LANTHANUM BROMIDE SCINTILLATION DETECTOR FOR GAMMA SPECTROMETRY APPLIED IN INTERNAL RADIOACTIVE CONTAMINATION MEASUREMENTS

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Apariţia de noi tipuri de detectori cu scintilatori anorganici $LaBr_3(Ce)$ şi $LaCl_3(Ce)$, a generat necesitatea testării lor pentru diferite aplicaţii. Ca o alternativă a detectorului NaI(Tl), utilizat, în mod tradiţional, în sistemele gama de monitorizare a I-131, în tiroidă, a fost considerat pentru caracterizare, un detector $LaBr_3(Ce)$, de dimensiuni 1.5" x 1.5". Rezultatele au fost evaluate, urmarindu-se conturarea unui raspuns referitor la potrivirea unui astfel de detector pentru detecţia I-131 în tiroidă. Au fost obţinute spectre comparative pentru cei doi detectori, NaI(Tl) si $LaBr_3(Ce)$, în aceeaşi geometrie de măsură.

By invention of the new types of nuclear detectors based on inorganic scintillators, $LaBr_3(Ce)$ and $LaCl_3(Ce)$, it appeared the opportunity they to be tested in different applications. As an alternative to the traditional NaI(Tl) scintillation detector, used in gamma spectrometric systems for I-131 monitoring in thyroid, in the present paper, it was considered for testing, a 1.5" x 1.5" $LaBr_3(Ce)$ detector. Spectra were measured and analysed for two detectors, NaI(Tl) and $LaBr_3(Ce)$ of comparative size, in the same measuring geometry. Discussion of the results and conclusions on the suitability of the new detectors for I-131 monitoring are presented.

Keywords: inorganic scintillators, thyroid monitoring, gamma spectrometry

1. Introduction

In vivo monitoring of the I-131 retention in thyroid is performed by gamma spectrometric measurements in the Human Monitoring Laboratory-Whole Body Counter from IFIN-HH for people working in nuclear medicine department from hospitals and in radiopharmaceuticals production activities [1-3]. The accuracy of these measurements is important for radiation protection evaluations, in the process of estimation of the committed effective doses and radiological risk. For this reason, a sensitive detector appropriate to detect photons with energies of several hundreds keV of I-131 is necessary, in conditions of good shielding,

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suitable collimation and calibration. The energy spectrum of I-131 decay contains the gamma lines of 80.2keV, 284.3keV, 364.5keV, 637keV and 723keV from which the 364.5keV one is dominant due to its high yield, of 0.812[4]. Taking into account the characteristics of I-131 energy spectra, room temperature detectors, namely, scintillation detectors based on NaI(Tl) crystals, with good efficiency and reasonable resolution, were considered appropriate for iodine monitoring in thyroid. These detectors were traditionally used, until now, in thyroid assessment gamma spectrometry systems [3]. In the last years, a new type of scintillation detector material $LaBr_3$(Ce), available in small sizes, with better resolution, became attractive for different applications, as high energy physics research, medical imaging, non-destructive testing, geological exploration, etc. [5-6].

The present paper presents, according to our knowledge, the first attempt to explore the suitability of the $LaBr_3$(Ce) detector for *in vivo* measurement of I-131 activity retention in thyroid, by evaluating the performances of such a detector, in comparison with a classic NaI(Tl) detector, of comparative size.

2. Characteristics of $LaBr_3$(Ce) detectors

The properties that make the $LaBr_3$(Ce) scintillator detector attractive for different applications based on gamma-ray spectrometry are:

- very good energy resolution
- very fast light output decay, enabling high count rate applications
- high temperature stability
- high gamma detection efficiency
- operation at room temperature
- promising technology for manufacturing crystal at larger sizes

These properties compared to those of the NaI(Tl) scintillator detector are presented in Table 1 (data from Ref.[5]).

Table 1

Comparison between the characteristics of $LaBr_3$(Ce) and NaI(Tl) scintillators of comparative size. Ref.[5]

Parameter	$LaBr_3$(Ce)	NaI(Tl)
Crystal density (g/cm^3)	5.29	3.67
Thickness for 50% attenuation of 662 keV gammas (cm)	1.8	2.5
Light yield (photons/MeV)	63000	39000
1/e decay time (ns)	26	250
Temperature coefficent of light output (%/C between 0C and 50C)	<0.02	-0.3
Resolution	3-4%	6-7%

Of particular relevance for the present work is the fact that $LaBr_3$(Ce) scintillator crystal has an important peculiarity given by the presence of La-138, a long-lived naturally occurring radioisotope of lanthanum. The La-138 has a radioactive half-life ($T_{1/2}$) of $1.05x10^{11}$ years and it acts as a gamma and beta self-contaminant of the scintillator crystal. Its specific decay is presented in Fig.1 [4].

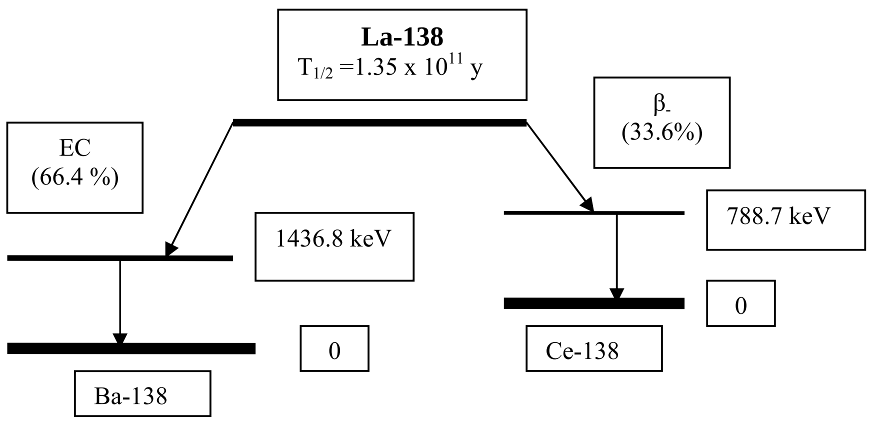

Fig.1 The La-138 decay scheme from Ref.[4]. See text for details.

La-138 decays by EC-electron capture (66.4%) to Ba-138 and by beta-minus decay (33.6%) to Ce-138., both being stable radioisotopes.
During the electron capture decay of the La-138 to Ba-138, is produced the gamma ray of 1435.8 keV (100%) and the Ba K x-ray peak, at the 34.7 keV.
The beta decay of La-138 to Ce-138, contributes itself with a beta continuum background with the endpoint energy of 255 keV and with the gamma ray of 788.7 keV (100%). This detector background radioactivity acts as a priory limiting factor in using this scintillation detector in low background measurements. It is the goal of the present paper to investigate this issue in details.

3. Experimental results

The experimental system we used for the measurements was a standard gamma spectrometry one, based on a 1.5“ x 1.5” $LaBr_3$(Ce) detector, type BriLanCe-380 manufactured by Saint-Gobain Crystals[7]. Its associated electronics include the phototube high voltage power supply 556 ORTEC module, the CANBERRA 2005 Preamplifier and analog and digital signal processing NIM modules. The data acquisition was performed using the dedicated multichannel analyzer emulation software, ORTEC- Gamma Vision 32 v-6 [8-11].

Radioactive standard point sources of Am-241(59.5keV), Ba-133(302.9keV, 356keV), Cs-137(661.7keV) and Co-60(1173.2keV, 1332.5keV) were used for the calibration of the spectrometer, in the energy range of 30 keV – 1600 keV. The pulse height spectrum obtained with these calibration sources is shown in Fig.2.

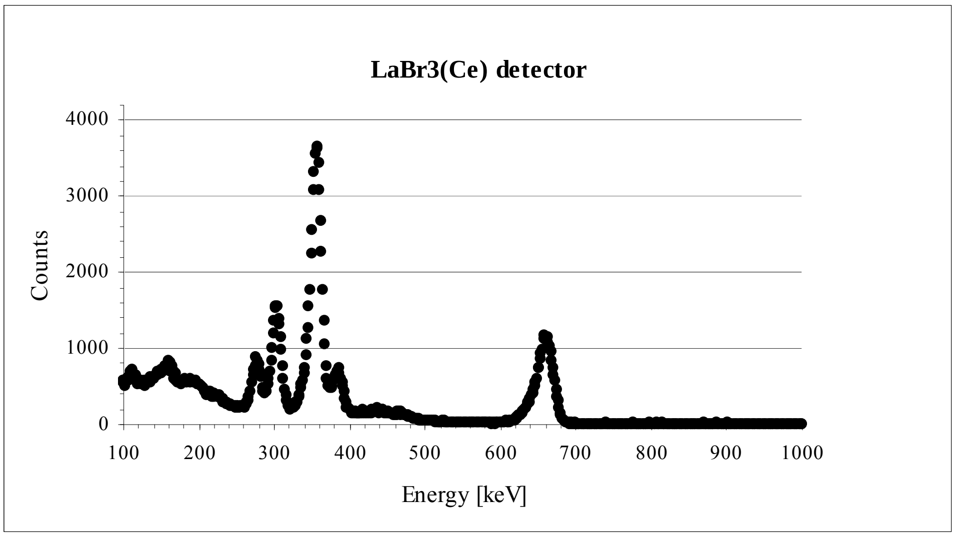

Fig.2 Mixed radioactive source calibration spectra. See text for details.

The energy calibration curve (Fig.3) can be described by a second order polynomial function. The excellent characteristics of the obtained fitting curve, namely, a correlation coefficient between experimental data and the output of the fitting curve, R=1, reveal the good linearity of the $LaBr_3$(Ce) spectrometer in the energy range we have considered.

The nominal energy resolution at the Cs-137 line energy of 661.7keV was measured for the $LaBr_3$(Ce) detector and it was found a value of 3.3%. This result confirms the excellent resolution of lanthanum bromide scintillation detectors comparing with the value of 6.8% we measured for a 2” x 2” NaI(Tl) scintillation detector. This significant difference in energy resolutions between the two types of scintillation detectors is shown, in detail, in Fig.4. The energy resolution of $LaBr_3$(Ce) detector is a characteristic that is considered for the evaluation of its suitability for the monitoring of I-131 in thyroid. The most important aspect on this evaluation is the effect of the La-138 decay lines on the background spectra, taking into account its decay scheme.

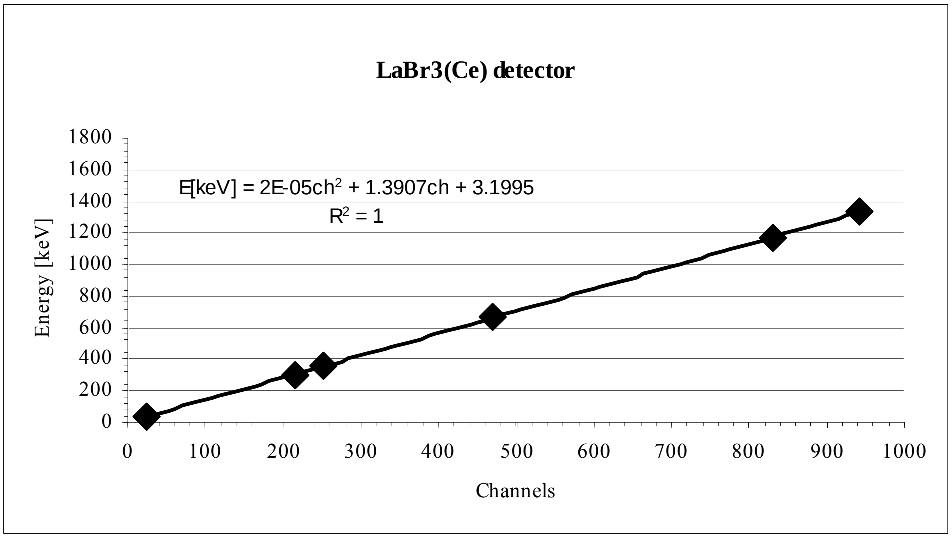

Fig.3 Energy calibration curve. R^2 is R-Square correlation coefficient between experimental data and the output of a fitted curve

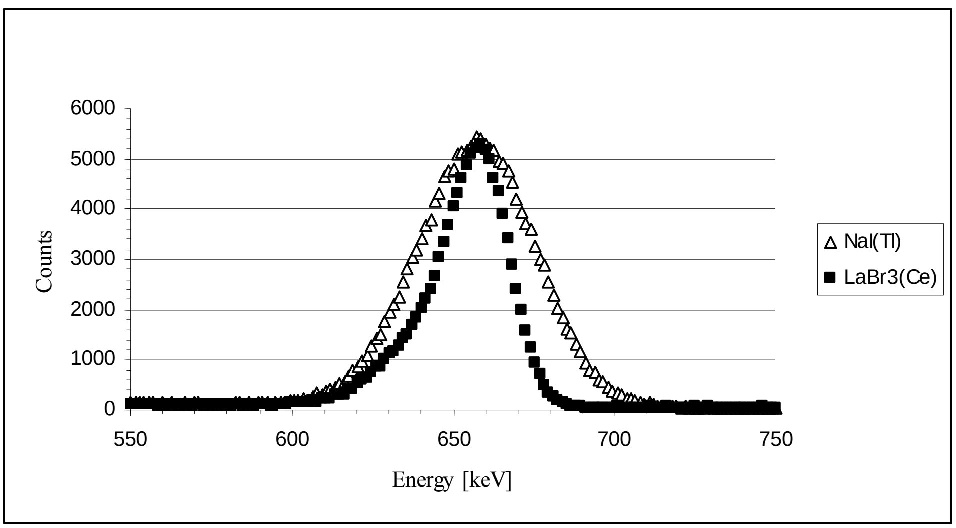

Fig.4 Cs-137 spectra (detail) for LaBr3(Ce) and NaI(Tl) detectors

In a qualitative analysis of the background spectra obtained with the thyroid gamma spectrometric system based on the 1.5" x 1.5" $LaBr_3$(Ce) detector, in the energy interval 30keV- 1600keV, there are two dominant peaks. The first peak, at the energy of 34.7keV, is due to the BaX-rays associated with the EC process. The second peak is obtained by the overlap of the 1435.8keV gamma ray, produced during the electron capture decay of the La-138 to Ba-138, with the 1460.8keV gamma ray of K-40 from the natural background.

In the same background spectra there are two energy intervals with specific characteristics. In the first one, 300 keV-700keV, could be distinguished the gamma rays of Ac-228, Tl-208, Pb-214 and Bi-214 from Th-232 and U-238 natural decay series. The second one has the energy range of 750 keV-1050keV in which the background spectra shape is due to the gamma ray of 788.7keV in coincidence with a beta particle.

From these two intervals, the first one, 300 keV – 700 keV, is of interest for our purpose, taking into account the value of 364.5 keV of the main energy gamma line of I-131 used for the estimation of activity by spectrometric methods.

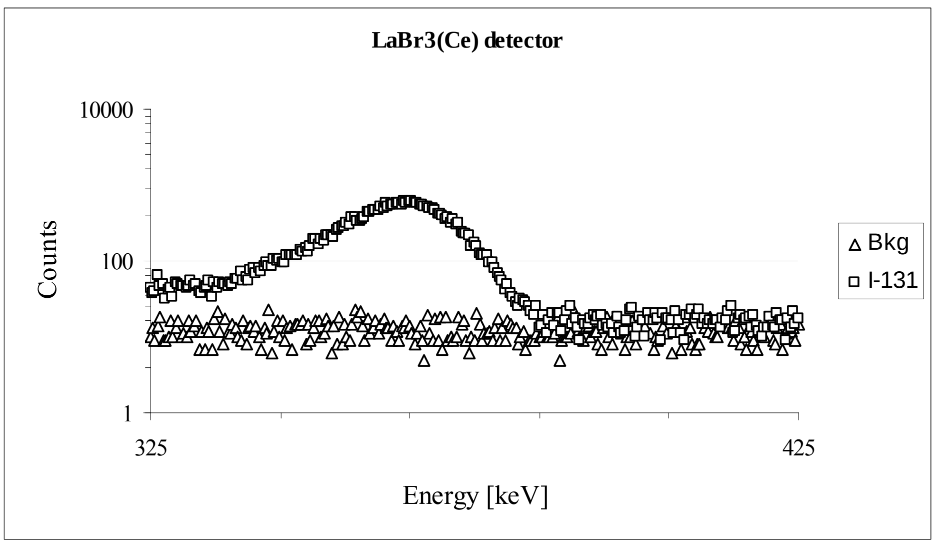

Fig.5 Background and I-131 pulse height spectra measured with LaBr3(Ce) detector. See text for details

A quantitative analysis of the pulse height background and I-131 spectra (Fig.5) was made in order to evaluate the suitability of the $LaBr_3$(Ce) to be used for the detection of I-131 in thyroid. From the background spectra were determined the count rates (cps) for the energy interval 300keV -700keV and for the interval 325keV – 425keV, corresponding to the region of interest of the I-131 peak.

The pulse height spectra of I-131 was obtained with a volume source of 20ml solution of I-131 inserted in ORTEC neck phantom and placed at 10 cm distance from the $LaBr_3$(Ce) detector[10]. Both spectra were considered to determine the Minimum Detectable Activity [MDA] for I-131 in the measuring conditions simulating the real monitoring conditions and thyroid geometry. Also, it was calculated the resolution of the $LaBr_3(Ce)$ detector at 364.5keV, the I-131 energy line of interest. All the results were declared in Table 2, together with those obtained, in the same measurement conditions, for the 2” x 2” NaI(Tl) scintillation detector. The uncertainties due to counting statistics are given in terms of one standard deviation (1σ).

Table 2

Results obtained from background and I-131 spectra of $LaBr_3$(Ce) and NaI(Tl) scintillators of comparative size.

Parameter	$LaBr_3$(Ce)	NaI(Tl)
cps in the energy interval (300keV-700keV)-background spectra	9.17 ± 0.03cps	4.91 ± 0.02cps
cps in the ROI of 364.5 keV I-131 peak – background spectra	1.223 ± 0.004	1.114 ± 0.003
MDA [Bq] for 364.5keV of I-131	20.2 ± 0.1	15.1 ± 0.1
Resolution at 364.5 keV [%] – I-131 spectra	3.7 ± 0.1	8.7 ± 0.1

It is noticed that the value of cps, in the energy interval (300keV-700keV), from LaBr3(Ce) background spectra is higher than that obtained with NaI(Tl) detector. This is due to the contribution to the background of the natural La-138 radioisotope, present in the Lanthanum crystal.

The values of cps for the energy interval corresponding to the region of interest of the 364.5keV of I-131 peak present a slight difference. This result is important because it reveals that the energy interval considered is not affected, by the La-138 radionuclide with a consistent contribution. Also, we observe the high difference between the cps for the whole energy interval (300keV-700keV) and that corresponding to the 364.5keV of I-131 peak. This is due to the presence of natural radionuclides, namely, the Bi-207, Ac-228, Tl-208, Pb-214 and Bi-214, with important branching ratios in the energy interval 450keV-600keV.

As expected, the different values for the MDA [Bq], calculated according to the formula from Ref.[12], at the 364.5keV line of I-131, for both detectors, demonstrate the dependence of the MDA on the background. The resolution of LaBr3(Ce) detector at the energy 364.5keV is, consistently, higher than the resolution of the NaI(Tl) detector.

6. Conclusions

The scintillation detectors remain a very good choice of the room temperature detectors to be used for *in vivo* measurements. Tests were made on a 1.5”x 1.5” $LaBr_3$(Ce) detector, using a standard NIM associated electronics. The results revealed the presence of the important effect of the long-lived naturally occurring radioisotope of lanthanum La-138 on the background spectra. Taking into account our specific application, there were determined relevant features of the detector, in the energy interval 325keV – 425keV, corresponding to the region of interest of the 364.5keV line of I-131. They were compared with the same characteristics determined for a NaI(Tl) scintillation detector of comparative size, considered as a common room-temperature detector used for iodine monitoring in thyroid. From the quantitative analysis of the background spectra, for both detectors, there were obtained valuable results. These consist in similar values for cps in the energy interval of interest and for the MDA corresponding to the 364.5keV energy of I-131. The excellent energy resolution of the $LaBr_3(Ce)$ detector, significant higher than that of the NaI(Tl), was confirmed. Our detailed evaluation is based on promising results for our peculiar application. As a conclusion, we can recommend the lanthanum bromide scintillation detectors for *in vivo* measurement of I-131 activity retention in thyroid, by gamma spectrometry.

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