IAEA
International Atomic Energy Agency

LESSON 7:

TRITIUM AND C-14 MONITORING

TRITIUM MONITORING

Tritium Monitoring

- Background and significance
- Techniques
- Real Time Measurements
- Estimation of low energy beta activity

Background and Significance

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- Tritium is a low energy beta emitter with E_{max} of 18 keV and needs special techniques to measure

- Tritium significantly produced in PHWR reactors.
  - Nearly 2/3 of internal dose is due to tritium intakes

- Fusion reactors use tritium as fuel

- To a lesser extent:
  - H-3 is a byproduct of nuclear ternary fission (0.01%) in light water
  - H-3 found due to neutron capture by Li^6 and B^{10} in control rods

- H-3 can be found as tritiated water (HTO), Gaseous tritium (HT) and organically bound tritium (OBT)

Tritium Dosimetry and Radiotoxicity

- ☐ Exposure to tritium oxide (HTO) is by far the most important type of tritium exposure and HTO enters the body by inhalation or skin absorption.
- ☐ The total effective dose from airborne HTO exposure is about 10,000 times more than the total effective dose from an equal exposure to HT gas.
- ☐ Tritium is classified as low toxicity according to IAEA classification scheme (Group 4).
- ☐ Tritium should be in higher concentration to pose airborne contamination hazard.
- ☐ Therefore, safe handling of tritium is recommended.

Tritium Contamination

- ❑ Highly contaminated metal or plastic surfaces may release some of the loosely-bound tritium immediately after exposure to the contaminating tritiated atmosphere or liquid. This is called outgassing or offgassing and can result in significant airborne concentrations.

- ❑ Tritium as HT or HTO will readily adsorb onto the surface of most metals (such as stainless steel, copper, or aluminum), plastics, and rubbers.
  - ▪ The tritium will remain fairly close to the surface unless the metal is heated to a high temperature.

- ❑ HTO diffuses even metallic cladding resulting in contamination in unexpected places

MONITORING TECHNIQUES

Tritium Monitoring

In the workplace tritium can exist as a gas (HT) or as an oxide vapour in the form of HTO.

- The sampling and measurement technique depends on the chemical form of tritium.

Due to its low beta energy direct monitoring of tritium is not possible unless the air is mixed thoroughly with the detector medium.

Monitoring of tritium may be accomplished by real time measurement or by air sampling and delayed measurement of the sample in an appropriate measurement assembly.

Tritium Sampling

1

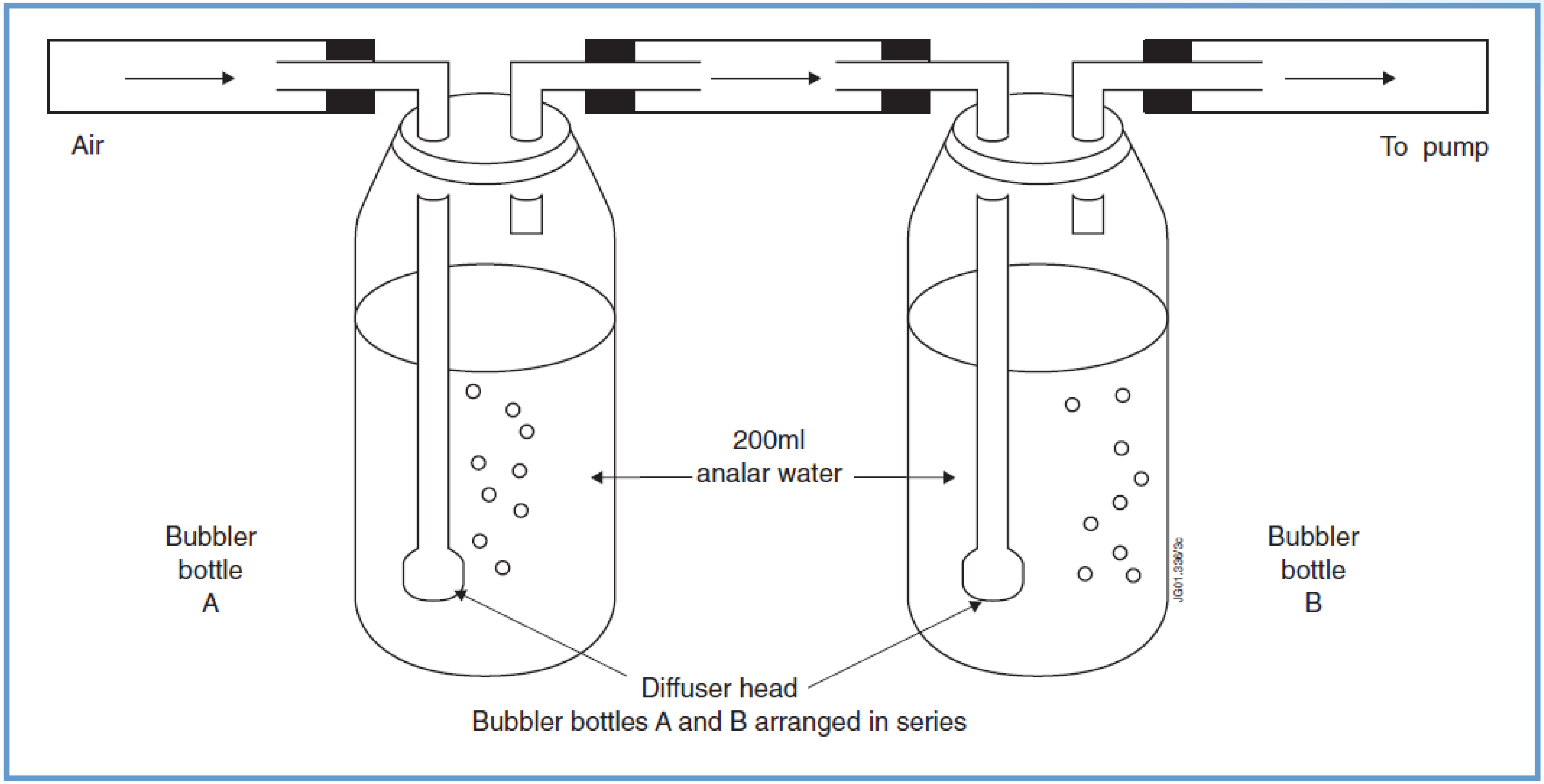
Sampling of tritiated water vapour can be carried out by the following techniques:

- Dynamic sampling
  - Condensation either using a cold strip or a cold finger.
- Passive Sampling
  - Condensation either using a cold strip or a cold finger.
  - Absorption in solid absorber such as silica gel, zeolite, etc.

2

The activity of water collected from the bubblers or from the solid absorbers (through heating or condensation) is measured by liquid scintillation

Bubbler for HTO sampling



Bubbler bottles A and B arranged in series

Diagram Bubbler sampler operation

Delayed Measurement using Bubblers

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The sampling air passes through two sets of bubblers in series filled with clean de-mineralized water and are separated by an oven.

- The oven and second bubbler are not required if it is known that tritium is present only in the form of water vapour.

The first bubbler traps the tritiated vapour.

The tritium gas is oxidised in the oven using a suitable catalyst to convert it into HTO.

When sampling with bubblers during a long time, for example 1 week, it is recommended to use a bubbler equipped with a cooling device in order to avoid the evaporation of the water.

Teflon should be avoided for sampling lines

HTO and HT Sampling

Airborne and gas form Tritium sampling device

Courtesy: Southern Scientific

HTO and HT Sampling

- Trapping yield of 99%
- Reduced evaporation due to the cooled trapping system, allowing weekly collection
- Easy to use, with instant opening cabinet for sample retrieval
- Accurate – electronic, accredited airflow meter

Dynamic Samplers

Good points:

- Rugged equipment.
- Sensitive method.
- High sampling rate.
- Sampling rate adjustable (typically from 10 up to 60 l/h).
- High collection efficiency (>95%).
- Stability of the sampling flow rate.
- Sampled volume is accurately known.
- Possible to use for a long time sampling.

Dynamic Samplers

Weaknesses:

- Require external power.
- Dilution of HTO sampled.

Check for:

- Temperature of the oven.
- Change the catalyst as required by the manufacturer (generally once a year).

Dynamic Samplers

- ☐ 20 ml vial containing tritium free water or water absorbent as the HTO trap
- ☐ In some cases, solid absorbers such as zeolite or silica gel are used as a tritiated water trap.
- ☐ The top of the vial is fitted with a diffusion orrifice
- ☐ The steady flow into the vial is maintained by the concentration gradient produced by the reduced level in the closed vessel compared with ambient air
- ☐ A desorber is required to remove the water from the solid absorber
    - The desorbed water is collected in a device at low temperature.

Passive Samplers

Good points:

- Small samplers.
- Require no external power
- Inexpensive to purchase, maintain and service.
- Simple to use.
- No dilution of the HTO sampled

Passive Samplers

Weaknesses:

- Two different samplers to know HT and HTO concentration (one with a a catalyst).
- Only use for short sampling time in case of total tritium sampler (between 15 and 30 minutes).
- Low sampling rate.

Check for:

- Evaporation of the liquid.
- Change the catalyst after each sampling for total passive sampler.

REAL TIME MEASUREMENTS

Real Time Measurement

For real time measurement of tritium location of the detector should be done judiciously so that area background radiation is low.

For real time measurement, the sampled air passes through a measurement cell.

Measurement cell is warmed to prevent condensation

Because of the low beta energy of tritium, the measurement cell is the detector itself, for example an ionization chamber or proportional counter or a plastic scintillator cell of the flow-through type.

Real Time Measurement

- ❑ A prefilter is put upstream of the measurement cell to trap particulates from the air sampled.
  - ▪ The prefilter should include an iodine trap if necessary - iodine can be a precursor to noble gases - ensure does not impact the result
  - ▪ The pre-filter should not trap or temporarily retain tritium or decrease the flowrate or the pressure inside the measurement cell over the limits specified by the manufacturer

- ❑ Interference of noble gas should be minimised while tritium measurement.
  - ▪ Compensation for noble gases contribution is difficult to achieve in ionization chamber based instruments.
  - ▪ In the case of proportional counters or plastic scintillators, the contribution due to noble gas can be reduced by taking advantage of difference in pulse height produced by beta particles of noble gas and tritium.

EXAMPLES OF REAL TIME MONITORS

Tritium air monitoring using ionization chamber

Fixed ionisation chamber instruments are most suitable for gaseous form.

Tritium beta particle ionizes the surrounding gas.

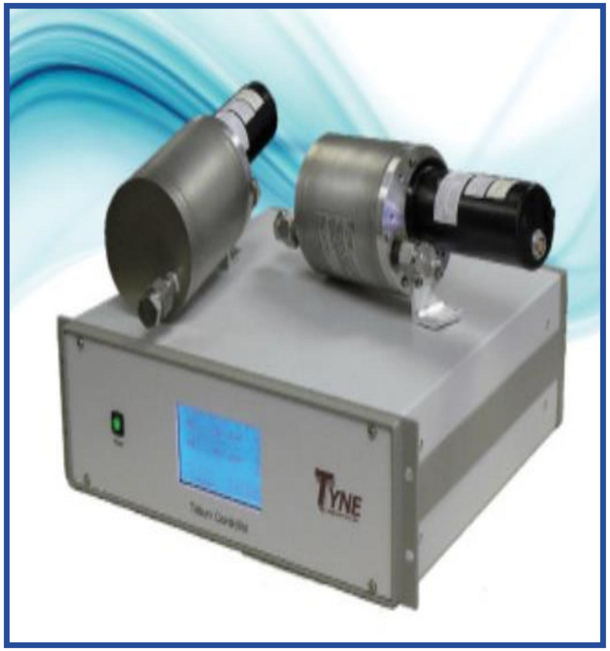
Portable instrument could supplement the fixed ones

Chamber volumes ranges from tenth to few tens of litres depending on required sensitivity – flow rates a few lpm

Gamma and radon compensation is crucial.

Portable and Installed Tritium Air Monitore

Portable tritium monitor
With dual ion chamber



Installed tritium monitor
With dual ion chamber

Ionisation Chambers

Good points:

✓

- Small detector due to the distance covered by the beta particle of tritium in air.
- Good sensitivity for tritium.
- Low maintenance cost.

Weaknesses:

✗

- Generally non specific for tritium measurement.
- Sensitive to temperature.
- Sensitive to pressure: the response is quite proportional to the pressure.

CARBON -14 MONITORING

Sources of ^{14}C in Nuclear environment

Carbon-14 and tritium are weak beta emitters and do not present an external radiation hazard.

The major ^{14}C producing neutron activation reactions in nuclear power reactors are:

(a) The $^{14}N(n,p)^{14}C$ reaction

(b) The $^{17}O(n,\alpha)^{14}C$ reaction

(c) The $^{13}C(n,\gamma)^{14}C$ reaction

(d) The $^{15}N(n,d)^{14}C$ reaction

(e) The $^{16}O(n,^{3}He)^{14}C$ reaction

Chemical Form of Carbon -14

C-14 can be in the forms of CO_2, CO or hydrocarbons.

Techniques for sampling $^{14}CO_2$ in air can be either active or passive.

Carbon mono-oxides and hydrocarbons can be converted to CO_2, by a catalyst at high temperature.

This is required as it is easy to measure C-14 activity in CO_2 form.

The real time monitors used for tritium sampling may be suitable for C-14 also.

Sampling

- The carbon in CO_2 form may be trapped by bubbling through a solution of sodium hydroxide or in a solid absorber like zeolite.

  - The air to be sampled passes through bubblers filled with sodium hydroxide solution in the range 0.8–4M (or tubes filled with solid absorber), an oven and a second set of bubblers (or tubes filled with solid absorber).
  - The first bubbler (or tube) traps carbon in CO_2 form.
  - The organic molecules are broken and the carbon oxidized to CO_2 form in the oven using a catalyst made of Platinum/Alumina pellets.
  - CO_2 is trapped in the second bubbler or solid absorber connected at the outlet of the oven.

Release of C-14 for Measurement

When the CO_2 is trapped in a solid absorber it has to be desorbed by heating at the end of the sampling time.

  - A temperature of 400 degrees Celsius is required for zeolite

CO_2 desorbed is trapped by bubbling through a solution of sodium hydroxide.

C-14 activity counted in LSS using Toluene based scintillator.

Calibration and Testing

The following should be calibrated:

- The flow meter associated with the sampling assembly should be calibrated against a national standard.
- The oxidation efficiency should be determined by sampling standard tritrium gas or organic carbon. Application of low concentration hydrogen or carbon and mass spectrometry can also be used for determination of the oxidation efficiency.
- The measurement assembly with a standard source of tritiated water, C-14 or, for monitors, with a radioactive source

Functional checking should include the detection efficiency of the measurement assembly and the sampling flow rate or volume

ANALYSIS METHODS

Liquid Scintillation Counting

For Liquid Scintillation Counting the sample to be counted is mixed with a mixture of certain organic chemicals, in a specified proportion, called a cocktail.

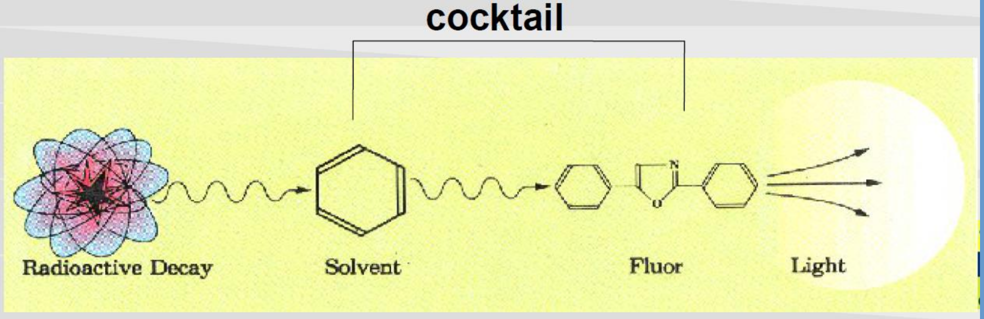
Generally the cocktail consists of a primary solvent (Dioxane for aqueous samples and Toluene for organic samples), a secondary solvent (Naphthalene only for aqueous samples), a primary solute (PPO) and a secondary solute (POPOP).

A vial containing the sample and the cocktail is placed in the measurement cell of the liquid scintillation counting system.

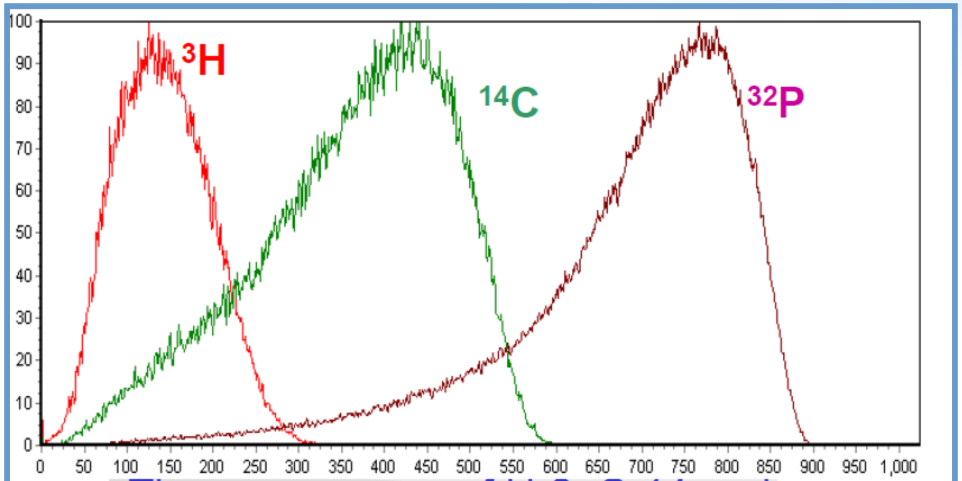
Liquid Scintillation Counting

- The scintillations produced in the cocktail by beta particles from tritium are detected by suitably positioned photomultiplier tubes.

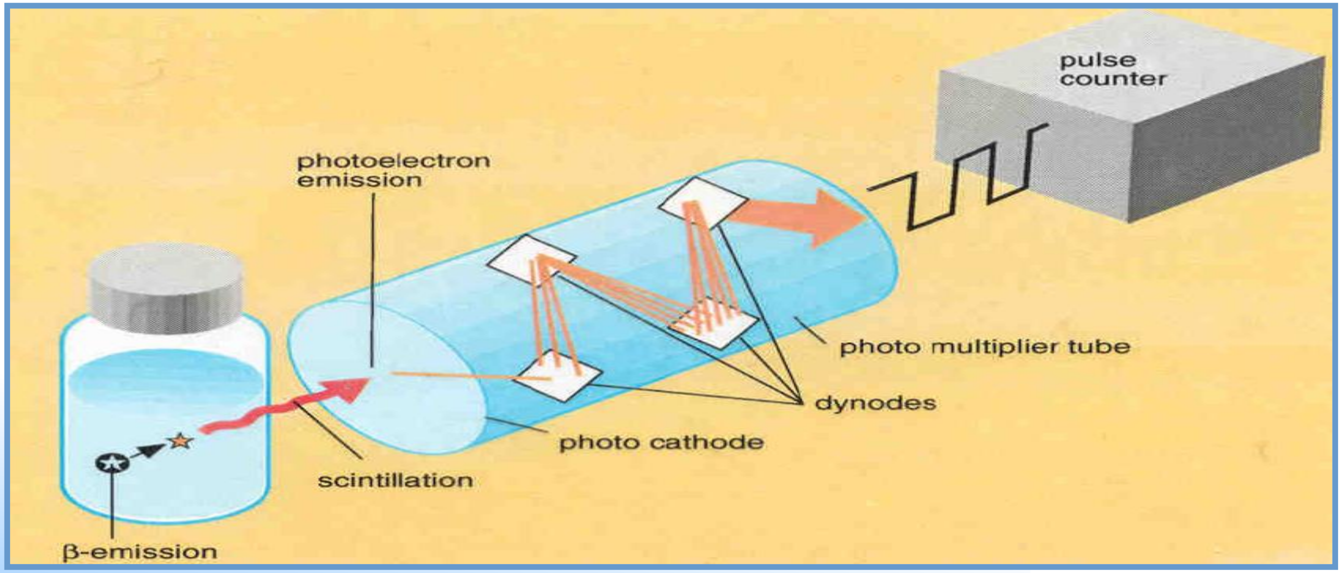
- The light output resulting from the scintillations can get reduced in the cocktail due to various mechanisms, commonly known as quenching. This will result in a reduction in the efficiency of the detector.



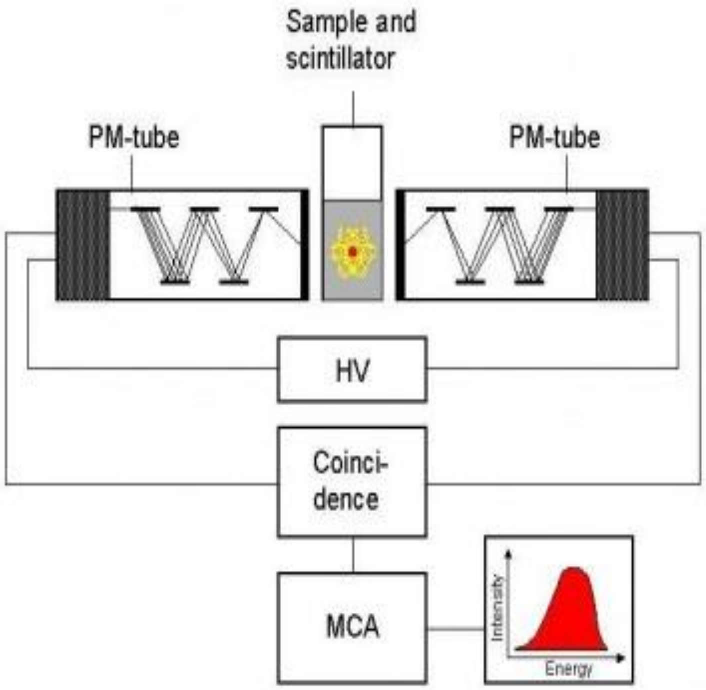
Beta Spectrum and Counting



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

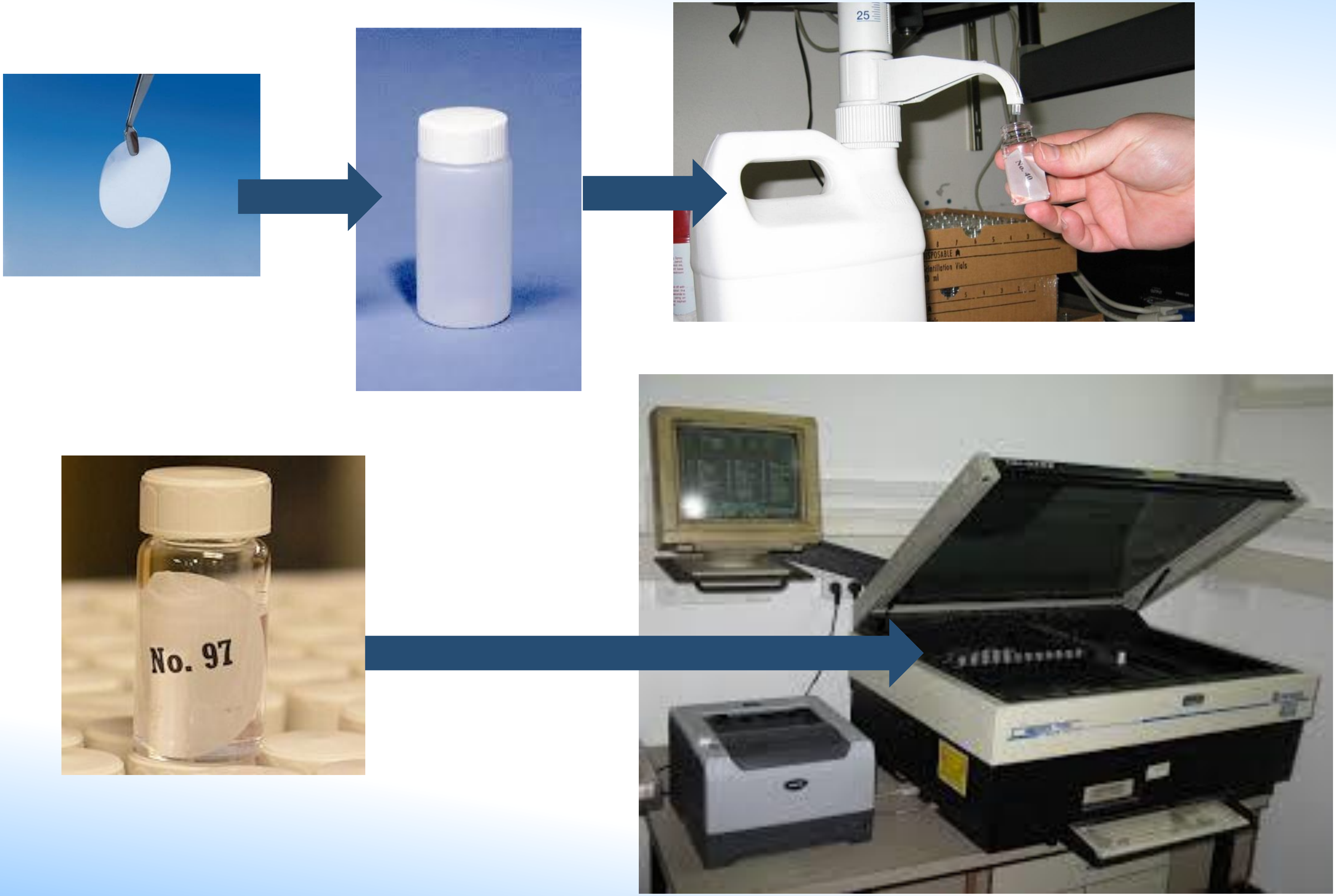


Liquid Scintillation Operations



- ☐ Light flashes detected
- ☐ Number determined
- ☐ Intensity determined
- ☐ Number of flashes per minutes is recorded as counts per minute
- ☐ Converted to nuclear desintegration rate
- ☐ Data analysis

Liquid Scintillation Counting



Liquid Scintillation Counting

Interference in liquid scintillation counting;

- Background (noise and PMT crosstalk)
- Static electricity
- Wall effect (organic solvent penetrate the wall)
- Radionuclide mixture (spectral overlapping due to presence of two radionuclides in sample)
- Luminescence
  - Chemiluminescence
  - Bioluminescence and photoluminescence.

Practical Aspects

- Solvent has to be compatible with the liquid scintillation cocktail.
- Spills of cocktail on outside of vial should be avoided to prevent quenching.
- Process should be clean to prevent luminescence or quenching.
- Make sure energy range is correct for radionuclides to be detected.