Development of novel opaque and transparent barrier films for VIP-encapsulation
Part-II: Barrier film production for VIPs

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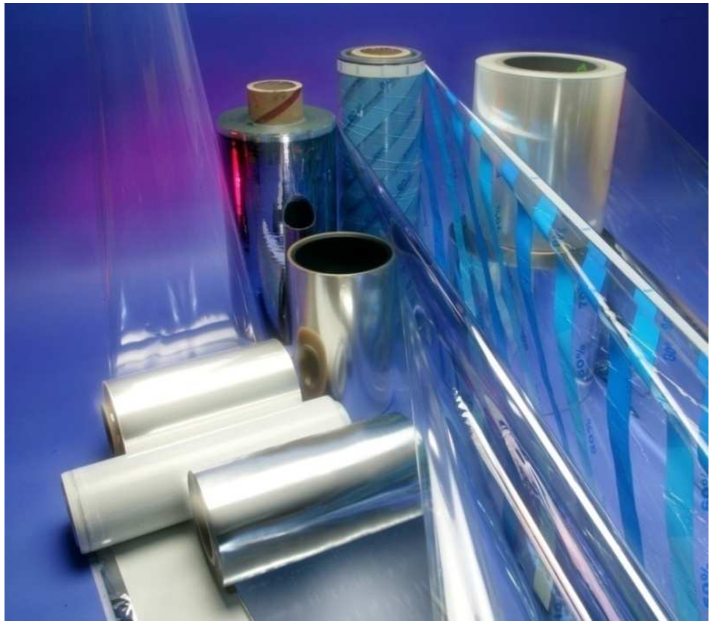

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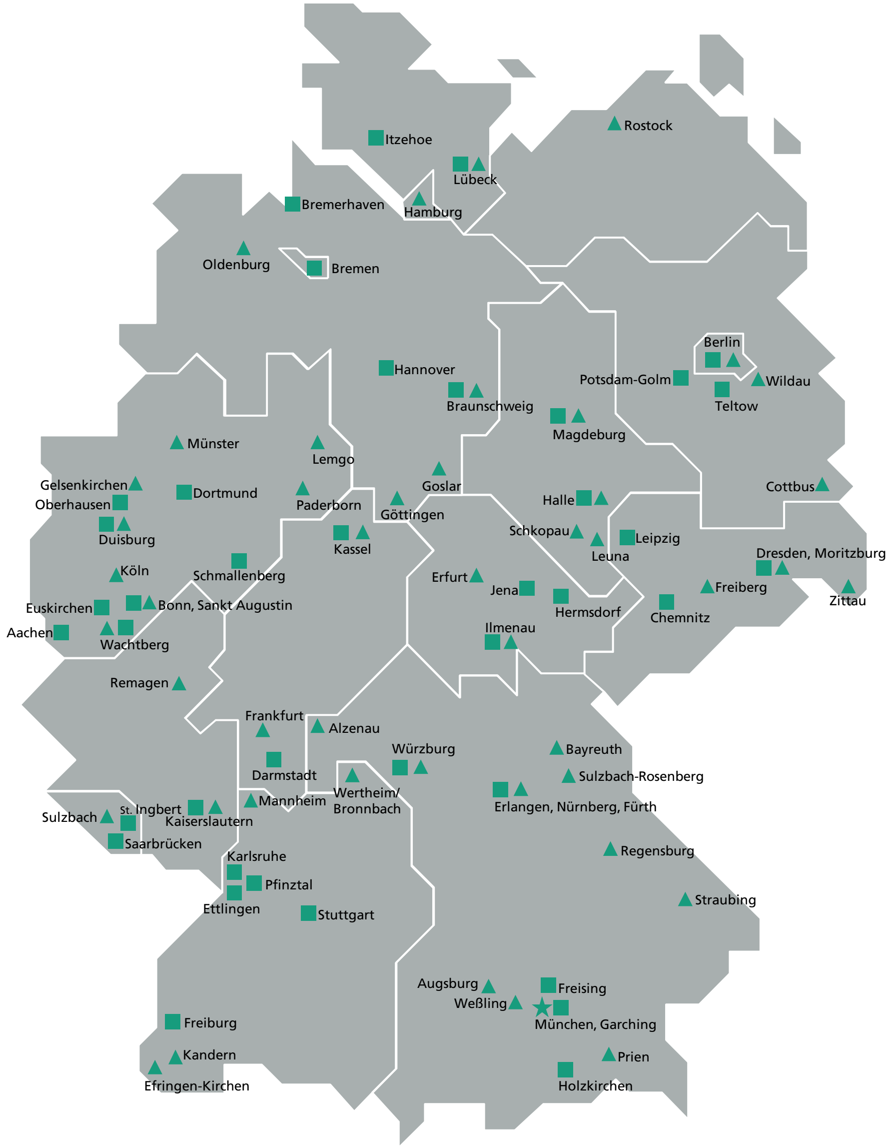

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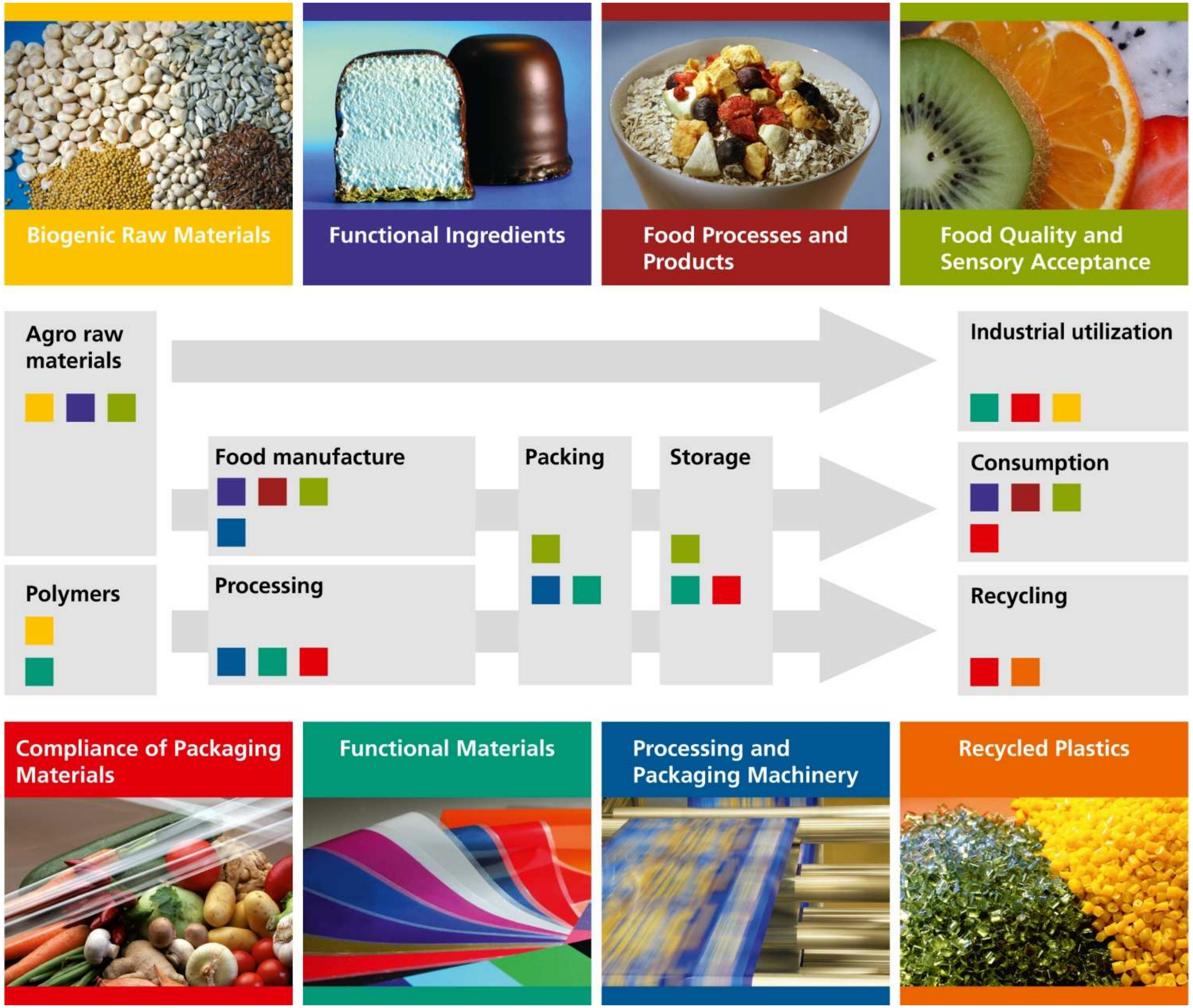

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Development of novel opaque and transparent barrier films for VIP-encapsulation – Part-II: Barrier film production for VIPs

- Barrier requirements of vacuum insulation panels (VIPs)
- State-of-the-art barrier films for VIPs
- Development of novel barrier films for VIPs
- Results for opaque barrier films
- Results for transparent barrier films
- Summary and outlook

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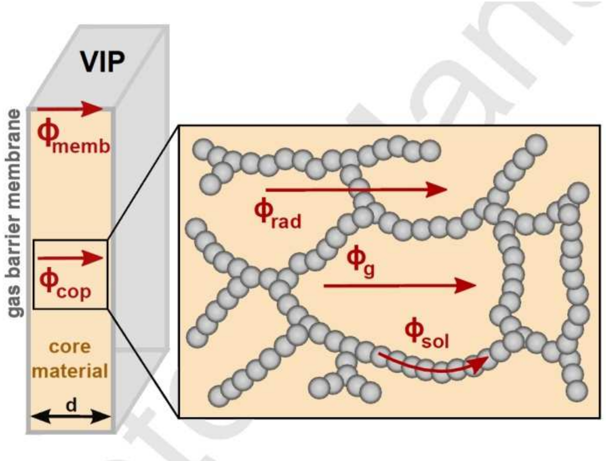
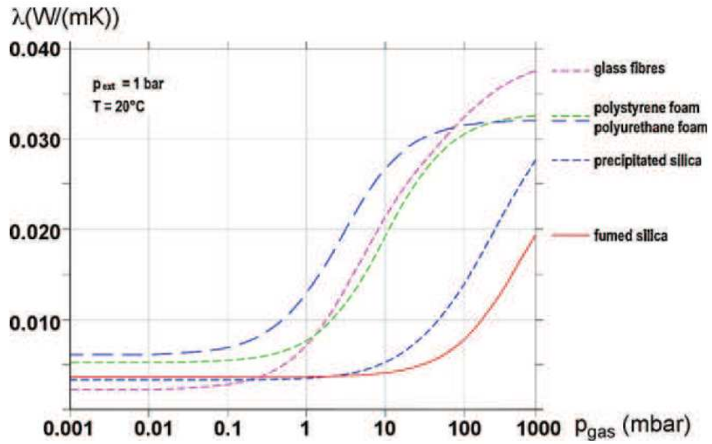
Heat transport in VIPs

- **Contributions to the heat transport**
  - Heat radiation (Φ_{rad})
  - Heat conductance within the solid matrix of the core material (Φ_{sol})
  - Heat conductance through the gas within the pores of the core material (Φ_g)
- **Φ_g increasing with gas pressure**

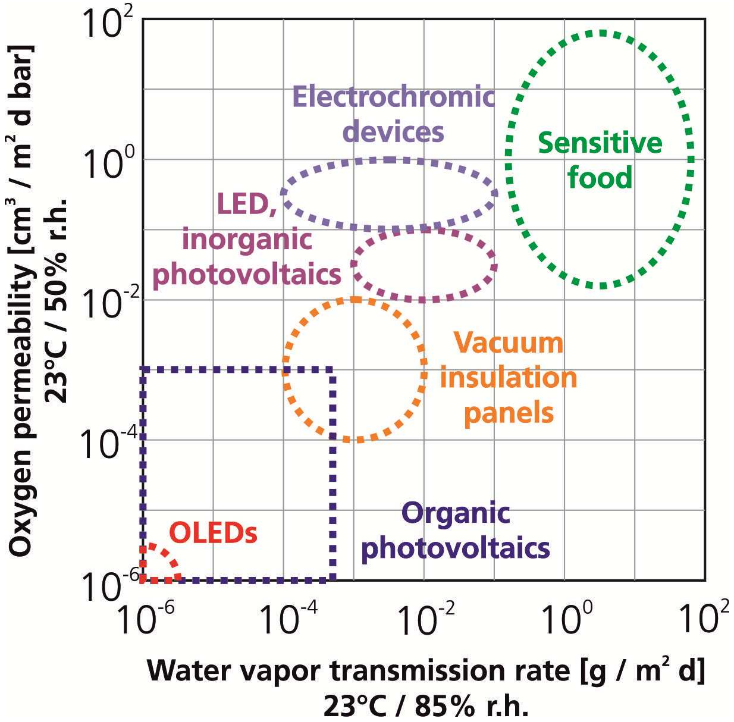
⇒ **VIP encapsulation:** protection against oxygen, nitrogen and water vapour

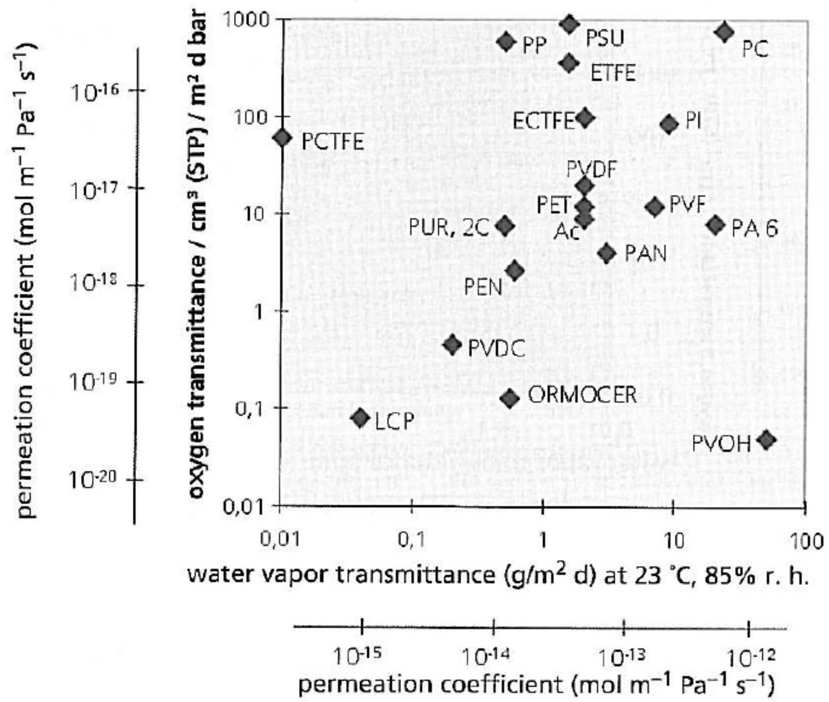




Left: M. Bouquerel et al., Energy and Buildings (2012), doi:10.1016/j.enbuild.2012.07.034
Right: R. Baetens et al., Energy and Buildings 42 (2010), 147–172

VIP encapsulation





Aluminum foil

- Nearly impermeable
- Disadvantage: Heat bridge effect

Polymer based film

- Disadvantage: Barrier performance not sufficient

⇒ Application of barrier layers

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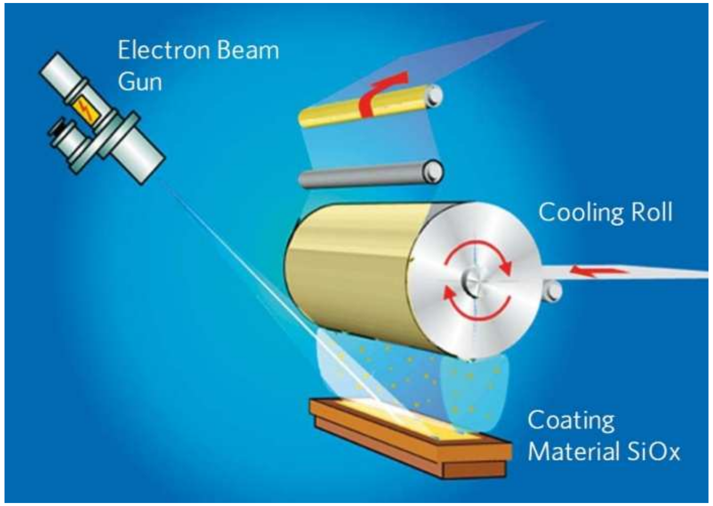
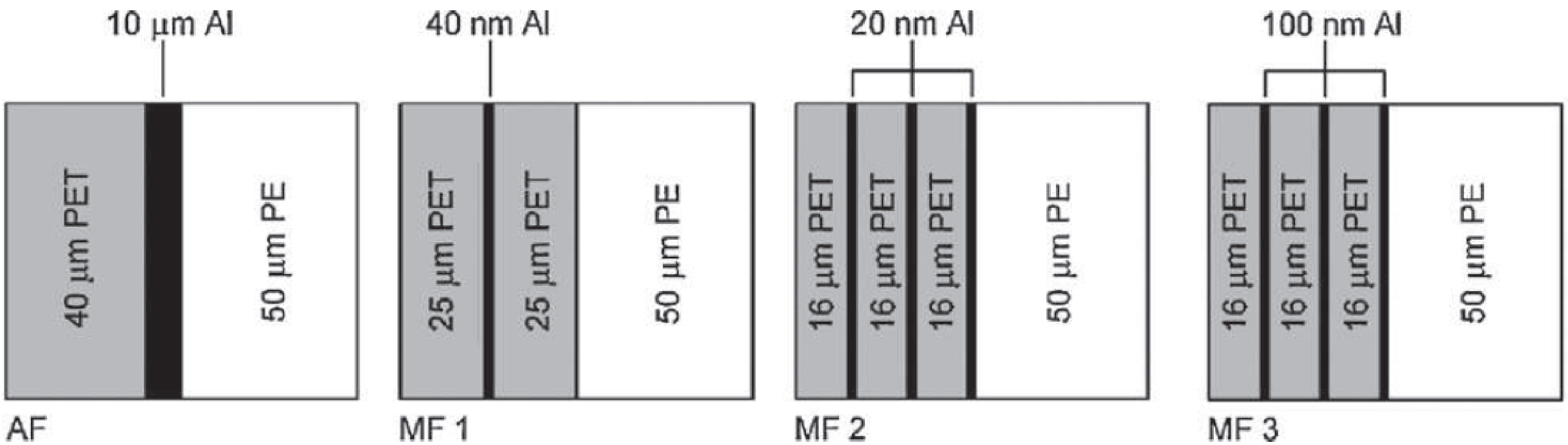
Left: Fraunhofer IVV
Right: H.-C. Langowski, in: O. G. Piringer, A. L. Baner (eds.), Plastic Packaging - Interactions with Food and Pharmaceuticals, 2008

State-of-the-art barrier films for VIPs

- **Physical vapour deposition of inorganic layers** on polymeric substrates
  - Thermal or electron beam evaporation
  - Aluminum or transparent oxides, e.g. SiO_x, AlO_x
- **Lamination** of films carrying barrier layers

⇒ **Barrier improvement of polymeric films**





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Top: Electron beam evaporation (Amcor)
Bottom: Laminates for VIP encapsulation (R. Baetens et al., Energy and Buildings 42 (2010), 147–172)

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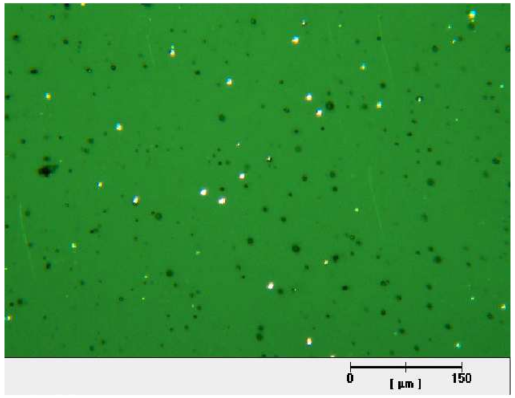
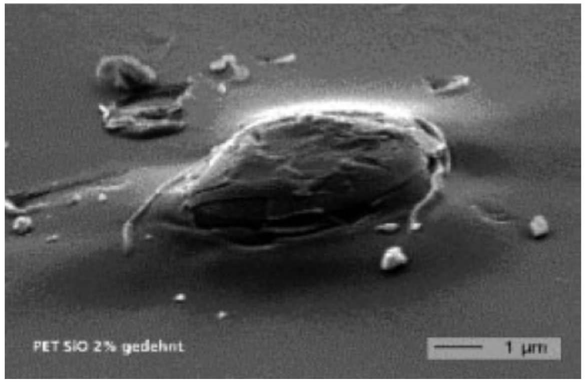
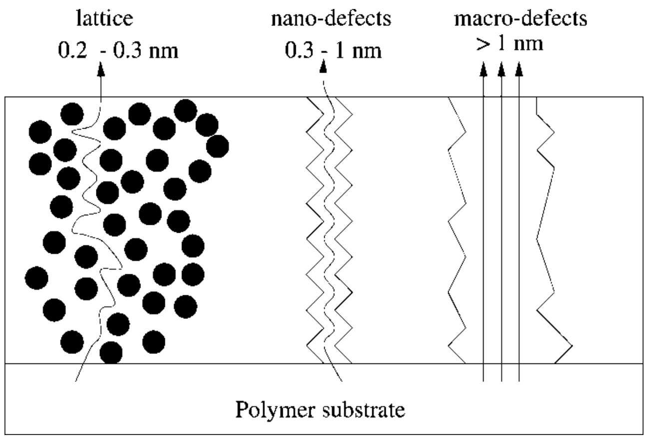
Permeation through inorganic barrier layers

- **Permeation of O_2 and H_2O through defects** within inorganic layers
- **Formation of defects**: Surface roughness, particles, mechanical stress
- **Additional permeation mechanisms for water vapour**
  - Nanodefects or grain boundaries
  - Assumption: Capillary condensation







Left: Defects in Al (IVV); Center: Defects in SiO_x (IVV)
Right: Permeation paths in SiO_x (A.P. Roberts et al., Journal of Membrane Science 208 (2002), 75–88)

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Development of Nanotechnology-based High-performance Opaque & Transparent Insulation Systems for Energy-efficient Buildings

NanoInsulate

- Development of cost effective **opaque and transparent VIPs**
- **Six times more energy efficient** than current solutions
- **Core materials**: Nanoporous polymer foams, aerogel-polymer composites
- **VIP lifetime > 50 years**
  - **O_2 permeability (at 23°C) < 10^{-3} cm^3(STP)·d^{-1}·m^{-2}·bar^{-1}**
  - **WVTR (at 23°C, 85%→0% r.h.) < 10^{-3} g·d^{-1}·m^{-2}**

Funded by the European Union 7th Framework Programme

Time: 2010 – 2014

11 project partners from 7 countries

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SEVENTH FRAMEWORK PROGRAMME

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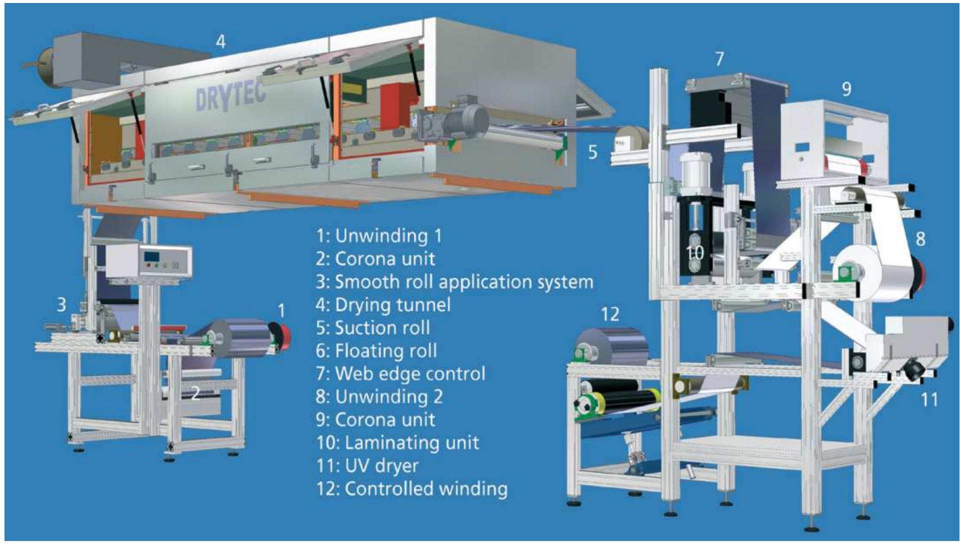
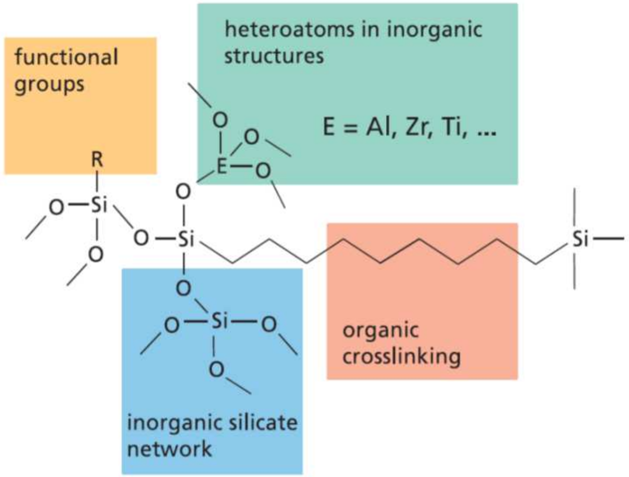
ORMOCER® layers

- **ORMOCER®s**: Inorganic-organic hybrid polymers
  - Mechanical protection of inorganic layers
  - High barrier performance on inorganic layers
  - Mechanical flexibility
  - Smooth surface
- **Application from liquid phase** by reverse gravure process
  - Curing of the lacquer by hot air
- **Combination of ORMOCER® with inorganic layers**





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Left: Lacquering and laminating machine of Fh IVV
Right: Structural elements of ORMOCER®s (Fh ISC)

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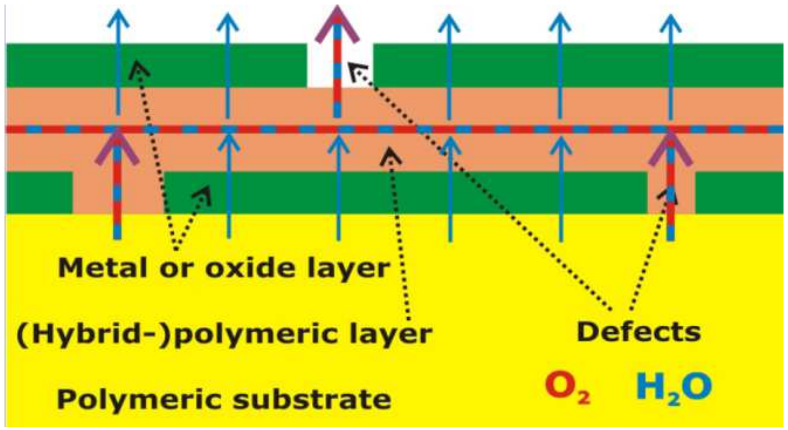
Permeation through alternating barrier layers

Barrier effects of the hybrid polymeric intermediate layer

- **Planarization of the inorganic layer**
  - ⇒ Decoupling of defects ⇒ Tortuous path effect
- **Improved mechanical flexibility**
- **Synergistic effect** between an inorganic layer and a hybrid polymer coated on it



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Left: Structure of alternating barrier layers (O. Miesbauer, IVV)
Right: SEM picture of alternating barrier layers (IVV)

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Novel barrier films for VIPs

PET substrate

- PET Melinex 401 (50 µm thickness)
  - Smooth surface; low elongation in MD direction during ORMOCER® curing
- „PET 1“
  - Low thickness 23 µm; Cost efficient

Inorganic layers

- Al (thermal evaporation): Opaque VIPs
- AlO_x (reactive thermal evaporation): Adhesion promoter
- SiO_x (electron beam evaporation): Transparent VIPs

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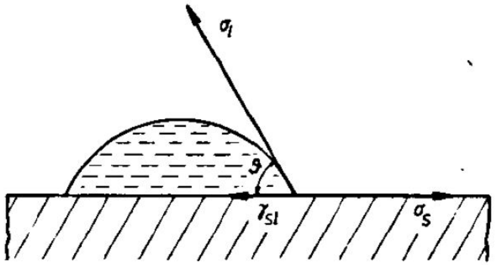
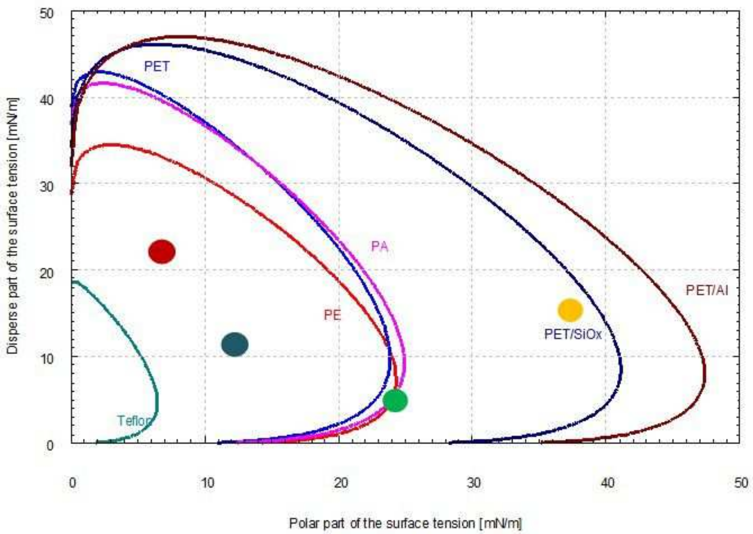
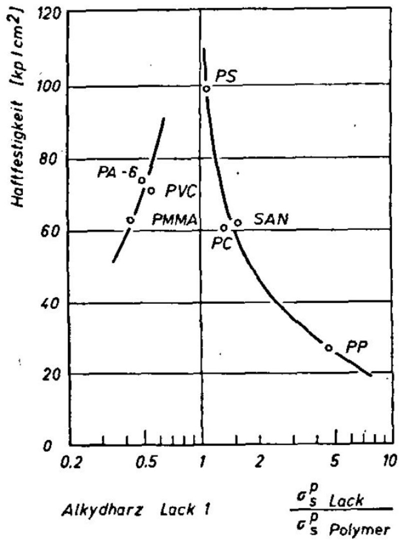
Adhesion between barrier layers

- **Combination of different materials ⇒ Adhesion is important**
- **Necessary for adhesion** of a lacquer on a substrate surface
  - Wetting of the lacquer
  - Adaption of surface energies of lacquer and substrate surface
- **Measurement of surface energies** (polar and disperse part): Contact angle method







Left (Contact angle) and right (Ratio of polar parts of surface energy vs. adhesion strength): H. Potente, R. Krüger, Farbe und Lack 84 (1978) 2, 72–75
Center (Wetting envelopes): I. Wallaschek, O. Miesbauer, IVV, 2011

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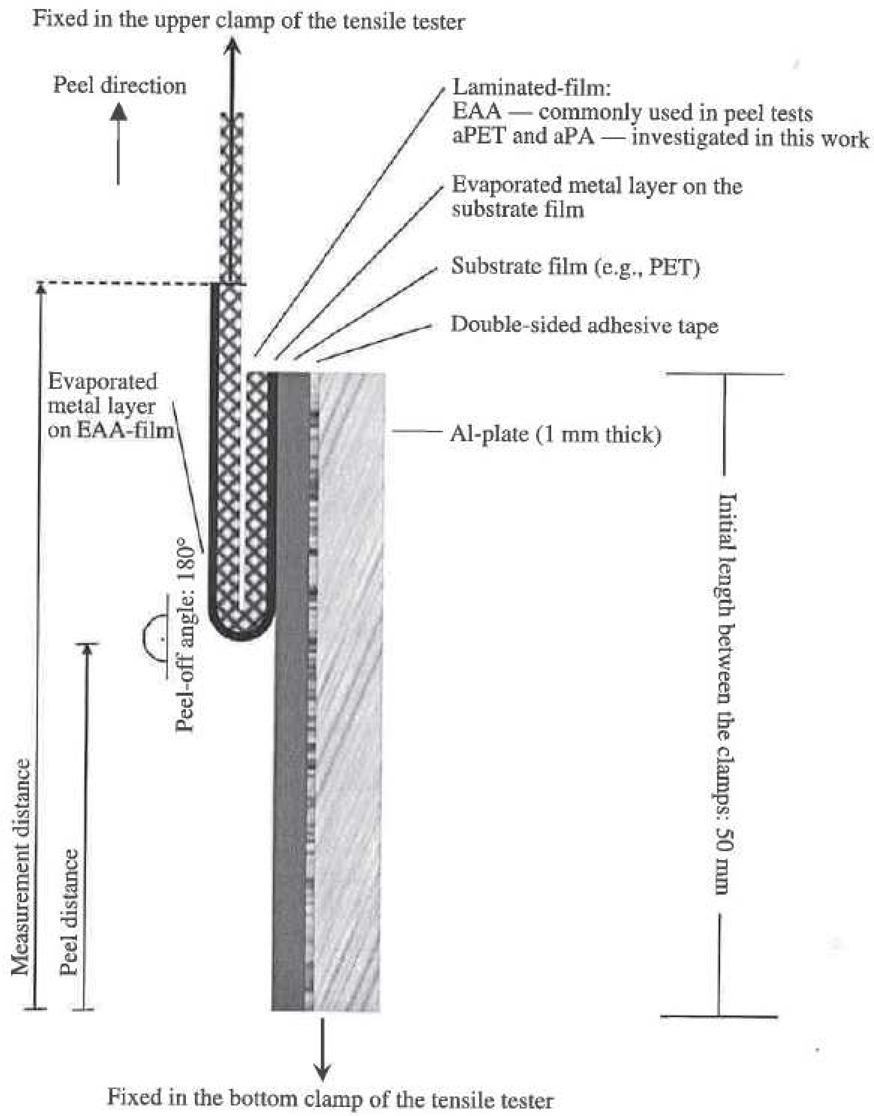
Adhesion between barrier layers

- **Measurement of adhesion strength:** EAA peel test
- EAA: Ethylene acrylic acid copolymer



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EAA peel test: M. Jesdinszki et al., J. Adhesion Sci. Technol. 26 (2012), 2339-2356

Results for opaque barrier films: Adhesion

- **Good adhesion (> 2 N/15mm) of ORMOCER® on Al or AlOx only for shown structures**
- No adhesion of ORMOCER® on PET 1 / Al in contrast to PET Melinex / Al
- **AlOx works as adhesion promoter** for ORMOCER® on PET 1 / Al
- **Choice of PET 1 / Al / AlOx / ORMOCER®** for further development since PET 1 is preferred substrate

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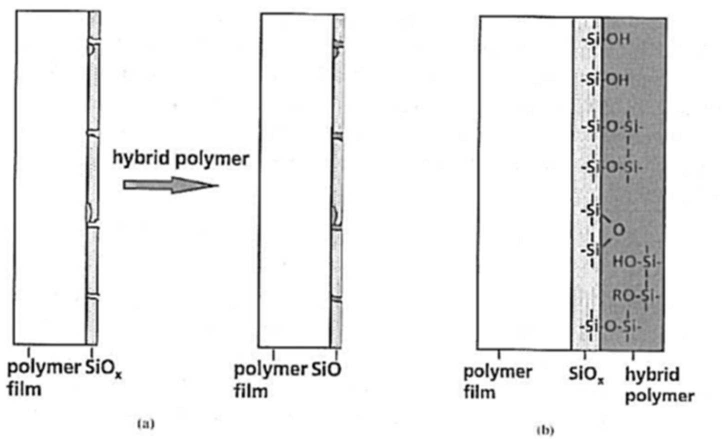
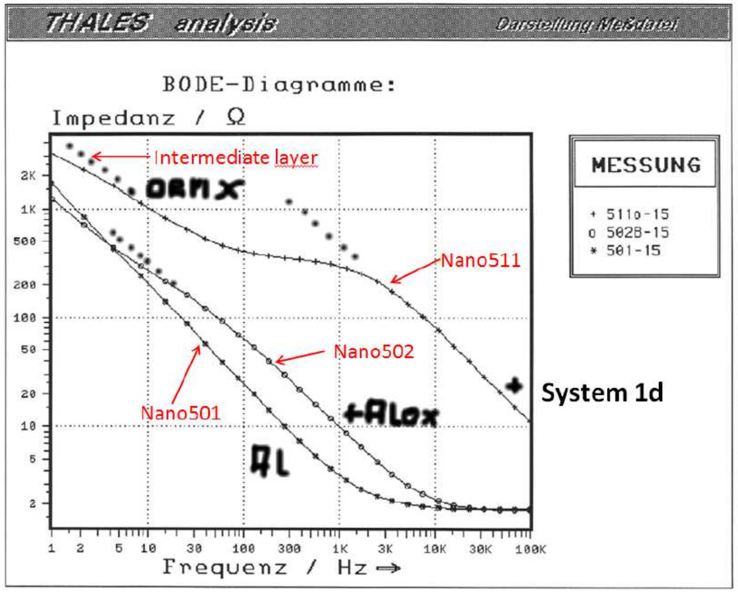
Results for opaque barrier films: Adhesion

- **Surface of Al: Passivation with AlOx layer** due to reaction with O_2 from air
- No adhesion of ORMOCER® on PET 1 / Al / AlOx (passivation)
- Good adhesion of ORMOCER® on PET 1 / Al / AlOx (deposited)
- Explanation by **electrochemical impedance spectroscopy**
  - **Possible penetration of ORMOCER®** into deposited porous AlOx layer

  ⇒ **Good adhesion**

  - Confirmation of **synergistic effect**





Left: Synergistic effect (S. Amberg-Schwab et al., J SOL-GEL SCI TECHN 13 (1998), 141–146); Right: Impedance spectroscopy of PET 1 / Al / AlOx (deposited) / ORMOCER® (J. Hollaender, IVV, 2012)

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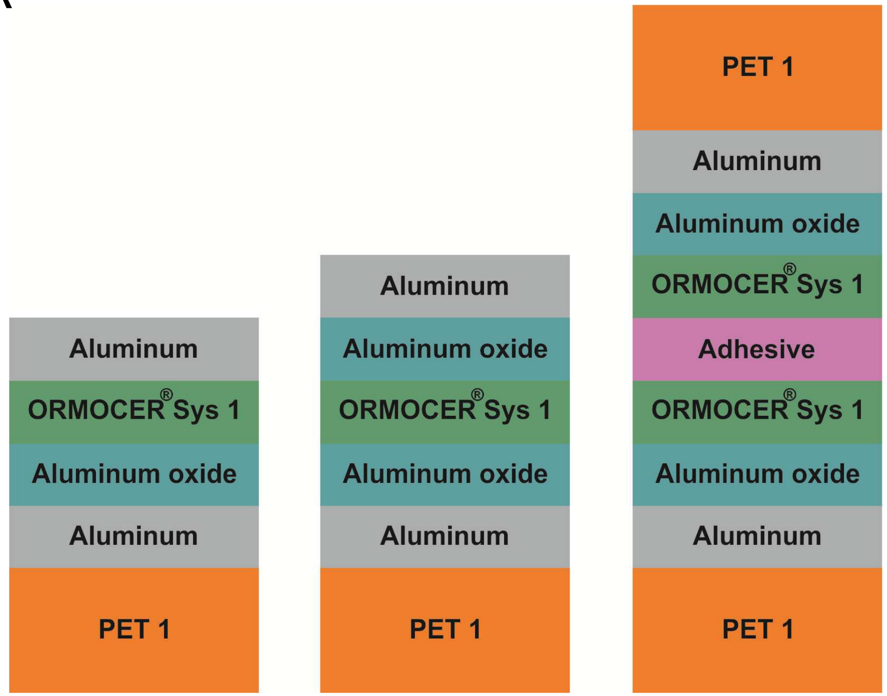
Results for opaque barrier films: Adhesion

- Moderate adhesion of Al directly on ORMOCER®
- **AlOx works as adhesion promoter** for Al on ORMOCER®
- Alternative: **Face-to-face laminate** of PET 1 / Al / AlOx / ORMOCER®
  - Good adhesion obtained in face-to-face laminate of PET Melinex / Al / ORMOCER®



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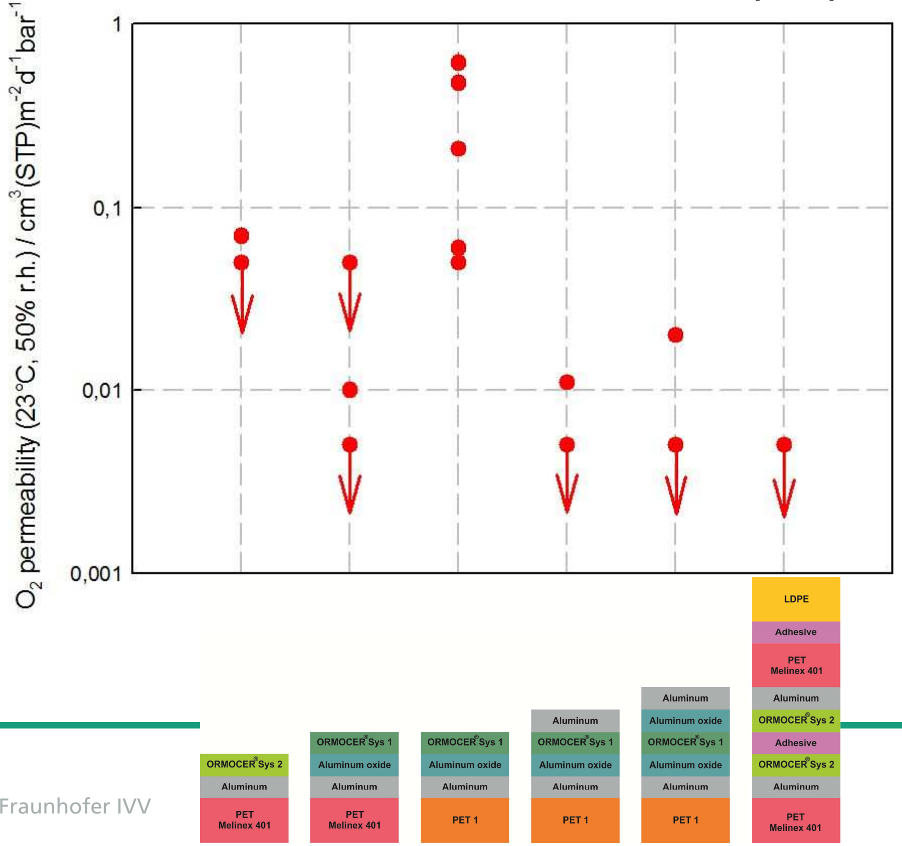
Results for opaque barrier films: Barrier performance

O_2 permeability (at 23°C, 50% r.h.) …

- … for inorganic layer on PET 1 higher than on PET Melinex
  - Possible explanation: Higher substrate roughness of PET 1
- … with AlOx adhesion promoter lower than without AlOx
  - Possible explanation: Synergistic effect of ORMOCER® on top of AlOx
- … for final structures < $5 \cdot 10^{-3}$ $cm^3(STP) \cdot d^{-1} \cdot m^{-2} \cdot bar^{-1}$



WVTR (at 23°C, 85%→0% r.h.) …

- … for PET 1 / Al / AlOx / ORMOCER® / Al // LDPE: ≤ 0.001 $g \cdot d^{-1} \cdot m^{-2}$

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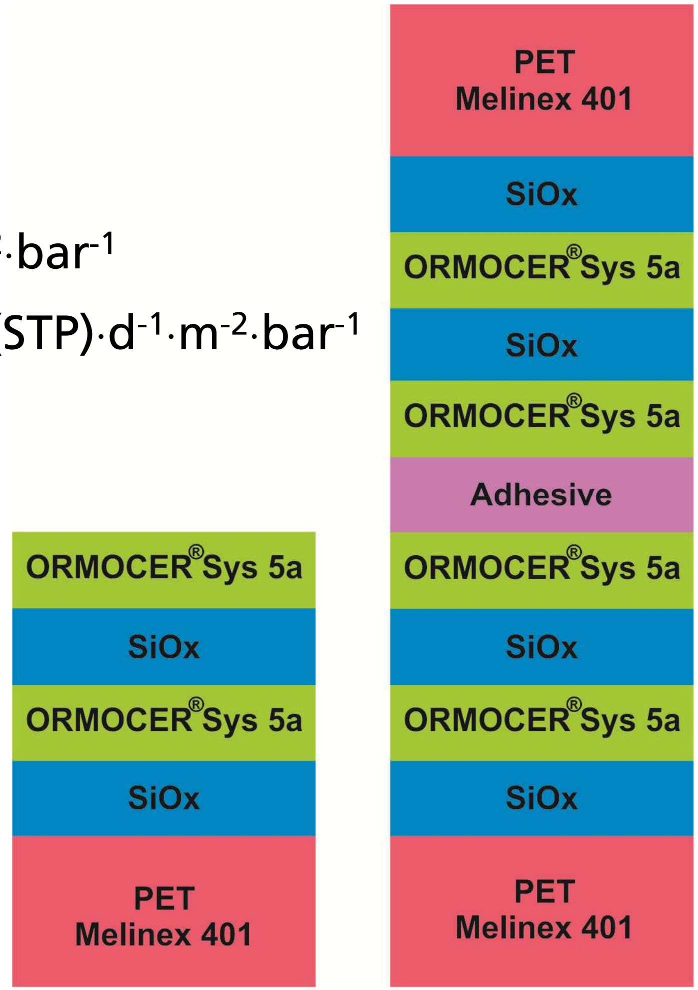
Results for transparent barrier films: Barrier performance

O_2 permeability (at 23°C, 0% r.h.) …

- … for single film < $5 \cdot 10^{-3}$ $cm^3(STP) \cdot d^{-1} \cdot m^{-2} \cdot bar^{-1}$
- … for face-to-face laminate < $1 \cdot 10^{-4}$ $cm^3(STP) \cdot d^{-1} \cdot m^{-2} \cdot bar^{-1}$



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Summary and outlook

- **Novel barrier films for VIP encapsulation:** combination of inorganic and ORMOCER® layers
- **Measured O_2 permeabilities (23°C) < $5 \cdot 10^{-3}$ $cm^3(STP) \cdot d^{-1} \cdot m^{-2} \cdot bar^{-1}$**
  - Required O_2 barrier performance almost reached
  - Measurement of O_2 and water vapour permeabilities for finally developed films in progress
- **Choice of a suited sealing film**
- **Final evaluation of novel barrier laminates** by measurement of the increase of gas pressure within VIPs encapsulated with these films
- **AlOx layers as adhesion promoter** necessary within opaque barrier films
  - Further work to be able to omit the AlOx layers in industrial production process

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Thank you for your attention!

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