Efficient Liquefaction of Hydrogen: Results of the IDEALHY Project

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Zusammenfassung: Die Verflüssigung von Wasserstoff gilt heute als energie- und kostenintensiv. Im Projekt IDEALHY wurde ein effizienter Prozess für zukünftige Großanlagen entwickelt. Dieser kann den spezifischen Bedarf an elektrischer Energie von derzeit rund 11 bis 15 kWh/kg flüssigen Wasserstoffs auf rund 6,4 kWh/kg verringern. Umweltbilanzen und wirtschaftliche Analysen sind Teil des Projektes. Der Artikel fasst die kurz vor Ende des Projektes verfügbaren Ergebnisse zusammen.

Abstract: Hydrogen liquefaction today is regarded as energy intensive and costly. The IDEALHY project has developed an efficient and cost-effective process for future large-scale plants. It can reduce the specific electricity consumption from current levels of 11 to 15 kWh/kg liquefied hydrogen to about 6.4 kWh/kg. Life cycle and economic assessments are part of the project. The article summarises results shortly before completion of the project.

1. Introduction

Hydrogen is expected to be an important future clean transport fuel. In the absence of a pipeline network, liquid hydrogen (LH_2) can be the most effective way to supply larger refuelling stations in the medium term. However, today hydrogen liquefaction is expensive, energy-intensive and relatively small-scale. For example, the liquefaction plant in Leuna/Germany, one of the most recently commissioned installations, has a capacity of 5 tonnes per day (t LH_2/d) and requires approximately 11.9 kWh/kg LH_2 [1]. Plants in the USA are reported to be between 12 and 15 kWh/kg LH_2 [2]. Large plants with capacities of up to 54 t LH_2/d have been operated in the past in connection with the Apollo project.

The aim of the IDEALHY project (Integrated Design for Efficient Advanced Liquefaction of Hydrogen, November 2011 to October 2013) has been to advance the technology for the liquefaction of hydrogen at scales from 50 t LH_2/d and, especially, to reduce the specific electricity consumption. The main elements of the project are:

- Technology analysis and conceptual liquefaction process assessment,
- Process optimisation, including component development [3],
- Hazard and risk assessment, and mitigation measures [4],
- Life cycle and economic assessment, and
- Planning and preparation of a large-scale demonstration.

This paper introduces the liquefaction process that has been developed and then concentrates on the life cycle and economic assessment. Most of the project reports and a number of conference contributions that focus on individual aspects have been made public [5].

2. The IDEALHY Preferred Process

Several processes proposed or realised in the past were collected and compared using identical boundary conditions and component efficiencies. From this a “Preferred Process” has been developed which promises a power consumption of less than 6.5 $kWh_{el}/kg\ LH_2$.

The main characteristics of the Preferred Process that contribute to an improved overall efficiency are compared with conventional technology in Table 1. Within the Preferred Process, the compressor driving the Brayton cycles is by far the largest power user (about 10 MW_{el}). Efficiency is enhanced by employing a turbo compressor instead of a dry piston or oil lubricated unit. The refrigerant here is Nelium 25, a mixture of 75% helium and 25% neon, combining excellent heat transfer (helium) with a high molecular weight (neon) that is required to make the use of turbo compressors possible. In the future, turbo compressors which operate at higher circumferential speed than today may permit a smaller neon share. Further details of component selection and Preferred Process details are discussed in [6].

Table 1: Comparison between IDEALHY and Existing Processes [7].

	Currently used process	**IDEALHY Preferred Process**
Hydrogen pressure in process	20 bar	80 bar
Pre-cooling	Open LN_2	Mixed refrigerant closed loop
Brayton cycle refrigerant	Hydrogen or helium	Nelium
Brayton cycle compressor	Dry piston compressor or oil lubricated screw compressor	Turbo compressor
Final expansion	Throttle valve or ejector	Gas bearing turbines or piston expander

The process flow diagram is shown in Figure 1. The process can be split into five stages:

- Compression of the feed,
- Chilling,
- Pre-cooling with a mixed refrigerant (MR, consisting of nitrogen, methane, ethane, propane and butane) down to about 130 K,
- Cryogenic cooling with Brayton cycles to 26.8 K, and
- A final expansion and liquefaction stage, resulting in LH_2 at 22.8 K, a para-hydrogen content of 98% and 100% purity.

Compression of the feed from assumed 20 bar to 80 bar and chilling of all streams entering the cold boxes are located in the upper section of Figure 1. Chilling reduces temperatures from assumed ambient 293 K (20°C) to 279 K (6°C). The pre-cooling and cryogenic cooling down to 80 K is located in one cold box, while the last cryogenic cooling stage is located in a separate cold box. Residual impurities are removed at 80 K level in switchable adsorbers.

Cryo-cooling is performed by two overlapping Brayton cycles with a common compression train (Nelium compressor on the top right of Figure 1). In the final step, the hydrogen is liquefied through two expansion turbines (T7 and T8 in Figure 1) from 80 bar to 2 bar.

The outlet stream of turbine T8 contains a certain amount of flash gas, which is warmed up to ambient temperature, compressed, cooled, condensed and throttled back into the storage vessel. Further details on the process stages can be obtained from IDEALHY Deliverable D5.22 [7].

The total electrical power requirement for the Preferred Process has been calculated to about 13.3 MW_{el} for a plant with 50 t LH_2/d capacity. This results in a specific power consumption of 6.4 kWh_{el}/kg LH_2. This figure is valid for the liquefaction process itself including electric motor losses.

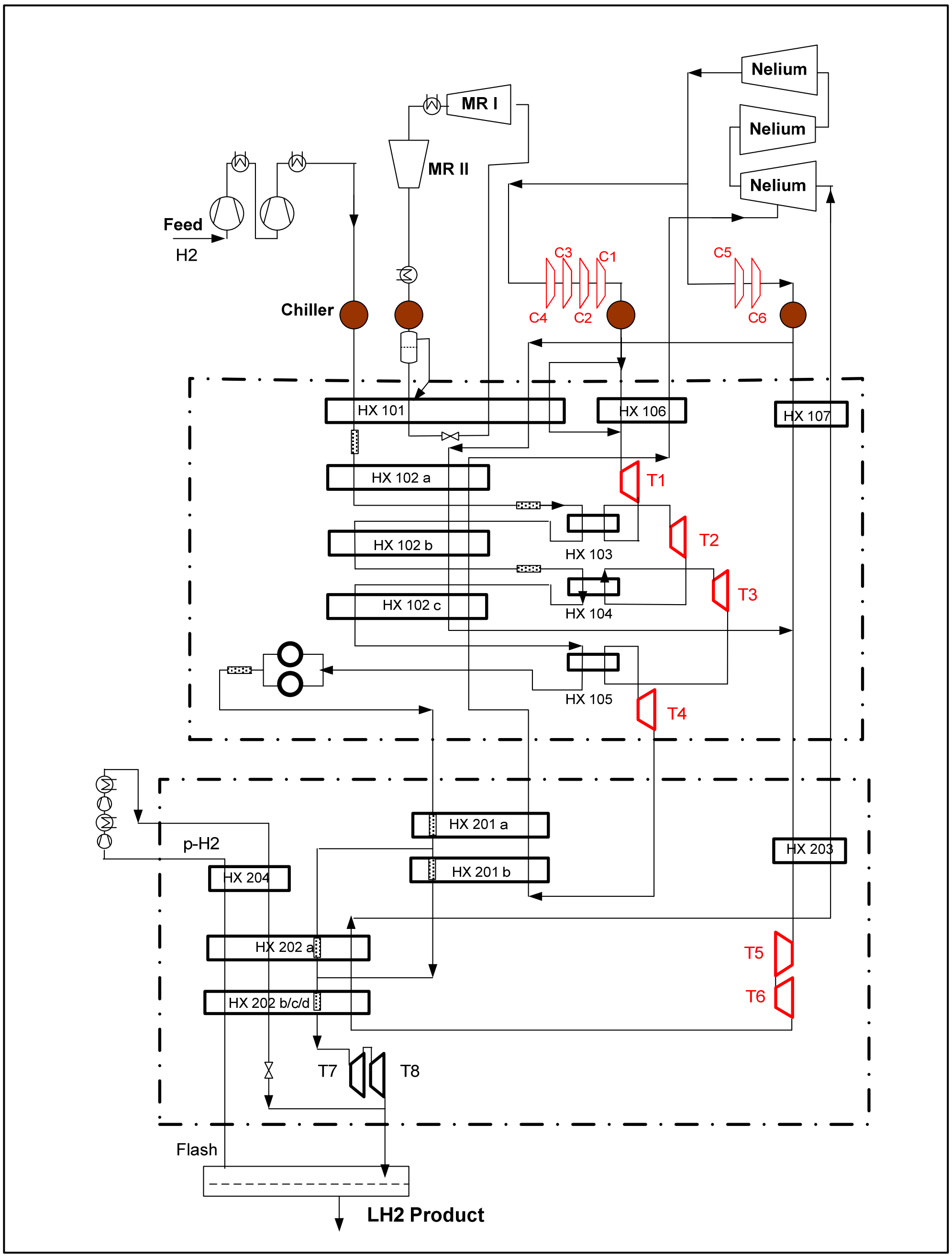

Figure 1: Flow Diagram for the IDEALHY Preferred Process [7].
The upper rectangle depicts the cold box for components above 80 K; the lower one represents the cold box for lower temperatures. Both are vacuum insulated. C = turbo compressor, HX = heat exchanger, p-H2 = para-hydrogen, T = turbo expander.

3. Life Cycle and Economic Assessment

3.1 Overview

The objectives of the life cycle assessment (LCA) and economic assessment have been to evaluate and compare the environmental impacts and economic costs and benefits of relevant pathways for the supply and liquefaction of hydrogen, the delivery of LH_2 to fuelling stations and the subsequent use of re-gasified hydrogen in road passenger vehicles. The comparison is carried out relative to current pathways based on crude oil from conventional sources and compressed gaseous hydrogen. The approach is further explained in the Baseline Results Report; IDEALHY Deliverable D3.13 [8].

Several ways of generating hydrogen are considered [9, 10]:

- Electrolysis with surplus wind electricity;
- Electrolysis with electricity from concentrated solar power;
- Reformation of natural gas, with and without carbon capture and storage (CCS); and
- Gasification of brown coal with and without CCS.

For all elements of the pathways, MS Excel workbooks have been developed. It is planned to publish them on the IDEALHY website [5], in particular the one that maps the liquefaction plant employing the Preferred Process.

At the time of completing this paper, the work that maps the liquefaction plant based on the Preferred Process has almost been completed whereas analysing entire pathways is at an intermediate stage. Therefore, this paper in the following sections focuses on assessing the liquefaction plant. Further findings will be reported in IDEALHY Deliverable D3.16 [11] and Deliverable D3.17 [12].

3.2 Methodology

The specific environmental impact categories that have been selected for the LCA are:

- Primary energy (PE) inputs in the form of energy from depletable resources, such as fossil and nuclear fuels, and
- Prominent greenhouse gas (GHG) emissions consisting of carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O).

The economic costs addressed by this assessment consist of:

- Internal costs, in €, which exclude taxes and financial incentives.

The LCA procedures incorporated in the spreadsheet workbooks are consistent:

- With the Renewable Energy Directive (RED) of the European Commission [13] for regulatory purposes (approximating to attributional LCA) and
- With consequential LCA for policy analysis purposes.

In particular, the RED methodology stipulates the exclusion of total GHG emissions associated with the construction, maintenance and decommissioning of plant, equipment, machinery and vehicles, whereas consequential LCA requires their inclusion. The results presented in this paper are based on the RED methodology.

The estimated emissions of CO_2, CH_4 and N_2O can be converted to equivalent (eq.) CO_2 by means of Global Warming Potentials (GWPs). Values of GWPs depend on the chosen time horizon under consideration. Additionally, these values are subject to revision from time-to-time by the Intergovernmental Panel on Climate Change (IPCC) as scientific understanding improves. In the context of LCA, the GWPs adopted are governed by the choice of methodology. Currently, the RED specifies GWPs of 23 kg eq. CO_2/kg CH_4 and 296 kg eq. CO_2/kg N_2O for a 100 year time horizon based on the IPCC Third Assessment Report [14][1].

3.3 Life Cycle Assessment Results

The analysis in the following assumes a 50 t LH_2/d hydrogen liquefaction plant operating at full load over 8,000 hours per year. Besides the liquefaction *process* as introduced above, the liquefaction *plant* has further requirements:

- Components such as an instruments air compressor, a vacuum pump for the cold boxes, the control system, safety devices, lighting, etc. involve additional power use.
- In order to operate a cooling system that mainly serves the inter- and after-coolers of the compressors, water pumps and /or the fans of a wet cooling tower will consume further electrical power. Water will be evaporated.
- A storage tank for the liquefied hydrogen is necessary. Although it will be very well insulated, a certain influx of heat will occur and result in boil-off, causing a higher mass flow through the flash gas cycle than induced by the Preferred Process.

This means that an estimated further 725 kW_{el} are consumed by the *plant*. In addition, a certain amount of hydrogen is lost, partly due to leakage through the sealing of the feed and flash gas cycle compressors and partly in the course of regenerating the adsorber that collects impurities. In total, this results in about 1.017 kg feed hydrogen being required for 1 kg LH_2 output.

The specific electricity consumption of the *plant* thus amounts to 6.76 kWh_{el}/kg LH_2, which is about 6% more than the 6.4 kWh_{el}/kg LH_2 for the *process*.

However, to counteract this, there are opportunities to reduce specific consumption, depending on the hydrogen generation process upstream of the liquefaction plant:

- High-pressure electrolysers are expected to come on the market in the future. If they provide hydrogen at 80 bar, a feed compressor will not be required. This will save 0.74 kWh_{el}/kg LH_2, reducing the power consumption of the *plant* to just above 6 kWh_{el}/kg LH_2.
- When hydrogen is generated from steam methane reforming and the natural gas is shipped to the site in a liquid state, the 'cold' released on re-gasification can be utilised. Assuming that enough cold is available continuously, it could replace the MR cycle. This would reduce the electrical power for pre-cooling from over 1,380 kW_{el} to just 100 kW_{el}, for a pump circulating nitrogen as a secondary refrigerant. The specific electricity consumption of the *plant* would then be reduced by about 0.62 kWh_{el}/kg LH_2 to below 6.15 kWh_{el}/kg LH_2.
- In order to further reduce electricity consumption, a combination of both options would be desirable. However, steam reformers operate at only 20 – 30 bar.

Considering the above 6.76 kWh_{el}/kg LH_2 electricity consumption as well as the hydrogen losses and water consumption, Table 2 shows the results with respect to depletable PE (fossil and nuclear) and associated total GHG emissions. It demonstrates that the location of the plant is very important

[1] More recent equivalent GWPs of 25 kg eq. CO_2/kg CH_4 and 298 kg eq. CO_2/kg N_2O are given in the IPCC Fourth Assessment Report [15].

since these results are strongly influenced by the actual sources of electricity used, which are assumed to be from the respective national grids.

Table 2: Primary Energy Inputs and Greenhouse Gas Emissions for Selected Locations. Figures for 2009 are based on national energy balances [16], expected figures for EU-27 in 2030 from GEMIS [17].

		2009				2009	2030
Impact Category	**Unit**	**Norway**	**Germany**	**United Kingdom**	**Australia**	**EU-27**	
Specific depletable PE input	MWh / t LH_2	0.49	17.32	18.34	22.39	16.85	11.57
Specific total GHG emissions	kg eq. CO_2 / t LH_2	99	3,572	3,597	6,737	2,442	1,958

3.4 Economic Assessment Results

For a 50 t LH_2/d liquefaction plant, an investment of 105 million €[2], a payback period of 20 years, an internal rate of return of 10% and annual fixed costs for operation and maintenance (O&M) of 4% of the investment are assumed. These figures do not apply to a first-of-its-kind demonstration installation but to a second or third plant. The costs for electricity are set to 100 €/MWh_{el} and those for water to 1.25 €/m^3. Hydrogen losses are accounted for at 2 €/kg feed.

With 8,000 operating hours per year, as above, the figures in Table 3 result, with specific liquefaction costs of about 1.72 €/kg LH_2. When the assumed power costs are halved to 50 €/MWh_{el}, 1.38 €/kg LH_2 follow. For comparison, the costs of hydrogen generation from large-scale steam methane reforming are currently 1.00 – 1.50 €/kg [19].

Table 3: Results of the Economic Analysis of Hydrogen Liquefaction based on the IDEALHY Preferred Process.

Cost Factor	Unit	Costs	Unit	Specific costs	Share in costs
Annuity	million € / a	12.33	€ / kg LH_2	0.74	43%
Fixed O&M costs	million € / a	4.20	€ / kg LH_2	0.25	15%
Variable costs - Electricity - Water - Hydrogen	million € / a	 11.27 0.55 0.24	€ / kg LH_2	 0.68 0.03 0.01	 39% 2% 1%
Total costs	million € / a	28.60			
Total specific costs			€ / kg LH_2	1.72	
			€ cent / kWh LH_2 (LHV)	0.05	

3.5 Discussion

The Preferred Process developed in the IDEALHY project is intended for large plants where power efficiency becomes decisive. It should be usable in plants up to 100 or even 150 t LH_2/d. The Preferred Process has quite a number of internal degrees of freedom which can be adjusted in order

[2] This is an estimate available at the time of completing this paper. More elaborate information on costs will be compiled in IDEALHY Deliverable D2.7 [18].

to obtain an overall optimisation. The optimum choice of parameters will depend mainly on the individual efficiencies of the components, such as compressors and expanders. For this reason, a complete optimisation – which should lead to even lower power consumption than currently estimated – can only be performed after additional development work has been carried out. An approach towards optimisation and towards establishing a demonstration plant is outlined in IDEALHY Deliverable D5.22 [7].

The figures related to depletable PE input and GHG emissions in Table 2 need to be considered as estimates. Rather than the absolute values, the significant differences between possible sites for a plant (country or region) should be noted. Since, for example, the PE mix for producing electricity in Norway is largely based on hydro, the resulting burden related to these two impact categories is low, in particular compared to Australia with a large share of brown coal power plants.

Regarding Germany, the share of renewable energy in electricity generation has increased from 16.4% to 22.9% in 2012. Therefore, the specific PE input and GHG emissions related to operating a liquefaction plant would be significantly lower today and will be even lower when a large-scale plant based on the Preferred Process becomes operational. It is sometimes argued, however, that the European electricity grid is integrated so closely that it is advisable to refer to EU averages rather than national figures. Comparing the 2009 and 2030 figures for EU-27 in Table 2 establishes a reduction of PE input by about 30% and of GHG emissions by about 20%.

Electricity and capital investment play major roles with respect to costs of hydrogen liquefaction plants. Concerning the former, on-site power generation could be an option, given the high load factor. This would save grid fees and de-couple GHG emissions from the national or EU-mix.

The economic competitiveness of highly efficient large-scale hydrogen liquefaction and its overall benefits with respect to PE input and GHG emissions will depend on the results of comparison with other pathways for fuel delivery [12].

4. Conclusion

The outcomes of the IDEALHY project bear the potential to revise the notion that liquefaction of hydrogen is inefficient and costly. A promising technical concept has been developed. The next crucial step consists in raising support for a demonstration plant in parallel to fostering the improvements of key components.

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6. References

[1] M. Bracha, L. Decker, Großtechnische Wasserstoffverflüssigung in Leuna, Deutsche Kälte-Klima-Tagung, 2008.

[2] R. Drnevich, Hydrogen Delivery – Liquefaction & Compression, Strategic Initiatives for Hydrogen Delivery Workshop, 7 May 2003, available at www1.eere.energy.gov/hydrogenandfuelcells/pdfs/liquefaction_comp_pres_praxair.pdf [accessed 23 September 2013].

[3] M. Börsch, Report on Components Development, Deliverable 2.5 of the IDEALHY Project (Publishable Summary), 2013, to be published at [5].
[4] B.J. Lowesmith, G. Hankinson, S. Chynoweth, Safety Issues of the Liquefaction, Storage and Transportation of Liquid Hydrogen: Studies in the IDEALHY Project, International Conference on Hydrogen Safety, Brussels, 2013.
[5] www.idealhy.eu/publications.
[6] H. Quack, I. Seemann, M. Klaus, Ch. Haberstroh, D. Berstad, H.T. Walnum, P. Nekså, L. Decker, Selection of Components for the IDEALHY Preferred Cycle for the Large Scale Liquefaction of Hydrogen, Cryogenic Engineering Conference and International Cryogenic Materials Conference, Anchorage, 2013.
[7] D. Berstad, L. Decker, A. Elliott, H. Quack, H. T. Walnum and P. Nekså, Schedule for Demonstration Plant including Options for location, Deliverable 5.22 of the IDEALHY Project, 2013, available at [5].
[8] N. D. Mortimer, O. Mwabonje and J. H. R. Rix, Baseline Results Report, Deliverable 3.13 of the IDEALHY Project, 2012, available at [5].
[9] K. Stolzenburg and R. Mubbala, Liquid Hydrogen Pathway Report, Deliverable 3.14 of the IDEALHY Project, 2012, available at [5].
[10] N. D. Mortimer, C. Hatto, O. Mwabonje and J. H. R. Rix, Hydrogen Production and Utilisation Report, Deliverable 3.15 of the IDEALHY Project, 2013 (in preparation), to be published at [5].
[11] K. Stolzenburg and R. Mubbala, Hydrogen Liquefaction Report, Deliverable 3.16 of the IDEALHY Project, 2013 (in preparation), to be published at [5].
[12] N. D. Mortimer, C. Hatto, O. Mwabonje and J. H. R. Rix, Techno-Economic Analysis and Comparison Report (Publishable Summary), Deliverable 3.17 of the IDEALHY Project, 2013 (in preparation), to be published at [5].
[13] European Commission, Directive 2009/28/EC of the European Parliament and of the Council of 23 April '09 on the Promotion of the Use of Energy from Renewable Sources and Amending and Subsequently Repealing Directives 2001/77/EC and 2003/30/EC, Brussels, Belgium, 5 June 2009.
[14] Intergovernmental Panel on Climate Change, Climate Change 2001: The Scientific Basis; Contribution of Working Group 1 to the Third Assessment Report of the IPCC, Cambridge University Press, Cambridge, United Kingdom, 2001, www.grida.no/publications/other/ ipcc_tar/ [accessed 23 September 2013].
[15] Inter-governmental Panel on Climate Change, Climate Change 2007: Synthesis Report: Fourth Assessment Report, 2007, www.ipcc.ch/publications_and_data/ar4/wg1/en/contents.html [accessed 23 September 2013].
[16] Approximate analysis of electricity generation statistics based on: International Energy Agency, Energy Balances, Paris, France; e.g. link for Germany: http://www.iea.org/statistics/ statisticssearch/report/?country=GERMANY&product=balances&year=2009 [last accessed 23 September 2013].
[17] Internationales Institut für Nachhaltigkeitsanalysen und -Strategien, Global Emissions Model for Integrated Systems: GEMIS Version 4.81, Darmstadt, Germany, 2013, www.iinas.org.
[18] Efficiency and Cost Calculations (Publishable Summary), Deliverable 2.7 of the IDEALHY Project, 2013 (in preparation), to be published at [5].
[19] References quoted in: T. Grube, B. Höhlein, Kosten der Wasserstoffbereitstellung in Versorgungssystemen auf Basis erneuerbarer Energien, in: J. Töpler, J. Lehmann (Editors), Wasserstoff und Brennstoffzelle, Springer, Berlin, October 2013.