Integrated design for demonstration of efficient liquefaction of hydrogen (IDEALHY)

Fuel Cells and Hydrogen Joint Undertaking (FCH JU)

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Publishable summary

In the now concluded process selection phase (Work Package 1, WP1) of IDEALHY the most promising hydrogen liquefaction concept was selected for further development in the succeeding Work Package 2 (WP2). The liquefier concept selected was described by a set of process characteristics with respect to hydrogen pressure level and process pathway, refrigeration process types and refrigerants for pre-cooling and cryogenic cooling, and expansion devices for final expansion and liquefaction.

This report summarises activities related to process modelling and simulation work for the most promising IDEALHY liquefaction concept. A particular emphasis in this report is on assumptions made for the central process units and sub-system of the liquefaction process.

In order to maximise energy efficiency a mixed-refrigerant cycle was decided upon for pre-cooling of hydrogen from feed temperature to the range 100–140 K. The current pre-cooling level is 132 K. For the cryogenic Brayton cycle, selection of refrigerant was not concluded in WP1 but has later been defined as a mixture between helium and neon, carefully optimised with respect to optimal compander and compressor train design and minimum cooling temperature.

From parallel activities in development of key process components and sub-systems, such as compressors, expanders, heat exchangers and catalysis, some of the recent findings have affected the original process design and required these to be modified. One example is the initial assumption that the liquefaction process would be once-through; this has had to be modified, conceding a certain rate of hydrogen flash gas during final expansion. The generation of hydrogen flash gas can be related to new, higher estimates for the minimum temperature level generated in the helium/neon Brayton cycle, and this has resulted in an additional reheat and recompression loop for the hydrogen gas. Another process modification made in WP2 is the introduction of a chiller system for additional cooling of high-pressure helium/neon to 279 K. The introduction of the chillers was identified as a means of reducing the compressor suction volume and the overall power consumption.

With the current process assumptions, based on ongoing work on the various components' development in WP2, the net power consumption is estimated to be 12.8 MW for 50 t/d liquefaction rate. This corresponds to a specific liquefaction power of 6.15 kWh/kg, which is slightly above the stated IDEALHY objective of 6 kWh/kg.

Further work in WP2 includes development and optimisation of process components for application in the IDEALHY liquefaction process and will be complemented by reports on efficiency and cost as well as an engineering audit. Advances in components' development beyond the current state may enable further improvements in specific liquefaction power and results closer to those defined by the IDEALHY objectives.

Key words

Hydrogen Liquefaction, Brayton Cycle, Mixed Refrigerant Cycle, Compressors, Heat Exchangers, Ortho-Para Conversion, Process Modelling and Simulation

Table of Contents

1 Introduction

As one of the main activities in WP1 of the IDEALHY project, a broad range of hydrogen liquefaction processes, existing and conceptual, were benchmarked subject to a defined set of rather realistic large-scale boundary conditions and process unit assumptions.

The main types of process configurations benchmarked were Claude cycles with hydrogen as refrigerant, Brayton cycles with mainly helium but also neon as refrigerant, and mixed-refrigerant pre-cooling combined with Brayton cryogenic cooling.

In addition to the liquefier benchmarking, the design, operating parameters and not least the interdependencies for the main sub-processes of the overall processes were considered in greater detail. The sub-processes and parameters considered were primarily:

- Hydrogen pre-compression process: pressure level; impact on other sub-processes, particularly cryogenic cooling cycle and final expansion and liquefaction stage;
- Hydrogen pre-cooling cycle: temperature level; process type; refrigerant; impact on overall power consumption; machinery;
- Hydrogen cryogenic cooling cycle: temperature level; process type; refrigerant; impact on overall power consumption; feasibility of machinery; impact on final expansion and liquefaction stage;
- Final expansion and liquefaction stage: expander type; impact on outlet state of hydrogen

With a strong emphasis on minimisation of power consumption as well as feasibility of required process components such as machinery and heat exchangers, the most promising concept was chosen, based on the process analyses performed in WP1. The benchmarking gave clear answers to what configurations and sub-process combination should be selected. The following conclusions for the most promising concept were made:

- 60–80 bar pre-compressor discharge pressure to reduce heat capacity peak around the critical point of hydrogen;
- Mixed-refrigerant pre-cooling cycle to enable gliding temperature profiles, optimised refrigerant composition and high energy efficiency. Hydrogen outlet temperature somewhere in the range of 100–140 K;
- Once-through process (no recycling of hydrogen gas) if practically feasible;
- Partially temperature-overlapping Brayton cycles for cryogenic cooling of hydrogen to a temperature between 24 and 30 K;
- Liquid expanders for final expansion and liquefaction stage.

The selection of refrigerant for the Brayton cycle was not made in WP1, but was defined later as a mixture of helium and neon, carefully optimised with respect to optimal compander and compressor train design and minimum cooling temperature.

This report documents the work on process design and simulations of the selected conceptual high-efficiency hydrogen liquefaction process. As will be observed, some of the conclusions made in WP1 have been modified during WP2, because further process design and optimisation is governed by the on-going and parallel work on component assessment in Task 2.2.

2 Process modelling, optimization and feasibility evaluation

Modelling and simulations of the complete liquefaction cycle were performed on an overall process level. Process components such as compressors, expanders and heat exchangers were modelled as general units, with input parameters such as efficiency figures derived from the detailed process unit studies in Task 2.2.

Two simulation environments were utilised in the process modelling, Aspen HYSYS and Microsoft Excel. A comparison of these tools was performed at an earlier stage in WP1 and results are described in D1.3.

In the following more detailed information is given on the process modelling and optimisation of each component, sub-process as overall system.

2.1 Compressor trains

As compressor trains for pre- and cryogenic cooling cycles make up the major portion of power consumption and CAPEX for high-efficiency liquefiers, these are also the subsystem for which relative improvements will yield the greatest overall improvement in absolute terms. Moreover, as the optimal design of a compressor train is highly sensitive to the selected refrigerant as well as suction and discharge pressure levels, compressors is a prioritised area of improvement in IDEALHY and the work conducted in Task 2.2.

2.1.1 Helium/neon compressors

In order to achieve the IDEALHY target for power consumption, an overall isothermal efficiency of approximately 70% is required. For conventional oil-lubricated screw compressors the best figure is about 54% (Ganni et al., 2008) and far from sufficient from an efficiency point of view. Hence, the only realistic alternative for the IDEALHY application is to use turbo compressors for light-gas compression in the cryogenic refrigeration cycle.

As turbo compressors are feasible for gases with molecular weight above approximately 6 kg/kmol, the refrigerant selected has to be a mixture of helium and neon. Based on the latest interaction with vendors a helium/neon mixture made up of 75% helium and 25% neon is currently regarded as a near-optimal trade-off for the large-scale liquefier concept under consideration. The search for an optimal helium/neon mixture has to take mainly two aspects into account: on the one hand maximising the average molecular weight and thus the achievable pressure ratio per turbo compressor stage, and on the other hand minimising/avoiding the amount of condensed neon (definitely avoiding freeze-out) in the very cold end at the outlet of the coldest expander in the cryogenic Brayton cooling cycle.

The resulting average molar weight of the helium/neon mixture is 8 kg/kmol and the maximum pressure ratio per impeller is therefore still very low for turbo compressors; 1.3 for the 75%/25% mixture compared to about 1.15 for pure helium. Due to pressure drop and other losses it is not efficient to intercool the refrigerant between each impeller. Based on information from compressor vendors, the optimal solution seems to be six intercooled compressor stages, each with 3-5 impellers. The helium/neon compressor train consumes about 75 % of the total power input and the cryogenic cooling cycle compressor train is hence by far the most power intensive process in the liquefaction cycle.

In the overall process simulation the compressor train is simulated as six compressors stages with isentropic efficiencies and pressure drop in intercoolers based on vendor data.

In the initial design from WP1, the helium/neon refrigerant was designed to be compressed from 1.85 bar to 23.6 bar with the compressor train, and further to 35 bar with the expander brake compressors. The large volumetric flow rates and low suction pressures were found challenging for the compressor design, and several measures were taken to reduce these problems. Firstly the suction pressure was increased, secondly the mass flow rate was reduced by increasing the overall pressure ratio, and finally only part of the refrigerant was expanded down to the lowest pressure at 2.4 bar. The current layout is shown in Figure 1, where a second partial low-pressure stream (LP II) enters the compressor train at the second stage inlet at 3.8 bar pressure.

As another means of further reducing the compressor suction volume and power consumption, additional aftercooling by an external chiller is proposed in the process design. The currently proposed location of these chillers is downstream of the high-pressure stages of the compander part of the helium/neon compressor train, as indicated in the overall process flow diagram shown in Figure 8. The chiller cools the refrigerant to 279 K after each of the two parallel high-pressure stages after regular intercooling, thus reducing the temperature of the low-pressure streams at the outlet of heat exchangers HX 104 and HX 105.

Another effect of increasing both low-pressure and high-pressure levels is reduction of intercooler losses. Intercooler pressure drop is more an absolute figure [bar] than relative [%] and the impact of pressure losses is hence less severe at elevated pressure levels. Furthermore, the introduction of chillers also contributes to reducing pressure losses on the low-pressure side of the helium/neon refrigeration cycle by reducing the duty of the above-mentioned heat exchangers. This effect has an additional contribution to reducing overall power consumption.

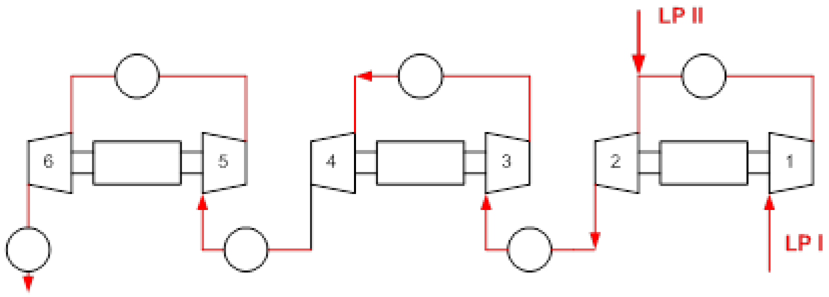

Figure 1: Current He/Ne compressor train layout

2.1.2 Mixed-refrigerant compressor

The mixed-refrigerant (MR) compressor has so far been modelled as a single-stage compressor with 85 % isentropic efficiency. A two-stage compression would increase the efficiency, but this evaluation will be a part of the techno-economic optimisation later in the project. In addition, depending on the refrigerant composition, one could risk partial condensation of the refrigerant in the intercooler, which would require a scrubber and a pump in parallel to the second stage compressor.

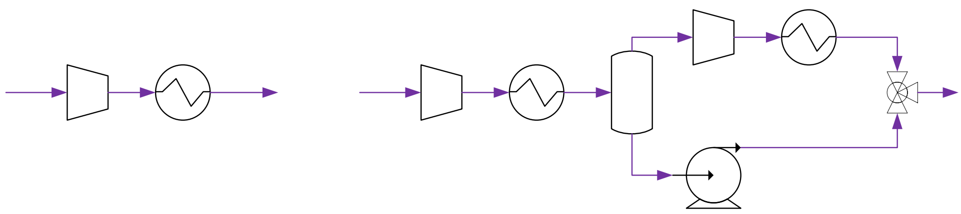

Figure 2: MR compression train layout

2.1.3 Hydrogen feed and recycle compressors

The hydrogen feed compression from 20 to 80 bar is assumed to be a two stage piston compressor. For process simulation purposes this is modelled with two intercooled stages with isentropic efficiencies and heat losses from vendors.

Due to the increased low pressure in the refrigeration cycle and the risk of neon freeze-out, it is not feasible to avoid hydrogen flash gas completely. Hence, the assumption of a once-through liquefaction process has been modified since WP1 was concluded. With the high hydrogen feed pressure it is found that the best solution is not to compress the almost pure para-H_2 flash gas back to the hydrogen feed at 80 bar, but to recompress the flash gas in a separate loop operating at considerably lower pressure. In the process simulations it is assumed that the retention time of hydrogen flash gas in the recycle circuit will be short enough to avoid partial reconversion back to ortho-hydrogen.

2.2 Expanders and J–T valves

Another obvious finding from the process benchmarking in WP1 is the significance of efficient expanders. Whereas J–T expansions should be avoided due to high entropy generation and associated irreversibilities, power-generating expanders with high efficiency are preferable for high-efficiency concepts. In addition to generating cooling the shaft power generated by cryo expanders should be recovered, in IDEALHY done through using brake compressor stages powered by expanders in the helium/neon compressor train.

2.2.1 Helium/neon expanders and brake compressors

In the IDEALHY liquefier there are a total of 5 turbo expanders in the helium/neon Brayton cycle, each powering a brake compressor stage in the compressor train. Three expanders are employed in series in the high-temperature Brayton cycle, and two are in series in the low-temperature Brayton cycle. The efficiency figures used for cryo expanders in the simulations are based on those stated by the vendors involved.

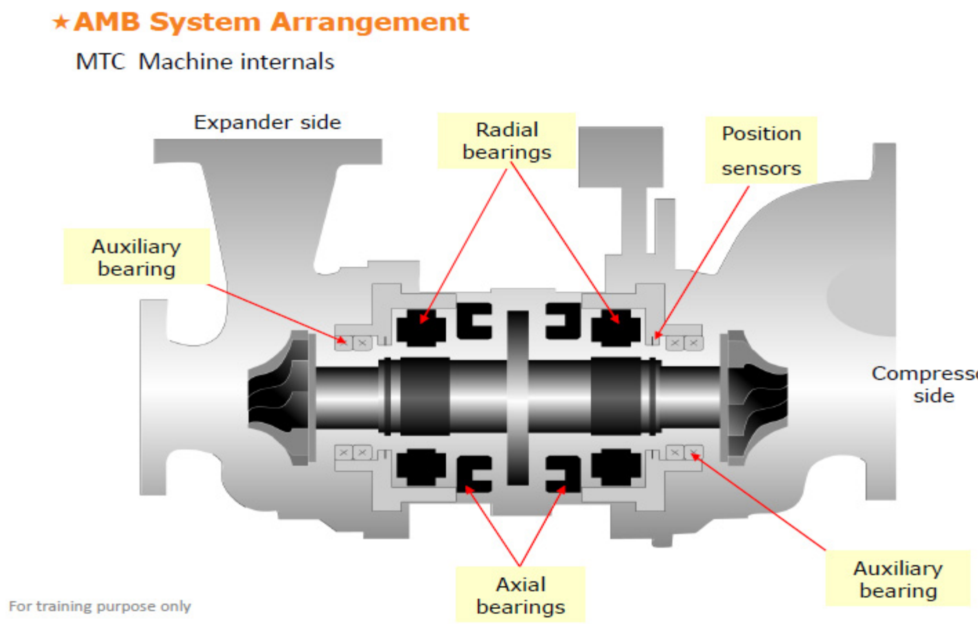

Figure 3: Expander/Compressor layout. By courtesy of SKF-S2M Magnetic Bearings

2.2.2 Wet hydrogen expanders

As described in D1.3, a wet expander will be used to expand and liquefy the cold hydrogen from 80 bar to the final storage pressure (2 bar). A two-stage expander with about 80 % isentropic efficiency per stage is available technology and will be used in the model.

2.2.3 J–T valves

For the hydrogen flash gas, a J–T valve is used for the expansion as the flow rate is too small for employing wet expanders as for the main hydrogen stream. This is an isenthalpic valve, which should be standard equipment as the flow rate is relatively low.

2.3 Heat exchangers

All the cryogenic heat exchangers are designed as plate-fin heat exchangers. Special design tools are needed to dimension these heat exchangers. However, for the purpose of process simulations, composite curve models with a defined pressure drop are used. Composite curve models combine the heat duty of all the hot streams into a hot composite cooling curve, and all the cold streams into a cold composite heating curve. The outlet temperature of either the hot or cold composite curve is defined, and the other is calculated based on heat balance. The outputs from the model are a mean temperature difference, heat transfer duty and a total UA value. UA is the overall heat transfer coefficient multiplied with the heat transfer area. This simplification assumes that it is possible to arrange the heat exchanger in a way that each hot stream gets the necessary cooling from the cold streams. For complex heat exchangers with many hot and cold streams, this is not always the case. The modelled heat exchanger is therefore calculated separately in specialised programs to check its feasibility.

2.3.1 Mixed-refrigerant heat exchanger

To evaluate the feasibility of the mixed refrigerant cycle, an initial design for the precooling heat exchanger (HX101) was sent for validation at Linde Engineering. The main challenge with mixed refrigerants in plate-fin heat exchangers is mis-distribution. The MR flow has to be divided on many layers on channels in each layer, giving ample risk of mis-distribution. Suitable header and distributor configurations are therefore vital for the heat exchanger design. However, these designs are based on experience and are proprietary information from vendors.

With the complexities involved in distribution of multiphase flows in plate-fin heat exchangers, it is proposed that the entire cooling should be done in one heat exchanger. Given the limited size and the risk of mis-distribution there is a limit to how thermodynamically efficient the heat exchanger can be designed. Initially, a limit of 40 NTU was set as a design criterion for the MR heat exchanger, and it is still thought that this reasonable. However, to evaluate the effect of this constraint, a case study was performed (see Figure 4).

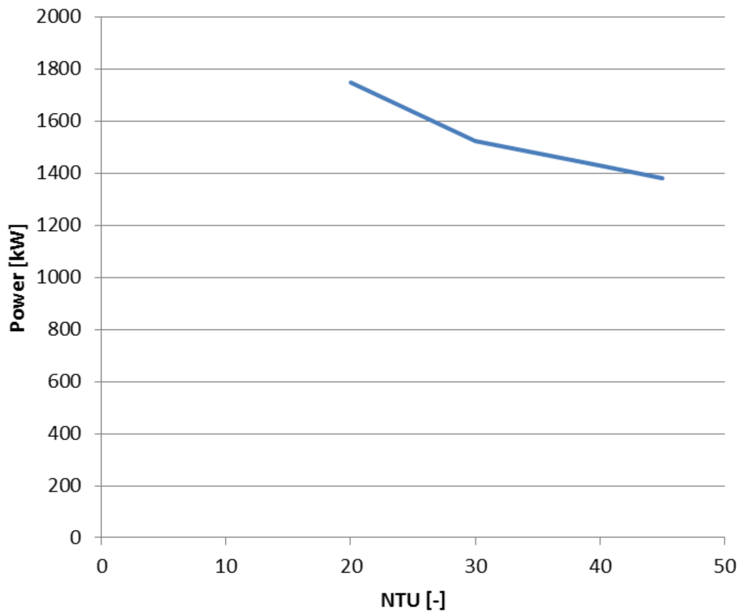

Figure 4: Estimates for power consumption vs. heat exchanger NTU for MR precooling to 132 K

To evaluate to what temperature it is efficient to use the MR cycle for precooling, it is necessary to evaluate the power consumption as a function of cooling temperature, with feasible heat exchanger configurations. As it is not realistic to do a detailed design for all these cases, the study is performed by using parameters from a base case heat exchanger design that is known to be feasible as boundaries for the cooling temperature evaluation study.

2.3.2 Hydrogen and helium/neon heat exchangers

Heat exchangers cooling the hydrogen from its feed to final temperature are assumed to be of plate-fin type. In cryogenic plants in general either plate-fin or coil-wound heat exchangers are used. Due to their higher efficiency and compact design (300–1,400 m^2/m^3 compared to 20–300 m^2/m^3 for coil-wound), plate-fin heat exchangers are the preferred type in the IDEALHY high-efficiency concept. The preferred hydrogen pressure level (80 bar) is below the upper bound for aluminium brazed plate-fin heat exchangers (100–115 bar) (ALPEMA, 2000) but limits the options for fin types. The fin types possible are further limited by fact that the hydrogen heat exchangers downstream of the purification unit must be filled with catalyst, typically in grains of a specific average diameter, These heat exchangers therefore need to have a wider pitch. Only a very restricted number of fin types can satisfy both conditions. From a thermodynamic point of view, these are not the most efficient fins. The size of the catalyst-filled heat exchangers is also driven by the required amount of catalyst. A thorough investigation of the effective reaction constant of the catalyst can help to minimise the final dimensions of the plate-fin heat exchanger blocks.

2.3.3 Intercoolers and chillers

The intercoolers are water-cooled and cool the compressed fluid to 298.15 K. To reduce the inlet temperature of the first He/Ne compressor stage, the high pressure flow from the brake compressors is cooled down to 279 K with a chiller operating with a conventional refrigerant, e.g. ammonia, propane or R134a.

The chiller will be a relatively standard refrigeration plant with evaporation temperature at about 274 K. The calculation of the chiller performance is not directly integrated to the main process calculation tool, but is calculated separately. Based on the actual chiller coefficient of performance (COP) for the chiller, the power consumption can be calculated. With an available cooling water temperature the COP would be expected to be in the range of 5–6.

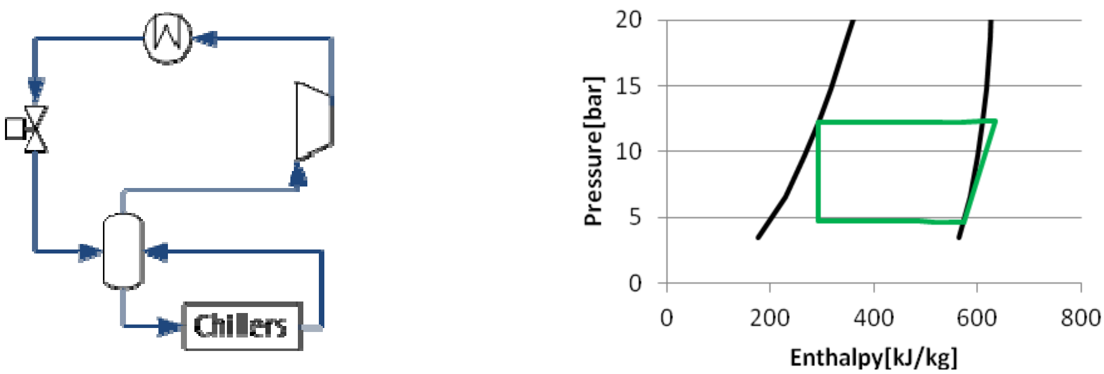

Figure 5: Possible chiller layout and ph-diagram for propane

2.4 Adsorption/final purification and ortho-para conversion

To reach the necessary purity level of the hydrogen product, a low-temperature purification stage is needed. In existing plants, this purification is located at the 80 K level, in combination with the first ortho-para conversion bed.

Initially it was planned to fill the heat exchangers with catalyst all the way from ambient temperature. However, since the catalyst will also be absorbing impurities, and the catalyst inside the heat exchangers is difficult to regenerate, this is not feasible. The temperature level where one can utilise catalyst-filled heat exchangers is therefore governed by the temperature of the final purification unit. This is still thought to be around 80 K.

Based on this, the configuration will be similar to the standard Linde Kryotechnik solution, with a combined purification and adiabatic conversion unit at about 80 K. Due to the temperature rise in the hydrogen during conversion, the feed must be re-fed to the previous heat exchanger before it is further cooled (see Figure 6). To further reduce the losses of the 80 K adiabatic conversion, an additional adiabatic converter is included at about 105 K. The catalyst will be placed outside the heat exchanger to allow for regeneration.

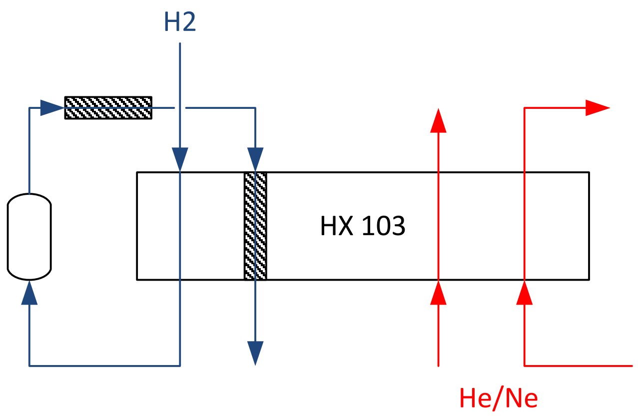

Figure 6: 80 K purification and conversion layout

2.5 Refrigerants and thermodynamic modelling

For the process simulations, the heat capacity and adiabatic expansion properties (heat capacity ratio) are the most important properties. For the ongoing and more detailed component calculations, other properties such as density, thermal conductivity and viscosity are also important.

The simulation and thermodynamic properties of hydrogen was discussed in D1.3. For the initial evaluation, the helium/neon refrigeration cycle was modelled as pure helium with thermodynamic data from NIST (Lemmon et al., 2010). For the high temperature part of the refrigeration, this is a valid assumption for the process calculations. Since the heat capacity ratios of the pure helium and the helium/neon mixture are equal, the mass flow has to be increased with the same ratio as the molecular weight.

However, as the temperature decreases, neon no longer behaves as an ideal gas, and the mixture does not have constant heat capacity ratio, the effect of which will be the subject of further investigation.

Table 1: Molecular weight, specific heat ratio and required mass flow rate for pure helium, 80%/20% and 75%/25% helium/neon mixtures

	Pure helium	80 % He 20 % Ne	75 % He 25 % Ne
Molecular weight (kg/kmol)	4.00	7.24	8.05
Specific heat ratio	1.67	1.67	1.67
Mass flow rate (kg/s)	3.97	7.19	7.99

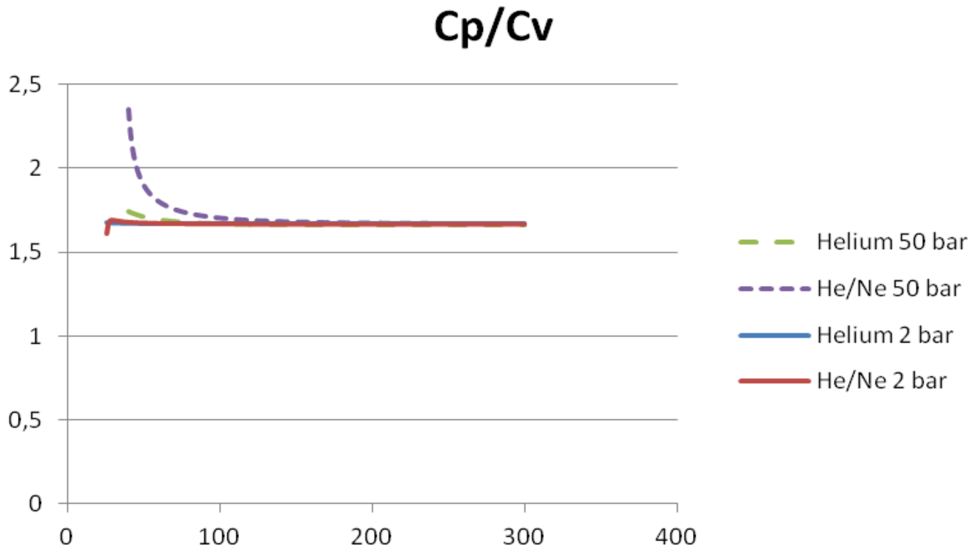

Figure 7: Heat capacity ratio for helium and He/Ne mixture at 50 and 2 bar, calculated as an ideal mixture with data from NIST (Lemmon et al., 2010)

3 Current configuration and key figures

For the current process configuration shown in Figure 8, key energy estimates are given in Table 2. From the original IDEALHY project plan, the following objective is stated for a 50 t/d liquefier: "The objective of the technical part of the project is to reduce this [24 MW] to less than 12 MW (less than 6 kWh/kg)." As can be observed the resulting specific power consumption for liquefaction is currently estimated to 6.15 kWh/kg, which is very close to the IDEALHY objective.

As work on key process components and interaction with vendors are still ongoing activities, the results in Table 2 may still be improved if better technical solutions, for the compressors, expanders and heat exchangers in particular, are identified. Moreover, the temperature level between precooling and cryogenic cooling, currently 132 K, will also be subject to further energy optimisation, which in turn may yield improved power results.

Table 2: Current power figures from simulations of the IDEALHY hydrogen liquefier

	Shaft power	Motor losses	Total	
Feed compression	1 450	70	1 520	kW
Flash gas	95	5	100	kW
Mixed refrigerant	1 421	70	1 491	kW
Brayton cycles incl. chiller	9 700	included	9 700	kW
		Sum	12 811	kW
		LH_2 flow rate	0.579	kg/s
		Specific power	6.15	kWh/kg

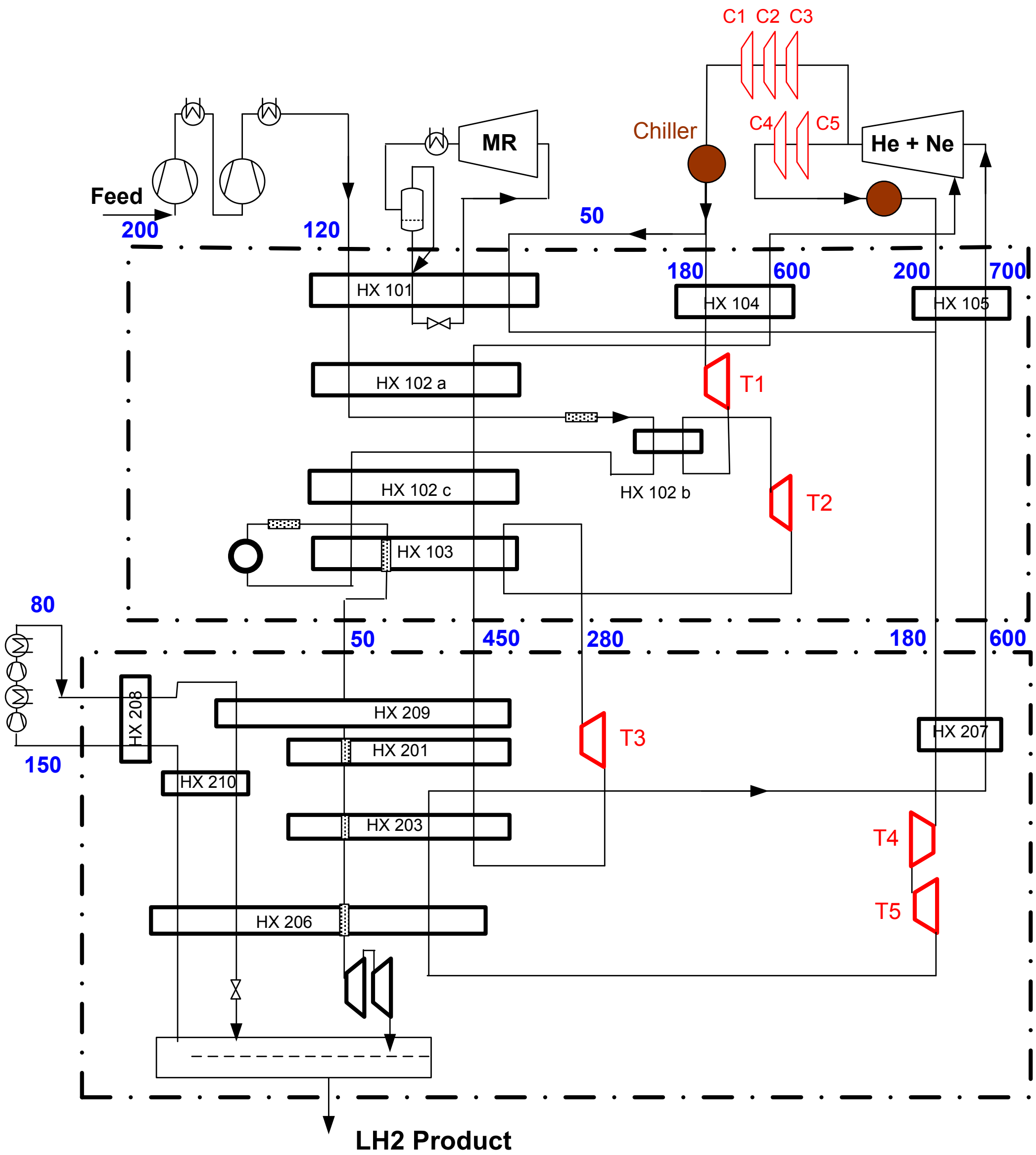

Figure 8: Process flow diagram for the IDEALHY high-efficiency hydrogen liquefier.

4 Integration with liquefied natural gas (LNG)

Steam reforming of natural gas is the most common method for hydrogen production. At LNG import terminals the LNG is re-gasified and at least partly warmed up for high-pressure pipeline distribution. The gas is compressed and fed into a pipeline or otherwise distributed. The energy for evaporation and warm-up comes either from sea water or by combustion of some natural gas. The cold released during evaporation and warming of the LNG to ambient temperature can be efficiently used to reduce both the energy requirements as well as the capital costs of hydrogen liquefaction.

The use of the cold from the LNG, its potential for reducing liquefier energy consumption and the process design for including LNG as a pre-cooling refrigerant, will be highly dependent on the amount of LNG available relative to the hydrogen liquefaction rate. The simplest and obvious solution would be to replace the MR precooling with LNG regasification directly, as there is a temperature match with the current liquefier configuration (precooling to 132 K). According to the simulation model, an LNG re-gasification rate of about 1.8 kg/s (57,000 tonnes/year) is required to supply the necessary pre-cooling for the current 50 t/d hydrogen liquefier. Power calculations from the simulation model show a reduction in power consumption of about 1,500 kW caused by this integration, resulting in a specific power consumption reduction from 6.1 kWh/kg to approximately 5.4 kWh/kg.

The second LNG integration option would be to use the LNG to lower the inlet temperature and thus the suction volumes for the helium/neon compressors, by replacing the chillers and high temperature recuperators (HX 104 and partly HX 105) – this could also be envisaged for the hydrogen feed compressors. Replacing chillers and high-temperature recuperators for the helium/neon would require an LNG flow rate of about 4.45 kg/s (140,000 tonnes/year). This would reduce the helium/neon compressor power by about 2.6 MW, and correspondingly reduce the specific power consumption from 6.1 kWh/kg to about 4.9 kWh/kg. However, certain other losses such as heat losses may have to be taken into account. Since the suction volume of the first helium/neon compressor stages are drastically reduced, this could be an advantage for the design of the compressor train. If even higher flow rates of LNG were available, intercooling in the helium/neon compression train would reduce the power consumption further.

If LNG is to be integrated with the IDEALHY liquefier there are several aspects to consider before the optimal solution can be determined. A fundamental question is whether the liquefier should be designed for two points of operation: with and without LNG, in case this flexibility is required.

From the two LNG integration alternatives described above, the first(replacing MR pre-cooling with direct heat exchange against pressurised LNG) represents the less complicated solution from the viewpoint of process flexibility. By replacing MR pre-cooling only, the operation of the helium/neon cycle and compressor train are affected to a much lesser degree. Since the compressors can be run at design point in both cases, this should enable high efficiency in both modes and thus be a more robust solution.

5 Conclusions and further work

This report documents the steady-state process modelling and simulation work for the liquefaction process selected as the most promising concept in IDEALHY. A particular emphasis in this report is on assumptions made for the central process units and sub-system of the liquefaction process: helium/neon compressors, mixed-refrigerant pre-cooling cycle, hydrogen compressors, helium/neon cryo expanders and brake compressors, heat exchangers, intercoolers and chillers, the adsorber and the ortho-para conversion catalyst.

With the current process assumptions, based on ongoing work on the various components' development in WP2, the net power consumption is estimated to be 12.8 MW for 50 t/d liquefaction rate. This corresponds to a specific liquefaction power of 6.15 kWh/kg, which is slightly above the stated IDEALHY objective of 6 kWh/kg.

Further work in WP2 includes development and optimisation of process components for application in the IDEALHY liquefaction process and will be complemented by reports on efficiency and cost as well as an engineering audit. Advances in component development beyond the current state may enable further improvements in specific liquefaction power and results closer to those defined by the IDEALHY objectives.

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7 Acknowledgements

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