

Radiochemistry

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Credits

Figures were used with permission from the following sources.

Part II. Chart of the Nuclides, General Electric Co.

Part IV. Fig. 5 from N.F. Johnson, E. Eichler and G.D. O'Kelley, "Nuclear Chemistry", Interscience Publishers, New York, 1963, p. 142.

Fig. 15 from N.F. Johnson, et al., p. 100

Part V. Fig. 1 from F. Nelson, T. Nurase and K. Kraus, J. Chromat., 13, 503 (1964)

Fig. 2 from K. Kraus and F. Nelson, Proc. of the Inter. Conf. on Peaceful Uses of Atom. Ener., (Geneva; United Nations), Vol. 7 (1956), Paper 837.

Part VII Fig. 2 from N.F. Johnson, et al, p. 159

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Fig. 9 " " " " " 25

Fig. 20 from W.H. Zoller and G.E. Gordon, Anal. Chem, 42, 261 (1970).

Part VIII Fig. 3 from R.B. Leachman, "Nuclear Fission", Scientific American, 1965.

Part IX Fig. 2 from General Atomic Co.

Part XI Fig. 1. from C.H. Wang, D.L. Willis and W.D. Loveland, "Radiotracer Methodology in the Biological, Environmental and Physical Sciences", Prentice-Hall Co., 1975, p. 195.

Fig. 4,5,6,8 from Technical Data in catalogs of Packard Instr. Co.

Most of the other figures were used or adapted from the author's (GRC) texts "Experimental Nuclear Chemistry" (out of print) and "Nuclei and Radioactivity".

COURSE OUTLINE

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A WORD TO THE USER

ACS AUDIO COURSES are instructional units on subjects of significant interest that are suitable for both individual study and group use.

The ACS Audio Course on "Radiochemistry," by Drs. Gregory Choppin and Patricia Baisden, is intended as an in-depth introduction to this important subject. This volume is the reference manual which is integrated with the audiotape of "Radiochemistry." The two together comprise the ACS Audio Course: neither is complete without the other. Because the lecturer refers constantly to the manual, each listener should have a copy of the manual as he listens, so he may follow these references.

This manual was not written to be a self-sufficient textbook. It was designed, rather, to be used as a workbook while listening to the tape. The listener is urged to take notes with the expectation that this activity will reinforce the learning process. The product is a personally annotated volume which should serve as an authoritative and up-to-date introduction to the subject.

Your comments and suggestions have proved to be exceptionally valuable guides for improving our educational programs. We hope you will continue to send them to us.

Department of Educational Activities
American Chemical Society

PART I

General Nuclear Concepts

Outline

- I. Definitions of Some Basic Nuclear Terms
- II. Nuclear Mass-Energy Relationships
 - A. Balancing Nuclear Reactions
 - B. Energy Conversion Factors
 - C. Comparison of Energy in Chemical and Nuclear Reactions
 - D. Calculation of Energy in Nuclear Reactions
 - E. Nuclear Binding Energy
 - 1. Binding Energy of Last Nucleon
 - 2. Binding Energy of a Deuteron
 - 3. Binding Energy per Nucleon
- III. Nuclear Radius
- IV. Nuclear Coulomb Barrier.
- V. Nuclear Models
 - A. The Shell Model
 - B. Liquid Drop Model
 - C. Unified Model
- VI. Problems

SECTION I

Some Basic Nuclear Terms

1. Nuclide: any nuclear species of a given number of protons and neutrons.

Nucleon: either neutron or proton.

Mass Number = $A \equiv$ number of nucleons = $N+Z$

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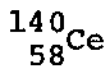
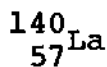
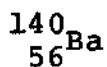
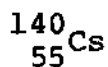
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Atomic Number = $Z \equiv$ number of protons

2. Isotopes: same Z ; different A



3. Isobars: same A ; different Z



4. Isotones: different A ; same N



5. Atomic Mass Unit (amu): 1 amu is $1/12$ the mass of ${}^{12}_6\text{C}$.

6. Masses of atomic particles

*

$$M_p = 1.00783 \text{ amu} = \text{mass of the proton} \quad \text{Mass of proton} = 1.00728$$

$$M_n = 1.00867 \text{ amu} = \text{mass of the neutron}$$

$$M_e = 0.00054860 \text{ amu} = \text{mass of the electron}$$

$$M_H = 1.00783 \text{ amu} = \text{mass of the hydrogen atom}$$

7. Units of radioactivity

$$\text{curie (ci)} = 2.22 \times 10^{12} \text{ dpm (disintegrations/min)}$$

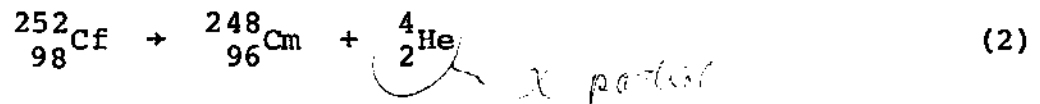
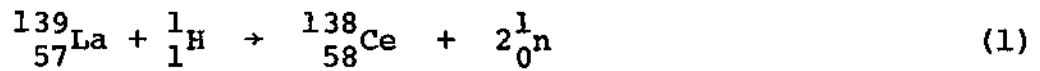
$$\text{millicurie (mci)} = 10^{-3} \text{ ci} \quad (\text{Defined: } 3.7 \times 10^{10} \text{ d/s})$$

$$\text{microcurie (}\mu\text{ci)} = 10^{-6} \text{ ci}$$

SECTION II

Nuclear Mass-Energy Relationships

A. Balancing Nuclear Equations



B. Conversion Factors

TABLE 1. Commonly used metric units: Systems

<u>Physical Quantity</u>	<u>mks</u>	<u>cgs</u>
Force (F)	newton ^N (nt)	dyne
Mass (m)	kilogram (Kg)	gram (g)
Length (d)	meter (m)	centimeter (cm)
Time (t)	seconds (sec)	seconds (sec)
Acceleration (a)	$\frac{\text{meters}}{\text{sec}^2}$	$\frac{\text{centimeters}}{\text{sec}^2}$
Work (W)	joule (J)	erg

From $F = m \times a$

$$\frac{N}{nt} \equiv \frac{Kg \cdot m}{sec^2} \quad \text{dyne} = \frac{g \cdot cm}{sec^2}$$

$$\frac{N}{nt} \equiv 10^5 \text{ dynes}$$

From $W = F \times d$

$$J \equiv \frac{N}{nt} \cdot m \equiv \frac{Kg m^2}{sec^2} \quad \text{erg} \equiv \text{dyne} \cdot \text{cm} \equiv \frac{g \cdot cm^2}{sec^2}$$

$$J \equiv 10^7 \text{ ergs}$$

1. Important energy conversion factors and definitions

NOTE: Energy and work are interchangeable

Electron Volt (eV): the energy acquired when an electron is accelerated through a potential difference of 1 volt.

Calorie (cal): the quantity of heat which must be added to 1 gram of water (1 atmosphere pressure) to change its temperature from 14.5 to 15.5°C.

1 cal = 4.18 J

$$\begin{aligned}
 1 \text{ eV} &= 1.602 \times 10^{-19} \text{ J} \\
 &= 1.602 \times 10^{-12} \text{ ergs} \\
 &= 3.829 \times 10^{-26} \text{ cal} \\
 &= 1.074 \times 10^{-9} \text{ amu}
 \end{aligned}$$

$$10^9 \text{ eV} = \text{GeV} \qquad 10^6 \text{ eV} = \text{MeV}$$

a. An important energy conversion factor is

$$1 \frac{\text{eV}}{\text{molecule}} = 23.06 \frac{\text{Kcal}}{\text{mole}} \qquad (4)$$

This is obtained by

$$\begin{aligned}
 1 \frac{\text{eV}}{\text{molecule}} &= \frac{3.829 \times 10^{-20} \text{ cal}}{\text{molecule}} \times \frac{\text{Kcal}}{10^3 \text{ cal}} \\
 &\quad \times \frac{6.023 \times 10^{23} \text{ molecules}}{\text{mole}}
 \end{aligned}$$

where $6.023 \times 10^{23} \frac{\text{molecules}}{\text{mole}}$ is Avogadro's number

b. Another frequently used conversion factor is

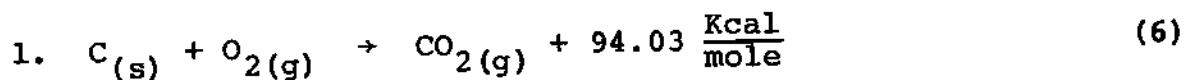
$$1 \text{ amu} = 931 \text{ MeV} \qquad (5)$$

This is obtained by

$$\begin{aligned}
 1 \text{ amu} &= \frac{1 \text{ eV}}{1.074 \times 10^{-9}} \times \frac{\text{MeV}}{10^6 \text{ eV}} \\
 &= 931 \text{ MeV}
 \end{aligned}$$

Handwritten notes:
 $E = mc^2$
 $1 \text{ amu} = 1.66 \times 10^{-27} \text{ kg}$
 $c = 3 \times 10^8 \text{ m/s}$
 $E = (1.66 \times 10^{-27}) (3 \times 10^8)^2$
 $E = 1.49 \times 10^{-10} \text{ J}$
 $1 \text{ eV} = 1.6 \times 10^{-19} \text{ J}$
 $\therefore \frac{1.49 \times 10^{-10}}{1.6 \times 10^{-19}} = 9.31 \times 10^8 \text{ eV}$
 $= 931 \text{ MeV}$

C. Comparison of Energy in Chemical and Nuclear Reactions



Using $E = \Delta M \cdot c^2$ and solving for the mass

$$\Delta M = \frac{E}{c^2} = \frac{94.30 \frac{\text{Kcal}}{\text{mole}}}{(3.0 \times 10^{10} \frac{\text{cm}}{\text{sec}})^2} \quad \text{where } c = 3.0 \times 10^{10} \text{ cm/sec}$$

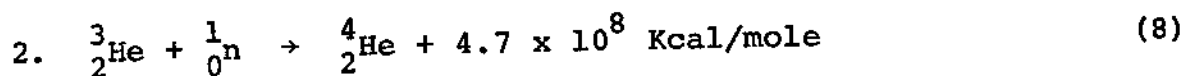
$$= 94.03 \text{ Kcal/mole} \times \frac{1 \text{ ev/molecule}}{23.06 \text{ Kcal/mole}} \times 1.602 \times 10^{-12} \text{ erg/ev}$$

$$\times 6.023 \times 10^{23} \frac{\text{molecules}}{\text{mole}} \times 9.0 \times 10^{20} \frac{\text{cm}^2}{\text{sec}^2}$$

Since $\text{erg} = \text{g cm}^2/\text{sec}^2$

$$= 4.37 \times 10^{-9} \left[\frac{\text{g} \cdot \text{cm}^2/\text{sec}^2}{\text{cm}^2/\text{sec}^2} \right] / \text{mole}$$

$$= 4.37 \times 10^{-9} \text{ g } (10^{-3} \% \text{ per mole of reactant mass}) \quad (7)$$



$$\Delta M = \frac{E}{c^2} = \frac{4.7 \times 10^8 \frac{\text{Kcal}}{\text{mole}}}{c^2} = 2.2 \times 10^{-2} \text{ g } \sim 0.5 \% \text{ of reactants mass} \quad (9)$$

D. Calculation of Energy (Q) in Nuclear Reactions

Convention used:

$$Q = -\Delta E$$

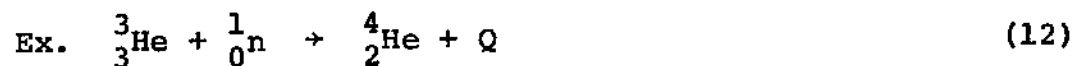
$$\Delta M = [\text{Sum of atomic masses of reactants}] - [\text{Sum of atomic masses of products}] \quad (10)$$

Note: Atomic masses (amu) = mass of nucleus + electrons

$$Q = \Delta M(\text{amu}) \times 931 \text{ (MeV/amu)} = \text{MeV} \quad (11)$$

Positive Q \equiv exoergic (occurs spontaneously)

Negative Q \equiv endoergic (requires energy to occur)



$${}^3_2\text{He} = 3.0160 \text{ amu}$$

$${}^1_0\text{n} = 1.0087 \text{ amu}$$

$${}^4_2\text{He} = 4.0026 \text{ amu}$$

$$\Delta M = [3.0160 \text{ amu} + 1.0087 \text{ amu}] - [4.0026 \text{ amu}]$$

$$= 0.221 \text{ amu}$$

$$\begin{aligned} Q &= 0.0221 \text{ amu} \times 931 \text{ MeV/amu} \\ &= 20.58 \text{ MeV} \end{aligned}$$

E. Nuclear Binding Energy

Binding Energy (BE): the energy released in the formation of a nucleus from the appropriate numbers of neutrons and hydrogen atoms

Ex: Calculate the BE of ${}^4_2\text{He}$



$${}^1_1\text{H} = 1.0078 \text{ amu}$$

$$\Delta M = 2[1.0078 + 1.0087] - 4.0026$$

$$= 0.03040 \text{ amu}$$

$$\text{BE} = 931 \text{ MeV/amu} \times 0.0304 \text{ amu}$$

$$= 28.30 \text{ MeV} \quad (14)$$

Binding energy per nucleon = Be/A

$$\text{BE}/A = \frac{28.30}{4} = 7.08 \text{ MeV/nucleon} \quad (15)$$

*After allowed to be
separated in 1949*

1. (Binding energy of last nucleon.

Ex. Calculate BE of last neutron and of last proton
in $^{235}_{92}\text{U}$.

Last neutron:

$$^{234}_{92}\text{U} + {}^1_0\text{n} = ^{235}_{92}\text{U} + Q \quad (16)$$

$$Q = M_{^{234}\text{U}} + M_{\text{n}} - M_{^{235}\text{U}} \times 931 \text{ MeV/amu}$$

$$= [234.04090 + 1.00867 - 235.04392] \text{amu} \times 931 \text{ MeV/amu}$$

$$\text{BE last neutron} = 5.26 \text{ MeV}$$

Last proton:

$$^{234}_{91}\text{Pa} + {}^1_1\text{H} = ^{235}_{92}\text{U} + Q \quad (17)$$

$$Q = [M_{^{234}\text{Pa}} + M_{^1_1\text{H}} - M_{^{235}\text{U}}] \times 931 \text{ MeV/amu}$$

$$= [234.04330 + 1.00783 - 235.04392] \text{amu} \times 93 \text{ MeV/amu}$$

$$\text{B.E. last proton} = 6.71 \text{ MeV.}$$

2. BE of a deuteron



BE = 2.2 MeV
BE/nucleon = 1.1 MeV

3. Binding energy per nucleon, BE/A

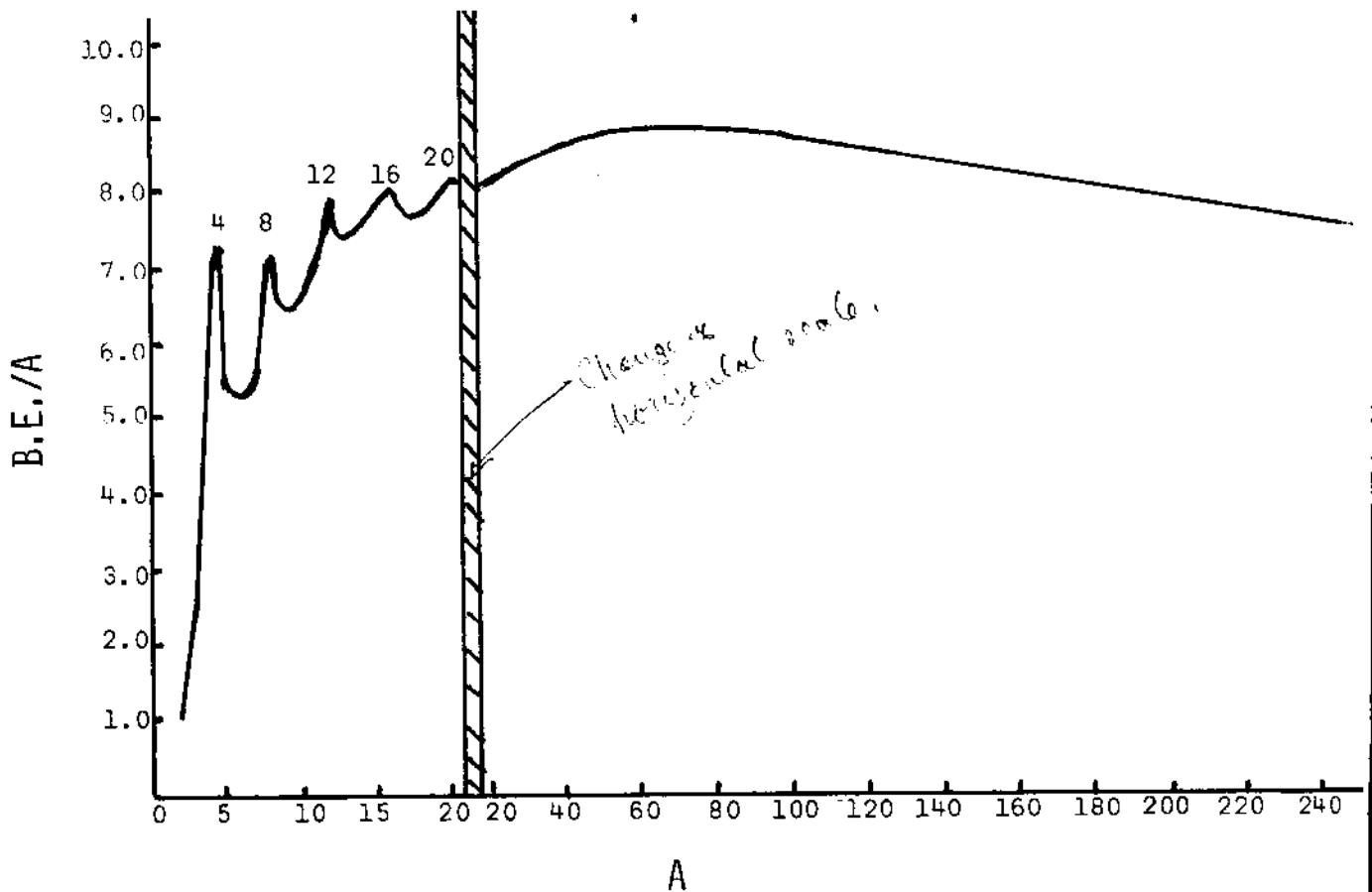


Fig. 1 Plot of the binding energy per nucleon (MeV/nucleon) as a function of the mass number.

Note: BE/A ~ 7-9 MeV

Magic Numbers - 2, 8, 20, 28, 50, 82, and 126.

Exoergic reactions:

- a) fusion of light elements ($A < 40$)
- b) fission of heavy elements ($A > 100$).

Discussion of magic numbers on the tape is confusing, since it does not distinguish clearly between proton number, neutron number, and mass number. The magic numbers refer to either proton number or neutron number, whereas Fig 1 is plotted as a function of mass number. Thus the peak of $A=4$ corresponds to the doubly magic nuclide with $Z=N=2$.

SECTION III
Nuclear Radius

For a sphere of incompressible particles

$$V \propto A$$

For a sphere, $V \propto R^3$

$$R \propto A^{1/3}$$

$$R = R_0 A^{1/3}; \quad R_0 = 1.4 \times 10^{-13} \text{ cm.}$$

Defining 10^{-13} cm as one femtometer (fm),

$$R = 1.4 A^{1/3} \text{ fm} \quad (19)$$

Examples: 1. For ^{238}U $R = 1.4 (238)^{1/3} \text{ fm} = 8.7 \text{ fm}$

2. For ^{80}Br $R = 1.4 (80)^{1/3} \text{ fm} = 6.0 \text{ fm}$

SECTION IV

Nuclear Coulomb Barrier

$$V_c(\text{ergs}) = \frac{(Ze)_1 (Ze)_2}{D(\text{cm})}$$

$$\begin{aligned} e &= \text{electrical unit charge} \\ &= 4.8 \times 10^{-10} \text{ esu} \end{aligned}$$

$$D = R_0 \times 10^{-13} (A_1^{1/3} + A_2^{1/3}) \text{ (cm)} \quad (20)$$

$$\therefore V_c(\text{ergs}) = \frac{Z_1 Z_2 e^2}{R_0 \times 10^{-13} (A_1^{1/3} + A_2^{1/3})}$$

$$V_c(\text{MeV}) = V_c(\text{ergs}) \times 6.24 \times 10^5 \frac{\text{MeV}}{\text{ergs}}$$

or

$$V_c(\text{MeV}) = \frac{1.439 Z_1 Z_2}{R_0 (A_1^{1/3} + A_2^{1/3})} \quad \text{when } R_0 \text{ is given in fm} \quad (21)$$

Example: Calculate the coulomb barrier in MeV for the reaction of ${}^{238}_{92}\text{U} + {}^4_2\text{He}$ $R_0 = 1.5 \text{ fm}$

$$(R_0 = 1.4 \text{ fm})$$

$$V_c(\text{MeV}) = \frac{1.439 \times 92 \times 2}{1.4 (238^{1/3} + 4^{1/3})} = 24.2 \text{ MeV} \quad (22)$$

Note: No coulomb barrier for a neutron.

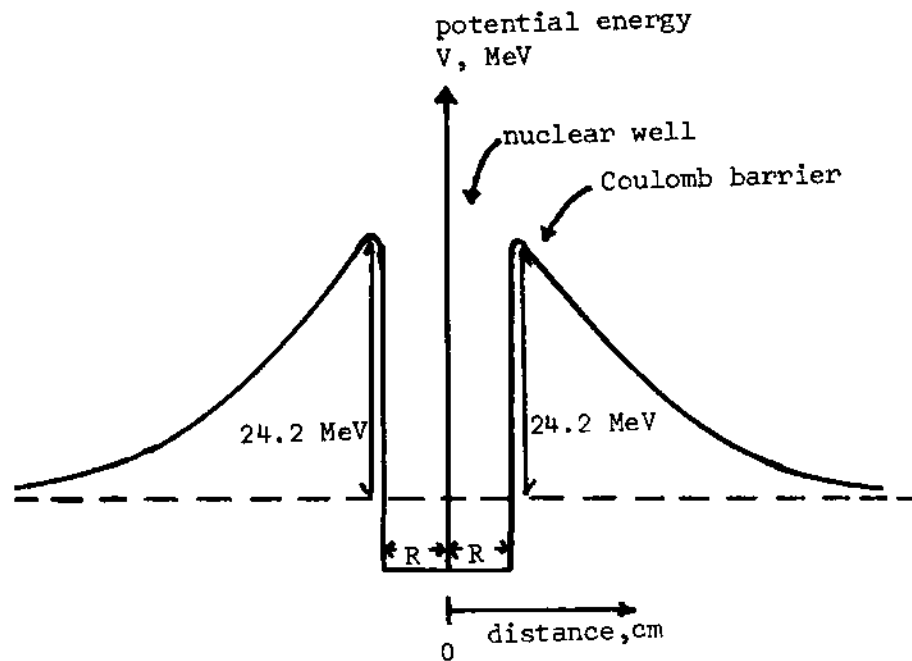


Fig. 2 Nuclear coulomb barrier and potential well for $^{238}_{92}\text{U}$ and ^4_2He .

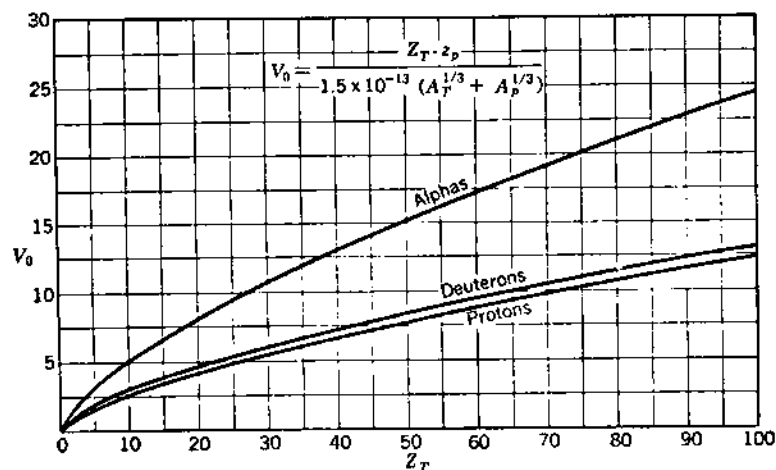


Fig. 3. The Coulomb barrier height as a function of Z_T , the atomic number of the target.

SECTION V

Nuclear Models

A. The Shell Model

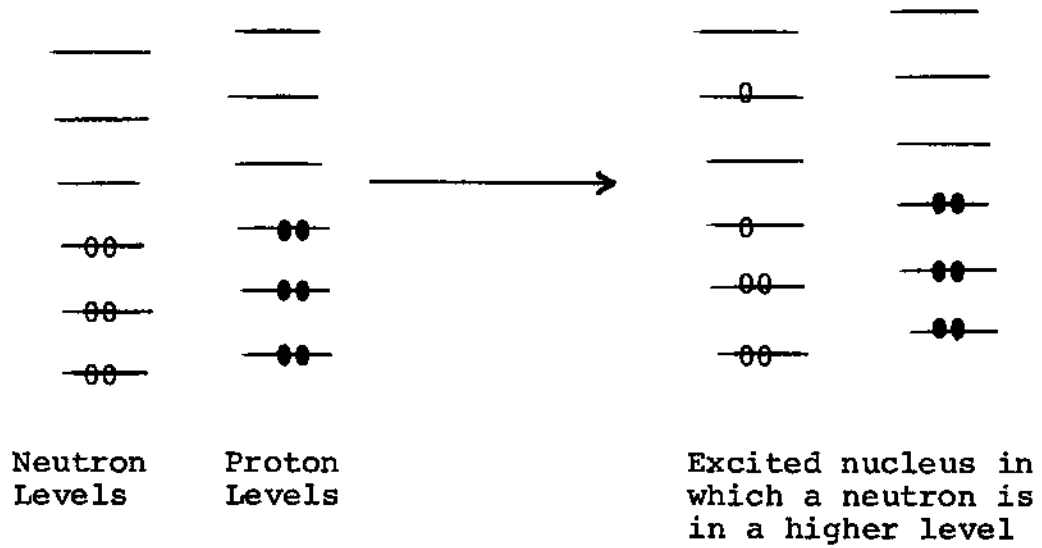


Fig. 4. Process of nuclear excitation by promotion of a neutron) in the shell model. The nucleus returns to the ground state by emission of one or more gamma rays.

B. The Liquid-Drop Model

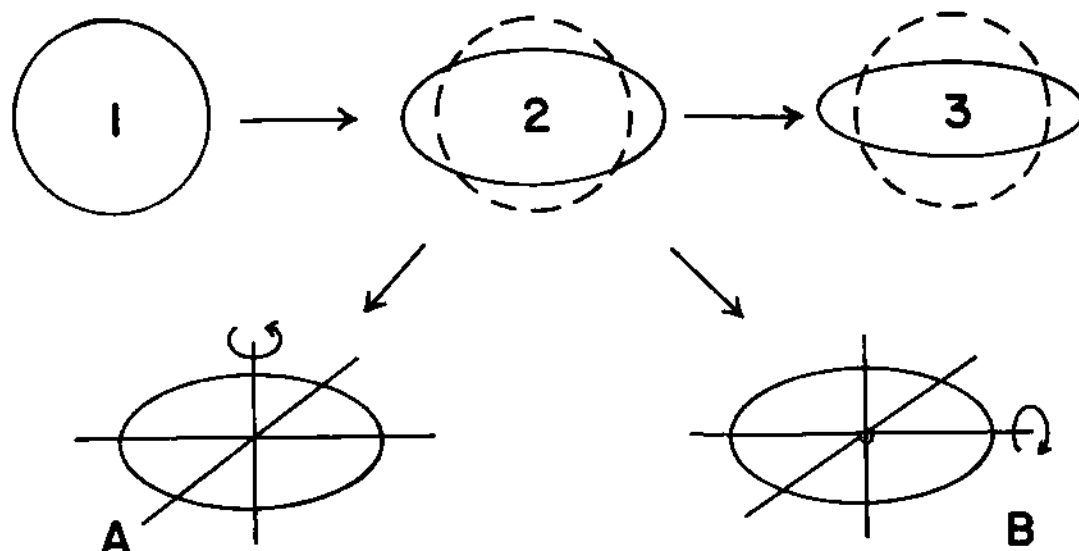


FIG. 5 Excitation of the nucleus into different vibrational (2 and 3) and rotational (A and B) states according to the liquid drop model.

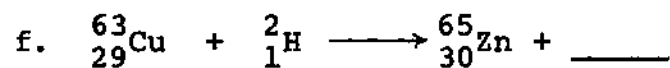
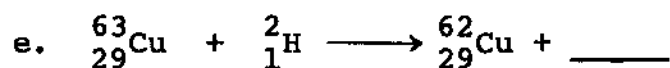
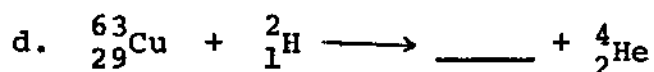
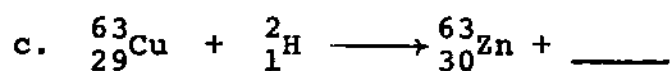
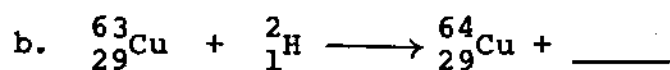
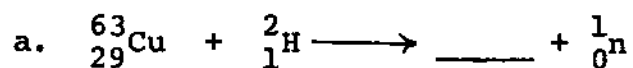
C. The Unified Model

1. Combination of shell and liquid drop models.
2. As Z or N values approach "magic numbers", the nuclear liquid droplet becomes more spherical and "stiffer" (requires more energy to deform).
3. For "stiff" nuclei, individual nucleon excitations require less energy than the rotational and vibrational excitations.

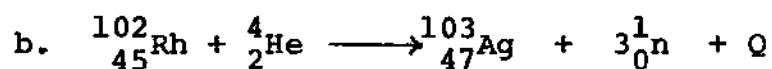
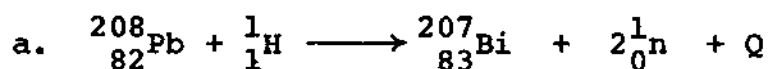
SECTION VI

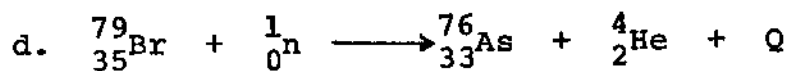
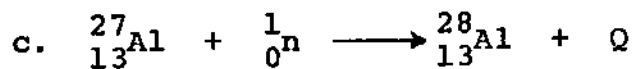
Problems

1. When the copper-63 nuclide is bombarded with deuterons, six different transmutations may occur. Complete the equations for these:



2. Calculate Q for the following reactions.





Atomic Masses (amu)

$${}_1^1\text{H} = 1.0078$$

$${}_{83}^{207}\text{Bi} = 206.9784$$

$${}_{35}^{79}\text{Br} = 78.9183$$

$${}_2^4\text{He} = 4.0026$$

$${}_{102}^{102}\text{Rh} = 101.9068$$

$${}_{33}^{76}\text{As} = 75.9242$$

$${}_0^1\text{n} = 1.0087$$

$${}_{47}^{103}\text{Ag} = 102.9083$$

$$\gamma \text{ ray} = \text{no rest mass}$$

$${}_{13}^{27}\text{Al} = 26.9815$$

$${}_{82}^{208}\text{Pb} = 207.9766$$

$${}_{13}^{28}\text{Al} = 27.9819$$

3. Calculate the total binding energy and the binding energy per nucleon for:

Masses (amu)

a. ${}_{12}^{24}\text{Mg}$

$${}_{12}^{24}\text{Mg} = 23.9850$$

b. ${}_{27}^{60}\text{Co}$

$${}_{27}^{60}\text{Co} = 59.9338$$

c. ${}_{79}^{197}\text{Au}$

$${}_{79}^{197}\text{Au} = 196.9665$$

4. Calculate the radii of the following nuclei using $R_0 = 1.4 \times 10^{-13}$ cm.

${}^4\text{He}$, ${}^{16}\text{O}$, ${}^{56}\text{Fe}$, ${}^{75}\text{As}$, ${}^{112}\text{Cd}$, ${}^{165}\text{Ho}$, ${}^{206}\text{Pb}$, ${}^{238}\text{U}$, and ${}^{256}\text{Fm}$.

5. Plot the radius values calculated in the preceding problem as a function of mass number and discuss the the resulting curve.
6. Calculate the coulomb barrier of a) ${}^{238}\text{U}$, and, b) ${}^{56}\text{Fe}$ nucleus to a proton when they are just in contact.
7. The density of metallic aluminum is 2.7 g/cm^3 . Calculate the density of the ${}^{27}\text{Al}$ nucleus, using $R_0 = 1.4 \text{ fm}$ and $M_{{}^{27}\text{Al}} = 27.0 \text{ amu}$, and compare the result with the 2.7 g/cm^3 for the atom in the metallic lattice.

PART II

Nuclear Stability and Radioactivity

Outline

- I. Stability Patterns
 - A. Odd-Even Rules for Nuclear Stability
 - B. Nuclear Forces
 - C. Neutron to Proton Ratio
- II. Modes of Radioactive Decay
 - A. Rules for Calculation of Nuclear Decay Energies
 - B. Alpha (α) Decay
 - C. Beta Decay
 - 1. Negatron (β^-) Decay
 - 2. Positron (β^+) Decay
 - 3. Electron Capture (EC)
 - D. Gamma (γ) Decay
- III. Decay Schemes
- IV. Decay Laws
 - A. The Decay Constant and Half Life
 - B. Specific Activity
- V. Parent-Daughter Relationships
 - A. Case I
 - B. Case II - Transient Equilibrium
 - C. Case III - Secular Equilibrium
- VI. The Chart of the Nuclides
- VII. Problems

SECTION I

Stability Patterns

<u>Abundance</u>	<u>Proton No.</u>	<u>Neutron No.</u>	<u>Mass Number</u>
60%	even	even	even
40%	even odd	odd even	odd odd
4 Nuclei:	odd	odd	even
(${}^2_1\text{H}$, ${}^6_3\text{Li}$, ${}^{10}_5\text{B}$, ${}^{14}_7\text{N}$)			

A. Odd-Even Rules for Nuclear Stability

1. Elements of odd Z have at most 2 stable isotopes.
2. Adjacent isobars, ${}^A_Z\text{M}$ and ${}^A_{Z+1}\text{M}$, cannot both be stable.

Ex: Technetium ($Z=43$) - no stable isotopes.

Stable Values of A .

${}^{42}\text{Mo}$	92	94	95	96	97	98	100
${}^{43}\text{Tc}$					[97]	[99]	
${}^{44}\text{Ru}$				96		98	99 100 101 102 104

[] indicates values of A most likely to be stable for $Z=43$.

3. Isobars separated by 2 units of Z can be stable

e.g. ${}_{28}^{64}\text{Ni}^{36}$ and ${}_{30}^{64}\text{Zn}^{34}$ stable but ${}_{29}^{64}\text{Cu}^{35}$ radioactive

(neutron numbers are superscripts on right).

B. Nuclear forces

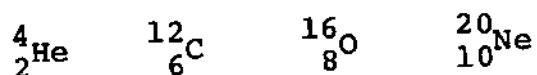
1. Coulomb electrostatic force: exists between protons; for nucleus it is repulsive, charge dependent, long range, proportional to Z^2/D (D = distance of proton-proton separation).

2. Nuclear force: exists between all nucleons and is attractive, charge independent, short range.

C. Neutron to Proton Ratio

1. Light nuclei

For stability $N = Z$; e.g.



2. As number of protons increase, in heavier nuclei when $Z=N$, p-p repulsion tends to outweigh sum of p-p, p-n, and n-n attractions. Thus, number of neutrons must increase at a more rapid rate than

number of protons. As Z increases, $n-p$ must increase (e.g.: $n/p = 1.6$ for uranium).

3. n/p for stability usually has a narrow range of values for a particular element.

		A of Stable Isotopes	n/p
even Z	50^{Sn}	112,114,115,116,117,118,119, 120,124	1.24-1.40
odd Z	49^{In}	113	1.31
	51^{Sb}	121,123	1.37-1.41

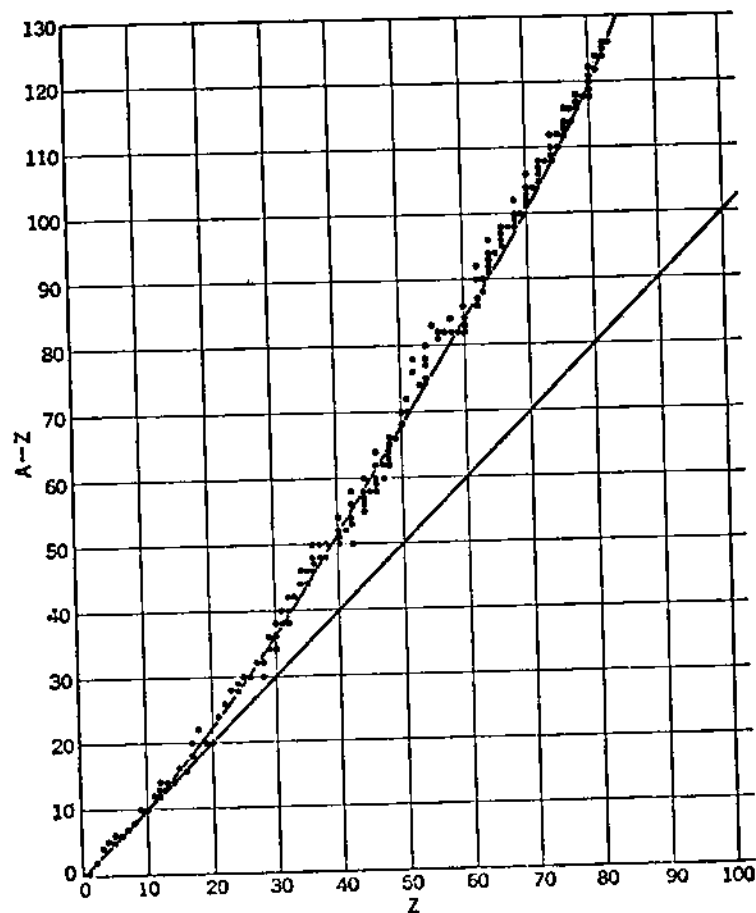


FIG. 1 Plot of the number of neutrons ($A-Z$) vs. the number of protons Z for stable nuclei ($>10\%$ abundance).

SECTION II

Modes of Radioactive Decay

3 modes of radioactive decay

1. Alpha (α) decay
2. Beta (β^- or β^+) decay and electron capture
3. Gamma (γ) decay

Spontaneous Fission

A. Rules for Calculation of Nuclear Decay Energies

The Q value, or the energy released may be calculated from the atomic masses if they are known.

Recall: $Q \equiv [\text{Sum of the atomic masses of the reactants}]$

$-\text{[Sum of the atomic masses of the products]}$

$$Q \equiv \Delta M \text{ amu} \times 931 \text{ MeV/amu}$$

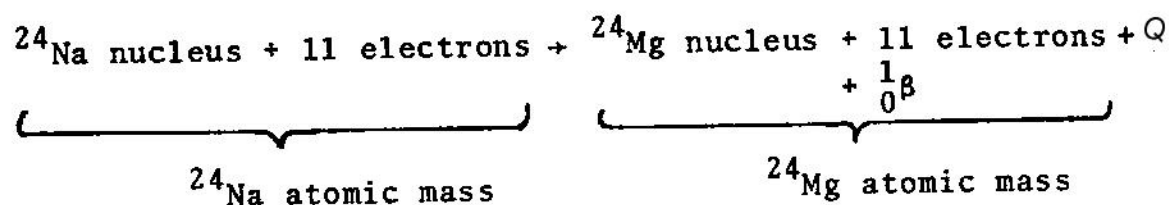
If Q is positive \equiv exoergic (occurs spontaneously)

negative \equiv endoergic (requires energy to occur).

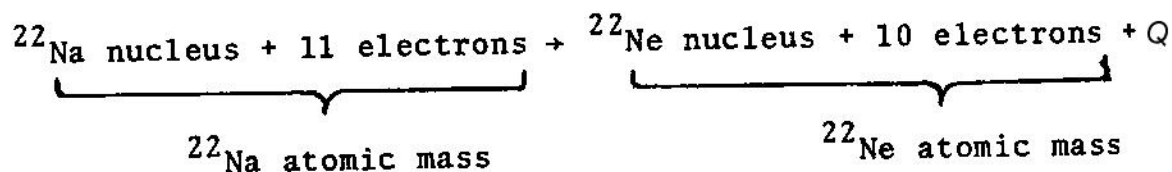
1. In equations involving negatron (β^-), electron capture (EC) or alpha (α) decay, it is not necessary to add or subtract electron masses.

Examples:

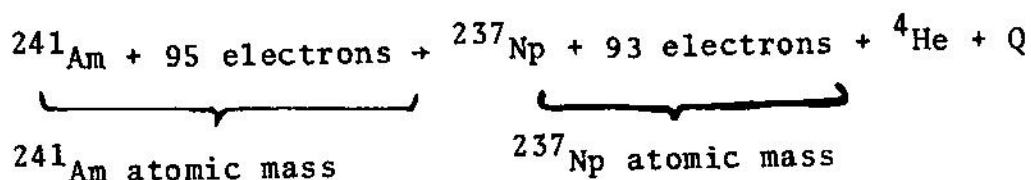
a) β^- Decay



b) EC Decay



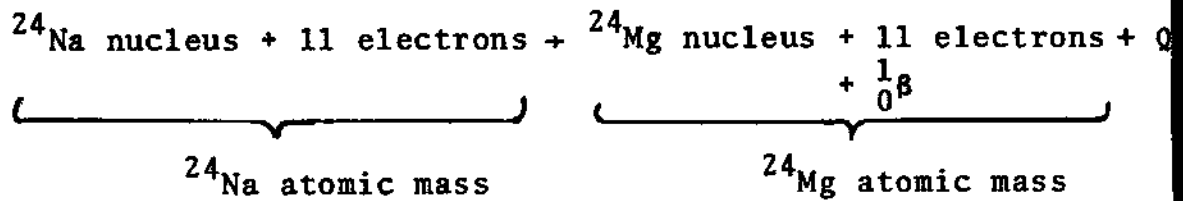
c) Alpha Decay



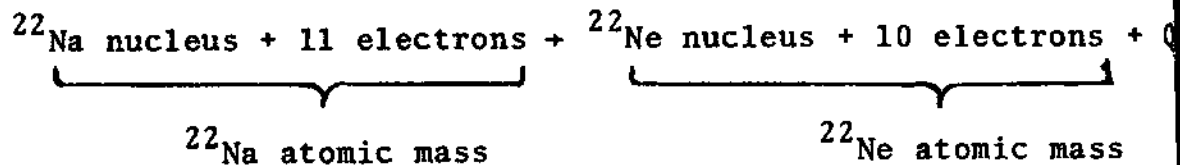
2. In equations involving positron (β^+) emission, it is necessary to subtract an additional two electron masses (i.e. 1.02 MeV) from the difference in masses of the initial and final product nuclei to obtain Q

Examples:

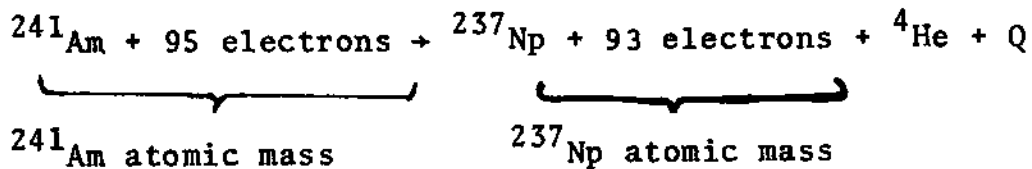
a) β^- Decay



b) EC Decay



c) Alpha Decay



2. In equations involving positron (β^+) emission, it is necessary to subtract an additional two electron masses (i.e. 1.02 MeV) from the difference in masses of the initial and final product nuclei to obtain Q

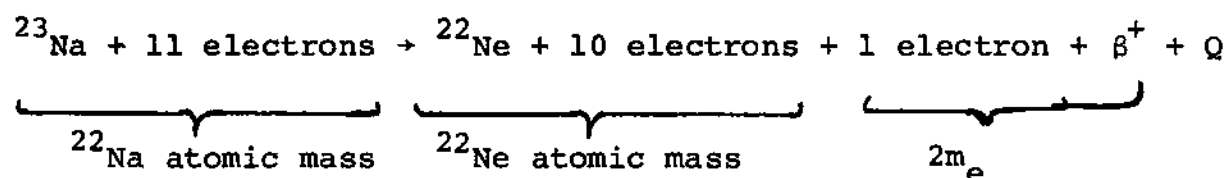
$$Q = [\text{Sum of atomic masses of the reactants}]$$

$$- [\text{Sum of atomic masses of the products}] - 1.02 \text{ MeV}$$

$$Q = \Delta m \text{ amu} \times 931 \text{ MeV/amu} - 1.02 \text{ MeV}$$

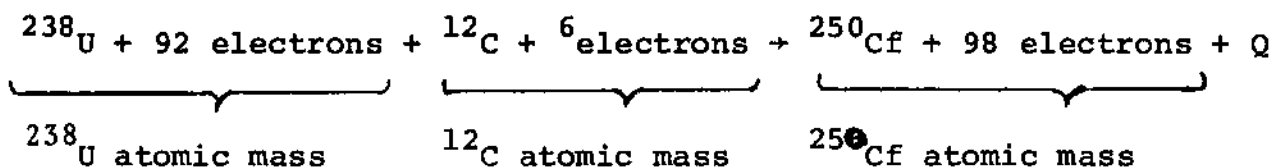
Example

β^+ Decay



3. It is not necessary to subtract electron masses in equations of nuclear reactions induced by charged particles or neutrons

Example



B. Alpha (α) Decay (Heavier Elements)



$$\Delta M = 238.0508 - 4.0026 - 234.0436$$

$$= + 0.0046 \text{ amu}$$

$$Q = (931 \text{ MeV/amu}) (0.0046 \text{ amu}) = 4.28 \text{ MeV}$$

Energy of the α particle

$$E_{\alpha} = \frac{M_{\text{Th}}}{M_{\text{Th}} + M_{\text{He}}} \times Q \approx \frac{A_{\text{Th}}}{A_{\text{Th}} + A_{\text{He}}} \times Q$$

$$= \frac{234}{238} \times 4.28 \text{ MeV} = 4.21 \text{ MeV} \quad (2)$$

Note: α -decay energy is discrete as transitions occur between states of definite energy.

$$E_{\text{recoil}} = E_{\alpha} \left(\frac{4}{238} \right) = 0.07 \text{ MeV} = 70 \text{ keV} \quad \text{carried off} \quad (3)$$

by residual
 ${}^{234}_{90}\text{Th}$ nucleus

Note: $Q = E_{\alpha} + E_{\text{recoil}}$

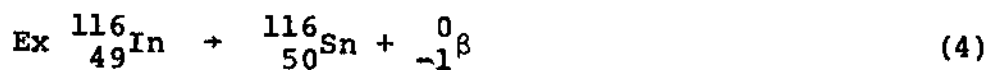
$$= 4.21 \text{ MeV} + 0.07 \text{ MeV}$$

$$= 4.28 \text{ MeV}$$

C. Beta Decay

1. Negatron (β^-) Decay

- a. Occurs throughout the periodic table.
- b. Neutron rich nuclides attempt to achieve a stable n/p ratio by β^- decay.



$$Q_{\beta^-} = 3.35 \text{ MeV}$$

$$\begin{aligned} E_{\text{recoil}} &= \frac{M_e}{M_{e^-} + M_{\text{Sn}}} \cdot Q_{\beta^-} = 5.49 \times 10^{-6} \text{ amu} \cdot Q_{\beta^-} \\ &= 1.59 \times 10^{-5} \text{ MeV} \approx 16 \text{ eV carried off by} \quad (5) \\ &\quad \text{residual } {}^{116}\text{Sn nucleus.} \end{aligned}$$

- c. Energy of emitted β^- particles from 0 to Q_{β}
(average energy $\approx 1/3 Q_{\beta}$)

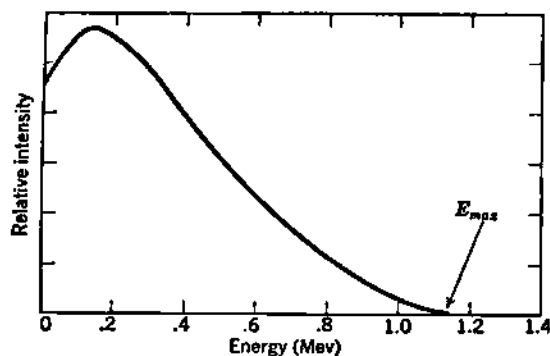
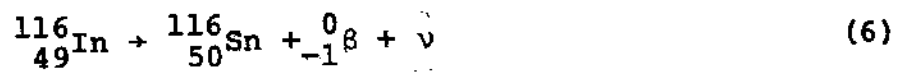


Fig. 2. Energy Spectrum for β^- rays

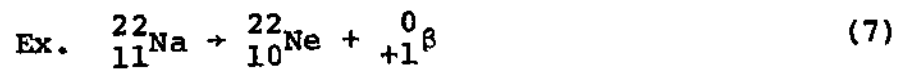
- d. Remaining energy carried off by neutrino,
(uncharged particle of zero rest mass).

Complete reaction



2. Positron (β^+) Decay

- a. Occurs if n/p ratio is too low



$$Q_{\beta^+} = 1.77 \text{ MeV}$$

- b. Energy of emitted positron averages about 0.4 Q_{β^+}

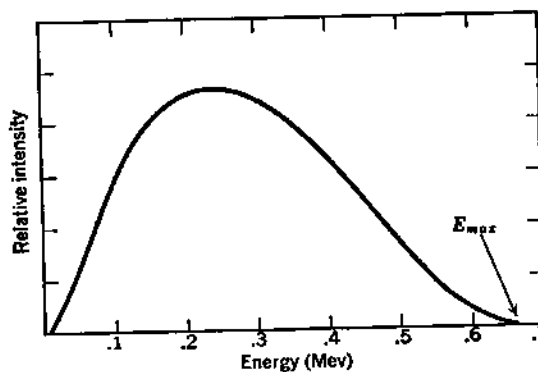


Fig. 3. Energy spectrum for β^+ rays

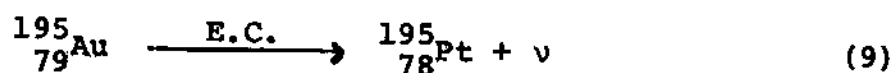
- c. Remaining energy carried off by neutrino (ν).

Complete reaction



3. Electron Capture

- a. Occurs when n/p ratio is too low
- b. Electron is absorbed from extranuclear electron shells of the atom (predominantly, the K shell)



$$\Delta m = 0.000283 \text{ amu} \times 931 \text{ MeV/amu} = 0.263 \text{ MeV}$$

$$Q_{\text{EC}} = 0.263 \text{ MeV}$$

- c. E.C. vs β^+

If the available decay energy [i.e. $\Delta m(\text{amu}) \times 931$ (MeV/amu)] is less than 1.02 MeV, positron emission is impossible and decay must take place by electron capture, E.C. If the available decay energy is greater than or equal to 1.02 MeV, decay can occur by either β^+ or EC or both.

$$^{22}_{\text{Na}} + ^{22}_{\text{Ne}} \qquad M_{^{22}_{\text{Na}}} = 21.99444 \text{ amu}$$

$$M_{^{22}_{\text{Ne}}} = 21.99139 \text{ amu}$$

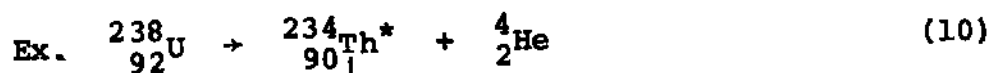
$$\text{Available energy} = (3.052 \times 10^{-3} \text{ amu}) (931 \text{ MeV/amu})$$

$$= 2.94 \text{ MeV}$$

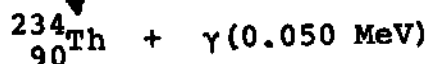
This reaction can go either by β^+ or EC or both. In reality $^{22}_{\text{Na}}$ decays 79% by β^+ and 11% by EC.

D. Gamma (γ) Decay

1. Must accompany another form of radioactive decay or occur as a consequence of a nuclear reaction which leaves a nucleus in an excited state.
e.g., follows α decay



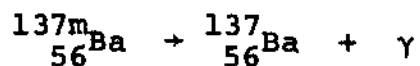
*=excited state



2. Metastable states occur when the excited state lasts a measurable amount of time.



m = metastable



3. Instead of γ -ray emission, orbital electron can be ejected; called a conversion electron.

$$E_e = E_\gamma - E_{BE}$$

E_e - energy of conversion electron

E_γ - energy of γ ray if it were to be emitted

E_{BE} - binding energy of orbital electron (conversion electron)

NOTE: In the examples given for α and β decay the energy of the recoiling nucleus was calculated. This energy imparted to the residual nucleus by the emission of a particle may be sufficient to rupture some or all of the chemical bonds holding the atom in a molecule.

The recoil energy in gamma decay is insufficient to cause bond rupture but such decay is associated with ionization of extranuclear electrons. This can result in bond rupture. The study of these processes are contained in a branch of nuclear sciences called "hot-atom" chemistry.

SECTION III
Decay Schemes

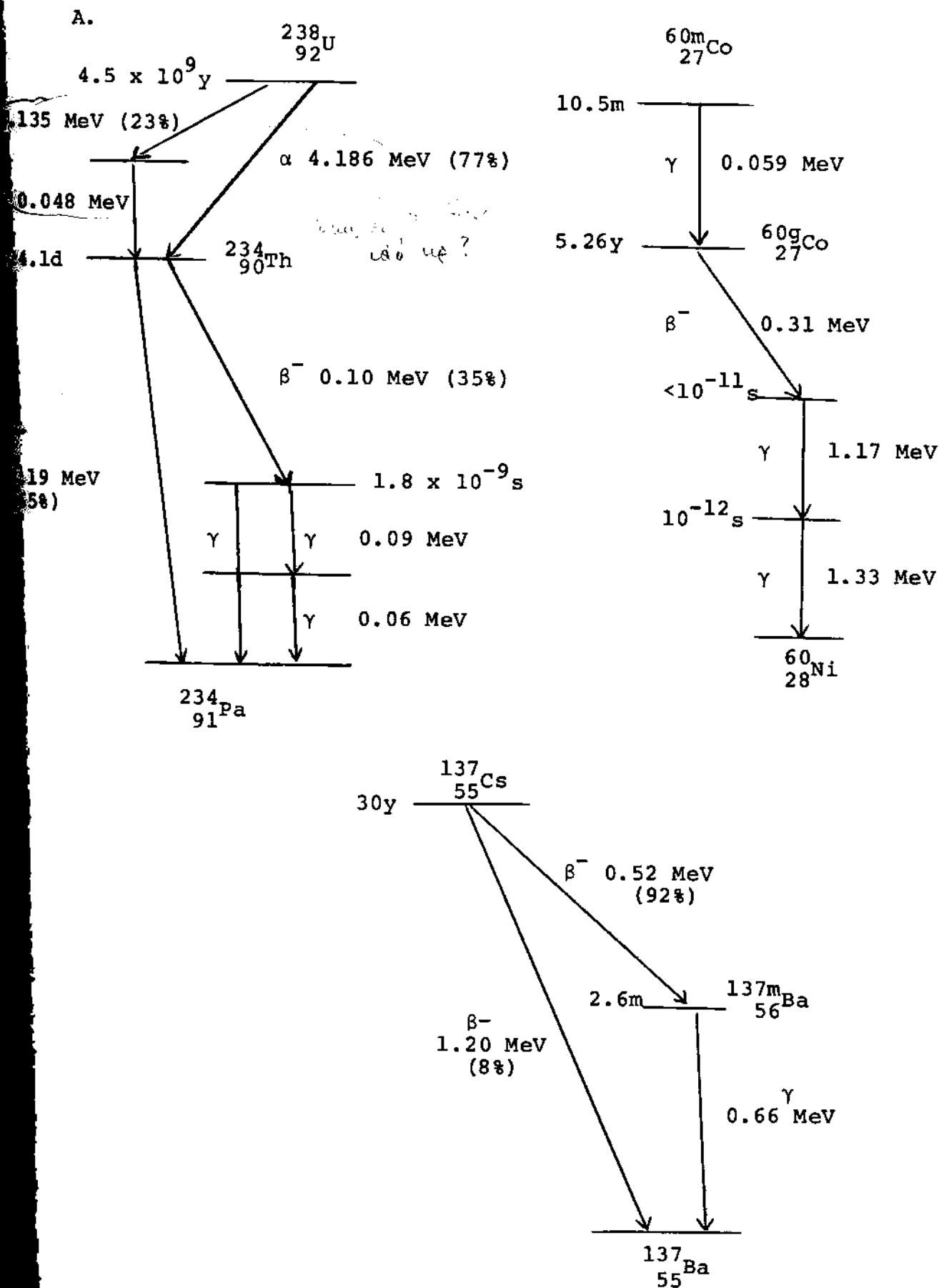


FIG. 4. Decay Schemes for $^{238}_{92}\text{U}$, $^{60\text{m}}_{27}\text{Co}$ and $^{137}_{55}\text{Cs}$

SECTION IV

Radioactive Decay Law

A. The Decay Constant and Half Life

$$A(\text{disintegration/min}) \propto N(\text{number of atoms})$$

$$A = \lambda N \quad (13)$$

λ = decay constant

$$-dN/dt = \lambda N$$

$$N_t = N_0 e^{-\lambda t}$$

$$\log_{10} N_t = \log_{10} N_0 - \frac{\lambda t}{2.3}$$

$$A_t = A_0 e^{-\lambda t} \quad (13a)$$

$$\log_{10} A_t = \log_{10} A_0 - \frac{\lambda t}{2.3} \quad (13b)$$

When $A_t = 0.5 A_0$, $t = t_{1/2}$, the half-life.

$$\frac{A_t}{A_0} = 0.5 = e^{-t_{1/2} \lambda}$$

$$-\lambda t_{1/2} = \log_e 0.5$$

$$t_{1/2} = \frac{-\log_e 0.5}{\lambda}$$

$$t_{1/2} = \frac{0.693}{\lambda} \quad (14)$$

The decay constant (and half-life) is a definite characteristic of the nucleus. The half-life is the time required for 1/2 of a statistically large number of radioactive nuclei to undergo decay.

- B. Specific Activity (SA) - total radioactivity of a given isotope per gram of the radioactive isotope (can also be per gram of a compound or per gram of the element).

$$SA \left(\frac{\text{dpm}}{\text{g}} \right) = \frac{0.693 N}{T_{1/2}} = \frac{0.693}{T_{1/2}} \times \frac{6.023 \times 10^{23} \text{ atoms/mole}}{\text{atomic weight grams/mole}} \quad (15)$$

where $T_{1/2}$ = half-life in minutes

N = number of radioactive atoms/gram

$$= \frac{6.023 \times 10^{23} \text{ atoms/mole}}{\text{atomic weight gram/mole}}$$

Note: atomic weight can again refer to the isotope, compound, or element

or

$$SA\left(\frac{Ci}{g}\right) = SA\left(\frac{dpm}{g}\right) \times \frac{\text{curie}}{2.22 \times 10^{12} \text{ dpm}} \quad (16)$$

Ex. Calculate the SA in $\frac{dpm}{gm}$ and $\frac{Ci}{gm}$ of ^{22}Na ($T_{1/2}=2.6y$).

(per gram of the radioisotope)

$$T_{1/2} = 2.6y = 1.367 \times 10^6 m$$

$$SA\left(\frac{dpm}{g}\right) = \frac{0.693}{1.367 \times 10^6 m} \times \frac{6.023 \times 10^{23} \text{ atoms/mole}}{22 \text{ g/mole}}$$

$$= 1.39 \times 10^{16} \frac{dpm}{g}$$

$$SA\left(\frac{Ci}{g}\right) = \frac{1.39 \times 10^{16} \frac{dpm}{g}}{2.22 \times 10^{12} c} = 6.252 \times 10^3 Ci$$

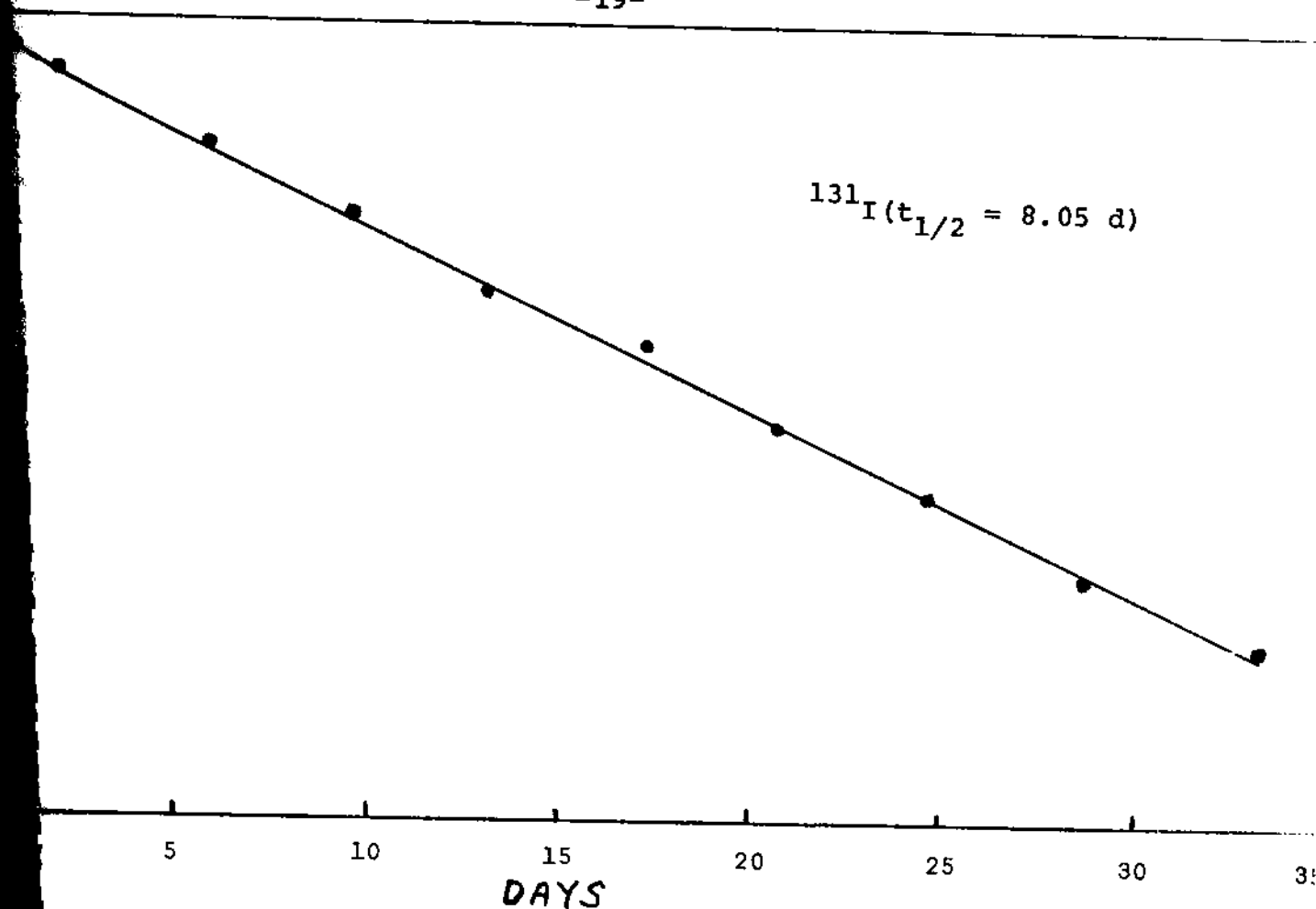


FIG. 5. Decay curve for ^{131}I

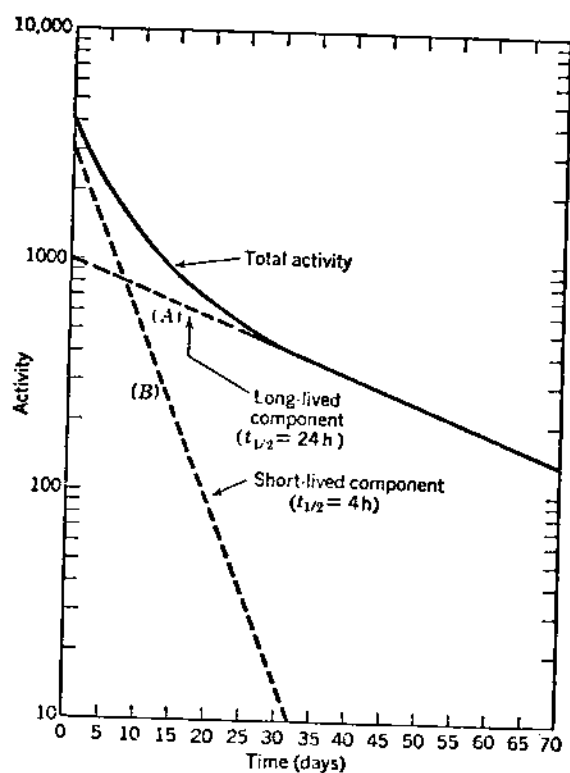
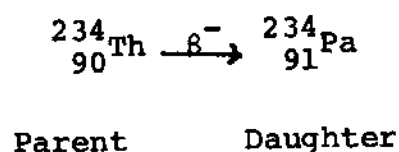


FIG. 6

Analysis of a Decay
Curve with Two
Components

SECTION V

Parent-Daughter Relationships



$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2 \quad (18)$$

Where N_1 and N_2 are the numbers of parent and daughter atoms, respectively, at time t .

$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^0 (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + N_2^0 e^{-\lambda_2 t} \quad (19a)$$

$$\text{At time } t = 0 \quad N_2^0 = 0$$

$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^0 (e^{-\lambda_1 t} - e^{-\lambda_2 t}) \quad (19b)$$

A. Case I $t_{1/2}$ (parent) < $t_{1/2}$ (daughter)

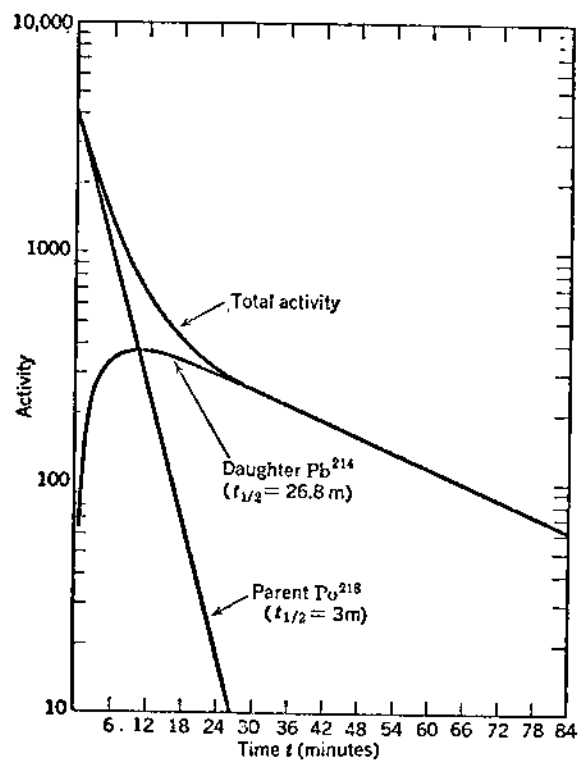


FIG. 7 Growth and decay curves for $^{218}\text{Po} \rightarrow ^{214}\text{Pb}$ system

B. Case II Transient Equilibrium: $t_{1/2}(\text{parent}) > t_{1/2}(\text{daughter})$

$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1 \quad (20)$$

$t_{1/2}(\text{p})/t_{1/2}(\text{d})$ roughly between 1 and 100

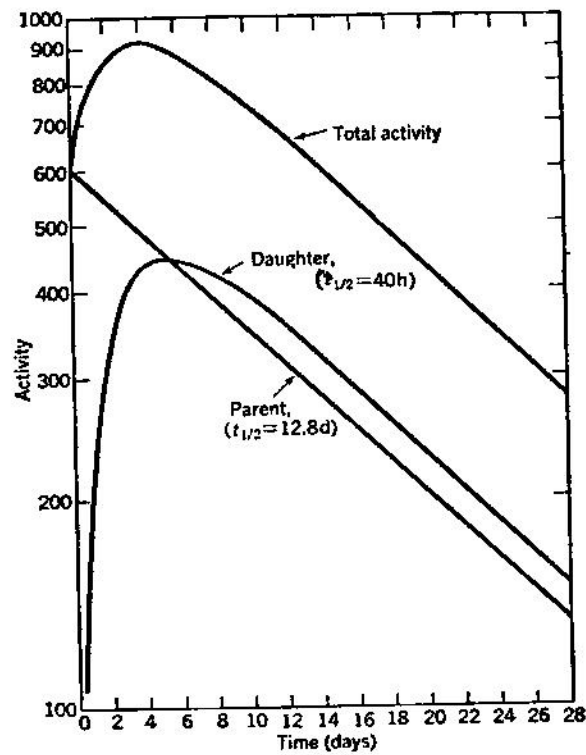


FIG. 8 Transient equilibrium for $^{140}\text{Ba-La}$ system.

Always: Growth of daughter with half life of shorter of the two
Decay of daughter with the halflife of the longer

C. Case III Secular Equilibrium: $t_{1/2}(\text{parent}) \gg t_{1/2}(\text{daughter})$

$$N_2 = \frac{\lambda_1}{\lambda_2} N_1 ; \quad \underline{A_2 = A_1}$$

$$t_{1/2}(\text{p})/t_{1/2}(\text{d}) > 100 \quad (21)$$

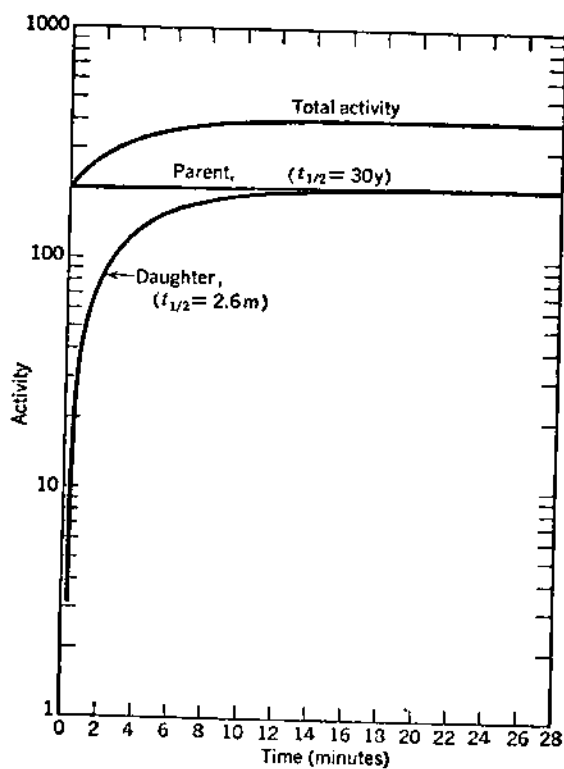


FIG. 9 Secular equilibrium for ^{137}Cs - ^{137}Ba system.

SECTION VI

The Chart of Nuclides

Much data on the radioactivity of nuclides is summarized in the Chart of Nuclides. You should familiarize yourself with how to find nuclides on the chart and extract the desired information.

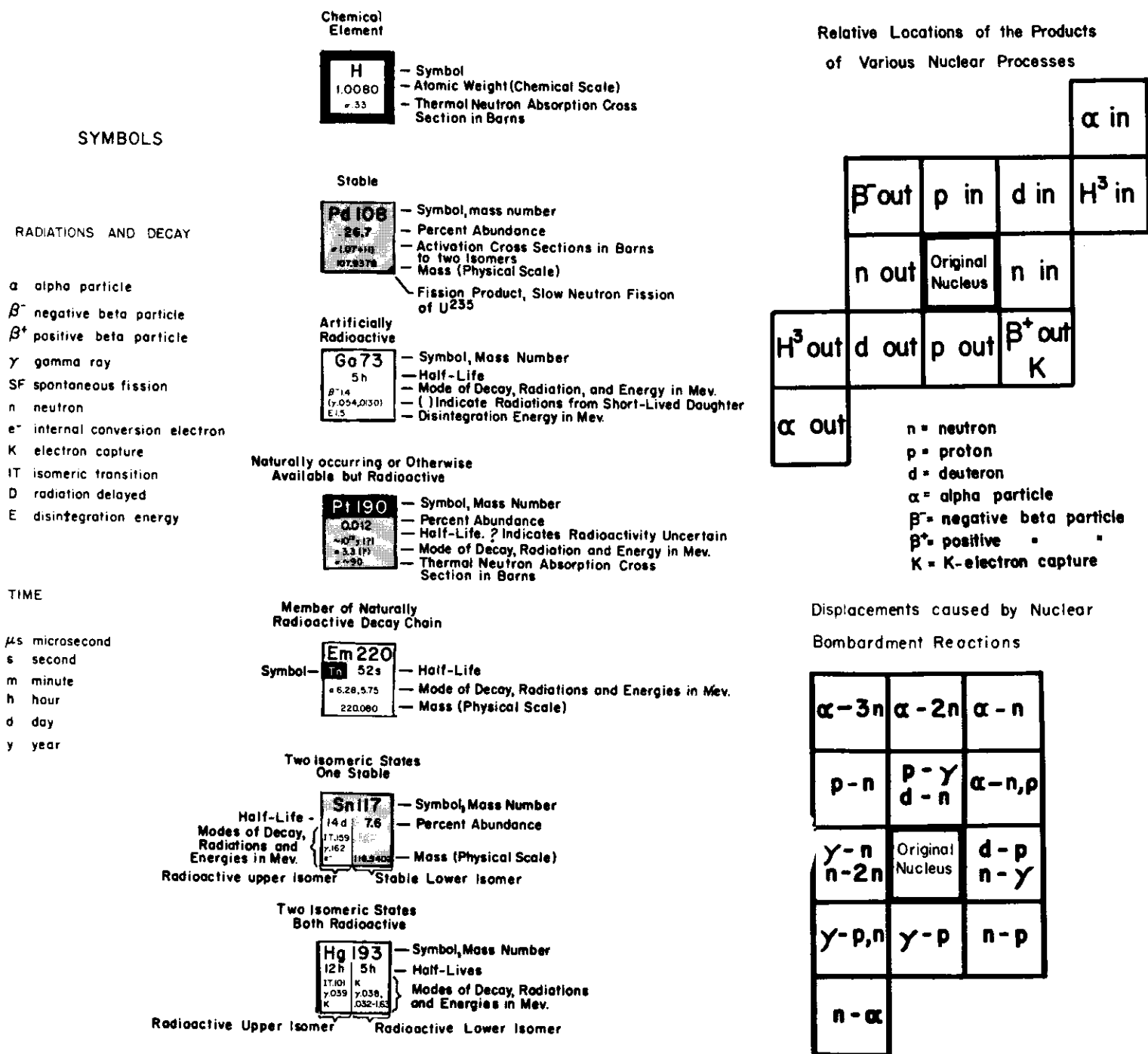


FIGURE 10

Chart of the Nuclides as prepared by General Electric Co. — symbols

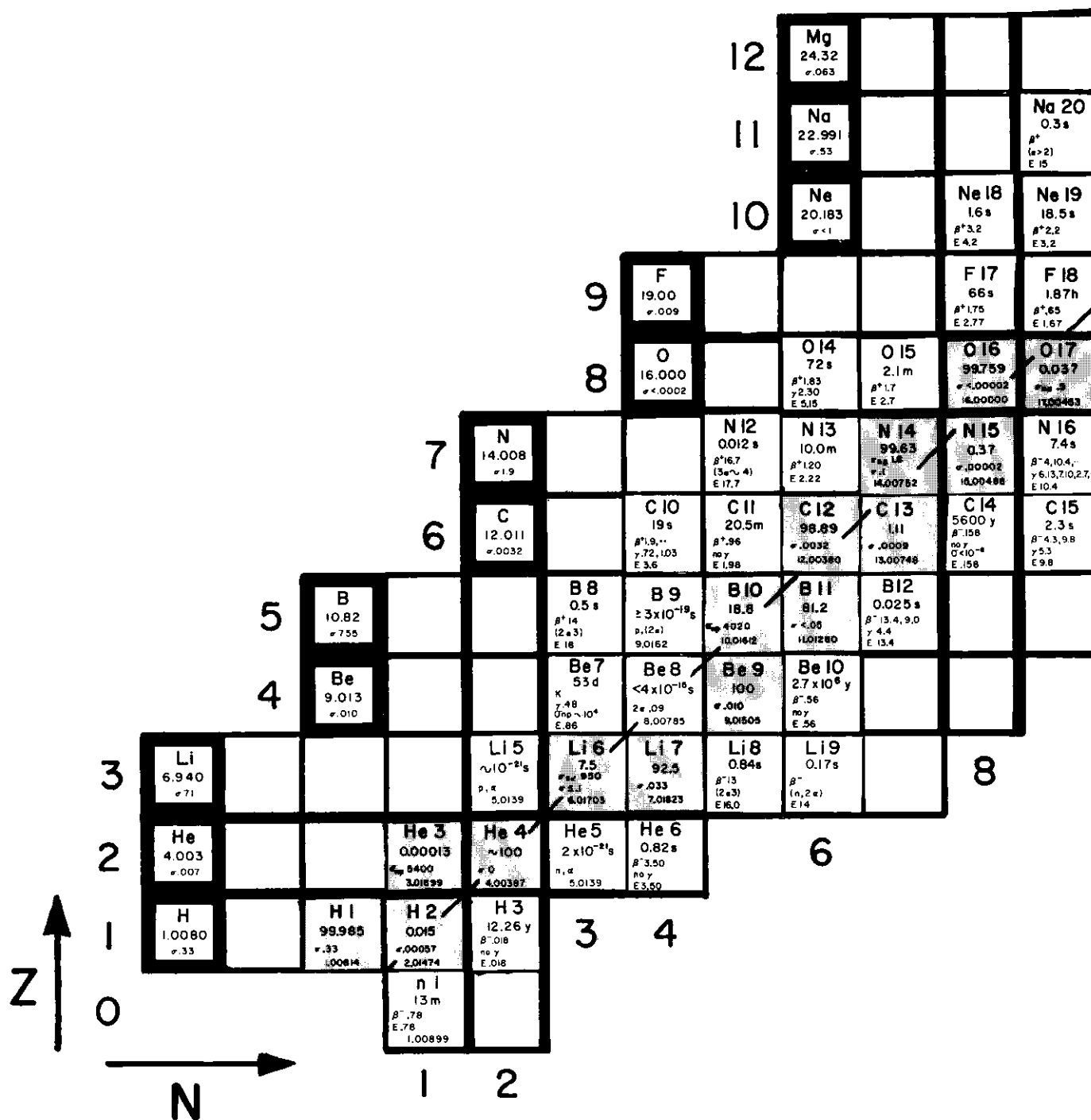


FIGURE 11
Chart of the Nuclides: Z=1→12; N=0→9

SECTION VII

Problems

1. Calculate the decay energy for the possible decay paths and predict the most probable mode of decay.

	amu
a.	
$^{22}_{11}\text{Na} \rightarrow ^{22}_{12}\text{Mg}$	$^{22}_{12}\text{Mg} = 21.9998$
$^{22}_{11}\text{Na} \rightarrow ^{22}_{10}\text{Ne}$	$^{22}_{11}\text{Na} = 21.9944$
	$^{22}_{10}\text{Ne} = 21.9914$
b.	
$^{32}_{15}\text{P} \rightarrow ^{32}_{16}\text{S}$	$^{32}_{16}\text{S} = 31.9721$
$^{32}_{15}\text{P} \rightarrow ^{32}_{14}\text{Si}$	$^{32}_{15}\text{P} = 31.9739$
	$^{32}_{14}\text{Si} = 31.9740$
c.	
$^{141}_{58}\text{Ce} \rightarrow ^{141}_{59}\text{Pr}$	$^{141}_{59}\text{Pr} = 140.9076$
$^{141}_{58}\text{Ce} \rightarrow ^{141}_{57}\text{La}$	$^{141}_{58}\text{Ce} = 140.9082$
	$^{141}_{57}\text{La} = 140.9108$
d.	
$^{239}_{93}\text{Np} \rightarrow ^{239}_{92}\text{U}$	$^{239}_{93}\text{Np} = 239.0529$
$^{239}_{93}\text{Np} \rightarrow ^{239}_{94}\text{Pu}$	$^{239}_{92}\text{U} = 239.0543$
$^{239}_{93}\text{Np} \rightarrow ^{235}_{91}\text{Pa}$	$^{239}_{94}\text{Pu} = 239.0521$
	$^{235}_{91}\text{Pa} = 235.0454$
e.	
$^{249}_{97}\text{Bk} \rightarrow ^{249}_{96}\text{Cm}$	$^{249}_{96}\text{Cm} = 249.0758$
$^{249}_{97}\text{Bk} \rightarrow ^{249}_{98}\text{Cf}$	$^{249}_{97}\text{Bk} = 249.0749$
$^{249}_{97}\text{Bk} \rightarrow ^{245}_{95}\text{Am}$	$^{249}_{98}\text{Cf} = 249.0763$
	$^{245}_{95}\text{Am} = 245.0663$

2. $^{90}_{39}\text{Y}$ decays by β^- emission to $^{90}_{40}\text{Zr}$. If the decay energy, Q , is measured to be -2.79 MeV and the mass of $^{90}_{40}\text{Zr}$ is determined by mass spectrometry to be 89.9047, what is the mass of $^{90}_{39}\text{Y}$?
3. In natural indium, $^{115}_{49}\text{In}$ is found in 95.8% abundance; in natural tin, $^{115}_{50}\text{Sn}$ is found in 0.38% abundance. Which would be expected to be unstable with respect to the other, and what approximate lower limit can be given the half life, since this nuclide is still found in nature, if the age of the elements is assumed to be 7×10^9 years?

Masses $^{115}_{49}\text{In} = 114.9039 \text{ amu}$ $^{115}_{50}\text{Sn} = 114.9033 \text{ amu}$

4. What is the mass of $^{235}_{92}\text{U}$ if the decay energy for the alpha decay of $^{239}_{94}\text{Pu}$ is 5.21 MeV?

$$^4_2\text{He} = 4.0026 \text{ amu}$$

$$^{239}_{94}\text{Pu} = 239.0521 \text{ amu}$$

5. Calculate the number of grams and the number of radioactive atoms in a millicurie of:

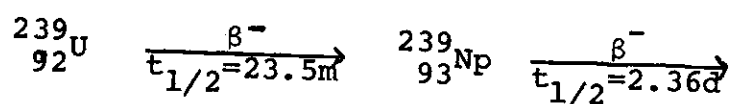
a. $^{36}_{17}\text{Cl}$ $t_{1/2} = 3 \times 10^5 \text{ y}$

b. $^{76}_{33}\text{As}$ $t_{1/2} = 27.6 \text{ h}$

c. $^{51}_{24}\text{Cr}$ $t_{1/2} = 27.8 \text{ d}$

6. What is the count rate of a sample nine half lives after a rate of 10^7 cpm is measured?

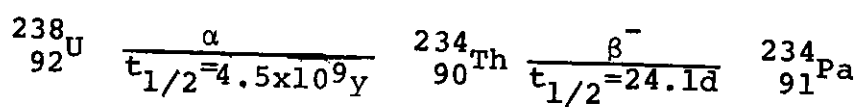
7. If a freshly prepared sample of $^{239}_{92}\text{U}$ of 10^6 dpm is allowed to stand for 6 hours, what will the total disintegration rate be at the end of that time?



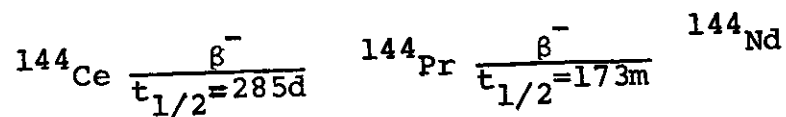
8. The following data were taken in an appropriate counter. Resolve these data into two activities and ascertain their half lives.

<u>t(hr)</u>	<u>dpm</u>	<u>t(hr)</u>	<u>dpm</u>	<u>t(hr)</u>	<u>dpm</u>
0	7000	0.10	2300	30	590
1	6000	12.5	1850	35	450
2	5300	15	1500	40	340
3	4600	17.5	1250	45	260
5	3800	20	1060	50	200
7.5	2860	25	780		

9. If a kilogram of $^{238}_{92}\text{U}$ is purified and then set aside for 2 months, what weight of $^{234}_{90}\text{Th}$ will be present in the uranium sample at the end of that time?

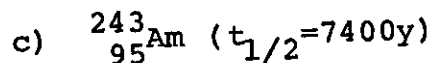
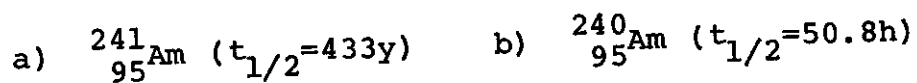


10. For the decay chain



when the ^{144}Ce and ^{144}Pr are in secular equilibrium,
what is the weight of ^{144}Pr per gram ^{144}Ce ?

11. Calculate the specific activity per gram of the isotopes
for the following



How does the specific activity vary with half-life?

PART III

Interaction of Radiation with Matter

Outline

I. Mechanism of Interaction

- A. Molecular Excitation
- B. Ionization

II. Absorption of Radiation

- A. Heavy Particles
- B. Electrons
- C. Gamma and X rays
 - 1. Photoelectric Effect
 - 2. Compton Effect
 - 3. Pair Production

III. Comparison of Absorption Ranges in Material

- A. Alpha Particles
- B. Beta Particles
- C. Gamma Rays

IV. Radiological Safety

- A. Units of Radiation
- B. Effects of Radiation Exposure
- C. Typical Values of Radiation Doses
- D. Maximum Permissible Exposure Levels in Man
- E. Recommended Working Conditions
- F. Decontamination Procedures

V. Problems

SECTION I

Mechanism of Interaction

A. Molecular Excitation

If the energy imparted by a radioactive particle is less than the energy required to ionize an orbital electron, excitation of electron(s) to higher energy levels of the molecule occurs. Deexcitation occurs by emission of low energy radiation in the form of x-rays, visible light, etc.

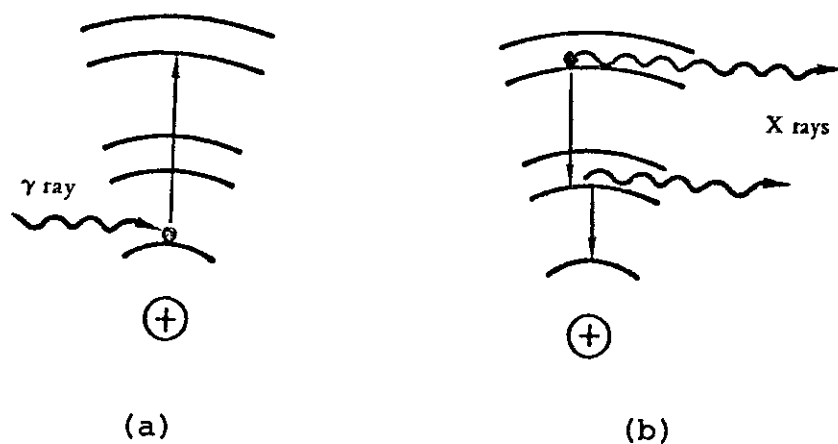


Fig. 1. Excitation by gamma ray: (a) the gamma ray excites the electron to a higher energy level; (b) the electron falls back to the original level in two steps, with X rays or visible light emitted in each step.

B. Ionization

If the energy imparted by radioactive particles is equal to or greater than the energy required to remove an orbital electron, ionization results in formation of an ion pair.

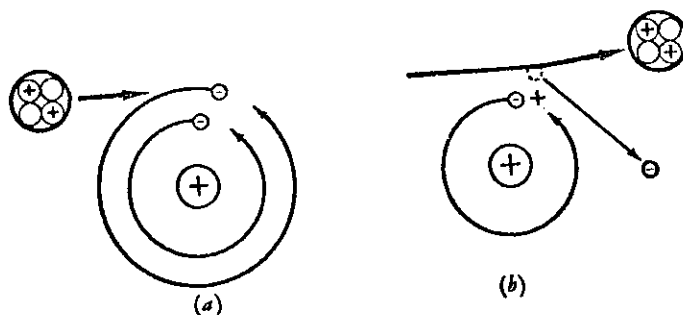


Fig. 2. Formation of an ion pair by α particle:
(a) alpha particle approaches an orbital electron
and (b) causes it to leave the atom, producing an
ion pair.

SECTION II

Absorption of Radiation

A. Heavy Particles:

Lose energy by interaction with coulomb field of orbital electrons. α particles traverse straight paths and have definite ranges in matter.

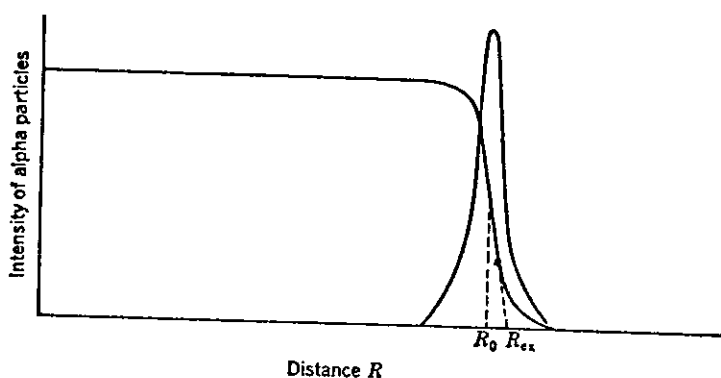


Fig. 3. Range curve for alpha particles.

Specific ionization - measures rate of energy loss

$-\frac{dE}{dX}$ per unit length in matter

$$-\frac{dE}{dX} \propto \frac{1}{v^2} \quad v = \text{velocity of } \alpha \text{ particle} \quad (1)$$

also,

$$-\frac{dE}{dX} \propto Z \quad Z = \text{atomic number of absorber} \quad (2)$$

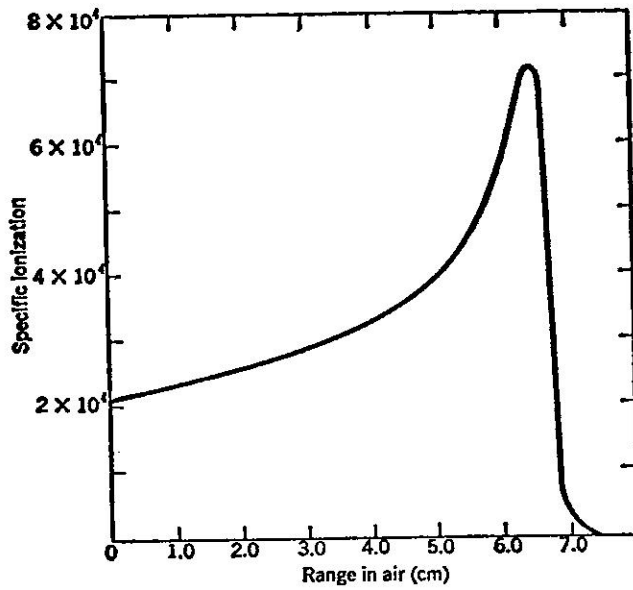


Fig. 4. Specific ionization of 7.7 MeV alpha particles in air. Important: ~ 34 eV lost per ion pair formed in gases; ~ 5 eV in solids and liquids.

TABLE I

W - AVERAGE ENERGY LOSS PER ION PAIR

<u>Gas</u>	<u>Particle</u>	<u>w(ev)</u>	<u>Gas</u>	<u>Particle</u>	<u>w(ev)</u>
Air	Electron	32.0	Helium	Alpha	42.7
Air	Proton	36.0	Neon	Alpha	36.8
Air	Alpha	35.5	Argon	Alpha	26.4
Hydrogen	Alpha	36.3	Methane	Alpha	29.4

B. Electrons:

Lose energy like α particles but for the same energy the velocity of electrons is much greater so lower specific ionization (remember - $\frac{dE}{dX} \propto \frac{1}{v^2}$).

Due to small mass, electron can lose large fraction of its energy in one collision. Large angle deflections result. Observed absorption follows exponential decrease with absorber thickness.

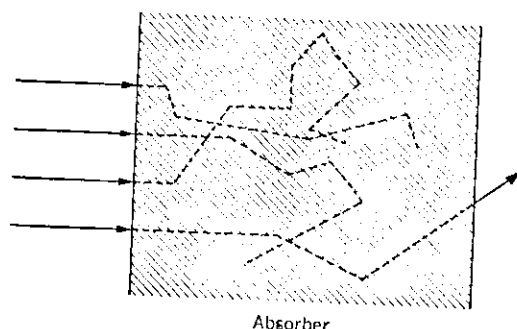
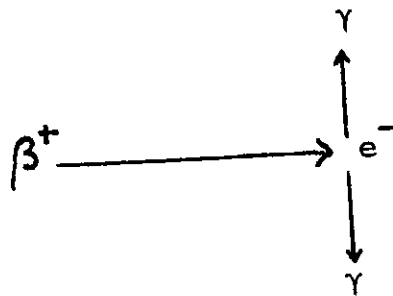


Fig. 5. Electron paths in an absorber

Bremsstrahlung - low energy X rays emitted when electrons are decelerated and bent in their paths by coulomb field of atomic nuclei.

Positron - interacts with electron, both annihilated and 2 gamma rays of 0.51 MeV each created.



C. Gamma and X rays:

Mechanism of interaction completely different than that of charged particle (α or electron); do not have a definite range in matter, exponential absorption.

1. Photoelectric Effect

Absorption of low energy γ rays by an atoms as a whole resulting in ionization

$$E_{e^-} = E_{\gamma} - E_{B.E.} \quad (3)$$

Where E_{e^-} = energy of photoelectron,
 E_{γ} = energy of gamma ray absorbed,
 $E_{B.E.}$ = binding energy of ejected electron

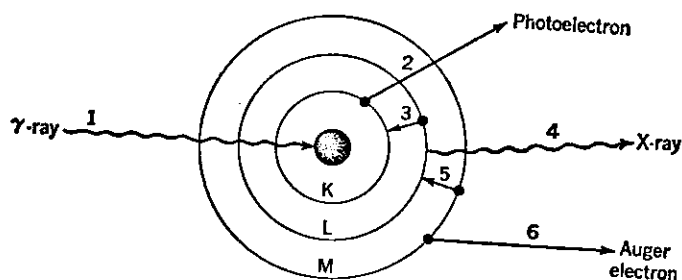


Fig. 6. The photoelectric effect: (1) complete absorption of the gamma ray by the absorber atom causing a photoelectron (2) to be ejected from the K-shell. The vacancy in an inner orbital results in electrons from higher levels falling down to fill all the lower vacancies, (3) and (5), x rays (4) characteristic of the absorber atom will be emitted with energies equal to the difference in binding energies of the two electronic levels involved in the transition. In place of an X ray, a low energy electron, known as an Auger electron(6) may be emitted.

2. Compton Effect

If the energy of the gamma ray is sufficiently large, rather than interaction with the atom as a whole, the γ ray may interact with any one of the orbital electrons as though it were essentially a free electron. Result: a Compton electron is ejected creating an ion pair.

$$E_{\gamma_2} = E_{\gamma_1} - E_{B.E. e^-} - E_{e^-} \quad (4)$$

If ejected e^- is from an inner orbital, X ray and Auger electron emission will occur as described for the photoelectric effect

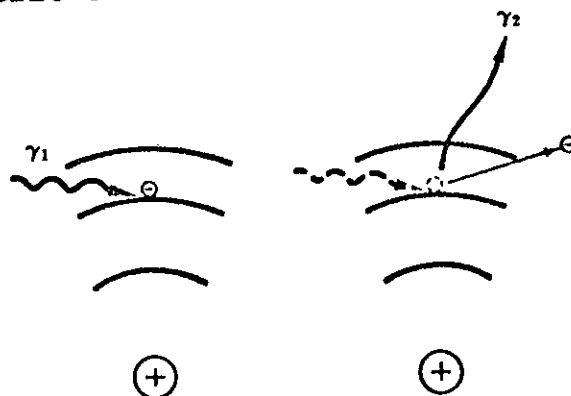


Fig. 7. Compton Scattering

3. Pair Production

Absorption of high energy γ rays cause formation of e^+ and e^- pair. Threshold gamma energy is 1.02 MeV.

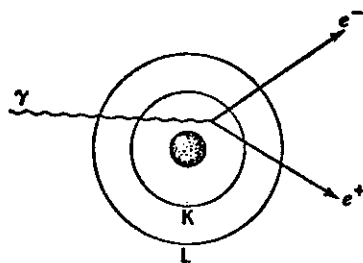


Fig. 8. Pair Production

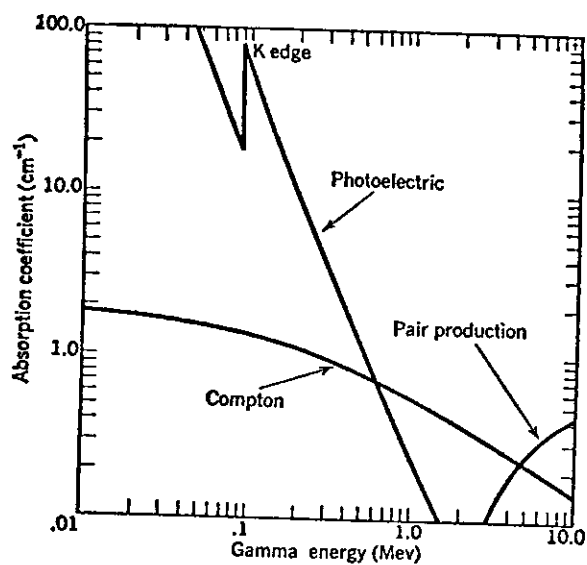


Fig. 9. Dependence on energy of absorption coefficients for lead.

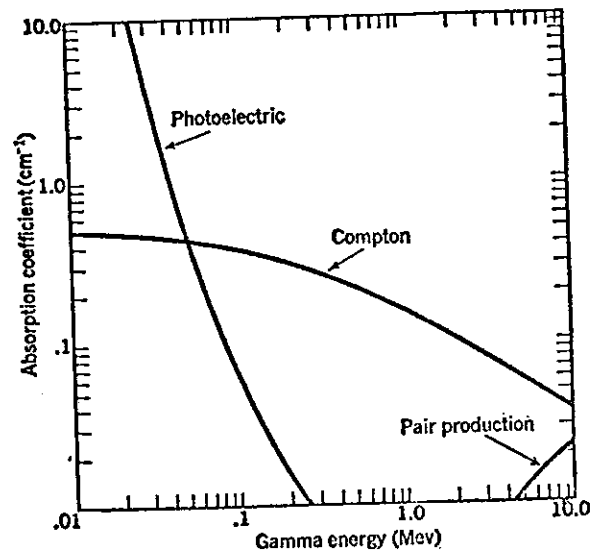


Fig. 10. Dependence on energy of absorption coefficients for aluminum.

SECTION III

Absorption Ranges in Materials

Range-distance of penetration in matter

A. Alpha Particle

$$\text{Range in air} = R = 0.309 E^{3/2} \text{ (MeV)} \quad (5)$$

Range in other absorbers: approximate

$$R = 0.173 E^{3/2} A^{1/3} \quad (6)$$

$$E = E_{\alpha}$$

A = atomic weight of absorber

$$\begin{aligned} R &= \text{thickness} \times \text{density} = \text{cm} \times \text{mg/cm}^3 \\ &= \text{range in mg/cm}^2 \end{aligned}$$

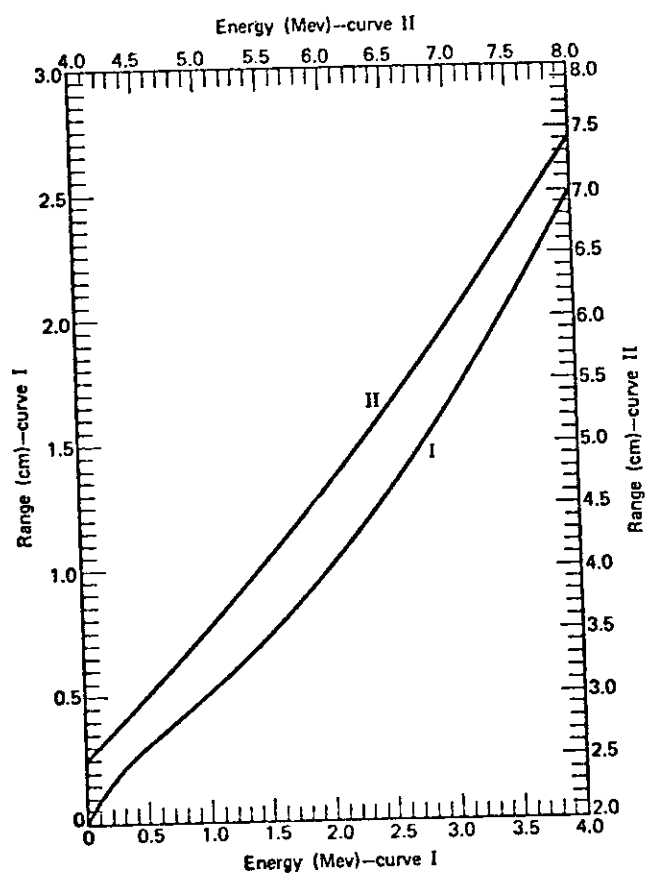


FIG. 11. Range-energy relation for alpha particles in air (15°C, 760 torr)

B. Beta Particles

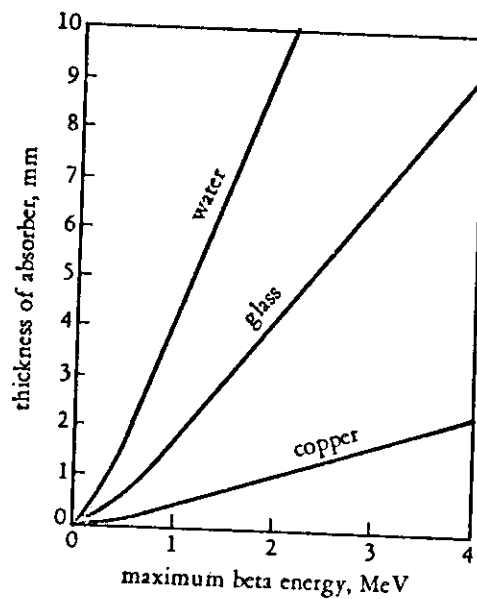


Fig. 12. Plot of absorber thicknesses required to stop beta rays completely.

C. Gamma Rays

Must consider contributions from photoelectric effect, Compton effect, and pair production.

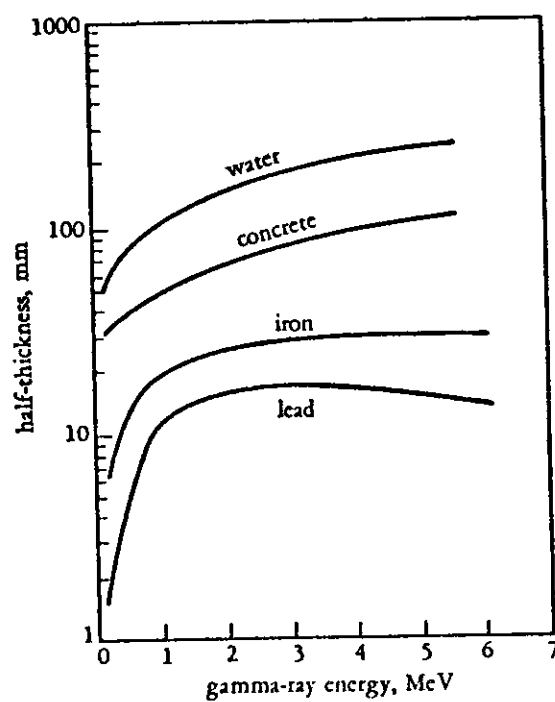


Fig. 13. Plot of absorber thicknesses required to reduce gamma ray intensity to one-half the original intensity.

SECTION IV

Radiological Safety

External Exposure - radiation source external to body

1. Alpha emitters - little or no danger, stopped by outer layer of dead skin tissue (can produce surface burn).
2. Beta emitters - can cause skin burns and biological damage.
3. Gamma and x-ray emitters - very penetrating, can damage internal organs.

Internal Exposure - introduction of radionuclide into body by ingestion, inhalation, etc.

1. Alpha emitters - bone seekers; high specific ionization can cause much radiation damage in a small volume.

2. Beta emitters - lower specific ionization, to evaluate danger you must consider particular biochemistry of the emitting element.
3. Gamma and x-ray emitters - cause damage over relatively large volume, comparable to external exposure.

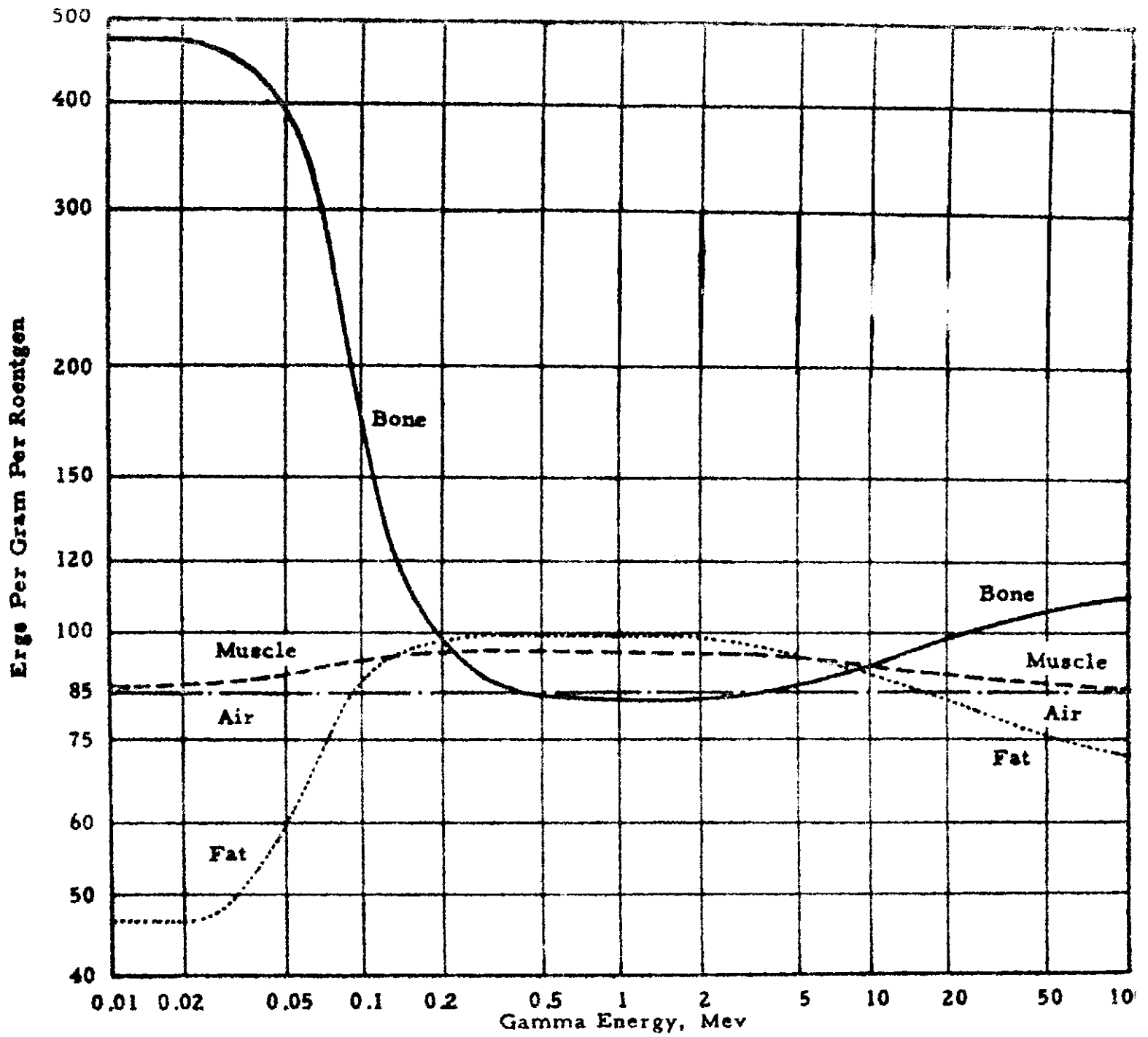


Fig. 14. Energy Absorption of gamma radiation in various tissues.

A. Units of Radiation

Since radiation produces ionization in its passage through matter, thereby losing energy, the extent of the ionization is used as a measure of the radiation field.

Roentgen \equiv quantity of γ or X-ray radiation which produces 1.16×10^{12} ion pairs in 1 gm of air.

Expresses intensity of radiation field, and not absorbed dose, limited to γ or X-ray radiation.

$$1.16 \times 10^{12} \frac{\text{ion pairs}}{\text{gm air}} = \text{absorption} \frac{87 \text{ ergs}}{\text{gm air}}$$

Rad \equiv dose of any nuclear radiation which results in the absorption of 100 erg/gm (any material)

Expresses absorbed dose, not limited to any particular type of radiation.

RBE (relative biological effectiveness) - ratio of absorbed dose of rads of γ radiation to absorbed dose of the given radiation required to give the same biological effect.

TABLE II

<u>Radiation</u>	<u>RBE</u>
x- or γ ray	1
β ray	1
Alphas	10
Slow Neutrons	5
Fast neutrons and protons (up to 10 MeV)	10

rem (roetgen equivalent man) - amount of radiation which produces the same biological effect as one rad of γ rays.

$$\text{dose (in rems)} = \text{RBE} \times \text{dose (in rads)} \quad (7)$$

B. Effect of radiation exposure

TABLE III

Effect of radiation doses on different organisms. LD-50/30 refers to a dose which has been found lethal for 50% of the organisms within 30 days. For micro-organisms about 10 times higher doses are required for killing than for inactivation.	
Micro-Organisms	<p>Enzymes inactive at $>2 \cdot 10^6$ rad</p> <p>Virus (dry) inactivated at 30 000-500 000 rad</p> <p>Bacteria inactivated at 2 000-100 000 rad</p> <p>Human cells inactivated at ≥ 100 rad</p>
Plants	<p>Flowers survive at 1 000 rad/d during the</p> <p>Trees do not survive at 100 rad/d growing season</p> <p>Trees normally survive at 2 rad/d (normally the spring)</p>
Animals	<p>LD 50/30 for amoeba 100 000 rad</p> <p>LD 50/30 for shells 20 000 rad</p> <p>LD 50/30 for hamster 900 rad</p> <p>LD 50/30 for rat 600 rad</p> <p>LD 50/30 for humans ~ 400 rad</p> <p>LD 50/30 for goat 350 rad</p>

C. Typical Values of Some Radiation Doses

TABLE IV

Sterilization of surgical supplies 2×10^9 mrem
Lethal dose (whole body).....	5×10^5 mrem
Cancer Therapy (to one region).....	5×10^5 mrem
Whole body diagnostic x-ray 2×10^4 mrem
Dental x-ray (complete).....	5×10^3 mrem
AEC limit for workers.....	5×10^3 mrem/yr
Natural background in Denver.....	130 mrem/yr
Natural background in Dallas30 mrem/yr
Chest x-ray.....	100 mrem
^{40}K in body16 mrem/yr
Living in a brick home35 mrem/yr
Living in a wooden house.....	11 mrem/yr
Radium dial watch (wrist).....	10 mrem/yr
Fallout from atomic tests.....	5 mrem/yr
Cross country jet flight (cosmic rays)...	1 mrem/yr

D. Maximum Permissible Exposure Level for Man

1. 5 rems/yr or accumulate

2. exposure of $5(N-18)$ rems

where N =age of individual

For any 3 month period, maximum = 3 rems

TABLE V

Maximum permissible amount of radioactivity in body (MPB) and concentration in air and water (MPC) for 24 h/day. τ_{eff} is the effective half-life of the radionuclide in the body. All values refer to the critical organ, i.e. the organ which has the highest tendency of accumulating the radioisotope. (According to Report of Committee II on Permissible Dose for Internal Radiation, Pergamon Press, 1960).

Nuclide	Critical Organ	τ_{eff} days	MPB μCi	MPC, $\mu\text{Ci}/\text{cm}^3$ water	air
^3H	Whole body	12	$2 \cdot 10^3$	0.05	$3 \cdot 10^{-6}$
^{14}C	Fat	12	300	$8 \cdot 10^{-3}$	10^{-6}
^{24}Na	Whole body	0.6	7	$4 \cdot 10^{-3}$	$6 \cdot 10^{-7}$
^{32}P	Bone	14.0	6	$2 \cdot 10^{-4}$	$2 \cdot 10^{-8}$
^{35}S	Testicles	76.4	90	$6 \cdot 10^{-4}$	$9 \cdot 10^{-8}$

Table V (cont.)

Nuclide	Critical Organ	τ_{eff} days	MPB μCi	MPC, $\mu\text{Ci}/\text{cm}^3$ water	air
^{42}K	Whole body	0.52	10	$8 \cdot 10^{-3}$	10^{-6}
^{51}Cr	Whole body	26.6	800	0.2	$4 \cdot 10^{-6}$
^{55}Fe	Spleen	388	10^3	$8 \cdot 10^{-3}$	$3 \cdot 10^{-7}$
^{59}Fe	Spleen	41.9	20	10^{-3}	$5 \cdot 10^{-8}$
^{60}Co	Whole body	9.5	10	10^{-3}	10^{-7}
^{64}Cu	Spleen	0.42	10	0.03	$2 \cdot 10^{-6}$
^{65}Zn	Whole body	194	60	10^{-3}	$4 \cdot 10^{-8}$
^{90}Sr	Bone	$6.4 \cdot 10^3$	2	10^{-6}	10^{-10}
^{95}Zr	Whole body	55.5	20	1	$4 \cdot 10^{-8}$
^{106}Ru	Kidneys	2.48	3	$4 \cdot 10^{-3}$	$5 \cdot 10^{-8}$
^{131}I	Thyroid gland	7.6	0.7	$2 \cdot 10^{-5}$	$3 \cdot 10^{-9}$
^{135}Xe	Whole body				10^{-6}
^{137}Cs	Whole body	70	30	$2 \cdot 10^{-4}$	$2 \cdot 10^{-8}$
^{140}Ba	Bone	10.7	4	$2 \cdot 10^{-3}$	$4 \cdot 10^{-8}$
^{144}Ce	Bone	243	5	0.08	$3 \cdot 10^{-9}$
^{198}Au	Kidneys	2.7	20	0.02	$9 \cdot 10^{-7}$
^{210}Po	Spleen	42	0.03	$7 \cdot 10^{-6}$	$2 \cdot 10^{-10}$
^{226}Ra	Bone	$1.6 \cdot 10^4$	0.01	10^{-7}	10^{-11}
^{232}Th	Bone	$7.3 \cdot 10^4$	0.04	$2 \cdot 10^{-5}$	$7 \cdot 10^{-13}$
^{238}U	Kidneys	15	$5 \cdot 10^{-3}$	$6 \cdot 10^{-4}$	$3 \cdot 10^{-11}$
^{233}U	Bone	300	0.05	$4 \cdot 10^{-3}$	$2 \cdot 10^{-10}$
^{239}Pu	Bone	$7.2 \cdot 10^{-4}$	0.04	$5 \cdot 10^{-5}$	$6 \cdot 10^{-13}$

E. Recommended working conditions

1. Care must be exercised to avoid contamination and ingestion or inhalation of radioactive material.

2. Laboratory Safety Rules:

a. No

1. Mouth pipetting
2. Smoking
3. Eating or drinking

b. Wear gloves

c. Lab benches should be covered with absorbant paper

d. When working with solids or liquids which might be inhaled use well ventilated hood or glove box

e. Exposure to personnel must be minimized by use of shielding and distance. Use of tongs and tweezers recommended in handling radioactive samples.

- f. Radioactivity must be stored in properly labeled containers under lock when not in use
- g. Monitor hands and working area frequently with both alpha and beta-gamma meters

Recommended working conditions for radionuclides of different hazards. For inexperienced personnel, 1/10 of the given values should be applied. 'A' refers to a low activity laboratory, 'B' to a semi-hot laboratory with good ventilation, and 'C' to a high activity laboratory with complete enclosure of the working area (hot laboratory).

Working Space	Radioactivity in millicuries		
	α	β	γ
A. Open laboratory bench (only if $t < 10d$ and the sample is dust free).....	0.1	0.1	0.1
B. Fumehood with stock solutions behind lead shielding.....	0.1	5	5
Fumehood with frontal shield...		5-300	5-100
C. Simple manipulator cell, 10 cm Pb.....		300	100-2 000
Advanced manipulator cell of the master-slave-type.....			2 000
Glove-boxes.....	>0.1		

F. Decontamination Procedures

- a. Work area - soak up solutions with tissues, wash area well, checking with survey meter.
- b. Personnel - for unbroken skin wash 2-3 minutes with mild soap, repeat as necessary. If necessary use solution of equal volumes of KMnO_4 (sat'd) and 0.2 N H_2SO_4 for 2 minutes, rinse with H_2O and then 5% solution NaHSO_3 for 2 minutes.

If cut, wash immediately in strong stream of water. For ingestion, treat as for toxic chemicals, induce vomiting.

SECTION V

Problems

1. If a 7.7 MeV α particle is found to have a range of 5 cm in air, estimate the number of ion pairs produced in this interaction.
2. How much energy is lost when an α particle interacts with helium to form 2×10^3 ion pairs?
3. What is the approximate range of a 6.3 MeV α particle in Al? In air?
4. Will the specific ionization per unit path length be greater for heavy particles or electrons?
5. For the interaction of electrons with matter the ratio between energy lost by radiation (such as bremsstrahlung) and ionization is given by

$$\frac{(dE/dx)_R}{(dE/dx)_I} = \frac{EZ}{800}$$

Compare the rate of energy loss by both processes for a 10 MeV electron when lead ($Z=82$) is used as the absorber.

Problems (cont.)

6. Which of the three mechanisms for the interaction of γ -rays with lead will be dominant if the energy of the γ -ray is 1 MeV? With aluminum?
7. What is the dose in rem received by a worker exposed to a field of radiation which is measured to be 50 mr for gammas? (1 mr=0.001 rad).
8. What is the total accumulated dosage (rems) recommended as the maximum permissible by age 25, 45, 60?

PART IV

Detection and Measurement of Radiation

Outline

- I. Gas Filled Detectors; Theory of Operation
 - A. Ionization Chambers
 - B. Proportional Counter
 - C. Geiger-Muller Counters
- II. Scintillation Counter; Theory of Operation
 - A. Organic Phosphors
 - B. Inorganic Phosphors
 - C. Types of Scintillation Counters
- III. Solid State Detectors
- IV. Comparison of Radiation Detection Devices
- V. Problems

SECTION I

Gas Filled Detectors: Theory of Operation

Depends on interaction of electric field of moving particle with detector material (gas) to produce ionization.

Recall: ~ 34 eV/ion pair

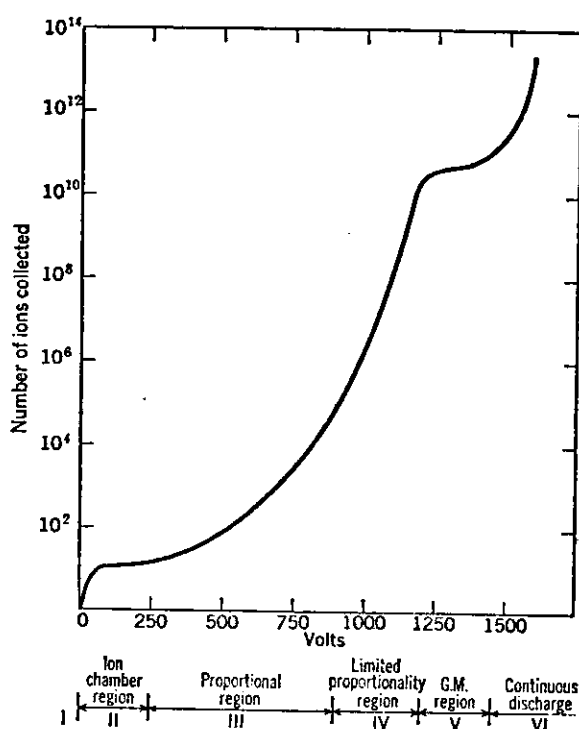


FIG. 1. The number of ion-pairs formed, as a function of detector voltage.

A. Ionization Chambers

1. Small pulse requires external amplification
2. Usually cheap and rugged

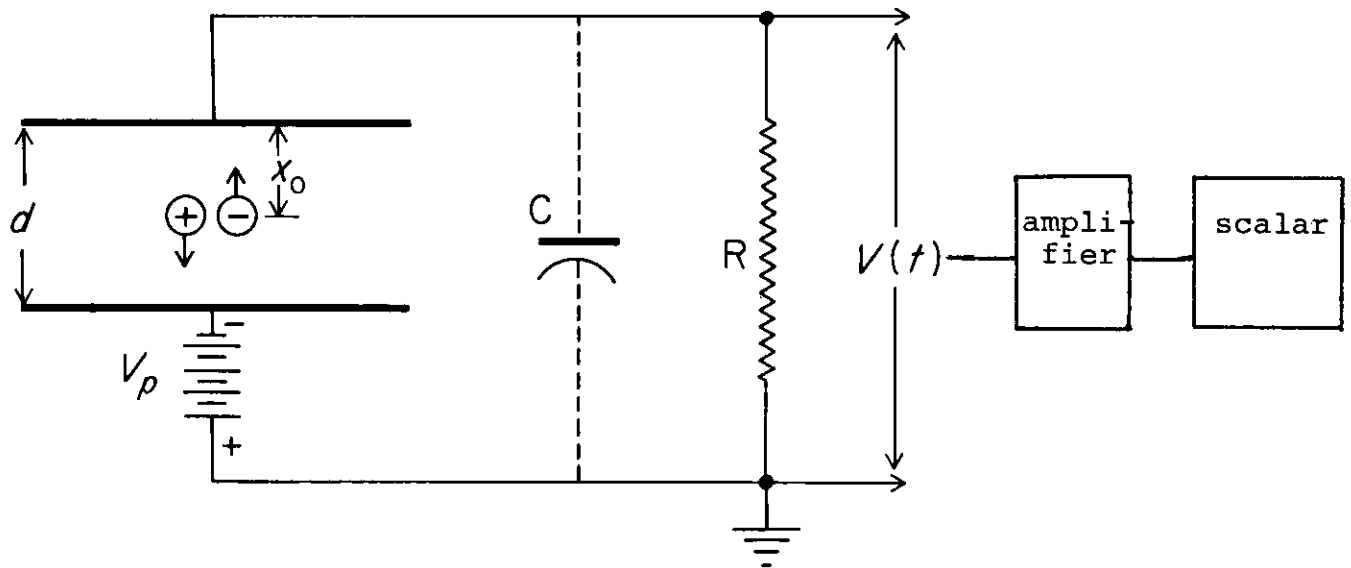


FIG. 2. Pulse-type ionization chamber.

EX: If a 4.2 MeV alpha particle dissipates all its energy ionizing the gas, how many ion pairs are formed?

$$\text{No. ion pairs} = 4.2 \times 10^6 \text{ eV} / 34 \text{ eV}$$

$$\text{Collected charge} = (1.2 \times 10^5) (1.6 \times 10^{-19}) \approx 2 \times 10^{-14} \text{ coulomb}$$

$$\text{If time of collection} = 10^{-5} \text{ sec}$$

$$\text{The average current} = \frac{2 \times 10^{-14} \text{ coulomb}}{10^{-5} \text{ sec}} = 2 \times 10^{-9} \text{ amp}$$

Important: The size of current pulse is proportional to the charge liberated and hence the amount of energy lost by particle in the gas.

B. Proportional Counters

Electric field strength at distance r from electrode is inversely related to r .

Ex: If applied voltage to cylindrical chamber of radius 1 cm is 1000 volts, the potential in vicinity of a center wire 0.001 inches in diameter $\sim 7 \times 10^4$ volts/cm. Causes acceleration of e^- 's leading to secondary ionization.

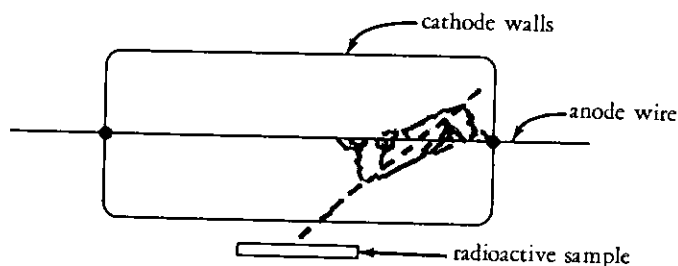


FIG. 3. Localization electron multiplication at the center anode wire in a proportional counter.

M = gas multiplication factor

$$= \frac{\text{number of electrons collected}}{\text{number of electrons in primary ionization}} \sim 10^3 - 10^5$$

Proportional counters can be constructed with or without "window". Source of radiation - outside for window, inside for windowless detectors.

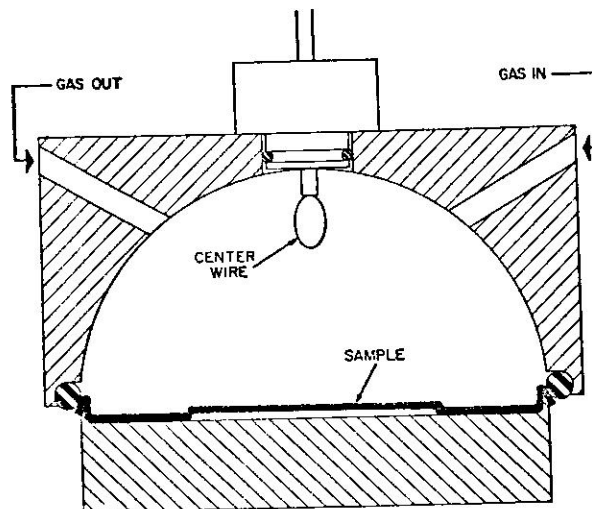


FIG. 4. Windowless flow chamber

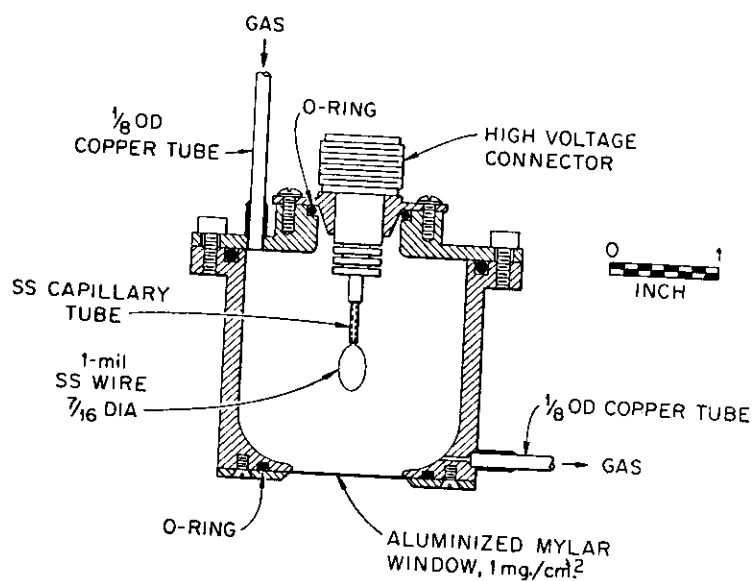


FIG. 5. End-window proportional counter for routine beta-ray counting.

Characteristics of Proportional Counters

1. Can distinguish between alpha and beta particles

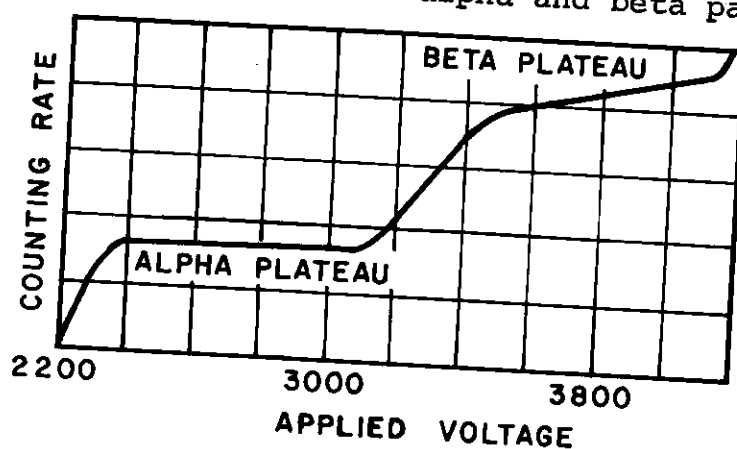


FIG. 6. A counting rate-voltage curve obtained with an end-window proportional counter and a source containing both an alpha and a beta emitter.

2. Can detect extremely high count rates

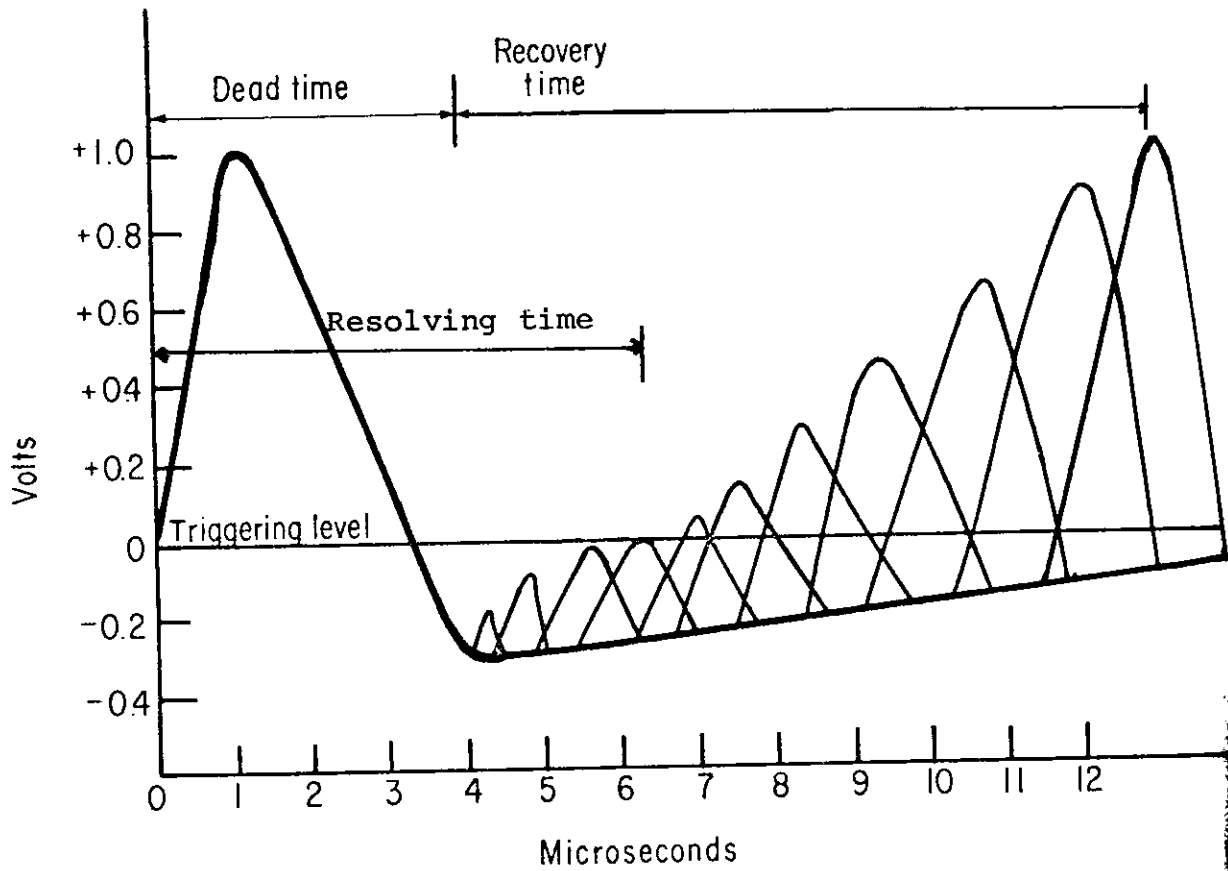


FIG. 7. Illustration of pulse shapes in a typical proportional counter system.

Dead time - time interval during which the detector is completely insensitive to additional ionizing particles.

Recovery time - that additional time beyond the dead time required for the pulse to regain its original amplitude.

Resolving time - average time interval for which the electronic detection system is insensitive (resolving time depends on the triggering level).

Resolving time determination - requires two standard sources of known activity

$$\tau = \frac{R_1 + R_2 - R_{12}}{2(R_1 R_2)} \quad (2)$$

τ = resolving time in sec

R_1 = cnt rate source 1 (cps)

R_2 = cnt rate source 2 (cps)

R_{12} = cnt rate of combined sources
1 and 2 (cps)

Typical dead time for a proportional counter 1-10 μ sec

C. Geiger-Muller Counters

1. Pulse size the same for all initial ionization whether 6 MeV alpha particle or 50 keV X-ray.
2. Large output pulse - requires less external amplification.

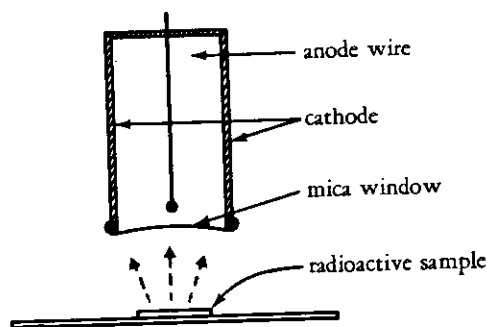
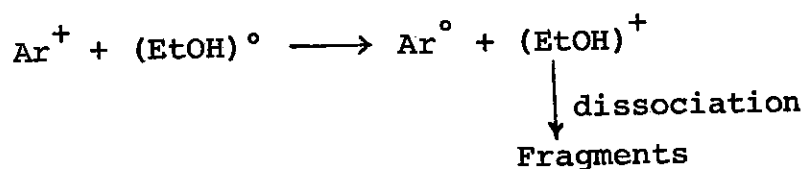


FIG. 8. Counting arrangement for an end-window Geiger counting tube.

3. Further avalanches prevented by quenching



4. Much longer dead time than proportional counter

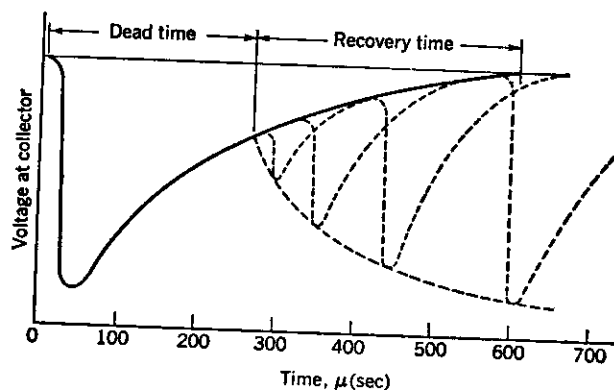


FIG. 9. Illustration of pulse shapes in a typical Geiger tube operating at a high counting rate. The dead time and recovery times are determined by the Geiger tube characteristics, but the resolving time depends on the triggering level of the electronic recording system.

5. Use primarily for beta counting

1. Alpha particles excluded by mica windows
2. Gamma rays - have too low a probability for ion pair formation per unit path length

SECTION II

Scintillation Counters: Theory of Operation

Requires the use of scintillators, organic or inorganic crystals with special properties. The nuclear radiation must be absorbed in the scintillator with subsequent re-emission of light photons.

The necessary properties of scintillators are:

1. high cross section for energy absorption
2. transparent to own radiation
3. emission of luminescent radiation must occur with high efficiency
4. radiation must have a wavelength to which the photomultiplier tube is sensitive

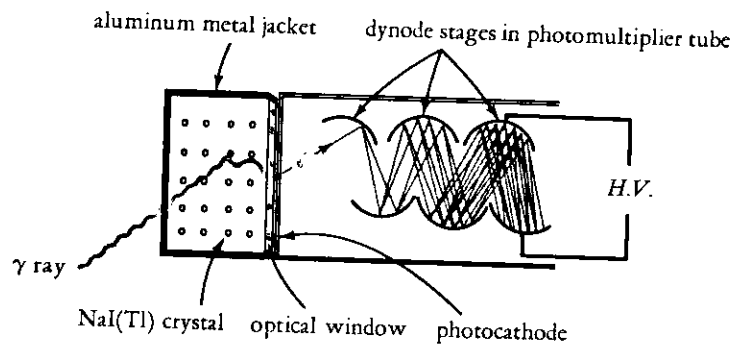


FIG. 10. A scintillation crystal with a photomultiplier tube. For simplicity, an electron multiplication (gain) of two at each dynode stage is shown; in practice, the gain is eight to ten per stage, with as many as ten dynode stages.

A. Organic Crystals (Phosphors)

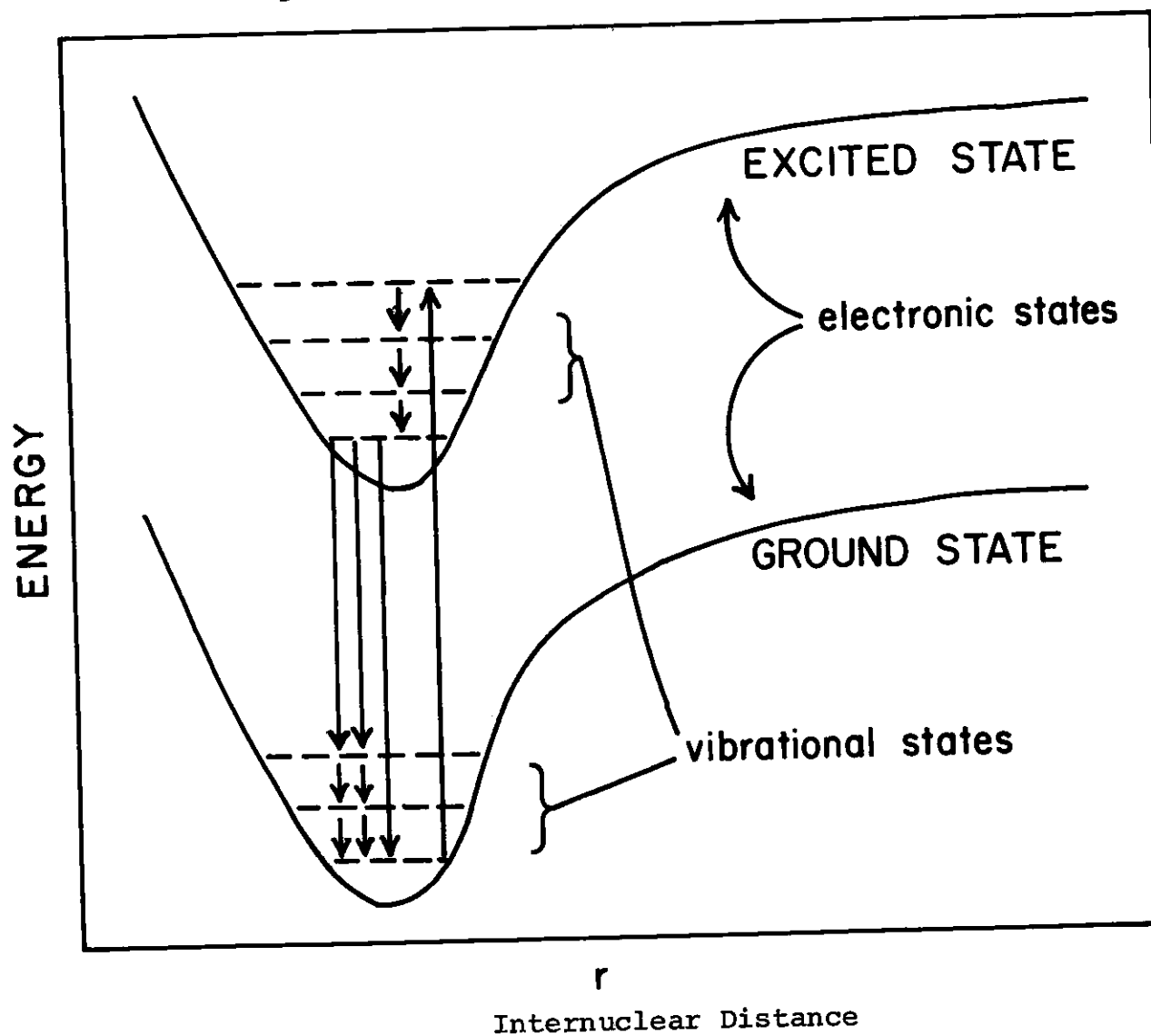


FIG. 11. Possible absorption and emission steps in phosph

Properties of Some Common Organic Phosphors

TABLE I

Material	Density (g/cm ³)	Wavelength of maximum emission (Å)	Decay time for emission	Relative pulse height from β particles
Anthracene	1.25	4400	3.2×10^{-8}	100
Stilbene	1.16	4100	6×10^{-9}	60
Plastic phosphors	1.06	3500-4500	$3-5 \times 10^{-9}$	28-48
Liquid phosphors	0.86	3500-4500	$2-8 \times 10^{-9}$	27-49

B. Inorganic Crystals (Phosphors)

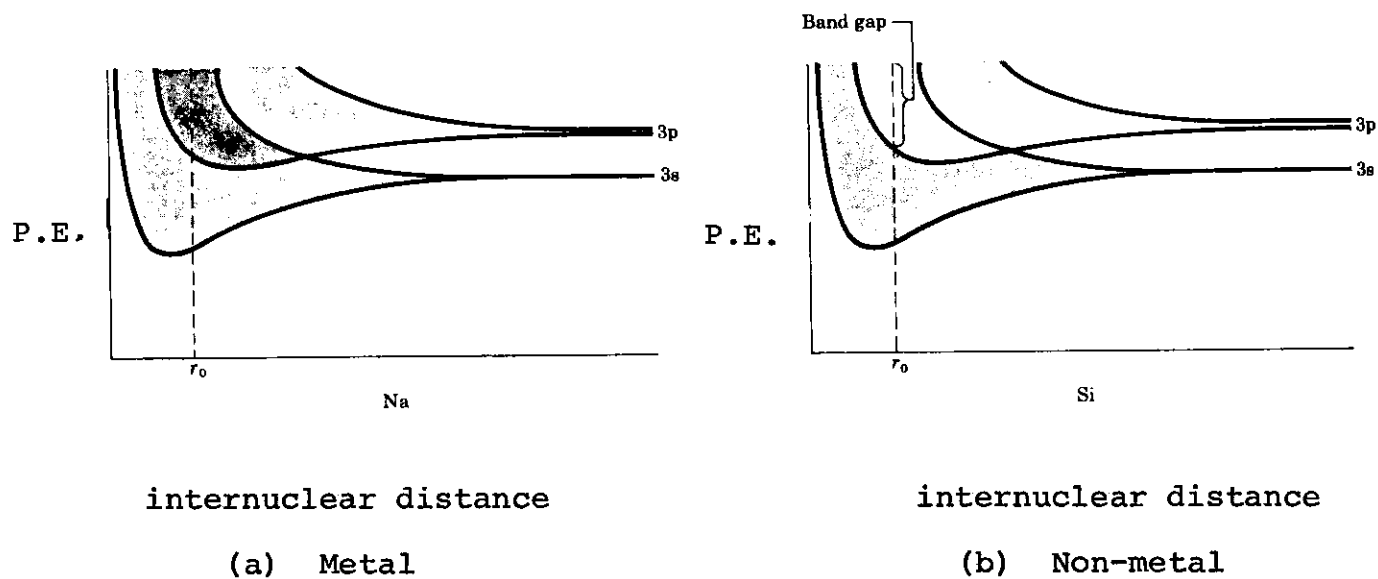


FIG. 12

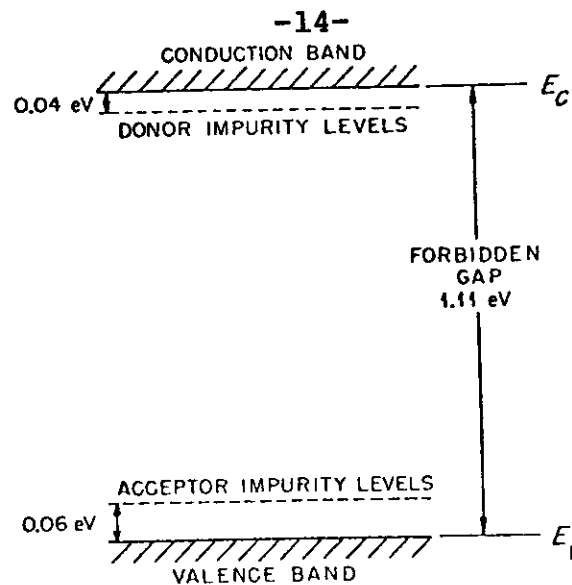


FIG. 13. Electronic Band Scheme for Non-metal.

Acceptor impurities: atoms with < 4 e's in valence shell "accept" additional e's, e.g. B in Si.

Donor impurities: atoms with > 4 e's in valence shell so donate e's; e.g. P in Si.

Properties of Some Common Inorganic Phosphors

TABLE II

Material	Density (g/cm ³)	Wavelength of maximum emission (A)	Decay time for emission (seconds)	Relative pulse height from β particles
NaI(Tl) *	3.67	4100	2.5×10^{-7}	210
ZnS(Ag) *	4.10	4500	1×10^{-5}	200

* (Tl) and (Ag) indicate small amounts of these elements added as impurity activators

C. Types of Scintillation Counters

1. Flat Solid - NaI(Tl) crystal

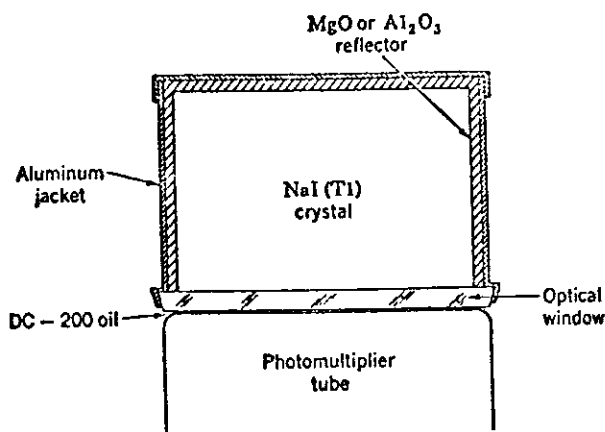


FIG. 14

2. Well Crystal

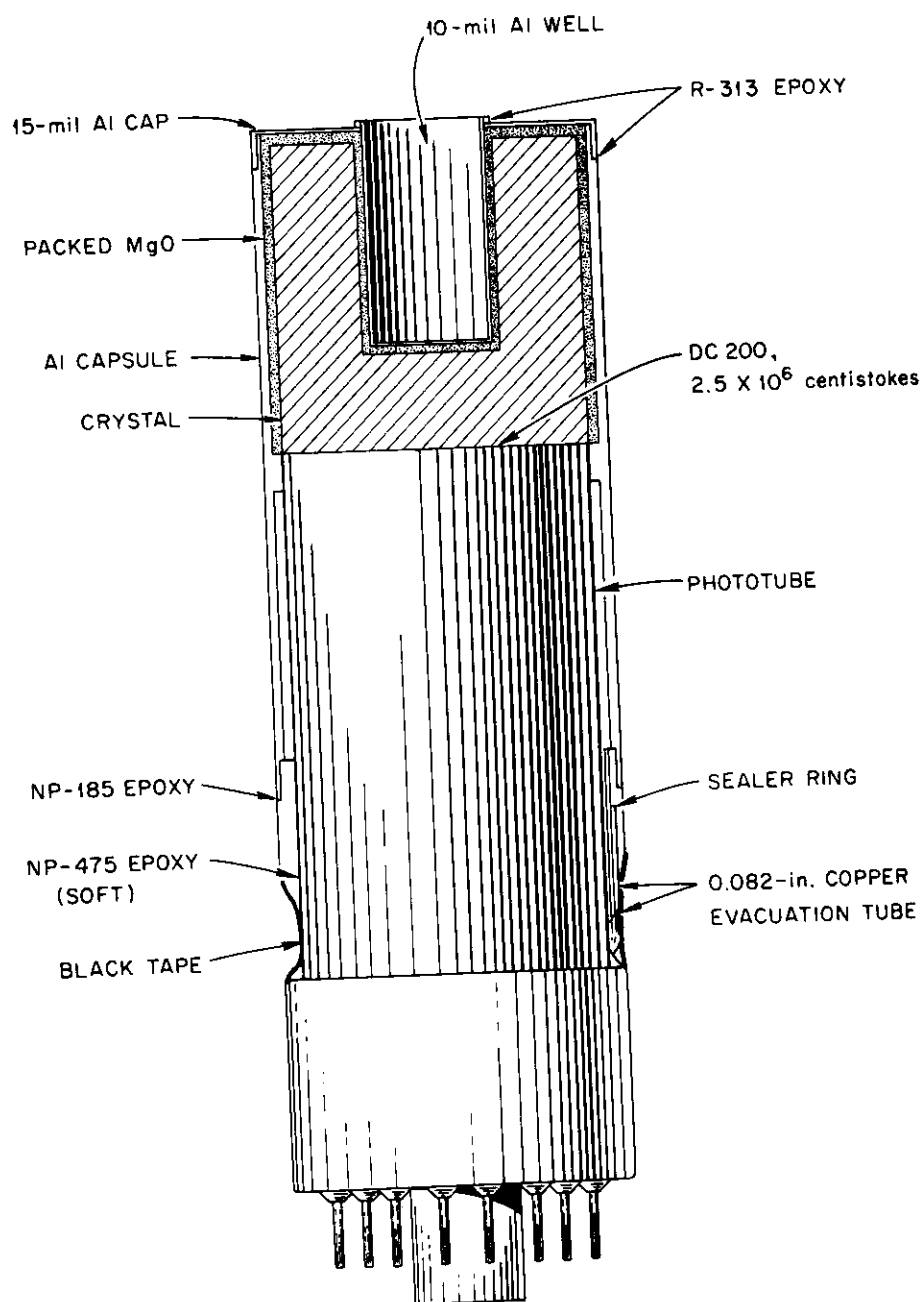


FIG. 15

C. Total Measuring Circuit

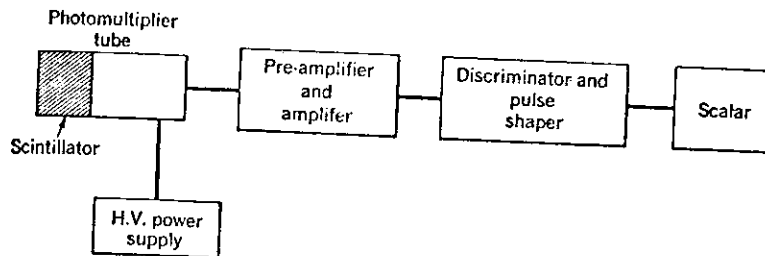
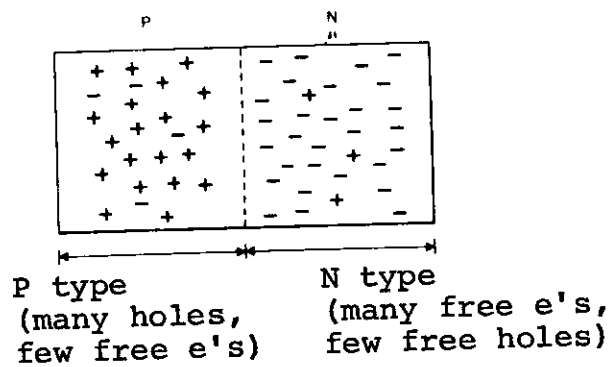


Fig. 16. Schematic diagram of a scintillation counting system.

SECTION III

Solid State Counters

(a) no bias voltage



(b) reverse bias

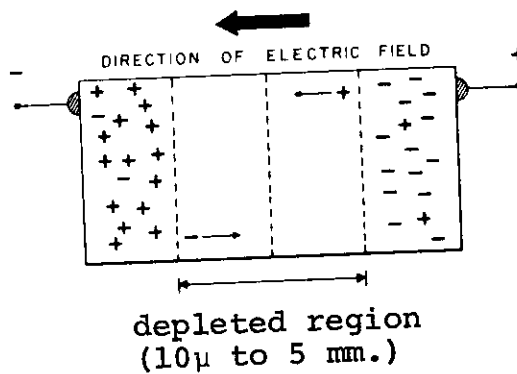


FIG. 17

SECTION IV

Comparison of Radiation Detection Devices

TABLE III

	Counter				
	Ionization chamber	Proportional	G.M.	Scintillation	Solid State
Normal detection state	gas	gas	gas	liquid or solid	solid
Radiation usually counted	α, β	α, β	α, β, γ	α, β, γ	α, β, γ
Multiplication of primary charge	1	10^4	10^8	10^8	10
Complexity of total system	medium	high	low	high	high
Particular advantages	simplicity	high count rates	simpli- city; adapta- bility	high count rates; high counting efficiency	excellent energy resolution

Problems

1. How many ion pairs are formed in a gas by a 5.4 MeV alpha particle?
2. Why does secondary ionization provide so much larger a percentage of the total ionization for beta particles than for alpha particles?
3. If the efficiency for a Geiger tube is $1 - e^{-N}$ where N is the number of primary ion pairs, calculate the efficiency for $N = 1, 2, 3, 4$, and 5. What does this tell you about the relative efficiency of Geiger-Muller counters for alpha, beta, and gamma rays (assuming they penetrate the window)?
4. The equation for the pulse size in a detector operating in the proportional region is

$$\Delta V = M \frac{ne}{c}$$

where M = gas multiplication factor, 10^4

n = number of electrons from the primary ionization

e = electronic charge, 1.6×10^{-19} coulombs

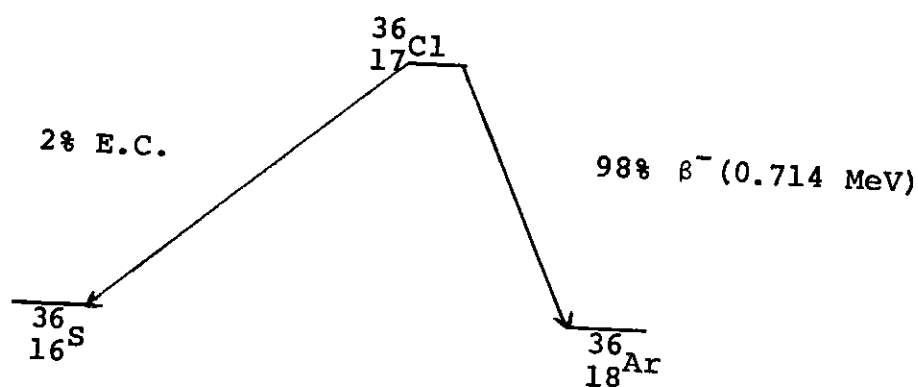
c = capacitance of the counter.

5. What is the total charge released when ^{244}Cm emits a 5.8 MeV alpha particle in an ionization chamber with air at 1 atm as the counting gas?

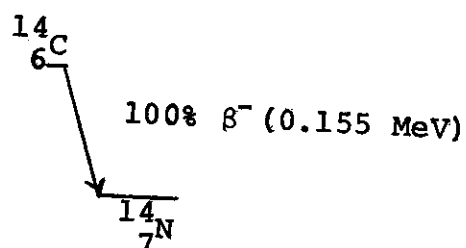
6. What is the range of a 5 MeV proton in aluminum?

7. What type of instruments would you consider best for counting each of the following and why?

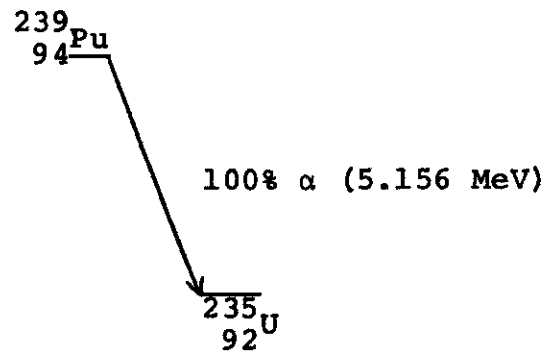
a. detection of $10^{-3} \mu\text{C}$ of $^{36}_{17}\text{Cl}$



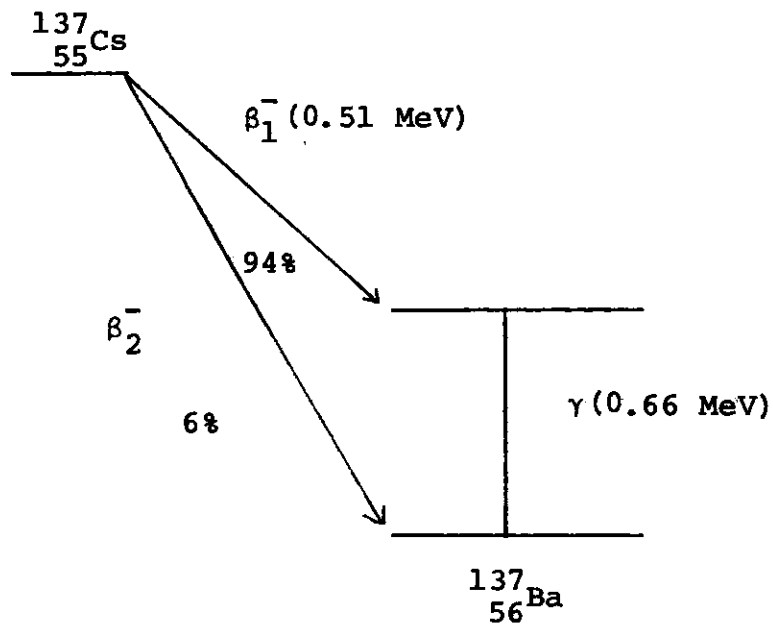
b. detection of 0.1 μC of $^{14}_6\text{C}$



c) detection of 10^{-5} μc of $^{239}_{94}\text{Pu}$



d) detection of 10^{-2} μc of $^{137}_{55}\text{Cs}$



e) What could be an alternative method of detection of each of the above radionuclides?

PART V

Counting Radioactive Samples

Outline

I. Preparation of Counting Samples

A. Purification of Counting Samples

B. Separation Techniques

1. Precipitation
2. Ion Exchange
3. Solvent Extraction
4. Chromatography
5. Distillation and Volatilization

C. Sample Preparation

1. Evaporation
2. Filtration
3. Electrodeposition
4. Sublimation
5. Liquid Samples
6. Gaseous Samples

II. Counting Efficiency

A. Detector Efficiency

1. Counting Efficiency
2. Resolving Time
3. Geometry
4. Backscattering
5. Self-absorption
6. Absorption

III. Problems

SECTION I

Preparation of Counting Samples

A. Purification - implies separation by analytical techniques of desired radionuclide from other contaminating radioactivities.

i) Choice of separation techniques depends on:

1. Tolerance level for radioactive impurities
2. Selectivity for the desired radionuclide
3. Speed
4. Volumes involved.

ii) Concentration of typical tracer level solution:

Ex: 1 ml of ^{60}Co solution (10^3 disintegrations/sec/ml)

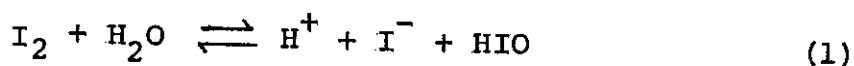
$$A = \lambda N = 10^3$$

$$N = 2.4 \times 10^{11} \text{ atoms/ml}$$

$$\text{concentration} = 4 \times 10^{-10} \text{ molar}$$

iii) Important problems due to LOW CONCENTRATION EFFECTS:

1. Adsorption of radioactive atoms on dust particles, vessel walls, pipets, etc.
2. Shift of chemical equilibrium, e.g.,



3. Differences in rates and mechanisms of reactions

iv) These problems may be avoided by adding a macro amount of:

1. isotopic carrier (stable isotope of the element being purified)
2. hold back carrier (stable isotope of the impurity)

B. Separation Techniques

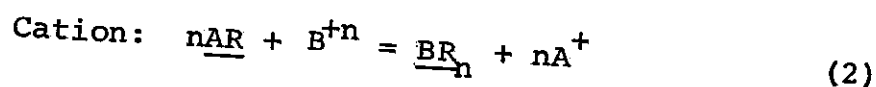
1. Precipitation - can be used to isolate desired radionuclide or to remove undesirable contamination. Technique requires the use of isotopic or hold back carriers.

Rules for maximum results:

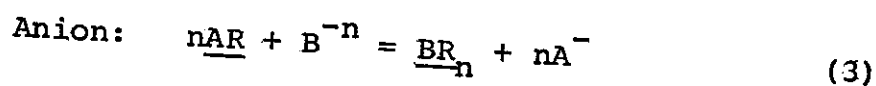
- a. slowly add relatively dilute reagent solutions to a hot solution of the radioactive sample
- b. if necessary to avoid hydrolysis, adjust pH of reagent
- c. digest solution by heating to minimize adsorption and to reduce level of trapped impurities
- d. wash carefully to avoid peptizing precipitate

- e. if practical, redissolve and reprecipitate sample
2. Ion exchange - use either cation or anion exchange resins

Equilibrium equations:



R = Dowex 50, strong acid cation exchanger



R = Dowex 1, strong base anion exchanger

- a. Effects of crosslinkage:

Crosslinkage - amount of divinylbenzene incorporated into the polystyrene matrix.

Increased cross linkage results in:

1. smaller changes in bed volume for different ionic forms and ionic strengths
2. increase in selectivity when latter is dependent on relative ionic radii
3. increase in capacity per unit volume of wet resin

4. slower kinetics of ion exchange due to decreased rates of ionic diffusion in resin

b. Effects of resin particle size

A smaller particle size results in:

1. faster attainment of equilibrium
2. increased efficiency of separation for a given volume of resin
3. a decreased flow rate for the same pressure

c. Advantages of ion exchange technique:

- i. Multi-step separation
- ii. Used for carrier free separations
- iii. Can be rapid and very selective

Disadvantages:

- i. Some experience in handling often necessary for useful results.

d. Cation Exchange

Factors effecting affinities of different cations for strong acid cation exchange resin:

- i. In dilute solutions, the affinity increases with increasing crystal radius of cation,
 $\text{Cs}^+ > \text{Rb}^+ > \text{K}^+ > \text{Na}^+ > \text{Li}^+$

- ii. In concentrated solution, selectivity may decrease and affinity order may reverse
- iii. The higher the charge on the cation the greater the affinity.

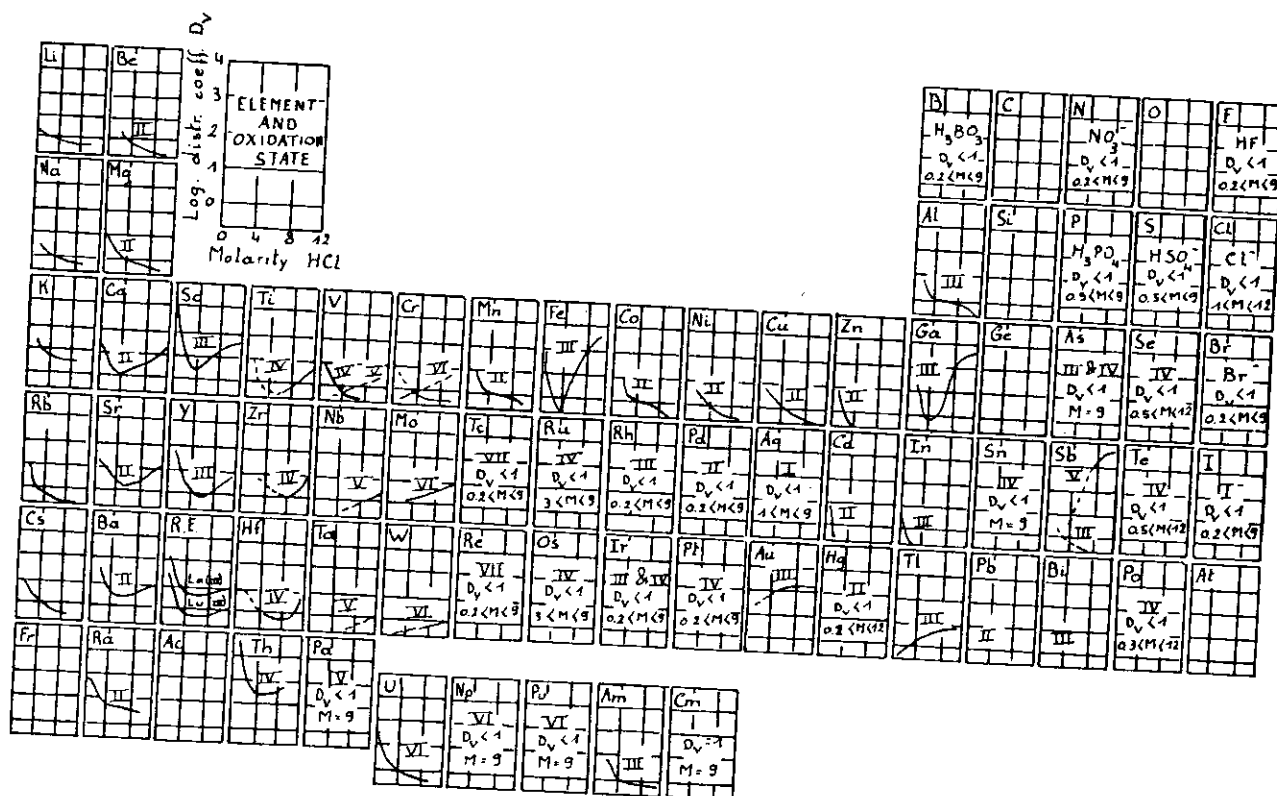
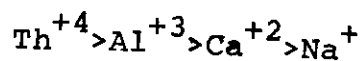


FIG. 1. Absorption of the element from HCl solutions by Dowex 50 x 4. (Nelson, Nurase, Kraus, J. Chromat. 13, 503 (1964).

e. Anion exchange

Order of selectivity:

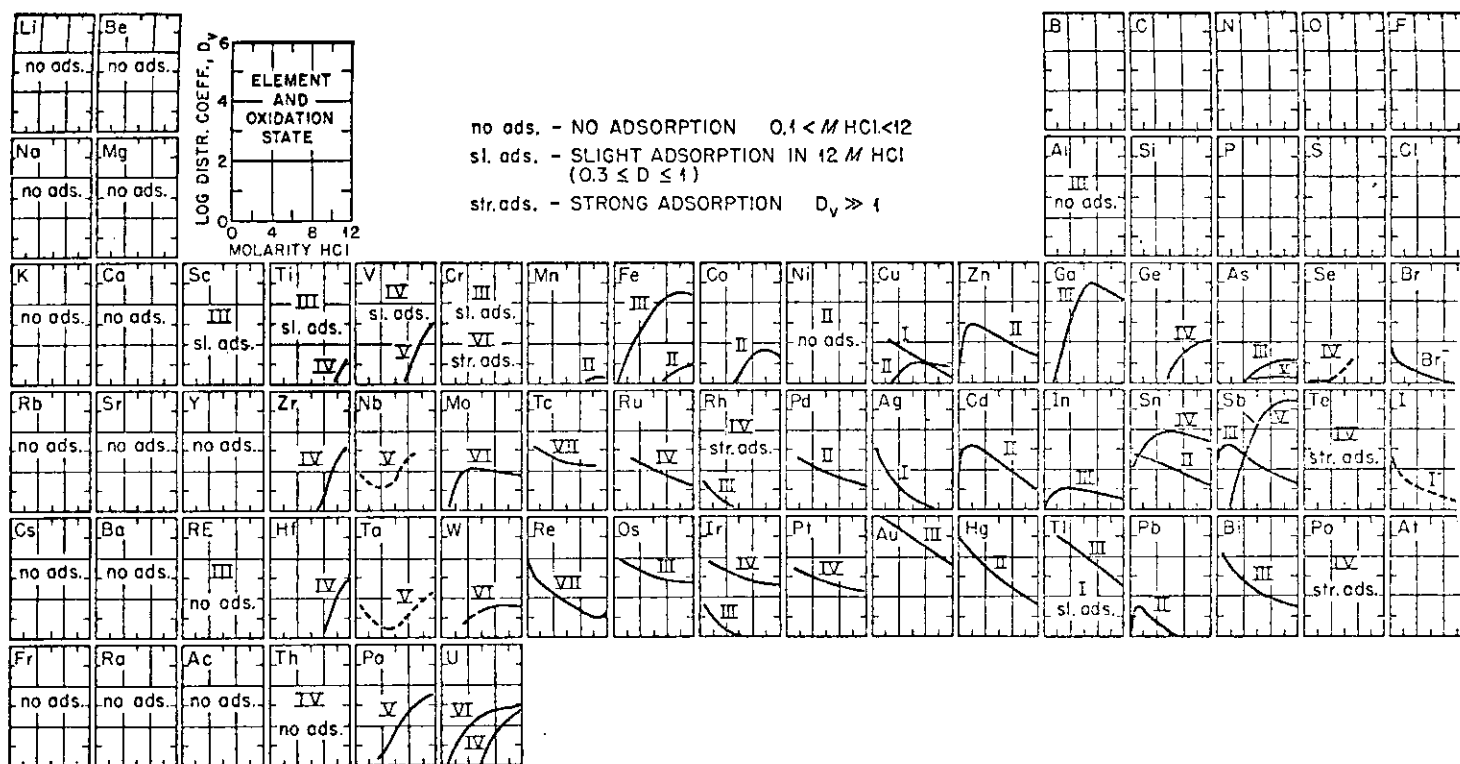
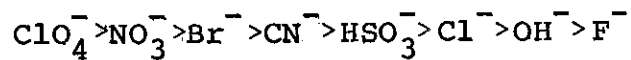


FIG. 2. Adsorption of the elements from hydrochloric acid by Dowex 1. Kraus and Nelson, Paper 837, Geneva Conf. Vol. 7 (1956).

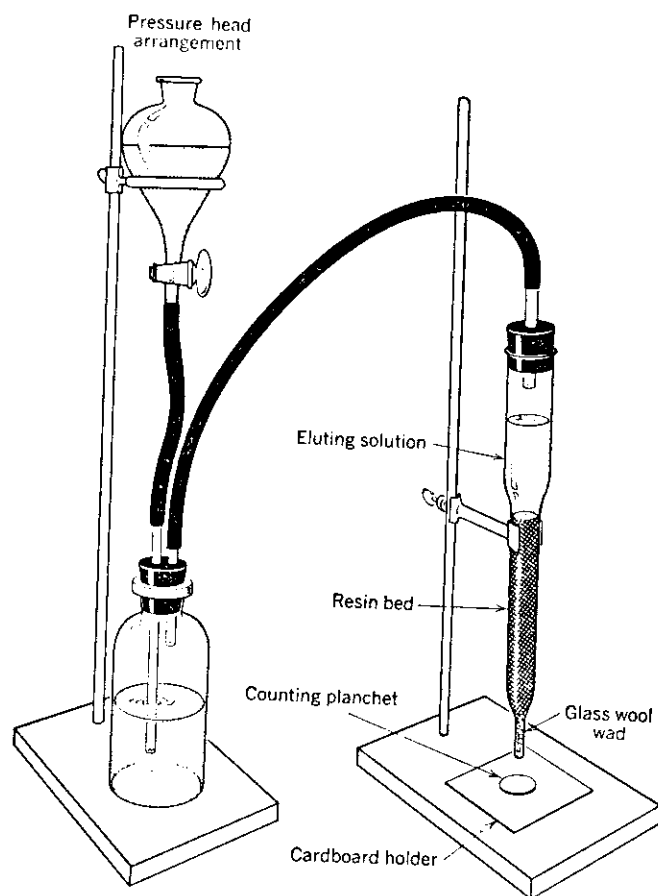


FIG. 3. Ion exchange elution column and pressure head.

3. Solvent Extraction - very similar to ion exchange.

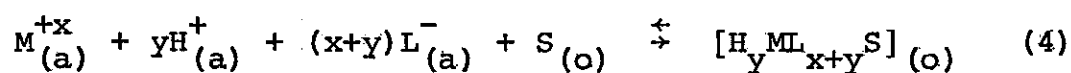
The desired metal ion is distributed between two immiscible liquid phases, usually water and

an organic solvent. The organic solvent may itself act as an extracting agent or the organic phase may consist of an extracting agent dissolved in an organic solvent.

a. Some typical solvent extraction systems

Cationic type:

- i. Extraction by an unionized organic solvent containing oxygen donor atoms - no diluent



Where M^{+x} - metal ion of charge x

H^{+} - hydrogen ion

L^{-} - monobasic ligand such as NO_3^{-}

S - organic solvent molecule acting as extractant

(a) - aqueous phase

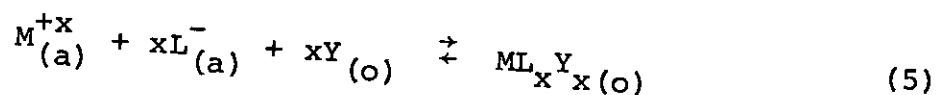
(o) - organic phase

Examples of S: diethyl ether

methy isobutyl ketone (MIBK)

isopropyl alcohol

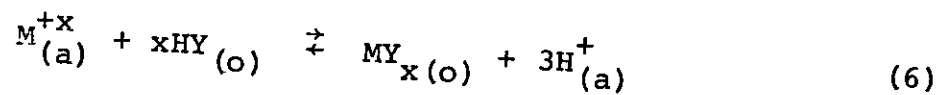
ii. Neutral phosphorous-based extractant dissolved in hydrocarbon diluent



Y - extracting agent

Ex: tributyl phosphate (TBP)

iii. Acidic extractants with oxygen donors dissolved in hydrocarbon diluent; usually extract through chelate formation



HY - acidic extractant

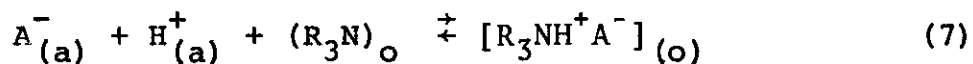
Ex: thenoyltrifluoroacetone (TTA)

di(2-ethylhexyl)phosphoric acid (HDEHP)

b. Anionic type:

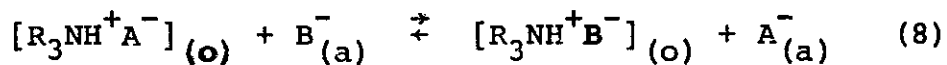
Amine extractants dissolved in a hydrocarbon diluent. Can be primary, secondary, or tertiary, amine or quaternary ammonium salts; Usually a high molecular weight amine.

Eqn. for tertiary amine (R_3N):

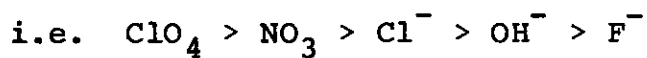


A^- = anion of an acid such as NO_3^- or a metal complex such as $FeCl_4^-$

$R_3NH^+A^-$ can undergo ion exchange:



Order of selectivity similar to that of anion exchange resins:



Advantages of solvent extraction:

- a. High selectivity
- b. Useful for carrier free separations
- c. Rapid, often faster than ion exchange
- d. Technique easier than ion exchange
- e. Wide variety of solvent extraction systems

Disadvantages:

- a. Single step separation

4. Chromatography

- a. Paper Chromatography - partition between a stationary aqueous phase supported by hydrophilic paper and a mobile organic phase.
- b. Electromigration - separation due to different velocities of ion under an electric field gradient. Both the solid and liquid phases are stationary with the liquid phase acting as a medium through which the ions migrates.
- c. Reversed-phase Chromatography - Solid support holds the organic phase as the stationary phase while the aqueous phase is mobile. Commonly

used as a column chromatographic technique in place of conventional ion exchange resins.

Ex: Extracting agent such a HDEHP absorbed on small glass beads (200-400 mesh), made into a column and separation on column by acidic eluant as in cation resin elution.

5. Distillation and Volatilization

- a. Distillation - similar to conventional method even at carrier free level.

TeCl_4 , SbH_3 , OsO_4 , I_2 are some compounds which can be distilled

Volatilization - when carrier free tracer is placed on surface and the temperature is raised, volatilization may begin below boiling point. Over narrow temperature range 90% or more of radioactive atoms are evolved from the surface.

C. Sample Preparation

1. Solution evaporation: good for small volumes of dilute or carrier free solutions. Solution spreads uniformly over counting planchet and solvent is evaporated under infrared lamp taking care to prevent splattering and consequent contamination of surrounding work area.

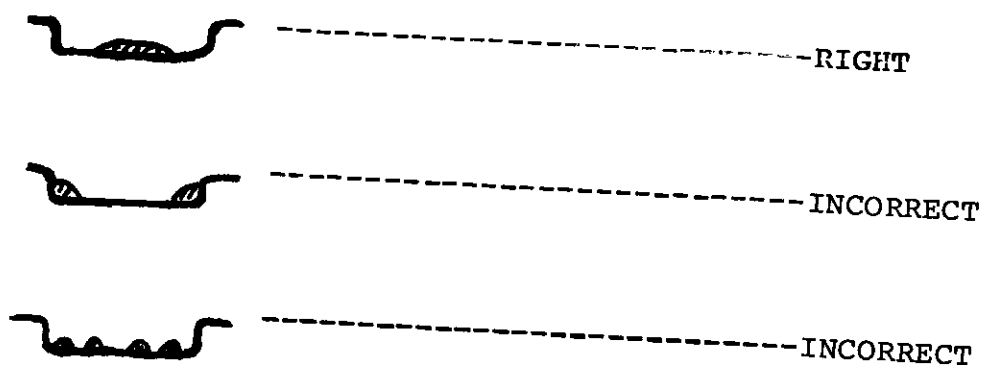


FIG. 4. Sample preparation by evaporation

2. If we have precipitation of carrier and radio-activity, the precipitate can be affixed to a counting pad by filtration. Pad then transferred to counting planchet.

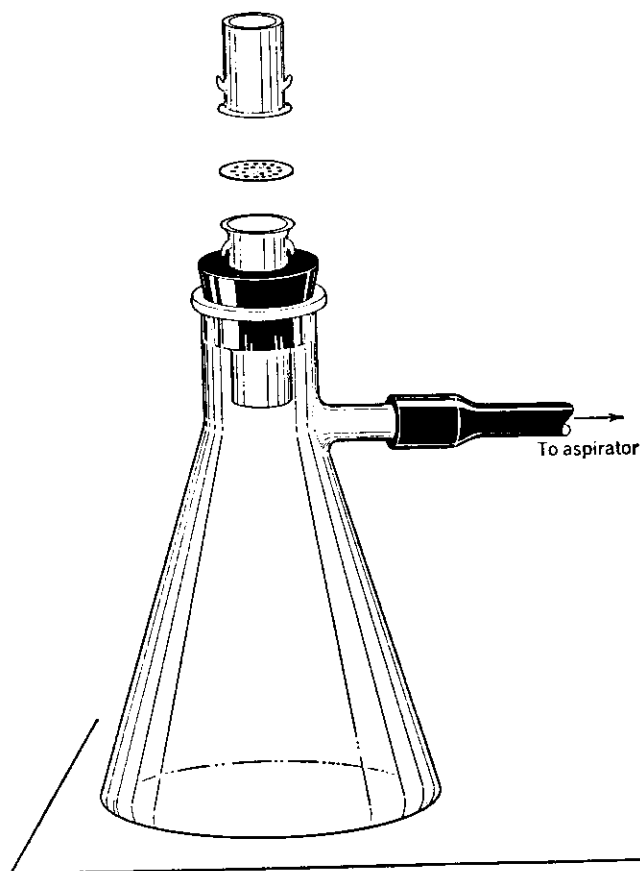


FIG. 5. Filter assembly for obtaining uniform deposits.

3. Electrodeposition: the desired cation is either deposited in metallic state or precipitated in an insoluble form on the electrode (usually a thin foil).

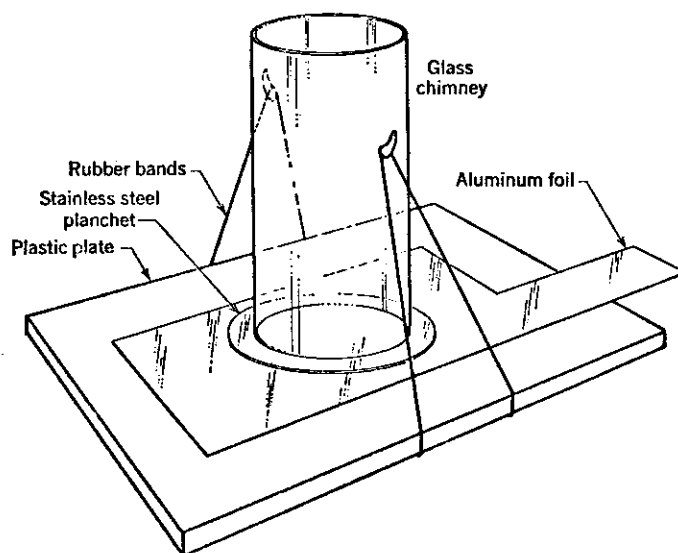


FIG. 6. Experimental arrangement for electrodeposition

4. Sublimation: elements or compounds are sublimed at high temperatures in a vacuum from a filament onto a counting planchet.
5. Liquid Samples: may be counted directly by liquid scintillation technique. Can also count gamma ray emitting radionuclides in a liquid sample using a NaI(Tl) well counter.
6. Gaseous Samples: usually avoided due to difficulties in handling.

SECTION II

Counting Efficiency

The following considerations are mainly for β emitting samples although in principle apply also to α and γ emitters.

A. Detector Efficiency = C

$$C = \frac{R \text{ (cpm)}}{R \text{ (dpm)}} = \frac{\text{measured count rate}}{\text{absolute disintegration rate}} \quad (9)$$

Also

$$C = f_e \cdot f_\gamma \cdot f_g \cdot f_b \cdot f_s \cdot f_a$$

1. f_e : counting efficiency of tube; $f_e = 1$ for good GM tube

2. f_γ : resolving time correction

Determine the resolving time τ for your particular counter system

Correction:

$$R_t = \frac{R_o}{1 - R_o \tau} \quad (10)$$

R_t = true cnt rate (cps)

R_o = observed cnt rate (cps)

τ = resolving time (sec)

3. f_g : geometry factor

4. f_b : back scattering factor; increases with increasing thickness of sample up to saturation or increases with z of sample backing material

5. f_s : self absorption factor

$$f_s = \frac{1}{\mu s} (1 - e^{-\mu s}) \quad (11)$$

s = sample thickness (mg/cm^2)

μ = absorption coefficient

$$\mu = \frac{22}{E^{1.33}} \text{ (cm}^2/\text{mg)}$$

E = energy of beta particle

6. f_a : absorption factor, includes absorption and scattering by sample covering, by air between sample and window, and by window.

NOTE: Counters are commonly calibrated by the use of SRM's (standard reference material) available from the National Bureau of Standards and from commercial suppliers. Rarely are the individual factors of Section II determined. Using the same planchets, sample position and counter all factors are invariant except the resolving time correction (a function of activity level) and the self absorption correction (a function of sample thickness).

SECTION III

Problems

1. Suggest one way of separating radioactive iodine from radioactive phosphate? [Hint: What would be the effect of the addition of AgNO_3 to a solution containing these two anions?]
2. A solution containing $^{210}\text{PbSO}_4$ ($t_{1/2} = 22$ yrs) was found to have a disintegration rate of 10,000 dpm per ml.
 - a. Can you precipitate the radioactive lead by the addition of 10 M SO_4^{-2} given the following information?
$$K_{sp} = [\text{Pb}^{+2}][\text{SO}_4^{-2}] = 1.1 \times 10^{-8}$$
 - b. How much inactive Pb carrier would be necessary to precipitate 99% of the ^{210}Pb in a 10M sulfate solution?
3. Twenty milliliters of a solution (solvent 1) containing 10^{-2} grams of iodine containing 25,000 dpm of I ($t_{1/2} = 8.07\text{d}$) is equilibrated with 10 ml of an immiscible solvent (solvent 2). Using the distribution coefficient K,

$$K = \frac{\text{conc. of solute in solvent 1}}{\text{conc. of solute in solvent 2}} = 20$$

calculate the weight of iodine in each solvent at equilibrium,

4. Using the results of problem 3, calculate the count rate in cpm of ^{131}I in each solvent assuming the efficiency of your counter is 7%.
5. What is the requirement for the purification of two substances by distillation whether they are radioactive or not?
6. Suggest a method for the separation of the following nuclides $^{40}\text{K}^+$, $^{232}\text{Th}^{+4}$, $^{140}\text{La}^{+3}$ and $^{131}\text{Ba}^{+2}$.
7. Two electrodes are placed in an aqueous solution of the radioactive metal chlorides of Zn, Fe, and Sn. The voltage which must be applied across the electrodes to cause deposition is

$$E_{\text{applied}} = E_{\text{cathode}} - E_{\text{anode}} + W$$

where W is the overvoltage. Given the following information, what are the possible cathode reactions? What is the anode reaction? Assuming $W = 0.2\text{V}$, at what applied voltages will these three metals be deposited?

	[E°(V)]
$1/2 \text{ Zn} \longrightarrow 1/2 \text{ Zn}^{+2} + \text{e}^{-}$	0.76
$1/2 \text{ Fe} \longrightarrow 1/2 \text{ Fe}^{+2} + \text{e}^{-}$	0.44
$1/2 \text{ Sn} \longrightarrow 1/2 \text{ Sn}^{+2} + \text{e}^{-}$	0.14
$1/2 \text{ H}_2\text{O} \longrightarrow 1/4 \text{ O}_2 + \text{H}^{+} + \text{e}^{-}$	-1.23
$\text{Cl}^{-} \longrightarrow 1/2 \text{ Cl}_2 + \text{e}^{-}$	-1.36

PART VI

Counting Statistics and Calibration

Outline

I. Counting Statistics

A. Definition of Errors

B. Distribution of Random Events

1. Poisson Distribution
2. Gaussian Distribution

- a. Standard Deviation for Total Count
- b. Standard Deviation for Counting Rate
- c. Confidence Levels and K

C. Error in Total Counting System

1. Standard Deviation for Sample Alone
2. Optimization of Counting System
3. Dead Time Corrections
4. Corrections for Low Levels of Radioactivity
5. Calculation for Preset Time and Preset Count
6. Relationship of Net Sample Count Rate to Background Count Rate
7. Choice of Counting Technique
8. Error Due to Electronic or Mechanical Misbehavior
 - a. $K > 3.5$
 - b. Chi-square Test

II. Radioactive Standards and Calibration

- A. Determination of Absolute Counting Rate
- B. Standard and Calibration Services

III. Problems

SECTION I

Counting Statistics

A. Definition of Errors

Determinate Error - usually systematic and constant.
Can usually be eliminated by redesign of experiment.

Random error - neither systematic or constant. Must
be evaluated by statistical methods.

Table 1 Notation

\bar{n} = arithmetic mean of all the measured values

n = a measured value of total counts

$P(n)$ = probability of occurrence of the value n

σ = standard deviation

p = probable error

R = count rate in counts per minute

t = time of measurement

N = number of measurements on a given sample

Subscripts

s = sample only

b = background

t = total (sample + background)

B. Distribution of Random Events

Binomial Distribution Law - correctly expresses probability but very difficult to use. Common practice is to use:

1. Poisson Distribution

$$P(n) = \frac{\bar{n}^n e^{-\bar{n}}}{n!} \quad (1)$$

A Poisson Distribution is not symmetric about \bar{n} , the average value, for low values of \bar{n} . See Fig. 1.

2. Gaussian Distribution

$$P(n) = \frac{1}{(2\pi\bar{n})^{1/2}} e^{-\frac{(\bar{n}-n)^2}{2\bar{n}}} \quad (2)$$

A Gaussian Distribution is symmetric about \bar{n} .

Note: If $\bar{n} \geq 100$ the Gaussian function may be substituted for the Poisson Distribution with no appreciable error.

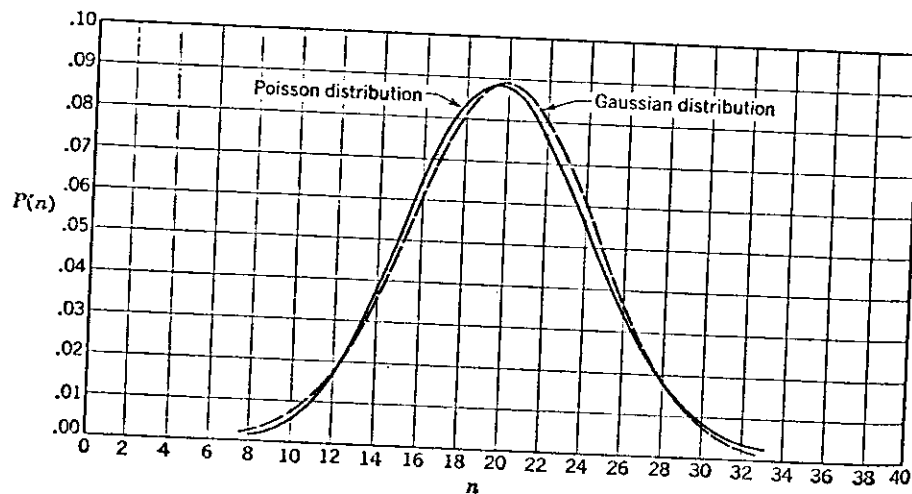


FIG. 1. The Poisson and Gaussian distributions for $\bar{n} = 20$.

- a. Standard Deviation (σ) for total count using Gaussian Distribution

$$\sigma = \left[\frac{1}{N} \sum_{i=1}^N (\bar{n} - n_i)^2 \right]^{\frac{1}{2}} = n^{\frac{1}{2}} \quad (3)$$

Note: σ corresponds to a 68% probability

Total count = rate x total time

Ex: Gross sample count rate = 1000 cpm

total time = 10 m

Total Count = 1000 cpm x 10 m = 10,000 counts

$$\sigma = (n)^{1/2} = (10,000)^{1/2} = \pm 100$$

Therefore: Counts $\pm \sigma = 10,000 \pm 100$ counts

or 9900 - 10,100 counts range for 68% probability.

Consequently, if this sample is counted 100 times, 68 times the total count will be between the limits of 9900-10,100 counts and 32 times, outside of these limits.

b. Standard Deviation (σ_R) in the counting rate (R)

$$\sigma_R = \frac{(n)^{1/2}}{t} = \left(\frac{R}{t}\right)^{1/2} \quad (4)$$

σ_R for previous example:

$$\frac{(10,000)^{1/2}}{10} = \frac{1000}{10} = \pm 10 \text{ cpm}$$

c. Confidence Levels and K

Confidence level - expresses the % probability of the true value occurring within certain limits

$$K = \text{number of standard deviations} = \frac{|\bar{n}-n|}{\sigma}$$

Table II. Table of Constants of Relative Error

Confidence Level (%)=(b)	K	
50	0.6745	Probable Error
68.27	1.0000	Standard deviation (1σ)
90.00	1.6449	Nine tenths Error
95.00	1.9600	Ninety-five hundredths error
95.45	2.0000	2σ
99.00	2.5758	Ninety-nine hundredths error
99.73	3.0000	3σ
99.9937	4.0000	4σ

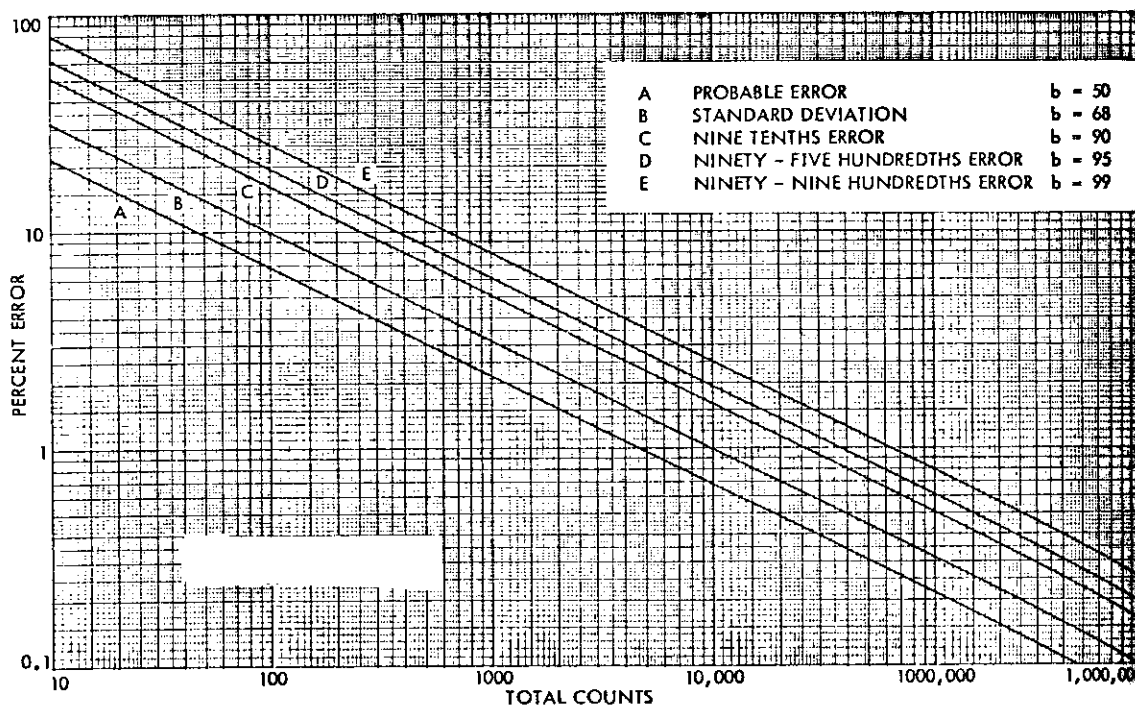


FIG. 2. Percent Error versus Gross Count

To determine the limits, Δn , corresponding to a certain confidence level for a total count of n

$$\Delta n = K_i \sigma = K_i (n)^{1/2} \quad (5)$$

where K_i is the K corresponding to the particular confidence level

Ex: total count $n = 10,000$

For probable error (or 50% confidence level)

$$\Delta n = 0.675 (n)^{1/2} = 0.6745 (10,000)^{1/2} = \pm 67$$

For 95% confidence level

$$\Delta n = 1.96 (n)^{1/2} = (1.96) (10,000)^{1/2} = \pm 196$$

C. Error in Total Counting System

Total count = sample count + background count

\therefore Sample count = total count - background count

1. (a) Standard deviation for total sample alone.

$$\sigma_s = (\sigma_t^2 + \sigma_b^2)^{1/2} = (n_t^2 + n_b^2)^{1/2} \quad (6a)$$

Frequently $\sigma_b \ll \sigma_t$

$$\therefore \sigma_s = (\sigma_t^2)^{1/2} = \sigma_t \quad (6b)$$

(b) Standard deviation for sample counting rate alone.

$$\sigma_{R_s} = (\sigma_{R_t}^2 + \sigma_{R_b}^2)^{1/2} = \left(\frac{R_t}{t_t} + \frac{R_b}{t_b} \right)^{1/2} \quad (7a)$$

Frequently $\sigma_{R_b} \ll \sigma_{R_t}$

$$\therefore \sigma_{R_s} = \left(\sigma_{R_t}^2 \right)^{1/2} = \sigma_{R_t} \quad (7b)$$

3. Optimization of counting conditions.

$$\frac{t_b}{t_t} = \left(\frac{R_b}{R_t} \right)^{1/2} \quad (8)$$

Ex: Suppose you are using a counter with a background count rate of 50 cpm and are only allotted one hour to count your sample which has a gross count rate of 2000 cpm. How should you proportion your allotted time between sample and background for minimum error?

$$R_b = 50 \text{ cpm} \quad R_t = 2000 \text{ cpm} \quad t_t = 60 \text{ min}$$

$$\frac{t_b}{t_t} = \left(\frac{R_b}{R_t} \right)^{1/2} = \left(\frac{50 \text{ cpm}}{2000 \text{ cpm}} \right)^{1/2}$$

$$\frac{t_b}{60 \text{ min}} = .158$$

$$t_b = 9.48 \text{ min.}$$

∴ You must take a 9.5 min background count and use the remaining time for sample counting (50.5 min).

4. Error in dead time corrections.

For GM counters with count rates greater than 8000-10,000 cpm or proportional counters with count rates greater than 10^5 cpm

$$\sigma_m = (n)^{1/2} \left(\frac{m}{n}\right) \quad m = \text{true count rate} \quad (9)$$

5. Calculations for minimum count rates.

$$R_{s(\min)} = \frac{1 + 2F_\sigma (tR_b)^{1/2}}{F_\sigma^2 t} \quad (10)$$

where $F_\sigma = \frac{\sigma_{R_s}}{R_s}$ and t is total time

Ex: Suppose you are using a counter (50 cpm background) to count your sample ($R_t = 200$ cpm) and given only one hour to complete the counting. For a desired fractional standard deviation of 1% what is the minimum sample count rate necessary?

$$F_Q = 0.1 \quad R_b = 50 \text{ cpm} \quad t = 60 \text{ min}$$

$$R_{s(\min)} = \frac{1+2(.01)(60 \text{ min} \times 50 \text{ cpm})^{1/2}}{(0.1)^2 (60 \text{ min})}$$

$$R_{s(\min)} = 349 \text{ cpm}$$

Is your experiment valid?

No, you do not have enough sample to count for your allotted time to give you 1% error. You need a sample which has a minimum counting rate of 349 cpm and your sample is only 150 cpm.

$$(R_s = 200 \text{ cpm} - 50 \text{ cpm})$$

6. Calculation for preset time (pst) or preset count (psc) for a predetermined accuracy.

$$\text{pst} = \frac{K^2 \times 10^4}{F^2} \times \frac{1}{R_s} \left[1 + \frac{2R_b}{R_s} \right] \quad (11)$$

$$\text{psc} = R_t \times \frac{K^2 \times 10^4}{F^2} \times \frac{1}{R_s} \left[1 + \frac{2R_b}{R_s} \right] \quad (12)$$

F = desired error as percentage

Ex: Suppose you had a total count rate of 600 cpm and the background rate was 50 cpm.

How long would you have to count your sample to obtain a 2% accuracy at 99% confidence level?

$$R_s + R_t - R_b = 600 \text{ cpm} - 50 \text{ cpm} = 550 \text{ cpm}$$

K (for 99% confidence level from Table II) = 2.5758; F = 2

$$p_{st} = \frac{(2.5758)^2 \times 10^4}{(2)^2} \times \frac{1}{550} \left[\frac{1 + 2(50)}{550} \right]$$

$$p_{st} = 35.6 \text{ m}$$

Suppose you counted the same sample to a preset count of 36,000, what would your percent accuracy be at a 95% confidence level?

$$p_{sc} = R_t \times \frac{K^2 \times 10^4}{F^2} \times \frac{1}{R_s} \left[1 + \frac{2R_b}{R_s} \right]$$

$$F = \left[R_t \times \frac{K^2 \times 10^4}{p_{sc}} \times \frac{1}{R_s} \left\{ 1 + \frac{2R_b}{R_s} \right\} \right]^{1/2} \quad (13)$$

K = 1.96 for 95% confidence level

$$F = \left[600 \text{ cpm} \times \frac{(1.96)^2}{36,000} \times 10^4 \times \frac{1}{550} \left\{ 1 + \frac{2(50)}{550} \right\} \right]^{1/2}$$

$$F = 1.17\%$$

Graphical Aids to Counting Statistics

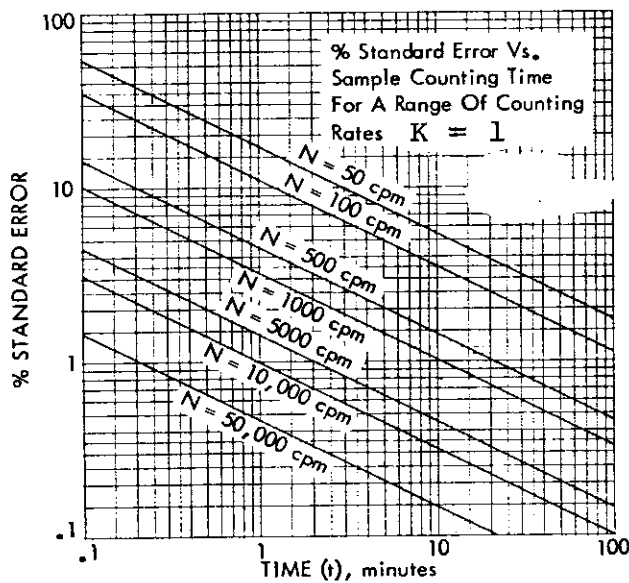


FIG. 3. Percent Error versus Counting Time ($R_b=12\text{cpm}$)

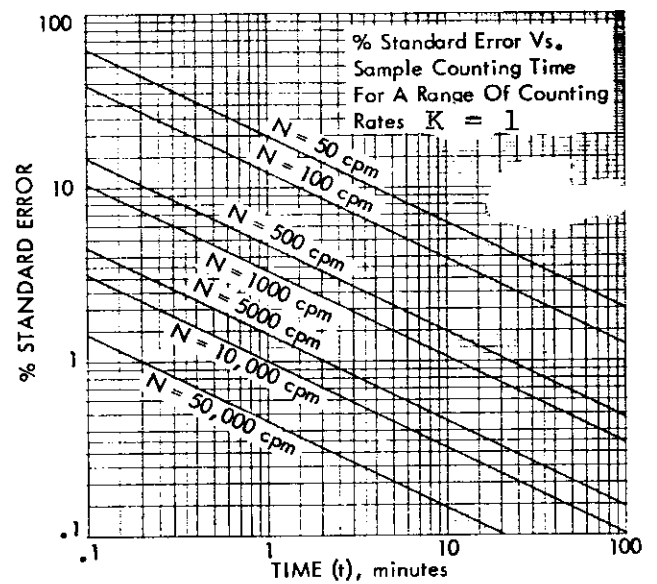


FIG. 4. Percent Error versus Counting Time ($R_b=25\text{cpm}$)

For $N = 50 \text{ cpm}$ what is the % standard error in a 10 min count for both background counting rates?

Fig. 3 - 5.5%

Fig. 4 - 6.3%

For $N = 10,000$ cpm and $t = 10$ min

Fig. 3 - 3.3%

Fig. 4 - 3.3%

7. Relationship of net sample count rate and background count rate.

Use following relationships: R_s = sample counting rate

$$\%F = K \times \frac{100}{(aR_s)^{1/2}} \times C \quad (14)$$

a = number of samples (or number of replicate counts on same sample)

K = number of standard deviations

F = desired error as %

Where C depends on counting conditions

$$C_1 = \frac{d}{d-1} \quad (\text{Background is counted for a long time to make error in background negligible}) \quad (15)$$

$$C_2 = \frac{\sqrt{d^2+1}}{d-1} \quad (\text{Preset COUNT is selected}) \quad (16)$$

$$C_3 = \frac{\sqrt{d^2+d}}{d-1} \quad (\text{Preset time is selected}) \quad (17)$$

$$\text{and } d = \frac{R_t}{R_b}$$

TABLE III

d	C ₁	C ₂	C ₃
1.1	11.000	14.866	15.20
1.2	6.000	7.813	8.124
1.3	4.333	5.477	5.763
1.4	3.500	4.301	4.650
1.5	3.000	3.605	4.025
1.8	2.250	2.574	2.861
2	2.000	2.236	2.449
3	1.500	1.581	1.732
4	1.333	1.378	1.491
5	1.250	1.275	1.369
6	1.200	1.216	1.296
7	1.167	1.178	1.245
8	1.143	1.152	1.212
9	1.125	1.132	1.173
10	1.111	1.116	1.164
25	1.0417	1.0425	1.063
50	1.0200	1.0206	1.031
75	1.0135	1.0136	1.020
100	1.0101	1.0101	1.015
250	1.004	1.004	1.006
500	1.002	1.002	1.003
750	1.001	1.001	1.002
1000	1.001	1.001	1.0015

Ex: When preset count is selected what is the percent error which corresponds to the following conditions:

$$R_t = 1250 \text{ cpm} \quad R_b = 50 \text{ cpm} \quad a = 1$$

K = one sigma or 1.000 (from Table II)

$$R_s = R_t - R_b = 1200 \text{ cpm}$$

$$\%F = K \times \frac{100}{(aR_s)} \times \frac{1}{2} \times C_2$$

$$d = \frac{R_t}{R_b} = \frac{1250}{50} = 25$$

From Table III, $d = 25$ corresponds to $C_2 = 1.0425$

$$\%F = (1.000) \times \frac{100}{(1 \times 1200)^{1/2}} \times 1.0425$$

$$\%F = 3.01$$

8. Choice of counting technique

Advantage of greater selectivity is accompanied by disadvantage of higher background

To determine which system will require shorter counting time for the same error

$$\frac{t}{t'} = \frac{\left[\frac{(aR_t)^{1/2} + (R_b)^{1/2}}{R_s} \right]^2}{\left[\frac{(aR'_t)^{1/2} + (R'_b)^{1/2}}{R'_s} \right]^2} \quad (18)$$

a = number of samples

$R_s = R_t - R_b$ (in cpm)

t = time (in min)

Unprimed - first system

Primed - second system

Ex: In a Geiger-Muller Counter, a sample registered a total count of 100 cpm with a background of 30 cpm. In a proportional counter the same sample gave 125 cpm with a 50 cpm background. If 3 such samples are to be counted which counter will give the same error in less time?

$$\text{GM } R_t = 100 \text{ cpm}$$

$$R_b = 30 \text{ cpm}$$

$$R_s = 70 \text{ cpm}$$

$$\text{Proportional } R'_t = 125 \text{ cpm}$$

$$R'_b = 50 \text{ cpm}$$

$$R'_s = 75 \text{ cpm}$$

$$a = 3$$

$$\frac{t}{t'} = \frac{\left[\frac{(3 \times 100)^{1/2} + (30)^{1/2}}{70} \right]^2}{\left[\frac{(3 \times 125)^{1/2} + (50)^{1/2}}{75} \right]^2} = \frac{.106}{.124} = .853$$

The ratio of $\frac{t}{t'}$ is less than one, (i.e. $t' > t$) and therefore the GM counter will provide the same error as the proportional counter in less time.

9. Errors due to electronic or mechanical misbehavior and data rejection

Two tests

a. Repetitive counts of sample

$$K = \frac{n - n_i}{\sigma_n} \quad (19)$$

If $K > 3.5$ - error non-statistical

Ex: Suppose the same sample was counted five times with the following results:

1. 504
2. 500
3. 502
4. 575
5. 502

Can any of your data be rejected?

The fourth count appears to be much too high; let's check:

$$\begin{array}{rcl} 504 & \bar{n} = \frac{2008}{4} = 502 & \sigma_{\bar{n}} = (\bar{n})^{1/2} = 22.4 \\ 500 & & \\ 502 & n_1 = 575 & \\ \frac{502}{2008} & K = \frac{502-575}{22.4} = 3.26 & \end{array}$$

$K < 3.5$ so the fourth count may not be rejected by this test.

b. Chi-Square Test

$$\chi^2 = \sum_{i=1}^N \frac{[(\text{observed value})_i - (\text{expected value})_i]^2}{(\text{expected value})_i}$$

If the average value is small (e.g., less than 100) you must use the Poisson Distribution to calculate the expected value.

Typically $\bar{n} > 100$ so the Gaussian Distribution can be used. In this case the expected value is identical to \bar{n} , the average value.

Thus for $\bar{n} \geq 100$

$$\chi^2 = \frac{1}{\bar{n}} \sum_{i=1}^N (n_i - \bar{n})^2 \quad (21)$$

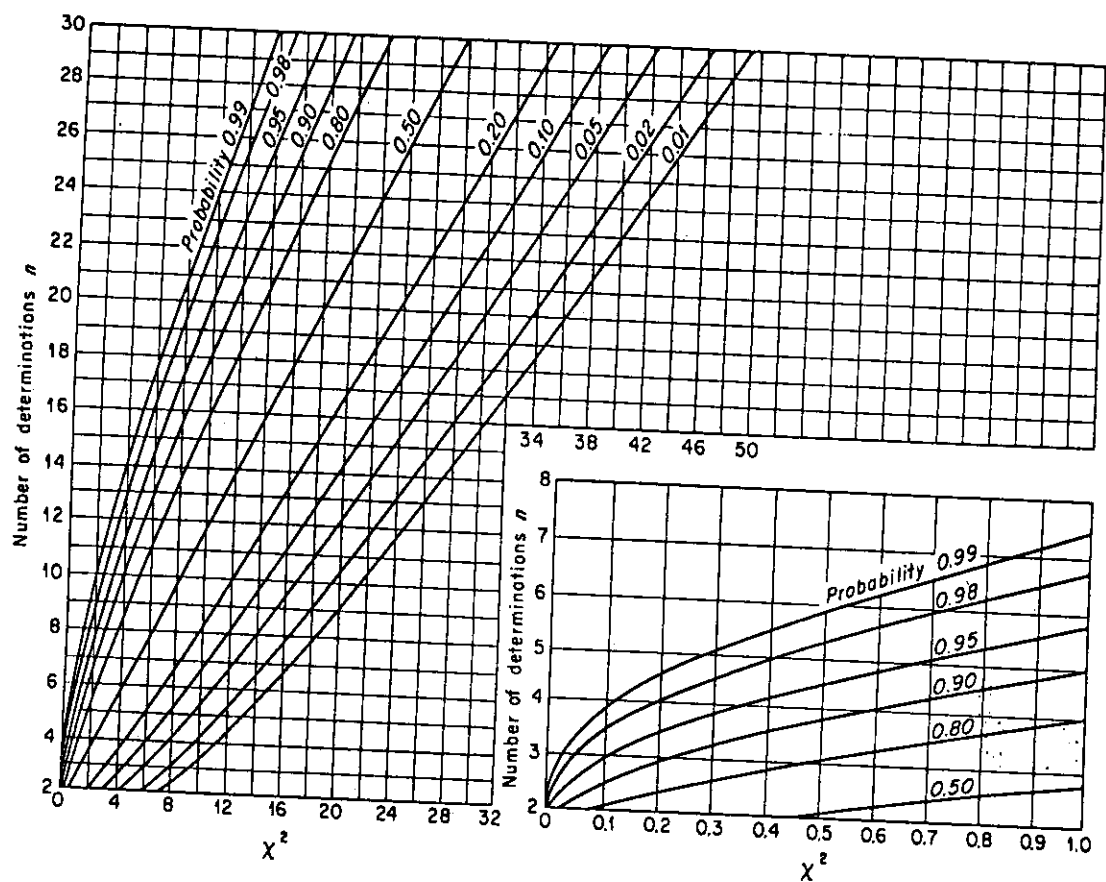


FIG. 5. The χ^2 distribution. The probability that the variations in a series of counting determinations are due to the randomness of the disintegration process. n is the number of determinations.

Example: A G-M counter was tested to see if it was operating properly by taking background counts in eight equal time intervals. The results are listed below

Interval	Count = n
1	312
2	298
3	318
4	309
5	330
6	306
7	291
8	326

The total number of counts observed = 2490 counts

$$\bar{n} = \frac{2490}{8} = 311$$

$\bar{n} > 100$ so eqn (20) was used

Interval	n	$n - \bar{n}$	$(n - \bar{n})^2$
1	312	1	1
2	298	-13	169
3	318	7	49
4	309	-2	4
5	330	19	361
6	306	-5	25
7	291	-20	400
8	326	15	225
<hr/>			
TOTAL 8	2490	0	1234

$$\chi^2 = \frac{1234}{311} = 3.97$$

Using Fig. 5 with $N=8$ and $\chi^2=1.29$ the probability ~ 0.80

Therefore, these 8 counts indicate that the probability that G.M. counter is operating properly is 80%.

Generally felt that better criterion is to reject values deviating from mean by more than 2σ (4.5% probability) or 3σ (0.27%).

SECTION II

Radioactive Standards and Calibration

A. Determination of Absolute Counting Rate - see Part V -
Section II.

B. Standards and Calibration Services

More information about Standard Reference Materials
can be obtained from:

"Standard Reference Materials", NBS, Spec. Publ. 260,
Gov't. Printing Office, Washington, D.C. (semiannual

or "Calibration and Test Services of the National Bureau
of Standards", NBS, Spec. Publ. 250, Gov't. Printing
Office, Washington, D. C. (1970) (revised pages
and new additions as needed.)

Guidelines for the user's of Radioactivity Standards can
be found in:

"User's Guide for Radioactive Standards": NAS-NS
3115(1974).

SECTION III

Problems

1. A sample with a gross counting rate of 5,000 cpm is counted for half an hour. What is the interval about the total count which corresponds to a 68% probability?
2. A sample counted for 15 minutes gave 9,000 total counts. A 30 minute background count registered 1200 counts. Calculate the count rate of the sample alone with its standard deviation and its probable error.
3. If only 30 minutes of counting time are available, calculate the desirable division of time between sample and background for minimum error if the background is approximately 30 cpm and total rate 100 cpm. What will the standard deviation for the sample be, expressed as a percentage error?
4. For a series of 100 counts, \bar{n} is 100 cpm. How many will give 127 cpm, 83 cpm, 100 cpm?
5. In a Geiger-Muller counter, a sample registered a total of 60 cpm with a background of 30 cpm. In a proportional counter the same sample gave 95 cpm with a background

of 60 cpm. If 3 such samples are to be counted, which system will give the same error in less time?

6. The following counting data were collected per minute with one sample: 3308, 3277, 3411, 3080, 3580, 3425, 3207, 3436, 3328, 3363 counts. Should any of these counts be rejected if it is desired to calculate a good average count rate?
7. A Geiger-Muller counter has a background of 18 cpm with a lead shield. Unshielded, the background rises to 48 cpm. For a sample which counts 60 cpm above background, what are the necessary counting times in the shielded and unshielded arrangements to obtain 5% probable error?
8. The same sample is counted with two different Geiger-Muller counters to determine whether their absolute sensitivity is the same. Counter 1 gave 500 counts in the same time interval that Counter 2 give 525 counts. Is this a statistically significant difference? Would the difference be significant if the two counts were 500 and 590?

9. Calculate the total number of counts that must be collected in each case for a count rate of 500 cpm to give probable errors of 0.1%, 0.5%, 1%, 5%, 10% and 50%.
10. Two successive counts in a proportional counter of the same sample for the same interval gave values of 5440 and 5600. Can the counter be considered to be operating normally? What would be the most probable conclusion if the two counts were 5440 and 5750?

PART VII

Pulse Height Analysis and Gamma Ray Spectroscopy

Outline

- I. Pulse-Height Analyzers
 - A. Single-channel Analyzer
 - B. Single-channel Differential Analyzer
 - C. Multi-channel Analyzers
- II. Gamma Ray Spectroscopy
 - A. NaI(Tl) Scintillation Spectrometers
 - 1. Gamma Ray Interactions with Crystals of different sizes
 - 2. Example of Low Energy γ -ray Interaction
 - 3. Example of Intermediate Energy γ -ray Interaction
 - 4. Example of High Energy γ -ray Interaction
 - 5. Diagram of a Typical NaI(Tl) Spectrometer
 - 6. Environmental Effects
 - a. Backscatter
 - b. Bremsstrahlung
 - 7. Sum Peaks
 - 8. Contrast of Ideal and Real Spectra
 - 9. Calculation of Compton Edge
 - 10. Resolution
 - 11. Calibration Procedure
 - 12. Calculation of Source Intensity
 - B. Solid State Detectors
 - 1. Gamma Ray Interactions
 - 2. Absorption Efficiency of several γ -ray Spectrometers
 - 3. Resolution Characteristic
 - 4. ^{239}Np Spectra in NaI(Tl) and Ge(Li)
 - 5. Other Spectra with Ge(Li)
- III. Problems

SECTION I

Pulse-Height Analyzers

I. Pulse-Height Analyzers

Height of pulse depends on energy of incident radiation.

Discriminator or pulse-height selector (PHS) - excludes pulses below a preselected level.

A. Single-Channel Analyzer

Make successive statistical counts at different "window", PHA settings, called channels, and determine number of pulses by successive subtraction. Then plot pulse-height distribution.

Sample data

PHS setting	Channel	No. of Pulses
0-100	1	0
100-200	2	2
200-300	3	5
300-400	4	2
400-500	5	0

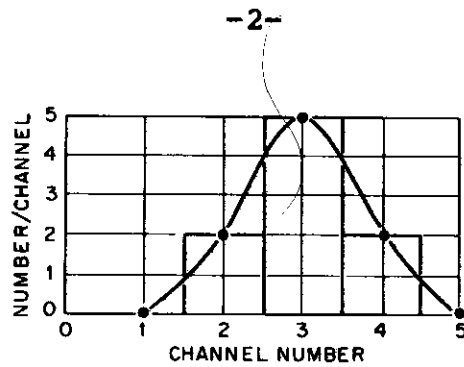


FIG. 1. Pulse-Height Distribution

B. Single-Channel Differential Analyzer

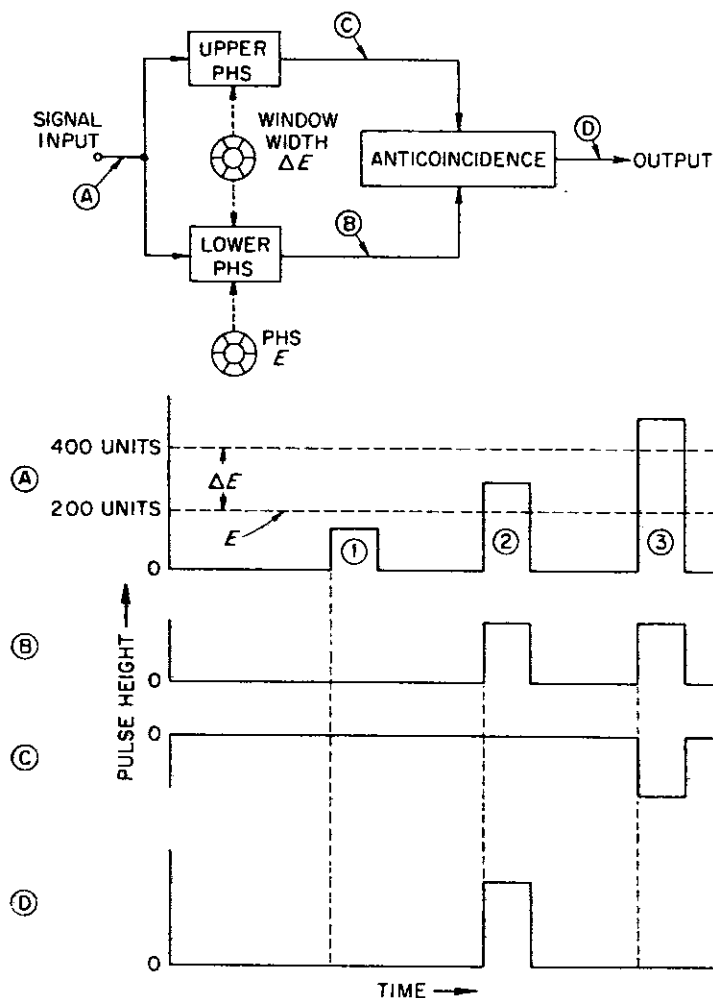


FIG. 2. Functional diagram of a single-channel pulse-height analyzer. Pulse shapes for the different part of the circuit are shown below the block diagram.

C. Multi-Channel Pulse Height Analyzers

Series of single channel analyzers each with a discriminator level slightly different. Used for α , β , and γ energy analysis however more helpful for α and γ where energy is discrete.

1. Pulse-height analysis for α 's:

Problem - thickness of source.

2. Pulse-height analysis for γ 's:

Can simultaneously measure a number of γ emitters.

SECTION II

Gamma Ray Spectroscopy

A. NaI(Tl) Scintillation Spectrometers

NaI crystal doped with Tl

Disadvantage-hygroscopic

Advantage-rather large size crystals are available.

Gamma rays interact with matter by: (Review Part III,
Section IIC)

1. Photoelectric Effect
2. Compton Scattering
3. Pair Production

All above processes occur in NaI(Tl) crystals. In all processes, γ ray interacts with I in NaI.

Note: γ rays are not directly detected - secondary electrons produced by above processes in the crystal give rise to light detected and amplified by phototube.

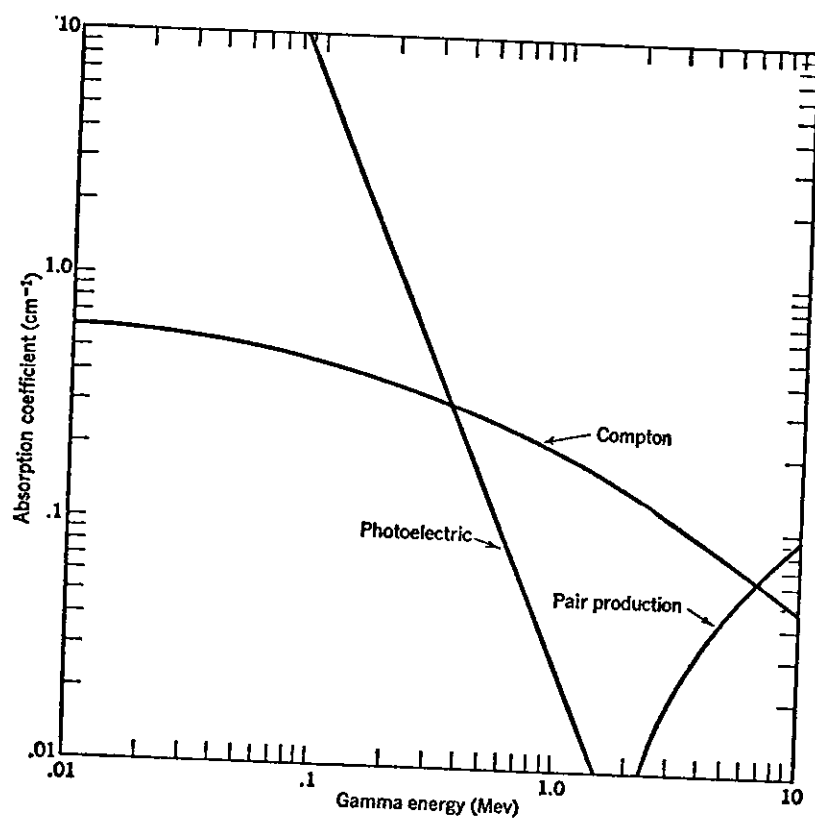


FIG. 3. Dependence of absorption coefficients for sodium iodide on energy.

1. Gamma Ray Interactions in Crystals of Different Sizes:

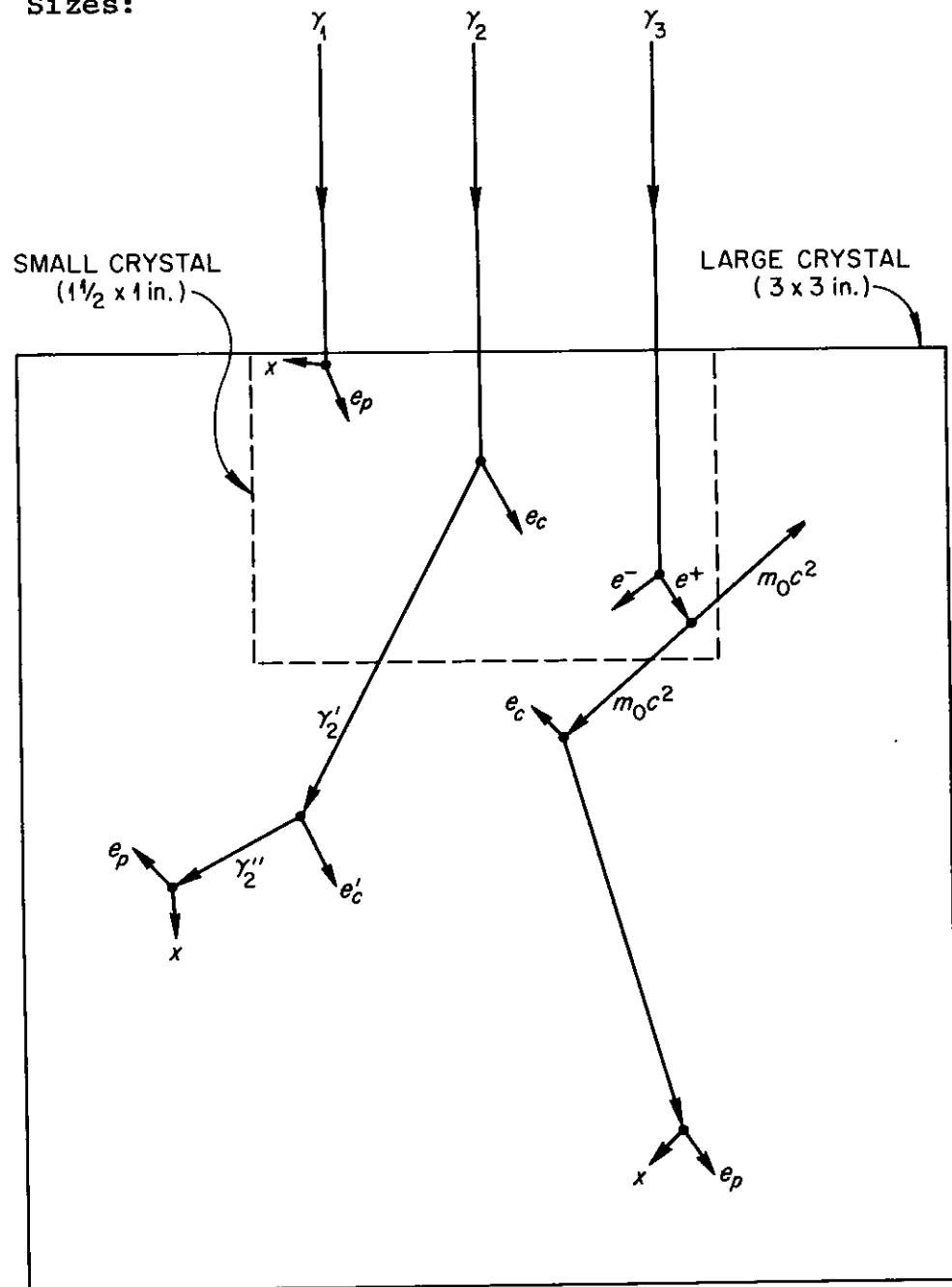


FIG. 4. Schematic representation of gamma-ray interactions within NaI(Tl) crystals of two sizes.

e_p - photoelectron (very short range)

x - x-ray (\sim 28 keV for iodine)

e_c - compton electron

γ' or γ'' - scattered (lower energy) γ rays

e^+ and e^- - ion pair

$m_0 c^2$ - annihilation photons (energy of each = 0.51 MeV)

2. Example of Low Energy γ -ray Interaction

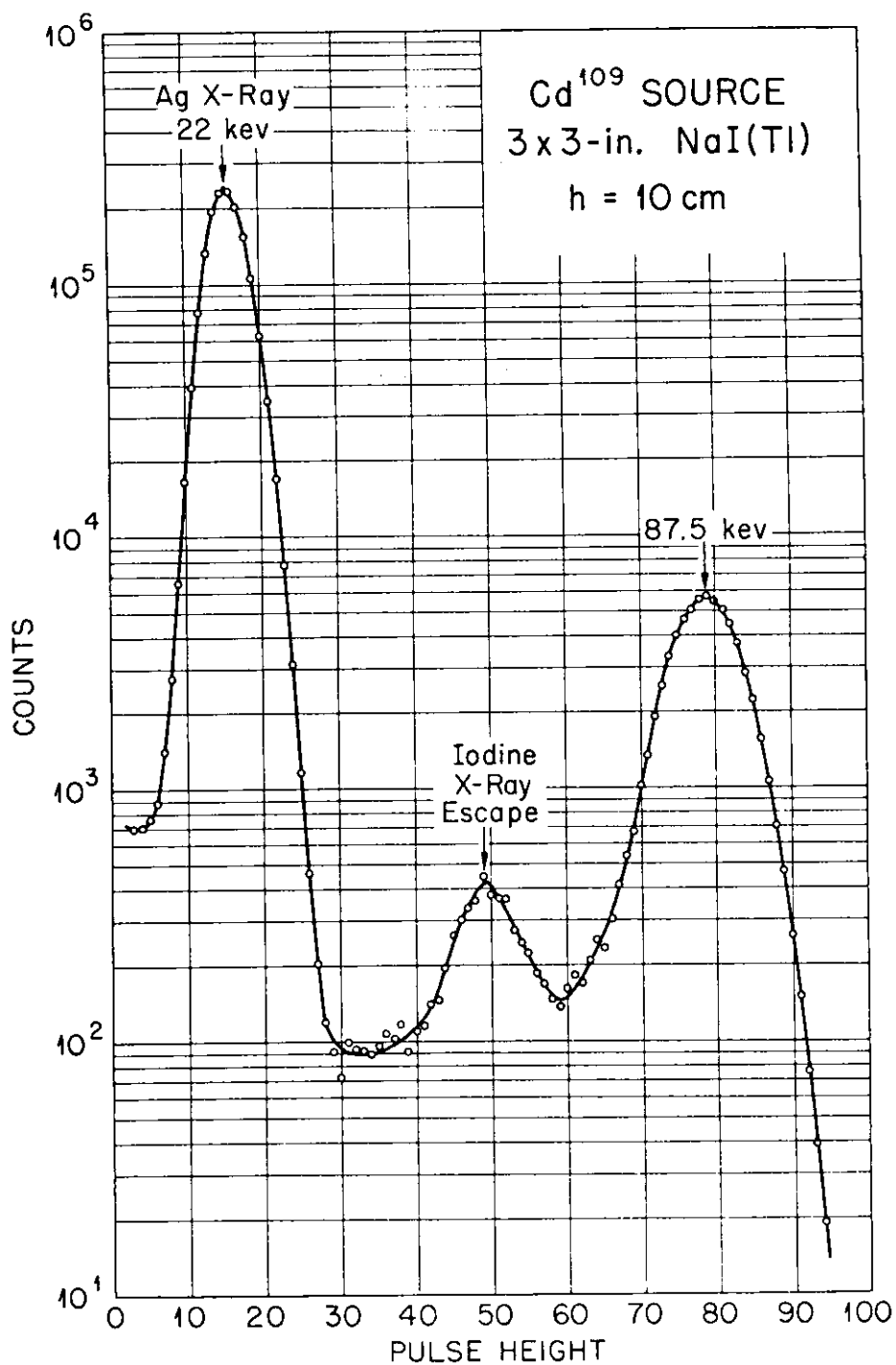


FIG. 5. Spectrum of 87.5 keV gamma rays and 22 keV X rays from a ^{109}Cd source, illustrating the phenomenon of X-ray escape following detection of 87.5 keV gamma rays.

3. Example of Intermediate Energy γ -ray Interaction

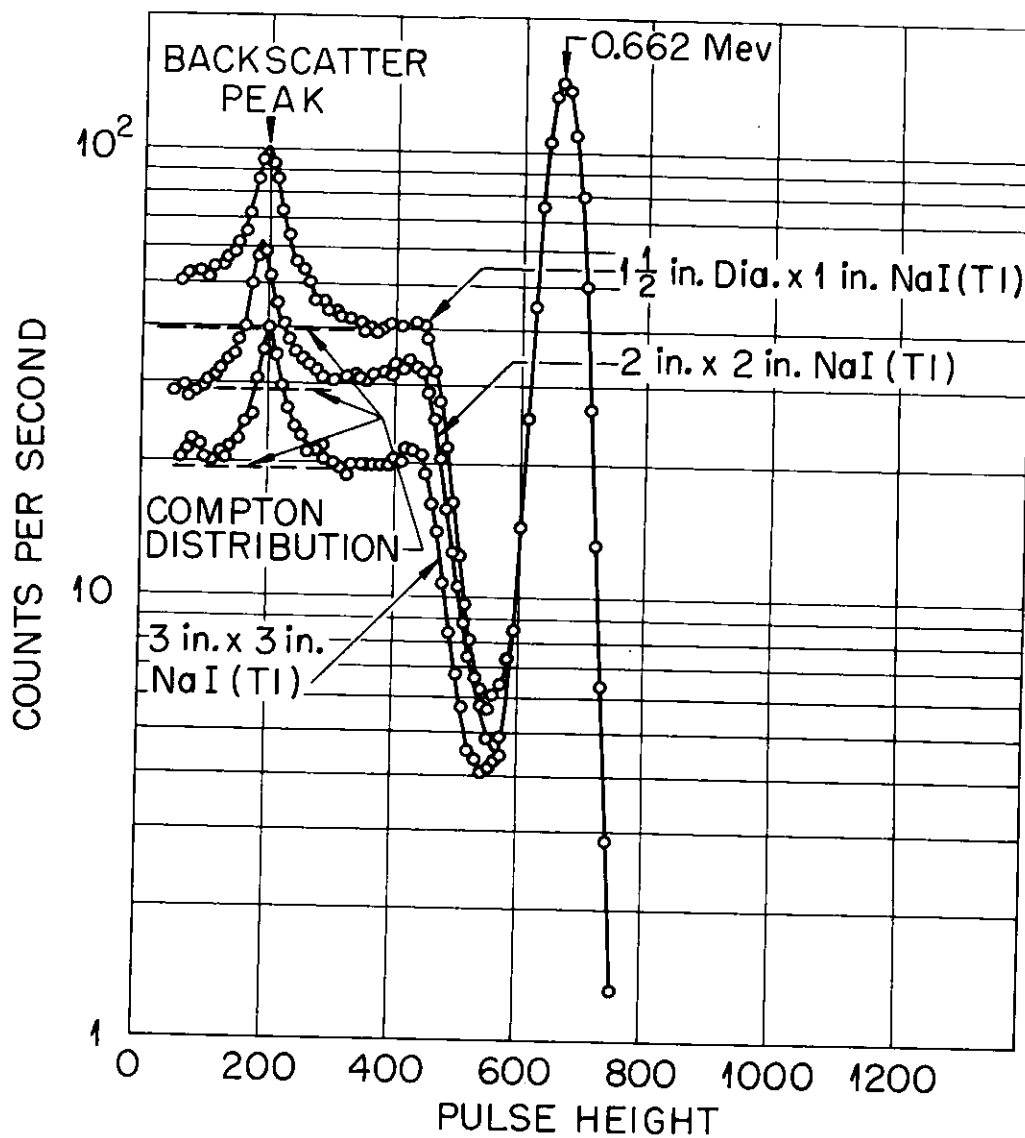


FIG. 6. Spectra obtained by measuring a ^{137}Cs source with NaI(Tl) spectrometers of three crystal sizes.

- A - Photo peak
- B - Compton peak
- C - Backscatter peak - due to scattering of gamma off of crystal shielding back into crystal - usually found at ≥ 200 . instrumental problem.

4. Example of High Energy γ -ray Interaction

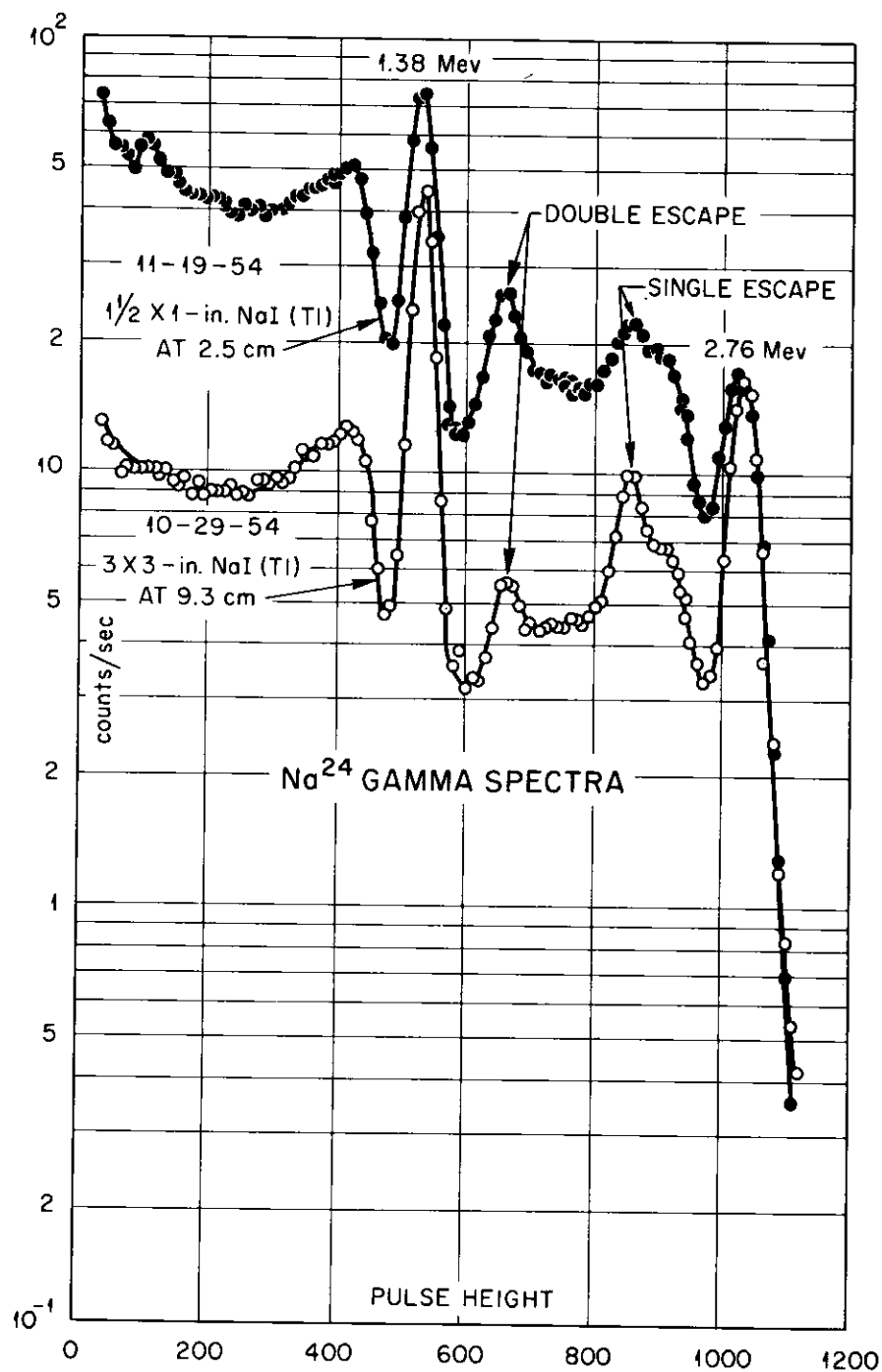


FIG. 7. Gamma ray spectra of ^{24}Na , using $1\frac{1}{2} \times 1$ inch and 3×3 inch NaI (Tl) spectrometers.

^{24}Na -2 γ -rays in this source $\gamma_1 = 2.76 \text{ MeV}$
 $\gamma_2 = 1.38 \text{ MeV}$

Note: Pair production usually not detected for
 γ -ray $< 1.5 \text{ MeV}$.

Summary:

γ_1 - low energy - photoelectric effect; e_p and x-ray

In Spectrum:

- a) Photopeak at energy of γ_1
- b) Iodine x-ray escape peak of energy = $\gamma_1 - \text{x-ray}$
- c) x-ray peak = energy of x-ray

γ_2 - intermediate energy - Compton and photoelectric effects. Size of crystal important.

In Spectrum:

- a) 1.5 x 1 in crystal
If γ_2' is stopped - photopeak at energy of γ_2
If γ_2' escapes - Compton peak - energy $\gamma_2' = \gamma_2 - \gamma_2'$

- b) 3 x 3 in. crystal
One large photopeak at energy of γ_2 (γ_2' captured).

Note: As γ ray energy increases, probability for x-ray escape diminishes.

γ_3 - high energy - pair production - positive and negative electron pair produced.

Energy of pair = $\gamma_3 - 1.02 \text{ MeV}$ ($2 m_{e^-}$)

Positron annihilates forming 2 photons of 0.51 MeV each 180° apart.

Small crystals - probably both annihilation photons escape

Large crystals - probably one annihilation photon and possibly both are stopped.

In spectrum:

3 peaks

- a) Full energy peak - due to multiple processes
- b) Single escape peak - loss of one annihilation photon
- c) Double escape peak - both annihilation photons lost.

5. Diagram of Typical NaI(Tl) Spectrometer

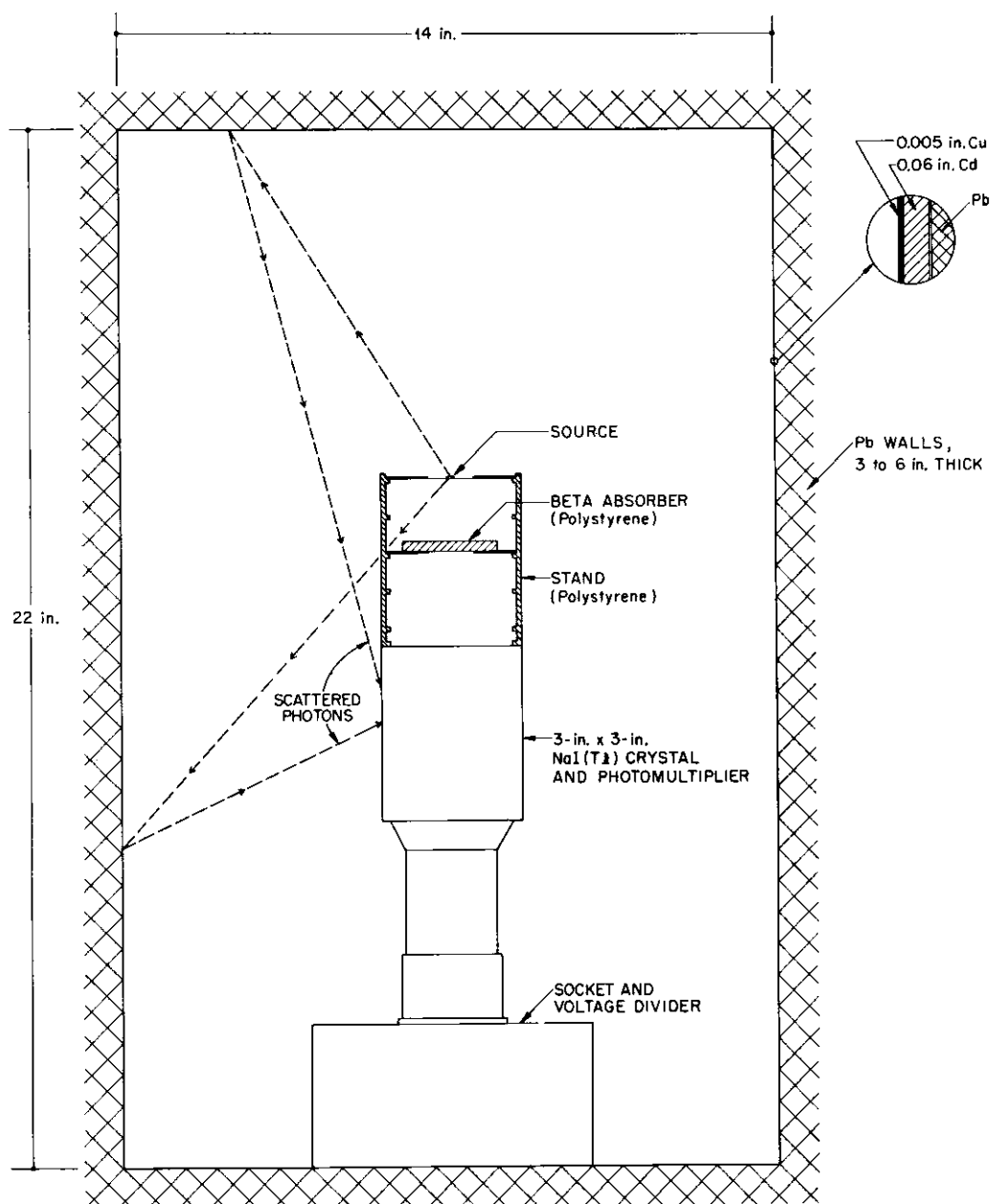


FIG. 8. Cross-section of a typical scintillation spectrometer installation, showing the 3x3 inch NaI (Tl) detector assembly, the lead shielding with "graded" liner, and the use of a low-mass support for the source and beta absorber. The origin of scattered photons is illustrated.

6. Environmental Effects

- a. Back Scatter - Results from Compton scattering in the walls of the detector shielding. If E_γ is larger ($E_\gamma > 1/2 mc^2$) the scattered γ will have a minimum energy approaching $1/2 mc^2$ - 250 keV. Thus a backscattering peak often shows up in the spectra at around 250 keV.
- b. Bremsstrahlung - Energy lost as radiation by charged particles passing through matter. Although most of the energy goes into excitation and ionization of the interacting medium some bremsstrahlung radiation is produced. It has a continuous energy spectrum from 0 to E_{\max} of the particle.

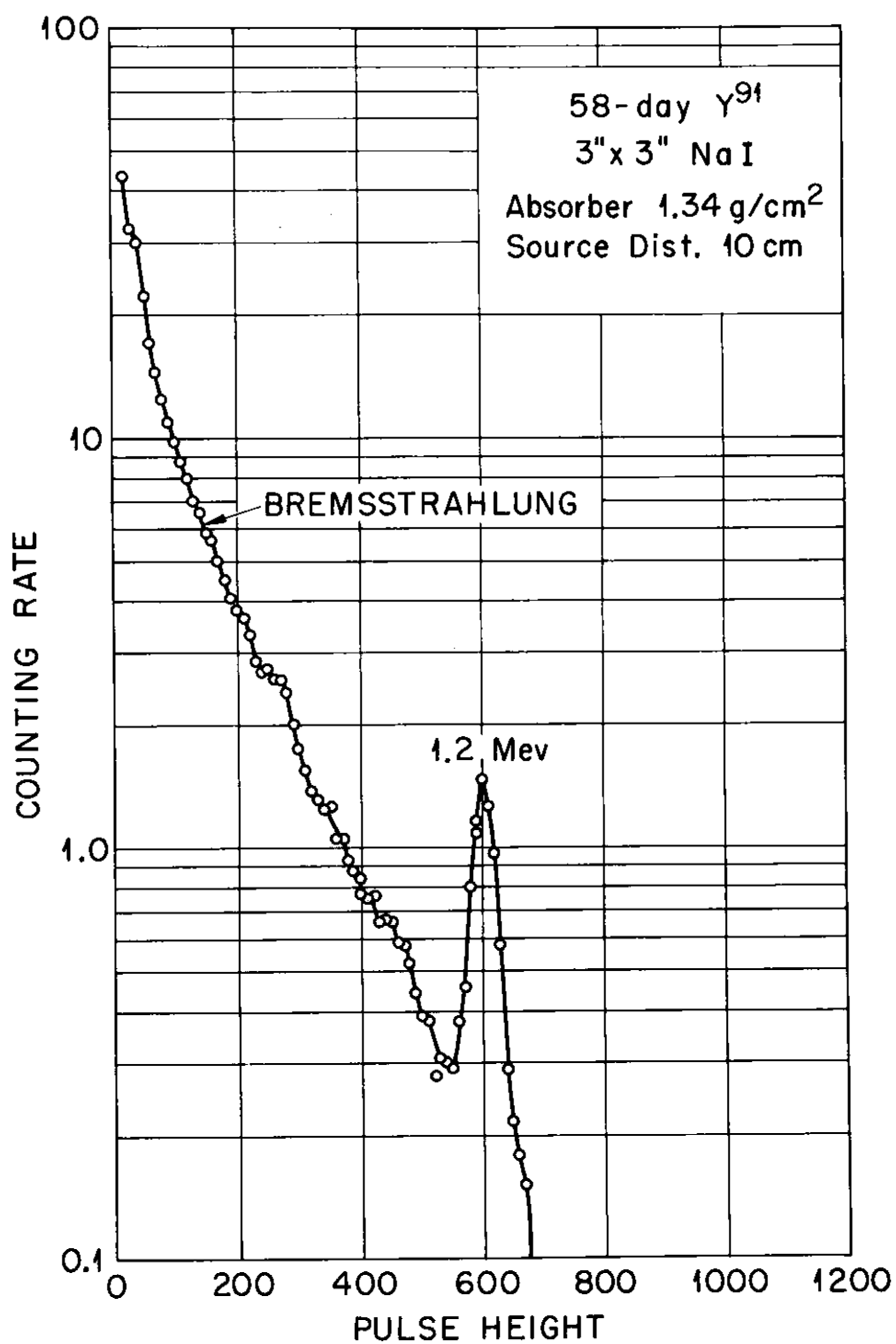


FIG. 9. Gamma-ray spectrum of 58-day γ^{91} , showing the bremsstrahlung spectrum characteristic of a source for which the beta-to-gamma intensity ratio is very large.

7. Sum Peaks

If two gamma rays from a source enter the detector crystal simultaneously, the total energy deposited may be the sum of the energy of the two gamma rays. May be coincident gamma rays or same gamma ray from high intensity source.

$$E_{\text{sum}} = E_{\gamma_1} + E_{\gamma_2} \quad \text{for coincident gammas}$$

$$E_{\text{sum}} = 2E_{\gamma_1} \quad \text{for two identical gammas}$$

Sum peak intensity decreased by increasing distance between sample and detector.

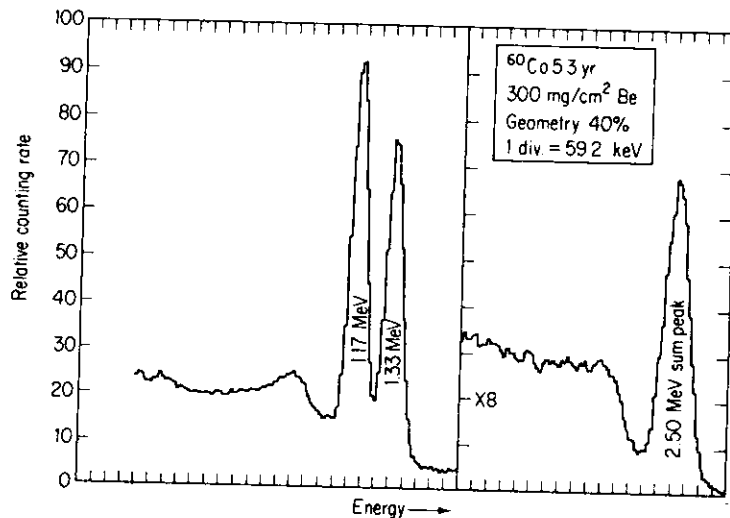


FIG. 10

FIG. 11

Spectra of Co showing sum peak.

8. Contrast of Ideal and Real Spectra

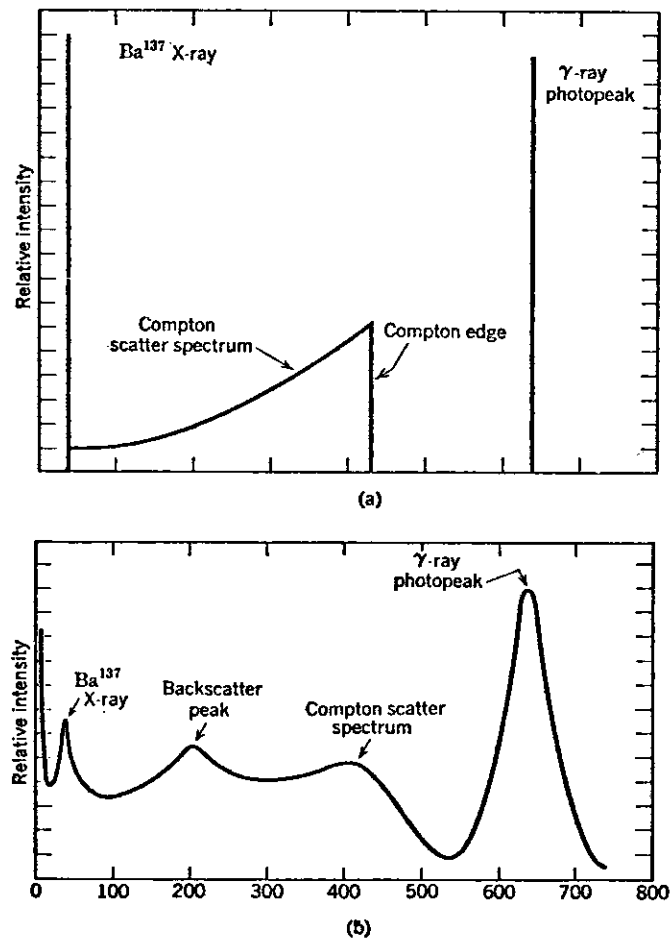


FIG. 12 (a) Gamma Spectrum from an ideal detector.

(b) Gamma spectrum from a real detector.

9. Calculation of the Compton Edge

$$E_e = \frac{4E_\gamma^2}{4E_\gamma + 1} \quad (1)$$

E_e = energy of electron

E_γ = energy of incident γ ray

10. Resolution-measure of ability to differentiate γ rays of similar energies.

$$\text{Resolution} = \frac{\text{FWHM}}{\text{Pulse height (or energy)}} \times 100 \quad (2)$$

FWHM = full width at half maximum in number of channels or of energy.

Ex: Pulse height = 635 keV; FWHM = 75 keV

$$\text{Resolution} = \frac{76 \text{ keV}}{635 \text{ keV}} \times 100 = 12\%$$

11. Calibration Procedure

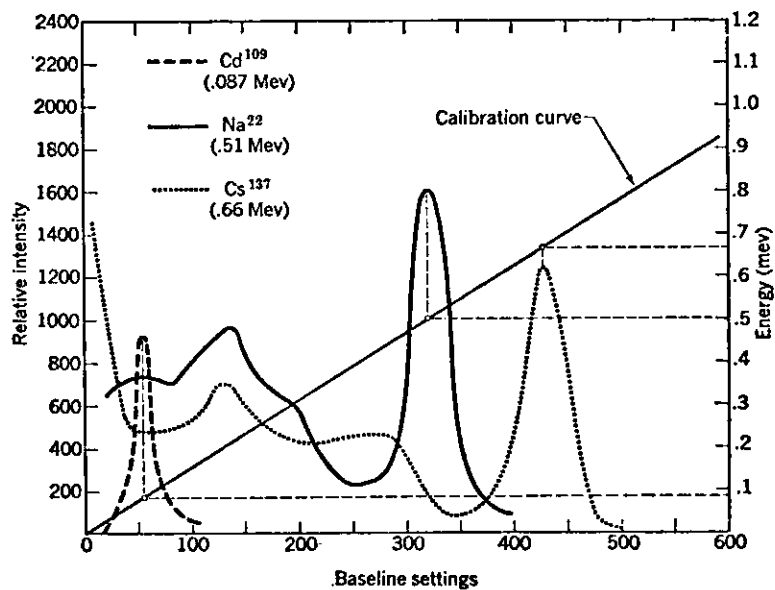


FIG. 13. An illustration of the method of using gamma spectra curves to obtain a calibration curve for the base-line settings as a function of energy. The calibration curve is drawn through the points determined by the base-line settings and the energy of the photopeaks of the standards.

B. Solid State Detectors

1. Gamma Ray Interactions

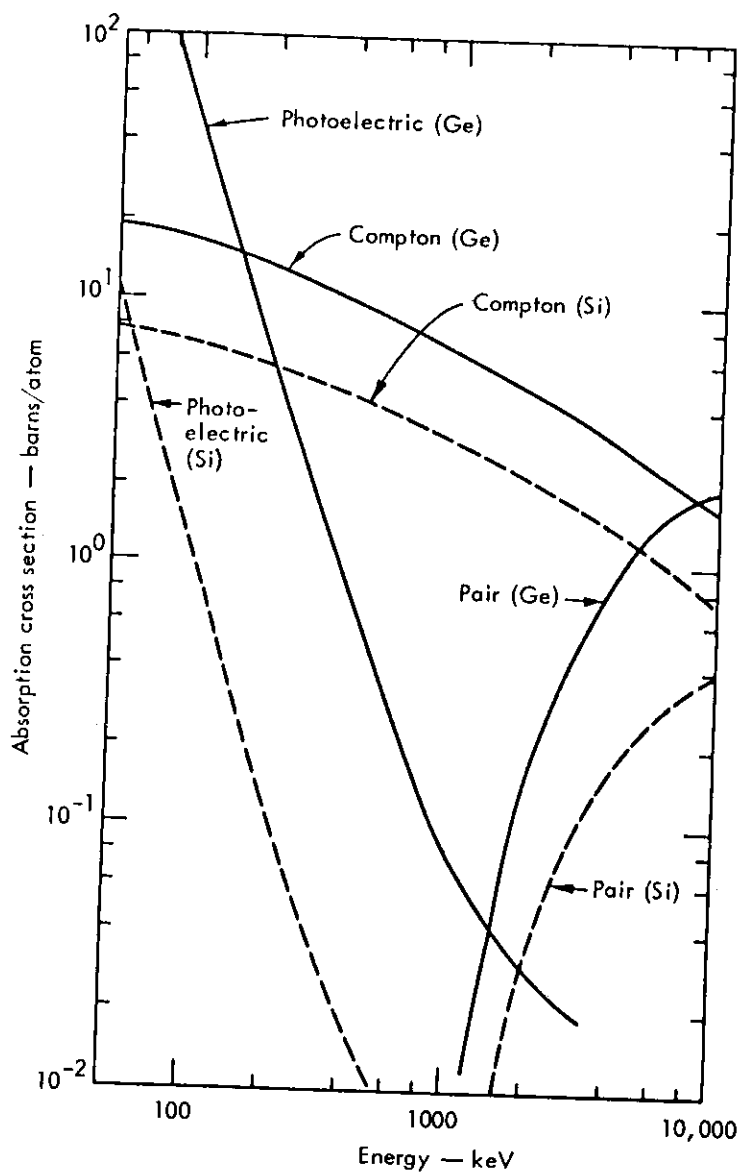


FIG. 14. The variation with energy of the photoelectric, Compton, and pair-production cross sections in silicon and germanium.

2. Absorption Efficiency of Several γ ray Spectrometers

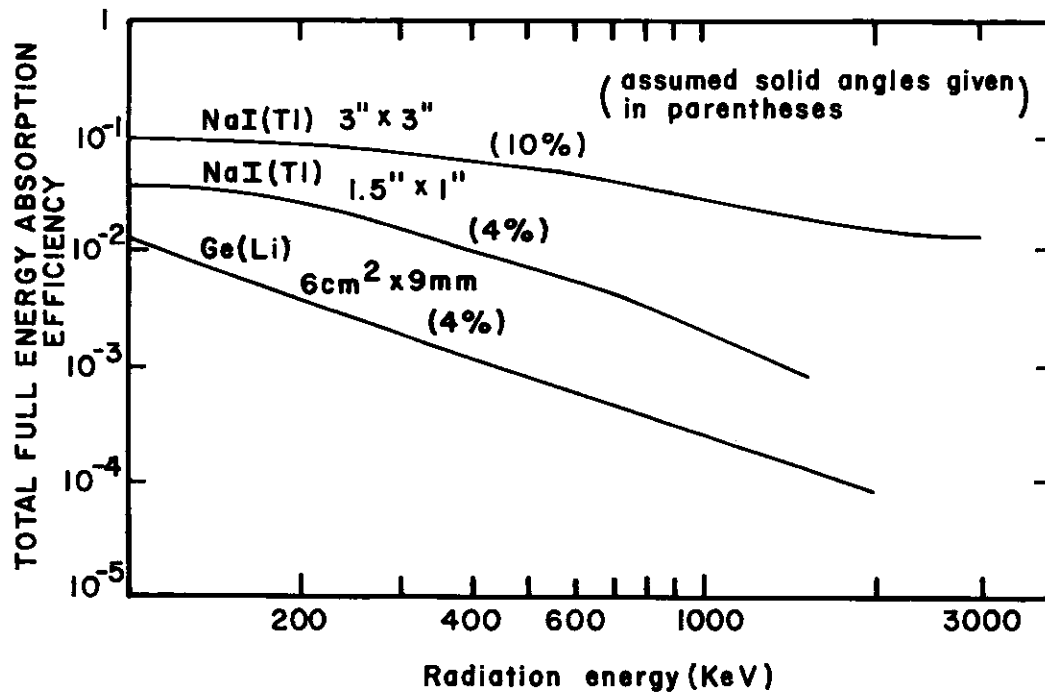


FIG. 15

3. Resolution Characteristics

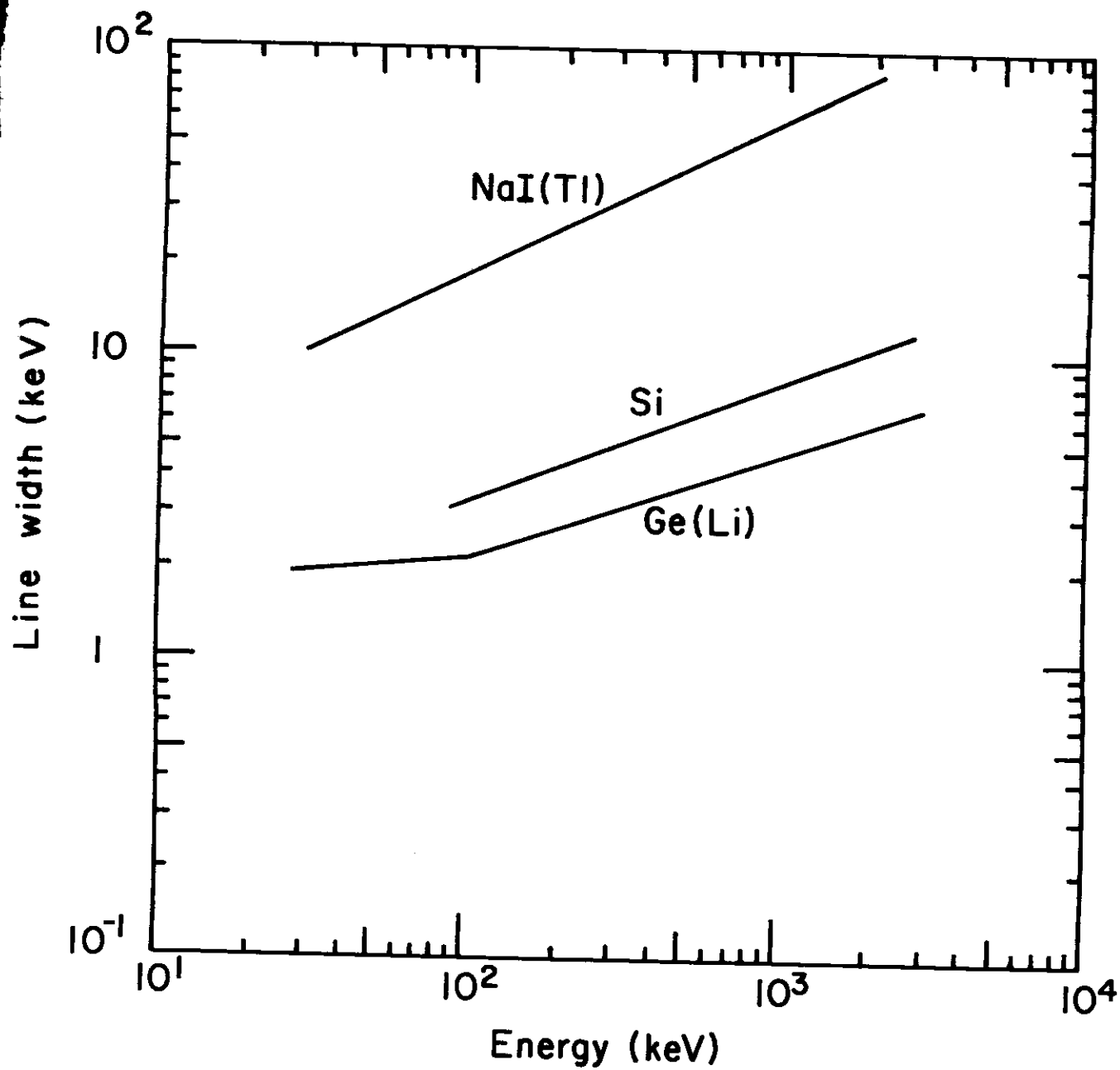


FIG. 16

4. ^{239}Np Spectra in NaI(Tl)

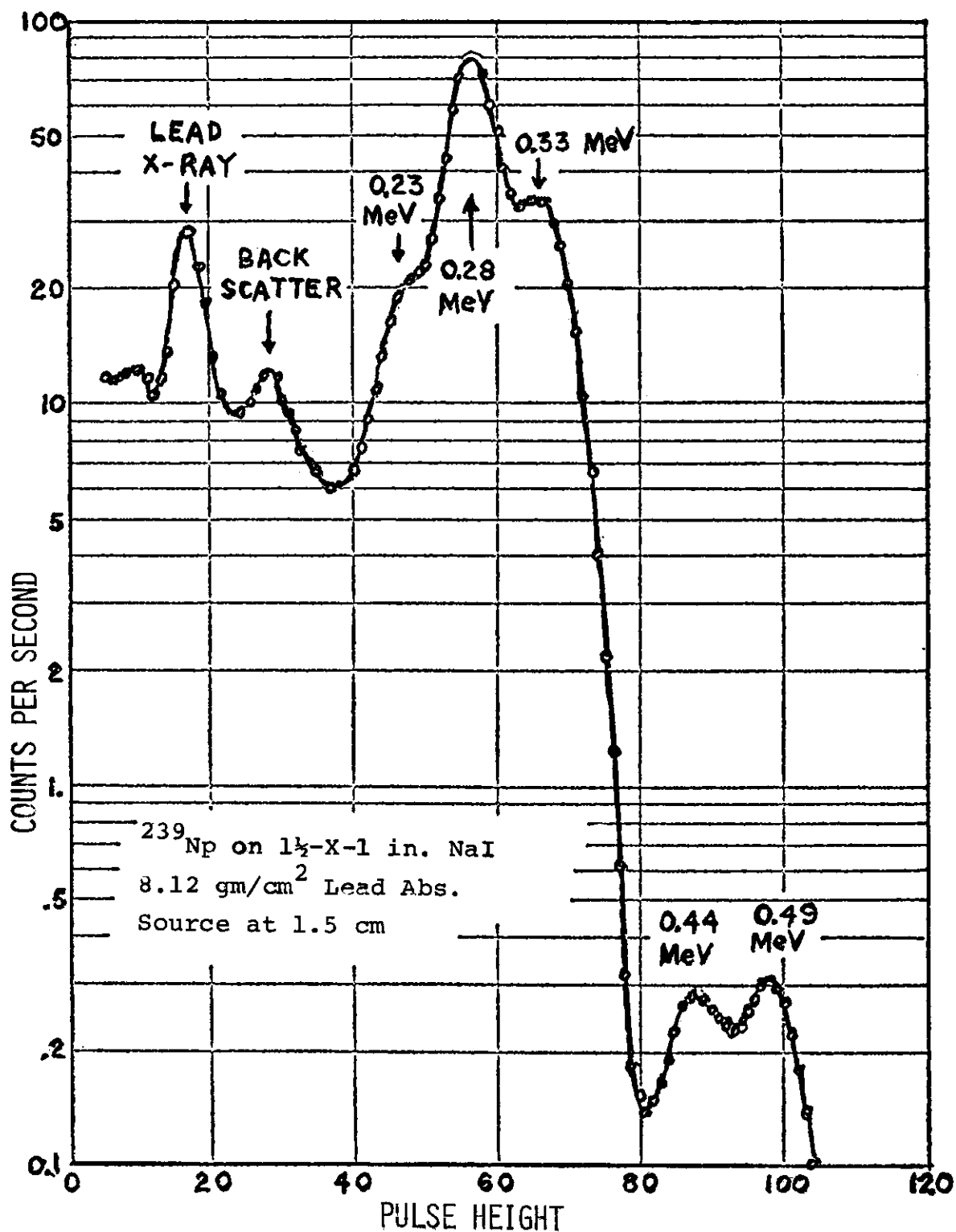


FIG. 17

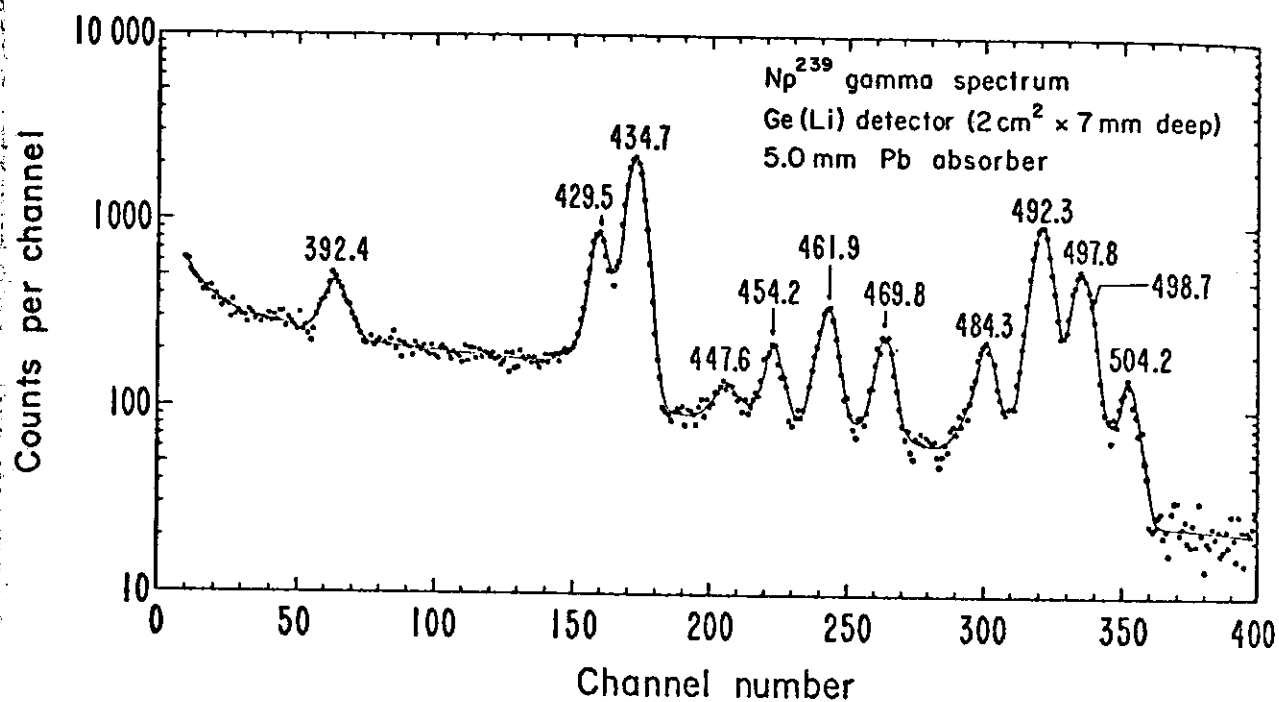


FIG. 18

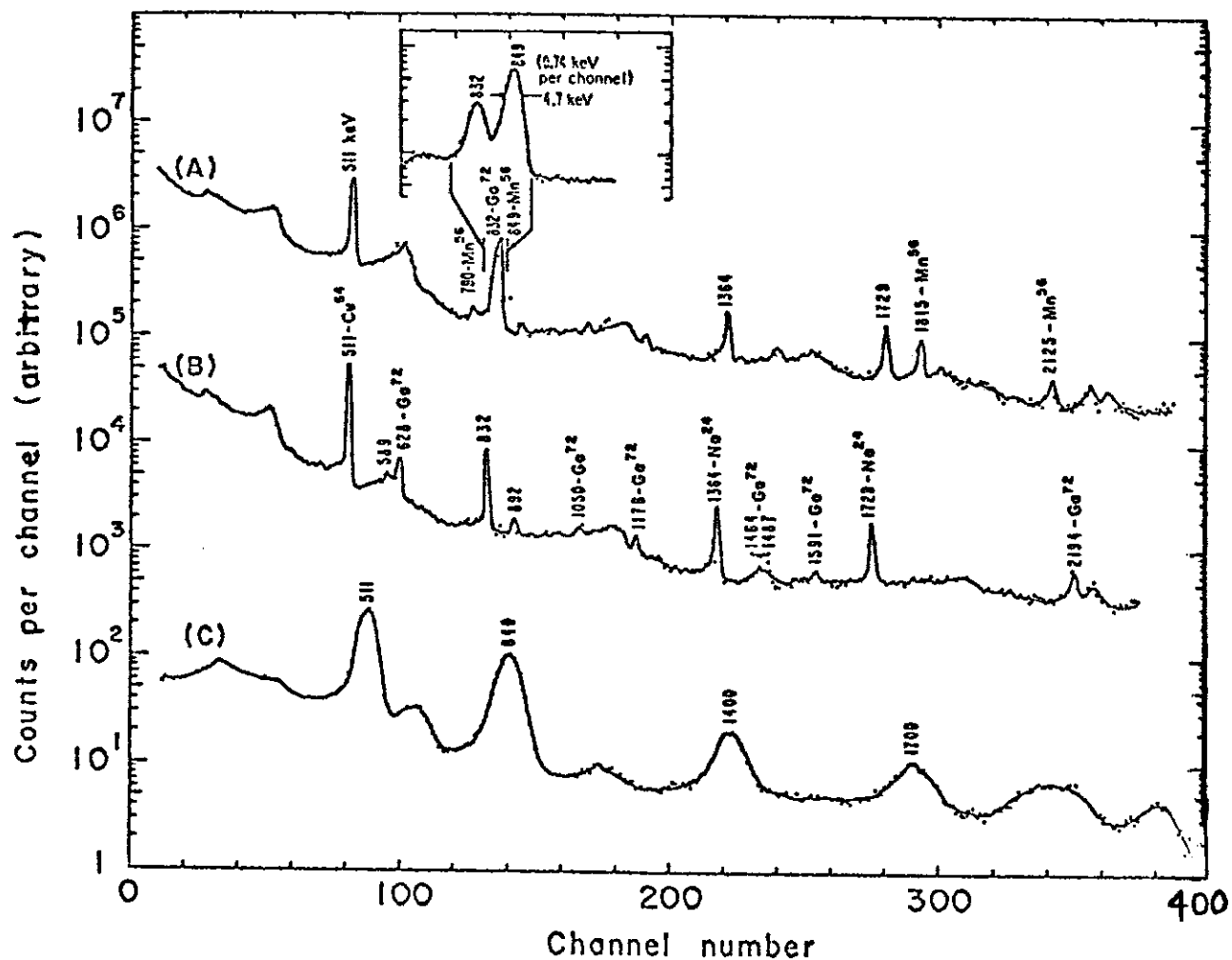


FIG. 19. Neutron Activated Al

- (A) Ge(Li) 2 hr irradiation, 5.2 hr after irradiation
- (B) Ge(Li) 2 hr irradiation, 54.7 hr after irradiation
- (C) NaI(Tl) 3" x 3" 2 hr irradiation, 5.7 hr after irradiation.

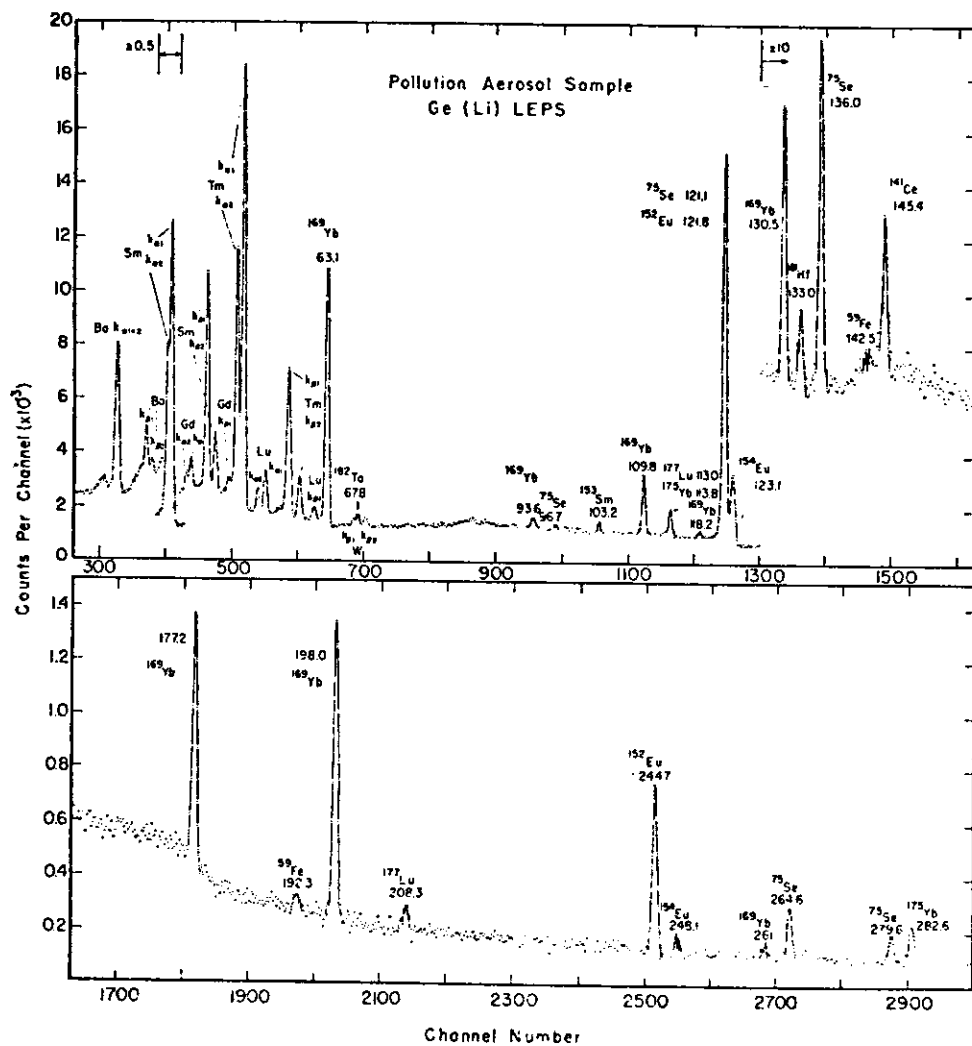


FIG. 20. Low-Energy Portion of the Gamma and X-ray spectrum of an irradiated millipore filter sample observed with the 0.5 cm³ Ge(Li) detector six days after a 9 hr irradiation. [W.H. Zoller and G.E. Gordon, Anal. Chem. 42 (2), 261 (1970)].

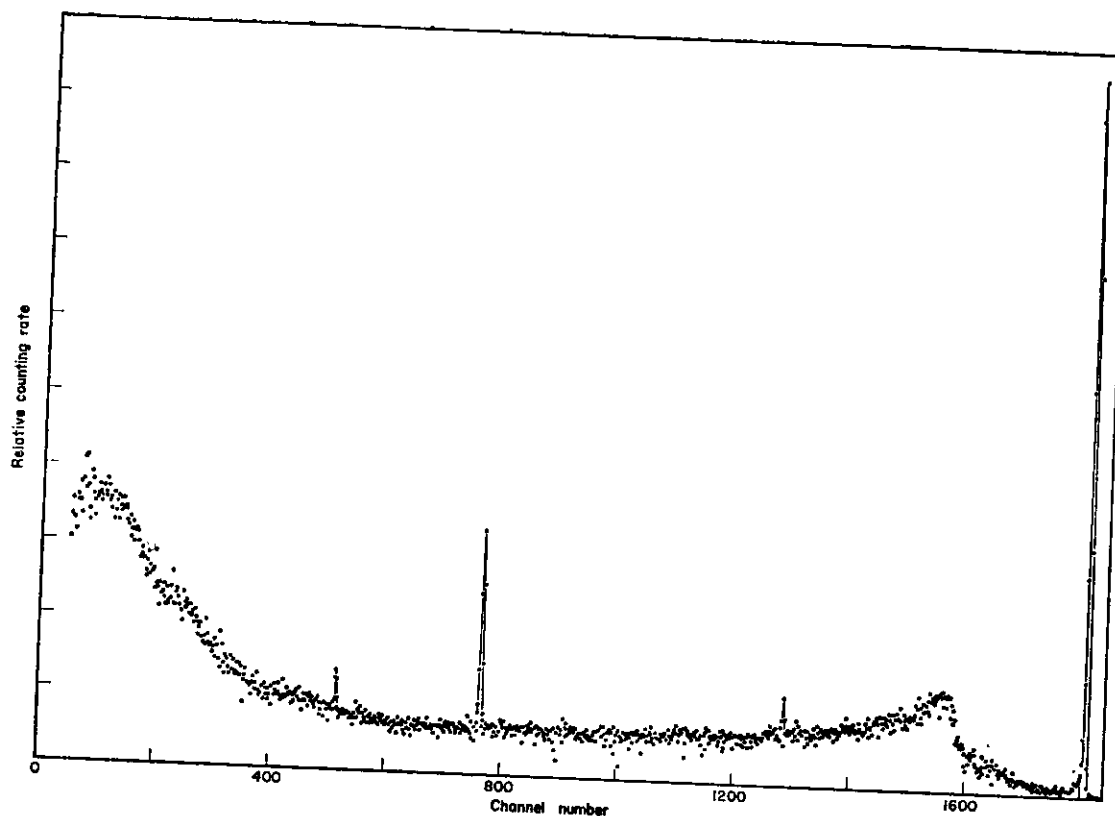
SECTION III

Problems

1. Calculate the Compton edge for the following γ ray emitting nuclides

	E (MeV)
a) ^{46}Sc	0.986, 1.314
b) ^{22}Na	0.51
c) ^{109}Cd	0.087

2. What will be the most probable mode of interaction between matter (i.e., a NaI(Tl) detector) and the γ rays emitted by the nuclides in problem 1? Explain your answers.
3. Figure 21 is a γ ray spectrum of ^{28}Al ($E_\gamma = 1.78$ MeV) obtained on a Ge(Li) detector. What would you expect to be the dominant mode of interaction? Make assignments for peaks a-d. Based on your assignments, at what energies would you expect to find peaks b-d?



4. For a given γ ray what is the effect of the sodium iodide crystal size on the percent of gamma rays completely absorbed? (Assume the crystal is cubic). For a given NaI(Tl) crystal size what is the effect of E_γ of the incident γ ray on the percent of gamma rays absorbed?

5. Plot the following NaI(Tl) counting data on semi-log paper to obtain the spectrum for $^{99m}\text{Te}(E_{\gamma}=140 \text{ keV})$

<u>Total Count</u>	<u>Pulse Height (keV)</u>
150	40
160	50
180	60
210	70
305	80
560	90
1050	100
800	110
620	120
700	126
1700	130
6000	136
10,000	140
4000	146
1700	150
300	156

What is the resolution based on the 140 keV photopeak? On the low energy side of the photopeak at 100 keV is the I x-ray escape peak. What is the energy of the x-ray?

6. Given below is the γ ray spectrum of ^{60}Co which has two gamma rays. $E_{\gamma_2} = 1.17 \text{ MeV}$ and $E_{\gamma_1} = 1.33 \text{ MeV}$. Identify all the peaks and humps in the spectrum as to energy. Theoretically, the Compton peak associated with a photo-peak has an energy given by the equation 1. Compare the theoretical values with the ones given in the spectrum.

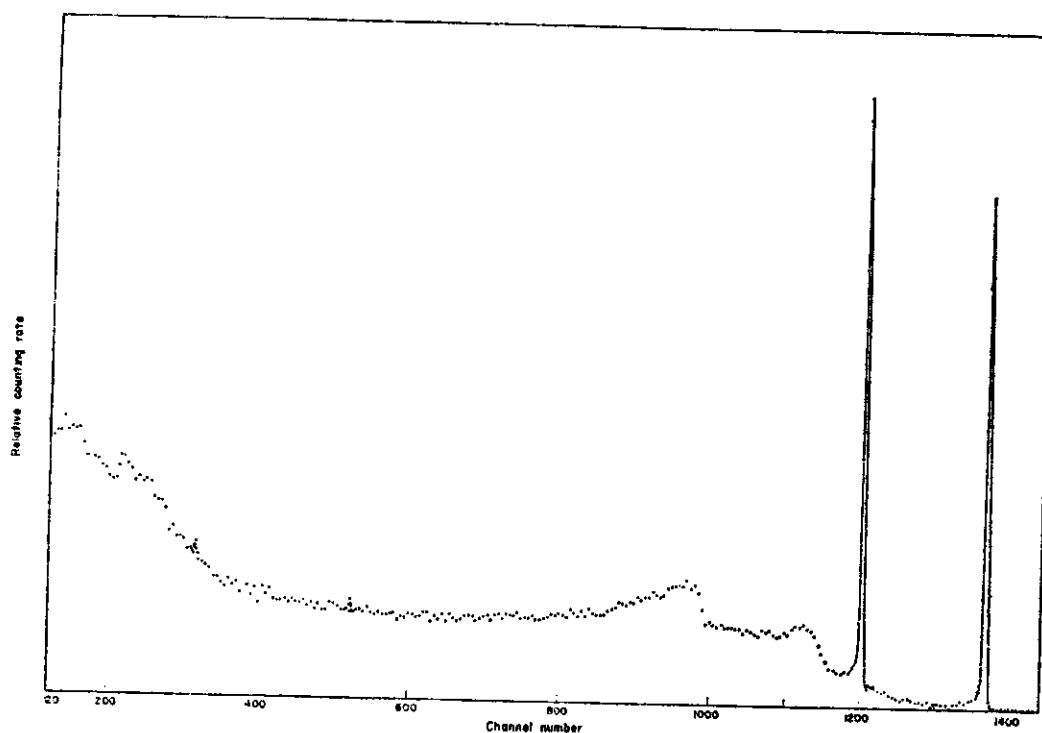


FIG. 22. Gamma spectrum of ^{60}Co

PART VIII

Induced Nuclear Reactions

Outline

- I. Introduction to Induced Nuclear Reactions
 - A. Notation for nuclear reactions
 - 1. Formal
 - 2. Shorthand
 - B. Schematic reaction diagram
 - C. Important quantities conserved
 - D. Types of projectiles
 - 1. Neutrons
 - 2. Charged particles
 - 3. Electromagnetic radiation
 - E. Threshold energy
- II. Types of Nuclear Reactions
 - A. Elastic scattering
 - B. Inelastic scattering
 - C. Spallation
 - D. Fission
- III. Reaction Cross Section
 - A. Total cross section
 - B. Partial cross section
 - C. Calculation of cross section
 - 1. Measurement of the beam
 - 2. Thin target
 - 3. Thick target
- IV. Reaction Mechanisms
 - A. Compound nucleus
 - B. Direct interaction
- V. Problems

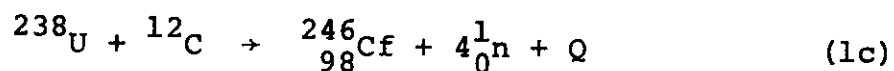
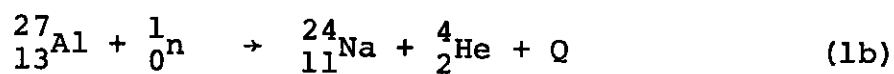
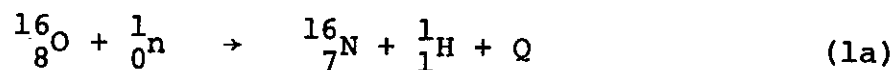
SECTION I

Introduction to Induced Nuclear Reactions

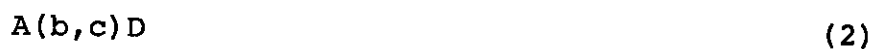
Induced Nuclear Reaction - Process by which changes are induced in nuclei by the interaction with other particles or sufficient energy.

A. Notation for nuclear reactions

1. Formal notation



2. Shorthand notation



A - target nucleus, irradiated material

b - projectile, bombarding particle

c - emitted particles

D - product or residual nucleus



B. Schematic reaction diagram

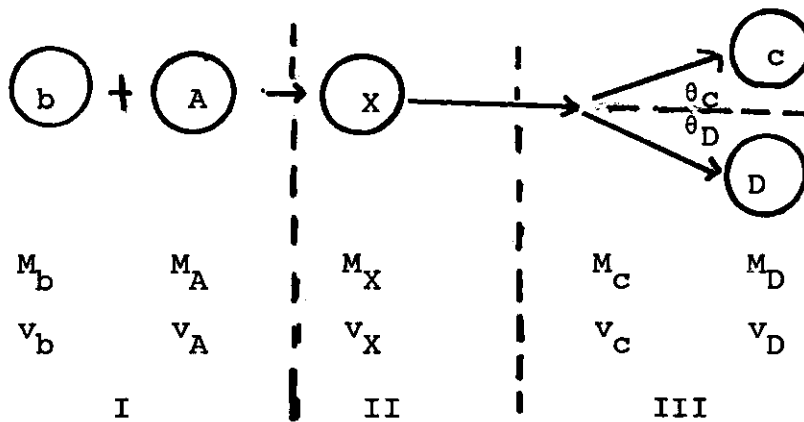


Figure 1.

I = collision stage

II = compound nucleus stage (maybe 10^{-14} s)

III = product formation stage

C. Important quantities conserved

1. Mass number; $A_A + A_b = A_C + A_D$ (3)

2. Nuclear charge; $Z_A + Z_b = Z_C + Z_D$ (4)

3. Total energy; $E_K = \text{kinetic energy}, E_M = \text{mass energy}$

$$(E_K + E_M^0)_A + (E_K + E_M^0)_b = (E_K + E_M^0)_C + (E_K + E_M^0)_D \quad (5)$$

4. Momentum: $M_A v_A + M_b v_b = M_X v_X = M_C v_C \cos \theta_C + M_D v_D \cos \theta_D$

$$\text{and } M_C v_C \sin \theta_C = M_D v_D \sin \theta_D \quad (7)$$

also, $M_b v_b = (M_A + M_b) v_X$ (assume $v_A = 0$)

$$E_{K(X)} = \left(\frac{M_A}{M_A + M_b} \right) E_{Kb} \quad (8)$$

5. Angular momentum.

D. Types of projectiles

1. Neutrons

Slow neutrons $E \leq 1 \text{ keV}$

Thermal neutrons; Maxwellian distribution of

velocities which at room temperature has a most probable energy, of 0.025 eV.

Fast neutrons; $E > 0.5 \text{ MeV}$

Obtain neutrons in nuclear reactors or special "neutron generators" which use reactions such as ${}^2\text{H}(d,n){}^3\text{He}$.

2. Charged Particles

Protons; ${}^1_1\text{H}$ or p

Deuterons; ${}^2_1\text{H}$ or d

Tritons; ${}^3_1\text{H}$ or t

Alpha particles; ${}^4_2\text{He}$ or α

Heavy ions; any charged particle heavier than α 's, such as ${}^3\text{Li}$, ${}^6\text{C}$, ${}^{36}\text{Kr}$, ${}^{54}\text{Xe}$.

Obtain charged particles from accelerators such as cyclotrons, synchrotrons, linacs, ~~hy~~lacs, etc.

E. Threshold Energy

E_{th} is minimum energy required for a nuclear reaction.

If $Q > 0$, exoergic, no threshold exists

If $Q < 0$, endoergic

$$E_{th} = -Q \left(\frac{M_b + M_a}{M_b} \right)$$

If b is projectile, then denominator should be M_a .

(9)

In bombardment by a charged particle such as p, d, α , or heavy ions, kinetic energy necessary to induce reaction $> E_{th}$ (due to coulomb repulsion)

For neutrons, no coulomb barrier so reaction can be induced by neutrons of zero kinetic energy.

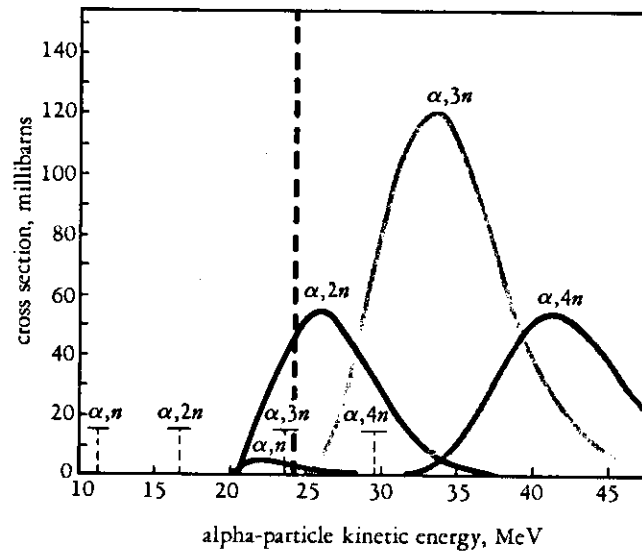


Fig. 2. Formation curves ("excitation functions") for $^{232}\text{Th} + \alpha$. Threshold energies are indicated by short dashed lines while the heavy dashed line indicates the calculated coulomb barrier.

SECTION II

Types of Nuclear Reactions

A. Elastic scattering

No change in identity of particle or target; target remains in ground state; total kinetic energy unchanged.



$$E_{K_A} + E_{K_b} = E'_{K_A} + E'_{K_b}$$

B. Inelastic scattering

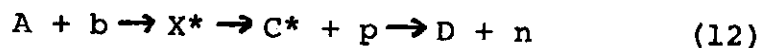
No change in identity of particle or target but target raised to excited state.



$$E_{K_A} + E_{K_b} > E'_{K_A} + E'_{K_b}$$

C. Spallation

After capture, nucleons emitted; e.g. $A(b, pn)D$:



D. Fission

After capture, nucleus splits into 2 nuclei.

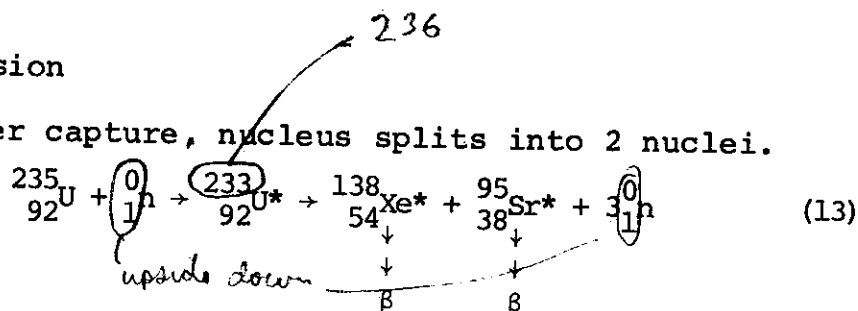
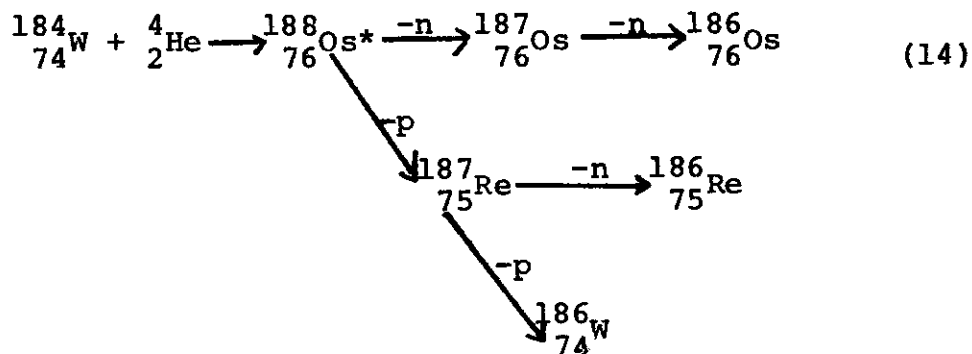
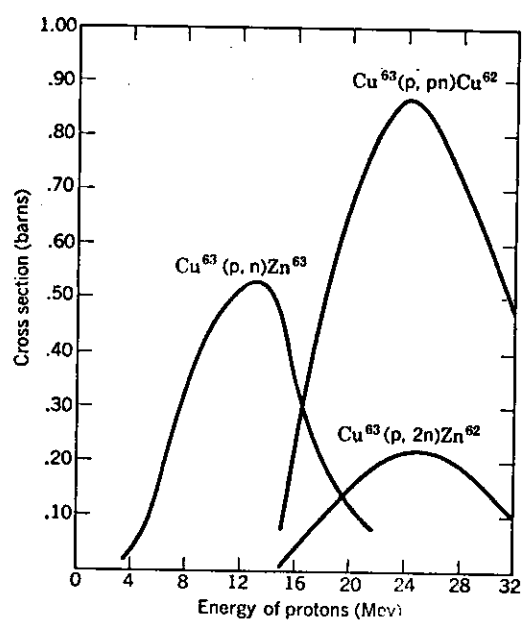


TABLE I. Approximate Fission Energy Balance

	Fission Energy, m.e.v.
Kinetic energy of fission fragments	170
Energy of gamma rays	16
Energy of beta decay	9
Kinetic energy of emitted neutrons	5
Total	200

E. Competing reactions.





At $E_p = 20$ MeV, ^{62}Cu , ^{62}Zn and ^{63}Zn all formed.

SECTION III

Reaction Cross Section

Cross section: probability of a nuclear reaction

Geometric cross sectional area of target nucleus

$$= \pi R^2 \text{ cm}^2 \quad (15)$$

assume $R = 1.4 \times 10^{-13} A^{1/3} \text{ cm}$, for $A = 200$:

$$\begin{aligned} \text{geometric cross section} &= (3.1416) (1.4 \times 10^{-13})^2 (200^{1/3})^2 \\ &= 2.1 \times 10^{-24} \text{ cm}^2 \end{aligned} \quad (16)$$

$$1 \text{ barn (b)} = 10^{-24} \text{ cm}^2$$

A. Total cross section

Sum of all reactions, both scattering and absorption

$$\sigma_T = \underbrace{\sigma_{\text{elastic}} + \sigma_{\text{inelastic}}}_S + \sigma_{\text{spallation}} + \sigma_{\text{fission}} + \text{etc} \quad (18)$$

B. Partial cross sections

Cross section for each individual reaction

See Figures 2 and 4.

C. Measurement of cross section

1. Beam intensity (for charged particles) or flux (for neutrons)

Charged particles:

$$1 \mu\text{A} = \frac{6.28}{n} \times 10^{12} \text{ particles/sec.}$$

$$\mu\text{C} = \mu\text{A} \times t \text{ (in sec.)}$$

*A convenient way to
do this*

Ex: calculate the particles/s of a $^{14}\text{N}^{+4}$ beam striking a target if the Faraday cup reading gives 7500 μC collected during a bombardment of 2 h.

$$\# \mu A = 7500 \mu C \times \frac{1}{2h} \times \frac{1}{60m} \times \frac{1}{60s} = 1.04$$

$$\# \text{particles/s} = 1.04 \times \frac{6.28 \times 10^{12}}{4} = 1.63 \times 10^{12} \quad (14)$$

2. "Thin" target - beam passes through target with very small attenuation

$$\frac{dN_i}{dt} = \phi_0 n \sigma_i X = N_1 \phi \sigma \quad (15)$$

$\frac{dN_i}{dt}$ - number of reactions occurring in target per unit time.

ϕ_0 - number of particles incident upon target per unit time.

n - number of target nuclei per cm^3 of target.

σ_i - cross section for the production of product (in cm^2).

X - target thickness in cm.

N_1 - $n \cdot X$ - total number target nuclei/ cm^2 .

$$N_i = N_1 \phi \sigma t \quad (16)$$

If product is radioactive with decay constant λ ,

$$\frac{dN_i}{dt} = N_1 \phi \sigma - \lambda_1 N_i \quad (17)$$

$$A_i = N_1 \phi \sigma (1 - e^{-\lambda_1 t}) \quad (18)$$

N.B. Charged Particles: normally express N_1 in number target atoms/ cm^2 and ϕ in particles/s (Beam is smaller than target)

Neutrons: normally express N_1 in total number target atoms and ϕ in neutrons/ cm^2/s . (Beam is larger than target)

3. Thick target - beam attenuated or completely stopped in target.

The loss in intensity of flux through thickness N is:

$$-d\phi = \phi n \overline{\sigma_i} dN \quad (19)$$

Integrating we get:

$$\phi_x = \phi_0 e^{-n\overline{\sigma_i} x} \quad (20a)$$

or

$$\phi_0 - \phi_x = \phi_0 (1 - e^{-n\overline{\sigma_i} x}) = N_i \quad (20b)$$

ϕ_0 = initial intensity

ϕ_x = intensity at thickness x

SECTION IV

Reaction Mechanisms

A. Compound nucleus mechanism

1. Target and projectile fuse
2. Excitation energy equilibrated among nucleons of compound nucleus
3. Excitation energy removed by nucleon evaporation and decay path roughly independent of formation path

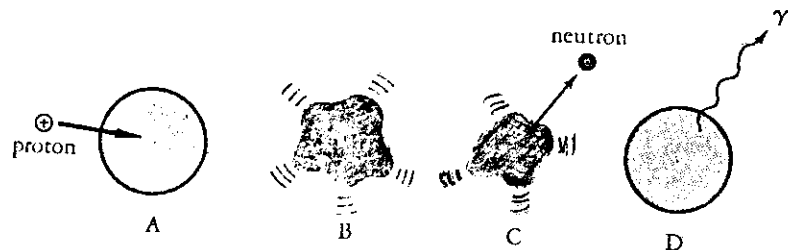


Figure 5. A simple representation of the formation of an excited compound nucleus B and its subsequent deexcitation by neutron evaporation C and gamma ray emission D.

B. Direct interaction mechanism

1. Projectile initially interacts with individual nucleons in target
2. After initial interaction, enters compound nucleus stage

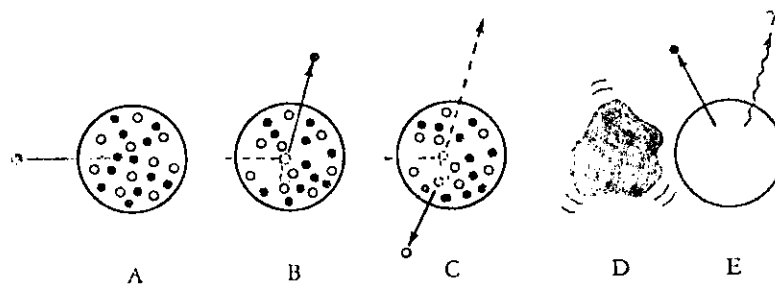
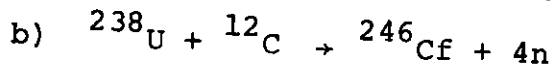
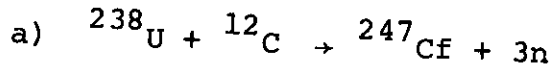


Figure 6. A simple representation of a high energy reaction in which a neutron (B) and a proton (C) are directly knocked out before formation of the compound nucleus D and subsequent deexcitation by evaporation and gamma ray emission E.

SECTION V

Problems

1. Calculate the energy thresholds for the following reactions:

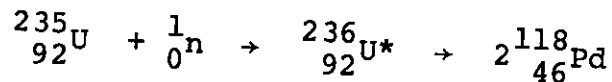


$$M_{^{238}\text{U}} = 238.1245; \quad M_{^{246}\text{Cf}} = 246.1451; \quad M_{^{247}\text{Cf}} = 247.1476;$$

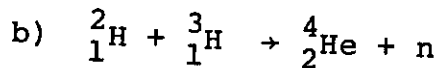
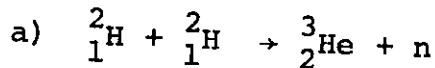
$$M_{^{12}\text{C}} = 12.0038.$$

2. Calculate the coulomb barrier for $^{238}_{92}\text{U} + ^{12}_6\text{C}$ using $R_0 = 1.4 \times 10^{-13}$ cm. Compare this answer with that of Problem 1.

3. Use Fig. 1 of Part 1 to estimate the energy release in nuclear fission in the reaction



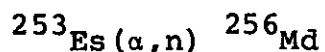
4. Calculate the energy released in the following thermo-nuclear reactions:



$$M_{^2\text{H}} = 2.0147; \quad M_{^3\text{H}} = 3.0170; \quad M_{^3\text{He}} = 3.0170; \quad M_{^4\text{He}} = 4.0038$$

5. How long must 25 g of cobalt be irradiated in a neutron flux of 10^{13} n/cm²/s in a reactor to produce 1000 Ci of ^{60}Co ? The $\sigma(n, \gamma)$ for ^{59}Co is 30.5 b and $t_{1/2}$ (^{60}Co) is 5.2y; assume no attenuation of the flux.

6. In a thick target experiment, a 0.01/cm thickness of Cd causes a 69% reduction in intensity of a neutron beam passing through it. The density of Cd is 8.65g/cm^3 . What is the cross section for Cd to these neutrons?
7. In planning the discovery experiments of element 101, Md, it was known that a thin target of only 10^9 atoms of ^{253}Es was available. The cyclotron could provide a beam of $100\mu\text{A}$ and the estimated cross section was 1 mb for the reaction:



How long an irradiation time was required per atom of ^{256}Md (ignore its decay)?

Part IX

Radionuclide Production and Activation Analysis

Outline

I. Radionuclide Production

- A. Reactor Production
- B. Production by Particle Accelerator
- C. Choice of Mode of Production
- D. Qualitative Aspects of Radionuclide Production
 - 1. Rate of Production of Desired Nuclide
 - 2. Rate of Decay of Product after Irradiation
 - 3. Determination of Necessary Irradiation Time

II. Activation Analysis (AA)

- A. Basic Activation Equation
- B. Comparative Method
- C. Types of AA
 - 1. Instrumental Activation Analysis (IAA)
 - 2. Radiochemical Activation Analysis (RAA)
 - 3. Comparison of IAA and RAA
- D. Typical Steps in AA
 - 1. Design of Experiment
 - 2. Sample Preparation
 - 3. Irradiation
 - 4. Chemical Isolation
 - 5. Measurement of Activities
- E. Advantages and Disadvantages of AA
- F. Applications of AA
 - 1. Biological
 - 2. Forensic Science
 - 3. Industrial
 - 4. Environmental

G. Charged Particle Activation Analysis

III. Problems

SECTION I

Radionuclide Production

A. Reactor Production: Radionuclides produced by neutron irradiation or by neutron induced fission. Nuclei produced are neutron rich so decay by β^- emission.

1. can irradiate more than one sample simultaneously
2. can irradiate large samples
3. thermal neutron capture cross-sections are relatively high;

$$\sigma_n \propto \frac{1}{E_n} \quad (1)$$

(thermal neutrons-relatively low energy neutrons with a thermal (Boltzmann) distribution of energies)

4. product nuclei are usually isotopic with target
5. fission products can be carrier free (not diluted by non-radioactive isotopes,

B. Production by Particle Accelerator: Radionuclides produced by bombardment of charged particles with emission of one or more light particles (neutrons,

protons, deuterons, etc.) Nuclei produced are neutron deficient and decay by E.C. or β^+ emission.

Characteristics:

1. only one sample irradiated at a time
2. only surface of solid sample is irradiated
3. charged particle reaction cross-sections are much lower than thermal neutron capture (σ_n , γ) cross section
4. product nuclei are usually not isotopic with target material

C. Choice of Mode of Radionuclide Production

1. Considerations involving time
 - a. distance from production site and shipping time
 - b. length of time of separation and purification
 - c. length of time of experiment
2. Consideration of specific activity
 - a. carrier free activity requires accelerator or fission produced activity

3. Consideration of cost

- a. economically, reactor produced activities are preferred

D. Quantitative Aspects of Radionuclide Production

Table of Notation

X - specific isotope of element X involved in the production of the desired radionuclide

Y - desired nuclide produced in the reaction Y may or may not be the same element as represented by X

N_X - number of atoms of X in a sample of w g. of the element of atomic weight M ; the total number of atoms of the element multiplied by the fractional isotopic abundance, f , of X.

$$N_X = 6.02 \times 10^{23} \cdot \frac{w}{M} \cdot f \quad (2)$$

N_Y - number of atoms of Y

λ - decay constant of Y in sec^{-1}

σ - cross section for the production of Y from X in cm^2

Φ - particle flux from reactor or accelerator

T - irradiation time

t - time after end of irradiation

1. Rate of Production of Desired Nuclide

$$A_Y = N_X \sigma \phi (1 - e^{-\lambda T}) \quad (3)$$

a. if $T \gg t_{1/2}$

$$A_Y = N_X \sigma \phi \quad (3a)$$

b. If $T \ll t_{1/2}$

$$A_Y = N_X \sigma \phi \lambda t \quad (3b)$$

c. Units of N_X and ϕ

TABLE I

	Reactor	Accelerator
N_X	Total number of target atoms	Total number of target atoms/cm ²
ϕ	neutrons/cm ² /sec	charged particles/sec

2. Rate of Decay of Product after Irradiation

$$A_Y = N_X \sigma \phi (1 - e^{-\lambda T}) e^{-\lambda t} \quad (4)$$

3. Determination of Necessary Irradiation Time

$(1-e^{-\lambda T})$ - saturation factor

When $T = t_{1/2}$ 50% saturation

$T = 2t_{1/2}$ 75% saturation

$T = 3t_{1/2}$ 87% saturation

Usually irradiate for 1 or 2 half lives

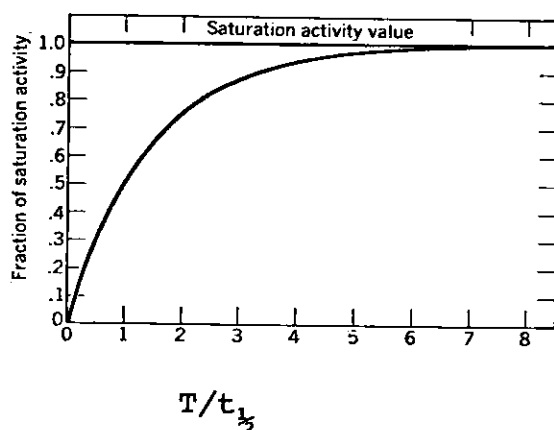


FIG. 1. The rate of approach to saturation in production of a radioactive species as a function of the irradiation time in half life units.

SECTION II

Activation Analysis (AA)

A method of elemental analysis using formation of radio-nuclides; neutron irradiation (neutron activation analysis, NAA) most common, but irradiation by various charged particles also used.

A. Basic Activation Equation

$$W = \frac{A \cdot M}{\sigma \Phi f (1 - e^{-\lambda T}) (6.02 \times 10^{23})} \quad (5)$$

W = weight of element irradiated

A = induced activity in dps at end of irradiation

M = atomic weight of that element

f = fractional isotopic abundance of the nuclide acting
as target

B. Comparative Method

Difficult to know cross section, σ , and flux, Φ , accurately
so use comparison with activity produced in standard sample.

$$\frac{\text{Weight of element in unknown}}{\text{Weight of element in standard}} = \frac{\text{Activity of element in unknown}}{\text{Activity of element in standard}} \quad (6)$$

Measurements are relative in both the production and detection steps. Some standards available from NBS (Table 2).

TABLE 2

Composition values for NBS-SRM 614, 615; trace elements in a glass matrix, 1 ppm from certificate of analysis (revised Aug. 3, 1972).

Element	Value
Antimony	1.06
Boron	1.30 \pm 0.2
Cadmium	0.55
Cobalt	0.73 \pm 0.02
Copper	1.37 \pm 0.07
Europium	0.99 \pm 0.04
Gallium	1.3
Gold	0.5
Iron	13.3 \pm 1
Lanthanum	0.83 0.02
Lead	2.32 \pm 0.04
Nickel	0.95
Potassium	30 \pm 1
Rubidium	0.855 \pm 0.005
Scandium	0.59 \pm 0.04
Silver	0.42 \pm 0.04
Strontium	45.8 \pm 0.1
Thallium	0.269 \pm 0.005
Thorium	0.748 \pm 0.006
Titanium	3.1 \pm 0.3
Uranium	0.823 \pm 0.002

C. Types of AA

1. Instrumental Activation Analysis (IAA)

The radionuclides produced in the activation are identified instrumentally by detecting their characteristic gamma spectra using Ge(Li) or NaI (Tl) spectrometry.

2. Radiochemical Activation Analysis (RAA)

The desired radionuclides are isolated using radiochemical techniques and counted.

3. Comparison of IAA and RAA

IAA is non-destructive, lends itself to the measurement of short-lived activities and to automated analysis.

RAA - original sample is altered; RAA may be necessary if interfering radionuclides prevent accurate measurement of the desired activities by IAA.

D. Typical Steps in AA

1. Design of experiment

a. Choice of nuclear reaction based on

- 1) reaction cross section
- 2) particle flux, ϕ , available at irradiation facility
- 3) decay scheme of desired activity
- 4) consideration of possible interfering reactions

b. Determination of irradiation time

2. Sample preparation

a. Sample and standard should be prepared in exactly the same manner, i.e. same chemical treatment.

b. Weights of sample and standard should be the similar and known accurately.

3. Irradiation of both sample and standard

4. Chemical isolation of activities of interest, if necessary

5. Measurement of activities in sample and in standard by appropriate detection system
 - a. short-lived nuclides: must count immediately after irradiation
 - b. Longer lived nuclides: irradiate for long periods, allow short lived activities to decay before counting
 - c. If γ ray counting, should follow decay of two γ rays in decay sequence of nuclide to verify radiochemical purity

E. Advantages and Disadvantages of AA

Advantages

1. Trace analysis - micro-to pico-gram (10^{-6} - 10^{-12} g) sensitivity level
2. Simultaneous determination of several elements possible
3. Can be nondestructive
4. Often accuracy of few percent at nanogram level
5. Freedom from reagent and laboratory contamination concerns

1. Cost of irradiation
2. Expensive equipment required
3. Precaution required for working with higher than normal level of radioactivity

Sensitivities are expressed as the micrograms of the naturally occurring element that must be present in the entire sample to be detected and determined by the Activation Analysis Service at General Atomic. By special arrangement, the sensitivities of many of the elements can be increased up to 100-fold. Sensitivities are for interference-free conditions.

Activation analysis sensitivities

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1 H NA																2 He NA	
3 Li 0.0008 _p	4 Be 15 _p																10 Ne 2
11 Na 0.004	12 Mg 0.5																18 Ar 0.002
19 K 0.2	20 Ca 4	21 Sc 0.001	22 Ti 0.1	23 V 0.002	24 Cr 0.3	25 Mn 0.0001	26 Fe 2 FS	27 Co 0.01	28 Ni 0.7	29 Cu 0.002	30 Zn 0.1	31 Ga 0.002	32 Ge 0.1	33 As 0.005	34 Se 0.01	35 Br 0.003	36 Kr 0.01
37 Rb 0.02	38 Sr 0.005	39 Y 0.4	40 Zr 0.8	41 Nb 3	42 Mo 0.1	43 Tc NA	44 Ru 0.04	45 Rh 0.005	46 Pd 0.03	47 Ag 0.004	48 Cd 0.005	49 In 0.00006	50 Sn 0.03	51 Sb 0.007	52 Te 0.03	53 I 0.002	54 Xe 0.1
55 Cs 0.001	56 Ba 0.02	57 ^L La 0.005	72 Hf 0.0006	73 Ta 0.1	74 W 0.004	75 Re 0.0008	76 Os 1	77 Ir 0.0003	78 Pt 0.1	79 Au 0.0005	80 Hg 0.08	81 Tl 1 _b	82 Pb 0.5 _p	83 Bi 1 _b	84 Po NA	85 At NA	86 Rn NA
87 Fr NA	88 Ra NA	89 ^A Ac NA															
			58 Ce 0.2	59 Pr 0.03	60 Nd 0.03	61 Pm NA	62 Sm 0.001	63 Eu 0.0001	64 Gd 0.007	65 Tb 0.03	66 Dy 0.00003	67 Ho 0.003	68 Er 0.002	69 Tm 0.2	70 Yb 0.02	71 Lu 0.0003	
			90 Th 0.2	91 Pa NA	92 U 0.003	93 Np NA	94 Pu NA	95 Am NA	96 Cm NA	97 Bk NA	98 Cf NA	99 Es NA	100 Fm NA	101 Md NA	102 No NA	103 (Lw) NA	

Key

Atomic number → 33 As ← Symbol

0.005 ← Sensitivity in micrograms

FS = Fast neutron flux

FS - Fast neutrons Fission spectrum, b - Beta count. p - Reactor pulse;
c - Bremsstrahlung radiation required. NA - Analysis not normally performed at GGA

FIG. 2. Table of activation analysis sensitivities as offered by the General Atomic Company, San Diego, Calif.

D. Applications of AA

1. Biological: trace elements in living systems, in vivo analysis, toxicology, metabolic and pathological studies.
2. Forensic science
3. Industrial: quality control, trace elements in metals, foods, semiconductors, etc.
4. Environmental: determination of trace elements in water, sediments, rocks, aerosols, etc.

TABLE 3

Estimated Detection Limits of Instrumental
Neutron Activation Analysis of Biological
Material ($\mu\text{g/g}$ Dry Tissue)

Element	*Typical Concentration	
	In Marine Organisms	Detection Limit**
Na	500-1500	0.05
K	1000-30,000	20
Rb	0.5-8	1
Cs	0.02-0.4	0.0001
Fe	1-500	2
Zn	10-200	0.5
Br	10-200	0.05
As	<1-50	0.3
Cd	<0.1-10	1
Ag	<0.001-5	0.001
Co	0.001-0.5	0.001
Cr	<0.02-1	0.02
Hg	0.05-5	0.02
Se	0.5-50	0.05
Sb	0.0001-0.05	0.001
Sc	0.00001-0.002	0.00005

* Typical Ranges in various tissues, with the low ranges usually associated with muscle and high ranges with liver.

** 300 mg of freeze-dried tissue; integral thermal neutron exposure of 3×10^{17} n/cm²; 20 min. and 1000 min. counts after decay periods of 3 to 5 days and 20 to 30 days, respectively; 20 to 40 cc Ge(Li) diode detection, except where specified.

TABLE 4

Estimated Minimum Detectable Concentrations of Pollutant Elements in Seawater by INAA and by NAA with Separations

Trace Element	Typical Reported Concentrations in Open Ocean (micrograms/liter)	Minimum Detectable Concentrations (in micrograms/liter)	
		INAA*	NAA with Separations**
Hg	0.02-0.2	0.05	0.001
Cd	0.06-0.7	16,000	0.001
Ag	0.002-0.05	1.0	0.003
As	2-3	Not Possible	0.0001
Cu	0.5-2	Not Possible	0.002
Cr	0.02-0.6	0.03	0.003
Zn	0.5-10	0.02	0.01
Sn	0.02	Not Possible	9
Se	0.08	0.2	0.02
Sb	0.2	0.02	0.00003

* 25 ml seawater; 24 hour irradiation at 10^{13} n/cm²/sec; 40 days decay; 1000 minute count on 20 cc Ge(Li) diode detector; based on 3 σ above Bkg-Compton contribution in peak areas.

** 500 ml seawater; elements chemically separated; 24 hour irradiation at 10^{13} n/cm² sec; 3 days decay; 500 minute count on a 20 cc Ge(Li) diode detector; based on twice Bkg contribution in peak areas.

TABLE 5

Estimated Detection Limits for the INAA of Trace Elements In Marine Sediments

Element	Typical Concentration Ranges in Marine Sediments ($\mu\text{g/gm}$)	INAA Sensitivity* ($\mu\text{g/gm}$)
Ag	0.01-0.5	0.1
Al	10,000-90,000	10
As	2.20	1
Ba	60-8100	100
Ce	40-70	5
Cd	205	10
Co	1-200	0.1
Cr	10-200	0.8
Cs	0.3-15	0.2
Cu	10-700	1
Dy	0.2	0.1
Eu	0.2-10	0.05
Fe	20,000-60,000	200
Hf	0.1-18	0.1
Hg	0.05-3	0.5
K	3000-30,000	5000
La	2-60	2
Lu	0.2	0.1
Mn	100-10,000	10
Na	2000-40,000	100
Pb	10-200	---
Rb	1-100	1**
Sb	0.5-15	0.1
Sc	0.2-30	0.02
Se	0.1-1	2
Em	0.5-30	0.3
Sn	0.5-15	500
Sr	200-2000	100**
Ta	0.03-3	0.01

TABLE 5 (con't.)

Tb	0.1-7	0.1
Th	0.3-10	0.08
V	10-500	10
Yb	1	0.3
Zn	5-4000	5**
Zr	100-400	70

* 100 to 800 mg of dried sediment; samples irradiated at optimum intervals ranging from 1 minute to 6 hours in a flux of 10^{11} to 10^{13} n/cm²/sec and counted at optimum intervals after the irradiation for 1 minute to 200 minutes; Ge(Li) diode detectors, 20 cc to 60 cc volumes

** Determined by counting on a coincidence-anticoincidence shielded Ge(Li) gamma ray spectrometer.

F. Charged Particle Activation Analysis

1. Usually used for elements of $Z < 10$; limited to surface analysis.

2. Reactions used:

(p,n)	(d,n)	(³ He,d)
(p,pn)	(d,2n)	(³ He,α)
(p,α)	(d,p)	

TABLE 6

Detection Limits for $^3\text{He}^\dagger$

<u>Element</u>	<u>Limits (ppm)</u>	<u>Surface Concentration ($\mu\text{g}/\text{cm}^2$)</u>
B	0.05	0.02
C	0.3	0.01
N	1.0	--
O	0.5	0.01
F	--	0.1

† 5 MeV ^3He , 0.5 μamp , 10 m irradiation

SECTION III

Problems

1. The disintegration rate of a nucleus produced in a reactor or an accelerator as a function of irradiation time is given by

$$A = \phi \sigma N_x (1 - e^{-\lambda T})$$

Discuss the variation of the saturation factor with T for the following cases

a) $T \gg t_{1/2}$

b) $T \ll t_{1/2}$

[Hint: e^{-x} where x is a large number $\rightarrow 0$, and e^{-x} where x is a small number can be approximated by $(1 - x)$]

2. The saturation activity is defined as the maximum activity in dpm that can be obtained for 1g of a given element in a reactor of known flux. Assuming a neutron flux of 10^{12} neutrons/cm²/sec calculate the saturation activities in dpm/ μ g for the following elements

Target isotope	Abundance %	$t_{1/2}$ of product	σ for (n, γ) in barn
a) ^{55}Mn	100	2.58h	13.3
b) ^{70}Zn	0.62	2.2 m	0.085
c) ^{127}I	100	25 m	5.6
d) ^{63}Cu	69.1	12.8 h	4.3

3. A target containing 10^{-4} g of ^{55}Mn was irradiated for four hours in a reactor with a neutron flux of 10^{13} neutron/cm²/sec. If the target was counted two hours after the irradiation was completed what would be the activity in dpm?
4. A sample of ^{127}I was irradiated ($I = 10^{12}$ neutrons/cm²/sec) for twice the half life of the product nucleus ^{128}I and then counted immediately on a counter which was 25% efficient. The observed count rate was 1.4×10^8 cpm. How much ^{127}I was originally present in the sample?
5. The activity of a 10^{-3} g sample of ^{70}Zn was found to be 10^5 dpm after irradiation for 4 minutes. Calculate the neutron flux of a reactor in neutron/cm²/sec.
6. Two samples containing ^{63}Cu are irradiated in a reactor. One of the samples is known to have 10^{-5} g of ^{63}Cu . The sample of known ^{63}Cu concentration was observed to have an activity of 1.7×10^4 dpm. The activity of the other sample determined on the same counter was found to be 0.9×10^4 dpm. What is the weight in grams of ^{63}Cu in the unknown sample? If two different detectors had been used for the counting of the samples how would you have corrected your data?

7. For the production of the following nuclides, choose and explain the basis for choice between cyclotron irradiation, reactor irradiation or fission.

^{137}Cs

^{35}S

^{14}C

^{197}Hg

^{147}Pm

^{36}Cl

^{58}Co

^{58}Co

8. The cross section of the product nucleus can be sufficiently large that second order neutron capture products are formed during irradiation of moderate duration. If the first order product is radioactive, then its concentration at any time is dependent on the decay constant and the cross section for the production of the second order product as well as the cross section for its own production. A destruction constant Λ_2 may be defined as

$$\Lambda_2 = (\lambda_2 + \phi\sigma_2)$$

The number of atoms of the second-order product at a time T is given by

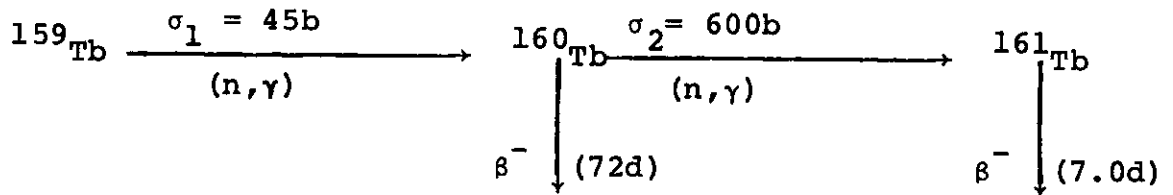
$$N_3 = \Lambda_1 \Lambda_2^* N_1^0 \left[\frac{e^{-\Lambda_1 T}}{(\Lambda_2 - \Lambda_1)(\Lambda_3 - \Lambda_1)} + \frac{e^{-\Lambda_2 T}}{(\Lambda_1 - \Lambda_2)(\Lambda_3 - \Lambda_2)} + \frac{e^{-\Lambda_3 T}}{(\Lambda_1 - \Lambda_3)(\Lambda_2 - \Lambda_3)} \right]$$

where $\Lambda_1 = \phi\sigma_1$

$$\Lambda_3 = \lambda_3$$

$$\Lambda_2^* = \phi\sigma_2$$

If a milligram of Tb_2O_3 is bombarded for 30d at a flux of 10^{14} neutrons/cm²/sec the following processes occur:



Calculate the activities of both ${}^{160}\text{Tb}$ and ${}^{161}\text{Tb}$.

9. In the same manner calculate what percent of the total activity at end of the bombardment in a 1 gram sample of gold foil will be due to ${}^{199}\text{Au}$ following a 1.5d irradiation with a flux of 10^{14} neutrons/cm²/sec.
 $[\sigma_{197}=96\text{b}, \sigma_{198}=3.5 \times 10^4\text{b}, t_{1/2}(198)=2.7\text{d}, t_{1/2}(199)=3.15\text{d}]?$
10. In a flux of 10^{13} neutrons/cm²/sec, how long an irradiation is necessary to reduce the amount of ${}^{186}\text{W}$ ($\sigma=37$ barns, 28.4% abundant) present in tungsten foil by 10%?

PART X

Tracer Methodology

Outline

- I. Basic Assumptions of Tracer Use
- II. Self Decomposition of Labelled Compounds
- III. Radiometric Analysis
 - A. Precipitation
 - B. Titration
- IV. Isotope Dilution Analysis
 - A. Direct Isotope Dilution
 - B. Reverse Isotope Dilution
 - C. Derivative Isotope Dilution
 - D. Substoichiometric Dilution Analysis
 - E. Summary of Requirements for and Advantages of Isotope Dilution
- V. Applications of Radiotracers
 - A. Chemical Separations
 - B. Solubility Studies
 - C. Solids
 - 1. Surface Area
 - 2. Diffusion
 - D. Kinetics and Mechanisms
 - 1. Hydrogen Migration
 - 2. Isotope Exchange
 - 3. Photosynthesis
 - E. Structural Studies
 - F. Geochronometry, Isotope Dating
 - 1. Requirements
 - 2. The Lead Method
 - 3. Radiocarbon Dating
- VI. Problems

SECTION I

Basic Assumptions for Tracer Use

1. Radioisotopes are chemically identical with stable isotopes of the same element.
2. Radioactive nature of the isotope does not change the chemical or physical properties.
3. No observable radiolysis; i.e., no observable perturbation in the chemical behavior of a system containing radioactive atoms as a result of chemical products formed in the system by the absorption of the decay energy.

NOTE: If tracer decays to radioactive daughter, must either:

- a. purify tracer before use and complete experiment and counting before significant amount of daughter activity grows in; or
- b. count tracer when parent and daughter are in equilibrium.

SECTION II

Self Decomposition of Labelled Compounds

Doses of 10^7 rads produce decomposition effects of the order of 1%.

For ^{14}C compounds if 1 millicuries per millimole in one year have ca. 10^7 rads

For ^3H compounds if 8 millicuries per millimole in one year have ca. 10^7 rads.

SECTION III

Radiometric Analysis

- A. Precipitation: a slight excess of radioactive reagent added to a solution of the unknown to precipitate the unknown quantitatively.

Ex: Analysis of Zn(II)

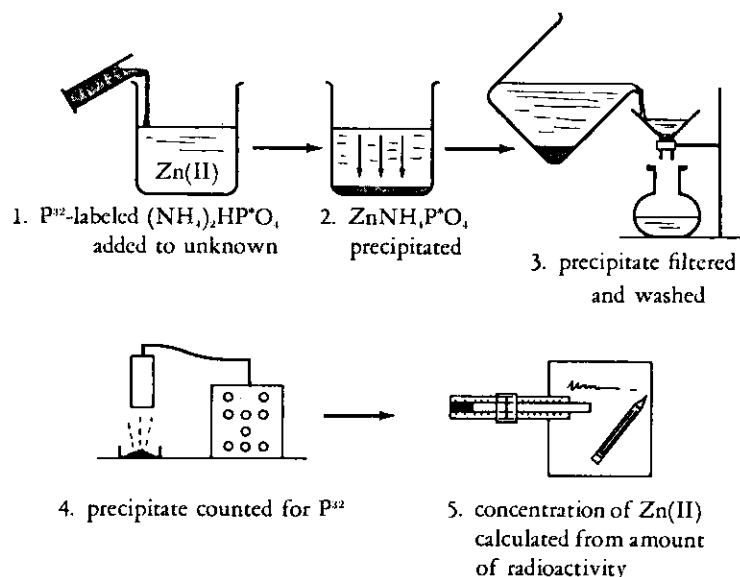


FIG. 1. Steps involved in the radiometric analysis of an unknown concentration of Zn(II) ions by use of a solution of ^{32}P -labelled $(\text{NH}_4)_2\text{HPO}_4$.

- B. Titration: place identical aliquots of unknown solution in a series of test tubes; varying aliquots of precipitation (or extracting) reagent added. After precipitation, supernatant counted and "titration" curve plotted.

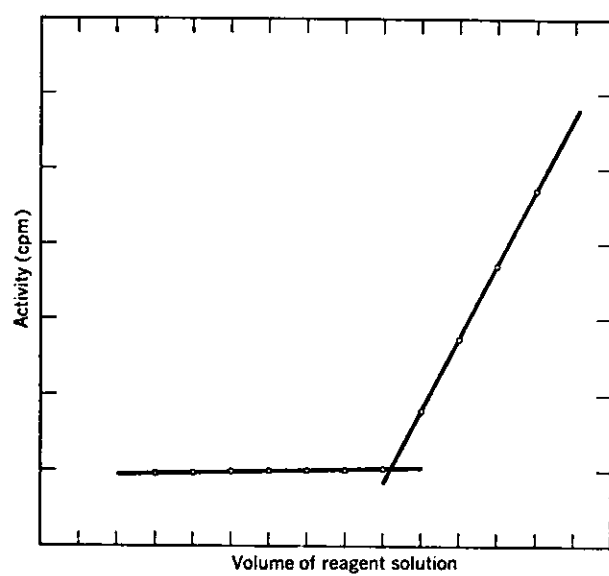


FIG. 2. Titration curve using radiometric analysis.

SECTION IV

Isotope Dilution

Valuable method for the analysis of complex mixtures of organic compounds.

A. Direct Isotope Dilution

Determination of the quantity of a particular compound in a mixture by the addition of a known amount of the same compound containing a radioactive label.

Method:

1. Addition of a known amount, W_a , of the radioactive compound with specific activity SA_a

$$SA_a = A_a / W_a \text{ (cpm/g)} \quad (1)$$

where A_a is the activity of the added radioactive compound in counts/min

2. Original sample and added W_a well mixed
3. Isolation of the desired compound in high purity but not necessarily quantitatively

4. Determination of the weight, W_f , and specific activity, SA_f , of the isolated pure compound

$$SA_f = A_f/W_f \quad (\text{cpm/gm}) \quad (2)$$

5. Calculation of the weight of the desired compound, W_i , in the original mixture

$$W_i = W_f \left[\frac{SA_a}{SA_f} - 1 \right] \quad (3)$$

Note: When the specific activity of the added radioactive compound, SA_a , is very high, $W_a \ll W_i$ and

$$W_i = W_f \left[\frac{SA_a}{SA_f} \right] \quad (4)$$

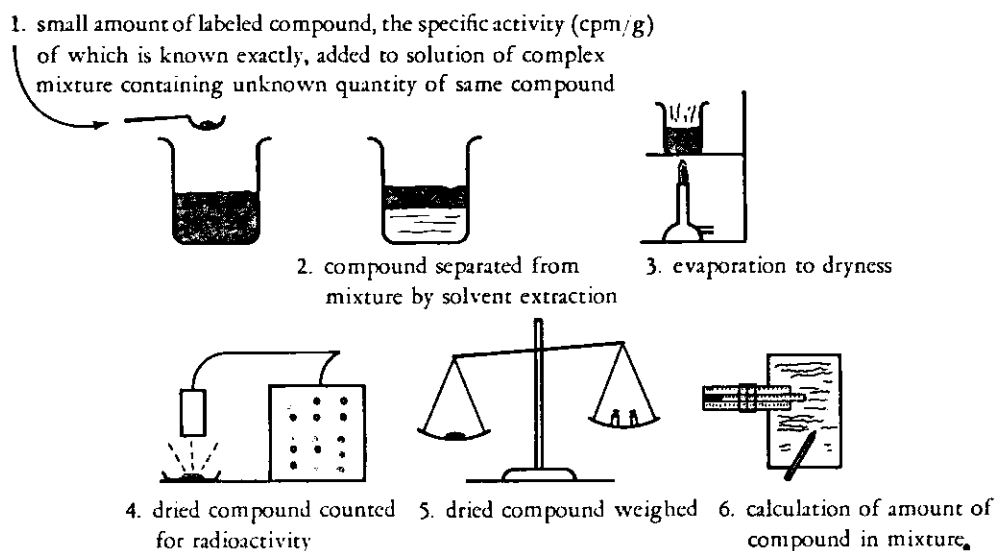


FIG. 3. Possible sequence of steps for determining the amount of one compound in a complex mixture by the technique of isotope dilution.

B. Reverse Isotope Dilution

Determination of a radioactive compound by the addition of the non-radioactive compound.

Method:

1. Determine the activity, A_1 , of the radioactive compound in the mixture

2. Mix in known amount, W_2 , of the stable compound
3. Again a high purity but not necessarily quantitative separation is performed yielding W_f of the pure compound
4. Measurement of the activity of the separated compound, A_2
5. Calculation of the amount, W_1 , of radioactive compound present in original mixture

$$W_1 = W_f \left(\frac{SA_1}{SA_2} \right) - W_2 \quad (5)$$

C. Derivative Isotope Dilution

A derivative of the unknown compound is made in high yield. A small sample of labelled derivative of known specific activity is added and the isotope dilution technique followed. Mathematically same as reverse isotope dilution.

D. Substoichiometric Dilution Analysis

Isolate equal but substoichiometric amounts of diluted and undiluted substances and compare activities. Can replace SA_1 and SA_2 in Eq. (5) by activities A_a and A_f .

E. Summary of Requirements and Advantages of Isotope Dilution

1. Requirements

Labelled compound must be chemically and radio-chemically pure

The labelled compound and analogous unlabelled compound in the mixture must be in complete equilibrium

Separation must be complete, high purity, but not necessarily quantitative

2. Advantages

Quantitative isolation unnecessary

Simple counting instrumentation sufficient

SECTION V

Application of Radiotracers

A. Chemical Separations

Can determine yield in a separation by adding initially a known amount of radiotracer (either isotopic or sufficiently similar chemically). Measure amount of activity after separation process.

$$\text{Yield (\%)} = \frac{\text{activity recovered}}{\text{activity added}} \times 100\%$$

B. Solubility Studies

Can measure K_{sp} by adding tracer before precipitation of insoluble compound, then determining activity in filtrate.

C. Solids

1. Surface area: add tracer to slurry of solid, allowing exchange or sorption by surface; e.g. use ^{212}Pb to measure surface area of PbSO_4 :

$$\frac{{}^{212}\text{Pb}_{\text{surface}}}{{}^{212}\text{Pb}_{\text{solution}}} = \frac{\text{Pb}_{\text{surface}}}{\text{Pb}_{\text{solution}}}$$

Found $5 \times 10^3 \text{ cm}^2/\text{g PbSO}_4$ as surface area

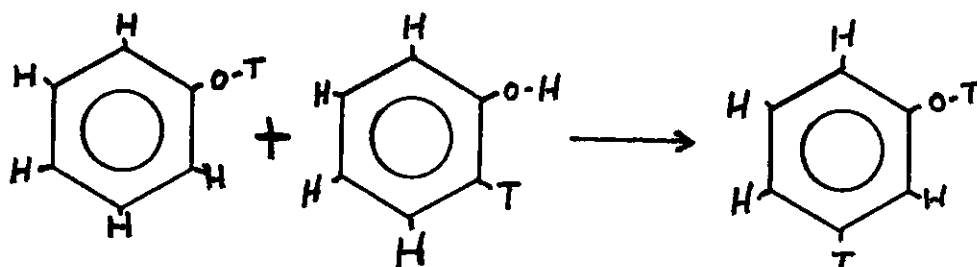
2. Diffusion: add tracer to surface, after period of time, solid divided into sections of known thickness, X , and their specific activity (SA) measured.

Diffusion coefficient, D , given by

$$\frac{d(\text{SA})}{dt} = \frac{Dd^2(\text{SA})}{dx^2}$$

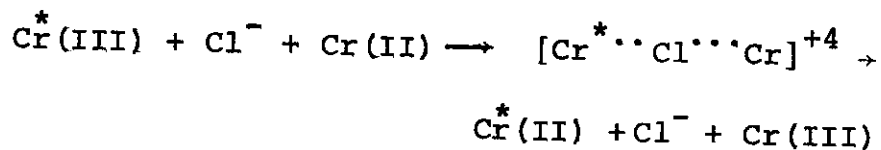
D. Kinetics and Mechanisms

1. Hydrogen Migration



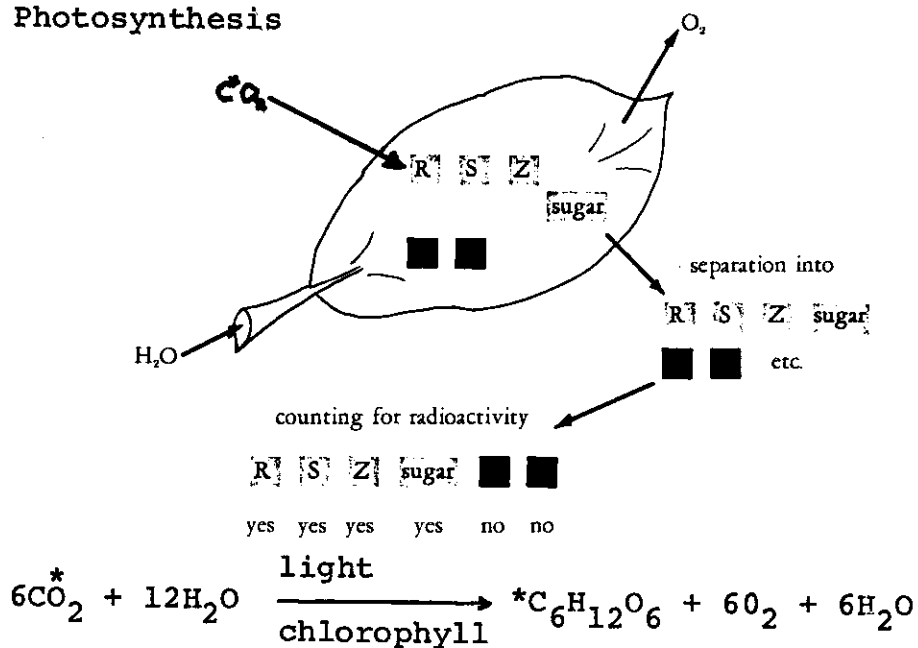
(T=tritium)

2. Isotopic Exchange

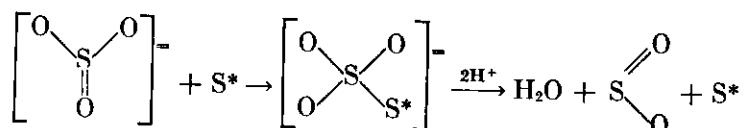


(6)

3. Photosynthesis



E. Structural Studies



F. Geochronometry, Isotope Dating

The decay of naturally occurring radioisotopes can be used for the measurement of age.

1. Requirements

- Decay rate of nuclide must be known
- Radioactive equilibrium must be established

- c. Sampling must be representative of material to be dated
- d. Analytical methods must be accurate
- e. The isotopic distribution must not have been altered by external processes

The range of applicability of a given method is approximately $10 T_{1/2}$ of the parent

2. The Lead Method

Based on the decay of uranium and thorium

	Establishment of equilibrium (years)
$^{238}\text{U} \rightarrow ^{206}\text{Pb} + 8\ ^4\text{He}$	10^6
$^{235}\text{U} \rightarrow ^{207}\text{Pb} + 7\ ^4\text{He}$	10^5
$^{232}\text{Th} \rightarrow ^{208}\text{Pb} + 6\ ^4\text{He}$	10^2

Example of calculation of age by Pb method

Assume a mineral which originally contained uranium but no thorium or lead. After a time t long enough for equilibrium (10^6 y for $^{238}\text{U} \rightarrow ^{206}\text{Pb}$), the ratio of ^{206}Pb to ^{238}U atoms is:

$$\frac{N_{206}}{N_{238}} = e^{\lambda_{238}t} - 1$$

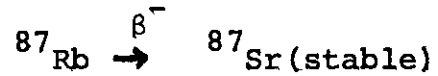
$$\begin{aligned}\lambda_{238} &= \text{decay constant for } ^{238}\text{U} \\ &= 0.154 \times 10^{-10}\end{aligned}$$

$$\text{For } t < 1.2 \times 10^9 \text{ y}$$

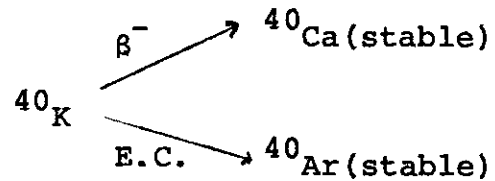
$$t = (N_{206}/N_{238})/\lambda_{238}(\text{y})$$

3. Rb and K Methods

Similar equations for use of



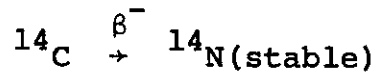
and



4. Radiocarbon Dating

Used for dating relatively young samples

1000 to 30,000 years



$$\lambda(^{14}\text{C}) = 1.245 \times 10^{-4} \text{ y}^{-1}$$

Given the biological specific activity of carbon as 15.3 dpm/g, the age of a sample, t , can be calculated after the determination of its specific activity, SA , by:

$$t = \frac{1}{\lambda} \ln \frac{15.3}{SA} \text{ (y)}$$

SECTION VI

Problems

1. One millimole of a compound with the empirical formula MCl_3 is dissolved in an aqueous solution containing 1 milliequivalent of radioactive Cl^- . At intervals, samples of the compound are isolated from aliquots of the solution. The activity of the compound increases slowly until it reaches $6/7$ of the total activity. What is the simplest molecular formula consistent with these data? If the original specific activity of the Cl^- was 1000 cpm per mg of $AgCl$, what will be its final specific activity?
2. A 2.0 ml sample of an aqueous solution containing 0.1 microcurie per ml of tritium is injected into the blood stream of an animal. After allowing sufficient time for complete circulatory mixing, a 1.0 ml aliquot of blood is removed and found to have an activity of 1480 dpm of tritium. From this, calculate the blood volume of the animal.
3. A Grignard reaction was run beginning with 1.55 gm of bromobenzene. After completion of reaction, 2.5×10^4 dpm of ^{14}C labeled benzoic acid was added to the reaction mixture. The benzoic acid was extracted once with petroleum ether and the extract was found to have 55 mg of benzoic acid with a specific activity of 190 dpm per mg. What was

the acid yield in the Grignard reaction?

4. A 0.1 gm sample of animal tissue was taken to be analyzed for histamine. After extraction of the histamine, it is reacted with ^{131}I labeled pipsyl chloride reagent with a specific activity of 2×10^6 cpm per micromole. After removal of unreacted reagent, 3000 micromoles of unlabeled pipsylhistamine are added to the mixture. After purification by recrystallization a 3000 micromole (33 mg) sample was found to contain 400 cpm. What was the amount of histamine in the sample?
5. A reaction mixture containing butyl alcohol was reacted with excess ^{14}C labeled acetic anhydride which had an activity of 5000 cpm per mg. The butyl acetate was isolated and found to have a total activity of 3.4×10^4 cpm. Calculate the amount of butyl alcohol in the reaction mixture.
6. A 1 ml sample of a solution containing trace amounts of lead and bismuth was subjected to paper chromatography in alcoholic hydrochloric acid solution. After development of the chromatogram, the wet strip was exposed to ^{35}S - labeled hydrogen sulfide gas whose activity was 2×10^5 cpm per microgram of H_2S . Upon drying and counting the strip, a spot corresponding to the position of PbS was found to have an activity of 1900 cpm while

a second at the correct position for Bi_2S_3 was found to have 475 cpm. What are the concentrations of lead and bismuth per liter in the solution?

7. A 10 ml sample of an unknown $\text{Ba}(\text{II})$ solution was titrated by addition of a 0.010 M sulfate solution containing 100,000 cpm per ml; 100 microliter additions were made and a filtered aliquot of the supernatant solution counted after each addition. No significant counts were observed until the 6th addition when 235 cpm were obtained; subsequent 100 μl additions gave the following count rates in the solution aliquot; 1170, 2080, 2980, and 3860 cpm. What was the concentration of the barium solution?

Part XI
Liquid Scintillation Counting (LSC)
Outline

- I. Principles of LSC
 - A. General Aspects of LSC
 - B. Factors Influencing Resolution
 - C. Liquid Scintillation Cocktails
 - D. Counting Vials
 - E. Sample Preparation
 - F. Chemiluminescence
 - G. Quenching
 - 1. Chemical Quenching
 - 2. Color Quenching
 - 3. Quench Corrections
 - a. Internal Standard Method
 - b. External Standard Method
 - c. Channels Ratio Method
 - H. Dual Labelling
- II. LSC for Alpha Emitting Nuclides
- III. Cerenkov Radiation and Counting
- IV. Problems
- V. References

SECTION I

Principles of LSC

A. General Aspects of Liquid Scintillation Counting (LSC)

Radionuclide is placed in solution or in a colloidal suspension in intimate contact with scintillation material

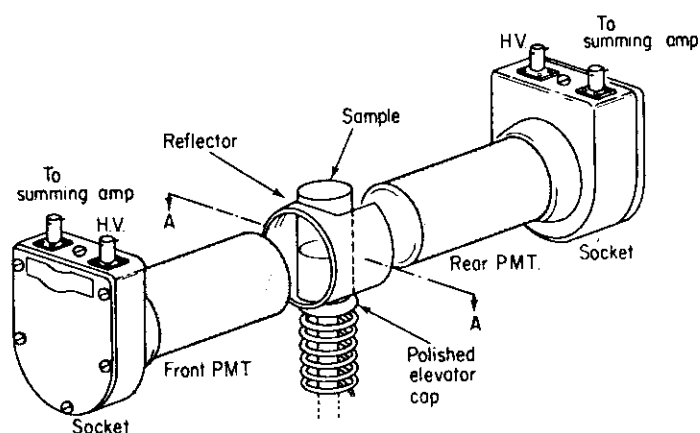


FIG. 1. Diagram of a typical scintillation counter.

Scintillation cocktail - solution of solvent + scintillator solutes

Advantages of LSC:

1. No self-absorption, thus high counting efficiency
2. LSC used to count weak β^- (e.g. ^{14}C , ^3H) but also higher energy β^- and EC. nuclides as well as α emitters
3. Possible to count two isotopes at the same time such as ^{14}C and ^3H - dual labelling

4. Easily automated analysis; often preset time or preset count option
5. Technology of sample preparation and counting highly developed

B. Factors Influencing Resolution in LSC

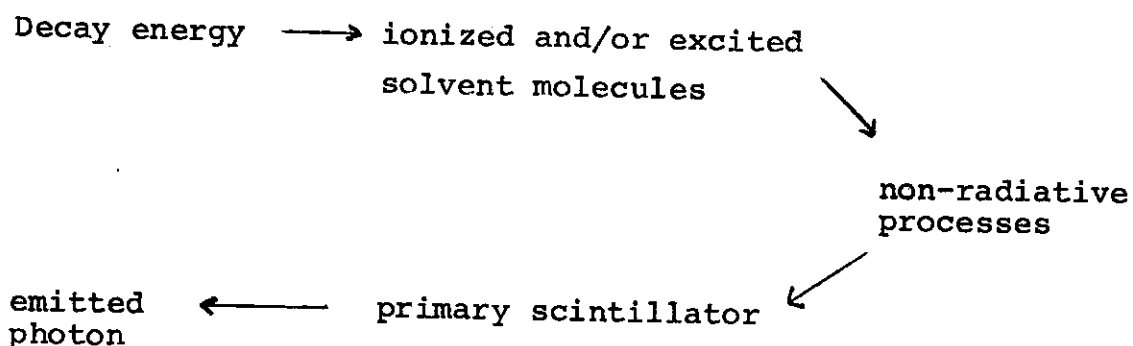
1. Energy of radiation: high detection threshold requires the background to be minimized for signal detection of low energy β
2. Type of radiation: α emitting nuclides are only 10% as efficient as β nuclides in converting the energy into photons
3. Instrumental: geometrical arrangement of detectors, photomultiplier tubes and associated electronic circuitry and the temperature of the scintillator are important
4. Scintillation cocktail: the composition of the basic scintillation solution and distribution of sample in the cocktail affect efficiency and resolution
5. Quenching: results in a decrease in the counting efficiency, and a shift in the pulse height spectrum

C. Liquid Scintillation Cocktail

1. Processes in LSC:

β (or α) radiation excites solvent molecules;
excitation transferred by collision to other molecules

and eventually to scintillator. This emits photons which have energy distribution related to fluorescence spectrum of scintillator.



2. Primary scintillator (solute) dissolved in appropriate solvent.
 - a. requirements - high degree of solubility of primary solute in solvent and considerable overlap of emission spectrum of solvent and the absorption spectrum of solute.
3. Secondary scintillator - added to cocktail as a wavelength shifter. Absorption spectrum of secondary scintillator must overlap with emission spectrum of primary scintillator.
4. Solvent:
 - 1) must efficiently transfer energy to scintillator
 - 2) must dissolve scintillator and sample in sufficient concentration

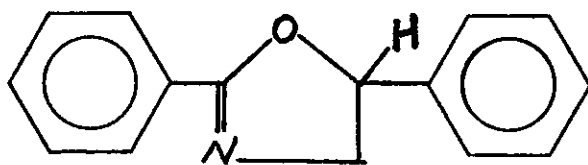
- 3) must not freeze at operating temperature
- 4) cost and purity also important

Common solvents usually aromatic hydrocarbons due to their higher efficiency for 1 .

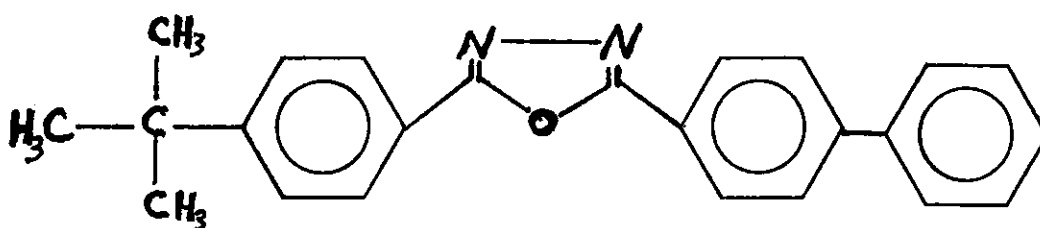
- a. Xylene - greatest pulse-height for a particular solute
- b. Toluene - cheaper, smaller absorption of fluorescent light thus recommended for larger volumes
- c. For aqueous samples - dioxane added to increase solubility
- d. For aqueous samples - quenching is a problem, naphthalene can be added to solvent to damp quenching

5. Some primary scintillators:

- a. PPO: 2-5-diphenyloxazole - very commonly used.
4-6 g/l for optimum results
- b. butyl - PBD: [2-(4-t-butylphenyl)-5-(4-biphenyl)-1,3,4, oxadiazole]. Excellent fluorescence properties and higher counting efficiency than PPO.
Not used in biological systems due to color quenching.



(a) PPO

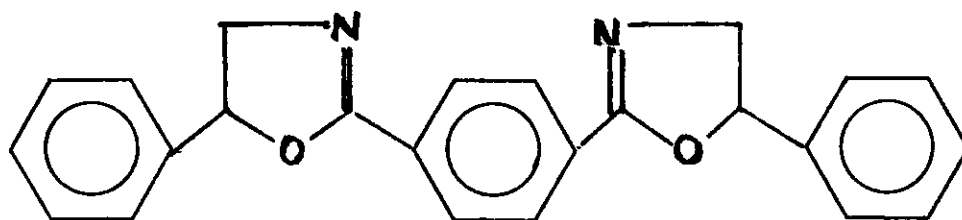


(b) butyl-PBD

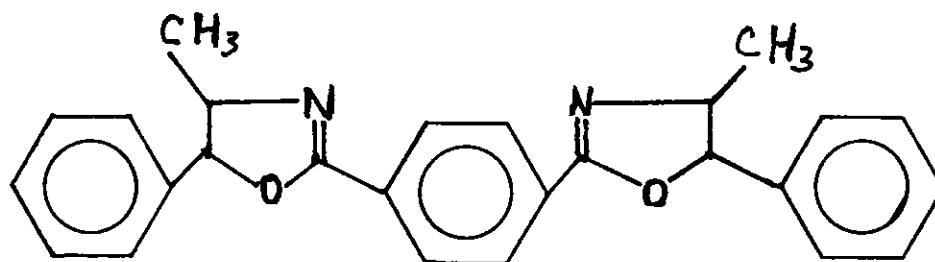
FIG. 2

6. Some secondary scintillators

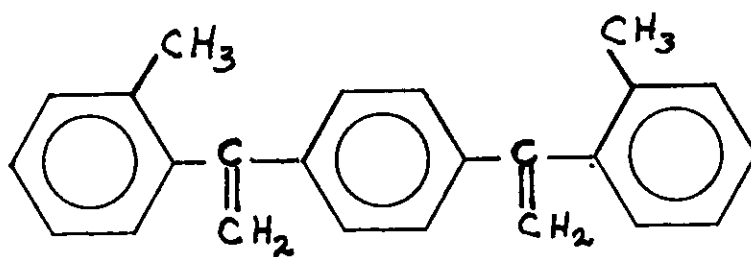
- a. POPOP: 1,4-bis-2(5-phenyloxaxolyl)-benzene
most common, usually used with PPO, efficient but poor solubility. 0.05-0.8 g/l concentration range.
- b. Dimethyl-POPOP: 1,4-bis-2(4-methyl-5-phenyloxaxolyl)-benzene 0.25-1.2 g/l concentration range.
- c. bis-MSB: p-bis-(O-methylstyryl)-benzene more soluble than POPOP or dimethyl POPOP.



(a) POPOP



(b) Dimethyl - POPOP



(c) bis-MSB

FIG. 3

TABLE 1

NAME	MOL. WT.	CONC. (M)	Toluene	
			λ_1 (Å)	λ_2 (Å)
PPO	221.25	10^{-2}	3650	3800
butyl-PBD	354.47	10^{-2}	3670	3820
bis-MSB	310.44	10^{-3}	4230	
POPOP	364.39	10^{-3}	4200	4410
DM-POPOP	392.44	10^{-3}	4300	

G. Counting Vials - usually 20 ml capacity

1. Glass-high background due to ^{40}K
2. Quartz-lower background but more expensive
3. Plastic-usually polyethylene, inexpensive; cannot be used for sample storage
4. Nylon-much like polyethylene but not permeable to solvents.

H. Sample Preparation

1. Biological Systems

- a. Dissolve directly in aromatic solvents
- b. Solubilization using quarternary ammonium bases-
avoid color quenching
- c. Wet combustion technique
60% HClO_4 ; 30% H_2O_2
- d. The alkalized emulsion cocktail

1. Insta-Gel (Packard Co.)

QUENCH RESISTANCE

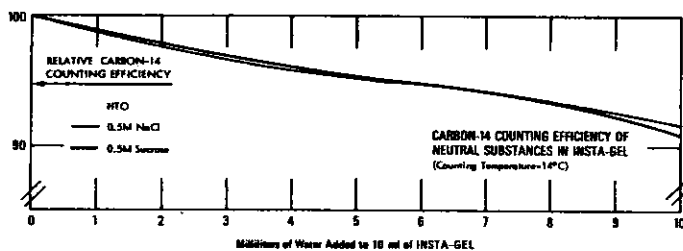


FIG. 4.

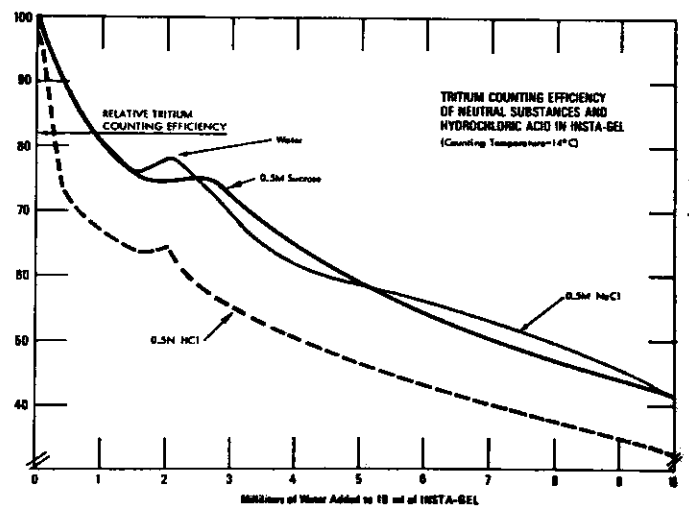


FIG. 5.

e. Milipore filters

1. Filters are either dissolved or suspended in gel.

F. Chemiluminescence

In LSC, arises from spurious luminescence events which results in an increase in the backcount.

Procedures for minimizing chemiluminescence:

Avoid preparation of samples under fluorescent lighting or direct sunlight.

Be sure digested sample is cooled before adding scintillation cocktail.

Add the maximum amount of water that will stay in solution when the sample is diluted with scintillation solution.

Neutralize digested sample to a pH of 6-7 with glacial acetic acid.

Dark adapt samples for 24 hours at 0°C before counting.

Adjust the liquid scintillation counter to operate at lowest possible temperature.

Increase lower window (discriminator) on the liquid scintillation counter to eliminate the spurious low-energy emissions. (This will not apply to tritium samples, but works nicely for high energy beta emitters such as ^{32}P .)

As a last resort: add small increments of methanol until acceptable background levels are obtained.

Note: Some of the above techniques, while reducing chemiluminescence, also reduce counting efficiency. Should experiment to ascertain conditions for the highest counting efficiency and lowest background.

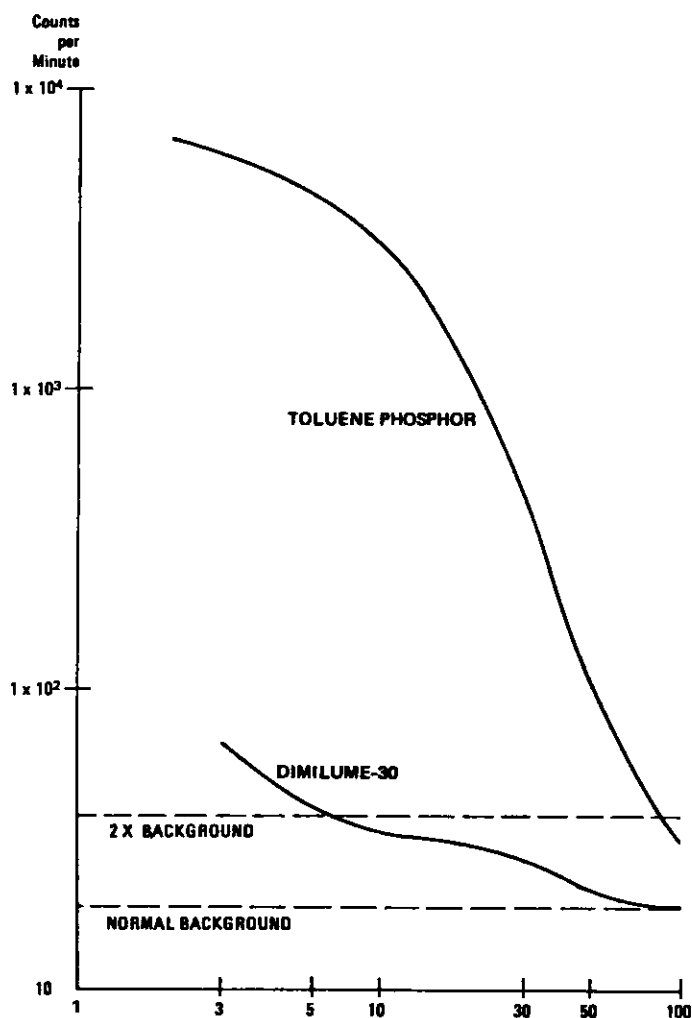


FIG. 6. TIME (MINUTES)
Reduction of Chemiluminescence at 24°C.
Sample: 0.1 ml blood plasma, 1ml SOLUENE-100,
and 10 ml phosphor.

G. Quenching - most probable source of error in LSC

1. Chemical Quenching - decrease in counting efficiency due to removal of excitation energy from scintillator via collisions before scintillator undergoes radiative de-excitation. Also shifts pulse height spectrum.

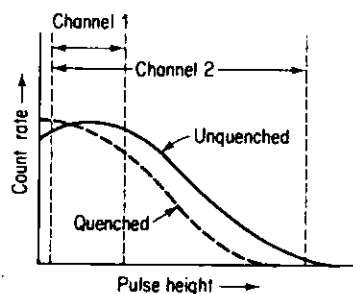


FIG. 7. Typical pulse height spectra for quenched and unquenched samples

- a. Effective chemical quenchers - molecules containing very electronegative atoms such as oxygen (air, water, ketones, aldehydes) or halides (organic halides)

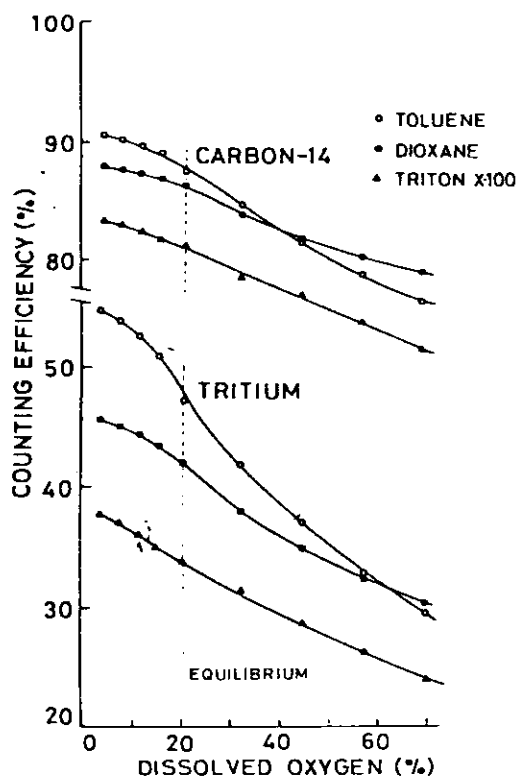


FIG. 8. Variation of counting efficiencies for T and C-14 vs. dissolved oxygen

2. Color Quenching - compounds which absorb photons from scintillator. Usually colored impurities can be removed from sample

3. Quench Corrections

a. Internal Standard Method

Unknown sample is counted

A known quantity of a standard is added, usually 5-10 nci (nanocuries, 10^{-9} curies), of the same nuclide as in the unknown.

Determine counter background by counting a vial containing only the scintillation cocktail

$$\text{Sample Activity (nci)} = \frac{A_s (R_1 - R_b)}{(R_2 - R_1 - R_b)} \quad (1)$$

R_b - background rate

R_1 - count rate of unknown sample

R_2 - count rate of sample + standard

A_s - activity of added standard

$$\text{Detector Efficiency (\%)} = \frac{(R_2 - R_1) \times 100}{A_s} \quad (2)$$

Disadvantages:

Original sample is altered by addition of the standard.

Not practical for large number of samples.

b. External Standard Method

External source used to introduce photoelectrons which simulate internal standards. Must prepare a set of quenched sources of known activity to calibrate system

Determine detector efficiency =

$$\frac{\text{observed count rate}}{\text{real disintegration rate}} \quad (3)$$

for each source

Count calibration sources with and without external source. Difference in count rates called gamma-increment. Calibration curve obtained from plot of detector efficiency vs. gamma increment

Advantage:

Many samples can be handled in a short time

Original sample not altered

Completely instrumental

Disadvantages:

Gamma increment linearly dependent on sample volume

Must be able to prepare samples reproducibly

Often difficult due to volatility of organic solvent of scintillation cocktail

c. Channels Ratio Method

Quenching not only reduces the counting rate but also shifts the pulse height spectrum. This method uses the shift as a measure of quenching and as a means to determine the counter efficiency

Requires at least two independent "windows"

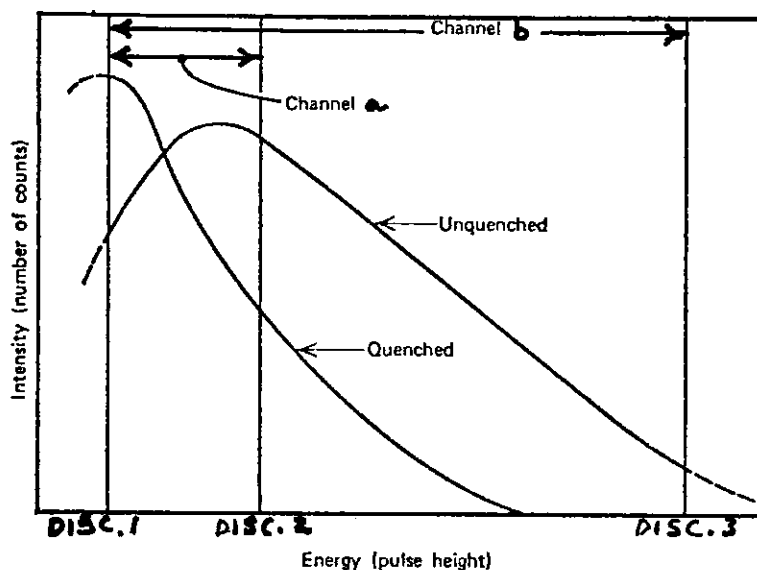


FIG. 9. Pulse height spectrum for an unquenched and quenched sample.

Window A: counts/unit time between discriminator settings 1 and 2.

Window B: counts/unit time between discriminator settings 1 and 3.

Calibrate system by preparing a set of quenched sources.

Determine detector efficiency for each calibration source.

Calibration curve obtained from plot of detector efficiency vs. ratio of counting rates in Windows A and B.

Advantage:

Purely instrumental

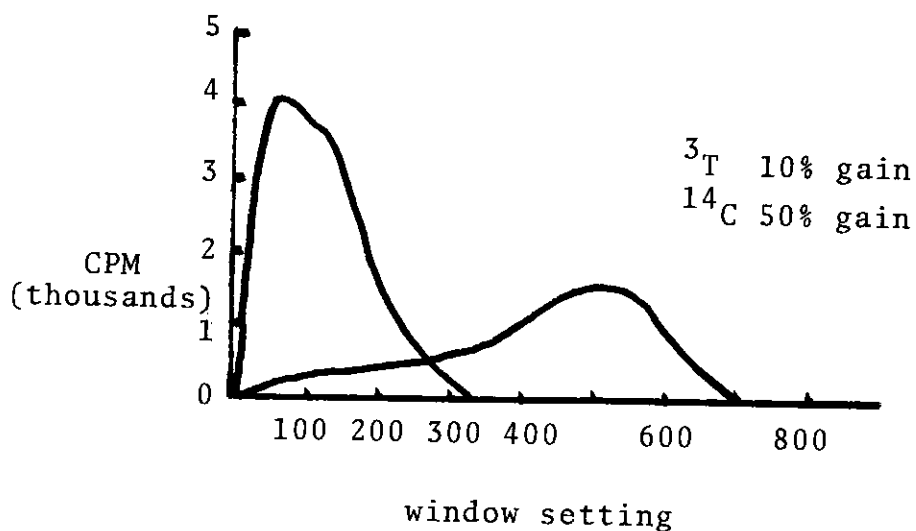
Disadvantage:

When count rate of sample is low counting statistics of monitor window must be considered.

H. Dual Labelling

Requires at least two independent windows.

Simultaneous determination of two isotopes in the same sample; e.g., ^{14}C and ^3H . Need a known standard for each nuclide. Obtain a pulse height spectrum of each standard as a function of amplifier gain. Choose appropriate gain settings in order to maximize the separation of the spectra.



FIG, 10. Pulse height of ^{14}C at 50% gain and ^3H at 10% gain.

Set the ^3H window discriminators at 1 and 2.

Set the ^{14}C window discriminators at 2 and 3.

Determine the count rate of the ^3H source in both window settings.

Do the same for the ^{14}C source.

Calculate the % of ^3H standard appearing in the ^{14}C window and the % ^{14}C standard in the ^3H window.

Calculate detection efficiency in cpm/nci for each standard in their respective windows.

When a mixture of these two isotopes are counted:

(wd = window)

$$\text{dpm}({}^3\text{H}) =$$

$$\frac{\text{cpm in } {}^3\text{H wd} - \frac{[(\% {}^{14}\text{C in } {}^3\text{H wd})(\text{cpm in } {}^{14}\text{C wd})]}{100}}{\text{detector efficiency of } {}^3\text{H}} \quad (4)$$

$$\text{dpm}({}^{14}\text{C}) =$$

$$\frac{\text{cpm in } {}^{14}\text{C wd} - \frac{[(\% {}^3\text{H in } {}^{14}\text{C wd})(\text{cpm in } {}^3\text{H wd})]}{100}}{\text{detector efficiency of } {}^{14}\text{C}} \quad (5)$$

SECTION II

II. LSC for Alpha Emitting Nuclides

Use of solvent extraction - radioactivity extracted into scintillation cocktail containing extracting agent.

SECTION III

Cerenkov Radiation and Counting

Cerenkov radiation is electromagnetic radiation produced when a particle is moving faster than the speed of light in the same medium. If the energy of the particle is sufficiently high, no primary scintillation may be needed due to the Cerenkov radiation produced in the solvent. The refractive index of the solvent determines the threshold energy for the emission of Cerenkov radiation. Therefore, lower energy particles may in fact be counted in a high refractive index solvent alone.

SECTION IV

Problems

1. An unknown sample registers 5050 cpm using LSC. A known standard of 8140 dpm is added to the sample and upon re-counting, the sample registers 11240 cpm. The background is already subtracted.
 - a) What is the counting efficiency?
 - b) What is the activity (in dpm) of the unknown?
2. Six aliquots of tritiated water were counted with LSC and the times in minutes required to register 10,000 counts were: 18.2; 17.8; 18.0; 13.1; 18.5 and 18.7. The background gave 1950 counts in 15 min. Calculate the average count rate and its standard deviation.
3. In a dual labeling experiment the efficiencies were determined to be: window 1, ^3H 40%, ^{14}C 40%; window 2, ^3H 3%, ^{14}C 60%. An unknown labeled with ^3H and ^{14}C gave 9000 cpm in window 1 and 5000 cpm in window 2. What was the activity of ^3H and of ^{13}C in the unknown?
4. A Grignard synthesis yielded impure 10g of benzoic acid. To a 100 mg sample, 5 mg of pure benzoic acid was added. The pure acid had a count by LSC of 12,000 cpm per mg. The spiked sample was purified and 8 mg of pure benzoic acid recovered with a count rate of 38400 cpm. What was the amount of benzoic acid in the impure product?

APPENDIX I TABLE OF CONSTANTS

Velocity of light	$c = 2.99776 \times 10^{10} \text{ cm sec}^{-1}$
Faraday constant	$F = 96500 \text{ abs coul per g equiv.}$
Electronic charge	$e = 4.8025 \times 10^{-10} \text{ abs esu}$ $= 1.60203 \times 10^{-19} \text{ abs coulombs}$
Planck constant	$h = 6.624 \times 10^{-27} \text{ erg sec}$
Avogadro's number	$N = 6.0228 \times 10^{23} \text{ mole}^{-1}$
Mass of electron	$m = 9.1066 \times 10^{-28} \text{ gm}$
Atomic weight of electron	$m = 5.4862 \times 10^{-4} \text{ (physical scale)}$
Mass of unit atomic weight	$M_O = 1.66035 \times 10^{-24} \text{ gm}$
Nuclear radius	$R = 1.4 \times 10^{-13} A^{1/3} \text{ (A=mass number)}$

ATOMIC WEIGHTS:

Hydrogen	$M_H = 1.00814$
Helium	$M_{He} = 4.00387$
Neutron	$M_n = 1.00899$

ENERGY EQUIVALENCE

1 atomic mass unit	$= 931 \text{ MeV}$ $= 1.49 \times 10^{-3} \text{ erg}$ $= 3.56 \times 10^{-11} \text{ cal}$
1 electron mass	$= 0.510 \text{ MeV}$
1 MeV	$= 1.07 \times 10^{-3} \text{ amu}$ $= 1.60 \times 10^{-6} \text{ erg}$ $= 3.82 \times 10^{-14} \text{ cal}$
1 eV/molecule	$= 23.06 \text{ kcal/mole}$

TIME:

Number seconds in a day	8.64×10^4
Number seconds in a year	3.1536×10^7
Number minutes in a day	1440
Number minutes in a year	5.2596×10^5
Number hours in a year	8.766×10^3

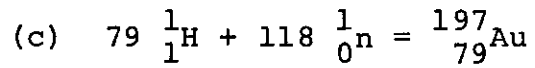
APPENDIX II ANSWERS

Part I

1. (a) ${}_{30}^{64}\text{Zn}$ (d) ${}_{28}^{61}\text{Ni}$
 (b) ${}_{1}^1\text{H}$ (e) ${}_{1}^3\text{H}$
 (c) $2 {}_{0}^1\text{n}$ (f) γ ray

2. (a) $\Delta m = + 0.01140$ amu
 $Q = - 10.61$ MeV
 (b) $\Delta m = + 0.025$ amu
 $Q = - 23.28$ MeV
 (c) $\Delta m = -0.0083$ amu
 $Q = + 7.73$ MeV
 (d) $\Delta m = - 0.0002$
 $Q = + 186$ keV

3. (a) $12 {}_{1}^1\text{H} + 12 {}_{0}^1\text{n} = {}_{12}^{24}\text{Mg}$
 $\Delta m = -0.21300$ amu
 $BE = + 198.30$ MeV
 $BE/A = 8.26$ MeV
 (b) $27 {}_{1}^1\text{H} + 33 {}_{0}^1\text{n} = {}_{27}^{60}\text{Mg}$
 $\Delta m = -0.56390$ amu
 $BE = + 524.99$ MeV
 $BE/A = 8.75$ MeV



$$m = -1.67630 \text{ amu}$$

$$BE = 1560.64 \text{ MeV}$$

$$BE/A = 7.92 \text{ MeV}$$

4.	${}^4_2\text{He}$	2.22 fm	${}^{165}_{67}\text{Ho}$	7.68 fm
	${}^{16}_8\text{O}$	3.52 fm	${}^{206}_{82}\text{Pb}$	8.27 fm
	${}^{56}_{26}\text{Fe}$	5.36 fm	${}^{238}_{92}\text{U}$	8.68 fm
	${}^{75}_{33}\text{As}$	5.90 fm	${}^{256}_{100}\text{Fm}$	8.89 fm
	${}^{112}_{48}\text{Cd}$	6.75 fm		

5. The radius doubles as the mass triples; i.e., there is very little change in size.

6. (a) $Z_{\text{U}} = 92$

$$Z_{\text{H}} = 1$$

$$E_{\text{B}} = 13.12 \text{ MeV}$$

$$R_{\text{U}} = 8.68 \times 10^{-13} \text{ cm}$$

$$R_{\text{H}} = 1.40 \times 10^{-13} \text{ cm}$$

(b) $Z_{\text{Fe}} = 26$

$$Z_{\text{H}} = 1$$

$$E_{\text{B}} = 5.53 \text{ MeV}$$

$$R_{\text{Fe}} = 5.36 \times 10^{-13} \text{ cm}$$

$$R_{\text{H}} = 1.40 \times 10^{-13} \text{ cm}$$

7. Radius of ${}^{27}\text{Al} = 4.20 \times 10^{-13} \text{ cm}$

$$\text{Volume of } {}^{27}\text{Al nucleus} = \frac{4}{3} \pi r^3 = 3.10 \times 10^{-37} \text{ Al atoms cm}^3$$

$$\text{Density} = 1.44 \times 10^{+14} \text{ g/cm}^3$$

Part II

1. (a) $E(\beta^-) = -5.03 \text{ MeV}$ $E(\beta^+) = 1.77 \text{ MeV}$
 (b) $E(\beta^-) = 1.68 \text{ MeV}$ $E(\beta^+) = -1.11 \text{ MeV}$
 (c) $E(\beta^-) = 559 \text{ KeV}$ $E(\beta^+) = -3.44 \text{ MeV}$
 (d) $E(\beta^-) = 6.61 \text{ MeV}$ $E(\beta^+) = 3.54 \text{ MeV}$ $E(\alpha) = 10.43 \text{ MeV}$
 (e) $E(\beta^-) = 1.10 \text{ MeV}$ $E(\beta^+) = 553 \text{ Kev}$ $E(\alpha) = 8.00 \text{ MeV}$

2. $M(^{90}_{39}\text{Y}) = 89.9017 \text{ amu}$

3. $^{115}_{49}\text{In} \xrightarrow{\beta^-} ^{115}_{50}\text{Sn}$ $E(\beta^-) 599 \text{ keV spontaneous}$
 $^{115}_{50}\text{Sn} \xrightarrow{\beta^+} ^{115}_{49}\text{In}$ $E(\beta^+) = -599 \text{ keV}$

4. $M(^{235}_{92}\text{U}) = 235.0439 \text{ amu}$

5.

Isotope	(min ⁻¹)	N(atoms)	mass (g)
(a) $^{36}_{17}\text{Cl}$	4.39×10^{-12}	5.05×10^{20}	3.02×10^{-2}
(b) $^{76}_{33}\text{As}$	4.18×10^{-4}	5.30×10^{12}	6.70×10^{-10}
(c) $^{51}_{24}\text{Cr}$	1.73×10^{-5}	1.28×10^{14}	1.09×10^{-8}

6. $A_t = 1.96 \times 10^4 \text{ cpm}$

7. $t_{1/2} \text{ parent} = 23.5 \text{ m}; t_{1/2} \text{ daughter} = 3.4 \times 10^3 \text{ m}$
 (none of the daughter decays before all of the parent has

decayed)

$$6.9 \times 10^3 \text{ dpm}$$

8. From Graph: component (B) $t_{1/2} = 12 \text{ hr}$
 component (A) $t_{1/2} = 3 \text{ hr}$
9. Weight of ${}^{234}_{90}\text{Th} = 1.44 \times 10^{-8} \text{ g}$
10. Weight of ${}^{144}\text{Pr/g } {}^{144}\text{Ce} = 4.21 \times 10^{-4} \text{ g}$

Part III

1. 2.3×10^5 ion pairs
2. 68 keV
3. Al: 8.2 mg/cm^2
 air: 4.9 cm (Compare with Curve II, figure 11, Section III
 (A))
4. Since the same dependence on velocity exists, i.e., $(\frac{dE}{dX}) \propto \frac{1}{v^2}$, the specific ionization per unit path length will be much less for electrons than for heavy particles.
5. The ratio of energy lost by radiation and ionization is ~ 1 . This implies that a 10 MeV electron will lose energy at approximately equal rates by both processes.

6. From Fig. 9, Pb: mostly pair production with some Compton effect

Fig. 10, Al: Compton effect only

7. 50 mr (α) = 1 rem
 100 mr (β) = 0.1 rem
 150 mr (γ) = 0.1 rem
8. 25 years - 35 rems
 45 years - 135 rems
 60 years - 210 rems

Part IV

1. 1.6×10^5 ion pairs
2. Because of the smaller mass a β^- particle may lose a large fraction of its energy in one collision. When this happens the primary electron will be deflected through a larger angle and have considerably lower energy. They in turn will cause secondary ion pair formation.
- 3.
- | N | Eff (%) | The rate of energy loss ($^{-dE/dx}$) |
|---|---------|--|
| 1 | 63 | for such particles is in the order |
| 2 | 86 | $\alpha > \beta > \gamma$. Therefore, the number of |
| 3 | 95 | primary ion pairs formed is least |
| 4 | 98 | for γ 's. G.M. counting should be |
| 5 | 99 | 100% efficient for α 's, and β 's |

As $N \geq 6$, $\text{Eff}(\%) \rightarrow 100\%$ since $N \gg 6$ but $< 100\%$ efficient for γ 's since N can be very small.

4. $\Delta V = 0.94V$

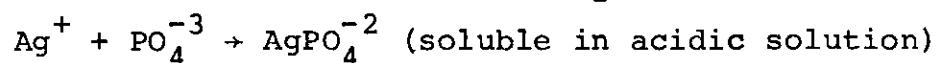
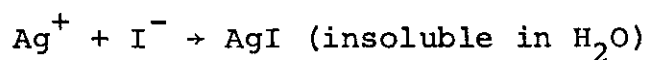
5. 2.73×10^{-14} coulombs

6. $R = 5.8 \text{ mg/cm}^2$

7. (a) 2.22×10^3 dpm. Count β^- using G.M. or proportional counter.
 (b) 2.22×10^5 dpm. Count rate too high for G.M. Also weak β^- probably wouldn't penetrate G.M. window (see Fig. 10)
 Use windowless proportional counter.
 (c) 22.2 dpm. Very low count rate. Can use ionization chamber on windowless proportional counter. Possible thin window proportional counter could be used.
 (d) 22.2×10^3 dpm. Proportional counter for β^- or count γ 's by NaI(Tl) scintillation counter.

Part V

1. By precipitation via



2. (a) No, one would need 40M sulfate ion
(b) 8.2×10^{-10}
3. solvent 1 - 4.9×10^{-4} g/ml
solvent 2 - 2.4×10^{-5} g/ml
4. solvent 1 - 43 cpm
solvent 2 - 1707 cpm
5. The vapor pressures of the two substances are considerably different and that of one is large enough to be useful.
6. Ion exchange chromatography using Dowex-50 and concentrated HCl.
7. $\text{Zn} \rightarrow \text{Zn}^{+2} + 2\text{e}^{-}$
 $\text{Fe} \rightarrow \text{Fe}^{+2} + 2\text{e}^{-}$ (cathode)
 $\text{Sn} \rightarrow \text{Sn}^{+2} + 2\text{e}^{-}$
 $\text{H}_2\text{O} \rightarrow 1/2 \text{O}_2 + 2\text{H}^{+} + 2\text{e}^{-}$ (anode)

The voltage which must be applied to cause deposition

	$E_{\text{app}}(\text{V})$
Zn	+ 2.19
Fe	+ 1.87
Sn	+ 1.57

Part VI

1. $\sigma = 387$; $149,613 \rightarrow 150,387$ counts
2. $R_s = 560 \pm 6$ cpm
Ignoring the contribution of the background error

$$R_s = 560 \pm 6$$

$$\text{Probable error} = \pm 4$$

$$3. \quad t_s = 13.6 \text{ m} \quad t_b = 16.4 \text{ m}$$

$$\sigma_s = \pm 2 \text{ cpm or } 3\%$$

$$4. \quad 3, 2, 4$$

$$5. \quad t_{GM} < t_{prop}, \text{ therefore G.M. will require less time for the same error}$$

$$6. \quad 3080, \text{ and } 3580 \text{ should be rejected}$$

$$7. \quad \text{shielded} - 4.8 \text{ m; unshielded} - 7.9 \text{ m}$$

$$8. \quad \text{No; yes}$$

$$9. \quad 4.5 \times 10^5, 1.8 \times 10^4, 4.5 \times 10^3, 180, 45, 1.8$$

$$\text{Use. eqn. 11 and assume } R_b \ll R_s, \text{ therefore } (1 + 2 \frac{R_b}{R_s}) \rightarrow 1 \text{ and}$$

$$R_s \rightarrow R_t$$

$$10. \quad (a) \text{ yes} \quad (b) \text{ not operating normally}$$

Part VII

$$1. \quad E_e = \frac{4E_\gamma^2}{4E_\gamma + 1}$$

$$(a) \quad 0.79 \text{ MeV for } E_\gamma = 0.986$$

$$1.10 \text{ MeV for } E_\gamma = 1.314$$

$$(b) \quad 0.342 \text{ MeV}$$

$$(c) \quad 0.022 \text{ MeV}$$

$$2. \quad (a) \quad E_\gamma = 1.314, \text{ Compton}$$

- $E_\gamma = 0.986$; Compton, some photoelectric
- (b) Compton with considerable photoelectric
 - (c) Primarily photoelectric, some Compton
3. Pair Production $E_\gamma > 1.5$ MeV
- (a) Full energy peak: due to multiple processes
 - (b) Single escape peak: $E = 1.78 - .51 = 1.27$ MeV
 - (c) Double escape peak: $E = 1.78 - 2(.51) = 0.76$ MeV
 - (d) Annihilation peak: $E = .51$ MeV
4. For constant E_γ as the size of the crystal increases the percent total absorption increases. For a constant crystal size, typically 3x3, the percent total absorption decreases with increasing E_γ .
5. From a graph, % resolution = 7.1%
- energy of x-ray escape peak = $\gamma_1 - E_{\text{x-ray}}$
- $140 \text{ keV} - 100 \text{ keV} = 40 \text{ keV} = E_{\text{x-ray}}$
6. (a) $E_{\gamma 1} = 1.33$ MeV
- (b) $E_{\gamma 2} = 1.17$ MeV
- (c) 1.12 MeV Compton edge for 1.33 MeV γ -ray
- (d) 0.96 MeV Compton edge for 1.17 MeV γ -ray

Part VIII

1. $^{247}\text{Cf} = 43.2$ MeV
2. 66.8 MeV
3. Ca. 236 MeV

4. (a) 3.44 MeV
(b) 17.87 MeV
5. 4.8 y
6. 815 b
7. 3×10^3 s

Part IX

1. (a) $t \gg t_{1/2}$, $e^{-\lambda t} \rightarrow 0$, thus the saturation factor, $(1 - e^{-\lambda t}) \rightarrow 1$; the activity of the product nuclide reaches a maximum.
(b) $t \ll t_{1/2}$, $e^{-\lambda t}$ can be approximated by $(1 - \lambda t)$, therefore, the saturation factor becomes $1 - (1 - \lambda t) = \lambda t$. The activity of the product nuclide increases proportionally with the irradiation time.
2. (a) 8.58×10^6 dpm Use: $A_2 = \frac{X(g) \sigma f \phi}{M}$
(b) 2.68×10^2 dpm
(c) 1.58×10^6 dpm Where M is molecular weight of de-
(d) 1.67×10^6 dpm sired element
3. 3.3×10^9 dpm
4. Irradiation time $2 t_{1/2} \rightarrow 75\%$ saturation = $(1 - e^{-\lambda t})$
Actual disintegration rate = 5.6×10^8 dpm = A_Y
 $A_Y = N_X \sigma \phi (.75)$
 $N_X = 1.3 \times 10^{20}$ atoms
 $^{127}\text{I} = 2.8 \times 10^{-2}$ g originally present
5. $\phi = 9.39 \times 10^{11}$ neutrons/cm²/sec

6. Weight of unknown sample = 5.29×10^{-6} g

Correct for efficiencies of the counters used

7. Isotope	Decay Mode	Mode of Production
$^{137}_{55}\text{Cs}$	β^{-}	reactor
$^{14}_6\text{C}$	β^{-}	reactor
$^{147}_{61}\text{Pm}$	β^{-}	reactor or fission
$^{58}_{27}\text{Co}$	β^{+}	accelerator
$^{35}_{16}\text{S}$	β^{-}	reactor
$^{197}_{80}\text{Hg}$	EC	accelerator
$^{36}_{17}\text{Cl}$	β^{-}	reactor
$^{60}_{21}\text{Co}$	β^{-}	reactor

8. ^{160}Tb : 2.06×10^{11} dpm
 ^{161}Tb : 7.86×10^{10} dpm

9. 17%

10. 8.5 yr.

Part X

1. M_2Cl_6 143 cpm
2. 295 ml
3. 132 mg
4. 2.2×10^{-4} mg
5. 5 mg

6. Pb = 0.058 mg/l Bi = 0.01 mg/l

7. $5.73 \times 10^{-4} \text{M}$

Part XI

1. (a) 76%

(b) 3660 dpm

2. 454 ± 7 cpm

3. ^{14}C : 7590 cpm

^3H : 14910 cpm

4. 750 mg.

APPENDIX III

Common Radioisotopes Listed According to Half-life

Half-life ^a	Isotope	Maximum Specific Activity ^b	Radiation	Production Method	
				Reactor	Cyclotron
12.3 h	¹³⁰ I	~40 Ci/g I	β, γ	(n, γ)	
12.36 h	⁴² K	>450 mCi/g K	β, γ	(n, γ)	
12.80 h	⁶⁴ Cu	~25 Ci/g Cu	EC, β^- , β^+ , γ	(n, γ)	
13.47 h	¹⁰⁹ Pd	~6 Ci/g Pd	β, γ	(n, γ)	
14.12 h	⁷² Ga	~2 Ci/g Ga	β, γ	(n, γ)	
14.96 h	²⁴ Na	~10 Ci/g Na	β, γ	(n, γ)	
17.4 h	¹⁹⁴ Ir	>30 Ci/g Ir	β, γ	(n, γ)	
19.2 h	¹⁴² Pr	>10 Ci/g Pr	β, γ	(n, γ)	
23.9 h	¹⁸⁷ W	~10 Ci/g W	β, γ	(n, γ)	
24 h	^{197m} Hg	~500 mCi/g Hg	EC, γ	(n, γ)	
26.4 h	⁷⁶ As	~4 Ci/g As	β, γ	(n, γ)	
35.34 h	⁸² Br	~1 Ci/g Br	β, γ	(n, γ)	
38.7 h	⁷⁷ As	CF	β, γ	(n, γ), β^-	
40.22 h	¹⁴⁰ La	~9 Ci/g La	β, γ	(n, γ)	
46.8 h	¹⁵³ Sm	~40 Ci/g Sm	β, γ	(n, γ)	
53.5 h	¹¹³ Cd	~50 mCi/g Cd	β, γ	(n, γ)	
64.0 h	⁹⁰ Y	CF, >1 Ci/g	β	Fission, β^-	
64.728 h	¹⁹⁸ Au	~60 Ci/g Au	β, γ	(n, γ)	
65 h	¹⁹⁷ Hg	~1 Ci/g Hg	EC, γ	(n, γ)	
66.7 h	⁹⁹ Mo	>140 mCi/g Mo	β, γ	(n, γ)	
67.2 h	¹²² Sb	~2 Ci/g Sb	β, γ	(n, γ)	
3.15 d	¹⁹⁹ Au	CF	β, γ	(n, γ)	
3.70 d	¹⁸⁶ Re	~15 Ci/g Re	EC, β, γ	(n, γ)	
4.535 d	⁴⁷ Ca	>150 mCi/g Ca	β, γ	(n, γ)	
5.013 d	²¹⁰ Bi	~50 mCi/g Bi	β, γ	(n, γ)	
5.270 d	¹³³ Xe	CF	β, γ	Fission	
6.7 d	¹⁷⁷ Lu	~20 Ci/g Lu	β, γ	(n, γ)	
7.5 d	¹¹¹ Ag	CF	β, γ	(n, γ), β^-	
8.05 d	¹³¹ I	CF	β, γ	(n, γ), β^-	
9.3 d	¹⁶⁹ Er	~1 Ci/g Er	β, γ	(n, γ)	
11.06 d	¹⁴⁷ Nd	CF	β, γ	Fission	
12.0 d	¹³¹ Ba	~10 mCi/g Ba	EC, γ	(n, γ)	
12.80 d	¹⁴⁰ Ba	CF	β, γ	Fission	
13.59 d	¹⁴³ Pr	CF	EC, γ	Fission	
14.28 d	³² P	CF	β		³² S(n, p) ³² P
15.0 d	¹⁹¹ Os	>400 mCi/g Os	β, γ	(n, γ)	
16.0 d	⁴⁸ V	CF	β^+ , EC, γ		⁴⁸ Ti(p, n) ⁴⁸ V
18.66 d	⁸⁶ Rb	~1 Ci/g Rb	β, γ	(n, γ)	
27.8 d	⁵¹ Cr	~500 Ci/g Cr	EC, γ	(n, γ)	
32.5 d	¹⁴¹ Ce	~2 Ci/g Ce	β, γ	(n, γ)	
35.0 d	⁹⁵ Nb	CF	β, γ	Fission	
35.1 d	³⁷ Ar	CF	EC		⁴⁰ Ca(n, α) ³⁷ Ar
39.5 d	¹⁰³ Ru	CF	β, γ	Fission	
42.5 d	¹⁸¹ Hf	~2 Ci/g Hf	β, γ	(n, γ)	
43 d	^{115m} Cd	~100 mCi/g Cd	β, γ	(n, γ)	
45.6 d	⁵⁹ Fe	~20 Ci/g Fe	β, γ	(n, γ)	
46.9 d	²⁰³ Hg	~1 Ci/g Hg	β, γ	(n, γ)	
50.0 d	^{114m} In	~1 Ci/g In	γ	(n, γ)	
52.7 d	⁸⁹ Sr	CF	β	Fission	
53 d	⁷ Be	CF	EC		⁷ Li(p, n) ⁷ Be
58.8 d	⁹¹ Y	CF	β, γ	Fission	
60.2 d	¹²⁵ I	CF	EC, γ	(n, γ), EC \rightarrow	
60.4 d	¹²⁴ Sb	~2 Ci/g Sb	β, γ	(n, γ)	
64 d	⁸⁵ Sr	CF	EC, γ		⁸⁵ Rb(p, n) ⁸⁵ Sr

APPENDIX III con't

Half-life ^a	Isotope	Maximum Specific Activity ^b	Radiation	Production Method	
				Reactor	Cyclotron
65.5 d	⁹⁵ Zr	CF	β, γ	Fission	
72 d	⁵⁸ Co	CF	EC, γ		⁵⁸ Ni(p, n) ⁵⁸ Co
75 d	¹⁸³ W	~500 mCi/g W	EC, β, γ	(n, γ)	
83.9 d	⁴⁶ Sc	>5 Ci/g Sc	β, γ	(n, γ)	
87.9 d	³⁵ S	CF	β		³⁵ Cl(n, p) S
106.7 d	⁸⁸ Y	CF	EC, γ		⁸⁸ Sr(p, n) Y
109 d	^{127m} Te	~500 mCi/g Te	β, γ	(n, γ)	
115 d	¹¹³ Sn	~300 mCi/g Sn	EC, γ	(n, γ)	
115.1 d	¹⁸² Ta	>500 mCi/g Ta	β, γ	(n, γ)	
120.4 d	⁷⁵ Se	~500 Ci/g Se	EC, γ	(n, γ)	
134 d	¹⁷⁰ Tm	~600 Ci/g Tm	β, γ	(n, γ)	
140 d	¹³⁹ Ce	CF	EC, γ		¹³⁹ La(p, n) ¹³⁹ Ce
144 d	¹⁵⁹ Dy	~1 Ci/g Dy	EC, γ	(n, γ)	
165 d	⁴⁵ Ca	~10 Ci/g Ca	β	(n, γ)	
183 d	¹⁹⁵ Au	CF	EC, γ		¹⁹⁵ Pt(p, n) ¹⁹⁵ Au
242 d	¹⁵³ Gd	~3 Ci/g Gd	EC, γ	(n, γ)	
245 d	⁶⁵ Zn	~3 Ci/g Zn	EC, β^+, γ	(n, γ)	
		CF			⁶⁵ Cu(p, n) ⁶⁵ Zn
255 d	^{110m} Ag	~1 Ci/g Ag	β, γ	(n, γ)	
270 d	⁵⁷ Co	CF	EC, γ		⁵⁶ Fe(p, γ) ⁵⁷ Co
280 d	⁶⁸ Ge	CF	$\beta^+, \text{EC}, \gamma$		⁶⁹ Ga(p, 2n) ⁶⁹ Ge
284 d	¹⁴⁴ Ce	CF	β, γ	Fission	
303 d	⁵⁴ Mn	CF	EC, γ		⁵⁴ Fe(n, p) ⁵⁴ Mn
330 d	⁴⁹ V	CF	EC		⁴⁹ Ti(p, n) ⁴⁹ V
368 d	¹⁰⁶ Ru	CF	β	Fission	
453 d	¹⁰⁹ Cd	~1 Ci/g Cd, CF	EC, γ	(n, γ)	¹⁰⁹ Ag(p, n) ¹⁰⁹ Cd
700.8 d	¹⁷¹ Tm	CF	β	(n, γ), $\beta \rightarrow$	
2.05 y	¹³⁴ Cs	~25 Ci/g Cs	β, γ	(n, γ)	
2.6 y	⁵⁵ Fe	~12 Ci/g Fe, CF	EC	(n, γ)	⁵⁵ Mn(p, n) ⁵⁵ Fe
2.62 y	¹⁴⁷ Pm	CF	β, γ	Fission	
2.62 y	²² Na	>1 mCi/mg Na, CF	$\beta^+, \text{EC}, \gamma$		²⁴ Mg(d, α) ²² Na
2.71 y	¹²³ Sb	CF	β	(n, γ), $\beta \rightarrow$	
3.81 y	²⁰⁴ Tl	~1 Ci/g Tl	EC, β	(n, γ)	
5.263 y	⁶⁰ Co	~50 Ci/g Co	β, γ	(n, γ)	
7.2 y	¹³³ Ba	~1 Ci/g Ba	EC, γ	(n, γ)	
10.76 y	⁸⁵ Kr	~21 Ci/g Kr	β, γ	Fission	
12.262 y	³ H	CF	β		⁶ Li(n, α) ³ H
12.7 y	¹⁵² Eu	>250 mCi/g Eu	EC, β, γ	(n, γ)	
16 y	¹⁵⁴ Eu	>250 mCi/g Eu	β, γ	(n, γ)	
27.7 y	⁹⁰ Sr	CF	β	Fission	
30.0 y	¹³⁷ Cs	CF	β	Fission	
30.2 y	²⁰⁷ Pb	CF	EC, γ		²⁰⁷ Pb(p, n) ²⁰⁷ Bi
48 y	⁴⁴ Ti	CF	EC, γ		⁴⁵ Sc(p, 2n) ⁴⁴ Ti
87 y	¹⁵¹ Sm	25 Ci/g Sm	β, γ	Fission	
92 y	⁶³ Ni	~10 Ci/g Ni	β	(n, γ)	
1.2 $\times 10^3$ y	^{166m} Ho	~1 mCi/g Ho	β, γ	(n, γ)	
5730 y	¹⁴ C	>2 Ci/g C	β		¹⁴ N(n, p) ¹⁴ C
2 $\times 10^4$ y	⁹⁴ Nb	0.2 mCi/g Nb	β, γ	(n, γ)	
8 $\times 10^4$ y	⁵⁹ Ni	~10 mCi/g Ni	EC	(n, γ)	
2.12 $\times 10^5$ y	⁹⁹ Tc	20 mCi/g Tc	β	Fission	
3.08 $\times 10^5$ y	³⁶ Cl	~10 mCi/g Cl	β	(n, γ)	
7.4 $\times 10^5$ y	²⁶ Al	~1 μ Ci/g Al	β^+, γ		²⁷ Al(p, pn) ²⁶ Al
2.7 $\times 10^6$ y	¹⁰ Be	~15 μ Ci/g Be	β	(n, γ)	
1.7 $\times 10^7$ y	¹²⁹ I	~260 μ Ci/g I	β, γ	Fission	

^ah = hour, d = day, y = year^bCf = carrier-free

APPENDIX IV

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K. Monographs of the Subcommittee on Radiochemistry of
the Committee on Nuclear Science, NAS-NRS.

Copies of the monographs listed below are available from the Clearinghouse for Federal Scientific and Technical Information, National Bureau of Standards, U.S. Department of Commerce, Springfield, Virginia 22151. The minimum price is \$3.00 for either one, two or three randomly selected publications. Additional individual copies will be sold in increments of three for \$3.00.

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Thorium	NAS-NS-3004 (1960)
Fluorine, Chlorine Bromine, Iodine	NAS-NS-3005 (1960)
Americium and Curium	NAS-NS-3006 (1960)
Chromium	NAS-NS-3007 (Rev.) (1963)
Rhodium	NAS-NS-3008 (Rev.) (1965)
Molybdenum	NAS-NS-3009 (1960)
Barium, Calcium, and Strontium	NAS-NS-3010 (1960)
Zirconium and Hafnium	NAS-NS-3011 (1960)
Astatine	NAS-NS-3012 (1960)
Beryllium	NAS-NS-3013 (1960)
Indium	NAS-NS-3014 (1960)
Zinc	NAS-NS-3105 (1960)
Protactinium	NAS-NS-3016 (1960)
Iron	NAS-NS-3017 (1960)
Manganese	NAS-NS-3018 (1960)
Carbon, Nitrogen, and Oxygen	NAS-NS-3019 (1960)
Rare Earths- Scandium, Yttrium, and Actinium	NAS-NS-3020 (1961)
Technetium	NAS-NS-3021 (1960)
Vanadium	NAS-NS-3022 (1960)
Tin	NAS-NS-3023 (1960)
Magnesium	NAS-NS-3024 (1961)
Rare Gases	NAS-NS-3025 (1960)
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Aluminum and Gallium	NAS-NS-3032 (1961)
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Radiochemistry con't.

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Tellurium	NAS-NS-3038 (1960)
Niobium and Tantalum	NAS-NS-3039 (1961)
Lead	NAS-NS-3040 (1961)
Cobalt	NAS-NS-3041 (1961)
Tungsten	NAS-NS-3042 (1961)
Germanium	NAS-NS-3043 (1961)
Platinum	NAS-NS-3044 (1961)
Iridium	NAS-NS-3045 (1961)
Osmium	NAS-NS-3046 (1961)
Silver	NAS-NS-3047 (1961)
Potassium	NAS-NS-3048 (1961)
Silicon	NAS-NS-3049 (Rev.) (1968)
Uranium	NAS-NS-3050 (1961)
Nickel	NAS-NS-3051 (1961)
Palladium	NAS-NS-3052 (1961)
Rubidium	NAS-NS-3053 (1962)
Sulfur	NAS-NS-3054 (1961)
Sodium	NAS-NS-3055 (1962)
Phosphorus	NAS-NS-3056 (1962)
Radium	NAS-NS-3057 (1964)
Plutonium	NAS-NS-3058 (1965)
Recent Proc. for As, At, Be, Mg, Ni, Ru and Se	NAS-NS-3059 (1974)
Neptunium	NAS-NS-3060 (1974)

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