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INITIAL RESULTS OF ARGON PURIFICATION IN THE LIQUID STATE*

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Summary

Argon, deliberately contaminated with air, has been purified in the liquid state by the use of molecular sieves to a level better than one part per billion (O_2 equivalent). This argon was maintained for a period exceeding 10 days with no detectable increase in impurity concentration. Twelve hours after transfer to a test detector, an attenuation length of 170 cm in a drift field of 1 kV/cm was measured for ionization electrons. These results are essential steps toward the realization of large liquid argon time projection chambers.

Introduction

For the past few years, we have pursued[1,2] the development of liquid time projection chambers (TPC)[3,4,5] due to our expectation that such detectors will open new possibilities for experimentation in physics. Potential applications of such detectors include experiments at reactors, at underground laboratories, and at medium energy and high energy accelerators.

Depending on the physics objective, the TPC medium may be Ar, Xe, CH_4, etc. We started this effort using Argon[1] due to its low cost, its ease of handling (inert gas), its ease of purification (low liquification temperature), as well as its possible uses.

With the liquid argon TPC, the applications of interest to us generally require massive detectors, e.g. reactor neutrino physics with detectors of order tons ($\bar{\nu}_e e^-$ elastic scattering, etc.) accelerator neutrino physics with detectors of order hundreds of tons ($\nu_\mu e^-$ elastic scattering, ν_μ oscillations, etc.), giant underground detectors of order thousands of tons (proton decay, solar neutrinos, etc.). Therefore, in addition to demonstrating long distance drifting of ionization electrons, we were driven to find a technique for argon purification at low cost which would be applicable for very large volumes. Standard techniques operate in the gas phase,[1,6,7] but the ideal technique would operate in the liquid phase.

The following describes results from initial tests of an argon purification system which operates in the liquid state. It is simple, inexpensive, applicable for very large volumes, and achieves impurity levels on the order of 1 ppb (O_2 equiv.). This corresponds to ionization electron attenuation lengths of about 150 cm at 1 kV/cm.

Liquid Argon Purifier/Storage System Tests

To investigate problems of long distance drifting of ionization electrons and track reconstruction in liquid argon, we constructed a test detector and a storage dewar.[2] The two dewars are coupled so that the high purity liquid argon can be rapidly transferred between them by means of a small pressure difference. The argon is maintained in the liquid state in each dewar by means of a liquid nitrogen reservoir pressurized to 40 psig.

Standard gas purification systems commonly employ cold molecular sieve to remove impurities.[1,7] To investigate whether molecular sieve is effective in removing impurities from argon in the liquid state, we modified the liquid argon storage dewar by the addition of molecular sieves, a convection heater, and an ionization detector (to monitor the liquid purity).

The cold walls of the LN_2 container are surrounded by vertical chimneys containing approximately 2.5 liters of type 4A molecular sieve in good thermal contact with the cold wall. The liquid argon is able to pass through these chimneys by convection. To enhance this convection, a heater constructed of 3 m of 32 gauge nichrome wire was installed. Approximately 30W could be applied to the heater before boiling occurred around the wire in the liquid argon. To monitor changes in the purity of the liquid argon, a gridded ionization chamber was installed in the argon volume beneath the LN_2 container. A change in the electronegative impurity level of the argon would cause a corresponding change in the collected charge produced by a ^{113}Sn internal conversion source located in the high voltage cathode. The drift distance (cathode to grid) was fixed at 3 cm, and the grid to anode distance, at 2 mm throughout this experiment. This purifier/storage system is shown in Figure 1.

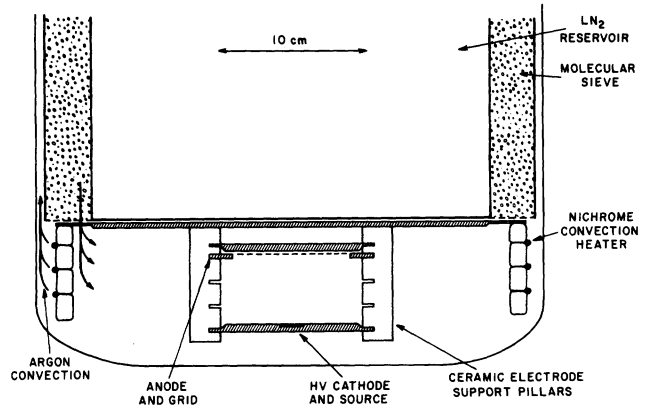

Fig. 1. Additions to the liquid argon purifier/storage system described in Reference 2. The heater enhances convection and the gridded ionization chamber monitors the argon purity.

Impurity Level Estimate

The ionization chamber in the storage dewar is

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used to estimate liquid argon purity. At a given drift field, $\vec{E}$, we measure the collected charge Q(E) from the ^{113}Sn source. A typical pulse height spectrum is shown in Figure 2. From a study of ionization recombination using ^{113}Sn, we have determined $Q_o(E)$, the charge escaping recombination in pure argon as a function of $\vec{E}$.[8] This is shown in Figure 3.

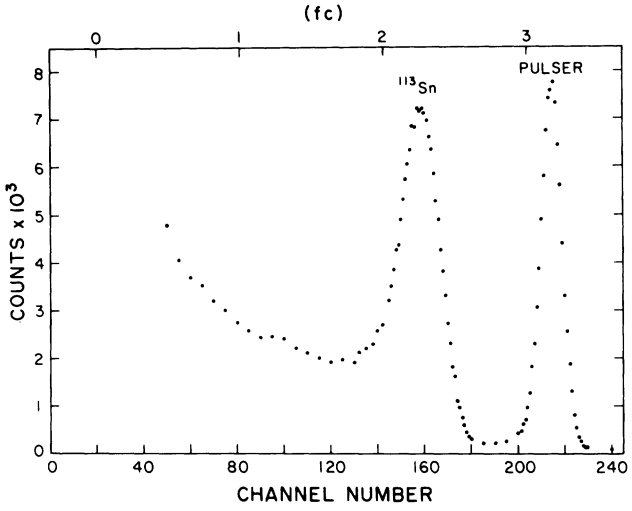

Fig. 2. A typical pulse height spectrum obtained with the ^{113}Sn internal conversion electron source at a drift field of 4.8 kV/cm. The pulser peak is at ∿ 3 fC.

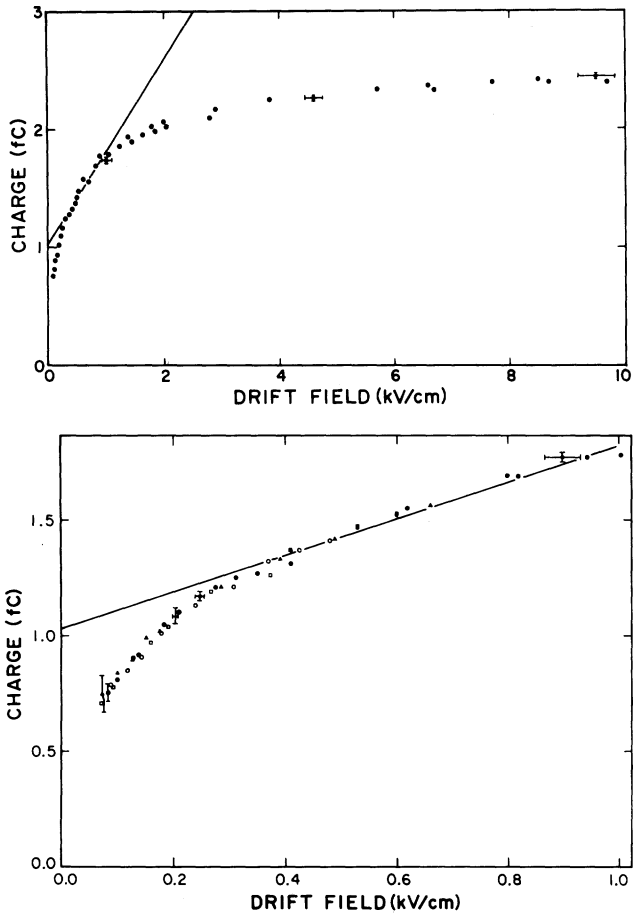

Fig. 3a & 3b. The free ionization electron charge, $Q_o(E)$, from a ^{113}Sn source is shown versus drift field, for high fields (a), and low fields (b). Impurity attachment effects have been removed.

For a given drift distance, x, and drift field, $\vec{E}$, the collected charge Q(E) is related to λ, the attenuation length of the drifting ionization electrons in liquid argon, i.e.

$$Q(E) = Q_o(E)e^{-x/\lambda} \quad (1)$$

This equation is used to determine λ. To convert λ to ρ, the impurity concentration, we assume the relationship,[9]

$$\lambda\rho = \alpha E, \quad (2)$$

where α is the attachment coefficient for a particular impurity (for O_2, α = 0.15 [cm^2-ppm-kV^{-1}]; for N_2, α is not well measured, but it is known to be substantially less electronegative than O_2[9]). We express impurities in terms of O_2 equivalent since the type of impurity is usually unknown.

This estimate of the impurity level should be used with caution since $Q(E)/Q_o(E)$ in the high purity liquid we used is typically close to one so that small uncertainties in Q(E) can contribute to large fractional variations in such a determination of ρ. The presence of large amounts of N_2 can enhance recombination without attaching electrons, and therefore reduce the free ionization charge.[10,11] However, because the most probable contaminant comes from external leaks, attachment to oxygen is expected to dominate enhanced recombination due to nitrogen.

Initial Purification and Liquification

The sieve material contained in the storage dewar was prepared by baking at 150°C for 12 hours under a vacuum of approximately 50 μ. Prior to condensing in the storage dewar, the argon was purified in the gaseous state by a process which normally provided purity levels better than 10 ppb (O_2 equiv.)[1] Approximately 16 liters of liquid argon were condensed as described over a period of about 36 hours.

This technique consistently produced liquid argon of purity at the level of 1 ppb (O_2 equiv.) which is significantly better than that obtained from the gas purification system alone. We attribute this improvement in the quality of the liquid to the presence of the sieve.

Purification in the Liquid State

Having established the liquid purity and its stability with time, we then contaminated it by introducing known amounts of air into the gas volume above the liquid. We enhanced the mixing of this air with the argon by inducing a refill cycle of the LN_2 reservoir which generated considerable agitation of the liquid argon. After contamination, the convection heater was turned on at 10W, and the collected charge, and hence purity, was recorded as a function of time.

Figures 4a and 4b show the effect of the addition of 310 and 350 ppb of air to the liquid argon by successive small and large injections respectively. It can be seen from the response of the ionization detector that the air is rapidly assimilated by the liquid argon, and that a recovery becomes apparent typically within an hour. After a period of approximately 24 hours, the purity has been restored to its original level of less than 1 ppb (O_2 equiv.).

After the early success with this liquid purification technique, we have not yet been able to reproduce these results where the liquid purity

improved to better than 1 ppb (O_2 equiv.). We believe that the present difficulty is a procedural one, i.e. incorrect preparation of the sieve prior to introducing the contaminated liquid. We are continuing to investigate this problem.

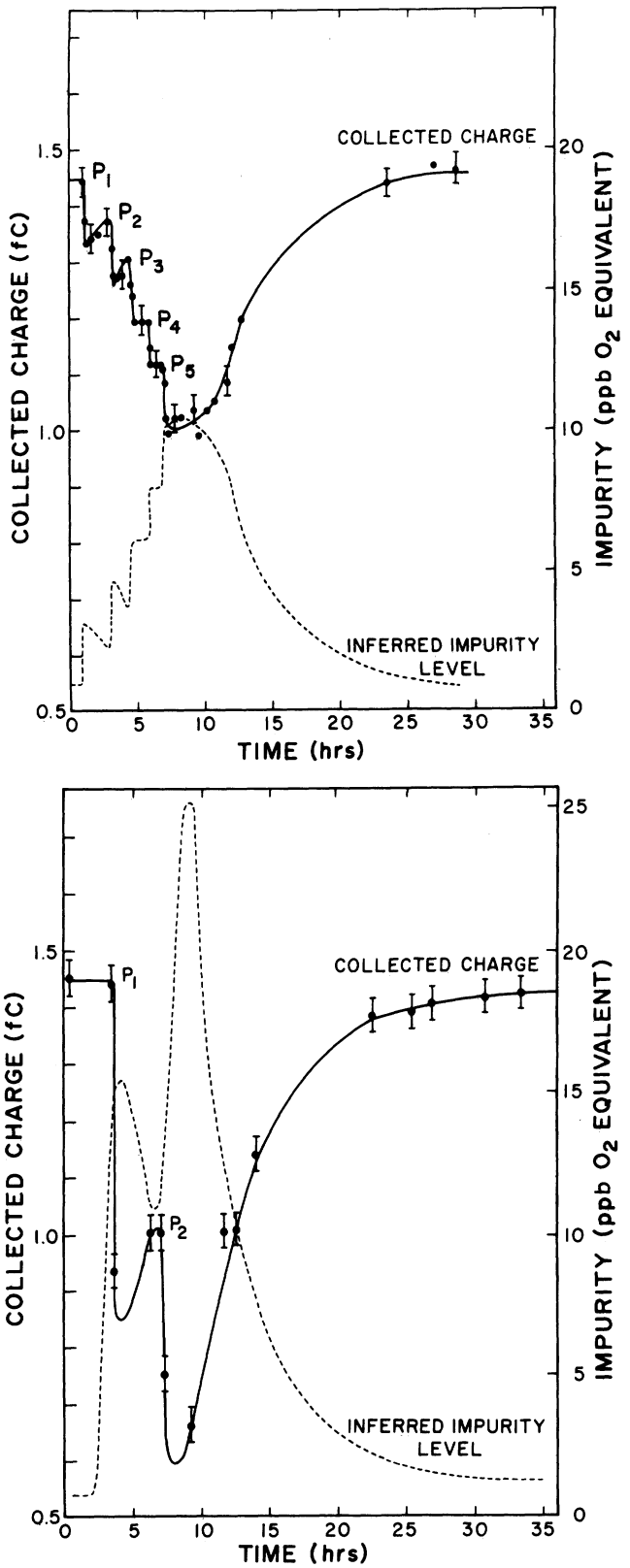

Fig. 4a & 4b. The collected charge with 5 injections $\sim$ 60 ppb each (a) and 2 injections $\sim$ 175 ppb each (b) of air is shown as a function of time. The injection points are marked by P's. The O_2 equivalent levels inferred from the collected charge is also shown.

Maintenance of Argon Purity

The collected charge in the ionization detector was observed over a period of 10 days during which it remained constant within errors ($\sim$ 1%). From the magnitude of this collected charge and its stability, we inferred that the liquid purity was always better than 1 ppb (O_2 equiv.). Therefore, the liquid deteriorated less than 1 ppb over 10 days in the storage dewar.

Measurements with the Test Detector

After the high level of purity had been restored, the liquid was transferred to the test detector which allows more detailed studies to be made. The ionization drift distance of this detector may be adjusted between 0.7 and 30 cm to allow an accurate measurement of the attenuation length, λ, and a separation of the two principle charge loss mechanisms, i.e. recombination, and attachment to impurities. This more accurate measurement of λ further substantiates the impurity level inferred in the storage dewar.

Attenuation Length Measurements

To measure the attenuation length, we record the collected charge from a ^{113}Sn internal conversion electron source as a function of the drift distance for a particular field.[1] The data collected at drift distances of 1, 3, 7 and 9 cm was used to deduce the attenuation length.[12] The dependence of λ upon E, approximately 12 hours after transfer to the detector, is shown in Figure 5. The attenuation length at 0.55 kV/cm (the field value also used in the storage dewar) was 95 cm, which infers an impurity level of 0.9 ppb (O_2 equiv.), assuming equation (2) and α = 0.15. The measured attenuation length of 170 cm at 1 kV/cm is the best reported to date and it is at least a factor of 4 better than reported by us last year.[2]

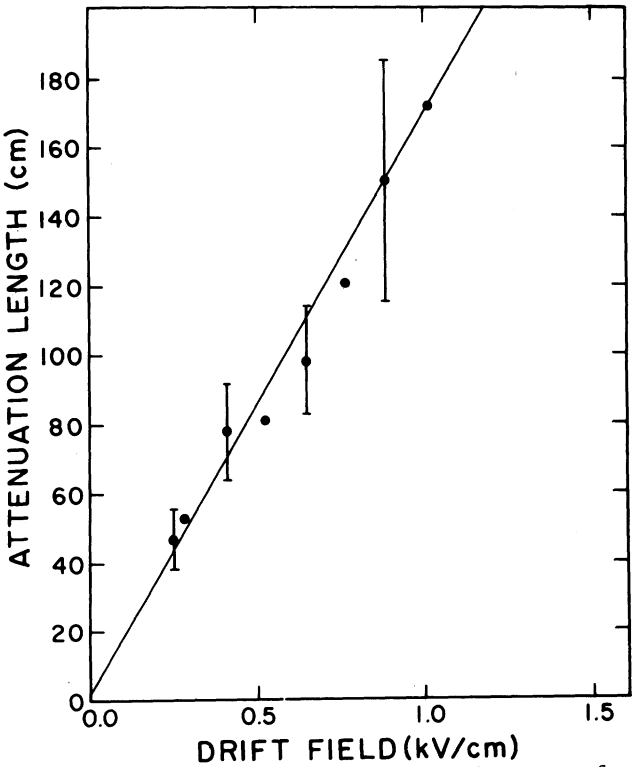

Fig. 5. The attenuation length, λ, is shown as a function of the drift field. These measurements were made about 12 hours after transfer to the test detector.

Figure 6 shows the measured attenuation length at 1 kV/cm as a function of time. The observed decrease in attenuation length as a function of time can be converted to an increase in the impurity level as a function of time. The rate of increase in the impurity appears to be a constant, as shown also in Figure 6, and it is equal to 0.6 ppb (O_2 equiv.) per day. This is about a factor of 5 worse than reported by us last year. Since the detector was not baked out prior to use, we believe that detector outgassing is the primary source of these impurities.

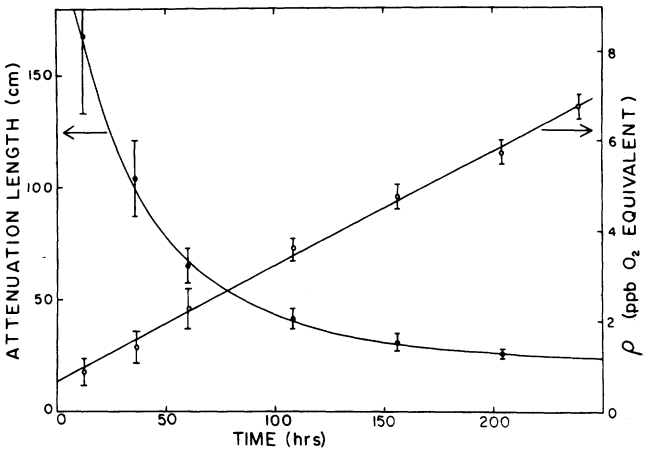

Fig. 6. The attenuation length and the corresponding impurity level is shown as a function of time after transfer of the liquid argon to the test detector.

High Voltage Discharge

Immediately after transfer of the pure liquid to the detector, we were unable to apply more than 8 kV to the cathode without evidence of electrical discharge. As time progressed and liquid impurity increased, the voltage where discharge begins also increased. The breakdown potential of liquid argon rises with increasing contamination. Extrapolation of existing data indicates that with impurities at the ppb level the breakdown potential is of the order of 100 kV/cm,[13] a value suspected to occur at a number of points within our detector.

Conclusions

These tests demonstrate that the quality of prepurified argon gas can be consistently improved to the 1 ppb (O_2 equiv.) level by condensing in the presence of molecular sieve, and that the major electronegative components of air can be removed from liquid argon by molecular sieves. However, our difficulties in subsequently reproducing liquid purification to the 1 ppb level shows that further study is required to understand the relevant parameters for its operation. It appears that with the extreme levels of purity attained, one must exercise care in detector design to avoid local high field regions where breakdown may occur.

The principal barrier to the realization of the liquid argon TPC has been the achievement and the maintenance of high liquid purity. This simple system has shown the capability to purify argon both in the gas and in the liquid state. Though much remains to be learned, we have shown that the basic purity requirements of large liquid argon TPC detectors can be met.

References and Footnotes

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