Modeling and Structural Optimization of Adsorption-Based Systems for the Removal of Carbon Dioxide from the Air*

Mayra G Gonzalez-Ramirez[a], Tiras Y. Lin[b], Thomas Roy[b], Thomas Moore[b], Du Thai Nguyen[b], Pratanu Roy[b], Sarah Baker[b], Lorenz Biegler[a], Grigorios Panagakos[a,c]

[a]Department of Chemical Engineering, Carnegie Mellon University, [b]Lawrence Livermore National Laboratory, [c]National Energy Technology Laboratory.

Motivation

Goal

- Design optimal devices for adsorption-based Direct Air Capture (DAC) technologies within the Topology Optimization (TopOpt) framework that are suitable for scale-up.

State-of-Art

- The use of DAC is imperative to handle hard-to-avoid CO_2 emissions [1].
- Contactor design improvements paired with sorbent material development is necessary for optimal devices that can be scaled-up.
- TopOpt has been used successfully for optimal structures in complex systems like flow batteries [2] and porous electrodes [3].
- **Computational Fluid Dynamics (CFD) is the only tool that can quantitatively predict the effect of design parameterization on device performance.**
- **Combining CFD and TopOpt is a powerful tool for seeking optimal device design for a given material.**

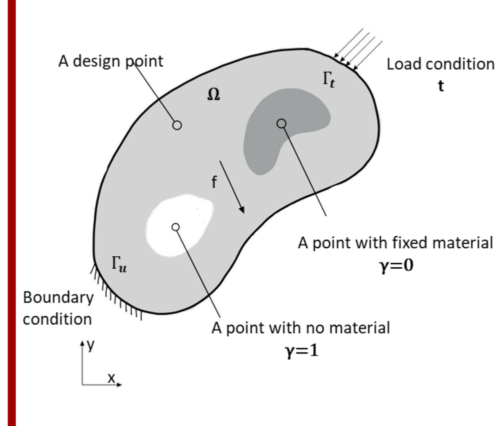
Approach

- Topology Optimization is a mathematical framework used to determine the optimal material distribution within a given design domain with respect to a certain objective function (e.g. adsorption rate).

Challenges

- Unique application of topology optimization.
- Considering multi-scale, packed bed adsorbers with complex, coupled physics within the Topology Optimization Framework.
- Defining representative nondimensional numbers that are appropriate for scaling up.
- Uniqueness of solution and global optimization not ensured a priori.

Topology Optimization Framework

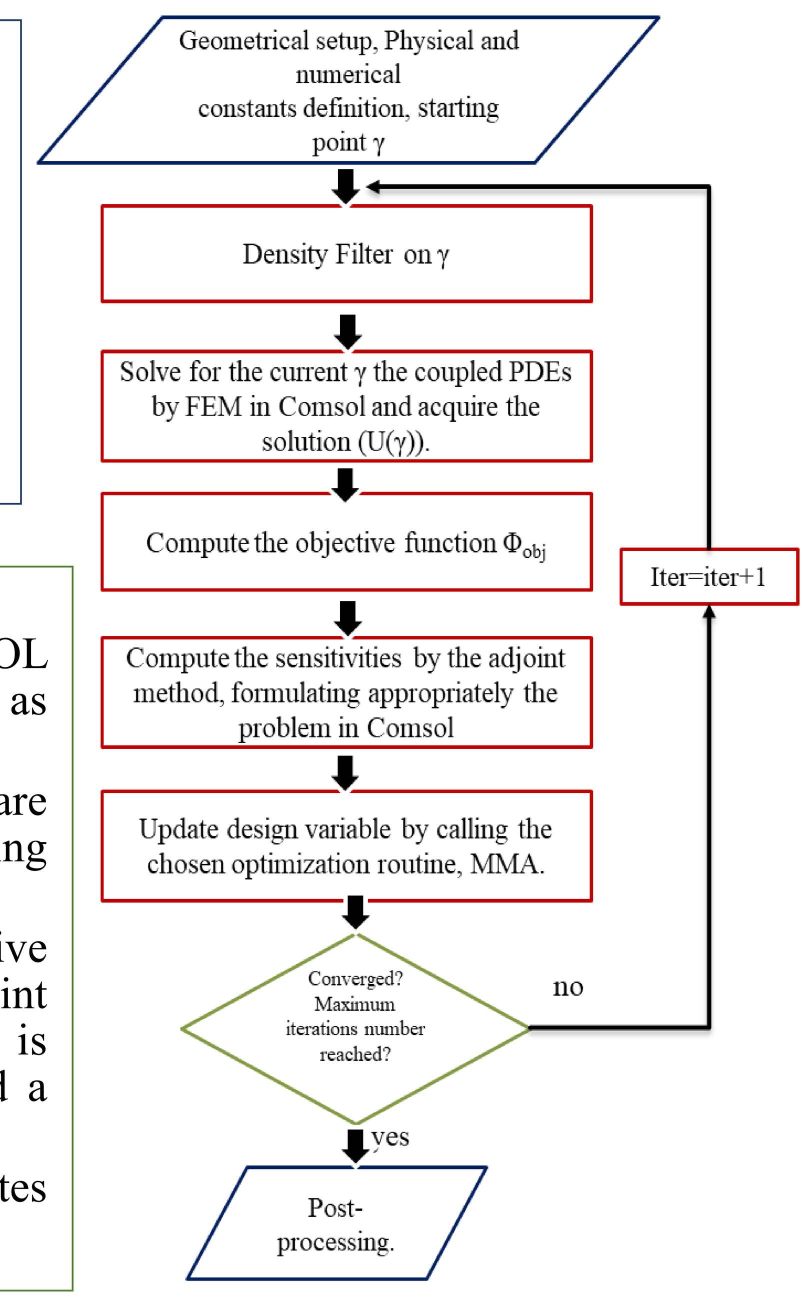


Concept

- The computational domain Ω with flow boundary conditions on ∂Ω contains material distributed as solid and void, based on the local $\gamma(\mathbf{r})$ values of each element. The local inverse permeability $\alpha(\mathbf{r})$ in the N-S equations is parameterized using the local $\gamma(\mathbf{r})$ [4], ranging between the limits of the open pore $\gamma = 1$ and dense material $\gamma = 0$.

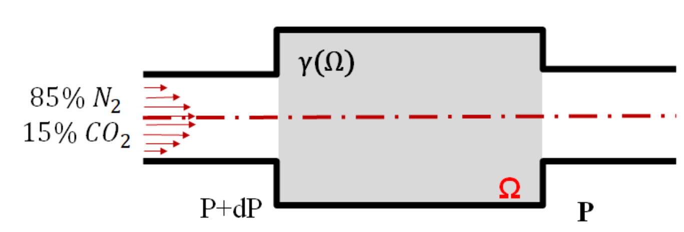
Methodology

- The iterative algorithm is carried out in the software COMSOL Multiphysics through LiveLink™, the COMSOL/MATLAB interface, as follows:
  - The Navier Stokes and mass transport equations at the k^{th} iteration are solved with a specific "guess" of the design variable $\gamma^k(\mathbf{r})$ producing the finite element solution $u^k(\gamma(\mathbf{r}))$.
  - Sensitivity analysis is performed, where the gradient of the objective function and associated constraints are evaluated as well as the adjoint equations for the Lagrange multipliers μ^k. The design variable is updated using the method of moving asymptotes (MMA) to yield a new guess γ^{k+1}.
  - The design variable is updated using the method of moving asymptotes (MMA) to yield a new guess γ^{k+1}.



Problem Statement

We wish to optimize one step in a post-combustion Pressure Swing Adsorption (PSA) process [5]. A feed containing N_2 and CO_2 is fed to the bed holding the sorbent.



The basis of this work is the formulation proposed by Olesen et al. [6]. The optimal design in [6] involves a complex γ distribution within the reactor. The algorithm optimizes the average reaction rate $\Phi(\gamma) = -\langle k(\gamma)c_i\rangle_\Omega$ by obtaining the optimal porosity field $\gamma(r)$, where $-\langle k(\gamma)c_i\rangle_\Omega$ is the kinetic reaction term. In our case, instead of reaction we treat adsorption instead for the DAC packed bed contactor. The overall constrained topology optimization model is:

Max adsorption rate $\min_{\gamma} \Phi(\gamma) = -\langle k(\gamma)c_i\rangle_\gamma$

$$s.t. \int_\Omega \gamma(x)dx - |\Omega| \le 0 \quad \textbf{Volume constraint}$$

$$\rho(\boldsymbol{u}\cdot\nabla)\boldsymbol{u} = -\nabla p + \eta\nabla^2\boldsymbol{u} - \alpha(\gamma)\boldsymbol{u} \quad \epsilon\,\Omega \quad \textbf{N-S}$$

$$\nabla\cdot\boldsymbol{u} = 0 \quad \epsilon\,\Omega \quad \textbf{Continuity}$$

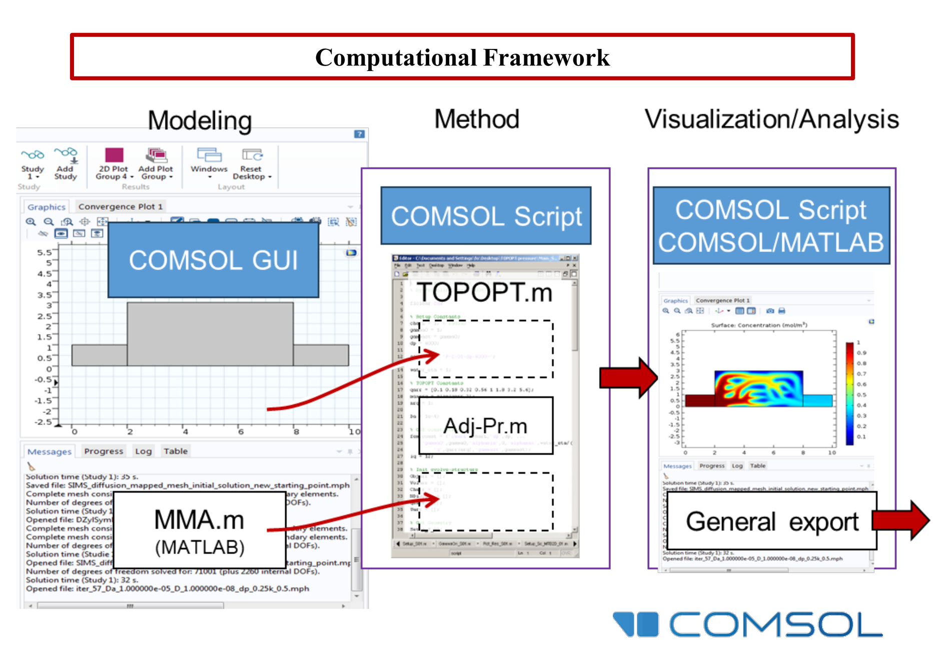
Single or Dual-site Langmuir Isotherm

$$(1-\varepsilon_b)\rho_s\, k_i(q_i^* - q_i) = \frac{\partial(uc_i)}{\partial x}$$

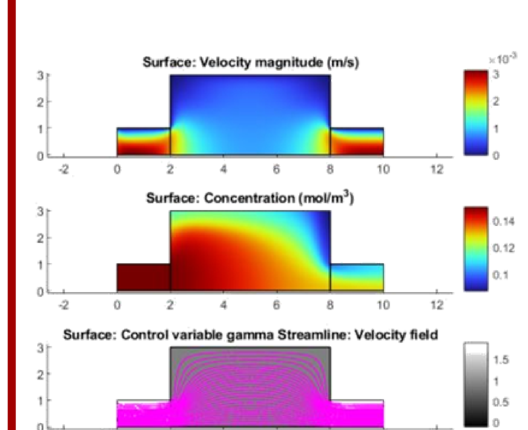
$$q_i^* = \frac{q_{1i}^* b_{1i} c_i RT}{1+\sum_j b_{1j}c_jRT} + \frac{q_{2i}^* b_{2i} c_i RT}{1+\sum_j b_{2j}c_jRT}$$

$$q_{mi}^s = k_{mi}^1 + k_{mi}^2 T, \qquad b_{mi} = k_{mi}^3 \exp\left(\frac{k_{mi}^4}{T}\right), \qquad m = 1,2$$

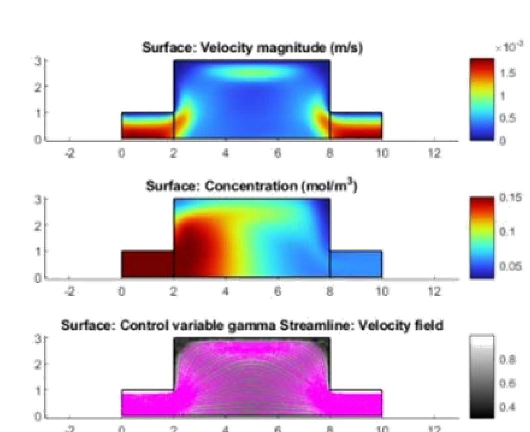
$$0 \le \gamma(\boldsymbol{r}) \le 1 \quad \textbf{Design variable}$$



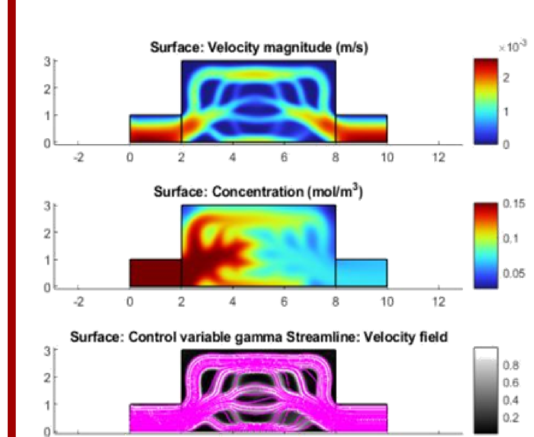
Preliminary Results



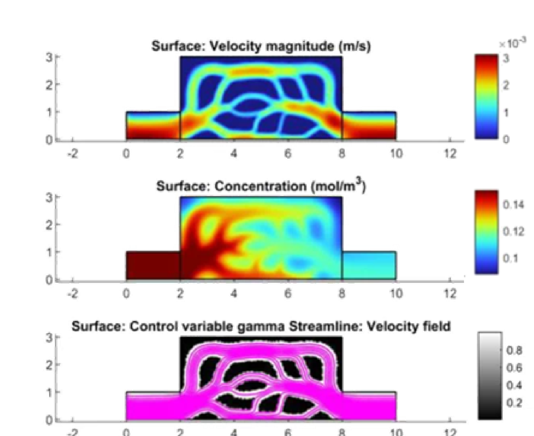
a) First design obtained with Topology optimization framework, k=1, $\Phi(\gamma) = -0.0036$



b) Preliminary design obtained with Topology optimization framework, k=80, $\Phi(\gamma) = -0.0046$



c) Preliminary design obtained with Topology optimization framework, k=180, $\Phi(\gamma) = -0.0050$



d) **Optimal design** obtained with Topology optimization framework, k=231, $\gamma = 0.6591$, ΔP=0.24 Pa, Da = 0.0001 m^2, $\Phi(\gamma) = -0.0065\frac{mol}{m^3\,s}$

Model

- 3207 sec
- 231 iterations

Size

- 3950 finite elements
- Triangular mesh
- 58011 degrees of freedom

- ❖ We obtained an optimal design for the process. The optimal value for the adsorption rate is 0.0065.
- ❖ **The acquired design increases the adsorption rate by 81% compared to the original design.**
- ❖ This value was achieved through the optimal material distribution that enhances the contact between the porous material and the gas stream.

Conclusions

Base
- Catalytic-packed bed reactors can be optimized to share underlying scaling properties. Therefore, this has the potential as a good starting point for packed bed DAC devices.

Our model
- The adsorption rate of the packed bed can be significantly increased by distributing the active porous material within the device using topology optimization.

Technology
- PSA processes could be a good starting point towards DAC design.
- Temperature Swing Adsorption (TSA) should be explored as well to determine the device potential.
- TSA process technology should be explored.

Future work
- TSA/PSA with packed bed physics within the TopOpt framework.
- Determine and evaluate dimensionless numbers that characterize the system

References

[1] Liu, H., Ampah, J. D., Zhao, Y., Sun, X., Xu, L., Jiang, X., and Wang, S. (2022). A perspective on the overarching role of hydrogen, ammonia, and methanol carbon-neutral fuels towards net zero emission in the next three decades. Energies, (16).

[2] Lin T.Y., Baker S.E., Duoss E.B. and Beck V.A. (2022). Topology Optimization of 3D Flow Fields for Flow Batteries. Journal of The Electrochemical Society. 169, 5.

[3] Roy T., Salazar de Troya M.A., Worsley M.A., Beck V.A. (2022). Topology optimization for the design of porous electrodes. Structural and Multidisciplinary Optimization volume 65, Article number: 171.

[4] Deng, Y., Wu, Y., and Liu, Z. (2018). Topology optimization theory for laminar flow

[5] Agarwal, A., Biegler, L. T., and Zitney, S. E. (2009). A superstructure-based optimal synthesis of PSA cycles for post-combustion CO_2 capture. AIChE Journal, 56(7):1813–1828

[6] Olesen, L. H., Okkels, F., and Bruus, H. (2006). A high-level programming-language implementation of topology optimization applied to steady-state navier-stokes flow. International Journal for Numerical Methods in Engineering, 65(7):975–1001.

Contact

mayragug@andrew.cmu.edu gpanagak@andrew.cmu.edu

lb01@andrew.cmu.edu Carnegie Mellon University

***Disclaimer:** This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344 and was supported by Laboratory Directed Research and Development funding under project 22-SI-006. LLNL Release Number: LLNL-PROC-847581.