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Title: COMPARISON OF 252CF TIME CORRELATED INDUCED FISSION WITH AmLi INDUCED FISSION ON FRESH MTR RESEARCH REACTOR FUEL

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# **COMPARISON OF $^{252}\text{CF}$ TIME CORRELATED INDUCED FISSION WITH AmLi INDUCED FISSION ON FRESH MTR RESEARCH REACTOR FUEL**

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MS in Nuclear Engineering

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May 10, 2017

# OUTLINE

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## OBJECTIVES

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- To calibrate Advanced Experimental Fuel Counter (AEFC)
  - $^{252}\text{Cf}$  in instead of traditional AmLi source
- To benchmark MCNP simulations using experimental results
- To investigate the effects of change in fuel assembly geometry to the count rates
- Finally, to show the boost in doubles count rates (coincidence rates) with  $^{252}\text{Cf}$  active source due to the time correlated induced fission (TCIF) effect

# MOTIVATION

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- MTRs around the world
  - With both highly enriched uranium (HEU) and low enriched uranium (LEU) fuel
  - $^{235}\text{U}$  can be separated directly from HEU fuel and diverted to the weapons program
  - Countries where MTRs are installed were committed to non-proliferation and their commitment needed verification
- Difficulties to obtain AmLi source in the US
- Better doubles rates obtained with  $^{252}\text{Cf}$  compared to AmLi in the past field trial in Uzbekistan in 2012 and 2014

## INTRODUCTION

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- The effective application of international safeguards to research reactors requires verification of spent fuel as well as fresh fuel.
- To accomplish this goal various nondestructive and destructive assay techniques have been developed in the US and around the world.
- The Advanced Experimental Fuel Counter (AEFC) is a nondestructive assay (NDA) system developed at Los Alamos National Laboratory (LANL). [1]

## INTRODUCTION (CONTINUED)

- Both neutron and gamma measurement capabilities
- Spent fuel assemblies are stored in water
  - The system designed to be watertight to facilitate underwater measurements by inspectors. [1]

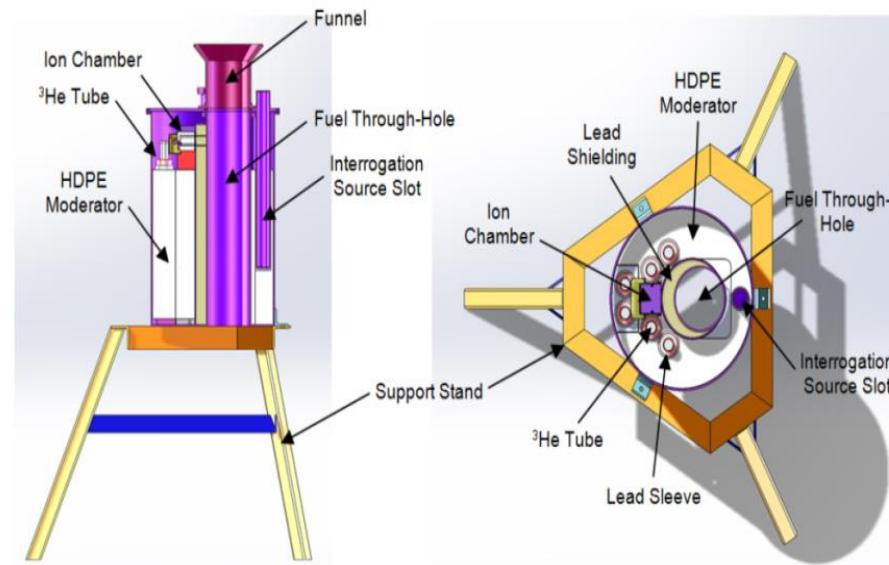


Figure 1: Mechanical design of the AEFC [1]

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## INTRODUCTION (CONTINUED)

- Six  ${}^3\text{He}$  neutron detectors and one ion chamber for gross gamma measurement
- Detectors are shielded with lead for high gamma doses
- Collimator for ion chamber
- Neutrons are moderated by HDPE and water
- Both active and passive neutron interrogation capabilities
- Active neutron measurement provides information about residual fissile mass
- Passive neutron measurement provides BU, IE, and CT [1]

## INTRODUCTION (CONTINUED)

- Passive gross gamma results provide information about BU, IE, and CT.
- In the past, active interrogation mostly used AmLi.
- In 2014 during the Uzbekistan field trial, AEFC was calibrated by  $^{252}\text{Cf}$ .
- Results showed better doubles count rates with  $^{252}\text{Cf}$  than AmLi. [2]
- Why would  $^{252}\text{Cf}$  give better coincidence results?
- $^{252}\text{Cf}$  was supposed to be complex due to its time correlated SF neutrons.

# INTRODUCTION (CONTINUED)

## Neutron Coincidence Counting

- In the AEFC, two or more neutrons are coincident if they are detected by any of the six  ${}^3\text{He}$  detectors within the specified gate window.
  - In this case, the gate window is 128  $\mu\text{sec}$ .

No. of Neutrons	1	2	3	4	n
No. of Coincidences	0	1	3	6	$n(n-1)/2$

- The coincidences measured can come from accidentals from background and active source. [6]

## INTRODUCTION (CONTINUED)

- Accidentals can be separated from the real coincidences from fuel.
- The process of separation is explained by the Rossi-Alpha distribution.

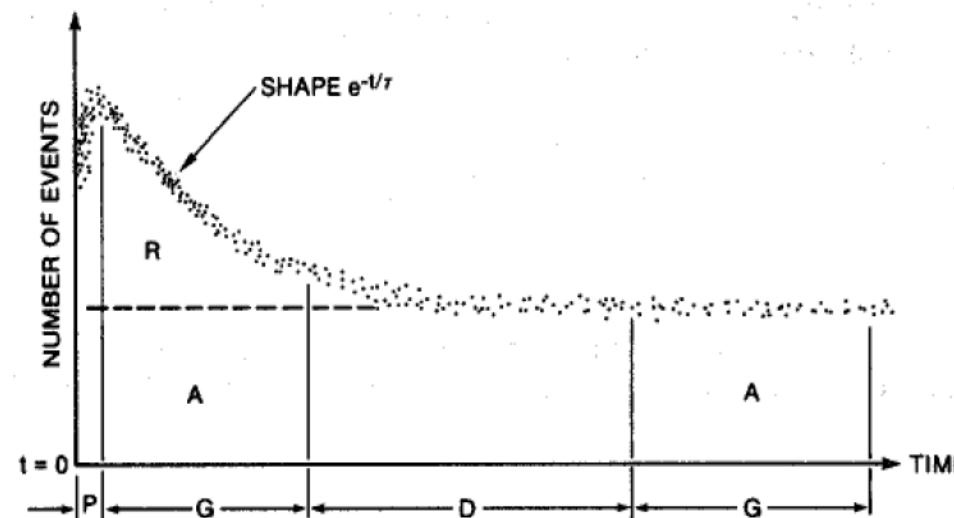


Figure 2: Rossi-Alpha distribution [5]

## INTRODUCTION (CONTINUED)

- P is a pre-delay, G is a gate length, and D is a long delay gate[5]
- First neutron detected inside any of six  ${}^3\text{He}$  detectors acts as a trigger and opens a time window.
- All neutrons detected within that specified time window are time correlated coincidences to the initial triggering neutron. [6]
- Subsequently, each neutron after the first triggering, neutron triggers its own window of equal time length and thus a distribution is produced. [6]

## INTRODUCTION (CONTINUED)

- If a random source (AmLi) is measured, a flat distribution will be obtained.
- If a source that emits time correlated neutron ( $^{252}\text{Cf}$ ) is measured, the distribution obtained will look like an exponential function given by

$$S(t) = A + R e^{-t/\tau}$$

- Where A is accidental coincidence and R is real coincidence [5]

## INTRODUCTION (CONTINUED)

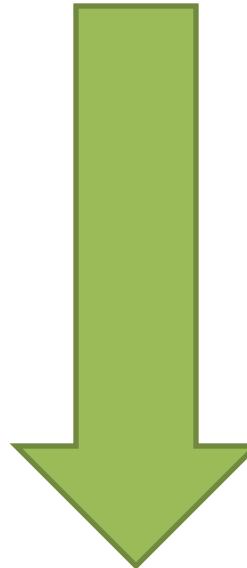
- Initially, coincidence counting concept was
  - source would produce random neutron
  - fuel would produce correlated neutron
  - net correlated neutrons from fuel itself
- However in the asymmetric system such as AEFC,  $^{252}\text{Cf}$  gives better results. [2]
  - Very small source-detector efficiency
  - Negligible contribution from the background (active source)
- Experimental - The source-detector is over moderated and efficiency is less than 1%
  - Fuel gets in the source-detector line of sight
  - The efficiency reduces even more

## INTRODUCTION (CONTINUED)

- MCNP - Fuel-detector efficiency is approximately 5%
- Effect of time correlated SF neutron background from  $^{252}\text{Cf}$  is negligible.
- Average energy of AmLi neutron 0.3 MeV is much lower than average energy of  $^{252}\text{Cf}$  of 2.3 MeV.
- AmLi neutrons are thermalized much faster than  $^{252}\text{Cf}$  neutrons.
- Probability of inducing fission in the fuel is much higher with AmLi.
- Once again, AmLi seems to be favorable to produce better coincidence results.

## INTRODUCTION (CONTINUED)

- The reason why  $^{252}\text{Cf}$  gives better coincidence results is



# INTRODUCTION (CONTINUED)

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Time Correlated Induced Fission (TCIF) Effect

## INTRODUCTION (CONTINUED)

- Term was brought up by Dr. Howard Menlove at the LANL.
- Related to the neutrons emitted in IF event from the fuel assembly coupled to the neutrons produced in a precursor fission event in an active interrogation source
- SF neutron from active source are time correlated with the **IF** neutron from the fuel.
- Combined multiplicity is higher with  $^{252}\text{Cf}$ .
- Only one IF event is possible per random  $(\alpha, n)$  reaction with AmLi.

## INTRODUCTION (CONTINUED)

- More than one IF per SF event with  $^{252}\text{Cf}$
- Boost in doubles rate is due to the boost in combined multiplicity due to the TCIF effect of  $^{252}\text{Cf}$ .

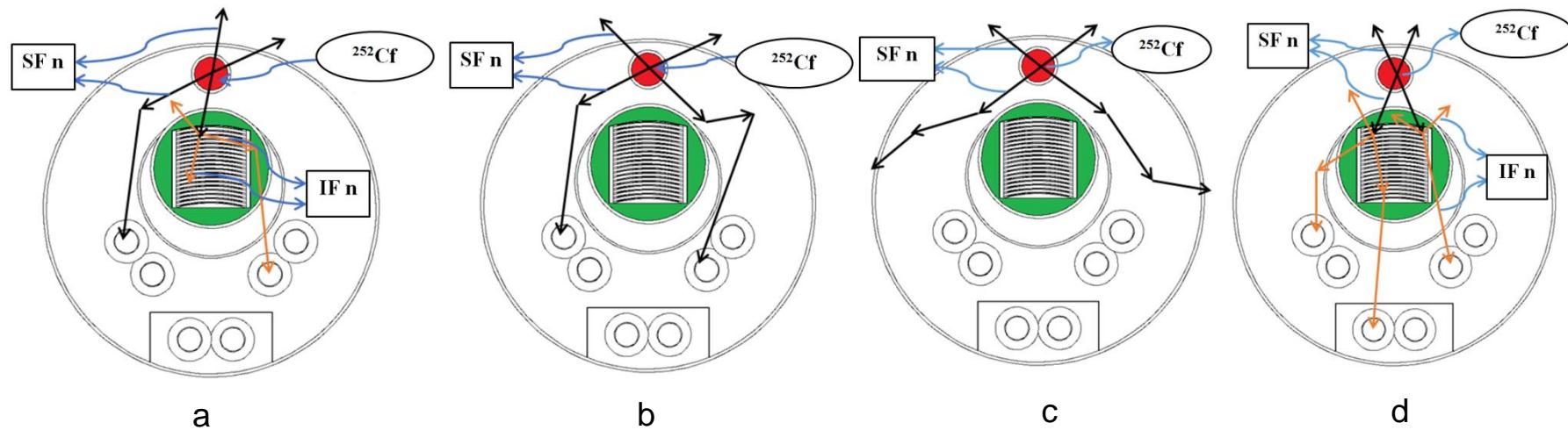


Figure 3: Possible neutron detection scenarios with  $^{252}\text{Cf}$  active sources

## MATERIALS USED

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- Clean AEFC system
- Water tank
- Crane to move AEFC and fuel assembly
- JSR-15 Shift Register
  - HV: 1680 V
  - Pre-delay: 4.5  $\mu$ sec
  - R+A or A Gate: 128  $\mu$ sec
- Laptop with INCC
- L-108 and O-187R, MTR type fresh fuel assemblies
- $^{252}\text{Cf}$  (A7866)- 45,670 n/s
- $^{252}\text{Cf}$  (A7869)- 170,695 n/s
- AmLi (N-165) – 37,940 n/s

## MATERIALS USED (CONTINUED)

- Key Dimensions
  - Full Assembly (FA) L-108: 108 cm long, while 60 cm active fuel meat
  - Partial Assembly (PA) O-187R: 90 cm long, while 60 cm active fuel meat
  - AEFC
    - Height: 112 cm
    - Axial center of  ${}^3\text{He}$  detectors at 67.67 cm above the base of the tank
    - Fuel through hole diameter: 12 cm
    - Length of  ${}^3\text{He}$  detectors: 25 cm

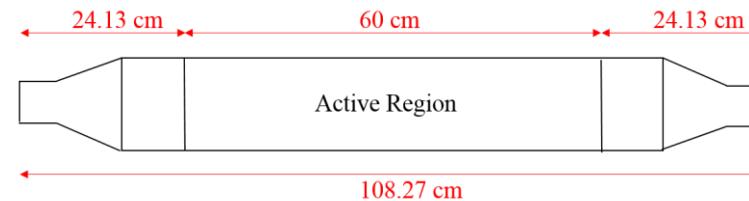


Figure 4: MTR fuel assembly

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## MATERIALS USED (CONTINUED)

- Water Tank Dimensions

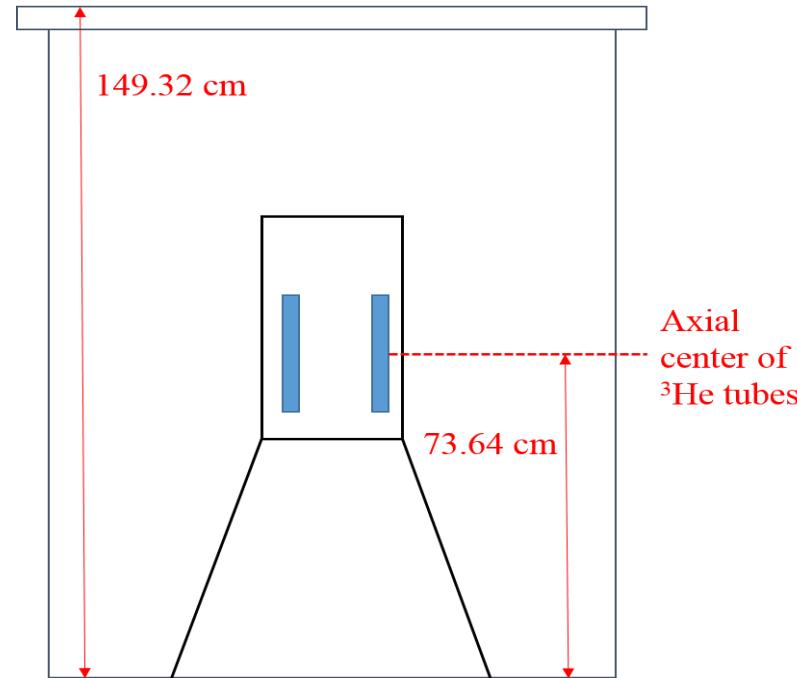


Figure 5: AEFC inside poly water tank

## MATERIALS USED (CONTINUED)



Figure 6: AEFC outside the water tank



Figure 7: AEFC + MTR inside water [4]

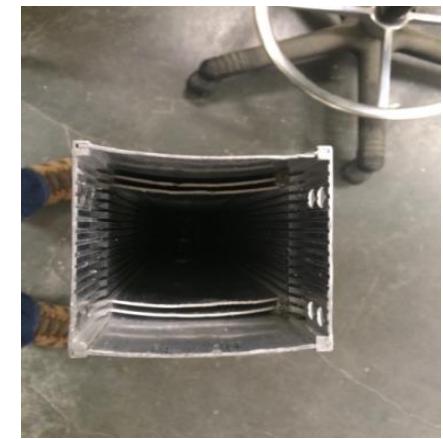


Figure 8: O-187R MTR fuel assembly

## METHODOLOGY

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### Experimental Measurement Procedure

- FA-<sup>252</sup>Cf measurements :
  - Completely lowered to the base of the water tank
  - 23 point measurements: 3 cm increment axially upward each time
  - Mid point measurement (13.67 cm above the base of the water tank)
  - Mid point measurement: fuel assembly rotated 90 degree
  - Bottom point measurement: 18 cm below the mid point measurement configuration
- FA-AmLi Measurements:
  - Mid point measurement: AmLi

## METHODOLOGY (CONTINUED)

- PA-<sup>252</sup>Cf measurements :
  - Mid point measurement (31.32 cm above the base of the water tank)
  - Mid point measurement: fuel assembly rotated 90 degree
  - Top and Bottom point measurement: 18 cm above and below the mid point measurement configuration
- PA-AmLi Measurements:
  - Mid point measurement: AmLi

## METHODOLOGY (CONTINUED)

- MTR fuel plates in fabricated fuel holder:
  - Mid point measurement: varying fuel plates with  $^{252}\text{Cf}(\text{A7869})$
  - With 1, 8, and 9 plates

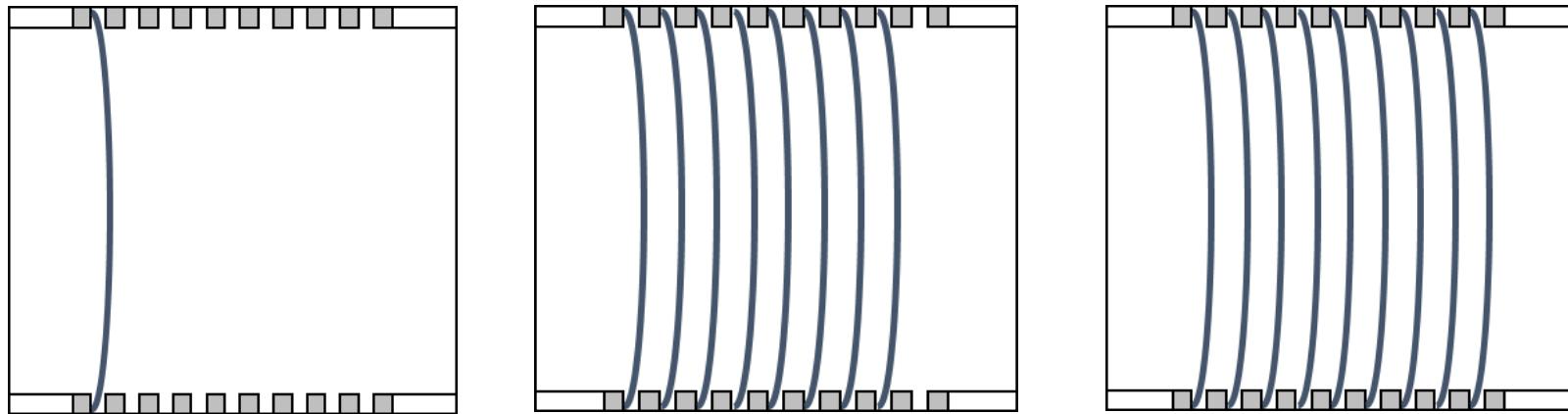


Figure 9: Varying Fuel plates in a fuel holder

## METHODOLOGY (CONTINUED)

### MCNP Simulation Procedure

- Replicated experiments
- Additional simulations
  - Varying fuel plates: 4, 6, 8, 10, ..., 18, 19 plates
  - Varying enrichment: 20% to 93.5%

# RESULTS AND ANALYSIS

- Count rates normalized to  $^{252}\text{Cf}(\text{A7866})$
- FA Benchmarking
  - Singles (S): within 5% up to 30 cm
  - Doubles (D): within 4% up to 30 cm
  - S and D diverging after 30 cm
  - $3 * \sigma$  error bars

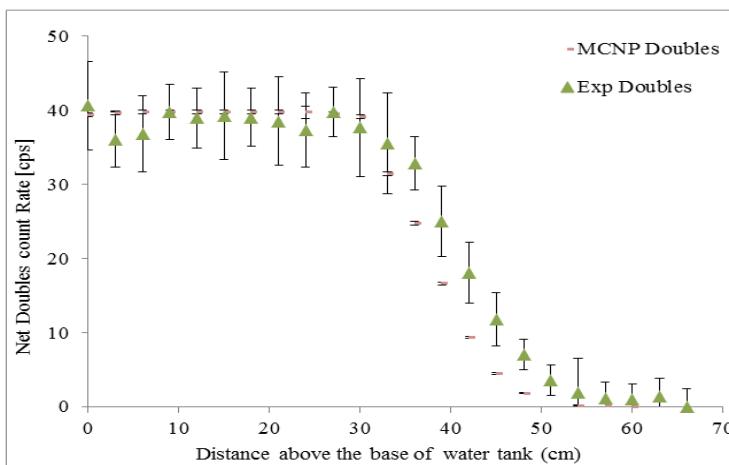


Figure 11: L-108 net doubles count rates

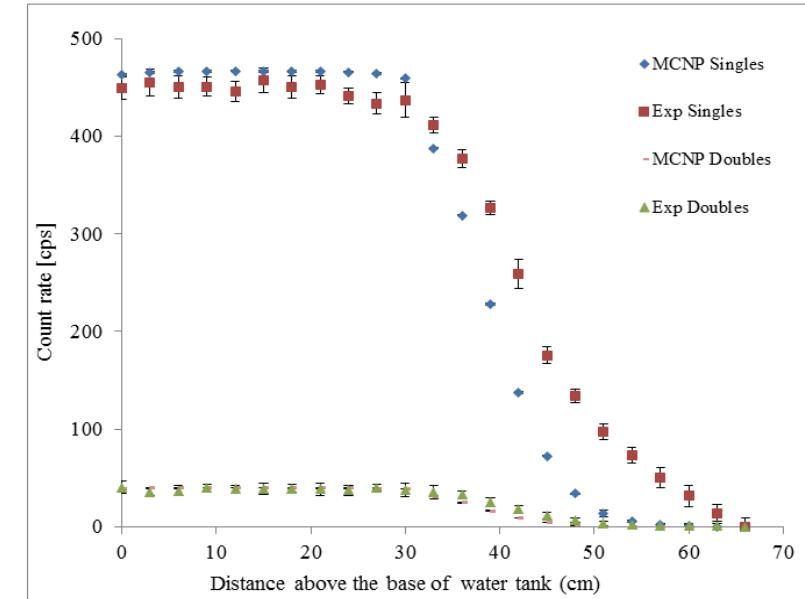


Figure 10: L-108 scan with  $^{252}\text{Cf}$  SF source

# RESULTS AND ANALYSIS (CONTINUED)

- PA Benchmarking
  - Singles within 25%
  - Doubles within 3%
  - $3 * \sigma$  error bars

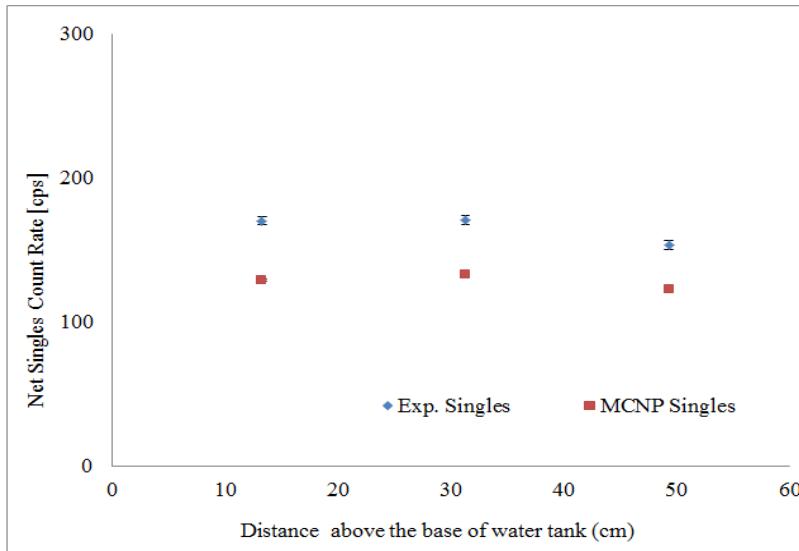


Figure 12: Three-point scan singles count rates

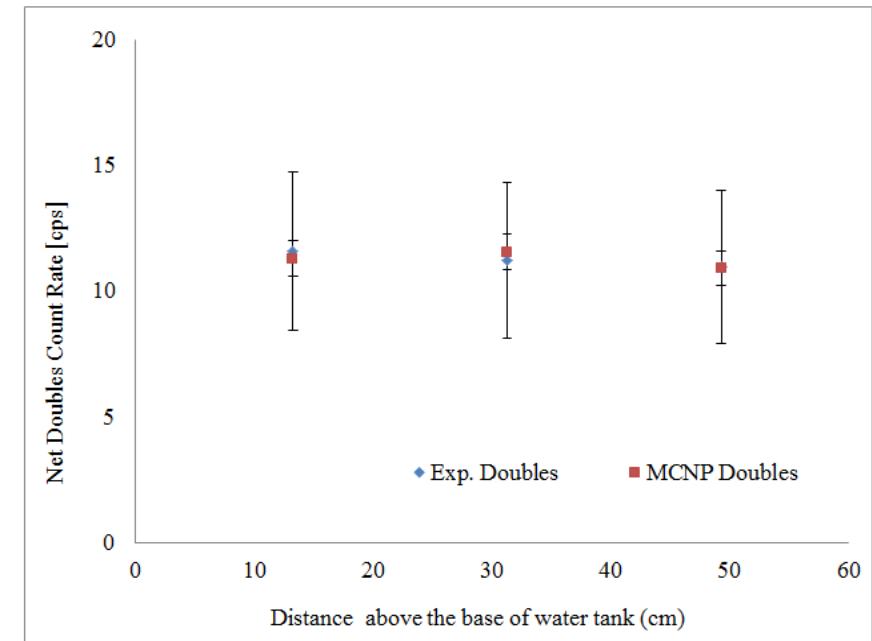


Figure 13: Three-point scan doubles count rates

# RESULTS AND ANALYSIS (CONTINUED)

- AECF Calibration
  - S and D count rates vs residual fissile mass
  - Linear fit: Exp. varying plates singles and MCNP varying plate singles
  - 2<sup>nd</sup> degree poly – concave down: MCNP varying enrichment singles
  - Linear fit: Exp. varying plate doubles and MCNP varying plates doubles
  - 2<sup>nd</sup> degree poly – concave down: MCNP varying enrichment doubles

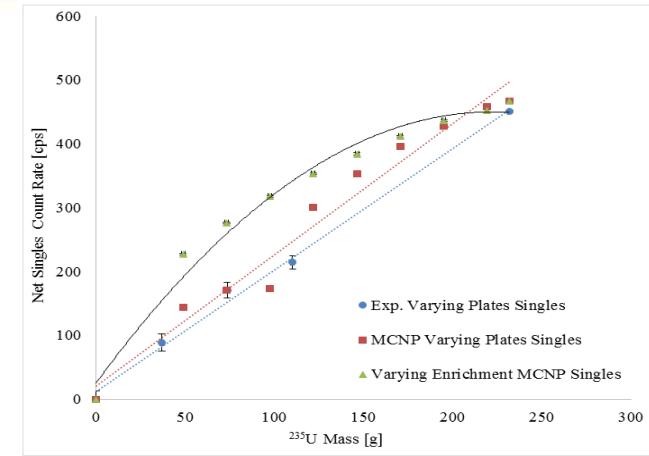


Figure 14: Singles count rate calibration

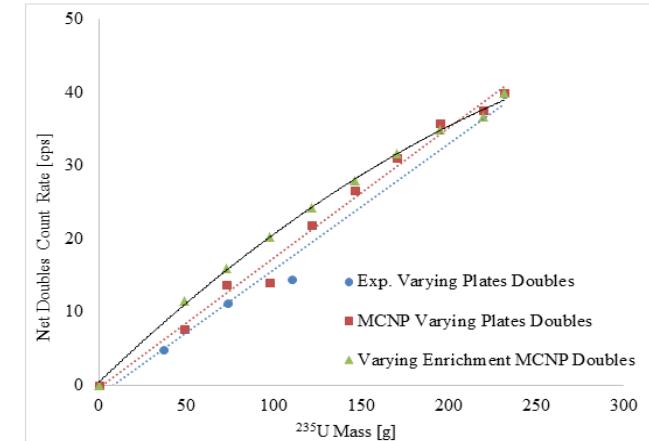


Figure 15: Doubles count rates calibration

# RESULTS AND ANALYSIS (CONTINUED)

- AECF Calibration
  - Singles, varying fuel plates Exp. and MCNP results, agree within 5%
  - Doubles within 20%
  - Higher count rates with varying enrichment in the middle
    - Higher  $^{238}\text{U}$  content
    - $^{235}\text{U}$  self shielding higher than  $^{238}\text{U}$

## RESULTS AND ANALYSIS (CONTINUED)

- AECF singles calibration with  $^{252}\text{Cf}$  vs AmLi- MCNP Results
  - Comparison of top, middle, bottom, and average singles
  - All agree within 5% with average
  - Homogenous distribution of  $^{235}\text{U}$  in fresh fuel
  - In burned fuel,  $^{235}\text{U}$  content is lowest in the middle compared to top and bottom
  - Helps distinguish fresh and used fuel
  - Normalized net singles: 31% higher with AmLi

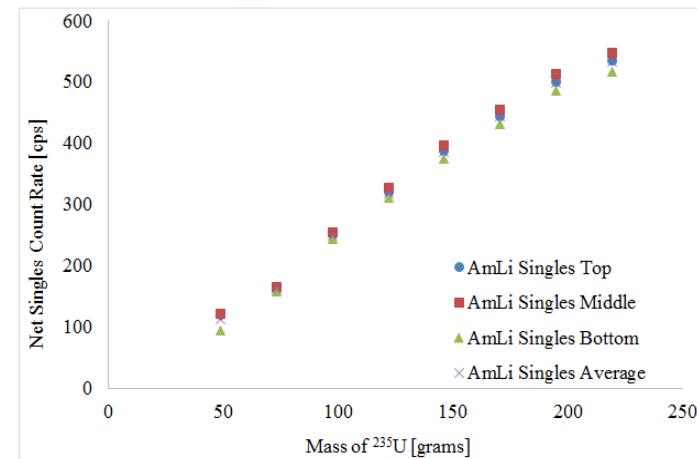


Figure 16: Singles calibration with AmLi source

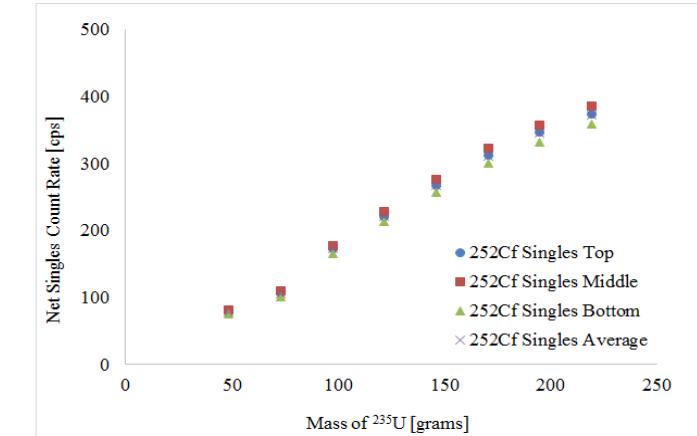


Figure 17: Singles calibration with  $^{252}\text{Cf}$  source

## RESULTS AND ANALYSIS (CONTINUED)

- AECF doubles calibration with  $^{252}\text{Cf}$  vs AmLi- MCNP Results
  - Comparison of top, middle, bottom, and average doubles
  - All agree within 4% with average
  - Normalized net doubles: 22 higher with  $^{252}\text{Cf}$

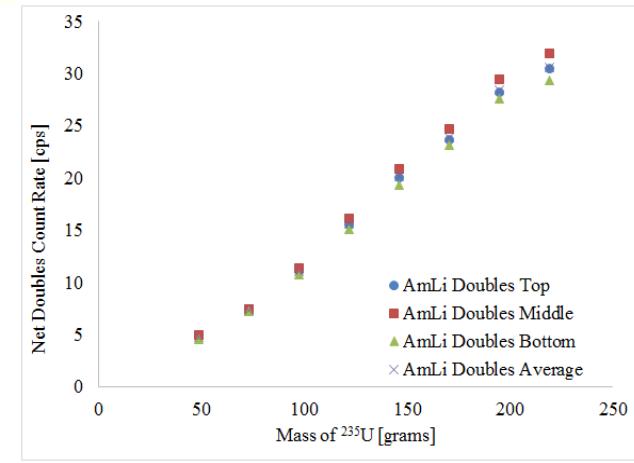


Figure 18: Doubles calibration with  $^{252}\text{Cf}$  source

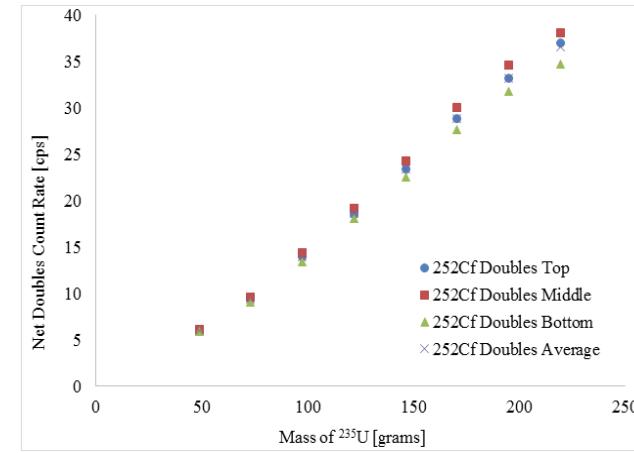


Figure 19: Doubles calibration with  $^{252}\text{Cf}$  source

# RESULTS AND ANALYSIS (CONTINUED)

	Singles	Doubles
Exp Original Configuration (FA)	450.71±3.41	39.78±1.34
Exp 90 deg rotated Configuration (FA)	427.35±1.95	38.38±0.69
Exp Original Configuration (PA)	170.54±1.025	11.21±0.334
Exp 90 deg rotated Configuration (PA)	154.07±1.621	10.99±0.424

- Comparison of count rates when geometry of fuel assembly is changed
  - FA: 5.18% fewer singles and 3.52% fewer doubles when rotated
  - Approximately  $7 * \sigma$  for singles and  $2 * \sigma$  for doubles
  - PA: 9.7% fewer singles and 1.96% fewer doubles when rotated
  - Approximately  $16 * \sigma$  for singles and  $1 * \sigma$  for doubles
  - More data is needed for confirmation
  - Bigger change in singles rate, while marginal change in doubles
  - The change needs to be accounted in verification measurements

# RESULTS AND ANALYSIS (CONTINUED)

- Experimental

	AmLi, N-165	$^{252}\text{Cf}$ , A7-866
<b>Singles IF/Source</b>	1.19E-02	9.70E-03
<b>Doubles IF/Source</b>	7.18E-04	8.36E-04

Source	Doubles IF/Singles IF
AmLi, N-165	6.03E-02
$^{252}\text{Cf}$ , A7-866	8.62E-02

- MCNP

	AmLi, N-165	$^{252}\text{Cf}$ , A7-866
<b>Singles IF/Source</b>	1.14E-02	9.88E-03
<b>Doubles IF/Source</b>	6.91E-04	1.01E-03

Source	Doubles IF/Singles IF
AmLi, N-165	6.02E-02
$^{252}\text{Cf}$ , A7-866	1.01E-01

## CONCLUSION

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- The singles and doubles rates in MCNP benchmarking agreed mostly within 5% and 4% respectively
- Calibration showed experimental and MCNP varying fuel plate singles agreed within 5% and doubles agree within 20%
- The varying enrichment curve showed higher count rates due to the lower self shielding of  $^{238}\text{U}$  compared to  $^{235}\text{U}$  and larges effects of water
- Singles and doubles of top, middle, and bottom measurements agree within 5% and 4% respectively with the average
- Measurements with 90 deg rotated assembly showed singles count rates as a function of geometry
- Normalized AmLi singles 1.23 times higher than singles from  $^{252}\text{Cf}$
- Normalized AmLi doubles 1.18 times lower than doubles from  $^{252}\text{Cf}$

## CONCLUSION (CONTINUED)

- Although  $^{252}\text{Cf}$  produced roughly 20% less singles than AmLi, there were more correlated neutrons within those single to result 17% higher doubles rates.
- The boost in the doubles count rates with  $^{252}\text{Cf}$  due to the boost in induced fission event due to the TCIF effect
- For future work, an experimental calibration of the AEFC with the fresh fuel containing higher percentage of  $^{238}\text{U}$  on average and varying  $^{235}\text{U}$  enrichment could be performed.
- The AEFC could be calibrated with fresh fuel rods containing various concentrations of  $\text{GdO}_3$  poison rods to simulate fission product absorption.

## REFERENCES

1. Menlove HO, et al., "Field Tests of the AEFC for Verification of Research Reactor Spent Fuel at the WWR-SM Reactor at the Institute of Nuclear Physics Uzbekistan." proc. INMM Annual Meeting; Orlando, Florida USA; 2012.
2. Menlove, Howard O, et al., "AEFC for the Verification of Research Reactor Spent Fuel – Field Experience to Date." Los Alamos National Laboratory, 16 Feb. 2017. Web. 14 Apr. 2017. <<http://permalink.lanl.gov/object/tr?what=info:lanl-repo/lareport/LA-UR-17-21223>>.
3. Menlove HO, et al., "The Optimization and Calibration of the AWCC Using 252Cf Interrogation and the Comparison with an AmLi Neutron Source." Los Alamos National Laboratory, USA, LA-UR-15-29620.
4. Menlove, HO, et al., "The Advanced Experimental Fuel Counter – a Portable Detector for the Verification of Research Reactor Spent Fuel." LA-UR- 11-01586, Los Alamos National Laboratory, USA, 04-05-2011.
5. Ensslin, N., "Principles of Neutron Coincidence Counting." Passive Nondestructive Assay Manual- PANDA. N.p.: Los Alamos National Laboratory, 2007, pages 457-491.
6. Trahan A C., "Utilization of the Differential Die-Away Self-Interrogation Technique for Characterization and Verification of Spent Nuclear Fuel." University of Michigan; Ann Arbor, Michigan USA; 2016.

# Questions ?