

A COMBINED EXPERIMENTAL AND COMPUTATIONAL STUDY ON THE EFFECT OF THE REACTOR CONFIGURATION AND OPERATIONAL PARAMETERS ON FORMATION, GROWTH AND DISSOCIATION OF CARBON DIOXIDE HYDRATE

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ABSTRACT

Clathrate hydrate-based technologies have been considered as promising alternatives for the effective management of the climate change risks related to emissions of carbon dioxide produced by human activities. Clathrate hydrates (also called gas hydrates) are nonstoichiometric crystalline ice-like substances, consisting of a hydrogen-bonded lattice formed by water molecules (host) and entrapped gas molecules (guest), which are not bonded with the lattice. Specifically, the water molecules form H-bonded frameworks of particular structures and enclose guests of various molecular dimensions such as methane, and CO₂, into a crystalline network of Eulerian polyhedral [1]. Depending on the size of the guest gas molecules, different types of Hydrate crystal structures can be formed. These ice-like inclusion compounds are formed under particular conditions of temperature and pressure. One of the advantages of gas hydrates is the drastic reduction of the volume of the enclathrated gas compared with that in the gaseous phase. The environmentally friendly profile of the technique, the low energy costs and its simplicity constitute some of the evident benefits of this process, comparing to conventional methods for capturing CO₂. Under standard temperature and pressure, the volume of gas stored in 1 m³ of CO₂ hydrate is about 120-160 m³ [2]. The large uptake of CO₂ in clathrate hydrates renders them a promising option for capture and storage [3,4], flow assurance and safe CO₂ pipeline transportations [5], or even disposing and sequestration [6], and also for CO₂-rich natural gas upgrade [7].

This work presents a combined experimental and computational investigation of the effects of experimental parameters, operational procedures, and experimental configuration characteristics on the phase diagrams of CO₂-H₂O systems and CO₂ hydrates formation, growth and dissociation conditions and a complete demonstration of the experimental procedure and the incipient and stability zone points. The operational modes involved (i) the incremental (step-wise) temperature cycling and (ii) the continuous temperature cycling processes, in the framework of an isochoric pressure search method. The determined (P, T) conditions for hydrate formation and dissociation, and the respective phase diagrams were compared with respect to the operational conditions, temperature cycling mode and utilized PVT configuration.

KEYWORDS: CO₂ hydrates, CO₂ capture, CO₂-H₂O phase diagrams, hydrate formation and dissociation.

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