

NE-UP: Improved Fission Neutron Data Base for Active Interrogation of Actinides

S.A. Pozzi, A. Enqvist, B. Wieger, and M. Flaska

*Department of Nuclear Engineering and Radiological Sciences,
University of Michigan, Ann Arbor, Michigan*

R.C. Haight

LANSCCE, Los Alamos National Laboratory, Los Alamos, New Mexico

J.B. Czirr, L.B. Rees

Department of Physics, Brigham Young University, Provo, Utah

M.A. Kovash

Department of Physics and Astronomy, University of Kentucky, Kentucky

P.V. Tsvetkov and M. Cuvelier

*Department of Nuclear Engineering, Texas A&M
University, College Station, Texas*



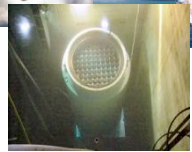
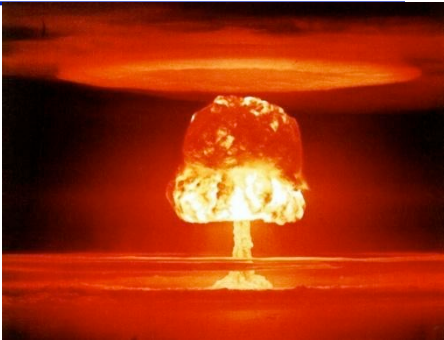
Detection for Nuclear
Nonproliferation Group



Grand Challenges for Nuclear Area

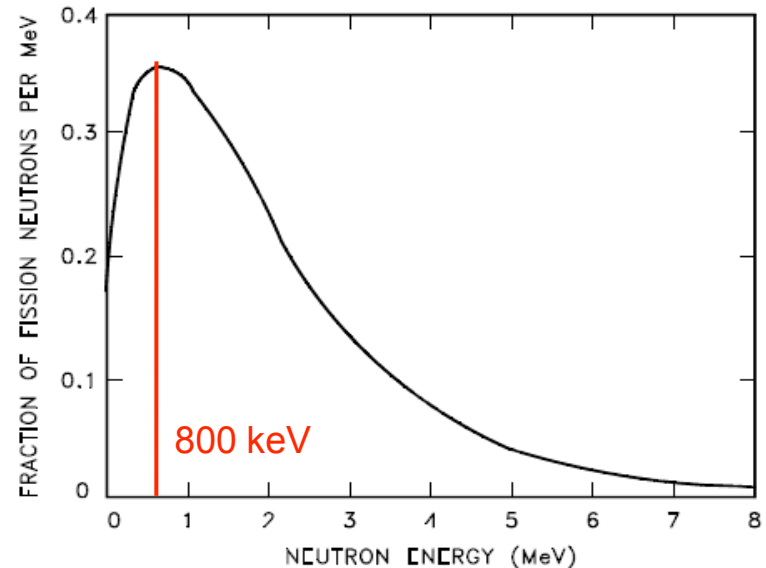
Nuclear Nonproliferation

- **Preventing Nuclear Terrorism**
- **Safeguarding Nuclear Fuel**
 - Requires advanced MC&A techniques to prevent diversions, ensure safety, and reassure the international community
 - Real-time accountability measurements for materials at all stages of the fuel cycle
 - Quantification of ^{239}Pu and other fissile isotopes
- **^3He -Replacement Technology – Candidates:**
 - High efficiency
 - Reliable neutron/gamma-ray discrimination
 - Neutron spectroscopic capabilities



Improved Fission Neutron Data Base for Active Interrogation of Actinides FY 2010 - 2013

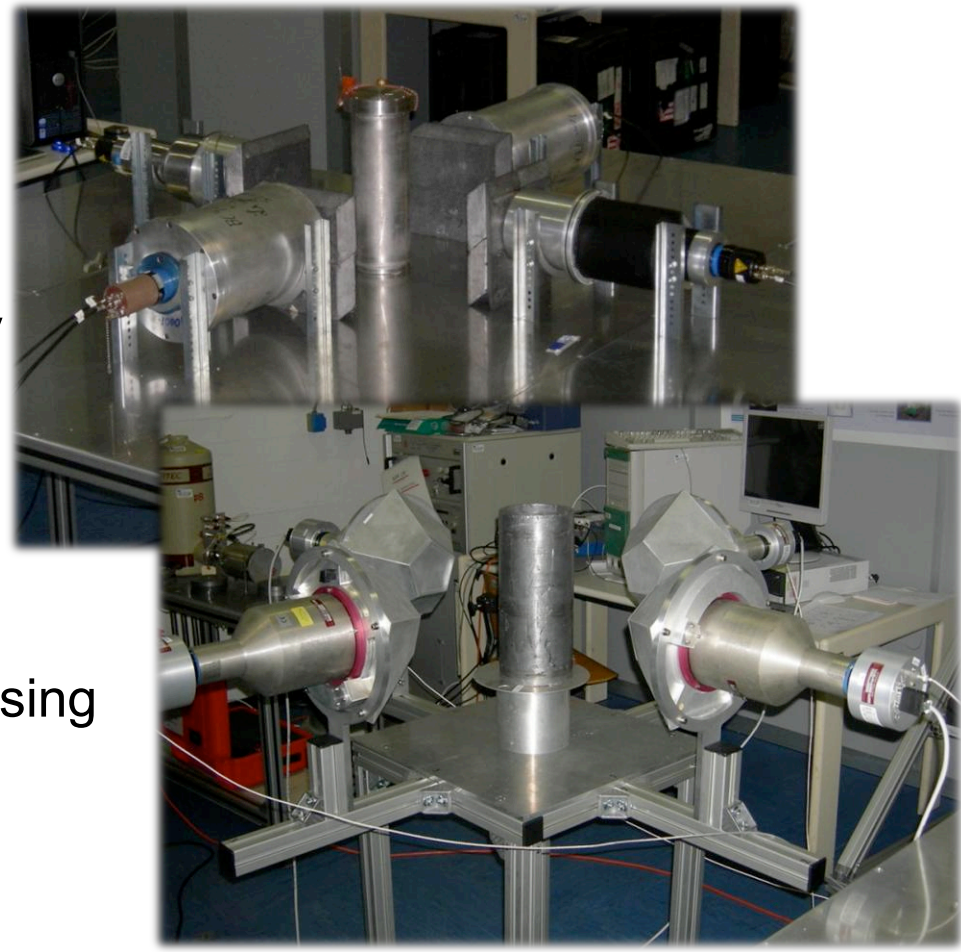
- Fission spectrum data libraries show inconsistencies and/or lack of data for relevant induced fission neutron spectra.
- Main region of interest where data are less reliable:
 - Below 1 MeV
 - Above 5 MeV
- Liquid scintillation detectors with optimized pulse shape discrimination (PSD) capabilities can be used in these ranges.



Project Leader

DNNG, University of Michigan

- Measurement systems based on a variety of liquid and plastic organic scintillators, including capture-gated scintillators
- These systems enable a variety of **unique measurements**:
 - Pulse height distributions
 - Time of flight
 - Cross correlations
 - Neutron/gamma multiplicity
- Measurements are performed using fast waveform digitizers and custom-made data-processing algorithms

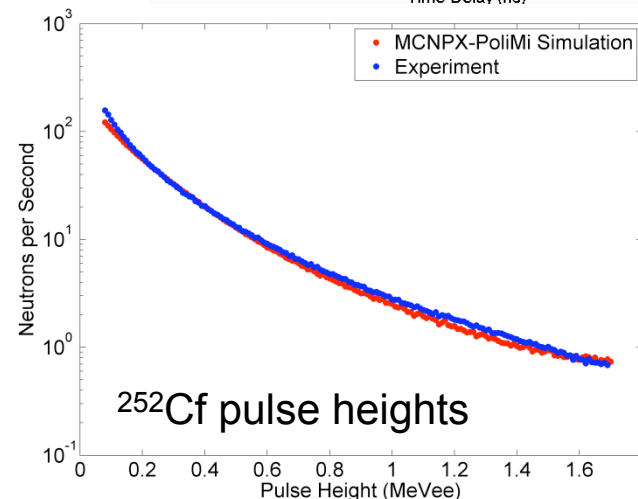
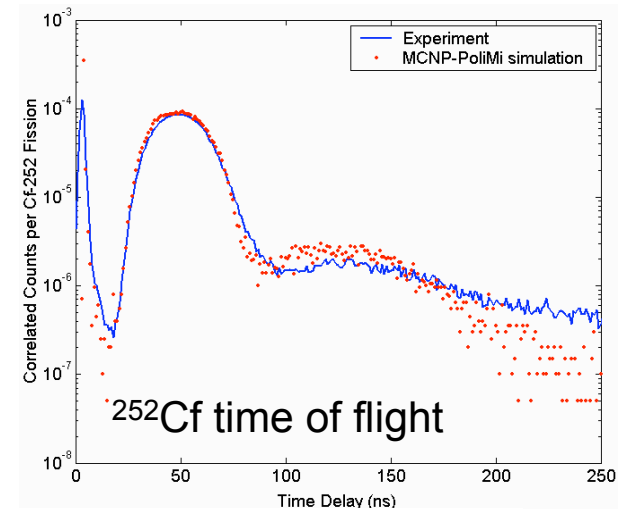




UM-DNNG Capabilities

MCNPX-PoliMi Code System

- The capabilities of MCNP-PoliMi were merged with MCNPX v2.6.0 to create a new code: MCNPX-PoliMi
- The code is suitable for high-fidelity detector response simulations:
 1. Nonlinearity in the light output from neutron collisions
 2. Varying light output from carbon and hydrogen collisions
 3. Pulse generation time within the scintillator
 4. Detector dead time
 5. Detector energy resolution

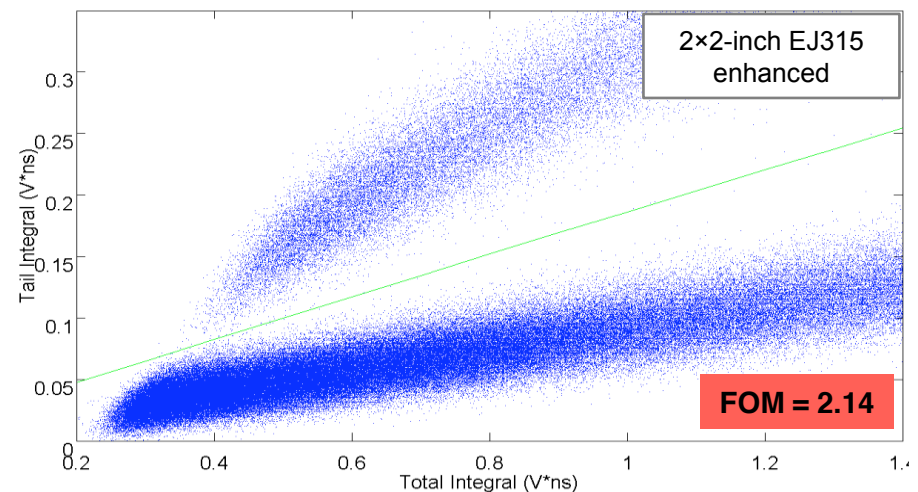
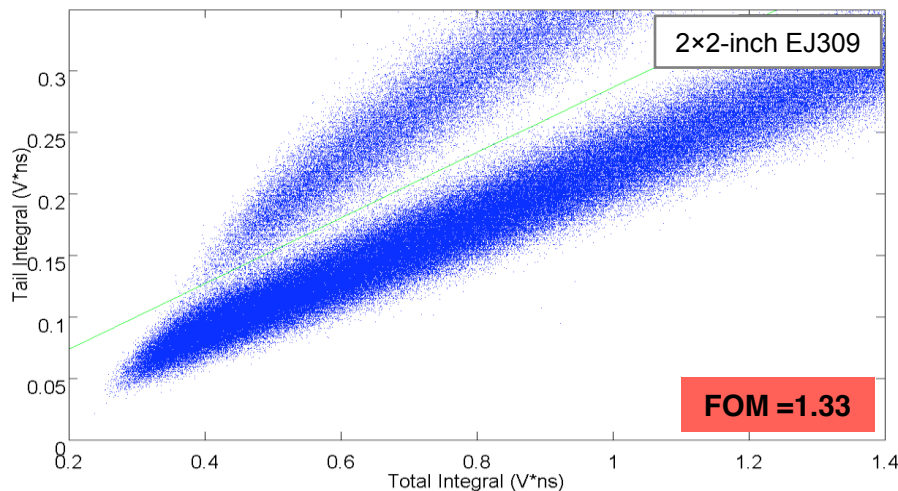
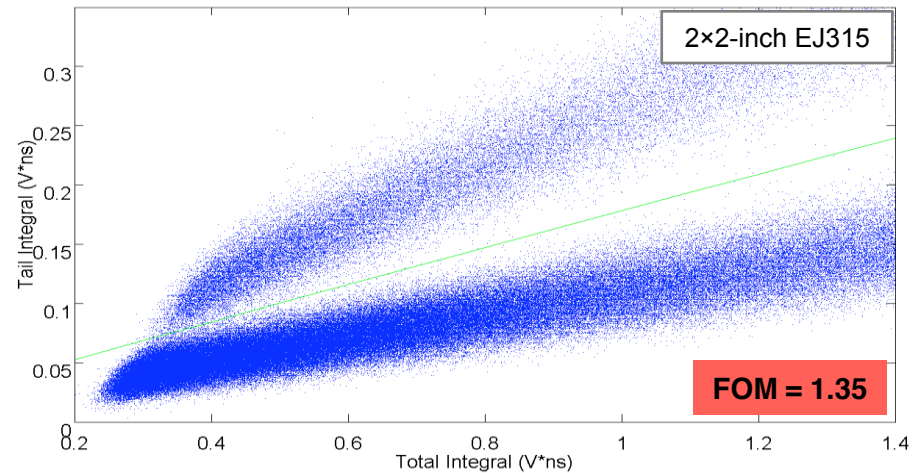


Detector Characterization

Pulse Shape Discrimination: Figure of Merit

Measurement details:

Detection threshold = 70 keVee
 Minimum detectable $E_N \sim 500$ keV
 Length of flight path = 60 cm
 ^{252}Cf fission source strength $\sim 15 \mu\text{Ci}$

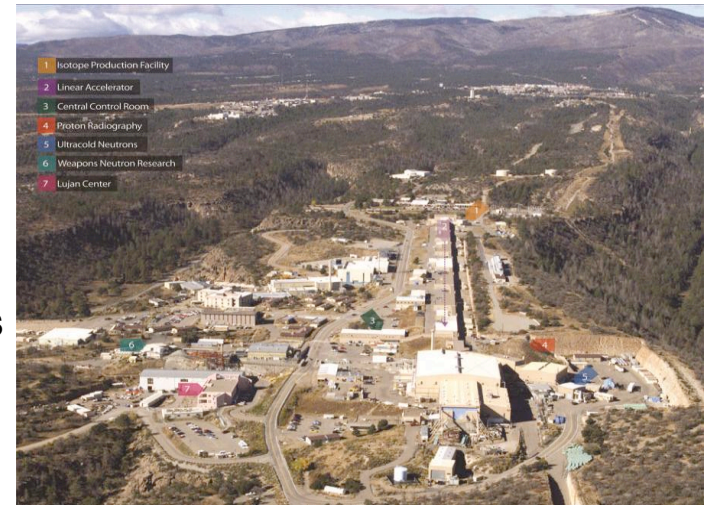


Project Collaborators

LANSCCE, Los Alamos National Laboratory

■ Experimental facility

- Beam time at LANSCCE/WNR neutron source
 - Pulsed “white” neutron source, 0.5 to 600 MeV
- Experimental area
 - 30-degree right beam line
 - 22.7-meter flight path to fission chamber
 - 1-meter flight path, fission to neutron detectors
 - Shielding, infrastructure, safety
- Fission chamber
 - ^{235}U present
 - ^{239}Pu future



- New experimental facility under construction (end CY 2011)

■ Neutron detector-efficiency calibration facility

- Time-tagged neutrons
- Demonstrated capability; available in future for NEUP collaboration

■ Data interpretation, consultation on manuscripts, etc.



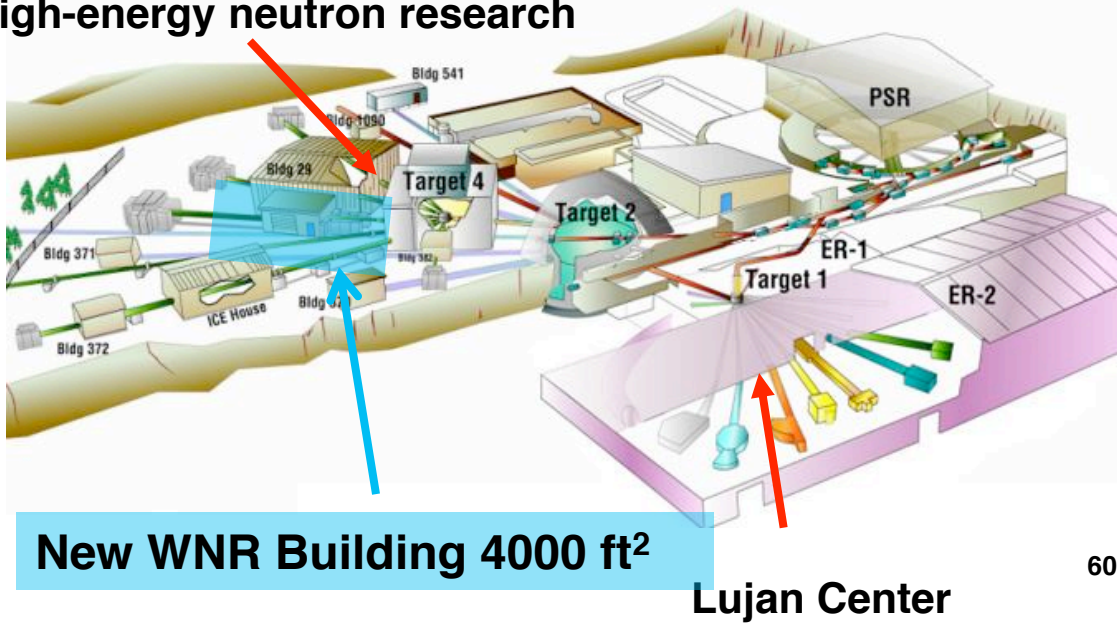
Project Collaborators

LANSCÉ/WNR and New Building

Weapons Neutron Research Facility

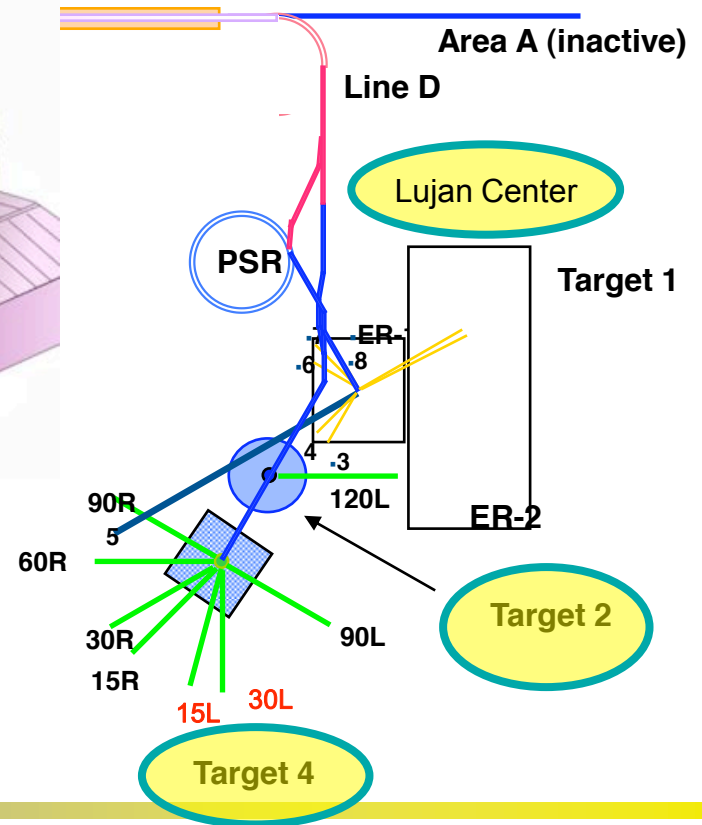
Target-4
High-energy neutron research

Linear Accelerator



New WNR Building 4000 ft²

Lujan Center



Project Collaborators

Brigham Young University

- A cadmium capture-gated detector works well as a neutron spectrometer - essentially all the incident neutron energy is lost in the scintillator prior to capture.
- Problem - Light output of organic scintillators is not linear with proton energy.

Characteristics for a good scintillation material

- High in H for moderating neutrons
- High Z for stopping gammas from Cd capture
- Linear light output for recoil protons

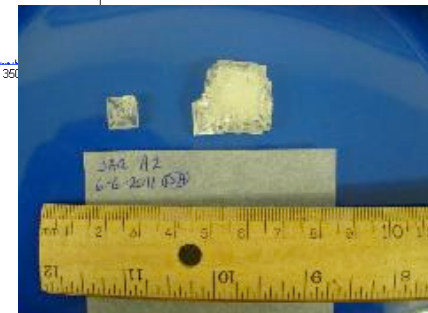
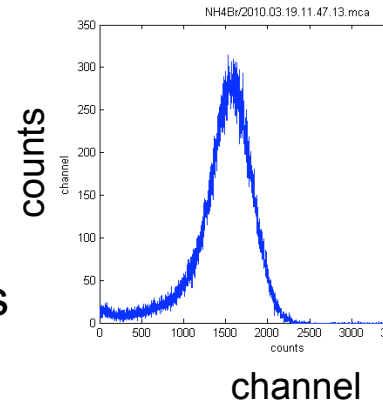
Possible candidates included ammonium halides

- NH_4Br with Eu^{2+} as an activator promising

Initial results look promising (crystals up to about 1cm across)

- Small clear crystals were grown with Eu_2Br included
- Light was produced when radiated by alphas
- Very challenging to scale up - it is hard to make large, good quality NH_4Br crystals
 - they tend to be microcrystalline, and results are not reproducible at this point

NH_4Br (Eu^{2+}) light output

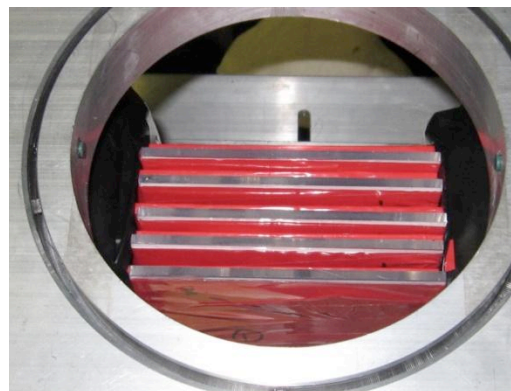
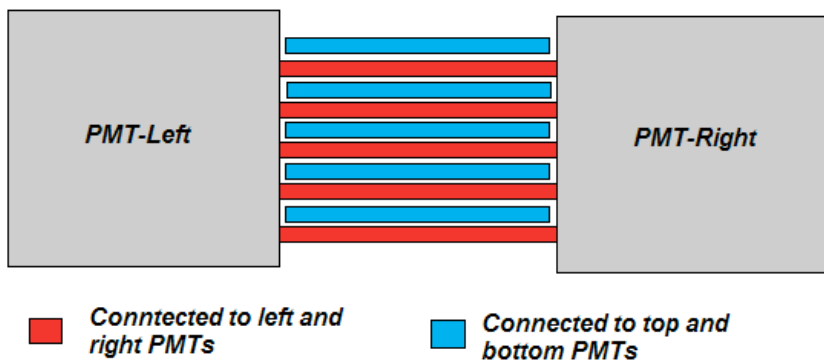




Project Collaborators

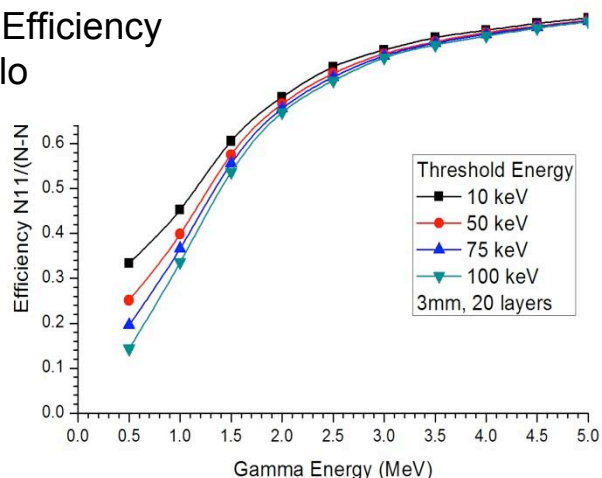
University of Kentucky

Layered Organic Scintillators - Exploit range difference between recoil protons and recoil electrons to distinguish incident neutrons and gammas

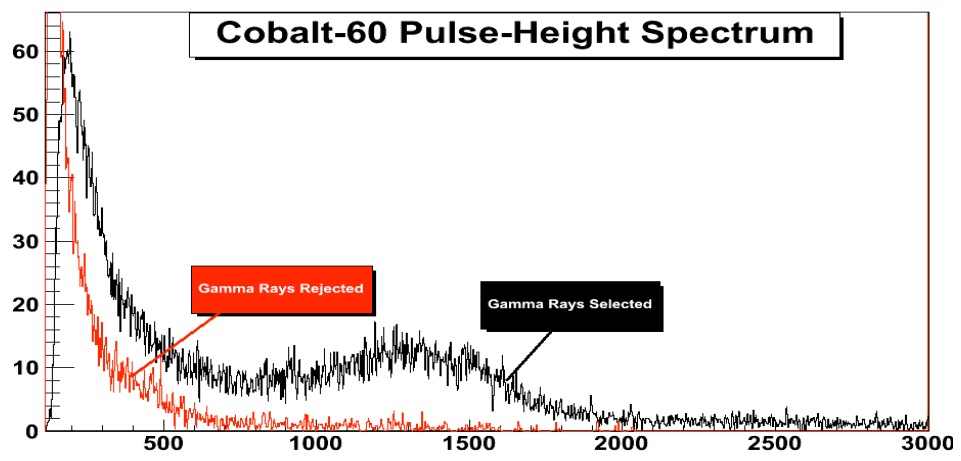


Prototype detector with 5 vertical scintillator layers

Gamma Rejection Efficiency
Geant4 Monte Carlo



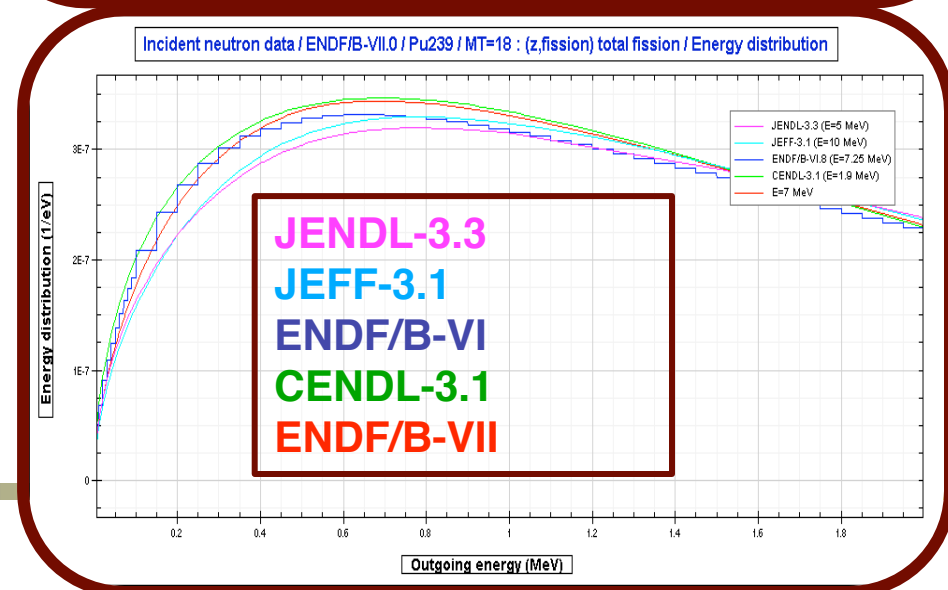
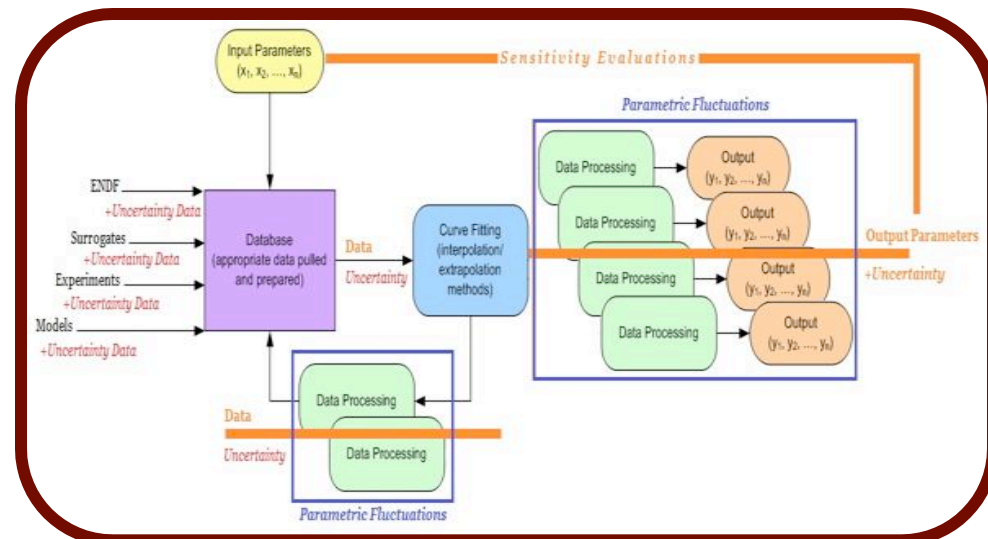
Cobalt-60 Pulse-Height Spectrum



Project Collaborators

Texas A&M University

- Development of high fidelity modeling approaches for system analysis and optimization
- Performance domain evaluations of advanced nuclear energy systems focusing on operation characteristics and waste management strategies vs. nuclear fission spectral data

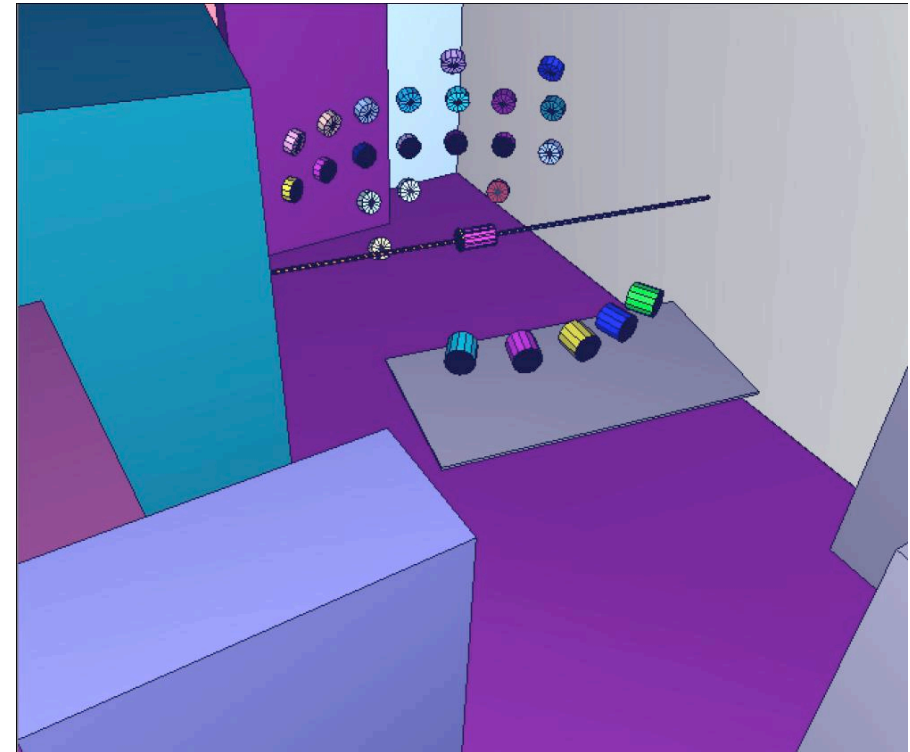




Experimental Setup at LANSCE

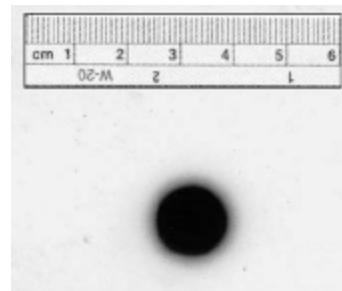
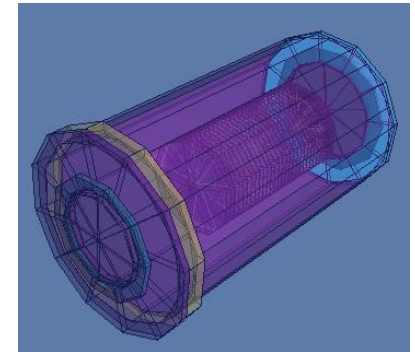
Measurement performed in July 2010

- WNR 30R beam
- Flight path 22.657 m
- White neutron source
 - Neutrons 0.5-600 MeV
- U-235 fission chamber (FC) from LLNL
- CAEN V1720, 12-bit, 250-MHz waveform digitizer
- Five EJ-309 liquid organic scintillation detectors - 80-cm distance between the FC and the detectors
- Threshold set to ~ 50 keVee (~ 350 keV neutron energy dep.)



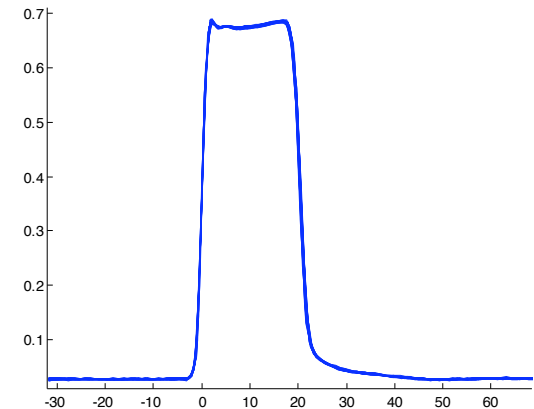
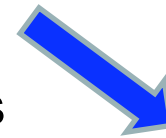
Fission Chamber Description

- Parallel-plate avalanche chamber (PPAC)
- 10 plates coated on each side with U-235
- Total deposited U-235 mass approximately 112 mg
- ~60 induced fissions per second
- ~30 triggers per second from alpha decays
- Target area is ~10 cm²; corresponding beam cross section is ~3 cm²
- Logical pulses have less than 0.2-ns spread – excellent timing



Experimental Data Acquired

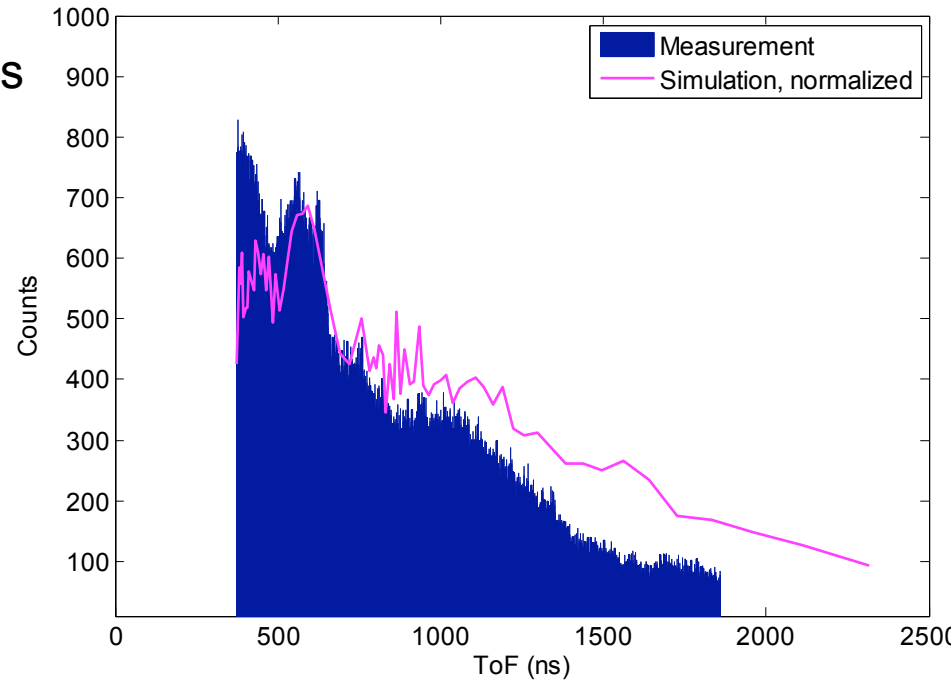
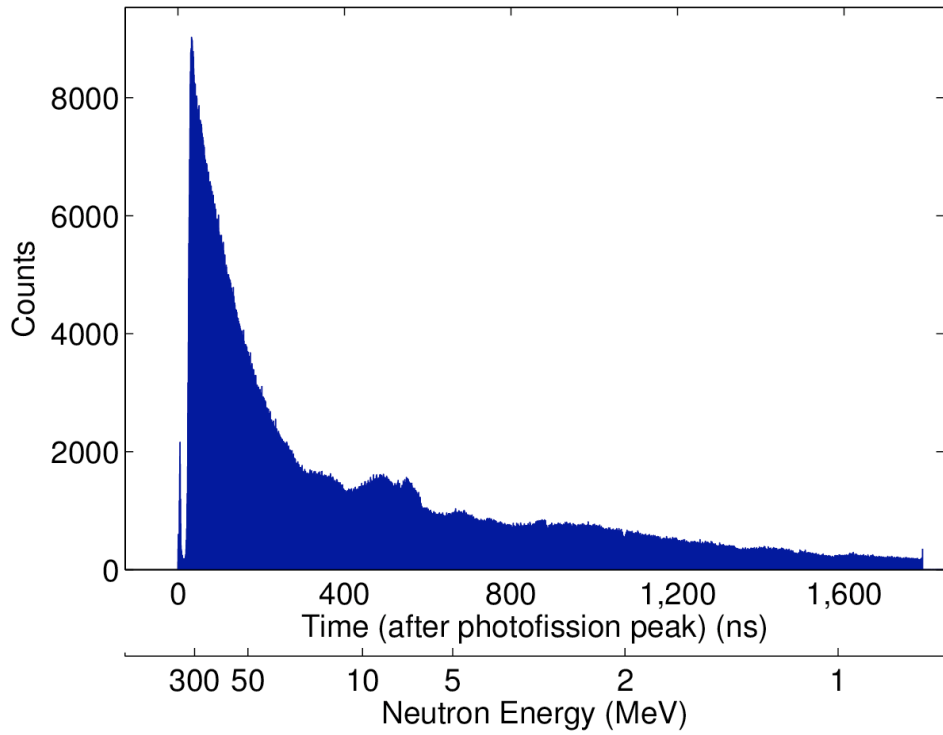
- Double time-of-flight (TOF) measurement
- Seven active channels used:
 - One beam trigger (long signal delay due to logical pulse creation and signal transport over 20 m)
 - One FC (logical square pulse shaping introduces delay, but very accurate timing characteristics)
 - Five channels for EJ-309 detectors
- Two neutron energies:
 - Fission-inducing neutron energy
 - Fission emitted neutrons from FC
- ~60 h of data were acquired
- ~1 neutron per second detected





Beam–Fission-Chamber TOF

Timing aligned with position of photo-fission peak (for 22.7-m flight path it occurs at 71 ns after spallation). 1–2-ns timing resolution observed.

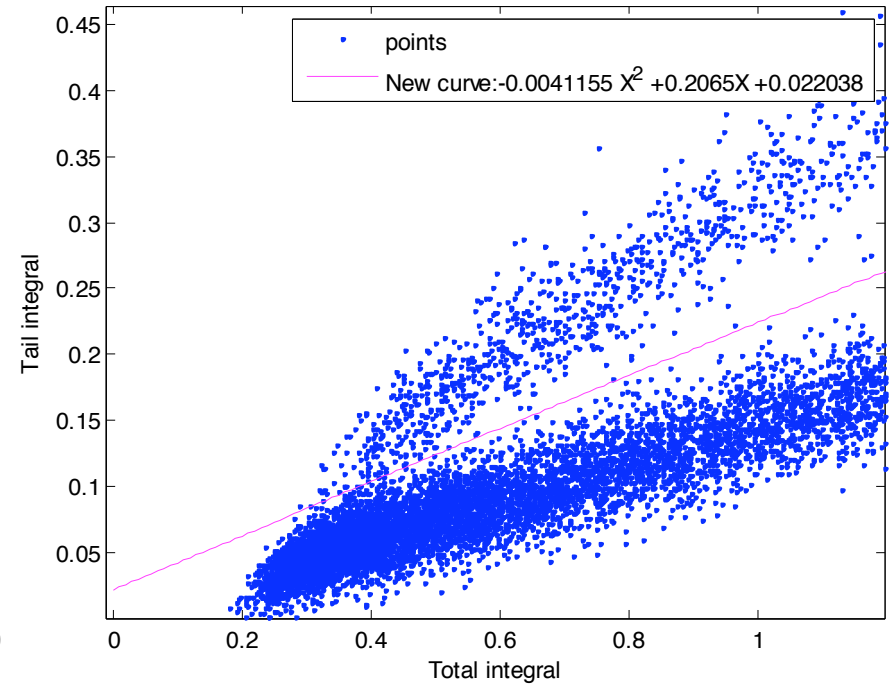
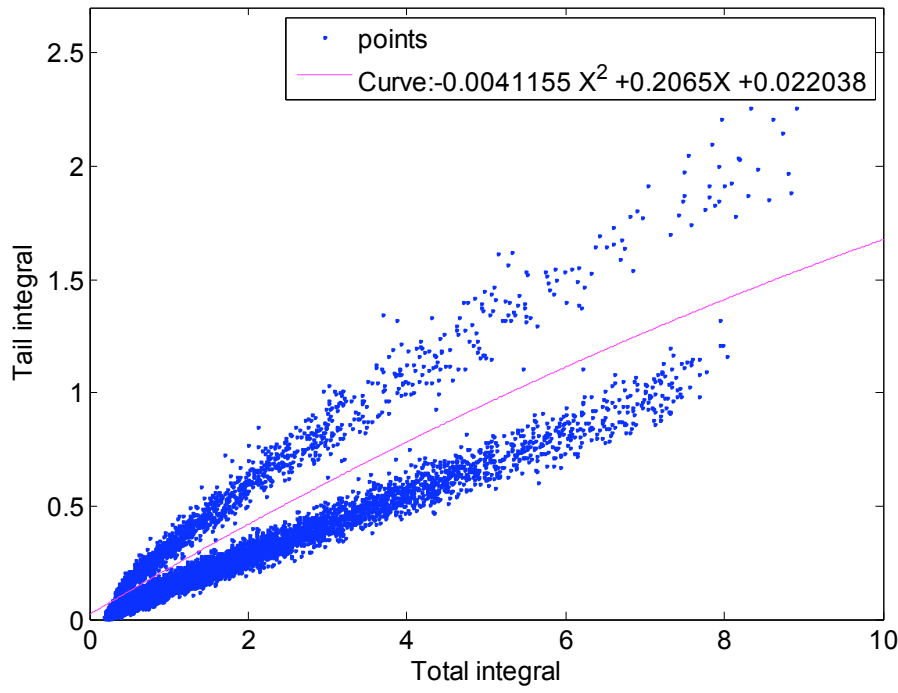


Preliminary comparison with simulated results, data for beam neutrons up to 20 MeV in energy.



Pulse Shape Discrimination

Benefit of liquid organic scintillators



Pulse shape discrimination for neutrons and gamma rays, tail integral vs. total integral of digitized pulse.

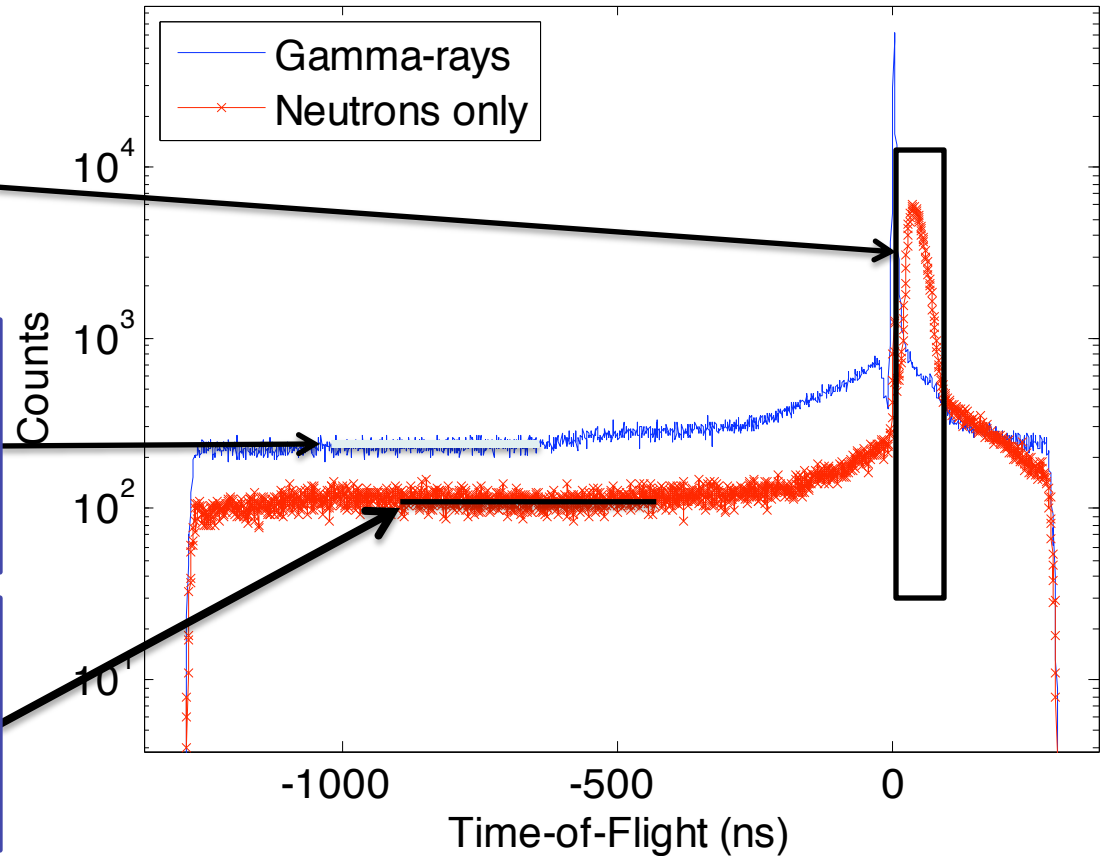


TOF Liquid-Scintillator Data

Neutron TOF region, slice for time/energy groups.

Gamma background (using the gamma-misclassification rate it will also make a correction factor for neutrons).

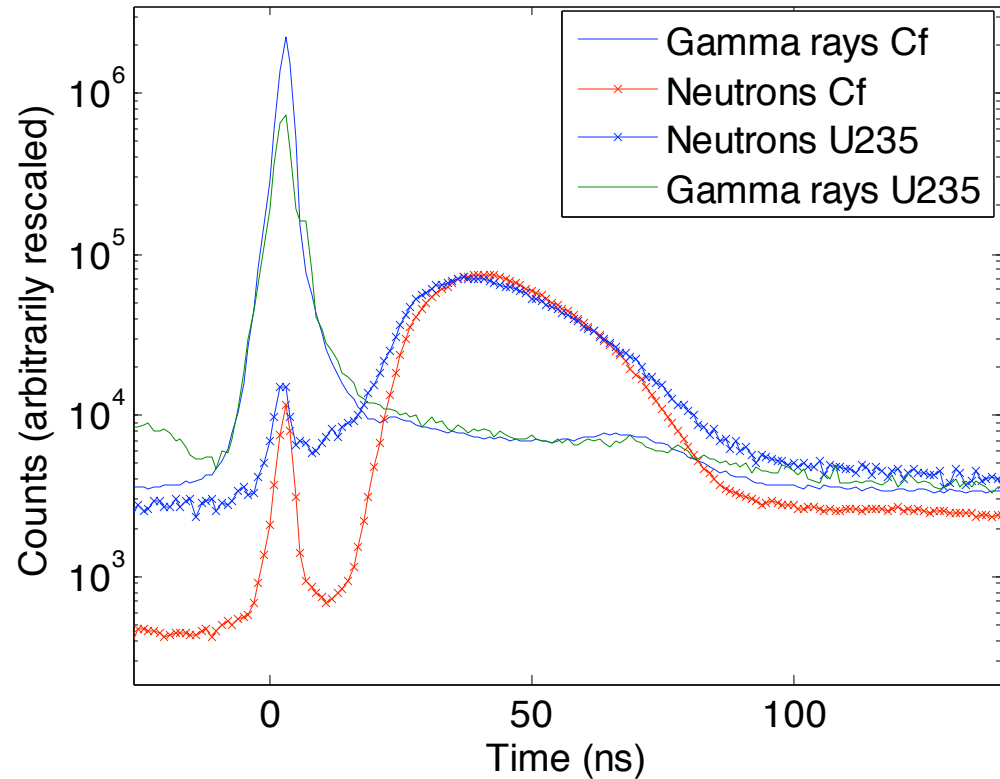
Neutron background to be subtracted from ToF-region.





TOF Liquid-Scintillator Data (Cont'd)

- LANSCE data (U-235) and Cf-252 efficiency TOF data fitted to approximately the same neutron amplitude for direct comparison.
- The similarities and differences of the different TOF spectra can be seen.
- High neutron background due to scatters from the neutron beam.



Data Analysis

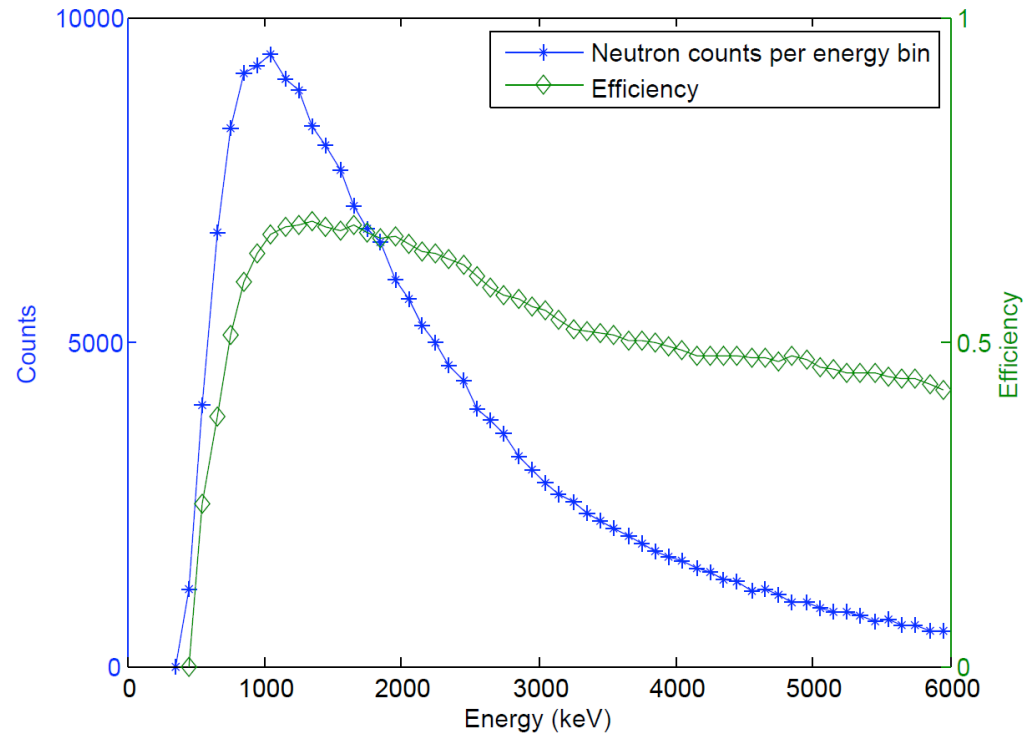
- To find the emitted neutron spectrum from the U-235 induced fissions we need to find the incoming flux on the detectors.
- Detector counts divided by the energy-dependent neutron detection efficiency.
- An identical setup of the same five EJ-309 detectors was created in the DNNG-lab.
- A Cf-252 source was used which has a fairly well-known spectrum; a sixth EJ-309 detector was used for fission timing.





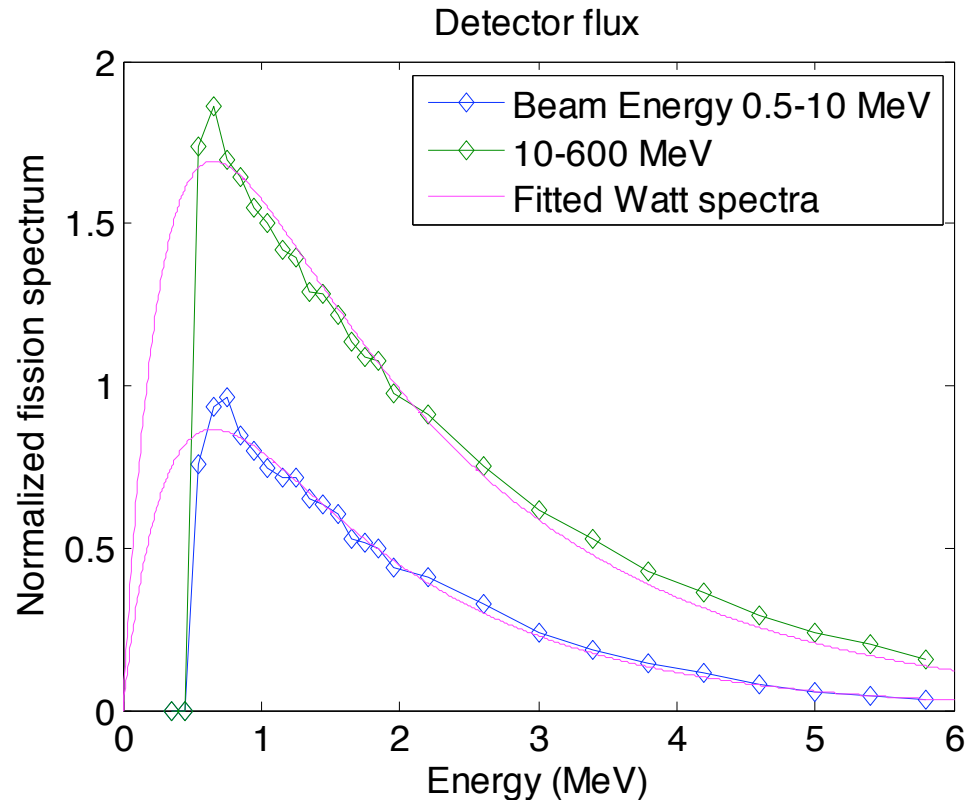
Neutron Efficiency and Detected Counts

- The counts are shown for all inducing neutron energies (0.5-600 MeV).
- At low energies data are limited by the detector efficiency going to zero near the threshold.
- At high energies detector counts and clipped pulses reduce the data likewise.



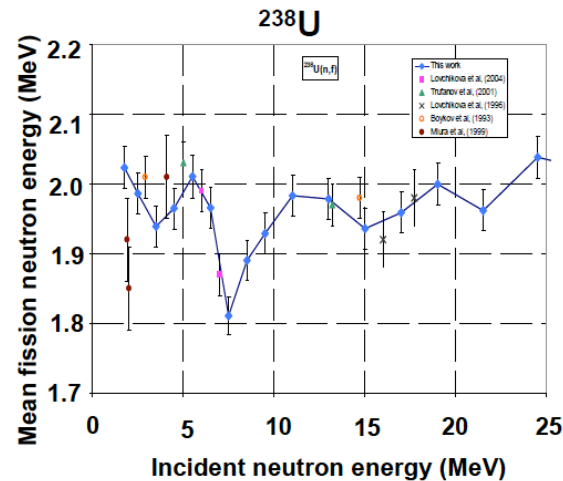
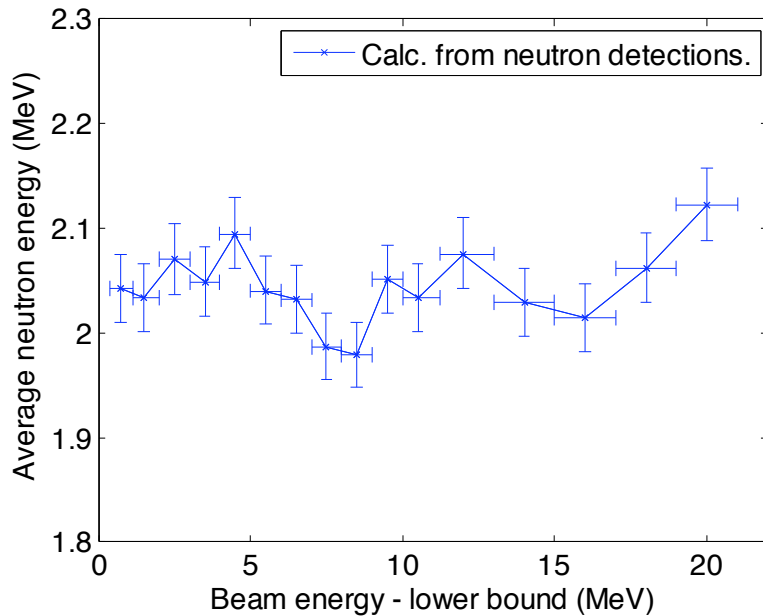
Unfolded Fission Spectrum

- Analyzing data as a function of the incoming beam energy allows for measurement of the number of emitted fission neutrons ($\bar{\nu}$) and their energy distribution.
- Normalization by detector geometrical efficiency and fission counts giving the absolute fluxes.
- At high beam-neutron energies more high-energy neutrons are emitted.





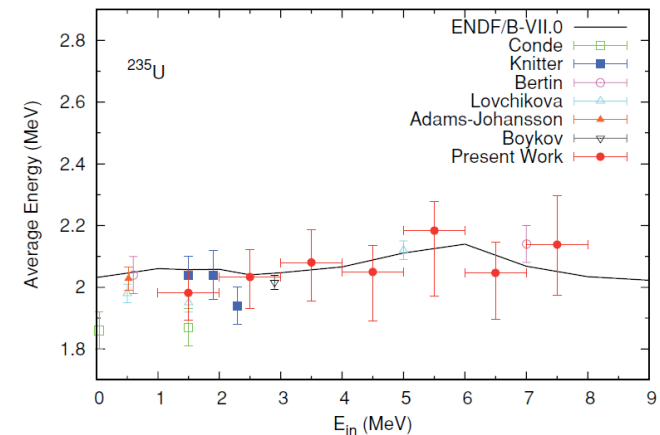
Average Energy of Detected Fission Neutrons (approx. 0.350 MeV – 6 MeV)



U-238

U-235

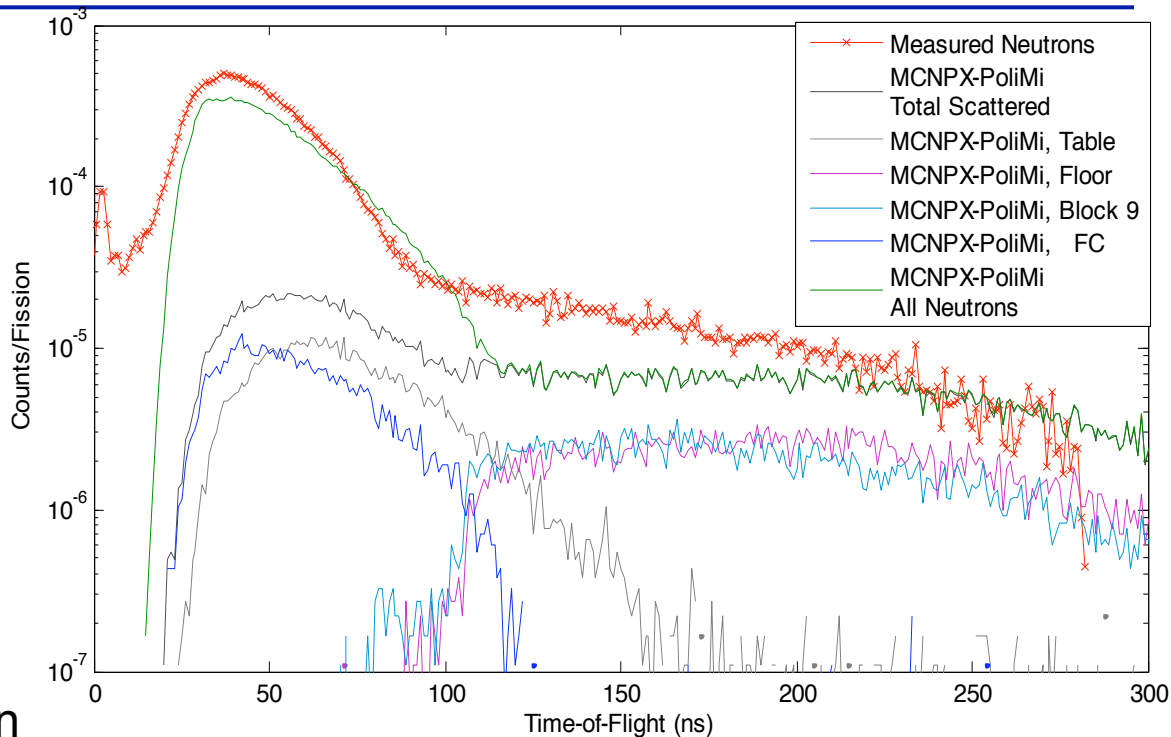
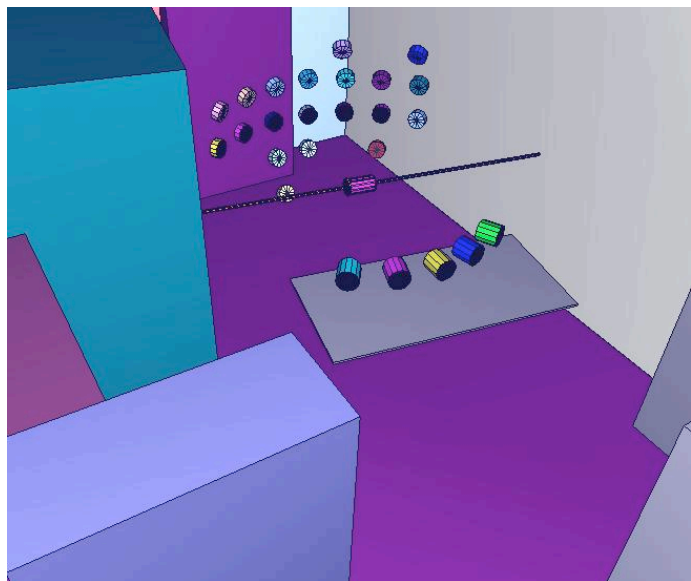
- Project results above (left); LANSCE data on right; energy calculated from TOF.
- (n, n'f) and (n, 2n'f) thresholds at 7.5 MeV and about 15 MeV are observed.





Room-Return Simulations

MCNPX-PoliMi



• Plot normalization is per fission

Table	Floor	Block 9	Other		
			Door	Rest of Room	FIGARO
43.87%	24.44%	21.87%	1.67%	7.13%	1.02%

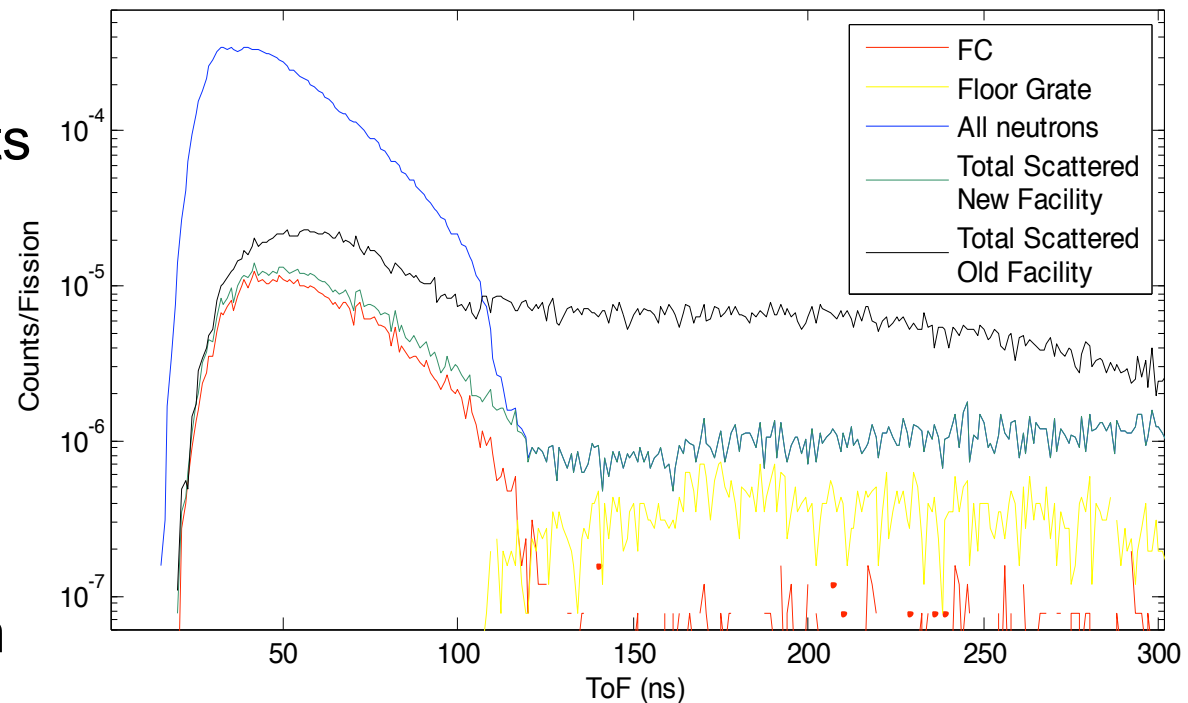
% of total detected scatters

- Measured data in red
- Other data from MCNP simulation
- In-scatter from fission chamber housing almost identical to “out-scatter” so that contribution can be neglected



New LANSCE Facility

- New facility features a 6-foot deep pit below the fission chamber.
- This modification results in a significantly lowered amount of neutron scattering in the room.
- In the lower energy regions (~ 300 keV), a reduction of almost a factor of ten can be seen between the old facility and the new facility.



Next Measurement Campaign

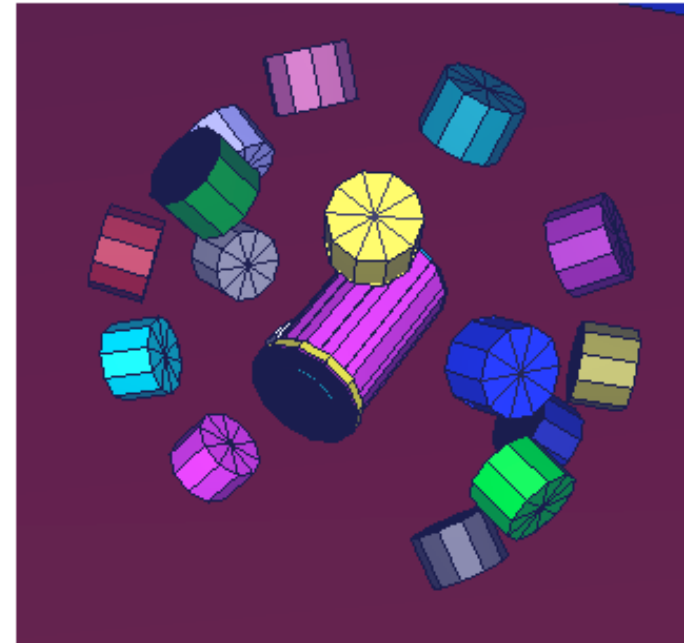
Within the next 6 months

- New LANSCE facility design:
 - New building with reduced room return will improve data acquisition.
- New additional detectors:
 - The DNNG group now possesses approximately 40 organic scintillators of various sizes to improve detection efficiencies.
- Additional improvement in analysis:
 - Using compound PSD methods lower energy neutrons can now be detected.
 - Measuring in the middle of an accelerator cycle will improve facility stability and the amount of recoverable data.



New NE-UP grant (11-1948)

- A new NE-UP grant was awarded (UM-LANL):
 - Basic Physics Data (FC4: Improved measurement techniques): Measurement of Neutron Multiplicity from Induced Fission
- Outcomes:
 - Neutron detection procedures for multiplicity distributions
 - Basic physics data on induced fission neutron multiplicity distributions for actinides of interest to the FCR&D
 - Energy-angle correlations of neutrons emitted from neutron-induced fission events





Detection for Nuclear Nonproliferation Group



NE-UP: Improved Fission Neutron Data Base for Active Interrogation of Actinides

S.A. Pozzi, A. Enqvist, B. Wieger, and M. Flaska

*Department of Nuclear Engineering and Radiological Sciences,
University of Michigan, Ann Arbor, Michigan*

R.C. Haight

LANSCCE, Los Alamos National Laboratory, Los Alamos, New Mexico

J.B. Czirr, L.B. Rees

Department of Physics, Brigham Young University, Provo, Utah

M.A. Kovash

Department of Physics and Astronomy, University of Kentucky, Kentucky

P.V. Tsvetkov and M. Cuvelier

*Department of Nuclear Engineering, Texas A&M
University, College Station, Texas*

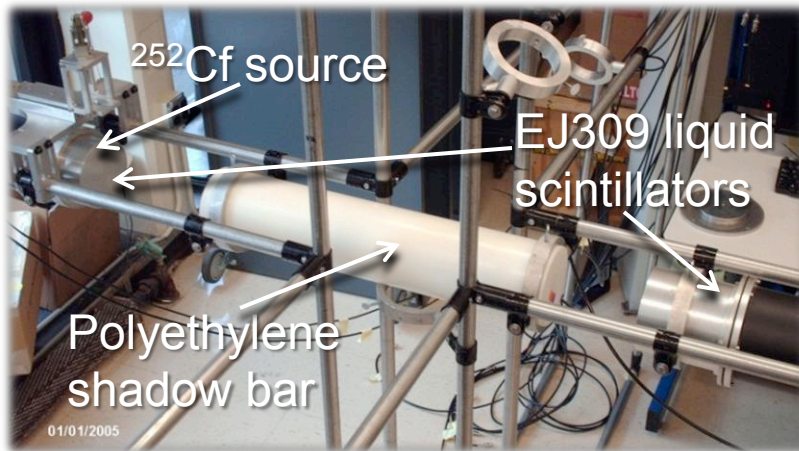
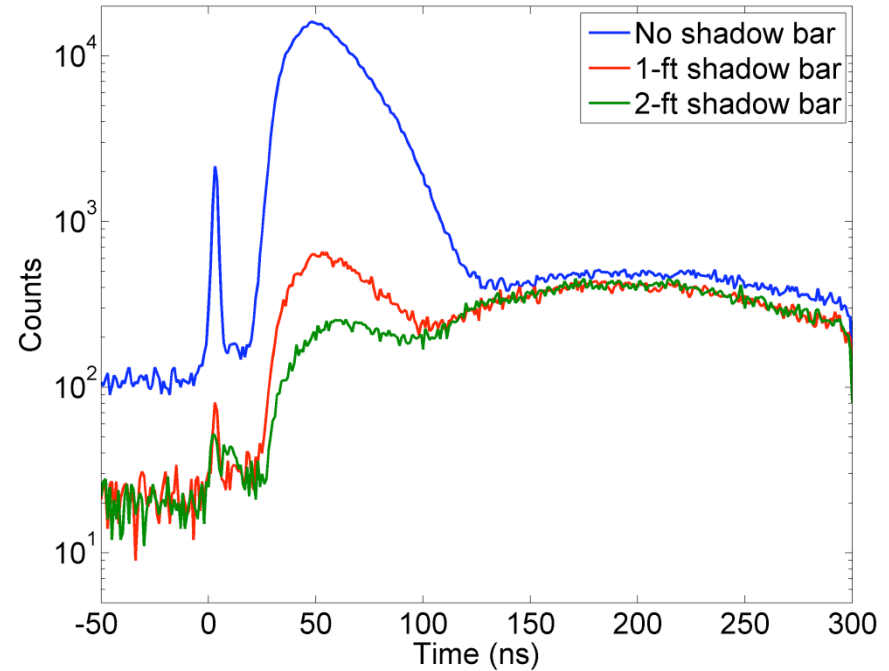


Detection for Nuclear
Nonproliferation Group

Detector Characterization

Efficiency Measurements

- Measurement technique
 - Measurements were taken with a polyethylene shadow-bar to determine room contribution
 - The room contribution is subtracted from bare time-of-flight measurement to obtain response matrix (or intrinsic neutron detection efficiency)



$$\begin{pmatrix} R_{11} & \dots & R_{1n} \\ \vdots & & \vdots \\ \vdots & & \vdots \\ R_{m1} & \dots & R_{mn} \end{pmatrix} = \begin{pmatrix} R_{11} & \dots & R_{1n} \\ \vdots & & \vdots \\ \vdots & & \vdots \\ R_{m1} & \dots & R_{mn} \end{pmatrix} - \begin{pmatrix} R_{11} & \dots & R_{1n} \\ \vdots & & \vdots \\ \vdots & & \vdots \\ R_{m1} & \dots & R_{mn} \end{pmatrix}$$

Neutron response matrix “Bare” TOF measurement Shadow bar measurement

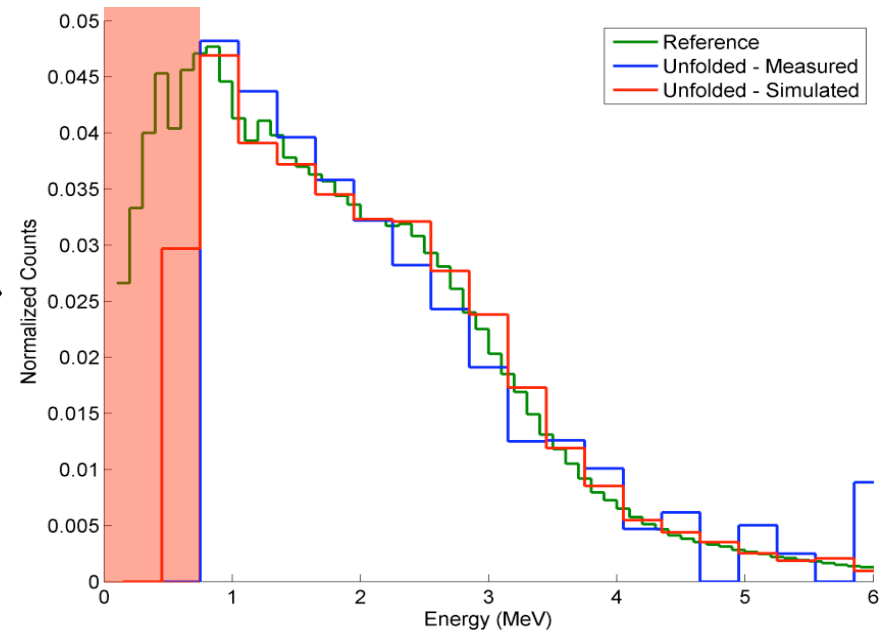
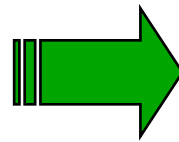
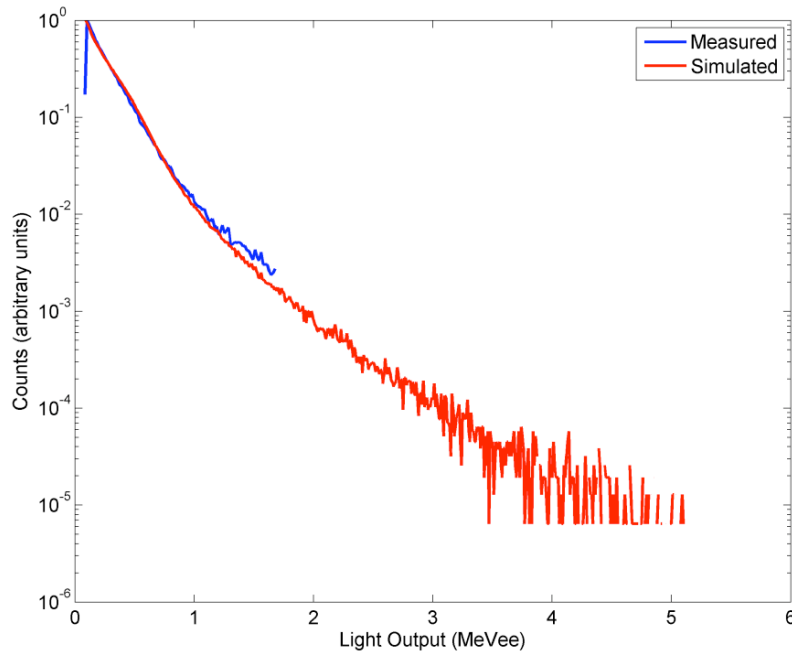


Algorithm Development

Neutron Spectrum Unfolding

- Neutron pulse height distributions are related to the neutron energy spectra
- Advanced algorithms are needed to “unfold” the energy spectra

$$N(L) = \int R(E_n, L)\Phi(E_n)dE_n$$





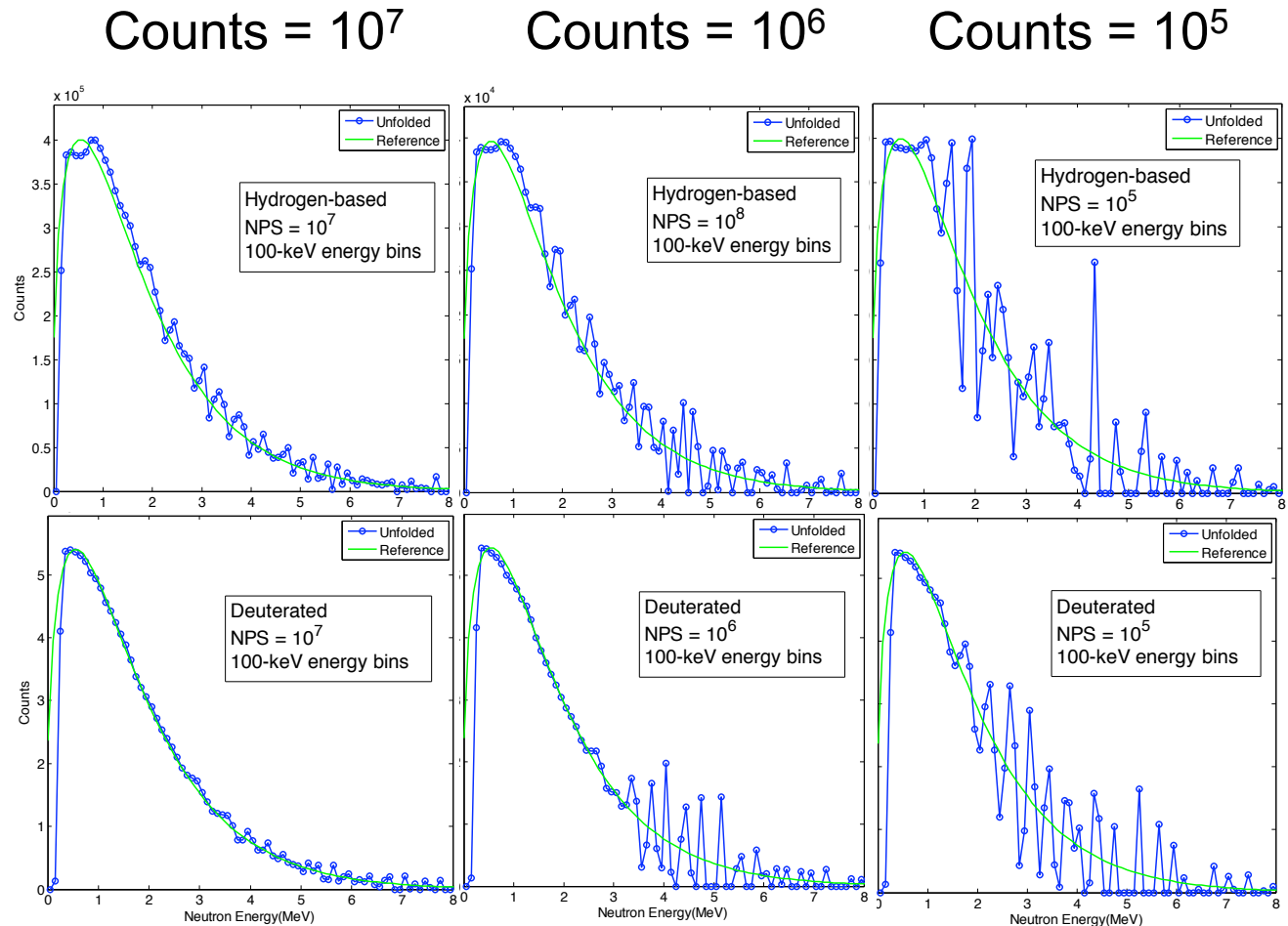
Spectrum Unfolding

Results: EJ309 (^1H) versus EJ315 (^2H)

100-keV bins

EJ309
Hydrogen-
Based

EJ315
Deuterium-
Based

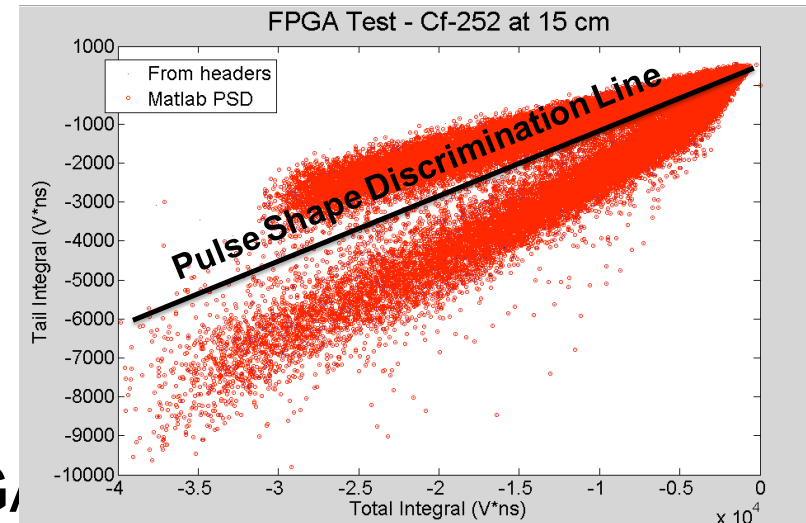




Electronics Development

Digitizing and Processing Board

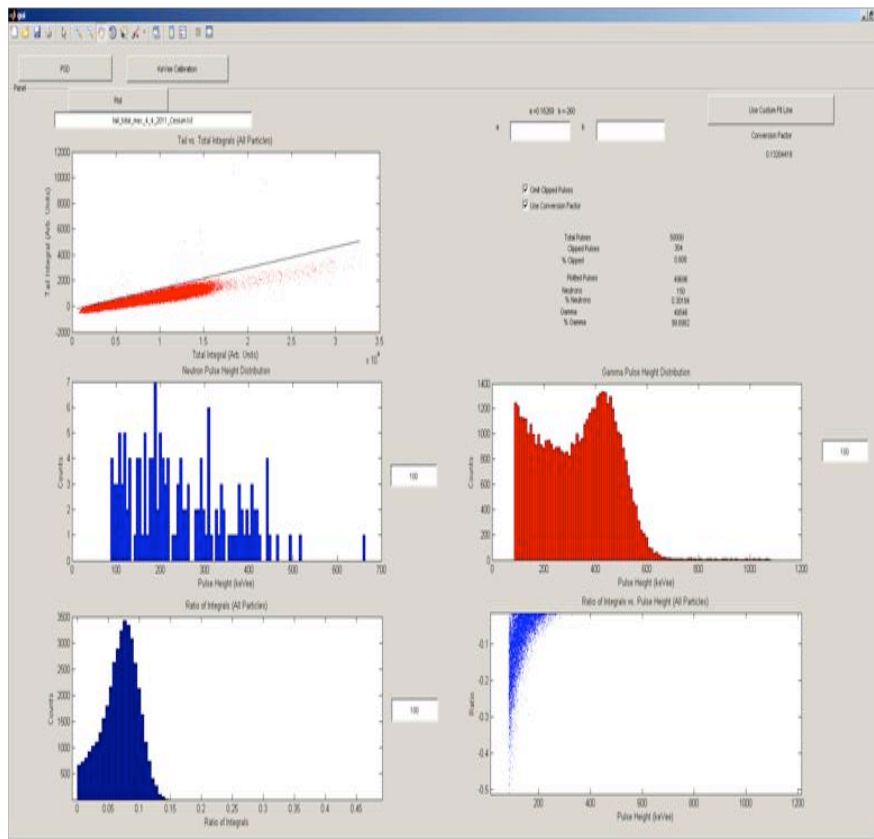
- Commercial board
 - 250 MS/s, 14 bits, 4 channels
 - Connects directly to a PC or laptop
- 4-channel amplitude threshold triggering and time-stamping
- Real-time preprocessing on FPGA
 - Time-of-peak and amplitude detection
 - Tail and total integration
 - Comparison to a programmable PSD line
- Customized software on a PC to visualize the tail/total integral results



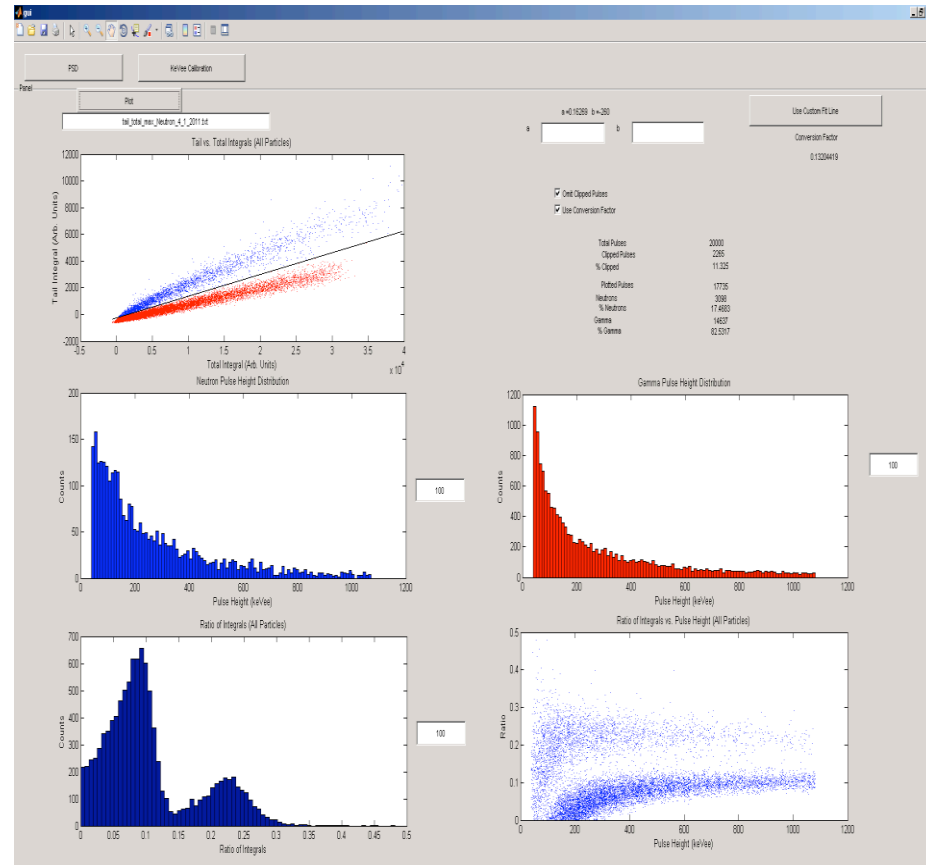
Electronics Development

Real-Time Graphical User Interface

^{137}Cs Gamma-Ray Source



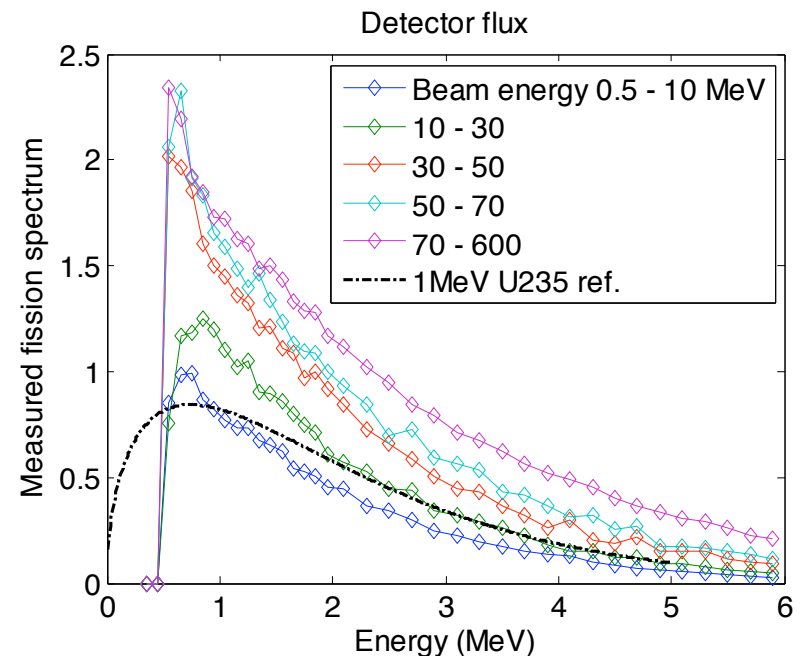
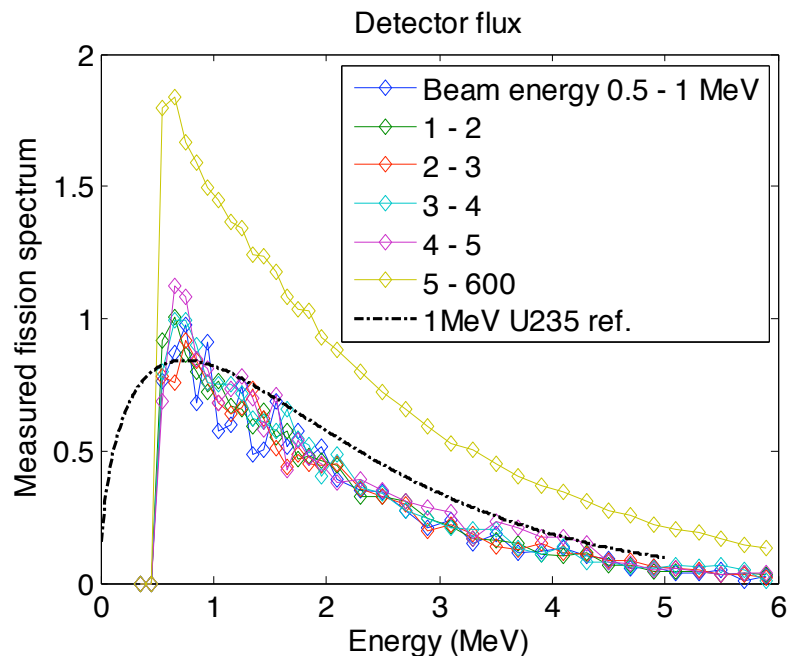
^{252}Cf Spontaneous Fission Source





Normalized Fluxes

- Normalization by detector geometrical ε and fission counts gives the absolute fluxes.
- Decent agreement with the plotted Watt-spectrum for 1-MeV n-induced fission (taken from MCNP) for low beam energies.
- At high energies the increased $\bar{\nu}$ becomes very pronounced.



Ongoing work

- Investigating the dependence of $\bar{\nu}$ (avg. neutrons) with neutron beam energy.
- Fitting the obtained fission spectra with Watt-curves and compare to literature.
- MCNP simulations to acquire the beam-FC spectra, and to verify the fission cross sections at high energies (>20 MeV).
- Prepare improvements and changes for the next measurement campaign to take place in the next 6 months at LANSCE.
- Data analysis improvements:
 - PSD algorithms
 - Pulse timing
 - Error propagation

Extra

- Pu239 data sample...

