COLDSPARK DRIVEN ENERGY AND COST-EFFICIENT METHANE CRACKING FOR HYDROGEN PRODUCTION

D5.1 Aspen/HYSYS models for lab and large-scale prototype based on results from WP1, WP2 and WP4

ColdSpark project partner	NORCE
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Deliverable number	5.1 - Basic Aspen/HYSYS models for lab and large-scale prototypes based on results from WP1 and WP2, WP4.
Description	Aspen/HYSYS models for lab and large-scale prototype are established for the ColdSpark® pilot testing and to support techno-economic / LCA analysis. The models are based on results from WP1, WP2 and WP4.
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Review History

Version	Date	Reviewer	Short Description of Changes
1	28.11.2023	Xin Tu, University of Liverpool (UoL) Sachin Maruti Chavan, University of Stavanger (UiS) Remya Ravindran Nair, SEID AS	Proposed use of input from WP1,2,4, and data as the basis for the model upgrade
2	05.12.2023	Sachin Maruti Chavan Remya Ravindran Nair	Updated according to the agreed report scope and input from WP1,2,4 with the project coordinator and WP leaders.
3	07.12.2023	Remya Ravindran Nair	General Review

Document Approval

Name	Role	Action	Date
Terje Hauan	Project Coordinator	*Approved*	08.12.2023

Nature of the deliverable

R	Document, report (excluding the periodic and final reports)	☒
DEM	Demonstrator, pilot, prototype, plan designs	☐
DEC	Websites, patents filing, press & media actions, videos, etc.	☐
DATA	Data sets, microdata, etc.	☐

DMP	Data management plan	☐
Ethics	Deliverables related to ethics issues.	☐
SECURITY	Deliverables related to security issues	☐
Other	Software, technical diagram, algorithms, models, etc.	☐

Dissemination level

PU	Public — fully open (automatically posted online on the Project Results platforms)	☒
SEN	Sensitive — limited under the conditions of the Grant Agreement	☐

ACKNOWLEDGEMENT

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The ColdSpark® project will validate a novel non-thermal plasma technology to produce hydrogen at an industrial scale from methane, with a process energy efficiency of 79%, achieving a conversion rate of 85% aiming at zero CO_2 emissions. This will be achieved by designing an industrial-relevant reactor that leverages the best features of the non-thermal plasma technologies, gliding arc and corona discharge, to ensure high efficiency and scalability. The innovation addresses for the first time the critical step of matching the reactor with a pulsed power supply. It enables a perfect fine-tuning of the cracking process parameters, to find the right electron density and energy distribution in the plasma reactor, to maximise energy efficiency. The up-and downstream gas management will be optimised to further contribute to the system's compatibility with the existing infrastructure. The project will develop and test a novel plasma reactor at a lab scale and validate it in conjunction with the power supply at a large scale, pursuing the industry's most power-efficient generation of hydrogen alongside high-value carbon. The technology will assess its application for both, natural gas and biomethane producers. A low energy cost (< 15 kWh/kg H_2 produced) without the need for catalysts and water, makes the proposed solution the most cost-competitive, environment-friendly, and less complex to implement. The reactor design and modularity bring lower CAPEX and OPEX and make it easily scalable and flexible. The project gathers the expertise of a mix of academic, research, and industrial partners from five countries, which bring both outstanding research and topic competence, as well as knowledge and access to the solution for end-user industries.

ColdSpark® is built on a strong consortium of 7 partners from Norway, Spain, Bulgaria, Germany, and the UK with SEID AS as a Coordinator.

More information about the project can be found at: http://www.ColdSpark.eu/

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EXECUTIVE SUMMARY

This report summarises the work performed from "WP5 – Desktop modelling of the definition of industrial set up". The main objective of this work package is to establish a framework and test on relevant models representing the planned experiments at both lab bench scale and large scale. Also, an upscaled model to support TEA/LCA/LCC analysis.

A HYSYS model has been created to serve as a basis for designing and scaling up experiments in the project. Preliminary work and results from ***WP1 - Development and optimisation of plasma methane cracking process at lab scale, WP2 - Gas separation and management of impurities, and WP4 - System components integration***, testing and validation have been considered for the models at various scales, and for upscaling and tuning the HYSYS models. In addition, an industrial scale model with simplified upscaling is used to give first-order estimates as input for LCA analysis (***WP6 - Sustainability and techno-economic assessment of plasma methane cracking process***).

This document describes the main features of the HYSYS model, based on preliminary laboratory observations and recommended design criteria for the planned large-scale testing in the future. More test data is needed to tune the models to real experiments at an appropriate scale and the final process design, especially when it comes to the gas feedback loop and energy consumption for the various plasma reactors and optimised H_2 purification process steps.

ABBREVIATIONS

FEED	Front End Engineering Design
LCA	Life Cycle Analysis
LCC	Life Cycle Cost
PFD	Process Flow Diagram
PSA	Pressure Swing Adsorption
UiS	University of Stavanger
UoL	University of Liverpool

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1 INTRODUCTION

The main objective of “***WP5 – Desktop modelling of definition of industrial set up***” is to create an industrial desktop front end engineering design (FEED) to validate the industrial feasibility and use model data.

This objective requires the following secondary objectives to be achieved within the project:

- SO1: Validate and fine tune the Aspen/HYSYS model using the testing results from WP4
- SO2: Upscale the design of Aspen/HYSYS model x10, optimising the system for its energy and material consumption.
- SO3: Study the potential of the industrial plant for the production of carbon products (nanotube, powder, etc…)
- SO4: Refinement of iterative modelling in interaction with the results from TEA/LCA/LCC (WP5)

This report contains the initial step to establish the framework consisting of modelling methodology and representative models to support the project, in particular the large-scale testing (WP4) and the TEA/LCA/LCC analysis (WP6). Several cases which best represent the already existing and planned laboratory setups for the project have been used to check the capability and flexibility of the modelling framework, and to allow tuning with experimental data. The final results will be based on the experimental data from ***WP1 - Development and optimisation of plasma methane cracking process at lab scale, WP2 - Gas separation and management of impurities, and WP4 - System components integration***, and will be reported in the planned deliverables ***5.2 – Aspen/ HYSYS model for or industrial scale fine-tuned with long test campaign*** in the future.

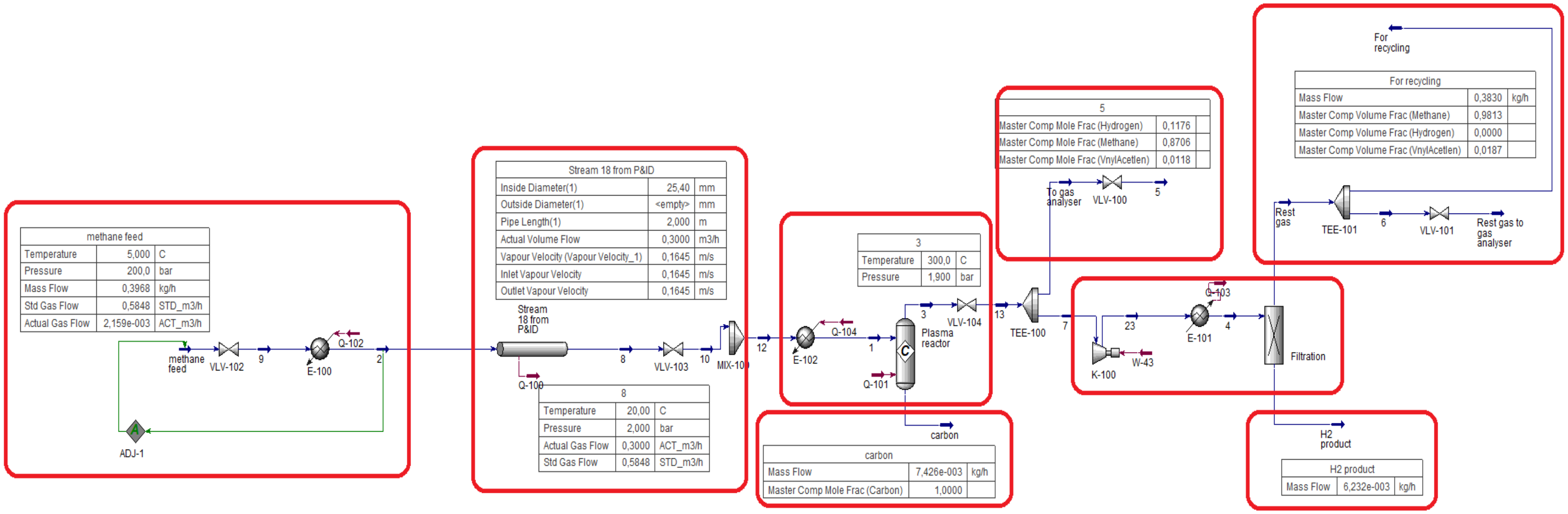
The start point is a simplified HYSYS model which has been created to design various laboratory set-up as required scales for various purposes. The model can be used to determine tubing size versus flow rate such as the maximum likely gas flow through each pipe section has been evaluated. Also, the efficiency of the plasma process has been taken into the model as manually entered parameters, these must be validated based on experimental results once available. Several byproducts were anticipated in this early-stage development of the model and incorporated into the model based on a python-script that used randomness in the recombination of the atoms from the plasma state.

The model has been tuned with the input parameters from the preliminary laboratory results and the design criteria for the upscaling to the set goals of the project. Several cases using the HYSYS model have been run, with different input flow, and with and without recycling of by-products.

The framework and the model will be continuously improved with the experimental data in the future project work.

2 DESCRIPTION OF THE HYSYS MODEL

A HYSYS model was built to simulate the conversion of methane into hydrogen, carbon and byproducts in the ColdSpark® plasma reactor. A screenshot of the PFD of the model is shown in Figure 1 below. Each of the red sections will be further outlined below.



methane feed		
Temperature	5,000	C
Pressure	200,0	bar
Mass Flow	0,3968	kg/h
Std Gas Flow	0,5848	STD_m3/h
Actual Gas Flow	2,159e-003	ACT_m3/h

Stream 18 from P&ID		
Inside Diameter(1)	25,40	mm
Outside Diameter(1)	<empty>	mm
Pipe Length(1)	2,000	m
Actual Volume Flow	0,3000	m3/h
Vapour Velocity (Vapour Velocity_1)	0,1645	m/s
Inlet Vapour Velocity	0,1645	m/s
Outlet Vapour Velocity	0,1645	m/s

8		
Temperature	20,00	C
Pressure	2,000	bar
Actual Gas Flow	0,3000	ACT_m3/h
Std Gas Flow	0,5848	STD_m3/h

3		
Temperature	300,0	C
Pressure	1,900	bar

carbon		
Mass Flow	7,426e-003	kg/h
Master Comp Mole Frac (Carbon)	1,0000	

5		
Master Comp Mole Frac (Hydrogen)	0,1176	
Master Comp Mole Frac (Methane)	0,8706	
Master Comp Mole Frac (VnylAcetlen)	0,0118	

For recycling		
Mass Flow	0,3830	kg/h
Master Comp Volume Frac (Methane)	0,9813	
Master Comp Volume Frac (Hydrogen)	0,0000	
Master Comp Volume Frac (VnylAcetlen)	0,0187	

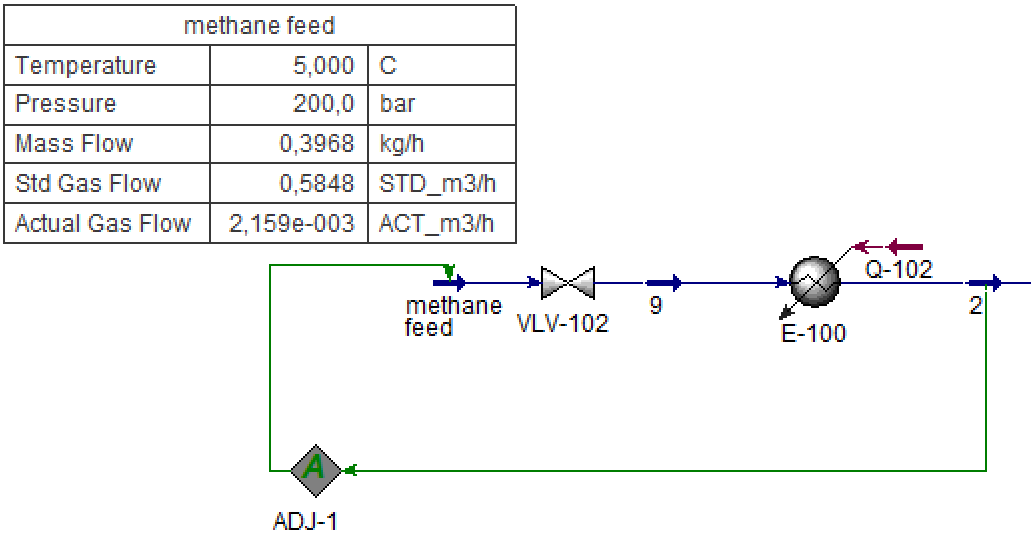
H2 product		
Mass Flow	6,232e-003	kg/h

Figure 1: Screenshot of the HYSYS PFD, and different sections to be outlined below.

Further information on how the plasma reactor is modelled in HYSYS is given in Appendix A – Behind the scenes of HYSYS simulations

2.1 SECTION 1 – GAS INLET

Figure 2 below shows the inlet facilities from methane gas storage tank. The gas is stored at 200 bar at anticipated ambient temperature of 5°C. The valve reduces the pressure to the desired line pressure of approximately 2 bara. The heater downstream of the valve increases the temperature to desired gas temperature of 20°C. The adjuster regulates the gas flow rate. In this case it is used a methane flow rate of 5 L/min at actual line conditions, which corresponds to 0.4 kg/h.

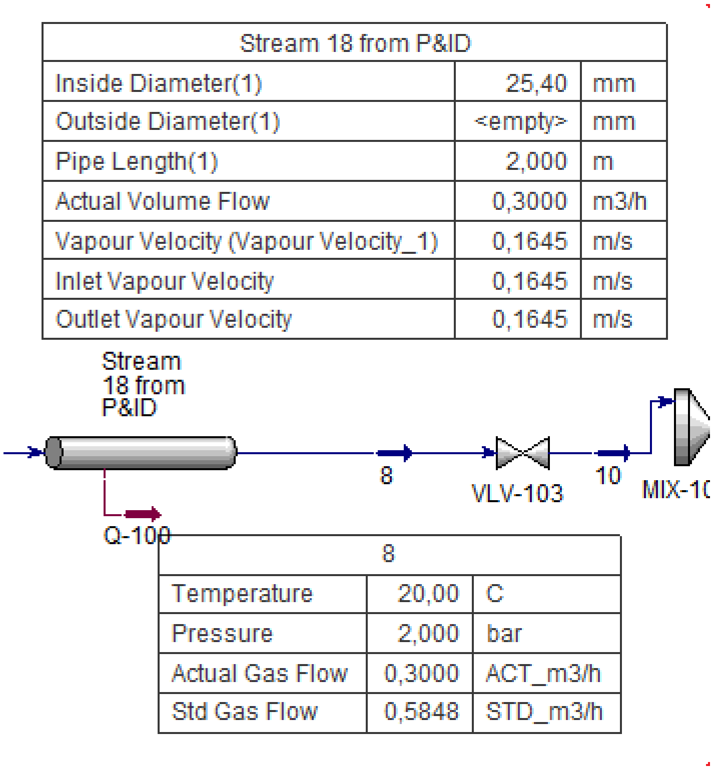


methane feed		
Temperature	5,000	C
Pressure	200,0	bar
Mass Flow	0,3968	kg/h
Std Gas Flow	0,5848	STD_m3/h
Actual Gas Flow	2,159e-003	ACT_m3/h

Figure 2: From gas storage tank into the system.

2.2 SECTION 2 – LINE CONDITIONS

The line conditions are shown in Figure 3 below. The flow rate, temperature and pressure are the same as in Figure 2. Two meters of piping has been used. In this 5 L/min case, a pipeline diameter of 1” is more than sufficient to handle the flow rate, as the gas velocity is well below the limitations given by NORSOK P-001 [6]. For cases with higher flow rates, it may be necessary to increase the pipeline diameter.



Stream 18 from P&ID		
Inside Diameter(1)	25,40	mm
Outside Diameter(1)	<empty>	mm
Pipe Length(1)	2,000	m
Actual Volume Flow	0,3000	m3/h
Vapour Velocity (Vapour Velocity_1)	0,1645	m/s
Inlet Vapour Velocity	0,1645	m/s
Outlet Vapour Velocity	0,1645	m/s

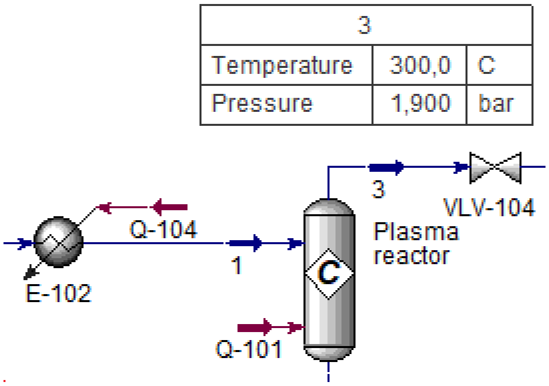
8		
Temperature	20,00	C
Pressure	2,000	bar
Actual Gas Flow	0,3000	ACT_m3/h
Std Gas Flow	0,5848	STD_m3/h

Figure 3: Line conditions.

The mixer on the right is there to enable the model to simulate a combination of gas from the methane storage and the gas mixture from the recycling line described in section 2.6.

2.3 SECTION 3 – THE PLASMA REACTOR

Figure 4 below shows the plasma rector part of the simulation. As HYSYS does not have a plasma module, a conversion reactor is used to model this instead. The upscaling is therefore also ONLY dependent on material balance and relationships observed in the laboratory. The possible differences in electrode arrangement leading to different contact volume of gas with the plasma field during the upscaling process is therefore not considered. Therefore, the results for the upscaled models are the first order estimates and must be fine-tuned in the future with experimental data. The temperature and pressure are set to give a prediction of the real outlet temperature. From the reactor, carbon is let out from the bottom, while gases leave from the top.



3		
Temperature	300,0	C
Pressure	1,900	bar

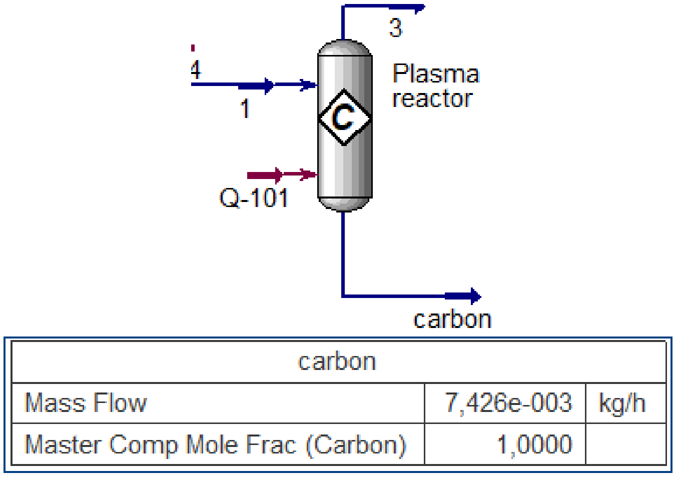
Figure 4: The plasma reactor.

Several reactions are possible to occur inside the reactor. In this preliminary model, only a few reactions are included, but the number can be increased when actual experimental data are available. Several byproducts are likely to occur during plasma cracking, such as ethane, propane, butanes, etc. These can be incorporated into the model once quantified from experiments.

2.4 SECTION 4 – PRODUCTS LEAVING THE PLASMA REACTOR

2.4.1 CARBON

From the bottom of the plasma reactor, the carbon product is collected, as shown in Figure 5. No gas is anticipated to follow this solid-state carbon.

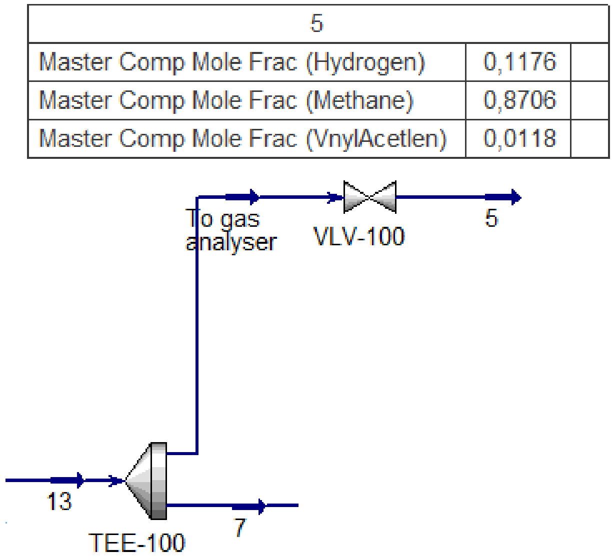


carbon		
Mass Flow	7,426e-003	kg/h
Master Comp Mole Frac (Carbon)	1,0000	

Figure 5: Solid carbon outtake from the plasma reactor.

2.4.2 GASES

The gases leaving the plasma reactor is sent to the separation and purification unit, where hydrogen is separated from the other products created in the reactor. One slip stream is sent to GC for analysis. This is illustrated on Figure 6.



5		
Master Comp Mole Frac (Hydrogen)	0,1176	
Master Comp Mole Frac (Methane)	0,8706	
Master Comp Mole Frac (VnylAcetlen)	0,0118	

Figure 6: Slip stream from plasma reactor for compositional analysis.

2.5 SECTION 5 – SEPARATION OF HYDROGEN FROM OTHER GASES

Figure 7 shows the preparation for separation of hydrogen from the other gases. The pressure and temperature are adjusted to suitable values for the desired separation process. From WP2 it has been suggested that 30% H_2 content should be achieved from the plasma reactor to get an efficient hydrogen separation process. This can be achieved by increasing the power supply for the plasma reactor, or by increasing the residense time in the reactor (rate and/or size of the reactor), according to preliminary data from WP1.

In this case the H_2 separation and purification unit is modelled as a component splitter. This can represent both a PSA separation or a filtration unit.

Hydrogen separated from other gases is sent for storage.

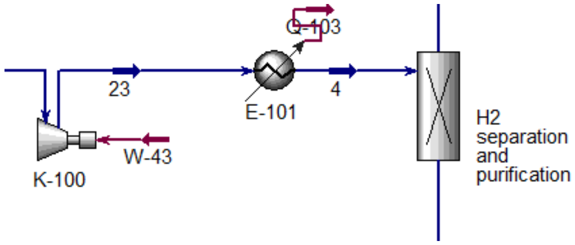
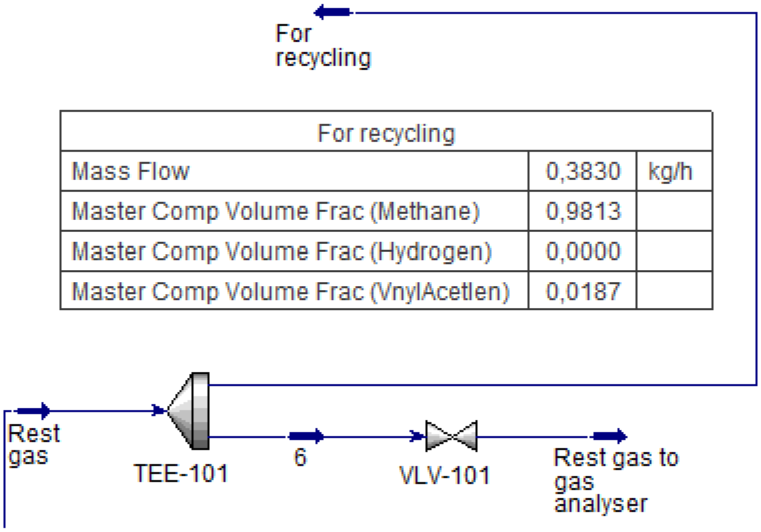


Figure 7: Separation of hydrogen from other gases.

2.6 SECTION 6 – TREATMENT OF BYPRODUCTS

When hydrogen is separated and sent for storage, there are still some remaining gases (unreacted methane and byproducts). A slip stream from this gas phase is sent for compositional analysis. The remaining gas can be stored, burnt, or emitted directly to air. This (potential) recycling line is shown in Figure 8. However, there is also an option to recycle these gases back into the loop by injecting them into the mixer upstream the plasma reactor (see Figure 3). In such case, pressure and temperature must be adjusted to meet the line conditions. Also, it is important to keep control over the total gas mass that enters the reactor in this case, and the model (and system) should include a flow controller to make sure that the correct amount of gas enters the reactor.



For recycling		
Mass Flow	0,3830	kg/h
Master Comp Volume Frac (Methane)	0,9813	
Master Comp Volume Frac (Hydrogen)	0,0000	
Master Comp Volume Frac (VnylAcetlen)	0,0187	

Figure 8: Handling of byproducts.

2.7 MODEL UPSCALING

The current parameters used in the HYSYS simulations are based on intuition and best guesses. Hence, the results that have been obtained are not based on an optimised model tuned by experimental data. Thus, the results shown in Table 1 below are only for illustrational purposes to indicate which parameters that can come out from a HYSYS simulation. The table covers lab scale experiments, but also covers upscaled experiments (up to 10,000 L/min of methane).

For all cases, pressure and temperature at reactor inlet is 2 bara and 20°C respectively. Power consumption for the plasma reactor has not been included for the cases. Also, for all cases, an increase in the power will result in an increase of reaction rates, as shown by preliminary experiments from WP1 (not public report).

The residence time and power consumption will not be directly modelled by HYSYS. However, actively use of the conversion parameters for each of the chemical reactions will simulate the effect of increasing power.

2.8 MODELLING RESULTS

Table 1 summarises the cases that have been modelled at various inlet flow rates. These cases with associated design parameters supply a first order estimates for the performance of the system and the effects of up-scaling. Note that these simulations anticipate up-scaling of the plasma reactor in line with the flow rate, which might not be the case.

Table 1: Summary of HYSYS results for lab scale (1-100) and upscaling (100-10 000 L/Min)

Flow rate (L/min)	0.5	1	5	10	50	100	1000	10000
Feed gas mass flow (kg/h)	0.04	0.08	0.40	0.79	3.97	7.94	79.35	793.53
Pipe diameter (mm)	25.40	25.40	25.40	25.40	25.40	25.40	25.40	76.20**
Gas velocity (m/s)	0.02	0.03	0.16	0.33	1.64	3.29	32.89 *	36.55 *
Output H_2 (mol-%)	11.63	11.63	11.63	11.63	11.63	11.63	11.63	11.63
Output CH_4 (mol-%)	86.05	86.05	86.05	86.05	86.05	86.05	86.05	86.05
Output C_2H_2 (mol-%)	2.33	2.33	2.33	2.33	2.33	2.33	2.33	2.33
Output H_2 (kg/h)	0.0006	0.0012	0.0062	0.0125	0.0623	0.1246	1.25	12.46
H_2 conversion efficiency (%)***	6.28	6.28	6.28	6.28	6.28	6.28	6.28	6.28
Unreacted CH_4 (kg/h)	0.037	0.073	0.37	0.73	3.67	7.34	73.39	730.74
Output C_2H_2 (kg/h)	0.0016	0.0032	0.016	0.032	0.161	0.322	3.22	32.05
Carbon (mg/min)	12.38	24.75	123.77	247.54	1237.7	2475.4	24753	32054

*Consider increasing piping diameter

** 3-inch piping used

*** Mass H_2 formed / (Mass methane x (4/16))

The main purpose of model upscaling is to estimate the dimension of the upstream and downstream process parameters to the large-scale lab testing with the planned H_2 capacity of 1 kg H_2/hour scale. and to use the experimental results to further upscale x10 to reach relevant industrial scale which form realistic basis for economic estimates and LCA/LCC analysis (WP6 - Sustainability and techno-economic assessment of plasma methane cracking process).

3 SUMMARY AND RECOMMENDATIONS FOR THE FUTURE WORK

3.1 SUMMARY

The modelling framework has been established and can be used to support the project. Although the calculations from HYSYS must be further tuned with experimental data, the estimates of H_2 and carbon production, the necessary hardware and pressure / temperature management. etc. can be used as indicators for the planning work.

3.2 RECOMMENDATIONS

The established framework must be further fine-tuned with experimental data in the future. The experimental data will also give guidance for the model upscaling process, for example reactor dimensioning, plasma electrodes arrangement, total energy input, flow rate versus residence time of CH_4 in the plasma field (from other modelling tools). These mechanisms and relationships can be described with the input parameters in the current framework to determine the H_2 and carbon output.

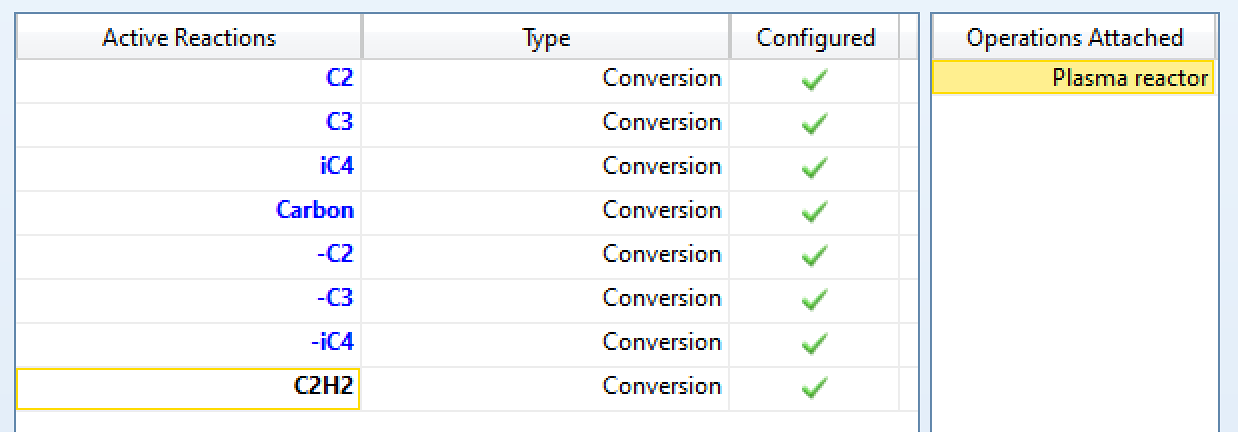
The downstream H_2 separation and purification process will need own process simulation, which may include Aspen Adsorption modelling (a separate module in Aspen/HYSYS. ref 1) for the VPSA performance and the required P/T to reach the design goal before recirculation of the uncracked CH_4 back to reactor. This is planned in WP2 and the outcome will be integrated with the WP5 models in the future.

4 REFERENCES

1. Aspen/HYSYS model version / user Guide
2. D1.4. Evaluation of the impact of impurities on the plasma cracking. University of Liverpool (internal report)
3. D2.1 Report on optimal design and process parameters for hydrogen separation and reactor gas recycling unit. EU ColdSpark® WP2 project report. 2023-11-30. University of Stavanger (sensitive report. for internal use)
4. D6.1 Methodology framework and baseline definition for life cycle studies (sensitive report. for internal use)
5. ColdSpark® work meeting documents for design and upscaling of SEID's ColdSpark® reactors (for internal use only).
6. NORSOK P-001 – Process design

APPENDIX A – BEHIND THE SCENES OF HYSYS SIMULATIONS

To enable the conversion reactor to act as the plasma reactor in the HYSYS model all the possible reactions taking place inside the reactor must be added. Some potential reactions are added in the menu shown in Figure 9 below, but the list can be extended based on experimental results.



Active Reactions	Type	Configured
C2	Conversion	✓
C3	Conversion	✓
iC4	Conversion	✓
Carbon	Conversion	✓
-C2	Conversion	✓
-C3	Conversion	✓
-iC4	Conversion	✓
C2H2	Conversion	✓

Operations Attached
Plasma reactor

Figure 9: Menu for reactions in the conversion reactor in HYSYS.

Each of the reactions are named after the product they produce. C2, C3, iC4 and carbon are thus reactions that produce ethane, propane, iso-butane and carbon respectively. The reactions denoted with a minus sign in front (-C2), are in the case where the byproducts are recycled back into the reactor, and they may be cracked and re-react to form methane.

The following reactions are defined in this model (but the list can be increased by demand):

C2: $2CH_4 \rightarrow C_2H_6 + H_2$

C3: $3CH_4 \rightarrow C_3H_8 + 2H_2$

iC4: $4CH_4 \rightarrow C_4H_{10} + 3H_2$

Carbon: $CH_4 \rightarrow C + 2H_2$

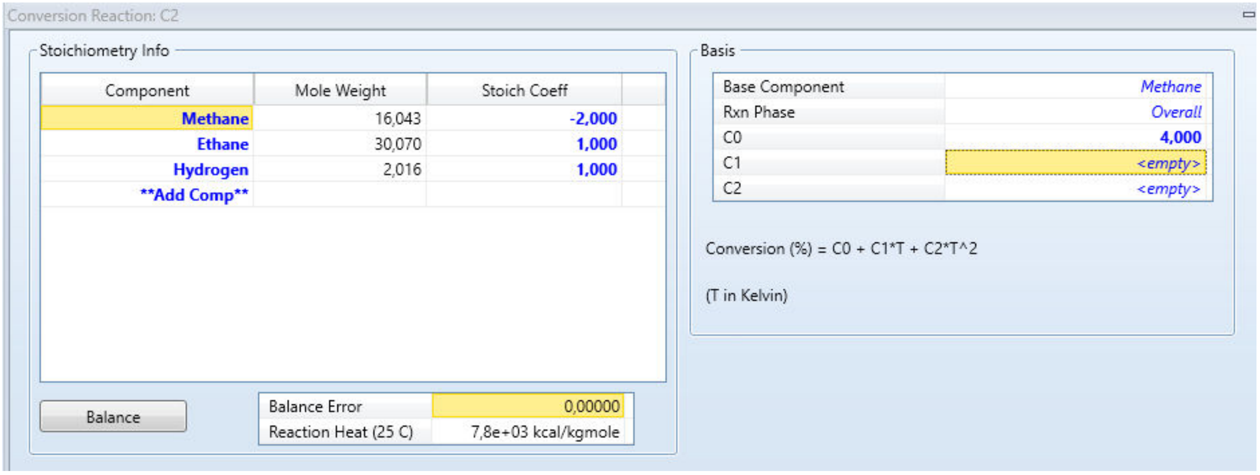
-C2: $C_2H_6 + H_2 \rightarrow 2CH_4$

-C3: $\mathrm{C_3H_8 + 2H_2 \rightarrow 3CH_4}$

-iC4: $C_4H_{10} + 3H_2 \rightarrow 4CH_4$

C2H2: $2CH_4 \rightarrow C_2H_2 + 3H_2$

For each of the reactions above, stoichiometric coefficients must be inserted. These coefficients are the same as in the equations above. In the case for C2 (ethane), shown in Figure 10 below, -2 for methane means that two molecules of methane are cracked to form one molecule of ethane and one molecule of hydrogen. The balance error of 0,000 means that the reaction is correctly balanced.



Component	Mole Weight	Stoich Coeff
Methane	16,043	**-2,000**
Ethane	30,070	**1,000**
Hydrogen	2,016	**1,000**
****Add Comp****		

Base Component	*Methane*
Rxn Phase	*Overall*
C0	**4,000**
C1	*<empty>*
C2	*<empty>*

Conversion (%) = C0 + C1*T + C2*T^2

(T in Kelvin)

Balance Error	0,00000
Reaction Heat (25 C)	7,8e+03 kcal/kgmole

Figure 10: Reaction coefficients for reactions defined.

To determine how much ethane is actually formed in the reactor, the conversion parameters must be set. This parameter will be manually adjusted based on experimental results.