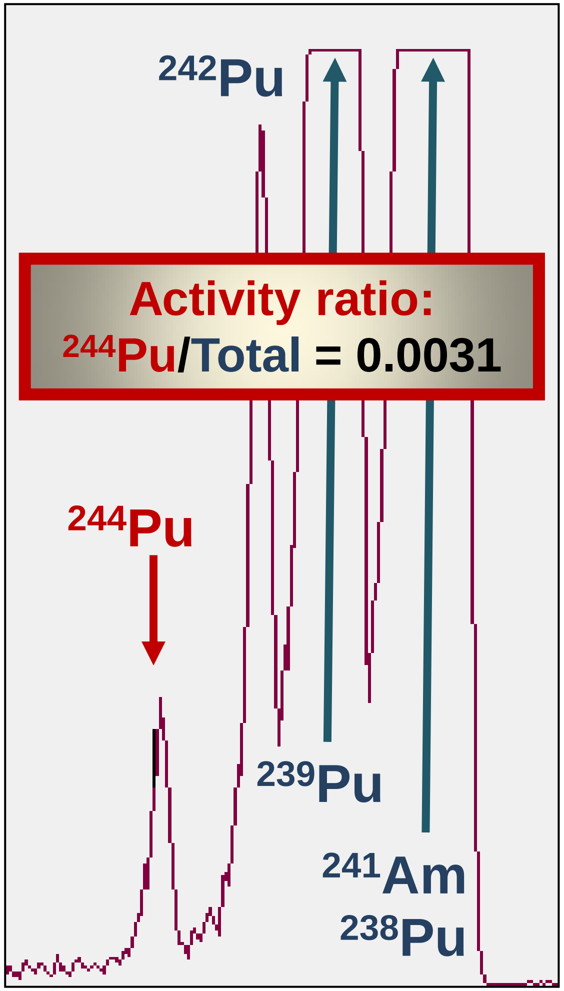
γ-spectrum of irradiated ^{244}Pu solution (1 ml / 10 μg)



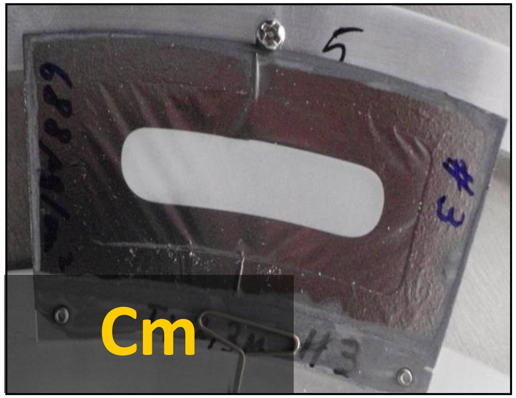
Yield determination: α-spectroscopy and NAA



α	NAA
401 (±40)	----
472 (±47)	----
490 (±49)	----
390 (±39)	----
673 (±67)	----
565 (±57)	673 (±61)
650 (±65)	724 (±65)
944 (±94)	790 (±71)
778 (±79)	785 (±70)



⇒ Limited precision in yield determination by α-spectrocopy

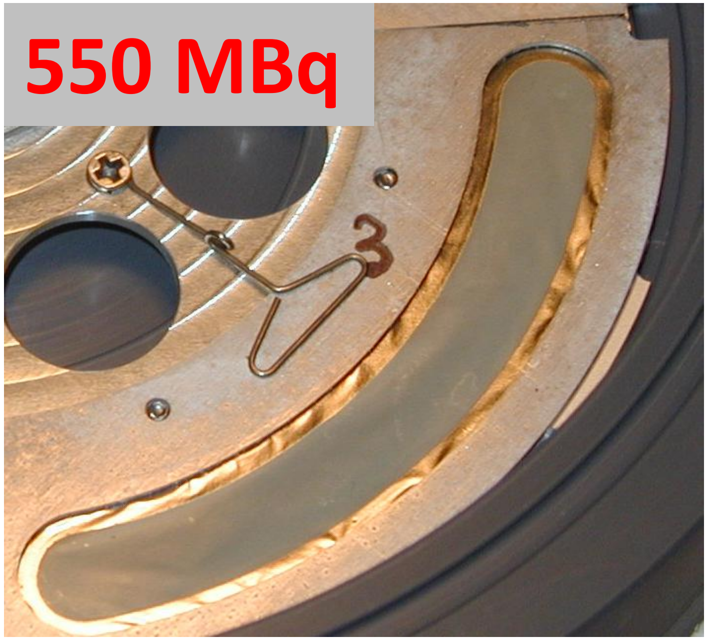


α	NAA
454 (±23)	429 (±39)
457 (±23)	454 (±41)
458 (±23)	458 (±41)
460 (±23)	458 (±41)
462 (±23)	455 (±41)
463 (±23)	458 (±41)
463 (±23)	457 (±41)
465 (±23)	435 (±39)

Deposition of Am and Cf by Molecular Plating

Deposition conditions for Am:

Solvent…………….Isobutanol
Plating time……….6 h
Voltage…………….150 – 200 V
Current density.….0.2-0.3 mA/cm^2
Stirring at………….1200 U/min
Temperature….…..15 C



Deposition conditions for Cf:

Solvent……………….Isobutanol
Plating time……….…4-5 h
Voltage………………..300 – 600 V
Current density….......0.2 mA/cm^2
Ultrasonic stirring......30 W
Temperature……....….15 C

Deposition of Am and Cf by Molecular Plating

Deposition of Am:

- Yield > 90%
- > 800 µg/cm² possible
- Cell made of PEEK
- Yield determination by γ-spectroscopy

Am	Thickness [µg/cm²]
^{243}Am	75
^{243}Am	758
^{243}Am	524
[illegible]	[illegible]
^{243}Am	634
^{243}Am	714

Deposition of Cf:

- Yield > 90%
- Cell made of PEEK
- Yield determination by γ-spectroscopy

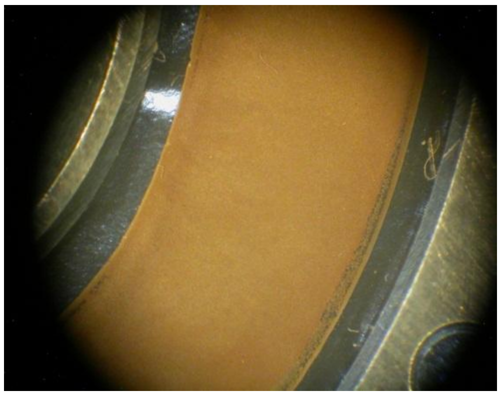
Cf	Thickness [µg/cm²]
^{249}Cf	70
^{249}Cf	484
^{249}Cf	537
[illegible]	[illegible]
^{249}Cf	454

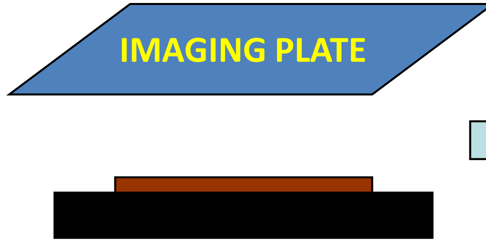
Prior to production of thick targets preparation of thin Targets:

- deposition test
- reference sample for yield determination by γ-spectroscopy

Target characterization: Radiographic Imaging (RI)

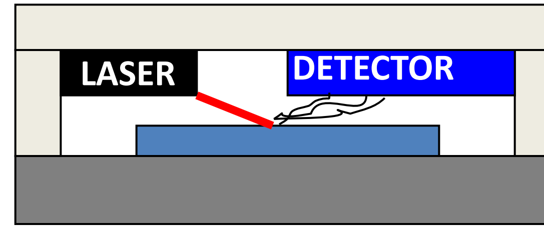
1 Production of Pu-244 target



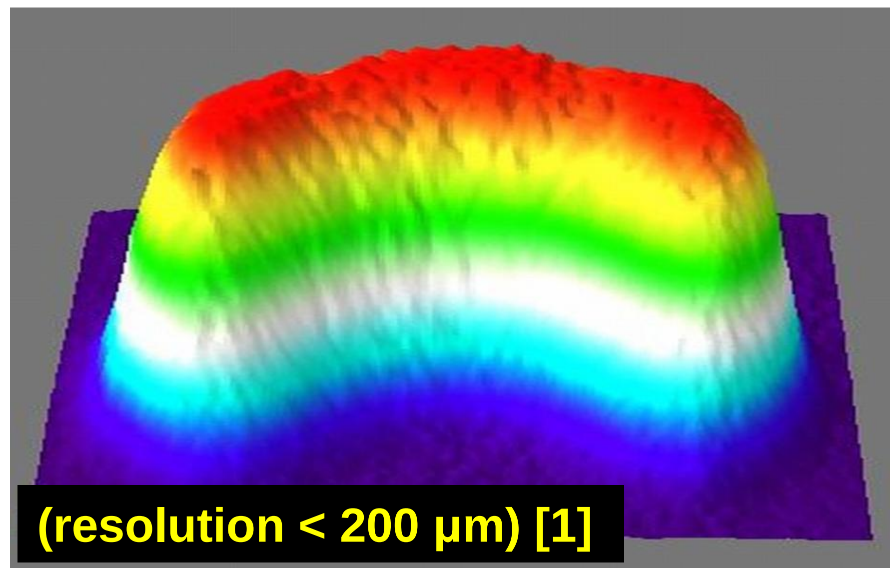


2 Imaging plate exposure to active target

3 Imaging plate reading process



Surface homogeneity



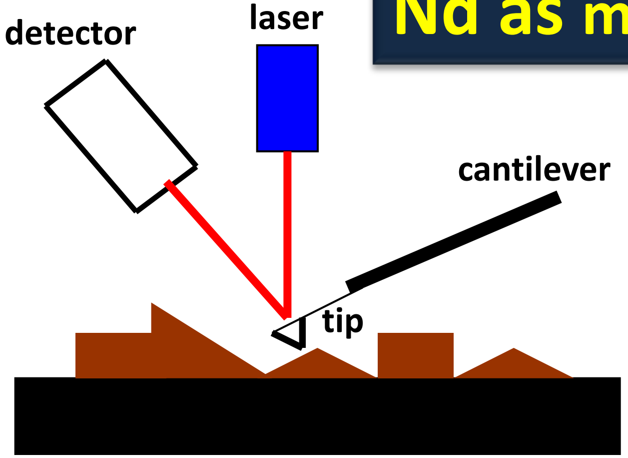
[1] D. Liebe, et al., Nucl. Instr. and Meth. A 590 (2008) 145

Target characterization: Atomic Force Microscopy (AFM)

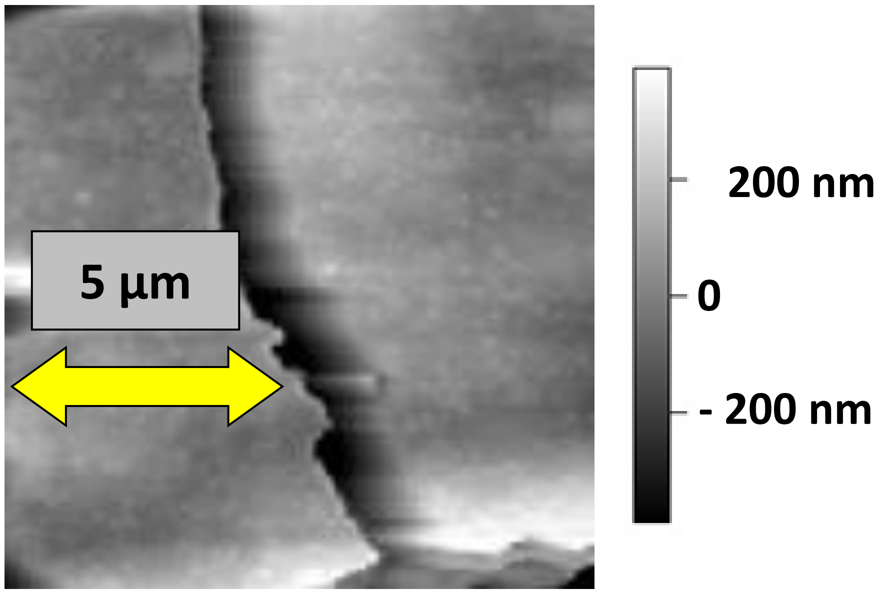
Nd as model element

1 Production of target

2 AFM target analysis



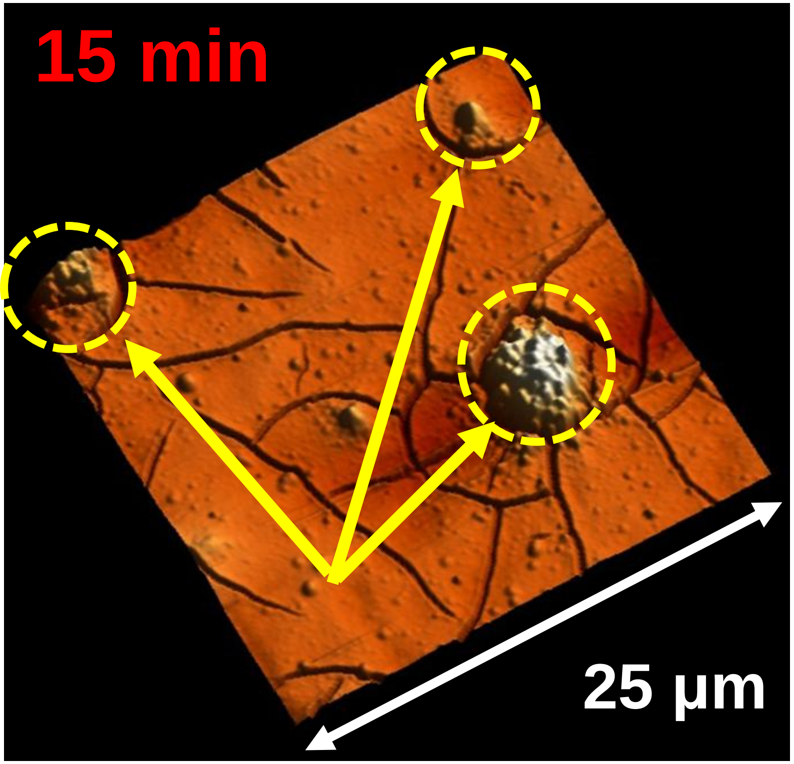
Surface roughness

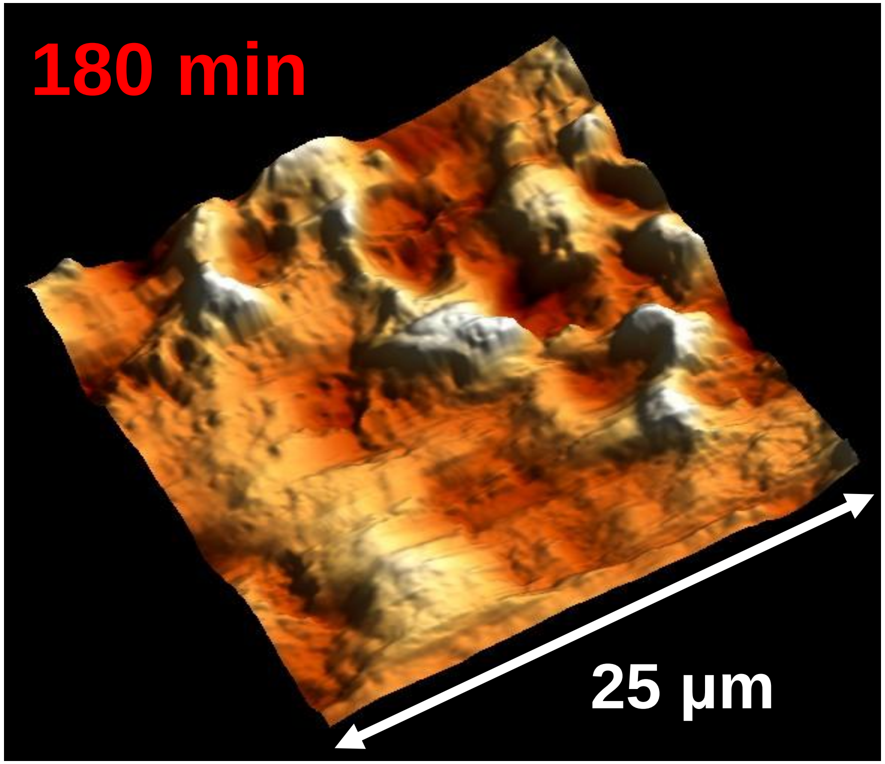


3 Typical obtained image

Studies on layer formation: Atomic Force Microscopy (AFM)

Nd as model element

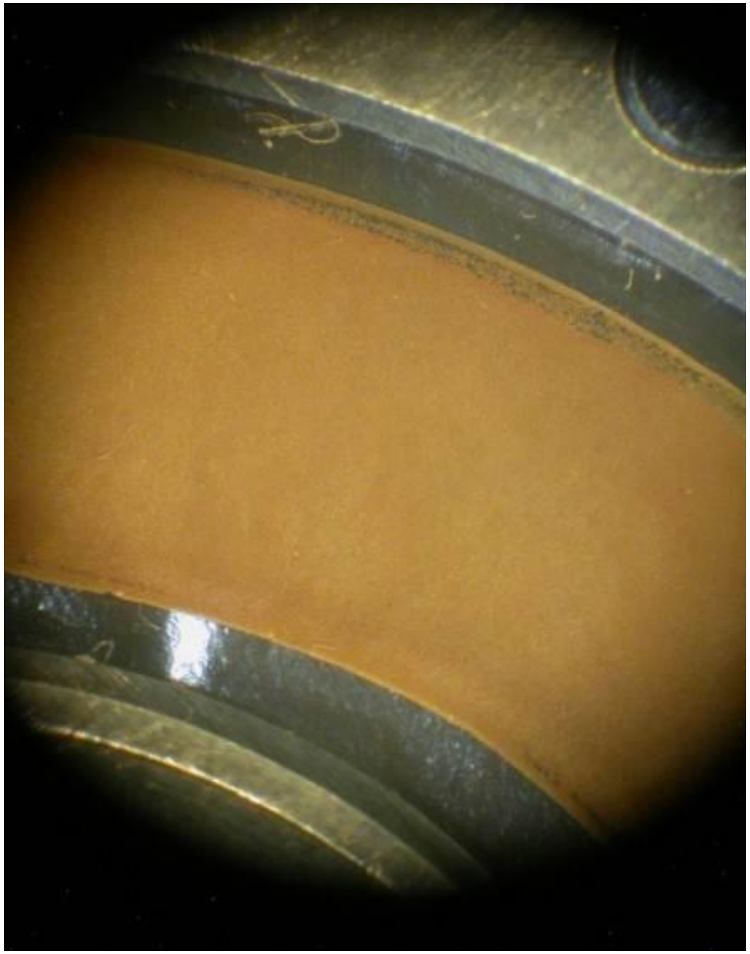




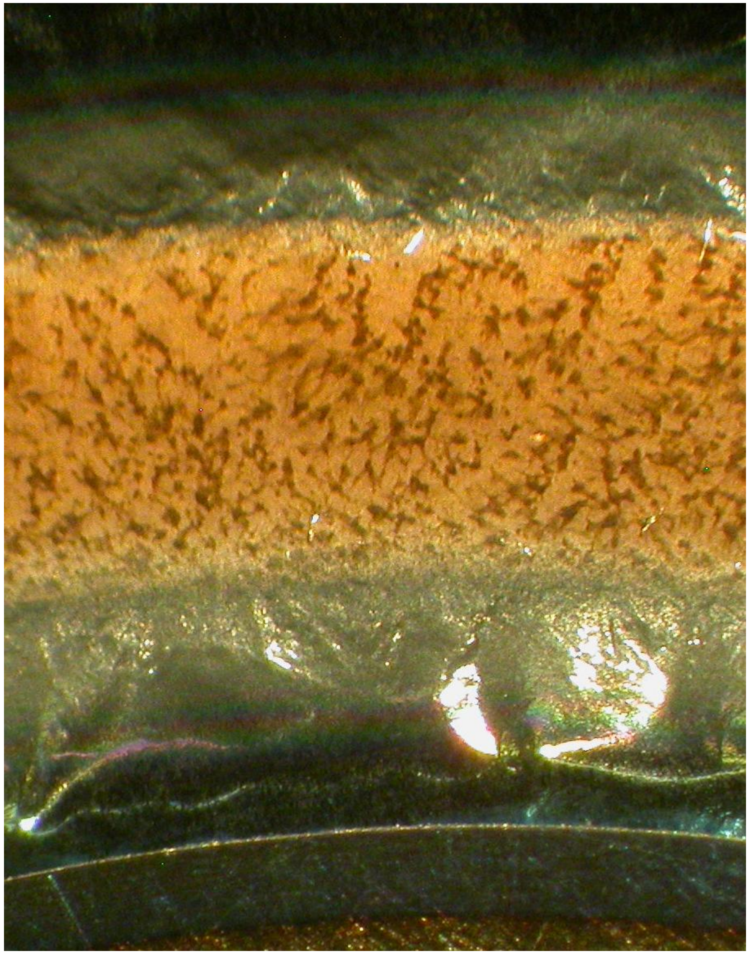
Deposition starts at spots (defects) on Ti-foil surface. Layer growth and final layer stability strongly depends on original backing surface roughness. Basic studies on layer formation and layer growth using AFM in solution. [A. Vascon et al., Nucl. Instr. and Meth. A, in press]

Irradiation of Pu-244 targets with Ca-48 beam: Layer morphology change

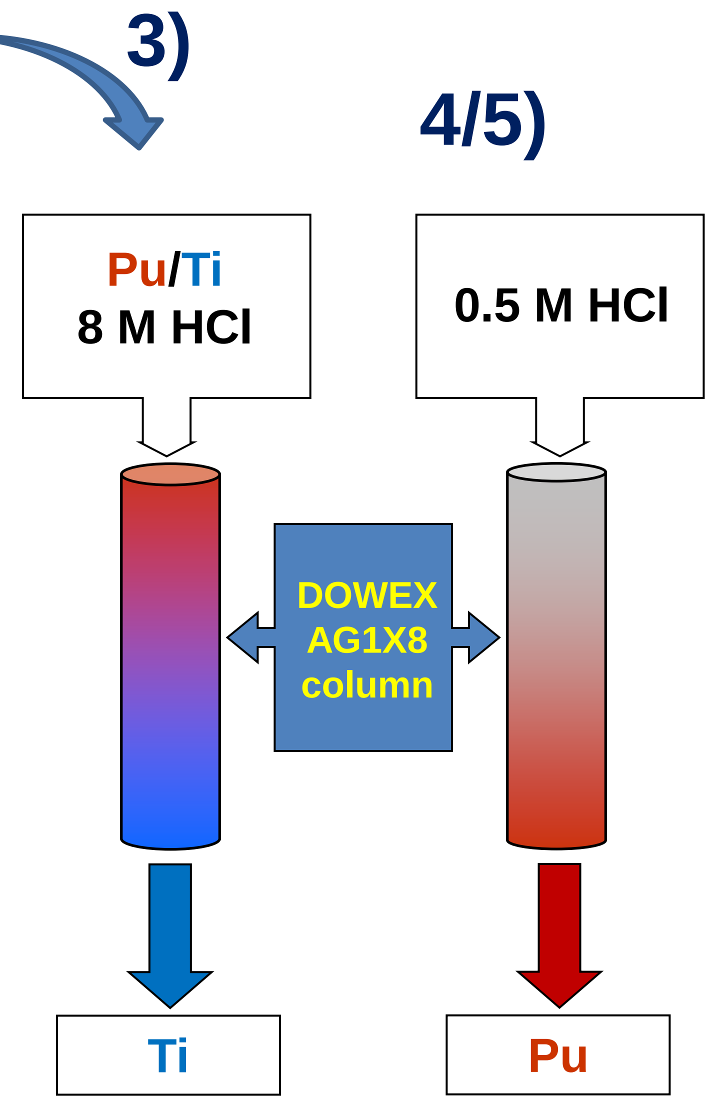
Fresh Pu-244 target layer



Pu-244 target irradiated with Ca-48. Beam dose > 5×10^{18}



Recovery of Pu-244 from used targets



1) Cut-out Ti-foil from Al-frame
2) Dissolve in hot, conc. HCl in a Teflon beaker
3) Evaporate to dryness and re-dissolve in 8m HCl
4) Removal if Ti with anion-exchanger DOWEX AG1X8.
5) Pu is eluted with 0.5 m HCl.
   Pu-recovery 89% (by NAA)

Alternative target production techniques I

- **Polymer-assisted deposition (PAD), LBNL work:**

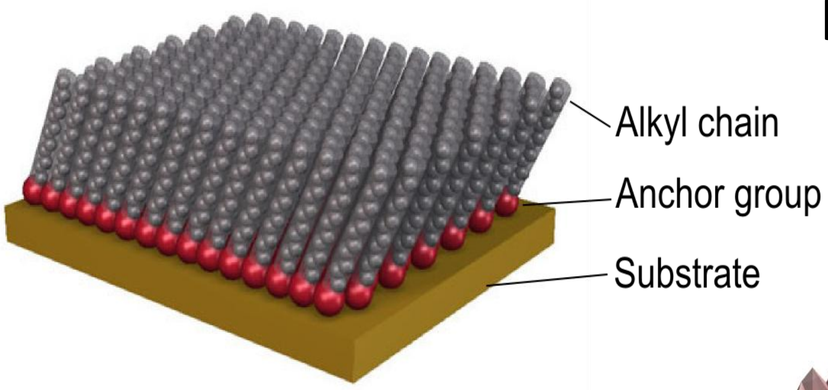
  Metal-oxide mixed with polymer solution. Spin-coating of silicon substrate with metal-organic film. Target thickness up to 600 µg/cm^2 possible. No irradiation tests with actinide elements so far

  [M. Garcia *et al*., Nucl. Instrum. Methods A 613 (2010) 396]

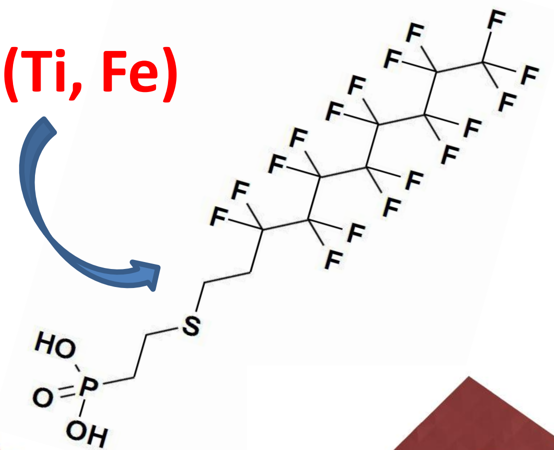
Alternative target production techniques II

- ## Superhydrophobic surfaces:

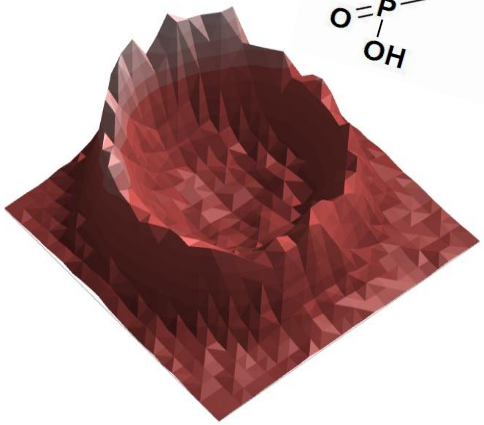
Modification of a substrate with self-assembled momolayer of alkyl chains. Homogenious deposition of metal-oxide/nitrate from aqueous solution by simple evaporation of single drops. No irradiation tests with actinide elements so far. [D. Renisch, Diploma Thesis, U. Mainz (2010)]



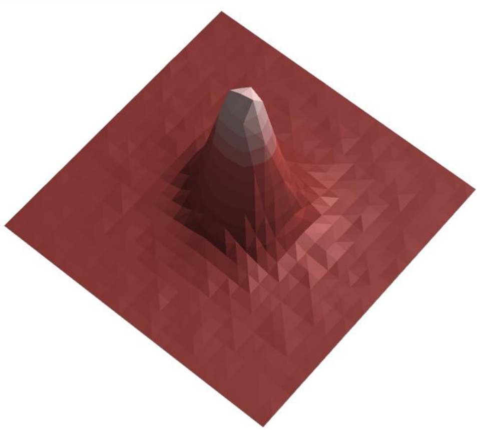
HDFP (Ti, Fe)



Evaporation of a single drop of Am-241(nitrate) solution. Activity distribution by RI



Untreated Ti-surface



Modified Ti-surface

Summary

- Rotating target wheels for TASCA and SHIP
- Molecular Plating of Pu, Cm, Am and Cf
- Target characterization methods include
  - α-, γ-spectroscopy, NAA
  - Radiographic Imaging (RI)
  - Atomic Force Microscopy (AFM)
- Recovery of used target material
- New developments in target production

Thanks to:

Lawrence Livermore National Laboratory providing Cm-248

Oak Ridge National Laboratory providing Am-243

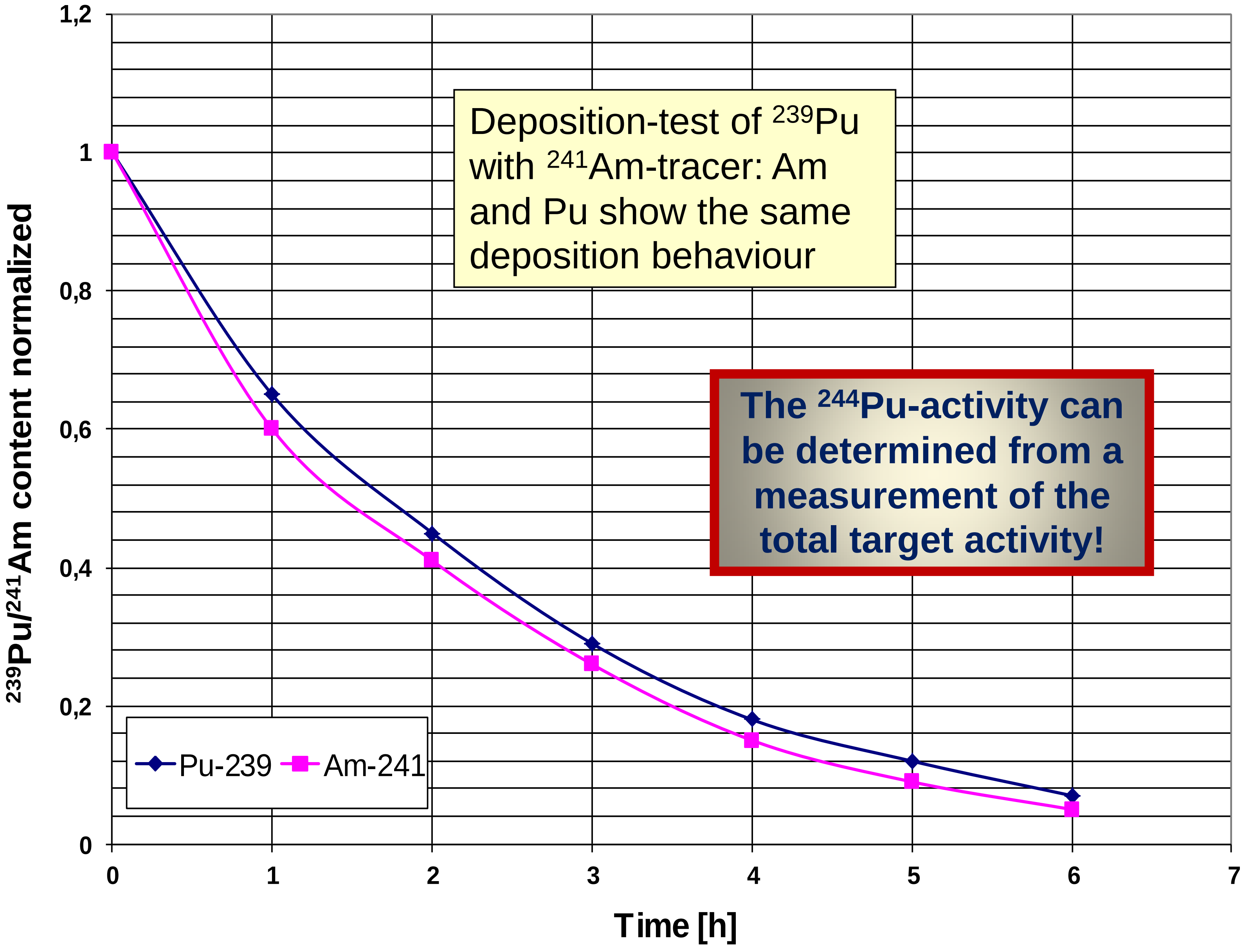
Lawrence Berkeley National Laboratory providing Cf-249

A. Kühnle, Institute of Physical Chemistry for AFM-measurements

BMBF for financial support

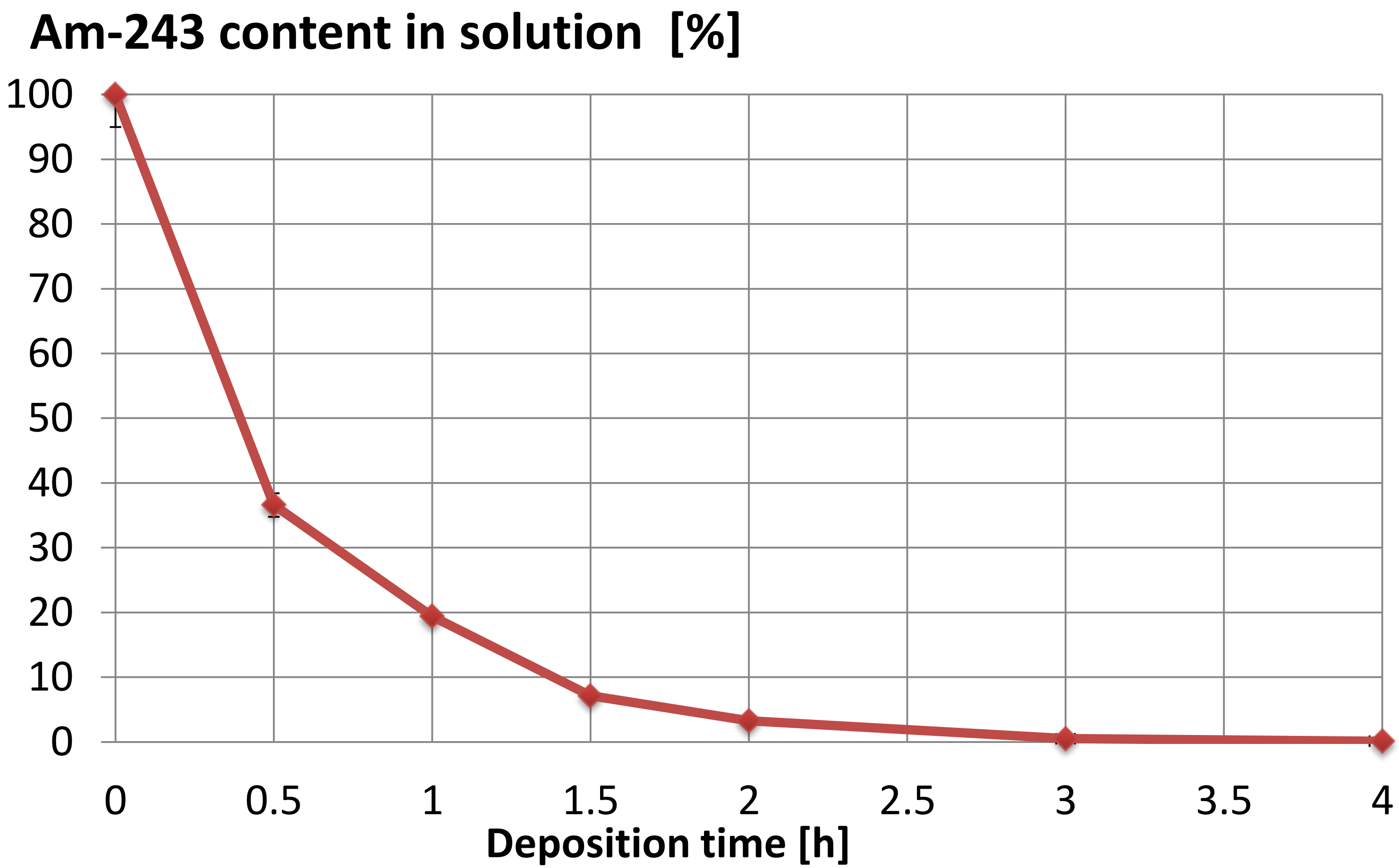
….and you for your attention

Simultaneous deposition of ^{239}Pu and ^{241}Am



Deposition kinetics of Am

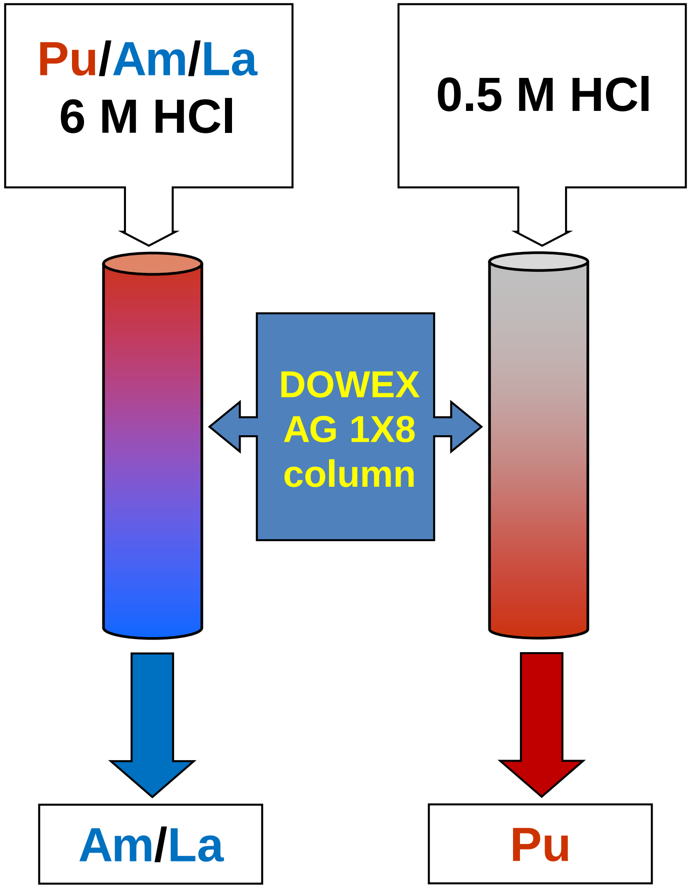
Prior to deposition and in subsequent 30-min steps 10 µl aliquot of supernatant solution evaporated to dryness to prepare sample for α-particle spectroscopy



Chemical separation of Am and Pu

^{241}Am 50.8 h	^{241}Am 432 a	
^{239}Pu 2.4×10^4 a	^{240}Pu 6563 a	^{241}Pu 14.4 a

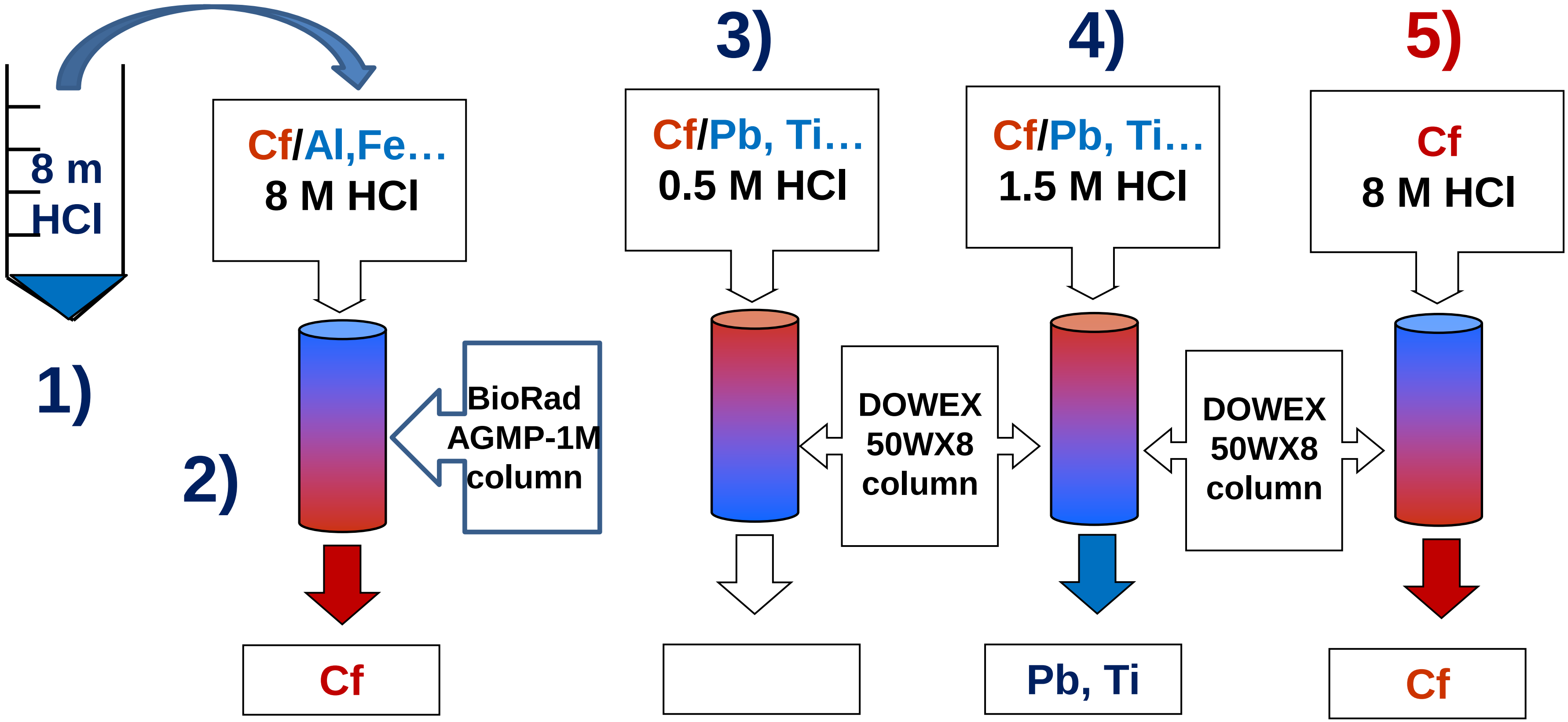
^{244}Pu stock solution contains ^{241}Am from decay of traces of ^{241}Pu.



Pu-Am-separation (if needed):

1. Co-precipitation of Pu (and Am) with $La(OH)_3$
2. Dissolvation of precipitate in 6 M HCl
3. Ion-exchange chromatography using DOWEX AG 1X8 anion-exchanger. In strong HCl-solution Pu forms anionic species that are retained.
   $\Rightarrow$ Pu is then eluted with 0.5 M HCl

Purification of Cf-249 prior to use



1) Dissolve in 8 m HCl
2) Purification step with anion-exchanger BioRad AG MP-1M
3) Evaporate to dryness and re-dissolve in 0.5 m HCl
4) Removal of Pb, Ti… with kation-exchanger DOWEX 50W-x88.
5) Cf is eluted with 8 m HCl. Almost 100% recovery

Purification of Cf-249 prior to use

^{244}Pu stock solution