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Title: Measurement of Delayed Neutron Cross-Sections for Proton Interrogation at 800-MeV

Author(s): Randy J. Spaulding
Chris L. Morris
Steve J. Greene

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Ph. D. Candidacy Examination

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Form 836 (7/06)

MEASUREMENT OF DELAYED NEUTRON CROSS-SECTIONS FOR PROTON INTERROGATION AT 800-MEV

RANDY J. SPAULDING

PH.D CANDIDACY EXAMINATION
DECEMBER 7, 2010

UNIVERSITY OF NEW MEXICO
DEPT. OF PHYSICS AND ASTRONOMY
ALBUQUERQUE, NM

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Overview

- Motivation: Standoff Interrogation
- Beta-delayed neutron emitters
- Isotopic groups of emitters
- Surrogate method for scattered background subtraction
- Cross-Section data
  - 800-MeV (LANSCE)
  - 4-GeV (AGS)

Motivation

"Today, we are declaring that nuclear terrorism is one of the most challenging threats to international security. We also agreed that the most effective way to prevent terrorists and criminals from acquiring nuclear materials is through strong nuclear security — protecting nuclear materials and preventing nuclear smuggling."

-- President Barack Obama, April 8, 2010

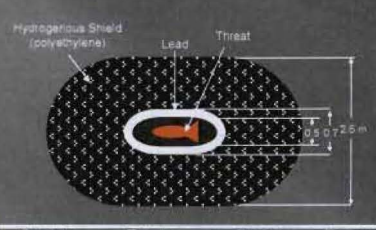
Maritime Nuclear Smuggling

- Millions of cargo containers enter the USA each year on enormous boxships
- Many smaller private craft have sufficient range and cargo capacity for SNM or a nuclear device

Proton Interrogation

- Goal
  - Reliable standoff (~500 m) interrogation of lightly shielded seaborne cargo from the air
    - Distance from populated areas enhances safety
- Requires
  - Compact low-power proton accelerator
  - Unique fissile material signature
- The signature
  - Intermediate energy protons (800 MeV-4GeV) produce a distinctive delayed neutron time signature (10s of seconds) from fissionable materials.

Active Interrogation: Why protons*?



Detection=10^6 fissions from a 10 10 cm target

Dose(p)=w_r 6.24 $10^9/(dE/dx)$
Dose(n,γ)=w_r 6.24 $10^9/[E/\lambda]$
w_r=10 for n; 1 for γ; 2 for p

Particle	Energy MeV	σ fission barn	Transmission	Dose MeV/cm²	Dose/detection Rad
n	14.1	2	1.74 E-04	1.57 E+04	25.148
γ	15	0.5	2.08 E-04	6.28 E+03	1.007
p	800	1	1.19 E-01	8.96 E+00	0.003

- Intermediate energy proton fission cross section is large
- Proton mean free path is long = large transmission
- Protons provide dose amplification in the target: 1 neutron/30MeV

*C. L. Morris, Fesseha Mariam and R. Schirato, *LANL*; John McGill, *GA*, *"Proton Interrogation", White paper presented to DNDO, 2005*

Potential Probes for Standoff Interrogation

Issue	Solution	Proton	Electron	Gamma	Neutron
produce DN signal	causes fission	✓✓	✓	✓✓	✓✓
shielded target	highly penetrating	✓✓	✗✗	✓	✓
directional production	accelerator primary	✓	✓	✗	✗
>100m standoff distance	charged particle focusing	✓	✓	✗	✗
high flux on target	low scatter/absorption	✓✓	✗✗	✗	✓
environmental safety	low dose per fission	✓✓	✗	✓	✗
signal-to-noise	low induced background	✗	✓	✓	✓

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Potential Probes for Standoff Interrogation

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environmental safety	low dose per fission	✓✓	✗	✓	✗
signal-to-noise	low induced background	✗	✓	✓	✓

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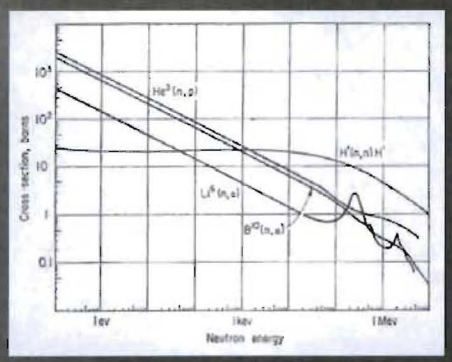
Neutron Detection



Neutron energy regimes
(approximate)

Neutron Energy, eV

Ultracold	Very Cold	Cold	Epithermal	Slow	Fast

Thermal = 0.025 eV

High-energy > ~14 MeV

Neutrons of interest

- Fission neutrons
  - Fission spectrum with tail above 2 MeV
- Delayed neutrons
  - Fissile elements <1 MeV
  - Fissionable elements <1 MeV
  - Lighter elements <2 MeV

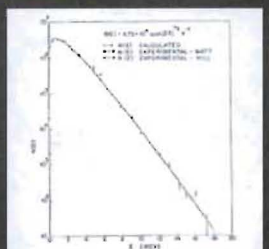


Fig. 4. Energy spectrum of fission neutrons. The calculated curve is based on the assumption of a Maxwellian distribution in the center-of-mass system for the neutrons emitted by a fission fragment.

Fission fragments comprise >100 DN emitters with half-lives ranging between $0.01 < t_{1/2} < 56s$

Approximate Half-life (sec)	Number of delayed neutrons per fission			
	^{235}U	^{238}U	^{239}Pu	Energy (MeV)
56	5.7 E-0	5.2 E-0	2.1 E-0	0.25
23	19.7	34.6	18.2	0.46
6.2	16.6	31	12.9	0.41
2.3	18.4	62.4	19.9	0.45
0.61	3.4	18.2	5.2	0.41
0.23	2.2	6.6	2.7	-
total delayed	0.0066	0.0158	0.0061	
total n/fission	2.5	2.43	2.9	
fraction delayed	0.0026	0.0065	0.002	

Spectroscopic S/N Enhancement

- Backgrounds of delayed neutrons--particularly ^{17}N--complicate detection of fissile materials
- Spectroscopic neutron detection at long delay times can provide a distinctive signature
  - In fissile targets delayed neutrons produce fission neutrons by inducing secondary fissions
    - k_{eff} amplification factors >5
  - 50% of fission neutrons are above 2 MeV
  - DN from ^{17}N are all below 2 MeV

Detector Selection

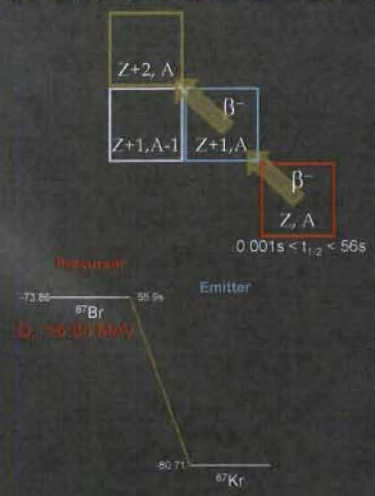
- ^{3}He (n, p) has large thermal cross-section and high Q-value
  - Gaseous proportional tubes:
    - Pros
      - Excellent discrimination → low background
      - Inexpensive to manufacture, reliable
    - Cons
      - Detection of fission-energy neutrons requires external moderation (HDPE) → energy information lost!
      - ^{3}He shortage raises cost

Detector Selection

- Liquid scintillators
  - Particle discrimination, good energy resolution ~5%
  - High background sensitivity makes them impractical for this application
- Light-Ion recoil provides energy information
  - H-recoil (plastic scintillator) very sensitive to background
  - Better alternative: high-pressure ^{4}He proportional counters
    - Provide particle discrimination for $E_n > 1$ MeV
    - Reasonable energy resolution (~15%) for $E_n < 10$ MeV

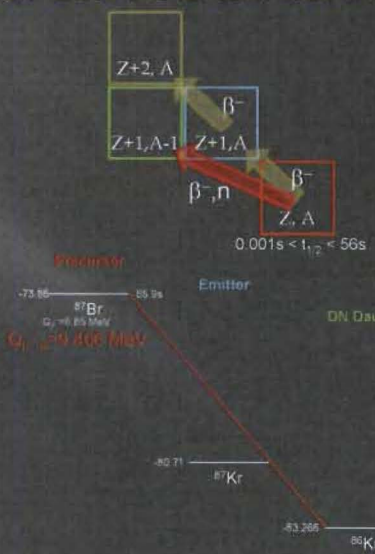
Beta-Delayed Neutron Production

- Isotopes that undergo delayed neutron emission following beta decay are characterized by a large difference in excess mass between the two isobars.
  - $Q_{\beta}^{-} > 5$ MeV (typ.)



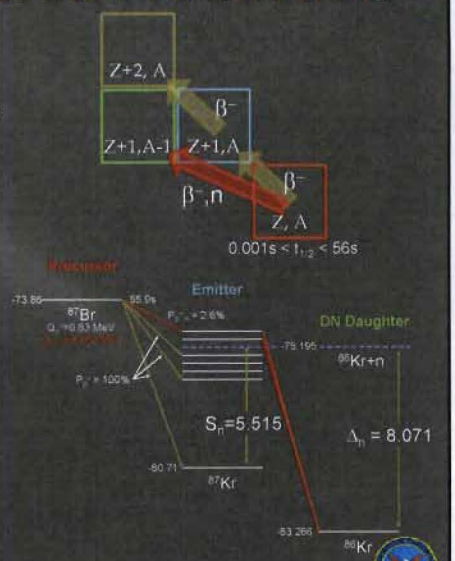
Beta-Delayed Neutron Production

- Isotopes that undergo delayed neutron emission following beta decay are characterized by a large difference in excess mass between the two isobars.
  - $Q_{\beta}^{-} > 5$ MeV (typ.)
  - $Q_{\beta^-,n} > S_n$ (8.071 MeV)



Beta-Delayed Neutron Production

- Isotopes that undergo delayed neutron emission following beta decay are characterized by a *large difference in excess mass* between the two isobars.
  - $Q_{\beta^-} > 5$ MeV (typ.)
  - $Q_{\beta^-} \approx \Delta_n$ (8.071 MeV)
- Lower-Z isobar (precursor) undergoes beta decay to an extremely short-lived excited state above the neutron binding energy of the emitter.



Delayed Neutron Precursors Appear in Clusters in Chart of Nuclides

Rb89	Rb90	Rb91	Rb92	Rb93	Rb94	Rb95	Rb96	Rb97	Rb98
Kr88	Kr89	Kr90	Kr91	Kr92	Kr93	Kr94	Kr95	Kr96	Kr97
Br87	Br88	Br89	Br90	Br91	Br92	Br93	Br94	Br95	Br96
Se86	Se87	Se88	Se89	Se90	Se91	Se92	Se93	Se94	
As85	As86	As87	As88	As89	As90	As91	As92		
Ge84	Ge85	Ge86	Ge87	Ge88					

Z

N

Clusters of Precursors Create Cascading Decay Chains

Rb89	Rb90	Rb91	Rb92	Rb93	Rb94	Rb95	Rb96	Rb97	Rb98
Kr88	Kr89	Kr90	Kr91	Kr92	Kr93	Kr94	Kr95	Kr96	Kr97
Br87 55.9 s	Br88	Br89	Br90	Br91	Br92	Br93	Br94	Br95	Br96
Se86	Se87 5.8 s	Se88 1.5 s	Se89	Se90	Se91	Se92	Se93	Se94	
As85	As86	As87 0.6 s	As88	As89	As90	As91	As92		
Ge84	Ge85	Ge86	Ge87	Ge88					

Z

N

Clusters of Precursors Create Cascading Decay Chains

Rb89	Rb90	Rb91	Rb92	Rb93	Rb94	Rb95	Rb96	Rb97	Rb98
Kr88	Kr89	Kr90	Kr91	Kr					
Br87 55.9 s	Br88	Br89	Br90	Br					
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As85	As86	As87 0.6 s	As88	As					
Ge84	Ge85	Ge86	Ge87	Ge88					

Z

N

Decay Chains are NOT Simple Exponential!

$$\frac{d}{dt}Se87 = -\lambda_{Se87} \cdot Se87(t)$$

$$\frac{d}{dt}Se88 = -\lambda_{Se88} \cdot Se88(t)$$

$$\frac{d}{dt}Br87 = -\lambda_{Br87} \cdot Br87(t) + \lambda_{Br87} \cdot \left(1-\beta_{Br87}\right) \cdot Se87(t) + \lambda_{Se88} \cdot \beta_{Se88} \cdot Se88(t)$$

where β_i = delayed neutron branching ratio

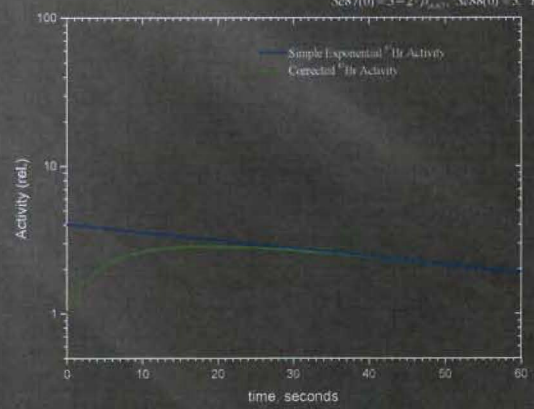
Decay of Beta Precursors is not Always Simple Exponential

$$\alpha_{Br87}(t) = \lambda_{Br87} \cdot \rho \cdot \left(K \cdot e^{-\lambda_{Br87} \cdot t} - \frac{Se87(0) \cdot \lambda_{Se87}}{\lambda_{Br87} - \lambda_{Se87}} \cdot (1 - \beta_{Se87}) \cdot e^{-\lambda_{Se87} \cdot t} - \frac{Se88(0) \cdot \lambda_{Se88}}{\lambda_{Br87} - \lambda_{Se88}} \cdot \beta_{Se88} \cdot e^{-\lambda_{Se88} \cdot t} \right)$$

where β_i = delayed neutron branching ratio

ρ = initial amount of each isotope created (appx. equal)

$Se87(0) = 3 - 2 \cdot \beta_{Se87}$, $Se88(0) = 3$, $K = BUNC$



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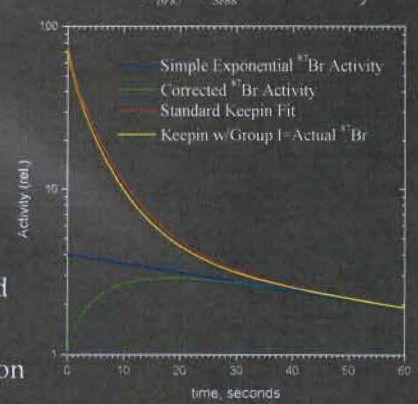
where β_i = delayed neutron branching ratio

ρ = initial amount of each isotope created (appx. equal)

$Se87(0) = 3 - 2 \cdot \beta_{Se87}$, $Se88(0) = 3$, $K = BUNC$

- Multigroup fits (e.g. Keepin) automatically correct for this (mostly)
- Small effect when comparing experimental data with calculated isotopic yields (5-20%)
- Likely negligible in this application

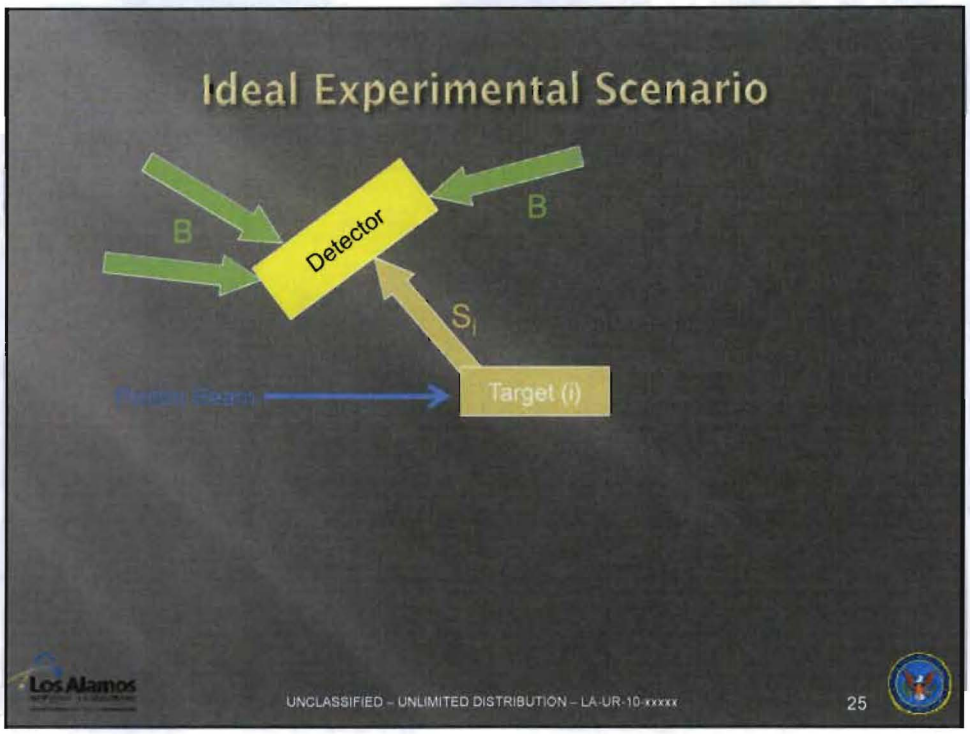


Decay Chains Can Be Quite Complex

Isotopic Target Group Definitions

- Grouped by atomic number based on half-lives of beta precursors that can be produced

Group	Atomic Number	Decay Characteristics Example Precursors
0	A < 8	No beta precursors
1	9 <= A < 17	Large cross-sections, very short half-life ^{9}Li (178 ms)
18	^{18}O	Enormous (p, 2p) cross-section ^{17}N (4.17 s)
2	17 <= A < 50	Cross-section drops with A ^{17}N, ^{22}O (2.25s), ^{16}C (747 ms), ^{18}N (624 ms), ^{19}N (271 ms)
3	50 <= A < 90	Cross-section and time signature fairly constant ^{51}Ca (10s), ^{48}K (6.8s), ^{49}K (1.26s), ^{50}K (472 ms), ^{44}Cl (560 ms)
4	90 <= A < 209	Cross-section increases slowly with A, small long-time tail ^{87}Br (55.65s), ^{137}I (24.5 s), ^{88}Br (16.29s), ^{85}As (2.02s), ^{91}Br (541 ms)
5	233 <= A	Large long-time tail, significant multiplication factor for fissiles


Ideal Experimental Scenario
B
B
Detector
S_i
Target (i)
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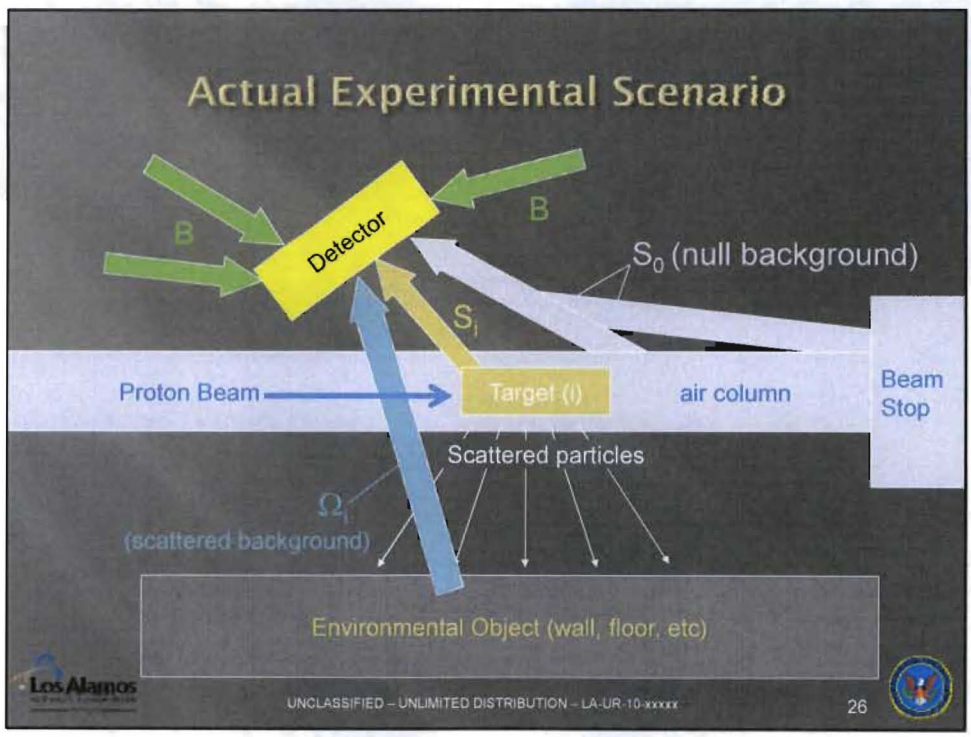
Actual Experimental Scenario
B
B
Detector
S_0 (null background)
S_i
Proton Beam
Target (i)
air column
Beam Stop
Scattered particles
Ω_i
(scattered background)
Environmental Object (wall, floor, etc)
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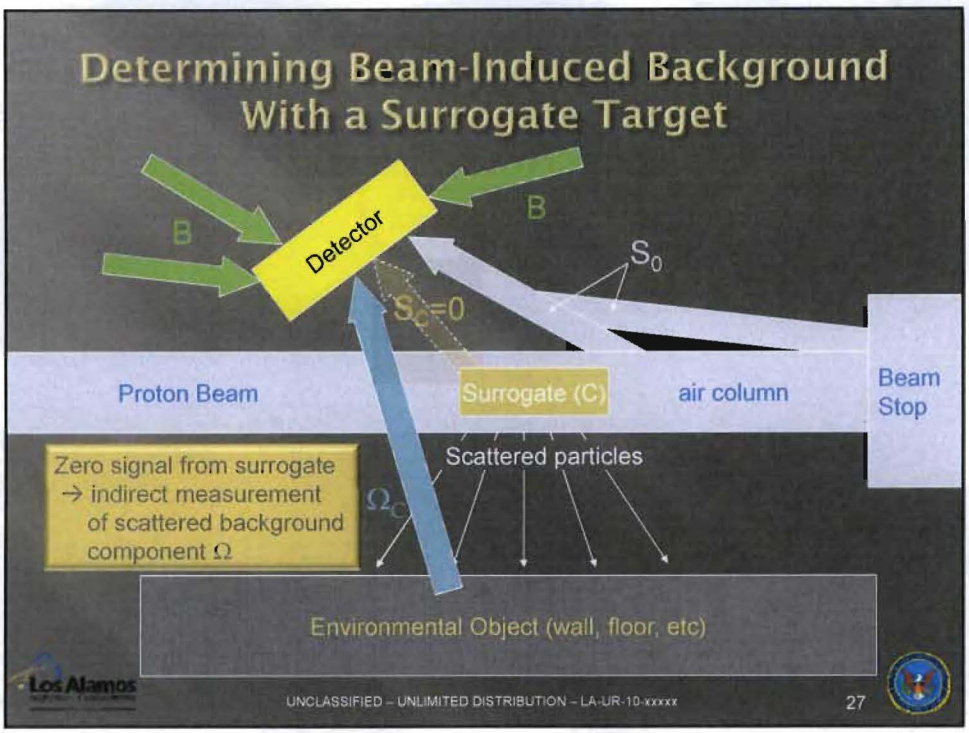

Parameters and Count Rates in Surrogate Experiments

- C_k = Count rate for in-beam targets
- S_k = Signal from various targets:
- B = Non-beam background count rate
- Ω_k = Scattered background component
- p_k = Protons in beam pulse
- κ_k = Fraction of protons passing through target
- ε = Detector efficiency*

*Assumed to be independent of target composition and time

$$S_k \equiv \frac{\sigma_k \lambda_k N_A}{A_k}$$

A_k = atomic mass
λ_k = mass thickness in g/cm^2

$$dC_k(t) = p_k \cdot \varepsilon(E) \cdot d^2 S_k(E,t) \cdot dE$$

$$\bar{\varepsilon}(t) \equiv \left[dS_k(t)\right]^{-1} \int \varepsilon(E) \cdot d^2 S_k(E,t)\, dE$$

$$dC_k(t) = p_k \cdot \bar{\varepsilon}(t) \cdot dS_k(t)$$

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$$\bar{\varepsilon}(t) \equiv \left[dS_k(t)\right]^{-1} \int \varepsilon(E) \cdot d^2 S_k(E,t)\, dE$$

$$dC_k(t) = p_k \cdot \bar{\varepsilon}(t) \cdot dS_k(t)$$

*Assumed to be independent of target composition and time

$$C_0 = \varepsilon \cdot \left(B + p_0 S_0\right)$$

$$C_C = \varepsilon \cdot \left\{B + p_C\left[S_0 + \kappa_C\left(S_C + \Omega_C\right)\right]\right\}$$

$$C_k = \varepsilon \cdot \left\{B + p_k\left[S_0 + \kappa_k\left(S_k + \Omega_k\right)\right]\right\}$$

Subscripts:
0 = Null target (air column + beam stop)
C = Surrogate target (Carbon)
k = Various interrogation targets

Parameters and Count Rates in Surrogate Experiments

- C_k = Count rate for in-beam targets
- S_k = Signal from various targets:
- B = Non-beam background count rate
- Ω_k = Scattered background component
- p_k = Protons in beam pulse
- κ_k = Fraction of protons passing through target
- ε = Detector efficiency*

$$S_k = \frac{\sigma_k \lambda_k N_k}{A_k}$$

$$dC_k(t) = p_k \cdot d\varepsilon(E) \cdot d^2 S_k(E,t)$$

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$$C_0 = \varepsilon \cdot \left(B + p_0 S_0\right)$$

$$C_C = \varepsilon \cdot \left\{B + p_C\left[S_0 + \kappa_C\left(S_C + \Omega_C\right)\right]\right\}$$

$$C_k = \varepsilon \cdot \left\{B + p_k\left[S_0 + \kappa_k\left(S_k + \Omega_k\right)\right]\right\}$$

Set surrogate signal to Zero and solve for scattered background

Surrogate Cross-Section Calculation

- Define $\eta_k \equiv \Omega_k / \Omega_C$ as the ratio of neutrons produced from beam scattered by the target divided by the neutrons produced from surrogate
  - Assumption of no delayed neutrons from surrogate valid for $t_D > 2s^*$ for Carbon

$$s_k = \frac{1}{\varepsilon \kappa_k}\left\{ c_k + \varepsilon B \gamma_k - \left(1 - \eta_k \frac{\kappa_k}{\kappa_C}\right) \cdot c_0 - \eta_k \frac{\kappa_k}{\kappa_C} \cdot c_C \right\}$$

where $\gamma_k = \frac{1}{p_0} - \frac{1}{p_k} + \eta_k \frac{\kappa_k}{\kappa_C}\left(\frac{1}{p_C} - \frac{1}{p_0}\right)$, $c_k \equiv C_k / p_k$,

*2 seconds is roughly 10 half-lives of ^{9}Li, the longest-lived precursor produced from a Carbon target

Benefits of the Surrogate Method

- Provides online measurement of scattered background component which is impossible to measure independently
- Eliminates enormous complexity involved with tracking all delayed neutron emitters in complex environment
- Flexibility: Practically any lower-group isotope can be used as a surrogate for a higher-group isotope
- η_k's fairly simple to calculate
  - Zeroth order: Interaction probability
  - Higher orders: Computer code, e.g. MCNPX
- True goal: demonstrate that backgrounds negligible
  - Signal independent of scattered background

Experimental Work

- Measure cross-sections spanning periodic table
  - Surrogates: Be, C
  - Structural, nonfissionable: O, Al, Fe, Sn
  - Heavy, slightly fissionable: W, Pb
  - Fissionable: U-238
  - Fissile: U-235
- Quantify detector energy response
  - Compare MCNPX models to experiment
- Validate surrogate method
  - CH2 versus pure C

Detectors

- Moderating He-3 detectors
  - Low He-3 pressure (0.3 bar) maximizes cost efficiency
  - Inexpensive, constructed at LANL

Single-tube detector "He3" or "Mark"

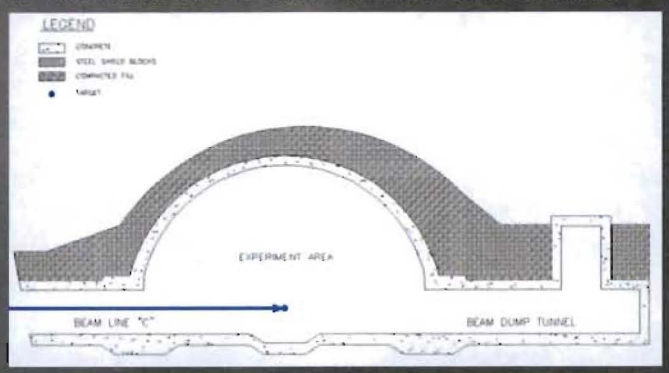
12"x2" ^{3}He in HDPE

Array of Neutron Detectors "ANDY"

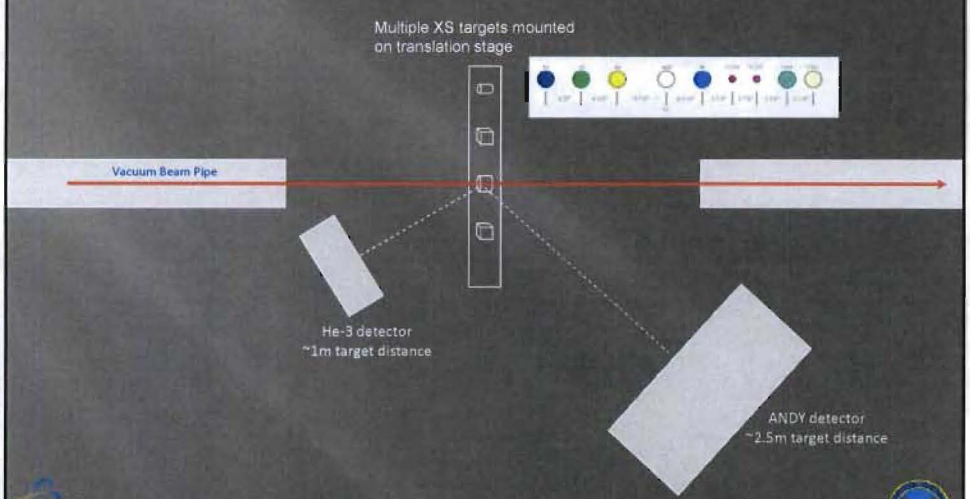
Differential moderation provides high efficiency plus coarse energy information

800-MeV Experiment @ LANL

- High-resolution spectrometer (HRS) dome at Area C at LANSCE
  - 46′-radius circular room and concrete dome



Experimental Setup for Cross Section Measurement



Conclusion and Future Work

- The proton-induced delayed neutron cross-section has been measured for several isotopes
- Signatures have been identified for fissionable materials.
  - Working on identifying fissionable vs. fissile.
- A framework is being developed to streamline benchmarking computer codes used to simulate these experiments
- Need to quantify variations in efficiency
- Need to validate surrogate method.
  - Construct DN decay curves from individual precursor yields?

Thanks to all who contributed to this work

- Randy Spaulding[1]
- Chris L. Morris[1]
- Steven J. Greene[1]
- Jeffrey D. Bacon[1]
- Gregory H. Canavan[1]
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- Mark F. Makela[1]
- Fesseha G. Mariam[1]
- Matthew M. Murray[1]
- Zhehui Wang[1]
- Laurie S. Waters[1]
- Fred J. Wysocki[1]
- Kris Kwiatkowski[1]
- Deborah Clark[1]
- Michael Brockwell[1]
- Kin Yip[2]
- Peter Vanier[2]

[1] Los Alamos National Laboratory
[2] Brookhaven National Laboratory