## **Electronic Supplementary Information**

## **One-Step Synthesis of Hierarchical Pentasil Zeolite Microspheres Using Diamine with Linear Carbon Chain as Single Template**

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Fig. S1. Powder XRD patterns of the as-synthesized ZSM-5 microspheres with different  $SiO_2/Al_2O_3$  synthesized by (a) EDA, (b) HDA, (c) DAOT, (d) DADC template at 428 K for (a) 5 days, (b) 5 days, (c) 10 days, (d) 10 days ( b:Y-axis shift: 300; c:Y-axis shift: 600; d:Y-axis shift: 900.)



Fig. S2. XRD patterns and IR spectra of the HDA-ZSM-5-60 and HDA-ZSM-5-30 with the crystallization time 1 d (a), 3 d (b) and 5 d (c).



Fig. S3. Powder XRD patterns of the as-synthesized HDA-ZSM-5-30 microsphere with  $H_2O/SiO_2=50$ .



Fig. S4. TG-DTG analyses of the EDA-ZSM-5-30 (A), HDA- ZSM-5-30 (B), DAOT-ZSM-11-30 (C) and DADC-ZSM-11-30 (D) samples.



Fig. S5. <sup>27</sup>Al NMR (A) and <sup>29</sup>Si NMR (B) spectra of the as-synthesized zeolites, EDA-ZSM-5-30 (a), HDA-ZSM-5-30 (b), DAOT-ZSM-11-30 (c), DADC-ZSM-11-30 (d), and HDA-ZSM-5-60 (e).



Fig. S6. SEM images of the ZSM-5-ind sample.

Table S1. XRD Crystallinity and IR Crystallinity of different crystallization time samples

samples	Crystallization time	XRD Crystallinity	IR Crystallinity
SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =60	1d	45.4%	46.4%
	3d	95.7%	84.4%
	5d	100.0%	83.4%
SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> =30	1d	0%	0%
	3d	93.6%	76.0%
	5d	100%	87.4%

## *Table S2*. The information of zeolite pentasil synthesized using diamines as template in the literatures

Molar compositions of synthetic mixtures	Template	Morphology	Reference
(R) <sub>26</sub> (K <sub>2</sub> O) <sub>12</sub> (Al <sub>2</sub> O <sub>3</sub> )(SiO <sub>2</sub> ) <sub>2</sub> (H <sub>2</sub> O) <sub>3580</sub>	1,8-Diaminooctane	intergrowth crystals ZSM-11 (10 x 15 μm)	[21]
106 SiO <sub>2</sub> : 26 Na <sub>2</sub> O: Al <sub>2</sub> O <sub>3</sub> : 17.3 H <sub>2</sub> SO <sub>4</sub> : 47 NaCl: 2400 H <sub>2</sub> O: 9 R	Diaminoalkane: H <sub>2</sub> N-(CH <sub>2</sub> ) <sub>3</sub> -NH <sub>2</sub> or H <sub>2</sub> N-(CH <sub>2</sub> ) <sub>9</sub> -NH <sub>2</sub>	Dispersed 5µm coffin-like ZSM-5/11 particles or 1µm spherical particles	[22]
$0.7-0.9[xR_2O+(1-x)Na_2O]:Al_2O_3:5-100SiO_2:$ $0-40H_2O$	1,6-diaminohexane or 1,2-cyclohexanedi amine	1.3 μm or 0.8 μm polycrystalline spherical particles	[23]
1.85 Na <sub>2</sub> O: Al <sub>2</sub> O <sub>3</sub> : 15.2 SiO <sub>2</sub> : 592 H <sub>2</sub> O : 19. 7 R	Ethylene Diamine	lath shaped	[24]
SiO <sub>2</sub> : 0.01Al <sub>2</sub> O <sub>3</sub> : 0.05CaO :0.1Na <sub>2</sub> O:R	1,6-diaminohexane	ca. 10 μm cauliflower-like morphology of Ca-MFI zeolites (in static condition)	[25]

samples	Size of prin	nary particles	Size of aggregates	$V_{Mesopore}$ <sup>c</sup>
	(nm) <sup>a</sup>	(nm) <sup>b</sup>	(µm) <sup>b</sup>	$(cm^3/g)$
EDA-ZSM-5-60	337.13	100-300	20-25	0.09
HDA-ZSM-5-60	238.85	100-150	4-6	0.12
DAOT-ZSM-11-60	240.95	50-80	6-8	0.06
DADC-ZSM-11-60	284.55	40-70	6-8	0.12
EDA-ZSM-5-30	230.54	100-200	15-20	0.09
HDA-ZSM-5-30	170.32	50-100	4-6	0.20
DAOT-ZSM-11-30	157.34	30-60	6-8	0.18
DADC-ZSM-11-30	236.69	30-50	7-8	0.09

Table S3. The size of the mesoporous zeolite samples

a calculated from the XRD. b obtained from SEM. c Mesopore Volume obtained from BET.