Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting Information for

From Lime to Silica and Alumina:

Systematic Modeling of Cement Clinkers using a General Force-Field

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Table S1. Structural data for selected crystalline phases of the lime-silica-alumina system. Buckingham potential MD data in italic and relative deviations between MD and experimental results in parenthesis.

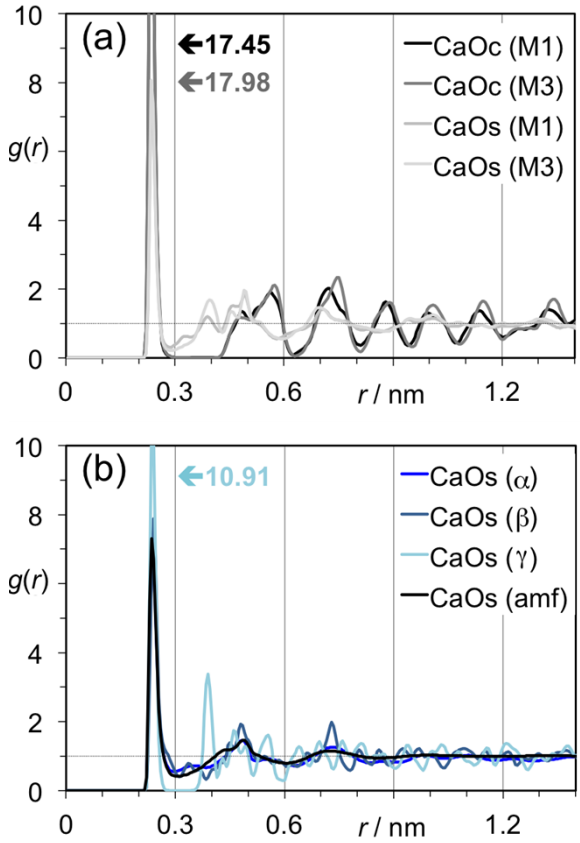
name	**alite**	**alite**	**belite**	**belite**	**belite**	**rankinite**	**wollastonite**	**wollastonite**	**-**	**mayenite**	**krotite**	**grossite**	**gehlenite**
allotrope	**M_1**	**M_3**	**α**	**β**	**γ**	**-**	**α**	**β**	**-**	**-**	**-**	**-**	**-**
formula	Ca_3SiO_5	Ca_3SiO_5	Ca_2SiO_4	Ca_2SiO_4	Ca_2SiO_4	$Ca_3Si_2O_7$	$CaSiO_3$	$CaSiO_3$	$Ca_3Al_2O_6$	$Ca_{12}Al_{14}O_{33}$	$CaAl_2O_4$	$CaAl_4O_7$	$Ca_2Al_2SiO_7$
CCN	C_3S	C_3S	C_2S	C_2S	C_2S	C_3S_2	CS	CS	C_3A	$C_{12}A_7$	CA	CA_2	C_2AS
space group	*Pc*	*Cm*	*$P6_3mc$*	*$P2_1/n$*	*Pbnm*	*$P2_1/a$*	*P-1*	*$P2_1/a$*	*Pa3*	*I-43d*	*$P2_1/n$*	*C2/c*	*$P\text{-}42_1\,m$*
reference	23	24	25	26	27	28	29	30	31	32	33	34	35
MD cells	2×8×5	2×8×3	10×11×8	10×11× 8	10×8×6	5×6×8	7×8×8	4×7×8	4×4×4	5×5×5	6×7×4	4×6×10	7×7×10
MD atoms	12960	15552	12320	13440	11200	11520	13440	13440	16896	14750	14112	11520	11760
a / Å	27.874 *28.071* (0.70)	33.083 *33.373* (0.88)	5.579 *5.593* (0.26)	5.502 *5.617* (2.09)	5.081 *5.247* (3.27)	10.600 *10.667* (0.64)	7.940 *7.934* (-0.08)	15.426 *15.445* (0.12)	15.263 *15.270* (0.05)	11.979 *12.015* (0.29)	8.700 *8.673* (-0.31)	12.840 *12.957* (0.91)	7.690 *7.722* (0.42)
b / Å	7.059 *7.060* (0.02)	7.027 *7.192* (2.34)	5.579 *5.628* (0.87)	6.762 *6.848* (1.27)	11.224 *11.296* (0.64)	8.920 *9.068* (1.65)	7.320 *7.358* (0.52)	7.320 *7.364* (0.60)	15.263 *15.270* (0.05)	11.979 *12.017* (0.31)	8.092 *8.171* (0.98)	8.862 *8.748* (-1.29)	7.690 *7.722* (0.42)
c / Å	12.258 *12.233* (-0.20)	18.499 *18.682* (0.49)	7.150 *7.029* (-1.69)	9.339 *9.381* (0.45)	6.778 *6.805* (0.39)	7.890 *8.415* (3.23)	7.070 *7.192* (1.73)	7.066 *7.191* (1.77)	15.263 *15.270* (0.05)	11.979 *12.017* (0.32)	15.191 *15.109* (-0.54)	5.431 *5.471* (0.74)	5.100 *5.069* (-0.61)
α / °	90.00 *89.99*	90.00 *90.02*	90.00 *90.05*	90.00 *90.00*	90.00 *90.00*	90.00 *90.00*	90.00 *89.95*	90.00 *90.00*	90.00 *90.00*	90.00 *90.01*	90.00 *90.00*	90.00 *90.00*	90.00 *90.00*
β / °	116.03 *115.81*	94.12 *93.93*	90.00 *89.96*	94.14 *96.94*	90.00 *90.00*	119.60 *120.75*	95.37 *93.32*	95.40 *93.54*	90.00 *90.00*	90.00 *90.02*	90.17 *90.13*	106.83 *106.22*	90.00 *90.00*
γ / °	90.00 *90.01*	90.00 *89.98*	120.00 *119.61*	90.00 *90.00*	90.00 *90.00*	90.00 *90.00*	103.43 *103.43*	90.00 *90.00*	90.00 *90.00*	90.00 *90.03*	90.00 *90.00*	90.00 *90.00*	90.00 *90.00*
V_{cell} / Å^3	2167.24 *2182.50*	4289.43 *4473.14*	192.73 *192.38*	346.55 *358.91*	386.54 *403.31*	648.66 *677.14*	397.82 *407.69*	794.33 *816.44*	3555.66 *3560.75*	1719.12 *1735.04*	1069.45 *1070.71*	591.54 *595.47*	301.59 *302.28*
d / g.cm^{-3}	3.150 *3.128* (-0.70)	3.182 *3.072* (-3.47)	3.073 *3.079* (0.18)	3.301 *3.194* (-3.24)	2.960 *2.838* (-4.13)	2.953 *2.830* (-4.17)	2.909 *2.840* (-2.38)	2.914 *2.836* (-2.67)	3.028 *3.027* (-0.02)	2.690 *2.671* (-0.85)	2.950 *2.942* (-0.09)	2.919 *2.938* (0.65)	3.019 *3.037* (0.60)

Table S2. Statistical distribution of $Q^n(m\text{Al})$ units in amorphous materials with ratio C/S = 1.5 according to the Loewenstein's rule. The corresponding MD result is reported in italic.

	$CaO:SiO_2:Al_2O_3$ 58.3:38.8:2.9					$CaO:SiO_2:Al_2O_3$ 56.5:37.6:5.9					$CaO:SiO_2:Al_2O_3$ 54.6:36.4:9.1				
*m*Al	Q^0	Q^1	Q^2	Q^3	Q^4	Q^0	Q^1	Q^2	Q^3	Q^4	Q^0	Q^1	Q^2	Q^3	Q^4
0	1.00	0.85	0.72	0.61	0.52	1.00	0.69	0.47	0.32	0.22	1.00	0.50	0.25	0.13	0.06
	1.00	*0.78*	*0.63*	*0.51*	*0.41*	*1.00*	*0.66*	*0.44*	*0.27*	*0.18*	*1.00*	*0.51*	*0.28*	*0.15*	*0.05*
1		0.15	0.26	0.33	0.37		0.31	0.43	0.44	0.41		0.50	0.50	0.38	0.25
		0.22	*0.33*	*0.38*	*0.45*		*0.34*	*0.44*	*0.45*	*0.30*		*0.49*	*0.50*	*0.41*	*0.31*
2			0.02	0.06	0.10			0.10	0.20	0.28			0.25	0.37	0.37
			0.05	*0.10*	*0.10*			*0.12*	*0.25*	*0.38*			*0.21*	*0.35*	*0.35*
3				0.00	0.01				0.03	0.08				0.12	0.25
				0.01	*0.03*				*0.04*	*0.14*				*0.10*	*0.24*
4					0.00					0.01					0.06
					0.00					*0.00*					*0.06*
Q^n %	12.8	35.6	35.3	14.5	1.88	7.12	30.1	37.3	21.4	3.46	4.14	23.7	37.8	27.9	6.50

Table S3. Statistical distribution of $Q^n(m\text{Al})$ units in amorphous materials with ratio C/S = 1.0 according to the Loewenstein's rule. The corresponding MD result is reported in italic.

	$CaO:SiO_2:Al_2O_3$ 48.6:48.6:2.9					$CaO:SiO_2:Al_2O_3$ 47.1:47.1:5.9					$CaO:SiO_2:Al_2O_3$ 45.5:45.5:9.1				
*m*Al	Q^0	Q^1	Q^2	Q^3	Q^4	Q^0	Q^1	Q^2	Q^3	Q^4	Q^0	Q^1	Q^2	Q^3	Q^4
0	1.00	0.88	0.78	0.68	0.60	1.00	0.75	0.56	0.42	0.32	1.00	0.60	0.36	0.22	0.13
	1.00	*0.85*	*0.72*	*0.62*	*0.48*	*1.00*	*0.74*	*0.53*	*0.39*	*0.30*	*1.00*	*0.64*	*0.41*	*0.26*	*0.17*
1		0.12	0.21	0.28	0.33		0.25	0.38	0.42	0.42		0.40	0.48	0.43	0.35
		0.15	*0.26*	*0.31*	*0.41*		*0.26*	*0.40*	*0.43*	*0.39*		*0.36*	*0.46*	*0.45*	*0.35*
2			0.01	0.04	0.07			0.06	0.14	0.21			0.16	0.29	0.34
			0.03	*0.06*	*0.09*			*0.07*	*0.16*	*0.24*			*0.14*	*0.25*	*0.33*
3				0.00	0.01				0.02	0.05				0.06	0.15
				0.01	*0.02*				*0.02*	*0.06*				*0.05*	*0.13*
4					0.00					0.00					0.03
					0.00					*0.00*					*0.02*
Q^n %	2.6	18.0	38.3	32.0	9.07	1.66	13.1	35.1	36.4	13.7	0.85	9.27	31.1	40.3	18.4


(a)
17.45
17.98
CaOc (M1)
CaOc (M3)
CaOs (M1)
CaOs (M3)
g(r)
r / nm
(b)
10.91
CaOs (α)
CaOs (β)
CaOs (γ)
CaOs (amf)
g(r)
r / nm

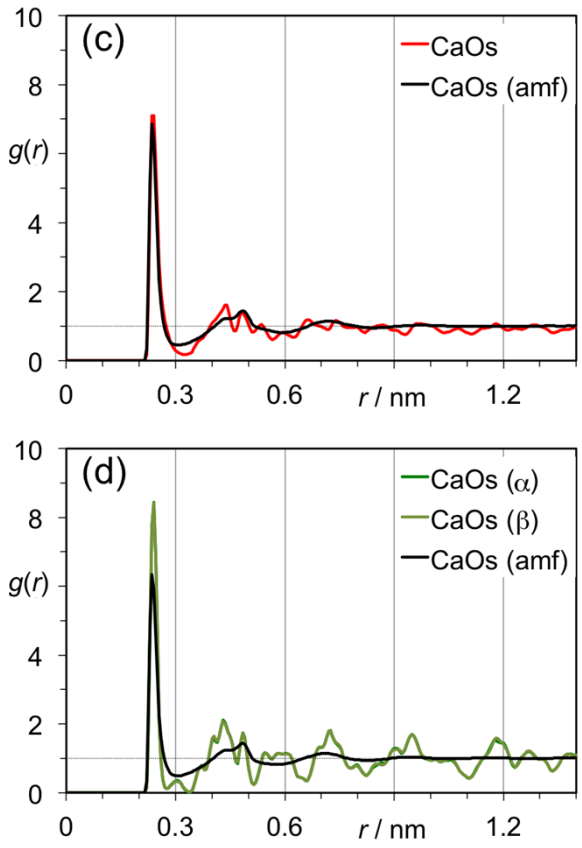
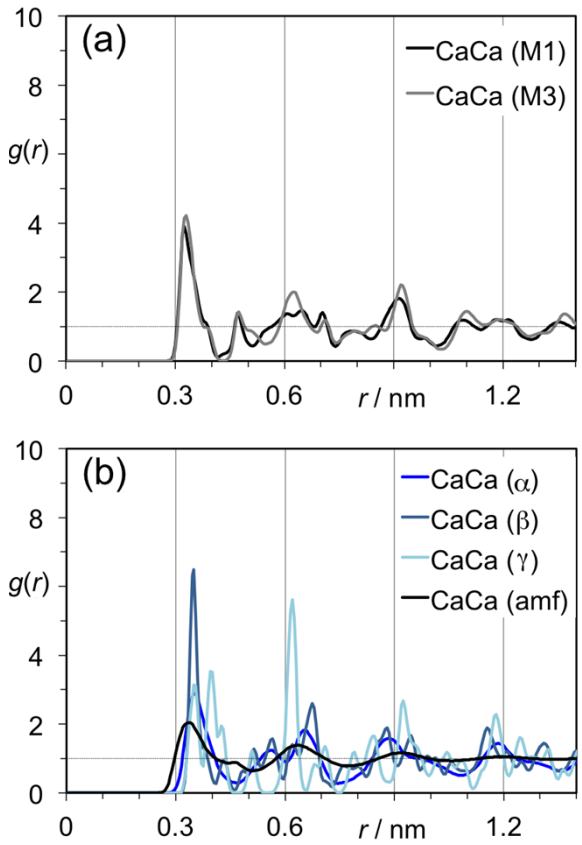


Figure S1. Radial pair distribution functions between calcium and oxygen atoms of: (a) M_1 and M_3 alite; (b) α-, β-, and γ-belite; (c) rankinite and (d) α-, β-wollastonite. The black lines in diagrams b-d represent the corresponding amorphous phases.


(a)
CaCa (M1)
CaCa (M3)
g(r)
r / nm
(b)
CaCa (α)
CaCa (β)
CaCa (γ)
CaCa (amf)
g(r)
r / nm

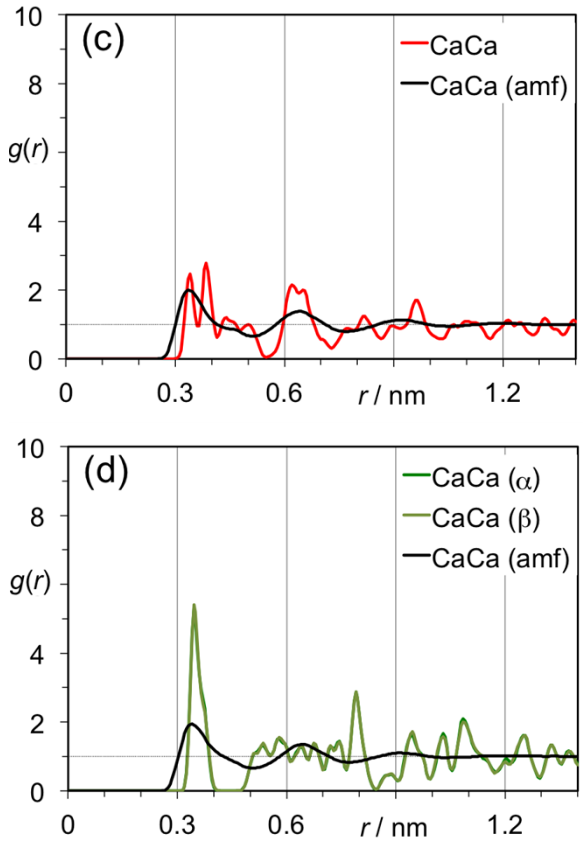


Figure S2. Radial pair distribution functions between calcium atoms of: (a) M_1 and M_3 alite; (b) α-, β-, and γ-belite; (c) rankinite and (d) α-, β-wollastonite. The black lines in diagrams b-d represent the corresponding amorphous phases.