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Exploring N₂ Capturing in Liquid Argon using Li-FAU Mol Sieve in the Iceberg Cryostat

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Liquefied ultra-pure noble gases are typically the chosen target for neutrino and dark matter experiments. Commonly, the required grade of purity of such cryogenic liquids in terms of oxygen contamination (< 100 ppt), which makes it necessary for the Gaseous Argon (GAr) and/or the Liquid Argon (LAr) to circulate through adsorption columns filled, typically, with BASF Cu-0226S and Mol Sieve 4A for capturing oxygen gas and water. Another requirement that emerged during the design of LBNF-DUNE is the purity of the LAr in terms of nitrogen concentration, as it was observed that both nitrogen and oxygen contamination concentrations in the LAr can cause the reduction of the LAr scintillation light emission. In particular, a decreasing behavior in the lifetime and the relative amplitude of the slow component is relevant from concentrations of 1 ppm of nitrogen and 0.1 ppm of oxygen. Recently, exploratory experiments performed in the Liquid Argon Purification Cryostat (PuLArC- ~ 90 L of LAr) at IFGW/Unicamp had been successfully used to purify LAr from nitrogen using an innovative solid adsorbent based on a Li-FAU Molecular Sieve. Here, we report experiments of nitrogen capturing in LAr carried out at the Iceberg Cryostat (~ 3000 L of LAr) at Fermilab. For the experiments, several controlled injections of nitrogen gas were made into the Iceberg cryostat, and the impurity concentrations in the LAr (both for nitrogen and oxygen) were monitored using a calibrated gas analyzer connected to the Iceberg during the purification process. The experimental data confirmed that the Li-FAU is capable of capturing the nitrogen gas from the LAr. A numerical methodology based on the mass and energy balances of the system was applied to better understand the behavior of the adsorption process and to help with the scale-up of the process. A Pore Diffusion Model (PDM) coupled with parameter optimization was proposed to predict the nitrogen concentration in the Iceberg cryostat over time. The model parameters applied in this work were determined based on experimental data acquired in the PuLArC. The model was able to accurately reproduce the data from the Iceberg, indicating that the mathematical model could be a valuable tool to design and operate the GAr/LAr system of LBNF-DUNE. Finally, the reported results of the experiments performed in the Iceberg unequivocally confirmed the Li-FAU capacity to capture nitrogen from LAr, invoking the discussion about the possibility of this adsorbent being used in other LAr-based experiments at Fermilab and CERN. It was possible to predict from simulations the necessary time to reach the required purity from an initial contamination concentration and also the number of purification cycles until solid saturation.

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