





GAMMA-SPECTROMETRY

INTRODUCTION

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General characteristics

Energy resolution

Peak area

Attenuation

Corrective factors



GENERAL CHARACTERISTICS

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



GENERAL CHARACTERISTICS

RESULTS DEPENDING ON EQUIPMENT CHARACTERISTICS :

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

Sample (material, shape)

Measurement conditions



¹⁵²Eu – Point source at 10 cm from the detector window (100 cm³) HPGe detector



Other features (continuum) : partial energy deposition, scattering, escape ...

BASIC FORMULA

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

or simply:

$$n_i = A \cdot I_i \cdot \varepsilon_i$$

A: activity (Bq)

- n_i : net peak area
- I_i : photon emission intensity
- ε_i : detection efficiency

-> 3 results depending on available input parameters:

$$\varepsilon_i = \frac{n_i}{A \cdot I_i}$$
 $A = \frac{n_i}{I_i \cdot \varepsilon_i}$ $I_i = \frac{n_i}{A \cdot \varepsilon_i}$

Hidden difficulties : peak area, efficiency for any energy (see "Fitting" section), corrective factors

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RADIOACTIVE DECAY DATA

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

$$T_{g} = I_{g} + I_{C}$$
Conversion coefficient: $\alpha_{T} = \frac{I_{\gamma}}{T_{\gamma}}$

$$I_{\gamma} = \frac{T_{\gamma}}{(1 + \alpha_{T})}$$

$$I_{\gamma} = \frac{T_{\gamma}}{(1 + \alpha_{T})}$$

$$I_{\gamma} = \frac{T_{\gamma}}{(1 + \alpha_{T})}$$

$$I_{\gamma} = \frac{T_{\gamma}}{(1 + \alpha_{T})} = \frac{9.91}{1.00181} = 9.89$$

$$I_{\gamma} = \frac{T_{\gamma}}{(1 + \alpha_{T})} = \frac{100}{1449} = 6.9$$



Example:

^{103m}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.

RADIOACTIVE DECAY DATA

^Drésent travail (2018)

[>]résent travail (2016)

HPGE DETECTOR

Size -> Efficiency P or N (low energies)

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



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Fluorescence

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n+ contact

HPGE DETECTOR

Different shapes, types and sizes



HPGE DETECTOR

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





SPECTROMETRY CHAIN

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: 10³ to 1.6 10⁴

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





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Energy resolution :

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

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

Resolving power : $\Delta E / E$ (%) : Some 10⁻³

(5 – 10 % for NaI(TI))

Characteristic of the spectrometer quality :

- separation of closely spaced peaks
- detection limit

ENERGY RESOLUTION

Initial photon emission : Monoenergetic line (source) (ΔE = some meV)

Widening and distortion of the resulting peak (spectrum)

Widening : Gaussian effects:

fluctuation of the number of charge carriers : ΔE_s electronic noise : ΔE_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



http://www.canberra.com/products/detectors/pdf/Germanium-Det-SS-C39606.pdf

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GAUSSIAN DISTRIBUTION

Distribution centered on energy E_0 with standard deviation, σ



Characteristics :

Full width at half maximum of the Gaussian

FWHM (ΔE) = 2,355 σ

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

$$G(E) = A \cdot \exp(\frac{(E - E_0)^2}{2\sigma^2})$$

STATISTICAL NOISE : ΔE_S

Cause : fluctuations of the number of charge carriers n

E : incident energy (some keV) $n = \frac{E}{w}$ w : mean pair creation energy (some eV)

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

Standard deviation of the number of charge carriers

-> Standard deviation of the deposited energy (n w)

$$\sigma_{ES} = w \cdot \sigma_n = w \cdot \sqrt{n} = w \cdot \sqrt{\frac{E}{w}} = \sqrt{w \cdot E}$$

STATISTICAL NOISE : ΔE_S

Example : (at 77K : w = 2.96 eV in Ge w = 3.76 eV in Si)

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

$$\sigma_{Es} = \sqrt{w \cdot E} = \sqrt{2.96 \cdot 10^6} = 1.72 \ keV$$
$$\Delta_{Es} = 2.355 \cdot \sigma_{Es} = 4.05 \ keV$$
$$???$$

The observed width is much lower (< 2 keV)

-> F = Fano factor : $F = \frac{Observed variance}{Poisson predicted variance}$

STATISTICAL NOISE : ΔE_S

$$\Delta_{ES} = 2.355 \cdot \sqrt{F \cdot \sigma_{ES}^2} = 2.355 \cdot \sqrt{F \cdot w \cdot E}$$

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



ELECTRONICAL NOISE : ΔE_E

Preamplifier – amplifier

- Independent on the energy
- Can be determined using a pulser : input at the preamplifier « test »



CHARGE COLLECTION : ΔE_{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)





$$\Delta E^2 = \Delta E_s^2(E) + \Delta E_c^2 + \Delta E_E^2$$

$$\begin{split} \Delta \mathsf{E}_{\mathsf{S}} &= \text{statistical noise (dependent on the energy)} \\ \Delta \mathsf{E}_{\mathsf{C}} &= \text{charge collection} \\ \Delta \mathsf{E}_{\mathsf{E}} &= \text{electronical noise} \end{split}$$

First approximation :
$$\Delta E^2 = K + \Delta E_s^2(E)$$

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

FWHM versus the energy



Linear fitting : $FWHM^2 = 0.00182E + 0.4141$ K = 0.414 keV (electronics + charge collection)

 $(2.355)^2$ F w = 0.00182 -> F = 0.110



OTHER PEAK SHAPE CHARACTERISTICS

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 : FWTM / FWHM = 1.82 FWFM / FWHM = 2.38

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







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REGION OF INTEREST

For quantitative analysis : determination of N(E) = 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



Regions of interest

FINDING PEAKS IN SPECTRUM

Automatic peak search







Gaussian x Top-hat filter

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

Seconde derivative : negative minimum at the position peak



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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,I] first channel = f; last channel = I









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

Beer-Lambert law

Mass attenuation coefficients

Beer-Lambert law

Attenuation of a narrow parallel photon beam



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

 μ 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: $\tau_i(E)$ $\tau \approx \text{const } Z^{4.5} \cdot E^{-3}$ (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 keV)

For practical use : tables function of Z and E Tables : cross sections ($1 \text{ barn} = 10^{-24} \text{ cm}^2$) or mass attenuation ($\text{cm}^2.g^1$)

Interaction	Linear attenuation coefficient (cm ⁻¹)	Mass attenuation coefficient (cm ^{2.} g ⁻¹)
Photoelectric	τ	au/ ho
Compton	σ	σ/ρ
Pair production	к	κ/ρ
Total	$\mu = \tau + \sigma + \kappa$	$\mu/\rho = \tau/\rho + \sigma/\rho + \kappa/\rho$

Photoelectric absorption coefficient = sum of photoelectric effect in each electronic shell (subshells):

$$\tau = \tau_{\rm K} + (\tau_{\rm L1} + \tau_{\rm L2} + \tau_{\rm L3}) + (\tau_{\rm M1} + \tau_{\rm M2+} \tau_{\rm M3+} \tau_{\rm M4+} \tau_{\rm M5}) + \dots$$

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

For E = Ei : absorption discontinuity: maximum ionisation probability in shell i

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



Germanium mass attenuation coefficient



Lead mass attenuation coefficient



Composition known -> calculation Composition unknown -> measurement

Calculation: Attenuation coefficients table

> XCOM (NIST Database)



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: H_2O Mass fraction = 1016 - 36.45 = 979.55

https://physics.nist.gov/PhysRefData/Xcom/html/xcom1.html

National Institute of Standards and Technology Physics Laboratory	XCOM
Element/Compound/Mit	xture Selection
following information:	
Help	
	Identify material by: O Element O Compound Mixture
	Method of entering additional energies: (optional)
	• Additional energies from file (Note: Your browser must be file-upload compatible)
	Submit Information Reset
XCOM	

520 0.9 SaCl 0.1	ted by a space for each compound. One compound per line. For example:
Note: Weights not summing to 1 will be normalized.	
801 56.45 820 575.55	
Dptional output title: Mixture HC/1N	Additional energies in MeV: (optional) (up to 75 allowed)
Fraph options:	Note: Energies must be between 0.001 - 100000 MeV (1 keV - 100 GeV) (only 4 significant figures will be us One energy per line. Blank lines will be ignored.
Total Attenuation without Coherent Scattering Coherent Scattering Incoherent Scattering	0.122 0.220
Photoelectric Absorption	tel include the standard grid
Photoelectric Absorption Pair Production in Nuclear Field Pair Production in Electron Field	Energy Range:

Constituents	(Atomic	Number	: Fraction	by	Weight)	
onstituents	(raconne	rapinet	. I Idettoll	05	or eight j	

- Z=1 : 0.108875 Z=8 : 0.856240
- Z=17 : 0.034884

To download data in spreadsheet (array) form, choose a delimiter and use the checkboxes in the table heading. After down

Delimiter:

- space
- (vertical bar)
- O tab
- O newline
- Download data Reset

		Scattering		1000	Pair Pr	oduction	Total Attenuation		
Edge	(required) Photon Energy	Coherent	 Incoherent	Photoelectric Absorption	In Nuclear Field	In Electron Field	With Coherent Scattering	Without Coherent Scattering	
	MeV	cm ² /g	cm ² /g	cm ² /g	cm ² /g	cm ² /g	cm ² /g	cm ² /g	
	1.000E-02	2.48E-01	1.53E-01	6.73E+00	0.00E+00	0.00E+00	7.14E+00	6.89E+00	
	1.500E-02	1.44E-01	1.68E-01	1.92E+00	0.00E+00	0.00E+00	2.24E+00	2.09E+00	
	2.000E-02	9.57E-02	1.76E-01	7.80E-01	0.00E+00	0.00E+00	1.05E+00	9.55E-01	
	3.000E-02	5.08E-02	1.82E-01	2.15E-01	0.00E+00	0.00E+00	4,47E-01	3.96E-01	
	4.000E-02	3.12E-02	1.82E-01	8.52E-02	0.00E+00	0.00E+00	2.98E-01	2.67E-01	
	5.000E-02	2.11E-02	1.79E-01	4.14E-02	0.00E+00	0.00E+00	2.42E-01	2.21E-01	
	6.000E-02	1.52E-02	1.76E-01	2.29E-02	0.00E+00	0.00E+00	2.14E-01	1.99E-01	
	8.000E-02	8.93E-03	1.69E-01	8.99E-03	0.00E+00	0.00E+00	1.87E-01	1.78E-01	
	1.000E-01	5.86E-03	1.62E-01	4.35E-03	0.00E+00	0.00E+00	1.72E-01	1.66E-01	
	1.220E-01	4.00E-03	1.55E-01	2.28E-03	0.00E+00	0.00E+00	1.61E-01	1.57E-01	
	1.500E-01	2.68E-03	1.47E-01	1.17E-03	0.00E+00	0.00E+00	1.51E-01	1.48E-01	
	2.000E-01	1.53E-03	1.35E-01	4.67E-04	0.00E+00	0.00E+00	1.37E-01	1.35E-01	
_	2 000 - 01	6 0 1E 01	1 178 01	1 245 04	0.000.000	0.000 .00	1 100 01	1 198 01	

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XCOM results





EXPERIMENTAL MEASUREMENT

Principle : Use the unknown matrix and a collimated photon beam



Two successive measurements

- Empty container
- Container filled with unknown matrix with thickness x

Experimental measurement

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:

$$\frac{u^{2}(\mu)}{\mu^{2}} = \frac{u^{2}(x)}{x^{2}} + \frac{1}{\ln^{2}\left(\frac{N_{0}(E)}{N(E)}\right)} \cdot \left(\frac{u^{2}(N_{0}(E))}{N_{0}^{2}(E)} + \frac{u^{2}(N(E))}{N^{2}(E)}\right)$$

Experimental measurement

Dedicated experimental arrangement :



Example of steel sample

⁶⁰Co in steel sample

		Eu-152	13,522	0,016							
			4,2671E+08	5,0E+05			1 a =	j			
			7,1119E+06	8,4E+03			1 a =	S			
			1,1853E+05	1,4E+02							
			4,9388E+03	5,8E+00		Date et heure	e de référence	25/03/20	018 12:00		
			1,3522E+01	1,6E-02							
	Eu-152 wit	hout material	Eu-152 w	ith mterial			Attenuati	on results			
						, ,					(
					Material mass	(m)	75,8032	g			μ
	Start	26/03/2018	Start	27/03/2018		Stand. unc.	0,0008	g			
	Real time	80226,6	Real time	77741,46	Diameter (D)		3,5	cm			
	Active time	77393,3	Active time	74906,12		Stand. unc.	0,02	cm			
	% TM	3,5%	% TM	3,6%	Height (x)		1	cm			
	Half-life cor	1,0001E+00	Half-life cor	1,0003E+00		Stand. unc.	0,01	cm			
	Decay corr.	1,0001E+00	Decay corr.	1,0001E+00	Density (ρ)		3,94	g.cm ⁻³			
						Stand. unc.	0,01	g.cm ⁻³			
											μ(α
Energy (keV)	NET	Rel. Stand. Unc	NET	el. Stand. Un	1/10	el. Stand. Un	μ (cm⁻¹)	el. Stand. Un	μ (cm².g ⁻¹)	Rel. Stand. Unc	
121,78	2248977	0,1%	328827	0,2%	0,1511	0,2%	1,890	1,0%	0,480	1,0%	
244,70	412482	0,2%	159477	0,3%	0,3995	0,3%	0,917	1,1%	0,233	1,1%	
344,28	1096300	0,1%	491001	0,1%	0,4628	0,2%	0,770	1,0%	0,196	1,1%	
411,12	66423	0,4%	32317	0,6%	0,5028	0,7%	0,688	1,4%	0,175	1,4%	
443,97	108192	0,3%	52366	0,4%	0,5001	0,5%	0,693	1,3%	0,176	1,3%	
778,90	399743	0,2%	226912	0,2%	0,5866	0,3%	0,533	1,1%	0,135	1,2%	
867,38	131623	0,3%	77692	0,4%	0,6099	0,5%	0,494	1,4%	0,125	1,4%	
964,08	454460	0,1%	273614	0,2%	0,6221	0,2%	0,475	1,1%	0,120	1,2%	
1085,84	371627	0,2%	228130	0,2%	0,6343	0,3%	0,455	1,2%	0,116	1,2%	
1112,08	428505	0,2%	266091	0,2%	0,6417	0,2%	0,444	1,1%	0,113	1,2%	
1212,95	43790	0,5%	27500	0,6%	0,6489	0,8%	0,432	2,0%	0,110	2,1%	
1299,14	53310	0,4%	34077	0,5%	0,6605	0,7%	0,415	1,9%	0,105	2,0%	
1408,01	638601	0,1%	418820	0,2%	0,6777	0,2%	0,389	1,1%	0,099	1,2%	

$$\mu(cm^{-1}) = \frac{-\ln\left(\frac{I}{I_0}\right)}{x}$$

$$\mu(cm^1.\,g^{-1}) = \frac{\mu(cm^{-1})}{\rho}$$

Example of steel sample

Example of steel sample







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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)



HALF-LIFE

Measurement at a date different from the reference time To get the counting rate at the reference time: $C_{\rm T}$

- T_{1/2} : radionuclide half-life
- T₀ : 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 $T_{1/2}$:

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

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



GEOMETRY CORRECTION

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



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 GEOMETRY

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
- -> Possiblitlity 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 transfer example



Efficiency transfer example

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
$$\Omega_{0} = 2\pi \left(1 - \frac{d_{0}}{\sqrt{d_{0}^{2} + r^{2}}} \right) \approx 2\pi \left(1 - \left(1 - \frac{r^{2}}{2d^{2}} \right) \right) = \pi \cdot \frac{r^{2}}{d^{2}}$$
$$\frac{\Omega_{m}}{\Omega_{0}} = \frac{d_{0}^{2}}{d_{m}^{2}}$$
$$\varepsilon_{m1}(100) = \varepsilon_{0} \ (100) \cdot \frac{d_{0}^{2}}{d_{m1}^{2}} = 0.0171 \cdot \frac{(10)^{2}}{(20)^{2}} = 0.0043 \qquad \Delta = \text{negligible}$$
$$\varepsilon_{m2}(100) = \varepsilon_{0} \ (100) \cdot \frac{d_{0}^{2}}{d_{m2}^{2}} = 0.0171 \cdot \frac{(10)^{2}}{(5)^{2}} = 0.0684 \qquad \Delta = +9\%$$

DE LA RECHERCHE À L'INDUSTRIE



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THANK YOU FOR YOUR ATTENTION !

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