Carnegie Mellon University

Hydrogen Purification and Storage in Electrolysers

Mathematical Modelling in Chemical Engineering

Carnegie Mellon University – April 2025

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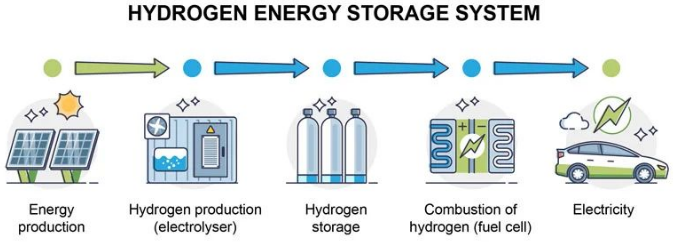
Motivation & Context



- Hydrogen is a key player in clean energy transitions.
- PEM (Proton Exchange Membrane) electrolysers are efficient systems for green hydrogen production.
- However, hydrogen must be purified and safely stored to be usable.
- Understanding the **diffusion behaviour through membranes** enables improved design and operation of these systems.
- Modelling these systems mathematically helps predict and optimise performance before experimental deployment.

Project Objective

- Develop a simulation model for hydrogen diffusion and purification through a PEM membrane.
- Study the effects of:
  - Feed pressure and temperature
  - Membrane thickness
  - Porosity (ϵ) and Tortuosity (τ)
- Compare steady-state and transient diffusion behaviors.
- Optimize the membrane configuration to maximize hydrogen flux.

System Setup

- **Material:** Nafion-based PEM membrane
- **Geometry:**
  - Membrane thickness: 20–300 μm
  - Length: 0.75–50 μm
- **Operating Conditions:**
  - Temperature: 350–500 K
  - Feed Pressure: 100–400 kPa
  - Permeate Pressure: 100 kPa
- **Transport Properties:**
  - Diffusivity (D): 1*10^-5 to 5*10^-10 m^2/s
  - Porosity (ϵ) and Tortuosity (τ) affect effective permeability

Steady-State Diffusion – Governing Equation & Setup

Steady-State Hydrogen Diffusion through PEM

- Governing Equation: $\frac{d^2C}{dx^2} = 0$ Derived from Fick's Second Law under steady-state (□C/□t=0)
- Fick's First Law of Diffusion (for flux computation): $J = -D\frac{dC}{dx}$
- Boundary Conditions: $C(0) = C_{\text{feed}} = \sqrt{p_{\text{feed}}}$ $C(L) = C_{\text{permeate}} = \sqrt{p_{\text{perm}}}$
- Numerical Solution Approach
  - Domain length L split into Nx points.
  - Central difference applied to second derivative: $\frac{C_{i+1} - 2C_i + C_{i-1}}{\Delta x^2} = 0$
- Matrix Formulation: AC=b where A Tridiagonal matrix from discretization and b is RHS with boundary conditions.

scipy.linalg.solve: Direct solution of linear system
scipy.optimize.root / fsolve: Nonlinear root finders used to solve the same system as residual function

Hydrogen Flux vs. Membrane Thickness vs. Diffusion Coefficient

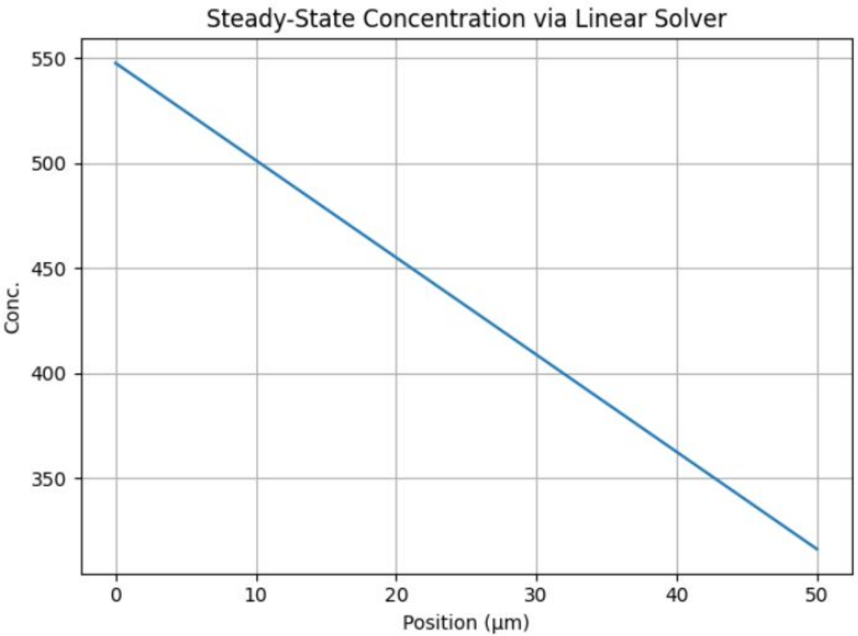
Effect of Membrane Thickness on Hydrogen Flux

- Equation Used: $J = \frac{P}{L}(\sqrt{p_{\text{feed}}} - \sqrt{p_{\text{perm}}})$ where $P = P_0 \exp\left(-\frac{E_a}{RT}\right)$ Temperature-dependent permeability, L: Membrane thickness.
- As membrane thickness increases, flux decreases sharply.
- Thinner membranes enhance transport, but trade-offs exist (mechanical strength, durability).

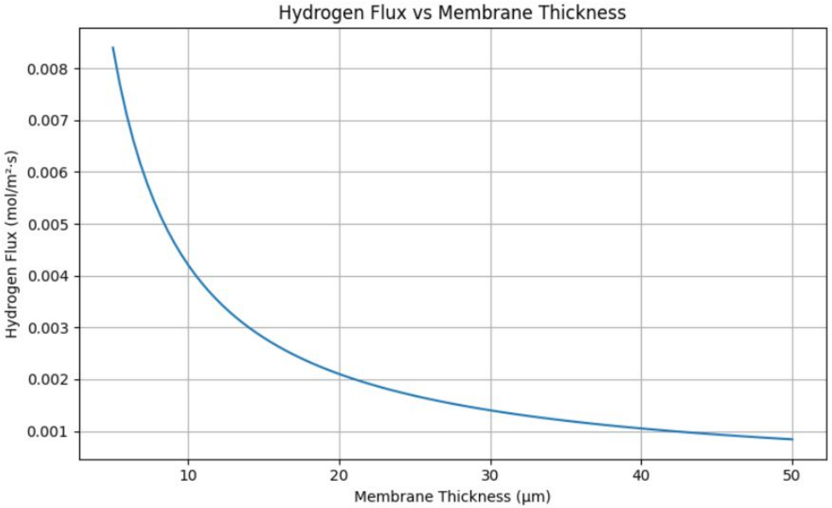
Effect of Diffusion Coefficient on Flux

- Equation Used: $$J = \frac{D(C_1 - C_2)}{L}$$
- Simulation Parameters:
  - $L=125\mu m$ fixed
  - $D=1\times10^{-10}$ to 5×10^{-10} m^2/s,
  - Boundary concentrations: $C_1 = 1.0$, $C_2 = 0.0$

Steady state: Concentration and flux

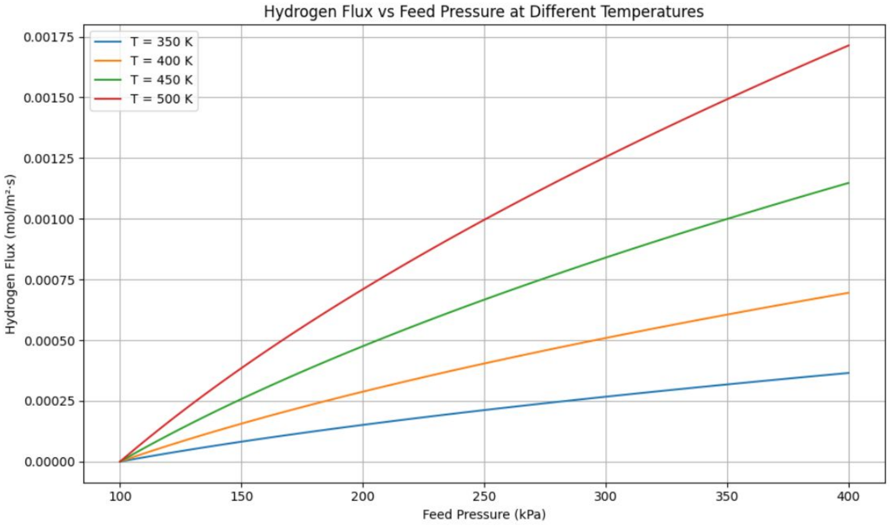


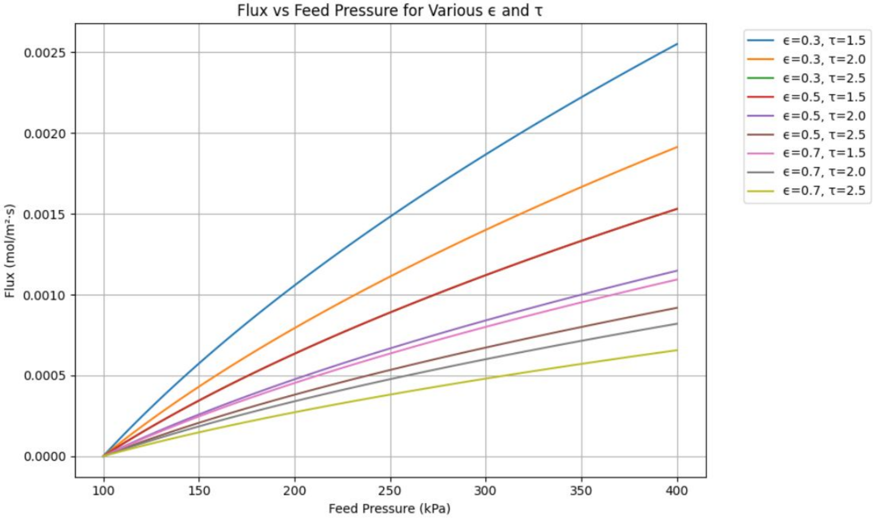
Steady state concentration solved by scipy.linalg. The fick's law gives linear value for conc. vs position



As the membrane thickness increases the flux decreases

Steady state: Flux vs feed pressure @ temp. and ϵ,τ



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Optimal membrane thickness = 5.00 µm
Maximum hydrogen flux = 8.401e-03 mol/m²·s
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Optimal Temperature = 450.0 K
Maximum Flux = 8.401e-04 mol/m²·s
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Transient State Simulation in H2 Diffusion

Governing equation: Fick's Second Law of Diffusion

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2}$$

Key parameters:

- Membrane length: $L = 0.75 \times 10^{-6}$ m
- Diffusion coefficient: $D = 1 \times 10^{-8}$ m^2/s

Boundary Conditions:

- Left boundary (x = 0): C = 1.0 (constant concentration source)
- Right boundary (x = L): C = 0.0 (zero concentration)

Initial Condition:

- C(x, t=0) = 0 for all interior points
- C(0, t=0) = 1.0 and C(L, t=0) = 0.0 at boundaries

Numerical Method Implementation

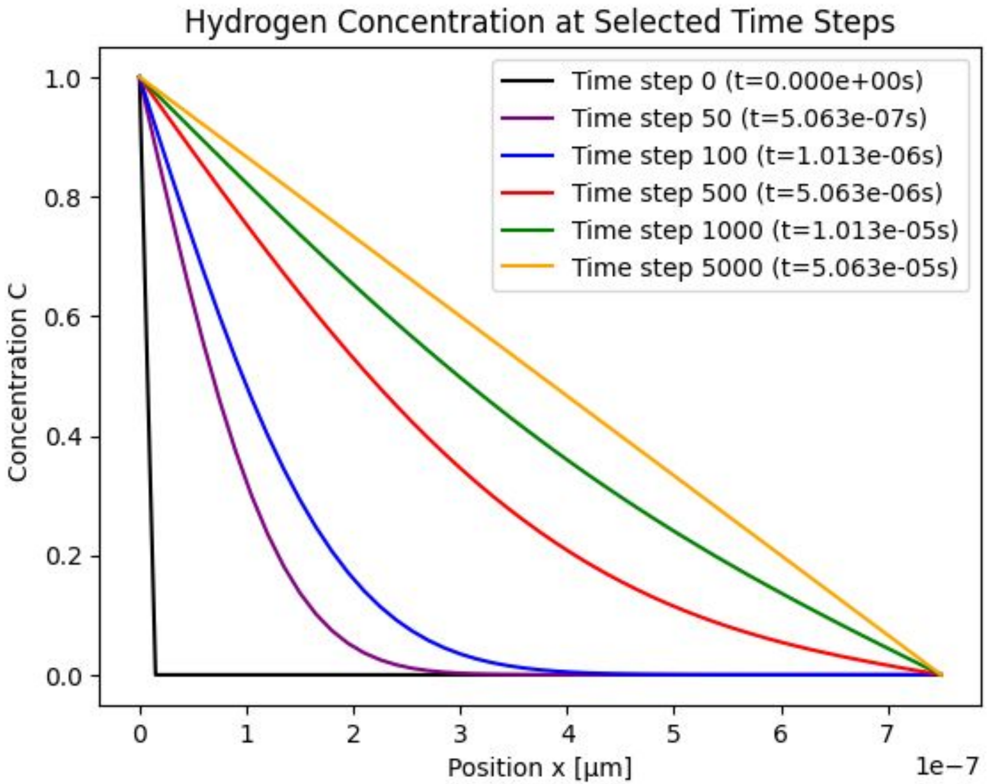
Discretization of the PDE

- The continuous PDE is converted to a discrete form using **Finite Difference Method**
- **Spatial discretization**:
  - Domain divided into Nx = 51 points
  - Grid spacing: dx = L/(Nx-1)
- Second derivative approximation:

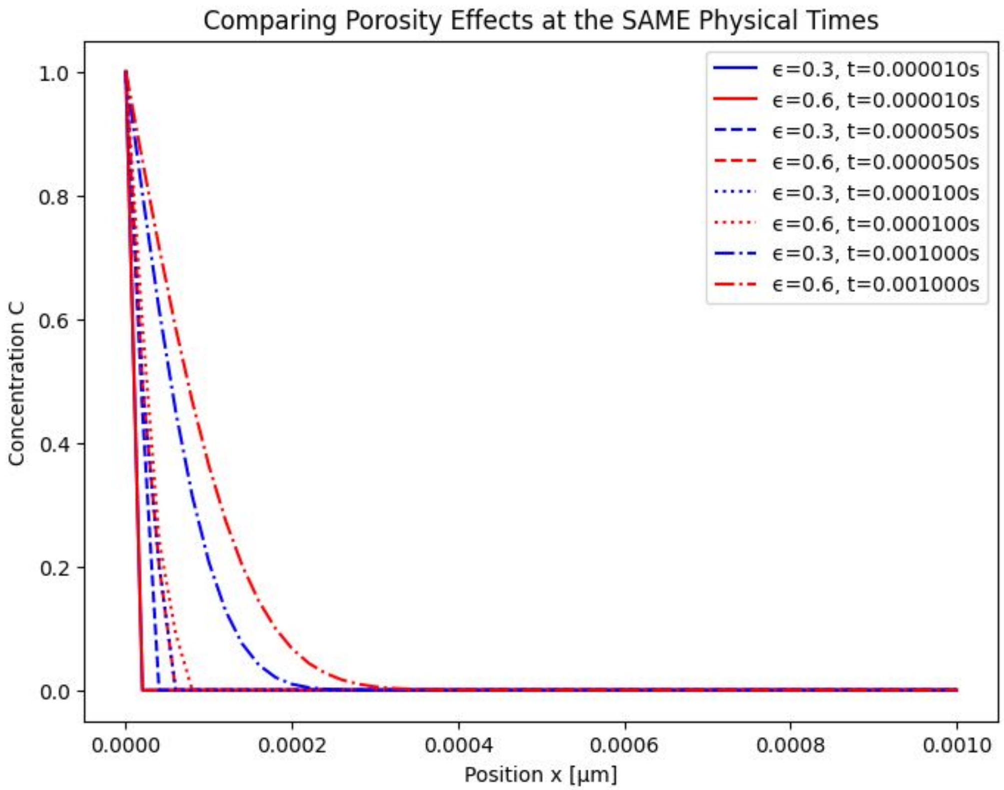
$$\frac{\partial^2 C}{\partial x^2} \approx \frac{C_{i+1} - 2C_i + C_{i-1}}{(\Delta x)^2}$$

$$\frac{C_i^{n+1} - C_i^n}{\Delta t} \approx D \frac{C_{i+1}^n - 2C_i^n + C_{i-1}^n}{(\Delta x)^2} \xrightarrow{\text{rearrange}} C_i^{n+1} \approx C_i^n + \frac{D\,\Delta t}{(\Delta x)^2}\left(C_{i+1}^n - 2C_i^n + C_{i-1}^n \right)$$

Result Demo: Vanilla Finite Difference method



Result Demo: Finite Difference + Porosity



Future Work

- Add temperature-dependence and permeability gradients
- Include non-Fick transport to simulate real membrane data
- Integrate experimental validation

Backup slides