Electronic Supplementary Material (ESI) for Inorganic Chemistry Frontiers.

SUPPLEMENTARY INFORMATION

Thermodynamic properties of the uranyl carbonate minerals roubaultite, fontanite, widenmannite, grimselite, čejkaite and bayleyite

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Contents: Page

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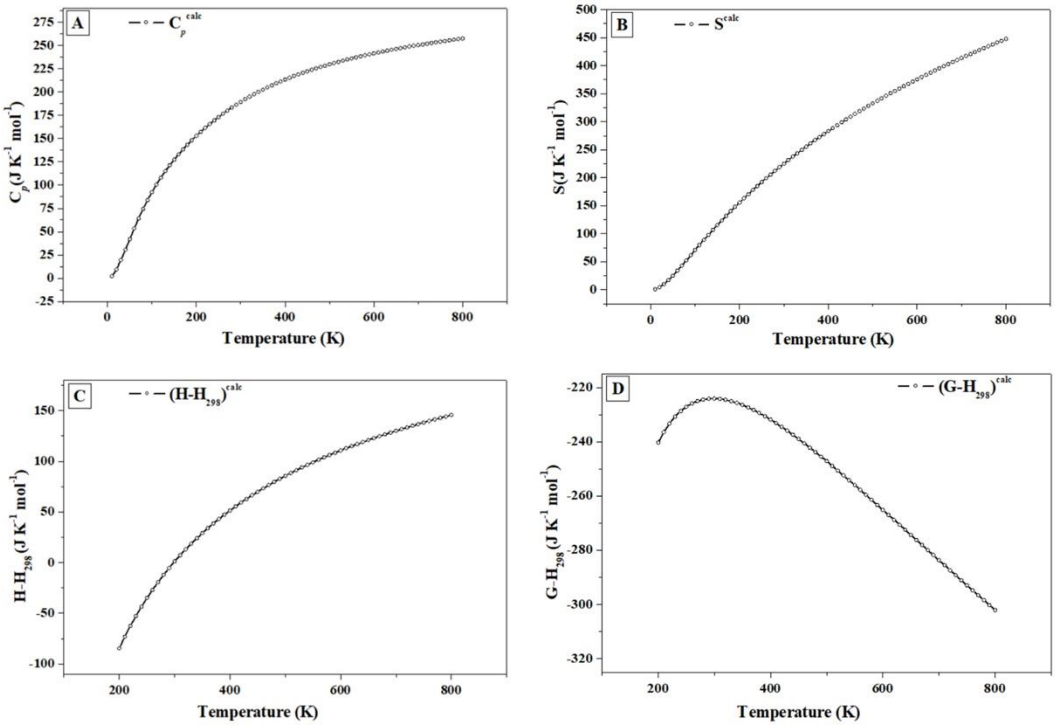

Fig. S.1. Calculated heat capacity, entropy, enthalpy and free energy functions of roubaultite.

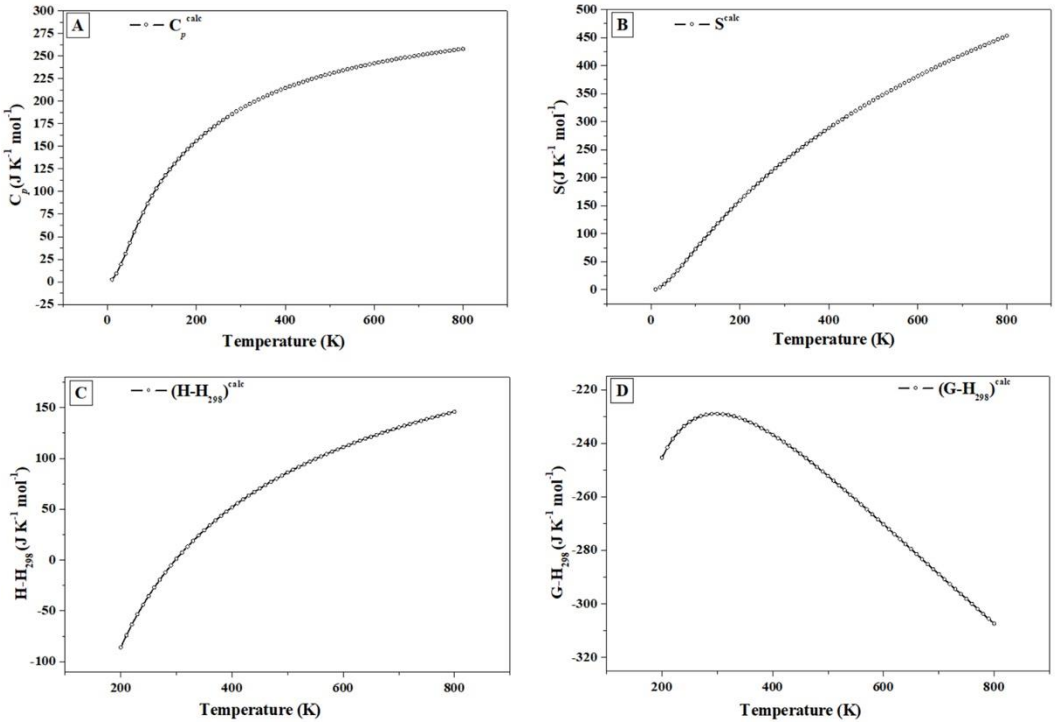

Fig. S.2. Calculated heat capacity, entropy, enthalpy and free energy functions of fontanite.

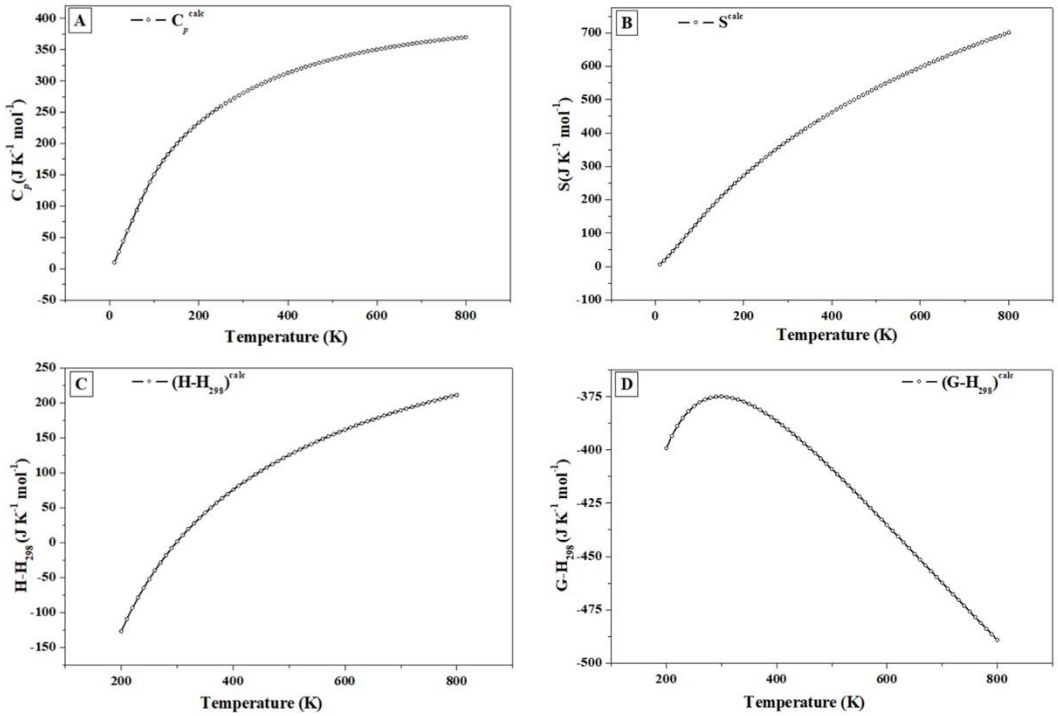

Fig. S.3. Calculated heat capacity, entropy, enthalpy and free energy functions of widenmannite.

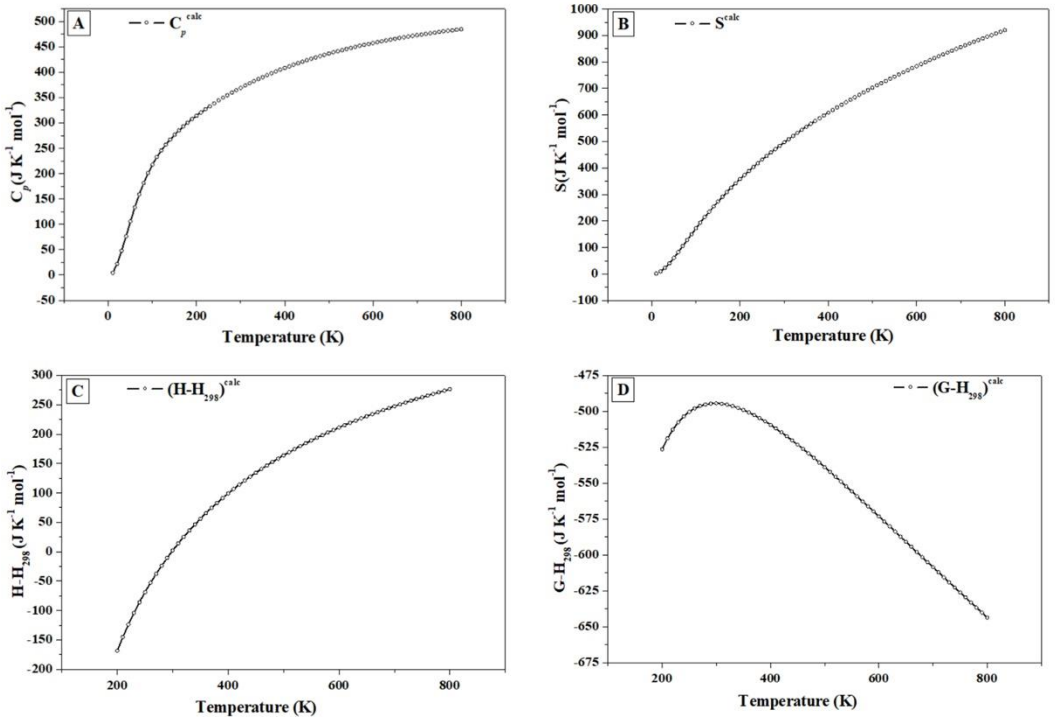

Fig. S.4. Calculated heat capacity, entropy, enthalpy and free energy functions of grimselite.

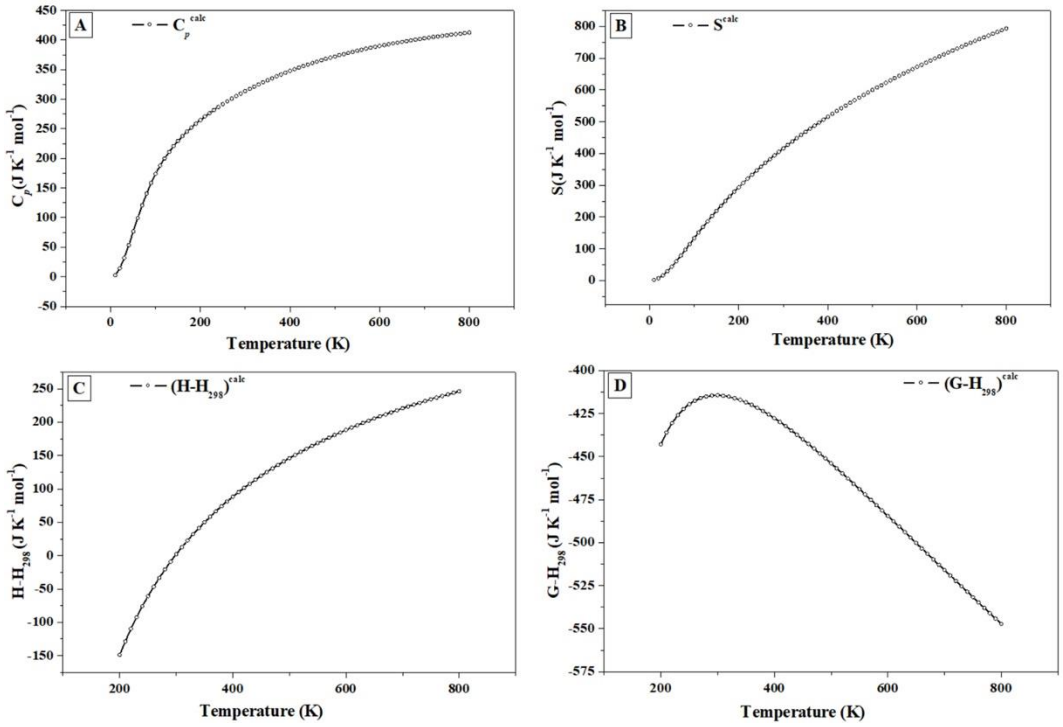

Fig. S.5. Calculated heat capacity, entropy, enthalpy and free energy functions of čejkaite.

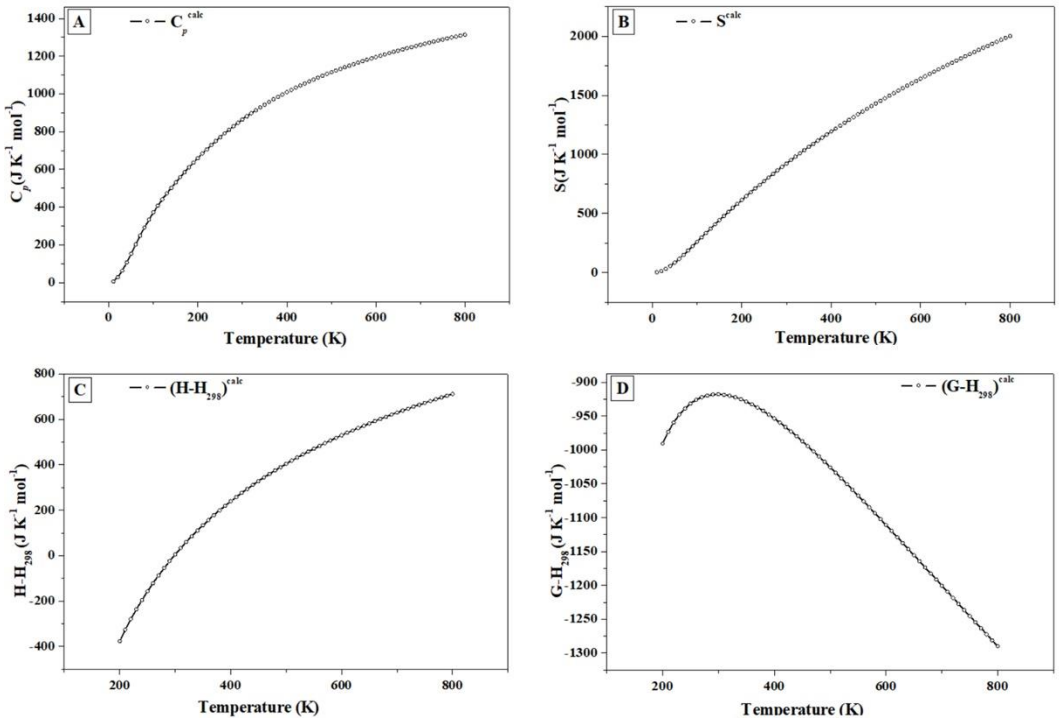

Fig. S.6. Calculated heat capacity, entropy, enthalpy and free energy functions of bayleyite.

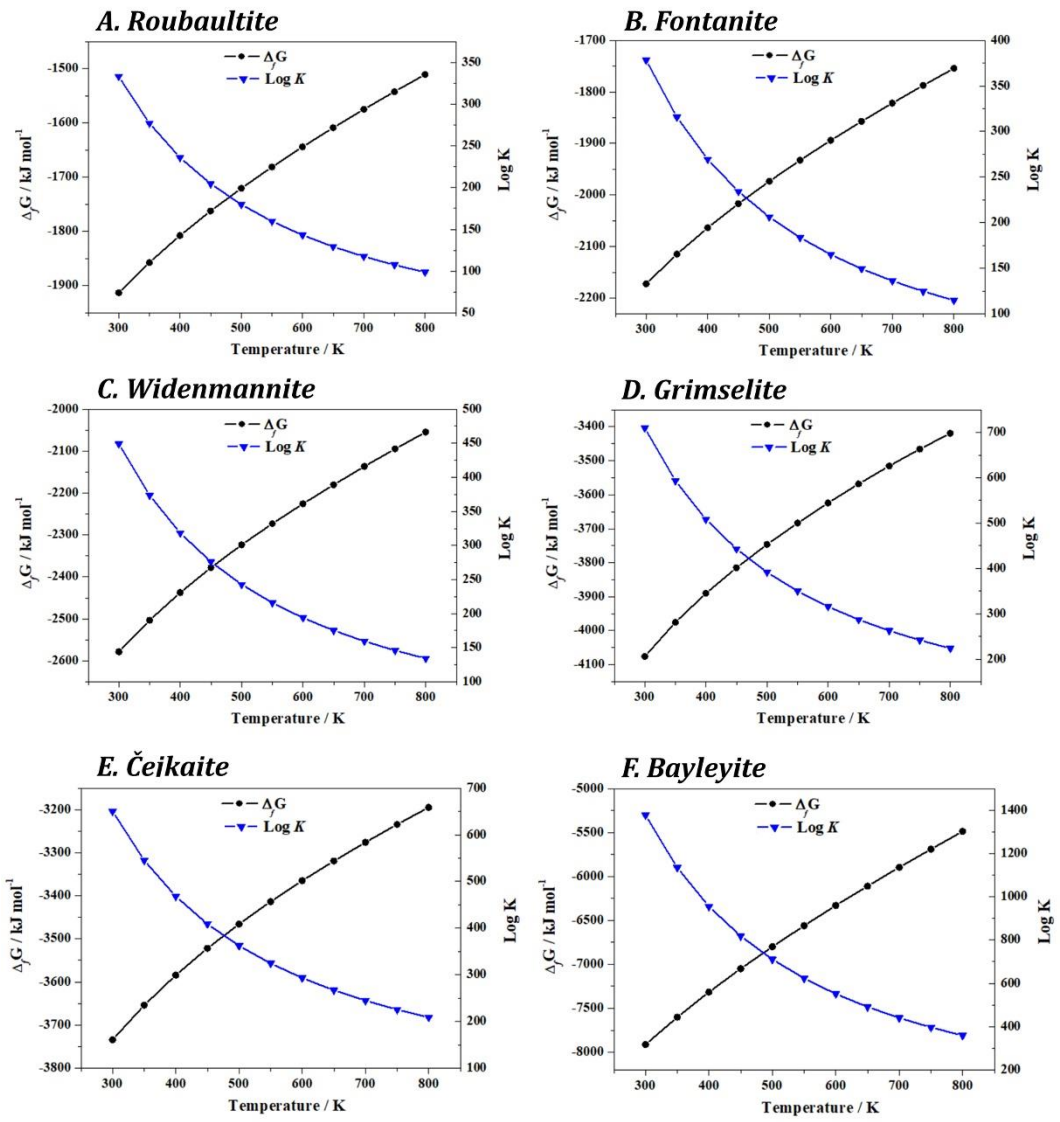

Fig. S.7. Computed Gibbs free energies of formation of roubaultite, fontanite, widenmannite, grimselite, čejkaite and bayleyite as a function of temperature and corresponding reaction constants.

Table S.1. Calculated enthalpies ($\Delta_r H$) and free-energies ($\Delta_r G$) of reaction and associated reaction constants (Log K) of reactions (I.A)-(I.F). The values of $\Delta_r H$ and $\Delta_r G$ are in units of $\mathrm{kJ \cdot mol^{-1}}$.

T(K)	$\Delta_r H$	$\Delta_r G$	Log K	$\Delta_r H$	$\Delta_r G$	Log K	$\Delta_r H$	$\Delta_r G$	Log K
	Reaction (I.A)			**Reaction (I.B)**			**Reaction (I.C)**		
298.15	-73.33	-26.70	4.68	-48.44	-13.06	2.29	13.46	11.30	-1.98
300	-72.97	-25.93	4.51	-48.14	-12.49	2.18	13.66	11.61	-2.02
320	-69.51	-17.97	2.93	-45.17	-6.69	1.09	15.46	14.65	-2.39
340	-66.68	-10.63	1.63	-42.71	-1.38	0.21	16.84	17.31	-2.66
360	-64.24	-3.63	0.53	-40.61	3.58	-0.52	18.03	19.83	-2.88
380	-62.61	2.59	-0.36	-39.00	8.07	-1.11	18.57	21.75	-2.99
400	-61.25	8.59	-1.12	-37.64	12.32	-1.61	18.99	23.61	-3.08
420	-60.27	14.24	-1.77	-36.57	16.30	-2.03	19.15	25.26	-3.14
440	-59.64	19.59	-2.33	-35.75	20.05	-2.38	19.06	26.73	-3.17
460	-59.35	24.67	-2.80	-35.17	23.58	-2.68	18.73	28.02	-3.18
480	-59.36	29.50	-3.21	-34.81	26.92	-2.93	18.17	29.18	-3.17
500	-59.68	34.11	-3.56	-34.65	30.09	-3.14	17.38	30.20	-3.15
T(K)	**Reaction (I.D)**			**Reaction (I.E)**			**Reaction (I.F)**		
298.15	-15.68	23.30	-4.08	1.63	-26.14	4.58	-134.42	-150.89	26.44
300	-15.51	23.77	-4.14	1.40	-26.50	4.61	-133.07	-150.88	26.27
320	-13.91	28.58	-4.66	-0.79	-29.95	4.89	-134.91	-150.67	24.59
340	-12.60	33.11	-5.09	-2.74	-33.13	5.09	-135.36	-150.43	23.11
360	-11.48	37.46	-5.44	-4.44	-36.04	5.23	-135.85	-150.19	21.79
380	-10.71	41.48	-5.70	-6.01	-38.80	5.33	-136.37	-149.98	20.62
400	-10.06	45.40	-5.93	-7.38	-41.34	5.40	-136.93	-149.81	19.56
420	-9.58	49.15	-6.11	-8.60	-43.71	5.44	-137.53	-149.67	18.61
440	-9.25	52.77	-6.26	-9.69	-45.93	5.45	-138.18	-149.55	17.75
460	-9.08	56.26	-6.39	-10.66	-48.00	5.45	-138.86	-149.47	16.97
480	-9.03	59.63	-6.49	-11.53	-49.94	5.43	-139.59	-149.40	16.26
500	-9.12	62.90	-6.57	-12.31	-51.77	5.41	-140.39	-149.34	15.60

Table S.2. Calculated enthalpies ($\Delta_r H$) and free-energies ($\Delta_r G$) of reaction and associated reaction constants (Log K) of reactions (II.A)-(II.F). The values of $\Delta_r H$ and $\Delta_r G$ are in units of $\mathrm{kJ \cdot mol^{-1}}$.

T(K)	$\Delta_r H$	$\Delta_r G$	Log K	$\Delta_r H$	$\Delta_r G$	Log K	$\Delta_r H$	$\Delta_r G$	Log K
	Reaction (II.A)			Reaction (II.B)			Reaction (II.C)		
298.15	-138.67	-91.22	15.98	-113.78	-77.58	13.59	-51.87	-53.22	9.32
300	-138.31	-90.43	15.75	-113.48	-77.00	13.41	-51.69	-52.90	9.21
320	-134.95	-82.35	13.44	-110.62	-71.07	11.60	-49.99	-49.73	8.12
340	-132.25	-74.88	11.50	-108.28	-65.63	10.08	-48.73	-46.94	7.21
360	-129.92	-67.73	9.83	-106.28	-60.51	8.78	-47.65	-44.26	6.42
380	-128.34	-61.29	8.42	-104.73	-55.81	7.67	-47.16	-42.12	5.79
400	-127.26	-55.30	7.22	-103.66	-51.57	6.73	-47.02	-40.28	5.26
420	-126.51	-49.58	6.17	-102.82	-47.52	5.91	-47.10	-38.56	4.80
440	-126.08	-44.12	5.24	-102.19	-43.66	5.18	-47.38	-36.98	4.39
460	-125.96	-38.89	4.42	-101.79	-39.98	4.54	-47.89	-35.54	4.04
480	-126.15	-33.90	3.69	-101.60	-36.48	3.97	-48.62	-34.23	3.72
500	-126.66	-29.12	3.04	-101.63	-33.15	3.46	-49.60	-33.04	3.45
T(K)	**Reaction (II.D)**			**Reaction (II.E)**			**Reaction (II.F)**		
298.15	-81.01	-41.22	7.22	83.31	57.85	-10.13	-34.98	-47.44	8.31
300	-80.85	-40.74	7.09	83.08	57.50	-10.01	-35.04	-47.40	8.25
320	-79.36	-35.80	5.84	80.99	54.12	-8.83	-35.66	-46.90	7.66
340	-78.18	-31.14	4.78	79.17	51.03	-7.84	-36.29	-46.36	7.12
360	-77.16	-26.63	3.86	77.59	48.19	-6.99	-36.95	-45.83	6.65
380	-76.44	-22.39	3.08	76.05	45.40	-6.24	-37.78	-45.45	6.25
400	-76.08	-18.49	2.41	74.95	43.08	-5.63	-38.41	-44.84	5.86
420	-75.83	-14.67	1.82	73.95	40.86	-5.08	-39.15	-44.34	5.51
440	-75.70	-10.94	1.30	73.04	38.75	-4.60	-39.99	-43.92	5.21
460	-75.69	-7.31	0.83	72.23	36.74	-4.17	-40.92	-43.55	4.94
480	-75.83	-3.77	0.41	71.51	34.85	-3.79	-41.91	-43.21	4.70
500	-76.10	-0.34	0.04	70.89	33.07	-3.45	-42.98	-42.90	4.48

Table S.3. Calculated enthalpies ($\Delta_r H$) and free-energies ($\Delta_r G$) of reaction and associated reaction constants (Log K) of reactions (III.A)-(III.F). The values of $\Delta_r H$ and $\Delta_r G$ are in units of $\mathrm{kJ \cdot mol^{-1}}$.

T(K)	$\Delta_r H$	$\Delta_r G$	Log K	$\Delta_r H$	$\Delta_r G$	Log K	$\Delta_r H$	$\Delta_r G$	Log K
	Reaction (III.A)			**Reaction (III.B)**			**Reaction (III.C)**		
298.15	-136.05	-32.28	5.66	-111.16	-18.64	3.27	-49.25	5.72	-1.00
300	-135.25	-30.76	5.36	-110.41	-17.33	3.02	-48.62	6.77	-1.18
320	-127.42	-15.16	2.47	-103.08	-3.88	0.63	-42.45	17.46	-2.85
340	-120.82	-0.79	0.12	-96.85	8.45	-1.30	-37.30	27.15	-4.17
360	-115.16	12.63	-1.83	-91.53	19.84	-2.88	-32.89	36.09	-5.24
380	-110.62	24.95	-3.43	-87.01	30.43	-4.18	-29.44	44.12	-6.06
400	-106.77	36.59	-4.78	-83.17	40.32	-5.27	-26.53	51.61	-6.74
420	-103.64	47.54	-5.91	-79.94	49.60	-6.17	-24.22	58.56	-7.28
440	-101.15	57.87	-6.87	-77.26	58.33	-6.92	-22.44	65.00	-7.72
460	-99.24	67.65	-7.68	-75.06	66.56	-7.56	-21.16	71.00	-8.06
480	-97.87	76.93	-8.37	-73.32	74.35	-8.09	-20.34	76.60	-8.34
500	-97.01	85.75	-8.96	-71.98	81.72	-8.54	-19.95	81.83	-8.55
T(K)	**Reaction (III.D)**			**Reaction (III.E)**			**Reaction (III.F)**		
298.15	-78.40	17.72	-3.10	31.66	-59.50	10.42	-135.65	-223.19	39.10
300	-77.79	18.93	-3.30	30.99	-60.64	10.56	-136.16	-224.00	39.00
320	-71.82	31.39	-5.12	24.44	-72.16	11.78	-141.23	-232.27	37.91
340	-66.74	42.94	-6.60	18.72	-82.78	12.72	-145.74	-239.89	36.85
360	-62.41	53.72	-7.79	13.79	-92.57	13.43	-149.79	-246.99	35.84
380	-58.72	63.85	-8.78	9.35	-101.82	14.00	-153.46	-253.66	34.87
400	-55.58	73.40	-9.58	5.51	-110.43	14.42	-156.80	-259.97	33.95
420	-52.95	82.45	-10.25	2.16	-118.51	14.74	-159.86	-265.97	33.08
440	-50.76	91.05	-10.81	-0.77	-126.13	14.97	-162.67	-271.68	32.25
460	-48.97	99.23	-11.27	-3.31	-133.33	15.14	-165.26	-277.15	31.47
480	-47.55	107.05	-11.65	-5.51	-140.15	15.25	-167.68	-282.38	30.73
500	-46.46	114.53	-11.96	-7.42	-146.61	15.32	-169.96	-287.40	30.02

Table S.4. Calculated enthalpies ($\Delta_r H$) and free-energies ($\Delta_r G$) of reaction and associated reaction constants (Log K) of reactions (IV.A)-(IV.F). The values of $\Delta_r H$ and $\Delta_r G$ are in units of $\mathrm{kJ \cdot mol^{-1}}$.

T(K)	$\Delta_r H$	$\Delta_r G$	Log K	$\Delta_r H$	$\Delta_r G$	Log K	$\Delta_r H$	$\Delta_r G$	Log K
	Reaction (IV.A)			Reaction (IV.B)			Reaction (IV.C)		
298.15	-987.61	-841.90	147.49	-962.72	-828.26	145.10	-900.82	-803.90	140.84
300	-986.44	-839.72	146.20	-961.60	-826.28	143.86	-899.81	-802.18	139.67
320	-972.26	-814.62	132.97	-947.93	-803.34	131.13	-887.30	-782.00	127.65
340	-960.68	-792.07	121.68	-936.71	-782.82	120.26	-877.16	-764.13	117.39
360	-951.19	-771.56	111.95	-927.56	-764.34	110.90	-868.92	-748.09	108.54
380	-943.77	-753.06	103.51	-920.16	-747.58	102.76	-862.58	-733.90	100.88
400	-937.82	-736.00	96.11	-914.22	-732.27	95.62	-857.58	-720.98	94.15
420	-933.21	-720.23	89.57	-909.51	-718.17	89.32	-853.79	-709.22	88.20
440	-929.73	-705.56	83.76	-905.85	-705.10	83.70	-851.03	-698.43	82.91
460	-927.23	-691.81	78.56	-903.05	-692.90	78.68	-849.15	-688.45	78.17
480	-925.55	-678.85	73.87	-900.99	-681.42	74.15	-848.02	-679.17	73.91
500	-924.60	-666.56	69.63	-899.57	-670.58	70.05	-847.54	-670.47	70.04
T(K)	Reaction (IV.D)			Reaction (IV.E)			Reaction (IV.F)		
298.15	-929.96	-791.90	138.73	883.23	750.12	-131.41	715.91	586.43	-102.74
300	-928.98	-790.02	137.55	882.18	748.32	-130.29	715.03	584.96	-101.85
320	-916.66	-768.07	125.37	869.28	727.30	-118.72	703.62	567.19	-92.58
340	-906.60	-748.33	114.96	858.59	708.49	-108.84	694.12	551.38	-84.71
360	-898.43	-730.46	105.98	849.82	691.61	-100.35	686.24	537.19	-77.94
380	-891.86	-714.16	98.17	842.49	676.19	-92.95	679.69	524.35	-72.07
400	-886.63	-699.19	91.30	836.56	662.16	-86.47	674.24	512.62	-66.94
420	-882.52	-685.32	85.23	831.73	649.26	-80.74	669.71	501.80	-62.41
440	-879.35	-672.38	79.82	827.82	637.30	-75.65	665.92	491.74	-58.38
460	-876.96	-660.22	74.97	824.68	626.13	-71.10	662.72	482.31	-54.77
480	-875.22	-648.72	70.59	822.16	615.62	-66.99	660.00	473.39	-51.51
500	-874.04	-637.77	66.63	820.16	605.69	-63.27	657.63	464.91	-48.57

Table S.5. Calculated enthalpies ($\Delta_r H$) and free-energies ($\Delta_r G$) of reaction and associated reaction constants (Log K) of reactions (V.A)-(V.F). The values of $\Delta_r H$ and $\Delta_r G$ are in units of $\mathrm{kJ \cdot mol^{-1}}$.

T(K)	$\Delta_r H$	$\Delta_r G$	Log K	$\Delta_r H$	$\Delta_r G$	Log K	$\Delta_r H$	$\Delta_r G$	Log K
	Reaction (V.A)			Reaction (V.B)			Reaction (V.C)		
298.15	-791.75	-651.57	114.15	-766.86	-637.93	111.76	-704.95	-613.57	107.49
300	-790.56	-649.52	113.09	-765.73	-636.08	110.75	-703.94	-611.98	106.55
320	-778.82	-628.40	102.57	-754.49	-617.12	100.73	-693.86	-595.78	97.25
340	-768.69	-608.92	93.55	-744.72	-599.67	92.13	-685.17	-580.98	89.25
360	-759.95	-590.83	85.72	-736.31	-583.61	84.68	-677.68	-567.36	82.32
380	-752.41	-573.95	78.89	-728.80	-568.47	78.14	-671.23	-554.79	76.26
400	-745.94	-558.15	72.88	-722.34	-554.42	72.40	-665.70	-543.12	70.92
420	-740.42	-543.29	67.57	-716.72	-541.23	67.31	-661.00	-532.27	66.20
440	-735.73	-529.28	62.83	-711.84	-528.82	62.78	-657.03	-522.15	61.99
460	-731.80	-516.02	58.59	-707.62	-517.11	58.72	-653.72	-512.67	58.21
480	-728.54	-503.44	54.78	-703.98	-506.02	55.06	-651.00	-503.77	54.82
500	-725.88	-491.47	51.34	-700.85	-495.50	51.76	-648.82	-495.39	51.75
T(K)	Reaction (V.D)			Reaction (V.E)			Reaction (V.F)		
298.15	-734.10	-601.57	105.39	638.34	501.39	-87.84	422.00	279.29	-48.93
300	-733.10	-599.83	104.44	637.28	499.66	-87.00	421.11	277.83	-48.37
320	-723.22	-581.85	94.98	626.83	481.99	-78.67	412.14	262.78	-42.89
340	-714.61	-565.18	86.83	617.59	465.62	-71.53	404.12	248.78	-38.22
360	-707.19	-549.73	79.76	609.52	450.48	-65.36	396.90	235.65	-34.19
380	-700.51	-535.05	73.55	602.16	436.09	-59.94	390.38	223.27	-30.69
400	-694.75	-521.34	68.08	595.73	422.69	-55.20	384.46	211.52	-27.62
420	-689.73	-508.38	63.22	590.02	410.06	-51.00	379.09	200.35	-24.92
440	-685.35	-496.10	58.89	584.95	398.13	-47.26	374.18	189.68	-22.52
460	-681.53	-484.44	55.01	580.44	386.81	-43.92	369.67	179.47	-20.38
480	-678.21	-473.31	51.51	576.41	376.05	-40.92	365.49	169.65	-18.46
500	-675.32	-462.68	48.34	572.77	365.79	-38.21	361.57	160.20	-16.74

Table S.6. Calculated enthalpies ($\Delta_r H$) and free-energies ($\Delta_r G$) of reaction and associated reaction constants (Log K) of reactions (VI.A)-(VI.F). The values of $\Delta_r H$ and $\Delta_r G$ are in units of $\mathrm{kJ \cdot mol^{-1}}$.

T(K)	$\Delta_r H$	$\Delta_r G$	Log K	$\Delta_r H$	$\Delta_r G$	Log K	$\Delta_r H$	$\Delta_r G$	Log K
	Reaction (VI.A)			Reaction (VI.B)			Reaction (VI.C)		
298.15	-526.35	-189.59	33.21	-501.46	-175.95	30.83	-439.56	-151.59	26.56
300	-524.34	-184.09	32.05	-499.51	-170.66	29.71	-437.72	-146.56	25.52
320	-507.18	-128.92	21.04	-482.84	-117.63	19.20	-422.21	-96.29	15.72
340	-492.81	-76.13	11.70	-468.84	-66.88	10.27	-409.29	-48.19	7.40
360	-479.48	-23.96	3.48	-455.85	-16.74	2.43	-397.21	-0.49	0.07
380	-472.61	22.21	-3.05	-449.00	27.69	-3.81	-391.43	41.38	-5.69
400	-466.82	67.82	-8.86	-443.21	71.55	-9.34	-386.58	82.84	-10.82
420	-463.80	111.17	-13.83	-440.10	113.22	-14.08	-384.38	122.18	-15.20
440	-463.48	152.39	-18.09	-439.59	152.85	-18.14	-384.78	159.52	-18.94
460	-465.80	191.64	-21.76	-441.63	190.56	-21.64	-387.73	195.00	-22.14
480	-470.66	229.13	-24.93	-446.10	226.55	-24.65	-393.13	228.81	-24.90
500	-478.01	265.06	-27.69	-452.98	261.04	-27.27	-400.95	261.15	-27.28
T(K)	**Reaction (VI.D)**			**Reaction (VI.E)**			**Reaction (VI.F)**		
298.15	-468.70	-139.59	24.45	1255.41	1090.72	-191.08	1921.54	1919.94	-336.36
300	-466.88	-134.40	23.40	1253.51	1086.59	-189.19	1919.79	1917.12	-333.79
320	-451.58	-82.36	13.44	1237.47	1046.18	-170.77	1905.06	1890.65	-308.61
340	-438.73	-32.39	4.98	1223.84	1007.85	-154.83	1892.50	1866.03	-286.67
360	-426.72	17.14	-2.49	1211.89	970.89	-140.87	1882.09	1843.34	-267.46
380	-420.71	61.11	-8.40	1203.97	937.69	-128.89	1873.80	1822.63	-250.53
400	-415.63	104.63	-13.66	1197.73	905.88	-118.29	1867.60	1803.88	-235.56
420	-413.11	146.08	-18.17	1193.88	876.19	-108.97	1863.42	1787.06	-222.25
440	-413.10	185.57	-22.03	1192.34	848.49	-100.73	1861.21	1772.08	-210.37
460	-415.53	223.23	-25.35	1193.06	822.66	-93.41	1860.90	1758.82	-199.72
480	-420.33	259.26	-28.21	1195.93	798.52	-86.89	1862.42	1747.15	-190.12
500	-427.45	293.85	-30.70	1200.92	775.88	-81.05	1865.73	1736.92	-181.45