LAr Purificarion for LNFB DUNE: Fluid Dynamics Analysis of the LAr Purification in Prototype Systems

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Background

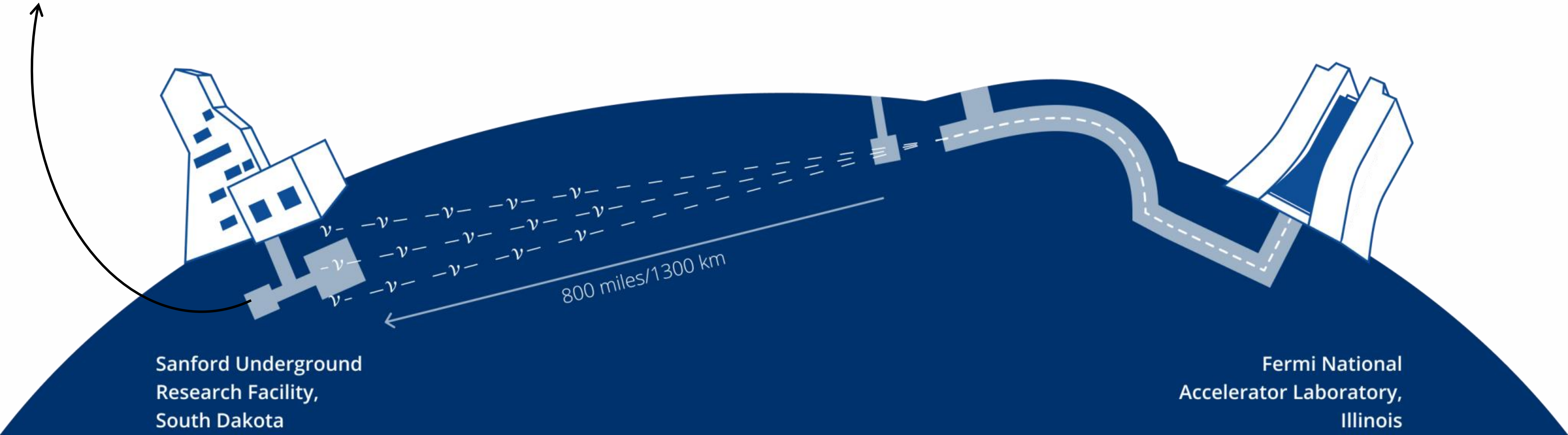
The DUNE

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)

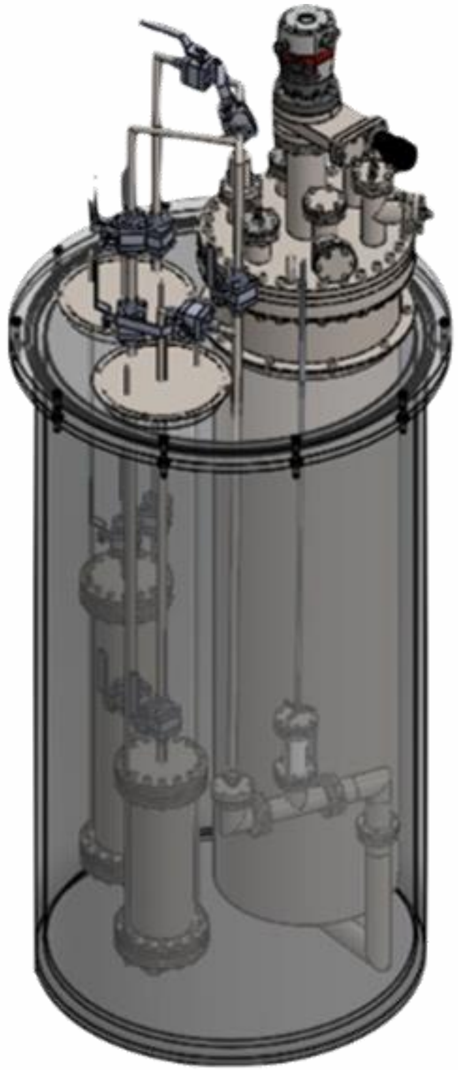


Motivation

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**

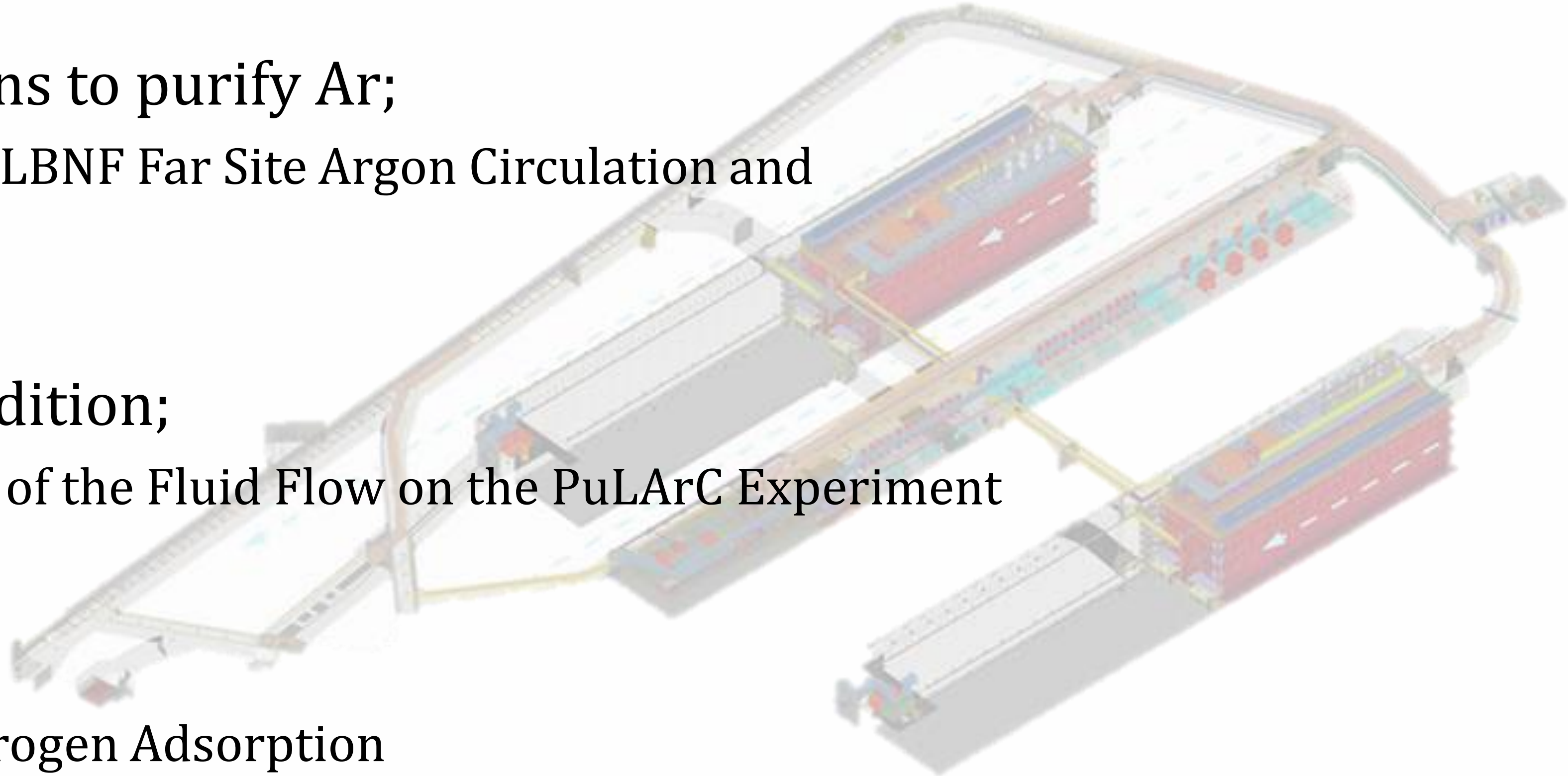


Presentation outline

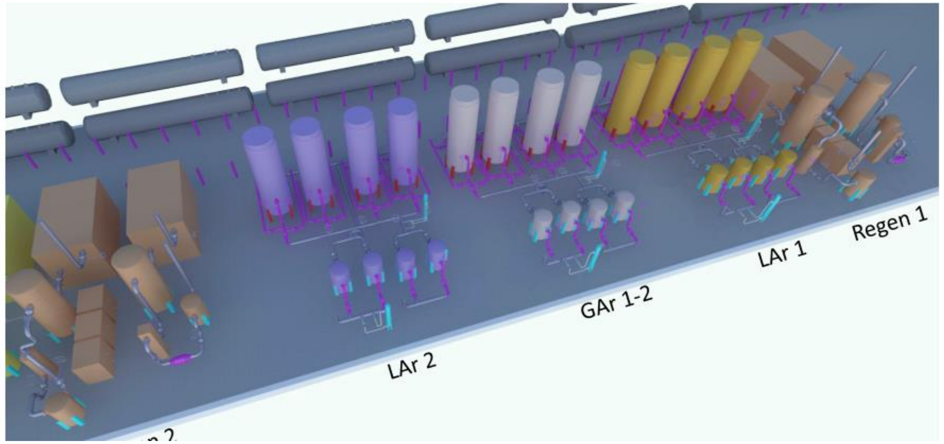
What is necessary to DUNE operates

To DUNE operates and get data, steps are necessary:

- Guarantee operation conditions to purify Ar;
  - Macroscopic simulations of the LBNF Far Site Argon Circulation and Purification Systems;
- Filters operate in optimal condition;
  - CFD Modelling and Simulations of the Fluid Flow on the PuLArC Experiment
- Media capabilities;
  - Modeling and Simulation of Nitrogen Adsorption



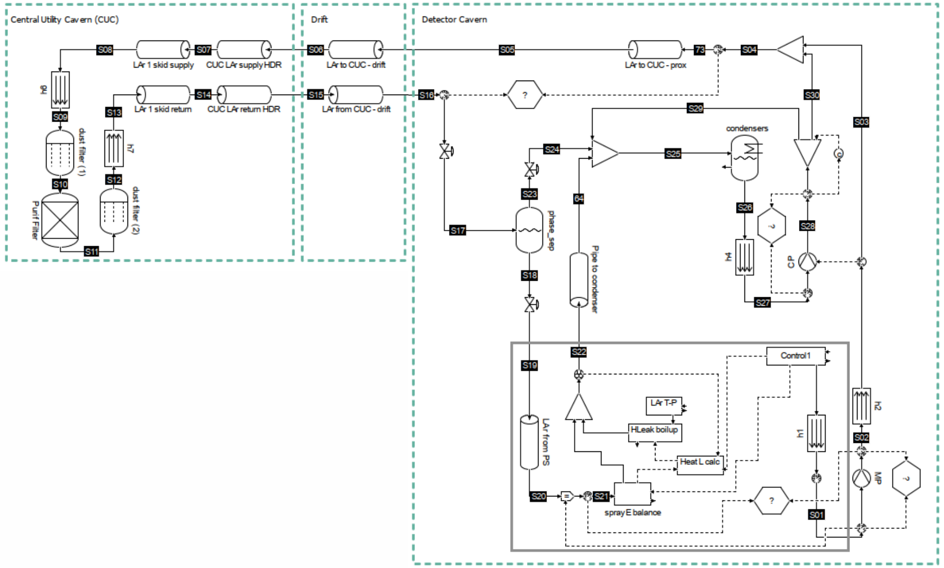
- Objective:
  - The main objective of this study is to simulate the liquid Argon (LAr) and gaseous Argon (GAr) circuit to guarantee operability, avoid design failures, and estimate the required pump power.
- Steps:
  - Identify pipe streams in the CAD and P&ID diagram – dimensions and elevation, number of accessories;
  - Implement the collected information into the macroscopic simulator: head loss, heat leak, pump power, different operating conditions.

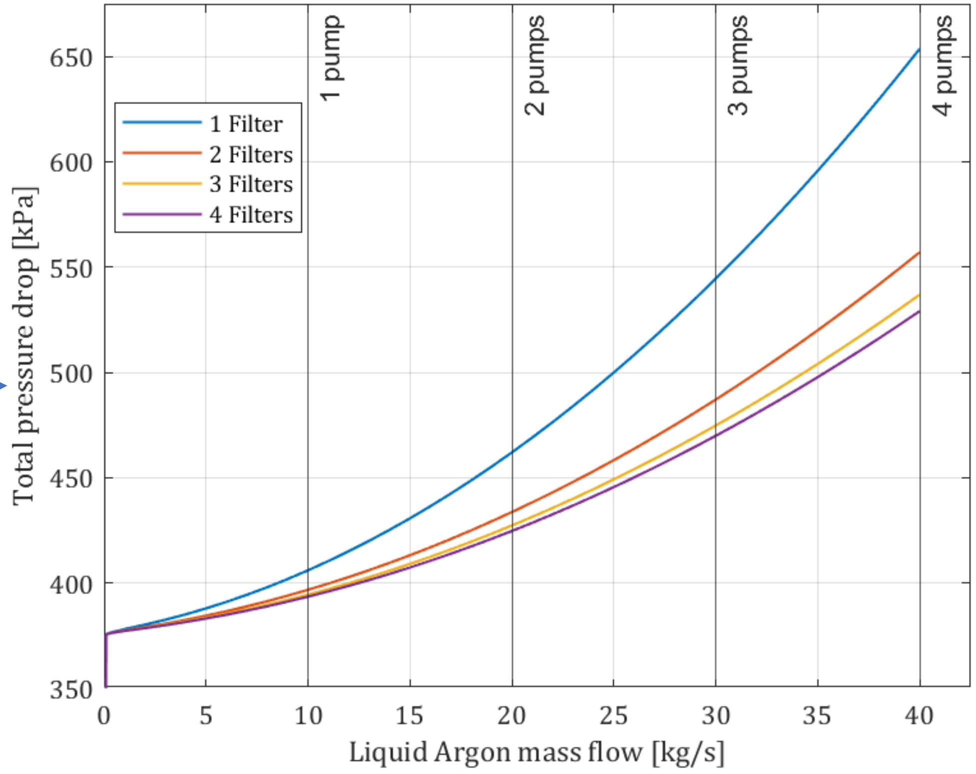


Macroscopic simulations of the LBNF Far Site Argon Circulation and Purification Systems

Pump power estimation

- Designing the Liquid Argon (LAr) pump for DUNE
  - Premise ⟶ Pump must provide pressure above 160kPa (Avoid LAr gasification)
  - ~342 m of pipeline !!





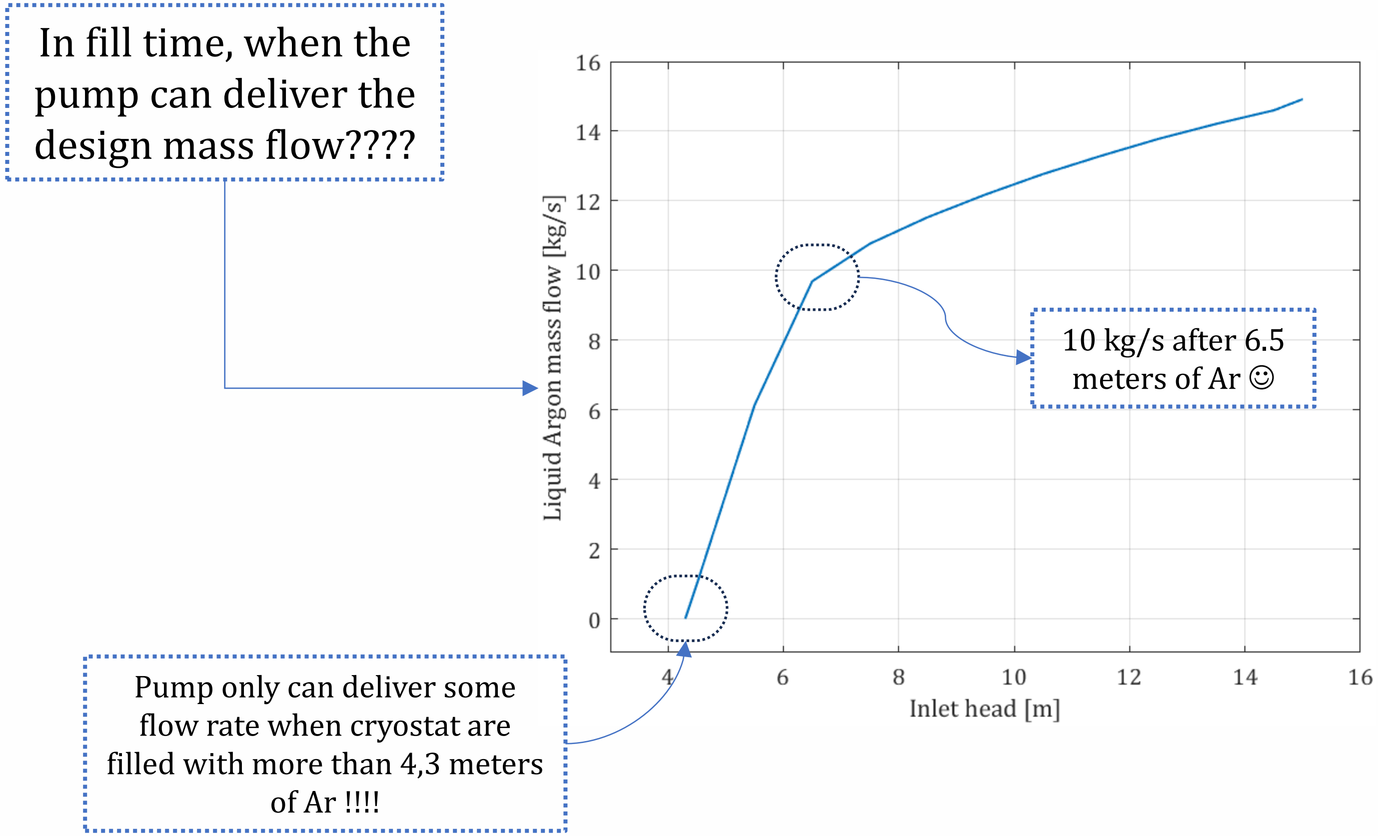
For the total pressure drop, almost 77% is due to elevation changes

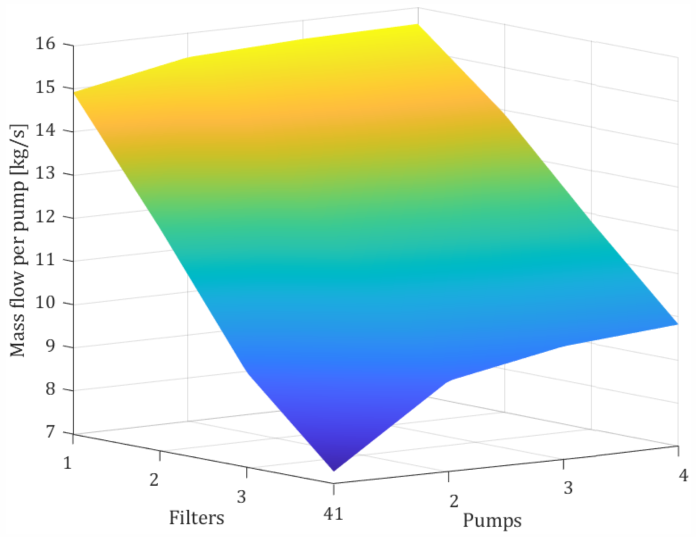
Each pump must provide ~ 5.5 kW to overcome the pressure loss.

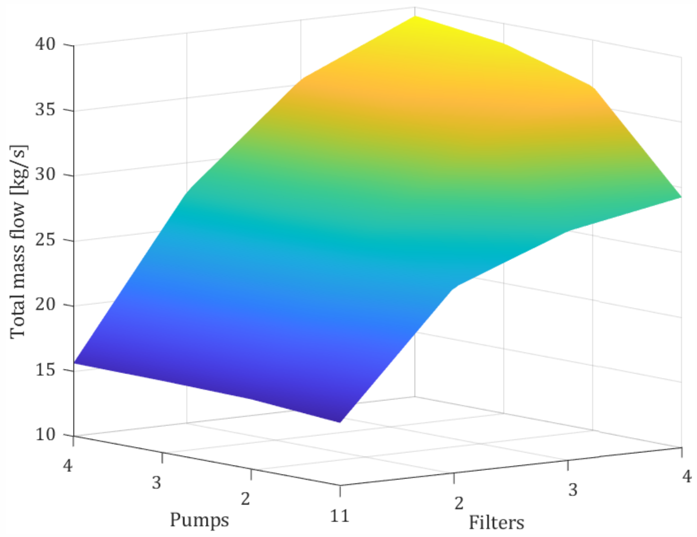
Pump design estimative

Theorical mass flow rate (kg/s)	10
Necessary power (kW)	5.46
Pump head (m)	27.8
Available NPSH (m)	15.81

- Explore the operational conditions:
  - Verify the mass flow when the cryostat is being filled (left);
  - Mass flow at 16 different operation scenarios (right)

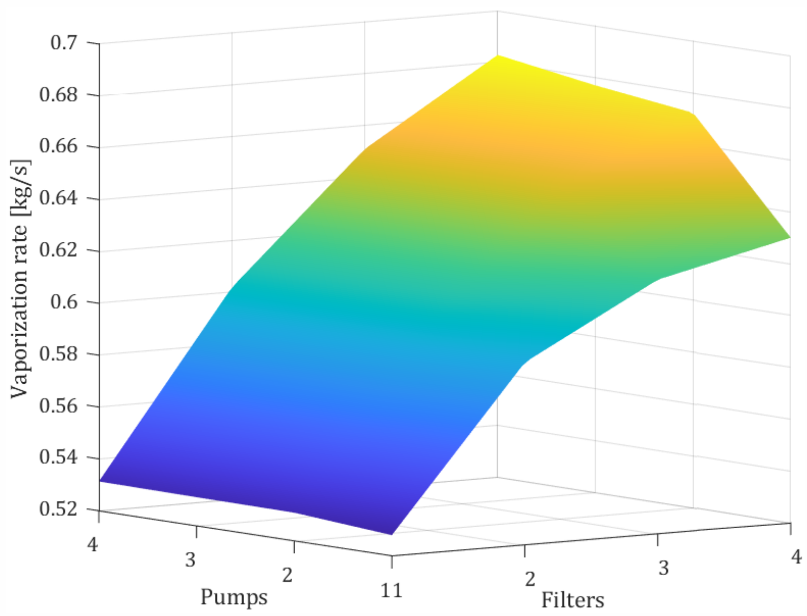


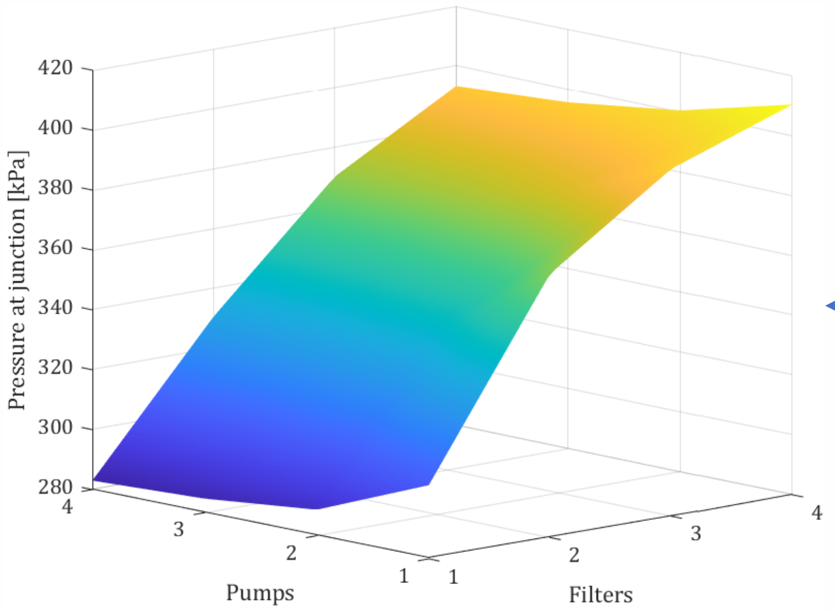




GAr estimation

- GAr circuit
  - Estimative of vaporization flow rate at cryostat;
  - Necessary pressure to feed of GAr in pipes ⟶ GAr pump;



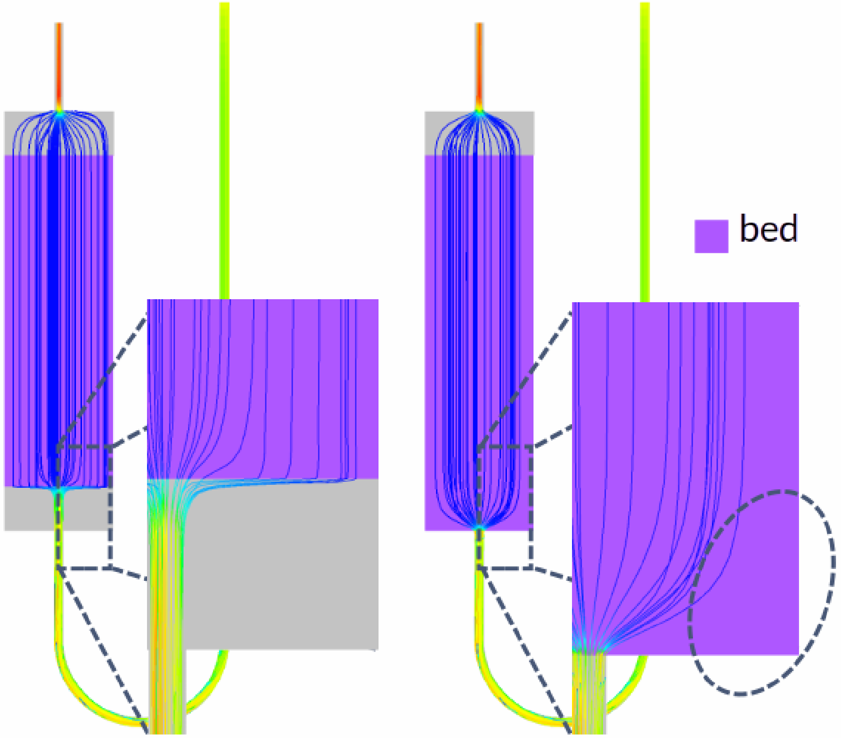


Vaporized Argon from cryostat must be liquefied, the junction are the reinjection position at pipeline

- The worst conditions: evaporation rate of 0.683 kg/s at a pressure of 410 kPa.
  - Low flow rate and height pressure pump are necessary!

CFD can helps to estimated important information to find the best setup in Lab Scale LAr Filter and answer some questions about the operability of Lab Scale facility

- In Purify filter, CFD helps to:
  - Find dead areas – Insert a gap before particle bed;
  - Calculated the pressure drop – ensure the pump mass flow;
- In Argon storage tank, CFD helps to:
  - Estimate the time frequency to measure the Ar concentration
- Solve Navier-Stoke equation



$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho v) = 0$$

$$\frac{\partial \rho v}{\partial t} + \nabla \cdot (\rho v v) = -\nabla p + \nabla \cdot \tau_{eff} + \rho g + F_{porous}$$

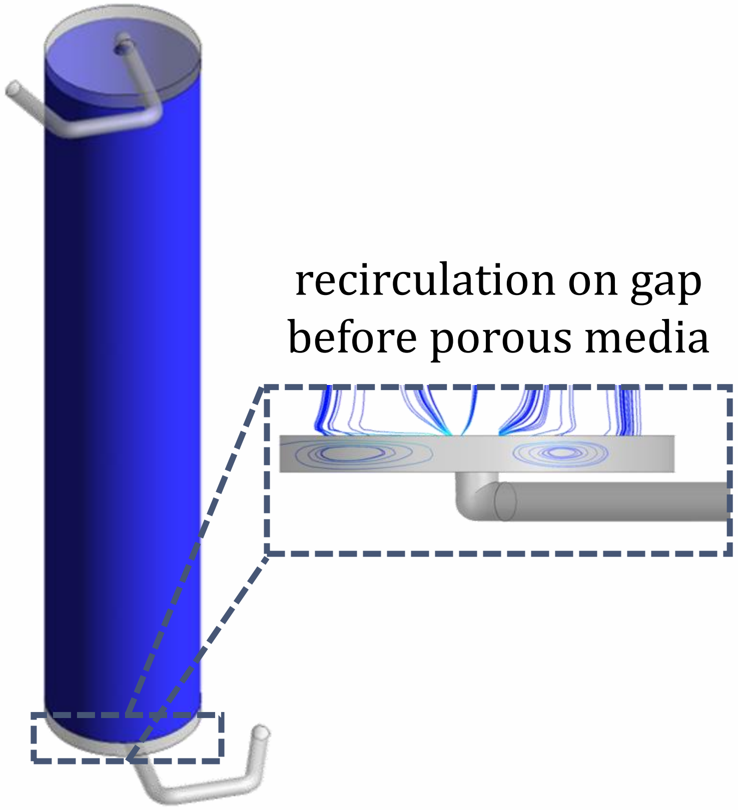
$$\frac{\partial \rho \alpha}{\partial t} + \nabla \cdot \left(\rho v \alpha - \Gamma_{Ar-N_2} \nabla \alpha\right) = 0$$

PuLArC CFD simulation

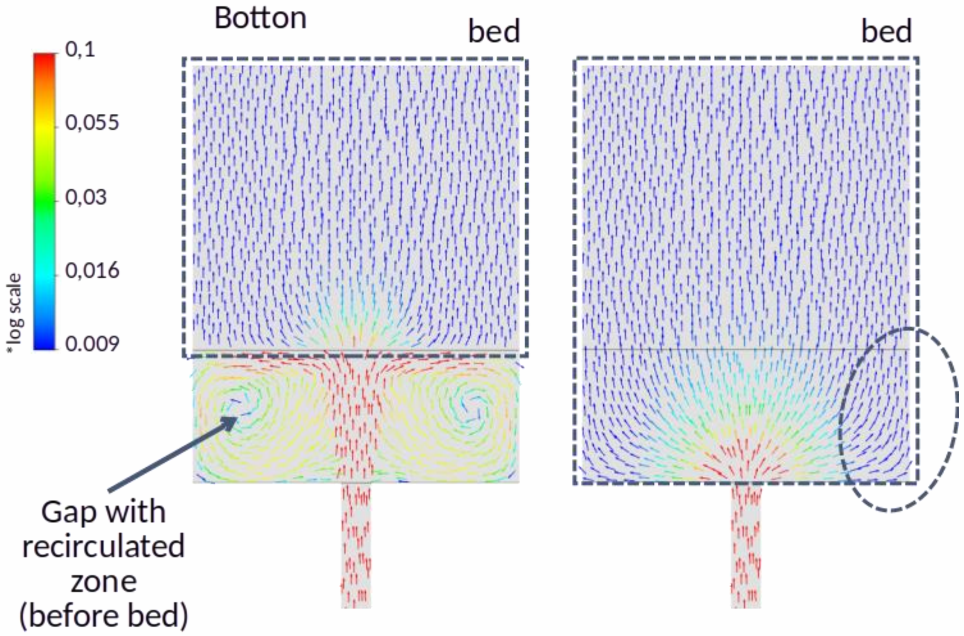
CFD in-house filter

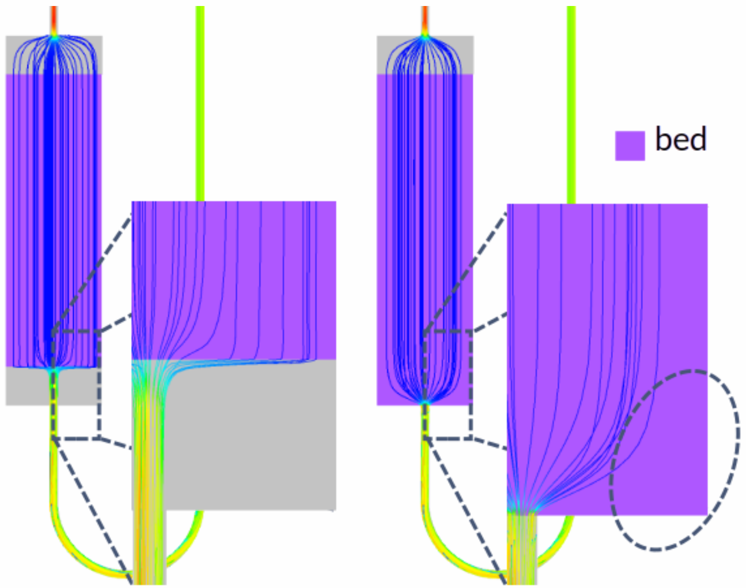
- **Pu**rification **L**iquid **Ar**gon **C**ryostat (PuLArC)

recirculation on gap
before porous media



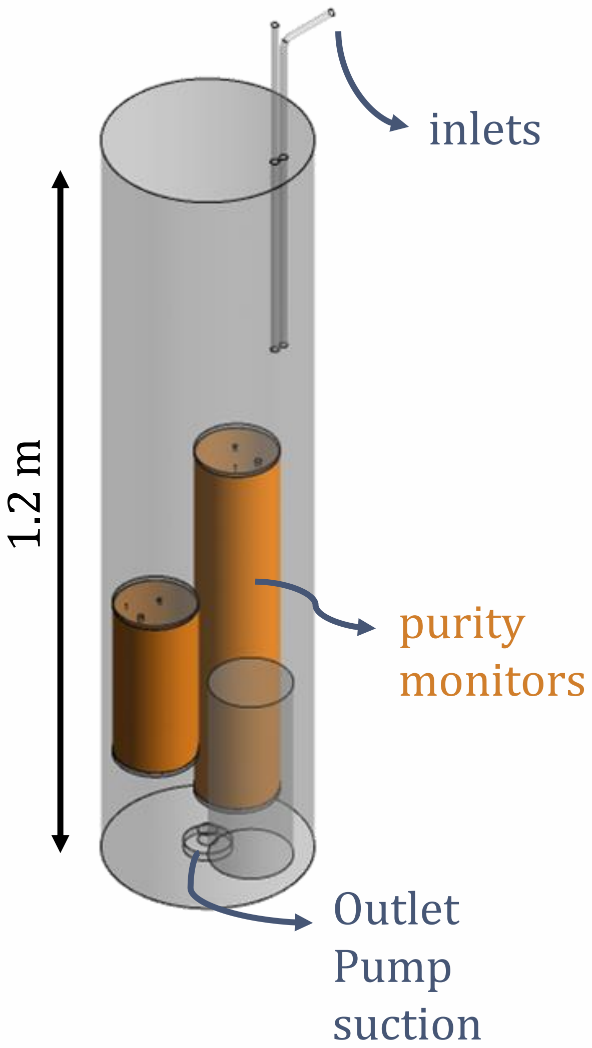
filtering medium
(porous domain)





PuLArC CFD simulation

CFD tank simulation



Geometry Simplification:

- Purity monitor considered hollow cylinders;
- Metallic meshes around the purity monitors modeled as porous media;

Computational Mesh:

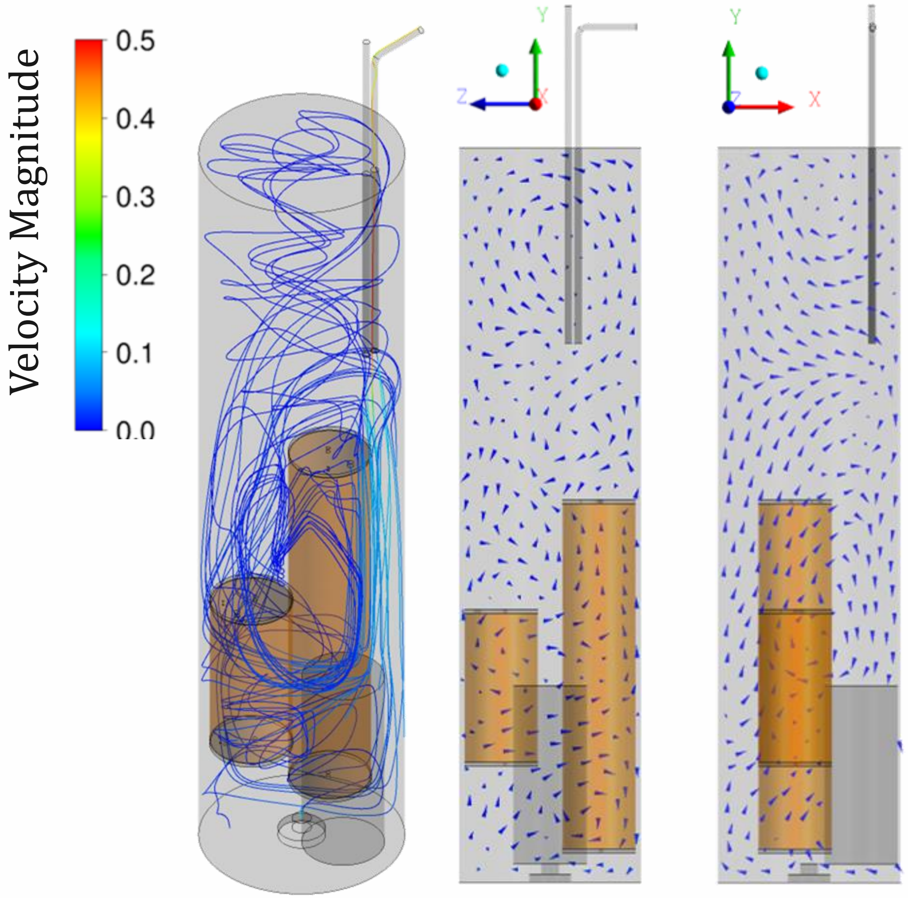
- Hybrid (hexahedral + polyhedral);
- ~ 2 million elements.

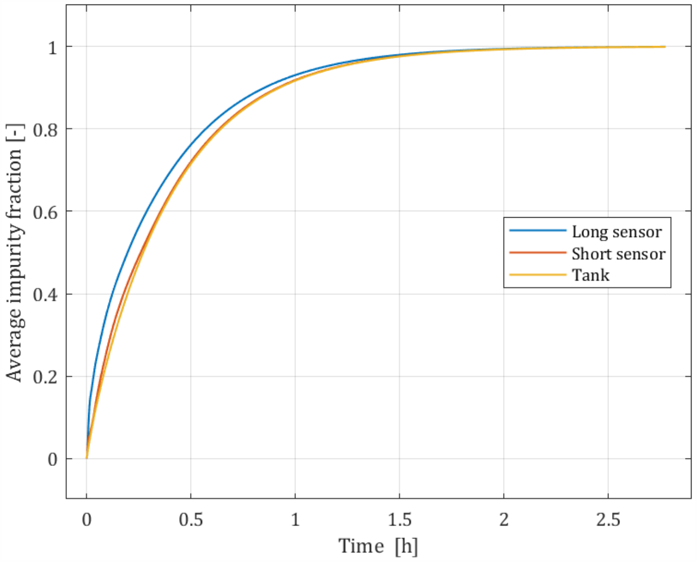
Boundary conditions:

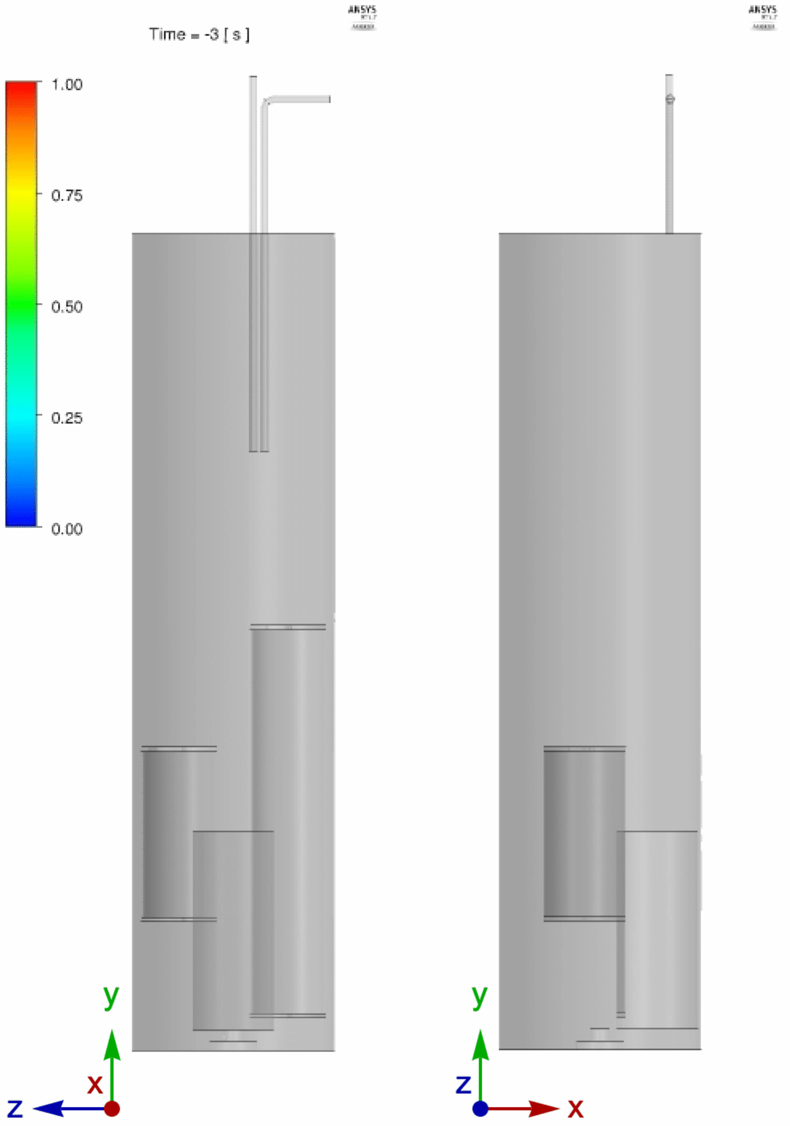
- Inlet of 4.3 L/min of liquid argon;
- Adiabatic walls.

PuLArC CFD simulation

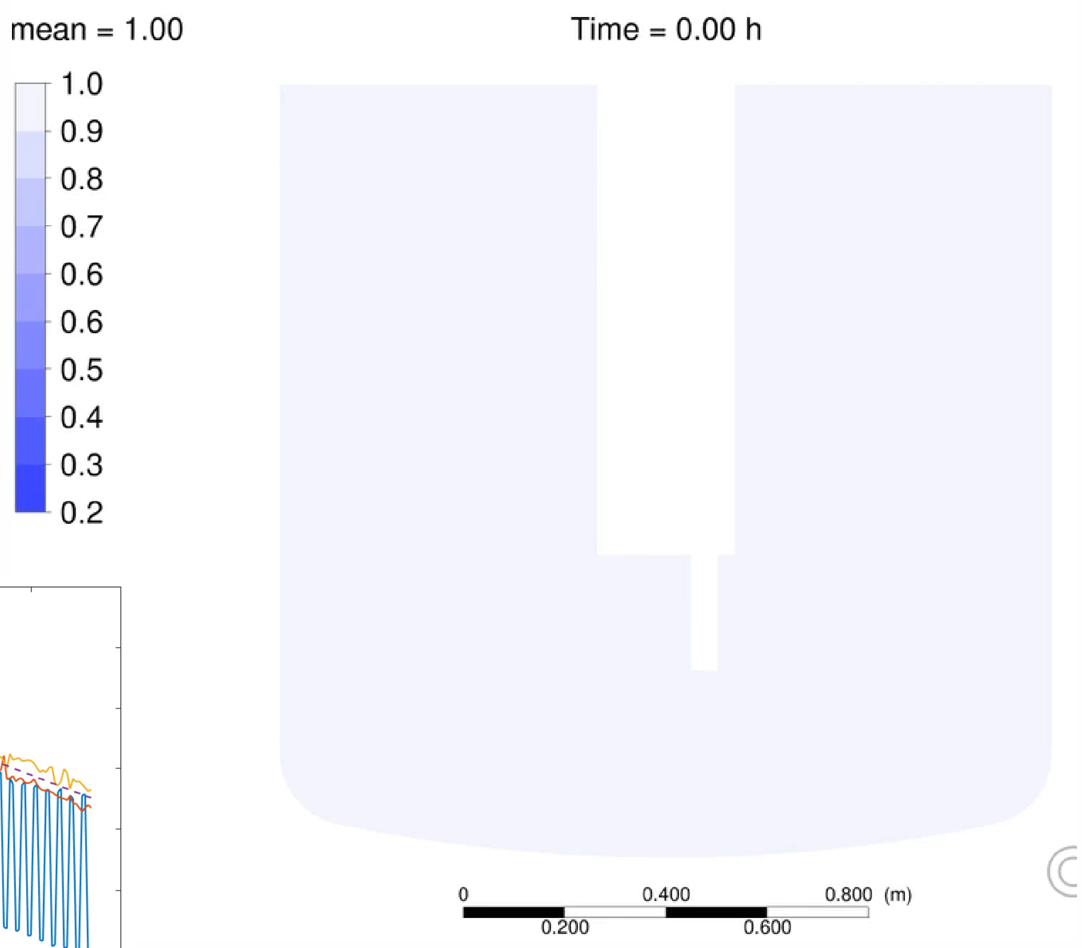
CFD tank simulation

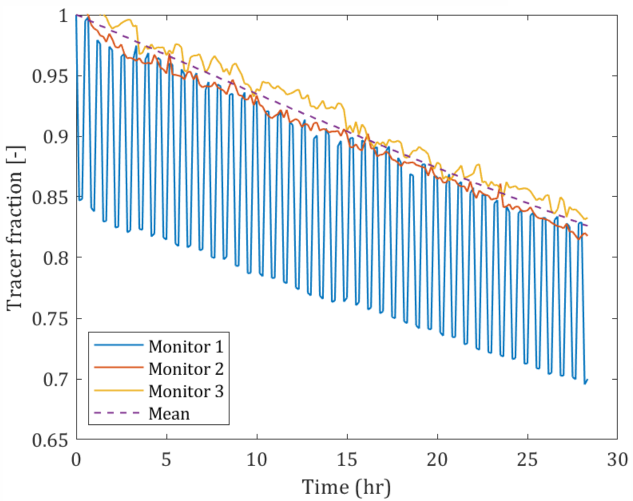






- **I**ntegrated **C**ryogenic **E**xperiment for **B**eam **E**nergy **R**esearch and **G**eneration (ICEBERG) Cryostat
- Simulation conditions:
  - Virtual filter inlet and outlet in the same position;
    - Periodic change of boundary;
  - Initial tracer concentration equal 1;
  - As filter inlet – Tracer fraction local flow value;
  - As filter outlet – Tracer fraction value equal to zero;
- 28 hours simulation;





Modeling and Simulation of Nitrogen Adsorption

Objectives

- Objective:
  - The main objective of this study is to use mathematical models to describe the adsorption phenomenon and estimate important information to capture the contamination of N_2 in Ar.

- Steps:
  - Development of a numerical code to solve the mathematical model and describe the adsorption of impurities in a fixed bed column;
  - Validation of the code with data from literature;
  - Simulate the experimental laboratory-scale filtration facility, evaluating the adsorption behavior of different solid particles and comparing their performances capturing nitrogen from liquid argon.

```
function dPhidt = funFERMI_Recirculation(~, phi, qmax, KLA, betaA)
% Initialize derivative vectors
dCAdt = zeros(N, 1);
dYAdt = zeros(N, 1);
dqAdt = zeros(N, 1);

% Extract variables from the state vector 'phi'
CA = phi(1:N);              % Concentration of component A
YA = phi(N+1:2*N);          % Adsorbed concentration
qA = phi(2*N+1:3*N);        % Equilibrium adsorption
V = phi(3*N+1);             % System volume
CAt = phi(3*N+2);           % Inlet concentration

% Set inlet concentration at z = 0
CA(1) = CAt;

%-- Internal Points (Numerical Solution) --------------------------------%
for i = 2:N-1
    % Calculate spatial derivatives
    dCAdz = (2*alpha*CA(i) + (0.5 - alpha)*CA(i+1) - (0.5 + alpha)*CA(i-1)) / dz;
    d2CAdz2 = (beta*CA(i+1) + beta*CA(i-1) - 2*beta*CA(i)) / dz^2;

     % Derivated R-P isotherm for monocomponent
    dqAdYA = qmax*( 1 + KLA*YA(i)^betaA - KLA*betaA*YA(i)^betaA) / (1 + KLA*YA(i)^betaA)^2;

    % Adsorption rate
    dYdt = ((ap * kL / ep) * (CA(i) - YA(i))) / (1 + rhoS*((1-ep)/ep)*dqAdYA);

    % Calculate time derivatives for concentration and adsorption
    dCAdt(i) = DL * d2CAdz2 - vz * dCAdz - ((1 - eb) / eb) * ap * kL * (CA(i) - YA(i));
    dYAdt(i) = dYdt;
    dqAdt(i) = dqAdYA * dYdt;
end

%-- Tank Equation -------------------------------------------------------%
Qin = Q - sum(dqAdt(2:N-1)) * msol / rhoL / (N-2);  % Adjust inflow rate
tau = V / Q;                                        % Time constant for outflow
tauin = V / Qin;                                    % Time constant for inflow
dVdt = Qin - Q;                                     % Volume change rate
dCAindt = ((CA(N) / tauin) - (CAt / tau)) - CAt * dVdt / V;
```

Experiments

Liquid argon purification

- Experiments at Unicamp – PuLArC **C**ryostat
  - ~ 1.2 kg of Li-FAU zeolite adsorbent;
  - N_2 contamination reduction: 40 ppm to 0.1-1.0 ppm (1-2 hours of active circulation)
- Experiments at Fermilab – 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)

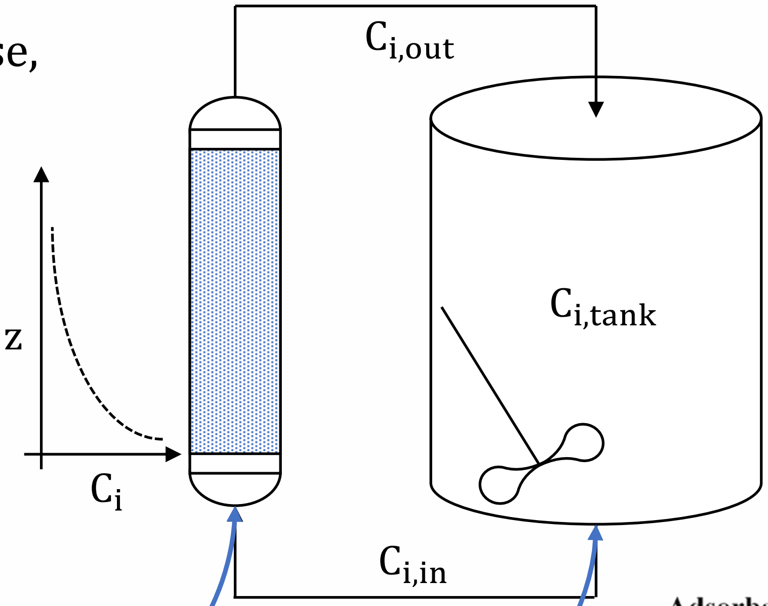
Mathematical Model

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

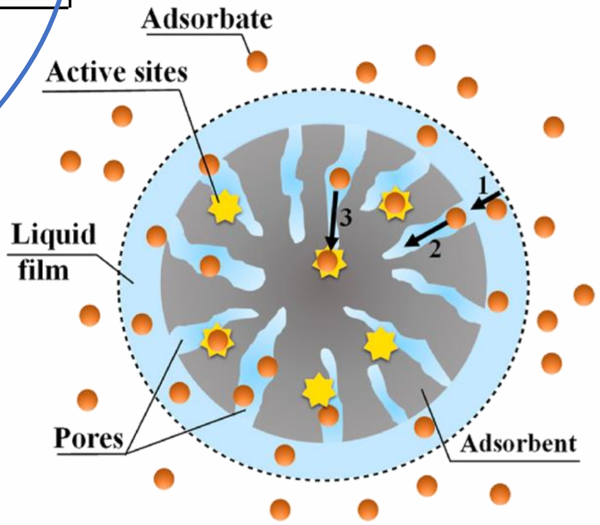
$$\frac{\partial Y_i}{\partial t} = \frac{K_L a_V}{\epsilon_p}(C_i - Y_i) - \rho_S \frac{\left(1-\epsilon_p\right)}{\epsilon_p}\frac{\partial q}{\partial t}$$

Solid-phase

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

Well-mixed tank

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



Intraparticle diffusion adsorption [1]

Mathematical Model

Closure equations

Adsorption isotherm:

Redlich-Peterson:

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

K_{RP}: constant related to adsorption capacity (m3/kg)

g: 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 :

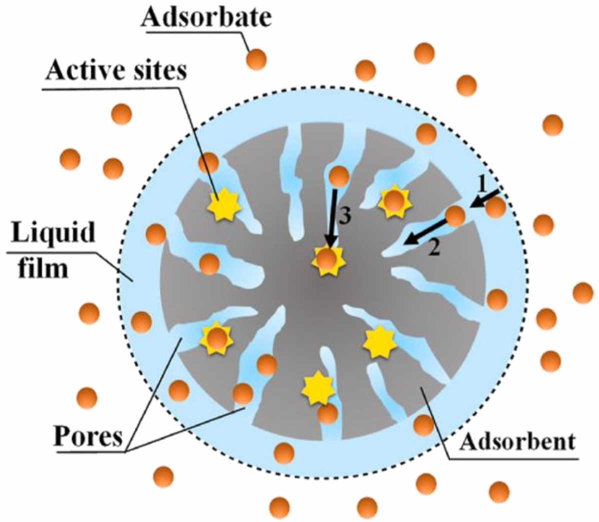
$$\frac{1}{K_L} = \frac{1}{k_e} + \frac{1}{\epsilon_p k_i} \qquad Sh = \frac{k_e d_p}{D_L} \qquad \frac{Sh}{Sc^{1/3} Re_m} = 0.91 Re_m^{-0.51} \qquad Sc = \frac{\mu_m}{\rho_L D_L}$$

ϵ_p: intraparticle porosity

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

D_L: molecular diffusivity of N_2 in Ar

τ: particle tortuosity



Intraparticle diffusion adsorption [1]

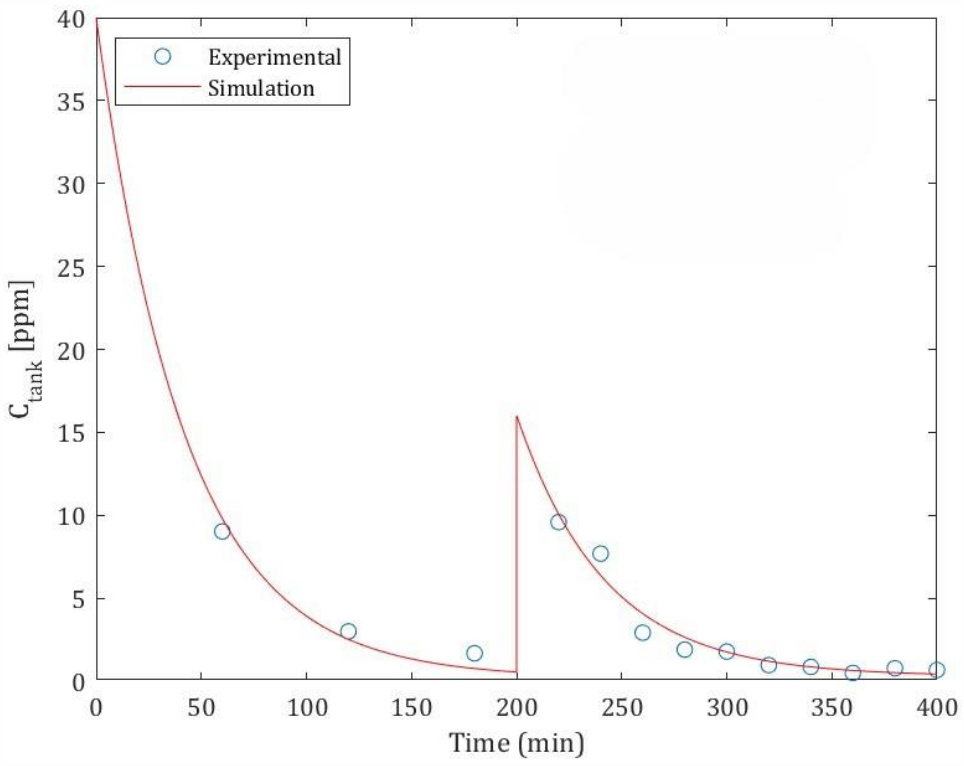
Model Solution

PuLarC parameters optimization

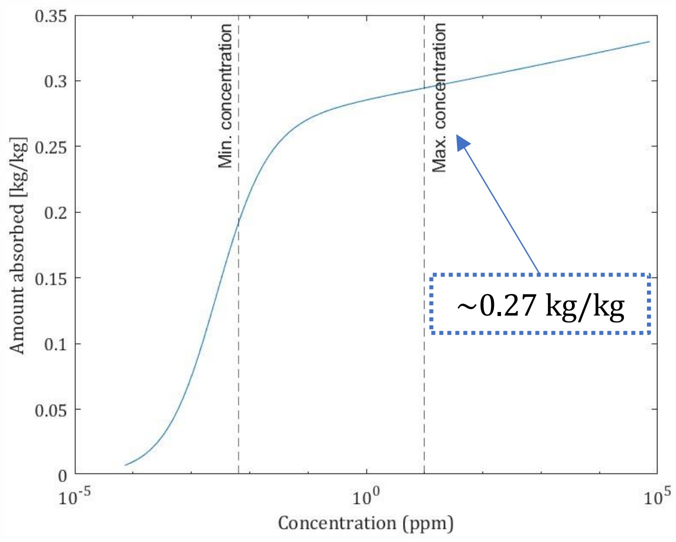
PuLarC simulation conditions:

- 2 cycles (media is not regenerated);
  - $C_{N_2,1} = 40$ ppm
  - $C_{N_2,2} = 16$ ppm
- $Q = 4$ L/min

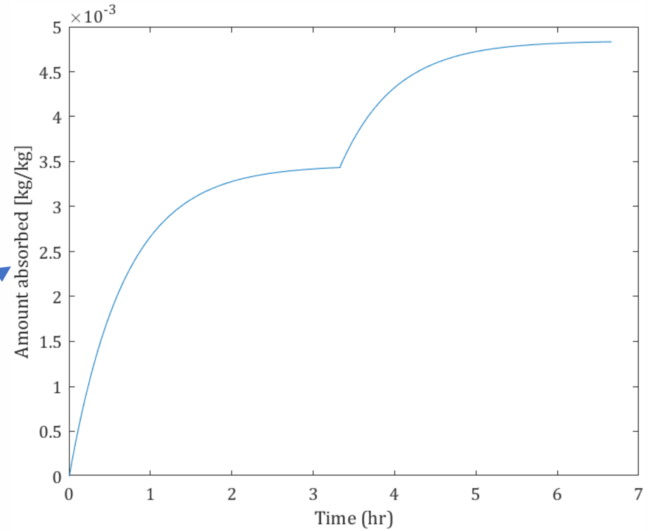
- tank volume: 0.09 m^3
- media level: 29 cm
- bed diameter: 95.5 cm
- bed porosity: 0.38
- $\rho_S = 936$ kg/m^3
- $d_p = 2$ mm



Regressed isotherm



~0.27 kg/kg



Adsorbent is far to saturation !!!!

Model Solution

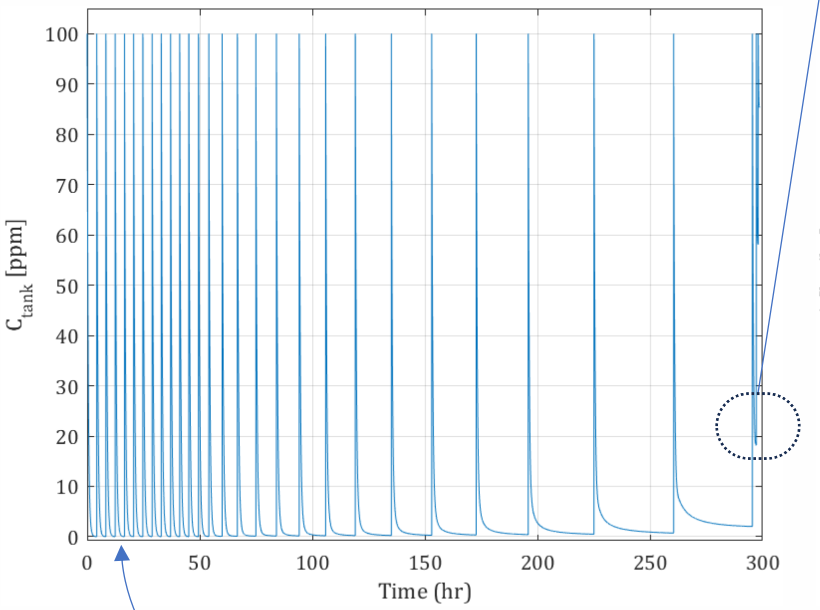
PuLarC simulation scenarios

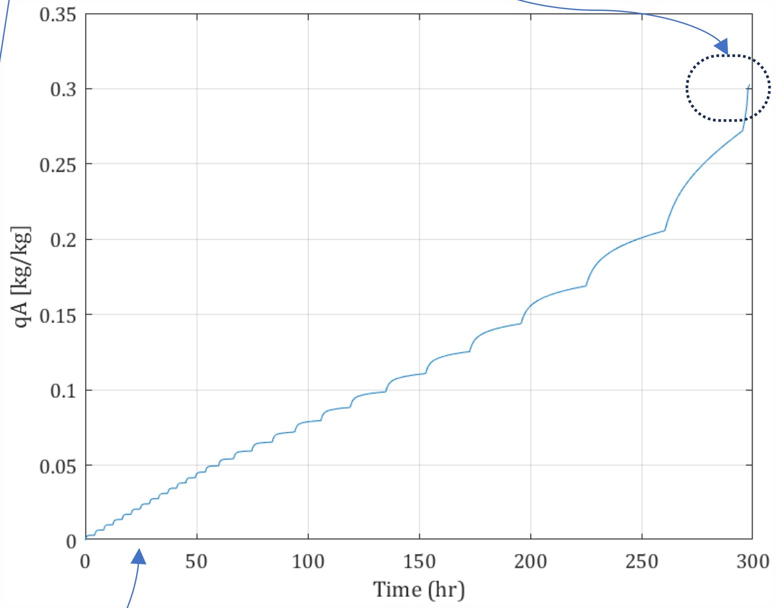
Simulation conditions:

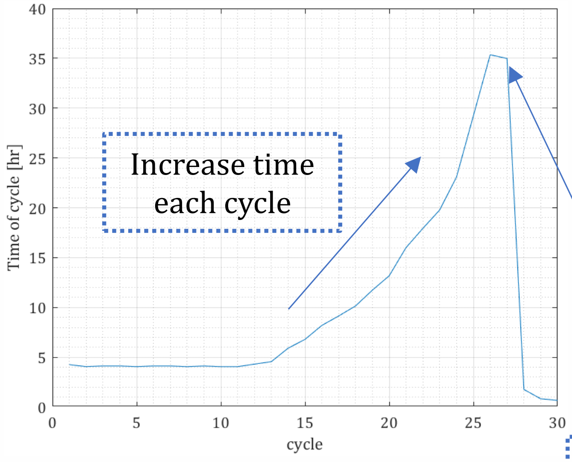
- Number of cycles to achieve the adsorbent saturation ???
- 100 ppm for each cycle;

28th cycle

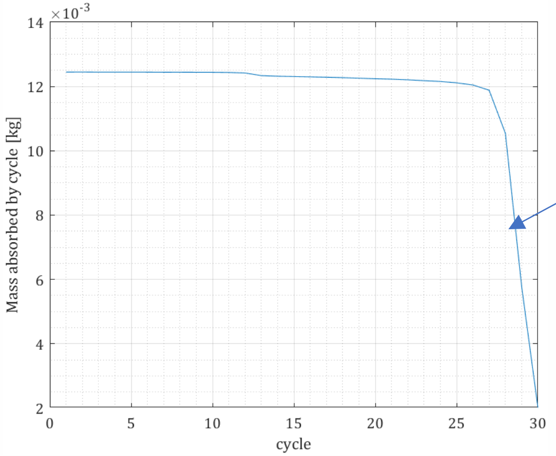
Max adsorption are reached







Increase time each cycle



After 28th cycle, no more mass can be adsorbed

Simulation stops when:

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

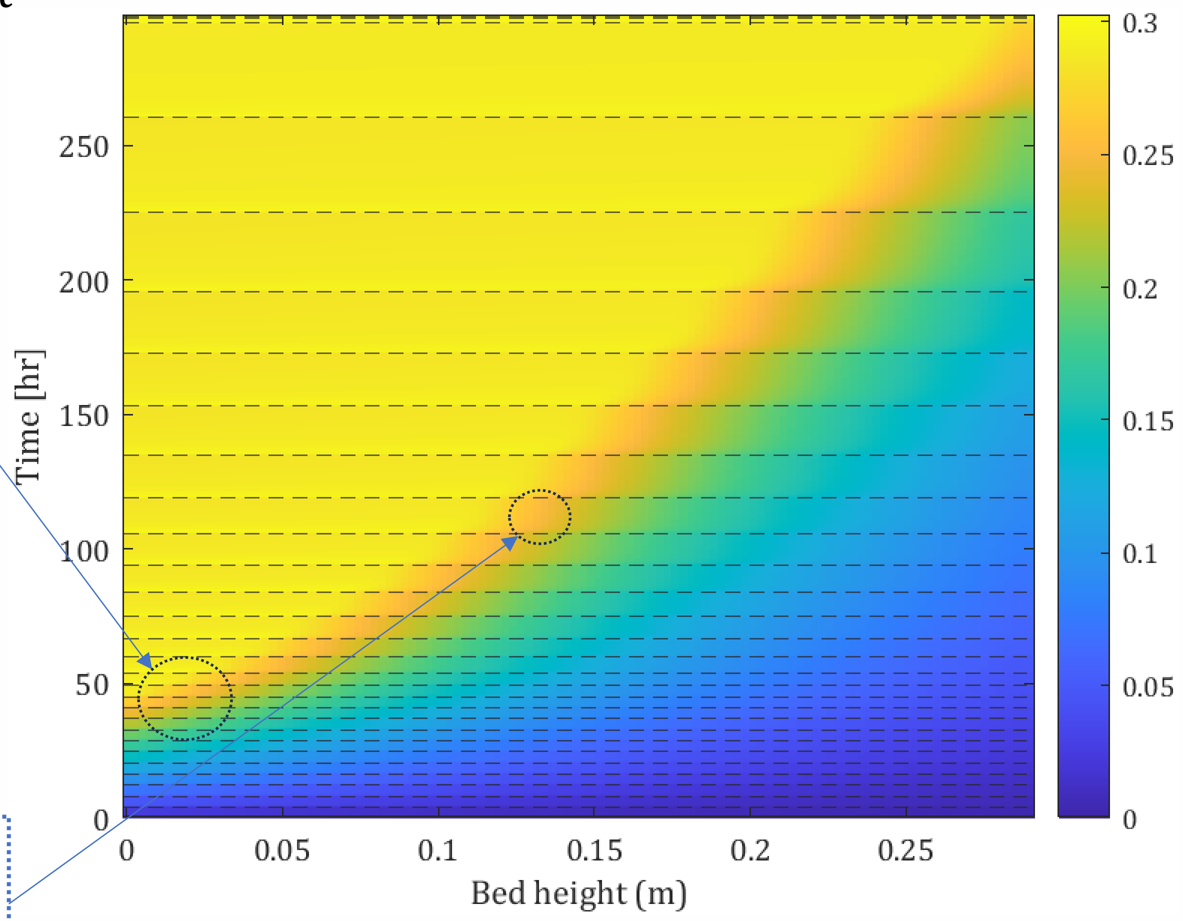
Model Solution

PuLarC simulation scenarios

Simulation conditions:

- Number of cycles to achieve the adsorbent saturation in all bed???
- 100 ppm for each cycle;

Colormap of N_2 in solid phase.
Dashed line represent the cycles



First centimeters of solid phase only saturated after ~45hr, or ~11th cycle

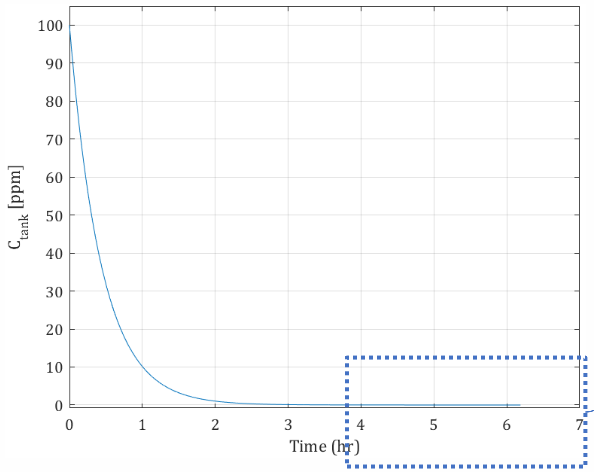
50% of bed are sutured, ~19th cycle

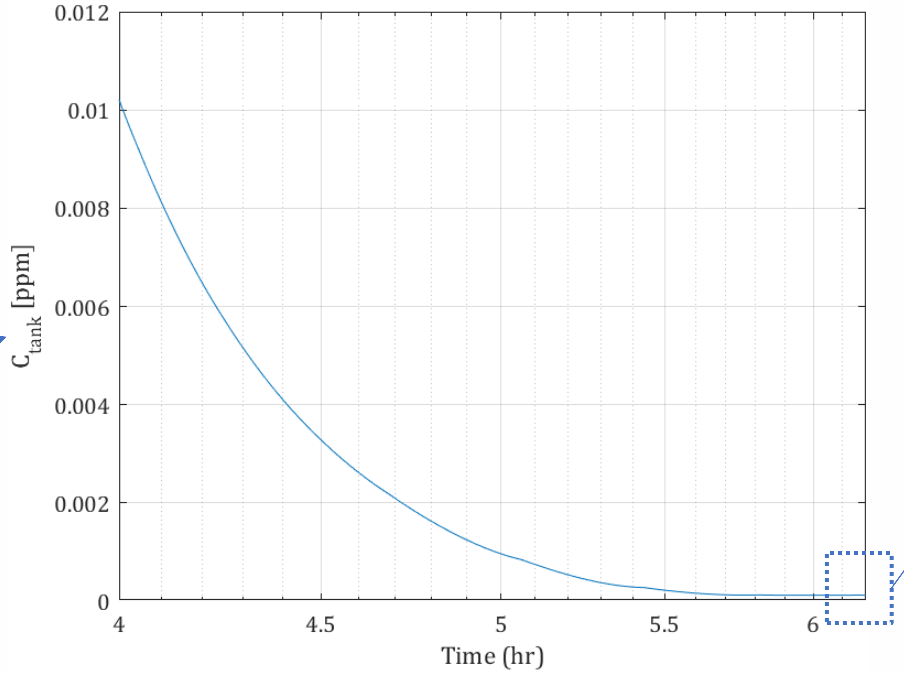
Model Solution

PuLarC simulation scenarios

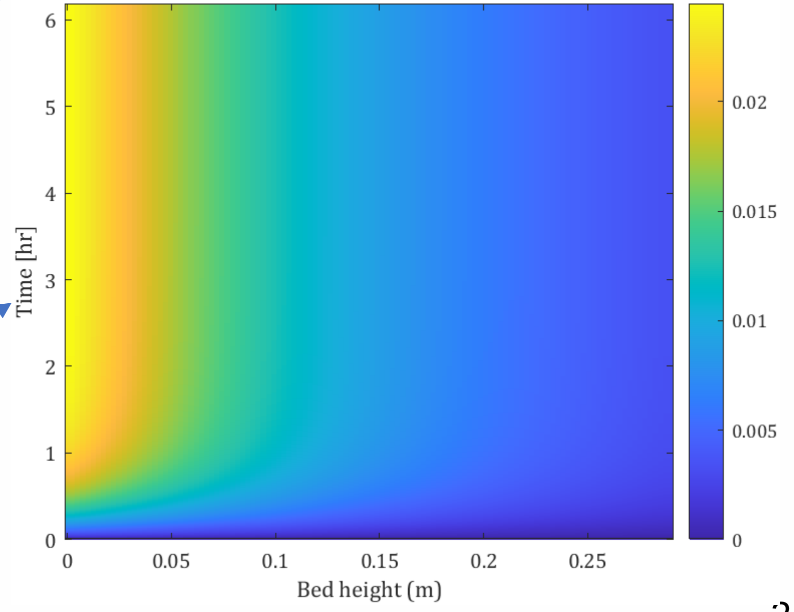
Simulation conditions:

- Minimum possible impurity??
- Initial ⟶ 100 ppm
- Target ⟶ 100 ppt





Target occurs at 6.2 hr



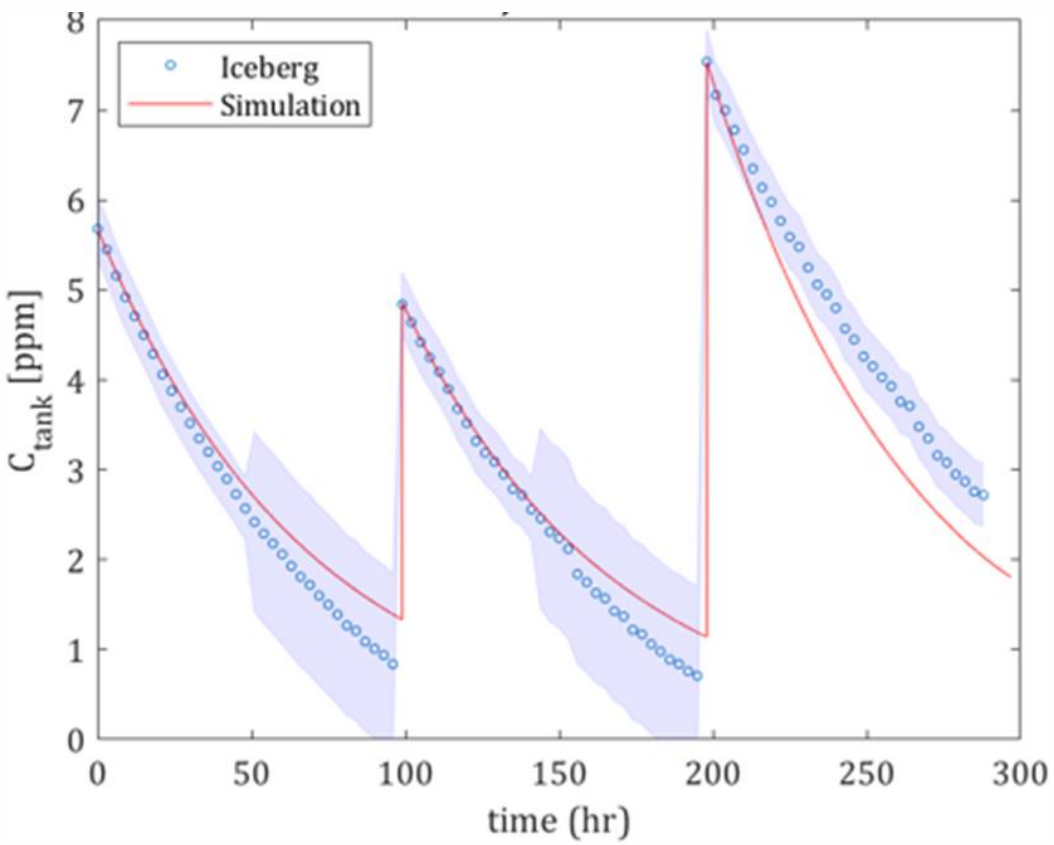
Solid is far to saturate, but system don't have concentration to fast adsorption

Model Solution

Iceberg simulation conditions:

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

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

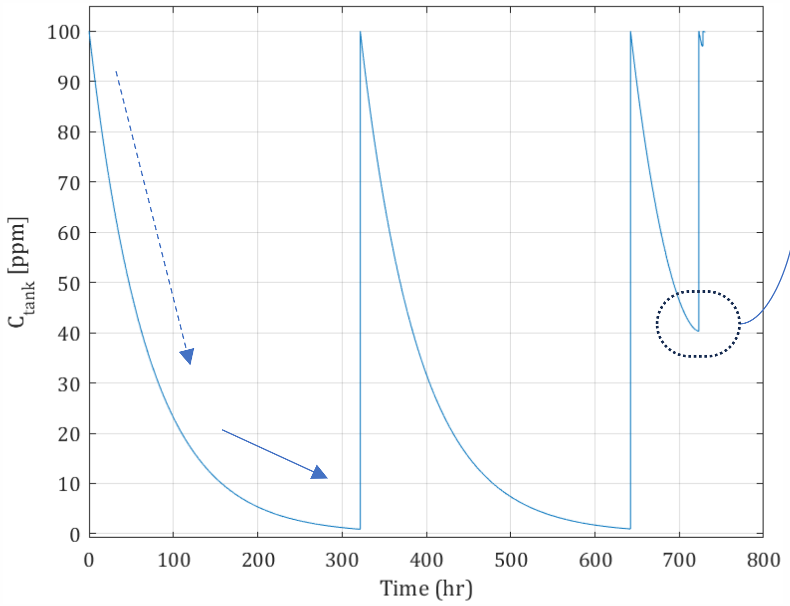


Model Solution

ICEBERG simulation scenarios

Simulation conditions:

- Number of cycles to achieve the adsorbent saturation ???
- 100 ppm for each cycle

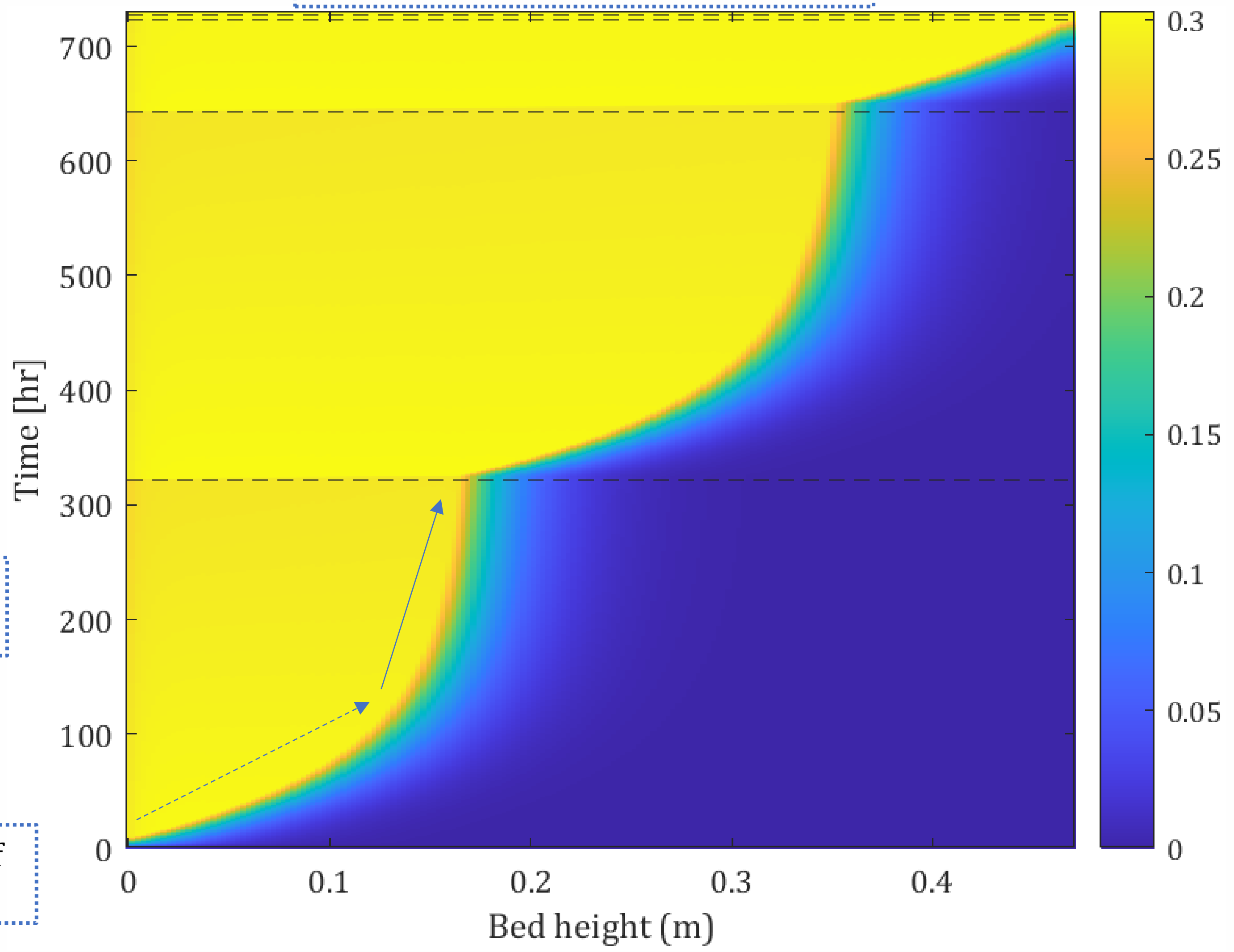


3^{th} cycle

~0.9 kg of N_2 are adsorbed

~0.3032 kg/kg of particle

Colormap of N_2 in solid phase. Dashed line represent the cycles

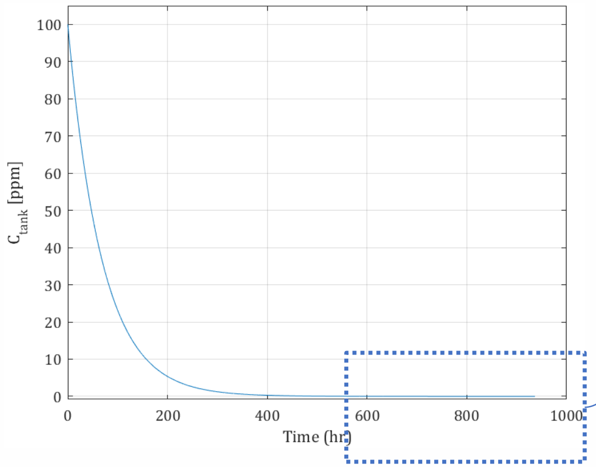


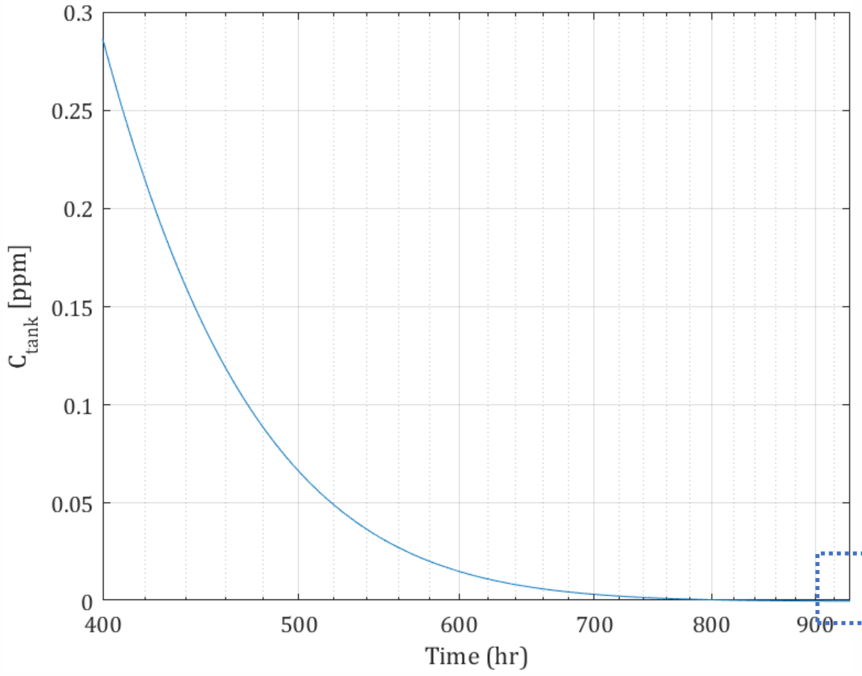
Model Solution

ICEBERG simulation scenarios

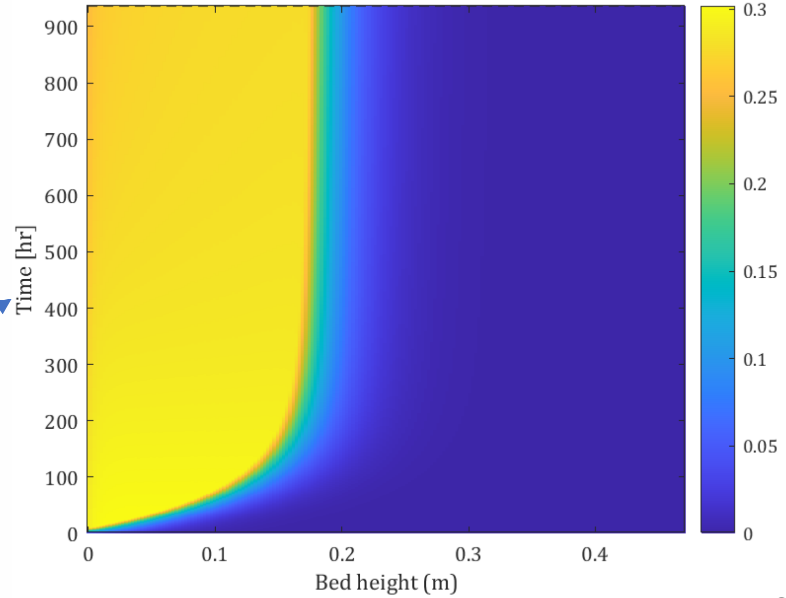
Simulation conditions:

- Minimum possible impurity??
- Initial ⟶ 100 ppm
- Target ⟶ 100 ppt





Target occurs at 934 hr



1/3 of the solid is saturated, but the system does not have enough concentration for fast adsorptio

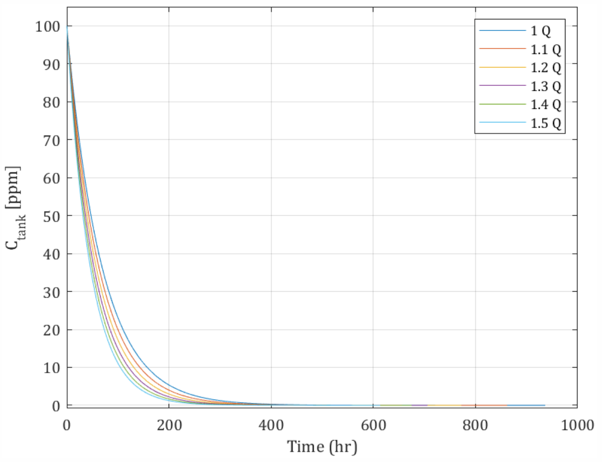
Model Solution

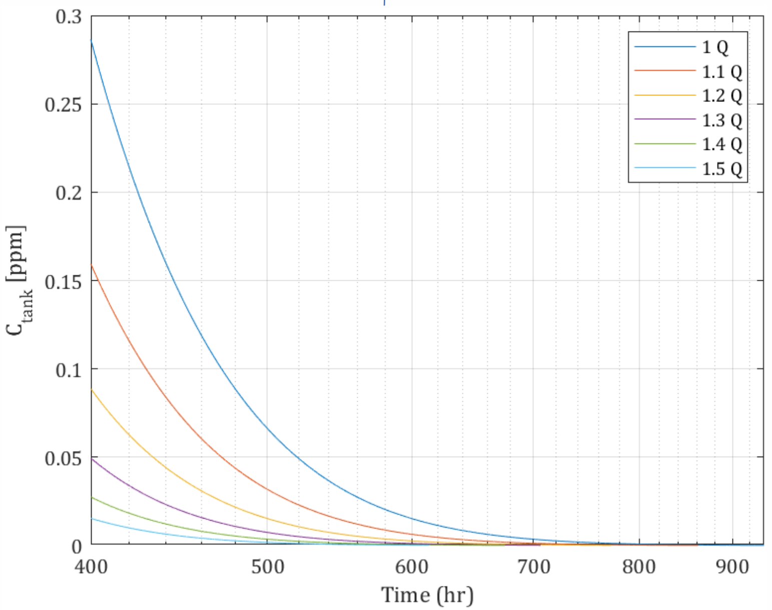
ICEBERG simulation scenarios

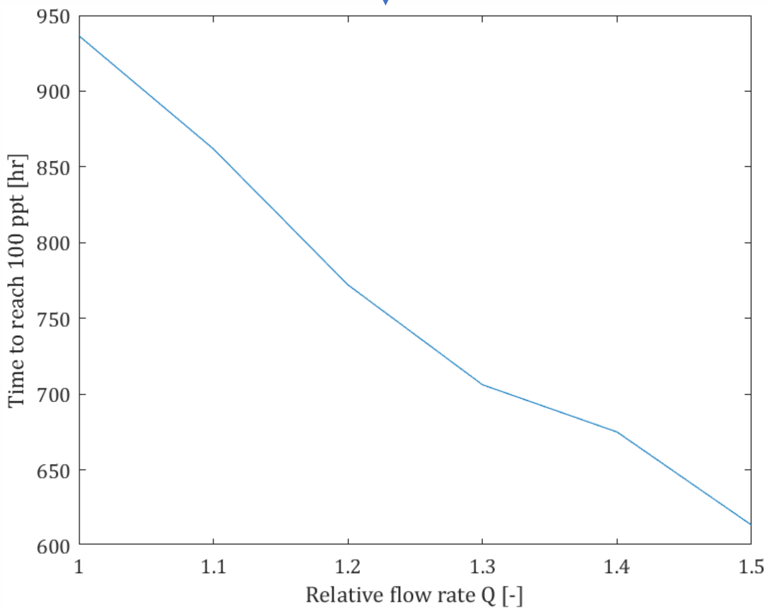
Simulation conditions:

- Minimum possible impurity??
- Initial ⟶ 100 ppm
- Target ⟶ 100 ppt
- Its possible to be faster????

1/3 of the solid is not saturated, there is still more capacity. Increasing recirculation will decrease the time.



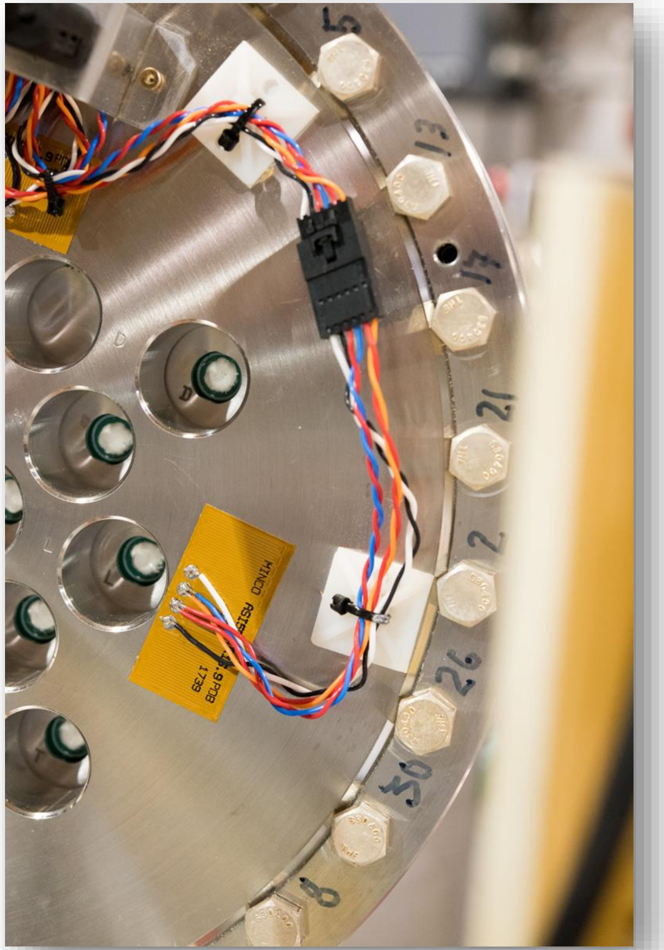




Final Remarks

Conclusion and Outlook

- About the LAr circuit in criostat
  - LAr Circuit:
    - Pump characteristics;
    - Mass flow rate in cryostat fill conditions;
    - Mass flow rate with N pumps and filter operation;
  - GAr circuit
    - Evaporation rate and Pump operation
- CDF analysis shows the mean path of LAr fluxes in filters and storage tank in PuLArC and ICEBERG experimental setups
- 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;



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LAr Purificarion for LNFB DUNE

Thank You for your attention!