



Funzionano



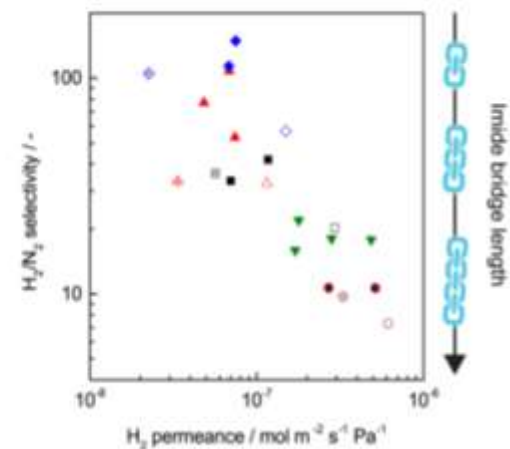
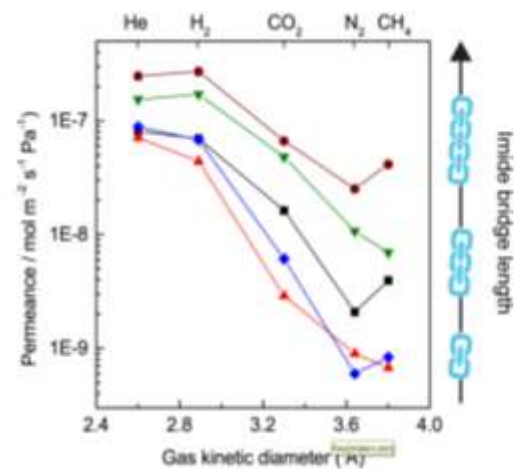
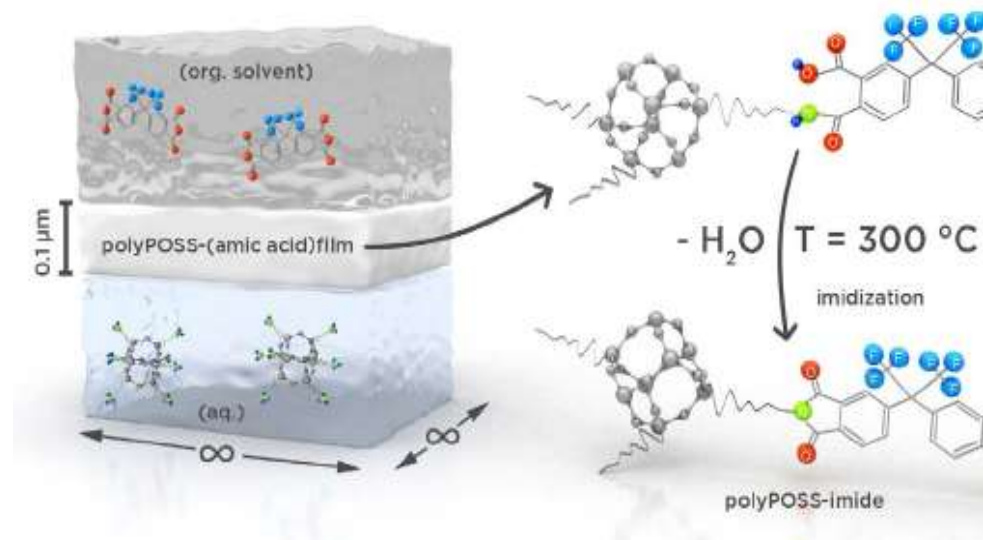
UNIVERSITY OF TWENTE.

# Mixed gas separation performance and upscaling of PolyPOSSimide membranes for H<sub>2</sub> purification

L. Ansaloni, E. Louradour, T.A. Peters, M. Pilz, C. Simon, F. Karimov, N.E. Benes, D. Sherban

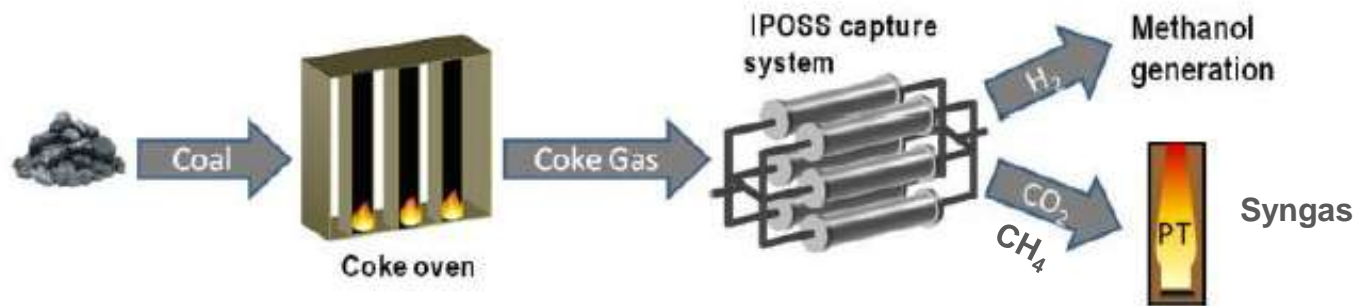
# Intro – iPOSS

- Hybrid iPOSS membranes for the separation of  $H_2$  in a pre-combustion environment
- Crosslinked network of alternating, covalently bonded imide and POSS groups
- Formed by an interfacial polycondensation (IP) of an di-anhydride and amine-POSS at the interface between two solvents, followed by a heat treatment
- H2020 - GENESIS project (NMBP20-2017)
  - Optimisation of POSS-building blocks to increase the thermal stability. Project aim 300 °C
  - Upscaling the membrane fabrication to tubular single channel and multi-channel element membranes
  - Demonstrate onsite at TRL6

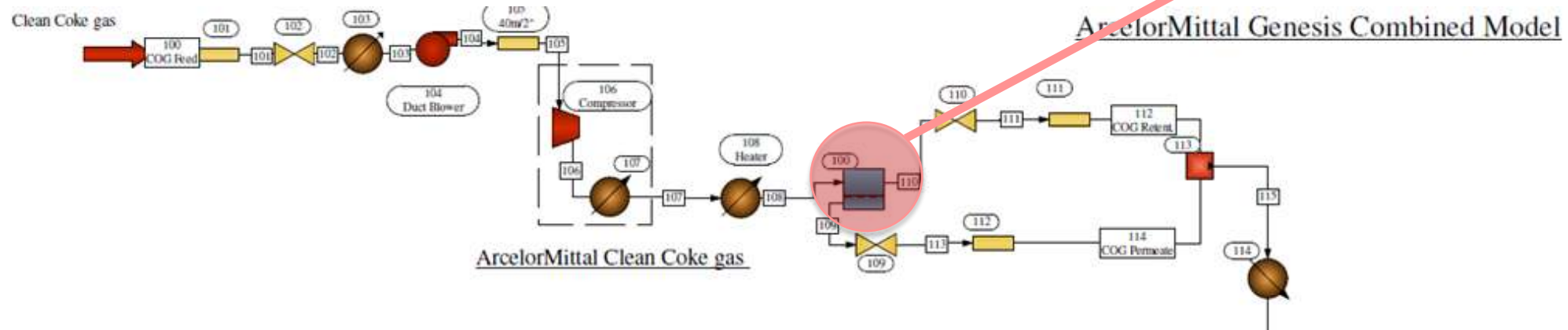


# Intro – demonstration

- Arcelor-Mittal – clean coke gas upgrading



Component	Content [vol.%]
N <sub>2</sub>	5.7
CO <sub>2</sub>	1.2
CO	4.0
H <sub>2</sub> O	5.6
CH <sub>4</sub>	21.2
H <sub>2</sub>	59.4
Ar + rest	2.9





# Intro – membrane upscaling

- Status: tubular single channel and multi-channel element membranes
  - Develop coating procedures for tubular membrane elements
  - Optimization and reproducibility of iPOSS layers on single CTI tubular asymmetric membrane supports
- Final aim
  - Synthesis of iPOSS layer on multichannel membrane supports (diameter 25mm, length 400mm, 61 channels of 2 mm diameter) with an effective area of 0,15 m<sup>2</sup>
  - Demonstration at membrane area: ~1 m<sup>2</sup>

small disks, 2014



GENESIS, 2018



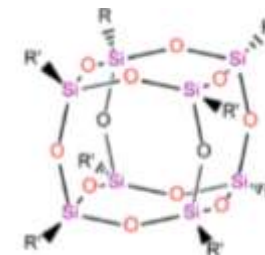
GENESIS, 2019-2020



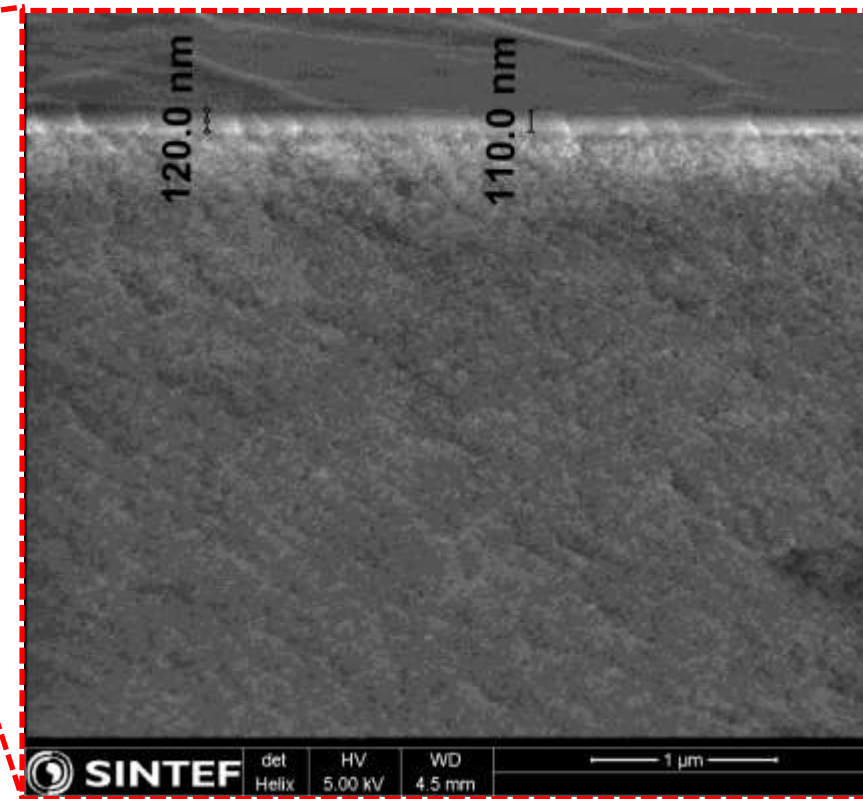
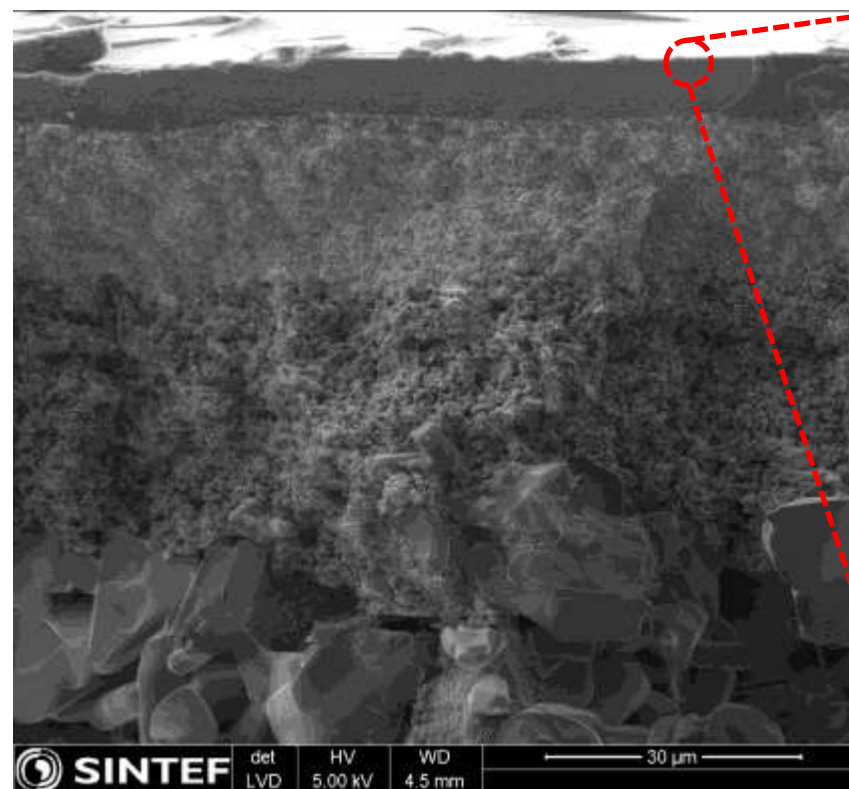
# iPOSS membrane



+



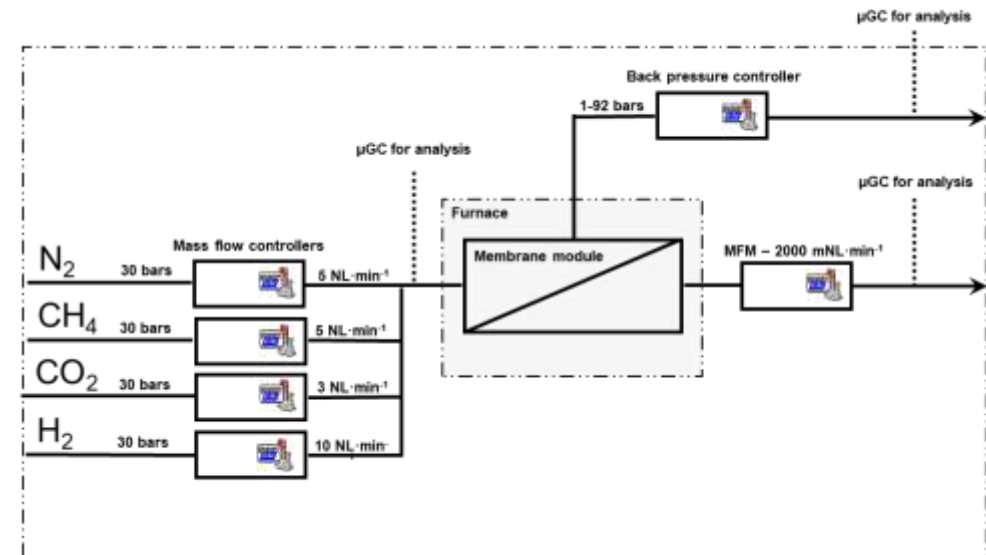
R =  $-(CH_2)_3-NH_3^+Cl^-$   
 R' =  $-(CH_2)_3-NH_2$   
 octa-ammonium POSS



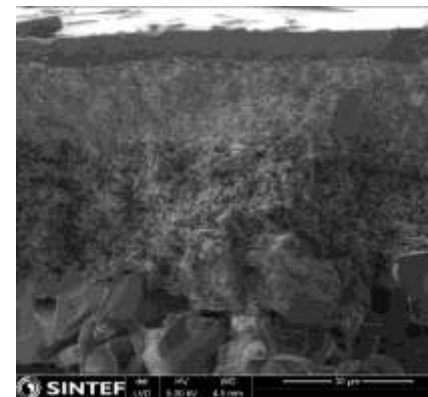
# Gas separation performance

- Gas permeation measurements
  - Pure gases, mixed (quaternary) gases
  - Dead-end mode; no active sweep
  - Up to 300 °C,  $\Delta P = 9$  bar

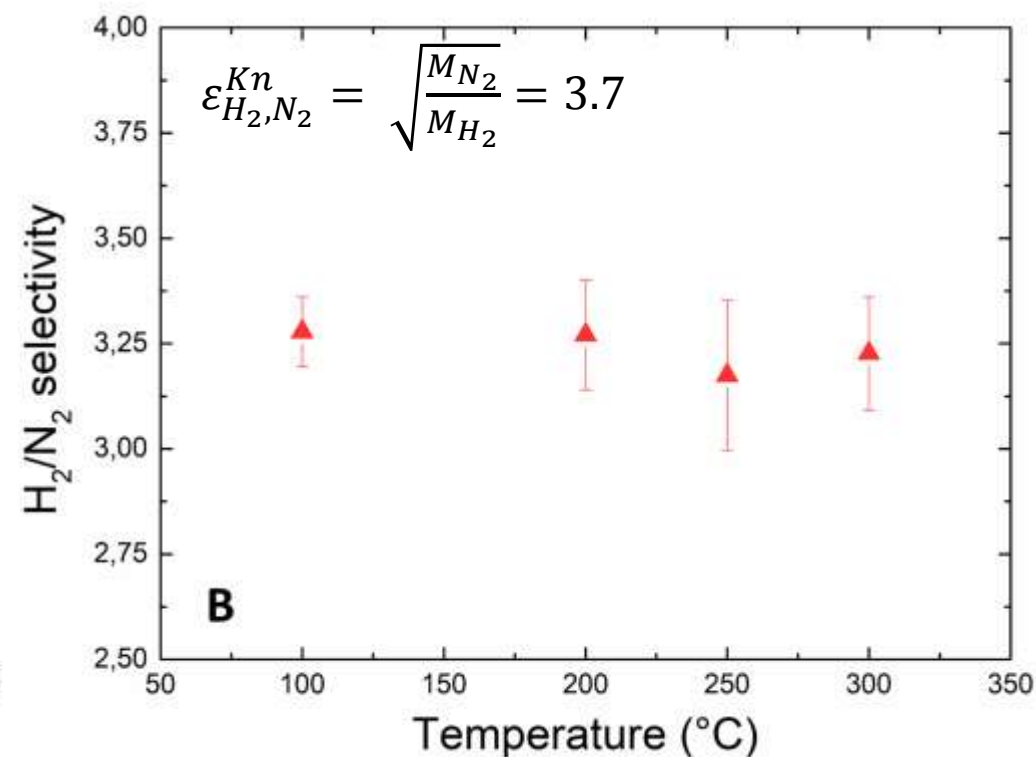
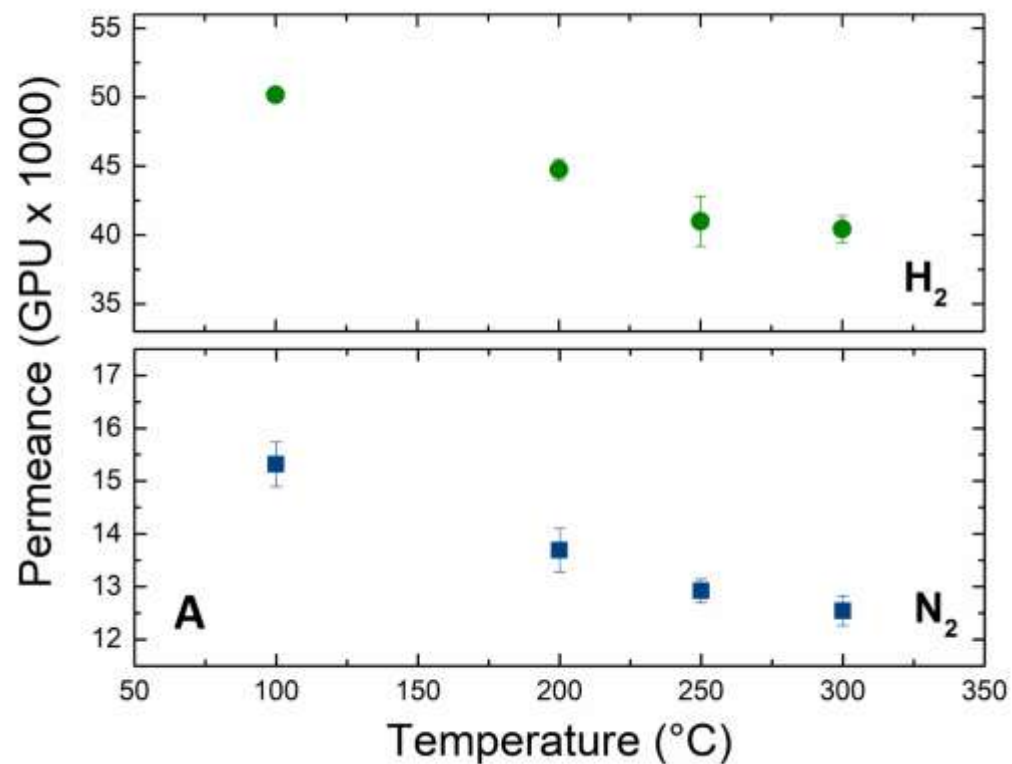
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# Gas separation performance

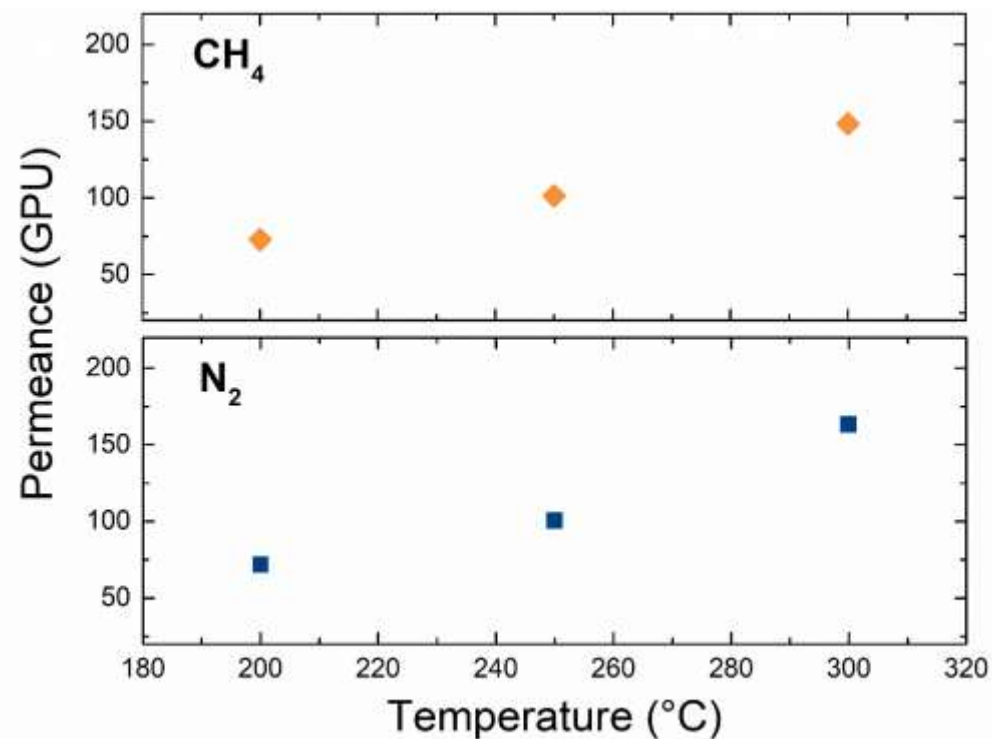
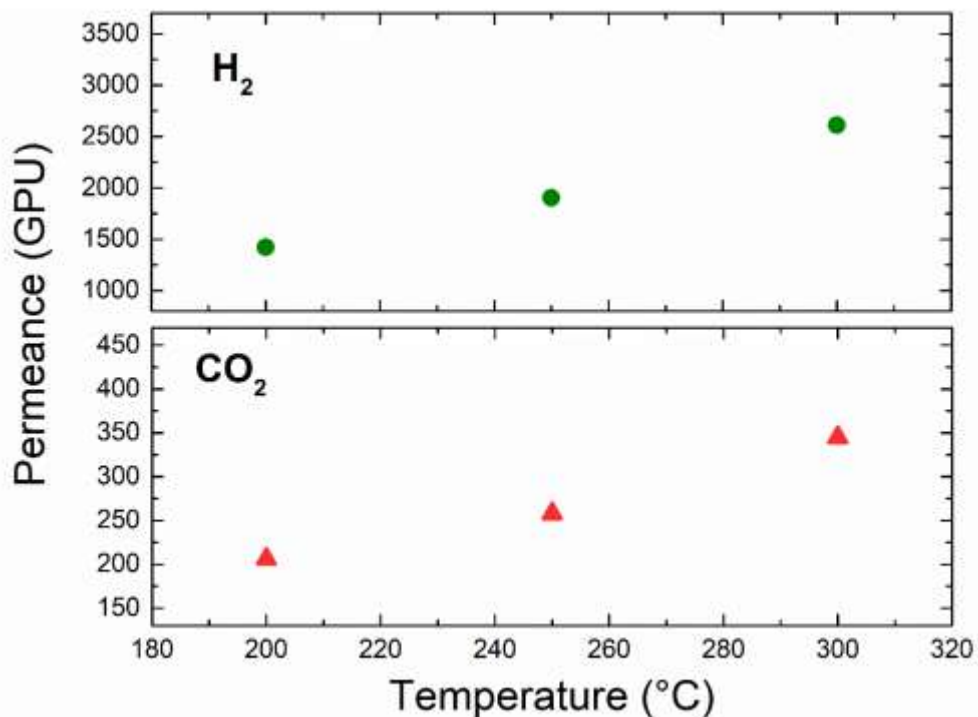
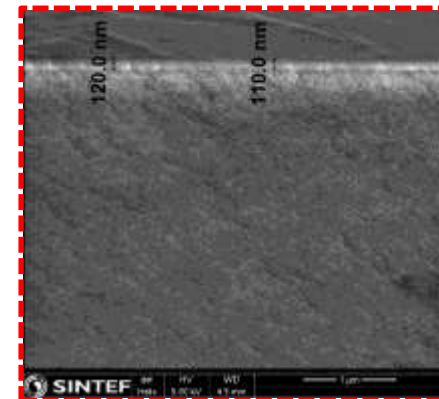


- Asymmetric tubular membrane support
  - Quality of support of uttermost importance



# Gas separation performance

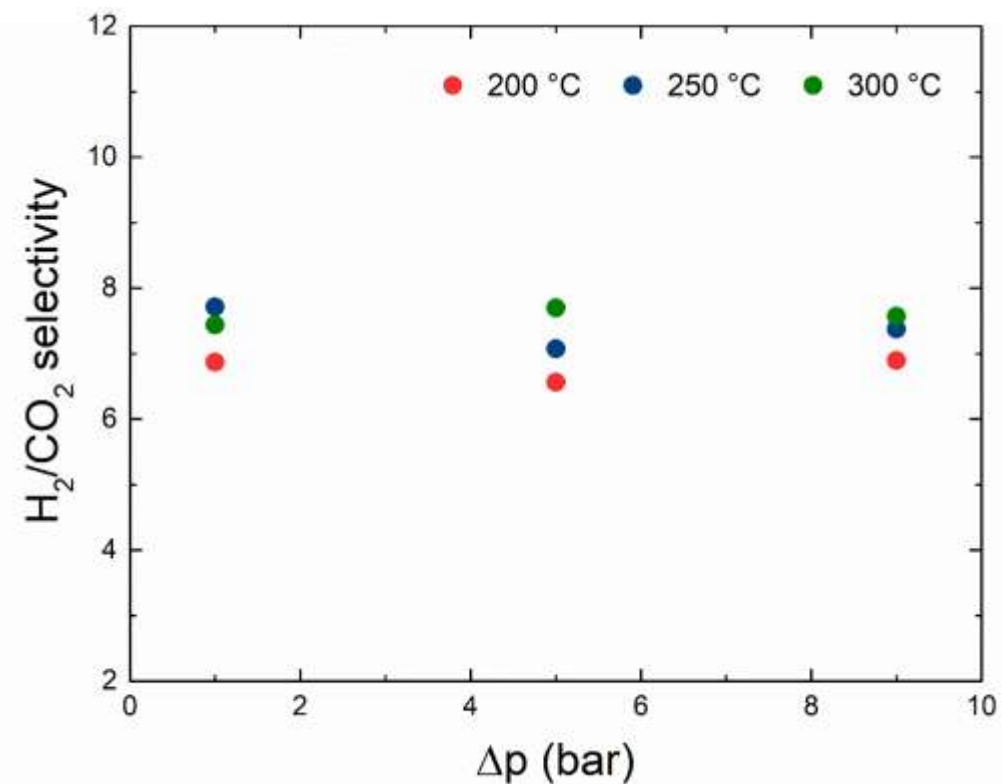
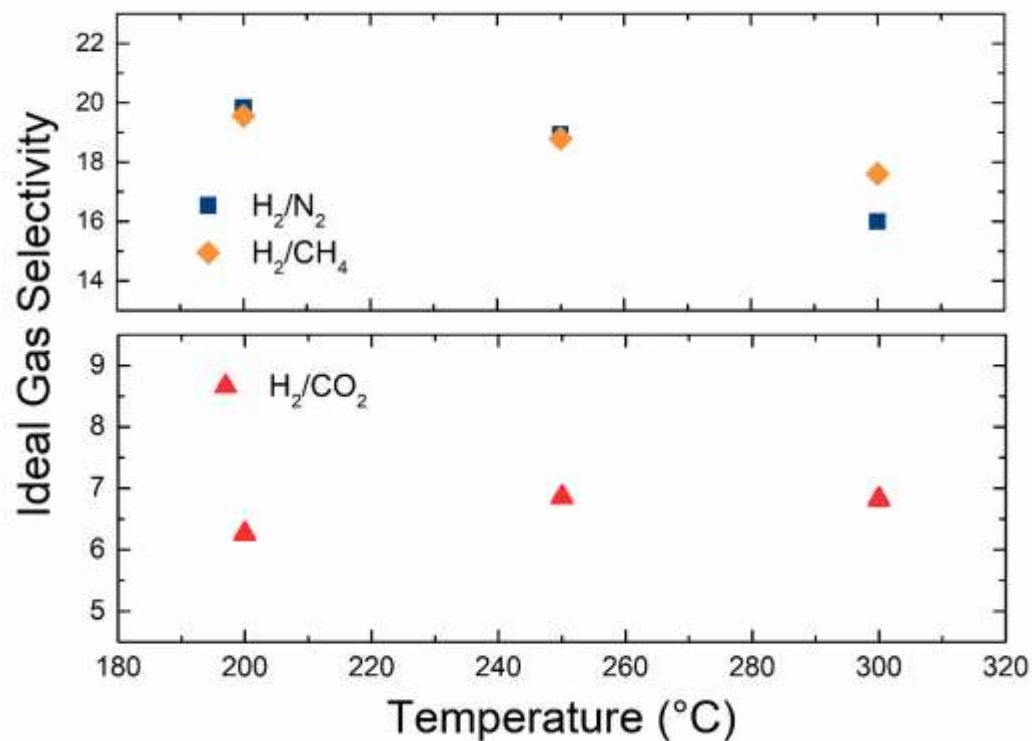
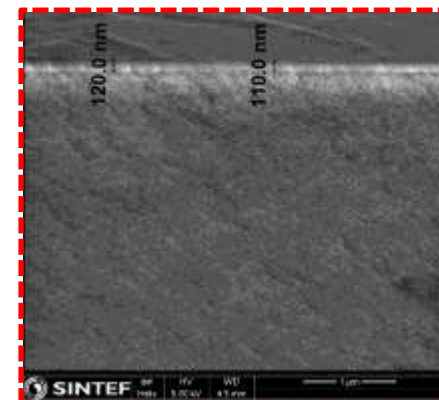
- Single gas – obtained at  $\Delta P = 9$  bar





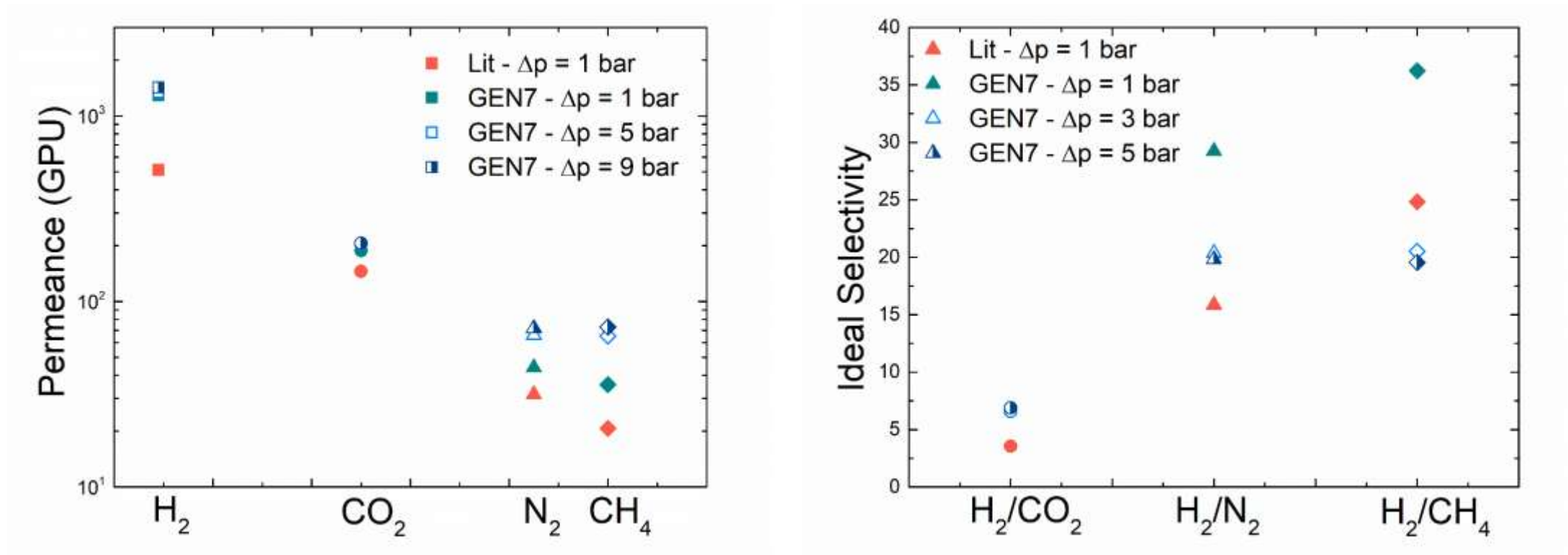
# Gas separation performance

- Single gas – permselectivity values



# Gas separation performance

- Literature comparison – 200 °C: small disks versus upscaled tubular membranes

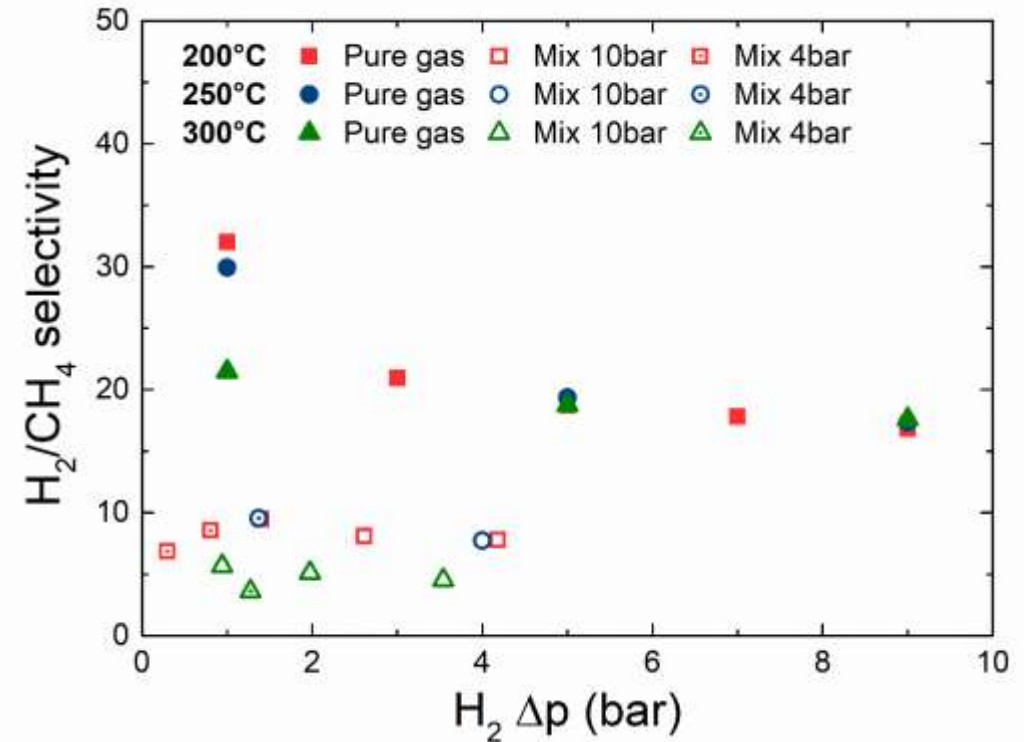
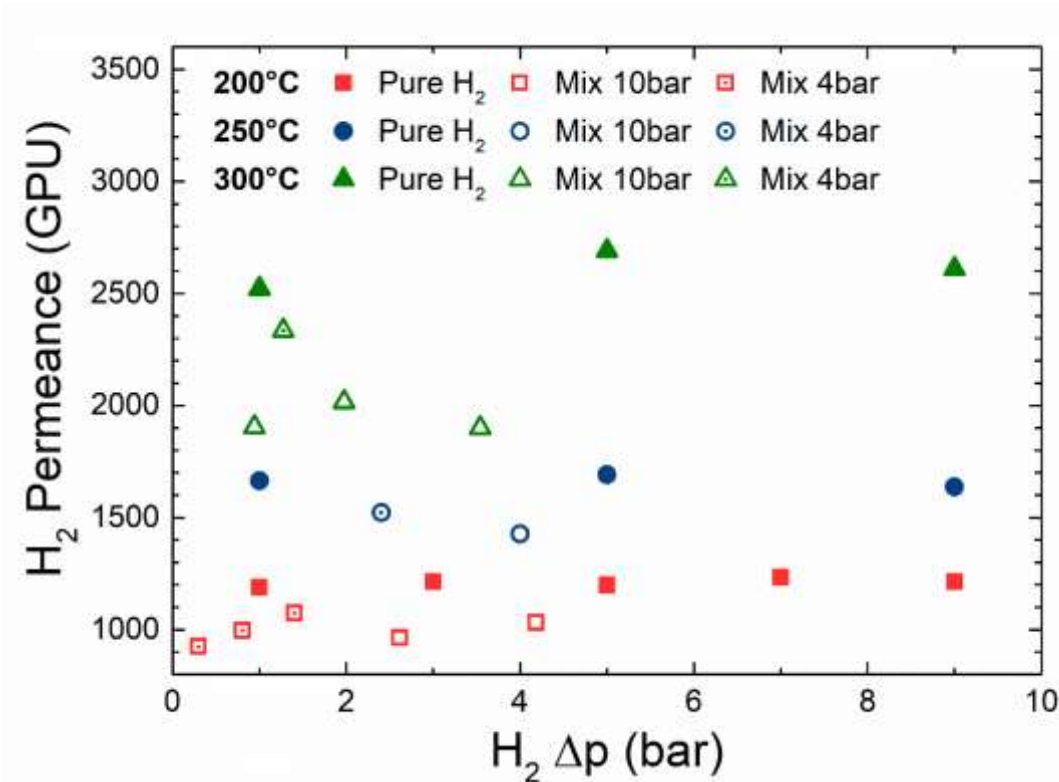


# Gas separation performance

## Mixed Gas

Mix 1 = 60% H<sub>2</sub>, 20% CH<sub>4</sub>, 10% CO<sub>2</sub>, 10% N<sub>2</sub>  
 Mix 2 = 40% H<sub>2</sub>, 30% CH<sub>4</sub>, 15% CO<sub>2</sub>, 15% N<sub>2</sub>  
 Mix 3 = 20% H<sub>2</sub>, 40% CH<sub>4</sub>, 20% CO<sub>2</sub>, 20% N<sub>2</sub>

- Gaseous mixtures

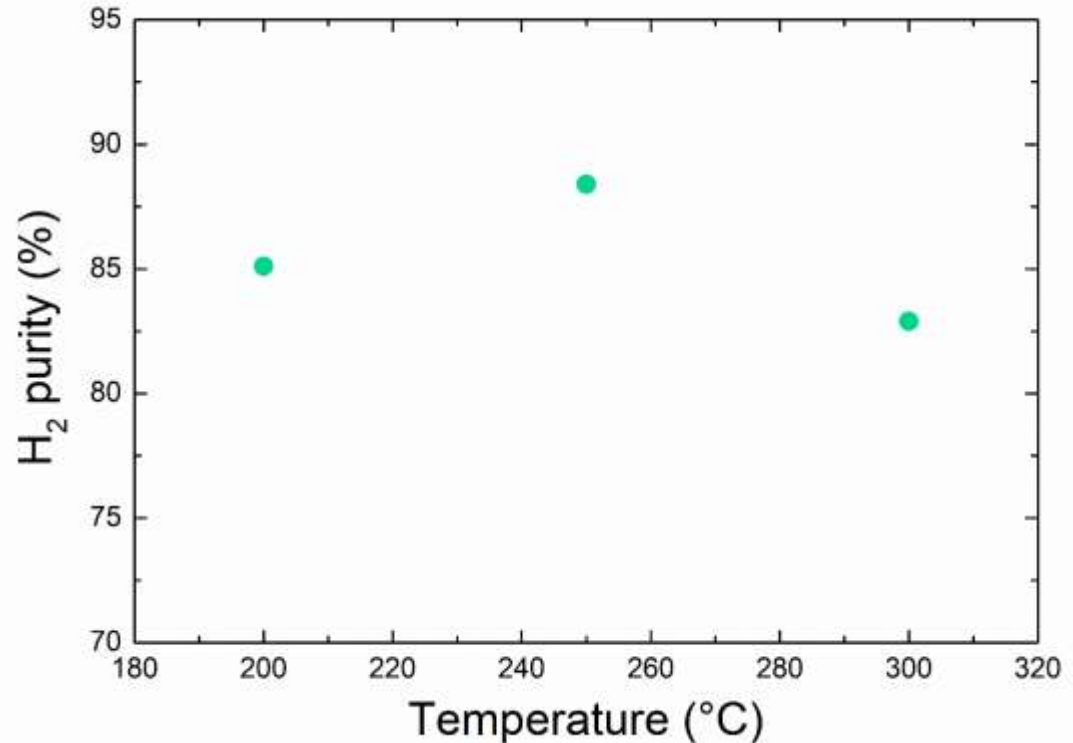
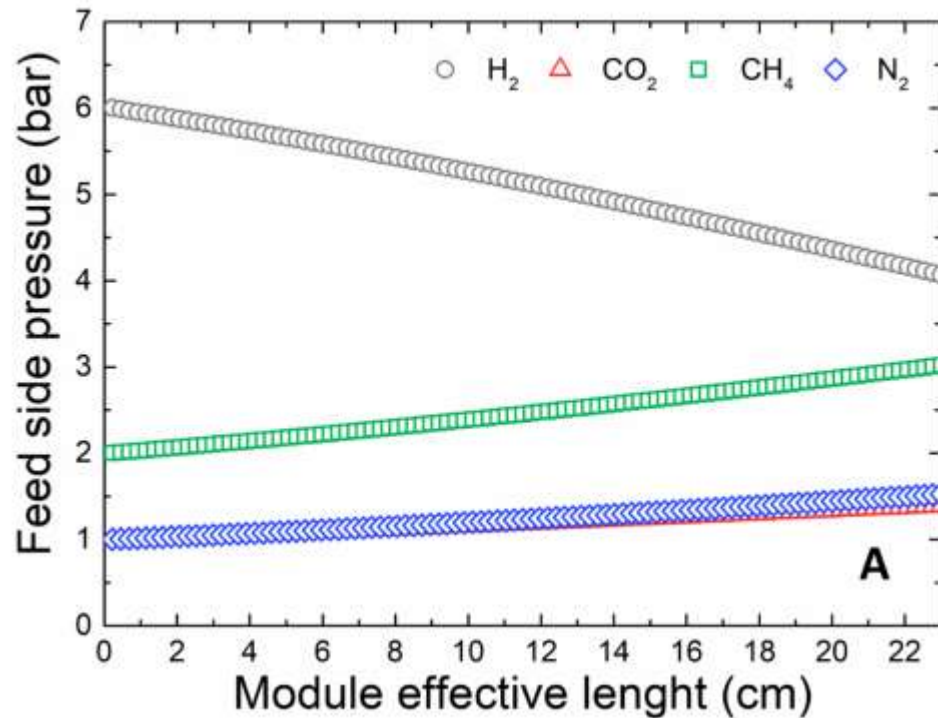


# Gas separation performance

## Mixed Gas

Mix 1 = 60% H<sub>2</sub>, 20% CH<sub>4</sub>, 10% CO<sub>2</sub>, 10% N<sub>2</sub>  
10 bar feed pressure, 3000 ml/min as total feed flow

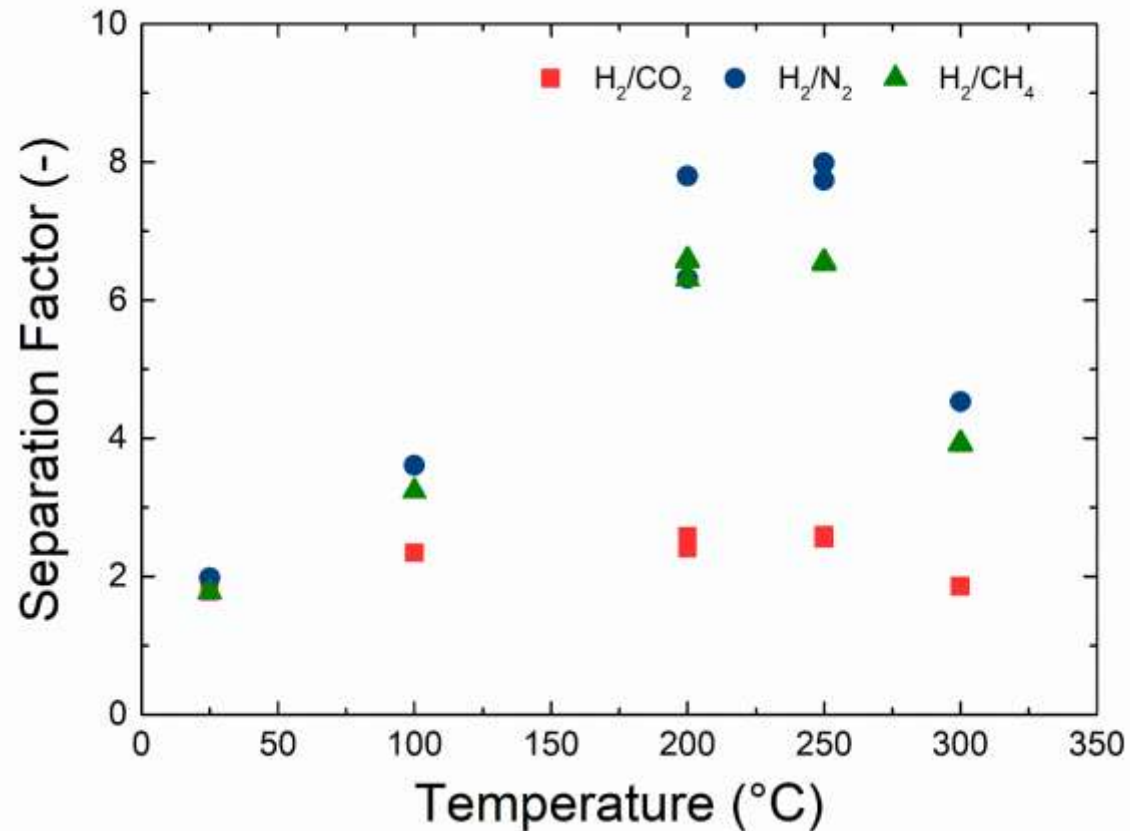
- Gaseous mixtures – module behavior





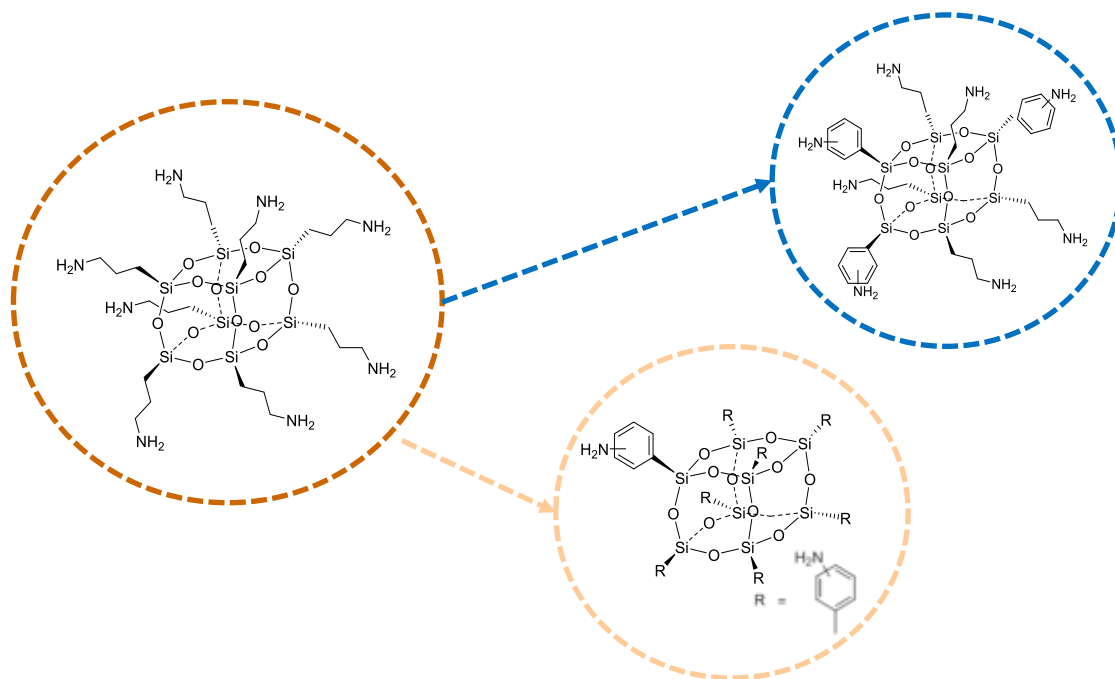
# Gas separation performance

- Gaseous mixtures – temperature effect on selectivity

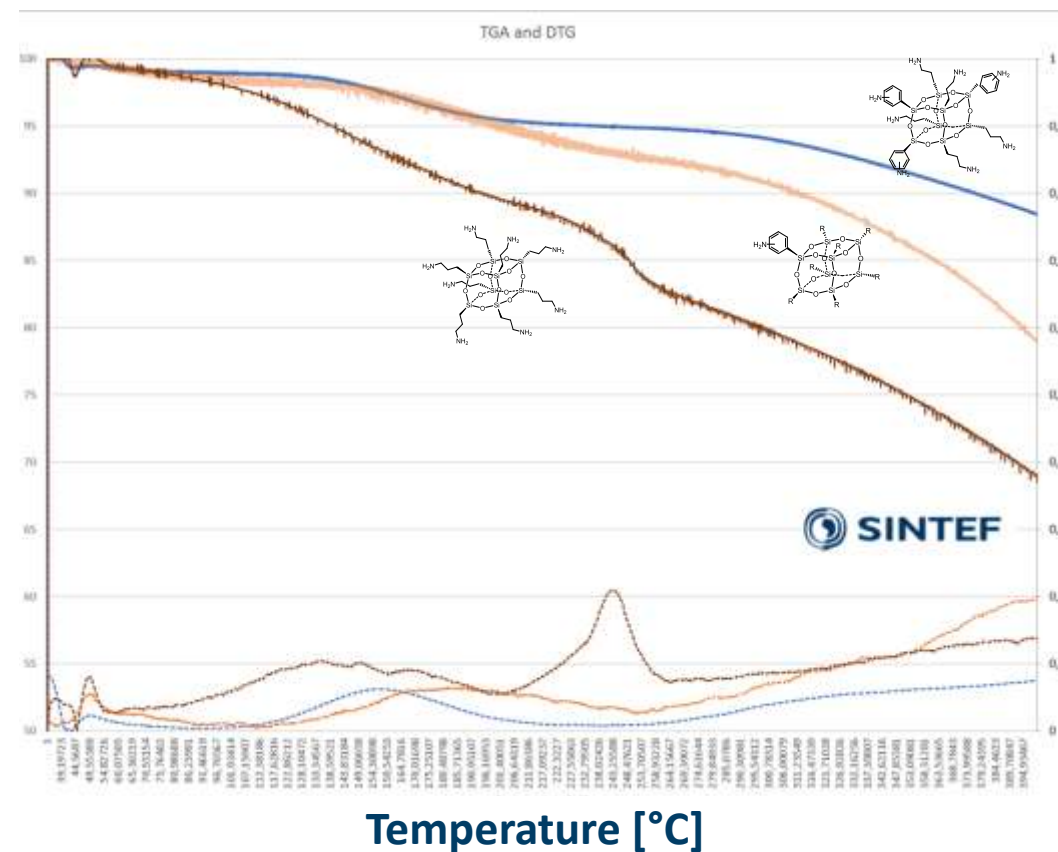


# Optimization of POSS building block

- Modification of amino-moieties
- Aromatic bridges are electropositive moieties enhancing thermal stability
- Mixed POSS particles showed the best performance

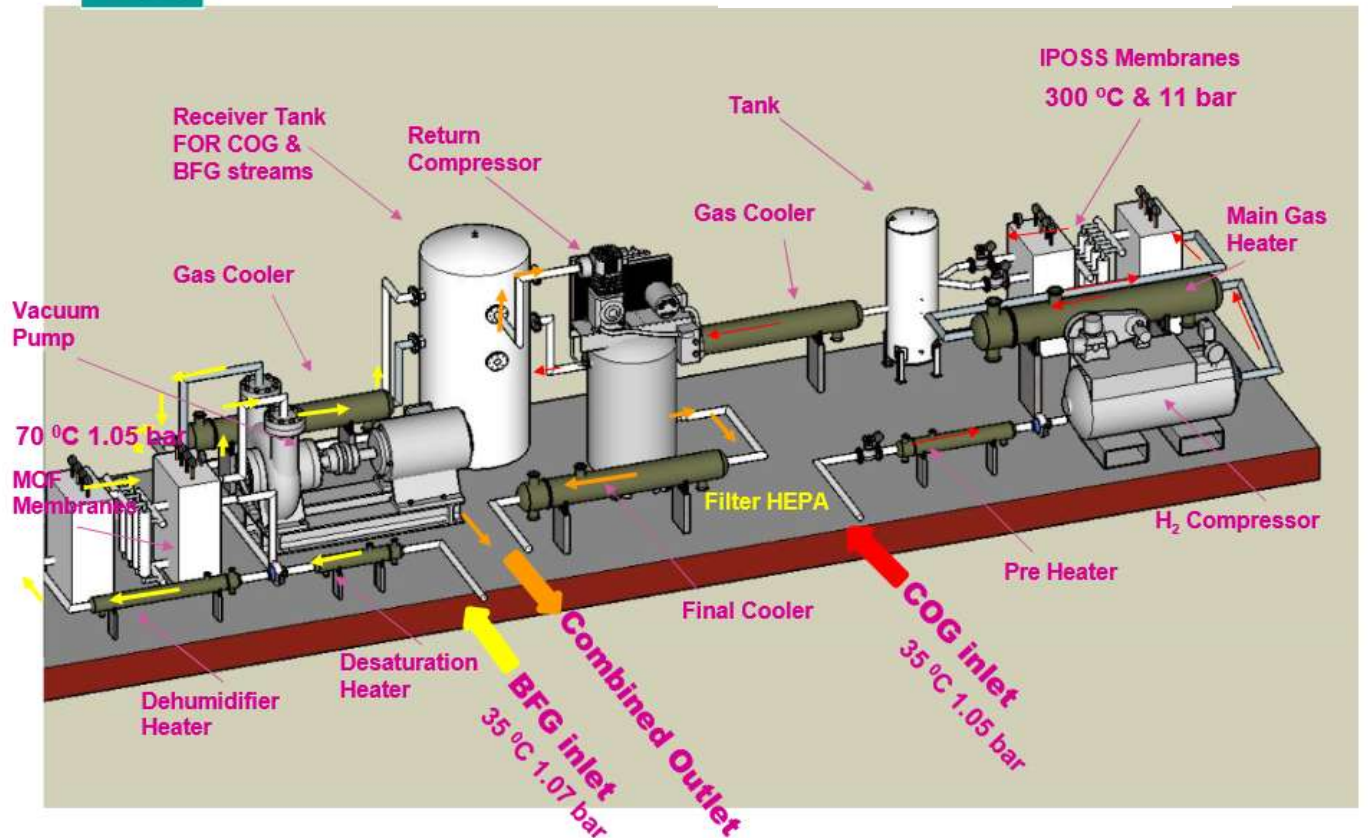


Weight [%]



# Demonstration at TRL6

- Arcelor Mittal iPOSS one-stage membrane system
  - Gent, Belgium
  - Clean coke gas:  $\sim 100 \text{ Sm}^3/\text{h}$
  - Membrane area:  $\sim 1 \text{ m}^2$
  - Date: 2020 – 2021
- On-going activities
  - Process design review
  - Conceptual and system design
  - Construction & installation



# Conclusions

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- Hyper-crosslinked nanoscale iPOSS membranes show promising performance for H<sub>2</sub> purification applications
- GENESIS aims to demonstrate the technology for the purification of H<sub>2</sub> from a coke gas (utilized in the steelmaking industry)
  - Membrane fabrication upscaled from  $\gamma$ -alumina coated discs to single channel tubular membranes
  - Gas permeation performance thoroughly investigated
  - Different types of amine-modified iPOSS have been produced in order to increase the thermal stability of the selective layer for  $T > 300$  °C
  - Demonstration activities onsite at Arcelor Mittal planned for 2020



# Acknowledgements

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- This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 760899. This publication reflects only the author's views and the European Union is not liable for any use that may be made of the information contained therein





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