Supplementary information. Crystal structures of hypothetical frameworks made of D4R-units exclusively, as obtained from the AASBU method and after energy minimizations of the silicate forms. The positions of oxygens are indicated for all hypothetical frameoworks, including the existing ACO topology and its tilted hypothetical variety.

ACO			***Im3m***
	x/a	y/b	z/c
O1	0.8104	0.8104	0
O2	¼	¼	3/4

ACO tilted			***P4/mnc***
	x/a	y/b	z/c
O1	0.8359	0.3359	3/4
O2	0.9381	0.8102	0.3110
O3	0.7473	0.8720	1/2

Structure T1 (one D4R-unit in *Cm*)			***Fmmm***
	x/a	y/b	z/c
O1	½	½	0.4483
O2	½	½	0.2789
O3	¾	½	¼
O4	¼	¾	0.3704
O5	0	0.8176	0.3782
O6	0.3123	0.4059	0.3267
O7	0.2729	½	½
O8	0.2885	0.5950	0.4193

Structure T2 (one D4R-unit in *P*-1)			***Cmmm***
	x/a	y/b	z/c
O1	0.2609	½	0
O2	½	½	0.1959
O3	0.2189	-0.0937	0.6828
O4	¾	¼	½
O5	0	0.1693	½

Structure T3 (one D4R unit in *Pna2*$_1$)			***Cmcm***
	x/a	y/b	z/c
O1	¼	-¼	0
O2	0	-0.2877	0.0674
O3	0.7882	-0.3726	0.0610
O4	0.8005	-0.2817	¼
O5	0.2338	-1/2	0
O6	0	-0.4516	0.1039
O7	0.2264	-0.4632	¼

Structure T4 (one D4R-unit in *P*-1)	***C2/m***

	x/a	y/b	z/c
O1	0.1186	½	0.9173
O2	0.0887	0.7085	0.7523
O3	0.0757	½	0.6083
O4	0.1400	0.7460	0.0088
O5	0	0.7211	½
O6	0.3401	½	0.9228
O7	0.2874	0.6872	0.7412
O8	0.2258	0.6447	0.9030
O9	0.2279	½	0.5586
O10	¼	¾	½
O11	0.1455	0.2935	0.5740

Structure T5 (one D4R-unit in *P*na2$_1$) ***P*na2$_1$**

	x/a	y/b	z/c
O1	0.4859	0.6490	0.5072
O2	0.5480	0.6089	0.7949
O3	0.6372	0.4925	0.7789
O4	0.6781	0.5786	0.0314
O5	0.5398	0.7391	0.7321
O6	0.3633	0.6620	0.7556
O7	0.3319	0.7143	0.0447
O8	0.4931	0.8012	0.0035
O9	0.1728	0.6680	0.8670
O10	0.3198	0.8367	0.1547
O11	0.6348	0.6657	0.2878
O12	0.6485	0.7095	0.9912
O13	0.4918	0.5160	0.0119
O14	0.4595	0.6208	0.1927
O15	0.3072	0.5816	0.0110
O16	0.4577	0.7461	0.2883

Structure T6 (one D4R-unit in *P*4) ***P*4/mmm**

	x/a	y/b	z/c
O1	½	0	0.6943
O2	½	0.2089	0
O3	0.6885	0	½
O4	0.6969	-0.3031	0
O5	0.6705	-0.1129	0.1828

Structure T7 (one D4R unit in *R*3) ***R*3**

	x/a	y/b	z/c
O1	0.5076	0.7506	0.1760
O2	0.5715	0.7279	0.9800
O3	0.5821	0.6636	0.8154
O4	0.6294	0.7604	0.7567

O5	0.5749	0.8391	0.9838
O6	0.6091	0.8043	0.2692
O7	0.6935	0.8874	0.2749
O8	0.6597	0.9243	0.9918
O9	0.6657	0.8293	0.4992
O10	0.7463	0.9957	0.1632
O11	0.6574	0.7289	0.9770
O12	0.7638	0.8414	0.9772
O13	0.6968	0.8054	0.2661
O14	0.7623	0.9256	0.9882
O15	0.7357	0.8709	0.7536
O16	0.8299	0.9204	0.8383
O17	0.6310	0.8696	0.7572

Structure T8 (one D4R unit in *P*3) ***P*6/mmm**

	x/a	y/b	z/c
O1	0.4847	0.2424	0
O2	0.6311	0	½
O3	0.5821	0.1642	0
O4	0.6653	0.0906	0.8173
O5	½	0	0.6952

Structure T9 (one D4R-unit in *P*4) ***P*4/mmm**

	x/a	y/b	z/c
O1	0.1156	½	0
O2	0	0.5763	0
O3	0	0.6848	0.3060
O4	0	0.8213	0
O5	0.8833	0.6905	½
O6	0.8959	0.7593	0.8175
O7	0.8768	0.8768	0
O8	0.8893	0.6200	0.8168

Structure T10 (one D4R-unit in *P*23) ***P*-43m**

	x/a	y/b	z/c
O1	0.0888	0.8079	0.2943
O2	0.9585	0.8515	0.2954
O3	0.8325	0.6521	0.1675
O4	0.0627	0.7491	0.0627
O5	0.0242	0.8256	0.1744
O6	0.8889	0.7337	0.2663
O7	0.9963	0.6289	0.3711
O8	0.9388	0.7534	0.3952
O9	0	½	0.3238
O10	0.1961	0.6167	0.1961
O11	0.1605	0.6916	0.3084
O12	0.0772	0.7368	0.4111