Methods in marine radioactivity and instrumentation

iER
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Objectives

- Present the basics of measurement for alpha, beta and gamma radiation.
- Show good examples of scientific results using these measurements. If we have time.

Basics: Common Types of Radiation

Alphas: An alpha is a particle emitted from the nucleus of an atom, that contains **2 protons and 2 neutrons**. It is identical to the nucleus of a Helium atom, without the electrons.

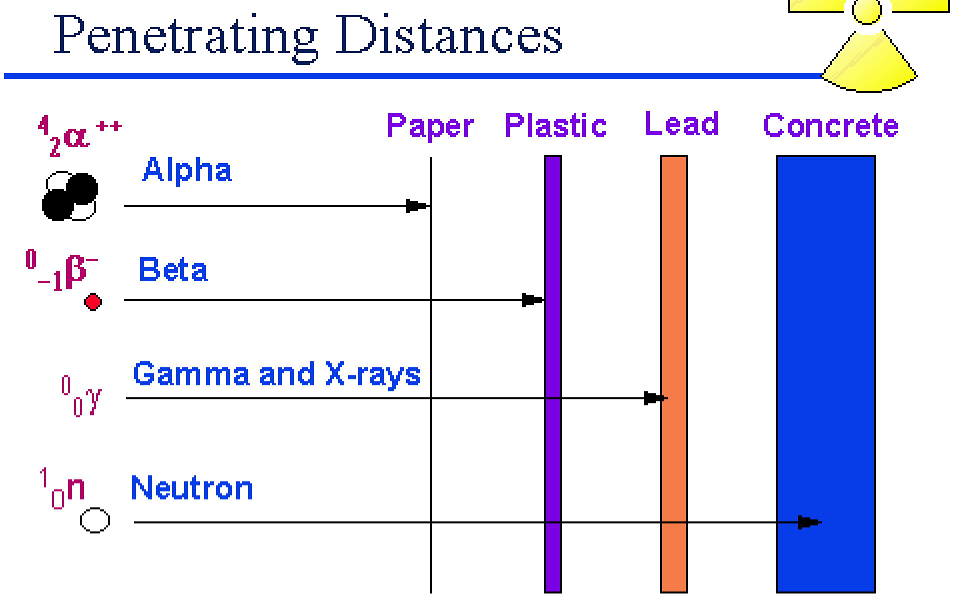
Betas: A beta is a high speed particle, identical to an electron, that is emitted **from the nucleus** of an atom

Gamma Rays: Gamma rays are **electromagnetic waves / photons** emitted **from the nucleus** (center) of an atom.

X rays: X Rays are **electromagnetic waves / photons** emitted **not from the nucleus**, but normally emitted **by energy changes in electrons**. These energy changes are either in electron orbital shells that surround an atom or in the process of slowing down such as in an X-ray machine.

Neutrons: Neutrons are **neutral particles** that are normally contained in the nucleus of all atoms and may be removed **by various interactions or processes like collision and fission**

Basics: Properties of Radiation



Alpha particles are heavy and doubly charged which cause them to lose their energy very quickly in matter. They can be **shielded by** a sheet of paper or the **surface layer of our skin**. Alpha particles are considered **hazardous only to a persons health if an alpha emitting material is ingested or inhaled**.

Beta and positron particles are much smaller and only have one charge, which cause them to interact more slowly with material. They are effectively shielded by thin layers of metal or plastic and are again considered **hazardous only if a beta emitter is ingested or inhaled.**

Gamma emitters are associated with alpha, beta, and positron decay. X-Rays are produced either when electrons change orbits within an atom, or electrons from an external source are deflected around the nucleus of an atom. Both are forms of high energy electromagnetic radiation which **interact lightly with matter**. X-rays and gamma rays are best **shielded by** thick layers of lead or other **dense material** and are **hazardous to people when they are external to the body.**

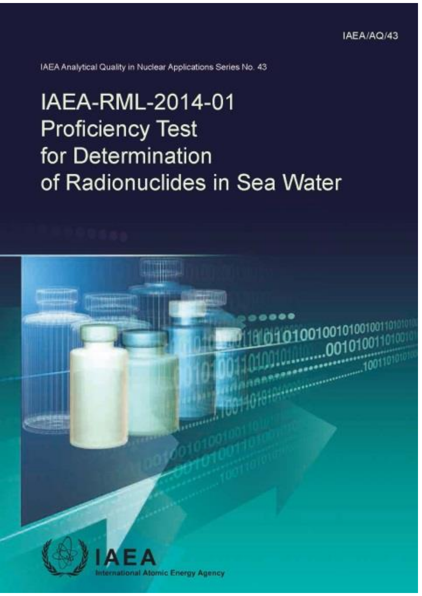
Neutrons are neutral particles with approximately the same mass as a proton. Because they are neutral they **react only weakly with material**. They are an external hazard best **shielded by** thick layers of **concrete**.

Eugen Kvasnak, Radiation and radioactivity, http://www.old.lf3.cuni.cz

Detectors

- Gas filled detectors
- Scintillation detectors
- Semiconductor detectors

List of method and instrument of measurements of radionuclides in seawater (IAEA PT 2014/AQ/43)


IAEA/AQ/43

IAEA Analytical Quality in Nuclear Applications Series No. 43

IAEA-RML-2014-01
Proficiency Test
for Determination
of Radionuclides in Sea Water

IAEA
International Atomic Energy Agency

IAEA-RML-2014-01
PROFICIENCY TEST FOR DETERMINATION OF RADIONUCLIDES IN SEA WATER:
IAEA/AQ/43

Radionuclide	**Methods**	**Instrument**
^{3}H	distilation of the seawater sample, electrolytic enrichment after distillation	liquid scitilation countiung (LSC)
^{90}Sr	chemically separeted ^{90}Y	gas-flow propotional counting (GPC)
^{134}Cs and ^{137}Cs	direct or pre-concentration (AMP/Cs compound, adsorption on hexacyanoferrates containing either copper, nickel or cobalt)	gamma spectrometry

List of half life, radiation, instrument and method of radionuclides in seawater

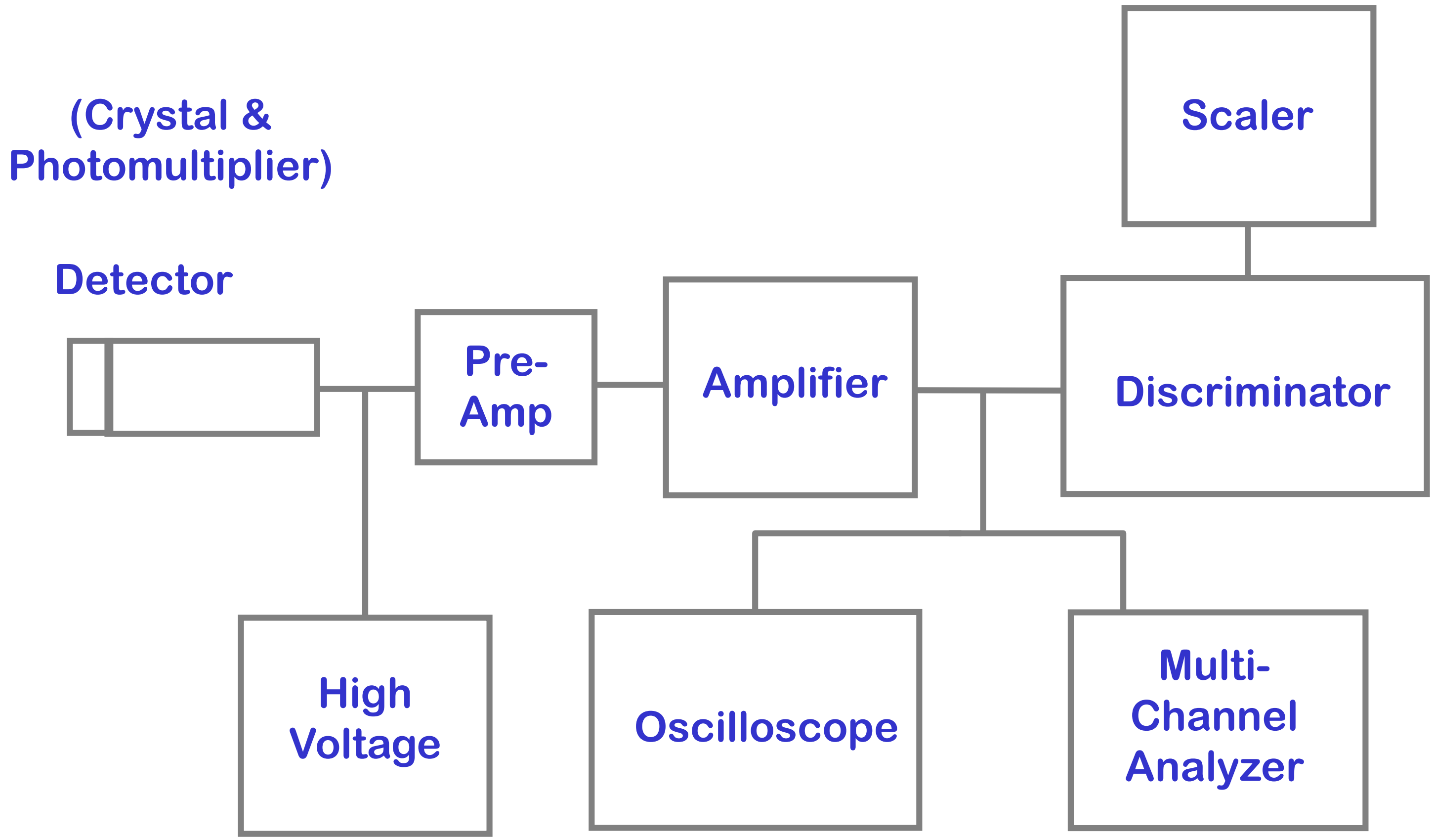
Radionuclides	Half life	Radiation	Instrument	Methods
^{3}H	12.32 y	β	liquid scitilation countiung (LSC)	distilation of the seawater sample, electrolytic enrichment after distillation
^{90}Sr	28.79 y	β(γ)	gas-flow propotional counting (GPC)	chemically separeted ^{90}Y or ^{90}Sr plus ^{90}Y
^{134}Cs and ^{137}Cs	^{134}Cs 2.065 y ^{137}Cs 30.167 y	β γ	gamma spectrometry	direct or pre-concentration (AMP/Cs compound, adsorption on hexacyanoferrates containing either copper, nickel or cobalt)
Plutonium isotopes	^{238}Pu 87.7 y ^{239}Pu 2.411 x 10^{4} y ^{240}Pu 6564 y	α γ	alfa spectrometry, AMS, ICP-MS	radiochemical separation

Liquid scintillation counting (LSC) often requires radiochemical separation

- Standard laboratory method for measuring radiation from beta-emitting nuclides.
- Samples are dissolved or suspended in a "cocktail" containing a solvent (historically benzene or toluene, and small amounts of other additives known as fluors).
  - Beta particles transfer energy to the solvent molecules, which in turn transfer their energy to the fluors;
  - Excited fluor molecules dissipate the energy by emitting light.
  - **Each beta emission (ideally) results in a pulse of light.**
  - Scintillation cocktails may contain additives to shift the wavelength of the emitted light to make it more easily detected.
- Samples are placed in small transparent or translucent (often glass) vials that are loaded into an instrument known as a liquid scintillation counter.

https://www.prl.res.in/~bapat/expt_tech_course/RadDetec/RadDetectInstruFunda.ppt

Generalized Detection System using a Scintillator



https://www.prl.res.in/~bapat/expt_tech_course/RadDetec/RadDetectInstruFunda.ppt

Alpha/beta/gamma counting by gas filled detectors

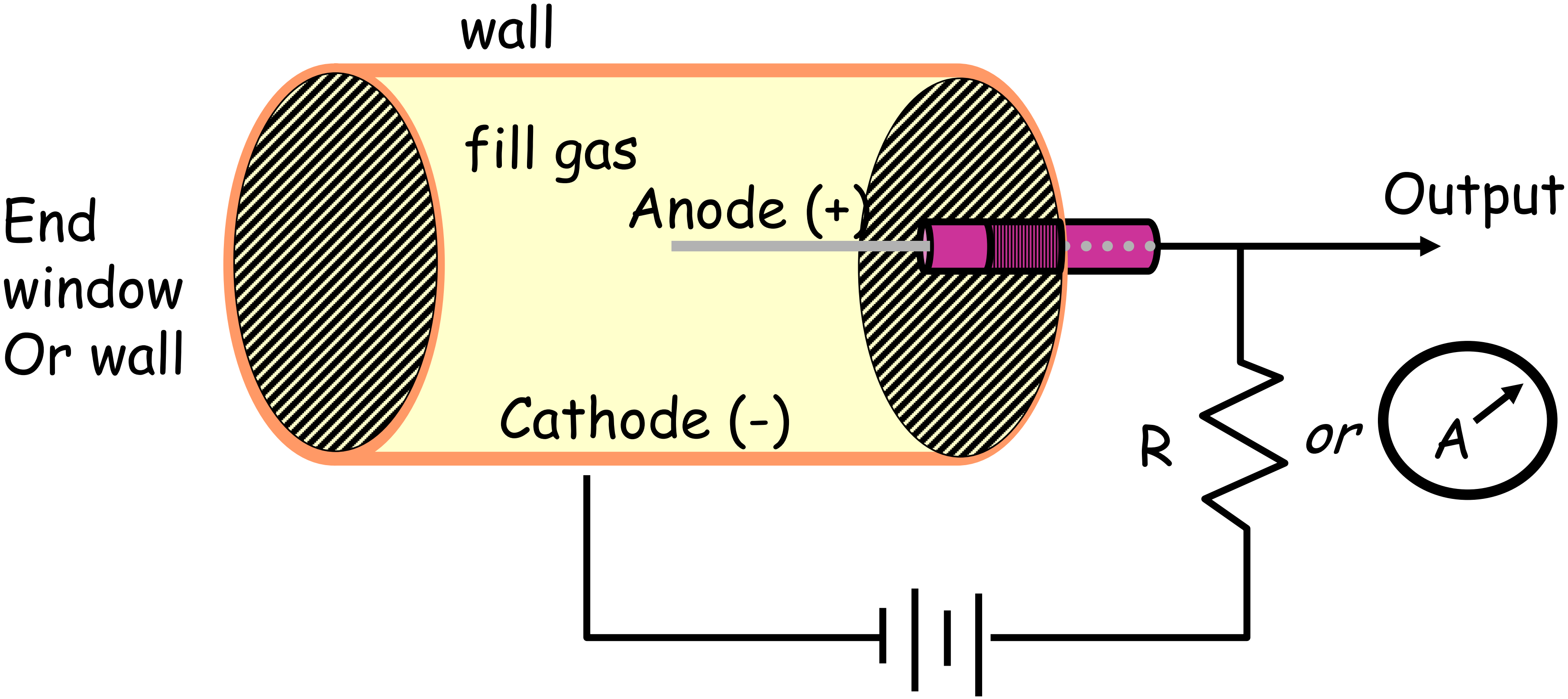
often need radiochemical separation

Gas-Filled Detectors – Components

- Variable voltage source
- Gas-filled counting chamber
- Two coaxial electrodes well insulated from each other
- Electron-pairs
  - produced by radiation in fill gas
  - move under influence of electric field
  - produce measurable current on electrodes, *or*
  - transformed into pulse

https://www.prl.res.in/~bapat/expt_tech_course/RadDetec/RadDetectInstruFunda.ppt

Gas- Filled Detectors – one example



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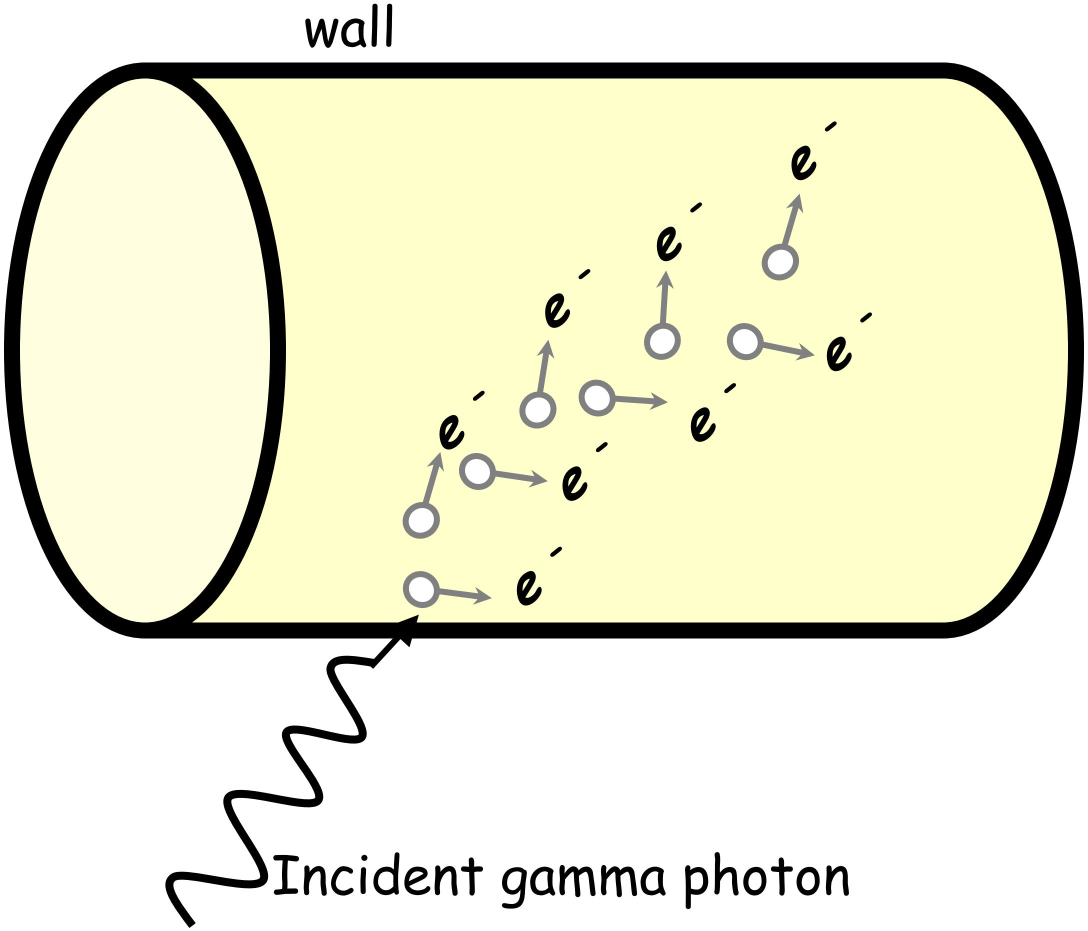
Indirect Ionization Process



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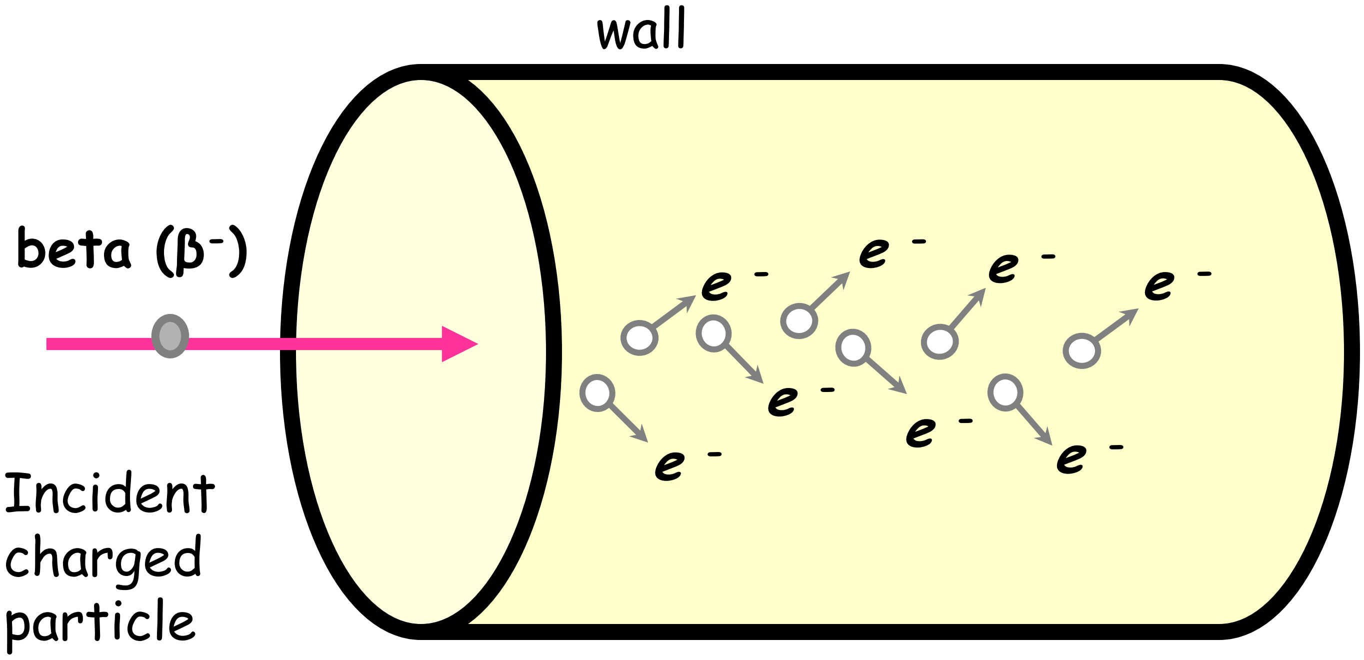
Direct Ionization Process



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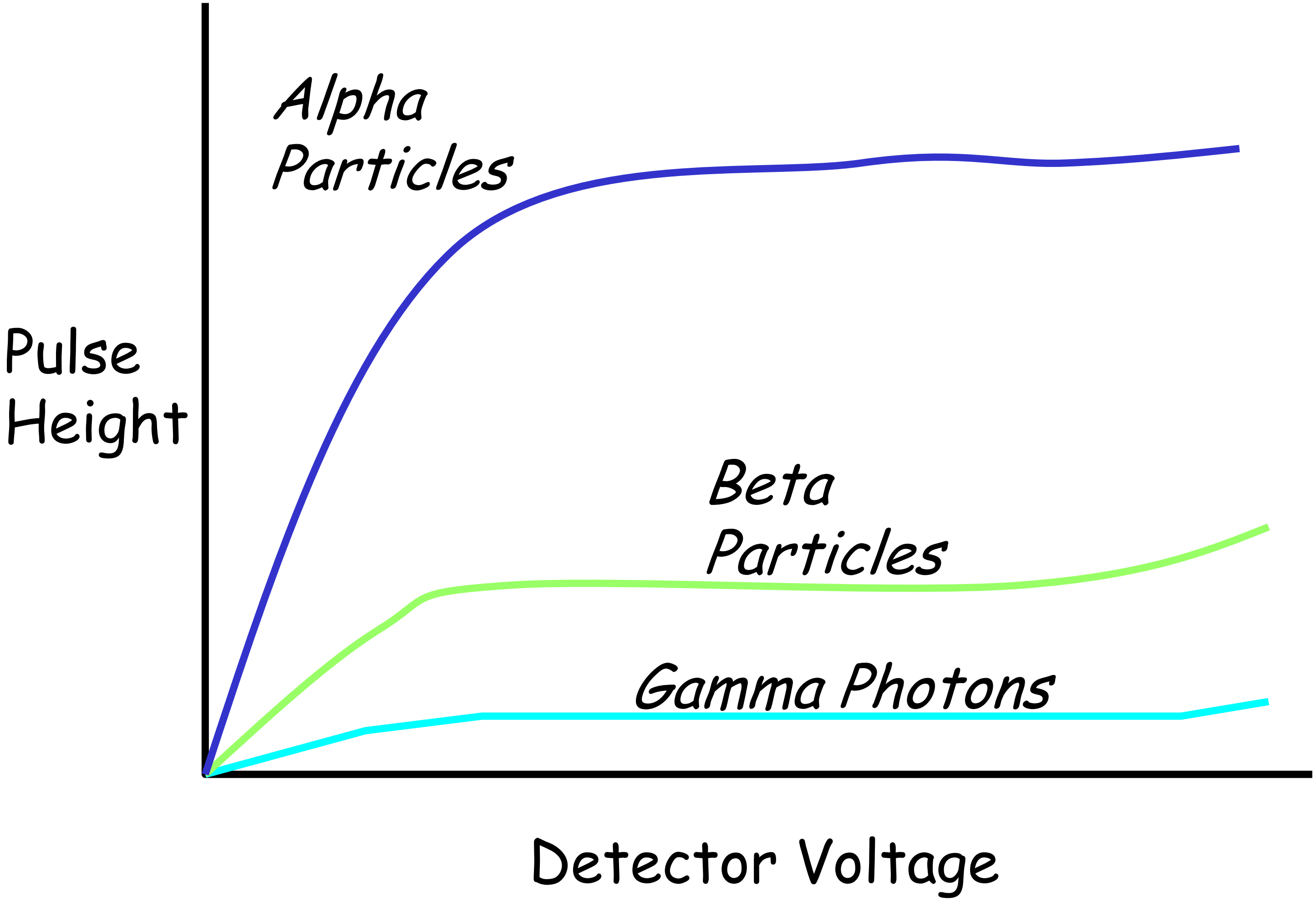
Pulse Height Variation



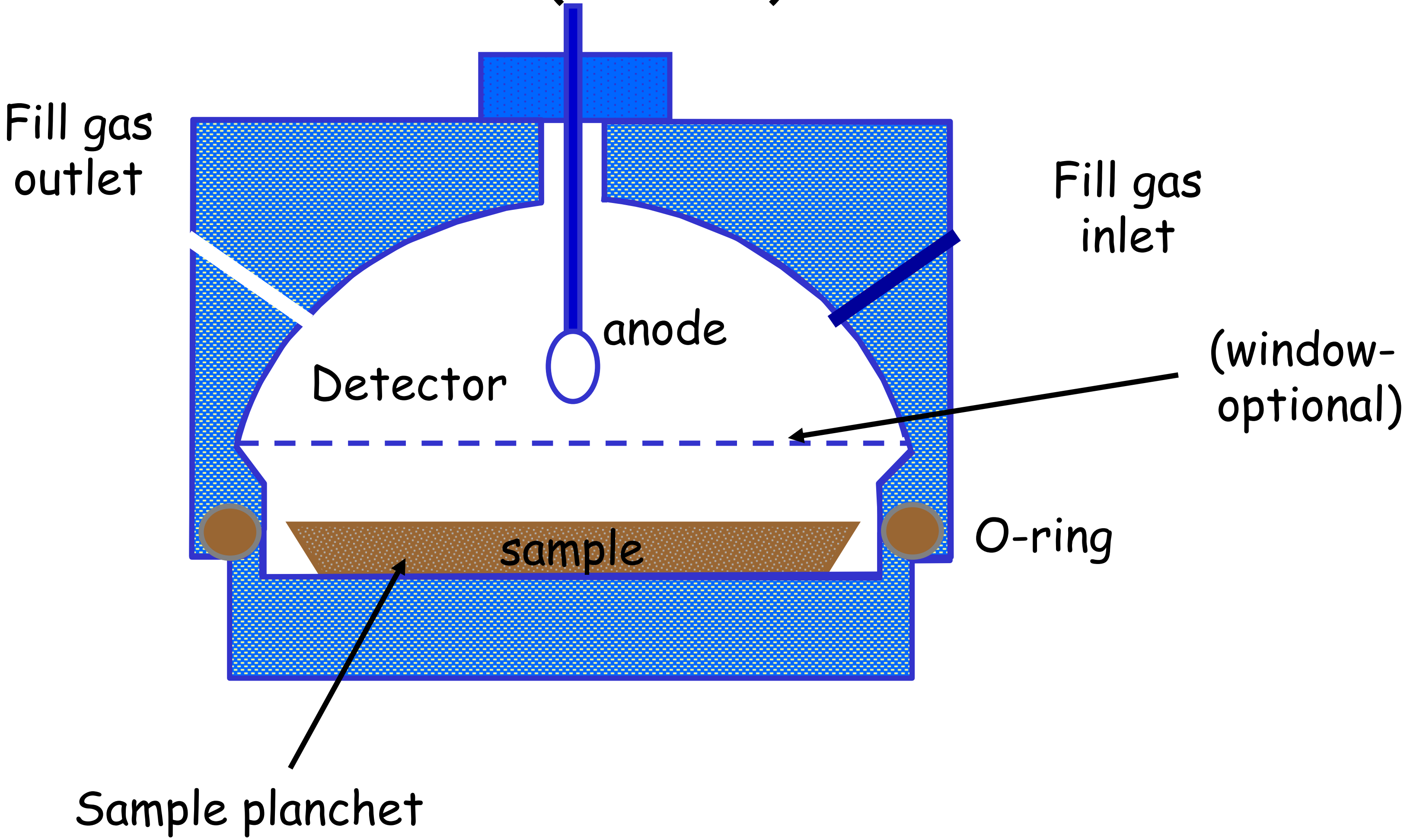
https://www.prl.res.in/~bapat/expt_tech_course/RadDetec/RadDetectInstruFunda.ppt

Other Aspects of Gas-Filled Detectors

- Accuracy of measurement
  - Detector Walls composed of air equivalent material or
  - tissue equivalent
- Wall thickness
  - must allow radiation to enter/ cause interactions
  - alpha radiation requires thin wall (allowed to pass)
  - gammas require thicker walls (interactions needed)
- Sensitivity
  - Air or Fill gas Pressure
  - see next graph

https://www.prl.res.in/~bapat/expt_tech_course/RadDetec/RadDetectInstruFunda.ppt

Gas-Flow Proportional Counter (GPC)



https://www.prl.res.in/~bapat/expt_tech_course/RadDetec/RadDetectInstruFunda.ppt

Gas Flow Proportional, continued

- Fill gas
  - selected to enhance gas multiplication
  - no appreciable electron attachment
  - most common is P-10 (90% Argon and 10% methane)

https://www.prl.res.in/~bapat/expt_tech_course/RadDetec/RadDetectInstruFunda.ppt

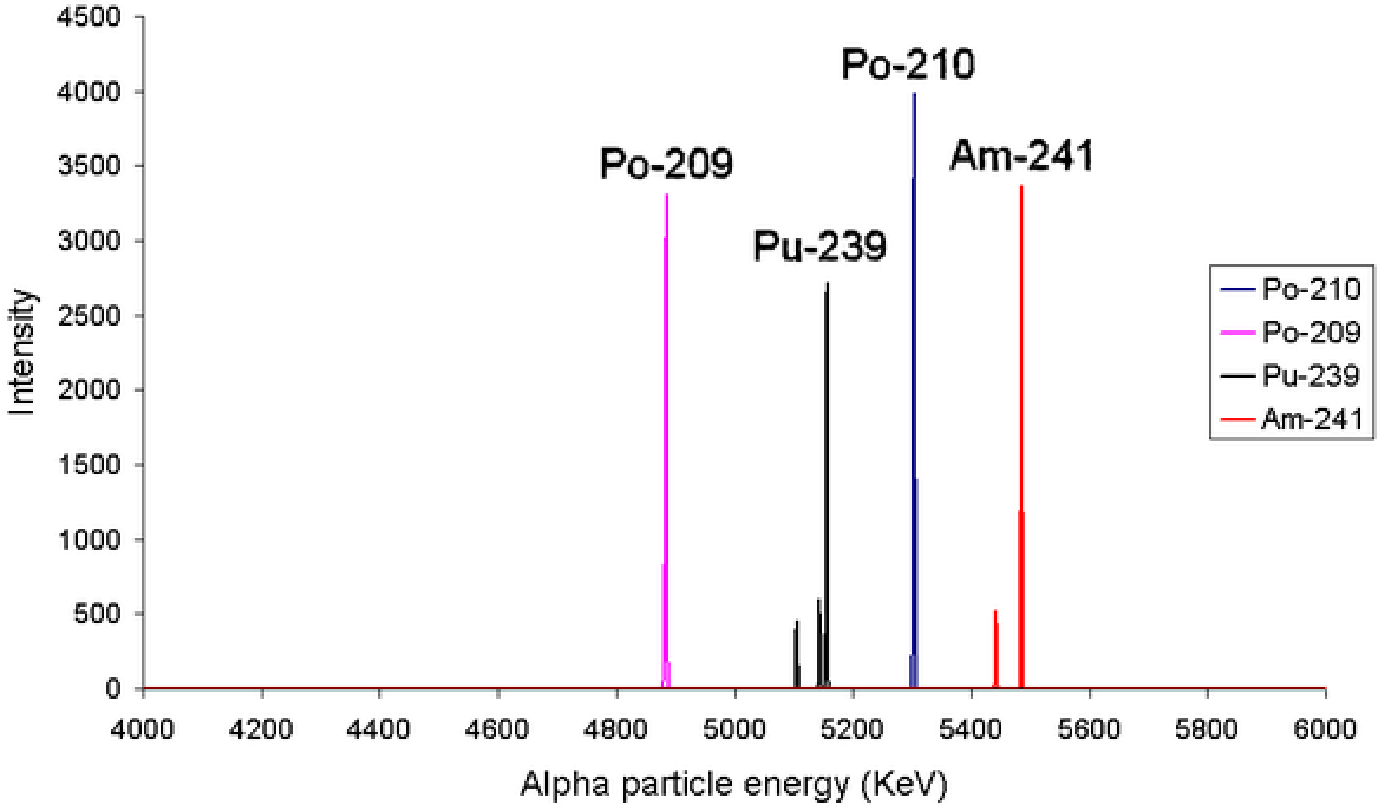
alpha spectrometry

- Stainless steel vacuum chamber
- Accommodates samples up to 50 mm (2 in.) diameter
- Supports the use of alpha detectors up to 1200 mm^2
- Integral vacuum gauge with vacuum/bias interlock
- Bias supply variable to 198 V dc, positive or negative
- Digital display of Counter/Timer, Chamber Pressure, Detecto Bias, Leakage Current, Pulser Energy, Discriminator Energy

http://www.canberra.com/products/radiochemistry_lab/alpha-spectroscopy.asp

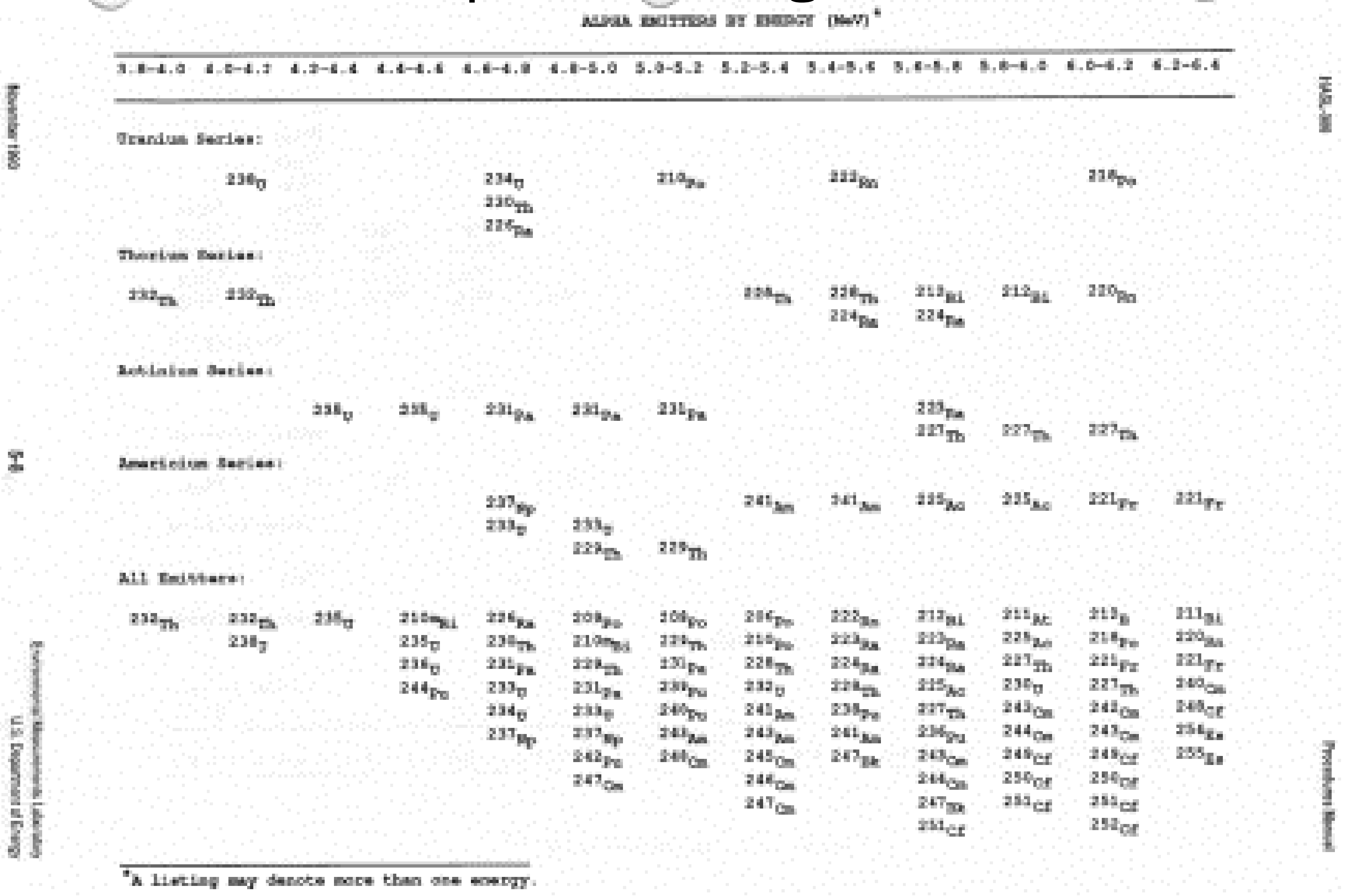
alpha spectrometry

need radiochemical separation



https://en.wikipedia.org/wiki/Alpha-particle_spectroscopy

Table of alpha emitting radionuclides



ALPHA EMITTERS BY ENERGY (MeV)*

3.8-4.0	4.0-4.2	4.2-4.4	4.4-4.6	4.6-4.8	4.8-5.0	5.0-5.2	5.2-5.4	5.4-5.6	5.6-5.8	5.8-6.0	6.0-6.2	6.2-6.4
Uranium Series:												
	^{238}U			^{234}U ^{230}Th ^{226}Ra		^{210}Po		^{222}Rn			^{218}Po	
Thorium Series:												
^{232}Th	^{232}Th						^{228}Th	^{228}Th ^{224}Ra	^{212}Bi ^{224}Ra	^{212}Bi	^{220}Rn	
Actinium Series:												
		^{235}U	^{235}U	^{231}Pa	^{231}Pa	^{231}Pa			^{223}Ra ^{227}Th	^{227}Th	^{227}Th	
Americium Series:												
				^{237}Np ^{233}U	^{233}U ^{229}Th	^{229}Th	^{241}Am	^{241}Am	^{225}Ac	^{225}Ac	^{221}Fr	^{221}Fr
All Emitters:												
^{232}Th	^{232}Th ^{238}U	^{235}U	^{210m}Bi ^{235}U ^{236}U ^{244}Pu	^{226}Ra ^{230}Th ^{231}Pa ^{233}U ^{234}U ^{237}Np	^{208}Po ^{210m}Bi ^{229}Th ^{231}Pa ^{233}U ^{237}Np ^{242}Pu ^{247}Cm	^{209}Po ^{229}Th ^{231}Pa ^{239}Pu ^{240}Pu ^{243}Am ^{248}Cm	^{206}Po ^{210}Po ^{228}Th ^{232}U ^{241}Am ^{243}Am ^{245}Cm ^{246}Cm ^{247}Cm	^{222}Rn ^{223}Ra ^{224}Ra ^{228}Th ^{238}Pu ^{241}Am ^{247}Bk	^{212}Bi ^{223}Ra ^{224}Ra ^{225}Ac ^{227}Th ^{236}Pu ^{243}Cm ^{244}Cm ^{247}Bk ^{251}Cf	^{211}At ^{225}Ac ^{227}Th ^{230}U ^{243}Cm ^{244}Cm ^{249}Cf ^{250}Cf ^{251}Cf	^{213}Bi ^{218}Po ^{221}Fr ^{227}Th ^{242}Cm ^{243}Cm ^{249}Cf ^{250}Cf ^{251}Cf ^{252}Cf	^{211}Bi ^{220}Rn ^{221}Fr ^{240}Cm ^{248}Cf ^{254}Es ^{255}Es

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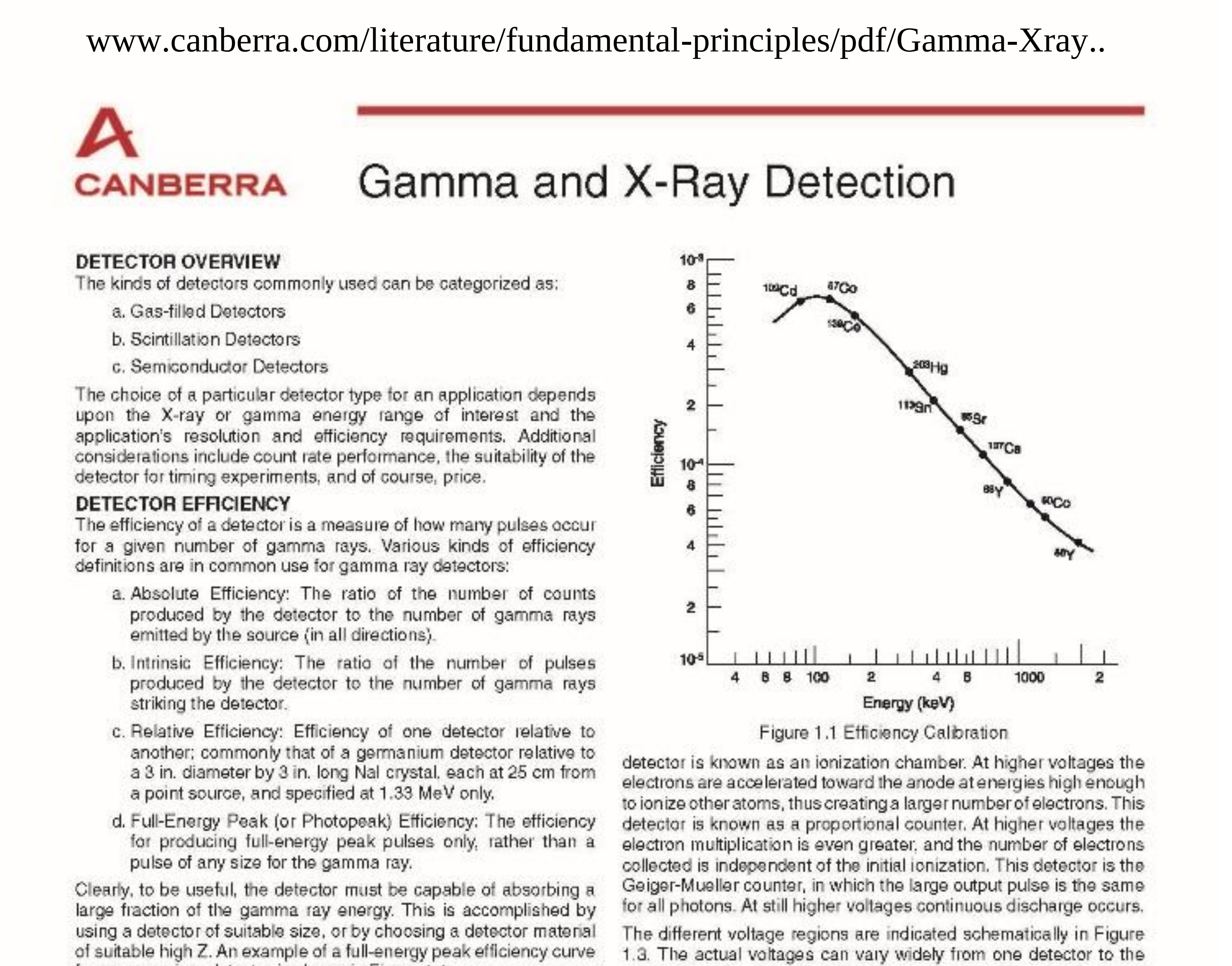
*A listing may denote more than one energy.

Gamma and X-Ray Detection

www.canberra.com/literature/fundamental-principles/pdf/Gamma-Xray..



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Gamma and X-Ray Detection

DETECTOR OVERVIEW

The kinds of detectors commonly used can be categorized as:

a. Gas-filled Detectors
b. Scintillation Detectors
c. Semiconductor Detectors

The choice of a particular detector type for an application depends upon the X-ray or gamma energy range of interest and the application's resolution and efficiency requirements. Additional considerations include count rate performance, the suitability of the detector for timing experiments, and of course, price.

DETECTOR EFFICIENCY

The efficiency of a detector is a measure of how many pulses occur for a given number of gamma rays. Various kinds of efficiency definitions are in common use for gamma ray detectors:

a. Absolute Efficiency: The ratio of the number of counts produced by the detector to the number of gamma rays emitted by the source (in all directions).

b. Intrinsic Efficiency: The ratio of the number of pulses produced by the detector to the number of gamma rays striking the detector.

c. Relative Efficiency: Efficiency of one detector relative to another; commonly that of a germanium detector relative to a 3 in. diameter by 3 in. long NaI crystal, each at 25 cm from a point source, and specified at 1.33 MeV only.

d. Full-Energy Peak (or Photopeak) Efficiency: The efficiency for producing full-energy peak pulses only, rather than a pulse of any size for the gamma ray.

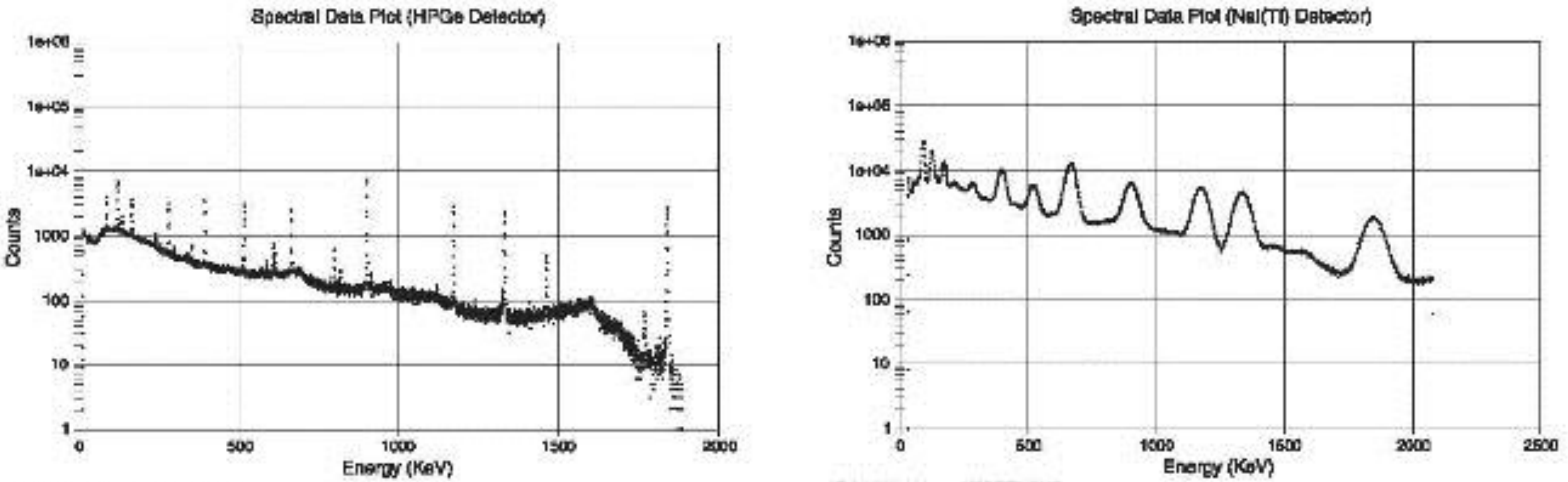
Clearly, to be useful, the detector must be capable of absorbing a large fraction of the gamma ray energy. This is accomplished by using a detector of suitable size, or by choosing a detector material of suitable high Z. An example of a full-energy peak efficiency curve

Figure 1.1 Efficiency Calibration

detector is known as an ionization chamber. At higher voltages the electrons are accelerated toward the anode at energies high enough to ionize other atoms, thus creating a larger number of electrons. This detector is known as a proportional counter. At higher voltages the electron multiplication is even greater, and the number of electrons collected is independent of the initial ionization. This detector is the Geiger-Mueller counter, in which the large output pulse is the same for all photons. At still higher voltages continuous discharge occurs.

The different voltage regions are indicated schematically in Figure 1.3. The actual voltages can vary widely from one detector to the

Sample spectrum



Datasource: NBSSTD.CNF
Live Time: 4000 sec
Real Time: 4020 sec
Start: 1 : 10.66 (keV)
Stop: 4096 : 1880.90 (keV)

Datasource: NBSSTD.CNF
Live Time: 1000 sec
Real Time: 1030 sec
Start: 1 : 5.40 (keV)
Stop: 1024 : 2085.16 (keV)

Figure 1.2

Operating voltages depend upon the fill gas as well as the geometry. For X rays, noble gases are often used, with xenon, krypton, neon and argon common choices. Xenon and krypton are selected for higher energy X rays or to get higher efficiencies, while neon is selected when it is desired to detect low energy X rays in the presence of unwanted higher energy X rays. Sometimes gas mixtures are used, such as P-10 gas, which is a mixture of 90% argon and 10% methane. Gas pressures are typically one atmosphere. The 2006 preamplifier available for proportional counters is shown in Figure 1.4.

The discharge produced by an ionization must be quenched in order for the detector to be returned to a neutral ionization state for the next pulse. This is accomplished by using a fill gas that contains a small amount of halogen in addition to a noble gas. The voltage drop across a large resistor between the anode and bias supply will also serve to quench the discharge since the operating voltage will be reduced below the plateau.

The Geiger-Mueller counter is inactive or "dead" after each pulse until the quenching is complete. This dead time can be hundreds of microseconds long, which limits the counter to low count rate applications.

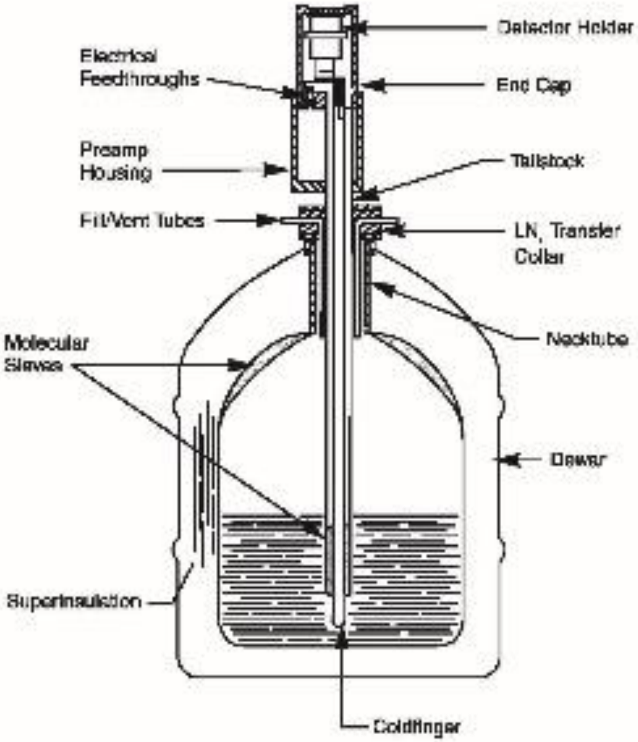


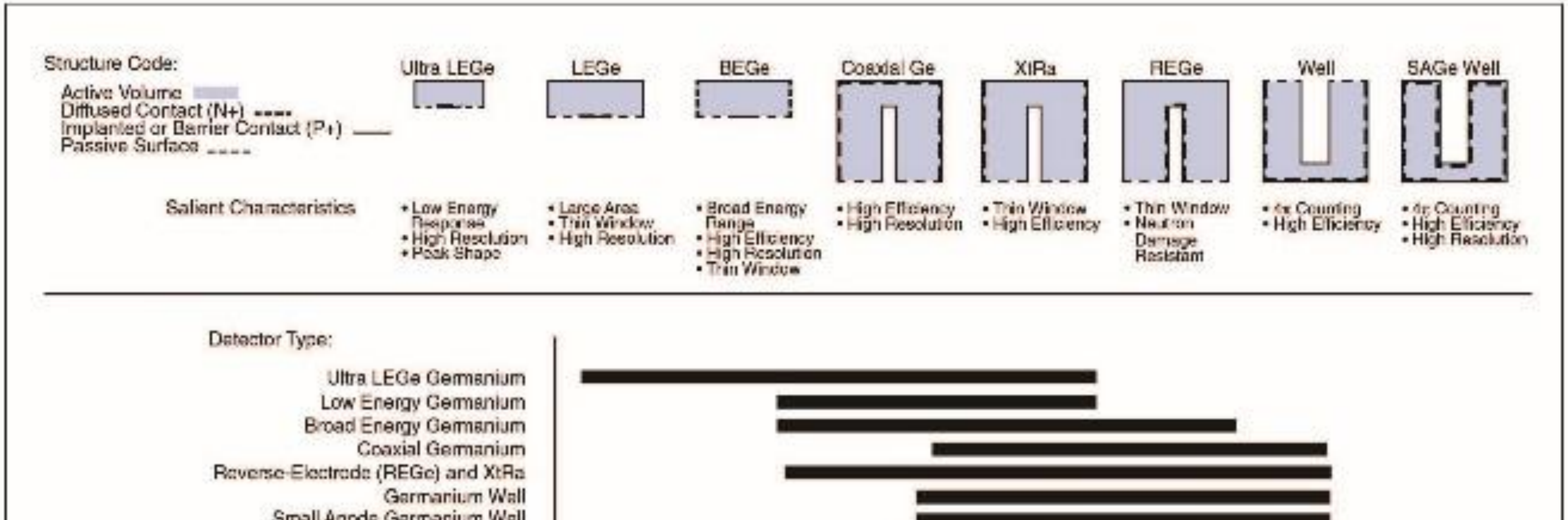
Figure 1.8 Model 7500SL Vertical Dipstick Cryostat

At low energies, detector efficiency is a function of cross-sectional area and window thickness while at high energies total active detector volume more or less determines counting efficiency. Detectors having thin contacts, e.g. Si(Li), Low-Energy Ge and Reverse Electrode Ge detectors, are usually equipped with a Be or composite carbon cryostat window to take full advantage of their intrinsic energy response.

Coaxial Ge detectors are specified in terms of their relative full-energy peak efficiency compared to that of a 3 in. x 3 in. NaI(Tl) Scintillation detector at a detector to source distance of 25 cm. Detectors of greater than 100% relative efficiency have been fabricated from germanium crystals ranging up to about 75 mm in diameter. About two kg of germanium is required for such a detector.

Curves of detector efficiency vs. energy for various types of Ge detectors can be found in the Detector Product Section of this catalog.

1. A.C. Melissinos, Experiments in Modern Physics, Academic Press, New York (1966), p. 178.



My message to you

- You can buy good instruments if you have money or you can produce instruments if you know basics and have good skill.
- You need to learn appropriate radiochemical separation techniques/methods to fit your objectives.
- Appropriate methods combined with good instruments will give you “best in the world” results.

Thank you for your attention!!

One good example: Ultra low background gamma-ray measurement of ^{134}Cs and ^{137}Cs in seawater at underground laboratories

Radiochemical separation of radiocaesium from seawater by improved AMP/Cs method (Aoyama and Hirose 2008)

and

Gamma-ray spectrometry at underground laboratory

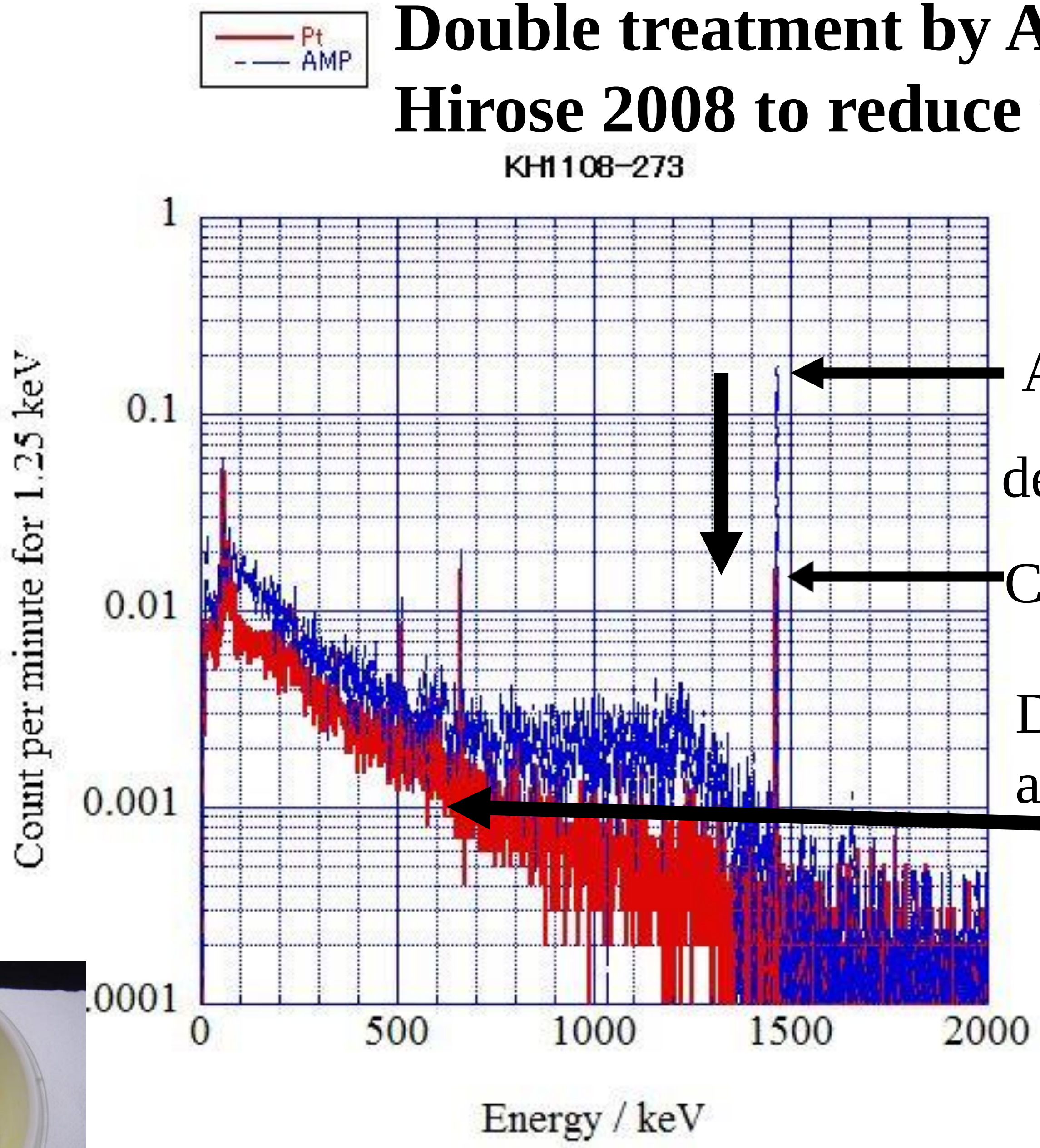
Underground laboratories in Japan

Ogoya underground laboratory
(Kanazawa University)
Y. Hahajima

Located inside a tunnel – a former Cu-mine
Covered by 135 m rock

Shields from old lead from
Kanazawa castle

Double treatment by Aoyama and Hirose 2008 to reduce ^{40}K

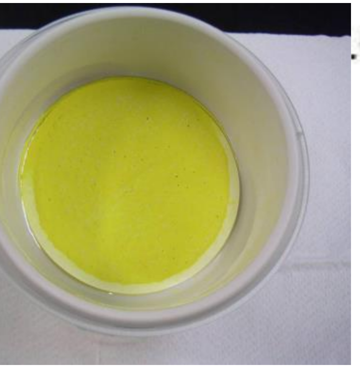


AMP/Cs compound

decrease 40K content

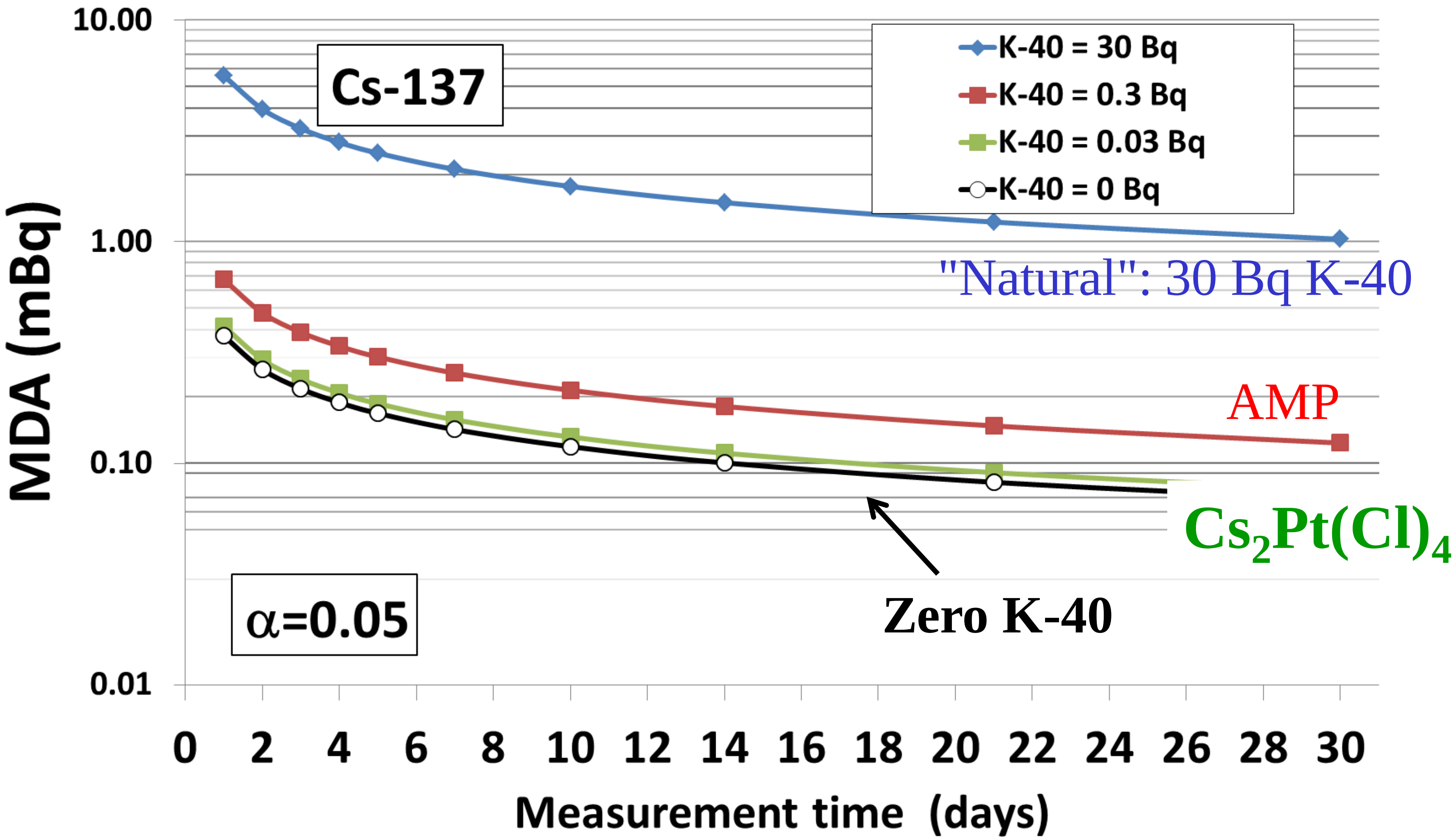
Cs2Pt(Cl)4 precipitat

Decrease compton
as expected



<- AMP/Cs compound

$Cs_2Pt(Cl)_4$ precipitate ->



Detection limits (MDA) for ^{137}Cs in a **<u>4-g sea water precipitate sample,</u>** measured in a 2-kg well-detector (Ge-12 in HADES), as a function of measurement time for different activities of ^{40}K in the sample.

Lutter et al., Nukleonika, 2015

Ca. 0.1 mBq of ^{137}Cs can be measured