



**National Centre
for Nuclear Research**
Świerk

Organic and inorganic scintillators for Neutron Detection

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A He-3 world crisis triggered a development of new techniques of neutron detection to replace commonly used He-3 detectors, so important in homeland security applications.

The demand of He-3 in USA in 2010 was estimated to be about 65 m³, whereas the supply was dramatically lower of about 20 m³ per year.

It triggered, in the recent years, intensive studies to find alternative detectors, particularly new scintillators.

Below, I will review our activity addressed to the new organic scintillators, in fast neutron detection, and some inorganic scintillators for thermal neutrons detection.



My talk is based mostly on the earlier studies addressed too:

- NORDBALL neutron wall
- French-Belgian DEMON arrangement
- EUROBALL neutron wall
- NEDA arrangement

and the present studies, carried out by our group at Świerk in Poland, addressed mainly to the homeland security, see for example, J. Iwanowska's PhD thesis:

<http://www.ncbj.gov.pl/en/dokument/comparative-studies-neutron-detectors-crisis-3he-supply>

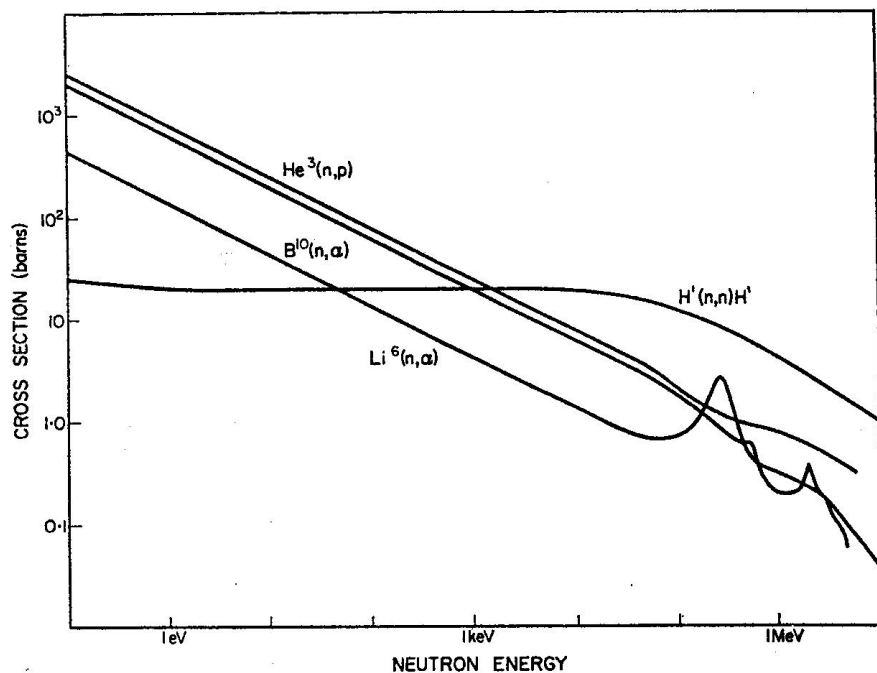


Advantages of organic scintillators in neutron detection

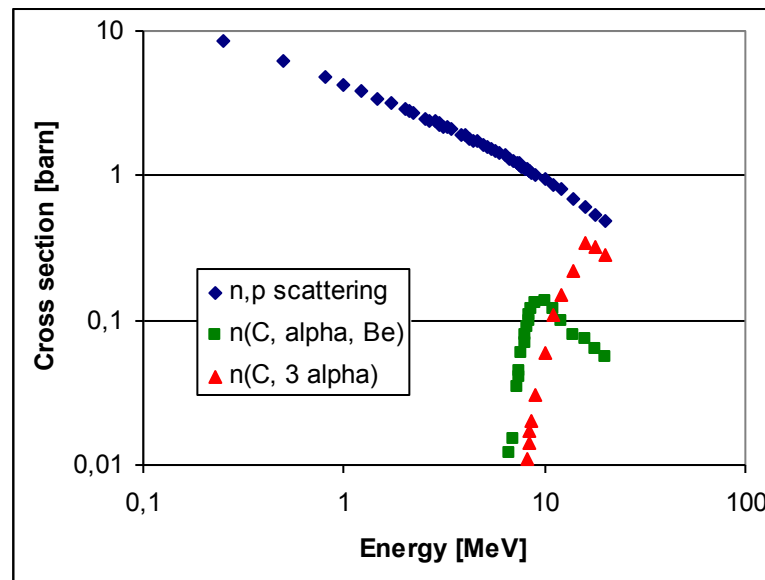
- large cross-section for n,p elastic scattering
- possible large volume – high detection efficiency
- fast output pulse of detectors
- high time resolution in time-of-flight experiments
- good gamma ray discrimination by PSD methods
- high counting rate capability



Detection efficiency



Cross section of n,p scattering in comparison to other reactions involved in the neutron detection

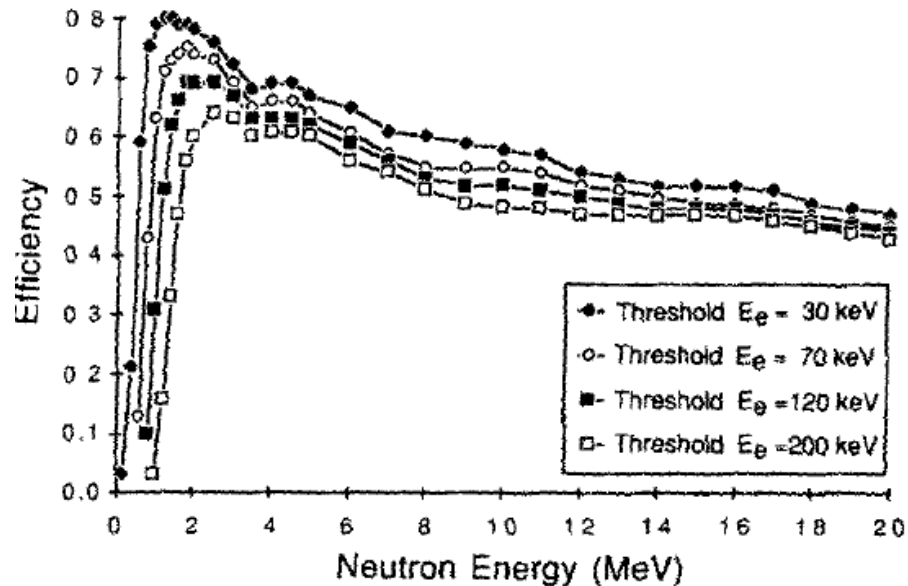


Different reaction channels responsible for neutron detection with energy up to 20 MeV



Detection efficiency-BC501A liquid scintillator

- number of H atoms: 5×10^{22} atoms/cm³
- number of C atoms : 4×10^{22} atoms/cm³
- continuous spectrum of recoil protons
- detection efficiency controlled by a low energy threshold



BC501A detector of NORDBALL

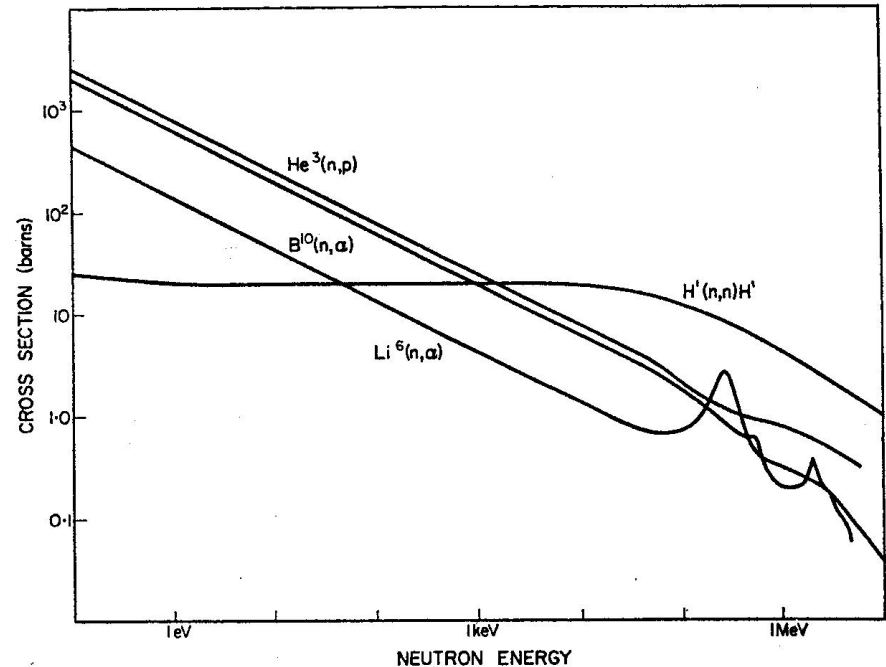
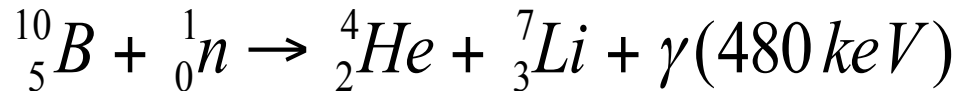
Volume 3.3 I,

detection efficiency for Am-Be neutrons:
about 60%.



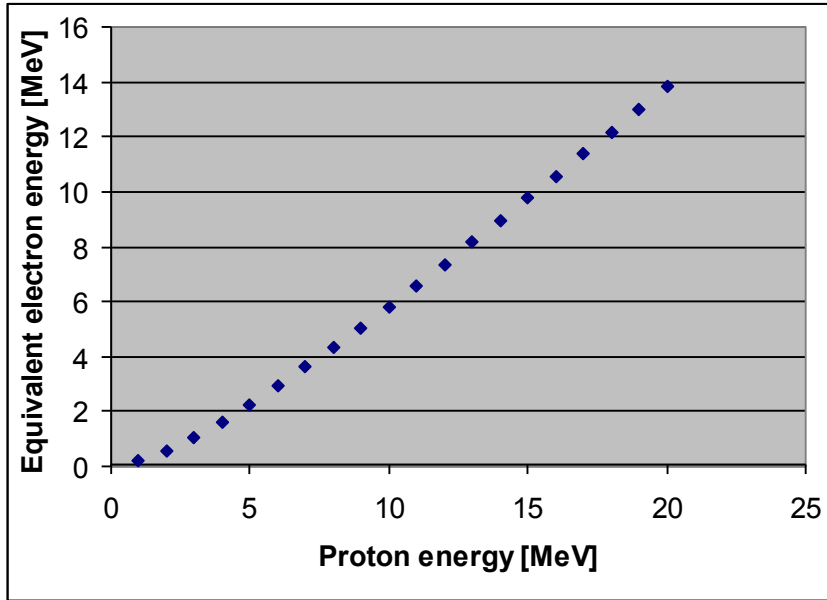
Liquid scintillators in neutron detection, B-10 loaded liquids

- Fast neutrons:
n,p scattering
- Thermal neutrons:
 ^{10}B loaded – neutron detection
via (n,α) reaction
 $Q = 2.3 \text{ MeV}$

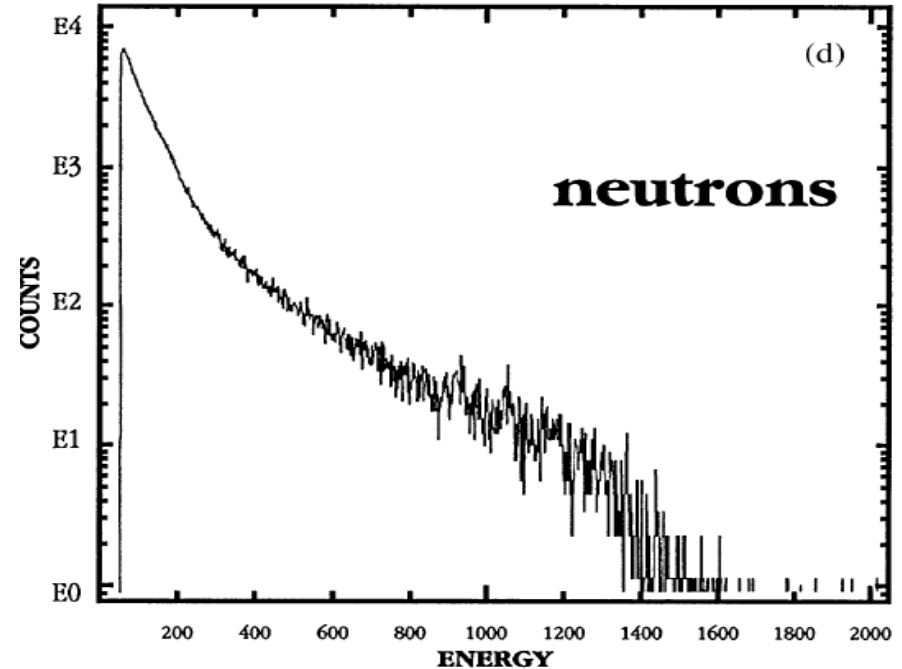




Energy spectra of neutrons



Equivalent electron energy corresponding to given energy of recoil protons (NE213/BC501A)

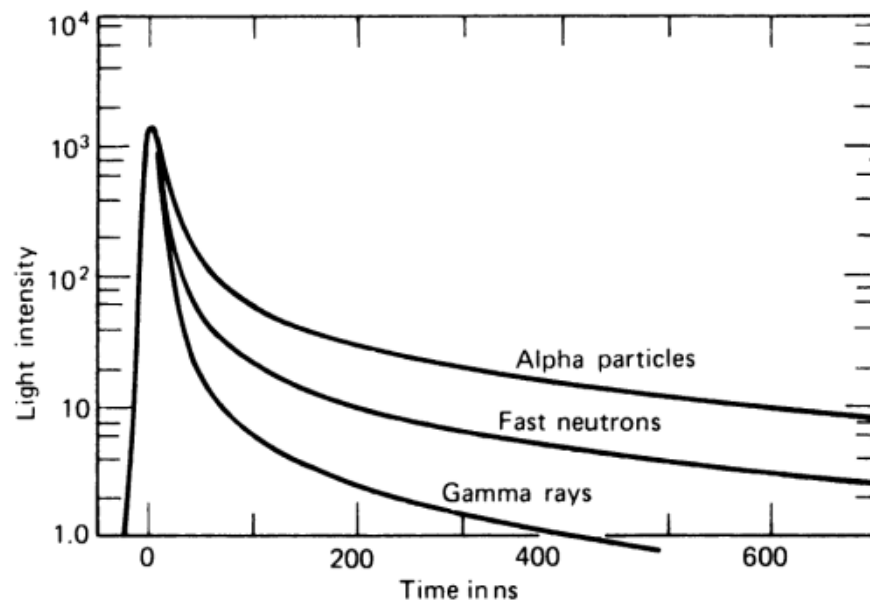


Spectrum of recoil protons from $^{246,248}\text{Cm}$, in BC501A scintillator of EUROBALL neutron wall



Neutron and gamma ray detection

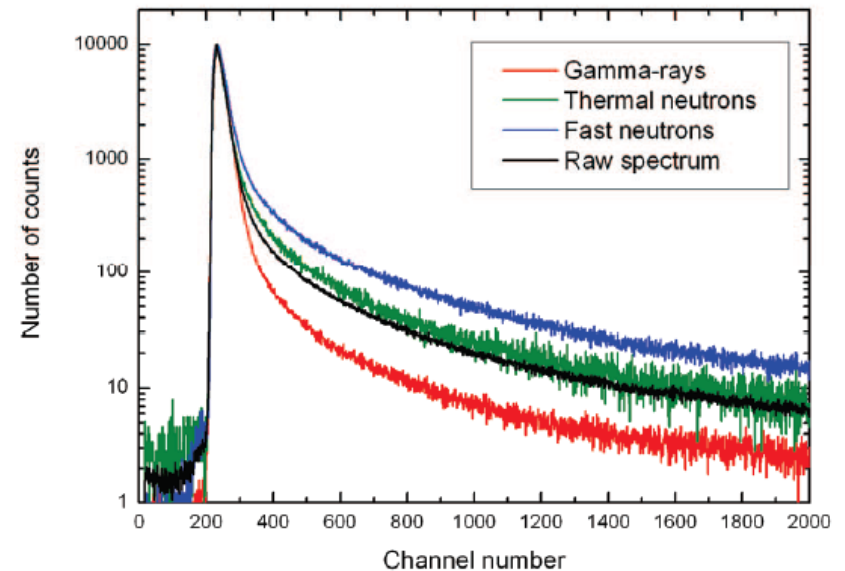
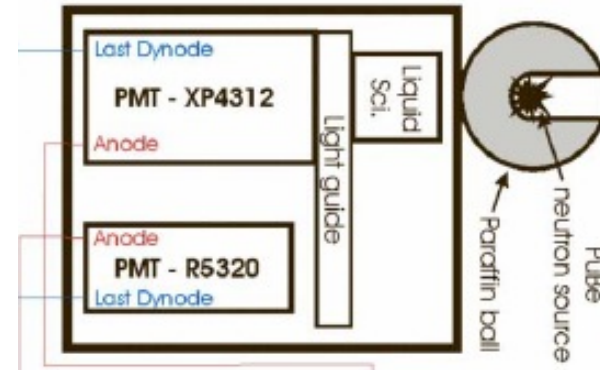
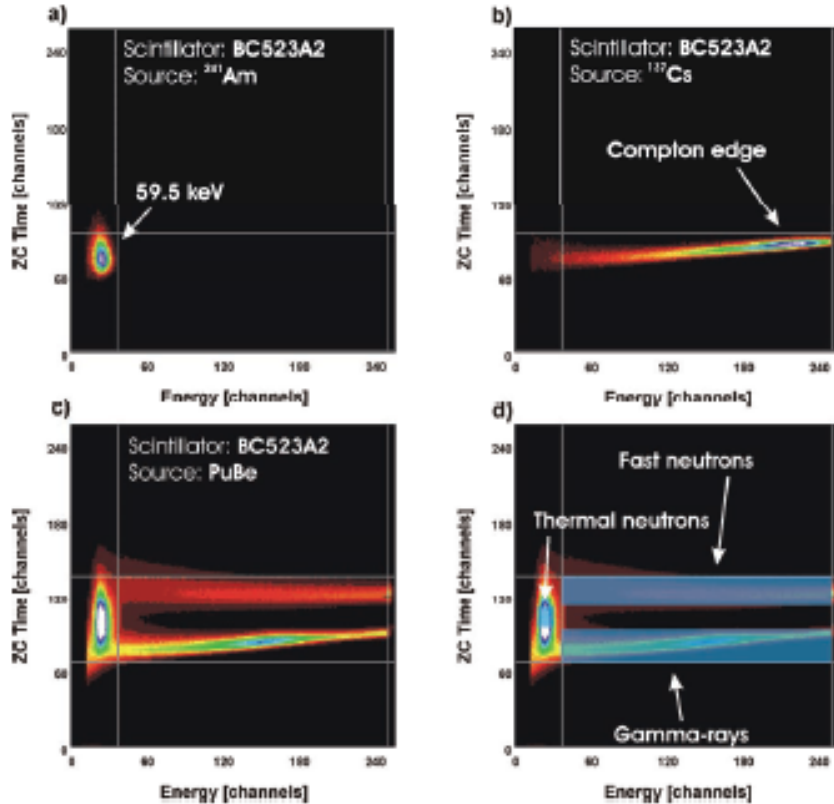
- large cross-section for Compton scattering in organic scintillators
 - 0.72 MeV – 20 barns
 - 2.2 MeV – 10 barns
- **n-gamma discrimination**



Schematical presentation of light pulses due to gamma rays, fast neutrons and alpha particles



Light pulses due to gamma rays and neutrons in ^{10}B loaded BC523A liquid





Decay components of light pulses due to gamma and neutrons

Fitting parameters of the three components calculated for the tested classic liquid scintillators

scintillator	particle	Fast component		Medium component		Long component	
		decay constant (ns)	%	decay constant (ns)	%	decay constant (ns)	%
BC501A	gamma	3.9 ± 0.4	85	27 ± 3	9	240 ± 20	6
	fast n	5.3 ± 0.5	43	40 ± 4	29	280 ± 30	28
EJ301	gamma	4.1 ± 0.4	88	20 ± 2	7	110 ± 10	5
	fast n	5.4 ± 0.5	54	41 ± 4	28	250 ± 30	19
EJ309	gamma	3.8 ± 0.4	81	30 ± 3	11	170 ± 20	8
	fast n	4.8 ± 0.5	46	38 ± 4	32	240 ± 20	22

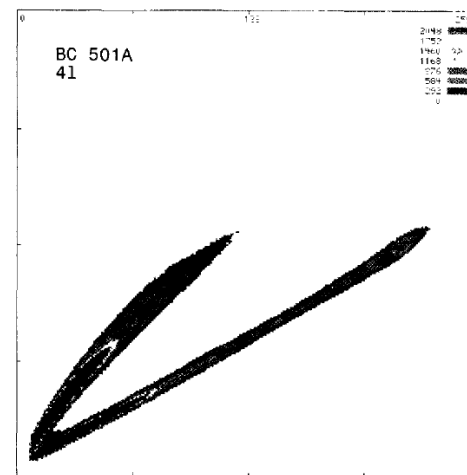
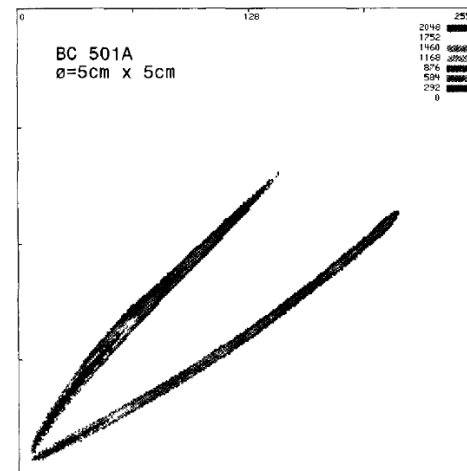
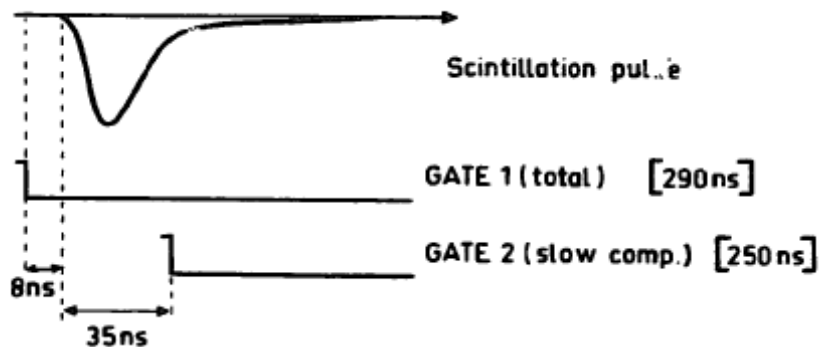
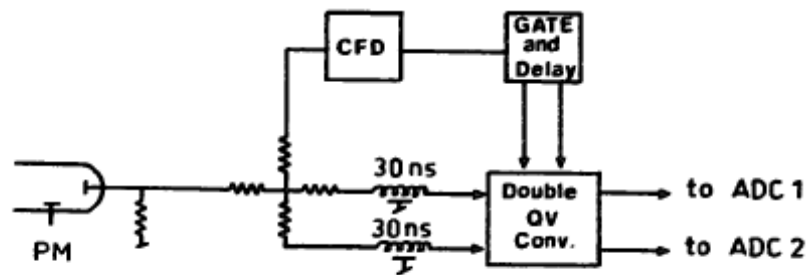


Organic scintillators

- 1960 – stilbene
- Liquid scintillators
 - NE213 BC501A EJ301
 - EJ309 – high flash liquid
 - BC523A, EJ301B, EJ309B – boron loaded liquids
- Composite crystals – mixture of stilbene or *p*-terphenyl and resin – Nikolay Galunov – Kharkov
- Plastics:
 - Lawrence Livermore Laboratory, USA– Natalia Zaitseva - Eljen Technology/ Scionix
 - Amcrys H, Ukraine
 - Saclay, France
- New stilbene – Natalia Zaitseva - Inrad Optics

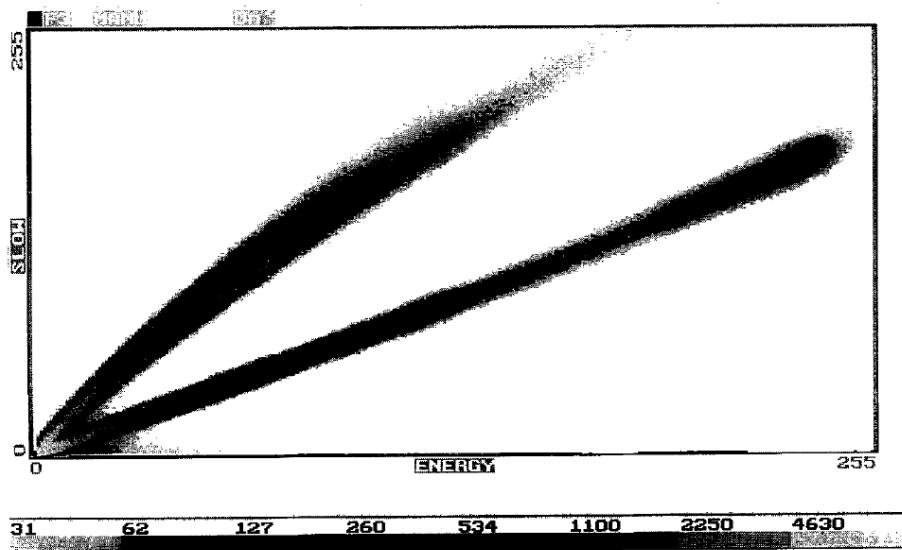


Charge comparison method

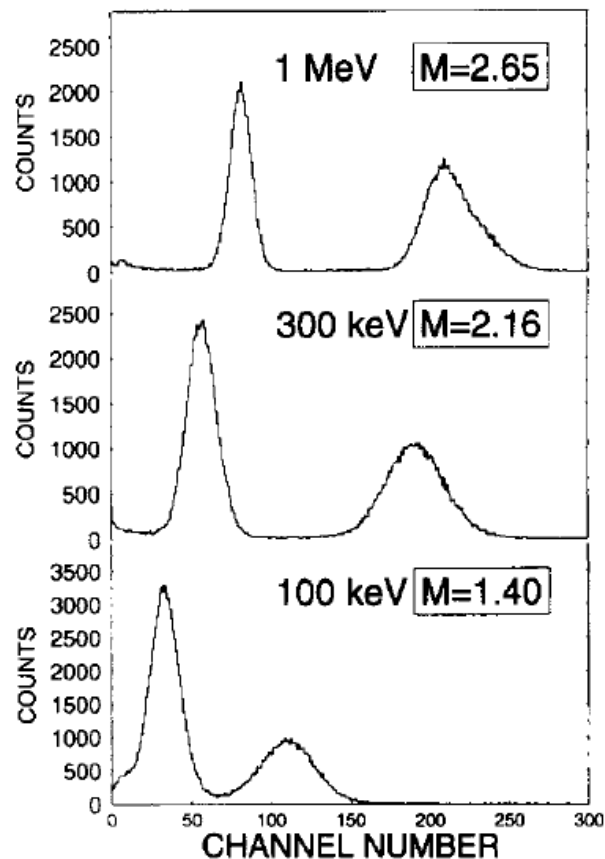




Charge comparison method



Ø5" × 5" BC501A at XP45212B,
Am-Be source, PHE number = 2300 phe/MeV

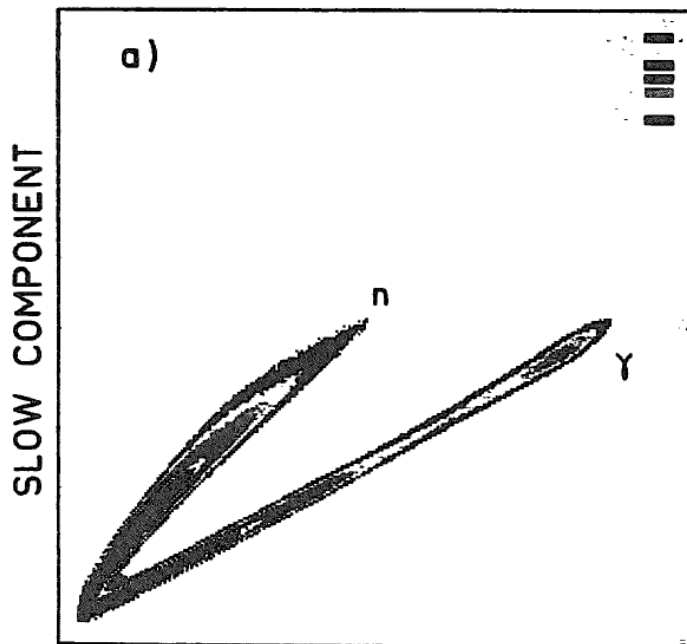


n – gamma discrimination at different
energy gates

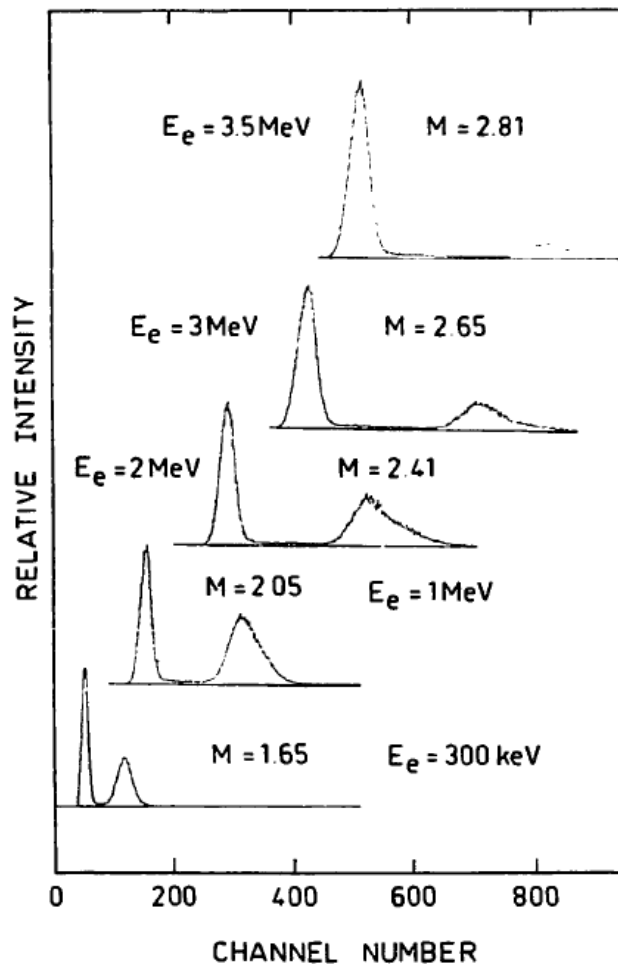


Charge comparison - optimization

Demon detector:
BC501A Ø16 cm x 20 cm
at XP45212B PMT



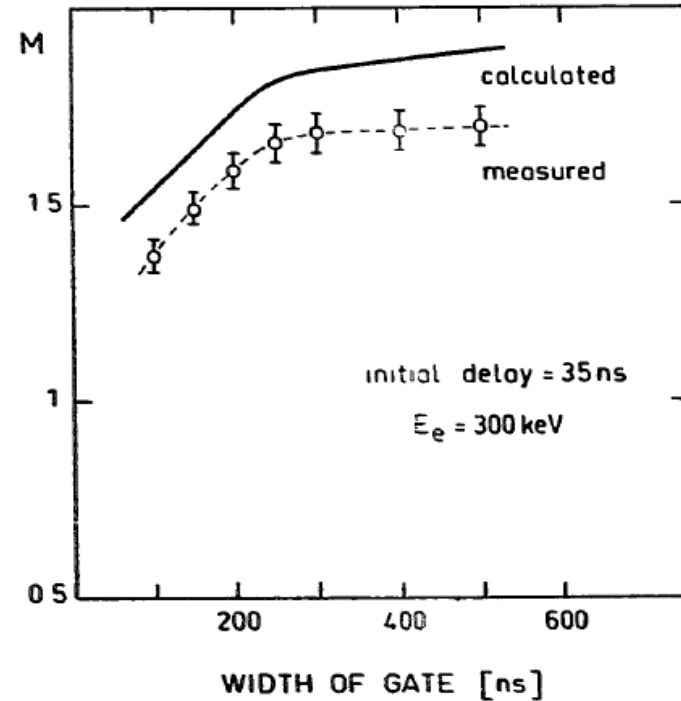
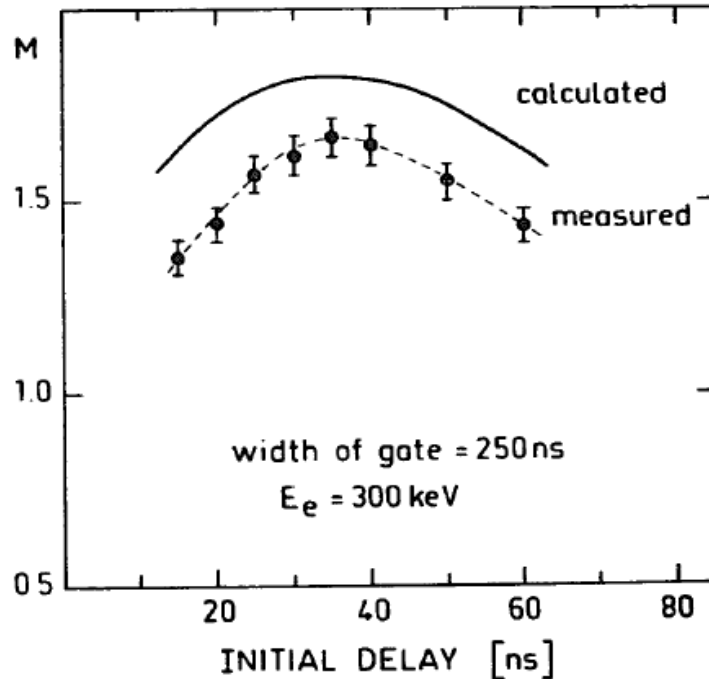
$$N_{\text{phe}} = 1100 \text{ phe/MeV}_{ee}$$





Charge comparison - optimization

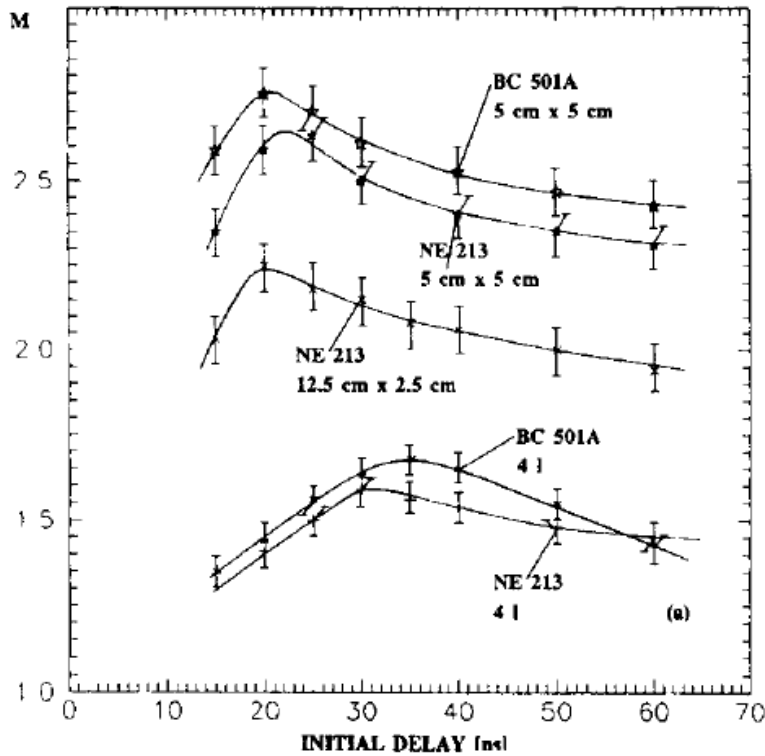
Demon detector:
BC501A Ø16 cm x 20 cm
at XP45212B PMT



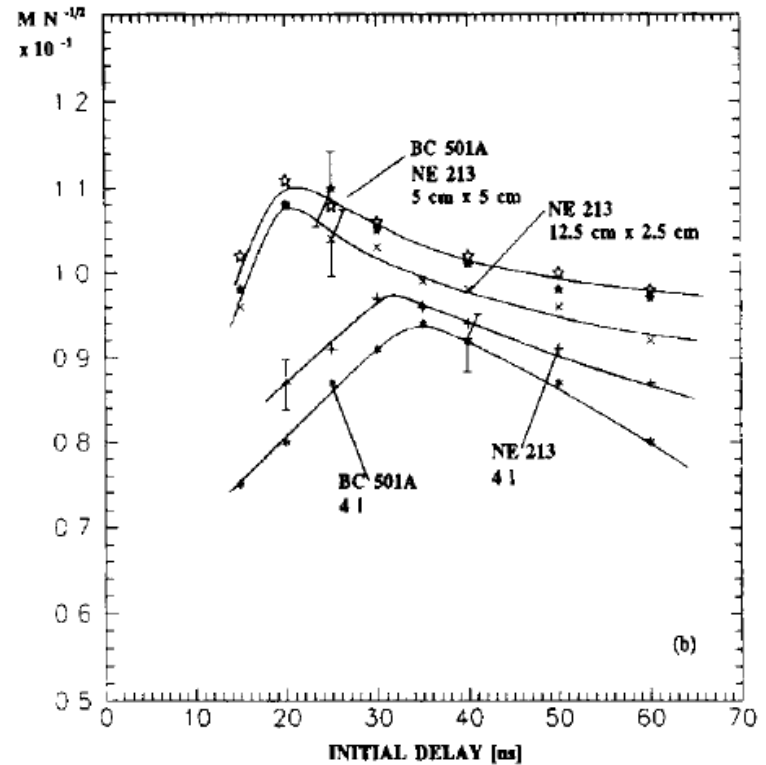
PHE number = 1090 ± 50 phe/MeV_{ee}



Charge comparison - optimization



M factor at 300 keV_{ee}

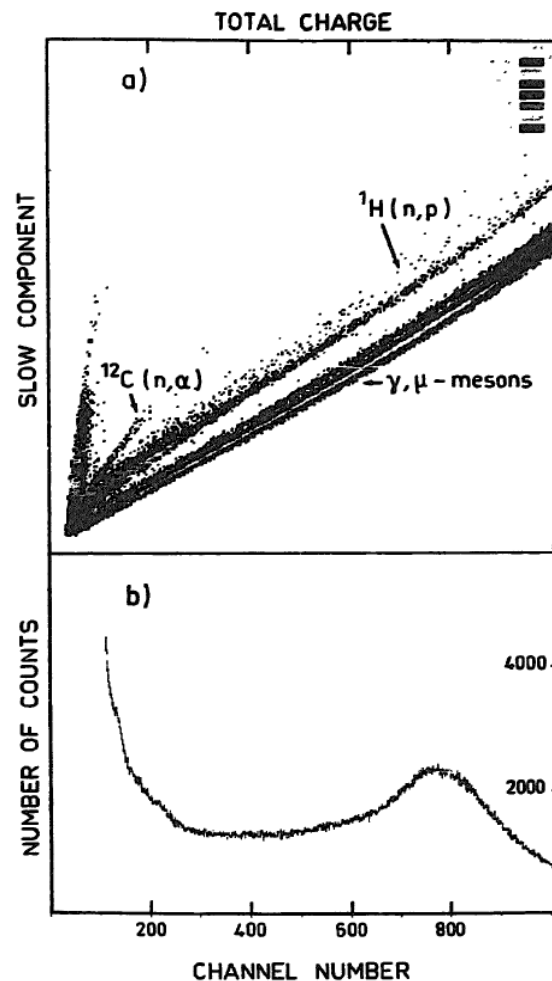


M factor normalized to the PHE number



Atmospheric neutrons

- Detector BC501A, $\varnothing 16$ cm x 20 cm
- Test in the laboratory in Strasbourg,
- Energy of neutrons up to 50 MeV
- Intensity of about 0.1-0.3% in relation to meson component
- Energy spectrum of mesons, straggling peak at about 30 MeV





Detection of high energy neutrons

Monoenergetic neutrons of **56 MeV** detected in BC501A:

A – gamma rays

B – protons, not fully stopped

C – recoil protons

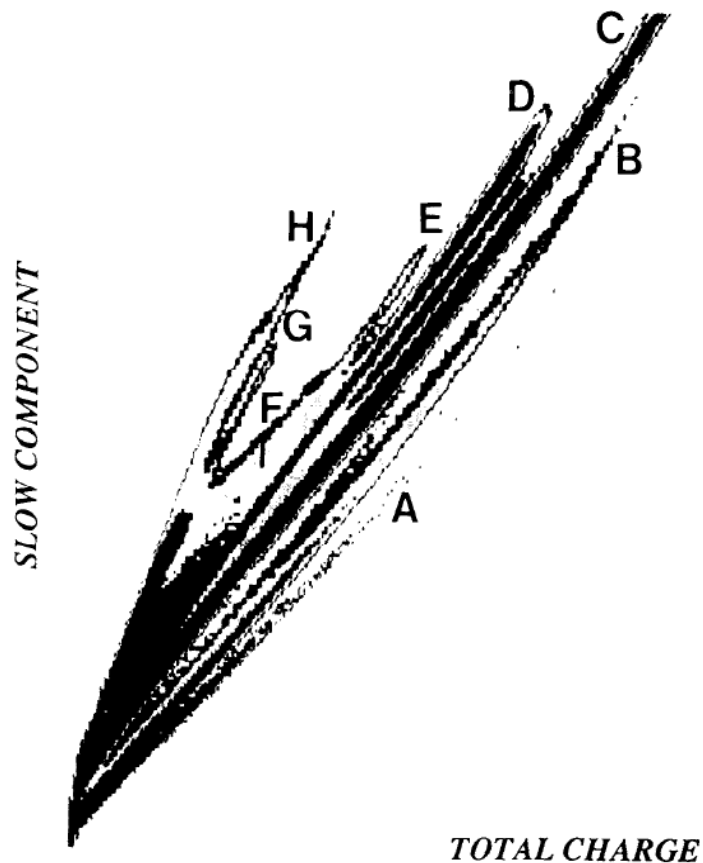
D – Deuterons

E – Tritons

F – Tritons and alphas

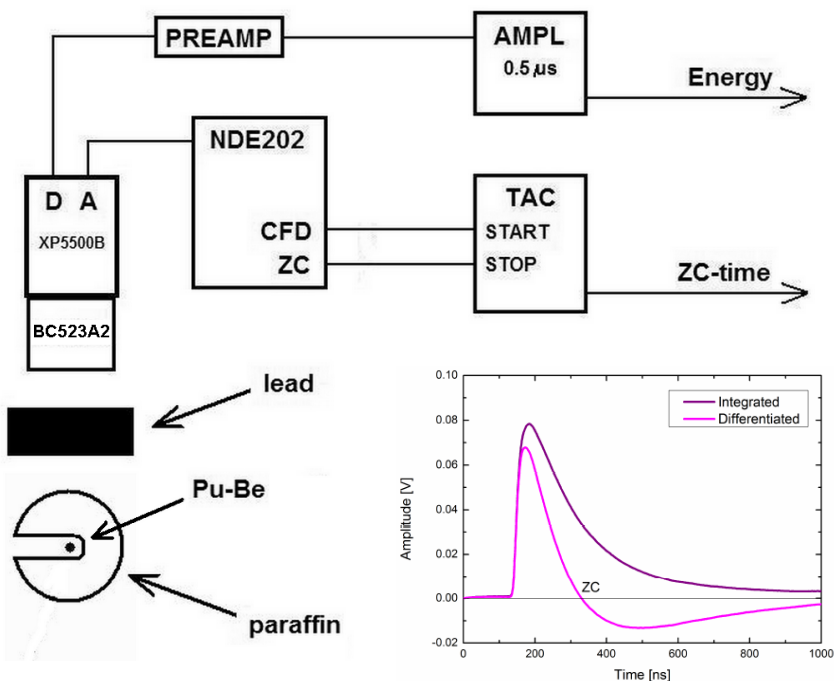
G – ^3He

H – alpha particles

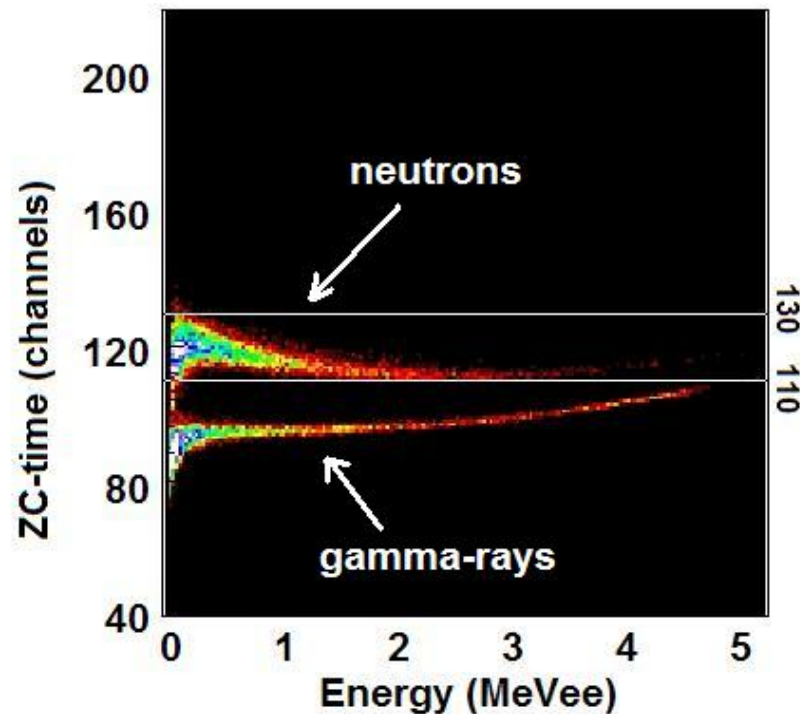




Pulse shape discrimination by Z/C method



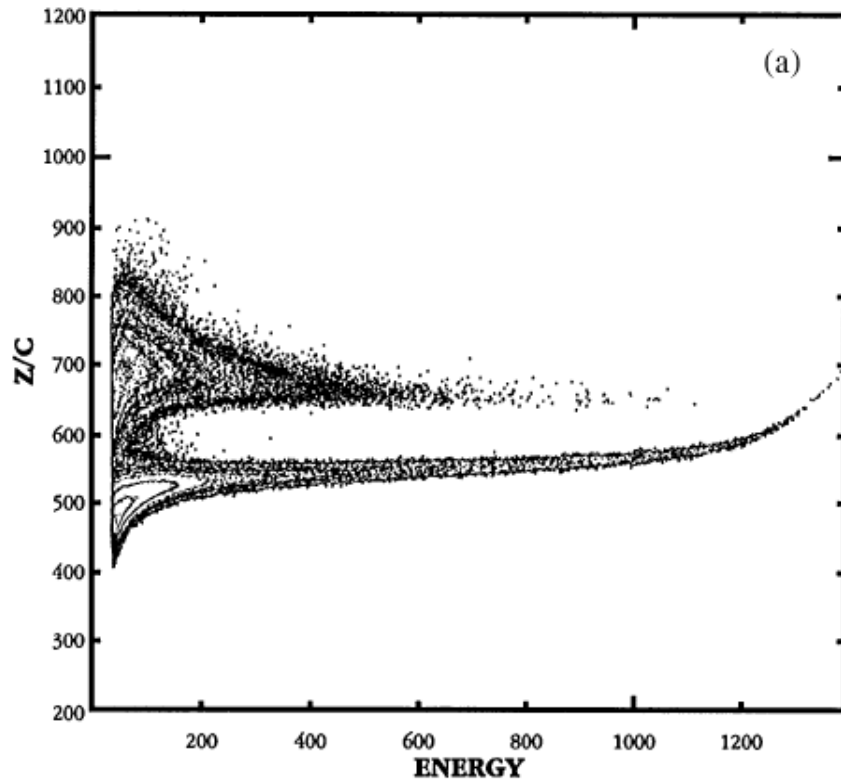
The experimental set up.
PSD by Z/C method.
Liquid scintillators coupled to slow
XP5500 or XP53X2 PMTs with high
quantum efficiency of about 35%!!!



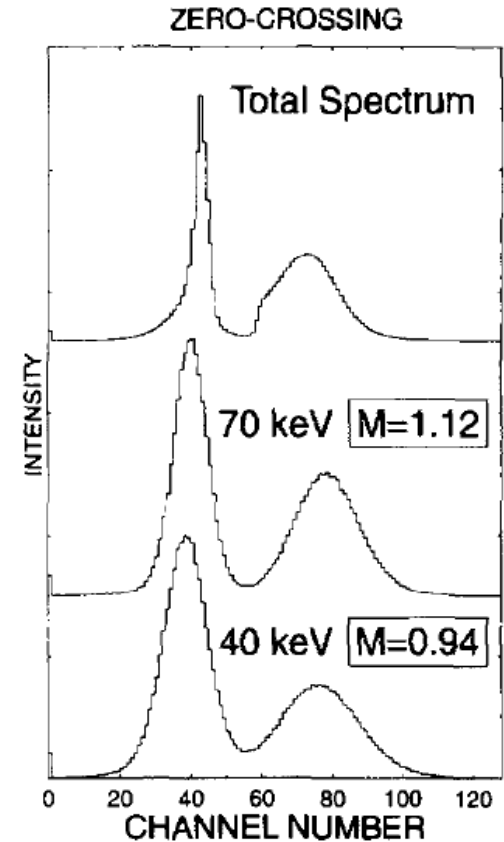
A 2D plot of ZC time vs. pulse height
measured with an unloaded EJ309 under
irradiation of a Pu-Be source



Pulse shape discrimination by Z/C method



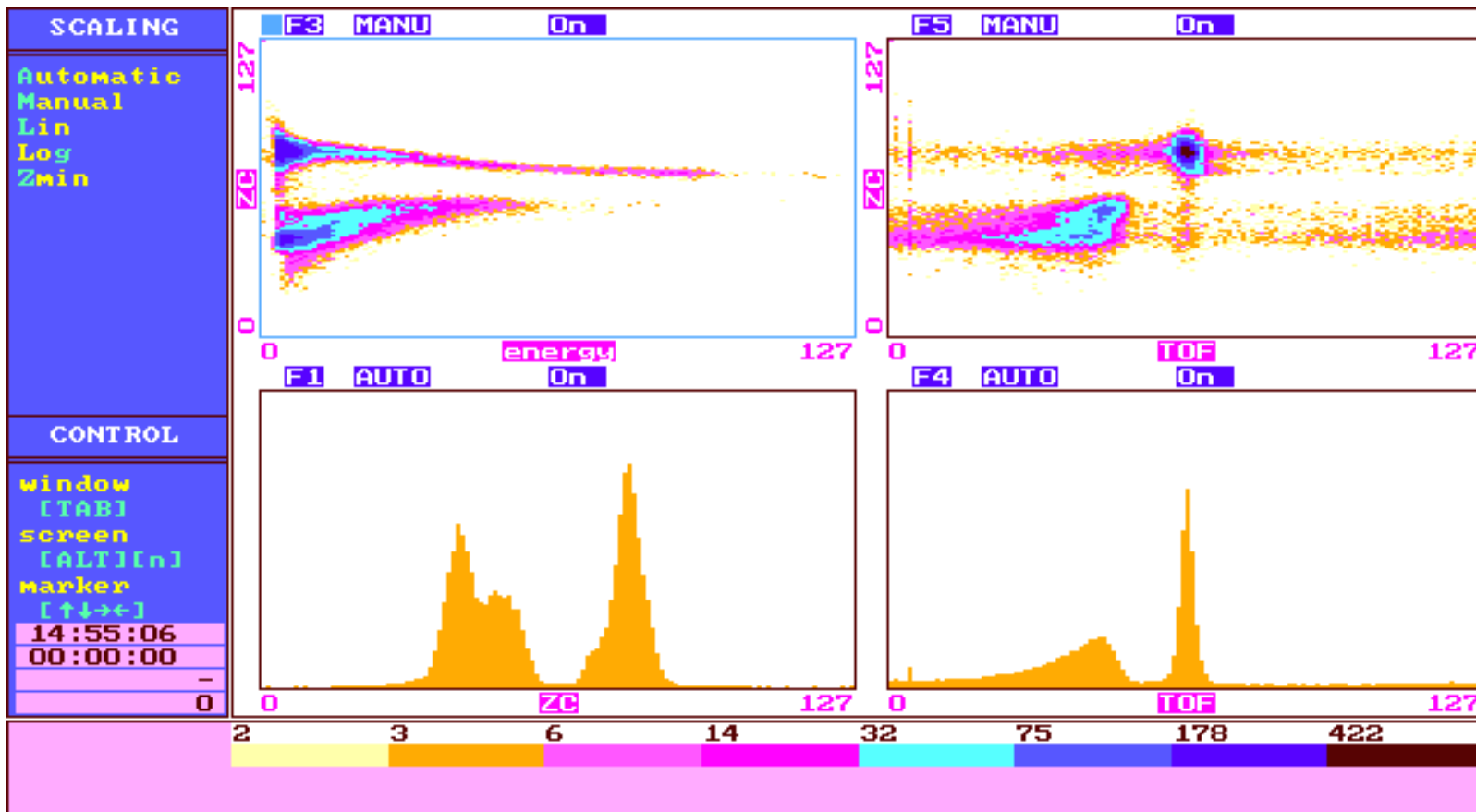
EUROBALL BC501A detector



5"x5" BC501A detector



Pulse shape discrimination by Z/C method

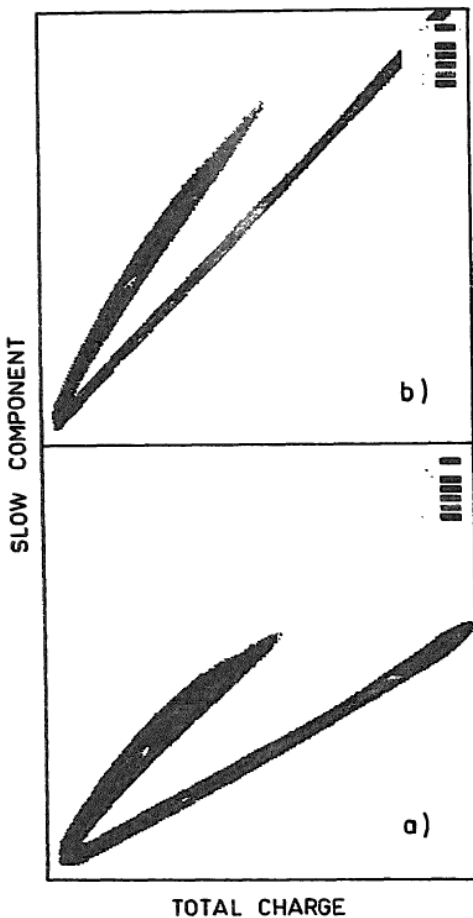


5"x5" BC501A, XP4512B PMT, EUROBALL neutron electronics, Pu-Be source

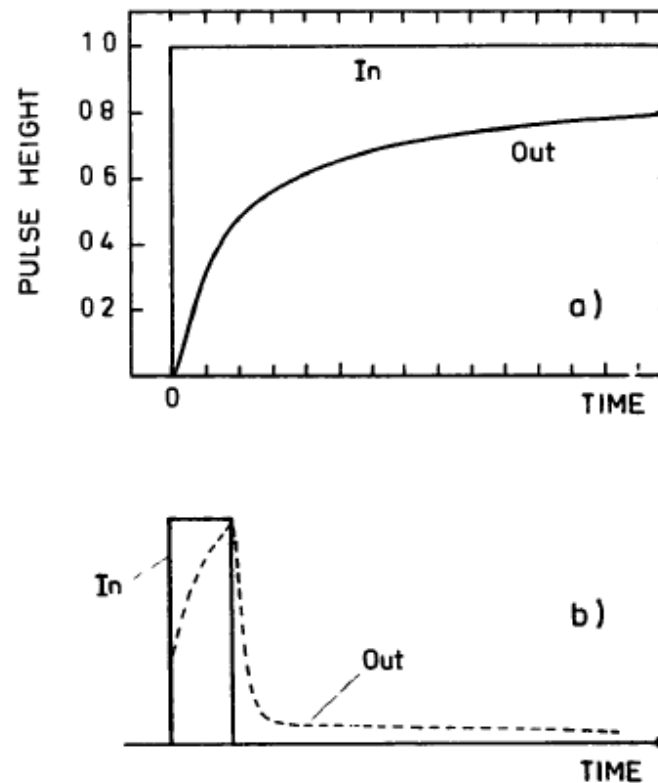


Input cable length effect

40 m
cable



2 m
cable



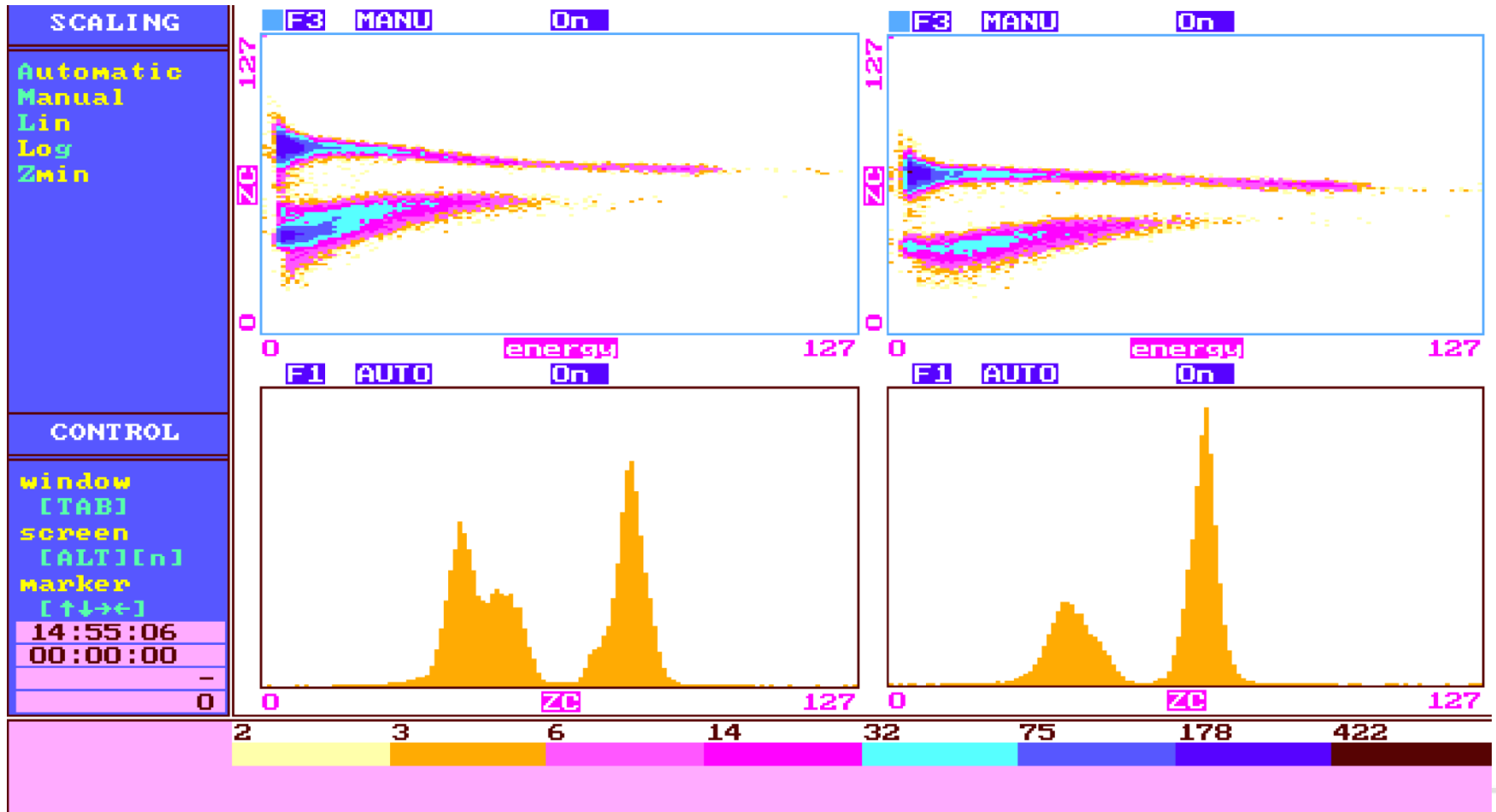
Distortion introduced by a long delay cable on the input pulse of the step function form (a), and on the short rectangular pulse (b).



Input cable length effect

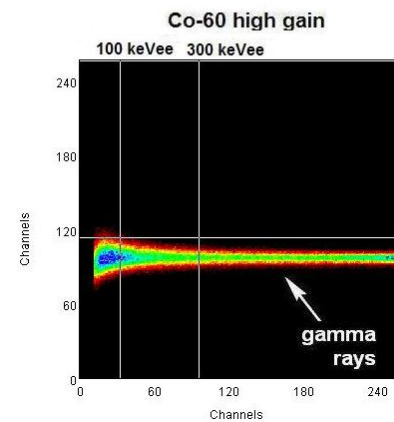
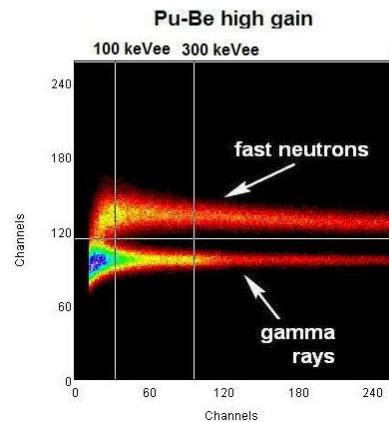
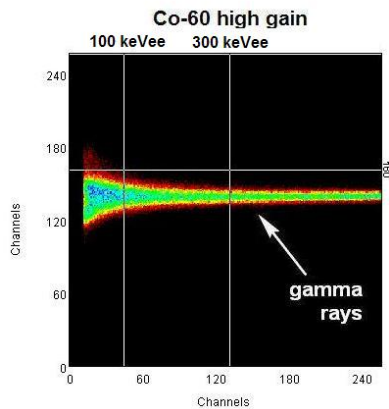
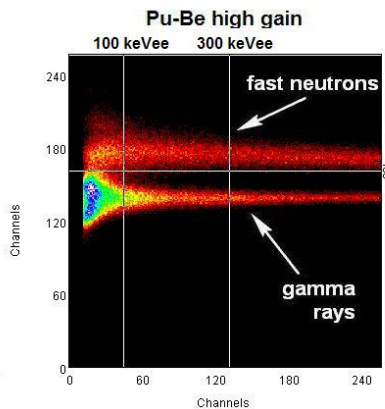
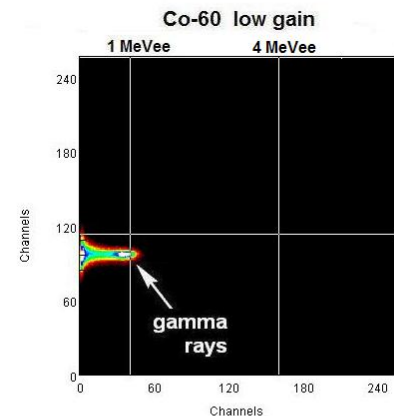
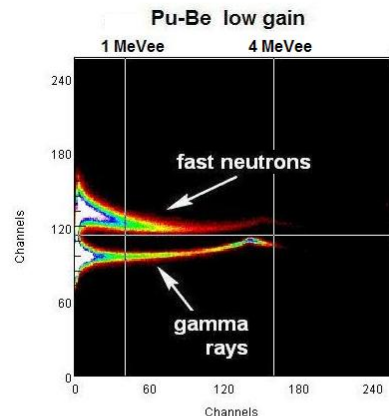
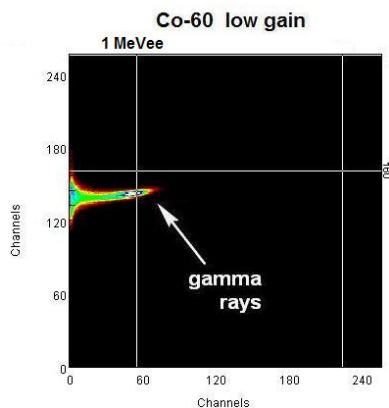
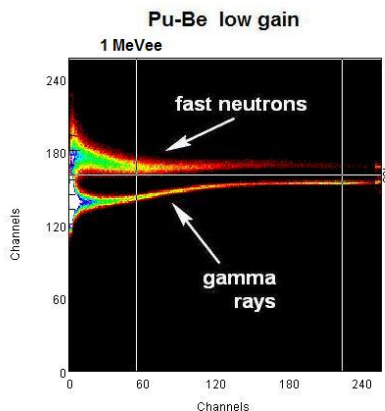
2 m cable

25 m cable





PMT comparison



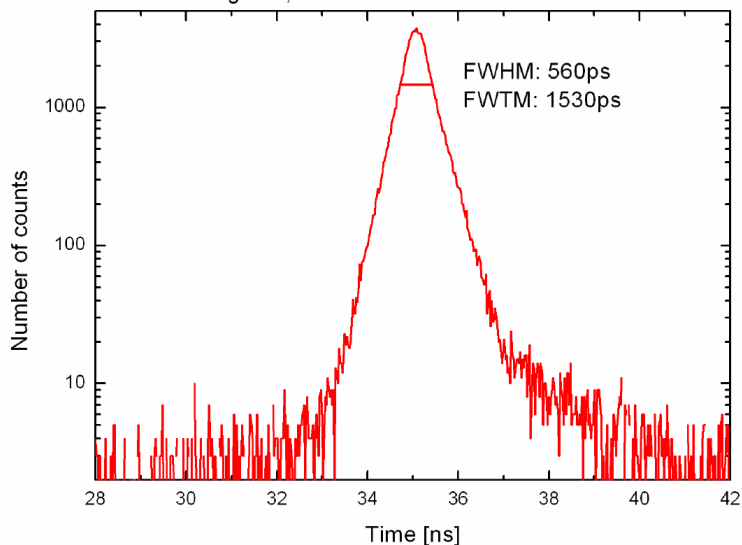
XP4512B,
PHE number = 1500 ± 50 phe/MeV

ETL9390,
PHE number = 3200 ± 100 phe/MeV

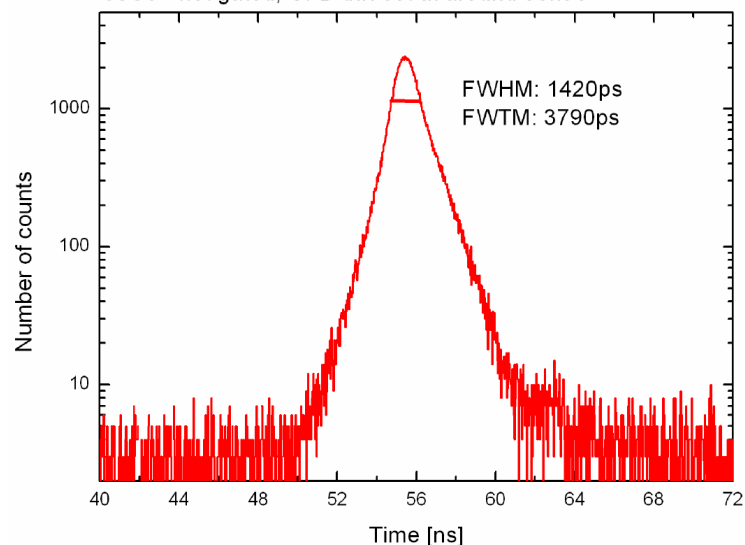


PMT comparison - timing

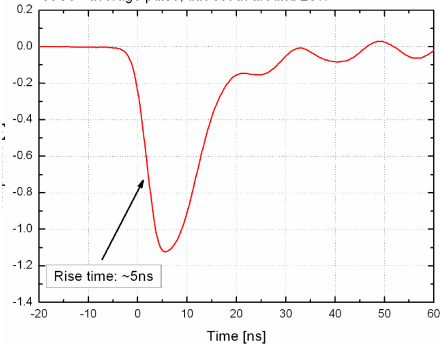
XP4512 + EJ309 5"
60Co - not gated, CFD thr. set at around 50keV



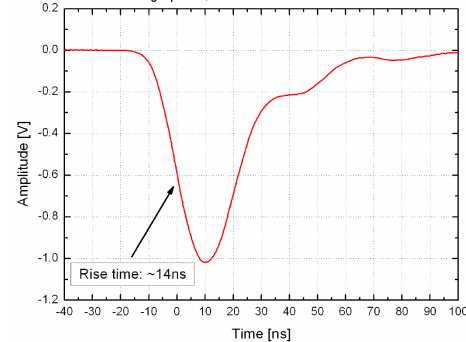
ETL9390 + EJ309 5"
60Co - not gated, CFD thr. set at around 50keV



XP4512 + EJ309 5"
60Co - average pulse, thr. set at around 20%



ETL9390 + EJ309 5"
60Co - average pulse, thr. set at around 50%





New organic scintillators

Scintillator	^{10}B loaded	Flash Point	Density [g/cm ³]	H:C Ratio	Light Yield ^{a)} [phe/MeV]
BC501A	NO	24 °C	0.87	1.21	1620
EJ301	NO	26 °C	0.87	1.21	2540
EJ309	NO	144 °C	0.96	1.25	2600
BC523A	YES, 4.4%	-8 °C	0.92	1.74	1540
BC523A2	YES, 2%	-8 °C	0.92	not known	2450
EJ339A2	YES, 2.5%	-8 °C	0.92	1.49	2190
EJ309B5	YES, 4.6%	144 °C	0.96	1.31	1850

BC*** – scintillators of Saint-Gobain Crystals

EJ*** - scintillators of Eljen/Scionix

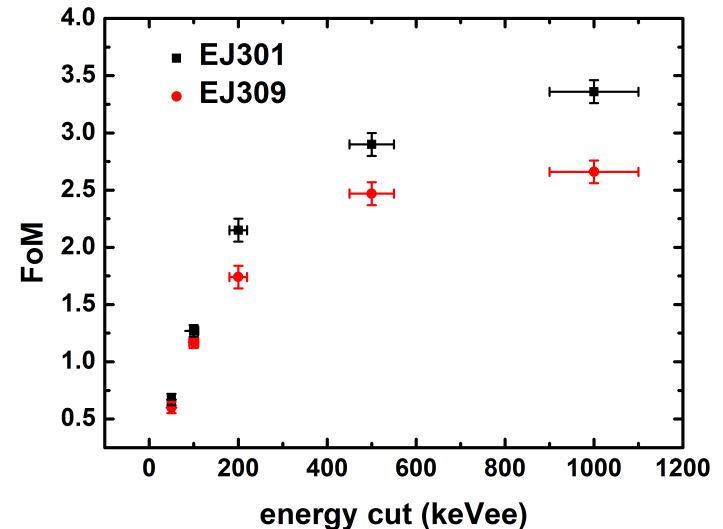
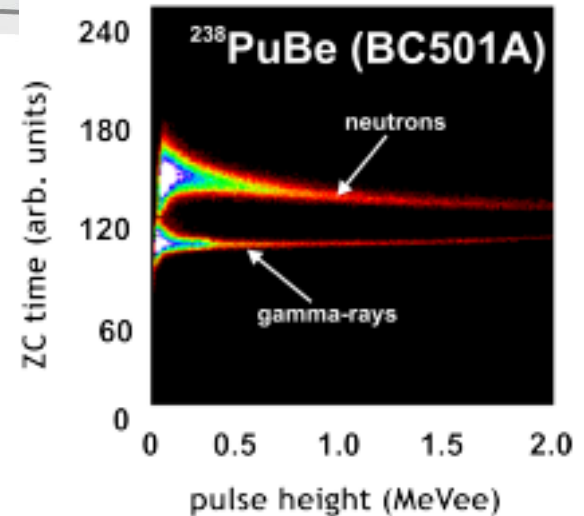
2" x 2" liquid cells at XP5500 PMT, QE = 35%

BC501A and BC523A – old!!!



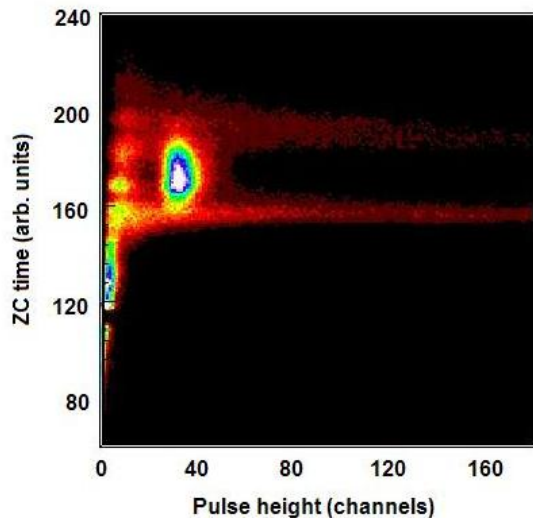
EJ309 vs EJ301 liquid scintillators

- EJ301 – BC501A – NE213 xylene-based liquid scintillator, low flash point at 20 °C, toxic, light output – 12000 ph/MeV
- EJ309 inorganic solvent, high flash point of 144 °C, low vapor pressure, low chemical toxicity, light output – 12300 ph/MeV, Slightly poorer n/gamma PSD

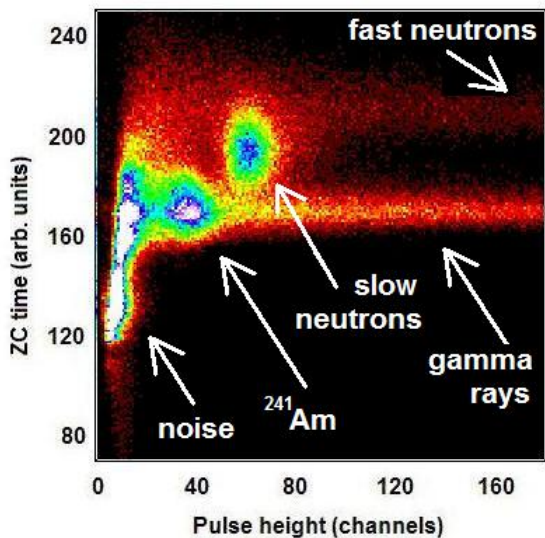
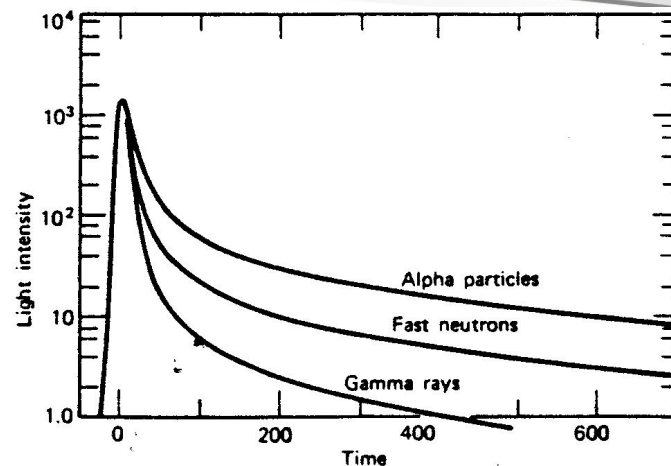




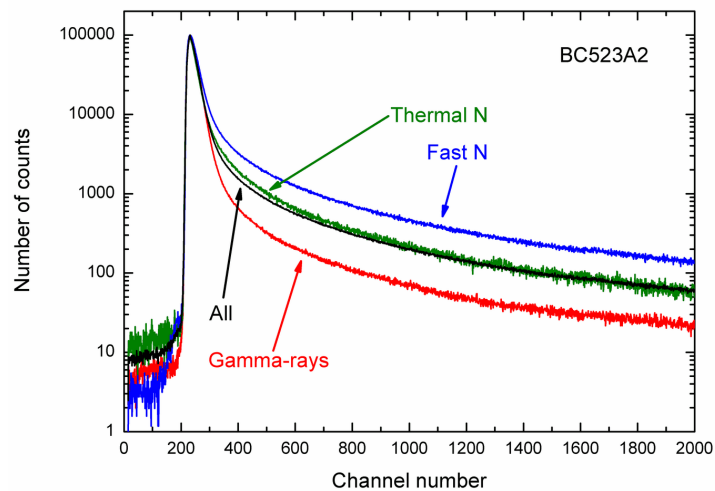
n/γ discrimination with B-10 loaded liquid scintillators



BC523A2

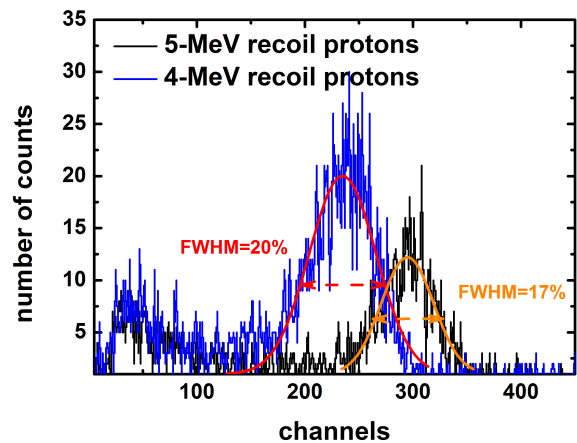
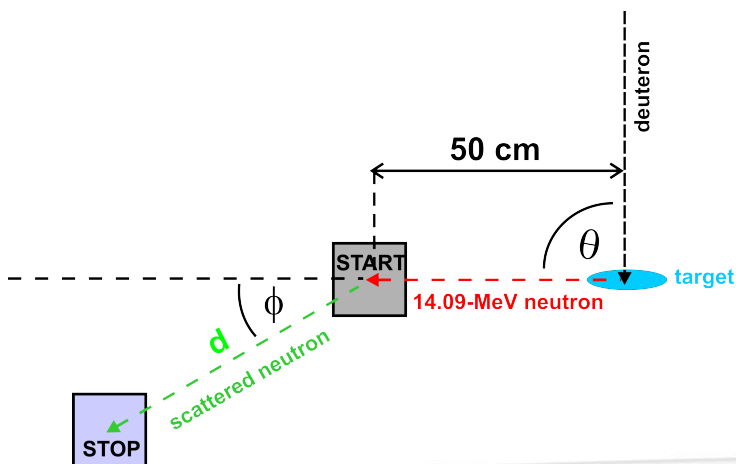
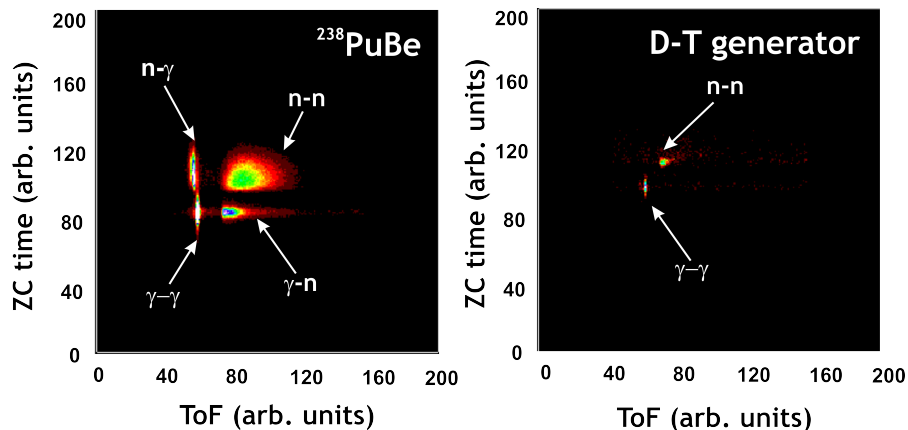
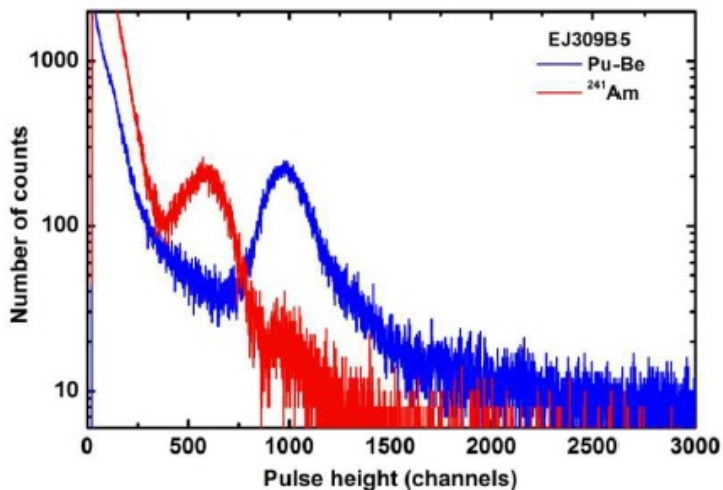


EJ309B5



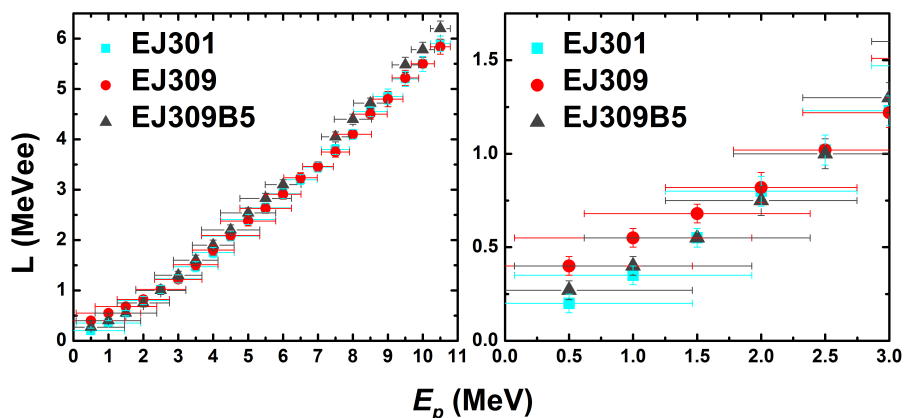


EJ309 vs. EJ301 calibration

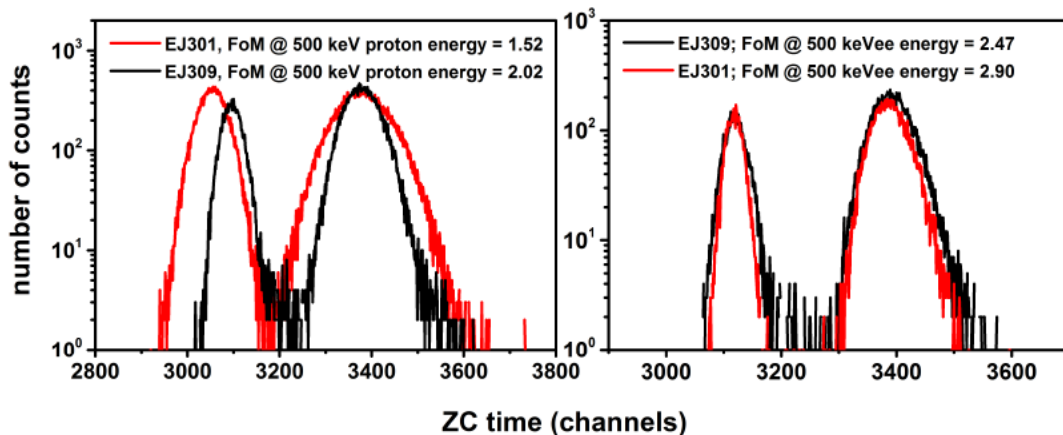




EJ309 vs. EJ301 calibration



- Calibration curves of the light output vs recoil proton energy as measured with EJ301, EJ309 and EJ309B5.
- Note a largest light output of EJ309 at low energies reflecting a weaker quenching of light due to protons.



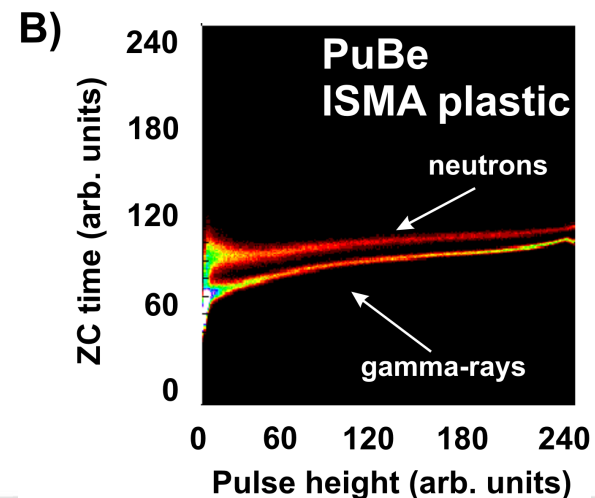
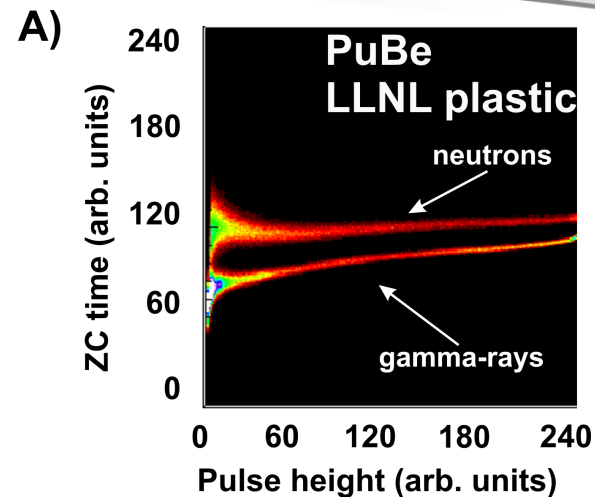
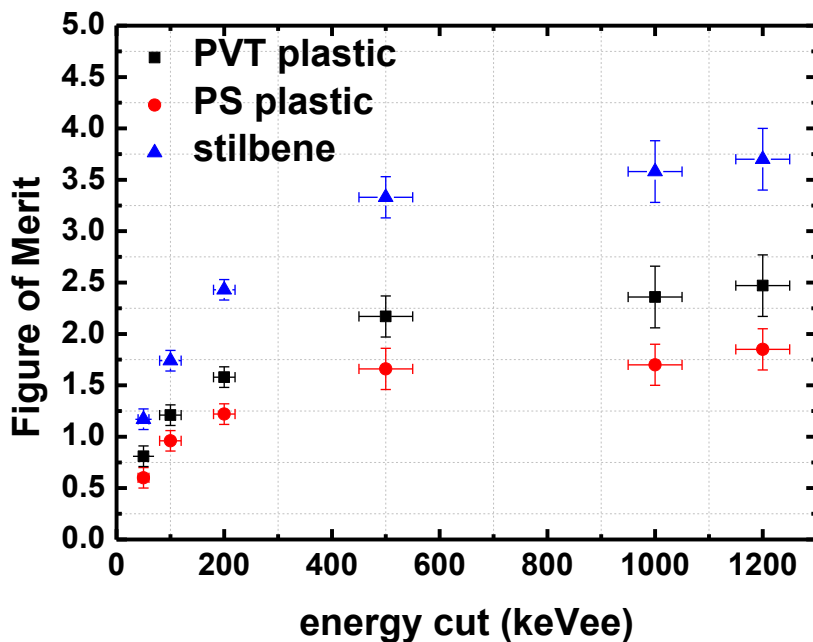
ZC time spectra for a 500 keV energy lost by protons and recoil electrons in EJ301 and EJ309.



Plastic scintillators – small samples

sample	photoelectron yield (phe/MeV)
PVT plastic	3800 ± 200
PS plastic	3100 ± 200
stilbene	4200 ± 200

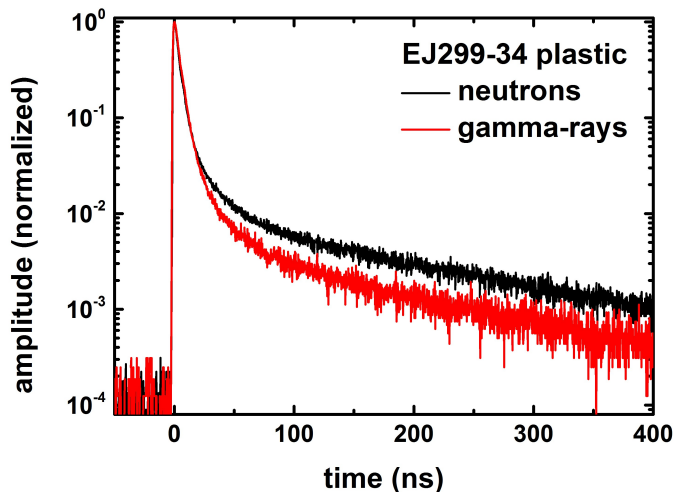
QE = 35%





Plastic scintillators – 2" x 2" samples

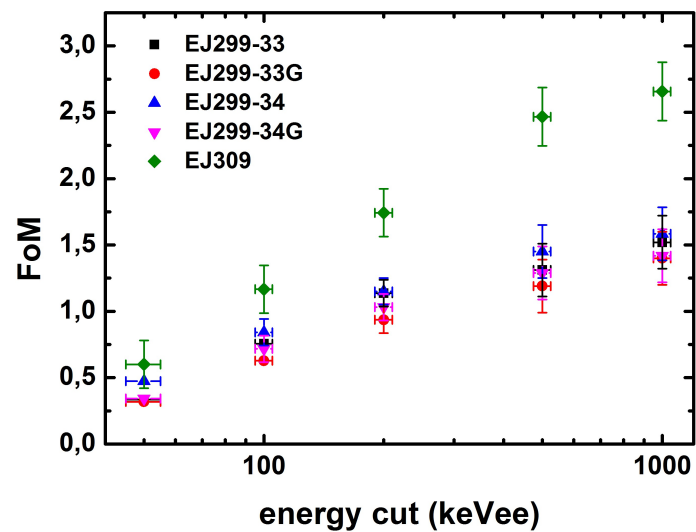
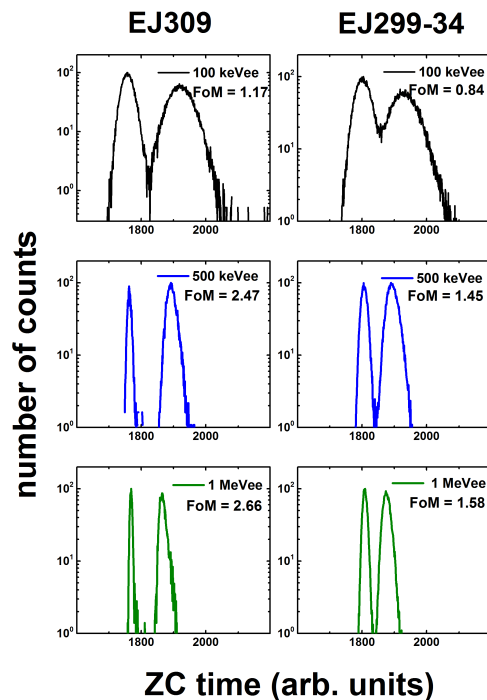
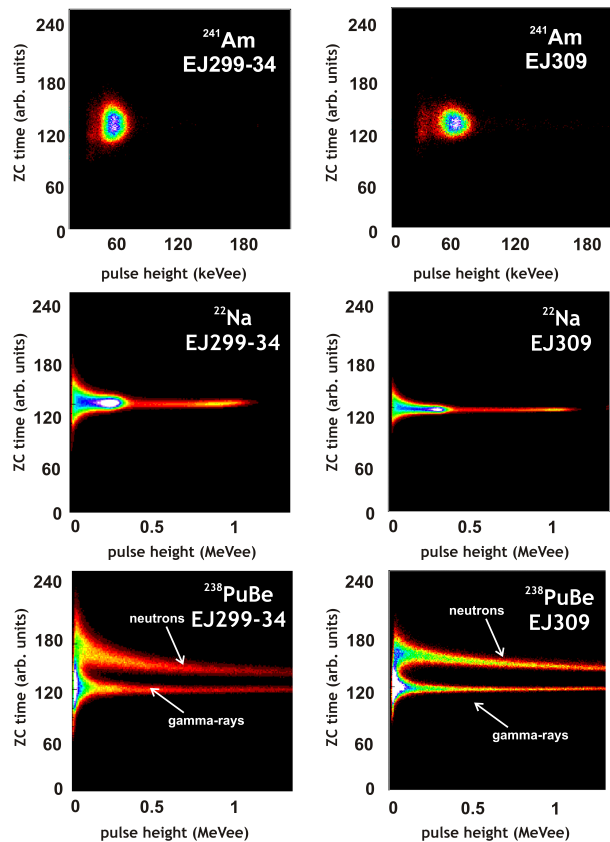
scintillator	EJ299-33	EJ309
light Output, % Anthracene	56	75
scintillation Efficiency, photons/1 MeV e ⁻	8600	11500
wavelength of maximum emission (nm)	420	424
number of H atoms per cm ³	5.13×10^{22}	5.46×10^{22}
number of C atoms per cm ³	4.86×10^{22}	4.37×10^{22}
number of electrons per cm ³	3.55×10^{23}	3.17×10^{23}
density (g/cm ³)	1.08	0.964



scintillator	photoelectron yield (phe/MeV)
EJ309	2600 ± 50
EJ299-34	2500 ± 50
EJ299-34G	2100 ± 50
EJ299-33	1950 ± 30
EJ299-33G	1600 ± 30

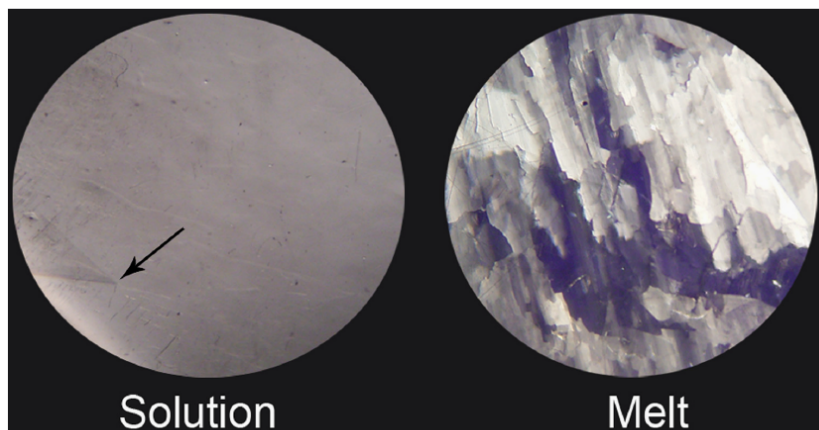


Plastic scintillators – 2" x 2" samples

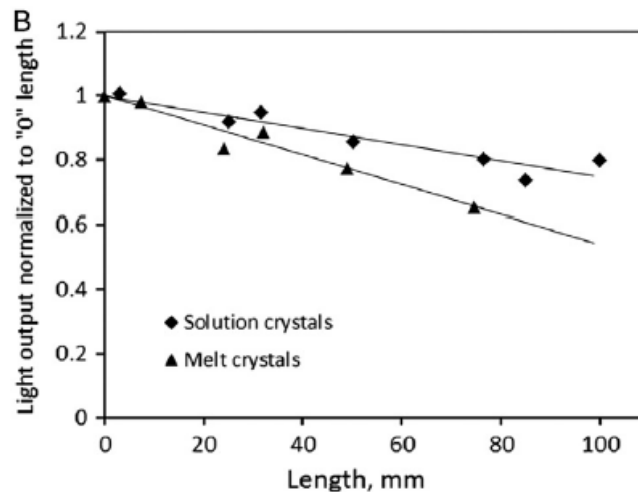
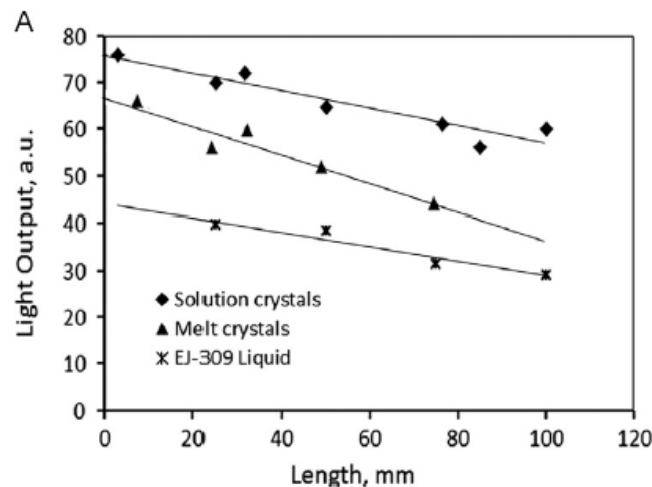




New stilbene – grown from solution

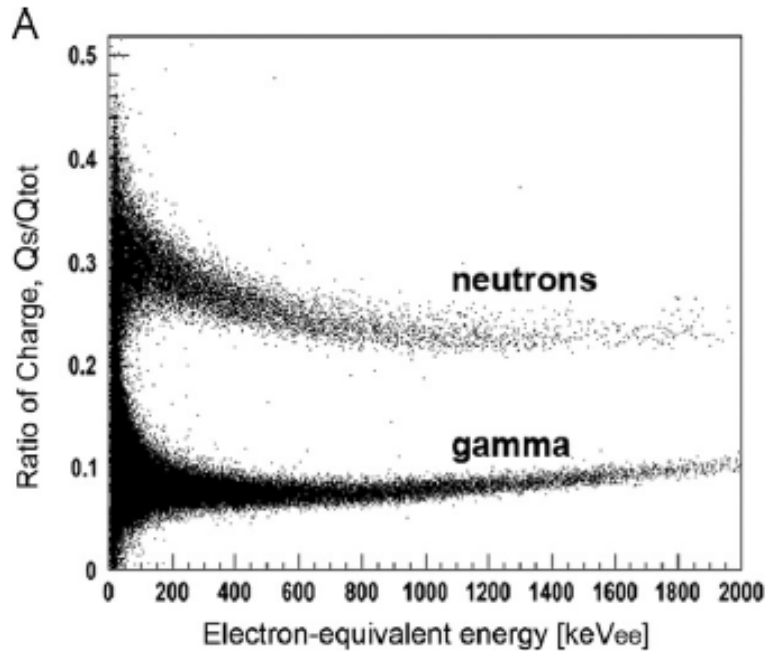


tested samples: $\varnothing 2''$, height up to 4''

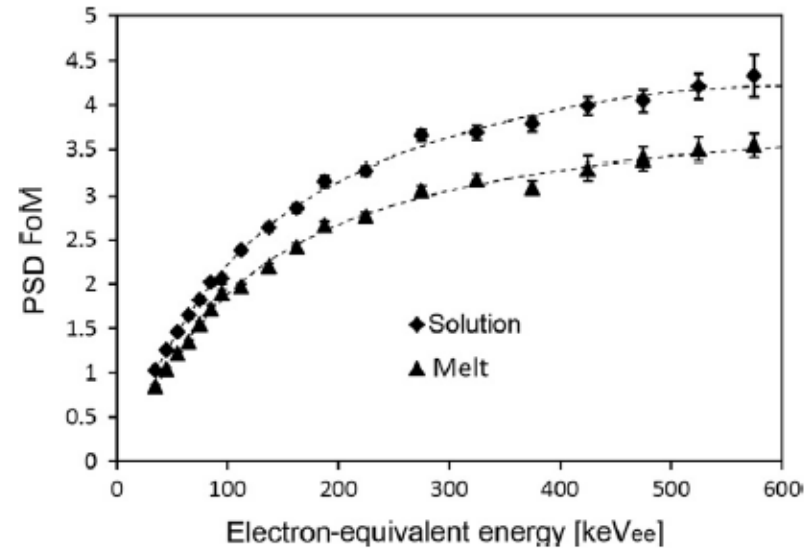
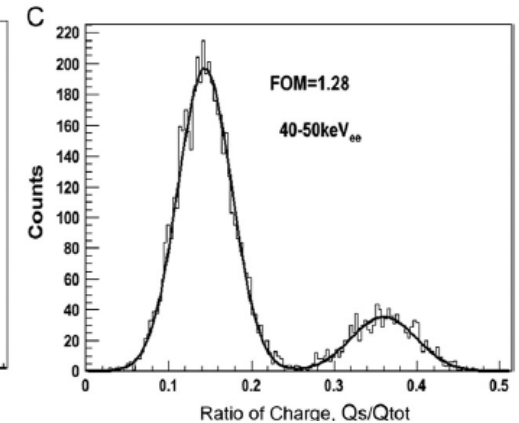
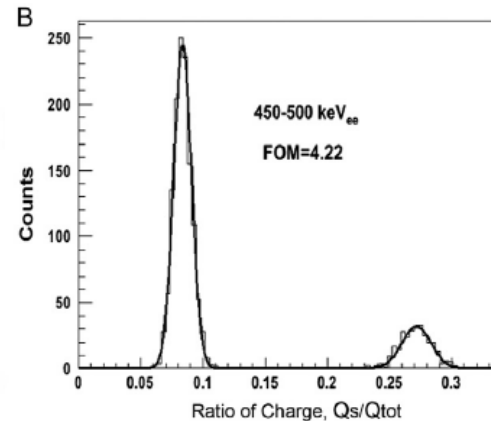




New stilbene – grown from solution

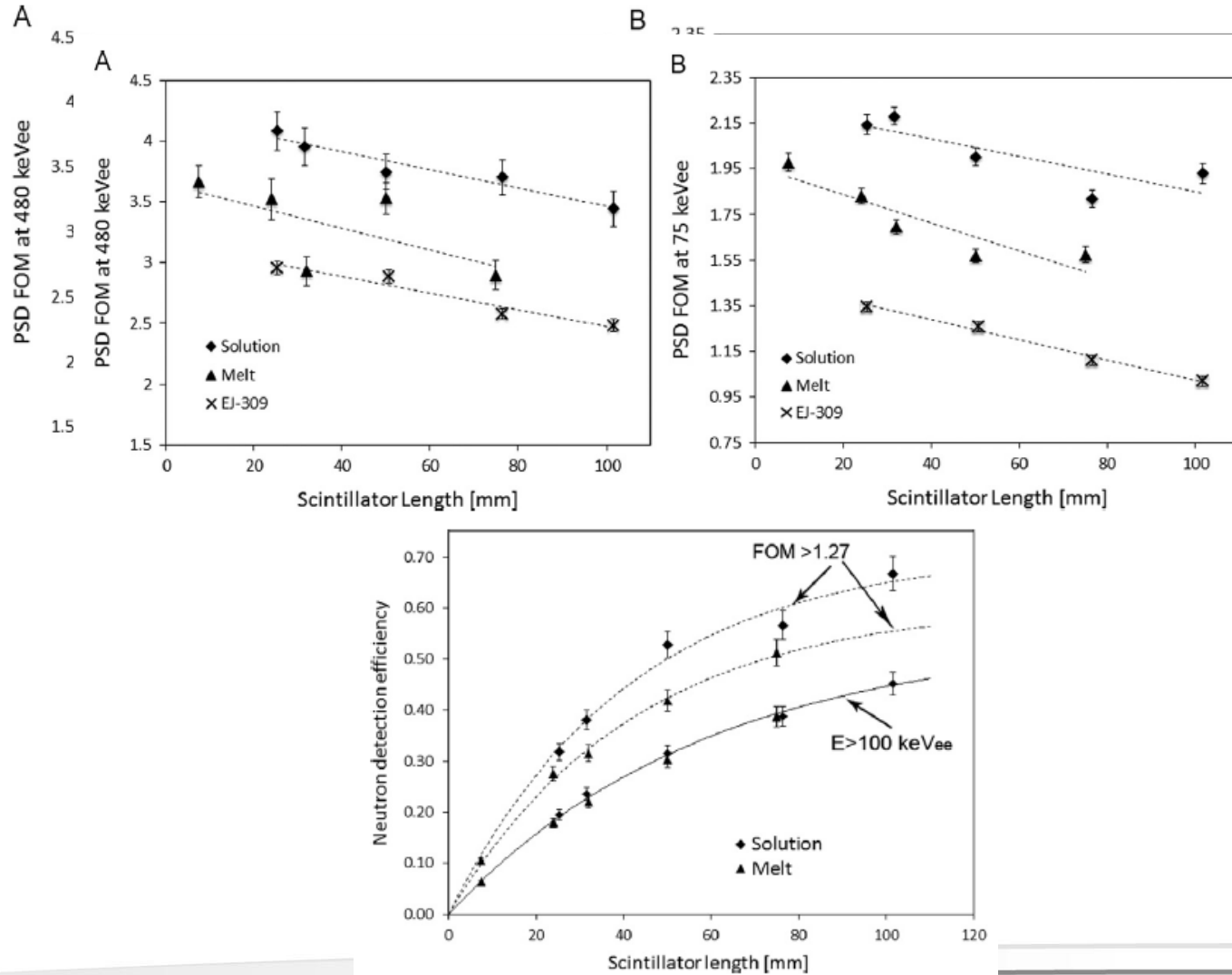


Ø51 mm x 75 mm





New stilbene – grown from solution

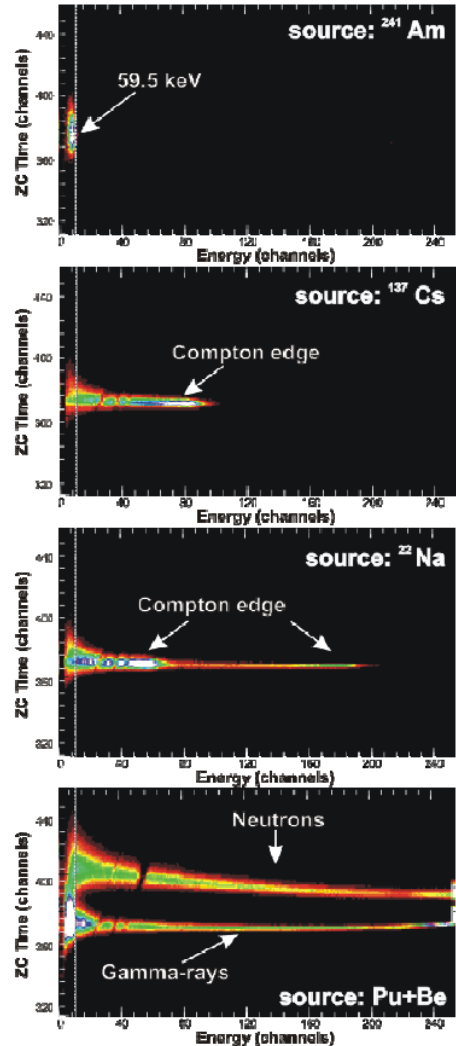




Stilbene and SiPMs in neutron detection



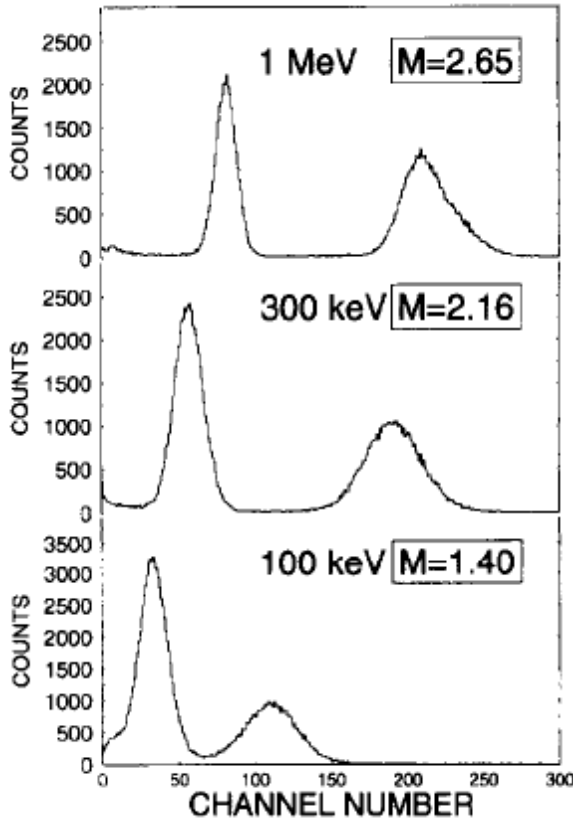
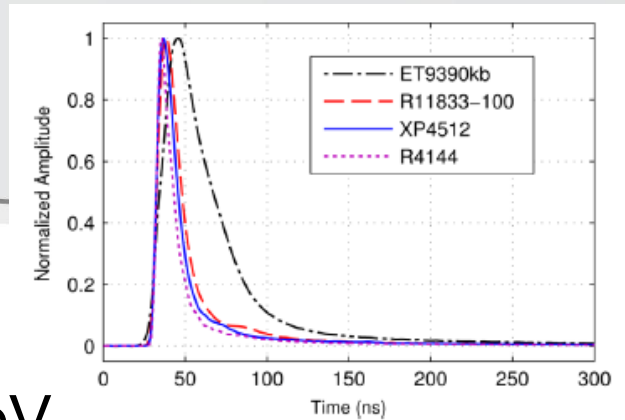
Stilbene: $\varnothing 25 \text{ mm} \times 25 \text{ mm}$
 MPPC array $12 \text{ mm} \times 12 \text{ mm}$



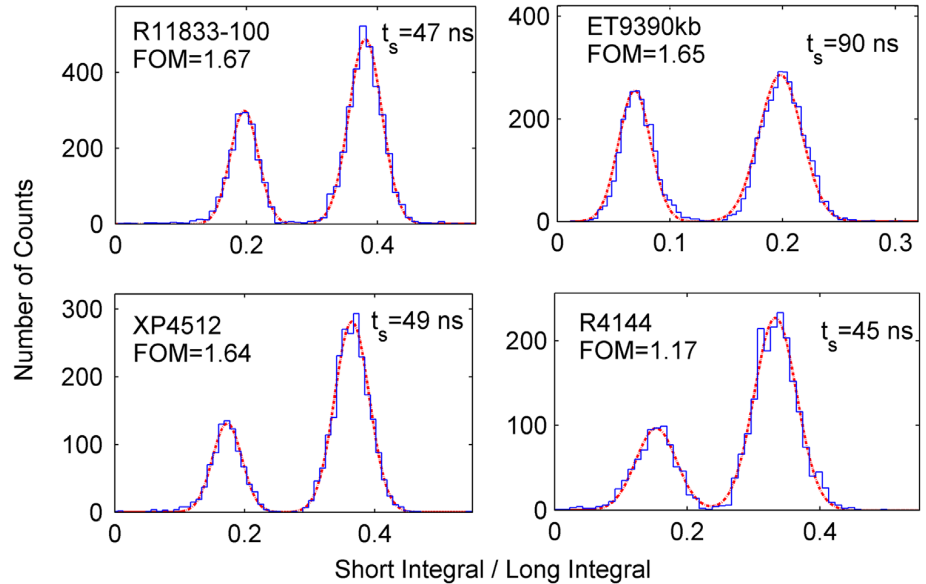
Phe/MeV	2240 ± 220
FoM 100 keVee	1.16 ± 0.06
FoM 300 keVee	2.13 ± 0.11
FoM 500 keVee	2.86 ± 0.14
FoM 1 MeVee	3.41 ± 0.17



Analog vs. Digital PSD



300 keV



5" x 5" BC501A at XP4512B
ZC analog method

5" x 5" BC501A at different PMTs
Digital charge comparison method

X.L. Luo, et al., NIM A767(2014)83



Limitations of the digital method

Summary of FOM results for the digitizers under investigation.

Digitizer model	ENOB for 1-V range	Sampling frequency (MHz)	FOM _{FWHM}	FOM _{valley}
V1720	9.14	250	1.26	1.06
V1751	9.04	1000	1.33	1.17
V1751	9.04	2000	1.33	1.34
Pixie-400	10.2	400	1.46	3.00

Energy threshold = 80 keV

ENOB – effective number of bits

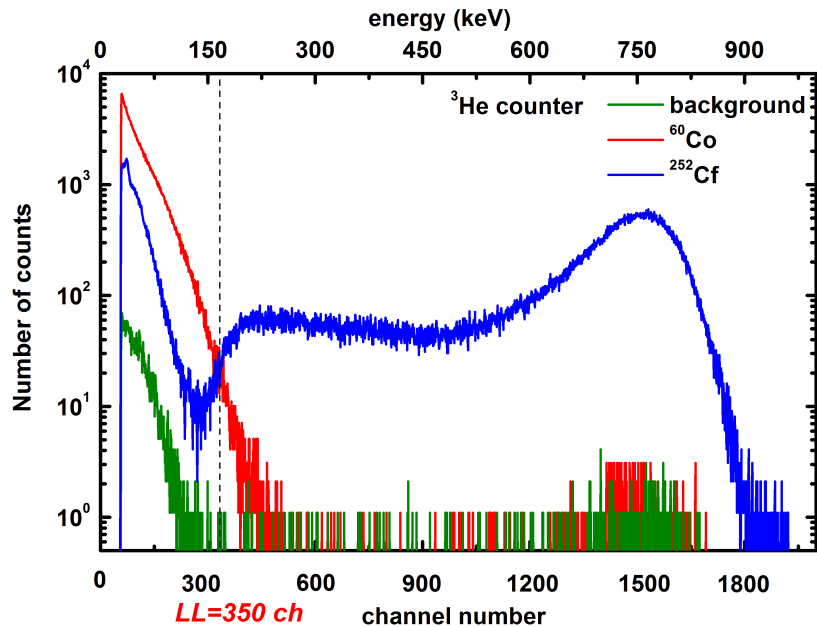


Further questions

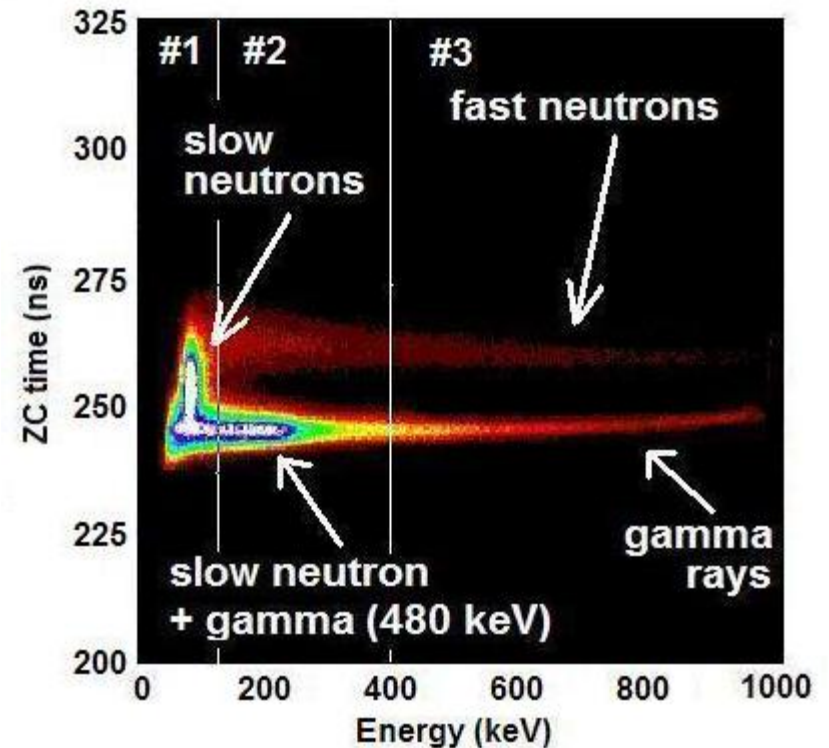
- Detection efficiency in comparison to He-3 detector
- Rejection of gamma rays in comparison to He-3 detector



Detection efficiency



Energy spectra measured with He-3 counter under irradiation of a ^{60}Co source, ^{252}Cf and background.



A 2D plot of ZC time vs. energy from EJ309B5 irradiated with the ^{252}Cf source.



Detection efficiency

detector	Source moderator	B (s ⁻¹)	G (s ⁻¹)	N(s ⁻¹)	ϵ_n	ϵ_γ
He-3	-	0.55	0.59	12.8	0.20(2)	<10 ⁻⁶
He-3	+	0.55	0.59	13.0	0.23(3)	<10 ⁻⁶
EJ309B5	-	0.11	6.43	3.89	0.34(4)	2.6(9)×10 ⁻⁵
EJ309B5	+	0.11	6.43	2.74	0.27(3)	2.6(9)×10 ⁻⁵

He-3 detector 2" diameter x 40" long

EJ309B5 3" x 3" liquid cell



Detection efficiency

The improvement of performance of EJ309B5:

- **application of pile-up rejection circuit** in the n/ γ discrimination electronics (to reduce γ -ray pile-ups)
- **increase of a lead shield** (to reduce γ -rays yield)
- **reduction of the detector size** (for further reduction of pile-ups of γ -rays from intense γ -ray sources)
- application of copper shield between the detector and lead shield (to reduce X-rays yield from lead)
- improved slow neutrons discrimination from γ -rays in low energy region (ROI #1).

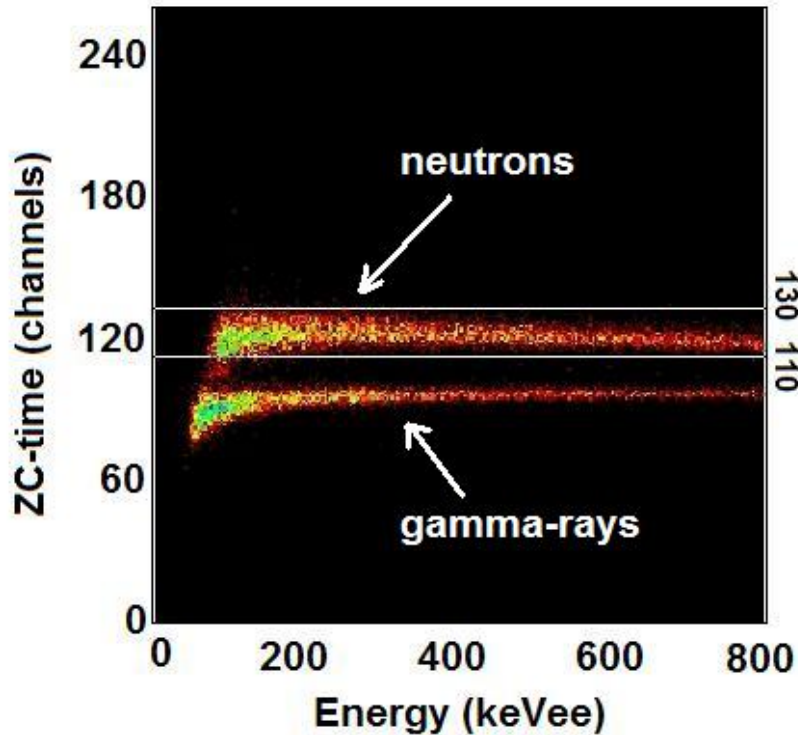


Suppression of gamma-ray sensitivity in liquid scintillators

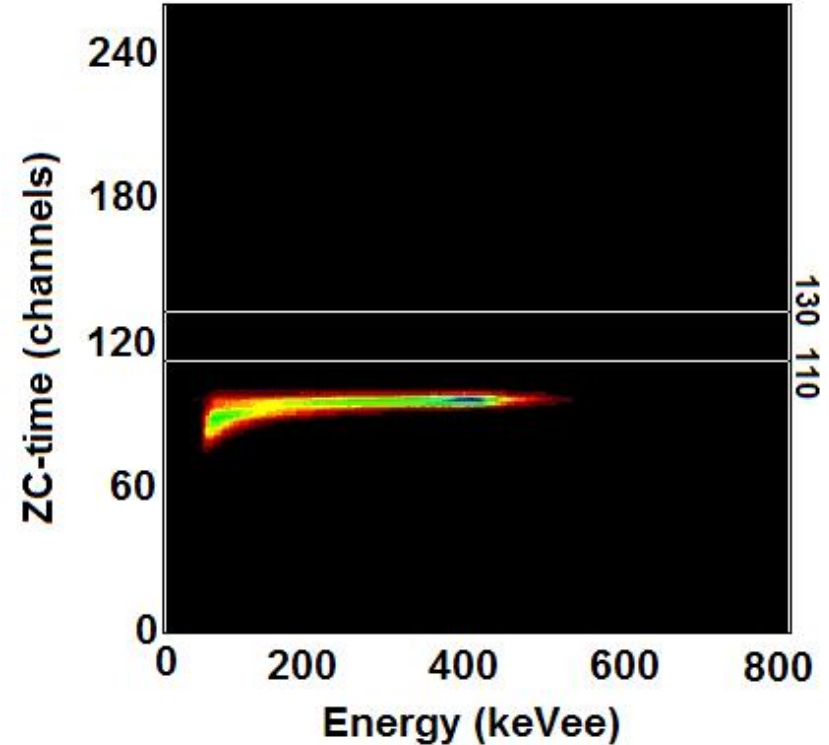
- 2" x 2" EJ309 liquid cell
- Z/C n- γ PSD with pile up rejection
- a strong field of gamma radiation from ^{137}Cs source, yielding 10 mR/h at the detector, as proposed in Neutron Detector Gamma Insensitivity Criteria
- Different shielding of neutron detector
- A comparison to a 2" x 20" He-3 counter surrounded with polyethylene shield.



Suppression of gamma-ray sensitivity in liquid scintillators



A 2D plot of ZC-time vs. Energy from EJ309 irradiated with the Pu-Be source.

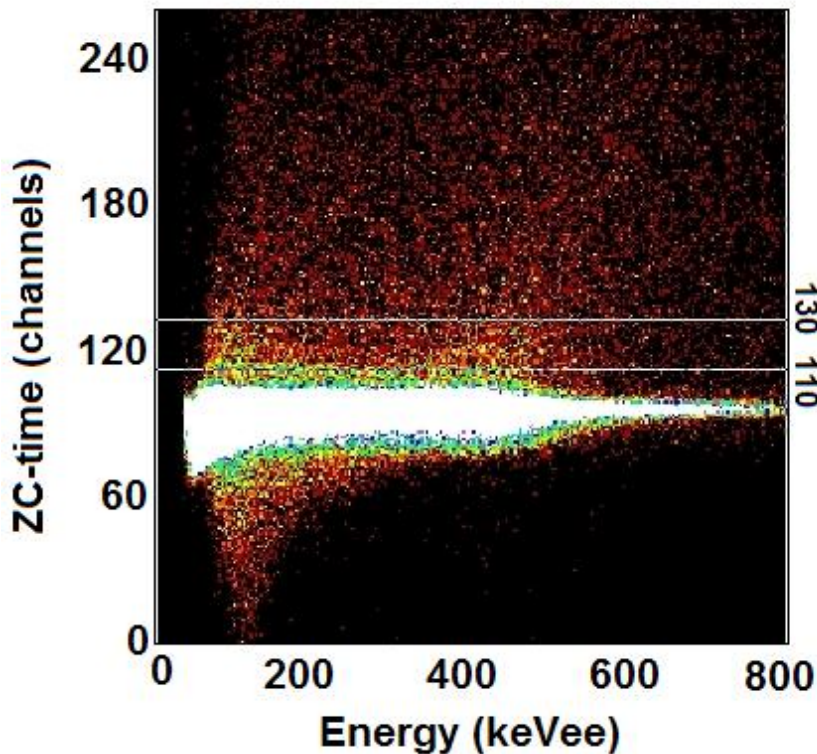


EJ309 irradiated with the ^{137}Cs source.

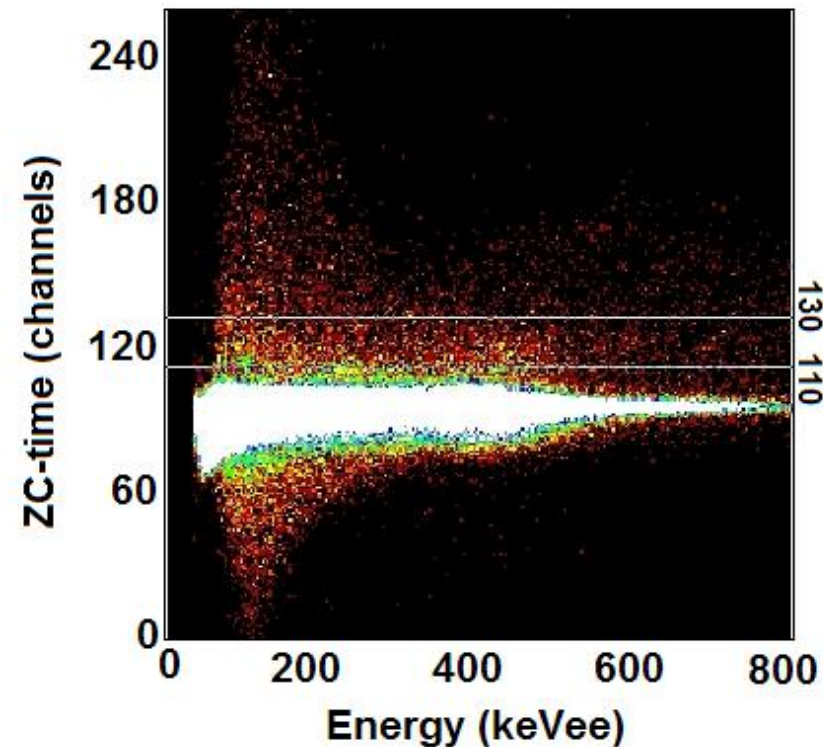


Suppression of gamma-ray sensitivity in liquid scintillators

no pile-up rejection



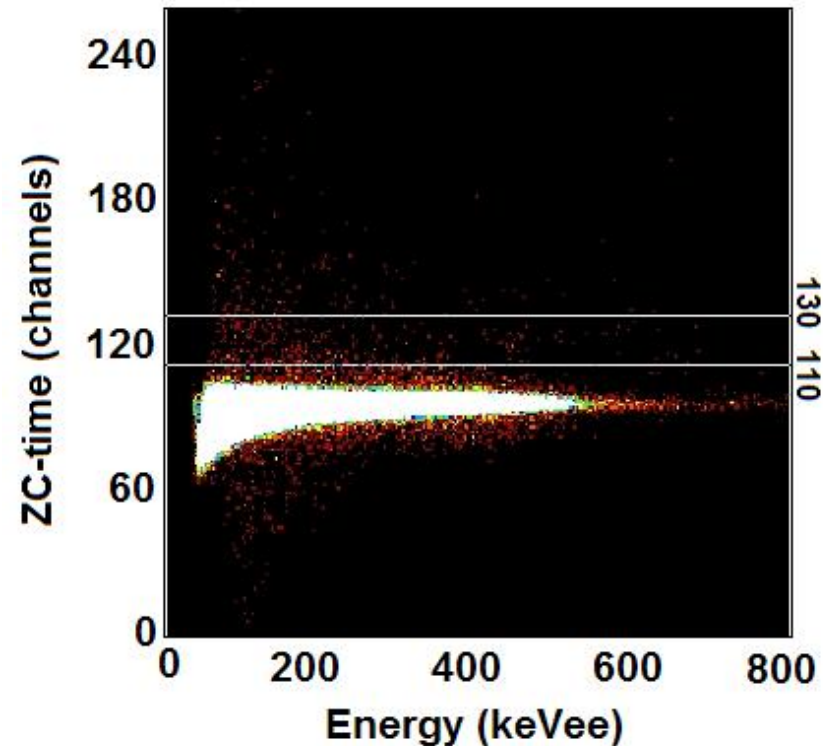
with pile-up rejection





Suppression of gamma-ray sensitivity in liquid scintillators

- Pile up rejection on
- The liquid cell shielded by lead and tin absorbers.





Suppression of gamma-ray sensitivity in liquid scintillators

Shield	No shield	Pb	Pb + Sn
Pb front	-	50 mm	50 mm
Pb side	-	10 mm	10 mm
Sn front	-	-	7 mm
Sn side	-	-	3.5 mm
γ -ray flux (kcps)	41	6.8	4.6
Relative neutron efficiency ^{*)}	1.25	0.84	0.70
intrinsic γ /neutron detection efficiency	3.1×10^{-4}	1.6×10^{-5}	5.1×10^{-6}

^{*)} relative to the intrinsic neutron detection efficiency of a polyethylene moderated He-3 counter irradiated with a Pu-Be source in the same geometry



First Conclusions

- A very efficient n/γ discrimination down to 20 keV_{ee}
- A high flash liquid scintillator showed an excellent n/γ discrimination capability
- A cheap solution due to PMT used in gamma spectrometry
- Unexpected quenching of slow component in the light pulses due to B events. Poor separation of thermal neutrons and gamma rays!!!
- Plastic scintillators with PSD capability still in the early phase of development



First conclusions

- New stilbene, grown from solvent, is a very attractive proposition. However, it has to be cheaper!
- Digital vs. analog n/gamma pulse shape discrimination,
- Optimisation of PSD in digital method – a large number of bits to assure a high dynamic range of input pulse height – important for low energies,



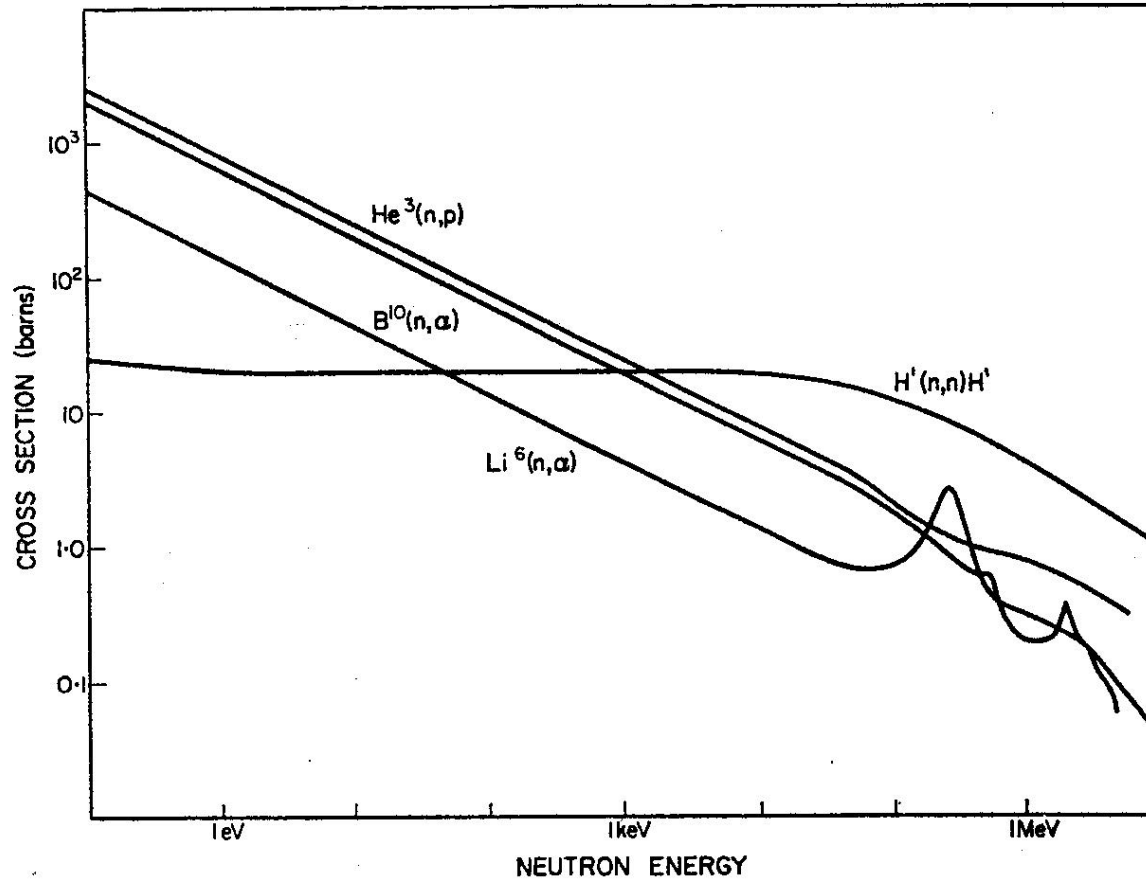
Inorganic scintillators in neutron detection

Neutron capture reactions of ^3He , ^{10}B and ^6Li

- $n + ^3\text{He}$ (0%*) $\rightarrow p + ^3\text{H} + 765\text{keV}$
 $\sigma = 5333 \text{ barn @ } 1.8 \text{ \AA } (\sim 0.025 \text{ eV})$
- $n + ^{10}\text{B}$ (20%*) $\rightarrow ^7\text{Li} + ^4\text{He} + 2.8 \text{ MeV}$ 7%
 $\rightarrow ^7\text{Li} + ^4\text{He} + 2.3 \text{ MeV} + \gamma$ (0.5 MeV) 93%
 $\sigma = 3838 \text{ barn @ } 1.8 \text{ \AA }$
- $n + ^6\text{Li}$ (7.5%*) $\rightarrow ^3\text{H} + ^4\text{He}$ (4.8 MeV)
 $\sigma = 941 \text{ barn @ } 1.8 \text{ \AA }$

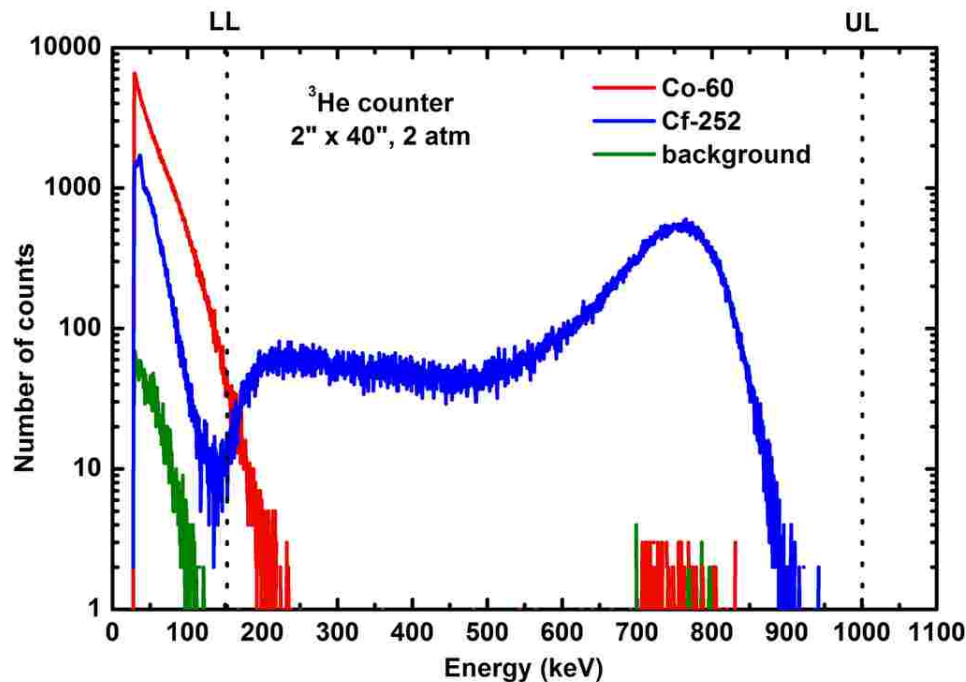


Inorganic scintillators in neutron detection





He-3 detectors



Energy spectra measured with He-3 counter under irradiation by γ -rays of a ^{60}Co source, ^{252}Cf and background.

Note the best gamma rays suppression due to a low atomic number and density of He, enhanced by a good pulse height discrimination



Classical slow neutron detectors

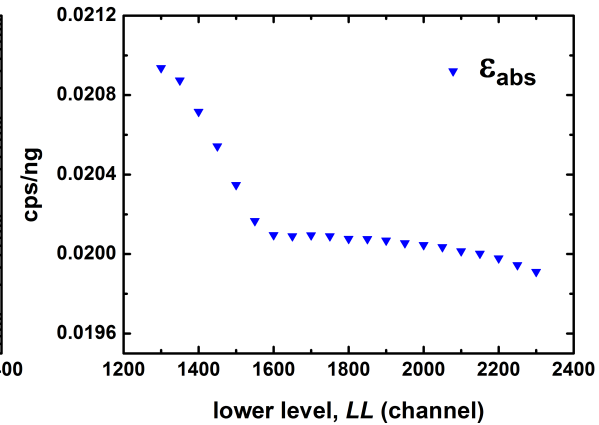
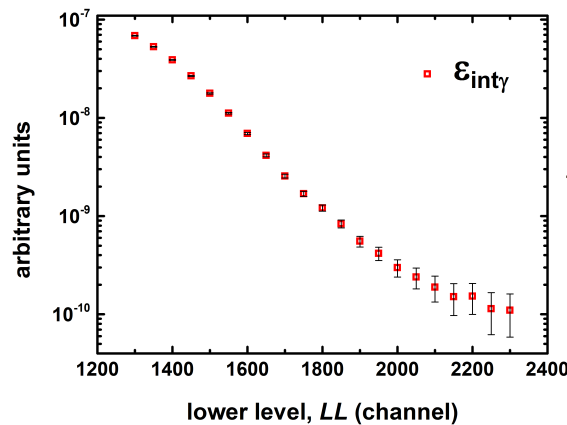
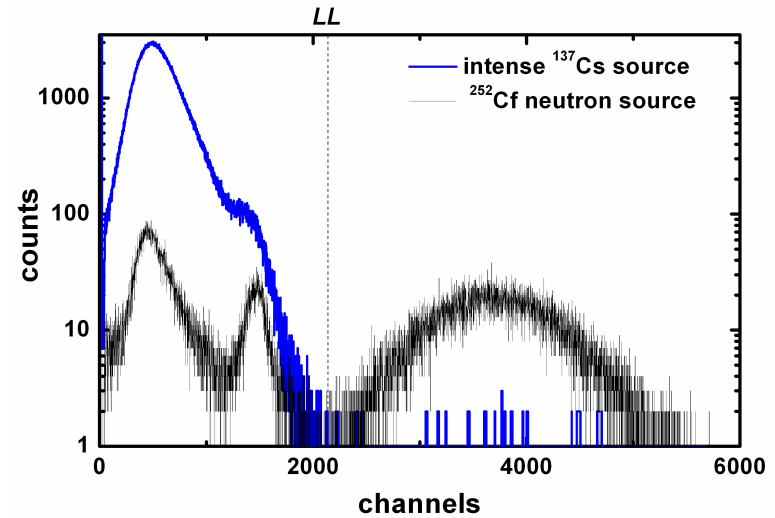
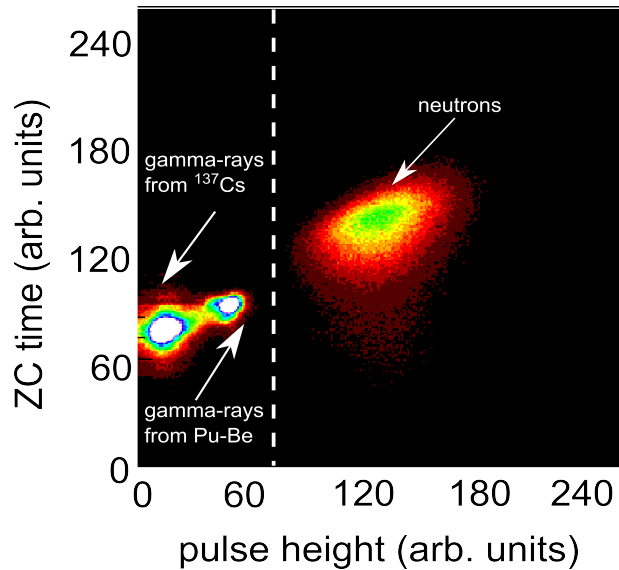
crystal	Neutron light output (ph/n)	Decay	α/β ratio	PSD
$^6\text{LiF/ZnS:Ag}$	160,000	80	0.44	+
$^6\text{Li-glass}$	~7,000	70	0.35	+
$^6\text{LiI:Eu}$	50,000	1,400	0.87	-



$^6\text{LiF}/\text{ZnS}:\text{Ag}$ screens

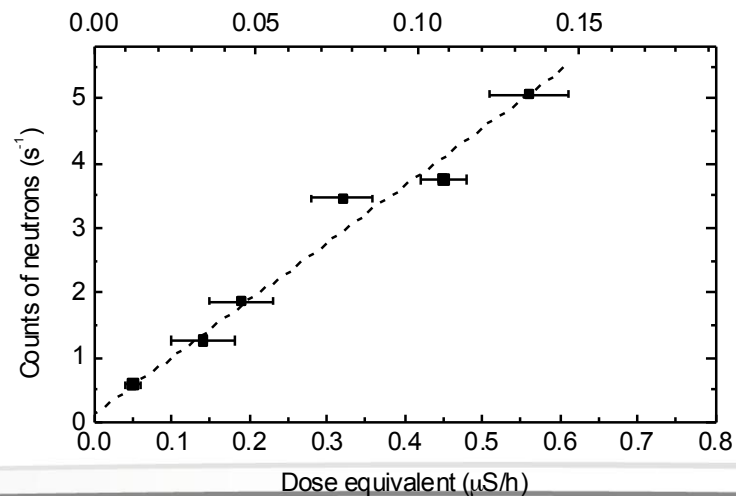
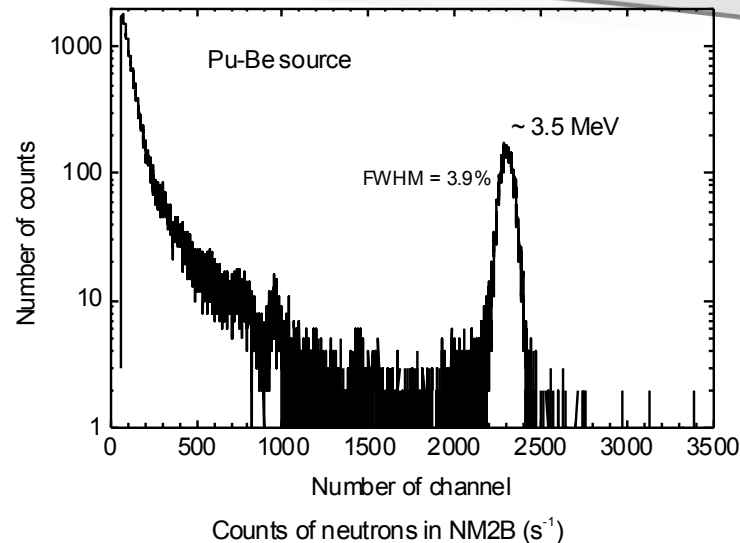
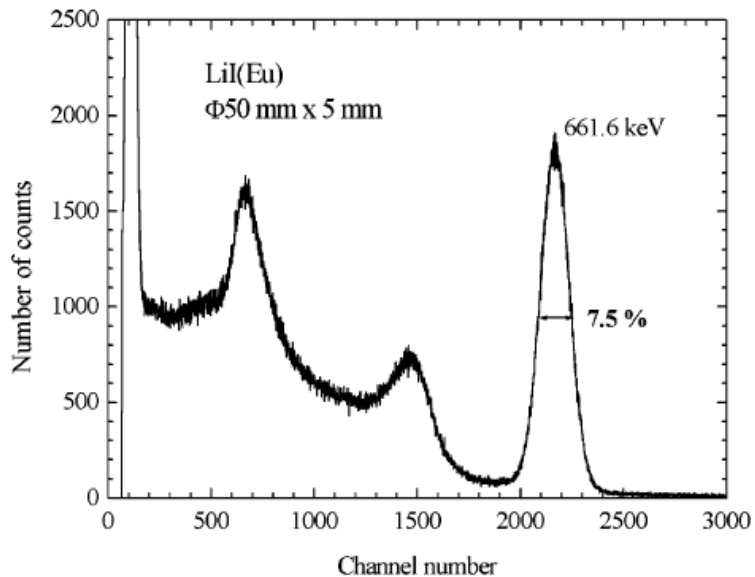


the screen with a light readout by scintillating fibers proposed by Saint Gobain for homeland security.





Lil:Eu



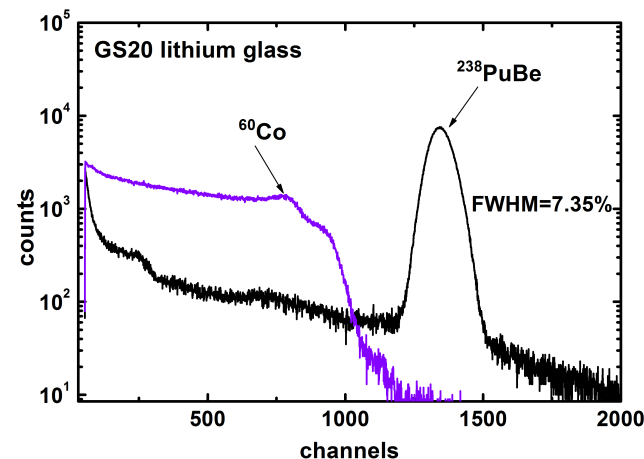
Light output 3000 phe/MeV (XP5200 PMT)

95% of thermal neutrons are stopped in 3 mm of Lil(Eu)

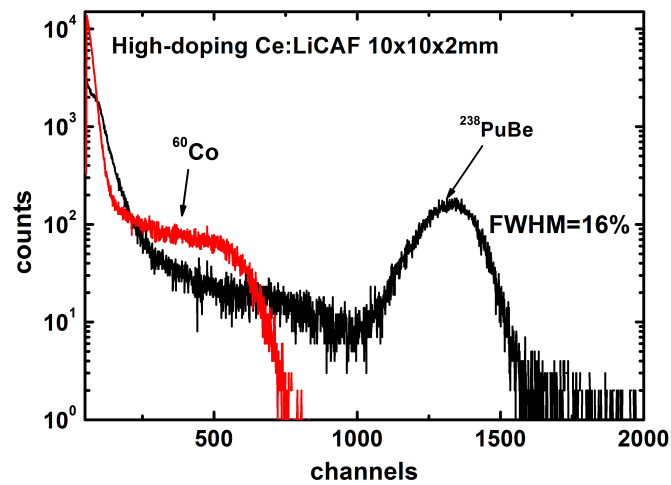


GS20 Li-glass and LiCAF:Ce ($\text{LiCaAlF}_6:\text{Ce}$)

scintillator	Light output (phe/MeV)
GS20	1300
LiCAF:Ce	~200
LiCAF:Eu	~8100
LiCAF:Eu (rubber)	~5600

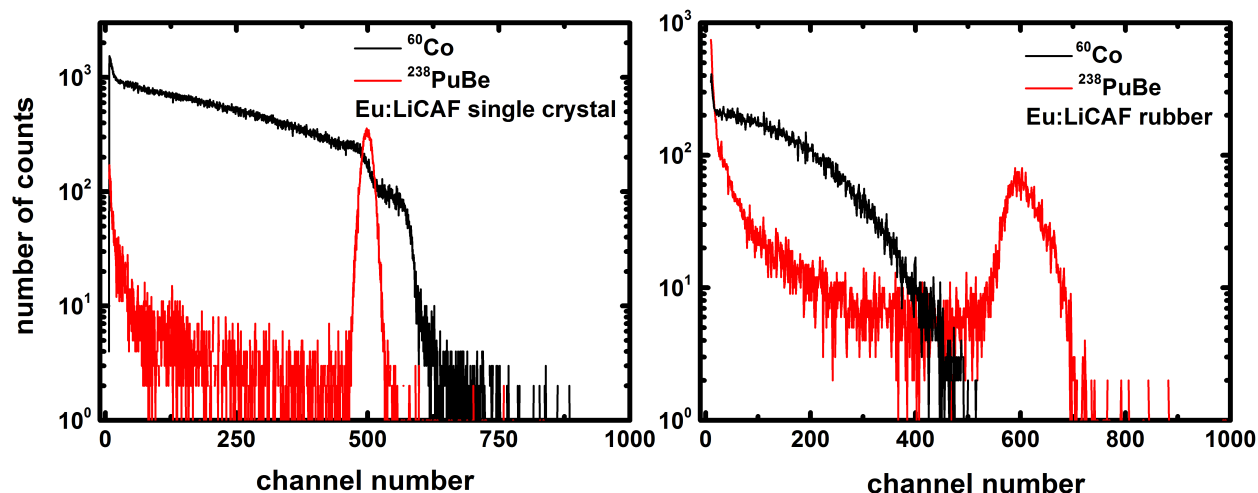


scintillator	Number of ^6Li atoms per cm^3	Intrinsic thermal neutron efficiency
GS20 $\varnothing 50 \times 2$ mm	$\sim 1,58 \times 10^{22}$	100%
High-doping Ce:LiCAF 10x10x2 mm	$\sim 0,5 \times 10^{22}$	35%
Low-doping Ce:LiCAF 10x10x2 mm	$\sim 0,5 \times 10^{22}$	32%
High-doping Ce:LiCAF $\varnothing 50.8 \times 2$ mm	$\sim 1 \times 10^{22}$	82%





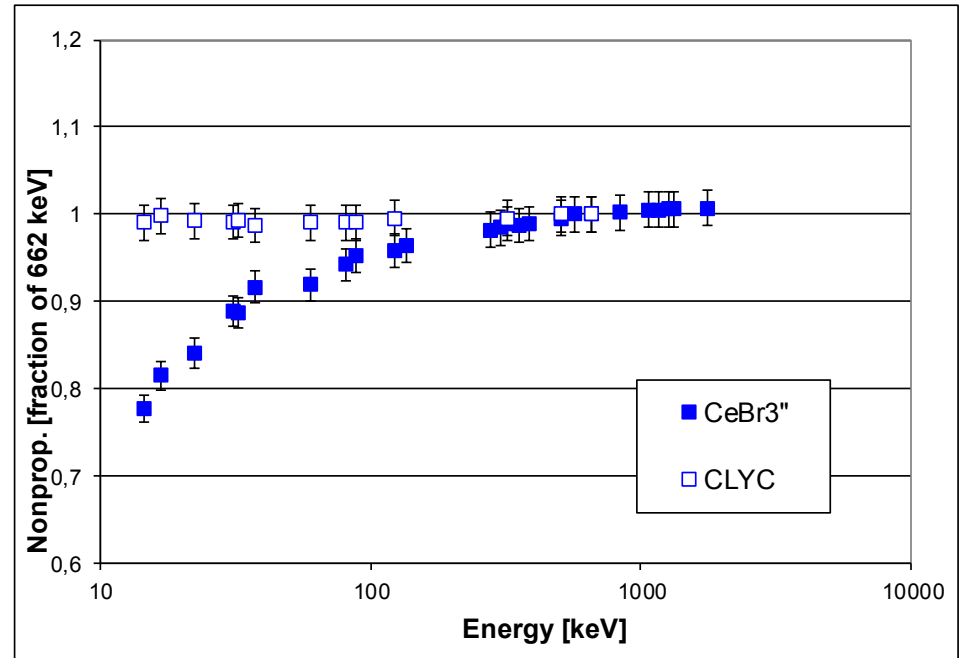
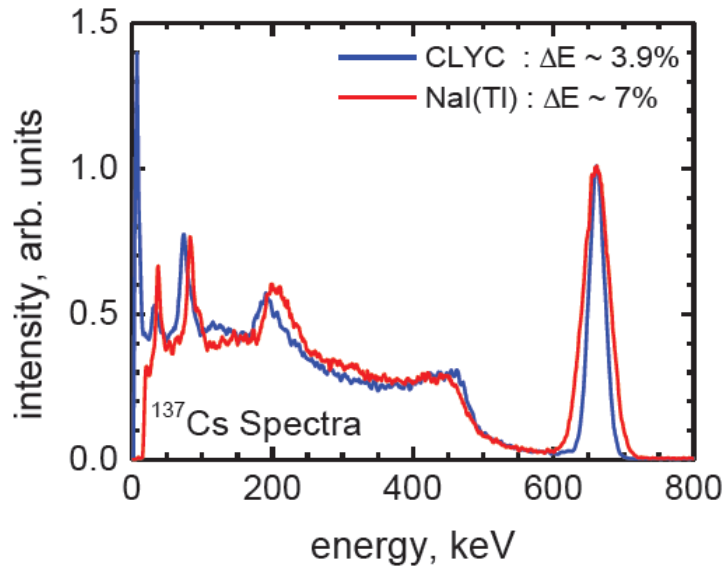
LiCAF:Eu and LiCAF:Eu rubber



- A limited detection efficiency and pulse height of the gamma ray response is the effect of a small size of LiCAF grain. However, it is sufficient to stop alphas and tritons produced by neutron capture in Li-6.
- Detection efficiency of neutrons in the rubber is about 3 times lower than in the crystal, but gamma rays rejection is more efficient. In the test with 7 MBq Cs-137 source (10 mR/h dose) the intrinsic efficiency of crystal of 10^{-4} was reduced to 10^{-6} in the test with the rubber.



CLYC scintillator ($\text{Cs}_2\text{LiYCl}_6:\text{Ce}$)



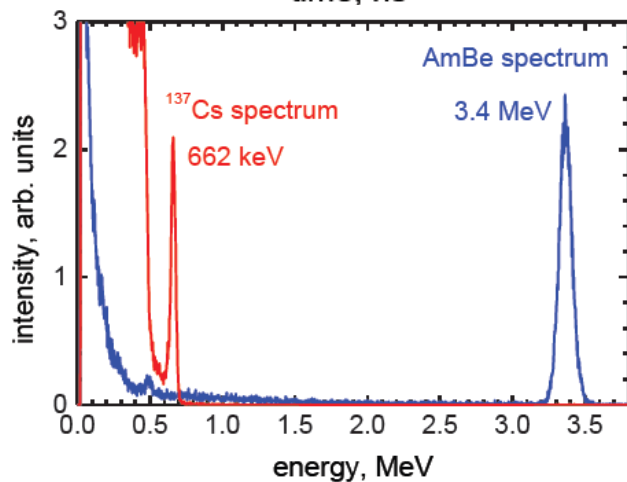
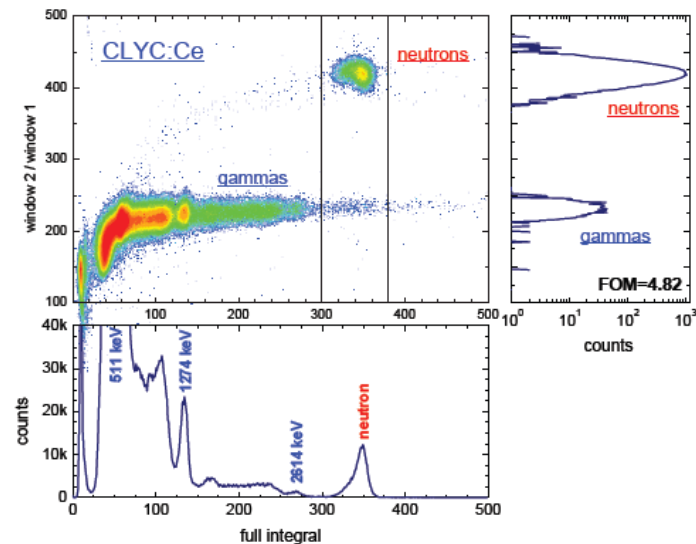
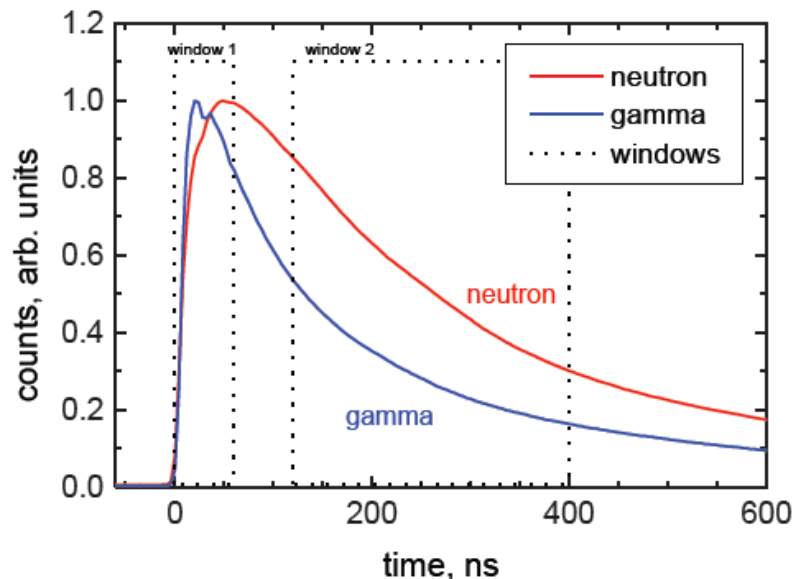
Light output 20,000 ph/MeV

Measured at NCBJ

Following RMD data



Thermal neutron detection in CLYC

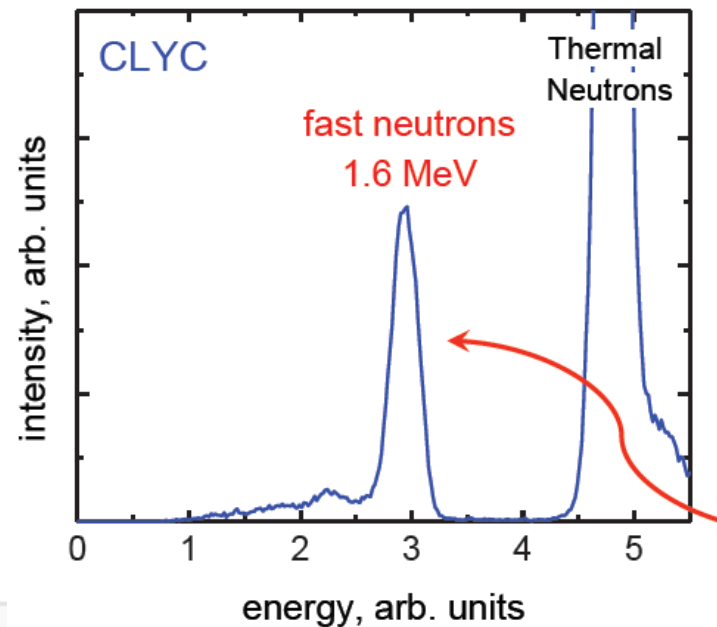
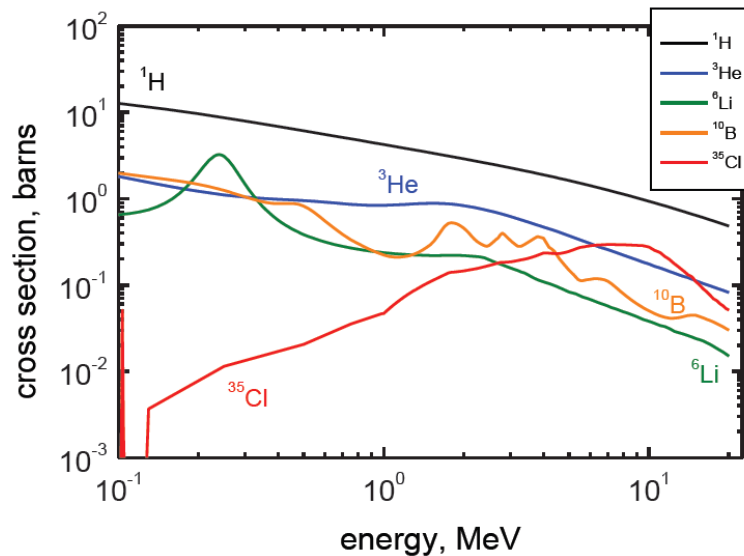
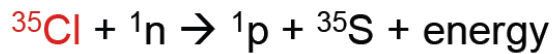
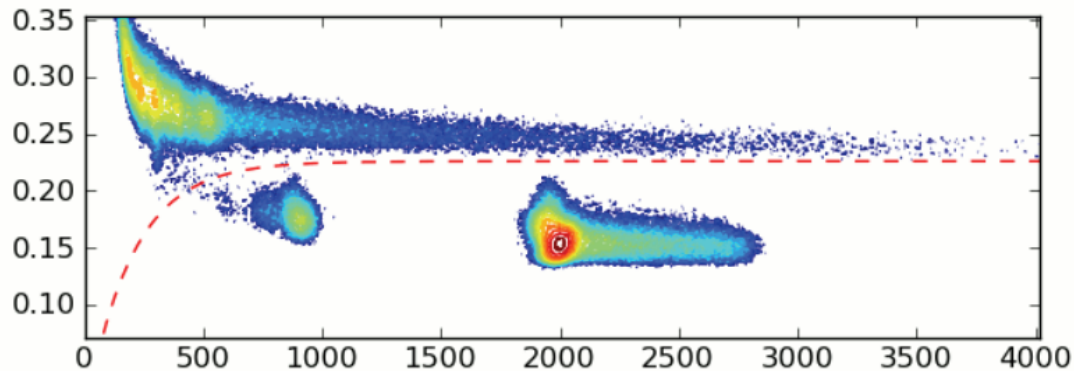


Thermal neutron detection:
 ${}^6\text{Li} + {}^1_0\text{n} \rightarrow \alpha + {}^3_1\text{H} + 4.8 \text{ MeV}.$

90% of thermal neutrons are stopped in 18.7 mm of CLYC



Fast neutron detection in CLYC





Summary of new developments

■ Organic scintillators:

- EJ 309 high flash liquid
- New stilbene – a very promising development, it has to be cheaper
- Plastic scintillators – still in the development phase

The most important application – physics, due to a good detection efficiency, a good n/gamma PSD and fast timing capabilities.

■ Digital n/gamma PSD



Summary of new developments

■ Inorganic scintillators

- CLYC - good performance in thermal neutron detection and gamma spectrometry.
- LiCAF rubber – a very interesting proposition of detector with a good suppression of gamma rays.
- LiI:Eu - renaissance of the old scintillator

Applications: handheld monitors in the homeland security equipment.



Border monitoring

$^6\text{LiF/ZnS:Ag}$ screens:



This is one of the best proposition to replace He-3 detectors in the border monitoring

