Medical radionuclide research activities at JRC-Geel

16th Varenna Conference on Nuclear Reaction Mechanisms
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Outline

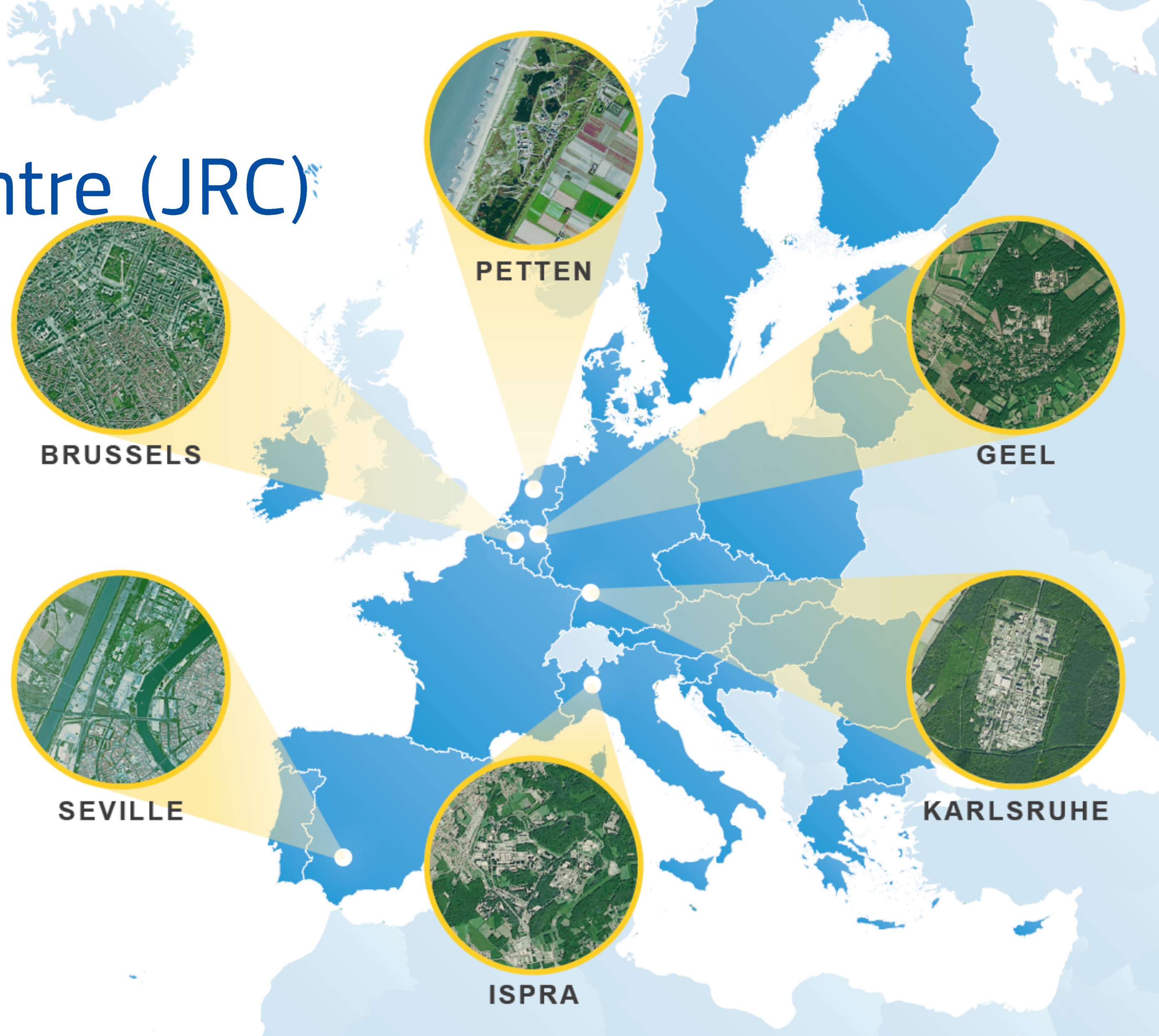
- The JRC and JRC-Geel accelerator facilities
- The new electron beamline for photonuclear reaction studies
  - Layout, instrumentation, main characteristics and challenges
- Radionuclide production via nanoparticle irradiation: the ir-NANO project
  - The concept, experiments, first results
- Summary

The Joint Research Centre (JRC)

The European Commission's science and knowledge service, supporting EU policies with independent evidence throughout the whole policy cycle

Headquarters in Brussels
and research facilities located
in **5 Member States**:

- Belgium (Geel)
- Germany (Karlsruhe)
- Italy (Ispra)
- The Netherlands (Petten)
- Spain (Seville)



Photonuclear reaction studies

The new GELINA electron beamline

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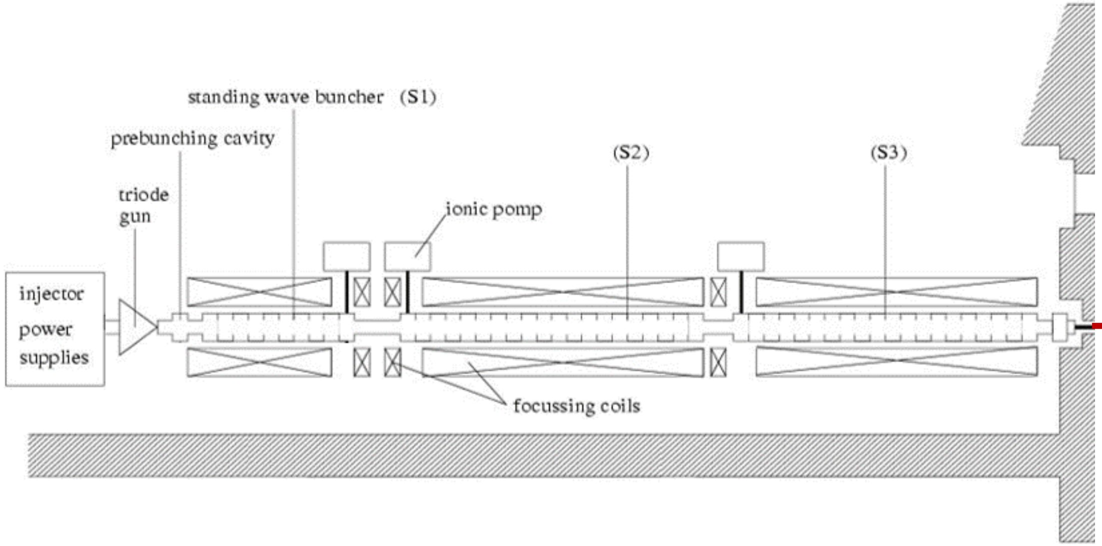
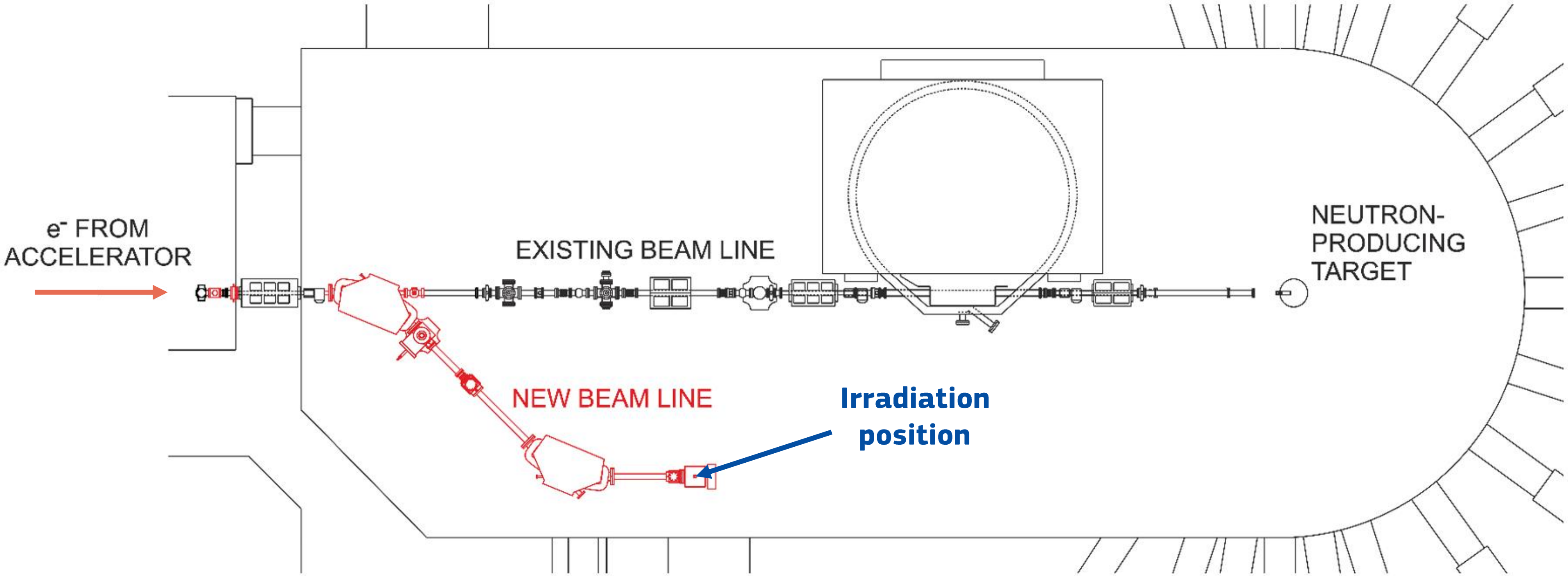
GELINA (Geel Electron LINAc)

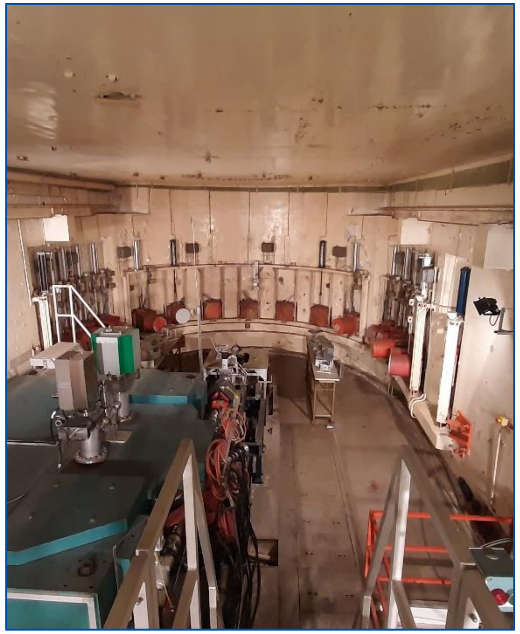
- ↙ 3 accelerating sections, E_{max} = 130 MeV
- Normally used to hit a uranium target with ~100±30 MeV electrons producing pulsed white neutron beams that propagate to experimental stations at different distances
- Study of neutron-induced reactions via ToF technique in the context of nuclear safety, security and safeguards



Neutron target

New beamline in the GELINA target hall

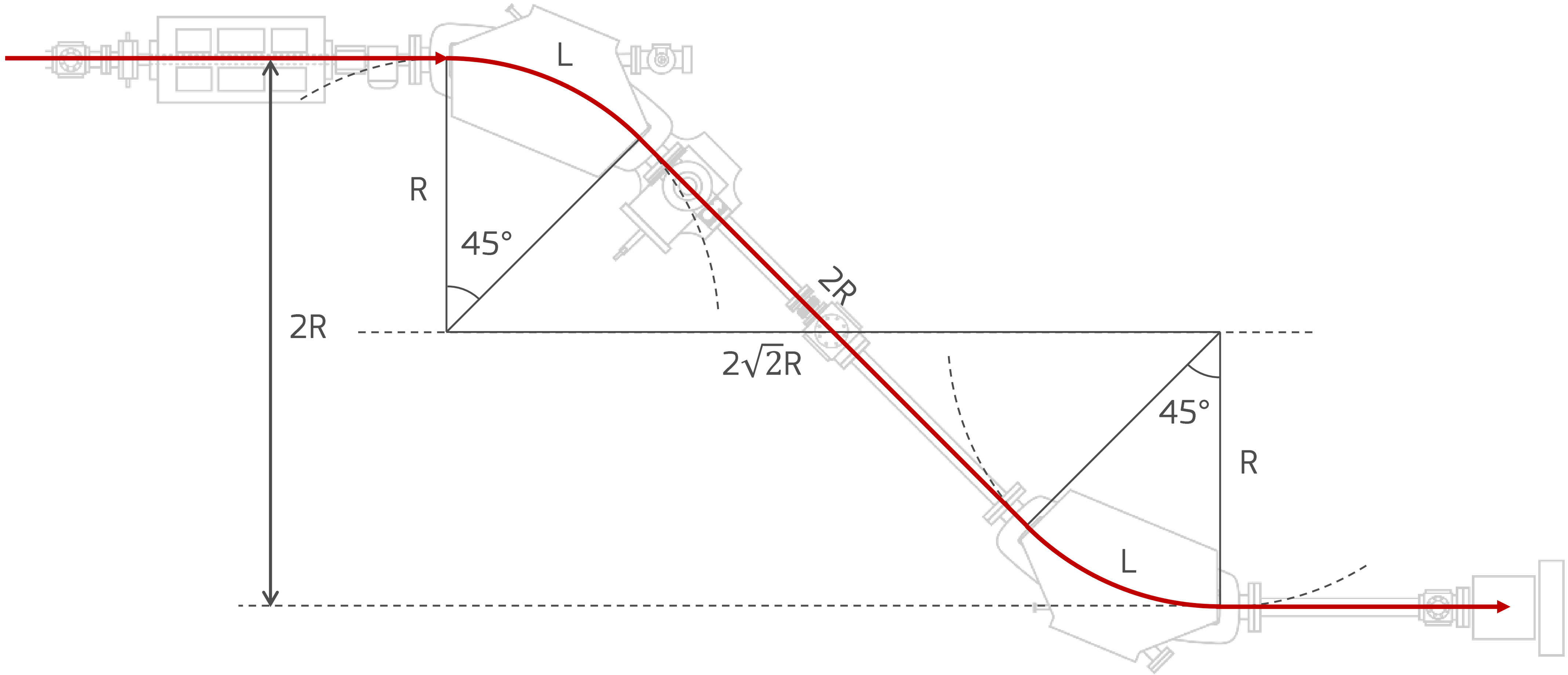




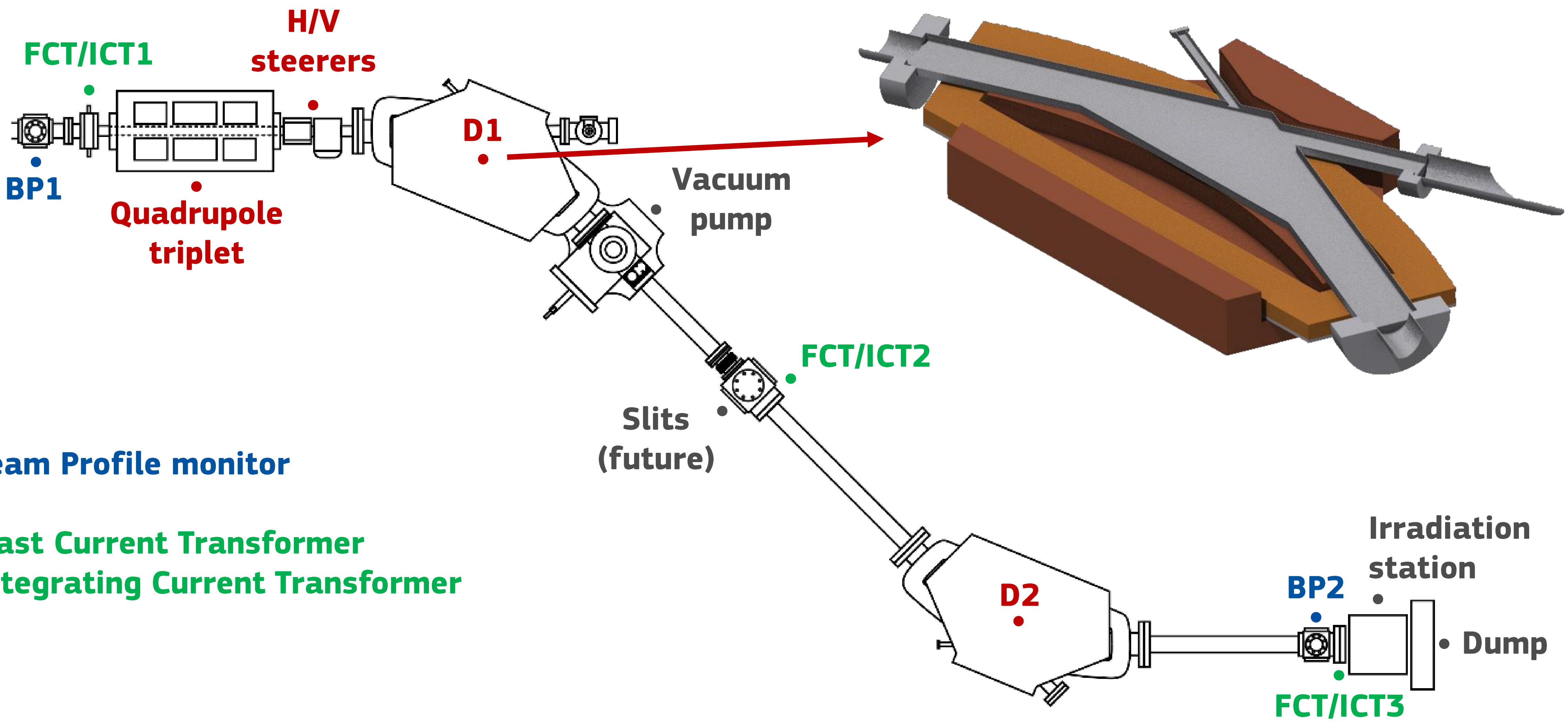
- The electrons are deflected off the existing beamline by two 45° bending dipoles
- Electron beams of selected energy are delivered onto a radiator (Pb) to generate Bremsstrahlung photons to be used for (γ,x) reaction studies

General layout

- Final beam displacement: 2.3 m (twice the dipole bending radius)



Beamline layout and instrumentation

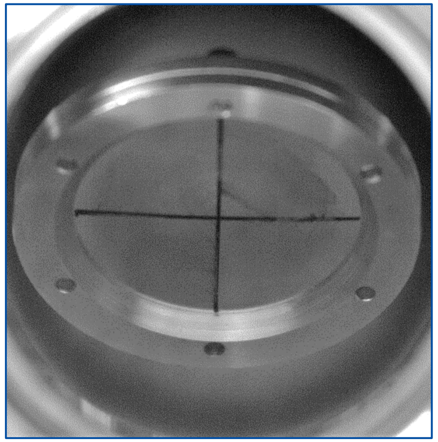


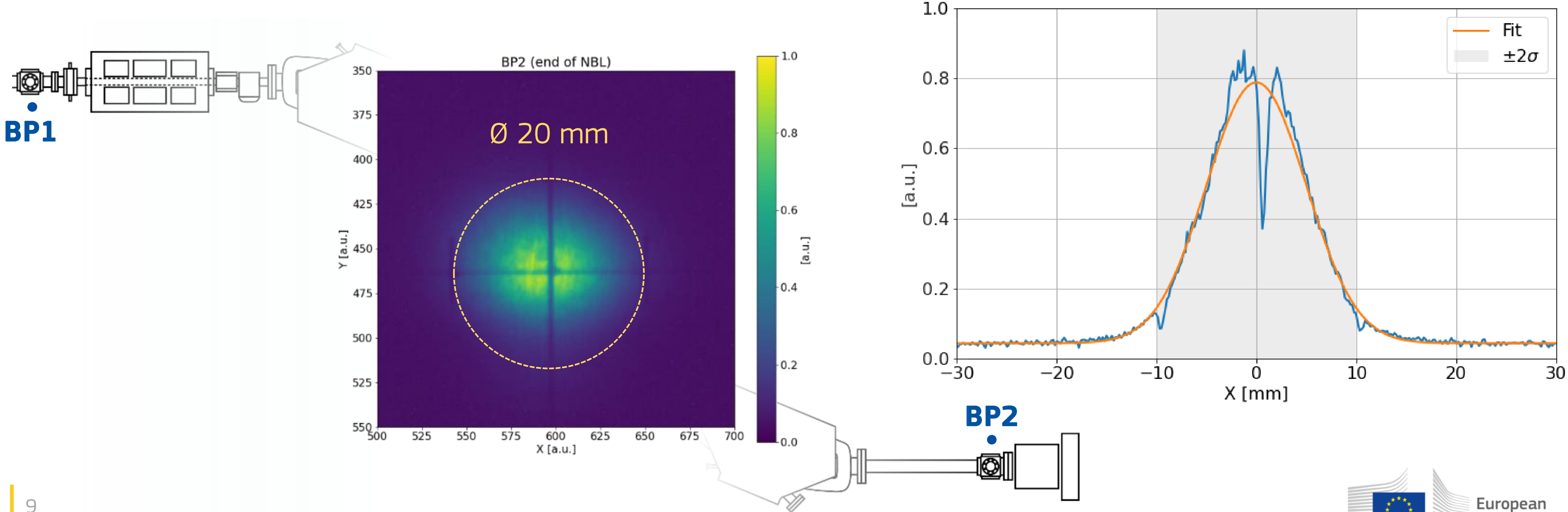
BP: Beam Profile monitor

FCT: Fast Current Transformer
ICT: Integrating Current Transformer

Electron beam profile

- Beam profile monitoring via optical transition radiation (OTR) on thin aluminium foils
- Insertable monitor at beamline entry, permanent monitor just upstream of irradiation location
- σ = 5 mm at beamline exit





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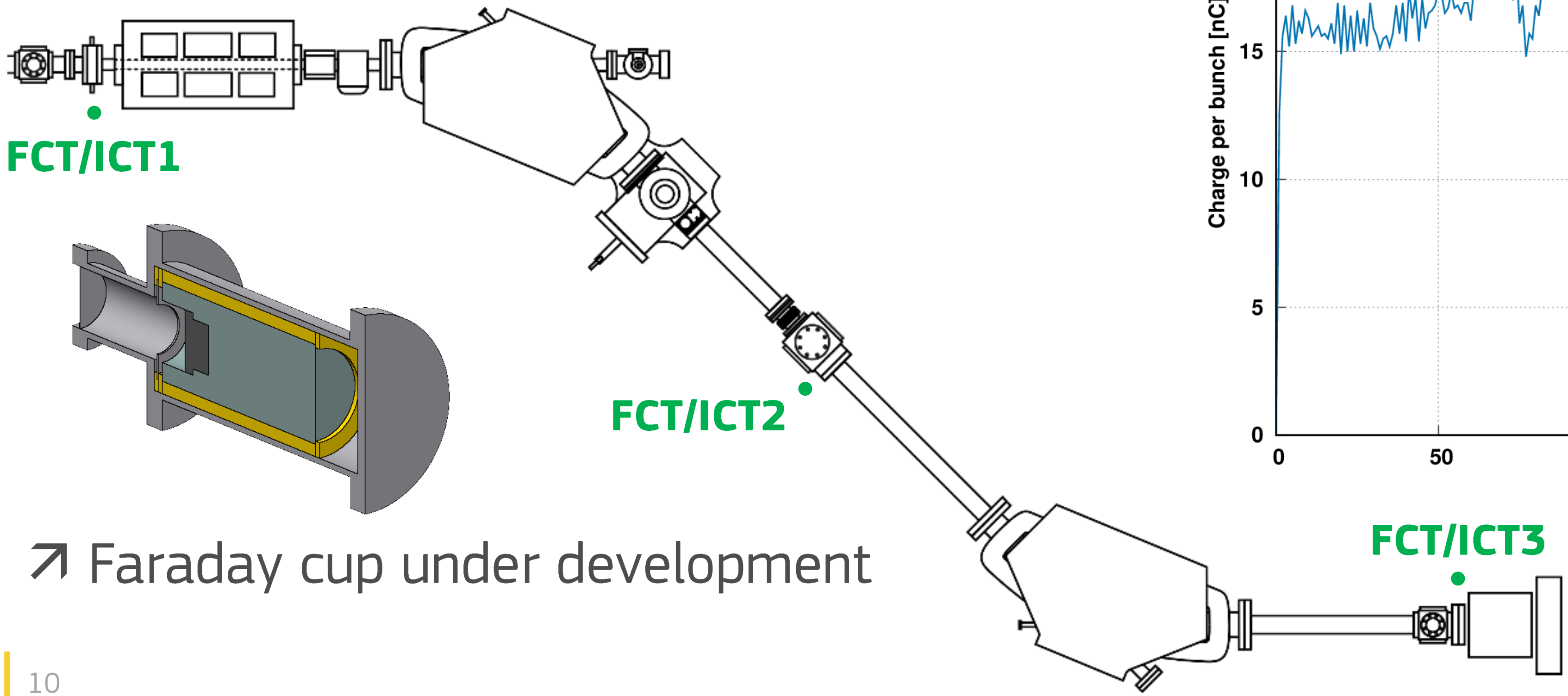
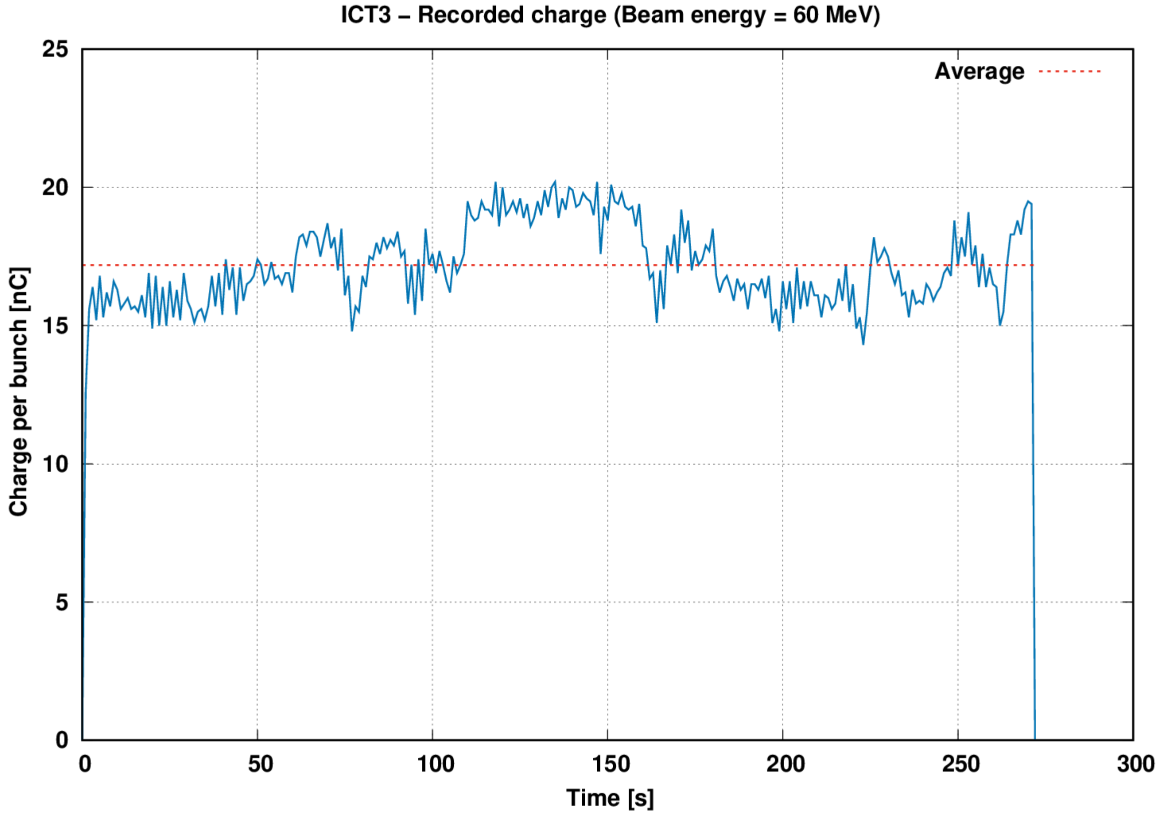
Electron beam current

- Bergoz integrating current transformers
- Current monitored at beamline entrance (i.e. accelerator exit), midpoint between two dipoles, beamline exit (i.e. current delivered on radiator)



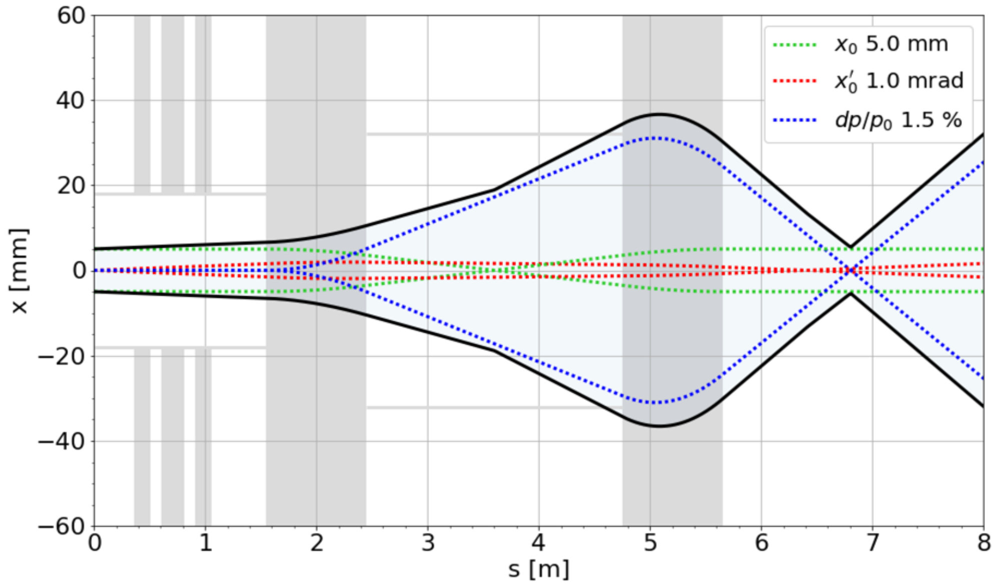
↗ Faraday cup under development

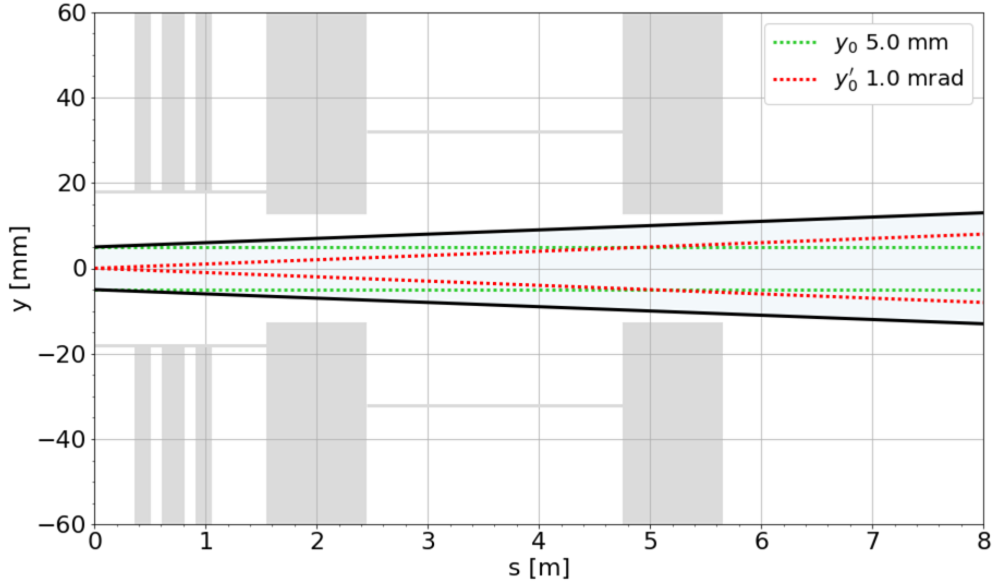
- Example: ICT3 current monitoring during 4.5 minutes ↓

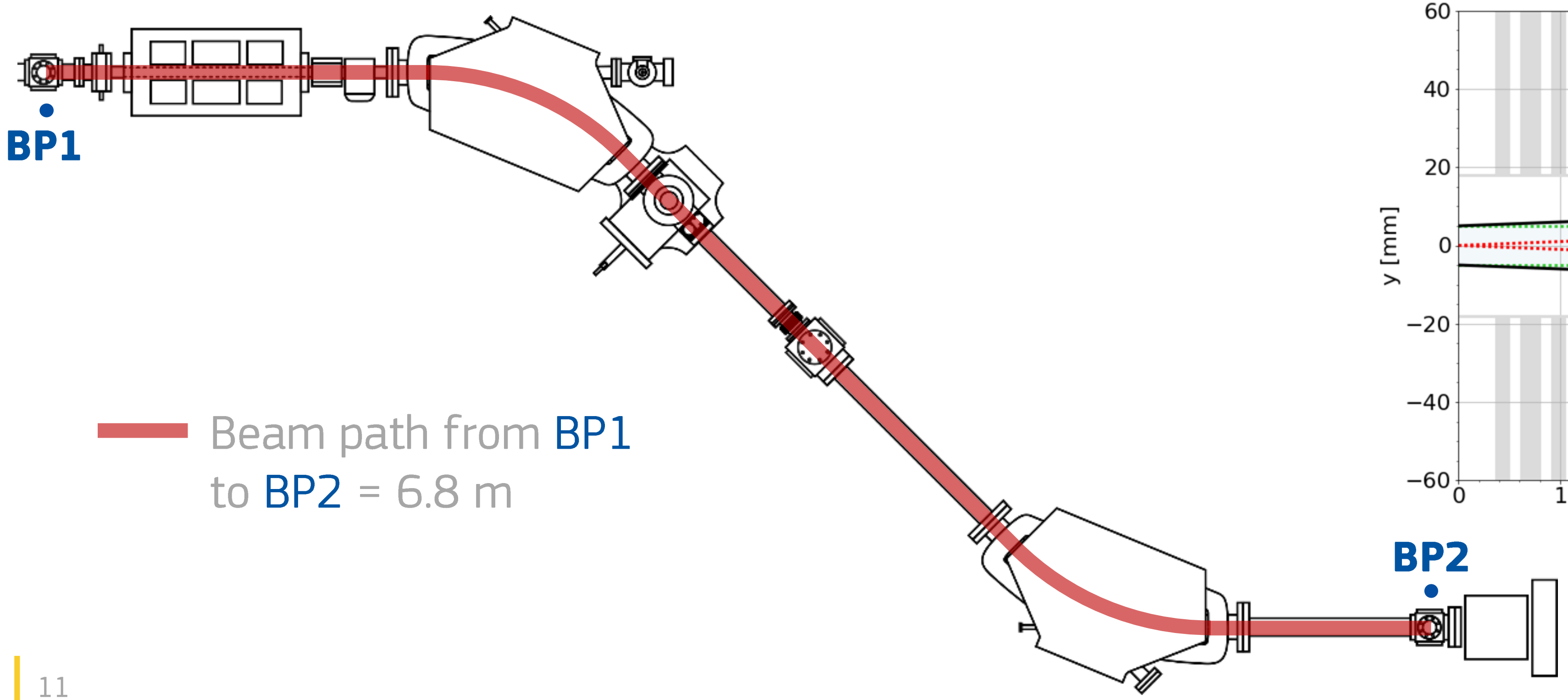


Beam optics

- Transverse beam dimension on the horizontal plane is dominated by the momentum spread
- The momentum acceptance is about ±1.5%
- Can $^{dp}/_{p}$ be reduced to this level?



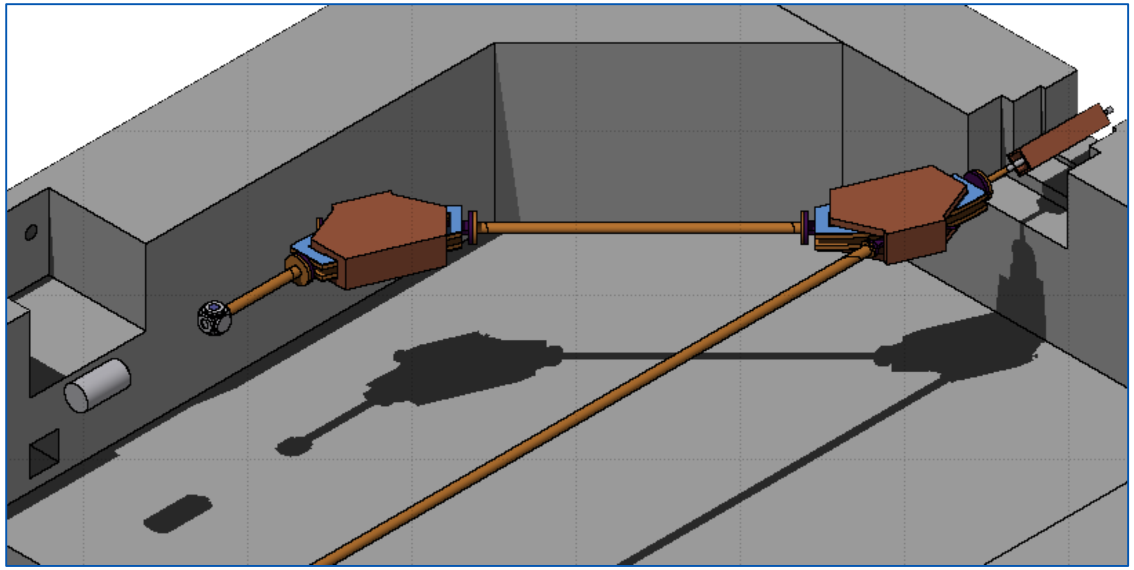


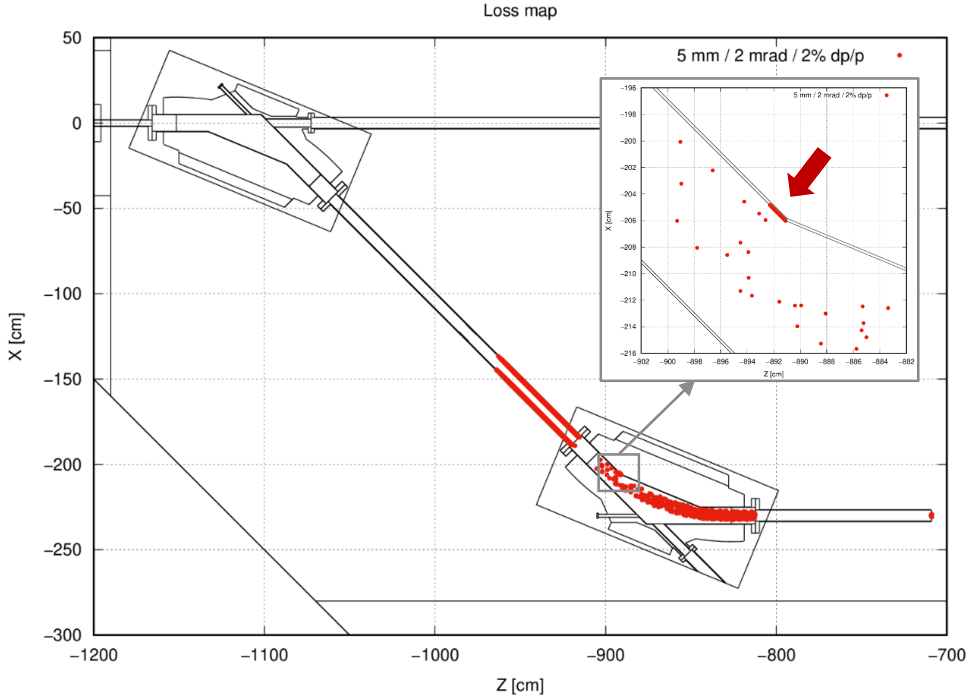


Beam path from BP1 to BP2 = 6.8 m

Momentum spread and beam losses

- Electron energy in normal operation is 70-130 MeV due to RF power depletion; this is a $^{dp}/_{p}$ of ±30% and would lead to unacceptable beam losses
- Expected beam losses for various conditions estimated with Monte Carlo model of beamline





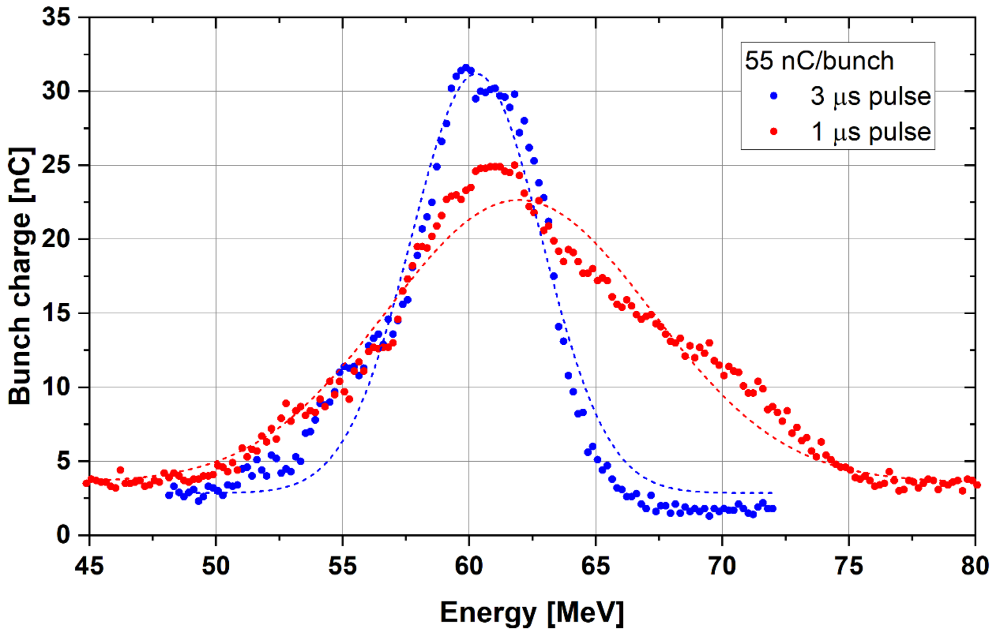
- Reduction of momentum spread through:
  - Longer pulses (2-3 µs compared to 10 ns in normal operation)
  - Reduced charge (tens rather than hundreds of nC/bunch)

Electron beam energy

- Beam energy spectrum can be obtained by observing the current on ICT2 (beamline midpoint, after first bend) as a function of current applied to D1

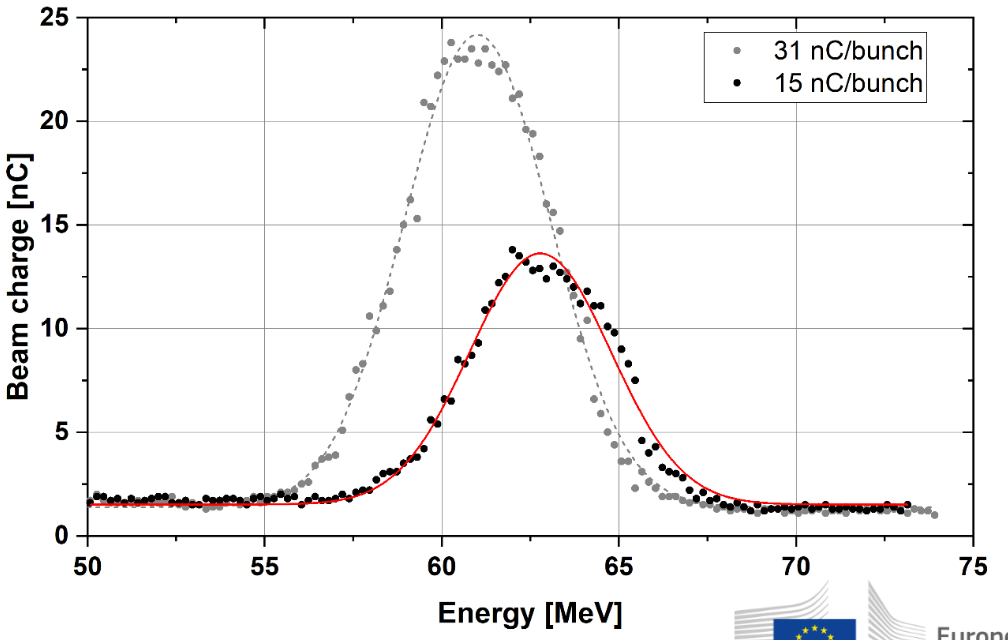
Effect of **longer pulse**

- 55 nC/bunch, 1 μs pulse
  - σ ~ 8.5%, 39% transmission
- 55 nC/bunch, 3 μs pulse
  - σ ~ 4.3%, 52% transmission



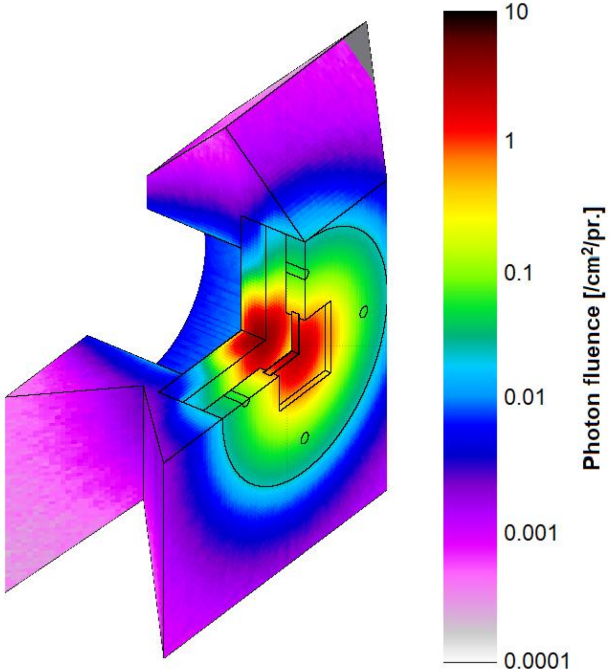
Effect of **lower current**

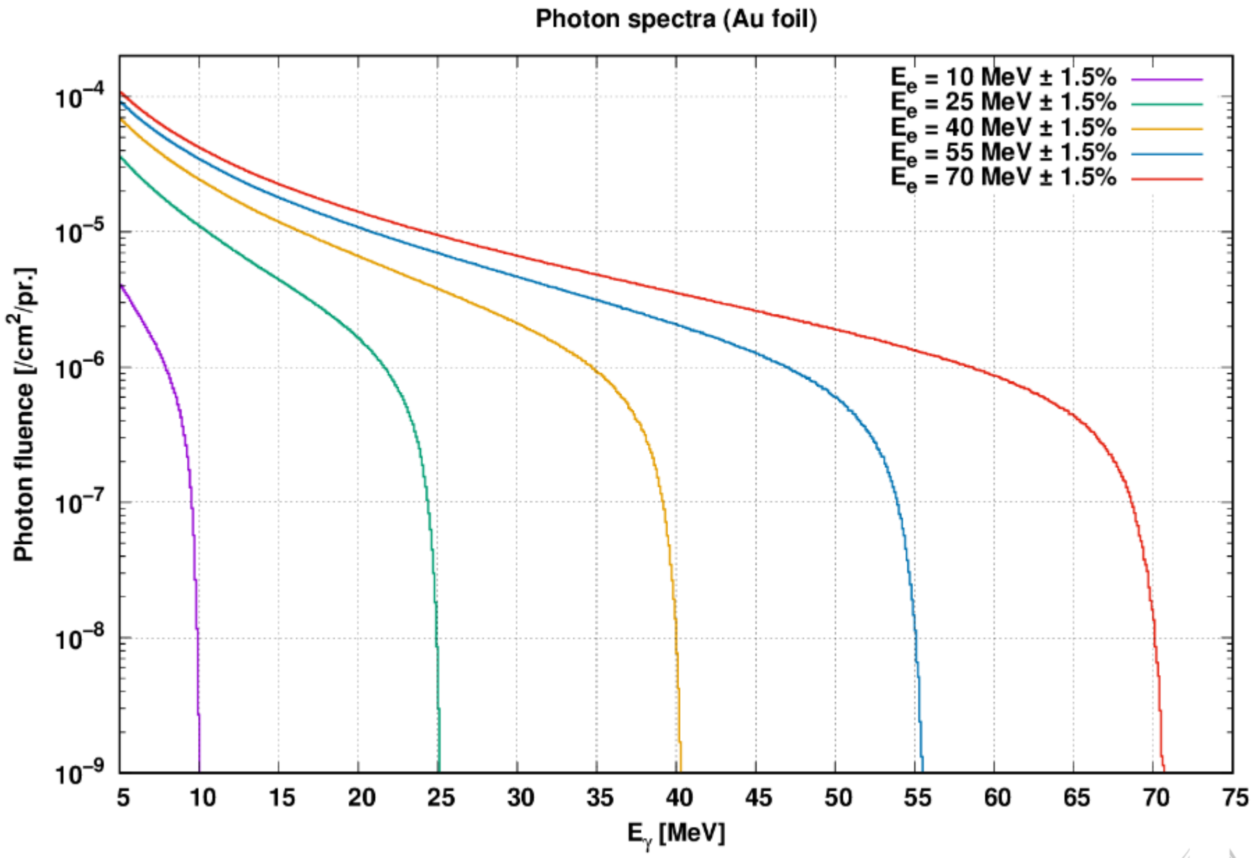
- From 30 to 15 nC/bunch
  - Slightly higher average energy
  - σ ~ 3.2%, ~80% transmission



Simulations

- Objective: validation of a Monte Carlo model reproducing experimental reaction rates
  - Use model for future irradiation planning
  - Use simulated photon fluence as ingredient in data analysis, estimate neutron contribution etc.





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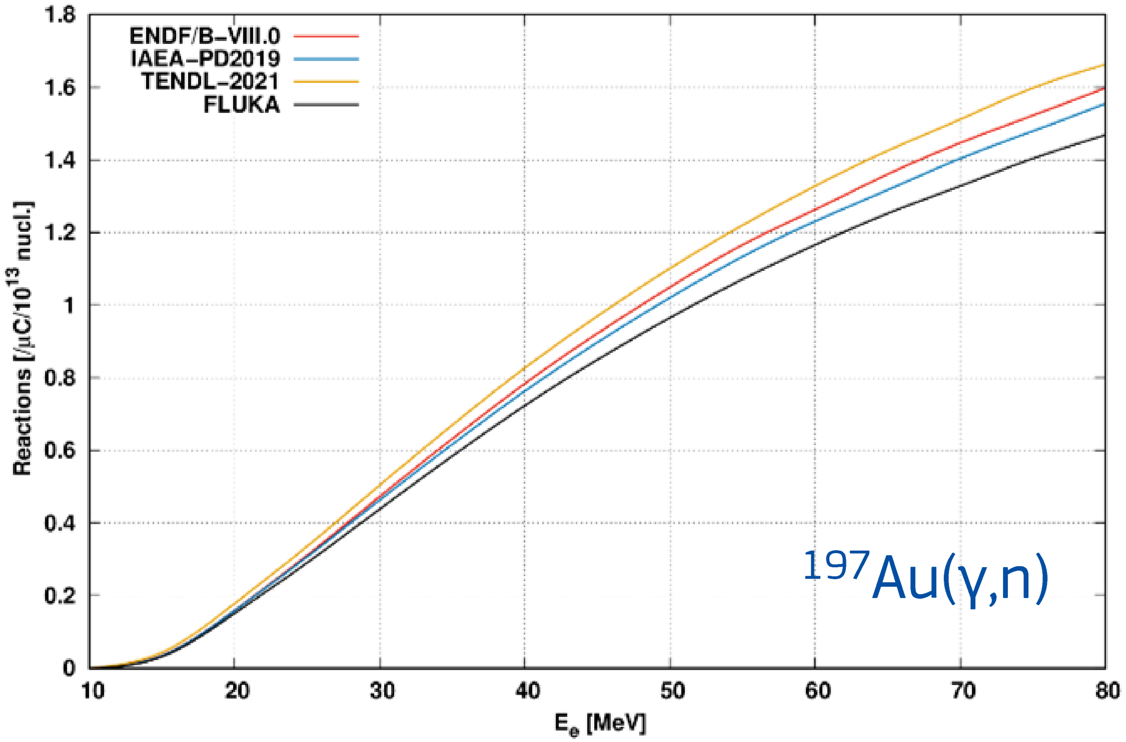
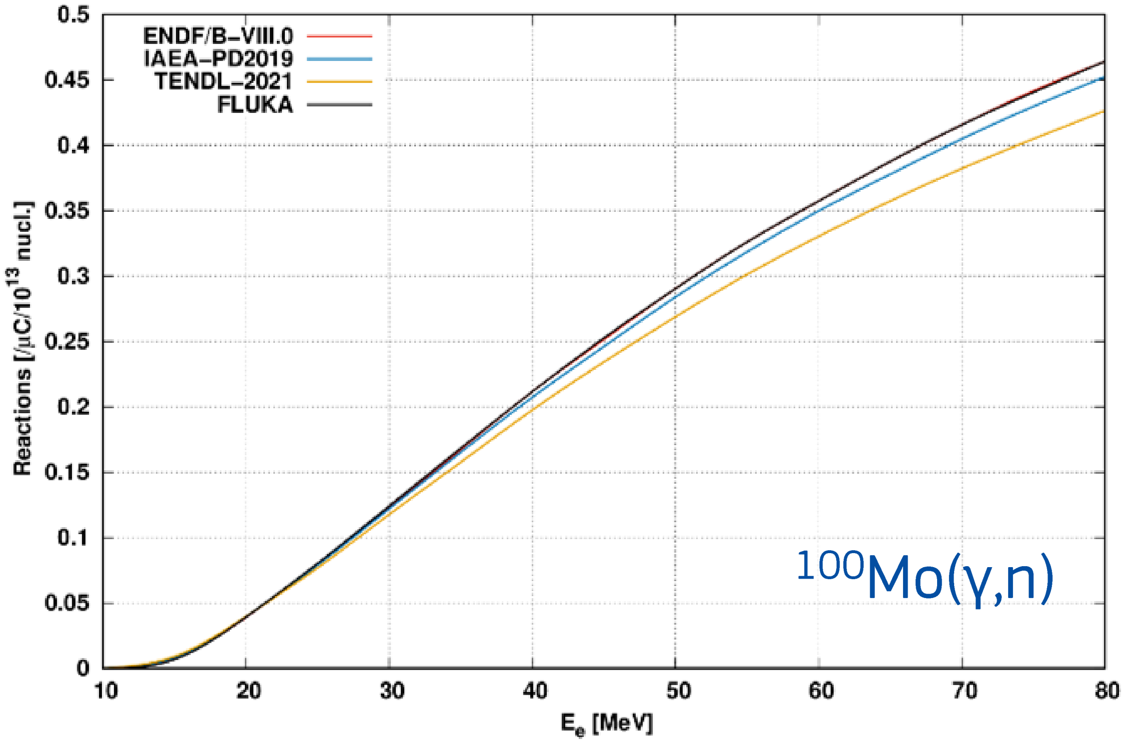
Reaction rates vs. beam energy

- Reaction rates (for the specific setup) obtained by convolution of simulated photon spectrum with reaction cross-sections from ENDF/B-VIII.0, IAEA-PD2019, TENDL-2021 libraries and FLUKA)

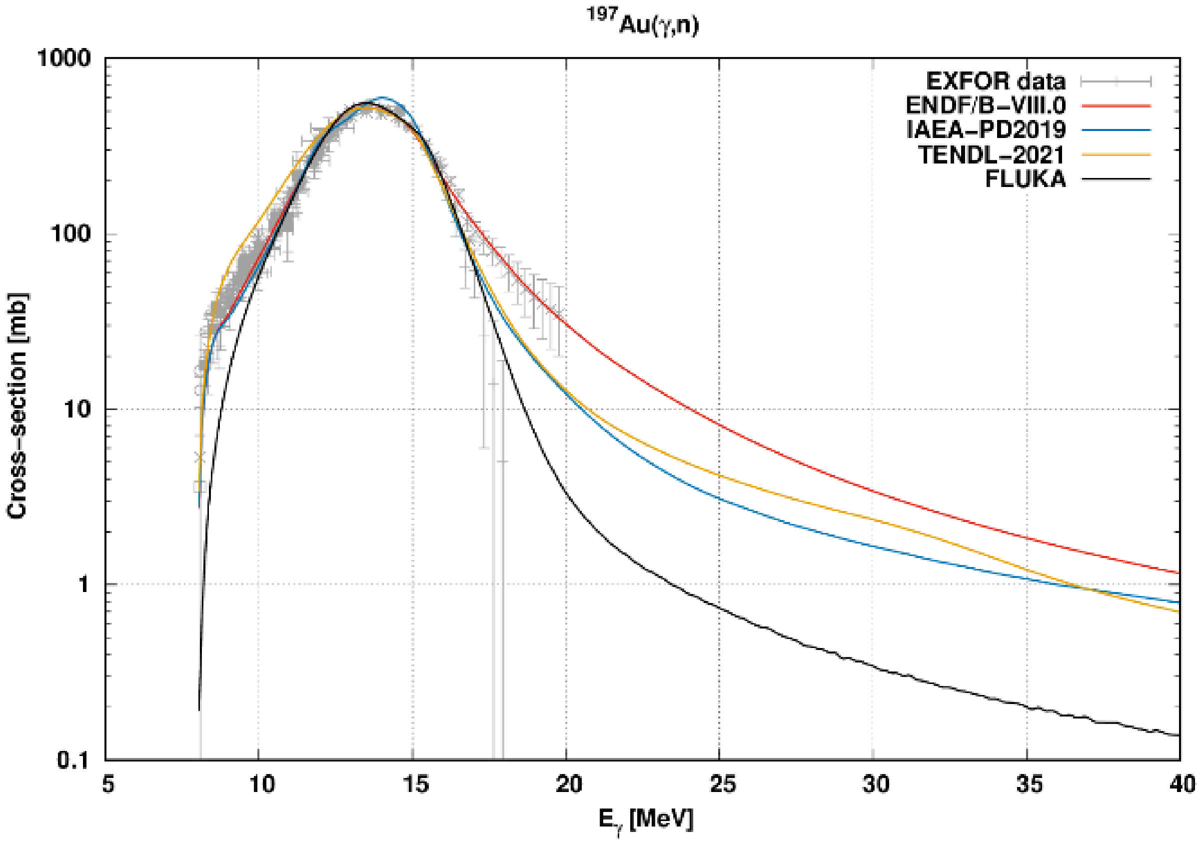


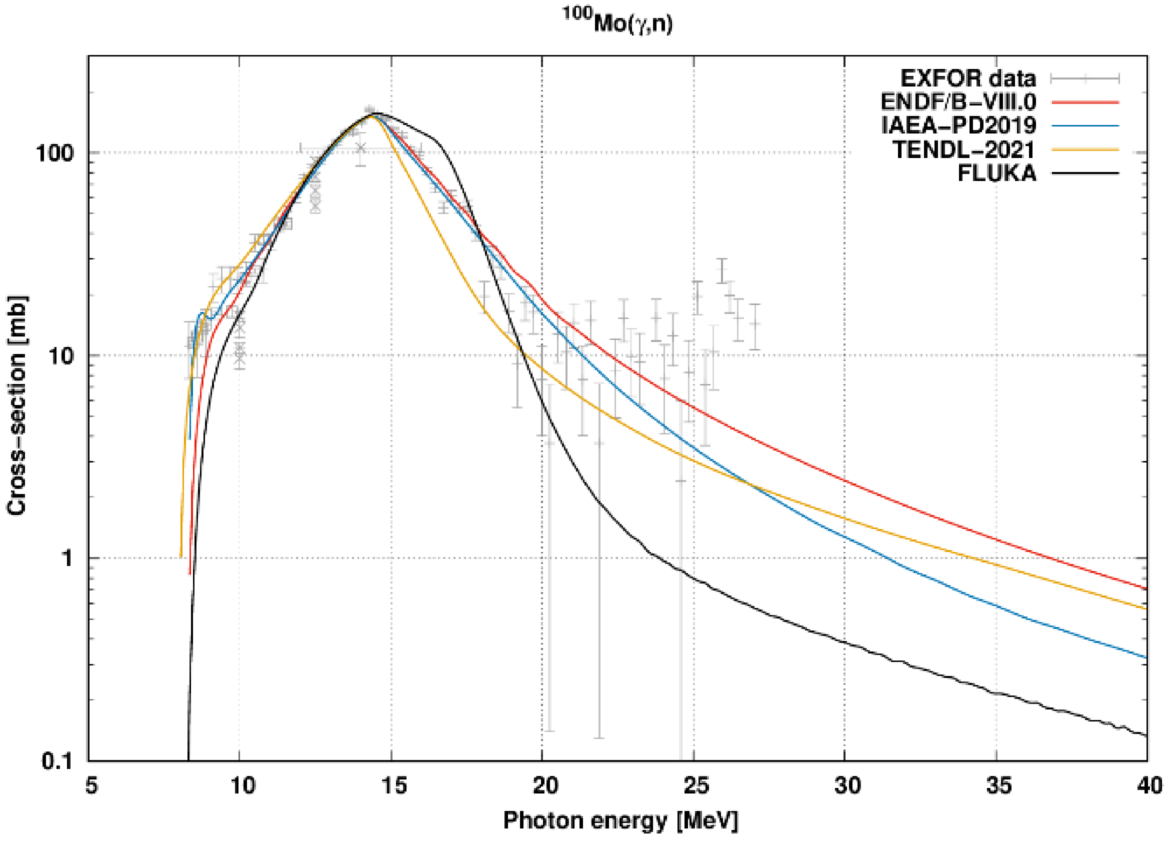


- Non-negligible differences even for gold which is routinely used as reference for photon activation analysis

Status of data

- This reflects both scarcity of data and diverging extensions to higher energies
  - E.g. ^{197}Au(γ,n) ↓, ^{100}Mo(γ,n) ↘





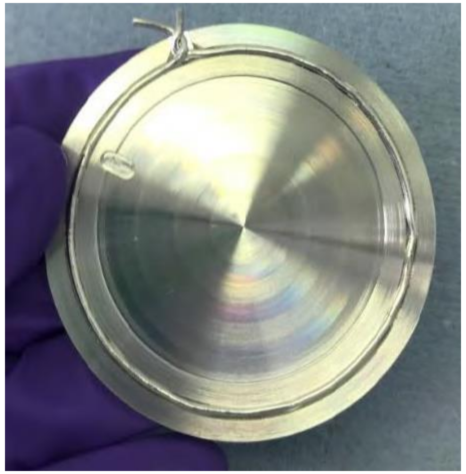
An ongoing project: ^{225}Ac for targeted α-therapy

- Production of ^{225}Ac via ^{226}Ra(γ,n) in collaboration with SCK-CEN (Belgium)

sck cen
Belgian Nuclear Research Centre

- Target development at SCK-CEN

- In progress: development of sample encapsulation (Al capsule with In seal →); irradiation of dummy samples in July 2023 to determine activation of the container, relevant impurities etc.



- Barium deposit included to check stability and study feasibility of activity measurement without removing target material from container

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Radionuclides via nanoparticle irradiation

The ir-NANO project

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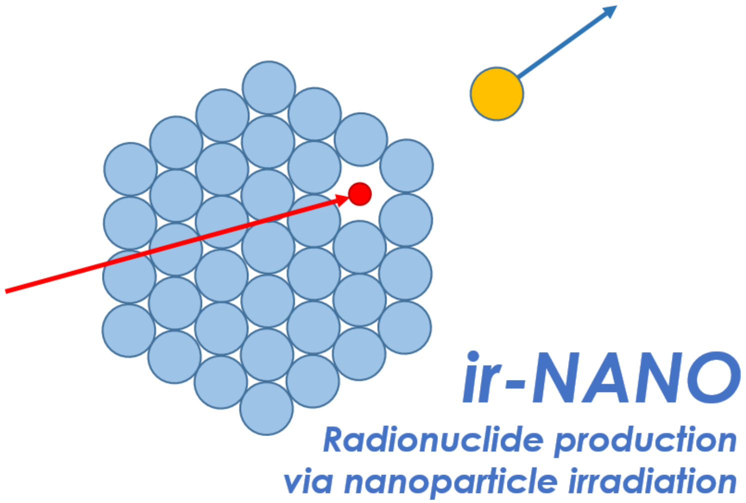
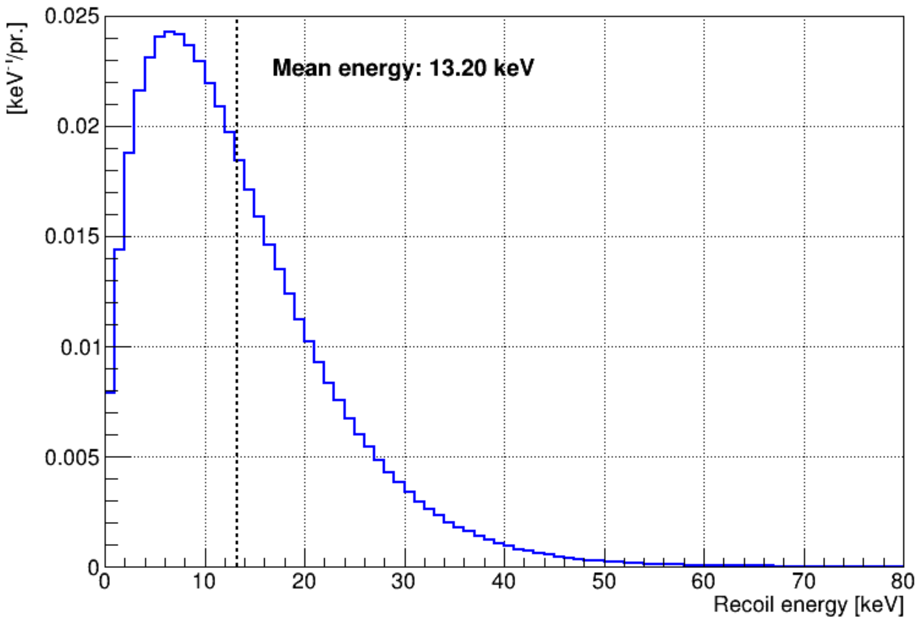
The idea

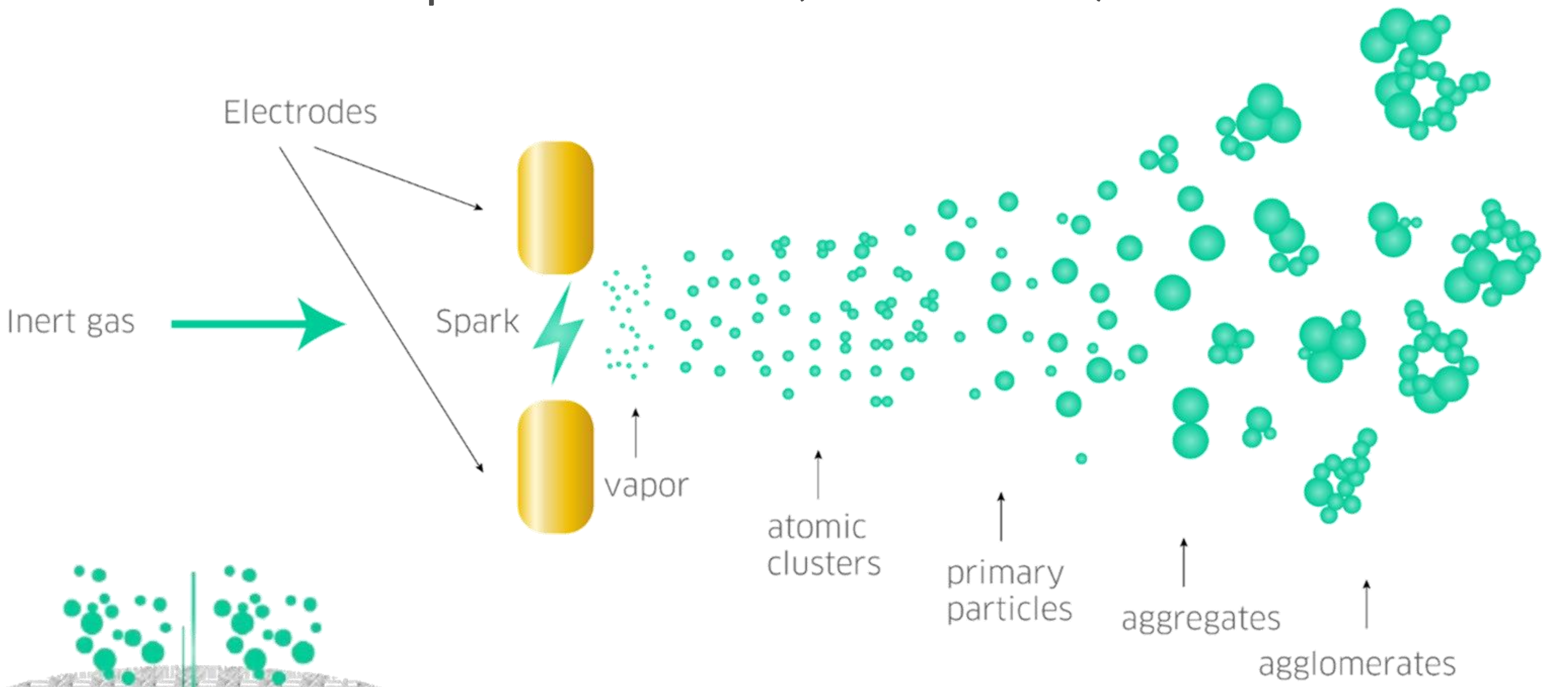
- Neutron or photon irradiation of Mo for ^{99}Mo production via ^{100}Mo(γ,n) or ^{100}Mo(n,2n)
- Molybdenum in nanoparticle form
  - ^{nat}Mo for the moment (9.74% ^{100}Mo)
- The basic idea: primary recoils can escape from the nanoparticle if its size is smaller than the range of the recoils, and be chemically extracted
  - E.g. energy spectrum of ^{99}Mo recoils from ^{100}Mo(γ,n) with 15 MeV photons →
- ir-NANO initiated as an Exploratory Research project, now part of regular work programme

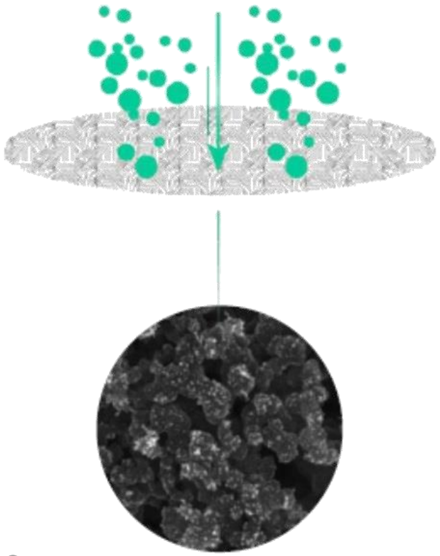




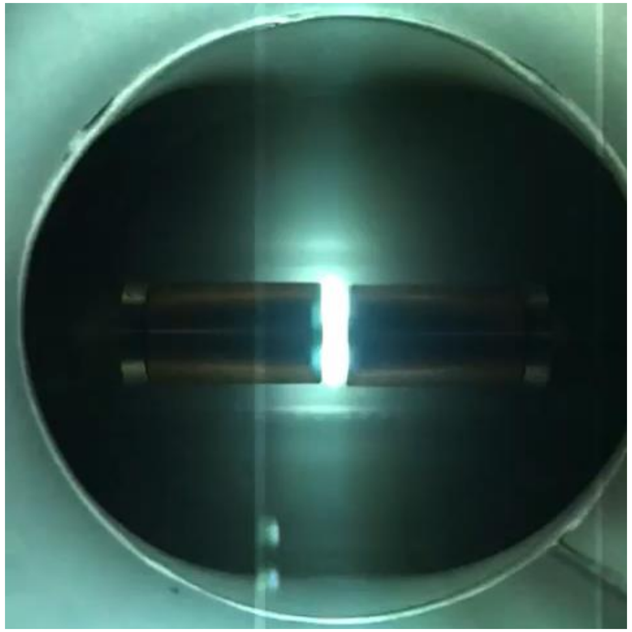
Nanoparticle production

- Production via spark ablation (JRC-Petten)



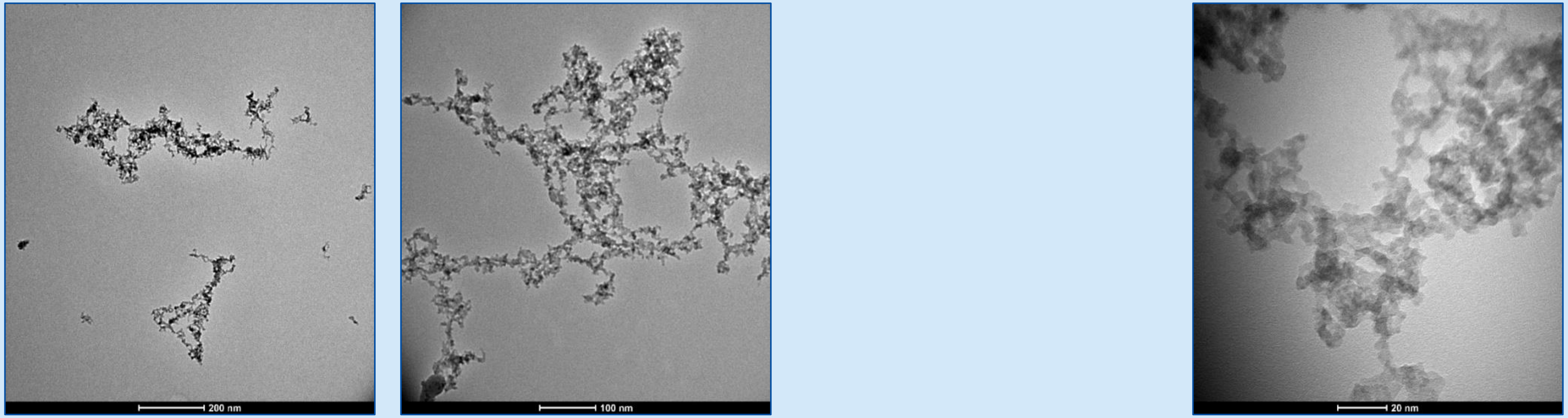


- ← Collection by filtration

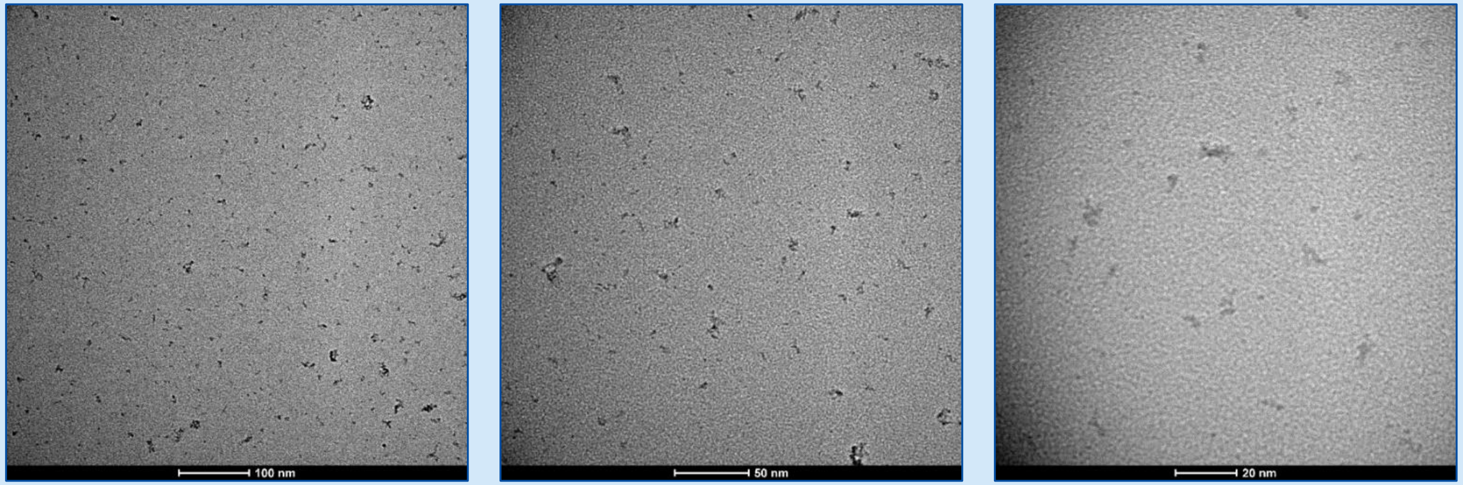


Nanoparticle production

- The inert gas flow defines the size of the agglomerates:
  - 1 L/min →



  - 10 L/min →



Neutron irradiations at MONNET

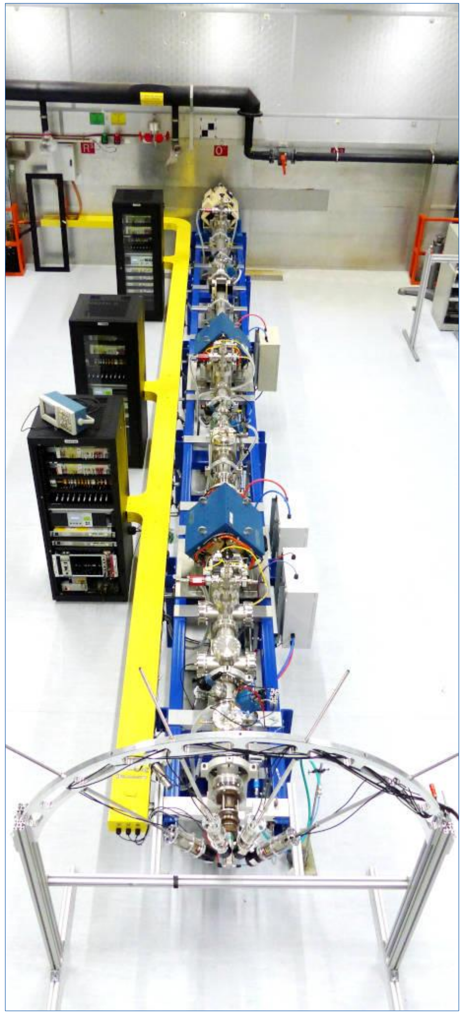
- 3.5 MV tandem van de Graaf light ion accelerator (protons, deuterons)

- Commissioned in 2020 (replacement of existing van de Graaf)

- Continuous or pulsed beam

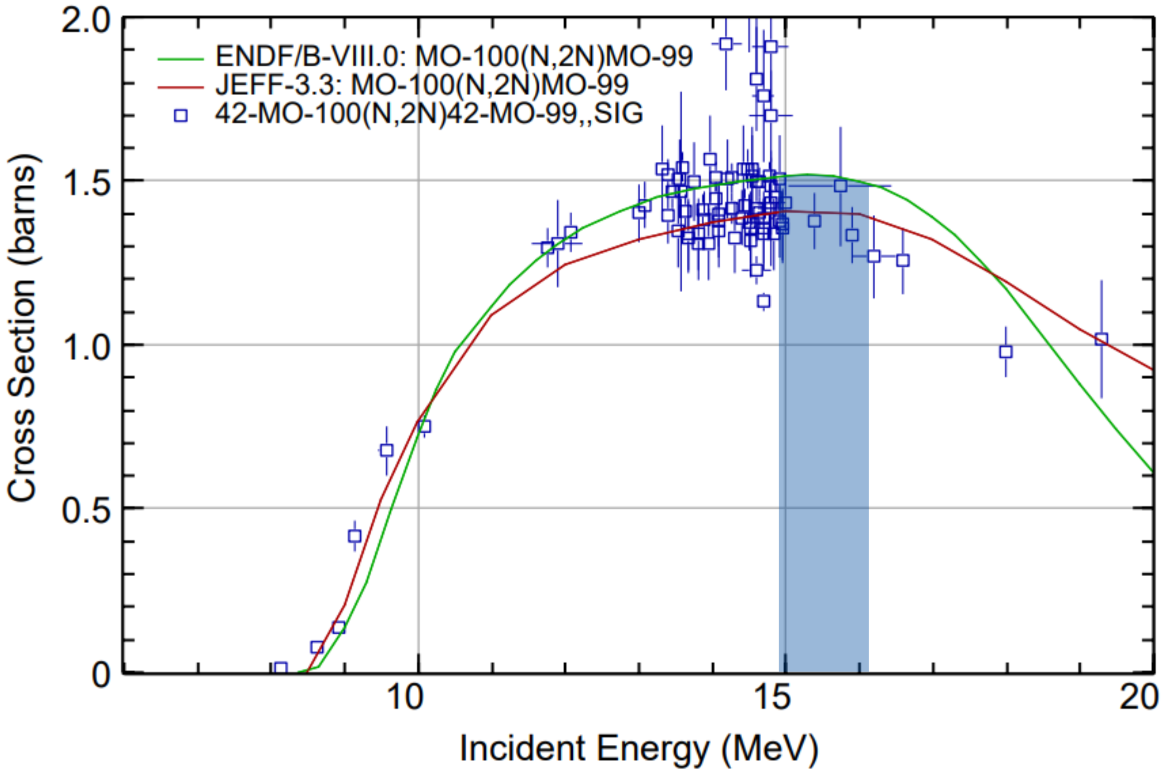
- Quasi mono-energetic neutrons produced by different clear reactions: ~500 keV to 19 MeV

- Neutron flux: 10^6 n/(s·sr) to 10^9 n/(s·sr)



Neutron irradiations at MONNET

- The $^{100}Mo(n,2n)$ reaction cross-section is highest for incoming neutrons of ~15 MeV energy



- Neutrons with an energy around 15.5 MeV, covering the cross-section maximum, were produced via the $^{3}H(d,n)$ reaction (deuteron irradiation of a tritium target with E_d~820 keV)

Neutron irradiations at MONNET

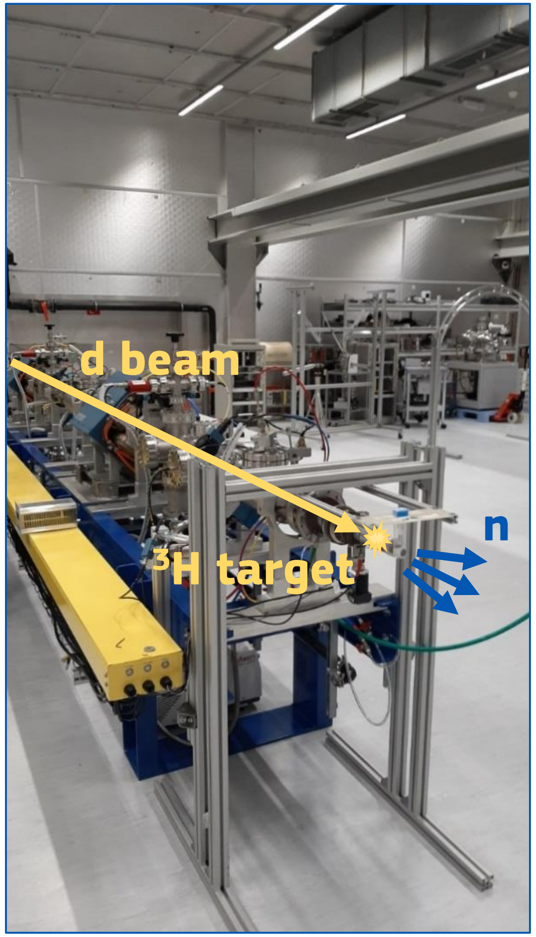
- ~20.6 mg / sample (i.e. ~1.35 mg of ^{100}Mo)

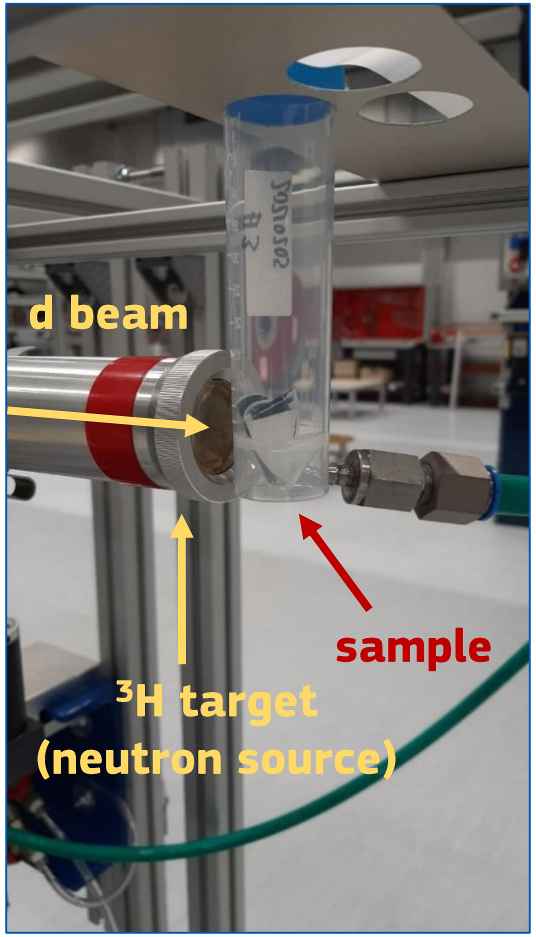
- Samples are placed at the end of the MONNET beamline, near the neutron source, in order to maximise the flux

- Irradiations last up to ~24 h; the neutron flux is monitored with ^{3}He counters

- Average integrated flux delivered on sample: 5.7×10^{11} cm^{-2}

- Average induced ^{99}Mo activity per sample: 18 Bq

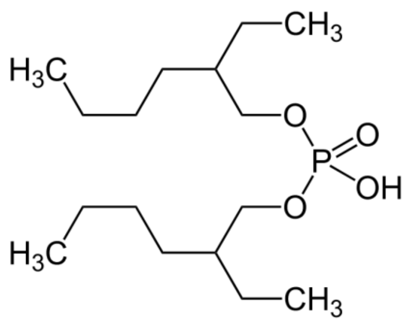




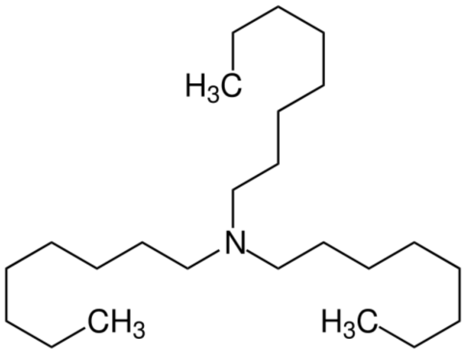
Extraction

- Solid-liquid extraction using extracting agents from metallurgy

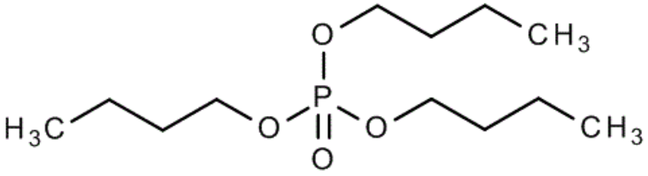
- MoO_3 agglomerates are not stable in water; chloroform preferred as solvent (octanol tried and discarded)



Bis(2-ethylhexyl)phosphate
HDEHP



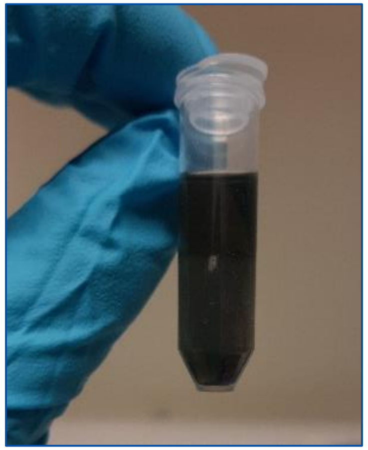
Trioctylamine
TOA

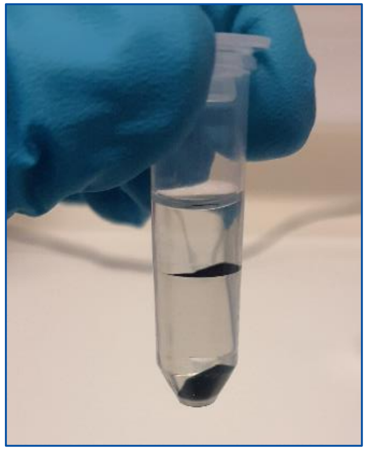


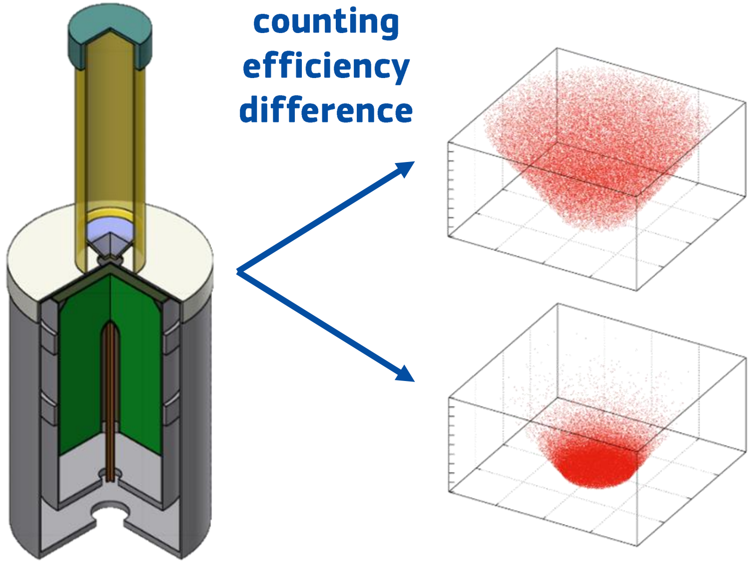
Tributylphosphate
TBP

Activity measurements

- Post-irradiation measurements of the induced ^{99}Mo activity are performed with HPGe detectors in Pb shielding…
- …followed by centrifugation and extraction of the supernatant…
- …and the final activity measurement



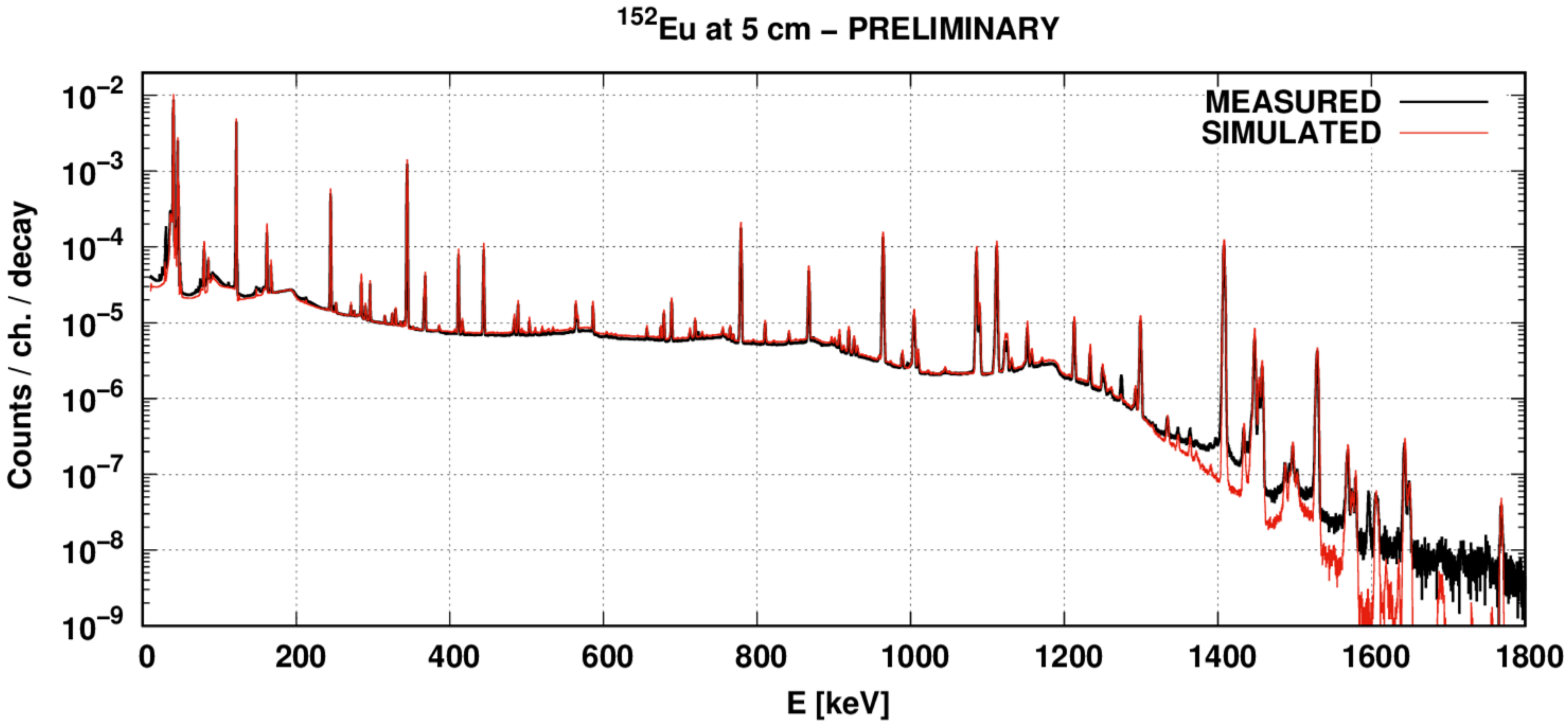


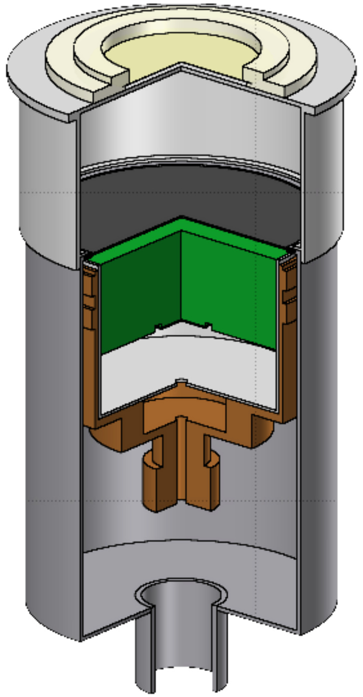


- A challenge: liquid samples with unknown or evolving radioactive material distribution at close counting geometries
- Monte Carlo models of the detectors needed to estimate the counting efficiency

Activity measurements

- Ongoing project to develop a full Monte Carlo model incorporating realistic decays to account for coincidence summing effects, important at close counting geometries





Recent results

- Post-irradiation material analysis performed at JRC-Ispra
- Different extracting solutions and conditions investigated

Extracting solution (chloroform-based)	Irradiation mode	Activity extracted	Mo extracted	A_{Spec} (Bq/g)	Enrichment Factor
PURE	Dry	-	0.02%	-	-
PURE	Suspension	-	0.01%	-	-
1% TOA	Dry	95%	0.29%	9.4E+04	328
1% TOA 10% TBP	Dry	79%	0.96%	7.1E+04	82
1% TOA 10% TBP	Dry	60%	0.58%	1.1E+05	103
1% 2BISEPO	Dry	28%	0.17%	9.2E+04	162
1% 2BISEPO	Suspension	77%	0.90%	1.0E+05	86

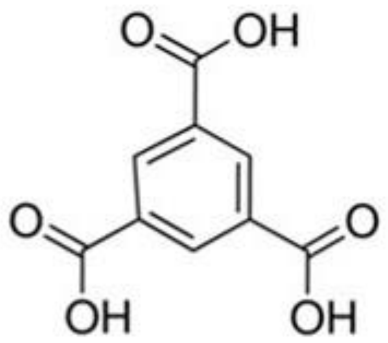
- Selective extraction of ^{99}Mo has been accomplished

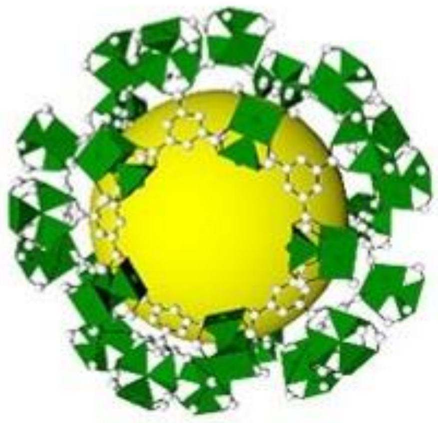
A new idea: the case of ^{51}Cr in metal-organic framework (MOF)

- ^{51}Cr is a useful red cell and platelet label; also used for spleen function studies and blood volume determination
  - EC decay to stable ^{51}V, $T_{1/2}$=27.7 d, main decay line: 320 keV
  - Suitable for most red cell survival and recovery studies
  - Produced in reactors by neutron activation

- The idea: irradiation of radiation-resistant chromium metal-organic frameworks (MOFs) for the production of ^{51}Cr via the recoil effect

- Will likely be tested at MONNET this year





MIL-100 (Cr)

Summary

- New electron beamline for photonuclear reaction studies at GELINA
  - Commissioning report approved by BelV in 2022: green light for experiments!
  - Still a lot to learn on machine operation far from normal regime
  - Hopefully, a new research programme is opening at the JRC and can become part of the Open Access programme

- ir-NANO project
  - Target preparation with high molybdenum purity, development of extraction procedure and selective extraction of ^{99}Mo
  - Alternative route via ^{100}Mo(γ,n) can be investigated at new GELINA beamline in the future
  - Upcoming test of ^{51}Cr production with MOFs
  - Further optimisation of irradiations and extraction procedure, and scaling up

Thank you!

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