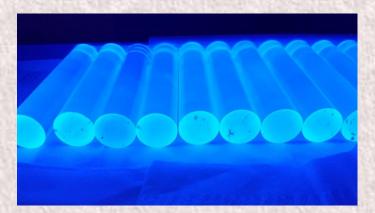
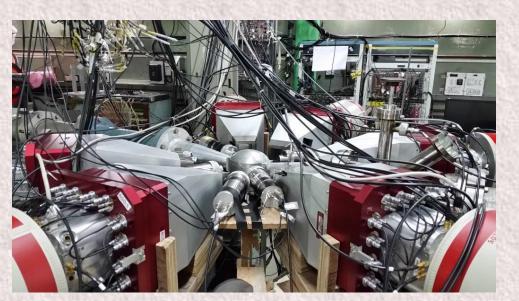
Production and testing of commercial scintillators



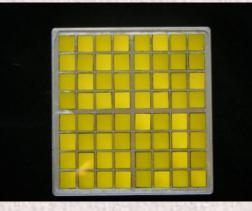
P. Schotanus

SCIONIX Holland B.V. Bunnik The Netherlands

www.scionix.nl







scintillation detectors from an industrial point of view :

Aim : to be able to produce larger quantities of devices with well defined specifications against affordable cost

Requirements

Why new materials ?

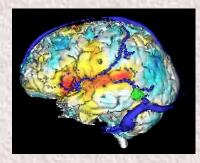
- Newer scintillation crystals, requirements
- To what level are new materials applied at this moment?

Inorganics / Organics

- Challenges and hurdles when going to large(r) scale production
- Advances in crystal readout : SiPms and actual applications
- A few examples



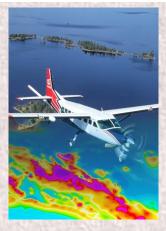
Scintillation detectors : standard tools in many disciplines





- Medical (SPECT/PET/CT) : Imaging, density
- Health Physics : Dose (particles, photons, neutrons)
- Security (finding/identifying sources) : spectroscopy
- High Energy Phyics (HEP) : particles, photons, imaging
- Nuclear Physice (high res.) Gamma rays spectr. Timing
- Geology: finding U, Th via spectroscopy,
- Mineral exploration (density, PNGA elemental analysis)
- Oil industry (density, well logging,)
- Space (gamma) spectroscopy, neutrons

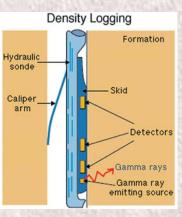






There is more around han 662 keV energy resolution

EACH DISCIPLINE has a vastly different specifications, definition of "better scintillators" depends on application !

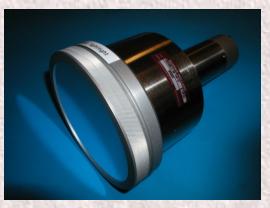






Advantages of scintillators : - large efficiency (density up to approx. 9 g / cc)

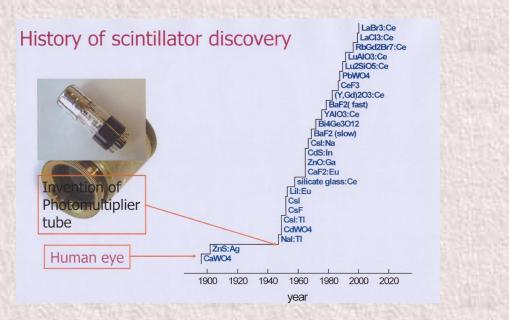
- inorganic crystals possible with large Z (La, Ce, Cs, Bi,Cd)
- speed (ns-microseconds)
- special shapes possible
- usually no cooling needed (unlike HpGe)
- can be relatively inexpensive (compete γ- ray spectr). < 4k€





Some crystals easy machinable (soft) like CsI(TI)



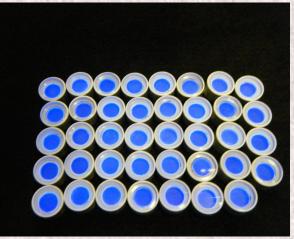


PLUS e.g. CLYC (Ce) Srl₂(Eu) CeBr₃ CLLB + other epasolites GAGG:Ce family La:GPS (Ce) and others...

Last 10- odd years <u>many</u> new (in)organic scintillation materials were discovered/published, most of them have not made it (yet) into industrial production...

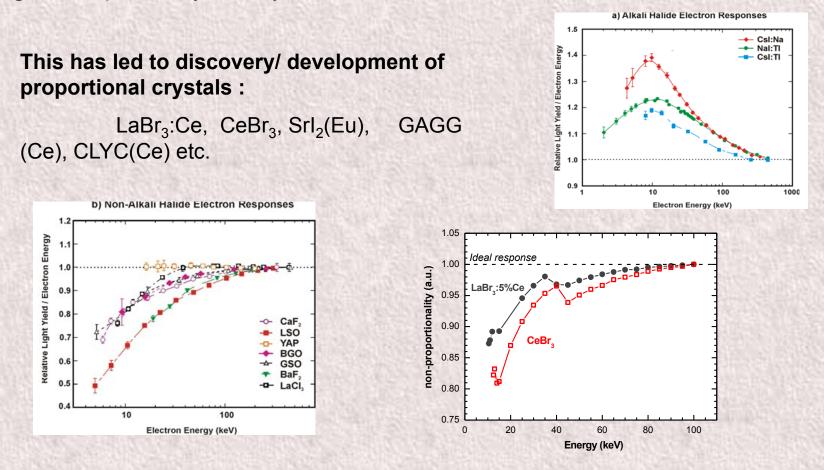
Q. What do we mean with novel/better scintillators ?

Q. What do we really need ?



It all depends on application !

scintillator research very much focused on **improved energy resolution for 662 keV** gammas pushed by security needs.....



Of course we would in general prefer : fast, bright, crystals BUT ..this is not always needed. Some other things we would like to see e.g. :

Environmental (gammas < 2.6 MeV)

~3-4 % energy resolution @ 662 keV as $LaBr_3$:Ce BUT low background as CeBr₃





Problem : cost !



e.g. In **X-ray spectroscopy** we really need fast (tens of ns) bright (> NaI(TI)) crystals at < 10 keV energy to allow quicker analysis Requested count rate MHz. CeBr₃ but high LO at low energies

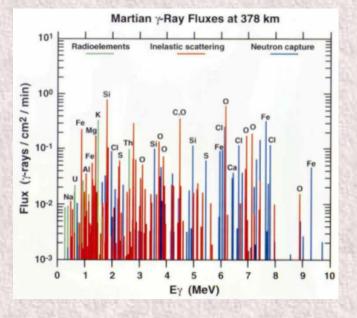
Oil Industry :

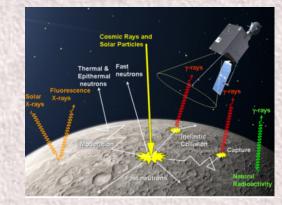
Porosity, saturation , densiry, multi phase flow

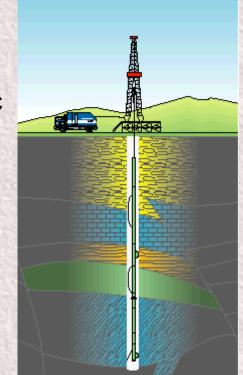
- Fast, High T high resolution crystals : LaBr₃:Ce, CeBr₃ O.K.

But also <u>(fast) plastics</u> able to operate (longtime) at 100 degrees C (MHz counting). Not available...

<u>Space</u>: High resolution low background (k-40) < 30 MeV, CeBr₃ O.K.





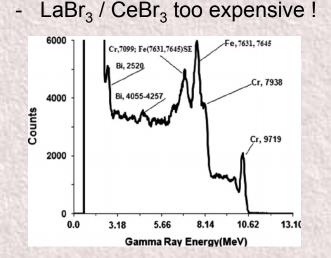


High Cost not real issue..

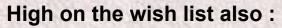
Mineral exploration using PNGA (coal analysis, cement)

- many kg crystals needed (> 5 kg per system)
- better energy resolution at 5 10 MeV, large Z required (comparable to BGO)

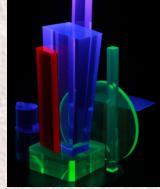








- organic scintillators with spectroscopic capability (W, Bi) under development
- Plastics with Neutron/ gamma capability (Li doped) idem



NEW materials imply new challenges

$\left(\frac{\Delta E}{E}\right)^{2} = \left(\frac{\Delta E}{E}\right)^{2}_{\text{sci, intr}} + \left(\frac{\Delta E}{E}\right)^{2}_{\text{stat, N}} + \left(\frac{\Delta E}{E}\right)^{2}_{\text{PMT, sci}}$

WHY?

For example

- Ideal (super) energy resolution is only achieved under optimum circumstances (maximum photoelectron detection and optimum homogeneity (items 1 and 3 in equation)

- **This all imposes :** selected QE or special high Cb cathode
 - PMT same size or larger than crystal
 - suitable glass or quartz windows needed
- i.e "Ideal" configuration for light detection is needed.

Not all crystal / readout configurations are possible to retain high resolution

For SiPm readout same : number of primary charge cariers needs to be optimised



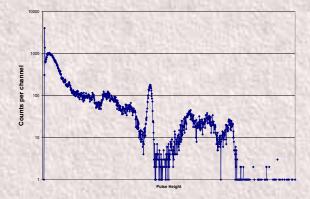


Where are we with application of "new" materials ?



LaBr₃ :Ce: Mature material used on > scale

High price prevents large scale us La-138 background is disadvantage Background 38x38 mm LaCl3 Feb 2006



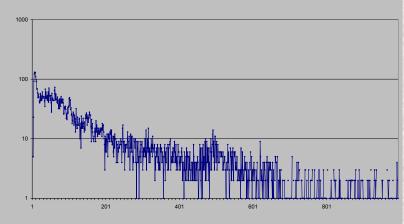
CeBr₃:

Hěllma[®] Materials

Absence of slow components allows use at MHz spectroscopy in plasma experiments

At high Multi (5-10 MeV) gamma energies resolution CeBr₃ only slightly worse than LaBr₃:Ce.





No beta background, very small alpha background (< 0.001 c/s/cc)

Rel. high cost (yield)

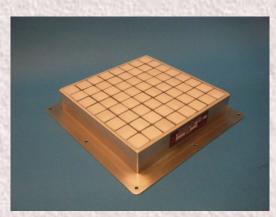
CeBr₃ used in :

Space applications (resolution)

Plasma Physics (speed)

PALS (time resolution)

Environmental (resolution / background)

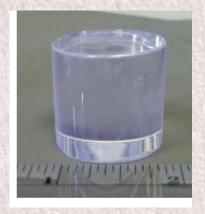


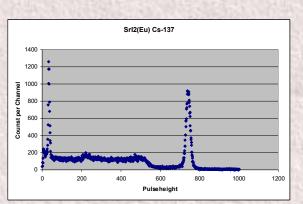
Time resolution comparable 127 / 119 ps Ackermann et al. (NIM A 786, 2015) 5-11 : energy resolution 5 % instead of 10 % @ 511 keV.

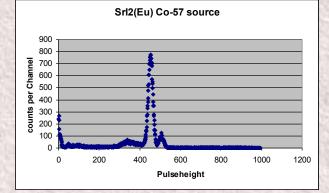
Srl₂(Eu) : very bright, proportional

small Stokes shift --→ self absorption ---- > larger crystals worse E-resolution !

Recently some very encouraging data were published though

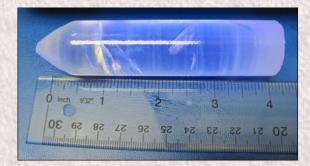






light yield very high ! → very good resolution at 100 keV Radiopure

Energy resolution approx. 3.5 % FWHM @ 662 keV Price relatively high = drawback Hardly used (yet) in commercial products



This crystal may find interesting applications in low background high resolution spectroscopy at low gamma ray energies !

More crystals of this "family" under development like

 $\frac{\text{KCal}_{3}(\text{Eu})}{\text{KCa}_{0.8}\text{Sr}_{0.2}\text{I}_{3}(\text{Eu})}$

Contain a lot of K-40 but crystals can be grown very fast (cost) (University of Tennessee)

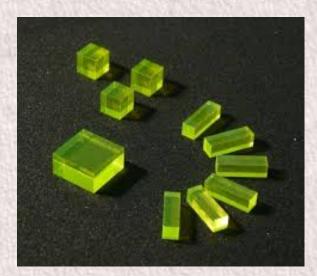
E-res 3.5 - 4.0 % @ 662 keV



GAGG (Gd₃Al₂Ga₂O₁₂) Ce

- 6.6 g/cc
- 520 nm max emission
- 56000 photons / MeV

Proportional, not hygrsoscopic FIRST non hygroscopic High LO crystal



High melting point (1850 °C) \rightarrow cost

Proportional, FIRST non hygroscopic High LO crystal

Not used frequently yet SiPm readout ? Special applications ?

No new generation scintillator will be the "ideal" material = illusion

Often bottom line is COST = growing yield

(material cost is seldom the real issue)

"Tuning" proportionality remains hot topic

Important factors for Industrial use are ?

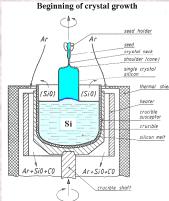
- what is the cost?
- can larger crystals be grown at usable size ?
- is the (superior) performance justified by the price ?



Crystal grown on Industrial scale remains a multiparameter system of which not all parameters can be always controlled 100 % :

- purity of materials (sub ppm) Background/absorption
- temperature during growth growing one crystal

- Inhomogeneities in dopant/cracks
- growing 100 crystals



Too tight specifications leads to (sometimes unaffordable) high cost !

 \leftrightarrow

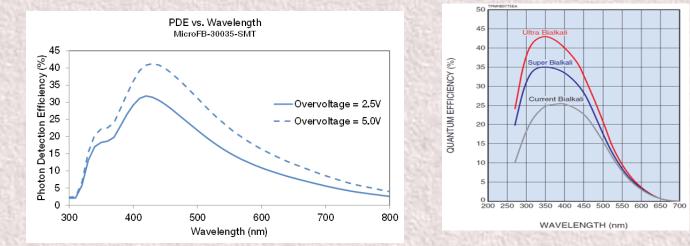
Price idea : Nal(TI) detector 76 x 76 mm : $2 \text{ k} \in 5 \text{ / cc}$ BGO detector 76 x 76 mm : 5-6 k€ 20 \$ / cc LaBr₃/CeBr₃ 76 x 76 mm : 35 k€ 100 \$ / cc For many applications standard inexpensive scintillators like e.g. Nal(Tl) / or standard plastic scintilators are 100 % sufficient and will not be replaced by novel scintillators due to low price of existing materials.

ADVANCES in scintillation light detection

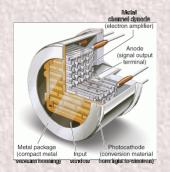
Range of possibilities for readout of scintillation light :

- PMTs
- (PIN) Diodes
- Avalanche Photodiodes (APDs)
- (Drift Diodes)
 - (EM) CCDs
 - Si-PMs (MPPCs)

The use of silicon technology allows higher wavelength emitting scintillators; PMTs dictated an emission in the 400 nm region.



Range of possibilities for readout of scintillation light :





PMTs

- Made of glass (fragile + K-40 background)
- Large signals, good S/N ratio, fast (ns)
- Large dimension, low price per cm²
- Sensitive to B fields
- Existing old technology (vacuum tubes)



APDs

- Amplification 100-1000
- rather Unstable (temp)
- rel expensive, small (max 10x10mm)

Silicon Photomultipliers (SiPMs, MPPCs)

PMTs

- (PIN) Diodes
- Avalanche Photodiodes (APDs)
- (Drift Diodes)
- (EM) CCDs
 - Si-PMs (MPPCs)

PIN diodes

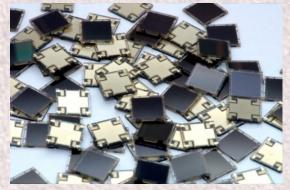
- No amplification (small signals)
- Maximum cm size
- Stable (temperature)

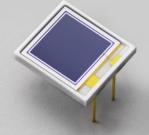
CCDs :

- DC measurement mostly
- Imaging
- For higher radiation fields

Drift diodes (for light detection)

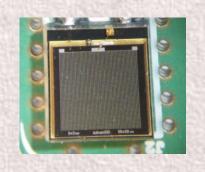
- small (not often used)
- still rel. expensive





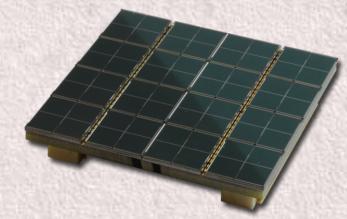
Large improvement of SiPms since 1.5 year does open up many possibities :

- Higher PDE
- Lower noise



PRO's

- Large gains (10⁶), fast
- Affordable cost (6x6 mm< 50-100 €) (mass production process)
- Higher basic QE than PMTs : Si
- Low voltage (30-70 V) e.g. ATEX



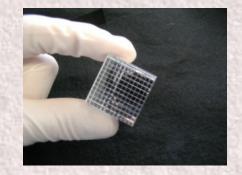
- high price per cm²
- filling factor

CON's

- pixel saturation easy (fast bright scintillators !)
- temperature dependent gain rel. large (+-1% / °C)

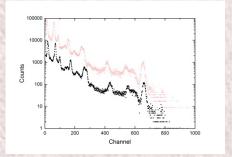
Readout of crystal pixels with SiPm obvious :

Med. Imaging (WLS) Fibers



Issues with SiPms :

A. Saturation / alinearity



Alinearity effects

- The higher the amount of pixels, the less problems with saturation. When all pixels fire = end of story
 Typical pixel size 35 micron = 19.000 pixels on 6x6 mm
 the slower the scintillator, the more pixels can recover
- to some level, energy calibration can be a solution

B. Limitation due to PDS / filling factor

- typical PDE for 6x6 mm is currently 52 % (more than UBA cathode)

C. Temperature behaviour

- Gain drift (typical -1.5 % per degree C)
- increase of noise
- D. Cost SiPm price per cm² is much higher than that of PMTs (100 € / cm²) compared to approx. 10 € / cm², 100 % scintillator coverage is not cost effective for larger crystals.

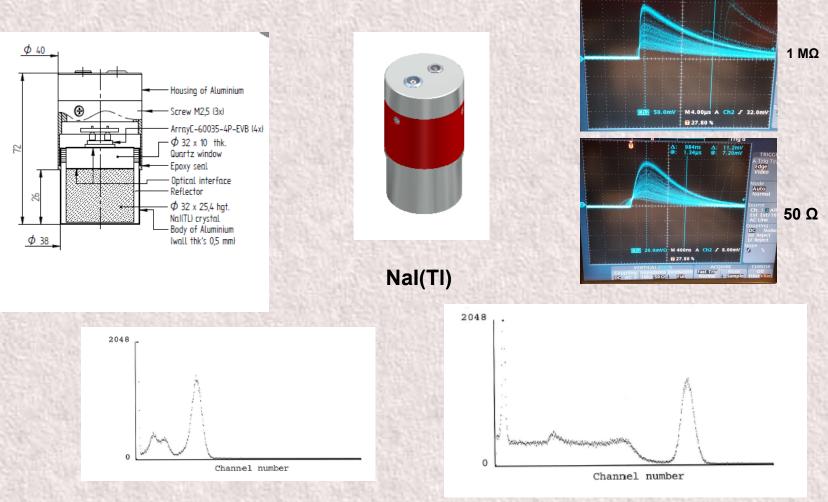
HOWEVER : p.e. statistics not always that important (e.g. with alkali halides)

→ Counting applications are fine

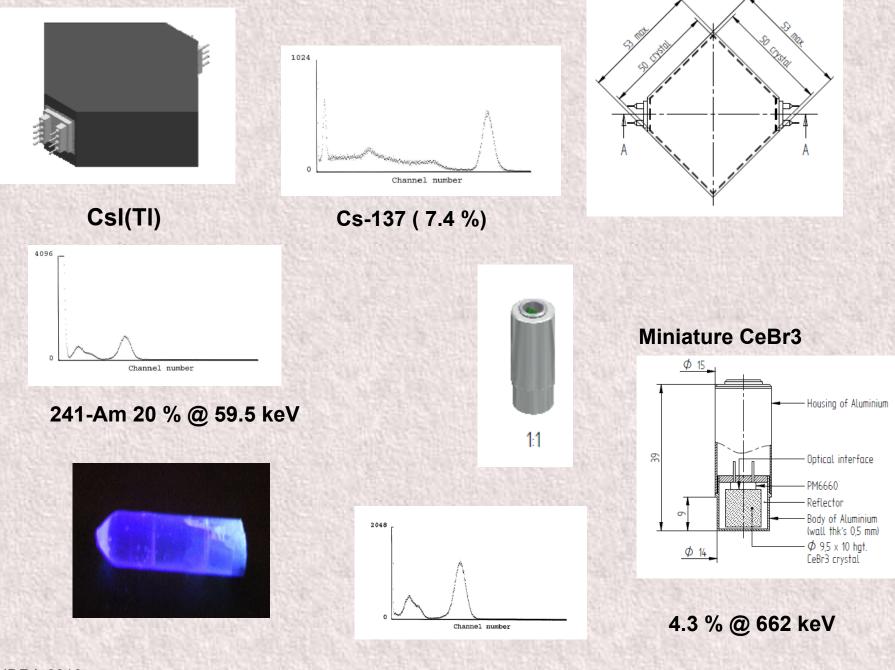
All parameters are totally different then with classic PMT readout

Am-241 20 % @ 59.5 keV

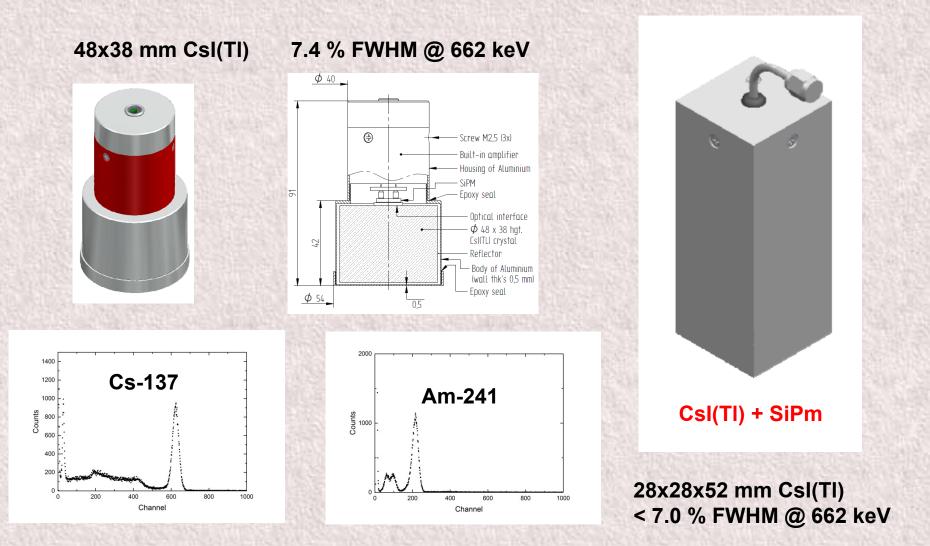
In many applications (especially with non proportional crystals), coverage of only a part of the scintillator with SiPm is sufficient for desired performance



7.5 % resolution @ 662 keV



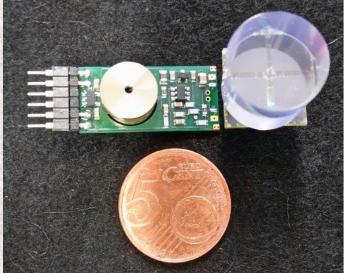
18.7 % @ 59.5 keV



When problems of saturation and temperature drift are overcome, current generation of Sipms are now usuable for numerous applications

Counting (e.g. densitometry) and limited spectroscopic applications wil be first

With little extra space, first order temperature compensated bias generator / preamplifier add a lot of functionality



Mechanical ruggedness of SiPms needs some attention

When using coincidence techniques also large volume plastic scintillators can be read out with arrays of SiPms (work in progress)

Detection of Neutrons :

	Physics (e.g. particle	e Physics, H	IEP)	
1	Security (SNM e.g. P	Pu, U)		
	Health Physics (dosi	imetry, ofter	n non spect	trometric)
Neutron energy	의 의원 문변 감독 전원	sed 0.025 eV rons > 50 k	State and State	
Interaction with Nucle				
Interaction with Nucle			A. Scatteri B. Nuclear	the state of the form of the state of the state of the state of the
- Elastic sca	ittering	(protons)		104
- Inelastic so	cattering +	prompt ga		10 ³ He
Nuclear Reactions e.g. : ${}^{10}B(n \alpha) {}^{7}Li$, ${}^{3}He(n p){}^{3}H$, ${}^{6}Li(n \gamma) {}^{3}H$,			nγ) ³H,	
	¹⁵⁷ Gd(n γ) ¹⁵⁸ Gd			$ \begin{array}{c} \begin{array}{c} \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\$

Most common detection method for neutrons

 ³He- tubes (pressurised) very unproblematic detector :

Easy to operate (no special electronics needed)

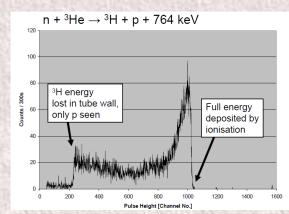
- gamma / neutron rejection > 10⁶
- No serious safety issues
- Large sizes possible (meters long)

World wide structural He-3 shortage \rightarrow availability / cost problems

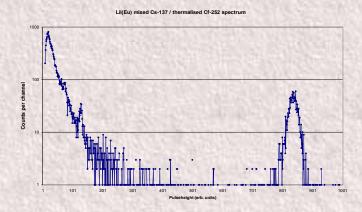
possible alternative : Detection of neutrons with scintillators :

- 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)



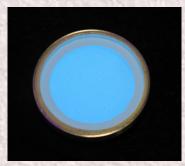


Usually , neutrons associated with gammas, both will interact with most scintillators but : Neutron + ${}^{6}Li \rightarrow$ alpha + Triton (4.78 MeV total) (particles !) \rightarrow Peak at e.g. > 4 MeV (In Lil(Eu))



Neutron / gamma separation possible via Pulse height.

3 mm Lil(Eu) absorbs 95 % of thermal neutrons



Lil(Eu) scintillator



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

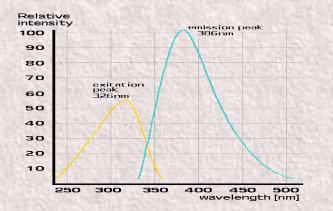
The ⁶Lil(Eu) alternative is an excellent solution for hand held instruments and dosimeters but is not an option for e.g. radiation portals

Alternative ⁶Li containing scintillators

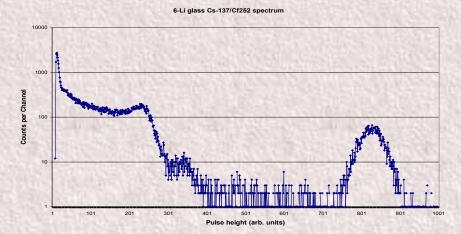
- ⁶Li Loaded glass scintillator (Ce doped)
- Low neutron peak location (approx. 1.8 MeV) implying more problem with gamma rejection
 - no large lengths possible (self absorption)

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)



90 % thermal neutron abs. in 1.5 mm



Neutron / gamma spectrum 6-Li glass



6-Li glass can be made in large sizes but cost is an issue

```
Cs<sub>2</sub>LiYCl<sub>6</sub>:Ce (CLYC)
( density 3.3 g / cc)
```

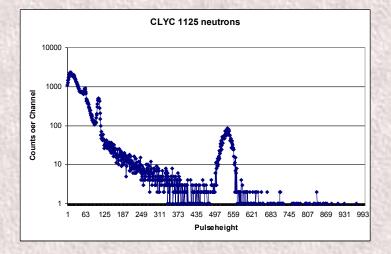
Proportional crystal:

Energy resolution approx. < 4-5 % (662 keV)

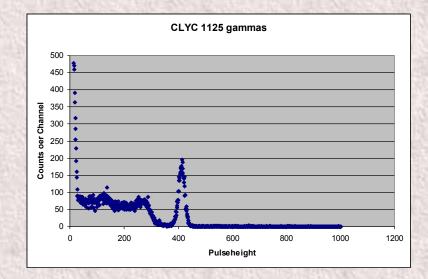
Neutron peak at 3.3 MeV

Fast neutron spectr. possible

12.7 mm needed for 90 % thermal neutron abs.



Cf-252 + Cs-137 spectrum in 1"x1"CLYC



Resolution approx. 4.5 % @ 662 keV



Problems :

- Cost (still) high
- Light output low
- Slow components

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

Digital electronics allows neutron gamma disc. based on PSD

CLYC used in identifiers more frequently. Cost prevent a wide scale use and replacement of Lil(Eu).

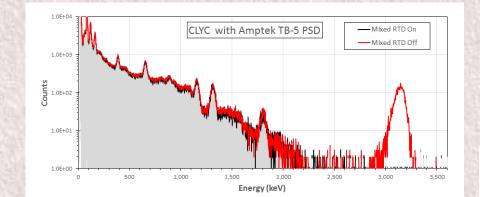
To achieve best energy resolution large (digital) shaping times are needed (? 4 µs)

CLYC ideal for small identifiers and PRDs

Gamma rejection with e.g. AMPTEK TB-5



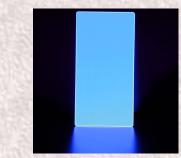




Large area neutron detection

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

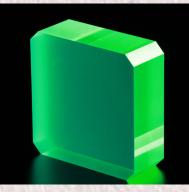


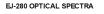
Detection Properties of Some EJ-426 Screens						
Theoretical NTH Efficiency						
0.5mm Thick						
0.41						
0.53						

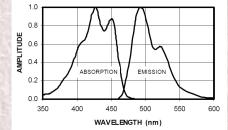
Problem is to get the light out (due to self absorption in powders)

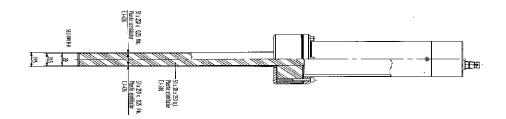
- Wavelength shifting (fibers) e.g. EJ 260 for large areas needed
- Due to non neglible gamma interaction in ZnS(Ag), PSD needed to obtain <u>He-3 comparable neutron</u> / Gamma discrimination

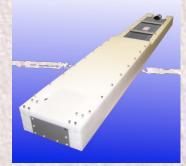
Commerical products available to replace He-3 in panels



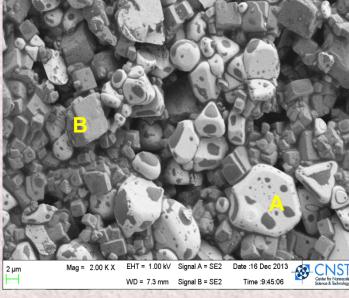








System may be furher optimized via grain size, binder etc. (ongoing R+D)



A- ZnS(Ag) grain B- LiF grain

D. Boron Loaded scintillators :

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

The other alternative :

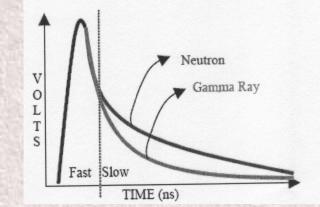
ORGANIC / Liquid scintillators

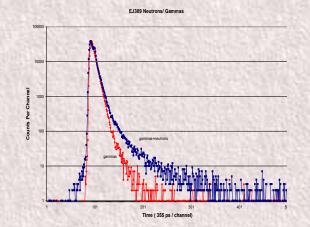
Some organic materials show <u>different pulse shape</u> for neutrons and gammas.

Liquid scintillators known since 1960's, most well known NE213(= EJ301 = BC501A), Xylene based.

Also organic crystals like **Stilbene**, **Anthracene** and **Para-terphenyl** are around for over 50 years. : "forgotten materials" untill recent !

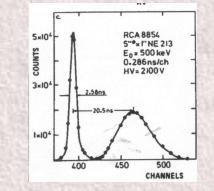






Different ways to do neutron/gamma separation with PSD

- 1. QDC with 20 ns and 1 microseconds gate (or different)
- 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

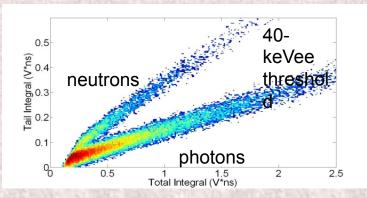
Other disadvantages of liquid scintillators :

- 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 :

open up applications



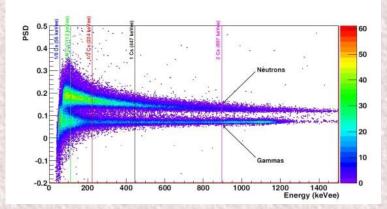
Advanced pulse sampling techniques using FPGAs allow on line 250 kHz throughput neutron / Gamma separation

> For Physics appications multichannel VMAE based fast digitizers are more frequently used

High flash point non toxic liquids New plastics with n-¥ PSD FPGA based pulse digitizing techniques







EJ301

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

Light output (rel. to antracene) :

Maximum of emission wavelength :

Photon yield / MeV electrons :

Properties :

No. C atoms per cc :

No. H atoms per cc :

No. electrons per cc :

Decay time short component

Decay time long components

Refractive index :

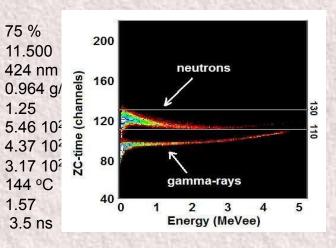
Density: H:C ratio:

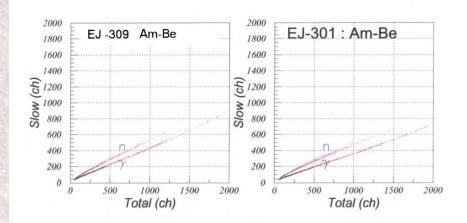
Flash point :

78 % 12.000 425 nm 0.874 g/ cc 1.21 4.0 10²² 4.810²² 2.3 10²³ 26 °C 1.50 3.2 ns 32.3, 270 ns

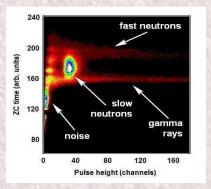
EJ309

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





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

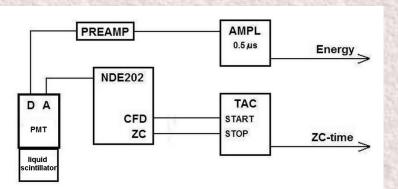


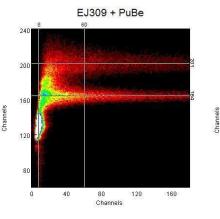
Boron peak at higher energies than with EJ301

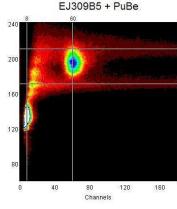
(100 keV versus 60 keV)

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

Courtesy Swiderski et al.





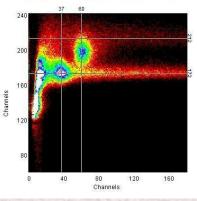


EJ309: N = 2530 phe/MeV @ 478 keV

EJ309B5: N = 1850 phe/MeV @ 478 keV

slow_n peak @ 100 keVee (!)

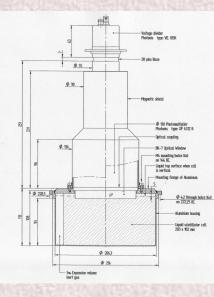
EJ309B5 + PuBe + 241Am + 60Co



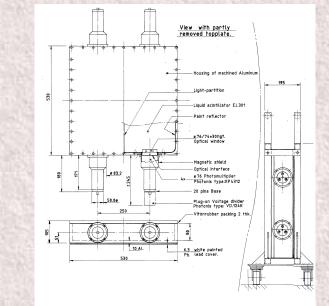
Design of Liquid cells for timing and PSD

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 orentations







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

SOLUTIONS THAT WORK

HOWEVER :

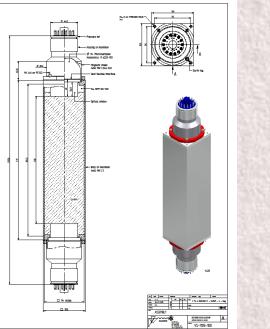
Neutron / Gamma PSD can be spoilt by a geometry where the light paths to the PMT are much different !







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



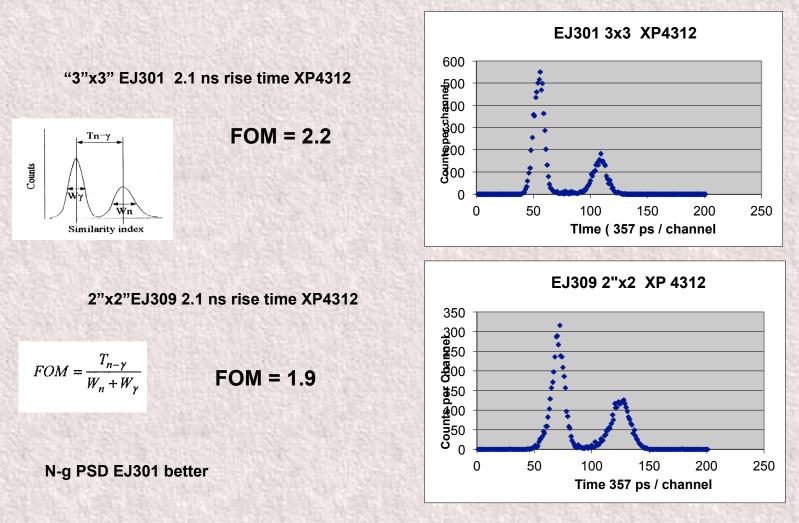
100x100x300 (2 PMTs)

O.K.

Some recent tests on several liquid scintilallator assemblies :

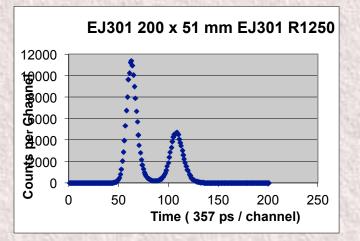
- What is influence of PMT on PSD ?
- What is influence of shape / PMT size

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



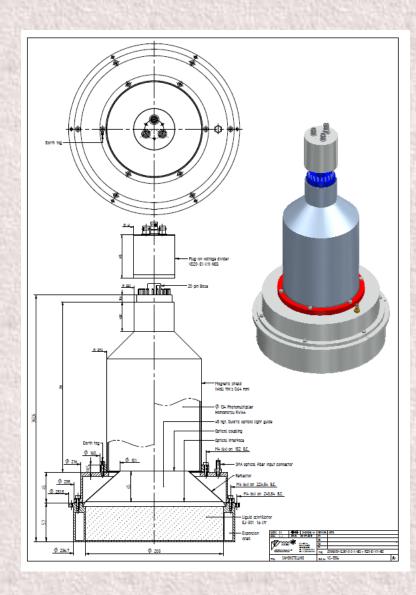
Larger cells, smaller PMTs

200x51 PMMA light guide R1250 PMT

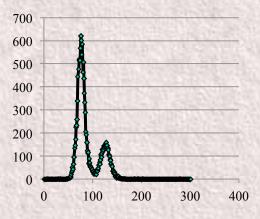


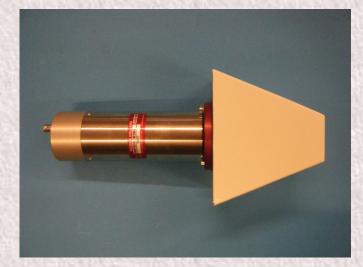
FOM = 1.88





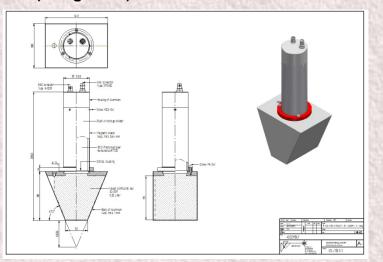
Pie shaped EJ309 cells 2 ns ET9214

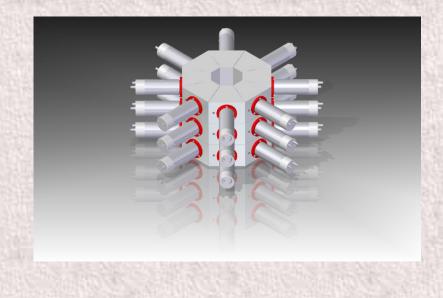




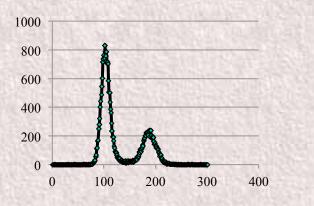
FOM = 1.22

Plutonium multiplicity counter (safeguards)

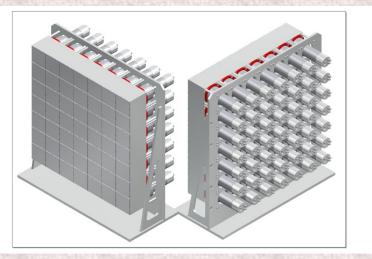


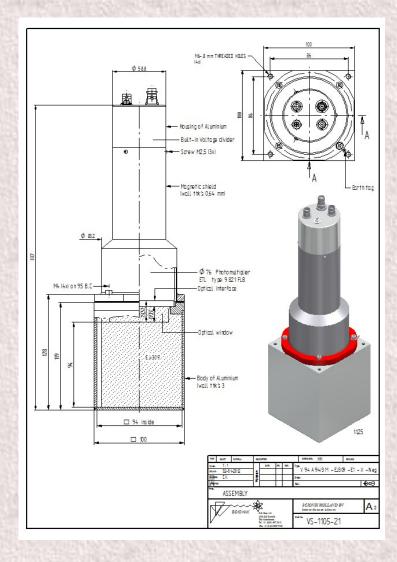


EJ309 94x94x94 mm 2.1 ns rise time ET 9821



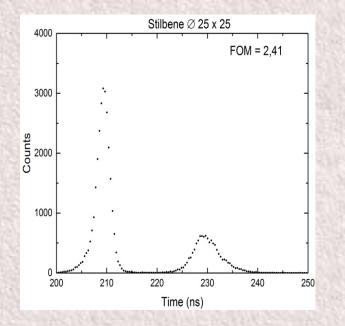
FOM = 1.75





organic crystals like Stilbene, Anthracene and Para-terphenyl are around for over 50 years. : "forgotten materials" untill recent !

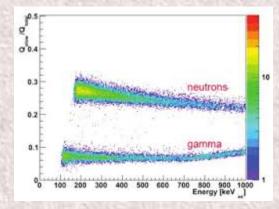
Recently renewed interest in organic crystals with PSD capability : Production on industrial scale (solution growth)

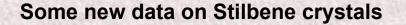


When price decreases promising material for neutron/gamma PSD with fast digitisers

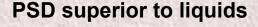


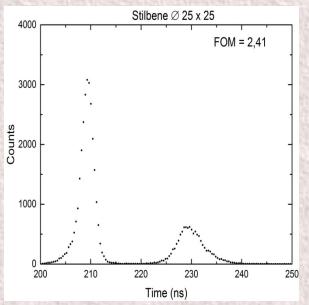
Solution grown Stilbene has almost 1.5 times more yield then bridgeman grown material (classic)

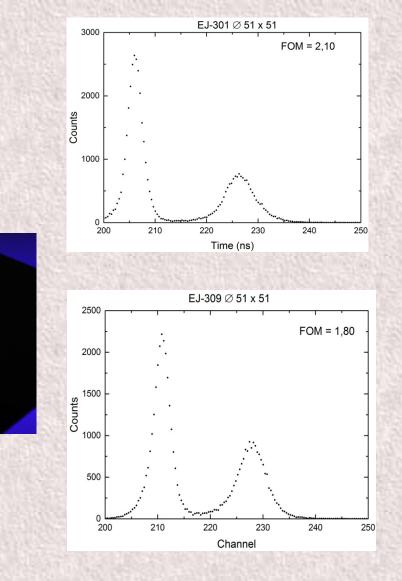




Recently renewed interest in organic crystals with PSD capability : Production on industrial scale (solution growth)







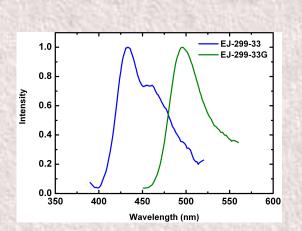
Stilbene remains rel. costly and toxic but other materials may follow....

Threshold in all cases 70 keV electron energy

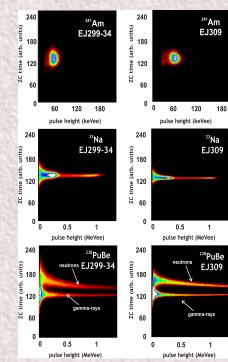
EJ-299-33A PSD PLASTIC SCINTILLATOR

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

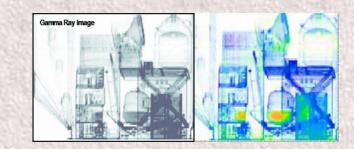


Also green emitting version avaiable





Decaytimes : 5 ns, 140 ns 150 ns (ratio diff. n-g)



Applied in neutron scanners

Very rapidly commercialised and widely used

Conclusions :

INORGANIC materials

- 1. Scintillator research flourishing ,many new materials developed, most of them not available commercially (yet). Cost remains issue
- 2. Li doped scintillators can replace He-3 tubes in some applications

ORGANIC materials

- 1. Since 50years organic scintillator research finally gets some interest ! More new organic materials may follow coming years.
- 2. The current availability of high flash point non dangerous goods liquid scintillators opens up possibilities to use these detectors where was prohibitive in the past.
- 3. The current availability of digital techniques allows the construction of novel instruments for neutron / gamma discrimination in mixed neutron / gamma fields
- 4. Liquid scintillators can be a replacement for He-3 tubes in **some** applications
- 5. Novel plastic scintillators offer neutron gamma separation via PSD; neutron / gamma separation not as good as liquids for can be adequate.
- 6. Organic crystals like Stilbene are getting back in the picture