Supporting Information

Syntheses, structure solutions, and catalytic performance of two novel layered silicates

Jie Liang, Jie Su,* Yanping Chen, Zhaofei Li, Kuo Li, Hao Zhang, Xiaodong Zou, Fuhui Liao, Yingxia Wang,* and Jianhua Lin*

List of Content

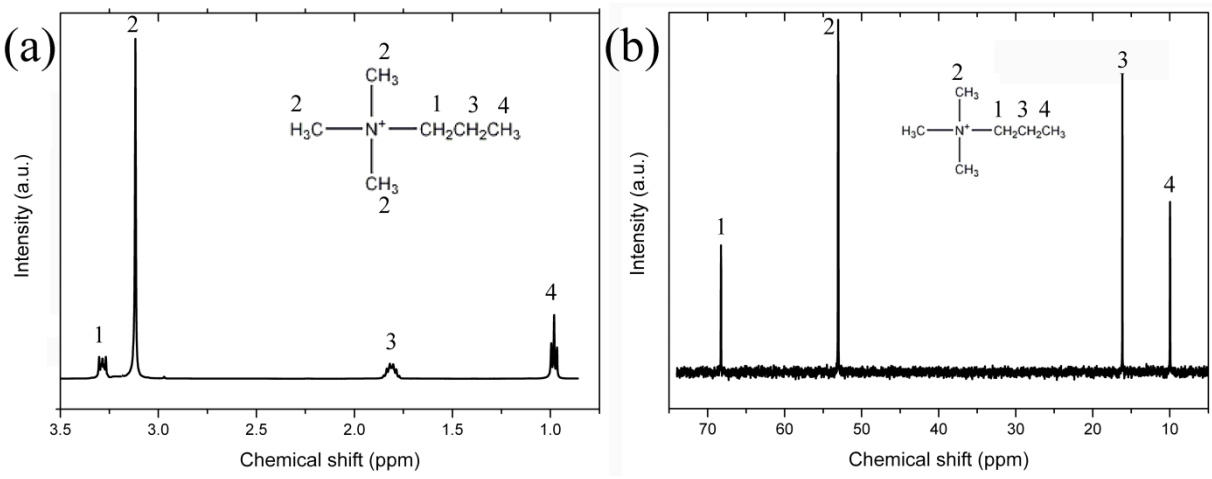

Figure S1. (a) ^{1}H and (b) ^{13}C NMR spectra of *n*-propyltrimethylammonium iodide.

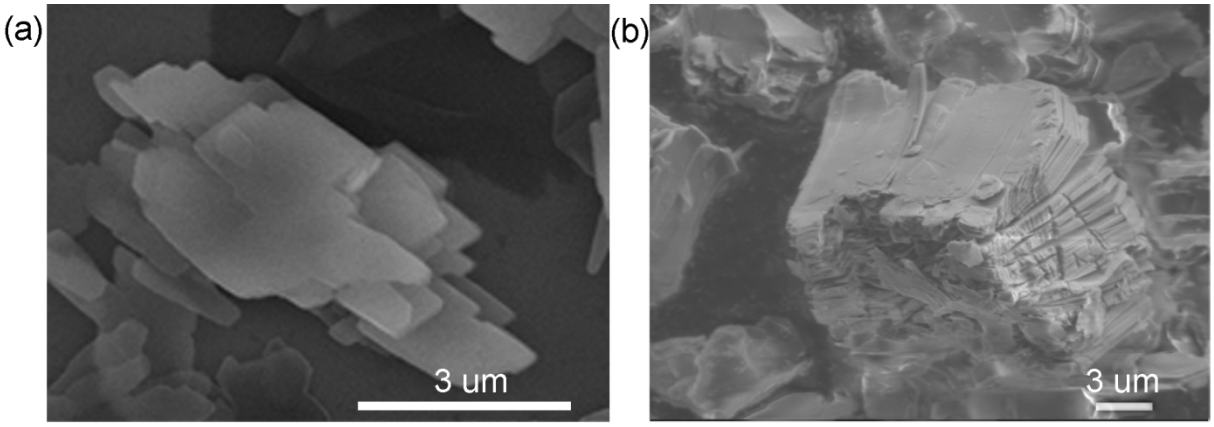

Figure S2. SEM images of PKU-13 and PKU-13a. Both PKU-13 and PKU-13a crystallize as aggregates of colorless crystals with plate-like morphology.

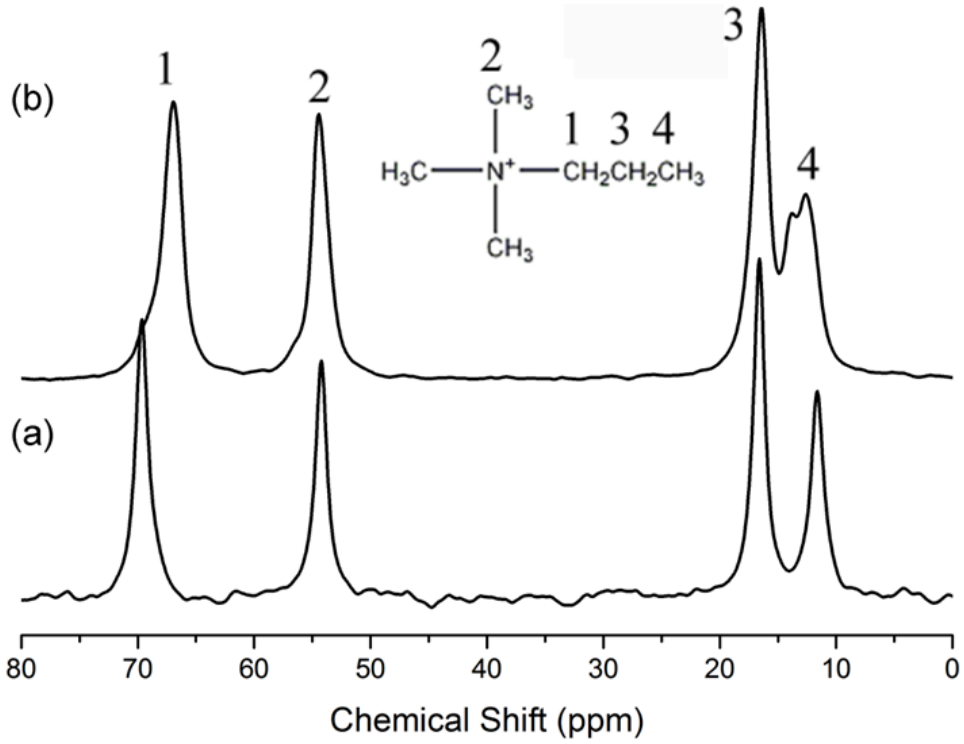

Figure S3. Solid state ^{13}C MAS NMR spectra of *n*-propyltrimethylammonium in (a) PKU-13 and (b) PKU-13a.

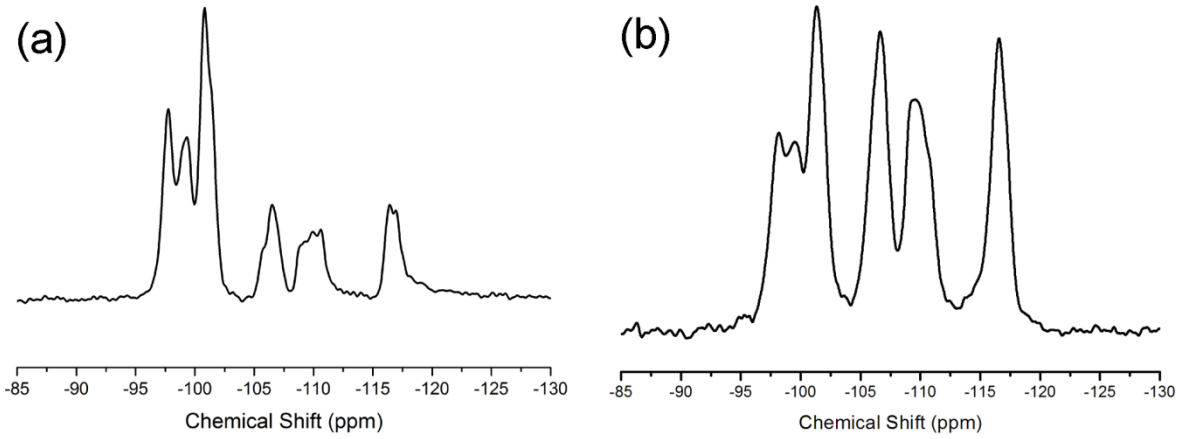

Figure S4. Comparison of (a) ^{1}H-^{29}Si cross-polarization (CP) NMR and (b) quantitative ^{29}Si NMR spectra of PKU-13. The contact time is 3 ms for the ^{1}H-^{29}Si NMR CP measurement.

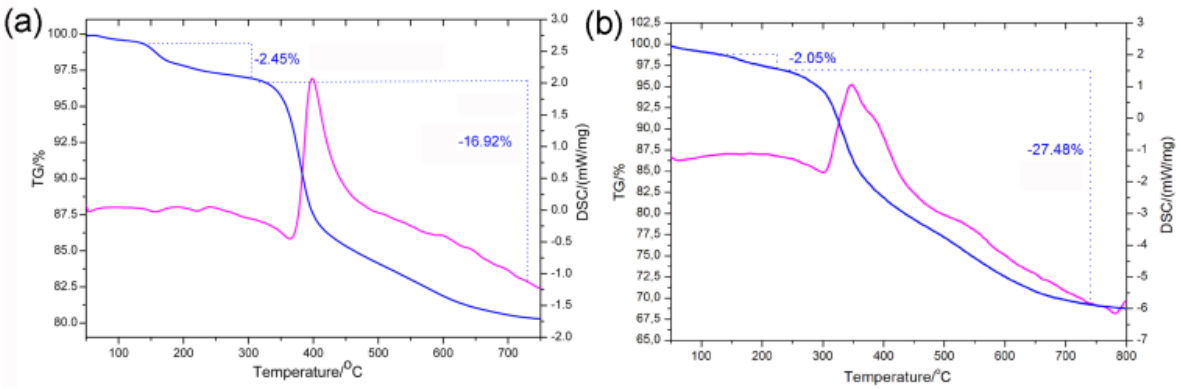

Figure S5. Thermogravimetric (TG) curves of PKU-13 and PKU-13a. In both TG curves of PKU-13 and PKU-13a, the first weight losses of 2.45% and 2.05% between 150 °C and 300 °C are due to the removal of one water molecule from the interlayer region (calcd. 2.24%, 2.09%). In the range of 320 °C and 700 °C, the dramatic weight losses of 16.92% and 27.48% correspond to the removal of the organic amine (calcd. 12.82%, 24.05%) plus one half water molecules from the dehydration of hydroxyls (calcd. 3.35% and 3.14%). After the experiments, the solids changed from white to black.

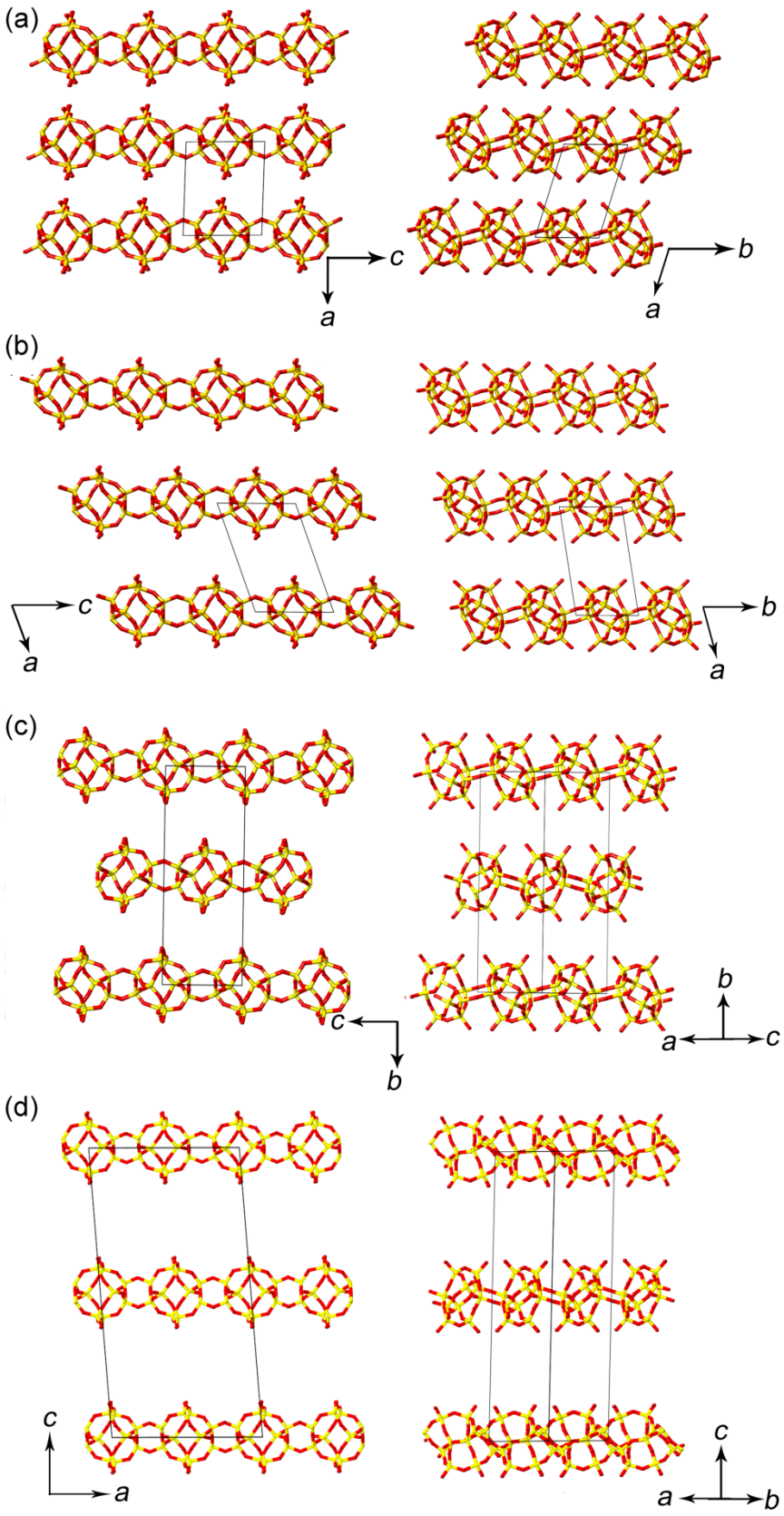

Figure S6. Framework comparison among (a) PKU-13, (b) PKU-13a, (c) HUS-2 and (d) HUS-7. The Si atoms are drawn in yellow and O atoms in red.

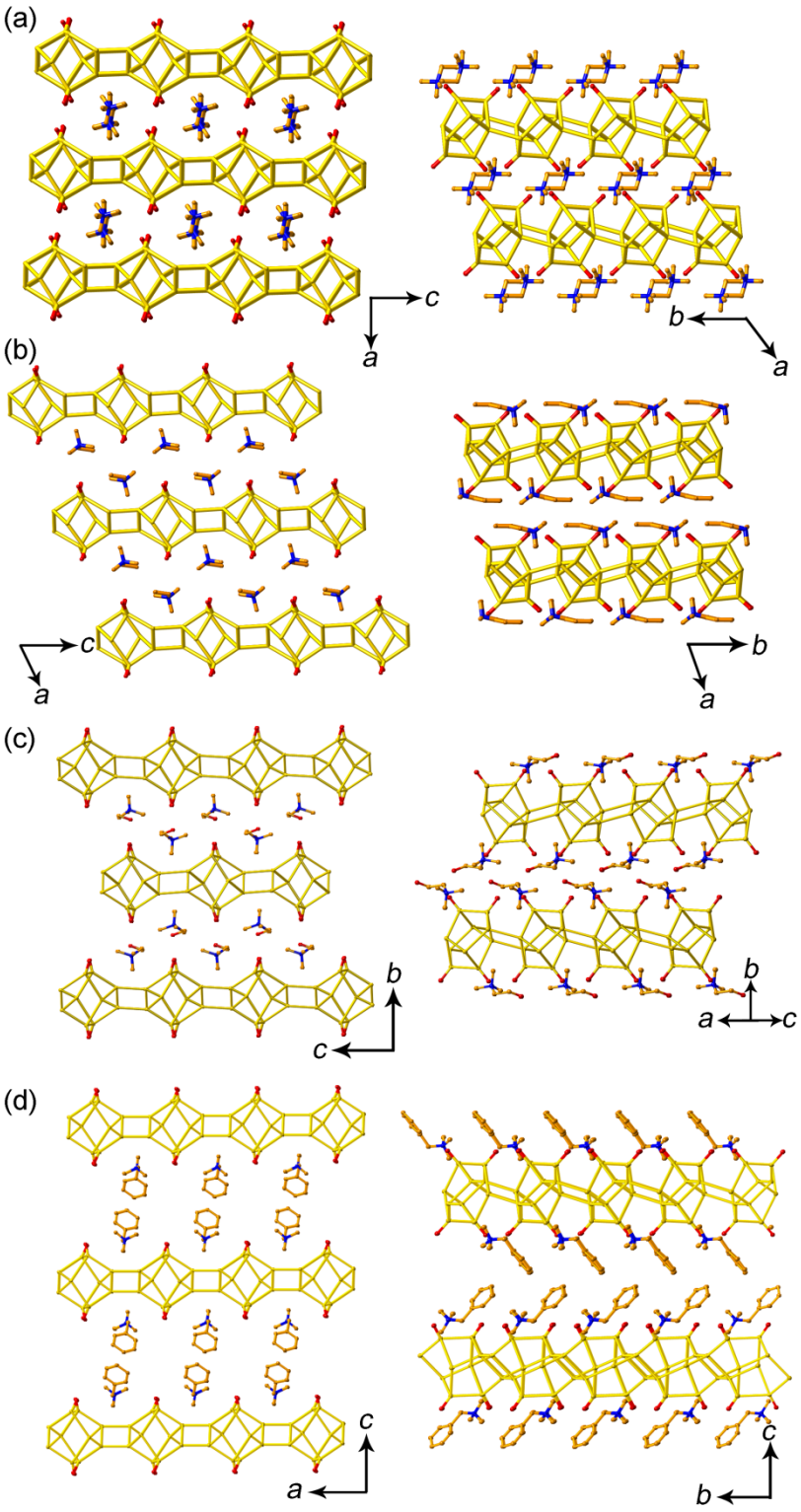

Figure S7. The configurations of different organic cations in (a) PKU-13, (b) PKU-13a, (c) HUS-2, and (d) HUS-7. The organic cations are propyltrimethylammonium, choline, and benzyltrimethylammonium in (a) and (b), (c), and (d), respectively. Silicon: yellow, oxygen: red, Carbon: orange, Nitrogen: blue.

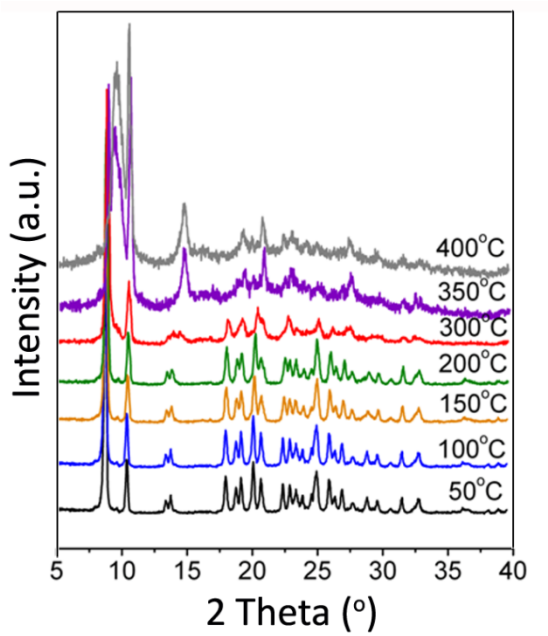

Figure S8. *In-situ* PXRD of PKU-13 heated from 50˚C to 400˚C.

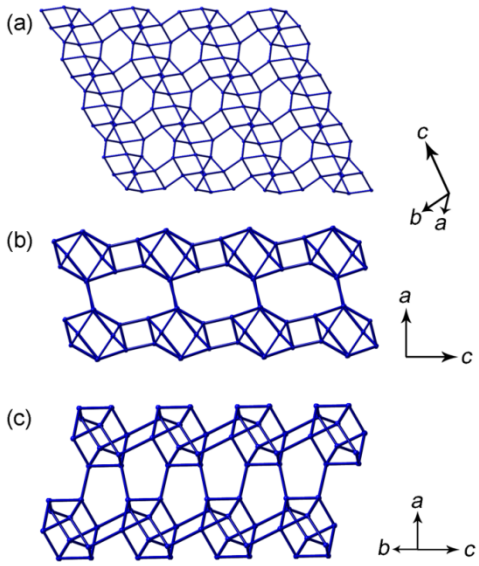

Figure S9. Crystal structure model of the hypothetical zeolite derived from r52 layers. The adjacent layers in the hypothetical zeolite are stacked in the same sequence as those in PKU-13.

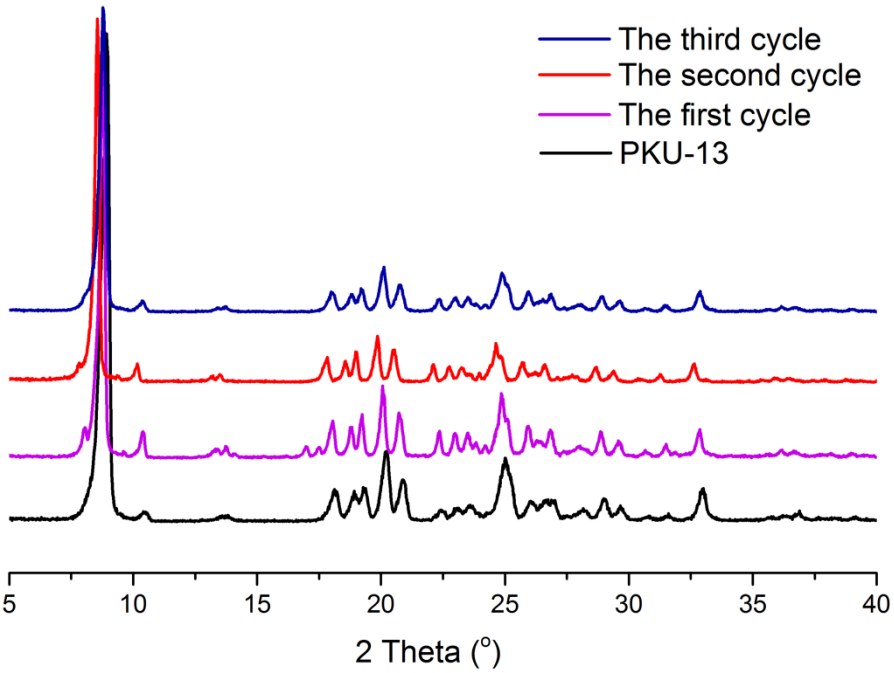

Figure S10. PXRD patterns of the PKU-13 samples after each catalytic recycle.

Table 1. Synthetic conditions for the novel layered silicates.

Sample	Synthesis conditions				Product
	SDA/SiO_2	H_2O/SiO_2	Tem.(°C)	Time(d)	
1	0.35	10	155	30	NON
2	0.35	15	155	30	NON
3	0.35	20	155	30	NON
4	0.5	12	155	30	**amorphous**
5	0.5	15	155	30	**PKU-13**
6	0.5	17	155	30	**PKU-13**
7	0.5	20	155	30	NON
8	0.75	10	155	30	amorphous
9	0.75	15	155	30	**PKU-13a**
10	0.75	20	155	30	amorphous

Table S2. The CHN analysis of PKU-13, PKU-13a and HAc treated PKU-13a.

Sample		C (wt%)	N (wt%)	H (wt%)
PKU-13	found	10.07	2.06	2.75
	calculated	9.51	1.85	2.26
After 1st catalytic cycle		9.67	2.03	2.63
After 2nd catalytic cycle		9.41	1.89	2.43
After 3rd catalytic cycle		9.28	1.75	2.47
PKU-13a	found	16.12	3.30	3.97
	calculated	16.79	3.26	4.00
HAc treated PKU-13a	found	6.41	0.89	2.00

Table 3. Atomic coordinates, isotropic displacement parameters (B_{iso}) and occupancies of PKU-13 obtained from Rietveld refinements.

Atom	*x*	*y*	*z*	B_{iso}	Occupancy
Si1	0.8850 (4)	0.3973(5)	0.1678(4)	0.9(1)	1
Si2	0.9006(4)	0.2055(5)	0.8226(4)	0.9(1)	1
Si3	0.2688(4)	0.5058(5)	0.5365(4)	0.9(1)	1
Si4	0.9850(4)	0.8058(5)	0.6724(4)	0.9(1)	1
Si5	0.7505(4)	0.1170(5)	0.5141(4)	0.9(1)	1
O1	0.9816(4)	0.3024(5)	0.2205(4)	1.44(2)	1
O2	0.7561(4)	0.4071(1)	0.2747(4)	1.44(2)	1
O3	0.8336(6)	0.2574(1)	0.9904(4)	1.44(2)	1
O4	0.0335(5)	0.3781(5)	0.8211(8)	1.44(2)	1
O5	0.3865(6)	0.4029(9)	0.5007(1)	1.44(2)	1
O6	0.1325(4)	0.3446(9)	0.4531(6)	1.44(2)	1
O7	0.1145(3)	0.8813(1)	0.5818(5)	1.44(2)	1
O8	0.9405(5)	0.0009(5)	0.7877(9)	1.44(2)	1
O9	0.7905(4)	0.1669(9)	0.6948(4)	1.44(2)	1
O10	0.6413(6)	0.9040(7)	0.4336(1)	1.44(2)	1
O11	0.7003(5)	0.3035(4)	0.5162(5)	1.44(2)	1
N	0.43014	0.30427	-0.00230	3.00(8)	0.5
C1	0.32970	0.33115	-0.08624	3.00(8)	0.5
C2	0.39106	0.32659	0.1584517	3.00(8)	0.5
C3	0.43196	0.09251	-0.07666	3.00(8)	0.5
C4	0.56734	0.42335	-0.02764	3.00(8)	0.5
C5	0.56590	0.62512	-0.03188	3.00(8)	0.5
C6	0.70170	0.76619	0.01012	3.00(8)	0.5

Table 4. Selected bond lengths and angles of PKU-13.

	Bond length / Å		**Bond length / Å**
Si1-O1	1.603(7)	Si3-O6	1.605(5)
Si1-O2	1.625(6)	Si3-O11	1.618(6)
Si1-O3	1.604(5)	Si4-O1	1.615(6)
Si1-O4	1.615(5)	Si4-O6	1.603(5)
Si2-O3	1.608(5)	Si4-O7	1.622(6)
Si2-O4	1.610(5)	Si4-O8	1.618(6)
Si2-O8	1.615(6)	Si5-O7	1.623(5)
Si2-O9	1.616(6)	Si5-O9	1.630(5)
Si3-O2	1.641(5)	Si5-O10	1.604(5)
Si3-O5	1.603(7)	Si5-O11	1.627(6)

	Angle / °		**Angle / °**
O1-Si1-O2	112.5(4)	O8-Si4-O6	109.4(4)
O1-Si1-O3	108.7(4)	O7-Si4-O1	110.6(4)
O1-Si1-O4	108.7(3)	O7-Si4-O6	109.5(3)
O2-Si1-O3	109.7(4)	O1-Si4-O6	111.0(4)
O2-Si1-O4	107.4(3)	O7-Si5-O9	106.4(3)
O3-Si1-O4	109.8(4)	O7-Si5-O10	109.9(4)
O3-Si2-O4	112.9(4)	O7-Si5-O11	108.4(4)
O3-Si2-O8	106.4(5)	O9-Si5-O10	113.7(5)
O3-Si2-O9	107.2(3)	O9-Si5-O11	106.7(3)
O4-Si2-O8	107.6(3)	O10-Si5-O11	111.4(4)
O4-Si2-O9	112.3(4)	Si1-O1-Si4	150.8(3)
O8-Si2-O9	110.4(4)	Si1-O2-Si3	135.2(3)
O5-Si3-O6	111.2(3)	Si1-O3-Si2	135.1(4)
O5-Si3-O11	112.7(4)	Si2-O4-Si1	153.6(4)
O5-Si3-O2	110.6(5)	Si3-O6-Si4	164.0(4)
O6-Si3-O11	109.4(4)	Si5-O7-Si4	160.3(6)
O6-Si3-O2	106.5(3)	Si2-O8-Si4	152.4(6)
O11-Si3-O2	106.2(4)	Si2-O9-Si5	149.1(4)
O8-Si4-O7	108.7(4)	Si3-O11-Si5	145.5(4)
O8-Si4-O1	107.6(4)		

Table 5. Atomic coordinates, isotropic displacement parameters (B_{iso}) and occupancies of PKU-13a obtained from Rietveld refinements.

Atom	*x*	*y*	*z*	B_{iso}	Occupancy
Si1	0.9063(4)	0.3973(5)	0.2214(6)	1.0(2)	1
Si2	0.9189(4)	0.2055(5)	0.8623(5)	1.0(2)	1
Si3	0.2293(4)	0.5058(5)	0.4216(6)	1.0(2)	1
Si4	0.9856(4)	0.8058(5)	0.6775(5)	1.0(2)	1
Si5	0.7907(4)	0.1170(5)	0.6290(3)	1.0(2)	1
O1	0.9922(4)	0.3307(5)	0.2324(5)	1.4(3)	1
O2	0.7924(4)	0.5702(1)	0.3815(4)	1.4(3)	1
O3	0.8649(4)	0.3708(9)	0.0633(4)	1.4(3)	1
O4	0.0363(4)	0.3357(5)	0.8030(8)	1.4(3))	1
O5	0.3220(5)	0.1303(8)	0.3380(1)	1.4(3)	1
O6	0.1108(3)	0.2603(10)	0.4023(6)	1.4(3)	1
O7	0.0999(4)	0.8029(1)	0.5365(6)	1.4(3)	1
O8	0.9404(7)	0.0300(5)	0.8154(7)	1.4(3)	1
O9	0.8328(3)	0.3196(9)	0.7838(4)	1.4(3)	1
O10	0.6990(4)	0.1464(9)	0.6225(1)	1.4(3)	1
O11	0.7388(5)	0.5110(5)	0.6534(6)	1.4(3)	1
N	0.65416	-0.03478	0.18141	7.0(8)	1
C1	0.58716	0.13489	0.33302	7.0(8)	1
C2	0.63742	-0.00876	0.03954	7.0(8)	1
C3	0.77470	-0.04518	0.16019	7.0(8)	1
C4	0.62568	-0.21603	0.20416	7.0(8)	1
C5	0.60677	-0.35314	0.05175	7.0(8)	1
C6	0.60439	-0.55845	0.05748	7.0(8)	1

Table 6. Selected bond lengths and angles of PKU-13a.

	Bond length / Å		**Bond length / Å**
Si1-O1	1.589(8)	Si3-O6	1.618(7)
Si1-O2	1.618(6)	Si3-O11	1.627(7)
Si1-O3	1.636(7)	Si4-O1	1.617(8)
Si1-O4	1.604(7)	Si4-O6	1.609(8)
Si2-O3	1.628(5)	Si4-O7	1.609(7)
Si2-O4	1.605(7)	Si4-O8	1.603(5)
Si2-O8	1.593(6)	Si5-O7	1.614(6)
Si2-O9	1.607(8)	Si5-O9	1.624 (7)
Si3-O2	1.620(6)	Si5-O10	1.610(8)
Si3-O5	1.591(7)	Si5-O11	1.625(7)

	Angle / °		**Angle / °**
O1-Si1-O2	113.8(4)	O8-Si4-O6	107.5(5)
O1-Si1-O3	109.9(4)	O7-Si4-O1	110.5(4)
O1-Si1-O4	111.4(4)	O7-Si4-O6	110.2(4)
O2-Si1-O3	105.6(4)	O1-Si4-O6	110.2(4)
O2-Si1-O4	108.0(4)	O7-Si5-O9	106.6(4)
O3-Si1-O4	107.9(5)	O7-Si5-O10	109.5(4)
O3-Si2-O4	108.4(4)	O7-Si5-O11	111.2(4)
O3-Si2-O8	107.4(5)	O9-Si5-O10	109.1(6)
O3-Si2-O9	108.6(3)	O9-Si5-O11	108.4(4)
O4-Si2-O8	109.8(4)	O10-Si5-O11	111.9(5)
O4-Si2-O9	110.0(5)	Si1-O1-Si4	148.0(4)
O8-Si2-O9	112.5(5)	Si1-O2-Si3	132.4(4)
O5-Si3-O6	109.4(4)	Si1-O3-Si2	138.8(4)
O5-Si3-O11	114.6(5)	Si2-O4-Si1	144.7(4)
O5-Si3-O2	110.6(6)	Si3-O6-Si4	161.3(4)
O6-Si3-O11	108.7(5)	Si5-O7-Si4	157.3(6)
O6-Si3-O2	106.7(4)	Si2-O8-Si4	149.6(5)
O11-Si3-O2	106.6(4)	Si2-O9-Si5	152.1(3)
O8-Si4-O7	110.2 (4)	Si3-O11-Si5	140.0(4)
O8-Si4-O1	108.2(4)		