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Hydrate-based Capture of Acidic Gases for Clean Fuels with New Synergic Additives

Zhi-Ming Xia^{a,b,c}, Xiao-Sen Li^{a,b,c,*}, Zhao-Yang Chen^{a,b,c}, Ke-Feng Yan^{a,b,c}, Chun-Gang Xu^{a,b,c}, Qiu-Nan Lv^{a,b,c}, and Jing Cai^{a,b,c}

^aKey Laboratory of Gas Hydrate, Guangzhou Institute of Energy Conversion, Chinese Academy of Sciences, Guangzhou, China ^bGuangzhou Center of Gas Hydrate Research, Chinese Academy of Science, Guangzhou, China ^cGuangdong Key Laboratory of New and Renewable Energy, China ^bSecond affiliation, Address, City and Postcode, Country

Abstract

The widespread need for the removal of carbon dioxide (CO₂) and hydrogen sulfide (H₂S) from process gas streams associated with hydrogen synthesis plants, upgrading of natural gas, landfill gas recovery, and enhanced oil recovery has led to a continuing interest in developing acid gas separation technologies. This work experimental investigated the hydrate formation processes for capturing acidic gases (CO₂ and H₂S) from mixture gas (biogas and natural gas) with a new synergic additive, which is composed physical gas solvent (TMS) and traditional hydrate promoter (TBAB). Based on the thermodynamic and kinetic studies, the results show that the hydrate formation process with the synergic additives can capture CO₂ and H₂S synchronously. The synergic additives not only can considerably moderate the equilibrium hydrate formation conditions, but also make high selectivity of CO₂ and H₂S over CH₄ during the hydrate formation process, and improve the formation rate and gas storage capacity through their excellent promote effect on the solubility and diffusivity of acidic gases. It will be of practical interest in relation to the fundamental study of enhancing gas hydrate formation and of potential importance for the industry application of gas hydrate.

Keywords: Capture, Acidic gas, gas hydrate, CO2, H2S, synergic additive

1. Introduction

Acid gases like carbon dioxide (CO₂) or hydrogen sulfide (H₂S) components are usually associated with the gas stream processes of hydrogen synthesis plants, upgrading of natural gas, landfill gas recovery, and enhanced oil recovery. All the gasification processes include an acid gas cleanup step, regardless of the feed stock used or the ultimate use of the synthesis gas produced. Approximately 5% of worldwide natural-gas reserves are contaminated with H₂S in excess of 15% concentration [1]. Over 30% of the current natural gas production in the US is in the form of low-quality natural gas that contains significant amounts of CO₂ and H₂S [1,2]. Acid gases must be removed from natural gas in order to: (1) Increase the heating value of natural gas; (2) Decrease the volume of gas transported in pipe-lines; (3) Reduce corrosion during the transport and distribution of natural gas and (4) Prevent atiomspheric pollution by SO₂, which is generated during the combustion of natural gas containing H₂S [2]. H₂S removal from natural gas is more challenging than CO₂ removal. Gas-pipeline specifications typically require removal down to levels of 1-3% CO₂ whereas H₂S needs to be reduced to as low as 3 ppm (v/v) total sulphur. H₂S also represents a greater health and safety hazard as compared to CO₂.

Biogas includes landfill gas, digester gas (from wastewater treatment plants) and biogas from the decomposition of animal waste or food processing waste is another burgeoning resource of acid gases[3]. It is naturally formed from the breakdown of organic waste materials in a low-oxygen (e.g., anaerobic) environment, and composes primarily of methane (typically 55% - 70% by volume) and carbon dioxide (typically 30% - 45%), and also includes smaller amounts of hydrogen sulfide (typically 50 - 2000 parts per million [ppm]). Due to its lower methane content (and therefore lower heating value) compared to natural gas, biogas use is generally limited to engine-generator sets and boilers adapted to combust biogas as fuel. Acid gases may have to be removed (selectively) from these gas streams for operational, economical or environmental reasons [4].

The most common process for the removal of H₂S and CO₂ is amine treatment. However, this becomes energetically and chemically demanding at high contaminant concentrations, which has led to work towards the development of other methods for contaminant removal. It is well known that natural gas emerges from the ground at a pressure of 5-25 MPa and both CO₂ and H₂S are known to form clathrate hydrates (abbreviated hydrates) at lower pressures than the hydrocarbon components of natural gas. So it is expect that the gas-separation technology utilizing hydrate formation can be effective for removing CO_2 and H₂S from natural gas. Gas hydrate crystallization processes has evolved as potential novel separation technology for carbon dioxide capture and sequestration, and separation of different gases from flue gas streams and so no [5],[6]. Compared with the conventional gas separation technologies, the hydrate-based gas separation method can consume the lowest energy potential due to the following factors: (1) The hydrate operating temperature is relatively moderation and the temperature difference between the formation and the decomposition sectors is less than 10 K, which is remarkably energy potential than that of conventional separation methods [7]. (2) The hydrate operating conditions are very close to the outlet condition of the natural gas or biogas, moreover, the hydrate operating process has the lower pressure loss due to the isobarically operation than that of conventional separation methods [7]. (3) The hydrate formation solution can be re-circulated used and almost without any material loss [8]. However, a commercially viable hydrate based CO₂ separation process demands a moderate operation condition, a rapid hydrate formation rate and a perfect gas storage capability [9-12]. Especially the last two essentials have been the two major bottlenecks for the gas hydrate application technologies. The precious researchers proposed that a higher solubility of hydrate forming gas molecules in water and larger contact area between the hydrate formers and water can reduces the mass transfer resistance and result in a faster hydrate formation rate[10, 13, 14].

It is usually difficult to experimental measure the mixture hydrate containing H_2S formation process due to the H_2S toxicity and corrosive to the device. That is why the experimental data are scarce in the open literature. As an innovative study to investigate the mixture hydrate formation with acidic gases, in this work, the hydrate-based capture of both CO_2 and H_2S process were studied based the thermodynamic, kinetic experiments. A new synergic additive composed physical gas solvent (Tetramethylene sulfone, TMS) and traditional hydrate promoter (tetra-n-butyl ammonium bromide, TBAB) was tested. This work will be of practical interest in relation to the development of hydrate-based acidic gas separation and of potential importance for the industry application of gas hydrate.

2. Experimental section

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