



## Power-to-gas process with high temperature electrolysis and CO<sub>2</sub> methanation

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### ► To cite this version:

Myriam de Saint Jean, Pierre Baurens, Chakib Bouallou. Power-to-gas process with high temperature electrolysis and CO<sub>2</sub> methanation. IRES: International Renewable Energy Storage Conference and Exhibition, Nov 2013, Berlin, Germany. pp.Session E1. cea-00958864

HAL Id: cea-00958864

<https://cea.hal.science/cea-00958864>

Submitted on 13 Mar 2014

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DE LA RECHERCHE À L'INDUSTRIE



# POWER-TO-GAS PROCESS WITH HIGH TEMPERATURE ELECTROLYSIS AND CO<sub>2</sub> METHANATION

**IRES 2013 – Session E1 |**

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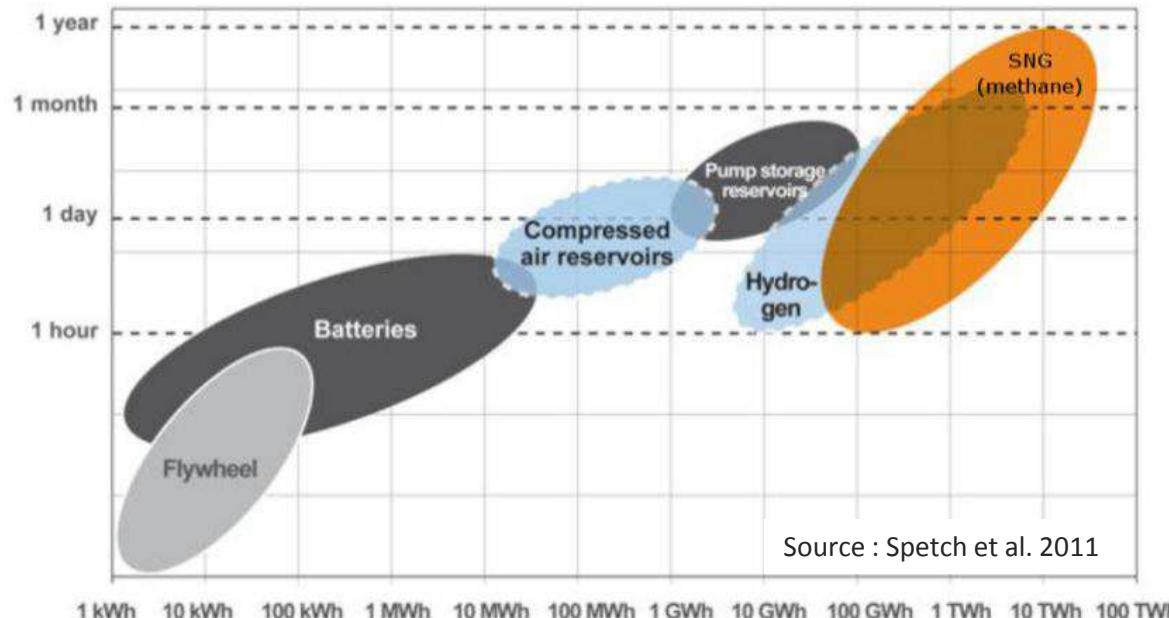
# **ENERGY BACKGROUND**

# POWER-TO-SNG : A SOLUTION FOR ELECTRICITY STORAGE



## Renewable ressource development : 3 issues for transportation and distribution electrical networks

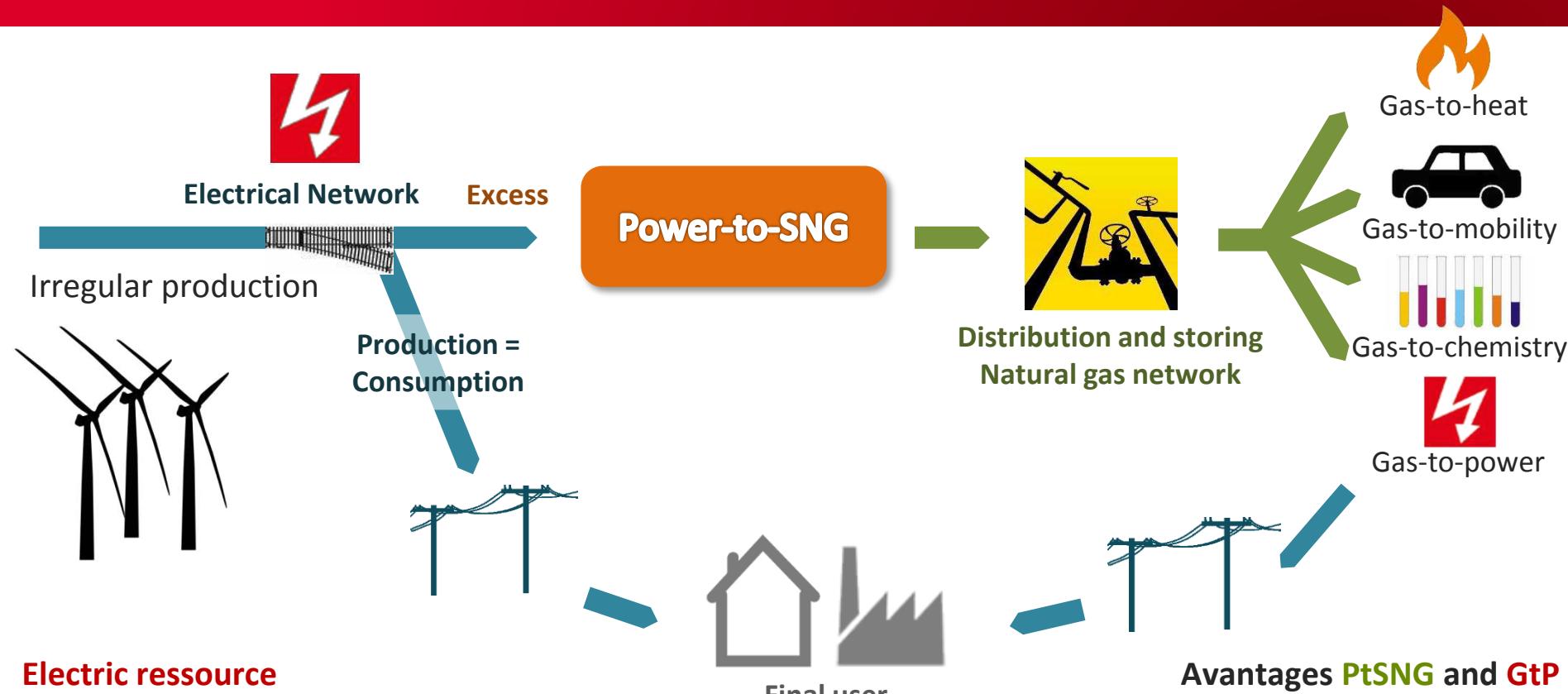
- High consumption periods
- Excess electric production
- Transportation of energy from production areas to consumption areas



**SNG**

Substitute  
Natural Gas  
(methane)

# A LINK BETWEEN TWO NETWORKS



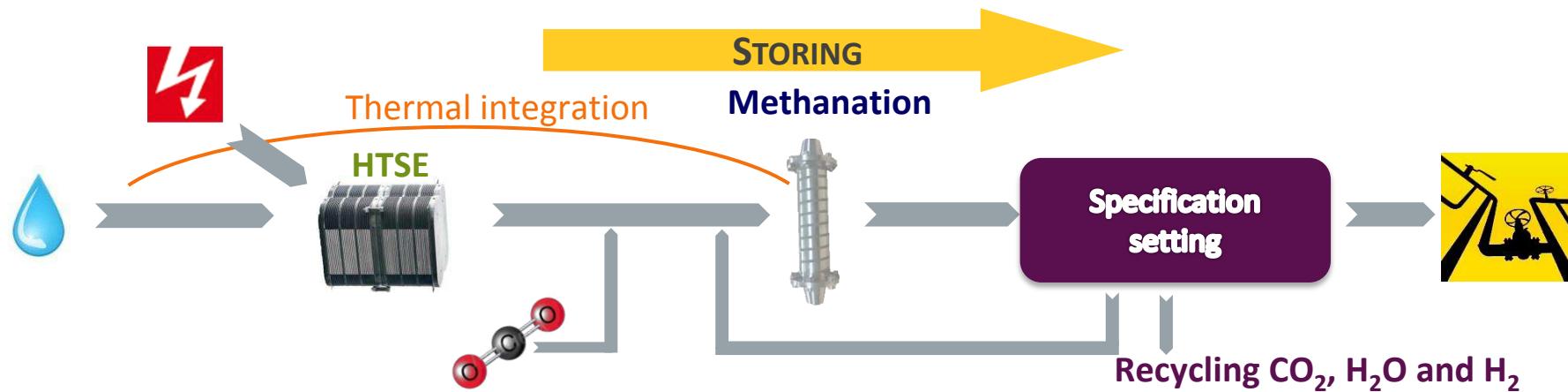
## Electric ressource

- Unstorable
- Irregular production
- Network congestion

- ## Avantages PtSNG and GtP
- Use of existing natural gas network
  - Mid or long term storage
  - Transportation
  - Production of electricity
  - **Connection of the 2 netwoks**

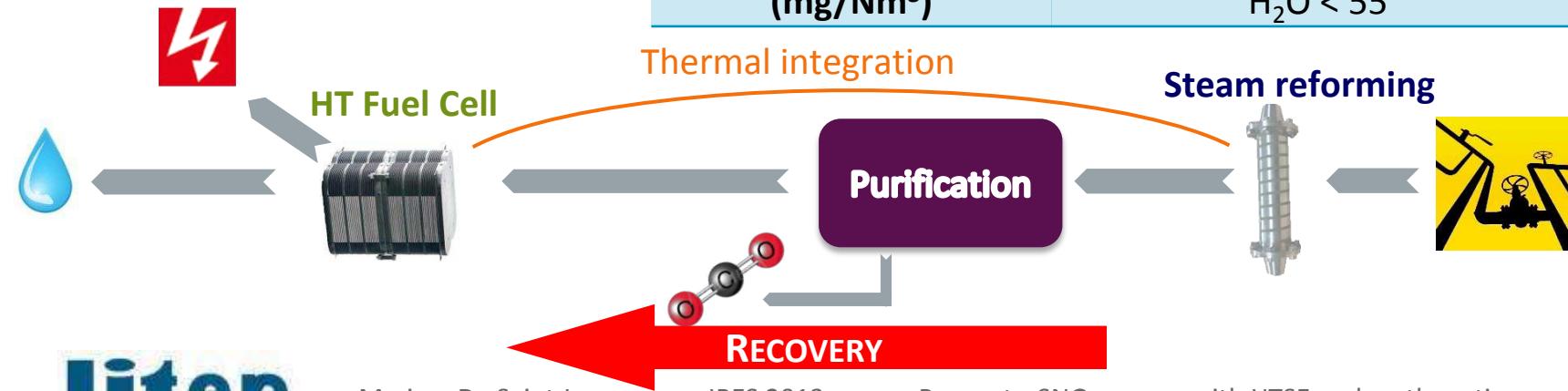
**POWER-TO-SNG PROCESS  
WITH HIGH TEMPERATURE STEAM  
ELECTROLYSIS AND CO<sub>2</sub> METHANATION**

# STUDIED POWER-TO-SNG PROCESS ARCHITECTURE

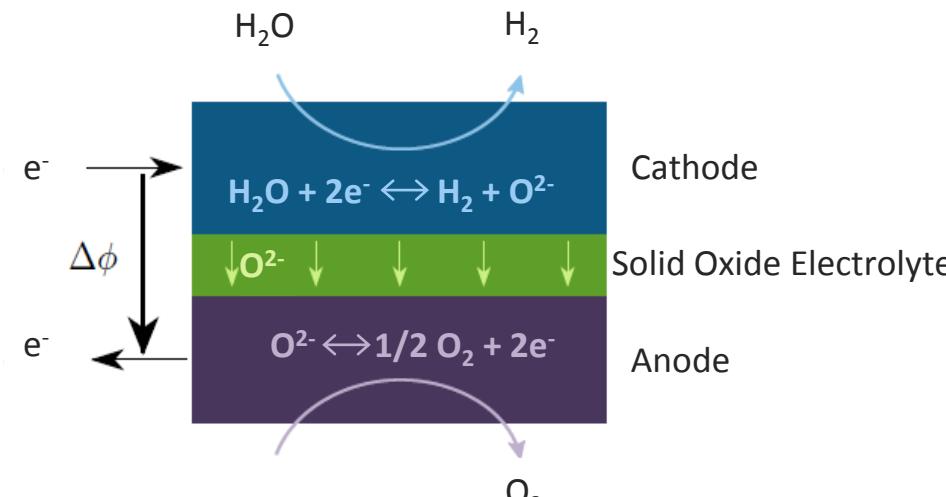


Wobbe index

$$W = \frac{HHV}{\sqrt{\rho}}$$



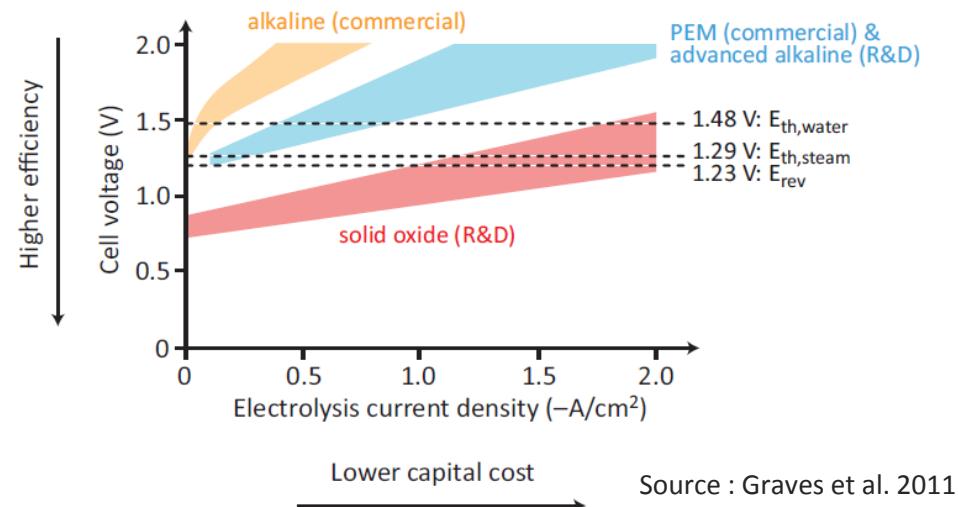
# HIGH TEMPERATURE STEAM ELECTROLYSIS



$T = 1073 \text{ K}$

## HTSE avantages

- High temperature :  $\Delta H$  decrease
- Irreversibility decrease
- High efficiency
- Reversible (SOEC / SOFC techno)
- Thermal behaviours : Exo, auto et endothermal
- Reactants :  $\text{H}_2\text{O}$  and / or  $\text{CO}_2$  : co-electrolysis



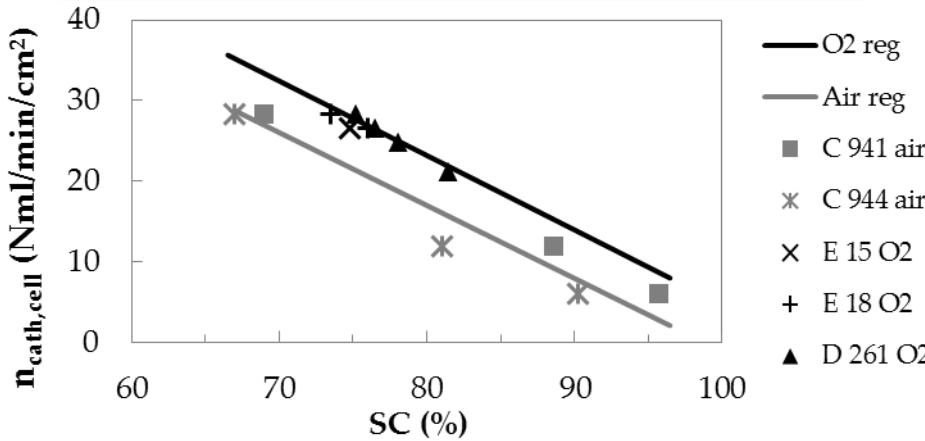
## HTSE current limitations

- R&D
- Cost
- Long-term degradation of performances

# SIMULATION : HTSE MODELLING AND VALIDATION

## HTSE Modelling : To determine $P_{elec}$ and $N_{cell}$ for a incoming flow

- SOEC technology
- $U_{op} = U_{tn}$  and SC : fixed values
- Molar and energy balances :  $P_{elec}$
- Electrochemical modelling : exp. law
- Determination of  $N_{cell}$  (and  $j$ )
- Correction with pressure and stack effects



Experimental data and interpolated law linking  $\dot{n}_{cath,cell}$  and SC for  $T = 1073$  K,  $P = 1$  bar,  $U_{op} = U_{tn}$ ,  $H_2 / H_2O = 10 / 90$ , on cells referenced C 941, C 944, D 261, E 15 et E 16.

## Experimental and phenomenological laws

$$\dot{n}_{cath, cell} = -0,829 \text{ SC} + 83,2$$

$$\dot{n}_{cath, cell} = -0,727 \text{ SC} + 81,8$$

with air sweep

with  $O_2$  sweep

$$R_{eq} = (U_{op} - U_{Nernst})/j$$

$\Omega \cdot cm^2$

## Pressure effect and stack effect

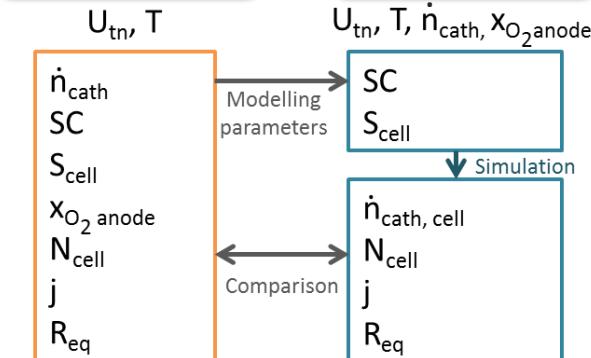
$$R_{eq} = R_{eq, P^0} P^{-0,09}$$

for  $P$  [1;10 bar]

$$R_{eq, Stack} = (R_{eq, cell} + 0,034) N_{cell}$$

## Experimental data

## HTSE modelling



Modelling : Calculation of  $j$  and  $N_{cell}$  with errors up to 40% → cell dispersion effect

# CO<sub>2</sub> METHANATION

## Sabatier reaction



## RWGS reaction



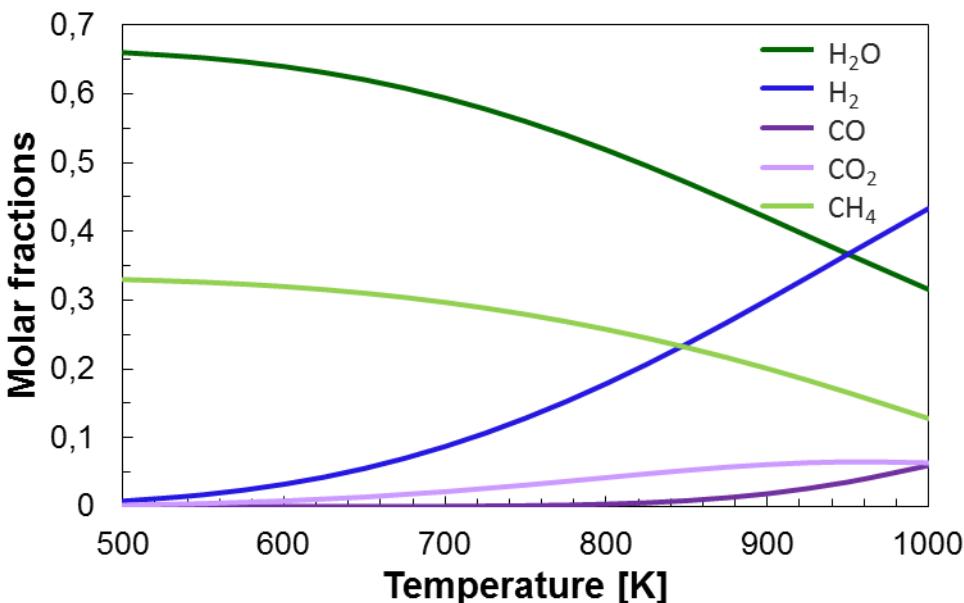
## CO methanation



## Carbon craking



- Catalysed reaction
- Favorable operating conditions for CH<sub>4</sub> production : P ↗ et T ↘



Equilibrium at P = 15 bar for H<sub>2</sub>/CO<sub>2</sub> = 4

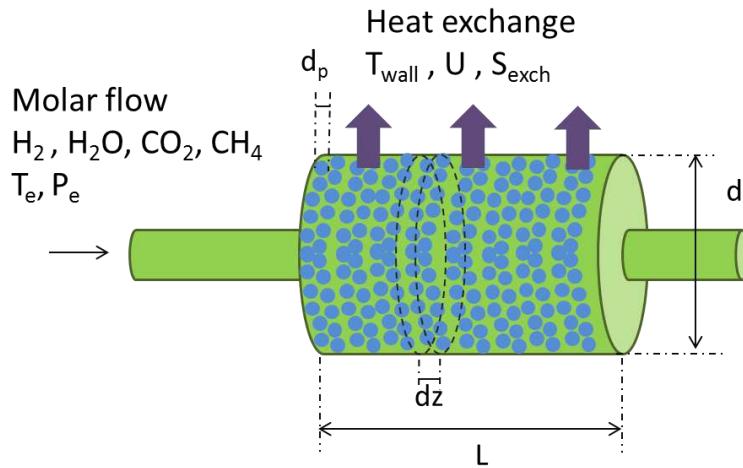
## Avantages of CO<sub>2</sub> methanation

- No CO at moderate T
- High CH<sub>4</sub> selectivity
- Exothermal reaction
- High conversion yield
- Existing catalysts

## Current limitations of CO<sub>2</sub> methanation

- Poor literature on kinetic laws
- Not a lot of experimental data published, preference given to syngas (CO + H<sub>2</sub>) methanation

## SIMULATION : METHANATION MODELLING



$$r \left[ mol.s^{-1}.m^{-3} \right] = 2691.7.10^3 e^{-64121/RT} \left( P_{CO_2}^n P_{H_2}^{4n} - \frac{P_{H_2O}^{2n} P_{CH_4}^n}{K_{eq}(T)^n} \right)$$

$$K_{eq}(T) = \exp \left( \frac{28183}{T^2} + \frac{17430}{T} - 8.254 \ln T + 2.87.10^{-3} T + 33.17 \right)$$

Pressure (bar)	1	2	30
n	0.225	0.5	1

Plug-flow reactor with fixed-bed catalyst and boundary conditions

### Methanation modelling

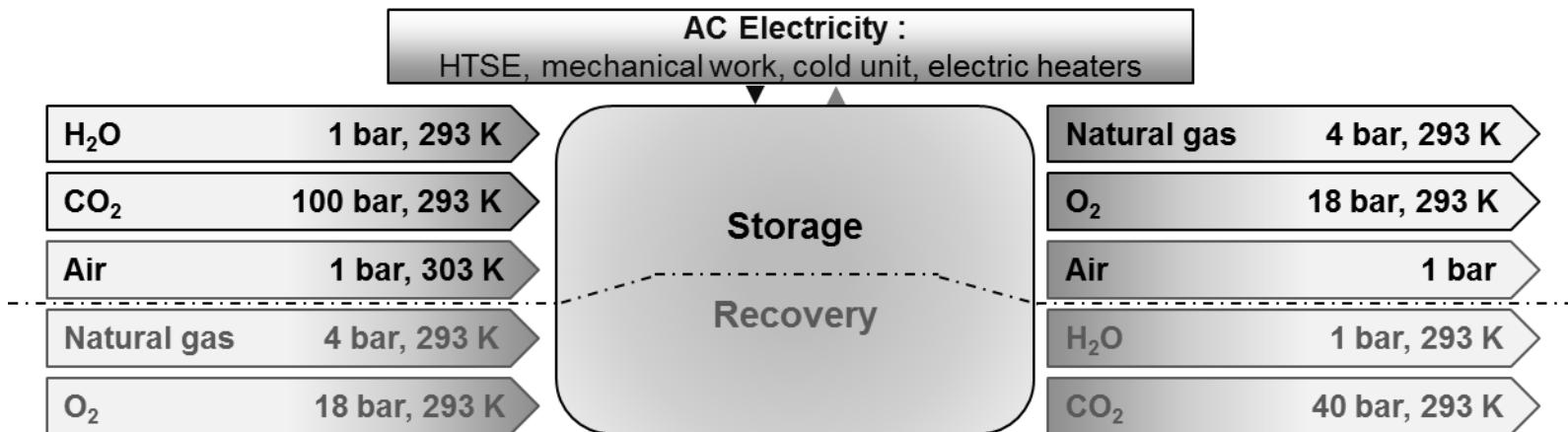
$$CO_2 + 4 H_2 \leftrightarrow CH_4 + 2 H_2O$$

- 1D plug-flow reactor modelling
- Kinetic law (cat Ru)
- Pressure  $\approx 16$  bar
- Adiabatic behaviour
- Inlet temperature = 573 K
- Outlet temperature < 973 K

Kinetic law from literature (Cat Ru) [Lunde 1974, Ohya 1997]

Simulation and experimentation agreement  
for  $n = 0.5$  ( $P = 2$  bar) for  $P_{exp} \in [3.4 ; 7]$   
Higher  $P$ , lower gap between simulation and experimentation,  $\forall n$  used

# SIMULATION : PERIMETER AND HYPOTHESES



**Pinch analysis module  
Process thermal integration**

$$\eta_{AC/DC} = 0.92$$

$$\Delta P_{hexch} = 0.2 \text{ bar}$$

$$\Delta T_{hexch} = 100-150 \text{ K}$$

$$\text{H}_2/\text{H}_2\text{O}_{HTSE} = 1/9$$

$$\text{H}_2/\text{CO}_2_{meth} = 1/4$$

**Cold Utility**

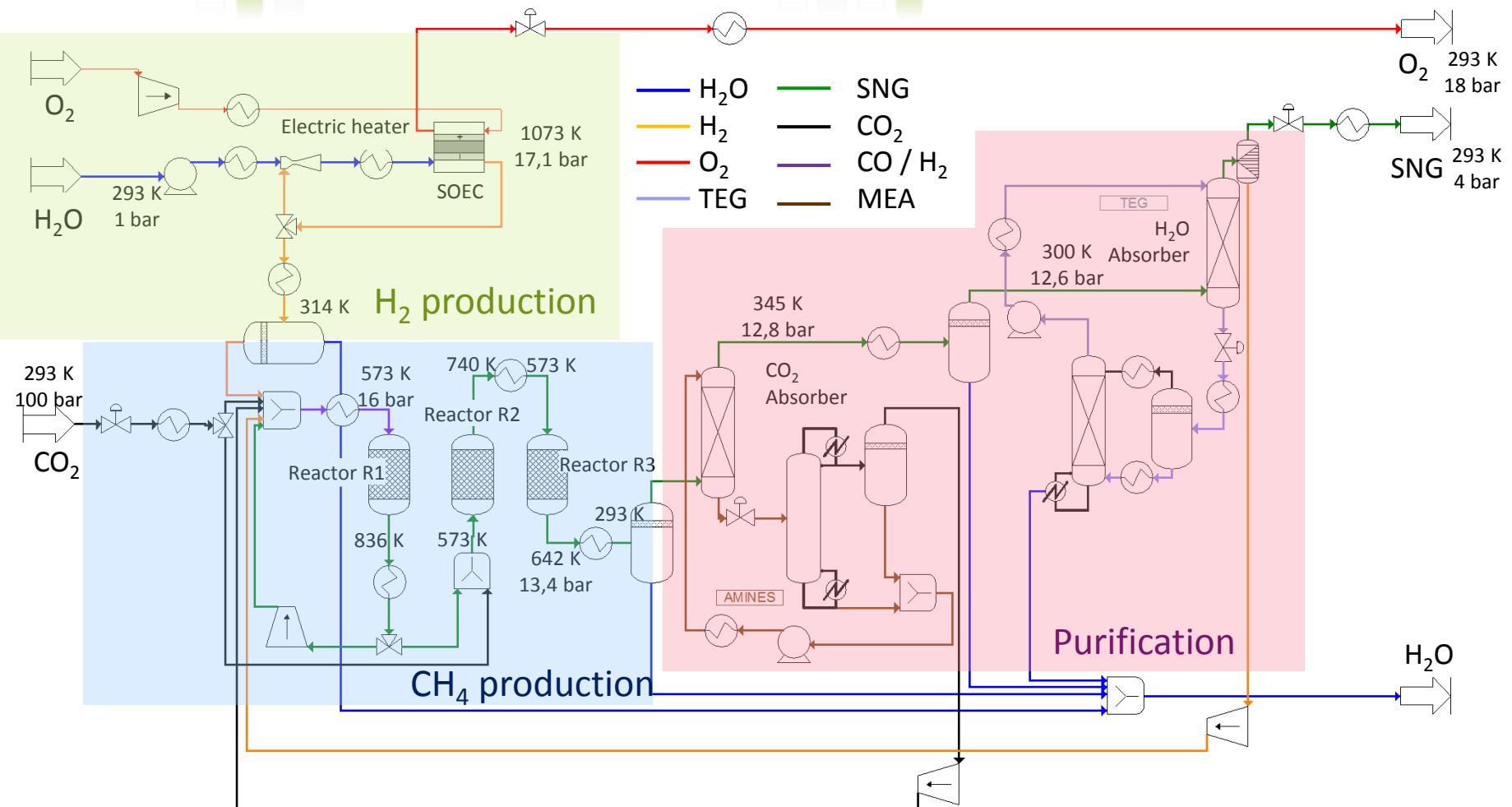
Cold unit (273 K)

$EER_{elec} = 1.73$

**Hot Utility**

Electric heaters

$\eta = 0.90$



### Energy Efficiency

$$\eta = \frac{\dot{n}_{SNG} HHV_{SNG}}{P_{elec, HTSE} + P_{elec, mech} + P_{elec, hot} + P_{elec, cold}}$$

### Theoretical Energy Efficiency

$$\eta = \frac{\dot{n}_{SNG} HHV_{SNG}}{P_{elec, HTSE}} = 0.89$$

## **SIMULATION RESULTS & CONCLUSION**

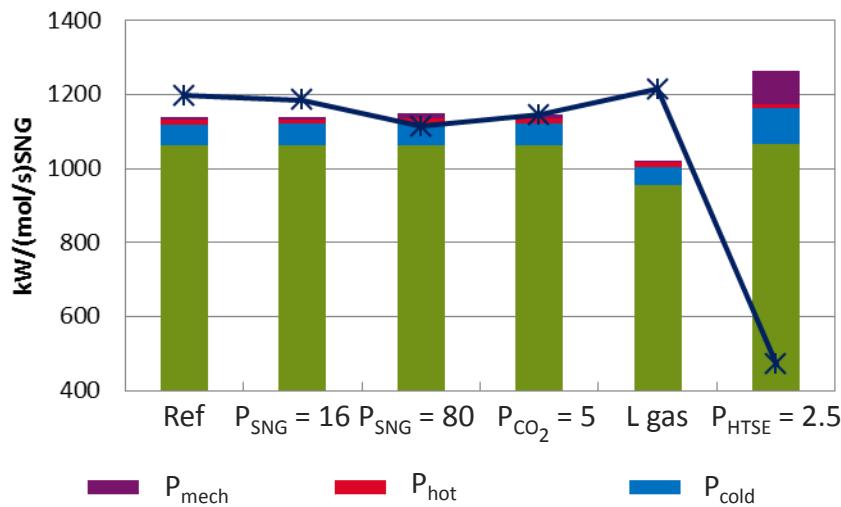
# SIMULATION : PARAMETRIC STUDY

$U_{op} = U_{tn}$ , SC = 75%,  $H_2/CO_2 = 4$ ,  $P_{meth} = 16$  bar,  $T_{HTSE} = 1073$  K

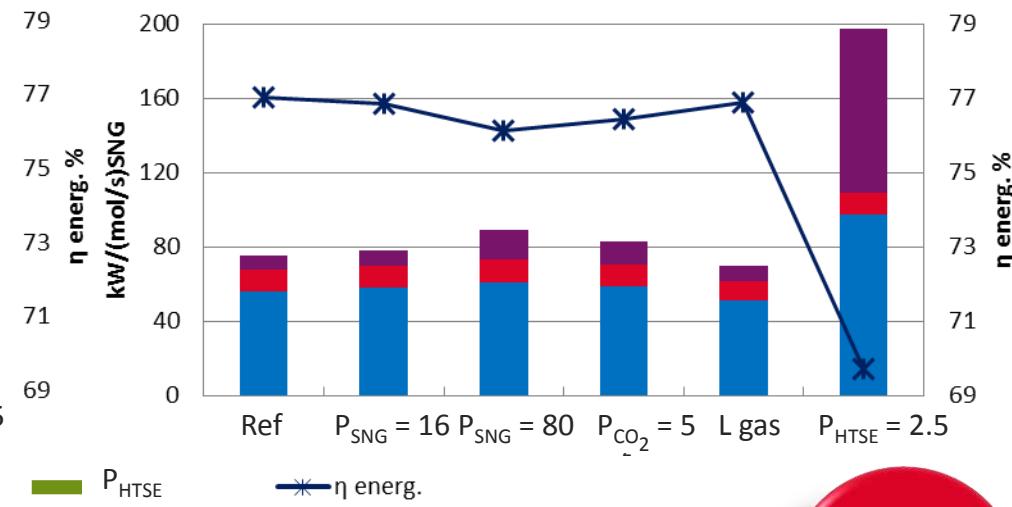
Reference case :  $P_{CO_2} = 100$  bar  $P_{SNG} = 4$  bar  $P_{HTSE} = 17$  bar H gas

Sensitivity :  $P_{CO_2} = 5$  bar  $P_{SNG} = 16$  and  $80$  bar  $P_{HTSE} = 2.5$  bar B gas

Electricity consumption



Electricity consumption except HTSE



- Injection on H or L gas network : no influence on energy efficiency  $\eta$
- Kind of network (transportation or distribution): high influence on  $\eta$
- $CO_2$  origine (separation or storage) : high influence on  $\eta$
- $P_{HTSE}$  : very high influence on  $\eta$  : loss of 7.4 pts (9.6%) regarding ref. case

$\eta_{HP} \approx 77\%$

$\eta_{LP} \approx 70\%$

# CONCLUSION

## Modelling

HTSE modelling  
for sizing with  
experimental law

Adequation  
between  
modelling results  
and experimental  
data

Kinetic law and  
modelling of  
methanation

Adequation  
between  
simulation results  
and observed  
performances

Scale-up of  
methanation  
stage

Purification of  
produced SNG

## Process simulation

Production of  
SNG matching  
with the  
specifications

Two gas qualities  
(H and L) are  
achievable

Higher  
efficiency if

The process  
is operated  
at high  
pressure

CO<sub>2</sub> from  
industrial  
storage is  
used



# Thank you for your attention

**POWER-TO-GAS PROCESS WITH  
HIGH TEMPERATURE ELECTROLYSIS AND METHANATION**



IRES 2013 |

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