

Renewable Energy

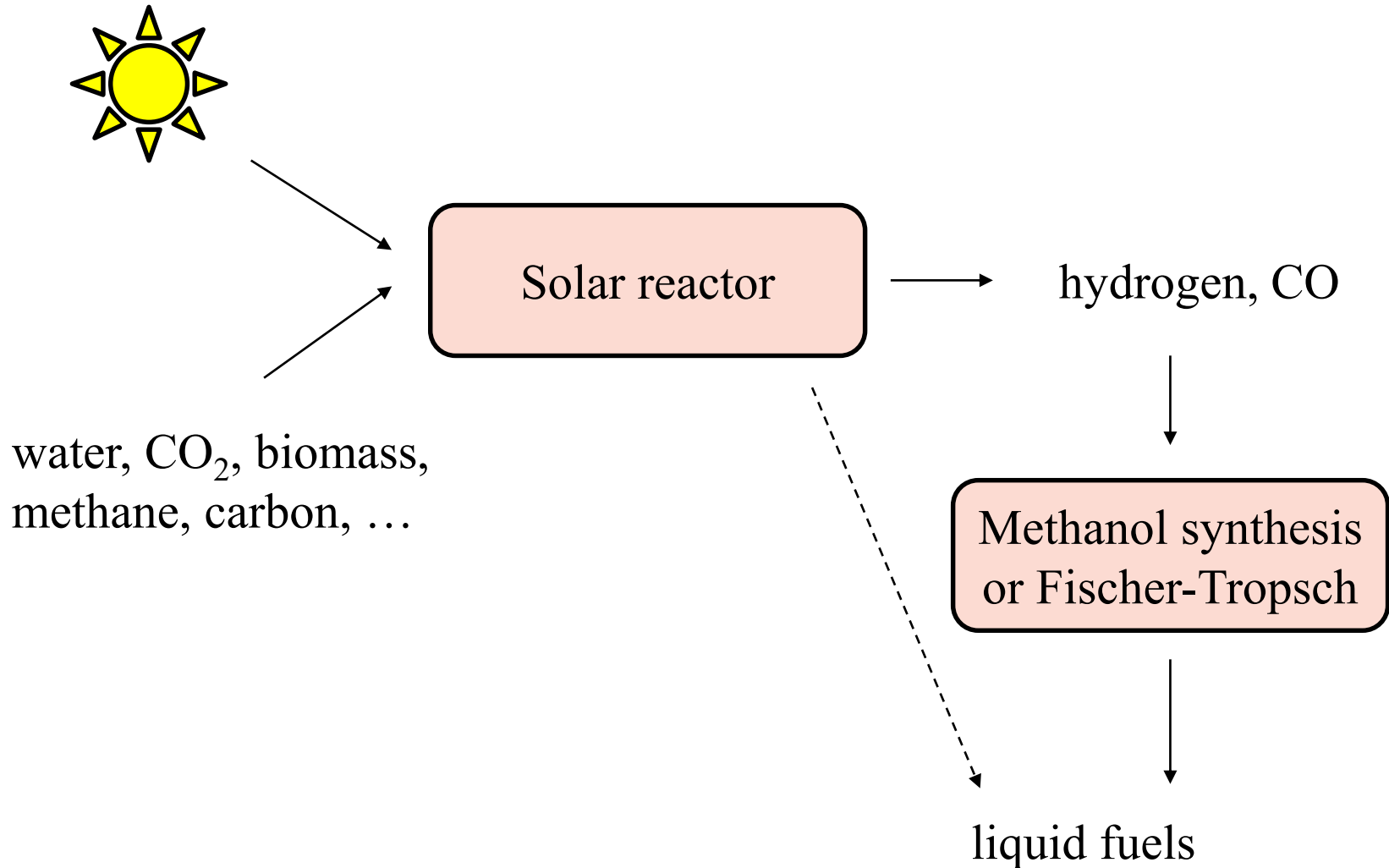
- Outline:
 - Conversion pathways solar-to-fuel
 - Hybrid pathways
 - Solar thermochemistry
 - Photochemistry

Learning outcomes of today's lecture

- Solar fuels:
 - How can solar energy be converted into fuels?
 - What is a hybrid pathway?
 - Why using fossil fuels together with solar energy?
 - What is solar thermochemistry and how can it be used for solar fuel processing?
 - Why is solar water-splitting via multi-step water splitting cycles preferred compared to direct thermolysis?
 - What is photoelectrochemistry and how can it be used for solar fuel processing?
 - What other chemical commodities or materials can be processed using solar energy?

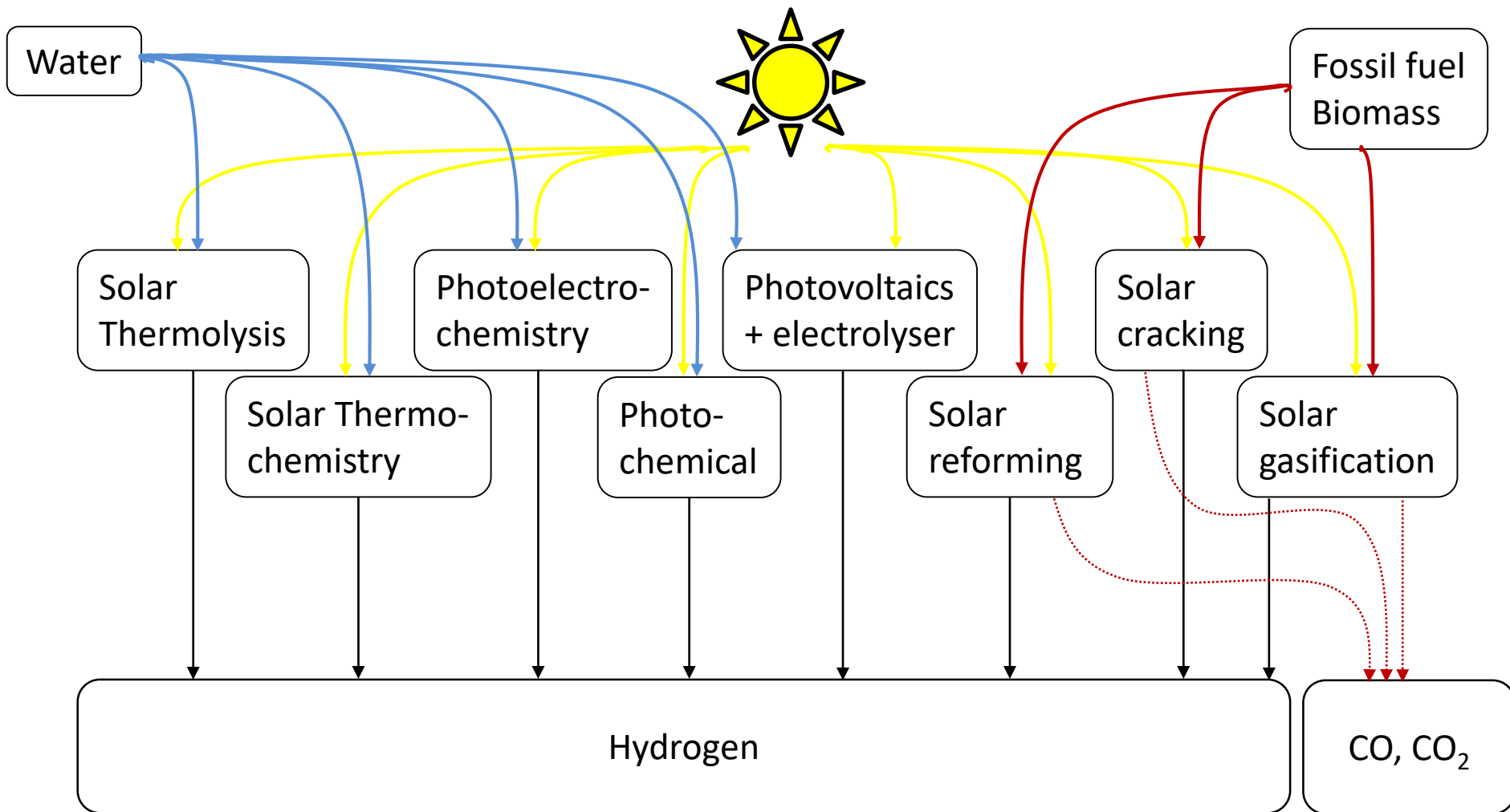
Conversion pathways

- Solar to fuels:



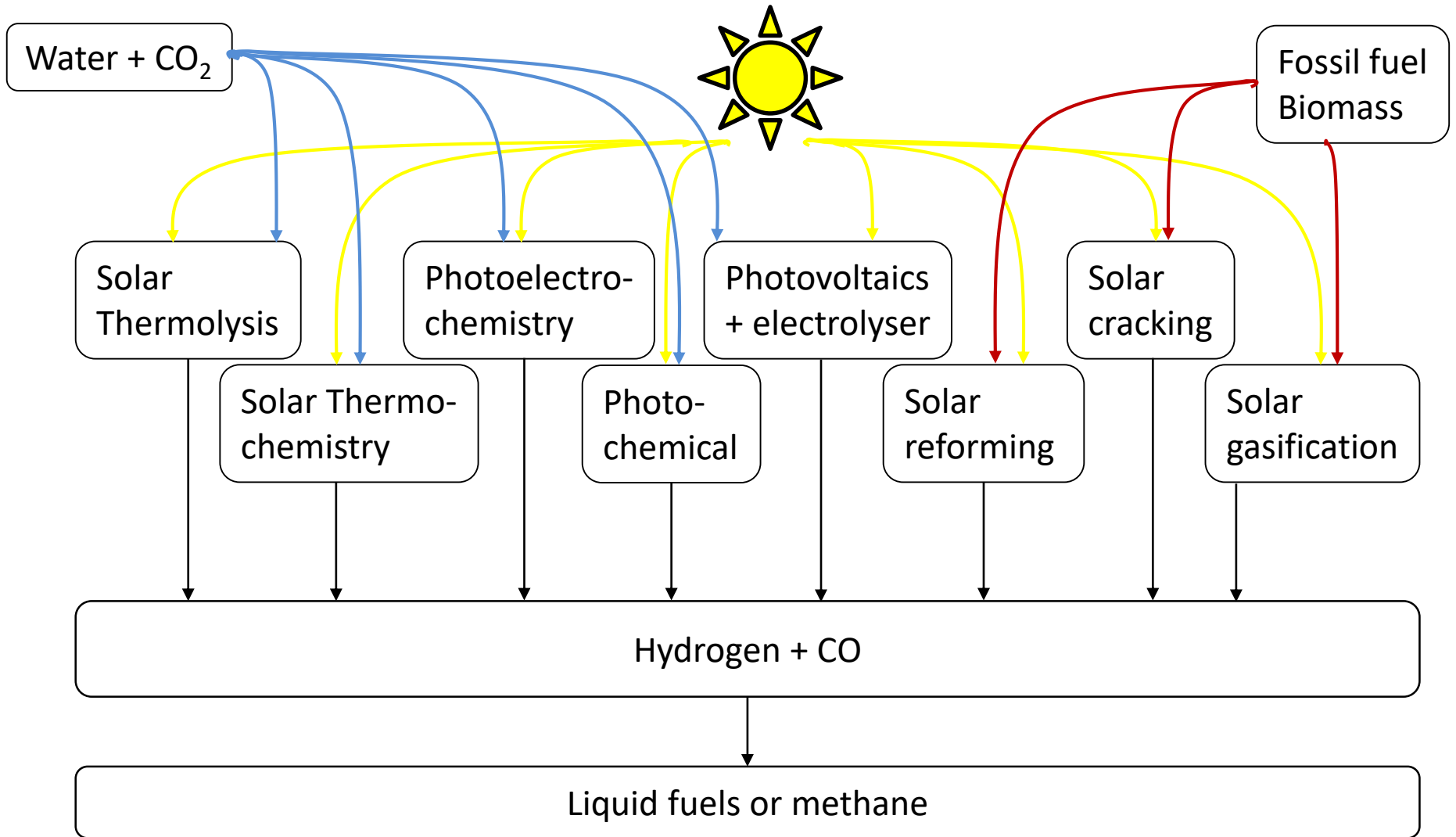
Conversion pathways

- Solar to hydrogen:



Conversion pathways

- Solar to synthesis gas (H_2+CO):

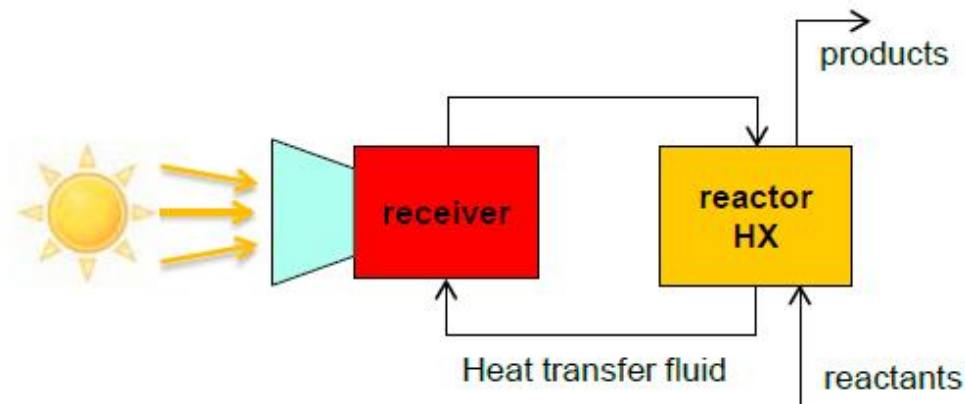


Conversion pathways

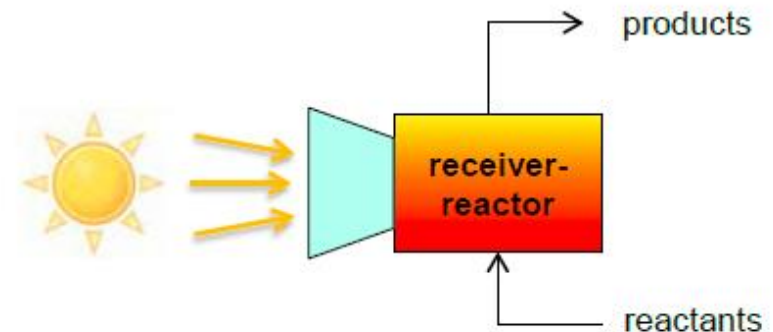
- General considerations:
 - What solar radiation concentration technology can be used (if needed)?
 - What solar reactor can be used and what are the requirements?
 - How can the sun be coupled into the process?
 - What can the reactor look like?
- Reactor concepts:

Decoupled receiver+reactor

Possibly with high-temperature storage



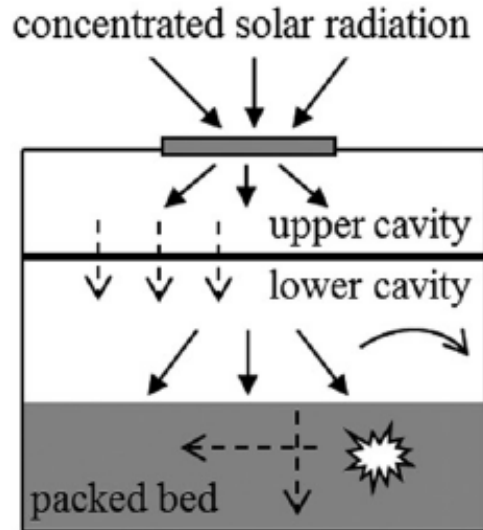
Coupled receiver-reactor



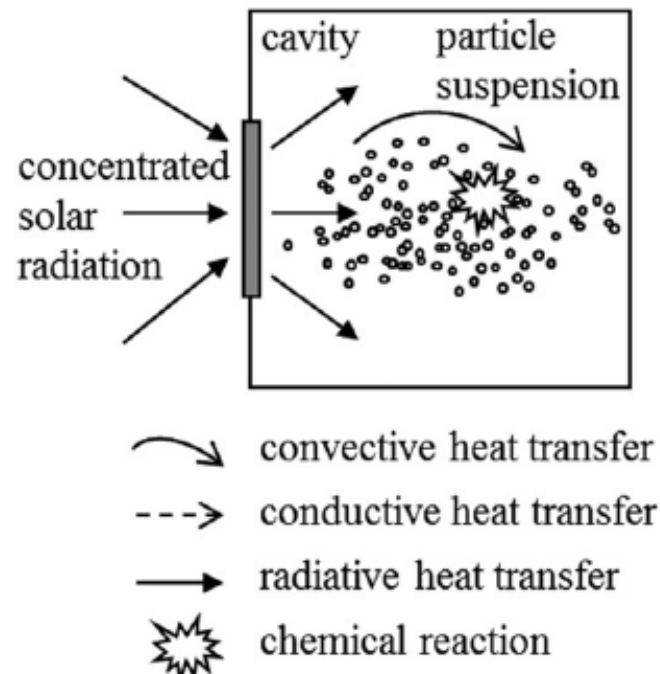
Conversion pathways

- Reactor concepts:

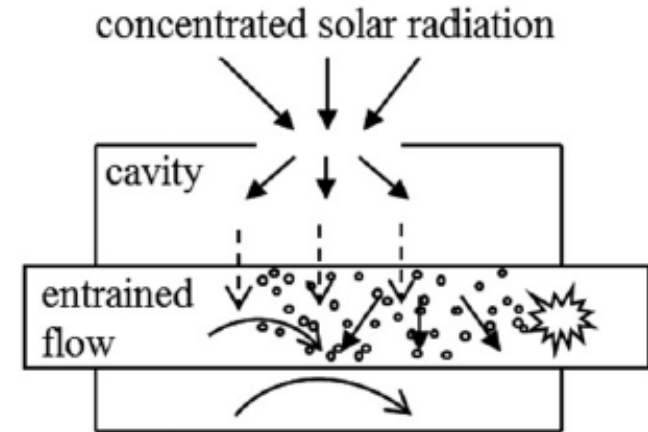
indirectly irradiated
packed-bed



directly irradiated
vortex-flow



indirectly irradiated
entrained flow

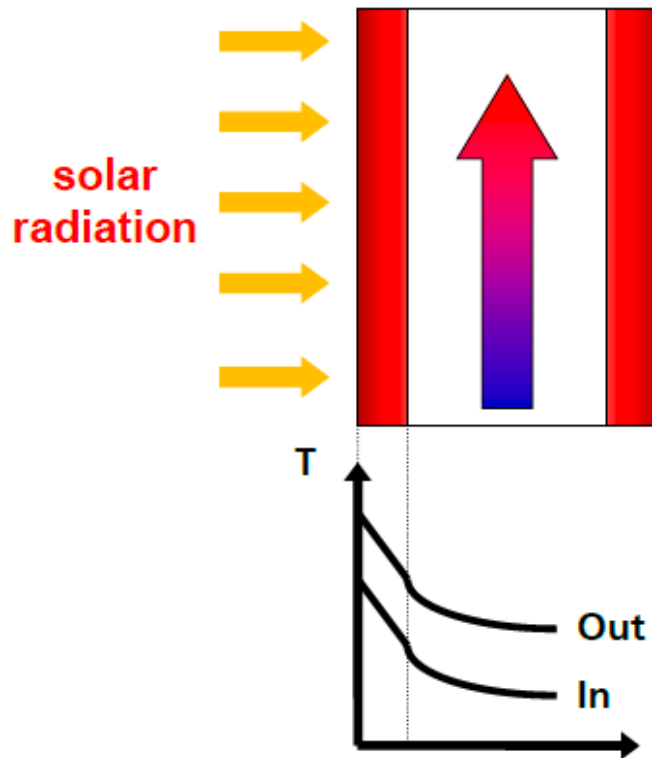


- convective heat transfer
- conductive heat transfer
- radiative heat transfer
- chemical reaction

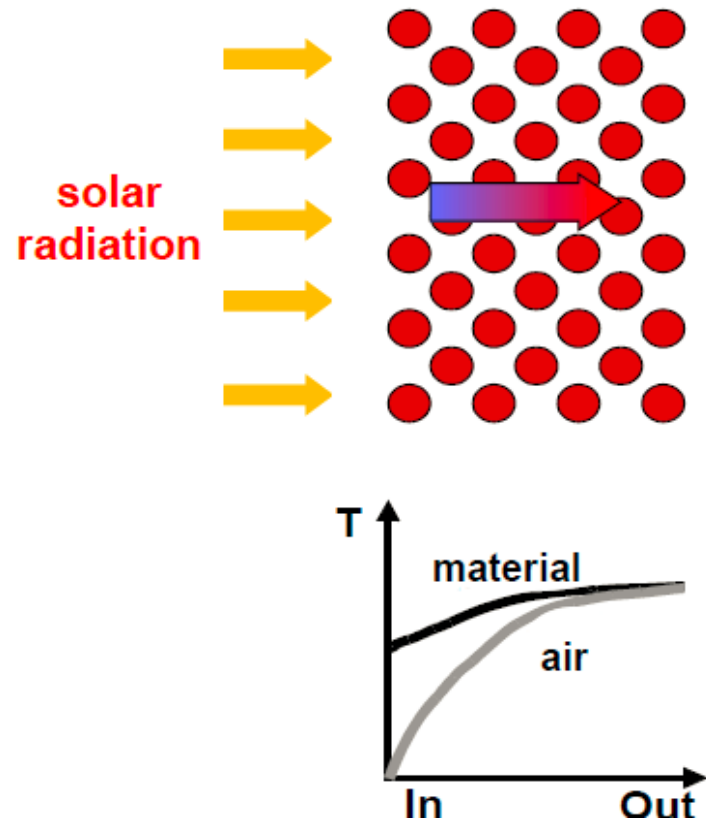
Conversion pathways

- Reactor concepts:

Tube receiver



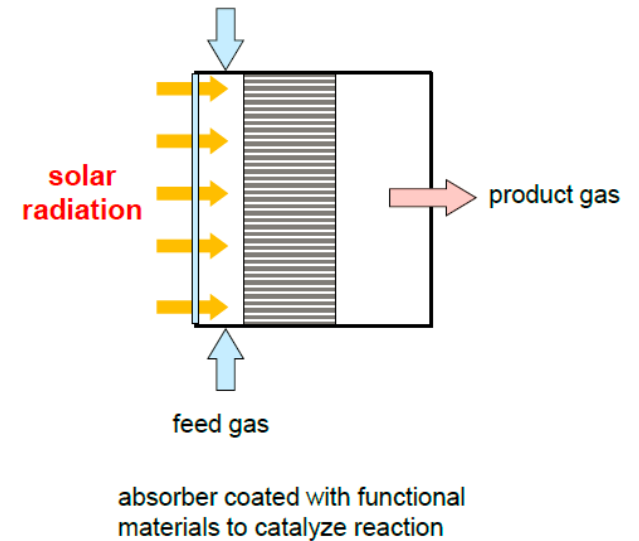
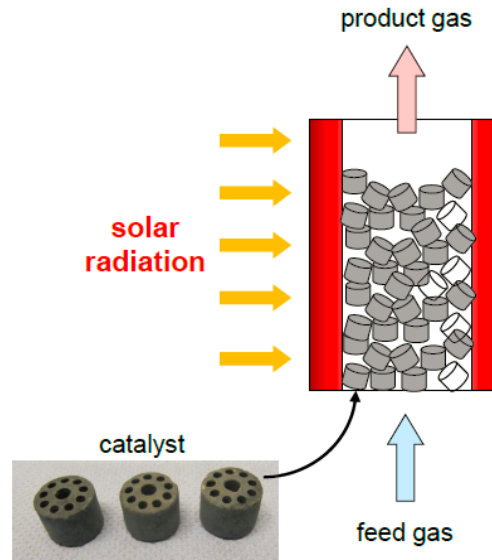
Volumetric receiver



- Also: open versus closed systems

Conversion pathways

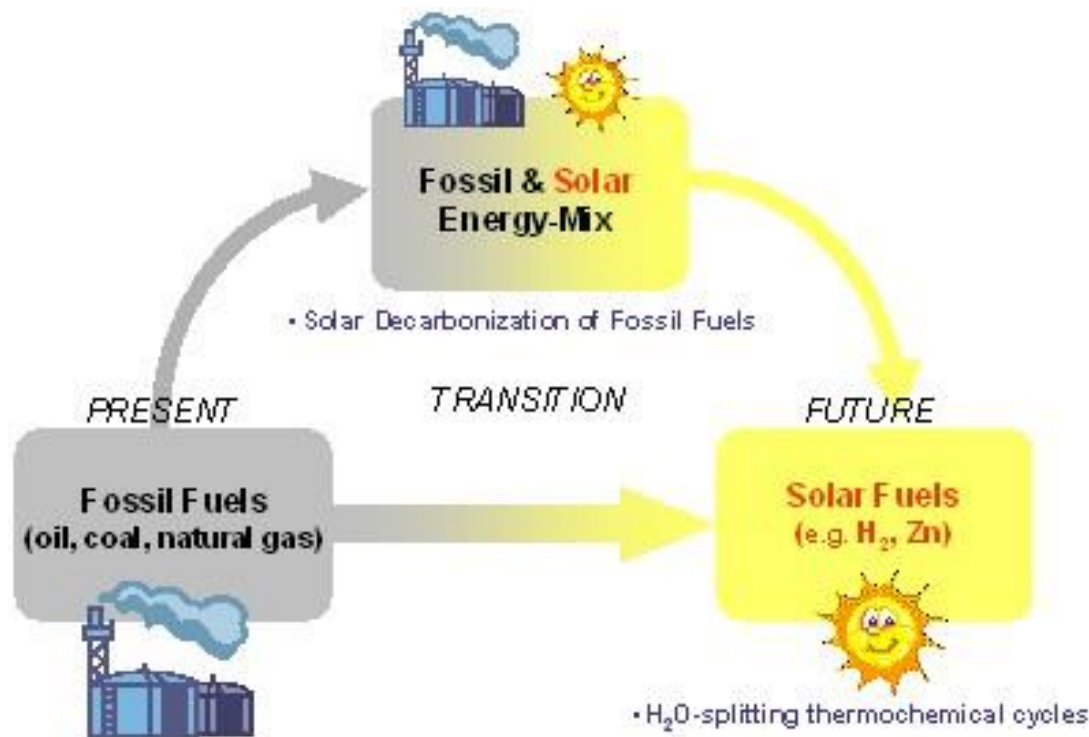
- Reactor concepts:
 - Stationary



- Moving:
 - Fluidized particle bed
 - Falling particle film
 - Rotating kiln
 - Moving particle bed

Hybrid solar conversion

- In the transition to a renewable future, hybrid pathways using fossil fuels exclusively as chemical source for the fuel production and solar energy as the process heat

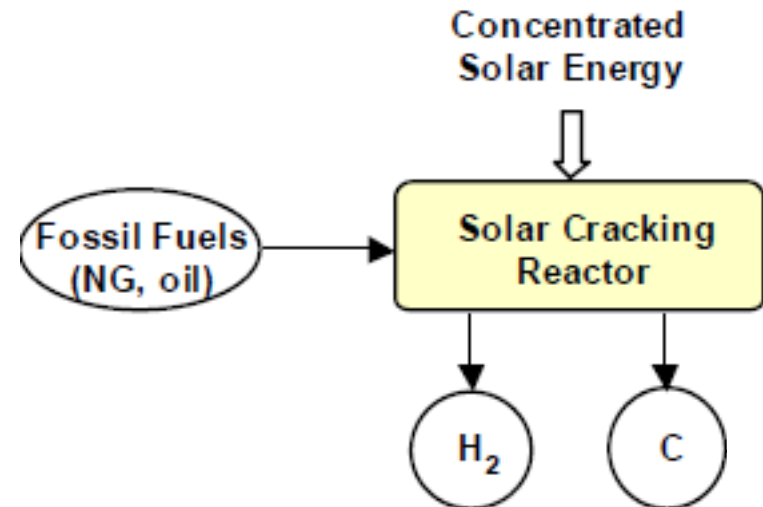


Hybrid solar conversion

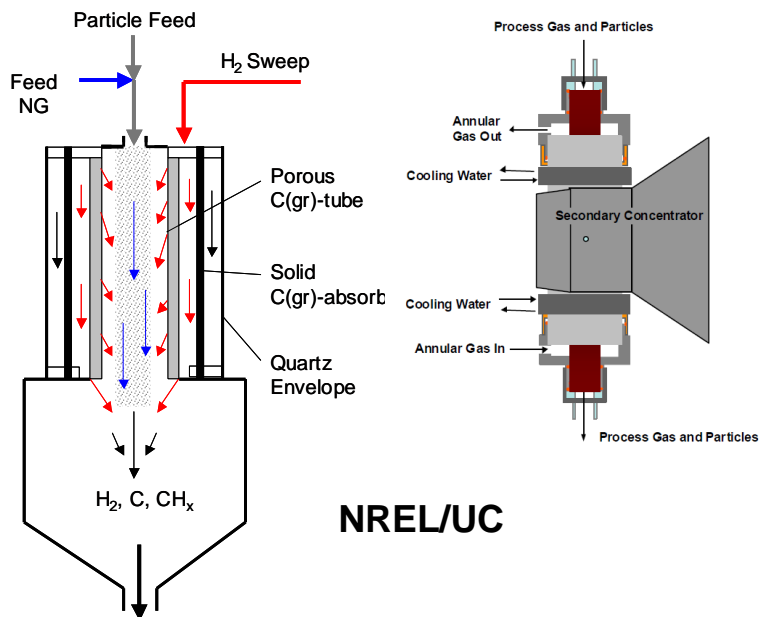
- **Thermal cracking:** complex organic molecules such as heavy hydrocarbons are broken down into simpler molecules such as light hydrocarbons, by the breaking of carbon-carbon bonds in the precursors at high temperatures and by using catalysts



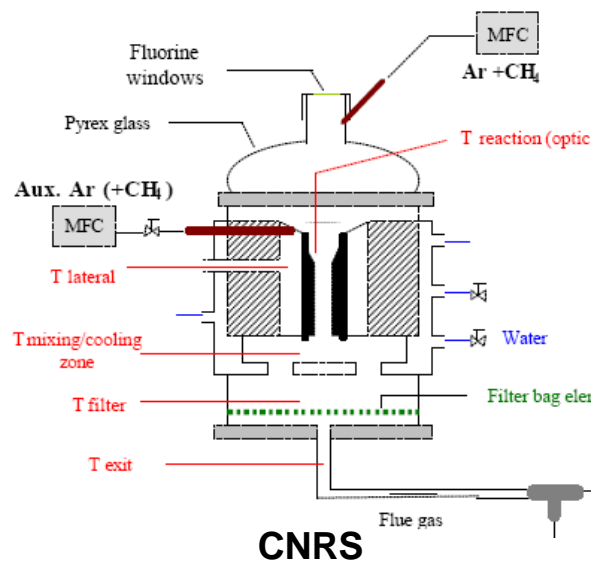
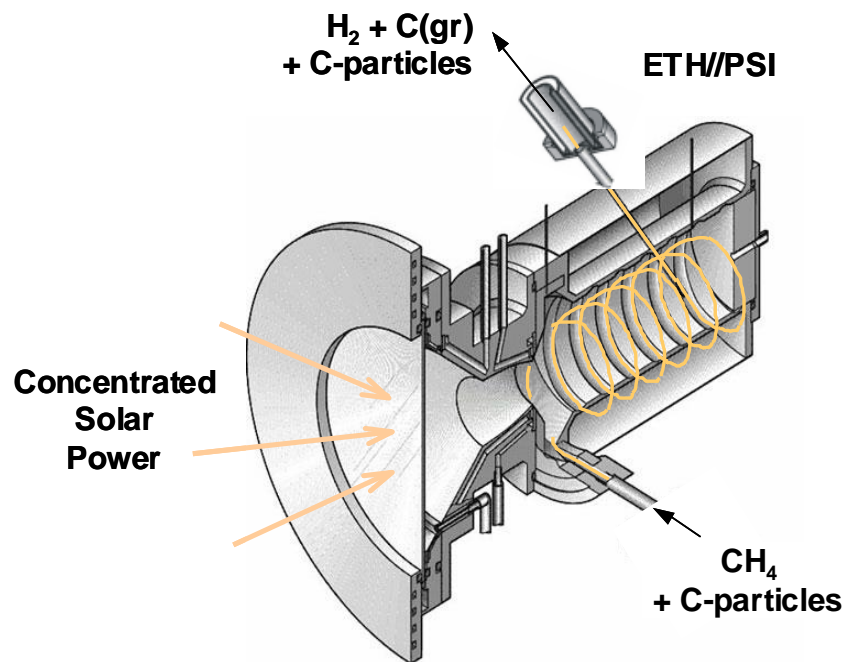
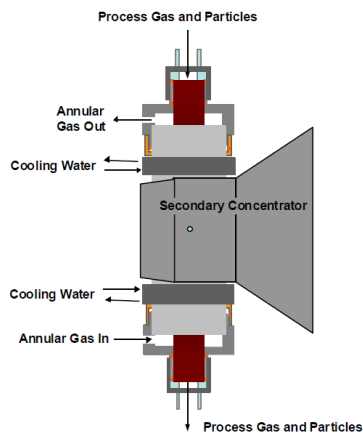
- General:



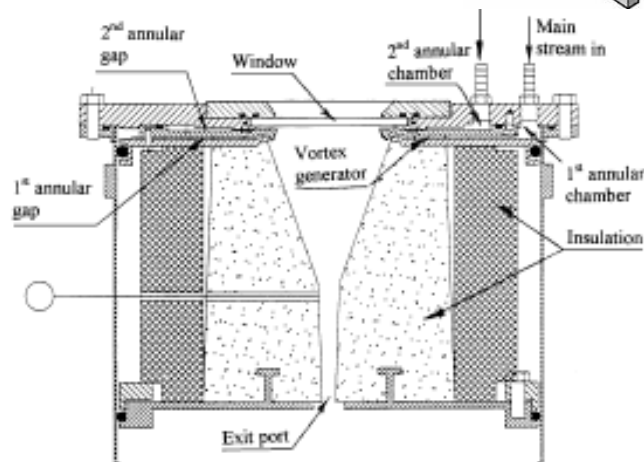
Solar Cracking of NG



NREL/UC



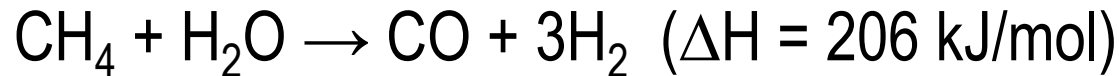
CNRS



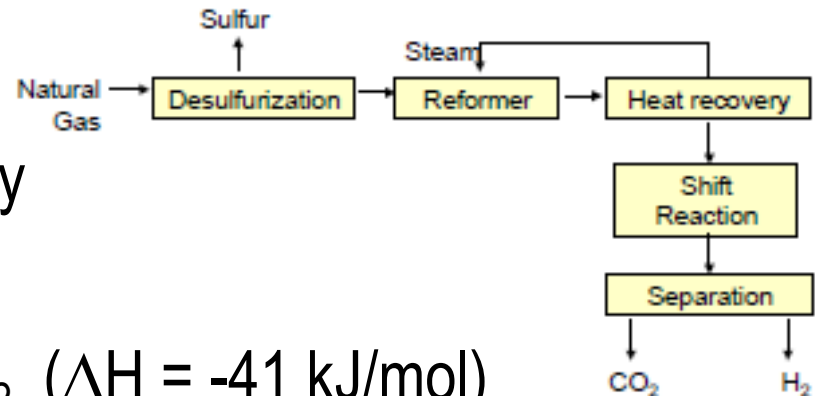
WIS

Hybrid solar conversion

- **Steam reforming:** uses light hydrocarbon feedstock, usually methane, reacts it at elevated temperatures with steam and catalytically converts the feed into hydrogen

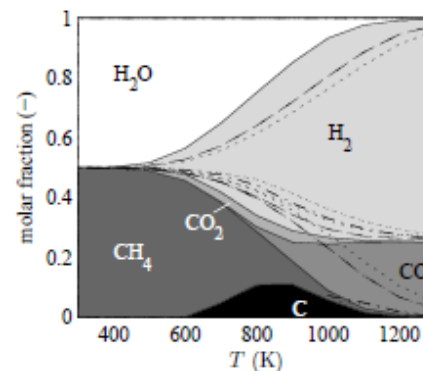
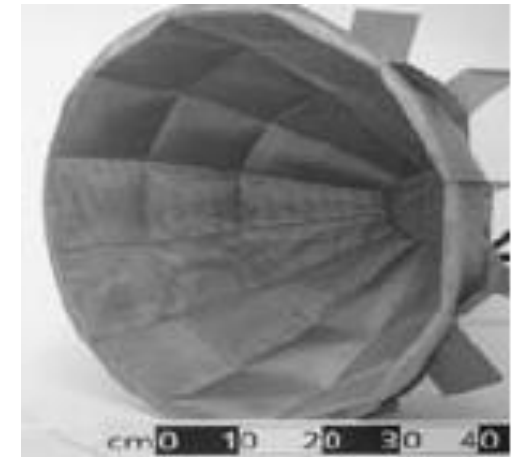
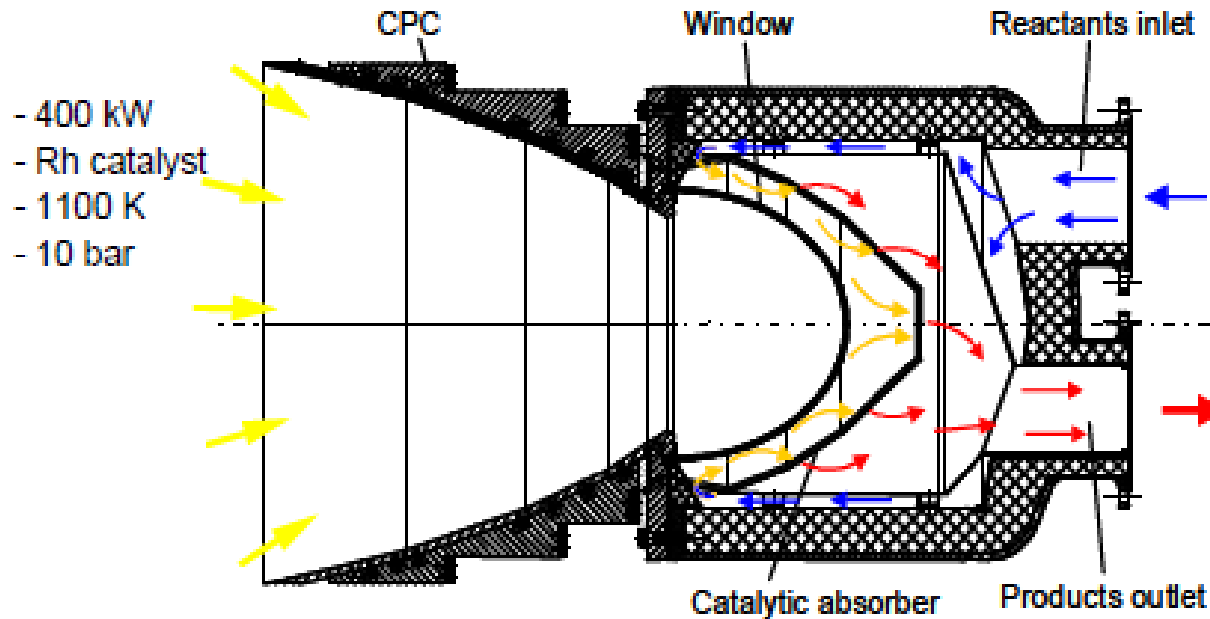


- Operates around 700 – 925 °C
- Can achieve 65 – 75% efficiency



Hybrid solar conversion

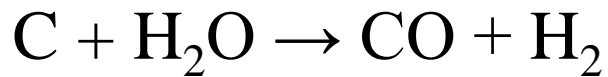
- Solar reactors developed for steam reforming
Solar gasification of methane ($\text{CH}_4 + \text{H}_2\text{O} \rightarrow \text{CO} + 3\text{H}_2$), DLR
SOLREF project



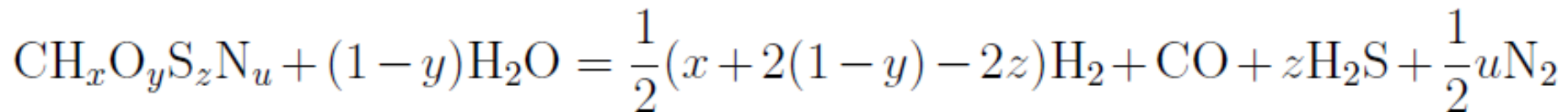
Hybrid solar conversion

- **Gasification:** uses carbonaceous materials, reacts it at high temperatures (>700 °C), without combustion, with a controlled amount of steam, oxygen, and/or CO_2 . Results in CO, H_2 , and CO_2 .

- E.g. for coal, or C-sources

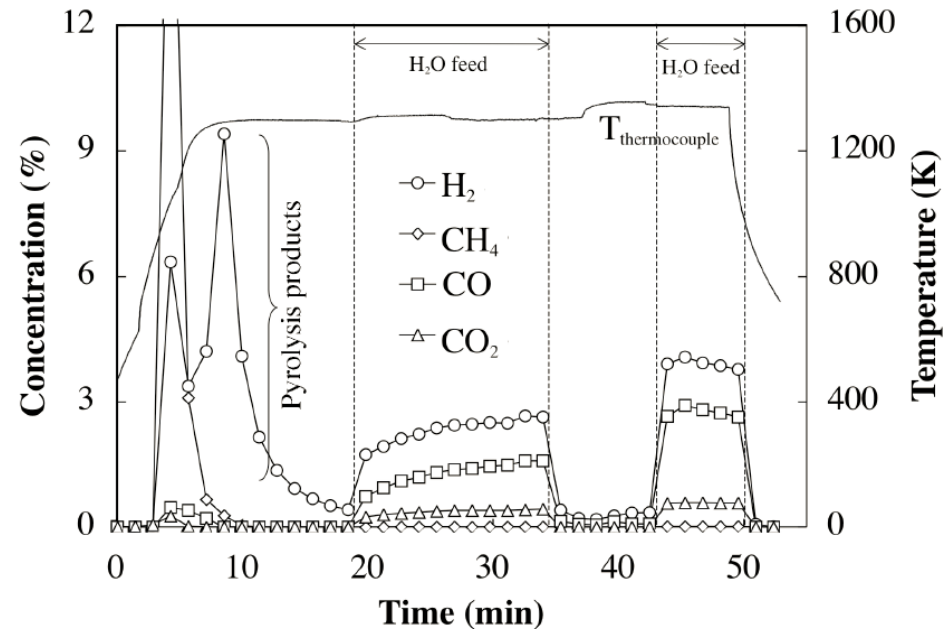


- More realistic (especially for biomass, or C-waste):



Hybrid solar conversion

- Consists of (sequential or simultaneous):
 - Dehydration
 - Pyrolysis (thermal decomposition in the absence of O_2 , devolatilization)
 - Gasification (heterogeneous gas-solid reaction of pyrolysis residue with reactive gas)
 - Combustion
 - Water-gas-shift

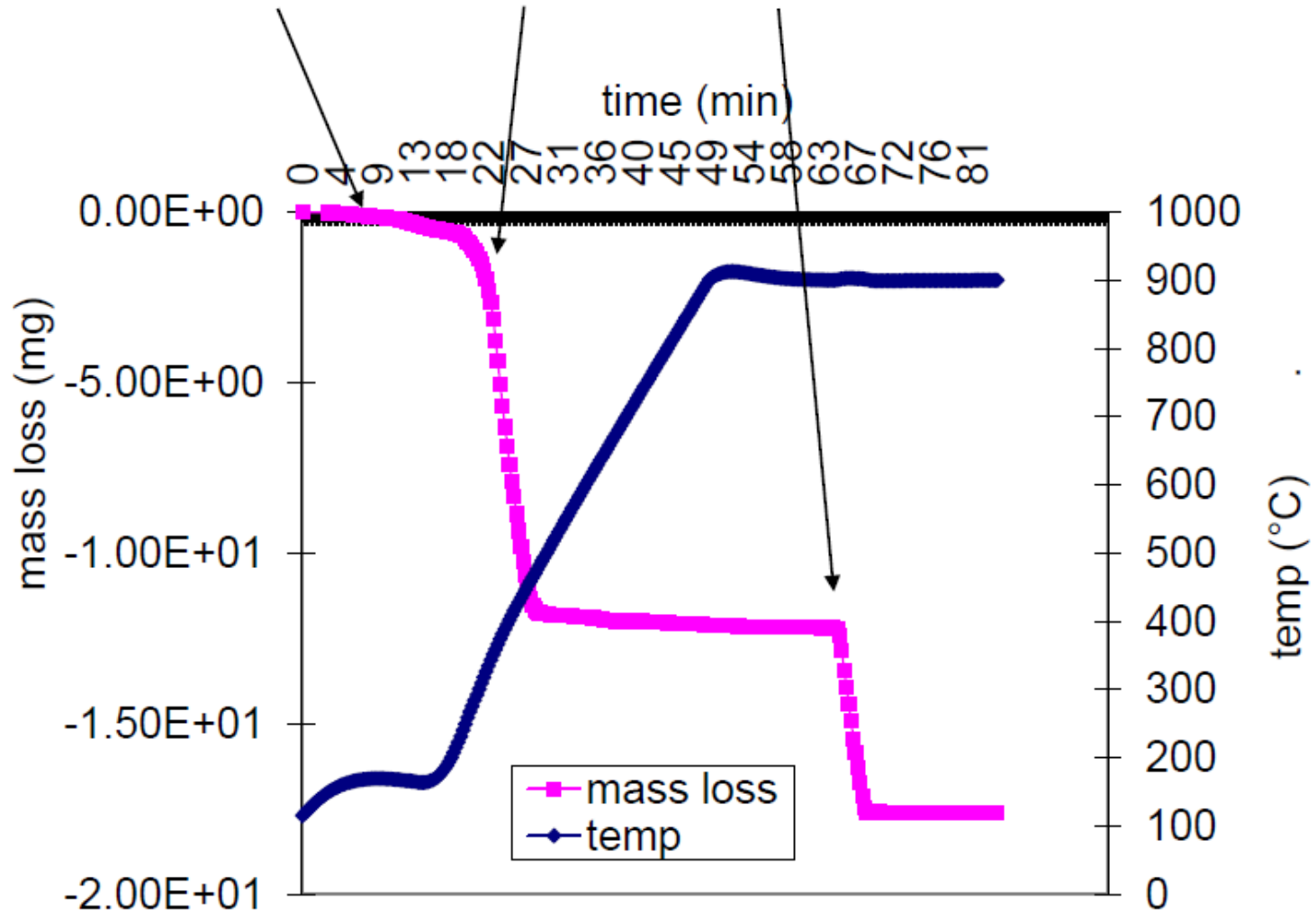


Trommer, Diss ETH, 2006.

Hybrid solar conversion

- Gasification (thermogravimetric experiment):

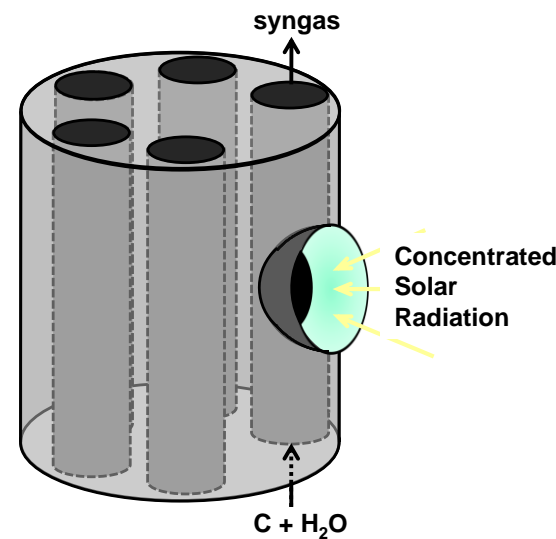
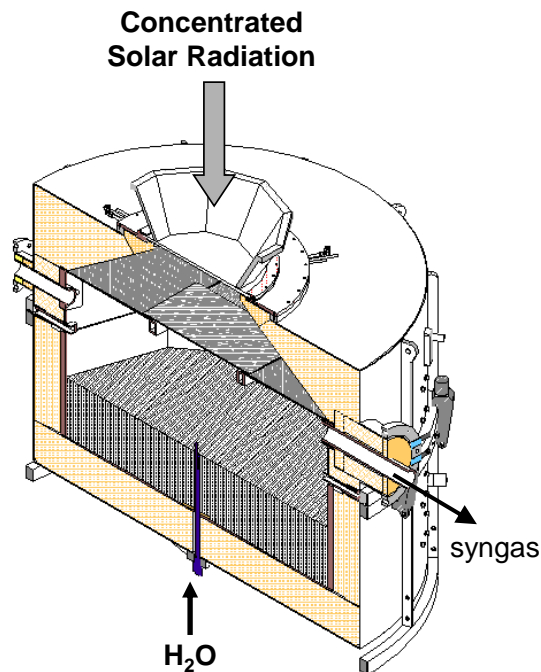
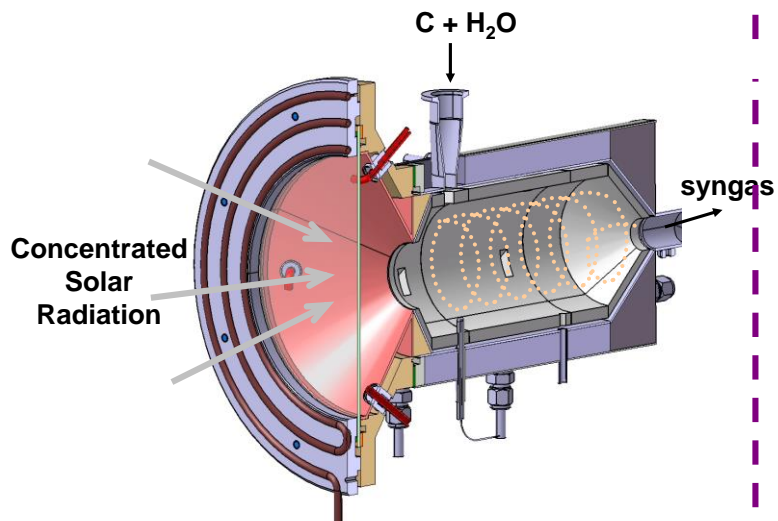
Dehydration, Pyrolysis, Gasification







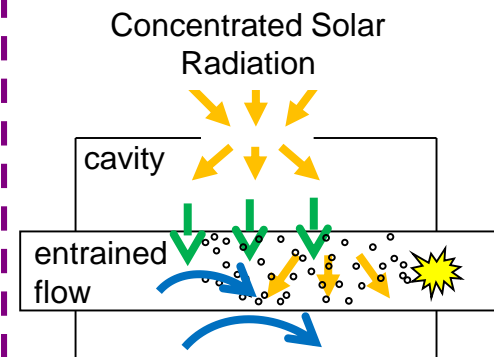
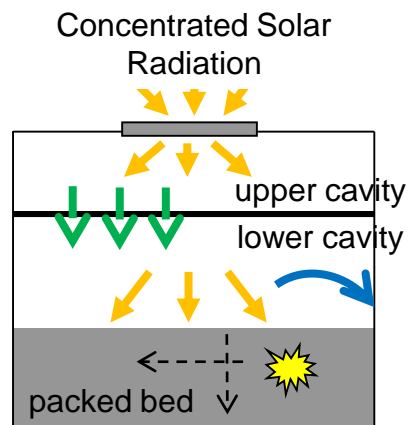
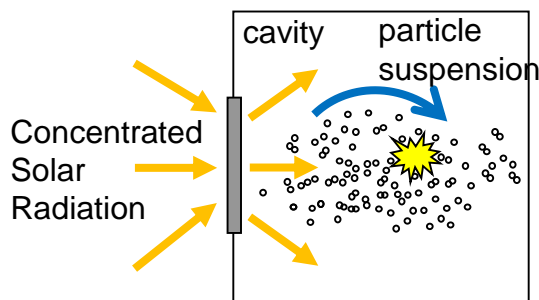
Solar Reactor Concepts

Direct-irradiation

Indirect-irradiation



-  convective heat transfer
-  conductive heat transfer
-  radiative heat transfer
-  chemical reaction

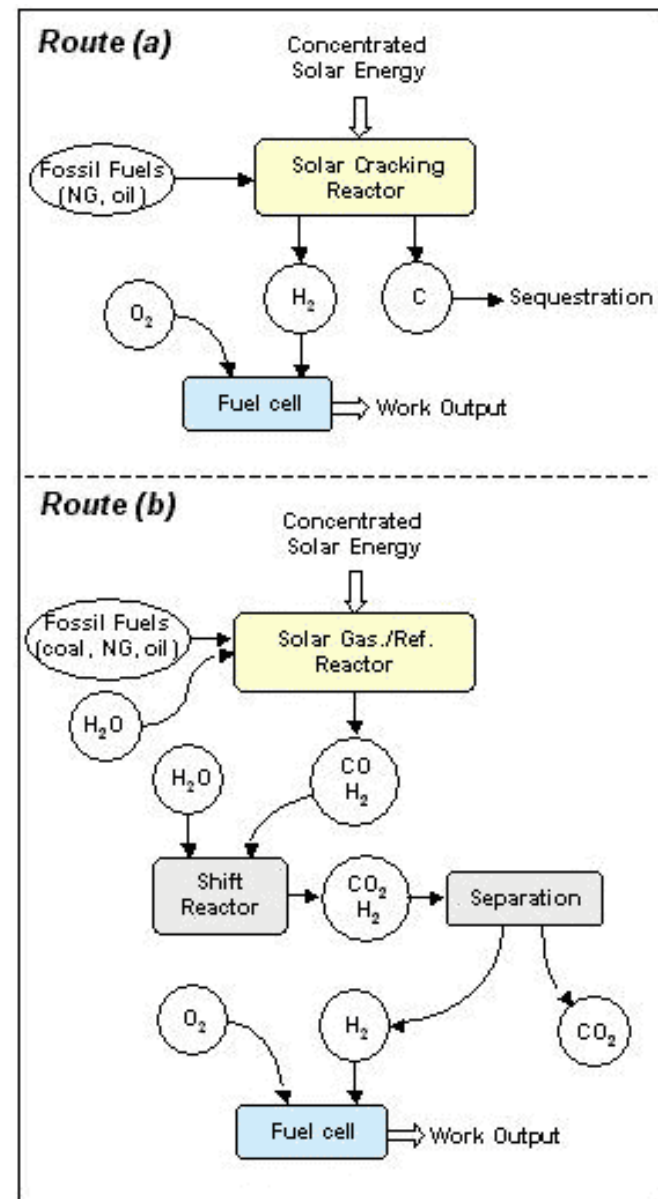


Hybrid solar conversion

- Hydrogen derived from fossil fuels has many impurities:
 - ~~• From combustion: CO_2 , CO , N_2~~
 - From the feedstock: sulfur
- Purification:
 - Desulfurization for gaseous feedstock: calcium-based slurries (SO_2 to sulfites and sulfates)
 - Desulfurization from solid/liquid feedstock: via catalysts into H_2S
 - ~~CO_2 removal:~~
 - ~~• temperature swing adsorption (solubility variation of CO_2 with temperature)~~
 - ~~• pressure swing adsorption (pressure dependent absorption of e.g. zeolites)~~
 - ~~• special membranes (cellulose)~~
 - CO removal from H_2 mixture: Hydrogen-permeable membranes made of metals (palladium)

Hybrid solar conversion

- Hybrid solar conversion
 - Advantage of hybrid process vs. conventional autothermal processes:
 - the gaseous products are not contaminated by combustion's by-products
 - the discharge of pollutants to the environment is reduced
 - the calorific value of the feedstock is upgraded
 - the fuel is decarbonized
 - there is no need for energy-intensive processing of pure oxygen

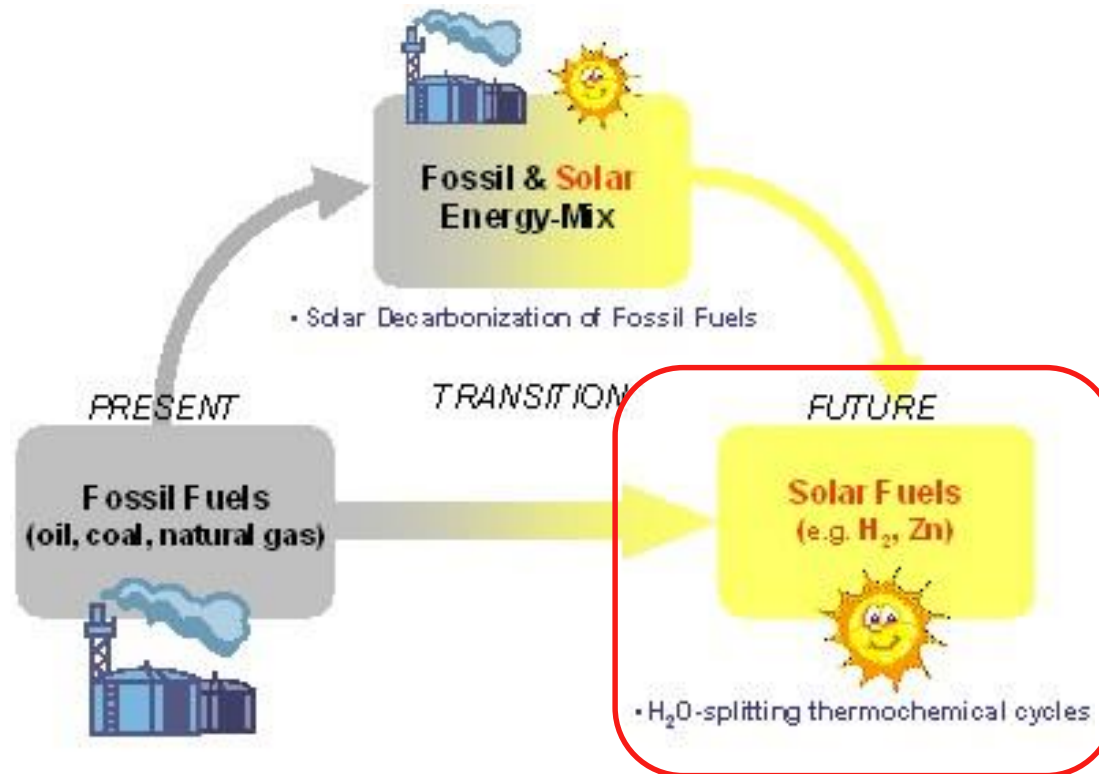


Renewable Energy

- Outline:
 - Conversion pathways solar-to-fuel
 - Hybrid pathways
 - Solar thermochemistry
 - Photochemistry

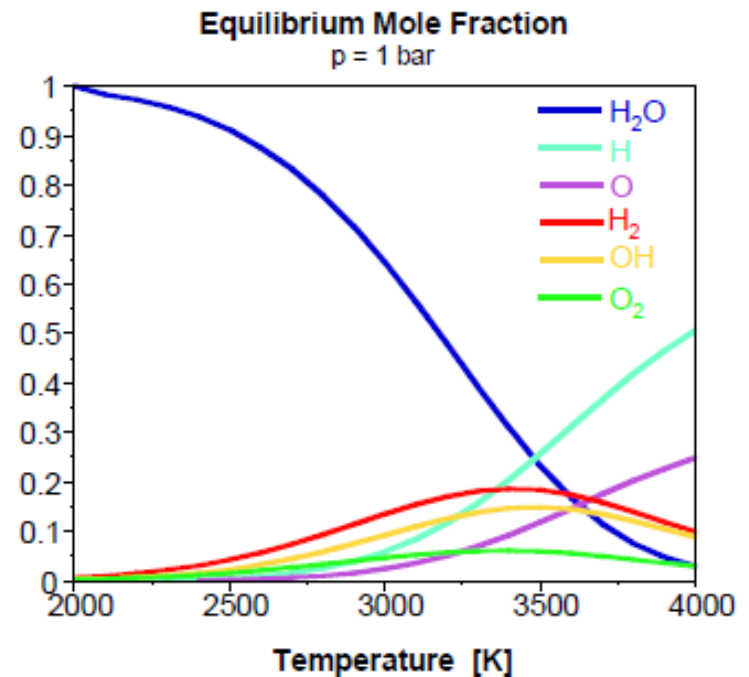
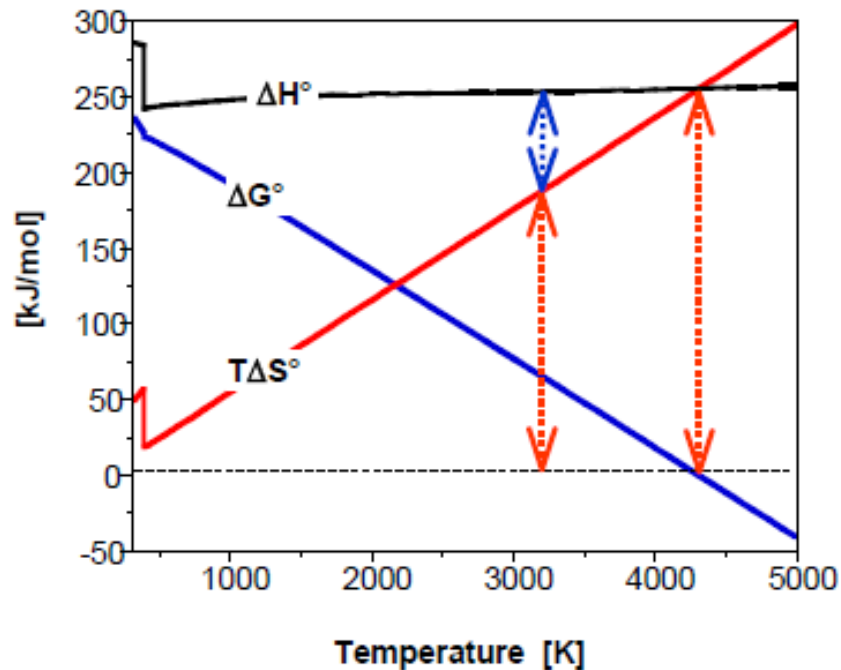
Solar thermolysis and thermochemistry

- In the transition to a renewable future, hybrid pathways using fossil fuels exclusively as chemical source for the fuel production and solar energy as the process heat



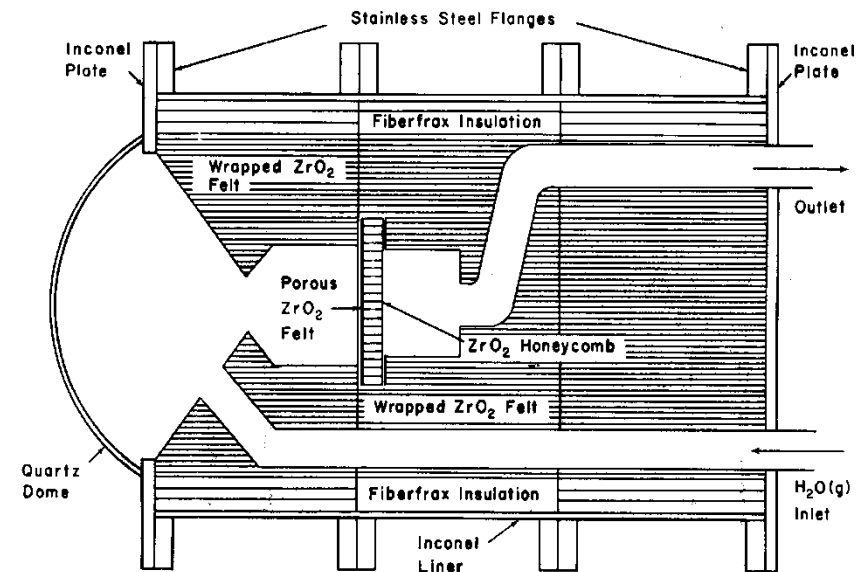
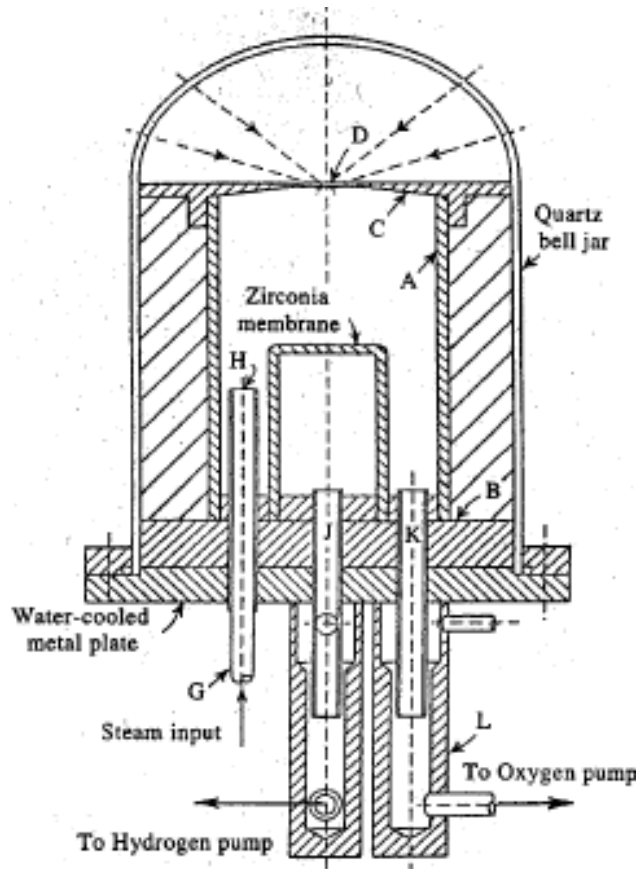
Solar thermolysis

- Solar thermolysis
 - Solar energy is used as process heat of chemical reaction
 - Direct thermolysis of water: $\text{H}_2\text{O} \rightarrow 1/2\text{O}_2 + \text{H}_2$



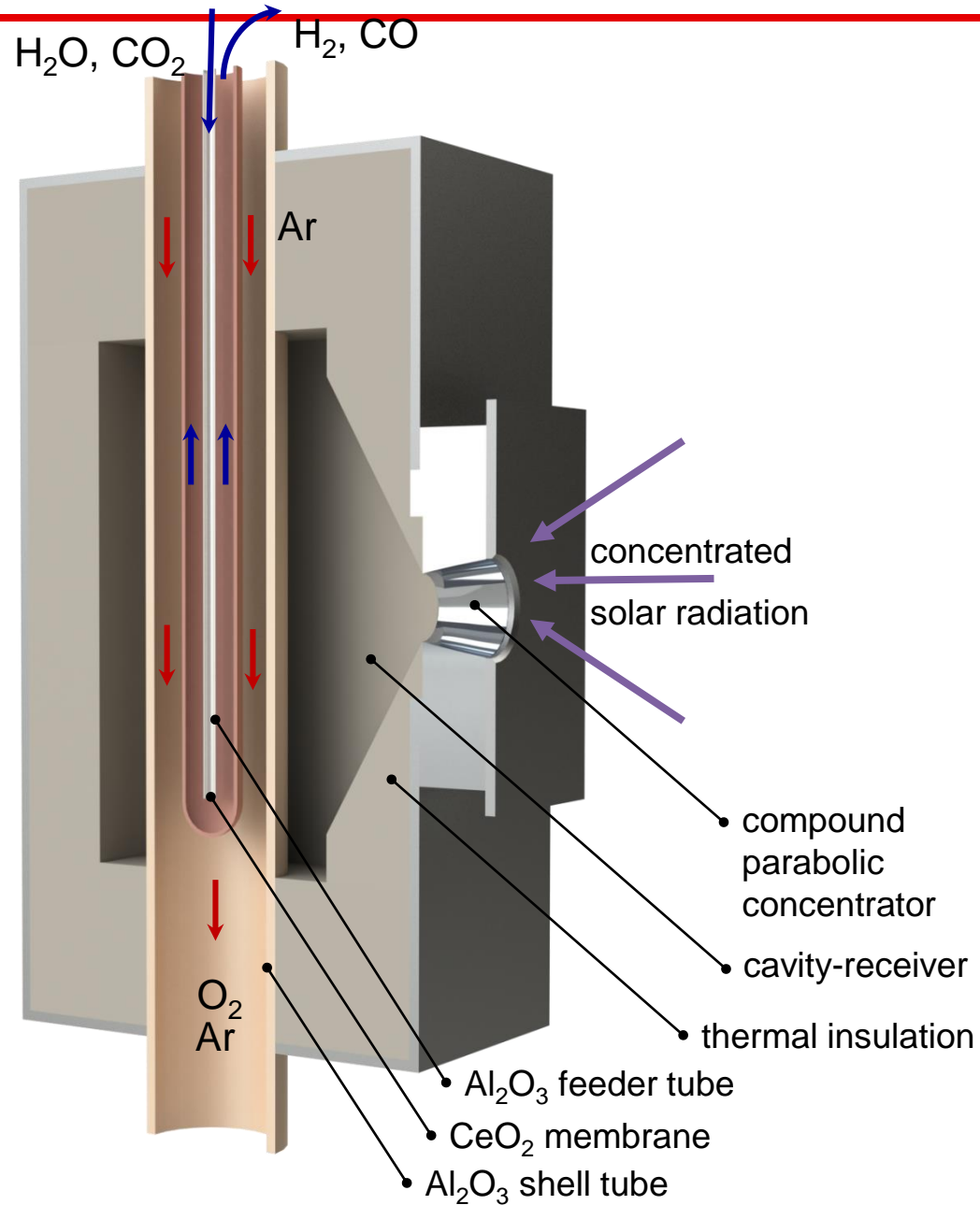
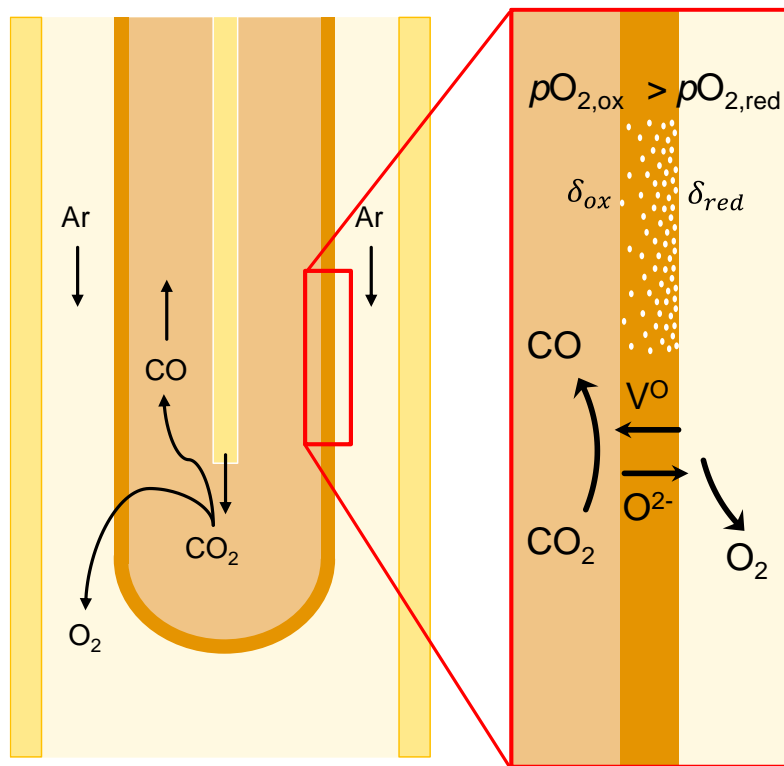
Solar thermolysis

- Reactor concept for solar thermolysis
 - Product separation by:
 - High temperature membranes
 - Rapid quenching of products



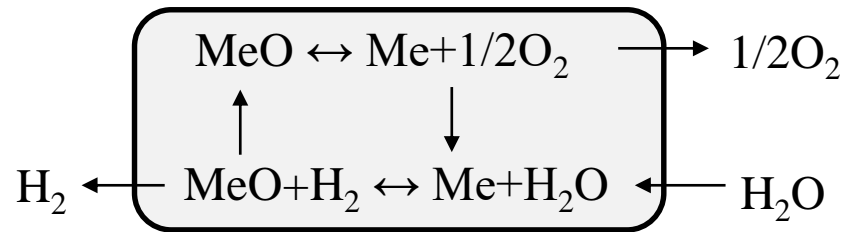
Fletcher E., *J. Solar Energy Engineering* **123**, 63-74, 2001

Solar Thermolysis

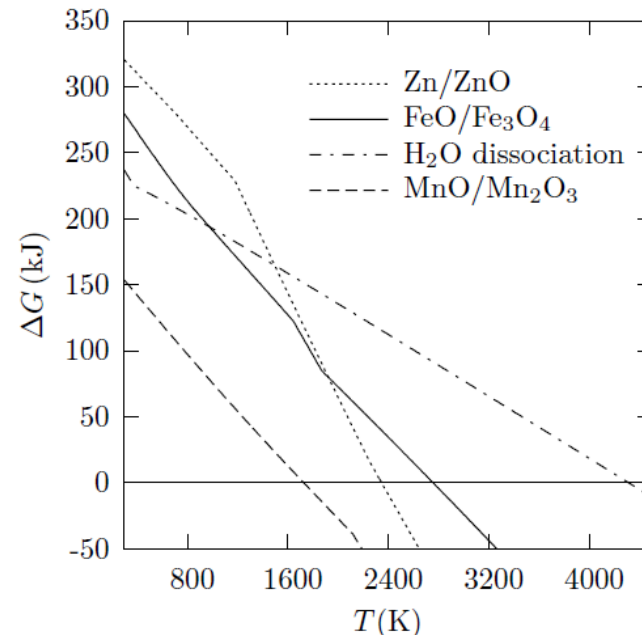


Solar thermochemistry

- Solar **thermochemical** cycles
 - Solar energy is used as process heat of chemical reaction
 - Multi-step water-splitting reactions:



- Omit explosive hydrogen and oxygen mixture since produced in separate steps
- Requires lower temperatures
- Possible redox pairs (Me/MeO):
 - Fe₂O₄/FeO
 - Ce₂O₃/CeO₂,
 - ZnO/Zn
 - SnO/SnO₂ ...



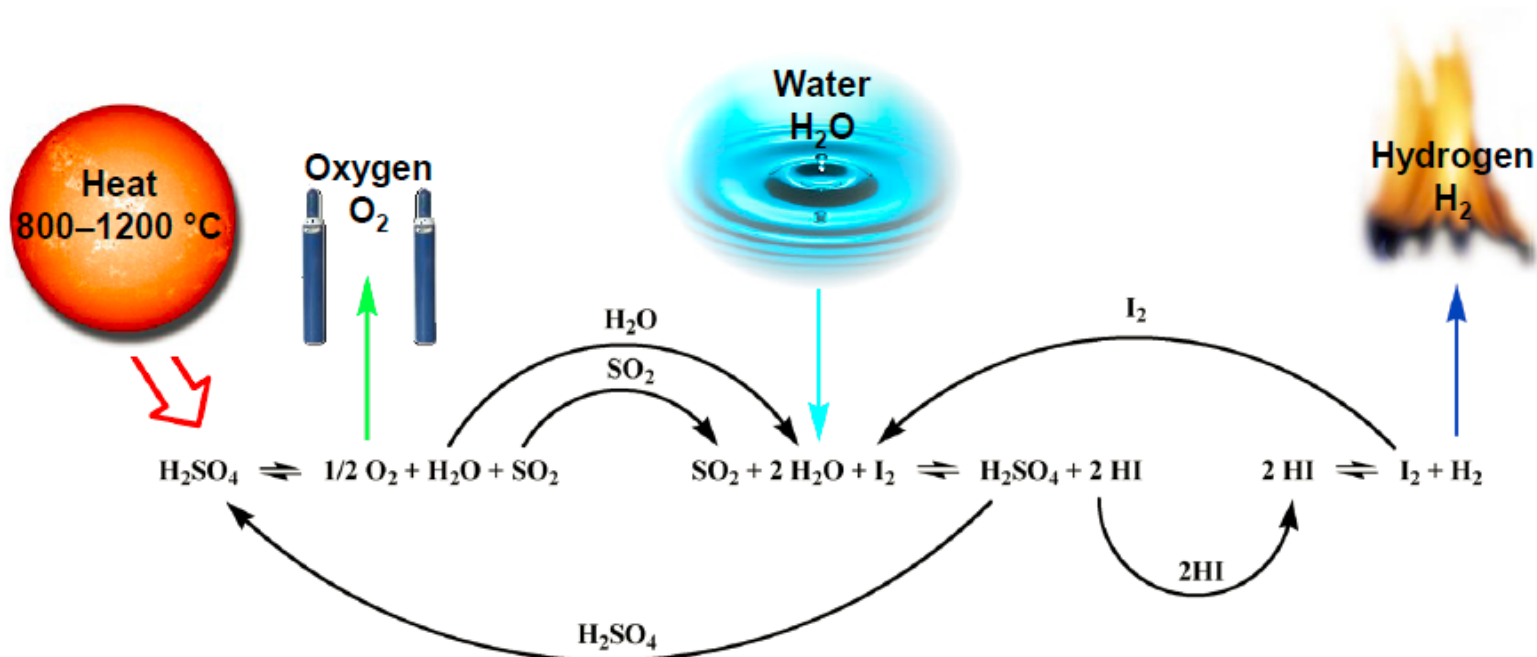
Solar thermochemistry

- Possible redox pairs for two-step cycles:

Cycle	Reactions	Cycle	Reactions
Zn/ZnO	$\text{ZnO} \rightarrow \text{Zn} + \text{O}_2$ $\text{Zn} + \text{H}_2\text{O} \rightarrow \text{ZnO} + \text{H}_2$	SoO ₂ /SiO	$\text{SiO}_2 \rightarrow \text{SiO} + 1/2 \text{O}_2$ $\text{SiO} + \text{H}_2\text{O} \rightarrow \text{SiO}_2 + \text{H}_2$
Fe ₃ O ₄ /FeO	$\text{Fe}_3\text{O}_4 \rightarrow 3 \text{FeO} + 1/2 \text{O}_2$ $3 \text{FeO} + \text{H}_2\text{O} \rightarrow \text{Fe}_3\text{O}_4 + \text{H}_2$	W/WO ₃	$\text{WO}_3 \rightarrow \text{W} + 3/2 \text{O}_2$ $\text{W} + 3\text{H}_2\text{O} \rightarrow \text{WO}_3 + 3\text{H}_2$
In ₂ O ₃ /In ₂ O	$\text{In}_2\text{O}_3 \rightarrow \text{In}_2\text{O} + 1/2 \text{O}_2$ $\text{In}_2\text{O} + 2\text{H}_2\text{O} \rightarrow \text{In}_2\text{O}_3 + 2\text{H}_2$	Hg/HgO	$\text{Hg} + \text{H}_2\text{O} \rightarrow \text{HgO} + \text{H}_2$ $\text{HgO} \rightarrow \text{Hg} + 1/2 \text{O}_2$
SnO ₂ /Sn	$\text{SnO}_2 \rightarrow \text{Sn} + \text{O}_2$ $\text{Sn} + 2\text{H}_2\text{O} \rightarrow \text{SnO}_2 + 2\text{H}_2$	Cd/CdO	$\text{Cd} + \text{H}_2\text{O} \rightarrow \text{CdO} + \text{H}_2$ $\text{CdO} \rightarrow \text{Cd} + 1/2 \text{O}_2$
MnO/MnSO ₄	$\text{MnSO}_4 \rightarrow \text{MnO} + \text{SO}_2 + 1/2 \text{O}_2$ $\text{MnO} + \text{H}_2\text{O} + \text{SO}_2 \rightarrow \text{MnSO}_4 + \text{H}_2$	CO/CO ₂	$\text{CO} + \text{H}_2\text{O} \rightarrow \text{CO}_2 + \text{H}_2$ $\text{CO}_2 \rightarrow \text{CO} + 1/2 \text{O}_2$
FeO/FeSO ₄	$\text{FeSO}_4 \rightarrow \text{FeO} + \text{SO}_2 + 1/2 \text{O}_2$ $\text{FeO} + \text{H}_2\text{O} + \text{SO}_2 \rightarrow \text{FeSO}_4 + \text{H}_2$	Ce ₂ O ₃ /CeO ₂	$\text{CeO}_2 \rightarrow \text{Ce}_2\text{O}_3$ $\text{Ce}_2\text{O}_3 + \text{H}_2\text{O} \rightarrow 2\text{CeO}_2 + \text{H}_2$
CoO/CoSO ₄	$\text{CoSO}_4 \rightarrow \text{CoO} + \text{SO}_2 + 1/2 \text{O}_2$ $\text{CoO} + \text{H}_2\text{O} + \text{SO}_2 \rightarrow \text{CoSO}_4 + \text{H}_2$	Mg/MgO	$\text{MgO} \rightarrow \text{Mg} + 1/2 \text{O}_2$ $\text{Mg} + \text{H}_2\text{O} \rightarrow \text{MgO} + \text{H}_2$
Fe ₃ O ₄ /FeCl ₂	$\text{Fe}_3\text{O}_4 + 6\text{HCl} \rightarrow 3\text{FeCl}_2 + 3\text{H}_2\text{O} + 1/2 \text{O}_2$ $3\text{FeCl}_2 + 4\text{H}_2\text{O} \rightarrow \text{Fe}_3\text{O}_4 + 6\text{HCl} + \text{H}_2$	SnO/SnO ₂	$\text{SnO}_2 \rightarrow \text{SnO} + 1/2 \text{O}_2$ $\text{SnO} + \text{H}_2\text{O} \rightarrow \text{SnO}_2 + \text{H}_2$
Mo/Mo ₂	$\text{MoO}_2 \rightarrow \text{Mo} + \text{O}_2$ $\text{Mo} + 2\text{H}_2\text{O} \rightarrow \text{MoO}_2 + 2\text{H}_2$		

Solar thermochemistry

- Three-step water-splitting cycles, e.g. sulfur-iodine:
 - further lower temperatures
 - but run in corrosive environment

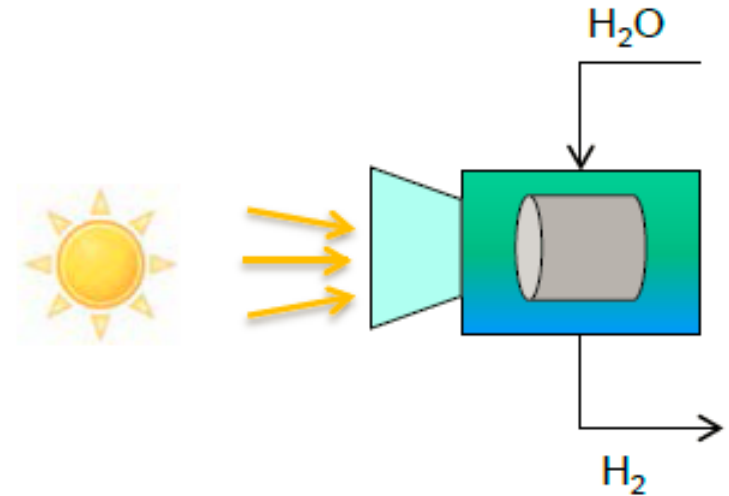
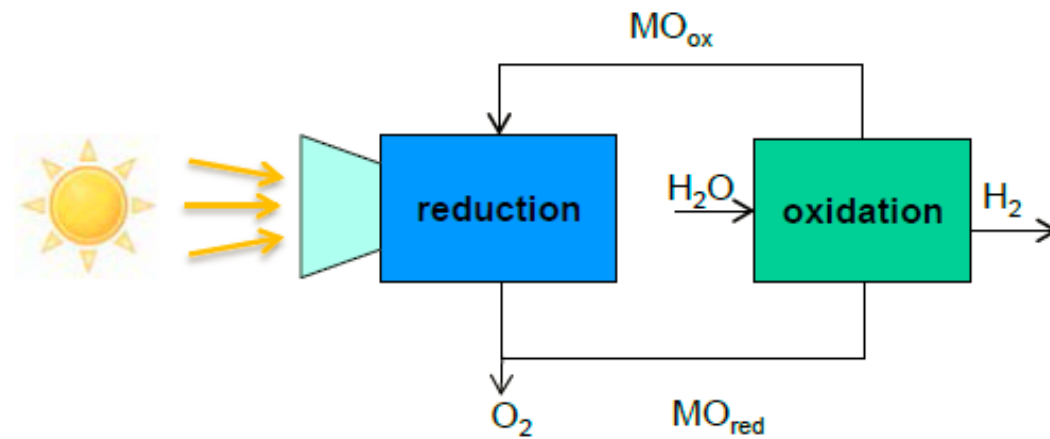


Solar thermochemistry

- Reactor concepts: two-step cycles

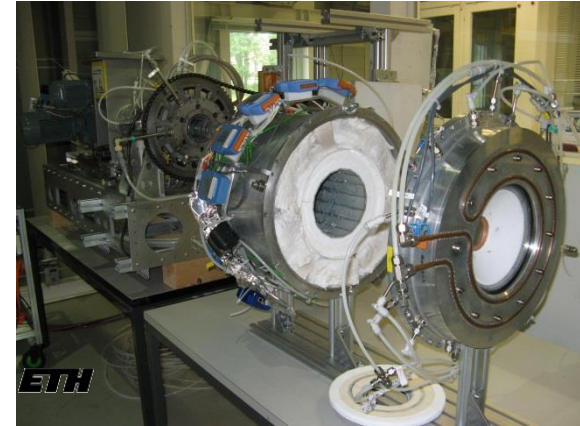
Moving material

Stationary material

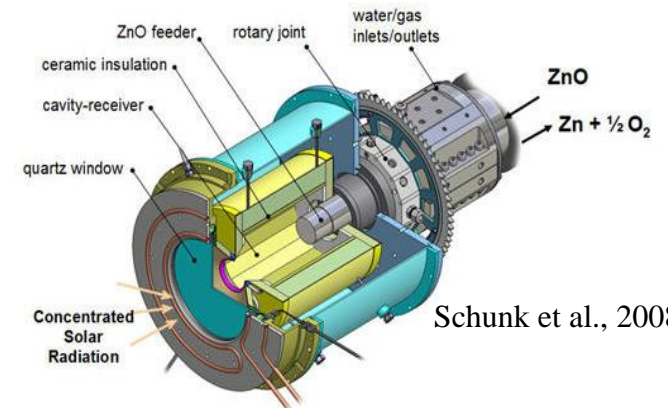
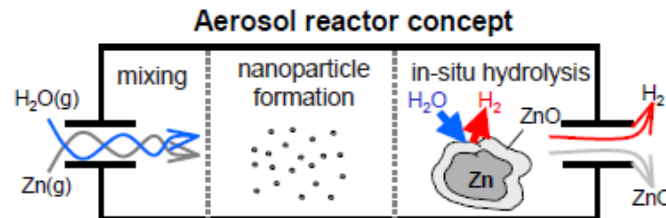


Solar thermochemistry

- Zn/ZnO-based proposed reactors, e.g. at ETH Zürich and PSI:
 - High-temperature reactor
 - 10 kW reactor
 - Reactor temperature: 2000 K
 - Peak concentration: 5800 suns



- Hydrolysis reactor:
 - Reactor temperature: 1263 K



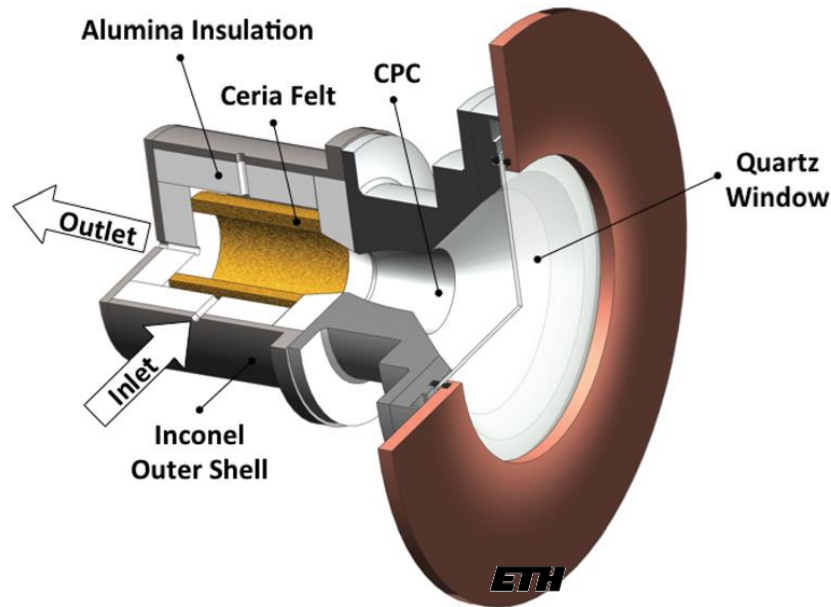
Schunk et al., 2008.

Melchior et al., 2009.

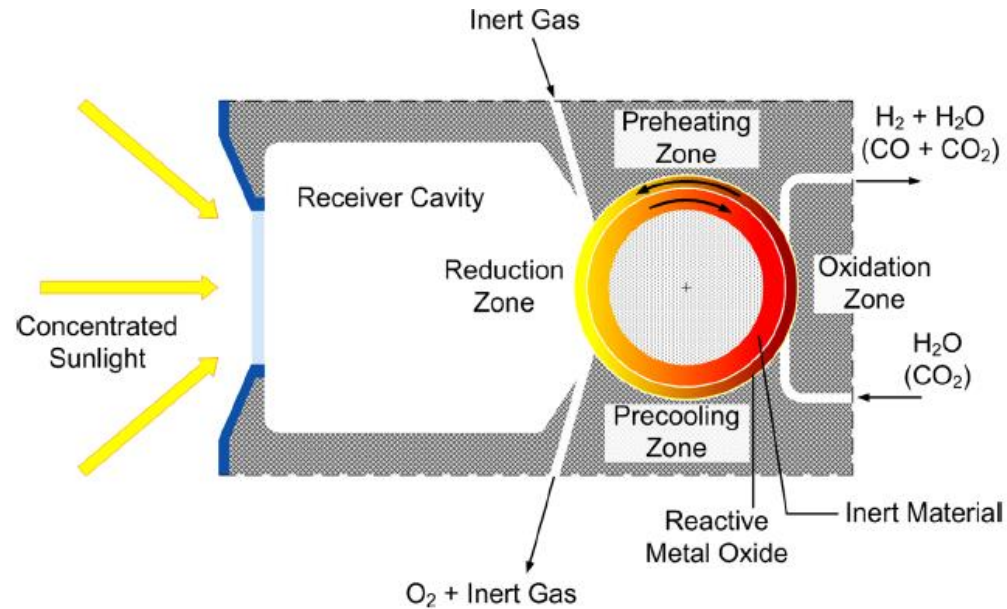
Solar thermochemistry

- Ceria-based proposed reactors, e.g.:

ETH Zürich



University of Minnesota

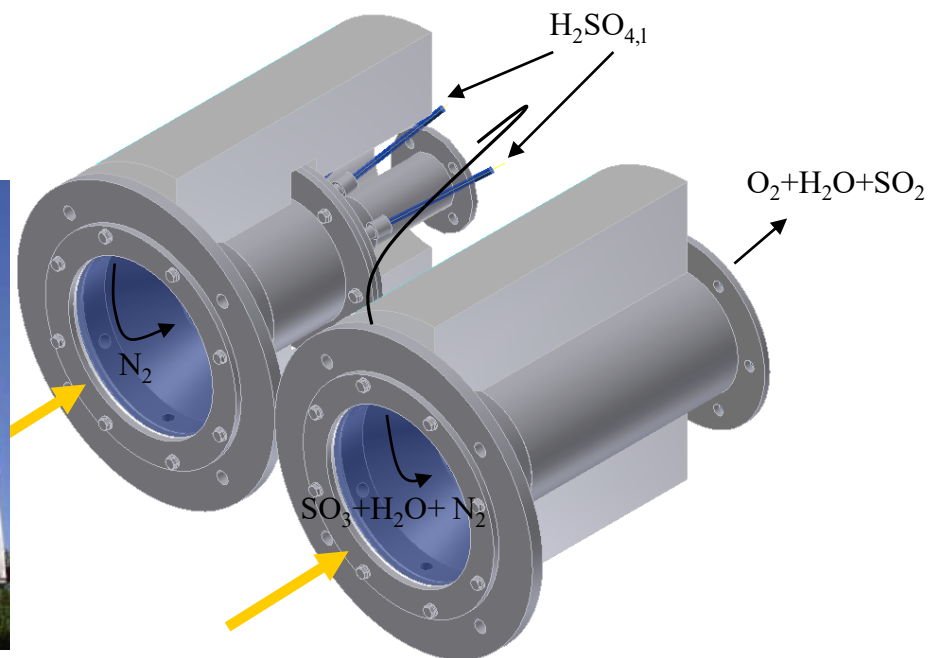
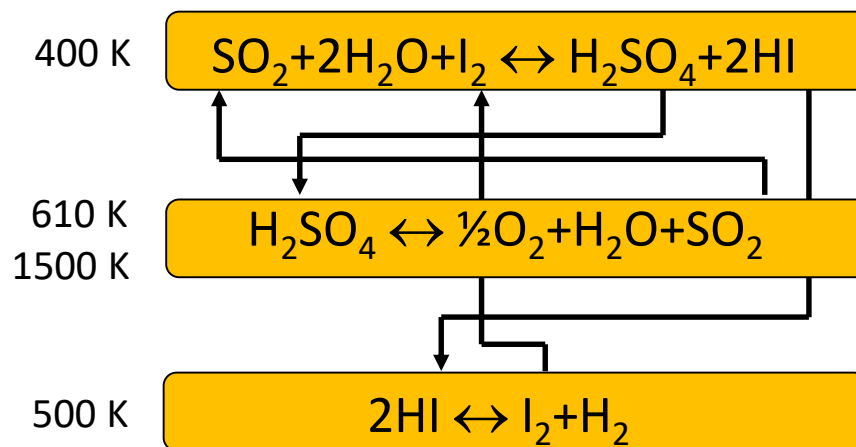


Temperature in reduction reaction: ~ 1800 K

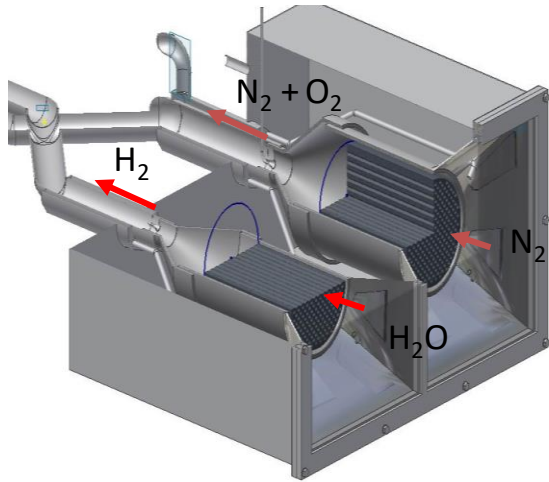
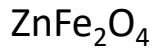
Temperature in oxidation reaction: ~ 1200 K

Solar thermochemistry

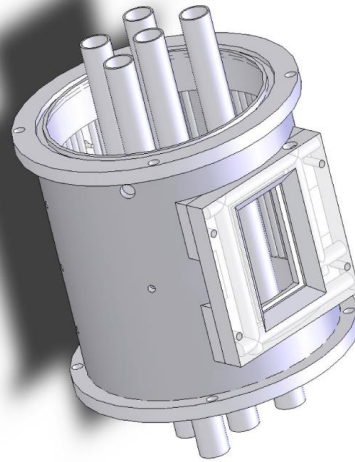
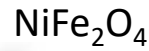
- SI-cycle, DLR Germany



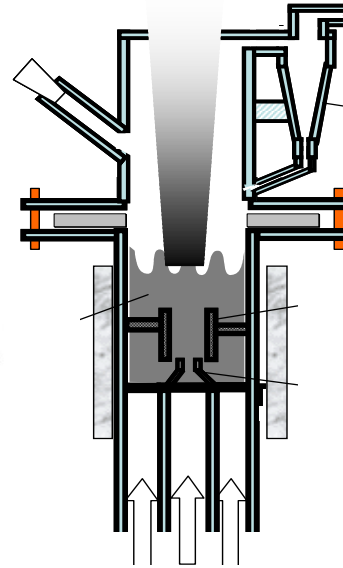
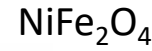
DLR, Germany



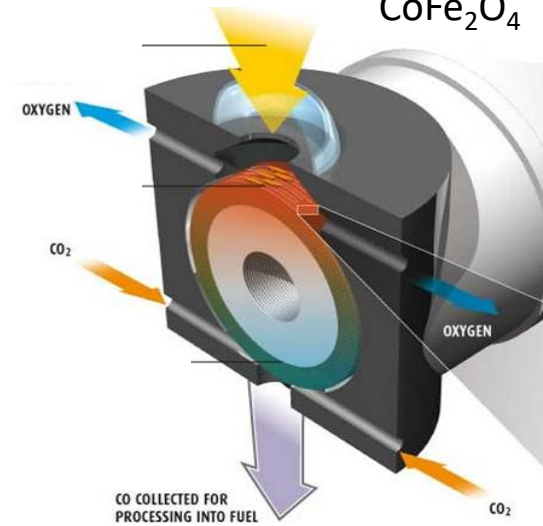
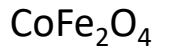
U. of CO, USA



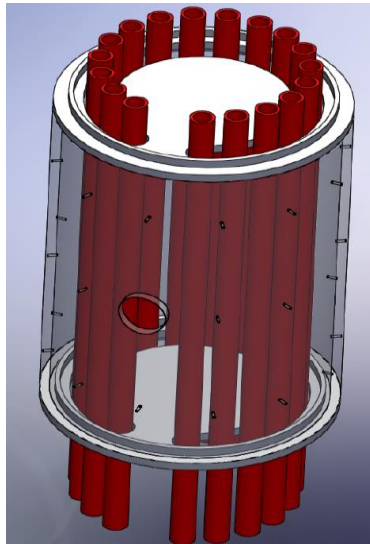
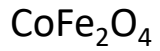
Niigata U., Japan



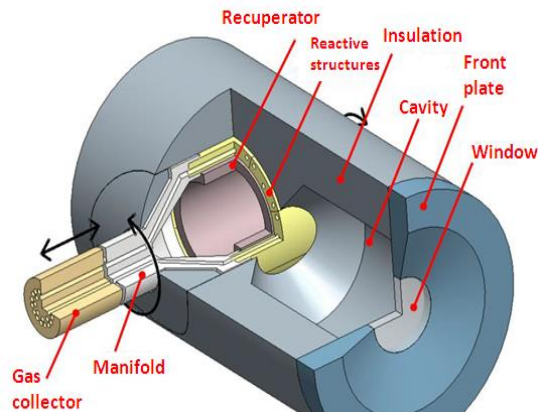
SNL, USA



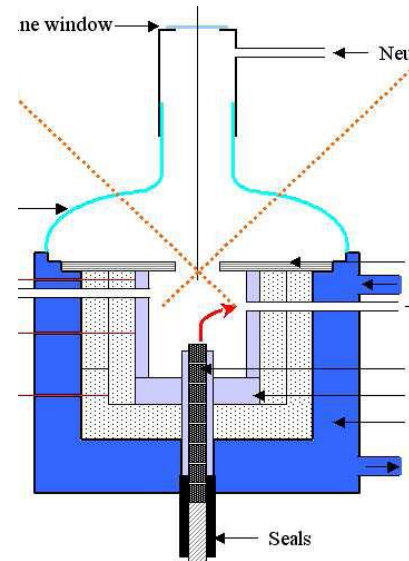
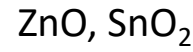
U. of FL, USA



U. of MN, USA



CNRS, France



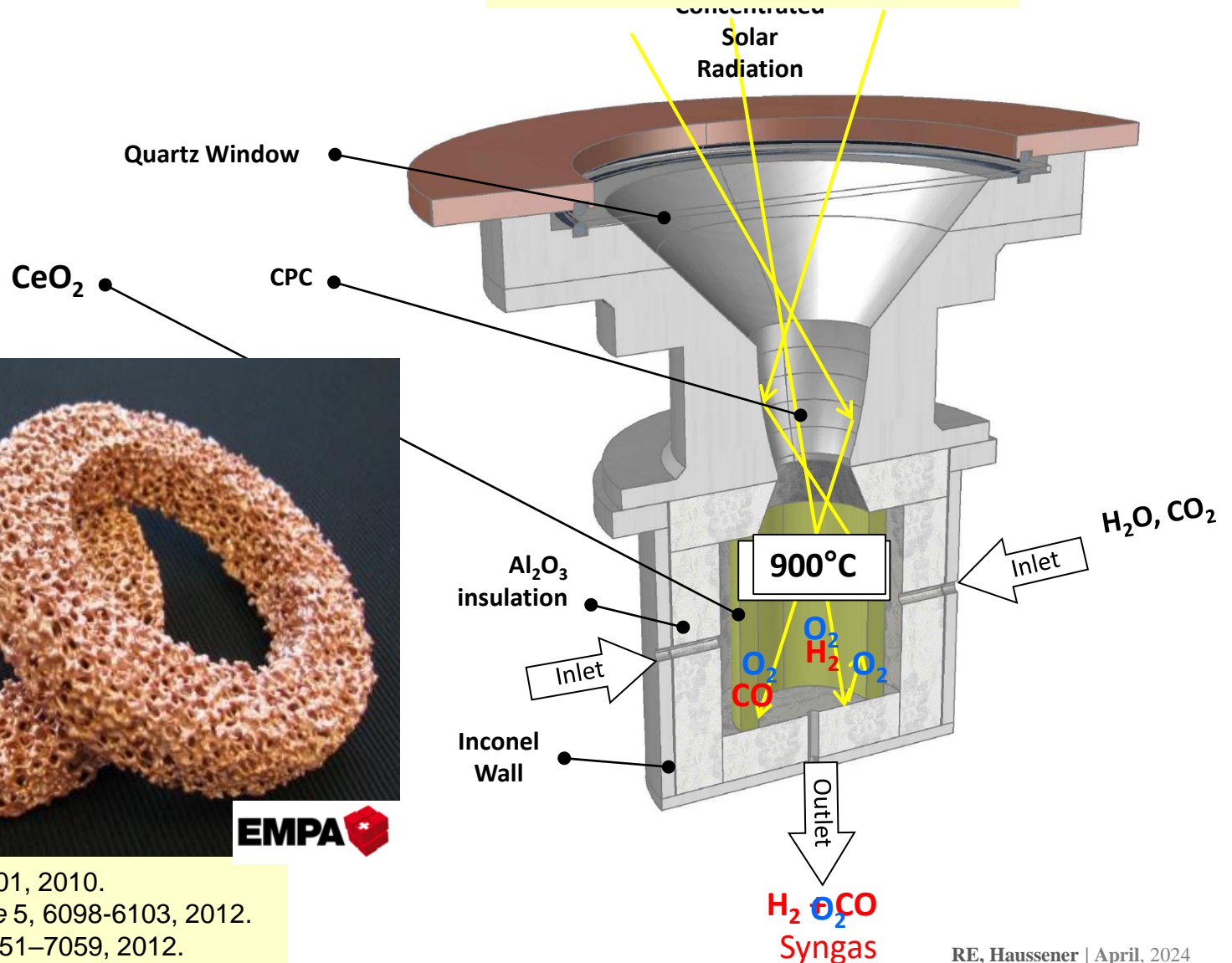
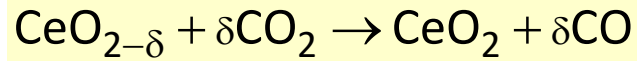
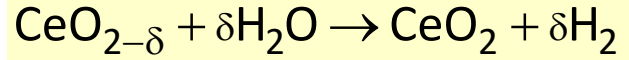
SNL, USA



Solar Reactor Technology



2nd step: Oxidation



RPC

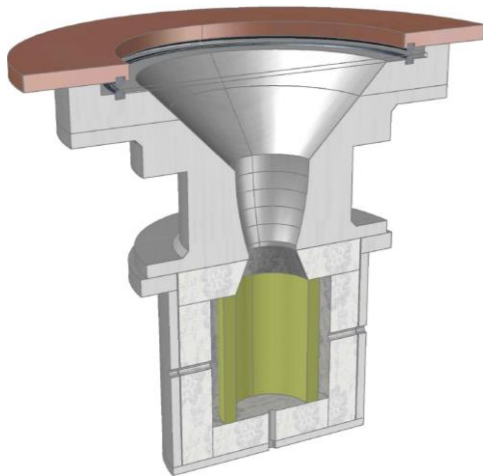


- Science 330, 1797-1801, 2010.
- Energy & Env. Science 5, 6098-6103, 2012.
- Energy & Fuels 26, 7051-7059, 2012.

Solar Reactor Technology

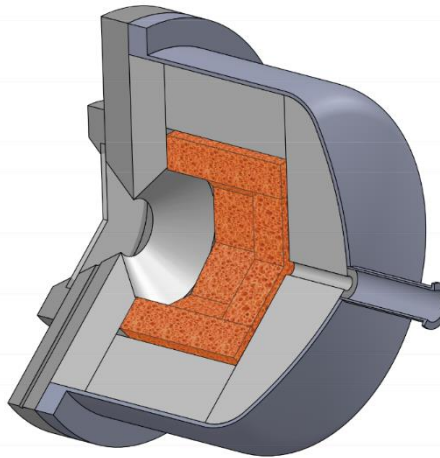


Generation 0
2 kW Lab-scale
Monoliths



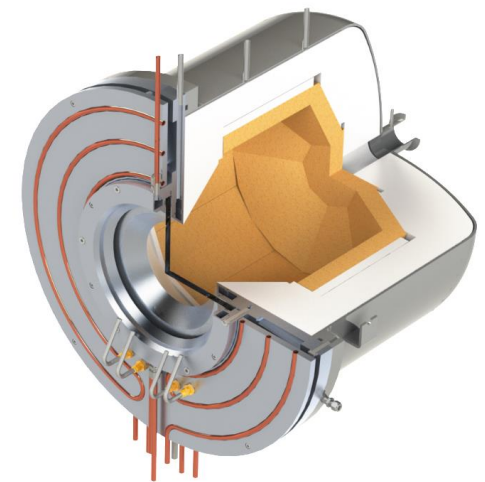
• Science 330, 1797-1801, 2010.

Generation 1
4 kW Lab-scale
RPC



• Energy & Env. Science 10;1142-1149, 2017.

Generation 2
50 kW Pilot-scale
RPC



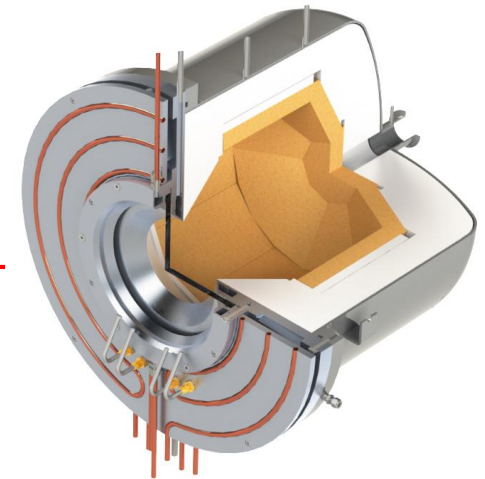
Solar Reactor Technology



EU Sun-to-Liquid Project



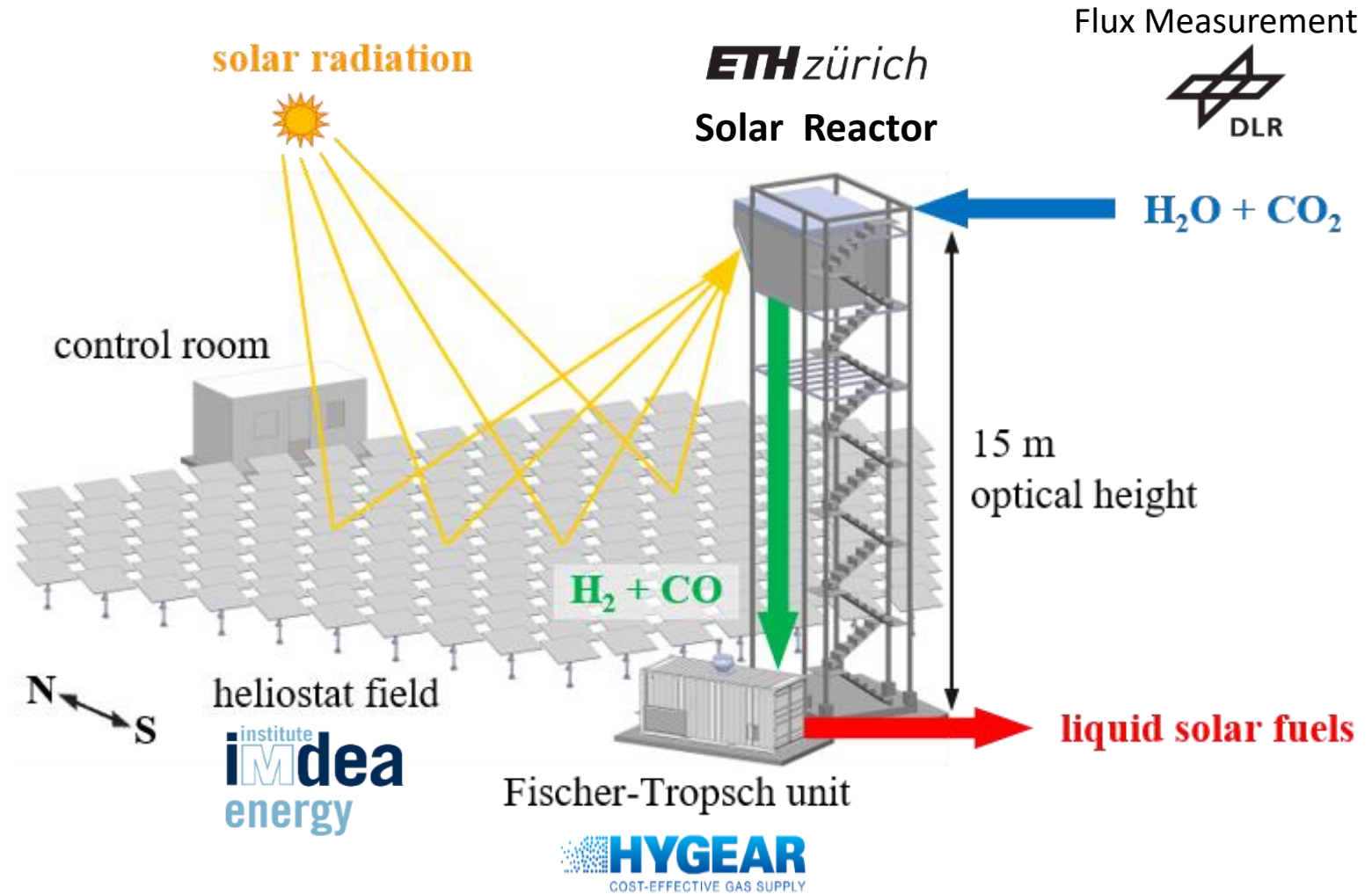
Generation 2
50 kW Pilot-scale
RPC



SUN to LIQUID
Fuels from concentrated sunlight

www.sun-to-liquid.eu

EU Sun-to-Liquid Project

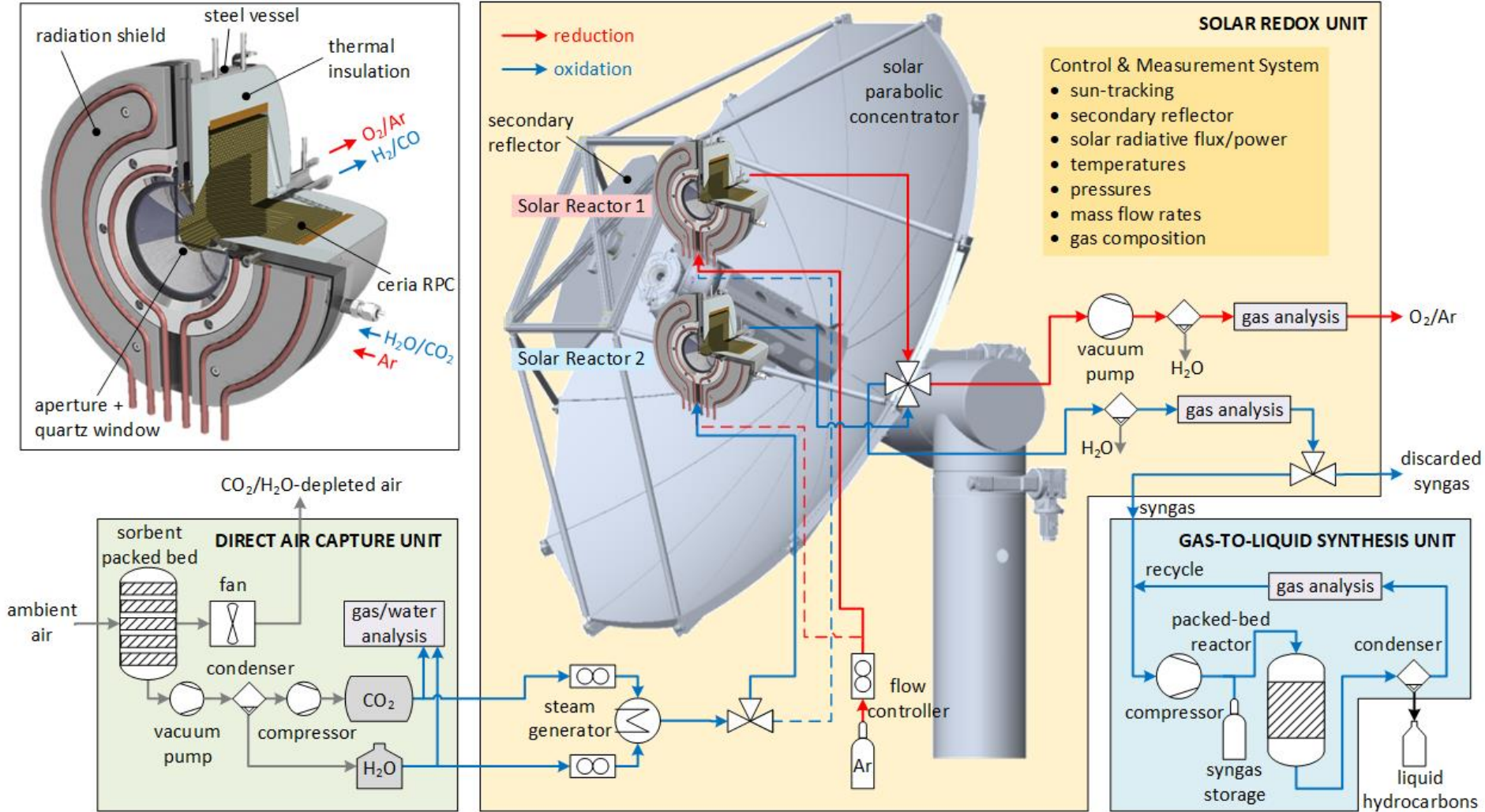


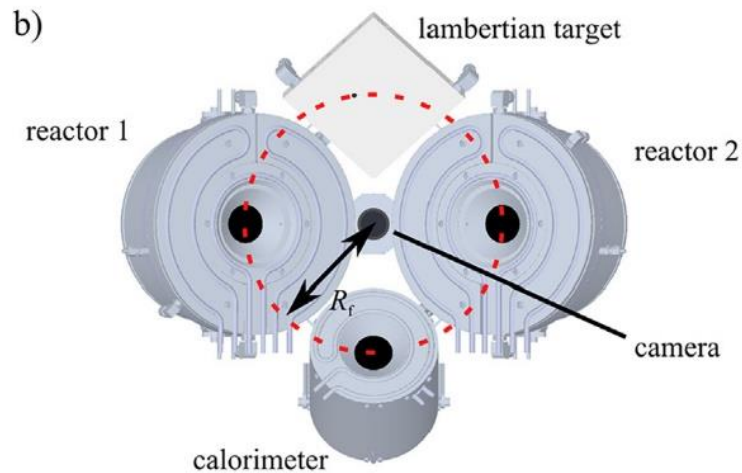
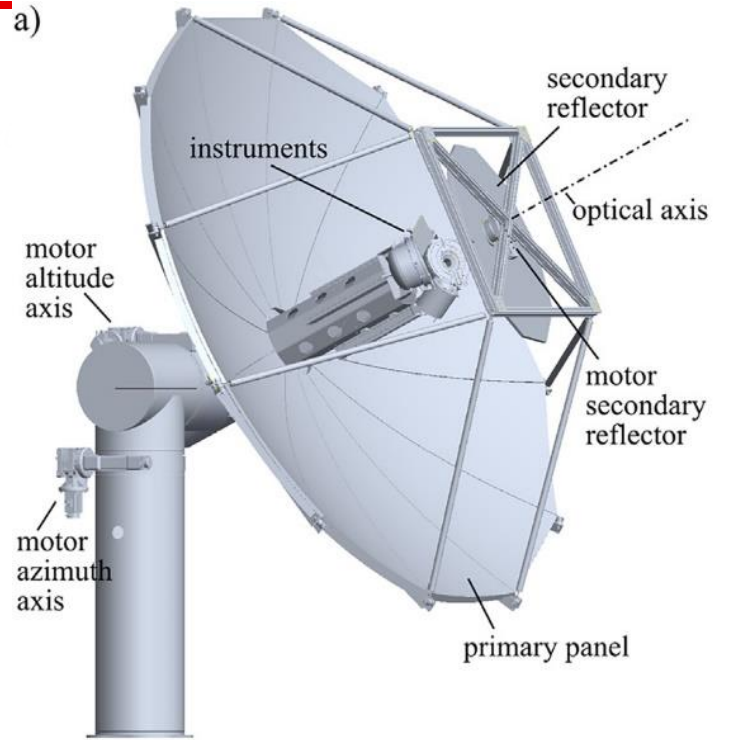
Fuels from Sunlight and Air

1st-ever production of carbon-neutral hydrocarbon fuels from sunlight and air
13-6-2019



Solar Mini-Refinery @ ETH



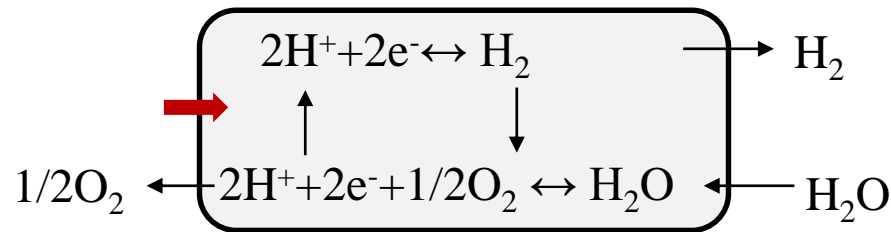


Renewable Energy

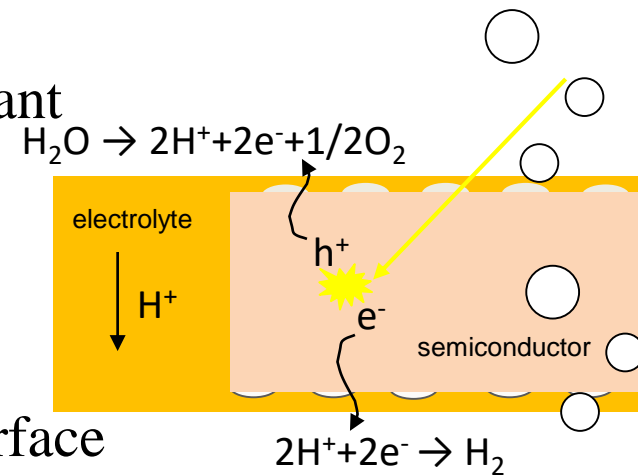
- Outline:
 - Conversion pathways solar-to-fuel
 - Hybrid pathways
 - Solar thermochemistry
 - Photochemistry

Photoelectrochemistry

- Photoelectrochemical processes
 - Solar energy is used as photon energy for the internal production of charge, which is separated at the solid-liquid junction
 - Multi-step water-splitting reactions ($E_0=1.23$ V):

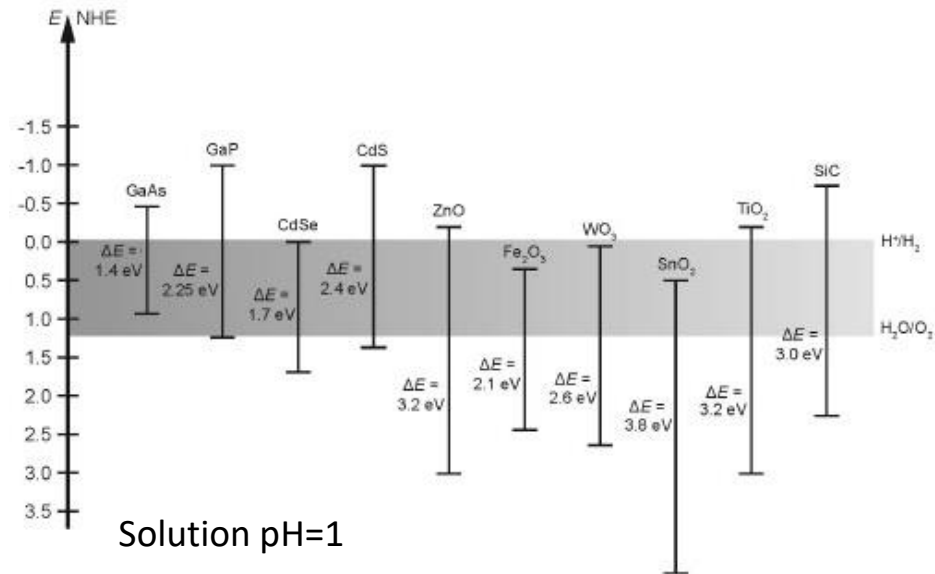


- Works at room temperature
- Spectral distribution of solar radiation important
- Processes:
 - Solar absorption
 - Electron-hole generation
 - Use electron and holes at liquid-solid interface
 - Ionic transport



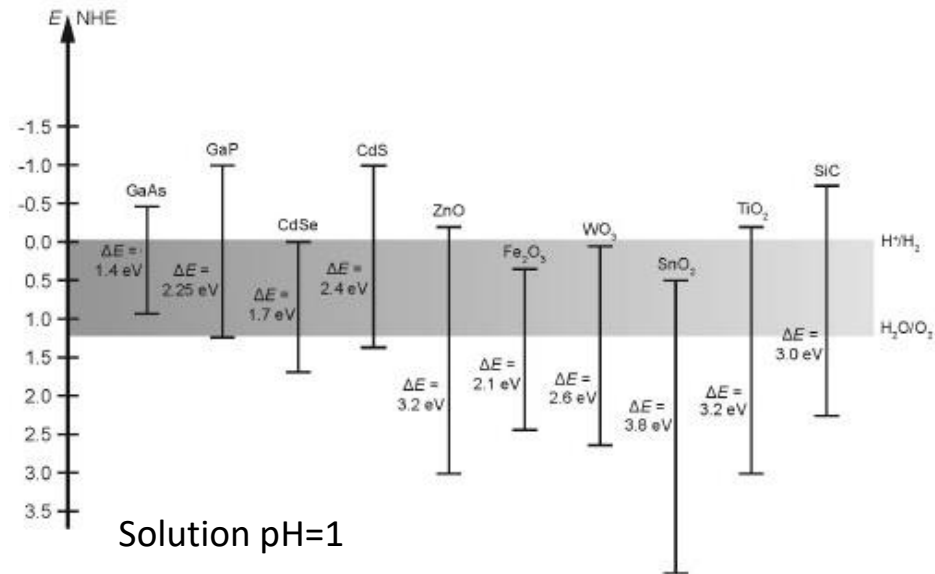
Photoelectrochemistry

- Stringent material requirements:
 - band gap size
 - suitable band edge position
 - high chemical stability in the dark and under illumination, as well as under highly acidic or base conditions
 - efficient charge transport in the semiconductor
 - selective and efficient electrochemical reactions
 - earth-abundance and low costs



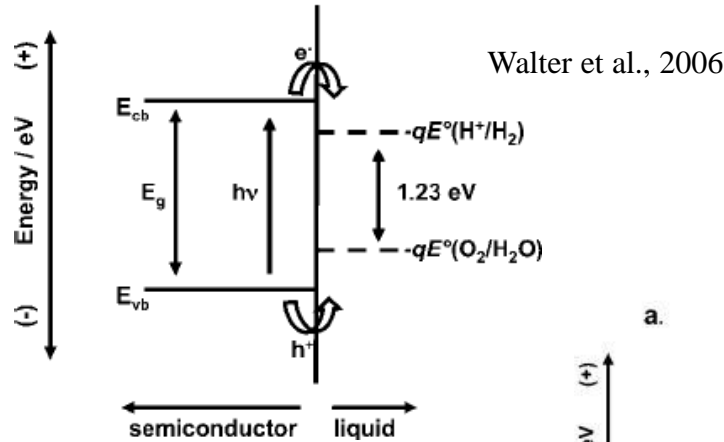
Photoelectrochemistry

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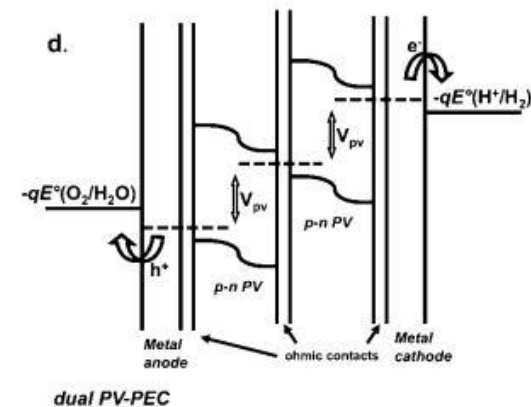
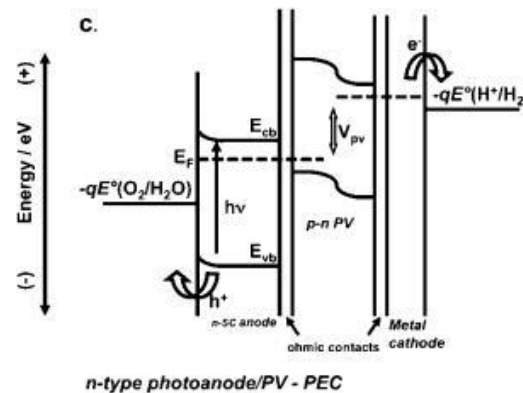
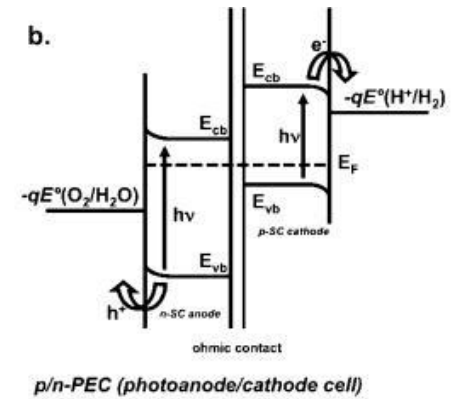
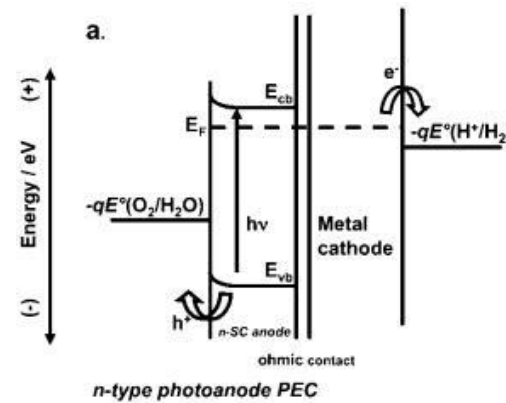


Photoelectrochemistry

- Band gap and band position of photoelectrode material must match reaction potentials:



- Various possible architectures:



Photoelectrochemistry

- Calculations:

- Photoactive material(s) will show

- diode-like current-potential behavior:

$$i = i_L - i_0 \left(\exp\left(\frac{qV}{kT}\right) - 1 \right)$$

Elementary charge

- Electrochemical system shows losses:

- Reaction overpotentials

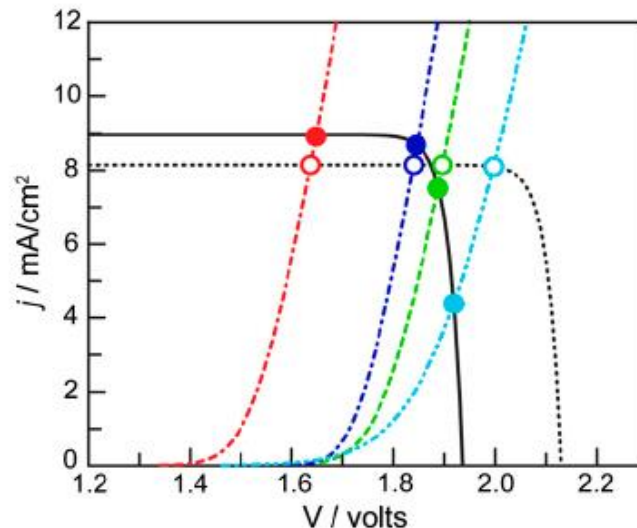
- Ohmic losses

$$E = E_0 + \eta_a + \eta_c + iR_{sol} + E_{mem} + E_{conc} > E_0$$

- Concentration losses

- Electrochemical load curve will show electrolyzer like load curve

- Intersection between both is operating point



Surendranath et al., 2012

Photoelectrochemistry

- Calculations:
 - Electrochemical system shows losses:

- Reaction overpotentials

- E.g. via Tafel equations:

$$\eta_a = a_1 \log\left(\frac{i}{i_{0a}}\right) \quad \eta_c = a_2 \log\left(\frac{i}{i_{0c}}\right)$$

Tafel slope

- Or Buttlar-Volmer:

$$i_R = i_{0a/c} \left[\exp\left(\frac{\alpha_a F \eta_{a/c}}{RT}\right) - \exp\left(\frac{\alpha_c F \eta_{a/c}}{RT}\right) \right]$$

Exchange current density Transfer coefficient

- Ohmic losses account for resistances in electrolyte, membrane, and solid conductor:

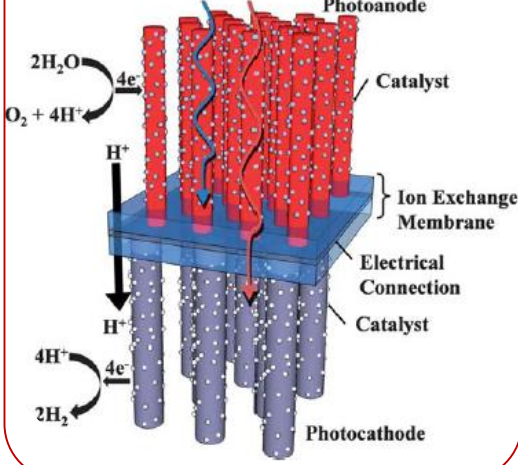
$$\Delta V_{\text{ohm}} = i \rho_{\text{sol}} l$$

resistivity Characteristic ion and electron path length

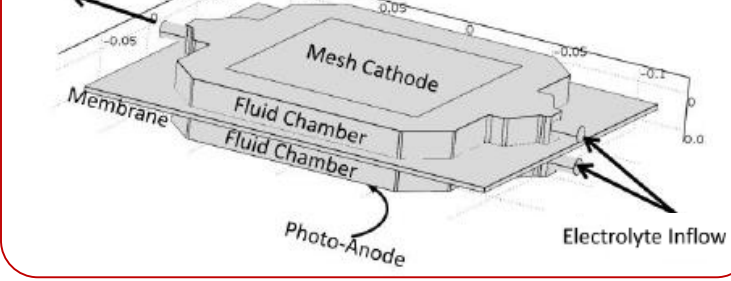
Photoelectrochemistry

- Proposed devices

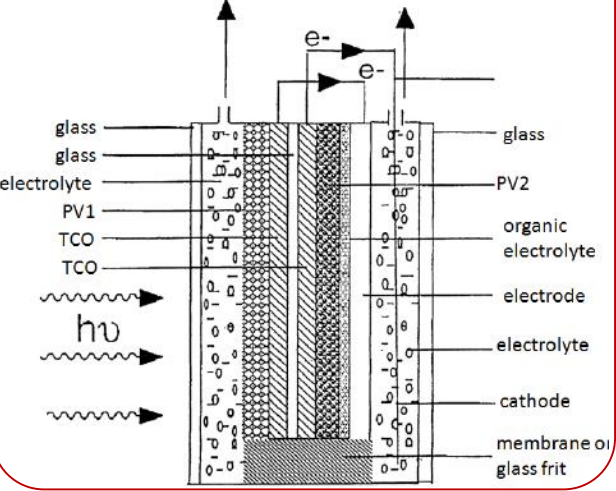
Spurgeon et al., 2011



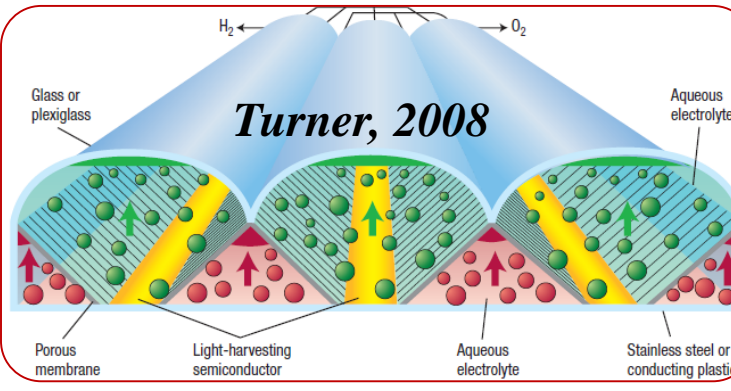
Carver et al., 2012



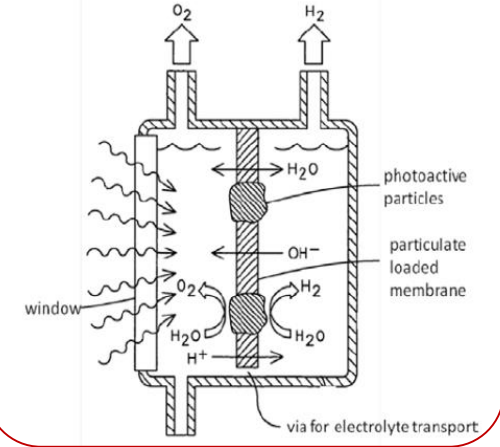
Grätzel et al., 2007



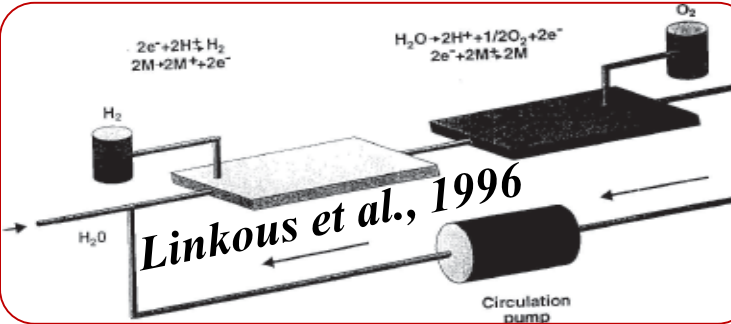
Turner, 2008



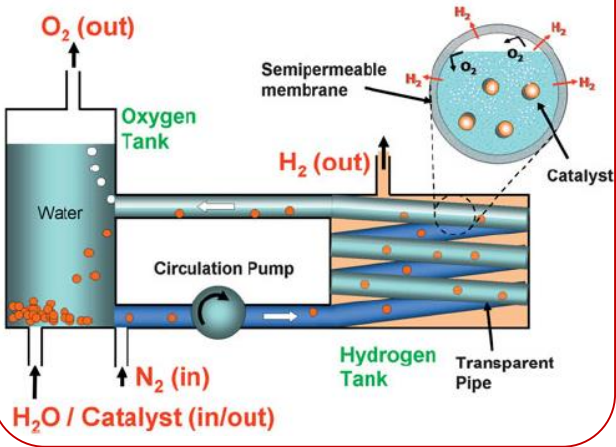
Miller et al., 2007



Linkous et al., 1996

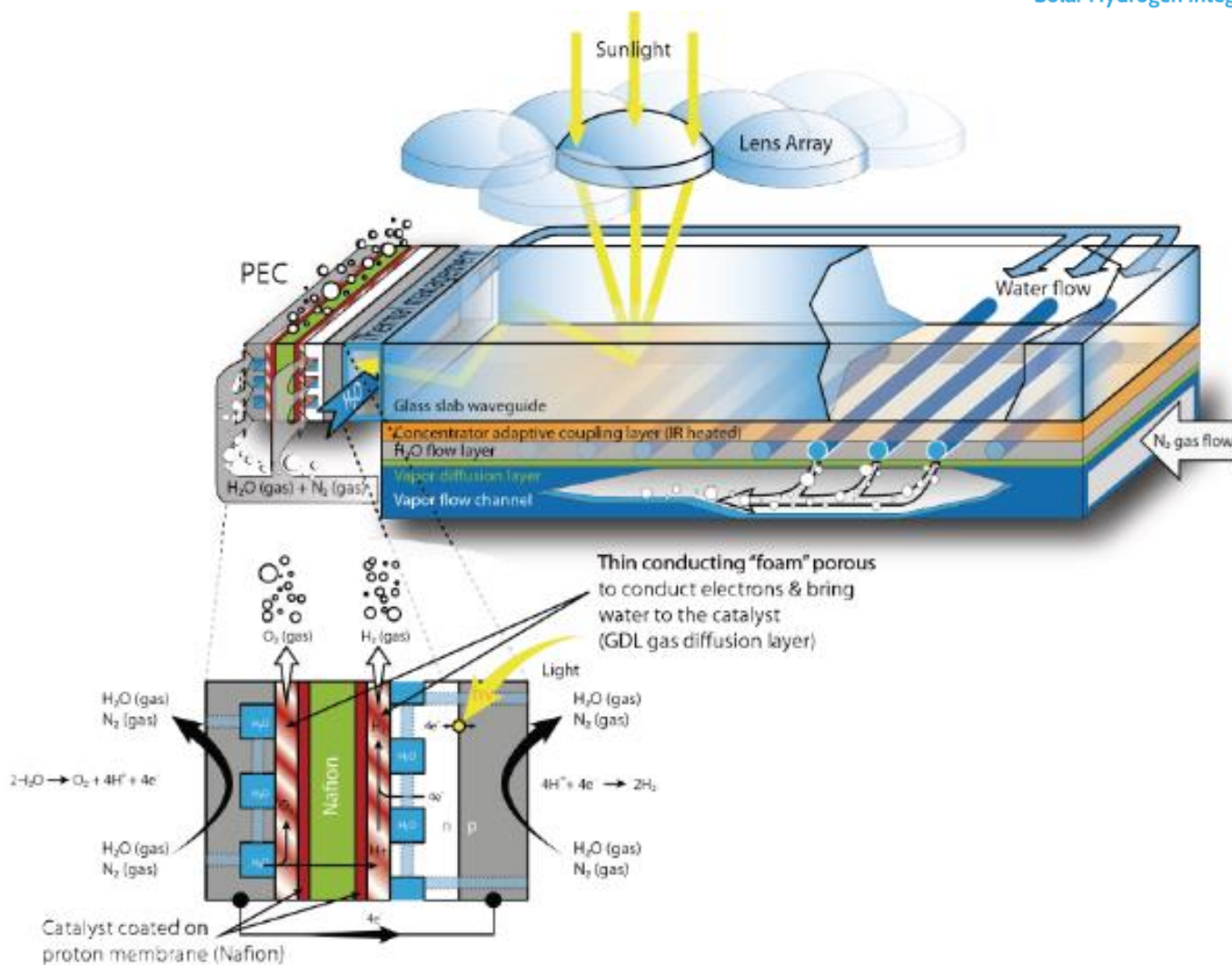


Parkinson et al., 2011



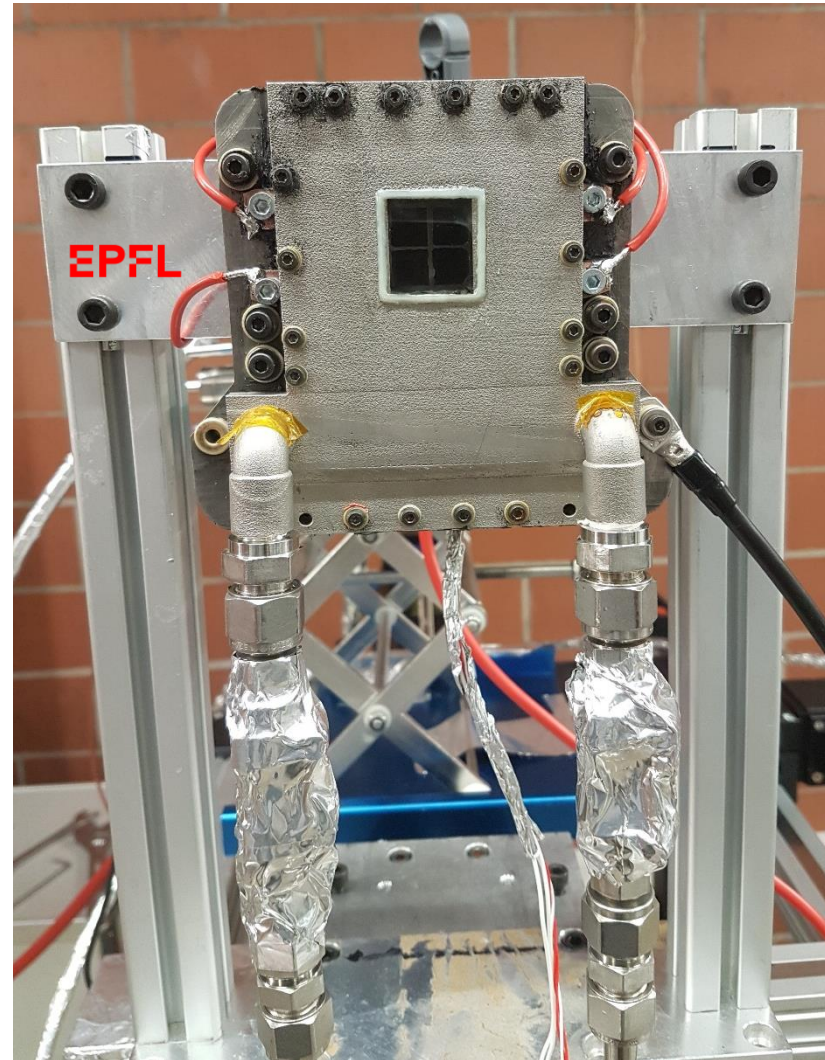
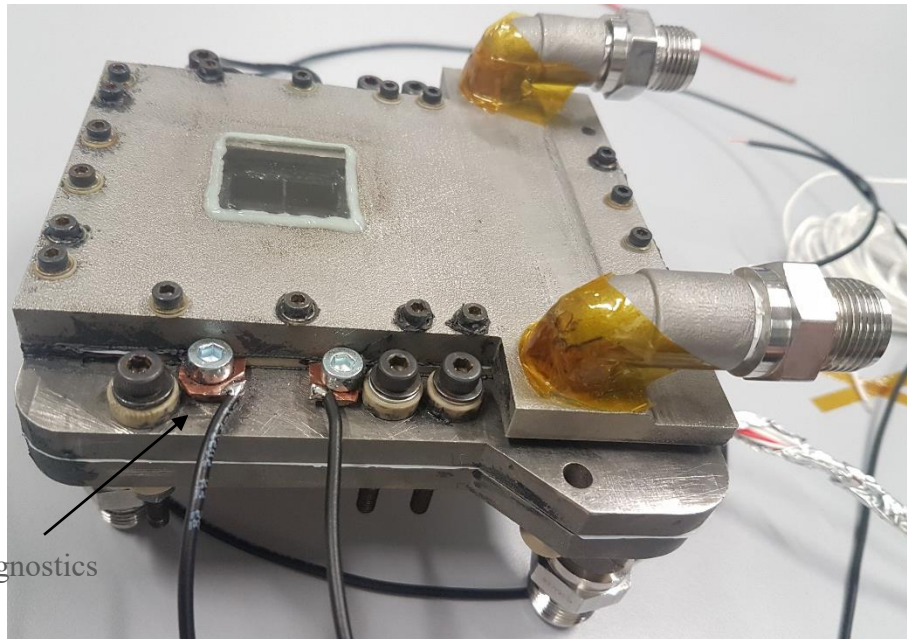
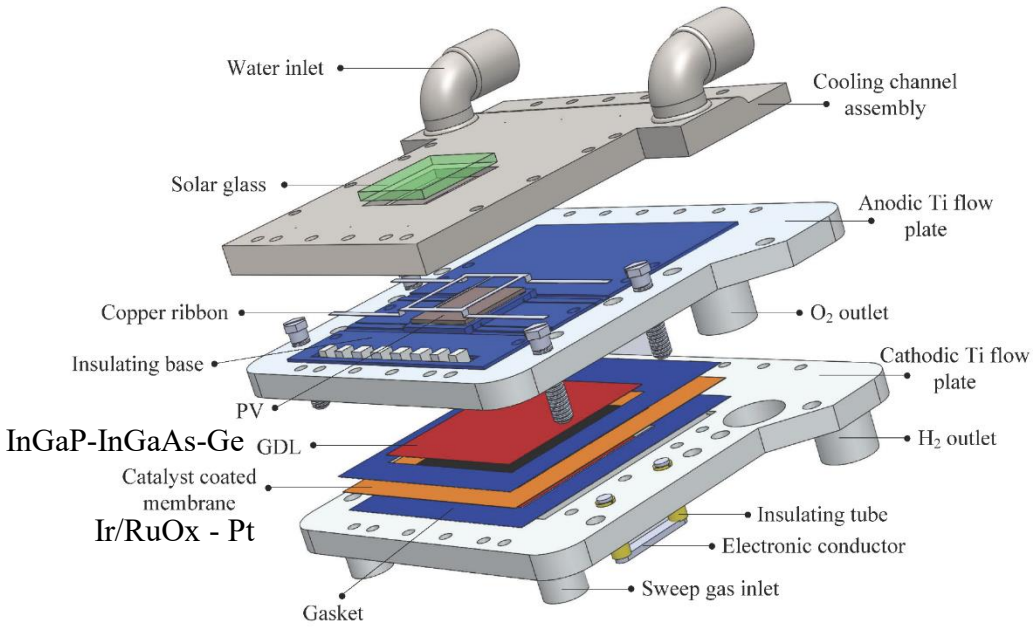
Photoelectrochemistry

- Proposed devices

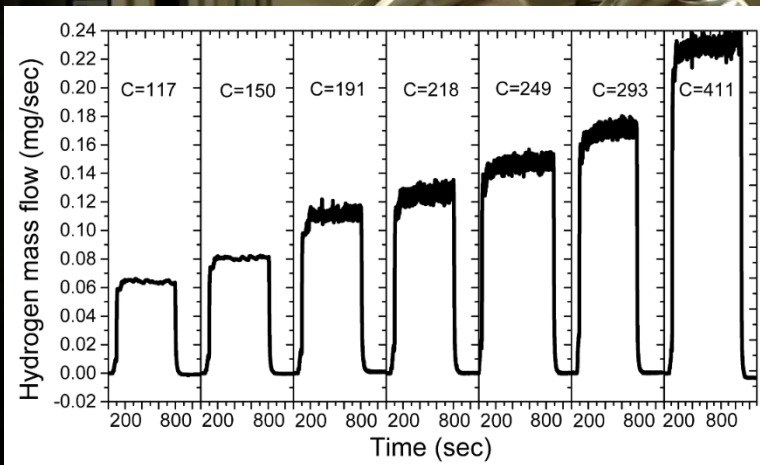
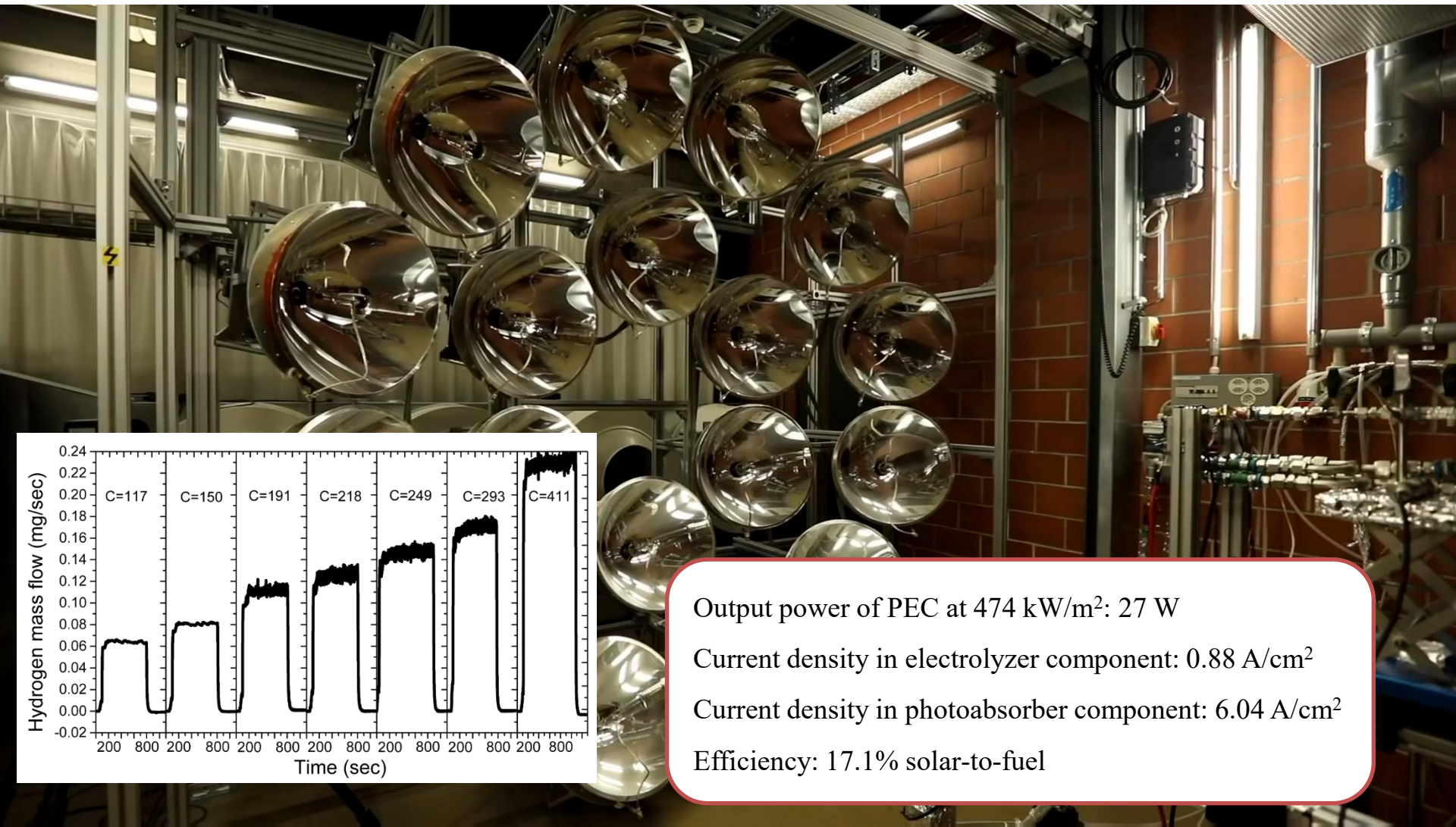


Photoelectrochemistry

US Patent 62/376923
EP Patent 16020308.9



Experimental demonstration



Output power of PEC at 474 kW/m²: 27 W

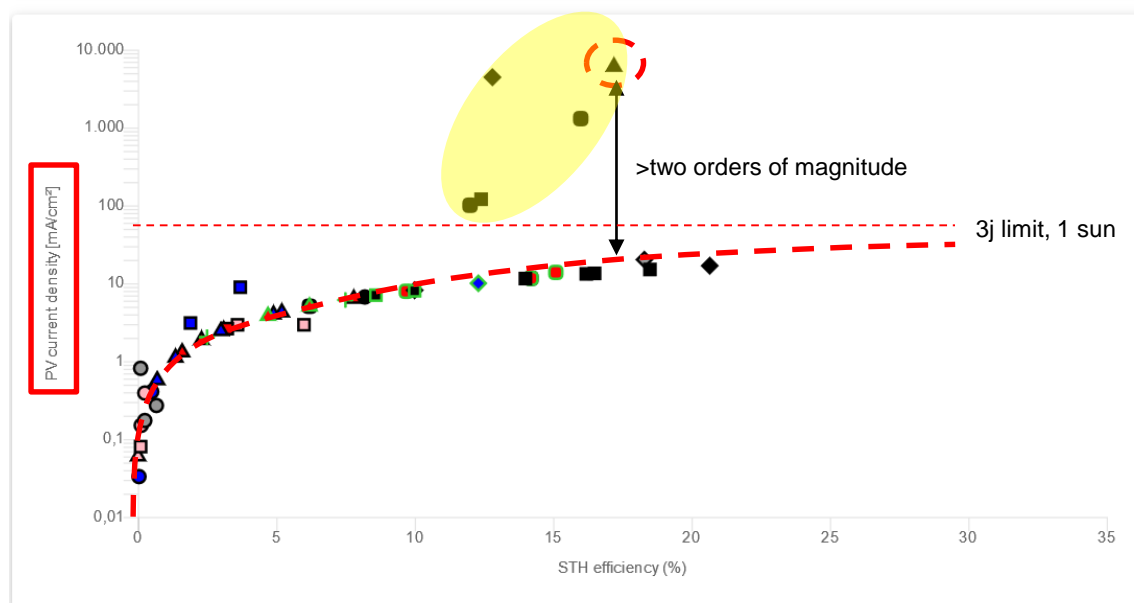
Current density in electrolyzer component: 0.88 A/cm²

Current density in photoabsorber component: 6.04 A/cm²

Efficiency: 17.1% solar-to-fuel

Comparison

- Dynamic and online tool: – <http://specdc.epfl.ch/>



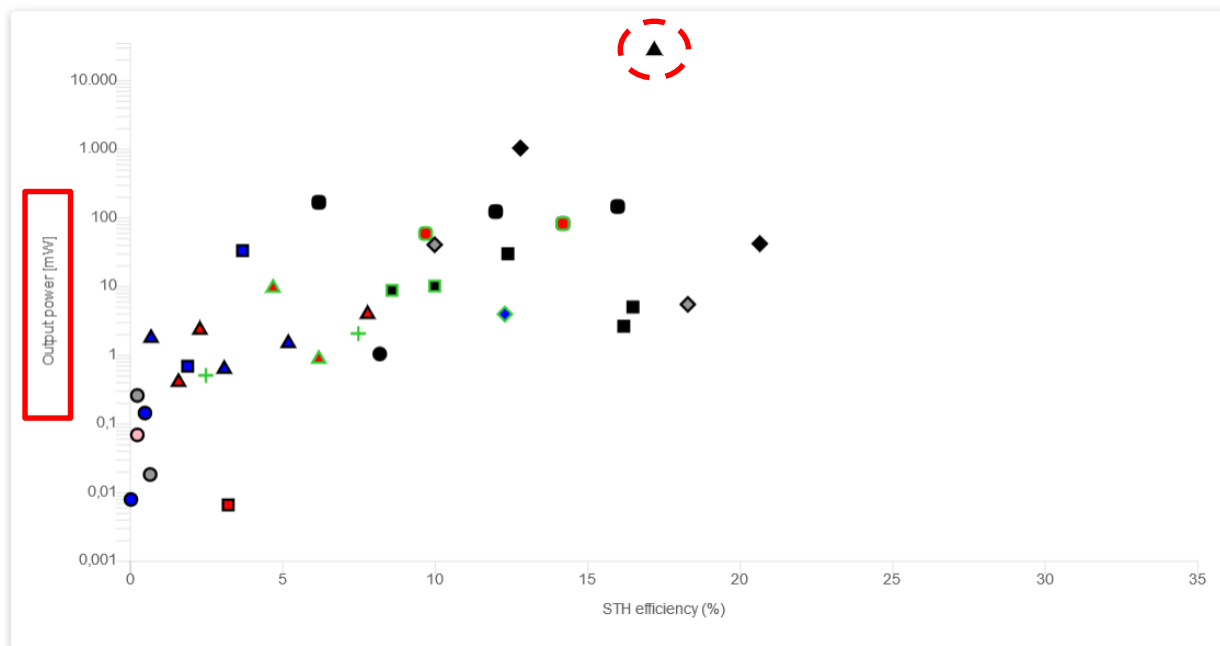
w/o multi-module demonstrations
w/o multiple electrolyzer demonstrations

LEGEND			
Fill color - PV / photoabsorber material	Boundary color - EC material	Symbol shape - PV / photoabsorber and EC configuration	
All III-V	Rare metal-based (expensive)	○ 2J, integrated PVs and catalyst	+ 3J, integrated PVs and catalyst
Partial III-V	Abundant (cheap)	□ 2J, integrated PVs, wired catalyst	△ 3J, integrated PVs, wired catalyst
All Si		◇ 2J, non-integrated PVs or catalyst	○ 3J, non-integrated PVs or catalyst
Partial Si			
Oxides and others			

Tembhurne, Nandjou, Haussener, *Nature Energy*, doi: 10.1038/s41560-019-0373-7, 2019

Comparison

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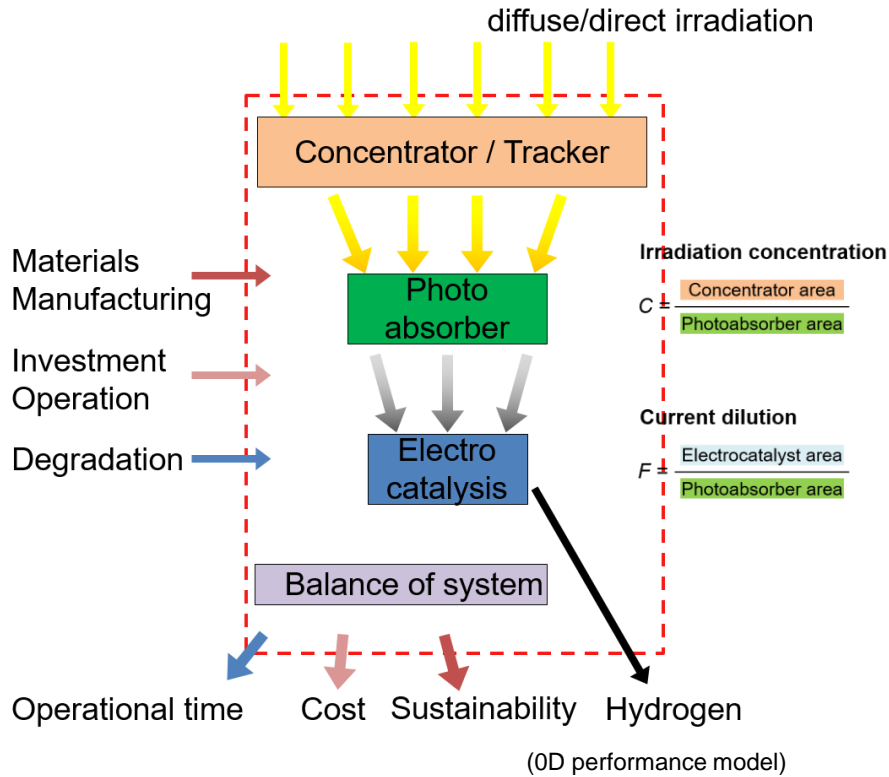
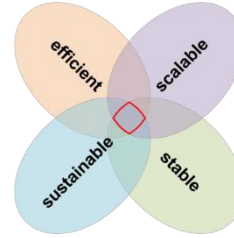
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Partial Si			
Oxides and others			

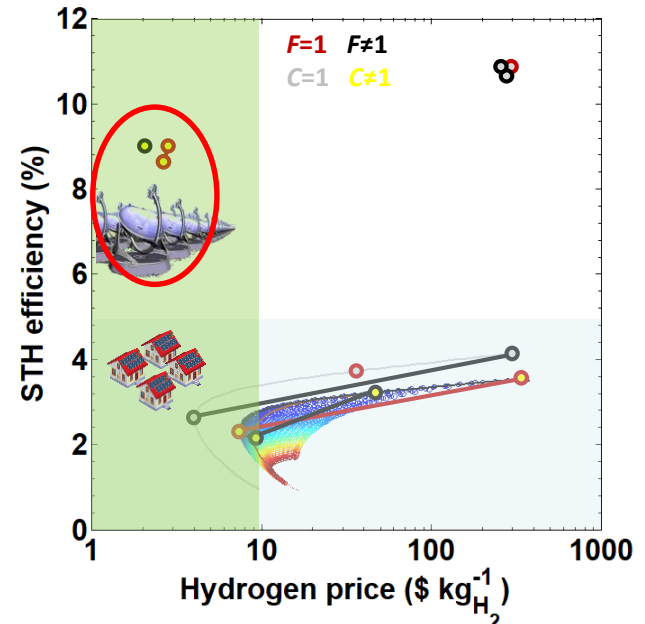
Tembhurne, Nandjou, Haussener, *Nature Energy*, doi: 10.1038/s41560-019-0373-7, 2019

Techno-Economics and LCA

- Promising device design groups



Photoabsorber: Si-V
Catalysts: Ni/Cu-ox



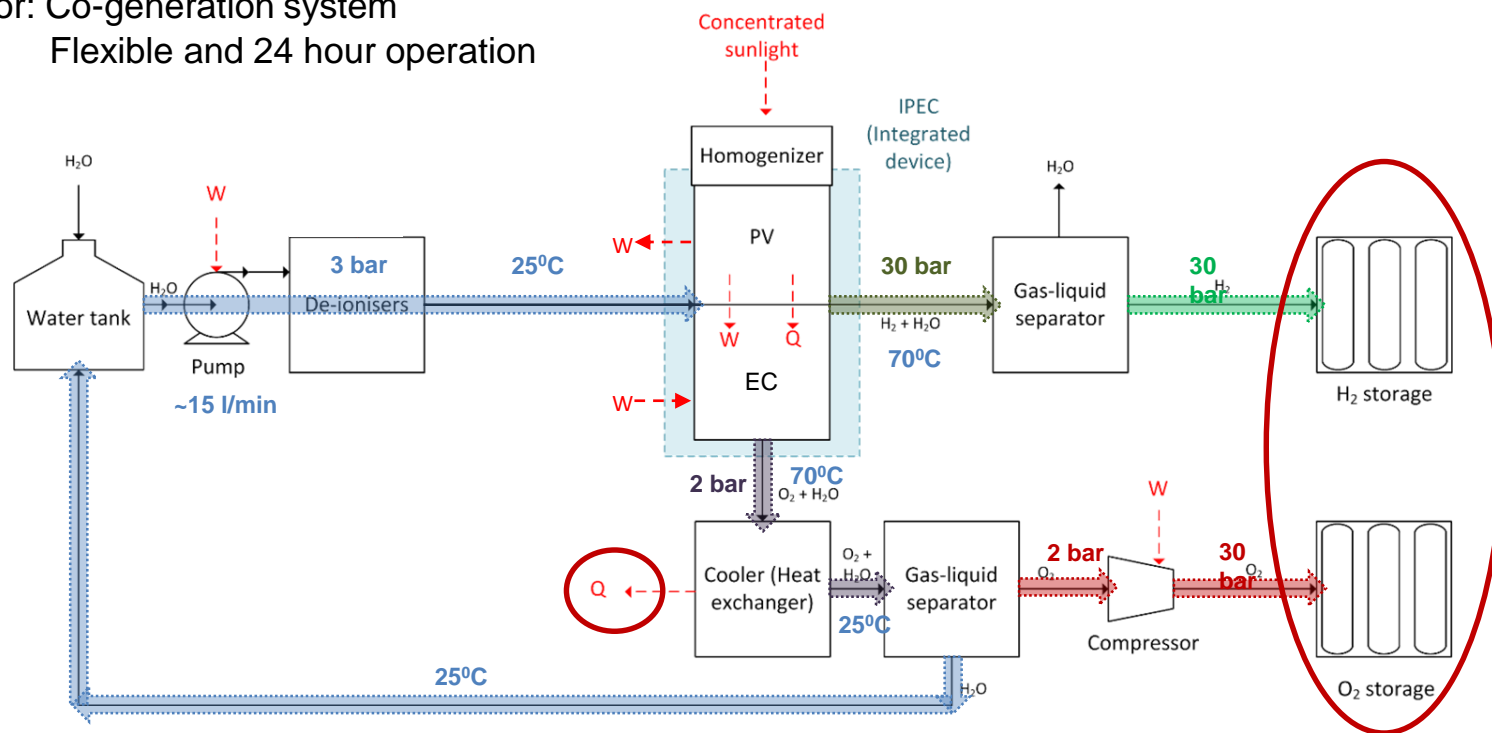
Dumortier, Temburne, Haussener, *Energy Environ Sci*, 8, 2015; <http://specdo.epfl.ch>

EPFL

Target: $\sim 0.5\text{kg H}_2/\text{day}$, kW-scale, long-term, on-sun demonstration

System Process Flow Diagram

Potential for: Co-generation system
Flexible and 24 hour operation

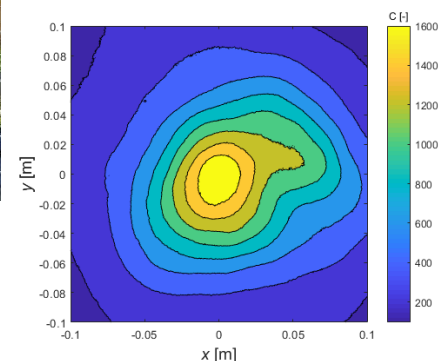
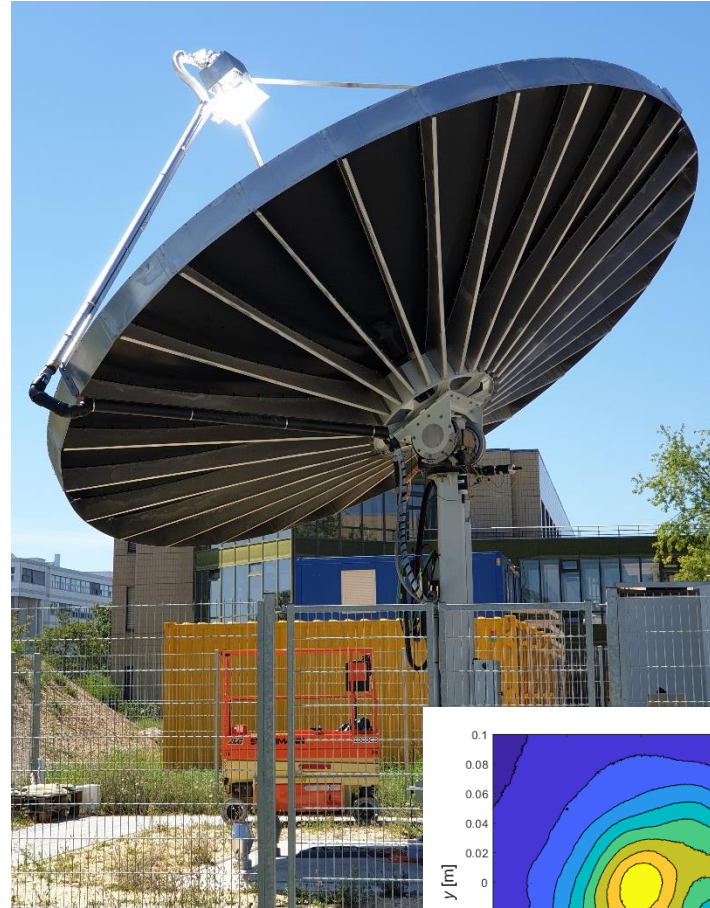


Tembhurne, Holmes-Gentle, Suter, Haussener, Nature Energy, 2023

Reactor and System in Operation

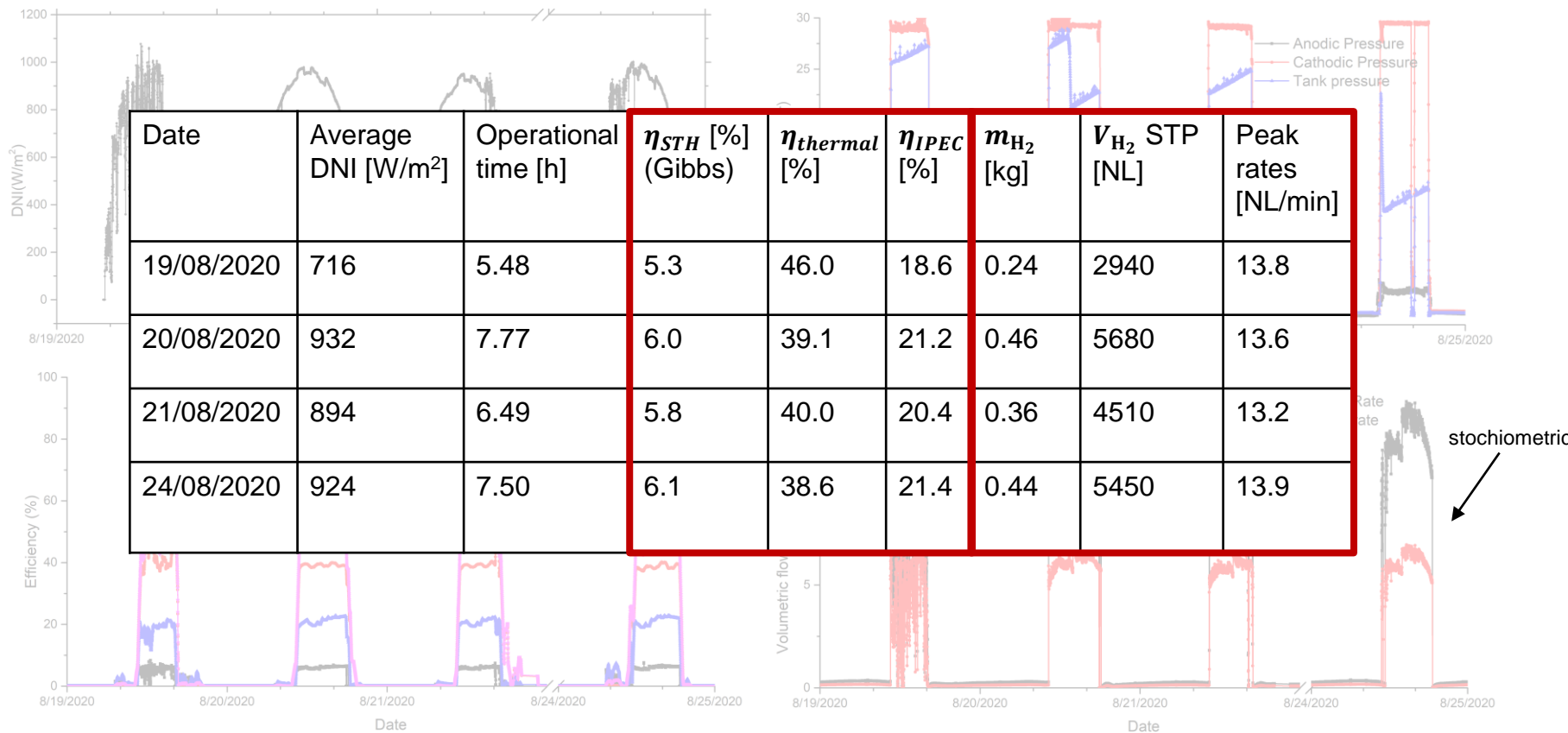


Tembhurne, Holmes-Gentle, Suter, Haussener, in preparation, 2020



Integrated System Test

- Full operation over multiple days in varying meteorological conditions

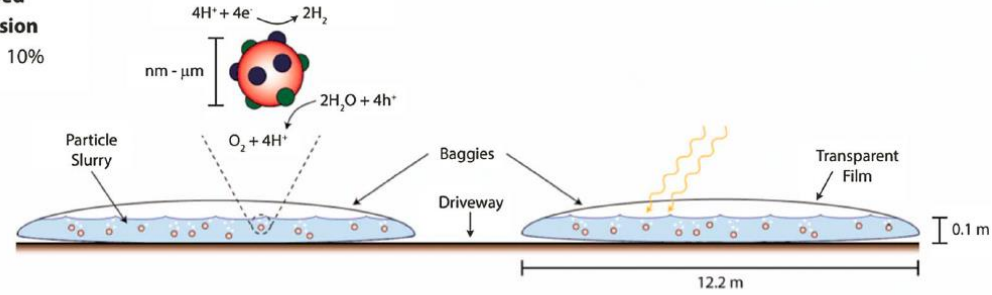


Tembhurne, Holmes-Gentle, Suter, Haussener, in preparation, 2020

Photoelectrochemistry

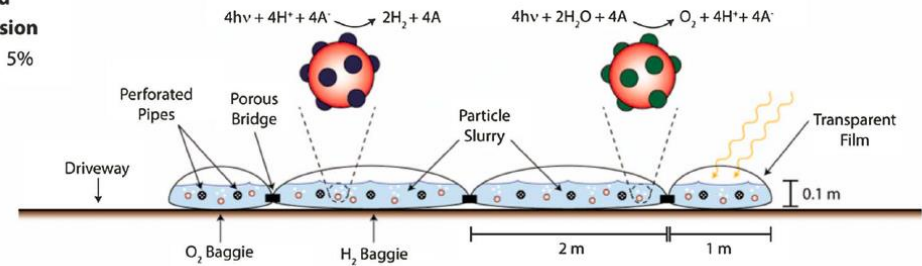
- Proposed devices

Type 1: Single Bed Particle Suspension
STH Efficiency 10%



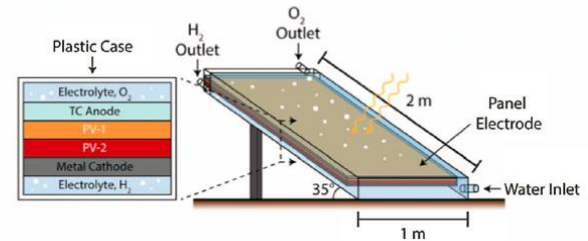
(a)

Type 2: Dual Bed Particle Suspension
STH Efficiency 5%



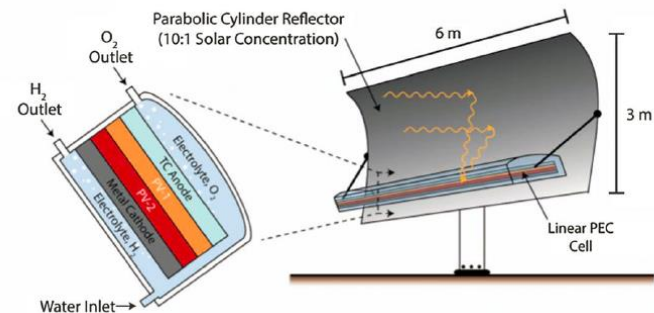
(b)

Type 3: Fixed Panel Array
STH Efficiency 10%

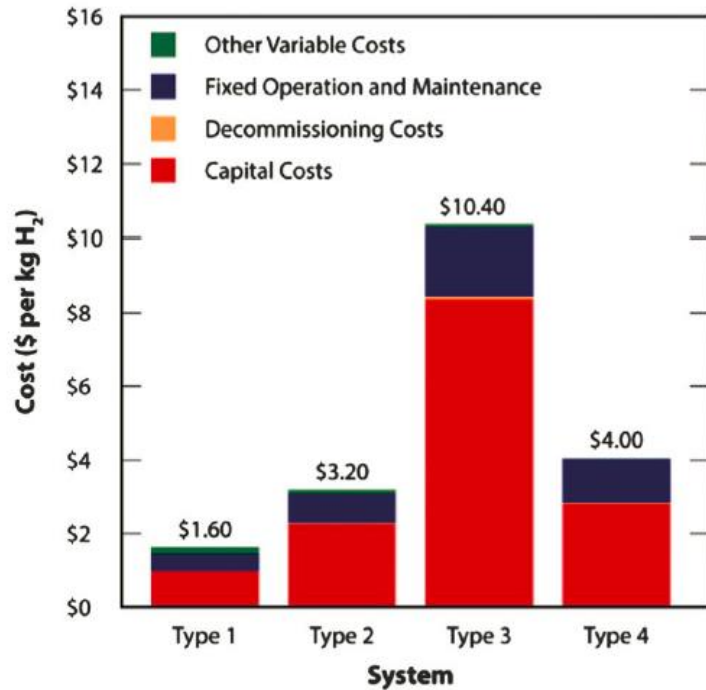


(c)

Type 4: Tracking Concentrator Array
STH Efficiency 15%



(d)

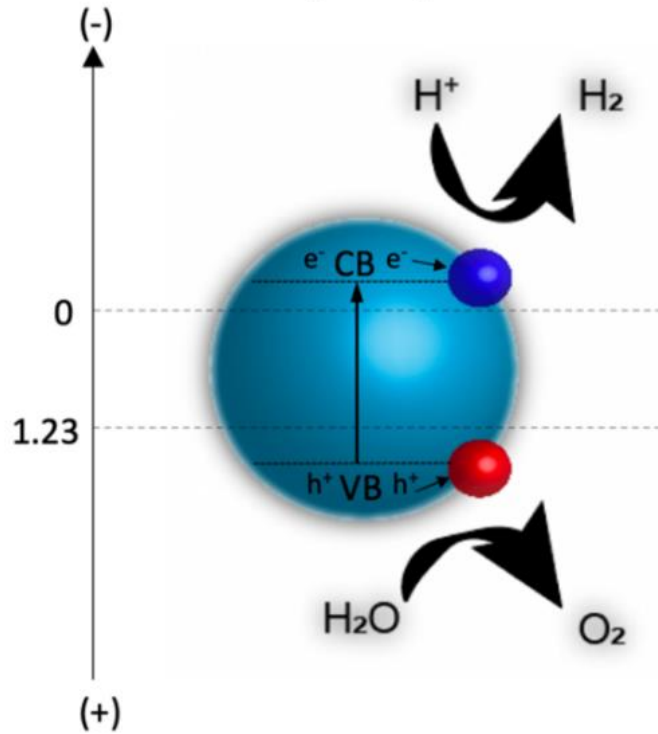


Pinaud et al., EES, 2013.

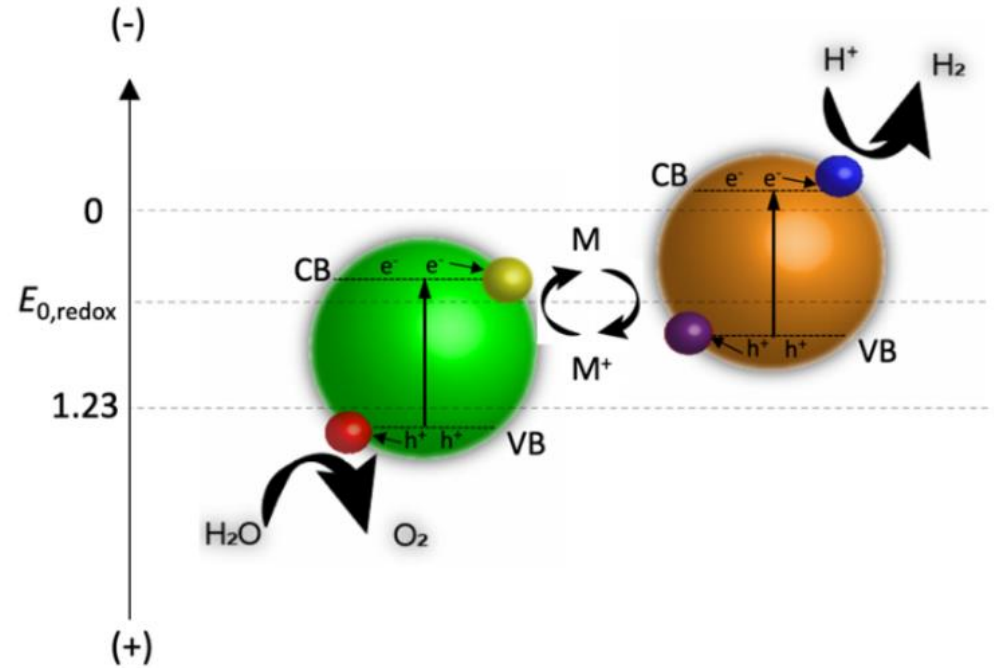
Photocatalysis

- Working principle

Potential V vs NHE (pH = 0)



Potential V vs NHE (pH = 0)

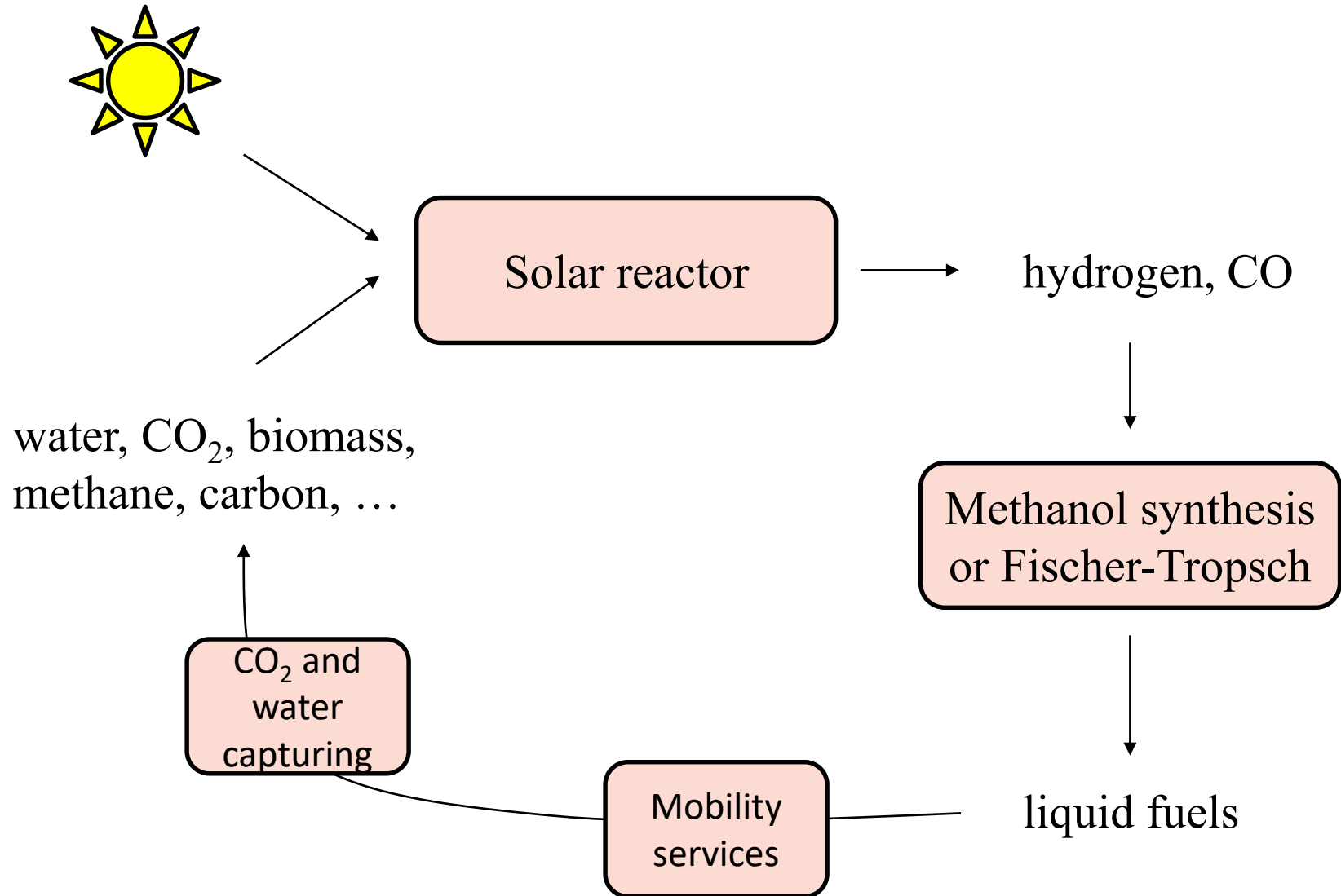


Renewable Energy

- Outline:
 - Conversion pathways solar-to-fuel
 - Hybrid pathways
 - Solar thermochemistry
 - Photochemistry

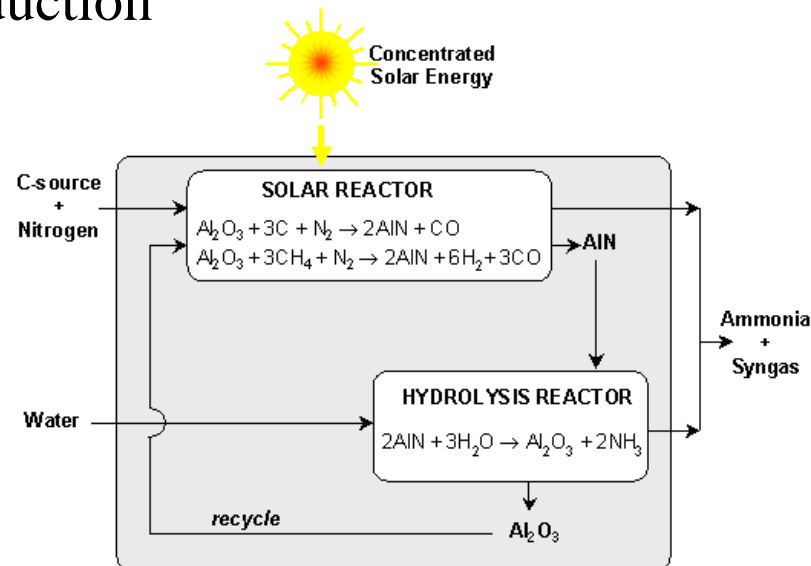
Sustainability issue

- Solar to fuels:



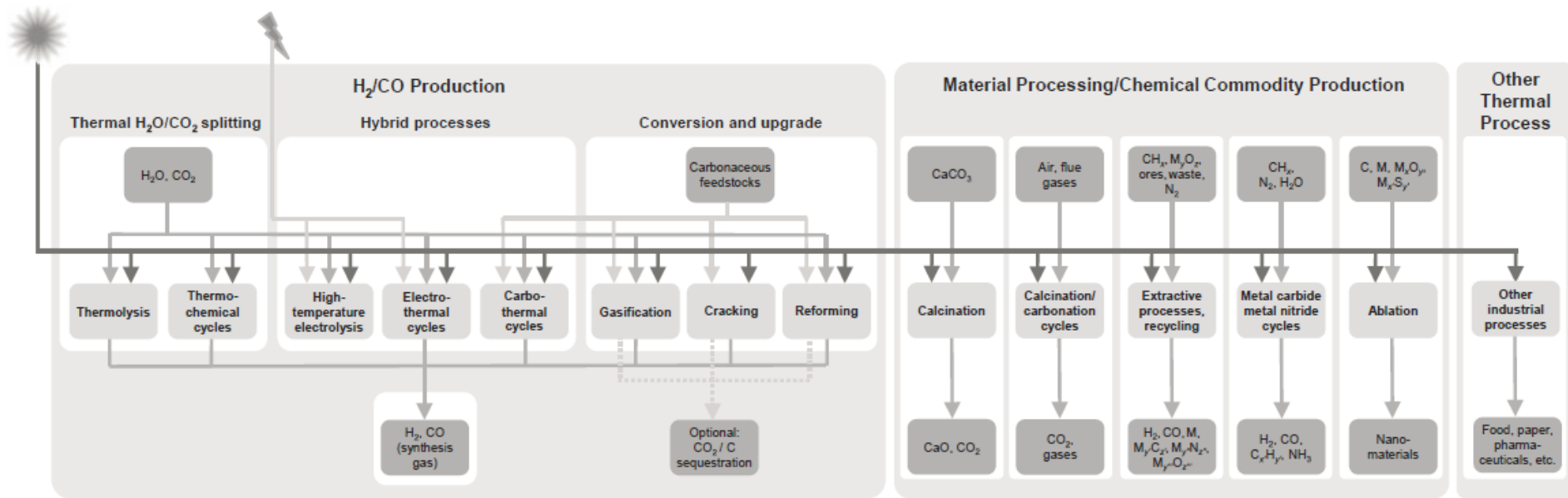
Solar materials

- Solar to materials:
 - In principle any other chemical reaction could be driven by solar thermochemistry or photoelectrochemistry if enthalpy of reaction matches solar irradiation, or equilibrium potential and band edge position matches solar irradiation and material combinations
 - E.g.:
 - Carbothermic reduction of alumina under near vacuum conditions
 - Ammonia production



Solar materials

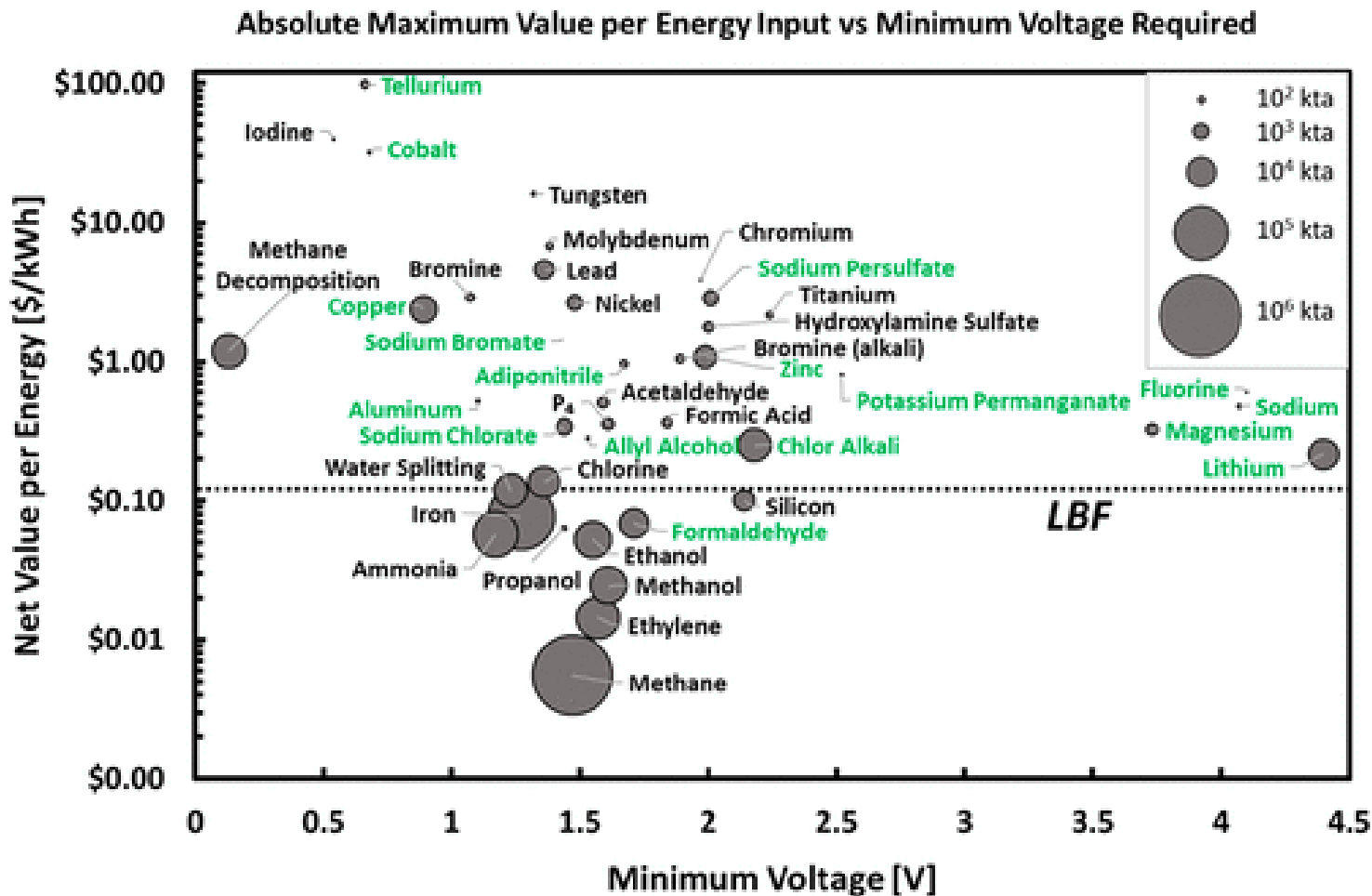
- Thermochemical:



Bader et al., 2016

Solar materials

- (Photo)electrochemical:

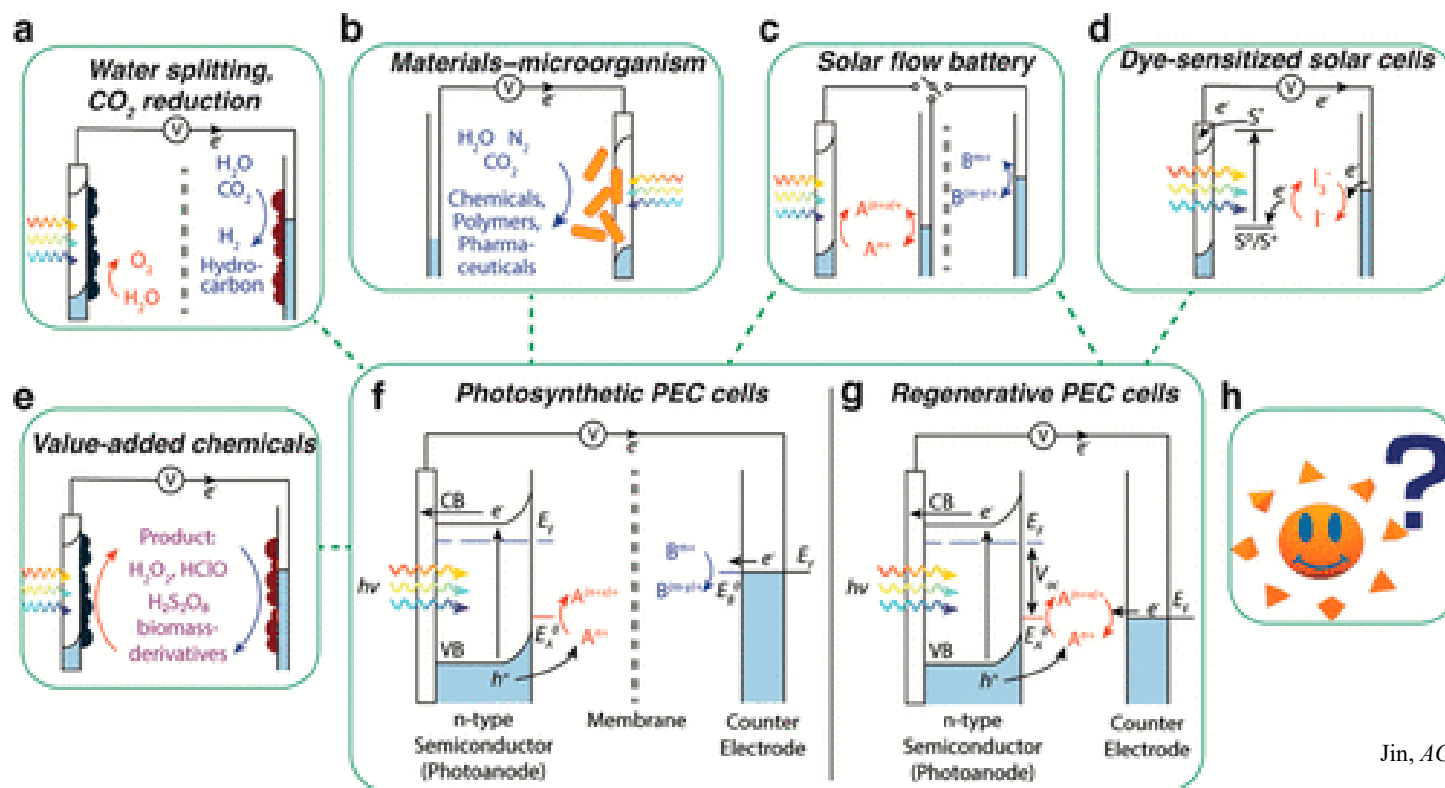


Maximum net value per energy input (log scale) plotted versus minimum voltage required for all electrochemical processes or electrochemical equivalents of thermochemical processes. For each point, the width of the circle corresponds to the relative market size. Processes highlighted in green are conducted electrochemically in industry, to any appreciable extent. The lower bound of feasibility (LBF) is plotted as the horizontal dashed line

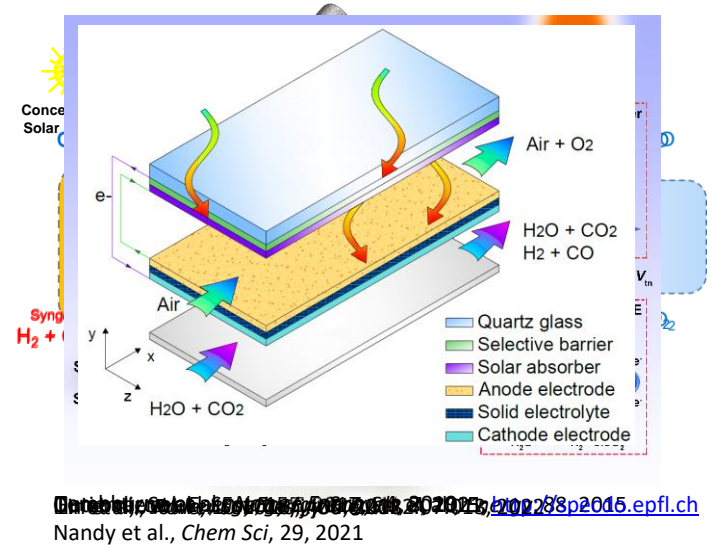
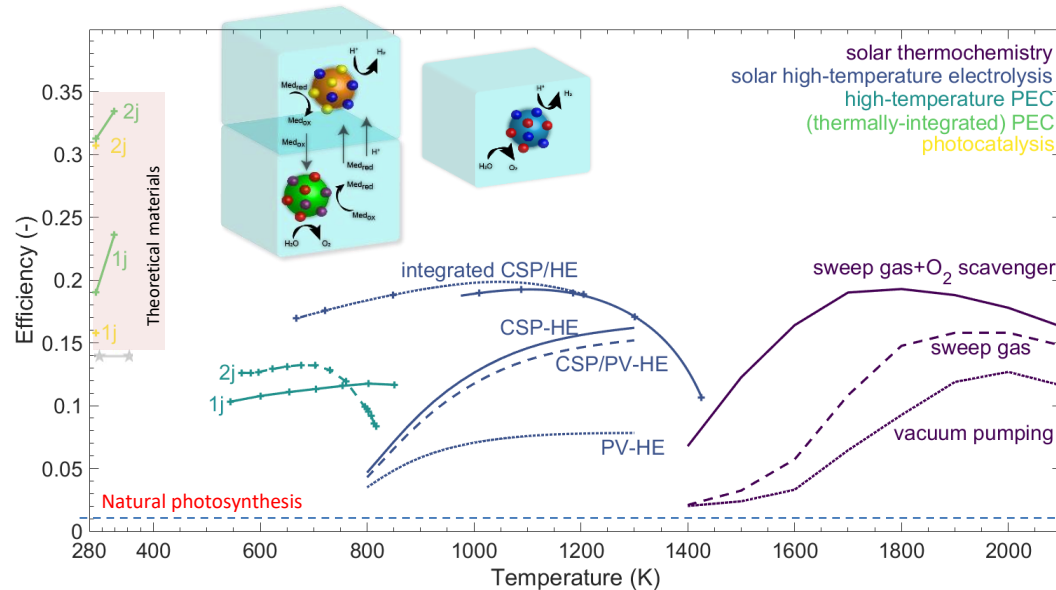
Palmer et al., Technoeconomics of Commodity Chemical Production Using Sunlight, ACS Sustainable Chemistry & Engineering, 2018

Other photo-driven chemistry

- Photo-electro-chemistry – more than fuels:
 - Oxidation of organic matter
(water treatment, air cleaning, medical disinfection, ...)
 - Photo-electrochemical nitrogen reduction
 - Photo-driven flowbatteries (I_3^-/I^- , VO_2^+/VO^{2+} ...)



Efficiency limits – Comparing solar fuel approaches



Comparison:

- Similar achievable STF efficiency for technical pathways
- Lower temperature requirements for HTE
- Less stringent requirement for heat recovery in HTE
- No requirement for p_{O_2} at OER (1 Pa for TCC, Air for HTE)
- But significant, unsolved thermo-mechanical material challenges for HTE
- For PEC: Comparable STF, low temperature, but corrosive environments

Learning outcomes of today's lecture

- Solar fuels:
 - How can solar energy be converted into fuels?
 - What is a hybrid pathway?
 - Why using fossil fuels together with solar energy?
 - What is solar thermochemistry and how can it be used for solar fuel processing?
 - Why is solar water-splitting via multi-step water splitting cycles preferred compared to direct thermolysis?
 - What is photoelectrochemistry and how can it be used for solar fuel processing?
 - What other chemical commodities or materials can be processed using solar energy?