



Review article

Recent advances in gas hydrate-based CO₂ captureHossein Dashti¹, Leonel Zhehao Yew¹, Xia Lou^{*}

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ABSTRACT

Hydrate-based CO₂ capture (HBCC) has received increasing attention, due to such advantages as the mild operating pressure and temperature, the ease of regeneration and its unique separation mechanism. This review paper is focused on the chemical additives and the mechanical methods that have been investigated to improve the CO₂ separation efficiency and energy consumption through HBCC technology. Detailed comparisons of the effects of various chemical additives and mechanical methods on gas consumption, operating conditions, hydrate induction time and CO₂ recovery are critically reviewed. The limitations and challenges of HBCC, in comparison with the conventional methods for CO₂ capture also are discussed.

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1. Introduction

Carbon dioxide (CO₂) capture is a continuous process which requires a significant amount of energy to operate. It contributes to around 70–90% of the total operating cost of the three-stage carbon capture and storage system that is commonly used for the reduction of CO₂ emissions (Herzog and Golomb, 2004). Current CO₂ capture employs adsorption, absorption and membrane technologies which are low in efficiency and usually require multiple stages. Continuous efforts have been made to search for alternative methods in the area of CO₂ capture so that the overall operating cost of the carbon capture can be reduced.

CO₂ is captured from the effluent of power plants either post combustion or pre-combustion. Post-combustion capture refers to the treatment of flue gas before being released into the atmosphere. The flue gas consists of approximately 15–20% CO₂ and 5% O₂, with the balance being N₂, and it is emitted from a full combustion process. Pre-combustion capture refers to the capture of CO₂ from the fuel gas, which is the partially combusted fuel containing approximately 40% CO₂ and 60% H₂. The high CO₂ content in the fuel gas allows more efficient capture. However, it can only be employed in an integrated gasification combined cycle (IGCC) power plant where the fuel is pre-treated to produce CO₂/H₂ syngas

and the CO₂ is separated from the syngas while the H₂ is fed into the combustion process. Post-combustion CO₂ capture is less effective than the pre-combustion method. However, it can be retrofitted to any plant without much modification (Spigarelli and Kawatra, 2013). Regardless, the gas systems that are discussed in this paper are mostly CO₂, or CO₂/N₂, or CO₂/H₂.

Hydrate-based CO₂ capture (HBCC) technology is a novel process that has received enormous attention, both from the industry and academic researchers, during the last two decades. The technology operates at mild pressures and temperatures, through a unique separation mechanism that is easy to regenerate and capable of separating gas mixtures, which might not be achievable via conventional technologies (Englezos and Lee, 2005). A significant number of studies have reported on the potential application of gas hydrates technology in CO₂ capture. These include some early work that was mostly focused on phase equilibrium studies of pure CO₂ hydrates, and more recent work that has focused closely on investigations of various chemical additives and mechanical methods for enhancement of the efficiency of CO₂ capture and separation. This paper will review the recent developments and research activities conducted on HBCC with a focus on chemical additives and mechanical approaches that are able to improve the selectivity, efficiency and kinetics of this technology. An introduction to the principles and significance of gas hydrates technology will be followed by detailed discussions of the current progress in technological improvements through the application of various chemical additives and mechanical methods. The key effects of the chemicals and mechanical methods, as well as the major outcomes

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of the research activities, will be summarised. In addition, the paper will give an account of the costs, limitations and challenges that are associated with HBCC, in comparison with the conventional technologies.

2. Hydrate-based CO₂ capture technology

2.1. Gas hydrates

Gas hydrates are solid clathrates made up of gas molecules (guests), such as methane (CH₄), CO₂, N₂ and H₂, that are caged within a cavity of hydrogen-bonded water molecules (host). They form under the favourable thermodynamic conditions of low temperature and high pressure, and they exhibit various structures depending on the size and chemical properties of the guest molecules (Sloan and Koh, 2008). Most small gas molecules, such as CO₂ and CH₄, form structure I (S_I). Structure II (S_{II}) hydrates form with larger gas molecules such as N₂ (Davidson et al., 1986) and propane. With the mixture of both small and large gas molecules, such as methane + cycloheptane, the structure H (S_H) may form (Sloan, 2003). The crystal structures of these hydrates consist of different water cavities. The most common forms of water cavity include the irregular dodecahedron (4³5⁶6³) and the pentagonal dodecahedron (5¹²), as well as the tetrakaidecahedron (5¹²6²), the hexakaidecahedron (5¹²6⁴) and the icosahedron (5¹²6⁸) that are often collectively described as 5¹²6^m, with *m* = 2, 4, 8 (Sloan and Koh, 2008). The term “5¹²” is used to indicate that a relatively smaller cavity contains 12 pentagonal faces, whereas the term “5¹²6^m” denotes a larger cavity with 12 pentagonal and *m* hexagonal faces, while “4³5⁶6³” illustrates a medium cavity which contains 3 tetragonal, 6 pentagonal and 3 hexagonal faces. A combination of 5¹² and 5¹²6² is more commonly seen in S_I, while the combination of 5¹² and 5¹²6⁴ is more commonly seen in S_{II}. In S_H, a combination of 5¹², 4³5⁶6³ and 5¹²6⁸ has been observed.

Formation of gas (in particular methane) hydrates has been a significant problem for the upstream oil and gas industry because they clog pipelines, valves, wellheads and processing facilities, thus reducing production and causing safety problems. Extensive research activities have been undertaken in order to prevent or mitigate the formation of gas hydrates (Kelland, 2006). Research on the enhancement of gas hydrate formation began in the late 19th

century after discovering the positive applications of gas hydrates for gas storage, separation, sequestration and desalination (Sloan and Koh, 2008).

2.2. Gas hydrate-based CO₂ capture

CO₂, as a small nonpolar hydrocarbon, forms S_I hydrates with a formula of CO₂·*n*H₂O (*n* = 5.75) when coming into contact with water molecules below the equilibrium temperature and above the equilibrium pressure (Sloan and Koh, 2008). Upon dissociation, one volume of CO₂ hydrates can release 175 volumes of CO₂ gas at standard temperature and pressure conditions, which is potentially useful for the separation of the CO₂ from flue gas. The equilibrium phase diagram of CO₂ hydrates is presented in Fig. 1, which is constructed using an equation from experimental data reported by Kamath (1984). The figure also shows that other gases, such as N₂, H₂, and O₂, form hydrates under different equilibrium conditions. The equilibrium curve of H₂ was obtained from Dyadin et al. (1999).

As one can see from the equilibrium diagrams, CO₂ has the lowest hydrate-forming pressure in comparison with other components in flue gas. Separating CO₂ from the other gases can be achieved by first forming a solid hydrate phase that is enriched with CO₂. Dissociating the hydrates, after separating the hydrate phase from the gaseous phase, leads to the recovery of CO₂ that is much higher in concentration than the initial feed. Studies have shown that the concentration of CO₂ in the hydrate phase is at least four times greater than that in the gas phase (Duc et al., 2007). This hydrate-based CO₂ capture process is illustrated in a flow diagram displayed in Fig. 2. In brief, the gas mixture is sent to the hydrate formation reactor, in which CO₂ hydrates form as the pressure increases and temperature decreases. The hydrate slurry is separated from the CO₂-lean gas in the separator and sent to a hydrates dissociation reactor, from which purified CO₂ is collected, and the CO₂-rich gas is recycled for further processing.

2.3. Parameters describing the HBCC process

The efficiency of hydrate-based CO₂ separation is often described by such parameters as hydrate induction time, gas consumption, hydrate equilibrium pressure, CO₂ recovery or split fraction (S.Fr.) and separation factor (S.F.).

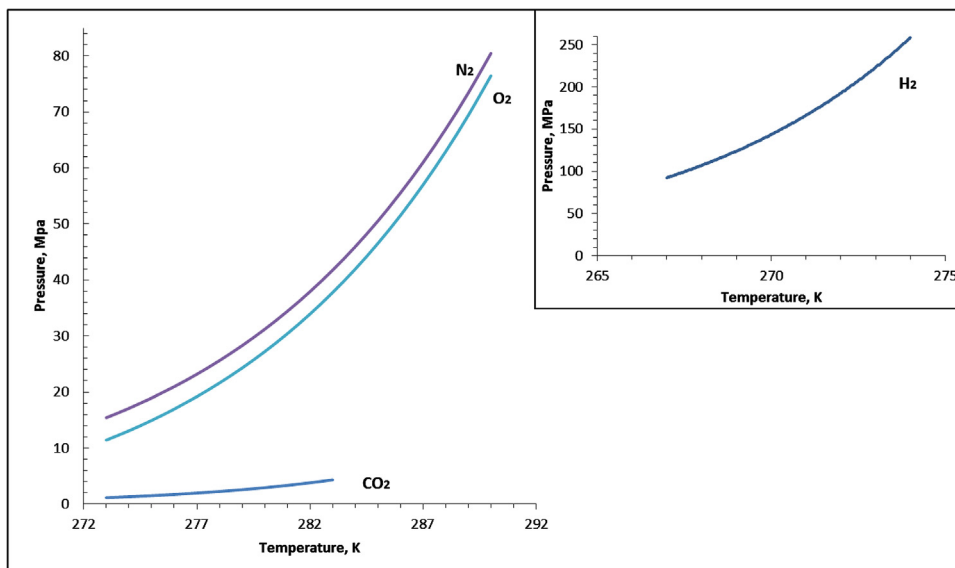


Fig. 1. The equilibrium phase diagrams of different hydrate formers.

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