Gamma Spectrometry of Naturally Occurring Radioactive Materials (NORM)

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MEASUREMENT OF ACTIVITY IN DECAY CHAIN SERIES

Growth of activity from decay of a parent

- When one radionuclide (the parent P) decays into another radionuclide (the daughter D), the rate of change of the number of daughter atoms must be the difference between the rate of growth from the parent and the rate of decay of the daughter:

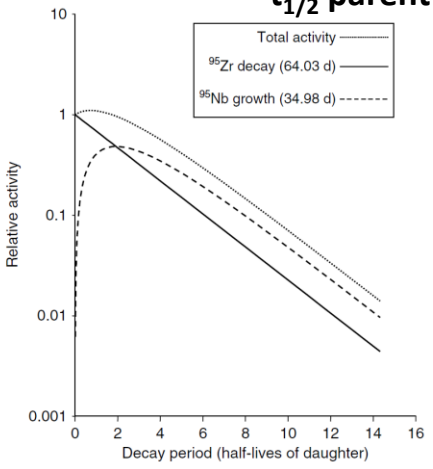
$$\frac{dN_D}{dt} = \lambda_P N_P - \lambda_D N_D = \lambda_P N_{P0} \exp(-\lambda_P t) - \lambda_D N_D$$

- riešenie rovnice:

$$N_D = N_{P0} \cdot \left[\exp(-\lambda_P t) - \exp(-\lambda_D t)\right] \cdot \frac{\lambda_P}{(\lambda_D - \lambda_P)} + N_{D0} \cdot \exp(-\lambda_D t)$$

$$A_D = A_{P0} \cdot \left[\exp(-\lambda_P t) - \exp(-\lambda_D t)\right] \cdot \frac{\lambda_D}{(\lambda_D - \lambda_P)} + A_{D0} \cdot \exp(-\lambda_D t)$$

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



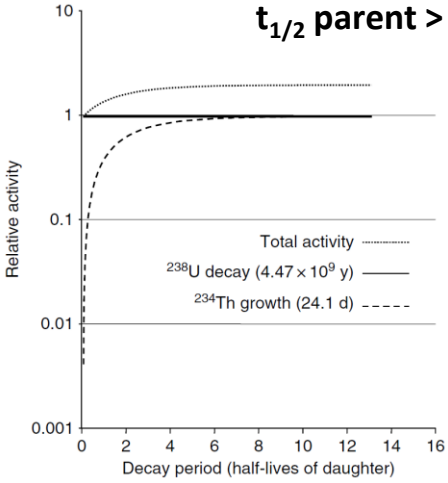
$$A_D = A_P \frac{\lambda_D}{\lambda_D - \lambda_P} = A_P \frac{t_{½P}}{t_{½P} - t_{½D}}$$

In a transient equilibrium, the activity of the daughter nuclide is in **constant ratio** to that of the parent nuclide and apparently decays with the half-life of the parent.

In Figure, we see the decay of the parent unaffected by the absence or presence of daughter and the growth of the daughter activity. (The time scale is in units of half-life of the daughter nuclide.) The total activity in the system is the sum of the parent and daughter activities.

Transient equilibrium is established after about **10 half-lives** of the daughter nuclide after which the daughter apparently decays with the half-life of the parent. As equilibrium is approached, the activity of the daughter nuclide becomes greater than that of the parent.

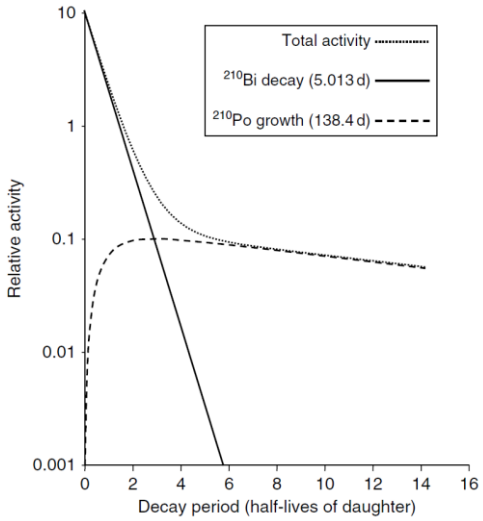
Case 2: Secular equilibrium
$t_{1/2}$ parent > > $t_{1/2}$ daughter



If the half-life of the parent nuclide is very long compared to that of the daughter, the equilibrium state is referred to as secular equilibrium. In such situations, where $t_{1/2D}$ becomes negligible, Equation becomes **A_D=A_P**, i.e. the daughter activity equals the parent activity.

$$A_D = A_P \frac{\lambda_D}{\lambda_D - \lambda_P} = A_P \frac{t_{½P}}{t_{½P} - t_{½D}} = A_P$$

Case 3:No equilibrium
$t_{1/2}$ parent < $t_{1/2}$ daughter

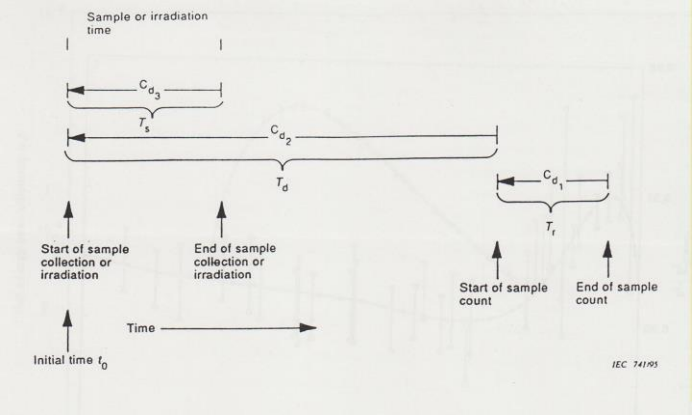


If the daughter half-life is greater than that of the parent then, obviously, the parent will decay, leaving behind the daughter alone. Figure shows the growth of daughter activity within an initially pure parent. No equilibrium is established; ultimately the decay curve will be that of the grown-in daughter.

Radioactive decay correction factors

halflife is in order of sampling or counting time

- **decay during counting interval T_r (real time of count)**
- **decay prior to the counting period T_d,**
- **decay during the sampling collection time T_S or the irradiation T_i (neutron activation analysis)**



$$C_{d1} = \frac{\lambda T_r}{1 - e^{-\lambda T_r}}$$

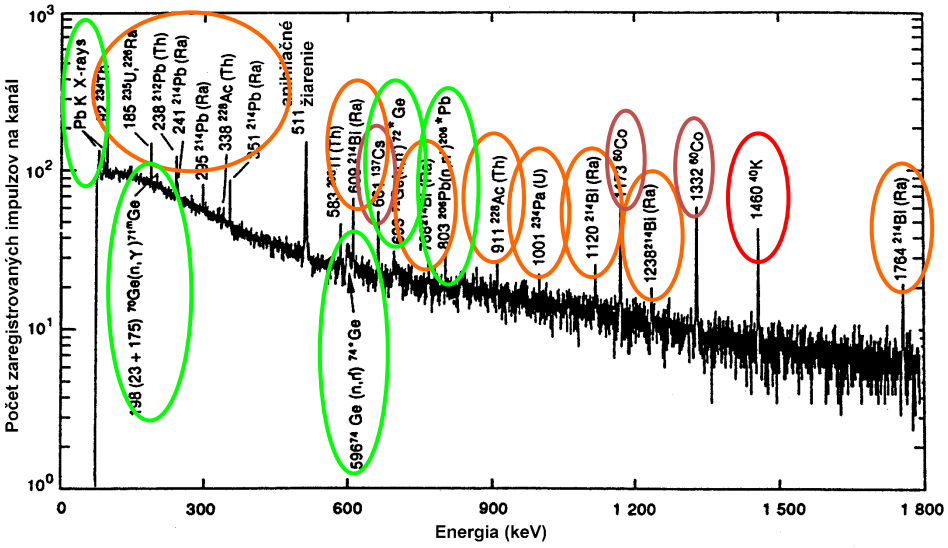
$$C_{d2} = e^{\lambda T_d}$$

$$C_{d3} = \frac{\lambda T_S}{1 - e^{-\lambda T_S}}$$

Environmental materials and background radiation

- The nuclides usually measured by gamma spectrometry are the primordial nuclides: ^{40}**K,** ^{235}**U,** ^{238}**U** and ^{232}**Th**.
- The **uranium** isotopes and ^{232}**Th** will be accompanied by their **daughter** nuclides.
- There are, of course, other naturally occurring nuclides, such as ^{14}**C**, which are produced continuously by nuclear reactions between high-energy particles with oxygen and nitrogen in the earth's atmosphere. Of those, only ^{7}**Be** is measurable by gamma spectrometry.

Meranie pozadia gama spektrometrom
s koaxiálnym detektorom HPGE



What is likely to be observed in background spectrum

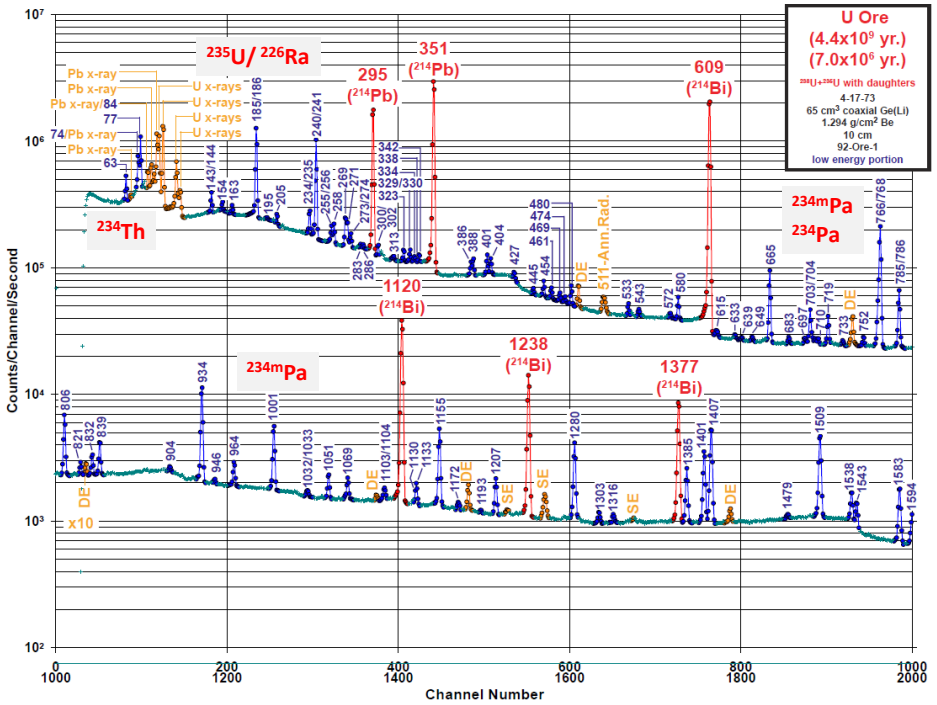
1) The primordial nuclides, ^{40}K, ^{235}U, ^{238}U and ^{232}Th and their daughters.
2) A few common reactor activation products that are often present in background (^{137}Cs).
3) A number of nuclides created by **neutron** reactions with the **detector and shielding materials** – the source of the neutrons involved might be **cosmic or proximity** to a nuclear reactor or accelerator.
4) The major 'fluorescence' **X-rays** from likely shielding materials – Pb, Sn, Cd and Cu.

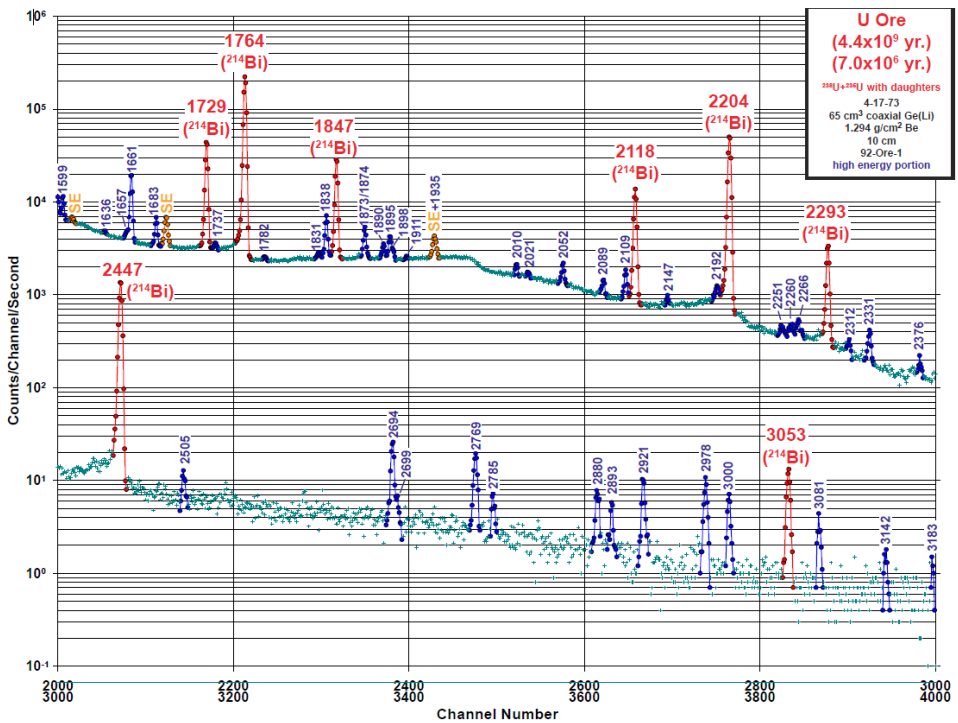
The uranium series – ^{238}U

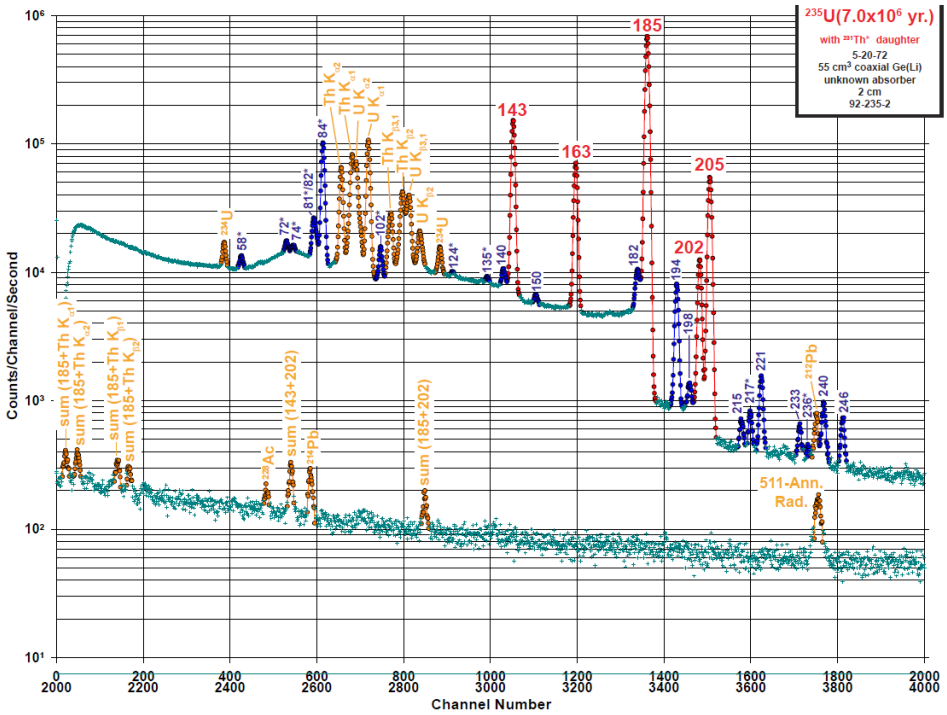
- ^{238}U comprises 99.25% of natural uranium. That decays by alpha emission to ^{234}Th which in turn decays to ^{234m}Pa and so on until stable ^{206}Pb is reached.
- If we look at the half-lives of the various nuclides they are all much less than the half-life of ^{238}U. This means that, in a natural, undisturbed source of uranium, every daughter nuclide will be in secular equilibrium with the ^{238}U. The activity of each daughter nuclide will be equal to the ^{238}U activity.
- There are 14 radionuclides in the chain and so the total activity of such a source will be 14 times that of the parent, or of any individual nuclide.

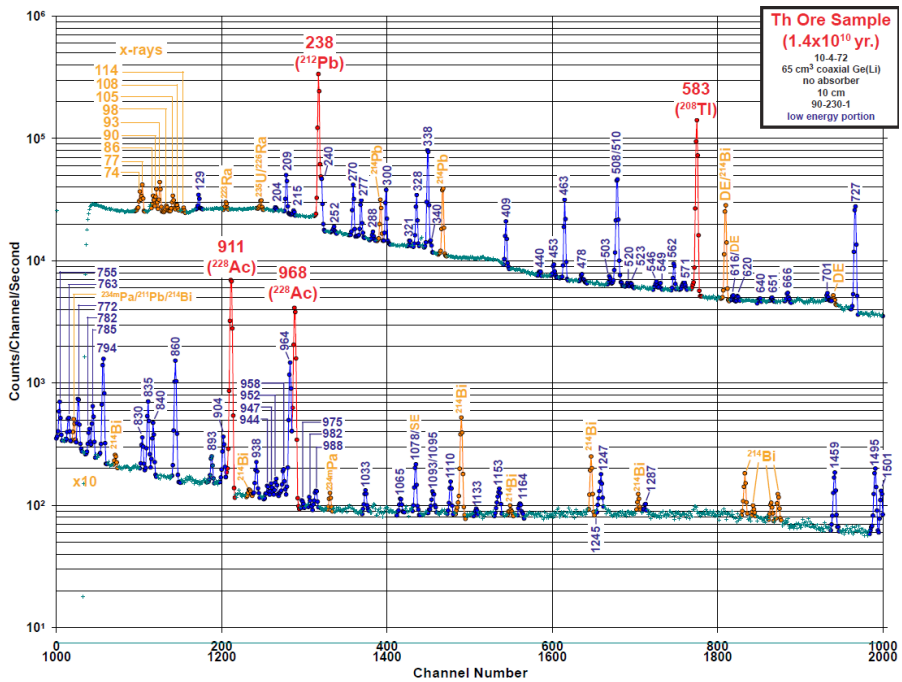
The uranium decay series – ^{238}U
14 decay stages, 8 alpha particles
Nuclides underlined are measurable by gamma spectrometry

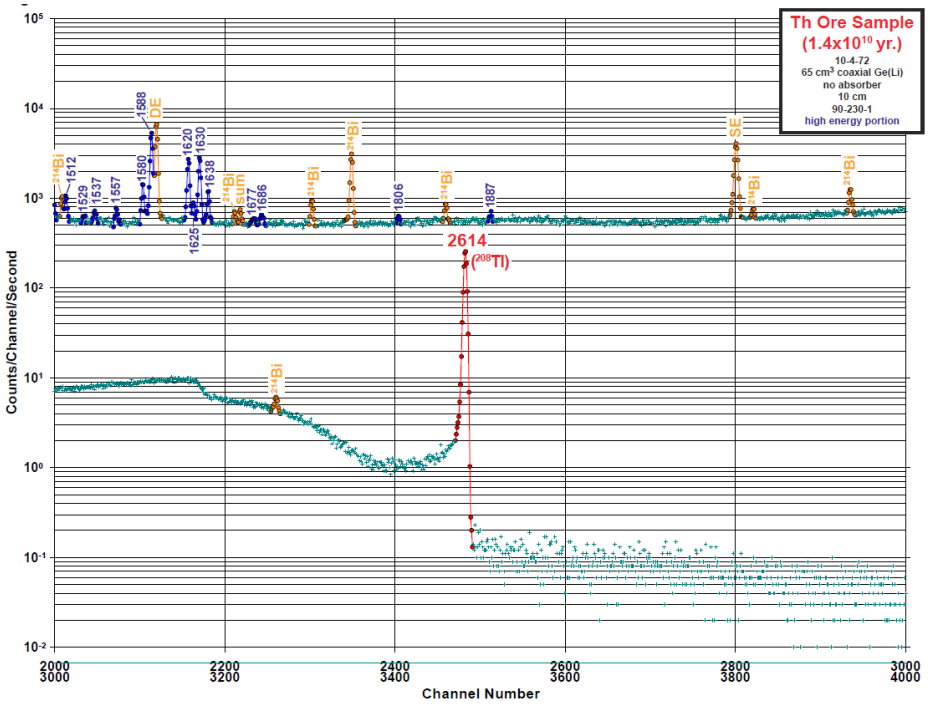
(1) **^{238}U** 4.468×10^9 y
↓α
(2) ^{234}Th 24.10 d
↓β ↓β
(3) ^{234m}Pa 1.17 m
↓IT
^{234}Pa 6.70 h
↓β ↓β
(4) **^{234}U** 2.455×10^5 y
↓α
(5) ^{230}Th 7.538×10^4 y
↓α
(6) ^{226}Ra 1600 y
↓α
(7) ^{222}Rn 3.8232 d
↓α
(8) ^{218}Po 3.094 m
↓α
(9) ^{214}Pb 26.8 m
↓β
(10) ^{214}Bi 19.9 m
↓β
(11) ^{214}Po 162.3 μs
↓α
(12) ^{210}Pb 22.3 y
↓β
(13) ^{210}Bi 5.013 d
↓β
(14) ^{210}Po 138.4 d
↓α
^{206}Pb STABLE

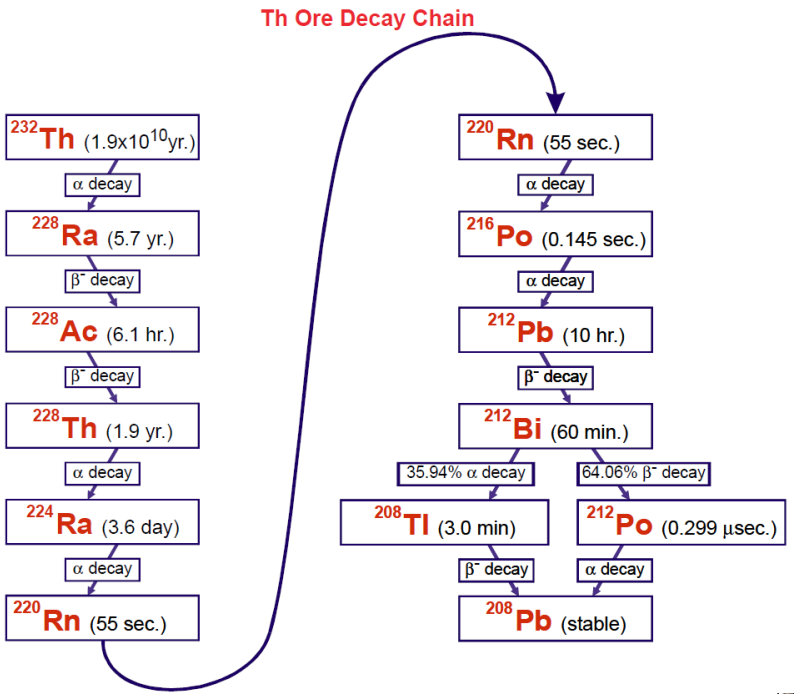

U Ore
(4.4x10⁹ yr.)
(7.0x10⁶ yr.)
²³⁸U+²³⁵U with daughters
4-17-73
65 cm³ coaxial Ge(Li)
1.294 g/cm² Be
10 cm
92-Ore-1
low energy portion
²³⁵U/ ²²⁶Ra
²³⁴Th
²³⁴ᵐPa
²³⁴Pa
351 (²¹⁴Pb)
295 (²¹⁴Pb)
609 (²¹⁴Bi)
1120 (²¹⁴Bi)
1238 (²¹⁴Bi)
1377 (²¹⁴Bi)
Pb x-ray
U x-rays
DE
SE
Sum
511-Ann.Rad.
x10
Counts/Channel/Second
Channel Number


U Ore
(4.4x10⁹ yr.)
(7.0x10⁶ yr.)
²³⁸U+²³⁵U with daughters
4-17-73
65 cm³ coaxial Ge(Li)
1.294 g/cm² Be
10 cm
92-Ore-1
high energy portion
1764 (²¹⁴Bi)
1729 (²¹⁴Bi)
1847 (²¹⁴Bi)
2204 (²¹⁴Bi)
2118 (²¹⁴Bi)
2293 (²¹⁴Bi)
2447 (²¹⁴Bi)
3053 (²¹⁴Bi)
SE
SE+1935
Counts/Channel/Second
Channel Number


^{235}U(7.0x10^{6} yr.)
with ^{231}Th daughter
5-20-72
55 cm^3 coaxial Ge(Li)
unknown absorber
2 cm
92-235-2
185
143
163
205
202
sum (185+Th K$_{\alpha1}$)
sum (185+Th K$_{\alpha2}$)
sum (185+Th K$_{\beta1}$)
sum (185+Th K$_{\beta2}$)
sum (143+202)
sum (185+202)
511-Ann. Rad.
Counts/Channel/Second
Channel Number


Th Ore Sample
(1.4x10^{10} yr.)
10-4-72
65 cm^3 coaxial Ge(Li)
no absorber
10 cm
90-230-1
low energy portion
x-rays
238
(^{212}Pb)
583
(^{208}Tl)
911
(^{228}Ac)
968
(^{228}Ac)
x10
Counts/Channel/Second
Channel Number


Th Ore Sample
(1.4x10^10 yr.)
10-4-72
65 cm^3 coaxial Ge(Li)
no absorber
10 cm
90-230-1
high energy portion
Counts/Channel/Second
Channel Number
2614
(208Tl)
DE
SE


Th Ore Decay Chain
232Th (1.9x10^10yr.)
α decay
228Ra (5.7 yr.)
β- decay
228Ac (6.1 hr.)
β- decay
228Th (1.9 yr.)
α decay
224Ra (3.6 day)
α decay
220Rn (55 sec.)
220Rn (55 sec.)
α decay
216Po (0.145 sec.)
α decay
212Pb (10 hr.)
β- decay
212Bi (60 min.)
35.94% α decay
64.06% β- decay
208Tl (3.0 min)
212Po (0.299 µsec.)
β- decay
α decay
208Pb (stable)

The actinium series – ^{235}U

11 decay stages, 7 alpha particles,
Only ^{235}U is measurable by gamma spectrometry

(1) **^{235}U** 7.04×10^8 y
↓α
(2) ^{231}Th 25.52 h
↓β
(3) ^{231}Pa 3.276×10^4 y
↓α
(4) ^{227}Ac 21.772 y
↓β
(5) ^{227}Th 18.718 d
+ α (1.38 %) to ^{223}Fr 22.00 m, then β
↓α
(6) ^{223}Ra 11.43 d
↓α
(7) ^{219}Rn 3.96 s
↓α
(8) ^{215}Po 1.781 ms
↓α
(9) ^{211}Pb 36.1 m
↓β
(10) ^{211}Bi 2.14 m
↓α
(11) ^{207}Tl 4.77 m
+ β (0.273%) ^{211}Po 516 ms then α
↓β
^{207}Pb STABLE

The thorium series – ^{232}Th

10 decay stages, 6 alpha particles
Nuclides underlined are measurable by gamma spectrometry

(1) ^{232}Th 1.405×10^{10} y
↓α
(2) ^{228}Ra 5.75 y
↓β
(3) ^{228}Ac 6.15 h
↓β
(4) ^{228}Th 1.9127 y
↓α
(5) ^{224}Ra 3.627 d
↓α
(6) ^{220}Rn 55.8 s
↓α
(7) ^{216}Po 150 ms
↓α
(8) ^{212}Pb 10.64 h
↓β
(9) ^{212}Bi 60.54 m
↓β (64.06%) ↓α (35.94%)
(10) ^{212}Po 0.300 μs ^{208}Tl 3.060 m
↓α ↓β
^{208}Pb STABLE

Radon loss

All of the decay series have within them a radon isotope. Radon is a gas. It will normally be trapped within a solid sample but if allowed to escape, for example, by grinding the sample, the **equilibrium** between the **postradon** nuclides, many of which have short half-lives and decay rapidly, **will be lost**.

In principle, this would alter the total activity of the sample and the dose rate from the sample. However, the half-lives of ^{219}Rn, in the actinium series, and ^{220}Rn in the thorium series, are very short and even if radon escapes, **equilibrium will be re-established within minutes**. That is not the case in the uranium series.

^{222}Rn in the ^{238}U decay chain

The seventh item in the ^{238}U decay chain is ^{222}Rn, with a half-life of 3.825 d. After loss of ^{222}Rn, there is ample time for the decay of the daughter nuclides preceding ^{210}Pb before re-growth of the ^{222}Rn.

If, as is often the case, post-radon nuclides were measured to estimate ^{238}U activity, loss of radon would affect the whole activity measurement process. The solution is simple – encapsulate the sample and wait for about **10 half-lives** of the ^{222}Rn to allow **equilibrium** to be re-established – **say one month**.

Having said that, experience shows that it is, in fact, possible to grind some geological materials without apparent loss of radon. However, that cannot be relied upon. Different materials have different radon-emanating powers, which will depend upon the moisture content and other factors.

Natural disturbance of the decay series

In general, if a material of natural origin is examined the expected equilibrium within the decay series will be found. There are occasions, however, when that is not so. **Groundwater** passing through rocks can **dissolve some of the elements** and transport them elsewhere where they may be deposited. For most nuclides, that is not a problem.

Those in the water will quickly decay and within the host rock will be quickly re-established. An exception is ^{210}Pb. Its 22.7 year half-life means that ^{210}Pb could be transported from one place to another, leaving a deficit in the host rock and an excess in the groundwater or at some other place where chemical conditions would cause the ^{210}Pb to be deposited or absorbed.

For such reasons, it is not wise to rely on measurement of ^{210}Pb alone as an estimate of the ^{238}U activity.

Table 16.1 Most significant gamma-rays emitted by the NORM nuclides

Nuclide	Source of P_γ data[a]	Half-life[b]	Gamma-ray energy (keV)[c]	Emission probability, P_γ (%)[d]	Comments
^{7}Be	LARA	53.22 d	**477.60**	10.44 (4)	—
^{40}K	XGAMMA	4.563×10^{11} d	**1460.82**	10.66 (13)	Probable interference from ^{228}Ac
^{235}U Series					
^{235}U	LARA	2.571×10^{11} d	**185.72**	57.2 (8)	44.8 % of composite peak – ^{223}Ra, ^{226}Ra and ^{230}Th interference correction needed
	—	—	**143.76**	10.96 (8)	^{230}Th interference – correction needed
	—	—	**163.33**	5.08 (7)	—
	—	—	205.31	5.01 (7)	Many interferences – not recommended
^{227}Th	DDEP	18.718 d	**235.96**	12.6 (6)	—
	—	—	**256.23**	6.8 (4)	—
^{223}Ra	LARA	11.43 d	**269.46**	13.7 (4)	Interference from ^{228}Ac
^{219}Rn	LARA	3.96 s	**271.23**	10.8 (7)	Interference from ^{228}Ac and ^{223}Ra
	—	—	**401.81**	6.4 (5)	—
^{238}U Series					
^{238}U	DDEP	1.632×10^{12} d	49.55	0.0697 (26)	Unusable – serious ^{227}Th interference

Nuclide	Source of P_γ data[a]	Half-life[b]	Gamma-ray energy (keV)[c]	Emission probability, P_γ (%)[d]	Comments
^{234}Th	LARA	24.10 d	**63.28**	4.8 (6)	—
	—	—	92.37	2.81 (26)	Measured together.
	—	—	92.79	2.77 (26)	Serious interference from ^{228}Ac when present. X-ray interferences.
^{234m}Pa	GRG	1.17 m	**1001.03**	1.021 (15)	No interferences. Slight summing in possible
	—	—	766.37	0.391 (9)	Interferences from ^{214}Pb and ^{211}Pb
	—	—	258.19	0.075 (3)	Serious interference from ^{214}Pb
^{226}Ra	XGAMMA	5.862×10^5 d (1600 y)	**186.21**	3.555 (19)	57.1 % of composite peak – ^{235}Ra and ^{230}Th interference correction needed
^{214}Pb	DDEP	26.8 m	**351.93**	35.60 (7)	—
	—	—	**295.22**	18.414 (36)	Insignificant interference from ^{212}Bi
	—	—	242.00	7.268 (22)	Interference from ^{224}Ra and deconvolution with 238.63 keV of ^{212}Pb needed
^{214}Bi	DDEP	19.9 m	609.31	45.49 (19)	Subject to TCS
	—	—	**1764.49**	15.31 (5)	—
	—	—	1120.29	14.91 (3)	Subject to TCS

Nuclide	Source of P_γ data[a]	Half-life[b]	Gamma-ray energy (keV)[c]	Emission probability, P_γ (%)[d]	Comments
	—	—	1238.11	5.831(14)	Subject to TCS
	—	—	2204.21	4.913 (23)	—
^{210}Pb	LARA	8.14×10^3 d	**46.54**	4.25 (5)	Slight interference from ^{231}Pa
^{232}Th Series					
^{232}Th	LARA	5.13×10^{12} d	63.81	0.27 (2)	Unusable – serious ^{234}Th interference
^{228}Ac	LARA	6.15 h	**911.20**	25.8 (4)	Subject to TCS
	—	—	**968.97**	15.8 (3)	Subject to TCS
	—	—	338.32	11.27 (19)	Subject to TCS – slight interferences from ^{223}Ra and ^{214}Bi
	—	—	964.77	4.99 (9)	Subject to TCS – interference from ^{214}Bi
^{212}Pb	XGAMMA	10.64 h	**238.63**	43.6 (3)	Deconvolution with 242.00 keV of ^{214}Pb needed
	—	—	300.09	3.18 (13)	Subject to TCS – slight interference from ^{231}Pa
^{212}Bi	XGAMMA	60.54 m	727.33	6.74 (12)	Subject to TCS – serious interference from ^{228}Ac
	—	—	**1620.74**	1.51 (3)	—
^{208}Tl	XGAMMA (Corrected for ^{212}Bi branching)	3.060 m	2614.51	99.7 (2)	Subject to TCS
		—	**583.19**	85.0 (5)	Subject to TCS
		—	**860.56**	12.5 (2)	Subject to TCS
		—	510.7	22.6 (2)	Subject to TCS and difficulty resolving from 511 keV annihilation peak

GAMMA SPECTROMETRY OF THE NORM NUCLIDES

The NORM activities being measured are low – environmental levels. The consequences are that the counting samples may be **large** and will almost always be measured **close** to the detector and, therefore, it will be necessary to take account of the natural background to the detector and be aware of the possibility of **true coincidence summing**.

If activities are high and the samples can be measured at some distance from the detector, all of the comments below relating to true coincidence summing can be disregarded, although if high enough, **random summing** may become a consideration.

Normal energy range recommended for NORM measurement: **30 to 2300 keV**.

There are many methods reported in the literature for the measurement of uranium isotopes using low-energy detectors. These methods are more applicable to high-activity samples.

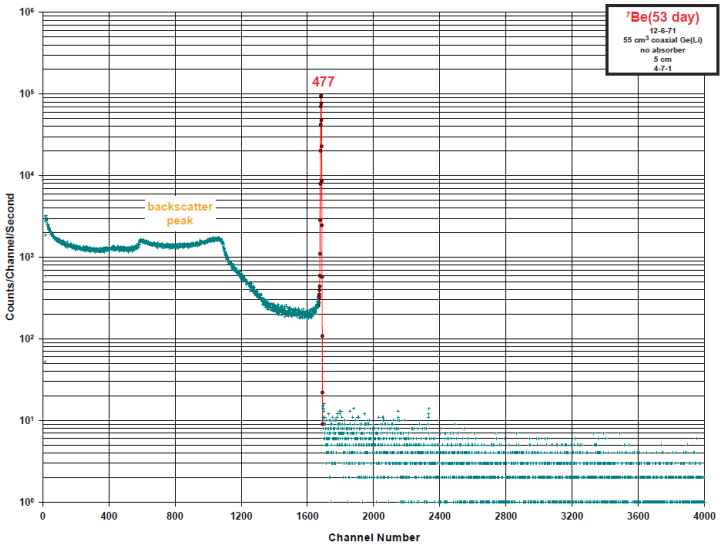
Measurement of ^{7}Be

^{7}Be is continuously generated in the atmosphere by **spallation reactions** of charged particles on oxygen and nitrogen. The nuclide is found in the gamma spectra of some natural waters and on environmental air filters.

Measurement is simple – the only gamma-ray emitted, **477.60 keV**, has no spectral interferences and there is usually no evidence of the nuclide in the background spectrum.

The nuclide has a relatively short half-life and is not supported by decay of a parent; therefore, decay corrections will usually be required to the time of sampling.

Measurement of ^{7}Be



Measurement of ^{40}K

^{40}K is very evident in background spectra. It is present as 0.17% of natural potassium and is present in wood and building materials and even in the bodies of the gamma spectrometrists.

The substantial presence of ^{40}K in the detector background and in many samples, with its long Compton continuum, severely restricts the limit of detection of the many nuclides emitting gamma rays at lower energies.

The gamma spectrometry of ^{40}K is straightforward but peaked-background correction is always necessary. There is a spectral interference from the 1459.91 keV peak of ^{228}Ac, which must be taken into account even when the activity of the 232Th daughters is low and the peak shape is not noticeably affected.

Resolution of the 186 keV peak

Of particular concern in the gamma spectrometry of NORM is the mutual interference between ^{235}U (185.72 keV) and ^{226}Ra (186.21 keV). These peaks are so close together that deconvolution in real environmental spectra is unlikely to give results that one can have confidence in.

In principle, it would be possible to perform a peak stripping operation using other peaks in the ^{235}U spectrum to estimate its contribution to the 186 keV peak. Unfortunately, the emission probability of the next most intense peak at 143.76 keV is only 1/5 of that of the 185.72 keV peak and itself needs correction for spectral interference. Bearing in mind that the peaks in the spectra are of environmental samples and are often of high uncertainty, any attempt at peak stripping gives ^{226}Ra results of very poor quality.

^{226}Ra and ^{235}U measurement

However, if one can be sure that the ^{226}Ra is in radioactive equilibrium with its parent ^{238}U and that the $^{235}U/^{238}U$ isotopic ratio is the expected natural value, the counts in the 186 keV peak can be mathematically apportioned between ^{226}Ra and ^{235}U.

From the specific activity of ^{235}U and ^{238}U (which can be taken from data tables or calculated from the isotope half-lives) and the gamma-ray emission probabilities, it is straightforward to calculate the proportion of counts in the 186 keV peak due to ^{226}Ra and to ^{235}U.

At the same time, it is also possible to make a small correction for an interference due to ^{230}Th. Corrections for ^{230}Th and ^{223}Ra interferences, one should apply to results for ^{235}U calculated from the 143.76 keV peak.

^{226}Ra and ^{235}U measurement

During the spectrum analysis, the 185.72 keV peak should be removed from the ^{235}U entry in the nuclide library. The analysis will then be performed assuming that the entire 186 keV peak is due to ^{226}Ra. We can then use the following correction factors to correct the ^{226}Ra value and derive an additional result for ^{235}U:

Corrected ^{226}Ra = 0,5709×Apparent ^{226}Ra

Estimated ^{235}U = 0,02662×Apparent ^{226}Ra

If, after peaked-background correction, the 186 keV peak area is not significant, an apparent ^{226}Ra upper limit should be calculated. This limit can then be multiplied by the factor 0.06224 (the ratio of the gamma-ray emission probabilities of ^{235}U and ^{226}Ra) to give an estimated upper limit for ^{235}U, which will be lower than that achieved by using the minor peaks of ^{235U}.

Other spectral interferences and summing

To measure **^{214}Bi** a more accurate result will be obtained by using only the 1764.49 keV peak, even though the 609.31 keV gamma ray has a much greater emission probability and none of the other major peaks suffer spectral interference, simply because this is the only usable peak not subject to significant summing.

Similarly, the non-summed 238.63 keV peak of **^{212}Pb** gives a more reliable result than the 300.09 keV peak, and it may be better to use only the non-summed, non-interfered 63.28 keV peak of ^{234}Th rather than risk deconvolution problems with the doublet 92,37+92,79 keV peak.

Other spectral interferences and summing

It is unfortunate that none of the peaks of ^{228}**Ac** or of ^{208}**Tl** are free of summing although there is scope for electing to measure only the one or two least-summed peaks. The major gamma-ray of ^{208}Tl, **2614.51 keV**, is a consequence of a final transition to the ground state. All other nuclear de-excitations pass through that level and, inevitably, the summing errors on the measurement of that gamma-ray are very large.

There would be little point in extending the energy scale of one's gamma spectrometer in order to measure the **2614.51 keV** gamma ray, in spite of its high emission probability.

Another problem peak in the gamma spectrometry of ^{208}**Tl** is that at 510.7 keV. This would need deconvolution from the 511.0 keV **annihilation peak** present in the background. Because of the Doppler broadening of the annihilation peak, the spectrum analysis programs available are unlikely to be able to do that reliably and the peak is best ignored.

CTBTO - Comprehensive Nuclear Test Ban Treaty Organization

Nuclides released in a nuclear explosion

The radioactive nuclides produced by detonation of a **nuclear weapon** are many and varied. In addition to the fission product nuclides, which we would expect, there are many nuclides created by activation of the components of the device itself and residues of the fuel.

The serious potential international repercussions of falsely claiming a test has occurred are such that in monitoring these nuclides care has to be taken that false positives are avoided and that only nuclides with real relevance are taken into account.

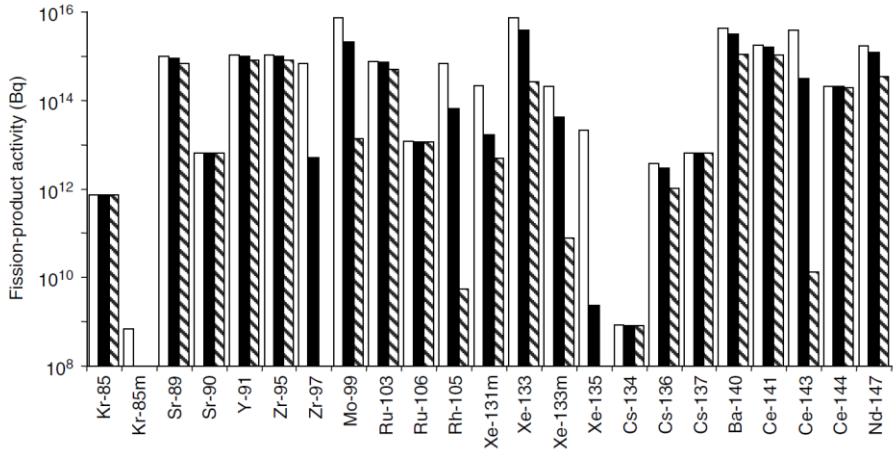
In principle, **measurement** of the pattern of nuclides released can provide information about the **type of nuclear device** and whether its detonation was above or below ground.

The CTBTO has produced a list of 47 relevant fission products, of which 28 have been detected in past monitoring exercises, and 45 relevant non-fission product nuclides, of which 17 have been previously detected. In this context, 'relevant' means that the nuclide would be produced in sufficient amount and its half-life is long enough to give a reasonable chance of detection some days after the test.

Nuclides released by a nuclear explosion

- Residues of fuel materials – ^{3}H, ^{6}Li, ^{7}Li, ^{232}U, ^{233}U, ^{234}U, ^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{241}Am.
- Non-fission product reactions of fuel materials – transuranics caused by neutron capture and subsequent decay within the fuel.
- Fission products – very many!
- Activation products:
  - of non-fuel materials, i.e. of the **constructional** materials;
  - in the **rocks** surrounding an underground explosion;
  - in the **ground** below a near-surface explosion;
  - in **sea** water around or near a sea or sea–surface explosion;

Activity of 24 fission product nuclides, created by a 1 kt nuclear explosion, after 5, 10 and 30 days.



1 kt (kiloton, non- SI, but the conventional unit), which is taken to be a $1{,}4\times10^{23}$ fissions per event (1 kt is 1/10 of the size of the bomb dropped on Hiroshima in 1945).

RASA – Radionuclide Aerosol Sampler/Analyser

Taking into account dilution of the volatilized fission products before reaching it, the monitoring station has to have the capability of measuring down to 10 μBqm^{-3} of ^{140}Ba. The measurement system is called RASA – Radionuclide Aerosol Sampler/Analyser. Detectors of at least 40% relative efficiency are specified to allow the required limit to be achieved in a reasonable time. Direct calibration over the range 88 to 1836 keV using spiked filter standards is used. Monte Carlo and other mathematical methods are not used. The measurement scheme is:

- Collect sample on air filter, collection efficiency 80% for 0,2 µm particles, for 24 h at a flow rate of at least 500 $m^3\ h^{-1}$.
- Allow a decay of no more that 24 h to allow short-lived natural nuclides (radon daughters) to decay.
- Measure the gamma spectrum for at least 20 h.
- Analyse the spectrum and report results within 72 h.

A Typical Radionuclide Monitoring Station Process

The Radionuclide Monitoring process involves collecting particulate matter from the air onto a piece of filter material in a high volume air sampler for ~24 hours.

After this time the filter is taken from the air sampler, compressed into a disk.

The disk is then placed in a chamber to allow natural radionuclides to decay for ~24 hours.

Finally, the filter sample is placed on a gamma detector

The data relating to the sampling conditions and radionuclides measured is then forwarded by satellite to the International Data Centre in Vienna where it is compiled and released to Countries participating in the Treaty.

Noble gas monitoring

The noble gas analysis system is called ARSA, the Automated Radioxenon Sampler/Analyser. This uses two different detection techniques, β-γ coincidence or highresolution gamma spectrometry. The nuclides sought are ^{131m}Xr, ^{133}Xe, ^{133m}Xe and ^{135}Xe, with a limit of measurement of 1 mBqm^{-3} for ^{133}Xe. The measurement scheme is as follows:

- Collect a sample of about 10 m^3 of air at a flow rate of 0,4 m^{3}h^{-1} over a period of 24 h.
- After removal of water and CO_2, xenon is absorbed on cold charcoal, purified and concentrated.
- Radon is removed by gas chromatography.
- Measurement by gamma spectrometry or β-γ coincidence over 24 h.
- Report results within 48 h.

Measurement of transuranic nuclides

A small proportion of thermal neutron interactions with ^{235}U generate ^{236}U, rather than induce a fission. This nuclide has a half-life of 2,34×10^7 years and can indulge in further thermal neutron capture to ^{237}U. That decays to ^{237}Np.

^{238}U (n,γ) ^{239}Uβ^- ––→ ^{239}Np β^- ––→ ^{239}Pu (n,γ) ^{240}Pu (n,γ) ^{241}Pu etc.

All of the plutonium isotopes, with one exception, decay by alpha particle emission.

The exception – ^{241}Pu betadecays to ^{241}Am. It follows that uranium which has been reprocessed, and materials contaminated by it, may contain all of these transuranic nuclides. Because of their low gamma-ray emission probabilities, low levels of the plutonium isotopes are not easily measured by gamma spectrometry.

Measurement of transuranic nuclides

Of the transuranic nuclides, only ^{241}Am, using its 59.54 keV gamma-ray, and ^{237}Np, 86.50 keV, are normally determined by gamma spectrometry.

^{237}Np also emits a gamma-ray at 29.37 keV but this is in an inconvenient region of the spectrum and is subject to serious self-absorption in most samples.

Even the 86.50 keV gamma-ray is likely to be affected by adjacent X-rays and other interfering gamma-rays. ^{237}Np is usually in radioactive equilibrium with its daughter, 27.0 d ^{233}Pa.

If equilibrium is assured, a more reliable measurement of ^{237}Np can be made by using the 311.90 keV gamma ray of ^{233}Pa. This gamma-ray is interference-free, less subject to self-absorption and provides a lower MDA. However, dissolving or leaching a sample will almost certainly destroy equilibrium between parent and daughter because 233Pa, which is carrier-free, readily absorbs on any available surface.

Waste drum scanning

Enrichment meters

Isotopic measurements of uranium alone are relatively easy. ^{235}U emits three gamma-rays with reasonable probability – 187.72, 143.76 and 163.33 keV – in decreasing order of probability.

^{238}U emits a gamma-ray of 49.55 keV with very low probability that is not easily usable, but its daughters, ^{234}Th and ^{234m}Pa, provide gamma rays with much greater emission probability. (It should be remembered that the long half-life of ^{234}U blocks the growth of any nuclides beyond it.)

At 63.28 keV, and a doublet at 92,37 + 92,79 keV, the ^{234}Th is not easily measured, except by high resolution spectrometry, and in the absence of Pu isotopes. ^{234m}Pa has two gamma-rays at 766.37 and 1001.03 keV, which have low emission probabilities but are measurable.

Enrichment meters

Instruments called 'Enrichment Monitors', which can incorporate low or high resolution detectors, make use of two regions-of-interest in the spectrum. One is centred on the 185.72 keV peak, in which counts are mainly due to ^{235}U, and one at some distance above to measure a portion of the Compton continuum created by the 1001.03 keV gamma-ray of ^{234m}Pa, which is indicative of ^{238}U. This forms the basis of a simple system easily understood that can use a low cost, low resolution detector. Recently, when the introduction of portable cooling systems was introduced, hand-held enrichment meters became feasible. Limitations of simple enrichment monitors are that they are only relevant to uranium which is in equilibrium with its daughters and samples must be of 'infinite thickness'. By that, is meant that the sample is so massive that all of the gamma-rays from the parts of the sample distant from the detector will be absorbed within the sample. Making the sample larger would have no effect on the spectrum. This avoids inaccuracies due to unknown self-absorption factors.

Special nuclear materials (SNMs) monitoring

- **Portal monitoring**, for personnel and vehicles, at exits from **nuclear facilities** to prevent removal, accidental or deliberate, of SNMs.
- Process **control** of reprocessing and fabrication.
- Routine **monitoring** inside plants for **inventory** purposes.
- Quality control checking of product.
- Monitoring of **plant** to confirm that no material is being unexpectedly held up within it.
- Monitoring of **waste** material before disposal.
- From an off-site perspective, monitoring of packages and premises suspected of holding SNMs.
- Monitoring at **international borders** to control international trafficking of SNMs.

Emission of spontaneous fission neutrons by uranium and plutonium isotopes

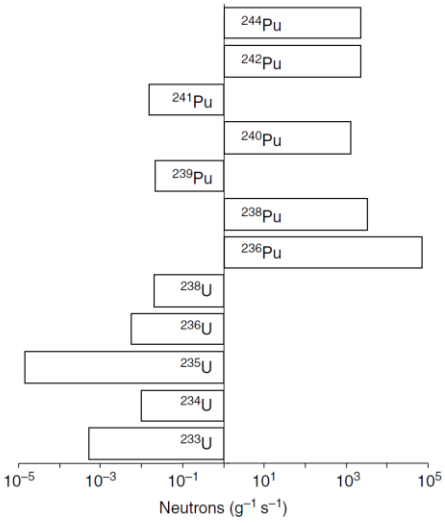


Figure shows the neutron emission rate from spontaneous fission for a number of relevant nuclides. It shows quite clearly that if a significant neutron emission is detected then one or more of the even numbered **plutonium** isotopes are present. In safeguards applications, neutron detector systems will often be combined with a gamma spectrometer.

The spontaneous emission rate of ^{235}U is not large enough to allow its measurement by passive neutron measurements, but active systems, in which neutrons from an **isotope source or neutron generator** are use to stimulate fission in the sample, are available.