(p,n) (n,gamma) standard comparison in service of “ Activation/Dismantling of materials and radionulides inventory in Accelerator Driven Systems ”

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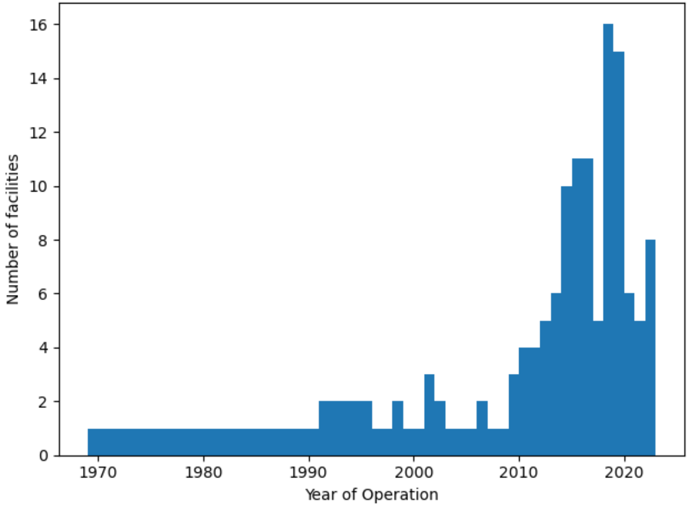
Jean-Michel **HORODYNSKI** (iRSD)

An increasing number of accelerators

← **IAEA** listed in **59** different countries **577** accelerators (as of 2020) with **19** manufacturers [1]

IAEA
International Atomic Energy Agency



When **PTCOG** catalogued, in **22** countries, **123** particle therapy facilities in clinical operation (as of May 2023) [2] with an acceleration since 2010 →

Particle Therapy Co-Operative Group

Such facilities are becoming a dismantling hazard

Accelerator Driven Systems : an emerging solution

ADS are a new gen nuclear reactor coupling a particle accelerator and a nuclear reactor for safer energy and allowing nuclear waste transmutation
Fission process is sustained by the spallation neutrons created with high energy proton beam

Key advantages :

- Reduce toxicity of existing nuclear waste from 300 000 years and more to less than 500 years.
- Intrisically safe-carbon free energy production due to its subcritical characteristics

Some ADS around the world :

- Transmutation Experimental Facility (TEF) [Japan]
  - Proton beam → 100 MeV (max. 400 MeV/250kW) [5]
- Multipurpose Hybrid Research Reactor for High-tech Applications (MYRRHA) [Belgium]
  - Proton beam → 600 MeV with 4 mA [6]
- Chinese Initiative Accelerator Driven Systems (CiADS) [China]
  - Proton beam → 250 MeV with 10 mA (max 1 GeV) [7]

Transmutex

START (**S**ubcritical **T**ransmuting **A**ccelerated **R**eactor **T**echnology) Project

- **Reactor :** molten lead reactor
- **Technology** : Fast neutron
- **Proton beam :** 5mA with 800 MeV (4 MW)

A decommissioning challenge

Different countries with different radioactive waste classification [3]:

- France, the classification of radioactive waste is based on level of radioactivity and half-life
- Japan, have chosen a classification by production chain
- Germany, classification mainly concerns the exothermic nature of the waste

ANDRA nagra. NUMO 原子力発電環境整備機構 BGE BUNDESGESELLSCHAFT FÜR ENDLAGERUNG …

IAEA International Atomic Energy Agency

↘ recommends for decommissioning plans before any facilities construction and must be regularly updated [4]

⇒ **radionuclide inventory**

Facilities have expanded their expertise by creating dedicated groups or developing software like ActiWiz (CERN)

PhD project –
"Activation/Dismantling of materials and radionulides inventory in ADS systems"

Objectives :

- Draft dismantling plans for future TMX prototypes
- First radionuclides inventory produced by activation
- ➢ Development of a dedicated tool for TMX installations, a hybride of analytical and MC codes

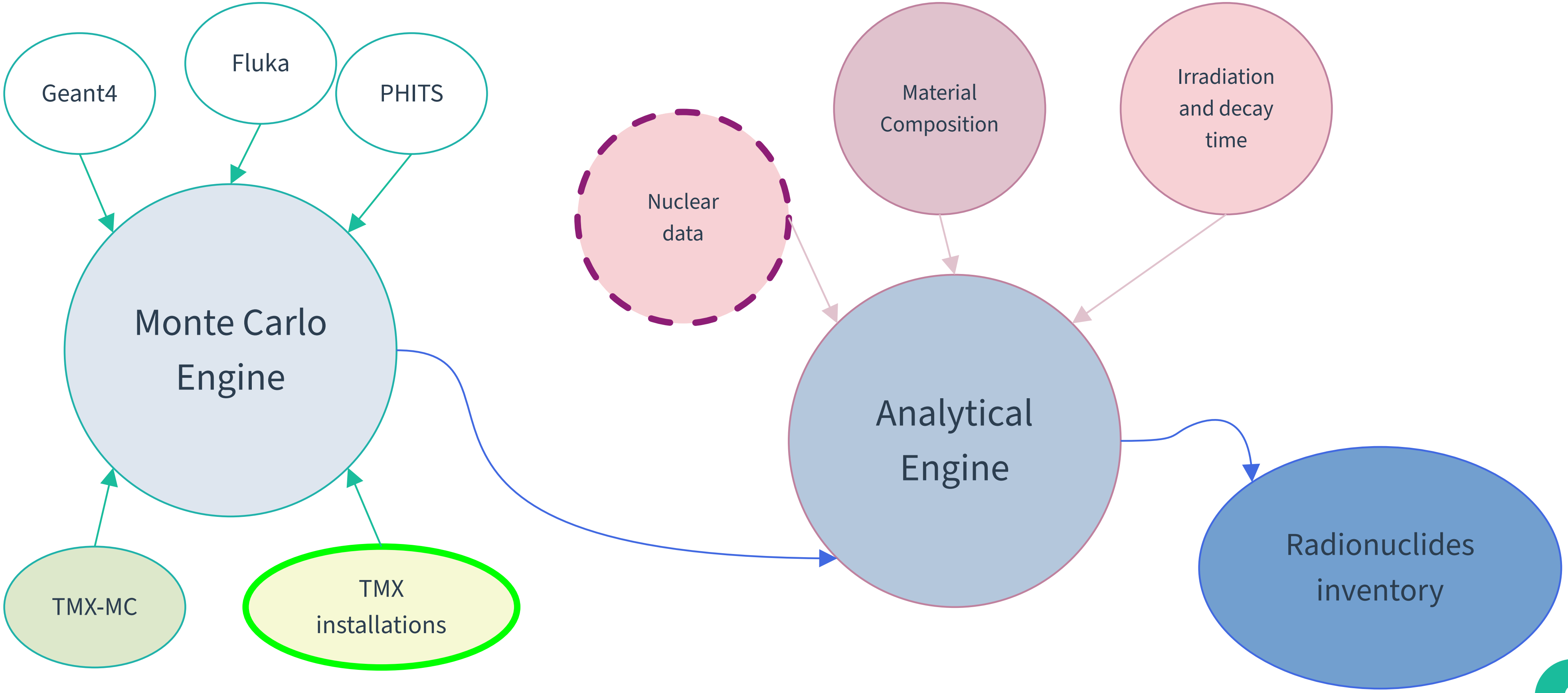
Milestones :

- State-of-the-art on available data for the reactions of interest (p, xn), (ɣ, xn), …
- Intervalidation of codes Fluka/Geant4/MCNP/PHITS for neutron activation

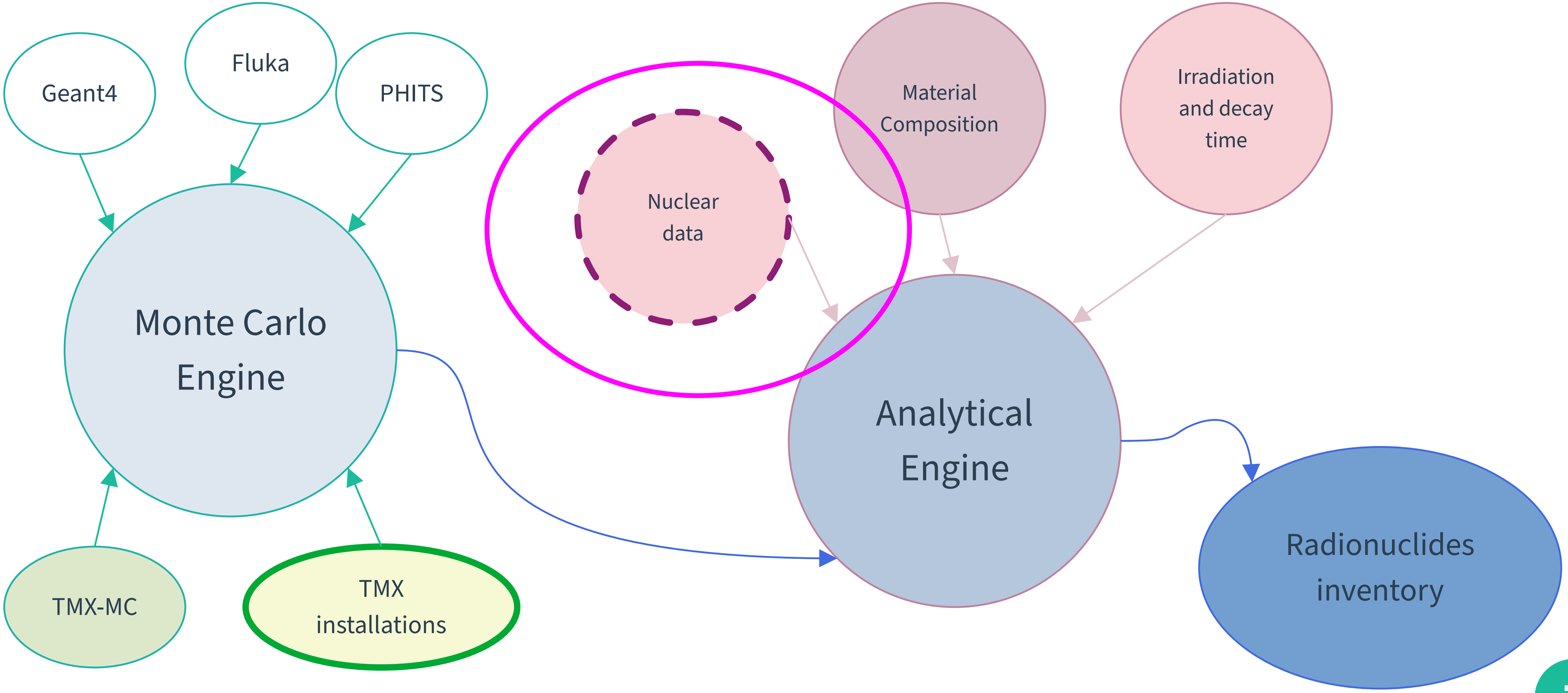
Context :

- IAEA listed 2 dozens of commonly found materials around accelerator facilities (inside most activated components) [4]
- Large energy spectrum of proton : from **1 MeV up to 1 GeV**
  - Studies split in several range of energies
    - Starters : below 20 MeV
    - Expanded over available facilities like Arronax, PSI

A journey to radionuclides inventory



A journey to radionuclides inventory



A journey in OECD JANIS cross section - myJanis

Idea :

Pythonic tool clean-up and automatised comparison of crossections from JANIS OECD (regrouping in particular ENDF, JENDL, JEFF, TENDL, …) and EXFOR, then MC codes

- Will reformat all files to a standardised structure
- Do average, delta, ratios to have systematic comparison of every models available
- Create an existence matrix, are models fully assertained by experimental data
- Collapse data (by integration) on ranges of interest
- Find threshold of reactions (mostly (p,n) and (ɣ, n))

A journey in OECD JANIS cross section - myJanis

Extraction status :

- ➢ For the materials present around accelerator (70 isotopes, all available models and reactions for :
  - Incident proton
  - Incident neutron have been extracted (only MT1, MT2 and MT102 were taken when available)
- ✗ EXFOR extraction → due to large file difference from JANIS + non aligment of data points with models
- ✗ MC extraction → in progress

Script status:

- Averaging functionalities ; interval reducing function (integration) implemented
- Deltas (abs, relative) / Ratio / Threhold extraction
- Plotting functions for σ(E) / Collapsing
- ✗ Jupyter conversion / Existence matrix

MyJANIS for Evaluated Nuclear Data Libraries

Step 1 : Original data : from JANIS filename should contain target/product/MT code

Incident energy	BROND-3.1	CENDL-3.2	ENDF/B-VIII.0	JEFF-3.3	JENDL-4.0u	JENDL/HE-2007	TENDL-2019
Incident energy							
Incident energy	σ(E)	σ(E)	σ(E)	σ(E)	σ(E)	σ(E)	σ(E)
0.00001	1177.24876	1179.88206	1177.264	1177.249	1177.24876	1179.89187	1178.66992
0.00001032234	1159.12562414624	1161.31874	1159.140240096	1159.12575584	1159.12562414624	1161.3284	1160.12567
0.000010625	1142.10897	1144.69823121508	1142.123	1142.109	1142.10897	1144.70774560484	1143.5222340166
0.00001065491	1140.56946018672	1143.05573	1140.58347248	1140.56947248	1140.56946018672	1143.06523	1141.88142
0.00001099835	1122.8921198232	1125.07228	1122.9059288	1122.8919288	1122.8921198232	1125.08164	1123.91645
0.00001125	1109.93934	1112.50764036093	1109.953	1109.939	1109.93934	1112.5168938379	1111.36472095186
0.00001135271	1105.07580293312	1107.37943	1105.089311744	1105.07547608	1105.07580293312	1107.38864	1106.24178
0.00001171864	1087.74823873408	1089.95774	1087.761208896	1087.74795872	1087.74823873408	1089.96681	1088.83798
0.000011875	1080.34426	1082.85950040046	1080.357	1080.344	1080.34426	1082.86851242339	1081.74703308684

t-065Tb159_p-065Tb160_c-MT102_raw.csv

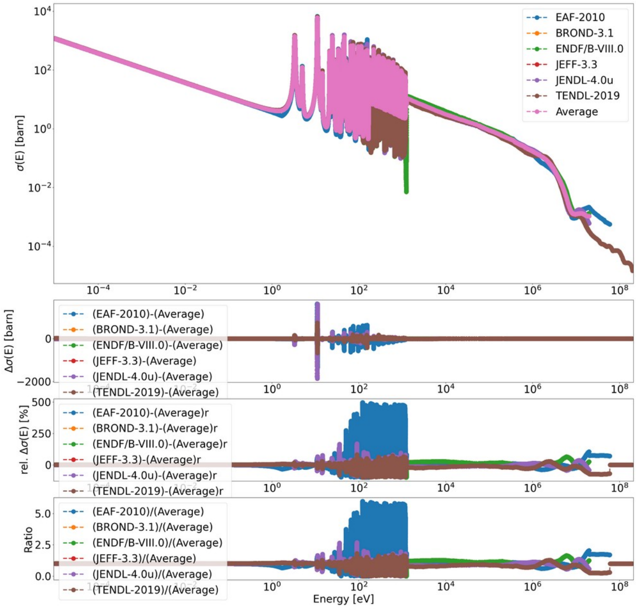
Step 2 : Reformatted data : unique header/standard infos (target, product, MT, model content)

energy	BROND-3.1	CENDL-3.2	ENDF/B-VIII.0	JEFF-3.3	JENDL/HE-2007	JENDL-4.0u	TENDL-2019
0.00001	1177.24876	1179.88206	1177.264	1177.249	1179.89187	1177.24876	1178.66992
0.00001032234	1159.125624	1161.31874	1159.14024	1159.125756	1161.3284	1159.125624	1160.12567
0.000010625	1142.10897	1144.698231	1142.123	1142.109	1144.707746	1142.10897	1143.522234
0.00001065491	1140.56946	1143.05573	1140.583472	1140.569472	1143.06523	1140.56946	1141.88142
0.00001099835	1122.89212	1125.07228	1122.905929	1122.891929	1125.08164	1122.89212	1123.91645
0.00001125	1109.93934	1112.50764	1109.953	1109.939	1112.516894	1109.93934	1111.364721
0.00001135271	1105.075803	1107.37943	1105.089312	1105.075476	1107.38864	1105.075803	1106.24178
0.00001171864	1087.748239	1089.95774	1087.761209	1087.747959	1089.96681	1087.748239	1088.83798
0.000011875	1080.34426	1082.8595	1080.357	1080.344	1082.868512	1080.34426	1081.747033
0.00001209621	1070.665466	1072.81728	1070.678266	1070.665266	1072.82621	1070.665466	1071.71513
0.0000124861	1053.606269	1055.9398	1053.619175	1053.606175	1055.94859	1053.606269	1054.85499

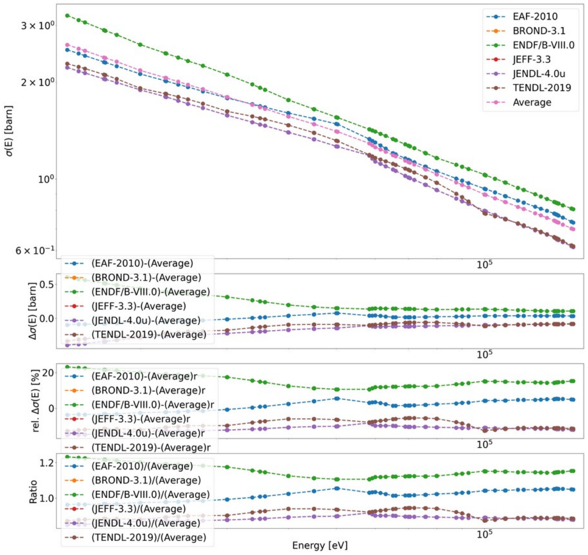
t-065Tb159_p-065Tb160_c-MT102_m-ABEFJT.csv

A journey in OECD JANIS cross section - myJanis

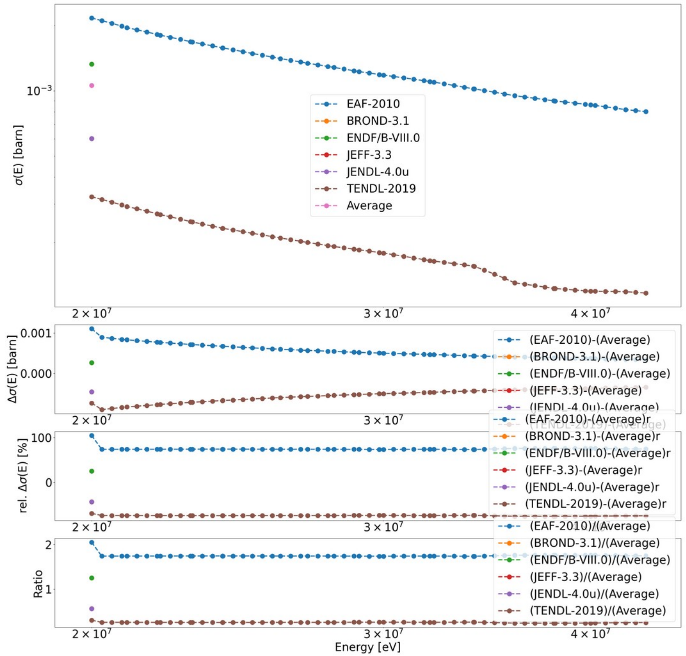
Step 3 : Generation of plots by energy sections ->



Full energy range



[1e4 – 1e6] eV



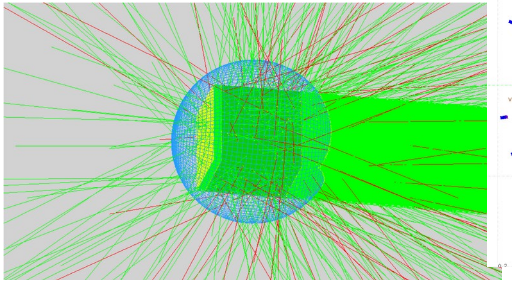
[20e6 – 40e6] eV

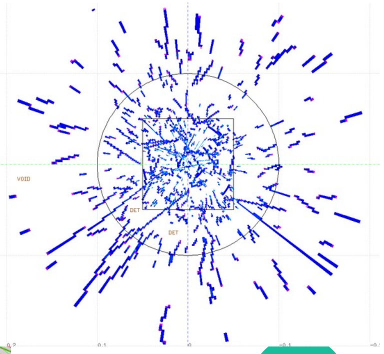
Mid conclusion

- Range existence still need to be compared
- EXFOR need to be extracted
  - Beware to very different structures →

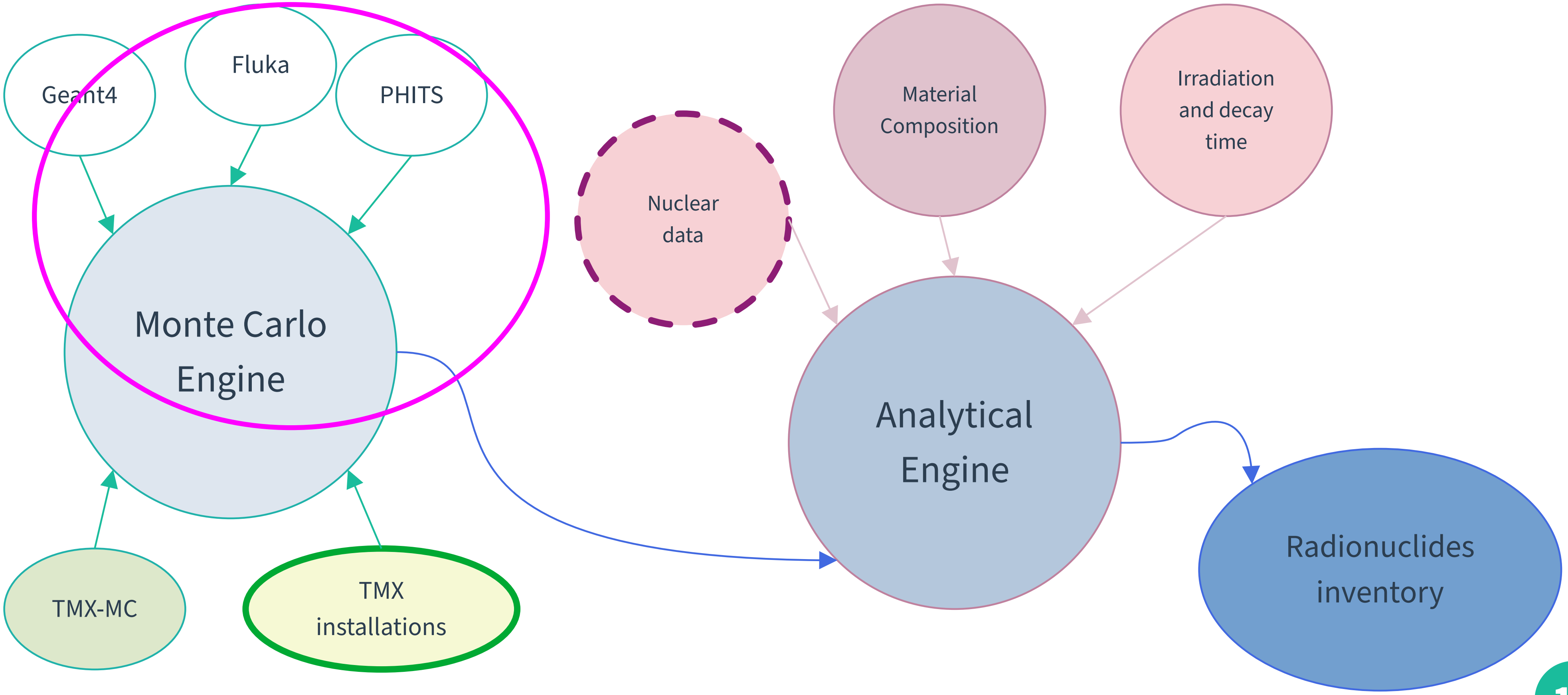
EN	A0172.002	A0172.002	A0172.002	A0172.002	A01[illegible]	A0923.002	A0923.002	A1466.002	A1466.002	A1466.002	A1466.005	A1466.005	A1466.005
EN													
EN	DATA	ERR-T	ERR-1	ERR-2	ERR R	DATA	DATA-ERR	EN-ERR-DIG	DATA	ERR-DIG	EN-ERR-DIG	DATA	ERR-DIG
1.840 MeV						30.5741 mb	20 %						
1.8615 MeV													
1.8649 MeV													
1.8668 MeV													
1.8696 MeV													
1.8725 MeV													
1.8740 MeV													
1.8759 MeV													
1.8784 MeV								0.0003 MeV	0.0005 b	0.0004 b			
1.8788 MeV													
1.8793 MeV								0.0003 MeV	0.0124 b	0.0004 b			
1.8796 MeV											0.00003 MeV	0.00052137 b	0.0001 b
1.8798 MeV											0.00003 MeV	0.00051856 b	0.0001 b
1.8799 MeV											0.00003 MeV	0.0010810 b	0.0001 b
1.8800 MeV											0.00003 MeV	0.0016437 b	0.0001 b
1.8802 MeV											0.00003 MeV	0.0031471 b	0.0001 b
1.8803 MeV											0.00003 MeV	0.0065324 b	0.0001 b

- Monte Carlo extraction :
  - PHITS the most ergonomic
    - Icntl = 3 → choice of energy based of corresponding in Evaluated Data Libraries
  - Geant4 → Use of Stepping Action and MT recognition
  - Fluka → Home-made fortran snippet needed
    - At least, particle yield for several slab of thickness 100 keV →

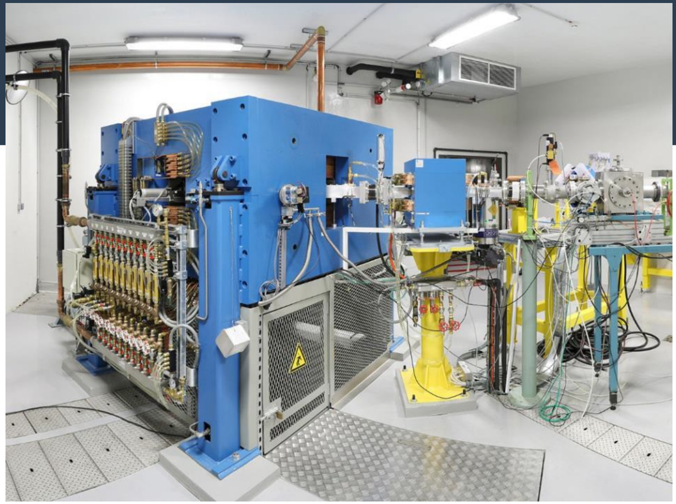




A journey to radionuclides inventory



Secondary Neutron Field from cyclotron



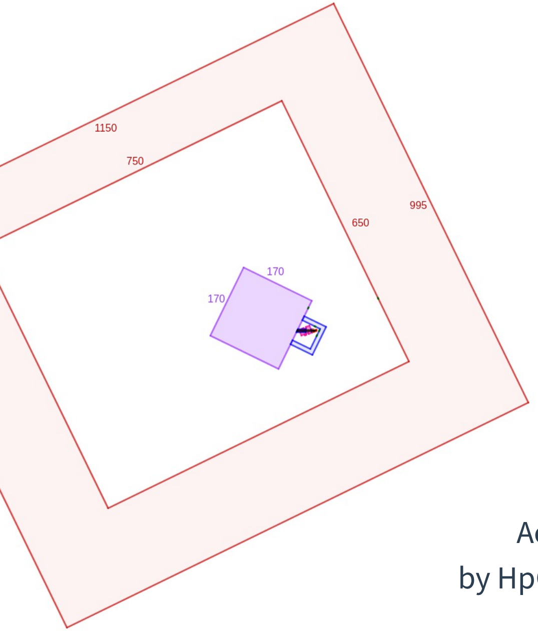
- TR24 from *Advanced Cyclotron System Inc*
- ***Irradiation line :***
  - Energy range : 16-25 MeV (lower can be achieved thanks to aluminium degrader)
  - Current : fA to 10 nA

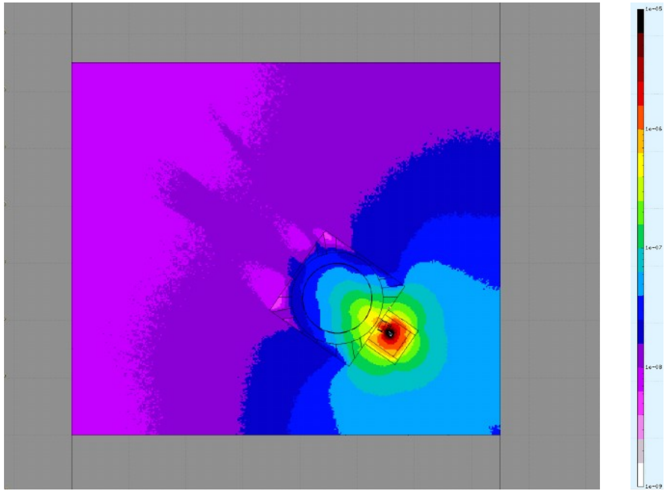
CYRCé production line :

- liquid $H_2{}^{18}O$ for ^{18}F production :
  - Irradiation of 20 minutes to 1 hours
  - 18 MeV with 80 µA (3.8 mL)
  - 16.5 MeV with 35 µA (1 mL)
- solid ^{64}Ni for ^{64}Cu production :
  - Irradiation of 4 to 5 hours
  - 11 MeV with 150-200 µA/h
- solid ^{89}Y for ^{89}Zr production :
  - < 13 MeV with 150-200 µA/h

Discrepancies between codes :
Activation around a medical isotope production unit

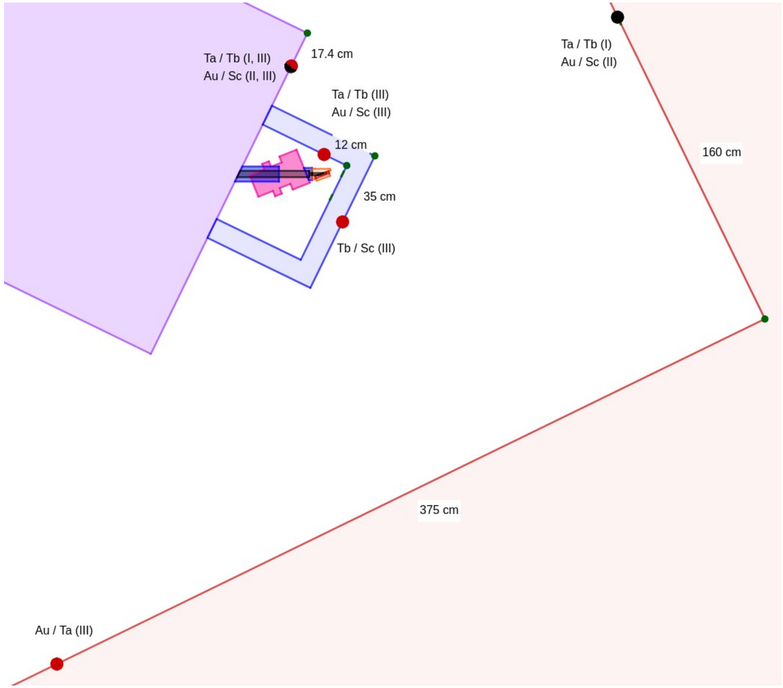
Measurement of activation of different samples inside the “casemate”





Expected neutron fluence inside the cyclotron room [Fluka]

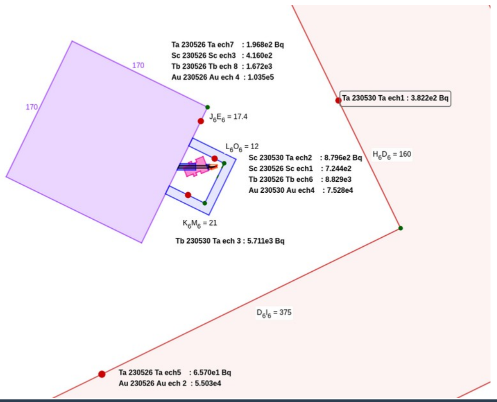
⇒ Tantalum/Terbium/Scandium/Gold

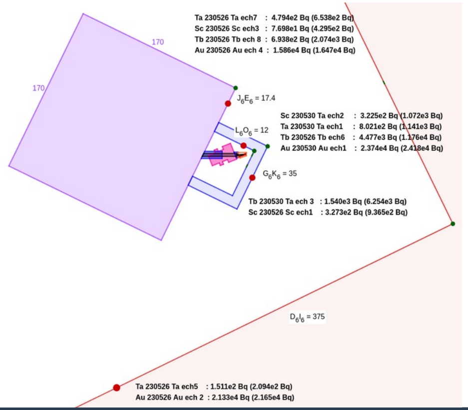


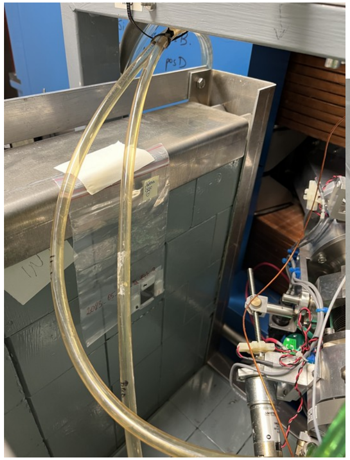
Activation estimation
by HpGe gamme spectrometry

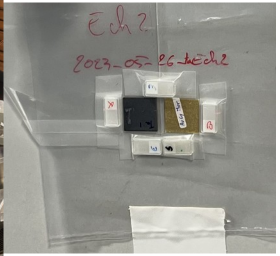
Furthering of experiment - I

- Run 2021 (I) /2022 (II) : Samples located on cyclotron yoke and nearby wall → 1 mL H_2O_{18} target with energy : 16.5 MeV ; current : 35 µA ; profile : 4*20 minutes
- Run III → Samples located on cyclotron yoke, nearby wall and inside biological shielding → solid 89Zr target with energy : 12.5 MeV current : 60 µA ; profile : 1*3h
- **Run IV** → Samples located on cyclotron yoke and nearby walls, inside biological shielding → 3.8 mL H_2O_{18} target with energy : 18 MeV current : 75 µA ; profile : 1*45min





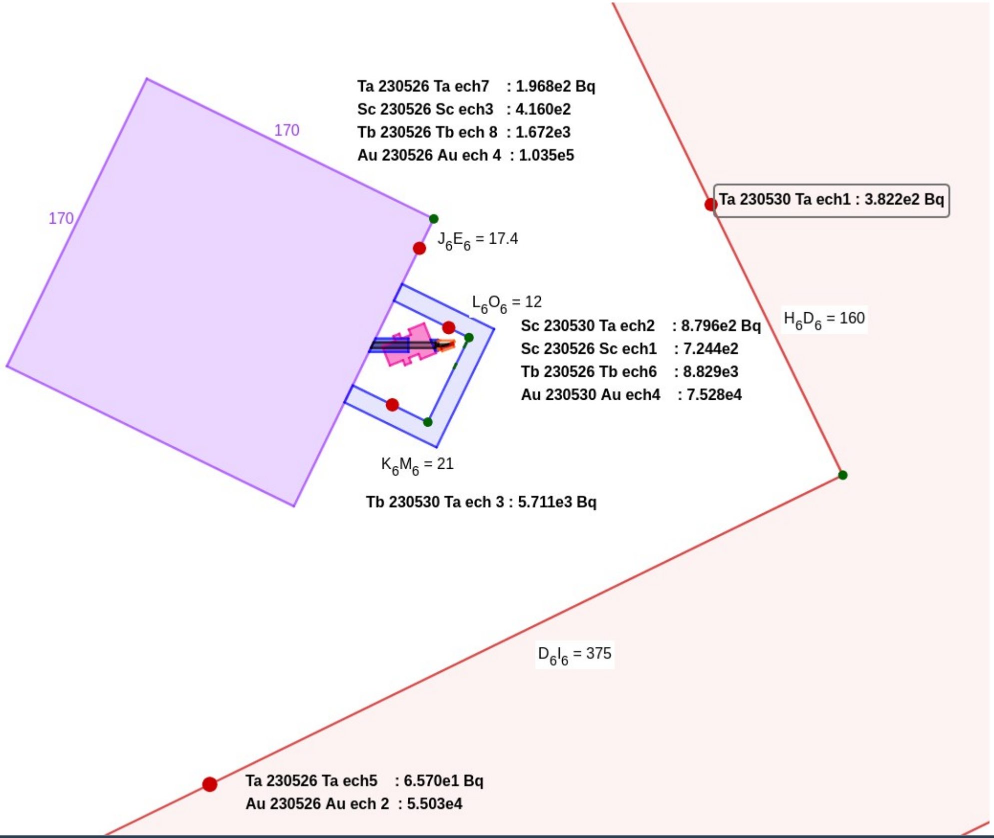


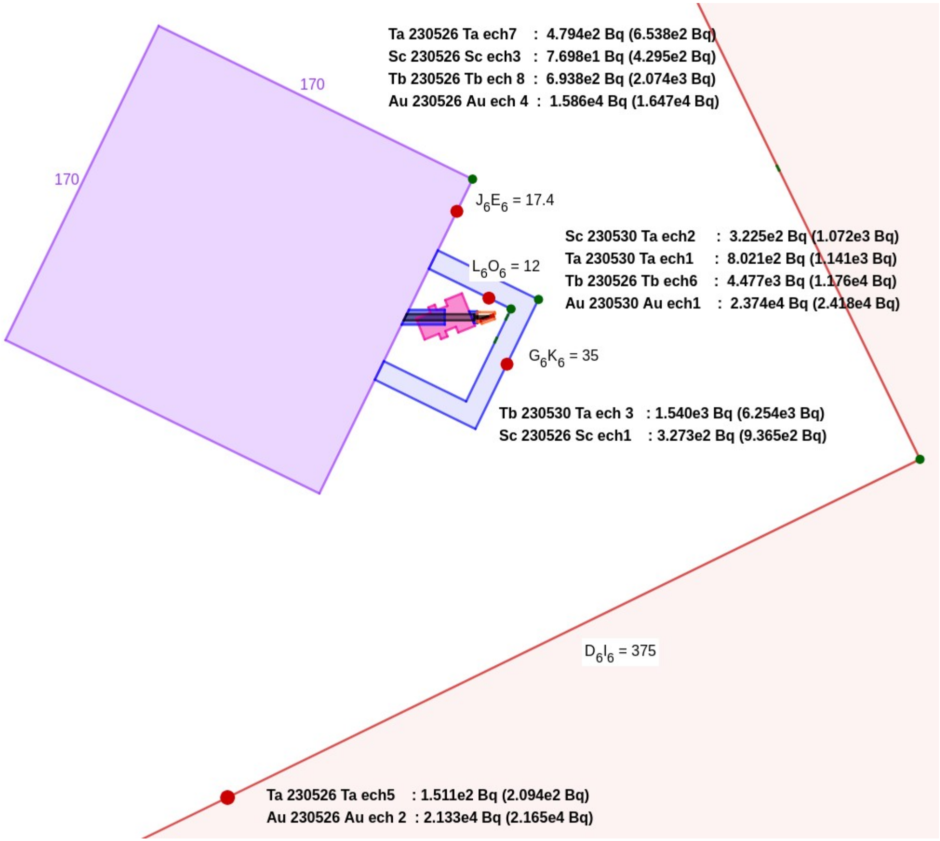


Furthering of experiment - II

Run III

Run IV



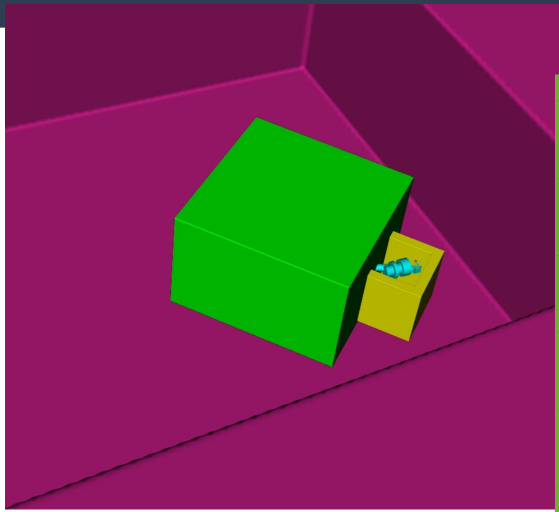


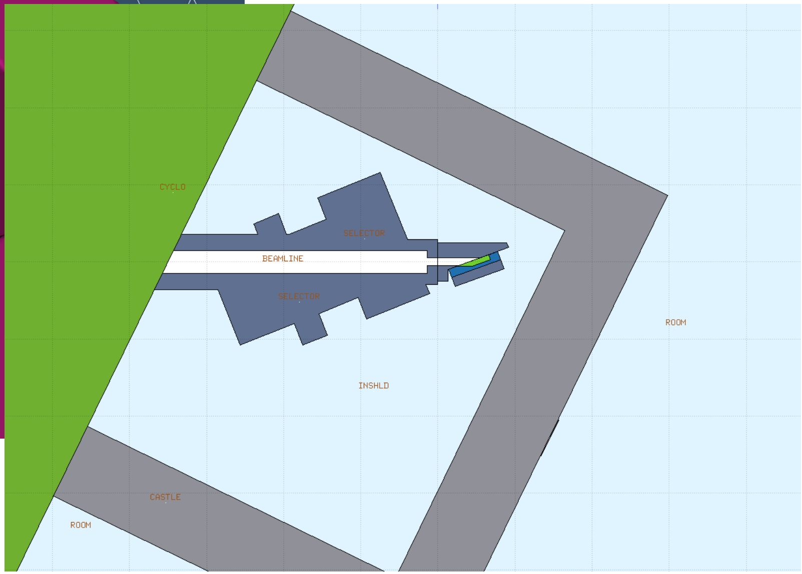
Monte Carlo simulations

Model of 3.8 mL H2O18 setup :

- Fluka
- FlukaCERN
- PHITS
- (MCNP)
- Geant4

Combinatorial geometry





Access to neutron spectrum :

Fluka/FlukaCERN → USRTRACK

PHITS → [T-Track]

Geant4 → Stepping Action (fGeomBoundary)

Access to Activity (Bq)

Fluka → RESNUCLEi

FlukaCERN → RESNUCLEi

PHITS → D-CHAIN

FISPACT-II

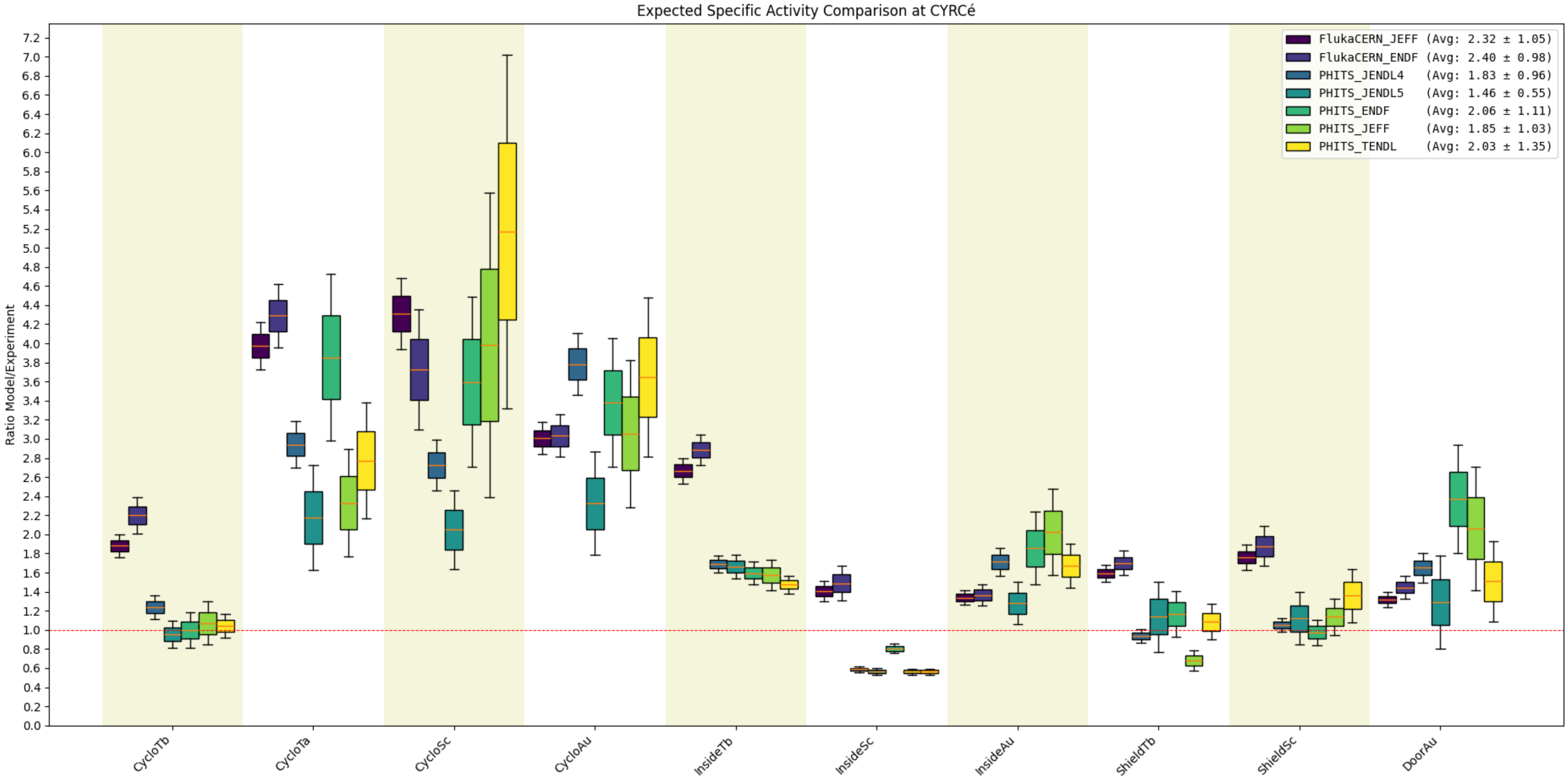
Access to decay gamma spectrum :

Fluka/FlukaCERN → USRTRACK + DCYTIMES

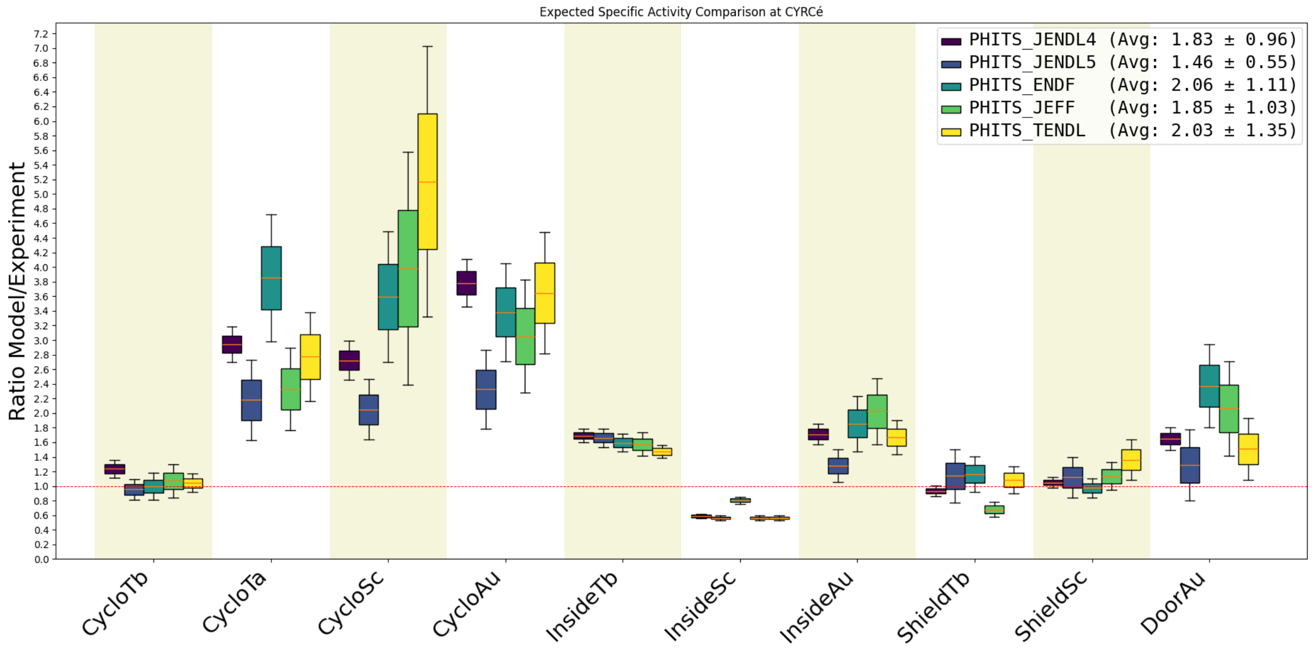
PHITS → D-CHAIN

Geant4

Compilation of results – Simulation/Experiment ratio



Compilation of results

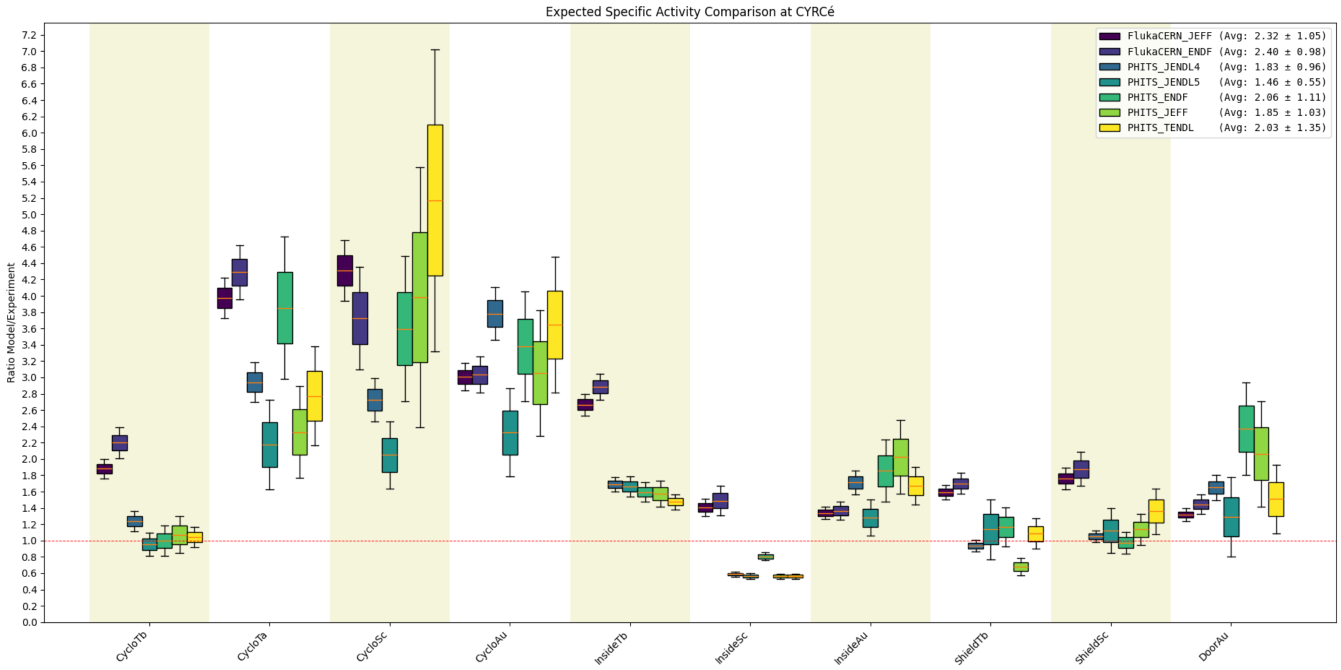


Systematic errors between codes is the biggest differentiator, especially for multi steps simulation

⇒ Experiments are the only validation

Biggest impact of evaluated data library choice is for instantaneous look (cyclotron yoke samples)

Compilation of results



Systematic errors between codes is the biggest differentiator, especially for multi steps simulation

⇒ Experiments are the only validation

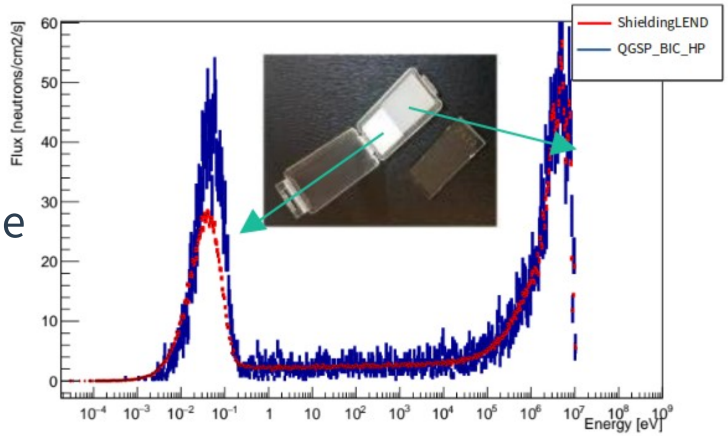
Biggest impact of evaluated data library choice is for instantaneous look (cyclotron yoke samples)

For this neutron secondary field, PHITS seems to have the best agreement.

Discrepancies starts at 18F production (overestimation of Fluka)

Conclusion – Take Home message

- **For well known, and simple reaction (i.e. only nCapture), nuclear library is mostly refinment factor**
  - But with different branching ratios, it becomes a mandatory consideration Especially for radio protection operators during facility functionment
- **What's to be done :**
  - Check the solid target to see if PHITS best competitor
  - Add FISPACT-II activity comparison to avoid local tambouille
  - Add comparison of neutron spectrum, with SSNTD (solid state nuclear track detector) as experimental discriminant



Merci !

THANKS !

Danke !

Trugarez !

谢谢 !