

Recent development in neutron detection with scintillators : Inorganic and Organic materials

P. Schotanus

SCIONIX Holland B.V.

Dedicated Scintillation Detectors

www.scionix.nl



Detection of Neutrons :

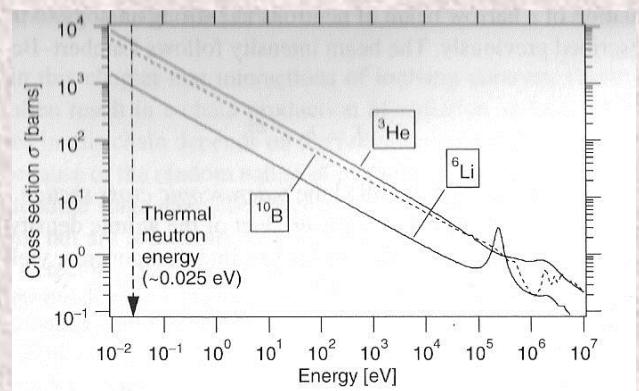
- Physics (e.g. particle Physics, HEP)
- Security (SNM e.g. Pu, U)
- Health Physics (dosimetry, often non spectrometric)

Neutron energy : **Thermalised 0.025 eV – MeVs**
(fast neutrons > 50 keV)

Interaction with Nucleus of absorber :
A. Scattering or
B. Nuclear reactions

- Elastic scattering (protons)
- Inelastic scattering + prompt gammas

Nuclear Reactions e.g. : $^{10}\text{B}(\text{n } \alpha) ^7\text{Li}$, $^3\text{He}(\text{n } \text{p})^3\text{H}$, $^6\text{Li}(\text{n } \gamma) ^3\text{H}$,
 $^{157}\text{Gd}(\text{n } \alpha) ^{158}\text{Gd}$



Most common detection method for neutrons :

1. ${}^3\text{He}$ - tubes (pressurised)

Most unproblematic detector :

- Easy to operate (no special electronic needed)
- gamma / neutron rejection $> 10^6$
- No serious safety issues
- Large sizes possible (meters long)



World wide structural He-3 shortage → availability / cost problems

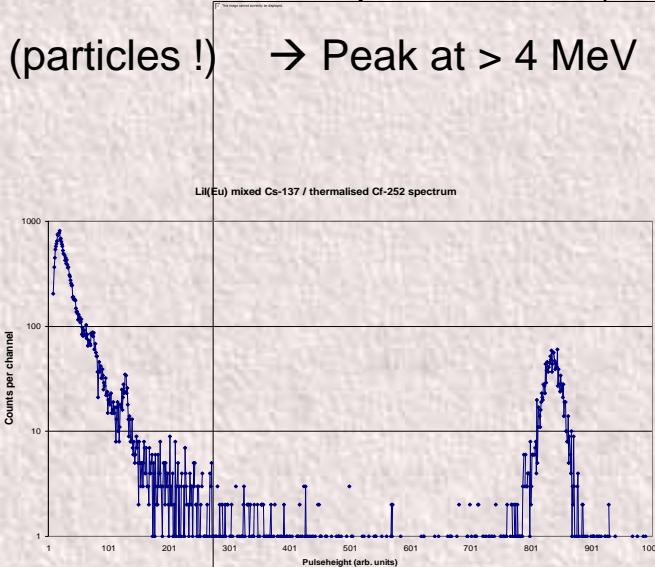
Possible alternative : Detection of neutrons with scintillators :

- A. Thermal neutrons via nuclear reactions on Li, B or Gd in the material.
- B. Fast neutrons via elastic (recoil) scattering in proton containing materials (organic scintillators)

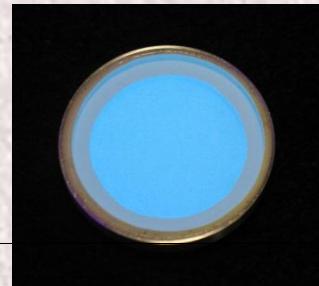
Usually , neutrons associated with gammas, both will interact with most scintillators but :



(particles !) → Peak at > 4 MeV



Neutron / gamma separation possible via Pulse height



Lil(Eu) scintillator

However Lil(Eu) 96 % enriched is a relative expensive material that cannot be made in large sizes.

The ${}^6\text{Li}$ (Eu) alternative is an excellent solution for hand held instruments and dosimeters but is not an option for e.g. radiation portals



Alternative ${}^6\text{Li}$ containing scintillators

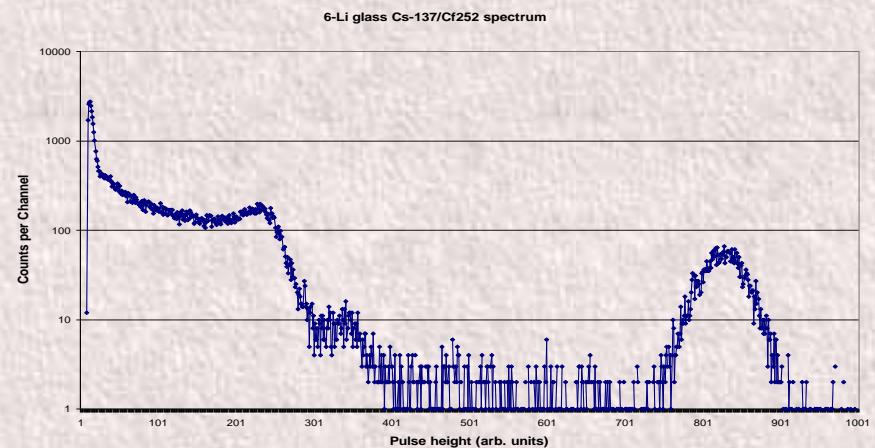
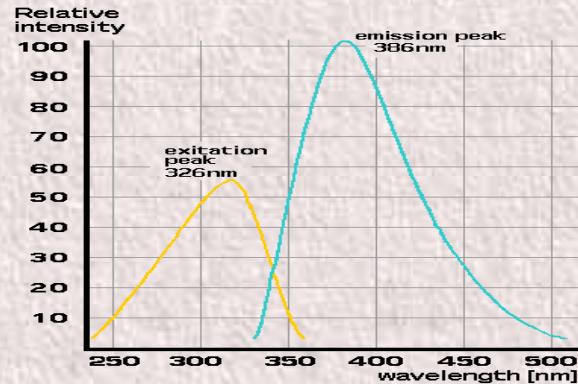
B

${}^6\text{Li}$ Loaded glass scintillator (Ce doped)

- Also expensive
- Low neutron peak location (approx. 1.8 MeV) implying more problem with gamma rejection
- no large lengths possible (self absorption)

An alternative if gamma flux is low and of low energy and if time resolution is an issue

(scintillation is fast, 60 ns decay time)



Neutron / gamma spectrum 6-Li glass

C. $\text{Cs}_2\text{LiYCl}_6:\text{Ce}$ (CLYC) crystals (density 3.3 g / cc)

Proportional crystal :

Energy resolution approx. < 5 % (662 keV)

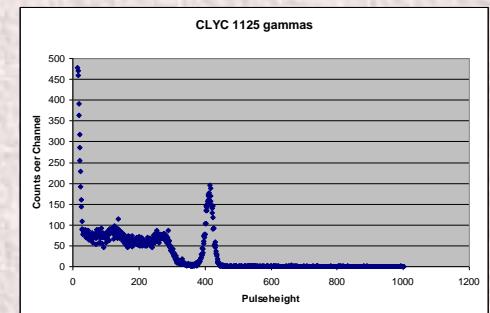
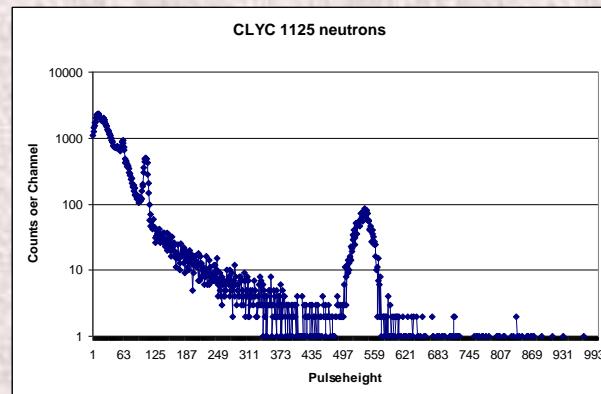
Neutron peak at 3.3 MeV



Neutrons no fast (CVL) component : Neutron / gamma discrimination possible using PHA and PSD

Problems :

- Cost high
- Crystal quality issues
- LO rel. low



Cf-252 / Cs-137 spectrum CLYC 1"x1"

Large area neutron detection

Screens of pressed powders 6-LiF and ZnS(Ag) : EJ 426

(cross section of 941 barns for 0.025 eV neutrons)

- 0.3 or 0.5 mm thick possible

Screen Type	$^6\text{LiF:ZnS}$ Mass Ratio	^6Li Density atoms/cc	Theoretical N _{TH} Efficiency 0.32mm Thick	Theoretical N _{TH} Efficiency 0.5mm Thick
EJ-426-0	1:3	1.14×10^{22}	0.29	0.41
EJ-426HD2	1:2	1.63×10^{22}	0.39	0.53

Detection Properties of Some EJ-426 Screens

Problem is to get the light out (due to self absorption in powders)

Wavelength shifting (fibers) e.g. **EJ 260** for large areas needed

- Due to non negligible gamma interaction in ZnS(Ag), PSD needed to obtain He-3 comparable neutron / Gamma discrimination

Commercial products available to replace He-3 in panels



D. Boron Loaded scintillators :

No inorganics known except for Li Borate glasses with low light output

The other alternative :

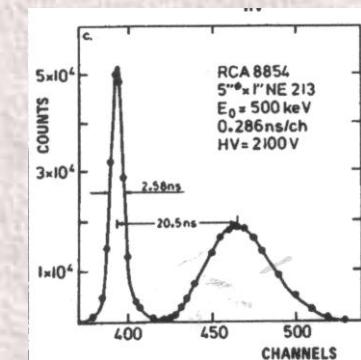
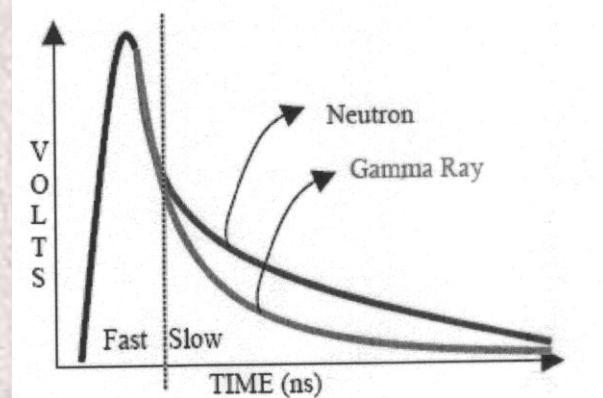
Liquid scintillators

Some activated organics liquids show different pulse shape for neutrons and gammas.

Liquid scintillators known since 1960's

Most well known NE213(= EJ301 = BC501A)

Xylene based.



Different ways to do neutron/gamma separation with PSD

1. QDC with 20 ns and 1 microseconds gate (or different)
2. Converts signals to time spectrum using a double delay line amplifier and CFDs (time spectrum shows two peaks)
3. Digitize the signal using wave form analyzer (500 Mhz to 1 GHz flash ADC)

Traditionally NIM electronics were used.

For many real field applications outside Physics this is very unpractical

Other **disadvantages** of liquid scintillators :

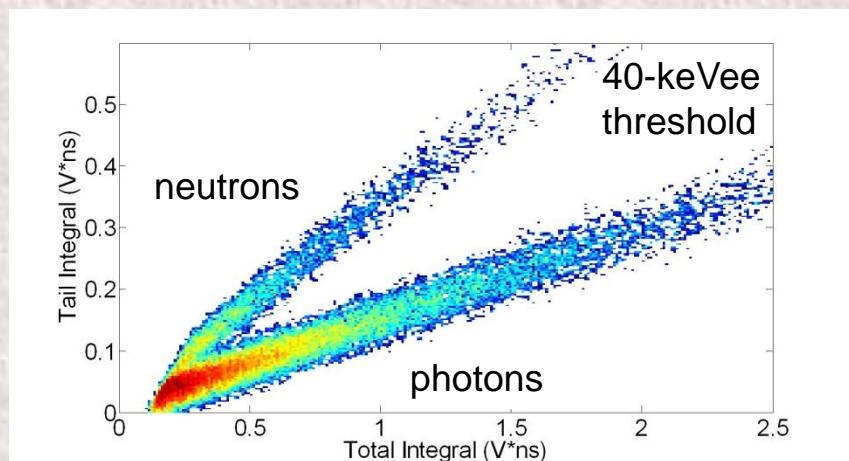
- Expand with increasing temperature (expansion volume is needed)
- Many types EJ301, EJ315 are low flash point (flammable) and toxic materials (transport / safety / handling issues)

New developments :

- High flash point non toxic liquids
- FPGA based pulse digitizing techniques



These new liquids open up applications



EJ301

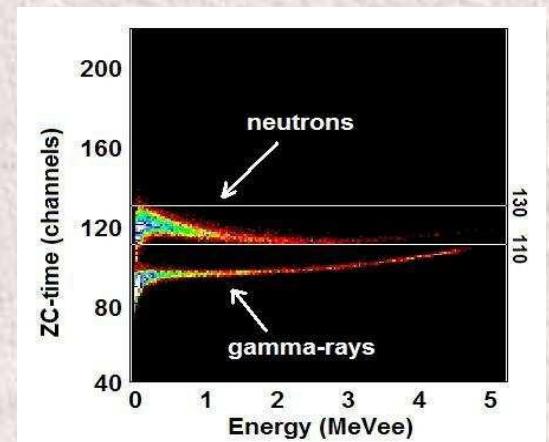
EJ301 is a scintillating liquid **equivalent to NE213 BC501A** specially designed for neutron / gamma discrimination.

Properties :

Light output (rel. to antracene) :	78 %	75 %
Photon yield / MeV electrons :	12.000	11.500
Maximum of emission wavelength :	425 nm	424 nm
Density:	0.874 g/ cc	0.964 g/ cc
H:C ratio:	1.21	1.25
No. C atoms per cc :	$4.0 \cdot 10^{22}$	$5.46 \cdot 10^{22}$
No. H atoms per cc :	$4.8 \cdot 10^{22}$	$4.37 \cdot 10^{22}$
No. electrons per cc :	$2.3 \cdot 10^{23}$	$3.17 \cdot 10^{22}$
Flash point :	26 °C	144 °C
Refractive index :	1.50	1.57
Decay time short component	3.2 ns	3.5 ns
Decay time long components	32.3, 270 ns

EJ309 (NEW)

EJ309 is a scintillating liquid especially designed for neutron discrimination It has a high flash point, **low vapour pressure** and no **toxicity (biodegradable)**.



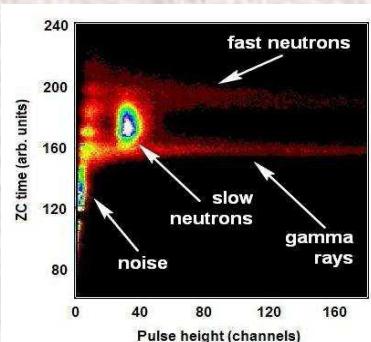
QUESTIONS :

- PSD EJ309 slight worse than EJ301 but how much ?

- Are fast PMTs always needed for good PSD ?

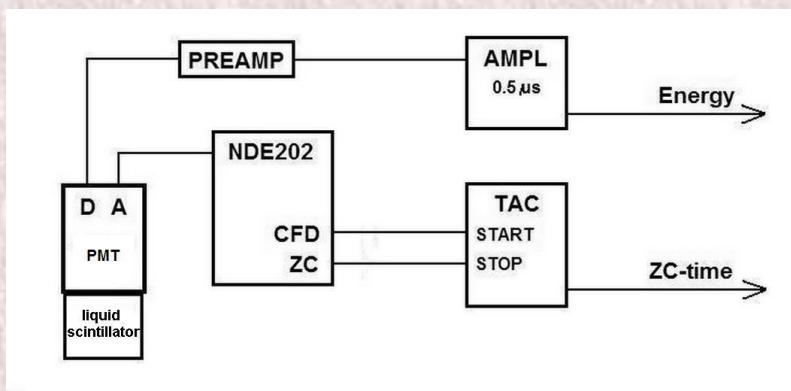
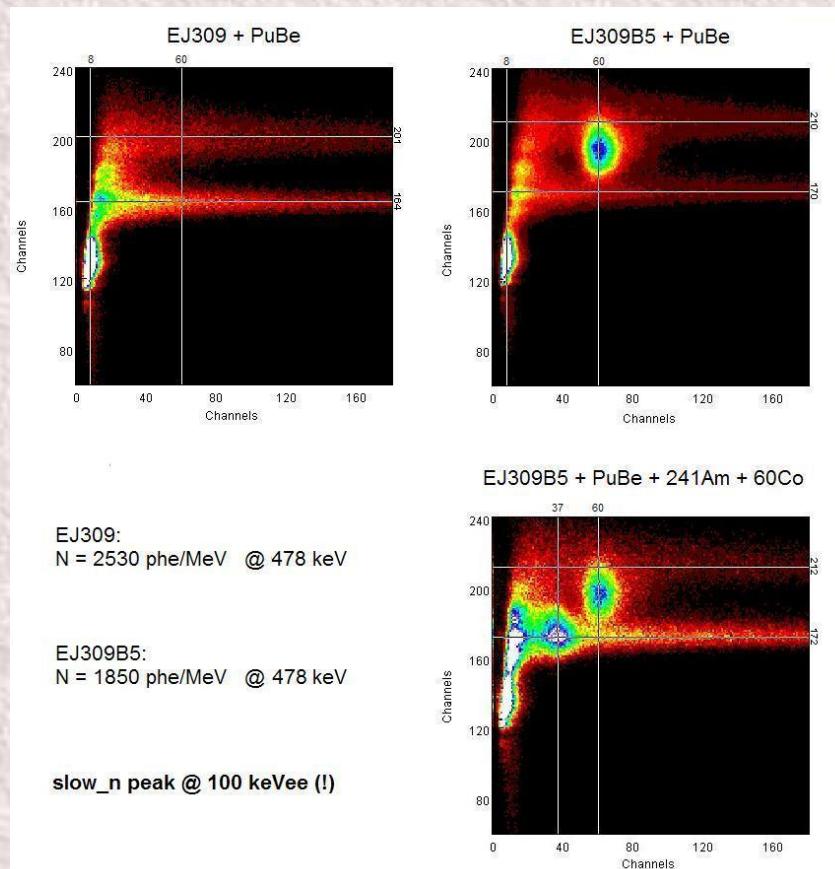
Some answers are given in the paper by Stevanato et al. NIM A 690 (2012) pp 96 but all depends on neutron energy...

When Liquid scintillators are Boron doped,
the neutron sensitivity is increased

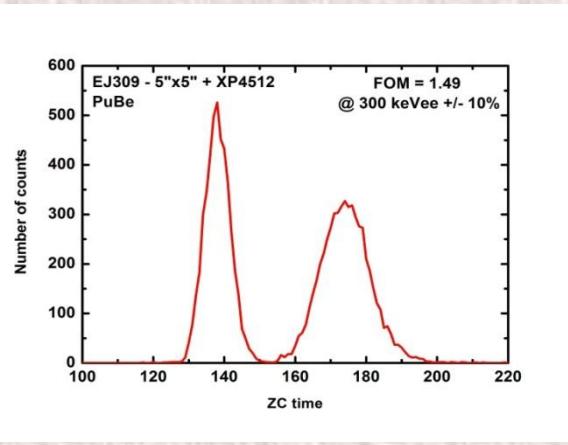
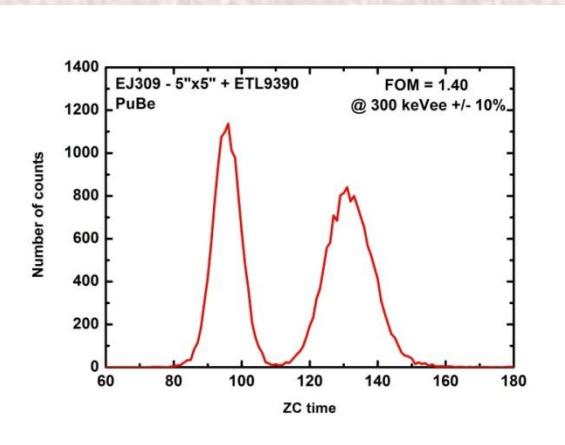


Boron peak at higher
energies than with
EJ301
(100 keV versus 60 keV)

Gammas, fast neutrons
and slow neutrons can
be separated by PSD



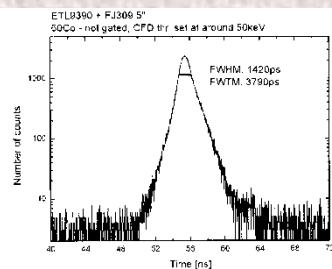
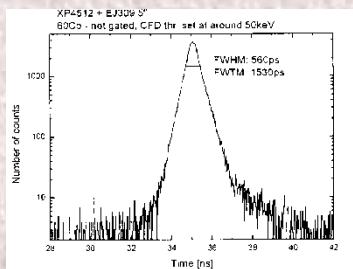
Effect of slow versus fast PMT



FOM is the same for a “fast” XP4512 as a “slow” ETL 9390
(data by Swiderski et al.)

Note that Fast PMTs suffer from a non-ideal photoelectron collection efficiency

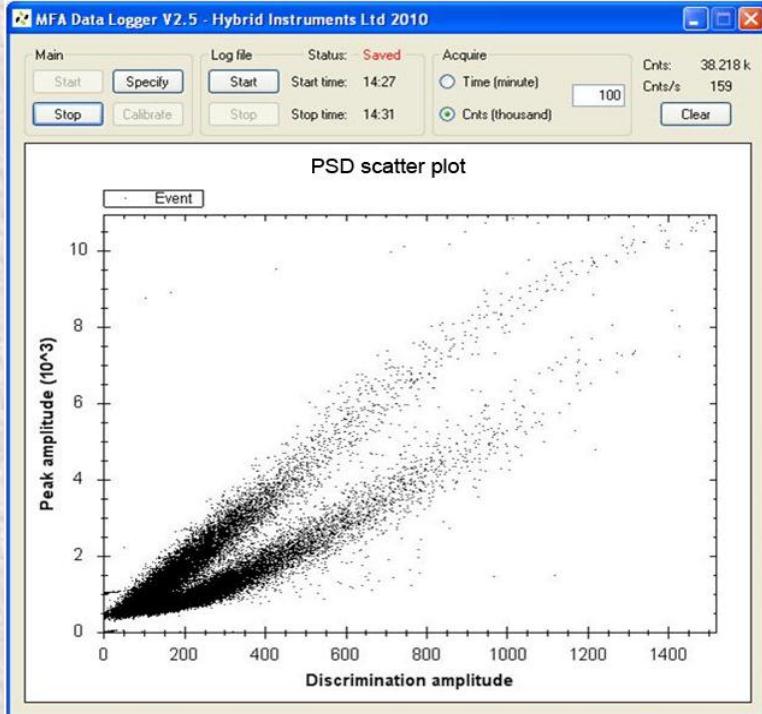
HOWEVER for some applications (neutron / Gamma coincidences) a optimum time resolution is needed (< 1 ns FWHM)



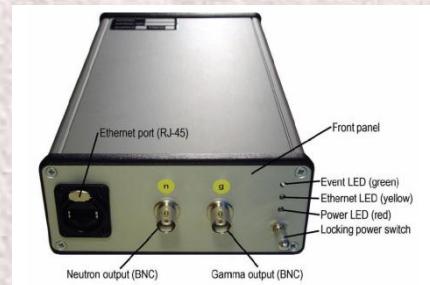
Fast PMTs offer better time resolution :
(500-600 ops versus 1-1.5 ns)

ELECTRONIC DEVELOPMENTS

Advanced pulse sampling techniques using FPGAs allow on line 250 kHz throughput neutron / Gamma separation



Instruments presented elsewhere in the workshop like the Mixed Field Analyser developed by Hybrid instruments (UK)

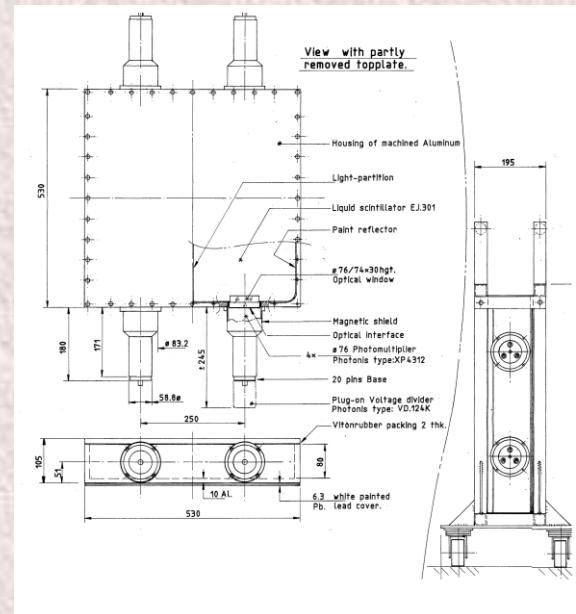
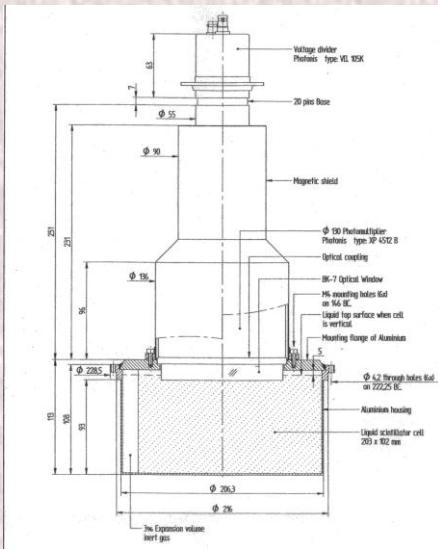


For Physics applications multi-channel VMA based fast digitizers are more frequently used

Design of Liquid cells for timing and PSD

General :

Liquids expand (T) and expansion space has to be available (3-5 % for temp. range $-20 - +50$ °C)
With properly designed so called dip-in windows optical interface between liquid and readout window can be guaranteed in all orientations

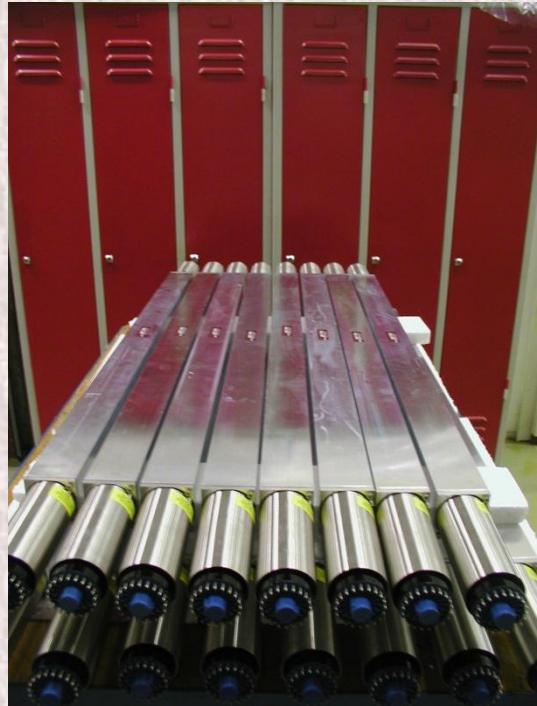


Larger sizes cells of different geometries also provide a good neutron / gamma PSD

SOLUTIONS THAT WORK

HOWEVER :

Neutron / Gamma PSD can be spoilt by a geometry where the light paths to the PMT are much different !



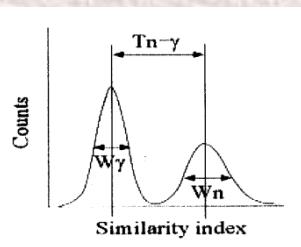
a 1:1 plug-in replacement of He-3 tubes by long liquid cells is not possible !

Some recent tests on several liquid scintillator assemblies :

- What is influence of PMT on PSD ?
- What is influence of shape / PMT size

All tests with analog NDE 202, threshold 100 keV (el),
10 mm Pb shielded weak Cf-252 source

"3"x3" EJ301 2.1 ns rise time XP4312



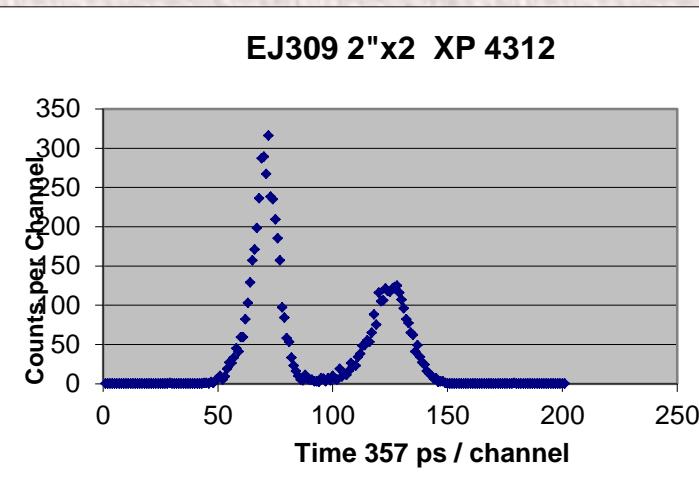
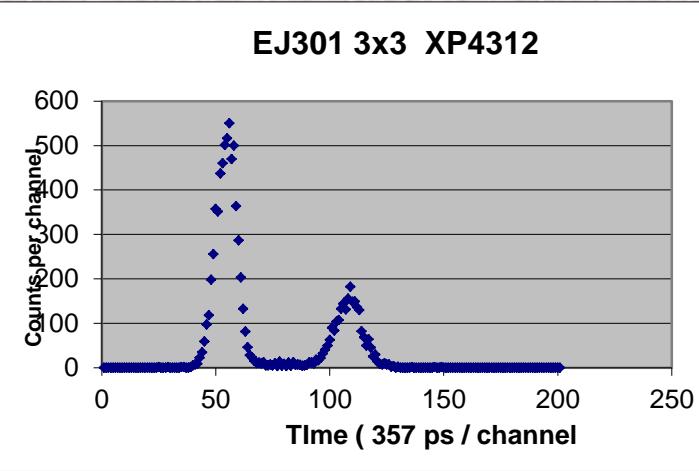
FOM = 2.2

2"x2"EJ309 2.1 ns rise time XP4312

$$FOM = \frac{T_{n-\gamma}}{W_n + W_\gamma}$$

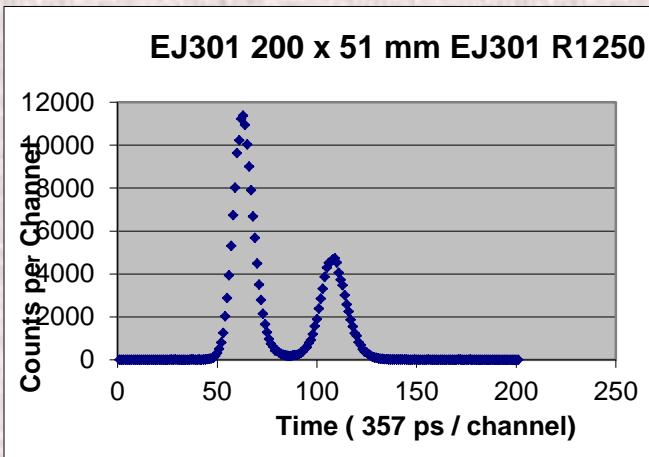
FOM = 1.9

N-g PSD EJ301 better

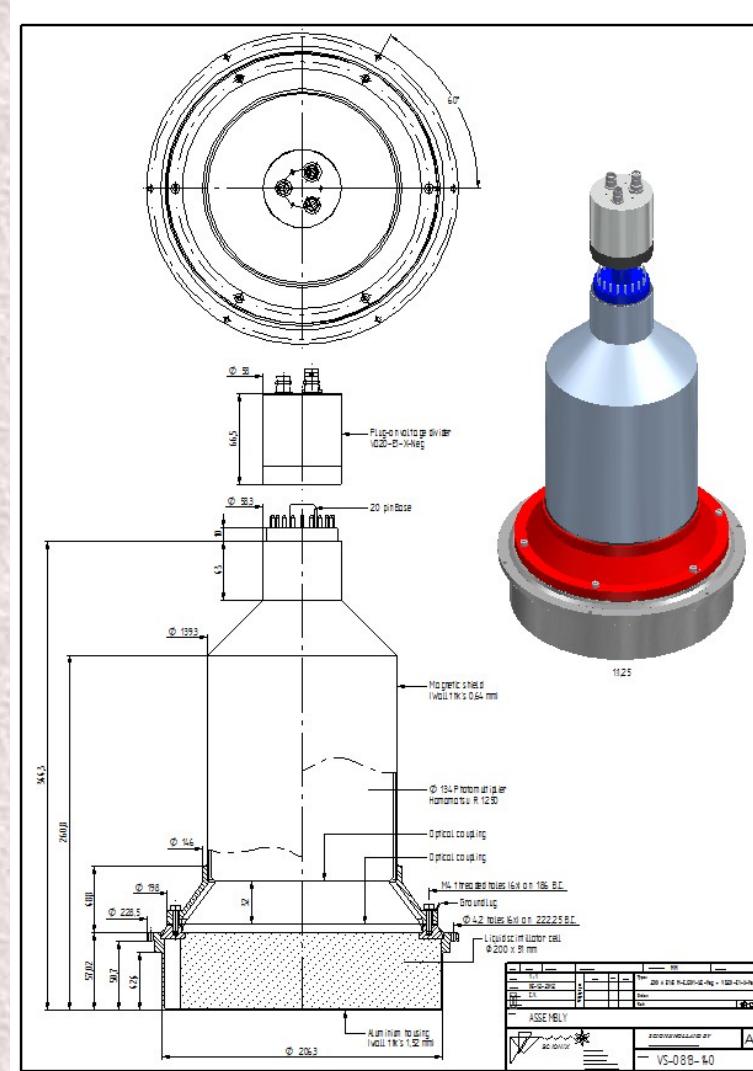


Larger cells, smaller PMTs

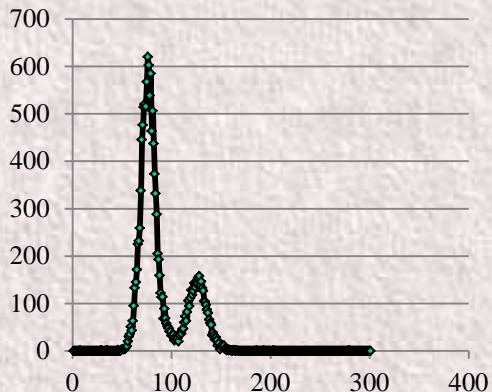
200x51 PMMA light guide R1250 PMT



FOM = 1.88

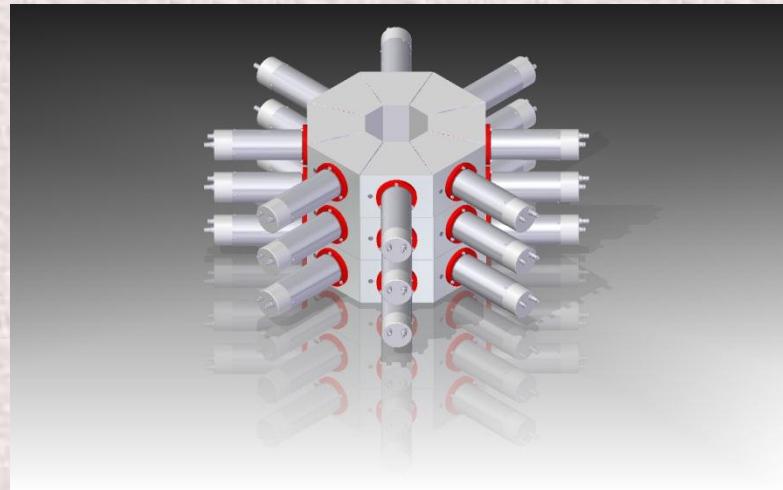
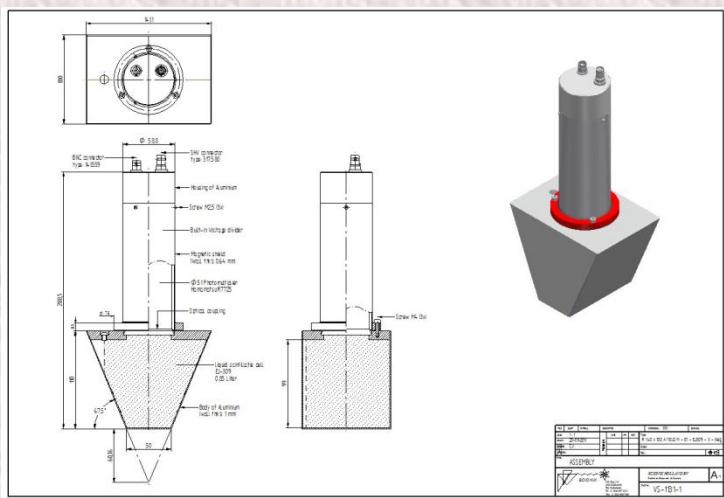


Pie shaped EJ309 cells 2 ns ET9214

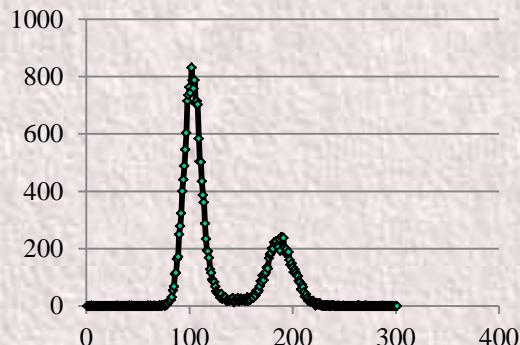


FOM = 1.22

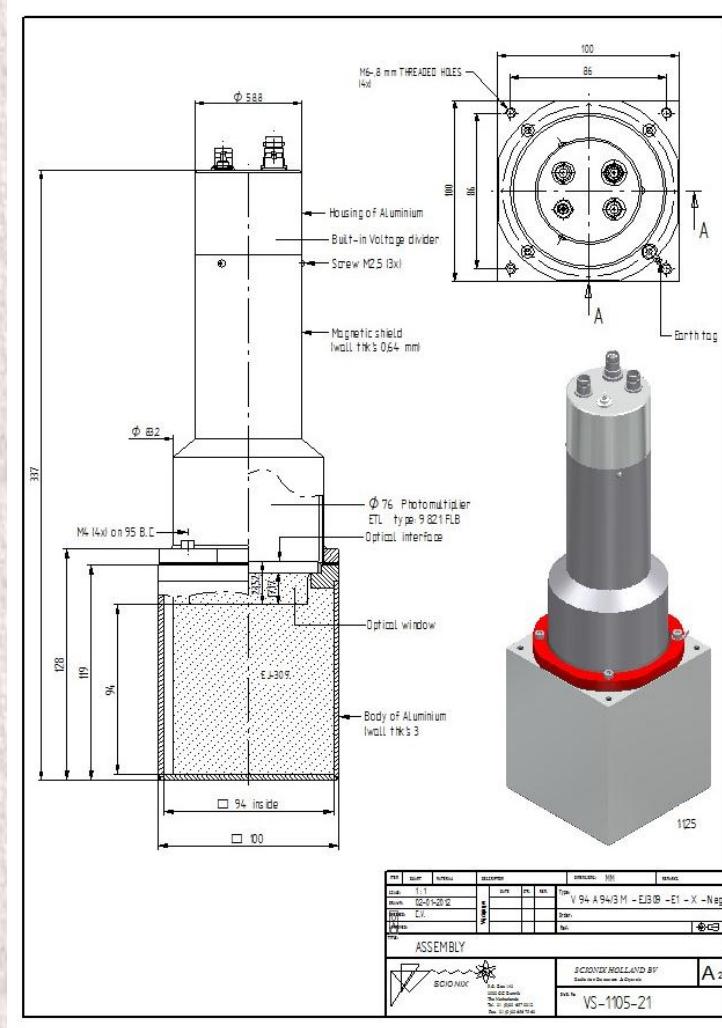
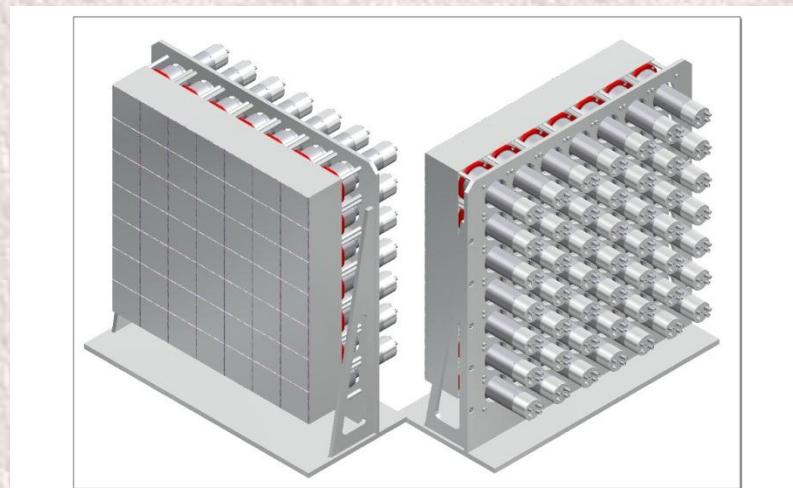
Plutonium multiplicity counter (safeguards)



**EJ309 94x94x94 mm
2.1 ns rise time ET 9821**



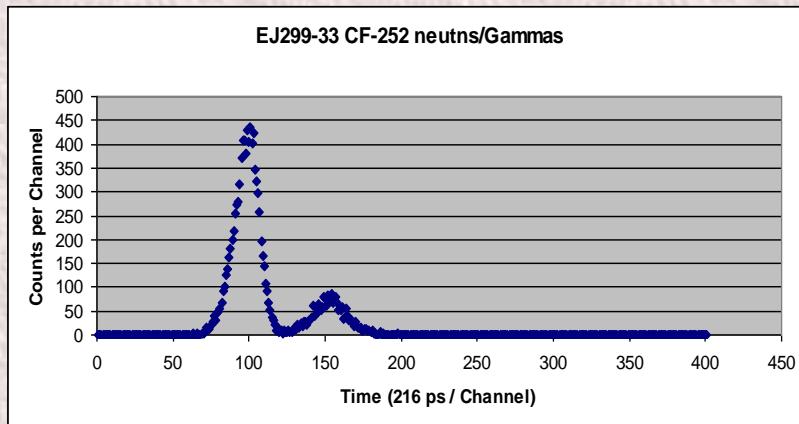
FOM = 1.75



Newest development : EJ299-33

Plastic scintillators with neutron / Gamma discrimination

Zaitseva et al. NIM A 668 (2012) pp 88-93. (LLNL)
(PVT based, w. special dye and activator)



51x51 mm 2 ns ET 9214

Neutron / Gamma
discrimination possible

FOM = 1.5

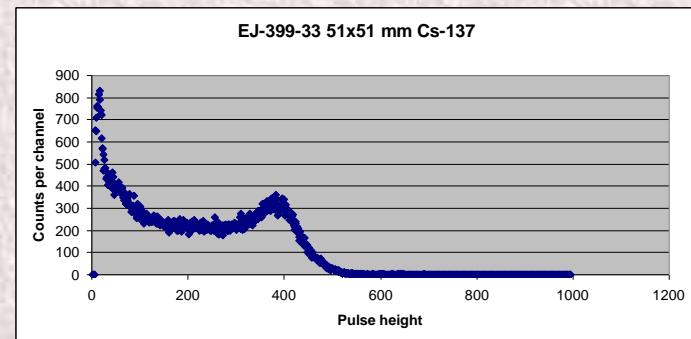
HOW GOOD is it really ?

Time spectra with analog non
optimised electronics



First 51x51 mm samples of EJ299-033

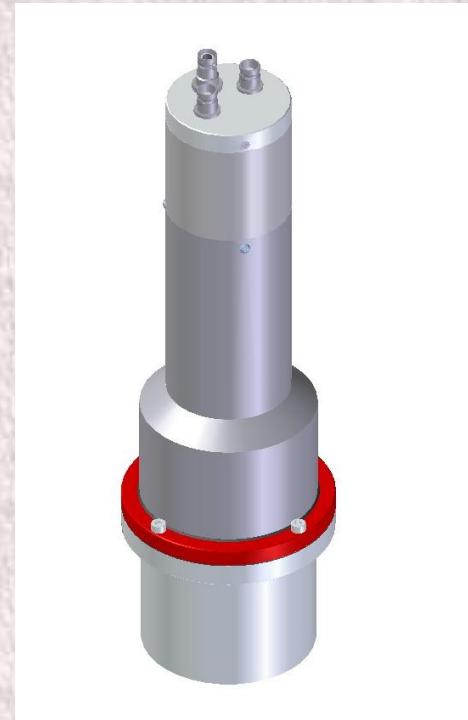
Material licensed to ELJEN



LO comparable to EJ309

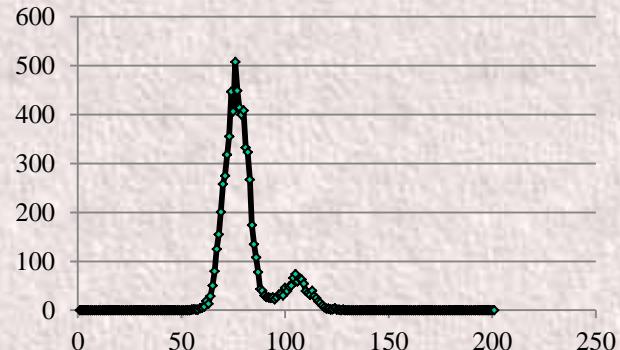
Important questions :

1. Optimum time resolution ?
2. Maximum dimension , effect on PSD ?
3. Cost ?



76 x76 mm 2 ns ET 9821

FOM = 1.25



**Advantage compared to liquid cells is obvious,
geometrical arguments to be further investigated**

Of course FOM threshold dependent !

EJ-299-33 PSD PLASTIC SCINTILLATOR PROVISIONAL DATA SHEET

This revolutionary plastic scintillator possesses pulse shape discrimination properties enabling the separation of gamma and fast neutron signals on the basis of their timing characteristics using conventional PSD electronics systems..

Physical and Scintillation Constants:

Light Output, % Anthracene	56
Scintillation Efficiency, photons/1 MeV e-	8,600
Wavelength of Max. Emission, nm	420
No. of H Atoms per cm ³ , x 10 ²²	5.13
No. of C Atoms per cm ³ , x 10 ²²	4.86
No. of Electrons per cm ³ , x 10 ²³	3.55
Density, g/cc:	1.08

Maximum sizes 152x152 mm made, larger sizes under development

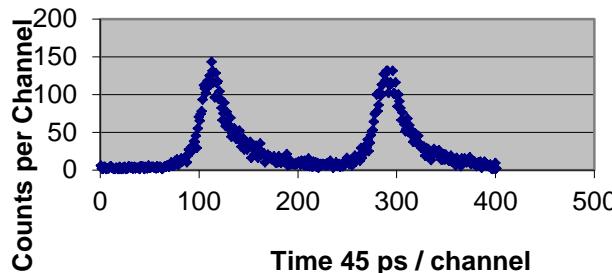
Decay times appox 6 ns and 80 ns



Co-60 time resolution for EJ230 cone coupled to XP2020 and 51x51 mm Ej309 cell coupled to 2 ns rise time ET 9214

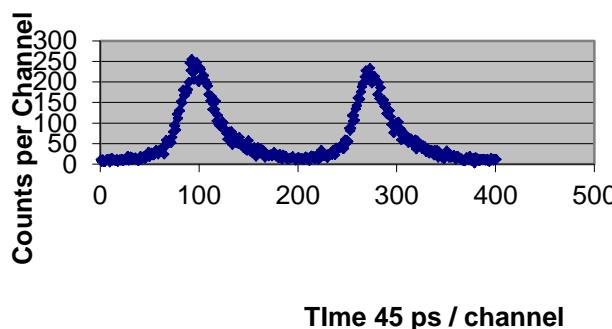
**Time resolution
1.62 ns FWHM**

Tlme resolution 51x51 EJ309 versus EJ230 cone



(Peaks 8 ns difference)

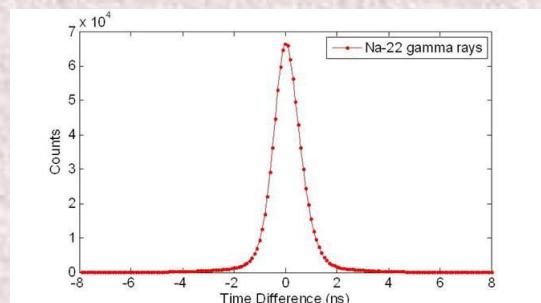
Time resolution EJ233-33 76x76 mm versus EJ230 cone



Co-60 time resolution for EJ230 cone coupled to XP2020 and 76 mm Ej299-33 detector coupled to 2 ns rise time ET 9821

**Time resolution
1.44 ns FWHM**

Flaske et al SORMA West 2012 EJ309 76x76 mm 1.2 ns FWHM



Conclusions :

1. The current availability of high point non dangerous goods liquid scintillators opens up possibilities to use these detectors where was prohibitive in the past.
2. The current availability of digital techniques allows the construction of novel instruments for neutron / gamma discrimination in mixed neutron / gamma fields
3. Liquid scintillators can be a replacement for He-3 tubes in **some** applications
4. The PSD performance of EJ301 remains superior to safer alternative liquids at the cost of flammability / toxicity issues. For many applications EJ309 is perfectly sufficient.
5. New EJ299-33 plastic scintillators slow neutron gamma PSD but the neutron-gamma separation is not as good as EJ309. Cost is comparable to liquid cells but in small sizes less expensive. Material much more costly than classic (EJ200) plastics due to expensive special casting process.

New activities taking place :

- A.Green emitting variants of EJ299-33
- B.More mechanical stable variants (EJ299-33 has low softening point !)
- C.Segmented (13x13x50 mm) EJ299-33 samples readout by SiPM
- D.Material compatibility tests