Supporting Information for

Influence of alkali metal cations on the local structure of amorphous precursors during zeolite crystallization

Kazuki Mori^a, Peidong Hu^{a,b}, Kentaro Kobayashi^c, Hiroki Yamada^d, Koji Ohara^c, Yutaka Yanaba^e, Tatsuya Okubo^a, Toru Wakihara^{a,b*}

^a Department of Chemical System Engineering, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan
^b Institute of Engineering Innovation, School of Engineering, The University of Tokyo, 2-11-16 Yayoi, Bunkyo-ku, Tokyo 113-8656, Japan
^c Faculty of Materials for Energy, Shimane University, 1060, Nishikawatsu, Matsue, Shimane 690-8504, Japan
^d Japan Synchrotron Radiation Research Institute / SPring-8, 1-1-1 Kouto, Sayo-cho, Sayo-gun, Hyogo 679-5198, Japan
^e Institute of Industrial Science, The University of Tokyo, 4-6-1 Komaba, Meguro-ku, Tokyo 153-8505, Japan *

*Email: wakihara@chemsys.t.u-tokyo.ac.jp (T. Wakihara)

Supporting fig



Fig. S1 Raman spectra of samples synthesized using (a): Na⁺, (b): K⁺. Dash and solid lines represent amorphous and crystalline species, respectively.



Fig. S2 Structure factor S(Q) of solid samples obtained with (a) Na⁺ and (b) K⁺.



Fig. S3 Partial reduced PDFs. $G_{ij}(r)$, of (a) FAU- and (b) MER-type zeolite calculated by $PDFgui^{1}$.



Fig. S4 Comparison between the experimental S(Q) (\bigcirc) and the results of RMC modeling (–).



Fig. S5 Structures of (a) K-14h and (b) K-14h obtained by RMC modelling. (c)Comparison of partial PDF, $g_{ij}(r)$, of Na-2h and K-14h.



Fig. S6 Distributions of T–O–T and O–T–O angle.



Fig. S7 Distributions of Al atoms in the 4Rs of Na-2h and K-14h.



Fig. S8 Distributions of T–O–T angles in FAU- and MER- type zeolites.

Synthesis period [h]	Si/Al [-]	Na/Al [-]	Yield [%]
0	1.6	1.20	35
1	1.7	0.93	32
2	1.7	0.90	35
3	1.7	0.84	35
5	1.7	0.94	38
6	1.7	0.92	37
7	1.7	0.87	37
13	1.7	0.88	40
14	1.7	0.90	41

Table S1 Molar compositions and solid yields of samples synthesized at 100 $^{\circ}$ C using Na⁺ and K⁺.

(a)	Sampl	es svni	thesized	using	Na ⁺
(a)	Sampi	es sym	inesizeu	using	INA

(b) Samples synthesized using $K^{\scriptscriptstyle +}$

Synthesis period [h]	Si/Al [-]	K/Al [-]	Yield [%]
0	1.8	1.10	27
12	1.8	0.89	32
13	1.8	0.89	32
14	1.8	0.88	34
15	1.8	0.91	37
16	1.8	0.90	41
17	1.8	0.90	42
24	1.8	0.92	45
48	1.8	0.90	44
72	1.9	0.93	45

Zeolite	Framework	Ring	Composite Building Unit (CBU)
FAU		4R 6R 12R	d8r pau
MER		4R 8R • : T (Si or Al) d6r sod

Table S2 Ring structures and composite building units of MER- and FAU- type zeolites.

Reference

C. L. Farrow, P. Juhas, J. W. Liu, D. Bryndin, E. S. Boin, J. Bloch, T. Proffen and S. J. L.
 Billinge, PDFfit2 and PDFgui: Computer programs for studying nanostructure in crystals, *J. Phys. Condens. Matter*