Electronic Supplementary Information

Tracking the Crystallization Behavior of High-Silica FAU During AEI-type Zeolite Synthesis Using Acid Treated FAU-type Zeolite

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Fig. S1. SEM image of the parent FAU (HSZ-320HOA).



Fig. S2. SEM images of sliced commercial FAU. (a) HSZ-320HOA (parent FAU, Si/AI = 2.8), (b) HSZ-350HUA (Si/AI = 5.0), and (c) HSZ-360HUA (Si/AI = 7.0).

x Na ₂ O/SiO ₂	0 min	15 min	30 min	1 h	2 h	4 h
0.05	-	Amor. + FAU	Amor. + FAU	Amor. + FAU	Amor. + FAU	_
0.10	_	FAU	FAU	FAU	FAU	FAU
0.15	Amor. + FAU	FAU	FAU	FAU + AEI	AEI + FAU	AEI

Table S1. Phases of products obtained at different heating times from starting reactants with various Na_2O/SiO_2 ratios.



Fig. S3. XRD patterns of products obtained from acid treated FAU at different crystallization times from the starting reactant with a chemical composition of 1.0 SiO_2 : $0.013 \text{ Al}_2\text{O}_3$: $0.05 \text{ Na}_2\text{O}$: 0.20 DMDMPOH: $5.0 \text{ H}_2\text{O}$.



Fig. S4. XRD patterns of products obtained from acid treated FAU at different crystallization times from the starting reactant with a chemical composition of 1.0 SiO₂: 0.013 Al₂O₃: 0.10 Na₂O: 0.20 DMDMPOH: 5.0 H₂O.



Fig. S5. XRD patterns of products obtained from acid treated FAU at different crystallization times from the starting reactant with a chemical composition of 1.0 SiO₂: 0.013 Al₂O₃: 0.15 Na₂O: 0.20 DMDMPOH: 5.0 H₂O.



Fig. S6. SEM images of products obtained at different crystallization times from the starting reactants with Na_2O/SiO_2 ratios of (a) 0.10 and (b) 0.15.



Fig. S7. S(Q)s of various materials, (a) parent FAU and AcT-FAU, and (b) products obtained from AcT-FAU at different crystallization times.

Table S2	. Si/Al ratio	s, yields,	and OSDA	contents	of products	obtained a	at different	crystallization	times from	the s	starting
reactant v	with a chen	nical com	position of	1.0 SiO ₂ :	0.013 Al ₂ O ₃	: 0.15 Na ₂ 0	D: 0.20 DN	IDMPOH: 5.0	H ₂ O.		

	15 min	30 min	1 h	2 h	4 h
Si/Al [–]	9.95	10.2	9.15	8.21	9.68
Solid yield [wt%]	3.39	2.19	3.40	4.00	5.60
Occluded OSDA [wt%]	16.5	15.3	16.0	15.1	15.5
Na/Al [–]	0.515	0.666	0.533	0.485	0.460

Table S3. Si/Al ratios, yields, and OSDA contents of products obtained at different crystallization times from the starting reactant with chemical composition a of 1.0 SiO₂: 0.013 Al₂O₃: 0.10 Na₂O: 0.20 DMDMPOH: 5.0 H₂O.

	0 min	15 min	30 min	1 h	2 h	4 h
Si/Al [–]	17.7	8.96	8.32	7.89	11.4	13.0
Solid yield [wt%]	9.40	3.21	5.00	5.01	18.6	24.0
Occluded OSDA [wt%]	12.7	16.3	15.3	16.1	16.7	16.4
Na/AI [–]	0.952	0.462	0.676	0.504	0.279	0.0146



Fig. S8. Thermogravimetry-differential thermal curves of products obtained at different synthesis times. (a) DTG, and (b) DDTA. DTG and DDTA mean the time derivative of TGA and DTA, respectively. The chemical composition of the starting reactant was 1.0 SiO₂: 0.013 Al₂O₃: 0.15 Na₂O: 0.20 DMDMPOH: 5.0 H₂O.



Void formation

Fig. S9. The proposed model for the formed voids in high-silica FAU.



 2θ [°] Fig. S10. XRD patterns of products obtained from the starting reactants with a chemical composition of 1.0 SiO₂: 0.013 Al₂O₃: *x* Na₂O: 0.0 DMDMPOH: 5.0 H₂O (*x* = 0.15, 0.25).