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# Reducing the exergy destruction in the cryogenic heat exchangers of hydrogen liquefaction processes

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## ABSTRACT

A present key barrier for implementing large-scale hydrogen liquefaction plants is their high power consumption. The cryogenic heat exchangers are responsible for a significant part of the exergy destruction in these plants and we evaluate in this work strategies to increase their efficiency. A detailed model of a plate-fin heat exchanger is presented that incorporates the geometry of the heat exchanger, nonequilibrium ortho-para conversion and correlations to account for the pressure drop and heat transfer coefficients due to possible boiling/condensation of the refrigerant at the lowest temperatures. Based on available experimental data, a correlation for the ortho-para conversion kinetics is developed, which reproduces available experimental data with an average deviation of 2.2%. In a plate-fin heat exchanger that is used to cool the hydrogen from 47.8 K to 29.3 K with hydrogen as refrigerant, we find that the two main sources of exergy destruction are thermal gradients and ortho-para hydrogen conversion, being responsible for 69% and 29% of the exergy destruction respectively. A route to reduce the exergy destruction from the ortho-para hydrogen conversion is to use a more efficient catalyst, where we find that a doubling of the catalytic activity in comparison to ferric-oxide, as demonstrated by nickel oxide-silica catalyst, reduces the exergy destruction by 9%. A possible route to reduce the exergy destruction from thermal gradients is to employ an evaporating mixture of helium and neon at the cold-side of the heat exchanger, which reduces the exergy destruction by 7%. We find that a combination of hydrogen and helium-neon as refrigerants at high and low temperatures respectively, enables a reduction of the exergy destruction by 35%. A combination of both improved catalyst and the use of hydrogen and helium-neon as refrigerants gives the possibility to reduce the exergy destruction in the cryogenic heat exchangers by 43%. The limited efficiency of the ortho-para catalyst represents a barrier for further improvement of the efficiency.

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## Introduction

Hydrogen represents a zero-emission fuel when used, for instance, in fuel cells for mobility or electricity generation. The only components emitted from these fuel cells are heat and water. The most common way to produce hydrogen is by reforming of natural gas (48%), but a large portion is also produced from oil (30%) and by coal gasification (18%) [1]. When steam-methane reforming, or other reforming technologies such as partial oxidation or auto-thermal reforming, is combined with pre-combustion CO<sub>2</sub> capture and storage, the processes can be used to provide a clean energy carrier for transportation or for generating electricity. On a long-term, a gradual conversion to a hydrogen-based society can be a way of mitigating the threat of accelerated global warming from anthropogenic greenhouse gas emissions.

Some of the largest technological challenges in a transition to a hydrogen-society are associated with the transport and storage of hydrogen. Hydrogen can either be liquefied (e.g. at 1.3 bar and 21 K), or compressed (typically in the range 200–700 bar and near ambient temperature). The preferred method of transportation depends on different circumstances such as the quantity of hydrogen, the distance of transportation and the preferred state of distribution and end use. The advantage with liquefied hydrogen (LH<sub>2</sub>) is that the energy density is almost 4.5 times larger than compressed hydrogen at 200 bar [2,3]. This reduces the necessary volume and weight of storage-facilities, which becomes particularly attractive if large quantities of hydrogen are going to be transported from remote locations and for distribution to filling stations in cities.

How beneficial liquefaction of hydrogen will be in comparison to compressed hydrogen depends to a large extent on how energy and cost efficient the hydrogen liquefaction process can be made. One of the current barriers for implementing large-scale plants for liquefaction of hydrogen is their high power consumption. The exergy efficiency of existing liquefaction plants is relatively low (25%–30% when factoring in the penalty for externally supplied liquid nitrogen for pre-cooling) and there is a large potential for improvement [4]. In currently operating hydrogen liquefaction plants, the specific energy requirement to liquefy hydrogen is 11.9 kWh/kg LH<sub>2</sub> for the Leuna plant [5], and slightly lower for newer plants. Several works have in recent years proposed novel solutions for lowering the specific energy requirements of the hydrogen liquefaction process [5–9]. The typical hydrogen liquefaction process consists of four consecutive stages [5,10]:

**Stage 1:** Pre-compression of the hydrogen feed gas, if required.

**Stage 2:** Pre-cooling of the hydrogen gas to about 80 K.

**Stage 3:** Cryogenic cooling of the hydrogen gas down to 20 K–30 K, including ortho-to-para-hydrogen conversion.

**Stage 4:** Final expansion and liquefaction of the hydrogen.

The processes presented in the literature differ most significantly in the pre-cooling and cryogenic cooling stages (Stage 2 and 3). Most of the current technology for hydrogen liquefaction is characterized by the nature of the refrigeration cycle that is used in the cryogenic refrigeration section, i.e.

whether a Claude cycle or a reverse Brayton cycle has been employed [10].

Small- to mid-scale hydrogen liquefaction plants are often placed in close vicinity to cryogenic air separation facilities such that liquid nitrogen can be used as refrigerant in the pre-cooling stage. Due to the limited global requirement for pure oxygen, liquefied nitrogen will not be available as a cheap refrigerant for future large-scale hydrogen liquefaction plants.

The most energy and cost efficient processes suggested in the literature [5,8,9], use mixed refrigerants instead of nitrogen to precool the hydrogen, where the mixed refrigerant consists of hydrocarbons, nitrogen and possibly other components such as neon [5]. A mixed refrigerant can have a tailor-made composition to enable a tight thermal match in the heat exchangers, leading to higher efficiency in the pre-cooling stage (Stage 2). The recent works by Berstad et al. [5], the IDEALHY-consortium [8] and Cardella et al. [9], present processes with very high overall energy efficiencies. In some of these processes, mixtures with neon in combination with either hydrogen or helium are used as refrigerants in the cryogenic cooling stage (Stage 3). One advantage of including neon in the mixture, is that neon increases the molecular weight of the mixture, such that conventional turbo compressors can be used with a maximum of 6–8 stages [9]. Turbo compressors can easily be up-scaled, and have generally higher efficiency and throughput capacity than oil-free piston compressors, which in overall can enable more efficient processes. Another advantage of using neon-mixtures at low temperatures is the enhanced heat transfer coefficient due to evaporating/condensing refrigerant.

There are currently knowledge gaps that need to be closed, both to realize the novel concepts proposed in the literature [5–9], and to develop even more energy efficient hydrogen liquefaction processes. For instance, it is crucial to have a tight thermal match between the hydrogen and the refrigerant, in particular at the lowest temperatures due to the otherwise high exergy destruction. An additional necessity in the hydrogen liquefaction process is to include catalyst to convert ortho-to-para-hydrogen (the protons in the H<sub>2</sub> molecule spin in the same (ortho) or in opposite directions (para)). Otherwise, the heat that is generated when liquefied ortho-hydrogen converts to para-hydrogen in e.g. storage tanks will lead to full evaporation, since the enthalpy difference of ortho-para conversion exceeds the latent heat of evaporation at low temperatures. The ortho to para conversion requires energy-demanding refrigeration, and the exergy destruction increases at decreasing temperature. The required conversion of ortho to para hydrogen is in modern liquefaction plants carried out continuously by filling the plate-fin heat exchangers with catalyst [11].

In this work, we evaluate strategies to reduce the exergy destruction and entropy production in the cryogenic plate-fin heat exchangers in a hydrogen liquefaction facility by taking advantage of a detailed mathematical model that incorporates the most important physical phenomena. To the best of our knowledge, such an analysis has not been presented before in the literature. We present first the model in Sec. **Theory**. Different cases are defined in Sec. **Cases**, where the cases differ in design parameters and choice of refrigerant. Results and discussion are presented in Sec. **Results** with concluding remarks in Sec. **Conclusion**.

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