Roadmap to economically viable hydrogen liquefaction

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Abstract

The distribution of hydrogen in liquid state has several advantages because of its higher volumetric density compared to compressed hydrogen gas. The demand for liquid hydrogen (LH2), particularly driven by clean fuel cell applications, is expected to rise in the near future. Large-scale hydrogen liquefaction plants will play a major role within the hydrogen supply chain. The barriers of built hydrogen liquefiers is the low exergy efficiency and the high specific liquefaction costs. Exergy efficiency improvements, however, are limited by economic viability. The focus of this paper is to present a roadmap for the scale-up of hydrogen liquefaction technology, from state-of-the-art plants to newly developed large-scale liquefaction processes. The work is aimed at reducing the specific liquefaction costs by finding an optimal trade-off between capital costs and operating costs. To this end, two developed hydrogen liquefaction processes were optimized for specific energy consumption and specific liquefaction costs, showing the potential to reduce the specific liquefaction costs by 67% for a 100 tpd LH2 plant compared to a conventional 5 tpd LH2 plant while achieving a specific energy consumption between 5.9 and 6.6 kWh per kg LH2 with technology that is or will be available within 5 years. The results make liquid hydrogen a viable distribution route for hydrogen for mobility.

Introduction

Air pollution and global warming coming along with global- ization, a growing world population and its ambition for higher living standard menace the future of mankind. For the last 200 years mobility, heating and power generation relied mainly on combustion of hydrocarbons causing carbon dioxide, soot and other pollutants. Carbon dioxide with more than 80% share is by far the main greenhouse gas (GHG) [1]. Transport, being responsible for one third of total GHG emission in the US [1] and about 20% in the EU, is the only major sector in the EU where greenhouse gas emissions are still rising [2]. Any technological progress in combustion engines has been outbalanced by an increasing individual mobility and the demand for larger and heavier vehicles. In recent years, focus has therefore been set on individual cars accounting for about 75% of all CO2 emissions coming from transport [2].

Hydrogen has come into focus as a potential future energy carrier as it reacts in a fuel cell with atmospheric oxygen releasing only electric energy, heat and water. Fuel cells can...
be used for mobility and stationary applications \[3\]. Several national initiatives, for instance in California, Japan and Germany \[3–5\], promote and support the installation of hydrogen refilling station (HRS) networks; car industries such as Hyundai and Toyota \[5\] have started the commercial production of fuel cell electric vehicles (FCEV) running on hydrogen, others such as Honda and Mercedes will follow \[5\].

These cars are equipped with gas tanks for highly compressed gaseous hydrogen (CGH\(_2\)) at 700 bar and near to ambient temperature \[6\]. Most of the refilling stations also operate on CGH\(_2\) because of its flexible availability and its low distribution costs for small quantities in short range. Individual mobility requires a minimum number of refilling stations even at a small number of FCEVs being operated. A reasonable network density of hydrogen refilling stations (HRS) requires a high investment in infrastructure e.g. for up to 400 HRS to be built in Germany by 2023 \[4\] with a total estimated investment of approximately €400 million. Starting on a small number of fuel cell cars leads to a very low traffic frequency at the HRS \[6\], causing significant amortization cost. A focus on vehicles with higher daily consumption would reduce this problem and bring down more quickly the cost in the hydrogen supply chain. Table 1 shows typical consumption rates for different vehicle types assumed in this paper based on \[3,5,6\], and estimates their resulting number being served by one CGH\(_2\) trailer, one liquid hydrogen (LH\(_2\)) trailer, one hydrogen source of 5 tons per day (tpd), for instance \[7\], and one of 50 tpd.

<table>
<thead>
<tr>
<th>Vehicle type</th>
<th>Assumed H(_2) consumption (tpd) [3,5,6]</th>
<th>Trailer [3]</th>
<th>Hydrogen source</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>CGH(_2) 1 ton H(_2) 3.5 tons H(_2)</td>
<td>LH(_2) 5 tpd 50 tpd</td>
<td></td>
</tr>
<tr>
<td>Passenger car</td>
<td>0.0004</td>
<td>2500</td>
<td>12,500</td>
</tr>
<tr>
<td>Bus/Truck</td>
<td>0.03</td>
<td>33</td>
<td>167</td>
</tr>
<tr>
<td>Train</td>
<td>0.25</td>
<td>4</td>
<td>20</td>
</tr>
<tr>
<td>Coastal ship</td>
<td>2</td>
<td>0.5</td>
<td>2.5</td>
</tr>
<tr>
<td>Large ship</td>
<td>10</td>
<td>0.1</td>
<td>0.5</td>
</tr>
</tbody>
</table>

Buses, trains and maybe ferry services are ideal candidates to establish a low cost hydrogen infrastructure for a hydrogen based green mobility. Due to their all day long operation they require significant amounts of fuel. According to Table 1, each fuel cell bus consumes in average 75 times and each train about 400–600 times of hydrogen compared to a fuel cell car [6], bringing down specific amortization costs for infrastructure to an acceptable level. One or two filling stations can serve all fleet vehicles overnight. The network density is not that of importance, instead these larger HRS can serve as distribution bases for relatively cheap hydrogen to car refilling stations. Production numbers of buses and trains are not so high allowing return on development already at smaller series. And space for fuel storage is also of minor priority.

Other fleets of fuel cell vehicles, such as trucks for retail and logistics, taxi and car sharing can also help to bring up the number of hydrogen consumers, keeping needs on infrastructure low. Ferry services [5] and coastal shipping would even inflate the hydrogen mobility. Assuming the estimate in Table 1, each ship would consume as much hydrogen as five thousand cars. Ship HRS infrastructure could in turn become the basis for a bus and train network. Although non private traffic contributes only to 25% percent of the total transport emissions, starting in public and logistics transport allows a much faster and economically viable market development in hydrogen mobility.

Distributing and storing the required hydrogen as a cryogenic liquid offers several advantages compared to CGH\(_2\). Because of the significantly higher volumetric density of liquid hydrogen (LH\(_2\)), the transportable load per LH\(_2\) trailer is significantly higher than in a CGH\(_2\) trailer \[3\], bringing down transport cost and trailer frequency at the station. Compared to CGH\(_2\), the delivery of LH\(_2\) becomes increasingly cost-efficient for larger transport volumes and over longer transport distances \[3\], as required by hydrogen mobility. Further on, the liquid hydrogen comes in guaranteed clean condition as any impurity will be frozen out in the liquefer plant.

The footprint of storage and infrastructure on the filling station is much smaller when compressing the cold hydrogen directly into the vehicles CGH\(_2\) or LH\(_2\) tank. The evaporation rate of the liquid hydrogen is of minor concern as the consumption rate is high enough for a regular operation.

Liquid hydrogen is produced by the cooling, expansion and the liquefaction of an expanded gaseous hydrogen feed gas stream from ambient conditions to a temperature of about 20 K [8]. The principles of hydrogen liquefaction and installed industrial liquefaction processes are described extensively in literature \[7–12\]. The hydrogen cooling in built industrial hydrogen liquefaction processes is typically performed in two refrigeration steps. For the hydrogen precooling to an intermediate temperature of about 80 K, a liquid nitrogen (LN\(_2\)) stream is used. For the cryogenic hydrogen cooling between 80 K and a liquefaction temperature of about 20 K, only helium and hydrogen are available as pure refrigerant fluids for a cryogenic refrigeration cycle [8]. A further challenge of industrial hydrogen liquefiers is the required catalytic ortho-para-hydrogen conversion [8,13].

The relatively low exergy efficiency of installed hydrogen liquefaction plants is the main draw back of a LH\(_2\) supply infrastructure. The specific energy consumption SEC of a state-of-the-art 5 tpd LH\(_2\) hydrogen liquefier with LN\(_2\) precooling is about 10 kWh per kg LH\(_2\) [14,15]. The future hydrogen mobility market will ask for large-scale hydrogen liquefaction plants with a significant improvement in exergy efficiency. Therefore, several studies for future large-scale hydrogen liquefaction plants were published since the late 1970s. An encompassing literature review is given in Refs. \[16–18\]. The majority of these publications focused on

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