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Photon emission intensities in the decay of U-235

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Abstract

New measurements of photon emission intensities in the decay of U-235 were performed. Source was prepared by deposition of a U-235 solution on glass plate. Standardization was carried out by defined solid angle alpha counting, giving the reference activity with 0.7 % relative combined uncertainty. Gamma spectrometry was performed with accurately calibrated high-purity germanium detectors. Corrections for source geometry and coincidence summing effects were applied. The reference line (185.72 keV) intensity was obtained with 1.3 % relative standard uncertainty.

Highlights

New measurements of the photon emission intensities in the decay of U-235 were performed.

Standardization was carried out by defined solid angle alpha counting.

Gamma spectrometry allowed determining the 185.72-keV emission intensity with 1.3 % relative combined uncertainty.

Absolute and relative emission intensities of gamma- and X-rays in the decay of U-235 and Th-231 are derived.

Keywords: U-235; Th-231; photon emission intensities; NORM

1. Introduction

Uranium-235 is the parent of one of the natural radioactive decay series and appears in the background of any spectrometer and is also classed as a NORM (Naturally Occurring Radioactive Material). Uranium-235 disintegrates by alpha emission to excited levels of thorium-231, with a half-life of $7.04 (1) \times 10^8$ years, producing thorium X rays and gamma rays in the energy range below 450 keV. Most of the emission intensities of these lines are very weak, since only X rays and 6 gamma rays have emission intensities higher than 0.01. Probably due to the weak specific activity of U-235 and its presence in any background measurement, only few experiments have been conducted to measure the photon emission probabilities associated to U-235 decay. Recommended values are based on the intensity of the 185.72-keV gamma ray which is used to normalize relative measurements. It must be noted that this reference peak overlaps with the 186.21-keV peak from Ra-226 decay in the experimental gamma-ray spectra. In the evaluation performed in 2008 (Xialong and Baosong, 2009), only six independent measurements of the absolute emission probability of the 185.72-keV gamma ray were available, leading to the recommended value of 57.1 (3) %. The progeny

in the U-235 decay chain (Figure 1) includes Th-231 (half-life= 25.52 h) which is in equilibrium with U-235: it decays by beta minus disintegration to excited levels of protactinium-231, producing Pa X rays and gamma rays with 352-keV maximum energy.

2. Experimental procedure

The source was prepared at JEC-Geel by deposition of a U-235 solution on glass plate. The impurities were quantified by isotope dilution mass spectrometry (U-234: 5.3 %, U-236: 0.110 % and U-238: 0.00005 %) what required to apply a corrective factor, $F_c=0.9458(7)$, to derive the U-235 activity from the source counting result. The deposit radius is 7.8(20) mm and the homogeneity was checked by autoradiography.

2.1 Alpha spectrometry

Standardization was carried out by defined solid angle (DSA) alpha counting, using a partially depleted PIPS detector (Canberra), in a vacuum chamber. The detector is connected linked to an analogic electronic chain with a multichannel analyzer which provides the experimental spectrum. The geometrical factor, $G = \frac{\Omega}{4\pi}$ (Ω being the solid angle), is derived from the source-to-collimator distance (166.29 (21) mm), the collimator and the source radii (20.255(2) mm and 7.8(20) mm, respectively) and is defined with 0.26 % relative combined standard uncertainty. Two spectra with a total acquisition time of 10^6 seconds have been recorded. The source activity, $A = 48.41(34)$ Bq is obtained as:

$$A = \frac{N \cdot F_c}{t \cdot G} \quad (\text{Eq. 1})$$

N is the net counting recorded by the detector and t is the acquisition live time. The uncertainty budget is presented in Table 1.

2.3 Gamma-ray spectrometry

2.3.1 Efficiency calibration

Gamma spectrometry was performed using two N-type high-purity germanium (HPGe) detectors: one for the low-energy range, and one for the high one, at different source-to-detector distances. The main HPGe detector (G9) using a 100 cm^3 germanium crystal is equipped with a 500- μm thick beryllium entrance window, linked to analog electronic chain. This spectrometer was calibrated using LNHB standard point sources, at the reference source-to-detector distance of 10 cm. The full-energy peak (FEP) efficiency, was derived from the net counting of peaks obtained from GAMMAVISION[®] spectroscopy software (ORTEC, 2016), or using COLEGRAM software (Ruellan *et al.*, 1986) to process complex region of interest with overlapping peaks. Coincidence-summing corrections are taken into account using the ETNA software (Piton *et al.*, 2000). The raw data (energy, efficiency) were fitted using two log-log polynomial functions, one for the low-energy range (15 keV to 150 keV) and one for the high-energy range (120 keV to 2 MeV). Experimental values and efficiency calibration fitted curves are plotted in Figure 1, with detail on the low-energy range presented in Figure 2. In the 120 keV to 150 keV region where both curves overlap, the relative difference between theses is less than 0.5 %. Relative standard combined uncertainties are about 0.6 % in the 120 keV to 1500 keV energy range, 1.0 % above 1500 keV and 1.0 % to 2.0 % from 30 keV to 120 keV. The secondary HPGe detector (GENIX) has a planar geometry with a 2.6 cm^3 germanium crystal and a 127- μm thick

beryllium window, connected to an analog electronic chain. Due to the small crystal size, it has a better energy resolution than the one of the large detector (full width at half maximum= 500 eV at 120 keV, instead of 800 eV for detector G9). The FEP efficiency is obtained for the reference source-to-detector of 8 cm, using the same procedure as for detector G9.

2.3.2 Photon emission intensities determination

Several spectra were obtained using the two HPGe detectors. The reference values are derived from spectra acquired with detector G9 during $t = (2.8 \times 10^6)$ seconds live time, the U-235 source being installed at the reference position. For each peak with energy E_i , the photon emission intensity, I_i is derived from the net counting of the full-energy peak, N_i obtained using GAMMAVISION® for well isolated peaks or COLEGRAM for the complex regions.

$$I_i = \frac{N_i \cdot C_{Gi} \cdot C_{Ci}}{A \cdot \varepsilon_i \cdot t} \quad (\text{Eq. 2})$$

A is the source activity (Bq) and t is the live time. The FEP efficiency, ε_i , is obtained from the calibration procedure. Corrections for the radionuclide decay are negligible. Corrections for source geometry, C_G , and coincidence summing effects, C_C , are computed using ETNA. The geometry effect due to the source area induces a corrective factor between 1.004 and 1.006, for the high and low energies, respectively. The coincidence summing corrective factor takes into account true coincidences between gamma rays and K and L X rays, and range from 1.000 to 1.021. The associated relative combined standard uncertainties are computed as:

$$\frac{u(I_i)}{I_i} = \left[\frac{u^2(N_i)}{N_i^2} + \frac{u^2(\varepsilon_i)}{\varepsilon_i^2} + \frac{u^2(A)}{A^2} + \frac{u^2(C_{Gi})}{C_{Gi}^2} + \frac{u^2(C_{Ci})}{C_{Ci}^2} + 0.2^2 + 0.28^2 \right]^{1/2} \quad (\text{Eq. 3})$$

The uncertainty associated to the count rate, $u(N_i)$, includes statistics and background subtraction, and the last term is added to take into account the peak area determination and the reproducibility. The uncertainty budget for the reference peak (185.72 keV) is presented in Table 2. The relative combined standard uncertainties associated to the six main absolute intensity values are between 1.3 % and 2.0 %.

Complementary measurement was performed with the source at 2 cm from the detector window, to derive the intensities of weaker peaks. Corrective factors for efficiency transfer were necessary and induced larger uncertainties for these peaks. In addition, the detector GENIX, with the source at 3 cm from the beryllium window, was used to record spectra for a detailed processing of the low-energy region. During the peak area determination, some energy regions required special attention, as detailed hereafter:

- Doublet 182.62 keV and 185.72 keV

The reference peak (185.72 keV) was analyzed together with the 182.62-keV peak, using COLEGRAM, the area of last one representing 0.7 % of the region of interest. The contribution of the background was subtracted as 4.6 % of the net peak area. The coincidence summing corrective factor applied was 1.0008. The resulting photon emission intensity is 0.576 (7). This result is compared with previous experimental data in Table 3.

- 70 keV to 100 keV (K X-ray lines)

There is a large number of peaks in the K-X ray region, due to U-235 decay (Th K X rays and several gamma-ray lines) and to other daughters in equilibrium (Ac and Pa K X rays). Moreover, background

peaks (Pb and Bi K X rays and U self-fluorescence K X rays are also present in the energy region ranging from 70 keV to 100 keV. This region was processed using COLEGRAM which allows fitting of experimental data with Gaussian or Voigt functions, according to the origin of the photons. Gamma-ray peaks are fitted using Gaussian shapes, whose individual widths follow a linear function versus the root square of the energy. X-ray peaks are fitted with Voigt functions, the Gaussian width evolving as the gamma-ray ones while the Lorentzian width are fixed values, derived from tabulated data: the intrinsic linewidth of the emitted photon is calculated as the sum of the width of the corresponding atomic levels (Campbell and Papp, 2001). An example of the peak fitting results is presented in Figure 3.

- 10 keV to 20 keV (L X-ray lines)

The case of the low-energy region is similar to the case of the K X-ray energy region since it includes L X rays of different elements. Here, these are processed using simple Gaussians, since the natural line width (Lorentzian) of L lines is much lower than the experimental Gaussian width and can be neglected in first approximation.

3. Results and discussion

Absolute and relative photon emission intensities in the decay of U-235 are presented in Table 4. The relative photon emission intensities, I_{iR} , are computed using the 185.72-keV peak (M) intensity as normalization factor:

$$I_{iR} = \frac{\frac{n_i C_{Gi} C_{Ci}}{\varepsilon_i A}}{\frac{n_M C_{GM} C_{CM}}{\varepsilon_M A}} \cdot 100 = \frac{n_i C_{Gi} C_{Ci}}{n_M C_{GM} C_{CM}} \cdot \frac{\varepsilon_M}{\varepsilon_i} \cdot 100 \quad (\text{Eq. 4})$$

In this case, the associated relative combined standard uncertainties are computed according to:

$$\frac{u(I_{iR})}{I_{iR}} = \left[\frac{u^2(N_i)}{N_i^2} + \frac{u^2(N_M)}{N_M^2} + \frac{u^2(\varepsilon_i)}{\varepsilon_i^2} + \frac{u^2(\varepsilon_M)}{\varepsilon_M^2} + \sum_j \frac{u^2(C_{ij})}{C_{ij}^2} + \sum_k \frac{u^2(C_{Mk})}{C_{Mk}^2} + u(\varepsilon_i, \varepsilon_M) \right]^{1/2} \quad (\text{Eq. 5}),$$

where $u(\varepsilon_i, \varepsilon_M)$ is the covariance taking into account the correlation between the efficiency values. In addition, Table 5 presents photons emission intensities in the decay of Th-232, which were determined in the same way. Detailed values for the L X-rays groups are given for thorium and for protactinium in the U-235 decay chain. In both tables, the absolute photons emission intensities are compared with the evaluated data (Nucleide, 2016). Most of the present results are in agreement with the tabulated data. However, some discrepancies are noticed in the 70-keV and 90-keV energy regions: the complex spectrum structure with gamma- and K X-rays from different nuclides, together with the strong influence of the background and uranium self-fluorescence make it difficult to unambiguously attribute the counting to individual lines. Nevertheless it is expected that these new experimental values will provide helpful information for further evaluation of the decay schemes of U-235 and Th-231.

Acknowledgement

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Tables captions

Table 1: Uncertainty budget for the activity measurement using DSA

Table 2: Uncertainty budget for the absolute photon emission intensity of the reference peak (185.72 keV)

Table 3: 185.72-keV absolute photon emission intensity: comparison with previous results (*Italics: doubtful results*)

Table 4: Relative and absolute photon emission intensities in the decay of U-235, and comparison with Nucleide database

Table 5: Relative and absolute photon emission intensities in the decay of Th-231, and comparison with Nucleide database (Normalization is calculated using the absolute intensity of the U-235 185.72-keV peak)

Figures captions

Figure 1: U-235 decay chain

Figure 2: Full-energy peak efficiency calibration versus the energy (lower panel: detail in the low-energy range)

Figure 3: Processing of the K X-ray region using COLEGRAM software

Contribution	Relative standard uncertainty
Statistics	0.41 %
Extrapolation	0.30 %
Geometrical factor	0.26 %
Backscattering (chamber)	0.10 %
Backscattering (window)	0.001 %
Counting time	0.01 %
Temperature	0.006 %
Impurities	0.074 %
Reproducibility	0.35 %
Total	0.7 %

Table 1: Uncertainty budget for the activity measurement using DSA

Contribution	Relative standard uncertainty
Source activity	0.7 %
Statistics	0.13 %
Peak area determination	0.2 %
Efficiency	1.0 %
Counting time and radionuclide decay	negligible
Geometry	0.1 %
Coincidence summing	0.01 %
Reproducibility	0.28 %
Total	1.3 %

Table 2: Uncertainty budget for the absolute photon emission intensity of the reference peak (185.72 keV)

Reference	Experimental value	Comment
Vaninbroukx and Deneke (1982)	0.575 (9)	Enriched material - Activity : mass spectrometry isotope dilution and alpha counting
Olson (1983)	0.561 (8)	Enriched material (97.66%) – NBS standard
Helmer and Reich (1984)	0.572 (5)	NBS standard
Lin and Harbottle (1991)	0.568 (13)	Enriched material (92.8%)
<i>Chatani (1999)</i>	<i>0.58 (2)</i>	<i>Indirect measurement - U-Al alloy and purified ²³¹Th source</i>
<i>Al-Saleh et al. (2006)</i>	<i>0.8019 (4)</i>	<i>Uranyle nitrate hexahydrate – powder sample</i>
Present result	0.576 (7)	Enriched material (94.58%)

Table 3: 185.72-keV absolute photon emission intensity: comparison with previous results
(Italics: doubtful results)

Energy (keV)	Relative intensity	Absolute intensity	
	This study	This study	Nucleide
Th L α X-rays	1.036 (26)	0.599 (15)	
Th L α X-rays	35.1 (8)	20.26 (44)	
Th L β X-rays	30.1 (6)	17.42 (38)	
Th L γ X-rays	4.20 (9)	2.43 (5)	
Th total L X-rays	70.4 (17)	40.7 (10)	40(22)
72.7	0.617 (12)	0.341 (18)	0.116 (20)
74.94	0.232 (5)	0.128 (7)	0.051 (6)
Th K α 2 X-ray	6.77 (10)	3.73 (19)	3.56 (7)
Th K α 1 X-ray	8.40 (13)	4.62 (24)	5.75 (11)
96.09	0.232 (5)	0.128 (7)	0.091 (11)
Th K β '1 X-rays	2.65 (4)	1.46 (8)	2.05 (5)
Th K β '2 X-rays	0.782 (12)	0.431 (22)	0.690 (19)
109.19	2.86 (4)	1.58 (8)	1.66 (13)
115.45	0.0441 (5)	0.0255 (3)	0.03 (1)
136.55	0.0468 (15)	0.0258 (15)	0.012
140.76	0.287 (22)	0.165 (13)	0.20 (1)
143.767	18.79 (24)	10.83 (14)	10.94 (6)
150.936	0.084 (23)	0.048 (13)	0.09 (3)
163.356	8.57 (12)	4.94 (7)	5.080 (3)
182.62	0.750 (25)	0.432 (14)	0.39 (5)
185.72	100.0 (12)	57.6 (7)	57.1 (3)
194.94	1.18 (3)	0.678 (18)	0.63 (1)
198.894	0.286 (25)	0.165 (14)	0.036 (2)
202.12	2.06 (4)	1.187 (24)	1.08 (2)
205.316	8.88 (12)	5.11 (7)	5.02 (3)
221.386	0.21 (3)	0.119 (20)	0.118 (5)
240.88	0.175 (4)	0.101 (5)	0.074 (4)
275.35	2.69 (4)	0.094 (5)	0.051(6)
275.49			0.032
291.65	0.074 (2)	0.0422 (25)	0.040 (6)
343.54	0.0126 (9)	0.0072 (6)	0.0032
345.92	0.0435 (23)	0.0248 (18)	0.040 (6)
387.84	0.049 (4)	0.0281 (26)	0.040 (6)

Table 4: Relative and absolute photon emission intensities in the decay of U-235, and comparison with Nucleide database

Energy (keV)	Relative intensity	Absolute intensity	
	This study	This study	Nucleide
Pa L α X-rays	2.88 (7)	1.66 (4)	
Pa L α X-rays	50.9 (11)	29.4 (6)	
Pa L β X-rays	49.9 (11)	28.8 (6)	
Pa L γ X-rays	8.53 (18)	4.93 (11)	
Pa total L X-rays	112.2 (24)	64.8 (14)	65 (3)
25.64	24.14 (31)	13.95 (18)	13.9 (7)
58.57	0.820 (18)	0.453 (25)	0.480 (16)
81.228	1.541 (30)	0.85 (5)	0.905 (23)
82.087	0.642 (16)	0.353 (20)	0.418 (13)
84.214	12.67 (19)	6.98 (36)	6.70 (7)
Pa K α 2 X-ray	0.420 (7)	0.231 (12)	0.37 (4)
99.278	0.249 (5)	0.137 (7)	0.137 (6)
102.27	0.585 (10)	0.322 (17)	0.441 (11)
Pa K β '1 X-rays	0.211 (4)	0.116 (6)	0.21 (2)
163.101	0.821 (14)	0.452 (24)	0.156 (5)

Table 5: Relative and absolute photon emission intensities in the decay of Th-231, and comparison with Nucleide database (Normalization is calculated using the absolute intensity of the U-235 185.72-keV peak)

Figure 1: ^{235}U decay chain

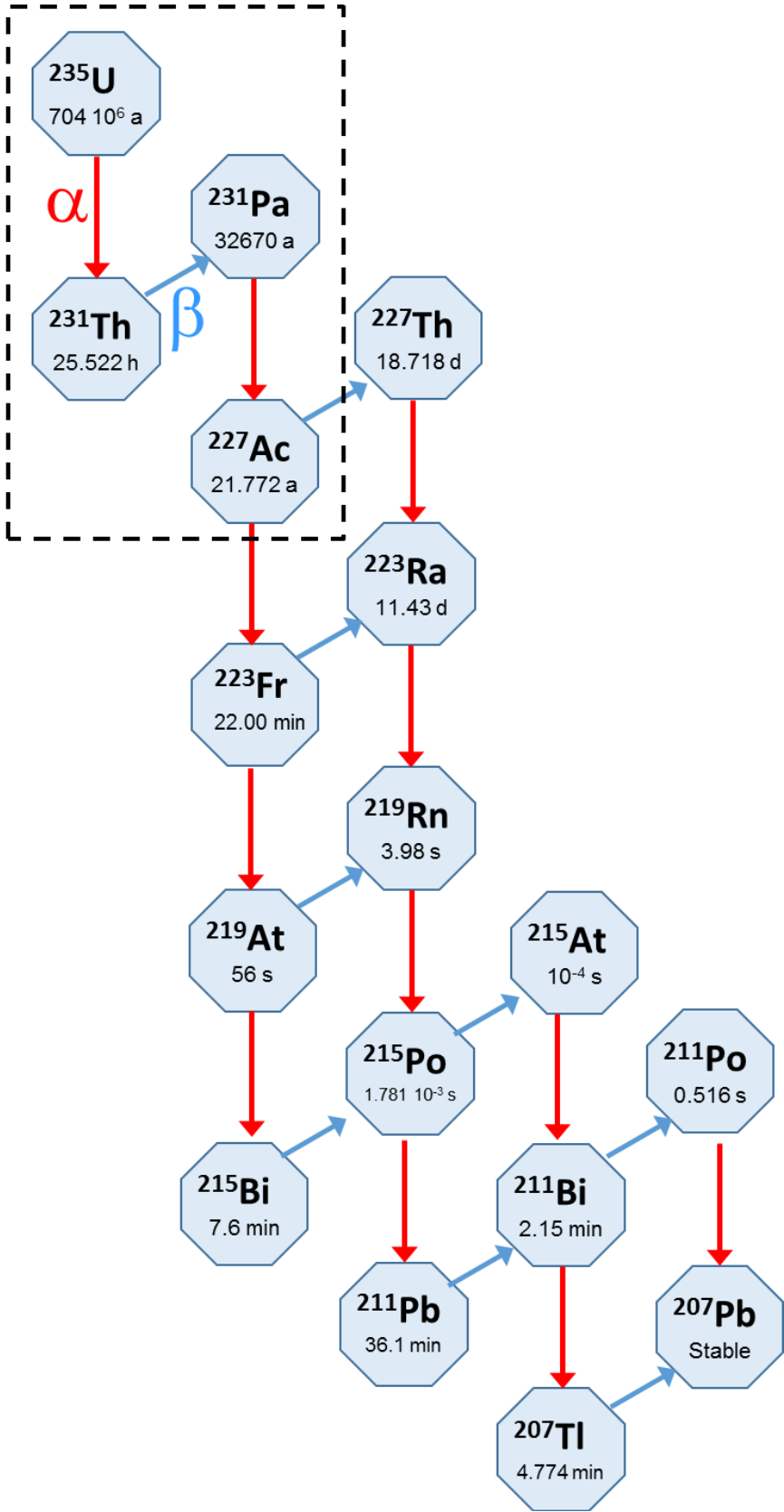


Figure2: Full-energy peak efficiency calibration versus the energy (lower panel: detail in the low-energy range)

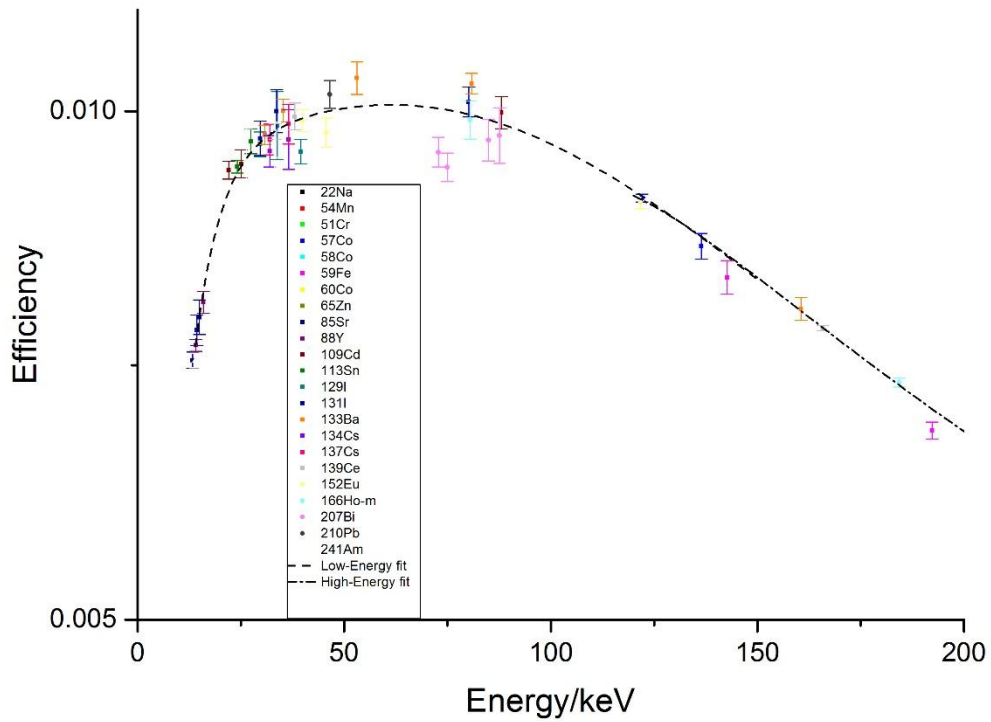
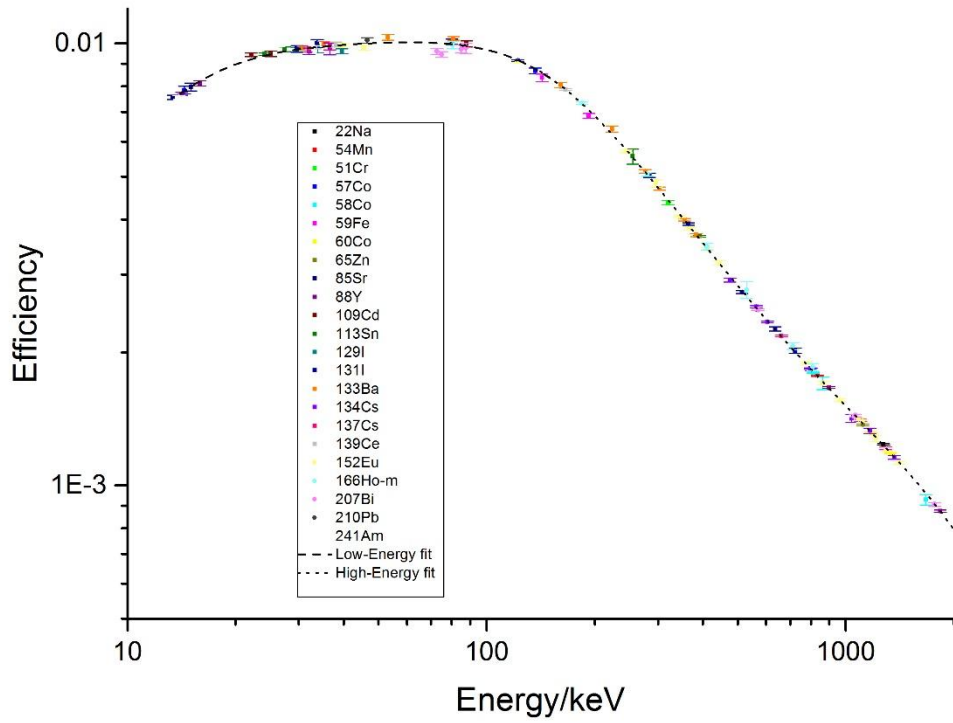


Figure 3: Processing of the K X-ray region using COLEGRAM software

