

**H.P. Mumm**

National Institute of Standards and Technology

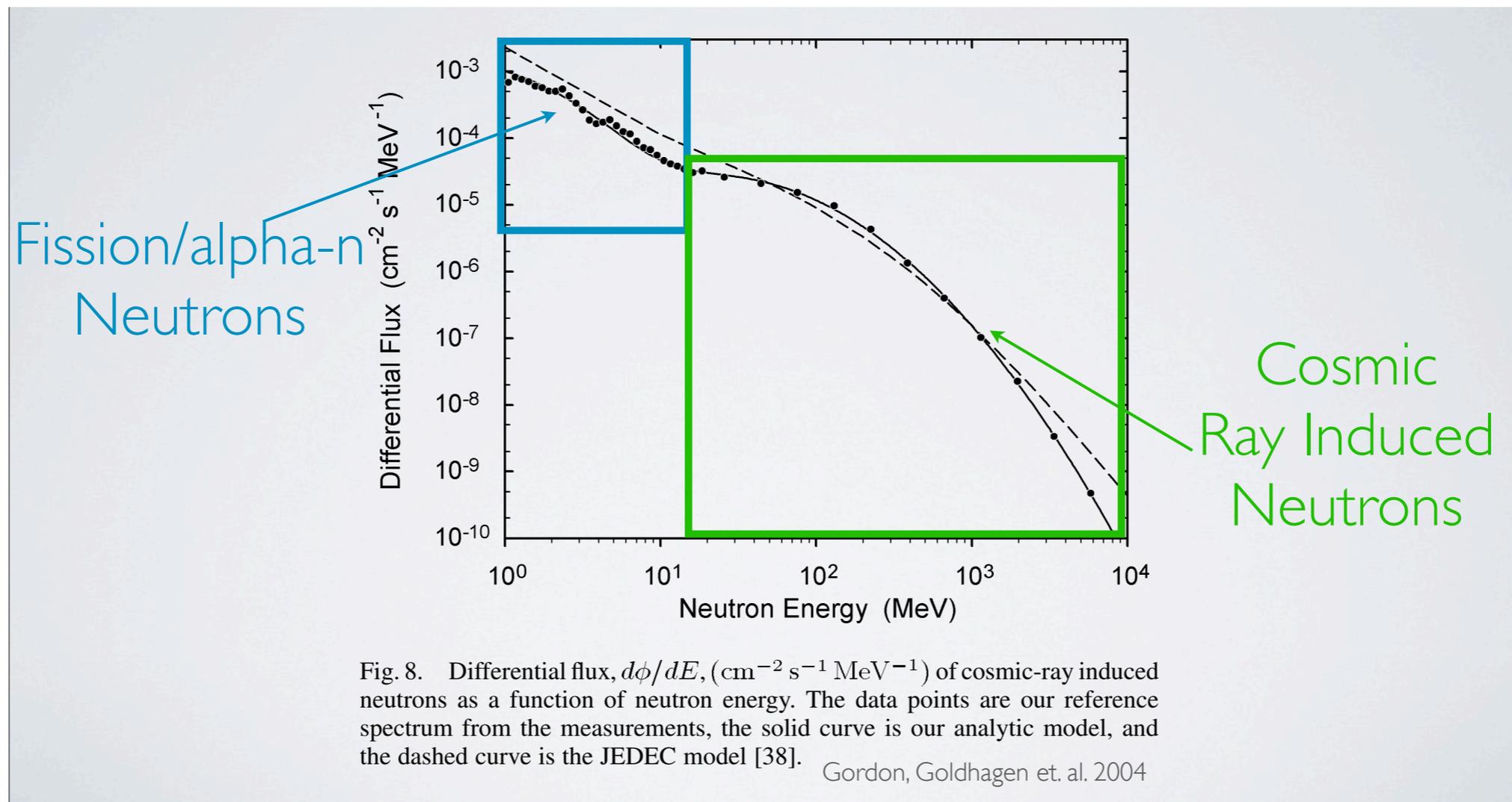
Most slides courtesy of Chris Bass

arXiv:1206.4036v1 [physics.ins-det] 18 Jun 2012



## How to build a highly efficient fast-neutron spectrometer

- fast neutrons (*up to GeV scale*) produced underground through
  - *( $\alpha,n$ ) reactions*
  - *spontaneous fission*
  - *spallation neutrons from cosmic rays*



## Limitations of traditional neutron detectors

- expensive and-or hard to get materials
  - non-linear light response
  - energy threshold
- 
- **Neutron capture on  $^3\text{He}$** 
    - use  $^3\text{He}$  proportional tubes in combination with scintillator
    - $^3\text{He}$  tubes very efficient and  $\gamma$ -insensitive
    - *$^3\text{He}$  expensive and limited availability*
    - *detector efficiency limited by geometry*
  
  - **Dope organic scintillator with a high neutron capture cross-section isotope**
    - PMTs measure recoil and capture signals
    - can provide a much cleaner signal between proton-recoil and neutron capture
    - *commercially-available doped scintillator are expensive*
    - *liquid scintillators potentially hazardous*
    - *nonlinear response*

## segmentation for improved energy resolution

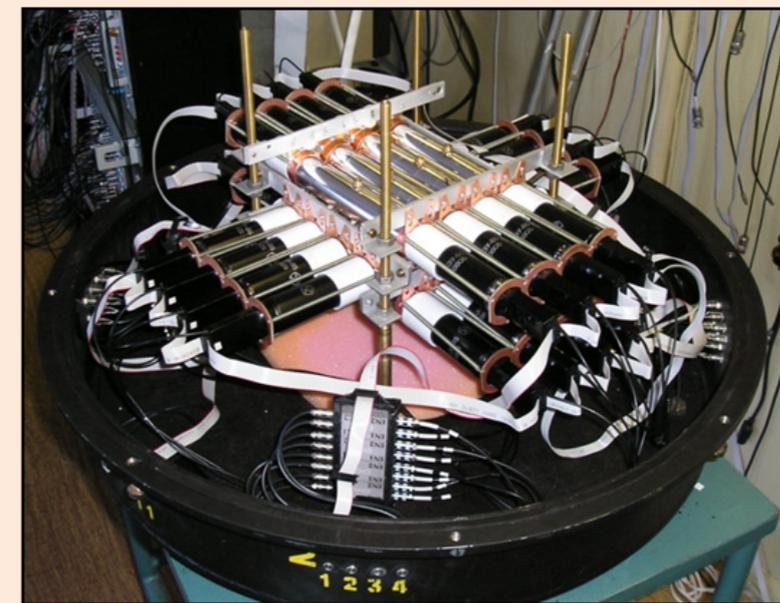
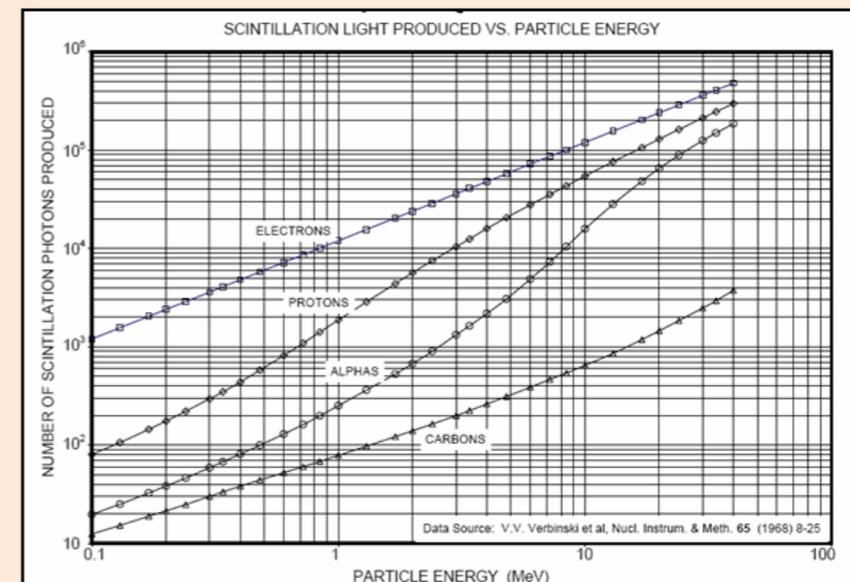
Organic scintillators exhibit nonlinear (but known) light response to charged particles.

**Sum of light intensity  
from energies of  
individual recoils**  $\neq$  **Light intensity  
for the summed  
recoil energy**

*(so, total collected light cannot definitely  
determine neutron energy)*

However, if light from each recoil can be measured separately, then neutron energy can be determined.

Optically separate sections of scintillator so that on average there is only one recoil event per section.



Slides courtesy of Chris Bass

Reactor Antineutrino Workshop. 12/11/2012

## capture nuclides

Choose capture material with high neutron capture cross section

Capture nuclide ensures that the neutron has completely thermalized

Isotope	Reaction	thermal neutron cross section (b)	Signatures
$^3\text{He}$	(n,p)	5330	p, $^3\text{H}$ : Q = 0.764 MeV
<b><math>^6\text{Li}</math></b>	<b>(n,<math>\alpha</math>)</b>	<b>940</b>	<b><math>\alpha</math>, <math>^3\text{H}</math>: Q = 4.8 MeV</b>
$^{10}\text{B}$	(n, $\alpha$ )	3840	$\alpha$ , $^7\text{Li}$ , $\gamma$ : Q = 2.8 MeV
$^{113}\text{Cd}$	(n, $\gamma$ )	21000	$\gamma$ -burst: Q ~ 8 MeV
$^{155}\text{Gd}$	(n, $\gamma$ )	61000	$\gamma$ -burst: Q ~ 8 MeV
$^{157}\text{Gd}$	(n, $\gamma$ )	260000	$\gamma$ -burst: Q ~ 8 MeV
$^{235}\text{U}$	(n,f)	580	fission fragments: Q ~ 200 MeV

- Plan was for lithium to be in aqueous solution
  - so scintillator must accept water-based solutions
  - such scintillator cocktails exist for Health Physics uses
- safety (for use underground/Reactors)
  - needs high flashpoint
  - non-toxic
- cheap
  - Zinsser Analytic developed a Scintillator that accepts aqueous solutions 40% vol. (Quickszint 164)
    - based on high flash point materials ( > 100 C )
    - relatively safe – contains only aromatic solvent (diisopropyl naphthalene) white oil, non-ionic surfactants, and fluors (PPO, bis-MSB)
- relatively cheap: ~ \$200/L

## lithium chloride solution chemistry

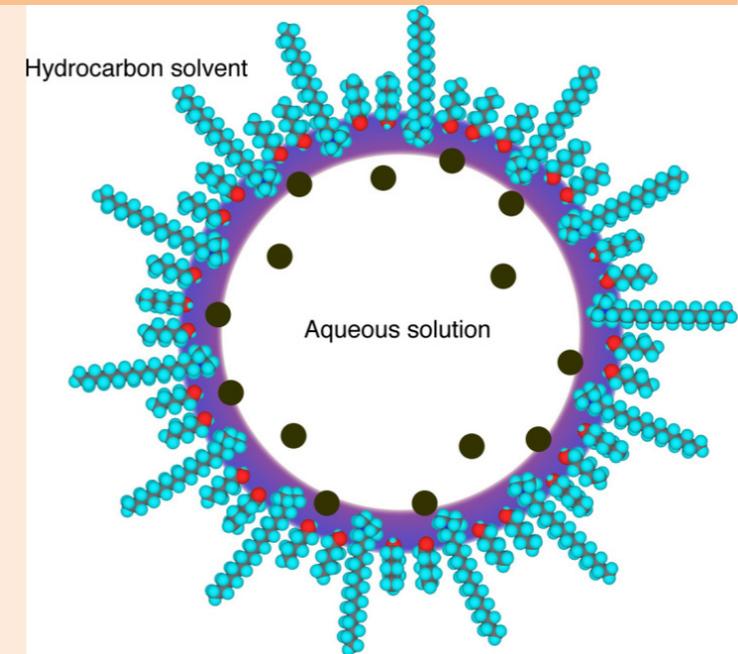


- excess hydrochloric acid can degrade scintillator
- lithium carbonate insoluble in water
- remove water to leave lithium chloride salt
  - lithium chloride hydrates form during heated water removal
    - bake out salt to eliminate hydrates (1-2 days)
- use lithium chloride salt to make LiCl(aq) solutions
  - (verify concentration by density)
- solubility of LiCl in water limits concentration of LiCl(aq) to ~ 12 M



## microemulsions

- isotropic mixtures of oil, water & surfactant that occur with minimal mixing and are highly stable
- surfactant molecules form a monolayer at interface between oil and water
- formation and stability governed by free energy of emulsification:



$$\Delta G^{\text{emuls}} = \Delta A \gamma_{\text{OW}} - T \Delta S^{\text{disp}} \rightarrow \Delta A \gamma_{\text{OW}} < T \Delta S^{\text{disp}}$$

interfacial area →  $\Delta A$   
 interface tension →  $\gamma_{\text{OW}}$   
 temp →  $T$   
 entropy →  $\Delta S^{\text{disp}}$

must be neg. for dispersion formation and thermo. stability

for microemulsion, area change is  $10^4$  to  $10^5$

pos., because microemulsion results in more random systems

must be very low: 0.01 mN/m

no surfactant:  $\gamma \sim 50$  mN/m  
 w/ surfactant:  $10^{-2}$  to  $10^{-4}$  mN/m

Slides courtesy of Chris Bass

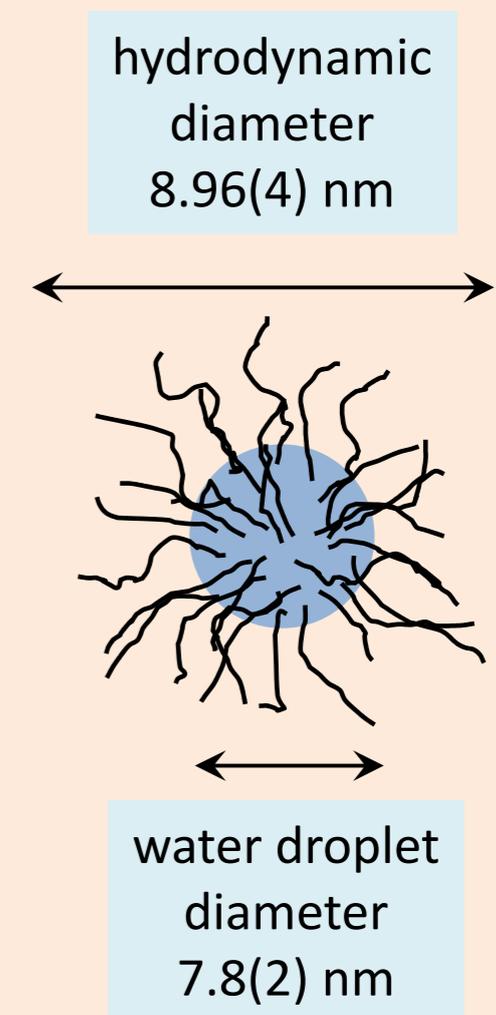
## dynamic light scattering studies

- DLS measures drop size based on intensity fluctuations of scattered light
- hydrodynamic diameter : includes surfactant tails that extend into the scintillator bulk
- surfactant in Quickszint : *nonylphenol ethoxylate*  $\rightarrow$   $C_9H_{19}$  hydrophobic tail
- assume random walk for extent of C-C and C-H bonds into bulk (570 pm)

### neutron captures occur inside water droplets

range of charged particles from  ${}^6\text{Li}(n,\alpha)t$  : **triton: 2.73 MeV 66.15  $\mu\text{m}$**   
(calculated using SRIM) **alpha: 2.05 MeV 10.70  $\mu\text{m}$**

- shifting of capture spectrum to lower energies
- loss of energy resolution in capture spectrum

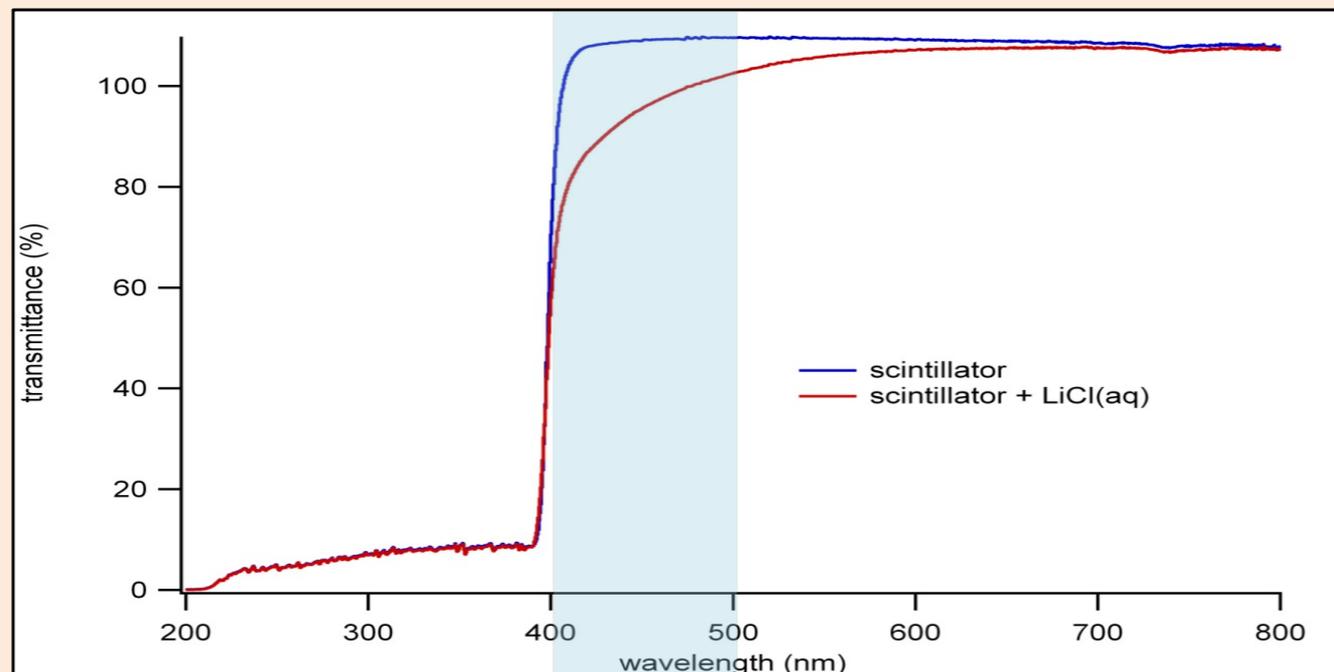


## Loading the liquid scintillator cocktail

- created batches of natural lithium-loaded scintillator:
  - concentration of LiCl(aq) in scintillator : between 0.1% and 10% by volume
  - concentrations of LiCl(aq) : 2.5 M, 5 M, 10 M
- characterize the mix ratio in terms of:
  - emulsion properties
  - optical properties
- optimize mix ratio for
  - maximum possible lithium-loading within the scintillator
  - maximize light transmission through scintillator

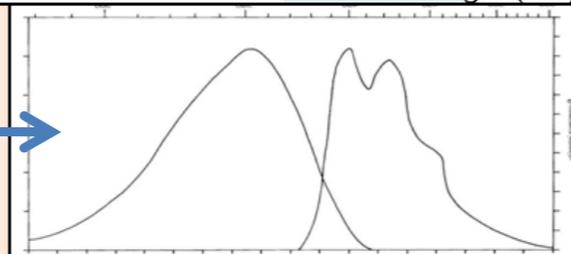


## UV-Vis photospectrometry

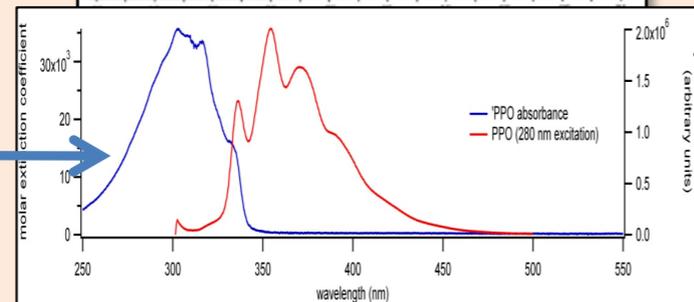


Average transmittance  
(over band) ~ 88%

secondary  
fluor  
(bis-MSB)



primary  
fluor  
(PPO)



Measure UV-Vis transmittance for  
samples of Quickszint with various  
loadings and concentrations of LiCl(aq)

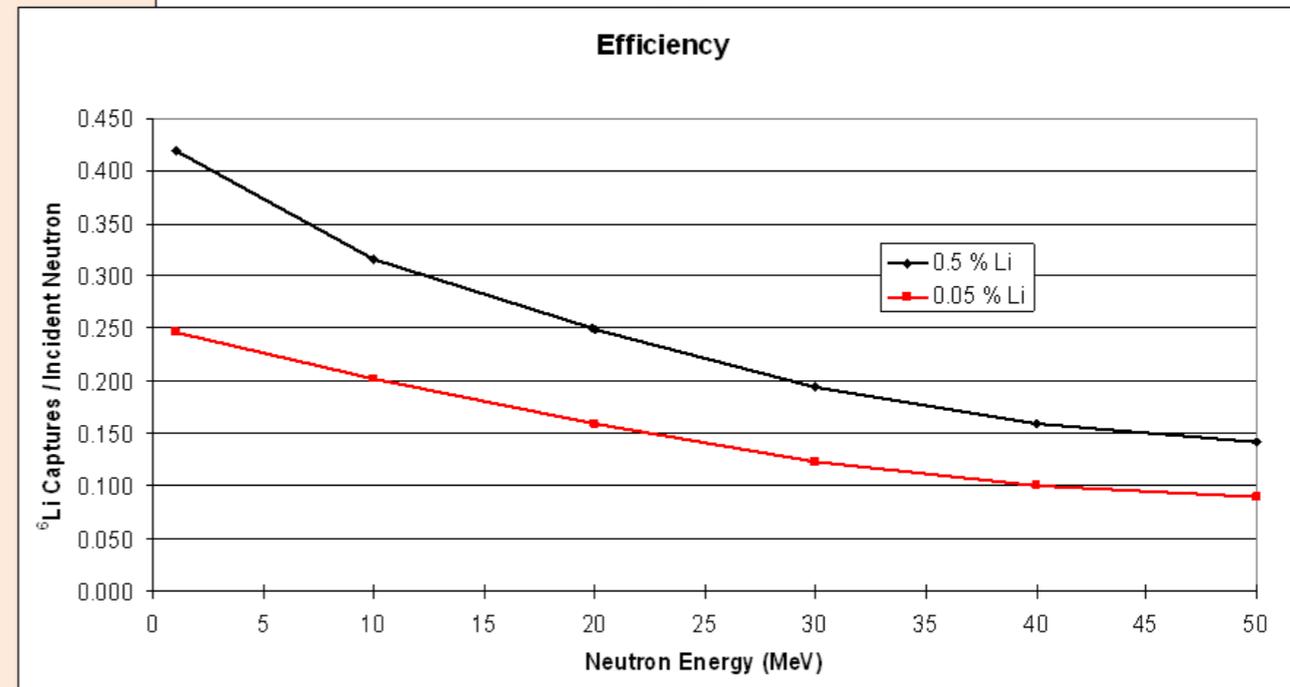
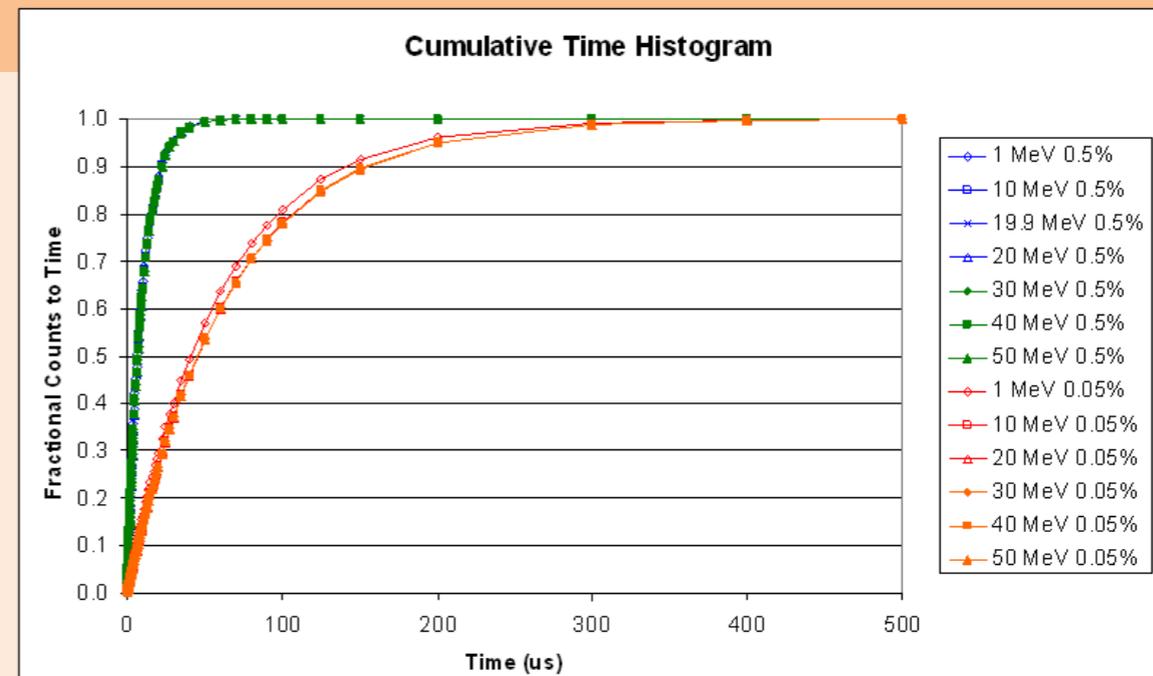


## MCNP lithium concentration studies

### MODEL:

segmented 50 cm x 50 cm cube  
(25 slabs)  
of  $^6\text{Li}$ -loaded liquid scintillator  
0.05% & 0.50% lithium by weight,  
with 95%  $^6\text{Li}$  enrichment

- load as much lithium into scintillator as possible for efficiency and capture times
- upper limit on loading due to optical attenuation



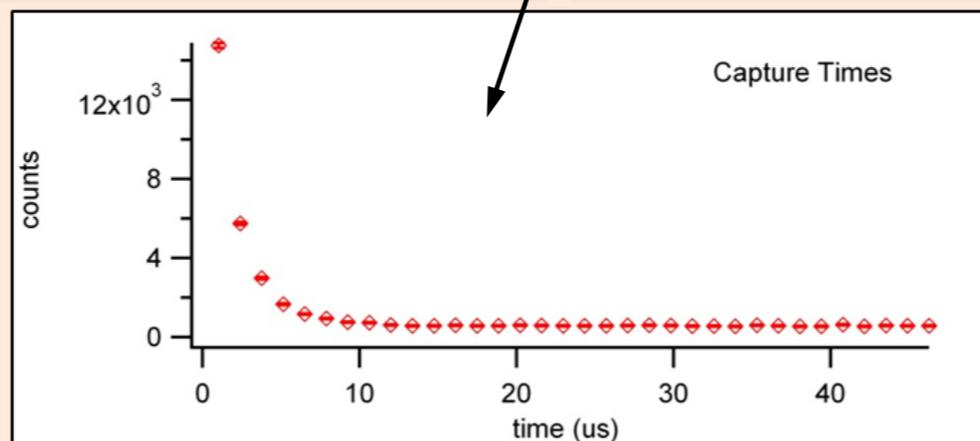
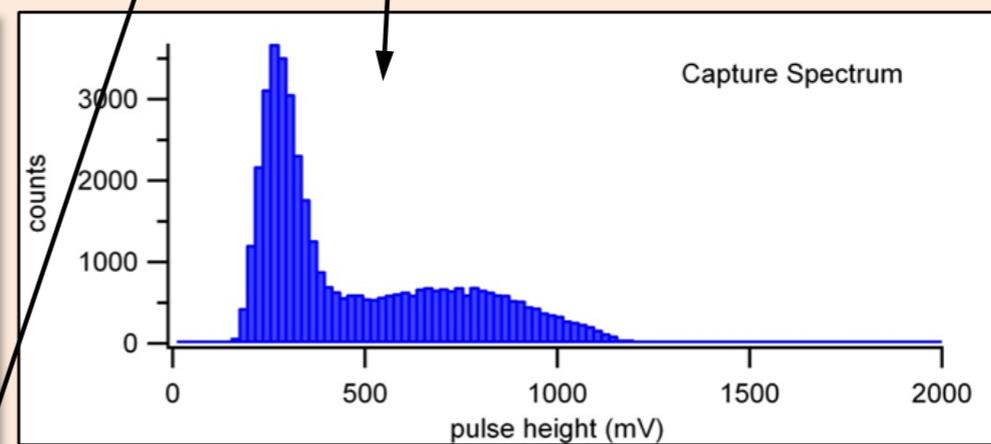
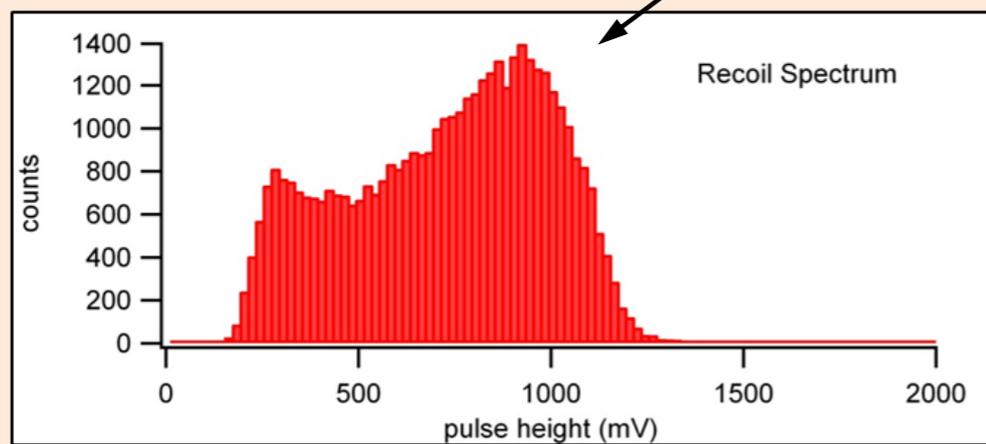
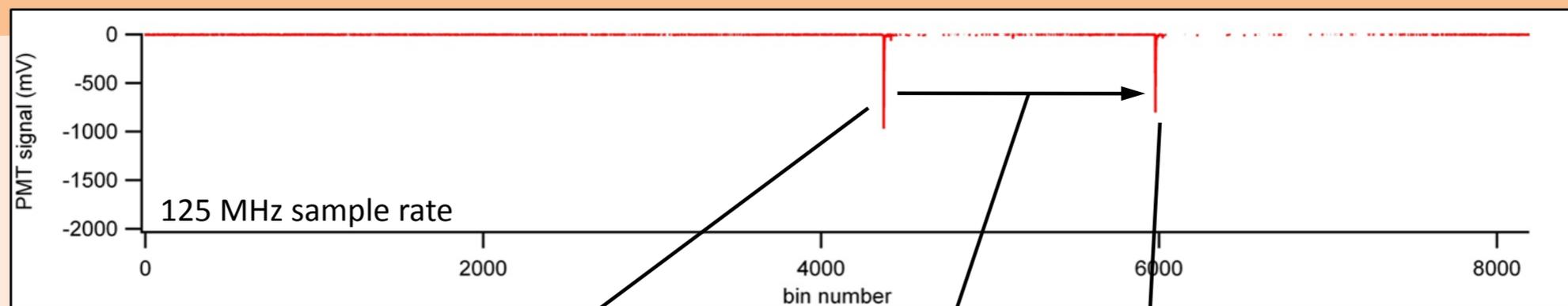
Slides courtesy of Chris Bass

## test detector design

- 125 mL cylindrical glass cell
- twin fill ports capped with Swagelock
- externally coated with Bicorn reflective paint
- coupled to Burle 8850 PMT with optical grease
- high linearity base
- shrouded within 1/8" wall aluminum tube



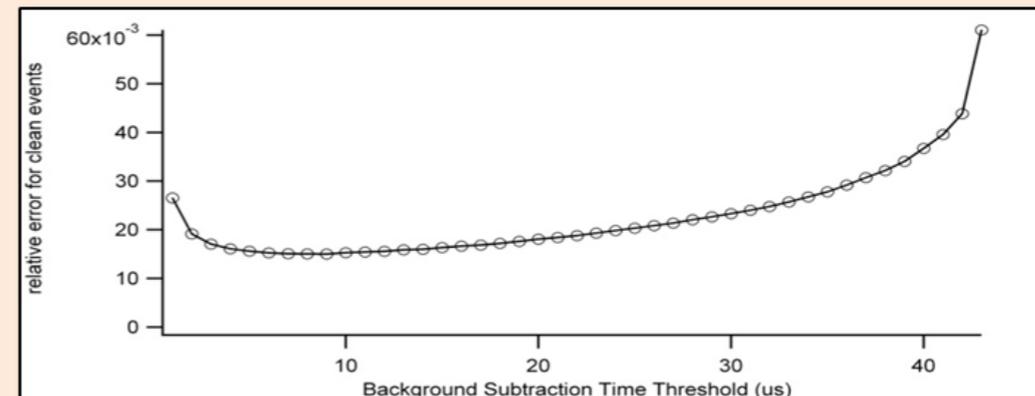
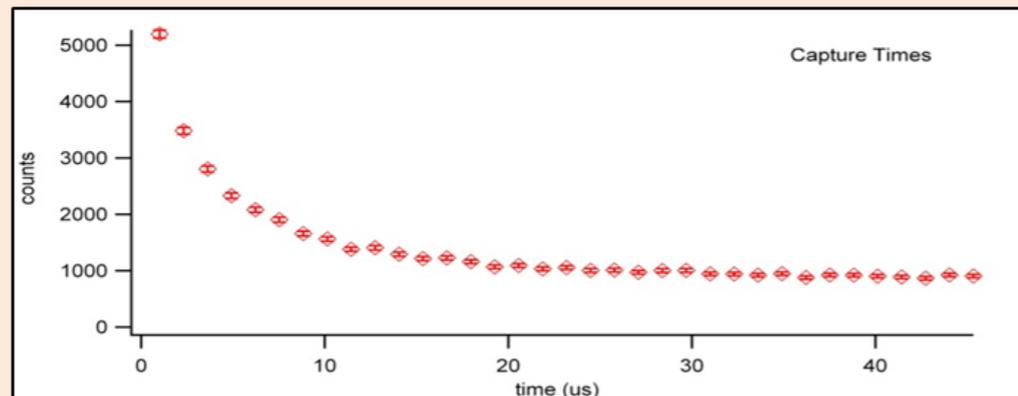
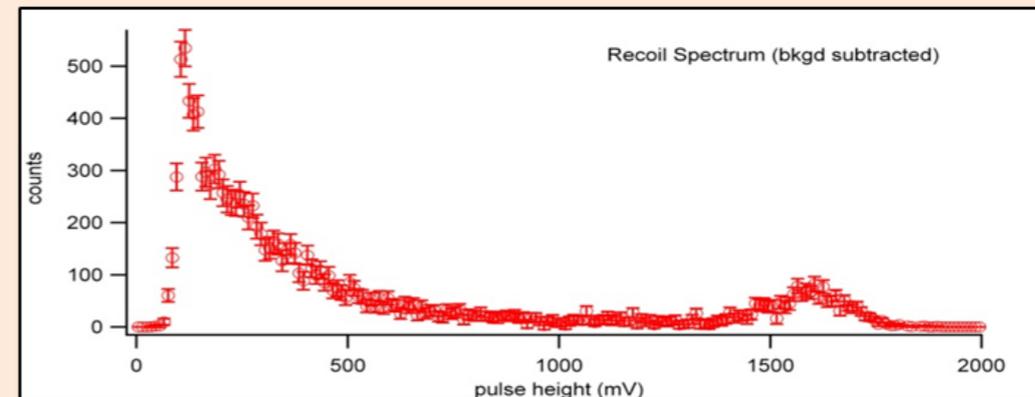
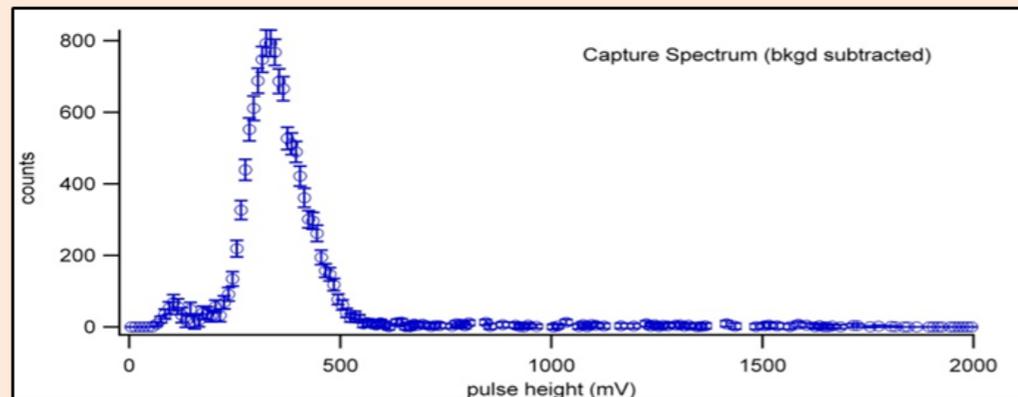
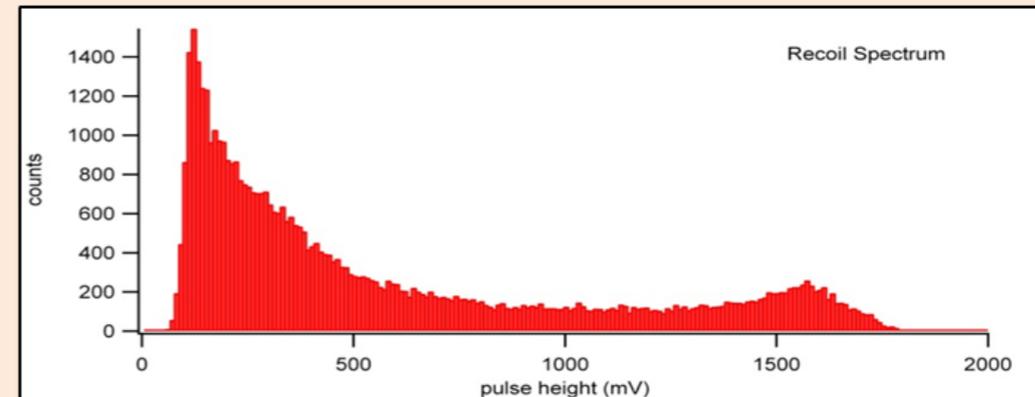
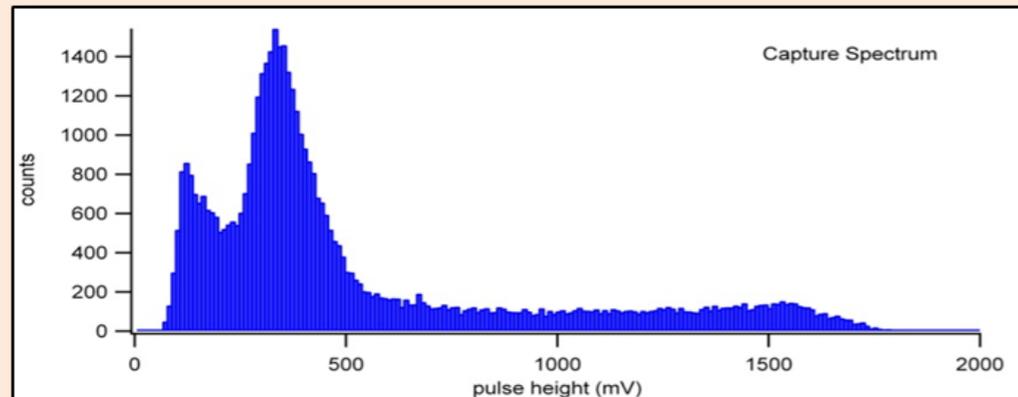
## waveform analysis



Slides courtesy of Chris Bass

Reactor Antineutrino Workshop. 12/11/2012

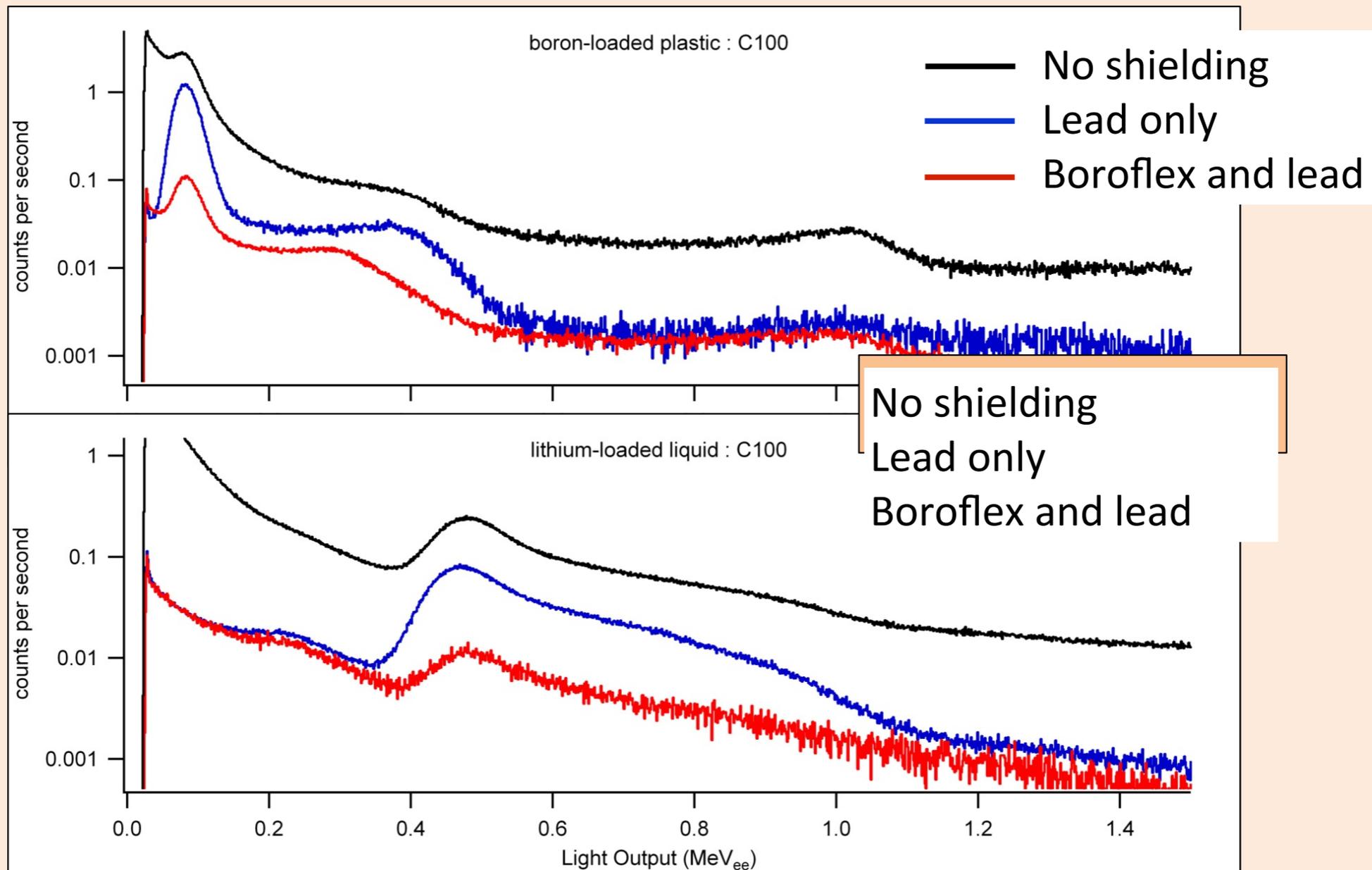
## 0.40% $^6\text{Li}$ -loaded w/ 14 MeV neutrons



Slides courtesy of Chris Bass

Reactor Antineutrino Workshop. 12/11/2012

## thermal neutron studies: C100 – downstream of thermal column & beam stop

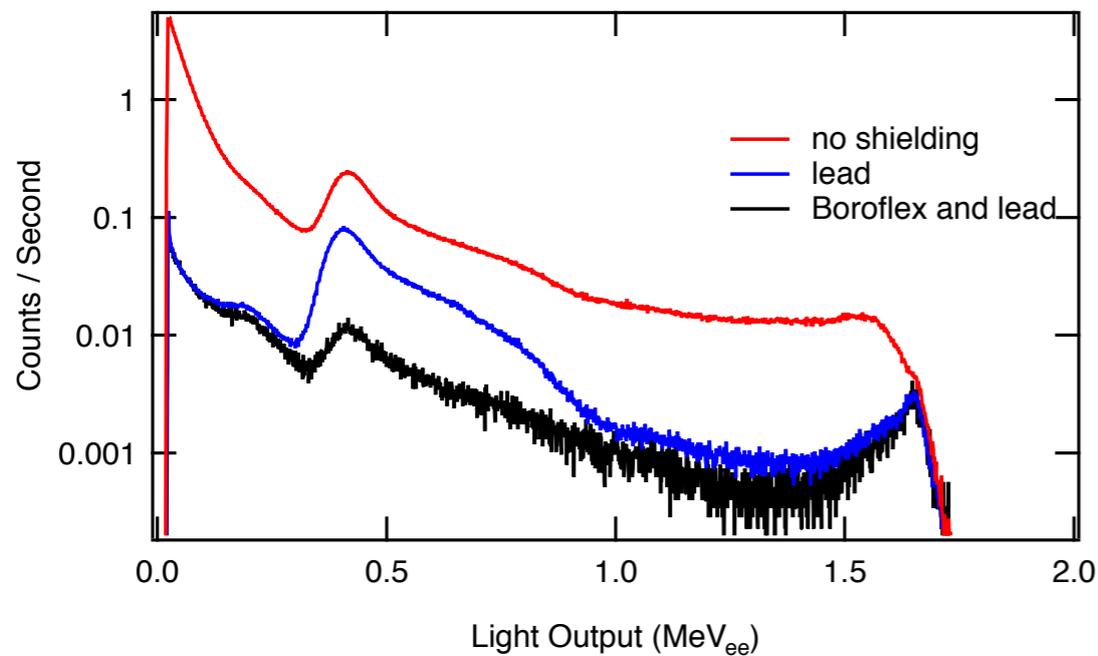


Slides courtesy of Chris Bass

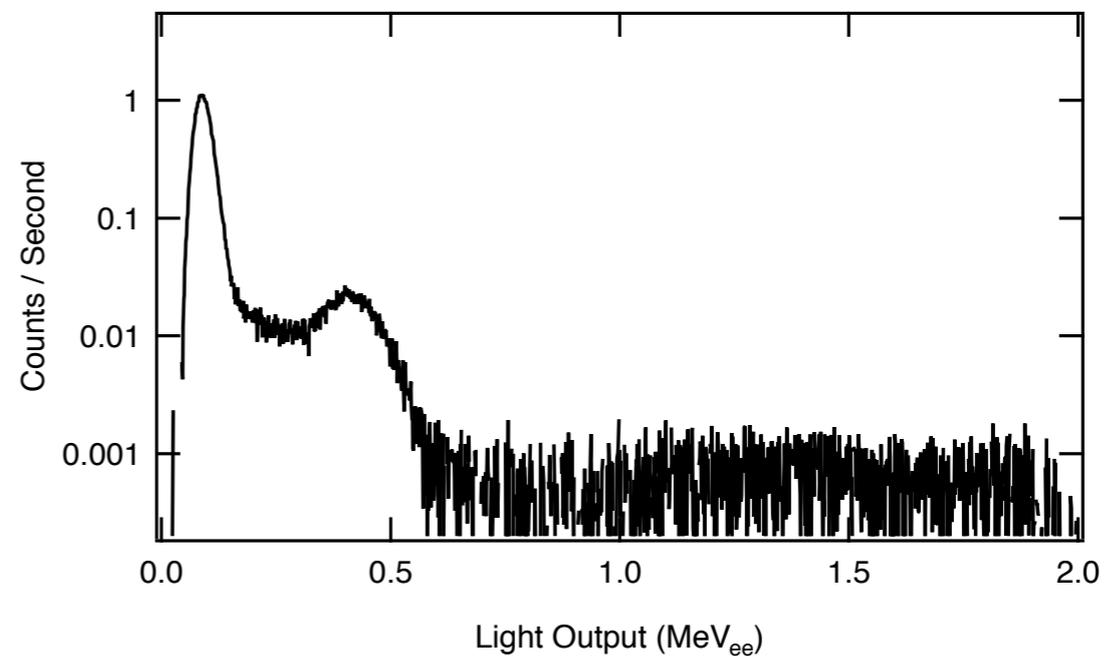
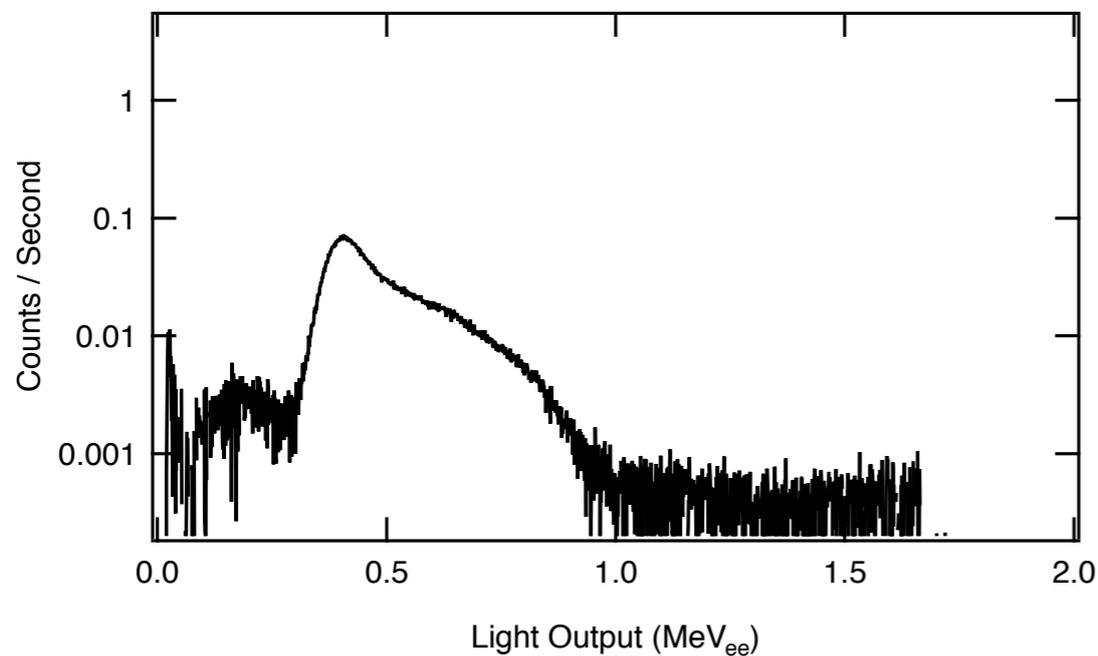
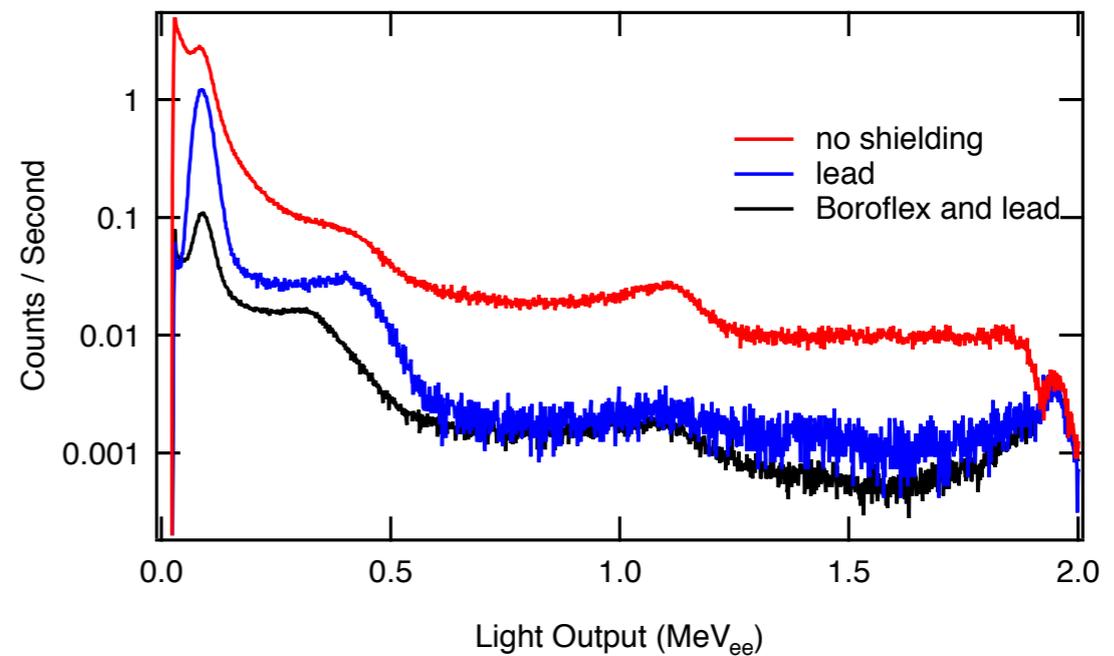
Reactor Antineutrino Workshop. 12/11/2012

# Thermal Neutron Measurements outside the Thermal Column

## $^6\text{Li}$ -loaded scintillator



## BC-454 boron-loaded plastic scintillator



Integrated fluence  $2.8 \text{ cm}^{-2}$

arXiv:1206.4036v1 [physics.ins-det] 18 Jun 2012

## continuing research for $^6\text{Li}$ -based spectrometer

- **scale up to large volume detector**

- current test cell : (1) PMT,  
est. efficiency < 0.1%



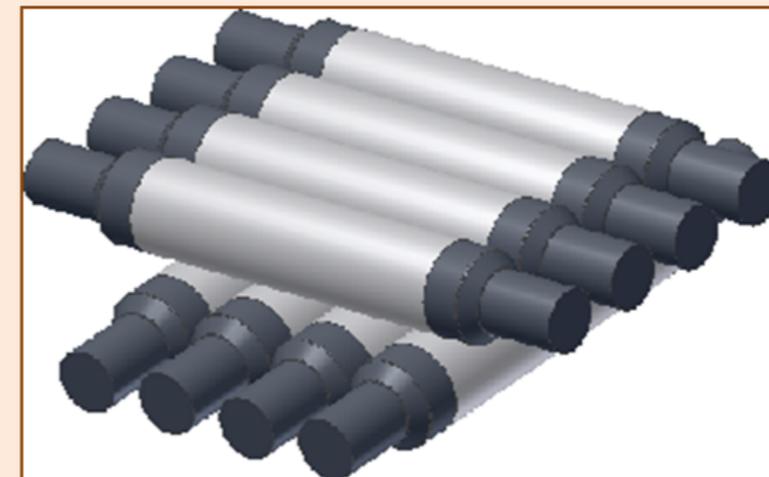
- larger volume cell : (2) PMTs  
MCNP studies suggest an efficiency at the  
few percent level for energies < 10 MeV



- **segmentation studies to optimize cell geometry and configuration**

- MCNP
- neutron measurements to for multiple cells with multiplicity

- **construct prototype for underground testing at *KURF***



## Conclusions:

Li-6 doped micro-emulsions can work well as high-efficiency neutron detectors

- Allow for fine segmentation (non-linear response compensation)
- Reasonably priced
- Safe and easy to work with

## Primary outstanding issues

- accurate measurement of attenuation length
- long term stability