Detectors on base of scintillation structures for registration of volumetric activities of gaseous and liquid media gamma radiation

V V Kadilin , V N Yurov, E V Ryabeva, V T Samossadny, E E Lupar , Yu A Trofimov, S V Kolesnikov, S B Chebishev and V O Nebolsin
National Research Nuclear University MEPhI (Moscow Engineering Physics Institute), Kashirskoe highway 31, Moscow, 115409

E-mail:EVRyabeva@mephi.ru

Abstract. The main aim of this research is the development and prototyping of the ionizing radiation detectors for the diagnosis of the physical processes used for monitoring the radiation situation at the thermal or fast neutrons reactors. In this article we present the experimental verification of applicability of the scintillation detectors based on $LaBr_3$(Ce) and $YAlO_3$(Ce). The experimental studies of the gamma-ray detection with several designs of the crystal scintillation detectors in gas and liquid are considered. It was shown that the measurement range in the liquid medium at the duration of one measurement of 100 seconds for ^{137}Cs equals from $3.79{\cdot}10^2$ Bq/l to $1.08{\cdot}10^8$ Bq/l for detector prototype based on $YAlO_3$(Ce).

1. Introduction

The aim of the research is the development and prototyping of ionizing radiation detectors for the diagnosis of the physical processes used for monitoring the radiation situation at the thermal or fast neutrons reactors. It is necessary to control the level of radiation exposure to humans and the environment in real-time measurements and to obtain a reasonable long-term forecasts of radiation exposure [1].

The proposed detector should have the highly sensitive radiometric channels and extended measurement ranges. The developed detectors must be capable of measuring the activity of fission gases ^{85}Kr, ^{85m}Kr, ^{88}Kr, ^{133}Xe, ^{135}Xe in the range from 10^4 Bq/m^3 to $3.7{\cdot}10^{14}$ Bq/m^3 ± 15% ensuring the identification of radionuclide in controlled gas-sample. It must provide the registration of gamma radiation of liquids to ensure the monitoring of volume activity of ^{137}Cs with limiting measured value $5{\cdot}10^7$ Bq/kg ± 15% and identification of radionuclide ^{134}Cs, ^{136}Cs, ^{138}Cs, ^{131}I, ^{133}I, ^{24}Na. The energy resolution for isotope ^{137}Cs should be no worse than 4–5%. The upper value of the operating temperature must be at least 80^0C. The dependence of the detection efficiency and energy resolution of the detector on the temperature must be insignificant [1,2].

Crystal detectors made from inorganic scintillators have high efficiency and high performance. They could be used in high temperature range, have good resistance to external mechanical influences and could be sufficiently large. The fist stage of the study was the choice and verification of applicability of the scintillators. After study of information on scintiallators [3] the following materials were chosen: $LaBr_3$(Ce) and $YAlO_3$(Ce).

Journal of Physics: Conference Series **675** (2016) 042010 doi:10.1088/1742-6596/675/4/042010

2. Prototypes of the scintillation detectors based on $LaBr_3$(Ce) and $YAlO_3$(Ce)

The detection unit for verification of applicability of the crystals for described tasks was developed.

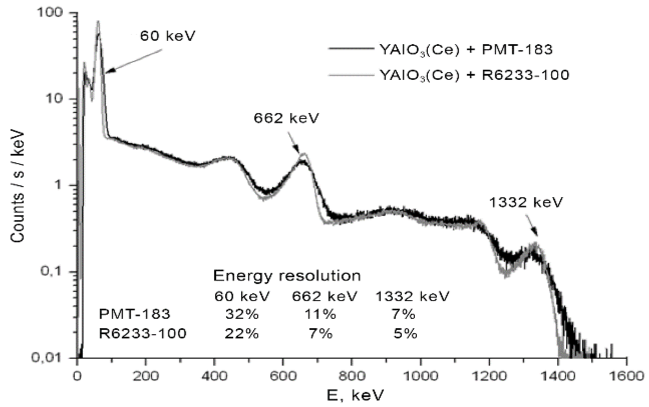

Figure 1. Energy spectrum with $YAlO_3$(Ce) scintillator.

For every scintillator the following experiments were done. The scintilator was irradiated by gamma-source with known spectra. The obtained waveform and the gamma spectra were recorded. The obtained spectra were analyzed and parameters of sensitivity were calculated. For testing the source ^{137}Cs isotope was used with activity ~ $0.6 \cdot 10^5$ Bq in the area of 4π. Figure 1 shows the spectra obtained by the crystal $YAlO_3$(Ce). The best energy resolution of the crystal was 5–7% for energy 1332 keV.Given the fact that this type of crystal has a low emission time – 27 ns, has a weakly pronounced temperature dependence of the light output and is not hygroscopic, it can be considered as a candidate for the implementation of the tasks of identification and activity measurements of radionuclide to a level of $3.7 \cdot 10^{17}$ Bq/m^3.

Energy resolution for crystal $LaBr_3$(Ce) at 662 keV line was equal to 4.2%. The scintillator $LaBr_3$(Ce) has a great light yield and, at the same time, small decay time (about 16 ns). A feature of the scintillator crystals $LaBr_3$(Ce) is relatively high intrinsic activity as a result of the decay of a radioactive isotope ^{138}La. To determine the contribution of the external background detector was surrounded by a layer of lead 10 cm thick. The measurements showed that the intrinsic background of crystal sample is $7.5 \cdot 10^{-3}$ counts/s/keV in the energy range 500–700 keV.

Figure 2. Prototype scintillation detector LABR1.

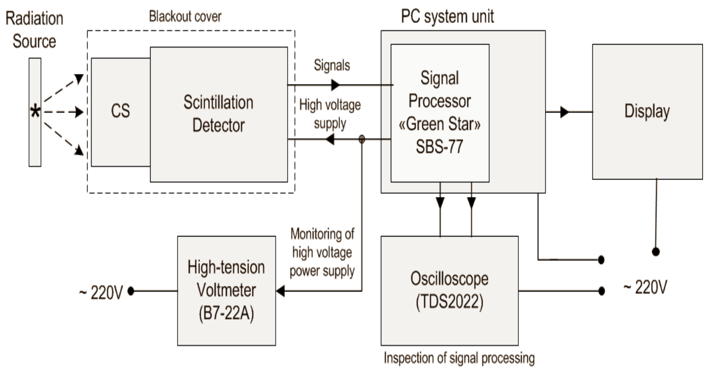

Figure 3. The test stand scheme.

3. Research of crystal scintillation detectors prototypes for the measurement of the activity of the gas environment

The source of ionizing radiation was set at the distance from the scintillation crystal's center. Pulse signals from PMT were processed by special board SBS-77 (firm "Green Star" [4]) installed in the system unit of a personal computer (PC). Form of signal is under the control by oscilloscope TDS-2022. The voltmeter V7-22A controls the PMT voltage. Modes of operation of SBS-77 and accumulated spectra are displayed on a PC monitor. The measurements of the gas activity were carried out. The sealed box with input and output connections for pumping gas through the test system was created for tests. The design of box allows to place inside it any of created detector prototypes. The scheme of the measuring stand is shown in figure 3.

The energy resolution of crystalline scintillation detectors LABR1, LABR2 and YAP1 were measured in the testing center for scientific equipment of Institute of Astrophysics NRNU MEPhI. Results of measurement are shown in figure 4.

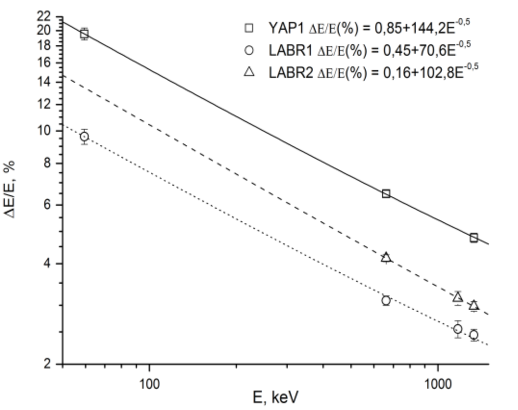

Figure 4. The energy resolution on the energy for the three detector prototypes.

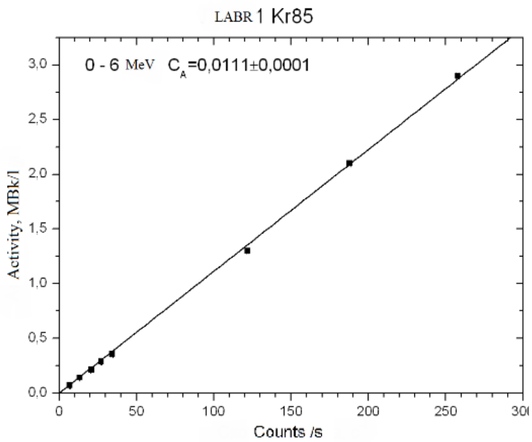

Figure 5. LABR1 counting rate on ^{85}Kr volumetric activity for full spectrum measuring range. Errors are within the data points.

The results confirmed the possibility of any of the prototypes made the sure identification of the radionuclides in the mixture of gaseous fission products ^{85}Kr, ^{85m} Kr, ^{88}Kr, ^{133}Xe, ^{135}Xe.
Crystal scintillation detectors based on $LaBr_3$(Ce) (LABR1 and LABR2) may provide identification of the nuclides ^{137}Cs, ^{134}Cs, ^{136}Cs, ^{138}Cs, ^{131}I, ^{133}I, ^{24}Na in media. Energy resolution of the crystal scintillation detector based on the $YAlO_3$ (Ce) YAP1 is not sufficient for such measurements.

The experiments showed linear dependence of the counting rate in complete absorption peak on the activity of gaseous ^{85}Kr (see figure 5). It is possible to estimate the minimal ^{85}Kr concentration which can be measured by the proposed detectors.

The *upper limit* is determined by the processing time of interaction which for the currently available processors it equal about 10^{-6} s. On the base of the experimentally obtained data the following values of the activity may be calculated for LABR1 and YAP1:

$$A_{upperLABR1} = 2.9\cdot10^9\ 10^6/250 = 1.16\cdot10^{13}\ \mathrm{Bq/m^3}\ ,$$

where $2.9\cdot10^9$ [Bq/m^3]/250[counts/s] is the rate for activity per one detector count (see figure 7), 10^6 – limit for detecting count rate. Using the same approach for YAP1:

$$A_{upperYAP1} = 2{,}86\cdot10^9\cdot10^6/160 = 1.8\cdot10^{13}\ \mathrm{Bq/m^3}.$$

If the measurement's time is 1s, number of events at the peak of total absorption will be about $5\cdot10^4$, which is enough to ensure accuracy about 1%.

The *lower limit* is determined by measuring the efficiency of the detector, the intensity of the background and the time of measurement. $I_{bacground}$=7.5·10^{-3}counts/s/keV for LABR1. When the measurement time 100s, the limit of the activity to be measured A_{lower}=2.1·10^{7} Bq/m^{3} for the prototype LABR1 and A_{lower}=4.5·10^{7} Bq/m^{3} for the prototype of YAP1.

4. The ability of models of crystal scintillation detector to register gamma radiation of liquids

The supposed liquid medium in most cases is water. The main peculiarity for registration of uniformly distributed radiation in the liquid medium from the radiation of the gas is a relatively high rate of absorption and scattering it in the medium itself.

In the fist approximation, the activity of uniformly distributed source may be presented:

$$A_1 = \frac{A_0}{\mu_{H_2O} l}(1 - e^{-\mu_{H_2O} l})$$

where A_1 – activity emerging from the source [Bq]; A_o – activity in the source [Bq]; μ – linear absorption factor [cm^{-1}]; l – thickness of the radiating source [cm].

The measurement with crystalline scintillation detectors should be conducted in a non-contact method and under moderate temperature conditions. This type of measurements improves the reliability of electronic components, and makes it easy to carry out preventive and calibration work with radiation detectors. The portion of unscattered radiation depending on liquid media and gamma energy may reach the detector in non-contact method. For ^{137}Cs isotope portion of unscattered radiation emitted from measuring volume of radioactive water with thickness 10 cm through 2 mm wall is approximately equal to 0.6.

The spectra of the following isotopes with the following main energy lines were experimentally investigated and analysed: 661.7 keV (^{137}Cs); 604.7 and 795.8 keV (^{134}Cs); 340.5, 818.5 and 1048.07 keV (^{136}Cs); 462.7 and 1048.07 keV(^{138}Cs); 364.48 keV(^{131}I); 529.8 and 875.3 keV(^{133}I);1368 and 2754 keV(^{24}Na). From the analysis of data on energy resolution it can be concluded that all listed lines can be identified with the detectors based on the $LaBr_3$(Ce) LABR1 and LABR2. Prototype based on the $YAlO_3$(Ce), YAP1 better be used for an energy of 662 keV due to its small thickness. Also because of lower energy resolution, it isn't able to identify the radionuclide ^{131}I in the presence of ^{136}Cs and ^{24}Na radionuclides.

For the prototype of the detector LABR1 measuring range in the liquid medium when the duration of one measurement – 100s for ^{137}Cs may be calculated on the base of results for gaseous media and taking into account correlated factors on portion of transmitted radiation. The calculated boundaries for measuring range are the following:

A_{lower} =1.77 10^{2} Bq / l; A_{upper} =6.96 10^{7} Bq / l.

For the prototype of the detector YAP1 measuring range in the liquid medium at the duration of one measurement of 100s for ^{137}Cs are the following:

A_{lower} =3.79·10^{2} Bq/l; A_{upper} =1.08·10^{8}Bq/l.

5. Conclusion

The prototyping and experimental investigation of the detectors based on crystal $YAlO_3$(Ce) and $LaBr_3$(Ce) are conduced. It is shown that when the measurement time is 100s the top level of measured activity of the gaseous ^{85}Kr medium is about 10^{7} Bq/m^{3}. The measuring range for models of detectors based on $YAlO_3$(Ce) and $LaBr_3$(Ce) in a liquid medium (water) for a duration of one measurement 100s for ^{137}Cs is as follows: A_{lower} = 3.79 10^{2} Bq/l and A_{upper} =1.08·10^{8} Bq/l and Alower=1.77·10^{2} Bq/l and Aupper =6.96 10^{7} Bq/l for prototypes correspondently.

Acknowledgments

Authors wish to acknowledge partial support from MEPhI Academic Excellence Project (contract № 02.a03.21.0005, 27.08.2013).

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