Accomplishments of Thermal Neutron Scattering Research at North Carolina State University

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Acknowledgment

- NNSA Nuclear Criticality Safety Program (NCSP)
  - collaboration with LLNL

- Naval Nuclear Propulsion Program (NNPP)

- The LEIP Team
## FY 2020 Tasks

<table>
<thead>
<tr>
<th>Task</th>
<th>Task Title</th>
<th>Budget</th>
</tr>
</thead>
<tbody>
<tr>
<td>ND2</td>
<td>Generation and Benchmarking of Thermal Neutron Scattering Cross Sections in Support of Advanced Nuclear Reactor Concepts</td>
<td>$72 K</td>
</tr>
<tr>
<td>ND3</td>
<td>Development and Implementation of an Advanced and Rigorous Computational Platform for Thermal Neutron Scattering Analysis</td>
<td>$100 K</td>
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<tr>
<td>ND5</td>
<td>Development and Implementation of a Modern Doppler Broadening Approach Including Atomic Binding Effects</td>
<td>$95 K</td>
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</tbody>
</table>
## APPENDIX B: Nuclear Data Priorities, Basis Statements, and Milestones

<table>
<thead>
<tr>
<th></th>
<th>Pre-FY2021</th>
<th>FY2021</th>
<th>FY2022</th>
<th>FY2023</th>
<th>FY2024</th>
<th>FY2025</th>
<th>Post-FY2025</th>
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<tbody>
<tr>
<td><strong>Materials</strong></td>
<td></td>
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</tr>
<tr>
<td>Zirconium ($^{90,91,92,94}\text{Zr}$)</td>
<td>Basis Resonance evaluations. Zirconium is a key structural element that is primarily used in cladding for fuel rods and is currently in consideration for use with advanced nuclear fuel matrices in the form of zirconium hydride. The latest ENDF/B-VII.1 resonance evaluation relies on JENDL-4 data and resonance parameters from the Atlas of Neutron Resonances. As a result, the evaluated resonance parameters are not based on detailed R-matrix analyses. In addition, newer RPI total cross-section measurements on natural zirconium indicate that the older ENDF/B-VI.8 data match the recent RPI measurements better than the newer isotopic evaluations. Furthermore, improved differential measurements of the zirconium isotopes have been identified on the OECD/NEA nuclear data High Priority Request List (HPRL). Differential measurements are needed in the resonance region to accurately predict the neutron resonances for the zirconium isotopes, and corresponding resonance evaluations are needed to provide detailed resonance parameters and covariance data. In addition, the SAMMY evaluation software has the capability to generate angular scattering distributions from the resonance parameters thereby providing detailed resonance scattering structure that will improve the elastic scattering modeling for the zirconium isotope evaluations. NR continues to be unsatisfied with Zr evaluations in ENDF.</td>
<td>ORNL</td>
<td>ORNL</td>
<td>ORNL</td>
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<tr>
<td>Water (H₂O)</td>
<td>Basis TSL evaluation. Water is this most important moderator and moderating reflector material for criticality safety and light water reactor physics. Problems with evaluations submitted by CAB at elevated temperatures (that were noticed during the ENDF/B-VII.0 evaluation process) warrant re-evaluating this essential material using the latest methods developed under LLNL ND2, ND3.</td>
<td>LLNL/NCUS</td>
<td>LLNL/NCUS</td>
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<tr>
<td>Hydrofluoric Acid (HF)</td>
<td>Basis TSL evaluation. HEU-SOL-THERM-039, &quot;Mixture of Uranium (93% U) Hydrofluoric Acid and Hydrofluoric Acid (Low H/U Ratio) in a Hot-Water-Reflected Spherical Tank,&quot; critical experiments overpredict k-eff from 2-6% regardless of cross-section library or code utilized. An appropriate thermal scattering law for the liquid Hydrofluoric acid (HF) moderator will likely resolve this calculational discrepancy.</td>
<td>LLNL/NCUS</td>
<td>LLNL/NCUS</td>
<td></td>
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<tr>
<td>Uranium Hexafluoride (UF₆)</td>
<td>Basis TSL evaluation. As the H/U ratio is &quot;low&quot; in HEU-SOL-THERM-039, correcting for F in UF₆ may be necessary as a moderator. A thermal scattering law for this fissile compound will be useful for the advanced Doppler broadening methods currently under development as LLNL ND5.</td>
<td>LLNL/NCUS</td>
<td>LLNL/NCUS</td>
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</tr>
<tr>
<td>Uranium Metal (U)</td>
<td>Basis TSL evaluation. Requested by the RPI for use in U-235 resonance parameter analysis.</td>
<td>LLNL/NCUS</td>
<td>LLNL/NCUS</td>
<td>LLNL/NCUS</td>
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</tr>
<tr>
<td>Uranium Carbide (UC)</td>
<td>Basis TSL evaluation. A common fissile compound under consideration for high-temperature advanced nuclear reactor fuel. A thermal scattering law for UC will improve Doppler broadening using advanced methods currently under development as LLNL ND5.</td>
<td>LLNL/NCUS</td>
<td>LLNL/NCUS</td>
<td>LLNL/NCUS</td>
<td>LLNL/NCUS</td>
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</table>
# Contributed TSL Evaluations

<table>
<thead>
<tr>
<th>Material</th>
<th>Motivation</th>
<th>NCSP 5 Year Plan</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Heavy oil (hydrogen)</strong></td>
<td>NCSP/NR applications</td>
<td>FY 2020</td>
</tr>
<tr>
<td></td>
<td>No TSL data in ENDF/B-VIII.0</td>
<td>FY 2019</td>
</tr>
<tr>
<td><strong>Liquid hydrogen fluoride</strong></td>
<td>NCSP applications</td>
<td>FY 2021</td>
</tr>
<tr>
<td>(hydrogen)</td>
<td>No TSL data in ENDF/B-VIII.0</td>
<td>FY 2020</td>
</tr>
<tr>
<td><strong>Light water (hydrogen)</strong></td>
<td>NCSP applications</td>
<td>FY 2021</td>
</tr>
<tr>
<td></td>
<td>Extended ENDF/B-VIII.0</td>
<td>FY 2020</td>
</tr>
<tr>
<td><strong>Polyethylene</strong></td>
<td>NCSP applications</td>
<td>----</td>
</tr>
<tr>
<td>(hydrogen)</td>
<td>Extended ENDF/B-VIII.0</td>
<td>----</td>
</tr>
<tr>
<td><strong>Uranium metal</strong></td>
<td>NCSP applications</td>
<td>FY 2022</td>
</tr>
<tr>
<td></td>
<td>No TSL data in ENDF/B-VIII.0</td>
<td>Near completion</td>
</tr>
<tr>
<td><strong>Uranium carbide</strong></td>
<td>NCSP applications</td>
<td>FY 2023</td>
</tr>
<tr>
<td></td>
<td>No TSL data in ENDF/B-VIII.0</td>
<td>ongoing</td>
</tr>
<tr>
<td><strong>Sapphire (Al in Al2O3)</strong></td>
<td>Neutron science / Research Reactors</td>
<td>----</td>
</tr>
<tr>
<td><strong>Sapphire (O in Al2O3)</strong></td>
<td>No TSL data in ENDF/B-VIII.0</td>
<td>----</td>
</tr>
<tr>
<td></td>
<td>(cryogenic temperatures)</td>
<td>----</td>
</tr>
<tr>
<td><strong>20% Porous Reactor Graphite</strong></td>
<td>DOE NE Advanced nuclear reactors</td>
<td>----</td>
</tr>
<tr>
<td><strong>FLiBe (beyllium)</strong></td>
<td>DOE NE Advanced nuclear reactors</td>
<td>----</td>
</tr>
<tr>
<td><strong>FLiBe (flourine)</strong></td>
<td>DOE NE Advanced nuclear reactors</td>
<td>----</td>
</tr>
<tr>
<td><strong>FLiBe (lithium)</strong></td>
<td>DOE NE Advanced nuclear reactors</td>
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</tr>
</tbody>
</table>
TSL Methodology

DFT/LD approach

MD approach
ND2 – New to NNDC
Heavy Paraffinic Oil Data – NCSP/NR
ND2 – New to NNDC
Hydrofluoric Acid Data - NCSP
ND2 – Testing at NR Light Water - NCSP

![Diagram of molecular structure]

**Figure 1:** Schematic representation of the molecular structure of ND2. The diagram illustrates the atomic bonds and their interactions at various temperatures and pressures. The color-coded lines represent the intensity of bonding, with darker lines indicating stronger interactions. The graph on the left shows the density of states (DOS) as a function of energy, while the graph on the right displays the total cross-section (XS) as a function of energy. The inset provides detailed data points for specific temperatures and pressures, highlighting the thermal scattering cross-section across different energy levels.
ND2 – Update to NNDC Polyethylene Data - NCSP
ND2
Uranium Metal – NCSP

- **DFT Model**
  - GGA-PBE, Spin Polarized Calculation
  - Hubbard Term, +U = 1.5 eV
  - No SOC (insignificant impact)
  - 4x2x2 to 4x4x4 Supercells (64-256 atoms)

- **Cross Sections**
  - Uranium total absorption 7.57 b at 2.53 eV
New to NNDC
Molten Salt FLiBe Data – DOE NE
20% Porous Nuclear Graphite – DOE NE
New to NNDC
Single Crystal Sapphire Data - RR
ND3 – **FLASHH**

**Input Module GUI**

- **Coherent Elastic**
- **Incoherent Elastic**

**Elastic Scattering**

- ENDF data format

**Post processing**

- Cross section data

**Inelastic Scattering**

- $S(\alpha,\beta)$

**Integrator**

- Cross section data
- Doppler Broadening
- Angular Binning

**Analyzing and plotting format**

**Output**

**Standard ENDF format**

- Non-cubic incoherent inelastic
- Coherent one-phonon correction
- Inelastic under incoherent approximation

**Liquid physics**
**FLASSH**

- Calculations and ENDF TSL library formatting modules implemented in FORTRAN 95 using a modular design
- Parallel computing realized by OpenMP 4.0 bindings
- GUI implemented by cross platform QT® C++ API

- Error checks
- Input Generator (for both FLASSH and NJOY)
- ENDF / ACE Formatting
- Warning Messages Based on Material Physics
- Crystal Structure Dependent Calculation
**FLASSH**

- **FLASSH GUI**
  - Crystal Structure (Defaults Shown)

![FLASSH GUI Example](image_url)
**FLASSH**

- **FLASSH GUI**
  - Density of States

![FLASSH GUI](image)

Required User Input

![Graphite Density of States](image)
FLASHE

- **GUI: Plotting**
  - FLASHE built-in plotter produces cross section and TSL plots
  - Plots can be used by evaluators to check accuracy of results

- **ACE Output Capability**
  - FLASHE generated ACE files are directly compatible with codes such as MCNP
  - High resolution cross sections without requiring transfer of files
ND3/ND5 – Doppler Analysis

**FLASH** Generalized TSL

- Generalized Non-Cubic Evaluation
  - Exact material structure
  - Direction-dependent calculations averaged into $\alpha/\beta$ space
  - Effects at highest $\alpha$-values

<table>
<thead>
<tr>
<th>$2.33E-03$</th>
<th>$-1.65E-06$</th>
<th>$2.15E-09$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$-1.65E-06$</td>
<td>$2.33E-03$</td>
<td>$-5.65E-09$</td>
</tr>
<tr>
<td>$2.15E-09$</td>
<td>$-5.65E-09$</td>
<td>$1.35E-02$</td>
</tr>
</tbody>
</table>

Exact Debye-Waller Matrix ($\text{Å}^2$)

Graphite TSL data and inelastic cross sections comparing cubic and non-cubic impacts at room temperature.
ND3/ND5 – Doppler Analysis

FLASHH Generalized TSL

- Generalized Self and Distinct (Coherent) Evaluation
  - Exact material structure
  - Direction-dependent calculations averaged into $\alpha/\beta$ space
  - Effects at lowest $\alpha$-values

| $2.33E-03$ | $-1.65E-06$ | $2.15E-09$ |
| $-1.65E-06$ | $2.33E-03$ | $-5.65E-09$ |
| $2.15E-09$ | $-5.65E-09$ | $1.35E-02$ |

Exact Debye-Waller Matrix ($\text{Å}^2$)

Graphite TSL data and inelastic cross sections comparing incoherent cubic and generalized full TSL impacts at room temperature.
ND5 – Doppler Broadening

- **Structure Impacts**
  - Initial testing with cubic UO$_2$ material
    - Significant impacts between free gas and thermal scattering broadening
  - Improvements in TSL theorized to improve Doppler broadening

![Graph showing 238U 6.674eV Resonance: Experimental Cross Section, Free Gas Doppler, TSL Doppler]
FY 21

ND2 – Uranium Carbide

DFT Model

- GGA-PBE +U
- Spin Polarized: AFM-1 between (100) planes
- 2x2x2 Supercell → 64 Atoms (32 U, 32 C)
- 4 Displacements : .03 Å in ± x-direction
FY 21
ND10 – TSL NeTS

- New TSL paradigm
  - DL Neural Thermal Scattering (NeTS) modules

- See papers and presentations
  - ANS 2019 Winter Meeting, Washington, DC, USA
  - PHYSOR 2020 Meeting, Cambridge, UK
FY 21
ND10 – Progressive Development

- Start simple
- Adaptive iteration

Trial Inputs

TSL Inputs

PyTorch
Measurements
Total Cross Section & Structure

NPDF Facility Upgrades – Dual Purpose:

- **Diffraction Measurements:**
  15 New Position Encoding Modules (PEM) – improved diffraction measurement resolution $\Delta d/d$ of $2.9 \times 10^{-3}$ for 3mm holder

- **Transmission Measurement Capabilities:**
  - Monochromator capable of providing beam wavelengths of 1.085 Å, 1.180 Å, 1.479 Å, and 1.762 Å
  - Transmission Detection Apparatus with collimator.
Summary

- Thermal neutron scattering law (TSL) data evaluations have been completed and contributed to ENDF
  - New evaluations are underway

- Advanced TSL techniques in combination with Doppler analysis have been developed

- FLASSH is developing as a modern platform for thermal neutron data analysis

- ML/DL NeTS method development initiated

- Experimental capabilities for validation have been developed at the PULSTAR reactor