Detecting well-shielded nuclear material in cargo containers via active neutron interrogation

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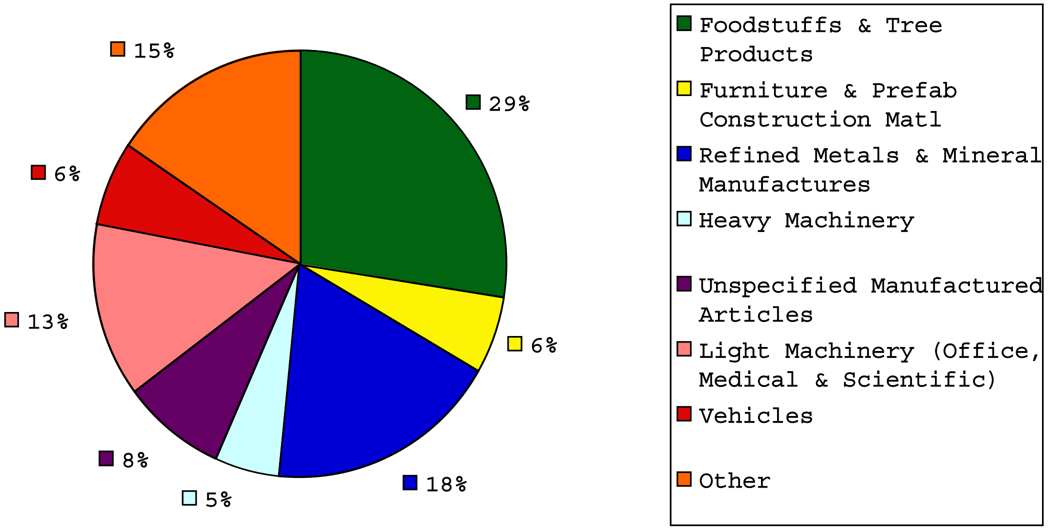
Potential danger at the world’s sea ports

- 90% of the world’s trade moves via sea-going containers
- Cargo is attractive for smuggling illicit material
    - Large volume and mass of material in each container
    - Cargo is non-homogeneous
- Volume of traffic is enormous
    - More than 6,000,000 containers enter the U.S. annually
    - U.S. west coast ports are processing 11,000/day— An average of 8/min on a 24/7 basis
- Successful delivery of one weapon of mass destruction in a container can be catastrophic

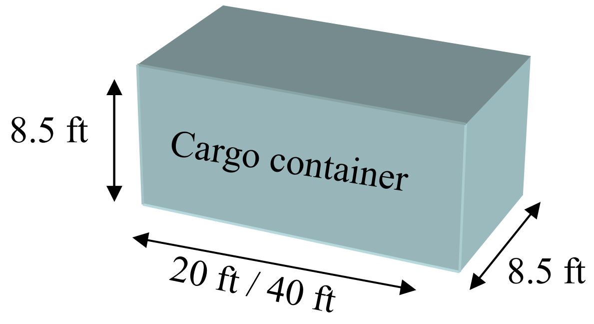
The Port of Oakland
San Francisco Bay, California

Top 10 foreign ports of origin			Top 10 domestic ports of entry		
Port of origin	Outbound to U.S.	% of total traffic	Port of entry	U.S. arrivals	% of total traffic
Hong Kong	558,600	9.8	Los Angeles	1,774,000	24.7
Shanghai	330,600	5.8	Long Beach	1,371,000	19.1
Singapore	330,600	5.8	NY / New Jersey	1,044,000	14.6
Kaohsiung	319,200	5.6	Charleston	376,000	5.2
Rotterdam	290,700	5.1	Savannah	312,000	4.3
Puson	285,000	5.0	Norfolk	306,000	4.3
Bremerhaven	256,500	4.5	Seattle	284,000	4.0
Tokyo	159,600	2.8	Tacoma	273,000	3.8
Genoa	119,700	2.1	Oakland	268,000	3.7
Yantian	114.000	2.0	Houston	233,000	3.3
Top 10 total	2,764,500	48.5	Top 10 total	6,241,000	87.0

The cargo is the challenge



- Cargo material is diverse
- Containers are very large
- Packing is inhomogeneous
- Need a reliable scan
- t_{scan} < 1 min / container



Scope of the project

- Concentrate on the threat with the gravest consequences—nuclear explosives
  - Uranium and plutonium with very high isotopic content of the nuclides ^{235}U and ^{239}Pu
  - Heavily shielded material

- Develop a prototype detection system for use at sea ports
  - Functions for a **range of material density:** $0 < \rho L < 150$ g/cm^2
  - Is **reliable:** False positive and false negative rates $< 10^{-3}$
  - Preserves the **flow of commerce** through the port:
    $t_{scan} < 1$ min / container

We need a useful signature unique to fissionable material

- Radiation must penetrate from deep within a cargo container to reach a detector outside and must be intense enough to be discriminated from background

- ^{235}U and ^{239}Pu are both radioactive and have unique gamma radiation signatures. Can we exploit these passive emissions?
  - ^{239}Pu ($t_{1/2} = 2.4 \times 10^4$ yr) emits weak gamma rays and neutrons
  - ^{235}U ($t_{1/2} = 7.0 \times 10^8$ yr) emits weak, low-energy gamma rays

- Active methods inject particles into container to produce fission reactions in fissile material and provide unique return signals

- We don’t expect to rely exclusively on active approaches
  - Passive radiation detection
  - Radiography to locate high-density components buried within an otherwise low-density cargo

Active interrogation

“Prompt”

$^{235}U(n,\gamma)^{236}U$

Detect capture γ-rays

Problem:

mass(U or Pu) < 10 kg
mass (other cargo) = 10,000 kg
S/N is very small
and need high energy resolution
detectors to identify U or Pu

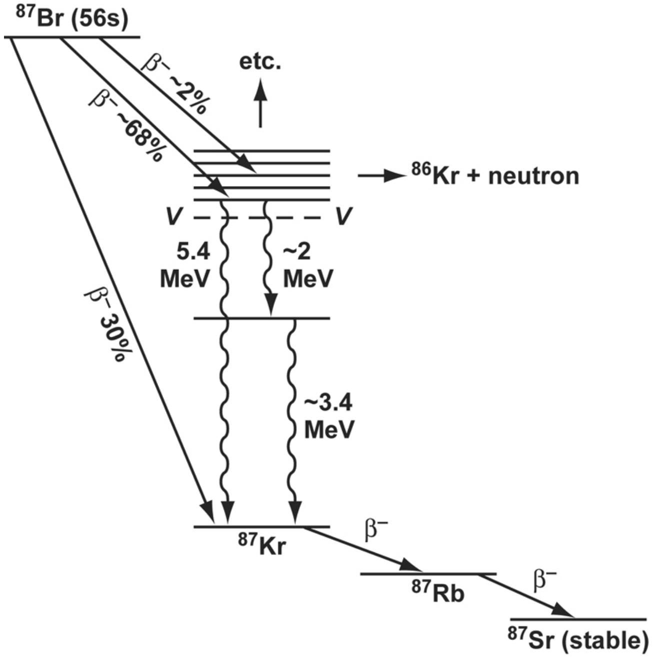
A word about the fission reaction and β-delayed gamma rays and neutrons

- Thermal-neutron induced fission reaction produces two fission fragments and zero to many neutrons. For example:

$n + {}^{235}U \rightarrow {}^{236}U^* \rightarrow {}^{90}Kr + {}^{143}Ba + 3n$

β-decay of the fission fragments frequently leaves the daughter nucleus in an excited state

- Sometimes above the binding energy of the last neutron => neutron emission
- More often to a high-energy state that de-excites by high-energy γ-ray emission
- γ-ray emission is 10 times more likely
- Both processes are fission signatures



Delayed n or γ?

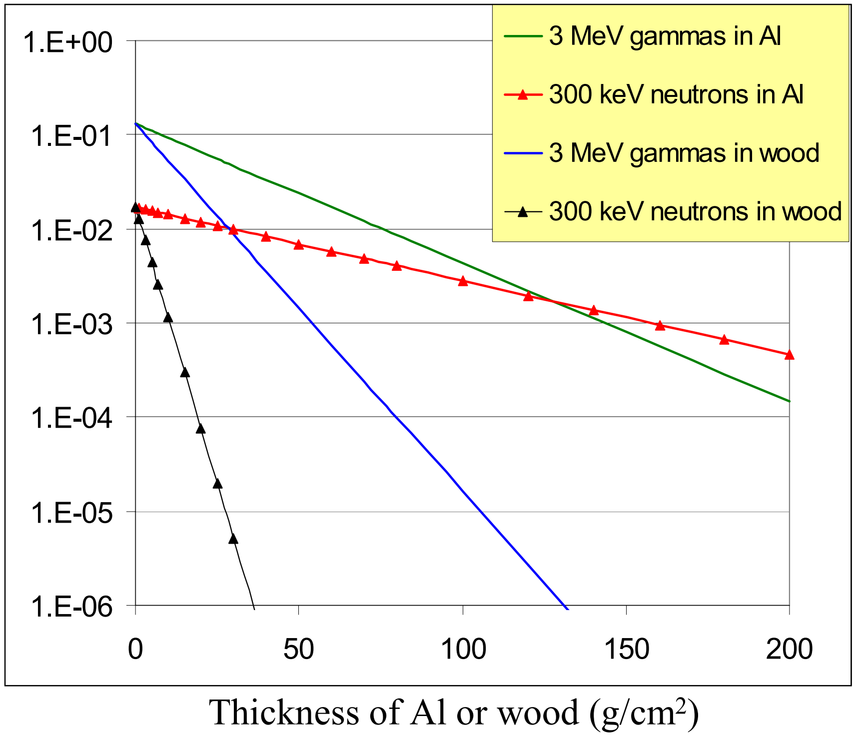
Yield / Fission

Delayed γ-ray yields are approx. one order of magnitude higher than delayed neutron yields

Yield /fission	^{235}U thermal fission	^{239}Pu thermal fission	^{238}U fast fission
Delayed neutrons [1]	**0.015**	**0.0061**	**0.044**
γ-rays at $E_\gamma > 3$ MeV [2]	**0.127**	**0.065**	**0.11**
γ-rays at $E_\gamma > 4$ MeV [2]	**0.046**	**0.017**	**0.03**

Attenuation [3]

Delayed neutrons are highly attenuated in hydrogenous material (estimate includes yield / fission)



The high energy γ-ray signal leaving thick hydrogenous cargo may be as much as 10^2 to 10^4 larger than the delayed-n flux.

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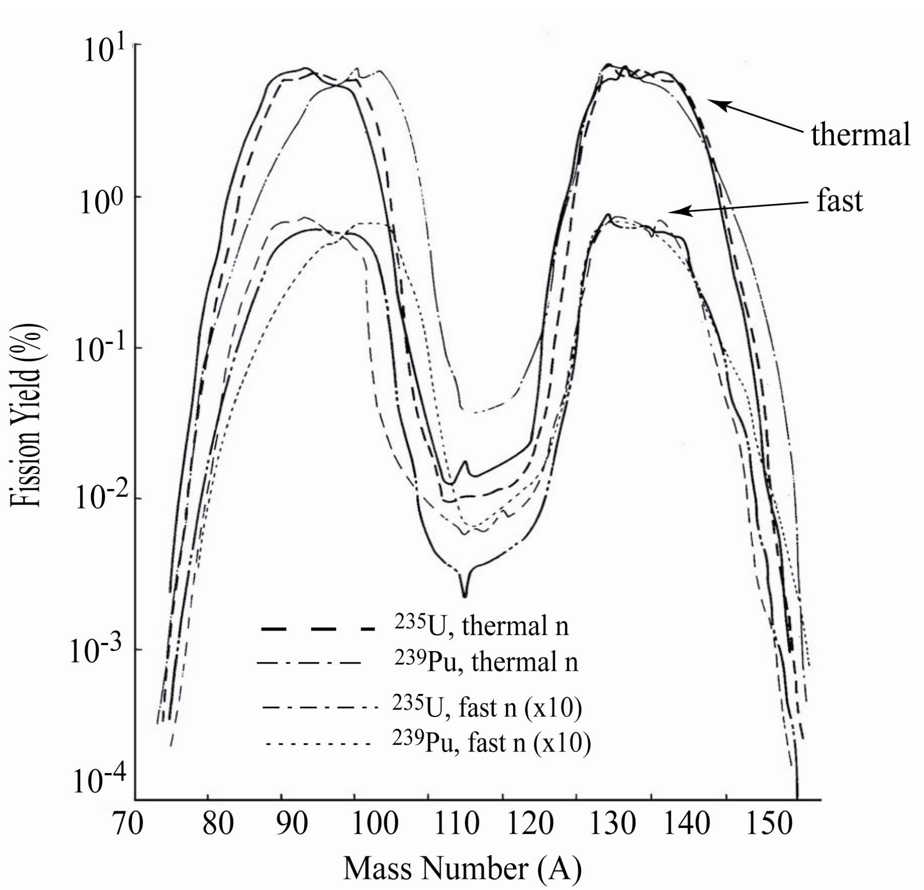
[1] LLNL Nuclear Data Group, 2003, http://nuclear.llnl.gov/CNP/nads/

[2] LBNL Isotope Explorer, 2003, http://ie.lbl.gov/ensdf/

[3] T. Rockwell III, Reactor Shielding Design Manual, D. Van Nostrand Co., New York (1956).

Neutron-induced fission-fragment mass distributions [1]



[1] www.kayelaby.npl.co.uk, T.R. England and B.F. Rider, (1992) OECD Report, NEA/NSC/DOC(92) p. 346

Can we use this signature to distinguish between ^{235}U and ^{239}Pu?

- Gamma-ray yield ratios
- Decay curves

High-energy gamma-ray yields in ^{235}U thermal neutron fission

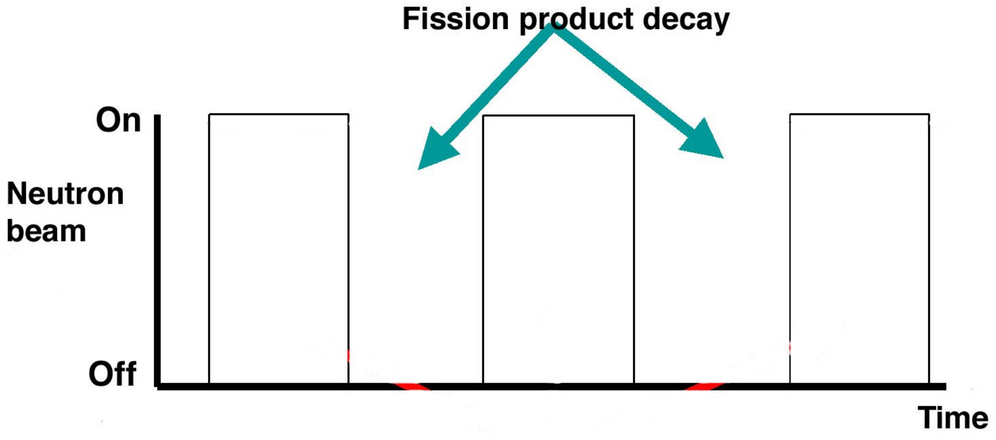
Nuclide	Half-life (sec)	> 4 MeV gammas per fission	> 3 MeV gammas per fission
^{85}Se	39.	0.0	0.0012
^{86}Br	55.	0.0013	0.0013
^{87}Br	55.	0.0045	0.0073
^{88}Br	16.	0.0045	0.0072
^{89}Br	4.4	0.0016	0.0021
^{89}Kr	189.	0.00064	0.0029
$^{90\text{-}m}Rb$	258.	0.00063	0.0036
^{90}Rb	156.	0.0089	0.016
^{91}Kr	8.6	0.000047	0.0020
^{91}Rb	58.	0.0052	0.017
^{92}Rb	4.5	0.011	0.012
^{93}Rb	5.9	0.00078	0.0073
^{94}Rb	2.7	0.00022	0.0015
^{95}Rb	0.38	0.000027	0.0011
^{95}Sr	25.	0.00052	0.0031
^{97}Y	3.8	0.0	0.017
$^{98\text{-}m}Y$	0.59	0.003	0.007
^{136}Te	17.5	0.0	0.0020
^{136}I	83.	0.0005	0.0011
^{138}I	6.5	0.00043	0.0010
^{140}Cs	63.	0.0	0.0038
^{141}Cs	25.	0.0	0.0017
^{142}Cs	1.8	0.00054	0.0014
Total, including activities not shown	Varying	0.0458	0.127

High-energy gamma-ray yields in ^{239}Pu thermal fission

Nuclide	**Half-life(sec)**	**>4 MeV gammas/fission**	**>3 MeV gammas/fission**
^{87}Br	55.	0.0015	0.0025
^{88}Br	16.	0.0013	0.0020
$^{90\text{-}m}$Rb	258.	0.00038	0.0021
^{90}Rb	156.	0.0025	0.0046
^{91}Rb	58.	0.0020	0.0063
^{92}Rb	4.5	0.0045	0.0049
^{93}Rb	5.9	0.00031	0.0029
^{95}Sr	25.	0.0003	0.0017
^{97}Y	3.8	0.0	0.013
^{98}Y	0.59	0.0024	0.0055
^{106}Tc	36.	0.0	0.0066
^{140}Cs	64.	0.0	0.0026
^{141}Cs	25.	0.0	0.0014
^{142}Cs	1.8	0.00037	0.0022
Total including activities not shown	Varying	0.017	0.065

High-energy γ-rays detected between neutron pulses are used to identify fissile material

- Fission product γ-rays integrated from 3 to 7 MeV between interrogation beam pulses are used to identify the presence of fissionable material
  - Distinguished from activation and background sources by their high energies ($E_\gamma > 3$ MeV)
  - And their characteristic decay times
- There is expected to be some γ-radiation between beam pulses due to activation of cargo
  - That radiation is expected to be low energy (< 2.5 MeV)
  - And mostly characterized by longer half-lives (typically $>> 1$ min)
- Detailed experimental evaluation of these assumptions and interferences is being conducted with real cargos to qualify this methodology



A combined solution

Arrival at port

Document screening

Passive screening

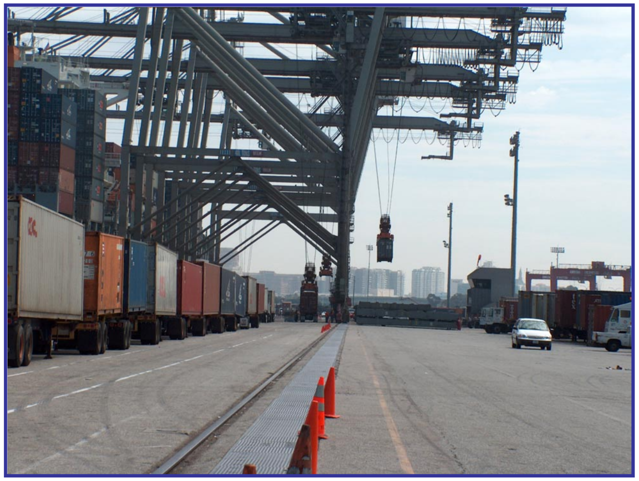
Radiography screening

Active interrogation

Unload container

Response

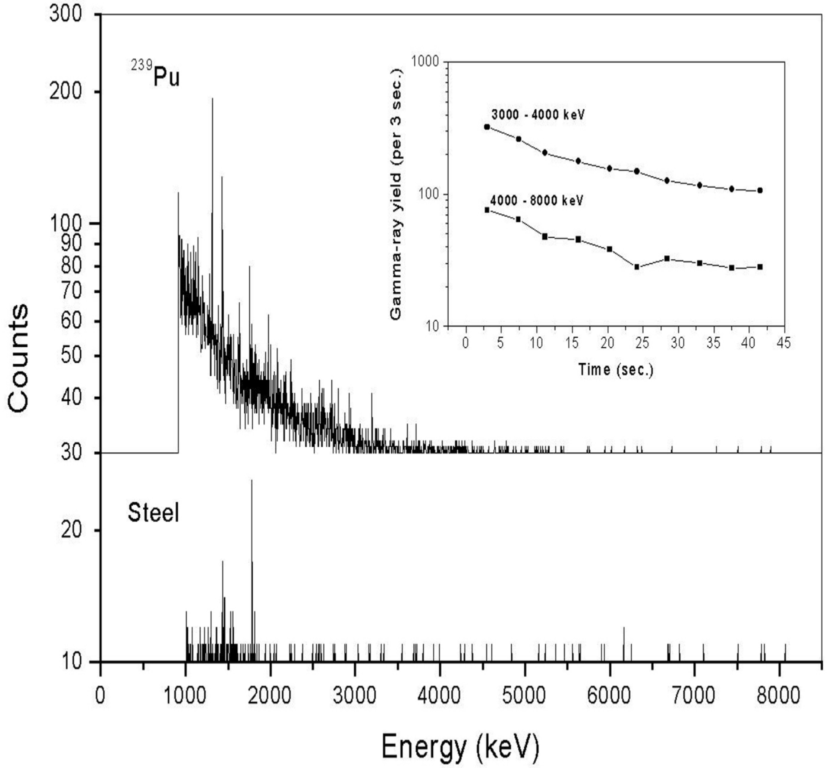
Cleared for delivery



β-delayed γ-rays above 3 MeV attributable to U, Pu

Experiment by Norman *et al.* 2004 [1]

- E_n = thermal
- Separate neutron irradiations of ^{235}U (93%), ^{239}Pu (95%), wood, polyethylene, aluminum, sandstone, and steel.
- Cycles of 30 s irradiation and 30 s counting.
- 10 sequential 3-second γ-ray spectra were acquired with a single coaxial 80% HPGe detector.



→

^{235}U(n_{th},f) and ^{239}Pu(n_{th},f):

Significant γ-ray intensity above 3 MeV.

Short effective half-life (approximately 25 s).

[1] **E. B. Norman** *et al.,* **NIMA 521 (2004) 608-610.**

[2] **E. B. Norman** *et al.,* **NIMA 534 (2004) 577.**

Fission Yields

Ratios of γ-ray intensity in HPGe (lines) and plastic (wide energy bins)

HPGe: Fission Product γ-ray line ratios

Target	^{95}Y 10.3 m	^{89}Rb 15.4 m	^{138}Cs 32.2 m	^{106}Tc 36 s
^{235}U	6.38%	4.72%	6.71%	0.40%
^{239}Pu	4.69%	1.71%	5.92%	4.03%

Plastic: Fission Product γ-ray bin ratios

Energy Bin (MeV)	0.5 – 1.5	1.5 - 2.5	2.5 - 3.5	3.5 - 4.5	4.5 - 5.5
^{235}U	324	58	16	4.6	1.00
^{239}Pu	565	105	24	5.3	1.00

$$\frac{^{235}U}{^{239}Pu} = \frac{I_\gamma^U(1.5-2.5) \Big/ I_\gamma^U(4.5-5.5)}{I_\gamma^{Pu}(1.5-2.5) \Big/ I_\gamma^{Pu}(4.5-5.5)} = 1.81$$

Cargo experiments with nat-U and E_n = 14 MeV

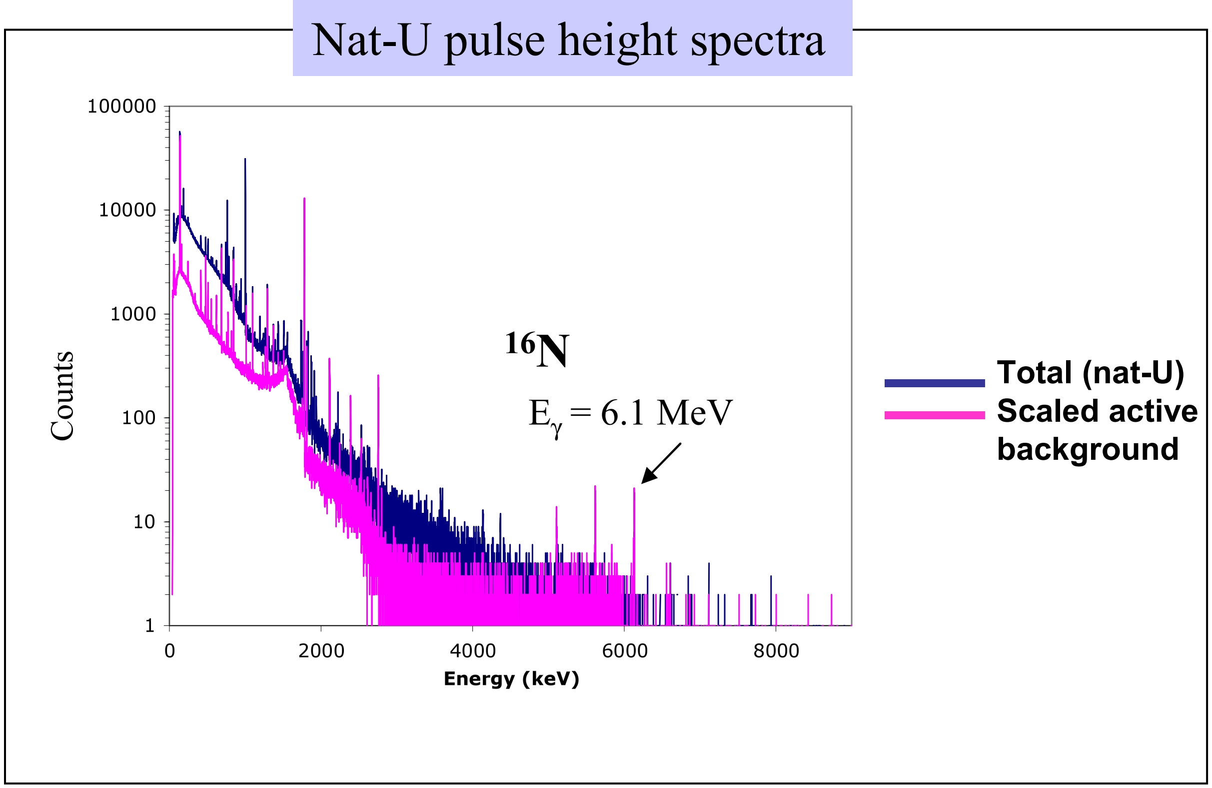
Target:

22 kg nat-U (150 g 235U)
cylinder within poly beads
3 m to generator
1.5 m to detector

Irradiation:

E_n = 14 MeV
10- 30 s irradiations
30 s count cycles
$Y_n = 2 \times 10^{10}$ n / s initial
$\Phi_n = 2 \times 10^{4}$ n / cm^2 / s at target

Background interference for $E_\gamma > 3$ MeV?



50% HPGe spectra after irradiation with 14 MeV neutrons, with and without the 22 kg nat-U target.

$^{16}O(n,p)^{16}N$:

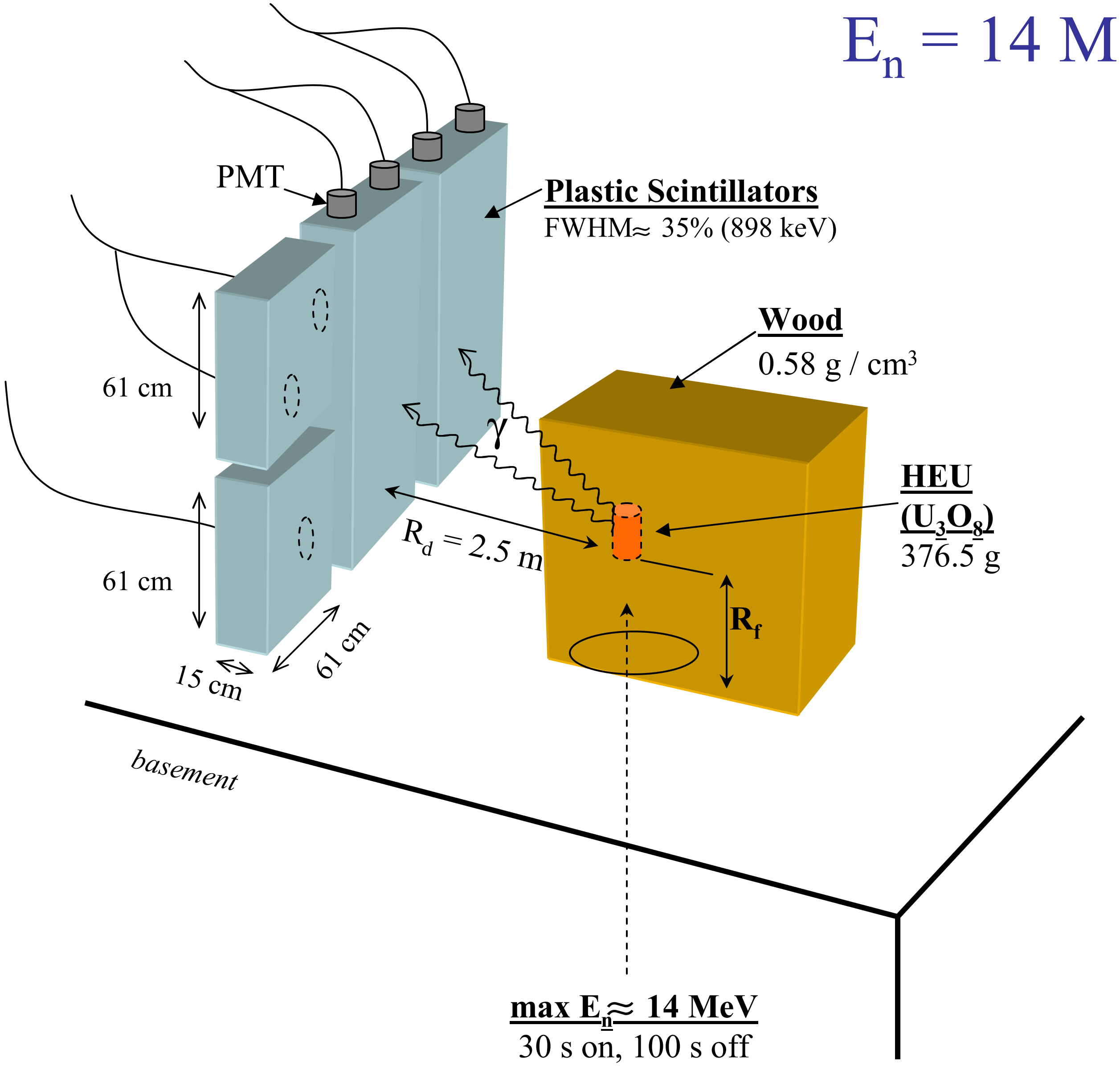
Threshold = 10.24 MeV

Q = -9.63 MeV

^{16}N $E_\gamma = 6.1$ MeV

^{16}N $t_{1/2} = 7.1$ s

Cargo experiments with HEU and $E_n = 14$ MeV



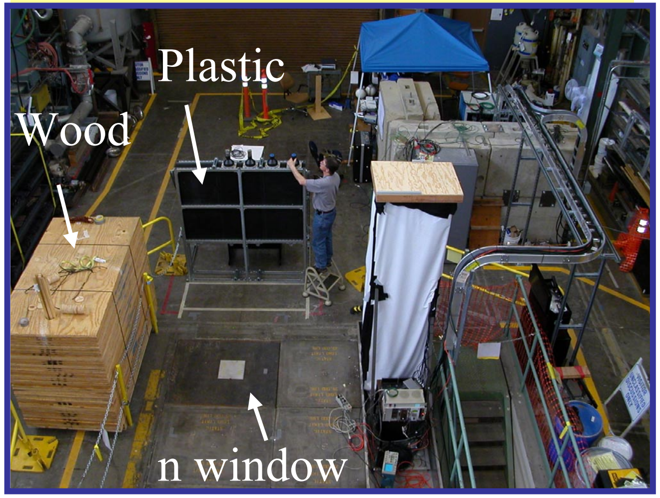
HEU embedded in plywood

R_f = 61 cm (40 g / cm^2 wood)

R_d = 2.5 m (60 g / cm^2 wood)

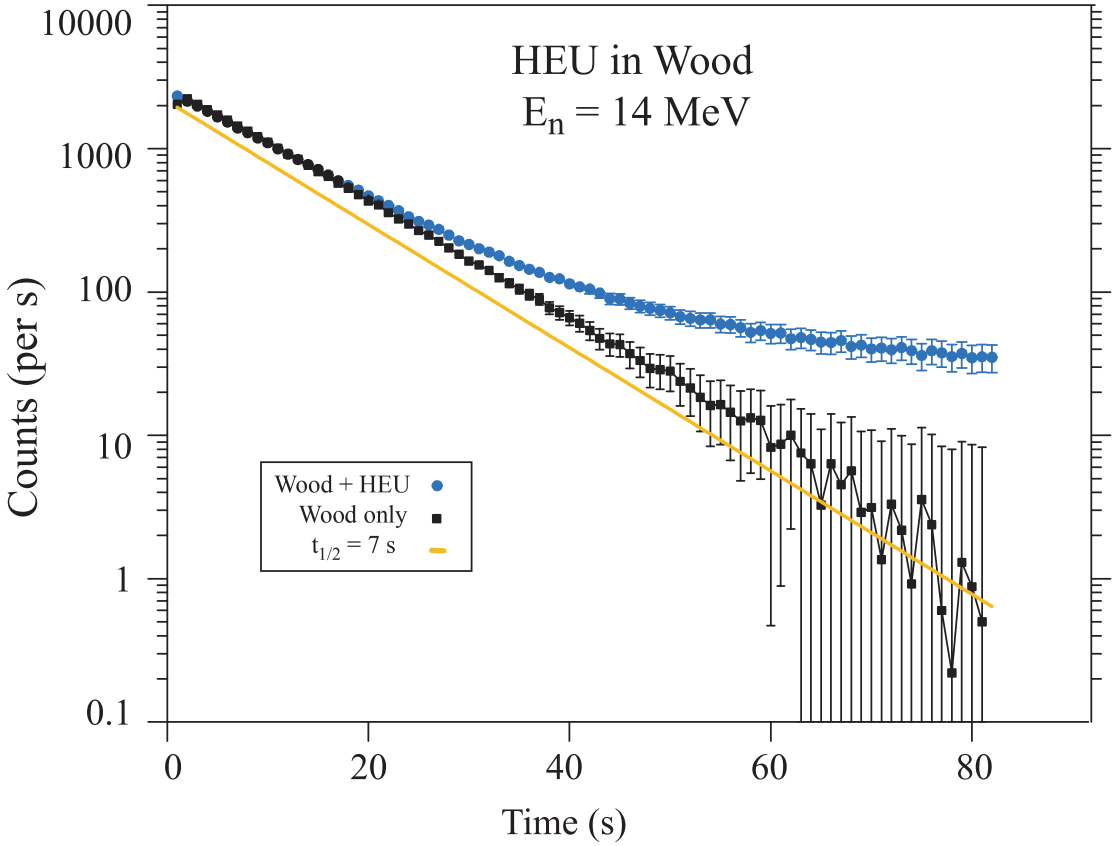
$Y_n \sim 6 \times 10^{10}$ n/s initial

$\Phi_n \sim 6 \times 10^4$ n/s/cm^2 at target



Decay curves show fission + ^{16}N contamination

3 MeV < E_γ < 4 MeV



$^{16}O(n,p)^{16}N$:

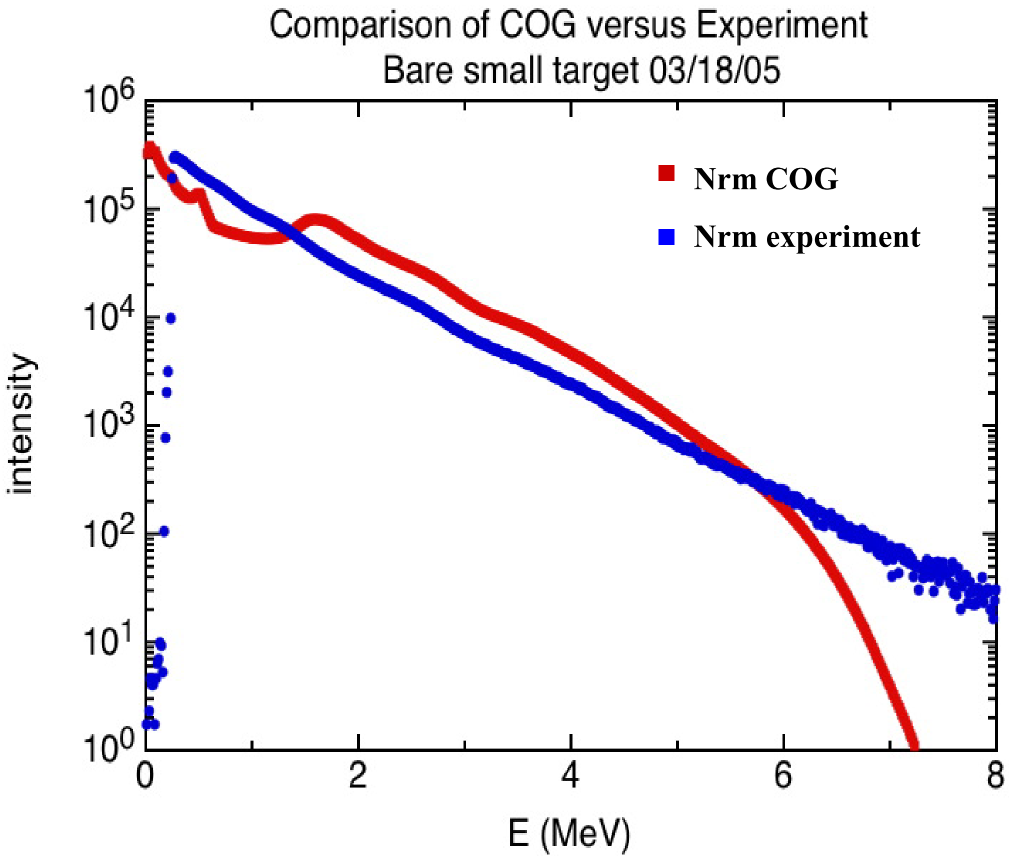
E_γ = 6.1 MeV

$t_{1/2}$ = 7.1 s

- E_n = 14 Mev
- 1 plastic detector
- 376.5 g HEU (U_3O_8)
- 50 irradiation cycles
- 3 MeV < E_γ < 4 MeV

→ We need E_n < 10.24 MeV!

Simulation vs. Experiment



Experiment:

Target: 276 g HEU (U_3O_8)
no cargo

Irradiation: E_n = 14 MeV
10- 30 s irradiations
100 s count cycles

Detector: 2’ x 4’ x 6” plastic

Simulation:

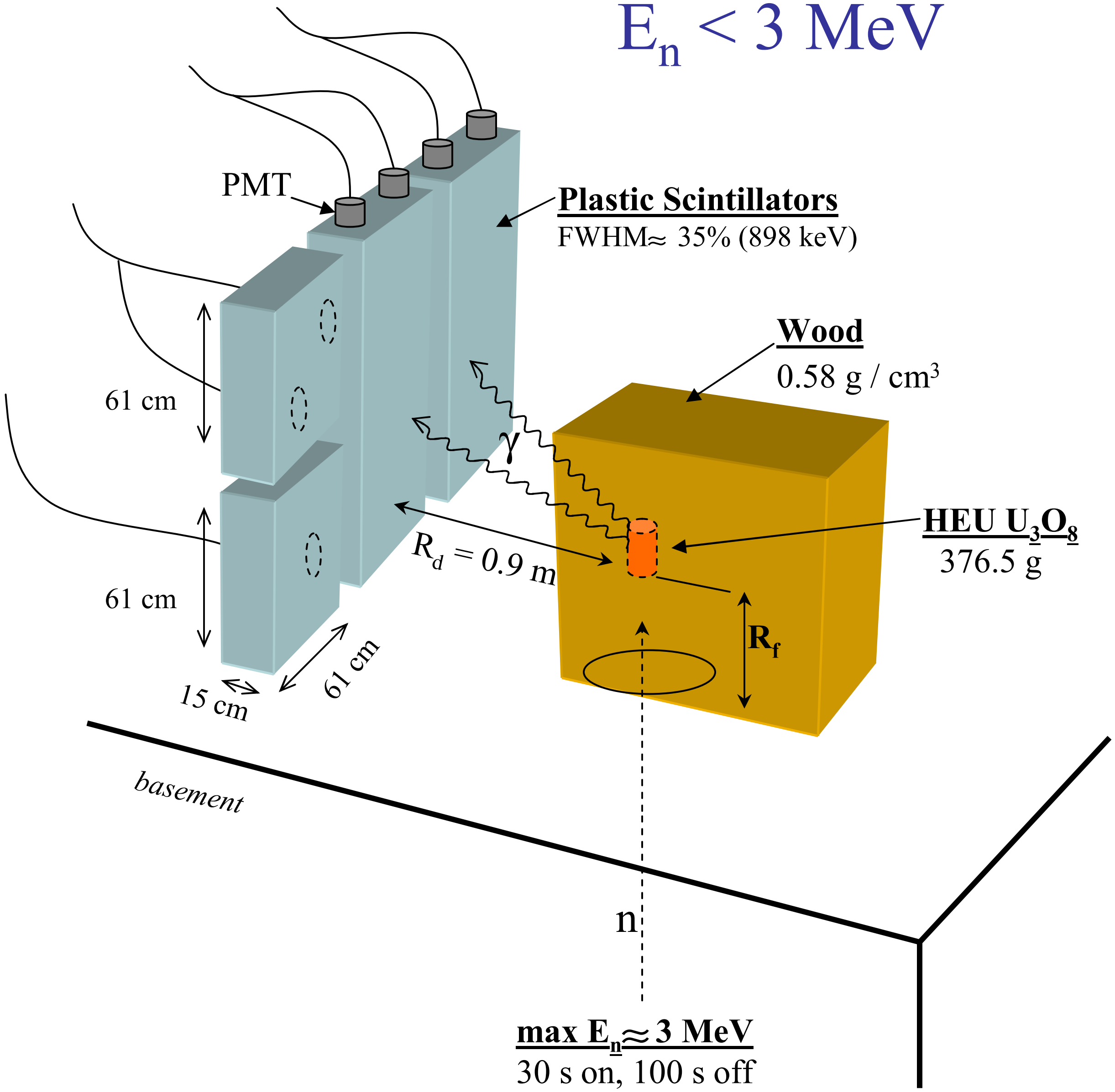
COG: Response function taken from 2’ x 2’ x 6” centrally-viewed detector as a simplified estimate

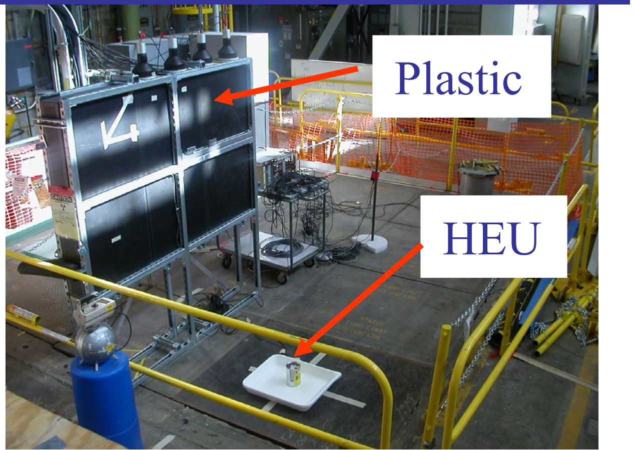
We now have $E_n < 10$ MeV with improved intensity

- RFQ d(d,n) generator from Accsys Technology
- High Energy Beam Transport delivered by LBNL
- $E_d = 4$ MeV at 100 μA
- Expect $E_n = 3$ to 7 MeV, $\Phi \sim 1\text{x}10^6$ n/cm^2/sec
  - Flat energy spectrum in this range
  - Forward-peaked angular distribution
- Deuterium gas target (installed 9/14/05)
  - Up to 15x higher flux as compared to a sealed target
  - Cross sections for d(d,n) rise rapidly with deuteron energy up to a maximum at $E_d \sim 5$ MeV

- Test experiment (9/1/05)
  - ^{12}C solid target
  - $E_d = 3.7$ MeV
  - E_n approx 1.5 to 3 MeV
  - Do we see Fission?   Background interferences?

We measured n + HEU
$E_n < 3$ MeV





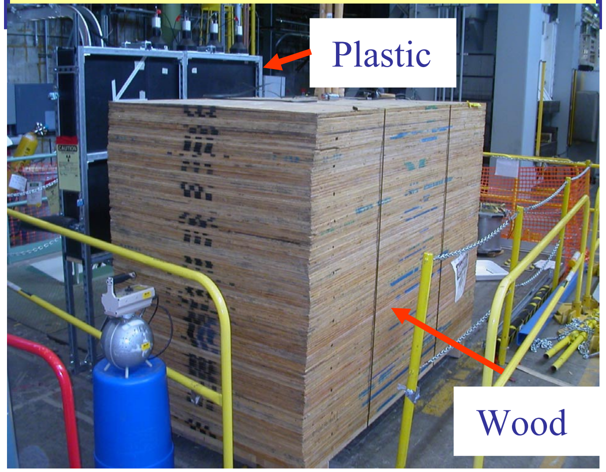
HEU + No Cargo

HEU + Wood

- $R_f = 1$ ft
- $R_f = 2$ ft
- $R_f = 3$ ft

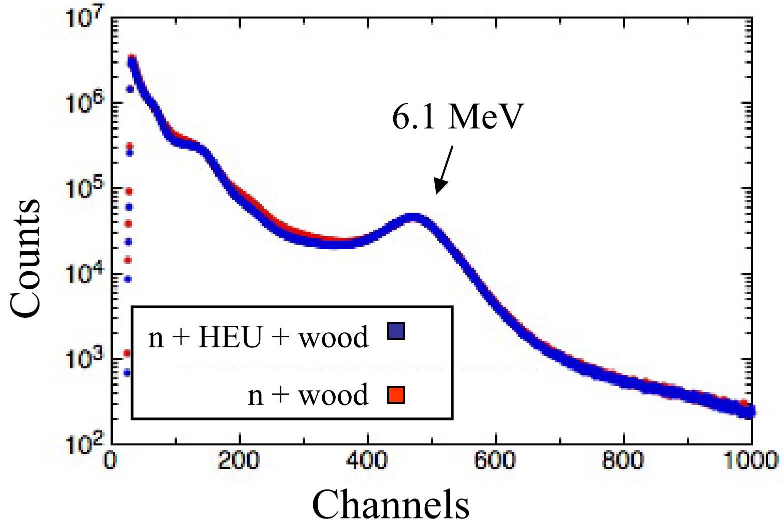
Teflon +No Cargo

($^{19}F(n,\alpha)^{16}N$)



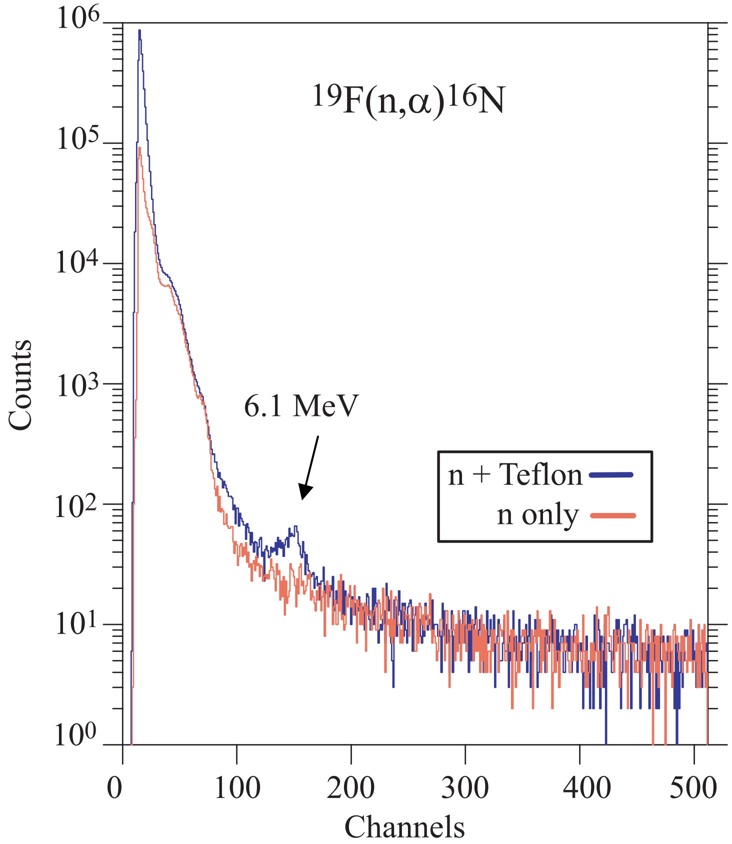
For $E_n < 3$ MeV, ^{16}N interference disappears

$E_n = 14$ MeV, 50 irradiations



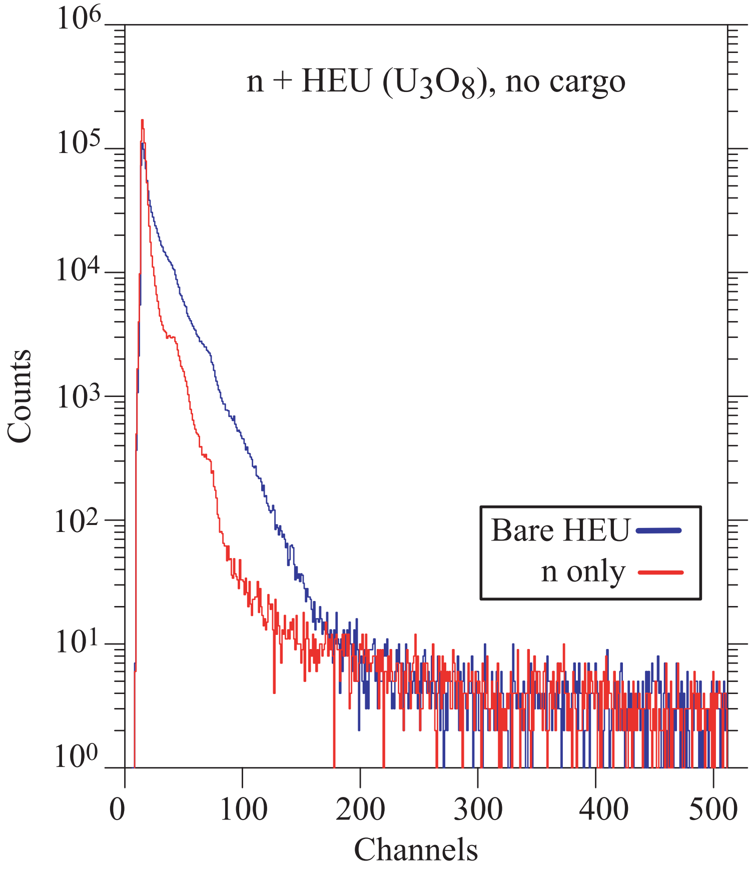
- 30 s neutrons on, 100 s off with γ counting
- 2 ft wood
- En = 14 MeV (d,t)
- $Y_n = 5 \times 10^{11}$ n/s

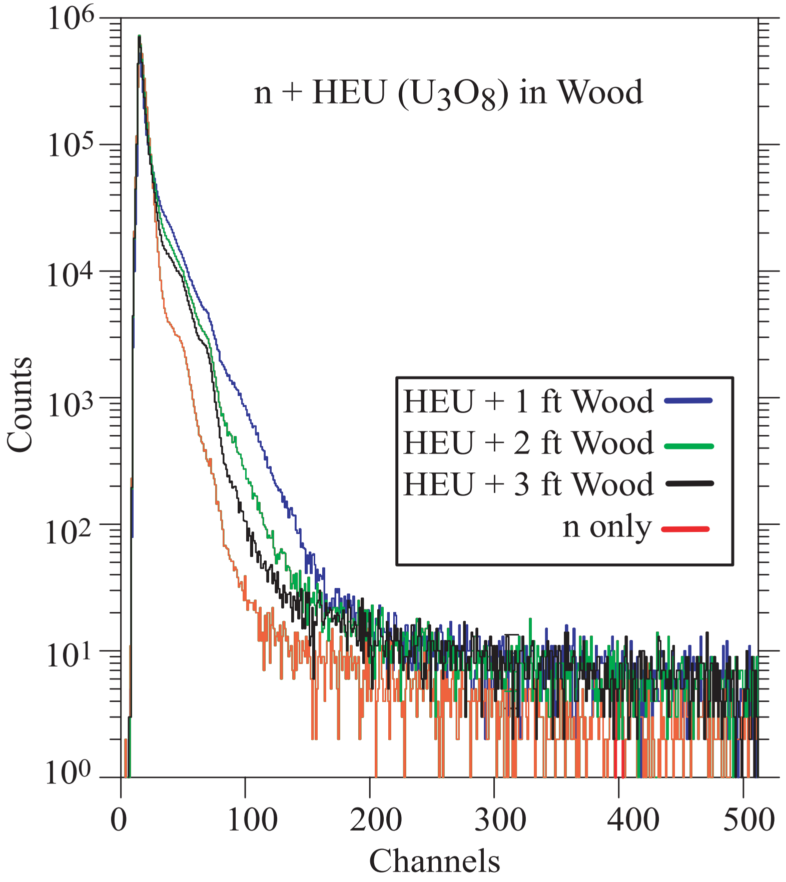
$E_n < 3$ MeV, 1 irradiation



Significant counts over background with $E_\gamma > 3$ MeV

$E_n < 3$ MeV, 1 irradiation

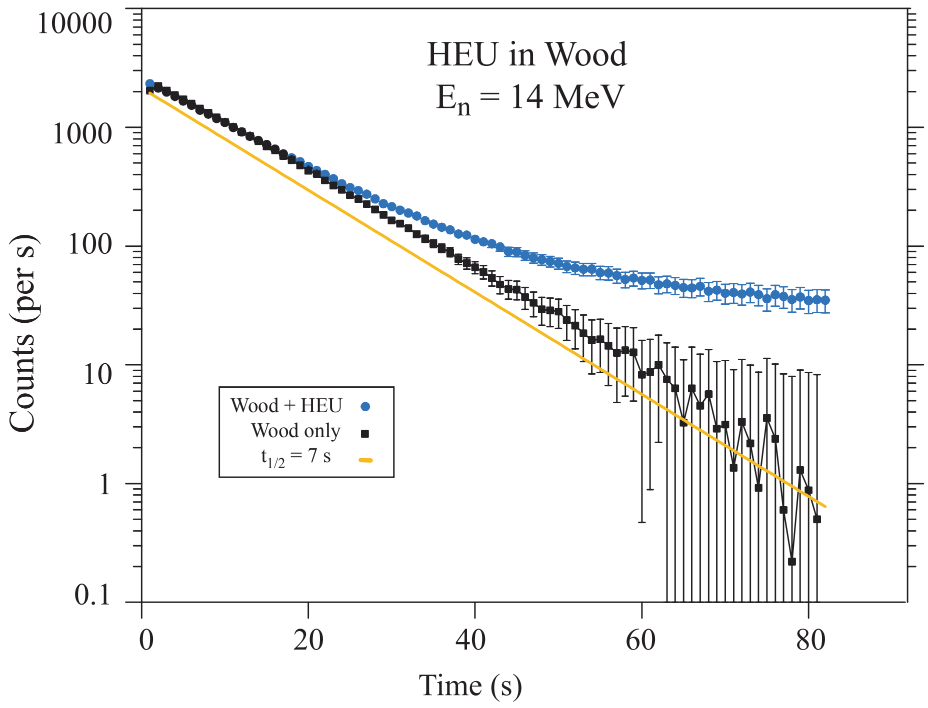




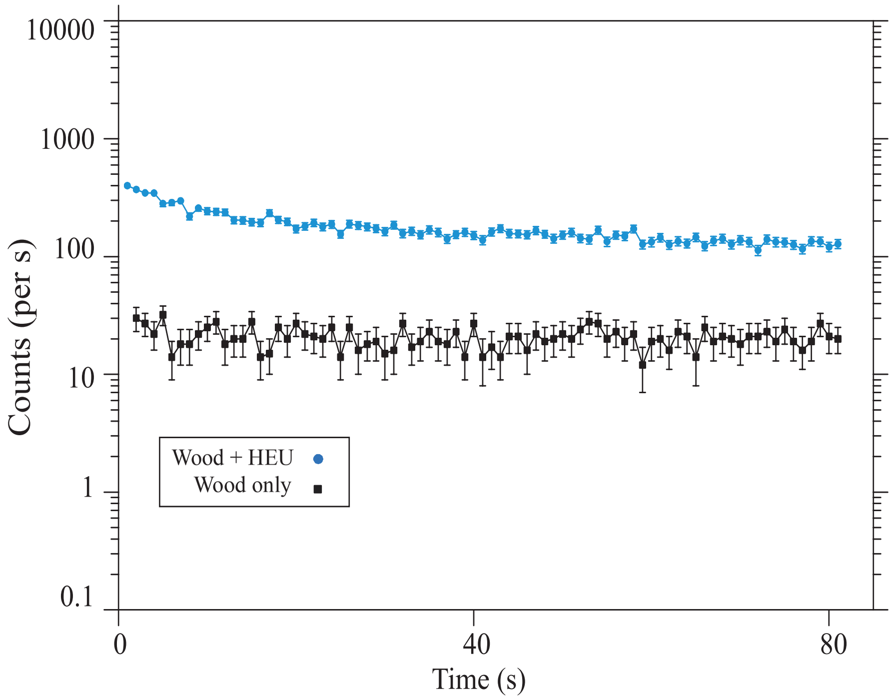
Active background is constant

Decay curves for 3 MeV < E_γ < 4 MeV

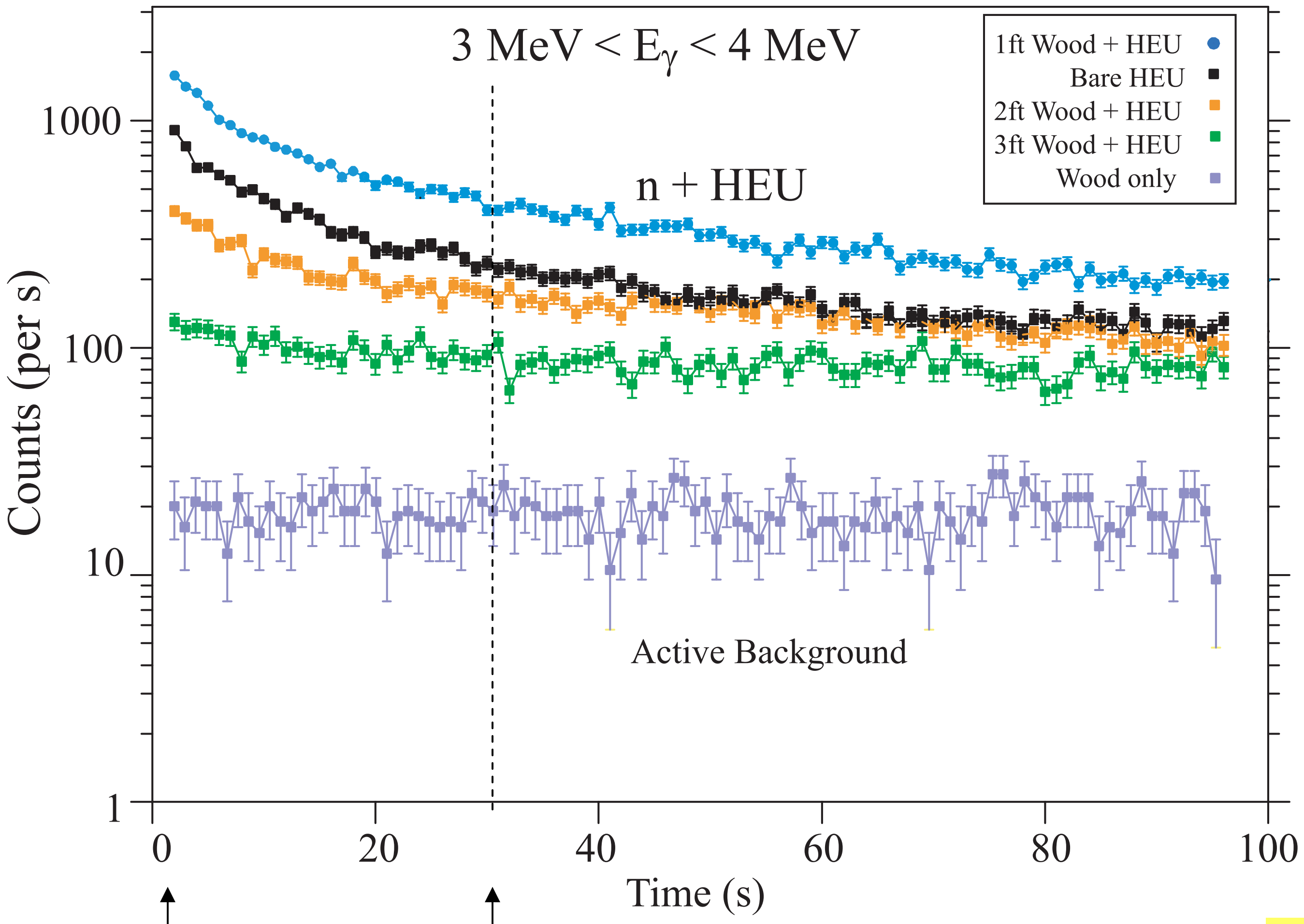
E_n = 14 MeV, 50 irradiations



E_n < 3 MeV, 1 irradiation



Fission in one cycle!

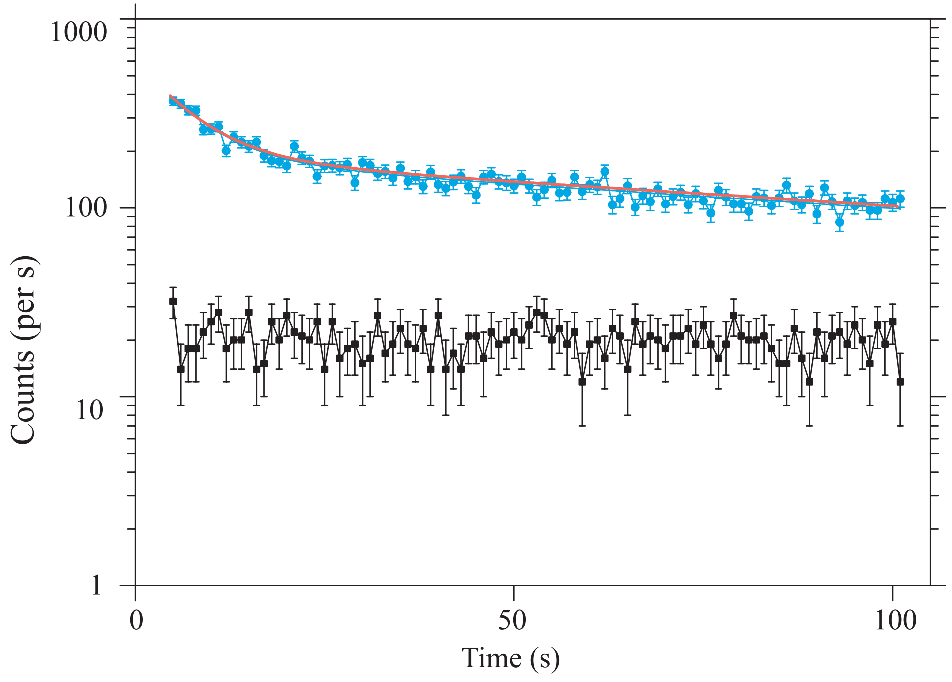


n-gen OFF

One minute since start of scan

→ 1 γ / g HEU

Decay times coincide with fission products



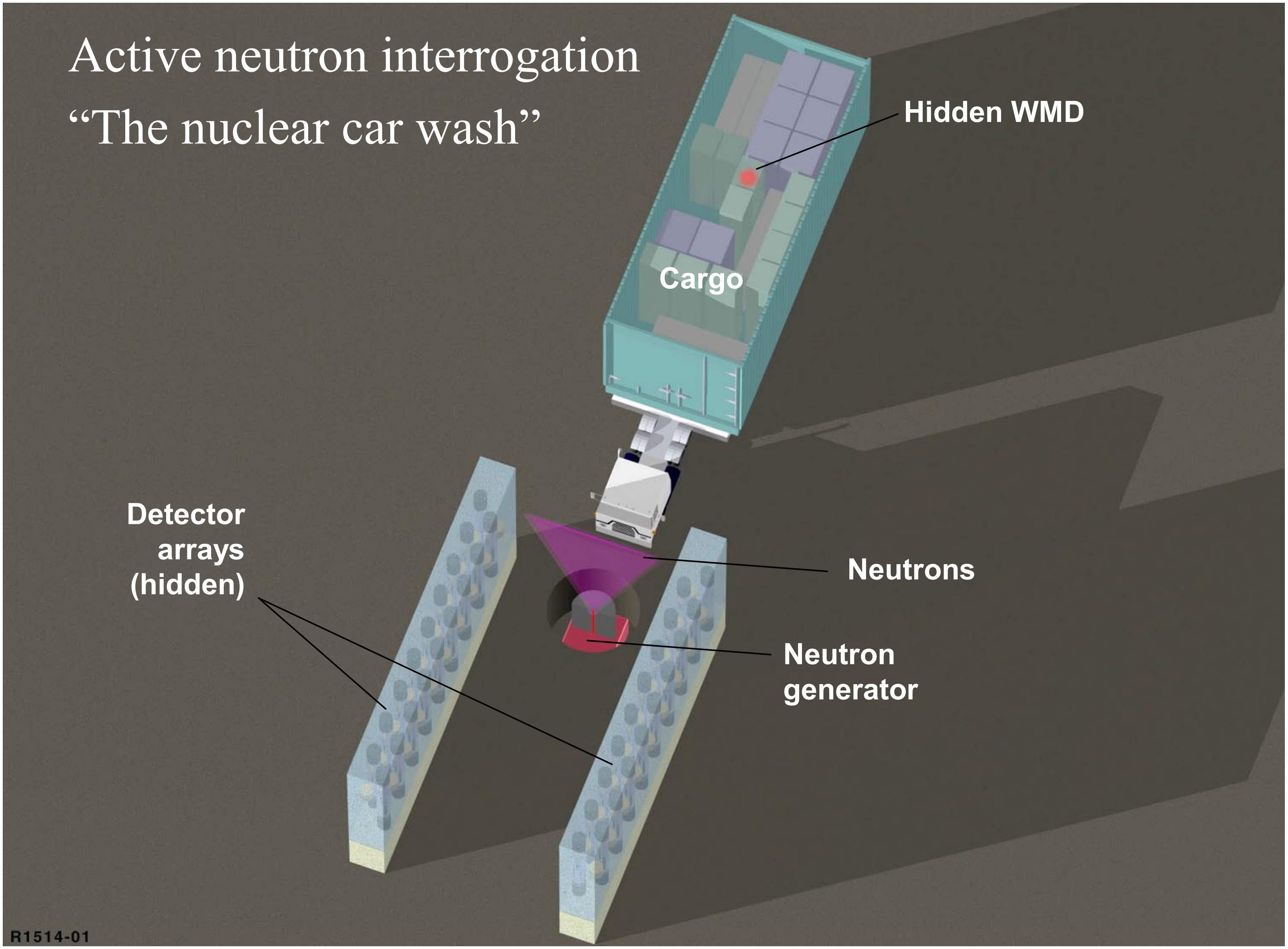
^{235}U Products with most intense activity at $E_\gamma > 3$ MeV and with $t_{1/2} < 10$ min

Product	$t_{1/2}$ (s)
^{90}Rb	156
^{91}Rb	58
^{92}Rb	4.5

Next Steps

- Install trolley system for automatic translation of a fully loaded container.
- Design and fabricate a prototype for field evaluation with real weapons and components.
- Design of deployable / commercial system.
- Combine with tomographic systems.

Active neutron interrogation
“The nuclear car wash”



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Douglas Manatt
Joe Mauger
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