Quasi-Differential Neutron Scattering Measurements of ^{181}Ta and Teflon from 1.5 to 20 MeV

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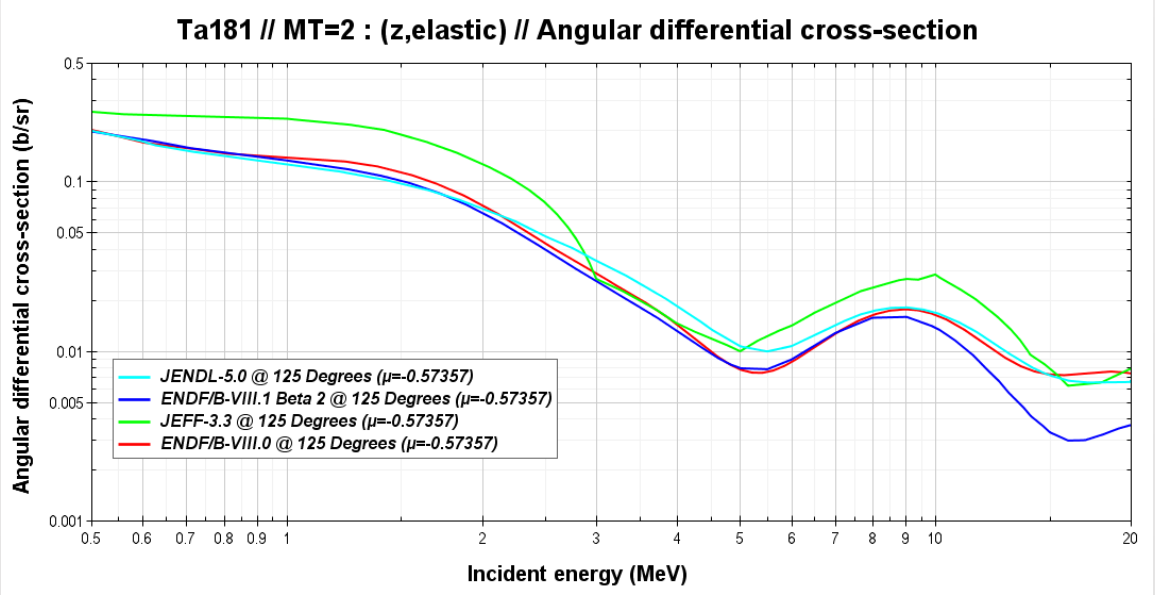
NCSP NUCLEAR CRITICALITY SAFETY PROGRAM

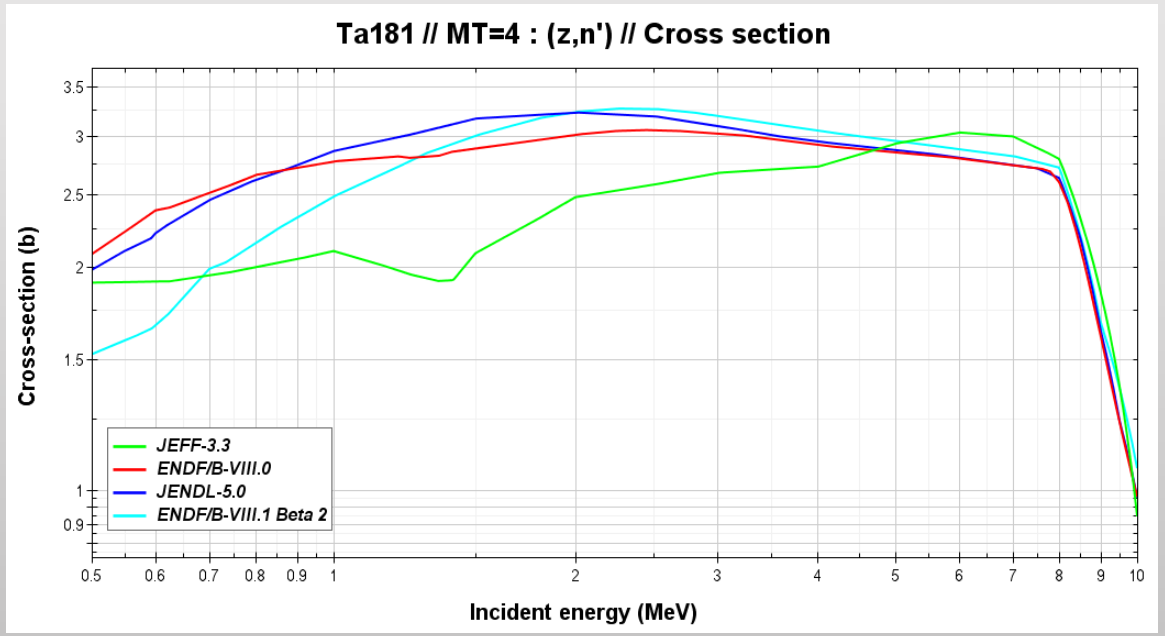
Presentation Overview

1. Motivation for the Experiments Performed
2. Upgrades to the Experimental Apparatus for Measurements
3. Quasi-Differential Experimental Methodology
    a) Pulse Shape Discrimination
    b) Validation Measurement of Carbon
4. Preliminary Results
5. Future Studies

Motivation for RPI Tantalum Experiment

- Due to special material and nuclear properties ^{181}Ta is relied on for the following applications:
  1. Neutron-producing targets for linear accelerators
  2. Recovering Uranium from fuel reprocessing[1]
  3. Casting of molten Plutonium metal[1]
- ^{181}Ta neutronics must be thoroughly understood to enhance the realms of scientific research, criticality safety, and national security

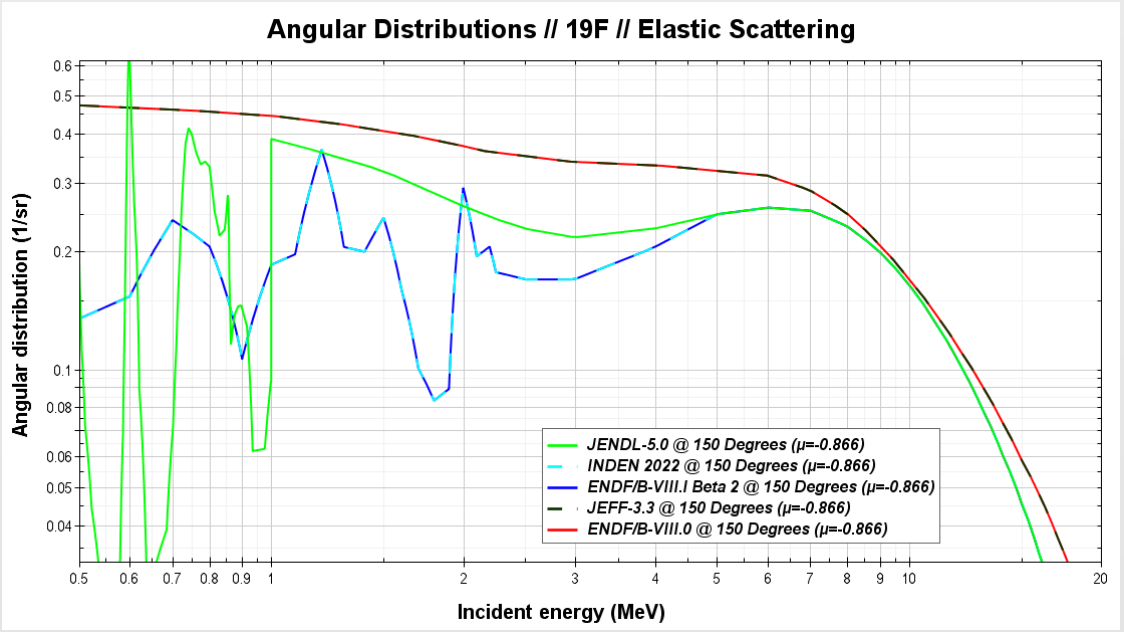


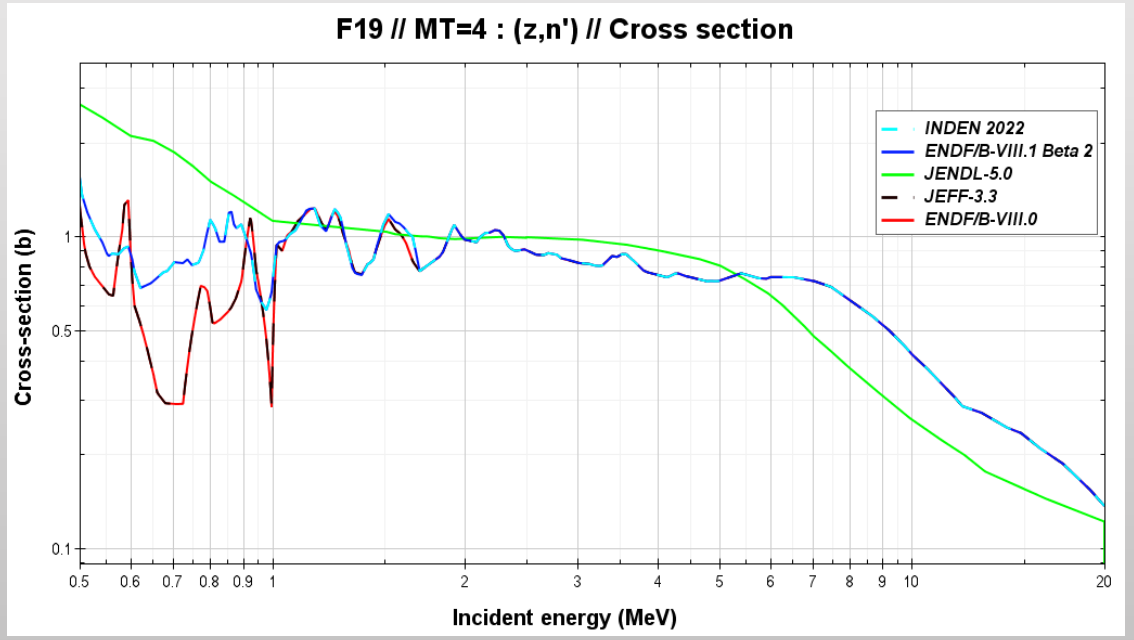


[1]Chambers, A. (2023) - *Five Year Execution Plan – United States Department of Energy Nuclear Criticality Safety Program FY2024 though FY 2028*

Motivation for RPI Teflon (CF_2) Experiment

- Measuring Teflon allows for validation of the ^{19}F evaluations
- Fluorine is relied on heavily in salts for Generation – IV reactor concepts:
  - Kairos Power – FHR
  - TerraPower – SFR and Natrium
  - Flibe LFTR (blanket and coolant)
- Fluorine is an integral component of Uranium manufacturing/enrichment (UF_6)
- Criticality safety, design, and operation of these reactor concepts are highly dependent on ^{19}F neutronics





The RPI High Energy Neutron Scattering System

Close-to-ideal neutron scattering array located ~30m from the neutron-producing target constructed with:

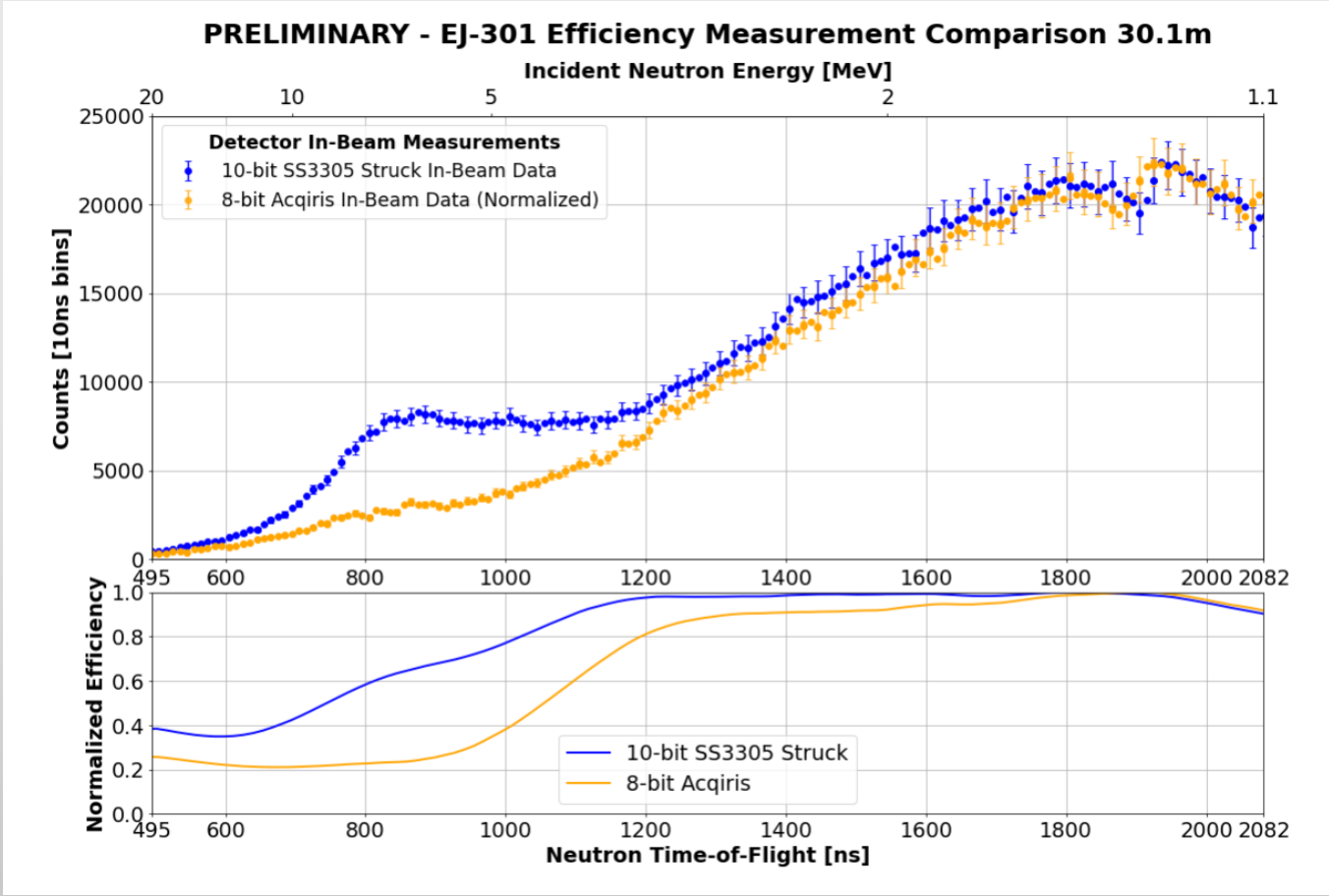
I. Eight 5in diameter EJ-301 organic liquid scintillator proton recoil detectors
   - Coupled to Photonis XP4572/B PMTs

II. Aluminum alloy detector mounting hardware and sample holder

III. A rigidly secured optical table allowing for seemingly infinite detector arrangements

Upgrade to Struck SIS-3305 10-bit Digitizer

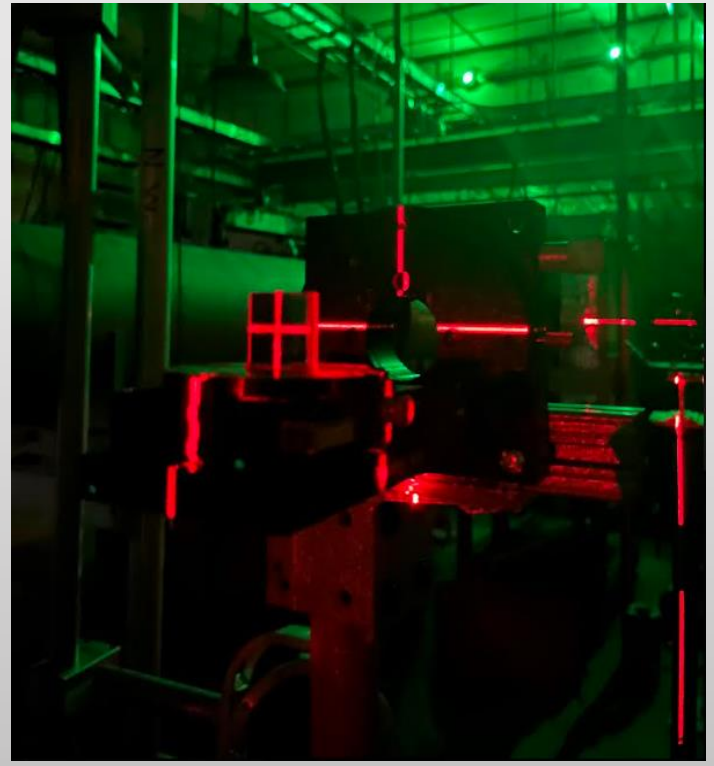
- System upgraded from Agilent-Acqiris AP240 8-bit to Struck SIS-3305 10-bit digitizer
  - Dynamic range of pulses increased from 256 bits to 1024 bits
  - Sampling resolution increased from 1.0*ns* to 0.8*ns*
- Upgrade yields increase in relative neutron detection efficiency of the system, largest gains in efficiency observed from 2 MeV – 20 MeV
- Comparison generated using pulse shape discrimination methods[1] different from the results presented in this work

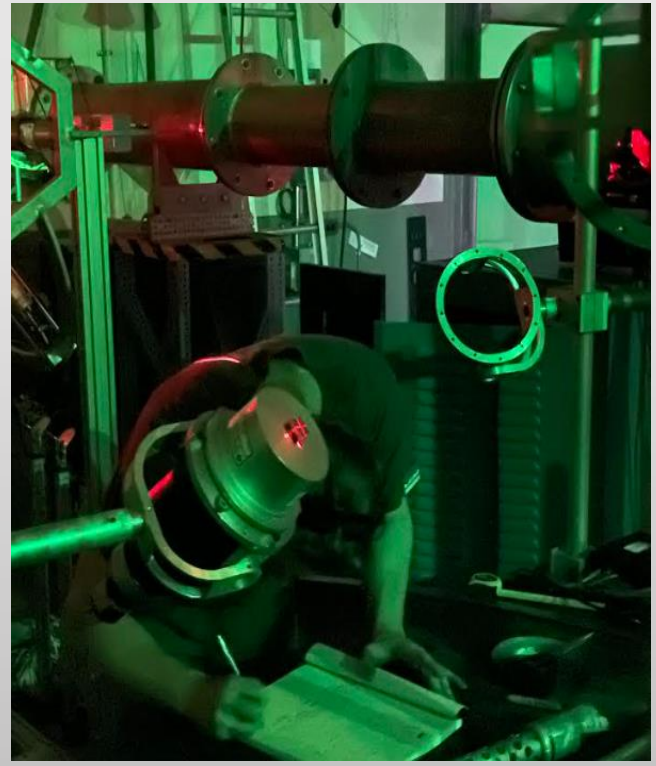


[1]A. M. Daskalakis, E. J. Blain, B. J. McDermott, R. M. Bahran, Y. Danon, D. P. Barry, R. C. Block, M. J. Rapp, B. E. Epping and G. Leinweber, "Quasi-differential elastic and inelastic neutron scattering from iron in the MeV energy range", *Annals of Nuclear Energy*, vol. 110, pp. 603 - 612, 2017

EJ-301 Detector Alignment Upgrade

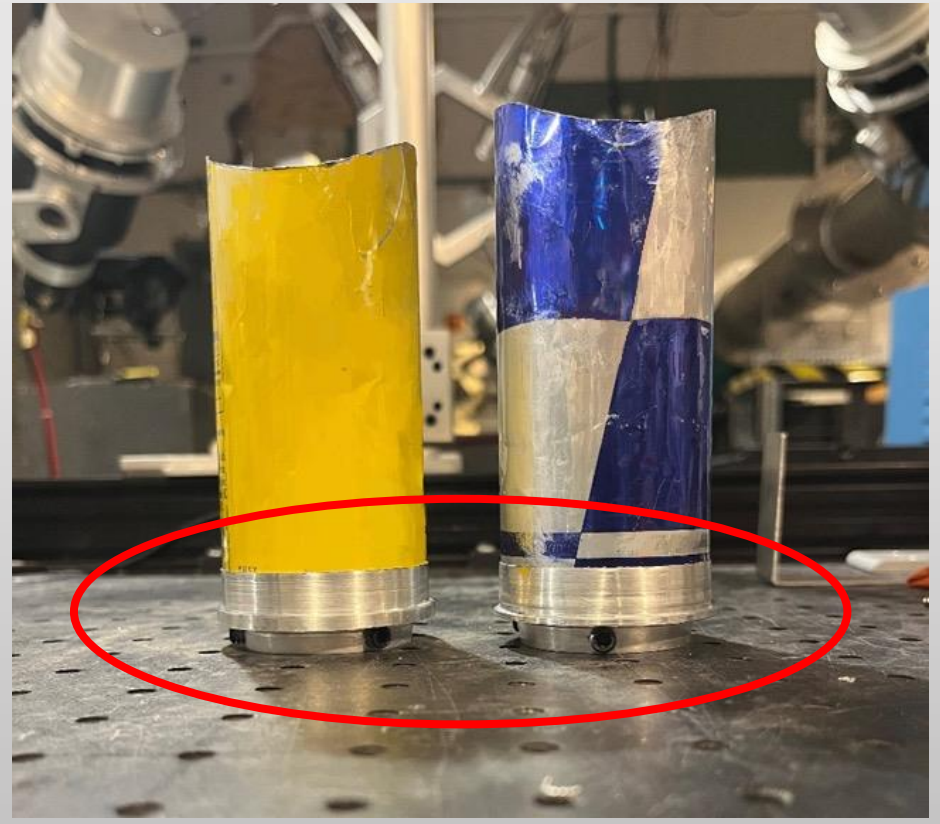
Alignment procedure using a beamline-centered self-leveling laser, mirror, and optical coincidence implemented to more accurately position detectors for experiments.





Sample Holder Upgrade

New fabricated aluminum ring/sleeve and securing mechanism are more rigid than previous solution – reducing sample position uncertainty



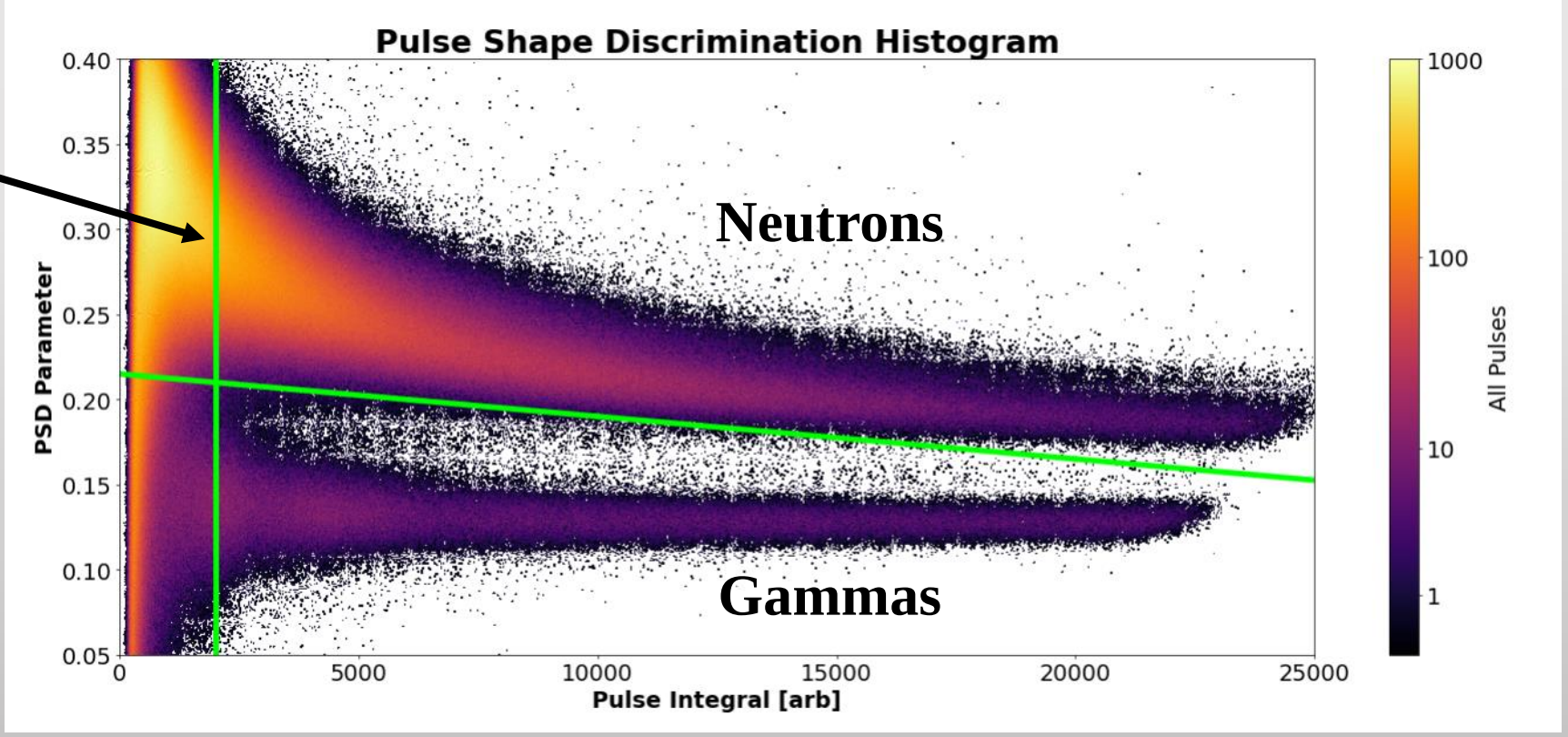
Quasi-Differential Scattering Experimental Conditions and Data Analysis

Quasi-Differential Measurement Methodology

1. Conduct differential neutron time-of-flight experiment on sample of interest, validation sample, and open beam
   - Due to sample size, the experiment is dominated by multiple scattering interactions
2. Perform MCNP transport calculation of validation (Carbon) measurement using measured neutron flux and detector efficiencies
   - This validates experimental geometry and reproduction of known validations sample
3. Perform MCNP transport calculation of sample of interest measurement using measured neutron flux and detector efficiencies
   - Differences present in nuclear data evaluations of the sample of interest are compared to the experimental data to validate performance or show needs for improvement

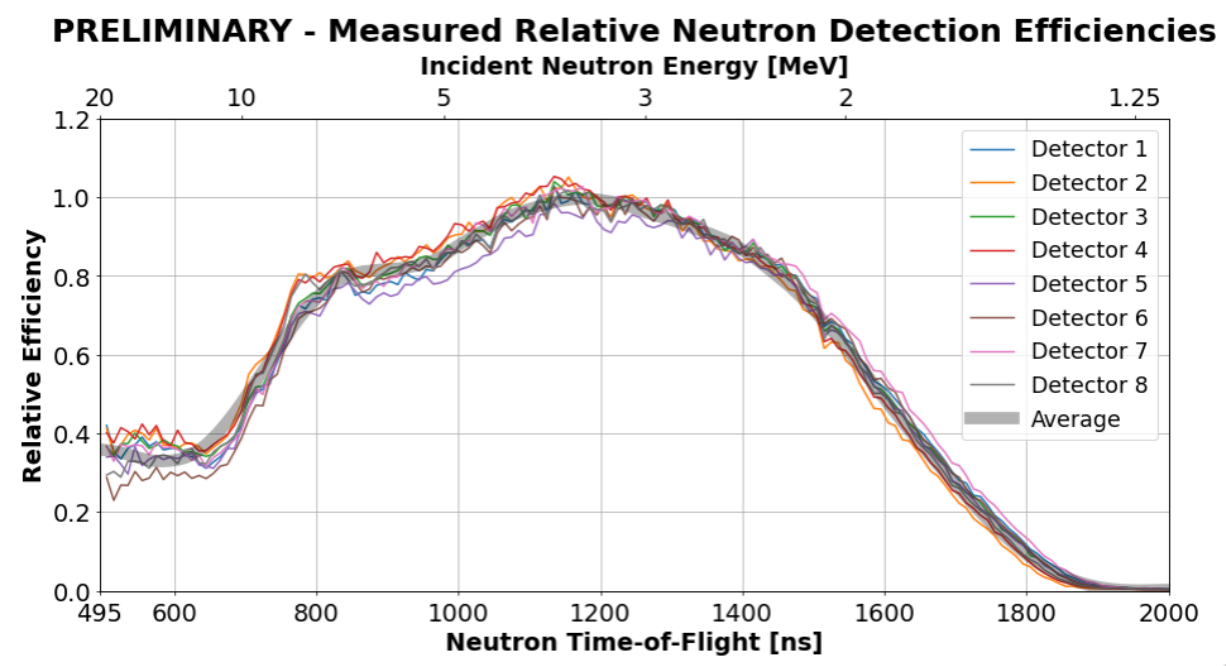
Pulse Shape Discrimination

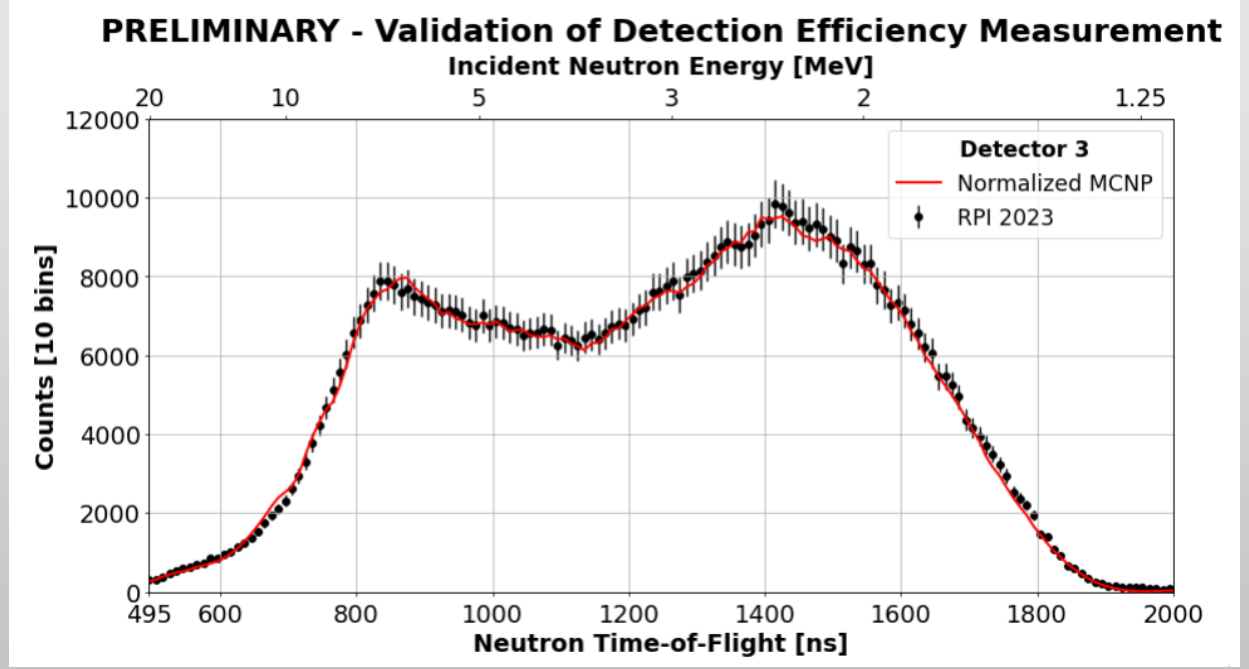
- Neutron and gamma pulses were separated based by the ratio of the tail integral of the pulse to the integral of the whole pulse
- Only pulse integrals above of 2000 were used for preliminary analysis



Detector Efficiency Determination

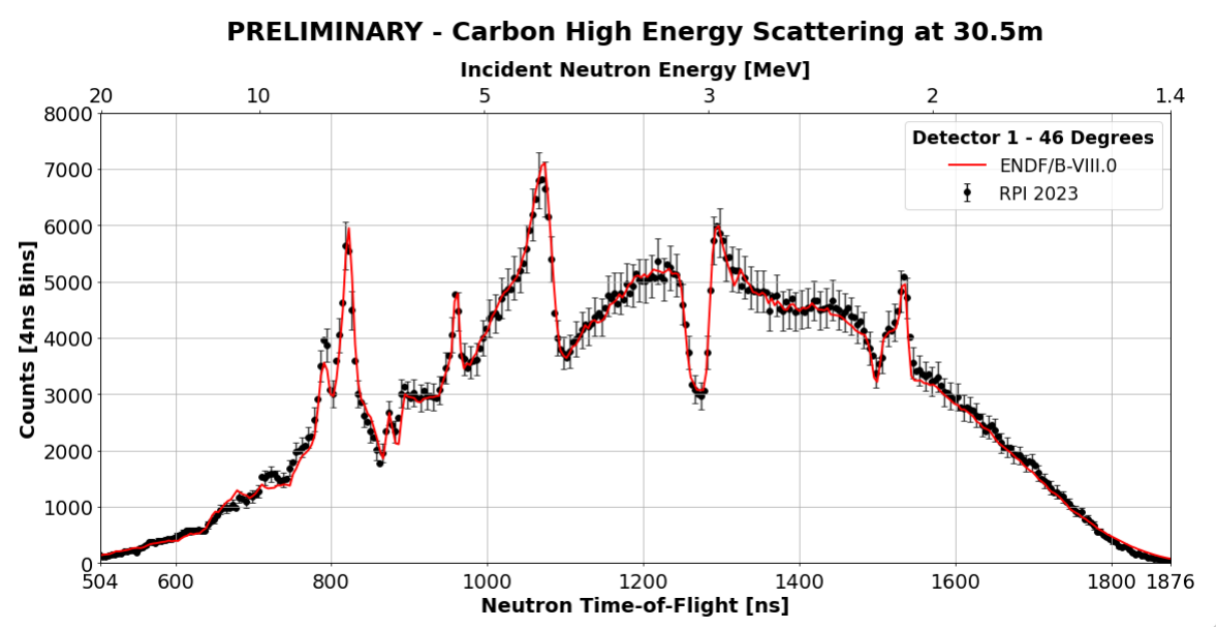
- EJ-301 detector efficiencies obtained from experiment and validated with SCINFUL
  - Further validated by reproducing detector efficiency measurement in MCNP6
  - The average neutron detection efficiency of all measurements was used in preliminary analysis

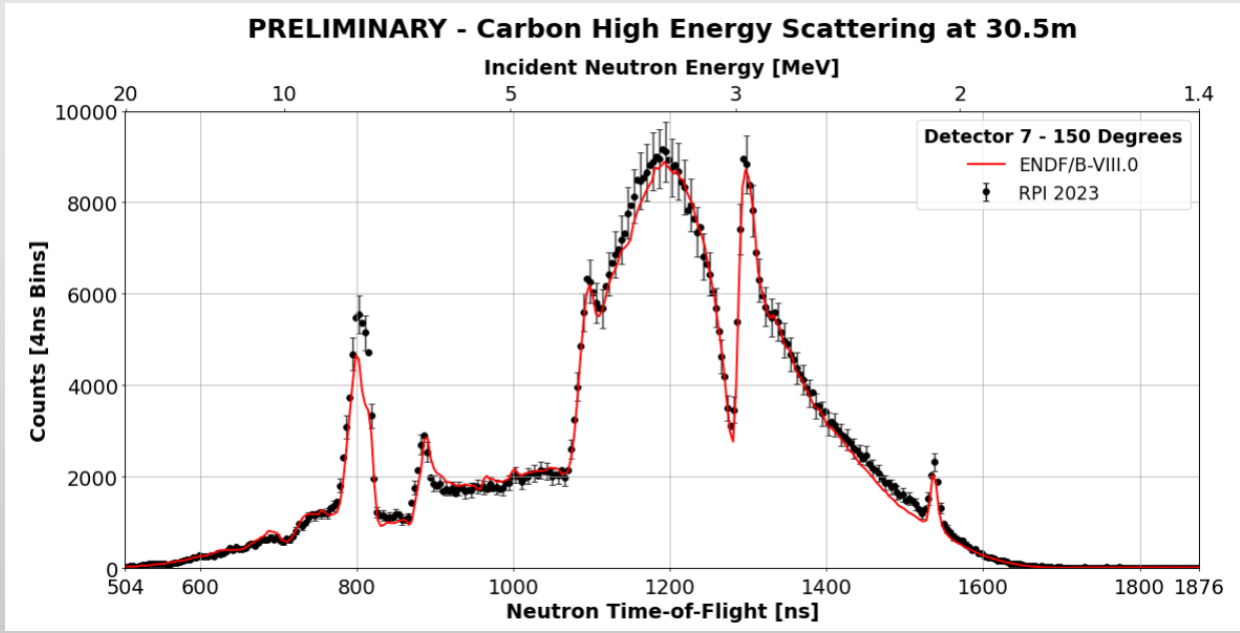




Validation Measurements of Carbon Sample

- An accurate quantification of the systematic uncertainty associated with the measurement has yet to be quantified, an arbitrary systematic uncertainty of 6% was used as an estimate from analyses performed on previous measurements[1,2]



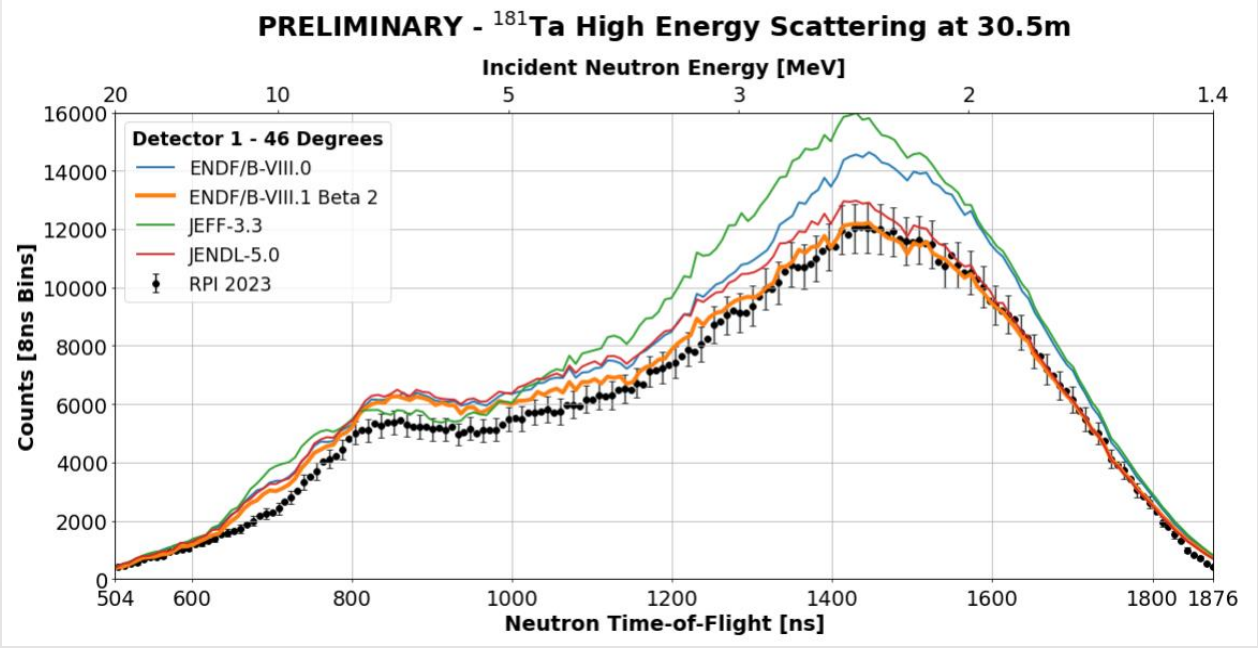


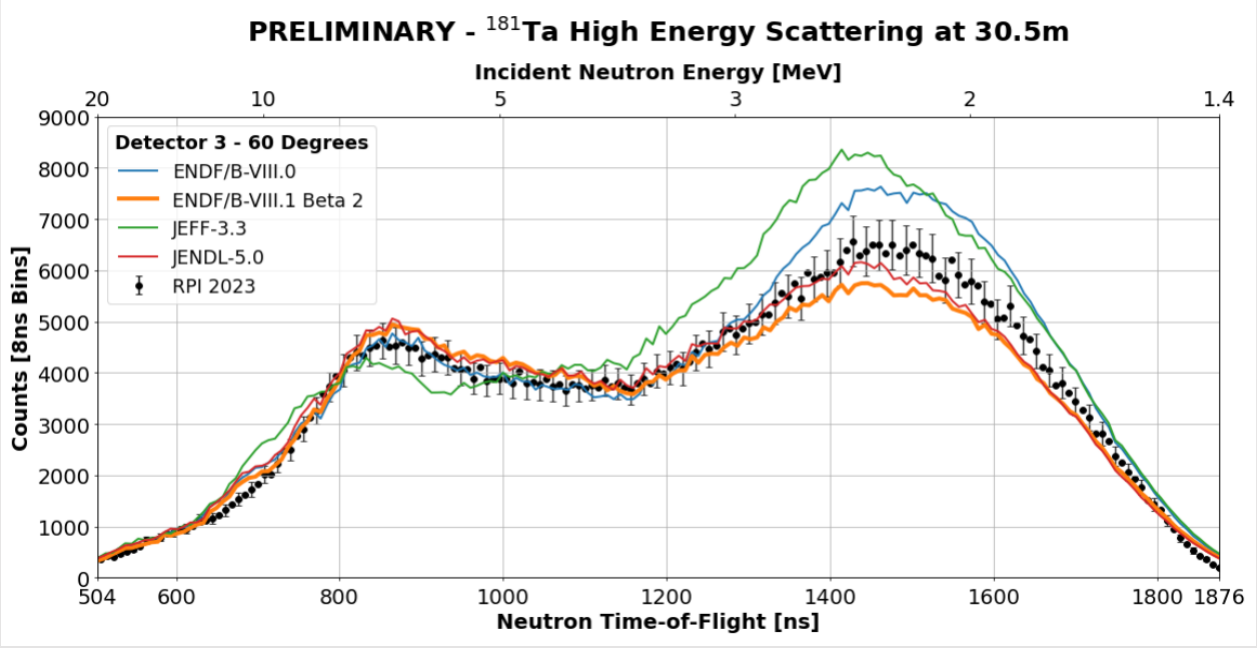
1. E. Blain, Y. Danon, D. P. Barry, B. E. Epping, A. Youmans, M. J. Rapp, A. M. Daskalakis and R. C. Block, "Measurements of Neutron Scattering from a Copper Sample Using a Quasi-Differential Method in the Region from 2 keV to 20 MeV", *Nuclear Science and Engineering*, vol. 196, no. 2, pp. 121-132, 2022, DOI:10.1080/00295639.2021.1961542
2. A. M. Daskalakis, E. J. Blain, B. J. McDermott, R. M. Bahran, Y. Danon, D. P. Barry, R. C. Block, M. J. Rapp, B. E. Epping and G. Leinweber, "Quasi-differential elastic and inelastic neutron scattering from iron in the MeV energy range", *Annals of Nuclear Energy*, vol. 110, pp. 603 - 612, 2017, DOI:10.1016/j.anucene.2017.07.007

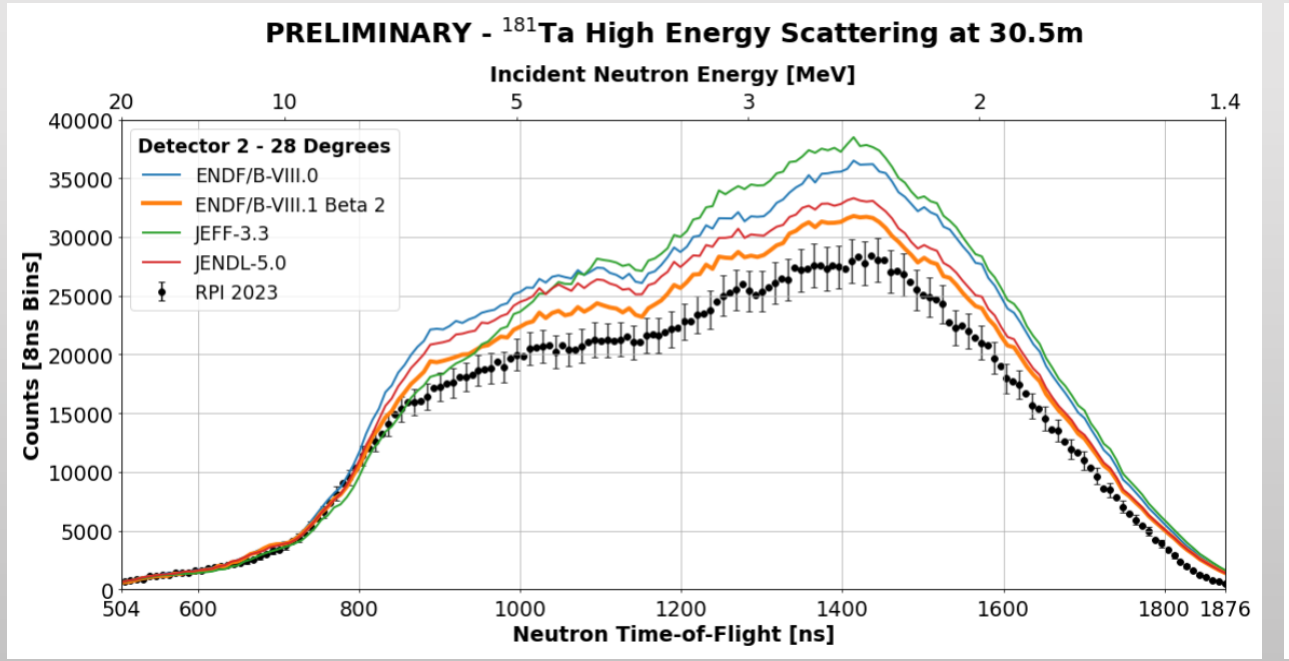
Preliminary Results from ^{181}Ta and Teflon Scattering Experiments

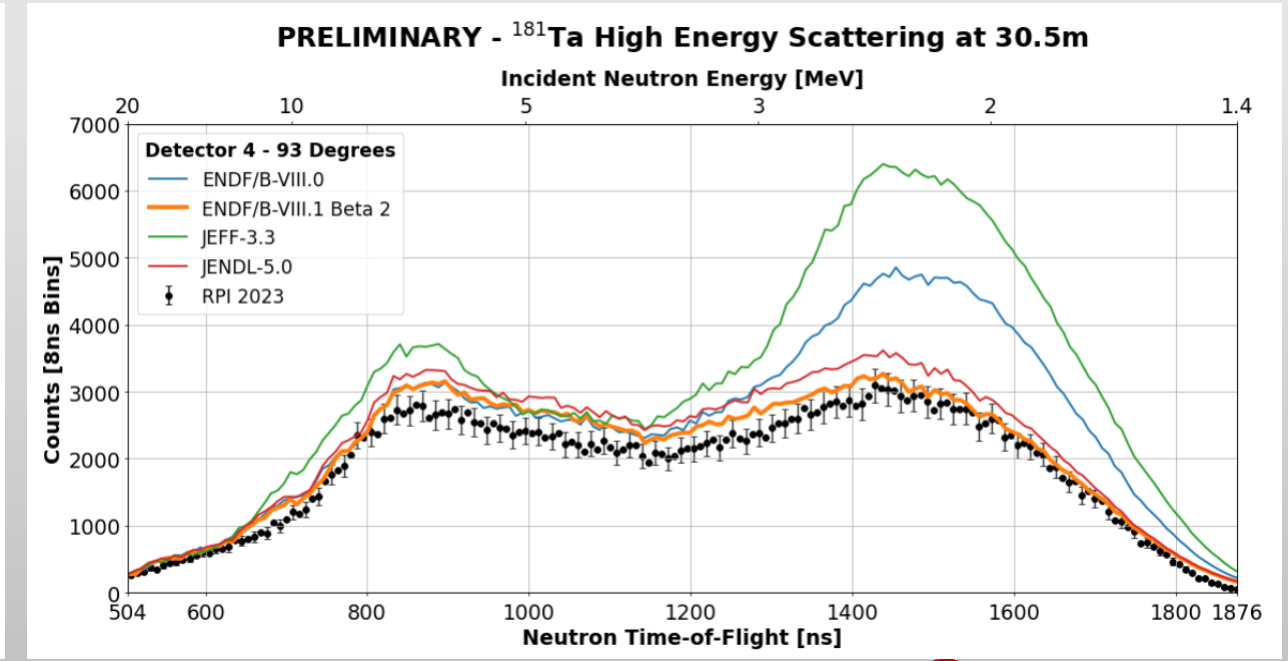
Preliminary Results From ^{181}Ta Experiment

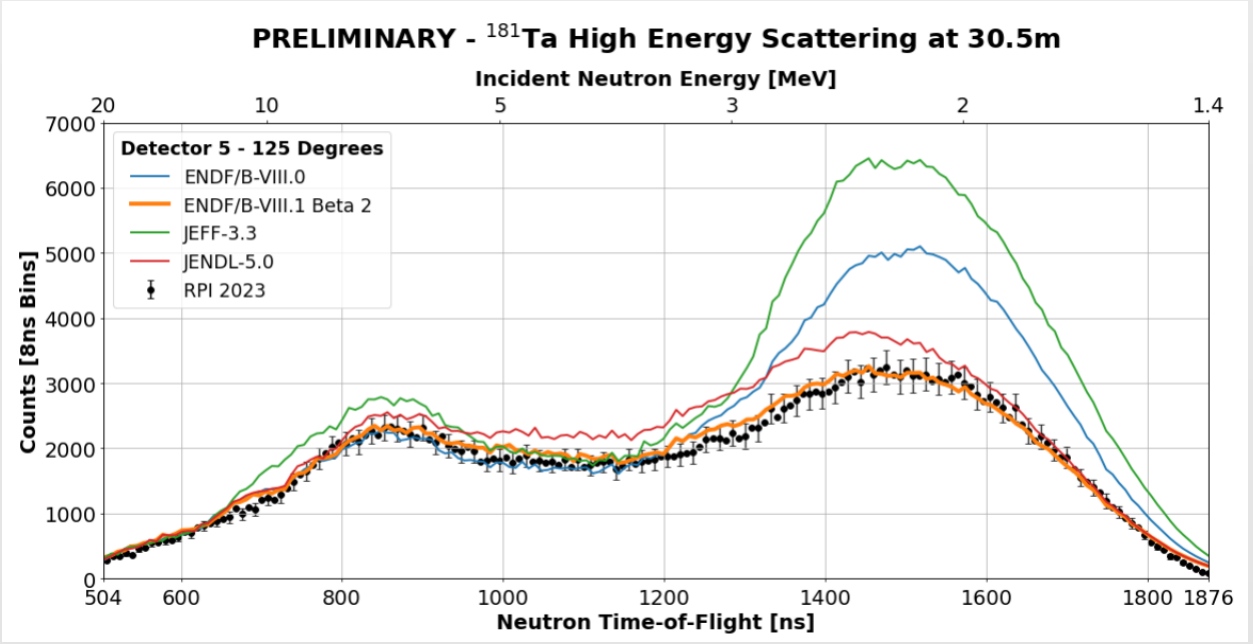
- First experiment presented is quasi-differential scattering measured from a 2.15*in* thick 3*in* diameter cylinder of ^{181}Ta
- Over 75 hours of data were collected during this experiment to obtain a high degree statistical accuracy
- Nuclear data from the following evaluations were used in MCNP6 to perform the transport calculations of the experiment:
  - ENDF/B-VIII.0
  - ENDF/B-VIII.1 (Beta 2)
  - JEFF-3.3
  - JENDL-5.0
- The estimation of 6% systematic uncertainty was also applied to these data

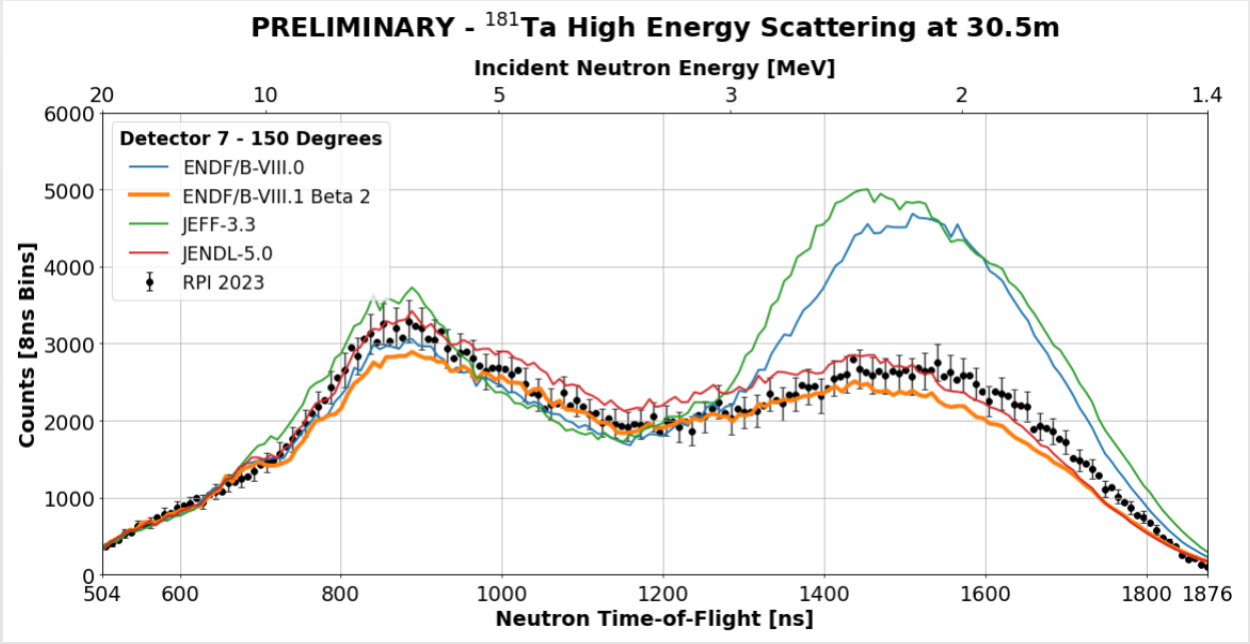

PRELIMINARY - 181Ta High Energy Scattering at 30.5m
Incident Neutron Energy [MeV]
20 10 5 3 2 1.4
Detector 1 - 46 Degrees
ENDF/B-VIII.0
ENDF/B-VIII.1 Beta 2
JEFF-3.3
JENDL-5.0
RPI 2023
Counts [8ns Bins]
0 2000 4000 6000 8000 10000 12000 14000 16000
504 600 800 1000 1200 1400 1600 1800 1876
Neutron Time-of-Flight [ns]

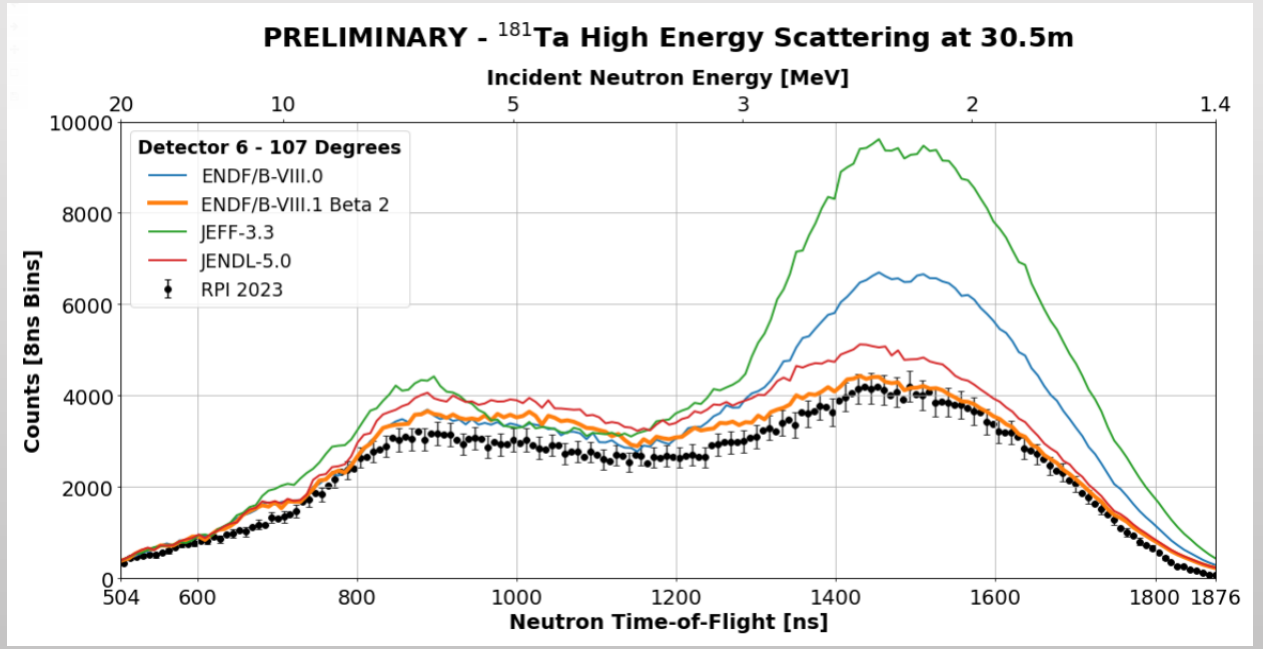

PRELIMINARY - 181Ta High Energy Scattering at 30.5m
Incident Neutron Energy [MeV]
20 10 5 3 2 1.4
Detector 3 - 60 Degrees
ENDF/B-VIII.0
ENDF/B-VIII.1 Beta 2
JEFF-3.3
JENDL-5.0
RPI 2023
Counts [8ns Bins]
0 1000 2000 3000 4000 5000 6000 7000 8000 9000
504 600 800 1000 1200 1400 1600 1800 1876
Neutron Time-of-Flight [ns]

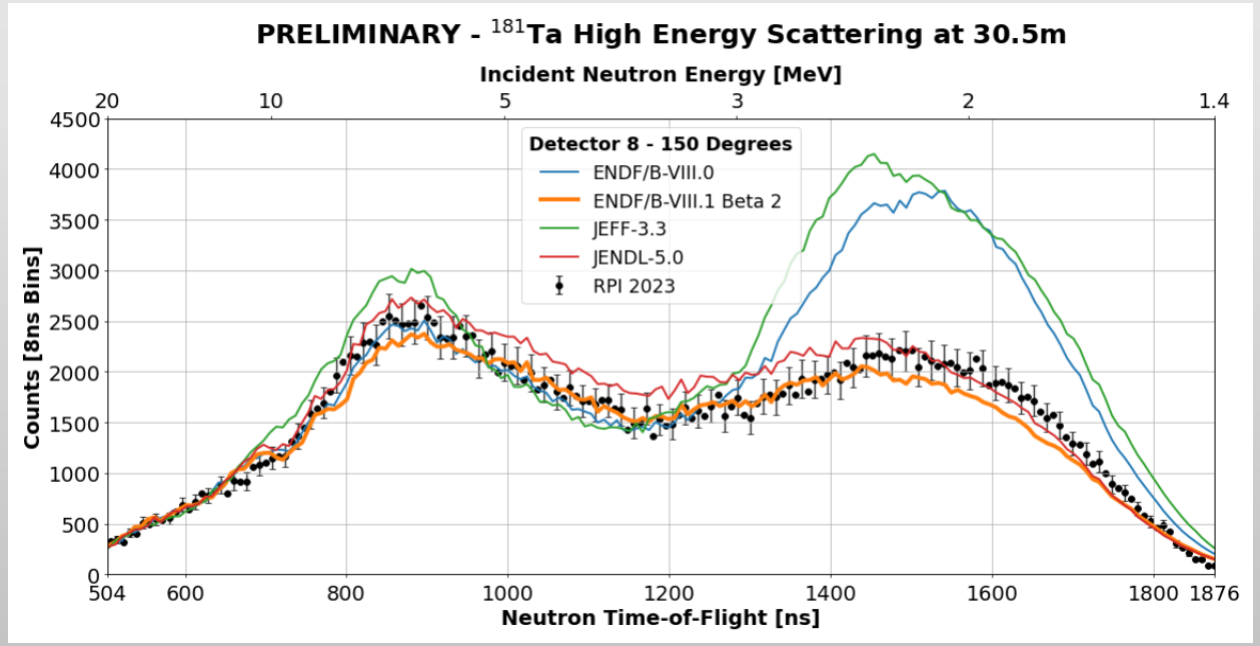

PRELIMINARY - 181Ta High Energy Scattering at 30.5m
Incident Neutron Energy [MeV]
20 10 5 3 2 1.4
Detector 2 - 28 Degrees
ENDF/B-VIII.0
ENDF/B-VIII.1 Beta 2
JEFF-3.3
JENDL-5.0
RPI 2023
Counts [8ns Bins]
0 5000 10000 15000 20000 25000 30000 35000 40000
504 600 800 1000 1200 1400 1600 1800 1876
Neutron Time-of-Flight [ns]


PRELIMINARY - 181Ta High Energy Scattering at 30.5m
Incident Neutron Energy [MeV]
20 10 5 3 2 1.4
Detector 4 - 93 Degrees
ENDF/B-VIII.0
ENDF/B-VIII.1 Beta 2
JEFF-3.3
JENDL-5.0
RPI 2023
Counts [8ns Bins]
0 1000 2000 3000 4000 5000 6000 7000
504 600 800 1000 1200 1400 1600 1800 1876
Neutron Time-of-Flight [ns]


PRELIMINARY - 181Ta High Energy Scattering at 30.5m
Incident Neutron Energy [MeV]
20
10
5
3
2
1.4
Detector 5 - 125 Degrees
ENDF/B-VIII.0
ENDF/B-VIII.1 Beta 2
JEFF-3.3
JENDL-5.0
RPI 2023
Counts [8ns Bins]
0
1000
2000
3000
4000
5000
6000
7000
504
600
800
1000
1200
1400
1600
1800
1876
Neutron Time-of-Flight [ns]


PRELIMINARY - 181Ta High Energy Scattering at 30.5m
Incident Neutron Energy [MeV]
20
10
5
3
2
1.4
Detector 7 - 150 Degrees
ENDF/B-VIII.0
ENDF/B-VIII.1 Beta 2
JEFF-3.3
JENDL-5.0
RPI 2023
Counts [8ns Bins]
0
1000
2000
3000
4000
5000
6000
504
600
800
1000
1200
1400
1600
1800
1876
Neutron Time-of-Flight [ns]


PRELIMINARY - 181Ta High Energy Scattering at 30.5m
Incident Neutron Energy [MeV]
20
10
5
3
2
1.4
Detector 6 - 107 Degrees
ENDF/B-VIII.0
ENDF/B-VIII.1 Beta 2
JEFF-3.3
JENDL-5.0
RPI 2023
Counts [8ns Bins]
0
2000
4000
6000
8000
10000
504
600
800
1000
1200
1400
1600
1800
1876
Neutron Time-of-Flight [ns]


PRELIMINARY - 181Ta High Energy Scattering at 30.5m
Incident Neutron Energy [MeV]
20
10
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3
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1.4
Detector 8 - 150 Degrees
ENDF/B-VIII.0
ENDF/B-VIII.1 Beta 2
JEFF-3.3
JENDL-5.0
RPI 2023
Counts [8ns Bins]
0
500
1000
1500
2000
2500
3000
3500
4000
4500
504
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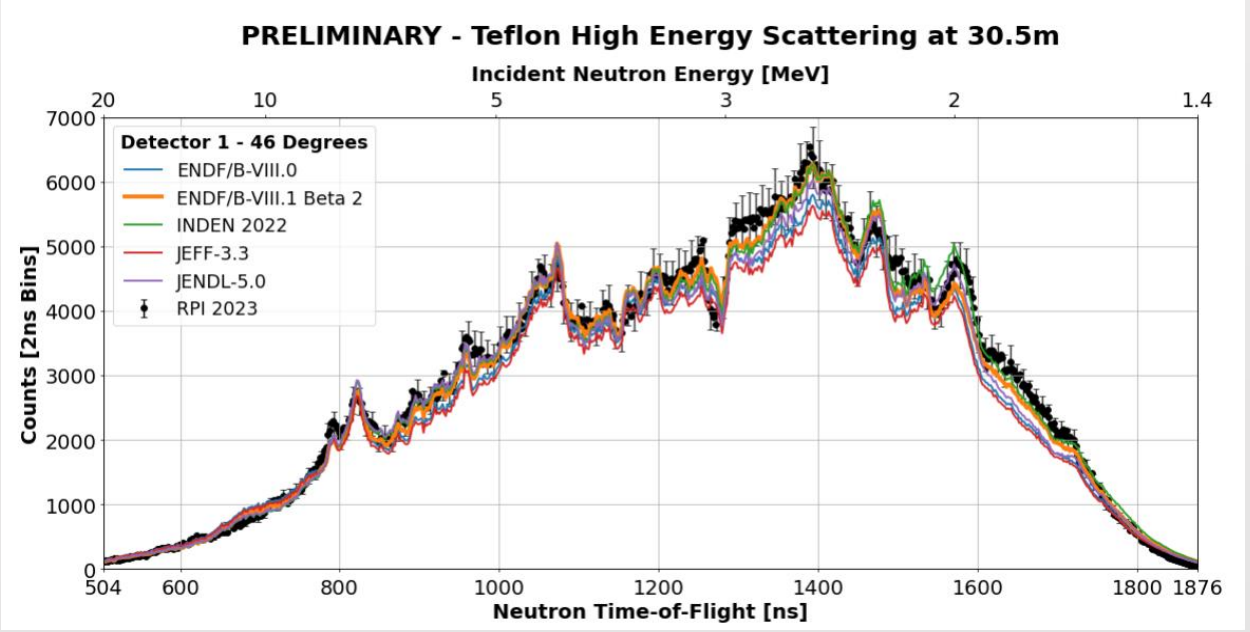
1000
1200
1400
1600
1800
1876
Neutron Time-of-Flight [ns]

Discussion on Preliminary ^{181}Ta Results

- ENDF/B-VIII.1 beta 2 evaluation best agrees with experimental data
  - Shape of ENDF/B-VIII.1 beta 2 evaluation seems accurate at all angles, but magnitude issues observed at most forward angles
- Large disagreements observed with ENDF/B-VIII.0 and JEFF-3.3 evaluations below 3 MeV
- JENDL-5.0 evaluation follows experimental data and ENDF/B-VIII.I beta 2 in some places, but more disagreements observed compared to beta 2

Preliminary Results From Teflon Experiment

- First experiment presented is quasi-differential scattering measured from a 1.95*in* thick 3*in* diameter cylinder of Teflon
- Over 135 hours of data were collected during this experiment to obtain a high degree statistical accuracy
- Nuclear data from the following evaluations were used in MCNP6 to perform the transport calculations of the experiment:
  - ENDF/B-VIII.0
  - ENDF/B-VIII.1 (Beta 2)
  - INDEN (2022 Evaluation)
  - JEFF-3.3
  - JENDL-5.0
- The estimation of 6% systematic uncertainty was also applied to these data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20
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Detector 1 - 46 Degrees
ENDF/B-VIII.0
ENDF/B-VIII.1 Beta 2
INDEN 2022
JEFF-3.3
JENDL-5.0
RPI 2023
Counts [2ns Bins]
0
1000
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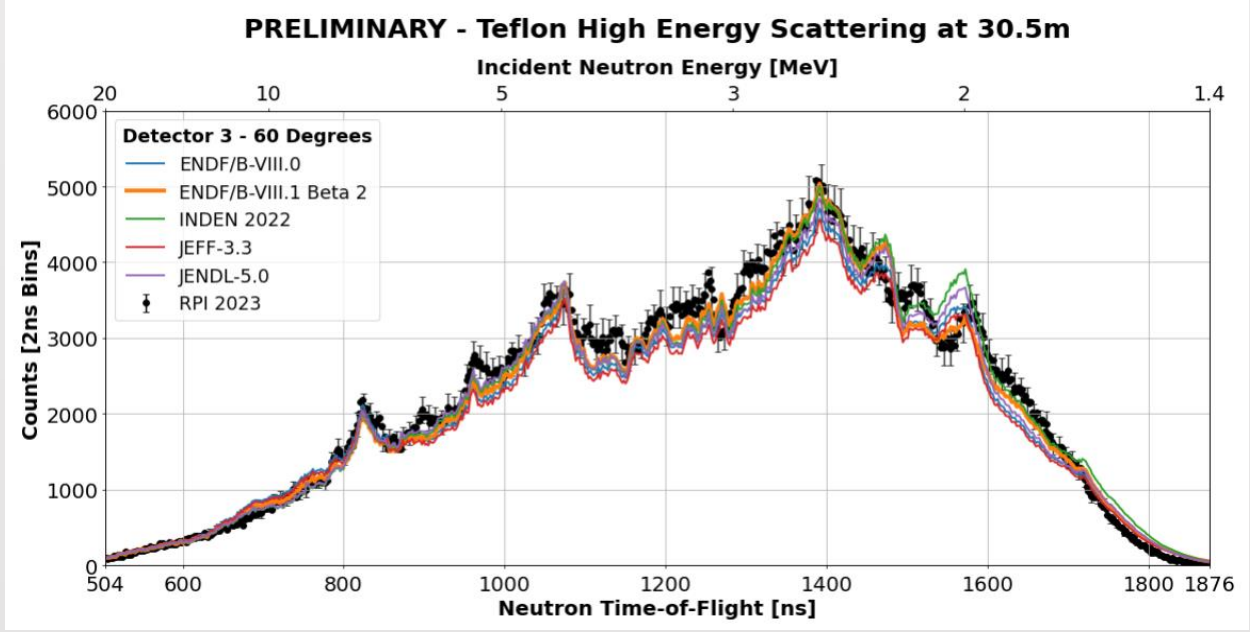
1876
Neutron Time-of-Flight [ns]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JEFF-3.3
JENDL-5.0
RPI 2023
Counts [2ns Bins]
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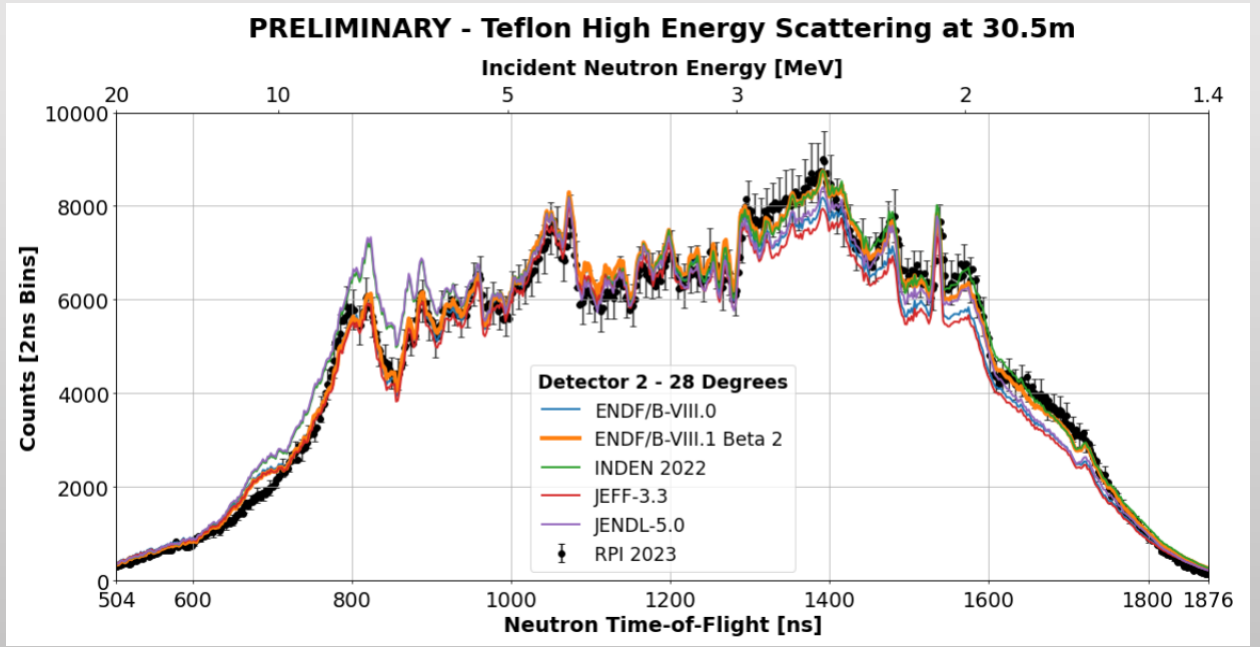
1876
Neutron Time-of-Flight [ns]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ENDF/B-VIII.1 Beta 2
INDEN 2022
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4000
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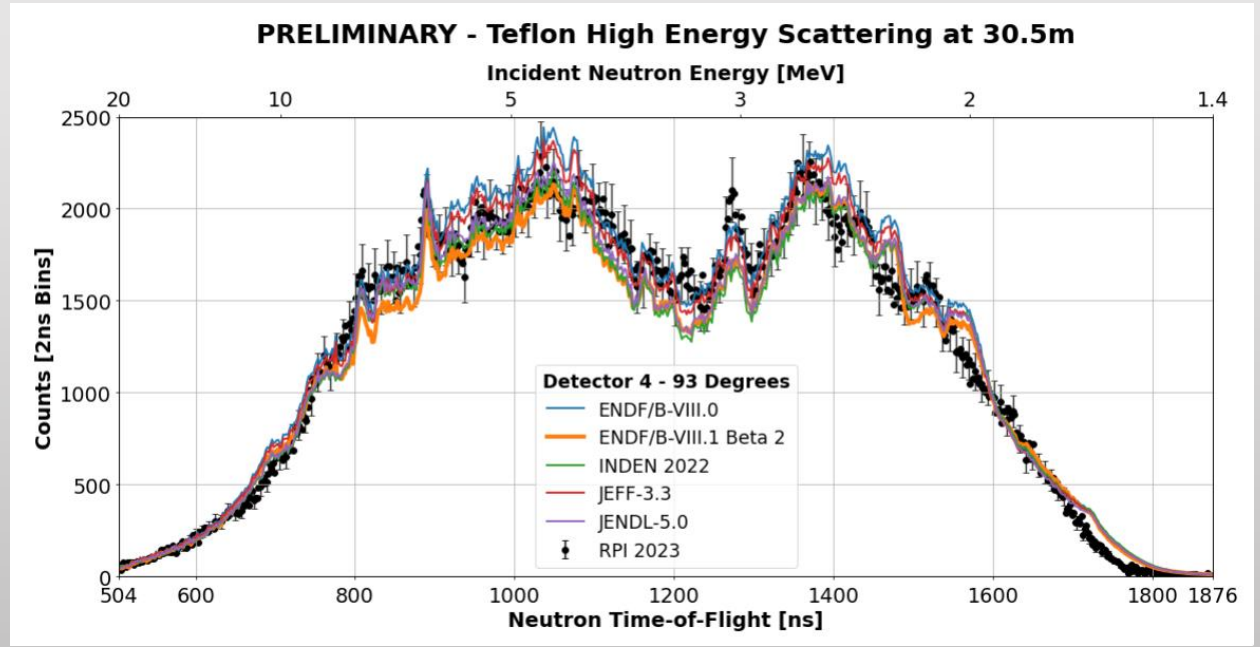
Neutron Time-of-Flight [ns]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RPI 2023
Counts [2ns Bins]
0
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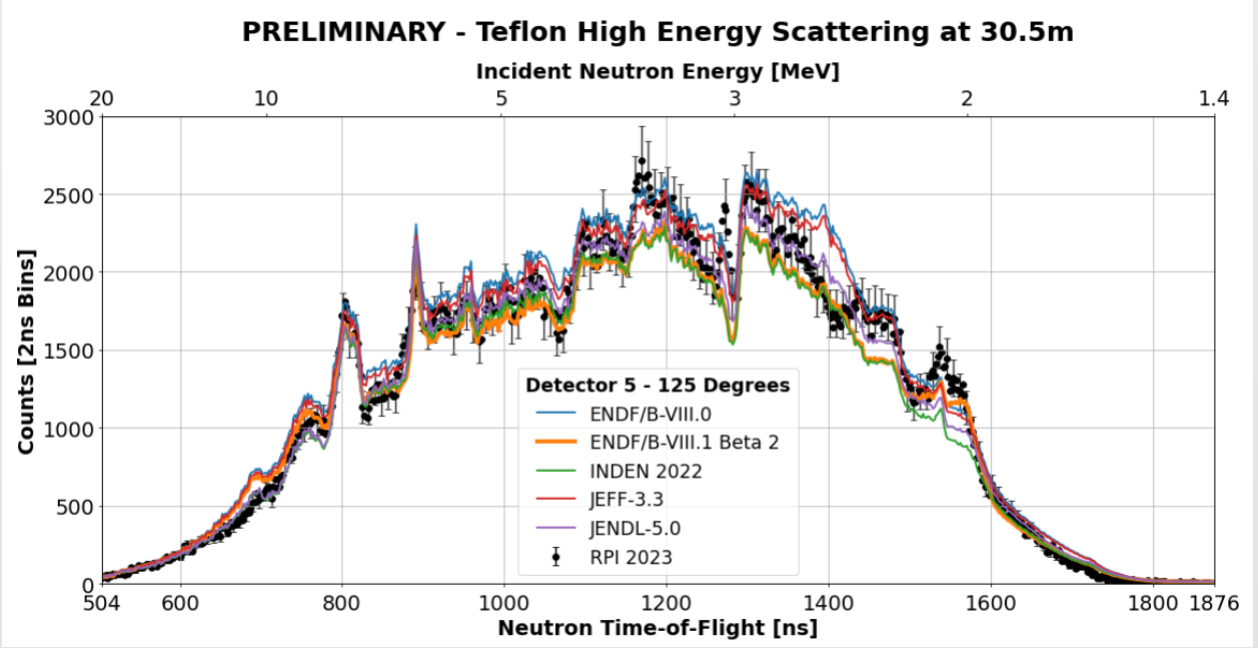
Neutron Time-of-Flight [ns]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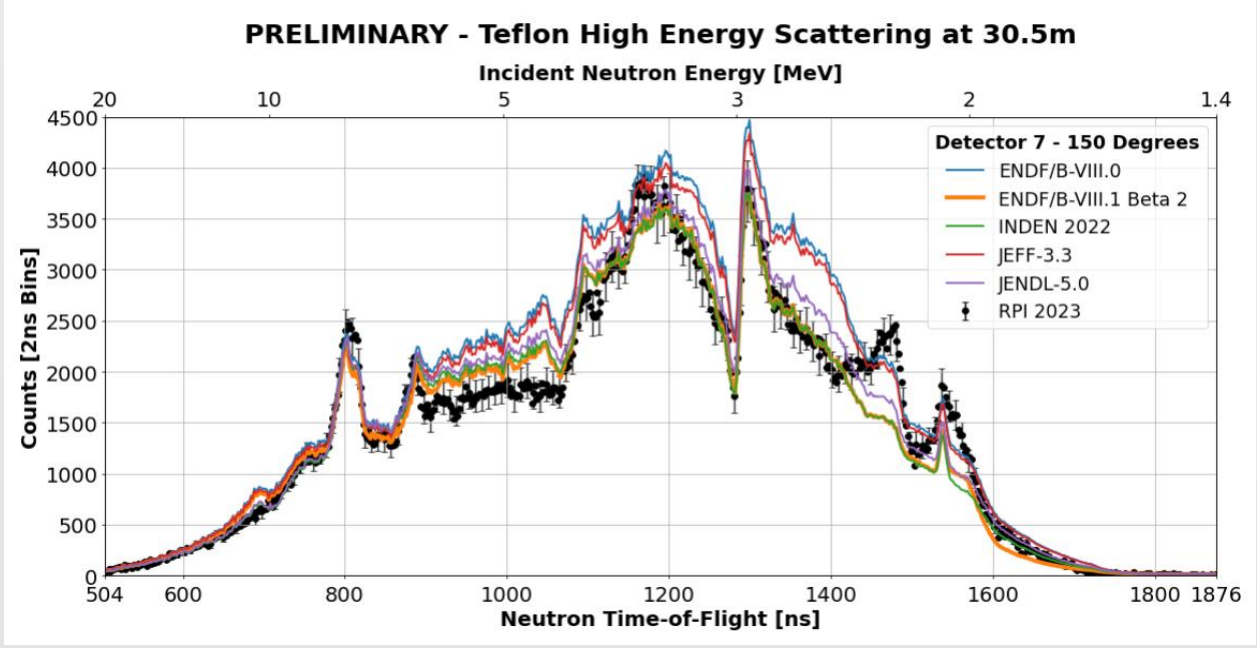
Counts [2ns Bins]
Neutron Time-of-Flight [ns]
Detector 5 - 125 Degrees
ENDF/B-VIII.0
ENDF/B-VIII.1 Beta 2
INDEN 2022
JEFF-3.3
JENDL-5.0
RPI 2023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Incident Neutron Energy [MeV]
Counts [2ns Bins]
Neutron Time-of-Flight [ns]
Detector 7 - 150 Degrees
ENDF/B-VIII.0
ENDF/B-VIII.1 Beta 2
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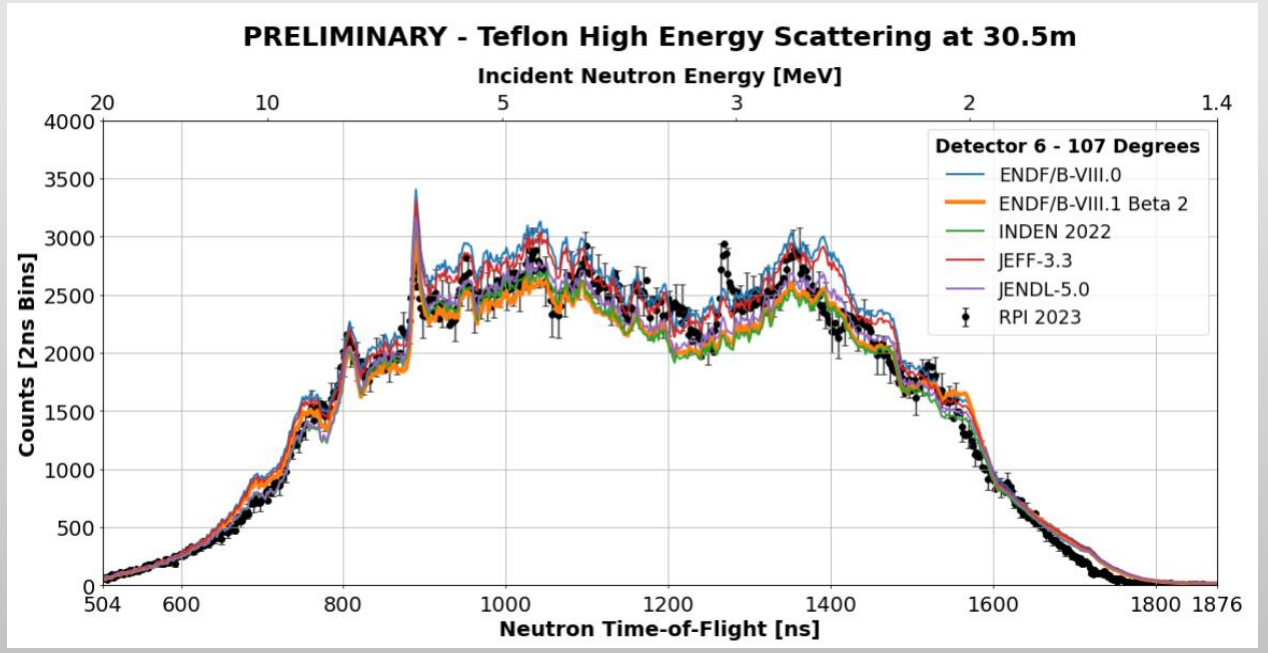
JEFF-3.3
JENDL-5.0
RPI 2023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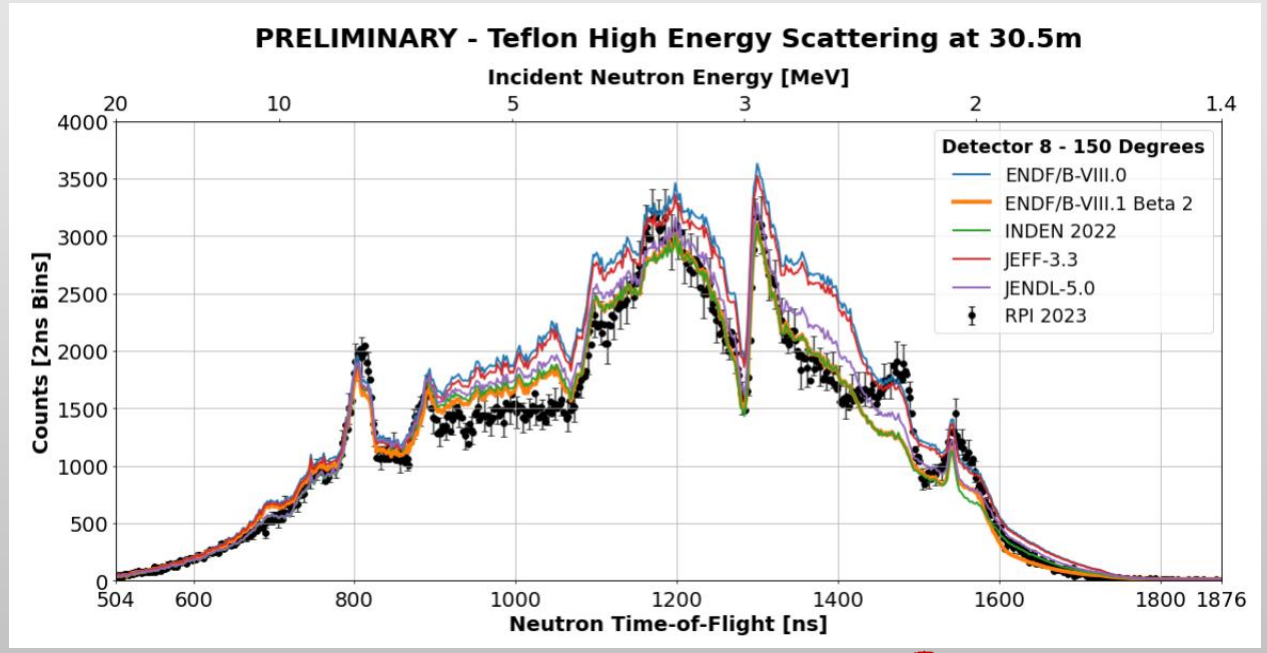
Incident Neutron Energy [MeV]
Counts [2ns Bins]
Neutron Time-of-Flight [ns]
Detector 6 - 107 Degrees
ENDF/B-VIII.0
ENDF/B-VIII.1 Beta 2
INDEN 2022
JEFF-3.3
JENDL-5.0
RPI 2023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Incident Neutron Energy [MeV]
Counts [2ns Bins]
Neutron Time-of-Flight [ns]
Detector 8 - 150 Degrees
ENDF/B-VIII.0
ENDF/B-VIII.1 Beta 2
INDEN 2022
JEFF-3.3
JENDL-5.0
RPI 2023

Discussion on Preliminary Teflon Results

- INDEN and ENDF/B-VIII.1 beta 2 evaluations observed to have best agreement with experimental data
- Evaluation and experimental agreement for forward angles is acceptable
  - ENDF/B-VIII.0 and JEFF-3.3 performance inferior to other evaluations in most regions
- Poor agreement between all evaluations observed at 150-degree detectors
  - Largest issues in big resonances and 3.5-6 MeV energy region
- Issues observed between 2 MeV and 3 MeV in 125-degree detector
- Resonance missing in evaluations seen in 107-degree detector at 3.1 MeV

Future Studies Needed for Final Experimental Results

Discussion on Pulse Shape Discrimination

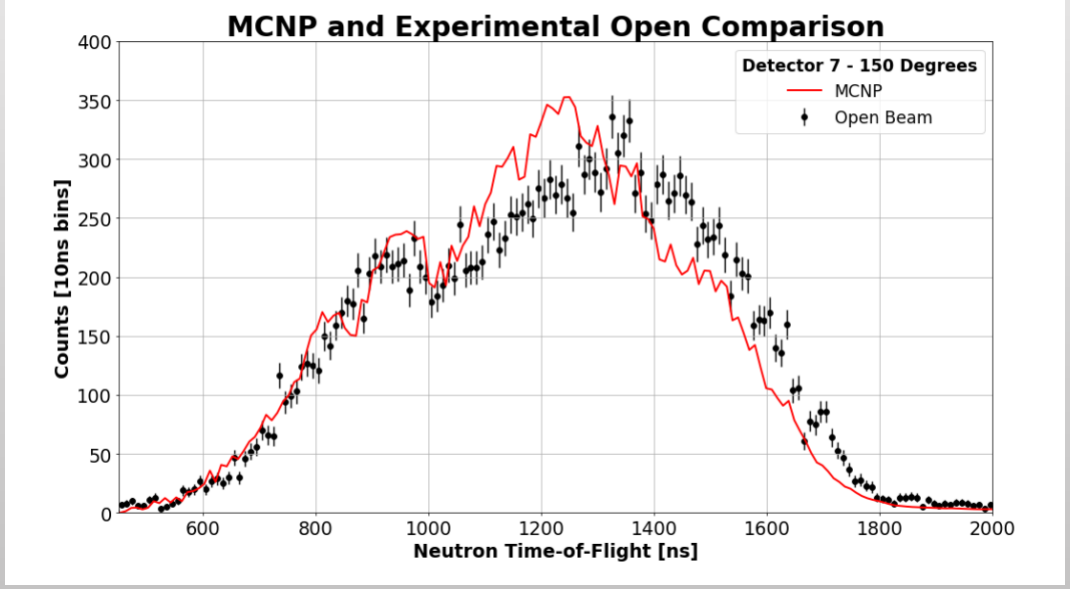
- Numerous pulse shape analysis methodologies are being explored at RPI to classify low integral pulses with high confidence
  - Goal: Increasing experimental region of interest to 0.5 MeV
- Methods being explored included:
  1. Pulse shape classification methods using reference pulse shapes for gammas and neutrons[1,2]
  2. Supervised and unsupervised neural networks
  3. Fourier filtering and frequency domain-based pulse shape analyses
  4. Continuous wavelet transform pulse shape analyses

1. A. M. Daskalakis, R. M. Bahran, E. J. Blain, B. J. McDermott, S. Piela, Y. Danon, D. P. Barry, G. Leinweber, R. C. Block, M. J. Rapp, R. Capote and A. Trkov, "Quasi-differential neutron scattering from ^{238}U from 0.5 to 20 MeV", *Ann. Nucl. Energy*, vol. 73, pp. 455-464, 2014, DOI:10.1016/j.anucene.2014.07.023
2. A. M. Daskalakis, E. J. Blain, B. J. McDermott, R. M. Bahran, Y. Danon, D. P. Barry, R. C. Block, M. J. Rapp, B. E. Epping and G. Leinweber, "Quasi-differential elastic and inelastic neutron scattering from iron in the MeV energy range", *Annals of Nuclear Energy*, vol. 110, pp. 603 - 612, 2017, DOI:10.1016/j.anucene.2017.07.007

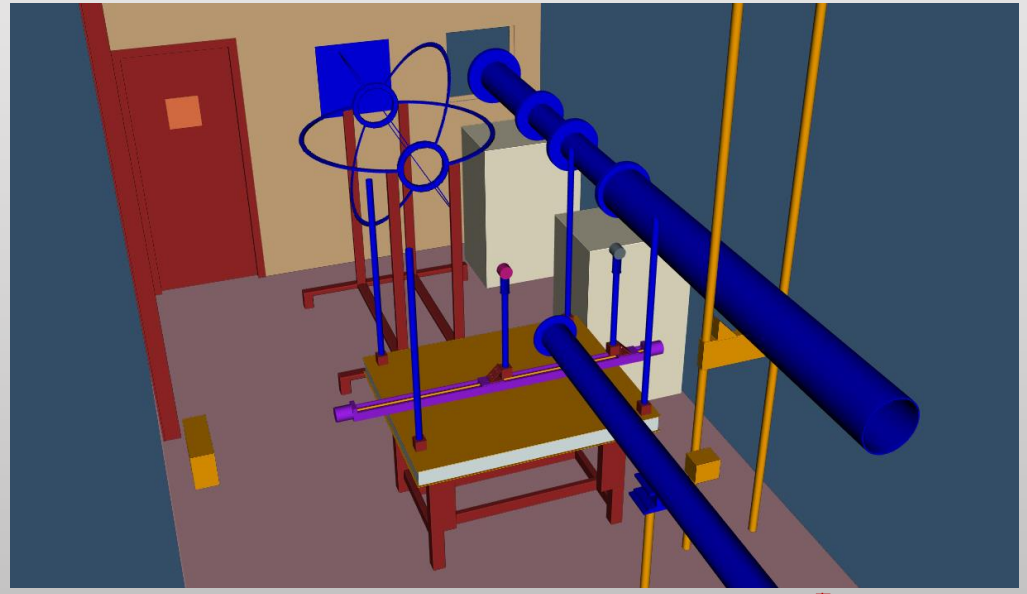
Improvements to MCNP Modeling

- Improvements to the MCNP modeling of the experimental geometry are needed to further reduce the systematic uncertainty of the experiment using the normalization factor technique
- Modeling of open beam needs improvement at 150-degree detector locations

Current Results



Future Implementation



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