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The ICARUS T600 liquid argon purification system

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Abstract

The ICARUS detector is a liquid argon time projection chamber with unique features that make it an ideal device to be used for several particle physics applications. After years of R&D activities, the ICARUS Collaboration proposed the construction of the T600 in strict partnership with industry to guarantee the necessary and viable scaling-up of the technology from prototypal dimensions to sized plants in order to study neutrino oscillations and matter stability in an effective way. The T600 represents the largest LAr detector (760 t LAr mass) ever realized. It was installed and successfully operated for 3 years inside the underground Gran Sasso Laboratory from May 2010 to June 2013. One of the most important issues for the success of the detector technology is the liquid argon purity. Purity requirements become stronger and stronger with the increase of the detector dimensions: for a plant of the ICARUS T600 size it is necessary to keep the residual electronegative impurity content to a level of the order of 0.1 parts per billion or better all over the argon volume during the whole detector run, thus allowing the ionization tracks, created by interacting particles inside LAr, to be transported with only slight attenuation along the drift path. In this paper we present the ICARUS T600 purification plant installed at Gran Sasso Laboratory and describe in details the solutions adopted for the LAr re-circulation and purification systems that permitted to reach impressive results in terms of LAr purity thus representing a milestone for any project involving LAr and developments at higher LAr mass scale.

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Keywords: LAr purification; ultra pure noble liquids; LAr TPC

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1. Introduction

LAr and other noble liquids (LXe, LKr) have unique characteristics to be used as detector media in particle physics with various applications going from rare event searches (dark matter, double beta decay, neutrino physics and proton decay) to high-energy physics. For this reason there is an increasing interest in developing large noble liquid detectors with high sensitivity and performance. All these detectors are based on ionization and/or scintillation light signals produced by interacting particles in LAr. Both processes are affected by impurity concentration in LAr depending on the detector characteristics and dimensions. Ionization electrons can be captured by residual compounds with high electron affinity such as O_2, H_2O and CO_2; the same for the scintillation (UV photons) production and transport that is also affected by residual nitrogen contamination, Acciarri et al. (2010).

These compounds are typically present in traces in the commercial noble liquids, for this reason one of the most important issues for the success of the noble liquid detector technology are the liquid purity and the purification methods. Purity requirements become stronger and stronger with the increase of the detector mass.

The ICARUS (Imaging Cosmic and rare Underground Signals) detector is a liquid argon Time Projection Chamber (LAr TPC), Rubbia (1977). This detection techniques guarantees high quality and granularity imaging of the interaction events similar to the bubble chambers, with the further advantages of being fully electronic, continuously sensitive and self-triggering; it also permits high resolution calorimetric measurements and is potentially scalable to huge masses that are necessary to perform rare events studies in a reasonable time scale. The operation principle is based on the fact that in highly purified liquid argon ionization tracks created by interacting particles inside LAr can be transported, practically unperturbed, over macroscopic distances (meters) by means of a uniform electric field up to a multi-wire anodic structure placed at the end of the drift path. Signals induced on wires by the drifting electrons are continuously sensed and recorded to provide three simultaneous projective views of the event, allowing a precise geometrical tridimensional reconstruction of the particle trajectory. Timing and triggering issues are performed by means of LAr scintillation light detection, allowing for absolute spatial reconstruction that is crucial for charge attenuation evaluation and high-resolution calorimetric measurements.

After years of R&D activities, Benetti (1993) and Cennini (1994), the ICARUS Collaboration proposed the construction of the T600 plant, ICARUS Collaboration (1995) that is the largest liquid argon detector ever realized with its 760 t LAr mass. It was firstly tested on surface and then installed in the deep underground Gran Sasso Laboratory to study neutrino oscillations and matter stability. Strict partnership with industry was necessary to perform an effective and viable scaling-up of the technology from prototypal dimensions to a sized plant.

For a plant of the size of the ICARUS T600, liquid argon purity is crucial for the TPC performance: residual compounds with high electron affinity have to be kept as low as 0.1 parts per billion (ppb) of oxygen equivalent, (corresponding to 3 ms free electron lifetime[†]) or even better, all over the argon volume during the whole detector run. For the scintillation photon production and transport, purity requirement could be less stringent: for instance O_2 concentrations has to be kept below 0.5 ppm and N_2 within few ppms.

The solutions developed by the ICARUS Collaboration over about 20 years of R&D to cope with argon purity specifications are essentially based on the following ingredients:

- Use of commercial argon filters operated directly in liquid phase: Hydrosorb^{TM} for water removal and Oxysorb^{TM} for oxygen removal. These filters capture CO_2 for thermal effects, while N_2 is not trapped;
- Adoption of ultra high vacuum components (such as standard UHV CF flanges) and techniques for detector construction;
- Precise commissioning procedures involving an initial vacuum phase to remove air and proper outgas all the internal surfaces followed by a fast cryostat LN_2 cooling phase and a minimized LAr filling phase with purified argon;
- Continuous gas and liquid argon re-circulation systems developed, both involving argon purification in liquid phase, to attain and maintain the required purity level.

[†] Free electron lifetime is the average capture time of a free ionization electron by an electronegative impurity in LAr: τ_{ele} [ms] ~ 0.3/N [ppb], where the N is the concentration of Oxygen equivalent impurities.

This approach brought to reiterate successful results; we cite for instance the value of 1.8 ms electron lifetime after 2 months operation in the 2001 surface test run on half of the T600 (275 m^3), Amerio et al. (2004). On a much smaller scale (120 liters prototype with 38 kg active mass) the remarkable electron lifetime of 21 ms was reached, Baibussinov et al. (2010).

2. The ICARUS T600 cryostat and argon purification plant

The ICARUS T600 cryostat was designed in strict partnership with Air Liquide Italia Company. The T600 test version was firstly operated on surface in 2001 to validate the implemented solutions for the cryogenic plant and verify detector performance, Amerio et al. (2004). Then the cryogenic plant was improved mainly for safety aspects connected with the underground operation and finally installed inside the Gran Sasso Laboratory Vignoli et al. (2006).

The T600 cryostat is composed by two identical adjacent parallelepiped aluminum LAr containers, the "East" and "West" modules, with external dimensions 3.9 w x 4.2 h x 19.9 l m^3 and internal volume of about 275 m^3.

Each module houses two TPCs separated by a common vertical cathode. The maximum drift length is 1.5 m. The anodic structure is composed by three parallel wire planes placed along three different directions. The T600 global number of sensible wires is 53248.

The two modules are independent from the point of view of argon containment and purification plants, while they have a common cooling shield and insulation vessel. The cooling shield surrounds the module pair and intercepts residual heat loss through insulation allowing maintaining LAr thermal uniformity within 1 K in the LAr volume. The N_2 shield is operated in forced way by means of one Barber Nichols pump (BNCP-51B-000 model).

Each module is equipped with two GAr and one LAr re-circulation systems.

The aim of the GAr re-circulation system is to purify the gas argon phase present on the cryostat ceiling to prevent diffusion of impurities from gas to liquid phase. Gas is dirtier with respect to the liquid, as it is in contact with warm outgassing materials (such as the signal plastic cables) and it could be polluted by possible small leaks due to the high number of joints on the overall 80 chimneys located on the cryostat top and hosting the read-out cables and the feed-through flanges.

The operation of the GAr re-circulation and purification system is based on the natural circulation of the fluid due to pressure differences: GAr collected from the top chimneys re-condense in a dedicated LN_2 heat exchanger and then drops back into the main cryostat just under LAr surface after passing through a LN_2 cooled Oxysorb^{TM} filter. The condenser is fed with liquid nitrogen by means of nitrogen forced circulation in order to keep it at the temperature required for an efficient re-condensation of the argon gas. Each GAr re-circulation unit collects gas from a row of 20 chimneys and guarantees a maximum rate of 25 Nm^3/h.

GAr re-circulation system is extremely useful during the Ar filling phase, because it allows recycling dirty warm gas as outgas phase is still on. During normal operation GAr re-circulation system results to behave as a precise pressure stabilizer and for this reason it has to be continuously active.

Forced re-circulation in liquid phase is instead designed to massively purify LAr just after cryostat filling to reach and maintain the highest purity level. In addition, it is suited for a fast restoration of the argon purity in case of accidental pollution during the detector run, as it guarantees a high re-circulation rate. It was dimensioned to guarantee full module volume re-circulation in about six days.

The key element of the liquid argon re-circulation system is the LAr transfer pump, which affects the plant design and performance. At the beginning of the ICARUS T600 project, the available solutions for pump choice were restricted to the following industrial products: submerged pumps or capsulated-motor cryogenic pumps.

In collaboration with Air Liquide Italia Service Company we selected the submerged pumps as highly preferred for the argon purity goal. In this kind of pump bearings are directly lubricated by means of a LAr flux. Capsulated-motor pumps have higher throughput than the submerged pumps but they have grease-lubricated bearings and result to be less safe for the high risk of argon contamination as pump is located in the gaseous phase not tightly separated from the liquid one.

During the first T600 module technical run in Pavia in 2001 it was implemented and successfully tested a CRYOSTAR Company submerged pump that allowed steady increase of the electron lifetime reaching the value of 1.8 ms after about 2 months of reliable operation.

During the T600 upgrade for the LNGS run we selected the ACD CRYO Company AC-32 model pump as the former pump was out of market.

The realized system is shown in Figure 1. Each module LAr re-circulation unit ensures LAr purification by forced liquid circulation through a battery of four Oxysorb/Hydrosorb^{TM} filter cartridges (connected in parallel), which are the same used during the initial cryostat filling. LAr is extracted at a medium height on one side of the vessel, purified and injected back at the opposite longitudinal side (20 m apart) uniformly distributing it at a lower height, close to the module bottom. The LAr submerged pump is located in an independent vessel that is connected to the main cryostat both in liquid and gas phase. The pump vessel has vacuum insulation with liquid nitrogen shield. The battery of filters is located in a separate vacuum vessel; cartridges are nitrogen cooled. The maximum re-circulation rate resulted to be about 1.5 m^3/h/unit, as result of the pump throughput and the filter battery impedance, thus compatible with specifications.

Liquid nitrogen circulation to cool the pump vessel, cartridges and all the Ar transfer lines is guaranteed by one of the three installed Barber Nichols (BNCP-51B-000 model) centrifugal pumps. LN_2 pump is fed by gravity with LN_2 from a phase separator vessel connected to the nitrogen storage tanks located above the cryostat.

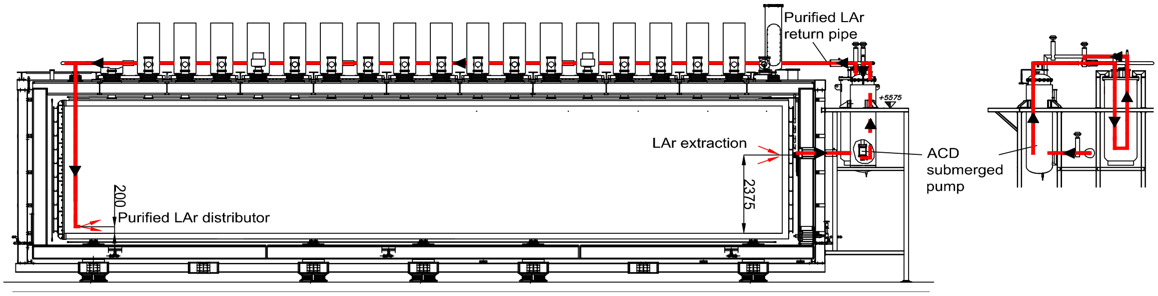

Fig. 1 Side and frontal views of the LAr re-circulation system of one T600 module at LNGS.

3. Argon purity trend and LAr re-circulation system performance

The commissioning of the T600 cryostat at LNGS followed the same successful approach adopted for the surface test run in 2001.

The two T600 modules were evacuated (down to to 10^{-2}-10^{-3} Pa) to remove air, to leak check the cryostat tightness and to perform an appropriate outgassing of the inner materials in order to ensure an acceptable initial LAr purity.

The cryostat cool-down phase was performed after stopping the vacuum pumping and keeping the internal volume in slight overpressure (100 hPa) with ultra-pure gas argon (Ar N60 quality) injection in order to minimize back-diffusion of air from residual leaks and enhance the cooling process. The whole phase lasted only about eight days in order to minimize the period of internal surface outgassing.

T600 filling was carried out in parallel on the two modules using commercial LAr ($H_2O \leq 1$ ppm, $O_2 \leq 0.5$ ppm, $N_2 \leq 3$ ppm) passed through two sets of filters: an extra cartridge with high purification capability was put in series at the entrance of the two Oxysorb/Hydrosorb^{TM} filter batteries. All the Ar gas re-circulation units were put into operation. The whole phase lasted about two weeks guaranteeing no-stop Ar filling at a global rate of about 2 m^3/h.

Just after the cryostat filling completion, with only the GAr re-circulation systems active, LAr purity was sufficiently high and uniform to detect ionization tracks over the full drift distance in both modules as soon as the detector was switched on: the free electron lifetime value was measured to be about 650 microseconds. Even for the LAr scintillation production and transport the LAr purity was immediately found to be acceptable thus permitting to use photomultiplier signals for timing and triggering purposes, since the very beginning, Rubbia (2011).

The LAr re-circulation system was put into operation about 1.5 months after the T600 filling due to several technical problems that lead to the need of upgrading the circuit. A precise and systematic measurement of the charge attenuation along muon tracks was then started to get a LAr purity evaluation of the whole volume and to check its uniformity.

With the liquid re-circulation turned on, the LAr purity steadily increased in similar way in both modules. Unfortunately along the three-years run several LAr re-circulation pump failures were experienced due to excessive usage or damage of pump bearing case that brought in most case to the lubrication filter saturation due to dust. The poor pump reliability led to consequent stop of the LAr re-circulation systems randomly in both systems, resulting in a sudden degradation of the purity in the corresponding module. On average the time interval between two consecutive faults was about 2,000 h. However in few months after the LAr-re-circulation start-up, thanks to pump redundancy (in support of the 2 installed LAr pumps two spare pumps were ready to be used in substitution of the installed ones), adequate spare part equipment and thanks to a prompts and accurate intervention procedure it was possible to guarantee a very fast pump substitution and maintenance (two to five days). As a consequence the free electron lifetime was always maintained much over the 3 ms lower limit (0.1 ppb impurity concentration) all over the run without affecting the T600 detector operation and performance. Purity was rapidly restored as soon as the LAr re-circulation is reactivated.

The analysis of the LAr purity during the T600 data taking demonstrated that similar purity trend were observed in both the T600 modules with an electron lifetime exceeding 7 ms, corresponding to a 12% maximum charge attenuation at longest drift distance. All the filter cartridges were precautionary changed only once along the T600 run.

Nitrogen contamination in argon was periodically measured on the T600 gas phase, using a custom set-up specifically developed for this purpose and based on a commercial mass spectrometer (Pfeiffer QMG 220). N_2 concentration was always found to be below the sensitivity of the instrument (1 part per million).

4. LAr re-circulation system upgrade

Given the frequent faults of the installed pumps, after one year of run we decided to upgrade one of the two LAr re-circulation system (the East module one) using similar pumps to the Barber Nichols (with external motor) used on the nitrogen circuit[‡], that were working in a very efficient and reliable way (exceeding 10000 working hours without interruptions). We started a joint development with Barber Nichols Company aiming at a pump for LAr re-circulation with similar performance of the ones used in the LN_2 circuit while preserving the argon purity. As a result, the Barber Nichols BNCP-32C-000 dedicated model was produced.

In order to host the new pump several major changes were introduced in the circuit design as it is shown in Figure 2. The new pump has a magnetic coupling and vacuum housing. It was located inside a new dedicated vacuum insulated cryostat and the vacuum housing of the pump was welded to the cryostat both at the mounting flange and at the inlet/outlet connections in order to ensure tight connections. A LAr/LN_2 heat exchanger was added up-stream of the pump aspiration to undercool liquid argon and to ensure mono-phase liquid state. A Venturi flow-meter was inserted down-stream of the pump output, profiting of the monophasic argon to measure the flux. Both the heat exchanger and the flow-meter were specifically developed for this application by Date Company.

The ACD CRYO pump and the whole LAr re-circulation system of the East module were stopped on March 20th, 2013. Major works were performed both on the argon and nitrogen circuits in order to introduce the new pump without stopping the East module data taking nor stopping the remnant part of the nitrogen cooling.

The new system was integrated in 2 weeks and the new pump was put into operation on April 4th, 2013. The effects of the prolonged stop and works didn't affect drastically the Ar purity that, after the new pump's switching on, started increasing again and faster than before. The new pump worked efficiently without stops for the last part of the run (up to the June 27th, 2013) allowing to further increase the electron lifetime well above the previously reached 7 ms maximum value.

[‡] Due to important changes in the cooling circuit design for the T600 installation at LNGS even for the LN_2 forced circulation a new type of pump was implemented with respect the surface test run. Instead of the capsulated-motor CRYOMEC pumps, Barber Nichols BNCP-51B-000 pumps were used. Barber Nichols BNCP-68-M1 pumps were instead used for LN_2 transfer in the Stirling Cryogenics BV cryocooler system.

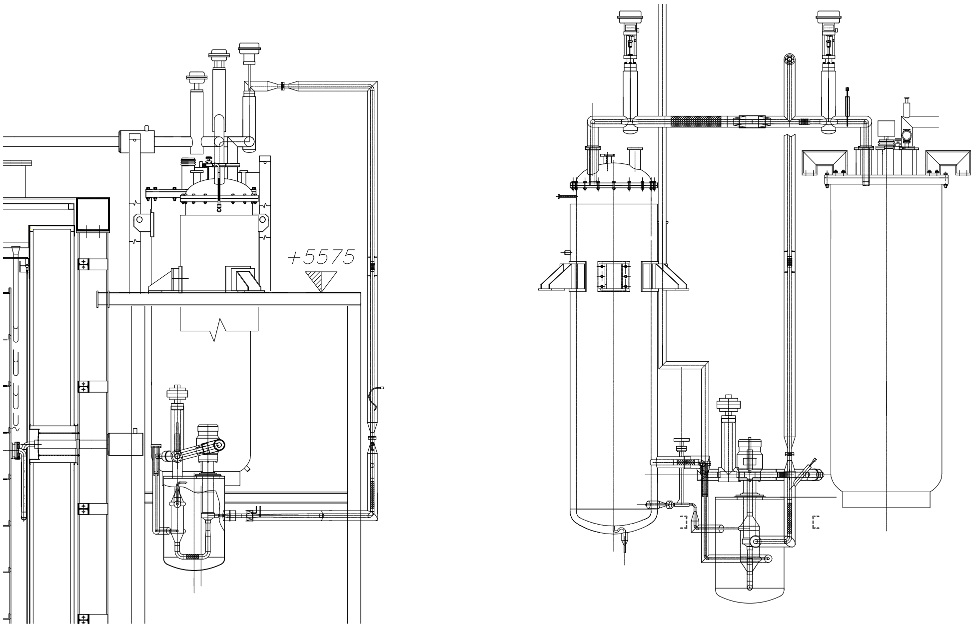

Fig. 2 Side and frontal views of the upgraded LAr re-circulation system of the East T600 module at LNGS.

5. Conclusion

The T600 run at LNGS lasted 3 years giving extraordinary results in terms of LAr purity: free electron lifetime exceeded 7 ms in both the modules, corresponding to an impurity content in LAr of less than 40 part per trillion O_2 equivalent giving a 12% maximum charge attenuation at the longest drift distance. In addition the new pump installed on the East module allowed to further increase the electron lifetime well above the previously reached 7 ms maximum value.

The ICARUS results represent a milestone for the development of any other noble liquid detector and pave the way to the construction of huge detectors, as required for example for the LBNE experiment, LBNE Collaboration (2012). Scaling-up of the ICARUS liquid argon purification technology is feasible; commissioning procedures can be tuned to bigger size plants too.

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