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# GAMMA-RAY DETECTOR BASED ON HIGH PRESSURE XENON FOR RADIATION AND ENVIRONMENTAL SAFETY

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Gamma-spectrometers based on compressed xenon gas assigned for monitoring the reactors and the radiation background at nuclear power plants, non-proliferation of radioactive materials, supervision and control over the radiation background in the environmentally disadvantaged areas, and other applications, are very promising detectors with excellent performance characteristics. This article reports on the results of the first stage of work on the creation of the portable γ-spectrometer based on compressed xenon that is unique for Ukraine. In order to work with ultra-pure gases under pressure, the complex cryogenic installation for Xe purification and detector filling was designed and manufactured. The installation was made of specially cleaned components, equipped with a heating system for the degassing of the inner walls, and is able of maintaining high vacuum down to $2 \cdot 10^{-9}$ mbar. A prototype ionization chamber for the use in portable HPXe detectors was developed and made. For the detector testing, a spectrometric channel based on high-quality electronic components was designed and manufactured. In the initial experiments, a study of the properties of the purified Xe mixed with the dopant $H_2$ was carried out. The assessment of the lifetime of charge carriers τ in the working gas at a pressure of 30 bar gave the value of $\tau > 150$ μs.

## INTRODUCTION

One of the problems of the modern radiation and environmental safety, in Ukraine and in the world as a whole, is the creation of accurate and reliable devices of radiation dosimetry and spectroscopy, the spectroscopy of γ-rays in particular. Gamma-spectrometers are necessary when performing tasks in applications such as the customs control (non-proliferation of radioactive materials), the monitoring and control of radiation background in environmentally disadvantaged areas, the monitoring of the reactors and the radiation background at nuclear power plants, the disposing of spent radioactive materials. Currently, the γ-spectrometers based on high-purity germanium (HPGe) and scintillators, such as NaI(Tl) and others, are widely used. Both types have advantages and some disadvantages. HPGe detectors that have the best spectral resolution of ~ 0.1% have to be cryogenically cooled, making their manufacture and operation quite expensive. On the other hand, the spectral resolution of the scintillator detectors is not very high (7…8%). Spectrometric detectors of γ-radiation based on high-pressure xenon gas (HPXe) have a high energy resolution (~ 1.7% for the 662 keV γ-quanta), high sensitivity, long operation life, and ability to work in a wide temperature range (from +20 to +180 °C), which makes them very promising.

The studies on the creation of a series of modern γ-spectrometers based on HPXe with a number of different potential applications have been carried out at KIPT since 2013. The creation of the HPXe detectors is associated with a number of serious technical demands. In particular, for the optimum spectrometric properties of the detectors, Xe must be highly purified (the electronegative impurities content should be $< 10^{-4}$ ppm); the operating gas pressure is up to ~ 50 bar; the operating voltage on the electrodes may reach 30 kV. This article reports on the results of the first stages of the work on the creation of a portable HPXe γ-spectrometer at KIPT.

## Xe GAS AS THE WORKING MEDIUM FOR THE IONIZATION CHAMBER

One of the most important characteristics of any substance used for detection of γ-radiation is its ability to absorb γ-quanta. This ability is characterized by the probability of interaction of γ-quanta with the substance. As known, γ-rays, as they pass through a substance, undergo mainly three types of interaction (excluding the nuclear reactions induced by the γ-quanta): the photoelectric effect, the Compton effect, and the formation of electron-positron pairs. The cross-sections of these interactions depend on the charge of the working material Z and the γ-quantum energy $E_\gamma$ as follows [1, 2]:

$$\sigma_{\text{ph}} \propto \frac{Z^5}{E_\gamma^{7/2}},\ \sigma_{\text{comp}} \propto \frac{Z}{E_\gamma},\ \sigma_{\text{pair}} \propto Z^2 \ln\left(2E_\gamma\right). \quad (1)$$

From the standpoint of the value of the atomic number, xenon (Z = 54), compared to other substances (for Ge Z = 32 and the NaI(Tl) Z = 50), is more preferred for the registration of γ-rays, especially in the energy region where the photoelectric effect is the main mechanism of the interaction of γ-quanta with matter.

Another important characteristic of the working substance of a γ-detector, which determines the efficiency of detection of γ-rays, is its density. The high compressibility of the gaseous xenon allows achieving a sufficiently high density (1.5…1.8 g/cm$^3$) at relatively low pressures (60…80 bar). The specific dependences of Xe density on pressure at various temperatures are shown in Fig. 1 [3].

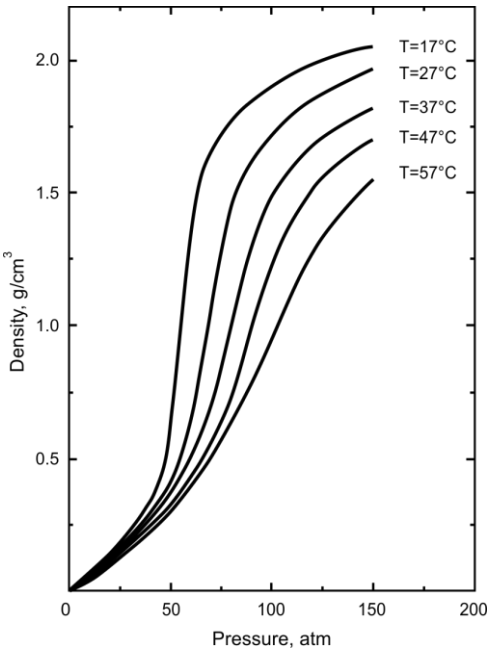


*Fig. 1. The dependence of the xenon gas density on the pressure at different temperatures [3]*

As shown by detailed studies of the spectrometric characteristics of HPXe γ-detectors, at the xenon density larger than 0.6 g/cm$^3$ the energy resolution of the detector begins to deteriorate. For this reason, the density of xenon in γ-spectrometers with high energy resolution should be about 0.5…0.6 g/cm$^3$. The mentioned density is several times lower than the density of liquid xenon (3.06 g/cm$^3$) or crystal NaI(Tl) (3.7 g/cm$^3$), and Ge (5.3 g/cm$^3$). In this sense, xenon is inferior to them in the detection efficiency of the gamma rays, as it is proportional to the density of the working substance.

Nevertheless, it is relatively easy to increase the detection efficiency of γ-rays of the HPXe γ-detectors without the deterioration of their energy resolution by increasing the working volume of the detector.

The average ionization energy in gaseous HPXe is relatively low, < 21 eV per electron-ion pair [4]. In addition, HPXe has low Fano factor – 0.13±0.1 [5]. From these two parameters, it follows that the theoretical energy resolution for the HPXe ionization chambers is ~ 0.5% at 662 keV.

There are other attractive qualities to the HPXe. For example, the ionization chambers based on HPXe are ideal for use in uncontrolled environments. In particular, it was shown that the detector characteristics do not change over a wide temperature range (from +20 to +180 °C) [6].

Some important properties of the pressurized xenon gas, compared with other detecting media, are listed in Tabl. 1 [1].

Table 1

Comparison of the characteristics of Xe and other detection media

| Medium | Density, g/cm$^3$ | Atomic number | w (eV/e-i pair) |
|---|---|---|---|
| NaI:Tl | 3.671 | 11. 53 | ~100 |
| Ge | 5.333 | 32 | 2.98 |
| $Cd_{0.8}Zn_{0.2}Te$ | 6 | 48. 30. 52 | 5 |
| $HgI_2$ | 6.43 | 80. 53 | 4.33 |
| HPXe | 0.5 | 54 | 21.9 |

It is also important to note that, in order to increase the electron drift velocity in the gaseous xenon as the working medium for γ-spectrometers, various accelerator molecular gases are commonly used. The optimum dopant is considered to be hydrogen [7, 8, 9, 10], the addition of which at the level of 0.2…1.0% can increase the carrier drift velocity by 5…10 times. Thus, in the initial stages of our work, which will be discussed, according to the given literary sources, the content of hydrogen in xenon of Xe+$H_2$ (0.25%) was selected.

## THE INSTALLATION FOR XENON PURIFICATION AND FILLING

The creation of the complex installation for xenon purification and filling is one of the most important stages of the work on the creation of the γ-radiation detectors based on HPXe. The main concept of the complex installation for xenon purification and filling has been developed after a careful study of the works by Dmitrenko et al. [11], Bolotnikov et al. [12], and others. Both the ideas gathered from the literature, as well as our own developments, were included in the final installation design.

Fig. 2 shows a schematic of the complex cryogenic installation for xenon purification and filling. The complexity of the installation is related to the necessity of obtaining a high vacuum level, in order to initially purify Xe and then work with pure Xe, as well as the need to fill the detector chamber with Xe under high pressure. Based on these requirements, the installation consists of a subsystem for creating a high vacuum level and the subsystem for the operation with high pressure. The vacuum part consists of the turbo-molecular pump Pfeiffer HiCube 80 Eco, the ion pump Gamma Vacuum TiTan CV, the vacuum line, a vacuum sensor Pfeiffer PBR 260 and the all-metal vacuum valves.

The vacuum part is connected to the high-pressure part, which consists of all-metal high-pressure valves, pressure gauges, the pipeline and the connected cylinders and vessels, including the ionization detector chamber itself. To prevent excessive pressure in the installation, there were safety bursting discs of the type Swagelok SS-RDK-16-1900 installed with a burst pressure of 131 bar. All of the purchased components that were used had a high degree of purification of the inner surfaces.

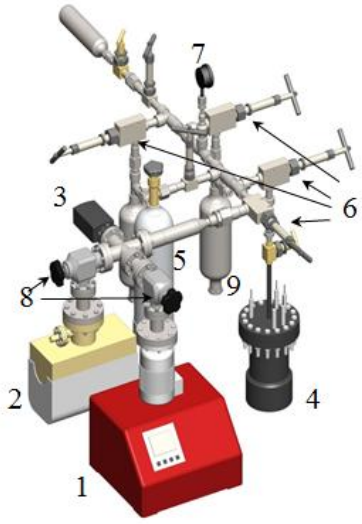


*Fig. 2. The schematic of the complex cryogenic installation for Xe purification and filling: 1 – turbo-molecular pump; 2 – ion pump; 3 – vacuum sensor; 4 – ionization chamber; 5 – cylinder with Xe; 6 – all-metal high-pressure valves; 7 – pressure gauge; 8 – vacuum valves; 9 – auxiliary cylinder for Xe transfer*

For the cryogenic purification of xenon and its mixtures [2], as well as for the transfer of gases in the installation, a thermoregulated nitrogen cryostat was designed and manufactured. The use of the precise proportional-integral-differential (PID) controller allows the cryostat to cool the installation vessels during the cryogenic purification and transfer in the temperature range of 173…300 K with an accuracy of ± 1K.

After the assembly and a prolonged evacuation of the installation, the vacuum level of the order of $2 \cdot 10^{-8}$ mbar was achieved (when evacuating all of the installation volumes). In order to prepare the installation further for the procedures of the detector filling with the purified gas, the installation must be heated to achieve the "degassing" of internal surfaces.

For this reason, the heating elements were installed onto the complex installation for xenon purification and filling that were controlled by the developed control cabinet. With the help of this heating system, long cycles of heating of the pipes, the vessels and the vacuum components can be carried out at temperatures up to 450 °C. The sensing of the heating temperatures was carried out by using the cable K-type (chromel-alumel) thermocouples.

Fig. 3 shows a graph of the pressure inside the installation measured at room temperature after multiple cycles of heating of different installation sections. The maximum temperature for heating of each component was selected according to the manufacturer's acceptable operating temperature data. As a result of the prolonged heating of all installation components, the improved vacuum level of the order of $P \sim 2 \cdot 10^{-9}$ mbar was achieved.

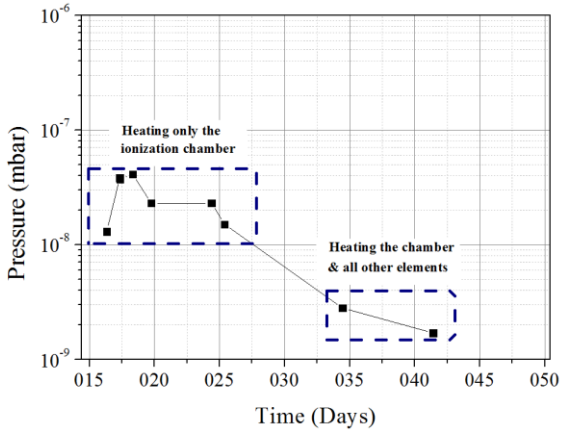


*Fig. 3. Pressure inside the Xe purification installation during the heating procedures (degassing)*

## THE IONIZATION CHAMBER BASED ON HIGH PRESSURE Xe

Out of all previously studied designs of the HPXe detector ionization chambers, the highest energy resolution (1.7%) was achieved in cylindrical chambers with three electrodes [11]. In the first version (the prototype) of the ionization chamber made in KIPT, such a configuration has been adopted as the basis (Fig. 4).

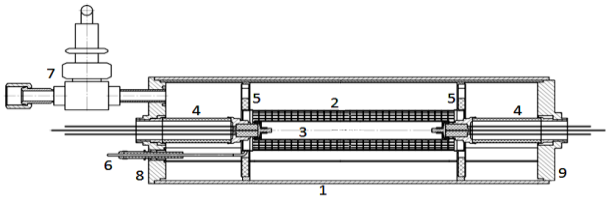


*Fig. 4. The drawing of the cylindrical ionization chamber of the HPXe γ-spectrometer: 1 – cathode-body; 2 – Frisch grid; 3 – anode; 4 – anode high-voltage feed throughs; 5 – ceramic supports of the Frisch grid; 6 – high-voltage feed through for the Frisch grid; 7 – filling valve; 8, 9 – side flanges*

The main elements of the design are: the steel body, that is also the high-voltage cathode; the intermediate electrode (the Frisch grid) that is transparent to carrier by ~ 96%; the central tubular anode. The side flanges, the cylindrical body, the Frisch grid and the guides supporting it are made of the A316 stainless steel; the central anode is made of a titanium tube, 20 mm in diameter and 180 mm in length. The design uses high-voltage feed throughs from the Ceram Tec Corp., which also support the anode. The Frisch grid made by the Constellation Technology Corp., is supported by high-voltage ceramic rings.

When operating the chamber in the detector mode, the electric field with the strength of $\geq 2$ kV/cm is created between the cathode and the grid and between the grid and the anode. To ensure such field, the voltage of -20 and -10.5 kV is applied to the detector cathode and the Frisch grid, respectively.

The main function of the intermediate electrode (the Frisch grid) is to eliminate the influence of the coordinates of the ionization event on the amplitude of the detected pulses [1].

The main design and calculated parameters of the ionization chamber are shown in Tabl. 2.

Table 2

Parameters of the developed and manufactured ionization chamber

| Parameter | Value and units |
|---|---|
| Working anode length | 180 mm |
| Anode outer diameter | 20 mm |
| Frisch grid length | 180 mm |
| Frisch grid outer diameter | 40 mm |
| Cathode inner diameter | 100 mm |
| Cathode wall thickness | 3 mm |
| Working gas volume | ≈1.41 l |
| Calculated cathode voltage | –20 kV |
| Calculated grid voltage | –10.5 kV |
| Calculated electric field near cathode | ≈2000 V/cm |
| Calculated electric field near anode | ≈15000 V/cm |
| Frisch grid penetration area | 96% |

## THE GAS PURIFICATION AND PURITY ASSESSMENT

For filling of the ionization chamber, the high purity xenon gas doped with hydrogen was used. Tabl. 3 shows the results of the elemental analysis of the mixture according to the gas chromatography measurements made by the company “Ingaz”, Mariupol.

Table 3

The content of the working mixture Xe+$H_2$

| Molecule | Quantity | Units |
|---|---|---|
| Xe | 99.7499 | % |
| $H_2$ | 0.25 | % |
| $N_2$ | 0.2 | ppm |
| $O_2$ | 0.1 | ppm |
| Kr | 0.1 | ppm |
| $C_nH_n$ | 0.1 | ppm |
| $CH_4$ | 0 | ppm |
| $CO_2$ | 0.1 | ppm |
| CO | 0 | ppm |
| THC | 0 | ppm |
| $SF_6$ | 0.1 | ppm |
| $C_2F_6$ | 0.1 | ppm |
| $H_2O$ | 0.1 | ppm |
| Total impurities | 0.9 | ppm |

By using the complex installation for gas purification and filing, the filling of the mixture into the detector ionization chamber was carried out, in this case to a pressure of 30 bar, corresponding to the gas density ~ 0.15 g/cm$^3$.

The method of assessing the lifetime of electrons in a working medium is based on the analysis of individual pulses measured in an ionization chamber at sufficiently small values of the electric field strength. Fig. 5 is the diagram of the connection of the electronic components in our pulse measurements. A two-electrode configuration was used for the radiation registration, i. e., the intermediate electrode and the anode of the chamber. The potential difference between the electrodes was 50 V, and the magnitude of the resulting electric field strength between the electrodes varied from 36 to 72 V/cm. For the lower values of the potential difference, the pulse registration was no longer observed. The signal was amplified with a charge-sensitive amplifier and digitized using a Tektronix MDO 3034 oscilloscope. For the ionizing radiation generating the pulses, we used cosmic muons [13], registering in the chamber with a rate of approximately once every 30 s.

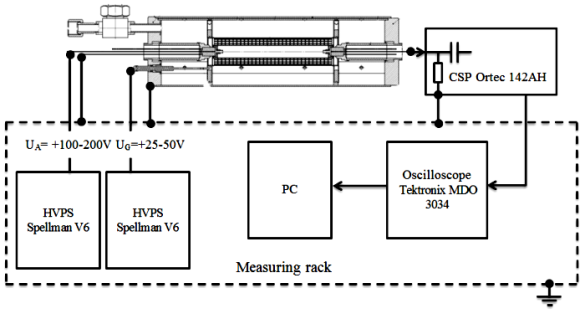


*Fig. 5. The ionization chamber wiring diagram for the electron lifetime measurements*

Fig. 6 shows one of the longest measured pulses. As one can see, the full pulse time is ~ 500 μs, and the rise time is ~ 150 μs.

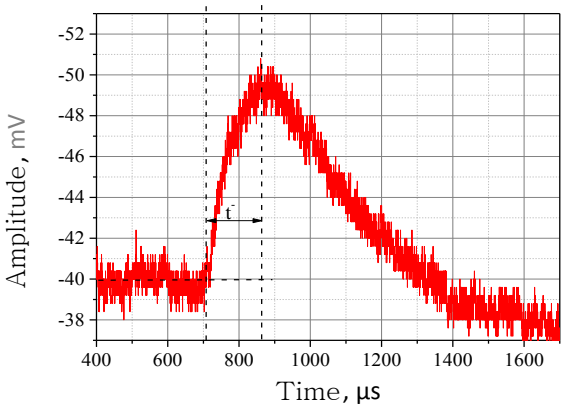


*Fig. 6. A measured pulse from a single cosmic muon*

With properly selected values of the electric field strength in the chamber, and for the values of the differentiating *RC* time constant of the amplifier lying within the range of $t^- << RC << t^+$ (where $t^-$ is the drift time of the electrons created by the ionization, and $t^+$ is the ions drift time), the pulse rise time is approximately equal to the electron drift time [1].

The relationship between the total charge Q generated in the working gas, the collected charge ΔQ, the electron drift time *t*, and the electron lifetime τ can be written as (2). From these equations, a conclusion can be drawn about the connection between the charge drift time (pulse rise time) and the lifetime: the lifetime is *at least* equal to the carrier drift time.

$$\frac{\Delta Q}{Q} = 1 - \exp\left(-\frac{t}{\tau}\right) \approx \frac{t}{\tau}, \quad \tau = t\frac{Q}{\Delta Q}, \quad \tau > t. \qquad (2)$$

Based on the analysis of multiple pulses measured by us, the longest rise time (the electron drift time) was determined, which was ~ 150 μs. Consequently, according to the described method, this value is the

lower limit of the electron lifetime, that is $\tau > 150$ μs in the $Xe+H_2$ (0.25%) mixture with the density of 0.15 g/cm$^3$.

## CONCLUSIONS

The γ-ray spectrometer based on high-pressure xenon gas was designed and built in KIPT. A complex cryogenic installation for the purification and filling of xenon, and mixtures thereof, has been designed and built, which is meant for the purification and transfer into the detector of high-purity Xe at pressure up to 60 bar. The installation is equipped with a heating system for degassing of internal surfaces, has the functionality of the cryogenic gas purification, as well as the ability to measure the degree of purity of the gas by measuring the lifetime of the charge carriers.

The experiments on filling the created detector ionization chamber with the purified mixture $Xe+H_2$ (0.25%), and the measurements of the lifetime of carriers in the working gas by using the single pulse analysis have been conducted. It was determined that the lower limit of the electron lifetime in the medium at a density of 0.15 g/cm$^3$ is $\tau > 150$ μs.

The studies of the spectrometric characteristics of the created detector, the studies of the effect of the gas pressure, purity and density on the detector characteristics, as well as the possible creation of the additional modules for the ultra-high purification of xenon and its mixtures, are planned.

**Acknowledgement**

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## ДЕТЕКТОР ГАММА-ИЗЛУЧЕНИЯ НА ОСНОВЕ КСЕНОНА ВЫСОКОГО ДАВЛЕНИЯ ДЛЯ ЗАДАЧ РАДИАЦИОННОЙ И ЭКОЛОГИЧЕСКОЙ БЕЗОПАСНОСТИ

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Гамма-спектрометры на основе сжатого газа ксенона, предназначенные для наблюдения и контроля за реакторами и окружающим фоном на АЭС, нераспространением радиоактивных материалов, радиационным фоном в экологически неблагоприятных зонах и других применений, являются очень перспективными детекторами с отличными эксплуатационными характеристиками. Данная статья посвящена результатам первой стадии работы по созданию уникального для Украины переносного γ-спектрометра на основе сжатого ксенона. Для работы с чистыми газами под давлением разработана и изготовлена комплексная криогенная установка очистки и напуска Xe. Установка создана на основе особо чистых комплектующих, оснащена системой прогрева для дегазации внутренних стенок и способна поддерживать высокий вакуум до $2 \cdot 10^{-9}$ мбар. Разработан и изготовлен прототип ионизационной камеры для использования в переносных

HPXe-детекторах. Для работы детекторов разработан и изготовлен спектрометрический тракт на основе высококачественных электронных блоков. В качестве первых испытаний проведены исследования свойств смеси очищенного Xe с легирующей добавкой $H_2$. При оценке времени жизни носителей заряда $\tau$ в рабочем газе под давлением 30 бар получены значения $\tau > 150$ мкс.

## ДЕТЕКТОР ГАММА-ВИПРОМІНЮВАННЯ НА ОСНОВІ КСЕНОНУ ВИСОКОГО ТИСКУ ДЛЯ ЗАВДАНЬ РАДІАЦІЙНОЇ ТА ЕКОЛОГІЧНОЇ БЕЗПЕКИ

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Гамма-спектрометри на основі стисненого газу ксенону, що призначені для спостереження і контролю за реакторами і навколишнім фоном на АЕС, нерозповсюдженням радіоактивних матеріалів, радіаційним фоном в екологічно несприятливих зонах і інших застосувань, є дуже перспективними детекторами з відмінними експлуатаційними характеристиками. Дана стаття присвячена результатам першої стадії роботи по створенню унікального для України переносного $\gamma$-спектрометра на основі стисненого ксенону. Для роботи з чистими газами під тиском розроблена і виготовлена комплексна кріогенна установка очищення і напуску Xe. Установка створена на основі особливо чистих комплектуючих, оснащена системою прогріву для дегазації внутрішніх стінок і здатна підтримувати високий вакуум до $2 \cdot 10^{-9}$ мбар. Розроблено та виготовлено прототип іонізаційний камери для використання в переносних HPXe-детекторах. Для роботи детекторів розроблений і виготовлений спектрометричний тракт на основі високоякісних електронних блоків. В якості перших випробувань проведені дослідження властивостей суміші очищеного Xe з легуючою домішкою $H_2$. При оцінці часу життя носіїв заряду $\tau$ в робочому газі під тиском 30 бар отримані значення $\tau > 150$ мкс.