Supporting information for:

Voronoi Tessellation Analysis of Clathrate Hydrates

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Table S1: Properties of Voronoi polyhedra of a perfect sI hydrate unit cell.

#	Polyhedron	n_{edges}	n_{vertices}	n_{faces}	S (Å^2)	V (Å^3)	η
1	5^{12}	30	20	12	184.82	204.52	1.335
2	5^{12}	30	20	12	184.82	204.52	1.335
3	$5^{12}6^2$	36	24	14	187.37	211.06	1.306
4	$5^{12}6^2$	36	24	14	187.37	211.06	1.306
5	$5^{12}6^2$	36	24	14	187.37	211.06	1.306
6	$5^{12}6^2$	36	24	14	187.37	211.06	1.306
7	$5^{12}6^2$	36	24	14	187.37	211.06	1.306
8	$5^{12}6^2$	36	24	14	187.37	211.06	1.306

S is the surface area; V is the volume.
$\eta = S^3/(36\pi V^2)$; $\eta = 1$ for a sphere

Table S2: Properties of Voronoi polyhedra of a perfect sII hydrate unit cell.

#	Polyhedron	n_{edges}	n_{vertices}	n_{faces}	S (Å^2)	V (Å^3)	η
1	5^{12}	30	20	12	183.34	200.39	1.338
2	5^{12}	30	20	12	183.34	200.39	1.338
3	5^{12}	30	20	12	183.34	200.39	1.338
4	5^{12}	30	20	12	183.34	200.39	1.338
5	5^{12}	30	20	12	183.34	200.39	1.338
6	5^{12}	30	20	12	183.34	200.39	1.338
7	5^{12}	30	20	12	183.34	200.39	1.338
8	5^{12}	30	20	12	183.34	200.39	1.338
9	5^{12}	30	20	12	183.34	200.39	1.338
10	5^{12}	30	20	12	183.34	200.39	1.338
11	5^{12}	30	20	12	183.34	200.39	1.338
12	5^{12}	30	20	12	183.34	200.39	1.338
13	5^{12}	30	20	12	183.34	200.39	1.338
14	5^{12}	30	20	12	183.34	200.39	1.338
15	5^{12}	30	20	12	183.34	200.39	1.338
16	5^{12}	30	20	12	183.34	200.39	1.338
17	$5^{12}6^4$	42	28	16	203.91	246.44	1.235
18	$5^{12}6^4$	42	28	16	203.91	246.44	1.235
19	$5^{12}6^4$	42	28	16	203.91	246.44	1.235
20	$5^{12}6^4$	42	28	16	203.91	246.44	1.235
21	$5^{12}6^4$	42	28	16	203.91	246.44	1.235
22	$5^{12}6^4$	42	28	16	203.91	246.44	1.235
23	$5^{12}6^4$	42	28	16	203.91	246.44	1.235
24	$5^{12}6^4$	42	28	16	203.91	246.44	1.235

S is the surface area; V is the volume.
$\eta = S^3/(36\pi V^2)$; $\eta = 1$ for a sphere

Table S3: All other types of polyhedra obtained from Voronoi polyhedra analysis. Polyhedra with triangular faces have many other different faces therefore only their total is shown. Please note total numbers in each column of this table and of Figure 4 in the main text add to the polyhedra numbers shown in the first subplot of Figure 4.

	Time (ns)							
Polyhedra	0	100	200	300	400	500	600	700
$4^2 5^8 6^1$	0	1	3	6	5	9	11	21
$4^2 5^8 6^2$	0	0	1	8	6	14	18	20
$4^2 5^8 6^3$	0	1	0	1	3	7	10	8
$4^2 5^8 6^4$	0	0	3	2	2	5	13	16
$4^2 5^8 6^5$	0	0	1	0	0	1	6	8
$4^2 5^8 6^6$	0	0	0	0	0	0	1	2
$4^2 5^8 6^7$	0	0	0	0	0	1	0	0
$4^3 5^6 6^1$	0	0	1	0	0	0	1	2
$4^3 5^6 6^2$	0	0	0	2	0	0	1	2
$4^3 5^6 6^3$	0	2	1	2	6	8	8	9
$4^3 5^6 6^4$	0	2	4	3	1	5	7	4
$4^3 5^6 6^5$	0	1	0	0	0	0	3	1
$4^3 5^6 6^6$	0	0	0	0	0	0	1	2
$4^3 5^6 6^7$	0	0	1	0	0	0	1	1
$4^3 5^6 6^8$	0	0	1	0	0	0	1	1
$4^4 5^4 6^1$	0	0	0	0	0	3	0	0
$4^4 5^4 6^2$	0	1	2	0	0	4	2	4
$4^4 5^4 6^3$	0	1	3	1	1	1	1	3
$4^4 5^4 6^4$	0	1	0	0	1	0	1	0
$3^m 4^n 5^o 6^p 7^q$	0	1	5	6	25	41	76	97

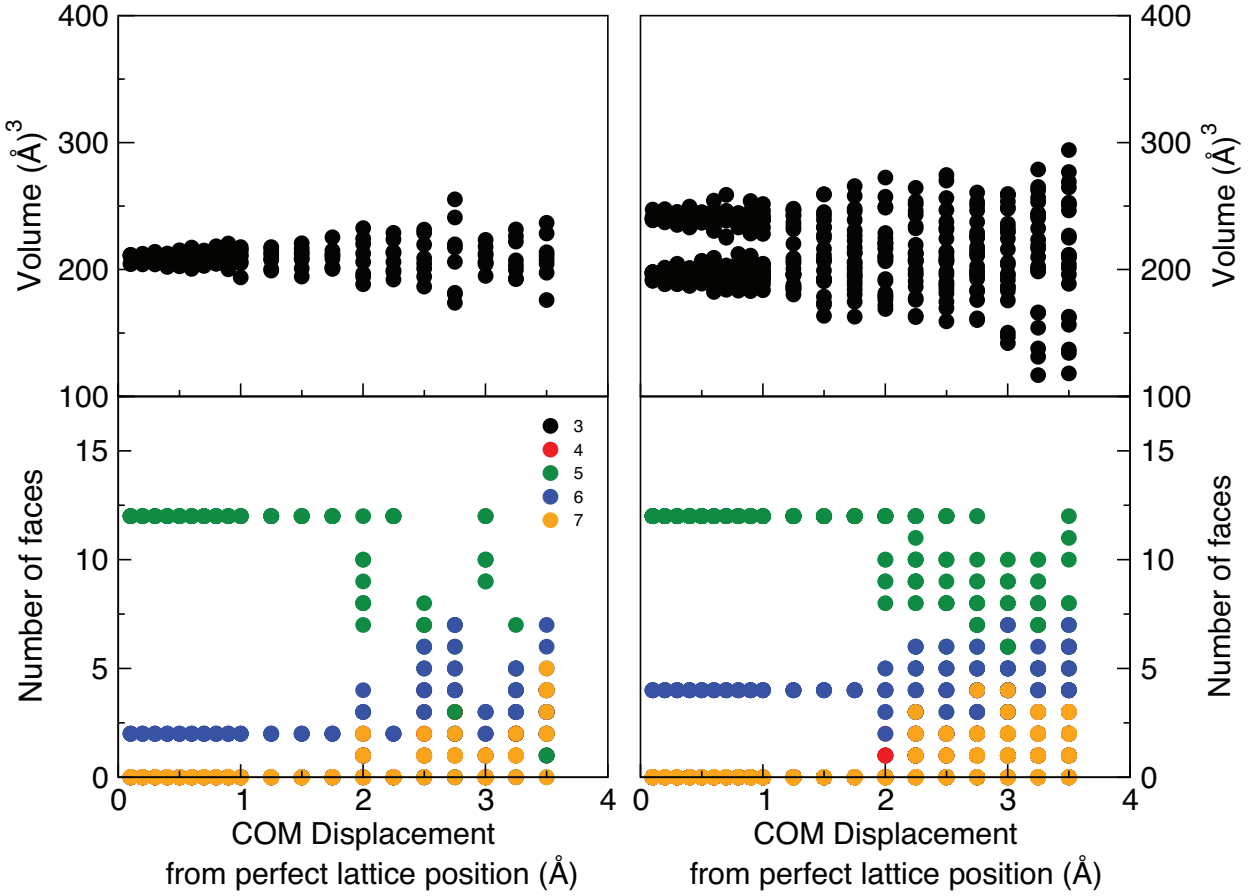

Figure S1: Voronoi polyhedra properties after tessellating randomly displaced methane coordinates from the center of the cages in sI and sII crystals.

Given below is a description of Voronoi polyhedra counterparts for the 5^{12} enclathrated methanes listed in Table 2. The number in parentheses is the quantity of the polyhedron type.

- Polyhedron containing a 5^{11} face is: $3^1 4^1 5^1 16^1 7^2 8^1$
- Polyhedra containing 5^{10} faces are all $4^1 5^{10} 6^2$
- Polyhedra containing 5^9 faces are: $3^1 4^1 5^9 7^2$ (1), $4^2 5^9 6^2 7^1$ (2), $3^1 5^9 6^3$ (1), $4^2 5^9 6^3 7^1$ (1), $3^1 4^1 5^9 6^2 7^2$ (1), $3^1 4^1 5^9 7^2$ (1), $4^3 5^9 6^4 7^1$ (1), $3^1 4^1 5^9 6^2$ (1)
- Polyhedra containing 5^8 faces are: $4^2 5^8 6^2$ (3), $4^1 5^8 6^5$ (1), $3^1 4^1 5^8 6^1 7^1$ (2), $3^1 4^1 5^8 6^3 7^1$ (1), $4^3 5^8 6^2 7^2$ (1), $4^2 5^8 6^1$ (3), $3^1 4^2 5^8 6^2 7^1$ (1)
- Polyhedra containing 5^7 faces are: $4^3 5^7 6^2 7^1$ (1), $3^1 4^2 5^7 6^3 7^2$ (2), $4^3 5^7 6^2 7^1$ (1), $3^1 4^2 5^7 6^4 7^1$ (1), $4^3 5^7 6^2 7^2$ (1)
- Polyhedra containing 5^6 faces are: $3^1 4^2 5^6 6^4 7^1$ (1), $4^3 5^6 6^4 7^4$ (1), $3^1 4^3 5^6 6^2 7^3$ (1), $4^4 5^6 6^3 7^2$ (1), $3^2 4^1 5^6 6^3 7^2$ (1), $3^1 4^1 5^6 6^2 7^2$, $3^1 4^2 5^6 6^3 7^1$ (1), $4^4 5^6 6^2 7^2$ (1)
- Polyhedron containing a 5^5 face is: $3^1 4^3 5^5 16^4 7^2$
- Polyhedra containing 5^4 faces are: $3^3 4^2 5^4 6^4 7^1$ (1), $4^5 5^4 6^3 7^3$ (1), $3^1 4^4 5^4 6^3 7^3$ (1), $3^1 4^3 5^4 6^5 7^1$ (1), $3^2 4^2 5^4 6^4 7^2$ (1)

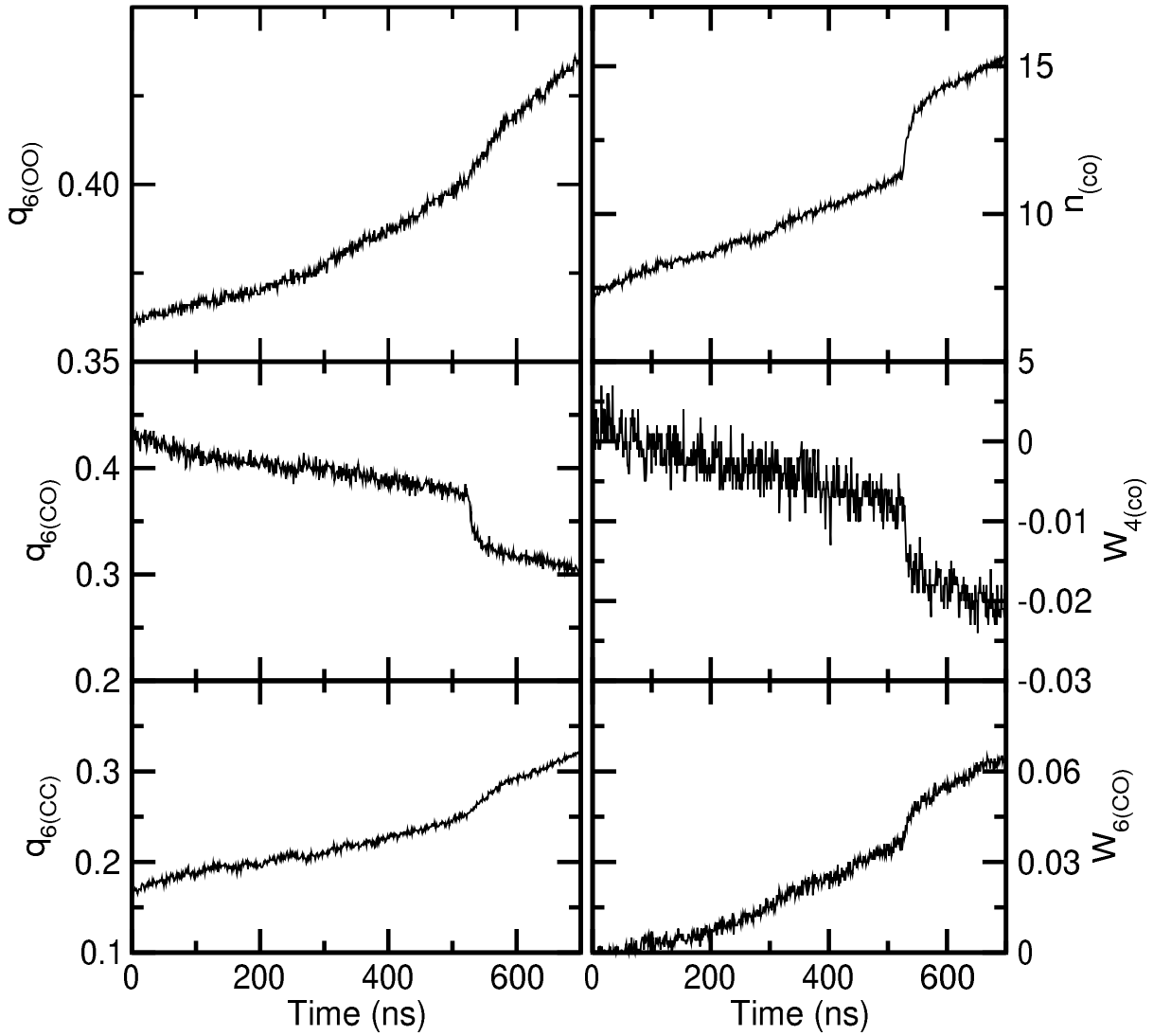

Figure S2: q_6, W_6, W_4, and coordination number order parameters calculated over the course of the trajectory for the entire system.

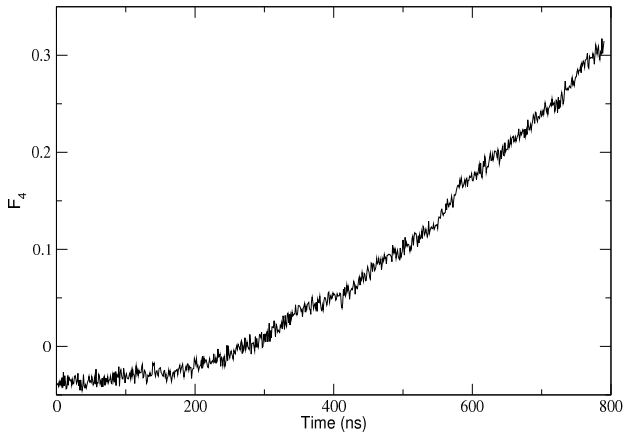

Figure S3: F_4 order parameter calculated over the course of the trajectory for the entire system.

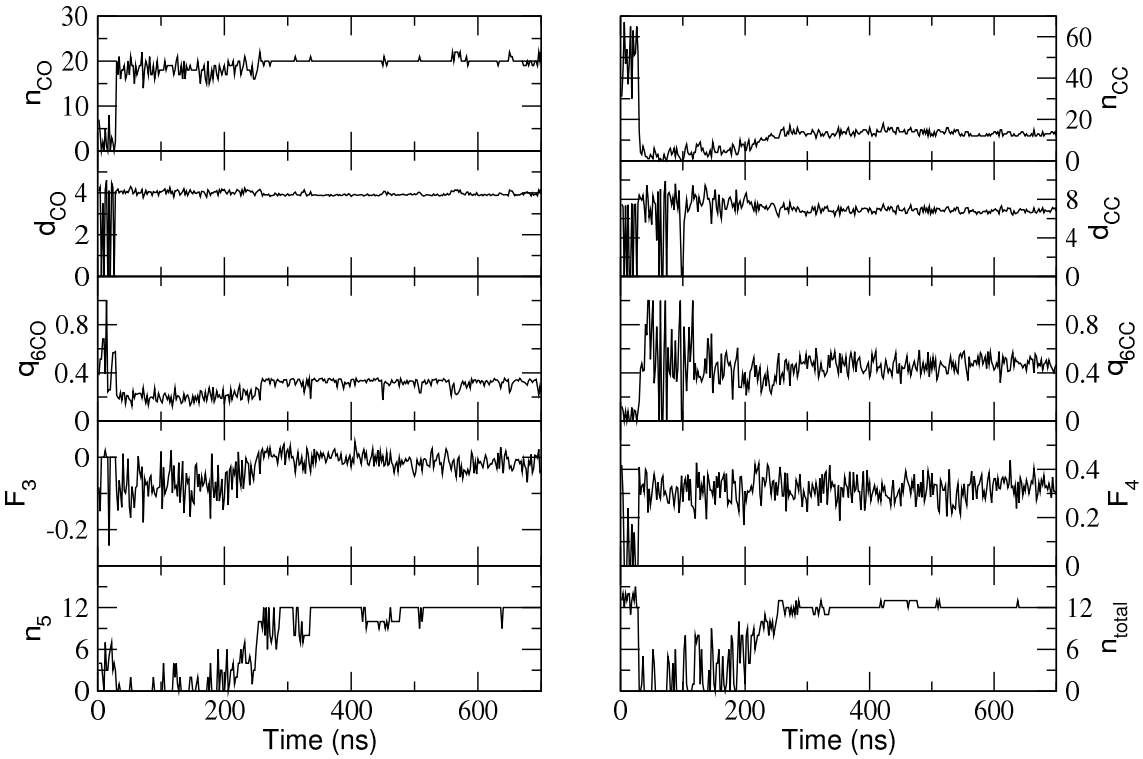

Figure S4: q_6, F_3, F_4, and coordination number order parameters calculated over the course of the trajectory for a specific methane.