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Supplementary Materials for

Reliable and practical computational description of molecular crystal polymorphs

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Published 11 January 2019, *Sci. Adv.* **5**, eaau3338 (2019)
DOI: 10.1126/sciadv.aau3338

The PDF file includes:

Other Supplementary Material for this manuscript includes the following:

(available at advances.sciencemag.org/cgi/content/full/5/1/eaau3338/DC1)

Supplementary Text

Discussion of Most-Stable Unobserved Structures

In this section, we briefly discuss several structures, which have not been observed in experiment but are very close in terms of stability to experimental structures. Let us first discuss the four systems with only one known experimental structure (XXII, XXIV, XXV, XXVI). We always discuss the relative stabilities in terms of our best ranking which includes PBE+TS vibrational free energies. For system XXII we find one structure (XXII-N3), which is only 0.5 kJ/mol less stable than the experimental structures. However, this structure is very similar to the experimental one. In our similarity search we find that 15 out of 20 molecules match with the experimental structure. For system XXIV the first 3 non-observed polymorphs (XXIV-N3, XXIV-N10, XXIV-N53) are found within 1.3 kJ/mol of the experimental structure. All of them are a partial match w.r.t. the experimental structure, i.e., at least 10 molecules out of 20 match with the experimental structure. In system XXV there are no structures within 3.8 kJ/mol. For system XXVI we find two structures which are within 0.3 kJ/mol of the experimental structure. Also in this case, both of them are very similar to the experimental structure. Structure XXVI-N5 is a partial match (16/20 molecules) and structure XXVI-N4 is in fact a complete match but has a large RMSD of 0.9 Å. Therefore, all structures which are within 1.3 kJ/mol of these experimental structures have several structural features in common with the experimental structure.

Finally, let us discuss the former drug candidate XXIII. For this system we find two structures which are described as more stable than any experimental structure (XXIII-N70 and XXIII-N5). Structure XXIII-N70 is a partial match for form A (13/20 molecules) and will be discussed in more detail below. Structure XXIII-N5 is a disordered structure with two molecules in the asymmetric unit ($Z' = 2$) and is virtually identical with form C. However, given the blind test settings for identical structures, it could not be excluded from the list since only 17 out of 20 molecules are a match. In addition, we find several $Z' = 1$ and $Z' = 2$ structures which are structurally sufficiently different and thermodynamically stable enough to potentially crystallize in experiment.

Our most stable structure (XXIII-N70) and form A (XXIII-N85) are structurally very similar, but only form A is observed experimentally despite the fact that structure N70 is predicted to be 4.1 kJ/mol more stable. Even our anharmonic free energy estimate does not significantly change the relative stability. This energy difference is too large to be attributed to a computational error. If form A crystallizes instead of structure N70, form A must be dynamically favored. The vibrational free energy stabilizes structure N70 by 2.8 kJ/mol compared to form A. Without the vibrational free energy calculation, structure N70 is still more stable than form A, but only by 1.3 kJ/mol (PBE0+MBD). The vibrational free energy difference is calculated assuming crystals of infinite size. Free energy may not yet stabilize structure N70 over form A during the nucleation phase, when crystallites are very small. However, since even without the free energy contribution structure N70 is more stable than form A, this observation cannot explain why structure N70 does not crystallize at all. Since the only notable difference between the two structure is the stacking of the sheets (see Figure S1), it may be concluded that under

the explored crystallization conditions crystal growth perpendicular to the sheets is significantly faster for the form A stacking than for the N70 stacking, potentially related to 2D nucleation on the surface. Extremely slow crystal growth may be the key to obtaining structure N70 rather than form A. Since structure N70 and form A share the same molecular conformation, the same hydrogen bonding and much of their surface chemistry, solvents favoring form A over forms B, C, D and E will also favor structure N70 over the other forms. Extremely long slurrying or extremely low melting starting from form A may be alternative ways to obtain structure N70. The main challenge will be to avoid conversion to forms B, C, D or E before conversion to structure N70 is achieved.

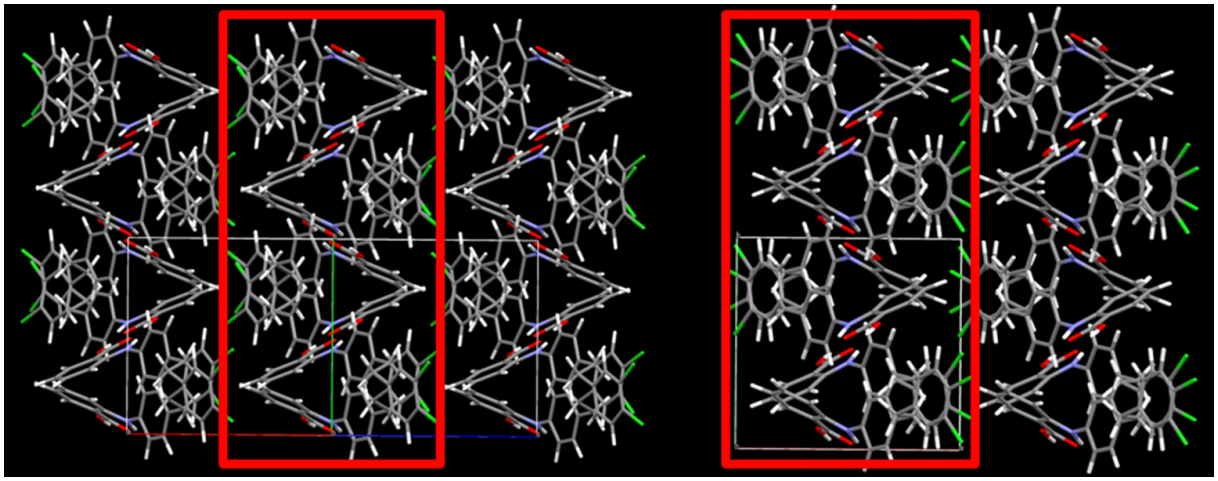

Fig. S1. Rank 1 (XXIII-N70) and form A (XXIII-N85) share a sheet structure (red boxes), in which the molecules are arranged according to the same pattern, but the sheets are stacked differently.

Table S1. Relative energies of a benchmark set of structures with small unit cells calculated with different methods. The benchmark set consists of the single $Z = 1$ structure and the top 7 $Z = 2$ structures after the PBE0+MBD ranking for system XXII, and the top 4 $Z = 2$ structures after the PBE0+MBD ranking for system XXIV. The energy of the most stable structure of a system after the PBE0+MBD ranking was set to zero for all methods. The letters after the slash indicate the used basis set and grid settings: *l* corresponds to the *light* species default settings for basis sets and grids in FHI-aims, while *t* refers to the *tight* species default settings. The label *rt* indicates the usage of the *really tight* grid settings together with tier-3 basis functions. All relative energies are in kJ/mol/molecule, or in the case of XXIV in kJ/mol/trimer. The method PBE0+MBD without a label for basis set and grid settings corresponds to the PBE0+MBD estimate used for the stability rankings in the main text and is described in the Methods section of the paper.

Structure	PBE+MBD/l	PBE+MBD/t	PBE+MBD/rt	PBE0+MBD/l	PBE0+MBD	PBE0+MBD/t
XXII-N17	0.0	0.0	0.0	0.0	0.0	0.0
XXII-N32	-0.1	0.6	0.6	0.4	1.0	0.8
XXII-N36	0.6	1.3	1.3	0.9	1.6	1.6
XXII-N9	-1.3	-0.3	-0.3	0.8	1.9	1.2
XXII-N92	1.8	1.8	1.8	2.1	2.1	2.1
XXII-N24	-2.4	0.6	0.8	-0.3	2.7	2.3
XXII-N39	0.5	1.1	1.0	2.3	2.9	2.4
XXII-N44	0.9	1.5	1.5	2.5	3.1	2.5
XXIV-N53	0.0	0.0	0.0	0.0	0.0	0.0
XXIV-N6	0.2	0.9	0.8	1.1	1.7	2.0
XXIV-N19	3.1	3.4	3.4	2.5	2.8	3.5
XXIV-N12	2.0	2.2	2.2	3.2	3.4	3.4

Table S2. Convergence of relative stabilities with basis set and grid settings. This table shows the mean absolute deviation (MAD) and the maximum absolute deviation (MAX) for all possible relative energies (within a system) from table S1 w.r.t. PBE+MBD calculations with *really tight* settings for basis set and grids.

Method	MAD [kJ/mol]	MAX [kJ/mol]
PBE+MBD/light	0.9	3.2
PBE+MBD/tight	0.1	0.3

Table S3. This table shows the MAD and the maximum absolute deviation (MAX) for all possible relative energies (within a system) from table S1 with regard to PBE0 + MBD calculations with tight settings for basis set and grids. The method PBE0+MBD without a label for basis set and grid settings corresponds to the PBE0+MBD estimate used for the stability rankings in the main text and is described in the Methods section of the paper.

Method	MAD [kJ/mol]	MAX [kJ/mol]
PBE+MBD/light	1.6	4.8
PBE+MBD/tight	0.8	1.8
PBE0+MBD/light	0.8	2.6
PBE0+MBD	0.4	0.8

Table S4. Stability ranking for system XXII in kilojoules per mole per molecule. The energy of the most stable structure in each ranking was set to zero and the structures are always ordered according to the highest available ranking level. The final ranking includes vibrational free energies calculated at 150 K. In addition, this table contains the number of molecules in the unit cell (Z), the space group (Symm.), and the density in g/cm^3.

Name	Z	Symm.	Density	PBE+TS	PBE+MBD	PBE0+MBD	PBE0+MBD+F_{vib}
XXII-N2 (Exp.)	4	P 21/n	1.677	1.7	0.7	0.0	0.0
XXII-N3	4	P 21/c	1.672	1.7	0.6	0.6	0.5
XXII-N4	4	P 21/c	1.688	2.5	1.2	1.0	1.9
XXII-N1	4	P n a 21	1.715	0.0	0.0	1.8	2.5
XXII-N19	4	P 21/n	1.669	5.2	4.1	3.9	4.1
XXII-N6	4	P 21/n	1.701	3.7	2.4	3.7	4.4
XXII-N7	4	P 21 21 21	1.703	2.8	1.9	4.3	4.8
XXII-N17	2	P 21	1.639	7.3	4.4	4.4	n/d
XXII-N27	4	P 21 21 21	1.668	6.1	5.1	4.5	n/d
XXII-N20	4	P n a 21	1.634	7.6	4.1	4.5	n/d
XXII-N35	4	P 21/n	1.673	6.3	4.6	4.8	n/d
XXII-N5	8	I 2/c	1.728	2.8	3.2	5.1	n/d
XXII-N8	4	P 21/c	1.709	4.0	3.8	5.2	n/d
XXII-N37	4	P 21/c	1.659	6.9	5.3	5.3	n/d
XXII-N32	2	P -1	1.659	7.2	4.9	5.4	n/d
XXII-N46	4	P 21/c	1.657	7.4	5.3	5.4	n/d
XXII-N13	4	P n m a	1.712	4.2	4.0	5.5	n/d
XXII-N22	4	P 21/c	1.609	7.8	3.9	5.6	n/d
XXII-N67	4	P 21/n	1.626	8.8	5.2	5.7	n/d
XXII-N52	4	P 21/c	1.663	6.8	5.5	5.9	n/d
XXII-N36	2	P -1	1.669	6.7	5.7	6.0	n/d
XXII-N42	4	P 21/n	1.659	6.9	5.3	6.0	n/d
XXII-N11	4	P n m a	1.712	4.0	3.8	6.1	n/d
XXII-N70	8	P b c a	1.607	8.0	6.0	6.2	n/d
XXII-N9	2	P -1	1.705	5.0	4.1	6.3	n/d
XXII-N34	4	P 21 21 21	1.660	7.6	5.3	6.3	n/d
XXII-N21	4	P 21/n	1.721	4.3	5.0	6.4	n/d
XXII-N18	4	P 21 21 21	1.664	7.4	4.4	6.4	n/d
XXII-N41	8	P b c a	1.684	6.4	5.6	6.5	n/d
XXII-N92	2	P 21	1.666	7.8	6.2	6.5	n/d
XXII-N53	4	P 21/c	1.661	8.0	5.9	6.6	n/d
XXII-N76	4	P -1	1.730	4.5	5.1	6.7	n/d
XXII-N16	4	P 21/n	1.708	4.6	4.6	6.7	n/d
XXII-N30	8	P 21/n	1.729	2.9	4.6	6.9	n/d
XXII-N10	4	P 21/n	1.727	3.1	3.7	6.9	n/d
XXII-N85	4	P 21/c	1.649	7.7	6.7	6.9	n/d
XXII-N26	4	P 21/n	1.657	7.2	5.0	7.0	n/d

Table S4. (continued) Stability ranking for system XXII.

Name	Z	Symm.	Density	PBE+TS	PBE+MBD	PBE0+MBD	PBE0+MBD+F_{vib}
XXII-N29	4	P 21/c	1.684	6.1	5.4	7.0	n/d
XXII-N47	4	P 21/c	1.664	7.3	6.3	7.0	n/d
XXII-N88	4	P c a 21	1.729	3.9	5.1	7.1	n/d
XXII-N24	2	P c	1.746	2.6	4.9	7.1	n/d
XXII-N25	4	P 21/c	1.724	3.2	5.7	7.2	n/d
XXII-N43	4	P 21/c	1.670	6.4	5.5	7.2	n/d
XXII-N58	4	P 21 21 21	1.696	5.8	6.0	7.3	n/d
XXII-N39	2	P 21/m	1.718	5.2	5.5	7.3	n/d
XXII-N83	2	P -1	1.614	8.6	6.5	7.3	n/d
XXII-N73	4	P 21/c	1.670	8.0	6.5	7.3	n/d
XXII-N12	4	P 21/c	1.706	4.8	4.9	7.4	n/d
XXII-N62	4	P 21/c	1.635	8.4	5.8	7.4	n/d
XXII-N44	1	P 1	1.730	5.8	5.8	7.5	n/d
XXII-N15	4	P 21/n	1.654	6.9	4.7	7.5	n/d
XXII-N40	4	P c a 21	1.744	4.0	5.7	7.6	n/d
XXII-N23	2	P m n 21	1.689	5.6	4.7	7.6	n/d
XXII-N33	4	P 21/n	1.698	5.5	6.0	7.7	n/d
XXII-N64	2	P 21	1.685	7.5	6.4	7.7	n/d
XXII-N31	8	P b c a	1.701	6.1	5.4	7.9	n/d
XXII-N86	4	P 21/n	1.698	6.2	6.7	8.1	n/d
XXII-N93	4	P c a 21	1.607	10.8	6.7	8.2	n/d
XXII-N48	4	P 21/c	1.701	6.2	5.5	8.2	n/d
XXII-N77	4	P 21/c	1.715	5.6	6.1	8.2	n/d
XXII-N28	4	P 21/n	1.689	6.3	5.1	8.2	n/d
XXII-N65	4	P 21/c	1.693	5.8	5.6	8.3	n/d
XXII-N84	8	C 2/c	1.668	8.4	7.1	8.3	n/d
XXII-N97	2	P -1	1.702	6.2	5.8	8.3	n/d
XXII-N66	4	P n a 21	1.620	10.6	5.7	8.3	n/d
XXII-N63	4	P 21 21 21	1.717	4.3	6.8	8.3	n/d
XXII-N45	8	P b c a	1.719	5.2	6.4	8.4	n/d
XXII-N55	4	P 21/c	1.664	7.6	6.8	8.4	n/d
XXII-N82	4	P 21/n	1.626	10.0	6.3	8.5	n/d
XXII-N71	4	P 21/n	1.708	5.4	6.5	8.6	n/d
XXII-N74	4	P 21/n	1.725	4.6	6.3	8.6	n/d
XXII-N57	4	P n m a	1.696	6.3	5.8	8.7	n/d
XXII-N61	4	P 21/c	1.717	6.8	6.3	8.7	n/d
XXII-N59	4	P 21/n	1.695	7.2	7.2	8.9	n/d
XXII-N69	2	P -1	1.752	4.9	7.7	9.0	n/d
XXII-N78	2	P 21	1.716	4.8	7.3	9.1	n/d
XXII-N72	4	P 21/c	1.723	5.0	7.5	9.2	n/d
XXII-N100	8	I 2/c	1.711	5.9	7.0	9.2	n/d
XXII-N51	4	P 21/c	1.689	7.6	6.6	9.4	n/d

Table S4. (continued) Stability ranking for system XXII.

Name	Z	Symm.	Density	PBE+TS	PBE+MBD	PBE0+MBD	PBE0+MBD+F_{vib}
XXII-N79	4	P 21/c	1.710	6.4	8.1	9.5	n/d
XXII-N87	4	P n a 21	1.718	6.1	7.9	9.6	n/d
XXII-N54	4	P 21/c	1.702	6.2	6.7	9.7	n/d
XXII-N89	2	P -1	1.662	7.3	6.7	9.7	n/d
XXII-N90	4	P 21/c	1.671	8.1	6.9	9.7	n/d
XXII-N81	4	P 21/c	1.691	7.7	7.5	10.0	n/d
XXII-N99	4	P -1	1.691	7.4	7.4	10.0	n/d
XXII-N75	8	P b c a	1.703	6.6	7.3	10.1	n/d
XXII-N80	4	P 21/c	1.705	6.9	7.3	10.1	n/d
XXII-N91	2	P -1	1.710	7.6	7.5	10.2	n/d
XXII-N95	4	P 21/c	1.687	7.4	7.4	10.2	n/d
XXII-N68	4	P 21/n	1.719	5.8	7.6	10.3	n/d
XXII-N96	4	P 21 21 21	1.639	9.0	6.7	10.6	n/d
XXII-N94	4	P 21/c	1.671	8.9	7.5	10.7	n/d
XXII-N98	4	P 21/c	1.665	8.6	7.2	10.7	n/d

Table S5. Stability ranking for system XXIII in kilojoules per mole per molecule. The energy of the most stable structure in each ranking was set to zero and the structures are always ordered according to the highest available ranking level. The final ranking includes vibrational free energies calculated at 300 K. In addition, this table contains the number of molecules in the asymmetric unit (Z'), the number of molecules in the unit cell (Z), the space group (Symm.), and the density in g/cm^3.

Name	Z'	Z	Symm.	Density	PBE+TS	PBE+MBD	PBE0+MBD	PBE0+MBD+F_{vib}
XXIII-N70	1	4	P 21/n	1.388	11.1	3.4	4.0	0.0
XXIII-N5	2	4	P -1	1.440	6.9	0.7	0.1	1.4
XXIII-E (Form E)	2	4	P -1	1.418	9.4	2.7	3.2	1.4
XXIII-N6 (Form C)	2	4	P -1	1.440	6.9	0.7	0.1	1.5
XXIII-N31	1	2	P -1	1.424	6.4	2.7	3.9	2.1
XXIII-N18	1	2	P -1	1.454	6.7	0.5	0.0	2.2
XXIII-N2	2	4	P -1	1.434	4.4	0.0	2.1	2.2
XXIII-N4 (Form B)	1	2	P -1	1.445	4.6	2.0	1.5	2.5
XXIII-N42	1	2	P -1	1.433	6.0	1.3	1.8	3.0
XXIII-N22	2	4	P -1	1.454	4.6	2.9	3.1	3.5
XXIII-N1	2	8	P 21/n	1.425	11.3	2.6	4.4	3.5
XXIII-N13	2	8	P 21/n	1.415	12.8	3.2	4.7	3.6
XXIII-N100	1	4	P 21/n	1.449	10.5	4.0	3.9	3.7
XXIII-N7	2	4	P -1	1.436	6.4	1.9	2.5	3.8
XXIII-N68	1	4	P 21/n	1.423	7.2	2.0	2.6	3.9
XXIII-N85 (Form A)	1	4	P 21/c	1.394	9.0	4.6	5.3	4.1
XXIII-N3	1	8	I 2/a	1.440	1.3	1.5	2.7	4.1
XXIII-N39 (Form D)	1	4	P 21/n	1.384	11.1	4.9	6.7	4.3
XXIII-N53	1	2	P -1	1.415	7.5	4.5	5.1	4.4
XXIII-N28	1	2	P -1	1.430	1.8	2.5	3.5	4.4
XXIII-N33	2	4	P -1	1.433	7.8	4.7	4.5	4.5
XXIII-N10	2	8	P 21/n	1.431	9.6	2.3	3.8	4.6
XXIII-N26	2	8	P 21/c	1.427	8.0	3.7	4.0	4.8
XXIII-N8	2	8	P 21/n	1.425	10.2	2.4	4.2	5.0
XXIII-N46	2	4	P -1	1.440	6.6	3.9	4.7	5.0
XXIII-N38	1	4	P 21/c	1.390	11.6	2.0	4.7	5.1
XXIII-N30	2	4	P -1	1.442	9.2	3.3	3.7	5.2
XXIII-N25	2	4	P -1	1.442	8.9	3.2	3.6	5.3
XXIII-N20	2	8	P 21/c	1.437	1.7	2.8	4.3	5.5
XXIII-N52	2	4	P -1	1.449	7.8	5.0	5.0	5.6
XXIII-N80	1	4	P 21/c	1.411	11.0	5.3	5.2	5.6
XXIII-N23	2	8	P n a 21	1.438	1.8	3.0	4.6	5.8
XXIII-N24	2	8	P 21/c	1.424	6.4	2.4	4.8	6.1
XXIII-N95	1	2	P -1	1.404	10.4	6.3	7.1	7.0
XXIII-N37	1	4	P 21/c	1.459	2.0	3.0	6.6	7.6
XXIII-N41	1	4	P 21/c	1.425	8.8	4.5	7.2	8.0
XXIII-N56	1	2	P -1	1.448	7.7	4.3	5.1	9.0

Table S5. (continued) Stability ranking for system XXIII.

Name	Z'	Z	Symm.	Density	PBE+TS	PBE+MBD	PBE0+MBD	PBE0+MBD+F_{vib}
XXIII-N49	1	2	P -1	1.414	10.0	3.7	5.8	9.2
XXIII-N91	1	2	P -1	1.414	8.7	6.0	7.2	9.2
XXIII-N43	1	4	P 21/n	1.451	8.6	5.8	6.4	9.3
XXIII-N66	1	2	P -1	1.465	0.0	3.8	7.2	9.8
XXIII-N62	1	4	P 21/c	1.406	8.8	5.0	7.2	10.3
XXIII-N88	1	4	P 21/c	1.440	6.4	6.7	8.7	10.9
XXIII-N48	1	4	P 21/c	1.411	8.1	5.8	8.0	11.4
XXIII-N61	1	4	P 21/c	1.459	7.7	5.1	7.5	11.9
XXIII-N81	1	4	P 21/c	1.443	5.9	5.0	8.4	12.5
XXIII-N35	2	16	C 2/c	1.459	6.5	4.5	4.5	n/d
XXIII-N76	2	16	C 2/c	1.421	3.0	3.4	4.8	n/d
XXIII-N47	2	4	P -1	1.445	7.9	5.0	5.0	n/d
XXIII-N21	2	8	P 21/n	1.416	13.0	3.8	5.0	n/d
XXIII-N83	2	4	P 21	1.425	6.1	5.3	5.1	n/d
XXIII-N40	2	4	P -1	1.431	8.6	4.2	5.2	n/d
XXIII-N60	2	4	P -1	1.446	8.0	5.2	5.2	n/d
XXIII-N59	2	4	P -1	1.447	7.9	5.2	5.2	n/d
XXIII-N27	2	16	C 2/c	1.435	3.0	3.3	5.2	n/d
XXIII-N57	2	4	P -1	1.447	7.8	5.1	5.2	n/d
XXIII-N54	2	16	C 2/c	1.450	7.0	5.0	5.3	n/d
XXIII-N63	2	4	P -1	1.433	8.7	5.4	5.4	n/d
XXIII-N36	2	4	P -1	1.428	9.4	5.2	5.4	n/d
XXIII-N32	2	8	P 21/c	1.440	10.0	4.2	5.6	n/d
XXIII-N64	2	8	P 21/c	1.437	4.0	4.6	5.6	n/d
XXIII-N29	2	8	P 21/c	1.429	12.0	4.3	5.9	n/d
XXIII-N71	2	4	P -1	1.447	7.9	4.5	5.9	n/d
XXIII-N73	2	16	I 2/c	1.445	7.4	5.5	6.0	n/d
XXIII-N97	2	4	P -1	1.410	10.3	5.7	6.1	n/d
XXIII-N67	2	4	P -1	1.420	9.0	5.8	6.2	n/d
XXIII-N90	2	4	P -1	1.437	7.5	5.7	6.2	n/d
XXIII-N77	2	4	P -1	1.452	7.4	4.5	6.3	n/d
XXIII-N45	2	16	I 2/c	1.424	5.6	4.3	6.3	n/d
XXIII-N65	2	4	P -1	1.422	8.9	5.8	6.3	n/d
XXIII-N86	2	8	C c	1.432	7.6	5.3	6.3	n/d
XXIII-N89	2	4	P -1	1.450	8.7	4.9	6.4	n/d
XXIII-N87	4	16	P 2/c	1.449	8.3	6.3	6.5	n/d
XXIII-N84	2	8	P 21/c	1.452	2.0	3.9	6.5	n/d
XXIII-N82	2	8	C c	1.424	7.3	5.4	6.6	n/d
XXIII-N44	2	4	P -1	1.431	11.0	4.8	6.6	n/d
XXIII-N55	2	8	P 21/c	1.409	6.7	5.0	6.6	n/d
XXIII-N34	2	16	I 2/c	1.420	4.6	4.3	6.7	n/d
XXIII-N69	2	4	P -1	1.436	8.9	5.2	7.0	n/d

Table S5. (continued) Stability ranking for system XXIII.

Name	Z'	Z	Symm.	Density	PBE+TS	PBE+MBD	PBE0+MBD	PBE0+MBD+F_{vib}
XXIII-N96	2	4	P -1	1.446	9.2	5.0	7.1	n/d
XXIII-N98	2	4	P -1	1.432	9.2	5.5	7.2	n/d
XXIII-N58	2	8	P 21/c	1.403	11.9	5.0	7.3	n/d
XXIII-N93	2	4	P -1	1.445	8.0	5.8	7.3	n/d
XXIII-N51	2	4	P -1	1.422	13.8	5.9	7.5	n/d
XXIII-N99	2	16	I 2/a	1.440	4.4	6.0	7.6	n/d
XXIII-N72	2	4	P -1	1.419	11.8	5.6	7.9	n/d
XXIII-N75	2	4	P -1	1.419	12.0	5.8	7.9	n/d
XXIII-N79	2	8	P 21/n	1.410	14.9	6.5	8.3	n/d
XXIII-N74	2	4	P -1	1.438	12.0	6.7	8.4	n/d
XXIII-N94	2	4	P -1	1.442	10.2	7.0	8.5	n/d

Table S6. Stability ranking for system XXIV in kilojoules per mole per trimer. The energy of the most stable structure in each ranking was set to zero and the structures are always ordered according to the highest available ranking level. The final ranking includes vibrational free energies calculated at 240 K. In addition, this table contains the number of molecules in the unit cell (Z), the space group (Symm.), and the density in g/cm^3.

Name	Z	Symm.	Density	PBE+TS	PBE+MBD	PBE0+MBD	PBE0+MBD+F_{vib}
XXIV-N2 (Exp.)	4	P 21/c	1.570	0.3	1.5	0.4	0.0
XXIV-N3	4	P 21/c	1.543	1.2	0.5	0.0	0.3
XXIV-N10	8	C 2/c	1.521	4.1	2.0	2.4	1.1
XXIV-N53	2	P -1	1.566	0.0	0.8	0.8	1.3
XXIV-N7	4	P 21/n	1.549	2.3	1.4	2.7	1.4
XXIV-N4	4	P 21/c	1.551	1.4	2.1	1.3	1.5
XXIV-N6	2	P -1	1.535	2.9	1.7	2.6	1.9
XXIV-N1	4	P 21 21 21	1.534	1.2	0.0	2.6	3.1
XXIV-N8	4	P 21/n	1.561	3.4	3.6	3.3	n/d
XXIV-N9	8	C 2/c	1.571	2.8	3.9	3.5	n/d
XXIV-N19	2	P -1	1.556	4.6	4.2	3.6	n/d
XXIV-N12	2	P -1	1.541	3.9	3.1	4.2	n/d
XXIV-N16	4	P 21/c	1.502	5.0	3.0	5.2	n/d
XXIV-N21	8	I 2/a	1.535	4.6	4.7	5.2	n/d
XXIV-N18	8	P c c n	1.507	4.9	4.1	5.5	n/d
XXIV-N33	4	P 21/c	1.571	5.6	5.7	5.7	n/d
XXIV-N22	8	I 2/c	1.505	5.8	4.8	5.8	n/d
XXIV-N43	4	P 21/c	1.580	7.4	6.8	6.2	n/d
XXIV-N11	4	P 21/c	1.479	6.0	3.3	6.2	n/d
XXIV-N23	2	P -1	1.556	5.4	5.2	6.3	n/d
XXIV-N40	8	P b c n	1.506	7.1	6.0	6.6	n/d
XXIV-N30	4	P 21/n	1.527	7.2	5.5	6.6	n/d
XXIV-N31	8	I b a 2	1.505	6.7	5.6	6.8	n/d
XXIV-N29	2	P -1	1.506	9.0	6.3	6.8	n/d
XXIV-N27	4	P 21/n	1.572	6.6	7.7	6.8	n/d
XXIV-N45	4	P 21/c	1.543	7.8	7.5	7.2	n/d
XXIV-N5	8	P b c a	1.522	4.6	4.3	7.2	n/d
XXIV-N63	4	P 21/c	1.527	9.8	8.1	7.4	n/d
XXIV-N14	4	P 21 21 21	1.523	7.2	4.7	7.4	n/d
XXIV-N13	4	P 21 21 21	1.518	6.8	4.2	7.4	n/d
XXIV-N44	8	C 2/c	1.549	8.0	7.5	7.5	n/d
XXIV-N50	8	P b c n	1.524	7.5	7.2	7.7	n/d
XXIV-N32	8	C 2/c	1.428	8.4	4.9	7.7	n/d
XXIV-N34	4	P 21/c	1.456	8.0	5.3	7.9	n/d
XXIV-N55	4	P 21 21 21	1.548	9.1	8.9	7.9	n/d
XXIV-N17	8	P b c n	1.602	6.5	6.6	8.1	n/d
XXIV-N24	4	P 21/n	1.536	7.9	6.7	8.2	n/d

Table S6. (continued) Stability ranking for system XXIV.

Name	Z	Symm.	Density	PBE+TS	PBE+MBD	PBE0+MBD	PBE0+MBD+F_{vib}
XXIV-N58	4	P 21/n	1.538	6.6	7.7	8.2	n/d
XXIV-N78	2	P -1	1.549	8.3	8.9	8.3	n/d
XXIV-N20	4	P n a 21	1.500	7.7	4.6	8.3	n/d
XXIV-N65	4	P 21/n	1.484	11.9	8.0	8.3	n/d
XXIV-N52	4	P 21/c	1.461	10.5	7.5	8.4	n/d
XXIV-N49	8	C 2/c	1.559	7.0	8.3	8.5	n/d
XXIV-N67	4	P -1	1.483	7.7	5.4	8.6	n/d
XXIV-N36	8	P c c n	1.513	8.3	7.2	8.7	n/d
XXIV-N62	8	P c c n	1.419	11.5	6.9	8.7	n/d
XXIV-N39	2	P -1	1.502	7.7	6.0	8.7	n/d
XXIV-N73	4	P 21/c	1.532	9.2	8.2	8.9	n/d
XXIV-N42	2	P -1	1.513	9.5	8.3	9.2	n/d
XXIV-N35	2	P -1	1.473	8.1	6.6	9.2	n/d
XXIV-N54	4	P 21/n	1.463	10.3	6.6	9.2	n/d
XXIV-N37	4	P 21/c	1.479	7.9	5.8	9.3	n/d
XXIV-N47	4	P 21/c	1.466	9.5	6.2	9.5	n/d
XXIV-N25	4	P 21 21 21	1.545	8.5	7.1	10.0	n/d
XXIV-N51	16	F d d 2	1.563	9.5	8.8	10.1	n/d
XXIV-N92	8	C c	1.537	9.3	7.5	10.3	n/d
XXIV-N85	4	P 21 21 21	1.550	9.9	9.9	10.3	n/d
XXIV-N64	4	P 21/c	1.519	11.6	9.3	10.3	n/d
XXIV-N60	2	P -1	1.449	11.7	8.1	10.5	n/d
XXIV-N70	16	F d d 2	1.479	10.2	9.2	10.6	n/d
XXIV-N66	2	P 21	1.555	9.0	9.2	10.8	n/d
XXIV-N90	4	P c a 21	1.472	11.4	8.1	10.8	n/d
XXIV-N94	8	P c c n	1.527	10.6	9.5	11.1	n/d
XXIV-N88	16	F d d 2	1.595	10.0	11.2	11.1	n/d
XXIV-N87	2	P -1	1.473	10.1	8.5	11.1	n/d
XXIV-N48	4	P 21/n	1.417	12.5	7.2	11.2	n/d
XXIV-N84	8	I 2/c	1.514	12.7	9.1	11.2	n/d
XXIV-N91	4	P 21/c	1.427	12.7	8.6	11.4	n/d
XXIV-N59	4	P 21 21 21	1.481	11.5	7.0	11.4	n/d
XXIV-N74	4	P 21/c	1.490	10.6	8.1	11.5	n/d
XXIV-N81	4	P 21/c	1.504	13.0	9.9	11.5	n/d
XXIV-N89	4	P 21/n	1.491	11.2	9.5	11.5	n/d
XXIV-N99	8	P c c n	1.522	10.0	10.2	11.7	n/d
XXIV-N97	4	P 21/c	1.551	11.9	11.0	11.9	n/d
XXIV-N77	4	P 21/c	1.498	11.3	9.7	11.9	n/d
XXIV-N61	2	P -1	1.494	10.1	8.6	12.0	n/d
XXIV-N75	4	P 21/c	1.508	11.7	9.6	12.1	n/d
XXIV-N95	4	P 21/c	1.486	10.7	9.6	12.1	n/d
XXIV-N93	4	P 21/n	1.532	12.9	12.1	12.2	n/d

Table S6. (continued) Stability ranking for system XXIV.

Name	Z	Symm.	Density	PBE+TS	PBE+MBD	PBE0+MBD	PBE0+MBD+F_{vib}
XXIV-N69	4	P n a 21	1.480	13.0	8.7	12.4	n/d
XXIV-N100	4	P 21/n	1.548	13.7	13.0	12.4	n/d
XXIV-N82	4	P 21 21 21	1.508	12.4	9.7	12.7	n/d
XXIV-N96	4	P 21 21 21	1.473	11.8	9.4	12.7	n/d
XXIV-N68	4	P 21/n	1.501	11.6	8.9	12.7	n/d
XXIV-N86	16	F d d 2	1.531	11.9	10.2	12.8	n/d
XXIV-N76	2	P 21	1.415	13.9	8.7	12.8	n/d
XXIV-N71	4	P 21/c	1.344	13.9	7.4	13.0	n/d
XXIV-N80	8	I 2/c	1.446	11.2	8.2	13.1	n/d
XXIV-N83	4	P 21/c	1.469	13.2	9.7	13.7	n/d
XXIV-N79	4	P 21/n	1.461	12.2	9.4	14.4	n/d
XXIV-N38	4	P 21/c	1.419	12.6	7.8	14.9	n/d
XXIV-N46	4	P n a 21	1.417	13.1	8.3	15.2	n/d

Table S7. Stability ranking for system XXV in kilojoules per mole per dimer. The energy of the most stable structure in each ranking was set to zero and the structures are always ordered according to the highest available ranking level. The final ranking includes vibrational free energies calculated at 300 K. In addition, this table contains the number of molecules in the unit cell (Z), the space group (Symm.), and the density in g/cm^3.

Name	Z	Symm.	Density	PBE+TS	PBE+MBD	PBE0+MBD	PBE0+MBD+F_{vib}
XXV-N6 (Exp.)	4	P 21/c	1.445	0.5	2.3	0.0	0.0
XXV-N5	8	P 21/c	1.431	0.0	0.0	1.0	3.8
XXV-N11	4	P 2/c	1.411	11.9	3.6	2.5	5.7
XXV-N1	2	P -1	1.453	4.4	3.2	4.2	7.0
XXV-N12	8	P b c n	1.397	13.8	5.1	4.2	n/d
XXV-N15	8	P b c n	1.409	13.3	4.4	4.3	n/d
XXV-N18	4	P c a 21	1.393	14.4	5.3	4.6	n/d
XXV-N22	2	P -1	1.431	7.0	6.7	4.7	n/d
XXV-N3	2	P -1	1.444	5.5	4.3	4.8	n/d
XXV-N16	4	P 21/c	1.422	4.0	3.9	5.1	n/d
XXV-N4	2	P -1	1.417	6.7	5.4	5.7	n/d
XXV-N2	4	P 21/n	1.437	7.6	3.8	6.0	n/d
XXV-N33	8	P b c n	1.407	14.1	7.4	6.1	n/d
XXV-N10	4	P 21/c	1.399	12.5	5.8	6.3	n/d
XXV-N29	4	P -1	1.425	10.9	8.3	6.8	n/d
XXV-N28	2	P -1	1.435	7.6	8.3	7.1	n/d
XXV-N19	4	P 21/c	1.417	13.3	8.5	7.1	n/d
XXV-N7	4	P 21/n	1.430	9.2	5.4	7.1	n/d
XXV-N36	8	P b c a	1.387	17.1	8.6	7.6	n/d
XXV-N25	4	P 21/n	1.404	11.3	6.9	7.8	n/d
XXV-N13	4	P 21/n	1.413	10.8	5.7	7.9	n/d
XXV-N27	8	I 2/c	1.419	14.5	8.8	8.2	n/d
XXV-N48	4	P 21/n	1.409	11.7	7.9	8.6	n/d
XXV-N26	2	P -1	1.418	12.6	7.9	8.7	n/d
XXV-N23	4	P 21/c	1.433	10.7	8.4	9.0	n/d
XXV-N9	4	P 21/c	1.433	10.6	7.8	9.1	n/d
XXV-N42	4	P 21/n	1.424	12.6	9.6	9.2	n/d
XXV-N20	8	P b c n	1.404	12.9	8.6	9.3	n/d
XXV-N49	2	P -1	1.401	11.0	9.7	9.6	n/d
XXV-N39	16	F d d 2	1.392	15.4	9.6	9.9	n/d
XXV-N30	4	P 21/c	1.411	18.0	9.7	9.9	n/d
XXV-N38	4	P 21/c	1.370	17.2	9.0	9.9	n/d
XXV-N32	2	P -1	1.408	15.3	9.2	10.2	n/d
XXV-N17	16	F d d 2	1.391	14.6	7.8	10.6	n/d
XXV-N44	4	P 21/c	1.395	16.2	9.4	10.9	n/d
XXV-N65	4	P 21/n	1.424	17.3	9.9	n/d	n/d
XXV-N24	4	P 21/c	1.395	14.6	10.0	n/d	n/d

Table S7. (continued) Stability ranking for system XXV.

Name	Z	Symm.	Density	PBE+TS	PBE+MBD	PBE0+MBD	PBE0+MBD+F_{vib}
XXV-N66	4	P 21/c	1.396	16.3	10.7	n/d	n/d
XXV-N60	8	A b a 2	1.427	11.6	10.8	n/d	n/d
XXV-N50	4	P 21/n	1.428	14.7	10.8	n/d	n/d
XXV-N45	1	P 1	1.387	18.7	11.1	n/d	n/d
XXV-N47	2	P -1	1.423	13.8	11.6	n/d	n/d
XXV-N59	8	P b c a	1.422	17.4	11.9	n/d	n/d
XXV-N43	4	P 21/c	1.417	19.6	11.9	n/d	n/d
XXV-N100	4	P 21/c	1.397	18.5	11.9	n/d	n/d
XXV-N71	4	P 21/c	1.418	18.4	12.0	n/d	n/d
XXV-N70	4	C c	1.410	14.7	12.0	n/d	n/d
XXV-N77	4	P 21/c	1.383	22.4	12.2	n/d	n/d
XXV-N61	4	P 21/n	1.409	16.1	12.3	n/d	n/d
XXV-N62	4	P 21/c	1.409	18.1	12.4	n/d	n/d
XXV-N40	4	P n a 21	1.437	15.0	12.5	n/d	n/d
XXV-N79	2	P -1	1.424	19.1	12.6	n/d	n/d
XXV-N55	16	I 41/a	1.396	18.2	12.6	n/d	n/d
XXV-N80	16	F d d 2	1.383	17.3	12.7	n/d	n/d
XXV-N57	8	P b c a	1.409	17.8	12.8	n/d	n/d
XXV-N52	8	P b c a	1.419	18.4	12.9	n/d	n/d
XXV-N53	2	P -1	1.437	14.0	13.0	n/d	n/d
XXV-N68	2	P n	1.377	20.6	13.0	n/d	n/d
XXV-N83	4	P 21/c	1.378	21.5	13.0	n/d	n/d
XXV-N81	2	P 21	1.415	15.0	13.0	n/d	n/d
XXV-N41	4	P 21/c	1.381	16.9	13.2	n/d	n/d
XXV-N64	4	P 21/c	1.408	18.7	13.2	n/d	n/d
XXV-N86	4	P 21 21 21	1.401	17.8	13.3	n/d	n/d
XXV-N58	4	P 21/c	1.423	17.3	13.4	n/d	n/d
XXV-N69	2	P -1	1.399	20.6	13.7	n/d	n/d
XXV-N73	2	P 21	1.427	19.7	13.8	n/d	n/d
XXV-N87	2	P -1	1.425	18.2	13.9	n/d	n/d
XXV-N84	8	I 2/a	1.363	27.3	13.9	n/d	n/d
XXV-N72	8	I 2/c	1.407	18.3	13.9	n/d	n/d
XXV-N78	4	P 21/c	1.379	21.3	13.9	n/d	n/d
XXV-N46	4	P 21/n	1.423	15.8	14.1	n/d	n/d
XXV-N89	4	P 21/c	1.420	15.0	14.3	n/d	n/d
XXV-N56	8	I 2/c	1.379	21.1	14.4	n/d	n/d
XXV-N63	2	P -1	1.416	19.2	14.5	n/d	n/d
XXV-N82	2	P -1	1.408	21.2	14.5	n/d	n/d
XXV-N74	4	P 21/c	1.404	18.9	14.7	n/d	n/d
XXV-N75	4	P 21/n	1.398	19.1	15.1	n/d	n/d
XXV-N85	8	I 2/c	1.429	16.3	15.2	n/d	n/d
XXV-N90	4	P n a 21	1.412	19.1	15.6	n/d	n/d

Table S7. (continued) Stability ranking for system XXV.

Name	Z	Symm.	Density	PBE+TS	PBE+MBD	PBE0+MBD	PBE0+MBD+F_{vib}
XXV-N91	4	P 21/c	1.383	20.7	15.7	n/d	n/d
XXV-N54	8	P b c a	1.407	18.9	16.1	n/d	n/d
XXV-N92	8	C 2/c	1.383	22.1	16.1	n/d	n/d
XXV-N88	8	P b c a	1.410	21.3	16.2	n/d	n/d
XXV-N94	8	C 2/c	1.387	23.8	16.6	n/d	n/d
XXV-N97	4	P 21/n	1.360	25.7	16.8	n/d	n/d
XXV-N95	2	P -1	1.395	27.7	17.0	n/d	n/d
XXV-N93	8	I 2/a	1.401	21.4	17.1	n/d	n/d
XXV-N96	4	P 21/n	1.387	19.2	17.2	n/d	n/d
XXV-N99	2	P -1	1.387	21.5	17.4	n/d	n/d
XXV-N76	4	P c a 21	1.401	22.4	17.5	n/d	n/d
XXV-N98	2	P -1	1.426	20.4	20.5	n/d	n/d

Table S8. Stability ranking for system XXVI in kilojoules per mole per molecule. The energy of the most stable structure in each ranking was set to zero and the structures are always ordered according to the highest available ranking level. The final ranking includes vibrational free energies calculated at 300 K. In addition, this table contains the number of molecules in the unit cell (Z), the space group (Symm.), and the density in g/cm^3.

Name	Z	Symm.	Density	PBE+TS	PBE+MBD	PBE0+MBD	PBE0+MBD+F_{vib}
XXVI-N1 (Exp.)	2	P -1	1.380	6.1	0.0	0.0	0.0
XXVI-N5	8	P 21/c	1.392	4.7	1.7	1.6	0.2
XXVI-N4	2	P -1	1.377	7.2	1.4	1.1	0.3
XXVI-N9	4	P 21/n	1.410	0.0	0.6	1.2	2.0
XXVI-N6	8	P 21/c	1.407	1.6	2.0	2.5	3.0
XXVI-N3	4	P -1	1.391	6.3	2.6	2.7	3.3
XXVI-N25	2	P -1	1.388	8.0	3.4	2.6	3.4
XXVI-N11	4	P -1	1.403	2.0	2.5	2.7	3.6
XXVI-N17	4	P -1	1.419	4.5	3.7	3.3	n/d
XXVI-N10	4	P -1	1.394	4.6	3.2	3.6	n/d
XXVI-N15	4	P -1	1.417	4.5	3.8	3.7	n/d
XXVI-N99	8	P 21/c	1.390	6.5	3.6	3.8	n/d
XXVI-N23	4	P -1	1.422	4.1	3.9	4.0	n/d
XXVI-N12	4	P 21	1.402	3.1	3.2	4.1	n/d
XXVI-N13	4	P -1	1.381	8.1	3.6	4.4	n/d
XXVI-N8	4	P -1	1.390	7.7	3.7	4.5	n/d
XXVI-N22	4	P -1	1.434	4.1	4.3	4.6	n/d
XXVI-N34	4	P -1	1.381	9.9	4.1	5.0	n/d
XXVI-N19	4	P -1	1.400	7.3	4.1	5.3	n/d
XXVI-N24	8	C c	1.390	8.5	4.0	5.3	n/d
XXVI-N42	4	P -1	1.376	10.4	4.5	5.6	n/d
XXVI-N28	4	P -1	1.374	9.8	4.4	5.9	n/d
XXVI-N40	4	P -1	1.380	9.5	4.6	n/d	n/d
XXVI-N18	4	P -1	1.390	7.9	4.6	n/d	n/d
XXVI-N48	4	P -1	1.414	6.1	4.8	n/d	n/d
XXVI-N63	4	P -1	1.397	8.2	4.9	n/d	n/d
XXVI-N33	4	P -1	1.376	9.4	4.9	n/d	n/d
XXVI-N32	4	P -1	1.379	9.6	4.9	n/d	n/d
XXVI-N38	4	P -1	1.414	5.9	5.0	n/d	n/d
XXVI-N41	4	P -1	1.378	9.4	5.0	n/d	n/d
XXVI-N26	4	P -1	1.426	4.4	5.0	n/d	n/d
XXVI-N72	4	P -1	1.382	9.3	5.0	n/d	n/d
XXVI-N20	16	C 2/c	1.381	9.3	5.1	n/d	n/d
XXVI-N27	4	P -1	1.420	4.6	5.1	n/d	n/d
XXVI-N62	4	P -1	1.412	6.4	5.1	n/d	n/d
XXVI-N30	4	P -1	1.374	9.5	5.1	n/d	n/d
XXVI-N16	4	P -1	1.411	5.8	5.2	n/d	n/d

Table S8. (continued) Stability ranking for system XXVI.

Name	Z	Symm.	Density	PBE+TS	PBE+MBD	PBE0+MBD	PBE0+MBD+F_{vib}
XXVI-N60	4	P -1	1.413	5.9	5.2	n/d	n/d
XXVI-N45	2	P 1	1.429	5.4	5.2	n/d	n/d
XXVI-N47	4	P -1	1.422	5.5	5.2	n/d	n/d
XXVI-N65	4	P -1	1.383	9.6	5.2	n/d	n/d
XXVI-N52	4	P -1	1.419	5.7	5.3	n/d	n/d
XXVI-N67	4	P -1	1.377	10.1	5.3	n/d	n/d
XXVI-N58	4	P -1	1.380	9.6	5.3	n/d	n/d
XXVI-N14	4	P -1	1.385	9.0	5.3	n/d	n/d
XXVI-N54	4	P -1	1.375	9.9	5.4	n/d	n/d
XXVI-N51	4	P -1	1.414	6.3	5.5	n/d	n/d
XXVI-N84	4	P -1	1.412	7.8	5.5	n/d	n/d
XXVI-N61	4	P -1	1.428	5.8	5.5	n/d	n/d
XXVI-N78	4	P -1	1.393	9.0	5.6	n/d	n/d
XXVI-N39	4	P -1	1.410	6.9	5.6	n/d	n/d
XXVI-N31	4	P -1	1.395	8.4	5.6	n/d	n/d
XXVI-N64	4	P -1	1.431	5.4	5.7	n/d	n/d
XXVI-N44	4	P -1	1.378	10.1	5.7	n/d	n/d
XXVI-N85	4	P -1	1.409	7.9	5.7	n/d	n/d
XXVI-N74	2	P 1	1.374	10.5	5.8	n/d	n/d
XXVI-N75	4	P -1	1.375	10.7	5.8	n/d	n/d
XXVI-N35	16	C 2/c	1.375	10.8	5.8	n/d	n/d
XXVI-N50	4	P -1	1.418	6.4	5.8	n/d	n/d
XXVI-N49	4	P -1	1.409	7.6	5.9	n/d	n/d
XXVI-N82	4	P -1	1.376	10.4	5.9	n/d	n/d
XXVI-N83	4	P -1	1.412	7.2	5.9	n/d	n/d
XXVI-N68	4	P -1	1.420	5.9	5.9	n/d	n/d
XXVI-N55	2	P -1	1.416	7.7	5.9	n/d	n/d
XXVI-N71	4	P -1	1.413	7.0	5.9	n/d	n/d
XXVI-N96	4	P -1	1.416	6.9	5.9	n/d	n/d
XXVI-N56	2	P 1	1.414	6.5	6.0	n/d	n/d
XXVI-N100	4	P -1	1.403	8.3	6.0	n/d	n/d
XXVI-N87	4	P n	1.395	8.4	6.0	n/d	n/d
XXVI-N29	16	C 2/c	1.410	8.1	6.1	n/d	n/d
XXVI-N86	4	P -1	1.424	6.2	6.1	n/d	n/d
XXVI-N94	4	P -1	1.391	8.8	6.1	n/d	n/d
XXVI-N70	4	P -1	1.419	6.7	6.1	n/d	n/d
XXVI-N77	4	P -1	1.428	5.1	6.2	n/d	n/d
XXVI-N93	4	P -1	1.386	11.5	6.2	n/d	n/d
XXVI-N73	4	P 21/n	1.401	3.6	6.2	n/d	n/d
XXVI-N91	4	P 21	1.387	10.8	6.2	n/d	n/d
XXVI-N92	4	P -1	1.372	12.0	6.3	n/d	n/d
XXVI-N80	8	C c	1.391	10.0	6.3	n/d	n/d

Table S8. (continued) Stability ranking for system XXVI.

Name	Z	Symm.	Density	PBE+TS	PBE+MBD	PBE0+MBD	PBE0+MBD+F_{vib}
XXVI-N57	4	P -1	1.425	5.3	6.3	n/d	n/d
XXVI-N89	4	P -1	1.415	7.4	6.4	n/d	n/d
XXVI-N79	4	P -1	1.382	10.7	6.4	n/d	n/d
XXVI-N37	4	P 21	1.382	10.2	6.4	n/d	n/d
XXVI-N81	4	P -1	1.410	7.1	6.5	n/d	n/d
XXVI-N46	8	C 2/c	1.421	8.3	6.5	n/d	n/d
XXVI-N88	4	P -1	1.421	5.9	6.5	n/d	n/d
XXVI-N43	8	C 2/c	1.387	7.0	6.6	n/d	n/d
XXVI-N59	16	C 2/c	1.396	8.8	6.7	n/d	n/d
XXVI-N90	8	C c	1.392	8.5	6.7	n/d	n/d
XXVI-N98	4	P -1	1.405	7.7	6.8	n/d	n/d
XXVI-N97	4	P -1	1.406	7.9	6.9	n/d	n/d
XXVI-N53	8	C 2/c	1.394	9.5	7.1	n/d	n/d
XXVI-N95	4	P 21/c	1.420	8.7	7.6	n/d	n/d
XXVI-N69	2	P -1	1.446	7.1	8.6	n/d	n/d

Table S9. Errors of PBE + TS–optimized structures with regard to experimental structures. This table shows the relative error in % for cell lengths (a, b, c), angles (α, β, γ), cell volume, and density. In addition the $RMSD_{20}$ is shown in Å for the settings described in the Methods section.

Exp. str.	Calc. Str.	a	b	c	α	β	γ	Volume	Density	$RMSD_{20}$
XXII	XXII-N2	0.46	0.94	2.07		0.76		2.99	-2.90	0.103
XXIII-A	XXIII-N85	-1.25	-2.12	0.06		-1.42		-3.09	3.18	0.198
XXIII-B	XXIII-N4	1.13	-0.92	-3.95	4.28	0.26	-1.39	-4.53	4.71	0.277
XXIII-C	XXIII-N6	-1.43	-1.24	-0.97	2.47	2.01	0.11	-3.19	3.30	0.237
XXIII-D	XXIII-N39	-3.56	0.74	1.24		2.11		-3.51	3.67	0.447
XXIII-E	XXIII-E	-1.15	-3.63	-1.69	-3.05	0.93	-0.55	-4.73	4.96	0.479
XXIV	XXIV-N2	2.17	-0.99	-0.78		1.04		0.11	-0.06	0.119
XXV	XXV-N6	-0.06	-1.30	-1.61		0.72		-3.43	3.51	0.129
XXVI	XXVI-N1	-1.02	-0.67	-1.95	3.30	0.73	0.86	-2.47	2.53	0.268

Table S10. Errors of the thermally expanded structures (corresponding to 300 K within the QHA) with regard to experimental structures. This table shows the relative error in % for cell lengths (a, b, c), angles (α, β, γ), cell volume, and density. In addition the $RMSD_{20}$ is shown in Å for the settings described in the Methods section.

Exp. str.	Calc. Str.	a	b	c	α	β	γ	Volume	Density	$RMSD_{20}$
XXIII-A	XXIII-N85-qha	-0.46	-0.73	0.28		-1.48		-0.70	0.74	0.193
XXIII-B	XXIII-N4-qha	2.84	0.75	-3.72	3.97	1.56	-1.49	-0.76	0.72	0.293
XXIII-C	XXIII-N6-qha	-0.69	-0.75	-0.36	2.03	2.06	0.23	-1.37	1.43	0.244
XXIII-D	XXIII-N39-qha	-2.34	2.13	1.50		2.07		-0.66	0.67	0.408
XXIII-E	XXIII-E-qha	0.35	-2.42	-1.49	-3.16	0.56	-1.02	-1.70	1.70	0.479

Data S1. All PBE + TS–optimized structures (light settings) and the thermally expanded structures for system XXIII. The structure numbers correspond to the rank in the initial structures provided by Neumann *et al.* (*4*). The structure labels corresponding to the experimentally confirmed structures are given in Table S9. The thermally expanded structures corresponding to 300 K are labeled with *qha*.