SANDA WP2/Task 2.2 report

Alberto Mengoni
on behalf of the Task 2.2 partners: ENEA, CIEMAT, JRC-Geel, Uni Lodz, IRSN

SANDA WP2/Task 2.2 definition

Task 2.2: Neutron capture cross sections
*Task coordinator: **ENEA**, partners: **CIEMAT, JRC, ULODZ, IRSN***

Subtask 2.2.1. Capture measurements of fissile isotopes.
CIEMAT, ULODZ and JRC will perform various cross section measurements at GELINA and n_TOF on the high priority reactions ^{239}Pu(n,g) and ^{239}Pu(n,f). The methodology developed within CHANDA for the absolute measurement of the ^{235}U alfa ratio will be applied to the ^{239}Pu case. A new ionization chamber built by ULODZ will be tested in a ^{239}Pu(n,f) measurement at JRC, which also deliver the ^{239}Pu samples. The combined measurement of the ^{239}Pu(n,g) and ^{239}Pu(n,f) cross sections will be carried out at CERN with the use of the Total Absorption Calorimeter.

Subtask 2.2.2. Capture measurement of stable isotopes.
ENEA will measure the ^{92,94,95}Mo(n,g) cross sections at GELINA and at the n_TOF facility with the high performance total energy detectors developed during the CHANDA project. The impact of the new evaluated nuclear data and their uncertainties will be verified in criticality safety and reactor applications at IRSN as end-user. The data will be part of an evaluation done in WP4 by IRSN.

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[SANDA] template WP18-20 v20180201

Milestones
M.2.11 “Measurement of the ^{239}Pu(n,g) at n_TOF”; M36
M.2.12 “Measurement of the Mo isotopes at GELINA and n_TOF”; M34

Deliverable: 2.3
Report on the ^{239}Pu(n,g), ^{94,95,96}Mo(n,g) cross measurements at n_TOF and GELINA
when: month 40

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$^{239}Pu(n,\gamma)$ neutron capture cross section and α ratio

- proposal for measurement at n_TOF submitted and approved by the CERN Research Board (December 2020)
- sample preparation procedure agreed between CERN and JRC-Geel

- The ionization chamber that is used for these measurements will be tested at JRC-Geel during 2021
- Measurements to be performed at n_TOF during 2022

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$^{94,95,96}Mo(n,\gamma)$ neutron capture cross section

- proposal for measurement at n_TOF submitted and approved by the CERN Research Board (December 2020)
- sample orders issued to Neonest AB, Sweden. Expected delivery Q1-2021

- Total cross section measurements to be performed at JRC-Geel (GELINA) during 2021
- Measurements to be performed at n_TOF during 2022

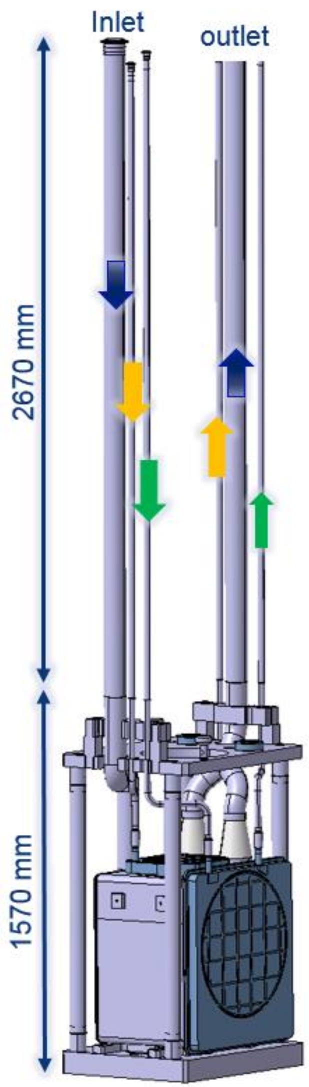
SANDA WP2/Task 2.2 report conclusion

At present, there is **limited delay (~4 months)** in the activity
since the schedule of the CERN accelerator complex
has not changed

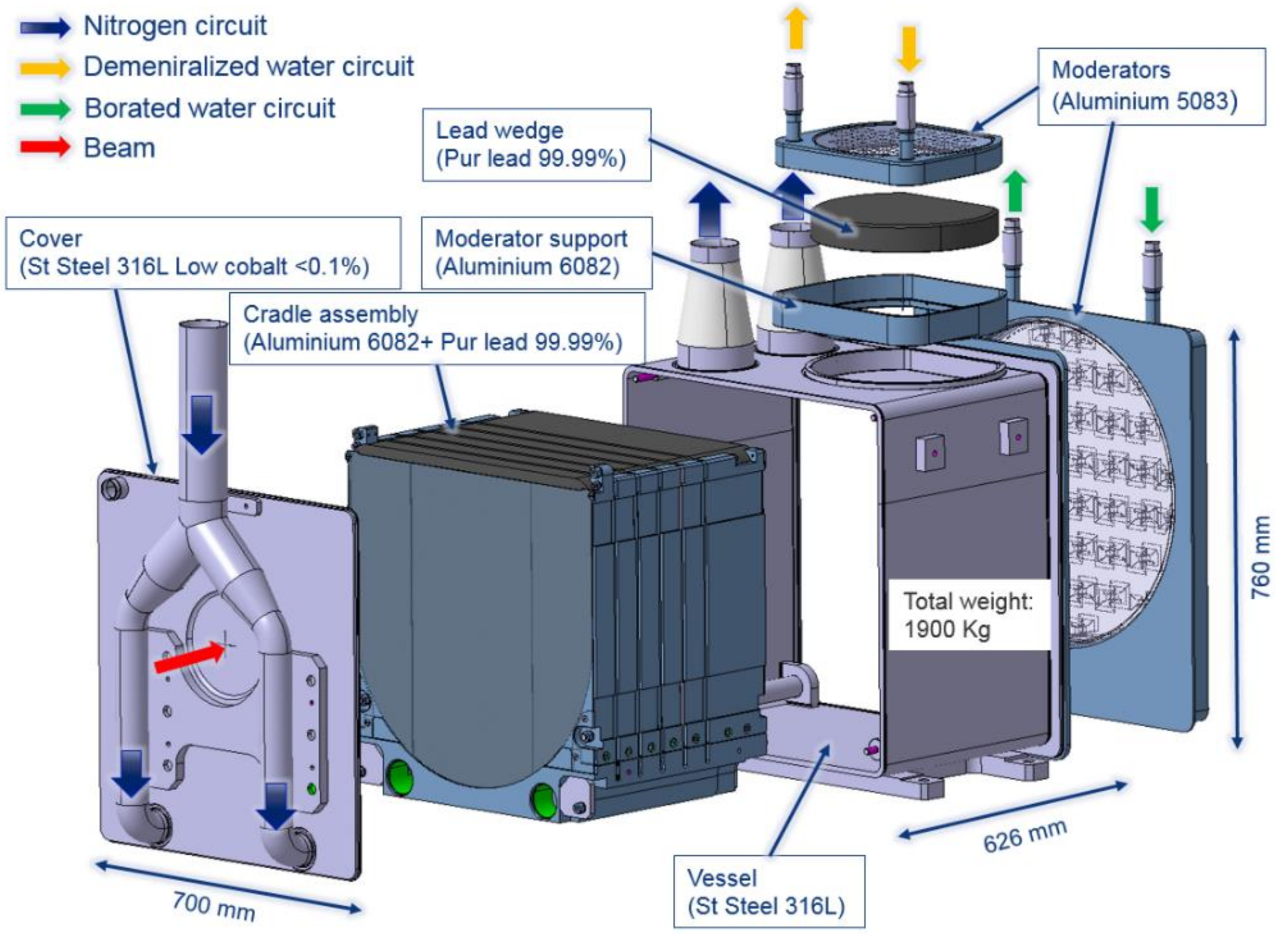
However...

Risks: mainly due to COVID
@ CERN: associated to the successful commissioning
and operation of the new target station
@JRC-Geel: for the production of the Pu-239 samples

n_TOF target #3

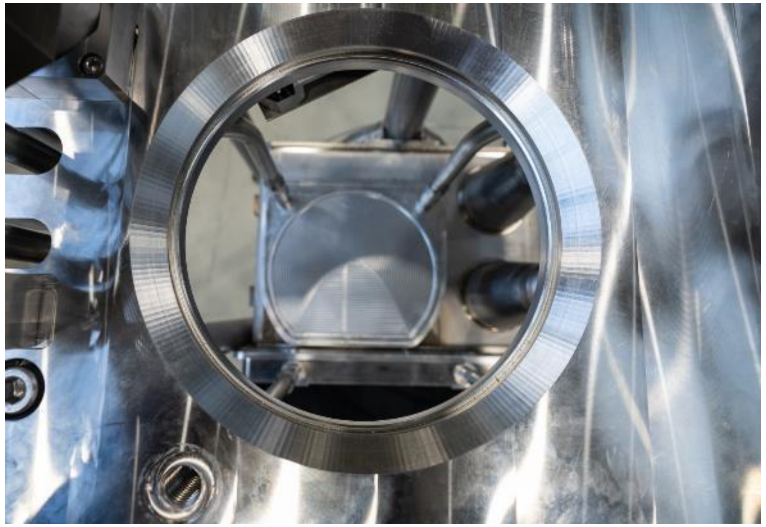


Production completed



courtesy of Oliver Aberle, CERN

n_TOF target #3 – team effort



courtesy of Oliver Aberle, CERN

n_TOF target #3 – team effort: Thank you!

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Infos from CIEMAT >>

Infos from ENEA >>

Infos from IRSN >>

Measurement of $^{239}Pu(n,\gamma)$ and α–ratio at EAR1 with TAC + fission detectors

V. Alcayne[1], J. Andrzejewski[2], M. Caamaño[4], F. Calviño[5], D. Cano-Ott[1], C. Domingo[6], I. Durán[3], B. Fernández[4], A. Gawlik[2], E. Gónzalez-Romero[1], C. Guerrero[7], J. Heyse[3], T. Martínez[1], E. Mendoza[1], J. Perkowski[2], A. Plompen[7], J.M. Quesada[7], P. Schillebeeckx[3], G. Sibbens[3], A. Tarifeño[5]

[1]CIEMAT, Spain
[2]University of Lodz, Poland
[7]JRC Geel
+
[4]U. Santiago de Compostela
[5]U. Politécnica de Cataluña
[6]IFIC – CSIC
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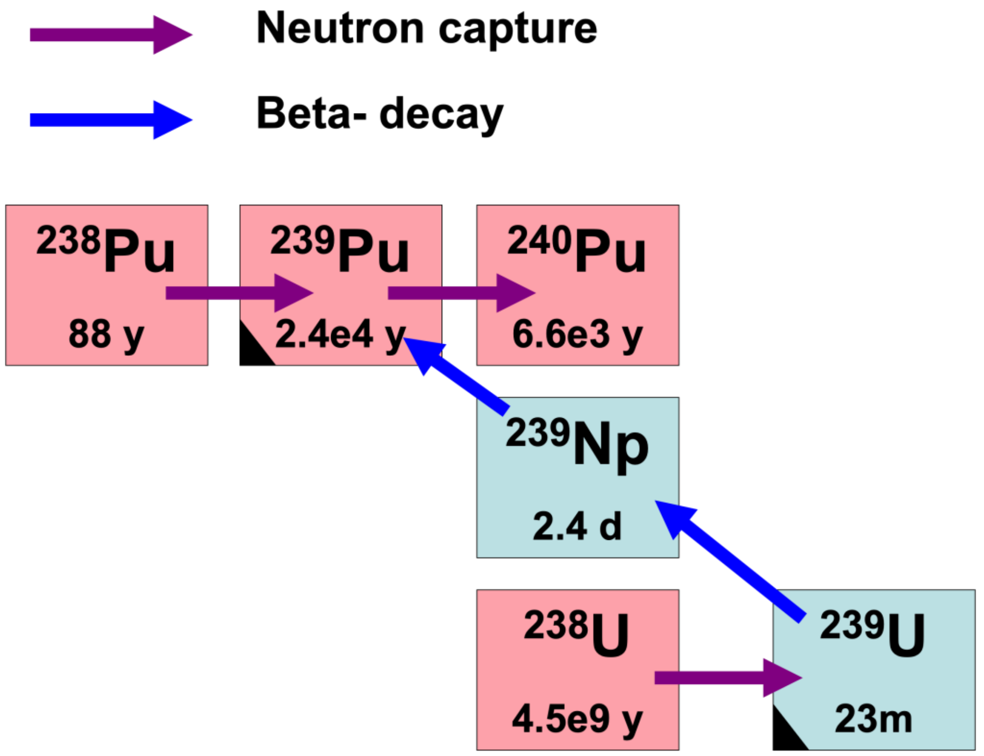
co-funded by the EU H2020 programme

^{239}Pu production in LWR

Standard fresh nuclear fuel for thermal reactors has a 5% ^{235}U and a 95% ^{238}U. ^{239}Pu is produced during the reactor operation mainly by neutron captures in ^{238}U + decays of ^{239}U and ^{239}Np. The ^{239}Pu is fissile and thus its neutron induced fission contributes to the power.



co-funded by the EU H2020 programme

Motivation

^{239}Pu is a very important isotope in actual Light Water Reactors (LWR), contributing up to **30% of the energy produced**.

After ^{238}U and ^{235}U, **^{239}Pu is the most abundant actinide in the spent nuclear fuel** (SNF).T The SNF contains ~1% mass contribution of ^{239}Pu (60 GWd/tU). On average, ~700 kg are produced every 4.5 years of LWR operation. The uncertainty in this mass is dominated by nuclear data (*A. Villacorta, PhD thesis*).

^{239}Pu is considered as a **major component of the nuclear waste** and also as a valuable resource. Pu-loaded fuel burning:

- MOX fuel is used in LWRs (France, …)
- Use of MOX and highly enriched fuels in ^{239}Pu is planned for Fast Reactors (Na-cooled, Pb-cooled, Gas-cooled) and some Small Modular Reactor designs.

^{239}Pu cross section data **are necessary** for improving the simulation capabilities for actual reactors (higher burnups, life extension, burnup credit). For these reasons, more accurate ^{239}Pu capture and fission cross section data are needed and those measurements have been listed in the NEA/OCDE **High Priority Request List**.

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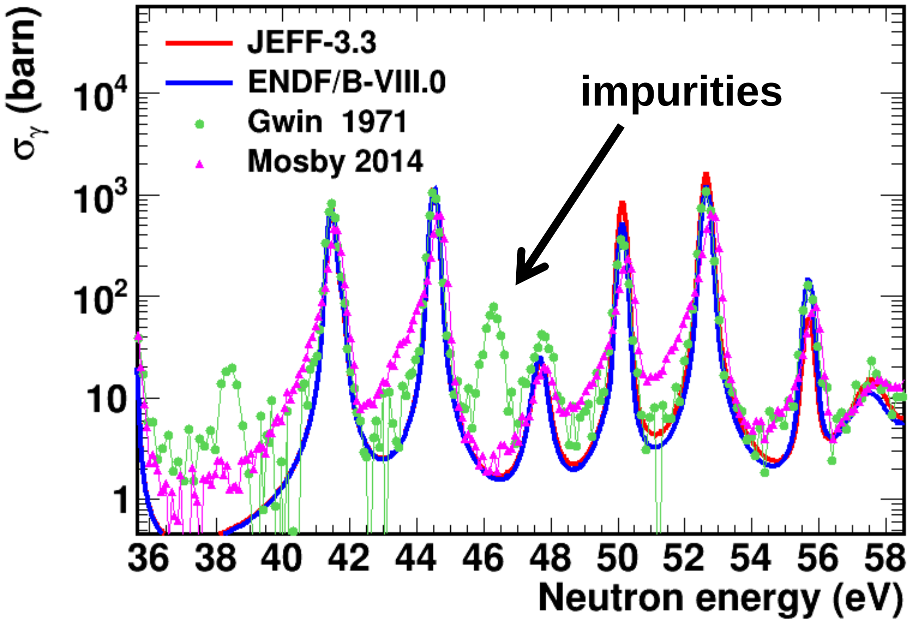
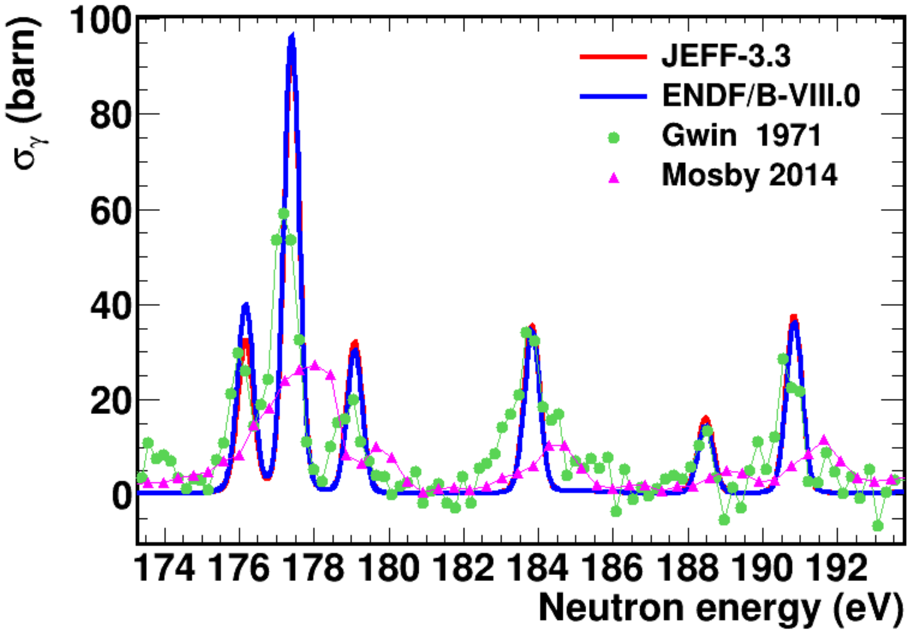
co-funded by the EU H2020 programme

Status of the $^{239}Pu(n,\gamma)$ cross section data

Two *high resolution resolution* capture data sets in EXFOR:

- Gwin et al., Nucl. Sci. Eng. **45**, 25 (1971) → (0.02 eV to 30 keV)
- S. Mosby et al., Phys. Rev. C 97, 041601 (2018) → (10 eV and 1.3 MeV, 3 papers, same measurement)

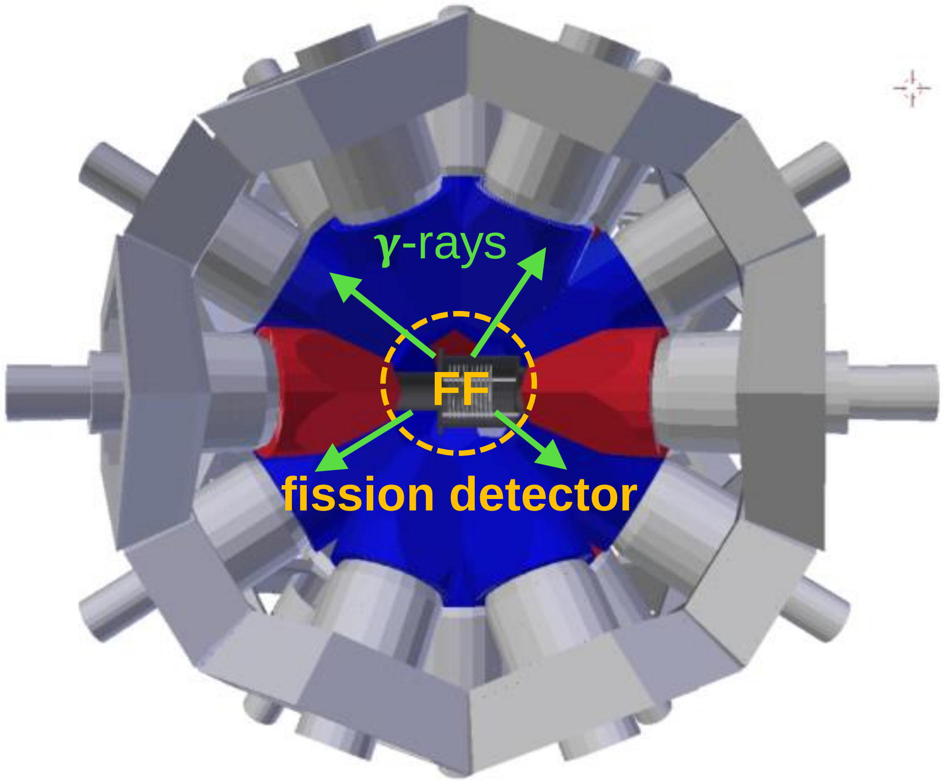




Experimental technique

Fission tagging: $\boldsymbol{\gamma}$-rays in coincidence (fission background) and anticoincidence (capture signal) with the fission detector. (*J. Balibrea et al.*, PRC 102, 2020)

$$Y_\gamma = \frac{c_{aco,\gamma} - \frac{1-\epsilon_f^*(E_n)}{\epsilon_f^*(E_n)} c_{tag} - c_{oth,\gamma}}{\epsilon_\gamma \phi_N}$$



c_{tot} = counts in the TAC.
c_{tag} = counts in the TAC in coincidence. with the ionisation chambers.
c_{oth}= background in the TAC.
ϵ_f^* = fission tagging efficiency.
ϵ_γ = capture detection efficiency.

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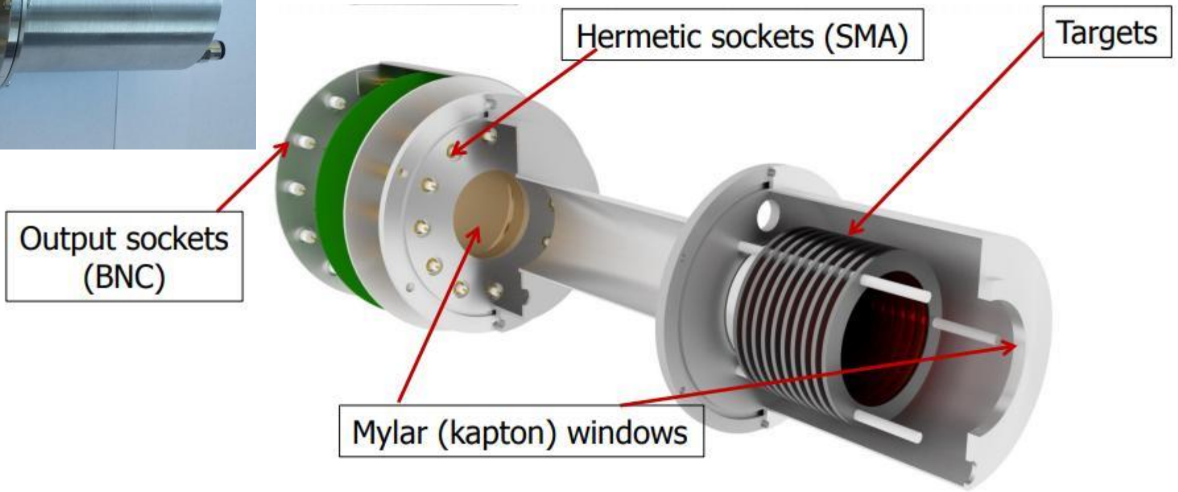
The new fission chamber

A new multi target fission chamber has been built taking into account the following important characteristics:

1. **Low mass** intercepting the neutron beam, to minimize the background in the TAC due to captures and elastically scattered neutrons.
2. Good **discrimination** between **alphas (2 MBq/mg)** and **fission fragments (5 mm gap)**.
3. Small **pile-up** effects.



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The samples will be provided by JRC-Geel

I. Ten thin ^{239}Pu **samples will be manufactured at JRC-Geel** with a total mass of ~**10 mg (1 mg/sample)**.

The samples will be placed inside the fission detector, in the center of the TAC. The limitations come from the signal-to-background ratio.

Overall uncertainties: ~3% below 100 eV and 4-6% between 100 eV and 1keV.

II. A thick sample, in order to extend the measurement to higher neutron energies.

This sample will be measured **without fission detector**. With **~80 mg** we could extend the measurement **up to 10 keV**.

Overall uncertainties: 3-4% between 100 eV and 10 keV.

Summary and Conclusions

INTC approved the measurement the α–ratio, (the fission) and the capture cross sections at the EAR1, using the n_TOF **TAC and ionization chambers** developed at the University of Lodz.

The ^{239}Pu **samples will be manufactured at JRC-Geel:**

- **10 thin samples** (**1 mg each**). **Fabrication started.**
- **One sample of 50-100 mg**. **Needs to be defined (meeting with JRC pending)**

One CIEMAT PhD student (Adrián Sánchez) has started to work on this topic.

Pending actions: arrangement of the transport and preparation of the documents for CERN (safety, radioprotection).

At present, there is no delay in the activity since the schedule of the CERN accelerator complex has not changed.

Risks: mainly due to COVID, associated to the operation of CERN, the successful commissioning & operation of the new target and the production of the Pu-239 samples.

SANDA WP2/Task 2.2 report

Infos from CIEMAT >>

Infos from ENEA >>

Infos from IRSN >>

EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

Measurement of ^{94,95,96}Mo(n,γ) relevant to Astrophysics and Nuclear Technology

September 10, 2020

M. Busso[1,2], D. M. Castelluccio[1,3], P. Console Camprini[1,3], N. Colonna[1], S. Cristallo[1,4], C. Domingo-pardo[5], A. Guglielmelli[1,3], J. Heyse[6], S. Kopecky[6], C. Lederer-Woods[7], A. Manna[1,8], C. Massimi[1,8], P. Mastinu[1], A. Mengoni[1,3], P.M. Milazzo[1], R. Mucciola[1,2], C. Paradela-Dobarro[6], T. Rauscher[9], F. Rocchi[3], P. Schillebeeckx[6], N. Sosnin[7], N. Terranova[1,3], G. Vannini[1] and the n_TOF Collaboration[10]

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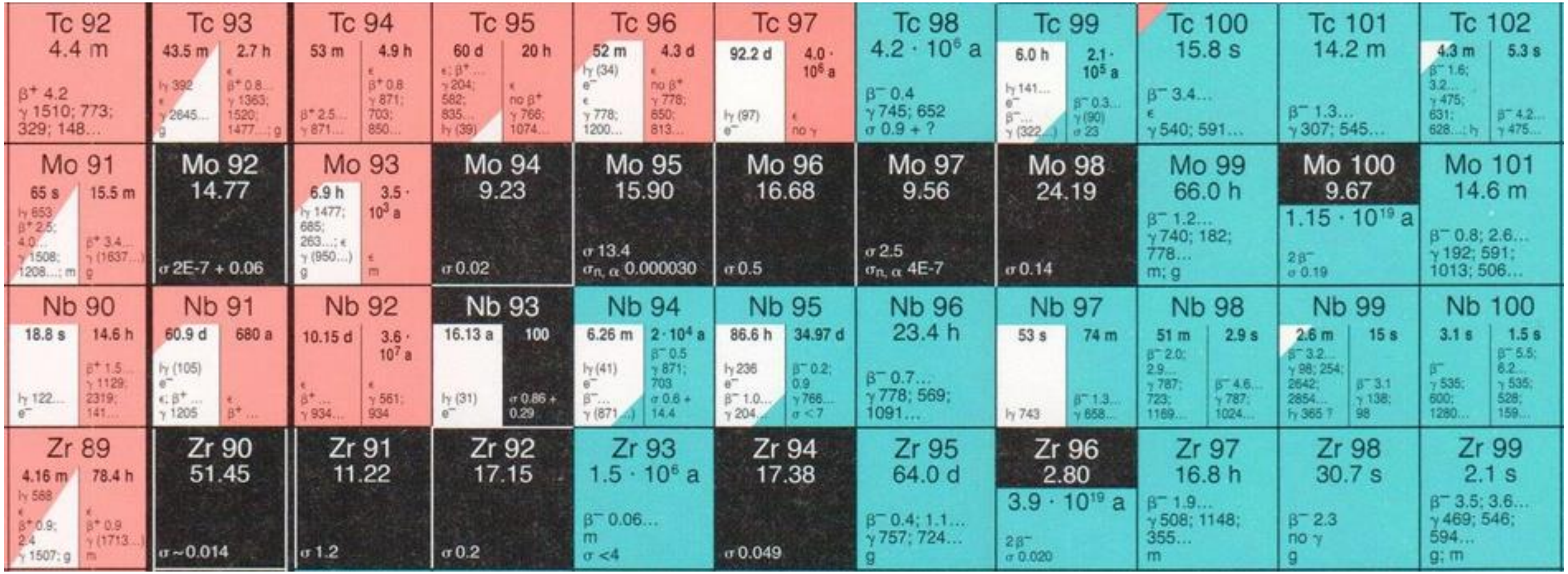
[9] *Department of Physics, University of Basel, Basel, Switzerland*
[10] *www.cern.ch/n_TOF*

Spokesperson: Cristian Massimi (cristian.massimi@bo.infn.it)
Technical coordinator: Olivier Aberle (olivier.aberle@cern.ch)

Courtesy of
Cristian Massimi
University of Bologna & INFN, Sezione di Bologna

Why $^{94,95,96}Mo$?

Molybdenum is relevant for **nuclear astrophysics** and **nuclear technology** and presently **known with large uncertainties**.



Why ^{94,95,96}Mo?

Molybdenum is relevant for **nuclear astrophysics** and **nuclear technology** and presently **known with large uncertainties**.

p process

s process & r process

s-only isotope

r-only isotope

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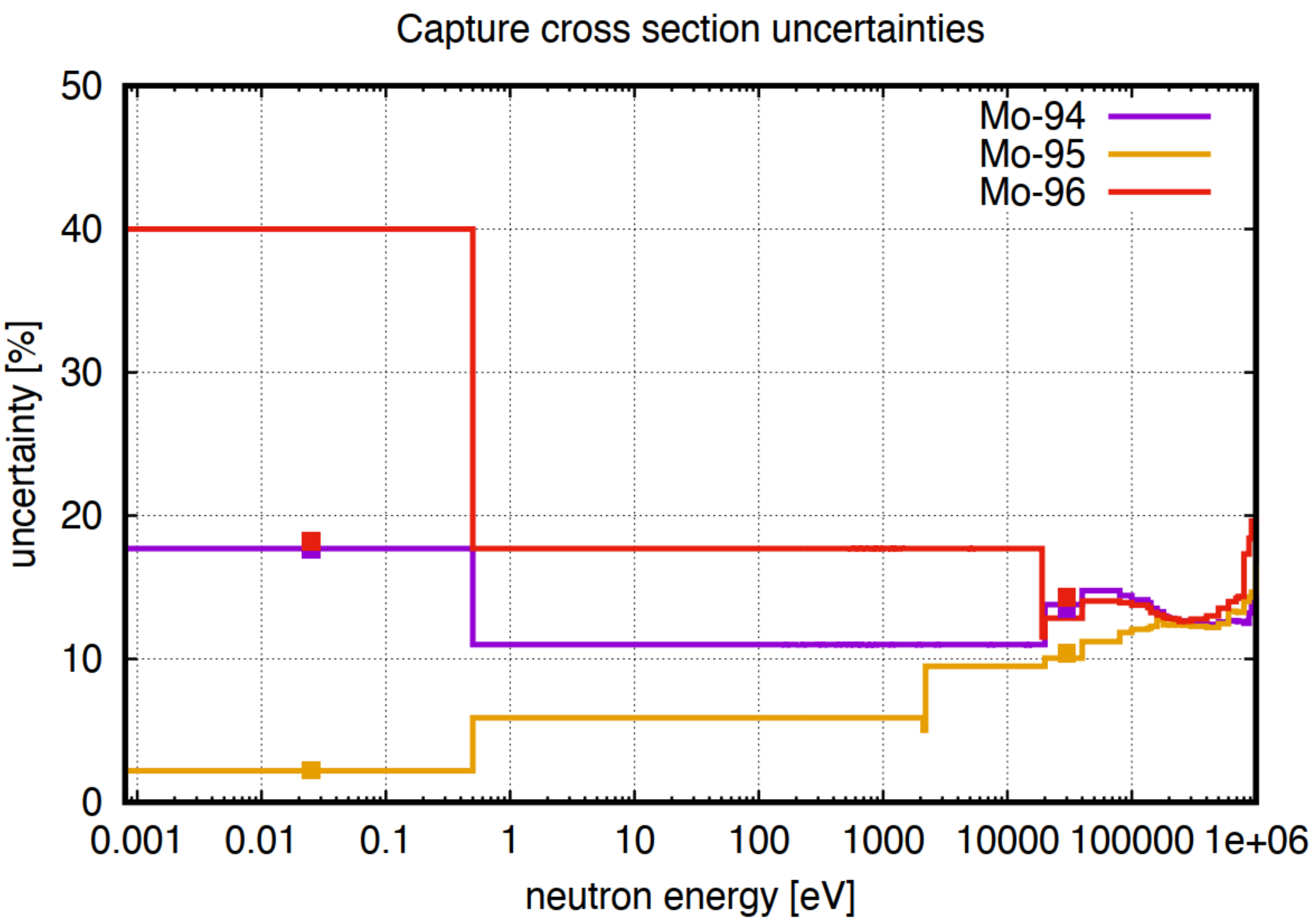
Why $^{94,95,96}Mo$?

nuclear technology

- **fission products** in nuclear reactors
- **Mo-based alloys** used to produce **nuclear fuel** for research, naval and space reactors
- ^{93}Mo ($T_{1/2}$ = 4000 yr) present in spent nuclear fuel (radiotoxicity)
- **$^{95}Mo(n,\gamma)$** reaction cross section related to **criticality safety studies**, based on a **burnup credit**
- Related to **safety assessments of spent nuclear fuel** transport, storage and final disposal
- handling of spent nuclear fuel in reprocessing facilities

Why ^{94,95,96}Mo?

Scientific relevance + other constraints (uncertainty, sample material, …) led to this proposal, limited to the 3 isotopes.



Data in the literature

Capture data

1. Weigmann and Schmid. **10 eV - 25 keV @ GELINA**, total energy detection principle using a **Moxon-Rae detector** and **natural molybdenum** samples.
2. Musgrove et al. **3 – 90 keV @ ORELA** (40 m) **F_6C_6** liquid scintillators and **enriched samples**.
3. Leinweber et al. **10 eV – 600 eV @ RPI** using **natural samples** and the capture detection system was a **total absorption detector** based **NaI**.

Transmission data

1. Leinweber et al. **10 eV – 2 keV @ RPI.**

Spin and parity

1. Sheets et al. **@ Los Alamos** using **DANCE** for n+^{94,95}Mo.

Proposed measurements

The measurements are proposed at both **EAR1** and **EAR2** measuring stations in order to accurately estimate the neutron capture cross-section for neutron energies **between thermal (25.3 meV) and 100 keV**.

Isotope	Mass [g]	Enrichment [%]	Areal density [atom/b]
^{94}Mo	2.0	98.90	1.81E-3
^{95}Mo	0.1	> 94.3	8.97E-5
	1.9		1.70E-3
^{96}Mo	0.2	> 95.7	1.74E-4
	1.8		1.59E-3

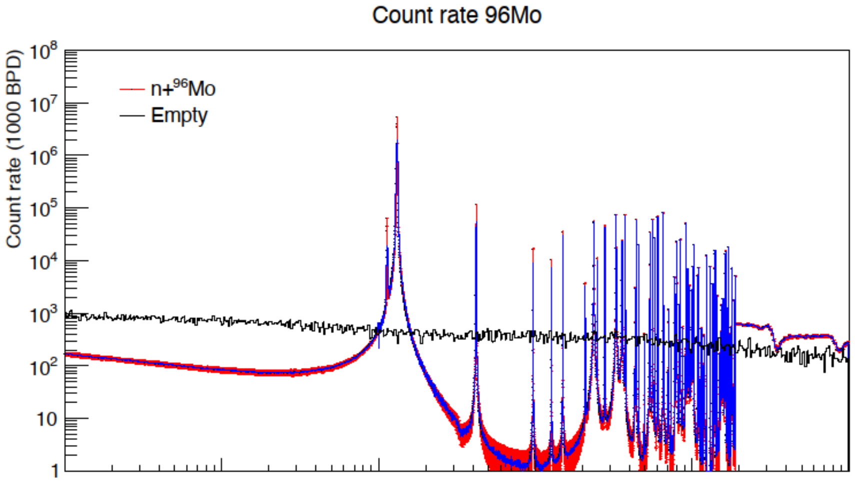
Table 1: Mass and areal density of the three enriched molybdenum samples.

Natural Mo sample will be also used

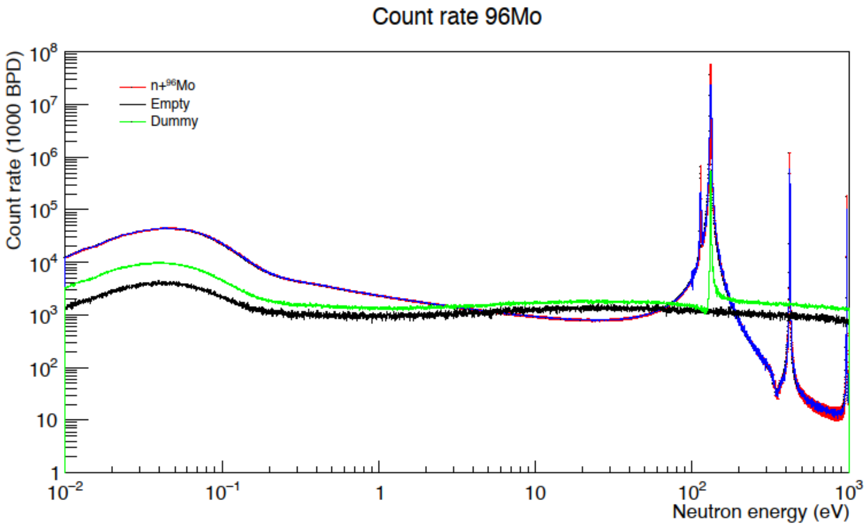
Metallic discs with a 30 mm diameter

Proposed measurements

Using C_6D_6



$20x10^{17}$ POT@ EAR1



$5x10^{17}$ POT@ EAR1

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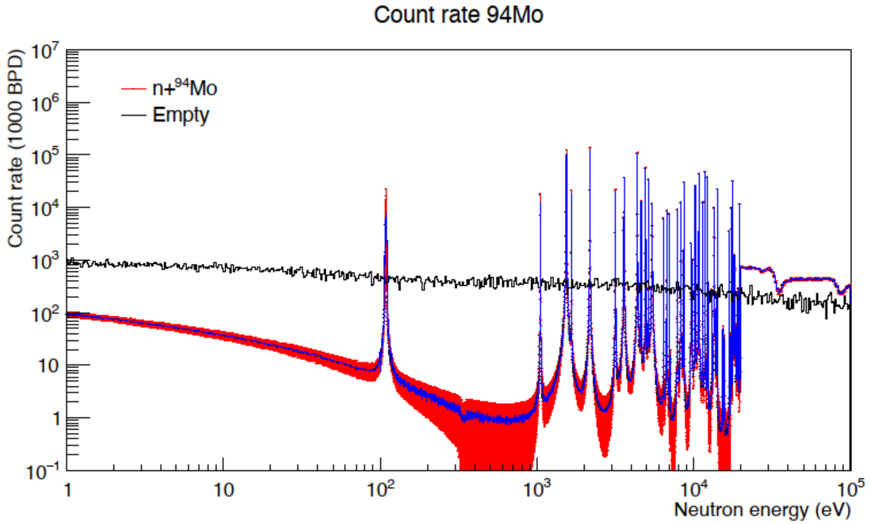
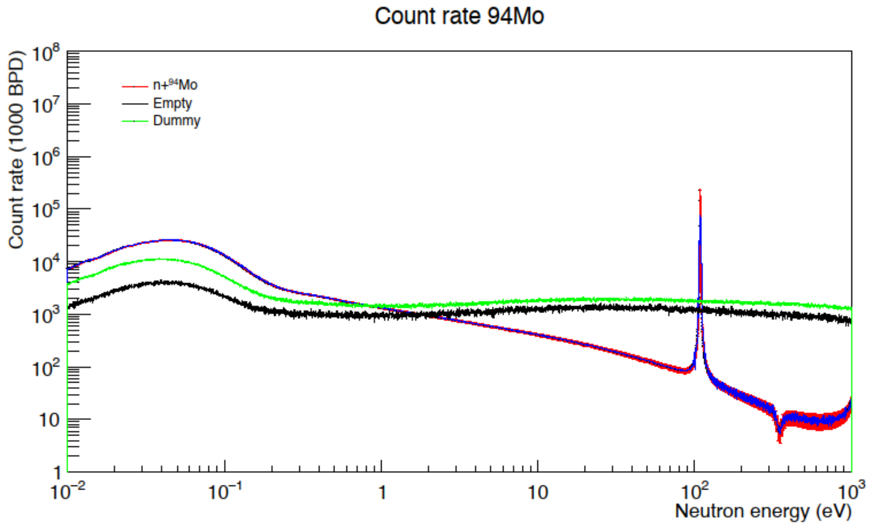
Proposed measurements

Using C_6D_6



$20x10^{17}$ POT@ EAR1



$5x10^{17}$ POT@ EAR1

Conclusions

To improve the status of evaluated data libraries for Mo isotopes and in particular **to improve the quality of the recommended capture cross sections**, a **collaborative effort** has been planned as part of the **SANDA project** supported within the EU Horizon 2020 framework programme:

- capture measurements at n_TOF
- Transmission measurements GELINA

using isotopically enriched Mo metallic samples.

Requested protons: 8×10^{18} protons on target
Experimental Area: EAR1 and EAR2

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Infos from CIEMAT >>

Infos from ENEA >>

Infos from IRSN >>

SANDA GENERAL MEETING
PROGRESS OF IRSN ON WP2&4 WORK PACKAGE
9-11 FEBRUARY 2021

IRSN
PSN-RES/SNC/LN
Pôle Sûreté Nucléaire
Service de Neutronique et des risques de Criticité
Laboratoire de Neutronique

N. LECLAIRE
L. LEAL

ASSESSMENT OF MOLYBDENUM NUCLEAR DATA

- Target: produce Mo evaluation with covariance for RP and cross sections
  - JEFF-33, ENDF/B-VIII.0: covariance for cross sections BUT no covariance for RP
  - JENDL-4.0: no covariance for RP and cross sections
- Resonance parameters retrieved from JEFF-3.1.1, JEFF-3.3, JENDL-4.0, ENDF/B-VII.1 and ENDF/B-VIII.0 evaluations
- Identification of available differential measurements in EXFOR
  - Lack of data for ^{95}Mo
- Use of JENDL-4.0 resonance parameters to initiate the evaluation process
  - Correspondence between spin groups and channel spins addressed
- Transmission and capture measurements of ^{nat}Mo at J-PARC (Japan) on various samples
  - ANNRI experimental device
  - 0.1, 0.5, 2 mm thick for capture and 0.5, 5 mm thick for transmission

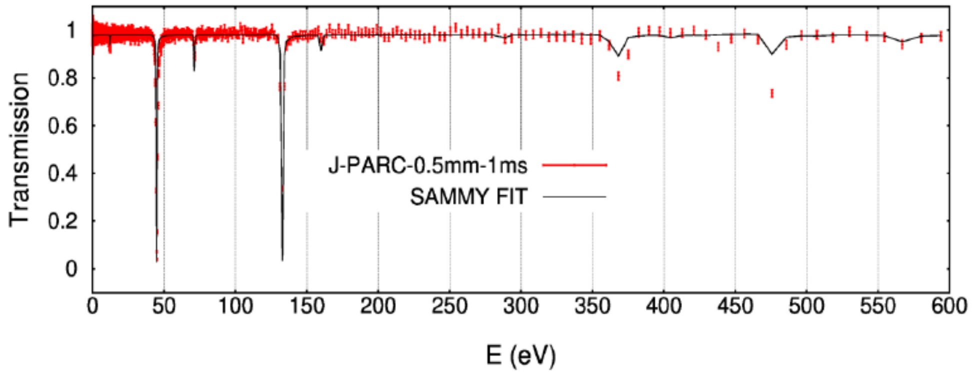
Isotope	Composition (%)	Thermal Cross Section (barns)	Resonance Integral (barns)
^{92}Mo	14.84	0.08±0.02	0.83
^{94}Mo	9.25	0.34±0.02	1.12
^{95}Mo	15.92	13.4±0.3	118±7
^{96}Mo	16.68	0.5±0.3	17±3
^{97}Mo	9.55	2.2±0.2	14.4±3.0
^{98}Mo	24.13	0.130±0.006	6.7±0.3
^{100}Mo	9.63	0.199±0.002	3.76±0.15

Library	Lower limit RR (eV)	Upper limit RR (eV)	Lower limit URR (eV)	Upper limit URR (eV)
JEFF-3.3	0	2141.2	0	206269
ENDF/B-VIII.0	0	2141.2	0	206269
JENDL-4.0	0	2000.0	0	400000

Working package WP2 & 4 – Measurement and assessment of nuclear data

ASSESSMENT OF MOLYBDENUM NUCLEAR DATA

- Fit of experimental data with SAMMY code (R-matrix)
  - Preliminary evaluation: sequential fit
    - Resolved resonance description
      - Use of $\chi 2$ as figure of merit
      - Generation of covariance matrix
      - Updated RP and RP covariance data at each step
  - Use of 0.5 mm sample in 0-600 eV
    - Low data resolution above 350 eV



- n_TOF and GELINA measurements of enriched Mo are planned/underway.

- Additional data: a) RPI transmission data for isotopes (^{95}Mo, ^{96}Mo, ^{98}Mo, ^{100}Mo); b) Transmission and capture data for ^{95}Mo from LANL

➡ **Need of enriched samples**

The END

Thank you!

The Task 2.2 partners: CIEMAT, JRC-Geel, Uni Lodz, IRSN and ENEA