

DISTILLATION PURIFICATION OF XENON FOR KRYPTON AND MEASUREMENT OF RADON CONTAMINATION IN LIQUID XENON

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ABSTRACT: We succeeded to reduce the Kr contamination in liquid xenon by a factor of 1/1000 with a distillation system in Kamioka mine. Then, the remaining radioactivities in purified liquid xenon were measured with the XMASS prototype detector. In this report, the distillation system and the remaining internal radioactivity levels are described.

1. INTRODUCTION

Liquid xenon is a good scintillator. It has a good photon yield (about 42photon/keV). Therefore, sensitivity against low energy events is good. The wave length (~175nm) is rather short, but it still can be observed by PMTs directly without a wave length shifter. The large atomic number provides a short radiation length for the low energy gamma rays. So, the external low-energy gamma rays would be absorbed in the outer region of the liquid xenon detector, and then the central region would become clean. The boiling point of liquid xenon is higher than other rare gas, like argon or neon, so it is relatively easy to handle. We can use liquid nitrogen to liquefy xenon. Because of rare gas and liquid, various purify methods can be applied. Xenon doesn't have long life radioactive isotopes.

A possible problem of liquid xenon for the low background experiments is that there is a contamination of ^{85}Kr during manufacture and refinement. Usual commercial “krypton-free” xenon still has ppb level krypton. It could become a serious background for the low background experiments. We have built a distillation system to reduce the Kr contamination mainly, and then processed 100kg of xenon. We have measured remaining krypton and radon levels after the distillation purification.

2. DISTILLATION PURIFICATION OF XENON FOR KRYPTON

Impurities, like CO_2 or H_2O , in xenon can be removed by adsorption method. A distillation method can be used to remove Kr, O_2 , N_2 , H_2 , or He in xenon, because their boiling points are lower than xenon. We have designed a distillation system to remove krypton in xenon. Table 1 shows a summary of the requirements for the purification system.

Item	Design value
Process speed	0.6 kg/hour
Yield (Kr-less gas / raw gas)	99%
Reduction factor for Kr in Xe	1/1000
Operation conditions	178 ~ 180 K, 2 atm.

Table 1 Requirements for the distillation system of Xe for Kr.

We have built the system in Kamioka Underground Observatory, ICRR. Univ. of Tokyo, which is located 1000m underground (2700m.w.e.) in Kamioka Mine in Gifu Prefecture in Japan. Figure 1 shows a photograph of the distillation system in Kamioka. Packings are used in the tower in order to accommodate 13 stages in the distillation tower, because the cavity height is limited in the underground laboratory.

We have processed 100kg xenon by using the distillation purification system in March 2004. It took about 1 week to process it. The raw xenon had about 3ppb krypton. We have separated about 1kg of “off gas” xenon with 330 ± 100 ppb krypton from the raw xenon. The amount of the purified xenon was about 99% of the raw xenon. The krypton contamination in the purified xenon was assessed by a special device made with an Atmospheric Pressure Ionization Mass Spectrometer (API-MS) system and a Gas Chromatography (GC) system.



Figure 1 The distillation system in Kamioka Observatory. The height of the distillation tower is about 3m.

3. KRYPTON ASSAY WITH GC+API-MS SYSTEM

The API-MS + GC system for krypton assay in xenon was originally developed at Taiyo Toyo Sanso Co. Ltd. (SAAN). We have measured the krypton contamination in the purified xenon by using their API-MS + GC system. In an API-MS, primary ionization is done by usual corona discharge. However, it is done in atmospheric pressure. Therefore, if the carrier gas (C) has a higher ionization potential than the target molecules (X), there is the secondary ionization by ion-molecule reaction of $C^+ + X \rightarrow C + X^+$.

In this reaction, most of the target molecules are ionized by charge exchange, and then the statistical sensitivity of the Mass Spectrometer part becomes high. Table 2 shows the ionization potential of related gases. In the API-MS detector, He or Ar is used as a carrier gas. They have higher ionization potentials than Kr, so it satisfies the condition. Xenon, however, has lower ionization potentials than Kr. Therefore, xenon has to be removed before API-MS detector so as not to ionize xenon atoms instead of krypton atoms. So, a GC system is used to separate krypton from xenon.

	He	Ar	Kr	Xe
eV	24.6	15.8	14.0	12.3

Table 2 Ionization potential.

Figure 2 shows a photograph of the CG + API-MS system at SAAN, and Figure 3 shows a schematic view of the GC + API-MS system.

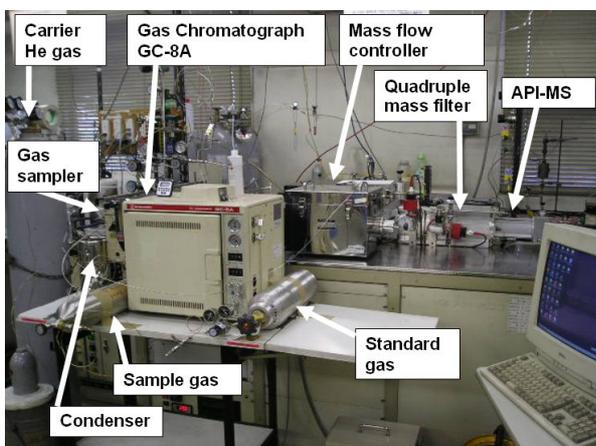


Figure 2 The Gas Chromatography and Atmospheric Pressure Ionization Mass Spectrometer system at SAAN.

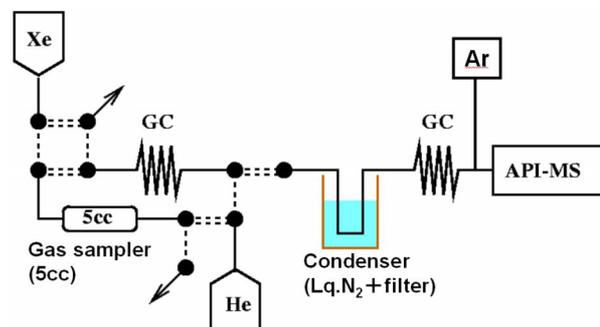


Figure 3 A schematic of the flow of the GC + API-MS system.

We have measured the krypton contamination in our purified xenon by using their API-MS + GC system in September 2004. In the first step of the measurements, xenon gas was fed into the gas sampler, and then 5cc of the gas was sampled. Then, the sample xenon gas was fed into the GC with He carrier gas. Only at the krypton timing, the sample gas was fed into the condenser, then krypton was trapped in the condenser. To increase statistical significance, we repeated these steps by 100 times. Finally, the condenser was heat up, the trapped gas was fed into the API-MS system with He and Ar carrier gases through the GC system. In the final step, only krypton timing was also selected. Actually, the API-MS counts ^{84}Kr which is most dominant krypton isotope. The abundance of ^{85}Kr is small. The $^{85}\text{Kr} / \text{Kr}$ ratio is about 10^{-11} . The measured ^{84}Kr counts are the following.

Sample xenon gas	184.0 ± 15.8 count
Carrier He gas	$(88.5 \pm 23.9) / 0.92$ count
Krypton 50 ppt standard gas	$(1176.9 \pm 35.7) / 0.88$ count

Then, the krypton concentration in the sample xenon gas was obtained as $3.3 \pm 1.1(\text{stat.})$ ppt. This corresponds to about 1/1000 factor reduction of krypton, because the initial krypton contamination in the raw xenon gas was about 3ppb.

4. RADON ASSAY WITH XMASS PROTOTYPE DETECTOR

XMASS is an underground experiment aimed at searching for rare phenomena under an ultra low background environment by using ultra pure liquid xenon in Kamioka mine, Japan. The main physics targets of XMASS are cold dark matter, neutrinoless double beta decays, and low-energy solar neutrinos. So far, we have developed a prototype detector and done 2 series of test experiments with it. By using the prototype detector, we have assessed the remaining radon in liquid xenon.

Figure 5 shows a schematic view of the prototype detector. The prototype detector consists of 100kg of liquid xenon and 54 low-background Photo-Multiplier Tubes (Hamamatsu R8778) developed for this experiment. It is installed in a heavy gamma ray shield in a clean room in Kamioka Observatory. The charge information from the PMTs are read by 54 channels of charge sensitive ADCs and a common Flash ADC for summed signals. The threshold of each PMT is about 0.4 photo electrons. The data acquisition trigger is applied when there are 4 multiple hits within 100ns time window. The typical trigger rate for normal runs is about 1.5Hz.

The radon assay was done through the following coincidence reactions.

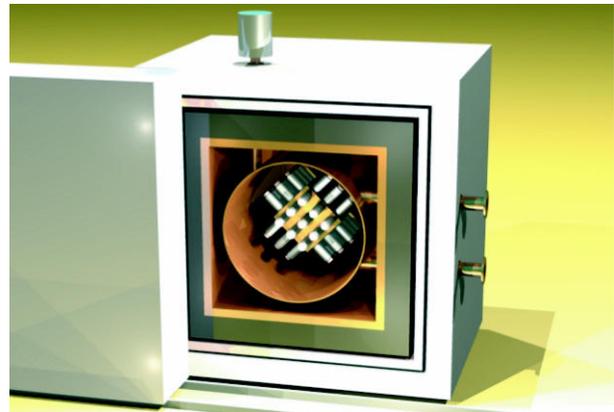
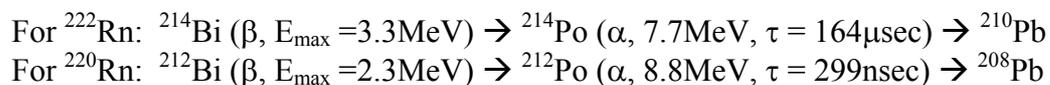


Figure 5 XMASS prototype detector. 100kg liquid xenon and 54 low-background PMTs are used. PMTs are attached to the 30L OFC chamber through 5mm thickness MgF_2 windows. They are put into a heavy shield in a clean room. The XMASS prototype detector is located in Kamioka Observatory, ICRR, Univ of Tokyo, in Kamioka mine.

In the test run in August 2004, we found 67 candidate events of the ^{222}Rn during 1.8 days observation. Figure 6 shows the result of the ^{222}Rn assay.

Figure 6(a) shows the daughter energy distribution of all the time-coincidence events within 1msec. In the low-energy region, there are accidental coincidence events. Therefore, we have applied a cut at 3.5MeV in Fig. 6(a). Then, the remaining events became 67 events. Figure 6 (b), (c), and (d) show the distribution of these 67 candidate events. They agree with expectations.

This measurement was done on 4th and 10th August, 2004. The live times are 0.8 day and 1.0 day, respectively. If we assume radioactive equilibrium, the remaining ^{238}U becomes like below.

$$4\text{th: } ^{238}\text{U} = (72 \pm 11) \times 10^{-14} \text{ g/g}$$

$$10\text{th: } ^{238}\text{U} = (33 \pm 7) \times 10^{-14} \text{ g/g}$$

The decrease of the remaining ^{238}U is consistent with the expected ^{222}Rn decay of 3.8 day half life. So, we think most of the remaining ^{222}Rn might come from outside the detector. We are planning to install a special device for radon reduction in future.

We didn't find any candidate events for ^{220}Rn . Then we have set a limit as $^{232}\text{Th} < 23 \times 10^{-14} \text{ g/g}$.

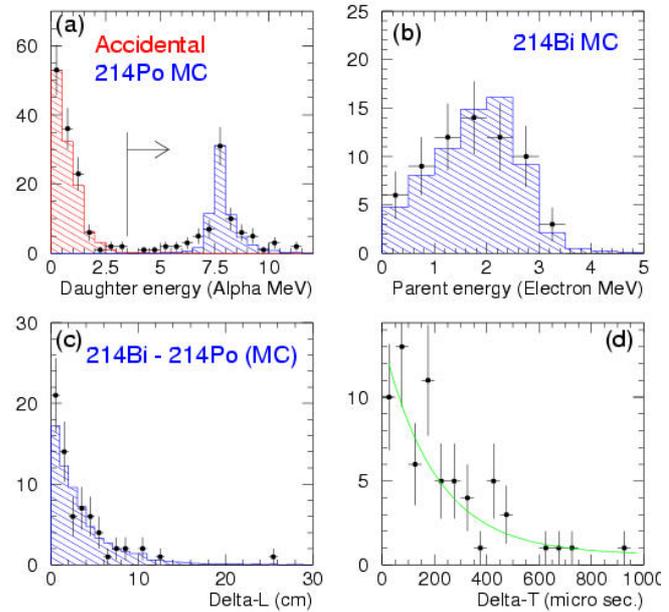


Figure 6 ^{222}Rn measurements in the test run in August 2004. (a) Energy distribution of the daughter event of the coincidence events within 1msec. (b) Energy distribution of the parent events. (c) Distance between the reconstructed vertex positions of parent and daughter events. (d) Time difference between the parent and daughter events. The fitted half life is $141 \pm 51 \mu\text{sec}$.

5. SUMMARY

We have built a distillation system of xenon for krypton removal in Kamioka Observatory. The measured reduction power was 1/1000 for krypton in xenon. The remaining krypton was measured by a GC + API-MS system. The radon contamination in liquid xenon was also assessed by the XMASS prototype detector. The current remaining impurities in purified xenon for XMASS experiment are the following.

$$^{238}\text{U} = (33 \pm 7) \times 10^{-14} \text{ g/g}$$

$$^{232}\text{Th} < 23 \times 10^{-14} \text{ g/g}$$

$$\text{Kr} = 3.3 \pm 1.1 \text{ ppt}$$

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