

Supplementary Information

Table S1. Density of CO<sub>2</sub> and CH<sub>4</sub> mixtures with different molar ratios at different temperatures and pressures

Gas	Condition	Density
CO <sub>2</sub>	323 K/15 MPa	696.80
25% CH <sub>4</sub> +75%CO <sub>2</sub>	323 K/15 MPa	400.62
	323 K/15 MPa	260.33
50% CH <sub>4</sub> +50%CO <sub>2</sub>	343 K/30 MPa	386.14
	363 K/45 MPa	447.54
	383 K/60 MPa	482.53
CH <sub>4</sub>	323 K/15 MPa	105.50

Tables S1 shows the densities of supercritical CO<sub>2</sub>/CH<sub>4</sub> mixtures at different mole ratios and the density of a 50% CH<sub>4</sub>+50% CO<sub>2</sub> mixture at different temperatures and pressures. The density of pure methane and CO<sub>2</sub> under the conditions of 323 K and 15 MPa is 701.08 kg/m<sup>3</sup> and 102.48 kg/m<sup>3</sup> respectively (according to NIST), which is very close to our calculated values. Additionally, the density of the supercritical CO<sub>2</sub>/CH<sub>4</sub> mixture calculated in our simulation is very similar to the density reported in the database by the Aimoli group [Aimoli et al., 2014]. This indicates that the system is in a critical state.

[1] C. G. Aimoli, E. J. Maginn and C. R. A. Abreu, *Journal of Chemical & Engineering Data*, 2014, 59, 3041-3054.