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STUDIES AND ASSESSMENTS ON THE RESPONSE OF A HIGH-PERFORMANCE SPECTROMETER TO ALPHA RADIATION

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Abstract. This paper describes studies and assessments of the main parameters of a high resolution multi-chamber alpha spectrometer that will be used in SALMROM, accredited laboratory in the Department of Life and Environmental Sciences of the IFIN-HH, for measurements of alpha radioactive concentrations in environmental samples. For the measurements there were used standard sources of ^{241}Am, ^{233}U, ^{244}Cm and of a mixture of radionuclides (^{239}Pu + ^{241}Am + ^{244}Cm). Topics covered include instrumentation, parameters, source characterization, measurements, analysis and discutions. Performances of the spectrometer based on eight ORTEC detectors were determined for all eight chambers and for all possible distances source-detector considering, also, the detector resolution and efficiencies repeatability for the energy lines of the alpha sources and the Minimum Detectable Activities.

Key words: energy calibration, efficiency calibration, repetability, alpha-ray spectrometry, octête Plus, alpha-ray measurement.

1. INTRODUCTION

Alpha-particle spectrometry is a measurement technique that has found many practical applications in various fields as nuclear decay data measurements, geological studies, or the measurement of low levels of activity in the environment. It represents a very useful and sensitive method for analyzing alpha-emitting nuclides, mainly because of the detector's very low background with a detection limit often 100–1000 times lower than γ-spectrometry [1, 2].

The Laboratory *SALMROM* for alpha, beta, gamma Spectrometry and Radon Measurements from the *Department of Life and Environmental Sciences* of the *Horia Hulubei National Institute for Physics and Nuclear Engineering*, Bucharest (IFIN-HH), was equipped with a state-of-the-art multi-chamber alpha spectrometer, type Octête Plus from ORTEC company to be used for activity measurements of alpha emitting radionuclides in environmental samples, in compliance with the laboratory's Quality Assurance documentation [3, 4].

The purpose of this paper is to present the main parameters of the alpha spectrometer, including the energy and efficiency calibration of the system using standard radiation sources, the Minimum Detectable Activities, the detector's resolution and efficiencies repeatability at the energy lines of the alpha standard sources.

2. METHOD AND MATERIALS

The experimental equipment is an integrated laboratory multi-detector/multi-channel analyzer alpha spectrometer based on eight chambers, type Octête, ORTEC, used for activity measurements of alpha-ray emitting radionuclides from environmental samples and samples of radioactive materials [4].

Silicon semiconductor alpha detectors operate at a low reverse bias voltage condition, normally between 50-100 volts DC. The voltage bias supply should be highly regulated to prevent noise and loss of resolution. The polarity of the bias depends on the type of detector, *e.g.*, surface, barrier, etc.

Each incorporated chamber is an independent alpha spectrometry system with a complex structure including: a high-performance ion-implanted-silicon charged-particle detector located in its own measuring chamber, operated in the energy range 0 ÷ 10 MeV, type ULTRA-AS, model BU-019-300-AS, of 300 mm^2 active area, its associated electronics namely, a variable detector bias supply, a preamplifier, a shaping amplifier with adjustable gain, a pulse stretcher and bias amplifier, a test pulser generator adjustable over the range 4 ÷ 10 MeV for noise measuring and chamber calibration, a monitor of the detector leakage current and a vacuum gauge. Also, each chamber is individually controlled by specialized ORTEC software as MAESTRO-32 or Alpha Vision-32, that make possible that the detector's bias voltage and leakage current to be read out on a personal computer. The level of the pressure inside the a measuring chamber could be read out and controlled in real time in the range 0÷30 Torr and when it has a certain internally preset value the protection vacuum interlock allows the automatic bias application [2, 4, 5, 6]. The alpha spectrometer is equipped with a Multi-Channel Analyser including 16-input multiplexer with individual controls for each channel and a successive approximation Analog Digital Converter with a conversion gain range from 64 to 4096 channels that can be selected through the PC software platform of MAESTRO-32 or Alpha Vision-32 programs [2].

To reduce the risk of vacuum leakage, all the eight chambers are connected to an integral vacuum manifold with a single connector. From the front panel of each channel, a valve VENT/HOLD/PUMP, interlocked to the detector bias, could be operated manually in order remove or apply the detector bias [4].

Inside each chamber there is a Sample Holder that allows the measurements of samples of 1÷5 mm diameter at different sample-detector distances between 4÷40 mm in 4 mm increments decided by the chamber wall slots. The reference environmental conditions for the measurements were as follows: temperature:

$(20 \pm 2)^0$ C; pressure: 1013.25 ± 0.2 hPa and humidity: 65% [7, 8]. The bias voltage applied to each detector was 50V that is a preset value. The measurements for the spectrometer's characterization were performed using alpha standard electro-deposited disk sources of radionuclides from the actinide series, described in Table 1. The sources were placed on each level of the chambers for the determination of detector's efficiencies and resolution at the energy lines of the certified standard disk sources in order to be performed the energy and efficiency calibrations of each channel of the alpha spectrometer.

Table 1

Alpha standard electro-deposited disk sources used in measurements

Standard radioactive alpha sources	**Active diameter/source diameter [mm]**	**Emission [$s^{-1}/2\pi r$]; Activity [Bq]**	**Manufacturer/ date**	**$T_{1/2}$ [year]**	**Obs.**
^{241}Am	5/22	$(7.5 \pm 0.15)\times10^3$/ 15000 ± 300	LMR IFIN HH/ 2011	432.6±0.6	disk source
^{233}U	5/25	145.2 ± 4.4/ 290.4 ± 8.7	CEA BNM LMRI/ 2010	$(159.2\pm0.020)\times10^3$	disk source
^{244}Cm	3/25	775 ± 23/ 1550 ± 46	CEA BNM LMRI/ 2010	18.11±0.03	disk source
^{239}Pu+ ^{241}Am+ ^{244}Cm (mixture of radionuclides)	5/25	(2001±60)/(2014±60) / (366±11) Total activity: 4381±122	AMERSHAM PLC	$(24.1\pm0.11)\times10^3$ 432.6±0.6 18.11±0.03	disk source

The levels with the best resolutions were considered as the optimum levels, and there were carried out repeatability measurements to study the concordance level between successive measurements. Considering the geometrical characteristics of the sources and of the detector and the distances source-detector in the range 4÷40 mm, there were determined the solid angles for all measurement geometries.

3. RESULTS AND DISCUSSION

3.1. DETECTOR EFFICIENCY

The ORTEC detector ULTRA-AS, model BU-019-300-AS, of 300 mm^2 active area, has an entrance window of 50 nm thickness. The ion-implanted-silicon charged-particle detectors are based on p-n junctions realized by the implantation of boron into a high purity silicon wafer, being obtained, after etching, the front contact. This represents the entrance window whose thickness controls the detector's resolution and the energy straggling of the alpha particles. The Figure 2 shows the average values of the efficiencies [4], for each alpha chamber level, considering all the measurements performed with standard alpha disk sources of ^{241}Am, ^{233}U and ^{244}Cm, for all eight alpha spectrometers and on all ten measurement levels of each one.

For all efficiency calculations it was considered the region of interest between the cannel 600 to 1935 of the pulse height spectra in order to be evaluated the background and integral area for a measuring live time of 1800 s.

Figure 1 shows that the efficiencies values of the alpha sources of ^{241}Am, ^{233}U and ^{244}Cm, considered as average values, are very similar for measurement levels higher than 1.5-2 × (detector diameter), when the straggling effect in the entrance window became insignificant. The efficiency of an alpha detector is the same for each alpha particle, being independent of their energy, but dependent on the measurement geometry.

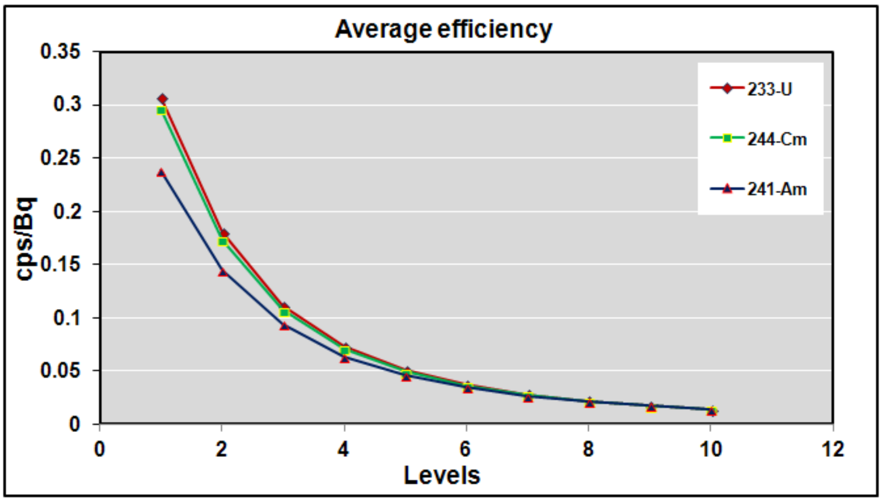

Fig. 1 – The average efficiency for ^{241}Am, ^{233}U and ^{244}Cm sources.

The maximum efficiency of the integrated alpha spectrometer with ULTRA-AS, model BU-019-300-AS detectors was 0.242 (cps/Bq) obtained for the chamber number 5 with the disk source of ^{241}Am. The efficiency of the alpha spectrometer has a rapid decreasing with the change of the measurement geometry, towards higher values of the source-detector distance. That is why, for high measurement efficiency, it is necessary to place the source as close as possible to the detector.

3.2. ENERGY CALIBRATION AND RESOLUTION

The full-width-half-maximum (FWHM) resolution of an alpha spectrometry system using commercially available detectors depends on several parameters that include: inherent energy resolution of the detector, charge carrier statistics, incomplete charge collection and variations in the energy loss in the dead layer, i.e., entry window thickness. The noise contributions of the non-detector system components to the energy spectrum are minimal for most alpha spectrometry systems [4]. The ORTEC detectors ULTRA-AS from each chamber were characterized by evaluating their response to alpha disk sources of ^{233}U, ^{239}Pu, ^{241}Am and ^{244}Cm. From the differential pulse height distributions representing the response of the alpha ULTRA-AS detectors to the energy lines of the sources, the energy calibration of the alpha spectrometer was performed and the energy resolutions of the detectors were determined, by calculating the full width at half maximum (FWHM) [7, 8]. Table 2 presents the peak positions corresponding to the energy lines of the Amersham plc source. Taking into account these results, it was obtained the energy calibration curve described by the following linear function:

$$\text{Energy [keV]} = a_0 + a_1 \times \text{Channel},$$

where: $a_0 = -12.46117$ and $a_1 = 4.98354$ are the coefficients of the fitting curve. The Residual Sum of Squares was determined. Its value of 10.20 reveals the goodness of the data fit curve.

Table 2

Energy calibration data

Radionuclides	**Energy [keV]**	**Channel**
^{239}Pu	5105.81	1027
^{239}Pu	5156.59	1037
^{241}Am	5388.26	1084
^{241}Am	5442.86	1095
^{241}Am	5485.56	1103
^{244}Cm	5762.65	1159
^{244}Cm	5804.77	1167

The values of the FWHM at the main energy lines of the ^{241}Am, ^{233}U and ^{244}Cm sources, are represented, in Figs. 2, 3 and 4 [4], for seven chambers, as a function of the distance of the source-detector, well defined by each level of the alpha chambers It can be seen, for all three alpha sources, that the variation of the resolution with the source-detector distance has 2 distinct parts: a fast decreasing of the FWHM for distances corresponding to the first 5 levels and stabilized values of the FWHM for the source-detector distances of the other 5 levels, in the range 24–40 mm [4].

In Table 3 the best values of the energy resolution of the detectors at the ^{241}Am line of 5.4856 MeV together with the corresponding efficiencies, in the same set-up conditions described before, are given for each chamber. For the eighth chamber (chamber 7) the data are not presented in Figs. 2–4 and Table 3 due to the presence of a high leakage current for this cell that alters the results.

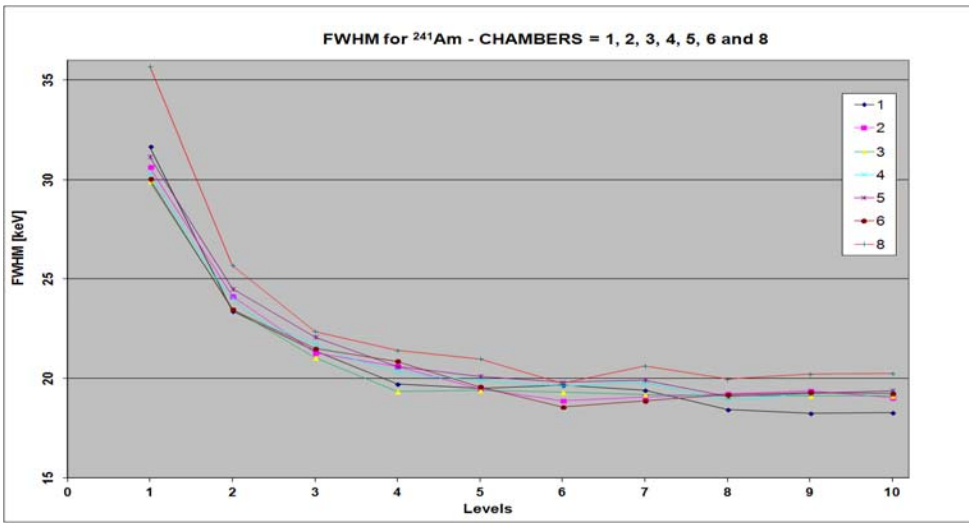

Fig. 2 – Detector FWHM at the ^{241}Am energy line of 5.4856 MeV.

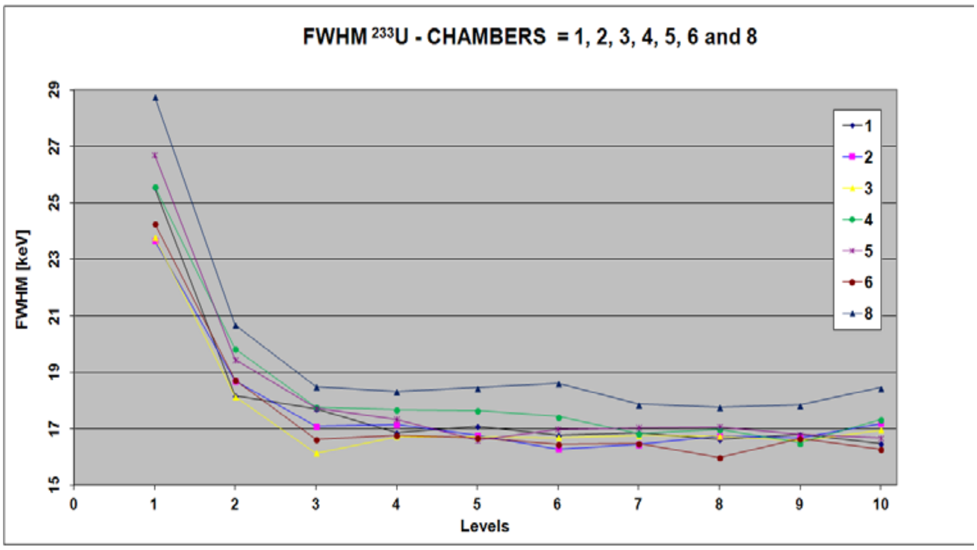

Fig. 3 – Detector FWHM at the ^{233}U energy line of 4.824 MeV.

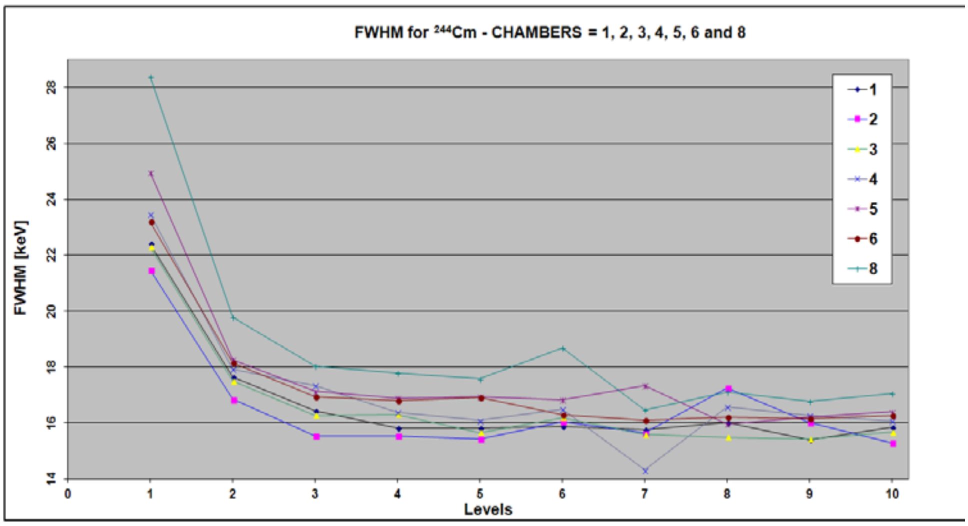

Fig. 4 – Detector FWHM at the ^{244}Cm energy line of 5.805 MeV.

Table 3

Optimum resolution for each chamber

Chamber/Level	Calculated Resolution [keV]
1/8	18.44
2/6	18.88
3/8	19.14
4/8	18.95
5/7	19.09
6/6	18.55
8/6	19.74

Figure 5 shows one spectrum of the Amersham disk source that contains a mixture of ^{239}Pu, ^{241}Am and ^{244}Cm radionuclides. It was placed on the level 7 of Chamber 5. It can be easily seen that the alpha multi–chamber spectrometer has a very good resolution to separate close peaks and multiple radionuclides can be well resolved.

3.3. REPEATABILITY MEASUREMENTS

Considering as optimum level of each chamber, that one corresponding to the optimum resolution, after one series of measurements, it was important to study the efficiency and the energy resolution repeatability, being performed ten measurements of 1800 s, with the ^{241}Am source.

The average values of the efficiencies and of the resolutions together with the relative standard experimental error values as a measure of their dispersion *S*(*n*–1) [%] and the relative standard experimental error of the mean values *S*(aver) [%] are presented in Table 4.

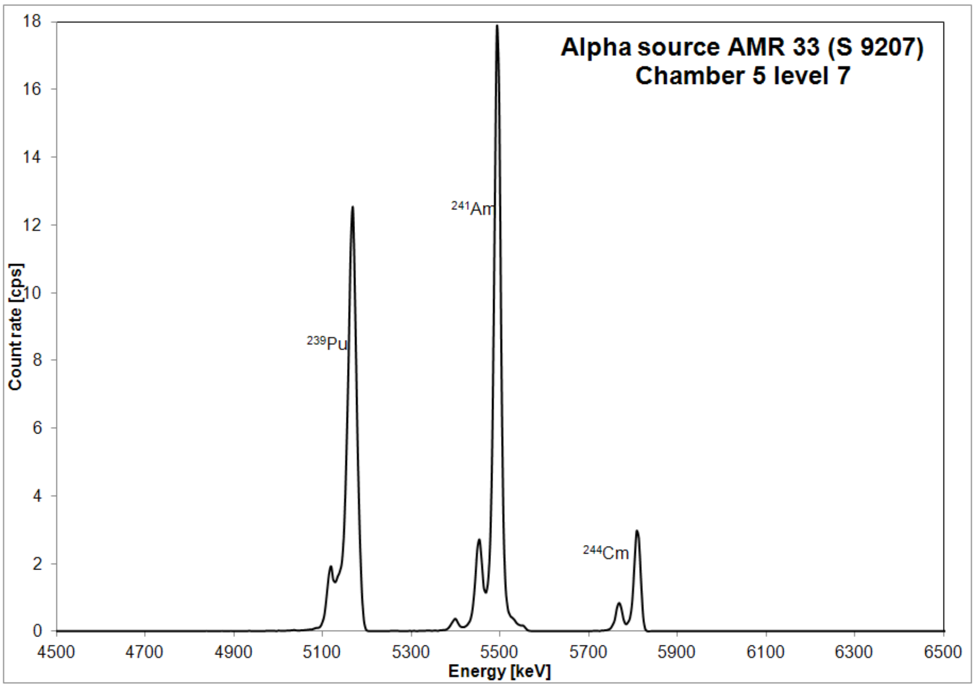

Fig. 5 – The spectrum for alpha source AMR 33, chamber 5, level 7.

Table 4

The efficiency and FWHM repeatability for ^{241}Am

Chamber	**^{241}Am**					
	Efficiency [cps/Bq]			**FWHM [keV]**		
	Efficiency average	***S*(*n*-1) [%]**	***S*(aver) [%]**	**FWHM average**	***S*(*n*-1) [%]**	***S*(aver) [%]**
1	0.0212	0.0730	0.0311	18.872	1.959	0.591
2	0.0338	0.2124	0.0906	19.213	2.974	0.897
3	0.0212	0.1912	0.0815	18.828	1.731	0.522
4	0.0211	0.9942	0.4239	19.042	1.072	0.323
5	0.0270	0.1605	0.0685	19.593	4.122	1.243
6	0.0351	0.2643	0.1127	19.009	3.271	0.986
8	0.0342	0.3624	0.1546	20.008	3.667	1.106

The standard errors of these values relative to the resolution of 19 keV warranted by the producer are in the range 5.3 % to 0.7%. Besides the FWHM of an energy peak spectra, the acquisition software MAESTRO-32 calculates, also, the alpha width at 20 % of the maximum height [FW(1/5)M] in order to be controlled the Gaussian shape of the peak.

It is known [6, 9, 10] that the alpha peak shape in an ion-implanted-silicon charged-particle detector is determined by characteristic physical processes that could induce asymmetries, distortions and tailing in the Gaussian shape of the peak. The good resolution of the alpha detector improves the shape of energy spectrum of the ^{241}Am, ^{233}U and ^{244}Cm sources, being resolved partially their energy lines.

Table 5 presents the experimental values of the FW(1/5)M, the Gaussian theoretical values and their associate relative standard errors for ^{241}Am, ^{233}U and ^{244}Cm sources, as average of the values corresponding to the levels from 6 to 10 with stabilized energy resolution values.

Table 5

Comparison between the theoretical and experimental peak shape of alpha sources

Disk source								
^{241}Am			**^{233}U**			**^{244}Cm**		
FW(1/5)M Gaussian [keV]	**FW(1/5)M Experiment** [keV]	**Relatie Error** [%]	**FW(1/5)M Gaussian** [keV]	**FW(1/5)M Experiment** [keV]	**Relative Error** [%]	**FW(1/5)M Gaussian** [keV]	**FW(1/5)M Experiment** [keV]	**Relative Error** [%]
29.53	32.02	-8.43	25.97	27.30	-5.12	25.35	26.61	-4.98
29.80	31.91	-7.08	25.76	27.04	-4.97	24.21	25.11	-3.72
29.18	31.61	-8.33	25.68	26.95	-4.95	24.96	26.25	-5.20
29.35	31.47	-7.22	25.67	26.88	-4.71	24.43	25.67	-5.10
29.32	31.38	-7.03	26.00	27.09	-4.19	24.52	25.71	-4.90

The values of the relative errors calculated for the main peaks of the alpha sources used for measurements emphasize the fact that compared with the Gaussian shape the broadening of the main alpha peak is more pronounced for the ^{241}Am source, respecting for those of ^{233}U and ^{244}Cm whose FW(1/5)M resolutions are evidently better.

The leakage currents were read out in real time for each chamber. The ion-implanted detectors have leakage currents in the tenths of nanoamperes (30-60 nA range). For chamber 7, it has been constantly observed the presence of a high leakage current (120–180 nA range) due to a mechanical damage of the detector's surface [4].

3.4. MINIMUM DETECTABLE ACTIVITIES [MDA]

The minimum detectable activities [MDA] of the alpha radionuclides ^{241}Am, ^{233}U and ^{244}Cm were calculated using the following equation [11]:

$$MDA[Bq] = \frac{3 + 4.65\sqrt{B}}{E_{ff}\,t},$$

where: B is the number of counts in the region of interest of the background, E_{ff} is the efficiency in cps/Bq and t is the counting time in seconds. The MDA values of the radionuclides ^{241}Am, ^{233}U and ^{244}Cm are declared in Table 6 for a measuring time of 1800 s. The results for MDA show an increasing of the MDA as function of the distance source-detector, because of the inverse proportionality relation between MDA and the counting efficiency [12].

Table 6

MDA for ^{241}Am, ^{233}U and ^{244}Cm

Level	**MDA [Bq]**		
	^{241}Am	**^{233}U**	**^{244}Cm**
1	0.006	0.004	0.005
2	0.010	0.008	0.009
3	0.016	0.013	0.14
4	0.023	0.019	0.021
5	0.032	0,030	0.030
6	0.043	0.037	0.038
7	0.055	0.052	0.050
8	0.071	0.068	0.068
9	0.084	0.084	0.089
10	0.108	0.108	0.100

4. CONCLUSIONS

The work conducted to fully characterize the alpha multi–chamber spectrometer was complex being performed many measurements in order to evaluate the resolutions of all eight ULTRA-AS detectors, to calibrate all the chambers of the spectrometer and to determine specific parameters. These parameters concern the radiation energy peaks that identify and quantify radionuclides, and include energy calibration, energy resolution, stability, repetability, peak-to-Compton ratio, and peak shape. The results obtained in the tests of repeatability in the same geometry conditions demonstrate the fact that the alpha spectrometer is correctly calibrated in energy and in efficiency and the measurement methods used are robust and can be validated.

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