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Reactor Metrology for TREAT Experiments

Tommy V Holschuh, Scott M Watson, James T Johnson, David L Chichester

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Reactor Metrology for TREAT Experiments

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Radiochemistry and Nuclear Measurements
Idaho National Laboratory

19 February 2020

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Overview

- TREAT Reactor Metrology (RMet) divided into two areas
  - Flux Wires
    - Ti, Fe, Co, Ni, Nb…
  - Fission Wires
    - DU, 19.43% UZr

- Flux wires give information about the neutron spectrum in a location of the reactor
  - Each isotope has different neutron interaction cross sections
  - Show deviations in spectrum compared to simulation

- Fission wires provide number of fissions in a location of the reactor
  - Using mass of wire and reactor energy → Energy Coupling Factor

Flux Wires

Fission Wires

Flux Wires

Fission Wires

Flux Wires

- Objective
  - Determine neutron energy spectrum
- Steps
  - Choose Wire
    - (Jim, Tommy)
  - Perform MCNP simulation to estimate neutron energy spectrum
    - (Jim, Kellen)
  - Irradiate wires in chosen position
    - (Kellen)
  - Count flux wire with detector
    - (Scott, Tommy)
  - Calculate activity for each flux wire
    - (Tommy)
  - Use software to “adjust” MCNP spectrum using calculated activities
    - (Tommy)

Flux Wires – Choose Wire

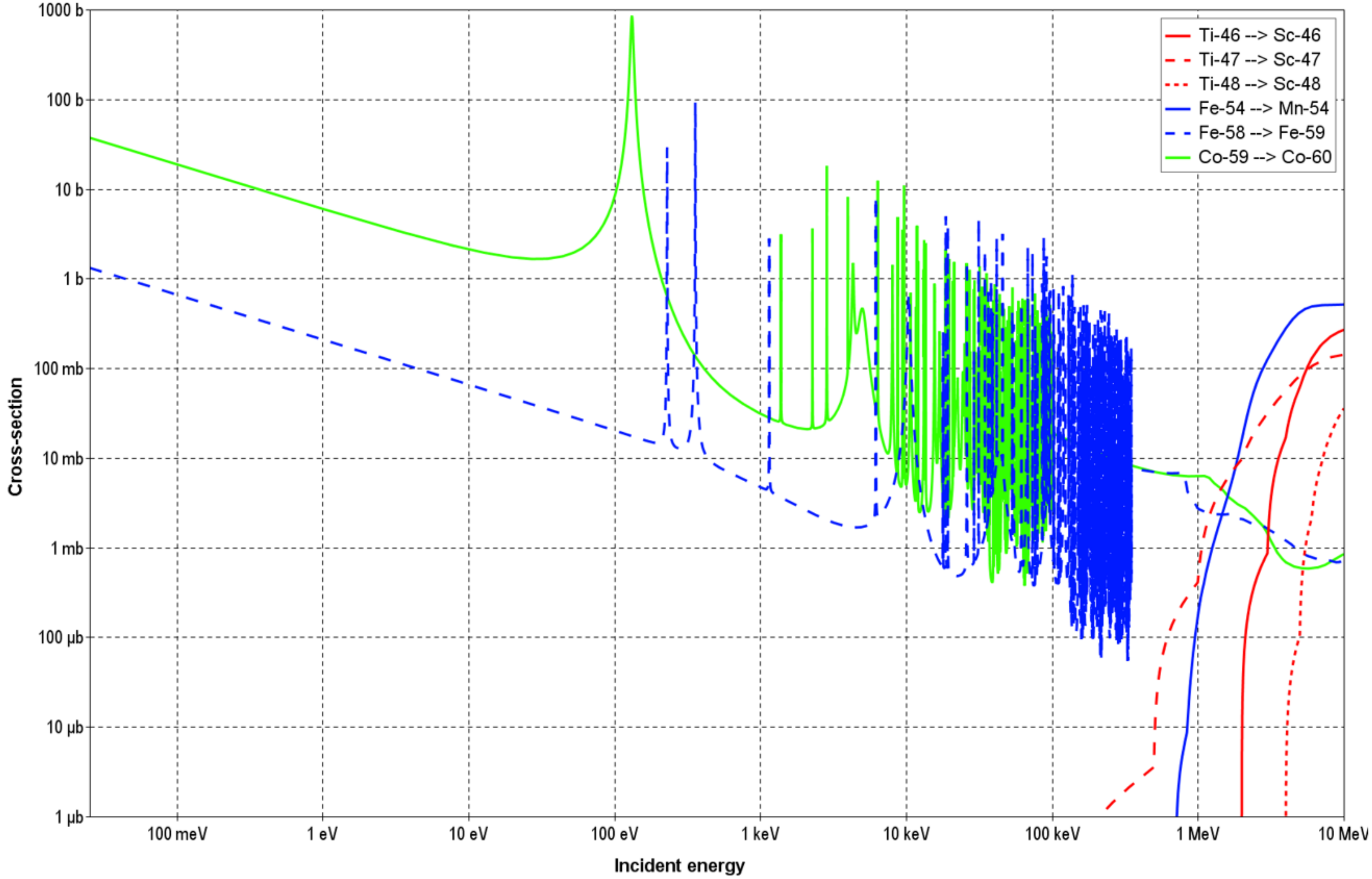
- Parent nuclide will have cross section of interest
  - n, gamma
  - n, p
  - n, alpha
  - n, n’
  - n, 2n
- Most useful wires would have multiple reactions
- Resulting daughter nuclide must be radioactive
- For gamma spectroscopy, must emit a gamma-ray
  - i.e. Sr-90 is beta only
- Half-life of usable value
  - Desirable to measure nuclide with half-life on order of decay time between irradiation and measurement
- Wire selection may change depending on reactor
  - TREAT is very thermal (which changes with temperature)

Flux Wires

Parent Isotope (Activation Wires)	Density (g/cc)	Natural Isotopic Abundance (%)	Daughter Isotope	Reaction Type
Ti-46	4.506	8.25 ± 0.03	Sc-46	Fast (n, p)
Ti-47		7.44 ± 0.02	Sc-47	Fast (n, p)
Ti-48		73.72 ± 0.03	Sc-48	Fast (n, p)
Fe-54	7.874	5.845 ± 0.035	Mn-54	Fast (n, p)
Fe-58		0.282 ± 0.004	Fe-59	Thermal (n, γ)
Co-59 (Al-0.1%Co)	8.9 (2.706)	100 (0.1)	Co-60	Thermal (n, γ)

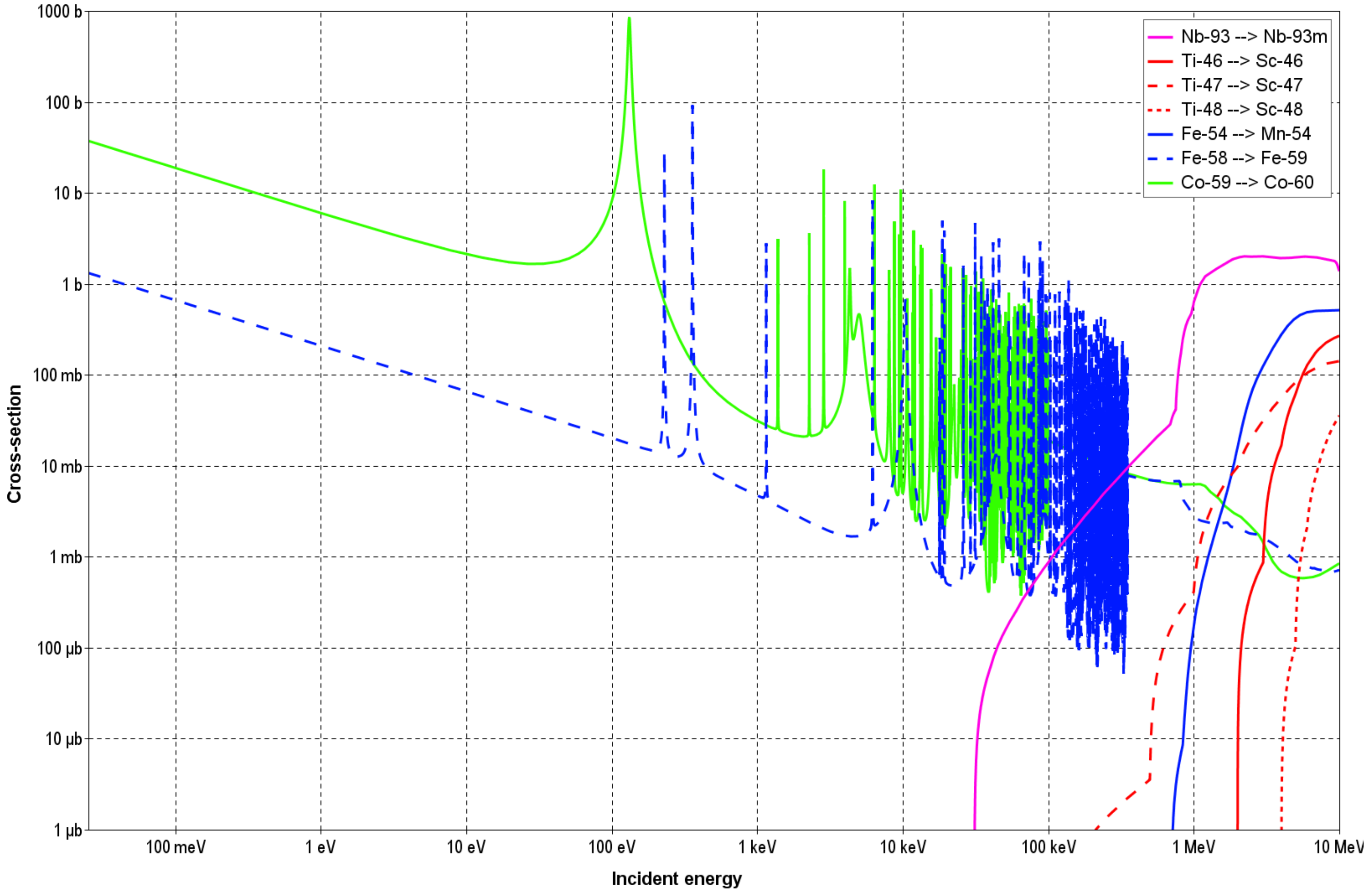
Daughter Isotope	Half Life	Decay Photon Energy (keV)	Absolute Yield (%)
Sc-46	83.79 ± 0.04 d	889.277 ± 0.003	99.984 ± 0.001
		1120.545 ± 0.004	99.987 ± 0.001
Sc-47	3.3492 ± 0.0006 d	159.381 ± 0.015	68.3 ± 0.4
Sc-48	43.67 ± 0.09 h	983.526 ± 0.012	100.1 ± 0.6
		1037.522 ± 0.012	97.6 ± 0.7
		1312.12 ± 0.012	100.1 ± 0.7
Mn-54	312.20 ± 0.20 d	834.848 ± 0.003	99.976 ± 0.001
Fe-59	44.495 ± 0.009 d	1099.245 ± 0.003	56.5 ± 1.8
		1291.590 ± 0.006	43.2 ± 1.4
Co-60	1925.28 ± 0.14 d	1173.228 ± 0.003	99.85 ± 0.03
		1332.492 ± 0.004	99.9826 ± 0.0006

Flux Wires



Ti-46 --> Sc-46
Ti-47 --> Sc-47
Ti-48 --> Sc-48
Fe-54 --> Mn-54
Fe-58 --> Fe-59
Co-59 --> Co-60
1000 b
100 b
10 b
1 b
100 mb
10 mb
1 mb
100 µb
10 µb
1 µb
Cross-section
100 meV
1 eV
10 eV
100 eV
1 keV
10 keV
100 keV
1 MeV
10 MeV
Incident energy

Flux Wires



Nb-93 --> Nb-93m
Ti-46 --> Sc-46
Ti-47 --> Sc-47
Ti-48 --> Sc-48
Fe-54 --> Mn-54
Fe-58 --> Fe-59
Co-59 --> Co-60
Cross-section
1000 b
100 b
10 b
1 b
100 mb
10 mb
1 mb
100 µb
10 µb
1 µb
100 meV
1 eV
10 eV
100 eV
1 keV
10 keV
100 keV
1 MeV
10 MeV
Incident energy

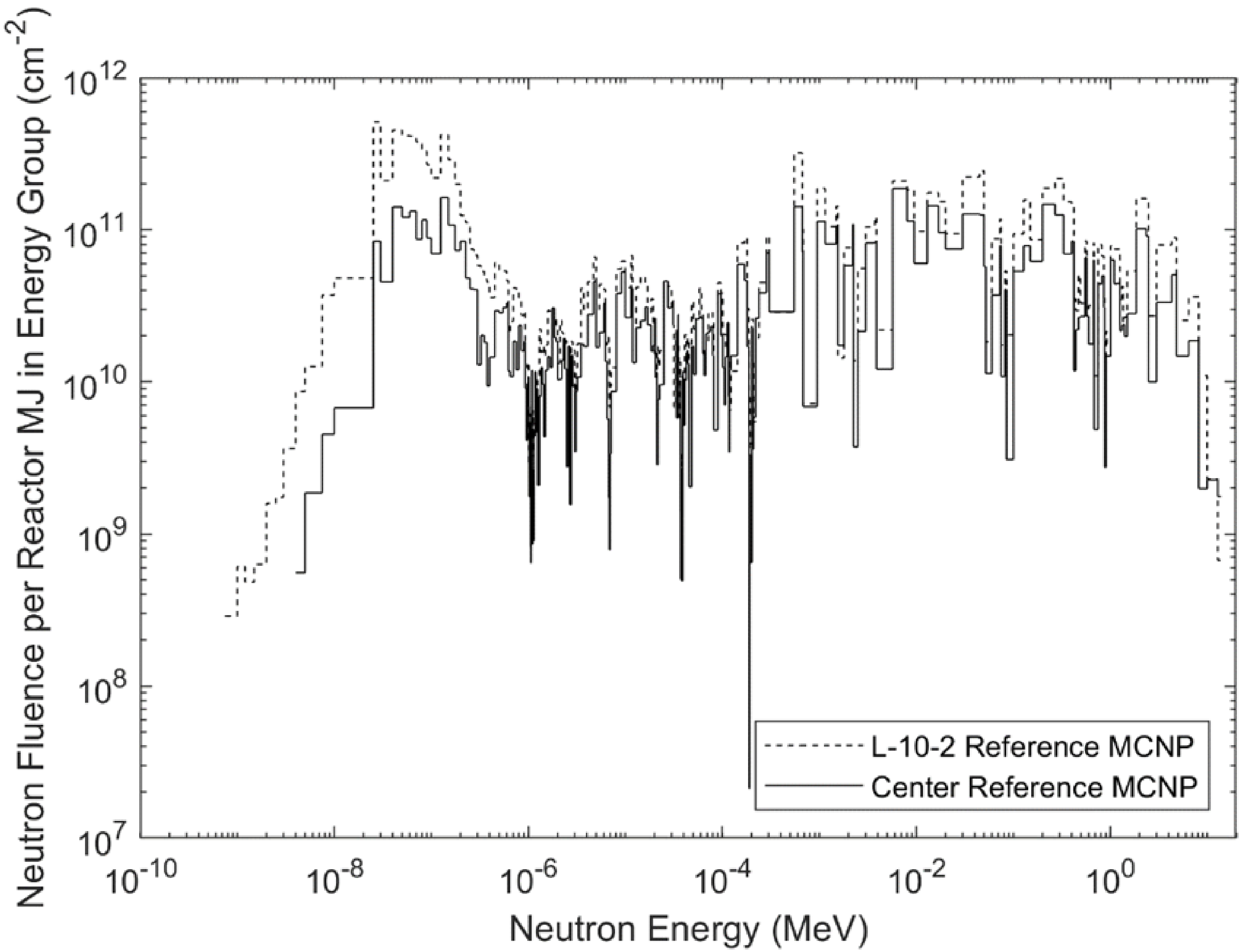
Flux Wires – Perform MCNP Simulation

- MCNP neutron tallies separated into energy bins will change depending on position in core
  - Center test position (M8, BUSTER)
  - Coolant channel (SPNDs)
  - Axial location

- Simulation may not accurately capture all phenomenon
  - It does its best

- Steady-state code package
  - Neutron spectrum does not change with time (temperature)
  - Does not incorporate control rod movements during reactor operations

Flux Wires



Neutron Fluence per Reactor MJ in Energy Group (cm^{-2})
10^{12}
10^{11}
10^{10}
10^{9}
10^{8}
10^{7}
10^{-10}
10^{-8}
10^{-6}
10^{-4}
10^{-2}
10^{0}
Neutron Energy (MeV)
L-10-2 Reference MCNP
Center Reference MCNP

Flux Wires – Calculate Activity

- TREAT Reactor Metrology Lab is located in IRC B5 laboratory
- Consists of one (or more) High-Purity Germanium (HPGe) detectors

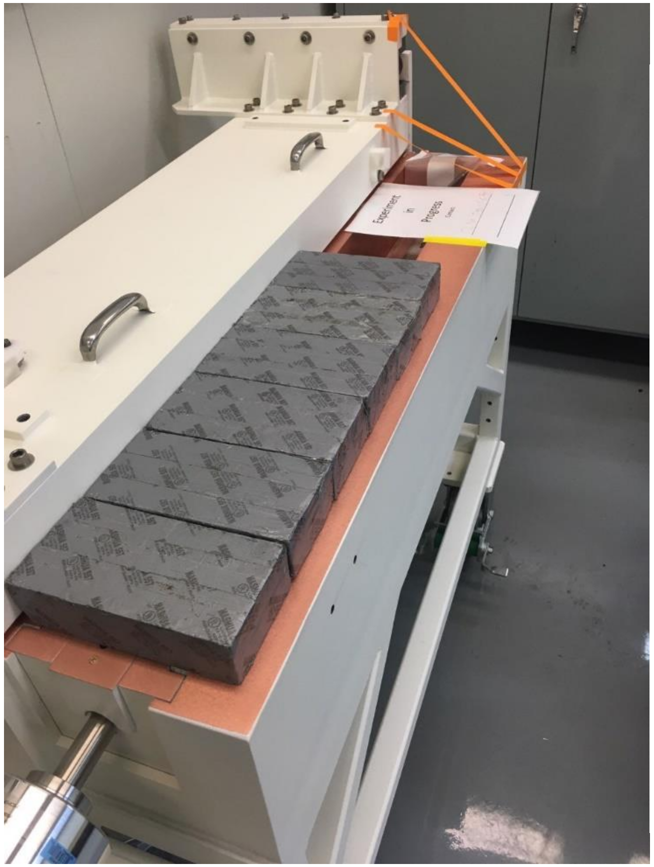
- Detectors are well-characterized
  - Calibration checks are performed routinely

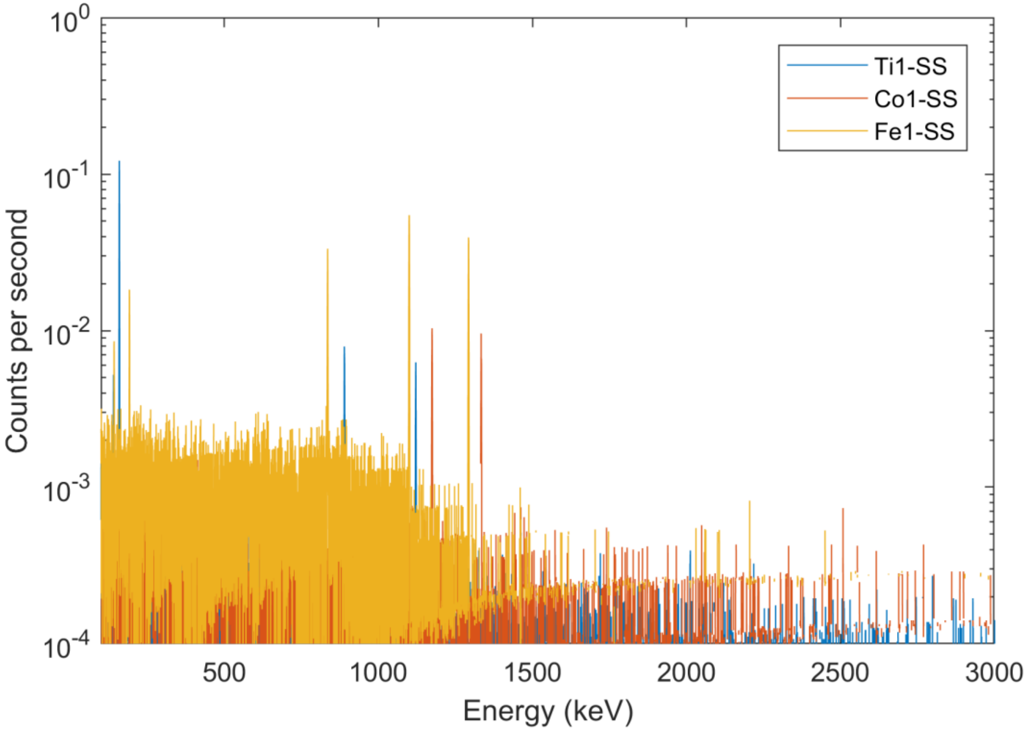
- Activity per gram of each irradiated flux wire is calculated from counting of gamma-rays emitted in characteristic peaks

- A = Activity per gram of parent isotope (Bq/g)
- λ = Decay constant (sec^{-1})
- C = Counts in photopeak for radionuclide
- t_d = Decay time between EOI and start of count (sec)
- η = Quantum yield of gamma-ray per disintegration
- ε = Absolute efficiency of detector at photopeak energy
- g = Self-shielding factor
- m = Mass of parent isotope (g)
- t_r = Real counting time (sec)

$$A = \frac{\lambda C e^{\lambda t_d}}{\eta \varepsilon g m \left(1 - e^{-\lambda t_r}\right)}$$

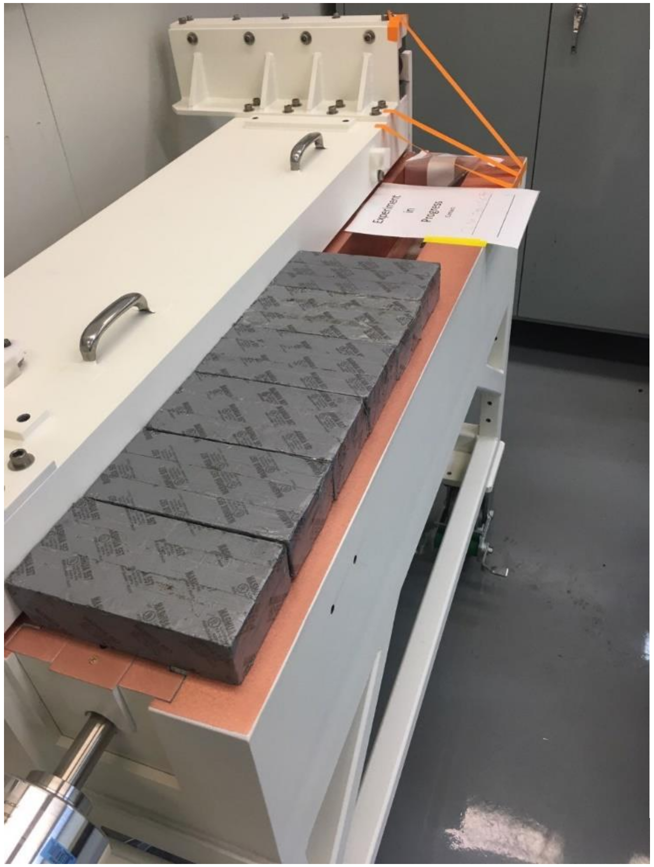
Flux Wires



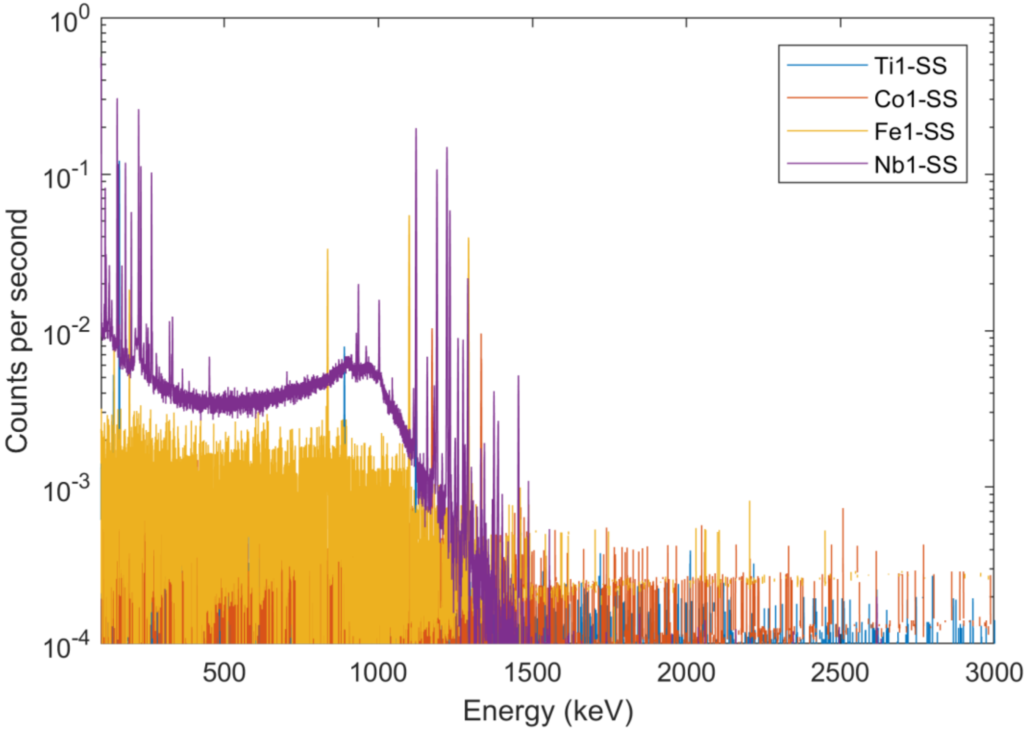


Ti1-SS
Co1-SS
Fe1-SS
10^0
10^{-1}
10^{-2}
10^{-3}
10^{-4}
Counts per second
500
1000
1500
2000
2500
3000
Energy (keV)

Flux Wires



Experiment in Progress

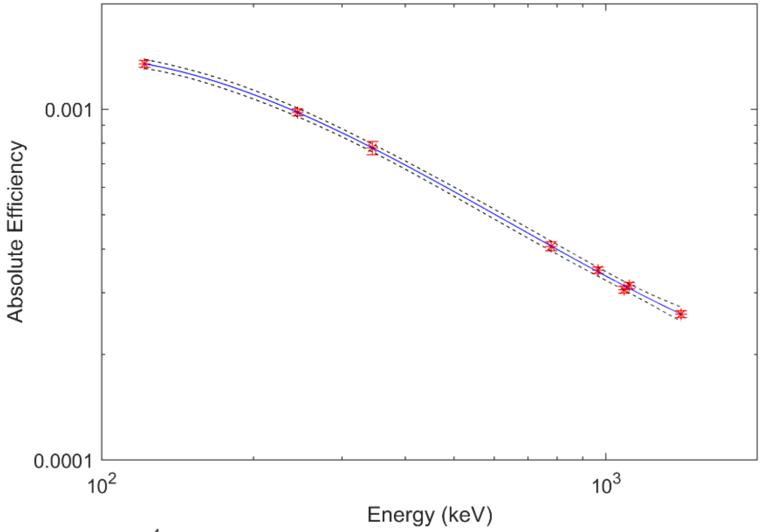


10^0
10^{-1}
10^{-2}
10^{-3}
10^{-4}
Counts per second
500
1000
1500
2000
2500
3000
Energy (keV)
Ti1-SS
Co1-SS
Fe1-SS
Nb1-SS

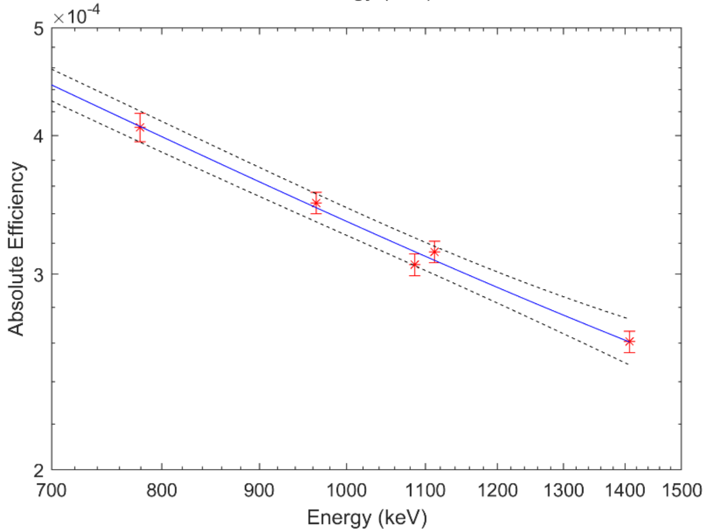
Flux Wires – Calculate Activity

- Largest effort is absolute efficiency for wire
  - Irradiated wire is counted
  - Check source used to calibrate detector (Eu-152)
    - Efficiency of detector as function of energy
  - MCNP model of check source is created, compared (Eu-152)
    - MCNP bias (<5%)
  - MCNP model of irradiated wire is created
    - Wire geometry is different from check source
    - Results are adjusted by MCNP bias
  - Efficiency for irradiated wire’s gamma rays in detector measurement is determined
- MCNP wire model must be performed for each unique wire/distance combination

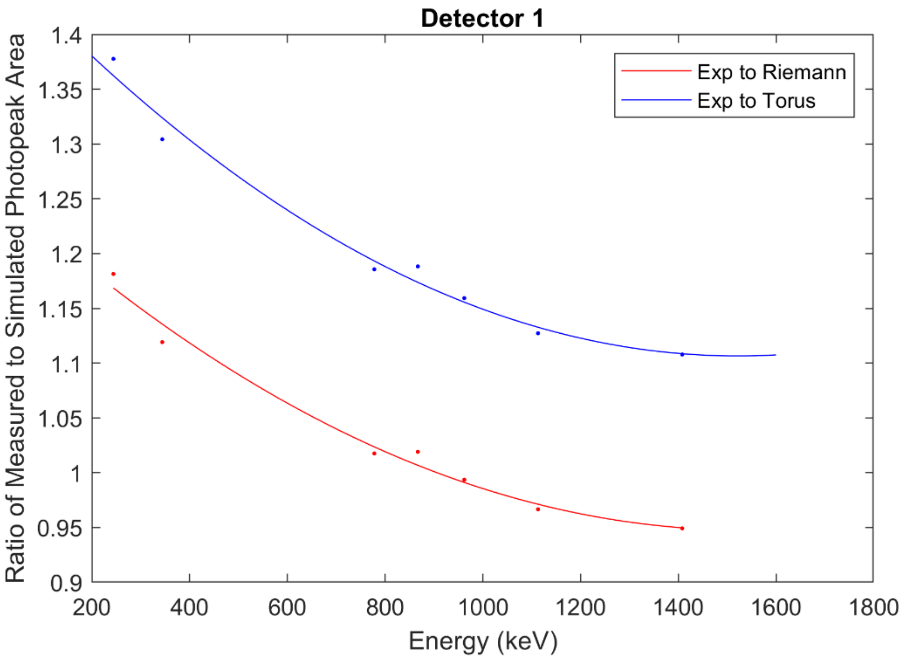
Flux Wires



Absolute Efficiency
0.001
0.0001
10^2
10^3
Energy (keV)



$\times 10^{-4}$
5
4
3
2
Absolute Efficiency
700
800
900
1000
1100
1200
1300
1400
1500
Energy (keV)



Detector 1
Exp to Riemann
Exp to Torus
Ratio of Measured to Simulated Photopeak Area
1.4
1.35
1.3
1.25
1.2
1.15
1.1
1.05
1
0.95
0.9
200
400
600
800
1000
1200
1400
1600
1800
Energy (keV)

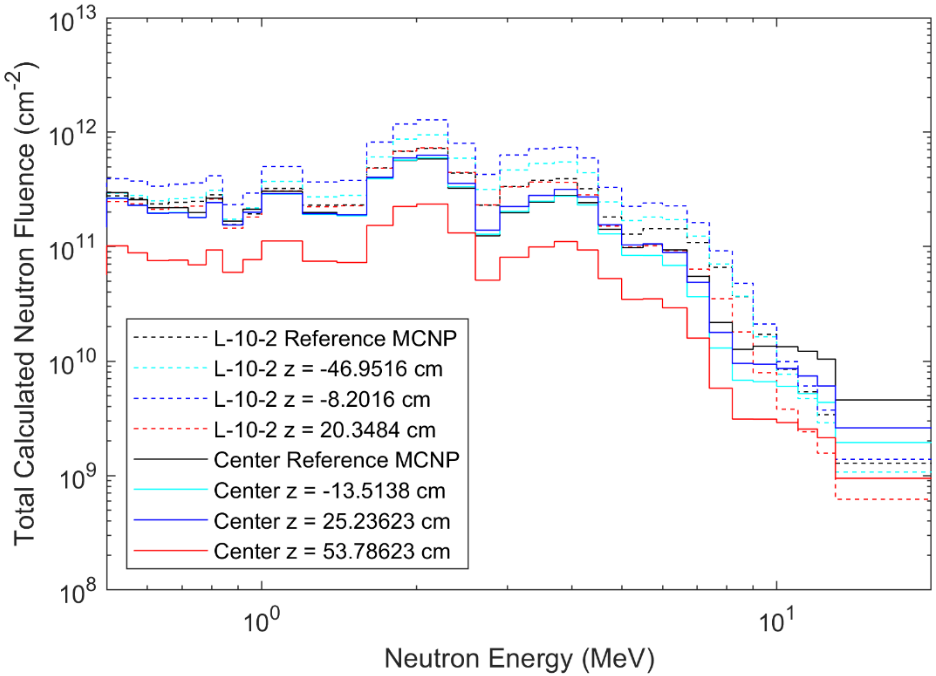
Flux Wires – STAYSL

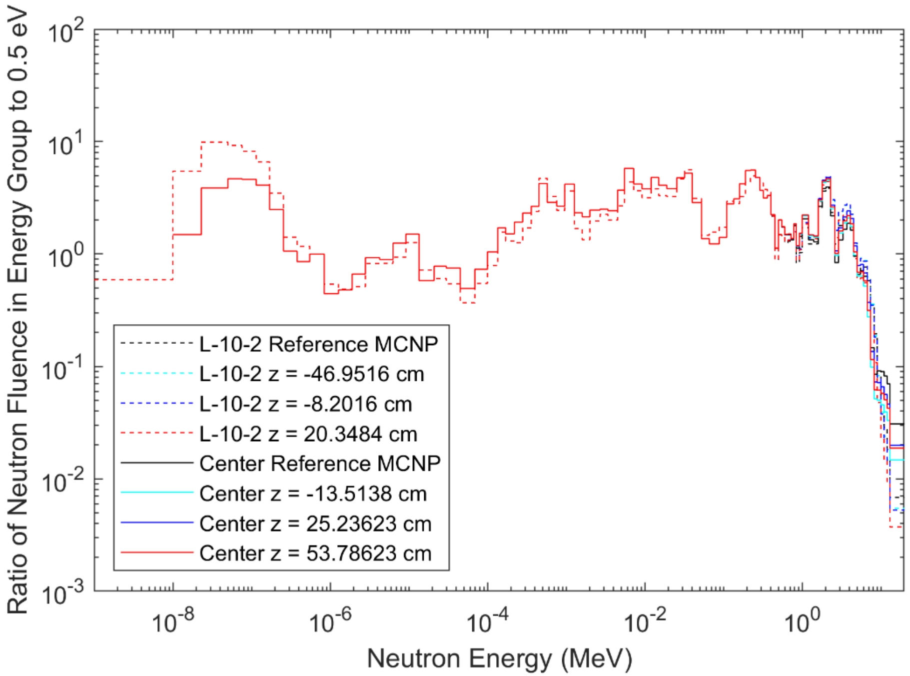
- PNNL STAYSL software package uses measured wire activities to “adjust” a guessed spectrum
  - Typically, wire activities are reported to end of irradiation (EOI)
  - STAYSL requires saturated activity in its calculation

  - MCNP provides hundreds of neutron groups
  - But typically less than 10 wire reactions are available
    - Solves with cross section data and covariance matrix
    - An underdetermined matrix

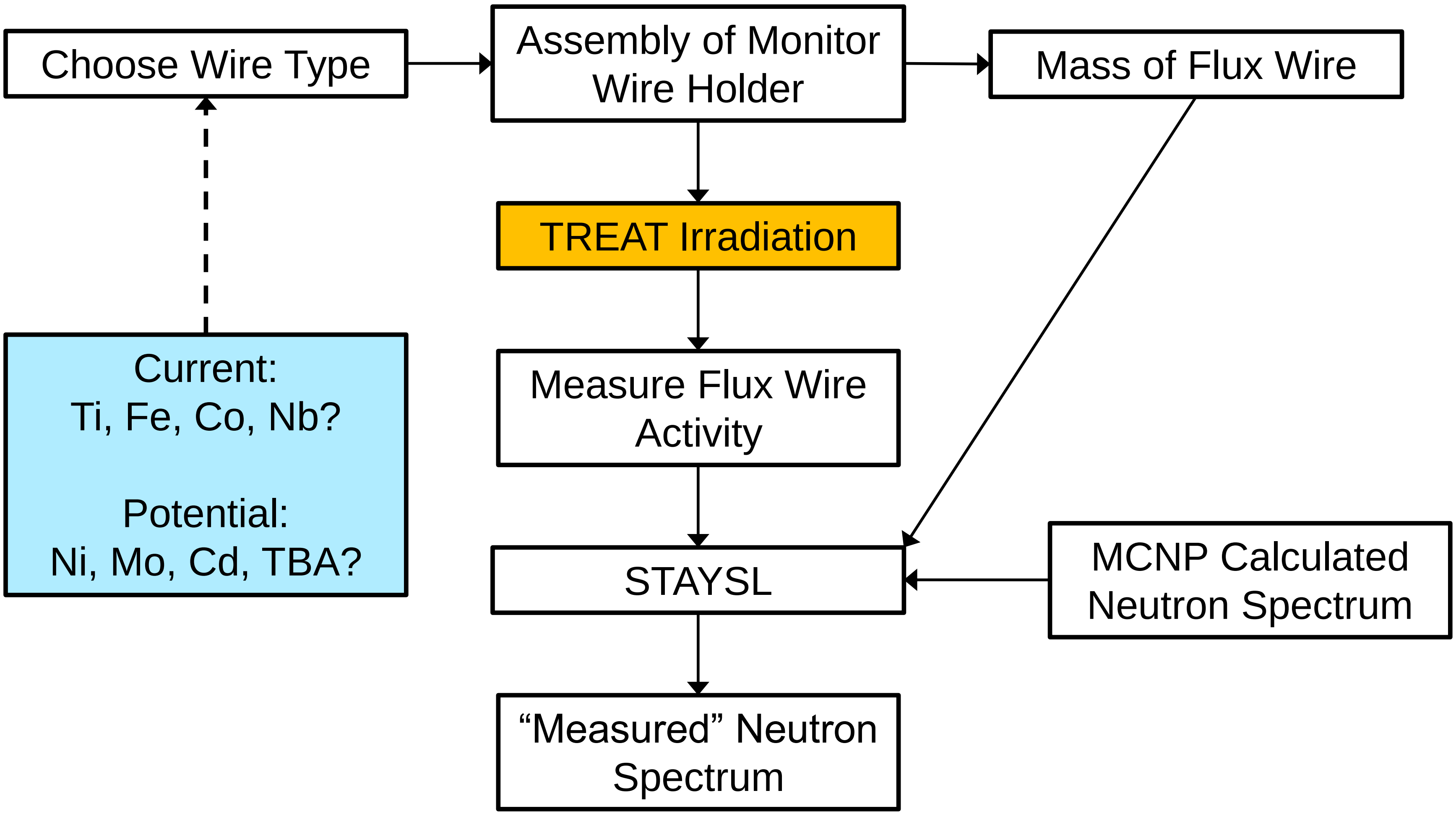
- STAYSL is very sensitive to initial MCNP “guess” for spectrum
- What usable information can STAYSL provide?
  - Relative correction for axial position
  - Ratio of thermal/fast fluence
  - Absolute neutron spectrum?

Flux Wires


Total Calculated Neutron Fluence (cm^{-2})
Neutron Energy (MeV)
10^{8}
10^{9}
10^{10}
10^{11}
10^{12}
10^{13}
10^{0}
10^{1}
L-10-2 Reference MCNP
L-10-2 z = -46.9516 cm
L-10-2 z = -8.2016 cm
L-10-2 z = 20.3484 cm
Center Reference MCNP
Center z = -13.5138 cm
Center z = 25.23623 cm
Center z = 53.78623 cm


Ratio of Neutron Fluence in Energy Group to 0.5 eV
Neutron Energy (MeV)
10^{-3}
10^{-2}
10^{-1}
10^{0}
10^{1}
10^{2}
10^{-8}
10^{-6}
10^{-4}
10^{-2}
10^{0}
L-10-2 Reference MCNP
L-10-2 z = -46.9516 cm
L-10-2 z = -8.2016 cm
L-10-2 z = 20.3484 cm
Center Reference MCNP
Center z = -13.5138 cm
Center z = 25.23623 cm
Center z = 53.78623 cm

Flux Wires – Conclusion



Flux Wires

Fission Wires

Fission Wires

- Objective
  - Determine total fissions (or coupling factor) for a given test position

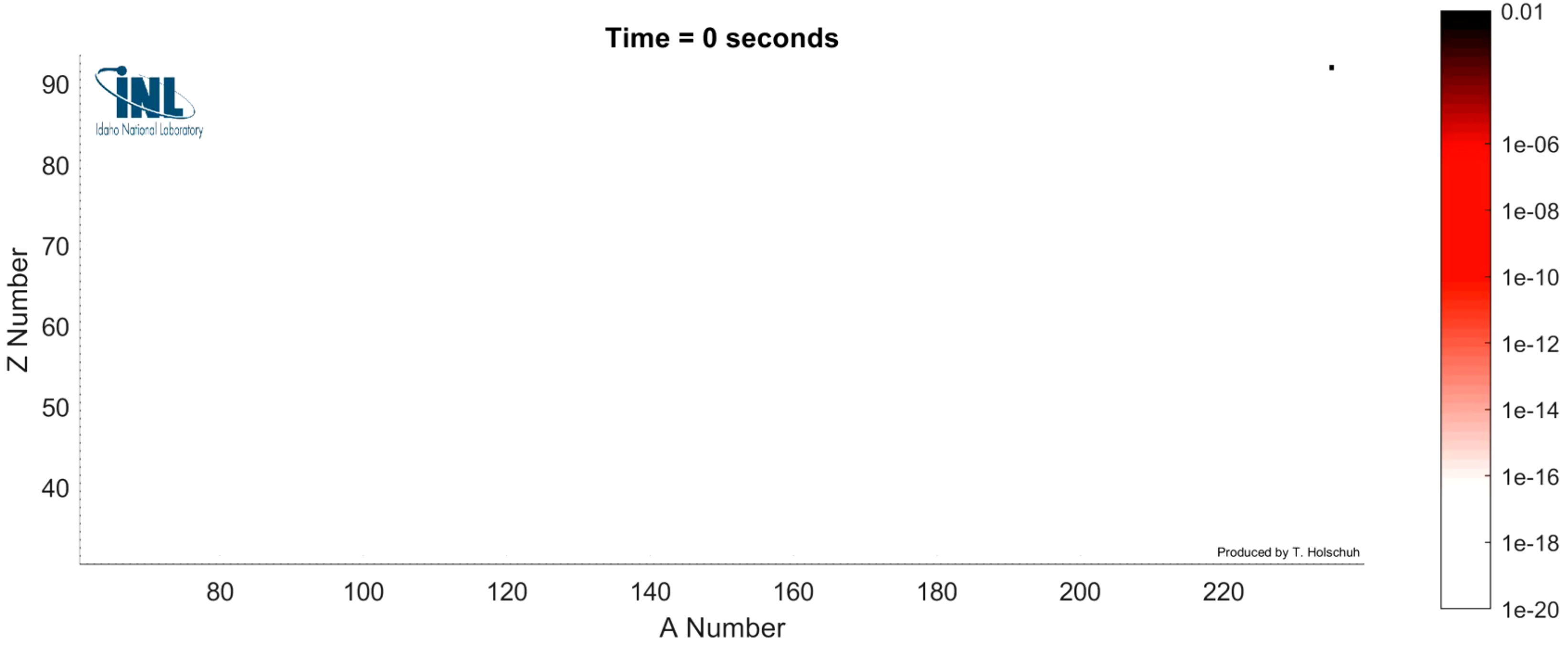
- Steps
  - Choose Wire
    - (Jim)
  - Irradiate wires in chosen position
    - (Kellen)
  - Count fission wire with detector
    - (Scott, Tommy)
  - Calculate number of fissions (or coupling factor)
    - (Tommy)

Fission Wires – Choose Wire

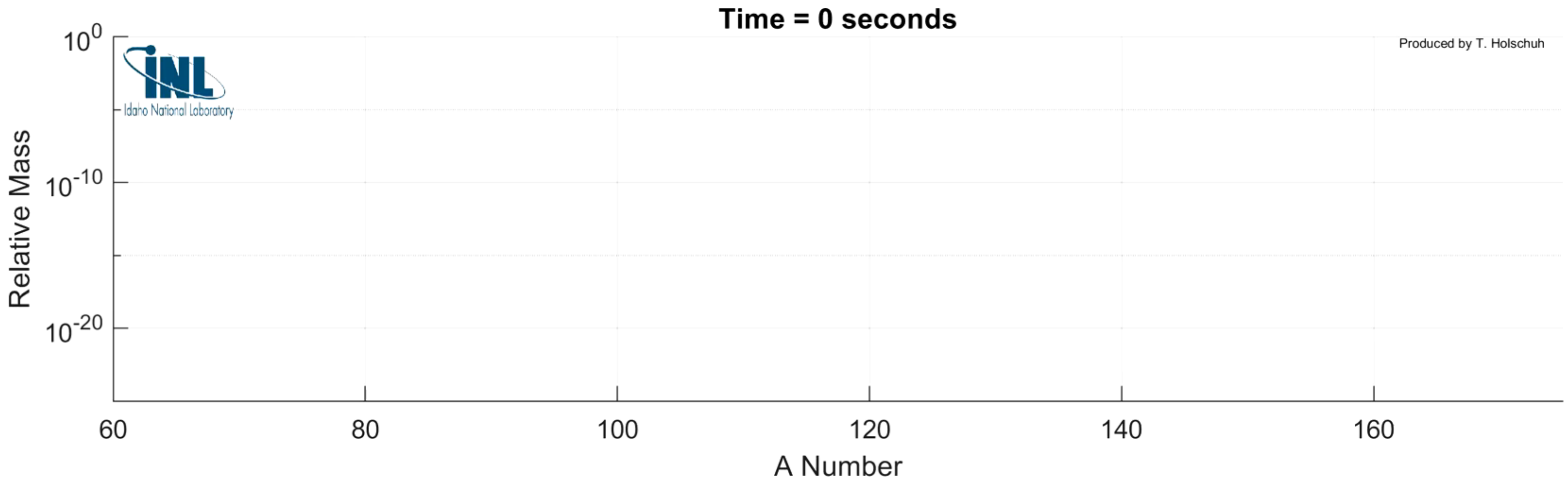
- Currently at TREAT, two choices for fission wires
  - Depleted uranium (DU) metal
    - TREAT has a large inventory
  - Low-enriched uranium-zirconium alloy (19.43% enriched UZr)
    - Higher melting point
    - Lower fraction of threshold fission from U-238
    - Inventory limited (legacy material)

- Characterization of fission products from U-235

Fission Wires



Fission Wires



Fission Wires – Calculate Fissions

- Only one fission product is necessary to calculate the number of fissions
  - More fission products allow for a lower uncertainty to be determined
- For gamma spectroscopy, fission products must emit a gamma-ray
- Half-life of usable value
  - Desirable to measure nuclide with half-life on order of decay time between irradiation and measurement
- Avoid gases (I-131, I-132, Xe-135)
- With UZr fission wire, zirconium fission products and their daughters cannot be used
- Uranium has a K-edge around 140 keV, attenuation in this region can be difficult (i.e. Tc-99m)
  - Avoid using gamma-rays less than ~200 keV

Fission Wires

- N = Number of fissions per gram of fissile isotope Activity per gram of parent isotope (Bq/g)
- C = Counts in photopeak for radionuclide
- λ = Decay constant (sec^{-1})
- t_d = Decay time between EOI and start of count (sec)
- F = Time-corrected fission yield
- η = Quantum yield of gamma-ray per disintegration
- ε = Absolute efficiency of detector at photopeak energy
- g = Self-shielding factor
- m = Mass of parent isotope (g)
- t_r = Real counting time (sec)

$$N = \frac{Ce^{\lambda t_d}}{F\eta\varepsilon mg\left(1-e^{-\lambda t_r}\right)}$$

Isotope	Half Life	Photon Peaks of Interest (keV)	Absolute Photon Yield per Disintegration (%)
Zr-95	64.032 ± 0.006 d	756.725 ± 0.012	54.38 ± 0.22
Nb-95	34.991 ± 0.006 d	765.803 ± 0.006	99.808 ± 0.007
Mo-99	65.924 ± 0.006 h	739.500 ± 0.017	12.20 ± 0.16
Tc-99m (Mo-99)	6.0072 ± 0.0009 h	140.511 ± 0.001	89 ± 4
Ru-103	39.247 ± 0.013 d	497.085 ± 0.010	91.0 ± 1.2
I-131	8.0252 ± 0.0006 d	364.489 ± 0.005	81.5 ± 0.8
I-132 (Te-132)	2.295 ± 0.013 h (3.204 ± 0.013 d)	522.65 ± 0.09	16.0 ± 0.5
Ba-140	12.7527 ± 0.0023 d	537.261 ± 0.009	24.39 ± 0.22
La-140	1.67855 ± 0.00012 d	487.021 ± 0.012	45.5 ± 0.6
Nd-147	10.98 ± 0.01 d	531.016 ± 0.022	13.4 ± 0.3

Fission Wires – Calculate Fissions

- Largest effort is absolute efficiency for wire
  - Irradiated wire is counted
  - Check source used to calibrate detector (Eu-152)
    - Efficiency of detector as function of energy
  - MCNP model of check source is created, compared (Eu-152)
    - MCNP bias (<5%)
  - MCNP model of irradiated wire is created
    - Wire geometry is different from check source
    - Results are adjusted by MCNP bias
  - Efficiency for fission product’s gamma rays in detector measurement is determined
- MCNP wire model must be performed for each unique wire/distance combination

Fission Wires – Calculate Coupling Factor

- Energy Coupling Factor (ECF) normalizes the fissions based on wire mass and total reactor energy released
  - Traditionally termed the power coupling factor (PCF)

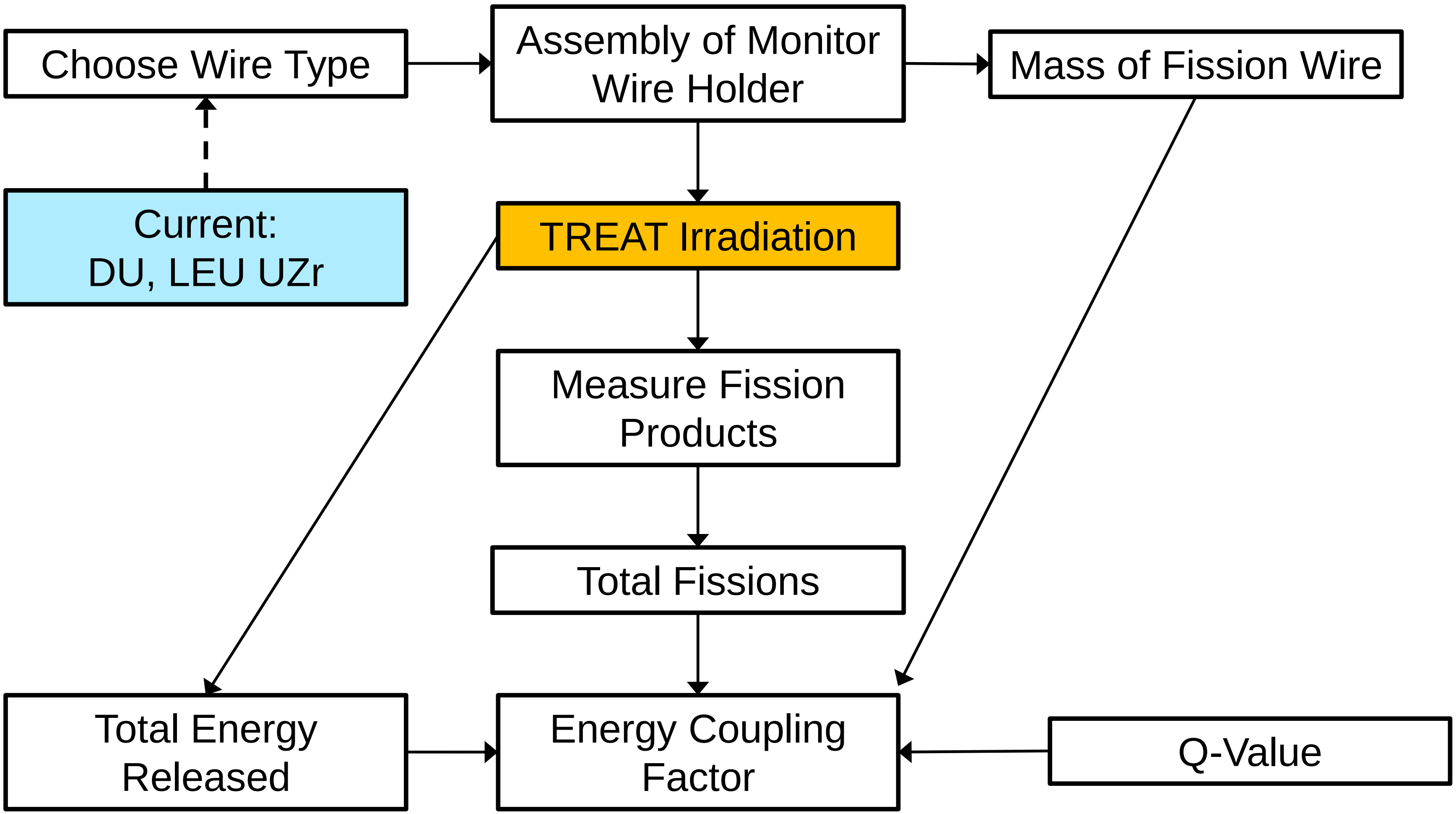
$$J = fissions \times \frac{MeV}{fission} \times \frac{1.602e-13\ J}{MeV}$$

$$ECF \equiv \frac{\text{Energy deposited in sample}}{(\text{material mass})(\text{Reactor Energy})} = \frac{J}{g-MJ}$$

- Q-value must be determined
  - In TREAT, value is 182 MeV/fission – see Jim Parry
  - In fuel, this value could be different (~190-200 MeV)

- ECF is compared to simulation with MCNP and/or appropriate MOOSE package

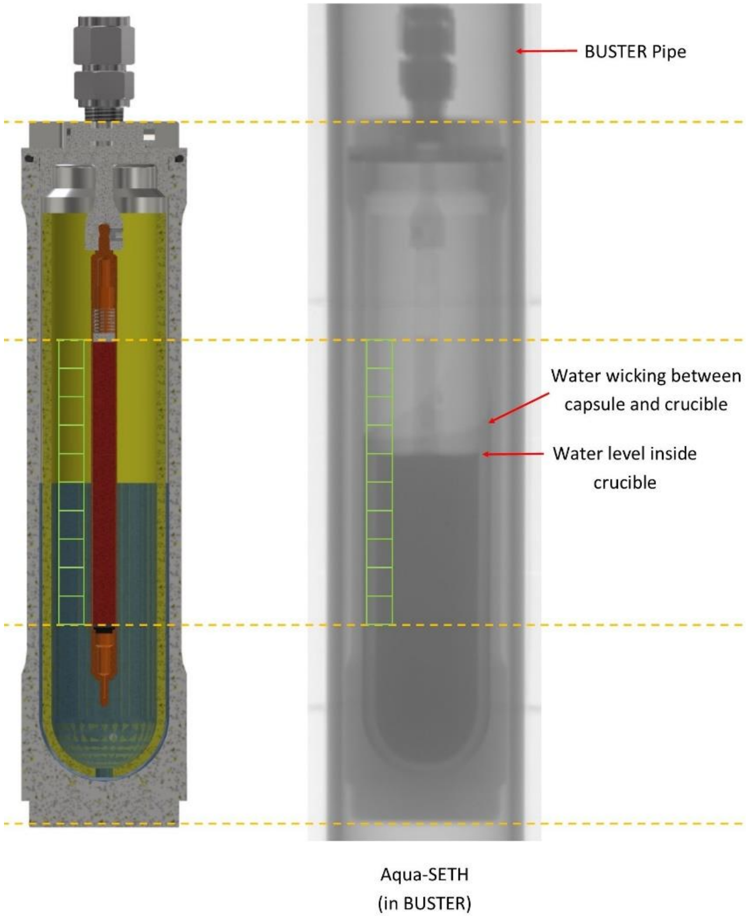
Fission Wires – Conclusion



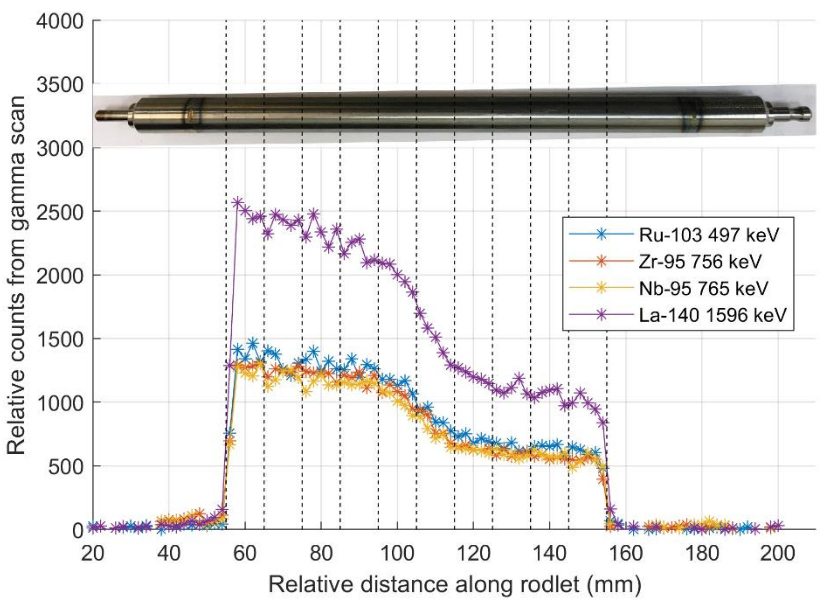
TREAT Reactor Metrology Fuel Measurements

- The methodology for fission wires can be extended to larger fission materials
  - SETH-A
    - ECF calculation
    - Gamma Scan (Jimmy)
  - Aqua-SETH
    - ECF calculation for entire rodlet
    - Gamma Scan – half submerged in water – ECF by pellet
    - Increase in ECF compared to dry environment
  - M-SERTTA
    - ECF calculation for entire rodlet
    - Gamma Scan – change due to enrichment – ECF by pellet
    - Changed set-up slightly and can perceive individual pellets within rodlet

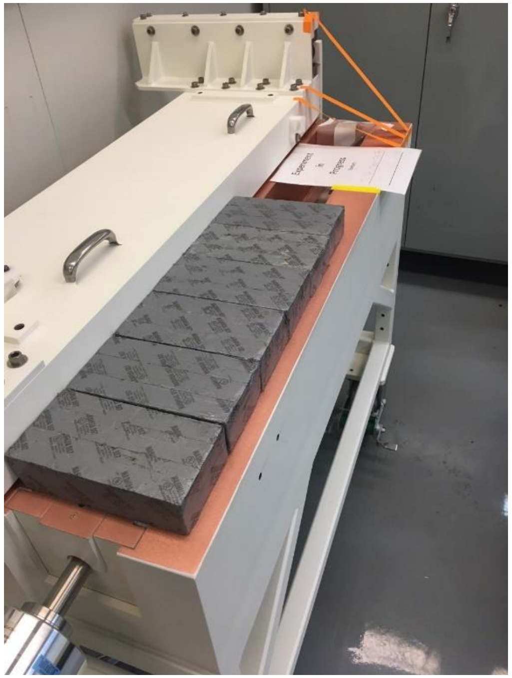
Aqua-SETH Measurements

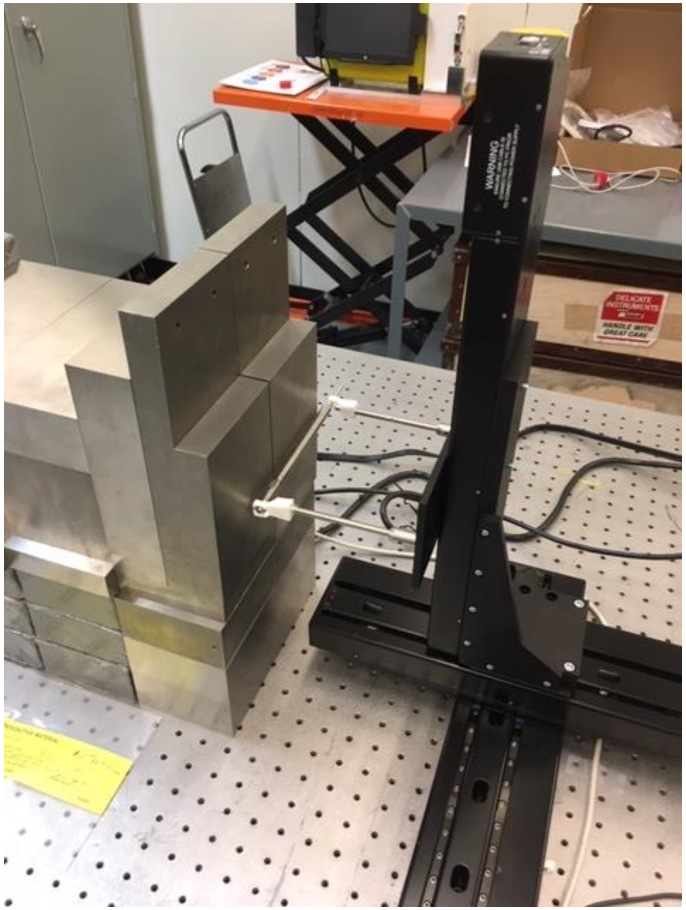


Pellet	ECF (J-g/MJ)	Unc (k=2)	Ratio to Pellet #9
1	0.828	0.074	2.055
2	0.874	0.079	2.168
3	0.864	0.078	2.143
4	0.825	0.074	2.047
5	0.729	0.064	1.809
6	0.563	0.049	1.396
7	0.448	0.039	1.113
8	0.417	0.036	1.034
9	0.403	0.034	1.000
10	0.366	0.031	0.908



M-SERTTA Measurements

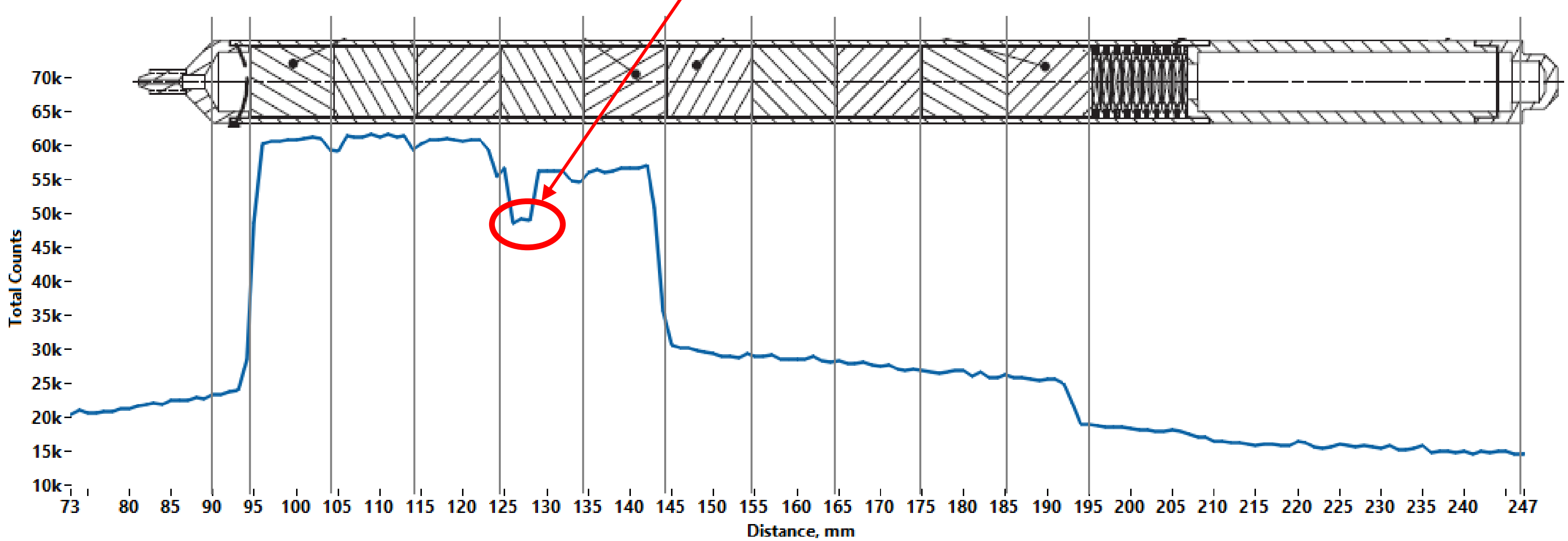




TREAT Reactor Metrology Fuel Measurements

- 4 inches of tungsten shielding + 1.5 +/- .25 mm fuel offset
- 1 mm slit width
- 2700 second dwell
- 1 mm step size
- Start Position 73 mm
- End Position 260 mm

These three points were recorded in a subsequent data set on a different
day.



TREAT Core Map

	A	B	C	D	E	F	G	H	J	K	L	M	N	O	P	R	S	T	U
1	ZRD	ZRD	108	398	283	324	247	319	408	H01	289	140	356	271	178	366	313	ZRD	SRC
2	ZRD	169	305	391	128	393	294	213	236	H02	224	285	115	383	358	367	196	198	ZRD
3	522	147	221	384	303	244	203	702	217	H03	186	728	164	142	161	342	346	286	287
4	194	298	382	314	389	701	145	183	257	H04	207	165	291	718	127	405	353	284	360
5	348	388	171	295	260	175	249	138	179	H05	173	344	302	117	120	148	182	312	399
6	155	369	226	700	230	228	231	241	296	H06	214	118	341	256	297	712	263	381	362
7	396	152	211	222	317	216	280	136	149	H07	197	269	277	215	156	242	253	177	349
8	328	141	726	258	121	722	160	321	114	H08	343	336	170	723	264	172	705	407	329
9	248	167	254	332	377	412	206	204	281	H21	378	229	309	279	403	300	157	116	272
10	347	133	113	112	359	290	210	340	166	M-8 Cal	322	104	416	282	529	106	310	306	414
11	395	129	239	331	190	163	275	259	371	HZD	126	223	372	523	202	252	135	245	386
12	392	397	729	278	262	716	243	530	185	153	209	233	304	721	192	292	714	413	355
13	102	311	123	238	323	250	330	526	502	521	500	503	531	417	265	237	246	288	299
14	318	232	240	711	184	154	159	137	501	528	527	525	101	109	334	713	357	273	333
15	387	168	401	132	187	119	144	327	201	122	520	370	199	227	208	220	320	143	162
16	351	415	406	364	111	717	189	255	174	301	110	200	293	720	100	394	326	124	139
17	107	361	379	363	103	146	180	724	195	130	150	703	188	225	380	338	266	131	375
18	ZRD	385	308	365	205	402	151	181	307	212	352	218	125	390	404	374	235	368	ZRD
19	ZRD	ZRD	191	409	315	337	134	373	339	251	335	354	176	345	400	234	325	ZRD	ZRD

- Red – Fuel Assemblies
- Yellow – Control Rods
- Green – Slotted Assemblies for Hodoscope
- Black – Graphite Dummy Assemblies
- Purple – Test Position