PROCEEDINGS OF SCIENCE

Purification of large volume of liquid argon for LEGEND-200

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The design, construction and performance of the system capable of purifying 65 m^3 of liquid argon to sub-ppm level designed for LEGEND–200 experiment is presented. The quality of the purified liquid argon is monitored in real-time during the purification process, by measuring the argon triplet state lifetime and simultaneous direct measurements of the concentrations of impurities such as water, oxygen, and nitrogen with a sensitivity of 0.1 ppm. The achieved argon triplet lifetime value measured inside the LEGEND cryostat, when filled in 70% of its capacity, was at the level of τ_3 = 1.3 μs. If needed, the system may also be used later to purify liquid argon filled into the LEGEND cryostat in the loop mode.

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1. Introduction

The LEGEND-200 experiment is currently being commissioned at the Laboratori Nazionali del Gran Sasso (LNGS) in Italy. Its main purpose is a quasi-background-free search for neutrinoless double beta decay of Ge-76. LEGEND-200 combines technologies like Liquid Argon (LAr) veto system developed in GERDA or underground formed copper developed by MAJORANA DEMONSTRATOR, as well as from other groups using world best experience to deploy up to 200 kg High Purity Germanium (HPGe) detectors for the neutrinoless double beta decay search. The goal is to achieve a background index below 2×10^{-4} cts/(keV•kg•yr) and obtain a discovery sensitivity for a half-life of 10^{27} year after 5 years of operation [1]. In LEGEND-200 LAr serves as a cooling medium for the HPGe detectors as well as a passive and active shield. The detector's cryostat has the capacity of 65 m^3 (~90 tonnes of LAr). The LAr active veto instrumentation is composed of light guiding fibers surrounding the HPGe detectors. The fiber shroud is coupled to SiPM arrays detecting the scintillation light of argon (128 nm). It has been already shown in the GERDA experiment that the LAr veto was a very powerful tool for background rejection [2].The scintillation properties of LAr like the attenuation length, the light yield and the triplet state lifetime (τ_3) are worsened by the presence of electronegative impurities such as oxygen, water and nitrogen. It is caused by quenching and absorption processes. Consequently, the efficiency of the LAr veto is significantly impacted by the level of argon purity. LLAMA (LEGEND Liquid Argon Monitoring Apparatus) [3] has been installed at the bottom of the cryostat to monitor the argon quality inside the cryostat during LEGEND filling, commissioning and operation. It consists of 16 SiPMs at various distances to the radioactive source, measuring LAr triplet lifetime, light yield and attenuation length. In order to achieve the best possible performance of the LAr detector, it has been decided to purify the argon acquired from a vendor while filling the cryostat. The purifying process was performed during initial filling of the LEGEND-200 cryostat in summer 2021. If needed, the LAr purification system may also be used later to purify LAr filled into the cryostat in the loop mode. A dedicated cryogenic pump has been installed on the bottom of the LEGEND-200 cryostat to circulate LAr between the purification system and the cryostat.

2. Design of the LEGEND Liquid Argon Purification System.

LEGEND Liquid Argon Purification System (LLArS) was designed to remove O_2 and H_2O and also N_2 from LAr while filling the Legend cryostat. The capacity of the LAr storage tank installed in LNGS for LEGEND used by the vendor to deliver argon is 8 tonnes. Therefore, the absorbent mass used for purification was chosen to process at least 8 tonnes between the regenerations of the system. LLArS (Fig.1) is integrated with the argon filling line and placed between the LAr storage tank and the cryostat. The argon is processed by two units, each consist of 2 traps placed in vacuum insulation. The traps are equipped with heaters and temperature sensors. Water (and N_2) removal module is based on two traps filled with 4 kg of Molecular Sieve 4 Å each. The oxygen removal module uses two traps, each filled with 5 kg of copper catalyst (Cu-0266 S®). Both adsorbers saturate with contaminants and need to be regenerated to restore theirs purification properties.

The established regeneration procedure is the following:

- For the O_2 traps:

Heating the saturated catalyst up to 250 ºC and flushing the traps with mixture of 5% H_2 in N_2 gas for reduction of CuO while monitoring level of released water (produced in the reduction reaction) and hydrogen gas at the exit of the line. About 800 volumes of the traps need to be exchanged to ensure the full reduction of the copper catalyst. Then H_2/N_2 gas is pumped out together with residual water to high vacuum (< 10^{-5} mbar).

- For the H_2O (N_2) traps:

Restoration of the molecular sieve is a mechanical process, then it is sufficient to heat the traps up to 250 ºC and pump it for several hours to reach high vacuum.

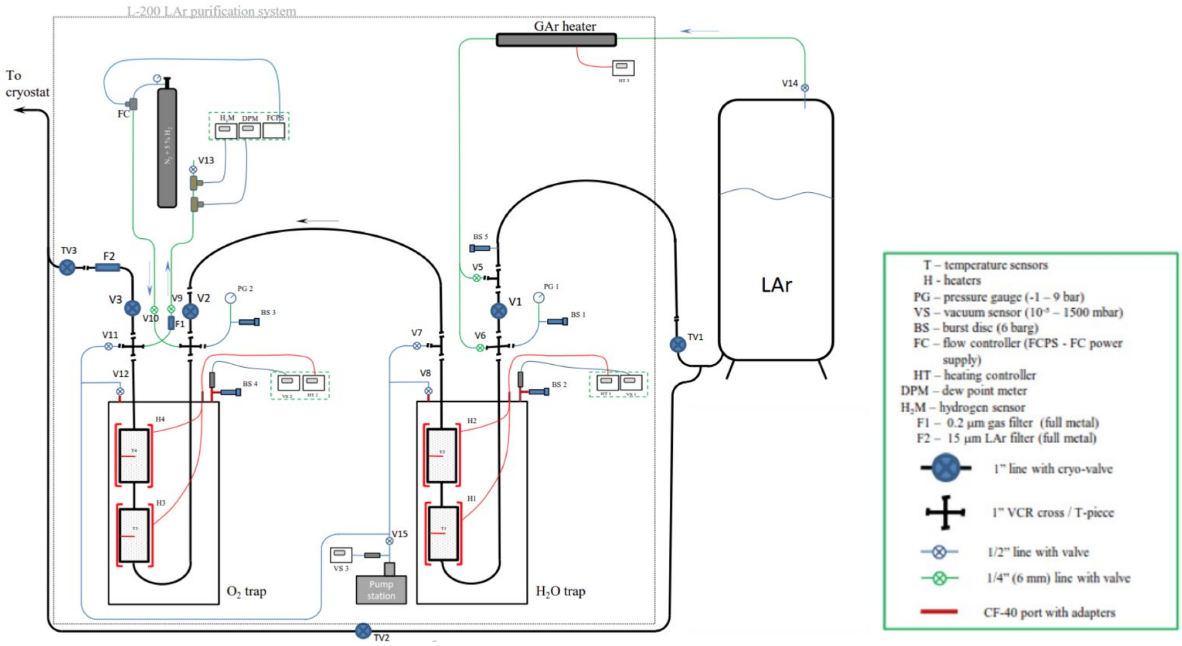

Figure 1. LEGEND LAr purification system (LLArS) instrumentation diagram.

The quality of the purified liquid during the LEGEND cryostat filling is monitored outside the cryostat in a real-time by two devices. The argon triplet lifetime measurement is performed with a dedicated Scintillation Analyzer. Simultaneous direct measurements of the concentrations of impurities such as water, oxygen, and nitrogen with a sensitivity of 0.1 ppm are realised with an Impurity Analyzer. The Scintillation Analyzer is based on a 60-liter metal-sealed dewar closed with a CF-160 flange. The LAr sample is viewed by two 2" TPB-coated PMTs registering the scintillation light. The four installed temperature sensors serve as LAr level monitoring. The DAQ unit is equipped with a 400 MHz digitizer. The pulse shape analysis of collected light allows to determine the long component – triplet state lifetime τ_3.

3. Operation of LLArS

The purification traps were constructed and tested in several small-scale tests at Jagiellonian University in Cracow showing good purification capabilities. The argon sample scintillation properties were measured using Scintillation Analyzer by determining τ_3 value. It was shown that argon with τ_3 = 0.80 μs prior to purification scintillates with τ_3 = 1.25 μs after.

The whole system was fully assembled and tested at Technische Universität München and then finally commissioned underground at LNGS in May 2021. The commissioning test's goal was to proof the elaborated filling procedure and check the entire cryogenic system by purifying about 4 tonnes of LAr and sending it to the cryostat. LAr with Linde certificate stating impurity level of: O_2 = 0.2 ppm, N_2 = 0.2 ppm, H_2O = 0.17 ppm was processed. Triplet lifetime was measured before (τ_3 = 0.85 μs) and after (τ_3 = 1.25 μs) the purification using the Scintillation Analyzer setup. The system performed well and as expected.

4. Filling the LEGEND-200 cryostat in summer 2021.

After the commissioning test, the system was fully regenerated and prepared for the cryostat filling campaign. The filling started with low argon flow (~150 kg/h), to cool down the cryostat slowly and reduce the temperature of the walls avoiding large temperature gradients. The process was monitored 24/7 due to safety and quality control reasons. The cooling phase was also used to determine the breakthrough curve of the purification system by monitoring the triplet lifetime as a function of the mass of the processed LAr. After purifying 12 tons the triplet lifetime dropped from 1.3 μs to about 1.1 μs and the system needed to be regenerated.

The standard LAr flow during the filling phase was kept at ~350 kg/h with continuous operation and monitoring. The system regeneration breaks were needed after each 12 tons of argon processed. LLArS performed as expected filling the LEGEND-200 cryostat up to 70 % of its capacity (i.e., 68 tonnes) with high purity argon characterized by τ_3 = 1.3 μs level (Fig.2), measured by LLAMA inside the cryostat.

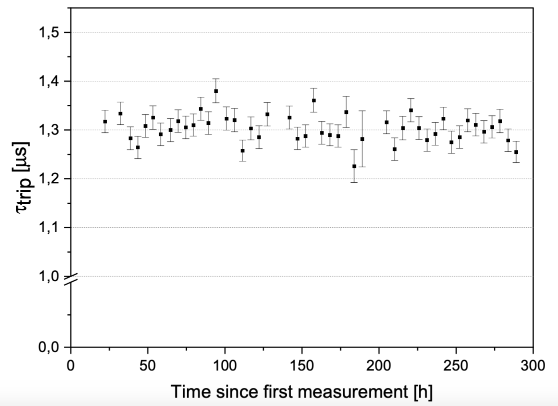

Figure 2. The τ_3(τ_{trip}) value monitoring performed by LLAMA during the LEGEND cryostat filling.

References

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