

# Argon Purification in the Liquid Phase

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### ABSTRACT

In the R&D programme for the ICARUS experiment, we have developed a practical procedure to purify liquid Argon in the liquid phase. Extreme purity is obtained, corresponding to electronegative impurity concentration below 0.1 ppb of O<sub>2</sub> equivalent. This corresponds to an electron lifetime in the range of several milliseconds equivalent to attenuation length of a few metres. The procedure has been tested up to flows of the order of 1000 litres of liquid per hour. The new technique makes it possible to operate very large amounts of ultrapure liquid Argon as foreseen for the multikiloton ICARUS experiment at Gran Sasso.

### 1. Introduction

The ICARUS collaboration proposed in 1985 [1] a multi-kiloton liquid Argon Time Projection Chamber to be run in the Gran Sasso Laboratory to search for rare underground phenomena ranging from proton decay to real time solar neutrino observations, as well as neutrino oscillations and relic supernovae neutrinos. The proposed detector is continually sensitive, self-triggering and provides three dimensional images of ionising events: it is an electronic bubble chamber.

Such a complex and ambitious project required an intensive R&D programme, that started four years ago. In particular the free electrons produced by ionisation in the liquid must be able to drift over distances longer than one metre, i.e. drift times of the order of several milliseconds, without substantial capture by electronegative impurities. This implies that the contamination of electronegative impurities must be kept around or below 0.1 ppb of Oxygen equivalent. We routinely employ simple techniques to obtain liquid Argon at this level of purity, as described in [2] where a three tons detector is described. The technique consists basically in flushing Argon gas, evaporated from liquid Argon, through Oxisorb[3] cartridges and molecular sieves. The gas is then liquefied back inside the detector. With the purifier installed for the 3 ton prototype we are able to purify 7000 litres of gas at NTP (equivalent to about 9 litres of liquid) per hour.

Purification directly in the liquid phase is the best way to substantially increase the purification rate and reduce the time necessary to fill a large detector. On the other hand it is not a priori guaranteed that the technique used for the gas works for the liquid, because it is not obvious that, due to the much higher density, the impurities have time to reach the active sites of the filter. To test this possibility we built the apparatus that will be described in the following sections. The technique also allows to make substantial savings in energy required to recondense the Argon after gas phase purification.

In Section 2 we describe the apparatus and in Section 3 the tests we made. Section 4 contains our conclusions.

### 2. Description of the apparatus.

A schematic view of the system is shown in the Fig. 1. The purifier consists of a 6 litres stainless steel cylinder, filled with a molecular sieve and an O<sub>2</sub> absorber. The molecular sieve is of the 5A type and has a volume of 2 litres, 1/3 of the total; it absorbs mostly water molecules. The absorption capacity depends on the temperature and partial pressure of the water vapour; it has been measured that 2 litres of molecular sieve can absorb more than 120

litres of water vapour before saturation; molecular sieves are also effective against other polar molecules. The O<sub>2</sub> absorber, 4 litres in volume, corresponding to 2/3 of the total, is the Oxisorb [3]; it consists of Chromium Oxide deposited on a SiO<sub>2</sub> support. In the apparatus described we can count on an absorption capacity of about 30 litres of O<sub>2</sub> at NTP. Notice that molecular sieve is located upstream in the filter, because Oxisorb is less effective in presence of water vapour.

We repeatedly measured, with the technique that will be described below, the free electron lifetime in the commercial liquid Argon [4] we employ. The corresponding contamination of electronegative impurities is calculated to be 0.5 ppm of O<sub>2</sub> equivalent. From the figures given above we can conclude that our test apparatus has a purifying capacity of the order of 70000 litres of liquid Argon, provided that the efficiency is 100% and that all the impurities in the system come from the supplied liquid.

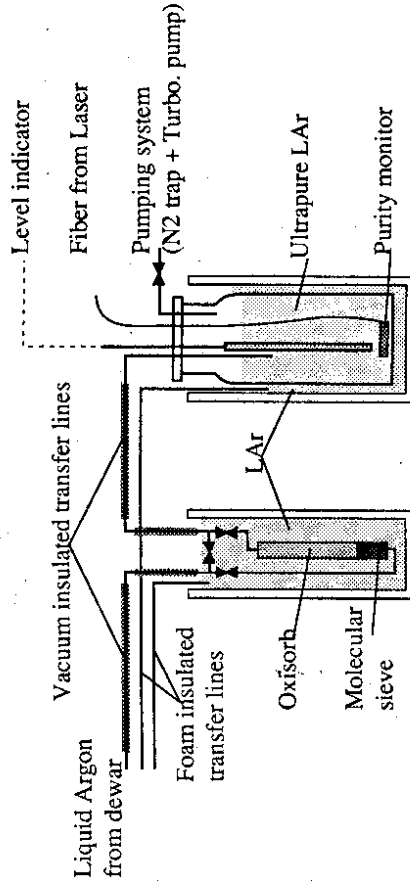


Fig.1. Schematic view of the liquid phase Argon purification system.

The purifier cylinder and the ultra pure Argon container, simulating the detector volume, are immersed into two separate liquid Argon baths; they are connected together and to the liquid Argon storage dewar through vacuum insulated cryogenic lines. The "detector" container hosts the purity monitor chamber on the bottom and a cylindrical capacitor (80 cm long) to measure the liquid level with the accuracy of 1 cm. The feedthrough for high voltage and signal cables and for the optical fibre bringing the laser light to the purity monitor are located on the upper flange. All the component of the cryogenic lines, purifier cylinder and ultra pure Argon container are made of stainless steel and are cleaned with the standard LEP ultra high vacuum procedure.

The measurement of the impurity concentration at levels much below the ppm is not trivial. Instead we measure the free electron lifetime, the quantity that is of direct interest for

our application, and infer the purity of the liquid. This purity monitor is an improved version of the one described in [5] and is schematically shown in Fig. 2. The photocathode is a stainless steel disk gold plated in the central region. The cathode is surrounded by a cylindrical piece, coated on the internal surface with Chromium Oxide by electrochemical deposition to absorb any stray diffused light. Electrons, photo-produced by a Laser pulse ( $\lambda = 266$  nm, 20 ns long), drift toward a first grid, called the window, driven by a field of 100V/cm (the cathode-window separation is 10 mm). The geometry allows the use of a moderately high field in this region, to have a high efficiency of injection of the photoelectrons in the liquid Argon conduction band, even for low drift fields. A fraction of the electrons crosses the window and enters a double gridded chamber. This fraction depends on the ratio of the field intensities after and before the window, and is, in practice, smaller than one. The consequent loss of electrons is not relevant, given the high value of the extracted charge.

The chamber has the same configuration as the one described in ref. [5]. The three grids are produced by photo-etching from stainless steel foils. They have a 2 mm square mesh made of 100  $\mu$ m diameter wires. The diameter of the window is 10 mm. The diameters of the cathode and anode grids are 30 mm. The drift distance between anode and cathode grids is 50 mm. The fields ratio across both grids is 2.5, well above the value of 1.9 needed to achieve complete transparency [6].

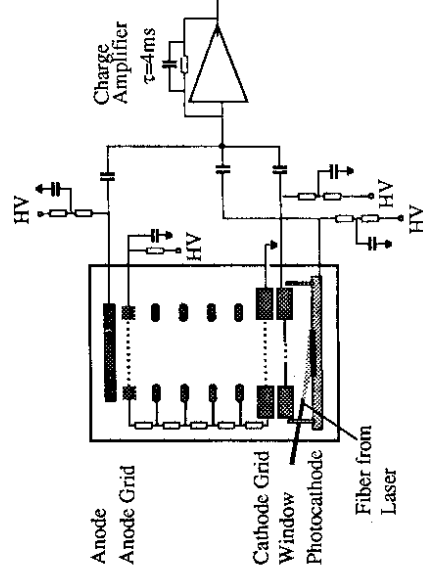


Fig.2. Purity monitor chamber.

The field in the drift region is made uniform by a series of stainless steel rings spaced by 10 mm. The voltage to the rings is given by a divider chain of 27 M $\Omega$  resistors.

absolute gain calibration. The integration constant of the amplifier is 4 ms. When the electrons drift between cathode and window, the amplifier receives two equal and opposite currents that cancel each other. When the electrons drift between the window and the cathode grid a positive current flows in the amplifier; as a consequence its charge output increases linearly reaching a value  $Q_c$ , the charge leaving the window. When the electrons drift between the grids, no current flows in the amplifier (at least if the grids screen perfectly). Finally when the electrons drift between the anode grid and the anode, the current is negative. The output charge signal has a negative step, whose height is the charge  $Q_a$  reaching the anode. An example of the charge output, to be discussed later, is shown in Fig. 3.

If no charge is lost in the drift volume (zero impurity concentration), obviously  $Q_a = Q_c$ ; if some charge is lost, corresponding to a lifetime  $\tau$ , the ratio of the charges is simply  $Q_d/Q_c = \exp(-T_d/\tau)$ , where  $T_d$  is the drift time. In conclusion the measurement of the ratio of the two charges and of  $T_d$  gives directly the lifetime. When the lifetime becomes much longer than the drift time, the charge ratio becomes equal to one within the errors and the monitor achieves its sensitivity limit. This limit is of course higher at lower values of the drift field (i.e. at higher values of  $T_d$ ). In practice we work at 40V/cm, corresponding to  $T_d = 282 \mu\text{s}$ , achieving a sensitivity of several milliseconds.

### 3. Testing liquid phase purification.

All the mechanical parts in the cryogenic circuit from the purifier to the container, including the container itself, have been degreased with demineralised water, baked at 150°C and evacuated at  $10^{-7}$  mbar or better.

The first test consisted in purifying the Argon in the gaseous phase, at a flow value of 14 equivalent liquid litres per hour, to check whether we obtained the same purity as with our usual procedure. The measured electron lifetime was unchanged above our sensitivity limit that is higher than 3-4 ms.

We then performed a series of purifications of Argon in liquid phase at different flows; the flux was regulated by choosing the appropriate pressure drop in the circuit. Each time we purified a volume of 130 litres. The flow values of our tests were: 125 litres of liquid per hour (l/h), 400 l/h, 470 l/h, 800 l/h.

Fig. 3 shows, as an example, the output pulse from the charge amplifier for the filling at 470 l/h. Interference from the firing of the Laser is visible as a positive spike 10  $\mu\text{s}$  before the signal and some oscillations at the beginning of the signal. The non-perfect flatness of the bottom of the signal is due to imperfect screening of the grids but does not interfere with the measurement.

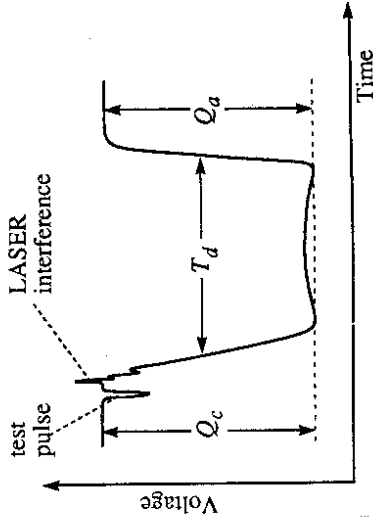


Fig.3. Charge signal from purity monitor for ultra pure liquid Argon purified at 470 l/h.

The figure shows that  $Q_a$  is equal to  $Q_c$  within the accuracy of the measurement; the drift time is  $T_d = 282 \mu\text{s}$ . The corresponding lifetime is higher than 3-4 ms.

For all the fillings we performed we found similar results: the electron lifetime was always above 3-4 ms. The maximum flux tested is essentially determined by the pressure difference between the liquid Argon storage dewar and the detector. Two or more similar cylinders in parallel, for example, can be used to increase the flux.

### 4. Conclusions.

We have shown that ultra pure liquid Argon can be obtained by direct purification of the liquid. The final purity obtained corresponds to an electronegative impurity concentration below 0.1 ppb  $\text{O}_2$  equivalent, equal to that obtained with similar procedures purifying the gas phase. The corresponding electron lifetime is larger than 4 ms. The flows are almost three orders of magnitude (the ratio of the densities) higher. As a consequence, the problem of filling a large scale detector is much simplified. The ICARUS experiment plans to build 5000 ton detector modules. One such module can be filled with ultrapure liquid Argon in a few weeks employing a small (5-10) number of liquid phase purification systems in parallel, built on the principles described here. A shorter filling time reduces also to a minimum the time spent by detector components at relatively high temperatures and not in contact with liquid; this implies that outgassing and contamination are diminished.

### References.

- [1] ICARUS collaboration, "ICARUS a proposal for the Gran Sasso Laboratory", INFN/AE-85/7 Frascati 1985.
- [2] P. Cennini et al. A three tons Liquid Argon Time Projection Chamber. DFPD 93/EP05. Padova Univ., submitted to Nuclear Instruments and Methods.
- [3] Oxisorb is a trade mark of Messer Griesheim GmbH
- [4] Supplied by Linde Gaz Industries (France).
- [5] A. Bettini et al., Nucl. Instr. and Meth. **A305** (1991) 177.
- [6] O. Bunenam, T.E. Cranshaw and A. Harvey, Can. J. Res. **27A** (1949) 191.