Supplementary Files

The Effective Volumes of Waters of Crystallization & the Thermodynamics of Cementitious Materials

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Table S1: Thermodynamic data for AFm and AFt phases, from *Cemdata18.*[1]

AFm-phases[a]	**$n(H_2O)$**	**V_m / $cm^3\ mol^{-1}$**	**S /J $K^{-1}\ mol^{-1}$**	**C_p /J $K^{-1}\ mol^{-1}$**	**C_p/S**
C_4AH_{19}	*19*	*369*	*1120*	1382.5	1.23
C_4AH_{13}	*13*	*274*	*831.5*	1141.5	1.37
C_4AH_{11}	*11*	*257*	*772.7*	1061.4	1.37
$C_2AH_{7.5}$	7.5	180	450	540.1	1.20
CAH_{10}	10	193	610	668.2	1.10
$C_4Ac_{0.5}H_{12}$	*12*	*285*	*713*	920.0	1.29
$C_4Ac_{0.5}H_{10.5}$	*10.5*	*261*	*668.3*	845.6	1.27
$C_4Ac_{0.5}H_9$	*9*	*249*	*622.5*	785.6	1.26
C_4AcH_{11}	*11*	*262*	*657*	881.6	1.34
C_4AcH_9	*9*	*234*	*640.6*	801.2	1.25
C_4AsH_{16}	*16*	*351*	*975*	1114.8	1.14
C_4AsH_{14}	*14*	*332*	*960.9*	1028.5	1.07
C_4AsH_{12}	*12*	*310*	*791.6*	948.4	1.20
$C_4AsH_{10.5}$	*10.5*	*282*	*721*	888.2	1.23
C_4AsH_9	*9*	*275*	*703.6*	828.2	1.18
C_2ASH_8	*8*	*216*	*546*	615.0	1.13
C_2ASH_7	*7*	*215*	*487.6*	562.6	1.15
$C_2ASH_{5.5}$	*5.5*	*213*	*454.8*	502.4	1.10
$C_4As_{0.5}ClH_{12}$	12	289	820	940.7	1.15
$C_4ACl_2H_{10}$	10	272	731	851.9	1.17
$C_4A(NO_3)_2H_{10}$	10	296	821	934.6	1.14
$C_4A(NO_2)_2H_{10}$	10	275	799	900.9	1.13
Ferrates					
C_4FH_{13}	13	286	630	1118.5	1.78
$C_4Fc_{0.5}H_{10}$	10	273	1270	841.2	0.66
C_4FcH_{12}	12	292	1230	892.5	0.73
C_4FsH_{12}	12	321	1430	967.6	0.68
$C_4FCl_2H_{10}$	10	278	1286	854.4	0.66

AFt-phases	n(H_2O)	V_m / cm^3 mol^{-1}	S /J K^{-1} mol^{-1}	C_p /J K^{-1} mol^{-1}	C_p/S
$C_6As_3H_{32}$	*32*	*707*	*1900*	2174.2	1.14
$C_6As_3H_{30}$	*30*	*708*	*1792.4*	2094.8	1.17
$C_6As_3H_{13}$	*13*	*411*	*1960.4*	1412.9	0.72
$C_6As_3H_9$	*9*	*361*	*646.6*	1252.7	1.94
$C_6Ac_3H_{30}$	30	650	1858	2121.1	1.14
$C_6F_2s_3H_{32}$	32	717	1937	2199.6	1.14
C_3ScsH_{15}	15	330	897.1	1071.2	1.19
C_3AH_6	6	150	422	445.4	1.06
$C_3AS_{0.41}H_{5.18}$	5.18	146	399	429.6	1.08
$C_3AS_{0.84}H_{4.32}$	4.32	142	375	412.9	1.10
Ferrates					
C_3FH_6	6	155	870	645.5	0.74
$C_3FS_{0.84}H_{4.32}$	4.32	149	840	434.4	0.52
$C_3A_{0.5}F_{0.5}S_{0.84}H_{4.32}$	4.32	146	619	416.3	0.67
$C_3FS_{1.34}H_{3.32}$	3.32	145	820	414.8	0.51

[a] The chemical formulae, in cement shorthand notation, are: A=Al_2O_3; C=CaO; F=Fe_2O_3; H=H_2O; S=SiO_2; c=CO_2; s=SO_3.

Table S2: Thermodynamics of hydrated mineral calcium silicates ex Table 10 of Blanc.[2-3] The final columns list the number of waters of crystallization per formula unit, $n(H_2O)$, molar volume, V_m/cm^3 mol^{-1}, molar entropy, S/J K^{-1} mol^{-1}, heat capacity, C_p/J K^{-1} mol^{-1}, and ratio C_p/S.

Mineral	Chemical Formula	$n(H_2O)$	V_m /cm^3 mol^{-1}	S /J K^{-1} mol^{-1}	C_p /J K^{-1} mol^{-1}	C_p/S
Crystalline C-S-H						
Gyrolite	$Ca_2Si_3O_7(OH)_2.2H_2O$	2	137.34	309.32	325.94	1.05
Xonotlite	$Ca_6Si_6O_{17}(OH)_2$	0	256.9	573.74	628.64	1.10
Tobermorite-14	$Ca_5Si_6(OH)O_{16.5}.10H_2O$	10	351.3	874.57	973.53	1.11
Tobermorite-11	$Ca_5Si_6(OH)O_{16.5}.5H_2O$	5.5	286.19	692.55	764.91	1.10
Foshagite	$Ca_4Si_3O_9(OH)_2.0.5H_2O$	0.5	160.66	295.07	309.38	1.05
Jennite	$Ca_9Si_6O_{16}(OH)_{10}.6H_2O$	6	456.4	839.25	933.21	1.11
Afwillite	$Ca_3Si_2O_6(OH)_2.2H_2O$	2	129.53	289.7	303.55	1.05
Hillebrandite	$Ca_2SiO_3(OH)_2.0.17H_2O$	0.17	72.58	179.71	177.46	0.99
Crystalline C-S-H Estimated						
Truscottite	$Ca_7Si_{12}O_{29}(OH)_4.H_2O$	1	478.73	927.68	1034.1	1.11
Okenite	$CaSi_2O_4(OH)_2.H_2O$	1	94.77	208.52	210.07	1.01
C2SH,α	$Ca_2(HSiO_4)(OH)$	0	71.12	122.38	111.88	0.91
Jaffeite	$Ca_6(Si_2O_7)(OH)_6$	0	174.38	326.19	344.9	1.06

Table S3: Thermodynamics of three hydrated sulfate minerals.[2] The final columns list the number of waters of crystallization per formula unit, $n(H_2O)$, molar volume, V_m/cm^3 mol^{-1}, molar entropy, S/J K^{-1} mol^{-1}, heat capacity, C_p/J K^{-1} mol^{-1}, and ratio C_p/S.

Mineral	Chemical Formula	$n(H_2O)$	V_m[4] /cm^3 mol^{-1}	S /J K^{-1} mol^{-1}	C_p /J K^{-1} mol^{-1}	C_p/S
Ettringite	$Ca_6Al_2(SO_4)_3(OH)_{12}\cdot 26H_2O$	26	705.1	1747.24	1794.55	1.03
		26		1900	2174.24	1.14
Monosulfoaluminate	$Ca_4Al_2(SO_4)(OH)_{12}\cdot 6H_2O$	6	308.2	747.26	783.68	1.05
		6		821	942.24	1.15
Gypsum	$CaSO_4\cdot 2H_2O$	2	74.5	193.8	186.2	0.96

References

1. Lothenbach, B.; Kulik, D. A.; Matschei, T.; Balonis, M.; Baquerizo, L.; Dilnesa, B.; Miron, G. D.; Myers, R. J., Cemdata18: A chemical thermodynamic database for hydrated Portland cements and alkali-activated materials. *Cem. Concr. Res.* **2019,** *115*, 472-506.
2. Blanc, P.; Bourbon, X.; Lassin, A.; Gaucher, E. C., Chemical model for cement-based materials: Thermodynamic data assessment for phases other than C–S–H. *Cem. Concr. Res.* **2010,** *40*, 1360-1374.
3. Blanc, P.; Bourbon, X.; Lassin, A.; Gaucher, E. C., Chemical model for cement-based materials: Temperature dependence of thermodynamic functions for nanocrystalline and crystalline C–S–H phases. *Cem. Concr. Res.* **2010,** *40*, 851-866.
4. Balonis, M.; Glasser, F. P., The density of cement phases. *Cem. Concr. Res.* **2009,** *39*, 733-739.