# Exploring $N_2$ Capturing in Liquid Argon using Li-FAU Mol Sieve in the Iceberg Cryostat

**Pedro Bianchi Neto**[a]
Renato Soccol Junior[a]
Flor de Maria Blaszczyk[e]
Sergey Koshelev[e]
Alan Hahn[e]
Daniel Correia[d,e]
Dilson Cardoso[c]
Roza Doubnik[e]
Mark Adamowski[e]
David Montanari[e]
Thiago P. M. Alegre[b]
Ana Amélia Machado[b]
Ettore Segreto[b]
José Mansur Assaf[c]
H. da Motta[d]
P. G. Pagliuso[b]
Dirceu Noriler[a]

[a] School of Chemical Engineering, UNICAMP, Brazil
[b] “Gleb Wataghin”, Institute of Physics, UNICAMP, Brazil
[c] Advanced Materials and Energy Research Center, UFSCar, Brazil
[d] Brazilian Center for Research in Physics, Rio de Janeiro, Brazil.
[e] Fermi National Accelerator Laboratory, Illinois, USA.

# Background

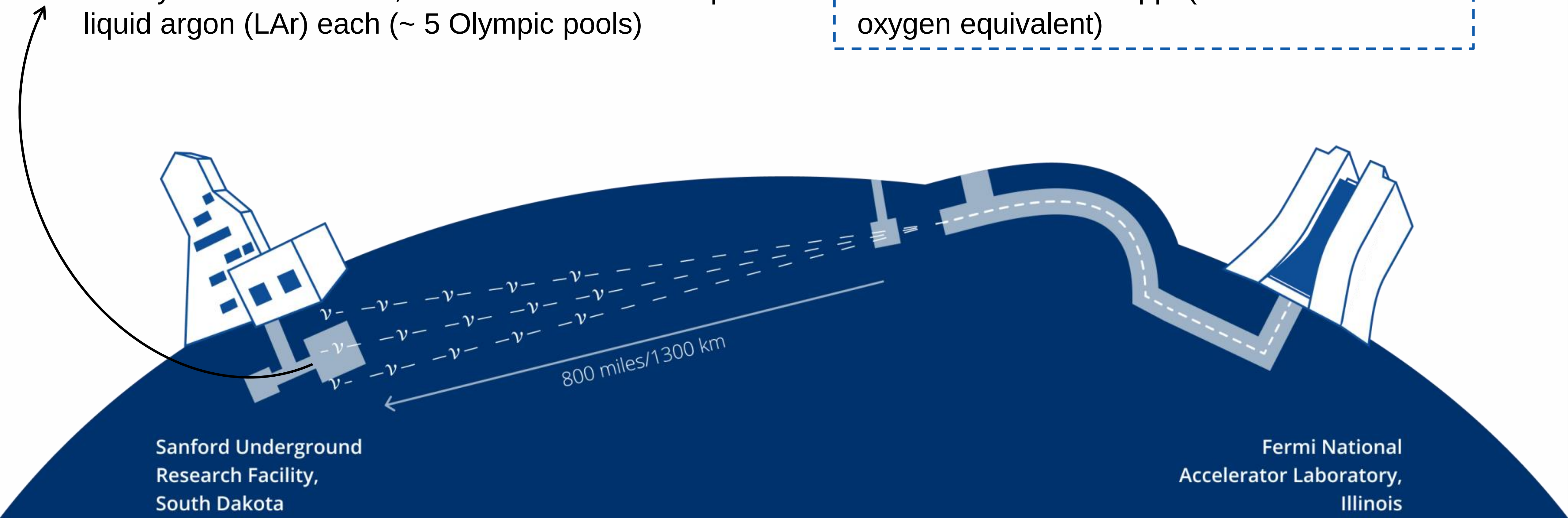
The DUNE

The **L**ong-**B**aseline **N**eutrino **F**acility (LBNF), located at the Sanford Underground Research Facility in Lead, South Dakota, hosts the **D**eep **U**nderground **N**eutrino **E**xperiment (DUNE).

- the cryostats contain 17,500 metric tons of ultra-pure liquid argon (LAr) each (~ 5 Olympic pools)

- How pure should the argon be for the experiment?

**Detector 1** → < ~ 100 ppt (or ~ 1.4 ml of oxygen equivalent)
**Detector 2** → < ~ 50 ppt (or ~ 0.7 ml of oxygen equivalent)



# Motivation

How do you reach such pure argon?

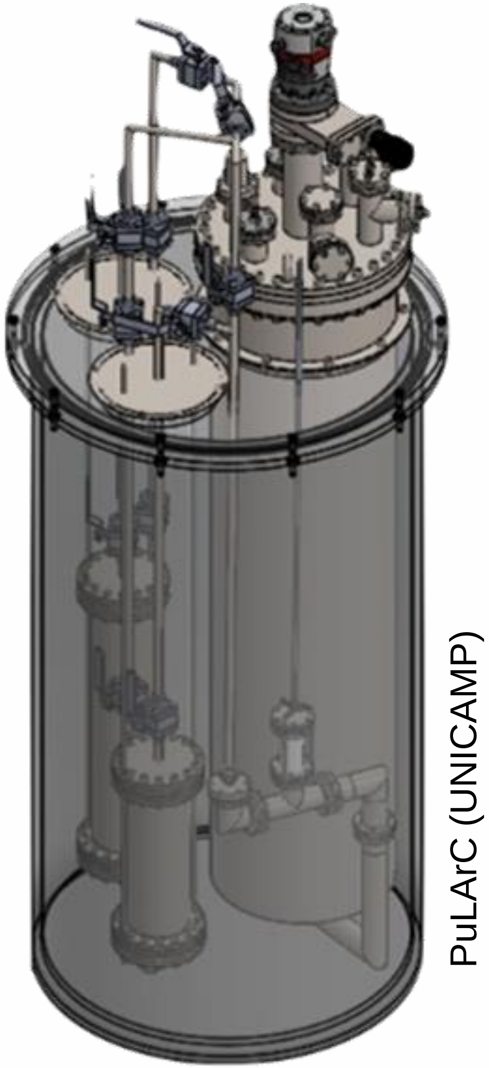
Contaminants in argon **significantly affect** experimental results:

- $N_2$, $O_2$ and $H_2O$

- The argon is purified using a **filtering media** (which captures $O_2$ and $H_2O$);
- As the typical filters can't remove $N_2$, it must be kept low in the argon purchasing specification.

An effective removal of $N_2$ impurities during liquid argon circulation was not known and it is desirable.

- A systematic study incorporating collaborative materials and methods, and combining numerical and experimental analysis;
- **Li-FAU zeolite**.



PuLArC (UNICAMP)

# Experiments

Liquid argon purification

Experiments at Unicamp – **Pu**rification **L**iquid **Ar**gon **C**ryostat (PuLArC)

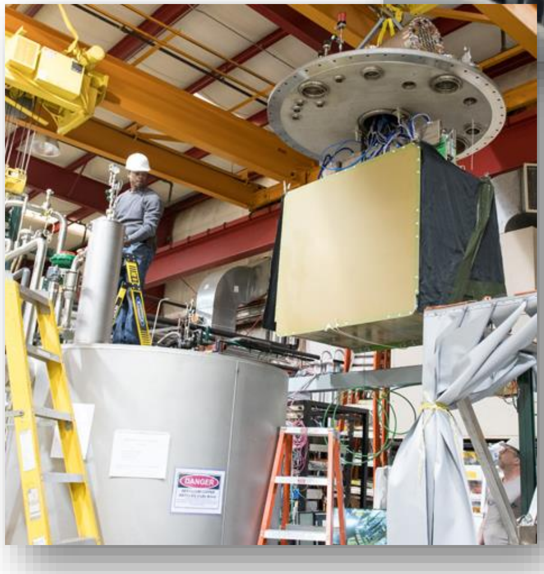
- ~ 1.2 kg of Li-FAU zeolite adsorbent;
- $N_2$ contamination reduction: 20-50 ppm to 0.1-1.0 ppm (1-2 hours of active circulation)

Experiments at Fermilab – **I**ntegrated **C**ryogenic **E**xperiment for **B**eam **E**nergy **R**esearch and **G**eneration (ICEBERG) Cryostat

- ~ 3 kg of Li-FAU zeolite adsorbent;
- $N_2$ contamination reduction: ~5 ppm to <1 ppm (96-hour cycles without active circulation).
- calibrated gas analyzer – monitor $N_2$ dissolved in LAr.

Measure concentration of $N_2$ dissolved in LAr with passing time: Recontamination after each cycle.

PuLArC (UNICAMP)



ICEBERG (Fermilab)

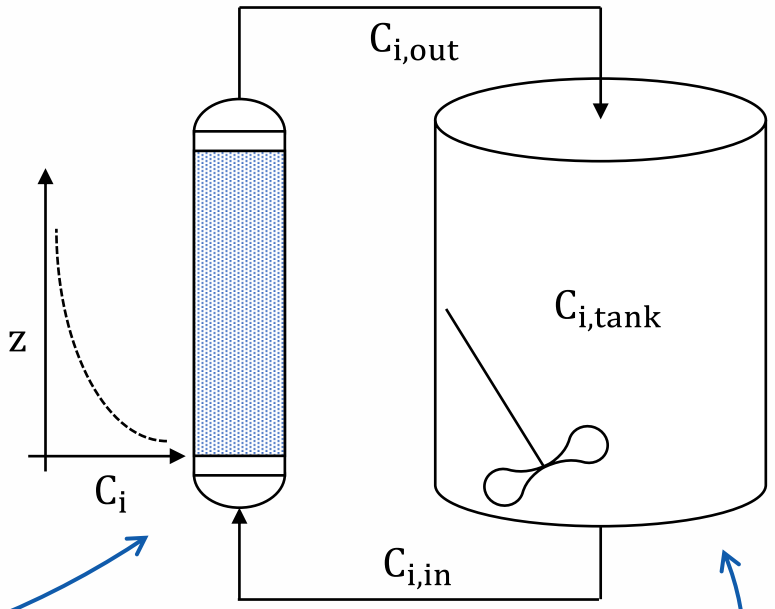
# Mathematical Model

How was the system modeled?

Transient mass balance in fluid phase, intra-particle, solid phase, and storage tank modeling.

**Hypotheses:**

- isothermal system;
- constant physical properties;
- plug flow;
- constant mass flow;
- uniform bed porosity;
- well-mixed tank.



Fluid-phase

$$\frac{\partial C_i}{\partial t} = D_L \frac{\partial^2 C_i}{\partial z^2} - v_z \frac{\partial C_i}{\partial z} - \frac{(1-\epsilon_b)}{\epsilon_b} K_L a_V (C_i - Y_i)$$

Intra-particle fluid-phase

$$\frac{\partial Y_i}{\partial t} = \frac{K_L a_V}{\epsilon_p}(C_i - Y_i) - \rho_S \frac{(1-\epsilon_p)}{\epsilon_p}\frac{\partial q}{\partial t}$$

Solid-phase

$$\frac{\partial q_i}{\partial t} = \sum_{=1}^{NC} \frac{\partial f_{eq}(Y_i)}{\partial Y_i}\frac{\partial Y_i}{\partial t} \qquad f_{eq}(Y_i) = \frac{K_{RP}\, Y_i}{1 + a_{RP}\, Y_i^{g}}$$

Well-mixed tank

$$\frac{\partial C_{i,tank}}{\partial t} = \frac{C_i Q}{V} - \frac{C_{i,in} Q_{in}}{V} - \frac{C_{i,in}}{V} \cdot \frac{dV}{dt}$$

# Mathematical Model

Closure equations

## Adsorption isotherm:

Redlich-Peterson:

$$f_{eq}(Y_i) = \frac{K_{RP}\, Y_i}{1 + a_{RP}\, Y_i^{g}}$$

constant related to adsorption capacity (m3/kg)

related to the max amount adsorbed in the first layer

adsorption intensity

Predicting the filter’s behavior is important to determine its capacity – specifying the saturation time and number of cycles needed to reach the required purity

## Mass transfer coefficient :

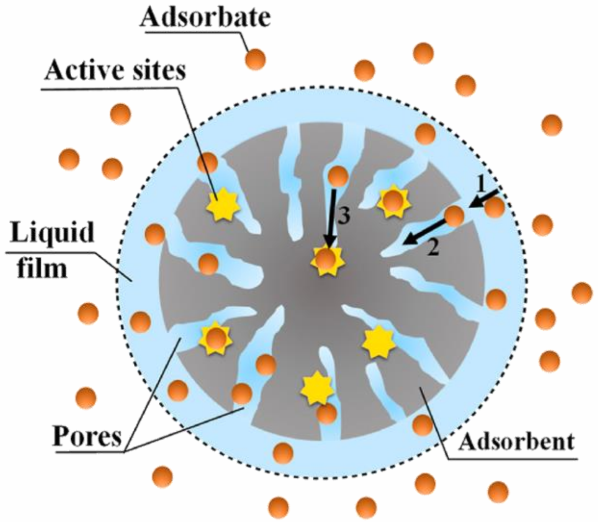
$$\frac{1}{K_L} = \frac{1}{k_e} + \frac{1}{\epsilon_p k_i} \qquad Sh = \frac{k_e d_p}{D_L} \qquad \frac{Sh}{Sc^{1/3} Re_m} = 0.91 Re_m^{-0.51} \qquad Sc = \frac{\mu_m}{\rho_L D_L}$$

intraparticle porosity

$$k_i = \frac{10\, D_L}{\tau\, d_p}$$

molecular diffusivity of $N_2$ in Ar

particle tortuosity



Intraparticle diffusion adsorption [1]

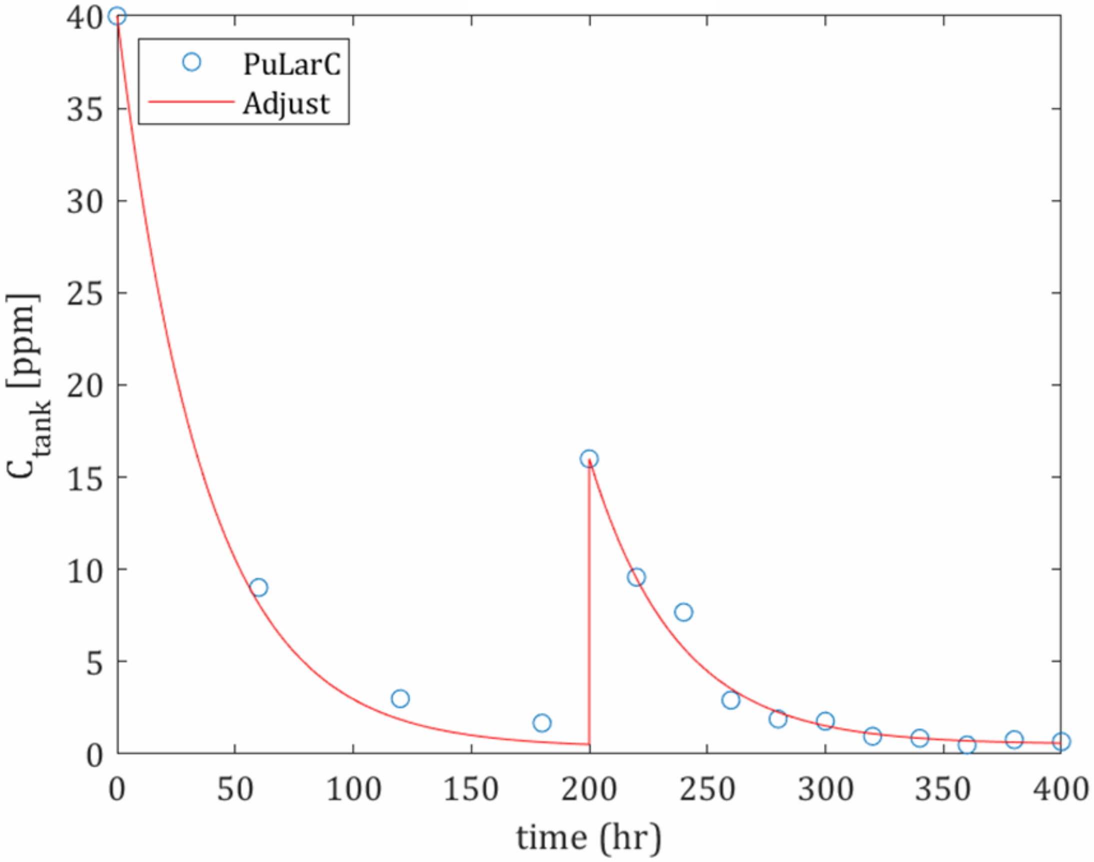
# Model Solution

PuLarC parameters optimization

## PuLarC simulation conditions:

- simulation – 2 cycles (media is not regenerated before the second cycle);
- $C_{N_2,1} = 40\ \mathrm{ppm}$
- $C_{N_2,2} = 16\ \mathrm{ppm}$
- $Q = 4\ \mathrm{L/min}$

- tank volume: $0.14\ \mathrm{m}^3$
- media level: $29\ \mathrm{cm}$
- bed diameter: $95.5\ \mathrm{cm}$
- bed porosity: $0.38$
- $\rho_S = 936\ \mathrm{kg/m}^3$
- $d_p = 2\ \mathrm{mm}$



$$f_{eq}(Y_i) = \frac{K_{RP}\ Y_i}{1 + a_{RP}\ Y_i^g}$$

| Isotherm parameters | |
|---|---|
| $K_{RP}$ | $1.69\ 10^6$ |
| $a_{RP}$ | $3.47\ 10^7$ |
| $g$ | $0.911$ |

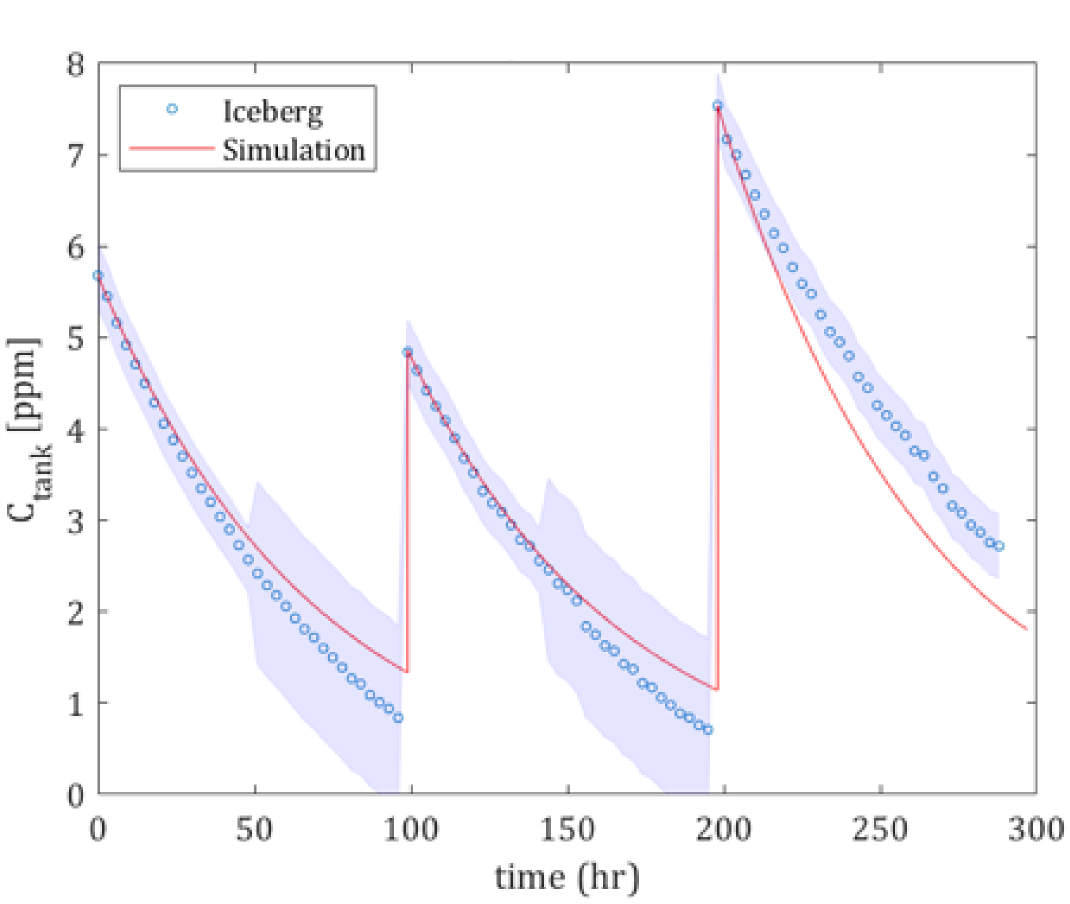
# Model Solution

Iceberg simulation with PuLarC parameter

## Iceberg simulation conditions:

- simulation – 3 cycles (media is not regenerated before the second and third cycles);
- $C_{N_2,1} = 5.68\ \text{ppm}$
- $C_{N_2,2} = 4.48\ \text{ppm}$
- $C_{N_2,1} = 7.54\ \text{ppm}$
- $Q = 0.61\ \text{L/min}$

- tank volume: $2.5\ \text{m}^3$
- media level: $47\ \text{cm}$
- bed diameter: $12\ \text{cm}$
- bed porosity: $0.38$
- $\rho_S = 936\ \text{kg/m}^3$
- $d_p = 2\ \text{mm}$



$$f_{eq}(Y_i) = \frac{K_{RP}\ Y_i}{1 + a_{RP}\ Y_i^g}$$

| Isotherm parameters | |
|---|---|
| $K_{RP}$ | 1.69 $10^6$ |
| $a_{RP}$ | 3.47 $10^7$ |
| g | 0.911 |

# Model Solution

Iceberg simulation with PuLarC parameter

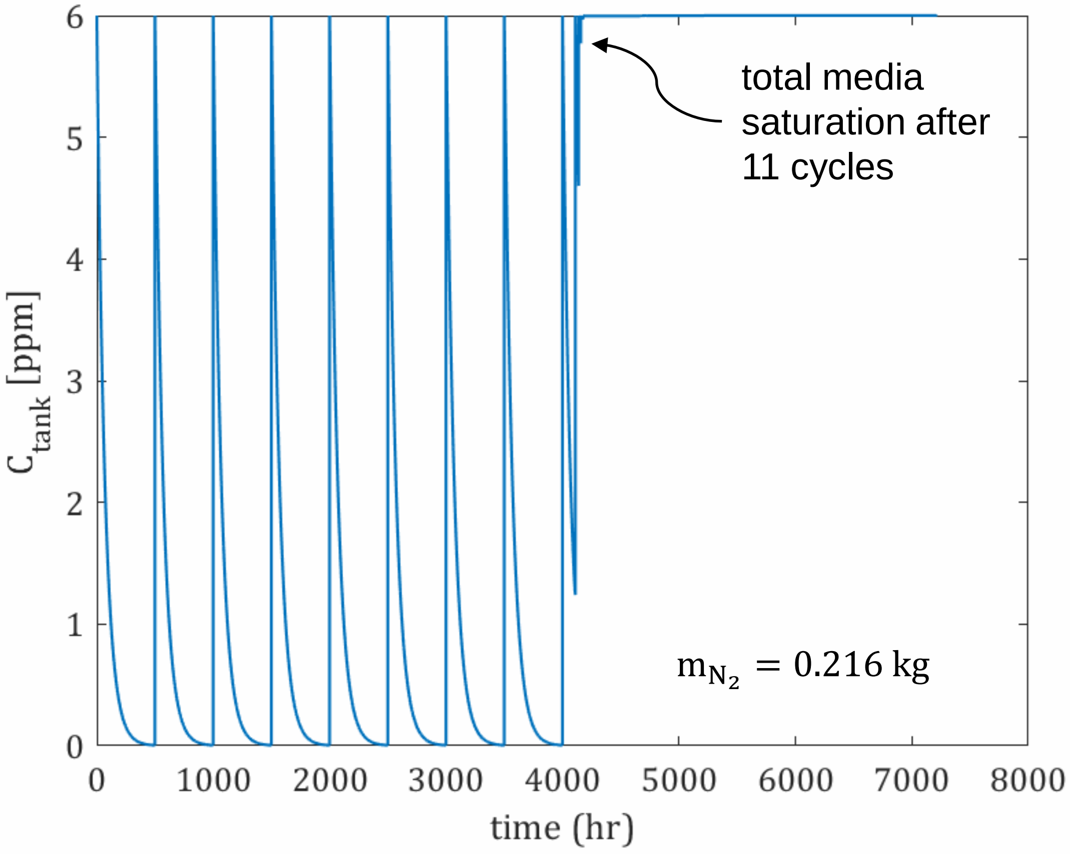
## Iceberg simulation:

- simulation – 20 cycles (media is not regenerated before each cycle);
- $C_{N_2,in} = 6\ \mathrm{ppm}$
- $Q = 0.61\ \mathrm{L/min}$

- tank volume: $2.5\ \mathrm{m}^3$
- media level: $47\ \mathrm{cm}$
- bed diameter: $12\ \mathrm{cm}$
- bed porosity: $0.38$
- $\rho_S = 936\ \mathrm{kg/m}^3$
- $d_p = 2\ \mathrm{mm}$

- Simulation stops when:

$$\frac{dC_{N_2}}{dt} < 10^{-8}\ \left(\frac{\mathrm{kg}}{\mathrm{m}^3\mathrm{s}}\right)$$



total media saturation after 11 cycles

$m_{N_2} = 0.216\ \mathrm{kg}$

$$f_{eq}(Y_i) = \frac{K_{RP}\ Y_i}{1 + a_{RP}\ Y_i^g}$$

| Isotherm parameters | |
|---|---|
| $K_{RP}$ | $1.69\ 10^6$ |
| $a_{RP}$ | $3.47\ 10^7$ |
| $g$ | 0.911 |

# Final Remarks

Conclusion and Outlook

- The successful testing of Li-FAU as an **effective adsorbent** for $N_2$ impurities marks a significant advancement for neutrino experiments, improving LAr quality. (see recent article at https://news.fnal.gov/2024/08/brazilian-researchers-discover-new-way-to-purify-liquid-argon-for-neutrino-experiments/)

- Promising results from the PuLArC and Iceberg cryostats – Li-FAU could be a viable alternative to traditional purification (molecular sieve 4A) in large-scale LAr cryostats.

- The purification media will improve the quality of the LAr used in experiments **+** allow for removing $N_2$ contaminants if air is accidentally introduced into the cryostat due to equipment malfunction.

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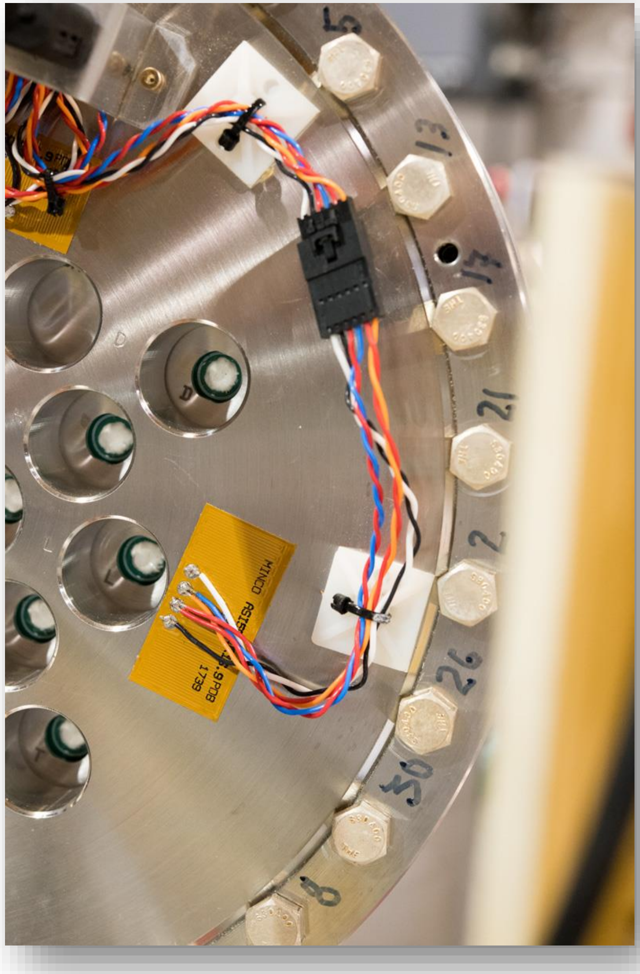
- A mathematical model was proposed for the adsorption process of nitrogen using the Li-FAU in liquid;

- The proposed model was able to match the experimental results obtained with the ICEBERG cryostat;


ICEBERG (Fermilab)

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# Exploring $N_2$ Capturing in Liquid Argon using Li-FAU Mol Sieve in the Iceberg Cryostat

Thank You for your attention!