Influence of feed gas compositions on replacement efficiency and structural transformation in the sH (CH $_4$ + MCP) – (CO $_2$ + N $_2$) replacement

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Natural gas hydrates have been considered as future energy resources due to their huge amount of deposits. Among various production methods, the ${\rm CH_4-CO_2}$ replacement has been studied as a promising technology which has a dual function of ${\rm CH_4}$ production and ${\rm CO_2}$ sequestration. In this study, ${\rm CO_2+N_2}$ gas were injected into the ${\rm CH_4}$ + methylcyclopentane (MCP) hydrate of structure H (sH) to examine the influence of feed gas compositions on structural transformation and guest exchange behavior in the sH hydrates. Three different gas mixtures (${\rm CO_2}$ 20%, 40%, and 60%) were used. The replacement efficiency was measured using GC and the structural transition was confirmed through $^{13}{\rm C}$ NMR and PXRD with Rietveld analysis. The experimental results showed that higher ${\rm CO_2}$ compositions in the feed gas contributed to the increased structural transition to structure I and also resulted in lower guest exchange due to lower ${\rm N_2}$ inclusion. Because of the trade-off between structural transition and N2 inclusion, the highest replacement efficiency was found at the feed gas composition of ${\rm CO_2}(40\%) + {\rm N_2}(60\%)$.