Supporting information for

Molecular behavior of hybrid gas hydrate nucleation: separation of soluble H_2S from mixed gas

Yi Lu[1], Chengyang Yuan, Xin lv[2], Qingping Li[2], Lei Yang[1]*, Lunxiang Zhang[1], Jiafei Zhao[1], and Yongchen Song[1]*

[1] *Key Laboratory of Ocean Energy Utilization and Energy Conservation of Ministry of Education, Dalian University of Technology, Dalian, 116024, China*

[2] *State Key Laboratory of Natural Gas Hydrate, Beijing, China, 100028*

*Corresponding Authors: leiyang@dlut.edu.cn (Lei Yang); songyc@dlut.edu.cn (Yongchen Song)

Table S1 Potential parameters for water, methane, H_2S.

Model	ε (kJ/mol)	σ(Å)	$q_H/q_C/q_O$	d_{OC} (Å)	d_{OH} (Å)	d_{OM} (Å)	∠H-O-H
TIP4p/ice	0.8822	3.1668	0.5897		0.9572	0.1577	104.52
OPLS/CH4	1.2300	3.7300					
TIP4p/S	2.23659	3.6900	0.6610				
TIP4p/MS	0.0	0.0	-1.2170				
TIP4p/HS	0.0	0.0	0.2780				

Table S2 Nucleation time for all hydrate simulation systems.

system/index	1	2	3	4	5	6	7	8	9	10
0%H_2S/CH_4	215	126	90	246	157	NAN	934	215	NAN	129
10%H_2S/CH_4	83	54	95	163	118	33	556	126	95	33
30%H_2S/CH_4	48	129	101	61	70	74	51	48	129	101
50%H_2S/CH_4	52	16	41	23	25	40	19	36	22	19
70%H_2S/CH_4	9	12	8	16	31	15	7	8	17	27
90%H_2S/CH_4	1	8	10	1	7	6	3	59	1	9
100%H_2S/CH_4	1	6	10	16	7	5	7	19	24	1

Table S3 Average induction time and nucleation rate for all hydrate simulation systems.

System (H_2S/CH_4)	0%	10%	30%	50%	70%	90%	100%
Induction time (ns)	411.2	135.6	82.1	29.3	15.0	10.5	9.7
Nucleation rate ($\times 10^{25}$)	0.462	7.139	11.65	32.457	62.418	87.436	94.386
			5				

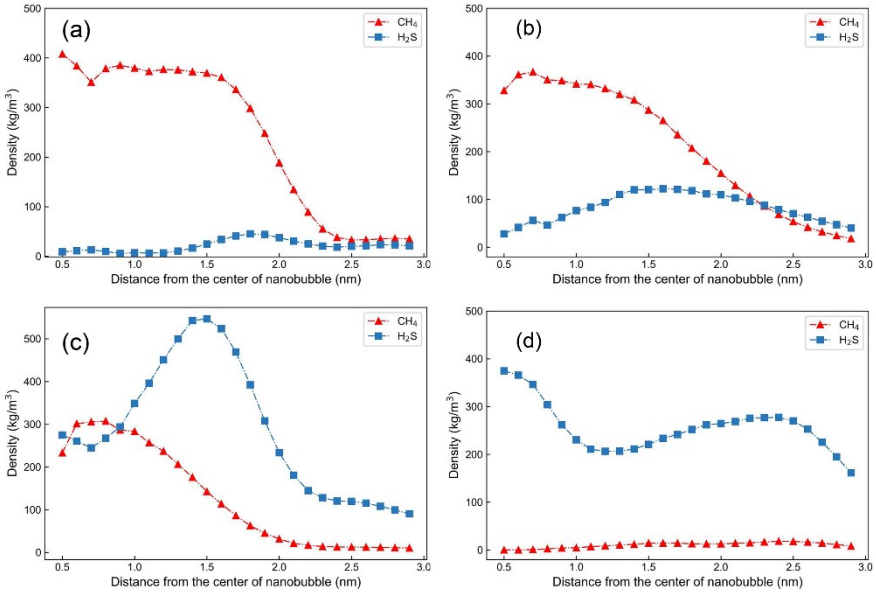

Figure S1 Radial density distribution of the two types of guest molecules in the nanobubble. (a)-(d) are the results for 10%, 30%, 70%, 90% H_2S system, respectively. The red and blue lines represent the results for CH_4 and H_2S.

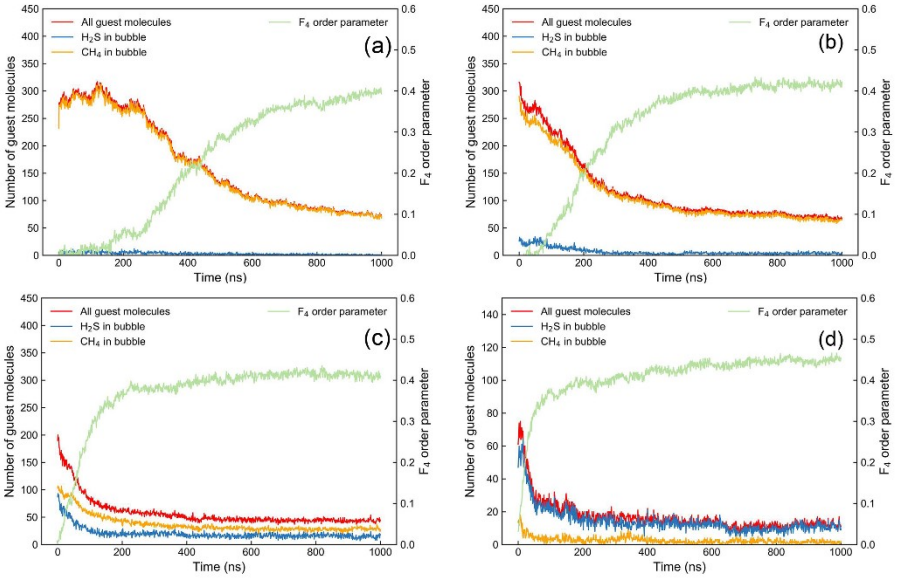

Figure S2 Evolution of the number of guest molecules in nanobubble and F_4 order parameter for (a) 10%, (b) 30%, (c) 70%, (d) 90% H_2S/CH_4 system.

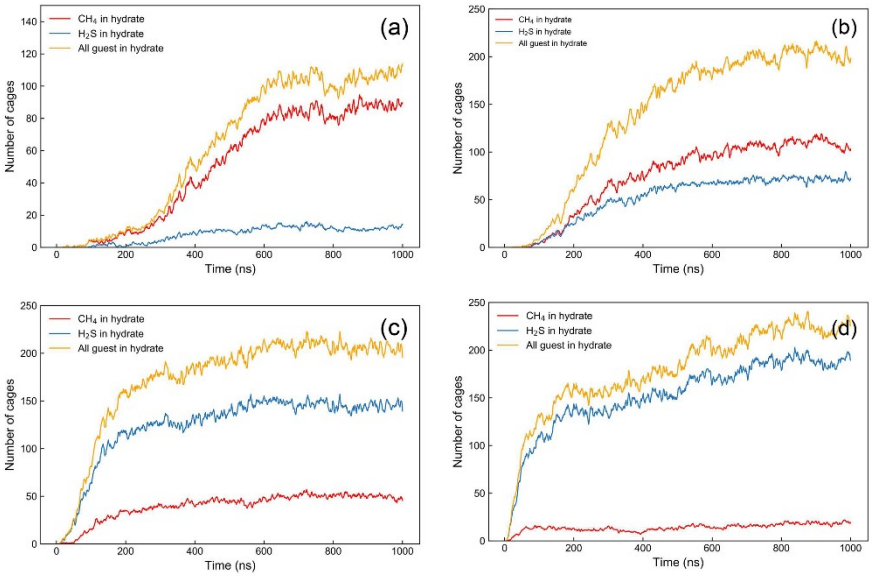

Figure S3 Evolution of cages occupied by mixed gas for (a) 10%, (b) 30%, (c) 70%, (d) 90% H_2S/CH_4 system.

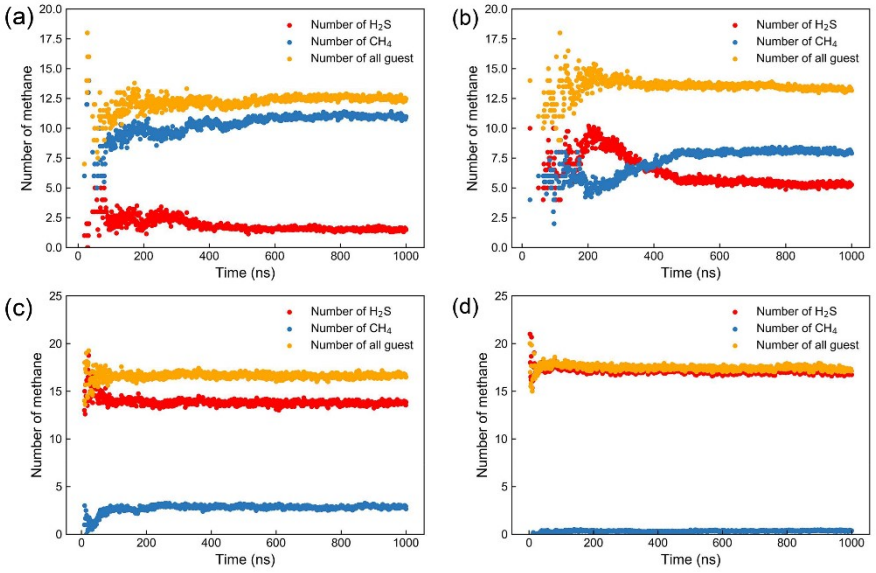

Figure S4 Example of typical evolution of the average number of guest molecules surrounding the cages for (a) 10%, (b) 30%, (c) 70%, (d) 90% H_2S/CH_4 system.

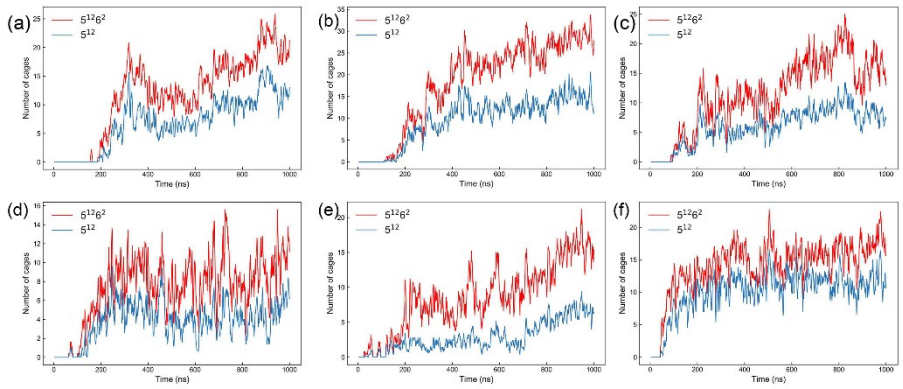

Figure S5 Example of typical evolution of the average number of 5^{12} and $5^{12}6^2$ cages in sI and SII crystal hydrate cluster for (a) 10%, (b) 30%, (c) 70%, (d) 90%, (f) 100% H_2S/CH_4 system.

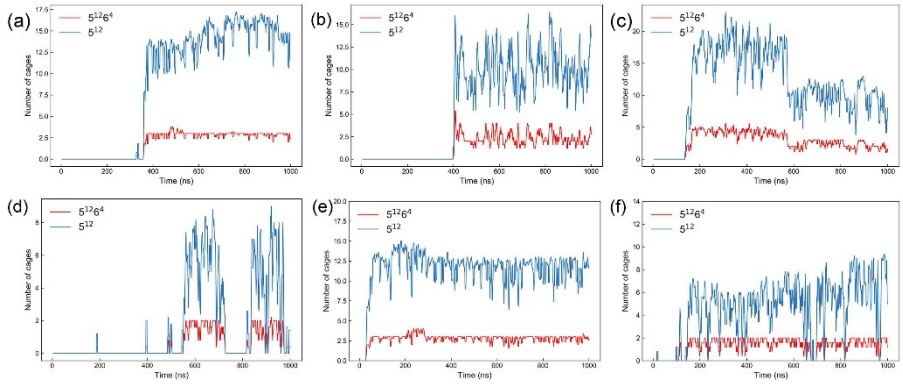

Figure S6 Example of typical evolution of the average number of 5^{12} and $5^{12}6^4$ cages in sI and SII crystal hydrate cluster for (a) 10%, (b) 30%, (c) 70%, (d) 90%, (f) 100% H_2S/CH_4 system.

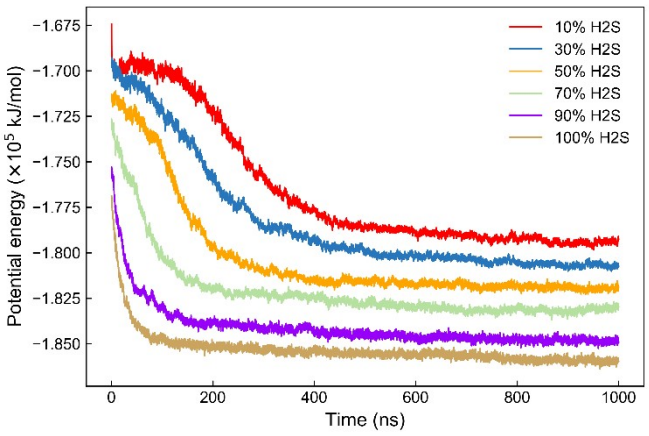

Figure S7 Typical potential energy evolutions for seven different initial H_2S/CH_4 proportion.

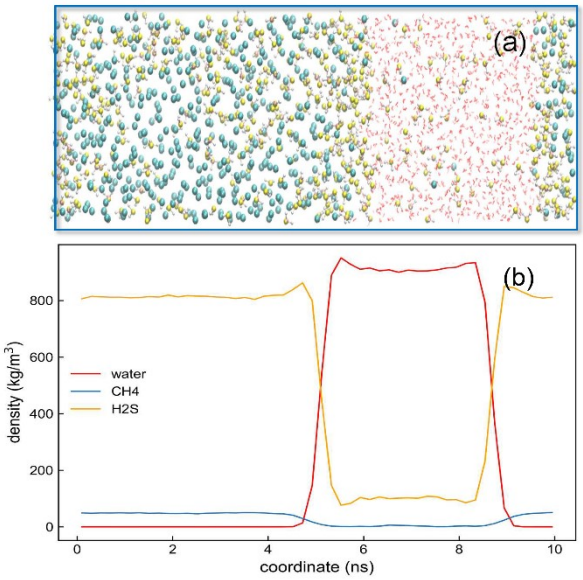

Figure S8 Typical final configuration and density distribution results for the simulation to calculate solubility with a flat interface. (a) typical configuration; (b) density distribution for the last 10 ns of the simulation.

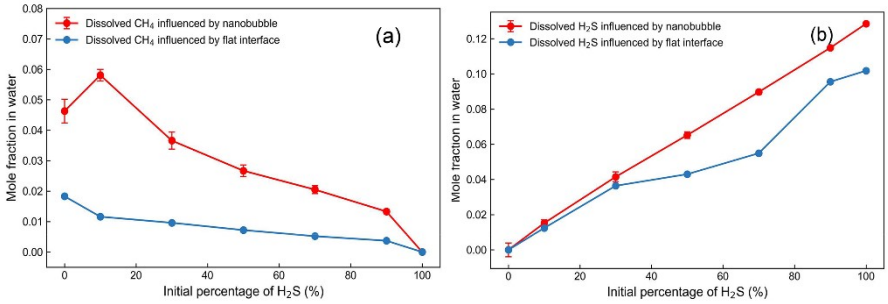

Figure S9 Solubility results for simulation with a flat interface at 250 K and 500 bar. (a) for methane; (b) for H_2S.