Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

SUPPLEMENTARY MATERIAL

Monte Carlo Simulations of the Separation of a Gas Mixture (CH_4 + CO_2) using Hydrates

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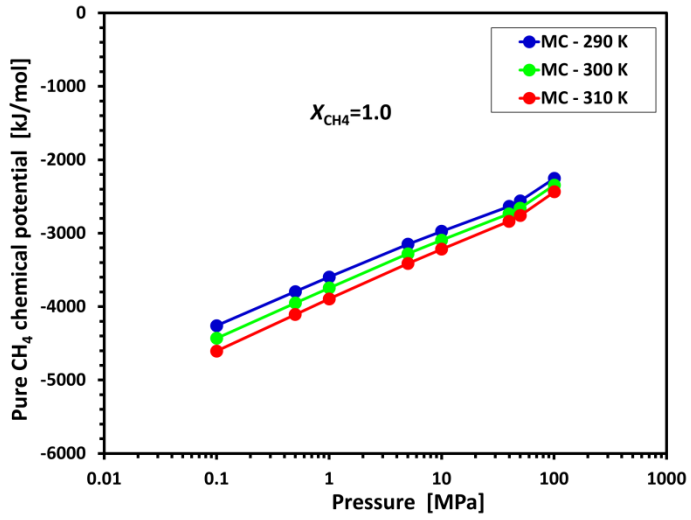
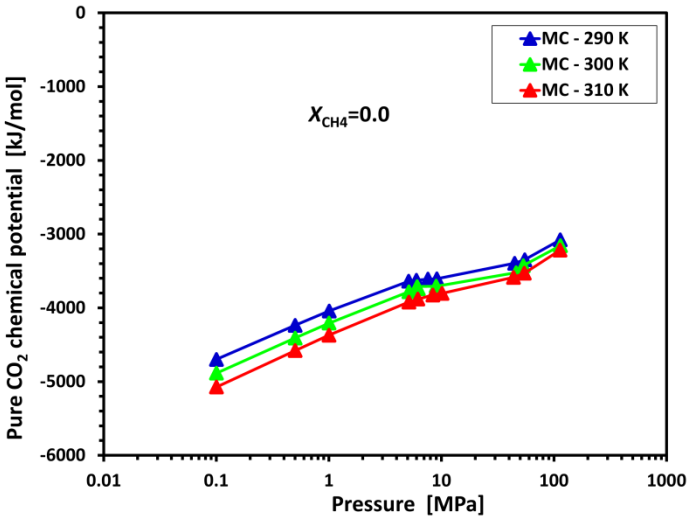


(a)



(b)

Figure S–1. Chemical potential of pure CH_4 (a) and CO_2 (b) as a function of pressure. GCMC data points are denoted with symbols (circles – CH_4, triangles – CO_2). Solid lines are guides to the eye only. Confidence limits are smaller than the symbol sizes.

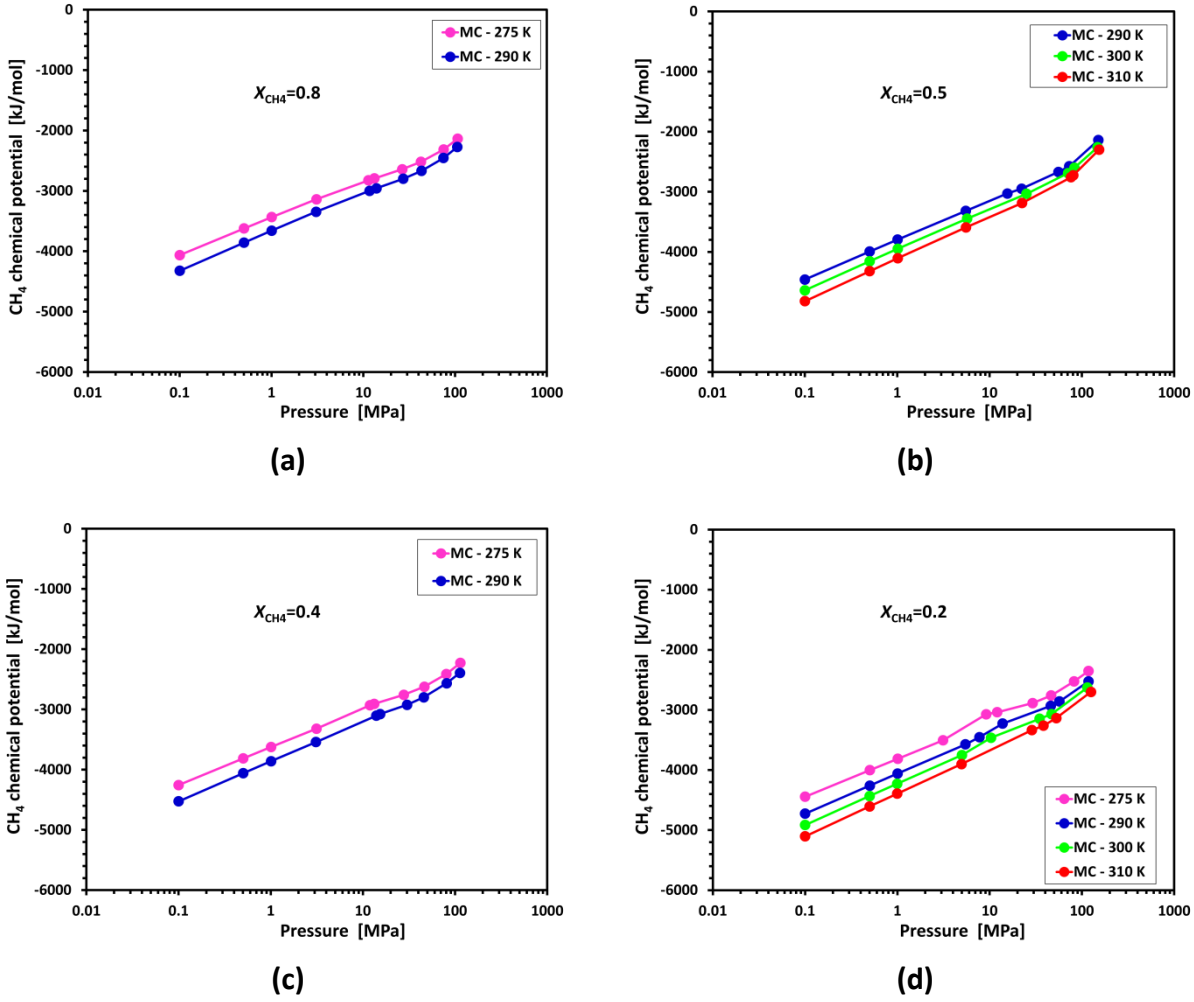


Figure S–2. Chemical potential of CH_4 as a function of pressure for a binary gas mixture containing CH_4 + CO_2 for four values of the mole fraction of CH_4. GCMC data points are denoted with symbols (circles – CH_4). Solid lines are guides to the eye only. Confidence limits are smaller than the symbol sizes.

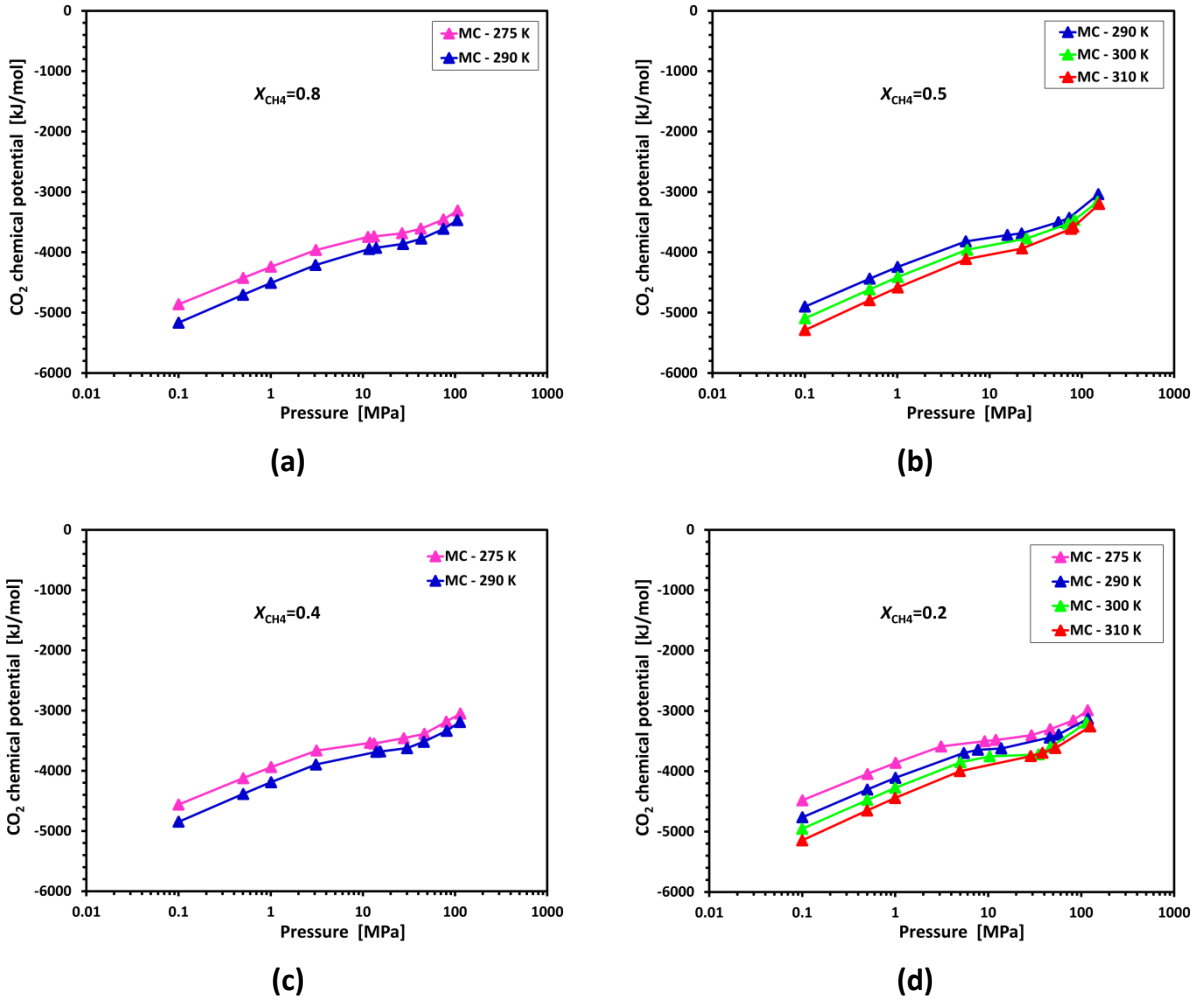


Figure S–3. Chemical potential of CO_2 as a function of pressure for a binary gas mixture containing CH_4 + CO_2 for four values of the mole fraction of CH_4. GCMC data points are denoted with symbols (triangles – CO_2). Solid lines are guides to the eye only. Confidence limits are smaller than the symbol sizes.

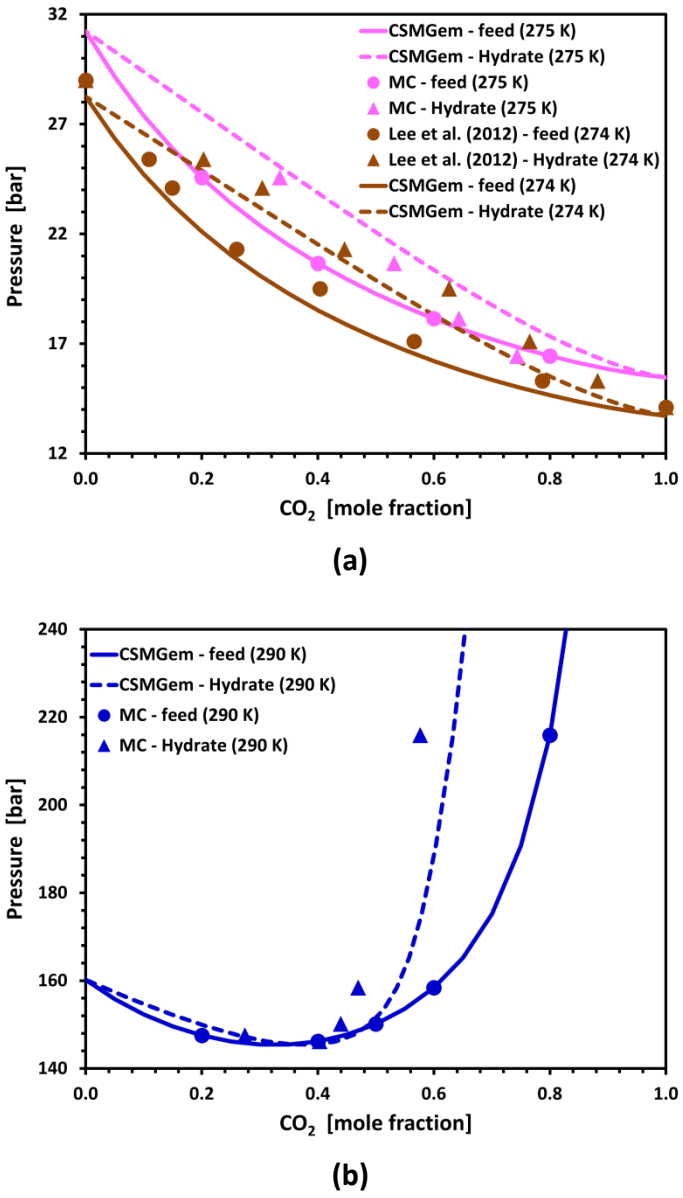


Figure S–4. Pressure as a function of CO_2 composition (mole fraction) in gas feed, *z*, and hydrate phase, *h*, for two temperatures denoted with different color: (a) 274 K (brown color), and 275 K (magenta color), and (b) 290 K (blue color). Lines denote calculations using the CSMGem simulator (Sloan and Koh, [18]), magenta and blue points denote GCMC simulations, and brown points denote experimental measurements reported by Lee *et al.* [44]. Confidence limits are smaller than the symbol sizes.

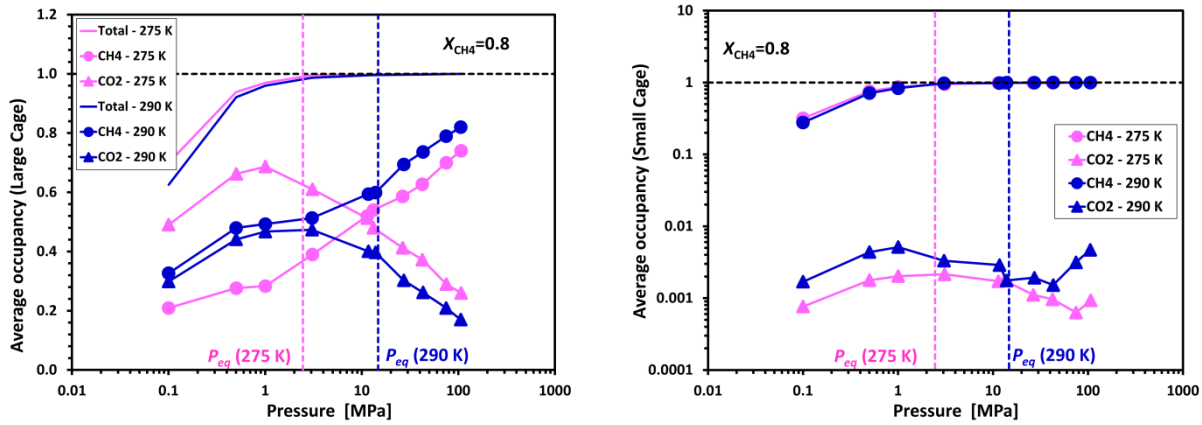


Figure S–5. Average cage occupancies for large (a) and small (b) cages as a function of pressure for a binary gas mixture containing CH_4 + CO_2 with the feed mole fraction of CH_4 equal to 0.8. GCMC data points are denoted with symbols (circles – CH_4, triangles – CO_2). Confidence limits are smaller than the symbol sizes. Lines are guides to the eye only. Vertical dashed lines denote the hydrate equilibrium pressure for the particular temperature. Color code: magenta – 275 K and blue – 290 K.

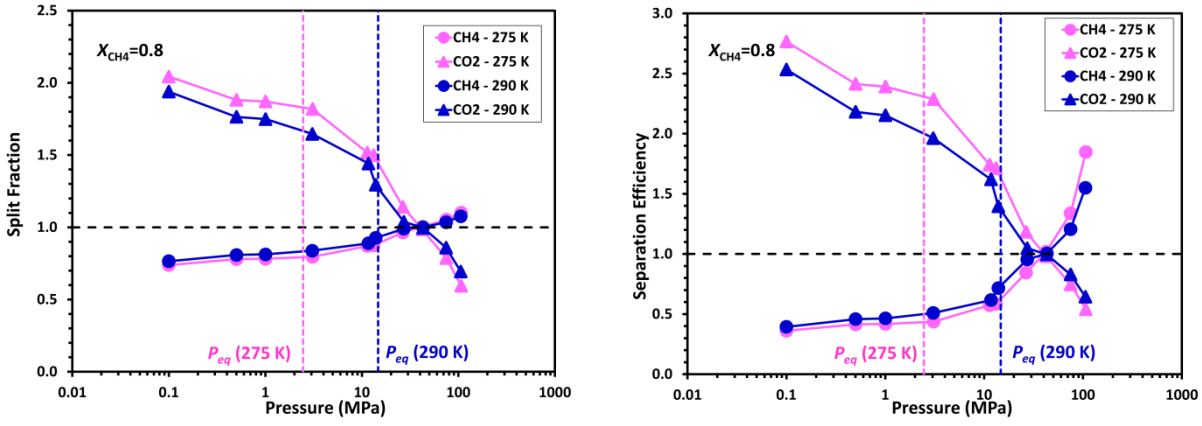


Figure S–6. Efficiency of the gas mixture separation using hydrates as a function of pressure for a binary gas mixture containing CH_4 + CO_2 with the feed mole fraction of CH_4 equal to 0.8: (a) Split Fraction, and (b) Separation Efficiency. GCMC data points are denoted with symbols (circles – CH_4, triangles – CO_2). Confidence limits are smaller than the symbol sizes. Lines are guides to the eye only. Vertical dashed lines denote the hydrate equilibrium pressure for the particular temperature. Color code: magenta – 275 K and blue – 290 K.

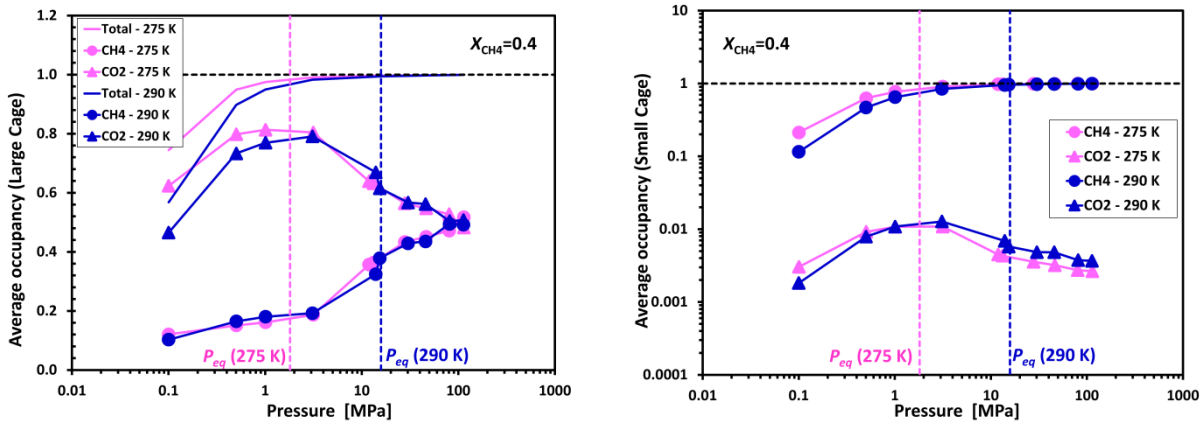


Figure S–7. Average cage occupancies for large (a) and small (b) cages as a function of pressure for a binary gas mixture containing CH_4 + CO_2 with the feed mole fraction of CH_4 equal to 0.4. GCMC data points are denoted with symbols (circles – CH_4, triangles – CO_2). Confidence limits are smaller than the symbol sizes. Lines are guides to the eye only. Vertical dashed lines denote the hydrate equilibrium pressure for the particular temperature. Color code: magenta – 275 K and blue – 290 K.

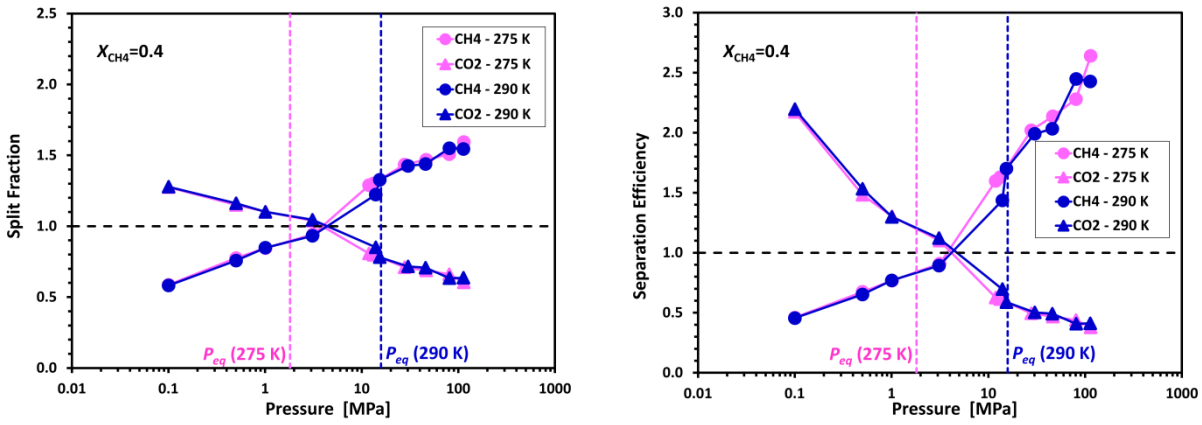


Figure S–8. Efficiency of the gas mixture separation using hydrates as a function of pressure for a binary gas mixture containing CH_4 + CO_2 with the feed mole fraction of CH_4 equal to 0.4: (a) Split Fraction, and (b) Separation Efficiency. GCMC data points are denoted with symbols (circles – CH_4, triangles – CO_2). Confidence limits are smaller than the symbol sizes. Lines are guides to the eye only. Vertical dashed lines denote the hydrate equilibrium pressure for the particular temperature. Color code: magenta – 275 K and blue – 290 K.

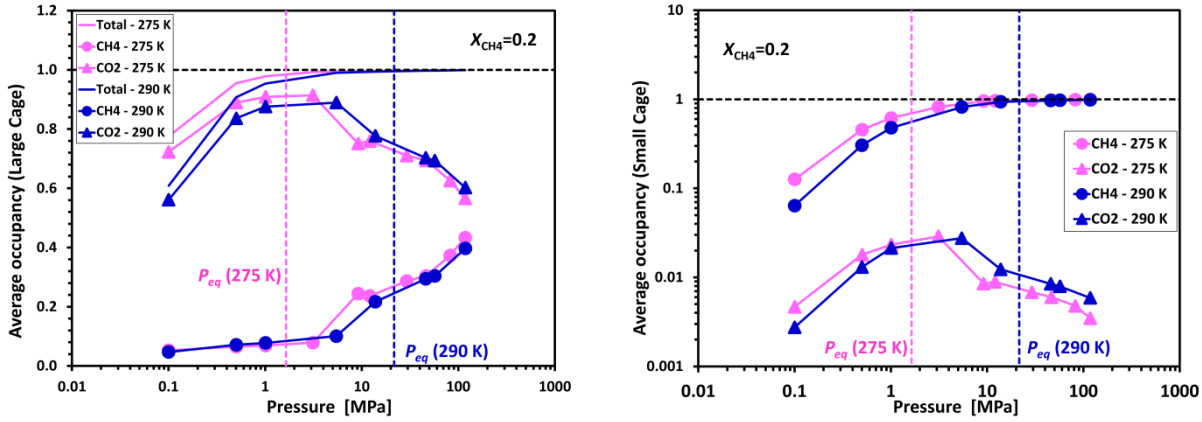


Figure S–9. Average cage occupancies for large (a) and small (b) cages as a function of pressure for a binary gas mixture containing CH_4 + CO_2 with the feed mole fraction of CH_4 equal to 0.2. GCMC data points are denoted with symbols (circles – CH_4, triangles – CO_2). Confidence limits are smaller than the symbol sizes. Lines are guides to the eye only. Vertical dashed lines denote the hydrate equilibrium pressure for the particular temperature. Color code: magenta – 275 K and blue – 290 K.

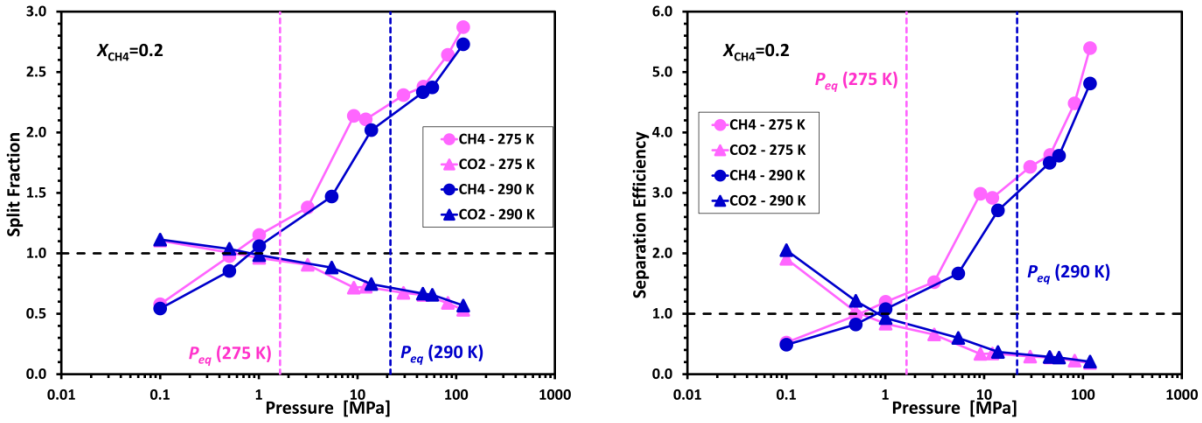


Figure S–10. Efficiency of the gas mixture separation using hydrates as a function of pressure for a binary gas mixture containing CH_4 + CO_2 with the feed mole fraction of CH_4 equal to 0.2: (a) Split Fraction, and (b) Separation Efficiency. GCMC data points are denoted with symbols (circles – CH_4, triangles – CO_2). Confidence limits are smaller than the symbol sizes. Lines are guides to the eye only. Vertical dashed lines denote the hydrate equilibrium pressure for the particular temperature. Color code: magenta – 275 K and blue – 290 K.