Computational Supporting information

Table 1. The crystal strucutures of 3-oxauracil[a] found within 7 kJ mol^{-1} of the global minimum in the lattice energy, generated by the MOLPAK/DMAREL search. The experimental crystal structure (ExptMinOpt) is also shown for comparison. The structures, denoted by the MOLPAK code, are available in shelx format from the authors.

Structure	Space	Lattice Energy	[b] Free energy		[c] Reduced Cell				Hydrogen Bonding		[d] Graph set		
	group	/kJ mol^{-1}	at 298 kJ mol^{-1}	Density/g cm^{-3}	a/Å	b/Å	c/Å	Angles/°	and motif		Level 1	Level 2	Level 3
AI41	$P2_1/c$	-99.736	-113.155	1.69	5.9906	7.0975	10.5621	α 98.343	N1-H1···O7	Dimer	R2,2(8)		
ExptMinopt	**$P2_1/c$**	**-99.693**	**-113.117**	**1.691**	**5.991**	**7.096**	**10.561**	**α 98.34**	**N1-H1···O7**	**Dimer**	**R2,2(8)**		
CC28	Pbca	-95.901	-110.112	1.704	6.5229	10.5147	12.849		N1-H1···O3	Jagged sheet	C1,1(4)	R2,2(8)	C2,2(8)
									N1-H1···O7				
AK7	$P2_1/c$	-95.444	-108.753	1.717	6.6109	7.7002	8.7658	α 101.348	N1-H1···O3	Jagged sheet	C1,1(4)	R2,2(8)	C2,2(8)
									N1-H1···O7				
AB99	P-1	-95.296	-109.029	1.712	6.0797	6.6072	6.7485	α 70.941	N1-H1···O8	Infinite ribbon of dimers	C1,1(6)	R2,2(8)	R4,4(20)
								β 67.674	N1-H1···O7				
								γ 62.908					
DD64	C2/c	-95.112	-108.591	1.652	5.8322	11.0235	14.4117	α 101.189	N1-H1···O7	Dimer	R2,2(8)		
AB24	P-1	-94.641	-108.469	1.649	4.1022	6.0574	9.7024	α 90.667	N1-H1···O7	Dimer	R2,2(8)		
								β 95.781					
								γ 108.099					
DE57	C2/c	-94.635	-108.271	1.632	4.1434	11.1736	19.8757	α 90.413	N1-H1···O7	Dimer	R2,2(8)		
CC73	Pbca	-94.465	-109.608	1.623	6.7515	7.0385	19.4769		N1-H1···O7	Ribbon	C1,1(4)		
DA50	Cc	-94.039	-105.526	1.624	6.2062	6.8664	10.8491	α 90.272	N1-H1···O8	Chain	C1,1(6)		
FA77	$P2_1/c$	-93.94	-109.840	1.675	6.0445	6.3893	12.9363	γ 116.179	N1-H1···O8	Infinite ribbon of dimers	C1,1(6)	R2,2(8)	R4,4(20)
									N1-H1···O7				
DE15	C2/c	-93.866	-107.687	1.622	6.4229	10.4228	13.8319	α 91.097	N1-H1···O3	Jagged sheet	C1,1(4)	R2,2(8)	C2,2(8)
									N1-H1···O7				
AS50	$Pna2_1$	-93.736	-108.796	1.671	5.2702	6.8377	12.4706		N1-H1···O8	Chain	C1,1(6)		
AK37	$P2_1/c$	-93.428	-107.728	1.736	3.6482	10.5014	12.1584	α 111.753	N1-H1···O8	Jagged sheet	C1,1(6)	R2,2(8)	C2,2(10)
									N1-H1···O7				
DE72	C2/c	-93.176	-106.436	1.684	6.5729	11.2661	12.9062	α 111.014	N1-H1···O8	Infinite ribbon of dimers	C1,1(6)	R2,2(8)	R4,4(20)
									N1-H1···O7				
AI49	$P2_1/c$	-93.123	-108.379	1.656	7.0279	7.4923	8.7912	β 101.619	N1-H1···O3	Chain	C1,1(4)		
AZ28	$P2_12_12_1$	-92.918	-107.594	1.646	4.7738	5.0962	18.7557		N1-H1···O7	Ribbon	C1,1(4)		
CB100	Pbca	-92.774	-107.544	1.625	6.5718	7.6646	18.3564		N1-H1···O7	Dimer	R2,2(8)		
AG6	Pc	-92.771	-108.412	1.672	3.7711	6.0927	9.999	α 102.075	N1-H1···O8	Chain	C1,1(6)		
CC104	Pbca	-92.728	-109.226	1.662	7.4677	10.1023	11.983		N1-H1···O3	Chain	C1,1(4)		
AF64	$P2_1$	-92.704	-106.751	1.665	4.7857	5.1496	9.1572	β 91.900	N1-H1···O7	Dimer	C1,1(4)		
DA38	Cc	-92.697	-104.727	0.829	7.4979	7.6717	8.0241	α 100.990	N1-H1···O3	Chain	C1,1(4)		
AQ3	$P2_12_12_1$	-92.574	-106.959	1.687	3.765	9.3707	12.6187		N1-H1···O8	Chain	C1,1(6)		
CB25	Pbca	-92.543	-107.544	1.659	7.3029	8.5272	14.5389		N1-H1···O3	Jagged sheet	C1,1(4)	R2,2(8)	C2,2(8)
									N1-H1···O7				
AM68	$P2_1/c$	-92.538	-106.787	1.642	5.093	7.9532	11.3078	β 93.218	N1-H1···O7	Dimer	R2,2(8)		
AM36	$P2_1/c$	-92.482	-107.646	1.623	4.0231	9.9288	11.6296	γ 95.201	N1-H1···O8	Chain	C1,1(6)		
AI46	$P2_1/c$	-92.46	-108.823	1.621	6.6323	6.9842	10.1214	β 98.720	N1-H1···O7	Ribbon	C1,1(4)		
AH13	$P2_1$	-92.419	-108.280	1.654	4.3576	5.2876	10.0234	γ 100.634	N1-H1···O3	Chain	C1,1(4)		
DE92	C2/c	-92.361	-106.009	1.732	3.6143	12.8024	19.0077	α 99.464	N1-H1···O8	Jagged sheet	C1,1(6)	R2,2(8)	C2,2(10)
									N1-H1···O7				
AI72	$P2_1/c$	-92.259	-107.411	1.68	3.7297	9.8077	12.4723	α 101.490	N1-H1···O7	Dimer	R2,2(8)		
AU81	$Pna2_1$	-92.223	-108.770	1.645	6.036	7.5053	10.0767		N1-H1···O3	Chain	C1,1(4)		

[a]All calculated structures are lattice energy minima calculated with the *ab initio* gas phase molecular and the intermolecular potential described in the text. The hypothetical structures are labelled according to the initial MOLPAK coordination geometry and order of density. [b]The Helmholtz free energy is estimated from the lattice energy, zero point intermolecular energy and temperature dependence of the rigid molecule internal energy and entropy, as derived from the k=0 second derivative properties. [c]The Niggli reduced cell parameters as calculated during the MOLPAK/DMAREL process are given for comparison. Only the reduced cell angles which are not 90 º are tabulated. All structures have one molecule in the asymmetric unit.. [d]Only the first three levels shown, calculated using RPluto.

Table 2. The crystal strucutures of 5-hydroxyuracil[a] found within 7 kJ mol^{-1} of the global minimum in the lattice energy, generated by the MOLPAK/DMAREL search. The experimental crystal structure (ExptMinOpt) is also shown for comparison. The structures, denoted by the MOLPAK code, are available in shelx format from the authors.

Structure	Space	Lattice Energy	[b]Free energy		[c]Reduced Cell				[d]Hydrogen Bonding		[e]Graph set		
	group	/kJ mol^{-1}	at 298 kJ mol^{-1}	Density/g cm^{-3}	a/Å	b/Å	c/Å	Angles/°	and motif		Level 1	Level 2	Level 3
AM64	$P2_1/c$	-120.427	-133.223	1.829	3.4572	10.5288	12.881	β 97.18	N1-H1···O9	Sheets	S1,1(5)	R2,2(10)	C1,1(5)
									O9-H9···O8				
									N3-H3···O7				
AM10	$P2_1/c$	-118.632	-133.026	1.831	3.3806	10.0209	13.9051	γ 99.41	N1-H1···O9	3D	S1,1(5)	R2,2(10)	R2,2(10)
									O9-H9···O8				
									N3-H3···O7				
ExptMinOpt	**P-1**	**-117.323**	**-127.364**	**1.831**	**6.151**	**6.371**	**7.232**	**α 66.78**	**O9-H9···O8**	**Sheets**	**S1,1(5)**	**R2,2(10)**	**R2,2(8)**
								β 80.05	**N3-H3···O7**				
								γ 63.12	**N1-H1···O7**				
AB90	P-1	-117.339	-127.092	1.831	6.1514	6.3701	7.2316	α 66.80	O9-H9···O8	Sheets	S1,1(5)	R2,2(10)	R2,2(8)
								β 80.07	N3-H3···O7				
								γ 63.13	N1-H1···O7				
AM39	$P2_1/c$	-116.382	-128.11	1.76	5.7353	7.8482	10.8837	γ 99.42	N1-H1···O9	Sheets	S1,1(5)	R2,2(10)	C1,1(5)
									O9-H9···O8				
									N3-H3···O7				
CD97	Pbca	-115.058	-129.455	1.721	6.057	10.8504	15.0413		N1-H1···O9	Sheets	S1,1(5)	R2,2(10)	C1,1(5)
									O9-H9···O8				
									N3-H3···O7				
CA76	P-1	-114.415	-125.184	1.775	5.1982	6.0124	7.8866	α 82.26	O9-H9···O8	Sheets	S1,1(5)	R2,2(10)	R2,2(8)
								β 89.59	N3-H3···O7				
								γ 78.98	N1-H1···O7				
FC6	$P2_1/c$	-113.734	-124.631	1.764	5.4293	7.2586	12.4002	β 99.20	O9-H9···O8	Sheets	S1,1(5)	R2,2(10)	C1,1(4)
									N3-H3···O7				
									N1-H1···O7				
AM98	$P2_1/c$	-113.344	-125.397	1.741	5.1852	7.8563	12.0026	β 91.774	N3-H3···O7	3D	S1,1(5)	C1,1(7)	C1,1(6)
									N1-H1···O8				
									O9-H9···O7				
AK1	$P2_1/c$	-113.301	-125.875	1.765	5.3006	7.5281	12.3354	α 107.72	O9-H9···O8	3D	S1,1(5)	C1,1(5)	C1,1(4)
									N3-H3···O7				
									N1-H1···O7				

[a]All calculated structures are lattice energy minima calculated with the *ab initio* gas phase molecular model of conformation A and the intermolecular potential described in the text. The hypothetical structures are labelled according to the initial MOLPAK coordination geometry and order of density. [b]The Helmholtz free energy is estimated from the lattice energy, zero point intermolecular energy and temperature dependence of the rigid molecule internal energy and entropy, as derived from the k=0 second derivative properties. [c]The Niggli reduced cell parameters as calculated during the MOLPAK/DMAREL process are given for comparison. Only the reduced cell angles which are not 90 º are tabulated. All structures have one molecule in the asymmetric unit. [d]Intermolecular hydrogen bonding only. [e]Only the first three levels shown, calculated using RPluto.