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**Determination of the activity of radionuclides
contained in volume samples**

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1. Measurement principle

1.1 Samples characteristics

The measurement must be performed for two types of solid samples : ash and clinker with the following characteristics :

Sample	Reference	Mass (g)	Density
Ash	N° 26390	13,5	0,29
Clinker	N° 26858	49,3	0,97

Elemental composition of each sample were determined by X-ray fluorescence and main elements (mass % > 1) are : O, Mg, Si, P, S, Na, K, Ca, Fe, Cu, Zn, Sb, Pb, Ti, Al, Cl. Radioactive elements were included in both samples ; estimated activities are given in table 1 :

Element	Ash		Clinker	
	Activity (kBq)	Relative uncertainty (%)	Activity (kBq)	Relative uncertainty (%)
⁵⁵ Fe	85,5	20	35,71	20
⁶³ Ni	11,82	20	5,27	20
¹⁴ C	0,5	20	0,25	20
³ H	0,63	20	< 0,5	-
²³⁹⁻²⁴⁰⁻²⁴² Pu	35,1	50	7,65	15
²³⁸ U	-	-	0,015	60
²⁴² Cm	-	-	0,01	40
²⁴⁴ Cm	-	-	0,05	40
²³³ U- ²³⁷ Np- ²³⁴ U	-	-	0,02	60
²³² U- ²⁴¹ Am- ²³⁸ Pu	1700	50	3,06	15
⁵¹ Cr	2,07	37	-	-
⁵⁴ Mn	1,7	10	2,12	10
⁵⁷ Co	-	-	0,1	13
⁵⁸ Co	8,76	5	3,45	10

⁶⁰ Co	32,76	6	25,21	10
⁹⁵ Zr	-	-	0,25	15
⁹⁵ Nb	-	-	0,42	22
¹⁰⁸ Ag ^m	0,21	25	0,05	24
¹¹⁰ Ag ^m	6,15	3	1,92	10
¹²⁴ Sb	1,18	20	0,06	35
¹²⁵ Sb	10	35	0,39	14
¹³⁴ Cs	8,62	38	0,27	12
¹³⁷ Cs	12,66	6	2,86	10

Table 1 : Estimated activities included in the samples

These samples are conditioned in 60 ml polypropylene [(CH₂-CH-CH₃)_n -density = 0,90 (2)] cylindrical containers. These containers have the following geometrical characteristics : internal diameter = 44,7 (2) mm , external diameter = 46,7 (2) mm, mass = 22,2 (1) g, filling height = 30 (1) mm.

Non radioactive samples and empty containers have also been supplied to perform the required absorption measurements.

1.2 Experimental procedure

The activity measurement concerns 13 gamma-emitting nuclides included in each sample, corresponding to energies ranging from 100 keV to 2 MeV. The measurement is performed by gamma-ray spectrometry. However, for these specific geometries (volume and matrix), there is no efficiency calibration available. Thus, it is necessary to compute the efficiency transfer between the reference calibration and the measurement conditions using the ETNA software [1]. The correct use of the software requires :

- the optimization of the geometrical parameters of the detector to get reliable results.
- the measurement of attenuation coefficients for both matrixes over the whole interest energy range.

2. Reference Calibrations

2.1. Detector characteristics

All the measurements are performed using a high-purity germanium (HPGe) coaxial detector (G1). The detector's characteristics as given by the detector's supplier are displayed in table 2.

Crystal diameter	48,0 mm	
Crystal length	52,7 mm	
End cap to crystal	3 mm	
Absorbing layers	0,5 mm of beryllium	
Inactive germanium	0,3 μm	
	warranted	measured
Resolution (FWHM) at 1,33 MeV, ^{60}Co	1,80 keV	1,73 keV
Relative efficiency at 1,33 MeV, ^{60}Co	10%	17,8%

Table 2. HPGe (G1) specifications

The output pulses are shaped and amplified by a Canberra 2020 amplifier (conventional Gauss shaping – time constant : 6 μs) then coded through an mutichannel buffer (EG&G ORTEC Model 919 Spectrum Master). Acquisitions are driven using Maestro® software that is also used for spectra display and processing. In general, the peak areas are computed according to Maestro® as the net area of a peak region whose background is linearly subtracted.

2.2. Point source calibration

The reference efficiency calibration was previously obtained using standard point sources at the reference position (10,35 cm between source and detector window). The nuclides used for the efficiency determination are : ^{54}Mn , ^{56}Co , ^{57}Co , ^{60}Co , ^{109}Cd , ^{133}Ba , ^{134}Cs , ^{152}Eu , ^{192}Ir and ^{241}Am . The activity (A) of each radionuclide was determined by direct methods such as coincidence method or liquid scintillation or by

a secondary method such as ionization chamber, that allows to get relative uncertainties of a few 10^{-3} .

For each peak, corresponding to energy E, with area N(E), the relevant full-energy peak efficiency, $\varepsilon(E)$ is :

$$\varepsilon(E) = \frac{N(E)}{A \cdot I(E) \cdot t} \cdot C \quad [1]$$

The gamma-ray emission intensities, I(E) are taken from the database NUCLEIDE [2] ; t is the acquisition time and C is the product of different corrective factors for effects such as coincidence summing, radioactive half-life, ...

Corrections for coincidence summing are determined using ETNA for multi-gamma emitting nuclides (^{56}Co , ^{60}Co , ^{133}Ba , ^{152}Eu , ^{134}Cs) ; the maximum correction factor is about 1,02.

The experimental efficiency values versus the energy are fitted using a log-log polynomial with the EFFIGIE software [2]. The resulting efficiency curve is displayed in figure 1 ; the associated relative uncertainties are around 0,5 %

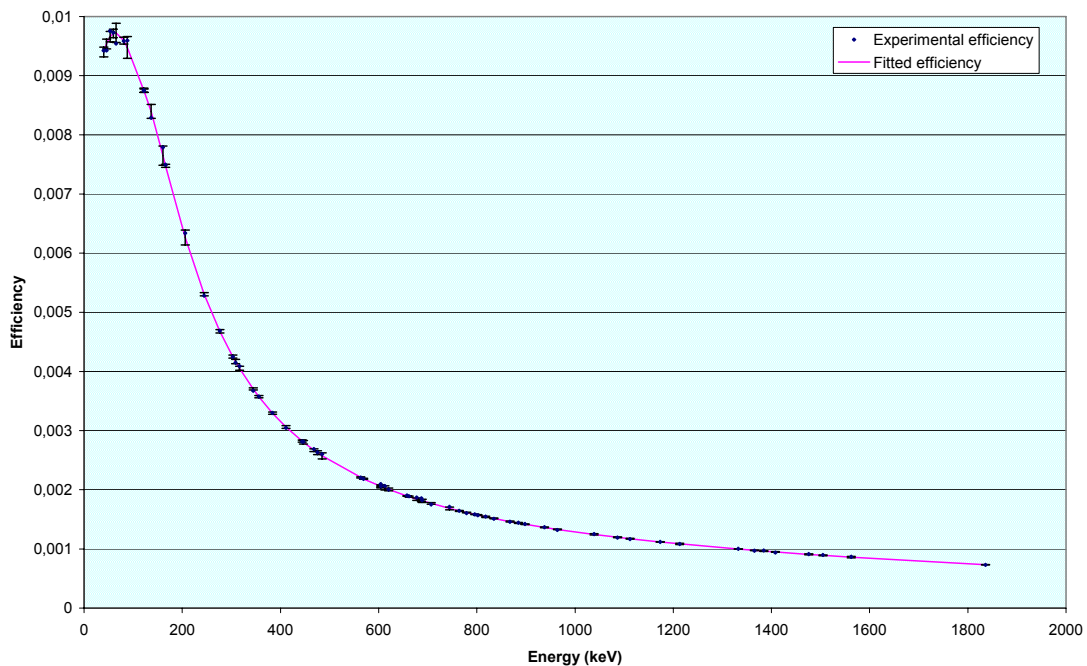


Figure 1 : Efficiency calibration for point source at 10,35 cm

2.3. Volume source calibration

Another efficiency curve was established using the so-called “SG50” volume geometry at 10,35 cm from the detector window. This 50 cm³ polyethylene [(CH₂-CH₂)_n – density = 0,92 (2)) container is filled with an hydrochloric solution (1mole.l⁻¹) including standard radionuclides. The container has the following characteristics : external diameter = 39 (1) mm, wall thickness = 1,2 (1) mm and bottom thickness = 1,4 (1) mm. The liquid filling height is deduced from the solution mass (48,202 g) and density (1,016 g.cm⁻³), that gives 4,51 cm.

The experimental efficiency values are obtained according to the same procedure as for the point sources, and also fitted using EFFIGIE. The resulting efficiency curve is displayed in figure 2 ; the associated relative uncertainties are about 2,0 %

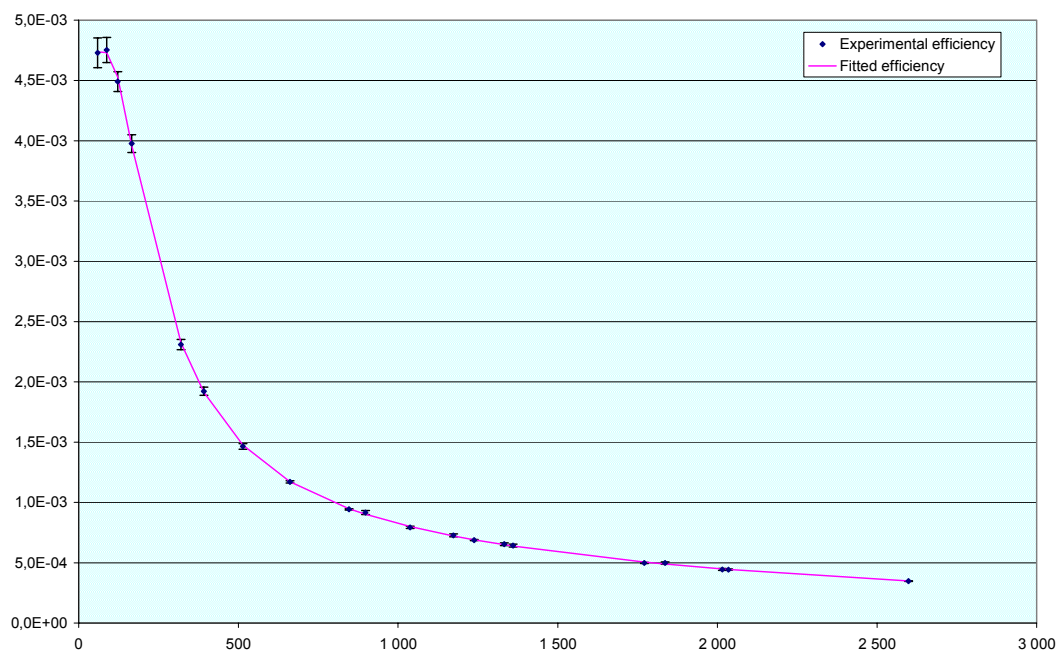


Figure 2 : Efficiency calibration for SG50 volume source at 10,35 cm

2.4 Efficiency transfer using ETNA software

The coefficients of efficiency transfer between both calibration geometries can be computed by ETNA, using the geometrical parameters according to the detector specifications. These computation results are compared with the experimental values in table 3.

Energy (keV)	Computed	Experimental	Ratio comp/exp
100	0,004639	0,004699	0,987
150	0,004189	0,004182	1,001
200	0,003478	0,003499	0,994
250	0,002863	0,002918	0,981
300	0,002414	0,002472	0,977
400	0,001824	0,001875	0,973
500	0,001470	0,001514	0,971
600	0,001242	0,001280	0,970
700	0,001088	0,001116	0,975
800	0,000971	0,000995	0,976
900	0,000880	0,000901	0,977
1000	0,000807	0,000825	0,978
1100	0,000744	0,000763	0,976
1200	0,000698	0,000709	0,984
1300	0,000651	0,000663	0,981
1400	0,000612	0,000623	0,983
1500	0,000577	0,000587	0,983
1700	0,000515	0,000525	0,982
2000	0,000439	0,000452	0,973

Table 3 : Computed and experimental values of the efficiency for SG50 geometry

3. Optimization of the geometrical parameters

The initial computation results show a deviation relative to experimental results. As this bias is nearly constant (about 3%) along the whole energy range, we suspect a wrong geometrical factor. Thus, to try to improve the geometric parameters

knowledge, it is necessary to perform complementary measurements and to compare them with the ETNA computation results using different parameters.

3.1 Displacement of a ^{137}Cs point source

We use a single ^{137}Cs point source that is positioned along the detector axis at different distances from the detector window, ranging from 2,35 cm to 18,35 cm. As there is only one gamma-ray emission (662 keV), there is no coincidence summing that would vary versus geometrical conditions : thus the ratio of the full-energy peak area for two distances directly corresponds to the ratio of relevant efficiencies. Second column of table 4 gives this ratio for the measurement distance relative to the reference distance (10,35 cm). Geometrical parameters of the detector (diameter, crystal-to-window distance) are slightly changed to try to fit the ETNA computed results with the experimental ones along the whole distance variation. The most coherent results of the efficiency ratios are given in third column of table 4. These are obtained with a crystal-to-window distance of 6 mm instead of the 3 mm given in the specifications.

Distance (cm)	Computed	Experimental	Ratio comp/exp
2,35	7,290	7,348	0,992
3,35	4,986	4,968	1,004
4,35	3,630	3,611	1,005
5,35	2,737	2,722	1,005
6,35	2,180	2,135	1,000
7,35	1,716	1,716	1,000
8,35	1,421	1,406	1,002
9,35	1,182	1,176	1,002
11,35	0,856	0,856	1,004
15,35	0,508	0,505	1,020
18,35	0,370	0,376	1,000

Table 4 : Comparison of computed and experimental efficiency ratios

3.2 Optimized transfer results using ETNA

With the optimized crystal-to-window distance, the ETNA computation transfer for the “SG50” geometry can be renewed. Table 5 presents the same results as table 3 with the optimized parameter.

Energy (keV)	Computed	Experimental	Ratio comp/exp
100	0,004647	0,004699	0,989
150	0,004225	0,004182	1,010
200	0,003508	0,003499	1,002
250	0,002897	0,002918	0,993
300	0,002439	0,002472	0,987
400	0,001839	0,001875	0,981
500	0,001484	0,001514	0,980
600	0,001254	0,001280	0,980
700	0,001098	0,001116	0,984
800	0,000980	0,000995	0,985
900	0,000888	0,000901	0,986
1000	0,000814	0,000825	0,986
1100	0,000751	0,000763	0,984
1200	0,000704	0,000709	0,992
1300	0,000656	0,000663	0,990
1400	0,000617	0,000623	0,991
1500	0,000582	0,000587	0,991
1700	0,000519	0,000525	0,990
2000	0,000443	0,000452	0,980

Table 5 : Computed and experimental efficiency ratios for SG50 geometry with optimized parameter

These computed results are consistent with the experimental uncertainties. However, the remaining bias indicate that 2 % uncertainty must be assigned to the efficiency transfer coefficient.

4. Determination of the attenuation coefficients

The attenuation coefficients have to be measured for ashes and clinkers. According to Beer-Lambert's law, a parallel photon beam with energy E and intensity $I_0(E)$, arriving upon normal incidence on a material of thickness x , is attenuated :

$$I(E) = I_0(E) \exp(-\mu(E)x) \quad [2]$$

$I(E)$ is the intensity of the transmitted beam, and $\mu(E)$ is the attenuation coefficient of material for energy E .

4.1 Experimental arrangement

The initial photon beam is provided by a ^{152}Eu point source emitting photons from 122 to 1408 keV thus allowing to cover the energy range of interest for the samples activity measurement. A first lead collimator (source collimator-thickness = 5 cm, hole diameter = 1 cm) is situated just under the source and another one (detector collimator) with the same characteristics is positioned above the detector window to get a nearly parallel beam. The ^{152}Eu spectra are recorded with the successive geometrical conditions :

- nothing between source and detector (Figure 3-a);
- empty container positioned above the detector collimator ;
- container filled with known sample mass for each matrix (Figure 3-b).



Figure 3 -a



Figure 3 -b

4.2 Results

From the first two series of spectra, it is possible to derive the attenuation coefficients of the container material. For each peak corresponding to energy E :

$$\mu(E) = \frac{1}{x} \cdot \ln\left(\frac{N_0}{N}\right) \quad [3]$$

where N_0 and N are the peak areas respectively without and with the container and x is the bottom thickness of the container.

Unfortunately, the results are not consistent and it is not possible to derive a coherent attenuation curve versus the energy. Due to limited schedule that did not allow to examine more carefully this problem, it was finally decided to use the tabulated data from XCOM [4].

The last two series of spectra are used to determine the attenuation coefficients of ash and clinker. The associated uncertainties are :

$$\left(\frac{u(\mu)}{\mu} \right)^2 = \left[\frac{u^2(x)}{x^2} + \left(\ln \frac{N_0}{N} \right)^{-2} \left(\frac{u^2(N)}{N^2} + \frac{u^2(N_0)}{N_0^2} \right) \right] \quad [4]$$

Except for 1112 keV and 1212 keV, the experimental values are obtained with 2 % mean relative uncertainties. These data are fitted using a quadratic log-log function and the resulting curves are drawn in Figures 4-a and 4-b. In a second step, these fitted values are input in ETNA to calculate the efficiency corresponding to each matrix in the volumic measurement conditions.

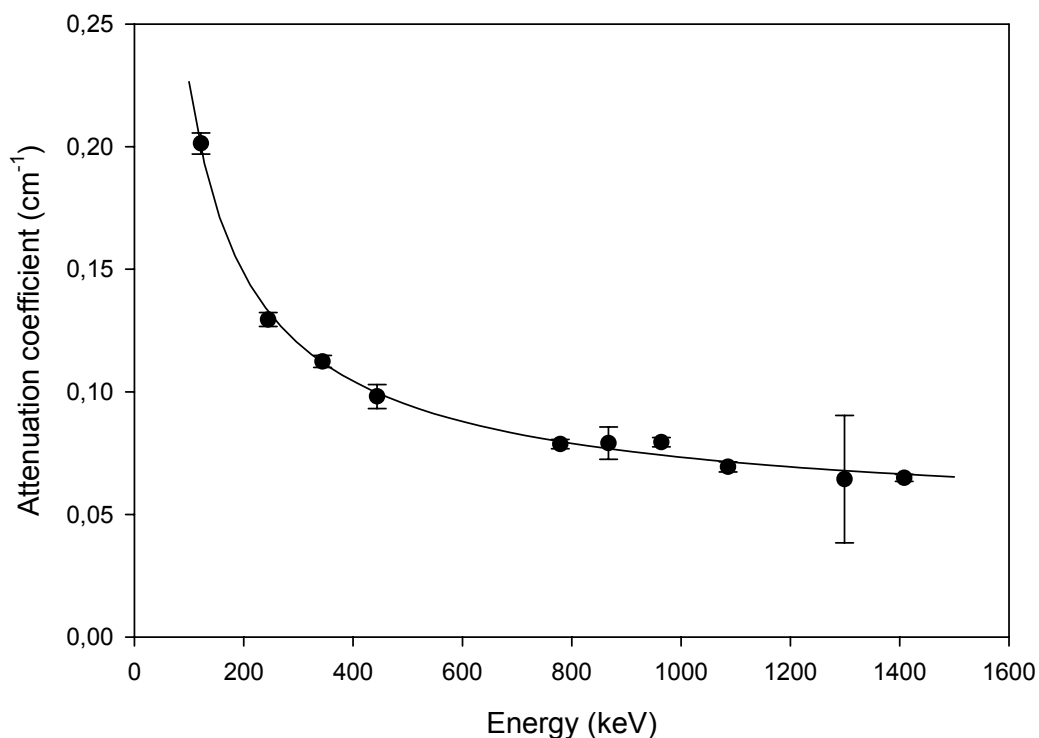


Figure 4-a : Attenuation coefficient of ash (experimental points with uncertainties and fitted log-log function)

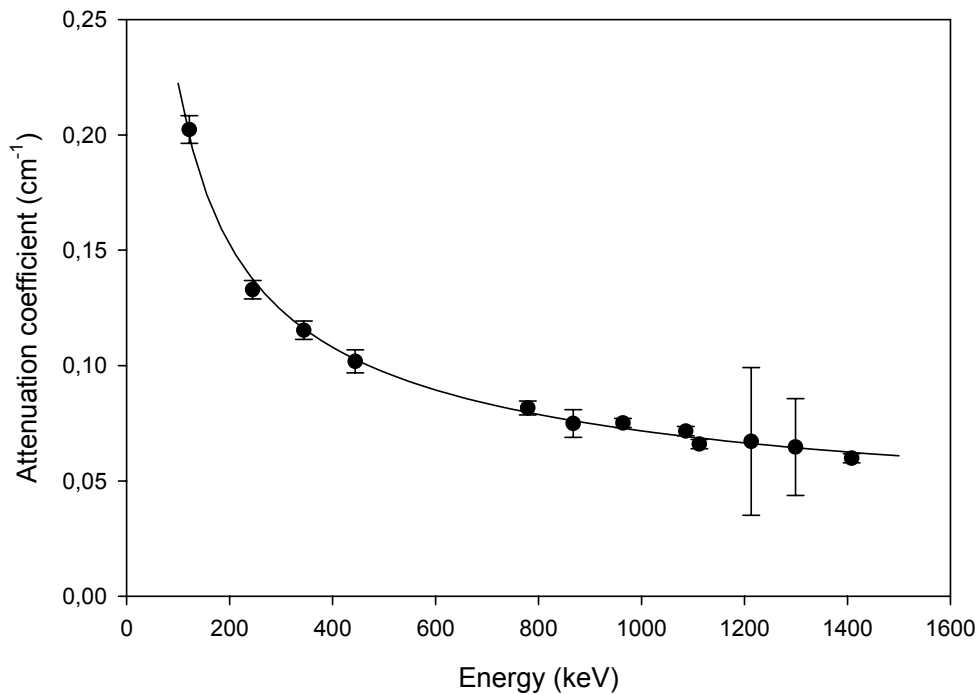


Figure 4-b : Attenuation coefficient of clinker (experimental points with uncertainties and fitted log-log function)

5. Determination of the activity of volume samples

5.1 Experimental conditions

To reduce efficiency transfer bias, samples of ash and clinker contained in the cylindrical geometry are measured at the 10,35 cm source-to-detector distance. Three measurements are processed for each sample, corresponding to 250 000 s acquisition time. With around 200 s⁻¹ counting rate, no pile-up correction is required. Figures 5 and 6 display the spectra of ash and clinker, respectively.

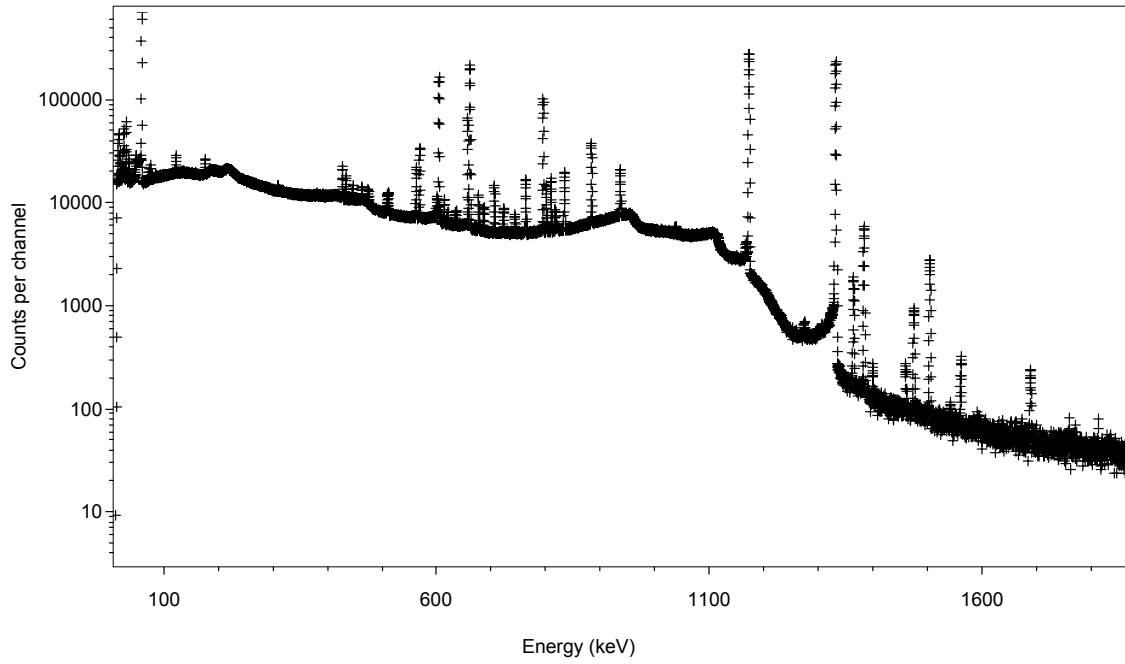


Figure 5 : spectrum of ash sample

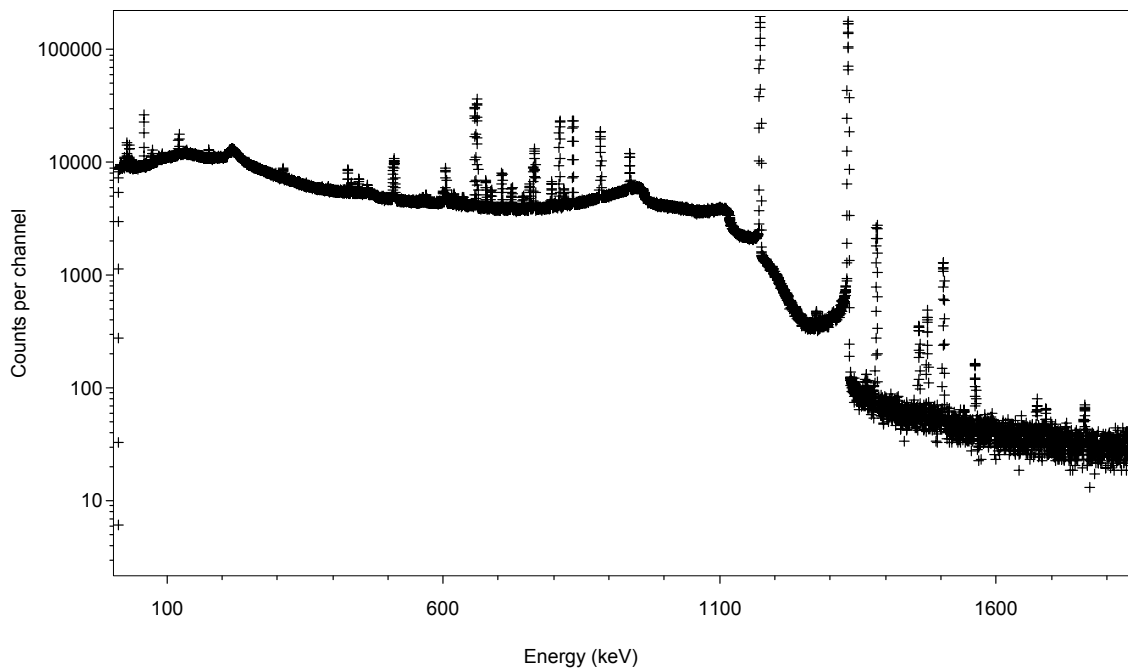


Figure 6 : spectrum of clinker sample

The peak areas are generally computed according to Maestro processing software. However, for regions with overlapping peaks, the COLEGRAM code [5] is used to

describe the individuals components and compute their respective areas. For this, a Gaussian function is fitted to the experimental data using a least-squares method. Figure 7 presents an example of deconvolution with COLEGRAM in the 760-770 keV energy region where peaks of ^{95}Zr (756,7 keV) and ^{95}Nb (765,03 keV) overlap.

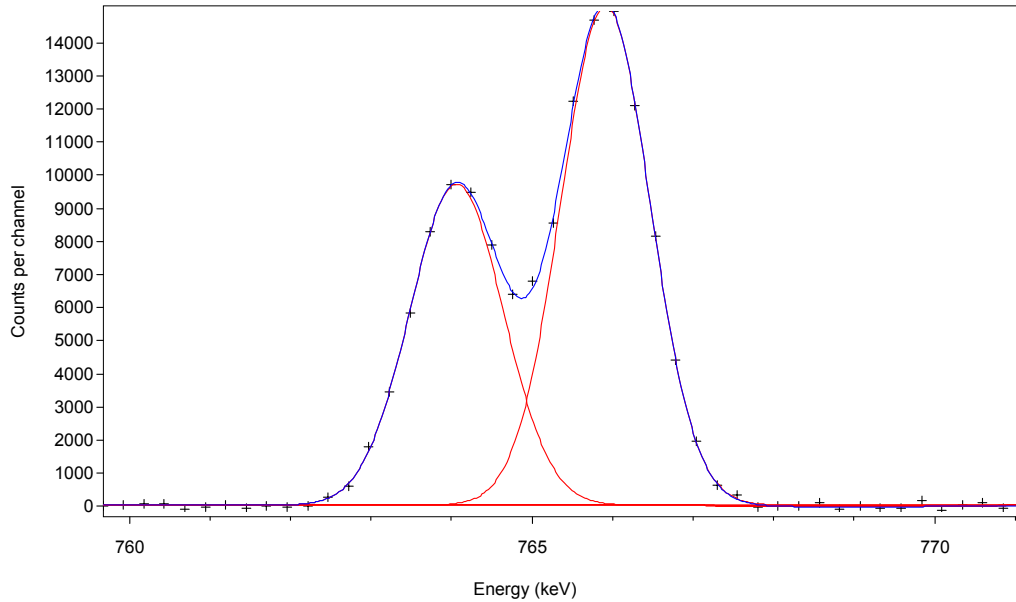


Figure 7 : Processing of the 760-770 keV energy region using COLEGRAM

5.2 Efficiency transfer

The efficiency transfer coefficients are computed by ETNA for both samples from the point source calibration efficiency, using the presently measured attenuation coefficients (see § 4.2). The resulting values are presented in table 6.

Energy (keV)	Ash efficiency	Clinker efficiency
100	0,005197	0,005223
120	0,005125	0,005123
150	0,004839	0,004822
200	0,004056	0,004037
250	0,003366	0,003345
300	0,002835	0,002817
400	0,002187	0,002176

500	0,001712	0,001707
600	0,001440	0,001437
700	0,001256	0,001255
800	0,001116	0,001116
900	0,001009	0,001010
1000	0,000920	0,000923
1100	0,000845	0,000848
1200	0,000791	0,000794
1300	0,000735	0,000739
1400	0,000689	0,000693
1500	0,000648	0,000652
1600	0,000611	0,000615
1691	0,000579	0,000583
1700	0,000577	0,000581
1800	0,000545	0,000549
1900	0,000516	0,000520
2000	0,000489	0,000493

Table 6 : ETNA computed efficiencies for the volume samples

5.3 Activity results

The activity of each radionuclide contained in the samples is determined according to equation [1] ; the results are given in table 7. The main corrective factors are due to coincidence summing, depending on the radionuclide decay scheme. These are calculated by ETNA software. For both matrixes, $^{108}\text{Ag}^m$, $^{110}\text{Ag}^m$, ^{134}Cs and ^{22}Na are most concerned, with about 1,5 % correction.

The reference dates are : 23/02/04 for ash and 26/02/04 for clinker.

Radionuclide	Ash		Clinker	
	Activity (Bq)	Combined standard uncertainty (%)	Activity (Bq)	Combined standard uncertainty (%)
⁵¹ Cr	80	21	Not detectable	
⁵⁴ Mn	1 030	3,6	765	3,7
⁵⁷ Co	80	8,4	30	7,1
⁵⁸ Co	780	3,7	675	3,7
⁶⁰ Co	32 000	3,6	13 300	3,6
⁹⁵ Zr	-	-	140	6
⁹⁵ Nb	-	-	490	3,8
¹⁰⁸ Ag ^m	230	4,3	22	11
¹¹⁰ Ag ^m	3 320	3	830	3,7
¹²⁴ Sb	85	4,7	5	7,5
¹²⁵ Sb	1 100	4,5	184	4,9
¹³⁴ Cs	7 680	3,6	115	3,7
¹³⁷ Cs	13 300	3,6	1 130	3,6
²⁴¹ Am	12 200	4,3	156	5,3

Table 7 : Activity contained in both samples

5.4 Uncertainties

The relative uncertainties quoted in table 7 correspond to the combined standard uncertainties ($k = 1$) [6]. Their computation takes into account :

- peak area (N) : statistical fluctuations ($N^{1/2}$) and fitting process;
- efficiency (according to the reference - point source - efficiency curve) ;
- efficiency transfer (2 % as defined above) ;
- coincidence summing (10 % of the corrective factor) ;
- gamma emission intensity (according to ref.[2]).

6. References

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