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# Optimal operation of a large-scale liquid hydrogen plant utilizing mixed fluid refrigeration system

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## ABSTRACT

This work proposes a new modified liquid hydrogen plant that uses a multi-component refrigerant (MR) refrigeration system. A plant capable of producing 100 tons of liquid hydrogen per day is simulated and optimized. Based on a previously published work, this paper represents an improvement for more realistic large-scale plant cycles. Importantly, this work contains a preliminary study of the variables and constraints together with methods for adjustment to achieve optimal steady-state operation. The optimization problem contains 23 variables and 26 constraints. Similar to the MR system, this system can be used to cool the normal feed hydrogen gas from 25 °C to the equilibrium temperature of –193 °C, and a simplified five-component mixture of refrigerant suggested for the plant is described. However, the novelty of this work relative to the previous work lies in the transition of the equilibrium ortho–para hydrogen gas from –193 °C to –253 °C via a new proposed configuration, which recommends a four H<sub>2</sub> Joule–Brayton cycle refrigeration system with optimization. Additionally, the plant optimization was conducted using two additional pinch temperatures (1 °C and 3 °C). The overall power savings is increased with a pinch temperature of 1 °C compared with that of 3 °C. The simulated overall power consumption of the proposed plant is 5.91 kWh/kg<sub>LH<sub>2</sub></sub>, which represents a 50% reduction compared with the current plant in Ingolstadt with an energy consumption of 13.58 kWh/kg<sub>LH<sub>2</sub></sub>. Pressure drops are also employed in the heat exchangers in the study simulation, but the results show that they do not have a significant impact on the overall plant total power consumption.

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## Introduction

Hydrogen has shown promise as an important energy source for future use in transportation vehicles. One of the main challenges in creating a hydrogen economy is the low efficiency of the current hydrogen liquefaction plant cycles.

Currently, the process for liquefaction of hydrogen gas is highly cost intensive and requires a large amount of energy for operation. It is therefore important that the plant is both well designed and also operated at conditions close to optimum.

A literature review for the development of large-scale hydrogen liquefaction processes worldwide from 1898 to 2009 was published by Krasae-in et al. [1]. Following that work,

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other researchers [2,3] used small-scale laboratory mixed fluid refrigeration systems to pre-cool hydrogen gas. Subsequently, Krasae-in et al. [4] proposed a new large-scale multi-component refrigerant (MR) system (generally referred to as a mixed refrigerant cycle [5–8]) with an efficiency greater than 50%. Finally, this paper proposes a more realistic and improved cycle with optimization superior to that previously proposed by the author [4], particularly due to the system used to cool the equilibrium hydrogen gas from  $-193\text{ }^{\circ}\text{C}$  to  $-253\text{ }^{\circ}\text{C}$ . The process considered in this paper is based on an MR system that can be used to cool the normal feed hydrogen gas from  $25\text{ }^{\circ}\text{C}$  to the equilibrium temperature of  $-193\text{ }^{\circ}\text{C}$ . A simplified five-component mixture of refrigerant suggested for the plant is also identified. However, a difference from the work in the previous paper is that the transition of the equilibrium ortho–para hydrogen gas from  $-193\text{ }^{\circ}\text{C}$  to  $-253\text{ }^{\circ}\text{C}$  is carried out via a newly proposed configuration for a four  $\text{H}_2$ -Joule–Brayton (J–B) cycle refrigeration system with optimization.

First, we present a review of the literature relevant to the optimal design of an MR system and describe the concept, justification, and rules for selecting the proposed process. Next, the optimal design of the proposed process is presented, including the optimization strategy. Importantly, different pinch temperatures and pressure drops in all of the heat exchangers are illustrated and are shown to have an effect on the total plant energy efficiency. Finally, methods for optimal control of the plant in practice are briefly explained.

## Process description

The first person to introduce the MR cycle was Podbielniak [5]. Later on, using the Podbielniak patent, Kleemenko [6] showed that the cycle efficiency depends on small temperature differences between the fluid being cooled and the multi-component refrigerants, a phenomenon that is related to the irreversibility of the heat transfer between the two streams. As a result, there are several US patents in existence on this technology for LNG. One of most interesting recommended cycles is an example proposed by Gaumer and Newton [7]. The concept of multi-component refrigerants (also known as mixed refrigerants) (Bottura [8]; Chrz 2010 [9]; Mafi et al. [10]; Bosma and Nagelvoort [11]) has been widely used in the liquefaction of natural gas for decades because of its reduced energy consumption compared with that of other conventional liquefaction cycles. The novelty of this mixed refrigerant system is described quite well by Flynn [12] and Venkatarathnam [13]. Until recently, certain large-scale LNG plants used more complicated MR cycles such as the “Dual” and “Cascade” MR processes previously described by Newton [14], Etzbach et al. [15], and Rentler and Sproul [16] and those recently invented by Kimble [17] and Robert and Agrawal [18] together with Cole and Bowen [19].

For a literature review of selected interesting papers on the optimization of the MR cycle, see references [20–23], which describe the concept adapted for use in this paper. Alabdulkarem et al. [20] and Tak et al. [21] explained the process for optimizing the MR cycles with different pinch temperatures. Nogal et al. [22] presented the concept optimizing the MR composition with no crossovers or minimum temperature

approach violations such that the hot and cold temperature profiles (composite curves) are verified for feasibility. Jensen and Skogestad [23] proposed a method for optimizing the MR cycle.

This paper contains a preliminary study on the optimal operation of a proposed cycle, as shown in the flowsheet of Fig. 1. For additional detail on the previous process, see Krasae-in et al. [4]. The newly modified and improved cycle is detailed in Section 2.2, the new proposed four hydrogen J–B cycles. The optimization concept for a mixed refrigerant cycle in this paper is taken from Jensen and Skogestad [23], who studied the optimum operation of refrigeration and LNG processes. According to Bracha [11], the purity of hydrogen from an actual large plant is 99%, which is quite high. Thus, it is assumed in this work that impurities do not play a significant role in the energy intensity of the plant.

The simulation and experiments on the small-scale plant described in Krasae-in et al. [3] reported that the trends of both the simulation and experimental data point in the same direction, and this paper extends this concept to the proposed large-scale plant with an MR refrigeration system. The large-scale MR cycle is modified from the small-scale MR process based on the test rig described in Krasae-in et al. [3]. The differences can be observed in the changes observed from the previous Fig. 7, Simulation data of the laboratory test rig with the proposed simplified composition compared to the experimental data, in Krasae-in et al. [3] to the current Fig. 1 in this paper, Simulation flow sheet for the proposed large-scale 100-TPD  $\text{LH}_2$  plant with MR and four  $\text{H}_2$  Joule–Brayton cascade cycles. These changes are described as follows. Ortho–para catalysts are included for ortho–para hydrogen gas conversion, single-stage compression is switched to two-stage to reduce the power consumption, and expansion valves are replaced by expanders to reduce the energy losses. In addition, a simple helium system or heat exchanger (HX5 in Fig. 7 of Krasae-in et al. [3]) is replaced by the four hydrogen J–B cycles. The simulation of the proposed large-scale plant is realized using a self-written C language program due to the lack of affordable commercial software, but PRO/II is used with the simulation of the test rig found in Krasae-in et al. [3]. The mathematical models (including both conservation of mass and energy) of all components: e.g., compressors, expanders, heat exchangers, O–P converters, liquid separators, mixers, and others are from the manual found in PRO/II that the author previously knew; this is including the mixed-refrigerant model. However, other commercially available process simulation programs can be used for the simulations, including HYSYS, HYSIM, and ASPEN PLUS, which are all similar. The new and optimized MR process has been particularly modified for a large-scale process with heat conversion by catalysts and has a simplified composition. The concept of energy analysis from Krasae-in et al. [2] is also adopted in this work for the large-scale analysis.

The concept, justification, and rules for cycle selection, as shown in Fig. 1, and reasons why the proposed system is superior to conventional choices are explained and clarified in this section. The reason for cooling normal hydrogen gas from  $25\text{ }^{\circ}\text{C}$  to an equilibrium temperature of  $-193\text{ }^{\circ}\text{C}$  using the MR cycle is given in the experiments by Krasae-in et al. [3]. The system is more efficient, simple, and reliable compared with pure refrigerant systems because the mixture of refrigerants

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