

### Nitrogen and Carbon Dioxide Hydrate Phase Behavior in Small Porous Silica Gels: Three-Phase Equilibrium and Thermodynamic Modeling

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In the present study, we examined methane hydrate in porous silica gels to see the effect of natural sandstones in deep sea environments. For developing the CH<sub>4</sub>-CO<sub>2</sub> swapping method, we first need many different types of phase equilibrium data including three major guests of CH<sub>4</sub>, CO<sub>2</sub>, and N<sub>2</sub>. Here, we adopted the saturated water in silica gel pores in order to substantially enhance active surface area for contacting and encaging gas molecules. We measured the three-phase hydrate (H)-water-rich liquid (L<sub>w</sub>)-vapor (V) equilibria of CO<sub>2</sub>+N<sub>2</sub> gas hydrates in 6.0, 15.0 and 30.0nm silica gel pores to investigate the effect of geometrical constraints on the phase equilibrium patterns. The hydrate stability region is shifted to a high pressure region as the pore size of silica gel decreases. We also confirmed that the calculated values of phase equilibria showed good agreement with experimental data. The overall experimental results are essential for both exploitation of the natural gas hydrate in marine sediments and sequestration process of carbon dioxide into the deep ocean.