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Hydrate-based carbon dioxide capture from simulated integrated gasification combined cycle gas

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Abstract

The equilibrium hydrate formation conditions for CO_2/H_2 gas mixtures with different CO_2 concentrations in 0.29 mol% TBAB aqueous solution are firstly measured. The results illustrate that the equilibrium hydrate formation pressure increases remarkably with the decrease of CO_2 concentration in the gas mixture. Based on the phase equilibrium data, a three stages hydrate CO_2 separation from integrated gasification combined cycle (IGCC) synthesis gas is investigated. Because the separation efficiency is quite low for the third hydrate separation, a hybrid CO_2 separation process of two hydrate stages in conjunction with one chemical absorption process (absorption with MEA) is proposed and studied. The experimental results show H_2 concentration in the final residual gas released from the three stages hydrate CO_2 separation process was approximately 95.0 mol% while that released from the hybrid CO_2 separation process was approximately 99.4 mol%. Thus, the hybrid process is possible to be a promising technology for the industrial application in the future.

Key words

integrated gasification combined cycle (IGCC); gas hydrate; CO_2 separation; H_2 purification; chemical absorption

1. Introduction

Carbon dioxide (CO_2) is considered as one main contributor to the greenhouse gases resulting in the global warming [1]. Near one third of all CO_2 emissions worldwide is produced by the fuel power plants [2]. Therefore, the prime target is to capture CO_2 from the fuel power plants to slow down the deterioration of the climate. Pre-combustion capture and post-combustion capture are two approaches extensively used for CO_2 capture from the fuel power plants [3]. Pre-combustion capture is capturing or removing CO_2 before the fuel being burnt while post-combustion capture is capturing or removing CO_2 from the flue gas [4]. Presently, the integrated gasification combined cycle (IGCC) is widely utilized in the coal-fired power plants. CO_2 capture from IGCC becomes one promising route of pre-combustion capture. In IGCC plant, coal is firstly gasified and then converted into a stream of hydrogen (H_2) and CO_2 [5]. Once CO_2 is removed efficiently, the resultant H_2 can be used as a clean fuel. Physical adsorption, chemical absorption, cryogenic fractionation and membrane process are the conventional technologies for capturing CO_2 from the fuel gas. However, the conventional technologies face some challenges, such as low capacity, technology feasibility, high cost and corrosion [6,7]. Therefore, novel and low cost technologies of carbon capture need to be developed.

Gas hydrate-based CO_2 capture technology is one of the novel technologies for capturing CO_2 from IGCC synthesis gas (mixture of CO_2/H_2). The gas hydrates are non-stoichiometric compounds formed by water molecules and small molecule gases such as CO_2, nitrogen (N_2), oxygen (O_2), H_2, methane (CH_4) [8]. The mechanism of hydrate-based CO_2 capture is selective partition of CO_2 component between hydrate phase and gaseous phase [9,10]. Because the equilibrium hydrate formation pressure of CO_2 is much lower than that of H_2 at the same temperature, it is expected that

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CO_2 is preferentially encaged into the hydrate phase. The hydrates are then separated and subsequently decomposed to create CO_2-rich stream while the rest constitutes CO_2-lean one. Thereby, CO_2 is captured from IGCC synthesis gas.

IGCC synthesis gas mainly contains approximately 40.0/60.0 mol% CO_2/H_2 gas mixture, and the outlet pressure of the synthesis gas is generally 3−5 MPa [3]. In our previous work [10], the concentration of CO_2 can be reduced from 40.0 mol% in the feed gas to approximately 13.5 mol% in the residual gas after one stage hydrate-based CO_2 separation. Therefore, in order to separate CO_2 from CO_2/H_2 gas mixture completely, two hydrate stages or even three hydrate stages are required. However, the equilibrium hydrate formation pressure is higher with the decrease of CO_2 concentration in the gas mixture. Thus, higher driving forces are required for the gas mixtures containing lower concentration of CO_2 to form gas hydrate, that is, more serious conditions are required for the gas mixtures containing lower concentration of CO_2 to form gas hydrate. Therefore, a hybrid process based on the hydrate and chemical absorption process is developed. Moreover, no complete system configurations with key hybrid process parameters were presented in the papers [11,12].

In our previous work, 0.29 mol% TBAB solution was proved to be an optimal hydrate formation promoter for the hydrate-based CO_2 capture from either flue gases or IGCC synthesis gas [9]. Thus, 0.29 mol% TBAB solution is adopted in this work directly. In addition, the equilibrium hydrate formation conditions for $CO_2/H_2/TBAB/H_2O$ systems with different CO_2 concentrations are measured firstly, and a hybrid process based on two hydrate stages coupled with a chemical absorption process is proposed for CO_2 capture. By the comparison of CO_2 capture efficiencies, the concentrations of H_2 in the residual gas, the energy consumptions between the third-stage hydrate-based CO_2 separation process and the hybrid process, and an optimal separation process are determined.

2. Experimental

2.1. Materials

A CO_2/H_2 gas mixture containing 39.6 mol% CO_2 was used in the work to simulate a pretreated fuel gas mixture. The gas mixture was supplied by Foshan Huate Gas Co., Ltd. Tetra-*n*-butyl ammonium bromide (TBAB) with 99.9% purity was supplied by Shanghai Sinopharm Chemical Reagent Co., Ltd., China. Monoethanolamine (MEA) with the purity of more than 99.0% was offered by Tianjin Fuyu Fine Chemicals Co., Ltd. The deionized water used with the resistivity of 18.25 m /cm was produced by an ultra-pure water system supplied by Nanjing Ultrapure Water Technology Co., Ltd., China. The glass-balls with the diameter of 3 mm were supplied by Yunhui Business Co., Ltd., China.

2.2. Apparatus

The experimental apparatus in this work is shown in Figures 1 and 2. The apparatus in Figure 1 was the same as that in our previous work [9]. The crystallizer (CR) with inner volume of 336 mL and the supply vessel (SV) with the inner volume of 1350 mL were made of 316 stainless steels. They were immersed in a glycol-water bath. On the front and back of the CR, there are two circular viewing windows made of Plexiglas. The maximum working pressure of the CR was 25 MPa. A magnetic stirrer (500 r/min) was employed to mix the contents in the CR. The temperatures of the gas phase and the liquid phase in the CR were measured using two Pt1000 thermocouples (JM6081) with uncertainties of ±0.1 K. The pressures in the SV and CR were measured with two Setra smart pressure transducers (model 552, Boxborough, MA, USA) with the uncertainty of ±20 kPa. In addition, the composition of the gas phase in the CR was determined with a HP6890 gas

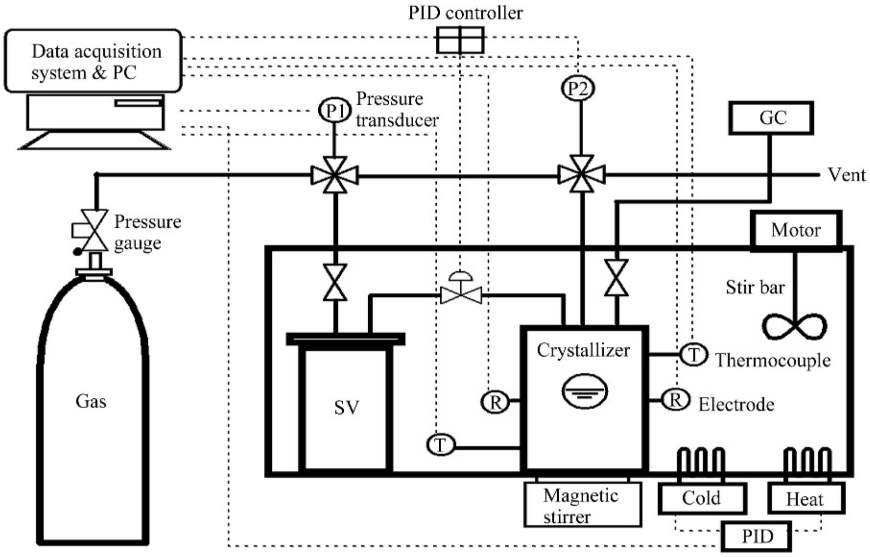

Figure 1. Experimental apparatus for hydrate process