HIGH-PRESSURE XENON DETECTORS FOR APPLICATIONS IN PORTAL SAFEGUARD SYSTEMS AND FOR MONITORING NUCLEAR WASTE

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ABSTRACT

Constellation Technology Corporation has developed a family of high-pressure xenon ionization chambers (HPXe) with mass of working medium between 100 g and 2 kg. The detectors demonstrate a good energy resolution (below 3%FWHM at 662 keV for 1 kg detector) that is close to the resolution of room temperature semiconductor detectors. The detectors employ relatively simple design, robust construction, and are available in a variety of pipe-like configurations of up to 1 m long. The sensitive area of HPXe may cover a few square feet and detect gamma rays with efficiency close to solid state detectors. Both in experimental modeling and computer simulations, HPXe has shown a good potential for detecting kilogram amounts of nuclear materials passing through the vehicle portal of a facility designated to store special nuclear materials. HPXe detectors have been tested at hard radiation conditions typical for a space orbital station. There is a positive experience of using HPXe at 120^{o}C with a specially designed preamplifier. There is no temperature and radiation degradation, as well as no charge trapping in xenon itself. Radiation stability of the working medium (Xe) is practically unlimited. All these features make HPXe detectors ideal candidates for monitoring nuclear waste tanks and other nuclear installations with hard radiation conditions.

INTRODUCTION

There is an urgent need for reliable and sensitive nuclear radiation systems suitable for detecting the accidental or intentional transport of special nuclear materials throughout portals at places such as border crossings, exits from nuclear storage facilities, gates of nuclear plants etc. (see, for example [1]). The detectors for these systems must be insensitive to environmental temperature extremes, rugged, and affordable. They must possess good efficiency and energy resolution but not require frequent servicing and maintenance, like using systematic cryogen supplement. Practically all of the nuclear materials (NM) emit gamma rays that constitute a characteristic signature for that material, penetrate construction materials, and can be used for detection and identification of NM. So far, two general categories of gamma ray spectrometers are used for these purposes:

- Semiconductor detectors (Germanium is the best of them), which have excellent energy resolution (~0.1% at 1 MeV)
- Scintillation detectors (NaI(Tl) is the most popular scintillator), which have the advantages of ambient-temperature operation, relatively low cost for unit of mass and are available in large massive crystals for construction of high-effective detection systems.

However, semiconductor detectors are very expensive (cost of ~1 cm^3 room-temperature detectors such as CZT could be up to $1,500/g) and sensitive to hostile environments; scintillation detectors suffer the disadvantage of poor energy resolution.

Since 1943, 176 underground storage tanks were constructed and filled with highly radioactive chemical waste [2]. The tanks typically are 23 m (75 ft) in diameter and have a capacity of up to

3,790 m^3 (1,000,000 gallons). The wastes are maintained at highly alkaline conditions at temperature up to 65°C. Fission and activation products generate gamma-ray dose rates up to approximately 11 Gy/h (1100 R/h) in the tank interior. *In situ* gamma-spectroscopy is used to measure gamma-ray-emitting radionuclide content and distribution within the tanks. This information is used to aid in tank characterization and is being developed for monitoring the effectiveness of waste retrieval. Currently, CdTe and CdZnTe detectors are used for gamma-spectrometry inside the waste tanks. However, CdTe and CdZnTe detectors have considerable charge trapping within the detector volume, which results in spectral degradation at high gamma-ray energy and high dose rates. The high temperature environment degrades spectrometric characteristics of these semiconductor detectors, which is why they are cooled by circulating ice water or are used for a very limited time.

The recent developments in xenon purification methods, material science, and sensitive electronics have generated significant gains in high-pressure ionization chamber technology. Today, high-pressure xenon is emerging as an important alternative detection medium for high resolution, room temperature gamma radiation spectrometry. Xenon effectively absorbs gamma rays, is mechanically and chemically stable, and can be used in extremely harmful conditions, such as intensive radiation fields, mechanical vibrations and shocks, high temperatures and pressures. There are no principal limitations on the dimensions of Xe detectors, which may be built in a variety of configurations such as, cylindrical or tunnel-like with dimensions large enough to pass loaded trucks through the detectors. Xenon is a relatively low cost material (market price for high-purity xenon is about $1/g).

The physical properties of xenon allow developing gamma-ray spectrometers with energy resolution close to room temperature semiconductor detectors, such as CdTe, CZT, and HgI_2. The statistic-limited (or the best theoretically achievable) energy resolution of any ionization detector is given by:

$$DE_i = 2.35\ [FWE\ ]^{1/2},$$

where E is the measured energy deposition, F is the Fano factor, and W is the average of electron-ion pair production [1]. One can see that the best achievable energy resolution is proportional to $(FW)^{1/2}$. For xenon of moderate density (<0.5 g/cm^3 for xenon), this factor has a relatively low value, comparable to that of the best semiconductors (Table 1). The theoretical limit for the energy resolution of xenon-filled detectors may be estimated to be 0.3-0.4% FWHM at 662 keV.

	F	***W*, eV**	**$(FW)^{1/2}$, $eV^{1/2}$**	***DE***
Si (77K)	0.08-0.13	3.8	0.5-0.7	0.38
Ge (77K)	0.06-0.13	3.0	0.4-0.6	0.31
CdZnTe (-40°C)	0.14	5.0	0.7	0.44
HgI_2	0.30	4.2	1.1	0.71
Ne + 0.5%Ar	0.050	26.2(Ar)	1.1	0.72
Xe	0.13	21.5	1.7	1.1
NaI(Tl)	1	26	5.1	3.2

Table 1. Physical Parameters of Noble and Gaseous and Solid State Detectors

HIGH-PRESSURE XENON DETECTORS

Constellation Technology Corporation has developed a family of high-pressure xenon ionization chambers (HPXe) with a mass of working medium between 100 g and 2 kg (Table 2). The detectors demonstrate a good energy resolution (below 3%FWHM at 662 keV for 1 kg detector – Fig.1) that is close to resolution of room temperature semiconductor detectors with only a few grams working mass. In energy resolution, they fulfil the niche between Germanium and scintillation detectors. The detection efficiency of HPXe is close to that of solid state detectors (Fig.2). The detectors employ relatively simple design, robust construction, and are available in a variety of pipe-like configurations of up to 1 m long. The sensitive area of HPXe may cover a few square feet and detect gamma rays with efficiency close to solid state detectors. Constellation HPXe spectrometers are constructed in portable and mobile systems available for testing in conditions of nuclear storage facilities or plants.

Detector	*HPXe-100*	*HPXe-500*	*HPXe-600*	*HPXe-1000*
Electrode system	2 electrodes	3 electrodes	3 electrodes	3 electrodes
Configuration of detector module: IC – ionization chamber; PA - preamplifier, HV– high-voltage supply	IC + PA	IC + PA + HV	IC	IC + PA
Mass of detecting medium, kg	0.1	0.8	0.5	1.8
Size of detector module, cm	∅4 x 13	∅12 x 50	∅9 x 62	∅12 x 100
Wall thickness, mm st.st.	1.5	2.7	2.0	2.7
Energy range, MeV	0.1-1.5	0.1-5.0	0.1-3.0	0.1-5.0
Energy resolution , %FWHM	4.5 (511 keV)	2.7 * (662keV)	5.0 (662 keV)	4.0 (662 keV)
Longevity, years	>6	>6	>6	>6
Energy resolution drift, % year^{-1}	<0.1	<0.1		<0.1
Temperature range	10-60^{o}C**	10-60^{o}C**	10-200^{o}C	10-60^{o}C**
Threshold of acoustic sensitivity		>60 dB at 662keV		
Required voltage	-4 kV	110 V AC/ battery	-12 kV, -9 kV	110V AC
Consumption power	<1 mW		<1 mW	0.5 kW

* Measured with pulse-gated HV power supply **Limited by charge-sensitive preamplifier

Table 2. Specifications of Constellation High-Pressure Xenon Detectors

HPXe-100 is a compact, simple two-electrode cylindrical ionization chamber exclusively constructed from stainless steel and ceramics. Being equipped with a high-temperature preamplifier, the detector could be used in very harmful conditions, for example, for monitoring nuclear waste tanks. Other possible applications include monitoring nuclear reactors and oil-well logging.

The detector HPXe-500 is constructed as a portable spectrometry system consisting of the detector module and readout system based on a laptop computer. The detector module HPXe-500 includes gridded cylindrical ionization chamber, preamplifier, and HV supply fastened directly to a flange of the high-pressure vessel. There are a charge-sensitive amplifier, high voltage supply, and high-voltage divider supplying voltages to both grid and cathode of the detector inside this unit. A HV supply working in “flipping mode,” i.e. generates high-voltage ~10 ms pulse when the readout system is disabled with veto-signal. This allows dramatically reduce electronic noise and improves energy resolution of the system (Fig.1). A compact readout system includes a laptop PC and PC/MCA card attached to the computer.

The detector HPXe-1000 is constructed as a complete mobile spectrometer system. The detectors is mounted inside a vibro-insolating housing attached to the mobile cabinet with readout electronics that includes a low-voltage power supply, PC computer with MCA card, and printer. The vibration and acoustic sensitivity of the detector was investigated with an acoustic generator installed on the vessel of a prototype detector. It was found that the energy resolution at 662 keV and 1.33 MeV is stable up to 70-dB acoustic level [5]. With the vibro-insulation used in the Constellation HPXe-1000 system, the system is capable of operating in industrial facilities and in the vicinity of roads with heavy traffic.

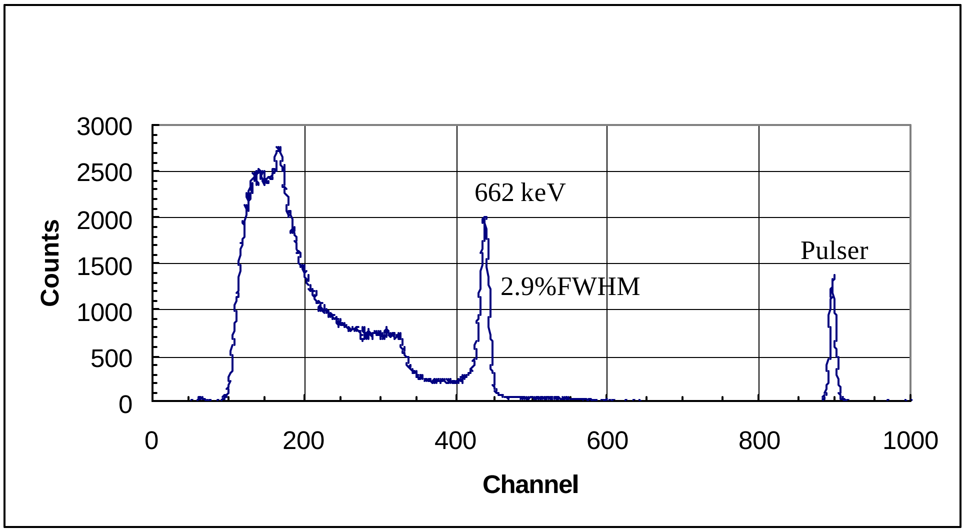

Figure 1. Spectrum of ^{137}Cs measured with HPXe-500 portable spectrometer

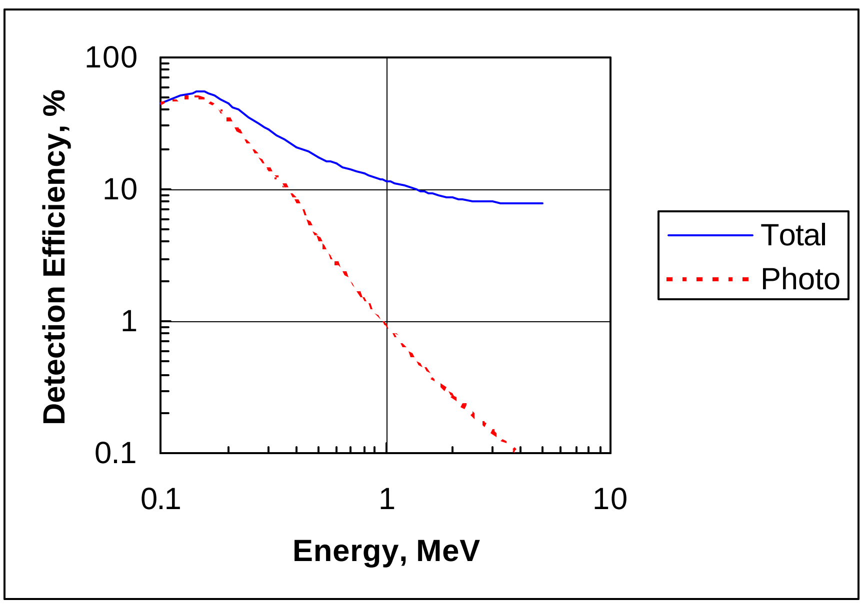

Figure 2. Calculated detection efficiency of HPXe-1000 and HPXe-500 gamma ray spectrometers filled with 0.32 g/cm^3 dense Xe in direction perpendicular to the detector axis

APPLICATION OF HPXe-1000 IN PORTAL SAFEGUARD SYSTEMS

The sensitivity of a prototype of the HPXe-500 detector for detection of special nuclear materials has been experimentally determined in a set of spectrometric measurements with a lead shielding of different thicknesses and compared with background spectra [3]. It was shown that ^{235}U and ^{239}Pu shielded with only a few millimeters of lead very efficiently masked most of their gamma ray signatures.

Under the contract with DTRA, the HPXe-1000 detector has been evaluated as a potential sensor for portal monitoring of nuclear materials. The following tests and investigations were undertaken:

1. The sensitivity of HPXe-500 for detection of two special nuclear materials, ^{235}U and ^{239}Pu, has been experimentally determined in a set of spectrometric measurements with different thickness of a lead shielding and compared with background spectra.
2. HPXe-1000 mobile spectrometer was tested to detect a set of calibrated gamma ray sources of ~1 mCi total activity in the range of 0.1 - 1.0 MeV placed in a shielding device called Nuclear Device Simulator, which consists of three enclosed cylindrical containers made of graphite, lead, and Nylon-6,6 as described in [4].
3. The experimentally determined background sensitivity and detection efficiency of HPXe-1000 was used for simulations of a vehicle portal of SNM storage facility. In these calculations:
    - A single portal monitoring site of 5x5 square meters on one side of the access road near to the storage facility outer wall/fence
    - 12 meters effective distance from source to detector
    - 10 to 100 sec of vehicle stop time for detector sampling
    - Different shielding configurations for weapon-grade plutonium in 0.1-10 kg amount

Analysis of all experimental and theoretical data shows that the Constellation HPXe detectors would have the capability to detect vehicle-borne weapon-grade Pu or enriched ^{235}U in kilogram amounts, without special shielding or with moderate shielding, and reasonable sampling times in the counting rate mode. For heavier shielding scenarios, neutron detection must be the primary means to initially detect the presence of neutron-emitting radioisotopes in a vehicle. If neutron detection should raise an alarm, there would be sufficient justification to conduct a more detailed inspection of a suspect vehicle using HPXe gamma detectors at closer range and/or longer sampling time. HPXe detectors filled with $Xe+^{3}He$ gas mixture could be used for detection as gammas and neutrons.

APPLICATION FOR NUCLEAR MONITORING IN HARMFUL CONDITIONS

Prototype of HPXe detectors with parallel-plate electrode system have been tested under hard radiation conditions typical for a space orbital station [6]. During 6 years of operation, the detector did not change energy resolution. There is a positive experience of using HPXe at 120^{o}C with a specially designed preamplifier [7]. Instead of a transistor, a ceramic vacuum tube has been used in this preamplifier. There is no temperature and radiation degradation, as well as no charge trapping in xenon itself. Radiation stability of the Xe working medium is practically unlimited. All of these features make HPXe detectors ideal candidates for monitoring nuclear waste tanks and other nuclear installations with hard radiation conditions.

Figure3 presents a spectrum of ^{137}Cs measured with the Constellation HPXe-100 detector in the presence of very intensive 6 MeV gamma-radiation resulting from neutron activation of water. The experiment models conditions in a heavy water nuclear reactor and demonstrates the ability of the detector recognize relatively low intensive nuclear radiation in intensive radiation fields.

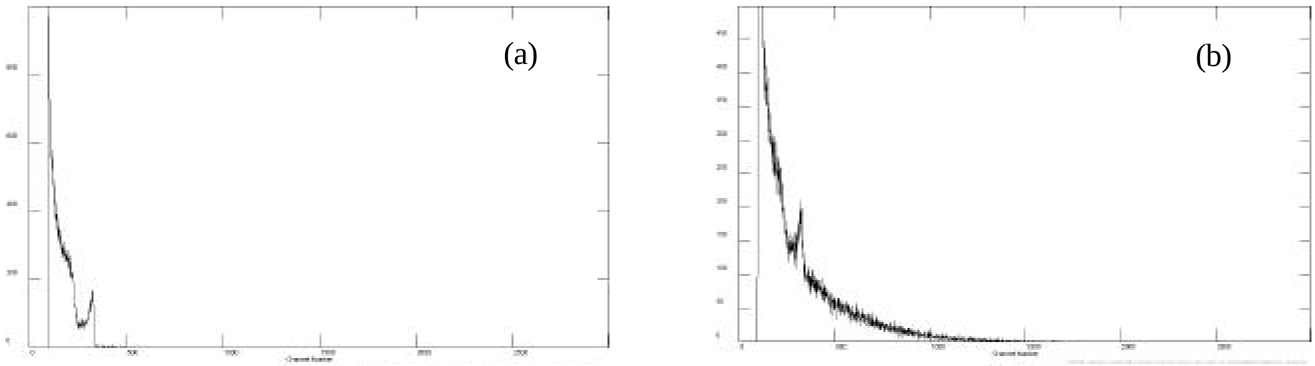

Figure 3. Spectral sensitivity of Constellation HPXe-100 detector to 5 mCi ^{137}Cs source in absence (a) and in presence (b) of 6 MeV radiation (2 mrem/h) from neutron activated water. Requisition time is 590 sec (a) and 308 sec (b).

SUMMARY

Constellation HPXe detectors are feasible for applications in detector systems for identification of nuclear weapons and weapon grade materials, portable spectrometers for use by law enforcement and military personnel in the field, autonomic nuclear monitoring systems in remote locations and storage facilities, and security portal systems to support nuclear non-proliferation. Both in experimental modeling and computer simulations, HPXe has shown a good potential for detecting kilogram amounts of nuclear materials passing through the vehicle portal of a facility designated to store special nuclear materials.

The HPXe detectors have been successfully demonstrated as high-sensitivity, robust, and stable gamma-ray spectrometers for use in harmful conditions. With the development of high-temperature preamplifiers, these detectors have no competitors between high-resolution gamma spectrometers for application in monitoring nuclear waste and nuclear reactors.

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