



Supplementary Materials

2 1. Analysis of the Instrumental Time Lag for a Thick Dense SBS Film and an Sbs TFC Membrane

SI Figure 1 gives a comparison of the time lag both , permeance, and permselectivity of a thick SBS film and a TFC SBS membrane. The trends for membranes are very similar and the small difference in the selectivity is probably due to the slightly different properties in bulk and in thin films. The virtually constant permeability and selectivity as a function of the sweep flow rate suggest that polarization phenomena in the permeate side are negligible, as this would have led to a decrease in permeability of the most permeable gas (CO₂) at the lowest sweep flow rates.



SI Figure 1. Effect of the sweep flow rate on the time lag (a), the permeances (b), and the
permselectivity (c) of a fast 5 μm thin film composite membrane (spheres) and a slower 159 μm thick
dense film (diamonds) for the N₂/O₂/CO₂ 80/10/10 vol % mixture. Sweeping gas at atmospheric
pressure.



13 2. Study of the Effect of Slow Pressure Increase on the Instrumental Time Lag

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 SI Figure 2. Increase of the feed pressure upon switching of the six-way feed valve from Argon purge

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 to the 80/10/10 vol % N₂/CO₂/O₂ mixture (a), corresponding permeation curves for O₂ (b), and

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 resulting instrumental time lag (c) determined with an SBS TFC membrane (area 1.77 cm⁻²). Feed flow

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 rate 200 cm³sTP min⁻¹ and sweep flow rate 30 cm³sTP min⁻¹.



18 3. Reaction of the Permeate Composition on Changes in the Feed Pressure

SI Fig. 3. Profile of the feed flow rates and feed pressure (a), and the permeate flow rates (b) as a function of time for the SBS thin film composite membrane with a 80/10/10 vol % N₂/CO₂/O₂ mixture.





SI Figure 4. Comparison of the increasing feed pressure as a function of time for individual pressure
 increase ramps (a) and for stepwise increasing pressure (b) for 2088 days aged sample SBF with the
 gas mixture CO₂/CH₄ 35/65 vol %.

25 5. Schematic Concentration Profile Development in Three Subsequent Pressure Steps





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SI Figure 5. Schematic representation of the development of the concentration profiles in the membrane after its first exposure to the gas and during two subsequent pressure increase steps.







31SI Figure 6. Individual time lag for CO_2 and CH_4 in a 2088 days aged sample of PIM-SBF with a32stepwise pressure increase (full circles). The empty circles show the corresponding points for the33pressure decrease steps. Points of the instrumental time lag Θ_0 for N₂, O₂, and CO₂ overlap.