ESPECTROMETRÍA GAMMA

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GRUPO RADÓN

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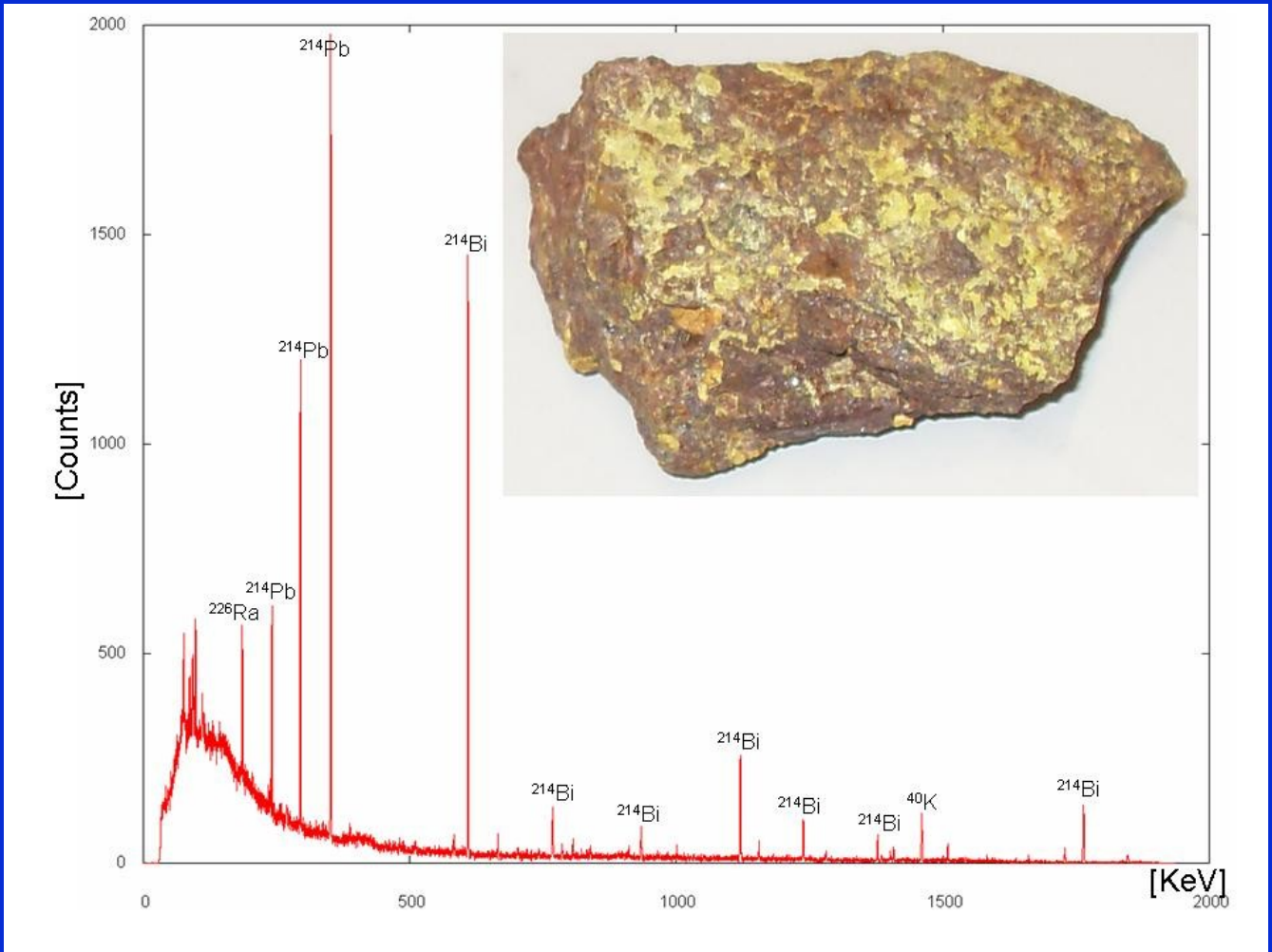
Laboratorio de Radiactividad Ambiental

Qué esperas obtener de esta clase ?



The gamma-ray spectrum of natural uranium, showing about a dozen discrete lines superimposed on a smooth continuum, allows the identification the nuclides 226Ra, 214Pb, and 214Bi of the uranium decay chain. ***(Wikipedia)***

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1. **Introduction**
2. Interaction of radiation with matter
3. Detection system
4. Examples: steel factories and natural radioactivity
5. Practical activity
6. Activity: “One minute paper”

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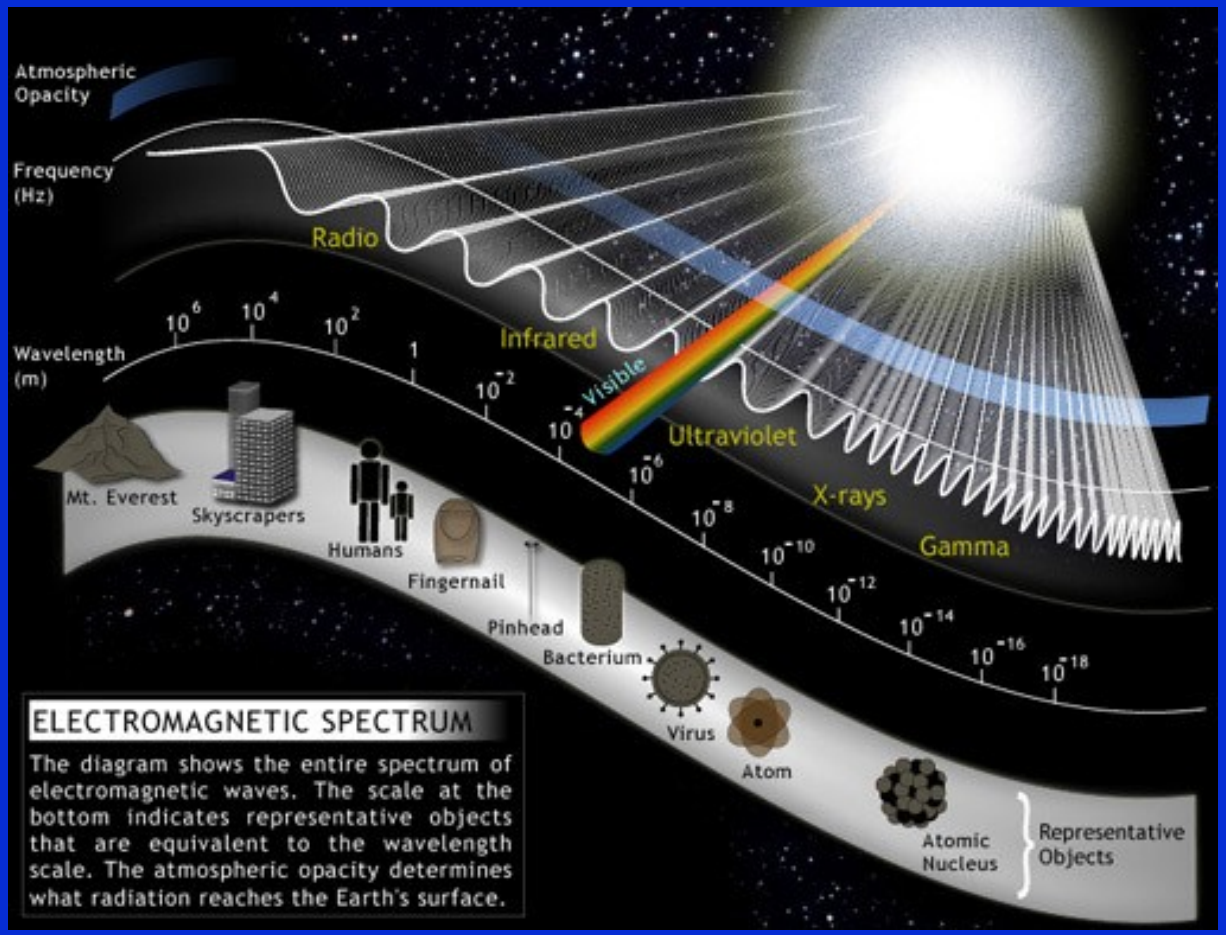
Ionizing radiation: Radiation with enough energy to detach electrons from atoms or molecules, thus ionizing them.



keV
10^3 eV

MeV
10^6 eV

Alpha decay

Beta decay

Gamma decay

LET'S REMEMBER

- **Activity (A):** Number of disintegrations per second
- **Half life ($T_{1/2}$):** Neccesary time for an isotope to decrease its nucleus by half
- **Decay constant (λ):** Probability of disintegration by time
- **Decay chain:** chained series of transformations (4 Natural decay chains)

$$A = A_0 \, e^{-\lambda t}$$

$$T_{1/2} = 0.693/\lambda$$

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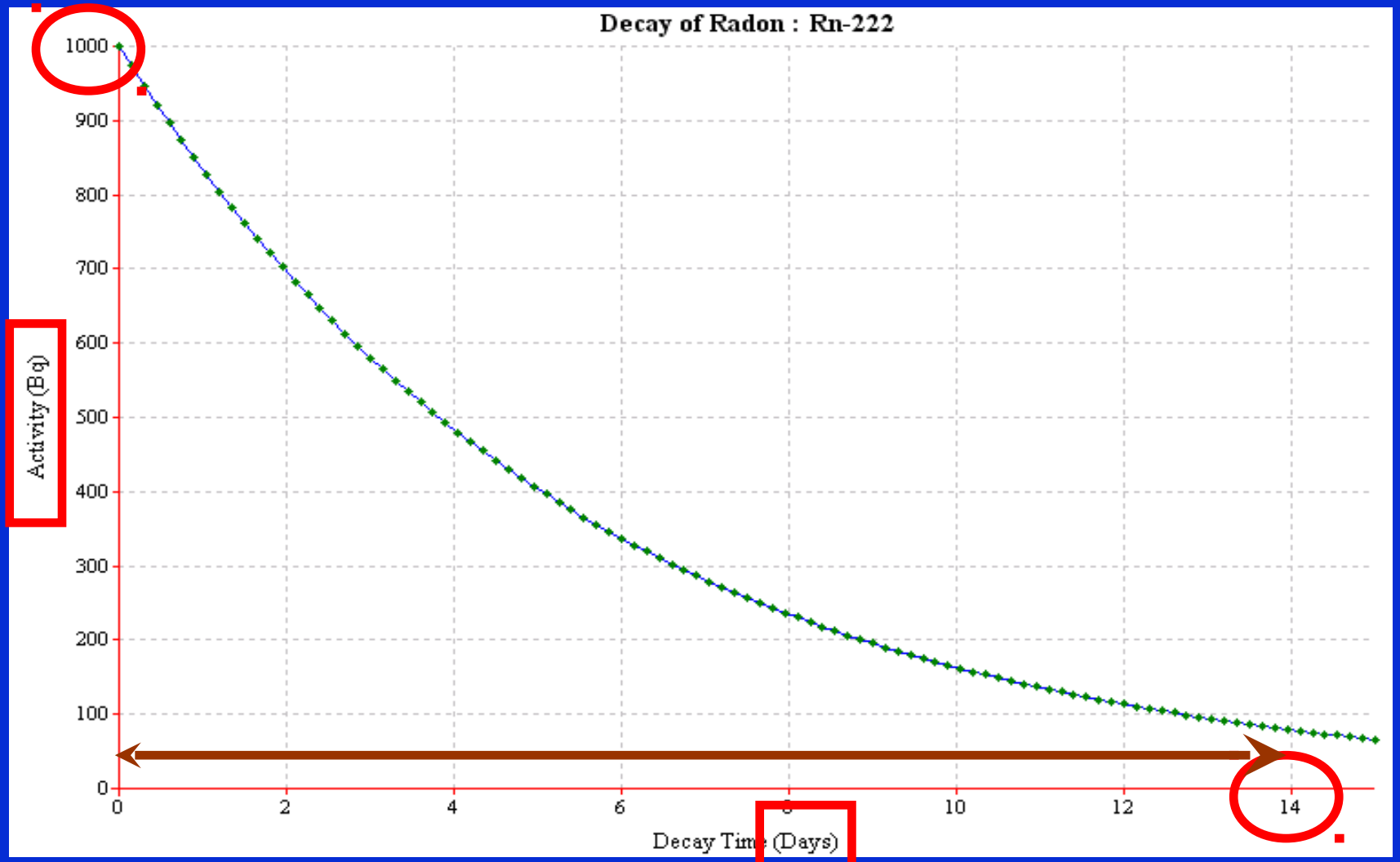

Units on radioactivity

- Becquerel (Bq) : unit of activity in the International System of Units ►1Bq = 1 DPS (disintegration / second.)
- Curie (Ci) : Old unit of activity ►1 Ci = $3.7 \cdot 10^{10}$ Bq
- Concentration : Bq/kg, Bq/l, Bq/m^3
- Sievert (Sv) : Unit for equivalent dose

Gammna decay: Photon's emission by a nucleus when reaching steady state of energy.

Interaction of radiation (photon) with matter (detection's material)

- **Gamma line is the fingerprint of a radionuclide**
- **One radionuclide can have several gamma lines with different probabilities and different energies**
- **X Rays ↔ Gamma Rays (with different energies)**
- **Gamma rays = Nucleus**
- **X Rays = Atomic crust**

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INTERACTION OF RADIATION WITH MATTER

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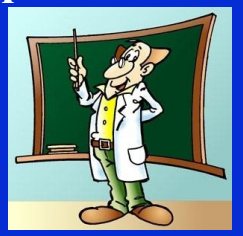
- Photoelectric effect
- Compton effect
- Pair production

- Charged particles, for example, alpha and beta particles, produce a signal within a detector by ionization and excitation of the detector material directly.



- Gamma photons are uncharged and consequently cannot do this
- Gamma-ray detection depends upon other types of interaction which transfer the gamma-ray energy to electrons within the detector material
- Excited electrons charge and lose their energy by ionization and excitation of the atoms of the detector medium, giving rise to many electron–hole pairs
- The absorption coefficient for gamma radiation in gases is low and all practical gamma ray detectors depend upon interaction with a solid
- The electron–hole pairs can be collected and presented as an electrical signal.

Photoelectric effect

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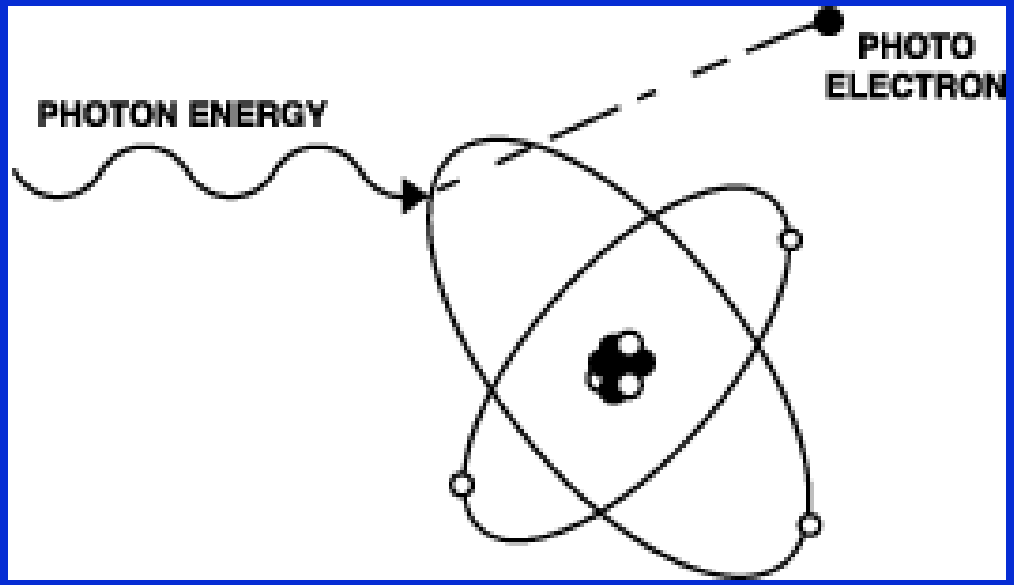
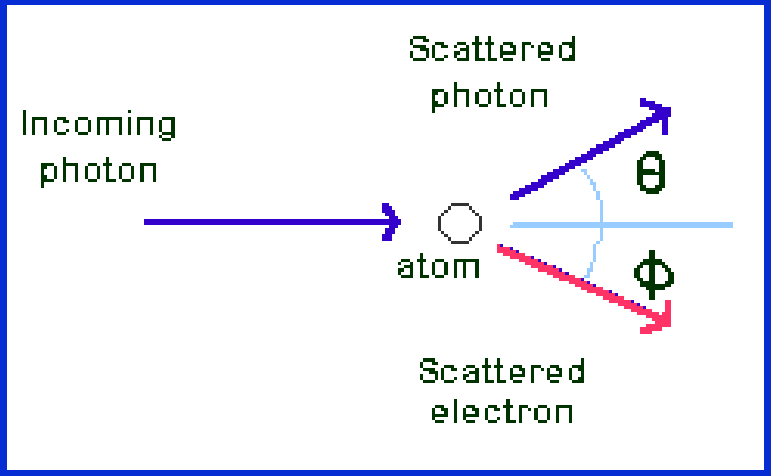
- The photon interacts with the atom and gives ALL its energy to one electron: one part of the energy is used as kinetic energy and the rest is used to remove electron from the atom



Compton effect

- "Elastic collision" : pool balls
- Main interaction of gamma rays
- The photon collides with electron and hands over part of its energy to it. The angle through which the photon is scattered, the energy handed on to the electron, and energy lost by the photon are interconnected



Pair production

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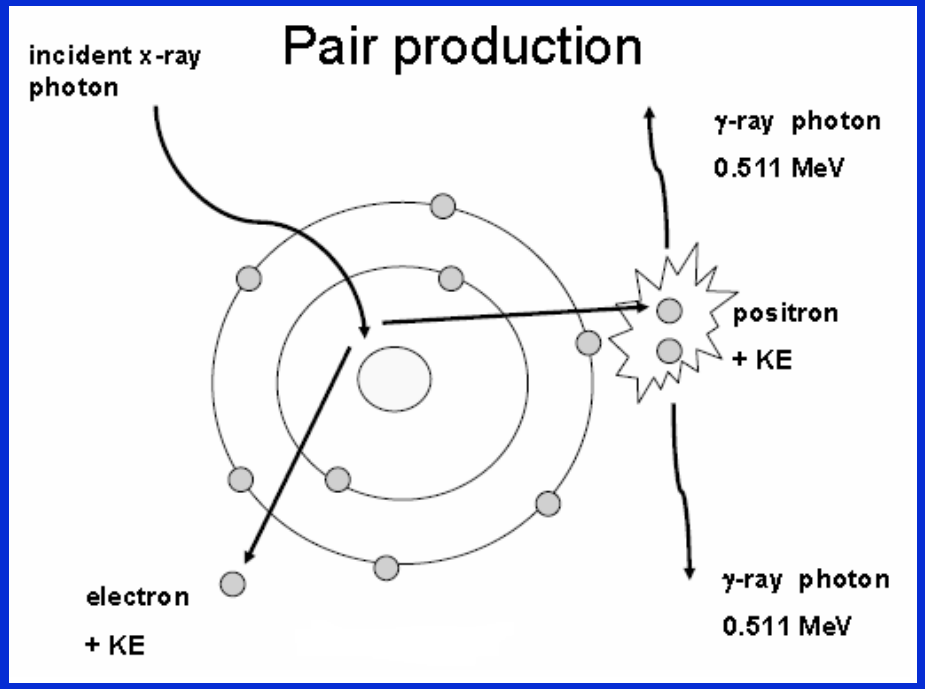
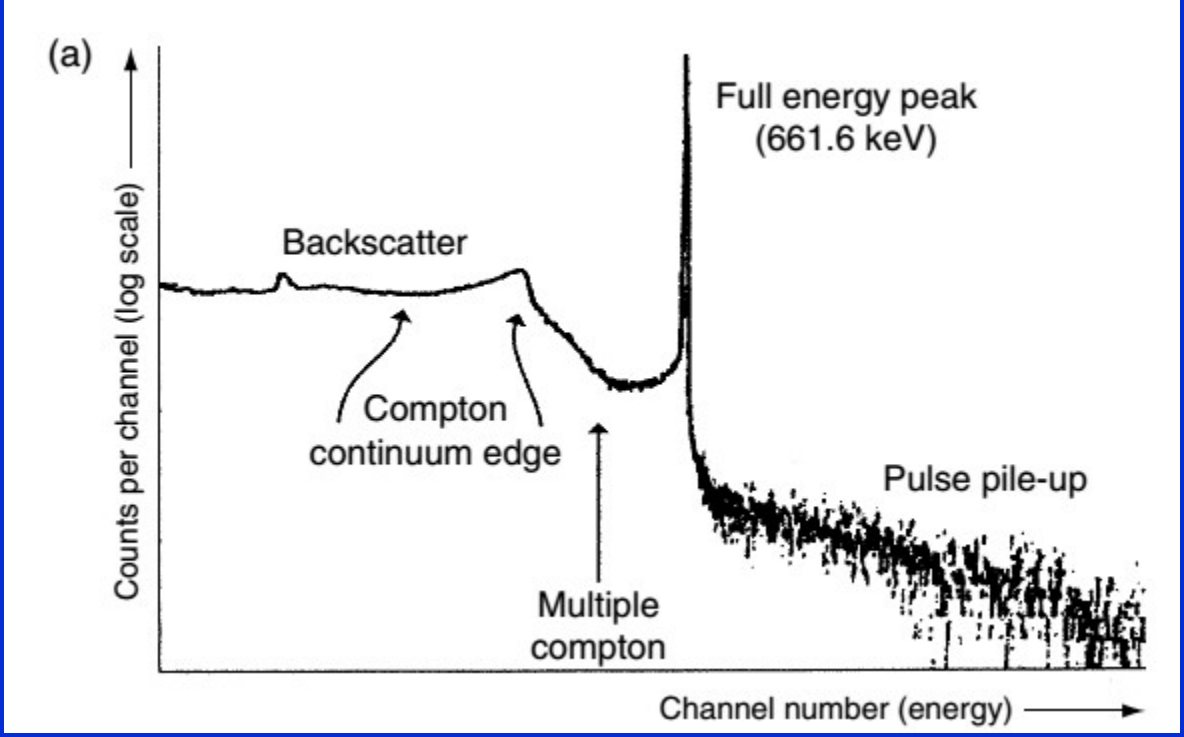
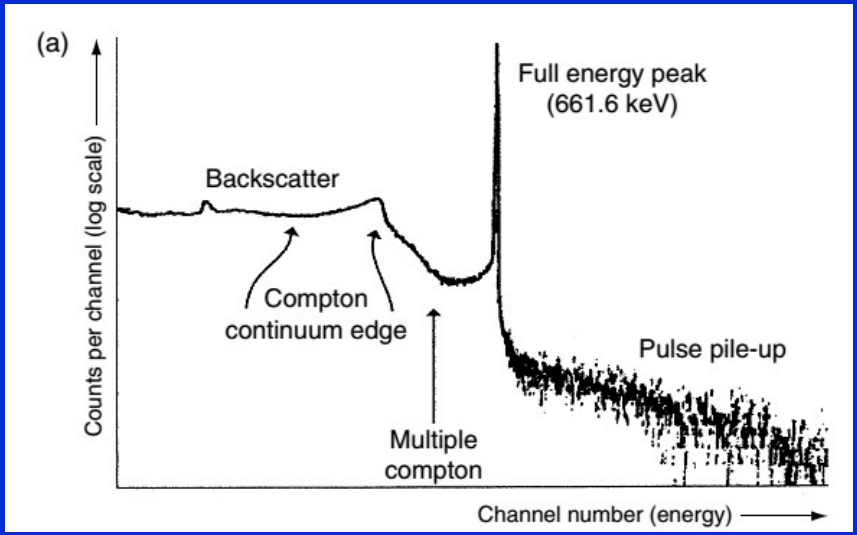
- When the photon with energy in excess of 1.02 MeV passes close to the nucleus of an atom, the photon disappears, and a positron (e^+) and an electron (e^-) appear
- Annihilation reaction: positron interacts with electrons and creates 2 annihilation photons each of 0.51 MeV

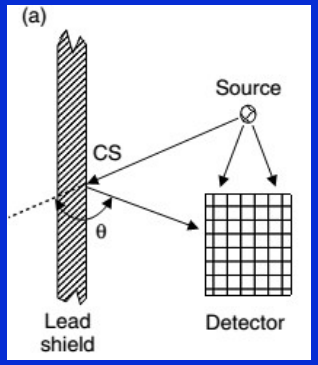


- Photoelectric interactions are dominant at low energy
- Pair production at high energy
- Compton scattering being most important in the mid-energy range
- In practice, evidence of pair production is only seen within a gamma-ray spectrum when the energy is rather more than 1022 keV

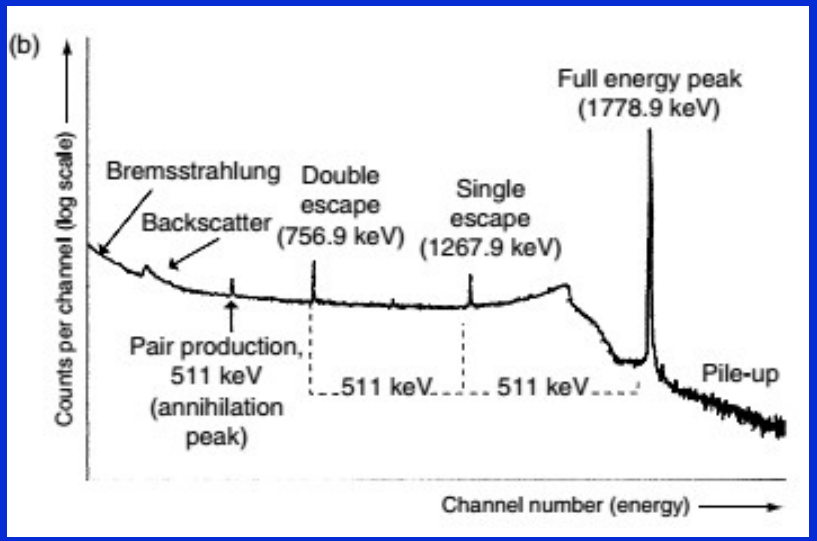


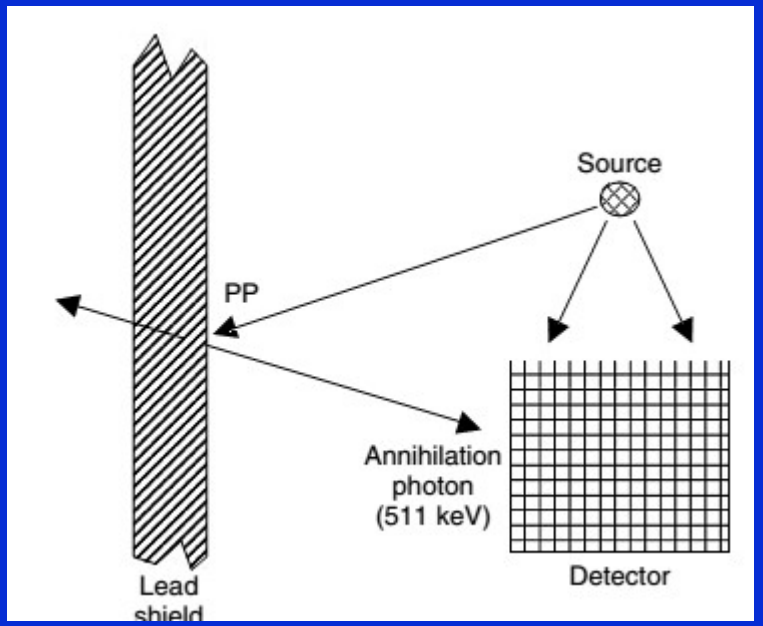


- Counts to appear in the spectrum above the full energy peak (apart from the natural background): due to random summing(sometimes referred to as pile-up), determined by the statistical probability of two gamma-rays being detected at the same time and therefore on the sample count rate
- The most troublesome photoelectric interactions will be those with the shielding, usually lead. There is a significant possibility that this fluorescent X-ray may escape the shielding and that it will be detected by the detector. The result will be a number of X-ray peaks in the gamma spectrum in the region 70–85 keV: problems for low energy measurements (solve with Cd and Cu)
- The normal geometric arrangement of source–detector shielding means that most gamma-rays are scattered through a large angle by the shielding: backscattering (difficult to solve)



Pair production: surroundings of the detector give rise to the annihilation peak at 511 keV in the spectrum. This is caused by the escape of one of the 511 keV photons from the shielding, following annihilation of the pair production positron. The annihilation peak is clearly visible in the spectrum of ^{28}Al (see Figure) but not in that of ^{137}Cs **(Why not ???)**





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- Instrumentation (HPGe) available in most of labs
- No radiochemistry is needed
- Pre-treatment of the sample is very simple
- A large number of radionuclides can be measured
- Detection limits acceptable for environmental determinations

- Each geometry needs different efficiency calibration
- Very dependent on density of sample
- Time consuming (several days in some cases)
- Maintenance of detector is critical (refrigeration)
- Monitoring of background levels is necessary

PRACTICAL POINTS

- Gamma spectrometry using germanium detectors is the best technique for identifying and quantifying radionuclides. This is due to the very sharply defined and characteristic energies of gamma-rays which are produced by the great majority of radionuclides.

- There are a small number of 'pure beta emitters', which do not emit gamma radiation. These cannot be identified by gamma spectrometry (^{3}H, ^{14}C, ^{90}Sr).

- X-ray energies will tell you the element present, but not which isotope.

- Decay schemes give vital information on whether gammas are in 'cascade'. This has great significance in true coincidence summing.

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DETECTION SYSTEM

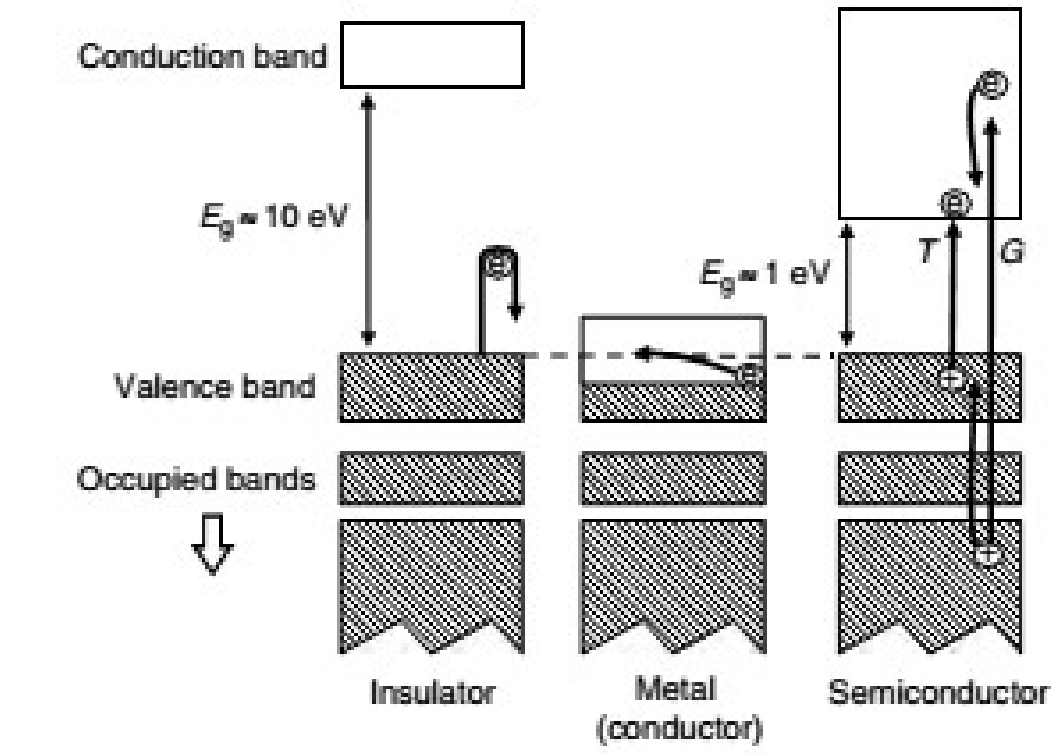
Radiation (gamma emission)
INTERACTS
with matter (detector's material)

It is neccesary to TRANSLATE the interaction
into electrical signal
which can be MEASURED

PROPERTIES OF AN IDEAL DETECTOR FOR GAMMA SPECTROMETRY.

- output proportional to gamma-ray energy;
- good efficiency, i.e. high absorption coefficient, highZ;
- easy mechanism for collecting the detector signal;
- good energy resolution;
- good stability over time, temperature and operating parameters;
- reasonable cost;
- reasonable size

The band structure of solids



Creation of charge carriers by gamma radiation

- The interaction of a gamma-ray with the semiconductor material will produce primary electrons with energies considerably greater than thermal energies
- Electric field, carriers will migrate up (electrons) or down (holes) the field gradient.
- The number of electron–hole pairs produced, ***n***, will be related directly to the gamma-ray energy absorbed
- One important component of the detector resolution is a function of ***n***
- Avoid trapping centers which can make difficult mobility of carriers: the detector material must be available, at reasonable cost, with a high purity and as near perfect as possible crystalline state

Suitable semiconductors for gamma-ray detectors

- have as large an absorption coefficient as possible (i.e. high atomic number);
- provide as many electron–hole pairs as possible per unit energy;
- allow good electron and hole mobility;
- be available in high purity as near perfect single crystals;
- be available in reasonable amounts at reasonable cost.

HPGe type *p* and *n*

A semiconductor material containing equal numbers of electrons and holes is described as an **intrinsic semiconductor**.

✓acceptor impurities when distributed throughout the semiconductor material give rise to extra energy states just above the valence band, called acceptor states. Germanium with this type of impurity would be called **p-type germanium** ('p' for positive acceptor impurities)

✓The impurity atom is a donor atom sitting in a donor site, it will introduce donor states just below the conduction band. Germanium with such impurities is **n-type germanium** ('n' for negative donor impurities).

✓The p-type material has an excess of holes and the n-type an excess of electrons.

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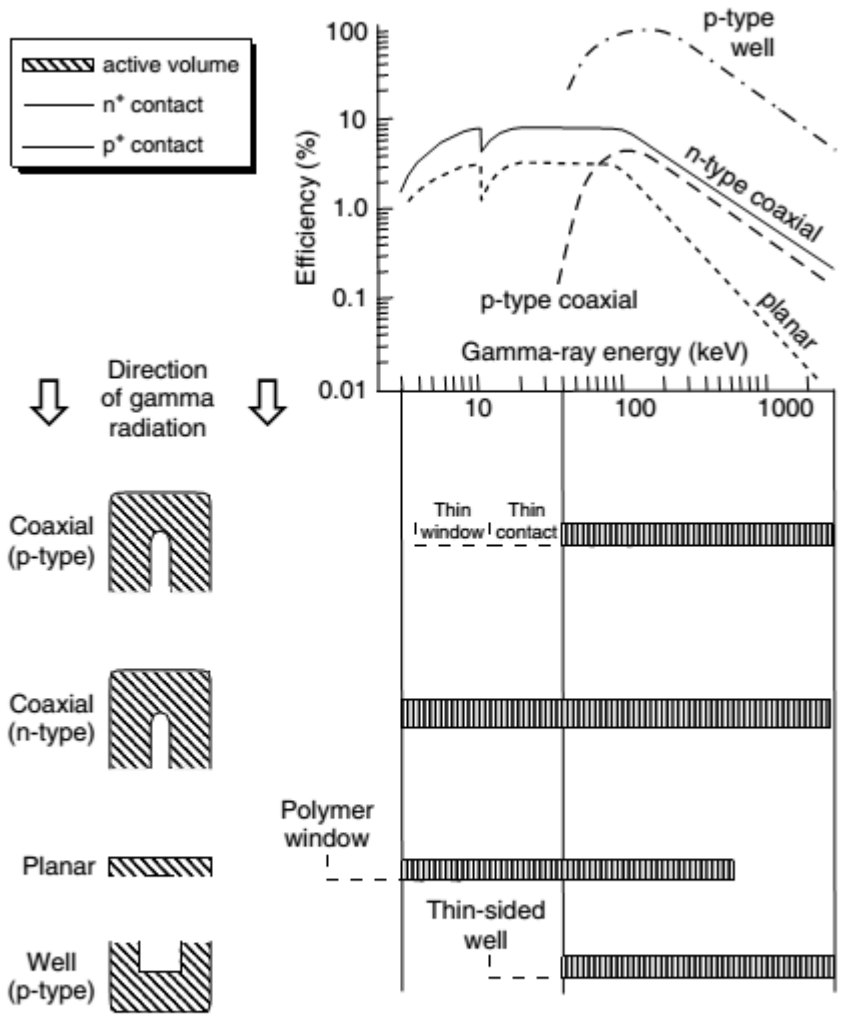
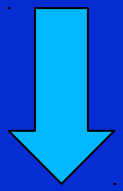


Figure 3.6 Configurations of detector generally available, together with schematic efficiency curves and an indication of the energy range over which they might be used

GERMANIUM DETECTORS ARE OPERATED AT LOW TEMPERATURE IN ORDER TO REDUCE ELECTRONIC NOISE AND THEREBY ACHIEVE AS HIGH A RESOLUTION AS POSSIBLE



The most common means of providing a suitably low temperature is cooling with liquid nitrogen (boiling point 77 K).

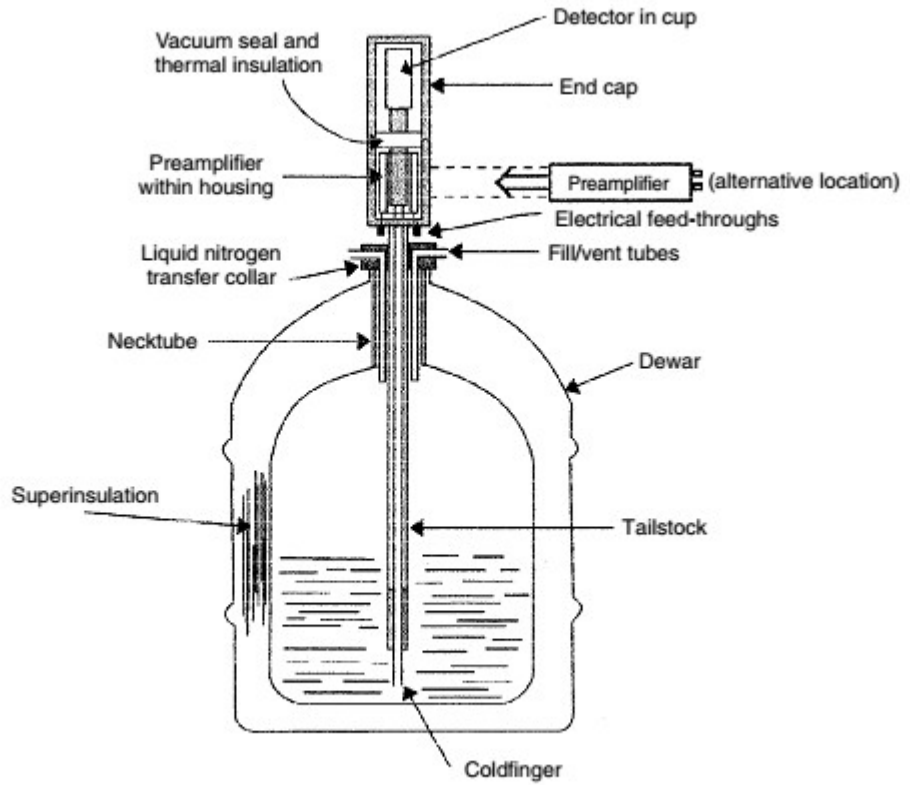


Figure 3.15 A typical germanium detector, cryostat and liquid nitrogen reservoir

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DETECTION SYSTEM

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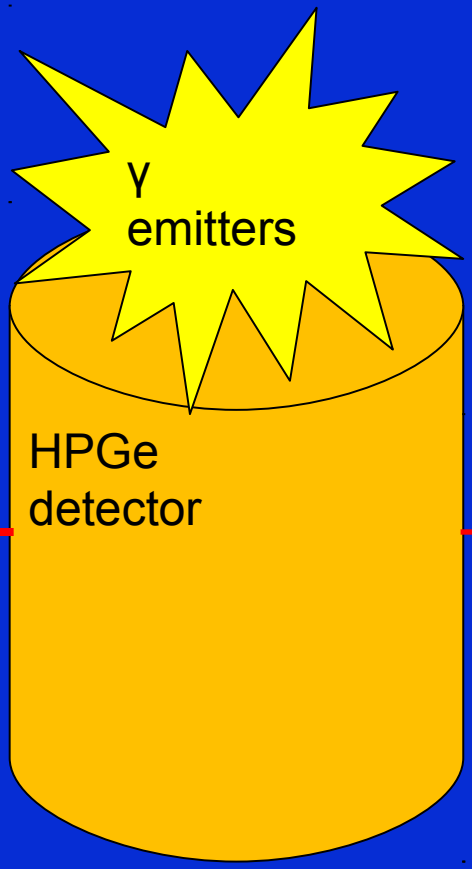


HV

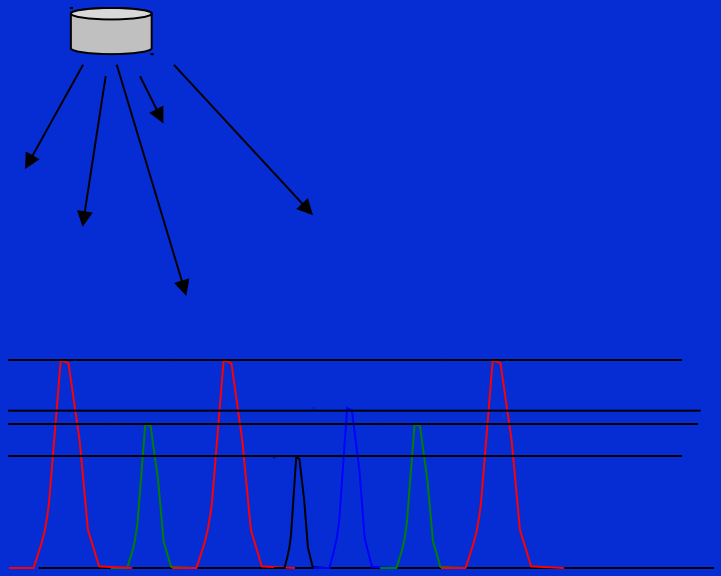
Pre Amp

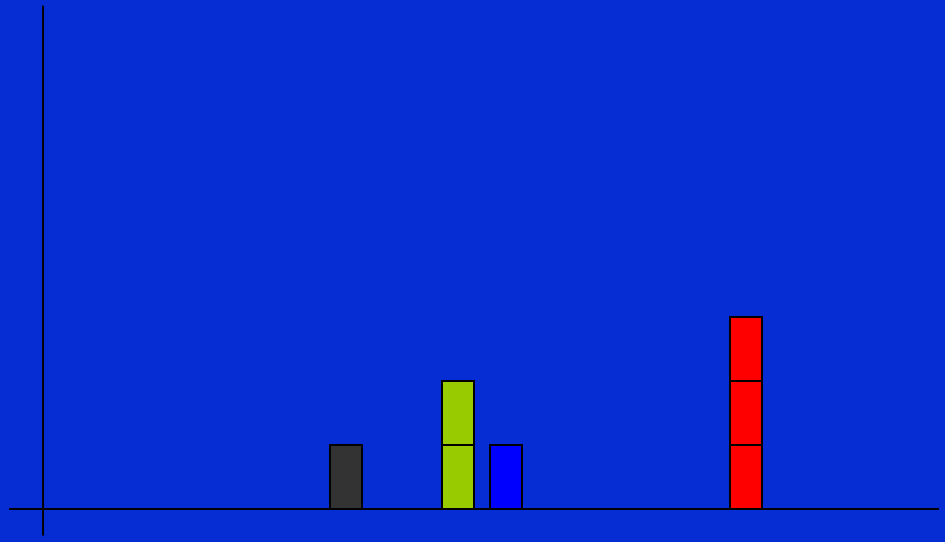
Amp

ADC (Analog-Digital-Converter)

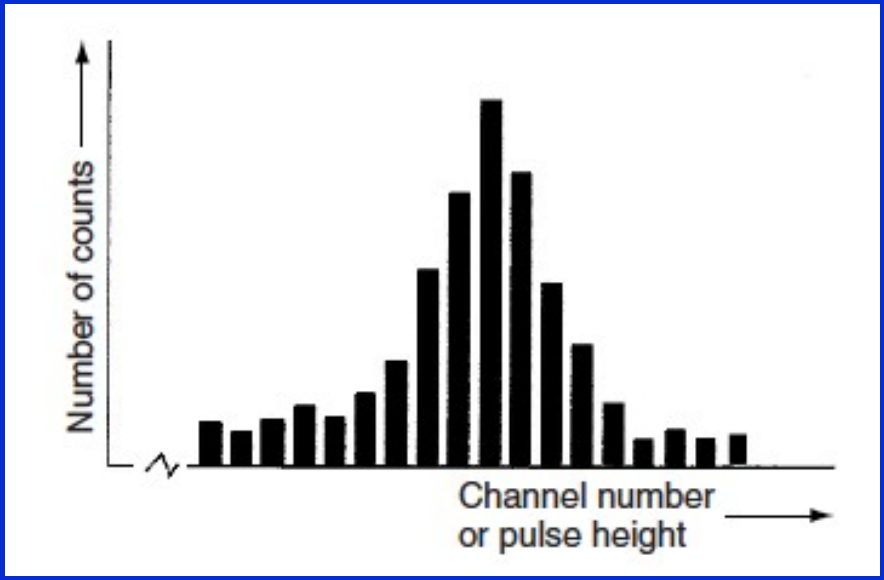
MCA (Multichannel analyzer)

- El detector genera una señal proporcional a la energía emitida por la fuente.
- El equipo acumula en cada canal el número de emisiones de una determinada energía.





MULTICHANNEL ANALYSER (MCA)



- from the output from the amplifier, it rejects out-of-range pulses;
- it measures the height of each of those accepted and adds a count into the memory location corresponding to the channel representing the voltage range;
- it displays the data as a spectrum and allows the data to be printed or saved to a data storage device.

➢ In principle, the relationship between pulse height (and therefore energy) and channel number would be exactly linear, passing through zero

MCA terms and definitions

- **Lower level discriminator (LLD)** – pulses below this level will not be analysed. Use this to reject electronic noise and low-energy X-rays.
- **Upper level discriminator (ULD)** – pulses above this level will not be analysed. Use this to reject very high energy pulses. This will often be left at its maximum, but still performs a useful function in rejecting high-energycosmic gamma-rays.
- **ADC zero level** – use this to adjust the energy calibration so that it passes through 0 keV. Not ideal for eliminating the effect of noise.
- **Digital offset** – this is a means of shifting the spectrum to lower channel numbers by subtracting a fixed number (the offset) from every channel number output by the ADC
- **Conversion range** –the maximum pulse height the MCA can accept, typically 10 V.
- **ADC resolution** is the total number of channels available within the ADC. It varies from model to model, but MCAs for germanium systems might incorporate a 16k (16 384), 8k (8192), or 4k (4096) channels ADC.
- **ADC conversion gain** is simply the number of channels actually used in a particular application – in everyday parlance, the spectrum size

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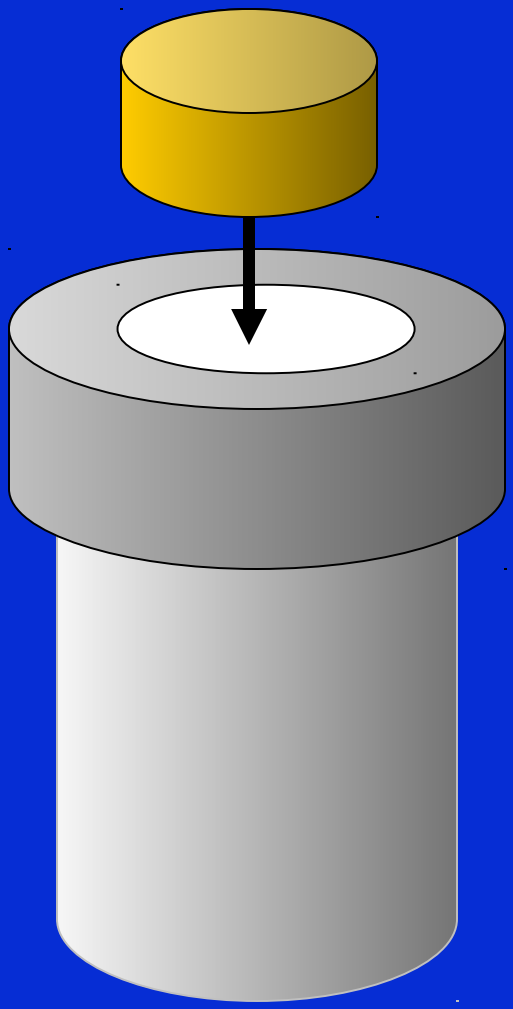
PROCESO DE MEDIDA:
GEOMETRÍAS, PATRONES Y CALIBRACIONES

- El patrón debe tener la misma geometría que la muestra a medir, la fuente radiactiva debe estar distribuida uniformemente en el patrón
- La posición de la muestra frente al detector deber ser fija, se precisan centradores.
- El programa determina la presencia de isótopos en función de la posición que ocupan en la pantalla del analizador (Energía), los desplazamientos del espectro, debidos a variaciones térmicas, pueden falsear los resultados. Es preciso realizar verificaciones periódicas.

Fuente de calibración

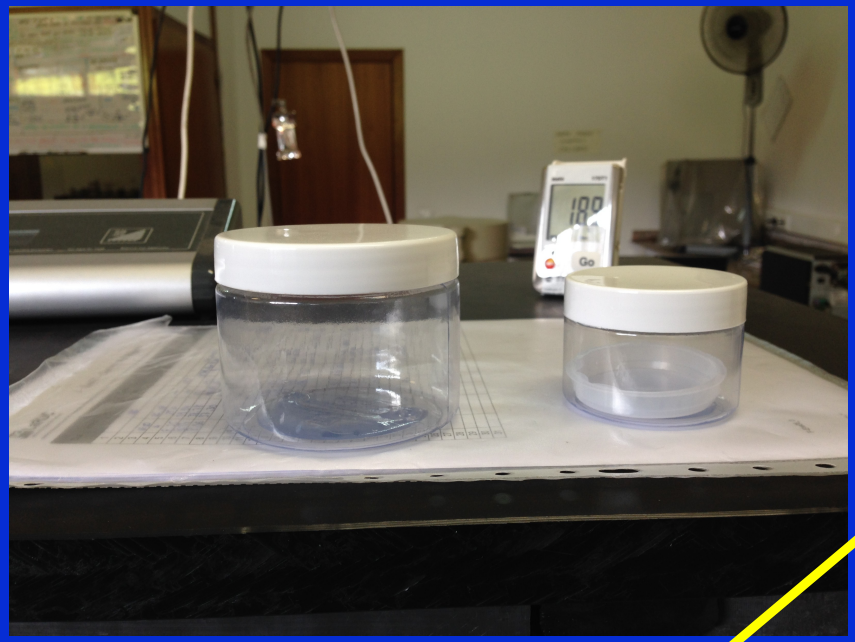


- Densidad similar a la muestra a medir (prensado de viruta de plomo).

- El patrón radiactivo debe estar distribuido uniformemente en todo el volumen. De ésta forma la autoabsorción será similar al caso de las muestras medidas en el laboratorio.

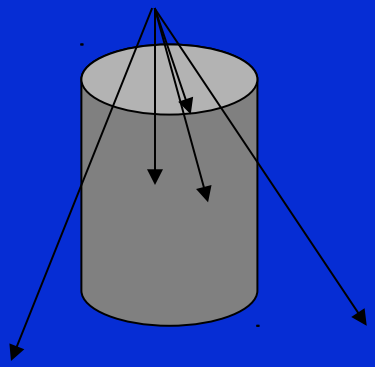
- Se mantiene la geometría mediante un centrador

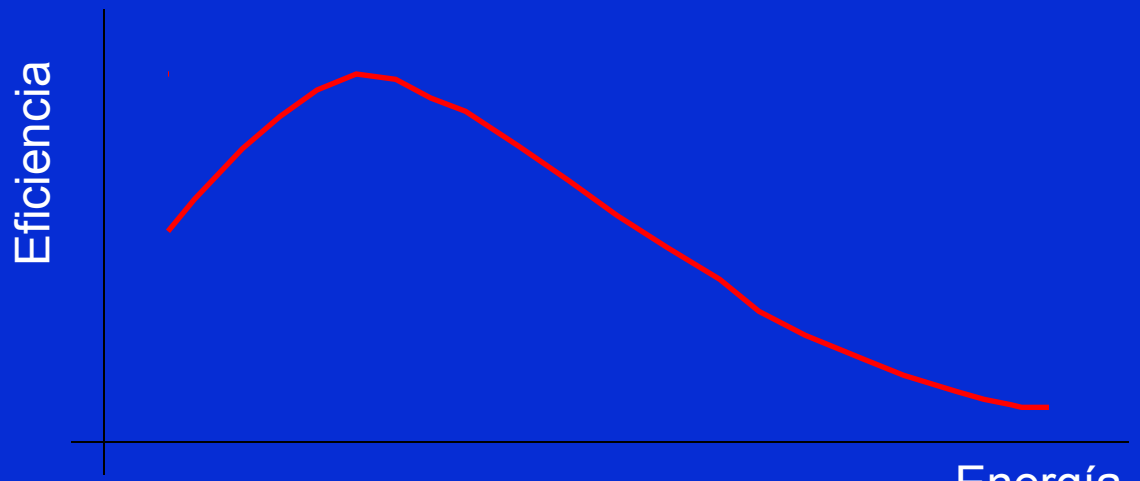
EJEMPLOS DE GEOMETRÍAS



Marinelli

- **La respuesta del detector no es lineal frente a la energía.**
- **Es preciso realizar una calibración Eficiencia/Energía**





DETECTION SYSTEM

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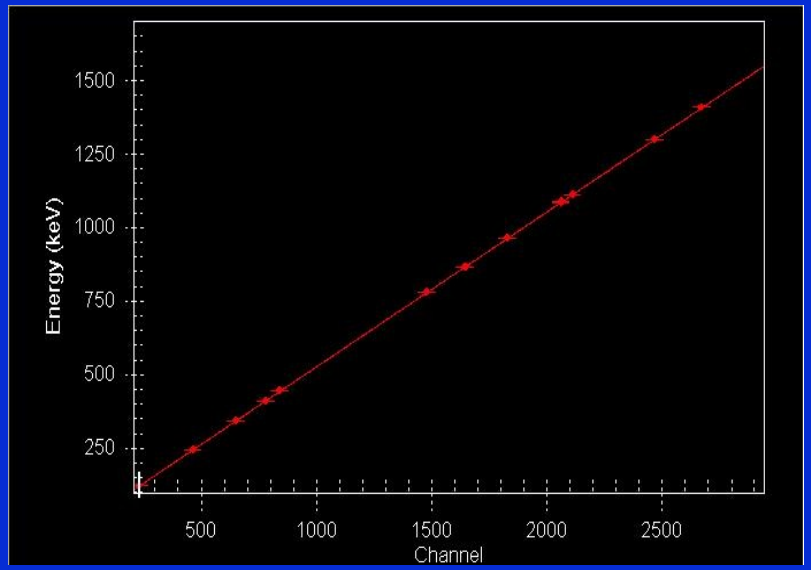
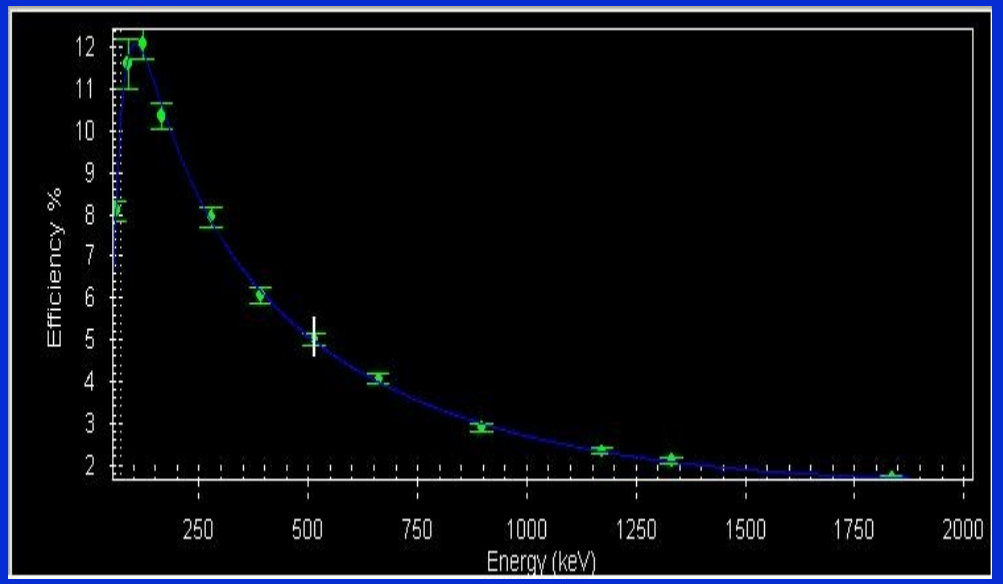
Calibration(standards)

Energy



Efficiency



Energy calibration

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The object of energy calibration is to derive a relationship between peak position in the spectrum and the corresponding gamma-ray energy

Energy calibration is accomplished by measuring the spectrum of a source emitting gamma-rays of precisely known energy and comparing the measured peak position with energy. It matters not whether the source contains a single nuclide or several nuclides: ^{152}Eu source for routine energy calibration

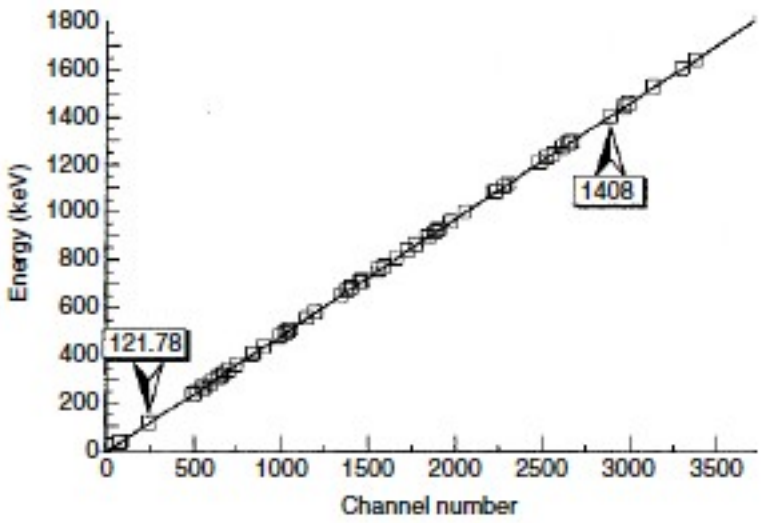
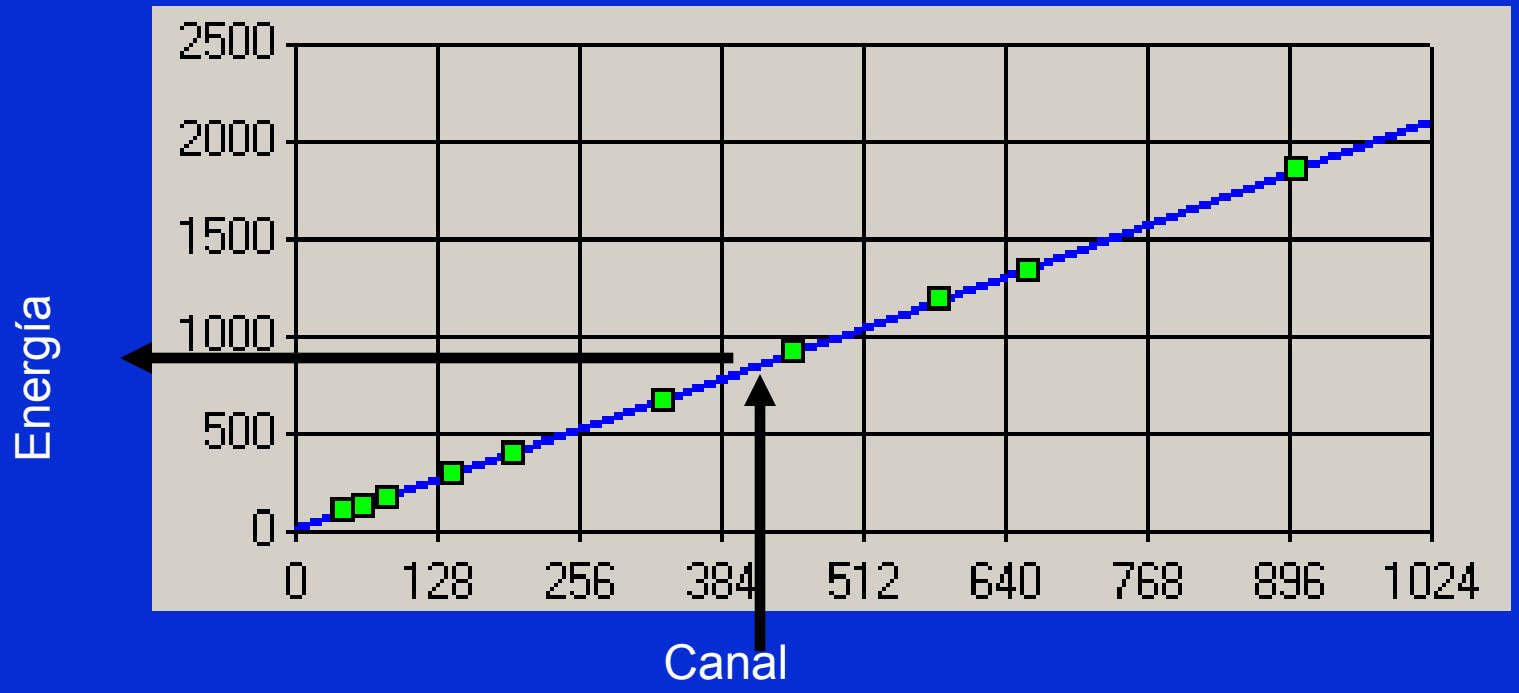


Figure 7.1 Energy calibration (55 points and the best fit straight line) using ^{152}Eu. The two marked points would be used for a two point calibration

Curva Energía / Canal



Energy - efficiency calibration

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- Relative efficiency is a general performance measure relating the efficiency of detection of the ^{60}Co gamma ray at 1332 keV of the detector to that of a standard sodium iodide scintillation detector.
- In gamma spectrometry, our intention is to relate the peak area in our spectrum to the amount of radioactivity it represents. For this, we need the absolute full energy peak efficiency. This relates the peak area, at a particular energy, to the number of gamma-rays emitted by the source and must depend upon the geometrical arrangement of source and detector.
- Absolute total efficiency relates the number of gamma rays emitted by the source to the number of counts detected anywhere in the spectrum. This takes into account the full energy peak and all incomplete absorptions represented by the Compton continuum.
- Intrinsic efficiency (full energy peak or total) relates the counts in the spectrum to the number of gamma rays incident on the detector. This efficiency is a basic parameter of the detector and is independent of the source/detector geometry.

$$\varepsilon = R/(S \times P_\gamma)$$

where R is the full-energy peak count rate in counts per second, S is the source strength in disintegrations per second (i.e. Becquerels) and P is the probability of emission of the particular gamma-ray being measured

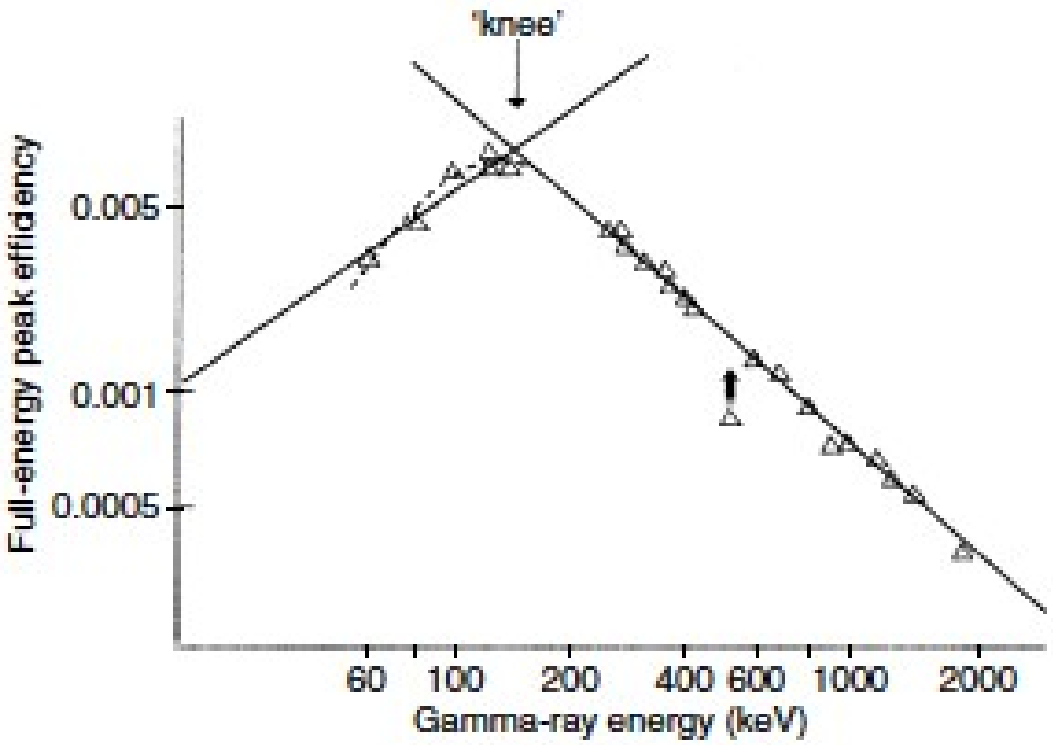
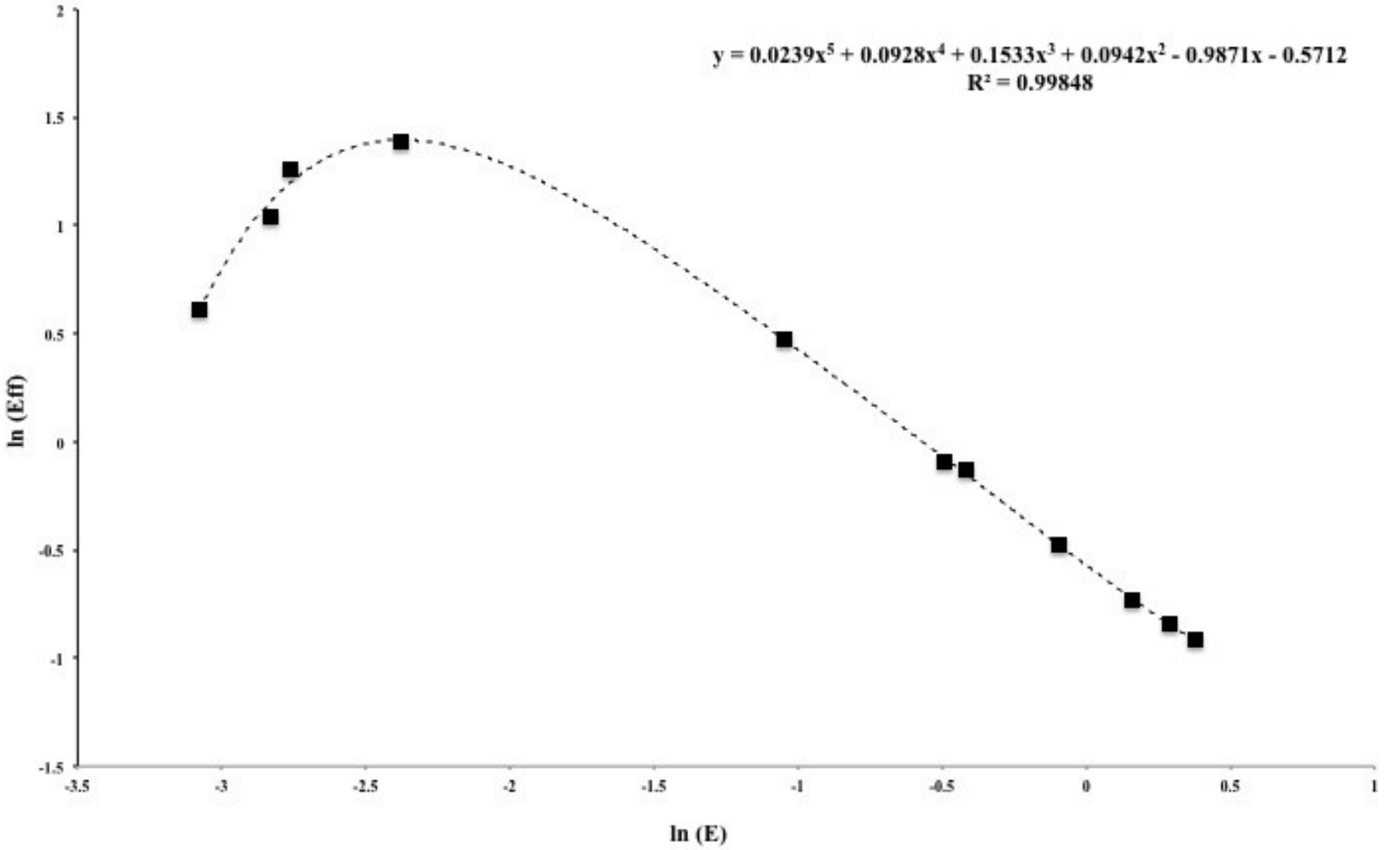
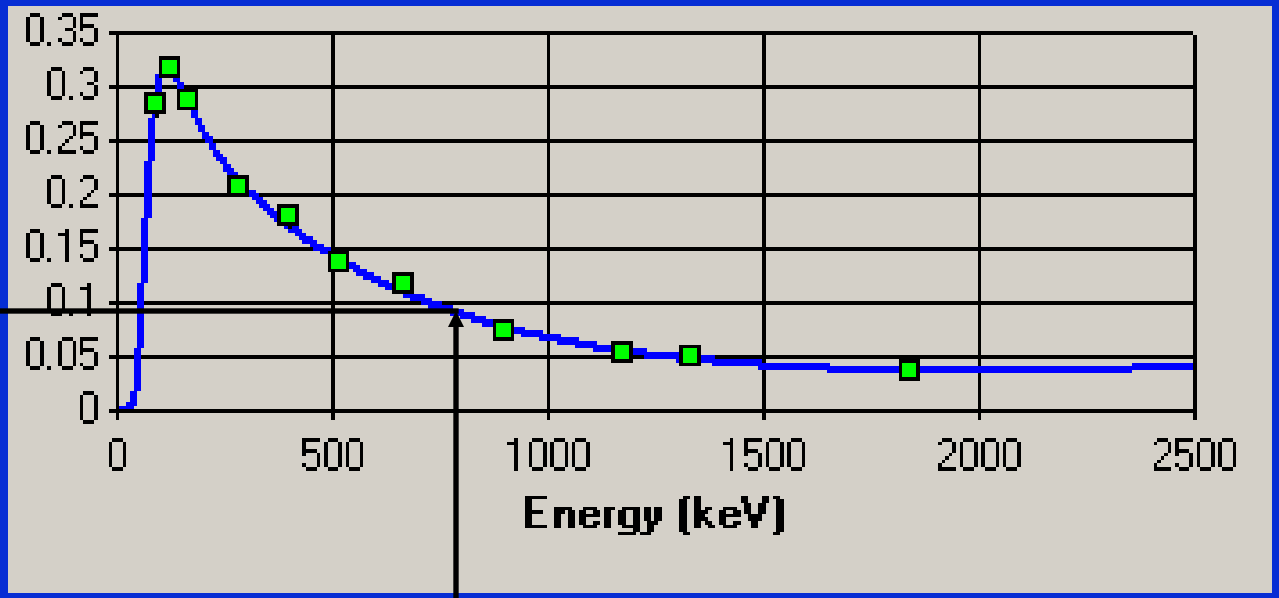


Figure 7.7 Efficiency curve for a p-type closed coaxial detector. The point lying below the line is that representing the 511 keV annihilation peak


y = 0.0239x5 + 0.0928x4 + 0.1533x3 + 0.0942x2 - 0.9871x - 0.5712
R² = 0.99848
ln (Eff)
ln (E)
2
1.5
1
0.5
0
-0.5
-1
-1.5
-3.5
-3
-2.5
-2
-1.5
-1
-0.5
0
0.5
1

Curva Eficiencia / Energía

Eficiencia del detector



Energía

ENERGY RESOLUTION

Resolution is a measure of the width of the peaks in a gamma-ray spectrum – the smaller the width, the better the detector, the higher the resolution.

FWHM: the Full Width of the peak at Half Maximum (keV)

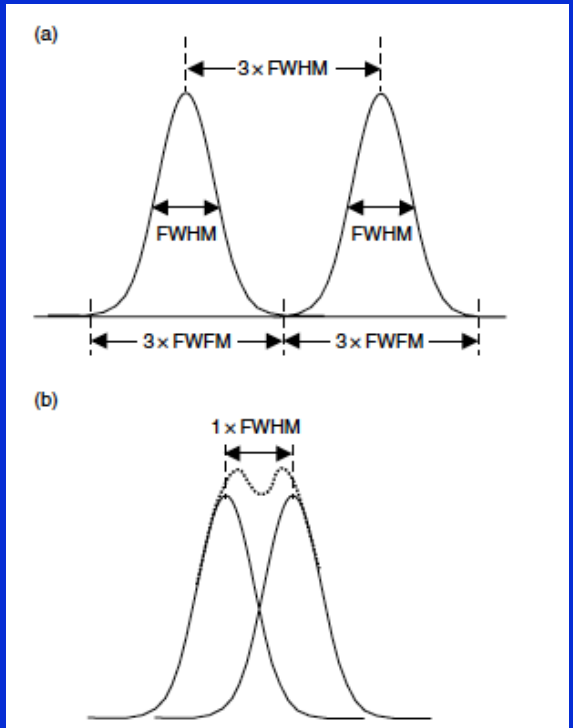


Figure 6.1 The influence of FWHM on the ease of discriminating between close energies: (a) centroids > 3 × FWHM apart should pose no problem; (b) centroids 1× FWHM apart require deconvolution programs

In practice, the measured shape of a gamma-ray peak in a germanium detector system is, to all intents and purposes, Gaussian

Table 6.3 Comparison of ultimate resolution of Ge and NaI(Tl) detectors

Material	ε(eV)[a]	F	FWHM at 661.67 keV	Resolution ratio	Source of data
Ge	2.96	0.058	0.794	0.031	Eberhardt (1970)
NaI(Tl)	170	1	25.0	0.031	See Knoll (1989), p. 312

[a] ε is the energy needed to create an electron–hole pair in germanium, or the energy needed to produce a photoelectron at the photocathode of the photomultiplier of the NaI(Tl) detector.

Si sólo se buscan determinados isótopos se puede realizar una calibración directa, en éste caso se mide la respuesta del detector frente a una fuente calibrada y se aplica posteriormente en la medida de la muestra

$$\text{Eficiencia} = \frac{\text{Área}}{\text{Actividad}}$$

DETECTION SYSTEM

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How to calculate activity

$$A\ (\mathrm{Bq/kg}) = \frac{I\ (\text{background corrected})}{T\ (\mathrm{s}) \cdot \varepsilon \cdot P \cdot M(\mathrm{kg})}$$

Let's remember some statistics …

The binomial distribution

In principle, the statistics of radioactive decay are binomial in nature. If we were to toss a handful of coins onto a table and then examine the arrangement, we would find coins in one of two dispositions – heads up or tails up. Similarly, if we could prepare a radioactive source and, during a particular period of time, monitor each individual atom we would see that each has only one of two possible fates – to decay or not decay

Confidence Limits

... we must quote our limits in such a way that we have a stated degree of confidence that the true value lies somewhere within them

Table 5.1 Coverage factors and the associated degree of confidence[a]

Coverage factor	Area within confidence limits (%)
1.0	68.3
1.645	90.0
1.96	95.0
2.0	95.5
2.326	98.0
2.576	99.0
3.0	99.9

[a] Confidence limit = coverage factor × s.

COUNTING DECISION LIMITS

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- Critical limit (LC) – a decision level: 'Is the net count significant?'
- Upper limit (LU) – 'Given that this count is not statistically significant, what is the maximum statistically reasonable count?'
- Detection limit (LD) – 'What is the minimum number of counts I can be confident of detecting?'
- Determination limit (LQ) – 'How many counts would I have to have to achieve a particular statistical uncertainty?'
- Minimum detectable activity (MDA) – 'What is the least amount of activity I can be confident of detecting?'

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5. Practical activity
6. Activity: “One minute paper”

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EJEMPLO: ACERÍAS

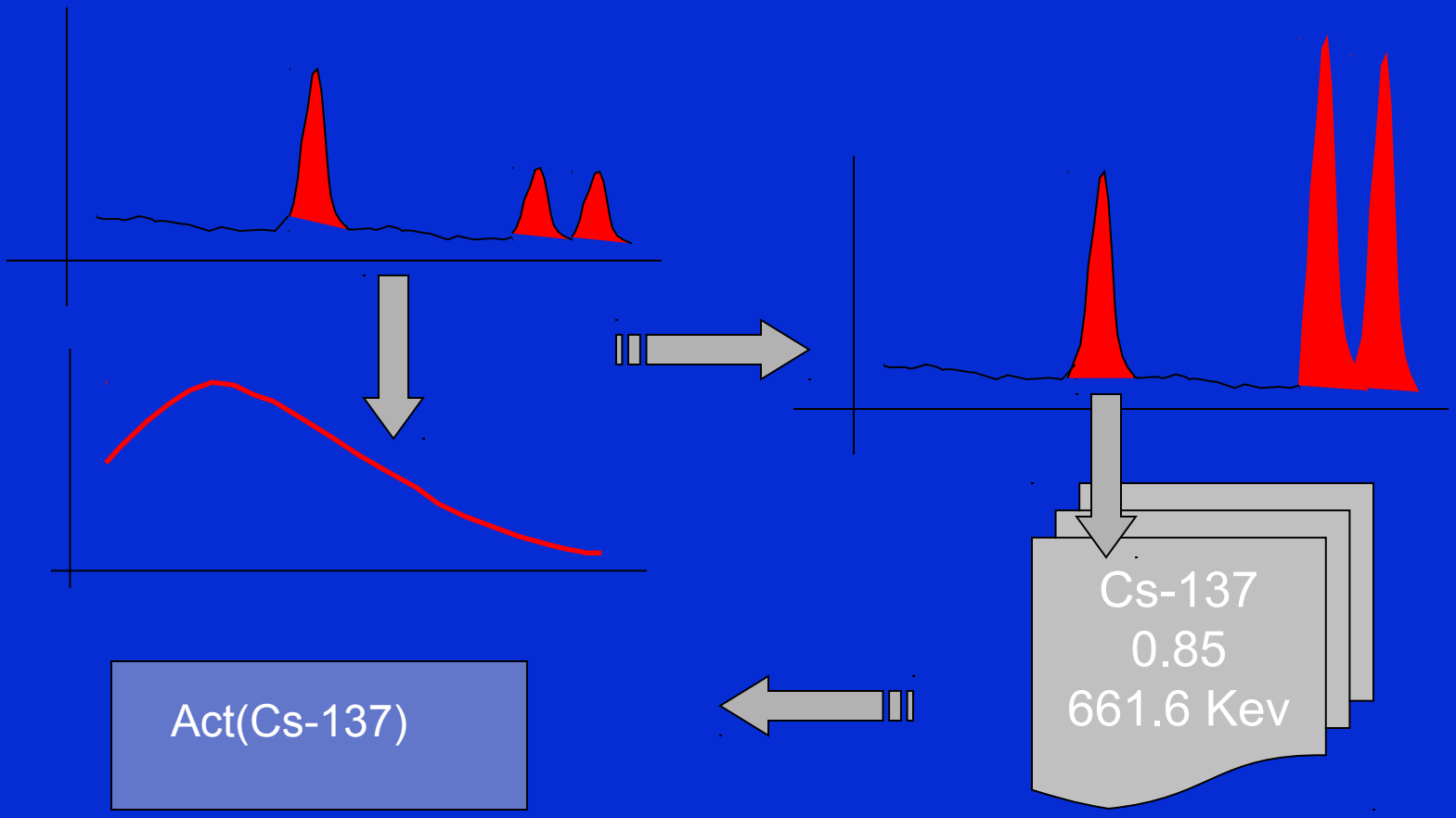
- En el caso más general se trabaja con todo el espectro

- El programa busca los picos, que corresponden a emisiones características de los diferentes isótopos de la muestra.

- Calcula el área de dichos picos y corrige el valor en función de la respuesta del detector.

- Determina el tipo de isótopo comparando la energía de los picos con una tabla de isótopos, librería, propia del material que se mide.

- Calcula la actividad del elemento en función del rendimiento gamma, cantidad de actividad que emite el isótopo en dicha energía.

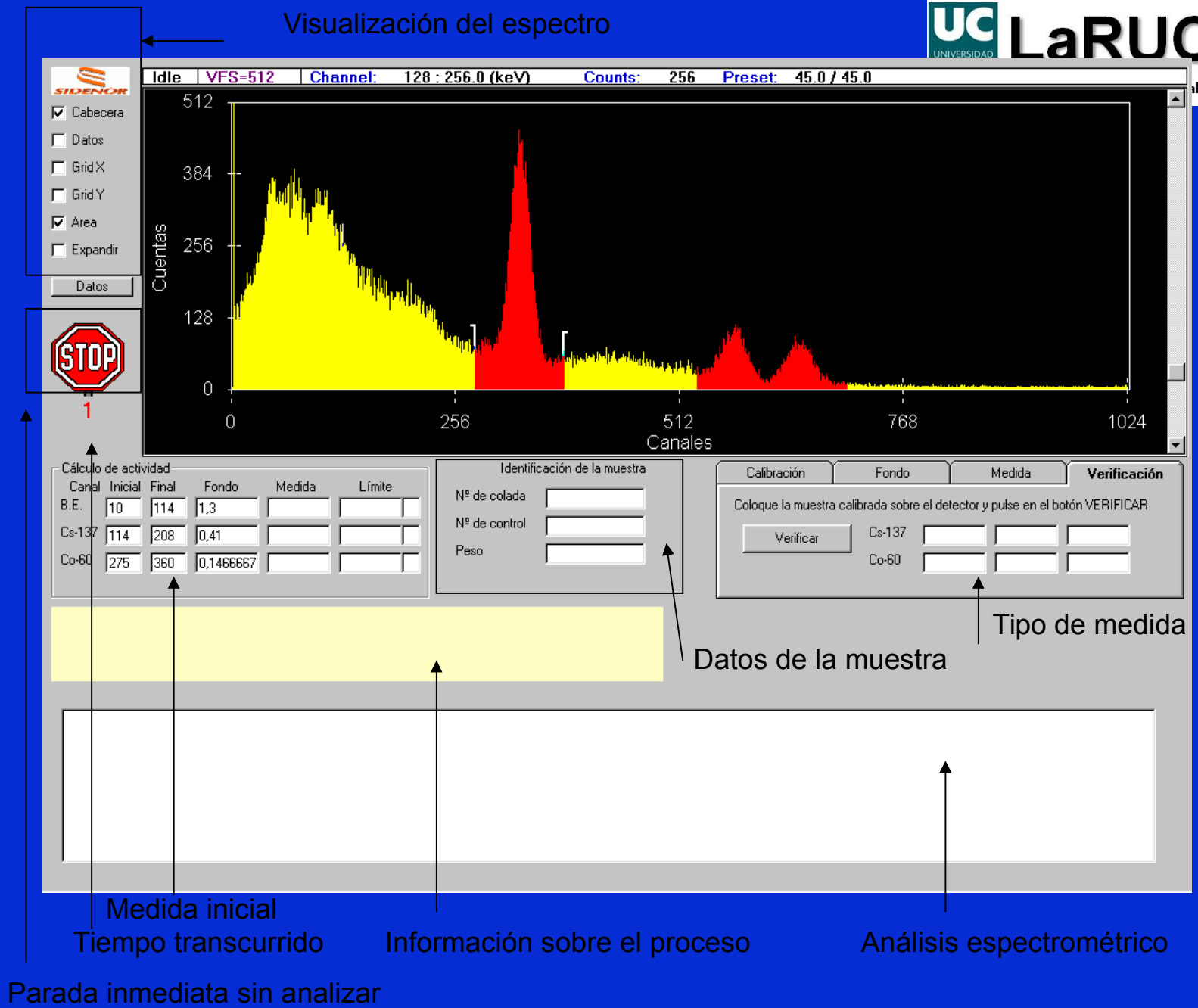

Cs-137
0.85
661.6 Kev
Act(Cs-137)

Análisis de picos

Peak No.		ROI start	ROI end	Peak centroid	Energy (keV)	FWHM (keV)	Net Peak Area	Net Area Uncert.	Continuum Counts
1		7-	19	15.89	34.60	5.23	1.51E+004	253.26	1.86E+004
2		21-	29	25.58	53.64	6.03	7.41E+002	338.36	5.24E+004
3	M	29-	374	42.94	87.76	10.70	2.17E+005	72.60	1.09E+005
4	m	29-	374	60.75	122.83	13.98	9.56E+004	36.08	1.20E+005
5	m	29-	374	82.92	166.55	17.46	1.43E+004	7.20	1.38E+005
.....									
15	m	535-	970	903.47	1842.51	74.77	6.12E+004	40.64	3.18E+004

Análisis de resultados

Nuclide Name	Id Confidence	Energy (keV)	Yield (%)	Activity (Bq /unit)	Activity Uncertainty
K-40	0.941	1460.81*	10.67	1.89081E+003	2.81162E+001
CO-57	0.820	122.06*	85.51	5.56557E+002	1.10762E+001
		136.48	10.60		
CO-60	0.984	1173.22*	100.00	2.68419E+003	2.03296E+001
		1332.49*	100.00	2.96881E+003	2.40518E+001
SN-113	0.915	255.12	1.93		
		391.69*	64.90	2.08184E+003	4.67840E+001
CS-137	0.990	661.65*	85.12	2.68463E+003	4.34523E+001


Visualización del espectro
Idle
VFS=512
Channel: 128 : 256.0 (keV)
Counts: 256
Preset: 45.0 / 45.0
Cabecera
Datos
Grid X
Grid Y
Area
Expandir
Datos
STOP
1
512
384
256
128
0
Cuentas
0
256
512
768
1024
Canales
Cálculo de actividad
Canal Inicial Final Fondo Medida Límite
B.E. 10 114 1,3
Cs-137 114 208 0,41
Co-60 275 360 0,1466667
Identificación de la muestra
Nº de colada
Nº de control
Peso
Calibración
Fondo
Medida
Verificación
Coloque la muestra calibrada sobre el detector y pulse en el botón VERIFICAR
Verificar
Cs-137
Co-60
Tipo de medida
Datos de la muestra
Medida inicial
Tiempo transcurrido
Información sobre el proceso
Análisis espectrométrico
Parada inmediata sin analizar

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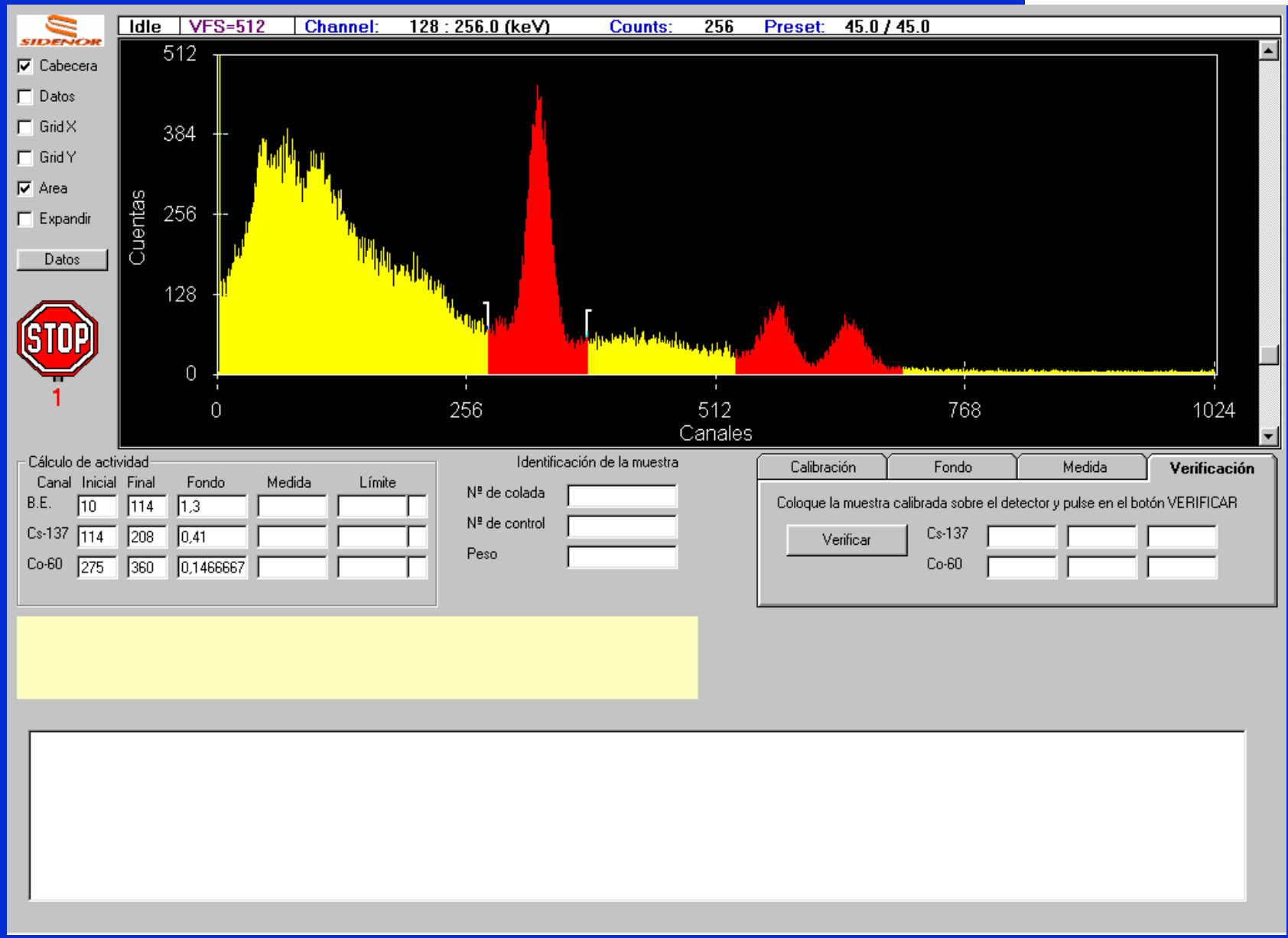
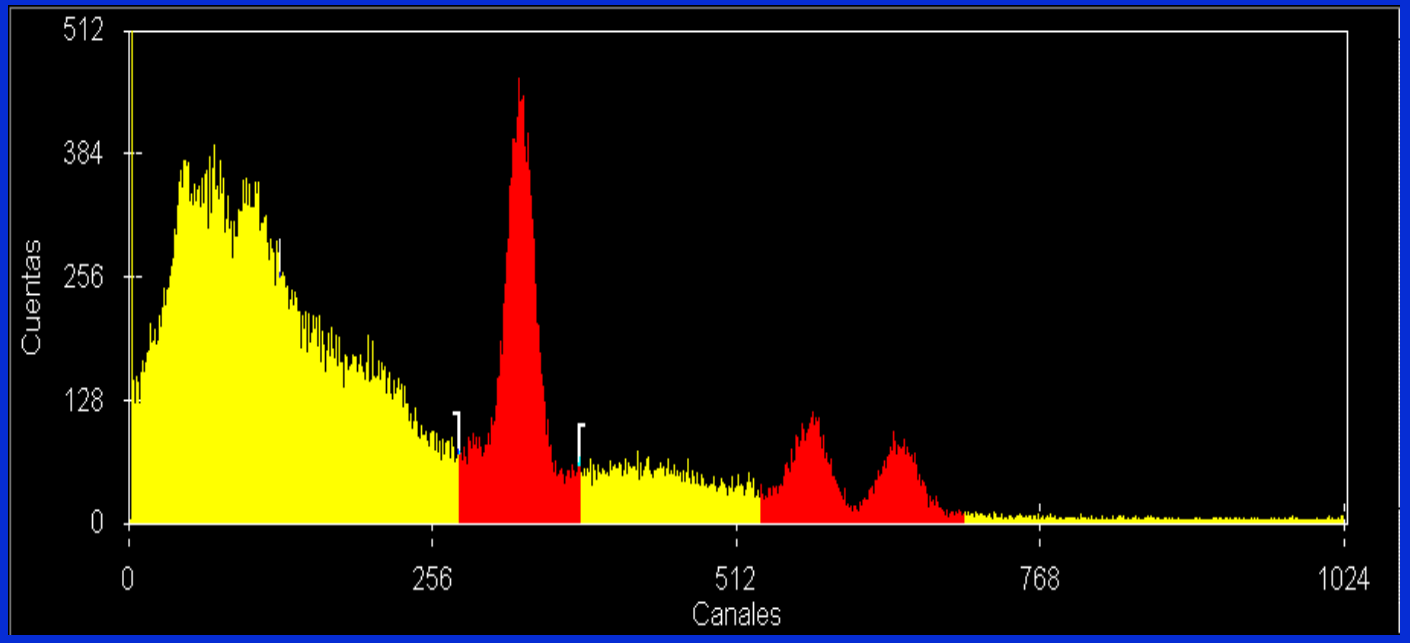
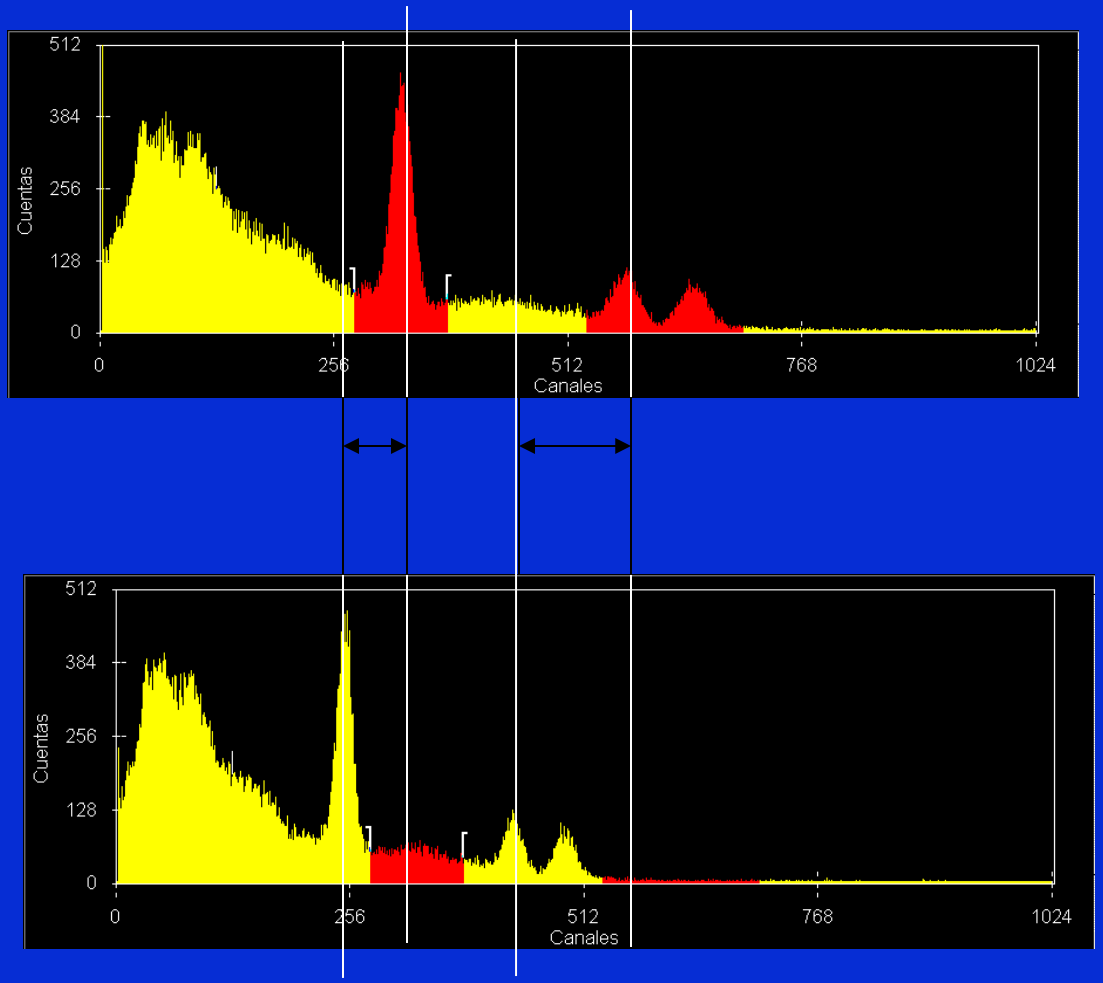


Verificación



El programa presenta en diferente color las zonas donde deben aparecer los picos de referencia. Si no aparecen en las bandas de color rojo es necesario recalibrar el espectro

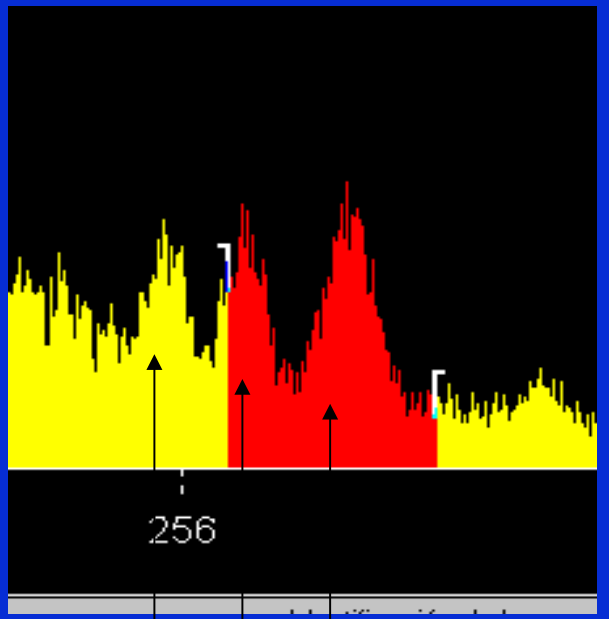
Verificación

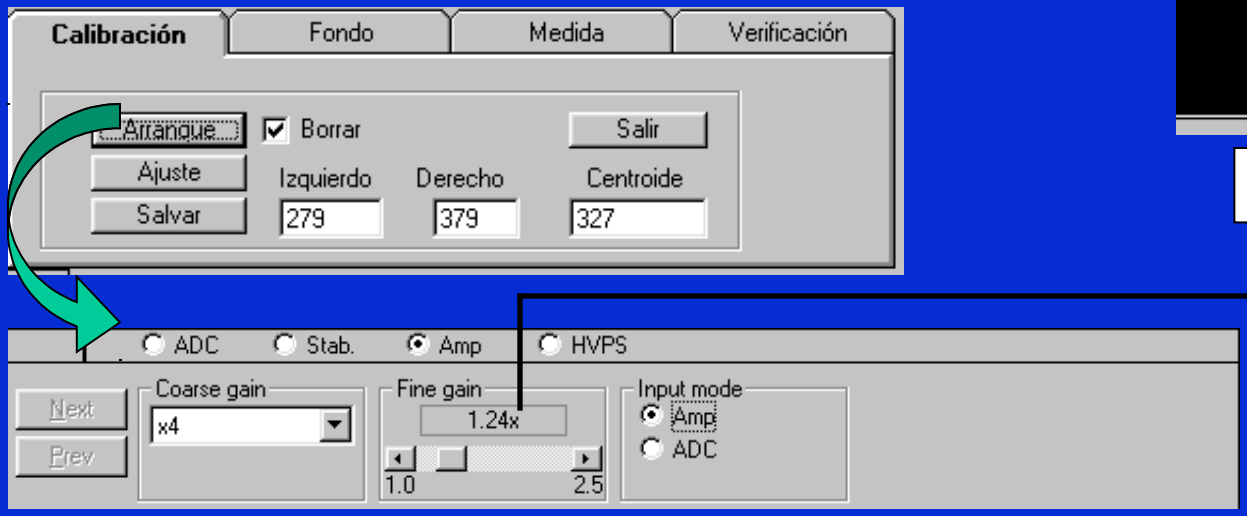


Básicamente debido a variaciones térmicas se producen desplazamientos de picos. La magnitud del desplazamiento es proporcional a la energía

Verificación

Para corregir el desplazamiento se varía la ganancia del amplificador, al aumentar éste valor se desplaza el pico hacia la zona de altas energías





Medida de fondo

Se emplea como valor de referencia para la primera fase de medida.

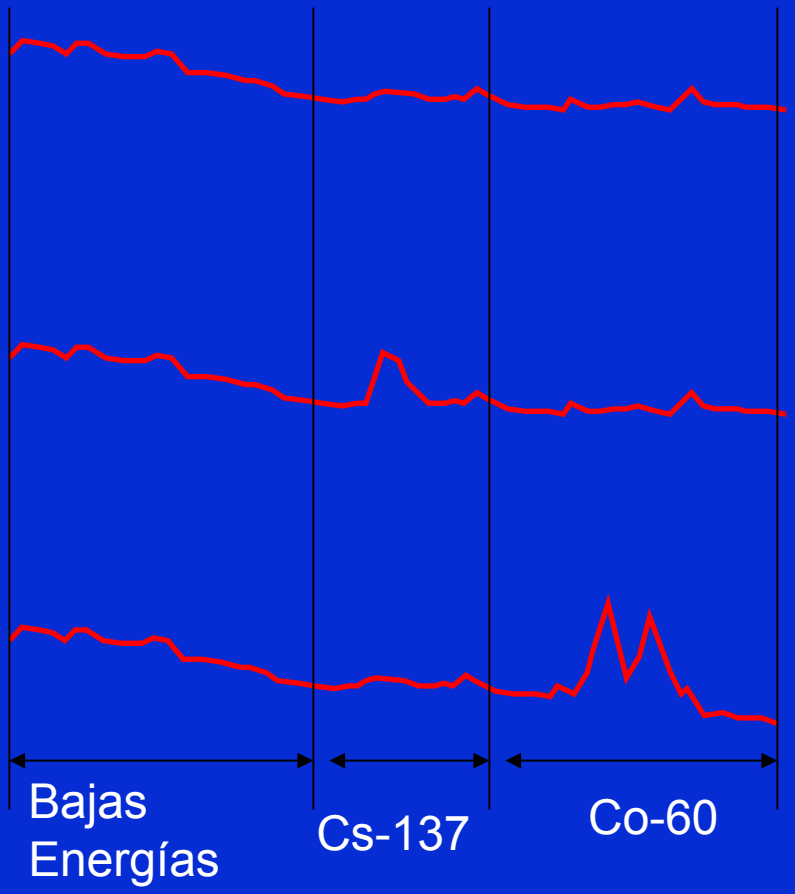
Se realiza de la misma forma que una medida normal.

El tiempo debe ser mayor que en empleado en una medida de muestras.

Proceso de medida

- Comparación con el fondo
  - Medida específica de fondo
- Análisis por espectrometría
  - Calibración, una por cada tipo de fuente
  - Edición de librerías en función de los isótopos a determinar
  - Verificación del sistema, la periodicidad dependerá de las condiciones ambientales del laboratorio.

Análisis inicial



Comparación con el fondo

Determina si el área en las zonas marcadas supera al fondo del equipo en un número determinado de desviaciones estándar.

Si se cumple ésta premisa se realiza una medida por espectrometría.

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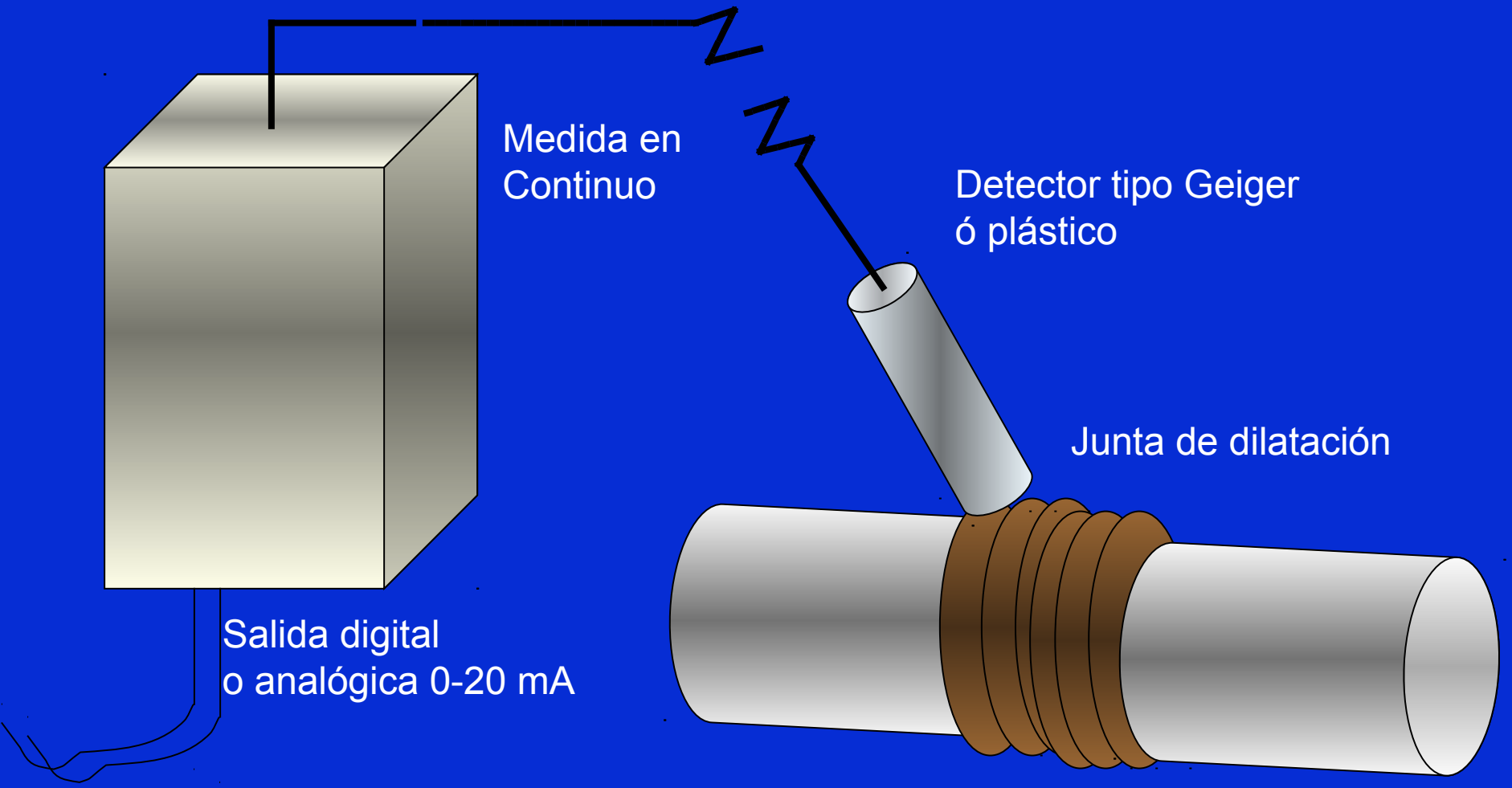
Medida por espectrometría

- Trabaja con librerías de isótopos específicas que pueden ser modificadas si se precisa.
- Necesita una curva de calibración de eficiencias que cubra el rango de medida.
- Puede identificar y cuantificar isótopos diferentes de los empleados en la calibración.
- Se puede definir la ventana de identificación de cada isótopo para evitar que se asigne la medida a un emisor con una energía próxima al especificado. Calcula límites de detección.

Medida específica de Cs-137



Medida específica de Cs-137

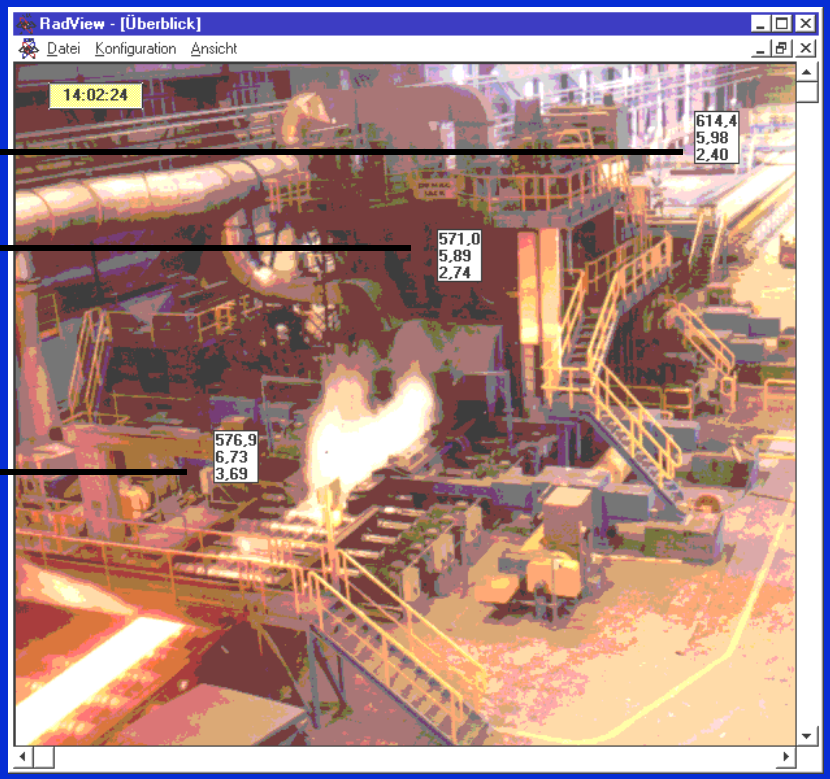
•El equipo controla continuamente La actividad medida en el detector.

• La programación se realiza desde un ordenador, los niveles de alarma, calibraciones, valores de alta tensión , tipo de comunicación,etc pueden ser modificados sencillamente.

• En el propio aparato se señalizan las alarmas.

• La medida se retransmite de forma automática a un ordenador remoto

Medida específica de Cs-137



Se puede programar una monitorización continua desde un ordenador para que controle todos los equipos y los niveles específicos de alarma de cada uno de ellos

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EJEMPLO: NATURALES

Radioactive equilibrium

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Growth of activity from decay of a parent

$$A_D = A_{P0}[\exp(-\lambda_P t) - \exp(-\lambda_D t)]\lambda_D/(\lambda_D - \lambda_P) + A_{D0}\exp(-\lambda_D t) \quad (1.12)$$

Transient equilibrium
$t_{1/2}$ parent > $t_{1/2}$ daughter

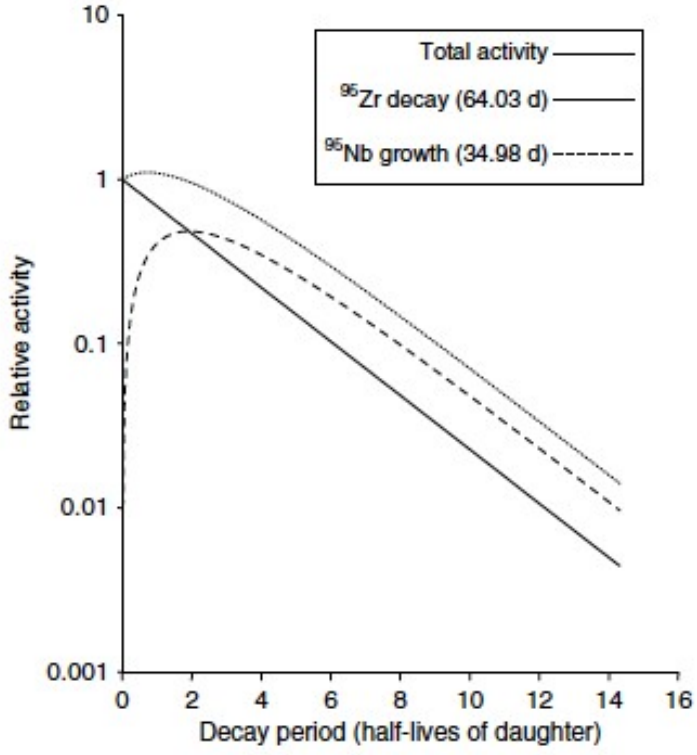

Figure 1.22 Transient equilibrium – relative activities of parent and daughter nuclides after separation

Secular equilibrium – $t_{1/2}$ parent >> $t_{1/2}$ daughter

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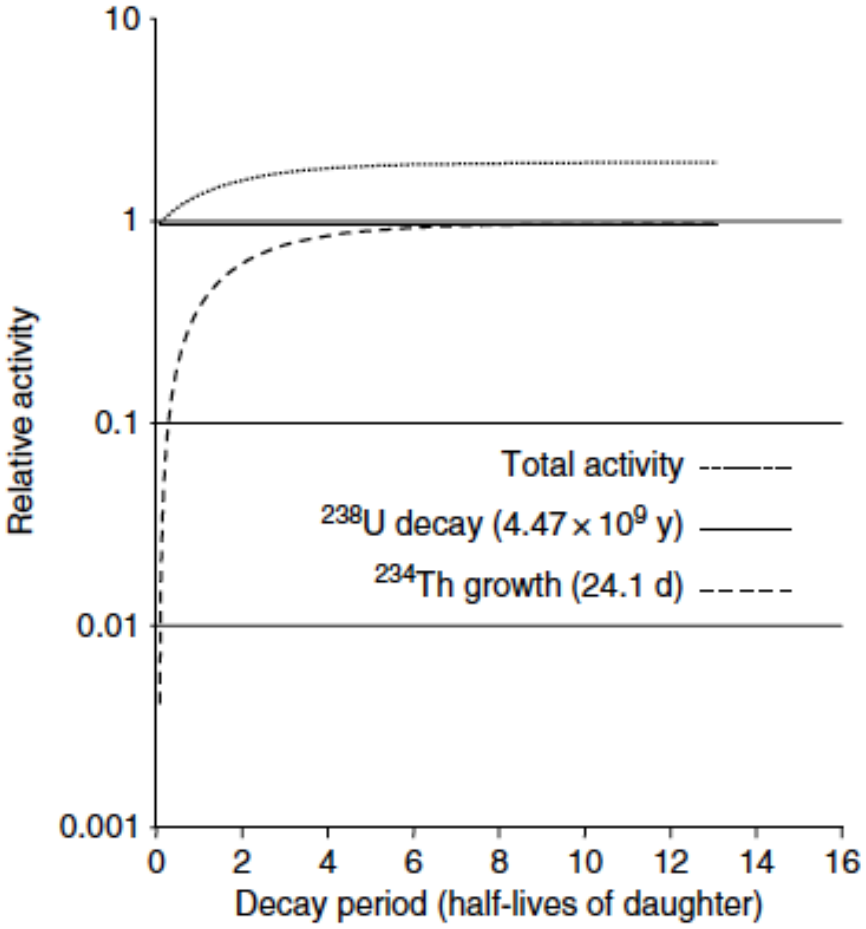

Figure 1.23 Secular equilibrium – relative activities of parent and daughter nuclides after separation where the parent half-life is much greater than that of the daughter

No equilibrium – $t_{1/2}$ parent < $t_{1/2}$ daughter

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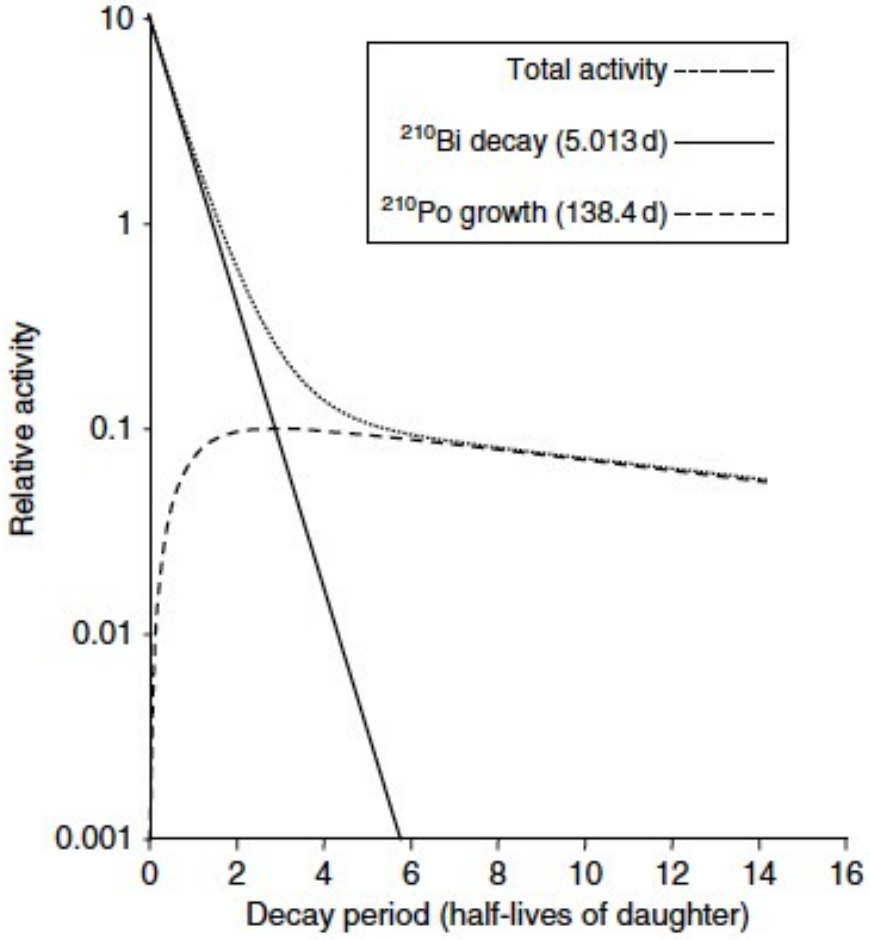

Figure 1.25 Relative activities of parent and daughter nuclides after separation where there is no equilibrium

Indicadores con pintura luminosa

Lentes o aleaciones de Th-232

More information ...

- http://www.unscear.org/ (United Nations Scientific Committee on the Effects of Atomic Radiation)
- http://www.nndc.bnl.gov/chart/ (Chart of nuclides)
- http://www.epa.gov/radon/pubs/citguide.html (A Citizen's Guide to Radon, EPA)
- Knoll, G. F. (1989). Radiation Detection and Measurement, 2nd Edn, John Wiley & Sons, Inc., New York, NY, USA.
- Currie, L.A. (1968). Limits for qualitative detection and quantitative determination, Anal. Chem., 40, 586–593.
- Currie, L.A. (2004). Detection and quantification limits: basic concepts, international harmonization and outstanding ('lowlevel') issues, Appl. Radiat. Isotopes, 61, 145–149.
- Practical Gamma-ray Spectrometry – 2nd Edition Gordon R. Gilmore © 2008 John Wiley & Sons, Ltd. ISBN: 978-0-470-86196-7

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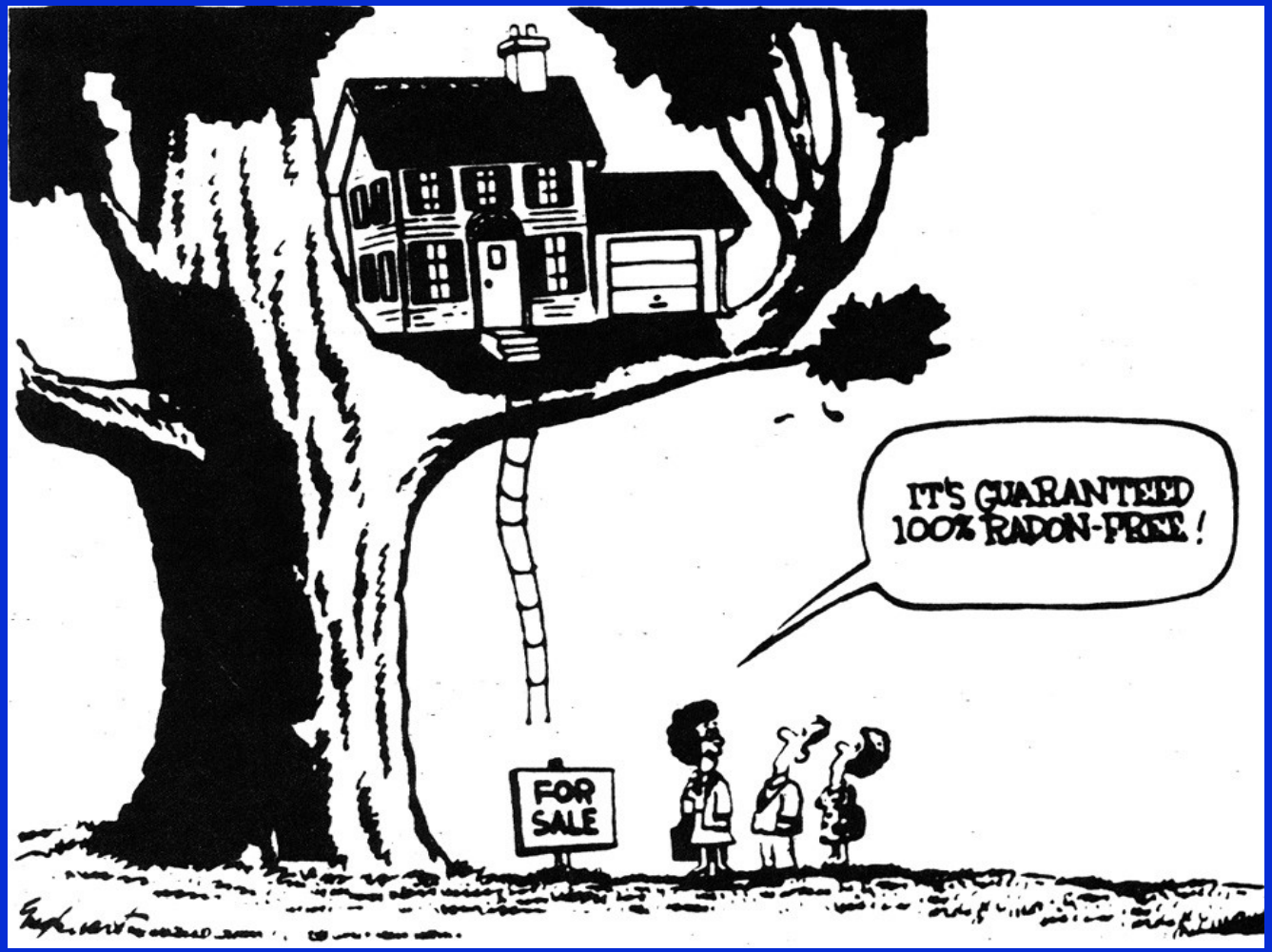
4. Examples: steel factories and natural radioactivity
5. **Practical activity**
6. Activity: “One minute paper”



Thank you very much !!!

Tack så mycket !!!

1. Introduction
2. Basic concepts
3. Radon Gas and Natural Radiation: risk assessment
4. Doses
5. Summary: Some key messages
6. **Activity: “One minute paper”**

1 minute paper ...