# Pulsed Neutron Die Away Experiments at Lawrence Livermore National Laboratory

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# Introduction

- Starting new integral experiment campaign at LLNL
- Aims to avoid compensating effects associated with criticality experiments
- Specifically target thermal neutron scattering law data
- Revive pulse neutron die-away experiments
  - J. Holmes at NNL

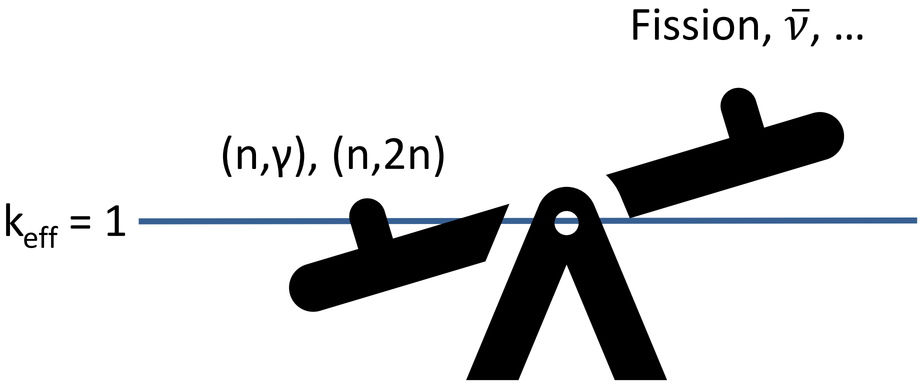
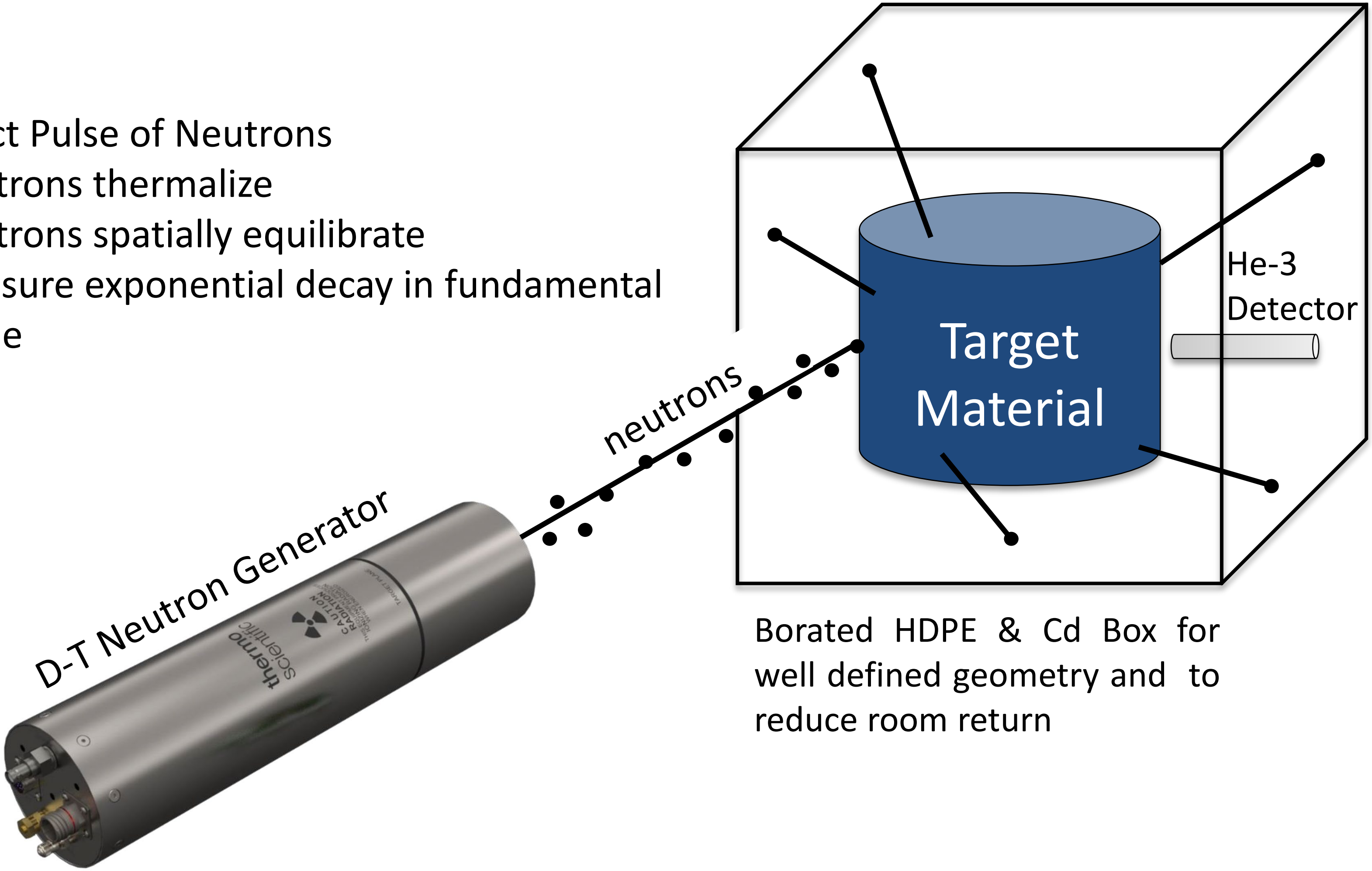


Figure: Compensating the criticality of an experiment with changes to different nuclear data

# Pulsed Neutron Die Away Experiments

1. Inject Pulse of Neutrons
2. Neutrons thermalize
3. Neutrons spatially equilibrate
4. Measure exponential decay in fundamental mode



Borated HDPE & Cd Box for well defined geometry and to reduce room return

# Integral Parameter: α eigenvalue

$$\phi(t) = \phi_0 \exp(-\alpha t) + R$$

$$\alpha = \overline{v\Sigma_a} + \overline{vD_0}\, B_0^2 - C B_0^4 + \cdots$$

- α: flux decay-time eigenvalue [$s^{-1}$]
- $D_0$ [$cm^2$-$s^{-1}$] is the asymptotic diffusion coefficient
- C: “cooling coefficient” [$cm^2$]
- $B_0^2$: geometric Buckling [$cm^{-2}$]
- *v* thermal neutron velocity (2.2 x $10^5$ cm/s)
- $\Sigma_a$ macroscopic absorption cross section [$cm^{-1}$]

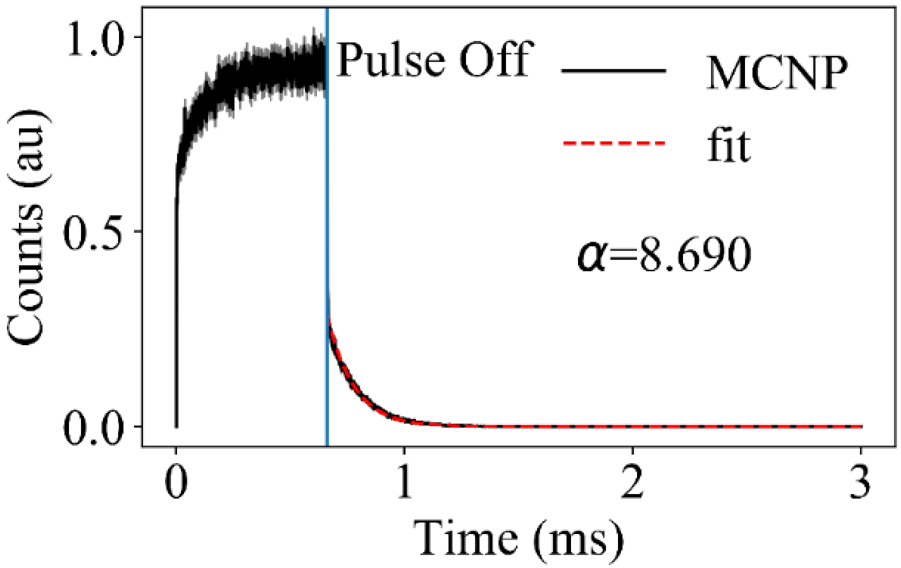


Figure: Example of pulsed-die-away curve modeled in MCNP

# Historical Experiments

- In 1950-70s, PNDA experiments were used to experimentally determine diffusion parameters of various moderators
  - Diffusion coefficient
  - Macroscopic absorption cross section
  - Extrapolation distance

Table: Examples of previous PNDA experiments

| Experiment | Target Material |
|---|---|
| **von Dardel & Sjostrand (1954)** | $H_2O$ |
| **Bracci & Coceva (1956)** | $H_2O$ |
| **deSaussure & Silver (1959)** | Beryllium |
| **Adam, Bod, Pal (1960)** | Diphenyl |
| **Serdula & Young (1965)** | Graphite |
| **Ritchie (1968)** | BeO |
| **Silver (1968)** | $H_2O$ Ice |
| **Saleita & Robeson (1971)** | $H_2O/D_2O$/Ice |
| **Drozdowicz & Woznicka (1987)** | Plexiglass |
| **Drozdowicz, Gillette (1999)** | Polyethylene |

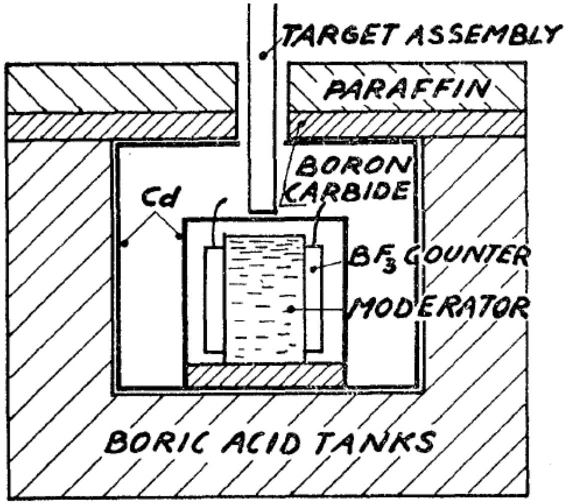


Figure: Experimental setup from von Dardel & Sjostrand

G. von Dardel and N. G. Sjostrand, "Diffusion Parameters of Thermal Neutrons in Water," Physical Review, vol. 96, no. 5, pp. 1245-1249, 1954.

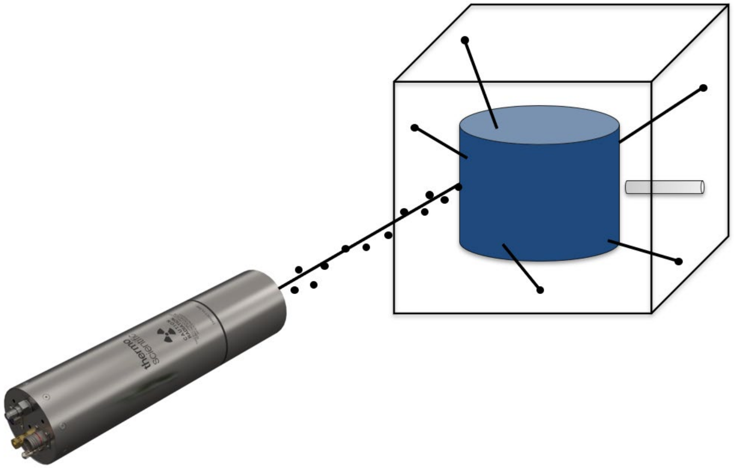
# Why PNDA for TSL Validation and Adjustment?

- Does not require fissile material
  - Non-nuclear facilities, reduced costs, fewer regulations, safer

- Very simple target shapes and compositions
  - Reduced uncertainties in benchmarks
  - Reduced material costs
  - Easy to change temperature

- Only sensitive to absorption and scattering of target medium
  - Reduces uncertainties from other nuclear data and compensating effects
  - Tune target size to vary effect of absorption vs. scattering

- Well conducted experiments have uncertainties of 0.1% - 0.5%



# Sensitivity Depends on Target Size

- Small targets (large Bucklings) are more sensitive to scattering
- Large targets (small Bucklings) are more sensitive to absorption

$$B_0^2 = \left(\frac{\pi}{H + 2\delta}\right)^2 + \left(\frac{2.405}{R + \delta}\right)^2$$

$$\alpha = \underbrace{\overline{v\Sigma_a}}_{\text{Absorption}} + \underbrace{\overline{vD_0}\, B_0^2 - C B_0^4}_{\text{Scattering}}$$

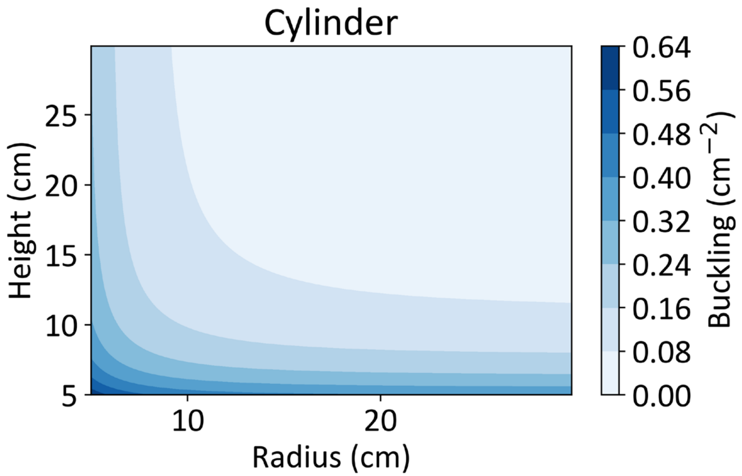


Figure: Buckling vs. cylinder dimensions

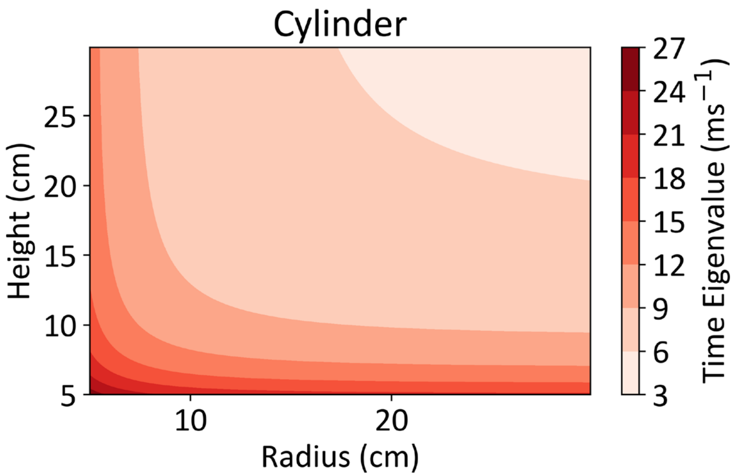


Figure: α vs. cylinder dimensions

# Sensitivity to TSLs

- Example: Historical water experiment in cylindrical geometry
  - A. Bracci & C. Coceva, “The diffusion parameters of thermal neutrons in water.” *Il Nuovo Cimento*, **4** (1956)

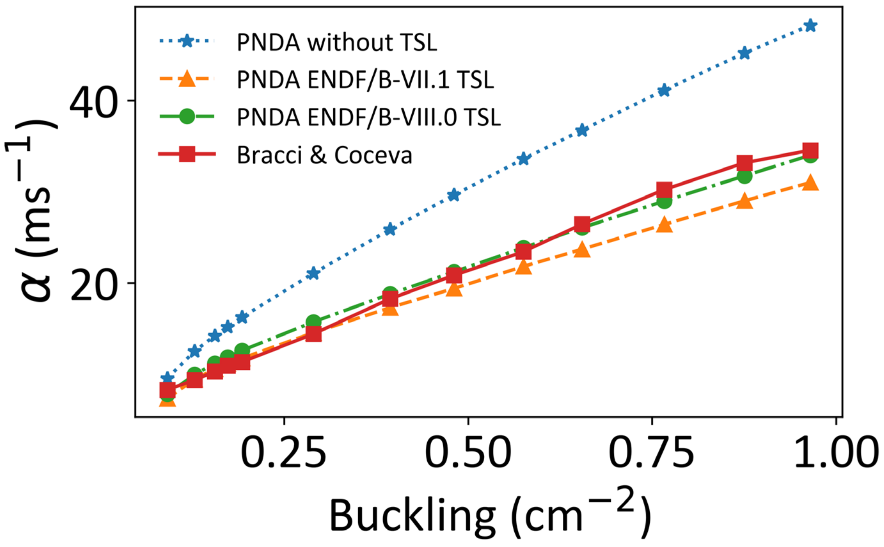


Figure: α vs. Buckling curve for experimental and simulated data

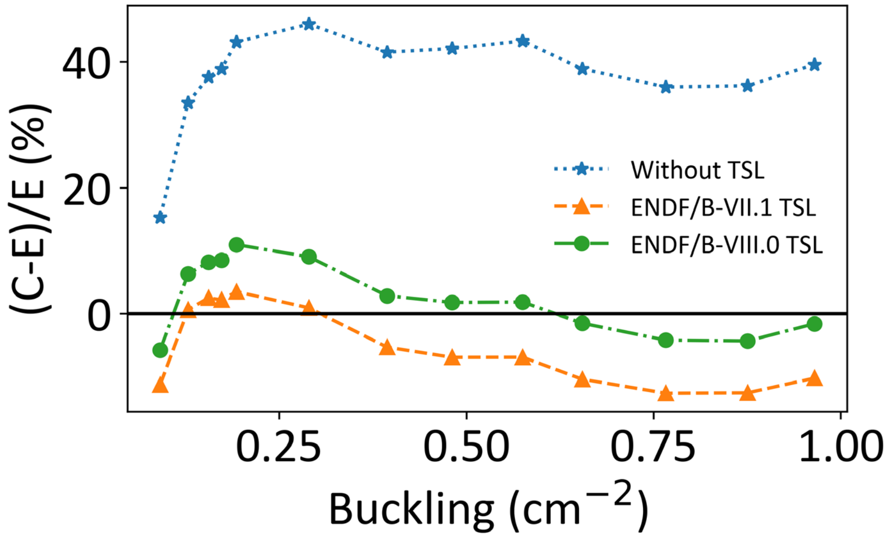


Figure: Bias of simulations without TSLs, with ENDF/B-VII.1, and with ENDF/B-VIII.0 TSLs

# Small Targets vs. Statistics

- Small targets desirable for sensitivity to TSLs
- They quickly leak neutrons, leading to poor detector statistics and poor fit of exponential decay

$$\phi_{fit}(t) = \phi_0 \exp(-\alpha t) + R$$

$$\chi^2 = \sum_i \left(\phi_{data}^{(i)} - \phi_{fit}^{(i)}\right)^2$$

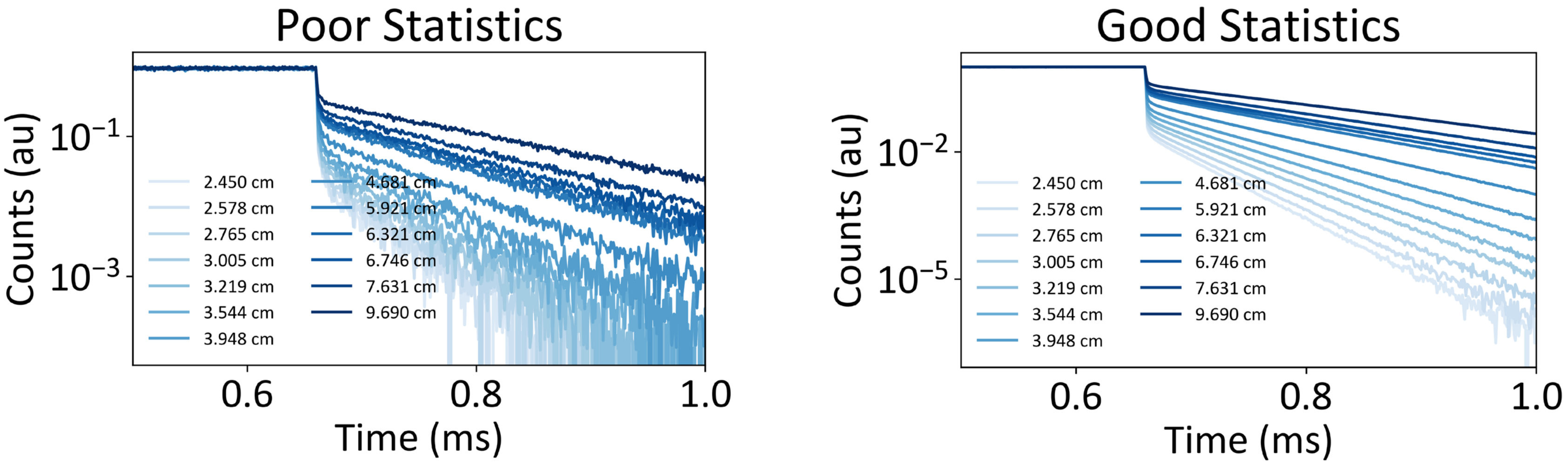
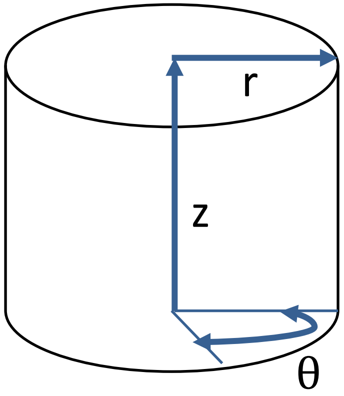


Figure: Statistical effects on die away curves with varying target sizes

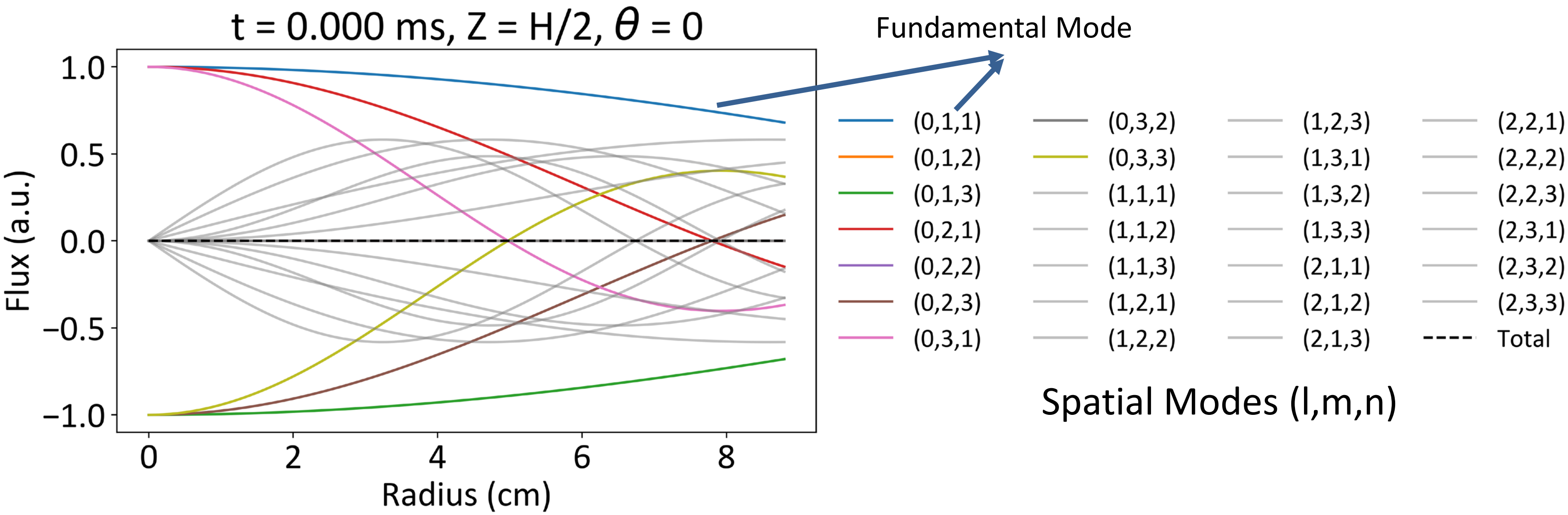
# Decay to Fundamental Mode

## Large Cylindrical Sample

$$\phi(r,\theta,z,t) = \sum_{l,m,n} C_{l,m,n} \sin\left(\frac{n\pi}{H}z\right) J_l\left(a_{l,n} r\right) \cos l\theta \, \exp\Big[-\big(\underbrace{\overline{v\Sigma_a} + \overline{vD_0}\, B^2_{n,m,l}}_{\alpha_{l,m,n}}\big)t\Big]$$



Focusing only on modes of Bessel function:



# Decay to Fundamental Mode

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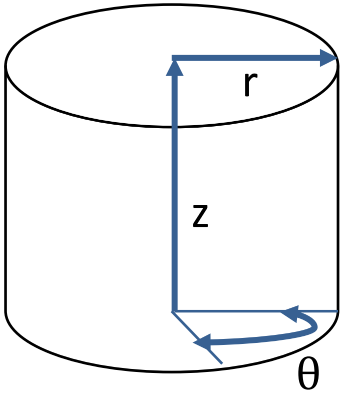

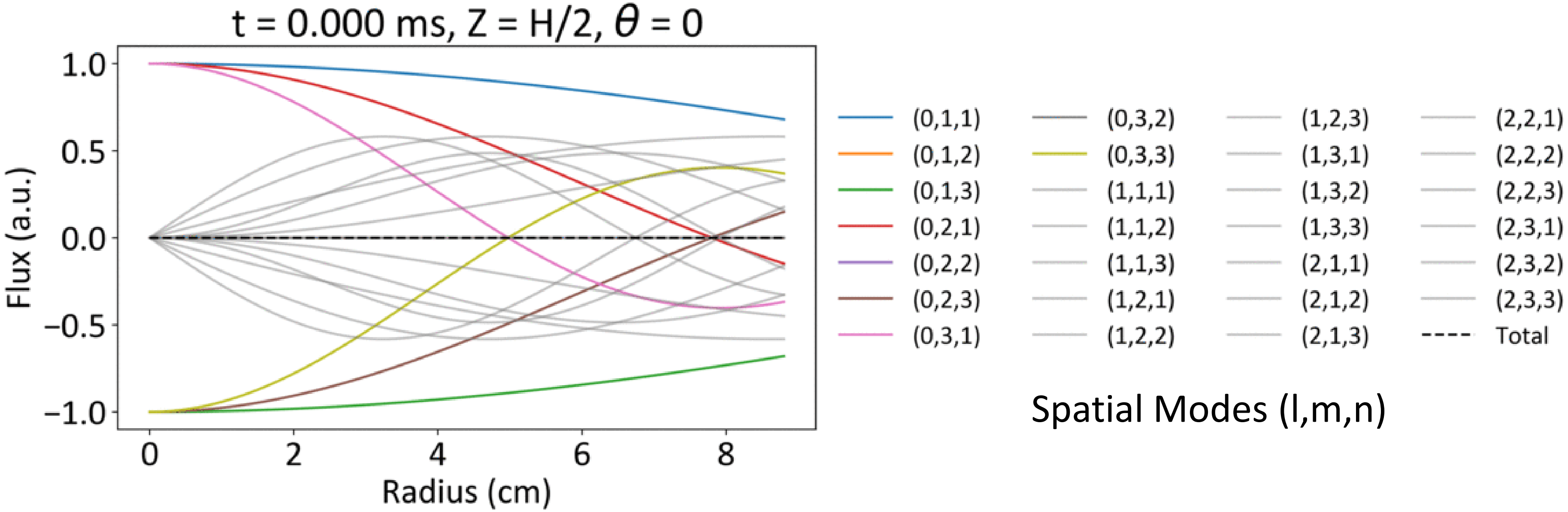

Spatial Modes (l,m,n)

# Decay to Fundamental Mode

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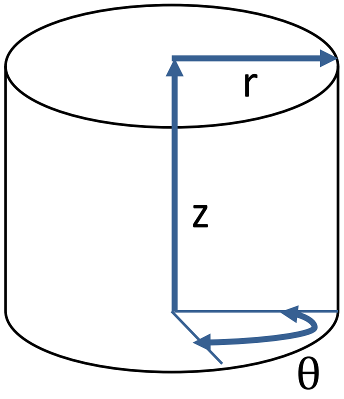
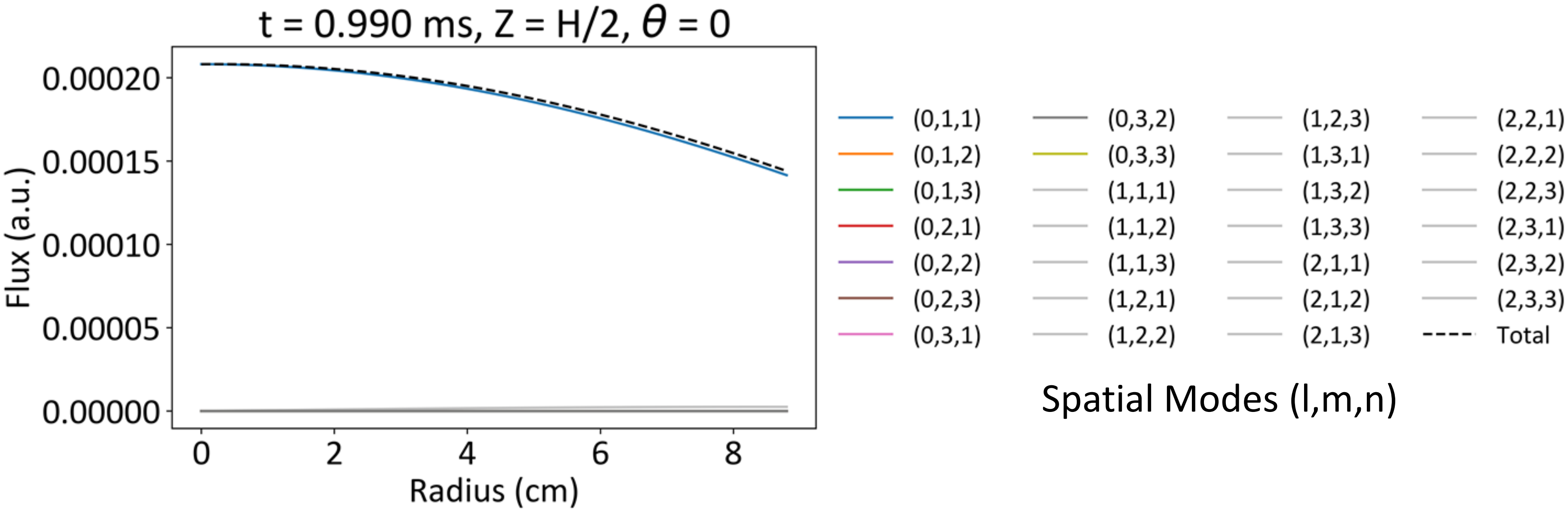


Flux in fundamental mode:


t = 0.990 ms, Z = H/2, θ = 0

Spatial Modes (l,m,n)

# Previous Data Assimilation Efforts for TSLs

- Preeminent work by D. Rochman and A.J. Koning
  - “Random Adjustment of the H in $H_2O$ Neutron Thermal Scattering Data.” *Nuclear Science and Engineering,* **17**, 2012
- Adjust model parameters in LEAPR (NJOY) with BMC-like method

$$F = 10^{\sqrt{\frac{1}{N}\sum(\log(E_i)-\log(C_i))^2}}$$

- Focused on H in $H_2O$ data at 293K
- Used criticality benchmarks
  - PST1, PST12, LSTL4, LMT1, LCT7, HST42…
- Demonstrated some non-linear effects
  - Depending on prior

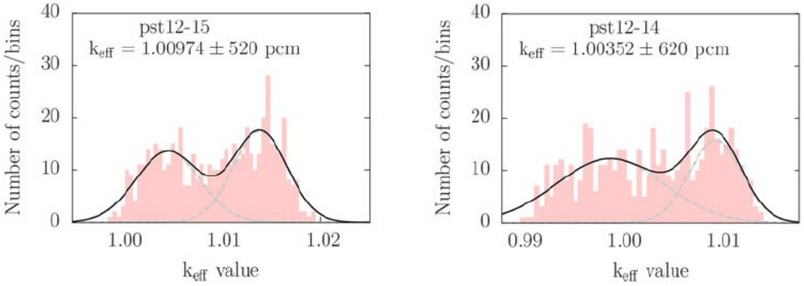


Figure: $k_{eff}$ distributions after sampling LEAPR parameters. From Rochman & Koning (2012)

# Proposed Adjustment Exercises

- **First Exercise:**
  - Replicate Rochman and Koning's approach
    - Using PNDA results ($H_2O$ should be completed summer 2021)
    - Using advancements in BMC methodology since 2012
  - Verify adjusted TSLs with criticality experiments
    - Avoiding compensation effects

- **Second Exercise:**
  - Common Scenario: No TSL data exists, use something close
    - Example: oil present, use H-$H_2O$ data
  - PNDA experiment is easier than new TSL evaluation from scratch
  - Using PNDA $\alpha$ eigenvalues, calibrate a new TSL
  - Verify with H-$H_2O$ → D-$D_2O$
    - Compare calibrated D-$D_2O$ to actual evaluation
  - Test with H-$H_2O$ → X-in-Oil
    - Assess with Rocky Flats critical experiments (HMF-48, HMM-11, PMF-42)

# Future Experiments

- Step through high-priority moderating materials
  - With existing TSLs
  - That are important and hopefully will soon have TSLs
- Current focus is on
  - Criticality safety
  - Hydrogenous materials
  - Room temperature
- Future focus
  - Non-hydrogenous
  - Low and high temperature scenarios
    - Hot Nevada desert
    - Idaho in winter

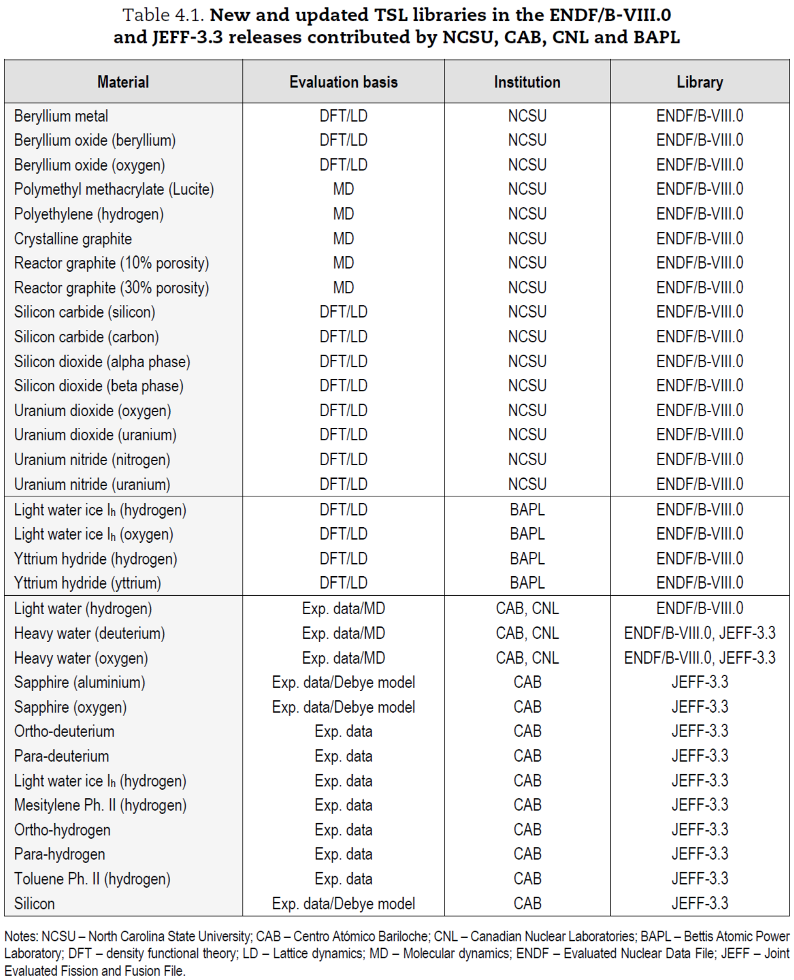


Table 4.1. **New and updated TSL libraries in the ENDF/B-VIII.0 and JEFF-3.3 releases contributed by NCSU, CAB, CNL and BAPL**

| Material | Evaluation basis | Institution | Library |
|---|---|---|---|
| Beryllium metal | DFT/LD | NCSU | ENDF/B-VIII.0 |
| Beryllium oxide (beryllium) | DFT/LD | NCSU | ENDF/B-VIII.0 |
| Beryllium oxide (oxygen) | DFT/LD | NCSU | ENDF/B-VIII.0 |
| Polymethyl methacrylate (Lucite) | MD | NCSU | ENDF/B-VIII.0 |
| Polyethylene (hydrogen) | MD | NCSU | ENDF/B-VIII.0 |
| Crystalline graphite | MD | NCSU | ENDF/B-VIII.0 |
| Reactor graphite (10% porosity) | MD | NCSU | ENDF/B-VIII.0 |
| Reactor graphite (30% porosity) | MD | NCSU | ENDF/B-VIII.0 |
| Silicon carbide (silicon) | DFT/LD | NCSU | ENDF/B-VIII.0 |
| Silicon carbide (carbon) | DFT/LD | NCSU | ENDF/B-VIII.0 |
| Silicon dioxide (alpha phase) | DFT/LD | NCSU | ENDF/B-VIII.0 |
| Silicon dioxide (beta phase) | DFT/LD | NCSU | ENDF/B-VIII.0 |
| Uranium dioxide (oxygen) | DFT/LD | NCSU | ENDF/B-VIII.0 |
| Uranium dioxide (uranium) | DFT/LD | NCSU | ENDF/B-VIII.0 |
| Uranium nitride (nitrogen) | DFT/LD | NCSU | ENDF/B-VIII.0 |
| Uranium nitride (uranium) | DFT/LD | NCSU | ENDF/B-VIII.0 |
| Light water ice $I_h$ (hydrogen) | DFT/LD | BAPL | ENDF/B-VIII.0 |
| Light water ice $I_h$ (oxygen) | DFT/LD | BAPL | ENDF/B-VIII.0 |
| Yttrium hydride (hydrogen) | DFT/LD | BAPL | ENDF/B-VIII.0 |
| Yttrium hydride (yttrium) | DFT/LD | BAPL | ENDF/B-VIII.0 |
| Light water (hydrogen) | Exp. data/MD | CAB, CNL | ENDF/B-VIII.0 |
| Heavy water (deuterium) | Exp. data/MD | CAB, CNL | ENDF/B-VIII.0, JEFF-3.3 |
| Heavy water (oxygen) | Exp. data/MD | CAB, CNL | ENDF/B-VIII.0, JEFF-3.3 |
| Sapphire (aluminium) | Exp. data/Debye model | CAB | JEFF-3.3 |
| Sapphire (oxygen) | Exp. data/Debye model | CAB | JEFF-3.3 |
| Ortho-deuterium | Exp. data | CAB | JEFF-3.3 |
| Para-deuterium | Exp. data | CAB | JEFF-3.3 |
| Light water ice $I_h$ (hydrogen) | Exp. data | CAB | JEFF-3.3 |
| Mesitylene Ph. II (hydrogen) | Exp. data | CAB | JEFF-3.3 |
| Ortho-hydrogen | Exp. data | CAB | JEFF-3.3 |
| Para-hydrogen | Exp. data | CAB | JEFF-3.3 |
| Toluene Ph. II (hydrogen) | Exp. data | CAB | JEFF-3.3 |
| Silicon | Exp. data/Debye model | CAB | JEFF-3.3 |

Notes: NCSU – North Carolina State University; CAB – Centro Atómico Bariloche; CNL – Canadian Nuclear Laboratories; BAPL – Bettis Atomic Power Laboratory; DFT – density functional theory; LD – Lattice dynamics; MD – Molecular dynamics; ENDF – Evaluated Nuclear Data File; JEFF – Joint Evaluated Fission and Fusion File.

# Questions, Comments, Discussion

- **References:**

  - G. von Dardel and N. G. Sjostrand, "Diffusion Parameters of Thermal Neutrons in Water," Physical Review, vol. 96, no. 5, pp. 1245-1249, 1954.
  - J. Holmes, M. Zerkle and D. Heinrichs, "Benchmarking a first-principles thermal neutron scattering law for water ice with a diffusion experiment," *EPJ Web of Conferences,* vol. 146, p. 13004, 2017.
  - J. Holmes, M. Zerkle and A. Hawari, "Validation of Thermal Scattering Laws for Light Water at Elevated Temperatures with Diffusion Experiments," in *PHYSOR 2020: Transition to a Scalable Nuclear Future*, Cambridge, United Kingdom, 2020.
  - D. Siefman, E. Heckmaier, W. Zwyiec, D. Heinrichs, “IER-501 CED-1: Preliminary Design of a New Pulsed-Neutron Die-Away Experimental Testbed for Thermal Scattering Law Benchmarks (PNDA),” *Lawrence Livermore National Laboratory*, LLNL-TR-820718, 2021

Lawrence Livermore National Laboratory

# PU-MET-FAST-042

NEA/NSC/DOC/(95)03/I
Volume I

PU-MET-FAST-042

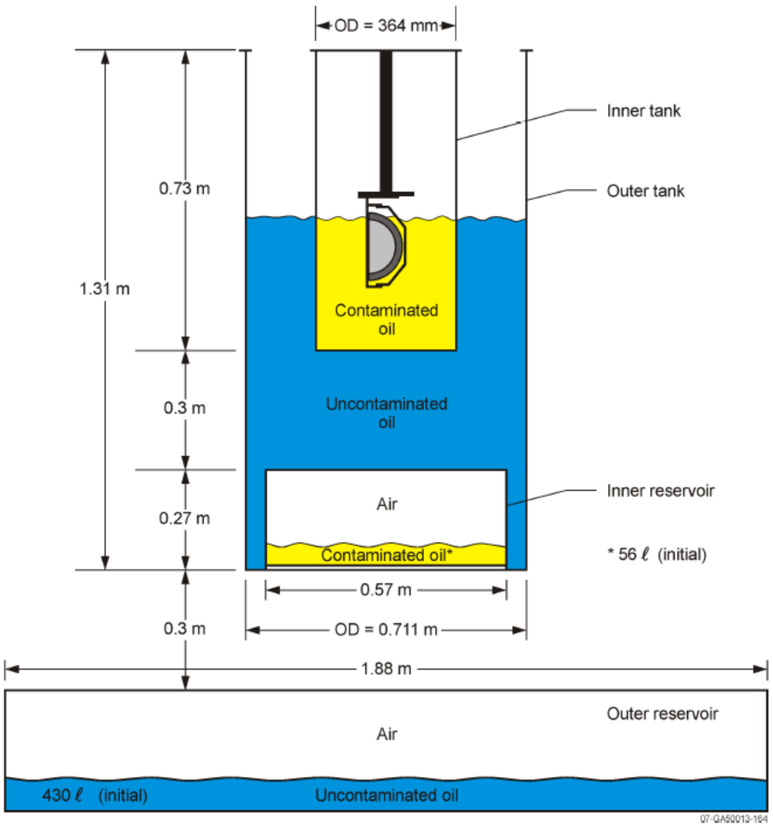


Figure 3. Schematic of the Experimental Setup.

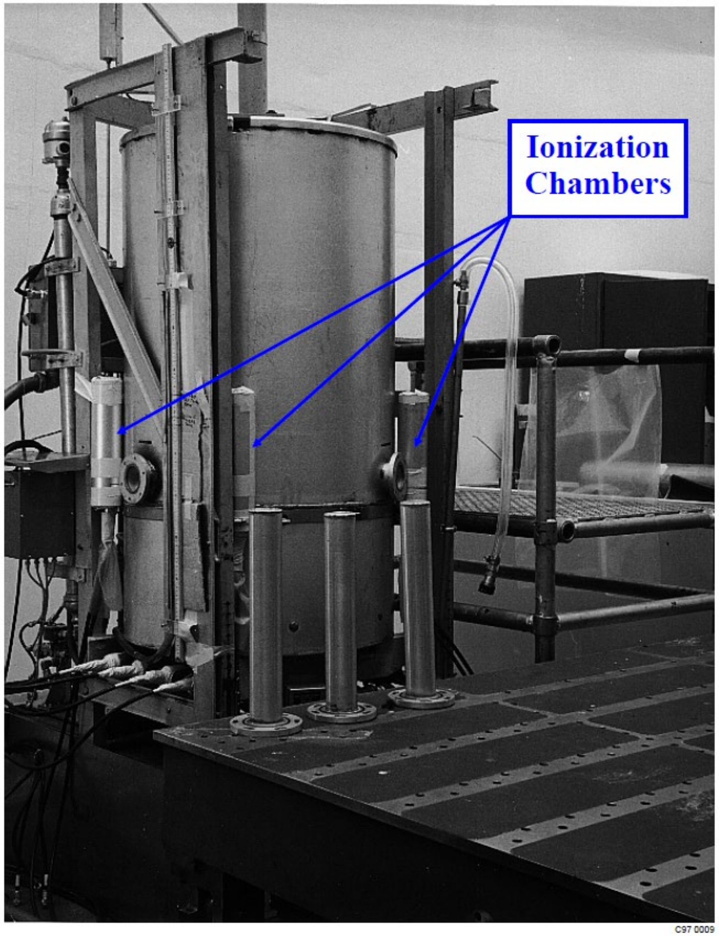


Figure 4. Photograph of the Outer Tank.

# HEU-MET-FAST-048

HEU-MET-FAST-048

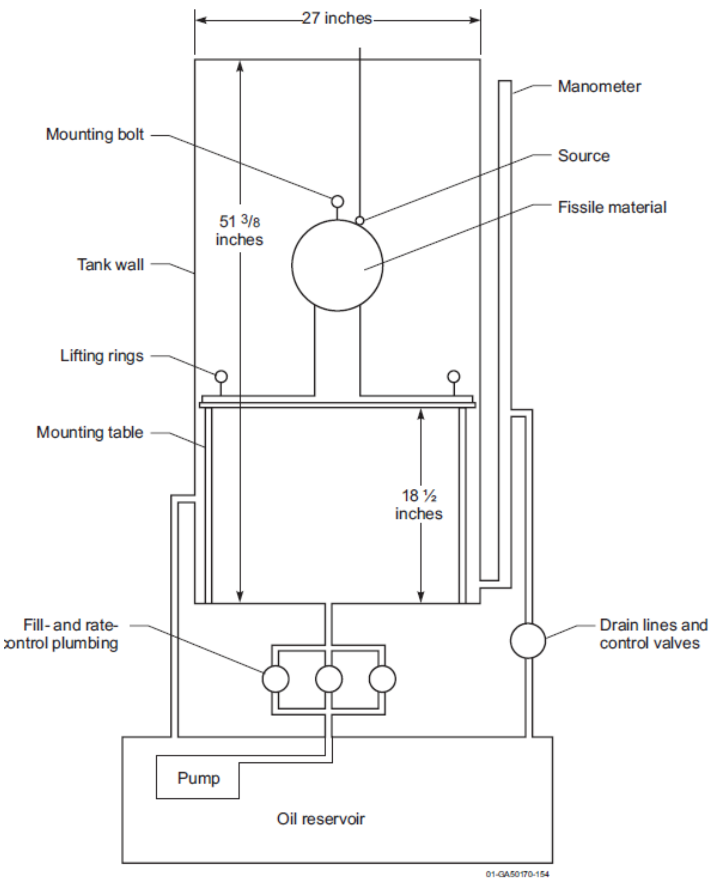


Figure 3. General Schematic of the Experimental Setup.

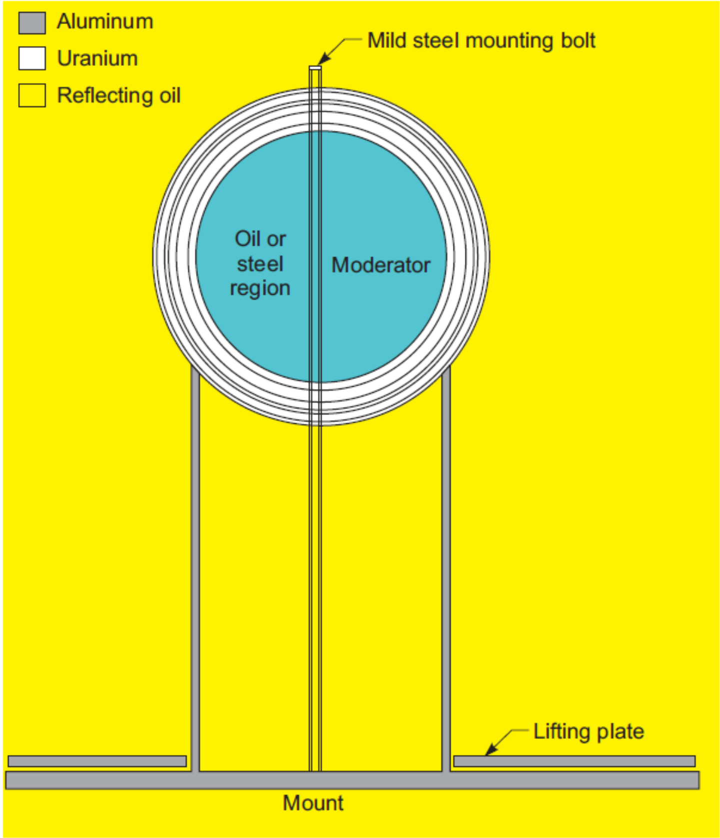


Figure 4. Generic Mount and Core Arrangement for Spherical Assemblies.