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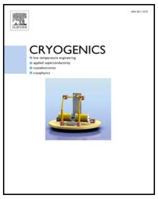

Research paper

Experimental study on CO_2 frosting and clogging in a brazed plate heat exchanger for natural gas liquefaction process

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ABSTRACT

The plate-fin heat exchanger (PFHE), which has been widely used in natural gas liquefaction (LNG) industry at present, has some disadvantages such as being sensitive to the impurities in the feed gas, such as water, CO_2 and H_2S. Compared with the PFHE, the brazed plate heat exchanger (BPHE), which has been applied in some boil off gas (BOG) recycling LNG plants of small to middle size, has simpler inherent structure and higher impurity tolerance. In this study the BPHE is suggested to replace the PFHE to simplify or even omit the massive CO_2 purification equipment for the LNG process. A set of experimental apparatus is designed and constructed to investigate the influence of the CO_2 concentration of the natural gas on solid precipitation inside a typical BPHE meanly by considering the flow resistance throughout the LNG process. The results show that the maximum allowable CO_2 concentration of the natural gas liquefied in the BPHE is two orders of magnitude higher than that in the PFHE under the same condition. In addition, the solid-liquid separation for the CO_2 impurity is studied and the reasonable separating temperature is obtained. The solid CO2 should be separated below 135 K under the pressure of 3 MPa.

1. Introduction

Natural gas has been regarded as an important alternative of oil and coal. It is widely used in modern industry since it produces less pollution as a fuel and the reserve is relatively rich as well. Liquefied natural gas (LNG) is one of the most important methods for the storage and transportation of natural gas. Over the past three decades, LNG industry has successfully brought many large remote gas fields to the gas markets that are unreachable by long pipeline [1]. Especially in countries with large area, more and more attentions have been shifted to scattered small natural gas reservoirs that were previously considered to be too remote and over cost for the exploitation and development [2], which promotes the research of the optimized LNG process and the development of the small-scale LNG plants. Distributed-scale LNG plant, which refers to the plant with LNG production capacities of 15–100 m^3 per day, is small enough to be packaged onto skids and manufactured in facilities [2]. It is much more beneficial than large-scale LNG plants when applied at small and remote natural gas reservoirs.

In general, there are two ways to reduce the size of the LNG plants. One is to optimize the liquefaction process and increase the energy efficiency. Worldwide researchers have been working on it for a long time and achieved significant progress. For instance, He et al. [3–5] recently proposed several novel designs for small-scale LNG plants in skid-mount packages, based on nitrogen expansion and mixed refrigerant cycles. The process optimization included comprehensive thermodynamic analyses and dynamic numerical simulation. Yuan et al. [6] put up with a novel liquefaction process adopting single nitrogen expansion with carbon dioxide pre-cooling and achieved the optimized liquefaction rate of 0.77 with unit energy consumption of 9.90 kW·h/kmol. The other option is to simplify or even avoid the complicated equipment for smaller size. Most small-scale LNG plants are equipped with massive impurity removal equipment to satisfy the strict purification standard definition, < 50 ppm for carbon dioxide (CO_2) and < 10 ng/Nm^3 for mercury [7], to prevent CO_2 freezing and mercury corrosion inside traditional plate-fin heat exchanger (PFHE) made of aluminum materials, which cause pretreatment system to large dimension and high cost. A compact heat exchanger with high heat transfer capacity is critical for small-scale LNG plants, especially those in skid-mount packages since natural gas can be liquefied when its temperature is cooled below 112 K at atmospheric pressure. [8] Plate-fin heat exchangers have been widely applied in LNG system for the large heat transfer area per unit of volume. The application of the PFHE in LNG system has been well studied in terms of internal structure, flow pattern distribution, heat transfer performance, material strength and stability over past decades [9–11]. Cao et al. [12] conducted an

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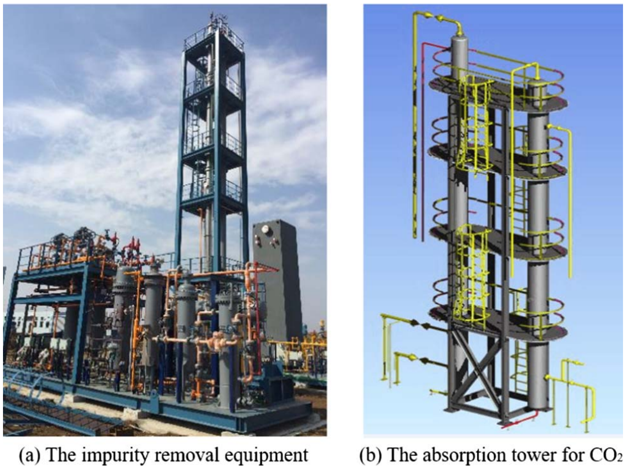
experiment with mixed refrigerant to study the heat transfer performance of the PFHE throughout the cool-down process of LNG and proposed some suggestions regarding its design in a mixed refrigerant cryogenic system. Ma et al. [13–15] recently investigated the stress characteristics of the plate-fin structure in an LNG heat exchanger by means of simulation and found how the peak value of equivalent stress in plate-fin structures changed in the cool-down process. G. Skaugen et al. [16] studied the non-ideal behaviors of the PFHE including Ledinegg instability and thermal oscillation during the condensation and evaporation of LNG. One detailed model of the PFHE was developed to calculate each layer individually and to investigate the effects of operation under static instability conditions. However, the literature reviews showed that little study was focused on the clogging phenomenon in this kind of heat exchangers. The inner structure of the PFHE is quite complex, which increases the sensitivity to the impurity within the natural gas. For instance, carbon dioxide (CO_2) is one of the major impurities, the freezing temperature of which is much higher than the liquefaction temperature of methane. It is obvious that the PFHE will be clogged by the frozen CO_2 when the natural gas is gradually cooling-down through the channels. As a result, large impurity removal equipment has been installed in the LNG process equipped with the PFHE, which cause pretreatment system to large dimension and high cost. Researchers have been investigating the CO_2 blockage under cryogenic conditions. Mesude Ozturk et al. [17] applied the Perturbed Chain-SAFT equation to model the phase behavior of CO_2 in methane and other hydrocarbons. The model can accurately predict the vapor-liquid equilibria, solid-vapor equilibria, and solid-liquid equilibria of hydrocarbon carbon dioxide systems and determine the CO_2 solubility in different hydrocarbon mixtures. It is helpful for optimizing the industrial process to reduce the carbon dioxide solidification condition. But the effect is limited since the CO_2 pretreatment equipment is still required. In fact, the maximum allowable CO_2 concentration of the feed gas for the heat exchanger can be influenced by many factors, such as the flow rate, the pressure, the type of the heat exchanger and so on. In the present paper, we focus on the type of the heat exchanger. A novel cold box equipped with brazed plate heat exchanger (BPHE) instead of the PFHE is proposed and designed to liquefy the natural gas. As seen from Fig. 2, it is apparent that the structure of the BPHE is much simpler than that of the plate-fin heat exchanger. The BPHE is less likely to be clogged when there is a small quantity of solid CO_2 impurity due to the simple internal structure. Therefore, the pretreating equipment of the LNG process can be simplified by applying the BPHE, which may largely reduce the impurity removal cost. Additionally, the BPHE has been applied in some small-scale LNG plants. [18] The cooling capacity of the BPHE can also meet the requirement of the LNG process. In the present paper, an experimental set-up was designed and constructed for the cool-down process of the methane/carbon dioxide (CH_4/CO_2) gas mixtures from room temperature to liquid methane temperature in a typical BPHE. The experiments were operated under the pressure of 3 MPa. The heat transfer area of the BPHE was appraised. The influences of the CO_2 concentration of the gas mixture on solid precipitation inside the BPHE was investigated by taking account of the flow resistance throughout the liquefaction process. The maximum allowable CO_2 concentration of the feed gas when using a BPHE to liquefy the natural gas was obtained, in which case the flow channels of the BPHE would not be clogged by the frozen CO_2 impurity. Additionally, further experiment was conducted to investigate the separation of the frozen CO_2 and the separating temperature was given.



(a) The impurity removal equipment (b) The absorption tower for CO_2

Fig. 1. The impurity removal equipment for a 50,000 m^3/d small-scale LNG plant.

2. Experimental setup

2.1. Experimental apparatus and procedures

The schematic and photograph of the experimental apparatus were shown in Fig. 3a and Fig. 3b.

The BPHE, which was made of stainless steel and manufactured by SWEP Company was installed inside a vacuum chamber for adiabatic insulation. The information of the BPHE used in the experiment is listed in Table 1.

CH_4/CO_2 gas mixture with different CO_2 concentration was used to simulate real natural gas. The gas mixture flowed into the channels of the heat exchanger where it was cooled by the liquid nitrogen. The methane was liquefied, and the CO_2 was frozen during the cool-down process. Then the solid CO_2 was separated in a solid-liquid separator and the liquid methane was heated by the water bath at room temperature. Gas samples were picked up at the outlet and the content of the samples was tested by the gas chromatograph. The performance of the solid-liquid separator could be evaluated by comparing the CO_2 concentration of the gas mixture before and after the liquefaction process. PT-100 is used as the temperature sensor and installed at the inlet and outlet of the BPHE and the separator. All the temperature data measured in experiments are the temperature of the fluid. Differential pressure transducers were installed at the inlet and the outlet of the heat exchanger in order to obtain the data of the pressure drop which could quantitatively indicate the flow resistance of the gas mixture. Therefore, whether the heat exchanger was clogged by the solid CO_2 or not could be confirmed.

2.2. Experimental conditions

In order to obtain the maximum allowable CO_2 concentration of the CH_4/CO_2 gas mixture, the feed gas with different CO_2 concentrations was used in this experimental test, as shown in Table 2. The pressure of the CH_4/CO_2 gas mixtures were under 3 MPa according to the data of the small-scale LNG plant with the liquefaction capacity of 50,000 Nm^3 per day which is shown in Fig. 1. Additionally, the BPHE was much smaller than the real one since it was limited by the scale of the laboratory. The flow rate of the fluid should be reduced at a specific proportion so that the velocity of the feed gas would be the same as the actual LNG process. The flow velocity in the PFHE equipped in the small-scale LNG plant shown in Fig. 1 is about 0.3 m/s. Thus, the flow rate could be obtained by the following equation.

$$Q_v = A \cdot u$$

where Q_v is the flow rate; A is the section area of the hot side of the BPHE used in the experiment and u is the flow velocity.

According to the result of the calculation, the flow rate of the feed gas in this experiment was designed as 35 standard liter per minute (SLPM), which was 1/1000 times of the flow rate in the LNG plant.

Although the liquefaction temperature of methane under the pressure of 3 MPa is 177 K, in actual LNG process the natural gas usually needs to be cooled to 120 K–140 K in case of evaporation during the