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Development of a method for liquid xenon purification using a cryogenic centrifugal pump

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Abstract

We are developing a new type of photon detector for an experiment to search for muons decaying into a positron and a gamma ray. In this experiment, the photon detector will utilize liquid xenon (Xe) as the scintillation material. Good transparency of the liquid Xe is required in order to gain the highest performance out of the detector. Impurities like water and oxygen must be removed efficiently for this purpose. We have developed a new purification system, dedicated to removing water from liquid Xe, by employing a cryogenic centrifugal pump and molecular sieves. The performance of the system is described in this article.

Keywords: Liquid xenon; Purification; Centrifugal pump; Low-temperature fluid

1. Introduction

Our group is developing a gamma-ray detector outlining liquid Xe as a scintillation material. This detector is planned to be used in a muon rare decay search experiment (MEG).

We built a prototype with 100-l volume to execute various developments, and have performed basic studies necessary for the construction and operation of the actual detector with 800-l liquid Xe [1]. We have proved in these studies that there is no difficulty for performing long-term stable operation of the detector as large as the actual one.

One of the most important results in these studies is that it is indispensable to remove remaining water in the liquid for obtaining the highest performance out of the detector [1]. Recently, we have developed a new purification system using a cryogenic centrifugal pump with superior performance, and executed a test for evaluating its performance [2]. In this article we introduce this new purification system and report on its performance.

2. Purpose of the test

The experiment, MEG, is searching for muons decaying into a positron and a gamma ray ($\mu \rightarrow e\gamma$ decay) at Paul Scherrer Institute in Switzerland, where the most intense muon beam in the world is available [3]. The $\mu \rightarrow e\gamma$ decay is suggested to exist by various theoretical models that incorporate new physics beyond the standard model of the elementary particle physics, and there is a high possibility that decay will be observed in this experiment.

In the MEG experiment, a gamma-ray detector with 800-l liquid Xe is adopted to detect gamma rays from $\mu \rightarrow e\gamma$ decays with good energy, angle, and time resolutions. The

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Table 1
Properties of liquid Xe

Property	Unit	
Saturation temperature at 0.10 MPa	T (K)	164.78
Latent heat (boiling)	Lv (J/kg) $\times 10^3$	95.8
Latent heat (melting)	Lv' (J/kg) $\times 10^3$	1.2
Specific heat	C_p (J/kg) $\times 10^3$	0.3484
Density	ρ (kg/m^3) $\times 10^3$	2.947
Viscosity	μ (Pa-s) $\times 10^{-4}$	5.08
Temperature/pressure at triple point	T_t (K)/P_t (MPa)	161.36/0.0815

advantageous features of liquid Xe are the first response of its scintillation light with a statistically sufficient amount of yield and strong stopping power for gamma rays due to its high atomic number and high density. It is also considered that we can easily increase the detector size without losing uniformity, because it is a liquid.

The properties of liquid Xe are given in Table 1. The liquid is kept at 167 K and 0.12 MPa during normal operation. Cooling is done with a pulse-tube refrigerator mounted on a cryostat. The refrigerator was developed and specially optimized for liquid Xe cooling by our group [4].

The wavelength of the scintillation light from liquid Xe has a central value of 174 nm in the ultraviolet region. The scintillation light is a result of photo emission from excited Xe molecules; thus, there is no absorption of the scintillation light by Xe, itself [5]. The scintillation light, however, can be absorbed by impurities, like water and/or oxygen, when they contaminate in the liquid because their photon absorption cross sections overlap the wavelength distribution of the scintillation light, causing a position dependence of the detector response [6]. This kind of dependence is crucial for our detector, which covers a large acceptance; it is necessary to substantially reduce any impurities. This is a reason why we developed a purification system, as already reported [1]. At that time, our purification system was designed to remove impurities by flowing gas Xe through a metal-heated getter after taking liquid Xe out of the cryostat and evaporating it (gas-phase purification). Purified Xe was returned to the cryostat and liquified again in it. However, the circulation speed was limited and additional cooling power was required to purify Xe in this method.

We recently developed a new purification method to improve the performance by employing a cryogenic fluid pump to circulate Xe in the liquid phase through a purifier. It is expected that this method will help us to minimize the cooling power consumption as well as to improve the purification speed.

3. Liquid xenon purification system

It was figured out during our study of the gas-phase purification that the main component contributing to scintillation light absorption was water. This is because the light absorption cross section of water is largest around the wavelength region of the Xe scintillation light. In addition, because we cannot heat the detector while evacuating the cryostat due to photomultipliers placed inside, water can easily stay and exude into the liquid after liquefaction. For this reason, we designed the new purification system to concentrate on removing water.

We prepared a purifier cartridge of 500 cm^3 filled with molecular sieves (MS13A) with a filling density of 0.59 g/cm^3. The cartridge is surrounded by a cylinder containing heaters with a low emission rate of dust and impurities provided by Watlow Electric Manufacturing Company. The purifier cartridge can absorb more than 24 g of water after regeneration, although that depends on the water contamination level in the liquid. We summarize the specifications of the purifier cartridge in Table 2.

We adopted a cryogenic centrifugal pump produced by BarBer-Nichols Inc., shown in Fig. 1. The specifications

Table 2
Specification of the purifier cartridge

Purifier cartridge dimensions	58 mm (dia.) 184 mm (height)
Absorbent	Molecular sieves (13 A) 1/16 inch, pellet type
Absorbent filling factor	0.59 g/cm^3
Water absorption capacity	>24 g
Design pressure	0.4 MPa
Operation temperature	165 K

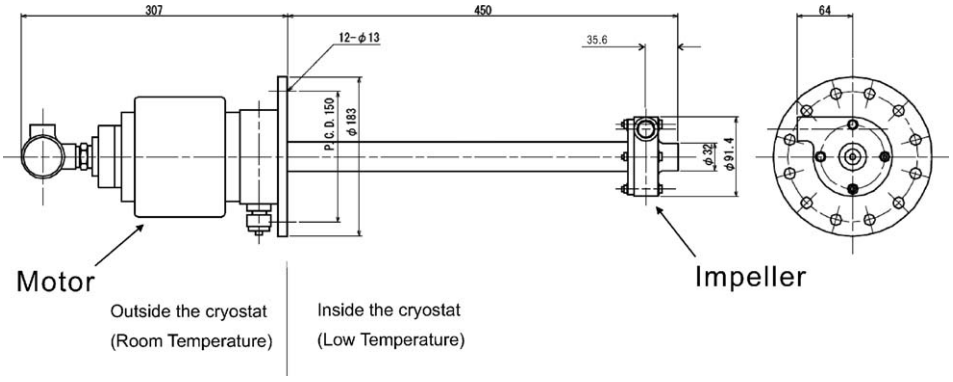

Fig. 1. Cryogenic centrifugal pump used in this purification test. The pump is designed to achieve a liquid-Xe flow rate of 100 l/h with a differential pressure of 0.2 MPa.

Table 3
Design point operating conditions of the cryogenic centrifugal fluid pump

Fluid	Liquid Xe
Inlet pressure (absolute)	0.1014 MPa
Inlet temperature	165 K
Flow rate	100 l/h
Differential pressure	0.2 MPa
Approximate operating speed	3175 rpm

of this pump are summarized in Table 3. It is possible to control the operating speed by modifying the input power frequency with a frequency inverter. At the normal working point (3175 rpm rotation speed of the pump head) the pump can flow 100 l of liquid Xe per hour. Under the normal operating condition, 5000 h of operation is guaranteed by the manufacturer, and after that maintenance work, such as exchanging bearings, is recommended. In the MEG experiment 800 l of liquid Xe will be used in the detector, which means that we can perform more than 600 purification cycles in 5000 h and do not need any maintenance work during the experiment.

We placed the centrifugal pump, purifier cartridge, and cryogenic valves controlling the liquid flow on a 40 cm diameter stainless-steel plate. The plate was mounted on a liquid-Xe detector prototype (Fig. 2). A safety valve with a pressure limit at 0.34 MPa was equipped at the exit of the purifier cartridge and a vacuum port was prepared outside of the cryostat in order to regenerate the molecular sieves. A detailed description of the Xe detector prototype can be found in Ref. [1].

We had been using a pulse-tube refrigerator for cooling purpose during prototype tests [4], but this time we used only liquid nitrogen because the refrigerator had to be removed due to a limitation of the space on the top plate. Fig. 3 shows a picture of the purification system and its schematic view.

Liquid Xe pumped from around the surface is passed through the purifier cartridge and then returned to the bottom of the detector through a teflon tube equipped inside the cryostat. Thus purified liquid Xe can be distributed uniformly inside the detector. A platinum resistance thermometer (PT100) is set at the teflon tube exit to confirm that Xe does not evaporate on the path of purification. Two more thermometers are placed just above the pump head in order to monitor the liquid surface level for avoiding the impeller from running idle. Because the height of the impeller from the inlet is about 3.5 cm (Fig. 1), a liquid surface level of about 5 cm above the inlet is sufficient. When we performed the purification test, the surface of the liquid Xe was kept at the level illustrated in Fig. 4, so that the liquid would not touch the liquid-nitrogen cooling pipe.

Several polonium α sources are placed in the detector, and a constant amount of scintillation light is always emitted around them. Because the range of the polonium α in liquid Xe is about 40 μm, light emission sources can be considered to be point-like. There are 238 photomultipliers [7] placed in the detector that detects scintillation light from liquid Xe, and output as an electric signal. The PMTs are placed at different distances from α sources, enabling us to estimate the absorption effect of scintillation light in liquid Xe by applying a solid-angle correction to the data. Based on this estimation we can evaluate the absorption length in liquid Xe by comparing it with a simulation by taking into account the effect of Rayleigh scattering. Additionally, it is also possible to estimate the impurity level in the liquid if we suppose that absorption in the liquid is caused only by water.

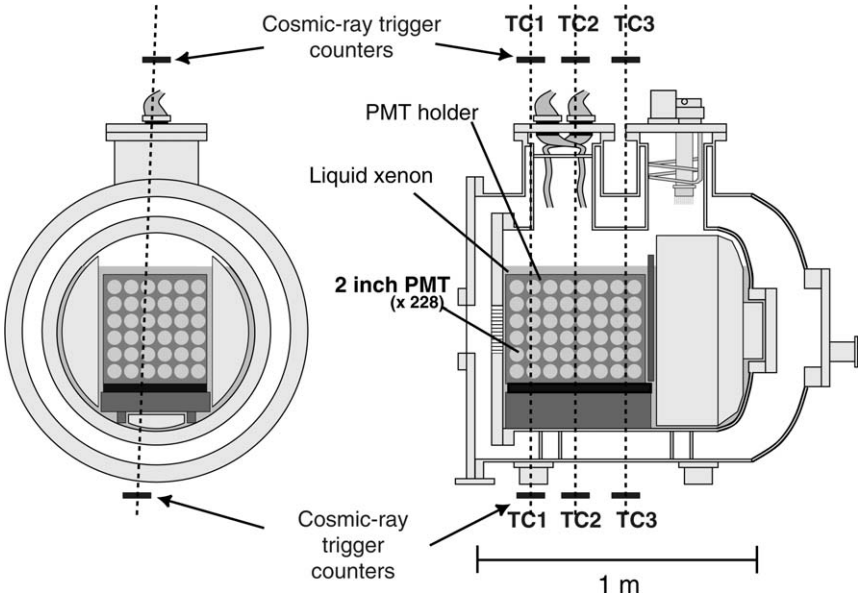

Fig. 2. Schematic view of the large liquid-Xe calorimeter prototype.

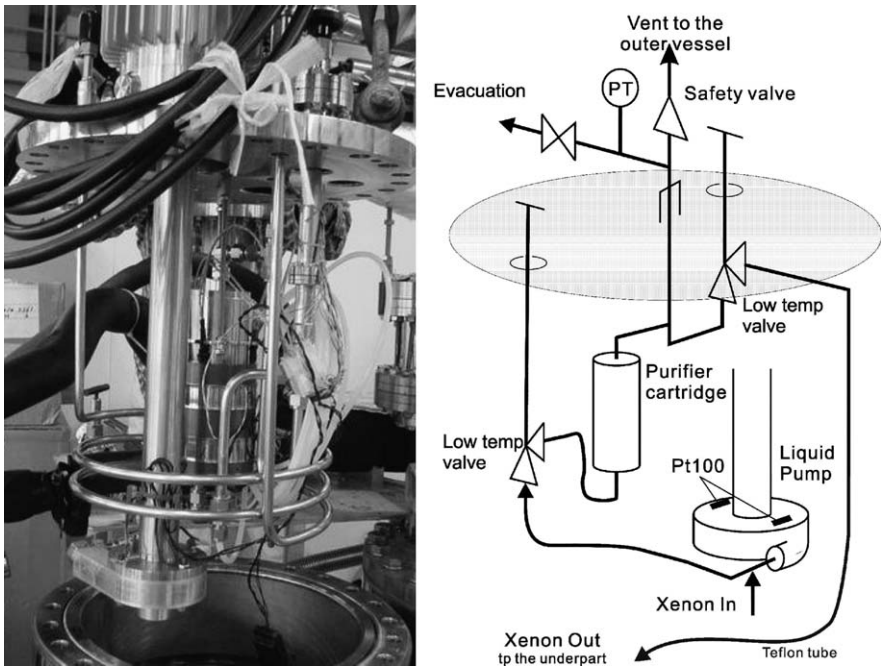

Fig. 3. Purification system to be inserted to the prototype detector (left). Schematic diagram of the purification system (right).

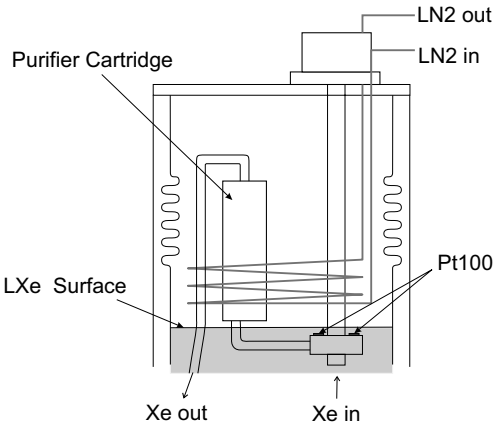

Fig. 4. Schematic view of the pump head, purifier, and liquid-nitrogen cooling pipe. The liquid-Xe surface level is monitored with two PT100 thermometers equipped above the pump head.

There are three sets of counters above and below the detector to trigger cosmic-ray events. These can also be used for evaluating the impurity level.

4. Liquid xenon purification test

4.1. Procedure

A purification test was performed in the following way. First the molecular sieves were regenerated in parallel with evacuation of the cryostat. The purifier cartridge was heated up to 250 °C and continuously evacuated for 4 h. Normal air (15 °C, 20% humidity) was fed in the cryostat afterward in order to artificially introduce water inside the detector, and then the cryostat was evacuated again. The liquid-nitrogen cooling pipe was cooled during the second and later evacuation for keeping water to stay in the cryostat. This procedure, feeding air and evacuating while cooling the pipe, was repeated three times before starting Xe liquefaction. Liquefaction was performed in the usual way, and after confirming the pump head in the liquid we started the pump operation. Fig. 5 shows the pressure and temperature of the cryostat and purifier cartridge as

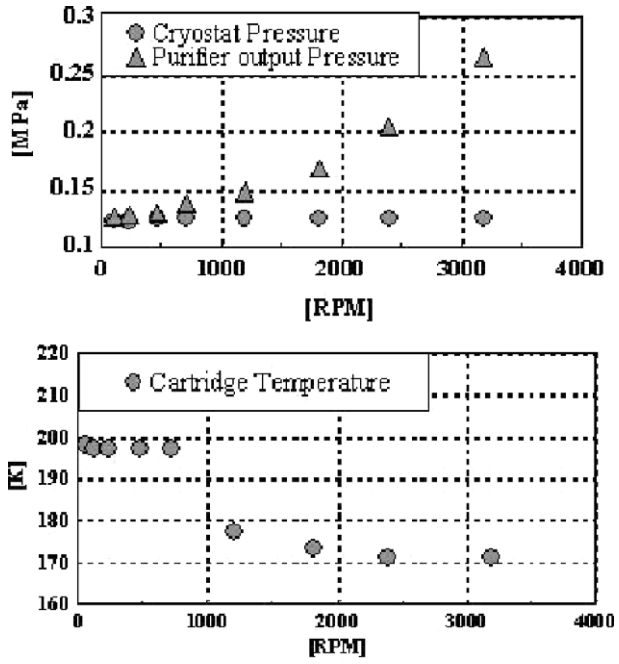

Fig. 5. Cryostat and purifier cartridge pressure (top) and cartridge temperature (bottom) as a function of the pump rotation speed.

a function of the rotation speed of the pump head. It can be seen that the purifier cartridge pressure increased as the pump rotation speed increased. We observed that the cartridge temperature suddenly decreased when the pump rotation speed exceeded 1000 rpm, indicating that the cartridge was filled with liquid and stable liquid circulation started at this point.

We started to count the purification time after confirming that purifier pressure and temperature had stabilized. When we were performing purification, we acquired α-particle and cosmic-ray event data to evaluate and monitor the absorption length. A detailed data analysis was executed offline to investigate the change of the absorption length.

We observed an increase of the liquid-nitrogen consumption immediately after we started to circulate liquid Xe. This is attributed to additional work provided to the system by the pump. It is reported in [1] that 52 W of cooling power is required for normal operation without using a pump. In this test 2.2-times amount of liquid nitrogen was consumed, indicating that an additional 62 W of heat was generated in the cryostat with pump operation.

We completed this purification test in 12 days and succeeded to operate the system for 70 h in total, corresponding to circulating 7000 l of liquid Xe.

4.2. Result

Here, we describe the analysis procedure of α-particle data. As already mentioned, it is necessary to take into account the Rayleigh-scattering effect for evaluating the absorption length. Therefore, we normalized the observed photomultiplier outputs at each stage of purification to those when the photomultiplier outputs saturated after sufficient purification cycles. This enabled us to investigate only the absorption effect because the Rayleigh-scattering effect does not depend on the impurity concentration in the liquid Xe. We then obtained a distribution of the normalized photomultiplier outputs as a function of the distance between the photomultiplier and the α source, and evaluated the slope parameter of the distribution by fitting with an exponential function. The slope parameter is equivalent to the absorption length of scintillation light in the liquid Xe. That slope parameter is shown in Fig. 6 as a function of the purification time. The shaded areas in the figure show periods when the purification system was operated.

We can plainly see in the figure that the absorption length improved rapidly after the purification was started, although the measurement accuracy was limited for an absorption length as long as 3 m (corresponding to a slope parameter of less than 0.003), because of a limitation due to the detector size. We observed at an early stage of purification that the absorption length fell back slightly when we stopped the pump. This was supposed to be caused by water dissociation from the material surface, and the impurity level reached equilibrium after a while. Needless

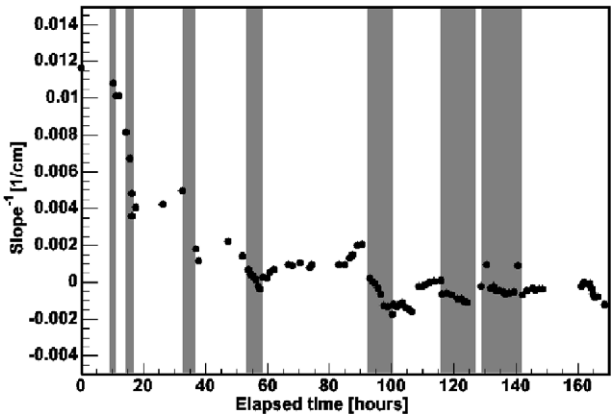

Fig. 6. Change in the slope parameter as a function of the purification time.

to say, this problem was resolved as we continued purification, and finally we did not observe any remarkable change of the absorption length, even 50 h later after we stopped the purification pump.

We also analyzed the cosmic-ray event data to evaluate the absolute value of the absorption length. We compared the data with a simulation that took into account a realistic Rayleigh-scattering effect. The absorption length was supposed to be infinite in this simulation. By comparing the total amount of light observed by all photomultipliers with the simulation, it was proved that the absorption length was consistent with infinity when the photomultiplier outputs saturated. Furthermore, the absorption length before starting the purification was estimated to be 85 cm by this method. This is consistent with that obtained in the α-event data analysis, where the absorption length was estimated to be 83 ± 3 cm.

4.3. Discussion

As described above, the absorption length improved as the impurity was removed by purification. Actually, this could be completed only in 22 h of actual purification time. Compared to the gas-phase purification case, in which 300 h had been necessary to remove impurities to well below the required level, the newly developed system can purify liquid Xe in an extremely short time. It is estimated that there was a 250 ppb water-equivalent impurity contained in the system at the beginning; the concentration was reduced down to 1/6 (about 40 ppb) in only 5 h, including the conditioning time of the system.

We show in Fig. 7 the water-equivalent impurity concentration estimated from the measured absorption length as a function of the integrated purification time. At the beginning the system was conditioned with a slower pump head speed ($\leqslant$2000 rpm), resulting in a slower decrease of the impurity concentration due to the low output pressure of the pump and a lower flow rate, as shown in Fig. 5. However, we could observe a rapid decrease of the impurity concentration after we had started to operate the pump

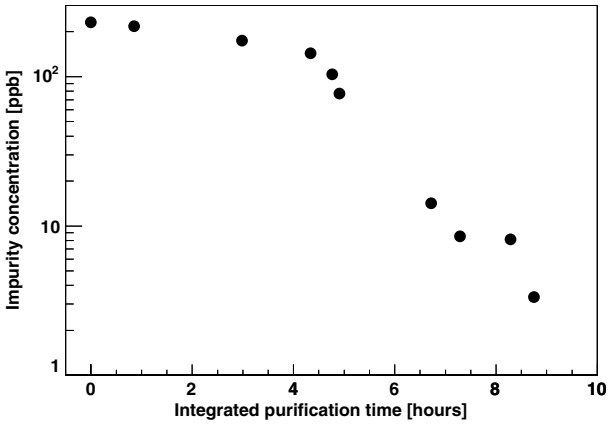

Fig. 7. Change in the water-equivalent impurity concentration as a function of the integrated purification time.

at the normal speed (3175 rpm). The impurity concentration can be reduced as an exponential function of the purification time if we suppose a simple model where the impurity removal rate is proportional to the concentration at that point. Although the measurement accuracy deteriorates when the absorption length is as long as the detector size, the impurity concentration changed exponentially, as shown in Fig. 7, indicating that the impurity was removed very efficiently.

An additional cooling power of 62 W was necessary for the pump operation. Power provided by the pump to the system was estimated to be 55 W, based on the pressure difference (2 Bar) and liquid-Xe flow rate (100 l/h), which is consistent with the required additional cooling power. This proves that the pump was working with the expected performance in the system.

We used only molecular sieves as an impurity absorber in this purification test because we focused on removing only water impurity in the system. There is, however, no principal problem to upgrade the system in such a way that oxygen can also be removed by using an oxygen absorber, like Oxisorb, in addition to the molecular sieves. Actually, it is reported that the oxygen-equivalent impurity concentration was successfully reduced to less than 0.1 ppb with a similar system [8] in liquid argon (Ar), where the impurity level was estimated by measuring the electron absorption cross section in liquid Ar.

5. Conclusion

We have developed a new purification method for liquid Xe using a cryogenic pump. We adopted a fluid pump with a flow rate of 100 l/h and molecular sieves as a water absorber, and successfully reduced the impurity concentration from 250 ppb to 40 ppb in 100 l of liquid Xe within 5 h. This is an adequate performance for the MEG photon detector as well as others using liquid-Xe scintillation light from the view points of the purification time and purity level.

Construction of the MEG liquid Xe photon detector is presently in progress. After completing it we plan to execute a full test of the detector operation with 828 photomultipliers installed. Based on the result obtained in this study, we also plan to perform a purification test of 800 l of liquid Xe in the liquid phase.

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