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Purity measurement of liquid methane by using the drift property of ionized electrons

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The purity is important parameter for the collection of electrons ionized in an ionization chamber filled with the liquid methane. The methane gas purification system was developed for the liquid methane ionization chamber. After the condensation of the purified methane gas in the ionization chamber mounted with ^{241}Am alpha source, ionized electrons were collected by a charge sensitive preamplifier. The concentration of impurity of oxygen gas was obtained by analyzing a collected charge distribution. Pulse height distributions of a charge sensitive preamplifier were obtained for the detection of γ rays and neutrons emitted from ^{60}Co, ^{137}Cs and Am-Be sources located outside the cryostat of the liquid methane chamber.

KEYWORDS: neutron detection, liquid methane, ionization chamber, purity monitor

I. Introduction

Nuclear data for neutron reactions in the intermediate energy region up to a few hundred MeV are important in various fields such as accelerator shield design and the evaluation of the irradiation damage to spaceship microelectronics. Therefore, it is necessary to detect high-energy neutrons with high efficiency to establish high-accuracy database for nuclear reactions. High-energy neutrons are detected by converting to protons through the neutron-hydrogen elastic scattering. In usual experiments of high-energy neutron reactions, large liquid organic scintillators are employed for the detection of neutrons with high efficiency. The amplitude of detection signals has dependence on incident positions of neutrons in large liquid organic scintillators due to the difficulties in collection of scintillating photons. Degradation in the energy resolution is caused by the variation in the amplitude of detection signals.

Recently, a large liquid argon time projection chamber (TPC) was developed for the research of neutrino physics in the ICARUS project[1)2)]. The large liquid argon TPC utilizes ionized electrons with drift length of a few meters in the liquid argon for precise measurements of the position and the energy deposition in neutrino reactions. Since methane molecules have symmetrical structure similar to noble gases, ionized electrons have a long mean free path in the liquid methane. Liquid methane TPC is expected to supply the three-dimensional image of recoil proton generated by the incident neutron. The total collected charges by electric field indicate the energy of recoil proton. Incident neutron energy is obtained by reconstruction of the kinematics between the incident neutron and the recoil proton. Oxygen molecules in liquid methane capture drifting electrons ionized by the energy deposition of the recoil proton.

In order to construct a large volume liquid methane ionization chamber for detection of high-energy neutrons with high efficiency and excellent energy resolution, concentration of oxygen molecules is kept to be sufficiently low in the liquid methane. Therefore, after condensation of purified gaseous methane, it is important to monitor the concentration of oxygen molecules in the liquid. Characteristics of drifting electrons indicate the concentration of oxygen molecules in the liquid methane. For monitoring oxygen impurity, drifting electrons are generated in the liquid methane. In reference 3)-5), photoelectrons were generated by UV-light irradiation of a metal disk placed in the liquid methane for drifting electrons.

We constructed a liquid methane ionization chamber. The ionization chamber consists of three disk-shaped electrodes placed in parallel. Negative high voltage was applied to a cathode electrode inserted between two anode electrodes connected to the ground potential. The gaseous methane was condensed in the ionization chamber by operating a miniature refrigerator. Before condensation, methane gas was purified in a purification system. An ^{241}Am alpha source was placed on the surface of cathode disk faced to the upper side of anode disk. Since the range of alpha particles is estimated to be 50 μm in the liquid, the ionized electrons were generated near the surface of the cathode disk. A conventional charge sensitive preamplifier was used to collect the electric charge of ionized electrons drifting between the electrodes. The concentration of oxygen in the liquid methane was evaluated by analyzing the collected electric charges. By applying electric field between the cathode disk and the lower side of anode disk, pulse height distributions of output signals of the preamplifier were obtained for irradiation with ^{60}Co and ^{137}Cs γ ray source and Am-Be neutron source.

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II. Experimental set-up

1. The ionization chamber

Figure 1 shows the schematic layout of the ionization chamber composed of three parallel electrodes made of disc-shaped stainless steel 45mm in diameter. A cathode electrode was inserted between two anode electrodes at spaces of 1.2 mm from the upper- and 3.0 mm from the lower-anodes. Each anode electrode was connected the ground potential through a resistor of 100MΩ. To generate ionized electrons for monitoring the concentration of oxygen molecules in the liquid methane, an ^{241}Am α source was placed on the cathode plate faced to the upper anode. A surface of the α source was covered with thin copper sheet with a 1.5 mm hole at the center for collimation of α rays.

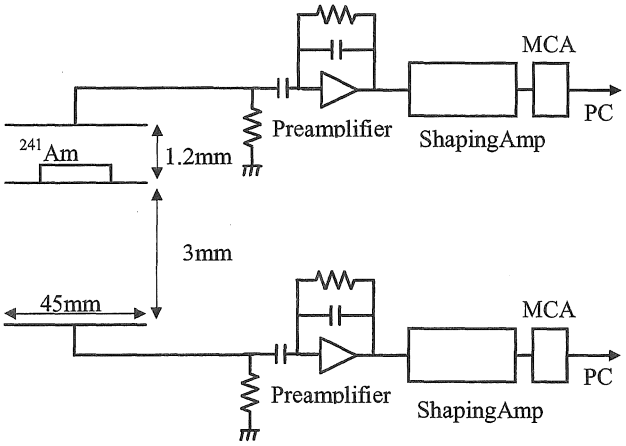

Fig.1 The schematic layout of the parallel ionization chamber

The ionization chamber was mounted in a stainless steel vessel 220cc in volume. In a large vacuum chamber , the vessel was connected to the cold head of a Gifford-MacMahon type refrigerator (Taiyo Toyo Sanso,TZ-65) through the copper braided thermal link. The electrode structure of the ionization chamber was suspended from the top flange of the vessel.

The electrical feedthroughs were welded on the top flange for high voltage supply and signal readout without breaking the vacuum isolation. Negative high voltage was supplied to the cathode through a RC filter network. Each anode electrode was connected to a charge-sensitive preamplifier with a feedback capacitor of 1pF. Two preamplifiers were contained in a preamplifier box. The preamplifier box was placed on the top flange outside the vessel to reduce the electrical noise induced in the signal readout line.

The output signal of the preamplifier was filtered by a shaping amplifier (Ortec 571) and processed by a multichannel analyzer. The pulse height of the output signal was calibrated by applying reference voltage pulses across a 1 pF capacitor connected to the preamplifier input.

The pulse height distribution measurements for ^{60}Co, ^{137}Cs γ-quanta and Am-Be neutron source were performed by the lower side of electrodes. Neutrons and γ-quanta were injected from external of insulating vessel via 1.5mm collimator.

2. Purification method

Gas flow diagram of the methane purifier system is shown in **fig. 2.** The gas line and the chamber were evacuated by a turbomolecular pump with baked at 150°C for a few days in order to reduce outgas emitted from the inner surface. The evacuation was finished at a vacuum of 1.0×10^{-3} Pa in the system. A commercial methane gas with a purity of 99.99%, which contains impurities of O_2<1ppm, N_2<20ppm, and CO_2<1ppm was introduced to the methane gas recovery vessel 400L in volume. The recovery vessel was filled with methane gas with the inner pressure of 0.2 MPa to prevent entering impurities from air. Before condensation, methane gas was purified by operating the purification system. Oxygen molecules in the methane gas are removed by using chemically binding on the adsorbent material of the Oxisorb (Messer) . A 13X-type molecular sieve trap removed moisture and CO_2 molecule.

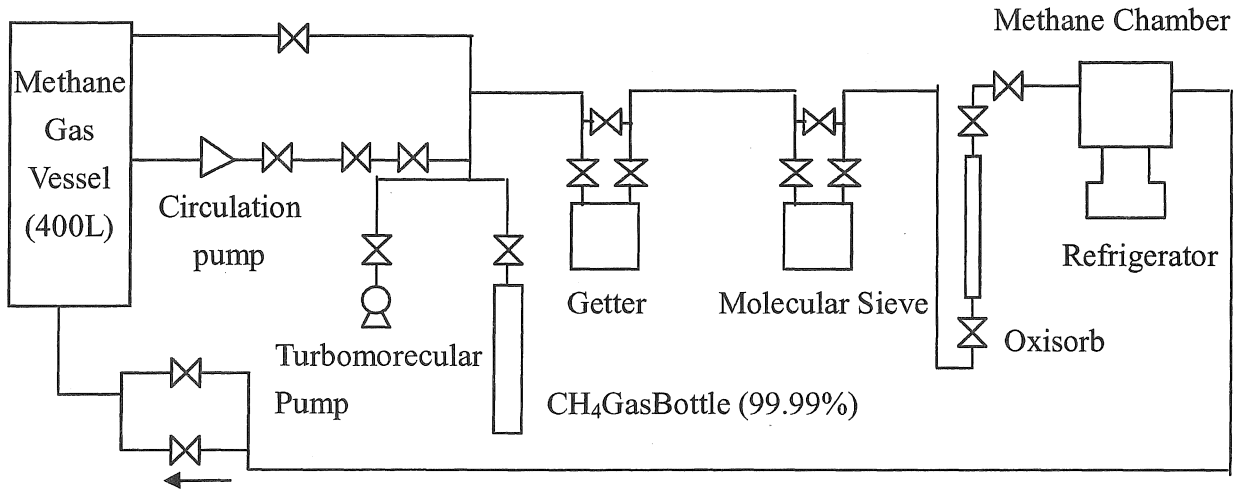

Fig.2 Gas flow diagram of the methane purifier system.

Furthermore, to improve methane purity getter-stabilized zeolites (SAES MicroTorr) was used. The methane gas was circulated in the purification system by operation of a bellows-type pump at 80°C for the purge of the inner surface of the pipe lines and the liquid methane vessel.

After purification of gaseous methane, the liquid methane vessel was cooled by operating the Gifford-MacMahon type refrigerator to condense the methane gas in the vessel. A platinum resistor was inserted into the hole of the copper clamp attached to the bottom of the liquid methane vessel for monitoring the temperature of the vessel. The liquid methane temperature was kept at –180°C within a variation of ±1°C by the use of a temperature controller (Taiyo Toyo Sanso,TS-1000) and an electric heater.

III. Results

The concentration of the oxygen molecules in the liquid methane was monitored by collecting electric charges of drifting electrons between the cathode- and the upper anode-electrodes.

The ionized electrons were generated in the vicinity of the upper surface of the cathode by energy deposition of α particles emitted from ^{241}Am α source. By applying the electric field between the upper anode and the cathode, the ionized electrons drifted towards the upper anode. Since oxygen molecule has negative electron affinity, drifting electrons were captured by oxygen. The amount of electronic charges Q of drifting electrons escaped from capture is expressed by the following equation,

$$Q = Q_o \exp\left(-XL^{-1}\right) \quad L^{-1} = CK \quad K = 1.4/E \quad (1)$$

where Q_o is the amount of electric charges of ionized electrons, X the drift length of electrons in liquid methane, C the oxygen concentration, K the attachment coefficient shown in the reference 6), E the strength of the electric field.

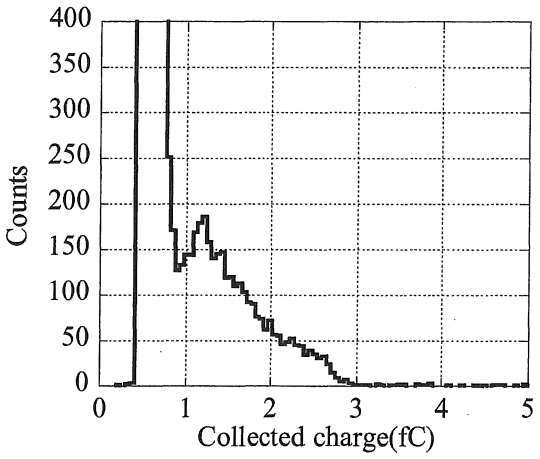

Fig.3 The charge collection distribution at 5kV/cm

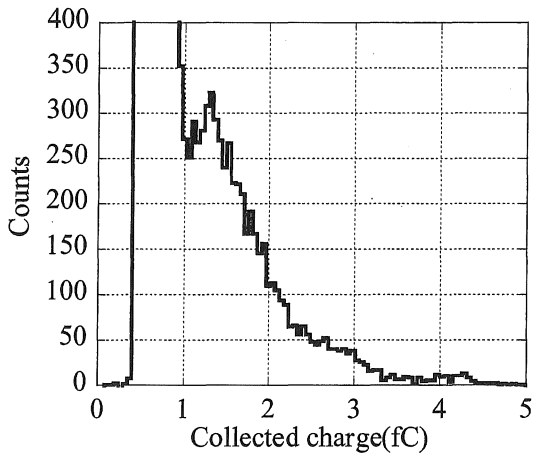

Fig.4 The charge collection distribution at 10kV/cm

The charge sensitive preamplifier collected the electric charges of the drifting electrons escaped from capture. The amount of electric charges escaped capture is indicated by the pulse height of output signal of the preamplifier. **Figures 3** and **4** show the collected charge distributions by applying the electric field of 5 kV/cm and 10 kV/cm, respectively. A peak appears in the distribution indicates the amount of electric charges of the drifting electrons escaped from capture. The values of collected charges were obtained to be 1.2fC at 5kV/cm and 1.3fC at 10kV/cm. The amount of collected charges increases with applied electric field. In **Figure 4**, the increase in the number of counts appears over the 3fC. According to the columnar recombination model, the amount of collected charge would increase with the angle between the α track and the direction of the electric field. A higher tail larger than 3fC in **Figure 4** may indicate electrons escaping from columnar recombination. The concentration of the oxygen molecules was evaluated to be 2-6ppm by using eq. (1). In the evaluation by using eq. (1), it is necessary to estimate the amount of electric charges of ionized electrons escaped from recombination in high density of ion-electron pairs created by α particles. In this work, 10% of ionized electrons were assumed to escape the recombination.

Figure 5 shows a pulse height distribution of liquid methane ionization chamber consisted of the cathode- and the lower-electrodes for irradiation with Am-Be, ^{60}Co and ^{137}Cs. The discrimination between ^{60}Co and ^{137}Cs as end of Compton edge is clearly shown. Response to the Am-Be source is continuously distributed over a high channel. Further investigation is necessary to extract the neutron events from the response to the Am-Be source.

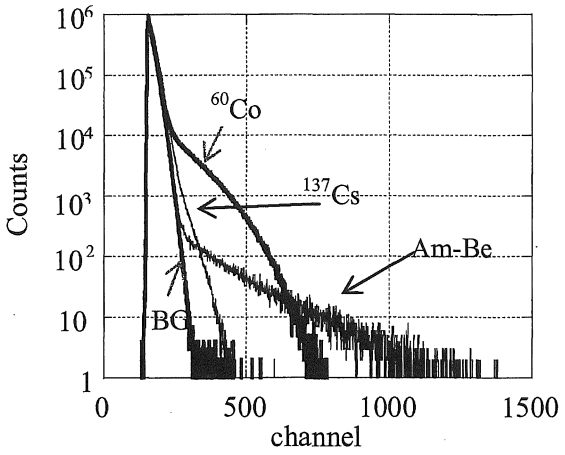

Fig.5 Response of liquid methane for Am-Be, ^{60}Co, ^{137}Cs

IV. Conclusion

The concentration of impurity of oxygen gas was obtained by analyzing a collected charge distribution of drifting electrons generated in the liquid methane by energy deposition of α particles. In this purity, pulse height distributions of a charge sensitive preamplifier were obtained for the detection of γ rays and neutrons emitted from ^{60}Co, ^{137}Cs and Am-Be sources located outside the cryostat of the liquid methane chamber.

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