Actinide and Lanthanide Target Developments using a Drop-on-Demand Printing System

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… the mechanical and electrical workshops at the JGU

… the radiation protection team

… all students and collaborators involved in the target printing topic in the past years

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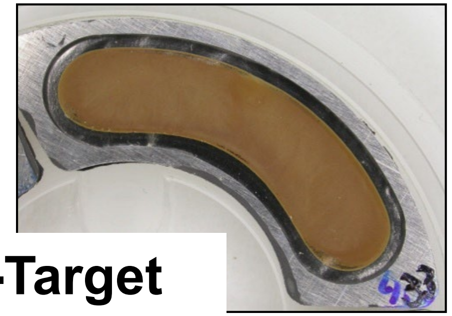
REQUESTS FOR RADIONUCLIDE TARGETS

Accelerator-based experiments (e.g. SHE)

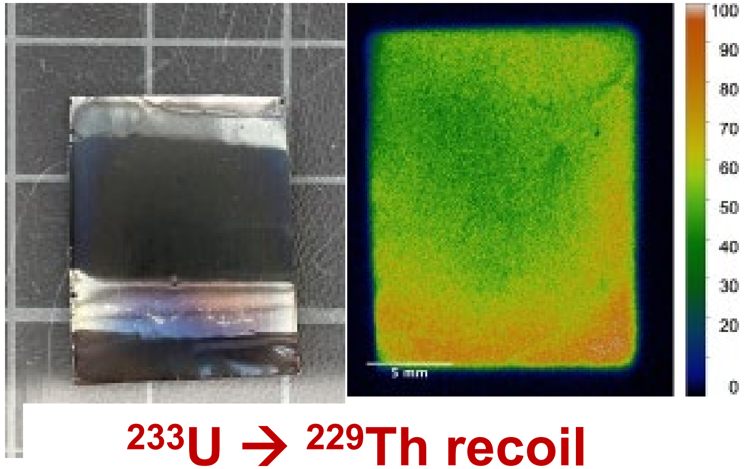
- thickness of about 1 mg/cm^2
- typical backing: 2 µm Ti
- mechanical/thermal resistance
- high homogeneity
- banana-shaped geometry for mounting on target wheels
- reliable technique, with high yields for rare material
- possibility to gain back unused material



Ti/^{244}Pu-Target

Recoil sources

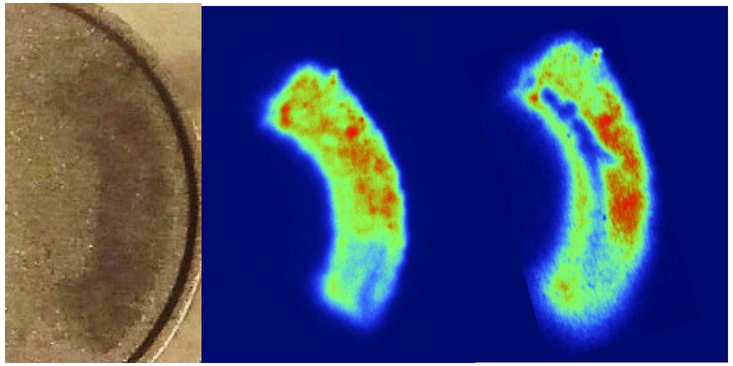
- high recoil rate
  - as much activity as possible
  - layer as thin as possible
- layer can be thicker, if energy of recoils does not matter
- for maximum recoil energy, the ideal source is a monolayer



^{233}U → ^{229}Th recoil source on Ti

Laser excitation/ablation

- broad variation of special requirements depending on the specific project
  - simple drop-and-dry targets
  - specific sample thickness in a few nm range + specific chemical composition



^{249}Cf target for a laser ablation source

TARGET REQUIREMENTS

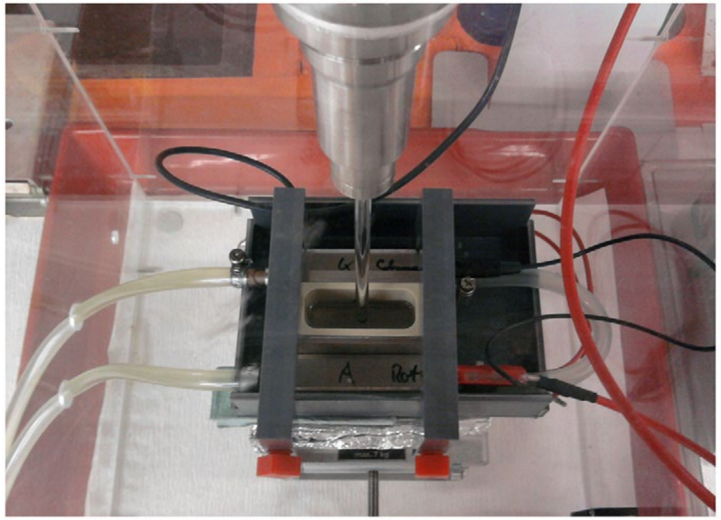
Broad variety of requirements

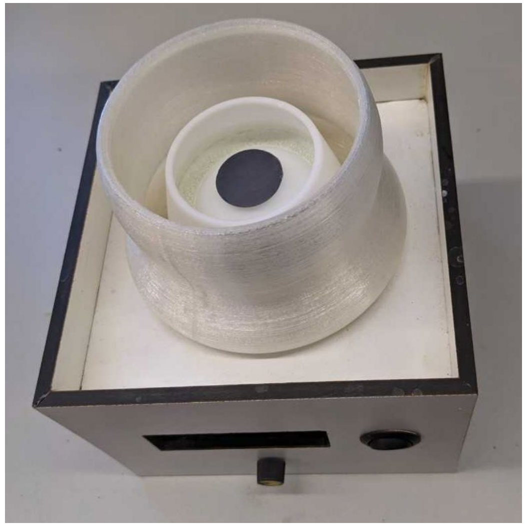
- Thickness
- Homogeneity
- Substrate
- Chemical form

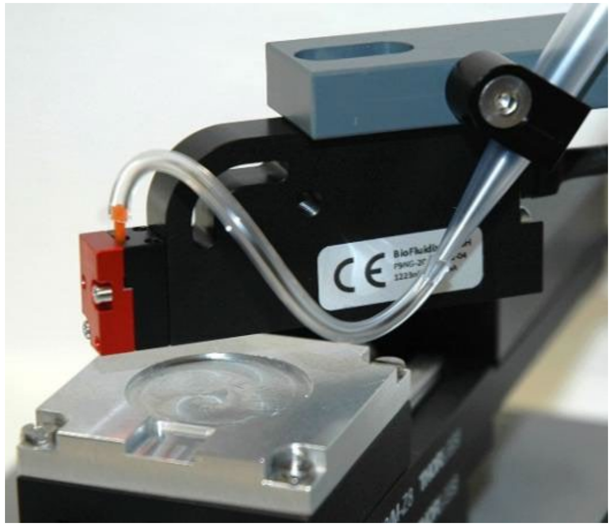
→

Necessity to establish a variety of different production methods

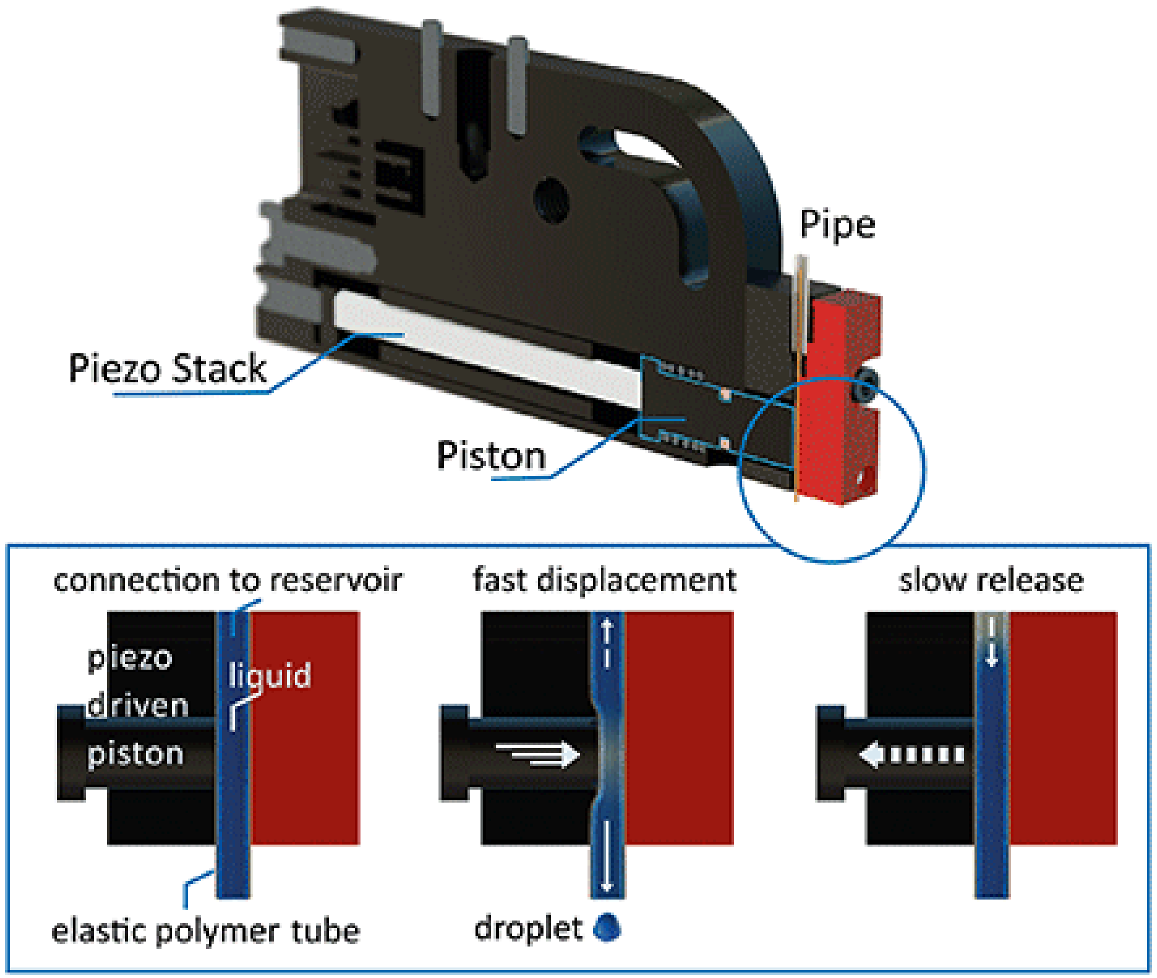
- Molecular plating
- Self-adsorption
- Spin coating
- Drop-on-Demand printing







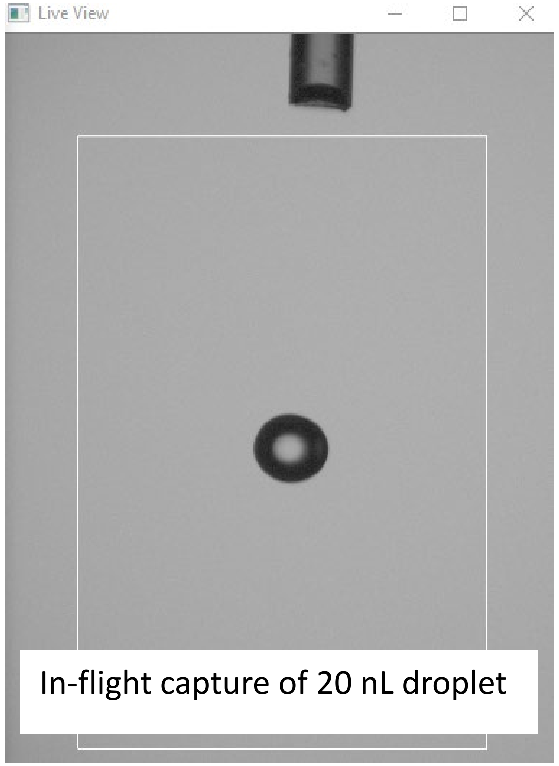
DROP-ON-DEMAND (DOD) PRINTING



Working principle of PipeJet® Nanodispenser
BioFluidix GmbH, http://www.biofluidix.com

Important settings:

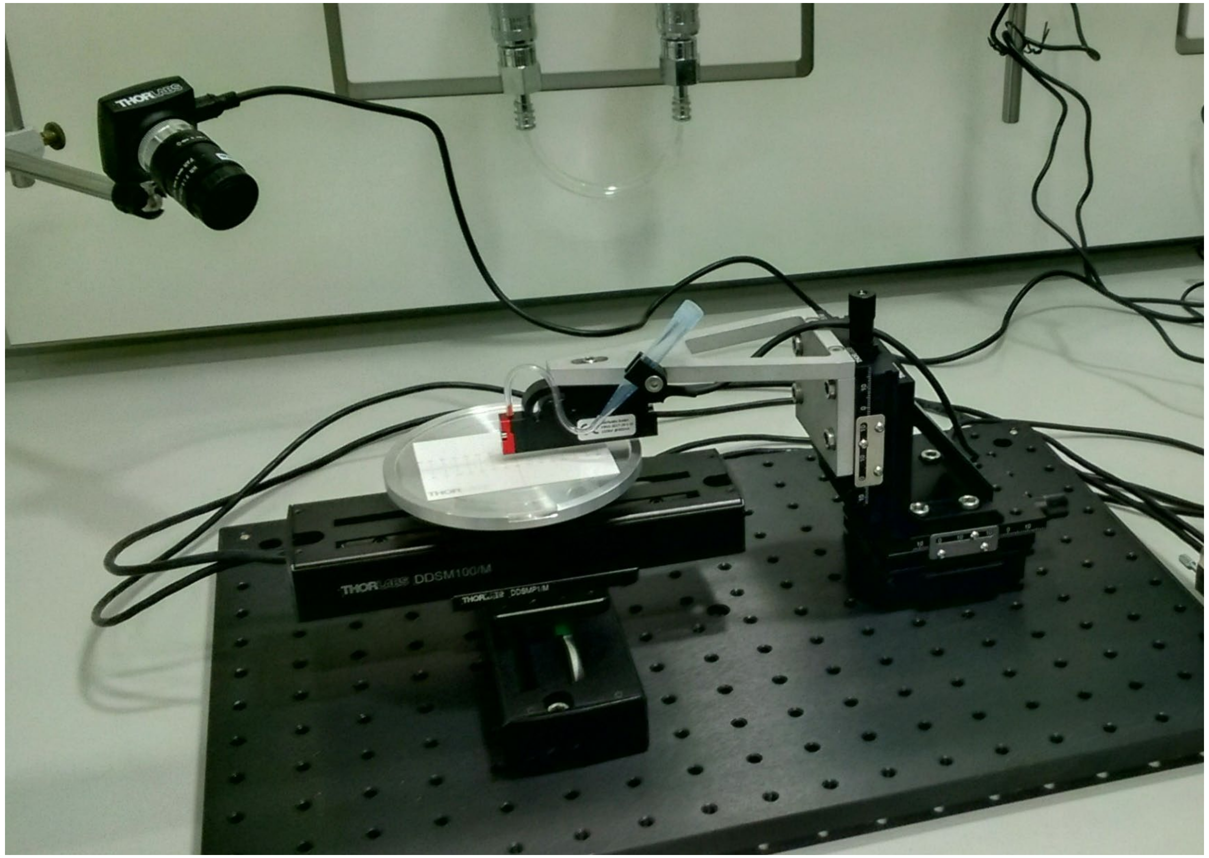
- **stroke range [µm]**
- **stroke velocity [µm/ms]**

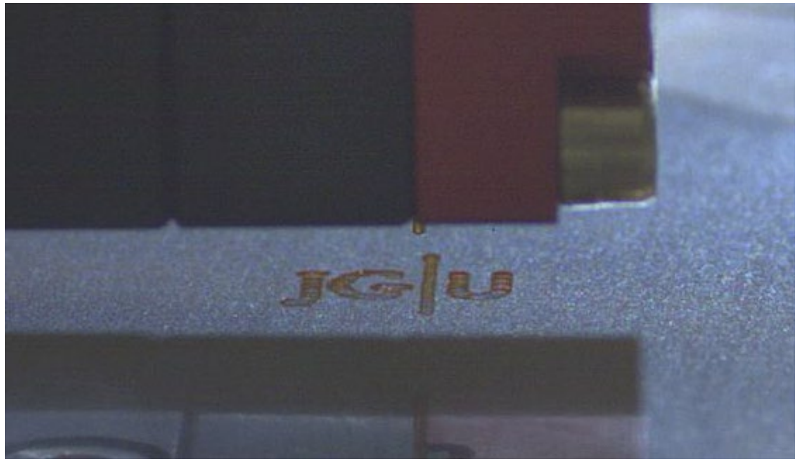


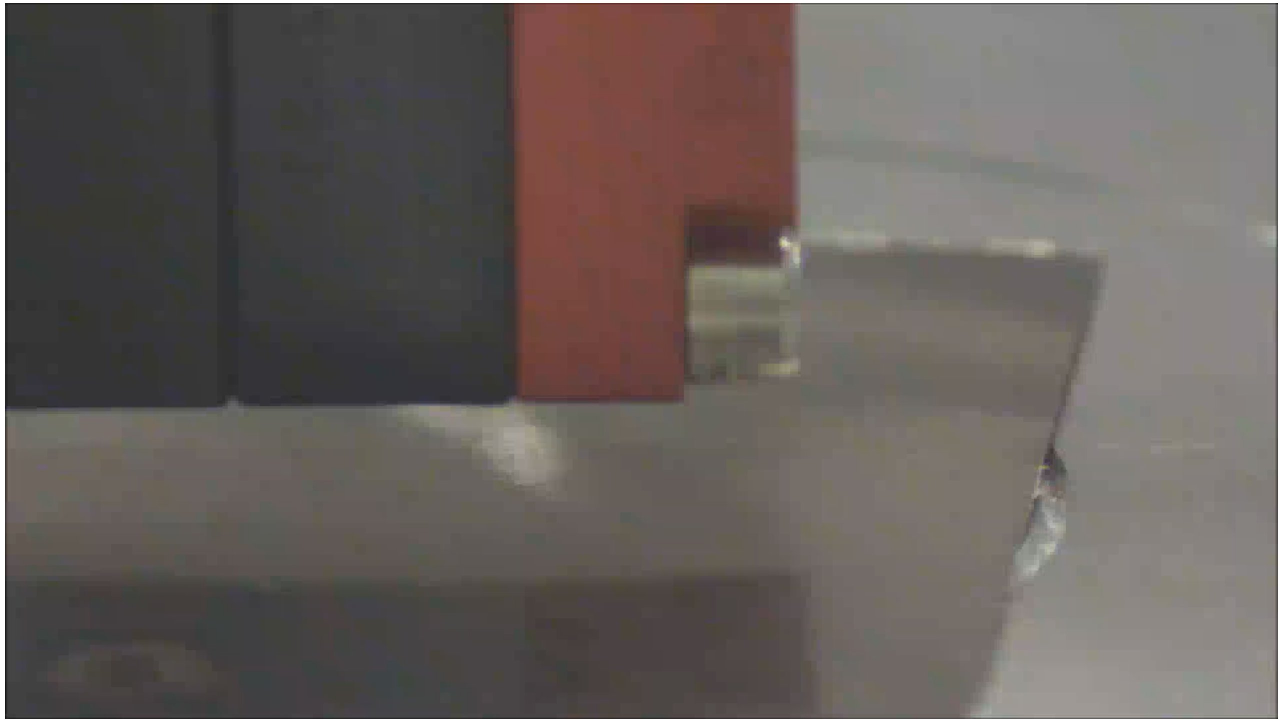
In-flight capture of 20 nL droplet

→ droplet sizes in the range 5 – 60 nL

DROP-ON-DEMAND PRINTING



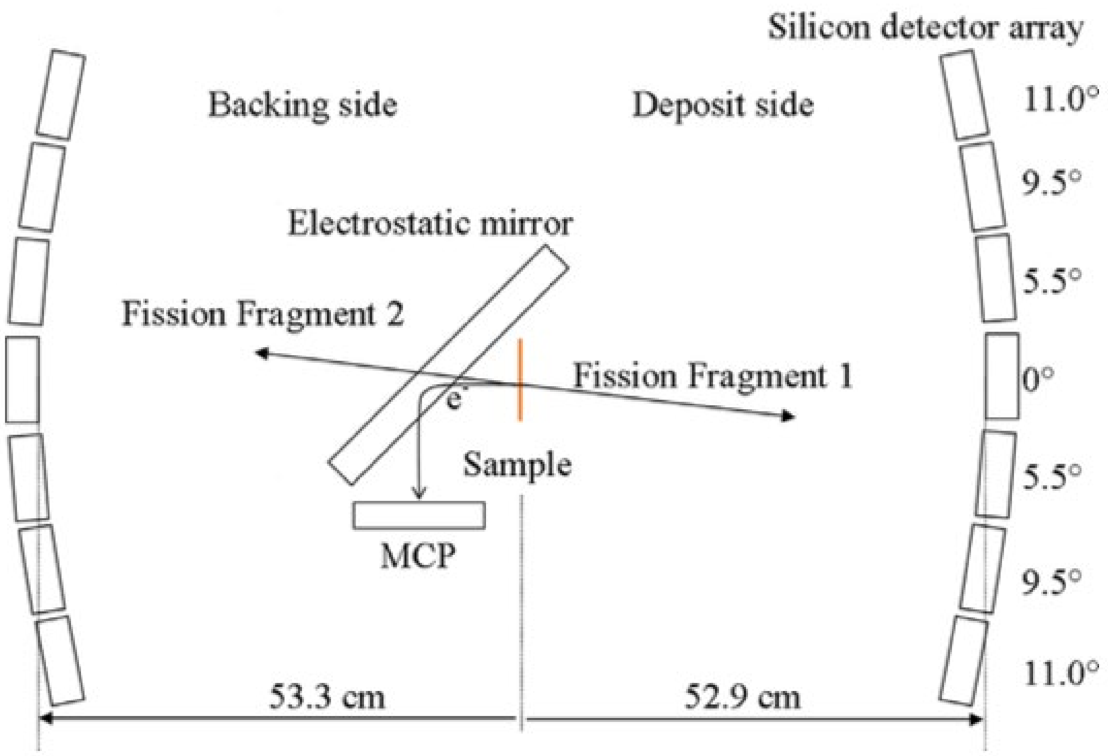




Combination of PipeJet dispenser with x-y-translation stages

R. Haas et al., NIM A 874 (2017) 43

CF-252 / CM-248 SOURCES FOR JRC GEEL



scheme of Verdi spectrometer

→ Determination of fission fragment masses and kinetic energy distributions

Source requirements:

- high energy resolution in 4π
- very thin backing: Au coated polyimide foil (24 µg/cm²)
- Cf/Cm layer with 1000 – 10000 fission/s, as thin as possible

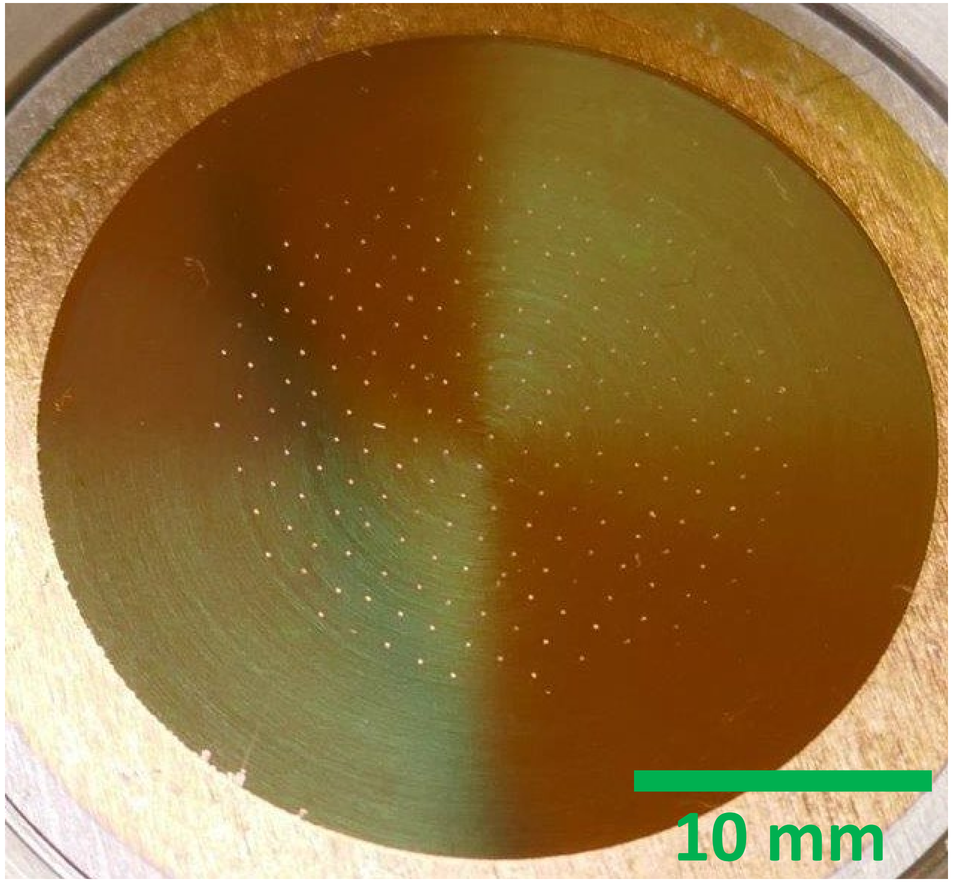
M.O. Frégeau et al., NIM A 817 (2016) 35

CF-252 / CM-248 SOURCES FOR JRC GEEL

Cf 252
2.647 a
α 6.118, 6.076...
γ (43, 100
155...), e⁻, sf
σ 20.3, σ_f 32

First attempt 2018

- printing from aqueous solution
- 221 x 20 nL drops, separated by 1 mm
- activity:
  - 330 kBq (alpha)
  - 10 kBq (sf)



Relative high hydrophobicity of substrate resulted in „thick" deposition
→ too high energy loss of fission fragments within the source

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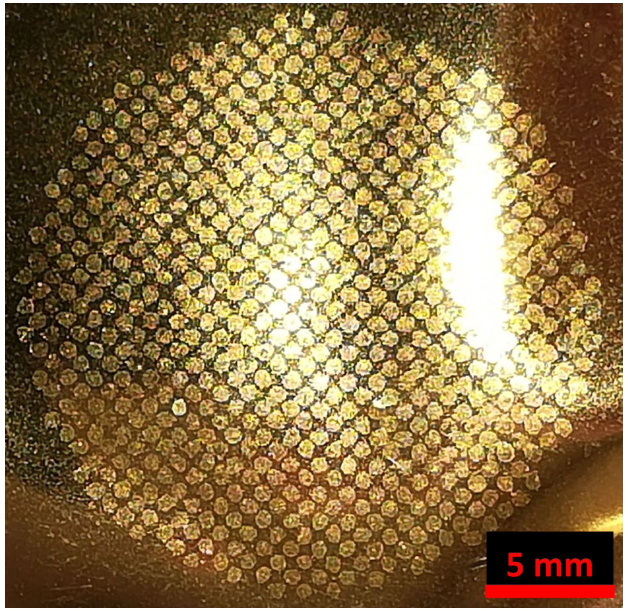
CF-252 / CM-248 SOURCES FOR JRC GEEL

New attempt 2021/2022

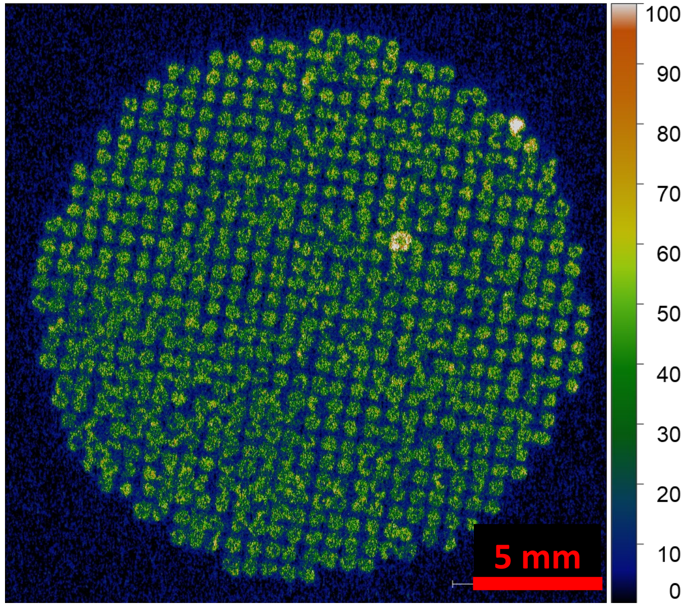
Cm 248
$3.48 \cdot 10^5$ a
α 5.078, 5.035...
γ, e⁻, g, sf
σ 2.63, σ_f 0.37

→ Tests with Am-243 in alcoholic solutions to control wetting behavior

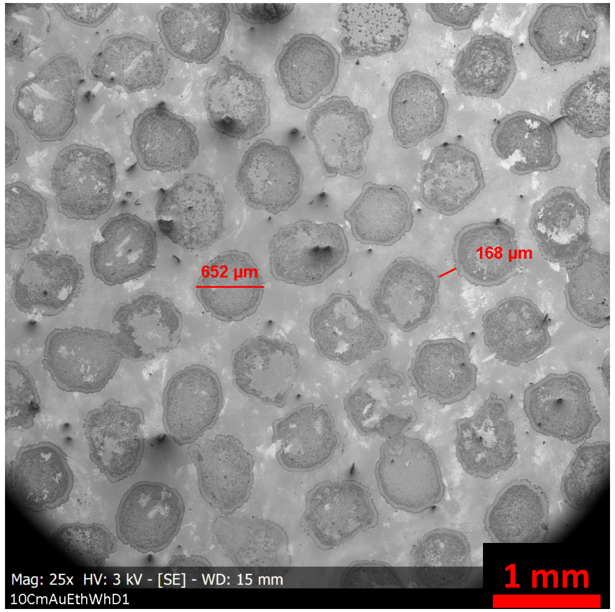
→ Improved printing pattern using ethanol/water mixture (70:30)



Microscope picture



Radiographic imaging



SEM picture

8 µg Cm-248
1270 Bq (α)
114 Bq (sf)
FWHM 19 keV
641 x 10 nL
680 µm deposit ø

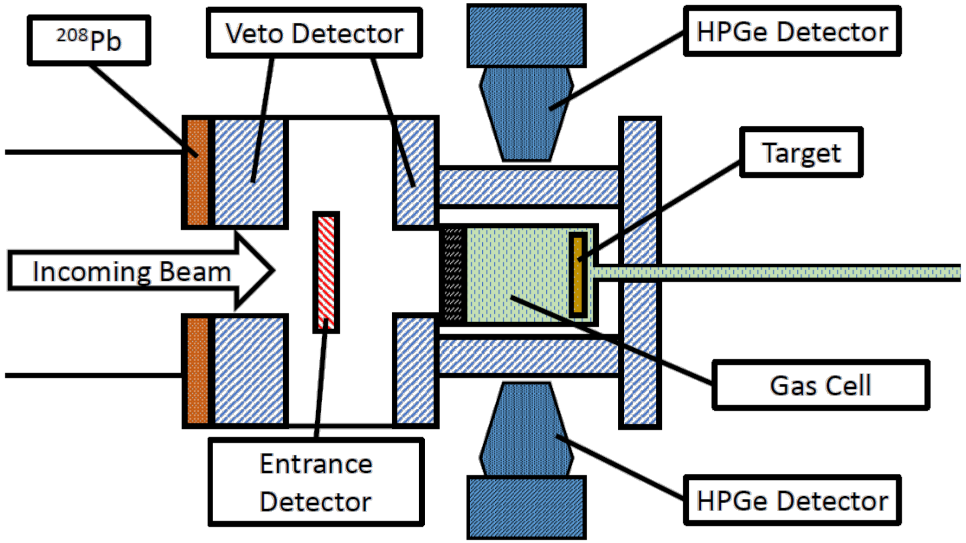
B.Sc. thesis of J. Lukacova (2021)
Research internship of Ch. Sirleaf (2022)

CM-248 / RA-226 TARGETS FOR MUX @ PSI

nuclear structure investigation from measurements of muonic X-rays

→ requirements:

- element of interest on the very top layer
- low Z backing → glassy carbon

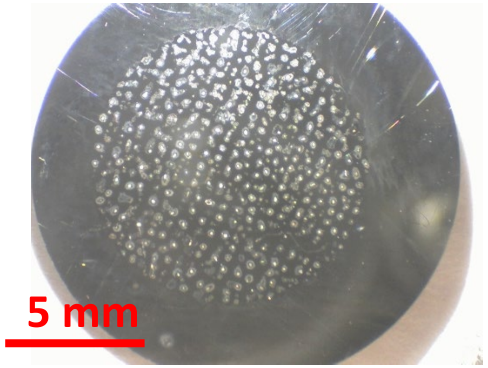


A. Knecht et al., arXiv:2004.03314v1 (2020)

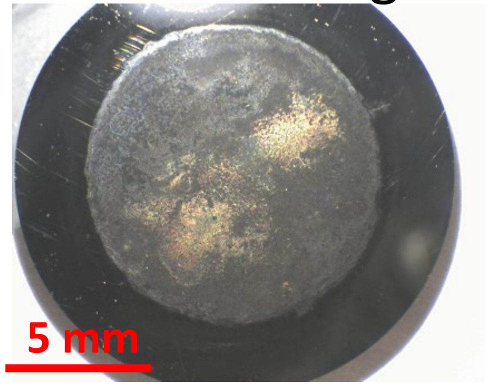
Beamtime 2018:

- Cm-248 worked with combination of MP + DoD
- did not worked for Ra-226

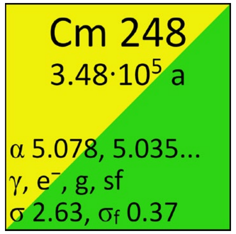
15 µg Cm-248 on glassy carbon backings



… DoD from aqueous solution



… MP + DoD

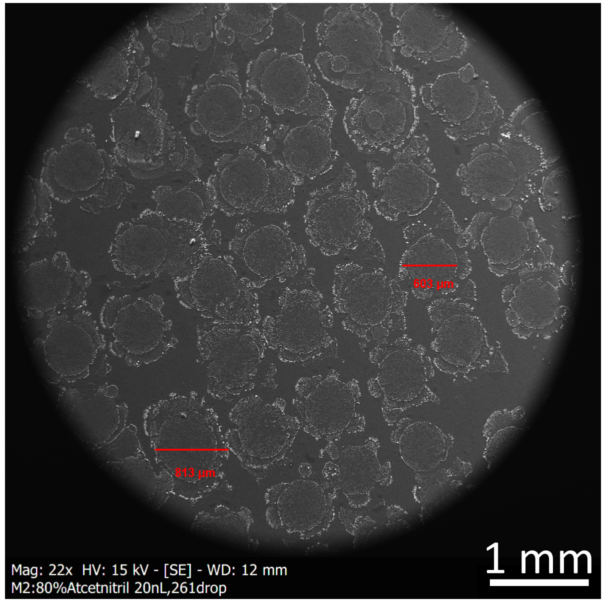


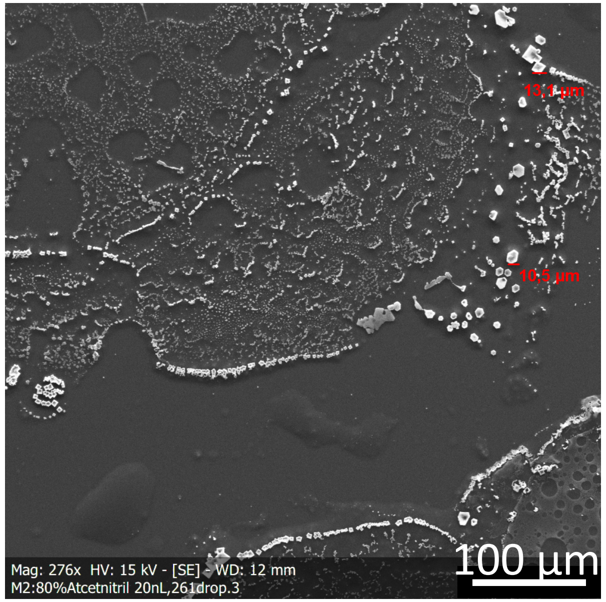
CM-248 / RA-226 TARGETS FOR MUX @ PSI

Ra 226
1600 a
α 4.7843, 4.601...
γ 186...
C14
σ 13.7, σ_f <5E-5

Ongoing challenge 2022: best route for Ra-226

- colleagues from PSI work on MP route
- we try to use DoD from organic solvents





5 µg Ba-nitrate on Sigradur

→ tests with EtOH/H_2O and acetonitrile/H_2O mixtures using enriched Ba-130

→ better distribution than printing from pure H_2O

→ but still a lot of micro-crystallization

Research internship of C. Bruhn (2022)

TH-232 AND U-233 TARGETS FOR JYFL

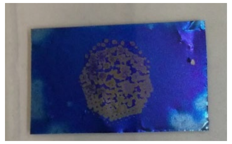
JYVÄSKYLÄN YLIOPISTO
UNIVERSITY OF JYVÄSKYLÄ

Subject of beamtime 2018:
study of the performance of DoD targets in a proton beam (50-60 MeV)

- mechanical stability?
- ability to deliver desired nuclear reaction products?
- comparison with a self-supporting metallic Th foil

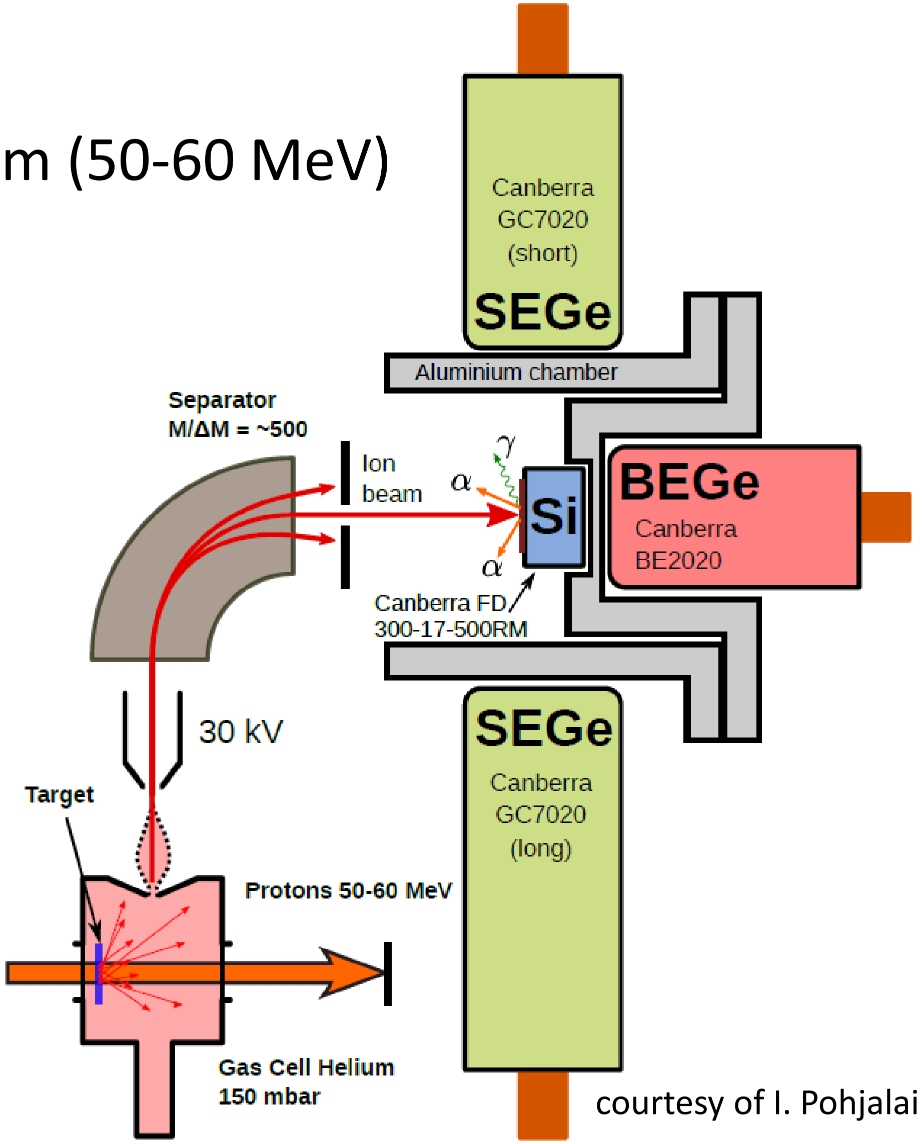
3.25 mg/cm² Th-232 foil

592 µg/cm² Th-232 on 20 µm Au foil



395 µg/cm² Th-232 on 25 µm Ti foil

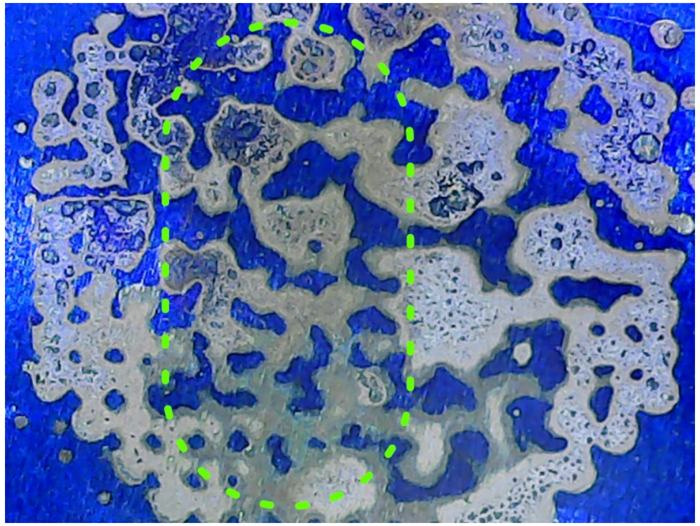
All DoD sample printed from aqueous nitrate solution and heat-treated afterwards @ 550°C



courtesy of I. Pohjalainen

TH-232 AND U-233 TARGETS FOR JYFL

Beamtime 2018



- max. intensity:
  20 µA @ 50 MeV for 5 min
- long-term stability:
  10 µA @ 50 MeV for 10 h
  ∫ = 3x10^{17} ions

courtesy of I. Pohjalainen

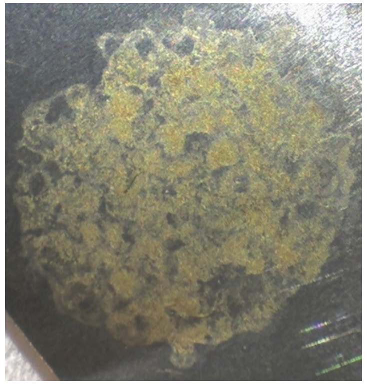
395 µg/cm² Th-232 on 25 µm Ti foil after irradiation

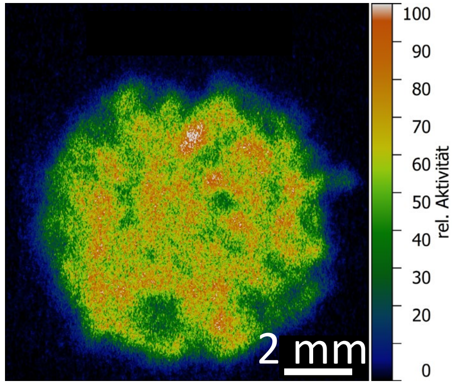
Results

– reaction products could be extracted from all used DoD targets
– all targets remained intact
– yield ≈ 30% compared to Th foil
  → inhomogeneous deposition lowers yield

Upcoming beamtime Oct. 2022:

→ test of U-233 DoD targets printed from isobutanolic solution





121 µg/cm² U-233 on 25 µm Ti

B.Sc. thesis of Ch. Sirleaf (2019)

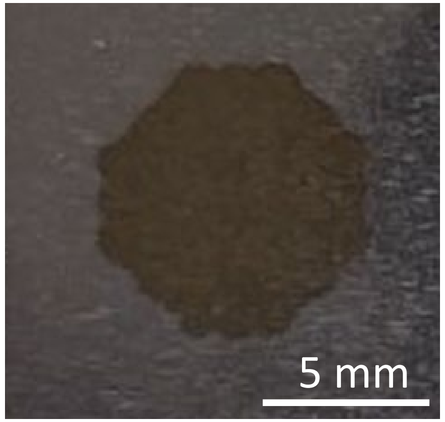
POLYMER ASSISTED DEPOSITION (PAD)

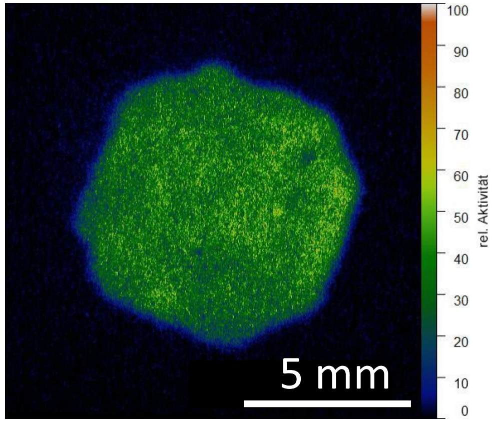
- idea: adding of polymer agent to create homogeneous deposition
- thermal decomposition of polymer necessary afterwards
- Polyethylenimine (PEI) solvable in water or ethanol

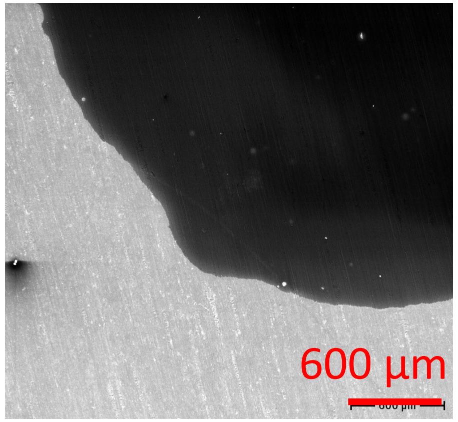
PEI monomer

<u>First experiments:</u>

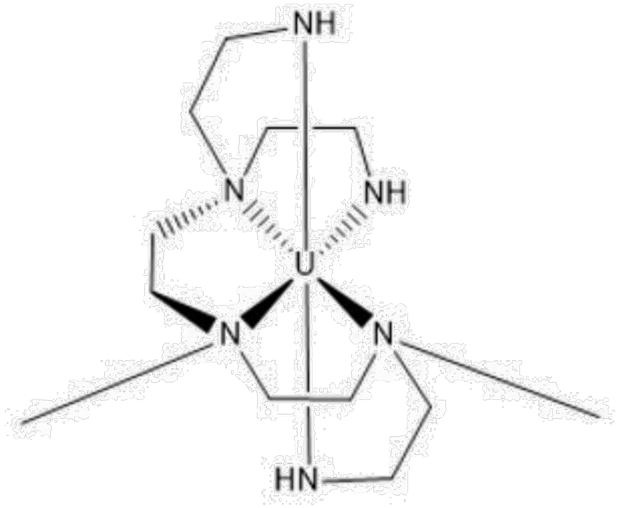
- solving of $UO_2(NO_3)_2$ in ethanol
- adding of PEI (1800 g/mol) solution with pH 6.5 – 9.2
- optional adding of EDTA for additional complexation







0.3 µg/cm² U-233 on Ti foil, after thermal decomposition

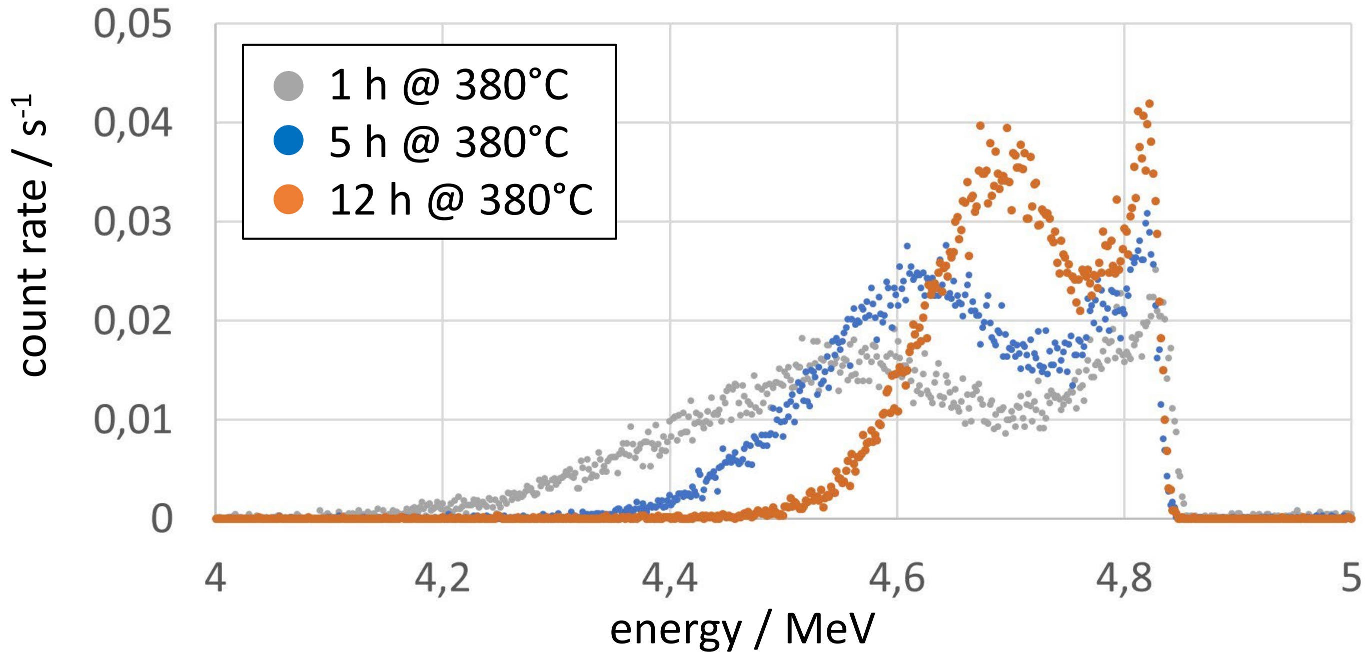


coordination of U by PEI

B.Sc. thesis of T. Schwab (2021)

POLYMER ASSISTED DEPOSITION (PAD)

α-spectra of 0.3 µg/cm² U-233 target at different stages of PEI decomposition



→ effect of proceeding polymer decomposition
→ layers with FWHM < 20 keV were possible

First PAD + DoD experiments promising

- tested two molecular weights (MW) (PEI-1800 and PEI-10000)
- lower MW provided better patterns and faster decomposition
- so far, stable layers only achieved with thickness < 10 µg/cm²
- higher U conc. resulted in inhomogeneous layers

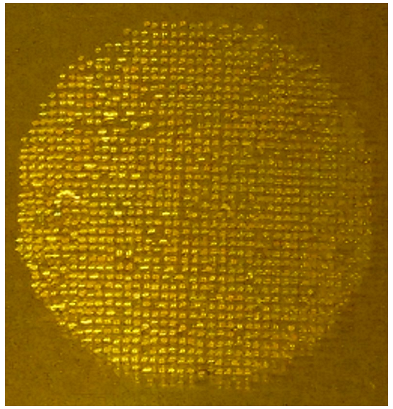
→ further investigations needed (e.g. using lower MW, but higher conc.)

B.Sc. thesis of T. Schwab (2021)

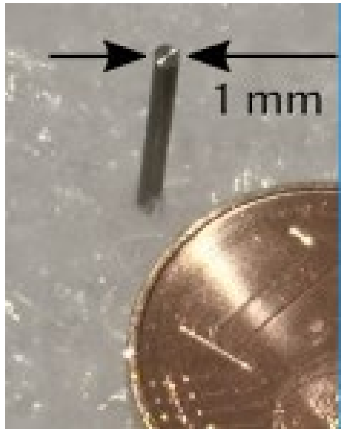
SUMMARY & OUTLOOK

DoD offers advantages compared to established electrodeposition methods

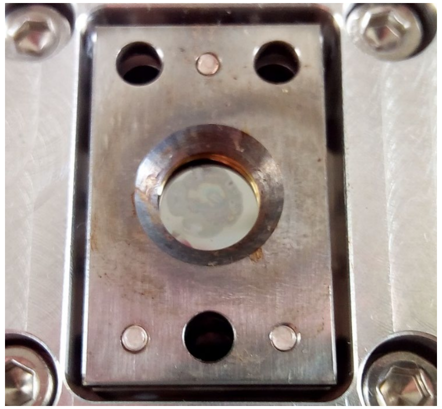
- no need of conductive substrate
- very flexible target geometry
- applicable to unconventional or fragile substrates
- broad variety of solvents applicable, including PAD methods



^{170}Er deposition on polyimide foil



Ho-163 sample holder for PENTATRAP



229Th layer on mounted Pyro detector for UCLA

→ more investigations on polymer-based solutions planned

→ idea for post-processing of inhomogeneous depositions with tetraethylene glycol (TEG)

Thank you for your attention!