



Review

A review of the hydrate based gas separation (HBGS) process for carbon dioxide pre-combustion capture



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ABSTRACT

In this work, a systematic review of the literature work done so far on the use of hydrate crystallization as a basis to develop data for the hydrate based gas separation (HBGS) process for the capture of CO₂ from fuel gas mixtures is presented. Such a gas mixture may arise in integrated gasification combined cycle (IGCC) power plants. A thorough assessment of the thermodynamic, kinetic factors and economic aspects of the HBGS process and critical comments are presented. Compared with competing technologies, high CO₂ capacity and the use of water as a solvent are key advantages for the HBGS process for CO₂ capture. Furthermore, in this review, a snapshot of the current state-of-the-art is presented and further research and development opportunities and pathways for commercializing the HBGS process for pre-combustion capture of CO₂ from IGCC power plants are discussed.

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1. Introduction

The escalating level of carbon dioxide in the atmosphere and that dissolved in ocean waters are major environmental concerns as they are linked to climate change and ocean acidification respectively. Power generation is one of the major sources of CO₂ emission, which contributes about 41% [1]. Increasing energy demands in India and China have resulted in the build-up of more than one power plant every week in these countries. Due to the strong global demand for energy, we are highly dependent on conventional fossil fuels like oil, natural gas and coal. Since fossil fuels are expected to continue as the primary source of energy, there is a need for CO₂ capture and sequestration to mitigate the environmental impact. One approach to reduce CO₂ emissions from the use of fossil fuels for power generation is to capture CO₂ prior to combustion. This approach is known as “pre-combustion” capture.

Pre-combustion capture is usually applied in integrated gasification combined cycle (IGCC) power plants. IGCC power plants use a gasifier to convert fossil fuels into syngas-mixture of CO and H₂ through partial oxidation. The syngas thus produced undergoes a

water gas shift (WGS) reaction to produce CO₂ and additional H₂. CO₂ can be captured and H₂ produced can be employed in gas turbines or fuel cells to produce electricity. This process can be easily adapted for natural gas and biomass. The mixture of CO₂/H₂ coming out of a WGS column is referred to as fuel gas mixture. On the other hand, the products of combustion in conventional thermal power plants are known as flue gas mixture, which is a mixture of CO₂, N₂ and O₂ after removal of particulates and SO_x. The pressure of this stream which is rich in CO₂ compared to flue gas mixture from a conventional power station ranges from 2.0 to 7.0 MPa [2]. High CO₂ partial pressure of fuel gas mixture makes the capture process more efficient compared to CO₂ capture from a flue gas mixture from a conventional power plant. Absorption, adsorption, membrane separation and cryogenic separation can be employed for CO₂ separation from the fuel gas mixture. These methods have their own advantages and disadvantages [3–8]. Absorption based on physical solvents like Selexol or Rectisol have been used in the industry for natural gas sweetening (removal of acid gases, CO₂ and H₂S) but there are several limitations in expanding them to CO₂ capture from power plants such as solvent regeneration efficiency, solvent degradation and vessel corrosion. The above factors affect the overall carbon removal efficiency from these solvents [6]. Other potential approaches like membrane separation, ionic liquids based absorption and adsorption are still at

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the laboratory scale testing for pre-combustion capture. Hence, there is a continued interest to develop novel, less energy intensive and environmental friendly processes to capture CO₂ from fuel gas streams.

Hydrate based gas separation (HBGS) process has been considered as a promising new/novel approach for CO₂ separation from fuel and flue gas mixtures [3–7,9–16]. The basis for separation is the selective partition of the CO₂ component of a fuel or flue gas mixture between the solid hydrate crystal phase and the gaseous phase upon hydrate crystal formation. Gas hydrates are non-stoichiometric crystalline compounds formed when guest molecules of suitable size and shape are incorporated in the well-defined cages in the host lattice made up of hydrogen bonded water molecules [17–19]. Although gas hydrate research has been of scientific interest since 1800s, three major focus areas are currently driving the research on gas hydrates, namely, flow assurance, natural gas hydrates in nature pertaining to energy recovery, and innovative technological applications [15,16,20–27]. A historical perspective of “gas hydrates” or “clathrate hydrates” research is presented in Fig. 1. As can be seen in the figure, there has been a phenomenal rise (data fit to exponential growth model for illustration) in research activity in the last decade or so. The top five subject areas where the publications belong to are: Earth and Planetary Sciences, Chemistry, Engineering, Chemical Engineering and Energy. Selective fractionation of gas components and high storage capacity in gas hydrates have motivated an extensive effort to employ hydrate crystallization for a wide range of innovative applications including gas separation, gas storage, seawater desalination, natural gas storage and transport, CO₂ sequestration, cold storage and refrigeration [28–53].

Extensive studies have also been reported in the literature for CO₂ capture from flue gas mixture for the HBGS process [9,11,12,54–69]. The limitation of employing HBGS process for CO₂ separation from a flue gas mixture is the low CO₂ concentration (10–20 mole %), the compression costs associated with the flue gas for applying the HBGS technology and additional requirement of large equipment for processing.

On the other hand, there are several advantages for applying the HBGS process for pre-combustion capture. The fuel gas stream exiting the water gas shift converter is in the range of 2–7 MPa [2] which is very favourable for hydrate formation. The other advantages of employing the HBGS process for CO₂ capture are the high storage capacity, energy efficiency, clean process (it does not use

toxic or hazardous materials) and environmentally benign (use of water as a solvent) process. Another advantage of the HBGS process is that it can simultaneously capture H₂S and SO₂, thus eliminating the need to have pre-treatment steps for pre-combustion capture [70–73]. Finally, it is noted that the gas from the dissociation of the hydrate crystals will be at elevated pressures thereby further reducing the compression cost for the transport of CO₂ for storage and sequestration. A schematic of the HBGS process for CO₂ pre-combustion capture is shown in Fig. 2.

Only a few review papers are available on this process [15,24,29]. A review of CO₂ capture from shifted synthesis gas, flue gas and sour natural gas or biogas using HBGS process is available in the literature [29]. The application of gas hydrates and a brief survey of experimental works on gas hydrate applications primarily summarizing the thermodynamic data of different gas/gas mixtures is available in the literature [24]. A recent review summarizes the hydrate phase equilibrium, promoters, experimental apparatus and methods employed in the literature.

In this work a systematic analysis of important parameters like gas uptake capacity, the rate of hydrate formation, separation efficiency (CO₂ recovery and separation factor) of the HBGS process in the presence of different promoters and different reactor configurations is presented. Such a review is needed to critically evaluate the advancement made so far and to identify the challenges to overcome in order to commercialize the HBGS process. A comprehensive assessment based on thermodynamic, kinetic parameters and cost estimation of the HBGS process is provided. Finally, future prospects and directions to commercialize the HBGS process are presented.

2. Thermodynamic feasibility of the HBGS process

CO₂ forms structure I (sI) hydrate and CO₂ molecules primarily occupy the large cages and a fraction of small cages of the resultant sI hydrate. On the other hand, Hydrogen forms structure II (sII) hydrate at pressures higher than 300 MPa at temperatures above freezing point of water. A cluster of two H₂ molecules can occupy the small cages while the large cages may have clusters of four H₂ molecules at certain experimental conditions [74]. Extensive phase equilibrium studies have been reported for pure CO₂ and H₂ [75–91]. Fig. 3 shows the hydrate phase equilibrium of pure CO₂ and pure H₂. At 275.6 K, the minimum pressure required to form pure CO₂ and pure H₂ hydrate were 1.6 and 366 MPa respectively. This significant difference in equilibrium hydrate formation condition forms the basis for HBGS process. The hydrate phase equilibrium of a typical fuel gas mixture consisting of 39.2 mol% CO₂ is also shown in Fig. 3. Typical fuel gas mixture exiting an IGCC power plant is in the range of 2.0–7.0 MPa. As can be seen in the figure, the fuel gas mixture can form hydrates from water in this pressure range. The minimum pressure required to form hydrate from fuel gas mixture at 275.6 K is 6.84 MPa. There are several papers in the literature that have reported phase equilibrium data for CO₂/H₂ gas mixture relevant to pre-combustion capture of carbon dioxide [92–95]. Spectroscopic measurements on solid hydrate phase has also been studied and in one such Raman spectroscopic study it has been reported H₂ does not occupy any of the cages of the sI hydrate structure formed from CO₂ + H₂ gas mixture [92]. The observation was based on the fact that the Raman peak of the H–H stretching vibration mode of H₂ was detected in the gas phase but not in the hydrate phase. Thus, it was concluded that H₂ behaves like a diluent gas in the CO₂ hydrate formation from the CO₂ + H₂ gas mixture. Another study based on Raman spectroscopy analysis also reported that H₂ does not occupy the hydrate cages in the formed hydrate [96]. On the other hand, it was shown by X-ray diffraction that H₂ may occupy the small cages of a hydrate formed by CO₂ and H₂ [97].

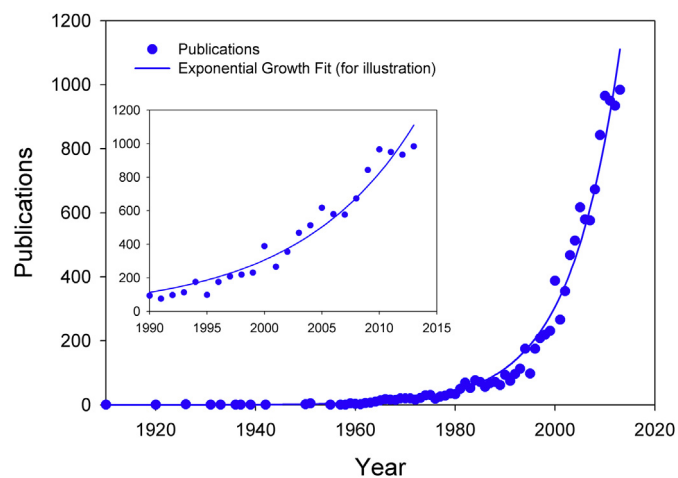


Fig. 1. Historical perspective of gas hydrate research. Data from Scopus on keyword search of “gas hydrates” or “clathrate hydrates”, Search performed on 08-Dec-2014, data presented until 2013.

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