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Tritium (H-3) Monitoring

Basic Principals, Sampling Techniques, and Examples

Presented at the 3rd RMCC Workshop Held at The Food and Environment Monitoring Centre

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USA

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Presentation Overview

Part 1: Basic Principals -- Tritium

- Chemical and Physical Characteristics
- Radiological Characteristics and Sources of Tritium
- Environmental Tritium Sampling
- Samplers
- Sample Preparation: Lots of chemistry here

Presentation Overview

Part 2: Sample Analysis Using Liquid Scintillation Counting (LSC)

- Tritium Detection and Measurement
- Principals of LSC
- Components of a Basic LSC System
- Basic LSC Energy Spectrum Analysis
- Counting Interference and Spectrum Analysis
- LSC Calibration
- Analysis Software (routines)

Tritium (H-3) Monitoring

Part 1: Basic Principals -- Tritium

Basic Principals -- Tritium

Chemical and Physical Characteristics

Part 1: Basic Principals – Tritium

- Chemical Characteristics:
  - T is an isotope of Hydrogen, ($^3H_2$)
  - HTO Oxide form: Upon release of $^3H_2$ to Environment

- Physical Characteristics:
  - Airborne Form: Gas; Vapor (HTO), Tritiated Dust or Aerosol
  - Contaminated Liquids (oils, water)
  - On Contaminated Equipment and Material

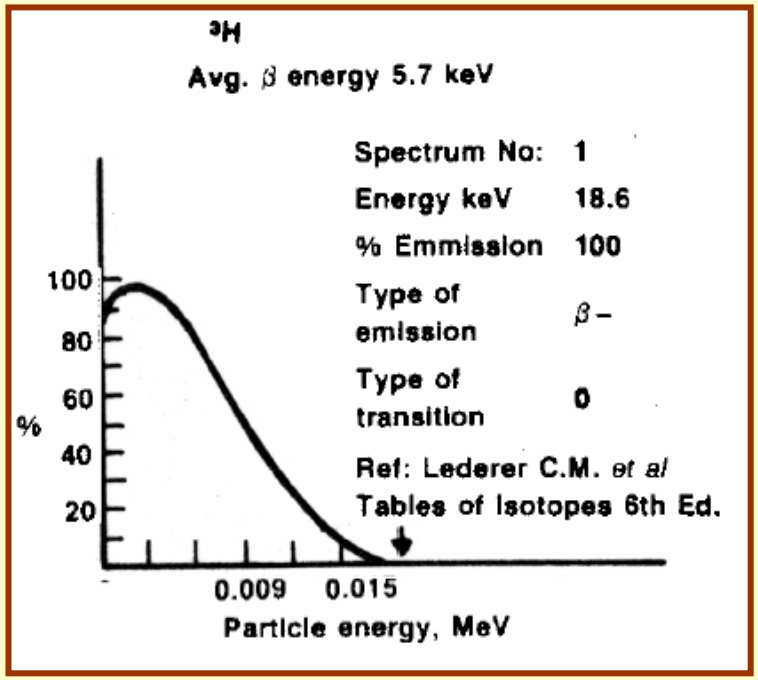
Basic Principals -- Tritium

Radiological Characteristics and Sources of Tritium

Part 1: Basic Principals -- Tritium

Radiological Characteristics

- Pure Beta Emitter, Half-life: 12.33 Y
- Maximum Beta Energy: 18.6 KeV
- Average Beta Energy: 5.7 KeV
- Specific Activity: 3.6×10^2 TBq/g, gas
  55 TBq/g, HTO
- Naturally-Occurring: ~ 555 Bq/L H_2O
  (Internal Dose: 0.015 mSv/yr)



- Annual Limit of Intake (ALI)
  - Gas: Not Applicable
  - HTO: 1×10^9 Bq
- Derived Air Concentration (DAC)
  - Gas: 8×10^9 Bq/m3 (~ 26,000X DAC HTO)
  - HTO: 3×10^5 Bq/m3

Basic Principals -- Tritium

Radiological Characteristics and Sources of Tritium

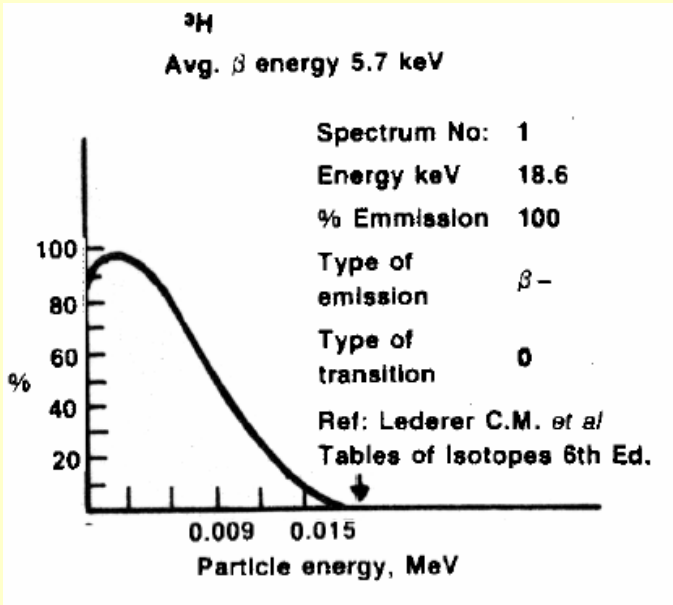
Part 1: Basic Principals -- Tritium

Sources of Tritium

- **Natural Sources:**
  - **$^{14}N(n,t)^{12}C$ and $^{16}O(n,t)^{14}N$ in Atmosphere**
  - **Almost exclusively HTO**
  - **Ave Concentration in Env water: 100 – 600 Bq/m^3**
- **Man-Made Sources:**
  - **Nuclear Industry: ~ 4x10^4 TBq/y**
    - **HWR: 3x10^2 – 2x10^3 TBq**
    - **LWR: 30 TBq/y per Reactor**

Uses of Tritium

- **Luminising Industry: 10 – 100 TBq/y**
- **Research and Teaching: 1 – 100 GBq/y**



Basic Principals -- Tritium

Radiological Characteristics and Sources of Tritium

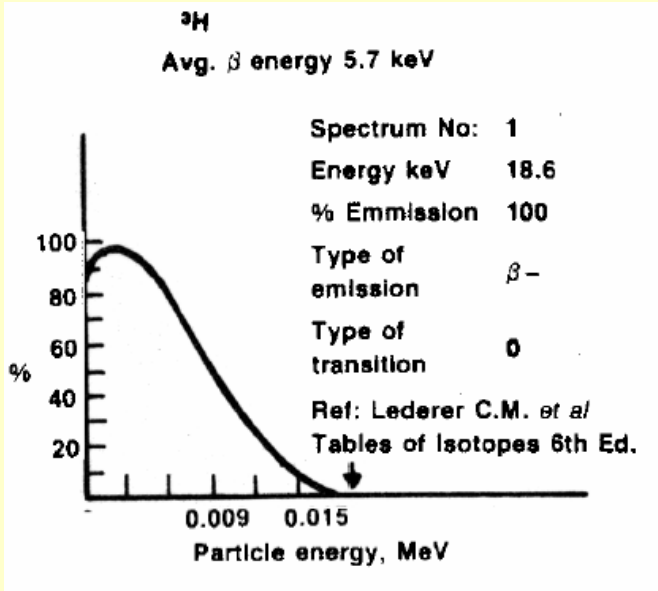
Part 1: Basic Principals -- Tritium

Human Health Impact

- **External Dose: Not Significant**

- **Internal Dose:**
  - **Through Inhalation or Ingestion**
  - **HTO Vapor Penetration Through Skin**
  - **Uniformly Distributed Upon Entry Into Body**
  - **HTO Dose ~ 26,000 Times Larger than T Gas**
  - **Biological $T_{1/2}$: 3-14 days (ave ~ 10 Days)**

- **Behavior in Environment:**
  - **HTO Contamination Spreads Faster Than Non-Gaseous Radionuclides**
  - **Behaves Very Much Like Water Vapor**



Basic Principals -- Tritium

Environmental Tritium Sampling

Active Sampling and Detection:

Commonly Utilized with Flow Through Ion Chambers
Or Proportional Counters:

- Filtered air is passed through a gas type detector
- Detector volume and flow rate determines sample size

Compensated Double Ion Chambers

Ion Chamber

T Monitoring Station

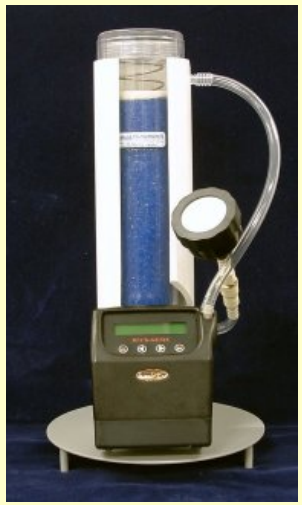
Basic Principals -- Tritium

Environmental Tritium Sampling

Passive Sampling:

Filtered air is passed through a medium that Traps the Tritium. The medium is then collected and transferred to a sample preparation and counting laboratory

Several Examples Will Follow:



Passive T Sampler
Example 1

The self-contained instrument consists of a pump and flow regulator to draw a constant sample (air) stream into a set of vials which collect the radioactive material.

Dual sets of vials are used to ensure that whatever may be missed by one vial is virtually certain to be trapped by a second one.

One set (of two) vials is used to collect tritium oxide, the air stream exiting from this set is passed through a small low temperature catalytic oxidizer and the resultant oxides are then trapped in the second set of vials.

A timer is mounted on the front panel of the instrument, as well as visual indicators to signal failure of sample flow.

A rotemeter and a low flow switch monitor the sampling flow rate.

Passive T Sampler
Example 2

A known volume of silica gel (400 gram) is placed inline with a low flow sampling pump.

Sample duration may be for a period of up to 2 weeks with on line power operation.

Battery power operation is approximately 8 hours with a fully charged battery.

Passive T Sampler
Example 3

Used at sites requiring monitoring of ambient air conditions for tritium gas or aerosols.

The unit uses a calibrated air flow pump to direct air through an oven with a non-asbestos catalyst to oxidize the tritium. The oxidized tritium and air bubbles into the water in the four collector bottles sequentially to trap the tritium in the collector liquid.

At sampling conclusion, the "tritified" water in the collection bottles is analyzed using liquid scintillation counting methodology.

Tritium concentration in the ambient air is calculated using the test duration, air flow rate, and tritium mass captured in the bottles.

Passive T Sampler

Example 4: Air monitoring Around a CANDU

Atmospheric water vapour is sampled monthly for tritium determination.

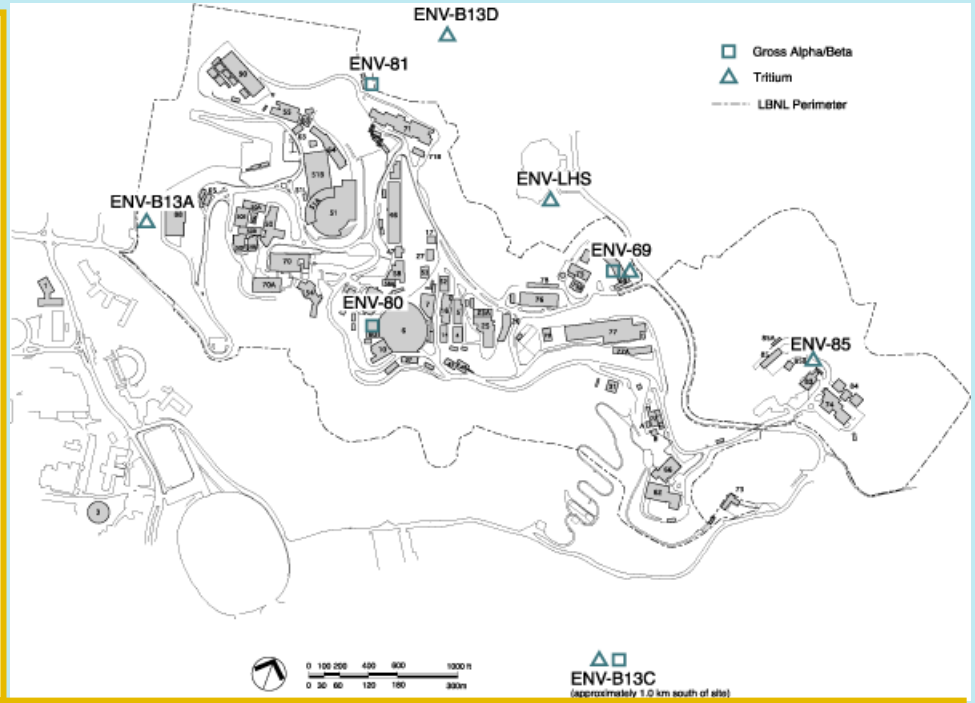
The sampling equipment consists of a tritium cell, containing molecular sieve material that absorbs water vapour as air is drawn through the cell, a flowmeter and a pump. The water vapor is recovered in the laboratory, and the tritium measured by liquid scintillation counting.

The flow rate of air through the tritium cell is reduced during the summer, from 0.2 to 0.07 m3/day, to prevent the sieve from becoming saturated through high humidity.

Ambient Air Monitoring Network: A Collection of Sampling Locations

The sites are chosen based on emission source locations, local wind patterns, and proximity to off-site residential areas and facilities.

Equipment at each site continuously samples outdoor air.



The sampling media are replaced and analyzed periodically.

Sampling Stations

Tritium Sampling is Commonly a Part of Sampling Stations Where Sampling for Several Radionuclides is Performed.

Sampling Station
Example: The Station

A network of environmental air stations includes several sampling stations to sample radionuclides in ambient air.

A typical station is shown at the right with its housing open for sample change out.

Sampling Station
Example: The Sampling Equipment

A pump pulls ambient air into the housing that protects the sampling apparatus and through the filter and cartridge.

Instrumentation within the housing records the total time the pump ran during the two-week sample period and the flow in the particle and the tritium sampling trains.

Sampling Station

Example: The Particulate and T Samplers

Each sampler is equipped with a filter to collect a particulate matter sample (for gross alpha/beta and radiochemical determinations);

and a silica gel cartridge to collect a water sample (for tritium determination).

Tritium (H-3) Monitoring

Part 2: Sample Analysis Using Liquid Scintillation Counting (LSC)

Tritium Detection and Measurement

- Portable Monitors
  - Commonly Ion Chambers
  - Battery Powered,
  - Light Weight,
  - Different Sensitivities,
  - Pre-settable Alarms.

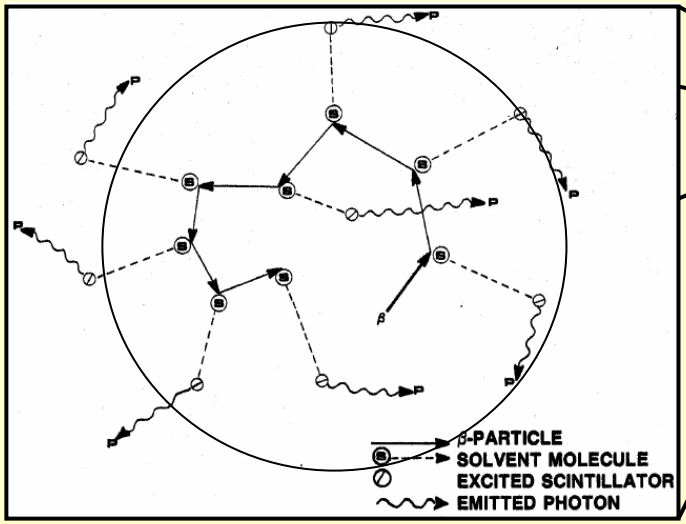
- Liquid Scintillation Counter (LSC)

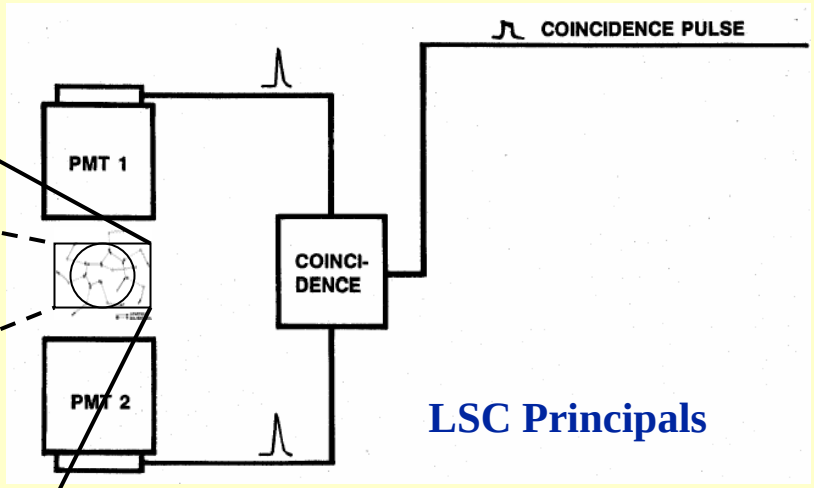
T Sample Analysis Using LSC

Principals of LSC (Coincidence):

Beta interacts with Cocktail and generated light which is counted by two PMTs in coincidence,

Beta Interaction with Cocktail



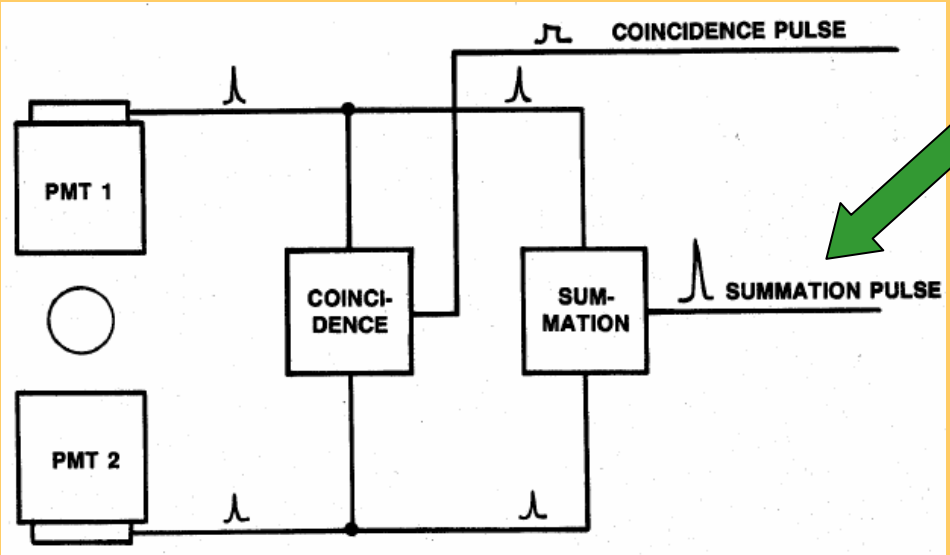


LSC Principals

T Sample Analysis Using LSC

Principals of LSC (Summation):

Beta interacts with Cocktail and generated light which is counted by two PMTs in Summation; this provides an output that is representative of the total energy of Beta, regardless of the location of Beta interaction in the Cocktail:

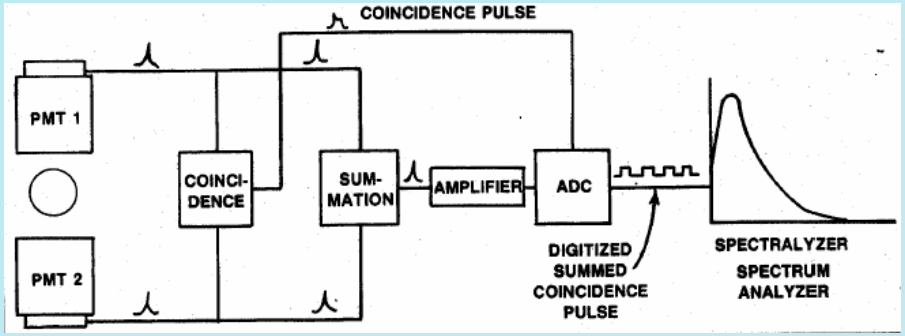


Sample Analysis Using Liquid Scintillation Counting (LSC)

Components of a Basic LSC Energy Spectrum Analyzer

Components:

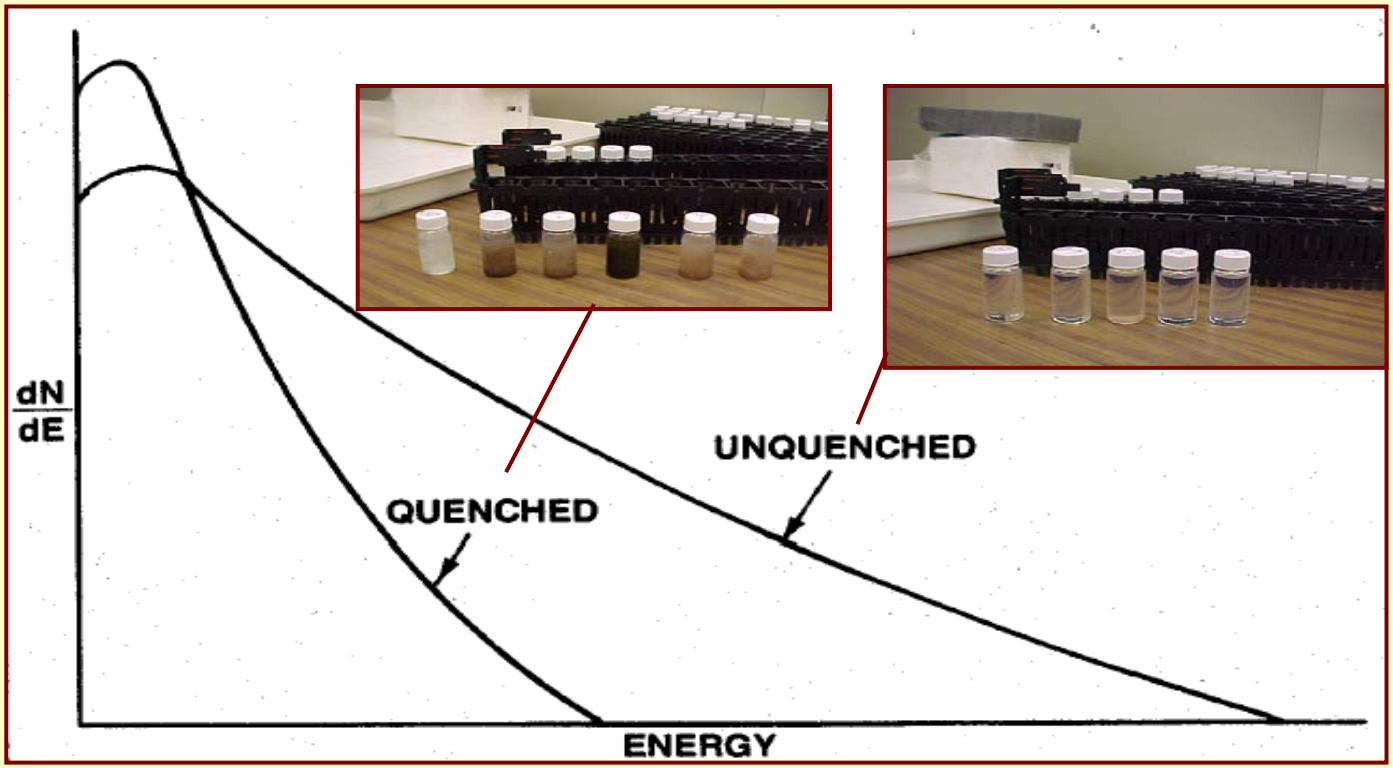
- Sample in Holder (Vial)
- Two PMTs
- Coincidence Circuit
- Summation Circuit
- Pulse Processing:
  - Amplifier
  - ADC
  - Spectrum Analyzer
  - Others



LSC Counting Interference and Spectrum Analysis

Quenching: May reduce the counting efficiency;

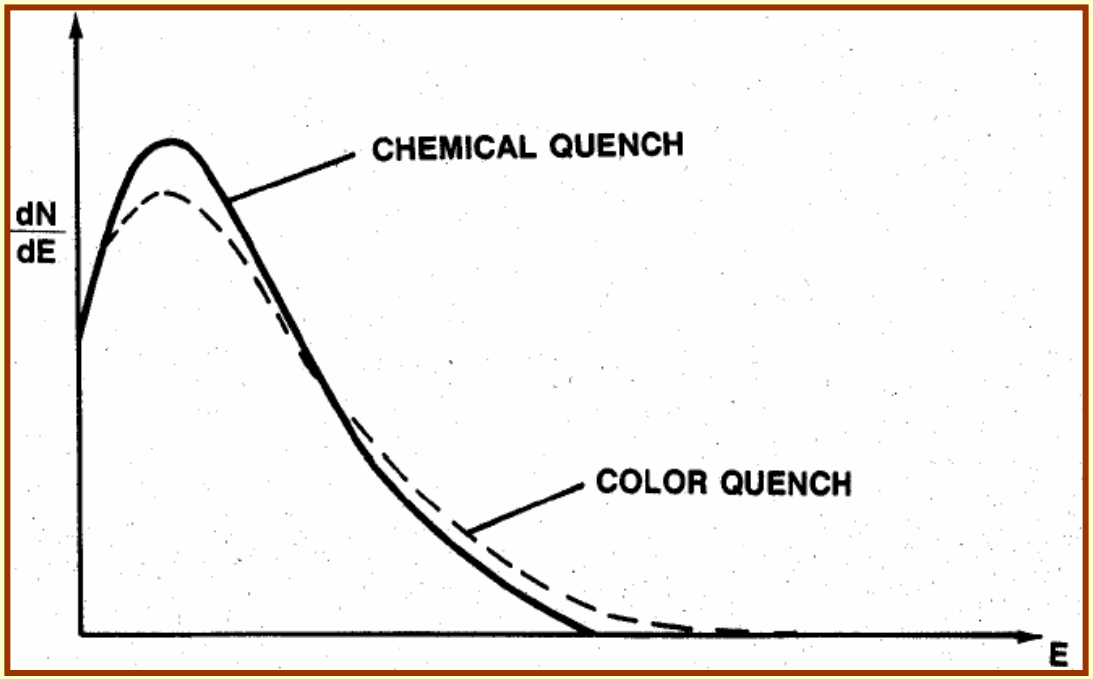
- Chemical quenching (impurity quenching) – causes energy losses in the transfer from solvent to solute:



LSC Counting Interference and Spectrum Analysis

Quenching: May reduce the counting efficiency;

- Color quenching – is the attenuation of light photons in the solution.



- Quenching results in the shift of the spectrum to lower energies.

LSC Counting Interference and Spectrum Analysis

Chemiluminescence: is production of light as a result of a chemical reaction.

- Most typically occurs in samples of alkaline pH and/or those containing peroxides, when mixed with emulsifier-type scintillation cocktails.

It can be controlled by:

- Using electronics logic on board scintillation counters, or
- Through additional sample preparation methods

Most modern LSC Systems perform a correction using on board electronics logic.

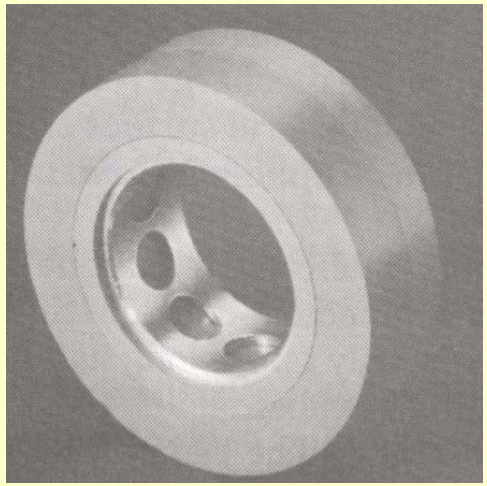
LSC Counting Interference and Spectrum Analysis

Static Electricity: Electrostatic Discharge (ED) is a phone producing interference in LSC.

- Teflon or human skin and polyethylene can generate electrostatic discharge.
- Static charge can develop in the scintillation vials or the cocktail.
- Static charge can on the vials can be produced as a results of handling.

ED will result in random spikes in the spectrum.

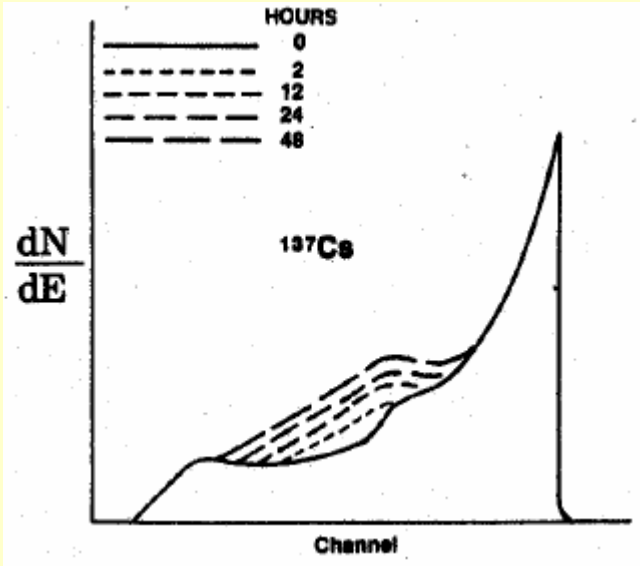
Electrostatic Controllers are used in modern LSCs to dissipate the static.



LSC Counting Interference and Spectrum Analysis

Wall Effects: Common organic solvents in the scintillation cocktail can penetrate the walls of plastic vials which then indirectly affect the counting accuracy of the sample.

The sample energy spectrum is not affected by this phenomenon but the Compton spectrum, induced by the external standard and used to calculate an external standard Quench indicating parameter, can become heavily distorted.



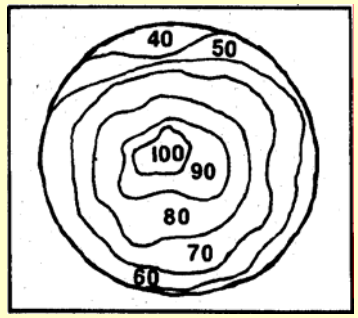
The impact is higher for increased quenching.

Modern LSC system provide for the correction due to this phenomenon.

LSC Counting Interference and Spectrum Analysis

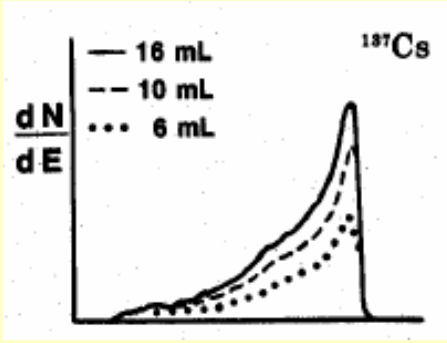
Scintillation Volume Variations: *Two problems*:

1) As the volume decreases, the light output falls on the less efficient areas of PMT. This results in the spectrum to shift towards lower energy (similar to quenching).



PMT areas of equal response

2) Smaller volume size skews the Compton spectrum from the external source, that is used in the energy calibration, and hence the results—diminishes spectrum.



Modern LSC provides for determining if the interference is a problem.

LSC Counting Interference and Spectrum Analysis

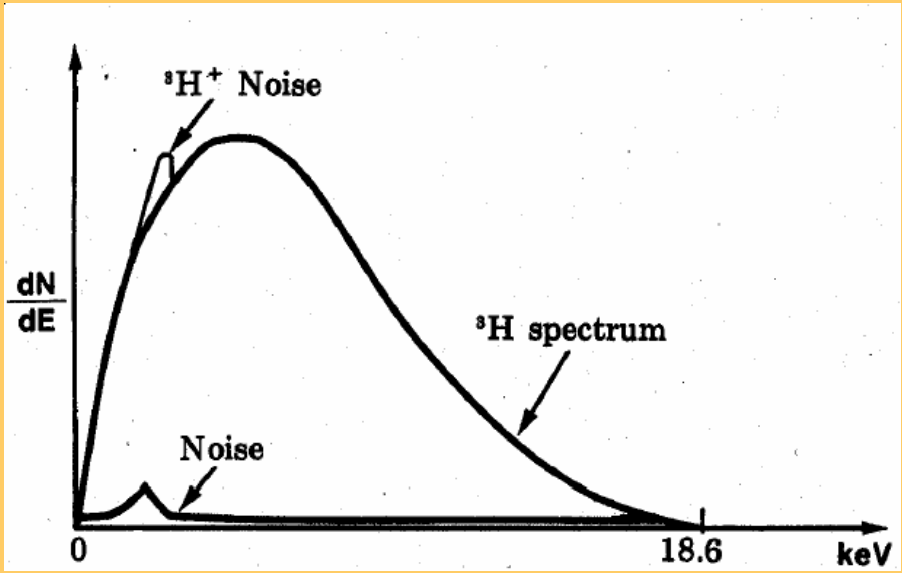
Heterogeneous Samples: Result in under estimation of the activity due to self absorption of Beta in the un-dissolved (insoluble) portion of the sample.

Can happen for samples that include compounds isolated on solid supports such as on paper chromatograms, filter paper and in polyacrylamide gels.

Chemistry is very important in the sample preparation for LSC.

LSC Counting Interference and Spectrum Analysis

Random Noise: Electronics noise can originate from the HV transients, line transmitted switching noises, and radiofrequency noise (switches, motors, relays and fluorescent lights). These noise pulses usually show up as single energy peaks superimposed on the spectrum:

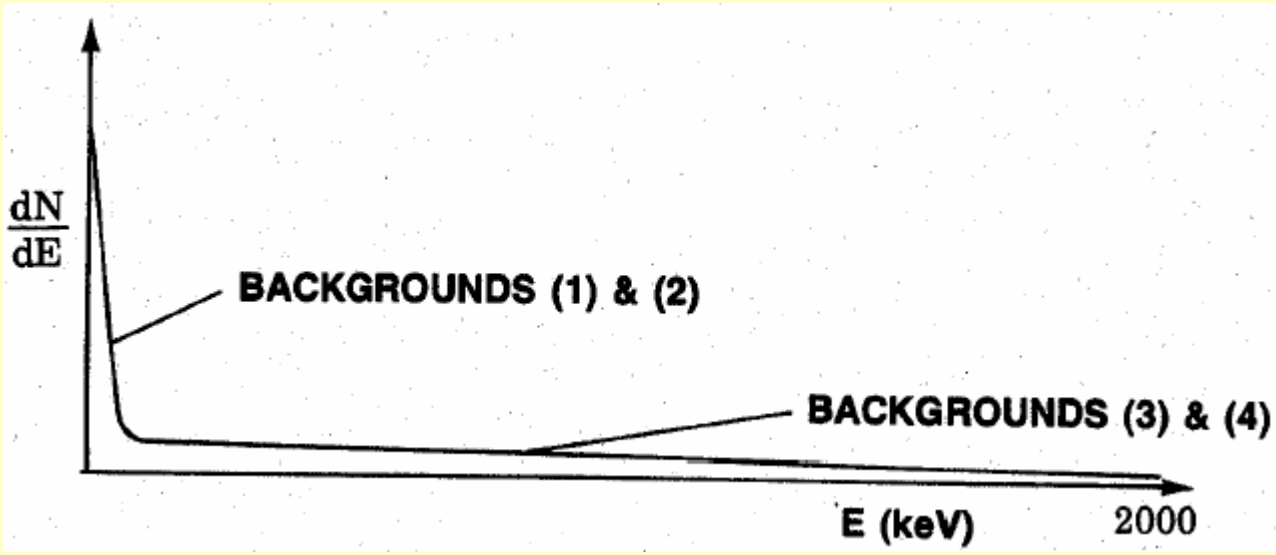


Digital filters can be used to eliminate these peaks—smoothing the spectrum

LSC Counting Interference and Spectrum Analysis

Background: Most common sources of background are:

1) Instrument ~ 10% : PMT (dark current, after pulse noise), low energy
2) Crosstalk ~ 22%: PMT cathode photoelectrons
3) Vila and PMT face ~ 37%: Material in vial and PMT (e.g. ^{40}K)
4) Scintillator (induced by cosmic rays) ~ 31%.

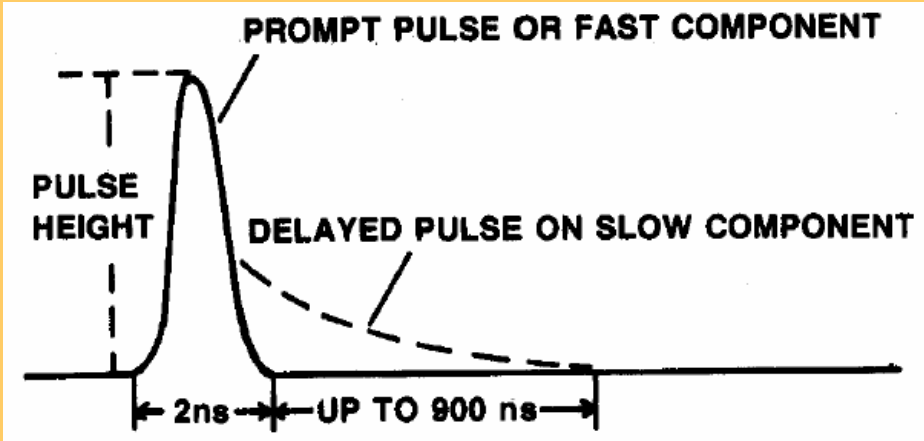


Sample Analysis Using Liquid Scintillation Counting (LSC)

Analysis Software (routines)

Note: In LSC

Beta energy is based on the area under pulse not the pulse height (as in Gamma Spect):



Modern LSC include the software package for analysis of spectrum and quantification of radioactivity contents of sample.

Reporting: General

Procedures must be established for management of all records associated with receipt, preparation, analyses, and disposal of samples and their derivatives:

1. These include the creation, distribution, use, maintenance, and disposition of the records.

2. The procedures are designed to comply with all applicable rules and regulations, such as local and state regulations, and the institution's procedures.

3. They provide for the systematic control of information (from creation to disposition) generated by any of the laboratory's information systems or media: paper, microfilm, or electronic.

Reporting: T Sample Analysis Data (1)

The Analysis report should, as a minimum, include:

- Sample information:
  - Description, Identification number (code), Collection Date and Time, Location of Collection
- Analysis information:
  - Date, time, and location (lab name) of analysis
  - Detector name, Identification number(code)
  - Calibration files (energy, efficiency, geometry), QC files (daily / weekly check files)
  - Minimum Detection Levels, MDL, (units must be consistent with the reported results for the sample), Decision Level, DL, for each nuclide analyzed
  - Volume, weight, or quantity of the sample
  - Counting time

Reporting: T Sample Analysis Data (2)

The Analysis report should, as a minimum, include:

- Analysis Results:
  - Calculated concentration or activity values in the units requested by the customer, Estimates of the uncertainty in the calculated concentration or activity
  - Identify the results that are below the MDA or DL
- Analyst and QA Review:
  - Name and signature of the analyst (dated)
  - Name and Signature of the QA reviewer (dated)
- Others
  - Other information requested by the customer per contract agreement

References

1. Merril Eisenbud, “Environmental Radioactivity,” 3rd Edition, Academy Press, 1987.
2. W. D. Ehmann and D. E. Vance, “Radiochemistry and Nuclear Methods of Analysis,” Wiley-Interscience, 1991.
3. Michael Kessler, “Liquid Scintillation Analysis, Science and Technology,” Packard Instrument Co, 1989.
4. Yutaka Kobayashi, “Liquid Scintillation Analysis-Packard Tri-Carb Laboratory Manual,” Packard Instrument Co, 1988.
5. Presentations by A. H. Mohagheghi, F. Ghanbari, and Sonoya Shanks, Sandia National Labs. Contact F. Ghanbari.