

SECTION 4

IRRADIATION INSTALLATIONS, DIAGNOSTIC AND RESEARCH METHODS

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PROPORTIONAL DETECTORS OF GAMMA-RADIATION BASED ON HIGH PURITY XENON GAS FOR RADIATION MONITORING

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This article is dedicated to a detailed investigation of properties of a proportional counter based on the Xe+2%CH₄ mixture. A setup for filling the detectors with high purity Xe gas and Xe-based gas mixtures was designed and made. An optimized design of the proportional counters was developed and investigated. The design parameters, such as the material and diameter of the anode, the number and material of the gas mixture inlets, composition and pressure of the gas were improved. The operating regimes of the proportional counters, as well as their thermal stability, were investigated.

INTRODUCTION

The development of new devices for γ -radiation monitoring would allow solving such urgent problems for Ukraine as the safety of operation of radiation-hazardous objects, which include: nuclear power plants; experimental nuclear facilities; enterprises for the production and processing of nuclear materials; enterprises that collect, store, transport, process and bury radioactive waste; warehouses and storages of radioactive materials and waste. Optimal radiation monitoring of critically important objects requires further improvement of monitoring during circulation and storage of nuclear materials, radioactive substances and radioactive waste to ensure radiation safety and, in addition, guarantees of nuclear non-proliferation.

Gamma-ray spectrometers/dosimeters based on xenon gas possess a number of advantages, compared to other devices. Gas-filled detectors, and those based on Xe in particular, have long operation life, and ability to work in a wide temperature range.

The most common configurations of gas-filled detectors include ionization chambers and proportional counters. Ionization chambers based on high-pressure Xe (HPXe) for γ -radiation detectors have been developed (e.g. [1, 2]) and used in various applications, which include radiation monitoring, space applications, non-proliferation, etc. The energy resolution of such spectrometric detectors reaches $\sim 1.7\%$ (for the 662 keV γ -quanta) [2].

The proportional mode of the detector operation differs from the operation in the ionization mode. The main idea behind the operation of a proportional counter is the process of multiplication of the charge carriers produced during the primary ionization, as illustrated in Fig. 1. The multiplication happens in the very non-uniform electric field region near the anode surface. The electric field in the gas volume of the detector is created by applying a potential difference between the detector's anode and the cathode.

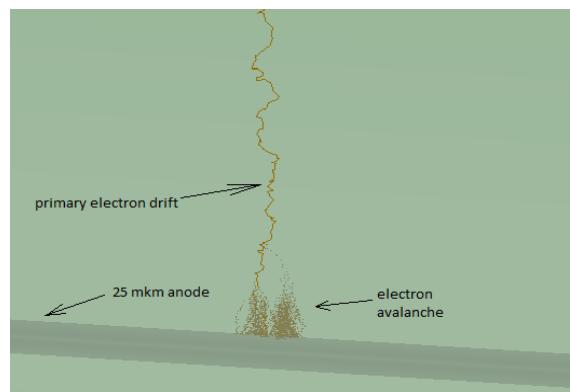


Fig. 1. An example of a simulated via Garfield++ [3] avalanche produced by a single electron in a cylindrical proportional counter filled with pure Xe. One can see the drift of the primary electron and the electron multiplication produced close to the anode surface

The amount of charge multiplication is called the gas gain. If the multiplication value is 1 then the operation mode changes from the proportional counter mode to the ionization chamber mode, and the output signal is produced by the primary ionization charge carriers only.

The niche of the proportional counters is between the high-precision expensive ionization chambers, and the Geiger-Muller detectors, which are quite cheap. Proportional counters based on Xe are smaller than the respective ionization chambers, and have reasonable sensitivity to radiation and spectrometric energy resolution. They are significantly less expensive, and, therefore, much more available.

The development of gas-filled proportional counters based on Xe and its mixtures is described in this work. A compact installation for filling the detectors with high-purity gases was developed and built. Xe-based proportional counters with modern design were developed and tested.

THE INSTALLATION FOR DETECTOR FILLING

The particular detector filling installation (the schematic is shown in Fig. 2) was designed for the transfer of gases of purity up to 6N (99.9999%) and is based on “Spectron Gas Control Systems GmbH” components. It allows the inlet connection of two high purity gas cylinders at pressure of up to 300 bar, Xe and CO₂, for instance, to create detecting medium gas mixtures of user’s choice. The pressure from the cylinders is reduced to up to 10 bar via the reducers, and the two gasses are mixed to produce a needed detector medium mixture. The installation consists of a number of two-way and three-way valves, reducers, pipes and monometers designed to work with high-purity gases. The installation is connected to the turbo-molecular pump Pfeiffer HiCube 80 Eco, and a vacuum sensor Pfeiffer PBR 260. Prior to gas filling, the installation and the connected detector are pumped out in order to perform degassing of the internal walls of the installation and the detector.

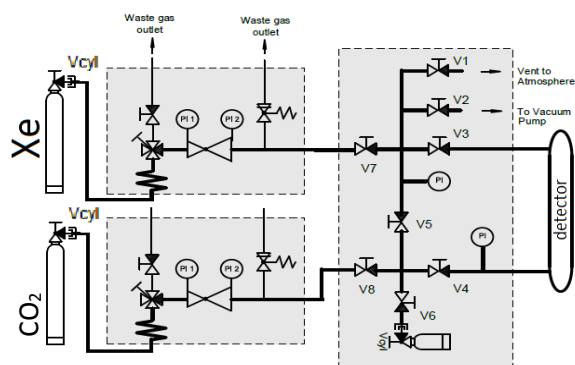


Fig. 2. The schematic of the installation for proportional counters filling

Fig. 2 shows the connection of a detector via two inlet tubes; the gas flow-through regime can be used during degassing and filling in this case. Using just one inlet tube is also possible.

THE PROPORTIONAL COUNTER BASED ON HIGH PURITY Xe

The design of our proportional counters has gone through several iterations, including those described in [4]. Fig. 3 shows the schematics for one of the first prototypes (a) and one of the most recent versions (b).

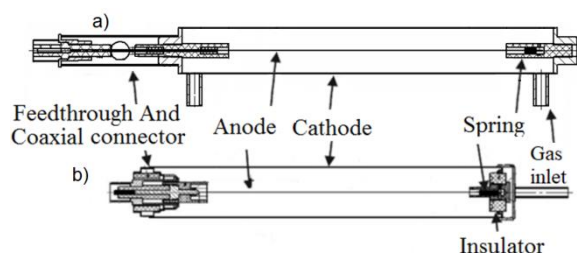


Fig. 3. The design schematics of a prototype (a) and newly developed (b) proportional counters

Parameters of the most recently developed and manufactured proportional counter

Parameter	Value and units
Working anode length	150 mm
Anode diameter	25.30 μ m
Cathode diameter	25.5 mm
Cathode wall thickness	0.3 mm
Anode voltage	1.2 kV
Gas	Xe+2%CH ₄
Pressure	1.25 bar

The counters’ main components are the cylindrical cathode tube (steel 12X18H10T), wire anode (CuBe), gas inlet tubes, an electrical feedthrough and a connector. The anode is tensioned with a spring. As can be seen, the recent design features an optimized geometry, feedthroughs from “Testbourne Ltd”, ceramic holders, it also has only one gas inlet tube made of copper, whereas the prototypes had two steel inlets.

The main design and operation parameters of the most recent proportional counter under consideration are shown in Table.

The previous detector design, which featured two gas inlet tubes and the anode diameter of 60 μ m was reported in a previous study [4], in which pressure values of up to 3 bar and several gas compositions have been evaluated. Higher pressure and anode diameter required relatively high anode voltage ($\sim$ 2500 V). In order to reduce this characteristic, in this study, the gas mixture Xe+2%CH₄ at 1.25 bar pressure and the 30 or 25 μ m CuBe anode wire were used, which led to a reduction of the operation voltage down to $\sim$ 1200 V.

THE DETECTOR FILLING

For filling of the proportional counters, the high purity xenon gas, doped with, CH₄, was used in this study. The gas purity was $\sim$ 99.999%. Typically, the detector is mounted onto the filling installation, after which the vacuum pumping down of the system is carried out for several hours. This step is followed by the degassing of the detector. For the degassing, flexible heating elements are wound onto the detector. The heating is carried out at temperatures from 110 to 250 $^{\circ}$ C, while pumping down, for tens of hours, and the effect of the environment temperature on the detector performance will be reported here. A detector is then filled with a desired gas mixture. The detector reported here was filled with Xe+2.08%CH₄ up to 1.25 bar.

MEASUREMENTS

The counting and spectrometric characteristics of the detector were studied by using a high performance measurement channel. It is comprised of an Ortec 142AH preamplifier, a 672 shaper amplifier, and a 927 ADC. Voltage up to 1500 V was applied to the anode by using an Ortec 659 high-voltage power supply through an RC filter. The gain value was 100, the time constant of the shaping amplifier was 3 μ s. The count rate and amplitude distributions of the detector signal were measured by using standard γ -sources ²⁴¹Am, ¹³⁷Cs, ¹⁵²Eu, ¹³³Ba.

The example spectra obtained at the anode voltage of 1200 V are shown in Fig. 4.

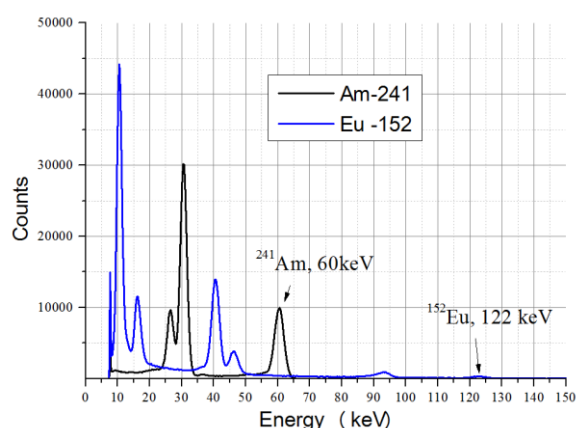


Fig. 4. Spectra measured with the proportional counter filled with Xe+2%CH₄ to 1.25 bar at 1200 V

By integrating the spectra of ²⁴¹Am measured at different anode voltages and dividing the obtained total count values by the spectrum acquisition time (typically 5 min), the pulse count rates were obtained (Fig. 5,a). As can be seen from the figure, the count rate increases when increasing the value of the anode voltage up to ~950 V, after which it reaches a relatively flat plateau.

By using the ~60 keV photopeak position in the ²⁴¹Am spectrum, the values of the gas amplification (gas gain) factor were calculated by the method described in [5] (see Fig. 5,b). Energy resolution values (see Fig. 5,c) at this photopeak were calculated from the ratio of the full width (in keV) at half maximum (FWHM) to the photopeak energy. As can be seen, the gas gain begins to increase exponentially (linearly on a logarithmic scale) starting from the anode voltage of ~1200 V. At the same voltage, the resolution of the detector reaches a minimum. The best energy resolution of the considered detector is ~6.5% for the γ -quanta energy of 60 keV. Thus, the value of the anode voltage of 1200 V was chosen for the operation of this detector in the spectrometric mode. For the pulse counting mode, the anode voltage values from 950 V and above are suitable.

TEMPERATURE STABILITY

As opposed to solid-state semiconductor detectors, gas-filled detectors have such an advantage as the intrinsic stability of operation at different temperatures [6]. The stability of our proportional counter filled with Xe-2%CH₄ to 1.25 bar was investigated. For this purpose, the spectrometric measurements were carried out at varied temperatures while holding the detector and the γ -source in the hot-cold chamber "Espec SU-241", all the electronics being at room temperature.

For convenience of the experiment, metallic radioactive sources were specially prepared and used: ¹⁷⁹Ta in the first experiment with the detector, which had 30 μ m BeCu anode and was degassed at 110 °C, and ⁵⁷Co in the second experiment with the detector, which had 25 μ m BeCu anode and was degassed at 250 °C. Radioactive ¹⁷⁹Ta was obtained by exposing a Ta metal foil to a beam of electrons in the 40 MeV electron accelerator at KIPT, and ⁵⁷Co was obtained by

exposing a Ni foil to the same electron beam; this procedure is described in [7].

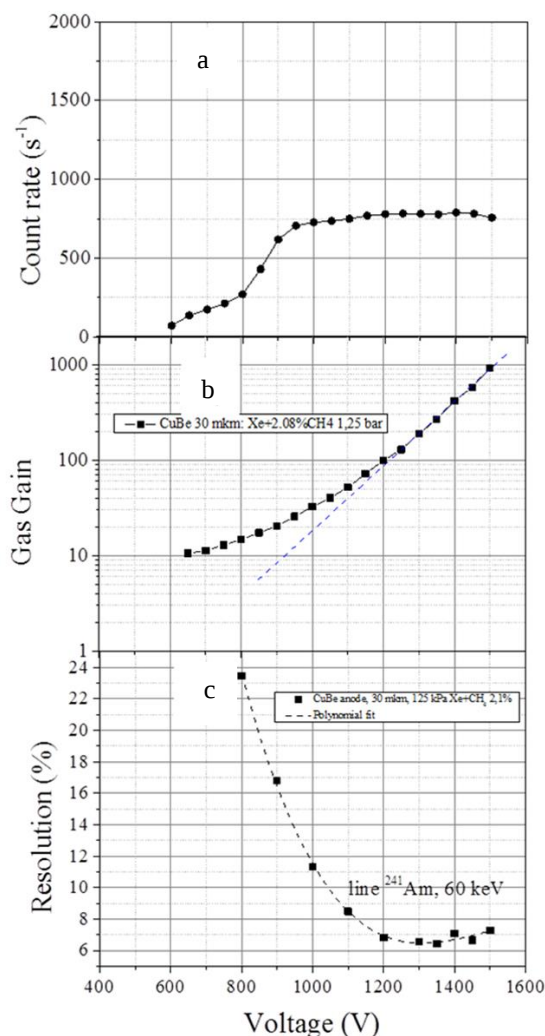


Fig. 5. Characteristics of the proportional detector as a function of the anode voltage: a – count rate; b – gas gain; c – energy resolution at 60 keV. Symbols correspond to the experimental data, the dashed lines are the exponential fit in (b) and polynomial fit in (c)

A typical room-temperature γ -spectrum of ¹⁷⁹Ta is shown in Fig. 6. It has several lines with good intensity in the observed energy range: 55.79 keV – 23.7%, 54.61 keV – 13.6%. In the presented spectrum, these lines are seen as one line at ~55 keV. ¹⁷⁹Ta γ -spectra measured at varied temperatures with a detector, which was "degassed" at 110 °C, are shown in Fig. 7. The spectra at low temperatures (to -40 °C) were also measured (not shown on the plot for the plot clarity).

The respective variation of the position of the combined ~55 keV photopeak versus measurement temperature is shown in Fig. 8. As can be seen, the peak position is fairly stable in the temperature range from -40 to +40 °C, with a very slight increase of the peak position with temperature, and at higher temperature the peak is significantly shifting to lower channel numbers. After bringing the detector back to room temperature, its performance is practically entirely restored.

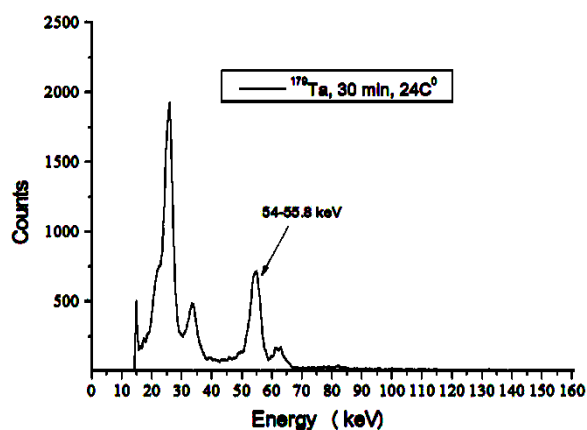


Fig. 6. Spectrum of ^{179}Ta measured at room temperature; x-axis is calibrated in the units of energy

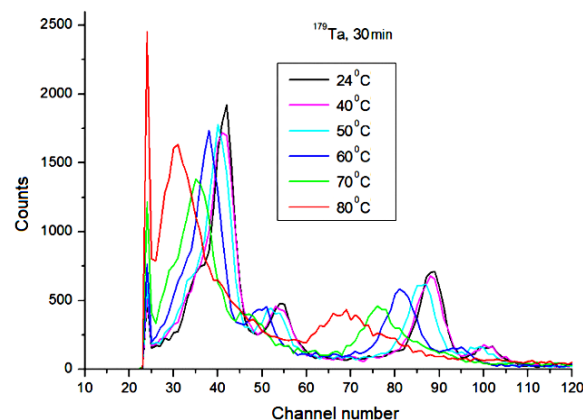


Fig. 7. ^{179}Ta γ -spectra measured at varied temperatures with a detector, which was "degassed" at 110 °C

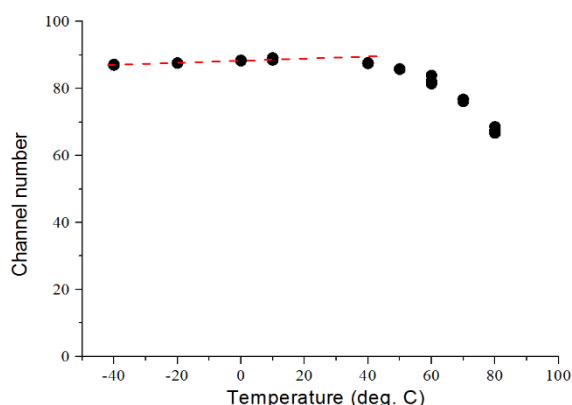


Fig. 8. Change of the position of the ~ 55 keV photopeak obtained from the data of Fig. 7

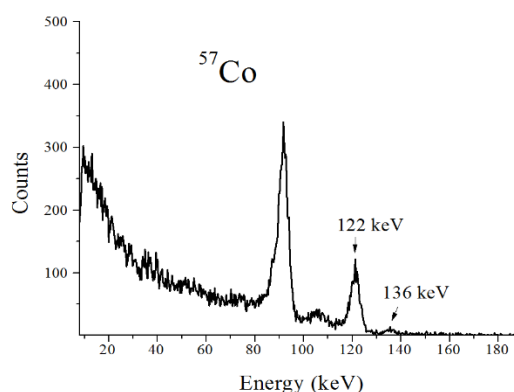


Fig. 9. Spectrum of ^{57}Co measured at room temperature; x-axis is calibrated in the units of energy

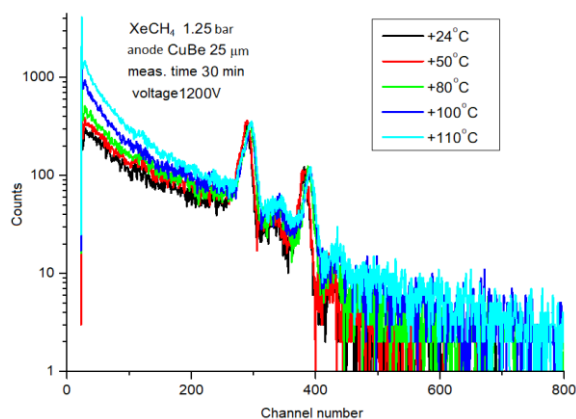


Fig. 10. ^{57}Co γ -spectra measured at varied temperatures with a detector, which was "degassed" at 250 °C

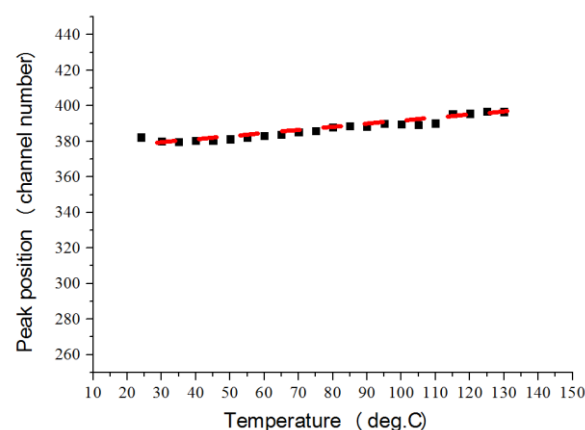


Fig. 11. Change of the position of the 122 keV photopeak obtained from the data of Fig. 10

A typical room-temperature γ -spectrum of ^{57}Co measured with a detector, which was "degassed" at 250 °C is shown in Fig. 9. It has the following γ -lines with good intensity in the observed energy range: 122.06 keV – 85.6% and 136.47 keV – 10.68%. The 14.4 keV line did not show in the measurements. The respective γ -spectra measured at varied temperatures with this detector are shown in Fig. 10, and the respective variation of the position of the 122 keV photopeak versus measurement temperature is shown in Fig. 11. The increase with temperature of the

background counts in the whole energy range is observed in the spectra. It can also be seen that the peak position is fairly stable (no shift to the left) up to 130 °C. There is somewhat of a shift of the peak position towards higher values with increasing temperature (~ 2% over 100 °C). Once the detector is calibrated and the operation temperature is being monitored during the spectral measurements, the shift of the peak and the impact of the background can easily be corrected.

DISCUSSION

The collection of charge, which is created in the ionization events, at the anode strongly depends on the voltage applied between the anode and the cathode, as illustrated in Fig. 5. The collection of some charge can be observed already at ~ 600 V for the given detectors, while full collection is achieved for voltages above 950 V. In Ref. [8], the relation of the width of a photopeak observed in a spectrum, which was obtained via a gas-filled detector, to the gas gain deviance is given by the equation:

$$\text{FWHM} = 2.35 \cdot \sqrt{(F + f) \cdot W \cdot E_\gamma},$$

where F is Fano factor of the gas; f is the variance of the gas gain (G); W is the mean energy for the electron-ion pair generation, and E_γ is the energy of the γ -photon.

The average ionization energy in gaseous Xe is relatively low, < 21 eV per electron-ion pair [9]. In addition, Xe has low Fano factor – 0.13 ± 0.1 [10]. The gas gain deviance f can be much higher than the Fano factor. Thus, lower f at lower G values, $G=1$ being the lowest, should result in better energy resolution. The extrapolation of the exponential part of Fig. 5,b suggests that $G=1$ should be achieved for $V \leq 600$ V. It is clear from Fig. 5,a that charge collection is not sufficient at such voltages, and, in order to achieve full collection in this regime, even yet higher purity gas and better gas filling techniques, than those described in this work, are required.

Gas-filled detectors are known to have an excellent temperature stability of operation. For example, the characteristics of ionization chambers based on HPXe were shown to be stable over a wide temperature range (from $+20$ to $+180$ °C) [11]. Our Xe-filled proportional counters also have quite good temperature stability, which seems to depend on the quality of the detector “degassing” prior to gas filling. The likely reason for the temperature-unstable behavior of the detector “degassed” at 110 °C is that a significant number of impurities, such as molecules H_2O , O_2 , etc. remain on the detector inner walls after such a “degassing” procedure. When increasing the temperature during measurements, these molecules desorb from the detector walls and act as gas impurities, which decrease the charge carrier lifetime, resulting in less charge collection on the anode and “left-shifting” of the spectra. “Degassing” of the detector at 250 °C cleans the inner detector walls from such impurities much better, and superior temperature stability is obtained. There is a clear trend in increasing of the number of background noise pulses with the increasing temperature, as well as minor peak shifting towards higher energies ($\sim 2\%$ over 100 °C), which could be taken into account during calibration. The reasons for the observed changes need to be further investigated.

CONCLUSIONS

Several versions of the γ -ray proportional counters based on high-purity xenon gas were designed and built in KIPT. A compact setup for the filling of the detectors with xenon, or mixtures thereof, has been designed and built, which allows filling of the detectors with high-purity gases at pressure values from 0.2 to 10 bar. The

setup provides some opportunity for heating and degassing of internal surfaces of the detector. However, an improvement in this system and procedures would likely reduce the number of impurities in the gas detecting medium and improve some of the detector characteristics. It was demonstrated that the temperature and time, at which the degassing procedure is carried out, impacts the performance stability of the detector when it operates at elevated temperatures up to 130 °C. Heating the detector (“degassing”) at 250 °C prior to filling it with the gas mixture ensures good operation temperature-stability.

Our latest γ -ray proportional counter design, which has a $25 \dots 30$ μm anode wire and $> 99.999\%$ pure $\text{Xe}+2\%\text{CH}_4$ gas mixture at 1.25 bar features the reduced operation voltage (~ 1200 V) and the energy resolution of 6.5% (at 60 keV), with the sensitivity range up to γ -ray energies of ~ 200 keV.

Obtaining complete charge collection at lower anode voltages, than those reported in this work, could improve the energy resolution.

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ПРОПОРЦІЙНІ ДЕТЕКТОРИ ГАММА-ВИПРОМІНЮВАННЯ НА ОСНОВІ ВИСОКОЧИСТОГО ГАЗУ КСЕНОНУ ДЛЯ РАДІАЦІЙНОГО МОНІТОРИНГУ

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Детально досліджено властивості пропорційного лічильника на основі суміші Хе+2%СН₄. Розроблено та виготовлено установку для заповнення детекторів газом Хе високої чистоти та газовими сумішами на його основі. Розроблено та досліджено оптимізовану конструкцію пропорційних лічильників. Покращено конструктивні параметри, такі як матеріал і діаметр анода, кількість і матеріал входів газової суміші, склад і тиск газу. Досліджено режими роботи пропорційних лічильників, а також їх термічну стійкість.