New exploratory testing conditions to understand the gas transfer mechanisms through VIPs' barriers

Mathias BOUQUEREL[1]
Thierry DUFORESTEL[2]

[1]Siniat International, Avignon, France
[2]EDF R&D, Les Renardières, France

IVIS 2013
19-20th September 2013
Dübendorf, Switzerland

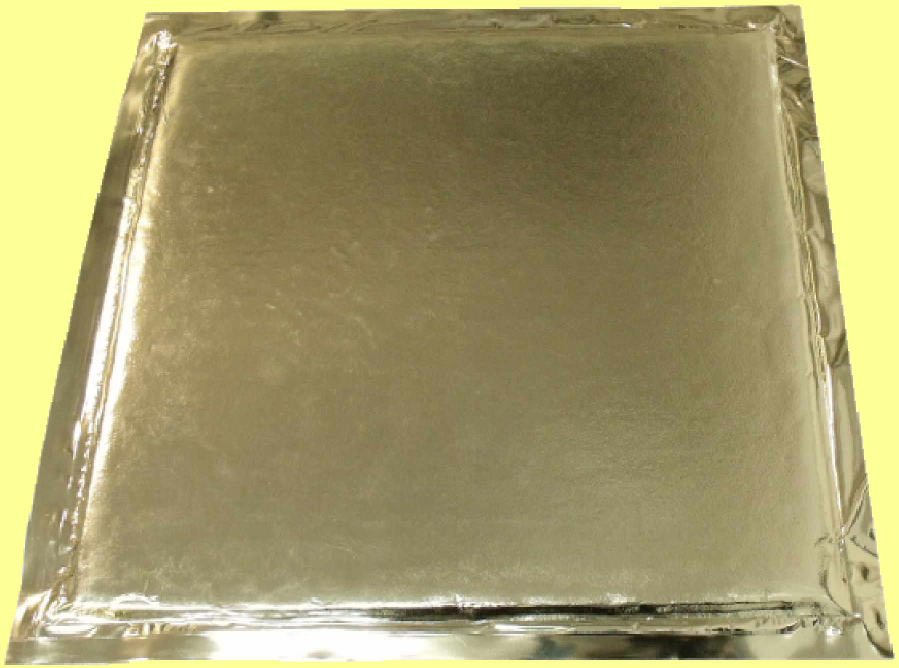
I. Introduction

II. Gas permeation through VIP barrier envelop: modeling

III. New exploratory conditions for gas permeation measurements

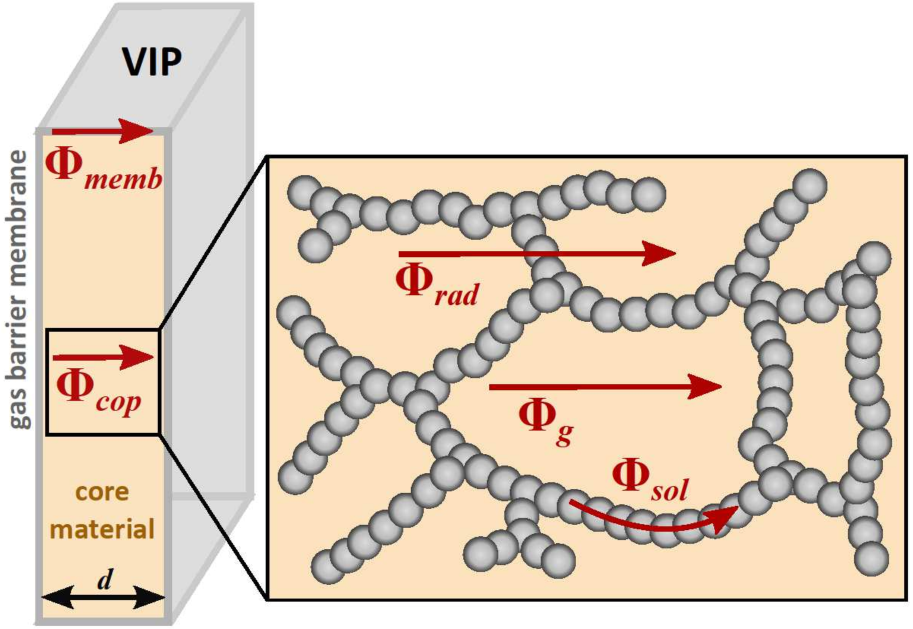
IV. Conclusion & outlook

I. Introduction: VIP thermal performance & aging



Thermal performance of VIPs: parallel fluxes model

- Heat fluxes in the core material:
  - Radiative heat transfer Φ_{rad}
  - Solid conduction Φ_{sol}
  - Gaseous heat transfer Φ_{g}
- Thermal bridge through the edge (gas barrier membrane) Φ_{memb}



- Parallel fluxes
- Independent fluxes

→ Elementary conductivities can be summed

$$\lambda_{tot} = \lambda_{rad} + \lambda_{sol} + \lambda_{g} + \lambda_{memb}$$

dry core moisture

$$\lambda_{tot} = \lambda_{rad} + (\lambda_{sol,dry} + \lambda_{u}) + \lambda_{g} + \lambda_{memb}$$

Thermal performance of VIPs: parallel fluxes model 2/2

$$\lambda_{tot} = \lambda_{rad} + (\lambda_{sol,dry} + \lambda_u) + \lambda_g + \lambda_{memb}$$

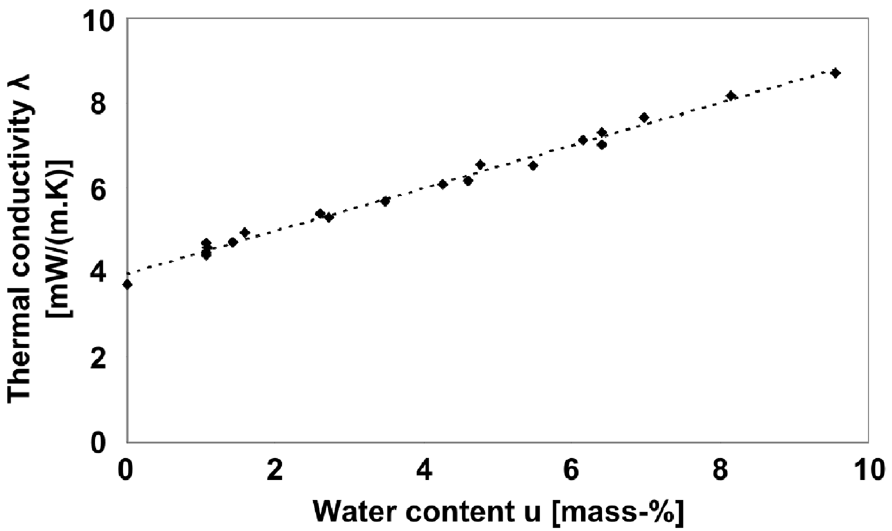
$\lambda_{rad} \simeq$ **1 mW/(m.K)**

$\lambda_{sol} \simeq$ **3 mW/(m.K)**

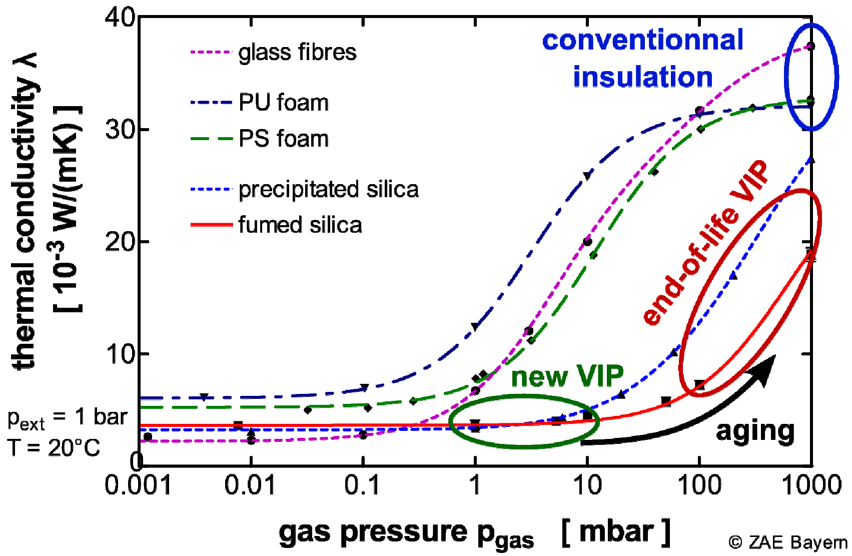
$\lambda_u \simeq$ **0 - 3 mW/(m.K)**

$\lambda_g \simeq$ **0 - 15 mW/(m.K)**

$\lambda_{memb} \simeq$ **1 mW/(m.K)**



[IEA Annex 39, 2005]



[IEA Annex 39, 2005]

Thermal performance & aging mechanism

$$\lambda_{tot} = \lambda_{rad} + (\lambda_{sol,dry} + \lambda_u) + \lambda_g + \lambda_{memb}$$

$\lambda_{rad} \simeq 1$ mW/(m.K)

$\lambda_{sol} \simeq 3$ mW/(m.K)

$\lambda_u \simeq 0 - 3$ mW/(m.K)

$\lambda_g \simeq 0 - 15$ mW/(m.K)

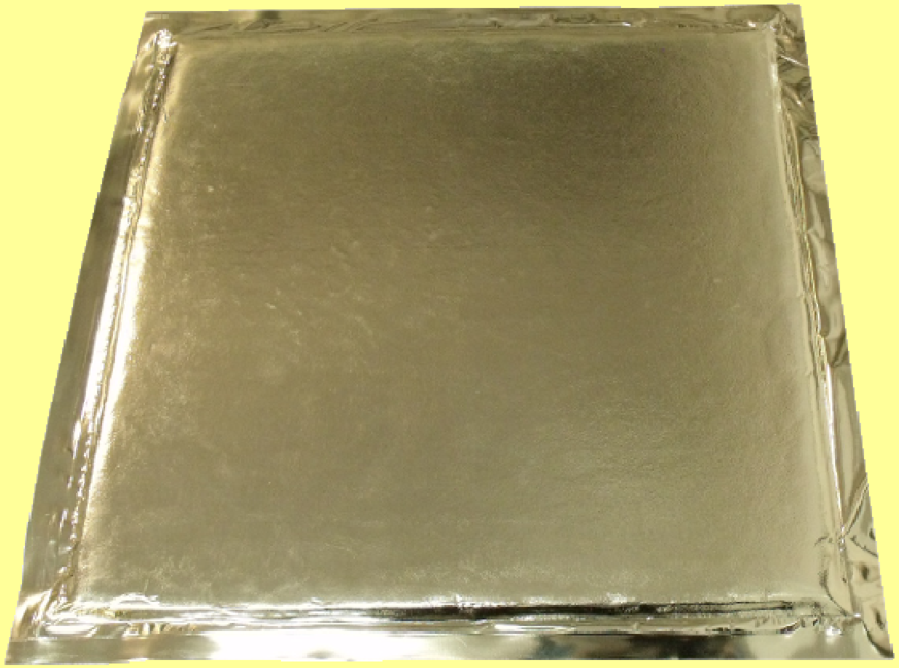
$\lambda_{memb} \simeq 1$ mW/(m.K)

Core material water content $u = f_1(t)$

Internal pressure $p = f_2(t)$

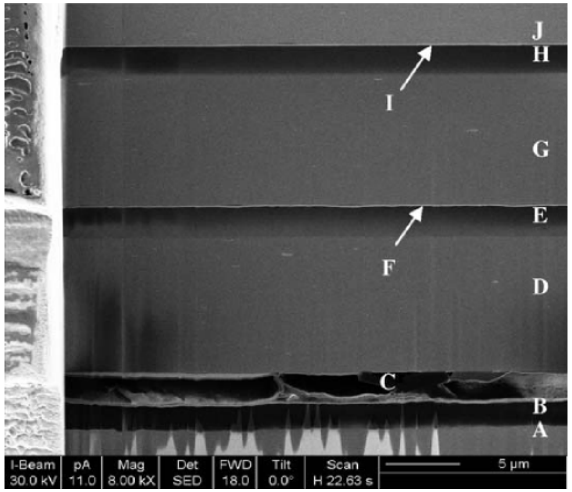
- The gas permeation through VIP envelopes should be quantified for the prediction of the thermal performance over the whole lifetime
- Dry air and water vapor should be quantified separately (not the same impact)

II. Gas permeation through VIP barrier envelop: modeling



State-of-the art gas permeation modeling
Multilayer membrane scale

[BRUNNER et al, Surface & Coatings Technology, vol 200, 2006]



J	PET	Polyethylene terephtalate	12 µm
I	AL	Aluminium	100 nm = 0.1 µm
H	PU	Polyurethane	2 µm
G	PET	Polyethylene terephtalate	12 µm
F	AL	Aluminium	100 nm = 0.1 µm
E	PU	Polyurethane	2 µm
D	PET	Polyethylene terephtalate	12 µm
C	AL	Aluminium	100 nm = 0.1 µm
B	PU	Polyurethane	2 µm
A	LDPE	Low Density Polyethylene	50 µm

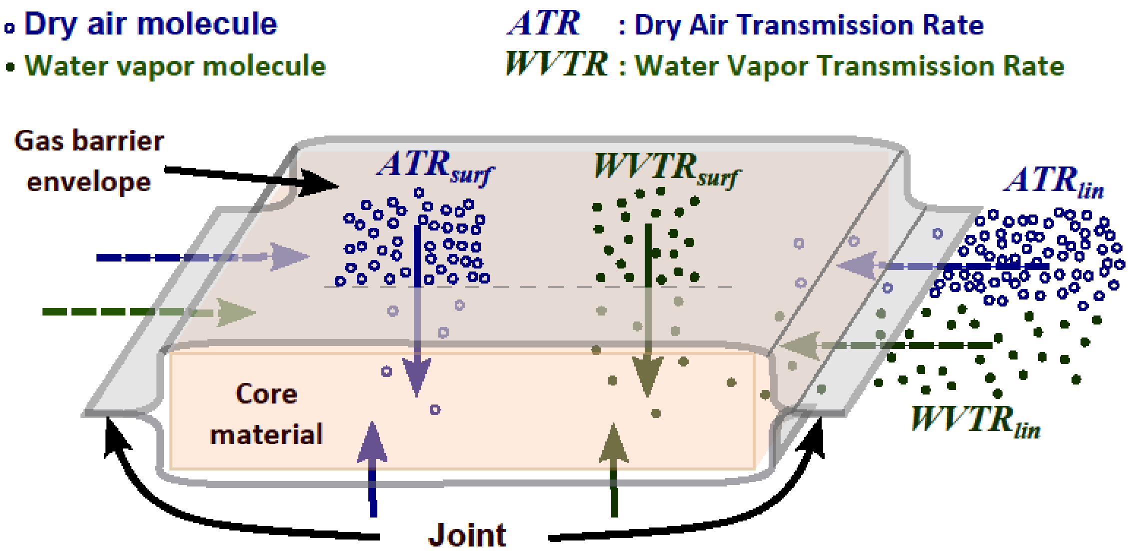
- At multilayer membrane scale: Sorption-Diffusion model (SD model)
  - Partial pressure gradient Δp_i is the driving force for gas transmission

$$GTR_{surf} = \Pi_{surf} \Delta p_i$$

  - Permeance Π is temperature dependant

$$\Pi_i = \Pi_i^0 \exp\left(-E_a / RT\right)$$

State-of-the art gas permeation modeling
VIP scale



- At whole VIP scale: 4 contributions to gas transfer
  - Two locations (faces & edges)
$$ATR_{tot} = A \cdot ATR_{surf} + P \cdot ATR_{lin}$$
$$WVTR_{tot} = A \cdot WVTR_{surf} + P \cdot WVTR_{lin}$$
  - Two gases (dry air and water vapor) $GTR_{tot} = ATR_{tot} + WVTR_{tot}$

- Final expression of water vapor or dry air permeation rate
$$ATR_{tot} = A \cdot \Pi_{air,surf} \cdot \Delta p_{air} + P \cdot \Pi_{air,lin} \cdot \Delta p_{air}$$
$$WVTR_{tot} = A \cdot \Pi_{vap,surf} \cdot \Delta p_{vap} + P \cdot \Pi_{vap,lin} \cdot \Delta p_{vap}$$

Alternative permeation model: Dual Pressure model

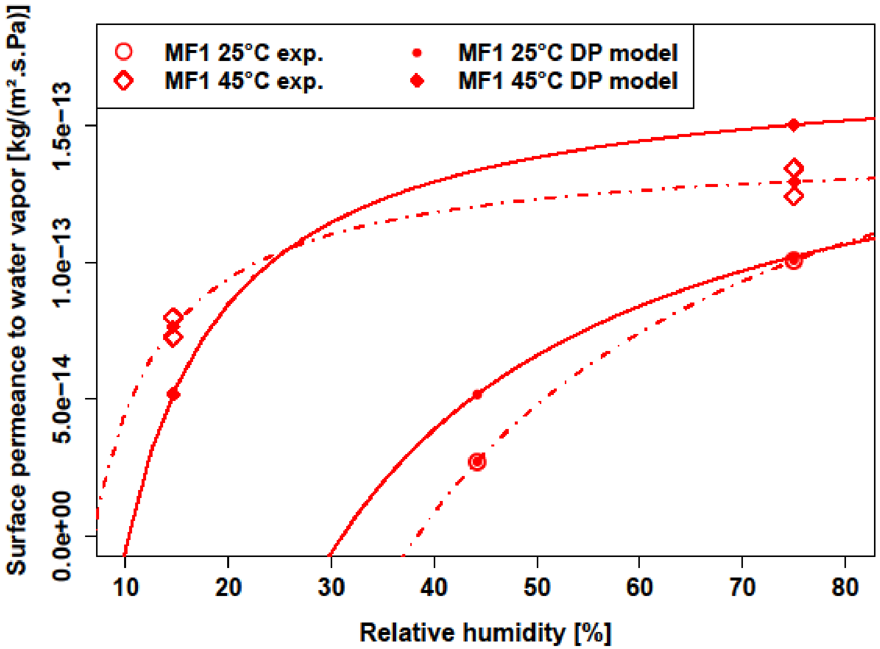
- Arguments in favor of a coupled model for mass transfer
  - Thermodynamics of irreversible processes
  - Competition or cooperation between gas components for sorption and diffusion
  - Influence of relative humidity on dry air transmission rates

- Alternative model proposed: the Dual Pressure model (DP)
  - Takes into account partial pressure gradient Δp_i and the total pressure gradient Δp_{tot}
  - Gas transmission rate: $$GTR_{surf} = K_1 \Delta p_i + K_2 \Delta p_{tot}$$
  - Apparent permeance: $$\Pi_{surf} = -\frac{GTR_{surf}}{\Delta p_i} = K_1 + K_2 \frac{\Delta p_{tot}}{\Delta p_i}$$

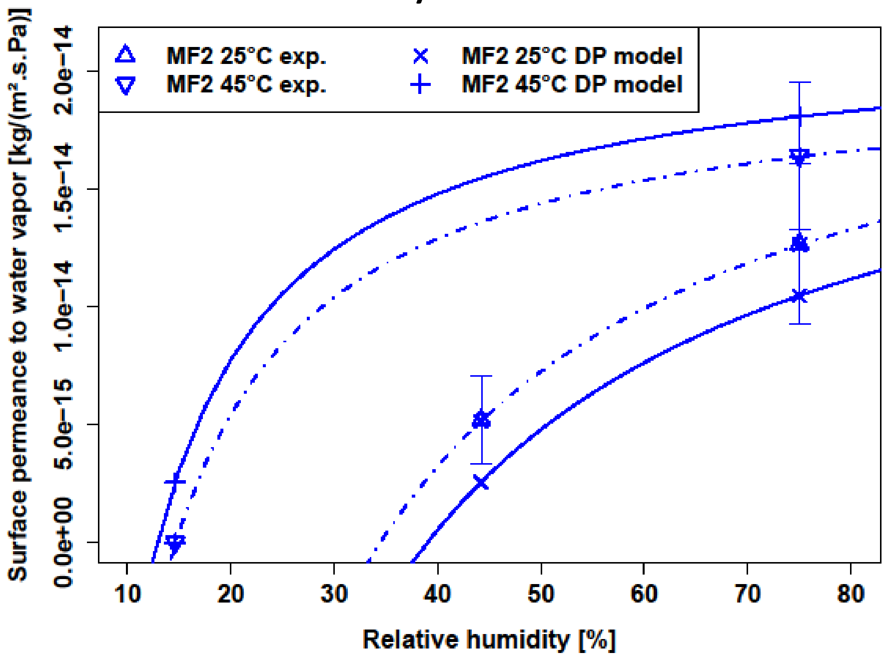
- Coefficients K_1 et K_2 can be identified on exp. data from IEA Annex 39 (VIP aging in climatic rooms)

Water vapor surface permeances from the DP model

Multilayer Barrier 1

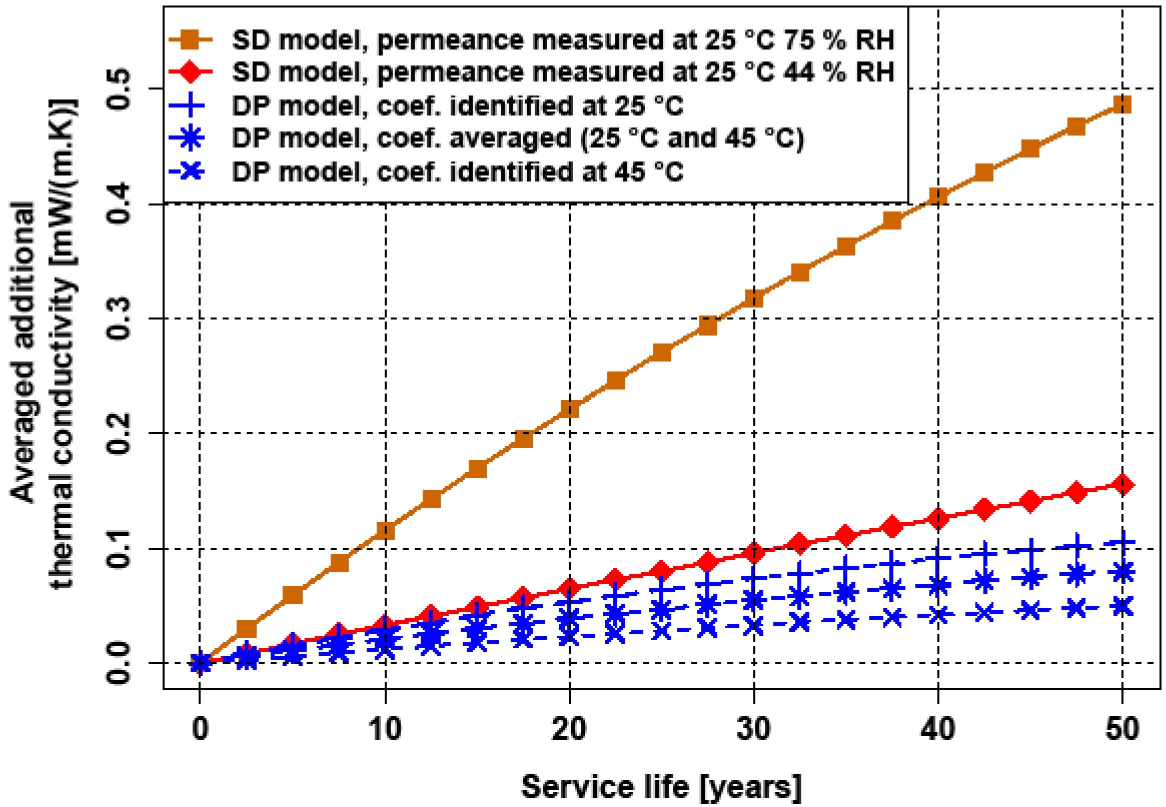


Multilayer Barrier 2



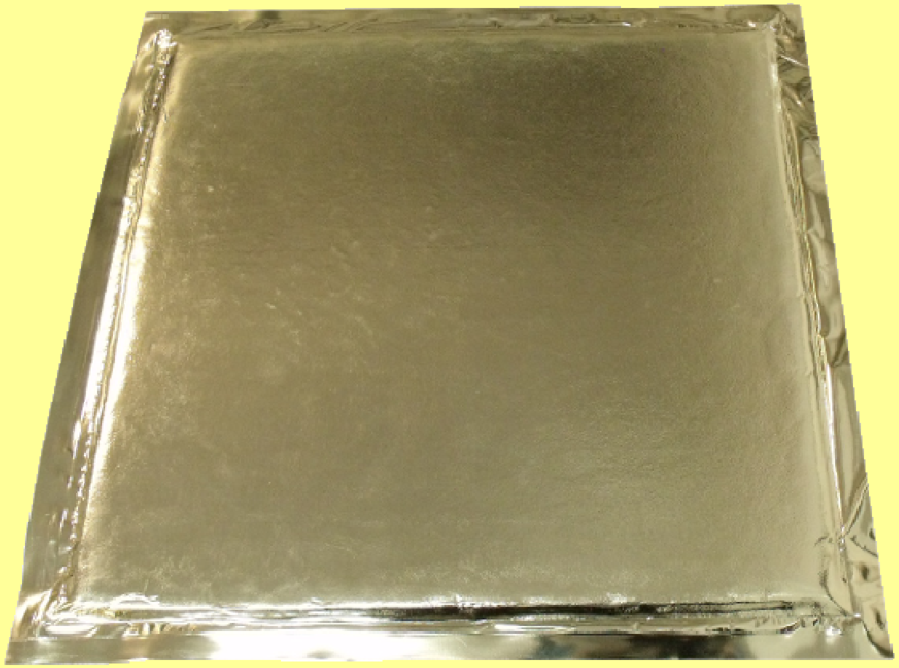
- Dotted lines: the parameter set (K_1 , K_2) is identified at each temperature
- Solid lines: the parameter set (K_1 , K_2) is averaged over all temperatures

Model behaviors:
Sorption-diffusion (SD) vs. dual pressure (DP)



- Π , K_1 and K_2 identified on exp. data from IEA Annex 39
- Climatic conditions during service: T = 25 °C, φ = 44 %
- DP model less sensitive to experimental conditions identification

III. New exploratory conditions for gas permeation measurements



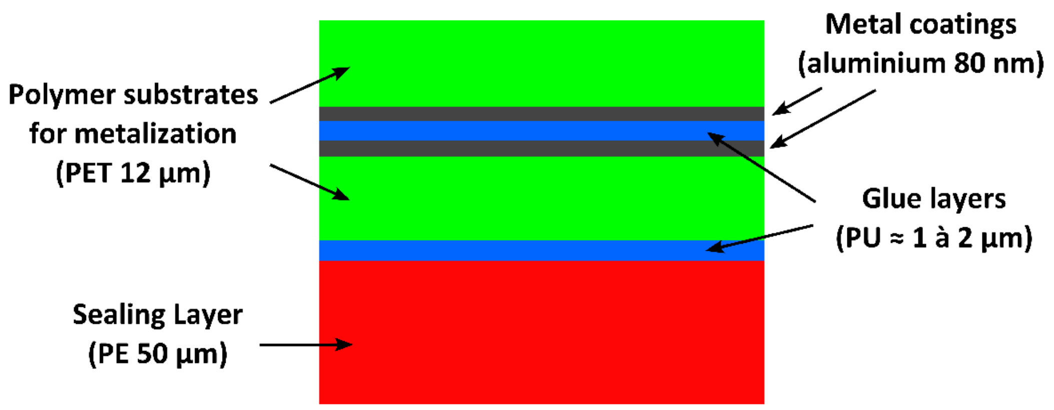
Exploratory experimental plan in new conditions

- Goal: to measure the influence of the water vapor molar concentration, at constant water vapor partial pressure → influence of the total pressure ?

Condition	Temperature T [°C]	Relative humidity φ [%]	Total pressure p_{tot} [Pa]	Water vapor molar concentration X_{vap} [%]
1	48	65 %	7 270	100 %
2	48	65 %	24 000	30 %
3	48	65 %	100 000	7 %

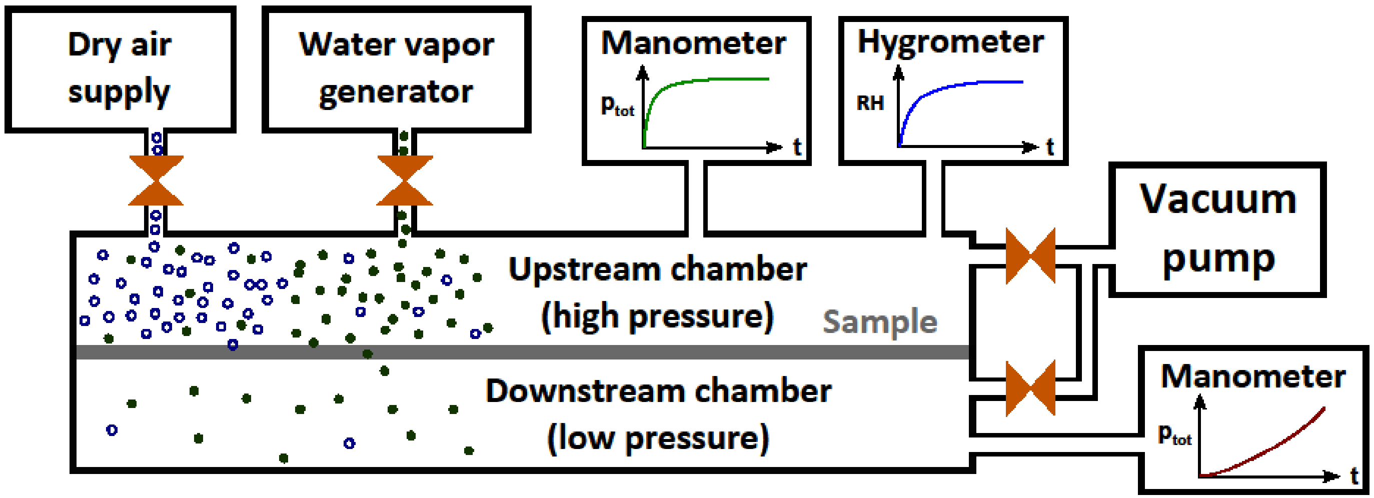
- Two experimental methods:
  - → Manometric method: measurement of the pressure increase
  - → VIP aging in climatic rooms: measurement of the mass increase

- Membrane MF8 : bilayer of an Al-coated PET (12 µm PET + 80 nm Al) + sealing layer PE 50 µm

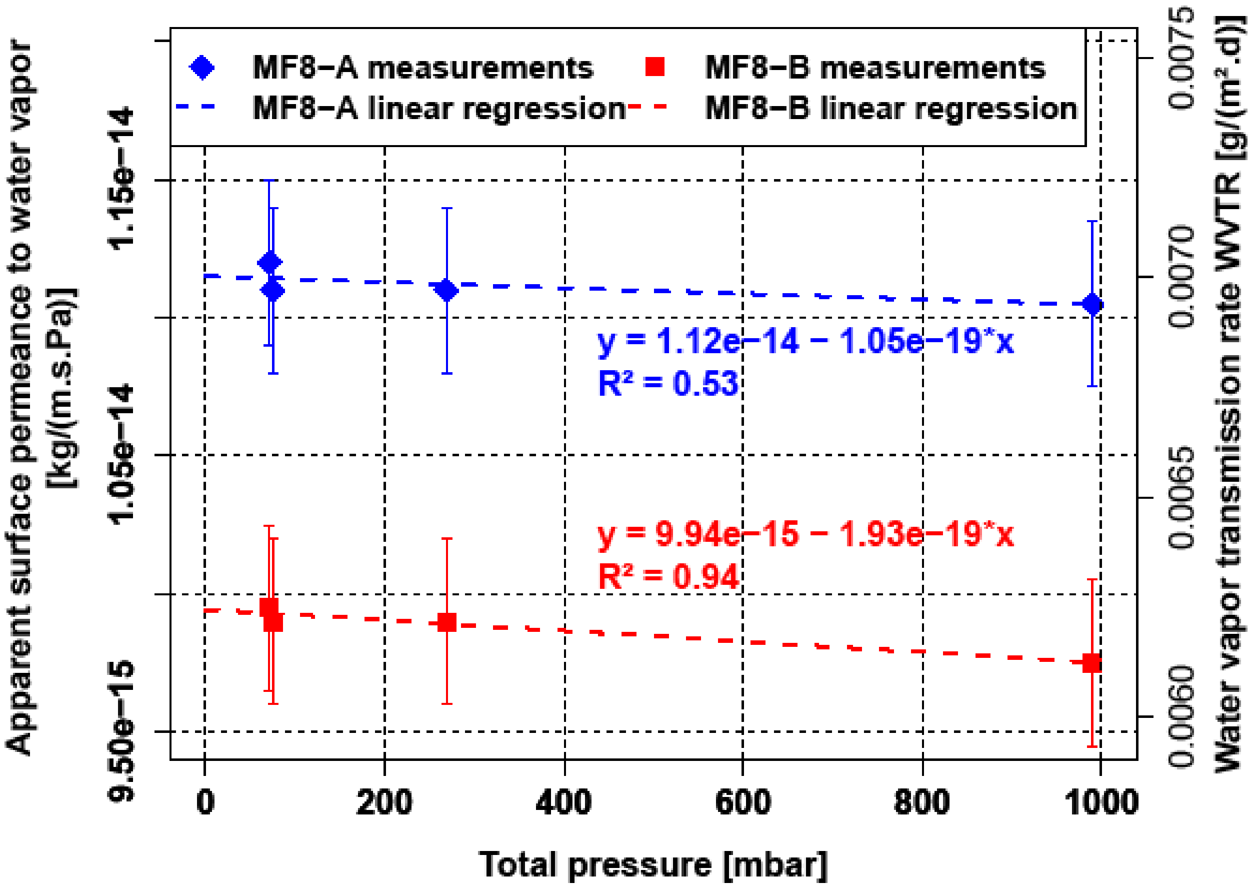


Experimental method 1: Manometric method at the scale of the barrier membrane

- Humid air configuration (modified “Deltaperm” device)
  - Upstream chamber : mixture of water vapor + dry air
  - Hard to measure the dry air transmission rate *ATR* ≃ 100 times lower than WVTR
  - The whole gas transmission rate *GTR* is measured, it is assimilated to the water vapor transmission rate *WVTR* (air transmission rate *ATR* negligible)



Manometric method measurements in humid air configuration: surface permeance to water vapor



- Very slight influence of the total pressure: total variation < 2 %

Experimental method 2: whole VIP scale
Accelerated gas permeation

- Accelerated aging of VIPs in climatic rooms, at high temperature & humidity

- Raw measurement data: mass and pressure increase over time
- What is needed for accurate measurements ?
  - Accurate measurements of core material hygroscopic properties
  - Good control of climatic conditions (temperature, RH, total pressure)
  - Large number of samples with various perimeter / area ratios
  - Combined measurement of mass and pressure increase
  - Long measurement period for stabilization (several weeks)

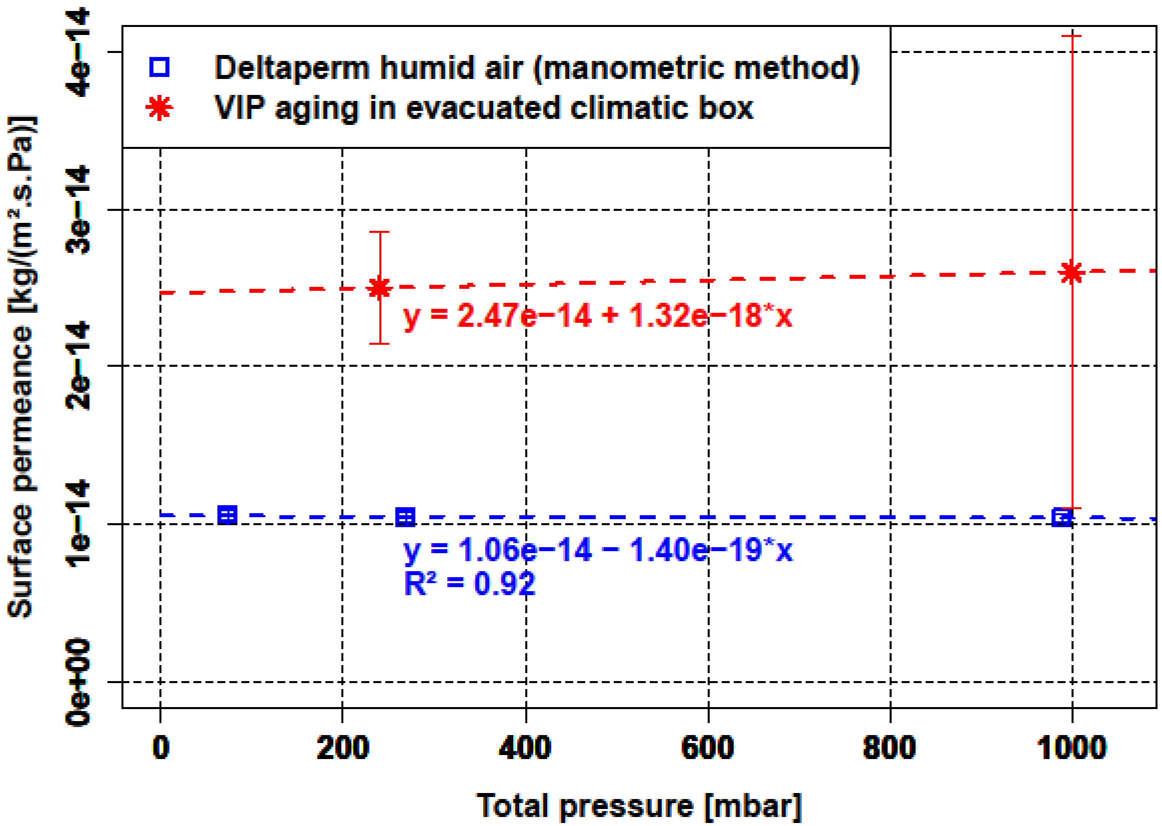
Accelerated VIP aging measurements: Surface and linear permeance to water vapor

Membrane and experimental condition	Total pressure p_{tot} [mbar]	Surface permeance Π_{surf} [kg/m².s.Pa]	Linear permeance Π_{lin} [kg/m.s.Pa]
MF8 / Condition 2	240 mbar	**$2{,}5 \times 10^{-14}$** $\pm 4 \times 10^{-15}$	**$9{,}2 \times 10^{-16}$** $\pm 10^{-15}$
MF8 / Condition 3	1 000 mbar	**$2{,}6 \times 10^{-14}$** $\pm 1{,}5 \times 10^{-14}$	**$2{,}7 \times 10^{-15}$** $\pm 3 \times 10^{-15}$
MF2 (IEA Annex 39) T = 45°C et φ = 75 % HR	1 000 mbar	$1{,}6 \times 10^{-14}$	3×10^{-15}

- The influence of the total pressure on the surface permeance seems to be negligible
- The linear permeance seems to be an increasing function of the total pressure

Membrane	Linear permeance to water vapor $\Pi_{lin,vap}$ [kg/m.s.Pa]	Linear permeance to dry air $\Pi_{lin,air}$ [kg/m.s.Pa]
MF8	$5{,}2 \times 10^{-16}$	$1{,}7 \times 10^{-16}$
MF2 (IEA Annex 39)	3×10^{-15}	8×10^{-19}

Experimental results: surface permeance to water vapor from both measurement methods



- Influence of the total pressure: negligible for both methods
- Influence of the method: factor 2.5 for the same barrier membrane

Influence of the total pressure

$$\Pi_{vap} = K_{1,vap} + K_{2,vap}\frac{\Delta p_{tot}}{\Delta p_{vap}} = K_{1,vap}\left(1 + \frac{K_{2,vap}}{K_{1,vap}X_{vap}}\right)$$

- Experimental data from IEA Annex 39

$$\frac{K_{2,vap}}{K_{1,vap}} \approx -0{,}01 \quad \& \quad X_{vap} \in [1{,}4\% - 7{,}2\%] \Rightarrow \frac{K_{2,vap}}{K_{1,vap}X_{vap}} \in [14\% - 70\%]$$

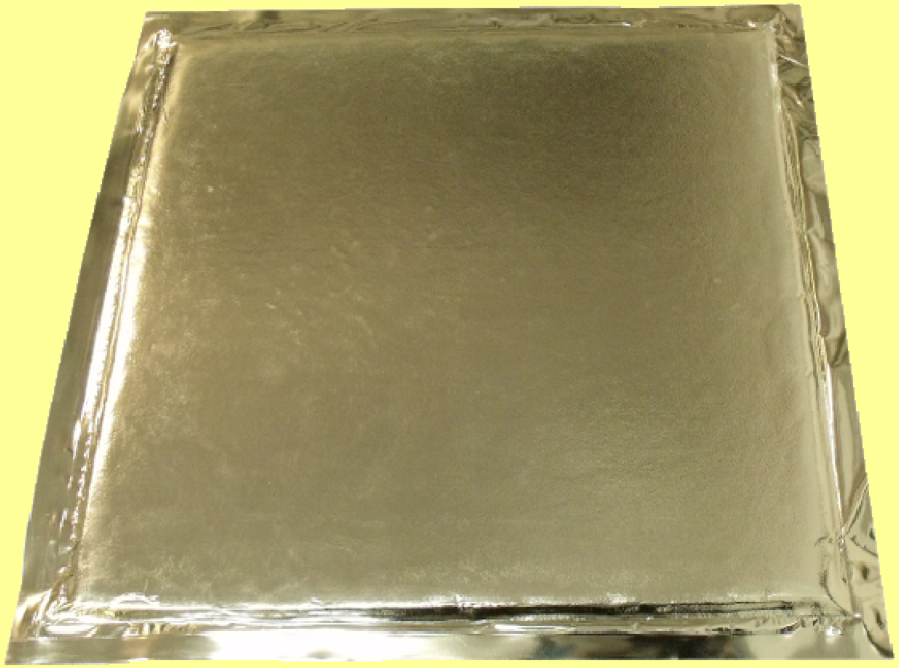
- Experimental conditions used here: T = 48 °C, ϕ = 65 % HR

$$\frac{K_{2,vap}}{K_{1,vap}} \approx -0{,}001 \quad \& \quad X_{vap} \in [7{,}2\% - 100\%] \Rightarrow \frac{K_{2,vap}}{K_{1,vap}X_{vap}} \in [0{,}1\% - 1{,}4\%]$$

- The influence of the total pressure may still exist, but may not be measurable in our conditions

  ➔ Dry conditions experiments required to estimate the potential influence

IV. Conclusion



Conclusion

Main aging phenomenon: **gas permeation through VIP envelopes**

→ Sorption-Diffusion model as a reference model, partial pressure as transfer driver

⇓ ⇓

Alternative model for mass transfer: Dual Pressure model (DP)

Takes into account both ***partial pressure*** and ***total pressure***

→ Both models are consistent with existing experimental data (but not extensive)

⇓ ⇓

Need for new experiments: ***influence of the total pressure ?***

→ **Two methods** for exploratory experimental plan

⇓ ⇓

Experimental results

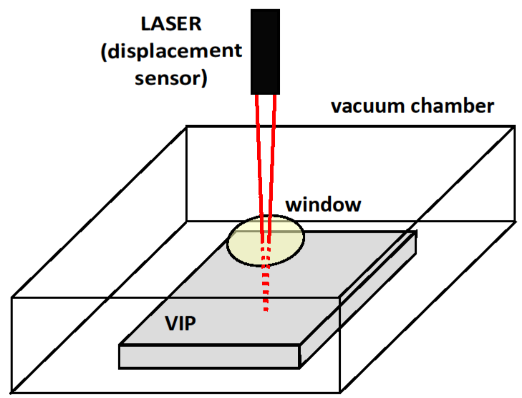
- No influence of the total pressure on surface permeance → exp. conditions ?
- Influence of the total pressure on linear permeance → uncertainty and air / vapor
- Influence of the method → sample integrity // sample size

Outlook

Three main matters for experimental exploration:

- Influence of the total pressure on ***surface permeance*** (VIPs' faces)
  - → Dual pressure model for **moderate temperature and humidity**:
  - → Measurements over **long periods** with **high precision balance**

- Influence of the total pressure on ***linear permeance*** (VIPs' edges)
  - → Accuracy of VIP aging measurements: **high precision balance** and **long periods**
  - → **Internal pressure measurement** through the lift-off procedure



- Influence of the experimental method: where does the factor 2.5 come from ?
  - → **Envelope sample integrity** after VIP manufacturing ? (samples from manufactured VIP)
  - → Sample size and selection → long term measurements on small VIPs
  - → Other causes: influence of the gas mixture on the low pressure side

Thanks for your attention !