Reactor Metrology for TREAT Experiments

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

March 2020
Reactor Metrology for TREAT Experiments

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

March 2020

Idaho National Laboratory
Idaho Falls, Idaho 83415

http://www.inl.gov

Prepared for the
U.S. Department of Energy

Under DOE Idaho Operations Office
Contract DE-AC07-05ID14517
Reactor Metrology for TREAT Experiments

Tommy Holschuh
Scott Watson
Jimmy Johnson
David Chichester

Radiochemistry and Nuclear Measurements
Idaho National Laboratory

19 February 2020

INL/MIS-20-57521
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

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

<table>
<thead>
<tr>
<th>Daughter Isotope</th>
<th>Half Life</th>
<th>Decay Photon Energy (keV)</th>
<th>Absolute Yield (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sc-46</td>
<td>83.79 ± 0.04 d</td>
<td>889.277 ± 0.003</td>
<td>99.984 ± 0.001</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1120.545 ± 0.004</td>
<td>99.987 ± 0.001</td>
</tr>
<tr>
<td>Sc-47</td>
<td>3.3492 ± 0.0006 d</td>
<td>159.381 ± 0.015</td>
<td>68.3 ± 0.4</td>
</tr>
<tr>
<td>Sc-48</td>
<td>43.67 ± 0.09 h</td>
<td>983.526 ± 0.012</td>
<td>100.1 ± 0.6</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1037.522 ± 0.012</td>
<td>97.6 ± 0.7</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1312.12 ± 0.012</td>
<td>101.1 ± 0.7</td>
</tr>
<tr>
<td>Mn-54</td>
<td>312.20 ± 0.20 d</td>
<td>834.848 ± 0.003</td>
<td>99.976 ± 0.001</td>
</tr>
<tr>
<td>Fe-59</td>
<td>44.495 ± 0.009 d</td>
<td>1099.245 ± 0.003</td>
<td>56.5 ± 1.8</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1291.590 ± 0.006</td>
<td>43.2 ± 1.4</td>
</tr>
<tr>
<td>Co-60</td>
<td>1925.28 ± 0.14 d</td>
<td>1173.228 ± 0.003</td>
<td>99.85 ± 0.03</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1332.492 ± 0.004</td>
<td>99.9826 ± 0.0006</td>
</tr>
</tbody>
</table>
Flux Wires

Flux Wires
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

\[ \text{Neutron Fluence per Reactor MJ in Energy Group (cm}^{-2}) \]

\[ \text{Neutron Energy (MeV)} \]

- 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

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

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

- Graph showing absolute efficiency vs. energy (keV) with data points and fitted curves.
- Graph showing ratio of measured to simulated photopeak area vs. energy (keV) for Detector 1, with data points and fitted curves labeled 'Exp to Riemann' and 'Exp to Torus.'
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
Flux Wires – Conclusion

Choose Wire Type

Assembly of Monitor Wire Holder

TREAT Irradiation

Measure Flux Wire Activity

STAYSL

“Measured” Neutron Spectrum

Mass of Flux Wire

Current:
Ti, Fe, Co, Nb?

Potential:
Ni, Mo, Cd, TBA?

MCNP Calculated Neutron Spectrum
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

Time = 0 seconds

Produced by T. Holschuh
Fission Wires

Time = 0 seconds

Produced by T. Holschuh
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
- $\lambda =$ Decay constant (sec$^{-1}$)
- $t_d =$ Decay time between EOI and start of count (sec)
- $F =$ Time-corrected fission yield
- $\eta =$ Quantum yield of gamma-ray per disintegration
- $\varepsilon =$ Absolute efficiency of detector at photopeak energy
- $g =$ Self-shielding factor
- $m =$ Mass of parent isotope (g)
- $t_r =$ Real counting time (sec)

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

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

\[
J = \text{fissions} \times \frac{\text{MeV}}{\text{fission}} \times 1.602 \times 10^{-13} J
\]

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

• 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
Choose Wire Type

Assembly of Monitor Wire Holder

Mass of Fission Wire

Current: DU, LEU UZr

TREAT Irradiation

Measure Fission Products

Total Fissions

Energy Coupling Factor

Total Energy Released

Q-Value

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

<table>
<thead>
<tr>
<th>Pellet</th>
<th>ECF (J-g/MJ)</th>
<th>Unc (k=2)</th>
<th>Ratio to Pellet #9</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.828</td>
<td>0.074</td>
<td>2.055</td>
</tr>
<tr>
<td>2</td>
<td>0.874</td>
<td>0.079</td>
<td>2.168</td>
</tr>
<tr>
<td>3</td>
<td>0.864</td>
<td>0.078</td>
<td>2.143</td>
</tr>
<tr>
<td>4</td>
<td>0.825</td>
<td>0.074</td>
<td>2.047</td>
</tr>
<tr>
<td>5</td>
<td>0.729</td>
<td>0.064</td>
<td>1.809</td>
</tr>
<tr>
<td>6</td>
<td>0.563</td>
<td>0.049</td>
<td>1.396</td>
</tr>
<tr>
<td>7</td>
<td>0.448</td>
<td>0.039</td>
<td>1.113</td>
</tr>
<tr>
<td>8</td>
<td>0.417</td>
<td>0.036</td>
<td>1.034</td>
</tr>
<tr>
<td>9</td>
<td>0.403</td>
<td>0.034</td>
<td>1.000</td>
</tr>
<tr>
<td>10</td>
<td>0.366</td>
<td>0.031</td>
<td>0.908</td>
</tr>
</tbody>
</table>
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    | Q    | R    | S    | T    | U    |
|---|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|      |
| 1 | ZRD  | ZRD  | 108  | 398  | 283  | 324  | 247  | 319  | 408  | H01  | 289  | 140  | 356  | 271  | 178  | 366  | 313  |      |      |      |
| 2 | ZRD  | 169  | 305  | 391  | 128  | 393  | 294  | 213  | 236  | H02  | 224  | 285  | 115  | 383  | 358  | 367  | 196  | 198  |      |      |
| 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  |      |      |      |      |
| 5 | 348  | 388  | 171  | 295  | 260  | 175  | 249  | 138  | 179  | H05  | 173  | 344  | 302  | 117  | 120  | 148  | 182  |      |      |      |      |
| 6 | 155  | 369  | 228  | 700  | 230  | 228  | 231  | 241  | 296  | H06  | 214  | 118  | 341  | 256  | 297  | 712  | 263  |      |      |      |      |
| 7 | 396  | 152  | 211  | 222  | 317  | 216  | 360  | 136  | 149  | H07  | 197  | 269  | 277  | 215  | 156  | 242  | 253  |      |      |      |      |
| 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  | 729  | 240  | 112  | 359  | 290  | 210  | 166  | M31  | 522  | 104  | 416  | 282  | 529  | 106  | 310  | 306  | 414  |      |      |
| 11| 395  | 129  | 739  | 331  | 190  | 163  | 276  | 259  | 371  |      | 126  | 223  | 372  | 523  | 202  | 252  | 135  | 245  | 386  |      |      |
| 12| 392  | 397  | 729  | 278  | 262  | 716  | 243  | 530  | 185  |      |      |      |      |      | 721  | 192  | 292  | 724  | 413  | 355  |      |      |
| 13| 102  | 311  | 729  | 240  | 184  | 154  | 159  | 137  | 501  |      |      |      |      |      |      |      |      |      |      |      |      |      |
| 14| 387  | 168  | 401  | 132  | 187  | 119  | 144  | 327  | 201  |      |      |      |      |      |      |      |      |      |      |      |      |      |
| 15| 351  | 415  | 406  | 364  | 111  | 717  | 189  | 255  | 174  |      |      |      |      |      |      |      |      |      |      |      |      |      |
| 16| 107  | 361  | 379  | 363  | 108  | 146  | 180  |      |      |      |      |      |      |      |      |      |      |      |      |      |      |      |
| 17| 385  | 308  | 365  | 205  | 402  | 151  | 181  | 307  |      |      |      |      |      |      |      |      |      |      |      |      |      |
| 18| ZRD  | ZRD  | 191  | 409  | 315  | 337  | 134  | 373  | 339  |      |      |      |      |      |      |      |      |      |      |      |      |
| 19| ZRD  | ZRD  | 191  | 409  | 315  | 337  | 134  | 373  | 339  |      |      |      |      |      |      |      |      |      |      |      |      |

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