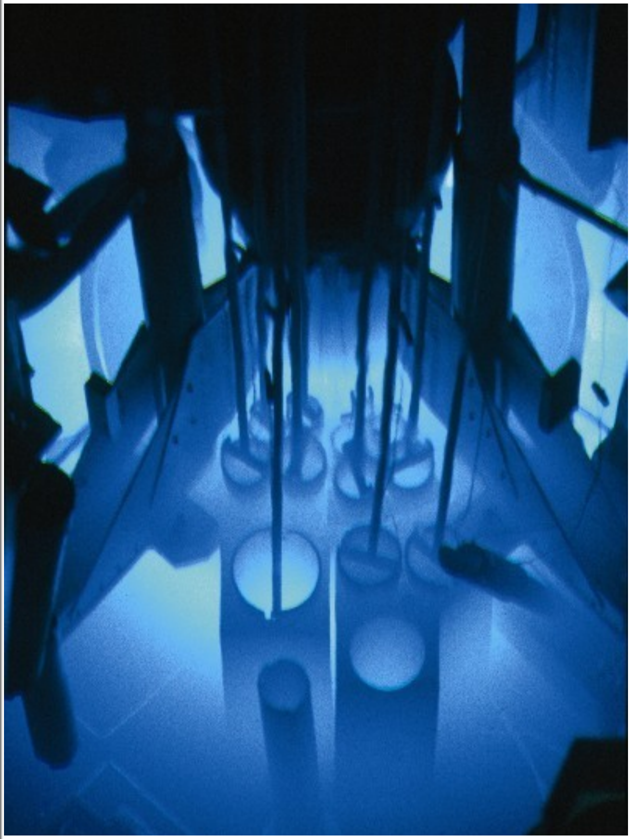
Challenges and Successes in Application of the Evaluated Nuclear Data for the Missouri University Research Reactor Core Irradiation Simulations

N.J. Peters
University of Missouri Research Reactor Facility
1513 Research Park Drive, Columbia, Missouri 65211 – U.S.A



http://www.murr.missouri.edu

University of Missouri Research Reactor

A 10 MW reactor that operates 24 hours a day, seven days a week, 52 weeks a year

165 full time-time employees

In 2014 produced **36 different isotopes** with 1175 shipments to 7 different countries

Each and every week MURR supplies the active ingredients for FDA approved **Quadramet®** and **TheraSpheres®**

MURR Isotope Production Activities

(Industrial and Research) Isotopes Produced in 2014		
Au-198	Ir-192*	Sb-122
Au-199	Kr-79	Sb-124
Ba-131	Mo-99	Sc-46
Ca-45	Na-24	Se-75
Cd-115	P-32	Sm-153*
Ce-141	P-33	Sn-117m
Co-60	Pd-109	Sr-89
Cr-51	Po-210	W-181
Cu-64	Rb-86	Y-90
Fe-59	Re-186	Yb-169
Lu-177*	Ru-103	Zn-65
Hg-203	S-35	Zr-95

MURR Isotope Production Research Activities

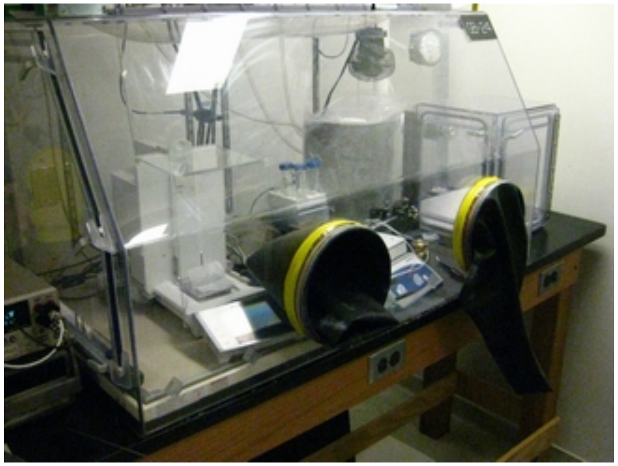
- Carrier free lanthanides
  - Indirect production (**Lu-177**, Pm-149, Ho-166)
    - DOE Advanced Nuclear Medicine Initiative
  - Electromagnetic isotope separation (**Sm-153**)
    - DOE SBIR
- Mo-99
  - n, gamma production for novel generator technologies
    - Industry partnership with **Northstar**
  - fission production with uranium recycle
    - Industry partnership with Northwest Medical Isotopes
- Rh-105
  - Carrier free from uranium fission using selective gas extraction
    - Subcontract with General Atomics/DOE Isotope Program
- Re-186
  - Accelerator production and separations for high specific activity
    - DOE Isotope Program
- As-72, As-77, Cu-67
  - Production of high specific activity with target recycle
    - DOE Isotope Program

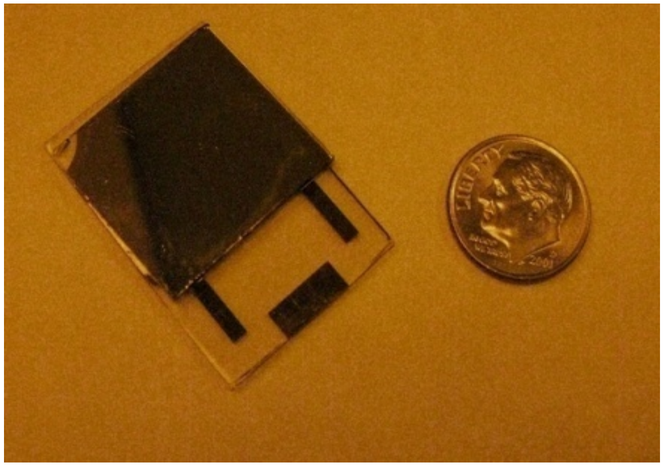
MURR Isotope Production Research Activities

- Po-210
  - Production and incorporation into nuclear batteries
    - Private Industry
- Pm-147, Gd-148
  - Collection of long-lived isotopes from primary cooling loop at FRIB
    - DOE Isotope Program
- Os-191
  - Production and incorporation into device
    - Industry partnership with CheckCap

Near Term DOE Isotope Program Supported Research Priorities

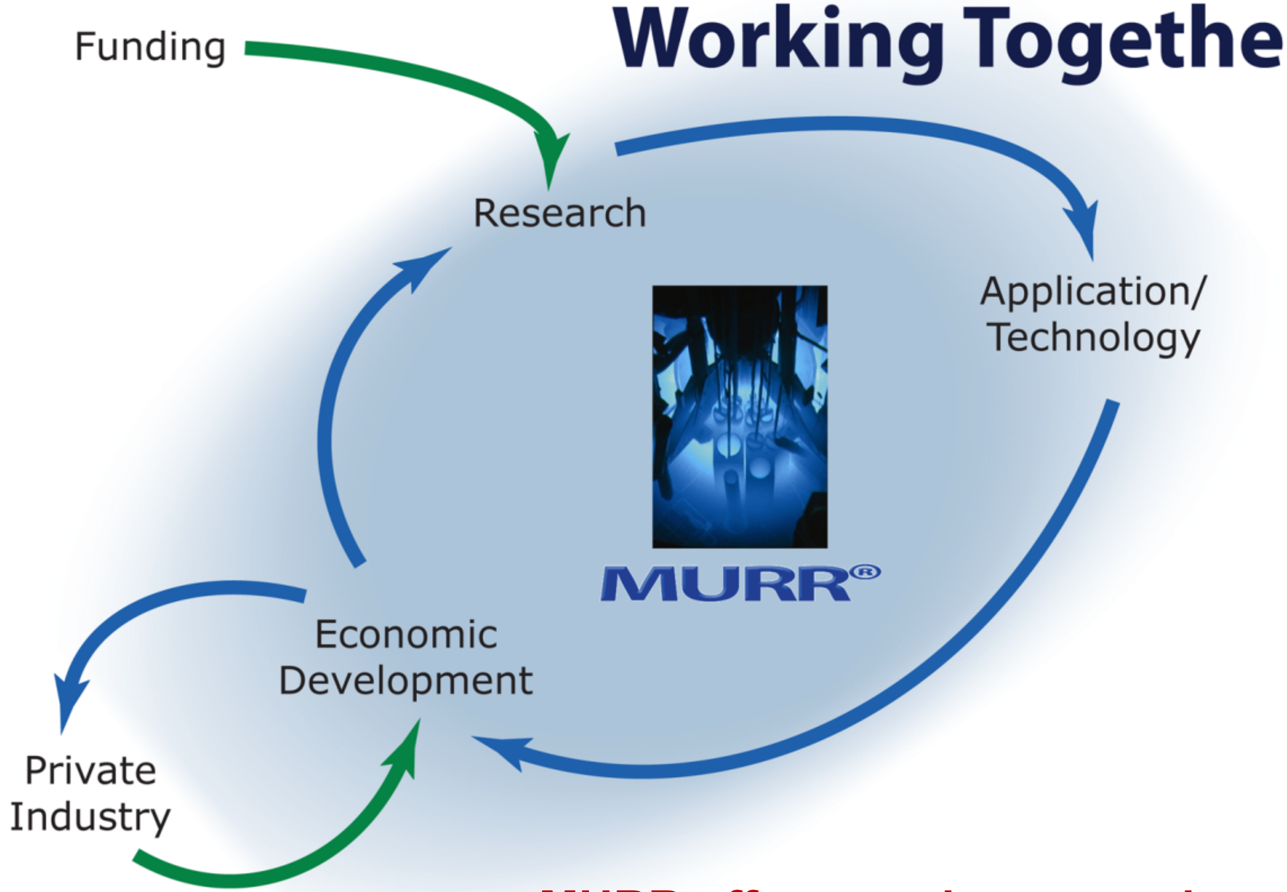
- Radioactive Isotope Separation (**Sm-153** and Sn-117m)
- Production of High Specific Activity As-72, As-77 and Cu-67 for Research and Clinical Applications: Effective design and recycling of targets and radioisotope separation
- Accelerator Production and Separations for High Specific Activity Re-186
- Development of Separation Chemistry for Collection of Long-Lived Radioisotopes (Pm-147 and Gd-148) from the Primary Cooling Loop at FRIB





University of Missouri Research Reactor

Academia and Industry Working Together



MURR offers a unique experience blending capabilities of a national laboratory, an industrial and a university research facility

University of Missouri Research Reactor: A US High-Performance Research Reactor

Facility	Power
Advanced Test Reactor, INL	250 MW
High Flux Isotope Reactor, ORNL	85 MW
Neutron Beam Split-Core Reactor, NIST	20 MW
University of Missouri, Columbia (MURR®)	10 MW*
Massachusetts Institute of Technology	6 MW
University of California-Davis	2 MW
Rhode Island Nuclear Science Center	2 MW
Oregon State University	1 MW
University of Texas, Austin	1 MW
North Carolina State University	1 MW
Pennsylvania State University	1 MW
Texas A&M University	1 MW
University of Massachusetts-Lowell	1 MW

Facility	Power
University of Wisconsin	1 MW
Washington State University	1 MW
Ohio State University	500 kW
Kansas State University	250 kW
Reed College	250 kW
University of California-Irvine	250 kW
University of Maryland	250 kW
University of Missouri, Rolla	200 kW
University of Arizona	100 kW
University of Florida	100 kW
University of Utah	100 kW
Purdue University	1 kW
Idaho State University	5 W
University Of New Mexico	5 W
Rensselaer Polytechnic Institute	1 W

University of Missouri Research Reactor : Basic Reactor Parameters

The MURR® is a **pressurized**, **heterogeneous reflected**, **open pool-type**, which is light-water moderated and cooled

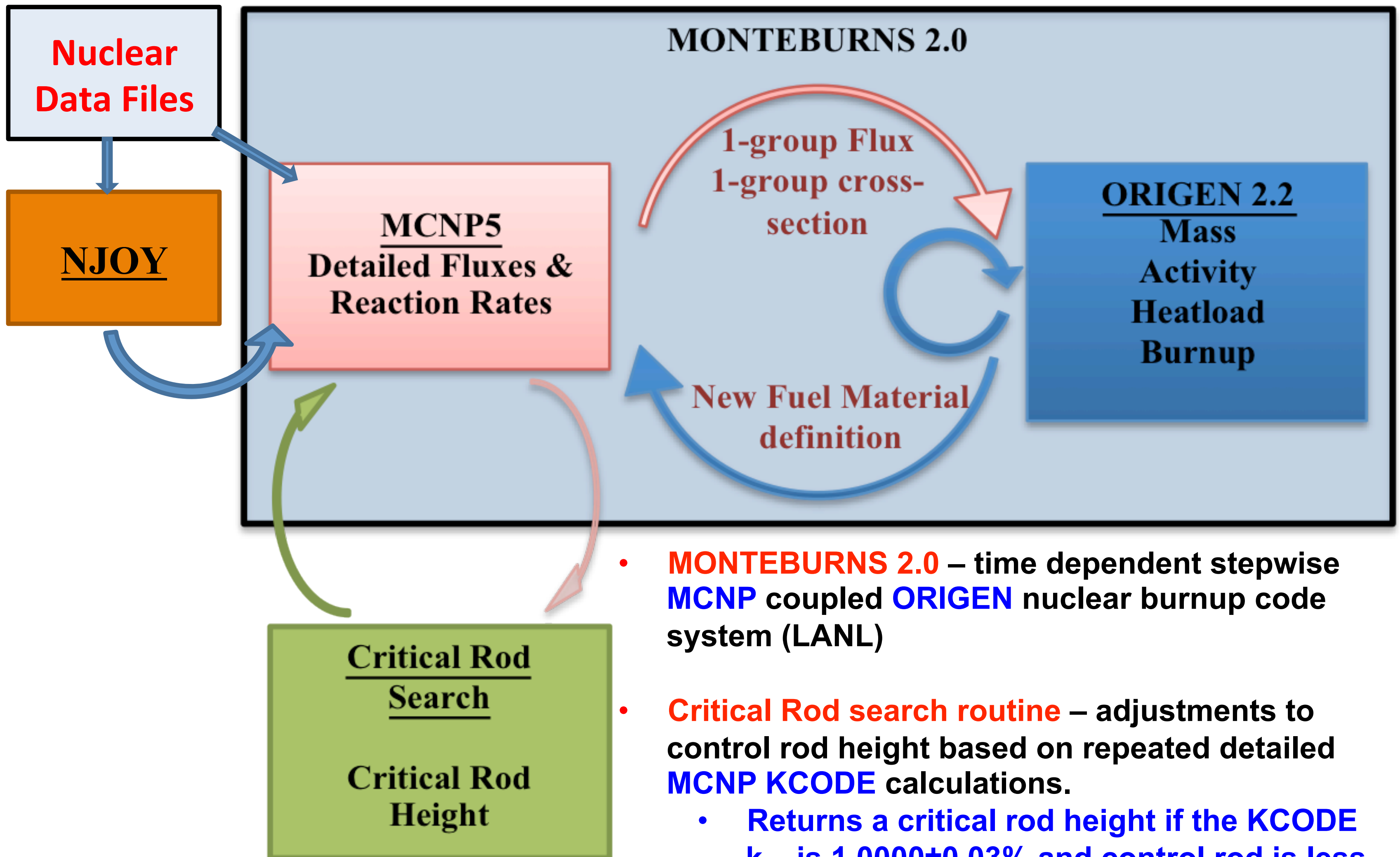
- Maximum power – **10 MW$_{th}$**
- **Peak flux in center test hole – 6.0E14 n/cm^2-s***
- Core – **8 HEU fuel assemblies**
- Excess reactivity control blades – **5 total: 4 BORAL® shim-safety, 1 SS regulating**
- Reflectors – **beryllium and graphite**
- Forced primary coolant flow rate – **3,750 gpm (237 lps)**
- Forced pool coolant flow rate – **1,200 gpm (76 lps)**
- Primary coolant temps – **120 °F (49 °C) inlet, 136 °F (58 °C) outlet**
- Primary coolant system pressure – **85 psia (586 kPa)**
- Pool coolant temps – **100 °F (38 °C) inlet, 106 °F (41 °C) outlet**
- Beamports – **three 4-inch (10 cm), three 6-inch (15 cm)**

University of Missouri Research Reactor: Core Models and Simulations

- **Use of state-of-the-art code systems and nuclear data libraries to develop high-confidence MURR core models and simulations**
  - **Key quantities to be predicted: neutron and gamma fluxes distributions**

- **Benchmarked MURR models and simulations are used for:**
  - **Predicting accurate reactor-core physics parameters for routine reactor operations (e.g, critical rod positions, excess core reactivity and worth's of samples and experiments etc.)**
    - **Nuclear data handling: NJOY**
    - **Irradiation transport and nuclear inventories: (MCNP, ORIGEN, MONTEBURNS)**
  - **Isotopes production (activity and heat generation predictions)**
    - **Nuclear data handling: NJOY**
    - **Irradiation transport and nuclear inventories: (MCNP/MCNPX, ORIGEN, MONTEBURNS)**
  - **HEU to LEU fuel conversion feasibility studies for the DoE's Material Management and Minimization Reactor Conversion Program**
    - **Nuclear data handling: WIMS-ANL, NJOY**
    - **Irradiation transport and nuclear inventories: (MCNP/MCNPX, Diff-3D, ORIGEN, REBUS, MONTEBURNS)**

University of Missouri Research Reactor Computational Methodology



- **MONTEBURNS 2.0 – time dependent stepwise MCNP coupled ORIGEN nuclear burnup code system (LANL)**
- **Critical Rod search routine – adjustments to control rod height based on repeated detailed MCNP KCODE calculations.**
  - **Returns a critical rod height if the KCODE k_{eff} is 1.0000±0.03% and control rod is less than maximum travel**

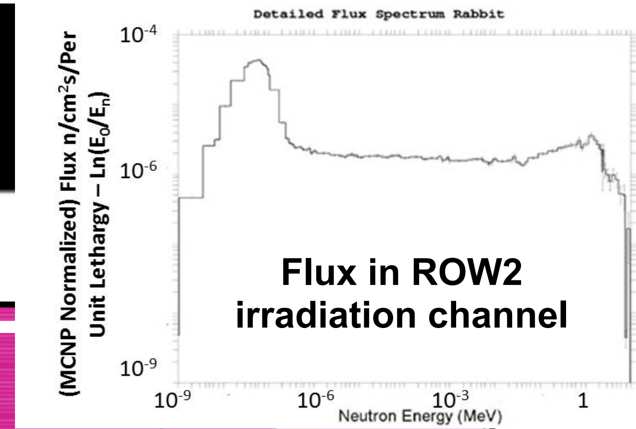
Detailed MCNP5 Steady-State MURR Core Model

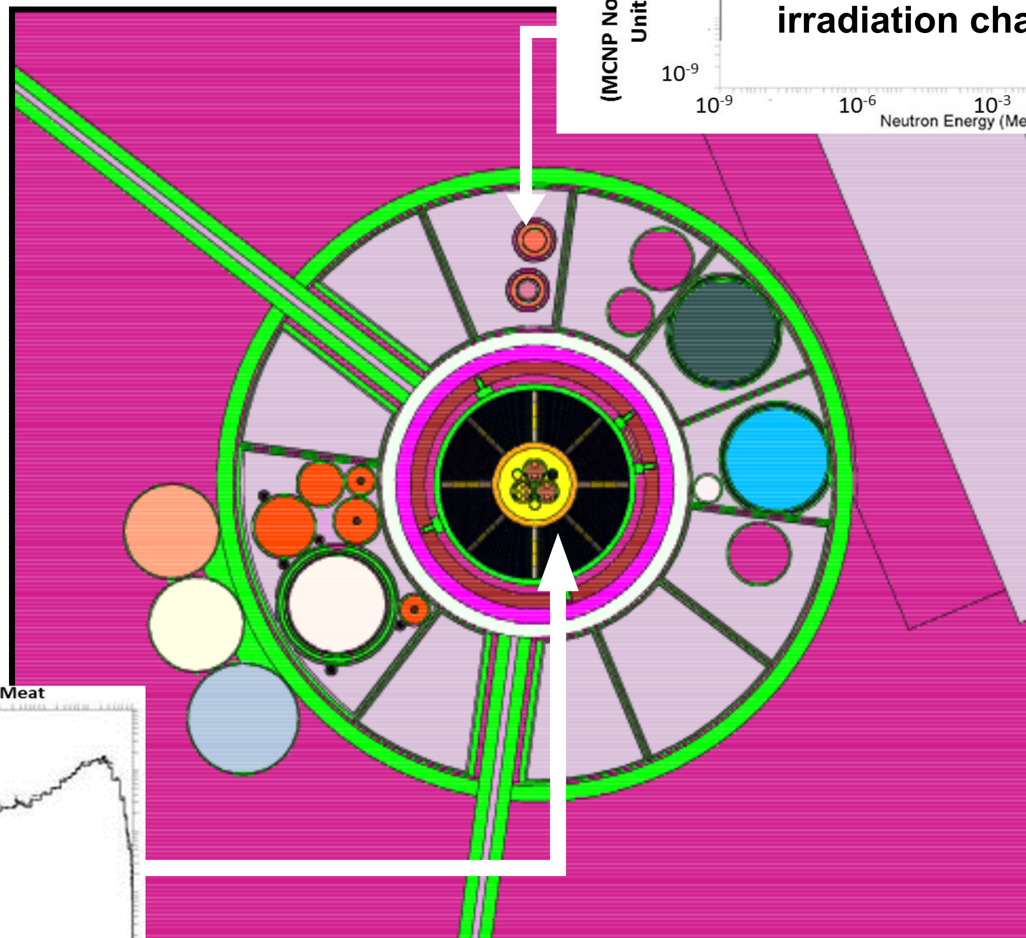
KCODE k_{eff} convergence prediction: *= 1.00043 ± 0.000028*

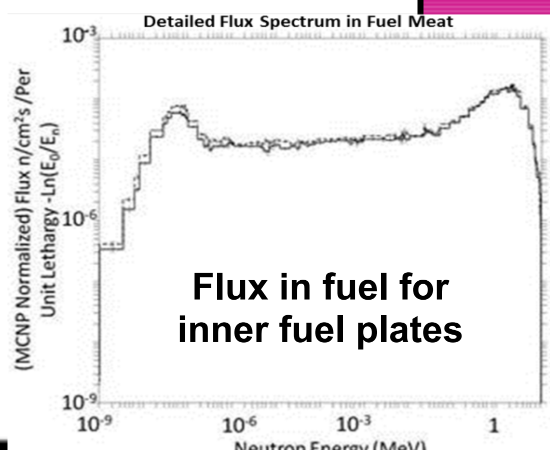
Source particle statistics: 50 million – 2 billion

Compute detailed flux spectrum (*φ (E)*) at any location in reactor

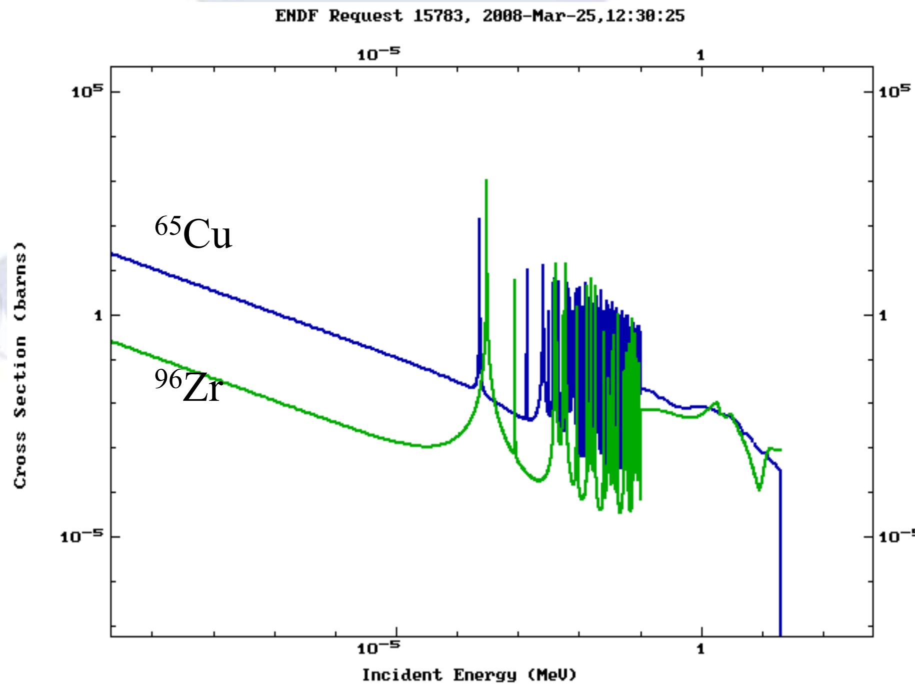
An accurate *φ (E)* is essential for predicting response functions including activation rates, and energy deposition rates







Predicting Activation Rates in ROW2 of the MURR Model



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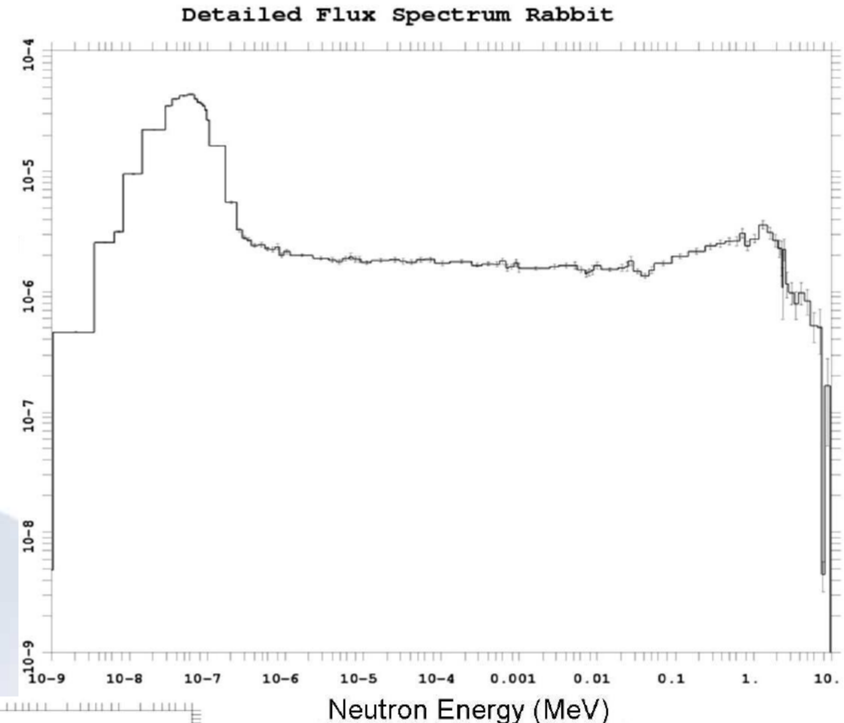
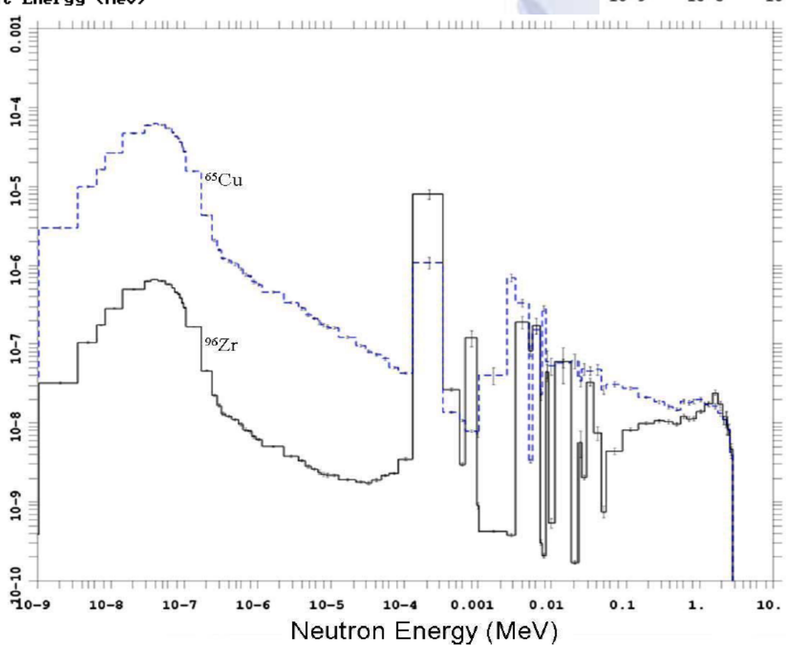


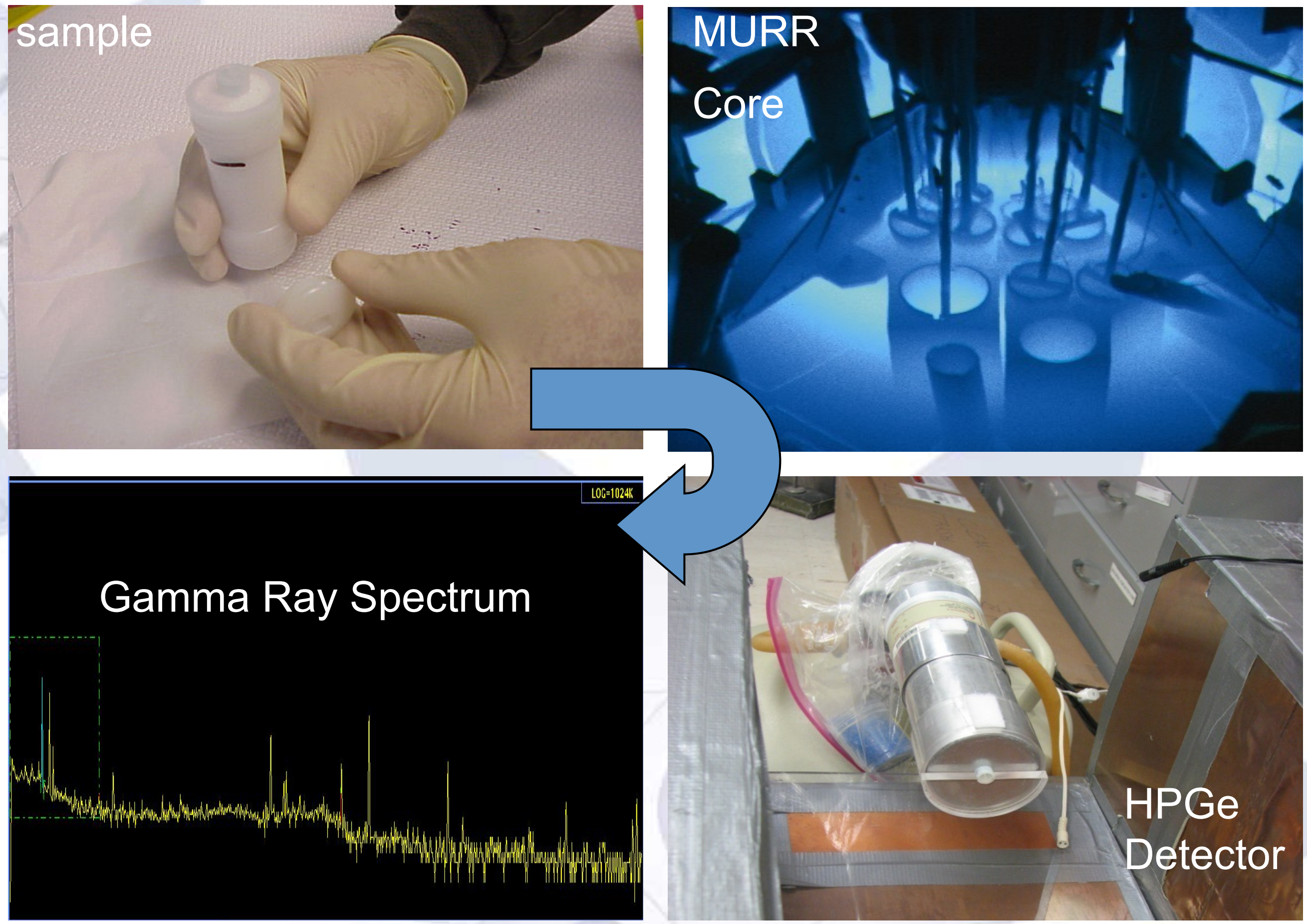


$$Rate = \int_{E1}^{E2} \sigma_{capture}(E)\phi(E)\, dE$$

Evaluated Nuclear Data File (ENDF), Database Version 04/07 http://www-nds.iaea.org/exfor/endf00.htm.

Benchmarking Measurements in ROW2

- **Activation experiments:**
  - Targets made from dilute single-elements standards to benchmark reaction rates
  - Targets made from NIST SRM to predict concentration
- **Sample prep:**
  - Gravimetric
  - Dry matter corrections (were done where necessary).
- **Irradiations:**
  - Sequentially; on the same day over 3 weeks
  - times range from 1 min – 1 hour in ROW2
  - decay times vary from 2 min - 1 week.
- **Counting:**
  - Standard gamma ray spectroscopy (HPGe)
  - Westphal and Live time correction used
  - Counting times vary so that the uncertainty on the counts ~1%.

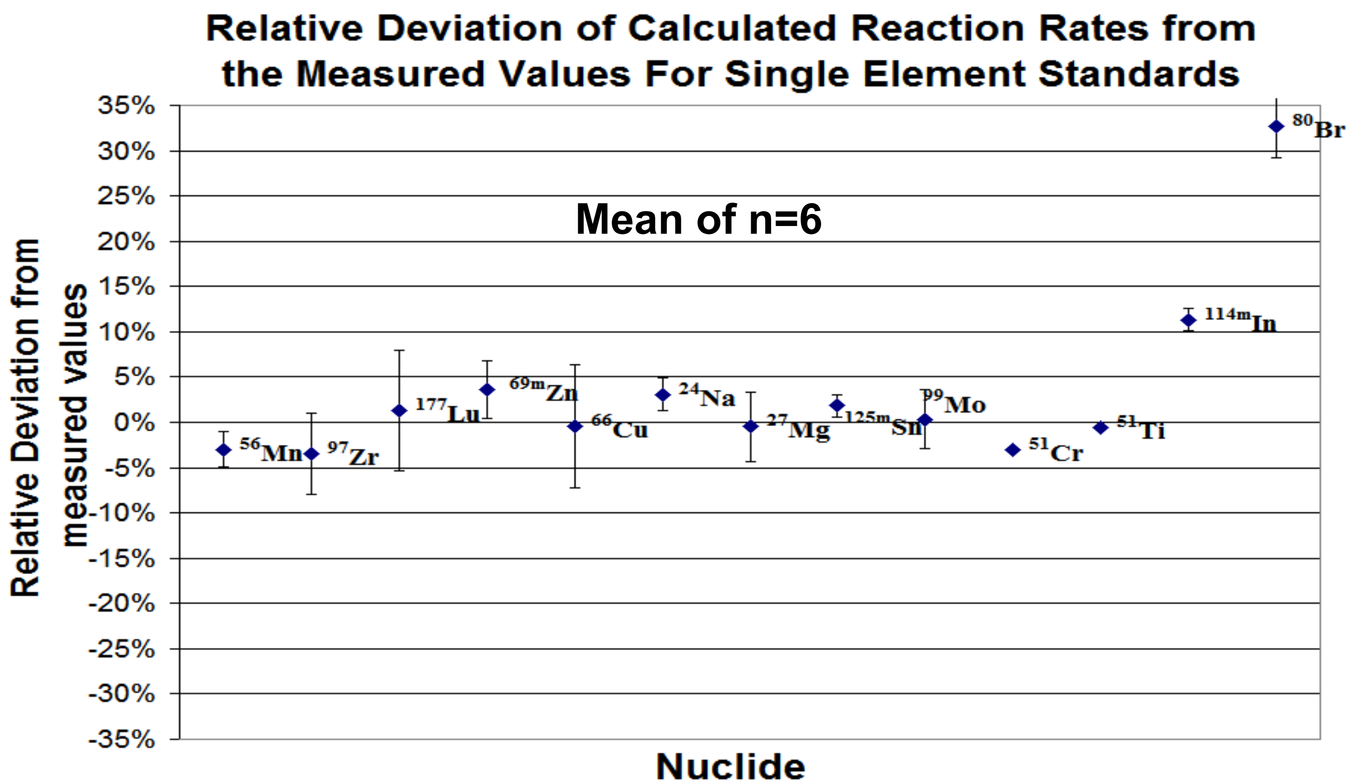


$$Rate = \overline{\sigma_c \phi} = \frac{R_c}{I \varepsilon n_t SDC}(C_{BR})$$

R_c – measured count rate (cps) I – gamma intensity
ε – detector efficiency n_t – number of targets nuclides
C_{BR} – correction for branching decay
S, D and C – corrections for Saturation, Decay and Counting, respectively.

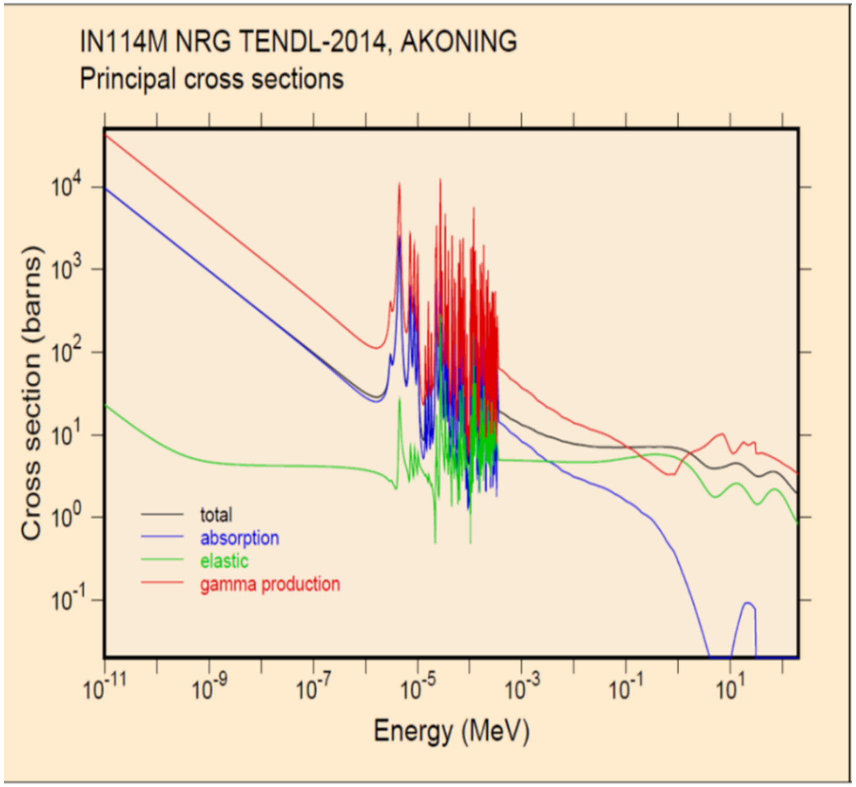
Benchmarking MURR Core ROW2 Flux Spectrum Using Intrinsic Reaction Rates

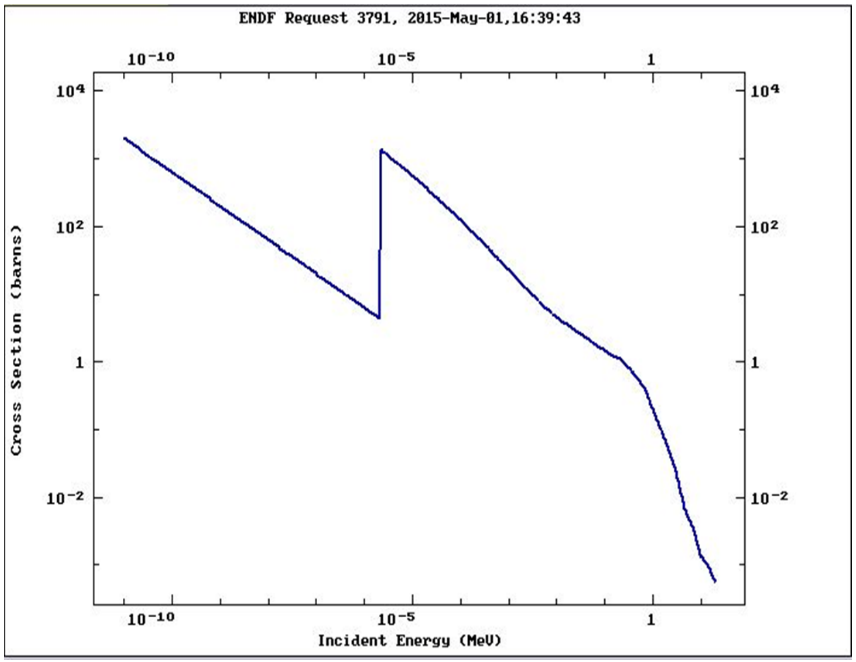
$$Rate = \int_{E1}^{E2} \sigma_{capture}(E)\phi(E)\, dE = \overline{\sigma_c\, \varphi}$$



- **Publication :** **N.J. Peters**, J.D. Brockman, J.D. Robertson, **"Using Monte Carlo Transport to accurately predict isotope production and activation analysis rates at the University of Missouri research reactor" J. Radioanal. N. Chem. 282: 255-259 (2009)**
- **MCNP predicted reaction rates used in new methodology in INAA, publication: N.J. Peters, J.D. Brockman, J.D. Robertson, "A new approach to single-comparator instrumental neutron activation analysis" J. Radioanal. N. Chem. 291(2): 467-472 (2011)**

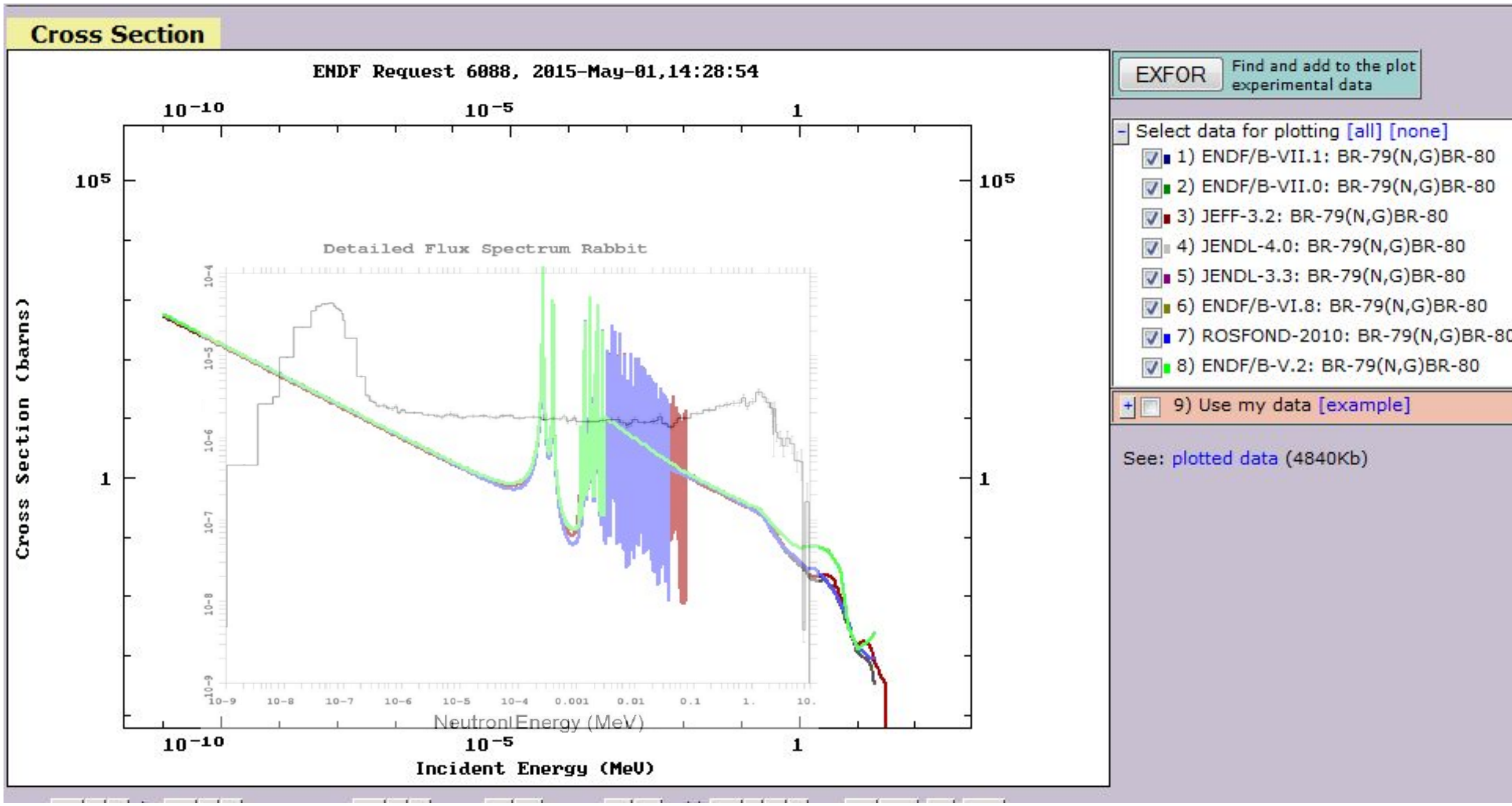
In-114(n, ϒ)In-114m Excitation Function: Accuracy and Consistency





http://www.nndc.bnl.gov/exfor/endf00.jsp
ftp://ftp.nrg.eu/pub/www/talys/tendl2014/neutron_html/neutron.html

Br-79(n, ϒ)Br-80 Excitation Function: Data Sets Accuracy and Consistency

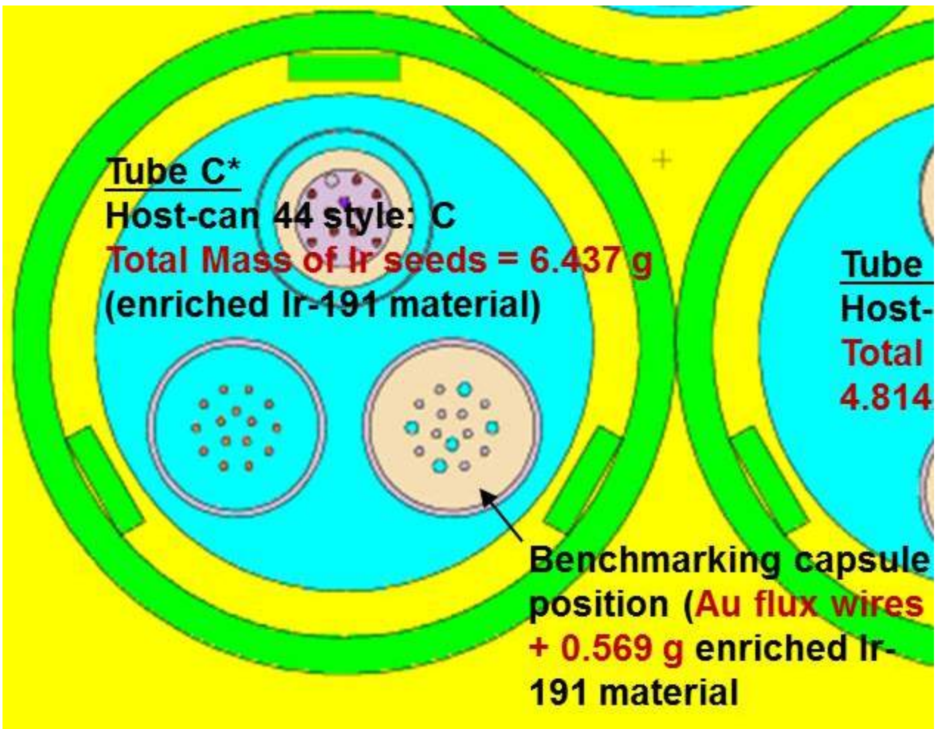


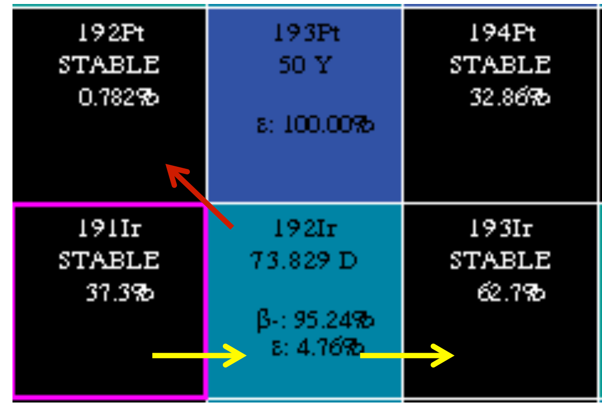
Is the excitation function Br-79(n, ϒ)Br-80 incorrect?

http://www.nndc.bnl.gov/exfor/endf00.jsp

Predicting Activity Production of Highly Absorbing Material: Enriched Ir-191 Targets

MCNP5 Iridium Flux trap Model



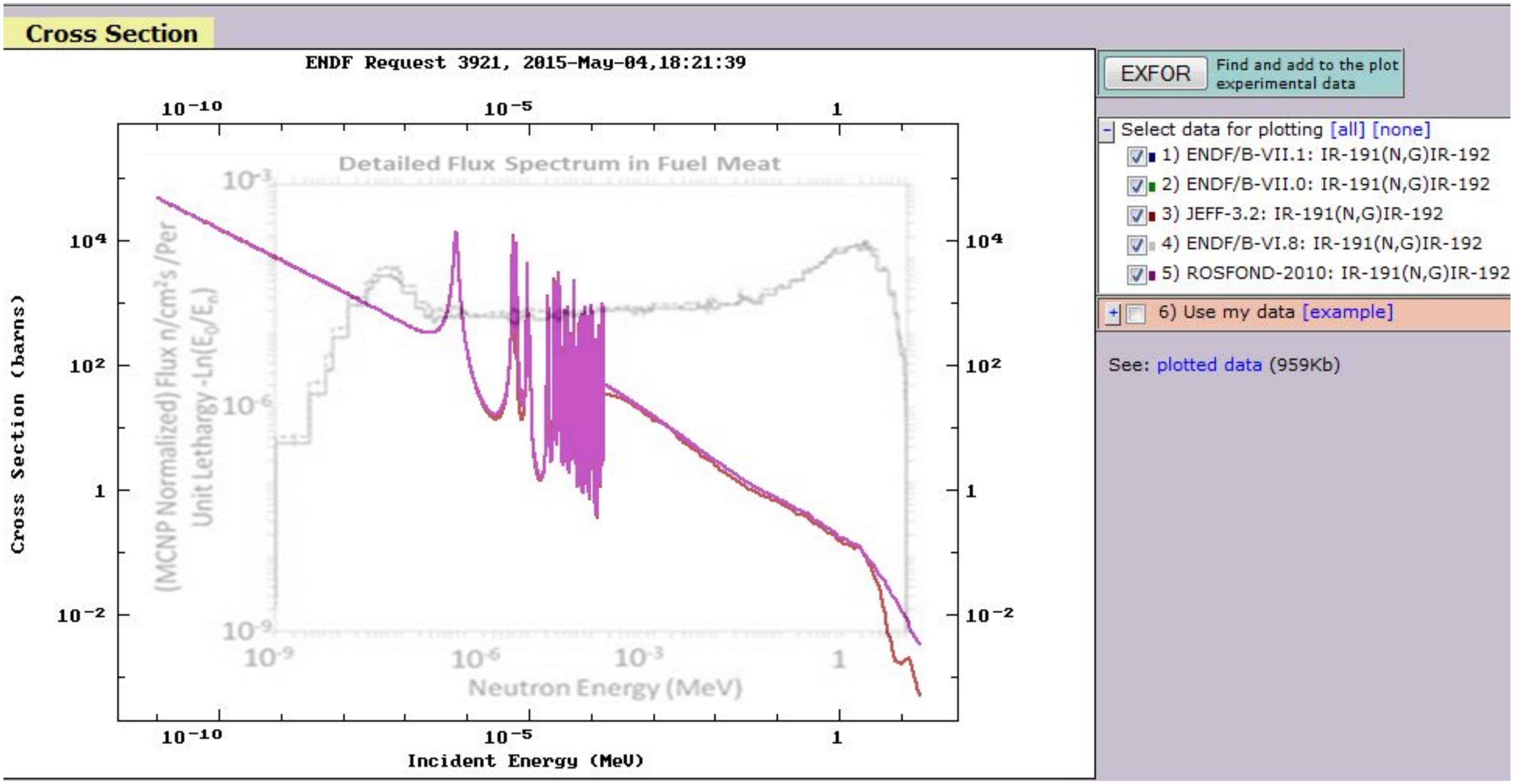


- **Irradiation simulation:**
  MURR MCNP5 KCODE model linked to ORIGEN2.2 isotope generation/depletion method - MONTEBURNS

- **Continuous energy data libraries: Ir-191 and Ir-192**

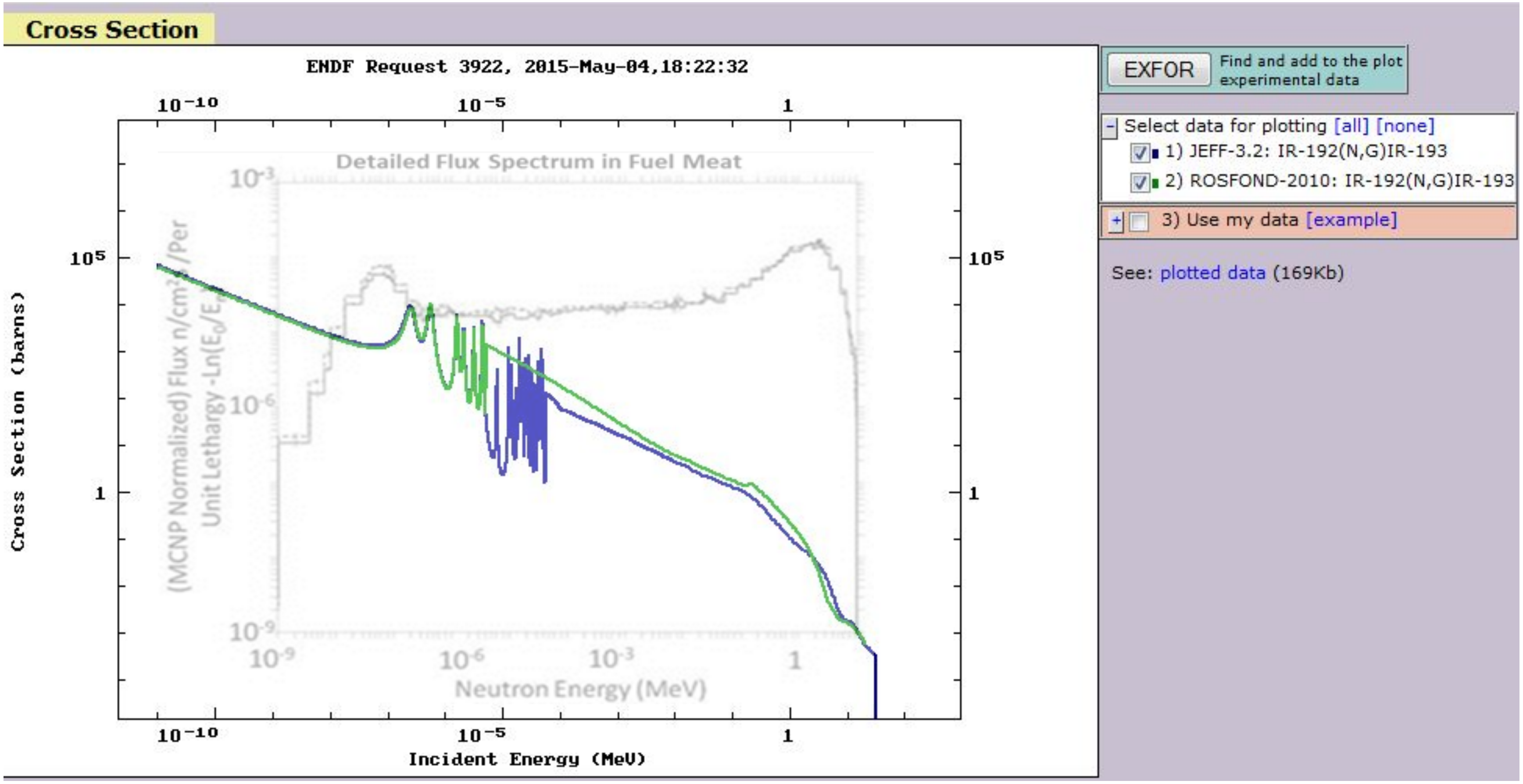
http://www.nndc.bnl.gov/nudat2/

Ir-191(n, ϒ)Ir-192 Excitation Function: Data Sets Accuracy and Consistency



http://www.nndc.bnl.gov/exfor/endf00.jsp

Ir-192(n, ϒ)Ir-193 Excitation Function: Data Sets Accuracy and Consistency

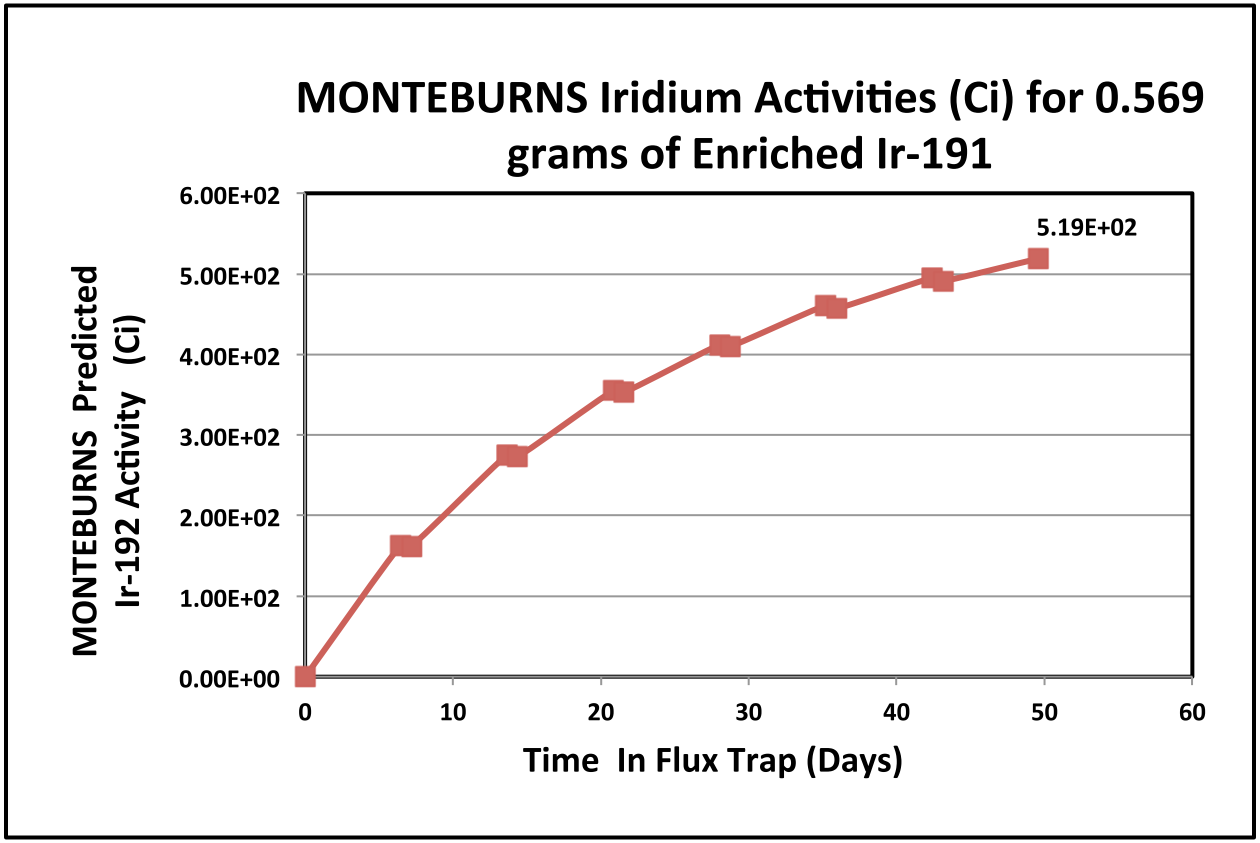


http://www.nndc.bnl.gov/exfor/endf00.jsp

Ir-192 Activity Prediction using MCNP–ORIGEN (MONTEBURNS) Simulations

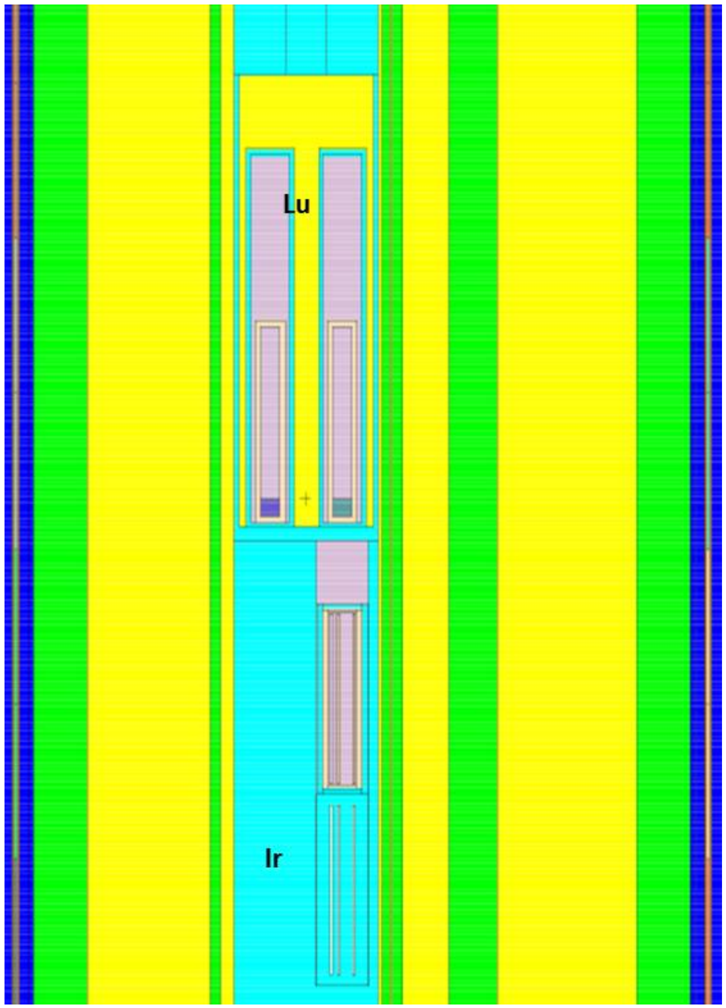
- 7-week irradiation of 0.569 grams of enriched Ir-191 in the MURR core flux trap configuration
- Ir-192 activity prediction: simulated using the ENDF /BVII.0 and ROSFOND data set for Ir-191 and Ir-192, respectively

	Totral Ir sample Mass grams	^{192}Ir EOI Activity Ci
Measured	0.5690	531.06
MONTEBURNS	0.56897	519.00
% Devation	-0.01%	-2.27%



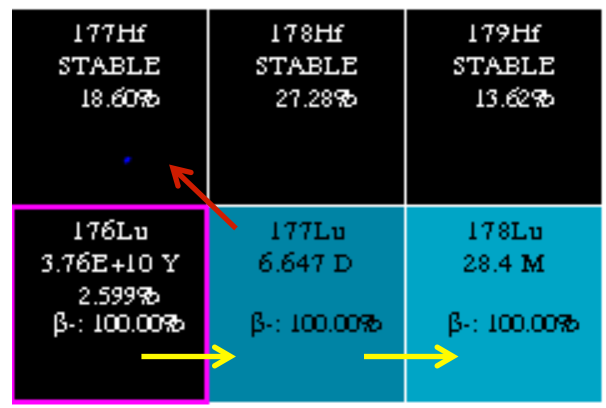
Modeling Activity Production Suppression due to Neighboring Strongly Absorbing Material

MCNP5 Lutetium/Iridium Flux trap Configuration



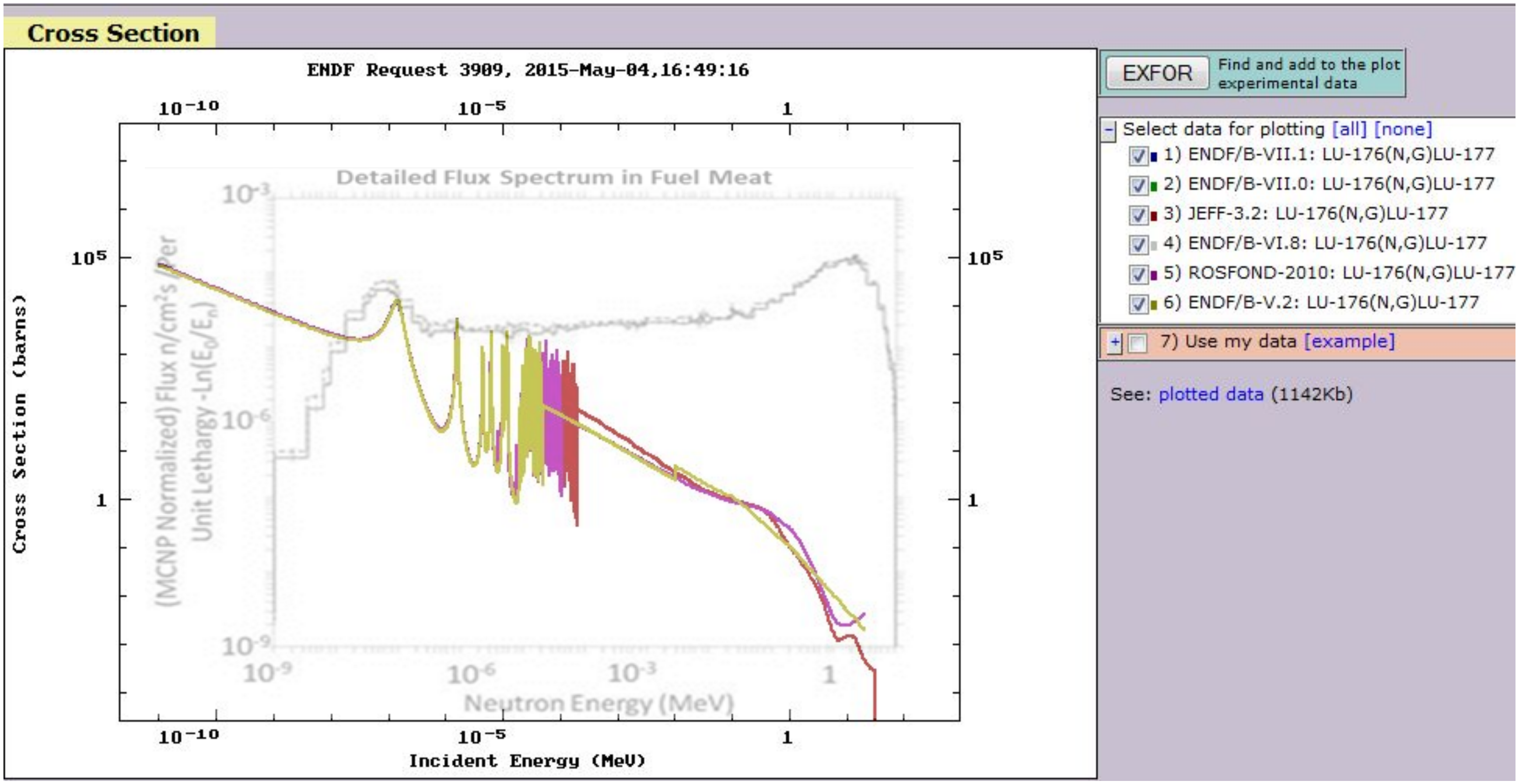
- **MONTEBURNS predicted Lu-177 activity suppression due to neighboring enriched Ir-191 targets**

- **Necessary cross-section data libraries for accurate prediction : Lu-176 and Lu-177**



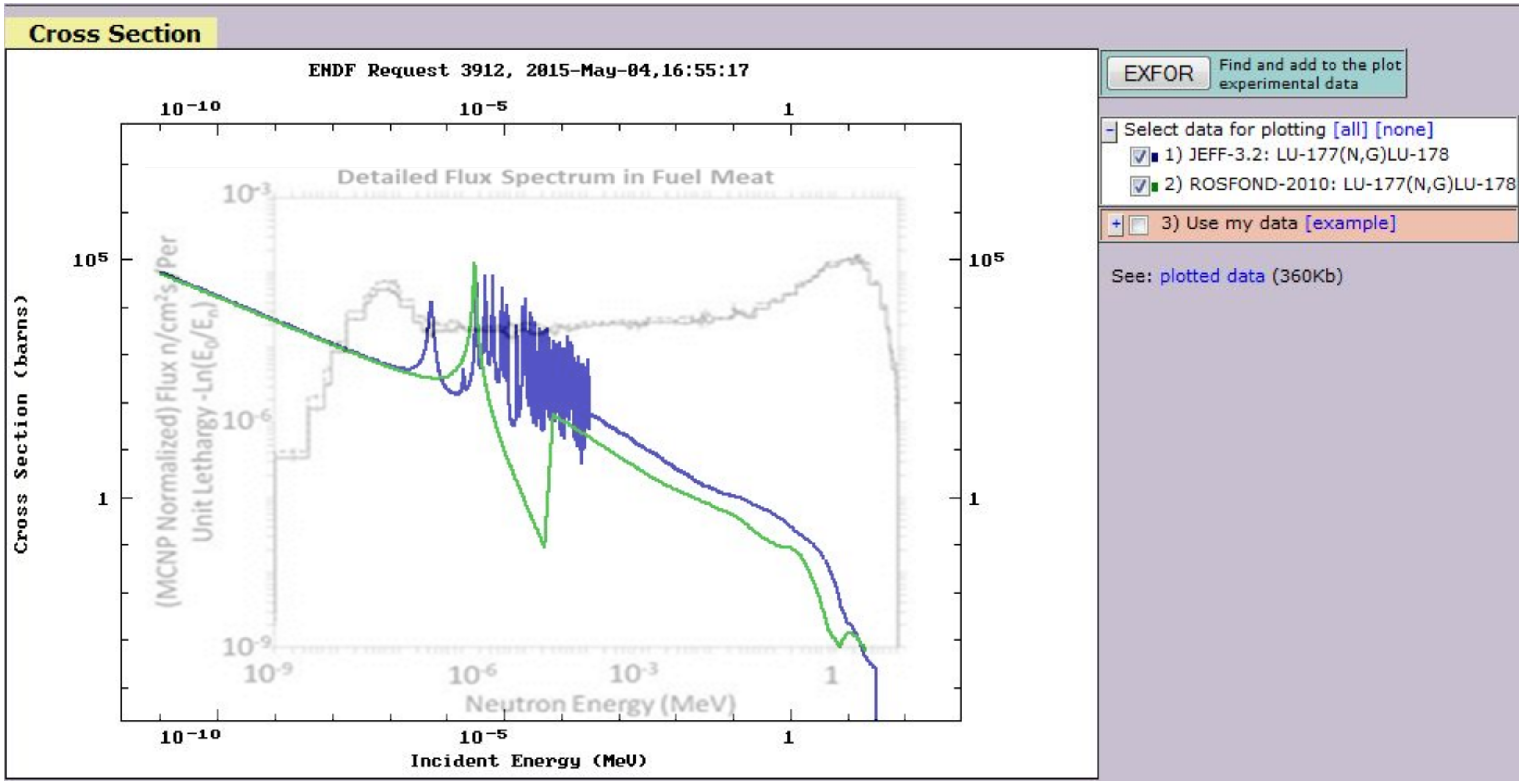
http://www.nndc.bnl.gov/nudat2/

Lu-176(n, ϒ)Lu-177 Excitation Function: Data Sets Accuracy and Consistency



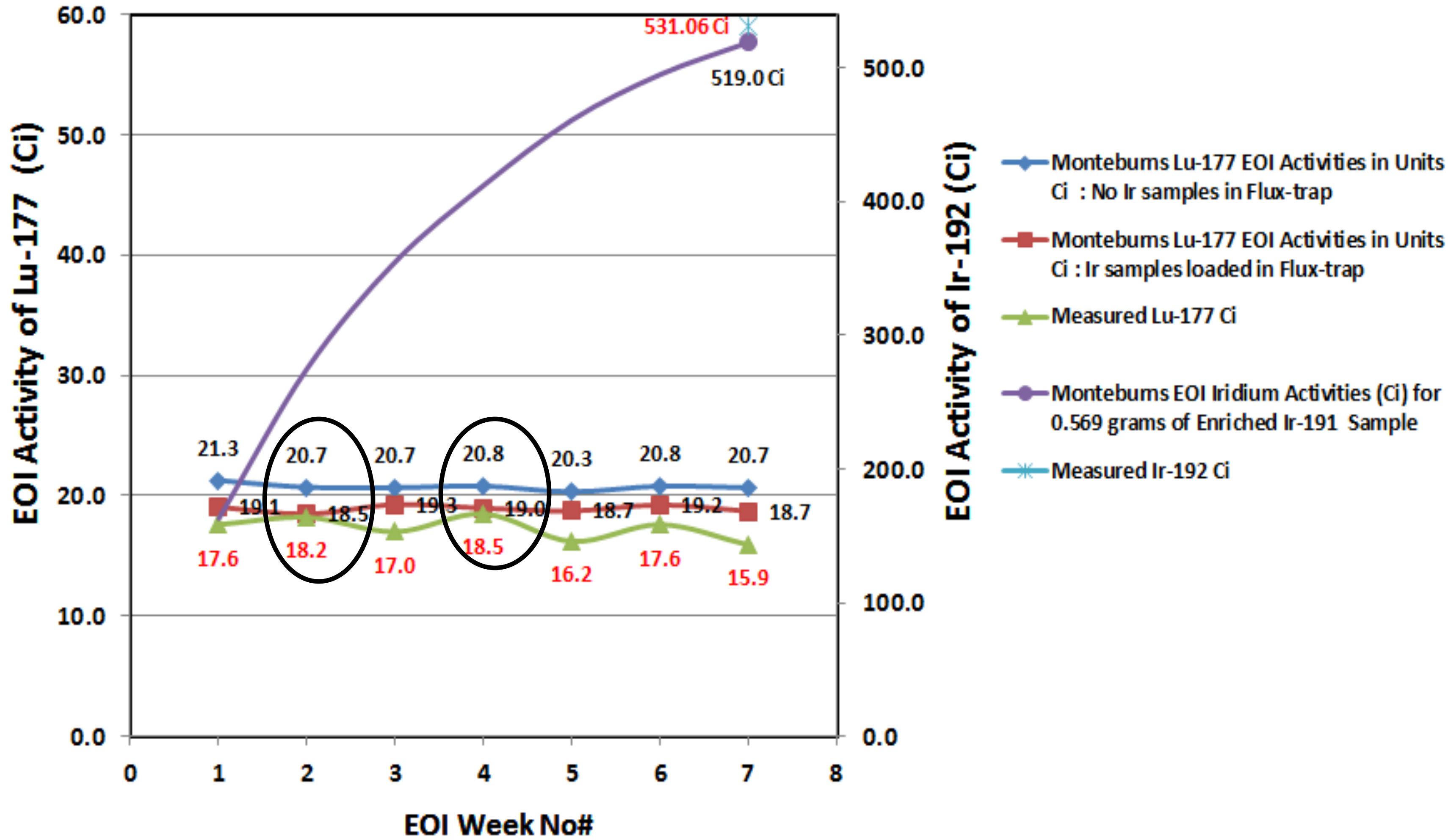
http://www.nndc.bnl.gov/exfor/endf00.jsp

Lu-177(n, ϒ)Lu-178 Excitation Function: Data Sets Accuracy and Consistency



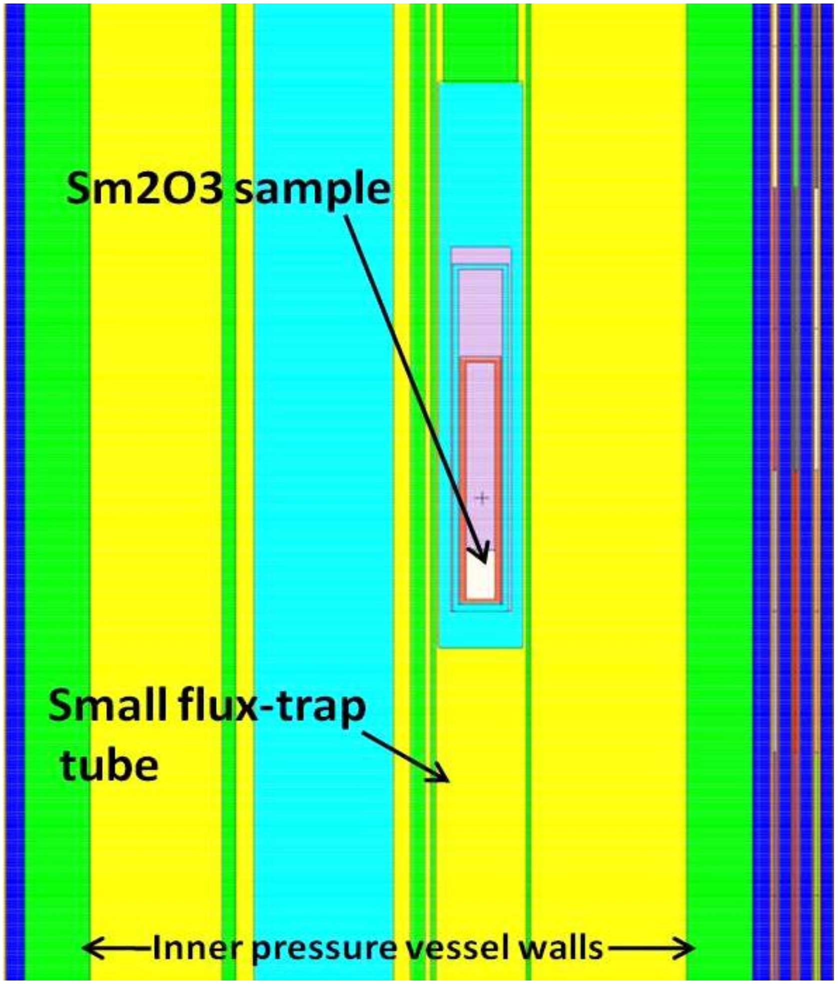
http://www.nndc.bnl.gov/exfor/endf00.jsp

Predicted Vs. Measured EOI Activities for Lu-177 and Ir-192 for Irradiation Period 5/31/2010 -7/19/2010

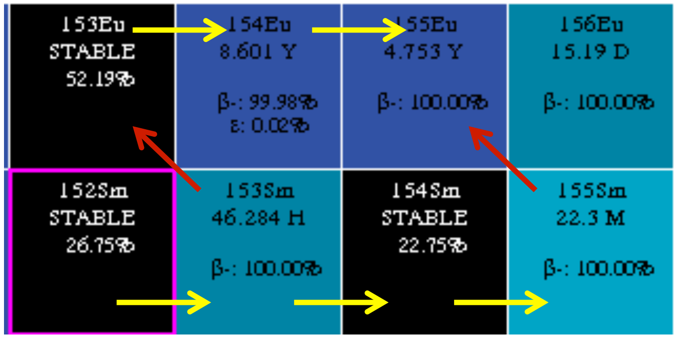


Irradiation Simulations for Studying Radionuclide Contamination in Products

MCNP5 Samarium Flux trap Configuration



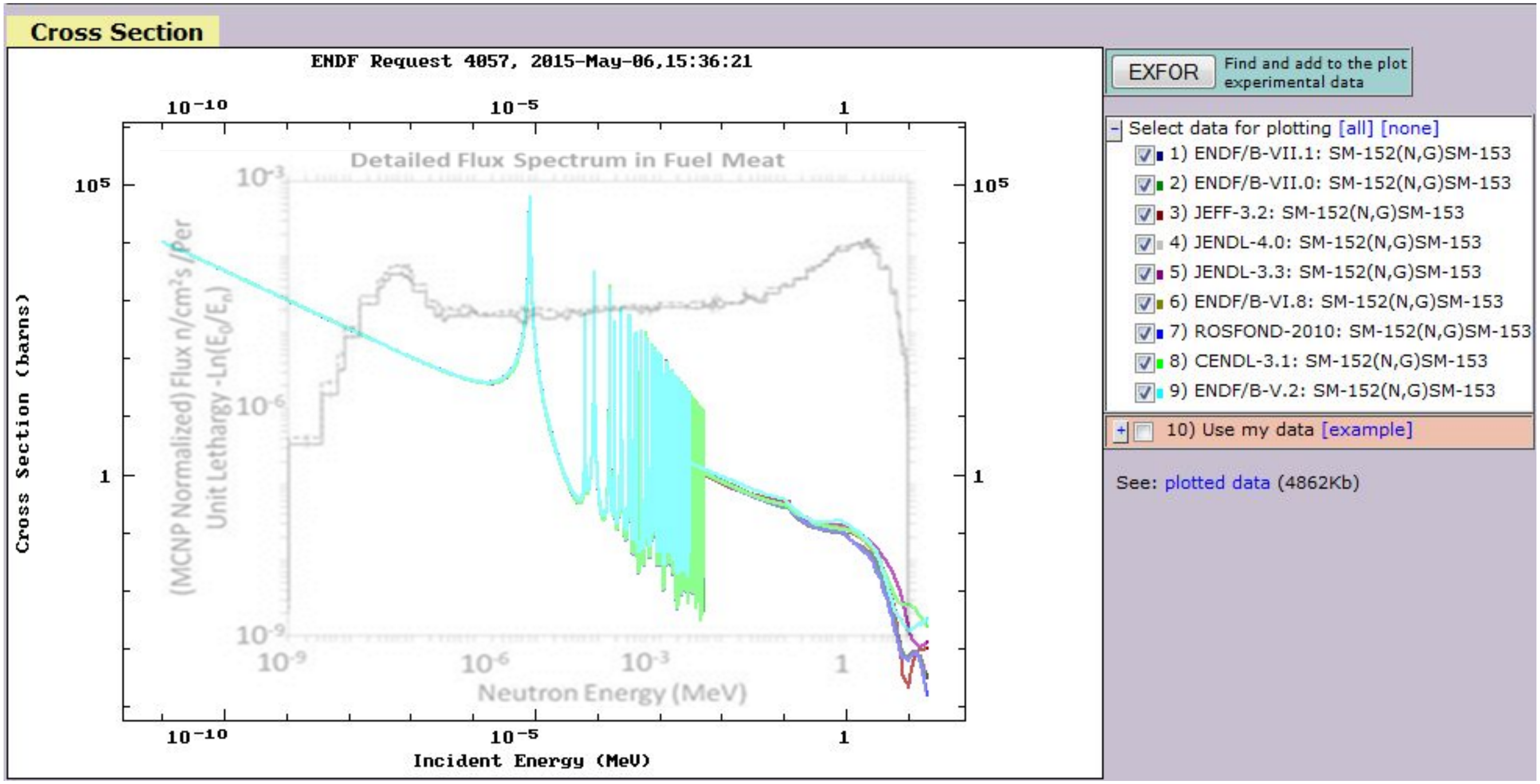
- **Prediction of Eu-154/155 contamination in product Sm-153 as product to contaminant activity ratio**



- **Necessary cross-section data libraries for accurate prediction : Sm-152, Sm-153, Eu-154 and Eu-155**

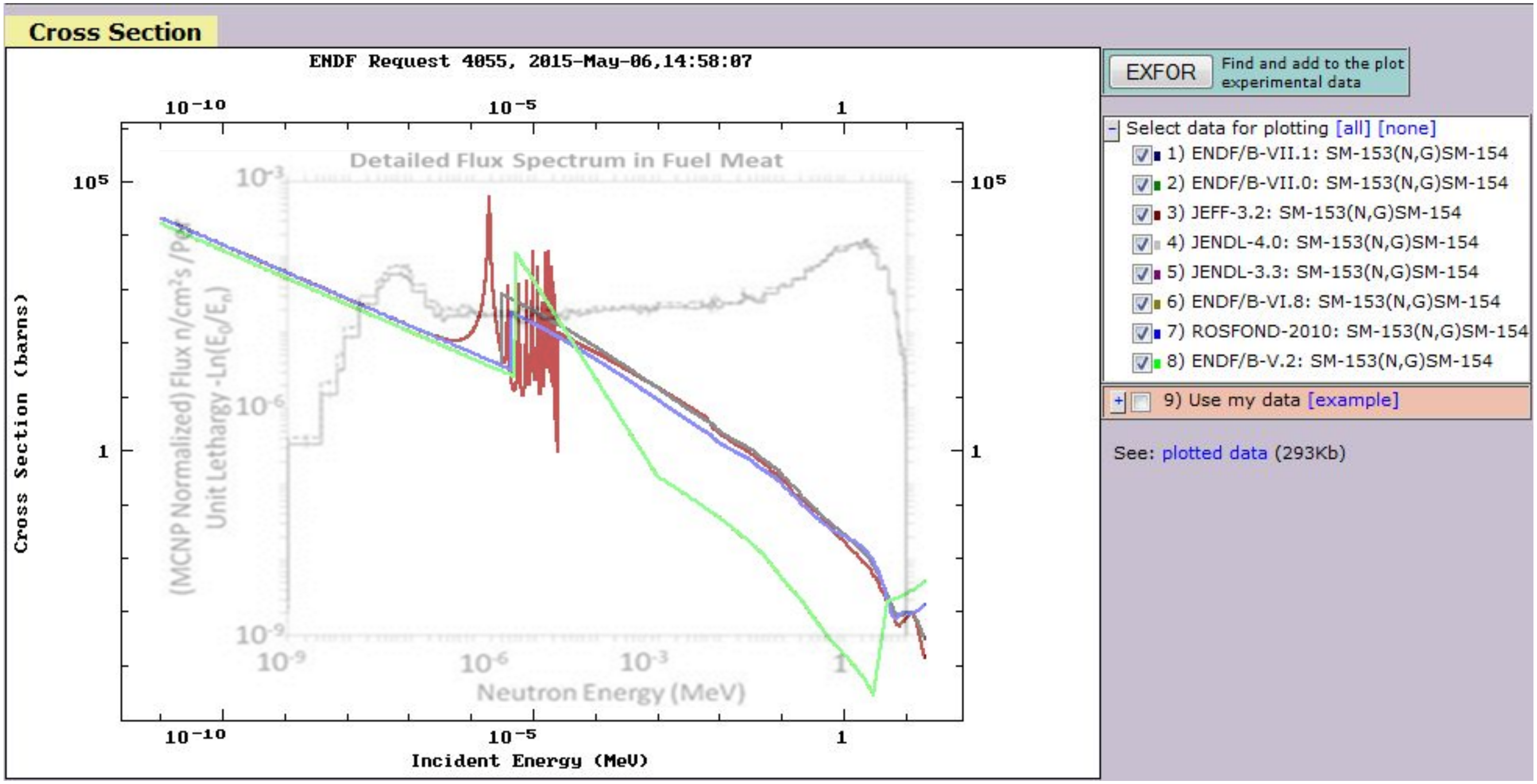
http://www.nndc.bnl.gov/nudat2/

Sm-152(n, ϒ)Sm-153 Excitation Function: Data Set Accuracy and Consistency



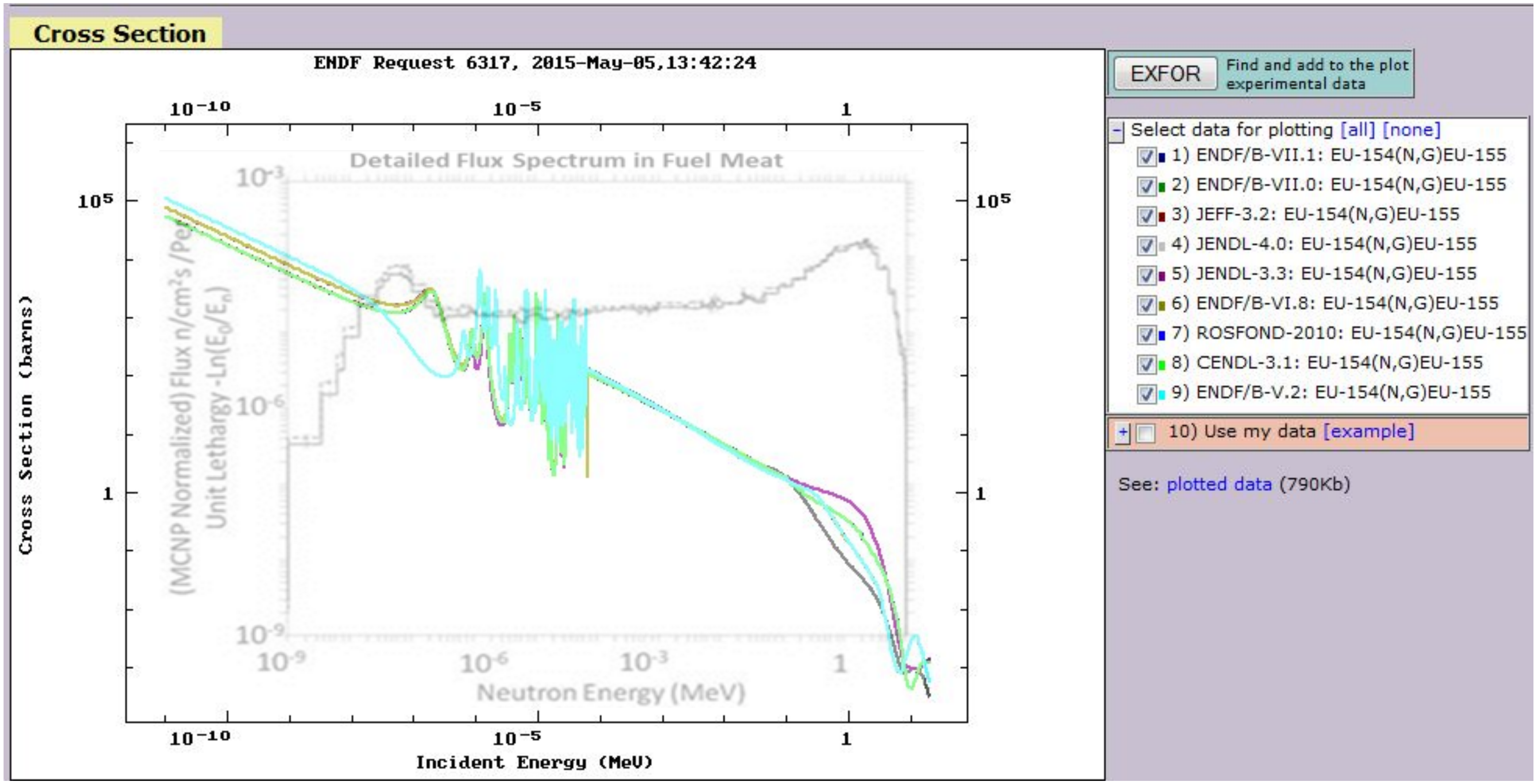
http://www.nndc.bnl.gov/exfor/endf00.jsp

Sm-153(n, ϒ)Sm-154 Excitation Function: Data Set Accuracy and Consistency



http://www.nndc.bnl.gov/exfor/endf00.jsp

Eu-154(n, ϒ)Eu-155 Excitation Function: Data Set Accuracy and Consistency



http://www.nndc.bnl.gov/exfor/endf00.jsp

Predicted Vs. Measured EOI Activities for Sm-153 and Eu-154

Mass of one Sm2O3 sample 0.0414 g **Irradtion Time: 151.49 hours**

Measured EOI Activity for x 2 sample **EOI Date: 10/22/2007**

Sm-153 = 351 Ci Eu-154 = 0.004525 Ci

MONTEBURNS EOI Activity for x 2 sample

Sm-153 = 352 Ci Eu-154 = 0.00544 Ci

Irr. Time Days	MB Activity Ci/g Sm-153	MB Activity Ci/g Eu-154	MB Activity Ci/g Eu-155	MB Activity Ratio A_{RN}
6.31	4.25E+03	6.57E-02	9.59E-03	5.65E+04
Irr. Time Days	**Measured Ci/g Sm-153**	**Measured Ci/g Eu-154**	**Measured Ci/g Eu-155**	**Measured Actvity Ratio A_{RN}**
6.31	4.24E+03	5.46E-02	N/A	7.76E+04

Predicting the Energy Deposition from Radiative Capture: Consistency and Accuracy

- **The spatial distribution of the energy deposited by gammas produced from radiative capture should be properly predicted for:**
  - **target heat generation during irradiation for isotope production**
  - **determining the effective energy release from new reactor fuel e.g., U-10Mo monolithic LEU fuel for RERTR conversion)**

- **MCNP uses the Energy-balance methodology to predict spatial energy deposition distributions**

- **The accuracy of Energy-balance methodology is limited by the integrity of the nuclear data set**

- **What is the Energy-balance Method?**
  - Energy conservation given as:

$$H(E)_n = E - \sum_i p_i(E)[E_{i,out}(E) - Q_i + E_{i,\gamma}(E)]$$

Here, E = is the incident energy

$p(E)_i$ = probability of reaction i at energy E

$E_{i\,out}(E)$ = average exiting particle energy for reaction i at neutron energy E

$E_{i\gamma}(E)$ = average exiting gamma energy for reaction i at neutron energy E

Q_i = Q-value of reaction i

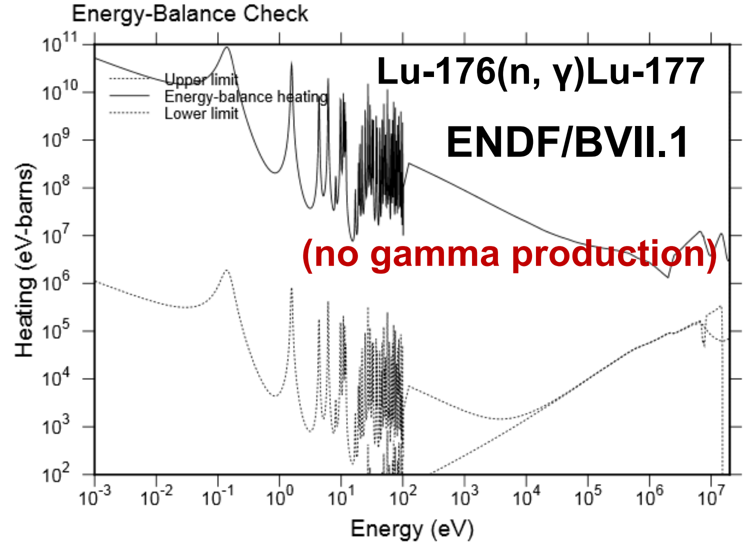
  - **The energy release of a reaction (H_i) is shown to depend on its excitation function ($\sigma(E)$)**

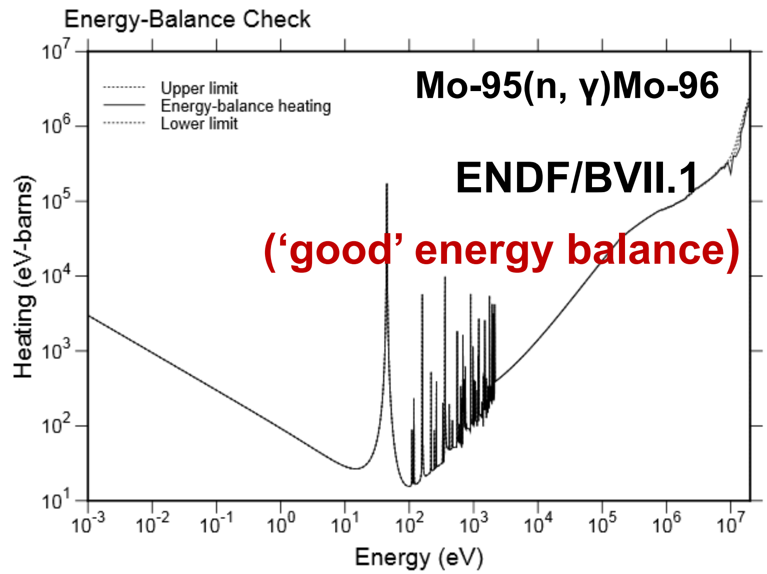
NJOY Energy Balance Checks for the ENDF/BVII.1

Major problems in the accuracy of the energy-balance method predictions are associated with gamma production data.

- **Known issues:**
  - **For small systems (relative to the mean free path of gammas), and where there are no gamma production data, the heating can be largely over predicted**
  - **Excessively large gamma production can under predict heating**
- **New issue:**
  - **Accuracy of heating depends on neutron spectrum energy cut-off**

ENDF Data consistency checked using NJOY (for Lu-176 and Mo-95 neutron capture)

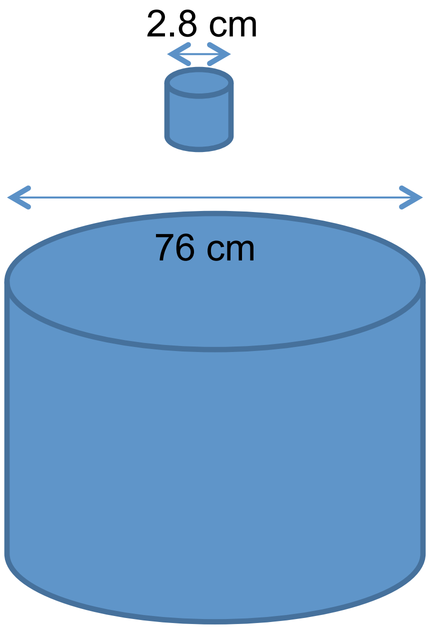


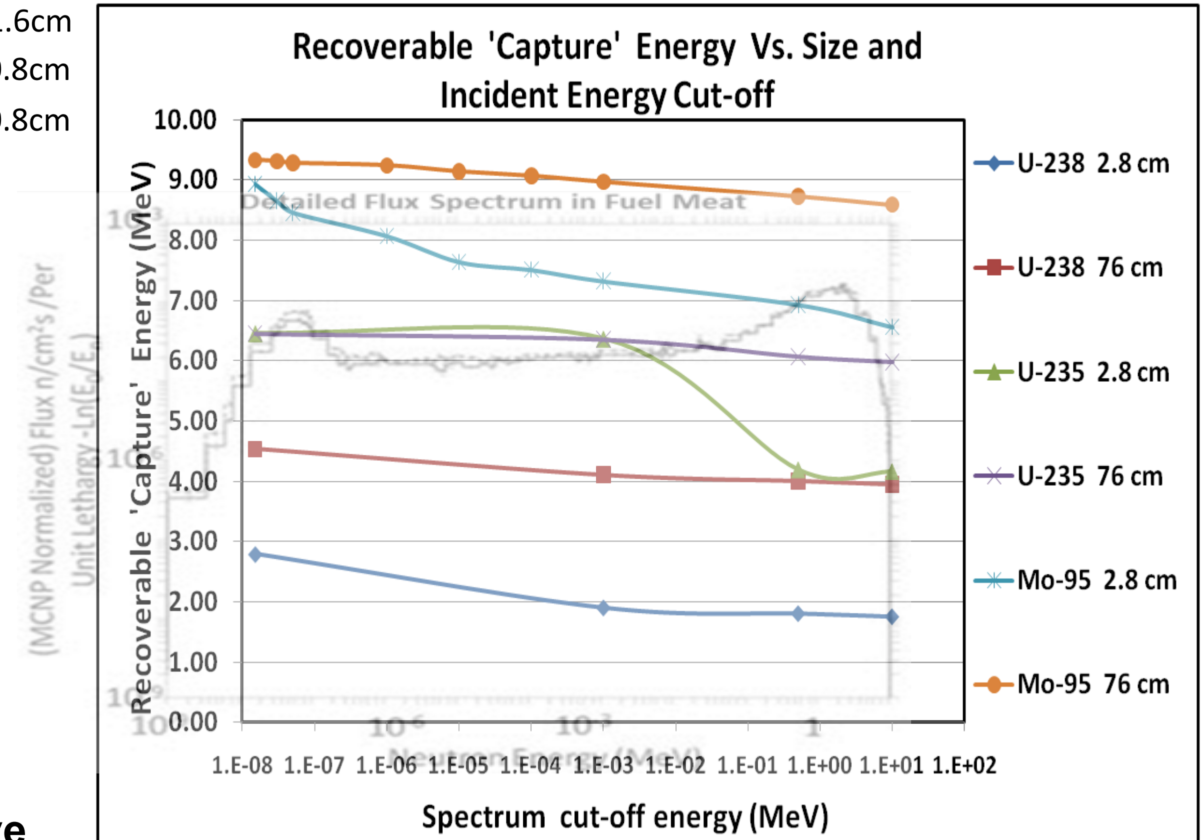


R.E. Macfarlane, https://t2.lanl.gov/nis/data/endf/ebalVII.1/summary.html

Effects of the Neutron Spectrum Energy Cut-off on Capture Heating

Mo-95(n, γ)Mo-96 Q=9.154MeV, mfp = 1.6cm
U-235(n, γ)U-236 Q=6.454MeV, mfp = 0.8cm
U-235(n, γ)U-236 Q=4.806MeV, mfp = 0.8cm





MCNP models with reflective boundary conditions

N. J. Peters, J.C. McKibben, K. Kutikkad, W.H. Miller, "**Refining the Accuracy of Predicting Physics Parameters at Research Reactors due to the Limitations in Energy Balance Method using MCNP and the ENDF Evaluations**", Nuclear Science and Engineering, **171(3)**: 210-219 (2012)

Practical Importance of Proper Gamma Production for Radiative Capture

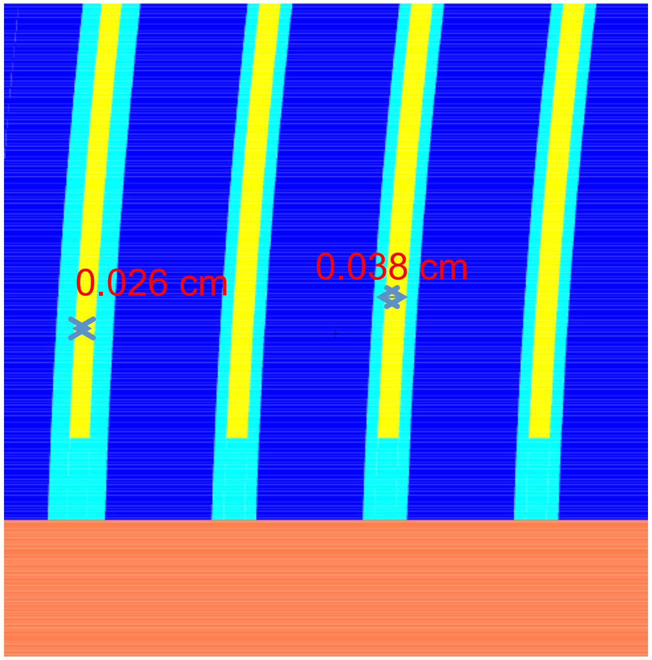
- **Heating limits for isotope production irradiations at MURR**
    - **Predicting the heating from capture gammas for a number of targets**

Gamma production is lacking for many target nuclides:

ENDF/BVII.1 NJOY Energy-balance checks
Te*(n, γ) no gamma production (**production of I-131**)
Ru*(n, γ) no gamma production
Rh-103(n, γ) no gamma production
Eu-154(n, γ) no gamma production
Eu-155(n, γ) no gamma production
Ce-141(n, γ) no gamma production
Ce-140(n, γ) no gamma production
Sn*(n, γ) no gamma production

- **MURR fuel conversion feasibility**
    - **Predicting the recoverable capture energy for the proposed U10Mo LEU matrix**

Extremely thin plates and lacking capture gamma production data for Mo and U



MURR MCNP5 U10Mo LEU Fuel Plates Model

Conclusion

- Where do we go from here?
  - Continual improvement of data sets
- How do we prioritize the requests for improved nuclear data sets?
    - Base on the need for isotope production
      - Medical and industrial (production and capture heat deposition applications)
    - Analytical techniques involving activation
      - INAA
    - The need for data on long-lived and 'important' short-lived nuclides