CHAPTER 4

RADIATION DETECTORS AND SURVEY INSTRUMENTS: THEORY OF OPERATION AND LABORATORY APPLICATIONS

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RADIATION INSTRUMENTATION

I. PORTABLE SURVEY INSTRUMENTS

The major principle for sensing and measuring radiations in survey instruments is based on the ionizations radiation produces when interacting in a gas filled detector. As described in the previous chapter, radiations passing through matter create ion pairs. In a detector, these ion pairs are collected to form an electrical signal through the use of an electric field. The signal, either a current or a pulse, is then used to register the presence or amount of radiation. There are a number of different types of radiation detectors, each operating on this basic principle, but designed for specific purposes. The two major types of portable radiation survey instruments, the Ion Chamber and Geiger Counter, are discussed below.

A. Ionization Chambers

Ionization chamber type instruments are designed to measure exposure rates of ionizing radiations in units of mR/hr or R/hr. The detector is usually cylindrical, filled with air and fixed to the display instrument. When radiation interacts with the air in the detector, ion pairs are created and collected generating a small current. Since we have defined the Roentgen in terms of the amount of ionization charge deposited in air, measurement of this ionization current will indicate the exposure rate.

B. Geiger Counter

The most common type of portable radiation survey instrument is the Geiger Counter, also know as a Geiger-Müller (G-M) Counter. The G-M counter's detector consists of a tube filled with a mixture of "Q-gas", containing 98% helium and 1.3% butane. This tube can usually be removed from the display instrument to survey an area. Instead of measuring the average current produced over many interactions, as in Ion Chambers, the output is recorded for each individual interaction in the detector. Thus, a single ionizing event causes the G-M tube to produce a "pulse" or "count". Because all pulses from the tube are the same size regardless of the number of original ion pairs that initiated the process, the G-M counter cannot distinguish between radiation types or energies. This is why most G-M counters are calibrated in "counts per minute" (cpm). However, G-M counters can be used to measure exposure rates in mR/hr or R/hr as long as the energy of the x- or gamma radiation is known and the instrument is calibrated for this particular fixed energy. At best, for a given x- or gamma ray energy, the count rate will respond linearly with the intensity of the radiation field. However, in most applications, the radioactive source will have x- or gamma rays of various energies which can result in erroneous and unreliable readings. Therefore, G-M counters are primarily used to detect the presence of radioactive material.

II. USE OF RADIATION SURVEY INSTRUMENTS

Radiation instruments are designed with a specific purpose in mind. Choose the instrument depending on your particular needs. Generally, Geiger Counters are more sensitive than Ion Chambers at monitoring low levels of contamination in the laboratory. If you wish to measure radiation levels in the laboratory, then an Ion Chamber is the proper instrument to use. Each instrument comes with an operating manual that describes its function and limitations such as warm-up time, battery life, operating temperature range, minimum sensitivities, etc. Outlined below are simple instructions on the proper use of portable radiation survey instruments.

- 1. <u>Read the instrument's operating manual</u>. Gain familiarity with the controls and operating characteristics.
- 2. <u>Check the batteries</u>. Most instruments have a battery check indicator. Replace weak batteries. Turn off the instrument when not in use. When storing the instrument for extended periods, remove the batteries to prevent damage from leakage.
- 3. <u>Check the operability of the detector</u>. Pass the detector over a radioactive check source (sometimes attached to the side or end of the instrument) to verify that the detector responds to radiation.
- 4. <u>Determine the instrument's response time</u>. By passing the detector at varying speeds over a check source, you can determine how long it takes for the detector to respond to the radiation. It is possible to miss contamination or radiation fields if the detector is moved too rapidly over the area being surveyed.
- 5. <u>Determine the operating background</u>. Note the instrument's response in an area free of contamination or radiation levels. This is normally due to natural sources of radiation called "background". Naturally occurring radiation arises from three sources: cosmic rays entering the earth's atmosphere, naturally occurring radioactive materials in the earth's crust, and naturally occurring radioactive materials within the body. Subtract this background value from the "gross" reading to obtain the "net" result due to the sample (S) itself:

 $\mathbf{S}_{\text{net}} = \mathbf{S}_{\text{gross}} - \mathbf{S}_{\text{background}}$

When using portable instruments, caution should be used when extending detector cords as this may generate electrical noise and register as "counts". Also, the window G-M tubes use to detect alpha and low energy beta particles are fragile and can easily break if dropped or punctured. In a mixed beta-gamma field, the reading due to beta radiation only will be the reading with the beta shield off the detector minus the reading with the beta shield on the detector.

III. CALIBRATIONS AND EFFICIENCY

In order for the results of a survey instrument to be meaningful, the instrument must be calibrated. Calibrations should be performed at least every six months or when battery or test

functions indicate a problem. Ion Chambers are usually calibrated against Cs-137, Co-60, or a x-ray radiation field. The true exposure rate is determined by multiplying correction factors (if any) by the reading on the instrument. G-M counters are usually calibrated against a specified reference standard at a fixed distance from the detector (usually 1 centimeter) and a variable pulse generator.

Efficiencies for instruments expressing results in terms of count rates can be calculated from the following formula:

 $Efficiency = \frac{Observed S \tan dard Count Rate (cpm)}{Known S \tan dard Di \sin tergration Rate (dpm)}$

The observed sample count rate can then be divided by the detector efficiency to obtain the actual disintegration rate.

Example: A Carbon-14 standard has a disintegration rate of 85,000 dpm. Your G-M counter measures a count rate of 4500 cpm. If the background is 250 cpm, what is the efficiency of the counter?

 $Efficiency = \frac{4500 \, cpm - 250 \, cpm}{85,000 \, dpm} = 0.05 \, cd \times 100 = 5\%$

IV. GEIGER-MÜLLER COUNTERS - DETAILED

A. G-M Tube Characteristics:

- 1. Radiation only triggers a discharge. Any particle which produces an ionization in the tube will produce a discharge.
- 2. Sensitive to x- and gamma rays, beta particles, and other charged particles.
- 3. Variety of detector shapes, sizes, and windows.
- 4. Large output signal (approximately 1 volt). Output pulse size is independent from the primary ionization which initiates it and gives no information as to the nature of the particle or its energy.

B. G-M Tube Description:

- 1. G-M tubes are usually cylindrical in shape. The tube contains two electrodes and a filling gas.
- 2. The collector electrode (anode) is mounted coaxially and insulated from the outer electrode (cathode) which is a ground potential and is usually part of the tube structure.
- 3. Filling gases which are usually noble gases, are used because they have low electron attachment coefficients to allow negative charge transfer in the tube from free electrons.

- 4. G-M tubes require a high voltage source to supply the voltage between the cathode and the anode.
- 5. Primary ionization in the G-M tube initiates a series of events that result in a sheath of ion pairs surrounding the anode. The free electrons are collected quickly because of their high mobility, the strong electric field near the anode, and their attraction to the positively charged anode. A positive ion sheath then results and terminates the buildup of the discharge by reducing the electric field strength.

C. Counting-Rate versus Voltage Curve:

- 1. The dependence of the pulse size on applied voltage in a gas-filled detector is shown in Figure 1.
- 2. A curve of counting-rate versus the voltage applied to the G-M tube is referred to as the characteristic curve as shown in Figure 2. The geometry of the radioactive source as well as the discriminator setting is held fixed while obtaining data for the curve.
- 3. Under normal operating conditions, the curve has a plateau with a small slope and a length of several hundred volts. A relative plateau slope of less than 10% per 100 volts is acceptable.
- 4. V_s is the voltage which produces pulses that are larger than the discriminator setting and are counted by the scaler. Alpha particles, because they are highly ionizing, require a lower starting voltage than beta particles.
- 5. The knee of the curve represents the condition under which all particles, no matter how small their primary ionization, produce output pulses that are larger than the discriminator setting.
- 6. The finite slope of the plateau is due to the extension of the sensitive volume of the tube as the voltage increases, and the production of occasional spurious pulses which increases as the voltage increases.



Figure 1 Output Pulse Height versus Applied Voltage for both alpha and beta particles for a G-M tube.

Applied Voltage (hundred volts)



Figure 2

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D. Production of the Discharge:

- 1. Electrons produced in the G-M tube by the primary ionization drift toward the anode undergoing collisions along their path. Electrons gain energy when traveling toward the anode and within the last few mean free paths of the anode, produce secondary ionizations.
- 2. The buildup of ionization is referred to as a Townsend avalanche. The initial avalanche terminates when all the electrons associated with it have reached the anode. The initial avalanche is followed by additional avalanches, each triggered by the preceding avalanche.

E. Quenching the Discharge:

- 1. As positive ions migrate to the cathode and are neutralized, an additional electron can be emitted as a result of this neutralization. This additional electron will result in another discharge unless it is quenched.
- 2. Self-quenched tubes contain a small percentage of a quenching gas in addition to the major constituent of the filling gas. There are two types of self-quenched tubes; organic-quenched and halogen- quenched.
- 3. In organic-quenched tubes, the energy released when the positive ion strikes the cathode goes into disassociating organic molecules rather than producing further ionization. When the organic gas is depleted to a sufficient extent the operation of the tube becomes erratic. This causes the plateau to be shorter and to have a larger slope.
- 4. In a halogen-quenched tube the quenching gas is not consumed in the quenching process. The diatomic molecules are dissociated in the quenching but recombine after a short time interval.

F. Resolving Time:

- 1. When radiation produces an ionization in a G-M tube, the electrons travel to the anode very quickly, but the positive ions are much slower and require between 100 and 400 lsec to reach the cathode. During the time it takes the positive ions to get to the cathode, the tube is insensitive.
- 2. Deadtime is the time interval after a pulse has occurred during which the counter is insensitive to further ionizing events. Recovery time is the time required for complete recovery of the pulse size after the end of the deadtime interval.
- 3. Resolving time is the minimum time interval by which two pulses must be separated to be detected as separate pulses. Resolving time approaches deadtime but can be as large as deadtime plus recovery time.
- 4. The phenomenon of a second or third ionizing particle striking the tube during the resolving time is known as coincidence. Coincidence results in an observed counting rate that is less than the true counting rate (coincidence loss).

5. The true counting rate, R_t , is related to the observed counting rate, R_o , and the resolving time, T_r , by:

$$R_1 = \frac{R_o}{1 - (R_o T_{r})}$$

G. Windows:

To keep absorption of alpha and beta particles to a minimum, windows are made with the smallest density thickness that is possible. Window thickness of 1 to 2 mg/cm^2 are commonly used.

H. Counting Efficiency:

- 1. The efficiency of detection depends on many factors, most of which do not remain constant from one experiment to another. G-M tubes are nearly 100% efficient in detecting alpha and beta particles that reach their sensitive volume but only 1-2% efficient in detecting x- or gamma rays.
- 2. The efficiency of a detector is given by: $E = Gf_b f_s f_w f_r e_b$
- 3. The geometry factor, G, accounts for the fact that only a portion of the particles emitted by the source move toward the detector.
- 4. Backscatter is the phenomenon by which particles which originally are emitted in a direction away from the detector are scattered back toward the detector. The backscatter factor, f_b , depends on the thickness and atomic number of the backing material.
- 5. Self-absorption factor, f_s , takes into account the effect of the finite thickness of the source on the number of particles reaching the detector. Some particles are absorbed by the source and some are scattered toward the detector.
- 6. The absorption factor, f_w , is the ratio of the actual counting rate to that which would be obtained if there were no absorption between the source and the interior of the detector. Absorption is due to the window, air, and any other absorber placed between the source and the detector.
- 7. Deadtime correction factor, f_t , depends on the counting rate and is considered only at high counting rates.
- 8. Intrinsic efficiency, e_b , is defined as the fraction of the particles which, upon entering the sensitive volume of the detector, produce a discharge.

V. LIQUID SCINTILLATION COUNTING

Liquid Scintillation Counting is the most common technique for the measurement of radioactivity of low energy beta emitters. Such emitters (H-3, C-14, S-35, Ca-45, etc) are difficult to detect using portable survey instruments since the beta may not be able to penetrate the thin window of the gas filled detector. In liquid scintillation counting, the sample is dissolved in a counting solution. The energy of the beta is absorbed by solvent molecules causing them to become excited. This excitation energy is transferred to a solute (known as a

scintillator) resulting in a flash of light or "scintillation" when the scintillator molecules return to the ground state. The number of scintillations emitted is proportional to the energy of the beta particle. A photomultiplier tube (PMT) is used to detect and amplify the light photons from the sample. The emitted light causes the emission of photoelectrons from the PMT which are multiplied by the PMT into a measurable electrical pulse. The height (amplitude) of the pulse is proportional to the number of photons which interact in the PMT. Therefore, the pulse height at the output of the PMT is proportional to the energy of the beta particle and the rate of beta emission in the sample. It is also possible to count very low energy gamma emitters by liquid scintillation since most of the gammas are absorbed in the counting solution.

Not all pulses from the PMT are due to radiation from the sample. Pulses are generated by the electronics, the PMT and from environmental background radiation. These "noise" pulses are identical to pulses due to scintillations from a sample. To distinguish the pulses, two PMT's are arranged in a "coincidence" mode. Because noise pulses are random events, it is unlikely that two PMT's will receive a pulse simultaneously. But most beta particles have sufficient energy to produce more than one photon in the solution. Therefore, it is probable that both PMT's will simultaneously receive photons due to a single beta decay event. A coincidence circuit is established to check if a pulse from one PMT is accompanied by a corresponding pulse from the other. The requirement that both PMT's receive a pulse within a certain time (coincidence resolving time) excludes the vast majority of noise pulses from the sample count.

Beta particles will produce PMT pulses up to a maximum amplitude. An upper level discriminator (ULD) can be introduced to the system which can exclude pulses which have a greater amplitude than the maximum amplitude for the nuclide of interest. A lower level discriminator (LLD) can be arranged to exclude all pulses smaller in amplitude than a given value. A gain control is used to determine the PMT pulse height to which a given discriminator setting corresponds. Changes in gain alter the amplitude of the pulses before analysis by the LLD and ULD. The limits of pulse height accepted by a pair of discriminators and a gain setting is referred to as a "window" (see Figure 3). Correct settings of gain controls and discriminators will discriminate between pulses of a given nuclide from those of another. To separate pulses from beta events in samples containing nuclides of differing energies, a number of separate channels of pulse height analysis are necessary. The instrument's operating manual should be referred to for specific procedures on how to optimize the counter for each particular nuclide to be analyzed.



Pulse height spectrum of a beta emitter showing the effect of gain. (Gain settings Ga, Gb, and Gc). Note that the pulse spectrum is centered between the window set by LLD and ULD to give the maximum counting rate (Gain setting Ga).

A. Optimum Counting Conditions

By adjusting the gain and discriminator settings, different counting windows can be established. Some windows may yield a high sample count as well as a high background count. The optimum setting (based on statistics) for the window settings is given by the Figure of Merit:

Figure of Merit =
$$\frac{S^2}{S+2B}$$

Where S = sample counts ; B = background Counts

The larger the Figure of Merit, the more significant the sample measurement is.

B. Counting Efficiency and Quenching

Counting efficiency depends on the windows used and the ability of the beta particle to interact with the scintillator to produce light-emitting events. A decrease in the ability to produce or transfer light to the PMT's is called quenching and occurs mainly from the

optical properties of the sample (i.e. color and/or turbidity) called photoluminescence, or the chemical composition of the sample, which is called chemiluminescence. Samples containing equal amounts of activity of the same nuclide can produce different count rates due to quenching.

Since quenching reduces the number of photons which reach the PMT's, the scintillations appear as beta particles of lower emission energies. The effect of quenching is a shift in the pulse height spectrum (see Figure 4). Thus, some low energy events which would normally exceed the coincidence threshold in unquenched samples will produce insufficient photons for detection in quenched samples.

Figure 4



Effect of Quenching on Pulse Height Spectrum

Because quenching occurs to some degree in all samples, a loss in counting efficiency will result. There are several techniques used to determine sample counting efficiency in a liquid scintillation counter but the three basic techniques are Internal Standard, Sample Channels Ratio and External Standard.

1. Internal Standard

The internal standard method for determining counting efficiency requires that the sample be counted in the usual manner, then a calibrated amount of a radioactive standard is added to the sample and the sample plus standard mixture is recounted. The increase in counts (due to the added standard) is used to determine the counting efficiency according to the following formula:

 $Efficiency = \frac{C2 - C1}{\operatorname{int} ernal s \tan dard \, dpm}$

where: C1 is net cpm of the sample without the internal standard and C2 is the net cpm of the sample with the internal standard.

In order to be most accurate, the material added as the standard should be of the same material as the sample, as to not introduce quenching, and added in a small volume (0.1 ml or less) so as to not alter the characteristics of the original sample. The amount of activity added must be accurately determined and should be equal to or greater than the sample activity.

2. <u>Sample Channels Ratio</u>

The channels ratio method of determining counter efficiency is based on the fact that the pulse height spectrum is always displaced when quenching occurs. A counter using two different channels for pulse height analysis can determine the shift. A set of quenched standards, each vial containing exactly the same amount of activity (dpm) but different amounts of a quenching agent, is used to establish a correlation between the ratios of the counts in the two channels and the corresponding efficiencies.

To use the channels ratio method to determine the efficiency of a single nuclide, one of the windows (Ch A) is set narrower than the normal window of analysis (Ch B) for that nuclide as shown in Figure 4. As the quenched standards set is counted, more and more counts will be shifted out of window B into window A. The counting efficiency in channel B and the net samples channels ratio (SCR) for each standard is calculated. A graph of the efficiency vs. SCR is obtained and a curve drawn:

Cou	nting Mode	: Single N	uclide (C-14)	(Counting Tin	ne: 1 N	linute				
Back	ground Co	unt Rate:	Channel A =	90 cpm;	Channel B =	27 cpr	n				
Que	iched Stand	lards Set:	10 samples, e	each conta	aining 97,60	0 dpm	of C-14				
Sam	ple #1 least	quenched	; #10 most qu	enched							
#	ChA	ChB	ChB/ChA	%			C	-14 E	ff vs. So	CR	
	cpm	cpm	SCR	Eff							
						100	1				
1	19787	87456	4.420	89.6						-	
2	22541	86171	3.823	88.3		80			/		
3	28738	82670	2.877	84.7				/	/		
4	34977	78970	2.258	80.9		60		/			
5	47505	71174	1.498	72.9	% EFF			/			
6	55311	62652	1.151	65.2		40					
7	63448	53135	0.8375	54.4							
8	65492	39859	0.6086	40.8		20	1/				
9	58511	24441	0.4177	25.1			ľ				
10	45768	11243	0.2457	11.5		0	1	2	3		4
									SCR		

SAMPLE CHANNELS RATIO CALCULATIONS

The SCR technique is of limited use in dual label counting and in low activity samples.

3. External Standard

The most widely used method for determining counting efficiency uses a highenergy gamma source positioned external to the sample vial in the counting chamber. The interaction of the gamma radiation with the vial produces electrons in the scintillation solution due to the Compton effect. The Compton electrons behave as beta particles, causing scintillations. The more quenched the sample, the fewer scintillations detected. These counts are then monitored by a separate (internal, usually factory pre-set) pulse height analyzer and directly related to an efficiency correlation graph, prepared from a set of quenched standards (see data table and graph page 12). Some counters use two channels of analysis to come up with an External Standard Ratio (ESR), which minimizes the effects of volume changes and changes in counting geometry.

EXTERNAL STANDARD RATIO CALCULATIONS

Background Count Rate: Ch A = 90 cpm; Ch B = 27 cpm Quenched Standard Sets: 10 samples, each containing 262166 dpm of H-3;	
Quenched Standard Sets: 10 samples, each containing 262166 dpm of H-3;	
10 samples, each containing 97600 dpm of C-14	
Sample #1 least quenched; #10 most quenched.	
C-14 Quenched Standard Set H-3 Quenched Standard Set	
Ch A Ch B % Eff Ch A Ch B % E	Eff
# cmp cmp ESR # cmp cmp ESR	
1 19787 87456 1.007 89.6 1 146489 59773 1.015 5	55.8
2 22541 86171 0.9745 88.3 2 124867 37083 0.9586 4	47.6
3 28738 82670 0.8976 84.7 3 107576 23921 0.8924 4	41.1
4 34977 78970 0.7816 80.9 4 89517 14000 0.7816 3	34.1
5 47505 71174 0.6622 72.9 5 64457 5728 0.6722 2	24.6
6 55311 63652 0.5567 65.2 6 42870 1989 0.5184 1	16.4
7 63448 53135 0.3921 54.4 7 28025 759 0.3449 1	10.7
8 65492 39859 0.1964 40.8 8 17313 255 0.1582	6.6
9 58511 24441 0.0322 25.1 9 9348 96 0.0154	3.6
<u>10</u> 45768 11243 0.0011 11.5 10 4770 41 0.0000	1.8

The External Standard method can be used to determine efficiency in any sample regardless of its radioactive content and is suitable for single and dual label counting as well as for samples of low activity.



C. Sample Preparation

In preparing samples for liquid scintillation counting, the physical and chemical characteristics of the sample determine the type of counting solution required. Many references are available on the types of solvents and scintillators for a particular application. The main objective is to produce a clear, colorless and homogeneous sample so that counting efficiencies can be determined by one of the described methods. However, it may not always be possible to achieve a homogeneous sample, for example when the radioactive material is isolated on filter paper, membrane filters, or gels. The determination of counting efficiency for such heterogeneous samples is a problem because it is difficult to duplicate the exact counting environment of the experimental samples. The proper method to determine the activity of a heterogeneous sample is to either elute the radioactivity or to solubilize the sample prior to counting. The internal standard method of efficiency determination is best suited for heterogeneous samples.

Another factor to consider when preparing samples for liquid scintillation counting is the introduction of high background count rates as a result of photoluminescence, chemiluminescence, and static electricity. In photoluminescence (also called phosphorescence), photons are generated by interactions of the ultra-violet component of light with the sample vial and contents. Therefore, samples should avoid exposure to direct sunlight and fluorescent light, and counting solutions should be stored in amber containers. Incandescent light will not cause photoluminescence. The level and duration of photoluminescence is a function of the light intensity and exposure time. When a sample has been photoactivated, it must be dark adapted until it decays to background levels.

In chemiluminescence, photons are generated during sample preparation as a result of chemical interactions of the sample components. The amount and duration is temperature dependent and the effect decays faster at higher temperatures. However, cooling the sample will slow down the effect to a point where the coincidence circuitry of the counter can discriminate between chemiluminescence and radioactive decay. Thus, storing samples in a refrigerator overnight should remove most of the background counts due to chemiluminescence and photoluminescence.

The usual method for detection of luminescence is to recount the sample after an appropriate interval. A decrease in the second count rate indicates a strong possibility of luminescence.

During dry seasons or when using an ambient counter, static electricity may be the cause of high background count rates. When a static charge that is deposited on a vial discharges, light photons are produced in proportion to the charge. If the vial is being counted at the time of discharge, an incorrect, high sample count rate will result. This problem can be reduced by humidifying the counter and by wiping the vials with a moist cloth or dryer static sheet.

D. Liquid Scintillation Counting - Detailed

Liquid scintillation counting is a technique that detects and measures the radioactivity in a sample. The radioactive sample is mixed with a solvent and fluor. The radiation emitted by the radionuclide produces ionization or excitation of the molecules of the surrounding fluid. Certain molecules contained in the fluid fluoresce when ionized or excited. The Liquid Scintillation Counter (LSC) is used to detect these flashes of light. The number of photons emitted by the fluid is proportional to the energy deposited in the fluid by the emitted particle.

Liquid scintillation counting is used primarily to detect low energy beta particles emitted by radionuclides incorporated in biological samples. Beta particles are emitted with energies ranging from zero to a maximum that is characteristic of the isotope (see Figure 5). The beta particle shares the maximum energy with the neutrino, hence one has a range of beta particle energies. The average energy of the beta particles is approximately one-third the maximum energy.



Figure 5 ³²P Beta Particle Energy Spectrum

1. <u>Physical Interactions</u>

Several interactions occur in the fluid as a result of the radioactive emission of particles. First,

Beta⁻ + solvent(1) $\rightarrow \rightarrow \rightarrow \rightarrow \ast$ solvent(1) {*denotes excited state}

The beta particle interacts with the solvent molecules and changes the molecules into ions, excited molecules, free radicals, or secondary particles. Excited solvent molecules are photoluminescent, that is, they are excited by radiation and re-emit radiation at the same or a different wavelength. Fluorescent emission lifetimes are in the order of nanoseconds, while, phosphorescence emissions have a much longer lifetime. It is fluorescent emissions that produce light photons detected in the LSC. Excited molecules can fluoresce or transfer energy to another molecule by resonance and diffusion-collision processes. The following interaction is a solvent molecule to solvent molecule transfer of energy:

*solvent(1) + solvent(2) $\rightarrow \rightarrow \rightarrow \rightarrow$ solvent(1) + *solvent(2)

The energy associated with the excited state of a solvent molecule can be quickly lost to the fluid by collision with other solvent molecules. There are usually 10⁴ to 10⁵ times as many solvent molecules as fluor molecules in a scintillation fluid thereby increasing the probability of this interaction. Diluting the fluid with molecules that do not participate in the process will reduce the efficiency of solvent-solvent interactions (dilution quenching). Usually, solvent-solvent interactions occur in less that 10⁻⁹ seconds. The energy transfer between an excited solvent molecule and fluor molecule is quantitative and non-radioactive.

```
*solvent(2) + fluor(1) \rightarrow \rightarrow \rightarrow \rightarrow solvent(2) + *fluor(1)
```

The excited solvent molecule does not emit photons that are absorbed by the fluor molecule. For this interaction to occur, the energy level of the solvent molecule must correspond to a discrete vibrational energy level of the fluor. The purpose of a fluor or solute molecule in a scintillation fluid is to accept the energy from the excited solvent molecule and emit a photon when returning to its ground state. When there is only one type of fluor, the fluor molecule to fluor molecule energy transfers are monoenergectic and reversible.

*fluor (1) + fluor(2) $\rightarrow \rightarrow \rightarrow \rightarrow$ fluor(1) + *fluor(2)

Primary fluors emit photons at a characteristic wavelength, whereas second fluors introduced into the fluid absorb the primary photons and emit secondary photons at a wavelength to which the phototubes are most sensitive. The number of photons emitted is related to the energy of the emitted particle.

2. <u>Solvent and Fluor Characteristics</u>

A good solvent will exhibit the following characteristics:

- a. Presence of π electrons about the molecule; π electrons are required for excitation to occur.
- b. Excited states with a long lifetime.
- c. Chemically compatible with the fluor and sample.
- d. Small absorption coefficient for light emitted by the fluor.

Typical solvents are aromatic hydrocarbons and include xylene, toluene, benzene, and dioxane. Another characteristic of solvent is quantum yield which is, the ratio of the number of photons emitted to the number of excited molecules. Xylene and toluene have good quantum yields.

Most fluors are oxazole and oxadiazole compounds. The most important fluor characteristics are:

- a. Emission of a photon of long wavelength, matching phototube photocathode sensitivity.
- b. A very short excited state lifetime.
- c. High solubility in the solvent and sample.
- d. High quantum yield.

The efficiency of energy transfer between an excited solvent molecule and fluor molecule is dependent on the relative concentrations. Figure 6 is a plot of the effect of fluor concentration versus photon yield which is a direct measure of energy transfer efficiency. Self-absorption, absorption of fluor emitted photons by fluor molecules, occurs at high fluor concentrations.



Some commonly used fluors are PPO (2,5-diphenyloxazole) and Butyl PBD [2-(4-t-Butylphenyl)-5-(4-biphenylyl)-1,3,4-oxadiazole]. Secondary fluors which are used for their longer wavelength photon include, POPOP [1,4-Bis-2-(5-phenyloxazolyl)-benzene], dimethyl POPOP [1,4-Bis-2(4-methyl-5-phenyloxazolyl)-benzene], and Bis-MSB [p-Bis(0-methyl-styryl)-benzene].

3. <u>Liquid Scintillation Counter</u>

Liquid scintillation samples are contained in small vials (glass or plastic) containing scintillation fluid. In the light tight counting well of the counter, photons emitted by the sample are reflected into the photomultiplier tubes (PMTs). When photons strike the photocathode (antimony-cesium alloy) of the PMT, photoelectrons are emitted. The photoelectrons are accelerated and focused into a dynode series and amplified ($x10^6$). The anode collects the electrons from the last dynode and produces a voltage pulse proportional to the electrons collected. Due to the proportionality between the energy of the radioactive particle, the number of excited molecules, the number of photons emitted by fluor molecules and the number of photoelectrons, the output pulse of the PMT is proportional to the energy of the beta particle.

Background or noise pulses are also produced by the PMTs and a coincidence circuit is used to reduce these pulses to acceptable limits. When pulses from the two PMTs occur within 20 to 40 nanoseconds of one another, the coincidence circuit produces a gate pulse. This pulse allows the pulse from the Pulse Height Analyzer (PHA) to pass through the coincidence gate and on to the scalers. Noise and background pulses are random and do not lead to production of gate pulses.

The output pulses from the PMTs are added in either a linear or logarithmic manner by the summing amplifier. A log amplifier produces a pulse with an amplitude equal to the log of the summed PMT pulses. Linear amplification generates a pulse linearly proportional to the summed PMT pulses.

After amplification, the pulse is analyzed by the PHA. The PHA has two limits, upper and lower. A set of upper and lower limits (discriminators) is called a window or channel. Only pulses with an amplification greater than the lower limit but less than the upper limit are passed. In this manner the LSC can differentiate pulses from different energy radiations. Two isotopes in the same sample can be counted simultaneously by discriminating pulses from the more energetic beta particles from the pulses from the less energetic beta particles.

When the PHA passes a pulse and a coincidence pulse arrives at the coincidence gate simultaneously, a pulse is sent to the scaler and registered. Most LSC's contain three channels for sample analysis and two channels for quench correction. The components of a LSC are shown in Figure 7 and Figure 8.

4. <u>Liquid Scintillation Counter Features</u>

Most liquid scintillation counters have five functional components:

- a) Sample transport system (STS)
- b) Electrical detection system
- c) Display system
- d) Counting program system
- e) Output system

The STS can be either a chain of vial holders or a tray which positions the vial over the counting well and also allows for sample identification. The STS is controlled either manually or automatically. The vial is lowered via elevator into the counting well and analyzed by the electrical detection system (PMTs, PHA, etc.). Some LSC's are refrigerated to help reduce thermal noise and chemiluminescence.

The display system presents visual information (the counts in the individual channel scalers, counting time and sample position number) while the sample is being counted. Some LSC's also display the sample count rate.

Counting programs allow the LSC to operate under various parameters such as type of isotope, channel settings, preset error requirements, counting time and number of counting cycles. Different instrument users can program the LSC for counting of their samples without interrupting the counting of other samples.

Output devices include: teletype writers, mechanical printers, magnetic tape cassette or conventional typewriters.







General schematic of a LSC with log amplification. Signals from the PMTs are monitored for coincidence and are also summed. When the coincidence requirement is met, the coincidence gate is opened to allow the same signal to be stored in scalers.

5. <u>Quenching and Quench Correction</u>

Any component in the vial which interferes with the basic interactions of the scintillation process produces a quenching effect. Quenching is a reduction in the total photon output of a sample which results from a reduction in energy transfer efficiency. Chemiluminescence is the process of producing an excited electronic state by the energy from a chemical reaction. It is one of several mechanisms by which molecules dissipate energy produced by a chemical reaction. In chemiluminescence, light emission does not involve, but is quenched by, oxygen. The intensity and duration of chemiluminescence are temperature dependent. Chemiluminescence decays faster (2 to 3 hours) at lower temperatures than at ambient temperature.

There are at least four basic types of quench; impurity, color, dilution, and absorption. Impurity quenching is caused by components in the vial that compete with fluor molecules for energy or react chemically with the fluor molecules to make them less reactive to energy transfer. Impurity quenchers include strong inorganic acids, oxidizing agents, and some organic compounds. Color quenching occurs when the quenching component absorbs photons emitted in the scintillation process before the photons can be detected by the PMTs. Colored samples such as chlorophyll and hemoglobin are quenching agents. In dilution quenching there is an insufficient number of fluor molecules in the scintillation fluid. This can be a result of using too large a sample which causes the final concentration of fluor molecules to fall below the optimum conditions. Beta absorption occurs in samples which are not in intimate, stable and physical contact with the scintillation fluid. Samples imbedded on support media (filter paper) and heterogeneous samples will exhibit beta absorption to a large degree.

Other examples of quenching are optical quenching (separation of scintillation fluid and sample into two liquid phases and fogging) and oxygen quenching (dissolved oxygen in scintillation fluid). The general effect of quenching is a reduction in the photon output of a sample and is shown in Figure 9.



6. <u>Counting Efficiency Determination</u>

To compare samples with various degrees of quench, sample activities (dpm) should be used. To determine the sample activity, the counting efficiency must be determined.

Counting efficiency = $\frac{sample cpm - background x100\%}{Sample dpm}$

Several methods are available for determining counting efficiency, they include: Internal Standard, Sample Channel Ratio (SCR), External Standard, and External Standard Channel Ratio (ESCR).

a) Internal Standard:

In this method the sample is first counted and the count rate determined. An amount of known radioactivity (a standard) is then added to the sample and the sample counted again. The counting efficiency can be determined using:

$$Efficiency = \frac{[sample with std.cpm - sample without std.cpm]}{std activity (dpm)}$$

NOTE: The counting efficiency of the standard is assumed to be the same as the counting efficiency of the sample.

Some advantages of the Internal Standard method are:

- A fast procedure for small numbers of samples.
- No quench curve is required.

Disadvantages of the Internal Standard method include:

- Sample must be opened and the standard introduced.
- Prone to errors in pipetting.
- Sample is altered by standard.
- Addition of standard may quench the sample.
- The standard should have the same chemical characteristics as the sample.
- b) Sample Channels Ratio:

The liquid scintillation spectrum is divided into two channels and a ratio of the counting rate for the two channels is expressed in the Sample Channels Ratio method. Quenching will cause the entire spectrum to shift to lower energies and thus change the ratio of the two counting channels. See Figure 9. for an illustration of this effect. A quench curve must be constructed using standards with the same or similar types of quenching agents as those present in the sample. A quench curve is a curve relating the counting efficiency of the standards to the indicator of quenching (i.e., SCR).

To use the sample channels ratio method:

- Prepare samples and standards. The standard should contain the same radionuclide as the samples.
- Count the standards using the same instrument settings as the samples. Use two channels to divide the spectrum.
- Plot standard counting efficiency versus SCR.
- Take SCR value for each sample and read the counting efficiency from the quench curve.
- Determine sample activity using:

Sample DPM = $\frac{sample \ count \ rate(cpm)}{counting \ efficiency}$

c) External Standard:

This method utilizes an external gamma ray source to produce Compton electrons in the vial, sample material, and scintillation fluid. Compton electrons will behave as beta particles and are counted as such. Quenching will affect the Compton electron spectrum and the beta spectrum in the same manner. As quenching increases, the Compton spectrum will decrease in total count rate. The amount of quenching is then expressed by the total gross count rate of the standard. A disadvantage of this method is the sample volume and vial position dependency of the external standard count.

d) External Standard Channels Ratio:

The ESCR method was developed to overcome the sample volume and vial position dependencies of the ES method. Two channels are used to monitor the Compton electron spectrum. Geometry and volume effects will alter the distribution of the channel counts and change the ratio of the two counting channels. To use this method, take the following steps:

- Prepare samples and standards just as in the SCR method.
- Count the standards using the same instrument settings as the samples. Use two channels to divide the Compton Electron spectrum.
- Plot the standard counting efficiency versus ESCR.
- Take the ESCR value for each sample and read the counting efficiency from the quench curve.
- Determine sample activity using:

$$Sample DPM = \frac{sample count rate (cpm)}{counting efficiency}$$

Figure 10. is an example of a quench curve for use in the ESCR method.



<u>Figure 10</u> A typical quench curve for tritium counting using the ESCR method.

7. <u>Dual Label Counting</u>

When two or more isotopes are present in a sample, both will produce detectable photons. The more energetic beta particles will produce pulses larger than less energetic beta particles. An example is a sample containing Carbon-14 and Hydrogen-3 (tritium). If there is a sufficient energy difference between the beta particle energies, in this case, the difference between 156 kev and 18 kev, a portion of the beta spectrum from the higher energy isotope can be counted without a contribution of counts from the other isotope. Figure 11 shows that as quenching increases, the number of counts from Carbon-14 registered in the tritium channel increases while the number of counts from tritium in the tritium channel decreases.



In dual label counting, two counting channels are required: for example, one counts the higher energy portion of the Carbon-14 spectrum and the other counts the lower energy spectrum. The lower energy portion of the spectrum is made up of counts from the lower energy isotope (³H) as well as a fraction from the higher energy isotope (¹⁴C). Three quench curves must be produced in order to make quench corrections using the ESCR method.

ESCR is plotted as a function of counting efficiency for:

- 1. ${}^{14}C$ in the ${}^{3}H$ channel.
- 2. ${}^{14}C$ in the ${}^{14}C$ channel.
- 3. 3 H in the 3 H channel.

To determine the activities of ${}^{3}H$ and ${}^{14}C$ in the sample, use the following equations:

CPM (¹⁴C channel) = DPM (¹⁴C) x Efficiency (¹⁴C in the ¹⁴C channel)

CPM (³H channel) = DPM (³H) x Efficiency (³H in the ³H channel +

DPM (14 C) x Efficiency (14 C in the 3 H channel)

VI. GAMMA SCINTILLATION COUNTING

The basic processes involved in scintillation counting are:

- 1. The absorption of radiation within the scintillator resulting in the excitation and ionization of atoms or molecules.
- 2. Photons are emitted when excited atoms or molecules return to their round state.
- 3. Photons are absorbed by the photocathode of the photomultiplier tube PMT) and result in the emission of photoelectrons.
- 4. The number of photoelectrons is amplified by the dynode series of the PMT. The photoelectrons are collected at the anode and produce a voltage pulse.
- 5. The pulse produced at the anode, which is proportional to the energy transferred to the scintillator, is amplified, analyzed and counted by a scaler.

To understand gamma scintillation counting, one must first investigate the manner in which gamma rays interact with matter, primarily solids and liquids.

A. Gamma Ray Interactions With Matter

Gamma rays interact with matter in several ways. The most important of these are:

1. <u>Photoelectric Effect</u>

In the photoelectric effect, the gamma ray interacts with an orbital electron, transferring all its energy to the electron and disappearing in the process. The electron is ejected with a kinetic energy equal to $hv - E_b$, where E_b is the binding energy of the orbital electron and ht is the energy of the gamma photon. The photoelectron will then undergo interactions with atoms in its path, transferring energy to the atoms until it comes to rest. The photoelectric effect is the predominate interaction at low gamma ray energies (less than 0.5 MeV).

2. <u>Compton Effect</u>

Gamma rays of medium energy (0.5 to 1.5 MeV) can undergo elastic collisions with orbital electrons. The gamma ray will transfer part of its energy to the electron and proceed in a new direction, at a lower energy (longer wavelength). The angular dependence of this type of collision with an electron is shown by:

$$h\nu' = \frac{h\nu}{1 + (1 + -\cos\theta)(hlm_o c^{2})}$$

Where hv' is the energy of the scattered photon, and θ is the angle between its direction and that of the primary photon. It is the energy of the primary photon and m_oc^2 is the rest mass energy of the electron expressed in MeV. Compton electrons make up a wide energy spectrum resulting from interactions of monoenergetic gamma radiation with matter. If an inner orbital electron is ionized, x-ray and Auger electron emission will occur.

3. <u>Pair Production</u>

When a gamma ray interacts with the nuclear force field, the photon can cease to exist and have all its energy converted into a positron and an electron. The gamma ray energy must be greater than 1.022 MeV (2 times the rest mass energy of an electron) and the excess energy ($h_v - 1.022$ MeV) is converted to kinetic energy of the positron and electron. After slowing down, the positron combines with a nearby electron and both are annihilated, producing two 0.511 MeV photons which can then participate in Compton and Photoelectric interactions.

B. Gamma Scintillation Detector

The operation of a gamma scintillation detector can be summarized as follows. Radiation produces a flash of light in the scintillator which is optically coupled to the PMT. The light photons strike the photocathode of the PMT and release photoelectrons. The photoelectrons are multiplied by factors of 10^5 to 10^7 and produce a voltage pulse at the anode.

The absorption of energy by a substance and its reemission as visible or near visible radiation is known as luminescence. The luminescence process in organic crystal (anthracene, trans-stilbene) scintillators is a molecular process. Radiation can raise molecules from the electronic ground state to an electronic excited state. The excited molecule can return to its ground state by emission of a photon or by dissipating the excess energy as heat in the surrounding media. The excited state lifetime is in the order of 10 nanoseconds. The scintillation process in inorganic crystal [(NaI(Tl), LiI(Eu), CsI(Tl)] scintillators can best be described in terms of the band theory of solids, as shown in Figure 12. Ionization occurs when radiation of sufficient energy causes electrons to move from their lattice sites into the conduction band. The electron vacancy in the valence band is referred to as a hole. The electron in the conduction band and the hole in the valence band move about independently in the crystal. Radiation can also cause excitation whereby an electron is moved into the exciton band. The electron-hole pair is now called an exciton and moves about the crystal together. The excitons, holes, and free electrons wander about the crystal until they are trapped at activator sites (impurities). By exciton capture or by the successive capture of an electron and hole, the activator site is raised to an excited state. The transition from the excited state to the ground state

releases a photon. The impurities used most frequently are Thallium (in NaI and CsI) and Europium (in LiI).





Crystal scintillators are manufactured as solid cylinders or flat disks. The scintillator is usually encased in aluminum for mechanical protection and control of ambient light. NaI(Tl) is hygroscopic and must be hermetically sealed. A glass or quartz window is used to allow light to escape the scintillator. Sometimes a light pipe is used between the scintillator and the PMT, which helps prevent light trapping in the scintillator and enables the light to be spread over a large photocathode surface. There must be good optical coupling between the scintillator and the PMT in order to reduce the amount of reflection at the interfaces.

Crystal scintillators should have the following characteristics:

- 1. High-energy conversion efficiency (energy of radiation to fluorescent radiation) in the scintillation process. Inorganic crystals are the most dense and are better gamma ray absorbers than organic scintillators.
- 2. Transparency to its own fluorescent radiation.
- 3. Excited states with short lifetimes.
- 4. Photon spectrum matching PMT photocathode sensitivity spectrum.

A photomultiplier tube consists of a photocathode (antimony-cesium) for releasing photoelectrons, a dynode series for producing electron amplification and an anode for collecting the current pulse. The photoelectrons emitted from the photocathode are focused and strike the first dynode. Secondary emission occurs, increasing the number of electrons which will strike the second dynode. This process of secondary emission occurs until the electrons are collected at the anode. The dynodes operate at increasing voltages and as the applied voltage is increased, the electron amplification also increases which

increases the voltage pulse on the anode. The output pulse on the anode is proportional to the number of photoelectrons originally released by the photocathode which is proportional to the number of photons produced in the scintillator. The number of photons produced in the scintillator is proportional to the energy deposited in the scintillator by the gamma photon.

C. Gamma Scintillation Spectrometer

One important characteristic of a gamma scintillation detector is its ability to give an indication of the energy of the incident gamma ray. Gamma scintillation spectrometry involves analysis of the output of a scintillation detector to determine the energies and intensities of incident gamma ray photons. Figure 13 is a block diagram of a gamma scintillation spectrometer.





The high voltage power supply provides the potential that accelerates electrons through the dynode series. A preamplifier, located close to the detector, is used to couple the detector to the amplifier and provide some amplification of the detector output pulse.

The linear amplifier supplies amplification and pulse shaping. The pulse shaping function permits the retention of information such as the time of occurrence and amplitude of the detector output. Pulse shaping also prevents overlooking the high gain stages of the amplifier and allows each shaped pulse to start from the same baseline.

The amplified and shaped pulses are sent to the pulse height analyzer (PHA) where they are sorted in terms of their amplitude. A component of the PHA is the discriminator circuit which accepts all pulses above a certain pulse amplitude and rejects those below this pulse amplitude. When the input pulse exceeds a preset discriminator level, called a threshold, the discriminator will generate an output logic pulse which is counted by the scaler. If the input pulse does not exceed the threshold, output logic pulse is produced. By varying the threshold setting, we can obtain an integral count of all pulses with an

amplitude greater than the threshold setting. Figure 14 shows an integral counting curve or spectrum.



Figure 14 Integral and Differential Spectrum from NaI(Tl) gamma scintillation spectrometer.

To obtain information about the energy of the incident gamma photons, a differential pulse height spectrum must be produced. The number of pulses within a specified amplitude (energy) range is determined with the use of a pair of discriminators, an upper level and a lower level (threshold). Pulses that have an amplitude larger than the threshold setting but less than the upper level discriminator setting fall in the window and will trigger the discriminator to produce an output logic pulse. If the pulse height is less than the threshold setting or greater than the upper level discriminator setting, no output logic pulse is produced. In this manner, when the input pulses have amplitudes or energies that fall within the window settings, an output logic pulse is produced and sent to the scaler. Figure 14. shows a typical differential pulse height spectrum. Each point on the curve represents the number of pulses of a particular amplitude (energy) received by the counting system in a specified amount of time. The points also correspond to a discrete amount of energy lost to the scintillation detector by the incident gamma photons. This type of spectrum allows us to determine the energy spectrum of a particular gamma ray source and to identify peaks corresponding to principle gamma ray energies.

D. Spectrum Analysis

The general form of gamma ray spectra is shown in Figure 15.



The total absorption peak or photopeak is due to those gamma photons which lose all their energy via the three main interactions in the scintillator. The degree to which the photopeak is broadened is a function of the energy resolution of the detector as well as variations in photon production, collection efficiency, photoelectron production and electron amplification processes.

The Compton distribution results from detector pulses following Compton interactions in which the scattered photons escape the scintillator and represents pulse heights that correspond to energies from zero up to the maximum energy available to an electron in a Compton interaction.

Other peaks that may be found in gamma ray spectra include:

- 1. Backscatter Peak
- 2. Annihilation Peak
- 3. Sum Peaks

A backscatter peak will appear in a gamma ray spectrum as a result of gamma ray (Compton) interactions with material (shielding, etc.) surrounding the detector. The maximum energy of a gamma ray backscattered 180° is 255 keV, regardless of the original energy.

An annihilation peak at 511 keV can be a result of the annihilation of a positron emitted by a gamma source. The positron will usually be stopped and annihilated in the aluminum housing of the scintillator. When the positron is annihilated, two 511 keV photons are produced and travel 180° from each other; thus if one enters the scintillator, the other will escape. If a high energy (>1.022 MeV) gamma ray undergoes a pair production interaction with the detector shielding material, one of the annihilation photons may enter the scintillator and deposit all its energy.

A sum peak is found in the spectrum of a gamma ray source that emits two or more gamma rays. The peak is equal to the sum of energies of the two or more gamma rays, provided that they are emitted simultaneously and enter the scintillator simultaneously.

E. Detector Resolution

The full width at half maximum or resolution of a scintillation detector is defined as the photopeak at one-half its maximum value, divided by the pulse height at the center of the photopeak. Resolution is usually measured at the photopeak from ¹³⁷Cs (662 keV). The observed resolution of a spectrometer system is a function of the effective resolution of the scintillator, the PMT, and the associated electronics.