Thermodynamic and kinetic inhibiton effects of diamines on the CH_4 hydrate formation

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In this study, piperazine ($C_4H_{10}N_2$, PZ) and hydrazine (N_2H_4, HZ) were selected as gas hydrate inhibitors to examine their dual inhibition effects on the CH_4 hydrate formation. The three-phase (hydrate (H)-liquid water (Lw)-vapor (V)) phase equilibria of the CH_4 hydrate with and without diamines were measured to identify their thermodynamic inhibition effects. HZ showed more significant thermodynamic inhibition effect than PZ. ^{13}C NMR measurement revealed that the structure of CH_4 hydrate with diamine is sI and the area ratio of the large to small cages of the hydrate is approximately 3.3 This result indicates that PZ and HZ are not be captured in the hydrate cages, and they just disrupt hydrogen bonding of water molecules of the host cages. The induction time and hydrate growth patterns were examined using in-situ Raman spectroscopy, and the result demonstrates that both PZ and HZ can also function as kinetic hydrate inhibitors. Furthermore, the interaction energy between hydrate cage and inhibitors was calculated by DFT to support the dual functional inhibition effects of diamines.