Meeting net carbon-neutral targets by 2050 requires new clean energy sources. Hydrogen can be produced as a zero-carbon fuel and will be a major contributor to meeting this objective. The significance of H₂ is illustrated by the formation of the Hydrogen Council in Q1 2017; the preparation of “The future of hydrogen” report by the IEA for the G20 meeting in Q2 2019; and the publication of “A hydrogen strategy for a climate-neutral Europe” by the European Commission in Q3 2020.

Zero-carbon H₂ can be produced through electrolysis of water using renewable electric power (green H₂); or through steam or autothermal reforming of natural gas, or gasification of coal (blue H₂). Blue H₂ production processes require carbon capture and storage to secure zero-carbon status.

Cost reductions will be needed for H₂ to displace fossil fuels. While technology advances in electrolyzers and lower-cost renewable power will, over time, reduce the cost of green H₂, the bulk of zero-carbon production will initially be blue H₂, which, at present, can be produced at greater scale and lower cost.

Demand for H₂ will be impacted by natural gas and carbon pricing. The latter provides governments the leverage to impact the rate and extent of H₂’s displacement of fossil fuels and influence H₂’s contribution to meeting the 2050 objectives.

**Liquid H₂ production.** Producing liquid H₂ (LH₂) reduces volumes by 800 and reduces storage and distribution costs. Liquefaction will, therefore, be central to many H₂-based schemes. All existing LH₂ plants are small (typically < 15,000 metric tpy) and use a combination of liquid nitrogen evaporative precooling and Claude H₂ expander cycle for final liquefaction. The requirement for liquid nitrogen means that larger LH₂ plants require integration with, or close proximity to, an air separation unit. Aside from constrained supply logistics, use of liquid nitrogen is thermodynamically inefficient, resulting in a liquefaction power demand of 10 kWh/kg–15 kWh/kg LH₂ on existing plants, equivalent to 30%–45% of the energy content of the H₂ feed. This level of power demand is unsustainable at the higher plant capacities required to produce bulk LH₂.

This article describes a low equipment count precooling concept integrated with an H₂ expander cycle for final cooling and liquefaction. The dual-expander precooling configuration uses natural gas, a single, low-cost refrigerant, and avoids the infrastructure needed to transfer, store and blend conventional liquid refrigerants. For final cooling and liquefaction, the cycle uses the H₂ feed as refrigerant and avoids external, high-cost refrigerants such as neon and helium.

Overall, the scheme achieves a favorable balance between capital and operating costs by combining simplicity, less equipment and low capital cost with a competitive power demand.

**Basis of design.** To provide a reference point for the data presented in this article, the basis of design is provided in TABLE 1. The basic premise is that the H₂ feed derives from either a hydrocarbon-based production plant, such as a steam reformer (blue H₂) or from electrolyzers (green H₂).

In addressing the plant production capacity and the desire to meet 2050 carbon targets, the authors have reflected the need to make a significant impact on consumption of liquid transportation and industrial fuels, which are presently used at an approximate rate of 4,000 metric MMtpy. To provide perspective, the Hydrogen Council projects H₂ consumption for transportation and industrial fuel of 38 exajoules per year in 2050 (1,050 metric MMtpy of oil equivalent). This would require an additional installed H₂ plant capacity
of approximately 400 metric MMtpy. If only 10% of the H₂ was required as LH₂, then this would require approximately 400 liquefaction plants, each with a capacity of 100,000 metric tpy.

Based on these challenging capacity parameters, the design capacity was selected as 100,000 metric tpy, which approximates the availability of maximum-sized electric-motor-driven H₂ compression equipment.

The specified H₂ arrival pressure of 25 bar is typical of the pressure of H₂ supply from steam reforming H₂ plants and existing PEM electrolyzers. This pressure affects the overall power requirement of the LH₂ plant.

It has been assumed that seawater at 20°C will cool a closed circuit of treated water cooling the process coolers and heat exchangers within the LH₂ plant.

**Methane precooling H₂ liquefaction.** A schematic of the overall liquefaction process is shown in FIG. 1. The illustrated concept integrates a dual-expander methane precooling process with a cold-end H₂ expander design. The precooling, however, could be combined with other cold-end arrangements using, for example, helium and/or neon refrigerants.

The precooling circuit, shown in red, was originally developed and patented for LNG application. The cold-end H₂ cycle is shown in green.

**Methane expander cycle.** The methane make-up gas to the precooling system is assumed to be pipeline-quality natural gas. Given the limited volume of the initial fill and make-up quantities, molecular sieve pretreatment provides a practical solution to remove carbon dioxide (CO₂), water vapor and residual hydrocarbons that would freeze in the liquefaction system. LH₂ plants approximate to LNG liquefaction or regasification facilities and with access to pretreated LNG feed gas or regasified LNG may not require pretreatment facilities.

The methane cycle operates as a closed circuit with the methane refrigerant compressed and circulated by the electrically driven recycle compressor, CP1. The compressed gas then flows to the cold box comprising multi-passage brazed aluminium heat exchangers, which cool the H₂ feed and the circulating H₂ refrigerant streams to approximately –160°C. After preliminary chilling in the cold box, the compressed methane flows to the expander wheels of the two expander-compressors, EC1/EC2. The high-temperature expander, EC1, typically discharges at 25 bar/–50°C, and the low-temperature expander, EC2, discharges at 10 bar/–125°C. The cold outlet streams from EC1/EC2 return to the cold box, where they cool the H₂ feed and the circulating refrigerant streams to approximately –120°C. EC1/EC2 are loaded by compressor wheels in series with the main recycle compressor, as illustrated in FIG. 2; they compress the circulating methane to 70 bar–100 bar at the inlet to the cold box.

The low-temperature expander, EC2, operates in partially liquefying mode and efficiently converts latent heat into mechanical work, improving cycle efficiency. This partial liquefaction (FIG. 2), which uses well-proven expander technology, reduces the total power demand of the H₂ liquefaction process and is a distinctive feature.

The condensed liquid formed in EC2 is separated from the gas phase in separator SP1 and is then flashed to near-atmospheric pressure. The resulting two-phase methane stream is evaporated and reheated by further heat exchange
with the H₂ feed and the circulating H₂ refrigerant streams, which are thereby cooled to –155°C. The evaporated and reheated flashed gas stream is recaptured to the system by the flash gas compressor, CP3, and routed together with the outlet gas stream from SP1 to CP1 suction for recompression and return to the expanders for further cooling duty.

The typical impact on precooling power demand of increasing liquids content at the outlet of EC2 is shown in Fig. 3. Power consumption is reduced as the liquid content of the expander outlet increases.

H₂ cooling and liquefaction. The H₂ cooling, liquefaction and refrigerant flows are shown schematically in Fig. 1. The two separate systems include the H₂ feed stream and the H₂ expander refrigeration cycle.

Feed gas H₂ is pretreated in molecular sieve adsorbers (AD1) to remove impurities, such as traces of CO₂ and water vapor, and fed to the cold box inlet. After precooling by the methane expander cycle to –150°C, the H₂ feed is routed to parallel, second-stage, regenerative cryogenic adsorbers (AD2) to remove residual trace impurities, mainly nitrogen and hydrocarbons. The adsorbers accomplish several key functions:

- Provide protection to the downstream ortho-H₂ to para-H₂ conversion catalyst
- Prevent freezing of the trace impurities in the downstream low-temperature regime
- Produce an ultra-high-quality LH₂ product, as required for certain industrial applications.

Multiple stages of ortho-H₂ to para-H₂ catalyst are provided, integrated with the cold end of the brazed aluminium exchanger system. Near-complete conversion of the ortho-H₂ to the para-allo-trope is essential prior to LH₂ storage, as residual ortho-H₂ to para-H₂ conversion in storage is exothermic, causing significant boil-off losses.

After ortho-H₂ to para-H₂ conversion and further expansion, the H₂ feed leaves the cold box as liquid H₂ at 2 bar/–250°C. It is depressurized, if required, and routed to storage.

The H₂ refrigerant system comprises a separate closed-cooling circuit with multiple expander stages that provide low-temperature refrigeration for liquefaction of the feed H₂. The H₂ in this second H₂ system consists essentially of unequilibrated “normal H₂” (75% ortho-H₂ plus 25% para-H₂).

The H₂ recycle compressor (CP2) delivers precooled H₂ to the expanders at 50 bar, with the first expander (EH1) typically discharging at 4.5 bar/–216°C and the last expander (EH2) discharging at 2.7 bar/–249°C. The expander outlet streams provide counter-current cooling of the H₂ feed gas and are then routed to the CP2 suction and returned to EH1/EH2 for further cooling duty. EH1/EH2 recover power, which is used to reduce the overall power demand of the system.

The H₂ expanders are both expected to require more than one impeller in series due to enthalpy drop limitations.

**Design issues.** In developing the overall system design, the authors have noted the difficulty in reconciling the available data on the properties of ortho-H₂. This has been addressed by modifying the reference state of ortho-H₂ in the National Institute of Standards and Technology’s REFerence fluid PROPer-ties (REFPROP) database.

**Alternative configurations.** A number of variants to the described system have been investigated, as described in the following sections.

**Nitrogen precooling refrigerant.** The use of nitrogen (or a nitrogen/methane mixture) in place of methane in the precooling system has been investigated. With nitrogen, a lower precooled temperature of approximately –190°C may be attained, but the overall specific power of the liquefaction process was found to be up to 10% higher than with methane. However, due to the lower attainable final temperature of the precooling stage (around –190°C), there is a reduction in the power requirement of around 20% for the relatively expensive H₂ compressor CP2, which may be significant in terms of capital cost.

**Low-temperature H₂ recycle compression.** Fig. 4 shows a variant of the H₂ liquefaction cycle that operates the H₂ recycle compressor with a significantly sub-ambient suction temperature. The recycled H₂ enters the first part of compressor CP2 typically at a temperature of –120°C. Alternatively, the inlet stream to compressor CP2 may be taken directly from the outlet stream of the H₂ liquefier unit at a lower temperature.

Depending on the inlet temperature of compressor CP2, its power consumption may be reduced by approximately 50%, relative to the configuration with a near-ambient inlet temperature, as shown in Fig. 1. While this leads to an approximately equivalent increase in the power demand for the precooling methane compression, operation of the H₂ compressor with a sub-ambient inlet temperature has advantages for H₂ liquefaction:

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**FIG. 3.** Relative power vs. liquid fraction from low-pressure expander.
• H₂ compression generally requires use of reciprocating compressors, as the density of H₂ is low for use in centrifugal compressors. Taking into consideration the relatively high investment and operational costs of reciprocating machines, the reduction in power requirement and cost of reciprocating compressors arising from the use of a sub-ambient inlet temperature may be significant. Furthermore, the differential power requirement imposed on the methane precooler circuit is then provided, in the configuration shown in FIG. 4, by relatively inexpensive centrifugal machines.

• Operation of the H₂ compressor with a sub-ambient inlet temperature increases the inlet density. As an example, at –120°C the inlet density is approximately twice the density at ambient temperature, facilitating the future use of lower-cost centrifugal compression in H₂ liquefaction.

Process performance. A realistic expected specific power of the H₂ liquefaction process is in the range of 6.7 kWh/kg–7.5 kWh/kg LH₂, depending significantly on the LH₂ delivery pressure and the efficiencies of the compressors and expanders. Other factors affecting the specific power include the disposition of the ortho-para conversion catalyst (in the heat exchangers, or external) and the selected precooler refrigerant (methane, nitrogen or combinations of these).

Relative process performance and configuration issues. Recognizing the inadequacies of present LH₂ technology for the scale required to support the H₂ economy, various configurations have been investigated by a number of parties. The IDEALHY consortium published data on a survey of various configurations of existing operating LH₂ facilities and new concepts. An extract from the consortium’s data is provided in TABLE 2, which shows a drive to improvement in both the precooler and liquefaction circuits. A measure of caution is required in interpreting this data, as the bases of design for the various processes are not identical and predicted machine efficiencies vary significantly.

In terms of power demand, the most promising schemes indicate a move towards single mixed-refrigerant (SMR) precooling and use of efficient, though expensive refrigerants, neon and helium, for the liquefaction stage. Other work indicates that SMR precooling combined with a high-pressure Claude H₂ cycle achieves power demand in the range of 6 kWh/kg–7 kWh/kg.

The authors sought to improve on these configurations with the following objectives:

- Simplify the plant and its operation
- Reduce capital cost
- Achieve a competitive power demand.

The described scheme achieves these objectives:

- The major equipment count for methane precooling (19 items) is > 25% lower than SMR processes (26 items), as refrigerant storage, blending and

### TABLE 2. Current technology and proposed new developments

<table>
<thead>
<tr>
<th>Technology</th>
<th>Status</th>
<th>Precooling cycle</th>
<th>Cryo-cooling and liquefaction cycle</th>
<th>Power demand, kWh/kg</th>
</tr>
</thead>
<tbody>
<tr>
<td>Linde–Ingolstadt</td>
<td>Operating</td>
<td>LN₂</td>
<td>H₂–Claude</td>
<td>13.6</td>
</tr>
<tr>
<td>Linde–Leuna</td>
<td>Operating</td>
<td>LN₂</td>
<td>H₂–Claude</td>
<td>11.9</td>
</tr>
<tr>
<td>Air Products</td>
<td>6 × operating</td>
<td>LN₂</td>
<td>H₂–Claude</td>
<td>12–15</td>
</tr>
<tr>
<td>Praxair</td>
<td>4 × operating</td>
<td>LN₂</td>
<td>H₂–Claude</td>
<td>12.5–15</td>
</tr>
<tr>
<td>Air Liquide</td>
<td>5 × operating</td>
<td>LN₂</td>
<td>H₂–Claude</td>
<td>12–15</td>
</tr>
<tr>
<td>WE-NET Study</td>
<td>Study</td>
<td>LN₂</td>
<td>H₂–Claude</td>
<td>8.53</td>
</tr>
<tr>
<td>WE-NET Study</td>
<td>Study</td>
<td>LN₂</td>
<td>Ne–Reverse Brayton</td>
<td>8.69</td>
</tr>
<tr>
<td>WE-NET Study</td>
<td>Study</td>
<td>LN₂</td>
<td>Ne–Reverse Brayton</td>
<td>8.58</td>
</tr>
<tr>
<td>Quack Study</td>
<td>Study</td>
<td>Propane</td>
<td>Ne/He–Reverse Brayton</td>
<td>6.93</td>
</tr>
<tr>
<td>Valenti and Macchi Study</td>
<td>None</td>
<td>He–Reverse Brayton</td>
<td></td>
<td>5.29</td>
</tr>
<tr>
<td>Sintef Study</td>
<td>Study</td>
<td>Mixed refrigerant</td>
<td>Ne/He–Reverse Brayton</td>
<td>6.2–6.5</td>
</tr>
<tr>
<td>Shimko Study</td>
<td>Study</td>
<td>None</td>
<td>He–Reverse Brayton</td>
<td>8.73</td>
</tr>
<tr>
<td>IDEALHY Study</td>
<td>Study</td>
<td>Mixed refrigerant</td>
<td>Ne/He–Reverse Brayton</td>
<td>6.4</td>
</tr>
</tbody>
</table>

FIG. 4. Low-temperature H₂ compression.
transfer facilities are not required

- Experience from the LNG industry indicates the power demand of dual-methane expander refrigeration incorporating partial liquefaction in the low-temperature expander is up to 10% lower than a typical SMR process, > 10% lower than triple-expander nitrogen processes and > 20% lower than dual-expander nitrogen processes
- Operations are simplified as precooling refrigerant composition adjustments are not required; the methane precooling system is self-adjusting
- Both the precooling and liquefaction refrigeration cycles utilize low-cost, readily available refrigerants that do not require storage or handling facilities
- The H₂ liquefaction cycle does not require expensive, hermetically sealed compression and expander equipment of the kind that is often required to reduce leakage of more costly refrigerants, such as helium and neon
- Considering the expected low specific power and low equipment count, the proposed plant arrangement is highly competitive with the projections shown in Table 2.

Cost estimate. A cost estimate was developed for the inside battery limit liquefaction plant. The inputs to this estimate were a sized equipment list and supplier pricing data. The equipment list is provided in Table 3.

The cost estimate for the 100,000-tpy production unit is US$380 MM, based on a Q1 2021 instant execution basis. The following methodology was applied to develop the cost estimate:

- **Mechanical equipment.** A costed equipment list was developed. The majority of equipment items were priced based on budget proposals received from equipment suppliers. Budget proposals from suppliers were benchmarked against proposals and actual costs from other projects to arrive at an overall anticipated cost. An allowance for 2 yr of spares and an allowance for first fill of lubricants were also included in the mechanical equipment cost.

- **Direct field cost.** This cost was factored from the mechanical equipment cost based on data from similar cryogenic projects of similar power demand. The overall project cost was obtained by addition of contingency, technology license fee and contractor’s EPC services and margin to the direct field cost.

- **Estimate exclusions.** The following costs were excluded from the estimate:
  - Owner’s costs
  - Costs for fees and permits
  - Finance costs
  - Insurance costs
  - Customs and import duties
  - Taxes
  - Forward escalation
  - Liquid H₂ storage costs
  - Offsites and utilities costs.

**Table 3.** List of major equipment for ISBL H₂ liquefaction plant

<table>
<thead>
<tr>
<th>Type of equipment</th>
<th>Number of items</th>
</tr>
</thead>
<tbody>
<tr>
<td>CP1 CH₄, recycle compressor, including motor</td>
<td>1</td>
</tr>
<tr>
<td>CP2 H₂, recycle compressor, including motor</td>
<td>Multiple reciprocating</td>
</tr>
<tr>
<td>CP3 CH₄, flash gas compressor, including motor</td>
<td>1</td>
</tr>
<tr>
<td>EC1 CH₄, expander-compressor</td>
<td>1</td>
</tr>
<tr>
<td>EC2 CH₄, expander-compressor</td>
<td>1</td>
</tr>
<tr>
<td>EH1 H₂, expander</td>
<td>&gt;2</td>
</tr>
<tr>
<td>EH2 H₂, expander</td>
<td>&gt;2</td>
</tr>
<tr>
<td>Cold box</td>
<td>1</td>
</tr>
<tr>
<td>SPI separator</td>
<td>1</td>
</tr>
<tr>
<td>AD1 H₂, feed gas adsorbers</td>
<td>2</td>
</tr>
<tr>
<td>AD2 cryogenic adsorbers</td>
<td>2</td>
</tr>
<tr>
<td>OP ortho-para conversion reactors</td>
<td>Integrated with BAHX</td>
</tr>
</tbody>
</table>

**Takeaway.** H₂ liquefaction by methane and nitrogen precooling processes incorporating a partially liquefying expander holds considerable promise as a low-cost, high-efficiency system. Significant engineering work, relevant to H₂ application, was carried out as part of the scheme’s earlier development for LNG application.

Early-stage use of H₂ as an energy vector, prior to its more widespread deployment, may require plants of a capacity below the 100,000-metric-tpy capacity selected as the basis of design. The application of the described process, given its simplicity and particular balance between operating and capital cost, may be enhanced at these lower plant capacity levels.

A patent for the described precooling process has been applied for covering methane and nitrogen refrigerants and mixtures of the two. A patent application has also been made for the use of the low-suction-temperature H₂ recycle compression system.

**LITERATURE CITED**


**NOTE**

The methane expander precooling system (OHL—Optimized Hydrogen Liquefaction) described in this article is owned and licensed by Gasconsult Ltd. Data on equipment performance was provided by BHGE.
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