GAMMA-SPECTROMETRY

INTRODUCTION
General characteristics

Energy resolution

Peak area

Attenuation

Corrective factors
Gamma-ray spectrometry is a fast and widespread method

- used to identify radionuclides and
- to determine their activity.

- Non-destructive method
- Simple sample preparation
- But relative method (requires calibration using standards)

**Mature technique**

- Button-push method *(which software ?)*
- Hidden difficulties / uncertainties factors
- Evolution
  - Large detectors
  - Digital electronics
  - Compton-suppression
RESULTS DEPENDING ON EQUIPMENT CHARACTERISTICS:

**HPGe detector** (other detectors for low-energy X-rays (Si(Li), SDD), room temperature detectors NaI(Tl), CdTe, CZT…), etc.)

Sample (material, shape)

Measurement conditions
$^{152}\text{Eu} - \text{Point source at 10 cm from the detector window (100 cm}^3\text{) HPGe detector}$

Peaks (full-energy peaks):
- position = energy -> radionuclide identification;
- net area = activity

$$N_p(E) = NE(E) \cdot \varepsilon(E) = A \cdot I(E) \cdot t \cdot \varepsilon(E)$$

Other features (continuum): partial energy deposition, scattering, escape …
\[ N_P(E) = NE(E) \cdot \varepsilon(E) = A \cdot I(E) \cdot t \cdot \varepsilon(E) \]

or simply:
\[ n_i = A \cdot I_i \cdot \varepsilon_i \]

\( A \): activity (Bq)
\( n_i \): net peak area
\( I_i \): photon emission intensity
\( \varepsilon_i \): detection efficiency

-> 3 results depending on available input parameters:

\[ \varepsilon_i = \frac{n_i}{A \cdot I_i} \quad A = \frac{n_i}{I_i \cdot \varepsilon_i} \quad I_i = \frac{n_i}{A \cdot \varepsilon_i} \]

Hidden difficulties: peak area, efficiency for any energy (see “Fitting” section), corrective factors
RADIOACTIVE DECAY DATA

- Decay scheme: Difference emission/transition
- Gamma transition = gamma emission + internal conversion

\[ T_\gamma = I_\gamma + I_C \]

- Conversion coefficient: \[ \alpha_T = \frac{I_\gamma}{T_\gamma} \]

\[ I_\gamma = \frac{T_\gamma}{(1 + \alpha_T)} \]

- \( ^{51}\text{Cr} \): \( T_\gamma = 9.91(2) \); \( I_\gamma = 9.89(2) \)
  \( \alpha_T = 0.00181 \)

\[ I_\gamma = \frac{9.91}{1.00181} = 9.89 \]

- \( ^{103m}\text{Rh} \): \( T_\gamma = 100 \); \( I_\gamma = 0.069(4) \)
  \( \alpha_T = 1448 \)

\[ I_\gamma = \frac{100}{1449} = 6.9 \]
Example:

$^{103m}\text{Rh}$ X-ray emission intensities (short half-life)

Difficulty to derive « recommended » data requires carefully detailed information

In October 2005, the General Meeting of the *International Committee for Radionuclide Metrology* (ICRM) formally approved the recommendation made by the Nuclear Data Working Group of using the DDEP evaluated decay data in all future nuclear data studies.
Size -> Efficiency
P or N (low energies)

Photon interaction -> movement of electrons -> electron-hole pairs

\[ E_e = E - U_e \]

Fluorescence

Photoelectric absorption

\[ E' = E - E_e \]

Rayleigh scattering

Compton scattering

Pair production

\[ m_e c^2 \approx 511 \text{ keV}, \text{ electron rest energy} \]
Different shapes, types and sizes

Different shapes, types and sizes -> influence on detection efficiency

« Relative efficiency » compared to 3" X 3" NaI (TI)

For 1332 keV ($^{60}$Co at 25 cm)
(FEP efficiency = $1.2 \times 10^{-3}$)
Detector + electronic modules (analog/digital)

Preamplifier
- RC
- Reset

Amplifier
- Adapt the shape of the pulse – time constant versus count rate

Multi-channel Analyzer
- Number of channels: $10^3$ to $1.6 \times 10^4$

It is important to characterize the whole spectrometry chain (detector + electronics)
General characteristics

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Energy resolution:

$\Delta E \text{ (keV)} : \text{FWHM : Full Width at Half Maximum of a peak}$

- Depends on the energy
- Varies from $0.16 \text{ keV (@6 keV)}$ to $2.5 \text{ keV (@1,3 MeV)}$, depending on the detector

Resolving power: $\Delta E / E \text{ (%) : Some } 10^{-3}$

(5 – 10 % for NaI(Tl))

Characteristic of the spectrometer quality:

- separation of closely spaced peaks
- detection limit
Initial photon emission: Monoenergetic line (source) ($\Delta E = \text{some meV}$)

Widening and distortion of the resulting peak (spectrum)

- **Widening**: Gaussian effects:
  - fluctuation of the number of charge carriers: $\Delta E_s$
  - electronic noise: $\Delta E_E$

- **Non Gaussian effects**:
  - Charges collection: $\Delta E_C$
  - Drift of the operating system (detector + electronics)
  - Pile-up

Result: in the resulting spectrum: 'peak' with finite width (keV) and more or less symmetrical shape

First approximation: Gaussian shape
ENERGY RESOLUTION

GAUSSIAN DISTRIBUTION

Distribution centered on energy $E_0$ with standard deviation, $\sigma$

Characteristics:

Full width at half maximum of the Gaussian

FWHM ($\Delta E$) = 2,355 $\sigma$

Area: $S = (2\pi)^{1/2} \sigma A$

$$G(E) = A \cdot \exp\left(\frac{(E - E_0)^2}{2\sigma^2}\right)$$
STATISTICAL NOISE : $\Delta E_S$

Cause: fluctuations of the number of charge carriers $n$

$E$: incident energy (some keV)

$w$: mean pair creation energy (some eV)

$n = \frac{E}{w}$

Hypothesis: Poisson statistics: $\sigma_n = \sqrt{n}$

Standard deviation of the number of charge carriers

$\rightarrow$ Standard deviation of the deposited energy ($n\ w$)

$\sigma_{Es} = w \cdot \sigma_n = w \cdot \sqrt{n} = w \cdot \sqrt{\frac{E}{w}} = \sqrt{w \cdot E}$
Example: (at 77K: \( w = 2.96 \) eV in Ge
\( w = 3.76 \) eV in Si)

\[ E = 1 \text{ MeV} \rightarrow \text{in Ge: } n = 3.3 \times 10^5 \]

\[ \sigma_{ES} = \sqrt{w \cdot E} = \sqrt{2.96 \cdot 10^6} = 1.72 \text{ keV} \]

\[ \Delta_{ES} = 2.355 \cdot \sigma_{ES} = 4.05 \text{ keV} \]

The observed width is much lower (\( < 2 \) keV)

\[ \rightarrow F = \text{Fano factor: } F = \frac{\text{Observed variance}}{\text{Poisson predicted variance}} \]
\[ \Delta_{Es} = 2.355 \cdot \sqrt{F \cdot \sigma_{Es}^2} = 2.355 \cdot \sqrt{F \cdot w \cdot E} \]

Fano factor < 1 (charges creation are not independent: correlations)

Measured for Ge and Si: value depending on the quality of the crystal?

Experimental values: (increasing versus time …)

Ge: 0.07 to 0.12

Si: 0.08 to 0.12
Preamplifier – amplifier

- Independent on the energy

- Can be determined using a pulser: input at the preamplifier « test »
CHARGE COLLECTION: $\Delta E_c$

Causes: trapping of charge carriers and ballistic deficit

- Trapping (impurities or crystal imperfections): loss of charge or slowing of the rate of charge collection

- Ballistic deficit (electron-hole mobility (15% difference in Ge))

Depends on the quality of the crystal and on the electric field (position of the interaction in the detector)

Consequence of the loss of charge: Low-energy tailing

Improvement:

- Increasing the voltage (recommended value by supplier)
- Rejecting pulses with slow rise-time
- Using collimator to avoid interaction in low electric field regions (front corners)
\[
\Delta E^2 = \Delta E_S^2(E) + \Delta E_C^2 + \Delta E_E^2
\]

\(\Delta E_S\) = statistical noise (dependent on the energy)
\(\Delta E_C\) = charge collection
\(\Delta E_E\) = electronical noise

First approximation:

\[
\Delta E^2 = K + \Delta E_S^2(E)
\]

\[
\Delta E^2 = K + 2.355^2 (F \cdot w \cdot E)
\]
\[ \Delta E^2 = K + 2.355^2 (F \cdot w \cdot E) \]

Linear fitting: 

FWHM\(^2 = 0.00182E + 0.4141 \]

K = 0.414 keV (electronics + charge collection)

\((2.355)^2 F \cdot w = 0.00182 \implies F = 0.110\)
FWTM (Full Width at Tenth Maximum)

FWFM (Full Width at Fiftieth Maximum)

These values can be compared to those characteristic of a Gaussian (ideal?) peak:

\[
\frac{\text{FWTM}}{\text{FWHM}} = 1.82 \\
\frac{\text{FWFM}}{\text{FWHM}} = 2.38
\]

Information about the peak shape, particularly on its base (collection defects)
General characteristics

Energy resolution

Peak area

Attenuation

Corrective factors
For quantitative analysis: determination of $N(E) = \text{peak area}$

(number of photons with energy $E$ that deposited all their energy in the detector)

Spectrum -> "Regions Of Interest" (ROI’s) containing one or several peaks
Automatic peak search
Peak region:
\[ Y(E) = G(E) + (aE + b) \]
(Gaussian + linear background)

Convolution by a « top-hat » filter
With zero area

Width = Gaussian width
(requires resolution calibration)

\[ Y^C = Y \otimes TF \]
Presence of a peak when \( Y^C > T \) (threshold)

Peak FWHM <-> channels where \( Y^C = 0 \)

-> definition of a ROI

Peak detection sensitivity is dependent on the threshold value:

T too low -> spurious peaks
T too high -> missing peaks
**DERIVATIVES**

First derivative: change of sign at the maximum peak position.

Second derivative: negative minimum at the position peak.

![Graph showing derivatives and automatic peak search](image-url)
Result of automatic peak search

Expert visual checking recommended!
Processing of a single peak ROI

- Peak superimposed on a continuous background
Background

Peak with energy $E_0$ in region $[f,l]$ first channel $= f$ ; last channel $= l$

- **Step**: 
  
  $F(i) = 1$ for $E < E_0$
  
  $F(i) = 0$ for $E > E_0$

- **Linear**: 
  
  $F(i) = 1 - \frac{(i - f)}{(l - f)}$

- **Galton curve**: 
  
  $F(i) = 1 - \frac{\sum_{f}^{i} Y_i}{\sum_{f}^{l} Y_i}$
## Background

<table>
<thead>
<tr>
<th>Background type</th>
<th>Gross area</th>
<th>Net area</th>
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<tbody>
<tr>
<td>Linear (33 channels)</td>
<td>943188</td>
<td>938907</td>
</tr>
<tr>
<td>Linear (25 channels)</td>
<td>942037</td>
<td>938191</td>
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<td>Linear (18 channels)</td>
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<td>Step (33 channels)</td>
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<tr>
<td>Galton (33 channels)</td>
<td>943188</td>
<td>938577</td>
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</table>
General characteristics

Energy resolution

Peak area

Attenuation

Corrective factors
Use of screen to reduce counting rate (X-rays, Bremsstrahlung)

Beer-Lambert law

Mass attenuation coefficients
Attenuation of a narrow parallel photon beam

\[ l(x) = l_0 \cdot e^{-\mu x} = l_0 \cdot e^{-\frac{\mu}{\rho} \rho x} \]

\( \rho \) = density (\( g.cm^{-3} \))
\( \mu \) = linear attenuation coefficient of material \( i \) for energy \( E \) (\( cm^{-1} \))
\( \rho x \) = mass thickness (\( g.cm^{-2} \))
\( \mu / \rho \) = mass attenuation coefficient (\( cm^2.g^{-1} \))

\( \mu \) depends on \( E \) and \( Z \)
Practical parameter: Attenuation (absorption + scattering) coefficient

Partial interaction coefficients depend on the energy, \( E \), and the material (\( Z \)):

- **Photoelectric absorption**: \( \tau_i(E) \)
  \[ \tau \approx \text{const} \cdot Z^{4.5} \cdot E^{-3} \] (dominant at low energies)

- **Compton scattering**: \( \sigma_i(E) \)
  \[ \sigma \approx \text{const} \cdot Z \cdot E^{-1} \]

- **Pair production effect**: \( \kappa_i(E) \)
  \[ \kappa \approx \text{const} \cdot Z^2 \] (only if \( E > 1022 \text{ keV} \))

For practical use: tables function of \( Z \) and \( E \)

Tables: cross sections (1 barn = \( 10^{-24} \text{ cm}^2 \)) or mass attenuation (\( \text{cm}^2 \cdot \text{g}^{-1} \))

<table>
<thead>
<tr>
<th>Interaction</th>
<th>Linear attenuation coefficient (( \text{cm}^{-1} ))</th>
<th>Mass attenuation coefficient (( \text{cm}^2 \cdot \text{g}^{-1} ))</th>
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<tbody>
<tr>
<td>Photoelectric</td>
<td>( \tau )</td>
<td>( \tau / \rho )</td>
</tr>
<tr>
<td>Compton</td>
<td>( \sigma )</td>
<td>( \sigma / \rho )</td>
</tr>
<tr>
<td>Pair production</td>
<td>( \kappa )</td>
<td>( \kappa / \rho )</td>
</tr>
<tr>
<td>Total</td>
<td>( \mu = \tau + \sigma + \kappa )</td>
<td>( \mu / \rho = \tau / \rho + \sigma / \rho + \kappa / \rho )</td>
</tr>
</tbody>
</table>
Photoelectric absorption coefficient = sum of photoelectric effect in each electronic shell (subshells):

\[ \tau = \tau_K + (\tau_{L1} + \tau_{L2} + \tau_{L3}) + (\tau_{M1} + \tau_{M2} + \tau_{M3} + \tau_{M4} + \tau_{M5}) + \ldots \]

If \( E < \) binding energy of shell \( i \), \( \tau_i = 0 \)

For \( E = E_i \) : absorption discontinuity: maximum ionisation probability in shell \( i \)

\( \tau \) variation versus the energy shows discontinuities corresponding to binding energies of electrons shells and subshells K, L, M...

Since \( \mu = \tau + \sigma + \kappa \)

\( \mu \) has the same discontinuities, function of the material atomic structure (Z)
Germanium mass attenuation coefficient

Ge binding energies:
- L1: 1.4143
- L2: 1.2478
- L3: 1.2167
- K: 11.1031
Lead mass attenuation coefficient

Pb binding energies:
- M1: 3.8507
- M2: 3.5542
- M3: 3.0664
- M4: 2.5856
- M5: 2.4840
- L1: 15.8608
- L2: 15.2000
- L3: 13.0352
- K: 88.0045
Composition known -> calculation
Composition unknown -> measurement

Calculation: Attenuation coefficients table

- XCOM (NIST Database)
- Example for HCl 1N
Defining the mass fraction of each compound for HCl 1N:

Matrix : HCl 1N = 1 mole of HCl in 1 liter of solution

HCl 1N density = 1.016 (1L = 1016 g)

Mass of one HCl mole = 1 + 35.45 = 36.45 g

Resulting input parameters for XCOM

Compound 1: HCl
Mass fraction: 36.45

Compound 2: H₂O
Mass fraction = 1016 – 36.45 = 979.55
Attenuation coefficients

Attenuation coefficients

XCOM results
Principle: Use the unknown matrix and a collimated photon beam

Two successive measurements
- Empty container
- Container filled with unknown matrix with thickness x
For each energy:

\[ N_2(E) = N_1(E) \cdot \exp (-\mu(E) \cdot x) \]

Thus:

\[ \mu(E) = \frac{1}{x} \ln \left( \frac{N_2(E)}{N_1(E)} \right) \]

Associated relative uncertainty:

\[
\frac{u^2(\mu)}{\mu^2} = \frac{u^2(x)}{x^2} + \frac{1}{\ln^2 \left( \frac{N_0(E)}{N(E)} \right)} \left( \frac{u^2(N_0(E))}{N_0^2(E)} + \frac{u^2(N(E))}{N^2(E)} \right)
\]
Dedicated experimental arrangement:
## Example of steel sample

### Example of steel sample

### 60Co in steel sample

<table>
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<td>0.1511</td>
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<td></td>
</tr>
</tbody>
</table>

### Material mass (m) and Density (ρ)

<table>
<thead>
<tr>
<th>Material mass (m)</th>
<th>Density (ρ)</th>
</tr>
</thead>
<tbody>
<tr>
<td>75,8032 g</td>
<td>3.5 cm</td>
</tr>
<tr>
<td>77741,46 cm</td>
<td>1 cm</td>
</tr>
<tr>
<td>74006,12 cm</td>
<td>3.94 g.cm⁻³</td>
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<tr>
<td>77393,3 cm</td>
<td>1,0001E+00</td>
</tr>
<tr>
<td>80226,6 cm</td>
<td>1,0001E+00</td>
</tr>
</tbody>
</table>

### Calculation of μ(cm⁻¹)

\[ \mu(\text{cm}^{-1}) = - \ln \left( \frac{l}{l_0} \right) \]

### Calculation of \( \mu(\text{cm} \cdot \text{g}^{-1}) \)

\[ \mu(\text{cm} \cdot \text{g}^{-1}) = \frac{\mu(\text{cm}^{-1})}{\rho} \]

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Example of steel sample

<table>
<thead>
<tr>
<th>Energy (keV)</th>
<th>(\mu) (cm(^{-1}))</th>
<th>(\mu) (cm(^2).g(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>1173,2</td>
<td>0,4212</td>
<td>0,1069</td>
</tr>
<tr>
<td>1332,5</td>
<td>0,3917</td>
<td>0,0994</td>
</tr>
</tbody>
</table>
OUTLINE

General characteristics

Energy resolution

Peak area

Attenuation

Corrective factors
Half-life correction
Decay correction (short half-lifes)
Attenuation correction (screen)
Self-attenuation correction (see “Self attenuation” section)
Geometry correction (source-detector distance)
Coincidence summing correction (see “Coincidence summing ” section)

Background peaks
Escape peaks (annihilation or characteristics Ge X-rays)
Dead time correction
In-flight annihilation correction (Beta plus emitters)
Measurement at a date different from the reference time
To get the counting rate at the reference time: \( C_T \)

\( T_{1/2} \): radionuclide half-life
\( T_0 \): reference time
\( T_m \): acquisition start time

\[
C_T = \exp \left( \ln(2) \cdot \frac{(T_m - T_0)}{T_{1/2}} \right)
\]
The radionuclide decays during the measurement

If $T_r$ not negligible versus $T_{1/2}$:

$$C_D = \frac{\ln(2) \cdot (T_r/T_{1/2})}{1 - \exp\left(-\ln(2) \cdot (T_r/T_{1/2})\right)}$$

Or:

$$C_D = \frac{\lambda \cdot T_r}{1 - \exp(-\lambda \cdot T_r)}$$

where

$$\lambda = \frac{\ln(2)}{T_{1/2}}$$

$T_{1/2}$: radionuclide half-life

$T_r$: acquisition time (clock - real time !)

$T_m$: acquisition start time
Efficiency calibration

- Characteristic of the detector.
- Defined for an incident energy and source-detector geometry

Sample with different geometry to be measured

- self-absorption
- geometry variation (solid angle)
Calibration sources
Large number of geometries and matrices:
- point source
- disk-shape samples (filters)
- cylindrical geometries
  - liquid
  - solid
  - gas
- Marinelli geometries (Environment)
  - liquid
  - solid
  - gas
Source-to-detector distance can be adapted according to the counting rate

Small distance : Higher count rate

- Smaller counting time
- Less influence of the environmental background
- Possibility to use smaller samples (less self-absorption)

Large distance : Lower count rate

- Less pile-up
- Reduction of the coincidence summing effect
- Accuracy of the position (relative uncertainty)

Compromise between counting time and corrective factors
Efficiency transfer principle (Moens)

Calibration geometry:
Full-energy peak efficiency: \( \varepsilon_0(E) = \varepsilon_0^G \cdot \varepsilon^I(E) \)

Where \( \varepsilon_0^G = \frac{\Omega_0}{4\pi} \)

Measurement geometry:
Full-energy peak efficiency: \( \varepsilon_m(E) = \varepsilon_m^G \cdot \varepsilon^I(E) \)

Where \( \varepsilon_m^G = \frac{\Omega_m}{4\pi} \)

\[ \varepsilon_m(E) = \varepsilon_0(E) \cdot \frac{\Omega_m}{\Omega_0} \]

Only valid for point sources
Volume: self-attenuation (+ matrix + source dimensions)
Efficiency calibration at 100 keV = 0.0171

Detector radius : r = 2 cm

Source - detector distance for calibration : d₀ = 10 cm

\[ \Omega_0 = 2\pi \left( 1 - \frac{d_0}{\sqrt{d_0^2 + r^2}} \right) = 0.122 \]

Source - detector distance for measurement (m₁) : \( d_{m1} = 20 \) cm

\[ \Omega_{m1} = 0.031 \text{ sr} \quad \frac{\Omega_{m1}}{\Omega_0} = 0.25 \]

Source - detector distance for measurement (m₂) : \( d_{m2} = 5 \) cm

\[ \Omega_{m2} = 0.449 \text{ sr} \quad \frac{\Omega_{m2}}{\Omega_0} = 3.7 \]
at 20 cm :

\[ \varepsilon_{m1}(100) = \varepsilon_0 (100) \cdot \frac{\Omega_{m1}}{\Omega_0} = 0.0171 \cdot \frac{0.031}{0.122} = 0.0043 \]

at 5 cm :

\[ \varepsilon_{m2}(100) = \varepsilon_0 (100) \cdot \frac{\Omega_{m2}}{\Omega_0} = 0.0171 \cdot \frac{0.449}{0.122} = 0.0629 \]

Approximation

\[ \Omega_0 = 2\pi \left(1 - \frac{d_0}{\sqrt{d_0^2 + r^2}}\right) \approx 2\pi \left(1 - \left(1 - \frac{r^2}{2d^2}\right)\right) = \pi \cdot \frac{r^2}{d^2} \]

\[ \frac{\Omega_m}{\Omega_0} = \frac{d_0^2}{d_m^2} \]

\[ \varepsilon_{m1}(100) = \varepsilon_0 (100) \cdot \frac{d_0^2}{d_{m1}^2} = 0.0171 \cdot \frac{(10)^2}{(20)^2} = 0.0043 \quad \Delta = \text{negligible} \]

\[ \varepsilon_{m2}(100) = \varepsilon_0 (100) \cdot \frac{d_0^2}{d_{m2}^2} = 0.0171 \cdot \frac{(10)^2}{(5)^2} = 0.0684 \quad \Delta = +9\% \]
THANK YOU FOR YOUR ATTENTION!