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Operation of a chemical looping pilot plant up to 150 kW fuel power – results for a nickel-based oxygen carrier and discussion of further scale-up scenarios

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and Hermann Hofbauer

- Critical issues in chemical looping combustion
- Dual circulating fluidized bed concept
- 120 kW pilot plant for gaseous fuels
- Experimental results with NiO-based particles
 - Chemical looping combustion
 - Chemical looping reforming
- Conclusion and outlook on scale-up

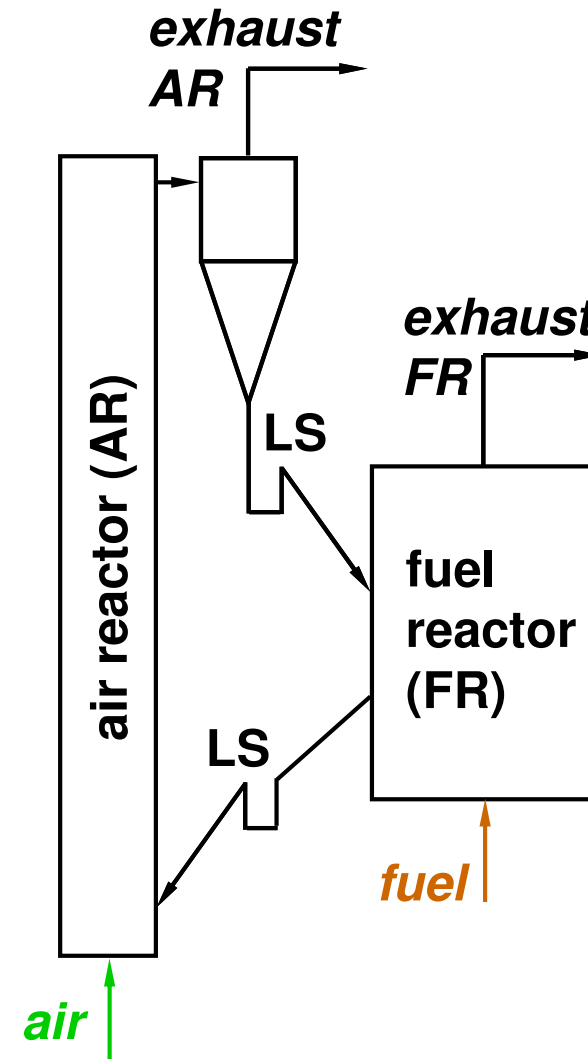
- Oxygen carrier particles
 - thermodynamic suitability
 - high reactivity
 - sufficient transport capacity
 - high mechanical stability
 - cyclic stability of reactivity and transport capacity
- Reactor system
 - excellent gas-solids contact in both reactors
 - sufficient solids circulation rate

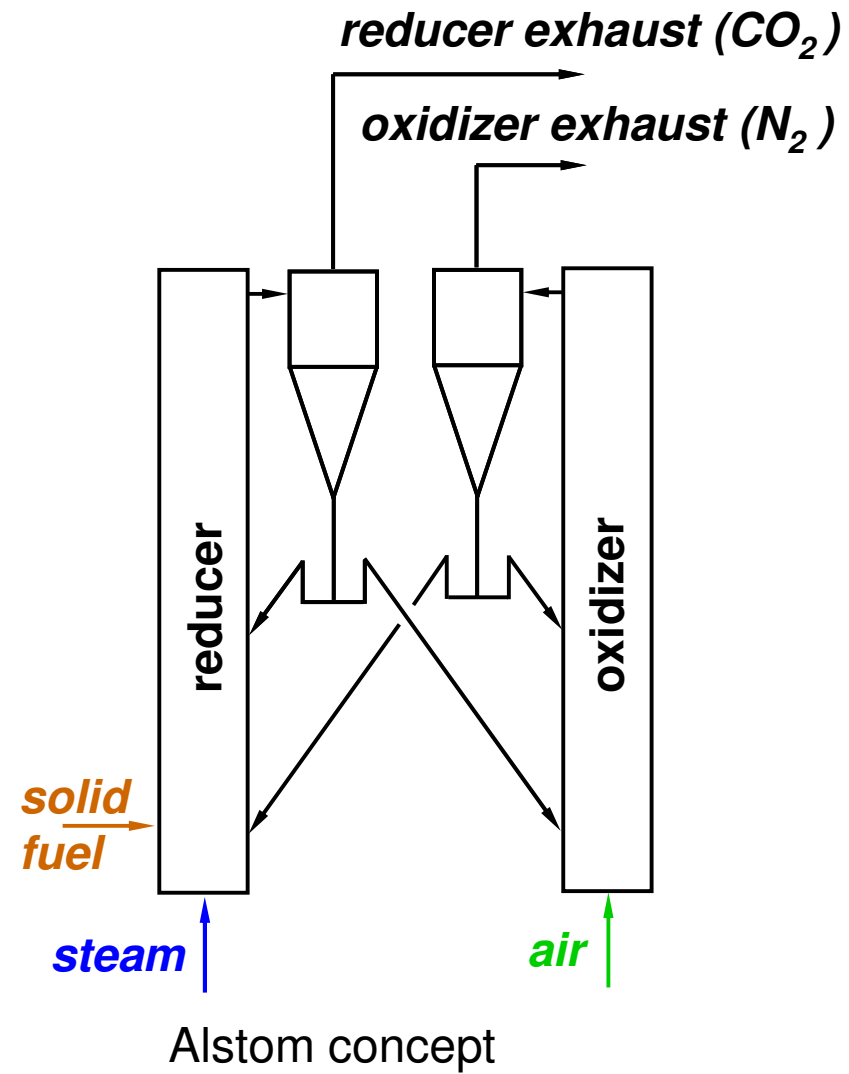
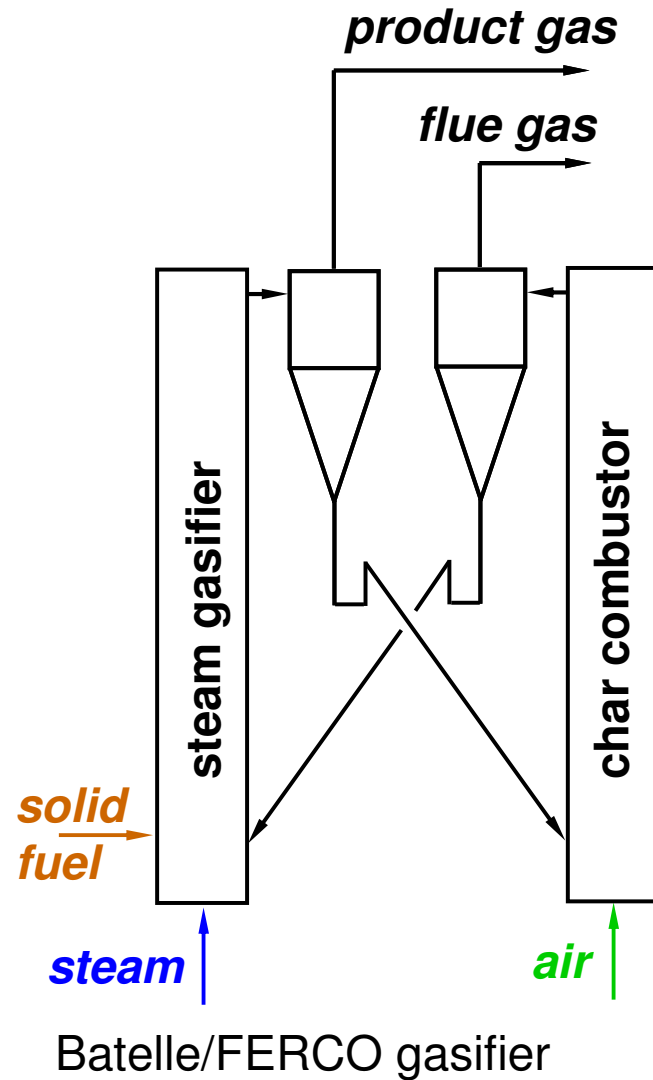
Dual fluidized bed technologies apart from FCC

Technology	Purpose of solids	Importance of gas–solid contact
(Biomass) gasification	Heat transport, catalyst	Partially for tar reforming in the gas generator
Sorption enhanced reforming	CO ₂ and heat transport, catalyst	High in the reformer/carbonator, low in the re-calciner (heat-driven)
Carbonate looping for CO ₂ capture	CO ₂ (and heat) transport	High in the absorber/carbonator, low in the re-calciner (heat-driven)
Chemical looping	Oxygen and heat transport	High in both reactors, no gas phase conversion without solids

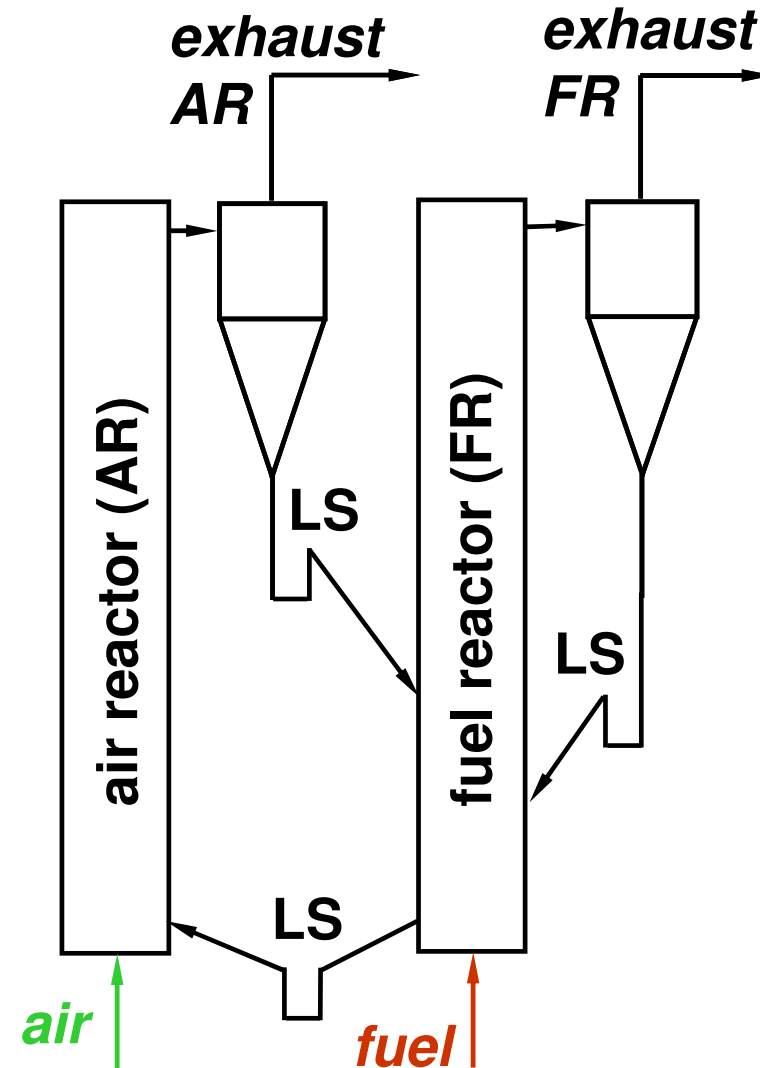
→ There is an ultimate requirement for good gas-solid contact in CLC systems

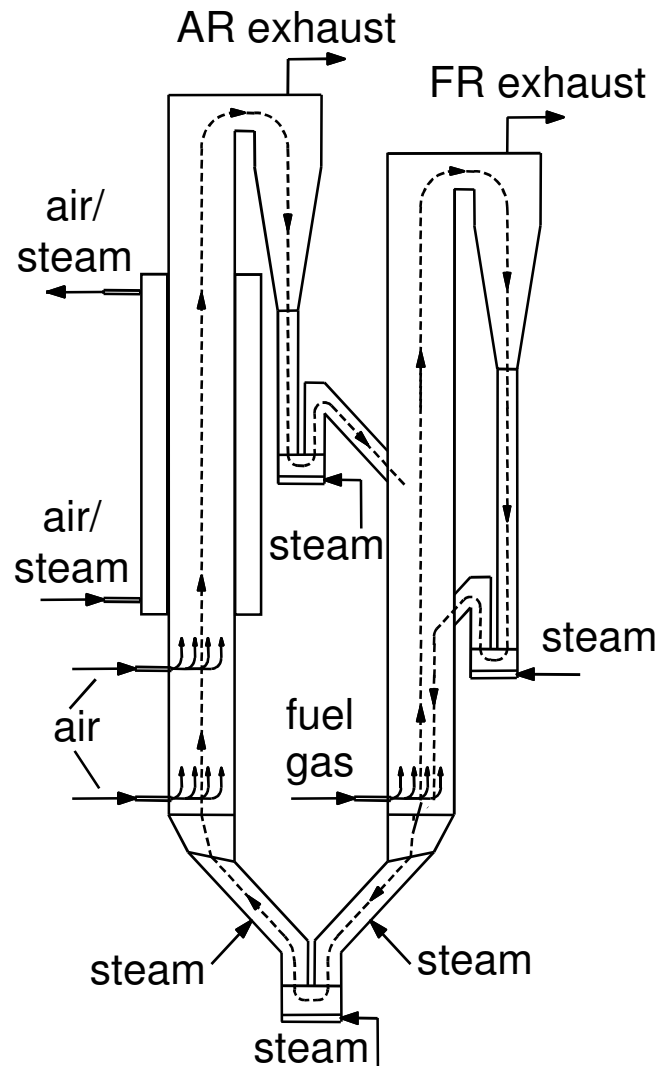
- Fast fluidized air reactor
- Bubbling bed fuel reactor
- Simple and robust
- Potential gas slip in FR (through bubble phase, no reactions in freeboard)
- Particle size mismatch
 - BFB typically $> 500 \mu\text{m}$
 - CFB typically $< 200 \mu\text{m}$
- High bed-cross sections in bubbling bed FR at industrial scale



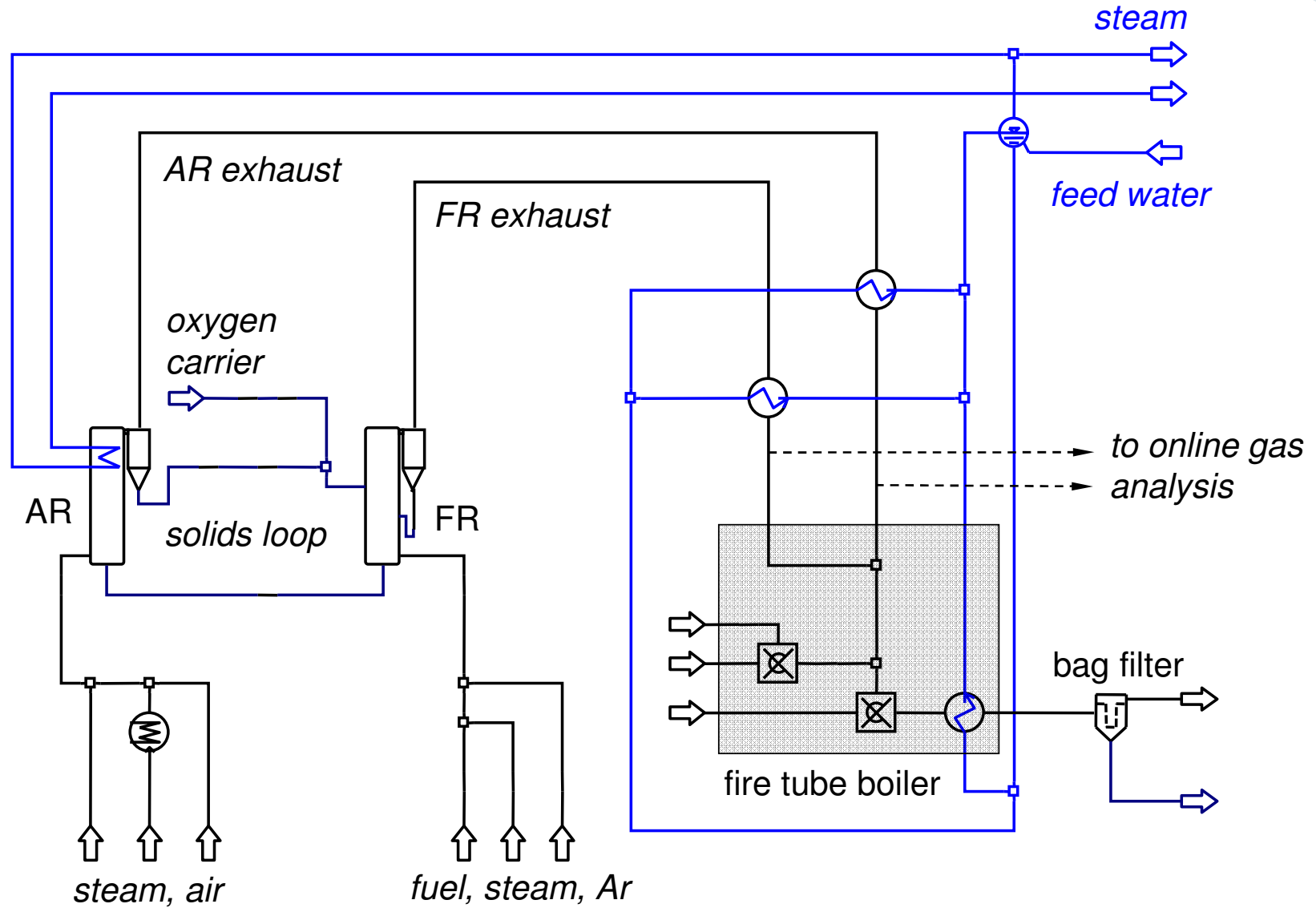


- Global solids circulation is controlled by air reactor fluidization only (eg. air staging)
- Fuel reactor can be optimized towards fuel conversion
- Inherent stabilization of global solids hold up due to the direct hydraulic link between the reactors
- Low reactor volume compared to bubbling fluidized beds (i.e. low specific solids inventory)
- High potential for scale-up

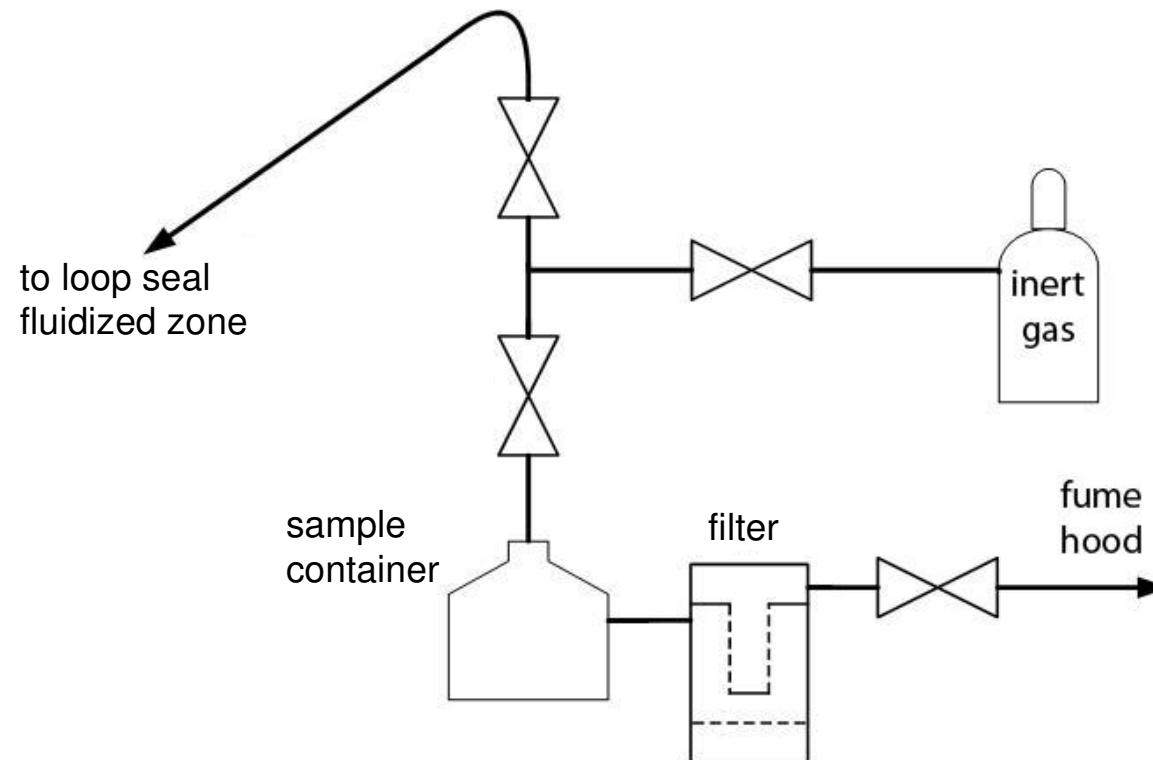


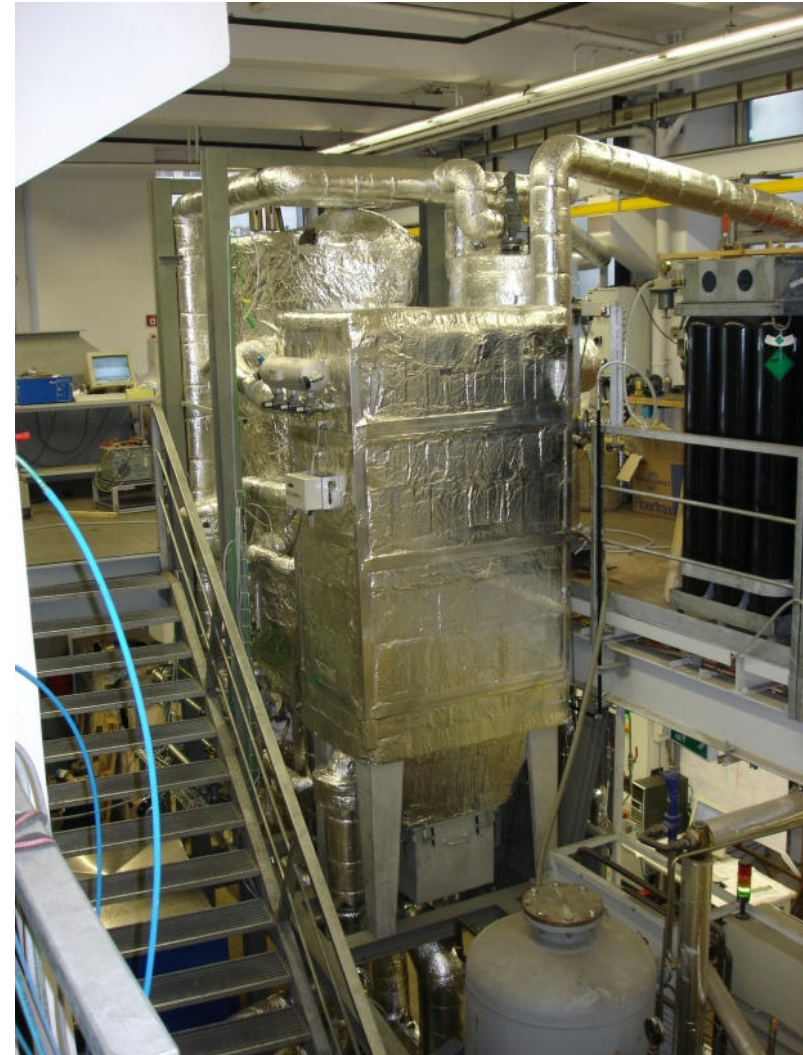
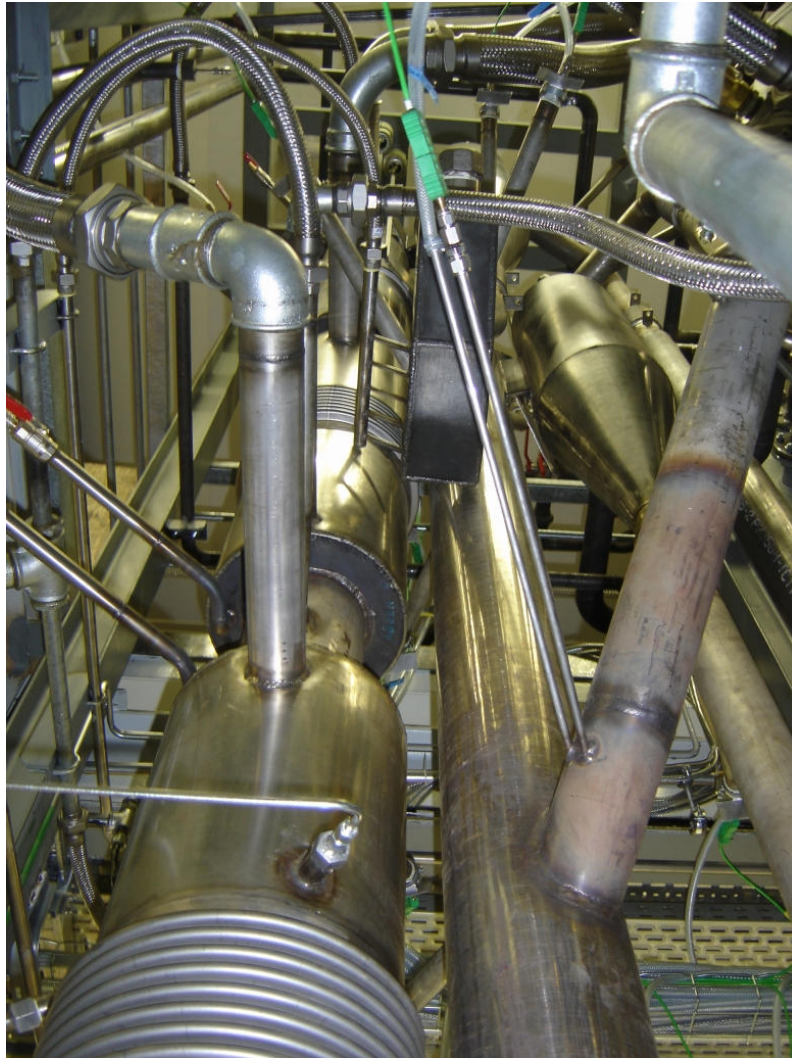


- bottom end = loop seal
- support fluidization in legs to lower loop seal (steam)
- air reactor: 4.1 m high, 0.15 m ID
- fuel reactor: 3.0 m high, 0.16 m ID
- design fuel: natural gas (98.6 % CH₄)
- alternative fuels: CO+H₂, C₃H₈
- design particle size: 120 μm
- Cooling system: double jackets attached to air reactor
- Exhaust gas cleanup: supported post combustion, fire tube boiler, bag filter



- Determination of average degree of oxidation in loop seals connecting air reactor and fuel reactor
- Determination of global solids circulation rate by comparison to gas phase mass balance

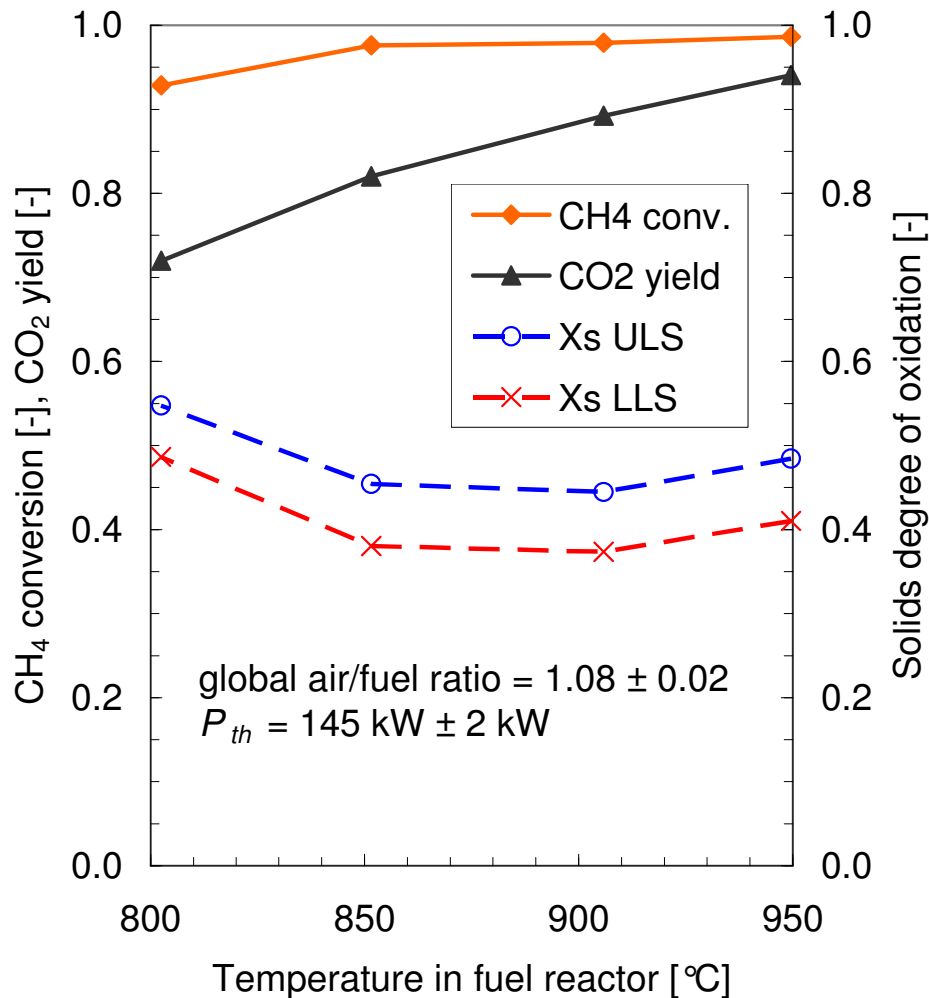




- Developed by Chalmers University of Technology within FP6 project CLC GAS POWER
- Manufactured by spray drying by Flemish Institute for Technology Research (FP6 - CLC GAS POWER)
- Total inventory 65 kg, “active inventory“ 30-35 kg

Parameter	Unit	Value
Oxygen carrier		Ni/NiO
Active NiO content	%	40
Oxygen transport capacity R_0	kg/kg	0.087
Mean particle size	μm	135
Apparent density	kg/m^3	3416

Effect of operating temperature



- NiO-based oxygen carrier
- $P_{th} = 145$ kW for natural gas
- global air ratio = 1.1

Methane conversion:

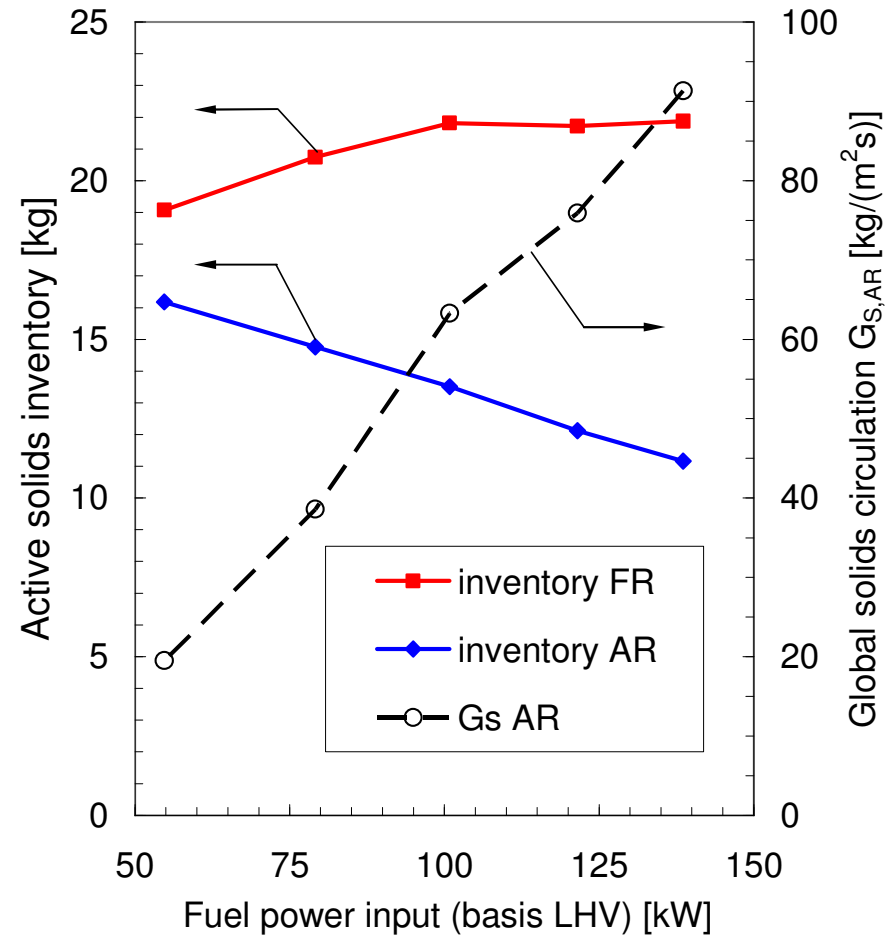
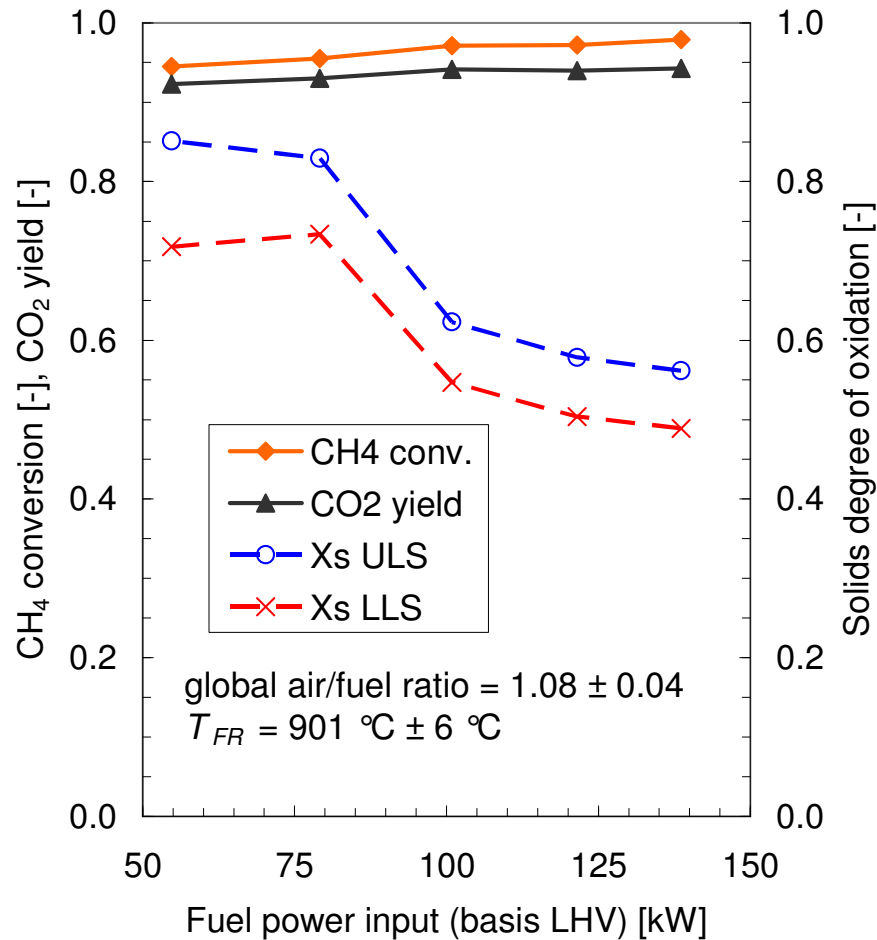
$$X_{CH_4} = 1 - \frac{y_{CH_4}}{y_{CH_4} + y_{CO} + y_{CO_2}}$$

CO₂-yield:

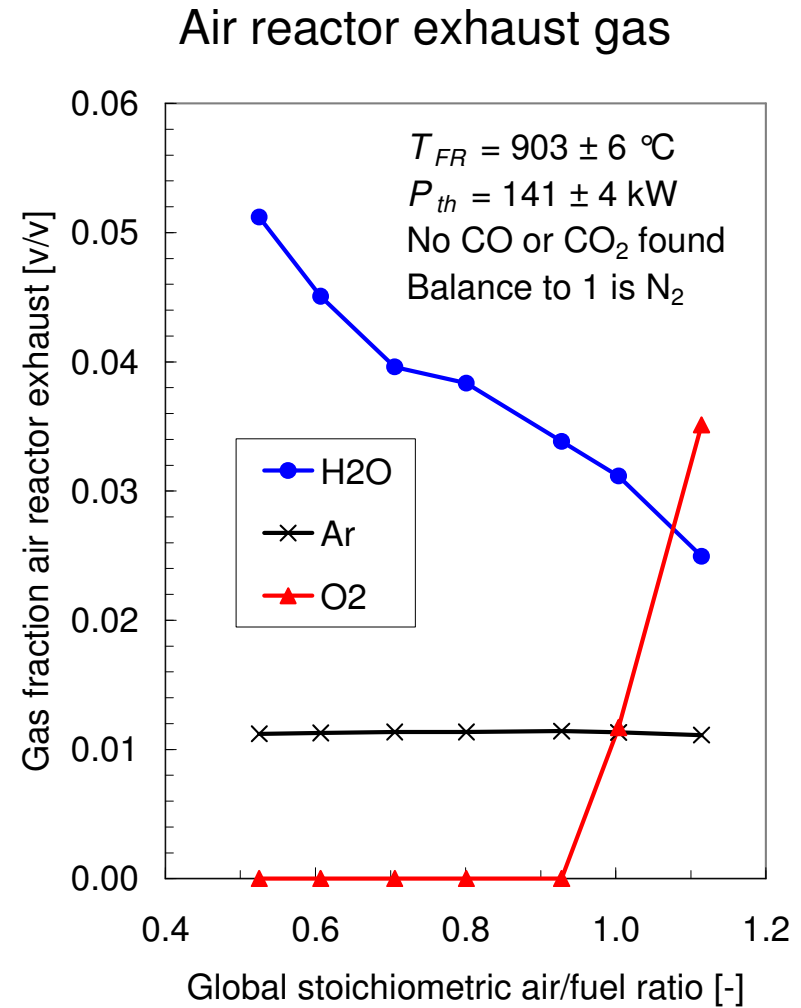
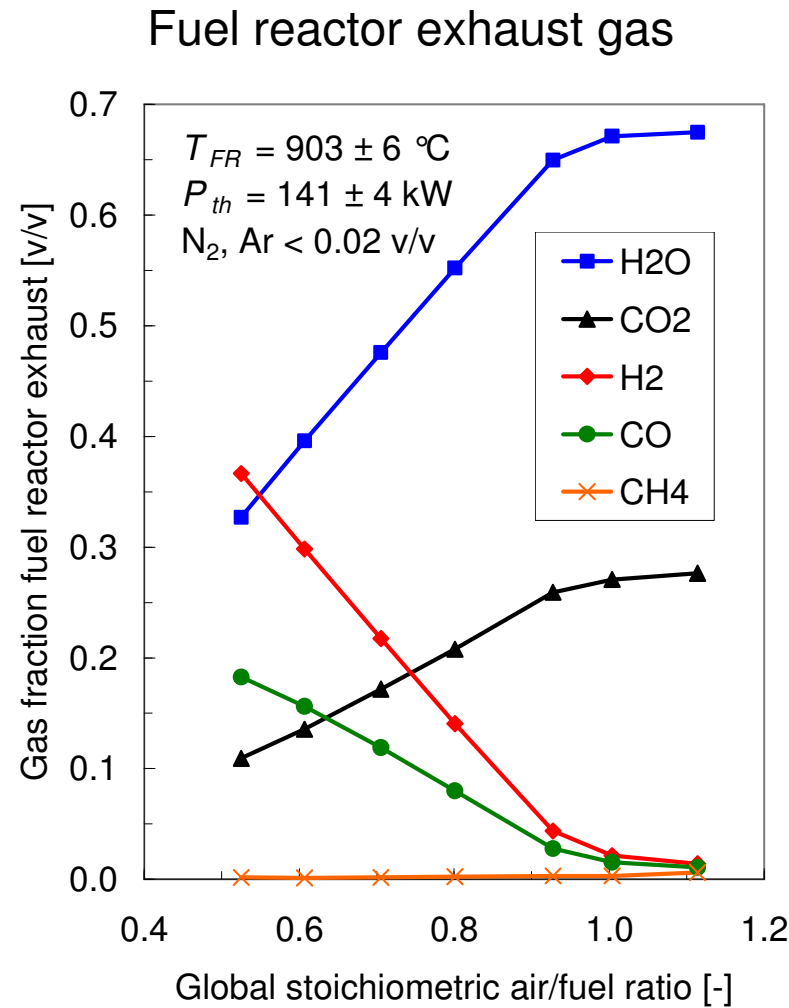
$$\gamma_{CO_2} = \frac{y_{CO_2}}{y_{CH_4} + y_{CO} + y_{CO_2}}$$

y_i ... mole fraction in fuel
reactor exhaust gas

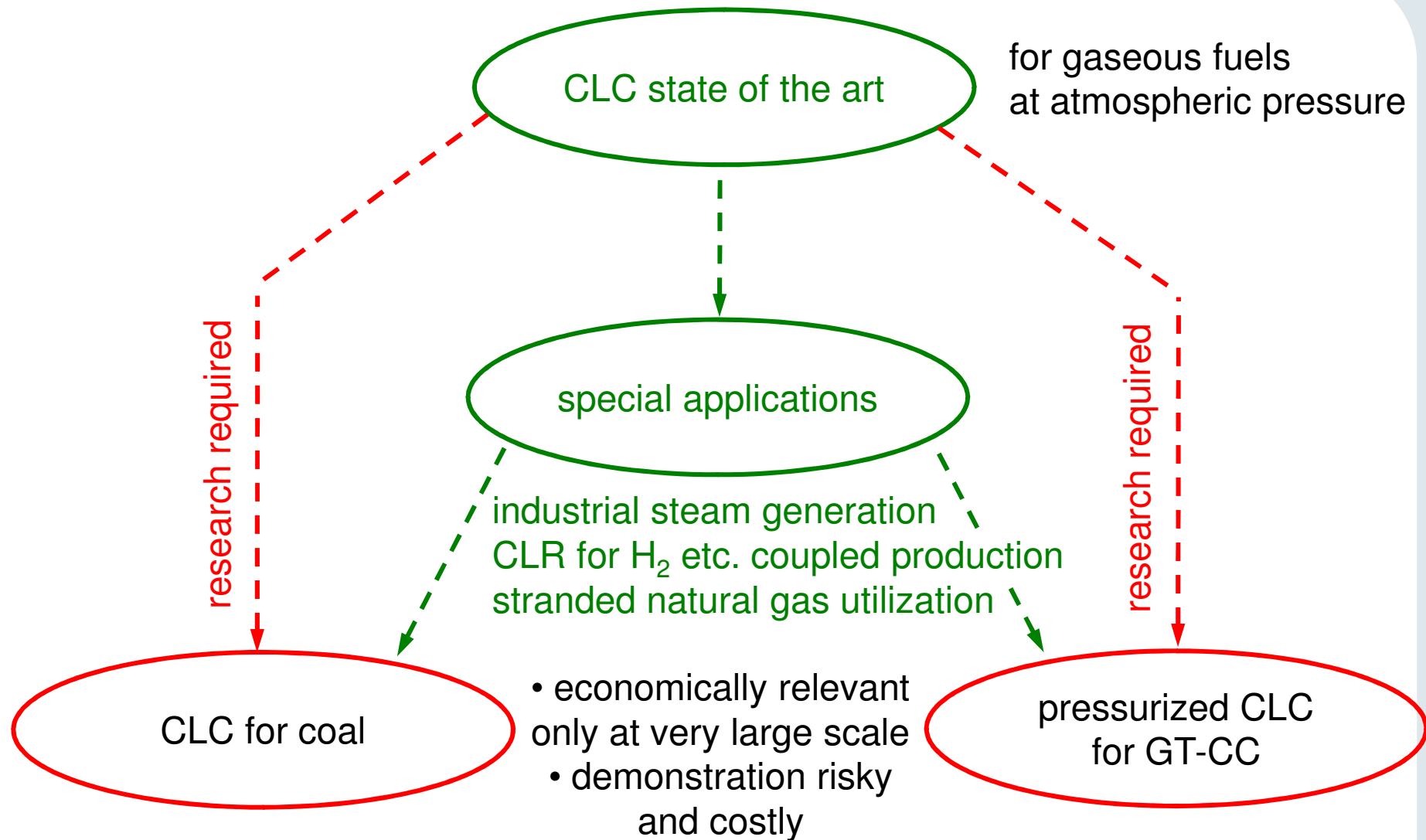
Effect of fuel power input



Effect of global stoichiometric air/fuel ratio



- DCFB reactor system:
 - scale-up ready candidate for large scale chemical looping application
- Pilot plant / NiO-based oxygen carrier:
 - CLC:
 - CH₄ conversion up to 99%
 - CO₂ yield up to 96% based on total carbon in fuel
 - good fuel conversion in spite of the limited riser heights
 - CLR:
 - Conversion to equilibrium in fuel reactor
 - No carbon deposition on particles at air/fuel ratios > 0.4
 - Quantitative O₂ absorption in air reactor
- Further work:
 - demonstrate CLC operation with oxygen carrier materials showing an improved performance/cost ratio
 - detailed reactor design issues
 - scale up to pilot/demonstration size (next step: 10 MW)





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