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SORPTION-ENHANCED REFORMING OF NATURAL GAS FOR PRE-COMBUSTION DECARBONISATION

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Sorption-Enhanced Reforming of natural gas for PRE-Combustion Decarbonisation

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Energy research Centre of The Netherlands

Vancouver, 10 September 2004

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Greenhouse gas mitigation is essential to the Energy Research Centre of the Netherlands

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ECN Priority Areas

Biomass

Solar Energy

Wind Energy

Renewable Energy in the Built Environment

Fuel Cell Technologies

Policy Studies

Clean Fossil Fuels

Energy Efficiency in Industry

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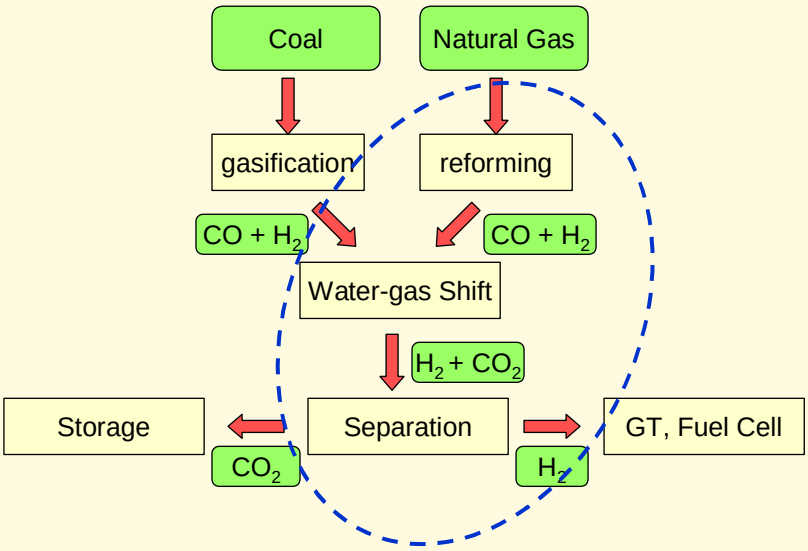
Pre-combustion decarbonisation

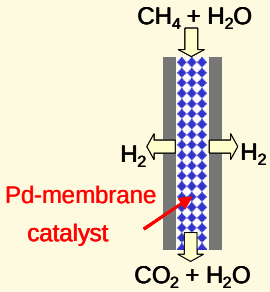


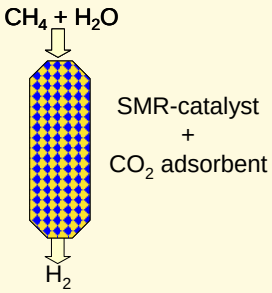
Separation-enhanced reactors

Steam reforming: $CH_4 + H_2O \rightleftharpoons 3\,H_2 + CO$ ($\Delta H = 206$ kJ/mol)

Water-gas shift: $CO + H_2O \rightleftharpoons H_2 + CO_2$ ($\Delta H = -41$ kJ/mol)

$CH_4 + 2\,H_2O \rightleftharpoons 4\,H_2 + CO_2$

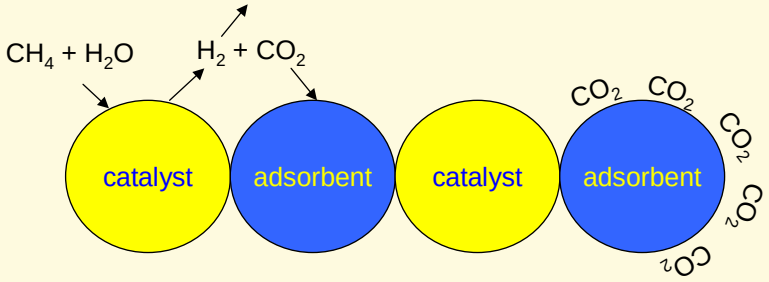




- Membrane reactors
- Sorption-enhanced reactors

Sorption Enhanced Reaction Process (SERP)

$$CH_4 + 2\,H_2O \rightleftharpoons 4\,H_2 + CO_2$$



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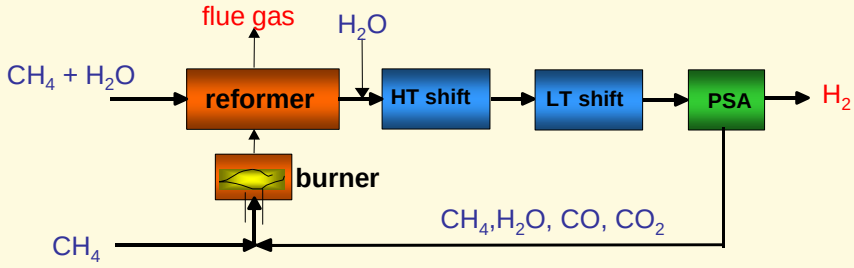
Conventional steam reforming of natural gas



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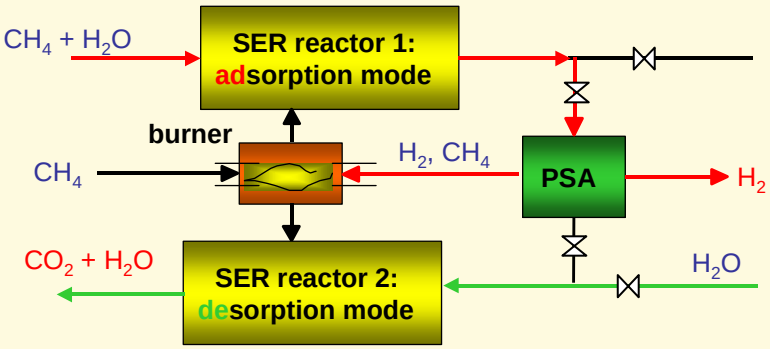
Sorption-enhanced reforming



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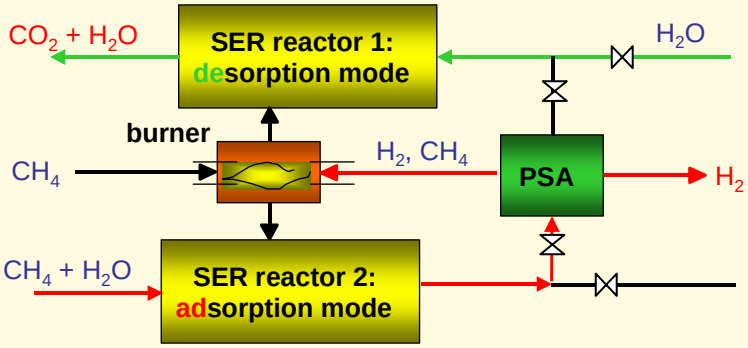
Sorption-enhanced reforming



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Results SERP
Base case system analysis

- Three main challenges:
  - High conversion of CH_4 (>95%) needed
  - Reduction of CO_2 stripping steam
  - 85% CO_2 capture

} High efficiency

- Approach
  - Improved/new catalyst and adsorbent
  - Process optimization (cycle times)
  - Innovative system configurations

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Experiments and Materials

- Experimental conditions
  - 100 ml/min flows
  - 1 – 5 grams adsorbent
  - pressure 1 to 5 bar(a)

- Materials
  - Commercially available (pre)reform catalysts or WGS catalysts
  - Hydrotalcite adsorbents

Assessment

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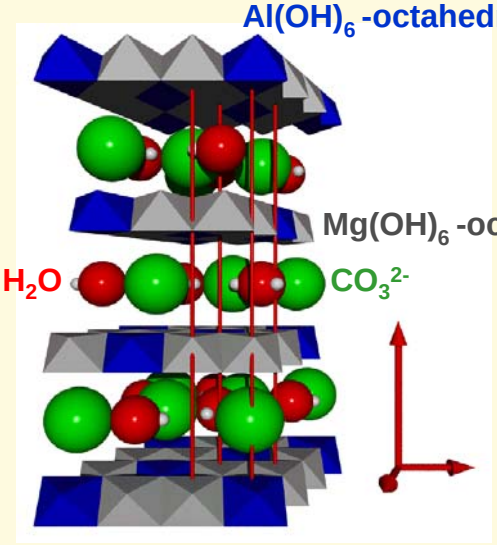
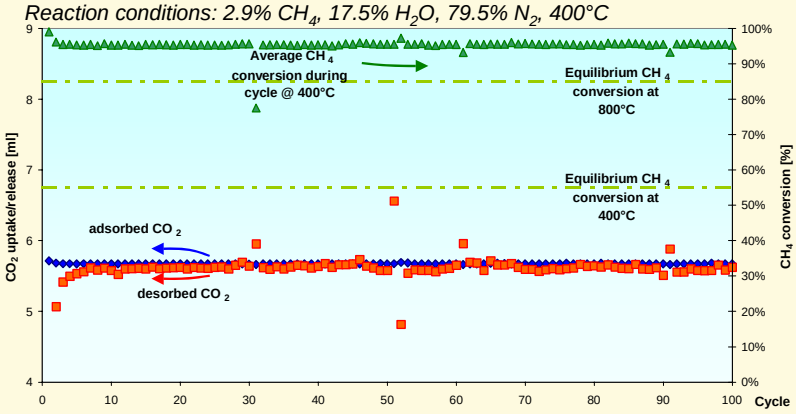
Adsorbent: hydrotalcite



general formula:
$Mg_{1-x}Al_x(OH)_2(CO_3)_{x/2}.nH_2O$
Promoted with K_2CO_3

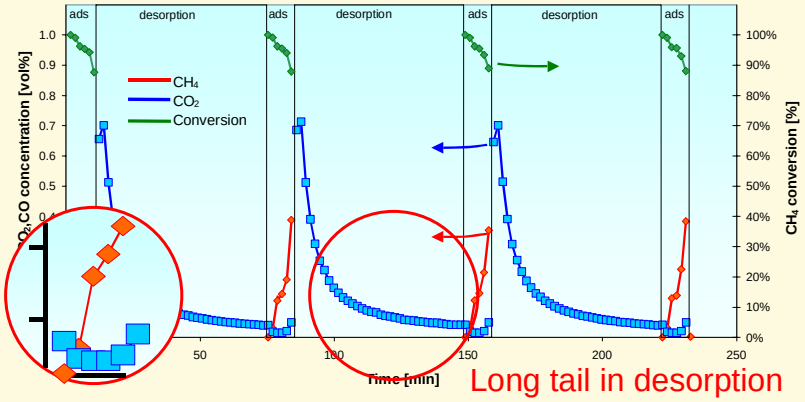
High average CH_4 conversion over 100 cycles

Reaction conditions: 2.9% CH_4, 17.5% H_2O, 79.5% N_2, 400°C



Typical experiment steam reforming of methane

Reaction conditions: 2.9% CH_4, 17.5% H_2O, 79.5% N_2, 400°C



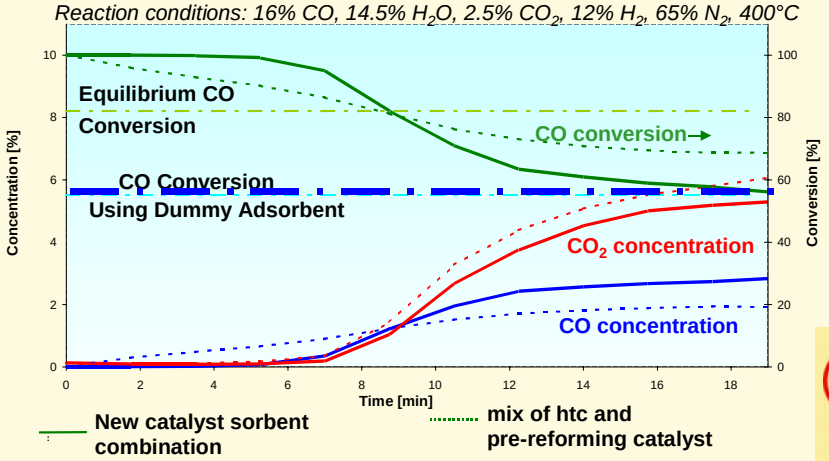
Long tail in desorption

Breakthrough of methane before CO_2

Efficiency Improvement in Water-Gas Shift Reaction

$$CO + H_2O \rightleftharpoons H_2 + CO_2$$

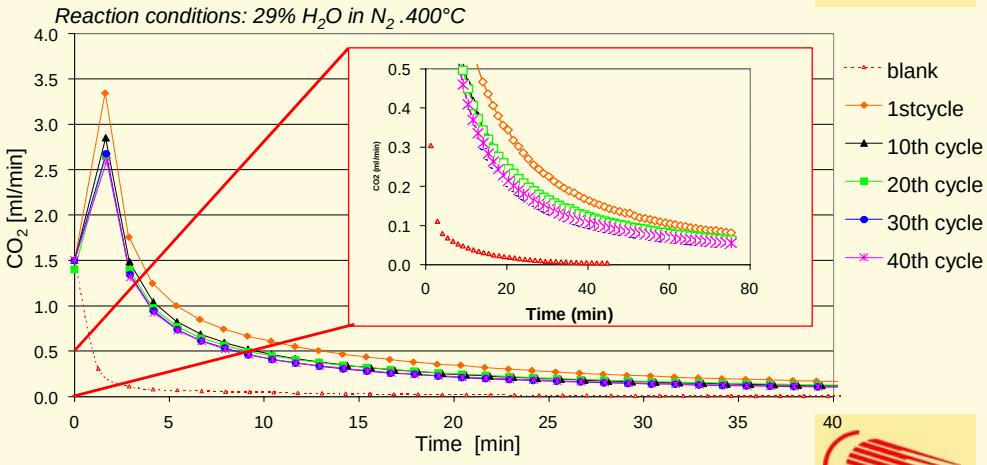
Reaction conditions: 16% CO, 14.5% H_2O, 2.5% CO_2, 12% H_2, 65% N_2, 400°C



New catalyst sorbent combination

mix of htc and pre-reforming catalyst

Regeneration of CO_2-loaded adsorbent



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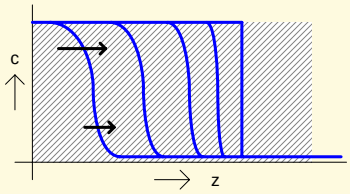
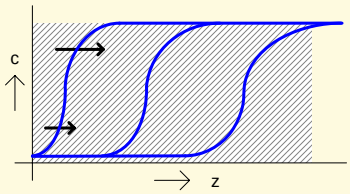
Modeling of the adsorption: Langmuir isotherm



Adsorption



Desorption

• For complete regeneration > 50 moles steam per mole CO_2 adsorbed is needed

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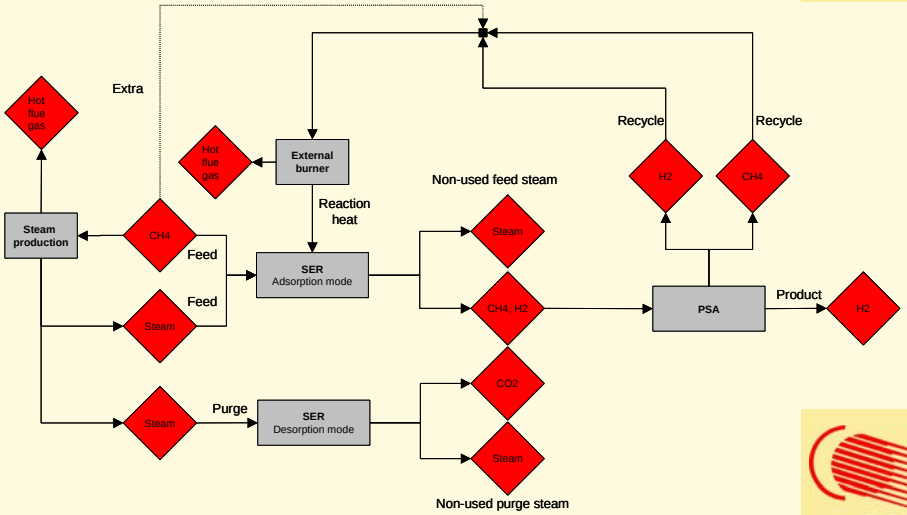
Systems modeling



Systeemberekeningen

steam/CO_2 ratio (-)	PSA efficiency (%)	feed steam efficiency (%)	purge steam efficiency (%)	H_2 production efficiency (%)	CO_2 capture efficiency (%)
50	85	0	0	27	25
50	85	*50*	*50*	42	39
50	*75*	50	50	39	41
25	75	50	50	53	56
10	75	50	50	70	74
5	75	50	50	78	82

= input

= result

= changed parameter

S/C 6

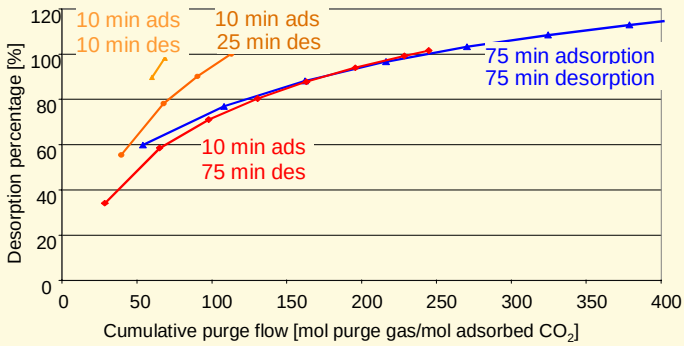
T = 491 °C

P_{ads} = 1.7 bar

kat/ads =2:3 (w/w)

- Low steam demand very important
  - lower desorption pressure
  - shallow desorption

Effect of shallow desorption



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- Low steam demand very important
  - lower desorption pressure
  - shallow desorption
  - novel adsorbents

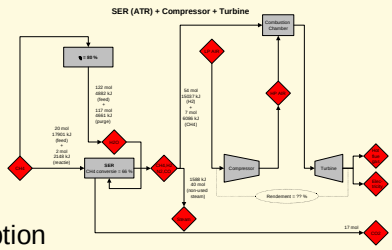
Development of novel adosrbents

- Modification of adsorbent:
  - Promising new hydrotalcites and promoters found
    - Alkali metal carbonates
  - Further development on going in co-operation with Utrecht University

Conclusions and future work SERP

- Conclusions
  - Methane conversion > 98% feasible at T=400 C
  - < 100 ppm levels of CO, CO_2 in H_2 gas
  - CO_2 removal efficiency of 85 % feasible

- R&D issues
  - Steam/CO_2 ratio < 5 (shallow ad- desorption mode)
  - Integration in GT-cycle
  - Reactor design



Acknowledgements

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  - Netherlands Ministry of Economic Affairs