Supporting Information

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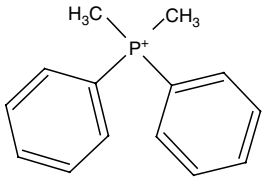
SI Text

SDAs Synthesis. Diphenyldimethylphosphonium hydroxide (Me_2Ph_2POH) was obtained by methylation of diphenylphosphine with methyl iodide in chloroform in the presence of K_2CO_3 as previously described for the synthesis of ITQ-27.

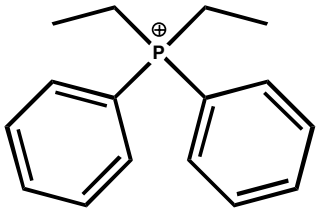


Diphenylphosphine, 10.80 g (0.058 mol), was dissolved in 50 mL of chloroform under nitrogen atmosphere. Then, 9.55 g of potassium carbonate sesquihydrate were added, and the mixture was stirred. Finally, 24.60 g (0.173 mol) of methyl iodide were added dropwise. After 48 hours, 8 g of methyl iodide were added again, and the mixture was left for a total time of five days.

The isopropanol was then eliminated and the solid washed with diethyl ether. Next, the product was dissolved in chloroform. The chloroform was evaporated, and the solid washed with diethyl ether and then dried under vacuum to give 18.426 g of diphenyldimethylphosphonium iodide (93.2% wt yield).

Next, 18.426 g of diphenyldimethylphosphonium iodide, previously dissolved in water, were converted to the corresponding hydroxide with 58.15 g of an anionic exchange resin in batch overnight, yielding 183.52 g of a 0.27 M solution of diphenyldimethylphosphonium hydroxide (92% of exchange yield) that was used as the structure directing agent (SDA) source.

Diphenyldiethylphosphonium hydroxide (Et_2Ph_2POH) was obtained by ethylation of diphenylphosphine with ethyl iodide in ethanol in the presence of K_2CO_3.



Diphenylphosphine, 50 g (0.27 mol), was mixed with 150 mL of ethanol under nitrogen atmosphere. Then, 37.12 g of potassium carbonate were added, and the mixture was stirred. Finally, 125.73 g (0.81 mol) of ethyl iodide were added dropwise. After 48 hours at 50 °C, the ethanol was eliminated and the solid washed with diethyl ether. The product was then dissolved in chloroform. The chloroform was evaporated and the solid washed with diethyl ether and then dried under vacuum to give 98.45 g of diphenyldiethylphosphonium iodide (98.5% wt yield).

Next, 98.45 g of diphenyldiethylphosphonium iodide, previously dissolved in water, were converted to the corresponding hydroxide with 270 g of an anionic exchange resin in batch overnight, yielding 271.75 g of a 0.94 M solution of diphenyldiethylphosphonium hydroxide (96% of exchange yield) that was used as the SDA source.

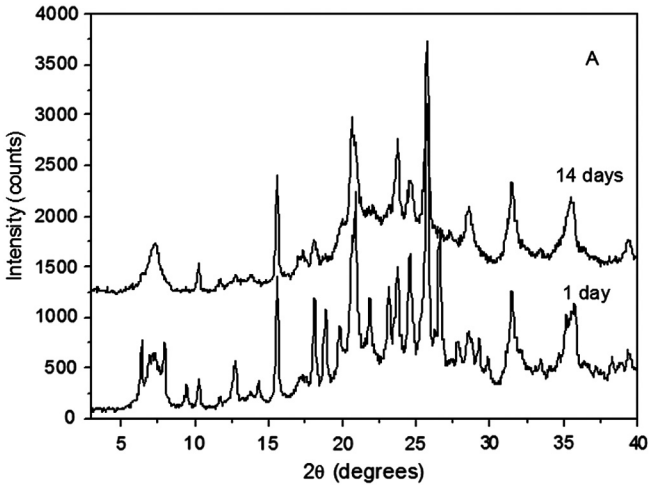


Fig. S1. X-ray diffraction patterns from gels with a Si/Ge = 5 using (*A*) Et_2Ph_2POH as the SDA.

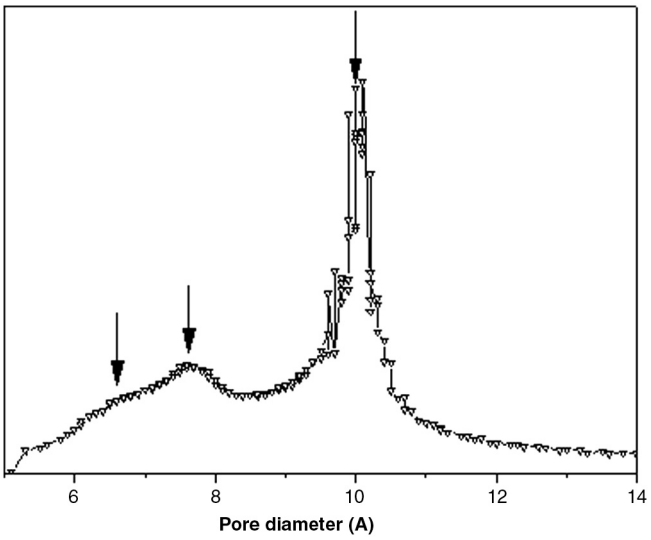


Fig. S2. Pore size distribution of Instituto de Tecnologia Química number 40 (ITQ-40) zeolite.

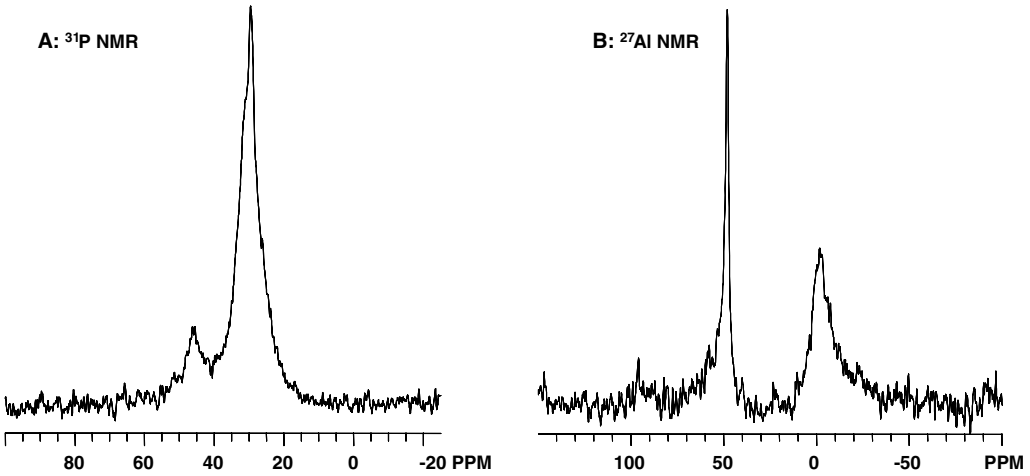


Fig. S3. (*A*) ^{31}P and (*B*) ^{27}Al magic angle spinning (MAS) NMR of as-synthesized ITQ-40.

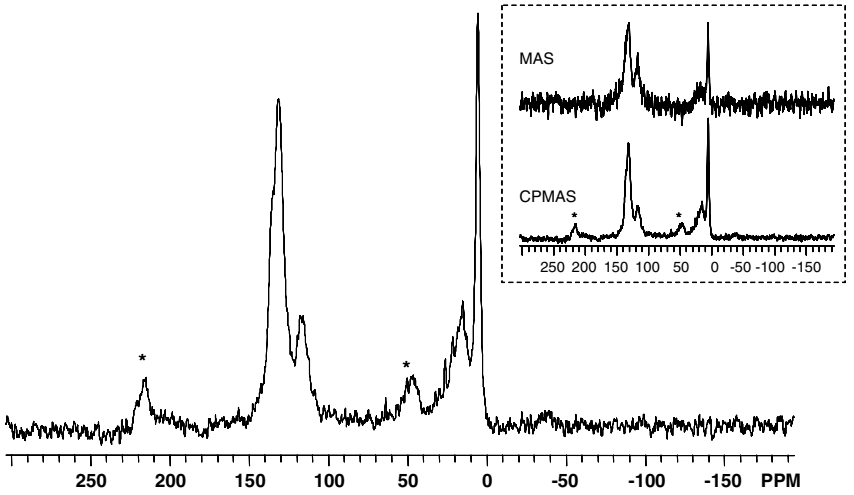


Fig. S4. ^{13}C NMR (50 MHz) of as-synthesized ITQ-40. Peak assignments (CH_3 at 5.6 ppm, -CH_3- centered around 21 ppm, protonated aromatics centered around 132 ppm, and the quaternary aromatic at 115 ppm) and integration of the MAS spectrum are consistent with the Et_2Ph_2POH SDA used in the synthesis, indicating that the SDA remained intact. (*Inset*) ^{13}C cross-polarized magic angle spinning (CPMAS) (acquired using 4.3-kHz spinning speed and 1-ms contact time) and Bloch decay (acquired using 8-kHz spinning speed and 90-s pulse repetition delay) spectra and the spinning sidebands are shown with asterisks.

Table S1. Final atomic coordinates from Rietveld refinement of calcined ITQ-40

Atom	No.	x	y	z	U_{iso}	Occupancy
Si,Ge	1	0.4716(7)	0.3314(7)	0.0335(5)	0.046(8)	0.574*
Si,Ge	2	0.3660(9)	0.4363(9)	0.0628(4)	0.046(8)	0.574*
Si,Ge	3	0.2266(5)	0.4531(9)	0.1208(5)	0.046(8)	0.574*
Si,Ge	4	0.2728(5)	0.5456(11)	0.2045(5)	0.046(8)	0.574*
Si,Ge	5	0.3333	0.6667	0.0788(9)	0.046(8)	0.574*
O	11	0.4659(39)	0.2329(20)	0.0515(9)	0.178(37)	1
O	12	0.5819(18)	0.4181(18)	0.0415(20)	0.178(37)	1
O	13	0.4501(44)	0.3237(25)	0.9823(7)	0.178(37)	1
O	14	0.4046(42)	0.3595(35)	0.0624(15)	0.178(37)	1
O	15	0.2907(17)	0.4087(17)	0.1022(7)	0.178(37)	1
O	16	0.4623(27)	0.5377(27)	0.0753(16)	0.178(37)	1
O	17	0.2270(9)	0.4540(17)	0.1724(5)	0.178(37)	1
O	18	0.2809(2)	0.5618(4)	0.1019(9)	0.178(37)	1
O	19	0.2449(11)	0.4898(22)	0.2500	0.178(37)	1
O	20	0.2219(13)	0.6110(6)	0.2001(11)	0.178(37)	1
O	21	0.3333	0.6667	0.0285(9)	0.178(37)	1

Estimated standard deviations for the last digit are given in the parentheses. Atoms 1 and 2 are in the double four-ring, atom 4 is in the double three-ring, and atoms 3 and 5 are in the $[6^4 5^3 4^3 3]$ cage
*Occupancy is for Si; the remainder, 0.426, is for Ge.

Table S2. Interatomic distances and angles from Rietveld refinement of calcined ITQ-40

SI1-O11	1.678(6)	O11-SI1-O12	106.6(15)
SI1-O12	1.676(6)	O11-SI1-O13	110.9(15)
SI1-O13	1.677(6)	O11-SI1-O14	110.4(13)
SI1-O14	1.675(6)	O12-SI1-O13	108.2(16)
		O12-SI1-O14	105.2(15)
SI2-O13	1.676(6)	O13-SI1-O14	115.0(14)
SI2-O14	1.677(6)		
SI2-O15	1.669(6)	O13-SI2-O14	115.8(16)
SI2-O16	1.678(6)	O13-SI2-O15	113.1(15)
		O13-SI2-O16	108.8(20)
SI3-O15	1.668(4)	O14-SI2-O15	108.1(10)
SI3-O15	1.668(4)	O14-SI2-O16	103.1(20)
SI3-O17	1.663(6)	O15-SI2-O16	107.1(15)
SI3-O18	1.665(6)		
		O15-SI3-O15	117.0(23)
SI4-O17	1.666(6)	O15-SI3-O17	111.2(10)
SI4-O19	1.666(6)	O15-SI3-O18	102.9(9)
SI4-O20	1.669(4)	O15-SI3-O17	111.2(10)
SI4-O20	1.669(4)	O15-SI3-O18	102.9(9)
		O17-SI3-O18	111.0(17)
SI5-O18	1.669(6)		
SI5-O18	1.669(6)	O17-SI4-O19	99.8(8)
SI5-O18	1.669(6)	O17-SI4-O20	112.7(12)
SI5-O21	1.619(6)	O17-SI4-O20	112.6(12)
		O19-SI4-O20	110.0(12)
SI1-O11-SI1	139.3(21)	O19-SI4-O20	110.0(12)
SI1-O12-SI1	150.9(26)	O20-SI4-O20	111.1(19)
SI1-O13-SI2	152.7(20)		
SI1-O14-SI2	139.8(17)	O18-SI5-O18	101.8(3)
SI2-O15-SI3	134.4(16)	O18-SI5-O18	101.8(3)
SI2-O16-SI2	151.9(32)	O18-SI5-O21	116.4(3)
SI3-O17-SI4	128.8(18)	O18-SI5-O18	101.8(3)
SI3-O18-SI5	175.0(12)	O18-SI5-O21	116.4(3)
SI4-O19-SI4	123.0(22)	O18-SI5-O21	116.4(3)
SI4-O20-SI4	127.4(19)		

T-O distances are in Å and T-O-T and O-T-O angles are in degrees. Estimated standard deviations for the last digit are given in the parentheses.