A CFD Analysis of the Operating Conditions of a Multitube Pd Membrane for H_2 Purification

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Introduction

The production of hydrogen is a major process for the chemical industry as well as for the energy sector. Using palladium membranes is an economic and efficient method to purify H_2. They remove H_2 by catalyzing the dissociation of the molecule at the surface and diffusing it through the lattice of the metal. The physics and geometry involved make the behavior of multitube membrane modules difficult to predict and simulate, but necessary for the implementation of this technology. This work uses COMSOL Multiphysics 4.4 to study the operating conditions of a seven membrane module. The influence of different Reynolds numbers on both, recovery and membrane utilization, is discussed. An optimum point to maximize the efficiency of the module is presented.

Computational Methods

To simulate the fluid flow and mass transport, the following equations were used:

Equation of motion:

$$\rho(\mathrm{u}\cdot\nabla)\mathrm{u} = \nabla\cdot\left[-\mathrm{pI} + \mu\left(\nabla\mathrm{u} + (\nabla\mathrm{u})^{\mathrm{T}}\right] - \frac{2}{3}\mu(\nabla\cdot\mathrm{u})\mathrm{I}\right)$$

$$\nabla\cdot(\rho\mathrm{u}) = 0$$

Species continuity equation:

$$\nabla\cdot\mathrm{j}_i + \rho(\mathrm{u}\cdot\nabla)\mathrm{w}_i = \mathrm{R}_i$$

$$\mathrm{j}_i = -\left(\rho\mathrm{D}_i^{\mathrm{m}}\nabla\mathrm{w}_i + \rho\mathrm{w}_i\mathrm{D}_i^{\mathrm{m}}\frac{\nabla\mathrm{M}_n}{\mathrm{M}_n}\right)$$

Sieverts' law was used as the boundary to simulate the H_2 flux across the membranes:

$$-\mathrm{n}\cdot\mathrm{N}_i = \bar{\mathrm{P}}_{H_2}\left[\sqrt{\mathrm{p}_{H_2}^{shell}} - \sqrt{\mathrm{p}_{H_2}^{tube}}\right]$$

Where $\bar{\mathrm{P}}_{H_2}, \mathrm{p}_{H_2}^{shell}, \mathrm{p}_{H_2}^{tube}$ are the H_2 permeance, H_2 partial pressure in the shell and tube side, respectively.

Simulation setup

The multitube membrane module consists of seven Pd membranes (Fig.1). Hydrogen enriched syngas is fed at the shell side while the permeated H_2 gas is collected from inside of the membrane tubes.

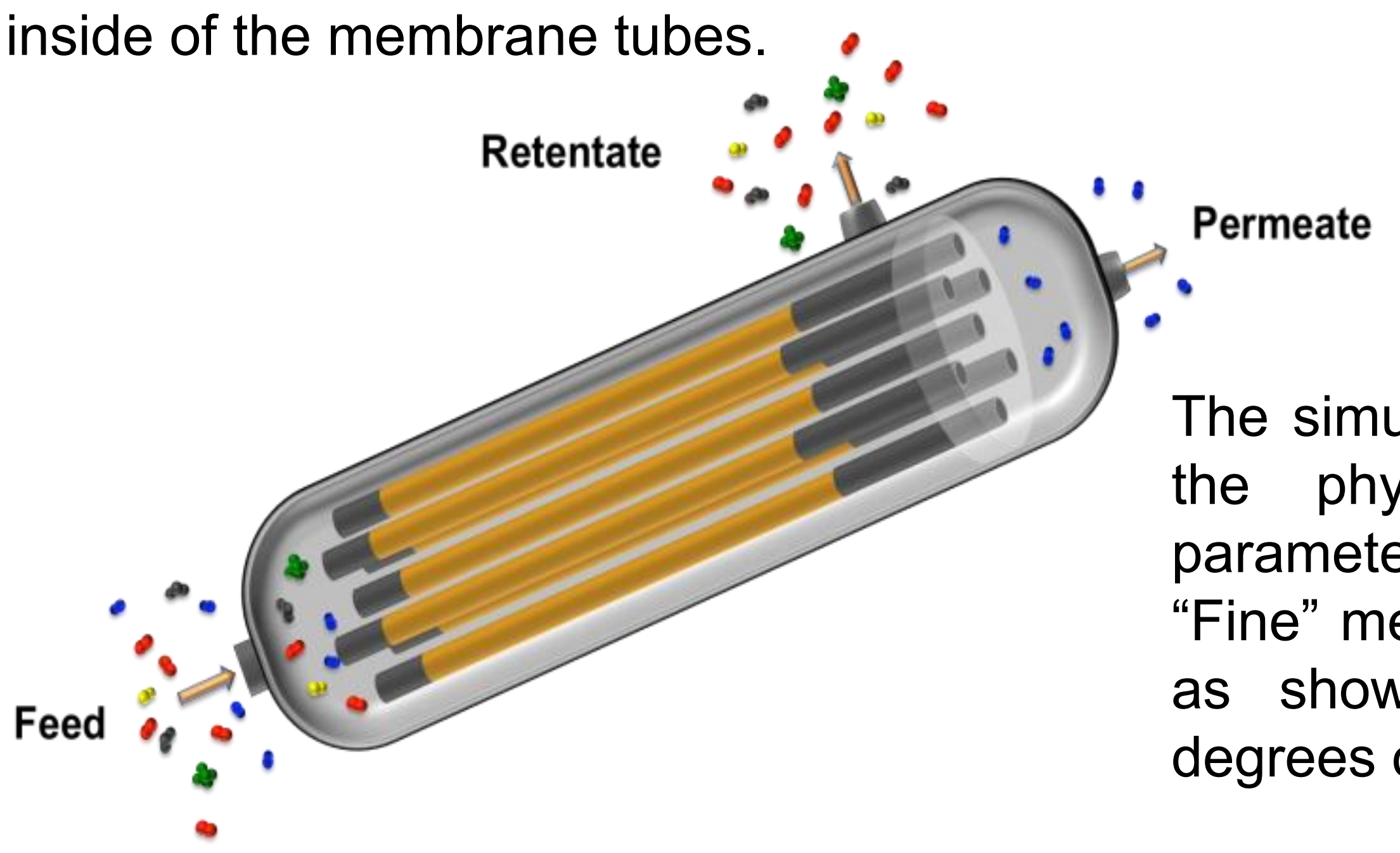


Fig 1. Representative sketch of the actual multitube membrane module

The simulation used "Reacting Flow" as the physics. Table 1. shows the parameters used in this simulation. "Fine" mesh was used for the simulation as shown in Fig 2. with 3,181,368 degrees of freedom.

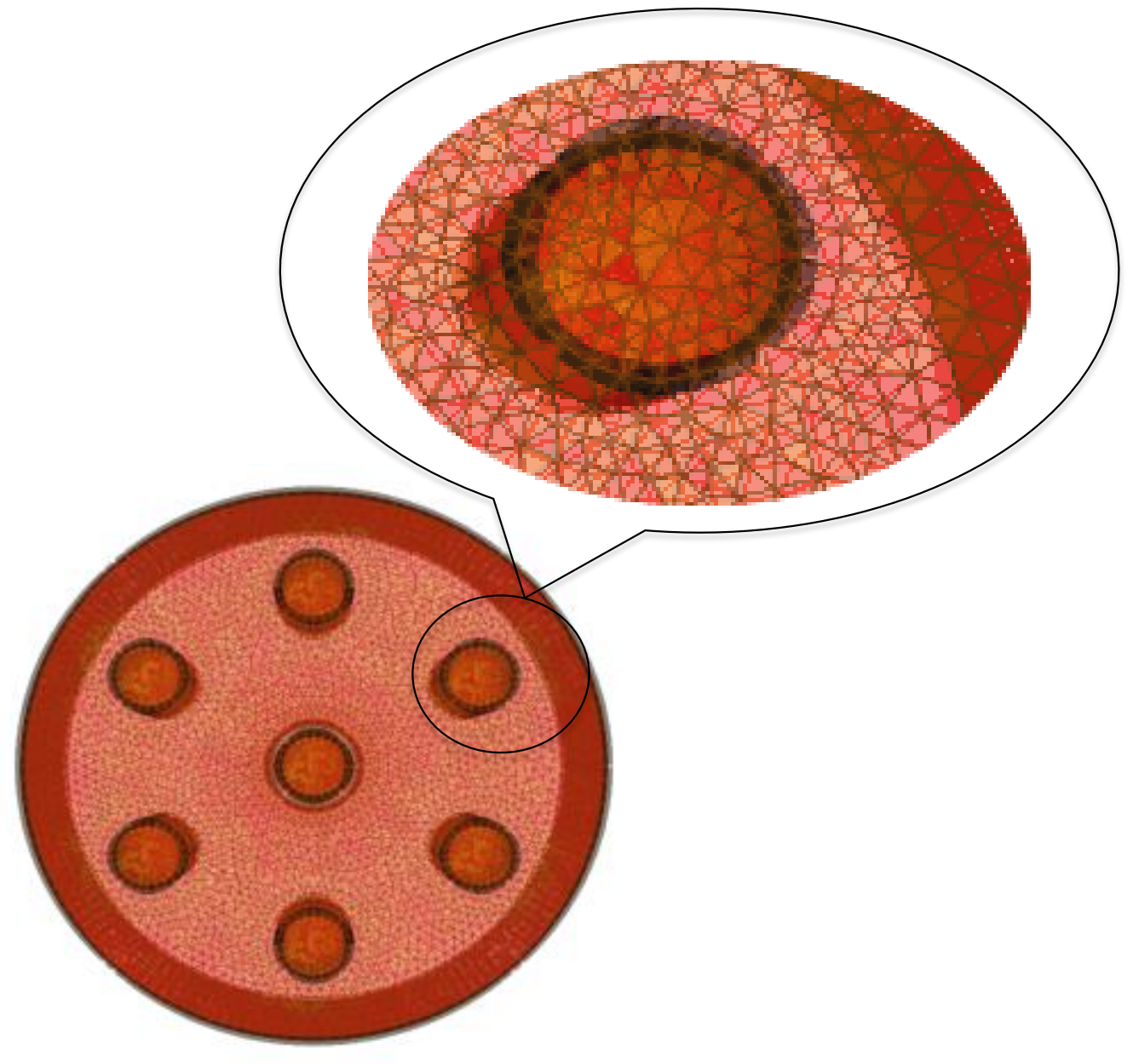


Fig 2. Cross-sectional view of the mesh used in the simulation

Table 1: Operational setting of the module

Shell Pressure / atm	12.6
Tube Pressure / atm	1
H_2 Permeance / mol $m^{-2}s^{-1}Pa^{-0.5}$	7.7×10^{-4}
Reynolds number	1-300
Initial gas composition / mole %	
H_2	43
N_2	50
CO	5
CO_2	1.5
CH_4	0.5

Results

Velocity Profile

The velocity of the gas decreases after the expansion between the inlet and the membrane module. Velocity increases and mixing occurs when the gases encounter the membranes.

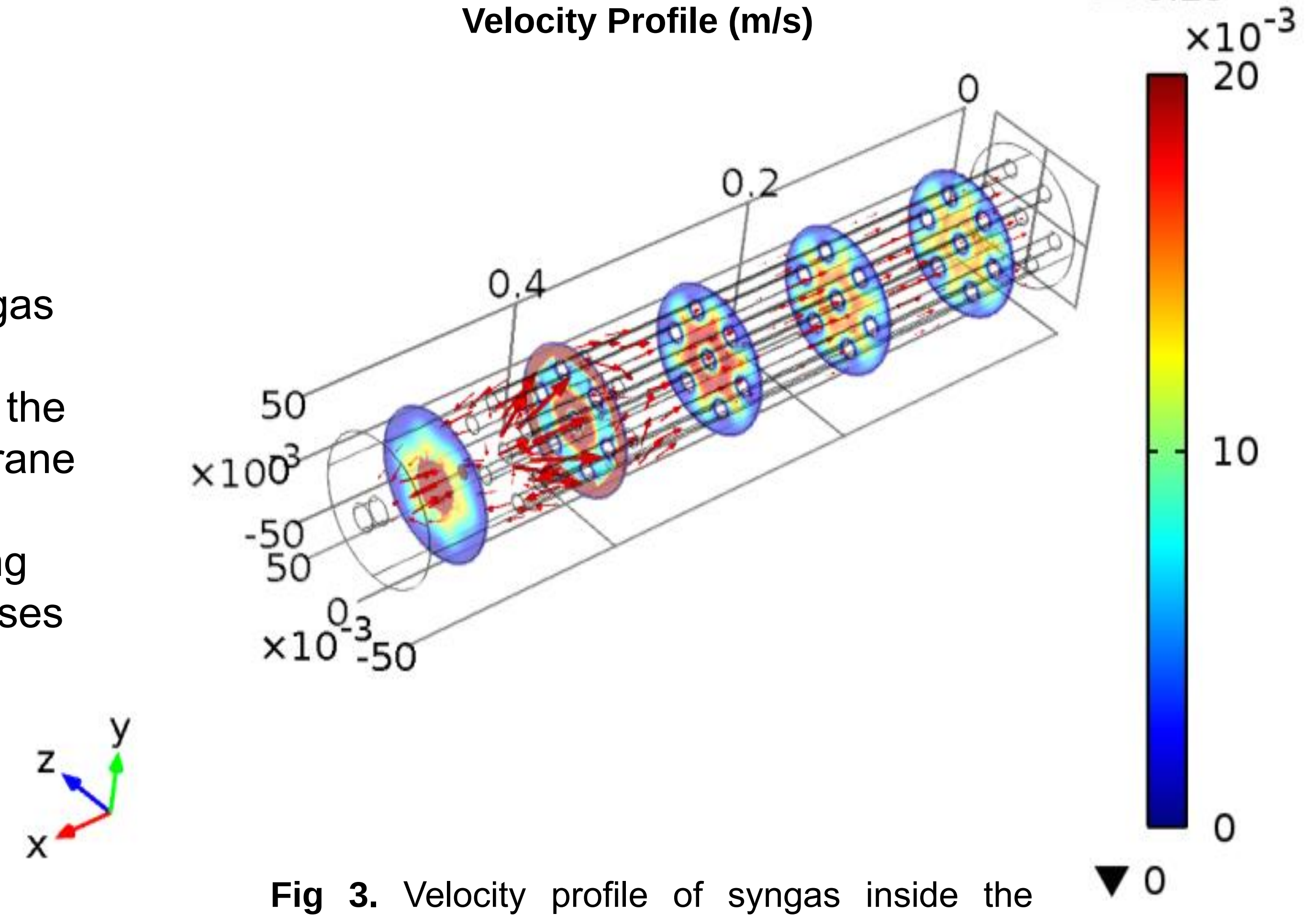


Fig 3. Velocity profile of syngas inside the membrane module at moderate Re numbers

H_2 Mole Fraction Profile

Different Re numbers displayed different H_2 mole fraction profiles within the membrane module. At low Re numbers, the H_2 recovery is maximized, but the membrane usage capacity is not exploited (Fig 4a). As the Re number increases, the membranes were used further down axially, but recovery decreased (Fig 4b).

Fig 5. shows a H_2 depleted boundary layer or concentration polarization. At high Re numbers, the boundary layer becomes thinner, but the H_2 recovery of the process reduces.

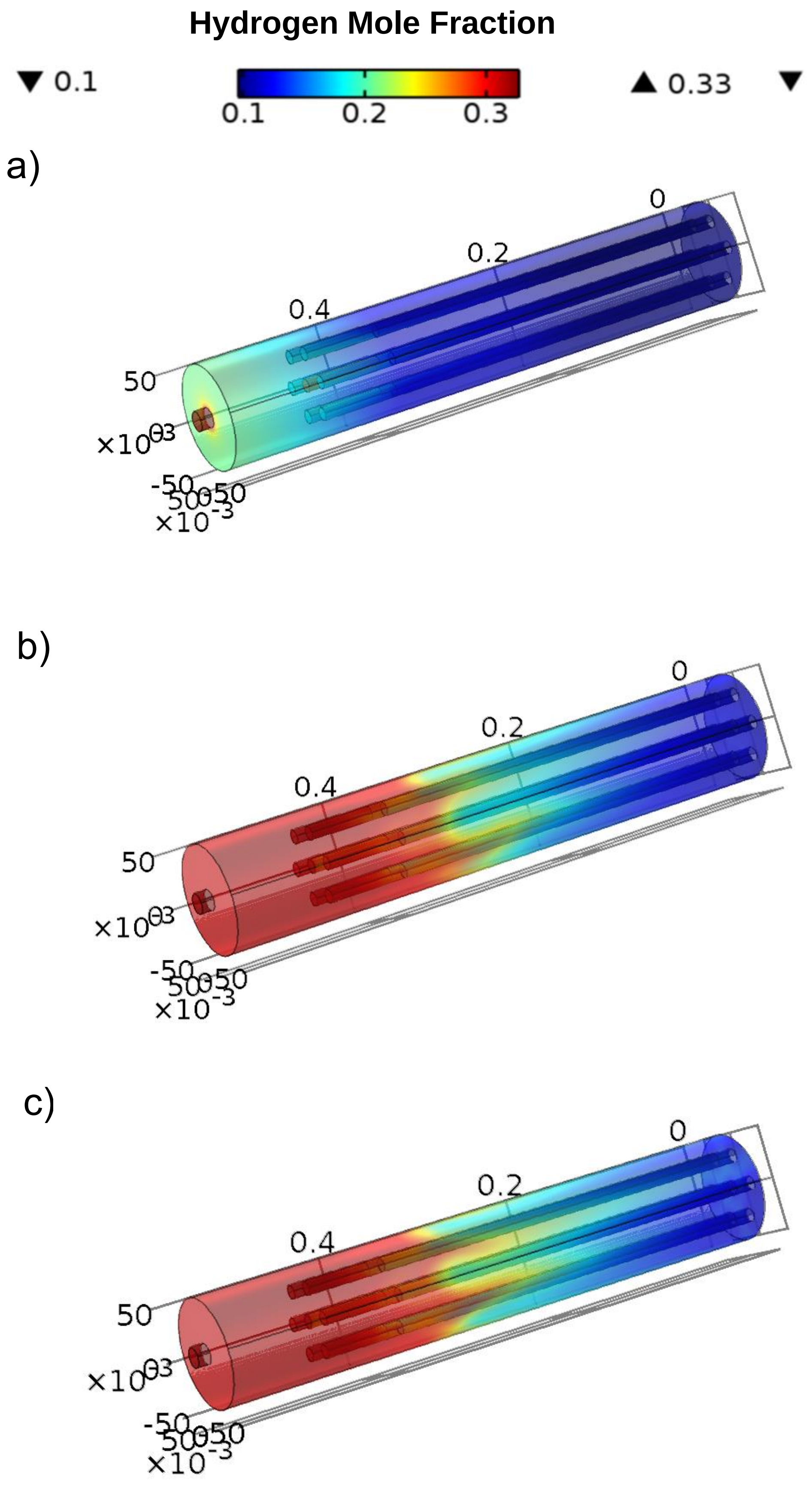


Fig 4. Axial view of the H_2 mole fraction distribution at different Re numbers: a) Low Re number, b) Medium Re, c) High Re

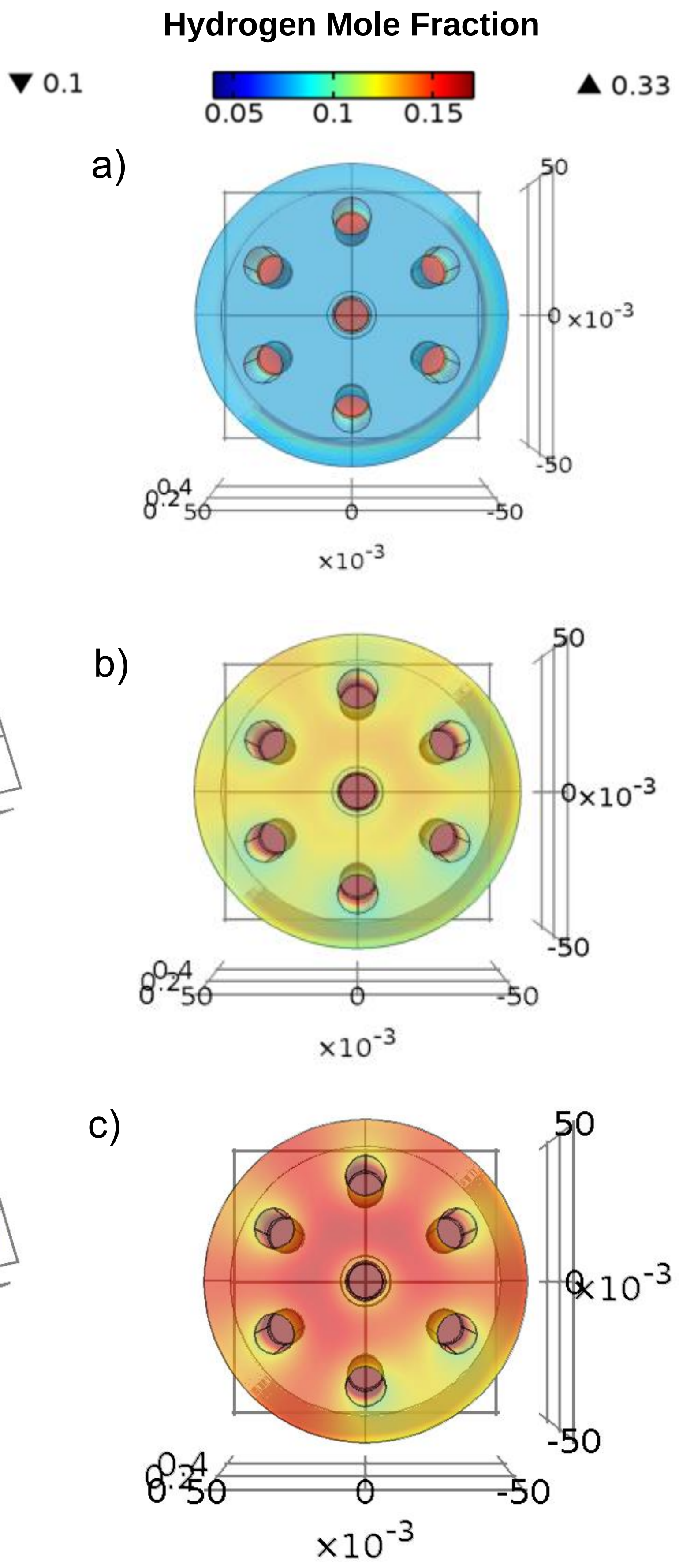
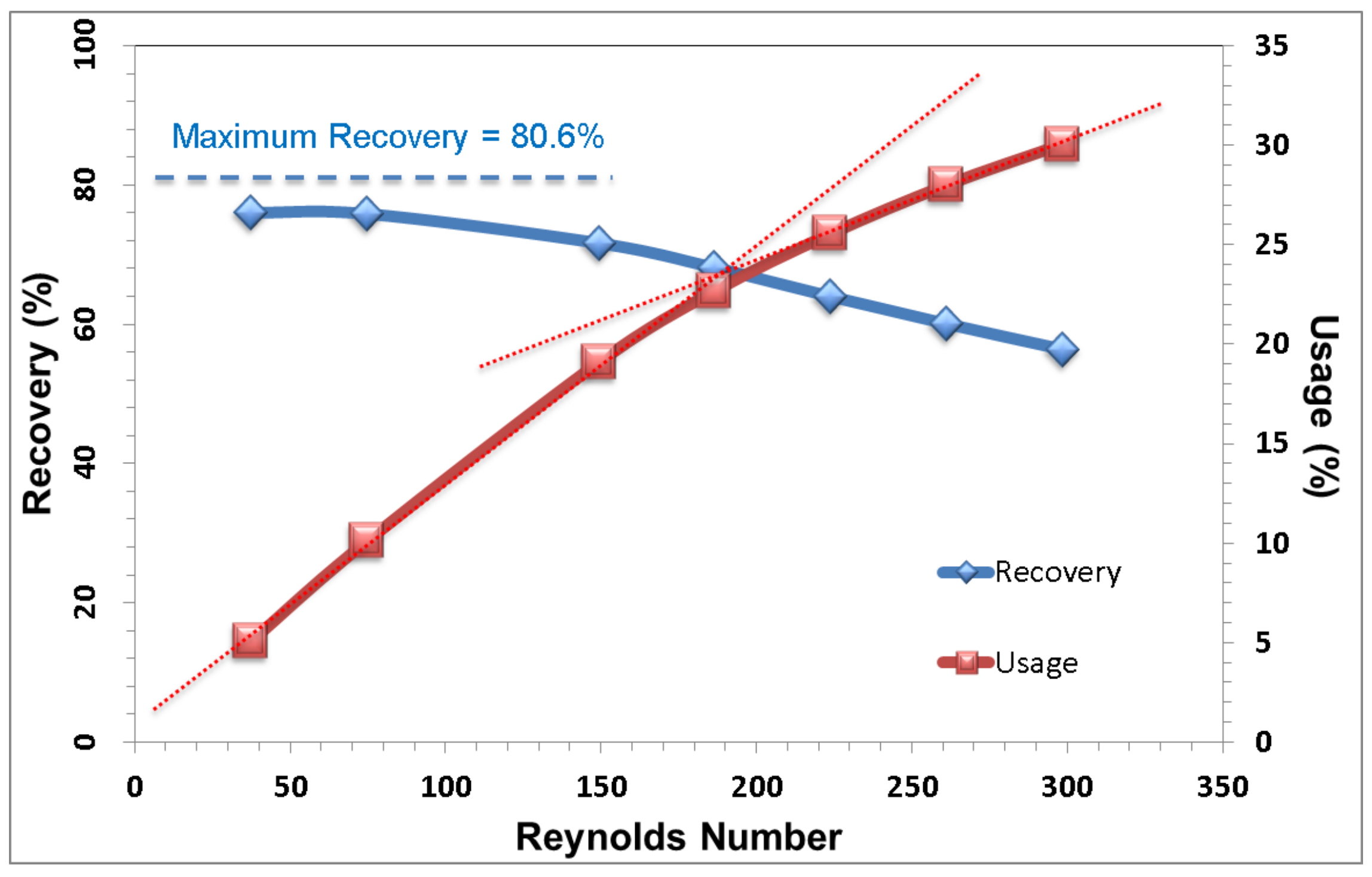


Fig 5. Cross-sectional view of the H_2 mole fraction distribution at different Re numbers: a) Low Re number, b) Medium Re, c) High Re



Recovery ($R_{H_2}(\%)$) and membrane usage ($\varphi(\%)$) were computed as:

$$R_{H_2}(\%) = \frac{F_{H_2}^{in} - F_{H_2}^{out}}{F_{H_2}^{in}}\cdot 100$$

$$\varphi(\%) = \frac{\gamma}{\gamma_{max}}\cdot 100$$

Fig 6. Recovery of H_2 and membrane utilization as a function of Re numbers

The optimum point for operating the module is represented at the intersection where the recovery and membrane utilization overlap (Fig 6.).

Conclusions

1. Low Re numbers displayed high recoveries, but the membranes were utilized ineffectively.
2. High Re numbers use the membranes more evenly and reduced concentration polarization, but H_2 recovery decreased.
3. A tradeoff between usage of the membrane and H_2 recovery was successfully depicted. An optimized Re number was presented balancing both properties.

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