Towards a liquid Argon TPC without evacuation: filling of a 6 m$^3$ vessel with argon gas from air to ppm impurities concentration through flushing

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Abstract. In this paper we present a successful experimental test of filling a volume of 6 m$^3$ with argon gas, starting from normal ambient air and reducing the impurities content down to few parts per million (ppm) oxygen equivalent. This level of contamination was directly monitored measuring the slow component of the scintillation light of the Ar gas, which is sensitive to all sources of impurities affecting directly the argon scintillation.

1. Introduction
GLACIER (Giant Liquid Argon Charge Imaging ExpeRiment) is a proposed very large LAr TPC with a well defined conceptual design [1, 2]. The proposed cryostat is a single module cryo-tank based on industrial liquefied natural gas (LNG) technology, with a cylindrical shape giving an optimal surface-to-volume ratio. The detector design is meant to be highly scalable, up to a total mass of at least 100 kton.

One severe key technical challenge of the GLACIER design is to achieve long drift paths of free ionization electrons in liquid argon over a very long distance, possibly as large as 20 meters. The GLACIER concept therefore relies on a double phase operation with a readout with charge amplification to compensate for the S/N loss due to diffusion and remaining charge attenuation from impurity attachment during the long drift path, but in addition requires very special care to achieve and maintain very high purity in the liquid argon, at the level of $\lesssim 20$ ppt oxygen equivalent [3], and a very high voltage to create the drift field to quickly drift the electron cloud across the liquid argon volume [4].

Argon purification systems based on oxygen reactants and molecular sieves, commercially available, have been studied since many years as part of the R&D for liquid Argon TPCs [5, 6], and are known to be very effective in reducing the oxygen contamination and also water, carbon oxides, fluorates elements or hydrocarbons to very small levels. During the ICARUS 50L exposure to the WANF neutrino beam at CERN [7], a free electron lifetime of 15 ms, corresponding to 20 ppt oxygen equivalent, was achieved using such standard (gas) purification methods.
Figure 1. Schematics of the apparatus, completed with recirculation and gas analyzing systems.

However, like the ICARUS 50L, all LAr TPCs operated up to now were housed in a vacuum vessel, and ultra-high vacuum evacuation was always performed prior to cooling and filling with liquid argon. This will not be possible for a LNG tank, because this kind of vessel, although a time tested solution, cannot withstand under- or over-pressures above 50-100 mbar, so evacuation is impossible. As of today, the experimental demonstration that high liquid argon purity can be achieved and maintained over long periods of operation in an LNG tank, is lacking. As a first step towards this milestone, we started studying new methods and scalable techniques to manage non-evacuable vessels.

2. Avoiding the ultra-high vacuum phase

The vacuum phase covers at least three important separate functions in providing conditions for a low level of impurities in LAr:

(i) it removes air from the vessel prior to cooling and filling with LAr;
(ii) it removes outgassing (water and other elements) from the detector materials and the vessel walls (baking is also sometimes performed);
(iii) it verifies the integrity (tightness) of the system itself.

In the present work, we focused on the item (i). Items (ii) and (iii) will be studied later. Concerning (i), it should be pointed out that commercially available (high grade) bulk liquid argon, which is handled, transported and stored without using high vacuum vessels, has an oxygen content of about 1 ppm, which is an acceptable starting point for further purification in the liquid phase. Therefore it looks very reasonable to expect that at least the “remove air” function can be easily achieved without vacuum. Prior to our test, we therefore anticipated that the air in a vessel could be efficiently displaced by a flow of Ar gas, and we have experimentally tested this technique for reducing the air concentration using a 6 m$^3$ vessel.

In our test, we first proved that oxygen can be effectively reduced from its air concentration (20%) to less than 0.1% with oxygen detectors. However, in order to be sensitive to all potential
contaminants that affect drift electron properties, we simultaneously instrumented our vessel with DUV photosensors (coated PMTs) to directly measure the argon scintillation light produced by embarked radioactive sources. Indeed, several components are observed in the scintillation of gaseous argon. The mean lifetime of the slowest (triplet) component (about 3 µs) is very sensitive to traces of impurities and can be very effectively used to monitor the impurities [8, 9] that affect the drift properties of electrons. See section 3.2.

To our knowledge, this is the first time this sort of experiment is conducted in a vessel of this size at such level of contamination. A test of argon gas flushing was previously reported in Ref. [10], however, the vessel was smaller than in our case and only the oxygen content was monitored. In our work, we therefore measured for the first time the level of several impurities by a direct observation of the argon scintillation light.

More experimental work is ongoing, in particular to address the outgassing and leak checking aspects, and results are expected in the near future.

3. Experimental setup
3.1. Ar vessel
A schematic of the apparatus, completed with a recirculation and gas analyzing systems, is shown in Fig.1. A 6 m³ vessel, previously used as a liquid argon dewar for testing of LAr/U calorimeters, was refurbished and made available for this test. The vessel is a double walled dewar, to allow for vacuum insulation in case of cryogenic operation. The walls of the inner vessel are stainless steel, while the top flange, 7 cm thick, is made out of aluminum, as the outer vessel. The diameter of the inner vessel is 200 cm and the total height of the dewar is 235 cm. On the top flange there are 12 KF sealed nipples for feedthroughs. The top flange is sealed with an o-ring. The tightness of the vessel has been measured through pressure decay leak testing, which allowed to determine a leak rate of 0.15 mbar lt/sec (which compares to a leak rate of ~10⁻⁶ mbar lt/sec characteristic of high vacuum vessels). This relatively large leak rate was deemed acceptable for our goals, since it corresponds - all things equal but a reversed pressure gradient - to a flow of 20 ppm of O₂ per hour; considering that the vessel is operated with an overpressure of ~ 100 mbar, the actual inflow of O₂ from air is likely to be rather negligible. The Ar gas transfer line provided a maximum flow of about 200 lt/min. The gas line was connected
Figure 3. The dependence of $\tau_2$ from the concentration of different impurities in Ar gas. Data from [9]. A fit with a Birks’ law-type function to the O$_2$ data is superimposed.

to a 35 mm diameter stainless steel tube which went all the way to the bottom of the vessel. The Ar gas was exhausted through a flow meter, which could measure a maximum flow of 100 lt/min.

3.2. Detection of impurity traces with PMTs
During operation, the vessel was equipped with two percent level O$_2$ monitors$^1$, which are sensitive to a minimum concentration of 0.1% O$_2$. The oxygen sensors were left hanging from the top flange, one at a depth of 30 cm and the second one 90 cm below.

Three photomultiplier tubes (PMT) were deployed inside the vessel to monitor the DUV argon scintillation light, at a distance of 40, 110 and 180 cm from the top flange. The PMTs are 2” ETL 9831KB, each housed in a cylindrical stainless steel holder with a 40 kBq $^{241}$Am ($\alpha$ emitter) source placed in front of the PMT, at a distance of 10 cm from the photocathode. There are large circular apertures on the sides of the holder, to allow for efficient gas circulation (Fig.2). Once inside the holder, a PMT sees a negligible amount of light from outside the holder itself, so that we could monitor the scintillation light of the argon at a given position. The inside of the PMT holder was lined with 3M foil specular reflector coated with 1 mg/cm$^2$ of tetraphenylbutadiene (TPB), rendering the reflection from the surface 90% diffuse [11]. In addition the PMT windows were coated with 0.05 mg/cm$^2$ TPB. TPB acts as a wavelength shifter for the DUV scintillation light of argon to approximately 430 nm (blue), within the high quantum efficiency range of the PMT [11]. The signal from each PMT was digitized at a sampling rate of 1 GS/s, using an Acqiris DP1400 digitizer.

$^1$ OXY-SEN model, from Alpha Omega Instruments.
Figure 4. PMT traces for alpha particles from an Am source. Top: an event at the beginning of the run, $\tau_2$ consistent with zero. Bottom: an event taken near the end of the run, with $\tau_2$ exceeding 1200 ns.

The scintillation light of pure argon gas has a fast and a slow component, with decay times $\tau_1$ shorter than 10 ns and $\tau_2$ of about 3 $\mu$s [12, 13]. The fast component is rather independent of the purity of the Ar, while the decay time of the slow component is sensitive to the concentration of impurities. In particular, very clean Ar shows a decay time exceeding 3 $\mu$s, while for $O_2$ concentration larger than 0.1% the decay time is 100 ns or less. See for example Fig. 3 (data from [9]), which is directly comparable to the measurements presented here since it was obtained with the same hardware and analysis procedure (in a smaller cell).

The quenching effect of impurities on the slow component of the scintillation light can be roughly described by

$$\tau_2[\text{ns}] = \frac{\tau'}{1 + k\rho[\text{ppm}]}$$

where $\tau'$ is the asymptotic value of $\tau_2$ for zero impurities, $\rho[\text{ppm}]$ is the concentration of impurities in ppm and $k$ is a constant; this function is reminiscent of the Birks’ law that describes the quenched light yield in scintillators for an increased $dE/dx$. For $O_2$, $\tau' = 2878$ ns and $k = 0.24$. This method is more sensitive for $\tau_2$ in the 500 to 2500 ns range (i.e. 0.5 to 10 ppm of $O_2$), while for smaller $\tau_2$ the variation is rather modest over a couple of orders of magnitude change in $O_2$ concentration.

For each measurement of the $\tau_2$ 10,000 events are acquired, a procedure which typically takes few seconds. Each waveform is fitted offline, to determine the value of $\tau_2$. The average value for
the 10,000 events is taken as the measured $\tau_2$ at a given time. Traces taken at the beginning and at the end of the run are shown in Fig.4.

4. Results
The 6 m$^3$ vessel was flushed with argon gas, not continuously, over more than a week. We started using a bottle (10 m$^3$ stp) of Ar57 (impurities less than 3 ppm) for a preliminary flushing which reduced the O$_2$ concentration from 20% to 0.5%, measured with one OXY-SEN sensor placed on the vessel exhaust. After 48 hrs without flushing any argon and the vessel with 150 mbar overpressure, the O$_2$ measured on the exhaust line actually improved, since purer argon had time to diffuse from the bottom to the top of the vessel. A flange at the top was then opened to insert the two OXY-SEN sensors inside the “clean” argon volume. Flushing with Ar57 was then restarted - as can be seen in Fig.5. Part of the data from the OXY-SEN readings are collected in Tab.1; the two sensors at different depths show very clearly the pure Ar gas acting as a piston, removing the residual air starting from the bottom to the top. The Ar57 flow was approximately constant at 80 lt/min, limited by the flowmeter on the exhaust line; this is equivalent to one volume change per 75 min. The vessel was always flushed with Ar57, except for the last 1.7 volume changes (starting at [02/25 14:00]) when Ar60 (impurities less than 1 ppm) was used, because the purification rate seemed to decrease, possibly due to $\sim$3 ppm of O$_2$ in the input gas.

Fig.6 and Fig.7 summarize the main results versus acquisition time. When the sensitivity limit of the OXY-SEN sensors was reached (0.1%), data have been taken continuously using the three PMTs. The decay time of the slow component of the Ar scintillation light ($\tau_2$) improves continuously due to the removal of impurities. The PMT near the bottom of the vessel (and the Ar inlet) consistently shows the largest $\tau_2$, and shows the reduction in of contamination earlier than the other PMTs, as expected from the Ar gas acting as a piston. The PMTs at the top
and in the middle show a similar $\tau_2$, and the top one shows larger variations, correlated with sudden changes in the flow rate and pressure, which is measured at the top flange. The Ar flow near the top flange is expected to be quite turbulent and non-uniform, since the outlet is - in practice - point-like when compared to the size of the flange itself. When the flushing stops, $\tau_2$ decreases by an amount roughly equivalent to the introduction of 2ppm of $O_2$, possibly coming from outgassing and/or leaks to the outside at a rate of 0.15 ppm per hour. The $O_2$ equivalent concentration at the end of the flushing procedure at the location of the bottom PMT is lower than 4 ppm, while the average of the 3 PMTs is slightly larger than 4 ppm. The $O_2$ equivalent concentration [ppm] vs time is shown in Fig.7 for $\tau_2 > 500$ ns, i.e. the optimal range for this technique. The $O_2$ concentration drops of about a factor of two after 5 hrs of flushing, i.e. 24,000 lt of Ar gas stp or 4 volume changes.

Table 1. $O_2$ concentrations as measured with OXY-SEN monitors (see text).

| Time&Date       | Exhaust $O_2$ [%] | Top $O_2$ [%] | Bottom $O_2$ [%] |
|-----------------|------------------|--------------|-----------------|
| 2010-02-19 15:30 | 19.1             | -            | -               |
| 2010-02-19 16:18 | 9.1              | -            | -               |
| 2010-02-19 16:28 | 6.9              | -            | -               |
| 2010-02-19 16:39 | 5.2              | -            | -               |
| 2010-02-19 16:48 | 4.2              | -            | -               |
| 2010-02-19 16:57 | 3.5              | -            | -               |
| 2010-02-19 17:06 | 3.3              | -            | -               |
| 2010-02-19 17:12 | 2.9              | -            | -               |
| 2010-02-20 10:29 | 1.4              | -            | -               |
| 2010-02-23 10:24 | -                | 0.3          | 0.3             |
| 2010-02-23 10:30 | -                | 0.3          | 0.3             |
| 2010-02-23 10:39 | -                | 0.3          | 0.3             |
| 2010-02-23 10:55 | -                | 0.3          | 0.3             |
| 2010-02-23 11:05 | -                | 0.25         | 0.3             |
| 2010-02-23 11:20 | -                | 0.25         | 0.3             |
| 2010-02-23 11:27 | -                | 0.2          | 0.3             |
| 2010-02-23 12:01 | -                | 0.2          | 0.3             |
| 2010-02-23 12:21 | -                | 0.2          | 0.25            |
| 2010-02-23 12:38 | -                | 0.2          | 0.25            |
| 2010-02-23 12:54 | -                | 0.2          | 0.2             |
| 2010-02-23 13:35 | -                | 0.15         | 0.2             |
| 2010-02-23 13:44 | -                | 0.1          | 0.2             |
| 2010-02-23 16:09 | -                | 0.1          | 0.15            |
| 2010-02-23 16:21 | -                | 0.1          | 0.1             |
| 2010-02-23 18:34 | -                | 0.1          | 0.1             |

5. Conclusions
The main results of this test is the purging of a 6 m$^3$ vessel from air to a residual contamination of few ppm of $O_2$ equivalent in pure Ar gas. The impurity concentration in argon has been
Figure 6. Fitted time constant of the slow component of the Ar scintillation light vs. time for three PMTs at different depths. Data taken during Ar flushing.

measured down to the few ppm range using a simple system, based on the quenching of the Ar scintillation due to impurities. The piston effect of argon displacing impurities has been seen both for percent level contamination and ppm level contamination. The final O\(_2\) equivalent concentration, after ten volume changes, was \(\sim 4\) ppm, which corresponds to a reduction of the O\(_2\) of about five orders of magnitude, faster than a simple exponential trend. The experimental setup was remarkably simple, and the reduction in concentration of impurities in the vessel proved to be rather insensitive to interruptions in the flushing procedure. This is an experimental demonstration that impurities in argon can be reduced easily and efficiently to ppm level without vacuum pumping.

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Figure 7. Same as Fig.6, converted in ppm of $O_2$ equivalent in argon.

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