

Thermodynamic Stability of CH₄/N₂/CO₂ Mixed Hydrates for Methane Hydrate Production by Replacement Method Using N₂/CO₂ Gas mixture

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In this study, thermodynamic stability and guest distribution of CH₄/N₂/CO₂ mixed hydrates with various compositions were investigated. Phase equilibria of the CH₄/N₂/CO₂ mixed hydrates were measured to determine the thermodynamic stability of gas hydrate deposits replaced by a N₂/CO₂ gas mixture. Moreover, composition analysis and ¹³C NMR measurement were also done to observe the guest behavior in the CH₄/N₂/CO₂ hydrates. The experimental results clearly show the preferential occupation of N₂ and CO₂ in the small 5¹² and large 5¹²6² cages of the CH₄/N₂/CO₂ hydrates, respectively. This study contributes to a better understanding on phase equilibria and guest dynamics related to CH₄ - flue gas replacement for CH₄ production and CO₂ sequestration in natural gas hydrate deposits.