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# Introduction in NAA

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# Introduction to Neutron Activation Analysis (NAA)


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# Introduction

Two features of (n,γ) reactor NAA are making its standardization potentially easy and accurate:

1. **the high penetrability of matter for neutrons**
2. **existence of a delayed signal (besides the prompt gamma's).**

Hence, standard and sample can be excited simultaneously and induced signals of both can be measured successively after a suited time following the end of irradiation.

# Introduction

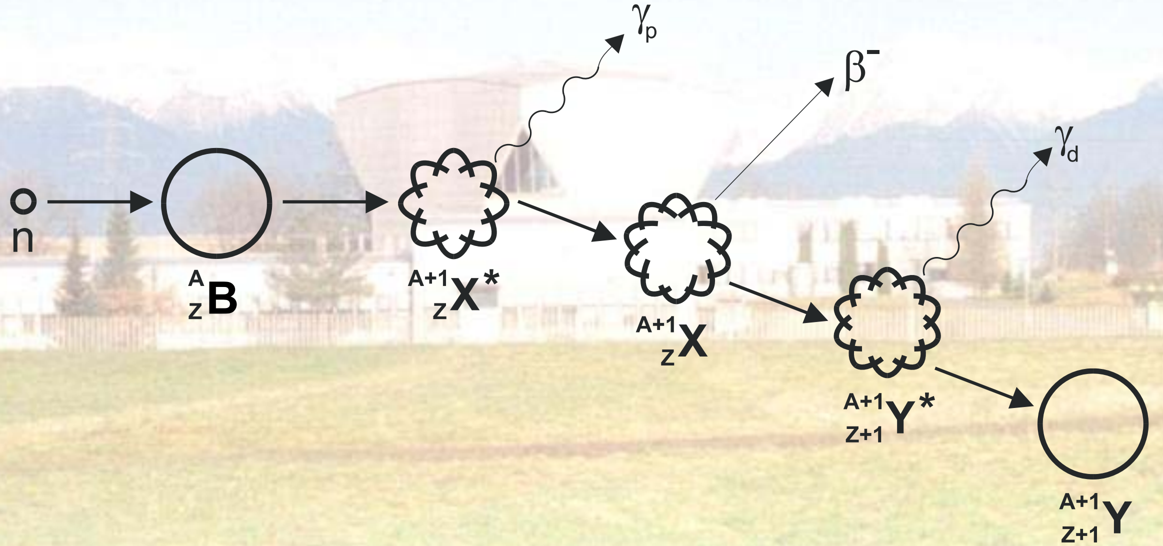
Other consequences:

- NAA is a bulk analysis method with multi-element capability (element concentration and measured signal is nearly matrix-independent).
- Matrix preparation can be kept simple.
- Treatment of sample (and standard) after irradiation is possible (enabling etching, dissolution, chemical separation - RNAA).
- High sensitivity (down to the $10^{-6}$, $10^{-9}$ or even to the $10^{-12}$ g/g) attainable for many elements.
- Reference method for certification of new CRMs or RMs.

# Introduction

Nuclear reactions: - direct (B + a $\rightarrow$ Y + b); ~ $10^{-22}$ - $10^{-21}$ s

- meta stable (B + a $\rightarrow$ X* $\rightarrow$ Y + b); ~ $10^{-16}$ - $10^{-14}$ s




[ e. g. $n + {}^{197}_{79}Au \longrightarrow {}^{198}_{79}Au^* \xrightarrow{\gamma_p} {}^{198}_{79}Au \xrightarrow{\beta^-} {}^{198}_{80}Hg^* \xrightarrow{\gamma_d} {}^{198}_{80}Hg$ ]

A typical (n,$\gamma$) reaction with $\beta^-$ decay.

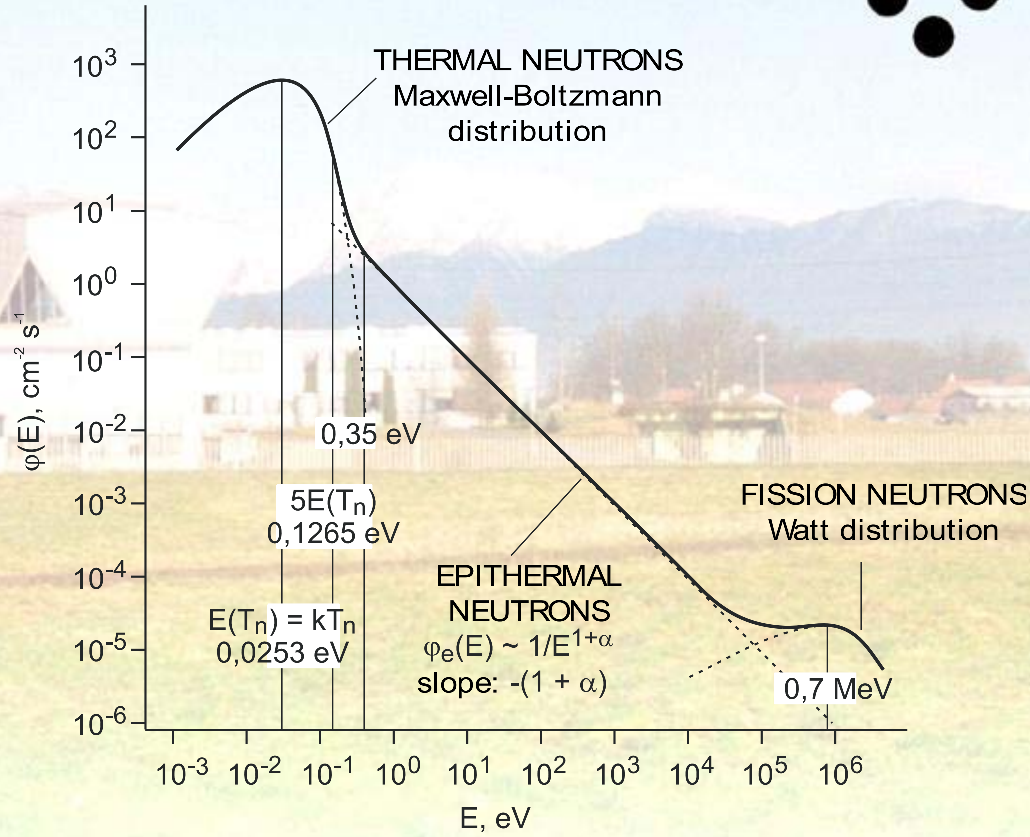
# Introduction

- NAA - four techniques:
  - Prompt Gamma Neutron Activation Analysis (**PGNAA**) (usually with energies < 0.001 eV); prompt gammy rays ($\gamma_p$) emitted by the compound nucleus *X** are measured
  - Thermal Neutron Activation Analysis (**TNAA**); from reactions in a well-thermalised reactor spectrum, the decay gamma rays ($\gamma_d$) are measured
  - Epithermal Neutron Activation Analysis (**ENAA**); resonance reactions are utilised by using suitable filters to remove thermal neutrons from the reactor spectrum and measuring decay gamma rays
  - Fast Neutron Activation Analysis (**FNAA**); high-energy neutron sources are used without the presence of a moderator for slowing down the neutrons and gamma rays are measured.

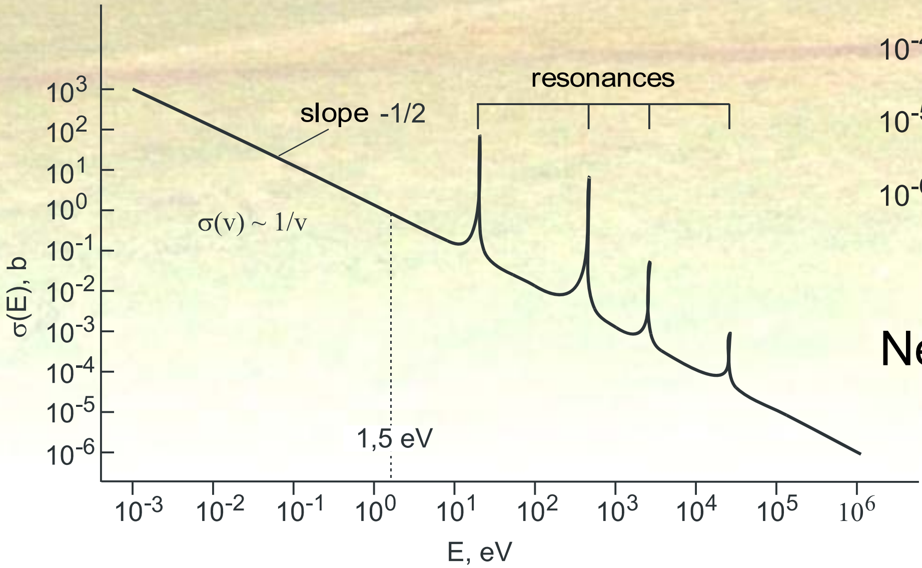
# (n,γ) reaction rate

**Specific reaction rate per target nuclide**

$$R_X = \frac{R}{N_1} = \int_0^\infty \sigma(E)\varphi(E)\,dE$$

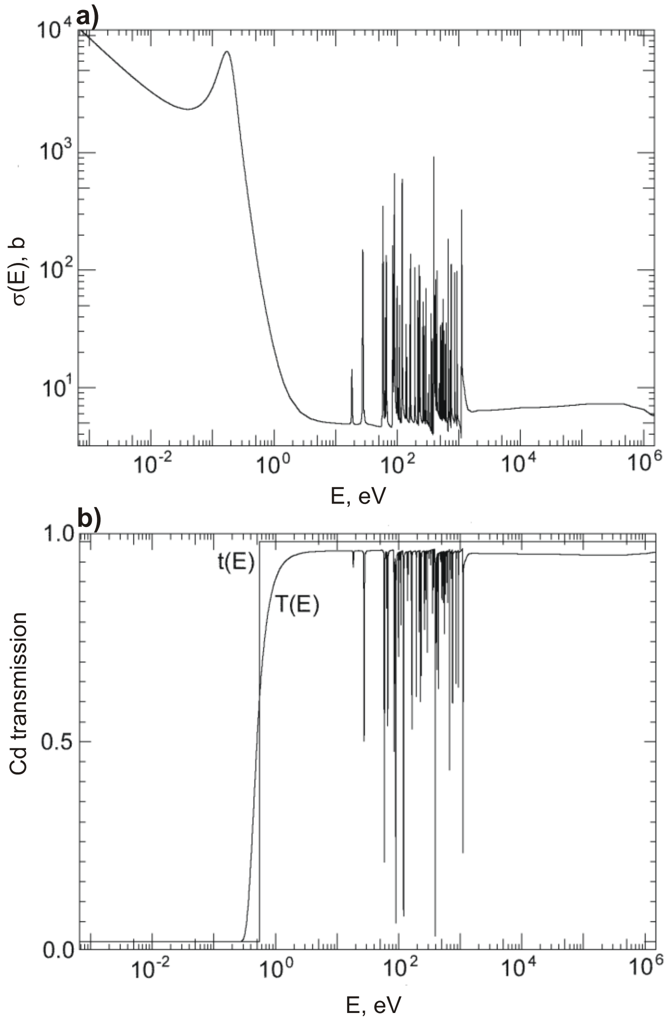



Neutron fluence rate distribution vs. E




Cross-section vs. E ($\sigma(\nu) \sim 1/\nu$)

# (n,γ) reaction rate




Cd total cross section and Cd transmission function T(E) for 1 mm Cd

Hogdahl convention:

$$R_X = \int_0^{v_{Cd}} \sigma(v)\, \varphi'(v)\, dv + \int_{E_{Cd}}^{E_2} \sigma(E)\, \varphi(E)\, dE = R_{X,th} + R_{X,e}$$

- all (n,γ) reactions are as follows:

$$\sigma(v) \propto 1/v$$

- $E_{Cd}$ = 0.55 eV

$$R_X = \sigma_0\, v_0\, n_{th} + \varphi_e\, I_0(\alpha) = \sigma_0\, \varphi_{th} + \varphi_e\, I_0(\alpha)$$

with: $\sigma_0 = \sigma(v_0)$ at reference velocity $v_0$ =2200 m $s^{-1}$, $\varphi_{th} = v_0\, n_{th}$ **fluence rate for thermal neutrons**, $n_{th}$ – thermal neutron density **and**

$$I_0(\alpha) = (1eV)^{\alpha} \int_{E_{Cd}}^{E_2} \frac{\sigma(E)\, dE}{E^{\,1+\alpha}}$$

**resonance integral** for epithermal part of spectra

# (n,γ) reaction rate

For better adjustment of (n,γ) reaction rate in Hogdahl convention it is necessary to introduce some correction factors:

- $G_{th}$ - thermal neutron self-shielding (nucleus density)
- $G_e$ - epithermal neutron self-shielding (density + resonance parameters)
- $F_{Cd}$ - Cd transmission factor for epithermal neutrons

$$R_X = G_{th}\,\varphi_{th}\,\sigma_0 + G_e\,\varphi_e\,I_0(\alpha)$$

**Cd-ratio:**

$$R_{Cd} = F_{Cd}\,R^*_{Cd} = 1 + \left(\frac{G_{th}\,f}{G_e\,Q_0(\alpha)}\right)$$ with $$f = \frac{\varphi_{th}}{\varphi_e}$$ and $$Q_0(\alpha) = \frac{I_0(\alpha)}{\sigma_0}$$

with $R_{Cd}$* - measured Cd ratio:

$$R^*_{Cd} = \frac{\int_0^\infty \sigma(E)\,\varphi(E)\,dE}{\int_0^\infty T(E)\,\sigma(E)\,\varphi(E)\,dE} = \frac{\int_0^{E_2} \sigma(E)\,\varphi(E)\,dE}{F_{Cd}\int_{E_{Cd}}^{E_2} \sigma(E)\,\varphi(E)\,dE}$$

# Activation equation

$$N_1^0 = \frac{w\,N_A\,\Theta}{M}$$

$$\frac{dN_1}{dt'} = -N_1\,R_{X,1}$$

$$N_1 = N_1^0\; e^{-R_{X,1}\,t'}$$

**taking into account** the removal of nuclei of the nuclide produced

$$\frac{dN}{dt'} = N_1\,R_{X,1} - N\,(\lambda + R_N) = R_{X,1}\;N_1^0\;e^{-R_{X,1}\,t'} - N\,(\lambda + R_N)$$

$$N = \frac{N_1^0\;R_{X,1}\;e^{-R_{X,1}\,t_{irr}}}{(\lambda + R_N - R_{X,1})}\left(1 - e^{\,(R_{X,1}-\lambda-R_N)\;t_{irr}}\right)$$

$$N = \frac{N_1^0\;R_{X,1}}{\lambda}\left(1 - e^{-\lambda\,t_{irr}}\right)$$

**not taking into account** the removal of nuclei of the nuclide produced

Burn-up factor

$$F_{burn} = \frac{\lambda\;e^{-R_{X,1}\,t_{irr}}\;\left(1 - e^{\,(R_{X,1}-\lambda-R_N)\;t_{irr}}\right)}{(\lambda + R_N - R_{X,1})\,\left(1 - e^{-\lambda\;t_{irr}}\right)}$$

$N^0_1$ - initial number of irradiated nuclei in the target
$N_1$ - number of target nuclide
N - number of radionuclide
*w* – mass of the investigated element
$N_A$- Avogadro constant=6.022045 $10^{23}$ $mol^{-1}$
M - molar mass
*Θ* - isotopic abundance
λ - decay constant = *ln(2)/$T_{1/2}$*
$t_{irr}$ - irradiation time
$R_{X,1}$ - specific reaction rate of target nuclide
$R_N$ - specific reaction rate of radionuclide

# Activation equation

Due to radioactive decay, the number of radioactive nuclei *N* decreases with time

$$N = \frac{N_1^0 \; R_{X,1}}{\lambda}(1 - e^{-\lambda t_{irr}})e^{-\lambda t}$$

The reaction rate of the radioactive nuclei *N*

$$\frac{dN}{dt} = -\lambda N$$

is proportional to the count rate measured by the detector.

The result is the **number of counts** in the full-energy peak ($N_p$) with the start of detection at time $t_d$ after the end of irradiation:

$$N_p = \frac{N_1^0 \; R_{X,1}}{\lambda}(1 - e^{-\lambda t_{irr}})e^{-\lambda t_d}(1 - e^{-\lambda t_m})\, \gamma \, \varepsilon_p$$

↓

$$\frac{N_p}{t_m} = N_1^0 \, R_{X,1} (1 - e^{-\lambda t_{irr}}) \, e^{-\lambda t_d} \, \frac{(1 - e^{-\lambda t_m})}{\lambda t_m} \gamma \, \varepsilon_p$$

$$S = 1 - e^{-\lambda \, t_{irr}}$$

Factor for saturation during irradiation

$$D = e^{-\lambda \, t_d}$$

Decay factor

$$C = \frac{(1 - e^{-\lambda \, t_m})}{\lambda \, t_m}$$

“Measurement” factor

# Activation equation

$$\frac{N_p}{t_m} = \frac{w N_A \Theta}{M} R_{X,1}\, S\, D\, C\, \gamma\, \varepsilon_p$$

$\varepsilon_p$ Full-energy peak detection efficiency, including gamma attenuation

$\gamma$ Probability of $\gamma$ emission

**Activity, *A*** [$s^{-1}$] or the total count rate in the detector:

$$A = N_1^0\, R_{X,1}\, \gamma\, \varepsilon_p = \frac{w N_A \Theta}{M} R_{X,1}\, \gamma\, \varepsilon_p$$

**Specific count rate ($s^{-1}$ $g^{-1}$)**

$$A_{sp} = \frac{N_A \Theta}{M} R_{X,1}\, \gamma\, \varepsilon_p$$

or

$$A_{sp} = \frac{N_p / t_m}{S\, D\, C\, w}$$

Specific count rate ($s^{-1}$ $g^{-1}$)

$$A_{sp} = \frac{N_A\, \Theta}{M} \frac{1}{F_{burn}} \left(G_{th}\, \varphi_{th}\, \sigma_0 + G_e\, \varphi_e\, I_0(\alpha)\right)\, \gamma\, \varepsilon_p$$

Specific count rate under Cd activation ($s^{-1}$ $g^{-1}$)

$$(A_{sp})_{Cd} = \frac{N_A\, \Theta}{M} \frac{1}{F_{Cd}} G_e\, \varphi_e\, I_0(\alpha)\, \gamma\, \varepsilon_p$$

Nuclear research reactor
**TRIGA Mark II** (250 kW)

- Short and long irradiation in the CC:

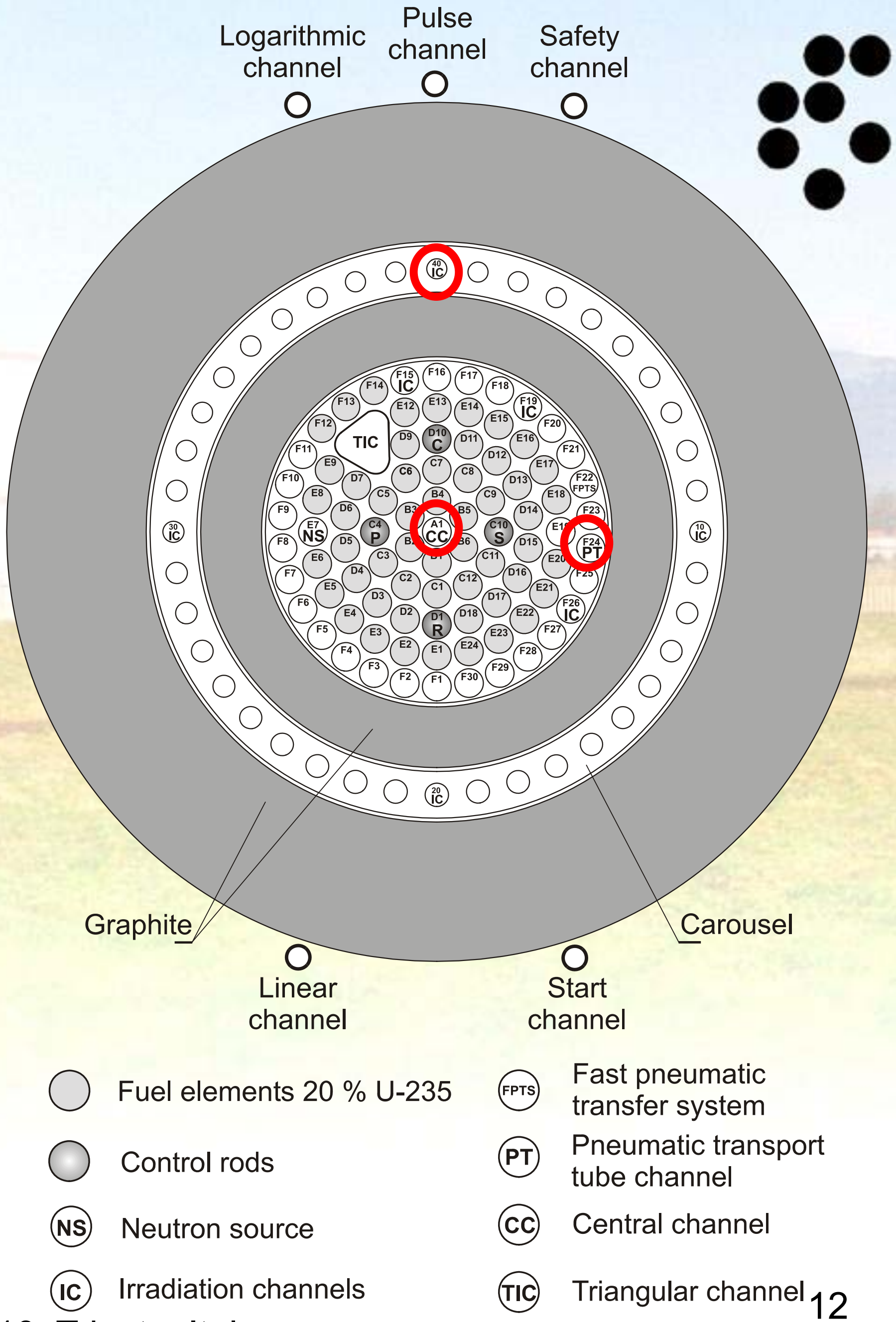
  $\varphi_{th} \sim 10 \cdot 10^{12}\ cm^{-2}\ s^{-1}$

- Short irradiation in the PT and in the FPTS (up-to 30 min.)

  $\varphi_{th} \sim 3.5 \cdot 10^{12}\ cm^{-2}\ s^{-1}$

- Long irradiation in the IC-40 (typically 20 hours)

  $\varphi_{th} \sim 1.1 \cdot 10^{12}\ cm^{-2}\ s^{-1}$



# Nuclear data of the target and nuclide formed

| El. | Nuclide | $T_{1,2}$ | $\sigma_{0,1}$, b | $I_{0,1}$, b | $\sigma_{0,N}$, b | $I_{0,N}$, b |
|---|---|---|---|---|---|---|
| Ru | $^{105}$Rh | 35.36 h | 0.32 | 4,3 | 16000 | 17000 |
| Eu | $^{152}$Eu | 13.516 y | 5900 | 1510 | 12800 | 1580 |
| Eu | $^{152m}$Eu | 9.113 h | 3304 | 1790 | 70000 | 1580 |
| Gd | $^{153}$Gd | 240.4 d | 735 | 2020 | 36000 | n. d. |
| Au | $^{198}$Au | 2.695 d | 98.65 | 1550 | 25100 | 31031 |
| n. d.: no data | | | | | | |

# Burn-up factor ($F_{burn}$) in irradiation channels of the TRIGA reactor (calculations)

**IC-40**: $\varphi_{th} \sim 1.0 \cdot 10^{12}\ cm^{-2}s^{-1}$
**PT**: $\varphi_{th} \sim 3.5 \cdot 10^{12}\ cm^{-2}s^{-1}$
**CC**: $\varphi_{th} \sim 10 \cdot 10^{12}\ cm^{-2}s^{-1}$

| El. | Nuclide | $T_{1,2}$ | $F_{burn}$ (20 h) | | |
|---|---|---|---|---|---|
| | | | IC-40 | PT | CC |
| Ru | $^{105}Rh$ | 35.36 h | 0.999 | 0.998 | 0.994 |
| Eu | $^{152}Eu$ | 13.516 y | 0.999 | 0.998 | 0.993 |
| Eu | $^{152m}Eu$ | 9.113 h | 0.998 | 0.993 | **0.980** |
| Gd | $^{153}Gd$ | 240.4 d | 0.999 | 0.995 | 0.987 |
| Au | $^{198}Au$ | 2.695 d | 0.999 | 0.997 | **0.991** |

| El. | Nuclide | ***F*<sub></sub>** | | | | | |
|---|---|---|---|---|---|---|---|
| | | **1 h** | **10 h** | **20 h** | **50 h** | **100 h** | **200 h** |
| Ru | $^{105}Rh$ | 1.000 | 0.997 | 0.994 | 0.988 | 0.980 | 0.973 |
| Eu | $^{152}Eu$ | 1.000 | 0.997 | 0.993 | 0.983 | 0.967 | 0.935 |
| Eu | $^{152m}Eu$ | 0.999 | 0.988 | 0.980 | 0.966 | 0.958 | 0.946 |
| Gd | $^{153}Gd$ | 0.999 | 0.993 | 0.987 | 0.968 | 0.937 | 0.879 |
| Au | $^{198}Au$ | 1.000 | 0.995 | 0.991 | 0.978 | 0.961 | 0.938 |

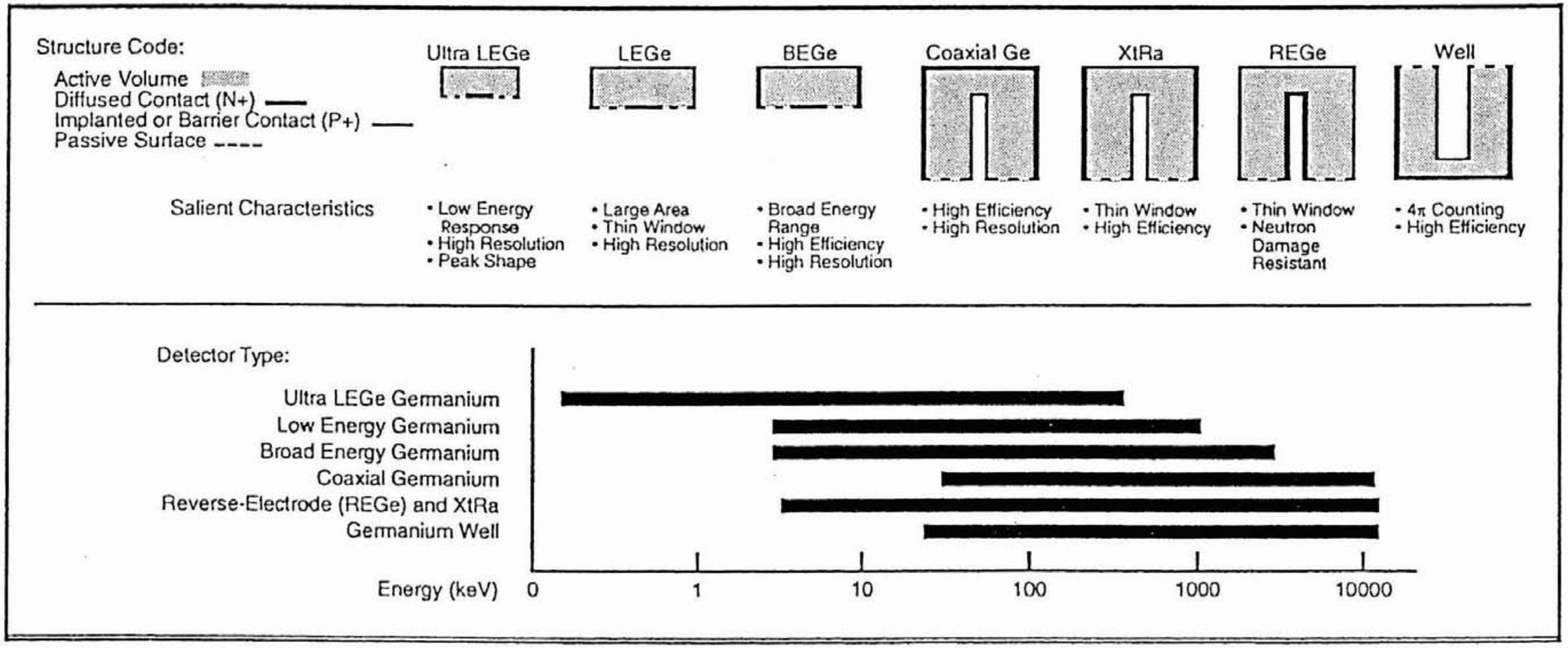
# (n, $\gamma$) Activation Analysis: Principles of standardization

**The mass of the element**

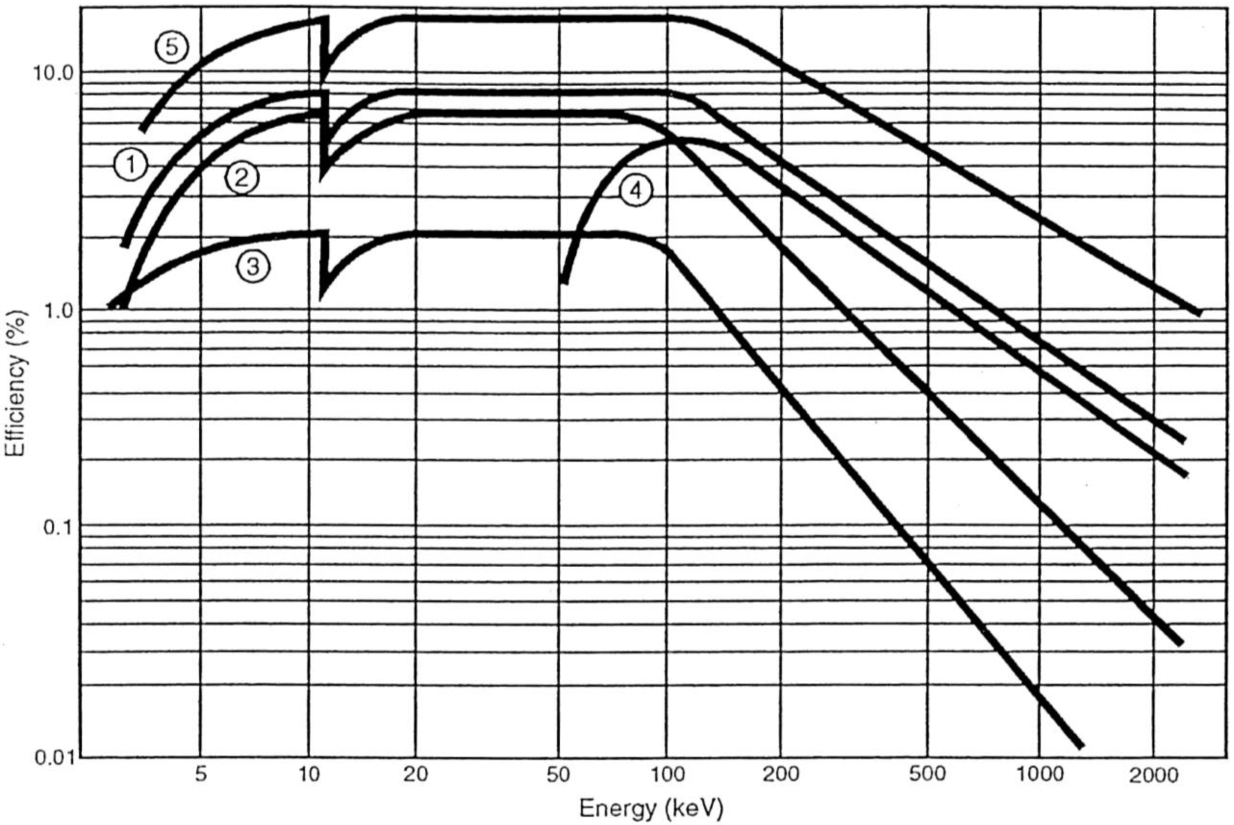
$$w_a = \frac{M_a}{N_A \Theta_a \gamma_a} \frac{\left(\frac{N_p / t_m}{S\,D\,C}\right)_a}{(G_{th,a}\,\varphi_{th,a}\,\sigma_{0,a} + G_{e,a}\,\varphi_{e,a}\,I_{0,a}(\alpha))\,\varepsilon_{p,a}}$$

1. **Relative standardization**
2. **Single-comparator standardization**
3. **Absolute (parametric) standardization**
4. **$k_0$-standardization**

# Types of Ge detectors



# Typical Absolute Efficiency Curves




Typical absolute efficiency curves for various Ge detectors
with 2.5 cm source to end-cap spacing

Code:
(1) REGe, 15% Relative Efficiency
XtRa, 15% Relative Efficiency
(2) LEGe, 10 cm² x 15 mm thick
(3) LEGe, 200 mm² x 10 mm thick
(4) Coaxial Ge, 10% Relative Efficiency
(5) BEGe, 5000 mm² x 30 mm thick

# Assembling an Energy Spectroscopy System