Measurement of the $^{238}Pu(n,\gamma)$ cross-section at EAR2

V. Alcayne[1], D. Cano-Ott[1], E. González-Romero[1], T. Martínez[1], E. Mendoza[1],G. Noguère[2], A. Perez de Rada[1], A. Sánchez-Caballero[1], A. B. Allannavar[3], V. Babiano[4], J. Balibrea-Correa[4], B. Bernardino Gameiro[4], F. Calviño[3], A. Casanovas[3], G. Cortés[3], G. de la Fuente Rosales[4], M. Diakaki[5], C. Domingo-Pardo[4], C. Guerrero[6], M. Kokkoris[5], J. Lerendegui-Marco[4] and the n_TOF Collaboration.

[1] Centro de Investigaciones Energéticas Medioambientales y Tecnológicas (CIEMAT), Spain

[2] CEA DES, Service de Physique des Réacteurs et du Cycle, Cadarache, France

[3] Universitat Politècnica de Catalunya, Spain

[4] Instituto de Fısica Corpuscular, CSIC - Universidad de Valencia, Spain

5 National Technical University of Athens, Greece

6 Universidad de Sevilla, Spain

GOBIERNO DE ESPAÑA MINISTERIO DE CIENCIA, INNOVACIÓN Y UNIVERSIDADES

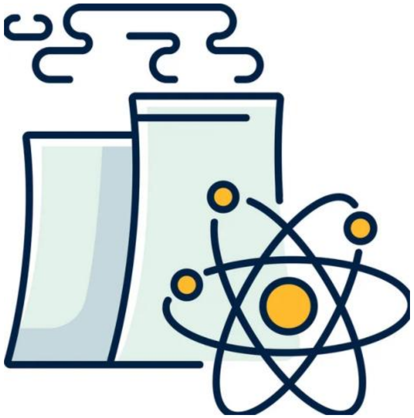
Ciemat Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas

UIN

Outline of the presentation

- Introduction and motivation
- Previous measurements and evaluations of ^{238}Pu
- Measurement at n_TOF EAR2
- Beam time request

Production of actinides in nuclear reactors



mainly alpha decay

mainly beta-decay

Neutron capture

Alpha decay

Beta- decay

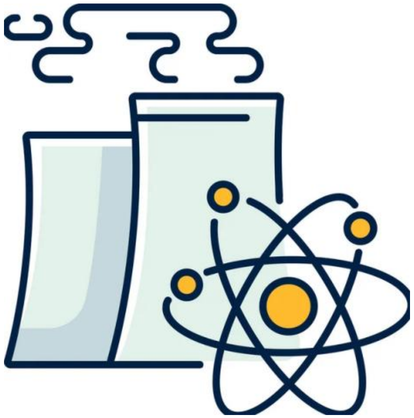
^{235}U
7.0e8 y

^{238}U
4.5e9 y

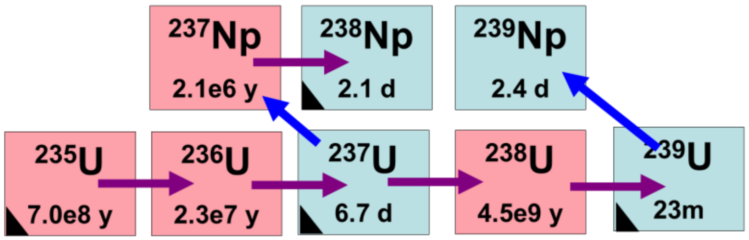
^{239}Pu ← Isotope
6.6e3 y ← $T_{1/2}$

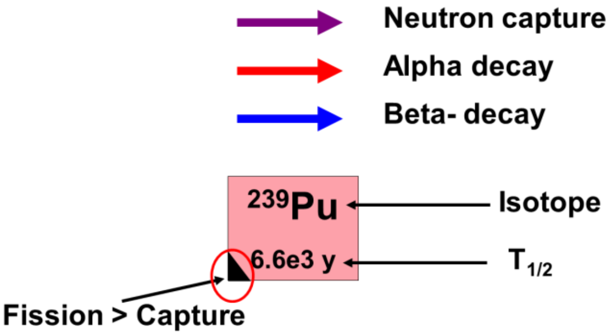
Fission > Capture

Production of actinides in nuclear reactors

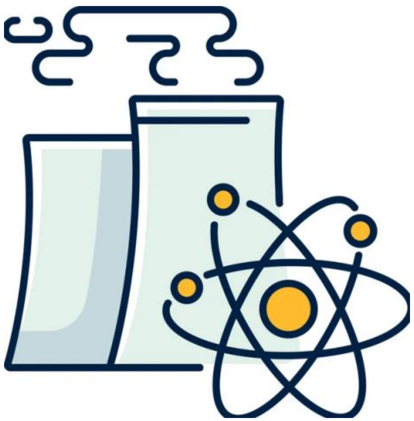


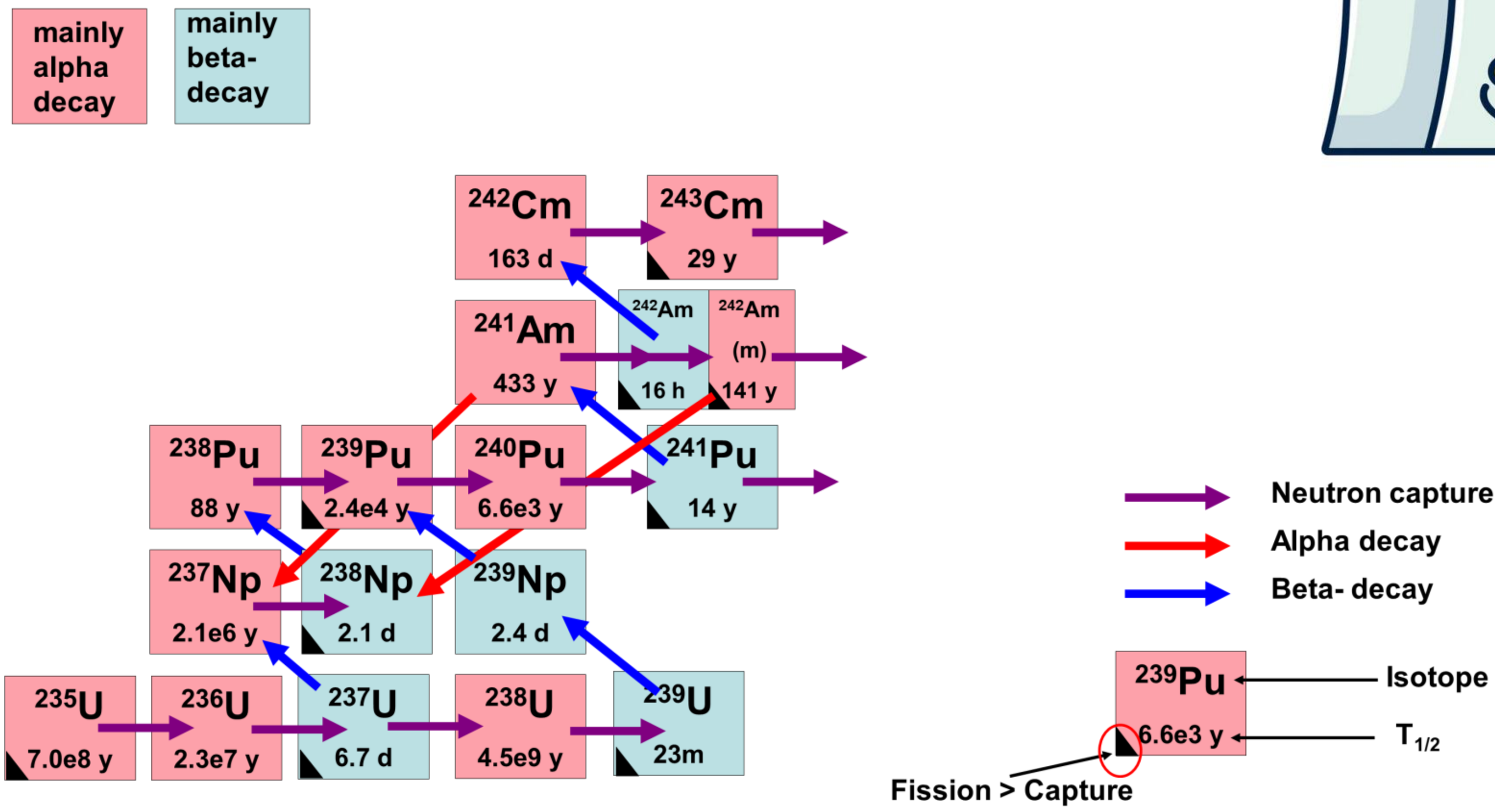
mainly alpha decay	mainly beta-decay



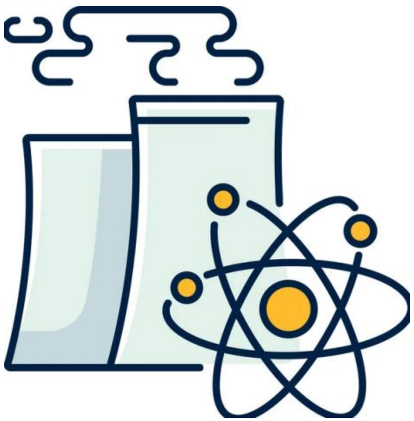


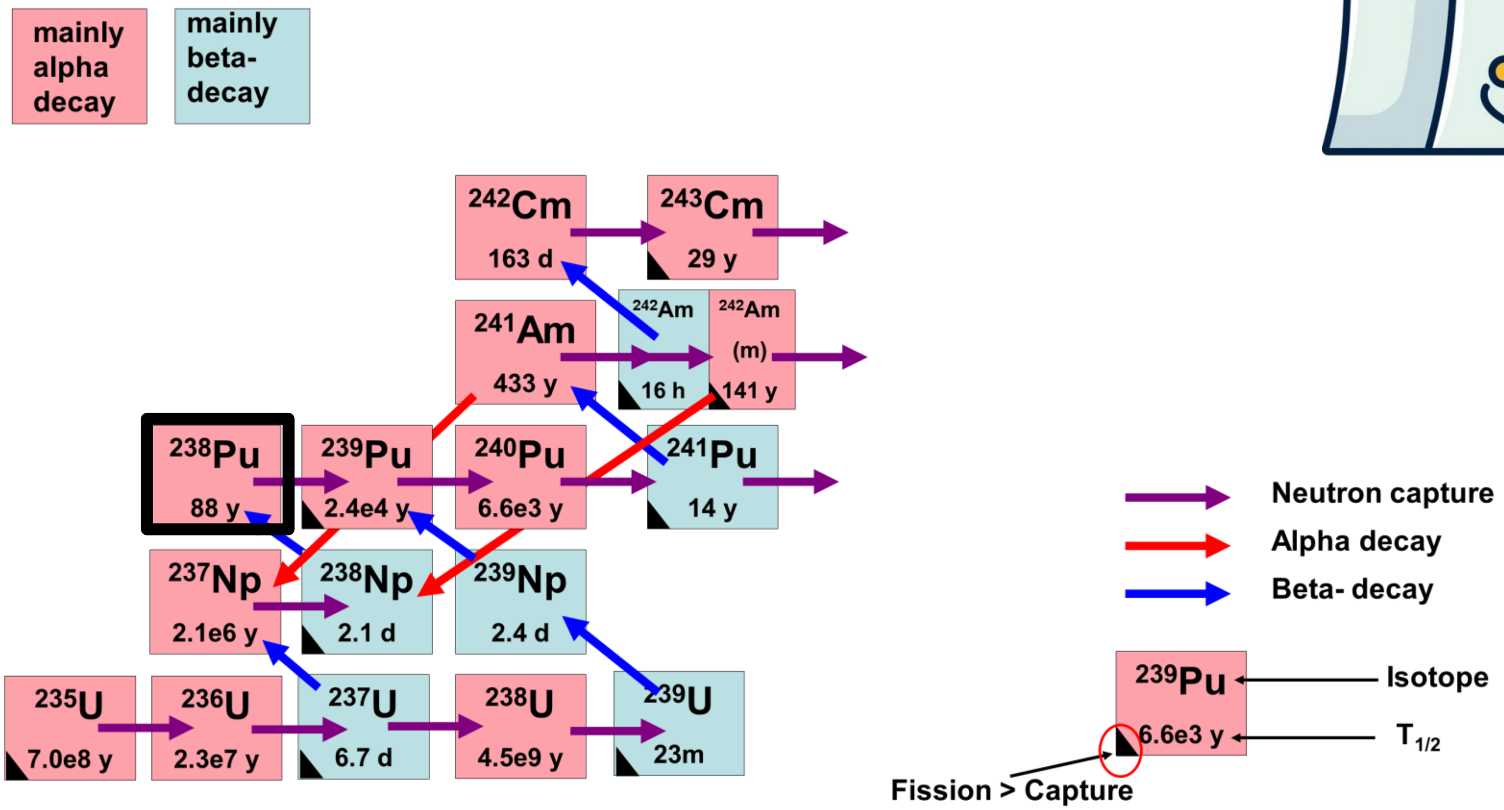
Production of actinides in nuclear reactors



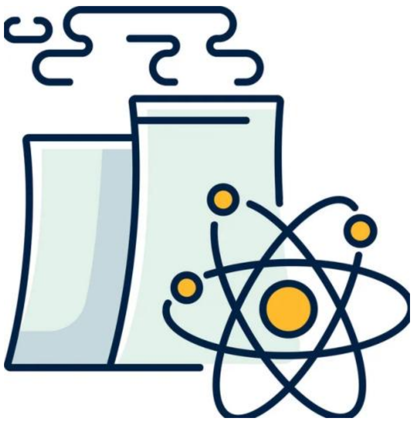


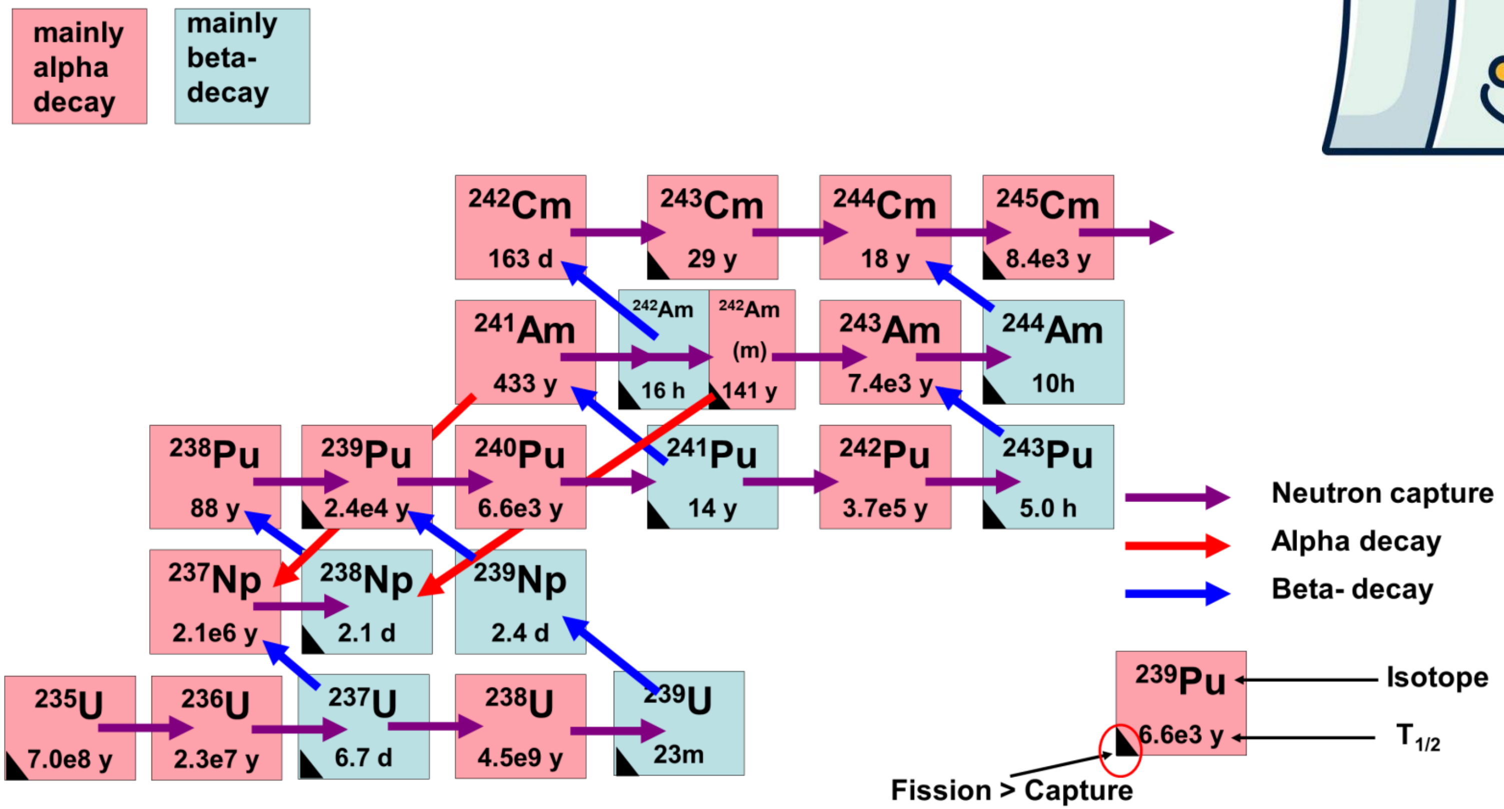
Production of actinides in nuclear reactors





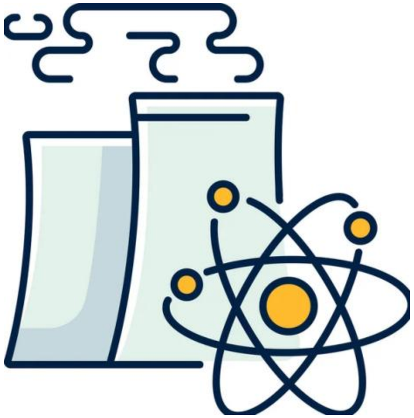
Production of actinides in nuclear reactors

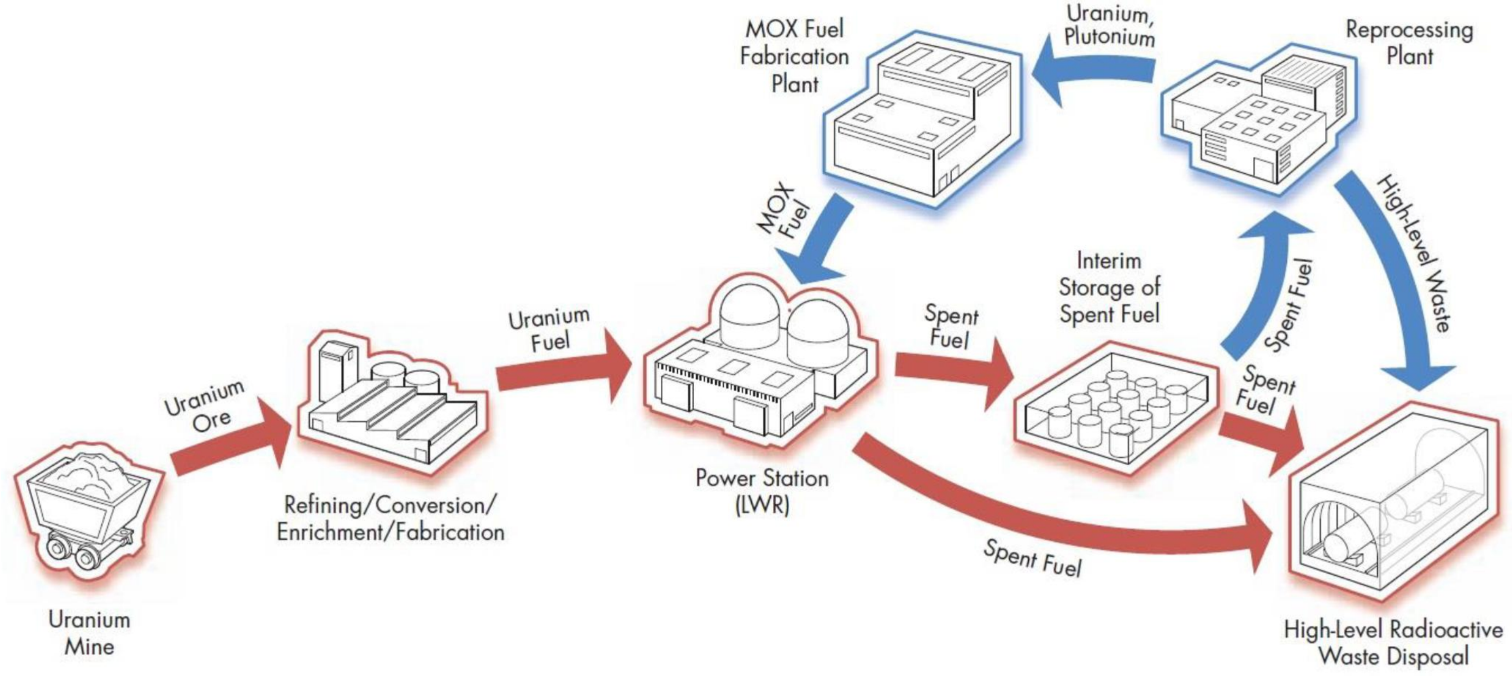




Production of actinides in nuclear reactors

- **All of this isotopes have to be** transmutated, reprocessed or dispose

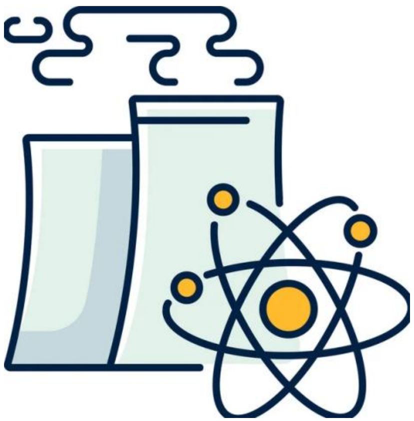


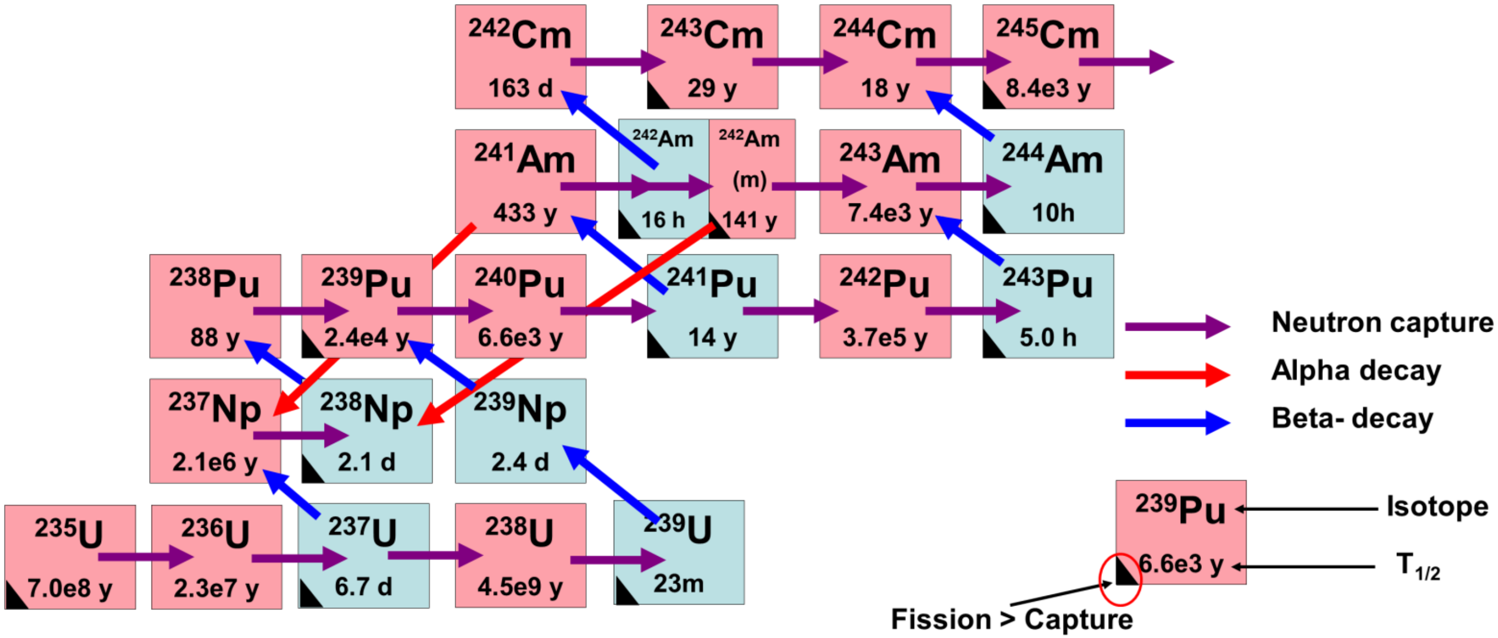


GOBIERNO DE ESPAÑA
MINISTERIO DE CIENCIA, INNOVACIÓN Y UNIVERSIDADES
Ciemat Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas

Production of actinides in nuclear reactors

- **Importance:** transport, transmutation, and storage of spent fuel.

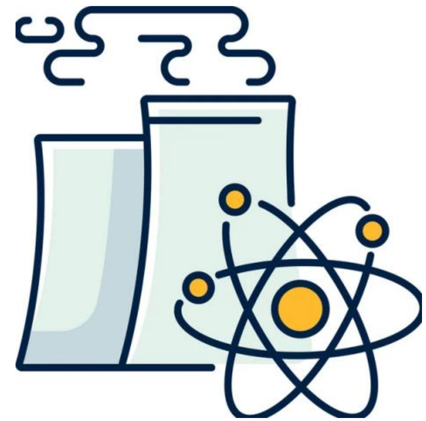


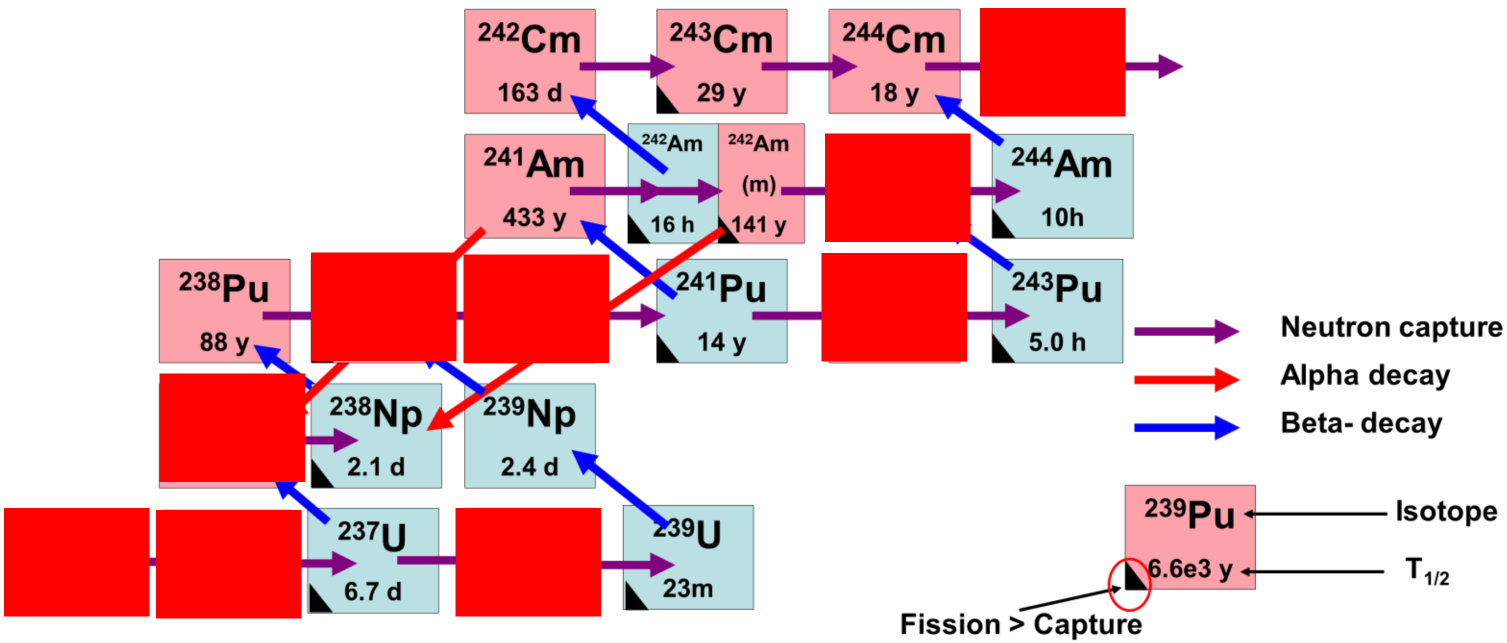


GOBIERNO DE ESPAÑA
MINISTERIO DE CIENCIA, INNOVACIÓN Y UNIVERSIDADES
Ciemat
Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas

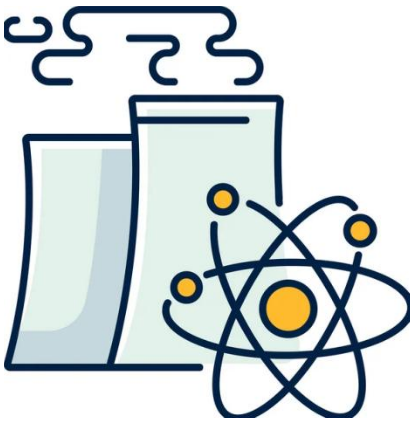
Production of actinides in nuclear reactors

- **Importance:** transport, transmutation, and storage of spent fuel.
- Isotopes with half-lives **longer than 1e3 year** are mainly important in the long term disposal.

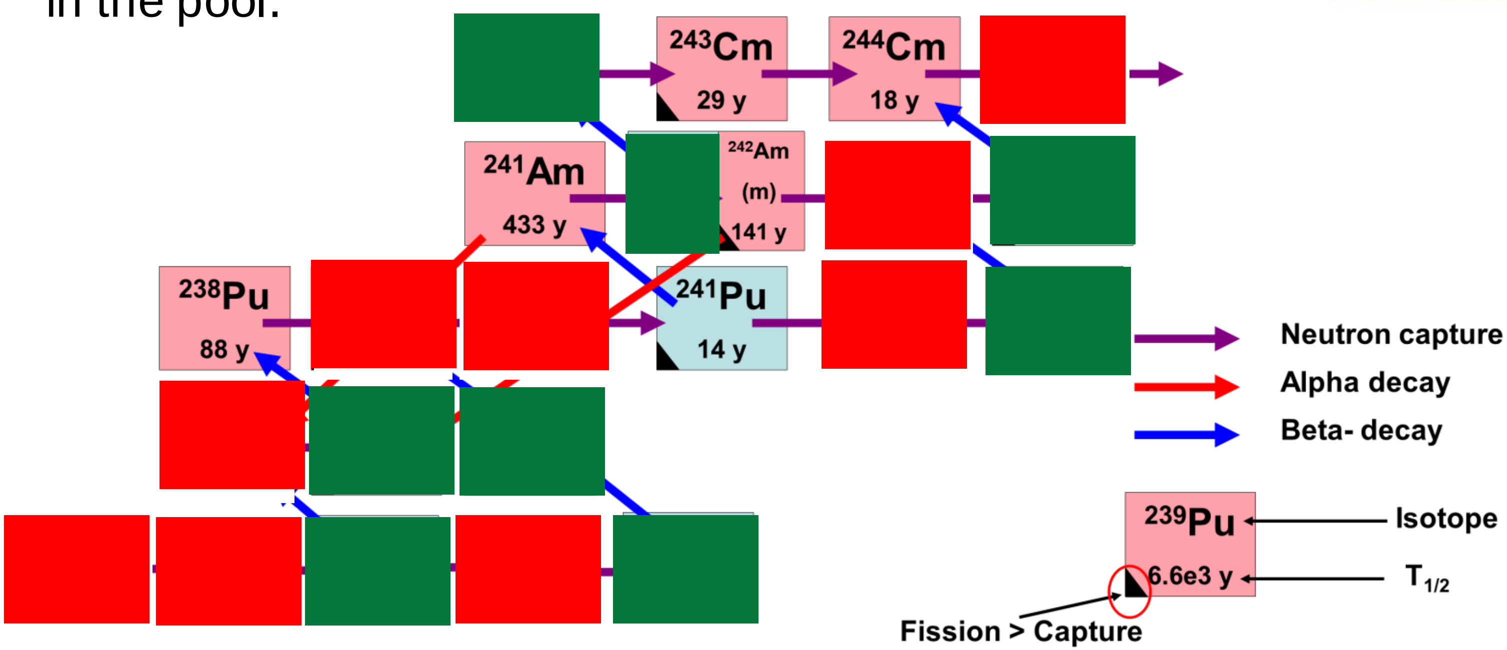




Production of actinides in nuclear reactors

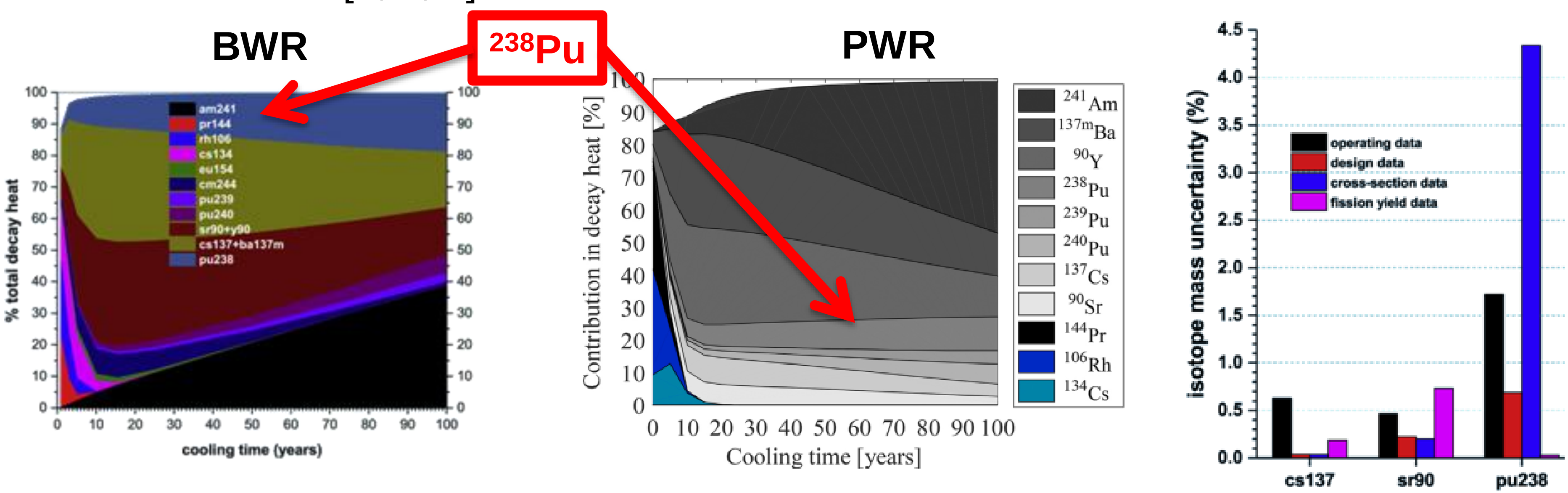


- **Importance:** transport, transmutation, and storage of spent fuel.
- Isotopes with half-lives **longer than 1e3 year** are mainly important in the long term disposal.
- Isotopes with half-lives **shorter than 1 year** decay in the pool.



Decay heat in nuclear reactors

- **Importance:** Key for transport, transmutation, and storage of spent fuel.
- **Main contributors to decay heat:**
  - **0–50 years:** Fission products dominate.
  - **>50 years:** Actinides, including ^{238}Pu, become significant.
- **^{238}Pu impact:**
  - ~10% of decay heat after 10 years of cooling.
  - Main α-emitter between 10 and 10^3 years after shutdown.
  - For BWR: After 15.6 years, ^{238}Pu content uncertainty leads to ~0.4% decay heat uncertainty [7].
- **Research focus:** Propagation of nuclear data uncertainties in decay heat estimation [7, 8, 9].



^{238}Pu for space exploration

- Missions beyond Jupiter require **Radioisotope Thermoelectric Generators (RTGs)** for electrical power.
- **Plutonium-238 (^{238}Pu)** is the ideal isotope:
  - Alpha-emitter with **87.7 years half-life**.
  - Provides sufficient decay heat and low radiation background.
- Example: Curiosity and Perseverance rovers used **4.83 kg of plutonium oxide**.

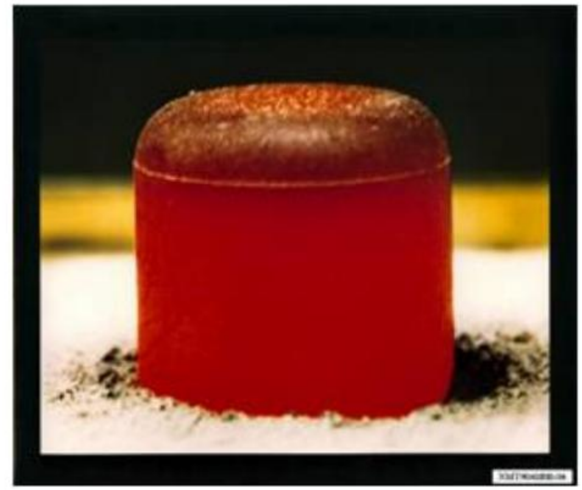
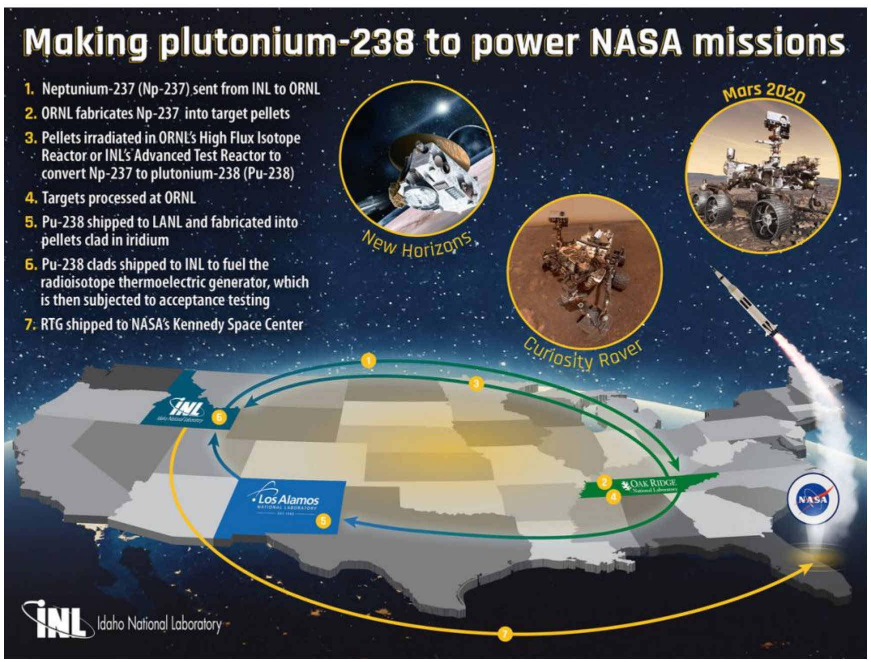


Figure 1. Ceramic fuel pellet before encapsulation

Images obtained from https://inl.gov/mars-2020/

^{238}Pu production

- **Post-Cold war:** Majority of ^{238}Pu produced in Russia.
- **U.S. production restarted:**
  - Idaho & Oak Ridge National Labs.
  - Current rate: ~400 g/year; Goal: 1.5 kg/year [2][3].
- **European efforts:**
  - ESA studies at BR2 reactor (SCK-CEN) [4].
  - Proven feasibility to reduce reliance on non-European partners [5].



ESA Contract No. 4000135477/21/NL/GLC/my

Executive Summary Report (ESR)

Pu-238 Production Feasibility Study

20 June 2022

	AREVA STUDY (EC 2010)		ORANO - SCK CEN - TRACTEBEL (EC 2022)
	Am241	Pu238	Pu238
CAPEX (M€)	120	400	160
OPEX (M€/yr)	12	25	9-17
k€/Watt	100-150	100-150	93-145

EC : Economic Conditions

Optimizing ^{238}Pu production

Preferred neutron energy range:

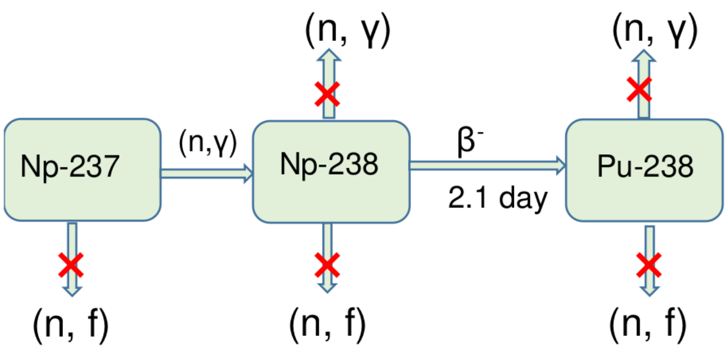
Resonance absorption region of ^{237}Np (1–600 eV) [6].

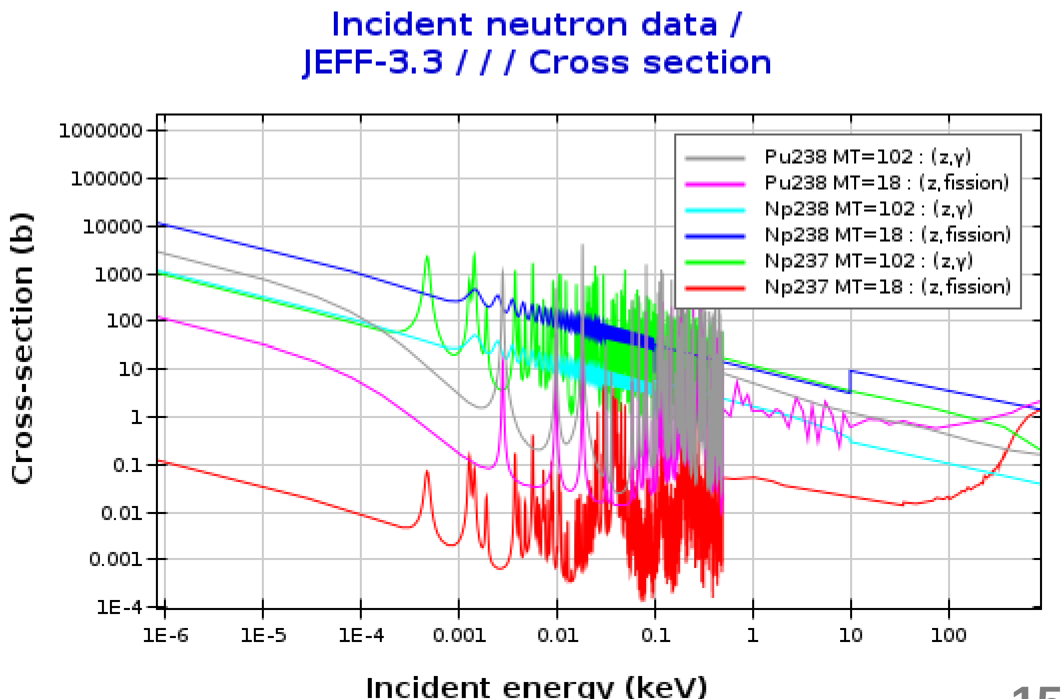
Undesirable reaction:

Neutron capture by ^{238}Pu → Produces ^{239}Pu (fertile, unsafe for reactors) [1].

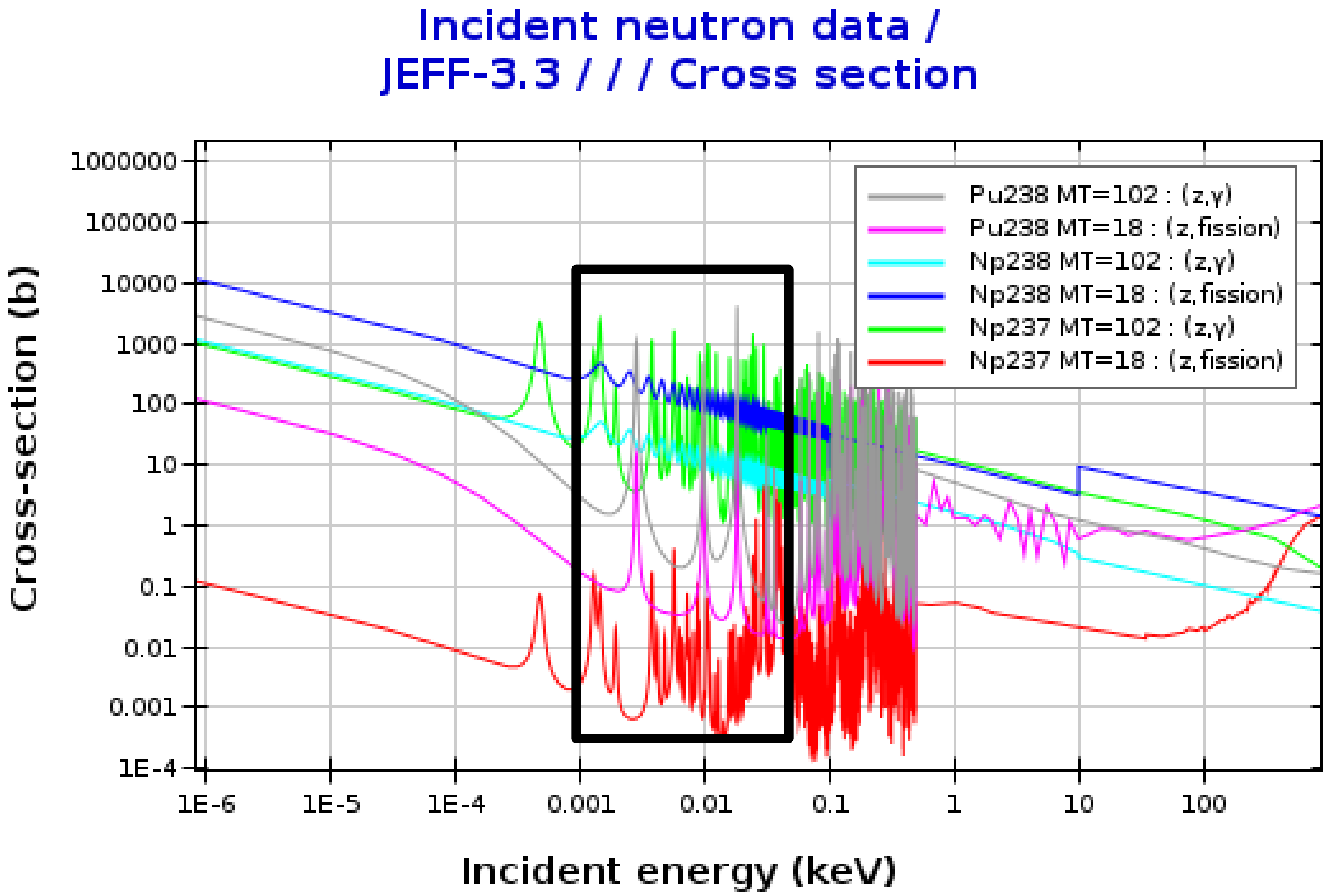
Key Focus:

Deeper understanding of the Resolved Resonance Region (RRR) for efficient ^{238}Pu production.





Optimizing ^{238}Pu production



Np

(

Integral measurements of ^{238}Pu

PROFIL & PROFIL-2 experiments:

- Conducted in the **Phénix fast reactor** [11].
- Irradiated nearly pure isotope samples for precise capture cross-sections and branching ratios [10].
- **Findings for ^{238}Pu:**
  - JEFF-3.1 library overestimates capture cross-section by ~2.5% [12]. The recent evaluations of this isotope has not change considerably.
  - Lack of significant updates due to limited new experimental data.

TABLE IV

C/E Ratios for the Samples Involving the Major Actinides of the Uranium Cycle, After Fluence Scaling

Nuclide	Reaction	Ratio	C/E
^{235}U	Fission	$^{235}U/^{238}U$	1: 1.007 ± 0.020 2A: 1.000 ± 0.002[a] 2B: 0.993 ± 0.003
	Capture	$^{236}U/^{235}U$	1: 1.000 ± 0.001[a] 2A: 1.000 ± 0.001 2B: 1.000 ± 0.001[a]
^{238}U	Capture $(n,2n)$	$^{239}Pu/^{238}U$ $^{237}Np/^{238}U$	1.018 ± 0.002 0.927 ± 0.028
^{238}Pu	Capture	$^{239}Pu/^{238}Pu$	1.024 ± 0.005

The small overestimation of the ^{238}Pu capture rate (~2.5% according to the PROFIL analysis) does not seem confirmed by the EXFOR trends (see Fig. 2). However, the differential data measurements are rather old and dispersed.

Integral measurements of ^{238}Pu

PROFIL & PROFIL-2 experiments:

- Conducted in the **Phénix fast reactor** [11].
- Irradiated nearly pure isotope samples for precise capture cross-sections and branching ratios [10].
- **Findings for ^{238}Pu:**
  - JEFF-3.1 library overestimates capture cross-section by ~2.5% [12]. The recent evaluations of this isotope has not change considerably.
  - Lack of significant updates due to limited new experimental data.

TABLE IV

C/E Ratios for the Samples Involving the Major Actinides of the Uranium Cycle, After Fluence Scaling

Nuclide	Reaction	Ratio	C/E
^{235}U	Fission	$^{235}U/^{238}U$	1: 1.007 ± 0.020 2A: 1.000 ± 0.002[a] 2B: 0.993 ± 0.003
	Capture	$^{236}U/^{235}U$	1: 1.000 ± 0.001[a] 2A: 1.000 ± 0.001 2B: 1.000 ± 0.001[a]
^{238}U	Capture	$^{239}Pu/^{238}U$	1.018 ± 0.002
	$(n,2n)$	$^{237}Np/^{238}U$	0.927 ± 0.028
^{238}Pu	Capture	$^{239}Pu/^{238}Pu$	1.024 ± 0.005

The small overestimation of the ^{238}Pu capture rate (~2.5% according to the PROFIL analysis) does not seem confirmed by the EXFOR trends (see Fig. 2). However, the differential data measurements are rather old and dispersed.

^{238}Pu in MYRRHA reactor

- **MYRRHA overview:**
  - Lead-bismuth-cooled fast spectrum reactor [13].
  - Uses MOX fuel with 30% plutonium.
  - Relies on accurate nuclear data for criticality and safety assessments.
- **Role of ^{238}Pu:**
  - Influences neutron economy via capture (n,γ) and fission (n,f) reactions.
  - Current data libraries show discrepancies [14].
- **Impact:**
  - Affects multiplication factor (k_{eff}) calculations.
  - Urgent need for updated evaluations and new experimental measurements to enhance simulation reliability.

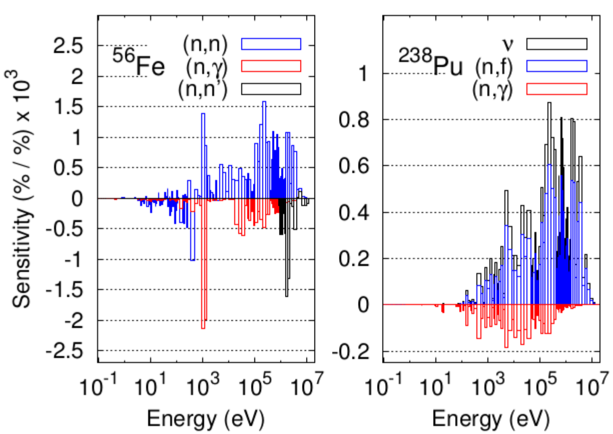
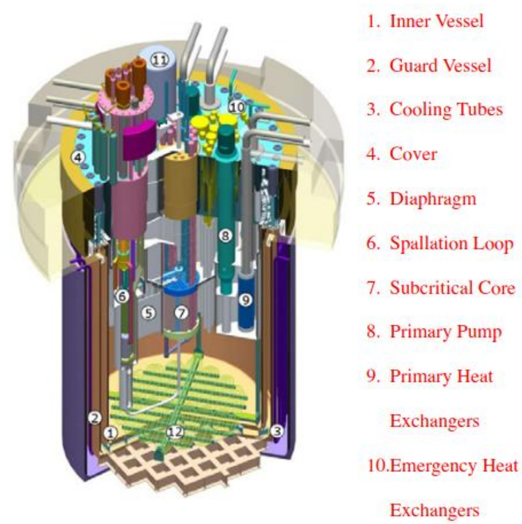
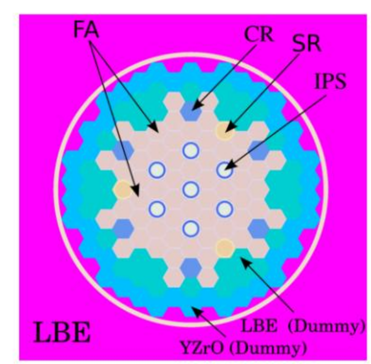


FIG. 2. Sensitivity profiles of the three most important reactions of ^{56}Fe and ^{238}Pu .



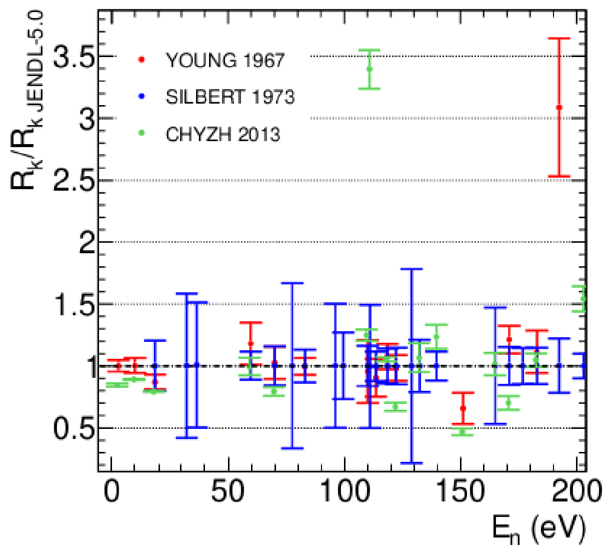


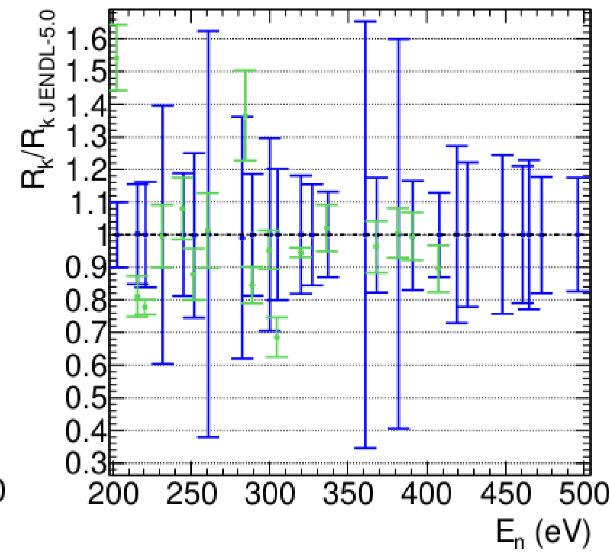
. MYRRHA model for the critical configuration, im-
ıted in KENO-VI (SCALE6.1).

Overview of ^{238}Pu neutron reaction measurements

- **Limited data availability:**
  - Few measurements exist for ^{238}Pu neutron reactions.
- **Fission reaction:**
  - Most studied channel, but additional data is needed there is an entry in HPRL [15–19].
- **Capture cross-Section:**
  - Thermal point, there are two old measurements [20, 21] and a more recent one performed at the ILL in 2009 [22].
  - Sparse data: Only **two capture measurements** and **one transmission measurement** available.

Experiment	Sample mass	Energy range
T. E. Young (1967)	~0.1 mg	Thermal-200 eV
M. G. Silbert (1973)	1.24 g	18.6-490eV
A. Chyzh (2013)	0.4 mg	Thermal-400 eV





Previous capture measurements

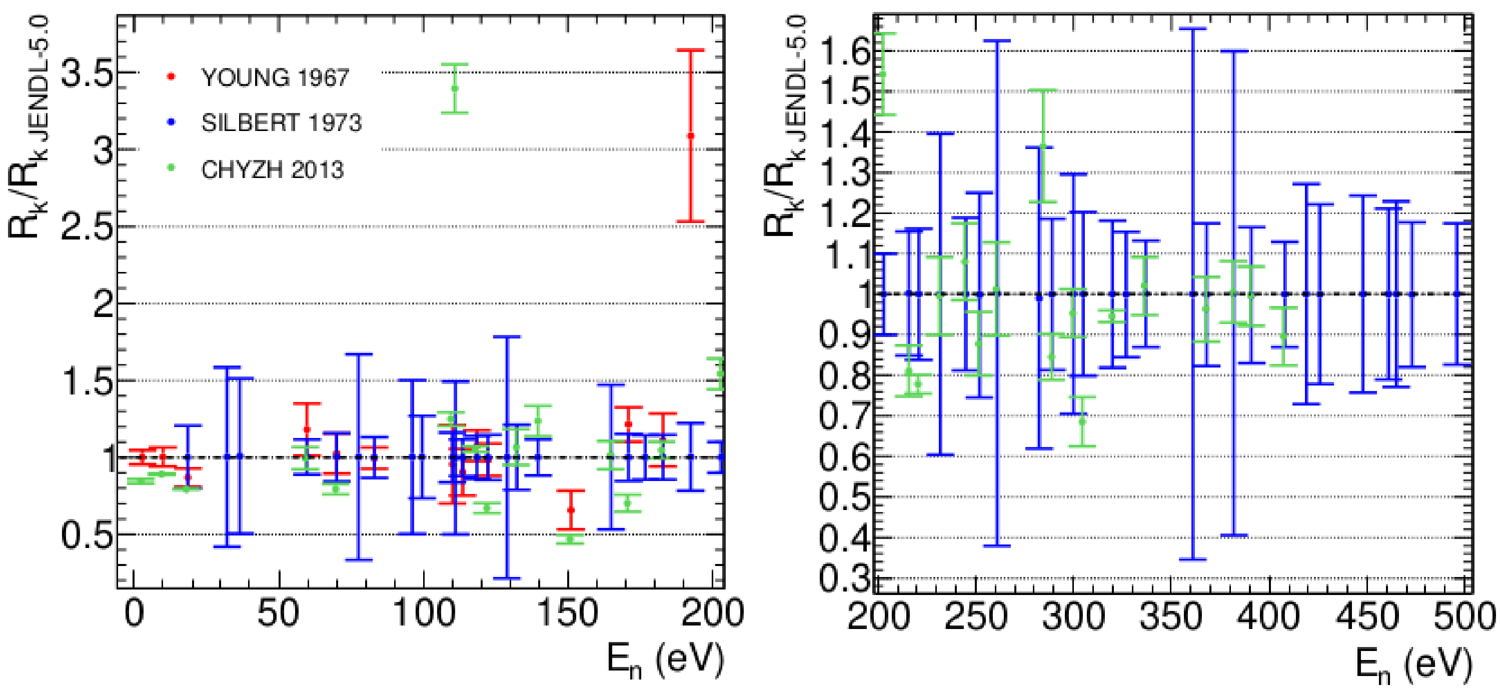
- **1967 (Young et al.) :**
  - Transmission measurement from thermal to 200 eV.
  - **Difficulties:** The data are not in EXFOR and there were considerable issues with contaminants in the sample such as actinides and water.

- **1973 (Silbert & Berreth):**
  - Capture measurement from 20 to 490 eV with a nuclear explosion, therefore is a single shot experiment.
  - **Difficulties:** Use of a single nuclear explosion unique challenges and high uncertainties in radiative kernels. Also, Differentiating capture γ-rays from other background signals was complex due to the intense neutron flux and multi-detector arrangement.

- **2013 (Chyzh et al.):**
  - Capture measurement from thermal to 400 eV with DANCE at Los Alamos
  - **Difficulties:** The extreme radioactivity of ^{238}Pu introduced significant background noise, complicating the isolation of neutron-capture events.

Overview of ^{238}Pu neutron reaction measurements

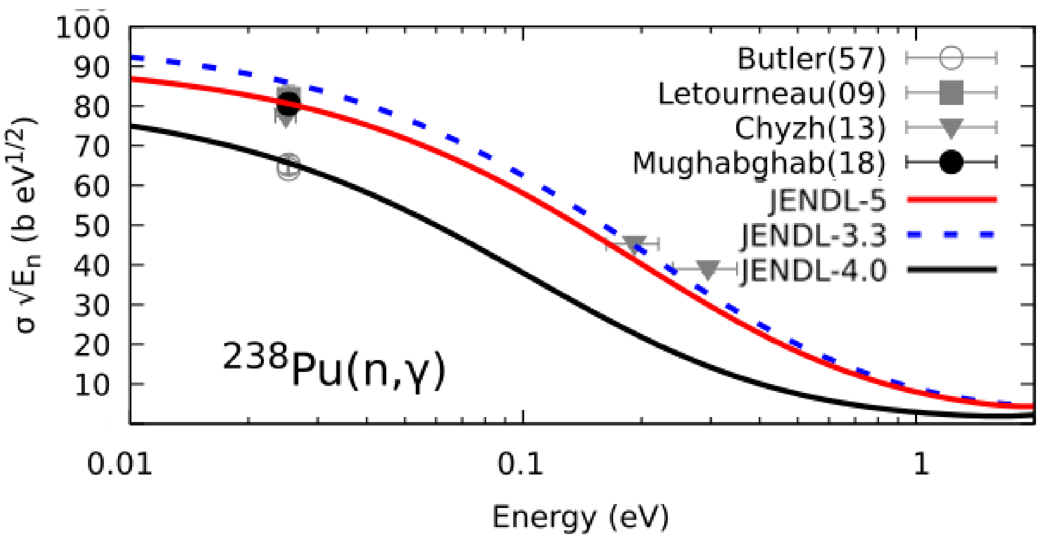
- Significant discrepancies between measurements (e.g., radiative kernels differ by 10–20%).
- Compatibility issues for higher resonance areas due to large uncertainties in earlier experiments.
- Therefore a new measurement is needed with a high instantaneous flux to reduce background and improve precision.

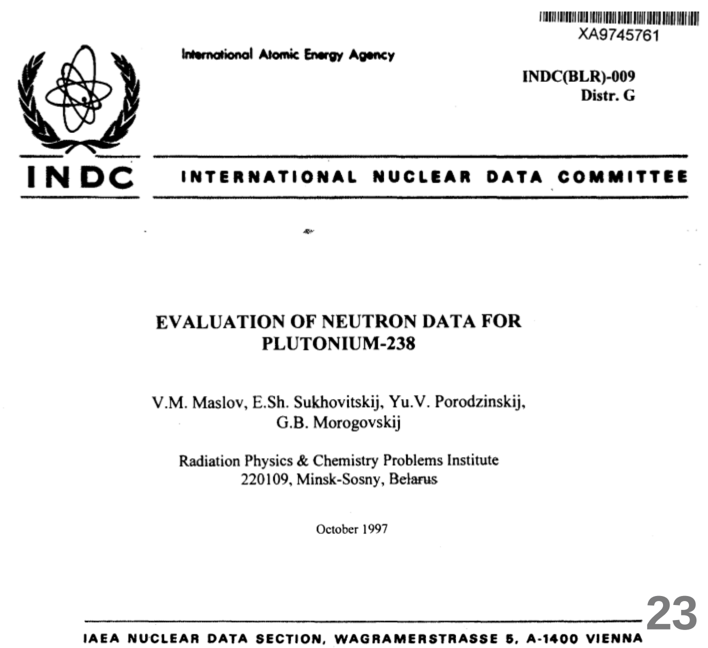


GOBIERNO DE ESPAÑA
MINISTERIO DE CIENCIA, INNOVACIÓN Y UNIVERSIDADES
Ciemat Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas
UIN

Evaluations of ^{238}Pu

- **Recent libraries:** JEFF-3.3, JENDL-4, JENDL-5, ENDF-VIII.
  - Based on Maslov's 1997 evaluation, combining data from Silbert and Young [31].
- **JENDL-5 updates:**
  - Negative resonances adjusted to match new thermal values from Chyzh and Letourneau [29].
  - Thermal capture cross-section increased by **30%**.
  - **RRR Parameters:** Remain unchanged.





XA9745761

International Atomic Energy Agency

INDC(BLR)-009
Distr. G

INDC INTERNATIONAL NUCLEAR DATA COMMITTEE

EVALUATION OF NEUTRON DATA FOR
PLUTONIUM-238

V.M. Maslov, E.Sh. Sukhovitskij, Yu.V. Porodzinskij,
G.B. Morogovskij

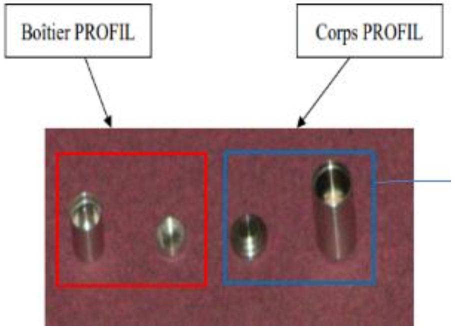
Radiation Physics & Chemistry Problems Institute
220109, Minsk-Sosny, Belarus

October 1997

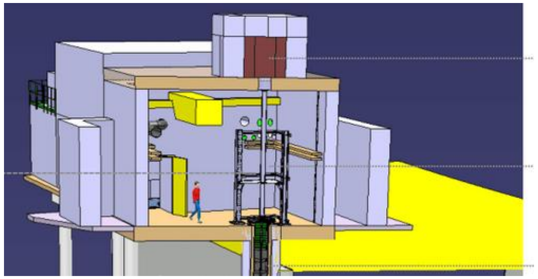
IAEA NUCLEAR DATA SECTION, WAGRAMERSTRASSE 5, A-1400 VIENNA

^{238}Pu n_TOF proposal

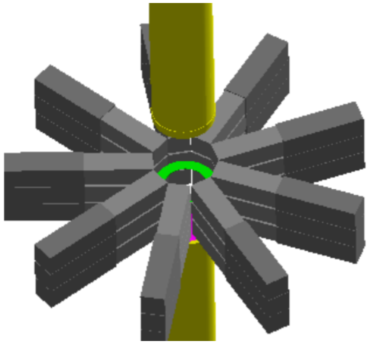
We propose to perform a capture cross section at n_TOF EAR2 to measure from 2 to 500 eV.



- **Sample:**
  - ~2 mg pellets provided by CEA Cadarache, in a stainless steel container.
  - Necessary enrichment for the measurement.



- **Experimental area:**
  - Location: n_TOF EAR2.
  - Energy range: 2-500 eV.
  - Experimental conditions: Strong instantaneous neutron flux required due to the short half-life ($T_{1/2}$ = 87.7 years) and high α-particle emission.



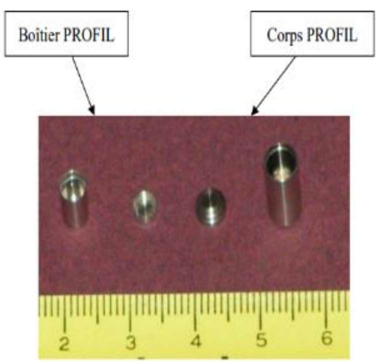
- **Detectors:**
  - Method: sTED with Pulse Height Weighting Technique (PHWT).
  - Sensitivity tailored to the experimental conditions and sample type.

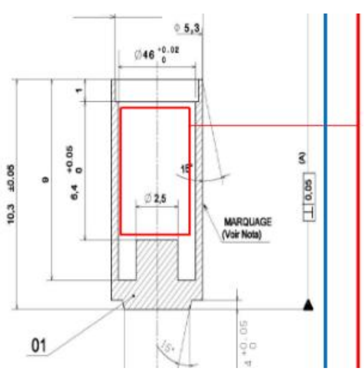
GOBIERNO DE ESPAÑA MINISTERIO DE CIENCIA, INNOVACIÓN Y UNIVERSIDADES

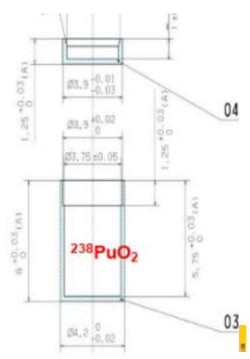
Ciemat Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas

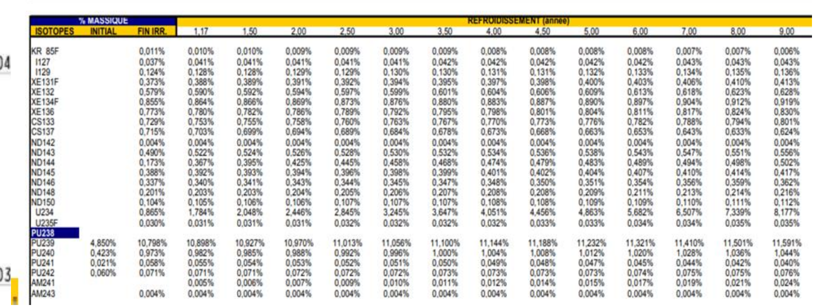
PROFIL samples of ^{238}Pu

- **Sample details:**
  - Four cylindrical ^{238}Pu samples, 4 mm height, 3.75 mm diameter.
  - Produced in the PROFIL and PROFIL2 experiments [10].
- **Irradiation:**
  - Irradiated in the Phenix reactor [11].
  - Contain ~2 mg of ^{238}Pu with ~70% enrichment.
- **Additional isotopes:**
  - Includes ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{234}U (~10%), and ^{137}Cs (major γ-ray emitter).
- **Container:**
  - Samples are housed in a stainless steel case.





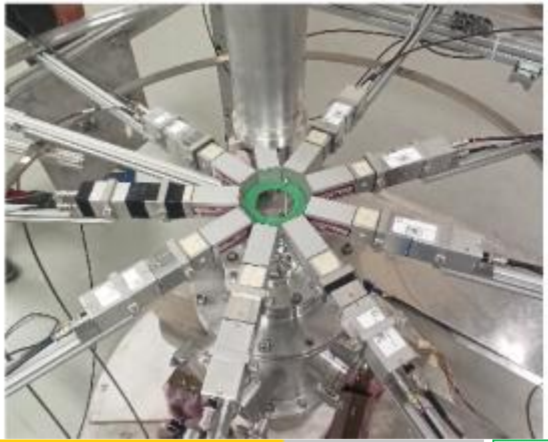


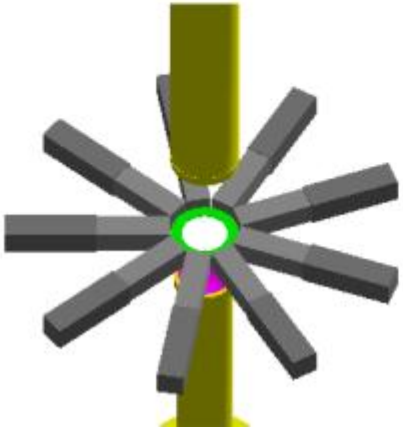


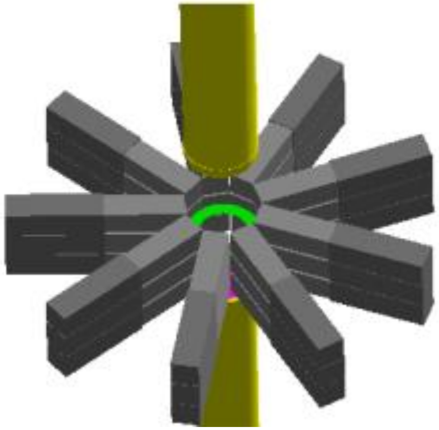
% MASSIQUE			REFROIDISSEMENT (année)												
ISOTOPES	INITIAL	FIN IRR.	1,17	1,50	2,00	2,50	3,00	3,50	4,00	4,50	5,00	6,00	7,00	8,00	9,00
KR 85F		0,011%	0,010%	0,010%	0,009%	0,009%	0,009%	0,009%	0,008%	0,008%	0,008%	0,008%	0,007%	0,007%	0,006%
I127		0,037%	0,041%	0,041%	0,041%	0,041%	0,041%	0,042%	0,042%	0,042%	0,042%	0,042%	0,043%	0,043%	0,043%
I129		0,124%	0,128%	0,128%	0,129%	0,129%	0,130%	0,130%	0,131%	0,131%	0,132%	0,133%	0,134%	0,135%	0,136%
XE131F		0,373%	0,388%	0,389%	0,391%	0,392%	0,394%	0,395%	0,397%	0,398%	0,400%	0,403%	0,406%	0,410%	0,413%
XE132		0,579%	0,590%	0,592%	0,594%	0,597%	0,599%	0,601%	0,604%	0,606%	0,609%	0,613%	0,618%	0,623%	0,628%
XE134F		0,855%	0,864%	0,866%	0,869%	0,873%	0,876%	0,880%	0,883%	0,887%	0,890%	0,897%	0,904%	0,912%	0,919%
XE136		0,773%	0,780%	0,782%	0,786%	0,789%	0,792%	0,795%	0,798%	0,801%	0,804%	0,811%	0,817%	0,824%	0,830%
CS133		0,729%	0,753%	0,755%	0,758%	0,760%	0,763%	0,767%	0,770%	0,773%	0,776%	0,782%	0,788%	0,794%	0,801%
CS137		0,715%	0,703%	0,699%	0,694%	0,689%	0,684%	0,678%	0,673%	0,668%	0,663%	0,653%	0,643%	0,633%	0,624%
ND142		0,004%	0,004%	0,004%	0,004%	0,004%	0,004%	0,004%	0,004%	0,004%	0,004%	0,004%	0,004%	0,004%	0,004%
ND143		0,490%	0,522%	0,524%	0,526%	0,528%	0,530%	0,532%	0,534%	0,536%	0,538%	0,543%	0,547%	0,551%	0,556%
ND144		0,173%	0,367%	0,395%	0,425%	0,445%	0,458%	0,468%	0,474%	0,479%	0,483%	0,489%	0,494%	0,498%	0,502%
ND145		0,388%	0,392%	0,393%	0,394%	0,396%	0,398%	0,399%	0,401%	0,402%	0,404%	0,407%	0,410%	0,414%	0,417%
ND146		0,337%	0,340%	0,341%	0,343%	0,344%	0,345%	0,347%	0,348%	0,350%	0,351%	0,354%	0,356%	0,359%	0,362%
ND148		0,201%	0,203%	0,203%	0,204%	0,205%	0,206%	0,207%	0,208%	0,208%	0,209%	0,211%	0,213%	0,214%	0,216%
ND150		0,104%	0,105%	0,106%	0,106%	0,107%	0,107%	0,107%	0,108%	0,108%	0,109%	0,109%	0,110%	0,111%	0,112%
U234		0,865%	1,784%	2,048%	2,446%	2,845%	3,245%	3,647%	4,051%	4,456%	4,863%	5,682%	6,507%	7,339%	8,177%
U235F		0,030%	0,031%	0,031%	0,031%	0,032%	0,032%	0,032%	0,032%	0,033%	0,033%	0,034%	0,034%	0,035%	0,035%
PU238															
PU239	4,850%	10,798%	10,898%	10,927%	10,970%	11,013%	11,056%	11,100%	11,144%	11,188%	11,232%	11,321%	11,410%	11,501%	11,591%
PU240	0,423%	0,973%	0,982%	0,985%	0,988%	0,992%	0,996%	1,000%	1,004%	1,008%	1,012%	1,020%	1,028%	1,036%	1,044%
PU241	0,021%	0,058%	0,055%	0,054%	0,053%	0,052%	0,051%	0,050%	0,049%	0,048%	0,047%	0,045%	0,044%	0,042%	0,040%
PU242	0,060%	0,071%	0,071%	0,071%	0,072%	0,072%	0,072%	0,073%	0,073%	0,073%	0,073%	0,074%	0,075%	0,075%	0,076%
AM241			0,005%	0,006%	0,007%	0,009%	0,010%	0,011%	0,012%	0,014%	0,015%	0,017%	0,019%	0,021%	0,024%
AM243		0,004%	0,004%	0,004%	0,004%	0,004%	0,004%	0,004%	0,004%	0,004%	0,004%	0,004%	0,004%	0,004%	0,004%

Detection setup

- **Current capabilities:**
  - sTED enables capture measurements at EAR2 from thermal to hundreds of keV.
  - Currently used in a 9-module array.
- **Future upgrade:**
  - Plans to upgrade to a 27-module array [35].
  - New detectors to be commissioned in 2025.
- **Configuration options:**
  - Initial use of 9-module setup (ε_{cap}=~6); potential switch to 27 (ε_{cap}=~18) -module configuration if performance is favorable.







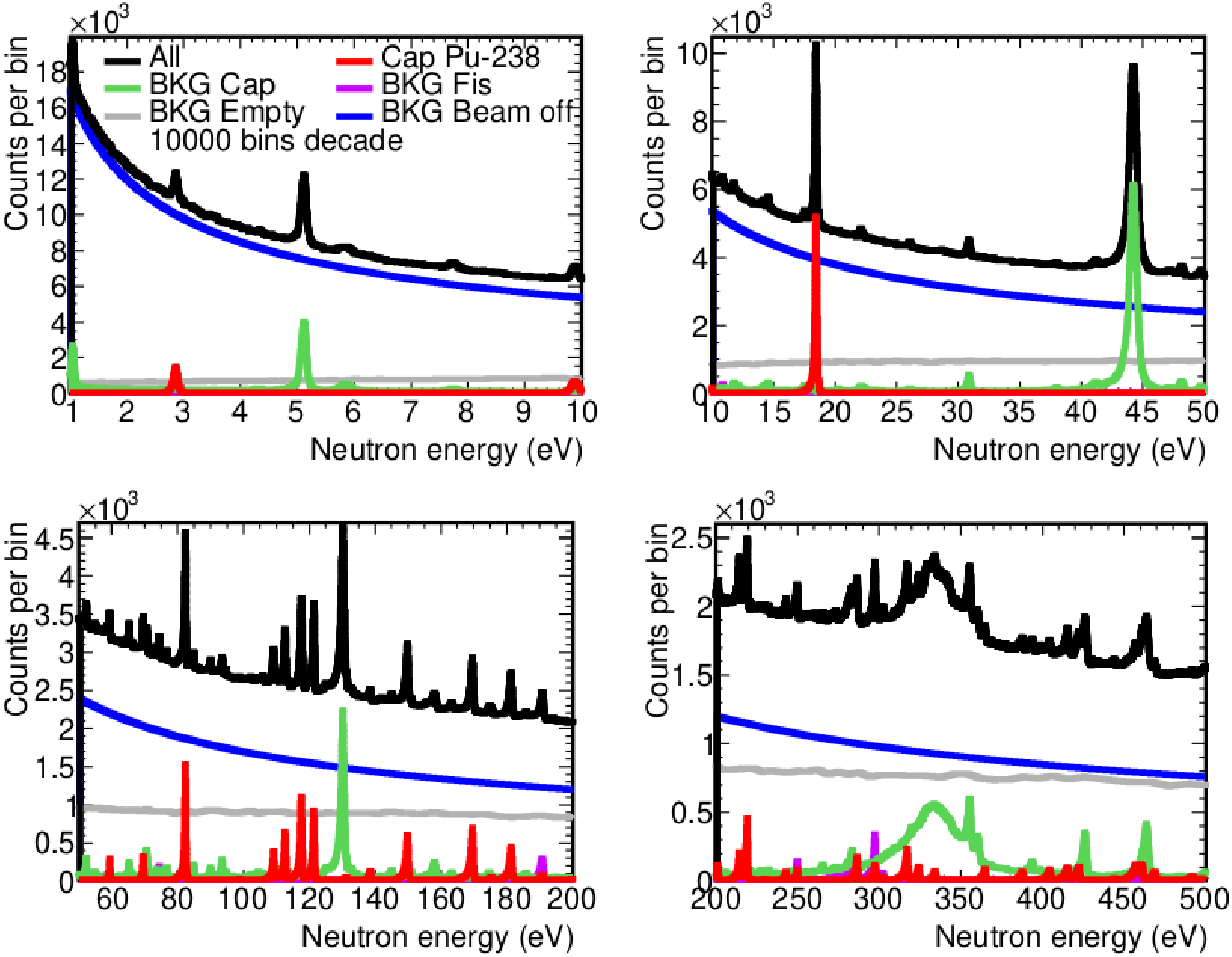
Counting rate estimates for ^{238}Pu

The counting rate estimates have been performed for 2 mg of ^{238}Pu, 9 sTEDs, including the RF and a total of $2 \cdot 10^{18}$ protons.

Different estimated backgrounds:

- Background produced by capture reactions in the isotopes of the pellet or in the stainless steel case (BKG Cap)
- Background produced by fission reactions in the actinides of the sample (BKG Fis)
- The empty background measured in previous experimental campaigns (BKG Empty)
- Background produced by the radioactivity of the sample mainly caused by ^{137}Cs (BKG Beam Off)

Counting rate estimate u^{238}Pu



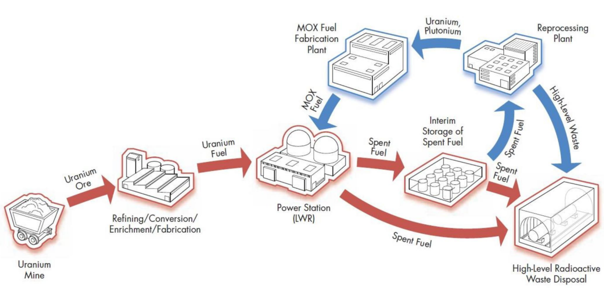
Estimation of number of protons

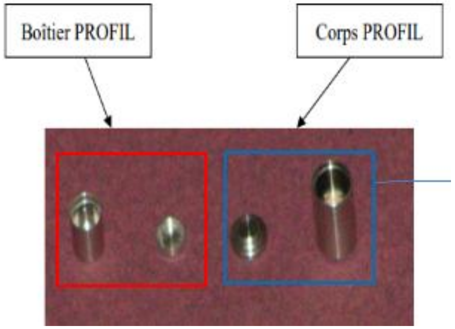
The estimated number of protons ($2 \cdot 10^{18}$) has been chosen to have uncertainties from both the subtraction of the background and statistics that are similar.

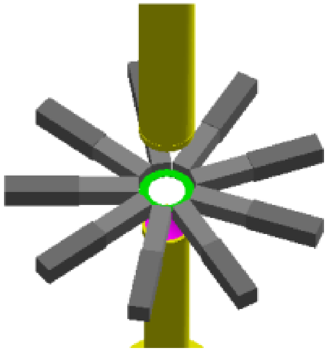
A total of 35 resonances would be measured, with half of them having an uncertainty lower than 10%.

The plan is to perform the **measurement in 2026**.The sample is not ready in 2025.

Measurement	Protons
Pu^{238}sample	$2 \cdot 10^{18}$
Dummy sample	$1 \cdot 10^{18}$
Auxiliary and Normalization	$1 \cdot 10^{18}$
Total	$4 \cdot 10^{18}$



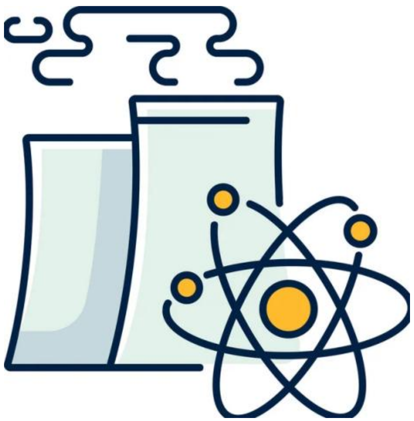


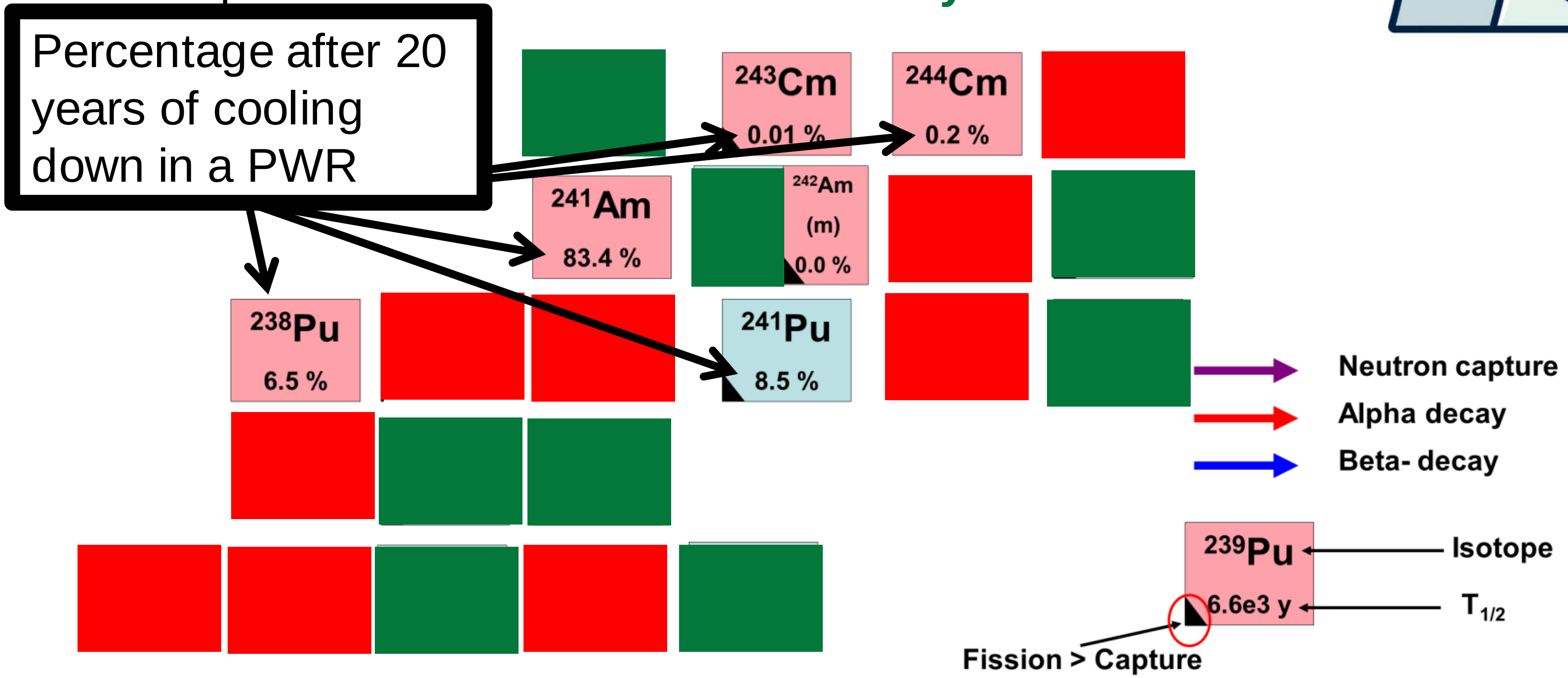


BACK-UP SLIDES

Production of actinides in nuclear reactors

- **Importance:** transport, transmutation, and storage of spent fuel.
- Isotopes with half-lives **longer than 1e3 year** are mainly important in the long term disposal.
- Isotopes with half-lives **shorter than 1 year**





GOBIERNO DE ESPAÑA MINISTERIO DE CIENCIA, INNOVACIÓN Y UNIVERSIDADES

Ciemat Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas

UIN