Supporting Information

for

Shin et al., “Targeted Synthesis of Two Super-Complex Zeolites with Embedded Isorecticular Structures”

Synthesis Details

The reagents used for zeolite syntheses included tetraethylammonium bromide (TEABr, 98%, Aldrich), NaOH (50% aqueous solution, Aldrich), $\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$ (98%, DC Chemical), aluminum hydroxide ($\text{Al}(\text{OH})_3 \cdot 1.0\text{H}_2\text{O}$ (Aldrich), colloidal silica (Ludox AS-40, DuPont), and deionized water. The final composition of the synthesis mixture was $5.2\text{TEABr} \cdot 1.9\text{Na}_2\text{O} \cdot 0.5\text{Ca}(\text{NO}_3)_2 \cdot x\text{Al}_2\text{O}_3 \cdot 7.2\text{SiO}_2 \cdot y\text{H}_2\text{O}$, where x and y are varied between $0.5 \leq x \leq 1.5$ and $150 \leq y \leq 390$, respectively. After being stirred at 80 °C for 24 h, the synthesis mixture was charged into Teflon-lined 23-mL autoclaves and heated to 145 °C under rotation (60 rpm) for 2 days. The solid products were recovered by filtration, washed repeatedly with water, and then dried overnight at room temperature.

Materials Characterization

Powder X-ray diffraction (XRD) patterns were recorded on a PANalytical X'Pert diffractometer (Cu K_α radiation) with an X'Celerator detector. Data were collected with a fixed divergence slit (0.50°) and Soller slits (incident and diffracted = 0.04 rad). **Crystal morphology and average size were determined by a JEOL JSM-6510 scanning electron microscope (SEM). The CO_2 , CH_4 , and N_2 adsorption isotherms were measured at 25 °C and at pressures up to 1.2 bar using a Mirae SI nanoPorosity-XG analyzer. Prior to the experiments, zeolite sample was evacuated at 250 °C for 6 h.**

Synchrotron powder XRD data for the solid product (a mixture of PST-25, PST-26, and PST-28) obtained from Run 8 in Table 1 were collected on the 9B beamline equipped with a ceramic furnace of the Pohang Acceleration Laboratory (PAL; Pohang, Korea) using monochromated X-rays ($\lambda = 1.4865 \text{ \AA}$). The detector arm of the vertical scan diffractometer consists of seven sets of Soller slits, flat Ge(111) crystal analyzers, anti-scatter baffles, and scintillation detectors, with each set separated by 20°. Data were obtained on the sample at room temperature in flat plate mode, with a step size of 0.01° and an overlap of 0.50° to the

next detector bank over the 2θ range $6^\circ - 126.5^\circ$. Profile fitting was performed in the 2θ range of $6^\circ - 70^\circ$ by the LeBail method^[S1] using the GSAS suite of programs^[S2]. **Simulated XRD patterns were obtained using the CrystalMaker/CrystalDiffract software package.**

Zeolite powders were dispersed in absolute ethanol and treated in an ultrasonic bath for 2 min. A droplet of the suspension was transferred onto a carbon-coated copper TEM grid and dried in air. The dried TEM grid was then loaded on to a Gatan-914 cryo-transfer holder at room temperature. Liquid nitrogen was used to cool down the TEM grid to below -100°C while the tip of the holder was kept in a sealed environment to prevent ice formation due to the water vapor in the air. The TEM holder was then quickly inserted into a JEOL JEM2100 TEM equipped with a LaB_6 filament and operated at 200 kV. The TEM experiment began after the temperature of the sample stabilized between -176°C to -177°C . The purpose of the cryo-treatment is to protect the large structures from distortion/damage caused by the high vacuum inside the TEM column. The SAED patterns were collected on a 14-bit Orius SC200D camera side-mounted at a 35mm port. A combination of low beam intensity and long camera exposure was used during the data collection to reduce electron radiation damage and enhance the collection of weak diffraction spots. We note that due to the large unit cells of the crystals, an extended camera length of 3-meters was used in order to observe the characteristic diffraction patterns that are located so close to the center beam. As a consequence of the large camera length, each pixel in the SAED became 0.01 1/nm. The pixel size of SAED collected using the conventional 100 mm camera length for our microscope setup is 0.16 1/nm. It means that at such large camera length, results obtained are very sensitive to the physical measurement and calibration, even at single pixel level. This is the reason why our SAED measurements differ slightly to the XRD measurements.

Structure Predictions of RHO-G7 and RHO-G8

In our recent work,^[S3] we have utilized the strong reflections approach not only for the identification and structure determination of the RHO family of zeolites with embedded isorecticular structures, but also for the prediction of its energetically-feasible, hypothetical structures of higher generations. In principle, higher generation structural models could be obtained from their three-dimensional electron density maps, which are calculated by adopting both amplitudes and phases from the lower generation. Given the huge unit cells (75 and 85 Å, respectively) and structural complexities of RHO-G7 (PST-26) and RHO-G8 (PST-

28), on the other hand, it is quite challenging to locate the T and O atoms from the dizzy electron density maps and to complete their structures. To solve this problem, we have developed another simpler and more convenient model building approach based on the structural coding in real space. The construction procedures for the super-complicated structures RHO-G7 and RHO-G8 are as follows:

1. RHO-G7

- 1) Take a structural fragment in RHO-G6 as shown in Figure S1a. All the atoms in the fragment locate in the region with $0 \leq x \leq 0.5$; $0 \leq y \leq 0.5$; $0 \leq z \leq 0.577$ ($= 0.5 \times a_{\text{RHO-G7}}/a_{\text{RHO-G6}}$); $x \leq z$; $y \leq z$; $x \leq y$. $a_{\text{RHO-G6}}$ (65 Å) and $a_{\text{RHO-G7}}$ (75 Å) are the unit cell parameter of RHO-G6 and RHO-G7, respectively.
- 2) The fractional coordinates x , y , and z for each of the atoms in the fragment are scaled by $a_{\text{RHO-G6}}/a_{\text{RHO-G7}}$ times, respectively.
- 3) Add the updated fractional coordinates x , y and z in the CIF file for RHO-G7. In case that some duplicated atoms are observed, the VESTA software is used to remove them.^[S4]
- 4) A total of 72 T and 168 O atoms in the asymmetric unit are found (Figure S1a).

2. RHO-G8

- 1) Take a structural fragment in RHO-G7 (Figure S1b). All the atoms in the fragment locate in the region with $0 \leq x \leq 0.5$; $0 \leq y \leq 0.5$; $0 \leq z \leq 0.566$ ($= 0.5 \times a_{\text{RHO-G8}}/a_{\text{RHO-G7}}$); $x \leq z$; $y \leq z$; $x \leq y$. $a_{\text{RHO-G7}}$ (75 Å) and $a_{\text{RHO-G8}}$ (85 Å) are the unit cells of RHO-G7 and RHO-G8, respectively.
- 2) The fractional coordinates x , y , and z for each of atom in the fragment are scaled by $a_{\text{RHO-G7}}/a_{\text{RHO-G8}}$, respectively.
- 3) Add the updated fractional coordinates x , y and z in the CIF file for RHO-G8. In case that some duplicated atoms are observed, the VESTA software is used to remove them.^[S4]
- 4) A total of 104 T and 240 O atoms in the asymmetric unit are found (Figure S1b).

Evolution of the numbers of T and O atoms in the RHO family of zeolites

Table S1 lists the numbers of symmetry-independent T and O atoms in the members (RHO-Gn's) of the RHO family up to 12th generation. All the numbers in Table S1 were determined from their structural models. It is of interest to note that the number of T and O atoms in each

generation follows several mathematical equations. In terms of the number of T atoms, odd and even generations of the RHO family follow two different equations, whereas the number of O atoms follows one single equation for all generations of the RHO family.

Number of T atoms in odd generations of the RHO family:

$$N_{T-odd} = \frac{1}{6}n^3 + \frac{1}{4}n^2 + \frac{1}{3}n + \frac{1}{4}$$

Number of T atoms in even generations of the RHO family:

$$N_{T-even} = \frac{1}{6}n^3 + \frac{1}{4}n^2 + \frac{1}{3}n$$

Number of O atoms in all generations of the RHO family:

$$N_O = \frac{1}{3}n^3 + n^2 + \frac{2}{3}n$$

where N_T and N_O are the numbers of T atoms and O atoms, respectively; n is the n -th generation.

Lattice Energy Calculations

All structures were optimized as a pure silica framework while maintaining the symmetry using the program GULP.^[S5] The Ewald method was utilized for the long-range coulombic interactions while the direct summation was used for the short-range interactions.^[S6] The atomic polarizability was described by the empirical core-shell model and was evaluated using the Catlow library.^[S7] Considering the large number of atoms in the framework of generation members of the RHO family of zeolites, especially of higher ones, the conjugate-gradient method utilizing multi-core CPUs was used before the final optimization with the rational functional optimization (RFO) method with a single core CPU.^[S8] Framework energies were evaluated relative to α -quartz, the most stable silica polymorph.^[S9]

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Figure S1. Structural fragments (left) of interest in (a) RHO-G6 and (b) RHO-G7 used for the construction of the complete structural models (right) of (a) RHO-G7 and (b) RHO-G8 after removing the duplicated atoms, respectively.

Figure S2. Simulated powder X-ray diffraction patterns of RHO-G1 to RHO-G8 in the RHO family of zeolites ($\lambda = 1.4865 \text{ \AA}$).

Figure S3. A selected zone of the experimental synchrotron PXRD pattern (top) of the product obtained from run 8 in Table 1, simulated PXRD pattern (middle) of the mixture with 20% RHO-G6, 45% RHO-G7, and 35% RHO-G8, and the PXRD patterns (bottom) of each of these three members (RHO-G6, red; RHO-G7, blue; RHO-G8, green) of the RHO family of zeolites. The CrystalDiffract program was used to simulate the XRD patterns of a number of mixtures with different fractions of RHO-G6 to RHO-G8.

Figure S4. Three-phase (PST-25, PST-26, and PST-28) LeBail fit for the product from run 8 in Table 1: observed data (crosses), calculated fit (solid line) and difference plot (lower trace) synchrotron diffraction patterns. Red, blue, and green vertical bars indicate the positions of Bragg peaks of PST-25, PST-26, and PST-28, respectively.

Figure S5. Adsorption isotherms at 25 °C of CO₂ (navy), CH₄ (green), and N₂ (pink) of the product (i.e., a mixture with RHO-G6, RHO-G7, and RHO-G8) obtained from run 8 in Table 1.

Figure S6. Experimental selected area electron diffraction patterns (taken from synthesized samples) and simulated electron diffraction patterns (based on hypothetical structural models) of (a and b) RHO-G4, (c and d) RHO-G5, (e and f) RHO-G6, (g and h) RHO-G7, and (i and j) RHO-G8 along the $[111]$ zone axis. It can be seen that strong reflection distributions are identical for all structures. The number of weak diffraction spots between the strong reflections systematically increases with the generation of the RHO family of zeolites.

Figure S7. SEM image of the solid product obtained from run 8 in Table 1 (see the main text) used for obtaining selected area electron diffraction patterns of PST-26 and PST-28.

Table S1. Numbers of the symmetry-independent T and O atoms in RHO-G n members of the RHO family of zeolites ($n = 1 - 12$).

Generation	Number of T atoms	Number of O atoms
RHO-G1 (zeolite Rho)	1	2
RHO-G2	3	8
RHO-G3 (paulingite)	8	20
RHO-G4 (ZSM-25)	16	40
RHO-G5 (PST-20)	29	70
RHO-G6 (PST-25)	47	112
RHO-G7 (PST-26)	72	168
RHO-G8 (PST-28)	104	240
RHO-G9	145	330
RHO-G10	195	440
RHO-G11	256	572
RHO-G12	328	728

Table S2. Atomic coordinates for the hypothetical RHO-G7 structure assuming the cubic space group $Im\bar{3}m$ with unit cell $a = b = c = 75 \text{ \AA}$.

Atom	x	y	z
T1	0.4129	0.1168	0.0872
T2	0.3867	0.2829	0.0872
T3	0.3874	0.1507	0.0467
T4	0.3197	0.2161	0.0203
T5	0.4542	0.2172	0.0204
T6	0.4129	0.3834	0.0458
T7	0.3460	0.1505	0.0468
T8	0.3874	0.1835	0.0205
T9	0.4126	0.2497	0.0461
T10	0.3204	0.1158	0.0868
T11	0.4531	0.3494	0.0201
T12	0.4126	0.2168	0.0205
T13	0.4796	0.1832	0.0205
T14	0.2802	0.2503	0.0874
T15	0.3464	0.3174	0.1126
T16	0.3459	0.1829	0.0207
T17	0.3461	0.1824	0.1533
T18	0.2531	0.1502	0.0470
T19	0.3196	0.2169	0.1134
T20	0.2130	0.1840	0.0204
T21	0.2798	0.1155	0.0868
T22	0.2800	0.2504	0.0458
T23	0.3870	0.2821	0.0456
T24	0.4130	0.1170	0.0461
T25	0.3873	0.1503	0.0872
T26	0.4538	0.0846	0.0192
T27	0.3874	0.1824	0.1129
T28	0.4798	0.0495	0.0197
T29	0.3459	0.1824	0.1129
T30	0.3460	0.1501	0.0873
T31	0.3460	0.3166	0.0202
T32	0.3459	0.2832	0.0872
T33	0.3193	0.2164	0.1537
T34	0.3459	0.2823	0.0461
T35	0.3205	0.1170	0.0459
T36	0.2803	0.2503	0.1800
T37	0.2797	0.2165	0.0202
T38	0.3870	0.3172	0.0200
T39	0.1869	0.1166	0.0869
T40	0.4126	0.3500	0.0201
T41	0.2531	0.1492	0.0874
T42	0.4543	0.1166	0.0461
T43	0.2799	0.0840	0.0195
T44	0.1860	0.0828	0.0194
T45	0.2126	0.1510	0.0472
T46	0.3202	0.2500	0.0875
T47	0.3204	0.0845	0.0196
T48	0.3873	0.0496	0.0196
T49	0.3202	0.2500	0.0461
T50	0.2125	0.0494	0.0196

T51	0.4126	0.0850	0.0195
T52	0.2543	0.1829	0.0205
T53	0.2540	0.0492	0.0196
T54	0.2794	0.1167	0.0455
T55	0.1470	0.1171	0.0465
T56	0.2129	0.1828	0.1129
T57	0.3463	0.0495	0.0200
T58	0.2796	0.2179	0.1137
T59	0.2532	0.1830	0.1531
T60	0.1200	0.0495	0.0189
T61	0.1464	0.1170	0.0871
T62	0.1873	0.1169	0.0463
T63	0.2539	0.1827	0.1130
T64	0.2130	0.1831	0.1539
T65	0.2126	0.1500	0.0873
T66	0.2795	0.2175	0.1537
T67	0.1456	0.0856	0.0188
T68	0.0802	0.0499	0.0198
T69	0.4540	0.2500	0.0460
T70	0.4124	0.2500	0.0876
T71	0.2801	0.2500	0.2199
T72	0.3203	0.2500	0.1797
O1	0.2758	0.1115	0.0670
O2	0.3669	0.1883	0.0237
O3	0.2330	0.1883	0.0238
O4	0.3243	0.2328	0.0330
O5	0.4577	0.2327	0.0334
O6	0.4330	0.2557	0.0431
O7	0.3420	0.1671	0.0333
O8	0.4083	0.2330	0.0330
O9	0.4331	0.2118	0.0236
O10	0.2578	0.1669	0.0338
O11	0.2666	0.0670	0.0243
O12	0.3339	0.1331	0.0425
O13	0.3917	0.1672	0.0331
O14	0.4102	0.2445	0.0671
O15	0.4579	0.1000	0.0329
O16	0.2999	0.2118	0.0239
O17	0.4090	0.1113	0.0671
O18	0.4001	0.2000	0.0248
O19	0.3437	0.3117	0.1331
O20	0.3243	0.1114	0.0671
O21	0.3909	0.2999	0.0998
O22	0.3668	0.3223	0.1099
O23	0.3907	0.1558	0.0669
O24	0.2999	0.2556	0.0431
O25	0.2752	0.2326	0.0337
O26	0.3329	0.2000	0.0247
O27	0.2087	0.1673	0.0327
O28	0.3666	0.2774	0.0905
O29	0.4081	0.1003	0.0333
O30	0.3999	0.3328	0.0251
O31	0.4328	0.3439	0.0245
O32	0.3424	0.1670	0.1001
O33	0.4669	0.2002	0.0248

O34	0.3670	0.2777	0.0433
O35	0.4329	0.3886	0.0429
O36	0.3324	0.1331	0.0912
O37	0.3997	0.2671	0.0916
O38	0.3430	0.1557	0.0673
O39	0.2767	0.2444	0.0671
O40	0.2998	0.2114	0.1096
O41	0.3259	0.2231	0.1332
O42	0.3329	0.2000	0.1579
O43	0.3423	0.3000	0.1001
O44	0.3666	0.3222	0.0244
O45	0.2999	0.1219	0.0895
O46	0.3247	0.1001	0.0335
O47	0.3915	0.1671	0.1003
O48	0.4001	0.2668	0.0422
O49	0.3326	0.1999	0.1087
O50	0.3246	0.2331	0.1007
O51	0.2668	0.2000	0.0244
O52	0.3327	0.2669	0.0424
O53	0.2753	0.1001	0.0328
O54	0.2581	0.1560	0.0674
O55	0.3896	0.2884	0.0668
O56	0.1992	0.0665	0.0246
O57	0.2674	0.1329	0.0915
O58	0.3997	0.1333	0.0426
O59	0.2767	0.2444	0.2000
O60	0.3000	0.1220	0.0437
O61	0.3242	0.2334	0.1670
O62	0.2326	0.1442	0.0429
O63	0.3669	0.1445	0.0900
O64	0.3669	0.1444	0.0435
O65	0.2657	0.1331	0.0426
O66	0.2752	0.2333	0.0997
O67	0.4006	0.1335	0.0912
O68	0.2328	0.0556	0.0230
O69	0.3331	0.2668	0.0915
O70	0.2089	0.1554	0.0672
O71	0.3433	0.2883	0.0671
O72	0.2997	0.2112	0.1580
O73	0.3425	0.2997	0.0328
O74	0.2676	0.1996	0.1098
O75	0.4666	0.0671	0.0248
O76	0.4574	0.3668	0.0324
O77	0.2330	0.1882	0.1564
O78	0.4086	0.3668	0.0331
O79	0.4106	0.3780	0.0669
O80	0.4006	0.0673	0.0246
O81	0.3670	0.1886	0.1100
O82	0.3236	0.2443	0.0671
O83	0.1431	0.1115	0.0669
O84	0.2013	0.1323	0.0435
O85	0.1911	0.1002	0.0320
O86	0.2999	0.2560	0.1761
O87	0.3904	0.2999	0.0332
O88	0.2569	0.1672	0.0998

O89	0.1674	0.1206	0.0437
O90	0.2752	0.2332	0.1672
O91	0.3335	0.0674	0.0245
O92	0.2576	0.1771	0.1333
O93	0.1387	0.0662	0.0236
O94	0.2329	0.1445	0.0908
O95	0.4331	0.0782	0.0240
O96	0.4331	0.1221	0.0433
O97	0.1906	0.1109	0.0673
O98	0.2327	0.1887	0.1098
O99	0.3000	0.0776	0.0235
O100	0.2752	0.2227	0.1335
O101	0.2670	0.2000	0.1573
O102	0.2104	0.1778	0.1333
O103	0.1667	0.1227	0.0891
O104	0.2088	0.1666	0.1003
O105	0.3670	0.0556	0.0233
O106	0.1995	0.1333	0.0904
O107	0.1425	0.1003	0.0326
O108	0.1646	0.0811	0.0214
O109	0.1000	0.0557	0.0236
O110	0.9653	0.1182	0.0347
O111	0.5653	0.5653	0.5195
O112	0.4662	0.4662	0.0250
O113	0.3998	0.3998	0.0919
O114	0.3997	0.3997	0.0422
O115	0.3326	0.3326	0.0252
O116	0.2666	0.2666	0.0420
O117	0.3324	0.3324	0.1083
O118	0.1998	0.1998	0.0247
O119	0.3325	0.3325	0.1578
O120	0.2664	0.2664	0.0918
O121	0.2669	0.2669	0.2247
O122	0.2661	0.2661	0.1753
O123	0.1999	0.1999	0.1576
O124	0.1999	0.1999	0.1080
O125	0.1327	0.1327	0.0914
O126	0.4650	0.4220	0.4650
O127	0.3635	0.6365	0.5509
O128	0.4761	0.1778	0.0000
O129	0.3436	0.1780	0.0000
O130	0.4565	0.2224	0.0000
O131	0.2070	0.0440	0.0000
O132	0.4620	0.0901	0.0000
O133	0.3900	0.1778	0.0000
O134	0.2754	0.0882	0.0000
O135	0.1876	0.0828	0.0000
O136	0.4759	0.3117	0.0000
O137	0.4739	0.0438	0.0000
O138	0.4104	0.2221	0.0000
O139	0.3419	0.0442	0.0000
O140	0.3232	0.2223	0.0000
O141	0.2104	0.1779	0.0000
O142	0.4568	0.3554	0.0000
O143	0.2580	0.0443	0.0000

O144	0.2564	0.1780	0.0000
O145	0.2762	0.2222	0.0000
O146	0.3247	0.0884	0.0000
O147	0.3933	0.0438	0.0000
O148	0.3437	0.3120	0.0000
O149	0.1418	0.0915	0.0000
O150	0.4087	0.0890	0.0000
O151	0.3905	0.3117	0.0000
O152	0.1217	0.0427	0.0000
O153	0.4768	0.4443	0.0000
O154	0.4098	0.3552	0.0000
O155	0.0755	0.0443	0.0000
O156	0.3248	0.1001	0.1001
O157	0.2753	0.1003	0.1003
O158	0.3915	0.0336	0.0336
O159	0.3421	0.0337	0.0337
O160	0.2577	0.0330	0.0330
O161	0.2090	0.0339	0.0339
O162	0.1920	0.1002	0.1002
O163	0.1426	0.1001	0.1001
O164	0.2089	0.1671	0.1671
O165	0.2576	0.1681	0.1681
O166	0.3429	0.1778	0.1331
O167	0.4669	0.3328	0.0251
O168	0.3000	0.2555	0.0901

Table S3. Atomic coordinates for the hypothetical RHO-G8 structure assuming the cubic space group $Im\bar{3}m$ with unit cell $a = b = c = 85 \text{ \AA}$.

Atom	<i>x</i>	<i>y</i>	<i>z</i>
T1	0.2463	0.1924	0.1004
T2	0.2238	0.1617	0.0180
T3	0.2461	0.1025	0.0402
T4	0.2824	0.1910	0.1002
T5	0.2230	0.1315	0.0773
T6	0.2239	0.0436	0.0174
T7	0.1883	0.1616	0.0999
T8	0.3642	0.0747	0.0172
T9	0.2832	0.0746	0.0174
T10	0.3416	0.0439	0.0173
T11	0.3053	0.2496	0.0405
T12	0.2233	0.1614	0.1349
T13	0.1878	0.0435	0.0174
T14	0.3413	0.2789	0.0176
T15	0.4229	0.1334	0.0413
T16	0.1295	0.1035	0.0409
T17	0.2830	0.2207	0.0407
T18	0.1650	0.1019	0.0760
T19	0.1881	0.1335	0.0419
T20	0.2472	0.2213	0.1588
T21	0.3057	0.0440	0.0176
T22	0.3642	0.3091	0.0176
T23	0.2830	0.2211	0.0773
T24	0.2829	0.1031	0.0405
T25	0.2467	0.0740	0.0172
T26	0.2829	0.1023	0.0766
T27	0.3416	0.2792	0.1358
T28	0.2464	0.1905	0.0178
T29	0.1639	0.0729	0.0172
T30	0.3647	0.1028	0.0406
T31	0.3053	0.2501	0.0771
T32	0.3052	0.1611	0.0997
T33	0.3418	0.1612	0.0998
T34	0.3415	0.2793	0.0994
T35	0.4005	0.1027	0.0406
T36	0.2820	0.1905	0.1353
T37	0.3055	0.1330	0.0412
T38	0.2468	0.2210	0.1945
T39	0.3644	0.3083	0.0997
T40	0.2828	0.2205	0.1943
T41	0.4002	0.3385	0.0403
T42	0.1291	0.1030	0.0770
T43	0.3054	0.2506	0.1597
T44	0.4229	0.1328	0.0772
T45	0.3645	0.1913	0.1354
T46	0.4227	0.0434	0.0174
T47	0.3056	0.2790	0.0179
T48	0.4003	0.1910	0.0180
T49	0.4822	0.0743	0.0174
T50	0.4595	0.0437	0.0176

T51	0.4003	0.3385	0.0770
T52	0.4004	0.0745	0.0171
T53	0.3416	0.1331	0.0771
T54	0.3647	0.3385	0.0404
T55	0.3410	0.2494	0.0401
T56	0.3641	0.2211	0.0774
T57	0.3054	0.1328	0.0772
T58	0.4004	0.1918	0.1000
T59	0.4593	0.1332	0.0415
T60	0.4228	0.2787	0.0178
T61	0.4228	0.3673	0.0407
T62	0.4594	0.3973	0.0180
T63	0.3417	0.1613	0.1353
T64	0.4004	0.2213	0.0774
T65	0.2469	0.2213	0.0772
T66	0.4821	0.1910	0.0181
T67	0.3053	0.1611	0.1351
T68	0.4000	0.3085	0.0176
T69	0.3418	0.1332	0.0411
T70	0.4004	0.2207	0.0407
T71	0.1883	0.1619	0.0179
T72	0.4004	0.1024	0.0768
T73	0.3646	0.3384	0.0771
T74	0.3416	0.1617	0.0179
T75	0.2468	0.2210	0.0404
T76	0.0706	0.0443	0.0175
T77	0.2464	0.1018	0.0766
T78	0.1056	0.0441	0.0170
T79	0.2825	0.1904	0.0180
T80	0.1284	0.0754	0.0172
T81	0.4594	0.2790	0.0179
T82	0.3642	0.1911	0.0180
T83	0.2239	0.1614	0.0999
T84	0.1656	0.1030	0.0405
T85	0.3645	0.1915	0.1001
T86	0.4230	0.3974	0.0179
T87	0.1879	0.1325	0.0775
T88	0.3643	0.2205	0.0407
T89	0.2459	0.1918	0.1355
T90	0.3056	0.2795	0.0996
T91	0.3056	0.2794	0.1357
T92	0.2831	0.2208	0.1585
T93	0.2231	0.1329	0.0415
T94	0.4227	0.2499	0.0404
T95	0.3406	0.2498	0.0770
T96	0.4597	0.1616	0.0180
T97	0.3054	0.1614	0.0180
T98	0.1883	0.1617	0.1357
T99	0.3647	0.1026	0.0769
T100	0.4229	0.1617	0.0179
T101	0.6592	0.7500	0.1592
T102	0.3052	0.2500	0.1948
T103	0.4595	0.2500	0.0405
T104	0.4228	0.2500	0.0772
O1	0.3605	0.2057	0.0293

O2	0.3608	0.0980	0.0592
O3	0.4708	0.4115	0.0221
O4	0.4630	0.1470	0.0294
O5	0.3232	0.1663	0.1382
O6	0.3024	0.1566	0.1175
O7	0.3232	0.2840	0.1387
O8	0.2862	0.1929	0.1171
O9	0.3231	0.2561	0.1556
O10	0.0877	0.0457	0.0185
O11	0.3235	0.0491	0.0210
O12	0.2057	0.1274	0.0800
O13	0.1256	0.0886	0.0296
O14	0.1467	0.0677	0.0225
O15	0.1763	0.1174	0.0802
O16	0.1838	0.1469	0.0887
O17	0.2436	0.1956	0.1174
O18	0.2351	0.1768	0.1391
O19	0.1182	0.0591	0.0217
O20	0.3825	0.0688	0.0211
O21	0.4111	0.1177	0.0805
O22	0.2273	0.1564	0.1176
O23	0.4126	0.1175	0.0378
O24	0.1779	0.1169	0.0382
O25	0.2648	0.1859	0.1393
O26	0.1681	0.0978	0.0589
O27	0.1684	0.0885	0.0281
O28	0.2431	0.2058	0.0879
O29	0.1471	0.1077	0.0379
O30	0.2940	0.0593	0.0216
O31	0.2355	0.1763	0.0962
O32	0.1470	0.1078	0.0788
O33	0.3607	0.3230	0.0291
O34	0.2432	0.2058	0.1472
O35	0.2649	0.0683	0.0210
O36	0.1850	0.1566	0.1174
O37	0.3022	0.2549	0.1770
O38	0.2273	0.1469	0.0883
O39	0.3828	0.1073	0.0384
O40	0.4625	0.3824	0.0295
O41	0.4196	0.3824	0.0290
O42	0.2648	0.2254	0.1557
O43	0.2056	0.1664	0.0965
O44	0.3028	0.2547	0.0590
O45	0.4039	0.3231	0.0287
O46	0.2938	0.2353	0.1982
O47	0.2853	0.2159	0.1769
O48	0.3535	0.1176	0.0801
O49	0.2848	0.2160	0.0592
O50	0.1267	0.0983	0.0588
O51	0.4207	0.3720	0.0591
O52	0.3014	0.2644	0.0288
O53	0.2356	0.1172	0.0802
O54	0.2647	0.1073	0.0385
O55	0.3527	0.1174	0.0376
O56	0.4114	0.0591	0.0218

O57	0.2432	0.0885	0.0290
O58	0.2350	0.1768	0.0216
O59	0.2861	0.2058	0.0891
O60	0.2940	0.2347	0.0374
O61	0.1836	0.1374	0.0591
O62	0.4411	0.0492	0.0207
O63	0.4033	0.3335	0.0588
O64	0.3607	0.2057	0.0885
O65	0.2341	0.1174	0.0376
O66	0.3538	0.0592	0.0217
O67	0.2943	0.2350	0.0804
O68	0.3448	0.2646	0.0297
O69	0.2858	0.2059	0.1466
O70	0.3828	0.1074	0.0790
O71	0.4707	0.0594	0.0219
O72	0.2947	0.2348	0.1555
O73	0.3232	0.1664	0.0970
O74	0.2057	0.1665	0.1383
O75	0.3617	0.3336	0.0589
O76	0.2865	0.0886	0.0294
O77	0.3615	0.3133	0.1175
O78	0.3232	0.2449	0.0382
O79	0.4627	0.2646	0.0290
O80	0.3826	0.3039	0.0213
O81	0.4039	0.0981	0.0590
O82	0.4045	0.2058	0.0884
O83	0.4410	0.1278	0.0383
O84	0.2058	0.0491	0.0204
O85	0.4203	0.1374	0.0591
O86	0.1765	0.0589	0.0218
O87	0.3529	0.2942	0.0956
O88	0.4710	0.1769	0.0219
O89	0.3441	0.2746	0.1175
O90	0.3015	0.2650	0.1465
O91	0.3232	0.1276	0.0793
O92	0.3525	0.2351	0.0809
O93	0.2943	0.1767	0.1394
O94	0.4194	0.2648	0.0293
O95	0.3015	0.1467	0.0885
O96	0.2650	0.2258	0.1967
O97	0.2056	0.1274	0.0372
O98	0.2649	0.1074	0.0789
O99	0.3233	0.1277	0.0385
O100	0.1839	0.1470	0.0286
O101	0.2279	0.1378	0.0592
O102	0.3452	0.2656	0.1516
O103	0.2941	0.1766	0.0218
O104	0.4041	0.3231	0.0884
O105	0.3531	0.1768	0.0219
O106	0.3021	0.1373	0.0592
O107	0.3528	0.2943	0.1393
O108	0.2939	0.1174	0.0804
O109	0.2941	0.1765	0.0958
O110	0.3531	0.2348	0.0374
O111	0.4116	0.1770	0.0219

O112	0.2862	0.2058	0.0291
O113	0.3530	0.2940	0.0221
O114	0.3608	0.3230	0.0884
O115	0.3825	0.3433	0.0381
O116	0.3826	0.2252	0.0792
O117	0.3606	0.0887	0.0291
O118	0.3016	0.2649	0.0887
O119	0.4209	0.2549	0.0590
O120	0.2946	0.1172	0.0375
O121	0.3525	0.1769	0.1393
O122	0.2647	0.1861	0.0965
O123	0.3033	0.2746	0.1174
O124	0.4115	0.2941	0.0221
O125	0.2648	0.1863	0.0213
O126	0.4409	0.3621	0.0376
O127	0.3445	0.1568	0.1175
O128	0.3441	0.2549	0.0589
O129	0.4046	0.0884	0.0292
O130	0.3451	0.1467	0.0886
O131	0.2858	0.0981	0.0591
O132	0.3013	0.1470	0.0293
O133	0.4113	0.3529	0.0808
O134	0.4409	0.2839	0.0213
O135	0.2443	0.2159	0.1767
O136	0.2444	0.2161	0.0590
O137	0.3611	0.1955	0.1174
O138	0.2348	0.0589	0.0217
O139	0.2276	0.1474	0.0296
O140	0.3615	0.2161	0.0592
O141	0.4412	0.1664	0.0210
O142	0.3449	0.2650	0.0881
O143	0.4032	0.2162	0.0591
O144	0.3530	0.1766	0.0961
O145	0.2438	0.0984	0.0589
O146	0.3826	0.1866	0.0974
O147	0.4118	0.2350	0.0809
O148	0.3230	0.2448	0.0802
O149	0.4115	0.2350	0.0372
O150	0.3230	0.2840	0.0217
O151	0.2431	0.2056	0.0297
O152	0.4116	0.3531	0.0370
O153	0.3826	0.3042	0.0965
O154	0.3826	0.2255	0.0381
O155	0.4411	0.2451	0.0385
O156	0.3824	0.3434	0.0798
O157	0.4045	0.2057	0.0292
O158	0.4408	0.4018	0.0213
O159	0.2648	0.2253	0.0794
O160	0.2646	0.2254	0.0381
O161	0.3825	0.1868	0.0209
O162	0.3452	0.1471	0.0292
O163	0.3448	0.1376	0.0592
O164	0.3231	0.2839	0.0967
O165	0.3232	0.1665	0.0208
O166	0.4707	0.2942	0.0222

O167	0.2057	0.1664	0.0209
O168	0.4191	0.1474	0.0290
O169	0.1762	0.1762	0.1394
O170	0.2347	0.2347	0.1985
O171	0.1170	0.1170	0.0370
O172	0.1763	0.1763	0.0955
O173	0.2344	0.2344	0.1550
O174	0.1171	0.1171	0.0803
O175	0.3525	0.3525	0.0808
O176	0.2938	0.2938	0.0223
O177	0.2346	0.2346	0.0809
O178	0.2942	0.2942	0.1395
O179	0.3524	0.3524	0.0370
O180	0.1765	0.1765	0.0218
O181	0.2347	0.2347	0.0373
O182	0.2940	0.2940	0.0956
O183	0.4113	0.4113	0.0221
O184	0.1570	0.0672	0.0000
O185	0.1118	0.0387	0.0000
O186	0.1250	0.0786	0.0000
O187	0.3032	0.2747	0.0000
O188	0.3614	0.3126	0.0000
O189	0.3445	0.2744	0.0000
O190	0.0701	0.0444	0.0000
O191	0.4791	0.4312	0.0000
O192	0.3599	0.0784	0.0000
O193	0.2865	0.0784	0.0000
O194	0.4615	0.3926	0.0000
O195	0.4625	0.0390	0.0000
O196	0.4205	0.2748	0.0000
O197	0.3469	0.0387	0.0000
O198	0.4210	0.3926	0.0000
O199	0.2266	0.1566	0.0000
O200	0.2441	0.1954	0.0000
O201	0.4775	0.0784	0.0000
O202	0.4619	0.2751	0.0000
O203	0.4025	0.3129	0.0000
O204	0.3006	0.0389	0.0000
O205	0.4792	0.3129	0.0000
O206	0.2423	0.0781	0.0000
O207	0.4792	0.1957	0.0000
O208	0.3616	0.1954	0.0000
O209	0.2282	0.0390	0.0000
O210	0.3031	0.1566	0.0000
O211	0.4071	0.0791	0.0000
O212	0.3443	0.1565	0.0000
O213	0.1854	0.1568	0.0000
O214	0.2849	0.1956	0.0000
O215	0.4616	0.1567	0.0000
O216	0.4206	0.1565	0.0000
O217	0.4185	0.0387	0.0000
O218	0.4030	0.1958	0.0000
O219	0.1837	0.0391	0.0000
O220	0.0664	0.0300	0.0300
O221	0.1103	0.0299	0.0299

O222	0.1838	0.1473	0.1473
O223	0.2277	0.1479	0.1479
O224	0.1256	0.0884	0.0884
O225	0.2431	0.2058	0.2058
O226	0.2868	0.2060	0.2060
O227	0.1841	0.0294	0.0294
O228	0.1689	0.0889	0.0889
O229	0.3015	0.1474	0.1474
O230	0.3011	0.0299	0.0299
O231	0.2277	0.0293	0.0293
O232	0.3454	0.0295	0.0295
O233	0.4192	0.0297	0.0297
O234	0.3451	0.1472	0.1472
O235	0.2430	0.0887	0.0887
O236	0.2865	0.0886	0.0886
O237	0.4629	0.0292	0.0292
O238	0.3606	0.0887	0.0887
O239	0.4048	0.0886	0.0886
O240	0.4420	0.5237	0.4420

Table S4. Numbers of seven different types of cages per unit cell in the RHO- G_n members of the RHO family of zeolites ($n = 1 - 8$).^[a]

	n	[4 ¹² 6 ⁸ 8 ⁶] <i>lta</i>	[4 ⁸ 8 ²] <i>d8r</i>	[4 ¹² 8 ⁶] <i>pau</i>	[4 ⁶ 6 ² 8 ⁶] <i>t-plg</i>	[4 ⁵ 8 ³] <i>t-oto</i>	[4 ⁶ 8 ⁴] <i>t-gsm</i>	[4 ⁷ 8 ⁵] <i>t-phi</i>
RHO-G1 (zeolite Rho)	1	2	6	0	0	0	0	0
RHO-G2	2	2	12	6	8	24	0	0
RHO-G3 (paulingite)	3	2	18	12	16	72	12	24
RHO-G4 (ZSM-25)	4	2	24	18	24	144	60	72
RHO-G5 (PST-20)	5	2	30	24	32	240	168	144
RHO-G6 (PST-25)	6	2	36	30	40	360	360	240
RHO-G7 (PST-26)	7	2	42	36	48	504	660	360
RHO-G8 (PST-28)	8	2	48	42	56	672	1092	504

[a] The number of cages was calculated according to the same rules given in ref. S3:
 $N_{lta} = 2$; $N_{d8r} = 6n$; $N_{pau} = 6(n-1)$; $N_{t-plg} = 8(n-1)$; $N_{t-oto} = 12n(n-1)$; $N_{t-gsm} = 2(n-1)(n-2)(2n-3)$; $N_{t-phi} = 12(n-1)(n-2)$.