Samples preparation and Introduction to the methods of radioactivity measurements

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Content

- ✓ Sample pretreatment
  - ✓ Types of environmental samples and their features
  - ✓ Target radionuclides
  - ✓ Main processes of treatments
- ✓ Introduction to the methods of radioactivity measurements
  - ✓ Main definitions
  - ✓ Ionizing radiation
  - ✓ Interaction with matter
  - ✓ Main detector types
  - ✓ Analytical radiometric methods
  - ✓ QA/QC

Sample pretreatment

Main goals:

- to homogenise sample matrix as well as possible
- to preserve sample for storage
- to prevent losses of a target radionuclide
- to prevent biological decay

Sample pretreatment

Types of samples:

- Liquids: surface and underground water, milk etc.
- Soil
- Plants: grass, wood, leaves, seeds, berry etc.
- Foodstuff

Sample pretreatment

Target radionuclides:

Principal nuclides	*Production mode*	*Half-life*	*Type of radiation*	*Principal food pathways*
^{3}H	A/FP	12.4 y	β	All foods
^{14}C	A	5.73 y		
^{35}S	A	87.4 days		Milk and crops
^{99}Tc	FP	$0.22 \cdot 10^{6}$ y		Crops, crustaceans and offal
^{90}Sr	FP	28.5 y		Milk, mollusks and animal products
^{60}Co	A	5.3 y	βγ	Mollusks
^{106}Ru	FP	1 y		Crops, mollusks and offal
^{125}I	A	60.1 days		
^{129}I	FP	$15.7 \cdot 10^{6}$ y		Milk
^{131}I	FP	8 days		
^{134}Cs	A	2.1 y		
^{137}Cs	FP	30.1 y		Milk, fish, shellfish and animal products
^{210}Pb	N	22 y		Mollusks and offal
^{210}Po	N	140 days	α	
^{238}Pu	A	87. 74 y		Mollusks and offal
^{239}Pu	A	24000 y		
^{241}Am	A	432	α γ	

Main processes

Liquid

Milk

^{137}Cs – direct measurement using a gamma-spectrometer

^{90}Sr – radiochemical separation

Storage – use preservative ($NaHSO_3$, methanal C_2HO) and keep in fridge

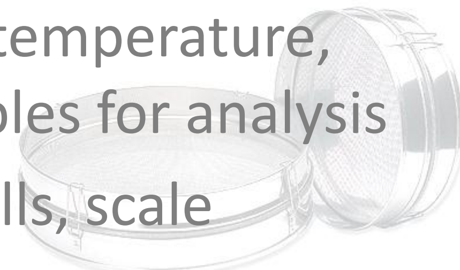
Sample pretreatment

Main processes

Soil

Drying at 105°C at an oven or at room temperature, sieving, grinding, collecting of subsamples for analysis

Equipment: oven, sieves (1mm), lab mills, scale



Gamma- emitters – direct measurement

β and α – radiochemical separation

Sample pretreatment

Main processes

Plant and foodstuffs samples

Drying at 105°C at an oven to stable weight, milling, collecting of subsamples for analysis

Moisture content – **obligatory**

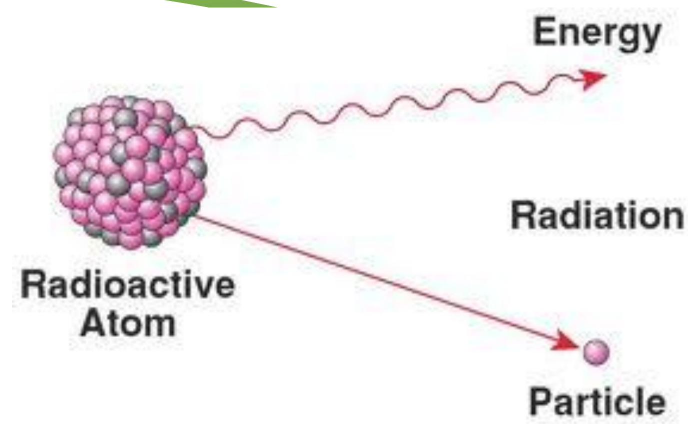
Equipment: oven, lab mills, blender, scale

Gamma- emitters – direct measurement

β and α – radiochemical separation

Methods

Main definitions



✓**Radioactive decay** - the spontaneous disintegration of unstable atomic nuclei or change of their energy state that is followed by ionizing radiation

✓**Radionuclide** – radioactive atom with fixed quantity of neutrons and protons. Radionuclides of the same chemical element – isotopes (**^{133}Cs, ^{134}Cs, ^{136}Cs, ^{137}Cs**).
(more than 2000 radionuclides have been identified)

✓**Ionizing radiation** is energy in the form of waves or particles that is able to remove electrons from atoms

Activity – decay rate

The decay rate is a nuclear property independent on temperature, pressure, chemical form of the isotope, and physical state of the substance.

$$A = \frac{dN}{dt}$$ ***BUT NOT concentration!!!***

^{137}Cs 1 Bq -- $3*10^{-10}$ mg
^{238}U 1 Bq -- 20 µg

SI unit Becquerel (Bq) (1 decay per second)

Becquerel is a measure of the rate (not energy!) of radiation emission from a source.

1 MBq=10^6 Bq
1 kBq=1000 Bq
1 mBq=0,001 Bq

Curie (Ci) – off-system unit,
1 Ci=$3,7\cdot10^{10}$ Bq

Methods
Main definitions

HALF LIFE

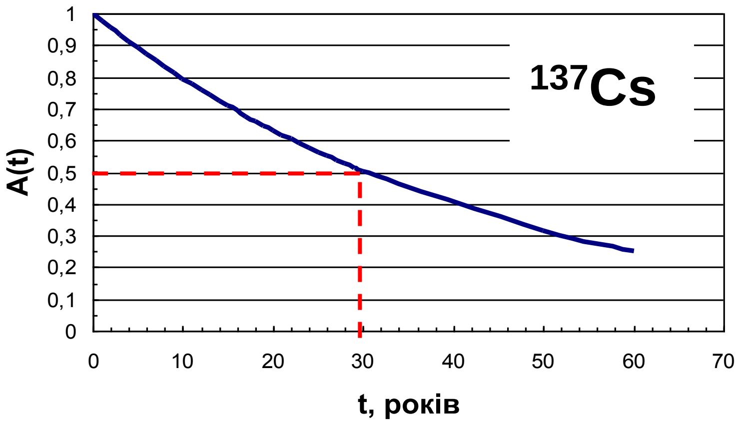
Radiation intensity from a radioactive source diminishes with time because more and more radioactive atoms decay and become stable atoms.

Half-life ($T_{1/2}$) is the period during which the radiation intensity decreases by half. This happens because half of the radioactive atoms will have decayed in one half-life period.

$T_{1/2}$ (^{137}Cs)= 30,17 years

$$A(t) = A(0) \cdot e^{-\lambda t}$$

$$\lambda = \frac{\ln(2)}{T_{1/2}} \approx \frac{0.693}{T_{1/2}}$$



Half-lives widely differ from one radioactive material to another and range from a fraction of a second to millions of years.

Methods
Ionizing radiation

The major types of radiation emitted during radioactive decay are alpha particles, beta particles, and gamma rays

Decay mode	Mass number		Atomic number	
	Parent	Daughters	Parent	Daughters
α- decay $^{238}Pu \Rightarrow {}^{234}U+\alpha+X$	A	A-4	Z	Z-2
β^- - decay $^{134}Cs \Rightarrow {}^{134}Ba+ \beta^- +\gamma$	A	A	Z	Z+1
β^+ - decay $^{22}Na+ \beta^+ \Rightarrow {}^{22}Ne+\gamma$	A	A	Z	Z-1
Isomeric transition (γ- ray ….) $^{137m}Ba \Rightarrow {}^{137}Ba +\gamma$	A	A	Z	Z

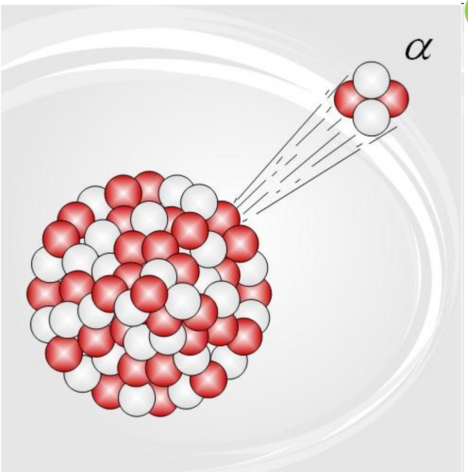
OTHER types of radiation

^{252}Cf	Jπ	Δ(MeV)	$T_{1/2}$	Decay Modes
	0+	76.0352	2.645 y 8	α : 96.91 % SF : 3.09 %

(A, Z) → (A – 1, Z) + n

^{223}Ra	Jπ	Δ(MeV)	$T_{1/2}$	Decay Modes
	3/2+	17.2349	11.43 d 5	α : 100.00 % ^{14}C : 8.9E-8 %

Alpha decay



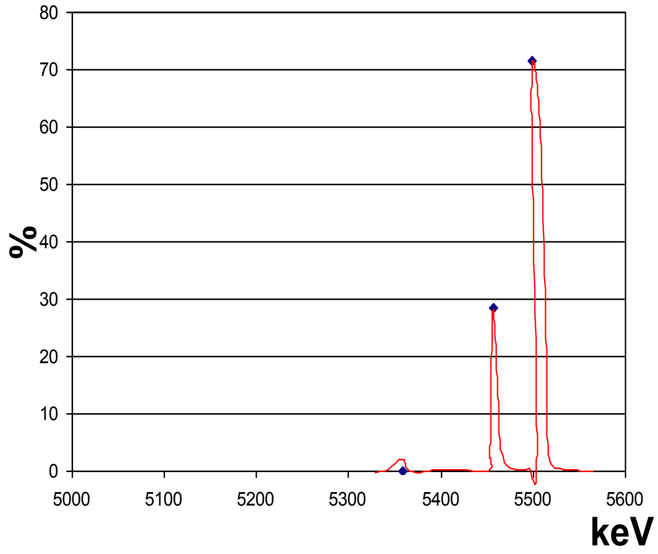
consist of two protons and two neutrons bound together into a particle identical to a helium nucleus

Charge **2+**

Average energy range - **3-7 Me**V

A radionuclide emits alpha particles that have discrete spectrum

Alpha particles are emitted by radioisotopes with atomic numbers greater than **82 (Pb)**.



Number of Alphas	**:**	**15**
Number of Betas	**:**	**0**
Number of Gammas	**:**	**33**
Number of X-Rays	**:**	**22**

$^{A}_{Z}R \Rightarrow {}^{A-4}_{Z-2}E + \alpha$

$^{238}\mathbf{Pu} \Rightarrow {}^{234}\mathbf{U} + \alpha + \mathbf{X}, \gamma$

α_1 - 5499,21 keV -71,5%

α_2 –5456,5 keV -28,4%

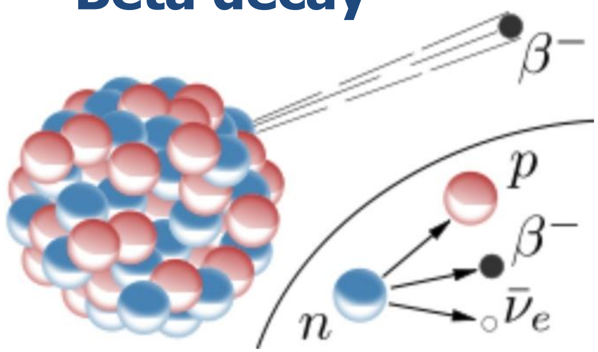
α_3 - 5358,3 keV -0,115%

X_1 – 43,49 keV – $3{,}8 \cdot 10^{-2}$ %

X_2 – 99,86 keV - $0{,}8 \cdot 10^{-2}$ %

X_3 – 152,72 keV - $1{,}1 \cdot 10^{-3}$ %

Beta decay



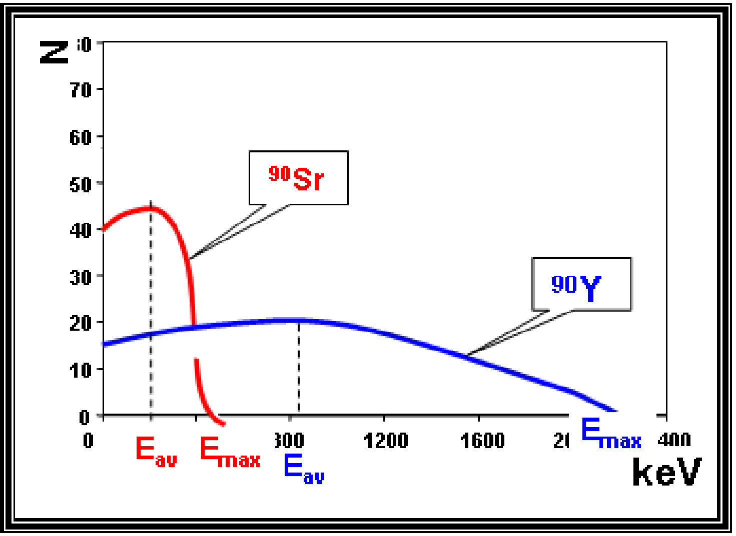
Beta particles are the electrons from the nucleus, the term "beta particle" being an historical term used in the early description of radioactivity.

During the decay, the weak interaction converts a neutron (n) into a proton (p) while emitting an electron (e−) and an antineutrino (v_e):

$n \rightarrow p + e^- + v_e$

A radionuclide emits electrons (positrons) that have continuous spectrum

The radiation hazard from betas is greatest if the beta-emitting radionuclides are ingested or inhaled

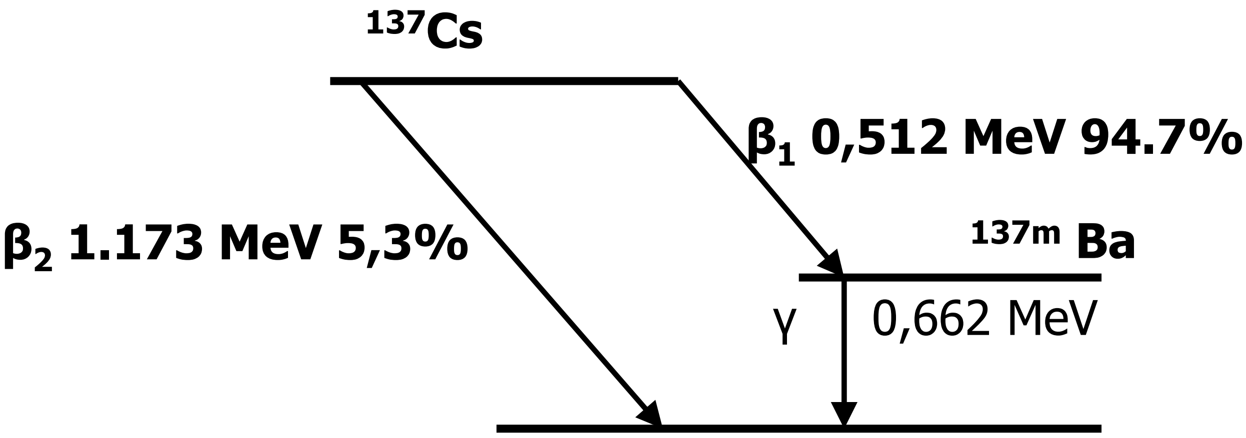


$E_{av} \approx 1/3\ E_{max}$

	E_{av} (keV)	E_{max}(keV)
^{90}Sr	200	544
^{90}Y	931	2245

Gamma ray

A nucleus which is in an excited state may emit one or more photons of discrete energies. The emission of gamma rays does not alter the number of protons or neutrons to the nucleus but instead has the effect of moving the nucleus from a higher to a lower energy state (unstable to stable). Gamma ray emission frequently follows beta decay, alpha decay, and other nuclear decay processes.

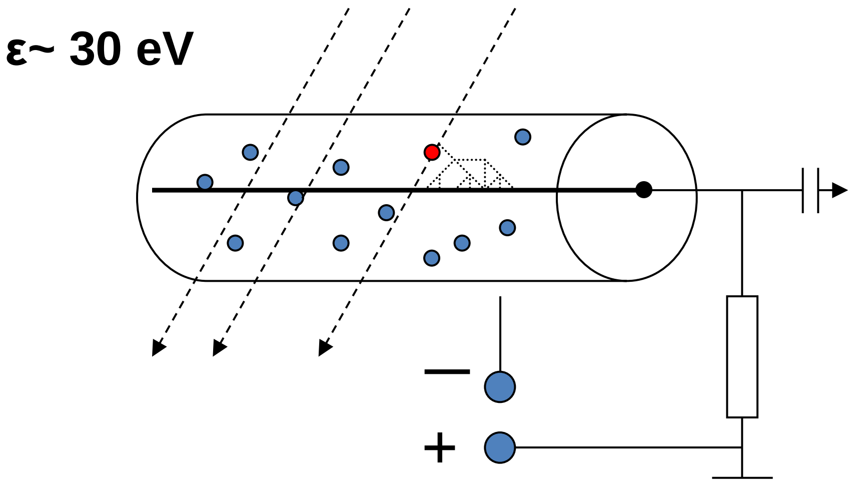


Interaction with matter

- ✓Charged particles and gamma quantum loss their energy on their way in matter due to interaction with atom electrons. Atoms of the absorber are getting ionized (exited).
- ✓The interaction is resulted in positive and negative charge accumulation, photon emissions.
- ✓Values of the accumulated charge or the photons energy sometimes are proportional to energy of an ionization particle or quanta.
- ✓Principles of all radiation detectors are based on secondary effects caused by interaction of irradiation with matter.

Gas filled detectors (GM)

detect the presence and intensity of radiation (particle frequency, rather than energy).



Are used for :

Measurement of gamma and X-ray radiation equivalent dose and dose rate.

Measurement of surface beta-particles flux density.

Solid scintillation detectors: NaI(Tl), CsI (Tl), ZnS

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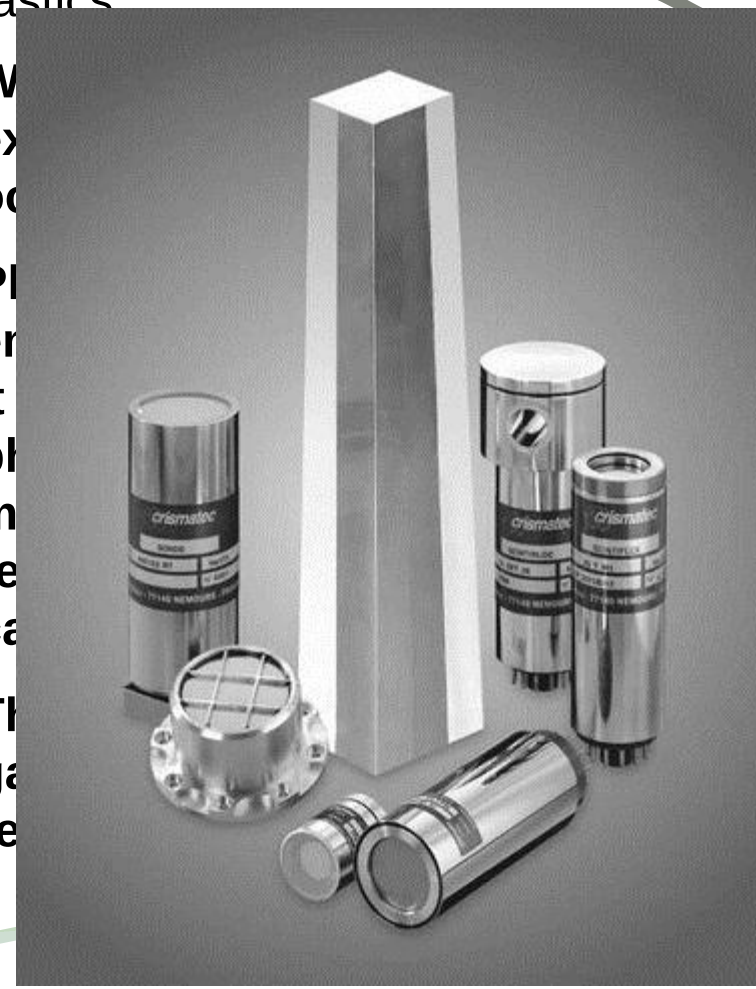
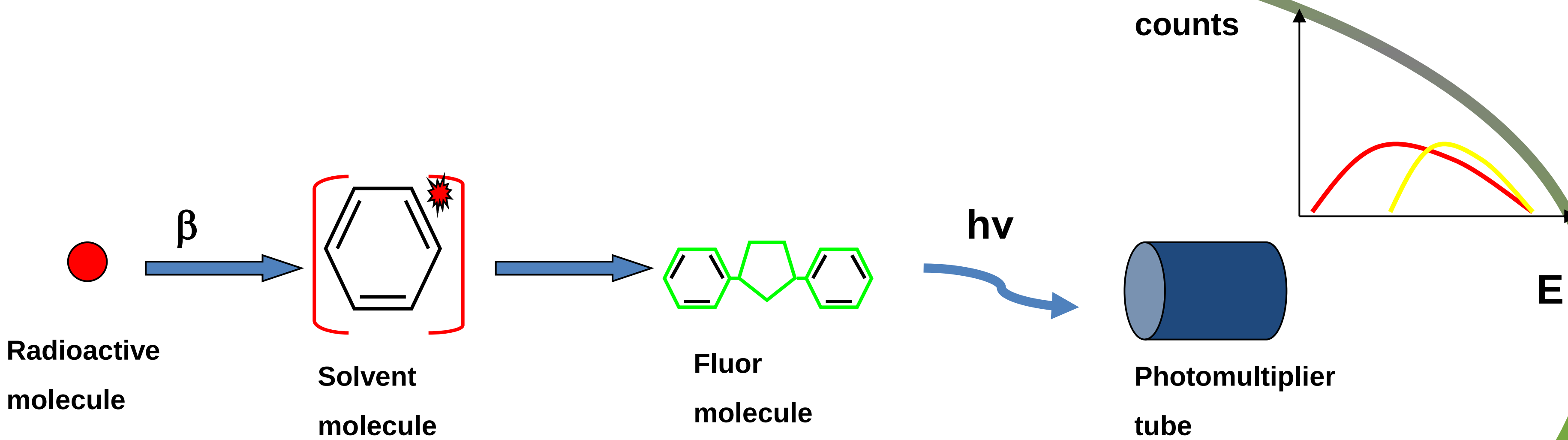


Liquid scintillator



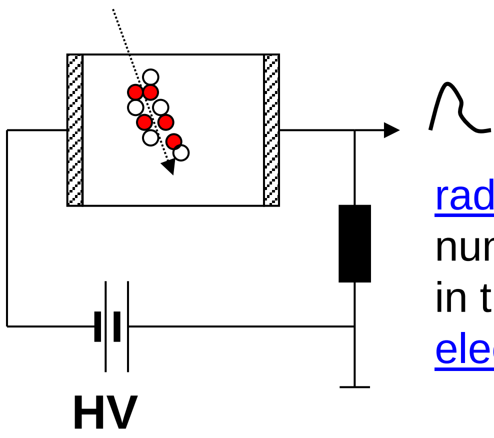
Emitted beta particles excite the solvent molecules

The energy of the solvent molecules is transferred to fluor molecules, which in turn emit light

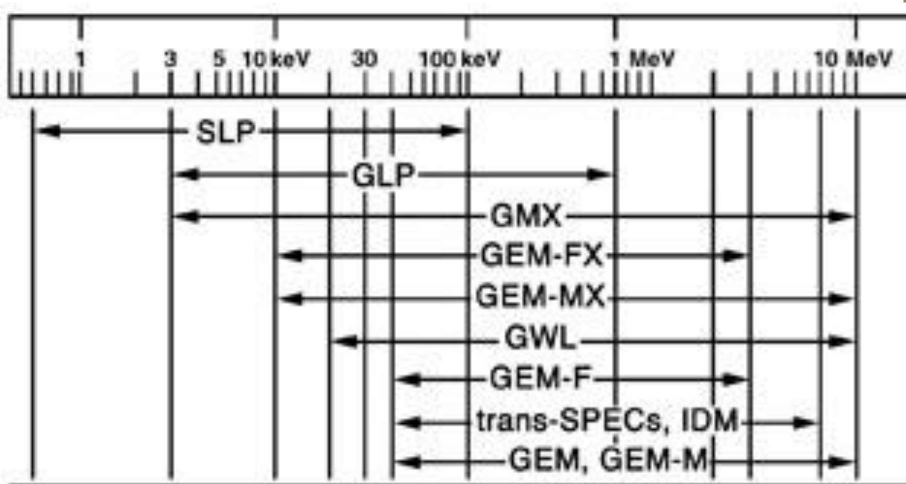
A unique application of liquid scintillators is counting of radioisotopes that emit low-energy beta particles, such as hydrogen-3 (^{3}H) or carbon-14 (^{14}C)

Semiconductor radiation detector

ε~ 3 eV



radiation is measured by means of counting the number of free charge carriers in the detector, which is arranged between two electrodes

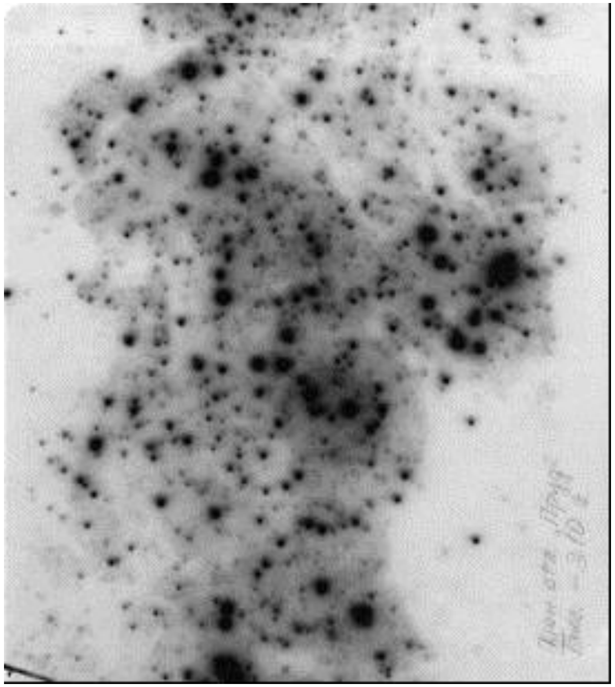


Methods
Detectors

Track detectors

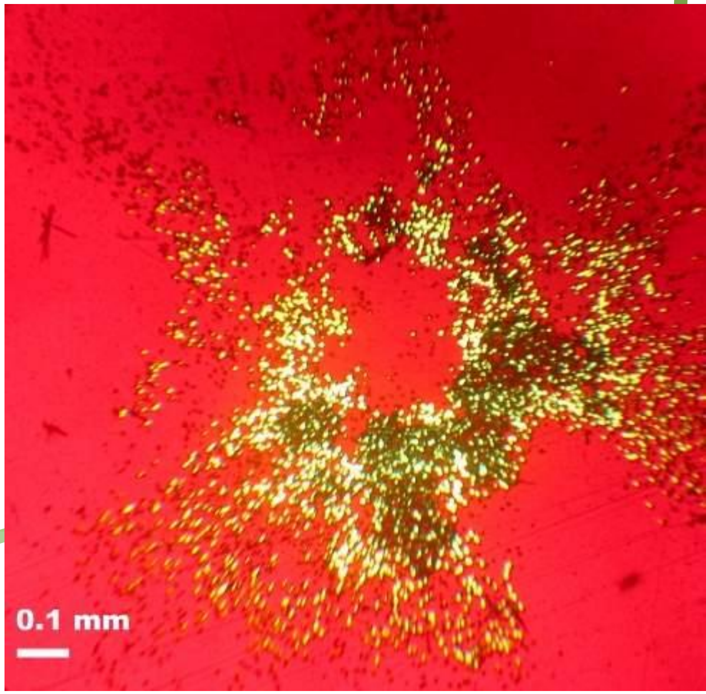
When a charged particle slows down and stops in a solid material, the energy that it deposits along its track can cause permanent damage in the material.

X-ray film



- Exposition
- Development
- Calculation

nuclear film



Analytical radiometric methods

Alpha emitting radionuclides

Digestion (solid)

Preconcentration

- Evaporation (water)
- Co-precipitation (solution, water)
- Solvent extraction

Chemical Separation

Anion-exchange chromatography

Extraction chromatography

Source Preparation

Electrolytic or

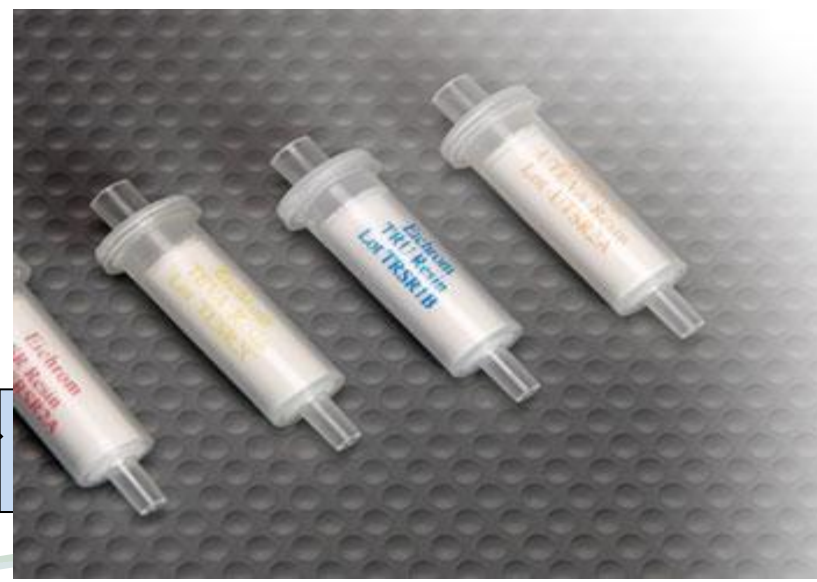
Chemical precipitation

U, Pu, Am, Cm ......

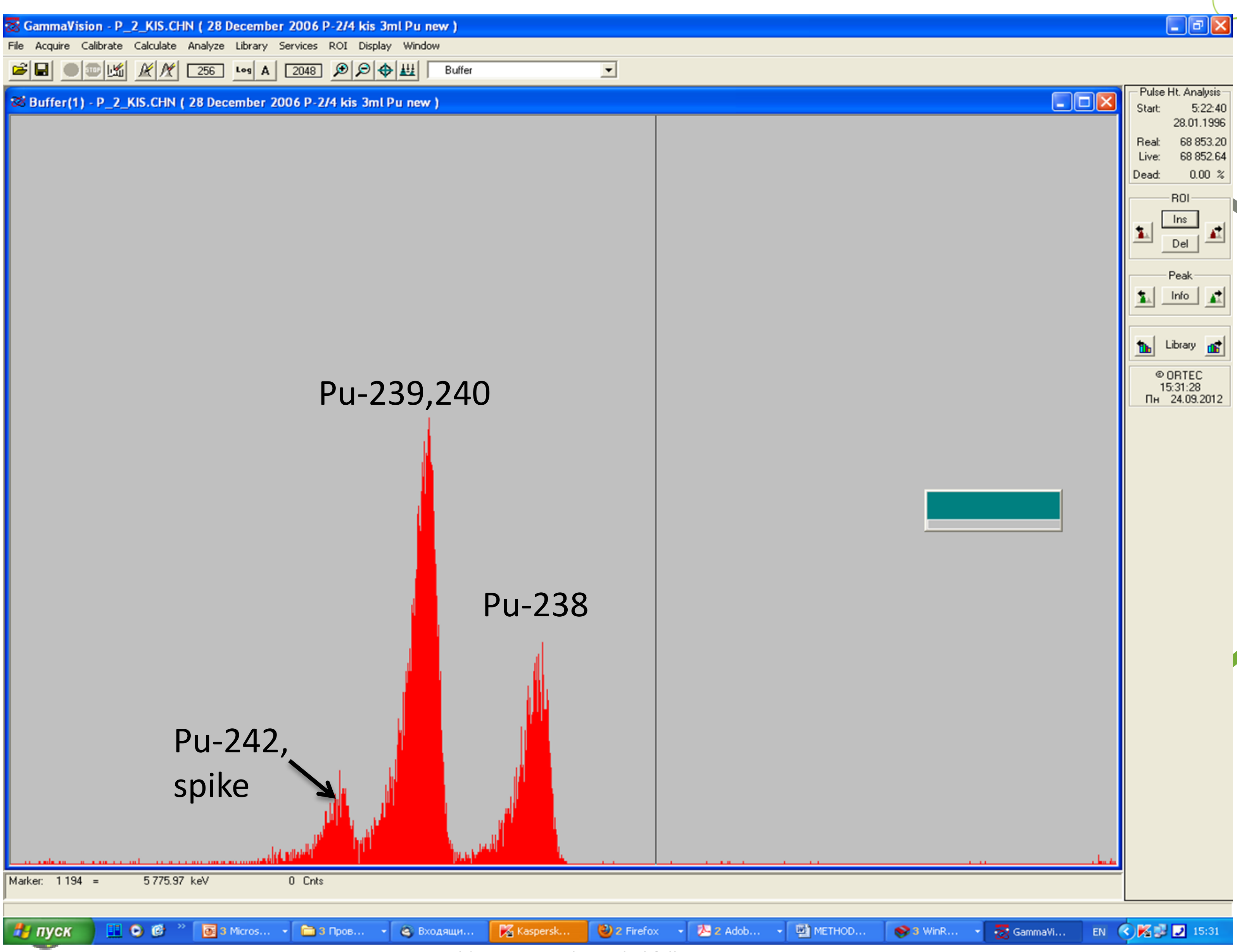
Anion-exchange chr.:

separation RNs in strong acids using anion-exchange resins (anionic nitrate or chloride complexes – $Pu(NO_3)^{2-}{}_6$)

Extraction Chromatography:

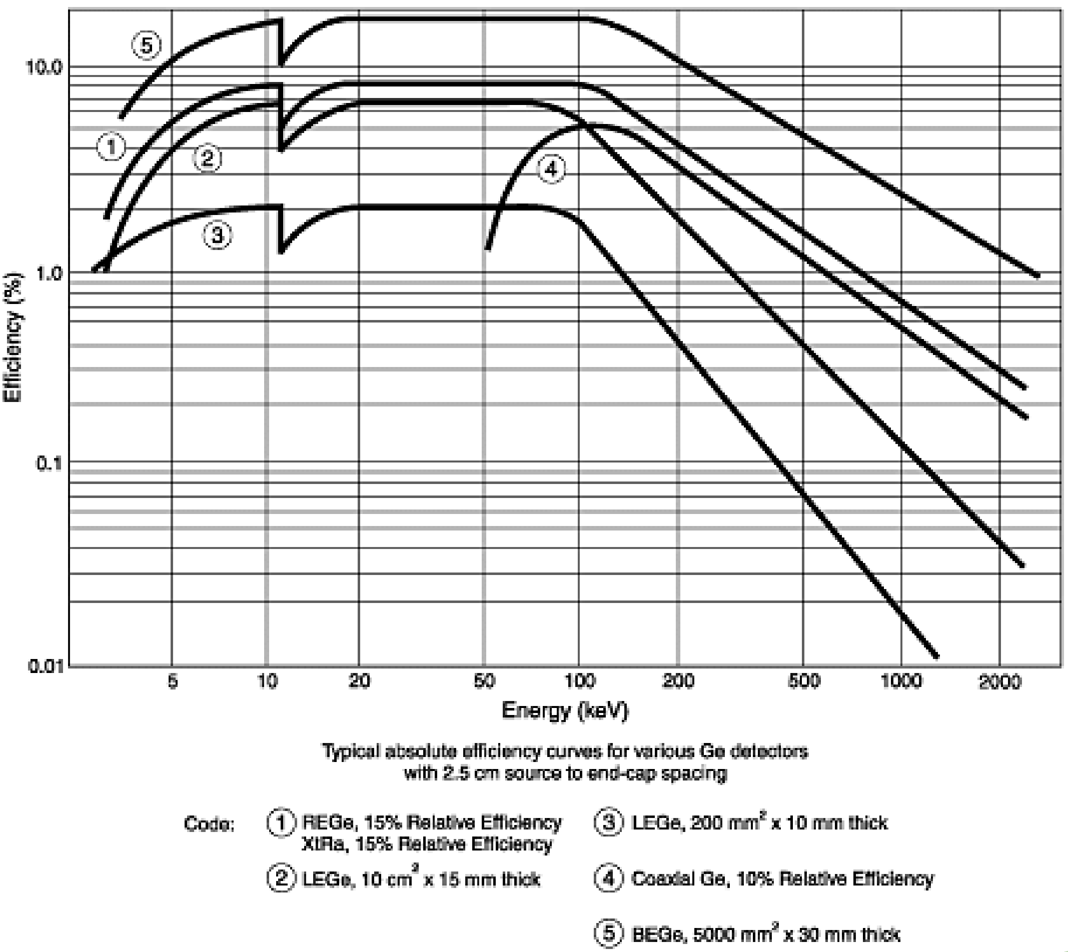


Analytical radiometric methods



Analytical radiometric methods

Gamma-spectrometry



Nuclides

(HPGe)

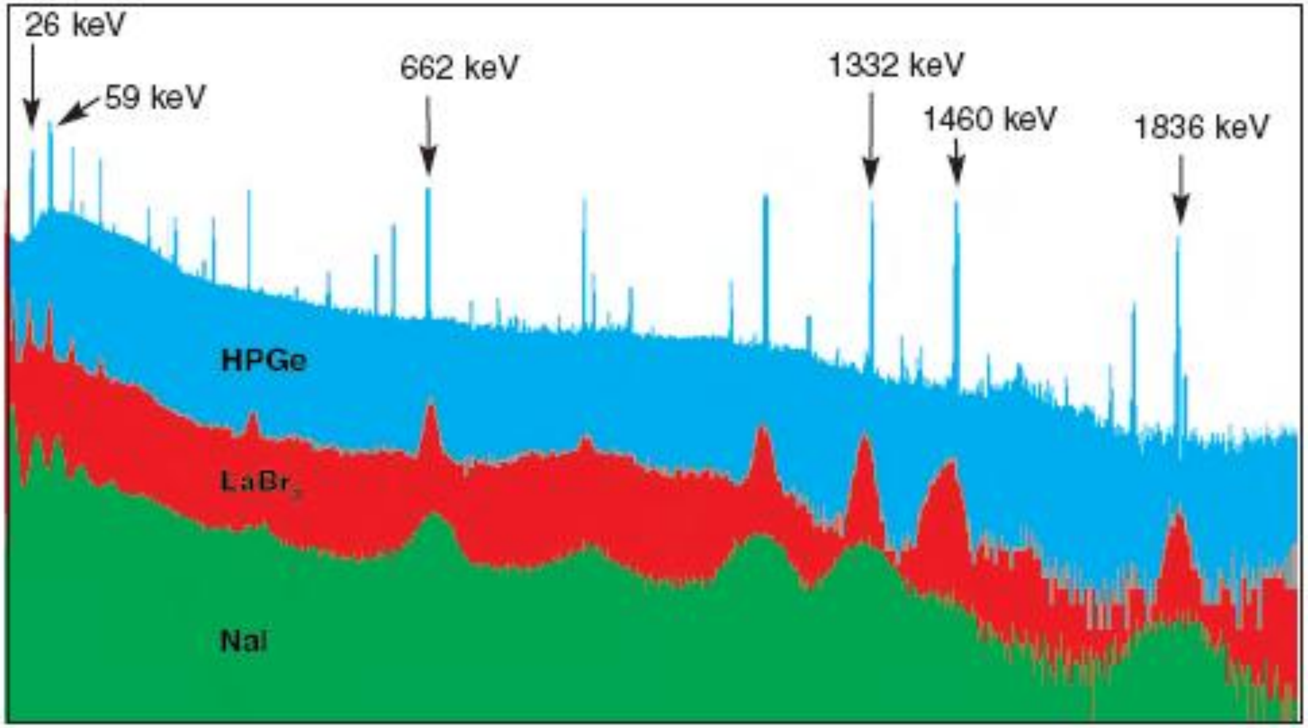
^{241}Am, ^{235}U ^{106}Ru, ^{131}I, ^{60}Co, ^{103}Ru, ^{134}Cs, ^{137}Cs, ^{144}Ce, ^{192}Ir

(HPGe extended energy range)

^{129}I, ^{210}Pb, ^{241}Am, ^{235}U ^{106}Ru, ^{131}I, ^{60}Co, ^{103}Ru, ^{134}Cs, ^{137}Cs, ^{144}Ce, ^{192}Ir

Analytical radiometric methods

Gamma-spectrometry



Advantage:

High energy resolution

Stability

Low MDA

Disadvantage:

Quit expensive

Cooling is needed

Advantage:

Inexpensive

Without cooling

High efficiency

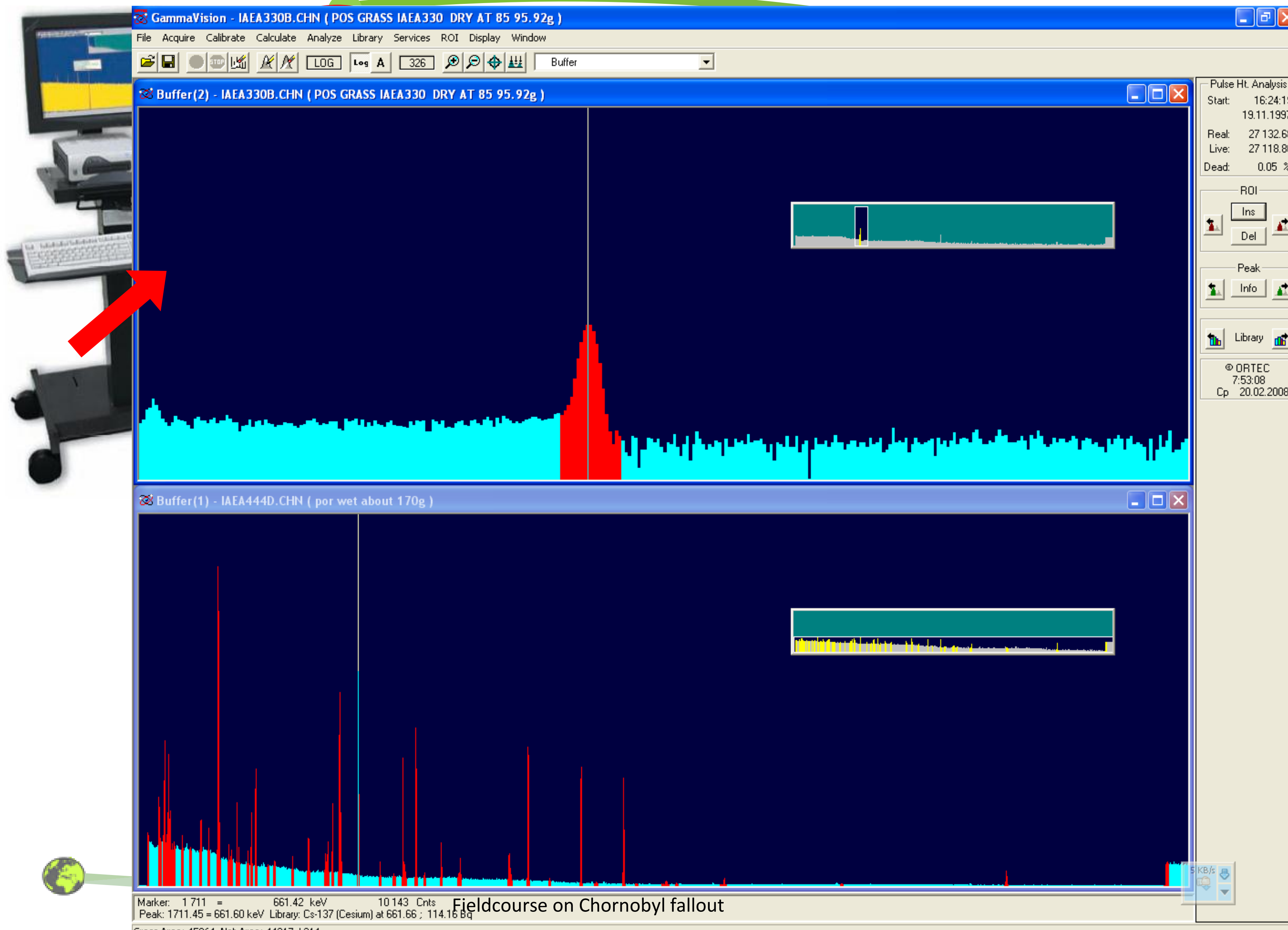
Disadvantage:

Low energy resolution

Higher MDA

Analytical radiometric methods

Gamma spectrometer with HPGe detector (ORTEC,USA)



Analytical radiometric methods

SCINTILLATION BETA-GAMMA-SPECTROMETERS SE-BG-01-“AKP”-150-63

PURPOSE

The spectrometers are designed for determination of qualitative and quantitative composition of radionuclides in samples and are used for determination of specific activity of the wide set of radionuclides (^{137}Cs, ^{90}Sr, Nat-Ra, Nat-Th, ^{40}K and others.)

PERFORMANCE

Gamma

Energy range of gamma-rays, MeV 0.1-3

Max. counting rate, s^{-1} $\leq 10^4$

MDA 1 hour exposition in 1L Marinelly cup, Bq

^{137}Cs	1.2
Nat-Ra	6.0
^{40}K	20
Nat-Th	3.0

Beta

Background intensity in energy range 200 keV-1200 keV, cps. <1.4

Energy range of registered beta-emission, MeV 0.1-3.5

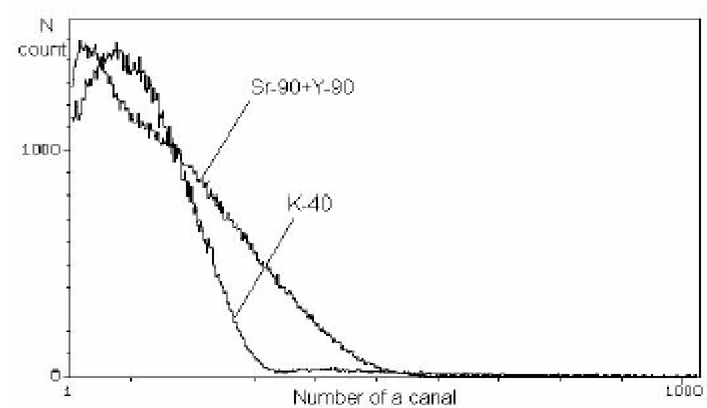
MDA by 2 hours exposition (ρ=1g/ml , Bq/sample)

	160 ml	10 ml
^{90}Sr	0.8	0.25
^{137}Cs	0.9	0.5
^{40}K	1.6	0.4

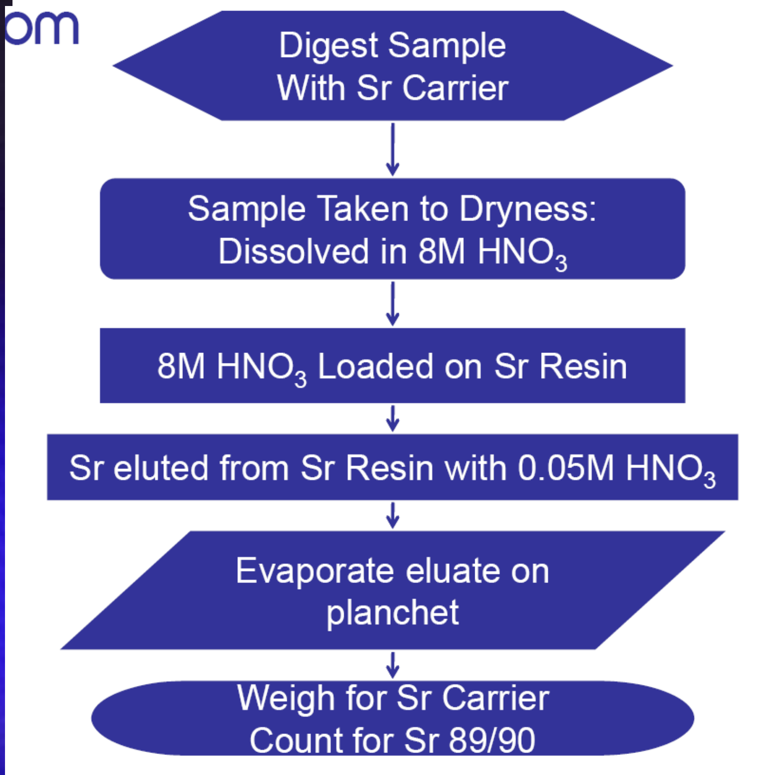
Beta emitting radionuclides

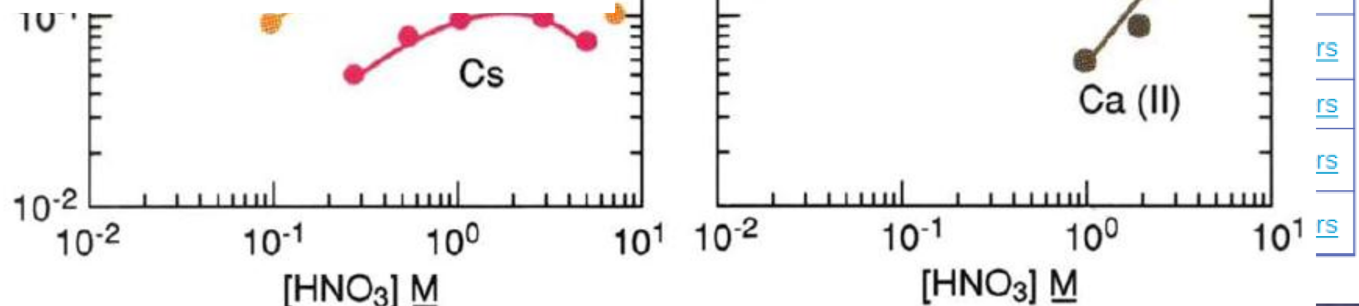
Radiochemical separation is needed in the most cases

Radionuclide	Instrumentation
^{3}H, ^{14}C, ^{99}Tc ^{35}S, ^{89}Sr, ^{90}Sr	Low background beta radiometers or liquid scintillation counting system
^{90}Sr, ^{137}Cs	**Beta spectrometer**

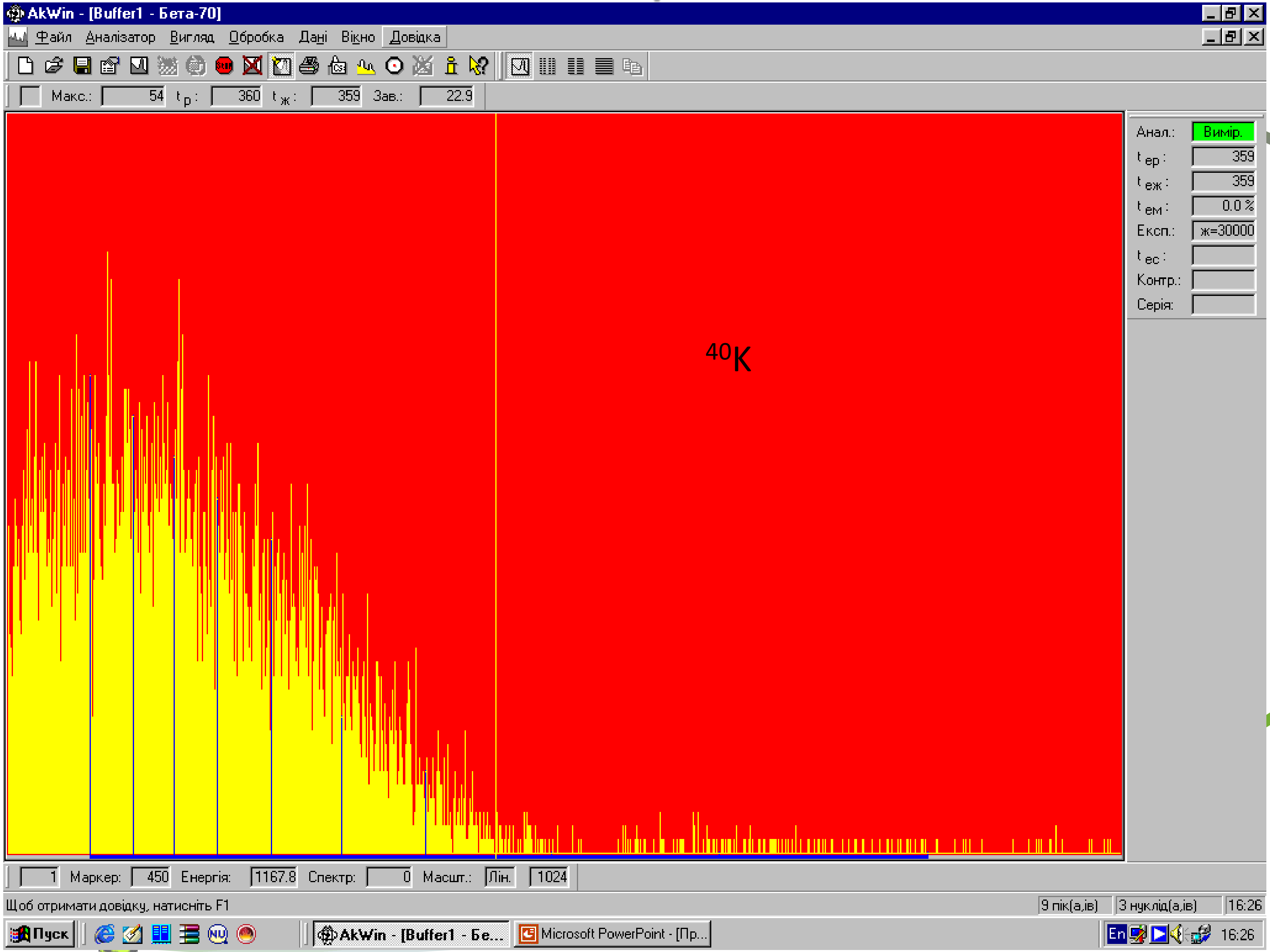


Acid dependency of k' for various ions at 23-25°C.
Sr Resin





Analytical radiometric methods
Beta spectrums



Quality Assurance/Quality Control

QA is an integral part of a laboratory analysis program. It provides a means to deliver valid and traceable results, an ability to identify and correct anomalies, repeatability, assurance that errors will be minimal and stochastic.

ISO/IEC 17025:2005 specifies the general requirements for the competence to carry out tests and/or calibrations, including sampling in following areas:

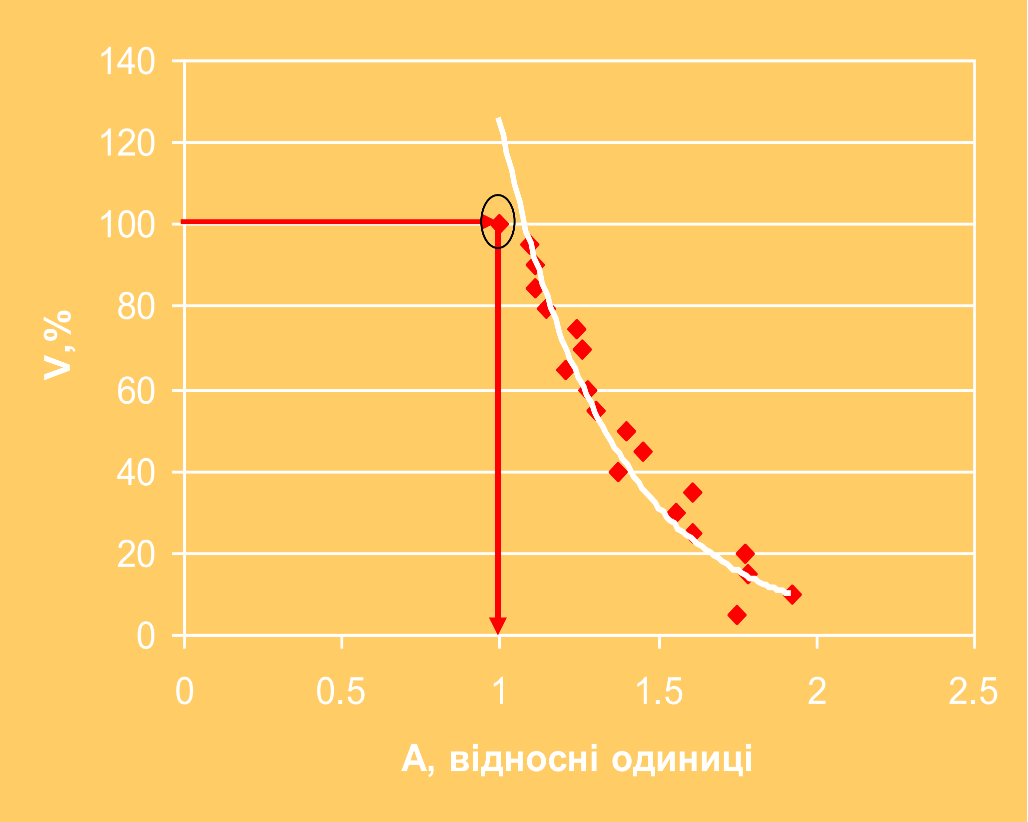
- ✓ Organization and management.
- ✓ Document control.
- ✓ Contracts and procurement.
- ✓ Corrective action.
- ✓ Control of records.
- ✓ Audits.
- ✓ **Personnel qualifications and training.**
- ✓ **Facility infrastructure and security.**
- ✓ **Validation of methods, uncertainty, and measurement traceability.**
- ✓ **Equipment.**
- ✓ **Analytical procedures.**
- ✓ **Calibration and maintenance.**
- ✓ **QC – reference standards, blanks, X-charts, proficiency testing.**
- ✓ **Documentation and reporting.**
- ✓ **Sampling.**

ISO/IEC 17025:2005

General requirements for the competence of testing and calibration laboratories

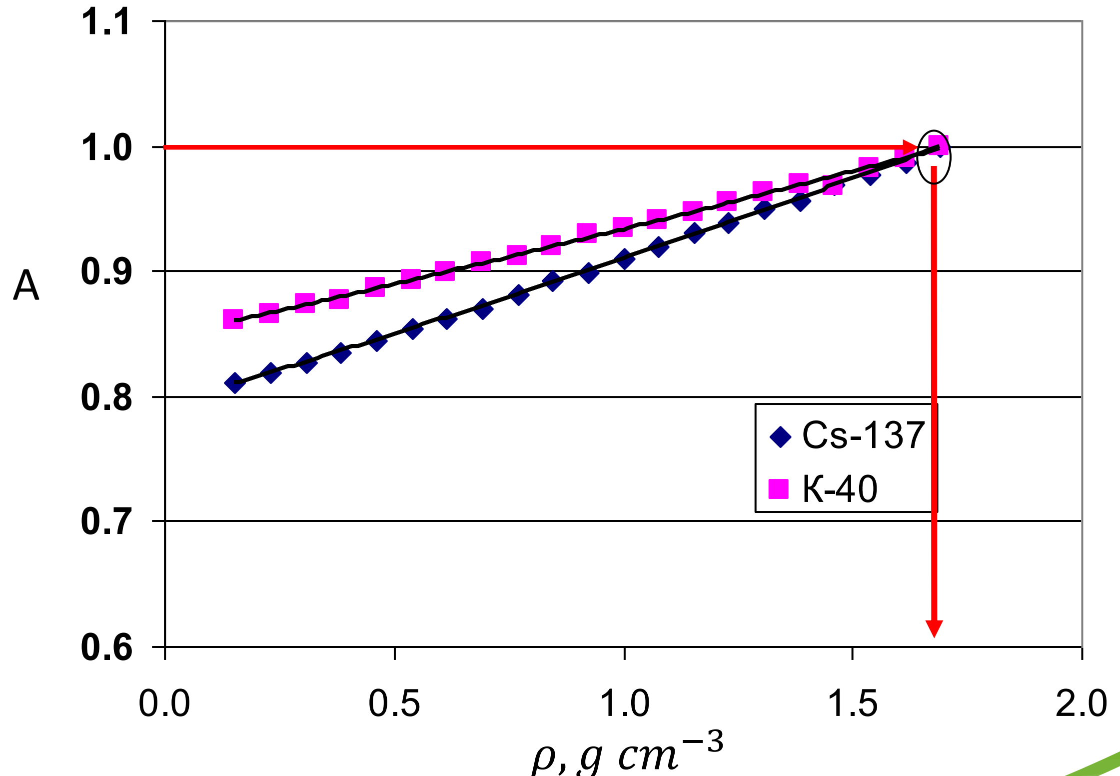
Quality Control

Geometry correction (gamma- spectrometry)
uncertainty sources



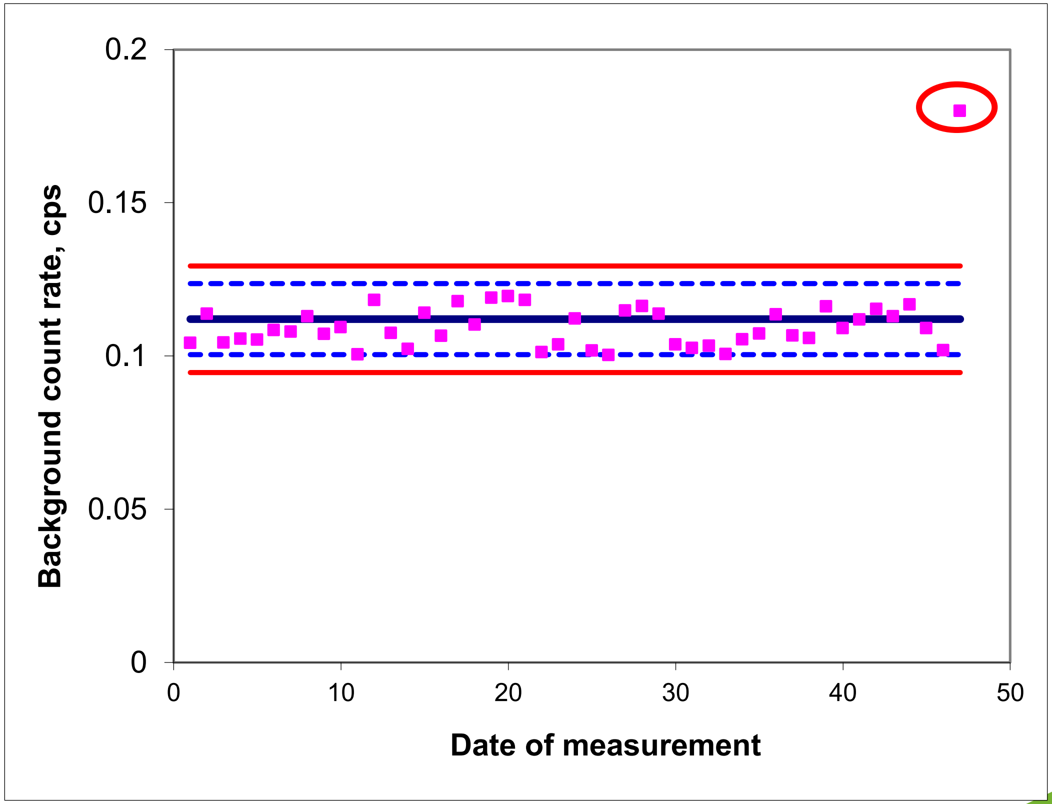
Quality Control

Density correction
uncertainty sources



Quality Control

Quality control X-charts



Quality Control

The sources of uncertainties and their quantification in gamma-spectrometry

Source	Symbol	Range, %	Typical value ,%
Counting	N	0.1-20	5
Emission probability	γ	0.1-11	<2
Self attenuation correction	K_3	0.1-5	<1
True coincidence correction	K_5	1-15	<3
Calibration	ε	1-5	2
Radiochemical procedures		1-10	3
Weight	M	0.01-1	<0.5

Thank you for your attention!!!