# Environmental gamma spectrometry for Polish Nuclear Power Plant – preliminary considerations

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Abstract

Poland faces a choice of an environmental radiation monitoring systems (ERMS) for newly designed Nuclear Power Plant (NPP). Contemporary radiation monitoring systems can be based on gamma detector cooperated with Multichannel Analyzer (MCA). This solution will give a tool for very fast assessment of radiological situation around NPP and allows authority to react in case of radiological circumstances. The level of radioactive isotopes of cesium and iodine must be the subject of permanent monitoring because of their large amount in the hypothetical radioactive elements fallout from NPP. Thus ERMS must be high sensitive for the mentioned radionuclides and simultaneously should provide information about other radionuclides present in contingency radioactive fallout. The above radionuclides are taken into consideration because of their dominant role in the radioactive fallout after nuclear disasters in 'Grand Slam': Windscale, TMI, Chernobyl and Fukushima.

Two types of gamma detector were examined for their further implementation for the ERMS. The pre-calibrated HPGe detector was applied to cooperate with Inspectot2000 MCA while the second spectrometric system was based on LaBr scintillation probe combined with Tucan 8k MCA. The mockup experiment was fulfilled with stainless sample obtained by neutron activation using Am-Be source with strength of 1.5 $10^7$ n/s. In results of activation radionuclides of $^{58}$Co, $^{60}$Co, $^{51}$Cr, $^{59}$Fe, $^{54}$Mn occurred in the sample. They were a source of gamma photons with energy ranging from 320 keV to 1332 keV and several other belonging to that limits. Basing upon the above source of gamma radiation the efficiency calibration of LaBr probe was done and two series of separate measurements were performed on both spectrometric systems. During each gamma spectrum analyzes peak net area, total area, background, resolution, efficiency, and activity were determined. These data allows to estimate Lowest Limit of Detection (LLD) and Relative Minimum Detectable Activity ($MDA_r$). Both of those parameters are useful for assessment spectrometric system ability. The LaBr probe shows better LLD and $MDA_r$ than HPGe detector however its resolution is worse by one order of magnitude. Thus preliminary test shows that LaBr with MCA could be taken into consideration as the future environmental gamma monitor for newly designed Polish Nuclear Power Plant.

Key Words: environmental gamma spectrometry, LLD, $MDA_r$

## 1. Introduction

The lesson taken from nuclear power plant accidents shows that the steps taken into consideration in initial phase of radiation protection management is essentialand must include accurate previewing studies of the environment around power plant. Nowadays the microprocessor technique allows to monitor practically unlimited number of physical parameters on line enabling the authority final assessment of radiological situation.

Contemporary radiation monitoring of environment around nuclear power plant can be based on gamma spectrometry method cooperated with MCA technique. For the regular and emergency monitoring it is necessary to record presence of some radionuclides in a ground layer around power plant and, as well, in few remote places.

The level of radioactive isotopes of cesium and iodine must be the subject of permanent monitoring because of their large amount in the hypothetical radioactive elements fallout from NPP. Thus monitoring system must be high sensitive for the mentioned radionuclide and simultaneously it should be flexible to react on other radionuclides present in radioactive fallout. The above radinuclides are considered because of their dominant role in the radioactive fallout after four main nuclear accidents (see Table 1).

For such an aim two types of gamma spectrometric systems with two different detectors are taken into consideration as the future source of information regarding any radioactive releases from soon constructed Polish nuclear power plant. First of them is a spectrometric system with HPGe detector while the second is equipped with LaBr probe. The aim of this work is to compare the above mentioned gamma spectrometric systems for their further implementation for environmental gamma spectrometry

Table 1 Radioisotopes fallout [Bq] during four main nuclear power plant disasters: Windscale (1957), Three Mile Island (1979), Chernobyl (1986) and Fukushima (2011).

| Windscale [1], [2] | | TMI [3] | | Chernobyl [4] | | Fukushima [5] | |
|---|---|---|---|---|---|---|---|
| $^{131}I$ | 7.40E+14 | $^{3}H$ | 5.40E+12 | $^{131}I$ | 1.76E+18 | $^{131}I$ | 1.60E+17 |
| $^{137}Cs$ | 2.20E+13 | $^{131}I$ | 1.11E+12 | $^{134}Cs$ | 5.40E+16 | $^{134}Cs$ | 1.80E+16 |
| $^{103}Ru$ | 3.00E+12 | $^{133}I$ | 1.48E+11 | $^{137}Cs$ | 8.50E+16 | $^{137}Cs$ | 1.50E+16 |
| $^{133}Xe$ | 1.20E+15 | $^{134}Cs$ | 3.70E+05 | $^{103}Ru$ | 1.68E+17 | $^{103}Ru$ | 7.50E+09 |
| $^{210}Po$ | 8.80E+12 | $^{137}Cs$ | 1.50E+06 | $^{106}Ru$ | 7.30E+16 | $^{106}Ru$ | 2.10E+09 |
| $^{89}Sr$ | 3.00E+12 | $^{133}Xe$ | 3.07E+17 | $^{133}Xe$ | 6.50E+18 | $^{133}Xe$ | 1.10E+19 |
| $^{90}Sr$ | 3,33E+11 | $^{133m}Xe$ | 6.30E+15 | $^{89}Sr$ | 1.15E+17 | $^{89}Sr$ | 2.00E+15 |
| | | $^{135}Xe$ | 5.55E+16 | $^{90}Sr$ | 1.00E+16 | $^{90}Sr$ | 1.40E+14 |
| | | $^{135m}Xe$ | 5.20E+15 | $^{140}Ba$ | 240E+015 | $^{140}Ba$ | 3.20E+15 |
| | | $^{85}Kr$ | 1.80E+15 | $^{132}Te$ | ~1.1EBq | $^{127m}Te$ | 1.10E+15 |
| | | $^{88}Kr$ | 2.30E+15 | $^{95}Zr$ | 196E+015 | $^{129m}Te$ | 3.30E+15 |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| | | $^{58}Co$ | 1.48E+08 | $^{141}Ce$ | 196E+015 | $^{95}Zr$ | 1.70E+13 |
| | | $^{60}Co$ | 3.33E+07 | $^{144}Ce$ | ~116E+015 | $^{141}Ce$ | 1.80E+13 |
| | | $^{89}Sr$ | 2.22E+06 | $^{239}Np$ | 945E+015 | $^{144}Ce$ | 1.10E+13 |
| | | $^{90}Sr$ | 2.22E+06 | $^{238}Pu$ | 0.035E+015 | $^{239}Np$ | 7.60E+13 |
| | | | | $^{239}Pu$ | 0.03E+015 | $^{238}Pu$ | 1.90E+10 |
| | | | | $^{240}Pu$ | 4.20E+13 | $^{239}Pu$ | 3.20E+09 |
| | | | | $^{241}Pu$ | ~6E+015 | $^{240}Pu$ | 3.20E+09 |
| | | | | $^{242}Cm$ | ~0.9E+015 | $^{241}Pu$ | 1.20E+12 |
| | | | | $^{99}Mo$ | >168E+015 | $^{91}Y$ | 3.40E+12 |
| | | | | | | $^{143}Pr$ | 4.10E+12 |
| | | | | | | $^{147}Nb$ | 1.60E+12 |
| | | | | | | $^{242}Cm$ | 1.00E+11 |
| | | | | | | $^{127}Sb$ | 6.40E+15 |

## 2. Quantitative and qualitative method for comparison of different gamma spectrometry systems

Due to comparison of a few gamma spectrometry methods some quantitative estimators are strongly required. The Lowest Limit of Detection (LLD) as well as Relative Minimum Detectable Activity ($MDA_r$) are facing this duty and both seem to be considered useful factors for performing such a task.

2.1 Lowest limit of detection

LLD can be understood as the lowest signal reliably exceeds N time the background standard deviation. N is typically chosen 3-5. Assuming Poison distribution, the standard deviation is equal to square root of the collected counts. Then for background of known CPM (counts per minute), or Bq(CPM) the LLD(counts) can be expressed as follows:

$$LLD(counts) = N\sqrt{t \cdot Bg(CPM)} \qquad \text{equation 1}$$

where:

*t* is a counting time in min.

At the same time the LLD(CPM) can be expressed as follows:

$$LLD(CPM) = N\frac{\sqrt{Bg(CPM)}}{t} \qquad \text{equation 2}$$

Quantity of LLD can be converted to the lowest detectable DPM (disintegration per minutes) by dividing by AFEPE (Absolute Full Energy Peak Efficiency):

$$LLD(DPM) = \frac{N}{AFEPE(E_i)} \cdot \frac{\sqrt{Bg(CPM)}}{t} \qquad \text{equation 3}$$

This value is expressed in [Bq per sample]. The LLD depends on time and geometry of measurement. When time of measurement increases, the value of LLD decreases respectively. It is due to better statistic of the measurement. This situation is preferred because of the lower limit of the detection. The LLD, however, depends also on the registration efficiency. Degradation of the efficiency affects an increase of the detection limit as a result of inverse proportion of the LLD to the AFEPE.

2.2 Relative Minimum Detectable Activity

It has been shown [6] that the background changes with detector. The $MDA_r$ is proportional to the square root of the background area divided by the efficiency at the specified energy, as shown in equation 4.

$$MDA_r = \frac{\sqrt{FWMH(E_i)Bg(E_i)}}{eff(E_i)} \qquad \text{equation 4}$$

where:

| | |
|---|---|
| $FWHM(E_i)$ | Resolution at energy $E_{iy}$ |
| $Bg(E_i)$ | Total background at energy $E_i$ |
| $eff(E_i)$ | AFEPE at energy $E_i$ |

## 3. Measurements

The two gamma spectrometry systems were used for assessment of their future applicability for environmental gamma spectrometry. The first one has been equipped with scintillation probe made from LaBr and Tukan 8k MCA. The second system was based on HPGe detector cooperated with I2k MCA. Due to the fact that HPGe detector was armed by the manufacturer on its numerical characteristic [7], [8], [9] it has also been used for cross calibration of the LaBr detector.

At the first stage of the researches the mockup experiment was performed. The stainless disc with 51 mm diameter and 3 mm thick was activated by Am-Be neutron source with strange 1.5 $10^7$ n/s. Activation time ranged between 2 and 72 hours and after 2 hours of cooling the sample was measured on both spectrometric systems.In activated sample radionuclides of $^{58}$Co, $^{60}$Co, $^{51}$Cr, $^{59}$Fe, $^{54}$Mn were detected. In this way the stainless sample was the source of photons with different energies dispersed among energy scale which was the advantage of mockup experiment.

The quantitative analyzes obtained with HPGe detector system was the background for cross calibration of the LaBr probe. In results of the above procedure the efficiency calibration of scintillation probe was performed. Efficiency of gamma quantum registration represented as the dependency of Absolute Full Energy Efficiency versus energy is presented in Fig. 1.

The spectrum of activated sample measured on HPGe detector system is presented in Fig. 2 while Fig. 3 presents spectrogram of the same sample obtained on spectrometric system armed with LaBr scintillation probe.

For all detected radionuckides the total peak area, net peak area, detection resolution, detection efficiency and activity, were read out from the system report files. After that $MDA_r$ as well as LLD were both calculated based on equation 1 and equation 4 respectively.

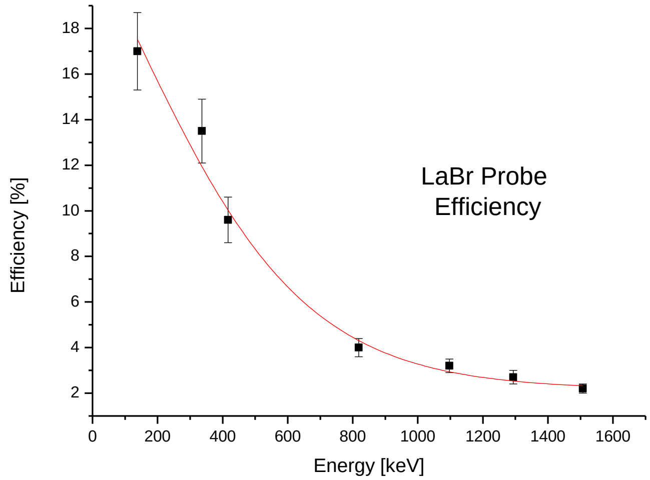


Fig. 1 The efficiency of the LaBr probe estimated during the mockup experiment

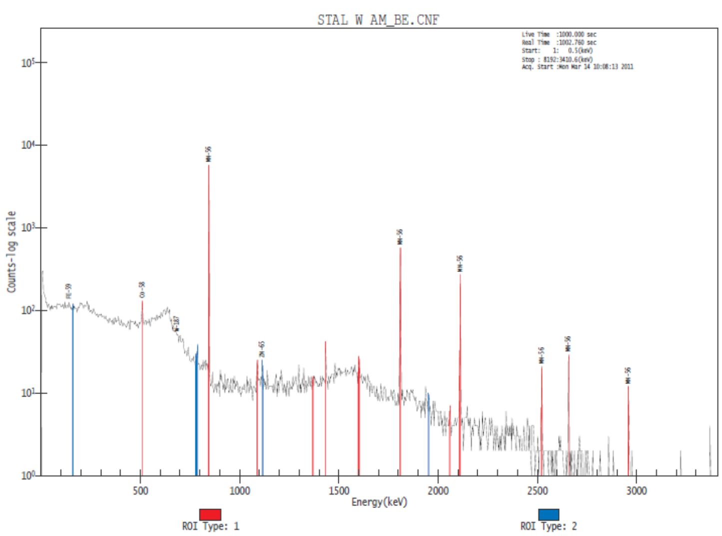


Fig. 2 Gamma-rays spectrum obtained during mockup experiment with a HPGe detector system

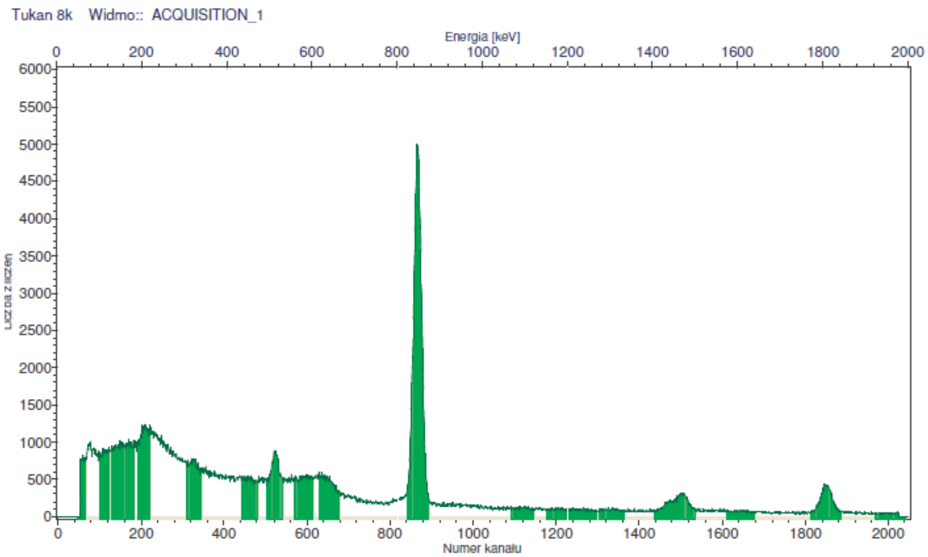


Fig. 3 Gamma-rays spectrum measured during mockup experiment, obtained with a LaBr scintillation probe.

## 4. Results

Table 2 presents results of quantitative analyze of activated stainless sample. Photons with energy ranging from 320 keV to 1332 keV and several other belonging to the mentioned diapason were registered. The most frequently photons emitted from $^{133}$I have energy 364 keV while energy 662 keV referes to photon released from $^{137}$Cs. It means that mockup experiment can represents contamination of the ground layer by most common radionuclides. To carry out further calculations of LLD and $MDA_r$ analysis of peak location, total peak area, net peak area, peak resolution and, radiation background were performed. The results of the mentioned analyzes for HPGe detector are presented in Table 3 which corresponds with spectrogram presented on Fig. 2. The results for LaBr detector which correspond to spectrogram on Fig 3 are presented in Table 4. The Relative Minimum of Detectable Activity for LaBr is higher than respective values for HPGe detector due to the better efficiency of the scintillation probe. The Lower Limit of Detection for LaBr probe is smaller in comparison with the HPGe (see Fig. 4). This means that the scintillate probe can reveal lower concentrations of radionuclides but some difficulty might appear during identifying these radionuclides as a result of the lower resolution of the probe in comparison with the HPGe detector resolution.

Table 2 Activities of radiunclides obtained in stainless sample as the results of two hours activation in Am-Be neutron source. Quantitative analyzes performed by pre-calibrated HPGe detector

| Nuclide Name | Id. Confidence | Energy (keV) | Yield (%) | Activity (Bq /Unit) | Activity Uncertainty |
|---|---|---|---|---|---|

| | | | | | |
|---|---|---|---|---|---|
| Cr-51 | 1.000 | 320.08 | 9.83 | 5.78E+01 | 2.20E+00 |
| Mn-54 | 0.996 | 834.83 | 99.97 | 1.67E+00 | 5.43E-02 |
| Co-58 | 0.996 | 511.00 | 29.8 | 7.09E+00 | 2.63E-01 |
| | | 810.76 | 99.45 | 7.84E+00 | 2.34E-01 |
| Fe-59 | 0.869 | 1099.22 | 56.5 | 8.74E-01 | 4.13E-02 |
| | | 1291.56 | 43.2 | 9.00E-01 | 4.97E-02 |
| Co-60 | 0.995 | 1173.22 | 100 | 3.24E-01 | 1.69E-02 |
| | | 1332.49 | 100 | 3.63E-01 | 1.81E-02 |

Table 3 Spectrometric characteristics of peak appearing in activation sample spectrogram presented on Fig. 2. For the measurement the HPGe detector was used.

| Radionuclide | Energy [keV] | FWHM [keV] | Total Area | Net Area | B-ground | Efficiency |
|---|---|---|---|---|---|---|
| Cr-51 | 320 | 1.24 | 30228 | 27482 | 2746 | 6.62E-02 |
| Co-58 | 511 | 1.36 | 11709 | 8721 | 2988 | 4.47E-02 |
| Co-58 | 810 | 1.52 | 19744 | 19193 | 551 | 3.08E-02 |
| Mn-54 | 834 | 1.53 | 4793 | 4187 | 606 | 3.02E-02 |
| Fe-59 | 1099 | 1.65 | 1272 | 910 | 362 | 2.44E-02 |
| Co-60 | 1173 | 1.68 | 960 | 634 | 326 | 2.32E-02 |
| Fe-59 | 1291 | 1.72 | 890 | 680 | 210 | 2.15E-02 |
| Co-60 | 1332 | 1.74 | 846 | 632 | 214 | 2.10E-02 |

Table 4 Spectrometric characteristics of peak appearing in activation sample spectrogram presented on Fig. 2. For the measurement the LaBr2 probe was used.

| Radionuclide | Energy [keV] | FWHM [keV] | Total Area | Net Area | B-ground | Efficiency |
|---|---|---|---|---|---|---|
| Cr-51 | 319.41 | 11.51 | 26464 | 952 | 25512 | 1.22E-01 |
| Co-58 | 512.17 | 16.41 | 29240 | 6384 | 22856 | 8.60E-02 |
| Mn-56 | 847.77 | 20.61 | 118535 | 104576 | 13959 | 4.70E-02 |
| Fe-59 | 1097.21 | 24.05 | 6702 | 94 | 6608 | 3.20E-02 |
| Fe-59 | 1293.93 | 33.12 | 6268 | 1095 | 5173 | 2.40E-02 |
| Mn-56 | 1810.39 | 28.2 | 16093 | 11421 | 4672 | 1.60E-02 |

Table 5 Value of LLD and $MDA_r$ for both spectrometric systems and radionuclides detected in respective quantitative analyzes

| Radionuclide | Energy [keV] | HPGe | | LaBr2 | |
|---|---|---|---|---|---|
| | | MDAr | LLD | MDAr | LLD |
| | | [Bq/sample] | | [Bq/sample] | |
| Cr-51 | 320 | 8.80E+02 | 5.39E-01 | 4.44E+03 | 1.05E-02 |
| Co-58 | 511 | 1.43E+03 | 3.50E+00 | 7.12E+03 | 1.57E-02 |

| | | | | | |
|---|---|---|---|---|---|
| Co-58 | 810 | 9.39E+02 | 4.02E-01 | | |
| Mn-54 | 834 | 1.01E+03 | 4.73E-01 | 1.14E+04 | 3.68E-02 |
| Mn-56 | 847 | | | 11412.17 | 0.036759481 |
| Fe-59 | 1099 | 1.00E+03 | 2.70E-01 | 1.25E+04 | 7.85E-02 |
| Co-60 | 1173 | 1.01E+03 | 2.43E-01 | | |
| Fe-59 | 1291 | 8.84E+02 | 1.36E-01 | 1.72E+04 | 1.18E-01 |
| Co-60 | 1332 | 9.18E+02 | 1.43E-01 | | |
| Mn-56 | 1810 | | | 2.27E+04 | 1.87E-01 |

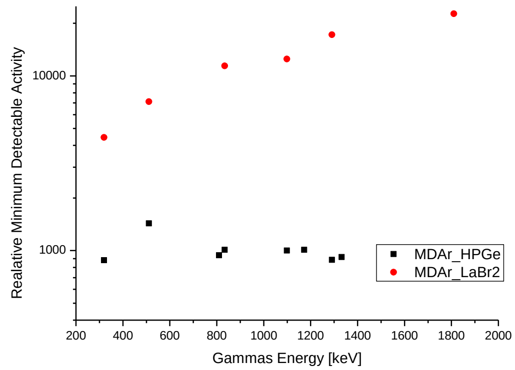


Fig. 4 Relative Minimum of Detectable Activity for LaBr probe and HPGe detector (in arbitrary units).

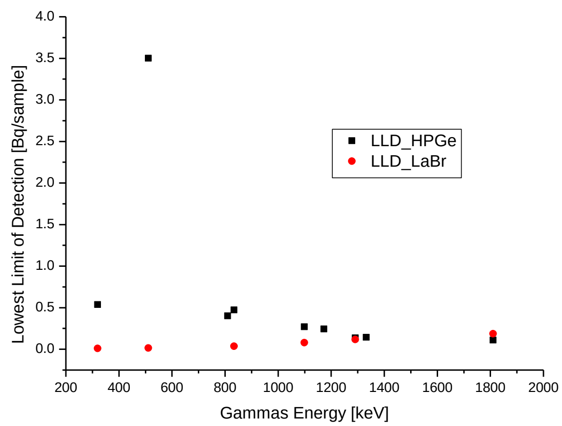


Fig. 5 Lowest Limit of Detection for both spectrometric systems. The lowest LLD the better system is.

## 5. Conclusions

The two gamma spectrometry systems, LaBr probe and HPGe detector, were compared for their future implementation for environmental gamma spectrometry in regular and emergency monitoring of the first Polish Nuclear Power Plant's vicinity. As quantitative estimators the Lowest Limit of Detection (LLD) as well as Relative Minimum Detectable Activity ($MDA_r$) were analyzed and compared after mockup experiment with activated stainless sample. The Relative Minimum of Detectable Activity of LaBr2 probe is higher than respective values for HPGe detector due to the higher efficiency of the scintillation probe. The Lower Limit of Detection for LaBr2 is smaller for scintillate probe, which means that, although, the scintillate probe can distinguish lower concentrations of radionuclides, some difficulty in identifying them might appear as a result of the lower resolution of the probe in comparison to the HPGe detector resolution.

The LaBr2 scintillation probe is suitable for environmental gamma spectrometry

## 6. Acknowledgments

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