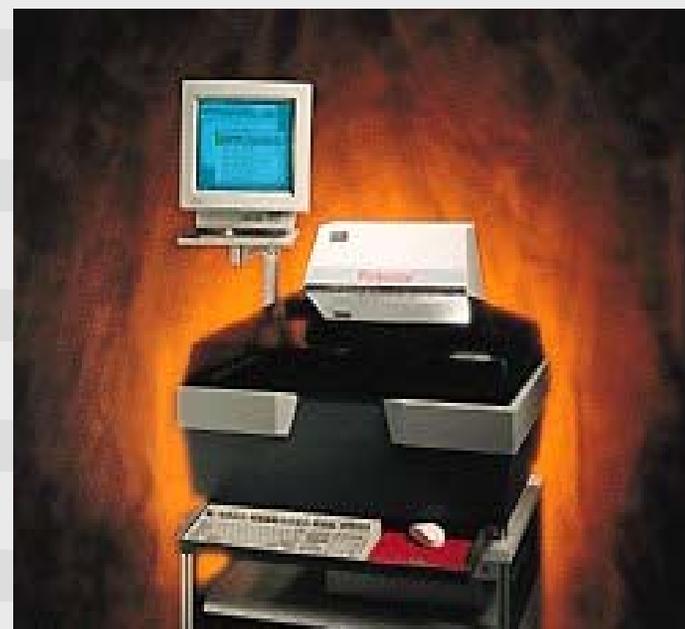


Liquid Scintillation Counting for the Determination of Beta Emitter — Principle and Application

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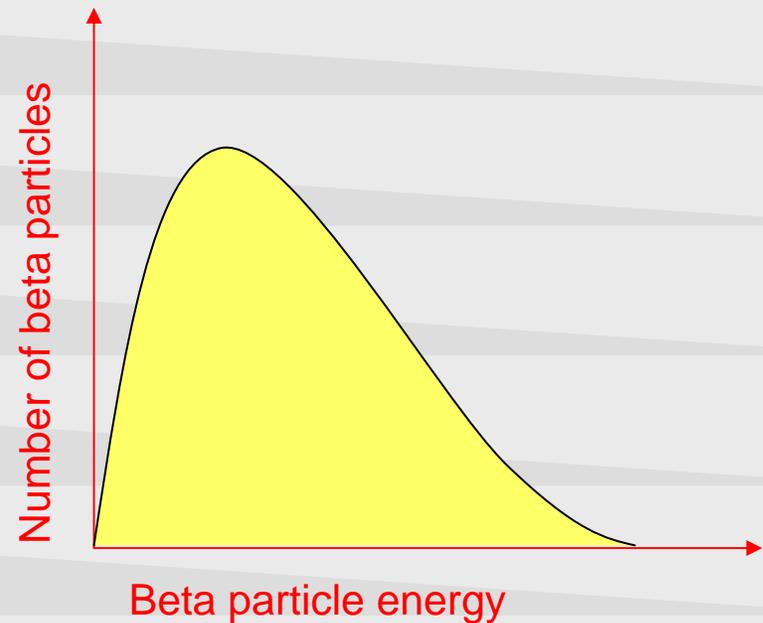
The Beta particles

- In beta decay two particles are emitted: an antineutrino ($\bar{\nu}$) and an electron, beta particle (β),



- The antineutrino shares the energy with the beta particle in the radioactive decay. The resulting beta radiation has a continuous distribution of energies from 0 to Max. decay energy.

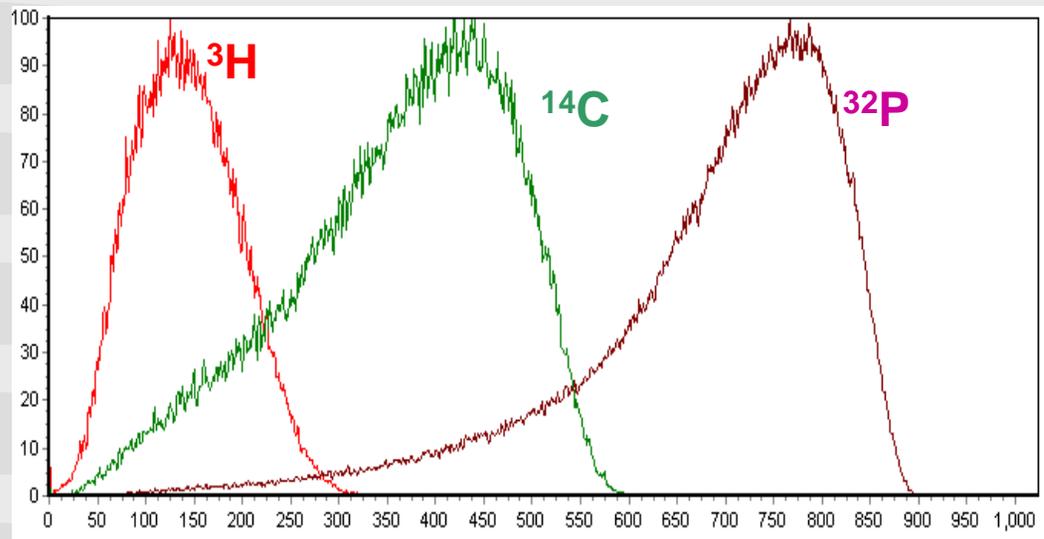
- Antineutrinos do not interact significantly with matter and thus do not feature in beta counting. However, beta particles can be detected by its reaction with matter. Normally, beta particles are detected by gas flow detector.



Detection of Beta Particles

- Due to the low penetrative power of beta radiation, the detection efficiency of beta emitter, especially the low energy beta emitters, are quite low, the quantification of beta emitting isotopes is quite difficult without LSC.
- The best possible contact is achieved when the sample is dissolved in the scintillation solution. By the counting the photons produced in the reaction of beta particles with the scintillator, the quantification of beta emitting isotopes can be easily carried out.

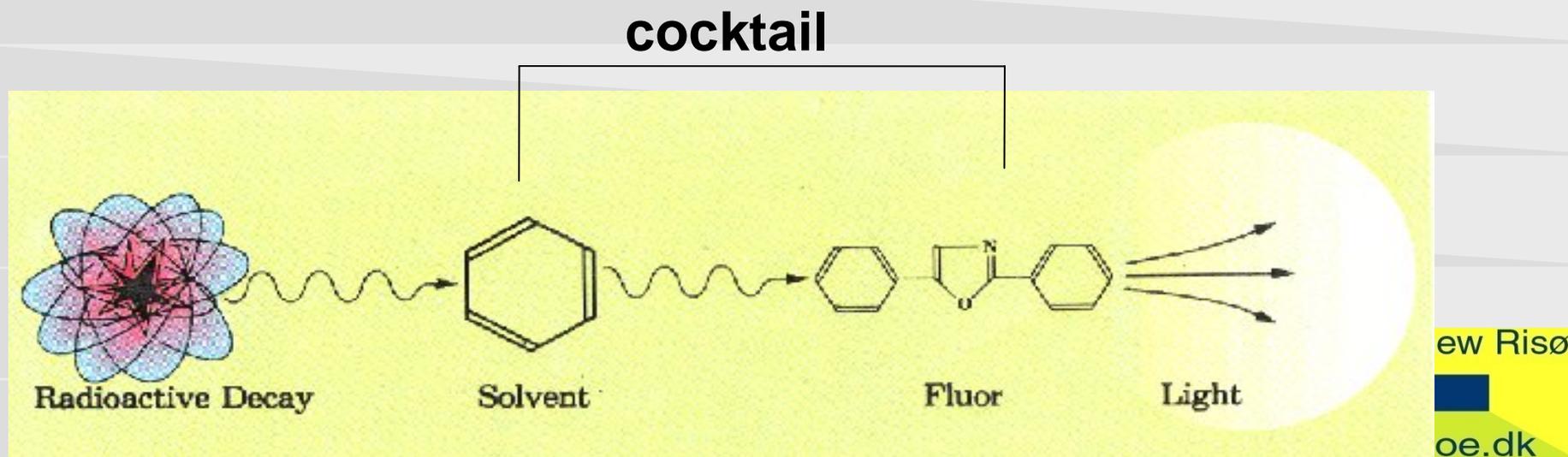
H-3, C-14 and P-32 LS beta spectra in logarithmic energy scale



The max. energy of H-3, C-14 and P-32 is: 18, 156 and 1700 keV.

Principle of LSC

In LSC, the radionuclide is mixed with a cocktail, which consists of **solvent** and **scintillator (fluor)**, the decay energy will be transferred to the cocktail, and converted to photons, by counting the photons using a PMT (photomultiplier tube), the activity of radionuclides is measured.



Aromatic Solvents for LSC

(1) Classic solvents with high toxicity, vapor pressure and low flammability.

(2) Safer solvent, with low toxicity, vapor pressure, odor, high flash point and no permeation through plastics.

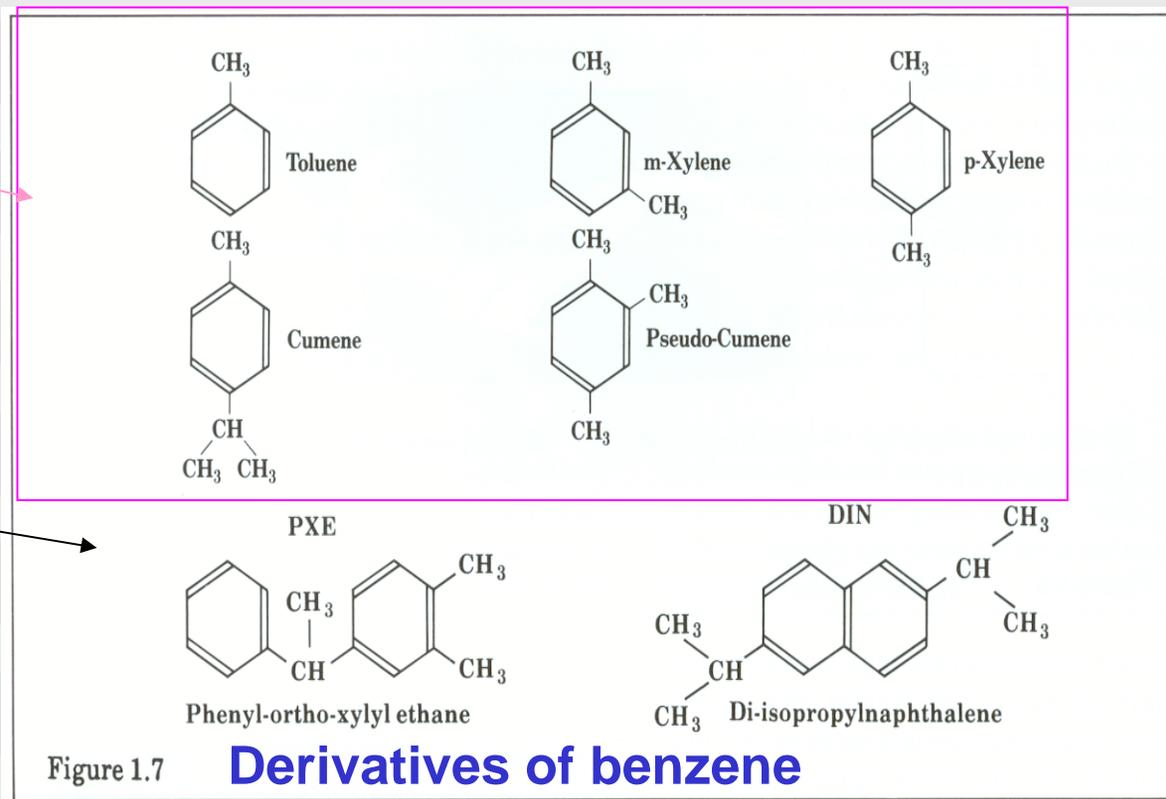


Figure 1.7

Derivatives of benzene

- Aromatic solvents is used as LSC solvent because of the high density of electrons associated with these solvents. When they react with beta particles, a large amount of fluorescence can be produced.

- PM tubes are not sensitive to the fluorescence wavelength of the aromatic solvents. A fluor is therefore used to capture the solvent energy and emit light at a wavelength more easily detected.
- Unfortunately the early PM tubes were not sensitive in the region of fluorescence emission of the primary scintillators. To solve this problem a wavelength shifter (secondary scintillator) was introduced. This absorbs energy from the primary scintillator and emits light at a wavelength more suitable to the PM tubes.

Primary Fluor

PPO — 2,5-diphenyloxazole (most popular)

PPD — 2,5-diphenyl-1,3,4-oxadiazole

PBO — 2-(4-biphenyl)-1,3,4-oxadiazole

PBD, BBD, Butyl-PBD, BBOT, TP

Secondary Fluor

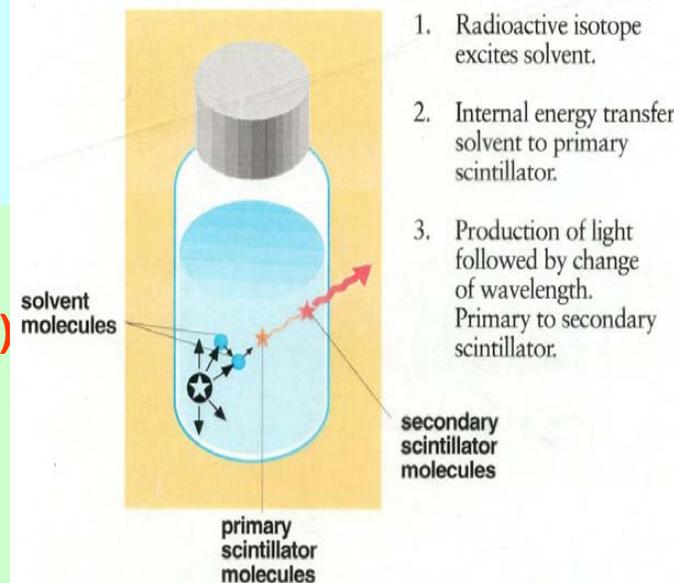
Bis-MSB: p-bis-(o-methylstyryl)benzene (widely used)

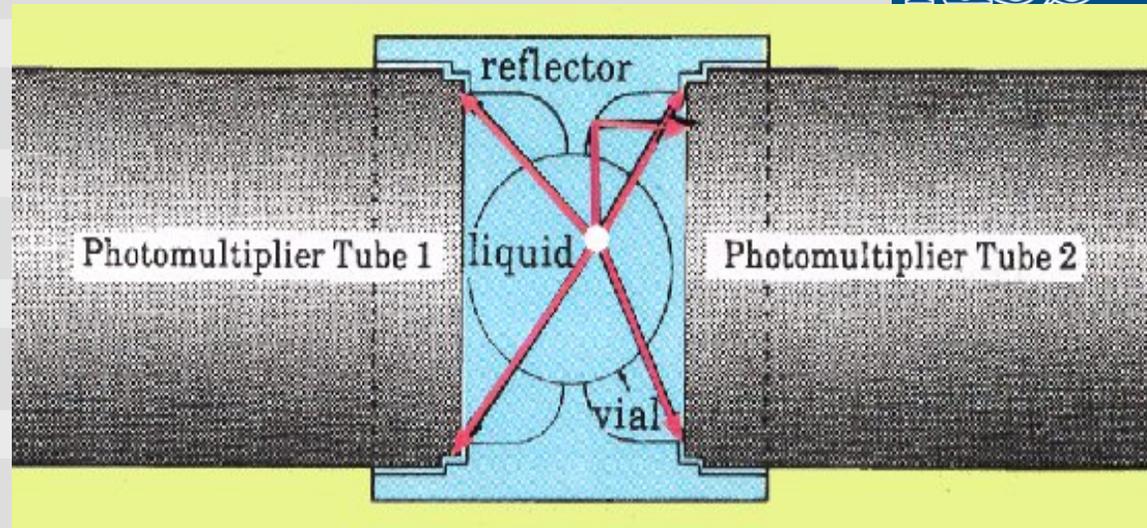
POPOP: 1,4-bis-2-(5-phenyloxazolyl)benzene

NPO: 2-(1-naphthyl)-5-phenyloxazole

Dimethy POPOP, NPD, BBO, PBBO

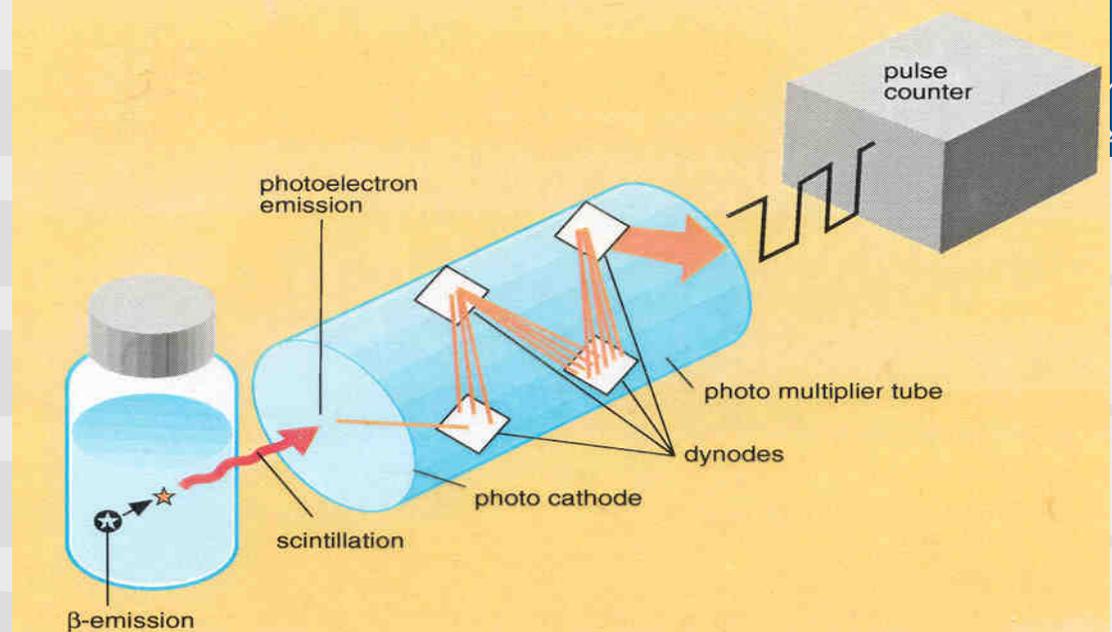
Liquid scintillation process (The energy transfer process)





- Photomultiplier (PM) tubes have been developed which can convert the minute flashes of light into measurable electrical pulses.
- Each individual radioactive decay event produces a pulse of photons. In general from **five to seven photons are generated for every 1 keV of decay energy and these are distributed uniformly in space.**

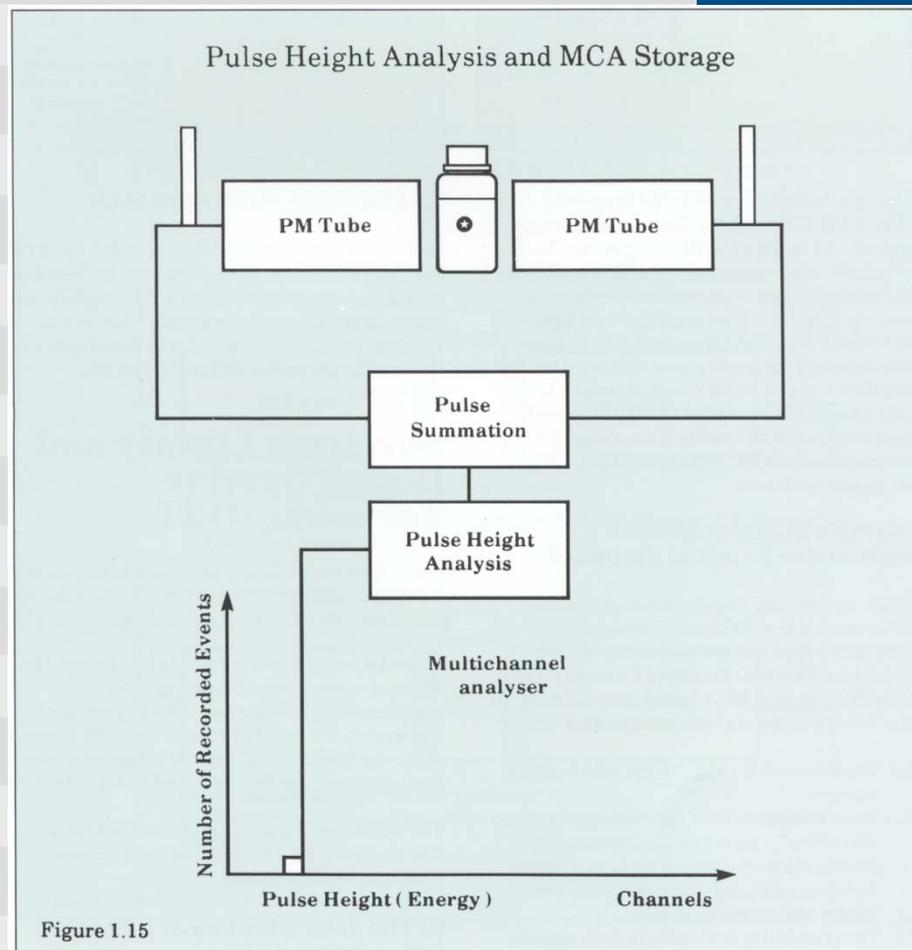
Counting Radioactive Decays



- The photons of light are collected by the optical reflector system of the scintillation counter to the light sensitive photocathode of the PM tubes and multiplied. **The electrical charge pulse obtained is proportional to the original beta particle energy.**
- A single photon striking the photocathode has at most a 30% probability of producing an electrical pulse. It is thus of prime importance that the energy transfer process is as effective as possible in order to maximise the counting efficiency, as there are losses associated with the full light generation process (quench).

MCA - Multi-Channel Analyzer

- A Multi Channel Analyser (MCA) is a memory that stores the electrical pulses from the PM tubes.
- Each individual decay event is stored at a location in the MCA depending upon its pulse height (decay energy). A digital picture of the energy distribution of the isotope is formed as more events are accumulated



Effect of Quench in LSC

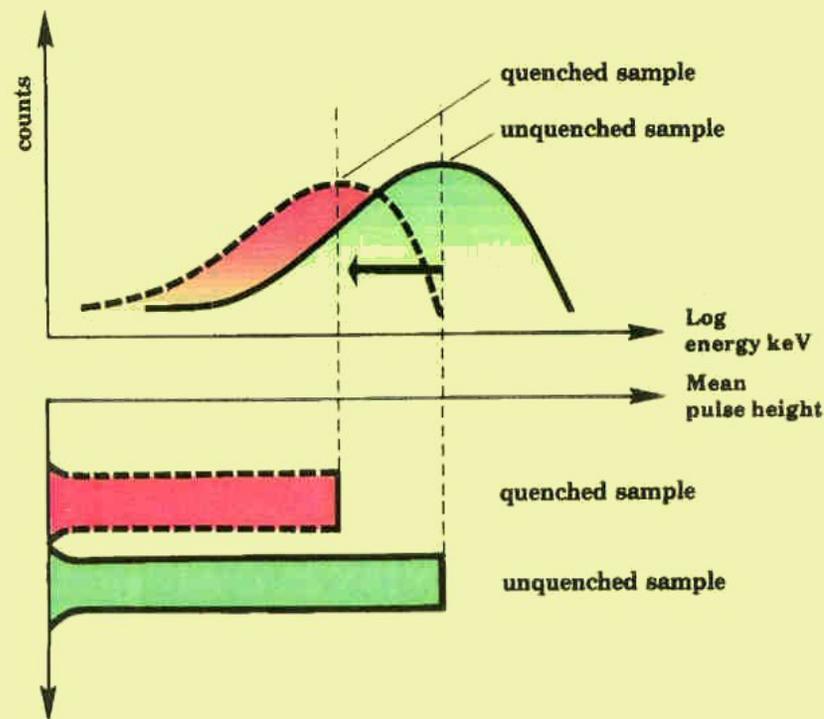
Quenching is a term used to describe energy losses occurring in the energy transfer process. There are three types of quenching:

- (i) Physical (ii) Chemical (iii) Colour

Physical Quenching

- Physical quenching occurs when a barrier impairs contact between the radioactive particle and the scintillator solution, or when the photons of light generated are absorbed by some solid within the vial.
- Radioactivity adsorbed onto active surfaces or filter disks, and two phase aqueous organic solutions are some of the most common sources of physical quenching.

Effect of Quenching on Energy Spectrum (Pulse Height Spectrum)



Color and Chemical Quench

Colour Quenching

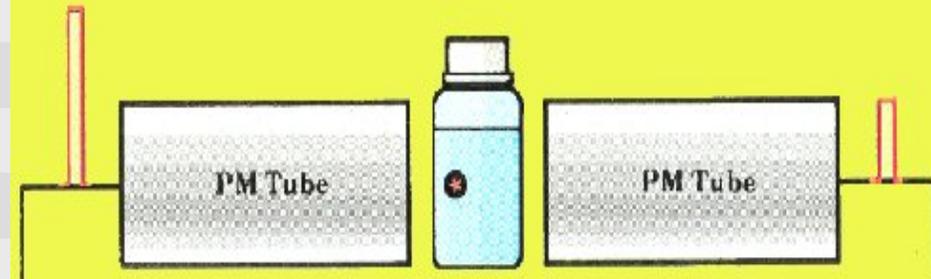
Colour quenching occurs after the fluorescence stage when light absorbing compounds interpose and lessen the number of photons leaving the scintillation vial. Fluorescence emission takes place in the blue region of the spectrum, therefore the order of severity of colour quenching is:

Red > orange > yellow > green > blue

Chemical Quenching

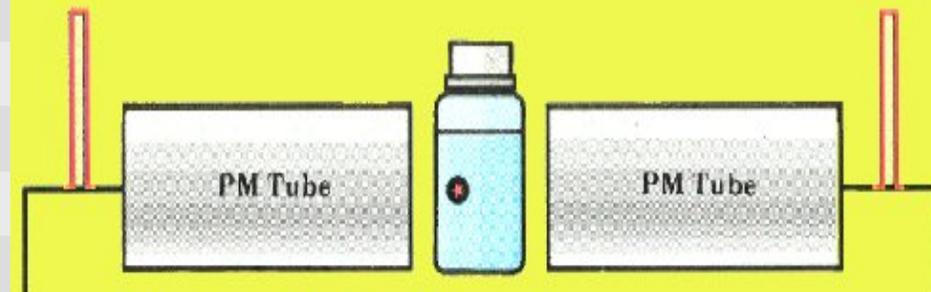
Any compound that does not have an aromatic structure will produce some quenching effect.

The Effects of Colour Quench and Chemical Quench on Pulse Amplitude Disparity



For a colour quenched sample:

An event occurring at one side of the vial produces two pulses of disproportionate height.

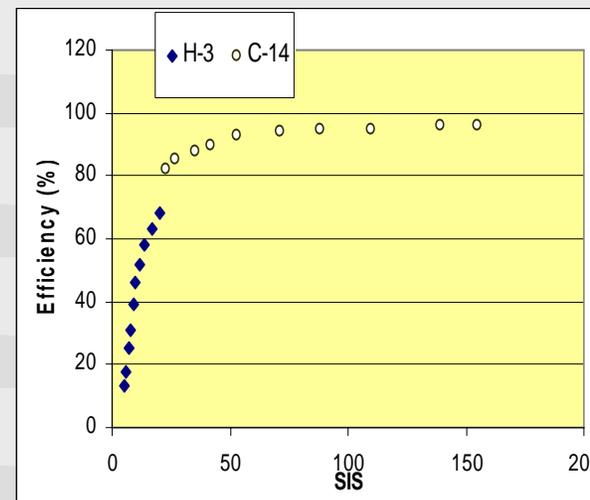
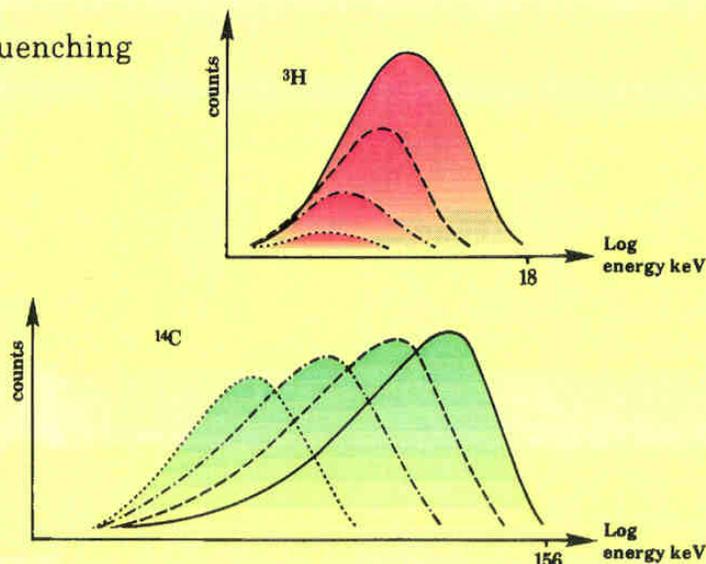


For a chemically quenched sample:

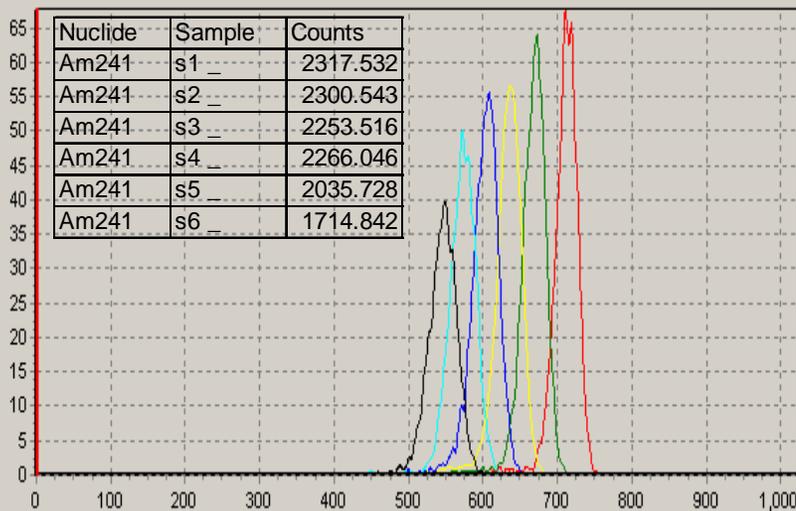
An event occurring at one side of the vial produces two pulses of identical height.

Effect of Quench in LSC

Effect of Quenching



Sample Spectrum



- Quenching causes a reduction in the photon yield from each radioactive decay event, results that the whole energy distribution is shifted down and counts (CPM) is reduced.

- The higher energy β emitter has a less effect on the reduction of counts than that of low energy emitter.

Quench Indices

- By Sample Spectrum Characterization

- SIS (spectral index of the sample)

It is a measure of the average energy of the sample calculated to represent an endpoint energy of the sample spectrum

- SQP(I) (Spectral Quench Parameter of the Isotope Spectrum)

- IC# (average energy of the sample spectrum expressed as a number)

- By External Standard

- SQP(E) (Spectral Quench Parameter of the external standard)

- tSIE (transformed spectral index of the external standard)

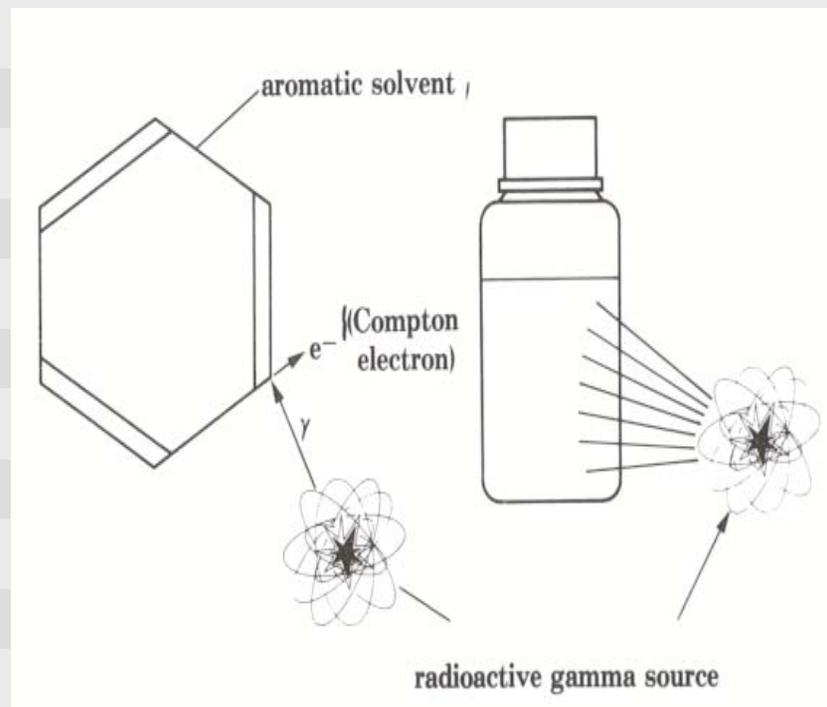
It is calculated on the basis of the tSIE being equal to 1000 for an unquenched ^{14}C sample, which is used for calibration and normalization

- H# (Horrock's number)

It is determined to be the inflection point of the descending edge of the external standard spectrum when plotted on a logarithmic channel number scale.

Quench monitoring using an external standard

- Compton electron spectrum of external standard shows the same quench effects as the sample spectrum.
- External source: ^{152}Eu , ^{226}Ra , ^{133}Ba , etc.
- Advantages over internal std method:
 - fast
 - nondestructive
 - allows simultaneous colour measurement
 - works equally well for low activity samples



- **Internal Standard Method**

$$E = \frac{C_{s+i} - C_s}{D_i}$$

$$D_s = C_s / E$$

E: Counting efficiency of the sample

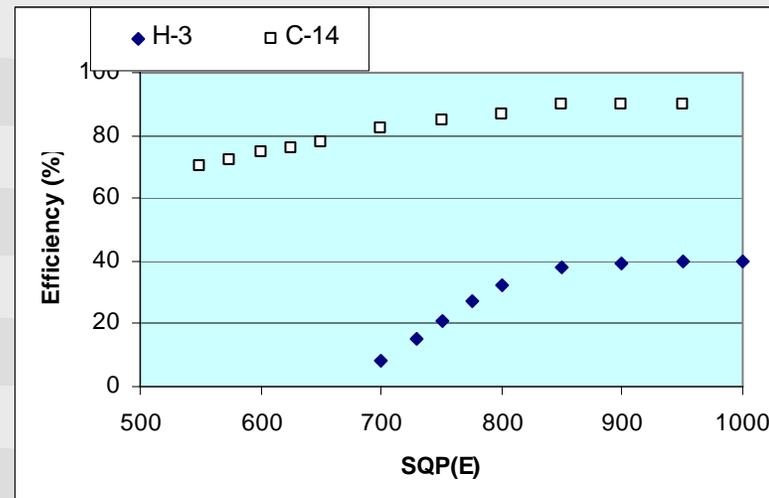
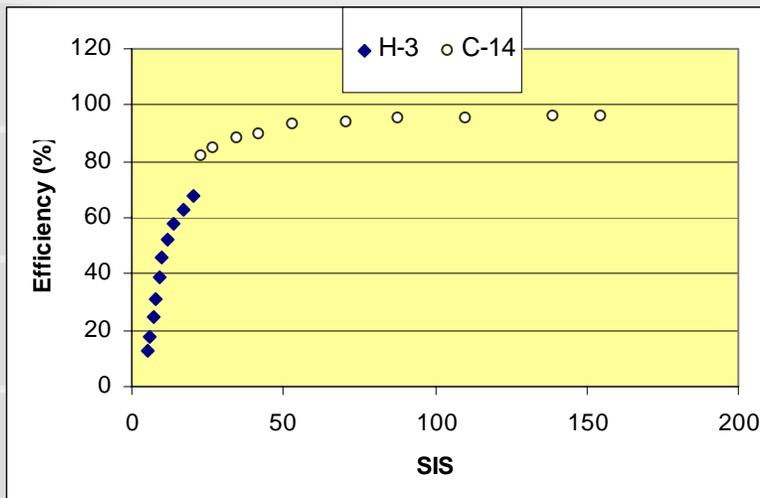
C_{s+i}: The count rate after the addition of internal standard

C_s: the count rate before the addition of internal standard.

D_i: The disintegration rate of the added internal standard

D_s: The disintegration rate of the sample

- **QIP (Quench indicating parameter) method**



Interferences in Liquid Scintillation Counting

- Background
- Static
- Wall effect
- Radionuclide mixtures
- Luminescence
 - Bioluminescence
 - Photoluminescence
 - Chemiluminescence

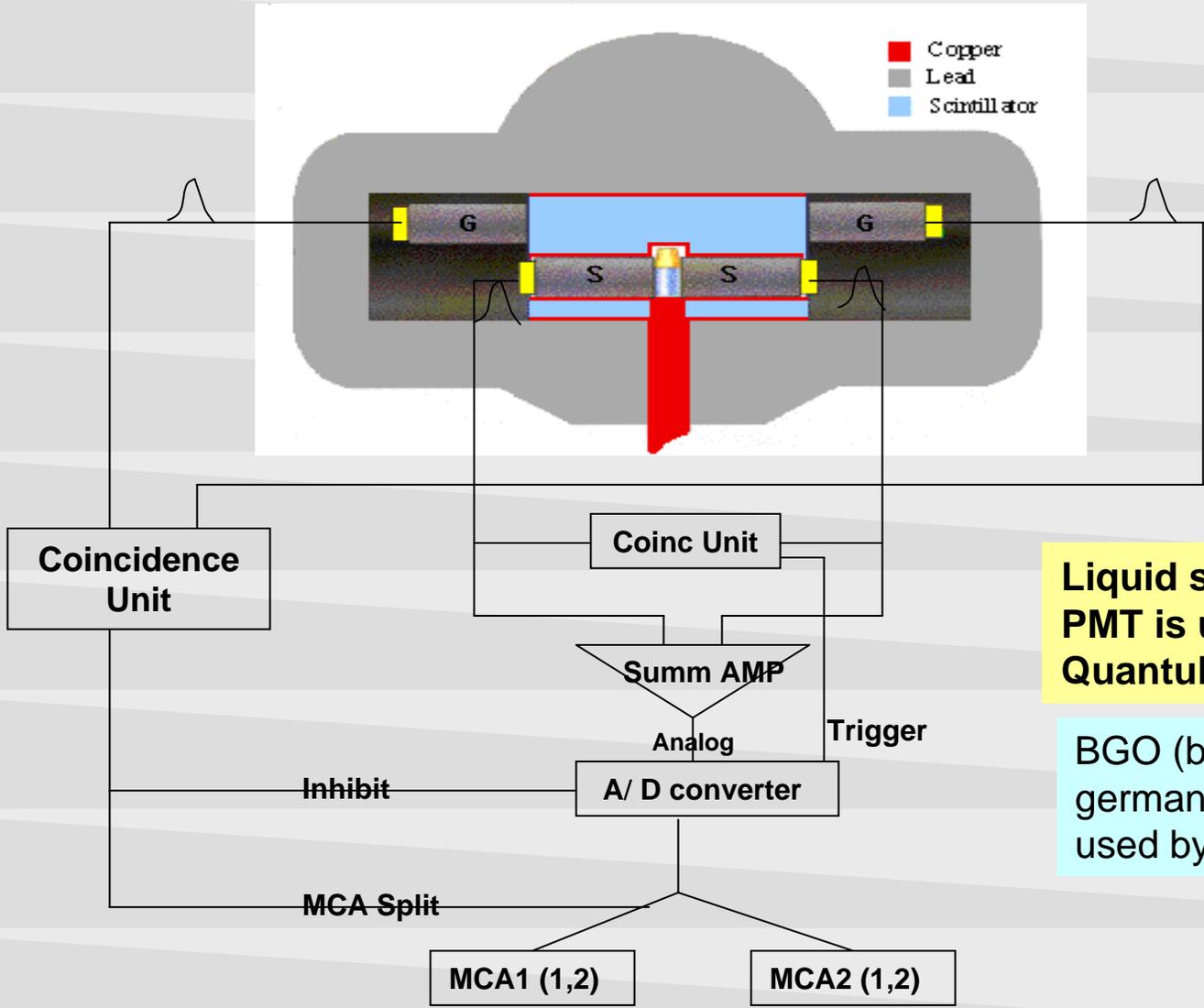
- **Interference from Background**

- Instrument noise and PMT cross talk
- External radiation, cosmic and environmental radiation

- **Method for reduction of the background**

- Coincidence circuitry
- Passive shield using lead and copper
- Active shield using guard detector to remove the effect from external radiation.
- Pulse amplitude comparison
- Low background vial, underground lab, temperature control etc.

Background reduction by passive and active shield



Liquid scintillator with PMT is used as guard in Quantulus

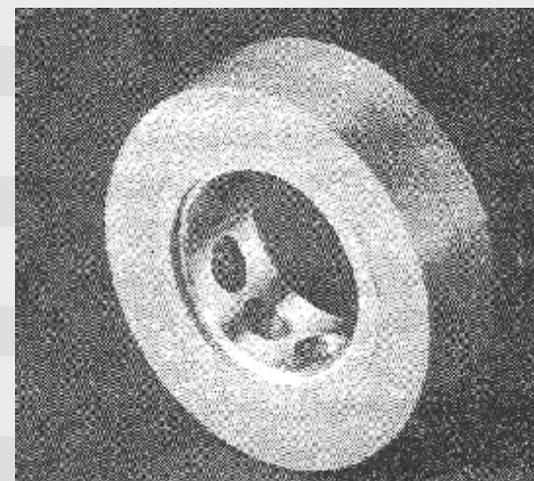
BGO (bismuth germanate) detector is used by Packard LSC

Static

- Static electricity may be generated on the scintillation vial in the sample preparation.
- Electrostatic discharge occurs randomly and produce photo, which interfering LSC.

Methods for reducing or eliminating static

- Use of electrostatic controller
- metal vial holder
- Selection of vial type
- Antistatic wipes
- Humidification of sample preparation and counting area



The electrostatic controller can generate a 360° field of electrically produced ions, when counting vial pass through it enters the field of electrically produced counterions, which neutralize all static electricity on the vial surface.

Wall effect

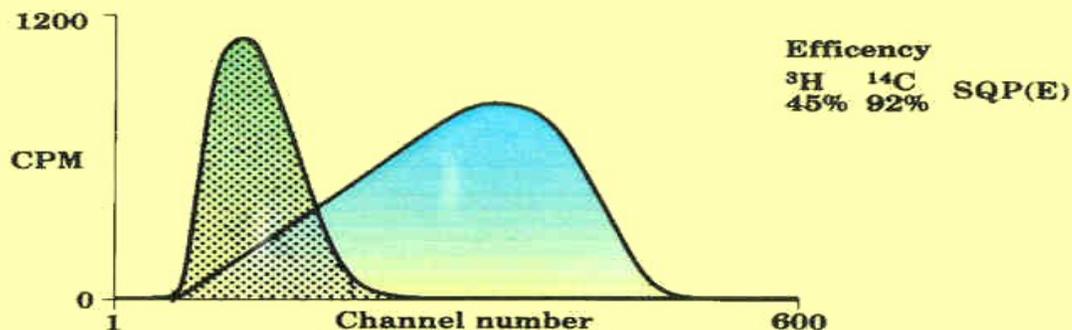
- Some organic scintillator from the cocktail can penetrate the wall of the plastic vial.
- Plastic vial with solvent penetration can scintillate, and causing a distortion of the external standard pulse height spectrum.
- This will result in an inaccurate quench correction.

Method to eliminate wall effect

- Use of new environmentally safer cocktail solvent
- Use of glass vial.



Radionuclides mixture



- Multiple radionuclides in a sample can present a sum spectrum. When the energy spectra of two radionuclides overlap, they interfere with each other.

Methods for quantifying the separate radionuclides

- Separation of individual radionuclides
- Using Dual or Triple Label Counting
 - Exclusion method
 - Inclusion method



Luminescence

- **Bioluminescence**

It is caused by the photons produced from biochemical reaction

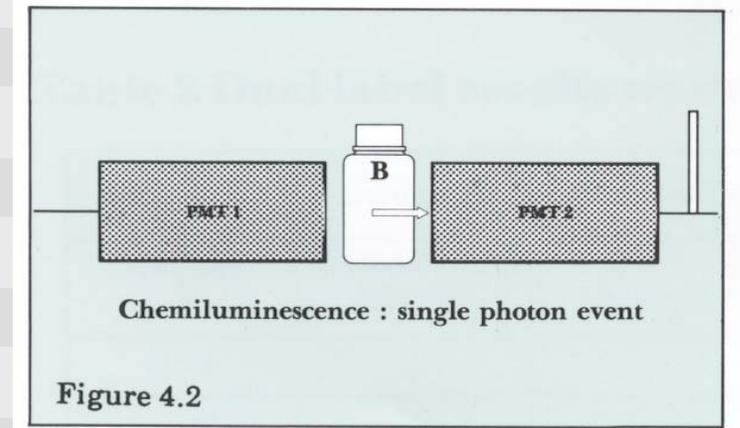
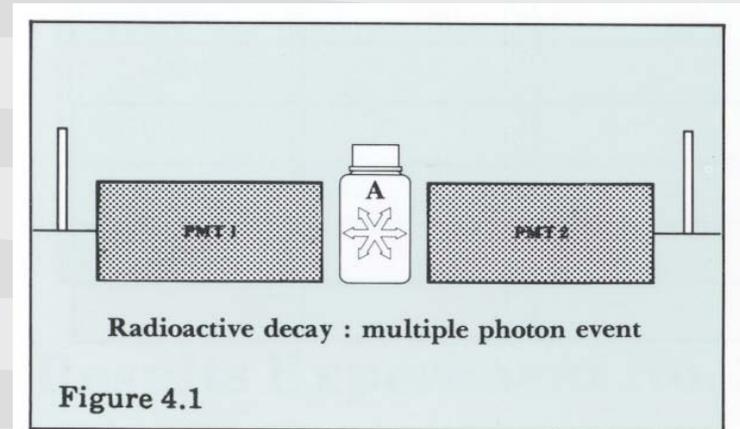
- **Photoluminescence**

It is the result of the exposure of the sample-fluor cocktail mixture to ultraviolet light. Photoluminescence is normally single photon and decay in minutes. So, it can be eliminated by keep in dark for 10-15 minutes.

- **Chemiluminescence**

This is the production of light in the scintillation cocktail due to a chemical reaction. When cocktail is mixed with sample solution, the pH effect and chemical interaction among the components cause molecular excitation and light emission.

- A radioactive decay is characterised by the production of multiple photons of light.
- A chemiluminescence event produces only a single photon of light.
- The luminescence spectrum occurs at the very low energy portion, generally in 0-6 keV region.



Luminescence Control and Elimination

- **Chemical method**
 - Neutralization of sample solution before mixing with cocktail.
 - Using special cocktail designed to reduce luminescence, such as Insta/Fluor, Hionic/Fluor etc.
- **Temperature Control**
 - Heating the sample to 40°C before measurement help drive the chemiluminescence reaction to its endpoint.
 - Cooling during measurement reduces counts from chemiluminescence.
- **Counting Region Setting**
 - Setting the counting region to over 10 keV will eliminate the luminescence.
- **Delayed Coincidence Counting**



α , γ Interaction in Liquid Scintillation Counting

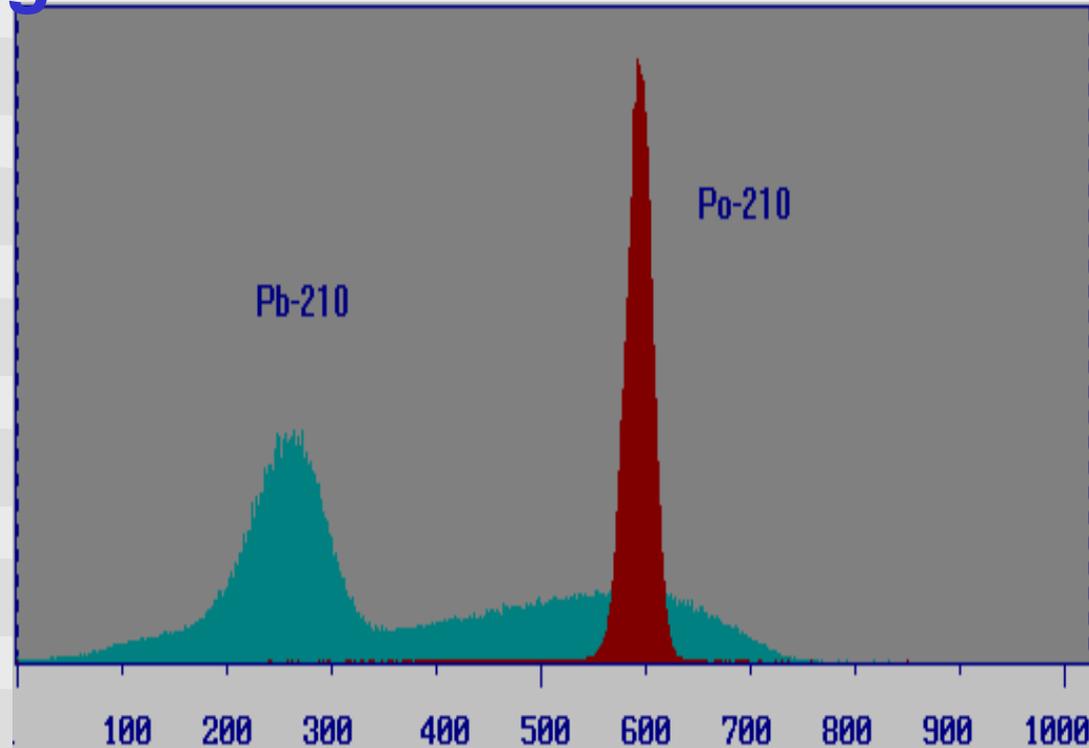
α Particles

✓ The higher charge and mass of α particle are responsible for less efficient excitation energy transfer to solvent and fluor.

✓ α particles produce light in the liquid scintillation cocktail at about 1/10 of light intensity per unit of particle energy of β particles.

✓ Its pulse height is equivalent to app. 1/10 of its original decay energy

✓ It is monoenergetic, so a single pulse height peak is seen for each α decay.



γ ray emission

γ ray is nonparticulate electromagnetic radiation, it makes only a minor contribution to excitation in liquid scintillation fluor by Compton electrons

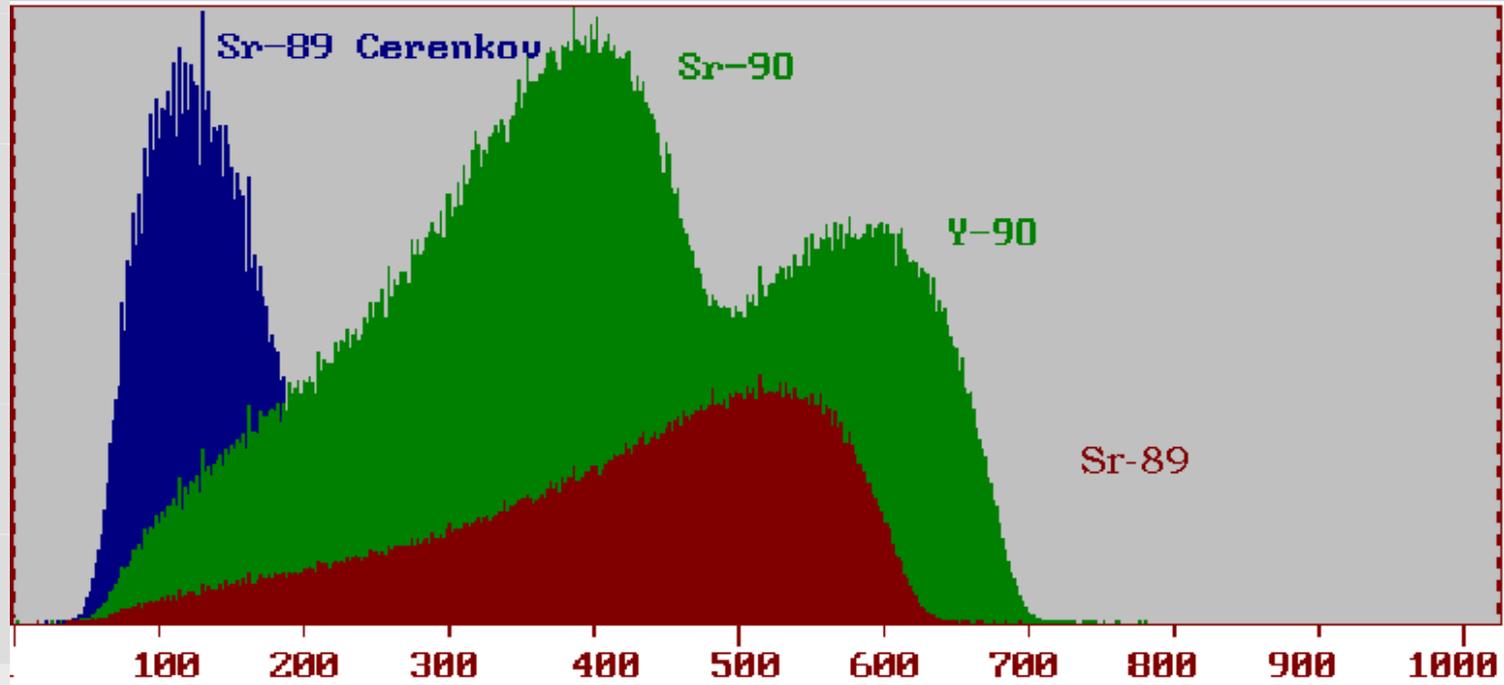
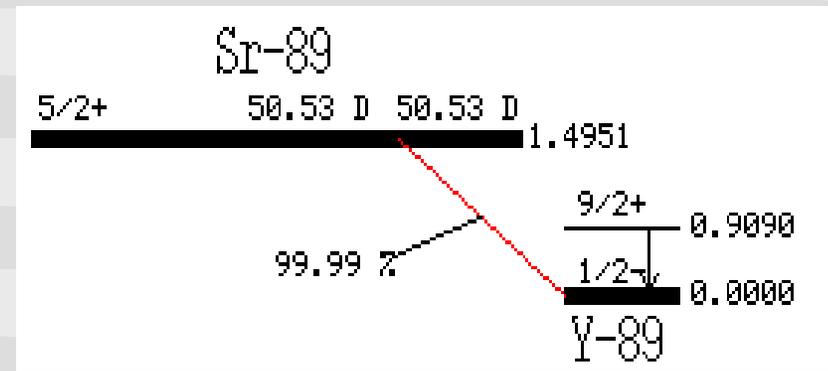
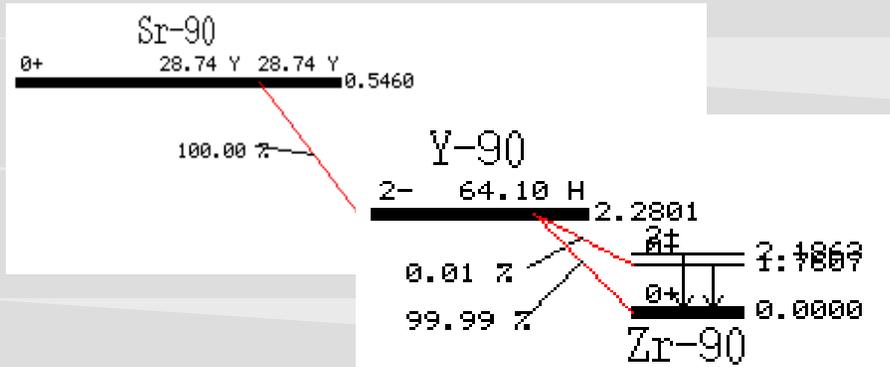
Measure many radiation types by LSC

- Beta particles
- Alpha particles
- Cerenkov radiation
- Electron Capture
- X-rays and Auger electrons
- Conversion electrons
- Gamma rays via Compton electrons
- Luminescence and fluorescence

Cherenkov Photon Counting

- Cherenkov effect
Charged particles, e.g. β , that possess sufficient energy can travel at a velocity exceeding the speed of light in the media such as water, organic solution. When this occurs, the charged particle produces Cherenkov photons, which extend from the ultraviolet into the visible wavelengths.
- Beta particles with energy excess of 263 keV in water can produce Cherenkov effect and can be detected by Cherenkov photons without cocktail.
- The Cherenkov photon is normally detected in the low energy counting region (0-50 keV).
- The Cherenkov counting efficiency is in the range of 35-50%

Determination of ^{89}Sr and ^{90}Sr by using β decay of ^{89}Sr and ^{90}Sr and Cherenkov radiation of ^{89}Sr and ^{90}Y



Auger Electrons Counting

- **Auger Electrons**

In the decay of internal conversion electron and electron capture, vacancies are left in electron shell, that can be filled by atomic electrons from higher energy level. In the process of falling to a lower energy shell to fill a vacancy, electron energy is lost as a photon of x radiation. This x radiation may collide with an atomic electron, resulting in the emission of the electron referred as an Auger electron.

- The energy of Auger electron is low, normally less than 10 keV.
- Electron Capture decay can be measured by counting Auger electrons by LSC

Electron Capture Counting

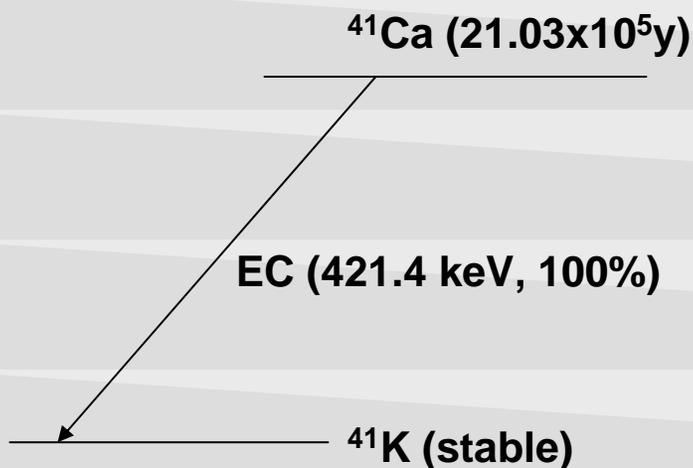
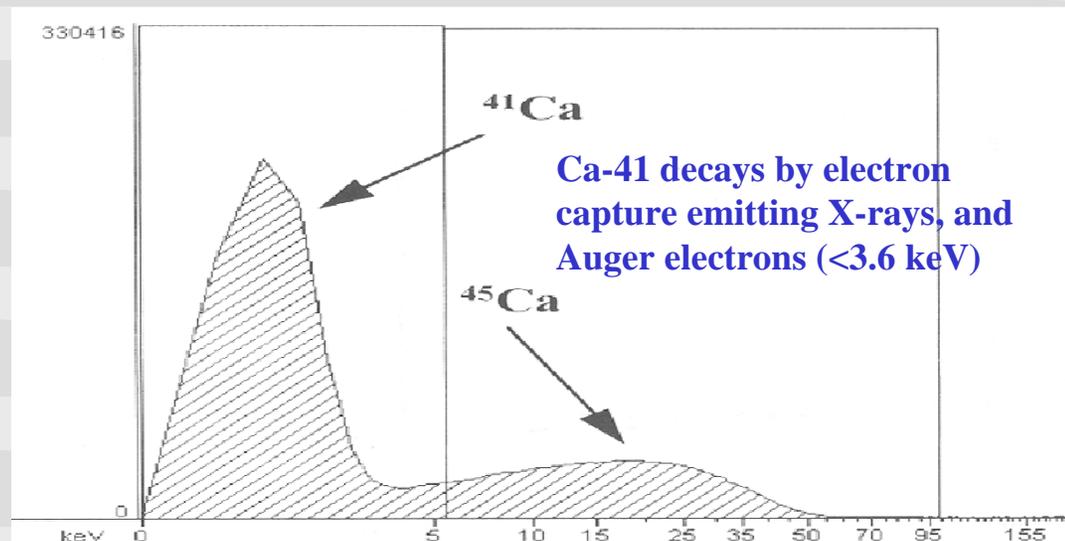
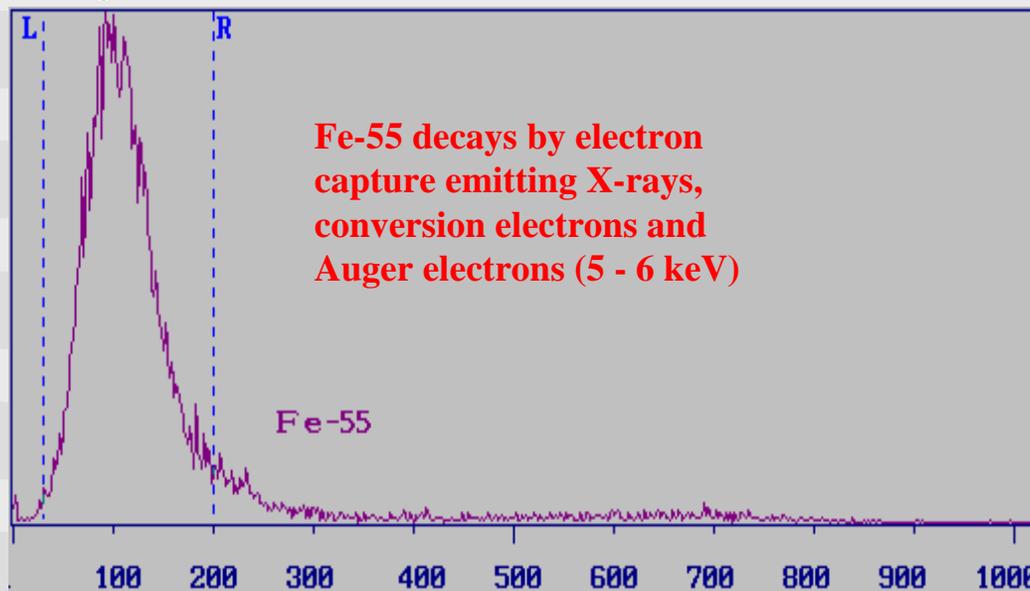
- **Electron Capture decay**

It is a decay process that an unstable nucleus capture a atomic electron, the absorbed electron combines with a proton to yield a neutron within the nucleus. The capture of atomic electron by the nucleus leaves a vacancy in the electron shell, and this is usually filled by an electron from an outer shell, resulting in the x radiation.

- The electron capture decay process may compete with β^+ emission.

- Electron capture decay normally companies with internal conversion electron and Auger electron emission, so it can be detected by using LSC.

Determination of Radionuclides with Electron Capture Decay



Discrimination of α/β spectra and simultaneous determination

- Since α particle produces about 1/10 as much light intensity as β particles per unit of radiation energy, α and β pulse height spectra often overlap.
- α particles emit 5 MeV or more energy which is able to create triplet state in solvent which de-excite to ground state slower than single state produced by β particles. The pulse decay time of α particles is 30-40 ns longer than β particles interaction with cocktail.
- Measurement of the pulse decay time and length allows identification of particle which cause it and enable simultaneous recording of pure α and β spectra, respectively, So called pulse shape analysis (PSA).

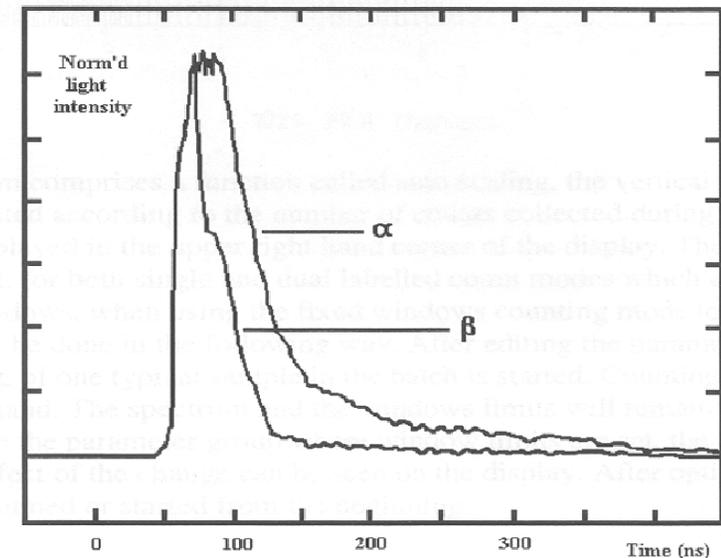
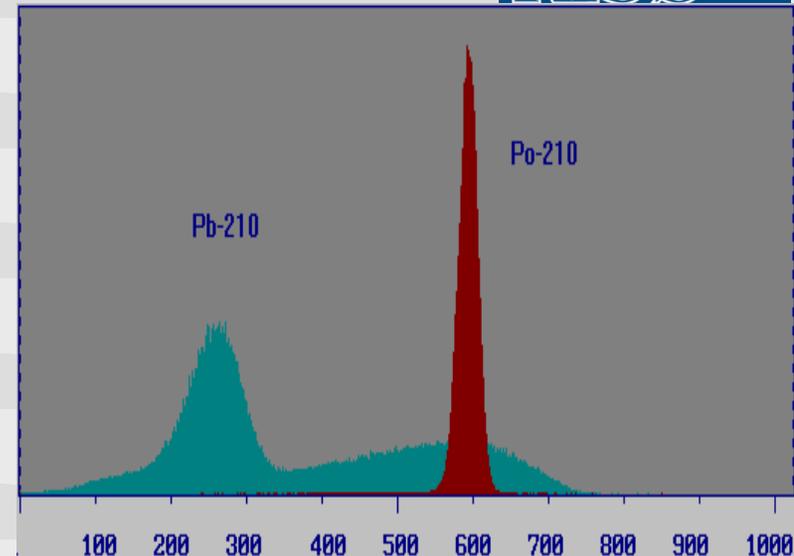
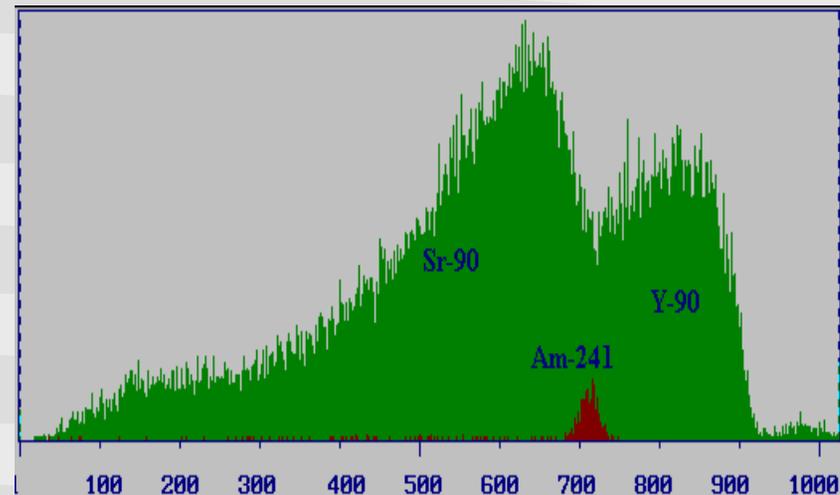
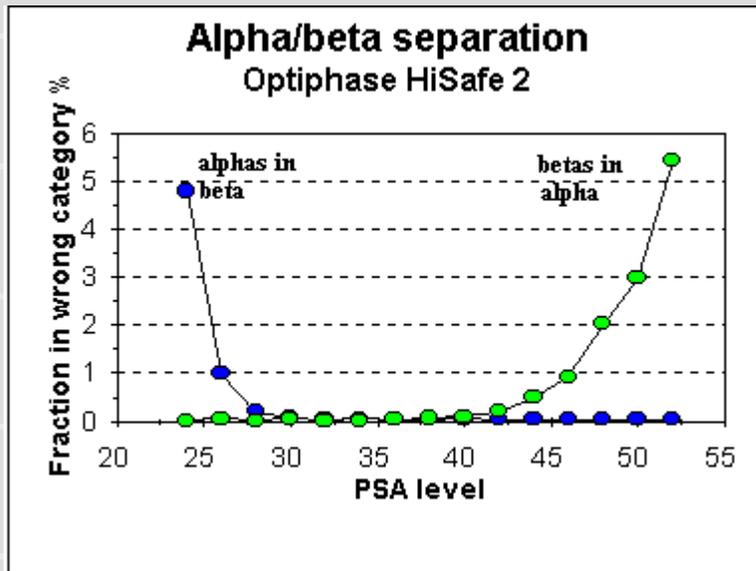


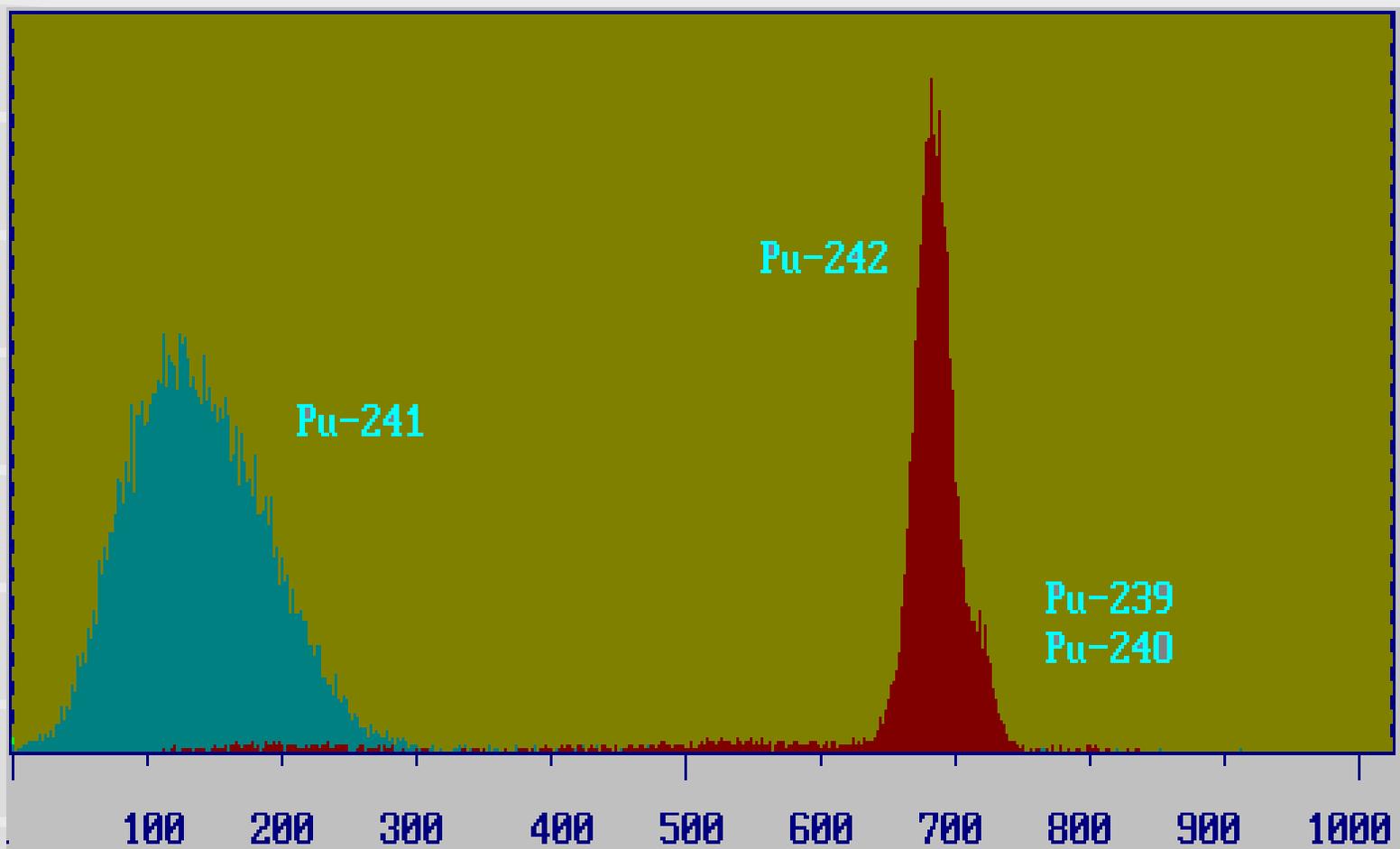
Fig. Pulse shape for event caused by α and β particles

Wallac Pulse Shape Analyzer

- It is a sensitive α/β separation device in commercial liquid scintillation counters
- Less than 0.02 Bq (0.5 pCi) easily detected in the presence of 2 kBq (50 nCi) of beta activity
- Minimal spill-over even at high quench levels

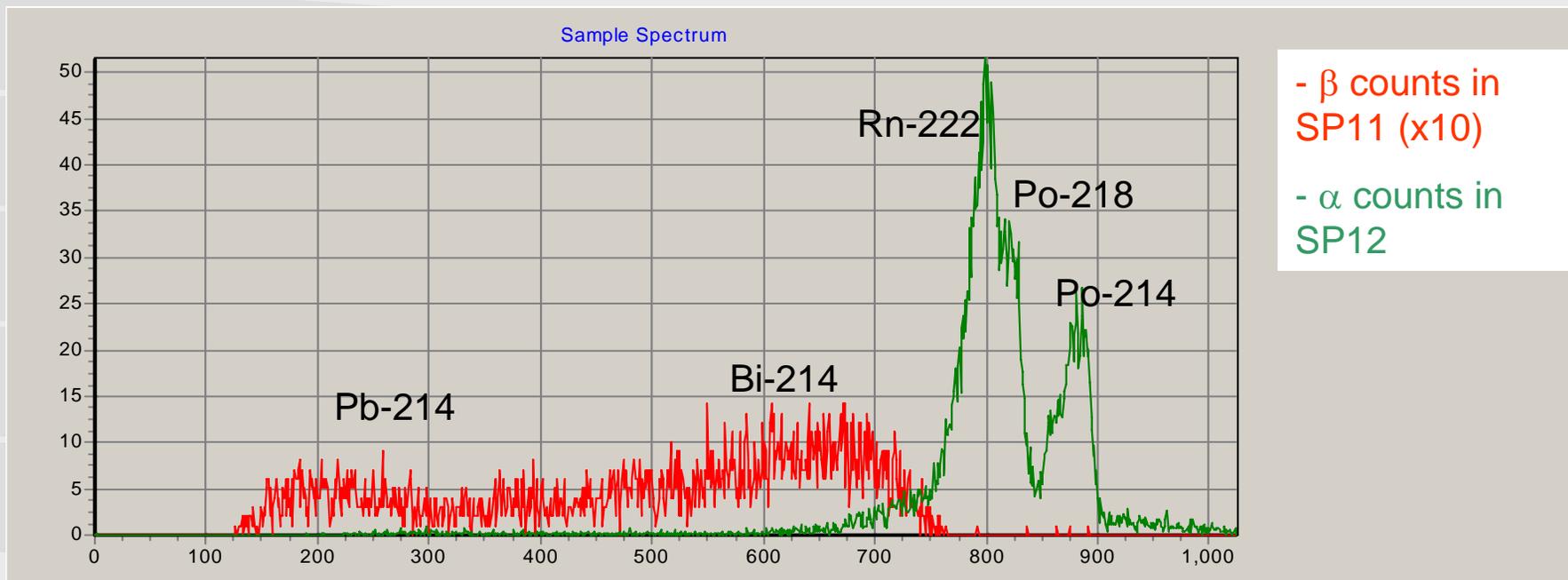


Low Level Pu by LSC

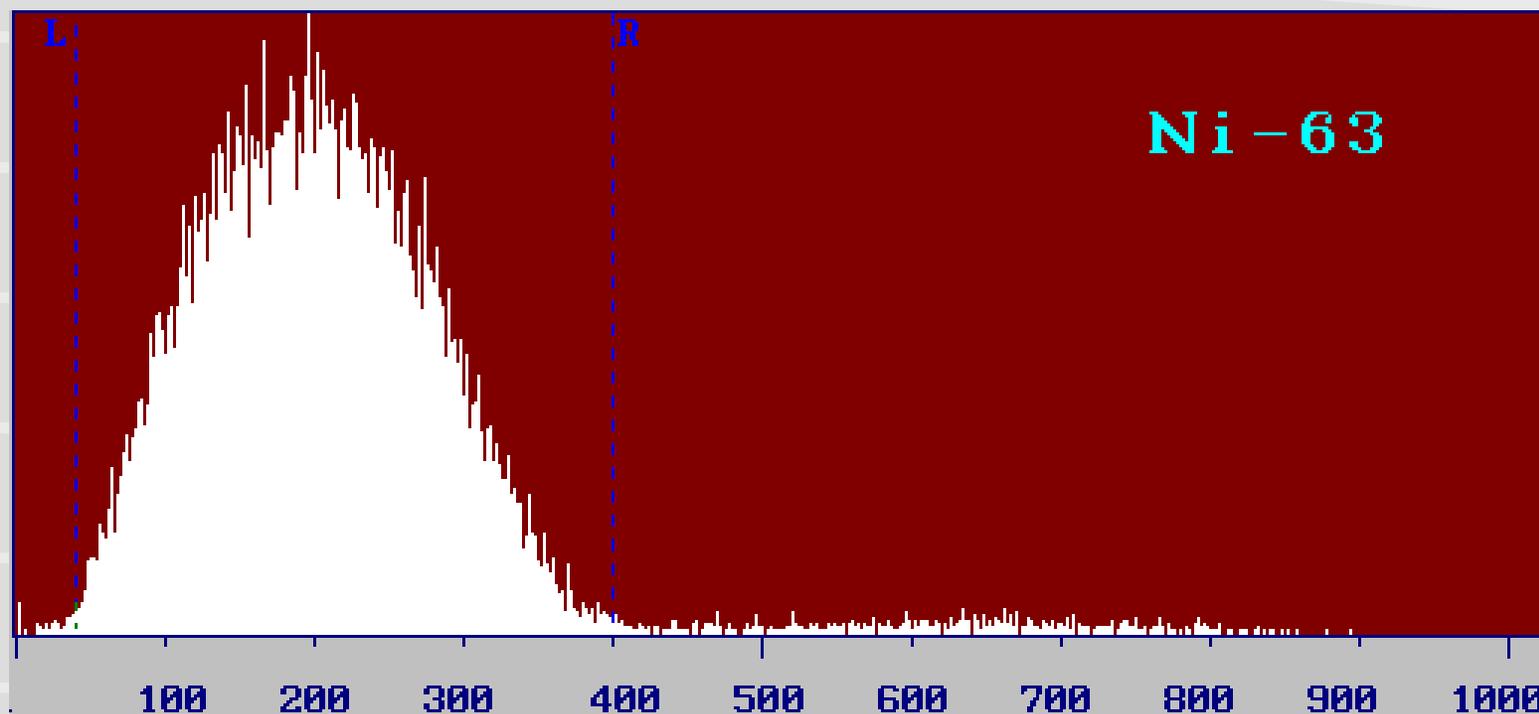


^{222}Rn in water

- 10 ml aqueous-immiscible extraction cocktail (such as ultima Gold F) is added to a 20 ml low diffusion PE vial.
- At sampling site, 10 ml water is added to the vial with cocktail, the vial is immediately capped tightly.
- By shaking, radon will be extracted to the cocktail, because the solubility of radon is much higher in the organic cocktail than in the water.



Low Level ^{63}Ni by LSC



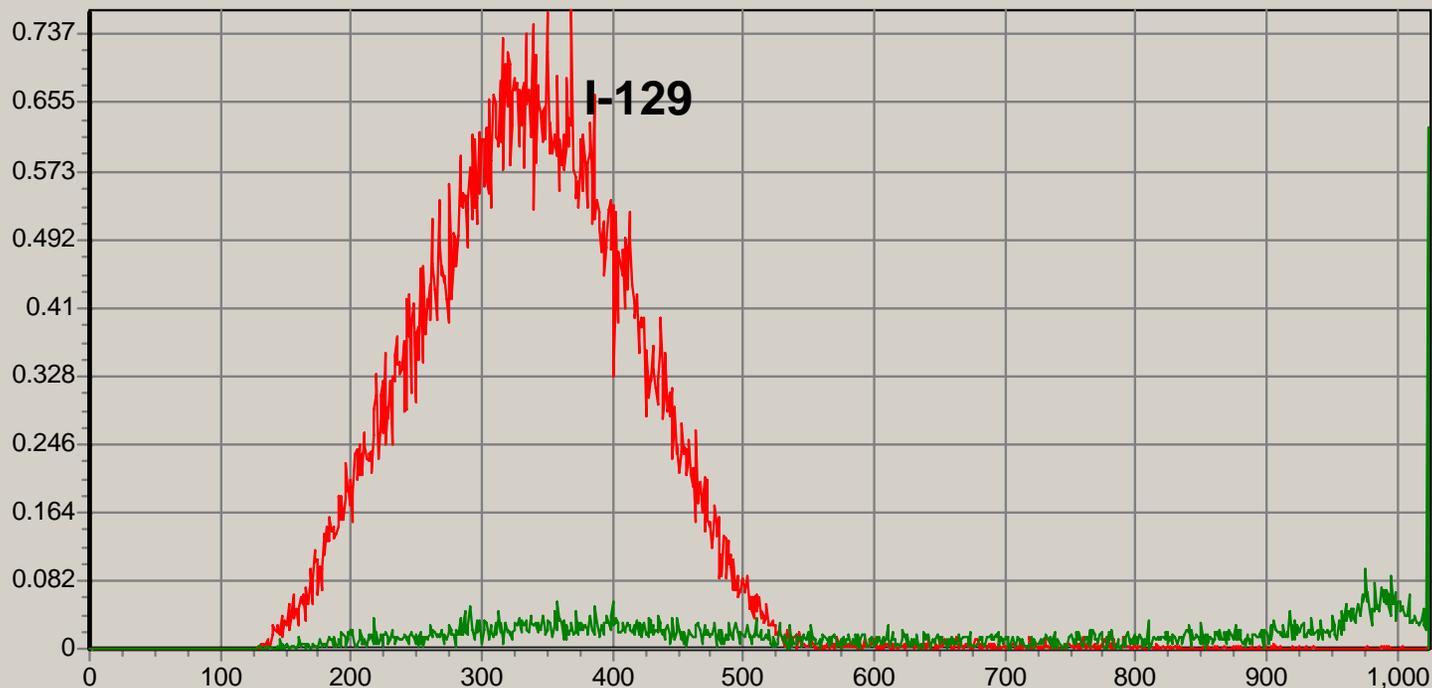
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^{129}I in seaweed

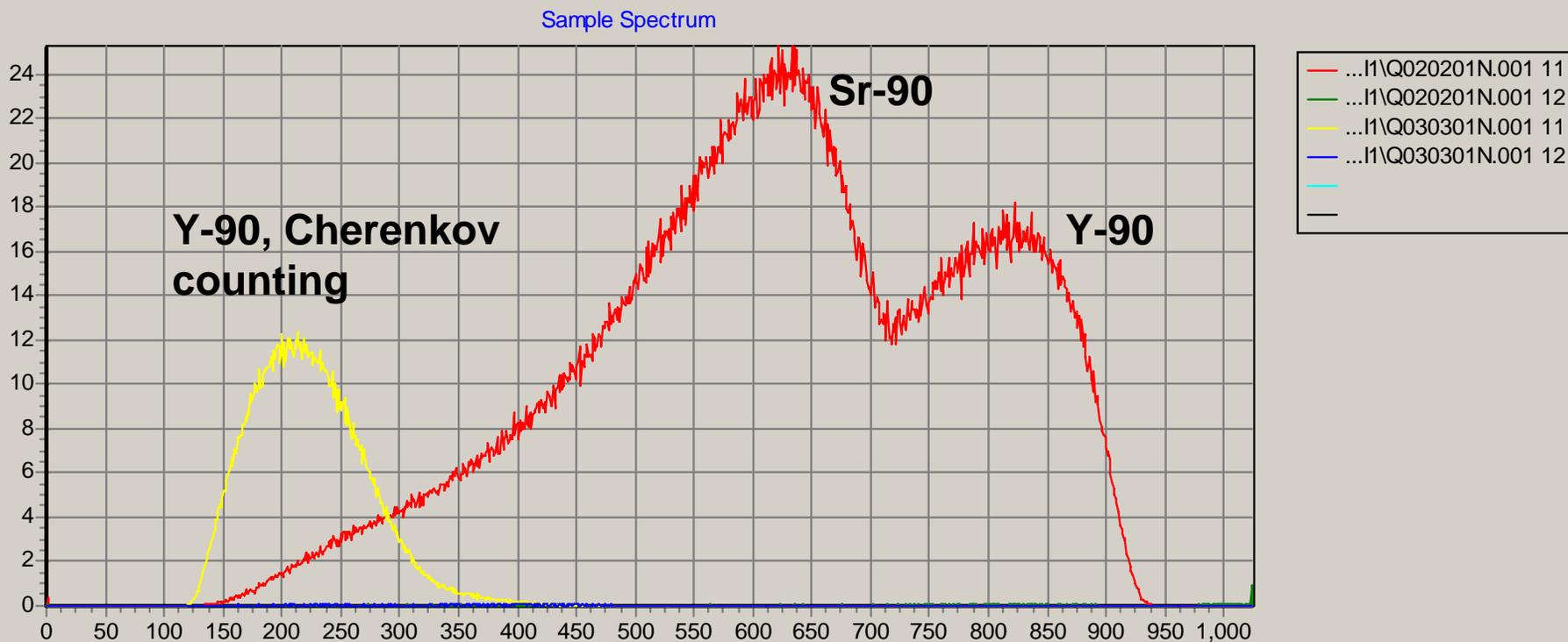
Sample Spectrum



- ^{129}I β counts in SP11
- background counts in SP12



Sr-90/Y-90 standard



Thanks !



The new Risø



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