Supporting information for

What is the actual supramolecular architecture of theophylline polymorphic modifications and co-crystals with water and iodine from energetic viewpoint?

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Table S1. Symmetry codes, bonding type, interaction energy of the basic building unit with neighbouring ones (E_{int}, kcal/mol) and the contribution of this energy to the total interaction energy (%) in the crystal of **1.**

Dimer	Symmetry operation	E_{int}, kcal/mol	Energy ratio (ER), %	Interaction
1-m1	-1/2+x,-y,1/2+z	-12.1	20.3	N1-H…N2 1.84 Å, 174°
1-m2	1/2+x,-y,-1/2+z	-12.1	20.3	N1-H…N2 1.84 Å, 174°
1-m3	-1+x,y,z	-8.1	13.5	stacking, C2…C5 3.42 Å
1-m4	1+x,y,z	-8.1	13.5	stacking, C2…C5 3.42 Å
1-m5	-1/2+x,-y,-1/2+z	-4.2	7.0	C1-H…C7(π) 3.09 Å, 133
1-m6	1/2+x,-y,1/2+z	-4.2	7.0	C1-H…C7(π) 3.09 Å, 133°
1-m7	-1/2+x,-1-y,-1/2+z	-2.0	3.3	C5-H…O2 2.70 Å, 155°
1-m8	1/2+x,-1-y,1/2+z	-2.0	3.3	C5-H…O2 2.70 Å, 155°
1-m9	x,y,-1+z	-1.9	3.2	C4-H…O1 2.47 Å, 149°
1-m10	x,y,1+z	-1.9	3.2	C4-H…O1 2.47 Å, 149°
1-m11	-1/2+x,-1-y,1/2+z	-1.6	2.7	H5B…H4A 2.48 Å
1-m12	1/2+x,-1-y,-1/2+z	-1.6	2.7	H5B…H4A 2.48 Å

Table S2. Symmetry codes, bonding type, interaction energy of the basic building unit with neighbouring ones (E_{int}, kcal/mol) and the contribution of this energy to the total interaction energy (%) in the crystal of **2**

Dimer	Symmetry operation	E_{int}, kcal/mol	Energy ratio (ER), %	Interaction
2-m1	-1/2+x,3/2-y,1+z	-10.8	18.2	N1-H…O2 1.79 Å, 153°
2-m2	1/2+x,3/2-y,-1+z	-10.8	18.2	N1-H…O2 1.79 Å, 153°
2-m3	x,y,-1+z	-8.0	13.5	stacking, C7…N4 3.40 Å
2-m4	x,y,1+z	-8.0	13.5	stacking, C7…N4 3.40 Å
2-m5	-1/2+x,3/2-y,z	-4.0	6.8	C4-H…O1 2.29 Å, 173°
2-m6	1/2+x,3/2-y,z	-4.0	6.8	C4-H…O1 2.29 Å, 173°
2-m7	2-x,1-y,-1/2+z	-3.3	5.6	C1-H…N2 2.28 Å, 151°
2-m8	2-x,1-y,1/2+z	-3.3	5.6	C1-H…N2 2.28 Å, 151°
2-m9	2-x,2-y,-1/2+z	-1.7	2.9	non-specific
2-m10	2-x,2-y,1/2+z	-1.7	2.9	non-specific
2-m11	3/2-x,-1/2+y,-1/2+z	-0.9	1.5	non-specific
2-m12	3/2-x,-1/2+y,1/2+z	-0.9	1.5	non-specific
2-m13	3/2-x,1/2+y,-1/2+z	-0.9	1.5	non-specific
2-m14	3/2-x,1/2+y,1/2+z	-0.9	1.5	non-specific

Table S3. Symmetry codes, bonding type, interaction energy of the basic building unit with neighbouring ones (E_{int}, kcal/mol) and the contribution of this energy to the total interaction energy (%) in the crystal of **3**

Dimer	Symmetry operation	E_{int}, kcal/mol	Energy ratio (ER), %	Interaction
3-m1	1/2-x,-1/2+y,1/2+z	-10.3	17.2	N1-H…N2 1.77 Å, 178°
3-m2	1/2-x,1/2+y,-1/2+z	-10.3	17.2	N1-H…N2 1.77 Å, 178°
3-m3	x,-1+y,z	-8.1	13.5	stacking, C1…N2 3.41 Å
3-m4	x,1+y,z	-8.1	13.5	stacking, C1…N2 3.41 Å
3-m5	1/2-x,-1/2+y,-1/2+z	-4.8	7.9	C1-H…O1 2.23 Å, 158°
3-m6	1/2-x,1/2+y,1/2+z	-4.8	7.9	C1-H…O1 2.23 Å, 158°
3-m7	1-x,2-y,-1/2+z	-2.0	3.2	C5-H…O2 2.59 Å, 133°
3-m8	1-x,2-y,1/2+z	-2.0	3.2	C5-H…O2 2.59 Å, 133°
3-m9	1-x,3-y,-1/2+z	-1.9	3.1	C5-H…O2 2.63 Å, 110°
3-m10	1-x,3-y,1/2+z	-1.9	3.1	C5-H…O2 2.63 Å, 110°
3-m11	x,y,-1+z	-1.7	2.8	non-specific
3-m12	x,y,1+z	-1.7	2.8	non-specific
3-m13	x,-1+y,1+z	-1.3	2.2	non-specific
3-m14	x,1+y,-1+z	-1.3	2.2	non-specific

Table S4. Symmetry codes, bonding type, interaction energy of the basic building unit with neighbouring ones (E_{int}, kcal/mol) and the contribution of this energy to the total interaction energy (%) in the crystal of **4.**

Dimer	Symmetry operation	E(int), kcal/mol	Energy ratio (ER), %	Interaction
Monomeric building unit (MBU)				
4-m1	-1-x,-1-y,-z	-20.3	32.3	N1-H…O1 1.77 Å, 178°
4-m2	-1+x, y, z	-8.1	12.9	C-H…π 2.74 Å, 141°
4-m3	1+x, y, z	-8.1	12.9	C-H…π 2.74 Å, 141°
4-m4	-x,-1-y,-z	-5.9	9.4	non-specific
4-m5	-1+x,-1/2-y,-1/2+z	-4.3	6.8	C1-H…O2 2.11 Å, 165°
4-m6	1+x,-1/2-y,1/2+z	-4.3	6.8	C1-H…O2 2.11 Å, 165°
4-m7	-x,-y,-z	-3.4	5.4	C-H…π 2.72 Å, 158°
4-m8	x,-1/2-y,-1/2+z	-1.8	2.9	non-specific
4-m9	x,-1/2-y,1/2+z	-1.8	2.9	non-specific
4-m10	1-x,-y,-z	-1.5	2.4	non-specific
4-m11	1-x,-1/2+y,1/2-z	-1.1	1.8	non-specific
4-m12	1-x,1/2+y,1/2-z	-1.1	1.8	non-specific
Dimeric building unit (DBU)				
4-d1	-1+x,y,z	-21.7	25.1	stacking, C6…C7 3.41 Å
4-d2	1+x,y,z	-21.7	25.1	stacking, C6…C7 3.41 Å
4-d3	-1+x,-3/2-y,-1/2+z	-4.5	5.3	C1-H…O2 2.11 Å, 165°
4-d4	-1+x,-1/2-y,-1/2+z	-4.5	5.3	C1-H…O2 2.11 Å, 165°
4-d5	1+x,-3/2-y,1/2+z	-4.5	5.3	C1-H…O2 2.11 Å, 165°
4-d6	1+x,-1/2-y,1/2+z	-4.5	5.3	C1-H…O2 2.11 Å, 165°
4-d7	-1+x,-1+y,z	-3.6	4.2	C4-H…N2 2.72 Å, 158°
4-d8	1+x,1+y,z	-3.6	4.2	C4-H…N2 2.72 Å, 158°
4-d9	x,-3/2-y,-1/2+z	-2.1	2.4	C5-H…N2 2.65 Å, 167°
4-d10	x,-3/2-y,1/2+z	-2.1	2.4	C5-H…N2 2.65 Å, 167°
4-d11	x,-1/2-y,-1/2+z	-2.1	2.4	C5-H…N2 2.65 Å, 167°
4-d12	x,-1/2-y,1/2+z	-2.1	2.4	C5-H…N2 2.65 Å, 167°
4-d13	-2+x,-3/2-y,-1/2+z	-1.6	1.8	non-specific
4-d14	-2+x,-1/2-y,-1/2+z	-1.6	1.8	non-specific
4-d15	2+x,-3/2-y,1/2+z	-1.6	1.8	non-specific
4-d16	2+x,-1/2-y,1/2+z	-1.6	1.8	non-specific
4-d17	-2+x,-1+y,z	-1.5	1.7	non-specific
4-d18	2+x,1+y,z	-1.5	1.7	non-specific

Table S5. Symmetry codes, bonding type, interaction energy of the basic building unit with neighbouring ones (E_{int}, kcal/mol) and the contribution of this energy to the total interaction energy (%) in the crystal of **5**

Dimer	Symmetry operation	E(int), kcal/mol	Energy ratio (ER),%	Interaction
Monomeric building unit (MBU)				
5-m1	-x,-1/2+y,1/2-z	-3.6	4.1	C-H…O 2.31 Å, 140°
5-m2	-1+x,1/2-y,-1/2+z	-2.1	2.4	non-specific
5-m3	-x,1/2+y,1/2-z	-3.6	4.1	C-H…O 2.31 Å, 140°
5-m4	-x,1-y,1-z	-14.2	16	stacking 3.39 Å
5-m5	-1+x,1+y,z	-3.5	3.9	C-H…O 2.61 Å, 114°
5-m6	1-x,-1/2+y,1/2-z	-3.2	3.6	non-specific
5-m7	1-x,-y,1-z	-10.5	11.8	stacking 3.47 Å
5-m8	1-x,-1/2+y,3/2-z	-1.6	1.8	non-specific
5-m9	x,1/2-y,-1/2+z	-4.5	5.1	C-H…N 2.2 Å, 164°
5-m10	1-x,1/2+y,1/2-z	-3.2	3.6	non-specific
5-m11	1-x,1-y,1-z	-21	23.6	stacking 3.29 Å
5-m12	x,1/2-y,1/2+z	-4.5	5.1	C-H…N 2.2 Å, 164°
5-m13	1-x,1/2+y,3/2-z	-1.6	1.8	non-specific
5-m14	x,3/2-y,-1/2+z	-0.9	1.1	non-specific
5-m15	x,3/2-y,1/2+z	-0.9	1.1	non-specific
5-m16	1+x,-1+y,z	-3.5	3.9	C-H…O 2.61 Å, 114°
5-m17	2-x,-y,1-z	-4.2	4.7	C-H…O 2.47 Å, 159°
5-m18	1+x,1/2-y,1/2+z	-2.1	2.4	non-specific
Dimeric building unit (DBU)				
5-d1	-1+x,y,z	-16.7	11.8	stacking 3.39 Å
5-d2	1+x,y,z	-16.7	11.8	stacking 3.39 Å
5-d3	x,-1+y,z	-11.2	7.9	stacking 3.47 Å
5-d4	x,1+y,z	-11.2	7.9	stacking 3.47 Å
5-d5	-1+x,1+y,z	-10.8	7.6	C-H…O 2.61 Å, 114°
5-d6	1+x,-1+y,z	-10.8	7.6	C-H…O 2.61 Å, 114°
5-d7	1-x,-1/2+y,1/2-z	-9.9	7	C-H…N 2.2 Å, 164°
5-d8	1-x,-1/2+y,3/2-z	-9.9	7	C-H…N 2.2 Å, 164°
5-d9	1-x,1/2+y,1/2-z	-9.9	7	C-H…N 2.2 Å, 164°
5-d10	1-x,1/2+y,3/2-z	-9.9	7	C-H…N 2.2 Å, 164°
5-d11	-x,-1/2+y,1/2-z	-6.1	4.3	C-H…O 2.31 Å, 140°
5-d12	-x,1/2+y,1/2-z	-6.1	4.3	C-H…O 2.31 Å, 140°
5-d13	2-x,-1/2+y,3/2-z	-6.1	4.3	C-H…O 2.31 Å, 140°
5-d14	2-x,1/2+y,3/2-z	-6.1	4.3	C-H…O 2.31 Å, 140°

Table S6. Symmetry codes, bonding type, interaction energy of the basic building unit with neighbouring ones (E_{int}, kcal/mol) and the contribution of this energy to the total interaction energy (%) in the crystal of **6.**

Dimer	Symmetry operation	E(int), kcal/mol	Energy ratio (ER),%	Interaction
6-d1	-1+x,y,z	-19	15.4	Stacking 3.35 Å
6-d2	1+x,y,z	-19	15.4	Stacking 3.35 Å
6-d3	x,y,z	-6.2	5	O-H…N 1.91 Å, 164°
6-d4	x,y,z	-6.2	5	O-H…N 1.94 Å, 164°
6-d5	1-x,-1/2+y,2-z	-4.2	3.4	C-H…O 2.64 Å, 126°
6-d6	1-x,1/2+y,2-z	-4.2	3.4	C-H…O 2.64 Å, 126°
6-d7	-x,-1/2+y,1-z	-3.9	3.2	C-H…O 2.64 Å, 126°
6-d8	-x,1/2+y,1-z	-3.9	3.2	C-H…O 2.64 Å, 126°
6-d9	-1-x,-1/2+y,1-z	-3.3	2.7	C-H…O 2.11 Å, 176°
6-d10	-1-x,1/2+y,1-z	-3.3	2.7	C-H…O 2.11 Å, 176°
6-d11	2-x,-1/2+y,2-z	-3.2	2.6	C-H…O 2.11 Å, 176°
6-d12	2-x,1/2+y,2-z	-3.2	2.6	C-H…O 2.11 Å, 176°
6-d13	-x,1/2+y,1-z	-1.1	0.9	C-H…O 2.50 Å, 154°
6-d14	1-x,-1/2+y,2-z	-1.0	0.8	C-H…O 2.50 Å, 154°

Table S7. Symmetry codes, bonding type, interaction energy of the basic building unit with neighbouring ones (Eint, kcal/mol) and the contribution of this energy to the total interaction energy (%) in the crystal of **7**.

Dimer	Symmetry operation	E(int), kcal/mol	Energy ratio (ER),%	Interaction
7-d1	-1-x,1-y,1-z	-13.4	9.9	π…π 3.21 Å
7-d2	1-x,1-y,1-z	-13.4	9.9	π…π 3.21 Å
7-d3	x,y,z	-9.9	7.3	I…N 2.89 Å, 177°
7-d4	x,1+y,z	-9.9	7.3	I…N 2.89 Å, 177°
7-d5	-x,-1/2+y,1/2-z	-5.8	4.2	C-H…N 2.72 Å, 166°
7-d6	-x,-1/2+y,3/2-z	-5.8	4.3	C-H…N 2.72 Å, 166°
7-d7	-x,1/2+y,3/2-z	-5.8	4.3	C-H…N 2.72 Å, 166°
7-d8	-x,1/2+y,1/2-z	-5.8	4.3	C-H…N 2.72 Å, 166°
7-d9	-x,1/2+y,1/2-z	-5.1	3.8	I…π 2.89 Å, 177°
7-d10	-x,1/2+y,3/2-z	-5.1	3.8	I…π 2.89 Å, 177°
7-d11	-1-x,1/2+y,3/2-z	-3.2	2.4	C-H…O 2.27 Å, 151°
7-d12	1-x,-1/2+y,1/2-z	-3.2	2.4	C1-H…O2 2.27 Å, 151°
7-d13	1-x,1/2+y,1/2-z	-3.2	2.4	C1-H…O2 2.27 Å, 151°
7-d14	-1-x,1/2+y,3/2-z	-1.6	1.2	non-specific
7-d15	1-x,1/2+y,1/2-z	-1.6	1.2	non-specific
7-d16	-1+x,1+y,z	-1.2	0.9	C4-H…I 3.20 Å, 128°
7-d17	1+x,y,z	-1.2	0.9	C4-H…I 3.20 Å, 128°