Supporting information

High quality synthesis of nanosized CHA zeolite by combination of starting FAU zeolite and aluminum sources

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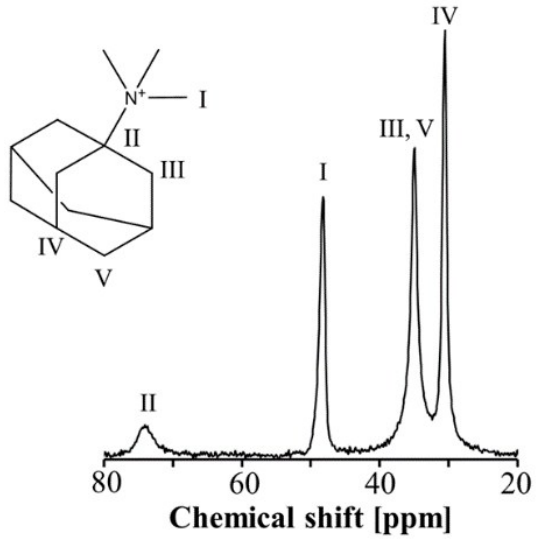

Figure S1. ^{13}C CP MAS NMR spectrum of CHA zeolite obtained using HSY+Al with Si/Al ratio of 20 (sample no. 18).

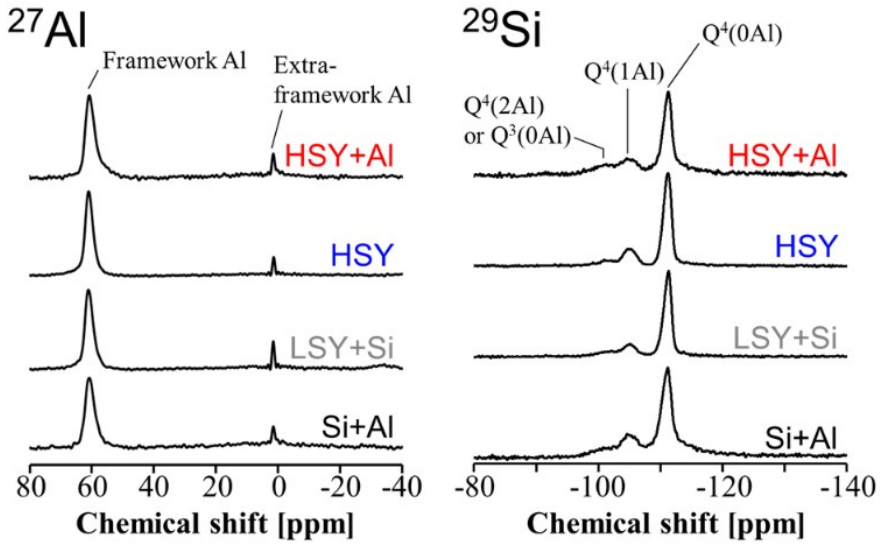

Figure S2. (left) ^{27}Al and (right) ^{29}Si MAS NMR spectra of the synthesized CHA zeolites (sample nos. 3, 9, 14, and 18, H-form).

Table S1 Synthesis conditions and characteristics of products obtained from various starting materials at different synthesis times[a]

Sample no.	Synthesis conditions								Product		
	Total Si/Al	Starting materials	Composed silica/alumina sources					Time [h]	Phase	Yield [%]	Si/Al[b]
			FAU			Fumed silica [Si mol%]	$Al(OH)_3$ [Al mol%]				
			Si/Al[b]	[Si mol%][c]	[Al mol%][d]						
21	20	Si+Al	-	-	-	100	100	1	Amorphous, $Al(OH)_3$	100	20
22	20		-	-	-	100	100	3	Amorphous, $Al(OH)_3$	77	13
23	20		-	-	-	100	100	4	CHA, $Al(OH)_3$	18	4.5
24	20		-	-	-	100	100	6	CHA, $Al(OH)_3$	91	14
25	20		-	-	-	100	100	8	CHA, $Al(OH)_3$	100	21
26	20		-	-	-	100	100	10	CHA	100	21
27	20	LSY+Si	2.8	14.0	100	86.0	-	2	FAU	11	7.9
28	20			14.0	100	86.0	-	4	FAU, CHA	35	-
29	20			14.0	100	86.0	-	6	FAU, CHA	83	22
30	20			14.0	100	86.0	-	12	FAU, CHA	80	23
31	20			14.0	100	86.0		14	CHA	84	23
32	26	HSY	26	100	100	-	-	2	FAU, Amorphous	51	18
33	26		26	100	100			3	FAU, Amorphous, CHA	16	18
34	26		26	100	100	-	-	6	FAU, Amorphous, CHA	61	18
35	26		26	100	100	-	-	8	CHA	68	20
36	20	HSY+Al	93	100	21.4	-	78.6	1	Amorphous, $Al(OH)_3$, FAU	27	7.5
37	20			100	21.4	-	78.6	2	CHA, $Al(OH)_3$	13	2.9
38	20			100	21.4	-	78.6	3	CHA	100	22

[a] Synthesis conditions: OSDA(*N*,*N*,*N*-trimethyladamanthylammonium hydroxide)/Si = 0.2, NaOH/Si = 0.1, H_2O/Si = 5, CHA seed (Si/Al = 16) = 2 wt.%, Temp = 140 °C.

[b] Determined by ICP.

[c] Calculated by the following equation: [Si in the Si source]/[total Si in the synthesis gel].

[d] Calculated by the following equation: [Al in the Al source]/[total Al in the synthesis gel].

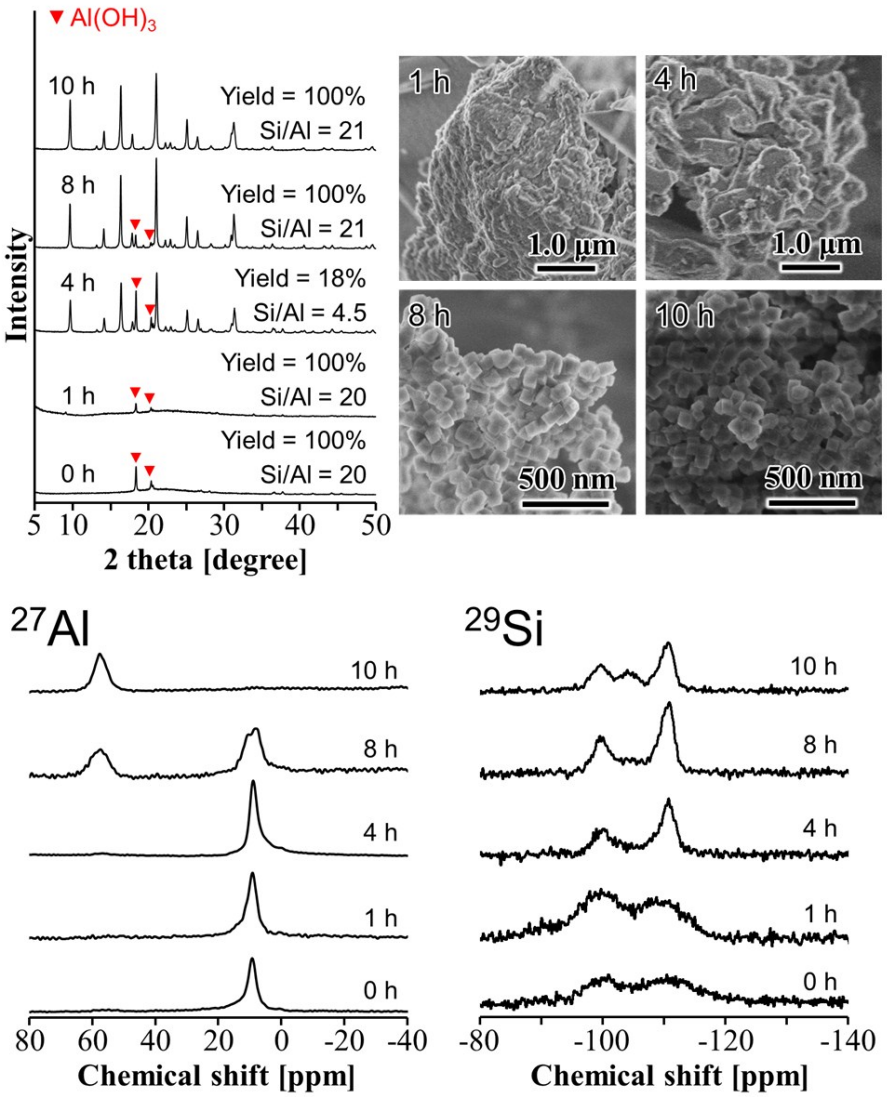

Figure S3. (left top) XRD patterns, (right top) SEM images, and (bottom) ^{27}Al and ^{29}Si MAS NMR spectra of solid products obtained from Si+Al with Si/Al ratio of 20 at different crystallization times (sample nos. 21–24).

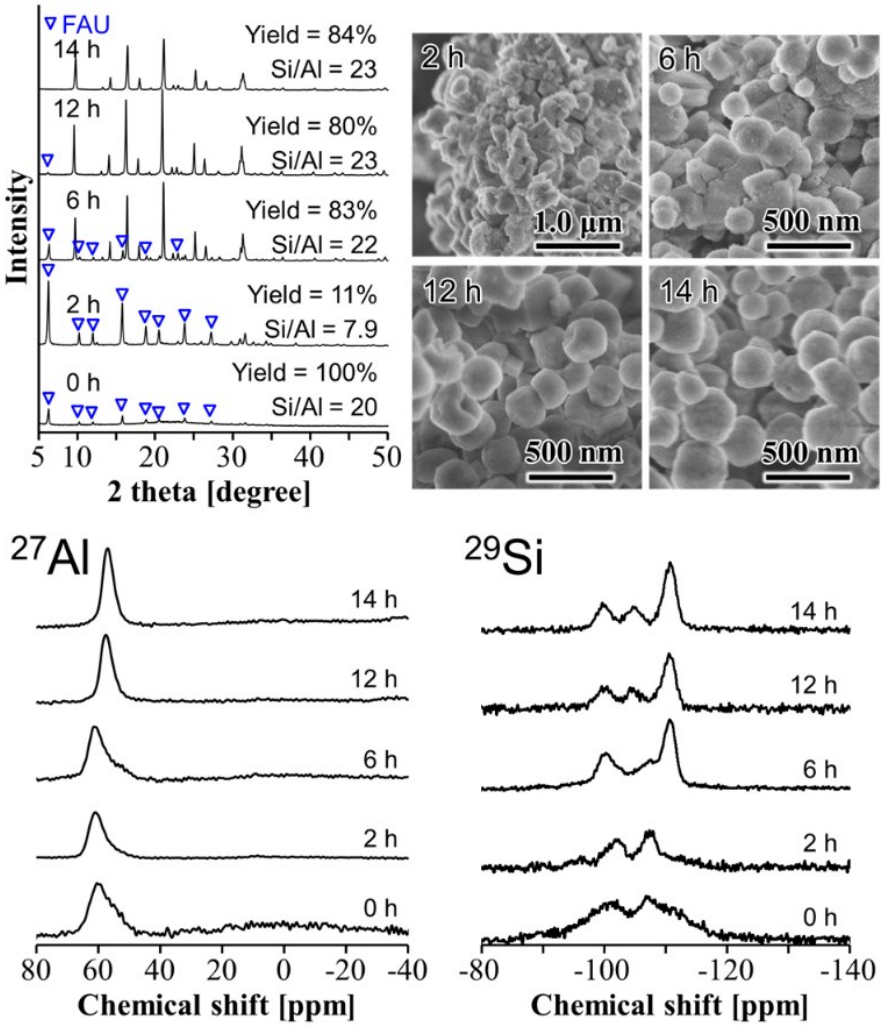

Figure S4. (left top) XRD patterns, (right top) SEM images, and (bottom) ^{27}Al and ^{29}Si MAS NMR spectra of solid products obtained from LSY+Si with Si/Al ratio of 20 at different crystallization times (sample nos. 25–28).

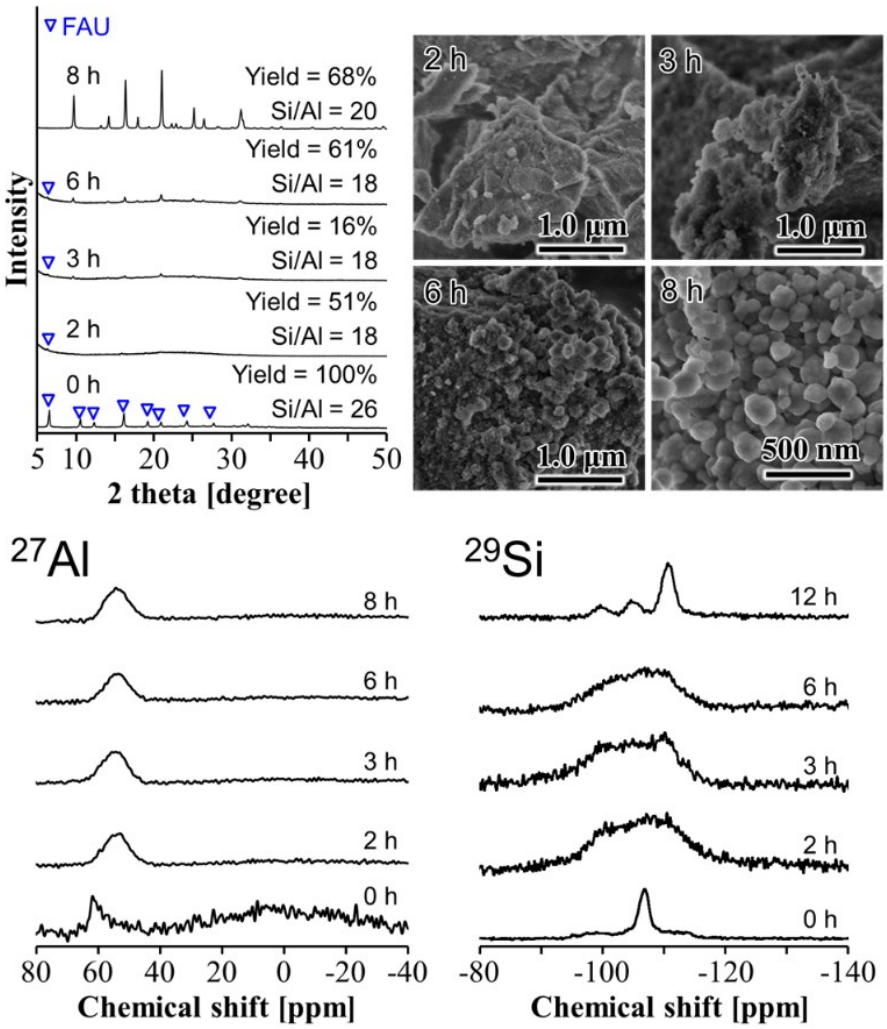

Figure S5. (left top) XRD patterns, (right top) SEM images, and (bottom) ^{27}Al and ^{29}Si MAS NMR spectra of solid products obtained from HSY with Si/Al ratio of 26 at different crystallization time (sample nos. 29–32).

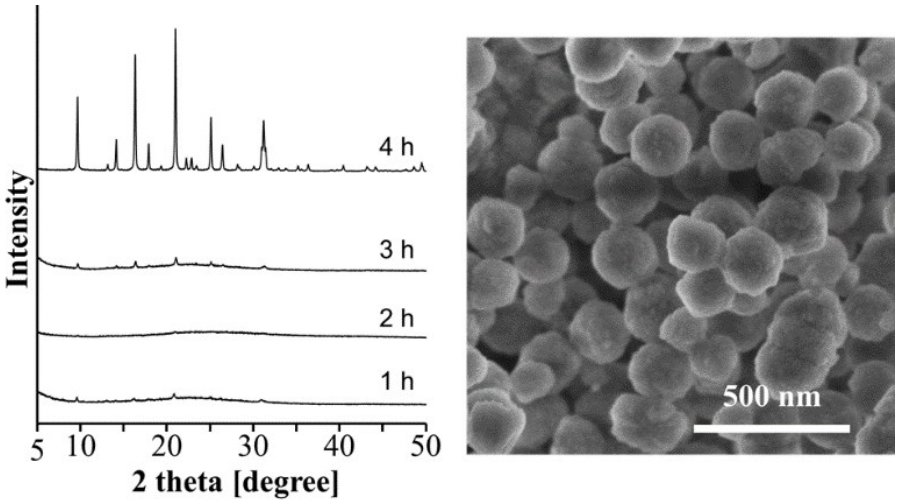

Figure S6. (left) Synthesis-time dependent XRD patterns of products obtained from HSY+Al when we used $NaAlO_2$ as an additional Al source instead of $Al(OH)_3$. (right) SEM images of product obtained after 4 h of hydrothermal treatment.

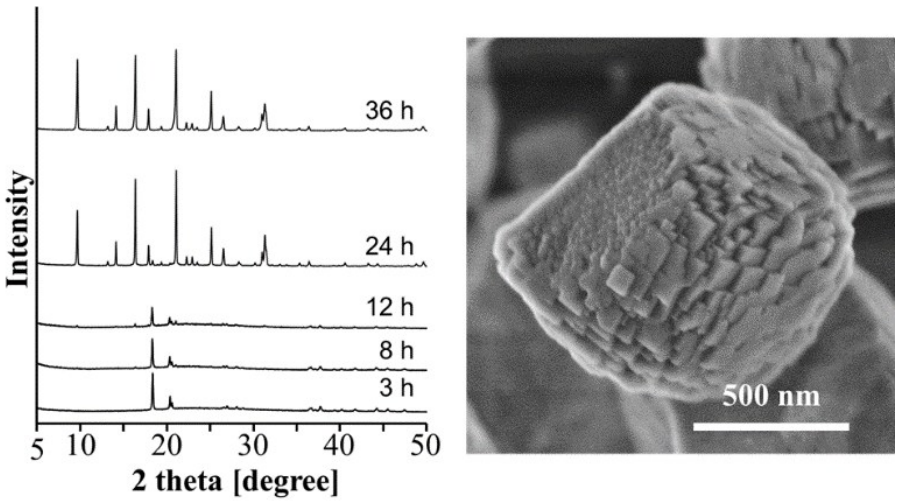

Figure S7. (left) Synthesis-time dependent XRD patterns of products synthesized using HSY+Al in the absence of CHA seed crystal. (right) SEM images of CHA zeolites obtained after 36 h of hydrothermal treatment.

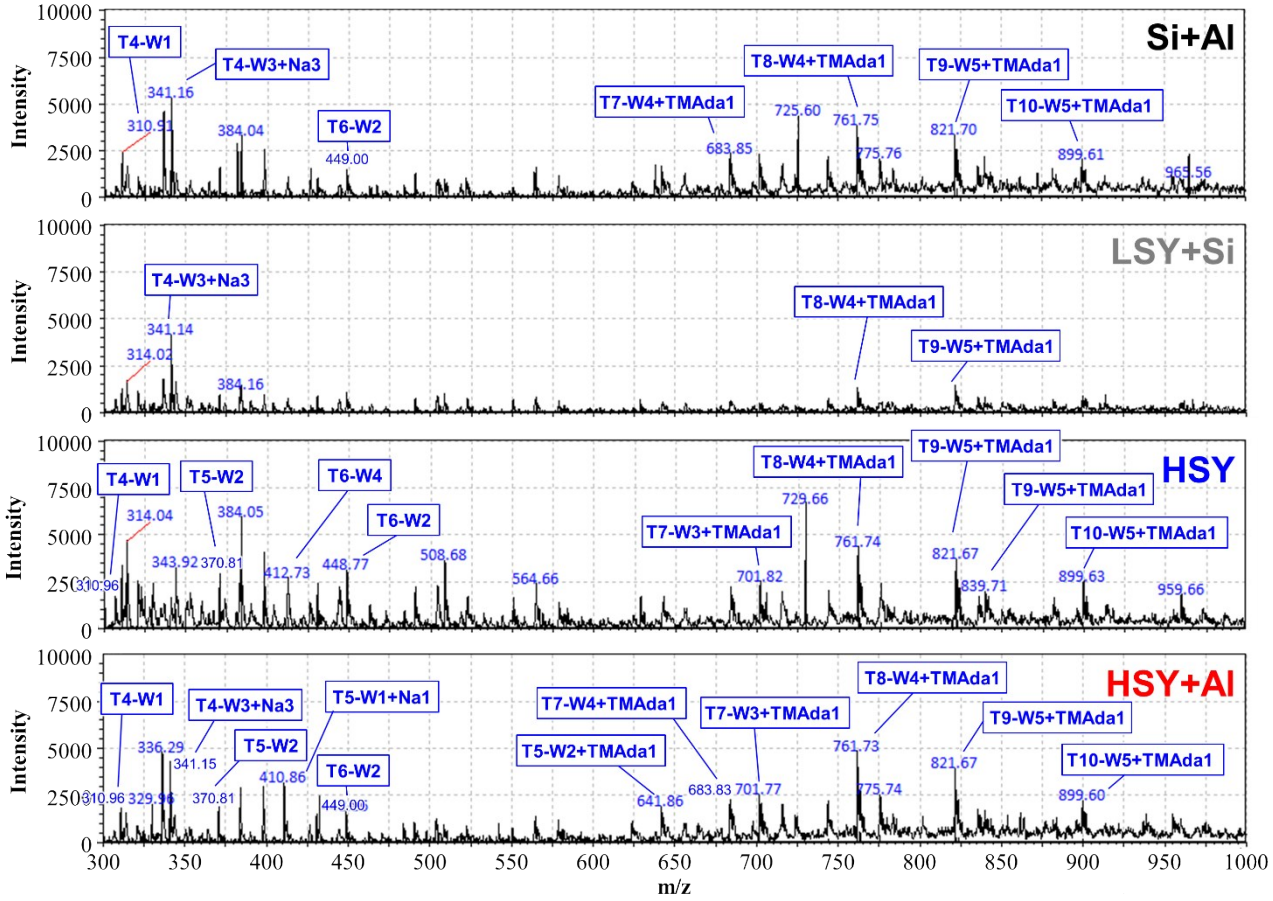

Figure S8. ESI-MS spectra of liquid phases obtained from various starting materials (sample nos. 23, 27, 33, and 37). The samples showing minimum solid yields were selected; synthesis times were 4, 2, 3, and 2 h for Si+Al, LSY+Al, HSY, and HSY+Al, respectively.

Table S2 Time-dependent product distribution for various CHA zeolite catalysts with different Si/Al and P/Al ratios (H-form).[a]

Catalyst			Time-on-stream	Product distribution [C-%]				
No.	Si/Al	P/Al	[min]	C_2H_4	C_3H_6	C_4H_8	C_2-C_4paraffin	others
CHA	22	0	15	37.8	19.1	9.3	6.4	27.4
			30	52.6	13.2	3	1.9	29.3
			45	58.7	10.4	2.2	1.3	27.4
			60	63.2	8.6	1.8	1	25.4
			75	64.1	6.4	0	0.7	28.8
			90	65.2	5.7	0	0.6	28.5
			105	66.1	4.8	0	0.6	28.5
			120	66.4	4.4	0.8	0.4	28
			135	67	4.2	0	0.5	28.3
			150	67.2	3.8	0	0.5	28.5
			165	68	4.1	0	0.5	27.4
			180	67.2	3.9	0	0.5	28.4
P-CHA	22	0.70	15	32.3	22.0	11.2	10.6	23.9
			30	45.1	19.2	5.0	3.0	27.7
			45	53.4	17.2	4.7	2.3	22.4
			60	57.0	13.6	3.1	1.6	24.7
			75	62.4	11.8	0.7	1.4	23.7
			90	65.3	9.4	0.4	1.7	23.2
			105	65.7	8.2	2.2	0.6	23.3
			120	66.7	7.3	0.0	0.7	25.3
			135	66.5	6.9	0.0	0.6	26.0
			150	67.9	5.9	0.0	0.7	25.5
			165	69.4	5.2	0.0	0.6	24.8
			180	68.5	5.5	0.0	0.7	25.3
P-CHA-2	11	0.38	15	15.4	27.4	12.2	13.7	31.3
			30	36.6	26.5	3.8	3.7	29.4
			45	51.3	18.1	1.90	2.1	26.6
			60	57.1	12.6	1.00	1	28.3
			75	62.2	9.6	0.30	1.4	26.5
			90	62.9	7	0.00	0.4	29.7
			105	65.3	6.3	0.30	0.6	27.5
			120	65.1	5.3	0.30	0.5	28.8
			135	66.8	4.4	0.00	0	28.8
			150	66.1	4.1	0.00	0.3	29.5
			165	66.7	4.3	0.00	0.5	28.5
			180	68	3.9	0.00	0.4	27.9

[a] Reaction conditions; Temp. = 400 °C, flow rate = 20 mL min^{-1} (C_2H_5OH/N_2 = 50/50 mol%), W/F = 0.025 $g_{cat}\,mL^{-1}min^{-1}$.

[b] Determined by ICP.

Figure S9. XRD patterns of CHA zeolite catalysts before and after ETO reaction.

Table S3 Comparison of initial propylene yields over various zeolites with different pore structures.

Catalyst name	Framework type	Si/Al	Time-on-stream [min]	Initial propylene yield[C-%]	Reference
P-CHA-2	CHA	11	15	27.4	This study
CHA	CHA	12.1	30	32.1	*Appl. Catal. A Gen.*, 2019, **575**, 204-213.
AEI	AEI	13.0	30	16.5	*Appl. Catal. A Gen.*, 2019, **575**, 204-213.
LEV	LEV	11.9	30	20.9	*Appl. Catal. A Gen.*, 2019, **575**, 204-213.
H-ZSM-5(Al)	MFI	37	30	23.5	*Appl. Catal., A*, 2011, **399**, 262-267.
HTON(sample no. 1)	TON	28.5	30	25.8	*J. Jpn. Pet. Inst.*, 2013, **56**, 22-31.