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Process optimization for large-scale hydrogen liquefaction

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ABSTRACT

The investment in the hydrogen infrastructure for hydrogen mobility has lately seen a significant acceleration. The demand for energy and cost efficient hydrogen liquefaction processes has also increased steadily. A significant scale-up in liquid hydrogen (LH₂) production capacity from today's typical 5–10 metric tons per day (tpd) LH₂ is predicted for the next decade. For hydrogen liquefaction, the future target for the specific energy consumption is set to 6 kWh per kg LH₂ and requires a reduction of up to 40% compared to conventional 5 tpd LH₂ liquefiers. Efficiency improvements, however, are limited by the required plant capital costs, technological risks and process complexity. The aim of this paper is the reduction of the specific costs for hydrogen liquefaction, including plant capital and operating expenses, through process optimization. The paper outlines a novel approach to process development for large-scale hydrogen liquefaction. The presented liquefier simulation and cost estimation model is coupled to a process optimizer with specific energy consumption and specific liquefaction costs as objective functions. A design optimization is undertaken for newly developed hydrogen liquefaction concepts, for plant capacities between 25 tpd and 100 tpd LH₂ with different precooling configurations and a sensitivity in the electricity costs. Compared to a 5 tpd LH₂ plant, the optimized specific liquefaction costs for a 25 tpd LH₂ liquefier are reduced by about 50%. The high-pressure hydrogen cycle with a mixed-refrigerant precooling cycle is selected as preferred liquefaction process for a cost-optimized 100 tpd LH₂ plant design. A specific energy consumption below 6 kWh per kg LH₂ can be achieved while reducing the specific liquefaction costs by 67% compared to 5 tpd LH₂ plants. The cost targets for hydrogen refuelling and mobility can be reached with a liquid hydrogen distribution and the herewith presented cost-optimized large-scale liquefaction plant concepts.

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Introduction

There is a growing worldwide concern for the impact of climate change and pollution. Alternative clean energy fuels

for the mobility and stationary power sector are an option to meet the targets for future greenhouse gas emissions and global warming, as indicated at the Paris Climate Conference in 2015 [1]. Hydrogen in combination with fuel cell technology is one of the most promising energy carriers [2]. Fuel cell

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electric vehicles (FCEV) produce clean water as exhaust and are capable of overcoming many disadvantages that are associated with battery powered electric vehicles, e.g. limited range and high charging time [3,4]. The investments in the hydrogen infrastructure for clean energy applications have therefore increased [5,6]. The future cost targets for hydrogen refuelling at the filling station are typically estimated by taking the cost of conventional fuels as a benchmark [4]. For Germany, the cost target for hydrogen refuelling was given between 5 € and 7 € per kg for hydrogen produced via electrolysis from renewable wind energy, liquefied and distributed in liquid state [7]. A similar future target for delivered hydrogen costs is given for the European Union [2].

Liquefying hydrogen is a cost-efficient way to store and transport large volumes of hydrogen over long distances [4]. Liquid hydrogen (LH₂) delivered prices in the USA were reported to be between approximately 5 and 10 \$ per kg LH₂ in 2010 [8], depending on location and volumes. Liquid hydrogen is produced by the cooling, purification, ortho-to para-hydrogen conversion, expansion and liquefaction of a hydrogen feed gas from atmospheric temperature to about 20 K [6,9]. The principles of hydrogen liquefaction are described in literature [9–11]. Up to date, the largest hydrogen liquefaction plants were built in the USA from the 1960s to the late 1970s for the space programs and were designed for liquefaction capacities of up to 55 metric tons per day (tpd) LH₂ [12–15]. In the USA, hydrogen liquefiers with a capacity of up to about 35 tpd LH₂ are in operation today [12,14,15]. More recently built hydrogen liquefaction plants are typically smaller, with plant capacities between approximately 5 and 10 tpd LH₂ [14,16,17]. The specific energy consumption SEC for a state-of-the-art 5 tpd LH₂ hydrogen liquefier is about 10 kWh per kg LH₂ [6,16,18].

In existing plants, the specific liquefaction costs SLC in € per kg LH₂, including all the capital (CAPEX) and operating expenses (OPEX) of the liquefaction plant, can take about 50% of the total hydrogen production costs. Large-scale liquid hydrogen production capacities exceeding today's plant sizes may soon be required to reduce the specific costs of the overall hydrogen value chain [6]. New large-scale hydrogen liquefaction concepts require exergy efficient process designs to reduce the SEC and thus the yearly OPEX. Therefore, a high number of conceptual design studies for future large-scale hydrogen liquefaction plants were published in literature since the late 1970s [14]. A literature review is given by the authors in Refs. [6,14,17,19]. The recent 50 tpd LH₂ IDEALHY conceptual process design, with a calculated specific energy consumption SEC of about 6.4 kWh per kg LH₂, is regarded as a benchmark for the SEC of large-scale hydrogen liquefaction plants [20–22].

The electricity costs, however, are a part of the yearly OPEX and only a fraction of the total SLC. The additional plant CAPEX required to improve the exergy efficiency are limited by economic viability. Besides challenges in the upscaling of the plant and equipment design, a substantial reduction of the SLC is required. The main goal in this paper is to minimize the specific liquefaction costs SLC through hydrogen liquefier process optimization. Novel optimization methods are outlined and applied to the newly developed large-scale hydrogen liquefaction designs [23] to reduce the specific energy

consumption SEC and the SLC. The process concepts are optimized for different design modifications as well as for different liquefaction capacities from 25 tpd to 100 tpd LH₂ while taking into account also sensitivities in, for instance, electricity costs. A final liquefier design case is selected for a detailed process and equipment design for implementation within the next 5 years.

State-of-the-art

A simplified block diagram for the hydrogen liquefaction process is depicted in Fig. 1. Built industrial hydrogen liquefaction processes typically include two refrigeration steps for different temperature ranges [9]. In a first step, the hydrogen feed gas is precooled with liquid nitrogen (LN₂) down to an intermediate precooling temperature of about 80 K with one or more aluminium brazed plate-fin heat exchangers [9,24]. Downstream of the heat exchangers, the hydrogen is passed through a cryogenic purification in 80 K adsorber beds to remove impurities, mainly air gases such as nitrogen, from the feed gas [9,25]. The adsorber vessels are typically installed in parallel to operate one vessel as adsorber while the other vessel is regenerated. The required catalytic ortho-to para-hydrogen conversion is a major challenge in hydrogen liquefiers [9,26–28]. The catalytic conversion is typically initiated after the precooling step and downstream of the feed gas purification. In efficient industrial liquefiers, the catalyst for the ortho-to para-hydrogen conversion is filled inside the process heat exchangers for a continuous conversion close to equilibrium [9,28,29].

The precooled hydrogen is cooled further and liquefied with a closed loop cryogenic refrigeration cycle. The cryogenic refrigeration cycle in built liquefaction processes is based either on a helium Brayton cycle or a hydrogen Claude cycle [9,30]. A Brayton cycle uses a turbine instead of a Joule–Thomson throttle valve for expansion [31]. The classical Claude cycle is a combination of a Joule–Thomson cycle and the Brayton cycle: a refrigerant stream is expanded in expanders while a second partial stream is cooled and expanded in a throttle valve to produce a two-phase stream [31]. The specific energy consumption SEC for a conventional 5 tpd LH₂ hydrogen liquefaction process with a hydrogen Claude cycle and LN₂ precooling is about 10 kWh per kg LH₂ [6,18,30].

Since the late 1970s, numerous conceptual design studies for future large-scale hydrogen liquefaction processes with capacities of up to 860 tpd LH₂ were published in literature [14,17,19]. A brief summary of selected relevant publications from literature is given by the author in Refs. [6,23]. In 1978, a hydrogen Claude cycle with nitrogen precooling was analysed in one of the first conceptual studies on large-scale liquefaction for 250 tpd LH₂ [32]. From 1994 to 2002, the WE-NET project in Japan studied different process concepts for 300 tpd LH₂, including a hydrogen Claude cycle, a helium Brayton cycle, a neon Brayton cycle and a helium-neon refrigerant mixture cycle [33–35]. The hydrogen Claude cycle with a closed loop nitrogen precooling cycle was chosen as preferred liquefaction process [33,35]. A hydrogen Claude cycle using liquefied natural gas (LNG) that was assumed to be available at LNG import terminals for precooling was investigated by

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