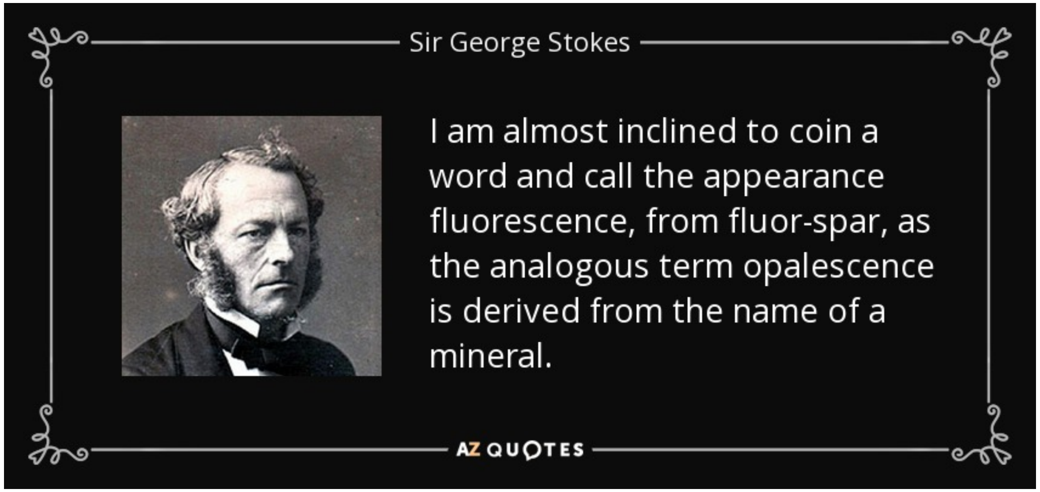
Scintillators
for neutron detectors: principles, traditional scintillators and emerging technologies

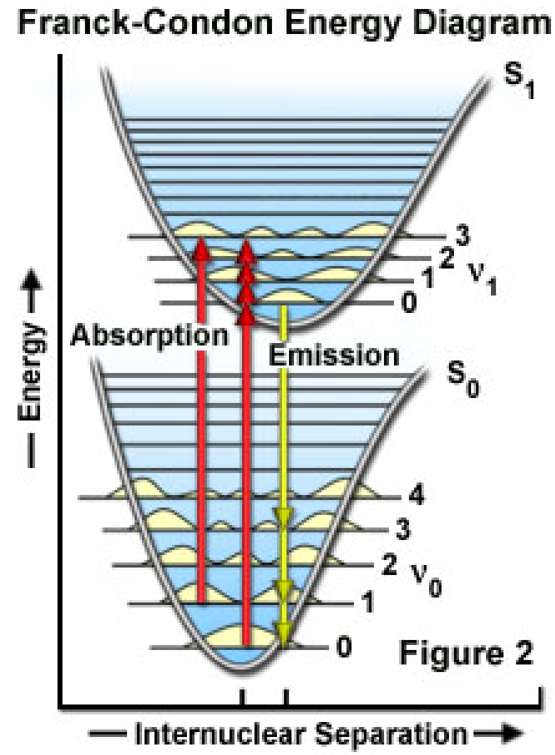
Francesco Mastinu, Sara Carturan

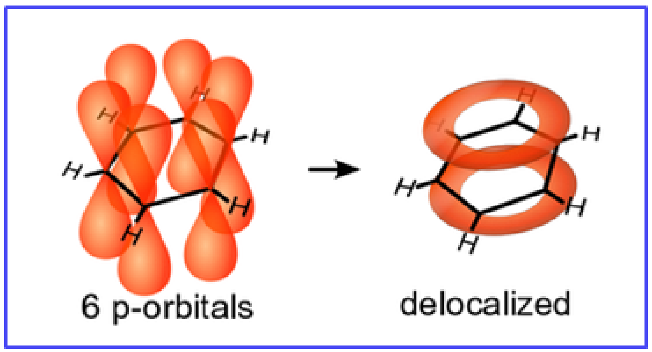
Outline

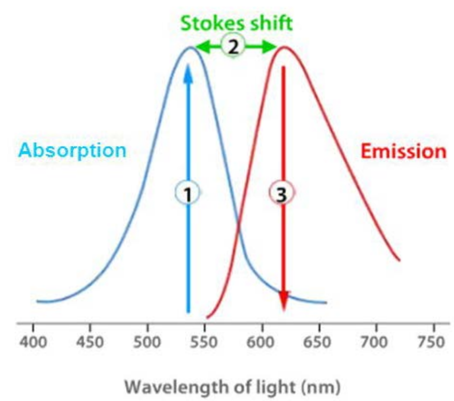
- Scintillator detectors: inorganic crystals, plastics and liquids
- Base principles of light emission in organic/inorganic materials: optical properties
- Design of a scintillator: energy transfer in plastics and liquids
- Ionization density: non-proportionality LY vs Energy→Ionization quenching
- Delayed fluorescence: basic principles of Pulse Shape Analysis
- PSA for n-γ discrimination: Liquid vs Plastic scintillators
- New organic scintillators: siloxane based (PSS)
- Solid PSS with PSD capabilities: effect of high Ist dye loading

Luminescence in molecules : basic principles



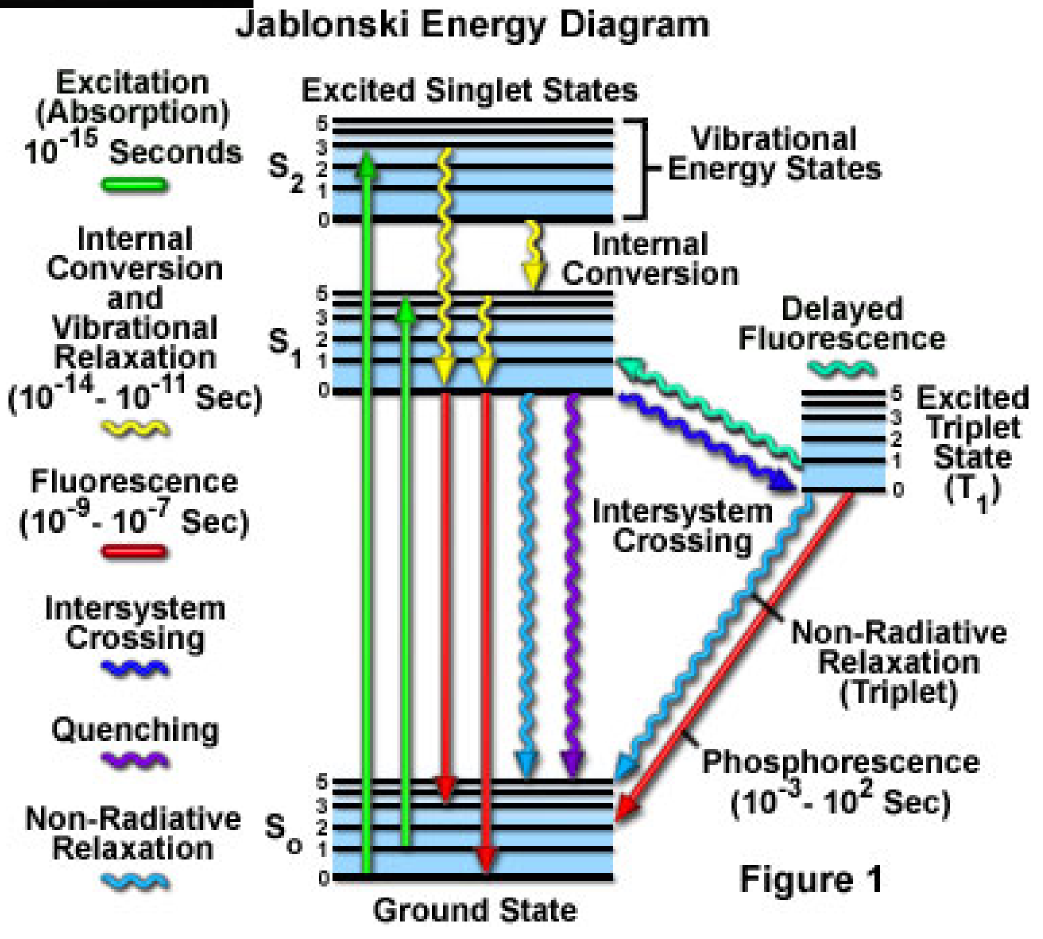






Molecules with delocalized electrons such as aromatic systems (benzene….) display fluorescence: absorption of photon (very fast, 10^{-14} sec, Eg some eV) from S_0 up to S_1 (higher vibrational levels), internal relaxation (fast, 10^{-12} sec), radiative decay by fluorescence (quite fast, 10^{-9} sec)

Radiative emission processes



Fluorescence is not the only enabled decay process….

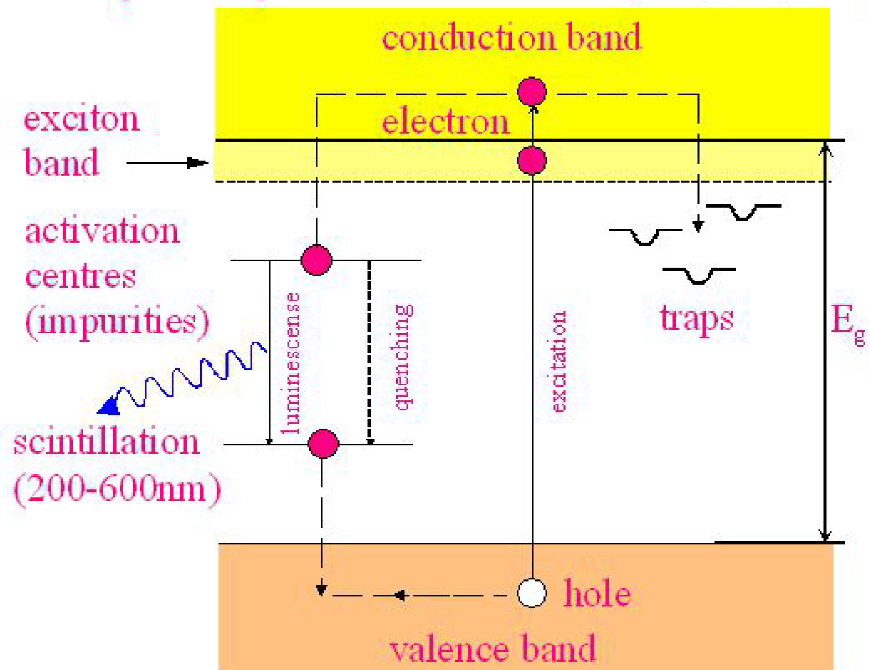
T1 population is possible (thought rate is very slow)→phosphorescence

T1 annihilation is possible (in case of high T1 concentration)→delayed fluorescence

PSD is enabled in those systems

Radiative emission in inorganic crystals

1a. Inorganic crystalline scintillators (NaI, CsI, BaF_2...



Band structure: VB CB, energy gap 5-10 eV

Excitation generates e-h pairs

Recombination →light emission: improved with doping: CsI(Tl)

Luminescence Decay Times:
ns-µs (activation)
ms (traps)

Longer τ vs fluorescence (larger energy gap)
ns-µs (activation)
ms (traps)

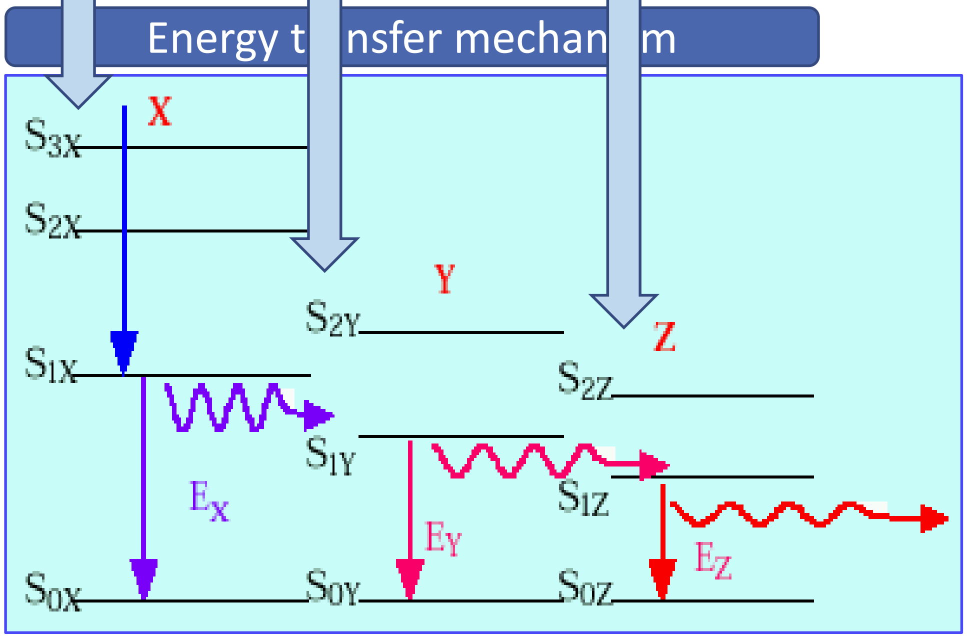
A GOOD organic scintillator: the basic RECIPE

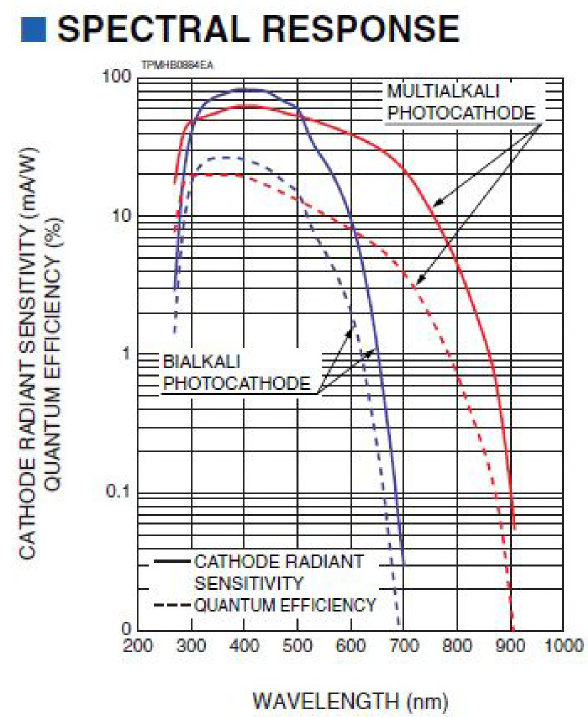
- **The cocktail (both liquids and solids)**

Solvent: absorbs the energy of impinging particle→UV emission (fluorescence)→ Energy transferred to 1st solute molecules

1st Solute (Primary dye): highly efficient fluor → **UV emission**

2nd Solute (Wavelength shifter): absorption overlaps with emission from Ist dye→emits in the **VIS range**→increase attenuation length (decrease of re-absorption phenomena)→better match with PMT sensitivity range (**350-450 nm**)

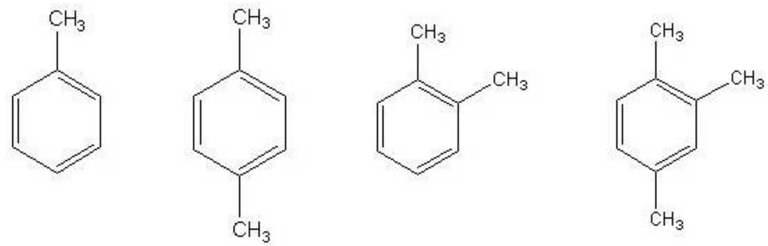




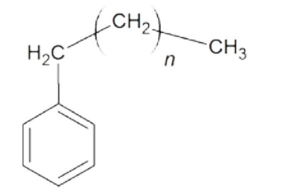
Organic scintillators: cocktails examples

Base solvents

Liquids

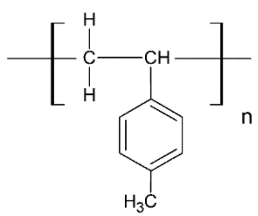


Toluene | p-Xylene | o-Xylene | Pseudocumene

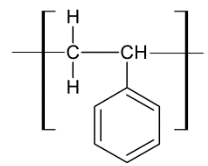


Linear alkyl benzene (LAB)

Solids



PVT (polyvinyltoluene)

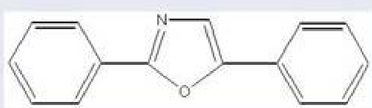
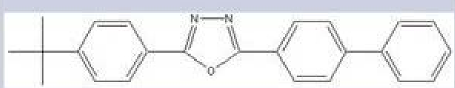
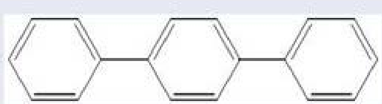


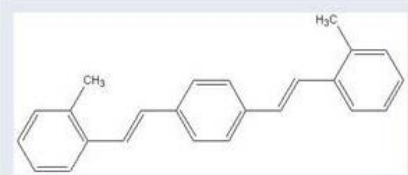
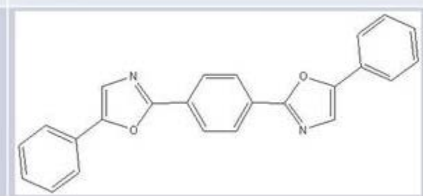
PS, polystyrene

Quantum Yield very low: 1-2%

Primary dyes (1-4 %wt.) QY ≈100%

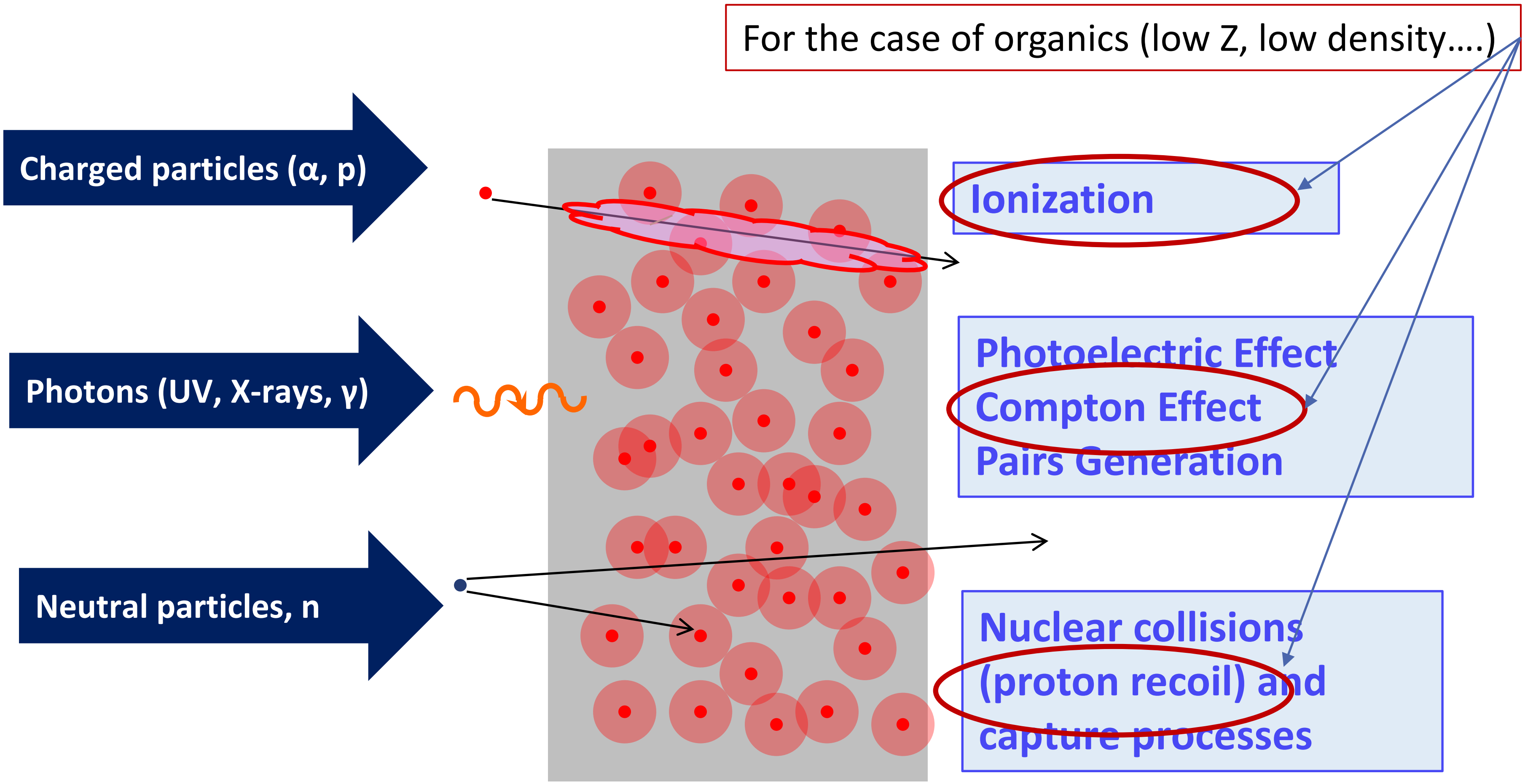
WLS dyes (<0.1 %wt.)

Scintillator	Molecular structure	Emission wavelength
PPO		357 nm
Butyl-PBD		363 nm
p-Terphenyl		340 nm

Scintillator	Molecular structure	Emission wavelength
Bis-MSB		477 nm
POPOP		410 nm

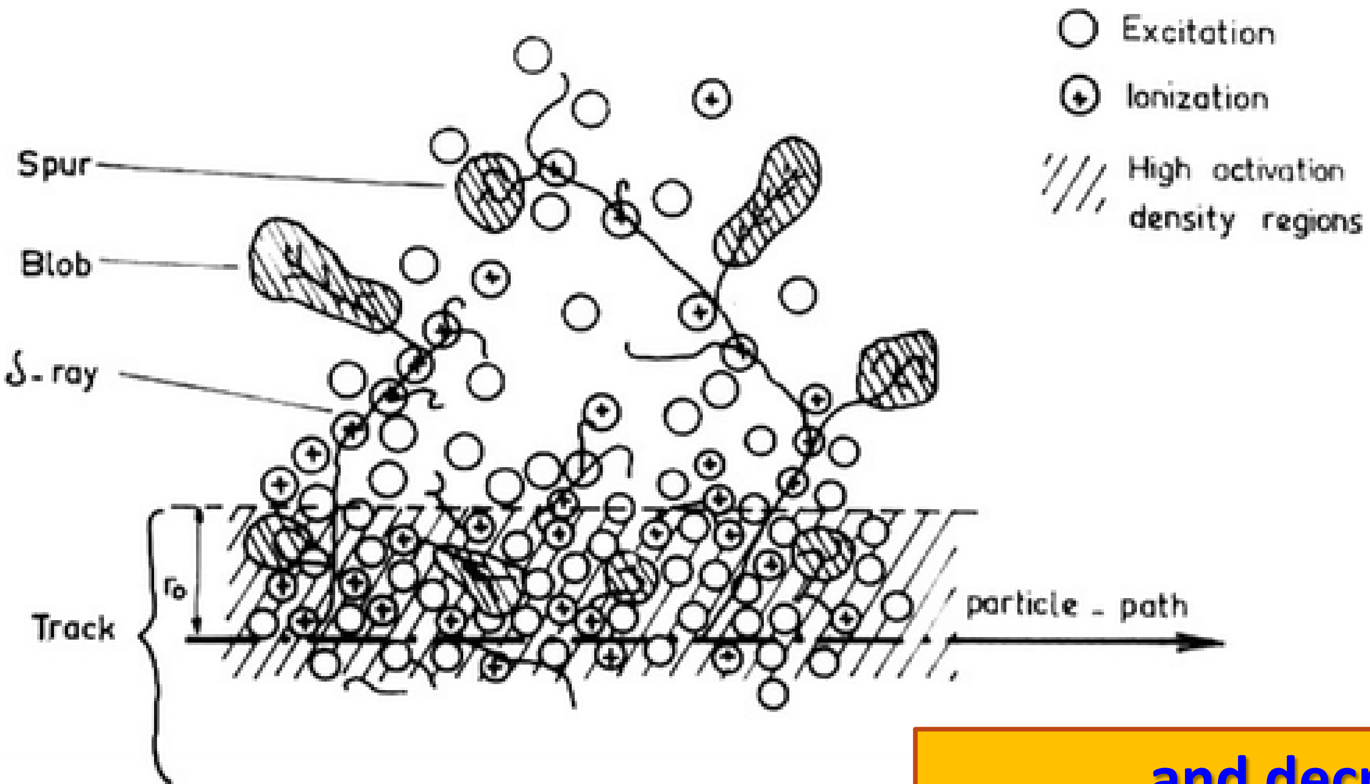
Scintillation → occurs when a particle looses energy through the medium producing excitation of the molecules and *radiative* de-excitation

- **Energy loss dE/dx** →can be a very complicated mechanism….(Bethe-Bloch)



Primary Ionizations: the ion kicks off electrons (Coulomb) from the nearby atoms all along its track→ molecules are excited up to very high ex levels (!)

Secondary Ionizations: fast secondary electrons (δ-rays) can efficiently ionize atoms far from the track→ again excitation but lower (in general) ionization density



High ionization density

Increase ionization *QUENCHING* !

and decrease LO…

Extent of Ionization Quenching and Light Yield

The extent of ionization quenching affects the Light Yield: different de-excitation mechanisms can take place→LY affected by particle's type and energy

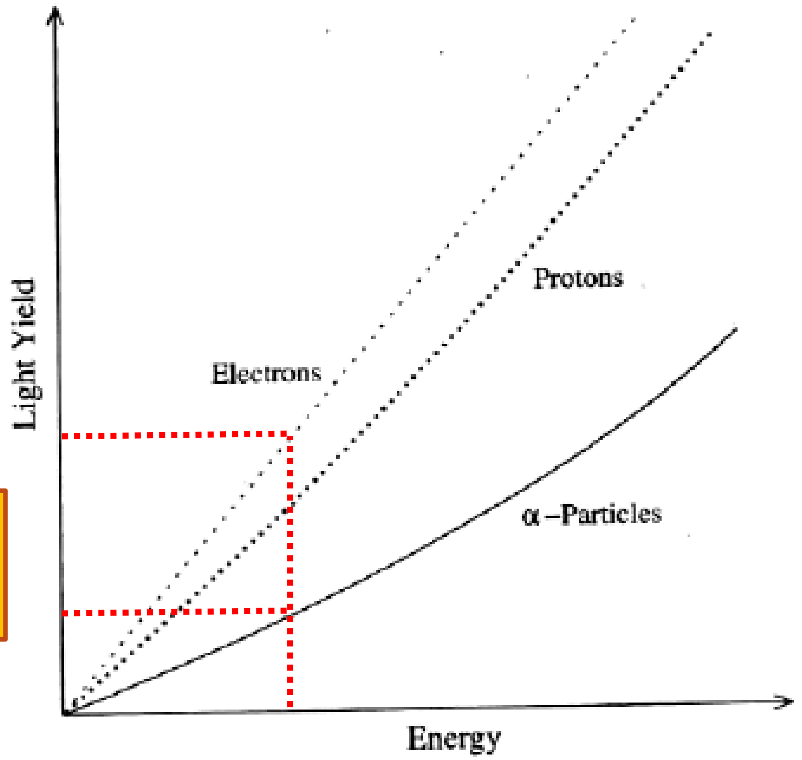
Light output for electrons (and γ-rays as well) is much higher than in the case of α with the same energy….

Higher ionization density leads to higher ionization quenching effects (non radiative decay channels are more probable to occur)

LY does not scale linearly with E (only for selected particles and conditions)

and the light pulse decay changes in shape as well… (PSD basis!)

LY for EJ212 (NE102)



Organic scintillators: a quick glance

STATUS of the ART

Plastic scintillators

- ❑ <u>good L.O.</u> (65% vs anthracene)
- ❑ different <u>shapes</u> and volumes
- ❑ <u>Fast</u> response time (2÷ 3 ns)
- ❑ relatively <u>low cost</u> (≈ 10€/g)

BUT ... Low radiation hardness (yellowing), **no PSD (but recently YES)**, sensitivity to **organic solvents** (crazing, swelling) (es. EJ212, NE102 etc.)

EJ212

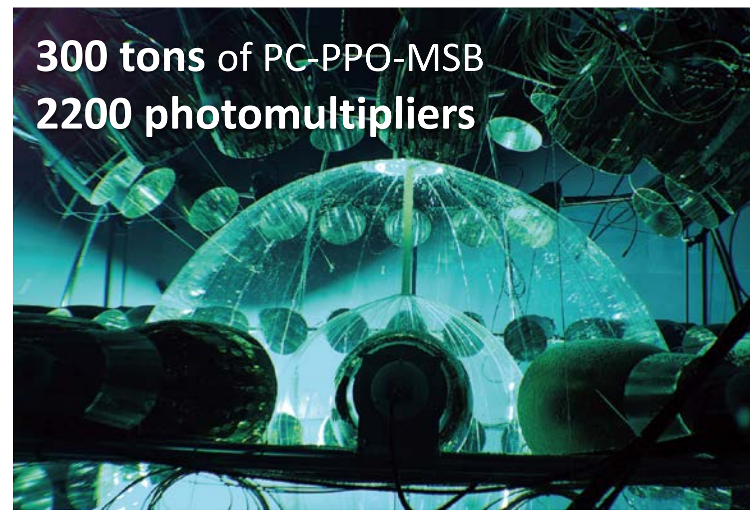
A sheet of NE102

Liquid scintillators

- <u>high L.O.</u> (80% vs anthracene)
- n-γ <u>pulse shape discrimination</u>
- <u>fast</u> response (3÷4 ns)

BUT...

Toxic, corrosive, **flammable**, explosive, high **volatility**, sensitivity to oxygen, **high cost**, **unfriendly** to environment (i.e. BC501, EJ301)



Borexino

OrIONe

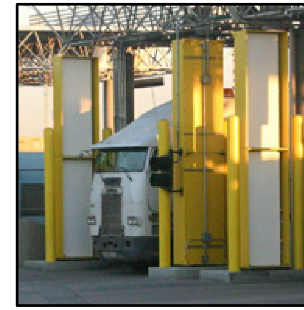
Neutron Detectors development: renewed interest

Wide range of applications

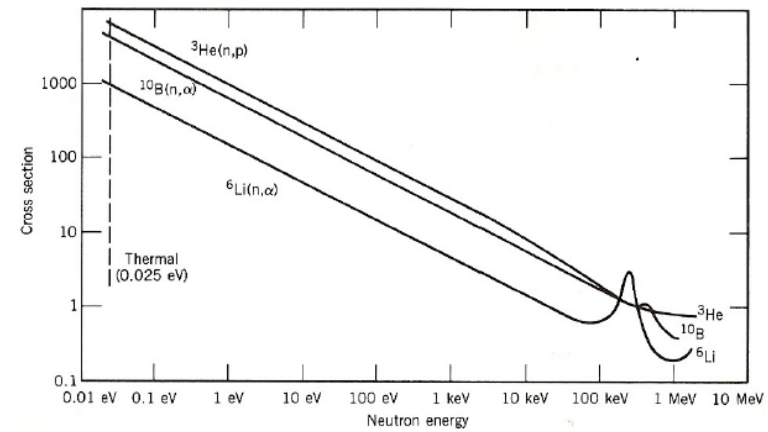
3He shortage

3000 €/liter

U.S. Energy Dept. 2009:
"No other currently available detection technology offers the stability, sensitivity and gamma/neutron discrimination of detectors using helium 3".



- ✓ **Detectors** in **Nuclear Physics experiments**
- ✓ **Monitors** in high neutron flux environments (Nuclear Plants, Spallation Neutron Sources, RIB accelerators…)
- ✓ **Homeland** security : **RPM→Radiation Portal Monitors**
- ✓ **Detectors** and **Monitors** for **Nuclear Medicine** (BNCT)
- ✓ **Materials Analysis** (neutron diffraction)



Organic Scintillators can work!

High Energy Neutrons (E>1 MeV) → Multiple scattering by Hydrogen and Carbon

Low Energy Neutrons (E<500 keV – 1 MeV) → Neutron Capture by B or nuclei dissolved in the siloxane rubber

$n + {}^6Li \rightarrow \alpha + {}^3He$ (Q=2.31 MeV)

$n + {}^{10}B \rightarrow \alpha + {}^7Li$ (Q=4.78 MeV)

$n + {}^3He \rightarrow p + {}^3H$ (Q=0.764 MeV)

Search for ….

- **GREEN scintillators** → non-toxic, non-flammable , eco-compatible
- **Resistant to chemicals, heat, radiation** → long-lasting transparency and L.O.
- **Able to detect fast AND low energy neutrons**→ doping with 6Li or 10B

inspired by the the work of Z. Bell and co-workers (Z.W. Bell et al., IEEE TNS 51 (2004) 1773; Z.W. Bell et al., "Organic scintillators for neutron detection," vol. 4784, Proc. SPIE 2002)

2008

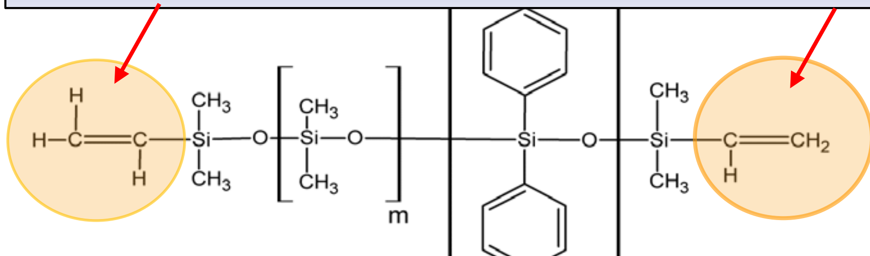
Production of polysiloxane based scintillators

- ✓ **Bio and Eco –compatible**
- ✓ **Elastomers:** maintain physical properties on a wide range of temperature (-100 up to 250°C), Tg of PS and PVT about 90°C
- ✓ **Resistant to most chemicals**
- ✓ **Resistant to radiation** (shown later)
- ✓ Can be modeled in **any solid shape**
- ✓ **Functional macromolecules** →suitable substituents along the siloxane chain → **phenyl groups** →**intrinsic fluorescence**

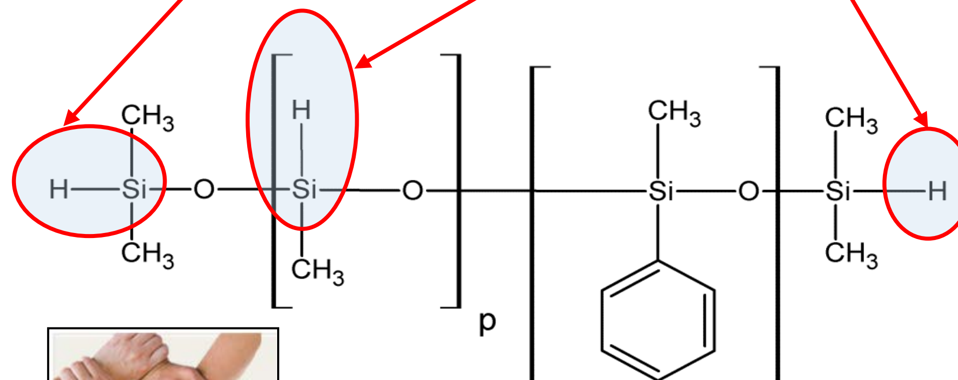
Polysiloxane synthesis: 2-part Pt-catalyzed addition

Reactive terminal vinyl groups – double bond

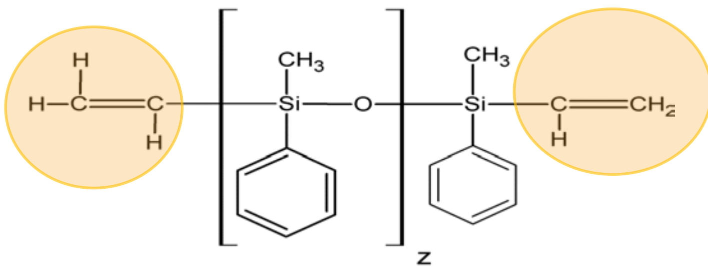
Cross-linker: reactive Groups Si-H



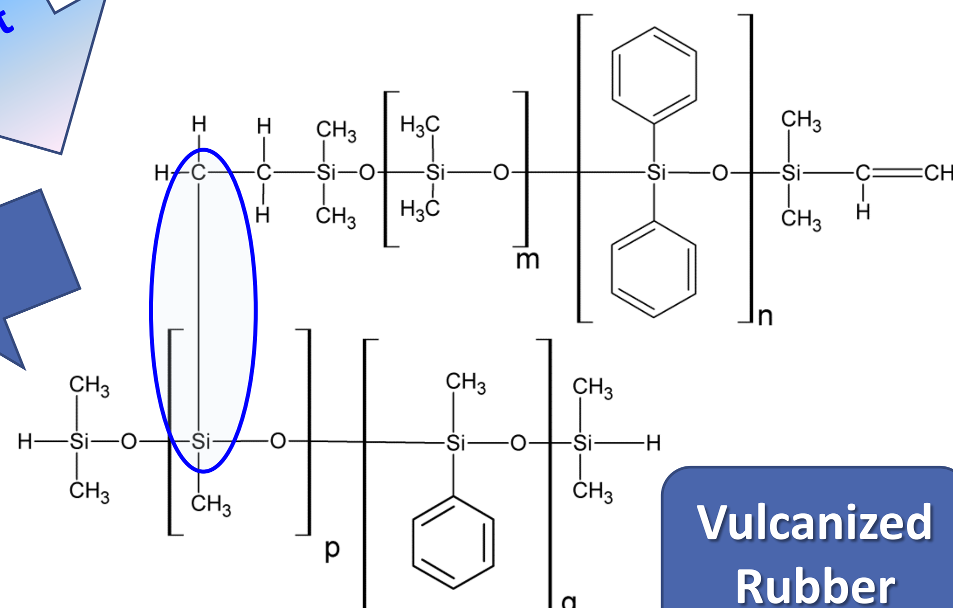
base resin A: n=22%



resin B: q=45-50%



base resin A: z=100%



- tailoring the amount of phenyl groups (Fluo, R.I., dyes solub.)
- mind the Pt content! (clustering...)
- Pt catalyst can interact with dissolved dyes

Vulcanized Rubber

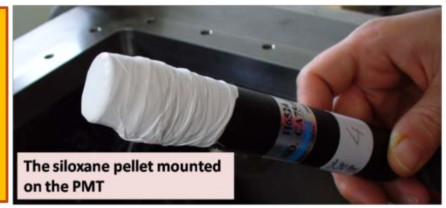
Blue emitting polysiloxane based scintillators

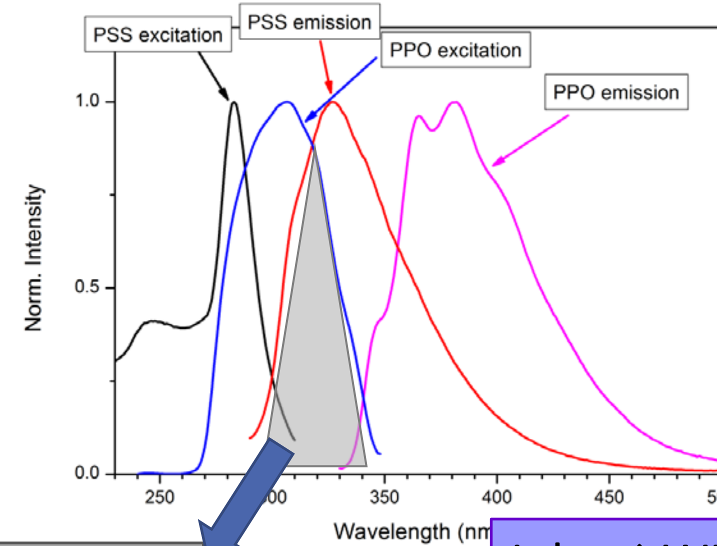
Quaranta et al. IEEE TNS , 57 (2010) 891
Quaranta et al. Opt. Mater. 32 (2010) 1317

Several combinations of base siloxanes, Ist dye and WLS (concentrations changing…)

- Optical features (exc ,em)
  - Design… energy transfer

- Light Yield vs EJ212
  - Alpha (^{241}Am source)
  - Gamma (^{60}Co source)

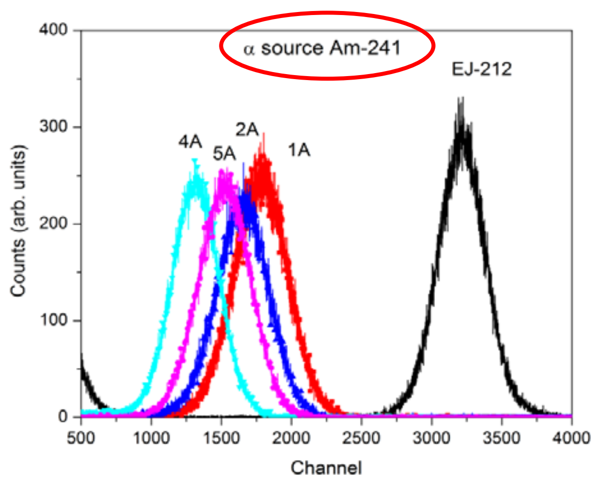


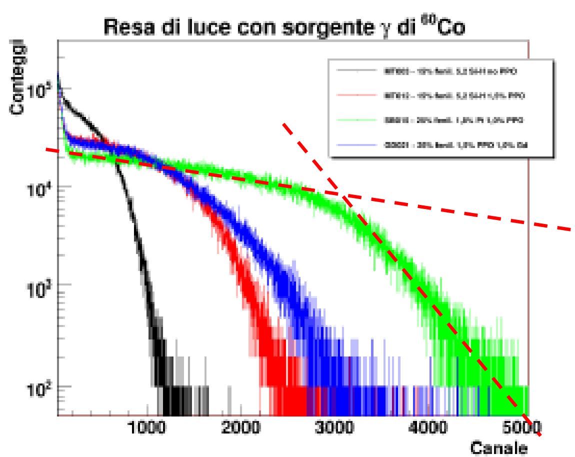


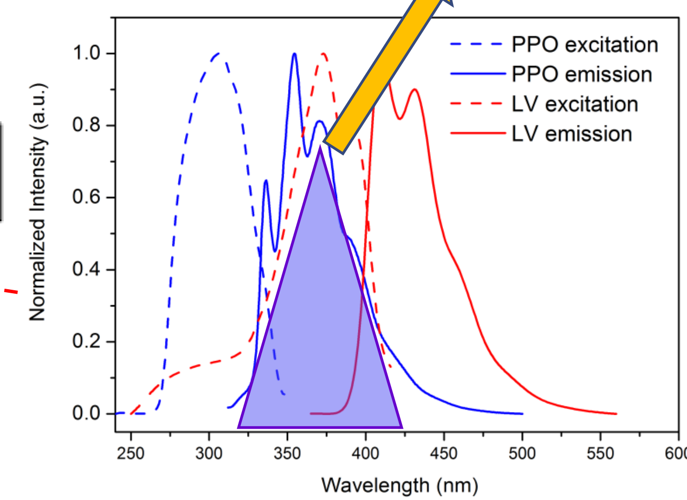
S. Carturan et al. Rad. Prot. Dosim. 143 (2011) 471

Matrix→I dye

I dye→WLS





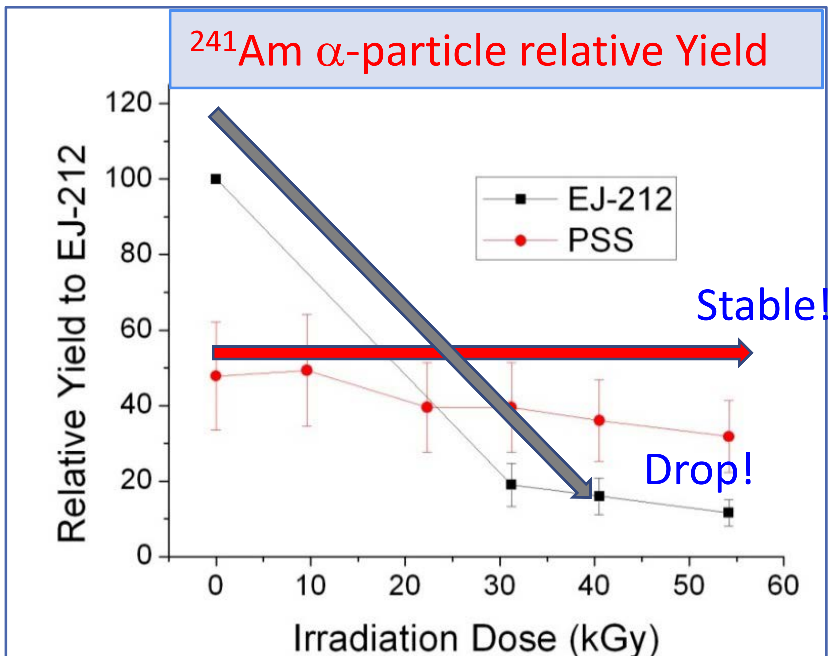


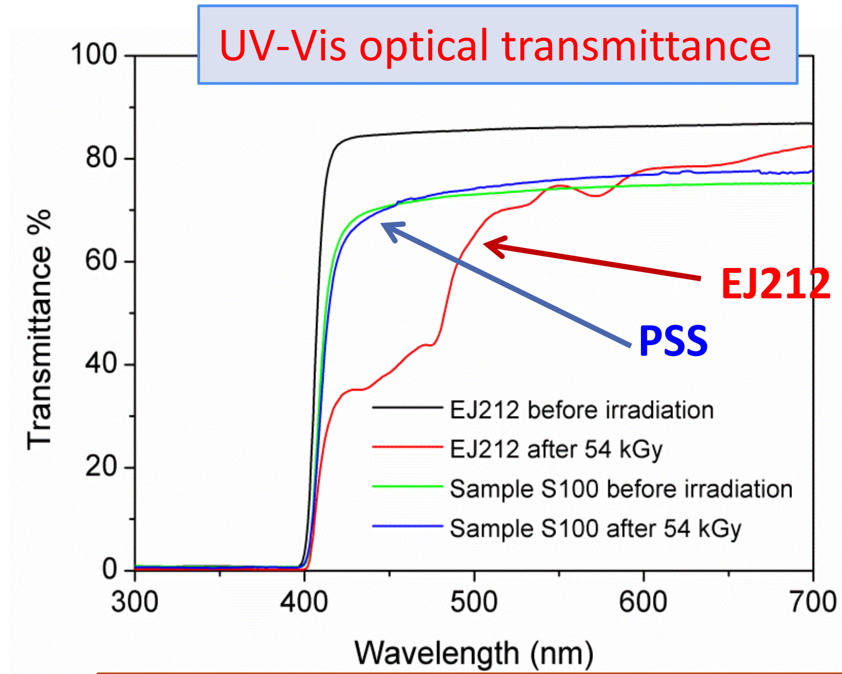
Polysiloxane based scintillators: performances

- ✓ Ease of manufacture, low cost (about 1 €/g)
- ✓ Functional groups → optical features tailoring
- ✓ Good dyes solubility → L.O. up to 70% EJ212
- ✓ Carborane solubility → fast/thermal n detected

^{60}Co γ-rays irradiated EJ212 and PSS scintillators (1%PPO, 0.02% LV); Calliope (ENEA facility)

Are RAD Resistant?





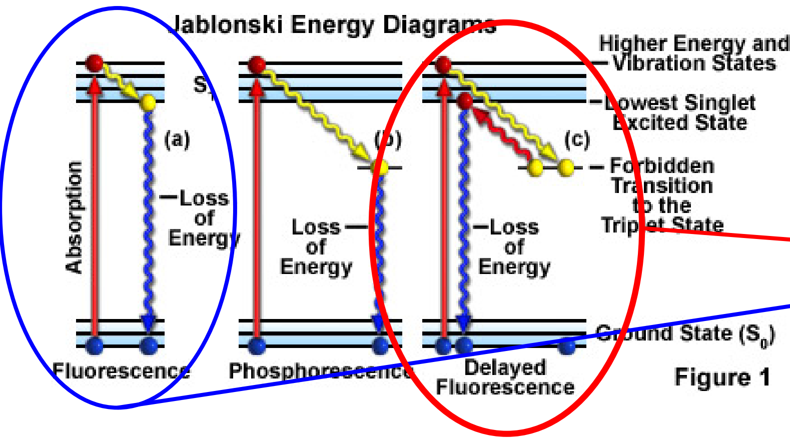
A. Quaranta, S. Carturan et al., *Mat. Chem. Phys*. 137 (2013) 951.

Higher Rad Res than EJ…

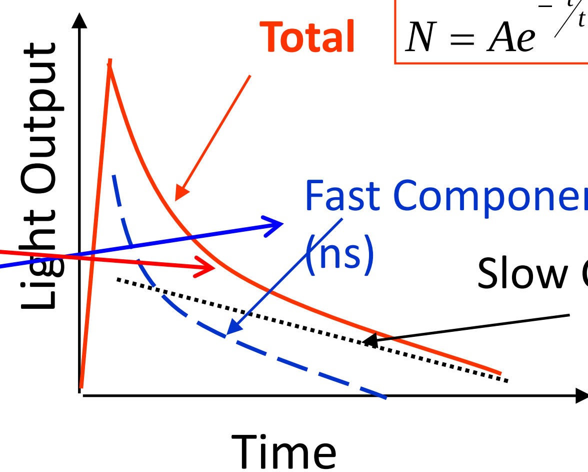
Pulse Shape Discrimination: basic principles

Is n-γ discrimination possible by PSA in PSS?

Different particles induce…. → **Different shape of light pulse**



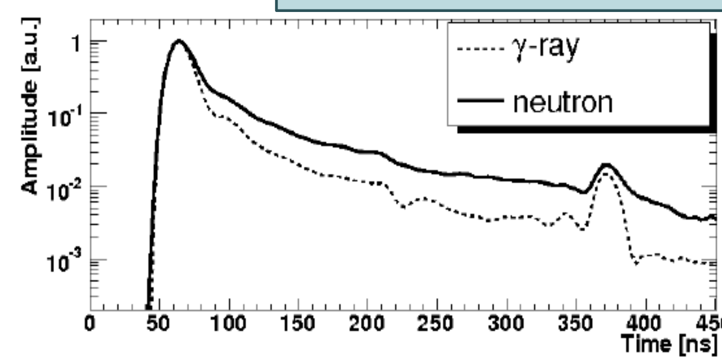
$$N = Ae^{-t/t_f} + Be^{-t/t_s}$$



Delayed fluo

Triplet-triplet annihilation
T1 + T1 → S0 + S1
S1→S0 + hν

Pulse shapes in BC501A



P. Söderström et al., NIM A 594 (2008)79

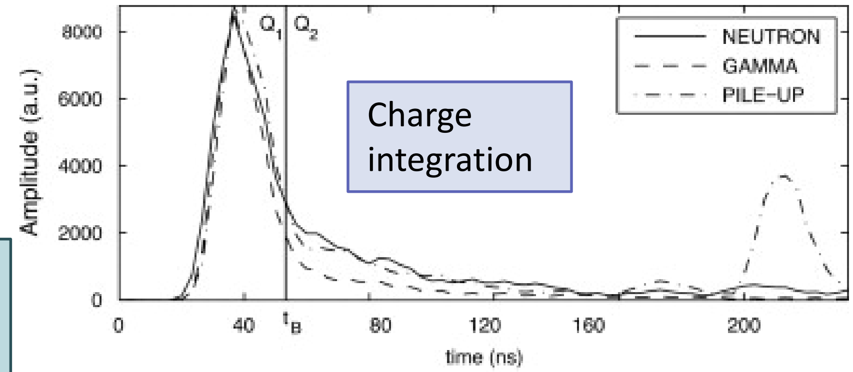
Incident particles with high ionization density (protons, alpha….) produce high concentration of T1 → increase slow component contribution →discrimination enabled

Pulse Shape Discrimination in solids ???

Several algorithms for n-γ PSD …

Literature on **LIQUIDS** only … T1 **diffusion**

Recent results on PVT **heavily** loaded with Ist dye (PPO): **INCREASED** concentration of T1!



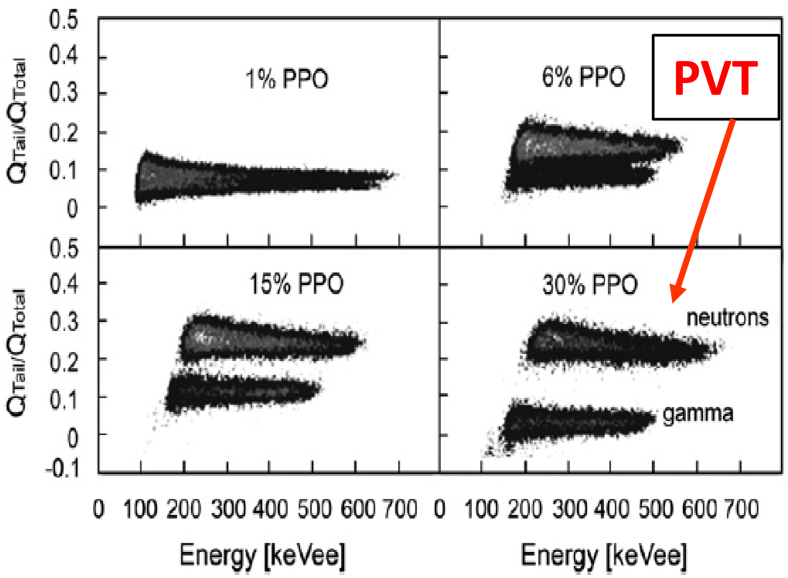
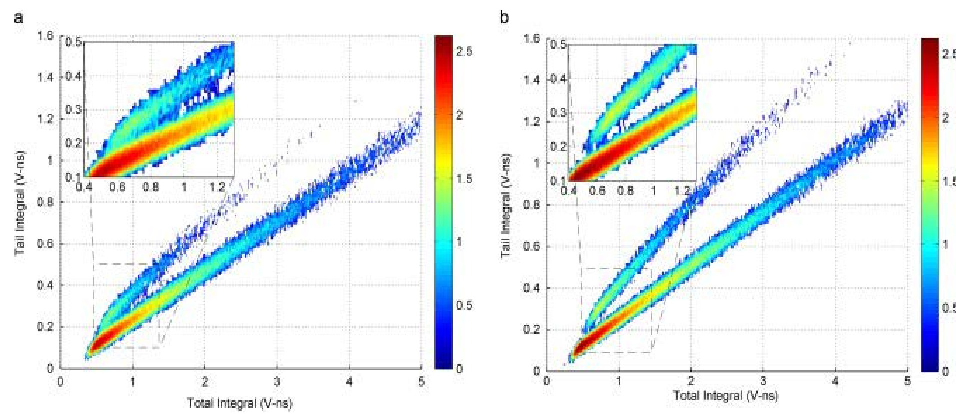


Fig. 3. Examples of experimental PSD patterns showing increase of neutron-gamma peak separation at increasing PPO concentration in a PVT polymer matrix.

N. Zaitseva et al., NIMA668(2012)88-93

PSD of γ-rays and neutrons using a 120-keVee threshold in EJ-299 (a) and EJ-309 (b)



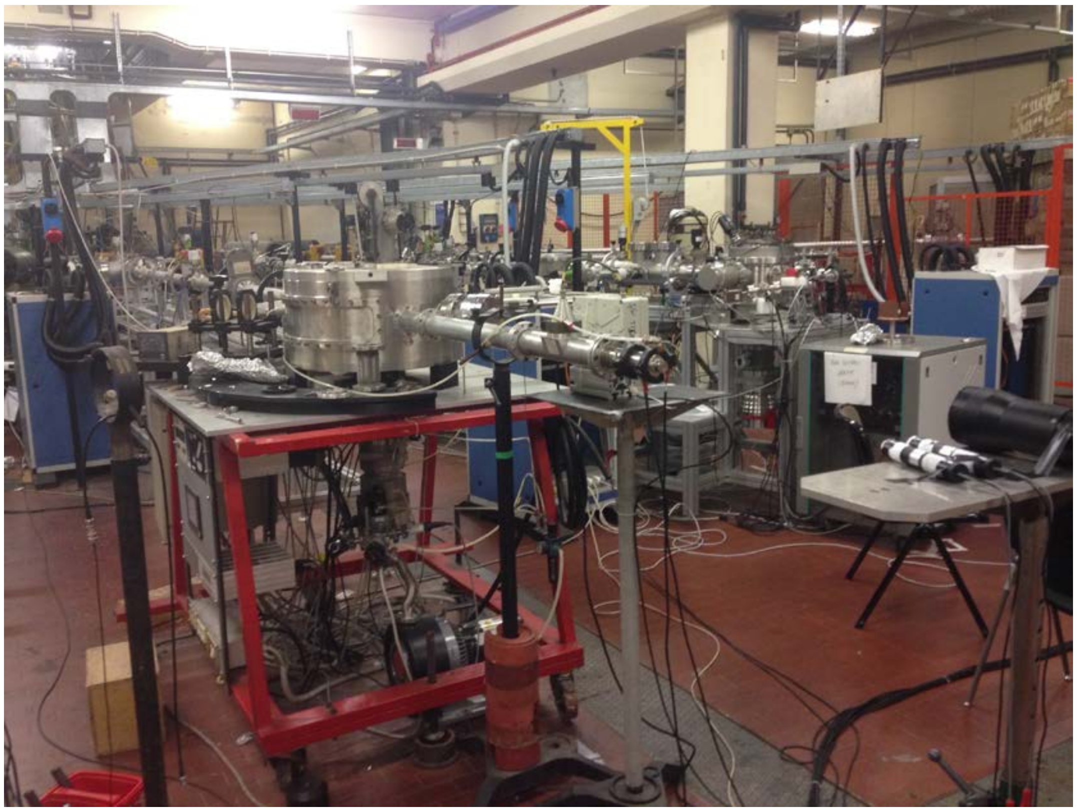
S. Pozzi et al., NIM A, 2013

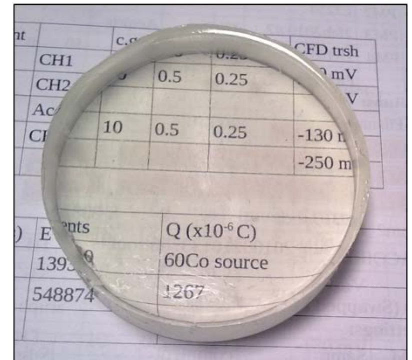
Could it work in PSS?

Loading with PPO… Experiment at CN accelerator

- Pulsed proton beam (2 ns, 3 MHz) on 7LiF target
- Proton energy 4.0 MeV → Main neutron peak 2.3 MeV 7Li(p,n)7Be (and a flash of γ-rays)

Testing PSD capabilities of…

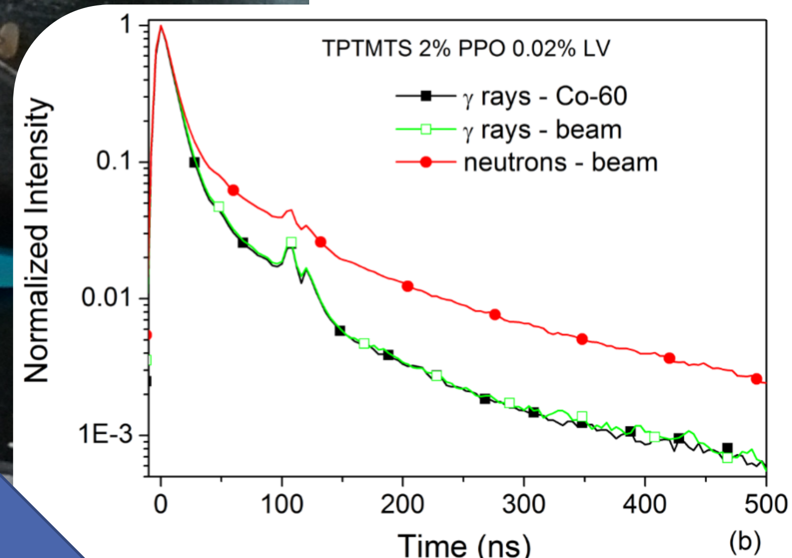




PSS 22%, PPO4%, LV0.02%

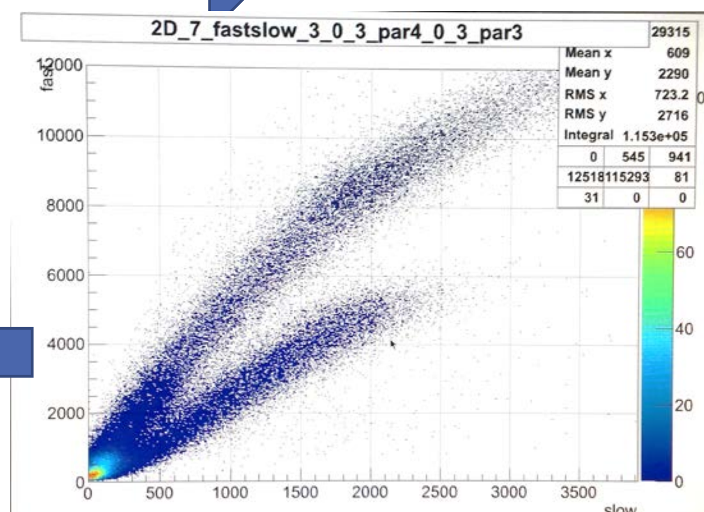
EJ-299-33

And compare them …



Pulse shape collection (thousands of waveforms)

Charge Comparison



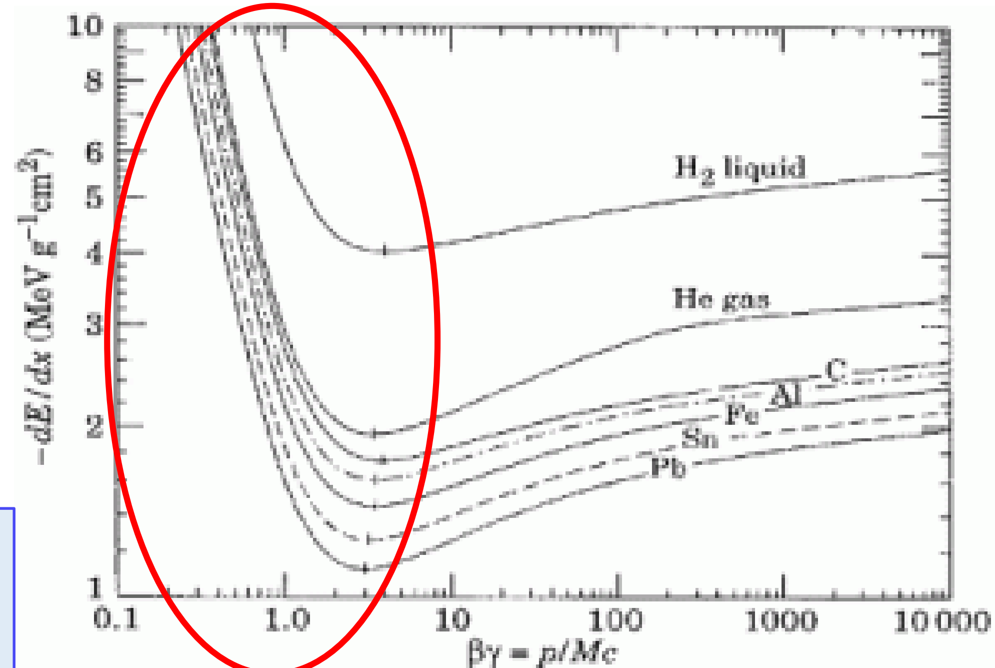
The floor to Francesco…

Spare Slides

- **Energy loss dE/dx vs Energy for charged particles** →Bethe-Bloch relationship (cumbersome…)

Maybe oversimplifying….

$$-\frac{dE}{dx} = \frac{4\pi e^4 z^2}{m_0 v^2} NB$$



Strong dependence on the atomic number of the impinging particle and its velocity

Slower moving particles, (i.e. particles with lower energy), lose energy more quickly as a function of distance!

This is due to the fact that slower moving particles when passing through a material have more time to interact with the electrons