

Extra-large pore JZO zeolites with tunable Si/Al ratios as efficient catalysts for degradable plastic monomer production

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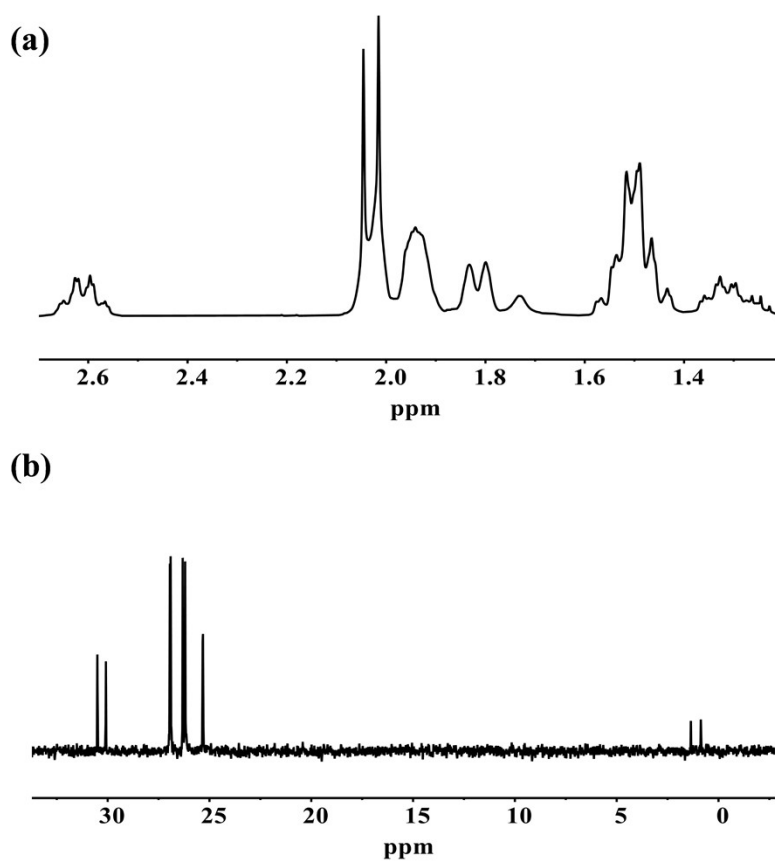


Figure S1. Liquid phase ^1H NMR (a) and ^{13}C NMR (b) spectra of TCyMPI in CDCl_3 .

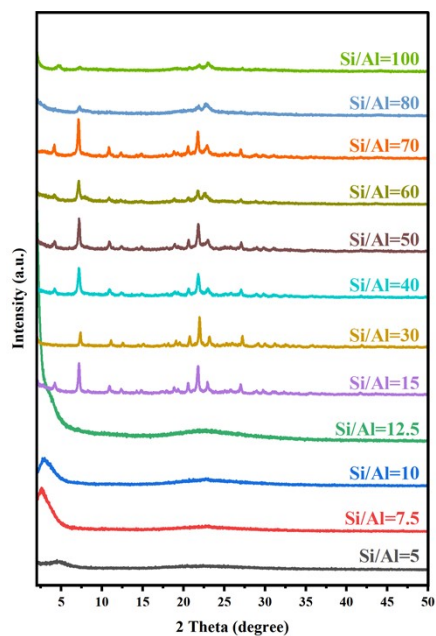


Figure S2. PXRD patterns of as-made JZO zeolites obtained with different Si/Al ratios after heating for 5 days. The gel molar composition is 1.0 SiO₂: 0.5 TCyMPOH: x Al₂O₃: 0.2 HF: 5 H₂O, 5 wt.% seed, with $x = 0.100, 0.067, 0.05, 0.040, 0.033, 0.017, 0.013, 0.010, 0.008, 0.007, 0.006, 0.005$ corresponding to Si/Al ratio of 5, 7.5, 10, 12.5, 15, 30, 40, 50, 60, 70, 80, 100, respectively.

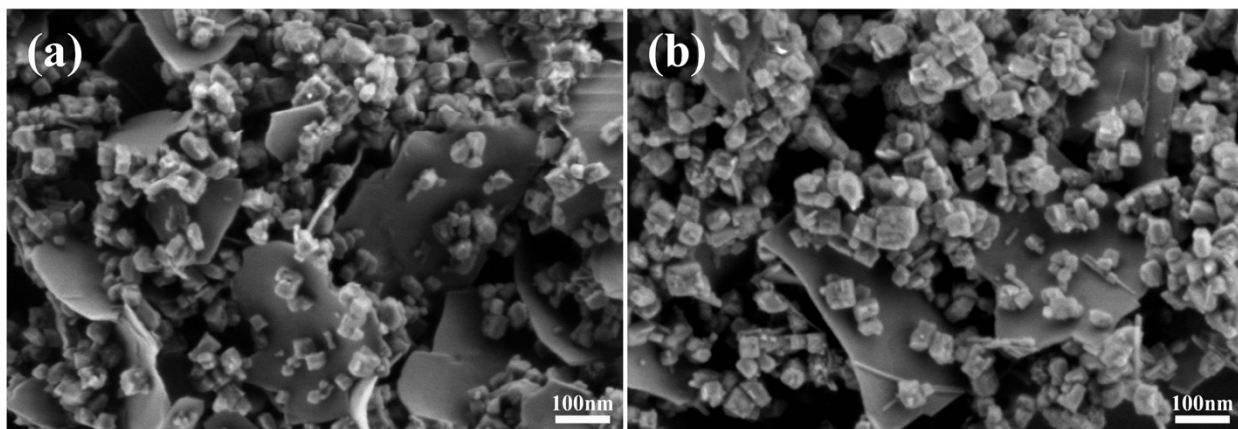


Figure S3. SEM images of as-made JZO zeolites obtained with a Si/Al ratio of 70 after heating for 11 days. 70-s-pre (a1), 70-n-pre (a2). The gel molar composition is 1.0 SiO₂: 0.5 TCyMPOH: 0.007 Al₂O₃: 0.2 HF: 5 H₂O, s-5 wt.% seed, n-no seed.

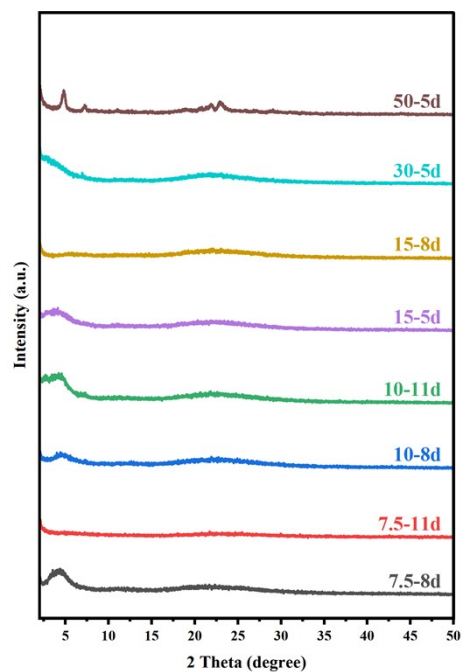


Figure S4. PXRD patterns of as-made JZO zeolites obtained with different Si/Al ratios over different crystallization times. The gel molar composition is 1.0 SiO₂: x Al₂O₃: 0.5 TCyMPOH: 0.2 HF: 10 H₂O, 5 wt.% seed, with $x = 0.067, 0.05, 0.033, 0.017, 0.010$ corresponding to Si/Al ratio of 7.5, 10, 15, 30, 50, respectively.

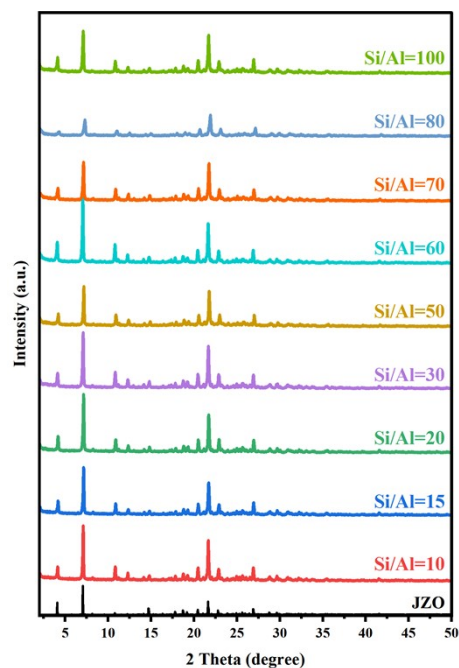


Figure S5. PXRD patterns of as-made JZO zeolites obtained with different Si/Al ratios after heating for 8 days. The gel molar composition is $1.0 \text{ SiO}_2 : x \text{ Al}_2\text{O}_3 : 0.5 \text{ TCyMPOH} : 10 \text{ H}_2\text{O}$, 5 wt.% seed, with $x = 0.050, 0.033, 0.025, 0.017, 0.010, 0.008, 0.007, 0.006, 0.005$ corresponding to Si/Al ratio of 10, 15, 20, 30, 50, 60, 70, 80, 100, respectively.

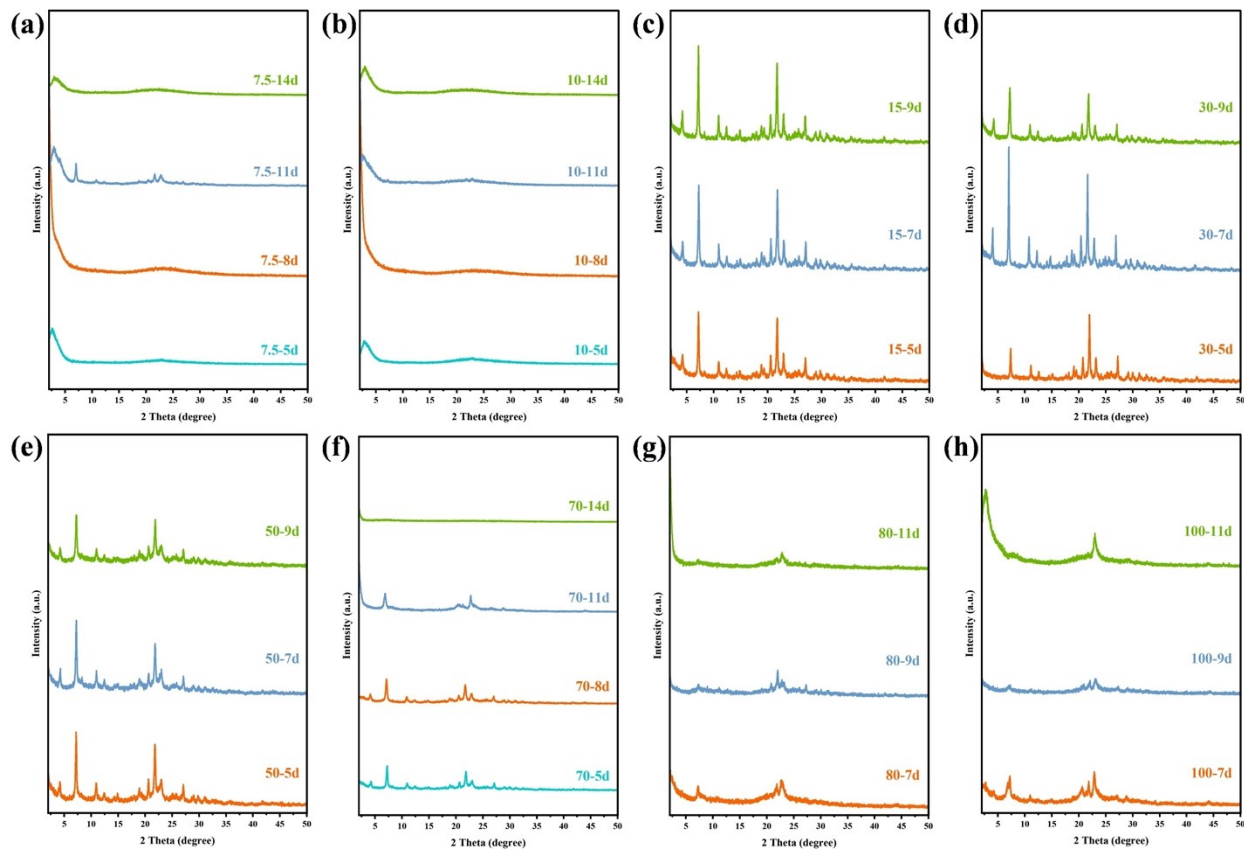


Figure S6. PXR D patterns of as-made products obtained with a gel Si/Al ratio of 7.5 (a), 10 (b), 15 (c), 30 (d), 50 (e), 70 (f), 80 (g) and 100 (h) over different crystallization times. The gel molar composition is $1.0 \text{ SiO}_2 : x \text{ Al}_2\text{O}_3 : 0.5 \text{ TCyMPOH} : 0.2 \text{ HF} : 5 \text{ H}_2\text{O}$, 5 wt.% seed, with $x = 0.067, 0.05, 0.033, 0.017, 0.010, 0.007, 0.006, 0.005$ corresponding to the Si/Al ratio of 7.5, 10, 15, 30, 50, 70, 80, 100, respectively.

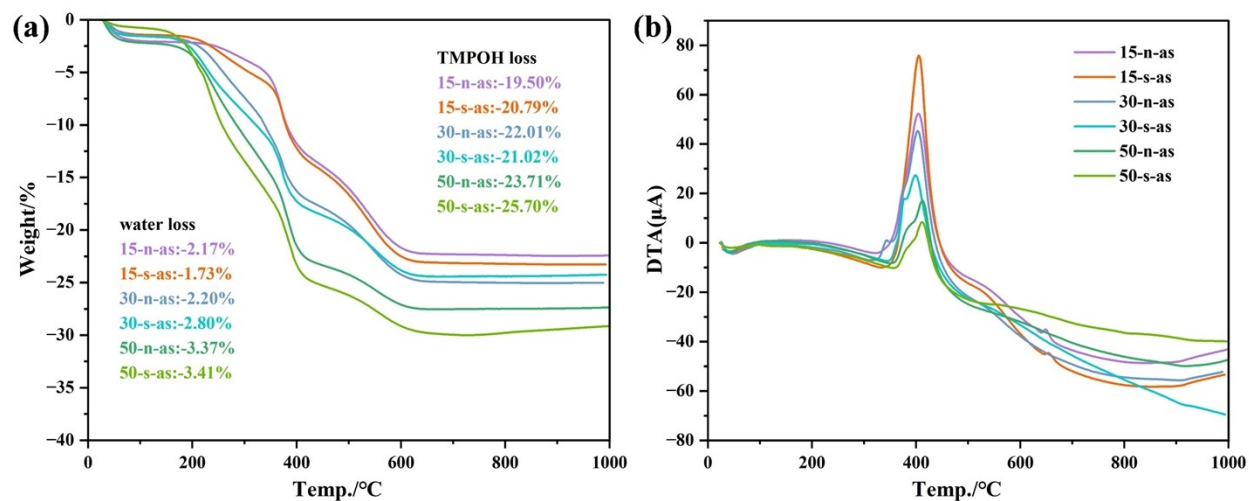


Figure S7. TG (a) and DTA (b) profiles of as-made JZO zeolites obtained with different Si/Al ratios. The gel molar composition is 1.0 SiO₂: 0.5 TCyMPOH: x Al₂O₃: 0.2 HF: 5 H₂O, with x = 0.010, 0.017, 0.033 corresponding to Si/Al ratio of 50, 30, 15, respectively. s-5 wt.% seed, n-no seed.

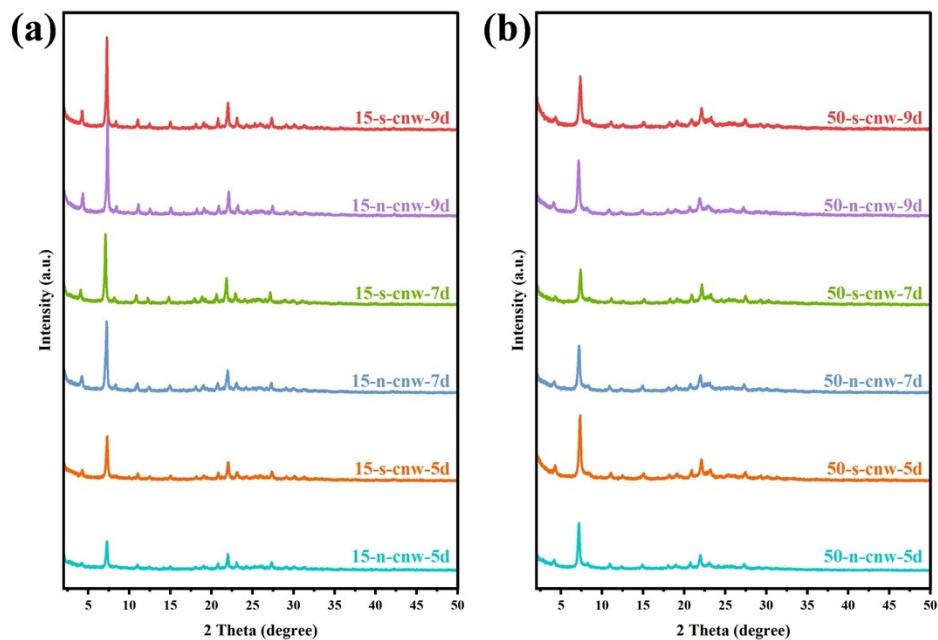


Figure S8. The XRD patterns of the cnw-JZO zeolite with Si/Al ratios of 15 (a) and 50 (b) after heating for 5, 7 and 9 days. The gel molar composition is 1.0 SiO₂: 0.5 TCyMPOH: x Al₂O₃: 0.2 HF: 5 H₂O, with $x = 0.010, 0.033$ corresponding to Si/Al ratios of 50, 15, respectively. s-5 wt.% seed, n-no seed.

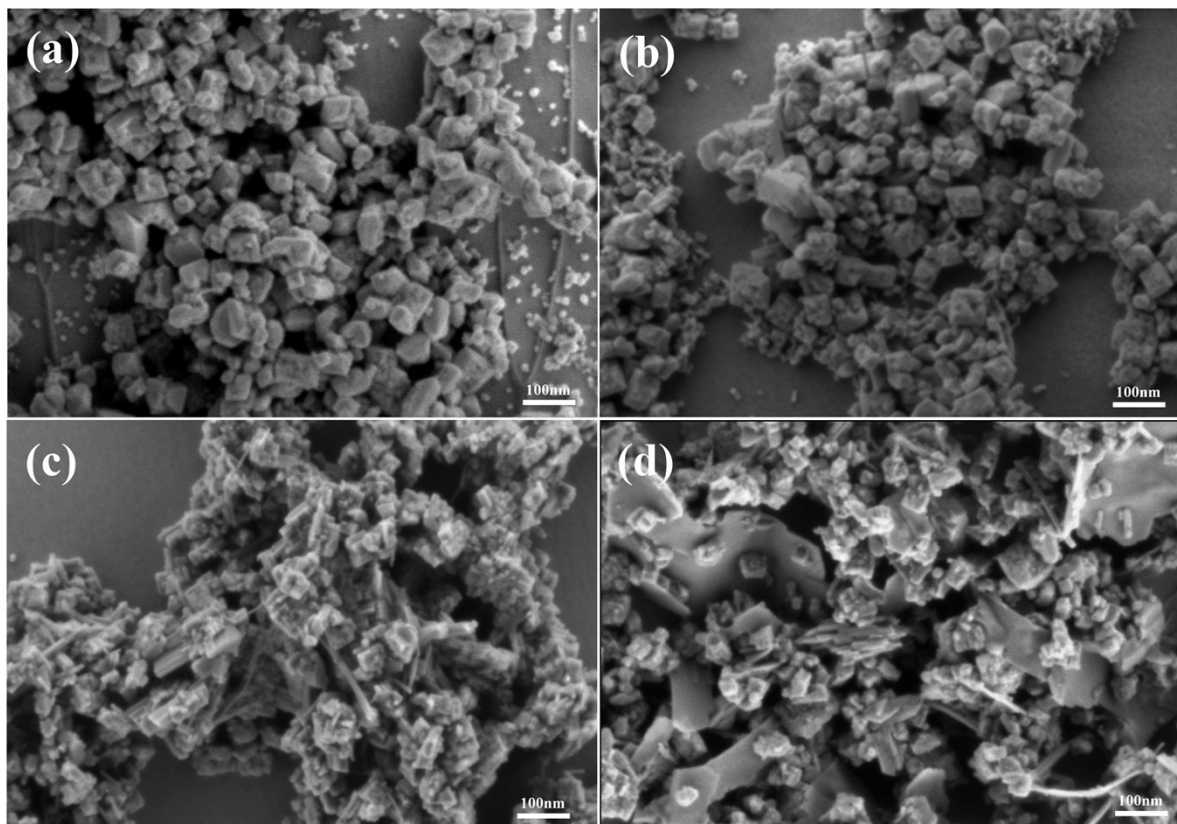


Figure S9. The SEM images of 15-s-cnw-5d (a), 15-n-cnw-5d (b), 50-s-cnw-5d (c) and 50-n-cnw-5d (d) after heating for 5 days. The gel molar composition is $1.0 \text{ SiO}_2 : 0.5 \text{ TCyMPOH} : x \text{ Al}_2\text{O}_3 : 0.2 \text{ HF} : 5 \text{ H}_2\text{O}$, with $x = 0.010, 0.033$ corresponding to Si/Al ratios of 50, 15, respectively. s-5 wt.% seed, n-no seed.

Supplementary Tables:

Table S1. Elemental analysis results of P-free JZO zeolites obtained with Si/Al = 70 using HF.

Zeolite	XRF		
	SiO ₂ %	Al ₂ O ₃ %	Si/Al
70-s-cnw	97.22	1.96	48.84
70-n-cnw	97.26	1.65	49.99

Table S2. Summary of synthesis and elemental analysis over samples obtained in hydroxide media.

Entry	Si/Al	H ₂ O/SiO ₂	Time (day)	seed	Phase	Yield (wt.%)	XRF		
							SiO ₂ %	Al ₂ O ₃ %	Si/Al
1	10	10	8	5 wt.%	JZO	3.4	-	-	-
2	15	-	-	-	JZO	15	84.5	6.75	10.6
3	20	-	-	-	JZO	17.5	94.5	4.79	16.8
4	30	-	-	-	JZO	11.3	95.4	4.05	20.0
5	50	-	-	-	JZO	13	97.4	2.22	37.3
6	60	-	-	-	JZO	7	-	-	-
7	70	-	-	-	JZO	6	-	-	-
8	80	-	-	-	JZO	6	-	-	-
9	100	-	-	-	JZO	4	-	-	-

Table S3. Textural properties of P-free JZO zeolites.

Sample	S_{BET} (m²/g)^[a]	V_{micropore} (cm³/g)^[b]
15-n-cnw	886.9	0.31
15-s-cnw	901.9	0.32
30-n-cnw	873.8	0.30
30-s-cnw	910.2	0.32
50-n-cnw	827.8	0.27
50-s-cnw	836.8	0.27

^[a]Apparent surface area calculated using the Rouquerol BET criteria. ^[b]The micropore volume was obtained by N₂@77K adsorption using the t-plot method.

Table S4. Acid sites concentration and strength for cnw-JZO zeolites.

Sample	Peak area			Adsorbed ammonia ($\mu\text{mol/g}$ catalyst)		
	Peak1	Peak2	Peak area ratio	Weak acid	Medium-strong acid	Total acidity
15-s-cnw	4543	14576	10: 32	47.14	255.33	351.35
30-s-cnw	3301	11853	10: 36	34.25	170.87	220.38
50-s-cnw	2060	7574	10: 37	21.34	78.59	99.93
15-n-cnw	5200	14311	10: 26	53.75	148.48	202.23
30-n-cnw	2885	14345	10: 50	29.93	148.84	178.77
50-n-cnw	2259	6003	10: 27	23.44	62.29	85.73