Preliminary Designs for Criticality Safety Benchmarks – Iron/Steel/Chromium Series

American Nuclear Society Winter Meeting & Expo, 2019

Nicholas Thompson, Jesson Hutchinson, Theresa Cutler, William Myers, David Hayes

November 20th, 2019
Integral Experiments

• Integral experiments are measurements with a neutron energy spectrum
  – Measurements are not made as a function of energy, but by some other indicator
  – Eg. A nuclear reactor measuring $k_{\text{eff}}$
  – Can be critical, subcritical, above critical, or even non-fissile (pulsed spheres, shielding)

• Valuable to nuclear criticality safety
  – Used to determining upper subcritical limits and biases

• Useful for improving nuclear data
  – Used to identify issues with nuclear data and isotopes/energy regions where nuclear data can be improved
Integral Experiments at NCERC

• The National Criticality Experiments Research Center (NCERC) is a general purpose critical experiments facility – the only one in the US.
• Operated by Los Alamos National Laboratory.
  – Long history of performing critical experiments
• Four critical assemblies
  – Also subcritical assemblies
• Very flexible
  – Can measure many materials – different fuels (HEU, Natural U, Pu, Np)
  – Fast, thermal and intermediate energy spectra
Why Iron?

• Iron is used in pretty much everything
  – Largest component of steel
  – Very important for many applications

• In 2013, CIELO (Collaborative International Evaluation Library Organization) project began
  – Part of OECD/NEA Working Party on Evaluation Cooperation (WPEC) Subgroup 40
  – Collaboration of many of the world’s experts in experiments, theory, simulations, and evaluations
  – They focused on improving nuclear data for the most important nuclides
  – Hydrogen, Oxygen, Iron, $^{235}$U, $^{238}$U, $^{239}$Pu

CIELO Collaboration Summary Results: International Evaluations of Neutron Reactions on Uranium, Plutonium, Iron, Oxygen and Hydrogen


Available online at www.sciencedirect.com


www.elsevier.com/locate/nds
Why Iron?

- So what happened?
- Iron evaluations were improved, but in some cases, lack of data
- For $^{56}\text{Fe}$ in particular:
  - Still issues with the resolved resonance region and fast scattering
  - Chromium and minor isotopes of Fe need to be measured more accurately
  - Potential issues with the low energy background

• The low energy background (from 10 to 100 keV) in $^{56}\text{Fe}$ capture has been partially motivated by performance of the evaluation on a single integral experiment hmi001 (ZPR-34/9). This experiment is, however, highly sensitive to $^{239}\text{Pu}$, as well as to $^{52}\text{Cr}$ and $^{58}\text{Ni}$. Future evaluations of these, and possibly other materials, can modify size of the background in $^{56}\text{Fe}$ capture or make it even redundant.
Why Iron?

Naming convention:
First letter
H = HEU
I = IEU
L = LEU
M = MIX
P = Pu

Second letter
M = Metal
C = Compound
S = Solution

Third letter
F = Fast
I = Intermediate
T = Thermal
M = Mixed

FIG. 30. (Color online) Results of the validation of ENDF/B-VIII.0 in criticality benchmarks sensitive to iron. Results for ENDF/B-VII.1, JENDL-4.0, and JEFF-3.2 are included for comparison.
Why Iron?

- Also an NCSP Priority (Five year plan)
  - NCSP funded evaluation work for Iron, has recognized Iron and Chromium as priorities

### Appendix B
Nuclear Data

| Priority Needs */ Additional Needs | Thermal scattering (Paraffinic Oil, HF, Silicone Oil, UO₂F₂, PuH₂, UH₃, Paraffin, U₃O₈, U₃Si₂, UC, PuO₂, etc.), ²³⁵Pu, Fe, Cr, ²³⁷Np, Pb, ⁵⁵Mn, Ti, ²⁴⁰Pu/²³⁵U, Th, Be, ³¹V, Zr, F, K, Ca, Mo, Na, La |

#### 2018

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Iron (⁵⁶Fe)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**Basis**

Revise high energy resonance region evaluation. Iron is a key element of structural materials in the DOE Complex (e.g., steel) and is used in many configurations (e.g., tanks, piping, admixed material that can serve as neutron absorber, etc.). ⁵⁶Fe has numerous resonances in the resonance range. Currently, the latest ⁵⁶Fe evaluation in the ENDF/B data files does not have detailed resonance parameters; rather, the evaluation provides a pointwise representation. The ⁵⁶Fe resonance evaluation will significantly improve radiation transport calculations for systems involving iron (i.e., critical benchmark analyses and criticality safety analyses of processes in the DOE Complex). Evaluation work was performed at IRSN in the past but was not apparently included in ENDF (this will be reviewed and considered for inclusion in ENDF).
Why Iron?

• Intermediate Iron (Steel) and Chromium benchmarks were also listed as priorities in 2011 – slide from Dr. Catherine Percher of LLNL

**Thermal/Epithermal eXperiments (TEX)**

• TEX Goals
  – Using available US Department of Energy fissile materials, create critical benchmarks to address the nuclear data and validation needs for criticality safety
  – July 2011 at Sandia National Laboratories, Albuquerque, NM
    • Representatives from US, UK, and France
    • Main take-aways
      – Intermediate spectrum experiments needed (only 2.1% of ICSBEP Benchmarks)
      – Test-bed assemblies that span multiple energy spectra are incredibly useful for nuclear data validation
      – Consensus prioritization of nuclear data needs (in order):
        • $^{239}$Pu, $^{240}$Pu, $^{238}$U, $^{235}$U, Temperature variations, Water density variations, Steel, Lead (reflection), Hafnium, Tantalum, Tungsten, Nickel, Molybdenum, Chromium, Manganese, Copper, Vanadium, Titanium, and Concrete (reflection, characterization, and water content)
      – LLNL and LANL completed 3 critical configurations in FY17 and 17 configurations in FY18
Why Iron?

• Cross Section Evaluation Working Group meeting was Nov 4-8
• Discussion of Iron – slide from Dr. Andrej Trkov of IAEA:

Iron cross sections

• CIELO=ENDF/B-VIII.0 evaluation
• Inelastic cross sections from Geel by Negret
• Leakage spectra from thick spheres with $^{252}$Cf source are under-predicted by up to 40% from 2 MeV to 6 MeV (shown by Simakov just before the release of ENDF/B-VIII.0)
• Patch to the Fe-56 is available that
  – Shows good performance in leakage spectra from thick spheres
  – Removes some deficiencies (e.g. near 300 keV)

New resonance evaluation is needed (some progress was made with L. Leal, including direct capture contribution)
Why Iron?

• Dr. Luiz Leal at IRSN is currently working on $^{54}\text{Fe}$ and $^{56}\text{Fe}$ evaluations.
• Additionally, J-PARC accelerator will be measuring transmission and capture below 50 keV for $^{54}\text{Fe}$, $^{56}\text{Fe}$ and $^{57}\text{Fe}$ using enriched samples.
• Finally, lack of benchmarks sensitive to Iron (especially in the intermediate energy region).
Why Chromium?

- Chromium is a large component of stainless steels (up to 20%)
- Also a high priority need, also lacks benchmarks
- Problems with resonances in 50Cr and 53Cr near 5 keV
- Slide from Dr. Trkov – one note, ZPR-6/10 (PMI-2) is an outlier

Chromium cross sections

- There is a problem with the ~5 keV resonances in $^{50,53}\text{Cr}$
- Raw measurements from ORNL/JRC and RPI agree in shape, but differ strongly after multiple scattering corrections
- ZPR-6/10 benchmark is highly sensitive to Cr cross sections near 5 keV
- New measurement is needed to resolve the discrepancy in measured data? (e.g. Lead-Slowing-Down measurement)
Why Iron and Chromium?

- Number of benchmarks sensitive to iron and chromium as a function of energy

**Natural Iron:**

\[ ^{54}\text{Fe} \approx 5.85\% \]
\[ ^{56}\text{Fe} \approx 91.75\% \]
\[ ^{57}\text{Fe} \approx 2.12\% \]
\[ ^{58}\text{Fe} \approx 0.28\% \]

**Nat Chromium:**

\[ ^{50}\text{Cr} \approx 4.345\% \]
\[ ^{52}\text{Cr} \approx 83.789\% \]
\[ ^{53}\text{Cr} \approx 9.501\% \]
\[ ^{54}\text{Cr} \approx 2.365\% \]
Why Iron and Chromium?

To summarize:
• Lack of intermediate benchmarks
• Issues with existing nuclear data
• Ongoing measurement and evaluation work
• New benchmarks will be useful for validating the quality of these evaluations

But why measure iron and chromium?
• Some issues with $^{56}\text{Fe}$ nuclear data are caused by lack of information on minor iron isotopes and chromium.
• By making integral measurements of various iron and chromium in different ratios, can help isolate the impact of chromium.
Proposed measurement series

- Three sets of material, each with multiple measurements
  - Basically an extension of the Zeus series
  - Carbon Steel (~99% Iron)
  - Stainless Steel 304 (or an iron-chromium alloy)
  - IronClad/C26M/FeCrAl
    - Accident Tolerant Cladding material – being tested in multiple commercial reactors
    - Developed by ORNL, GE Global Nuclear Fuel is manufacturing

<table>
<thead>
<tr>
<th></th>
<th>Carbon Steel AISI 1018</th>
<th>IronClad/C26M</th>
<th>SS 304</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe</td>
<td>98.81-99.26</td>
<td>79.95</td>
<td>66.5-74.0</td>
</tr>
<tr>
<td>Cr</td>
<td>0</td>
<td>12</td>
<td>18.0-20.0</td>
</tr>
<tr>
<td>Ni</td>
<td>0</td>
<td>0</td>
<td>8.0-10.5</td>
</tr>
<tr>
<td>Mn</td>
<td>0.60-0.90</td>
<td>0</td>
<td>≤2.0</td>
</tr>
<tr>
<td>Al</td>
<td>0</td>
<td>6</td>
<td>0</td>
</tr>
<tr>
<td>Si</td>
<td>0</td>
<td>0</td>
<td>≤0.75</td>
</tr>
<tr>
<td>N</td>
<td>0</td>
<td>0</td>
<td>≤0.10</td>
</tr>
<tr>
<td>C</td>
<td>0.14-0.20</td>
<td>0</td>
<td>≤0.08</td>
</tr>
<tr>
<td>P</td>
<td>≤0.04</td>
<td>0</td>
<td>≤0.045</td>
</tr>
<tr>
<td>S</td>
<td>≤0.05</td>
<td>0</td>
<td>≤0.03</td>
</tr>
<tr>
<td>Mo</td>
<td>0</td>
<td>2</td>
<td>0</td>
</tr>
<tr>
<td>Y</td>
<td>0</td>
<td>0.05</td>
<td>0</td>
</tr>
</tbody>
</table>
Proposed measurement series

• Will use the Comet critical assembly
• Initial design
  – Cylindrical metal plates
  – HEU Jemima plates (blue)
  – Iron/Steel plates (red)
  – Moderator plates (green)
• Energy spectra will be tuned by adding or removing moderator plates
• Goal
  – 1 fast configuration
  – 1 fast/intermediate configuration
  – 1 intermediate configuration
Previous measurements

- ZEUS – HEU plates, high density polyethylene (HDPE), carbon steel
  - HEU-MET-FAST-72 in ICSBEP
- Three cases
  - Case 1 and 2 – no HDPE
  - Case 3 – 12 plates of HDPE
- Case 3 was almost 50% Fast, 50% Intermediate

ZEUS: FAST-SPECTRUM CRITICAL ASSEMBLIES WITH AN IRON - HEU CORE SURROUNDED BY A COPPER REFLECTOR
Design optimization process

- Currently have a basic idea, partially based on previous ZEUS measurements and other constraints (size, weight, fuel availability, etc.)
  - Foil irradiations may also be added to obtain spectral information
- Newer experiments LANL has designed have much higher intermediate fluxes
- LANL has developed optimization tools to help design critical experiments as part of the ARCHIMEDES LANL LDRD
  - Isaac Michaud gave a talk on this on Tuesday:
    “Designing Critical Experiments Using Gaussian Process Optimization”

- Optimization tools will be used to determine:
  - Best moderating material and thicknesses
  - Best reflector material
    - Currently copper is used as a reflector material – to maximize sensitivity to iron, an iron reflector may better
Looking for input/feedback

- What would make this experiment most useful to you?
Thank you!

• This work was supported by the DOE Nuclear Criticality Safety Program, funded and managed by the National Nuclear Security Administration for the Department of Energy.

• This work also used research supported by the U.S. Department of Energy LDRD program at Los Alamos National Laboratory.