CATALYSIS SOCIETY OF TURKEY/ METU UZEM NOVEMBER 20 2020

Chemical Engineering and Catalysis:

from molecular to process scale

G.B. Marin

Laboratory for Chemical Technology

GHENT UNIVERSITY

LCT DRIVING CHEMICAL TECHNOLOGY

CSC Centre for Sustainable Chemistry

ERIC European Research Institute of Catalysis

SUNERGY

Outline

- Introduction
- Super Dry Reforming
- Oxidative coupling of methane
- Conclusions and Perspectives

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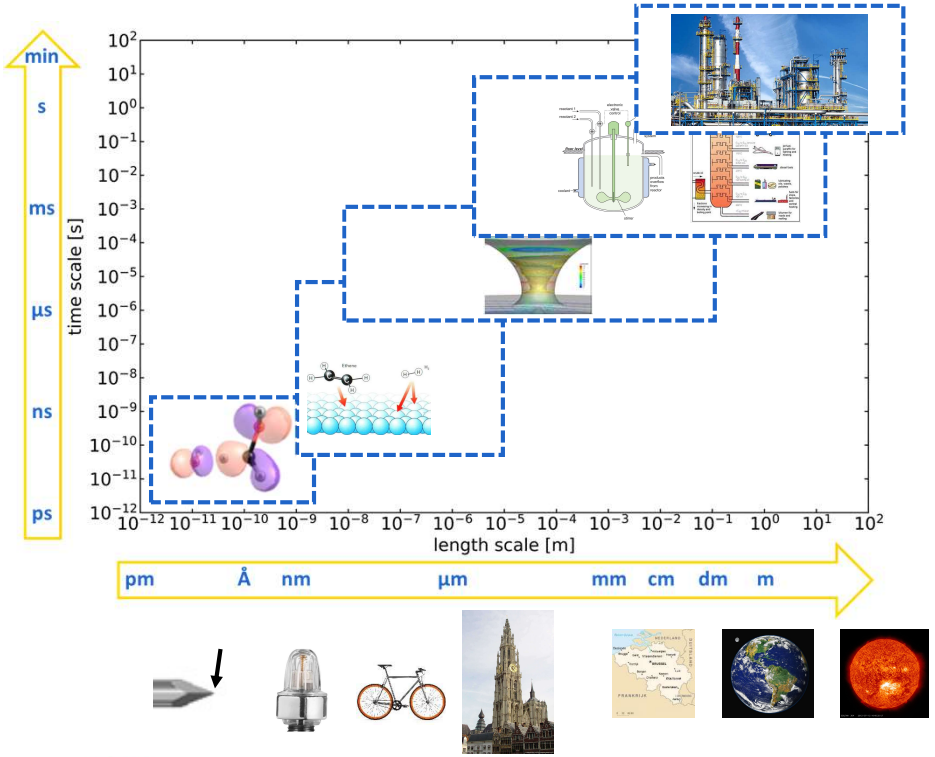
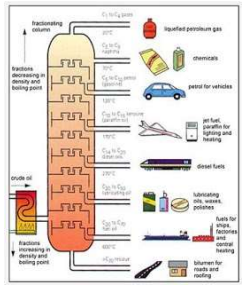
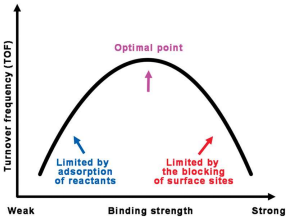
Setting the scene







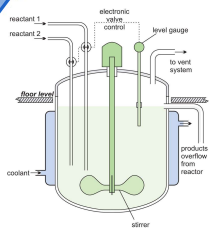
Process

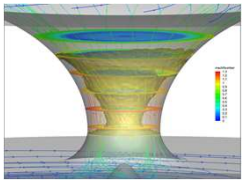
Separation Units

Transport Phenomena

Reactor

Kinetics

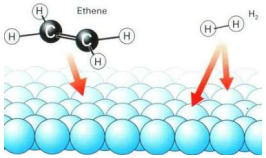


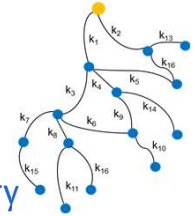


Catalyst Design

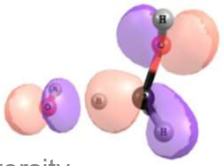
Surface Phenomena

Mechanism





Quantum Chemistry



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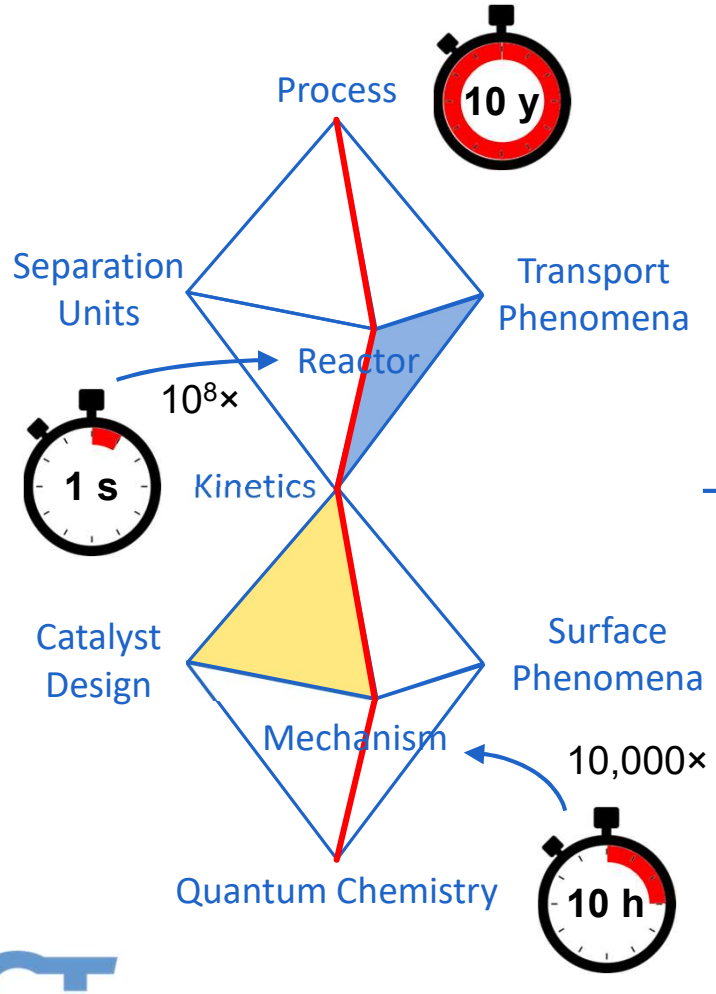
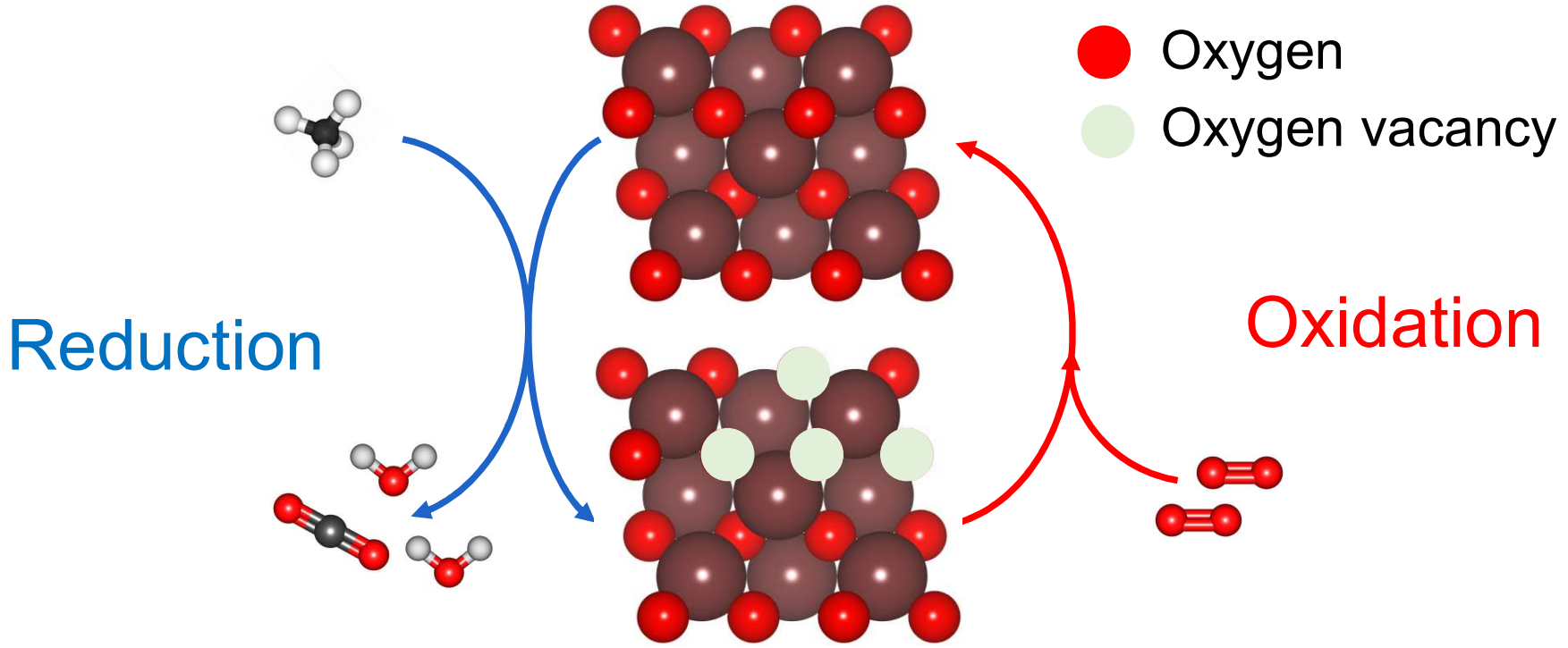
Multi-Scale Modeling



Catalysis: e.g. Mars van Krevelen **Re**d**ox** cycle



+ Diffusion of lattice oxygen from bulk to surface

Nibbelke R.H. et al., J. Catal. 1995, 156, 106 Menon U. et al., J. Catal., 2011 283,1
Alexopoulos K. et al., Appl. Catal. B 2010, 97, 381
Mathias F. et al., Angew. Chem. Int. Ed. 2018, 57 (38), 12430;
Angew. Chem. Int. Ed. 2019, 58, 2

Catalytic vs Cyclic/Chemical Looping process

$$CH_4 + 2O_2 \longrightarrow CO_2 + 2H_2O$$

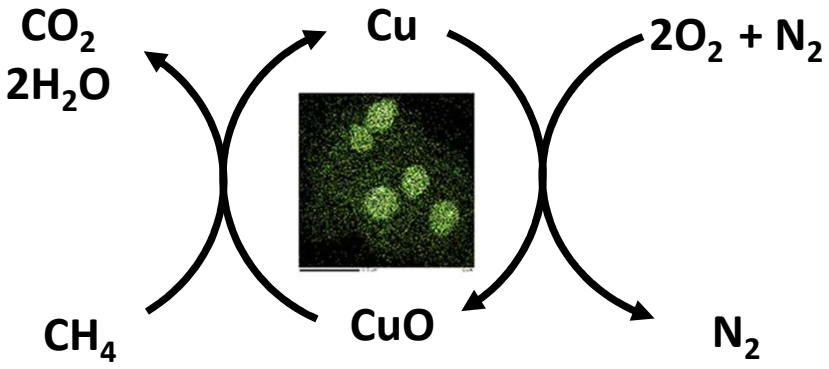
Catalytic process

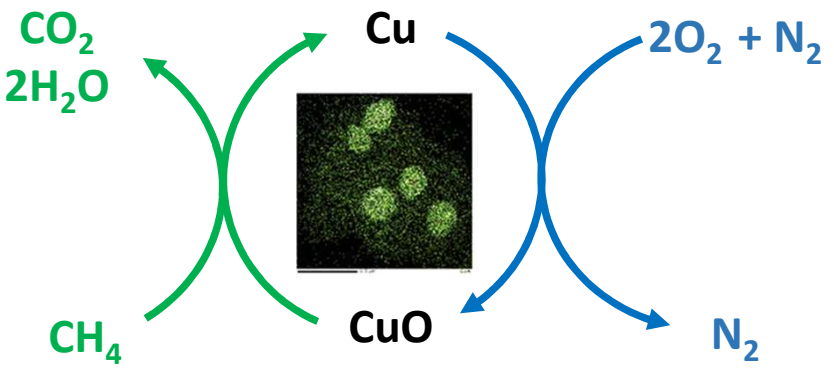
Single set of conditions

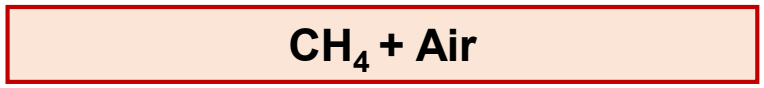
Chemical looping process

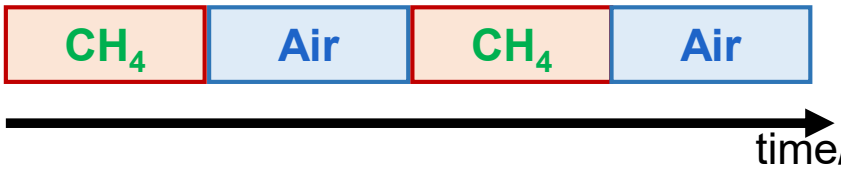
Different sets of conditions

e.g. Mars - van Krevelen mechanism









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Ishida M. et al.,"Evaluation of a CHEMICAL-LOOPING-COMBUSTION power generation system by graphic exergy analysis" Energy, 12 (1987),147

Chemical Looping allows to:

- combine reaction and separation
- improve selectivity e.g. Rashmi M. Contractor CES vol. 54 (1999) 5627-5632 : Dupont's CFB technology for maleic anhydride
- operate safely “within” explosion limits
- use cheap materials (CaO, FeO_x, …)
- circumvent equilibrium limitations
- mitigate carbon formation
- optimize heat management
- minimize exergy loss

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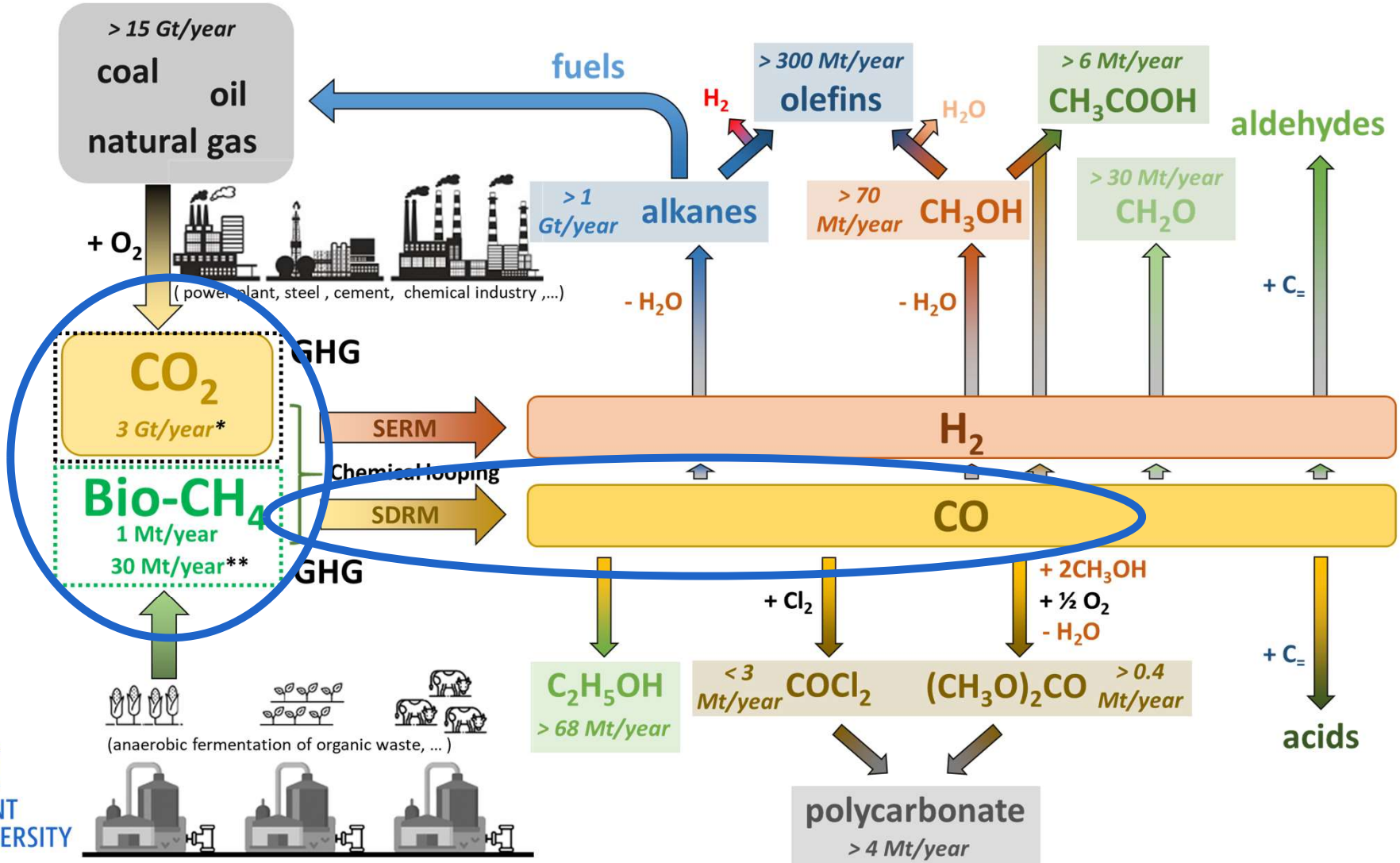
CO_2 capture and utilization (CCU)



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K. Verbeeck, L. Buelens et al., Energy & Environmental Science 11 (2018) 1788-1802 (DOI: 10.1039/C8EE01059E)

Chemical Looping: super dry reforming of CH_4

$$3CO_2 + CH_4 \rightarrow 4CO + 2H_2O$$

1: $CH_4 + 3CO_2 + Fe_3O_4 + 4CaO \rightleftarrows 2H_2O + 3Fe + 4CaCO_3$

$\Delta_r H^\circ_{1023K}$ (kJ $mol^{-1}_{CO_2}$)	$\Delta_r G^\circ_{1023K}$ (kJ $mol^{-1}_{CO_2}$)
-103	-32

2: inert + $4CaCO_3$ + $3Fe \rightleftarrows 4CaO + Fe_3O_4 + 4CO$ + inert

$\Delta_r H^\circ_{1023K}$ (kJ $mol^{-1}_{CO_2}$)	$\Delta_r G^\circ_{1023K}$ (kJ $mol^{-1}_{CO_2}$)
+212	+24

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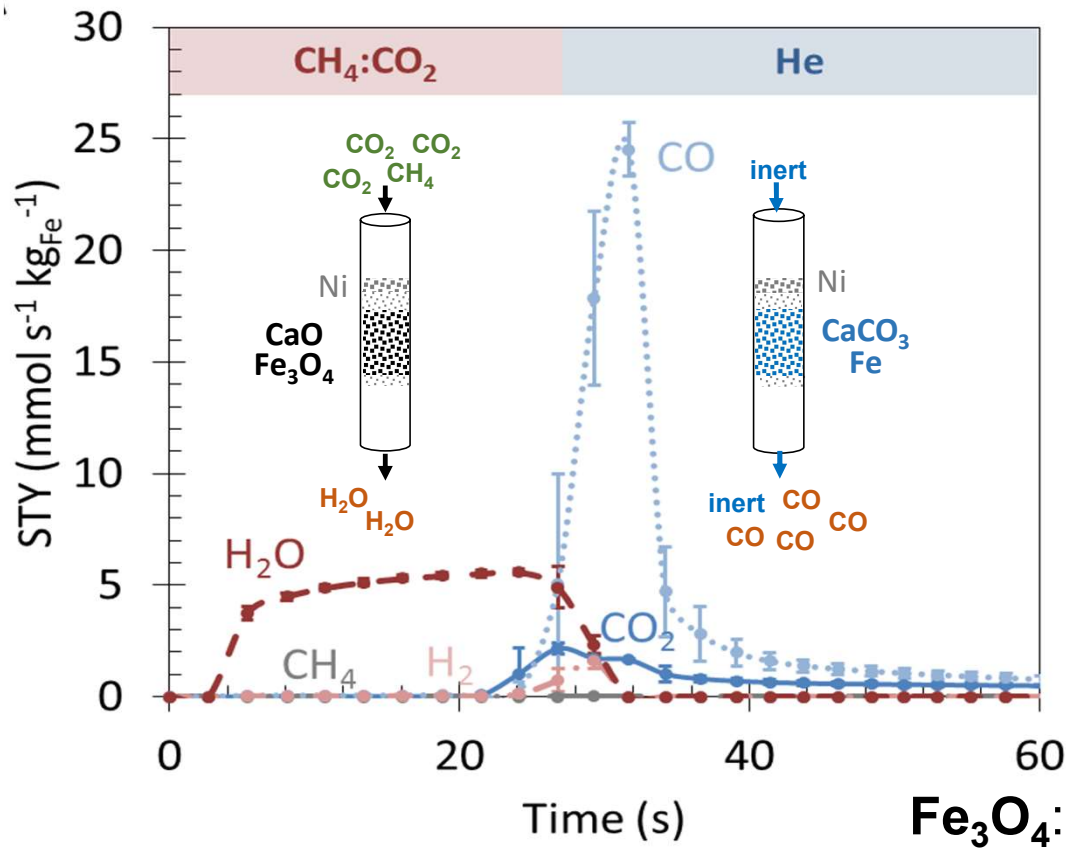
✓ Cheap materials

L. Buelens et al., Science 354 (2016) 449

T = 1023 K

Fixed bed: Permanent periodic regime

T=1023 K; CH_4:CO_2 = 1:3; after 25 cycles



Fe_3O_4: Oxygen carrier/Storage Material (OSM)

CaO : CO_2 sorbent

Ni : reforming catalyst

L. Buelens et al., Science 354 (2016) 449

Super dry reforming of CH_4

→ Step 1: $CH_4 + 3CO_2 + Fe_3O_4 + 4CaO \rightleftarrows 2H_2O + 3Fe + 4CaCO_3$

$\Delta_r H^\circ_{1023K}$ $(kJ\ mol^{-1}_{CO_2})$	$\Delta_r G^\circ_{1023K}$ $(kJ\ mol^{-1}_{CO_2})$
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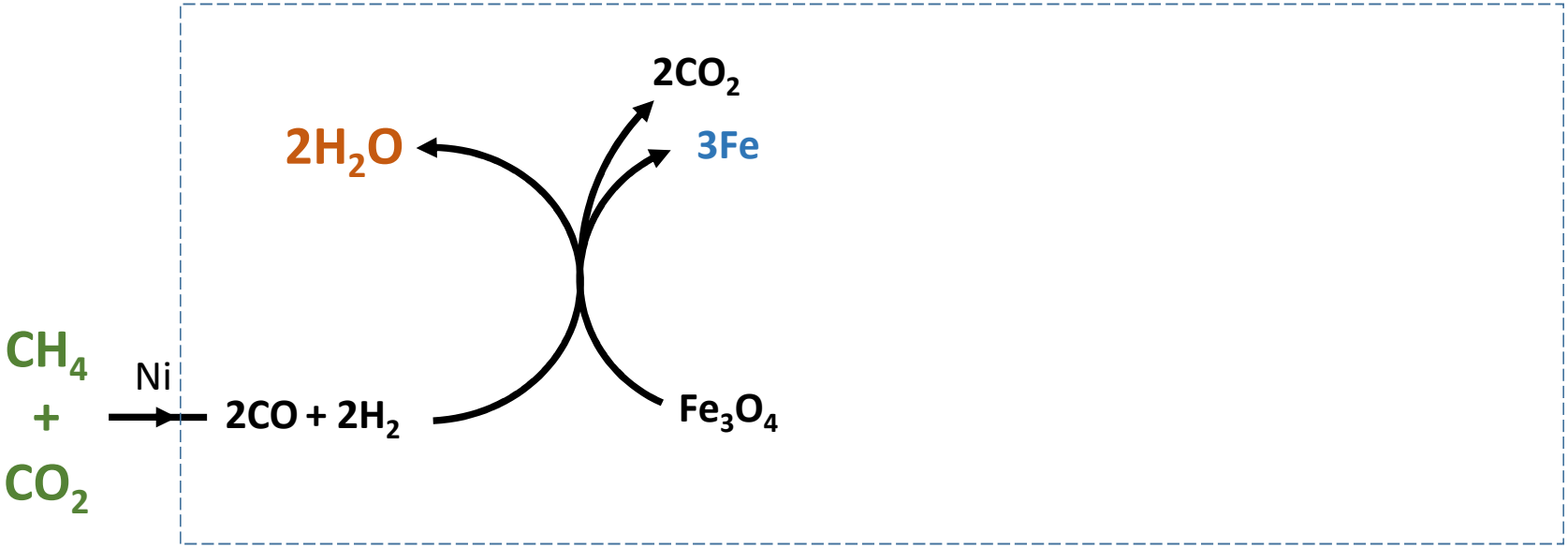

L. Buelens et al., Science 354 (2016) 449

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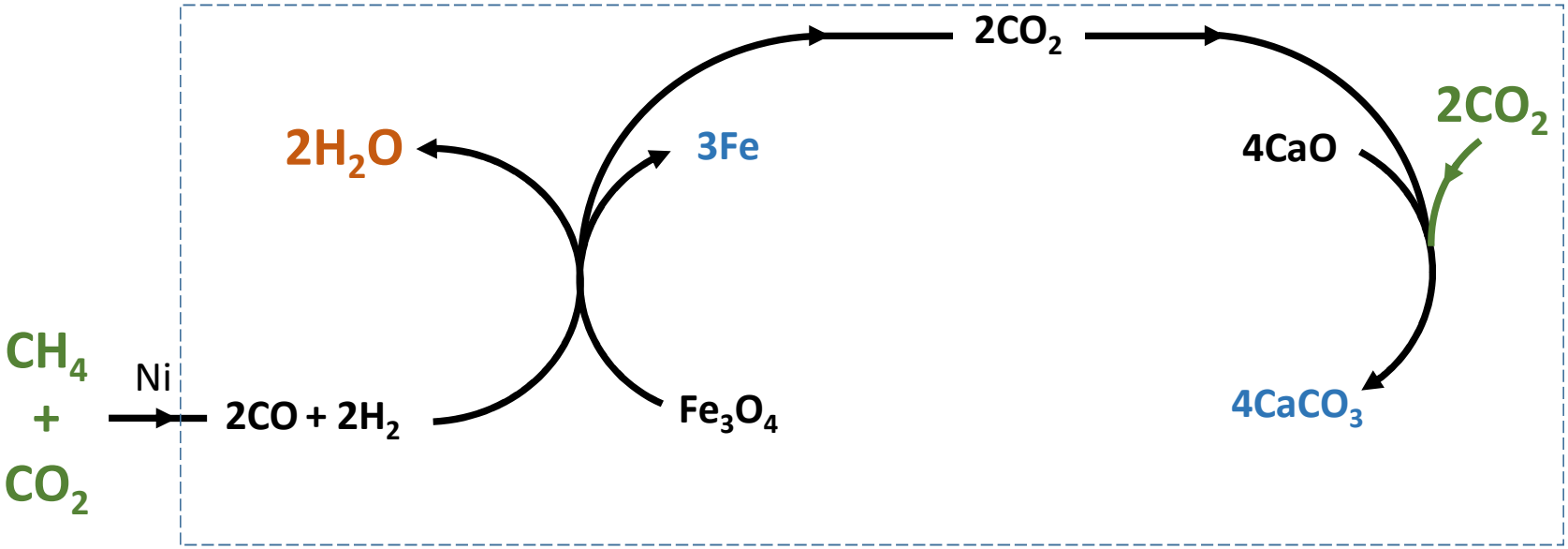

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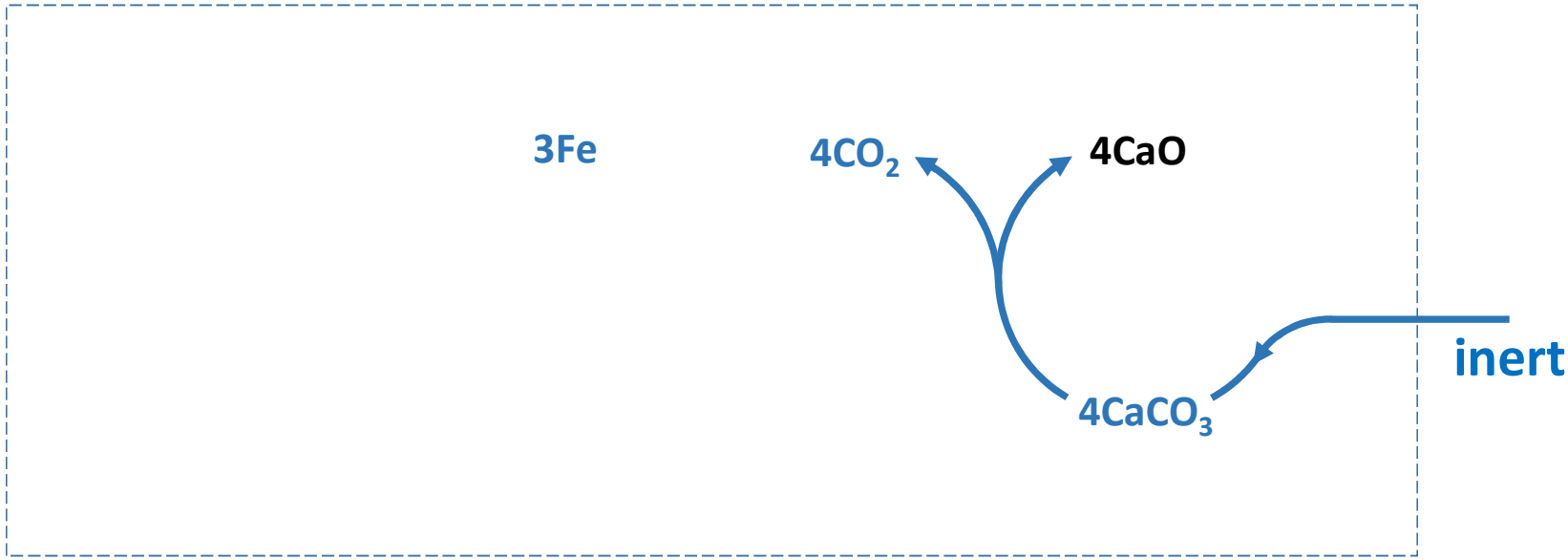
L. Buelens et al., Science 354 (2016) 449

Super dry reforming of CH_4

→ Step 1: $CH_4 + 3CO_2 + Fe_3O_4 + 4CaO \rightleftarrows 2H_2O + 3Fe + 4CaCO_3$

→ Step 2: inert + $4CaCO_3 + 3Fe \rightleftarrows 4CaO + Fe_3O_4 + 4CO$ + inert

$\Delta_r H^\circ_{1023K}$ $(kJ\ mol^{-1}_{CO_2})$	$\Delta_r G^\circ_{1023K}$ $(kJ\ mol^{-1}_{CO_2})$
-103	-32
+212	+24



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L. Buelens et al., Science 354 (2016) 449

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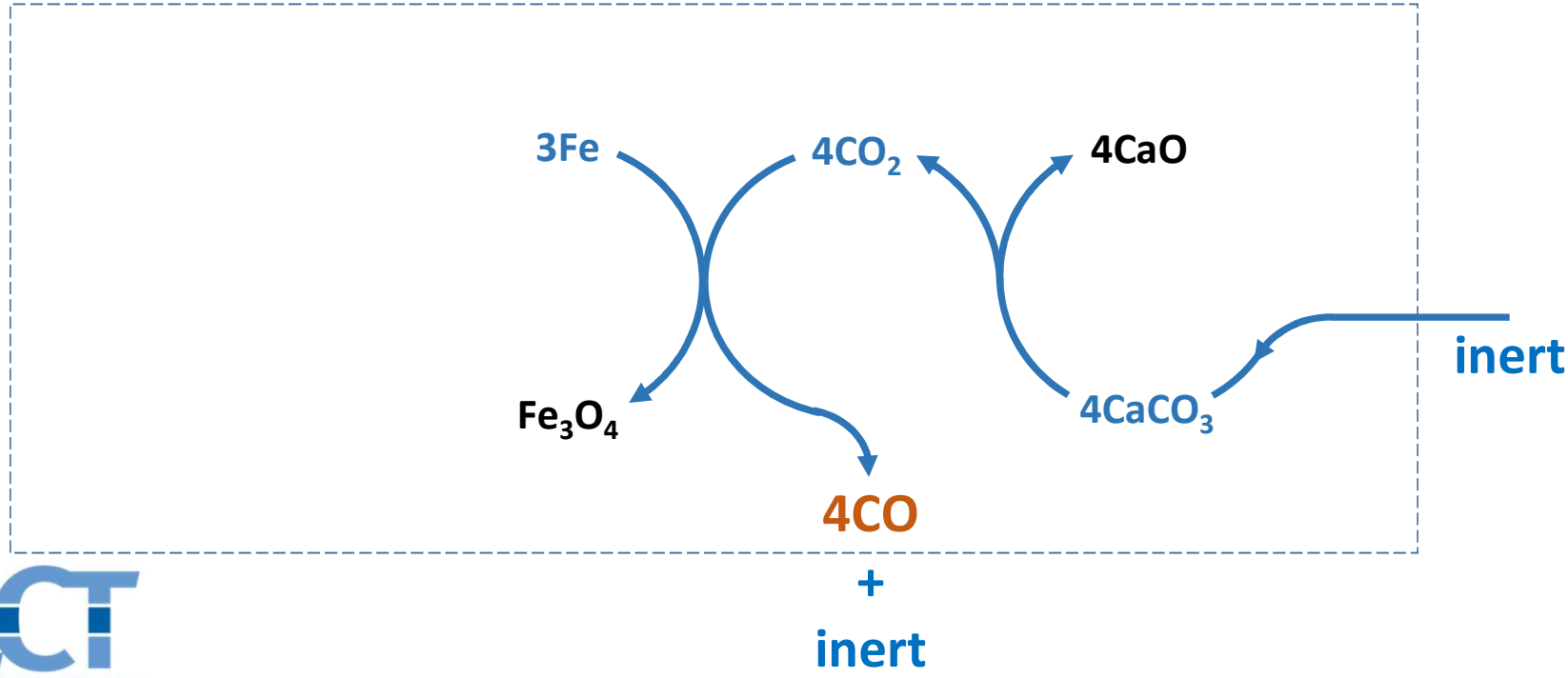
+212	+24



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Super dry reforming of CH_4

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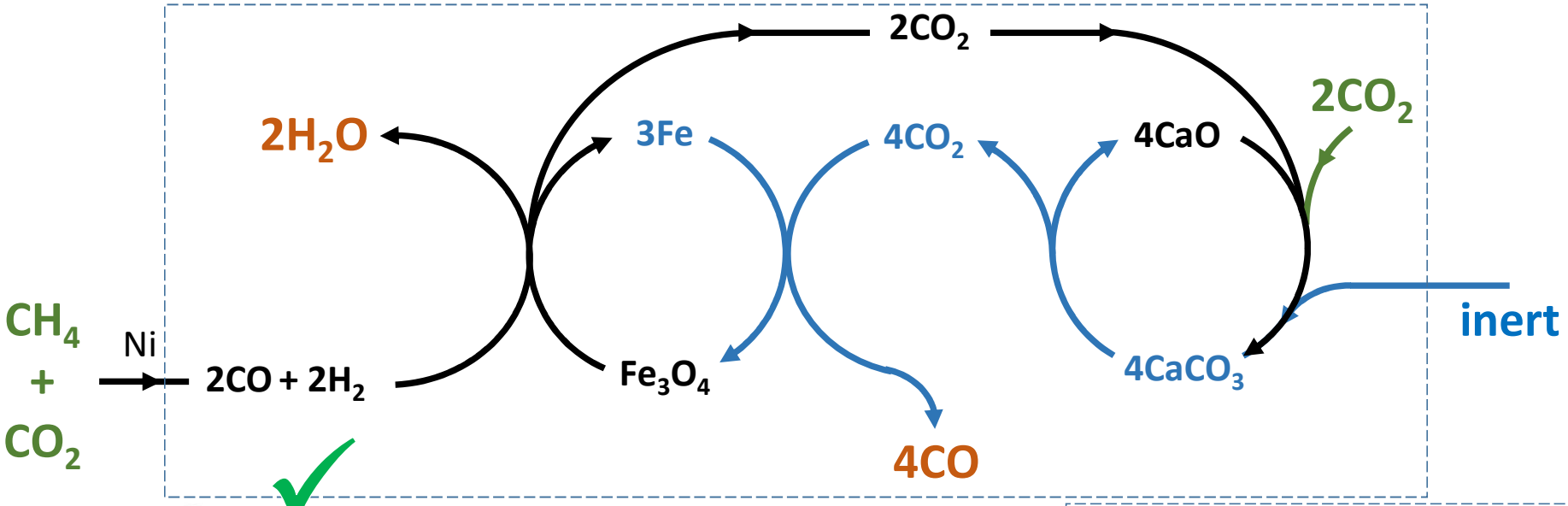
→ Step 2: inert + $4CaCO_3 + 3Fe \rightleftarrows 4CaO + Fe_3O_4 + 4CO$ + inert

$CH_4 + 3CO_2$ + inert $\rightleftarrows 4CO + 2H_2O$ + inert

$\Delta_r H^\circ_{1023K}$ (kJ $mol^{-1}_{CO_2}$)	$\Delta_r G^\circ_{1023K}$ (kJ $mol^{-1}_{CO_2}$)
-103	-32.4
+212	+23.8
+109	-8.6

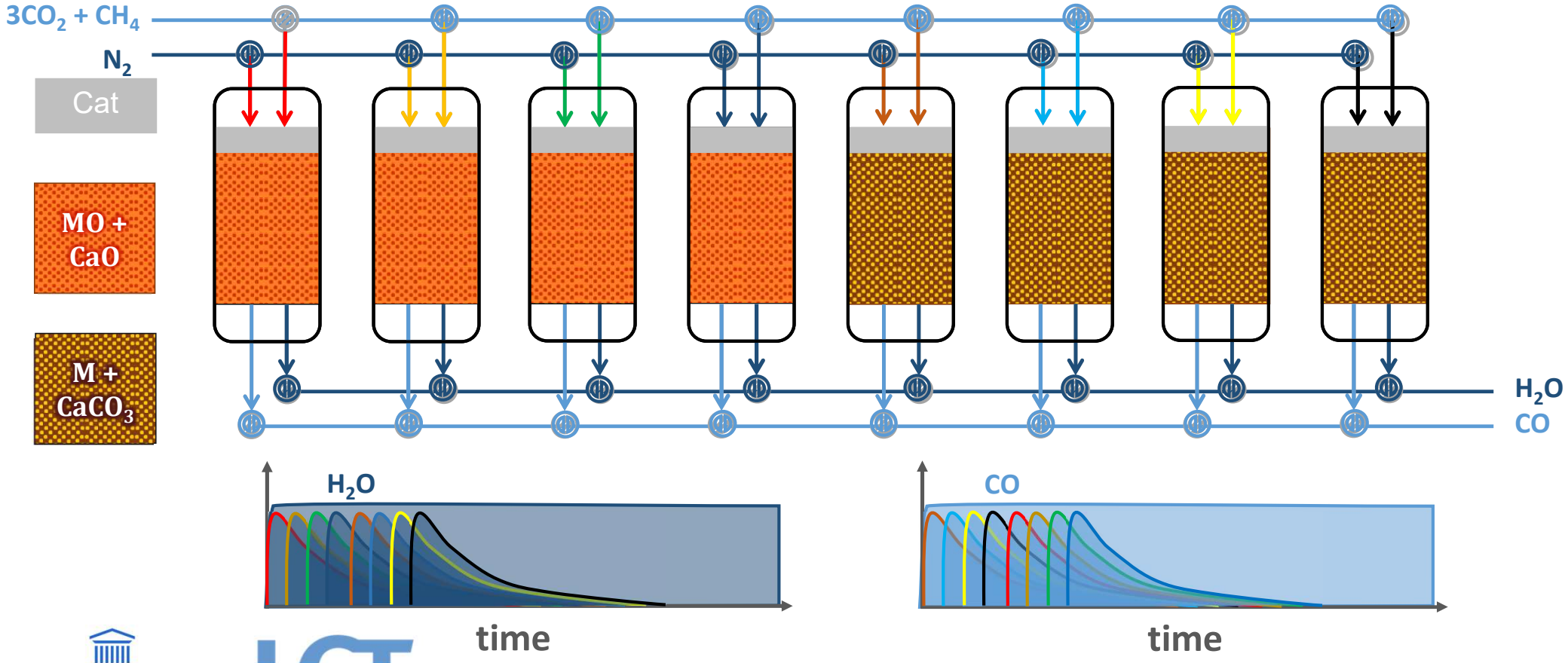


Circumvent equilibrium limitations + inert

RWGS: $2H_2 + 2CO_2 \rightleftarrows 2H_2O + 2CO$

L. Buelens et al., Science 354 (2016) 449

Permanent periodic regime: simulated moving bed



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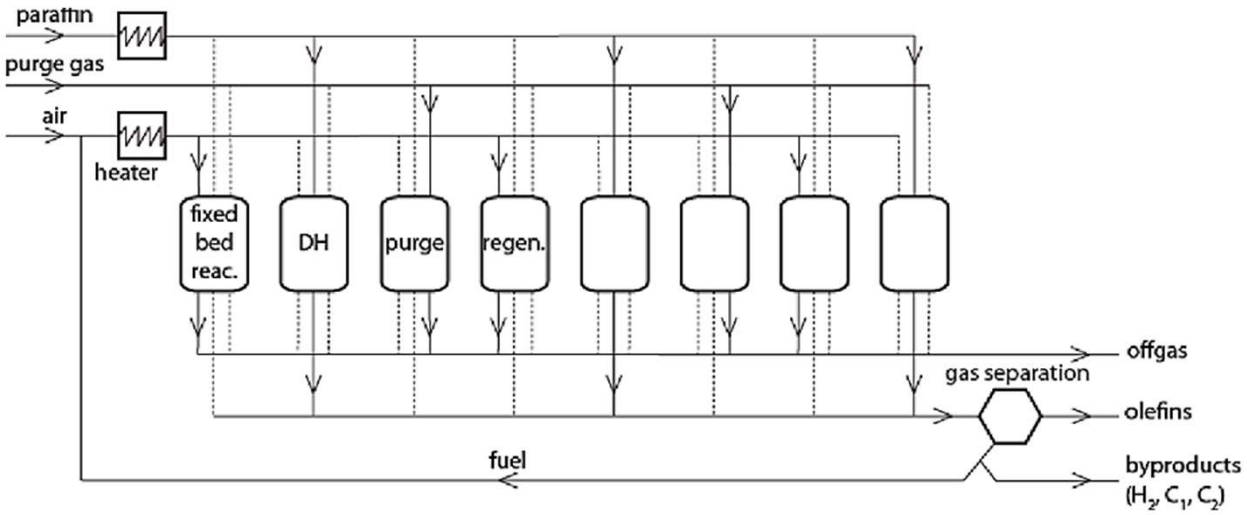
Industrial processes with similar technology

CATOFIN dehydrogenation unit



Cr_2O_3/Alumina

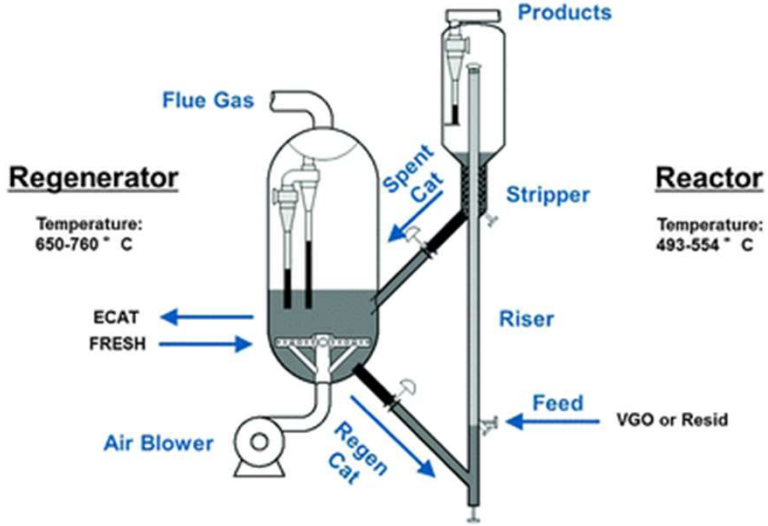
T: 575 °C

P: 0.2 - 0.5 bar

Period: hour

Sattler J.J.H.B. et al., Chem. Rev. 2014, 114, 10613

Fluid catalytic cracking (FCC)



Zeolite

T: 500 - 800 °C

Period: minute

Super Dry Reforming of CH_4

3 CO_2 per CH_4 at a CO space-time yield of 1-10 mol/m^3/s

Paul B. Weisz “The Science of the Possible: windows on reality”
CHEMTECH, July 1982, 425
Jean-Paul Lange et al. I&EC Research 2019,58,8674-8680 and
Catal. Sci. Technology 2016, 6, 4759-4767

Overall endothermic: how to reach and maintain 1023K?

Autothermal Dry Reforming

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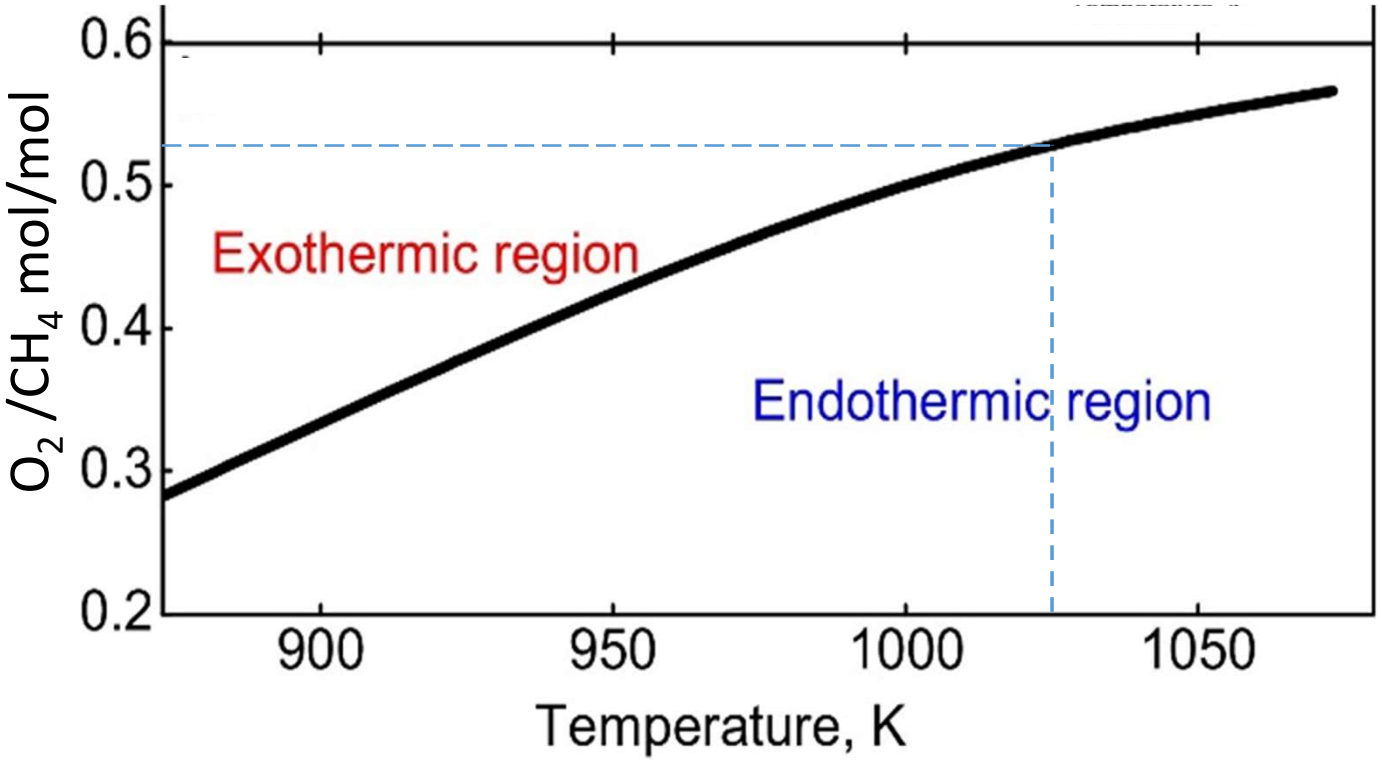
Chemical Looping with “combustion” of some methane

$CH_4 + 3CO_2 \rightleftarrows 4CO + 2H_2O$ $\Delta H^0_{1023} = 327\ kJ/mol_{CH4}$

$CH_4 + xCO_2 + (3 - x)/2O_2 \rightleftarrows (1 + x)CO + 2H_2O$ $\Delta H^0_{1023} = 0\ kJ/mol_{CH4}$

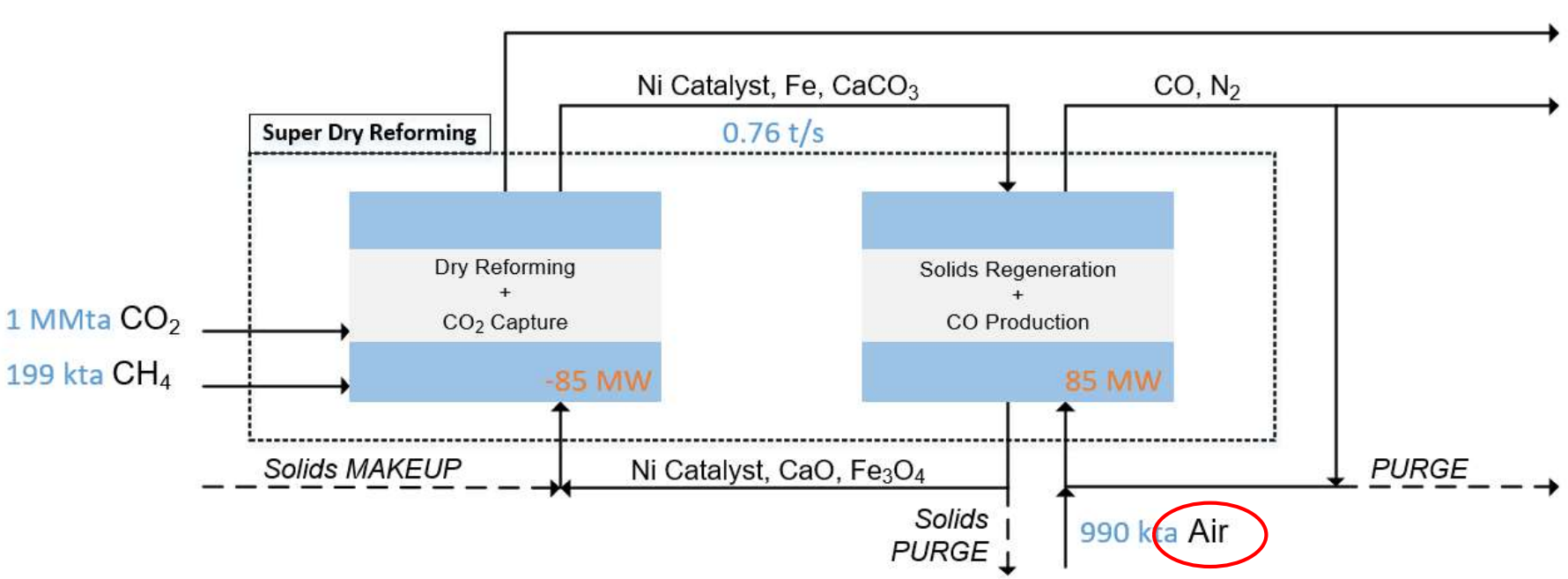
@ T=1023K

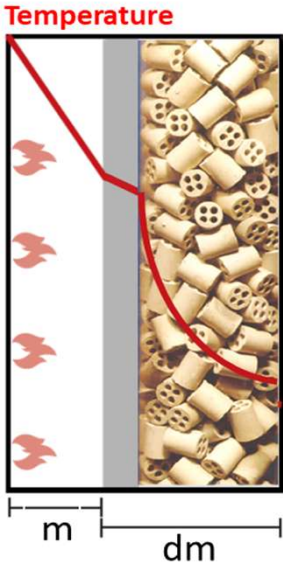
x ≈ 2



J.W. Hu et al. Journal of CO2 utilization, 16 (2016) 8-16; Appl. Cat. B, 231 (2018) 123-136

Autothermal dry reforming: flowsheet (circulating solids)





10 molCO/m3/s ➔ 100 Ton solids

✓ Optimize heat management

✓ Minimize exergy loss

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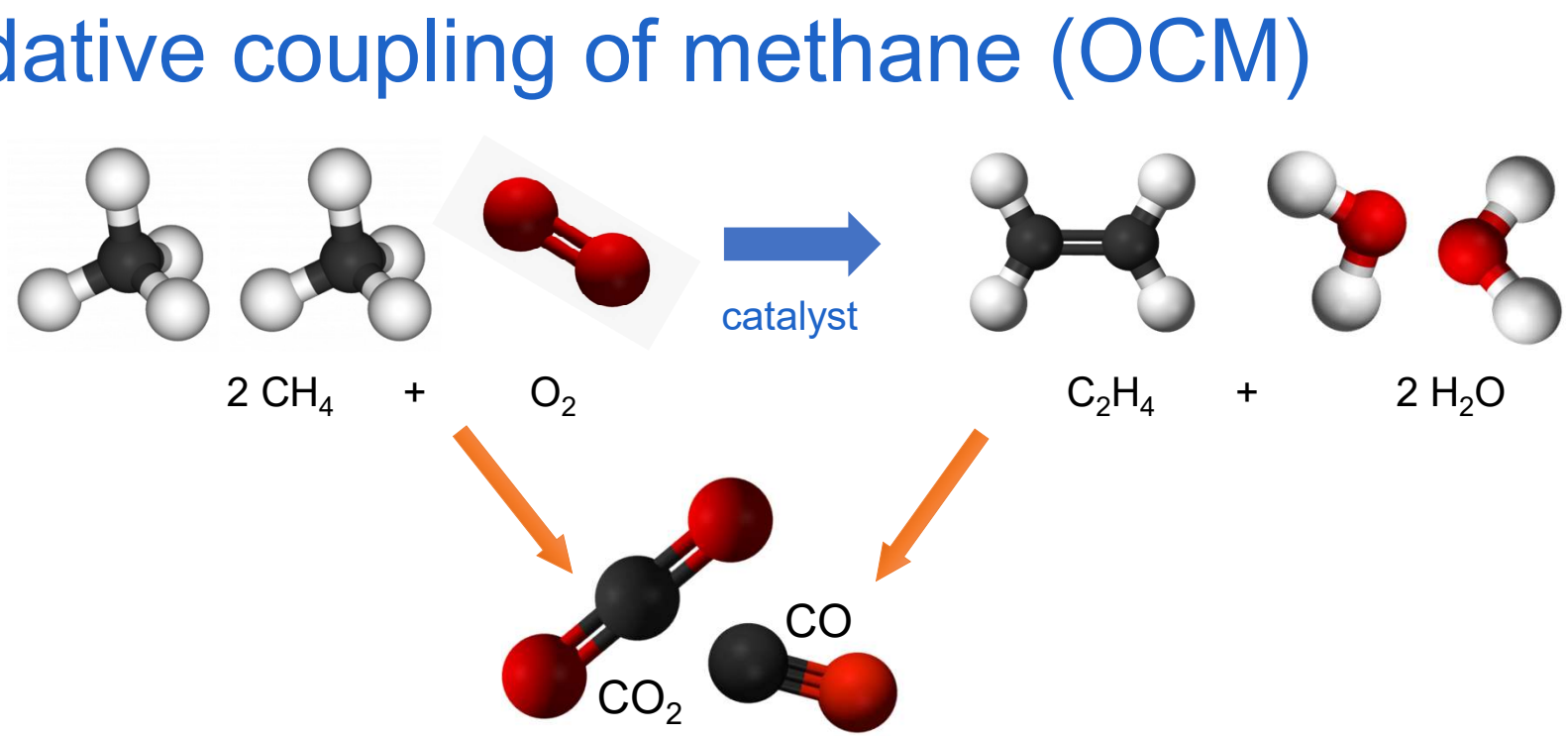
Oxidative coupling of methane (OCM)



Challenges

- × Inertness of CH_4: **high T**
- × Trade-off between conversion and selectivity: **low ethene yield**
- × High exothermicity: **heat management**

GAS PHASE CHEMISTRY

CATALYST &

REACTOR DESIGN

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Gas-Phase Reaction Network

Primary initiation

$CH_4 + O_2 \leftrightarrow CH_3\bullet + HO_2\bullet$

$CH_3\bullet$ generation

$CH_4 + H\bullet \leftrightarrow CH_3\bullet + H_2$

$CH_4 + O\bullet \leftrightarrow CH_3\bullet + OH\bullet$

$CH_4 + OH\bullet \leftrightarrow CH_3\bullet + H_2O$

$CH_4 + HO_2\bullet \leftrightarrow CH_3\bullet + H_2O_2$

$CH_3\bullet$ coupling

$CH_3\bullet + CH_3\bullet + M \leftrightarrow C_2H_6 + M$

$CH_3\bullet$ oxidation

$CH_3\bullet + O_2 \leftrightarrow CH_3O\bullet + O\bullet$

$CH_3\bullet + HO_2\bullet \leftrightarrow CH_3O\bullet + OH\bullet$

$CH_3\bullet + O_2 \leftrightarrow CH_2O + OH\bullet$

Dehydrogenation of ethane

$C_2H_6 + H\bullet \leftrightarrow C_2H_5\bullet + H_2$

$C_2H_6 + OH\bullet \leftrightarrow C_2H_5\bullet + H_2O$

$C_2H_6 + CH_3\bullet \leftrightarrow C_2H_5\bullet + CH_4$

$C_2H_6 + O\bullet \leftrightarrow C_2H_5\bullet + OH\bullet$

$C_2H_5\bullet + M \leftrightarrow C_2H_4 + H\bullet + M$

$C_2H_5\bullet + O_2 \leftrightarrow C_2H_4 + HO_2\bullet$

Hydrogen–oxygen reactions

$O_2 + H\bullet + M \leftrightarrow HO_2\bullet + M$

$H_2O_2 + M \leftrightarrow OH\bullet + OH\bullet + M$

Decomposition of C_2H_4

$C_2H_4 + O\bullet \leftrightarrow CH_3\bullet + CHO$

$C_2H_4 + OH\bullet \leftrightarrow CH_3\bullet + CH_2O$

Oxidation of $CH_3O\bullet$ and CH_2O

$CH_3O\bullet + M \leftrightarrow CH_2O + H\bullet + M$

$CH_2O + OH\bullet \leftrightarrow CHO\bullet + H_2O$

$CH_2O + HO_2\bullet \leftrightarrow CHO\bullet + H_2O_2$

$CH_2O + CH_3\bullet \leftrightarrow CHO\bullet + CH_4$

$CHO\bullet + M \leftrightarrow CO + H\bullet + M$

$CHO\bullet + O_2 \leftrightarrow CO + HO_2\bullet$

$CO + HO_2\bullet \leftrightarrow CO_2 + OH\bullet$

Dehydrogenation of ethene

$C_2H_4 + O_2 \leftrightarrow C_2H_3\bullet + HO_2\bullet$

$C_2H_4 + H\bullet \leftrightarrow C_2H_3\bullet + H_2$

$C_2H_4 + O\bullet \leftrightarrow C_2H_3\bullet + OH\bullet$

$C_2H_4 + OH\bullet \leftrightarrow C_2H_3\bullet + H_2O$

$C_2H_4 + CH_3\bullet \leftrightarrow C_2H_3\bullet + CH_4$

$C_2H_3\bullet + M \leftrightarrow C_2H_2 + H\bullet + M$

$C_2H_3\bullet + O_2 \leftrightarrow C_2H_2 + HO_2\bullet$

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Experimental conditions:

p_t= 1-10 bar ; T = 873–1123 K; CH_4/O_2 I_0= 4–10; He/CH_4 I_0 = 0–1.25; $V / F_{CH4,0}$ = 0.1–1.9 m^3 s mol^{-1};
CH_4 conversion = 2–15%, ***O_2 conversion*** = 10–100%

Chen, Qi, et al. Industrial & Engineering Chemistry Research 30.9 (1991): 2088–2097
Chen, Qi, et al. AIChE journal 40.3 (1994): 521–535

Surface Reaction Network

HYDROGEN ABSTRACTION BY ELEY–RIDEAL STEPS

$CH_4 + O* \leftrightarrow CH_3^{\bullet} + OH*$

$C_2H_4 + O* \leftrightarrow C_2H_3^{\bullet} + OH*$

$C_2H_6 + O* \leftrightarrow C_2H_5^{\bullet} + OH*$

$C_2H_5^{\bullet} + O* \leftrightarrow C_2H_4 + OH*$

$CH_3O^{\bullet} + O* \leftrightarrow CH_2O + OH*$

$CH_2O + O* \leftrightarrow CHO^{\bullet} + OH*$

$CHO^{\bullet} + O* \leftrightarrow CO + OH*$

$H_2 + O* \leftrightarrow H^{\bullet} + OH*$

$H_2O_2 + O* \leftrightarrow HO_2^{\bullet} + OH*$

$OH^{\bullet} + O* \leftrightarrow O^{\bullet} + OH*$

$H_2O + O* \leftrightarrow OH^{\bullet} + OH*$

$HO_2^{\bullet} + O* \leftrightarrow O_2 + OH*$

HYDROGEN ABSTRACTION BY SURFACE REACTION

$CH_3O* + O* \leftrightarrow CH_2O* + OH*$

$CH_2O* + O* \leftrightarrow HCO* + OH*$

$CHO* + O* \leftrightarrow CO* + OH*$

$C_2H_4O* + O* \leftrightarrow C_2H_3O* + OH*$

RECOMBINATION OF HYDROXYLS

$2OH* \leftrightarrow H_2O* + O*$

CO OXIDATION ON THE SURFACE

$CO* + O* \leftrightarrow CO_2* + *$

C–C CLEAVAGE ON THE SURFACE

$C_2H_3O* + O* \leftrightarrow CH_2O* + HCO*$

ADSORPTION STEPS

$O_2 + 2* \leftrightarrow 2O*$

$CH_3^{\bullet} + O* \leftrightarrow CH_3O*$

Sn/Li/MgO

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Nibbelke R.H. et al., J. Catal.,(156), 1995, 106-119

Kechagiopoulos, P. N., et al., Ind. Eng. Chem. Res., (53), 2013, 1825–1840

Catalyst and Kinetic Descriptors

- Reaction families:

H abstraction by Eley−Rideal reaction

H abstraction by surface reaction

Recombination of OH

CO oxidation on the surface

C−C cleavage on the surface

- Polanyi relation within a reaction family:

activation energy: $E_{a,i} = E_0 + \alpha \Delta_{r,i}H$

  - kinetic descriptors:α, E_0
  - a reaction family shares the same (E_0,α)

- Reference: abstraction from Methane

$D_1 = \Delta_{r,1}H$ catalyst descriptor

- Any reaction i in the family: e.g. from Ethane

$\Delta_{r,i}H = D_1 + Q_{C2H5–H} - Q_{CH3–H}$

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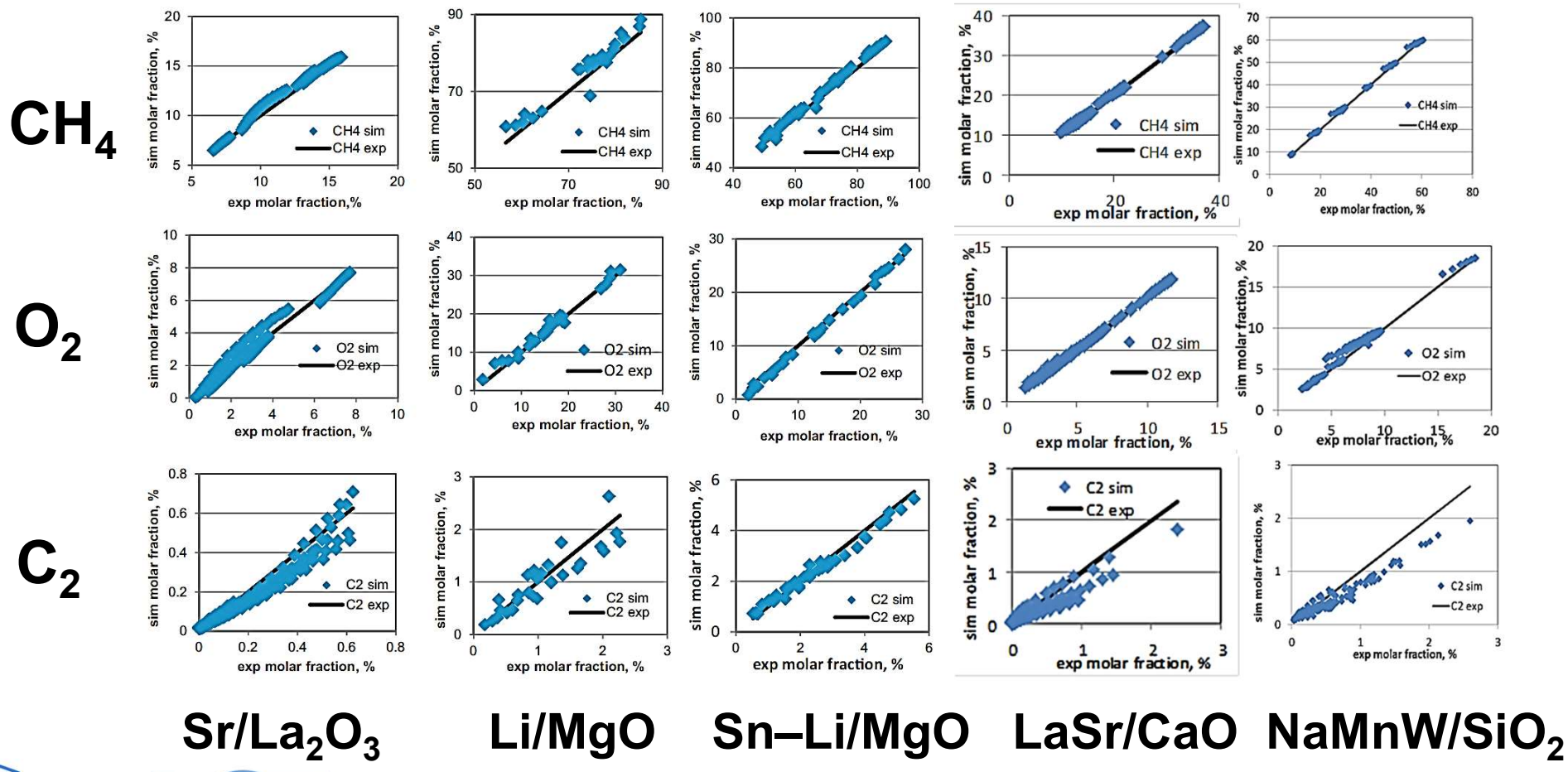
P.N. Kechagiopoulos et al., Ind. Eng. Chem. Res., (53), 2014, 1825–1840

Gas phase/surface reactions microkinetics



Alexiadis, V. I., et al. Applied Catalysis B: Environmental 150 (2014) 496–505

Catalyst descriptors

D1
Reaction enthalpy of H abstraction (kJ/mol_{CH4})

D2
Chemisorption heat O_2 (kJ/mol_{O2})

D3
Sticking probability of CH_3 on O* (-)

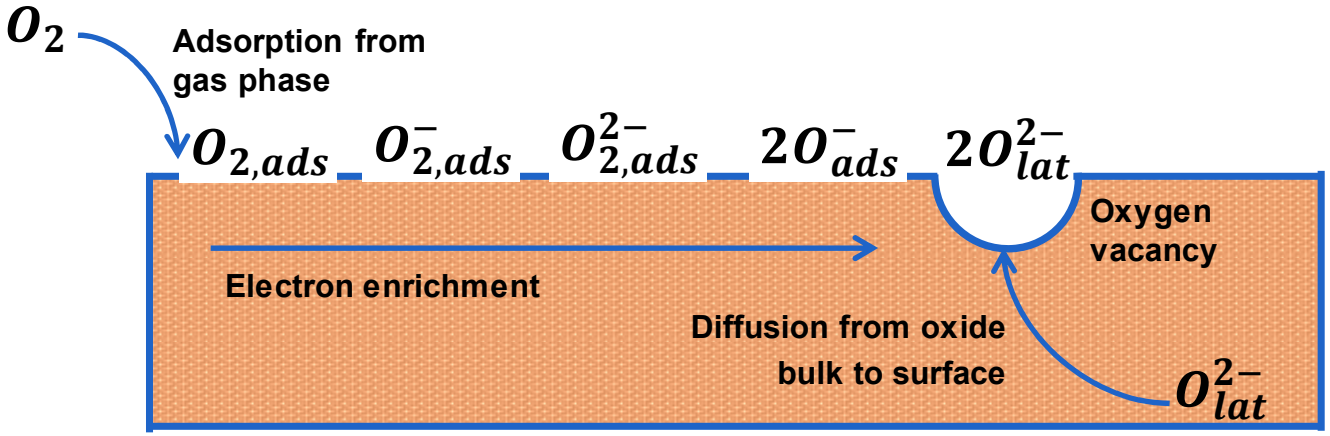
D4
Active sites Density (mol/m^2)

$CH_4 + O* \leftrightarrow CH_3^{\bullet} + OH*$

$O_2 + 2* \leftrightarrow 2O*$

$CH_3^{\bullet} + O* \leftrightarrow CH_3O*$

$\boldsymbol{Oxygen\ Pool\ Capacity}$



Nibbelke R.H. et al., J. Catal. 156), 1995, 106-119

Pirro, L. et al., Catal. Sci. Technol. (9), 2019, 3109-3125

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Catalyst descriptors and catalyst performance

	D_1	D_2	D_3	D_4
Unit	kJ mol^{-1}	kJ mol^{-1}	10^{-4}	10^{-6}mol m^{-2}
Sr/La_2O_3	44.4±0.2	119.5±3.5	6.5 ±0.5	10 ± 1
Li/MgO	91.2±0.2	73.6±2.2	1.1900±0.0002	0.4330 ±0.0009
Sn–Li/MgO	56.6±0.8	60.5±2.6	0.62±0.01	1.33 ±0.03
LaSr/CaO	65.1±1.6	139.6±5.3	1.14±0.03	6.4±1.5
$NaMnW/SiO_2$	81.400±0.002	44.0±1.6	0.10±0.02	0.457±0.005

Value ±95% confidence interval

HIGH ACTIVITY:
low D1, high D3,D4

LOW ACTIVITY:
high D1, low D4

HIGH SELECTIVITY:
low D2,low D3

D_1 – Reaction enthalpy of hydrogen abstraction from CH_4
D_2 – Chemisorption heat of O_2
D_3 – Initial sticking probability of CH_3
D_4 – Density of active sites

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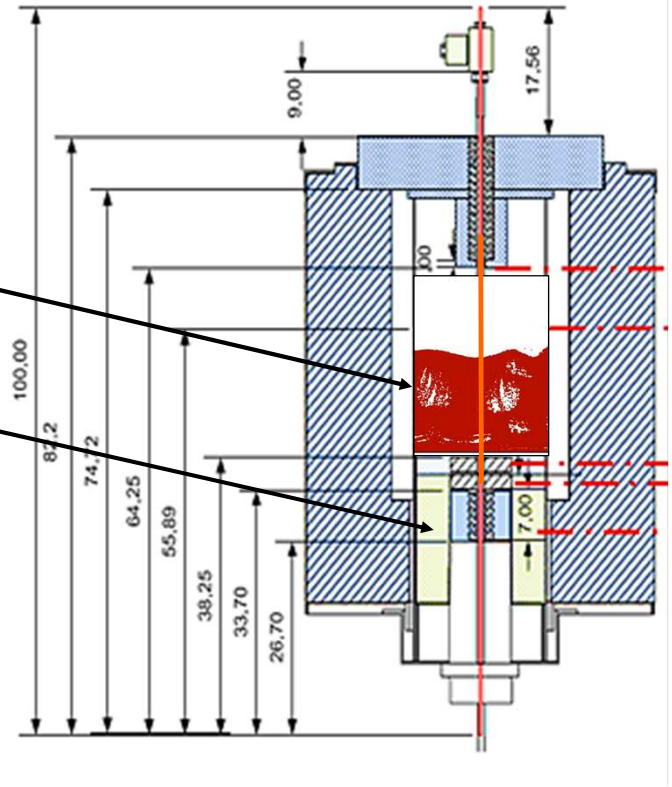
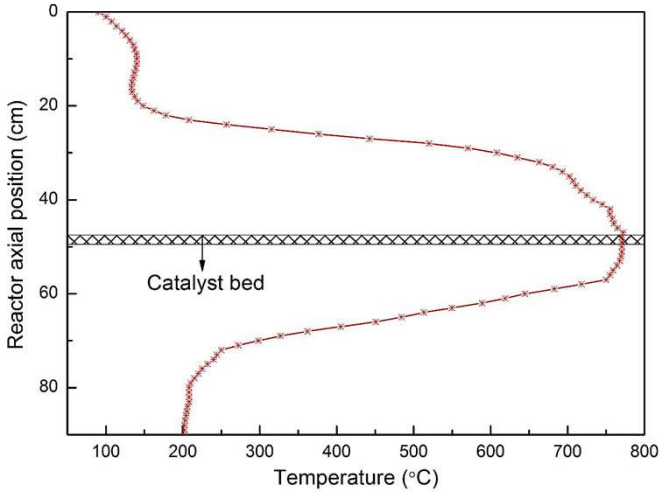
Alexiadis, VI, et al. Applied Catalysis B-Environmental , 199,252-259,2016

Experimental Set-Up: isothermal conditions

Fluidized sand bath

Air distributor





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Chemical Engineering and Catalysis

- Introduction
- Super Dry Reforming
- Oxidative coupling: heat transfer
- Conclusions and Perspectives

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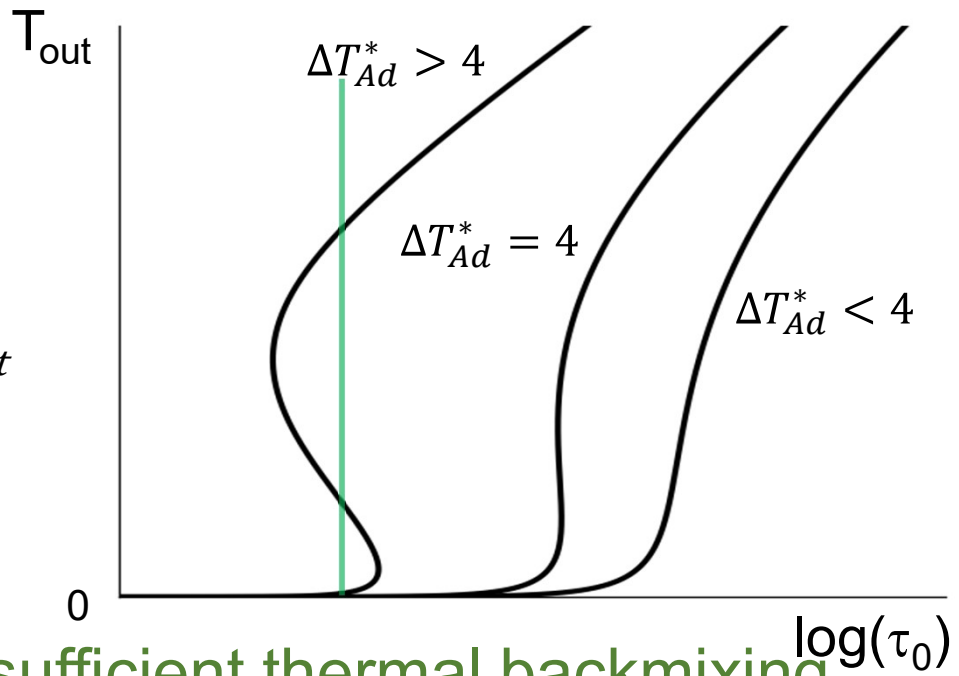
Exothermic reactions: steady-state multiplicity

Unfolding parameter: e.g. dimensionless adiabatic temperature rise ΔT_{Ad}^*

$$\Delta T_{Ad}^* = \frac{E}{RT_0^2}\Delta T_{Ad}$$

State variable:

e.g. *outlet temperature* T_{out} *or conversion* X_A



Positive feedback required: e.g. sufficient thermal backmixing

Bifurcation parameter: e.g. *inlet temperature* T_{in} *or* $\tau_0 = \frac{V}{F^0_{V(STP),CH_4}}$

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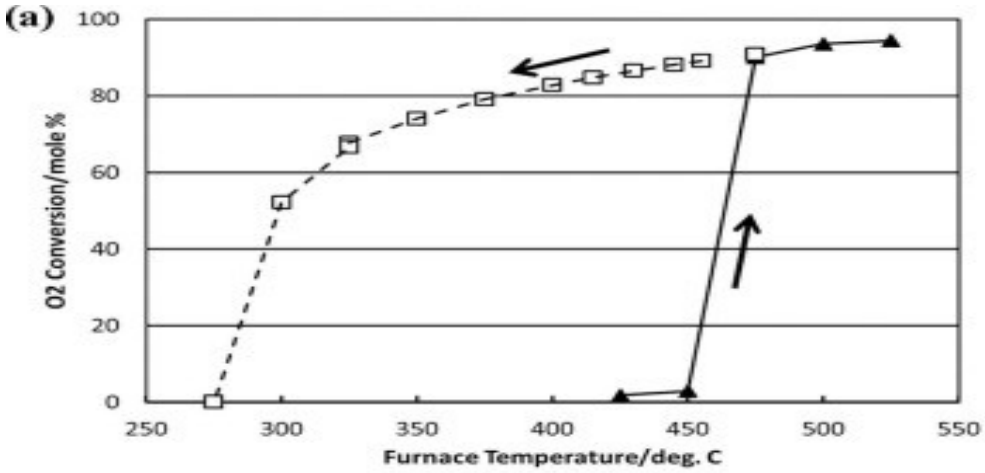
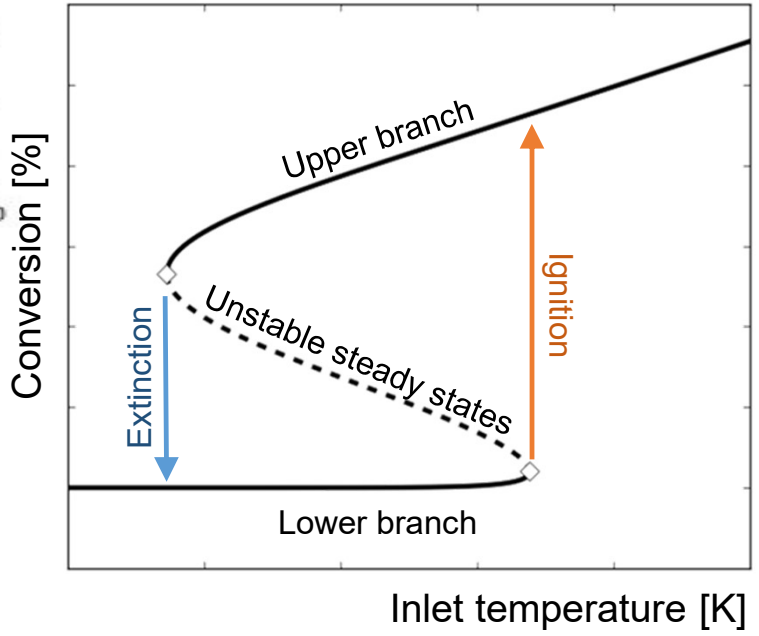
Steady-state multiplicity: Ignition/Extinction



S. Sarsani et al., Chem. Eng. J., (328), 2017, 484-496.



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Outline

- Introduction
- Super Dry Reforming
- Oxidative coupling: reactor design
- Conclusions and Perspectives

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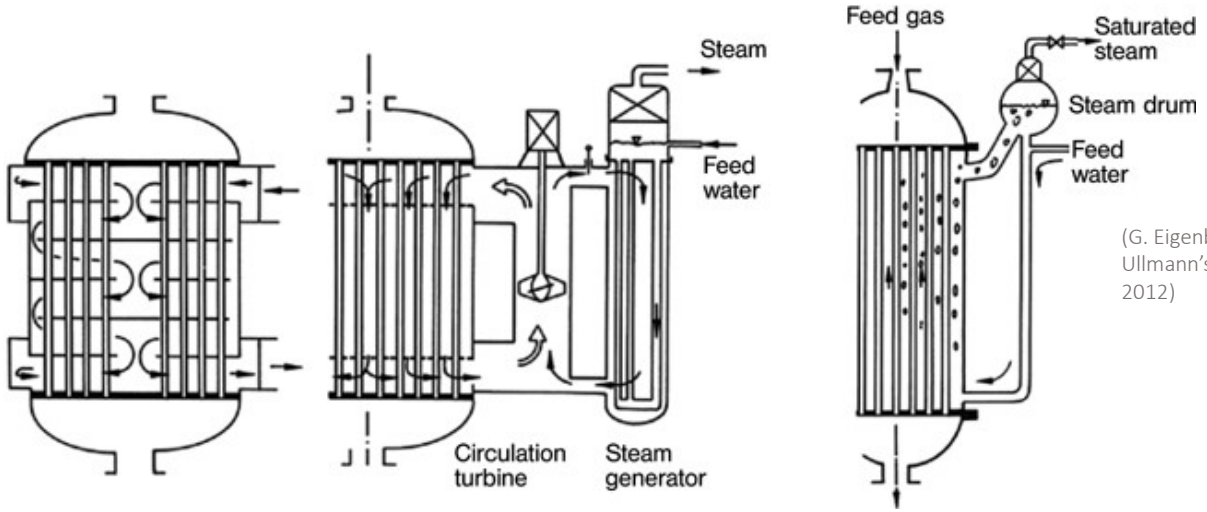
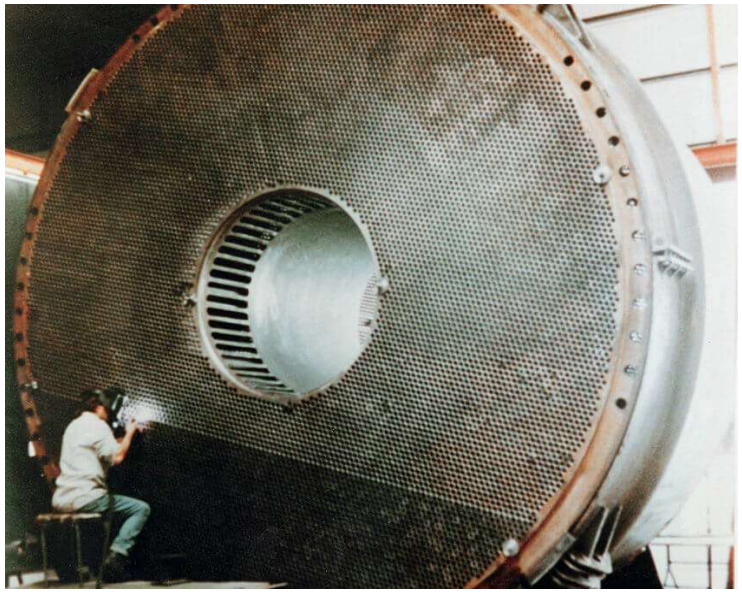
Industrial reactor : heat management by cooling?

cooled, multi-tubular reactor



(G. Eigenberger & W. Ruppel, Ullmann's Encycl. Ind. Chem., 2012)



(http://www.technobell.eu/chemical-process-technologies/phthalic-anhydride/)

> 200 multi-tube reactors (of the largest size possible ≅ 50000 tubes/reactor) for a single OCM plant

(J.H.B.J. Hoebink et al., Chem. Eng. Sci., 1994)
(L. Chen et al., Chem. Eng. J., 2020)

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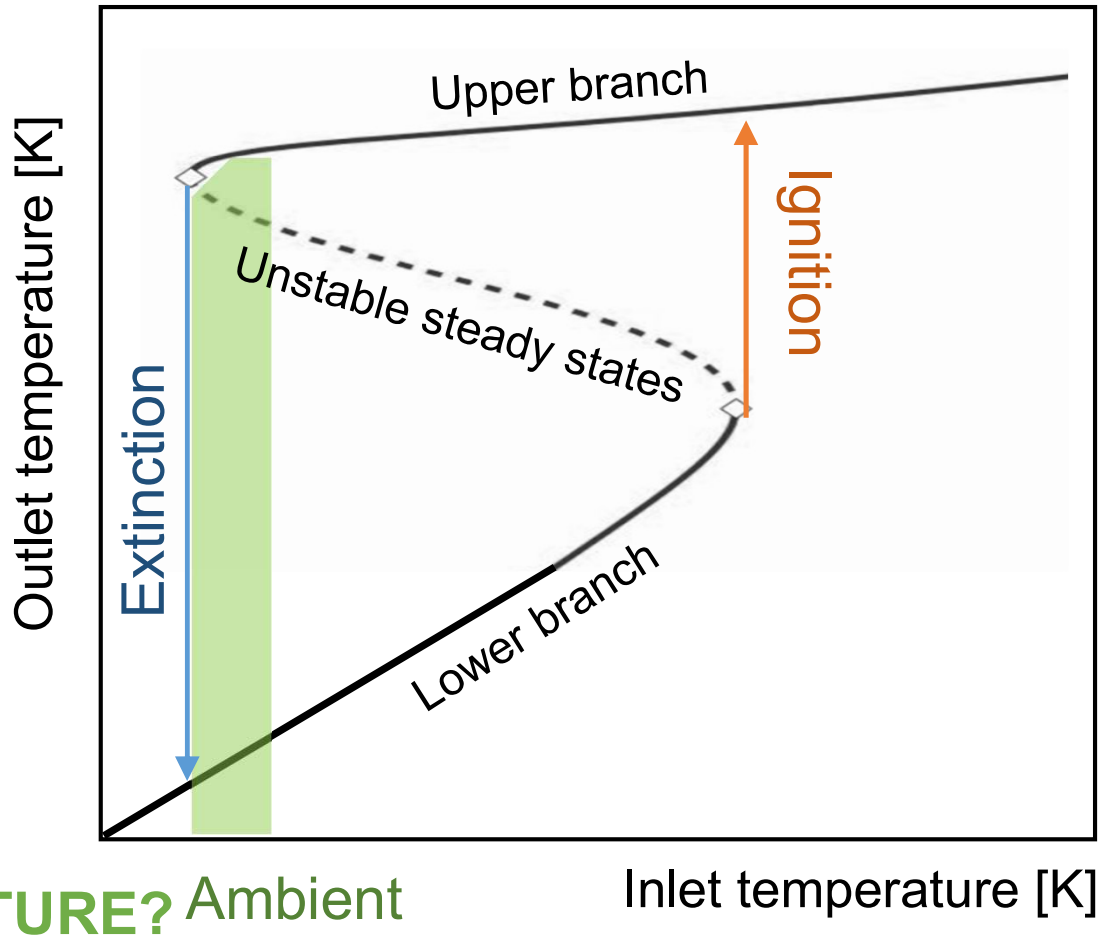
Industrial reactor : heat management



AUTOTHERMAL ADIABATIC OPERATION WITH AMBIENT INLET TEMPERATURE?

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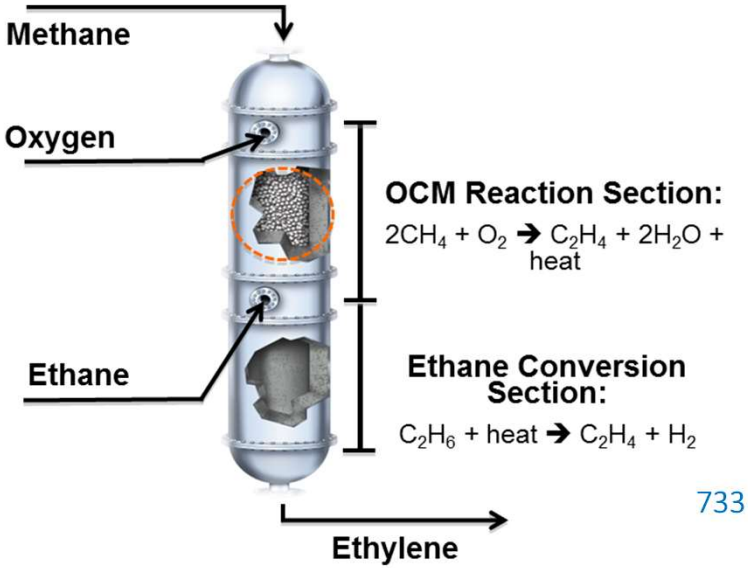
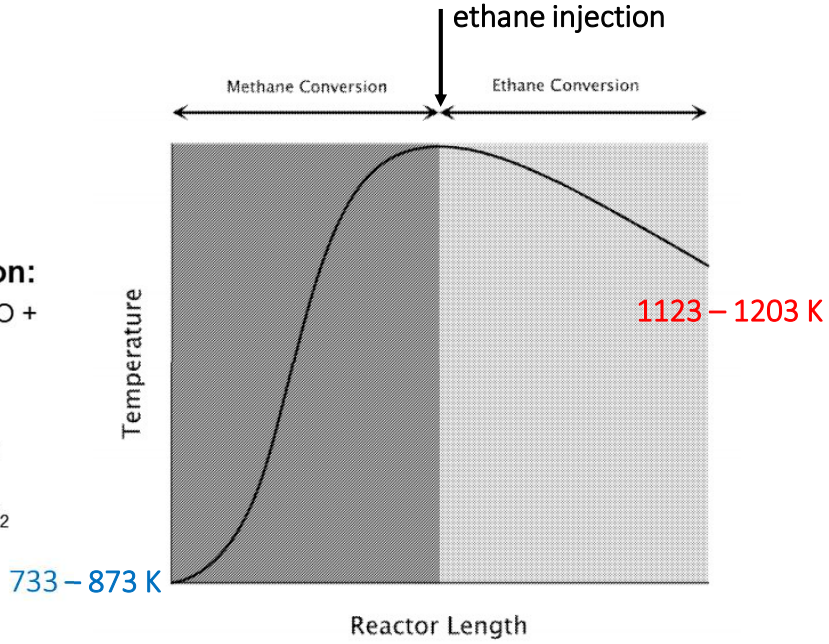
Adiabatic packed-bed reactors

siluria TECHNOLOGIES

http://siluria.com/Technology/Demonstration_Plant





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PETROCHEMICALS

McDermott buys Siluria for oxidative methane-coupling technology

by Alexander H. Tullo

AUGUST 9, 2019 | APPEARED IN VOLUME 97, ISSUE 32

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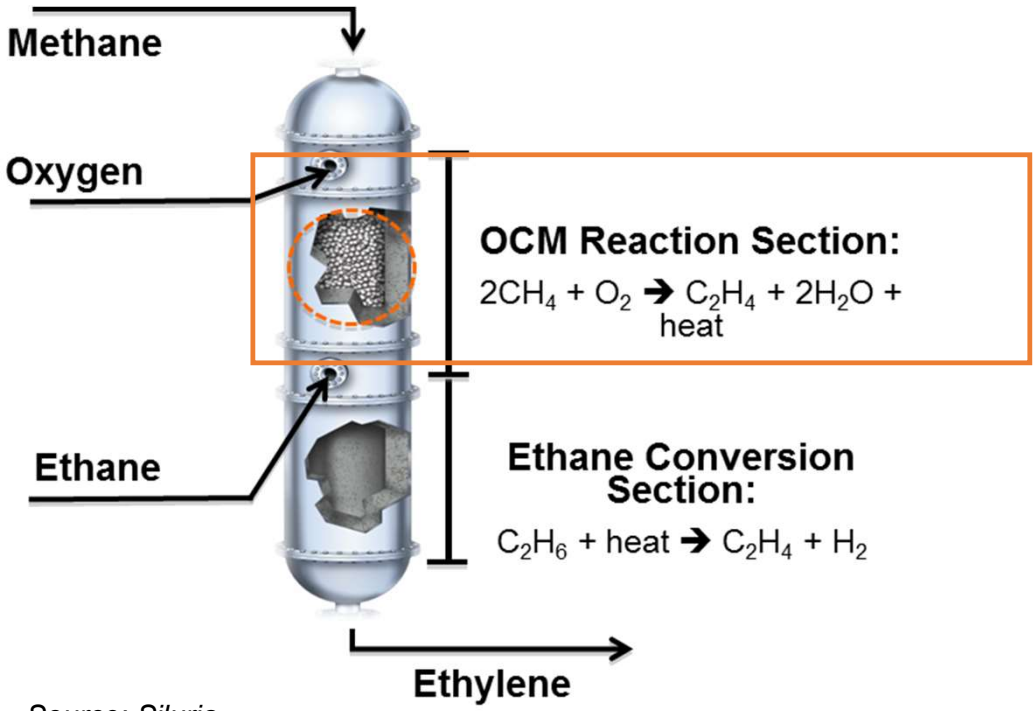
Adiabatic packed bed reactor

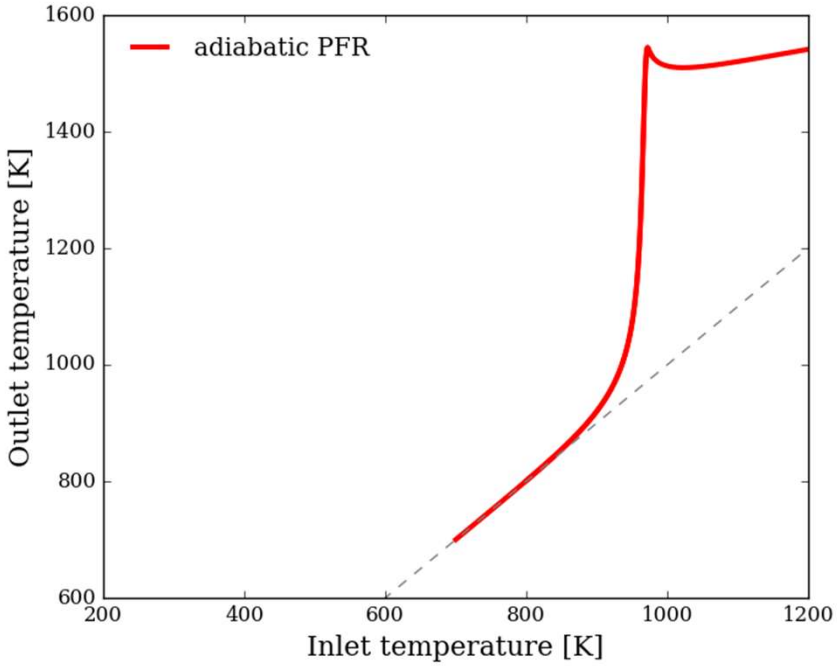


Source: Siluria

Plug flow reactor (PFR)

Sr/La_2O_3

$CH_4:O_2 = 6$, $V/F^0_{V(STP),CH4} = 0.01$ s, $m_{cat}/V = 1000$ kg_{cat}/m^3



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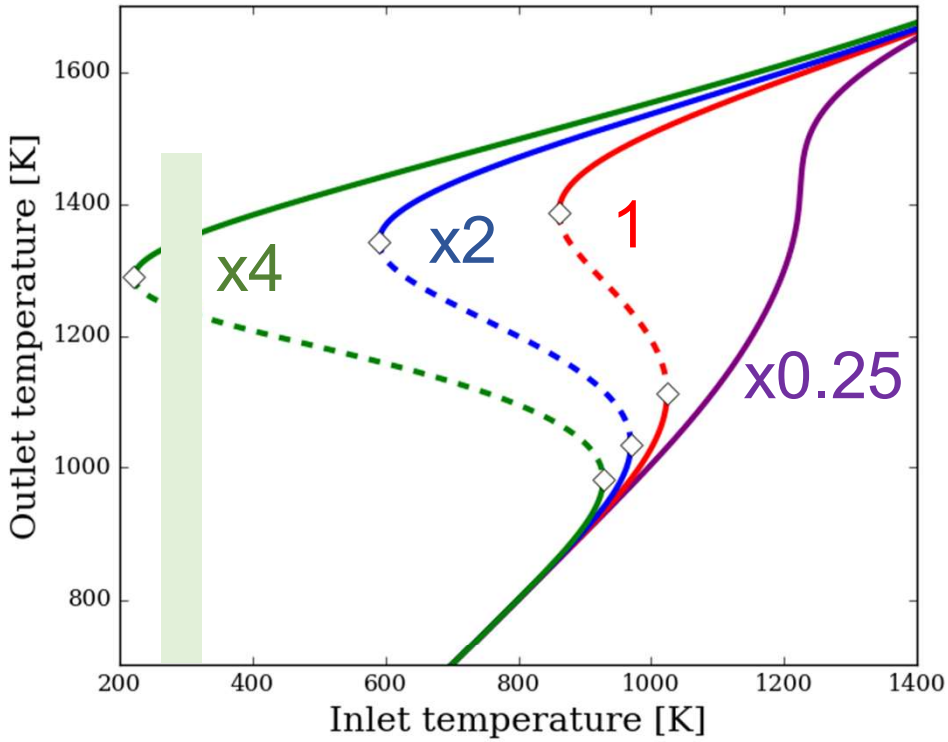
L.A. Vandewalle et al., Chem.Eng.Sci., (199), 2019, 635-651.

Adiabatic Continuous Stirred Tank Reactor (CSTR)



ambient

Sr/La_2O_3

P = 1 bar, CH4:O2 = 4 (no dilution),
V/F0V(STP),CH4 = 0.003 s

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Ideal reactor types

- Plug flow reactor **(PFR)**

  No species backmixing, no thermal backmixing

- Continuously stirred tank reactor **(CSTR)**

  Perfect species backmixing, perfect thermal backmixing

- Lumped thermal reactor **(LTR)**

  No species backmixing, perfect thermal backmixing

  (Zhe Sun et al. CEJ , 343, 770-788, 2018)

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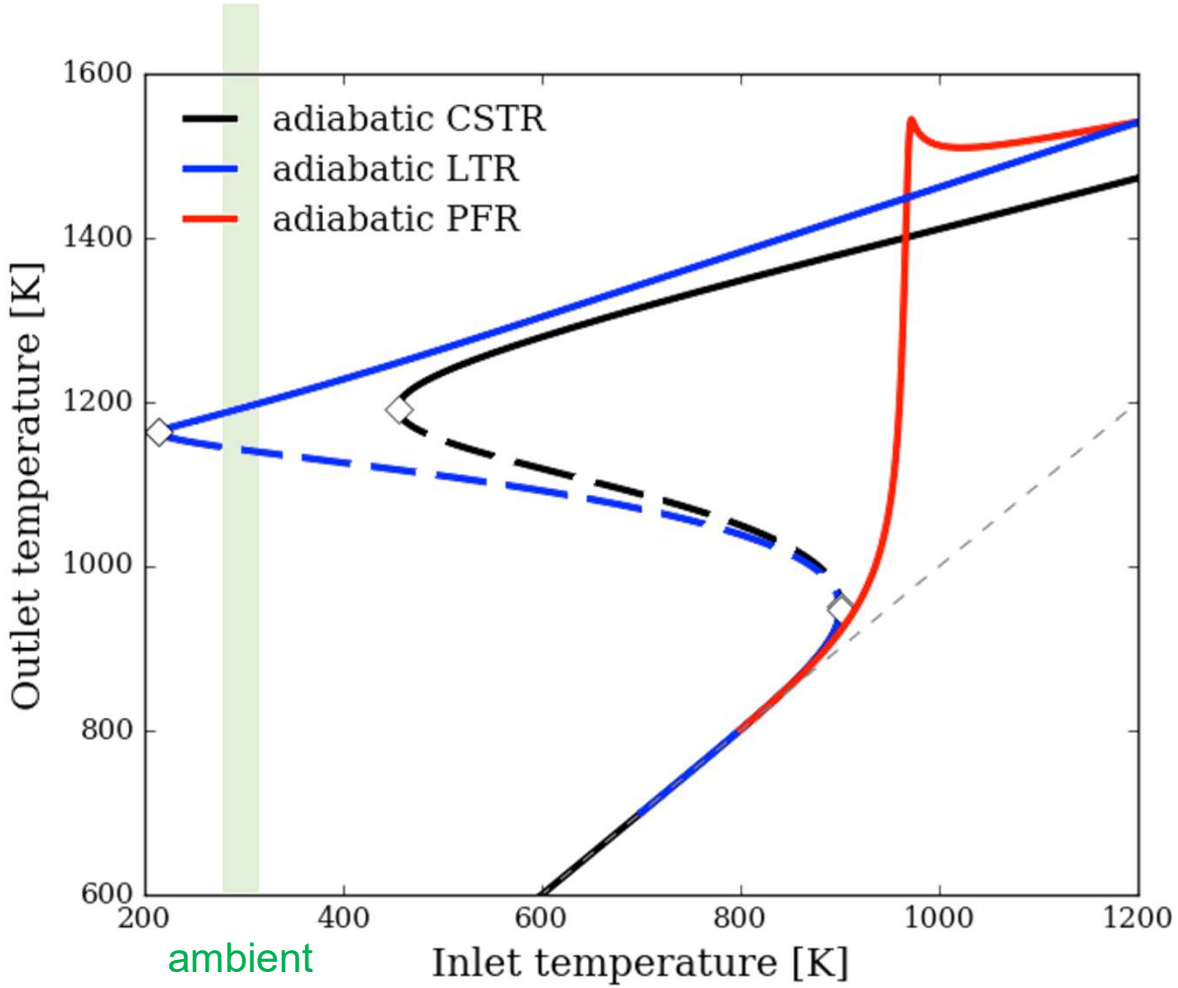
OCM with Sr/La_2O_3 : comparison of reactor types

LTR most promising

– Lowest extinction point (ambient inlet temperature possible)



Operating conditions:
P = 1 bar, $CH_4:O_2$ = 6,
$V/F^0_{V(STP),CH4}$ = 0.01 s,
m_{cat}/V = 1000 kg_{cat}/m^3

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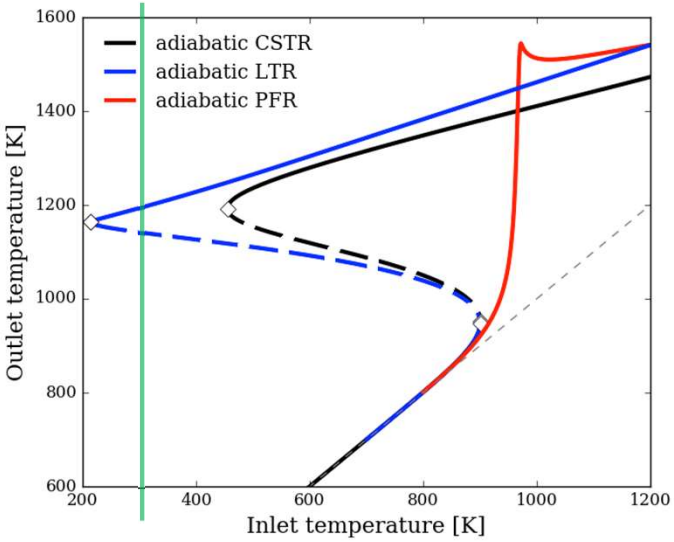
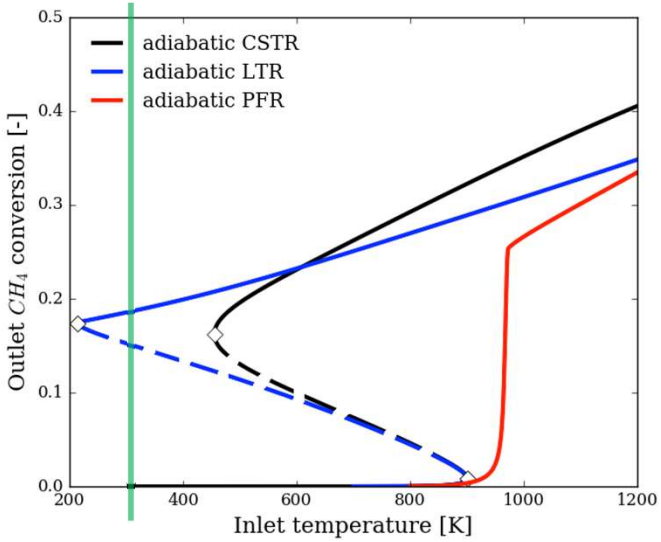
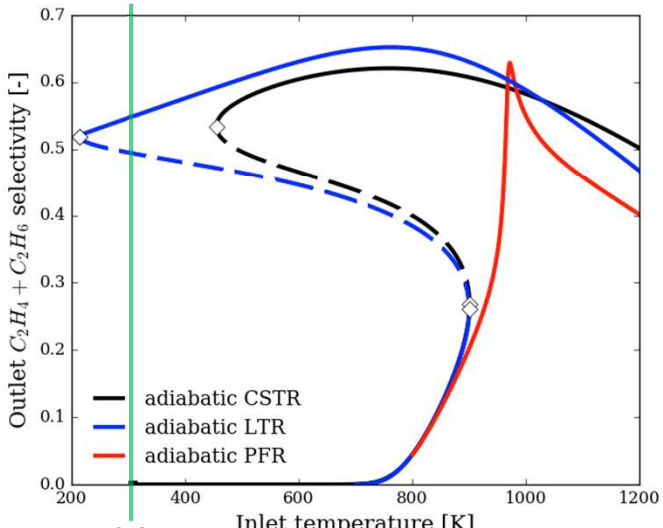
OCM with Sr/La_2O_3 : comparison of reactor types

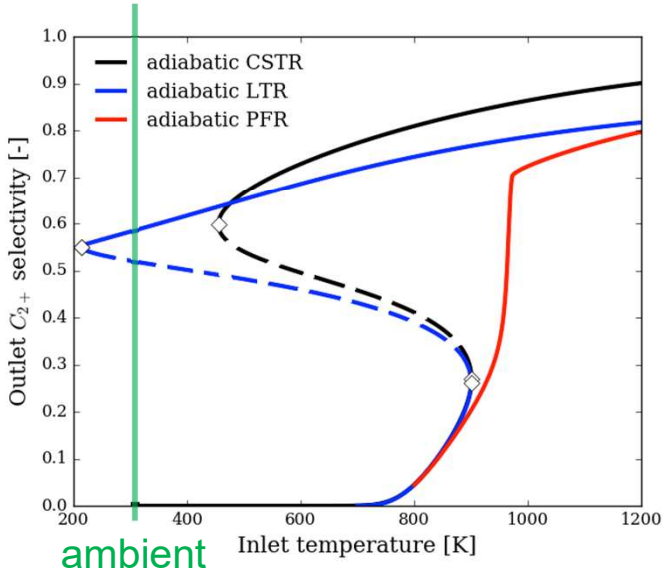
LTR most promising

– Lowest extinction point (ambient inlet temperature possible)









Operating conditions:
P = 1 bar, $CH_4:O_2$ = 6,
$V/F^0_{V(STP),CH4}$ = 0.01 s,
m_{cat}/V = 1000 kg_{cat}/m^3

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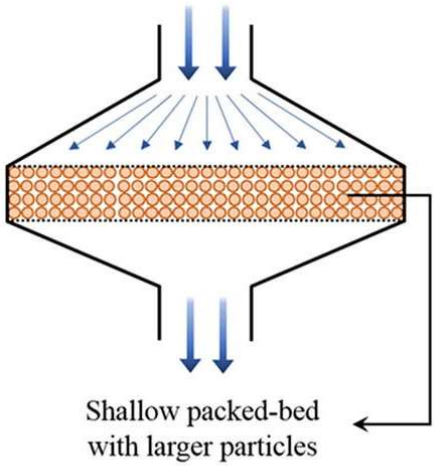
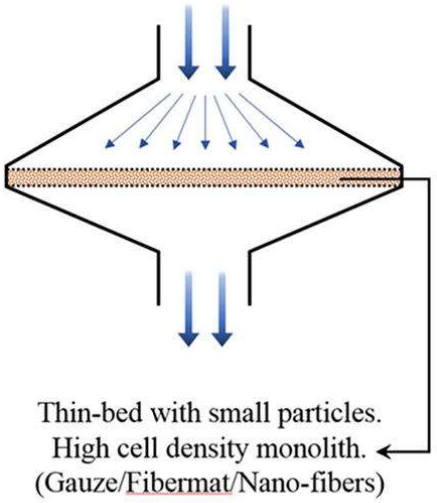
Lumped Thermal Reactors in real life

Key feature: little species but good thermal backmixing

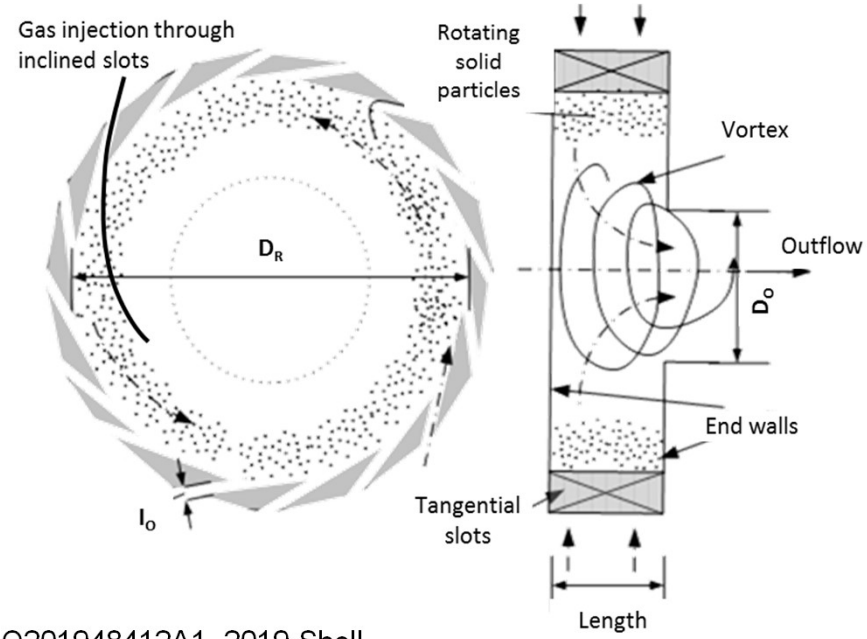
Shallow packed bed



High cell density monoliths



Gas-solid vortex reactor



Balakotaiah, V. et al. Chem. Eng. J. 2019, *374*, 1403–1419

Thin bed cofeed reactors for methane conversion, US4876409A, 1989 ARCO

Process for oxidatively converting methane to higher hydrocarbon products, WO2019048408A1, WO201948412A1, 2019 Shell

Vandewalle, L. A. et al. Chem. Eng. Sci. 2019, *198*, 268-289

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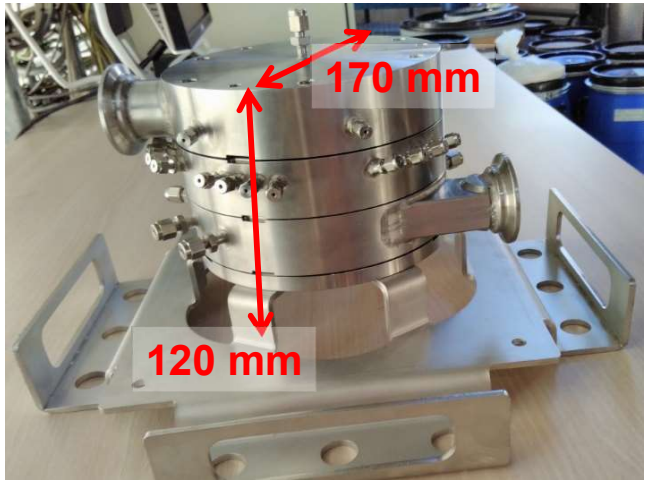
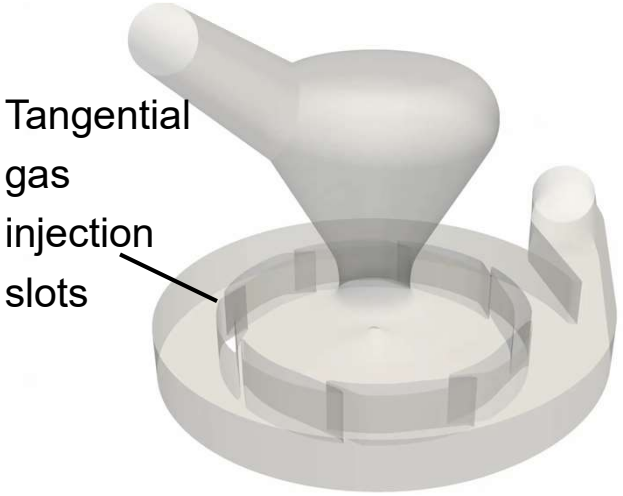
Gas-Solid Vortex Reactor (GSVR)





Rotating bed in
Vortex reactor

- High gas feed flow rates → small gas residence time
- High gas-solid slip velocity → good gas-solid heat and mass transfer
- Good thermal backmixing

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PROCESS INTENSIFICATION

M .N. Pantzali et al., AIChE J., (61), 2015, 4114-4125.

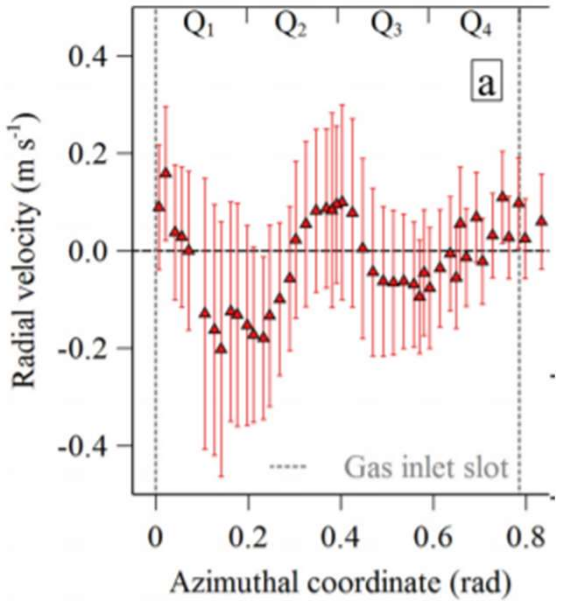
Thermal backmixing

Solid-phase energy equation:

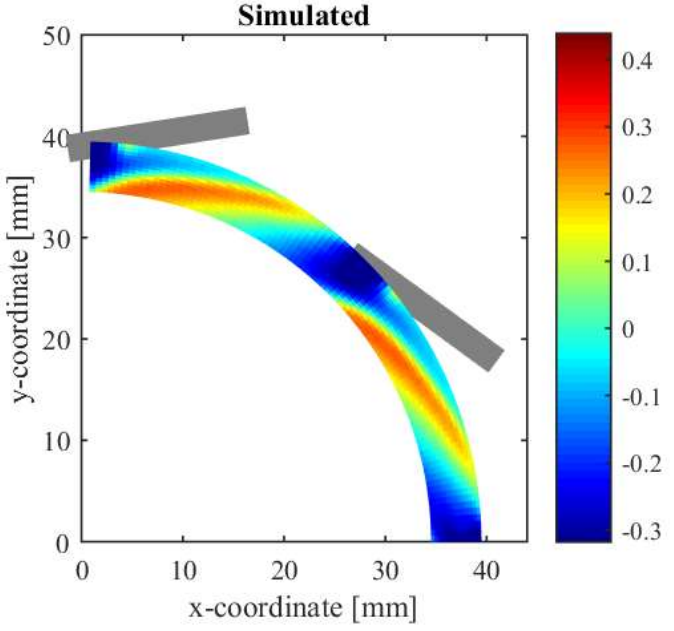
$$\frac{\partial(\varepsilon_s \rho_s h_s)}{\partial t} + \boldsymbol{\vec{\nabla} \cdot (\varepsilon_s \rho_s \vec{u}_s h_s)} = \vec{\nabla} \cdot \left(\varepsilon_s \rho_s \alpha_{\text{eff},s} \vec{\nabla} h_s\right) + \varepsilon_s \rho_s (\vec{u}_s \cdot \vec{g}) - Q_{sg} + \varepsilon_s Q_{r,s}$$

energy transport by moving particles

For GSVR: importance of radial motion of particles!!



Gonzalez-Quiroga et al., Powder Technology (2019)



Radial velocity [m/s]

Vandewalle et al., IECR (2019)

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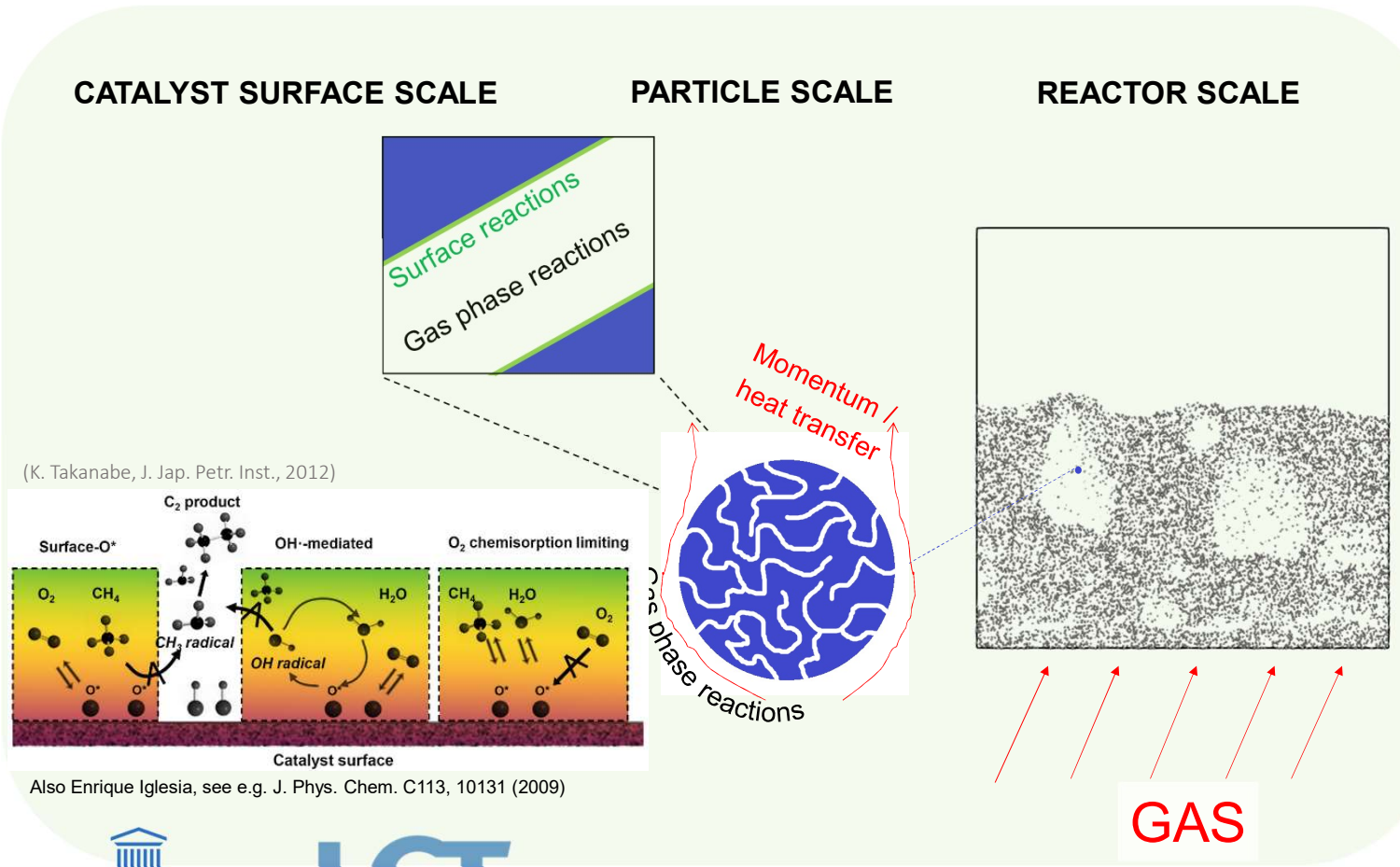
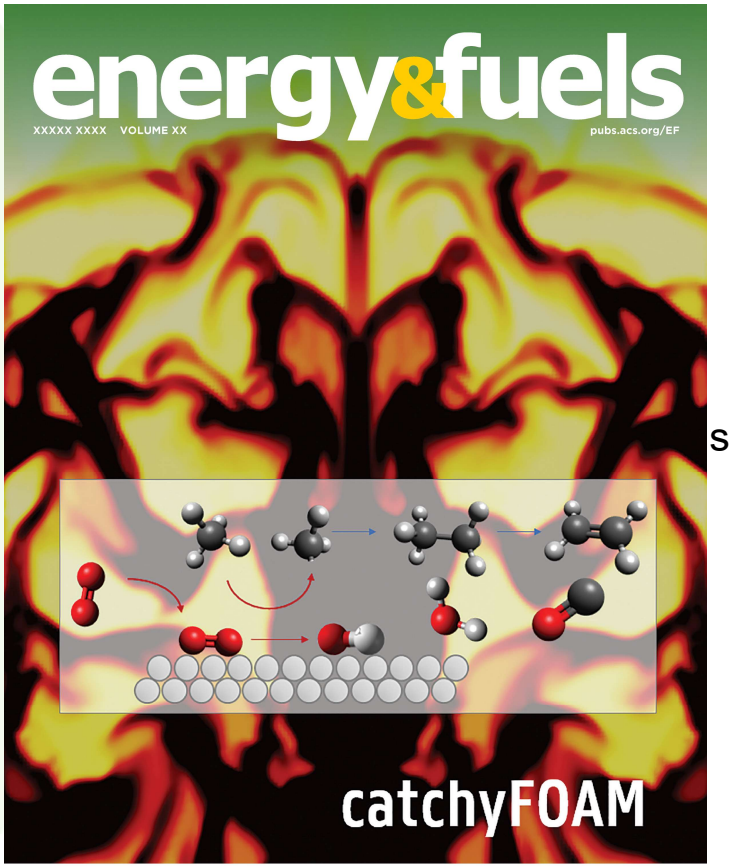
Computational Fluid Dynamics Euler-Euler code

CATalyticCHemistrYFOAM



Also Enrique Iglesia, see e.g. J. Phys. Chem. C113, 10131 (2009)



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Most Trusted. Most Cited. Most Read.
www.acs.org

Vandewalle L.et al, Energy & Fuels (2020)

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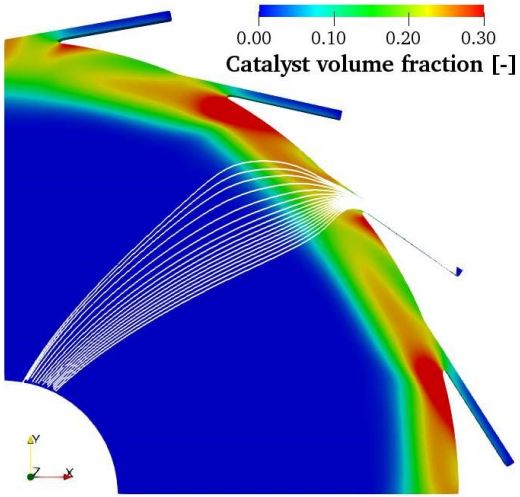
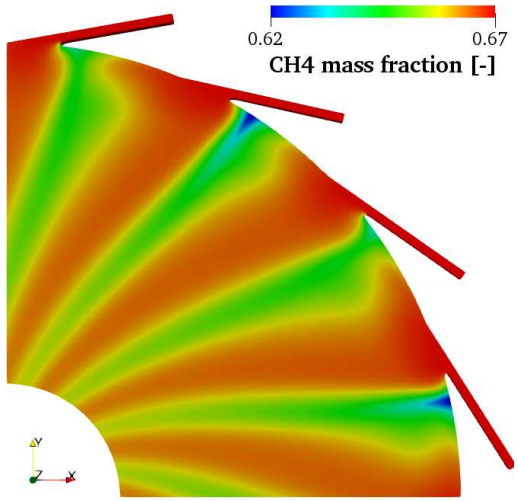
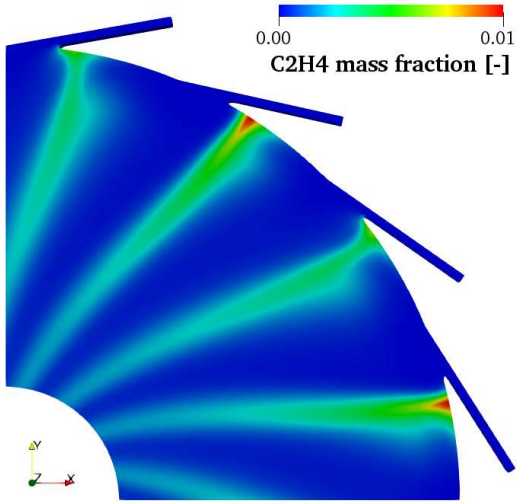
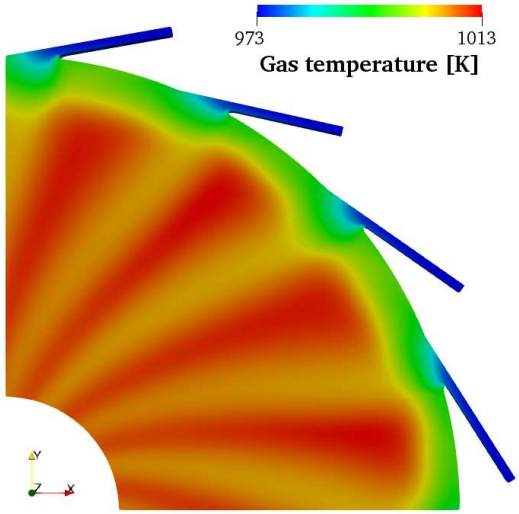
https://github.com/lavdwall/catchyFOAM

Adiabatic GSVR for OCM: T_{in} = 973 K









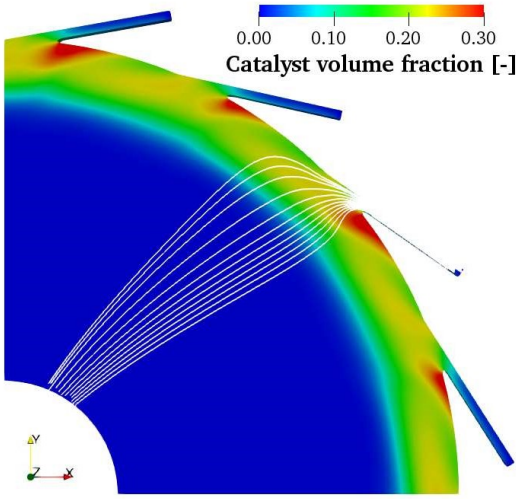
CH_4 conversion ~ 1.56 %
Local higher conversion in 'dead' zone between the slots

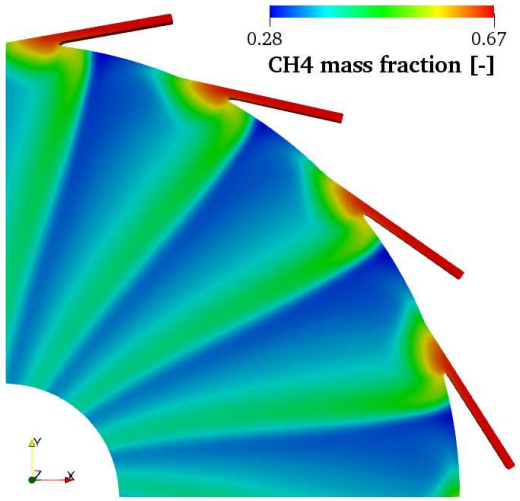
C_2 selectivity ~ 67 %
9.4 % C_2H_4 + 57.6 % C_2H_6

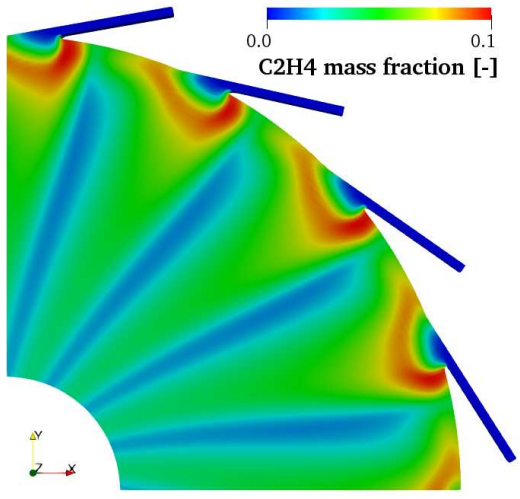
Temperature rise from inlet to outlet ~ 35 K

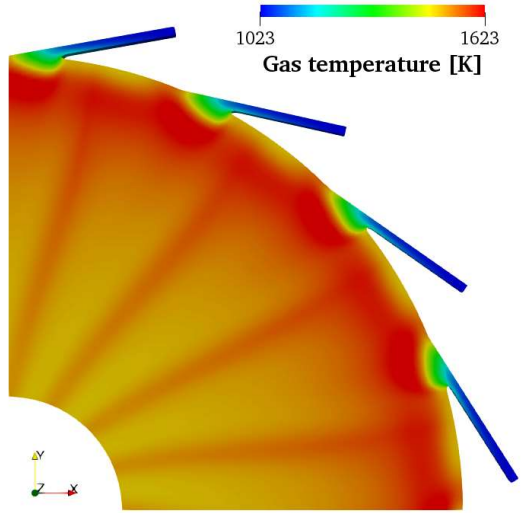
2D GSVR, 16 slots, D_R = 8 cm, $F_{V,g,inj}$ = 15 Nm³/hr, CH_4:O_2 = 4:1, P = 2 bar, T_{in} = 973 K (adiabatic)
12g catalyst (4%Sr-40%La/SiC, porosity 0.27, A/V = 18.29 10^6 m²/m³, d_s = 500 µm, ρ = 2950 kg/m³)

Adiabatic GSVR for OCM : T_{in} = 1023 K









IGNITION

CH_4 conversion ~ 44 %

C_2 selectivity ~ 79 %

14.5 % C_2H_4 + 0.3 % C_2H_6

+ 64.3 % C_2H_2

Temperature rise from inlet to outlet ~ 475 K

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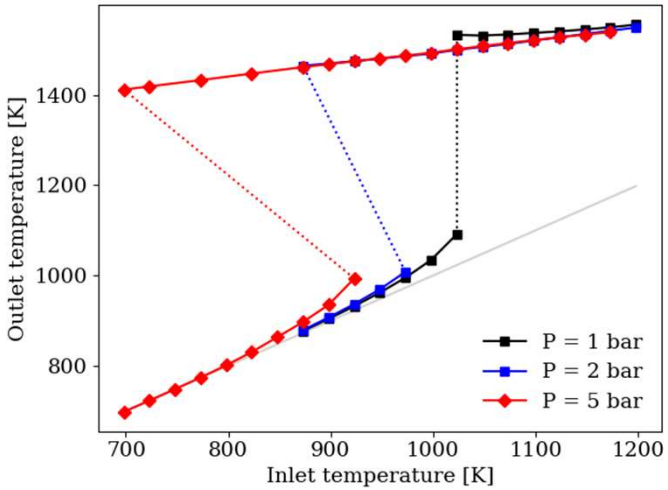
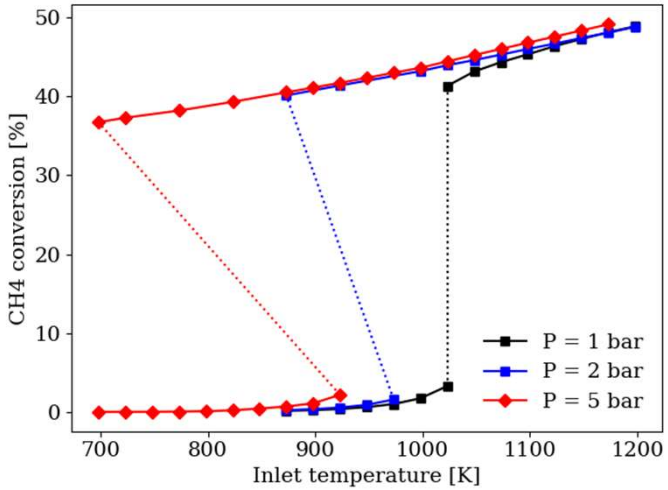
2D GSVR, 16 slots, D_R = 8 cm, $F_{V,g,inj}$ = 15 Nm³/hr, CH_4:O_2 = 4:1, P = 2 bar, T_{in} = 1023 K (adiabatic)

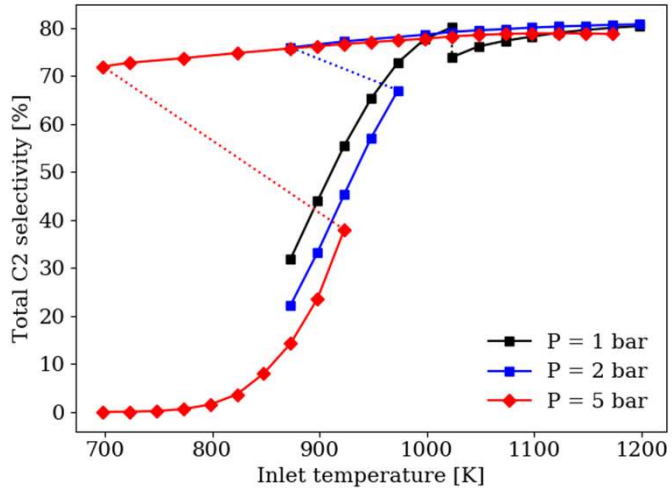
12g catalyst (4%Sr-40%La/SiC, porosity 0.27, A/V = 18.29 10^6 m²/m³, d_s = 500 µm, ρ = 2950 kg/m³)

Effect of pressure on ignition/extinction

Wider range of steady-state multiplicity at higher pressure







2D GSVR, 16 slots, D_R = 8 cm, $F_{V,g,inj}$ = 15 Nm³/hr, CH_4:O_2 = 4:1, P = 2 – 5 bar, T_{in} = 673 - 1198 K (adiabatic)
12g catalyst (4%Sr-40%La/SiC, porosity 0.27, A/V = 18.29 10^6 m²/m³, d_s = 500 µm, ρ = 2950 kg/m³)

Outline

- Introduction
- Super Dry Reforming
- Oxidative coupling
- Conclusions and Perspectives

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Conclusions

- Combine !
  - computational and experimental techniques
  - material and process development
  - chemical engineering and catalysis
    - Transport phenomena on lab and industrial scale
    - Autothermal adiabatic operation of exothermic reactions

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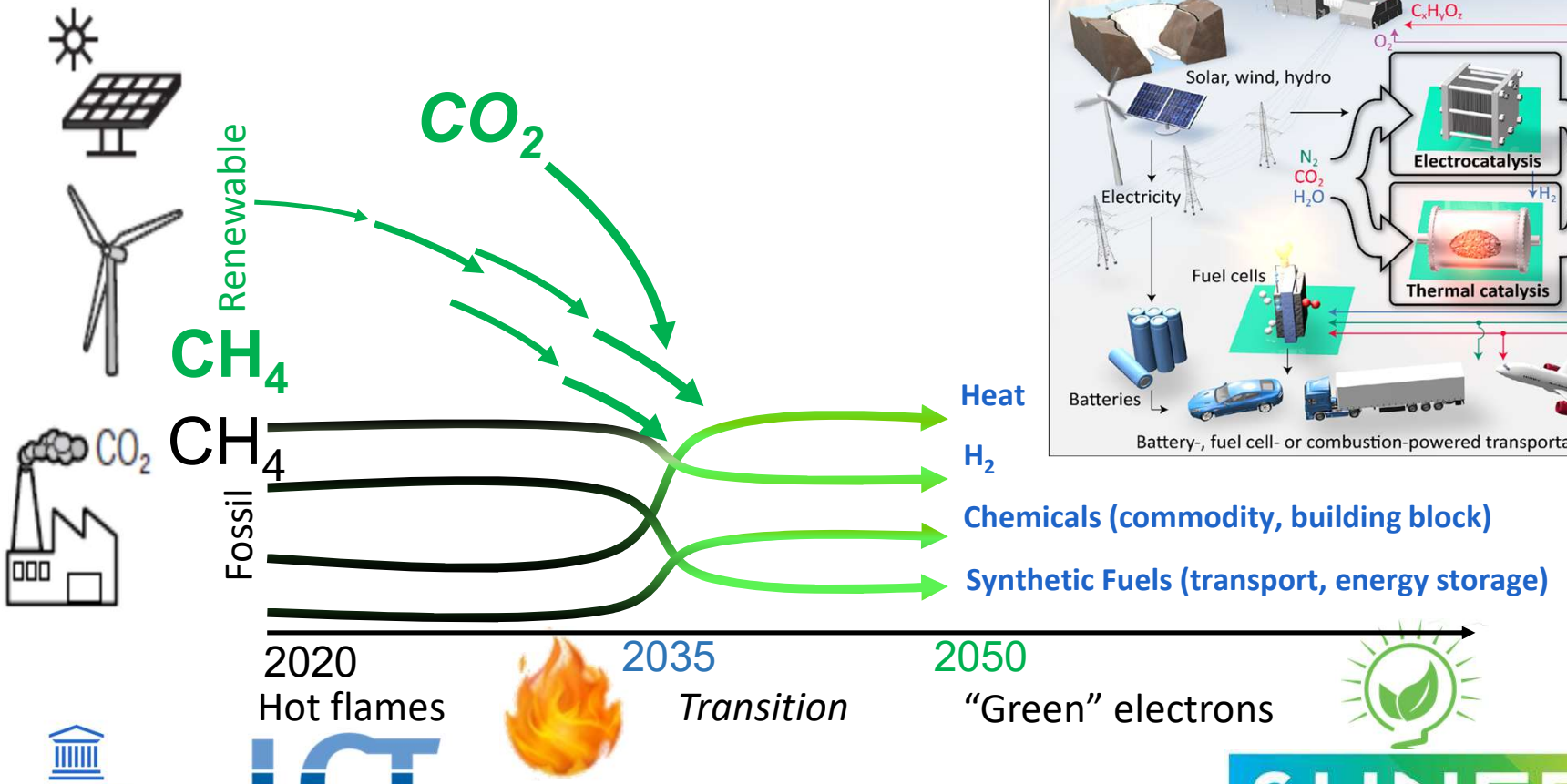
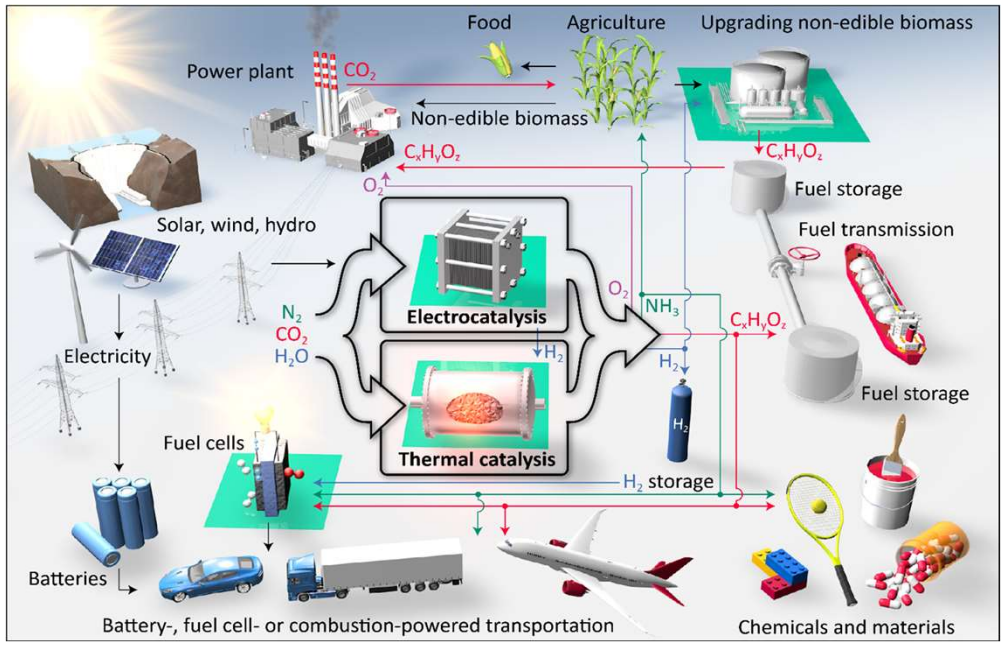
Perspectives: Chemical engineering and catalysis





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K.M. Van Geem et al., Science, 364 (6442) 6442 734 (2019)

SUNERGY

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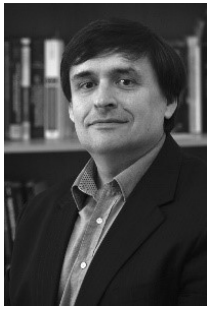











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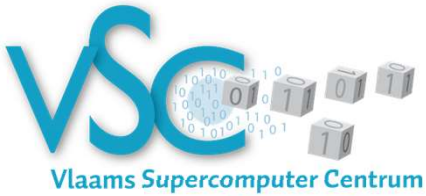

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