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GAS-FILLED GAMMA-RADIATION DETECTOR BASED ON HIGH-PURITY XENON

Sergey Sokolov*, Aleksey Pudov, Alexander Rybka, Vladimir Kutny, Alexander Abyzov, Gennadiy Kholomyeyev, Serhii Melnikov
National Scientific Center "Kharkiv Institute of Physics and Technology", Kharkiv, Ukraine
**Corresponding Author: sokolov@kipt.kharkov.ua*
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This paper reports on the development, manufacturing and testing of proportional γ-ray detectors filled with gas mixtures based on high-purity xenon. To let the gas mixtures into the detector, a special installation was developed and manufactured, consisting of components designed to work with high-purity gases. The influence of the gas pressure, its composition (pure Xe or its mixture with H_2, CH_4), and the voltage at the anode on the spectrometric resolution and gas gain of the detectors was studied. The addition of H_2 or CH_4 to xenon is used to increase the charge carrier drift velocity. These additives also stabilize the gas mixture, i. e. decrease the probability of gas breakdown at high voltage between the detector electrodes. Gas xenon, as well as its mixtures, of research purity grade (99.9999%) have been used. Proportional γ-ray detectors based on xenon gas can operate in both counting and spectrometric modes. To study the characteristics of the detectors, we used standard sources of γ-radiation ^{241}Am, ^{137}Cs, ^{152}Eu, ^{133}Ba. The best energy resolution values to date were obtained for a detector filled with a gas mixture of Xe + 2.1% CH_4 at a pressure of 2.5 bar and an anode voltage of 2500 V; they were ~ 9.5% for an energy of 40 keV and ~ 5% for 120 keV. In the manufacturing of detectors intended for radiation monitoring and identification of radioactive materials, in particular in nuclear power, complex materials science problems have been solved. To increase the thermal and radiation resistance, all elements of the detector construction are made of materials that are weakly activated by ionizing radiation. In addition, the design of the detectors is completely free of glass elements and organic components. The detectors are designed to monitor technological processes and to work as part of radiation monitoring systems, including those at nuclear power plants.
KEY WORDS: detector, gamma radiation, count rate, spectrometry, xenon, gas gain

Gas-filled counters are well known and used to register alpha-, beta-particles, protons, gamma quanta, neutrons in various fields of fundamental and applied research. Proportional gamma-ray counters-spectrometers filled with xenon gas have a number of advantages: a wide range of operating temperatures, high sensitivity, long-term stability, and high radiation resistance. The unique combination of physical properties of xenon gas makes it very attractive as an active medium in radiation detectors: high stopping power, low Fano factor, mechanical and chemical stability, as well as low energy required to create an electron-ion pair. Xenon detectors, in particular, have proven themselves well in astronomical applications in space [1], where they operate in highly aggressive radiation and temperature regimes.

The specificity of using detectors for radiation monitoring and identification of radioactive materials, in particular in nuclear power, requires the solution of rather complex materials science problems: the choice of materials with low activation in intense fluxes of ionizing radiation, including neutrons, the rejection of the use of glass elements, the complete absence of organic components in the design of the detector, the provision of very low (at the picoampere level) leakage currents between the detector electrodes.

This paper presents the first results of a study of the counting and spectrometric characteristics of a proportional gamma radiation detector of a special design filled with pure Xe.

DETECTOR DESIGN

Be, Al, Mg and Ti [2] are the structural elements that are more suitable, in terms of their activation properties, in the manufacture of detectors designed to operate in intense fields of ionizing radiation. Considering the properties of nuclei, it is undesirable to use alloys containing Mn, Co and Ag in the design of detectors. The prevalence of stainless steel in the manufacturing of the detectors is determined by its manufacturability, corrosion and temperature resistance, despite its activation in neutron fluxes. When iron and nickel, the main components of stainless steel, capture a neutron, they form radioactive nuclides with a half-life of more than three years.

For the manufacture of anodes, molybdenum or tungsten is typically used, and tungsten is used for anode tensioning springs. Molybdenum powder and copper solder are the main components used in sintering metal elements with ceramics. Currently, mainly kovar or titanium is used in metal-glass and metal-ceramic insulators.

Fig. 1 shows the schematic of the design of the developed and manufactured detector. The detector body is a seamless thin wall pipe made of 12X18H10T steel (Ø25.5×0.3 mm, length 250 mm). The design includes a coaxial vacuum current feedthrough with a "guard ring" and a high-voltage connector, as well as protective insulators designed to reduce edge effects. The cylindrical design was selected as the one most used, however, there have been studies of alternative designs, i.e. in [3,4], which are potentially interesting.

The disadvantages of metal-glass insulators, which are usually used in gas-discharge counters, are the limited abrupt temperature changes to 45°C and the limited neutron flux density up to 10^7 cm^{-2}s^{-1}. In this detector, for the

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manufacture of insulators, we used Macor ceramic glass manufactured by Corning Incorporated, USA. This new kind of glass-ceramic can be processed mechanically. The accuracy of machining is determined by the size of the mica crystals, the diameter of which is ~20 μm. The composition of the material is as follows: SiO_2 - 46%, MgO - 17%, Al_2O_3 - 16%, K_2O - 10%, B_2O_3 - 7%, F - 4%. Some mechanical, thermal and electrical properties of the ceramic are given in Table. 1.

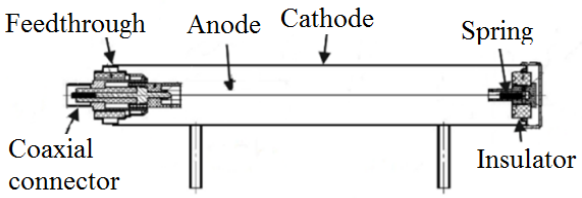

Figure 1. Detector design

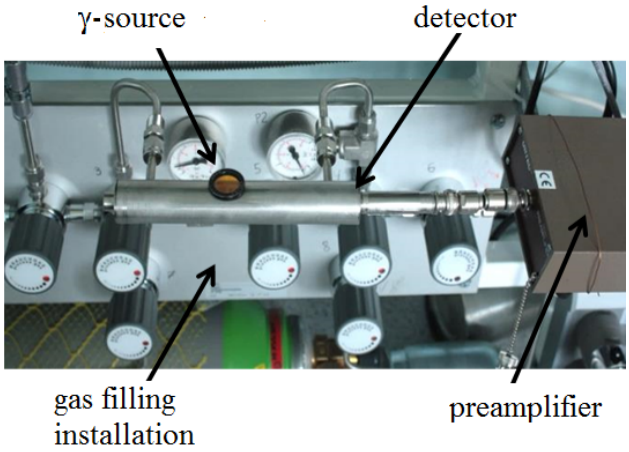

Figure 2. Installation for filling the detector with gas mixtures

Table 1.

Properties of glass-ceramics "MACOR"

Thermal expansion coefficient, 25 - 300ºC	$93 \cdot 10^{-7}$/ºC
Heat conductivity, 25ºC	1.46 Wt/m·ºC
Working temperature	800 ºC
Density	2.52 g/cm^3
Young's modulus, 25ºC	66.9 Gpa
Dielectric strength (for thickness 12 mm and 25°C)	62.4 kV/mm
Bulk resistance, 25°C	$>10^{16}$ Ohm·cm

The anode is a tungsten wire with a diameter of Ø60 μm, tensioned with a spring. Thus, the detector does not contain organic materials or glass.

To ensure the purity of the gas and prevent contaminants' release from the surface of the detector parts, various methods and cleaning technologies were used in the manufacture of all parts, and the detector as a whole. Sandblasting was carried out using aluminum oxide (Al_2O_3) particles with a size of 50 μm. We also used electrochemical polishing of the inner surface of steel pipes, electrochemical cleaning and polishing of thin tungsten wires, and final cleaning of the surface of all elements was carried out with dry ice CO_2. The detector was assembled by using laser welding. At various stages of cleaning and assembly, the control measurements of the resistance of the ceramic elements of the detector at high voltage were carried out.

To fill the detector with pure xenon and its mixtures, an installation was developed and manufactured, which used accessories from Spectron GCS GmbH (Fig. 2). Before filling, the detector, the valves and corresponding connecting tubes were evacuated and the detector was further degassed at an elevated temperature (~110ºC). The detector was filled with pure xenon or its mixture with H_2 (0.25%) or CH_4 (2.1%) to various pressures from 0.5 to 3 bar. These additives to xenon are used to increase the drift velocity of charge carriers [5] in gas, as well as to suppress secondary ionization processes. We used purified xenon gas, as well as purified xenon-hydrogen and xenon-methane gas mixtures of research grade purity (99.9999%). It was estimated that the gas in the detector had no more than 1 ppm contaminants. Table 2 shows the typical composition of impurities, in this case for the $Xe+H_2$(0.25%) gas mixture. The $Xe+CH_4$(2.1%) mixture has the same total amount of impurities and is assumed to have a similar impurity composition.

The counting and spectrometric characteristics of the detectors were investigated. A voltage bias from 100 to 2700 V was applied to the anode by using an Ortec 659 high-voltage power supply through an RC filter. The measurements were carried out by using an Ortec 142AH preamplifier, a 672 shaper amplifier, and a 927 ADC. The gain value was 100, the time constant of the shaping amplifier was 3 μs. The count rate and amplitude distributions of the detector signal were measured by using standard γ-sources ^{241}Am, ^{137}Cs, ^{152}Eu, ^{133}Ba.

Table 2.

Contaminants' composition in the gas mixture Xe+0.25%H_2.

% H_2	Contaminants, ppm. (not higher than)								
	Kr	O_2	N_2	Ar	C_nH_m	$CO+CO_2$	SF_6	S_2F_6	H_2O
0.25	0.1	0.1	0.2	-	0.1	0.1	0.1	0.1	0.1

RESULTS AND DISCUSSION

The dependence of the detector counting rate on the voltage measured for the γ-source ^{241}Am at different pressures of pure Xe or the xenon mixture Xe + 2.1% CH_4 is shown in Fig. 3. For all gas pressures in the detector, the counting rate of the detector begins to increase sharply when the voltage applied to the anode is above a certain threshold value. This threshold voltage shifts to higher values with increasing pressure in the detector. Thus, in practice, at higher pressure, a higher detector sensitivity is achieved, but higher voltage values are required.

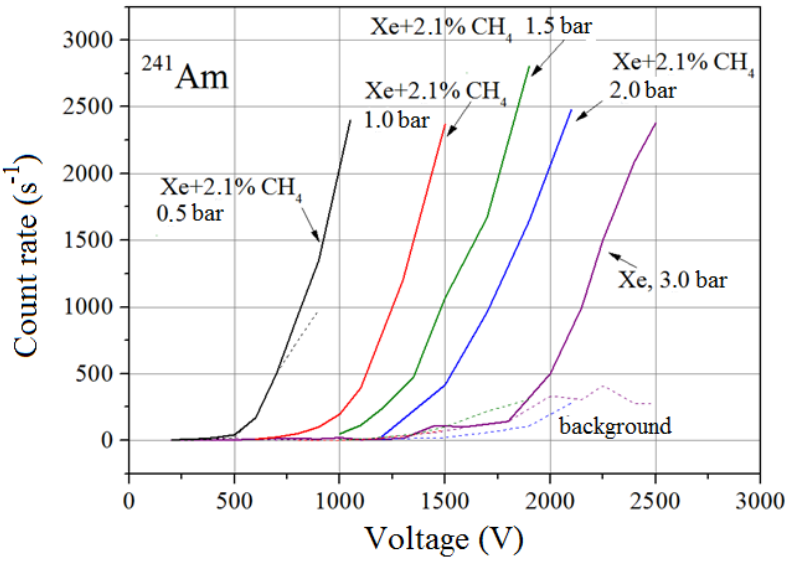

Figure 3. The count rate for ^{241}Am at different pressures of the gas mixture. Dashed lines represent the background measurements

The count rate measured as a function of the applied voltage for a detector filled with pure Xe up to a pressure of 1.5 bar is shown in Fig. 4. As can be seen, the number of counts in the background measurements is much lower than that in the measurements with the γ-source. Table 3 shows the signal-to-noise ratio values for the Xe+2.1%CH_4 mixture for the isotopes ^{241}Am, ^{133}Ba, ^{152}Eu at a pressure of 1.5 bar and anode voltage of 1900 and 2550 V.

Table 3.

Signal-to-noise ratio for the mixture Xe+2.1%CH_4 at 1.5 bar

Isotope	^{60}Am (59.5 keV)	^{133}Ba (80.9 keV)	^{152}Eu (121.8 keV)
Signal-to-noise ratio, 1900 V	8	11	16
Signal-to-noise ratio, 2550 V	25	34	50

With an increase in the pressure of the Xe+2.1%CH_4 mixture to 2.5 bar (Fig. 5), the look of the counting characteristics significantly differs depending on the type of the radiation source: the counting rate increases gradually with the anode voltage for ^{137}Cs, while for ^{241}Am it practically does not change at low voltage values, and above 1000 V begins to grow rapidly. This behavior corresponds to different energies of gamma rays emitted by these sources. Indeed, for ^{137}Cs, most of the detector signal is due to the broad spectrum of Compton scattered γ-quanta, since the quanta from the ^{137}Cs photopeak with an energy of 662 keV are not registered at such Xe pressures. On the other hand, ^{241}Am has several low-energy γ-lines (60 keV and below), and in the spectra of ^{241}Am, these lines (peaks) began to appear at voltages above 1000 V, therefore, the counting characteristic begins to increase significantly at these voltages.

Similar dependences were measured for the other mentioned gas mixtures and pressures, though it turned out that the addition of H_2 or CH_4 did not lead to significant changes in the counting characteristics of the detectors.

The examples of the measured amplitude distributions of the detector signal (spectra) are shown in Fig. 6. For comparison, the spectrum on the left shows a line obtained with an Ortec 419 precision pulse generator. As can be seen, the detector can register radiation with energies up to ~120 keV (^{152}Eu line). However, at a higher gas pressure in such detectors, it is possible to detect radiation with higher energies [6]. The energy resolution (from full width of a photopeak at the half-amplitude level, FWHM) of the detectors was determined from the photopeaks of different

γ-sources. For the detector filled with the Xe+2.1%CH_4 mixture at the pressure of 2.5 bar, and a voltage of 2500 V (Fig. 6), it ranged from ~9.5% for an energy of 40 keV to ~5% for 120 keV. Such values of the energy resolution of the detector are sufficient to identify a reasonable number of radioactive isotopes.

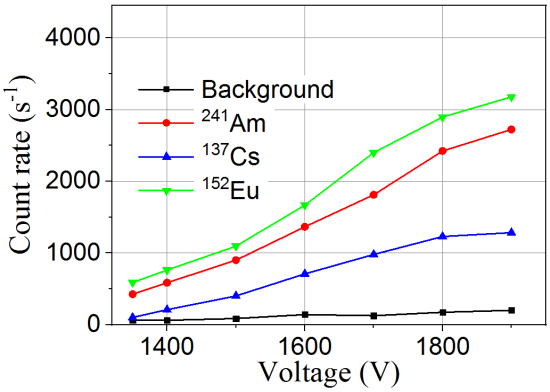

Figure 4. Voltage dependence of count rate for different isotopes, 1.5 bar, pure Xe

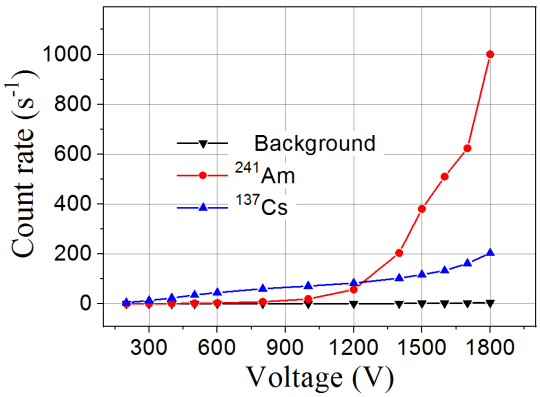

Figure 5. Voltage dependence of count rate for different isotopes, 2.5 bar, Xe + 2.1% CH_4

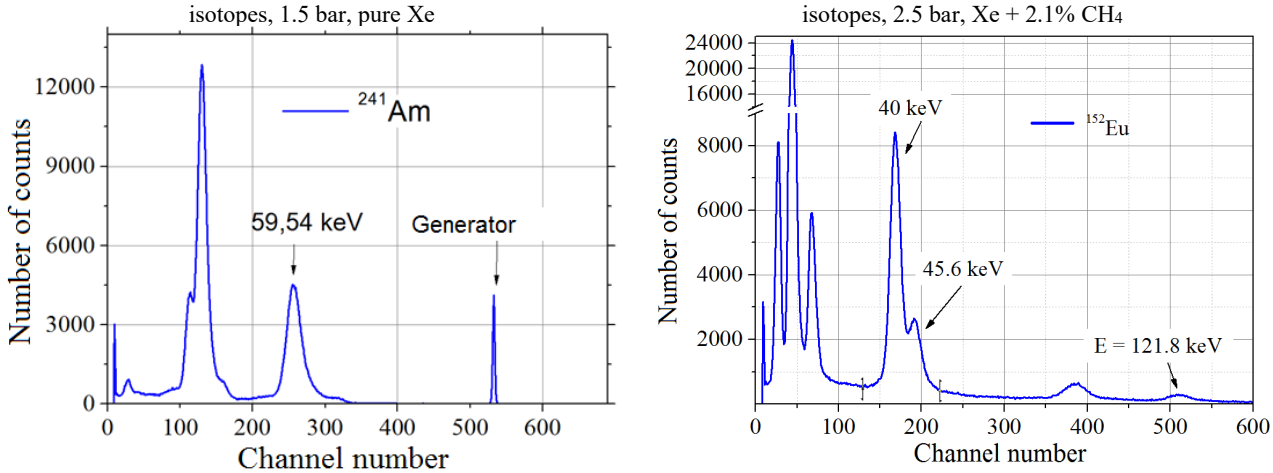

Figure 6. The measured spectra for ^{241}Am and ^{152}Eu, Xe+2.1% CH_4 mixture at 2.5 bar, and the anode voltage of 2500 V

The gas gain M is an important characteristic of a gas-filled detector operating in the proportional mode. For various values of pressure and composition of gas mixtures in the detector, it was determined by using the following formula [7]:

$$M = \frac{wVC_0}{Ae\epsilon_x}, \tag{1}$$

where w is the average energy required to create one ion-electron pair in a gas mixture (21.5 eV for Xe), V is the pulse amplitude, C_0 is the detector capacitance (in our case, 20 pF), A = 100 is the amplifier gain, e is the electron charge, ϵ_x is the photon energy (for example 60 keV for ^{241}Am).

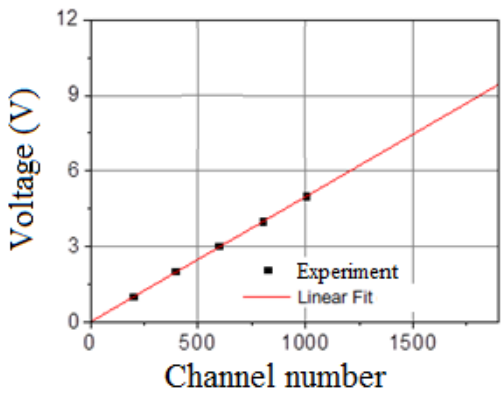

Figure 7. Dependence of the detector signal amplitude on the ADC channel number

Before determining the coefficient, the correspondence of the channel number of the measurement channel to the amplitude of the signal after the shaping amplifier was first carried out. The resulting dependence is linear (Fig. 7), with a linearity coefficient of 0.9999. Such a high degree of linearity indicates a high accuracy in determining the energy of spectral lines by the detector, which is necessary for the identification of radioactive isotopes.

The values of M depending on the applied voltage, calculated from the amplitude distributions for different values of pressure of the Xe+2.1%CH_4 mixture, are shown in Fig. 8. As can be seen, on a logarithmic scale, the dependence of the coefficient M on the applied voltage is nearly linear and shifts to the right with increasing gas pressure. Fig. 9 shows a comparison of the gas gain values for different gas compositions. As can be seen, the dependence of the gas gain on voltage is practically the same for the compositions of all mixtures, which were used.

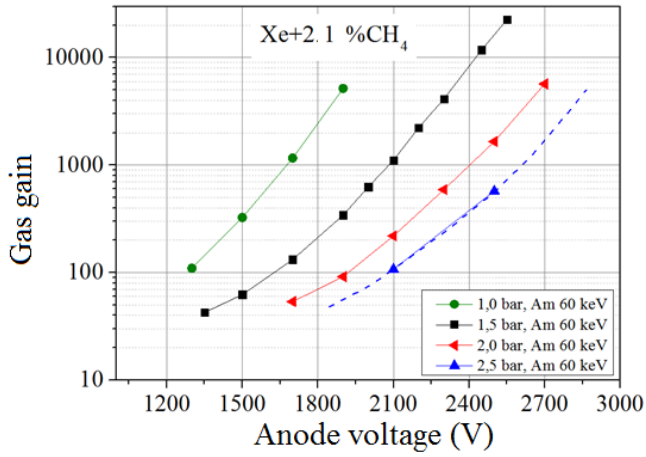

Figure 8. Gas gain versus anode voltage for Xe+2.1%CH_4

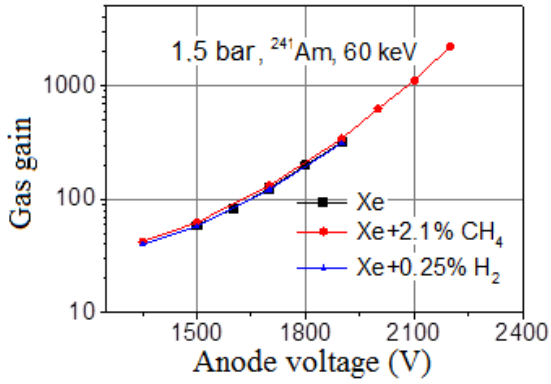

Figure 9. Gas gain as a function of the anode voltage at a pressure of 1.5 bar for clean Xe, Xe+2.1%CH_4 and Xe+0.25%H_2

Fig. 10 and Fig. 11 show the gas gain values calculated from the photopeaks of several different isotopes for pure Xe and a mixture of Xe + 2.1% CH_4, respectively. As can be seen in both cases, the *M* values calculated using different gamma lines practically coincide. This behavior suggests that the gas gain does not depend on the number of charge carriers created during the primary ionization event, which corresponds to the proportional operating mode of the detector. The dependence of *M* on voltage in a significantly wider range was obtained for the mixture of Xe + 2.1% CH_4, and it can be seen that for high voltages it is practically linear on a logarithmic scale (dashed line in Fig. 11). This behavior indicates that there is no effect of secondary ionization on the gas gain at the given applied voltages. The very fact that for the mixture of Xe + 2.1% CH_4, in contrast to pure Xe, it was possible to carry out measurements at a much higher anode voltage indicates the stabilization of the gas mixture upon the introduction of additives such as methane, i.e. lowering the probability of gas breakdown.

The results obtained are in good agreement with the values of the gas gain obtained for similar compositions of mixtures [8]. Note also that, in the case of using a thinner anode [9] in the detector, lower voltage to achieve the same detector performance would be required.

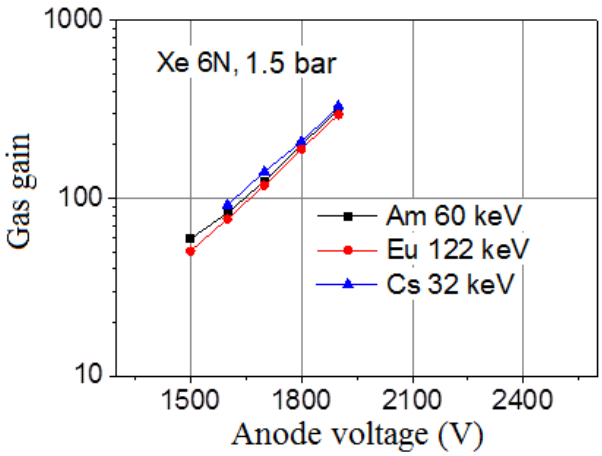

Figure 10. Gas gain for pure Xe at 1.5 bar, calculated for several gamma-ray energies

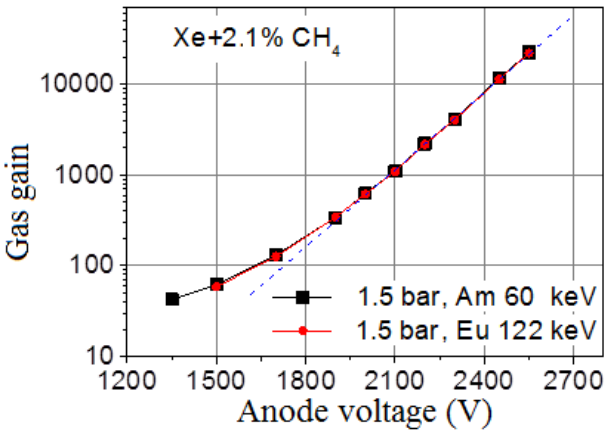

Figure 11. Gas gain for Xe + 2.1% CH_4 mixture at 1.5 bar, calculated for several gamma-ray energies

CONCLUSIONS

Thus, the γ-radiation detectors filled with xenon, which have both high counting (dosimetric) and spectrometric properties have been developed, manufactured and tested. The detectors were made without using glass or organic materials. The influence of gas pressure, its composition (pure Xe or its mixture with H_2, CH_4), and the anode voltage on the spectral resolution and gas gain of the detectors was investigated. The detectors have shown sufficient operational characteristics, which will allow them to be used in various fields of application, including at nuclear power plants.

To improve the characteristics of the detector, it is planned to use a thinner anode (Ø≤30 μm), as well as to use ^{10}B in the detector design, which will lead to the possibility of using it as a neutron detector. It may also be promising to investigate the effect of mixtures of noble gases such as Xe and Ne in attempts to reduce the anode voltage due to a process of energy transfer between the two species, which produces extra electrons [10].

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ORCID IDs

Sergey Sokolov, https://orcid.org/0000-0002-6134-905X; **Aleksey Pudov**, https://orcid.org/0000-0002-9499-379X
Alexander Rybka, https://orcid.org/0000-0003-4096-5539; **Vladimir Kutny**, https://orcid.org/0000-0001-8899-545X
Alexander Abyzov, https://orcid.org/0000-0002-1975-7586; **Gennadiy Kholomyeyev**, https://orcid.org/0000-0003-1779-9050
Serhii Melnikov, https://orcid.org/0000-0003-2494-1174

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ГАЗОНАПОВНЕНИЙ ДЕТЕКТОР ГАММА-ВИПРОМІНЮВАННЯ НА ОСНОВІ ВИСОКОЧИСТОГО КСЕНОНУ

С. Соколов, А. Пудов, А. Рибка, В. Кутній, А. Абизов, Г. Холомєєв, С. Мельніков
Національний науковий центр «Харківський фізико-технічний інститут»
Харків, Україна

Газонаповнені детектори іонізуючого випромінення знайшли широке застосування для вимірювання ядерних та радіоактивних матеріалів, а також в медицині, астрономії, матеріалознавстві. Пропорційні детектори γ-випромінення на основі газу ксенону можуть працювати як в рахунковому, так і в спектрометричному режимах. Для використання цих детекторів для радіаційного контролю та ідентифікації радіоактивних матеріалів, зокрема в атомній енергетиці, потрібно вирішити досить складні матеріалознавчі завдання. Це вибір матеріалів з низькою активацією в інтенсивних потоках іонізуючого випромінення, в тому числі нейтронів, відмова від використання скляних елементів, повна відсутність органічних компонентів в конструкції детектора. У даній роботі повідомляється про розробку, виготовлення та випробування пропорційних детекторів гамма-випромінювання, наповнених газовими сумішами на основі особливо чистого ксенону. Для напуску в детектор газових сумішей була розроблена і виготовлена спеціальна установка складається з компонентів, призначених для роботи з особливо чистими газами. Досліджено вплив тиску газу, його складу (чистий Xe або його суміші з H_2, CH_4) і напруги на аноді на спектрометричне розрізнення і коефіцієнт газового посилення детекторів. Добавка H_2 або CH_4 до ксенону застосовується для підвищення швидкості дрейфу носіїв заряду. Ці добавки також призводять до стабілізації газової суміші, тобто зниження ймовірності газового пробою при високій напрузі між електродами детектора. Використовувався газ ксенон, а також його суміші дослідницької марки чистоти (99,9999%). Для дослідження рахункових і спектрометричних характеристик детектора в роботі використовувалися стандартні джерела γ-випромінення ^{241}Am, ^{137}Cs, ^{152}Eu, ^{133}Ba. Кращі, на даний момент, значення енергетичного розрізнення були отримані для детектора, наповненого газовою сумішшю Xe + 2,1% CH_4 при тиску 2,5 бар і напрузі на аноді 2500 В; вони склали ~ 9,5% для енергії 40 кеВ і ~ 5% для 120 кеВ. Для підвищення термічної і радіаційної стійкості, всі елементи конструкції детекторів виготовлені з матеріалів, що слабо активуються під дією іонізуючого випромінення. Детектори призначені для контролю технологічних процесів і для роботи в складі систем радіаційного контролю, в тому числі на АЕС.

КЛЮЧОВІ СЛОВА: детектор, гамма-випромінення, швидкість відліку, спектрометрія, ксенон, газове підсилення

ГАЗОНАПОЛНЕННЫЙ ДЕТЕКТОР ГАММА-ИЗЛУЧЕНИЯ НА ОСНОВЕ ВЫСОКОЧИСТОГО КСЕНОНА

С. Соколов, А. Пудов, А. Рыбка, В. Кутний, А. Абызов, Г. Холомеев, С. Мельников
Национальный научный центр «Харьковский физико-технический институт»
Харьков, Украина

Газонаполненные детекторы ионизирующих излучений нашли широкое применение для измерения ядерных и радиоактивных материалов, а также в медицине, астрономии, материаловедении. Пропорциональные детекторы γ-излучения на основе газа ксенона могут работать как в счетном, так и в спектрометрическом режимах. Для применения этих

детекторов для радиационного контроля и идентификации радиоактивных материалов, в частности в атомной энергетике, требуется решить довольно сложные материаловедческие задачи. Это выбор материалов с низкой активацией в интенсивных потоках ионизирующих излучений, в том числе нейтронов, отказ от использования стеклянных элементов, полное отсутствие органических компонентов в конструкции детектора. В данной работе сообщается о разработке, изготовлении и испытании пропорциональных детекторов гамма-излучения, наполненных газовыми смесями на основе особо чистого ксенона. Для напуска в детектор газовых смесей была разработана и изготовлена специальная установка, состоящая из компонентов, предназначенных для работы с особо чистыми газами. Исследовано влияние давления газа, его состава (чистый Xe или его смеси с H_2, CH_4) и напряжения на аноде на спектрометрическое разрешение и коэффициент газового усиления детекторов. Добавка H_2 или CH_4 к ксенону применяется для повышения скорости дрейфа носителей заряда. Эти добавки также приводят к стабилизации газовой смеси, т.е. понижению вероятности газового пробоя при высоком напряжении между электродами детектора. Использовался газ ксенон, а также его смеси исследовательской марки чистоты (99,9999%). Для исследования счетных и спектрометрических характеристик детектора в работе использовались стандартные источники γ-излучения ^{241}Am, ^{137}Cs, ^{152}Eu, ^{133}Ba. Лучшие, на данный момент, значения энергетического разрешения были получены для детектора, наполненного газовой смесью Xe+2,1% CH_4 при давлении 2,5 бар и напряжении на аноде 2500 В; они составили ~ 9,5% для энергии 40 кэВ и ~ 5% для 120 кэВ. Для повышения термической и радиационной стойкости, все элементы конструкции детекторов изготовлены из материалов, слабо активируемых под действием ионизирующих излучений. Детекторы предназначены для контроля технологических процессов и для работы в составе систем радиационного контроля, в том числе на АЭС.

КЛЮЧЕВЫЕ СЛОВА: детектор, гамма-излучение, скорость счета, спектрометрия, ксенон, газовое усиление