

## Supporting Information

# One-Pot Synthesis of Ultra-Small Pt Dispersed on Hierarchical Zeolite Nanosheet Surfaces for Mild Hydrodeoxygenation of 4-Propylphenol

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### List of Supporting Materials

**Figure S1.** SEM images and particle size distribution of the synthesized Pt supported on silicalite-1 samples.

**Figure S2.** TEM images, EDS elemental mapping for Pt on STEM images, and Pt particle size distribution of the synthesized Pt supported on silicalite-1 samples.

**Figure S3.** XRD patterns N<sub>2</sub> adsorption-desorption isotherms of the synthesized bifunctional Pt supported on HZSM-5 samples.

**Figure S4.** SEM images and particle size distribution of the synthesized bifunctional Pt supported on HZSM-5 samples.

**Figure S5.** TEM images of the synthesized bifunctional Pt supported on HZSM-5 samples.

**Figure S6.** Pt particle size distribution of the synthesized bifunctional Pt supported on HZSM-5 samples.

**Figure S7.** XANE of various Pt supported zeolite samples

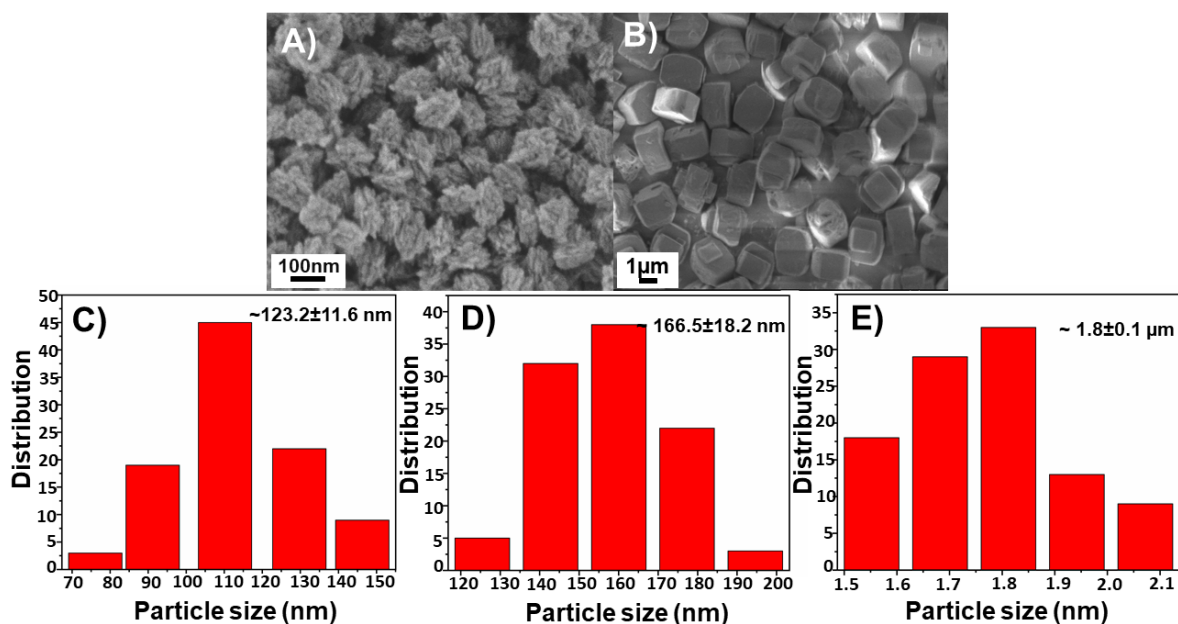
**Figure S8.** NH<sub>3</sub>-TPD profiles of the synthesized bifunctional Pt supported on HZSM-5 samples.

**Table S1.** Pt content and the relative crystallinity of the synthesized Pt supported on zeolites.

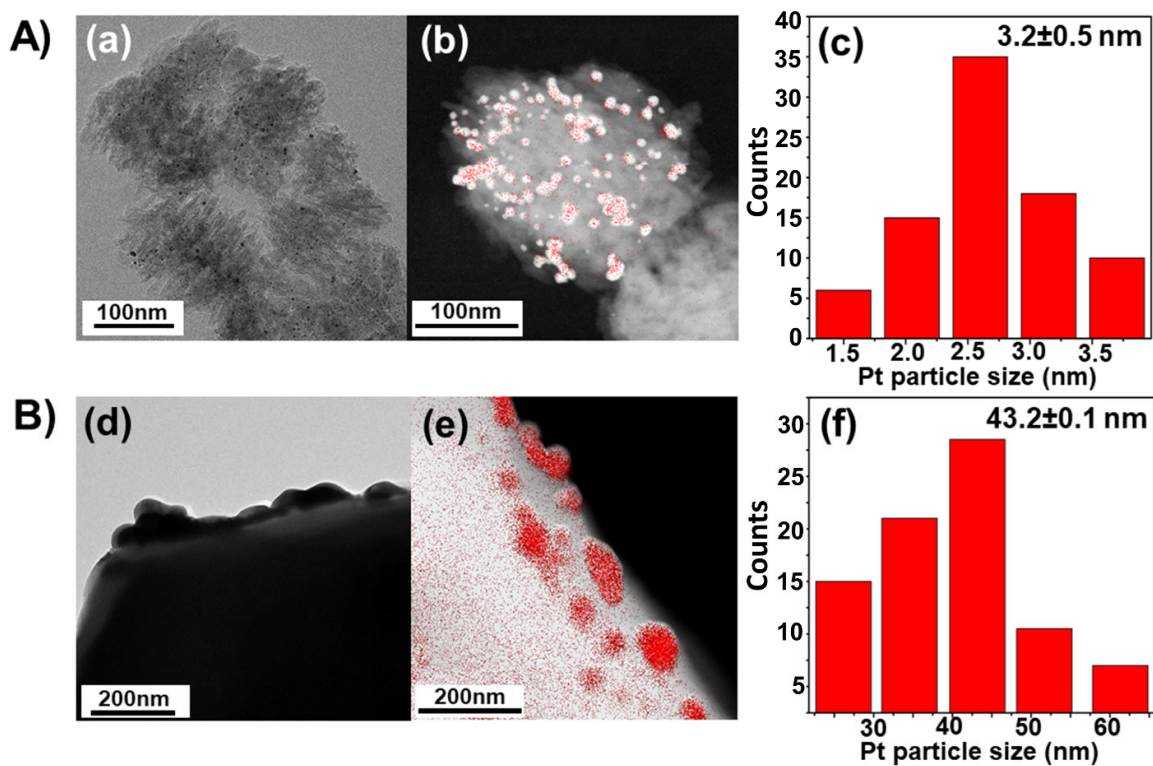
**Table S2.** Textural properties of the synthesized Pt supported on HZSM-5 samples.

**Table S3.** Summary data of the conversion and selectivity over Pt@HZSM-5NS (one) obtained at various reaction times.

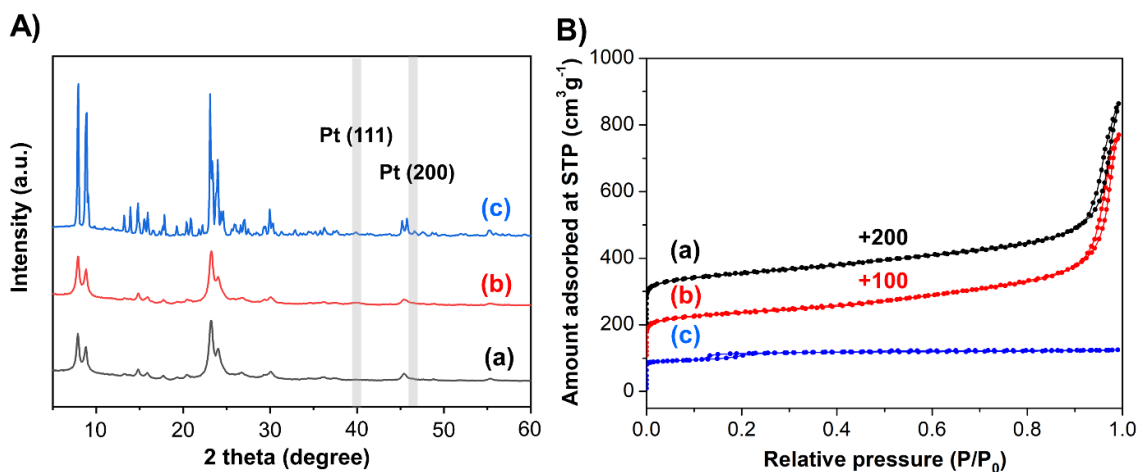
**Table S4.** Summary data of the conversion and selectivity over various catalysts.



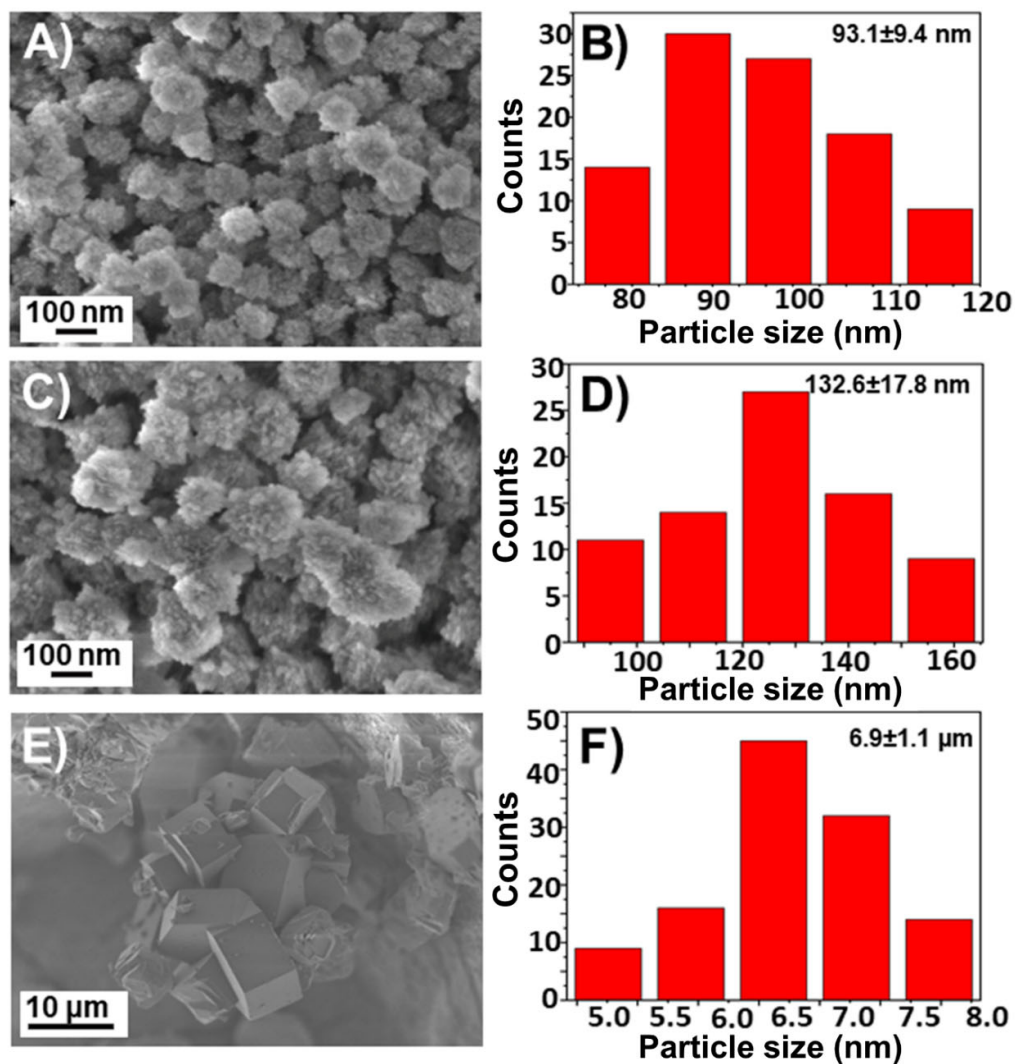
**Figure S1.** SEM images of (A) Pt/SiNS(imp) and (B) Pt/SiCON(imp), and particle size distribution of (C) Pt/SiNS(one), (D) Pt/SiNS(imp), and (E) Pt/SiCON(imp).



**Figure S2.** TEM images (a, d), EDS elemental mapping for Pt (red spots) on STEM images (b, e), and Pt particle size distribution (c, f) of (A) Pt/SiNS(imp) and (B) Pt/SiCON(imp).



**Figure S3.** (A) XRD patterns and (B) N<sub>2</sub> adsorption-desorption isotherms of (a) Pt@HZSM-5NS(one), (b) Pt@HZSM-5NS(imp), and (c) Pt@HZSM-5CON(imp).



**Figure S4.** SEM images (A, C, E) and particle size distribution (B, D, F) of Pt@HZSM-5-NS(one) (A, B), Pt@HZSM-5-NS(imp) (C, D), and Pt@HZSM-5-CON(imp) (E, F).

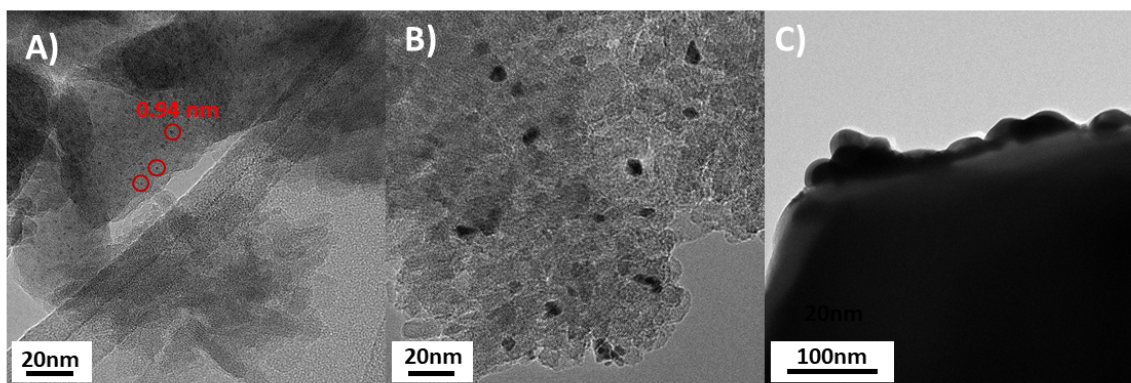


Figure S5. TEM images of (A) Pt@HZSM-5-NS(one), (B) Pt@HZSM-5-NS(imp), (C) Pt@HZSM-5-CON(imp).

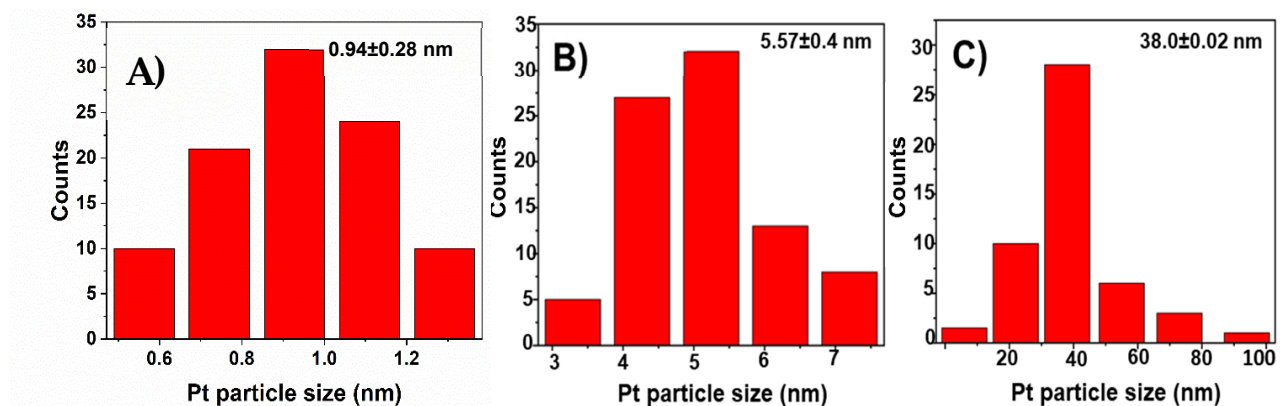


Figure S6. Pt particle size distribution of (A) Pt@HZSM-5-NS(one), (B) Pt@HZSM-5-NS(imp), and (C) Pt@HZSM-5-CON(imp).

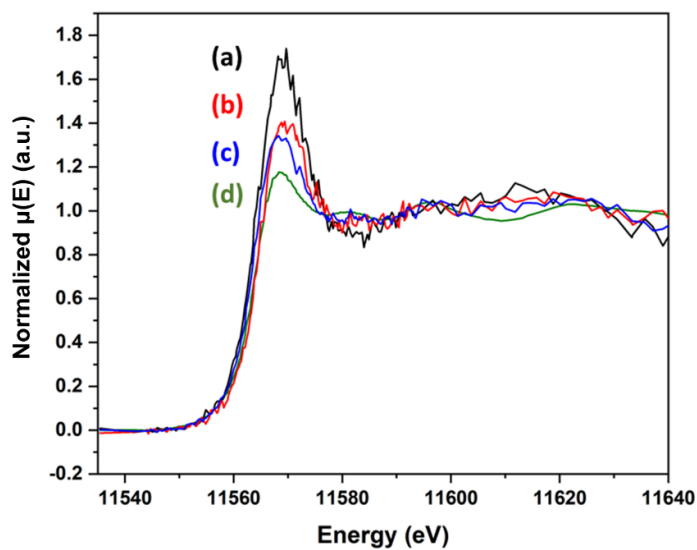
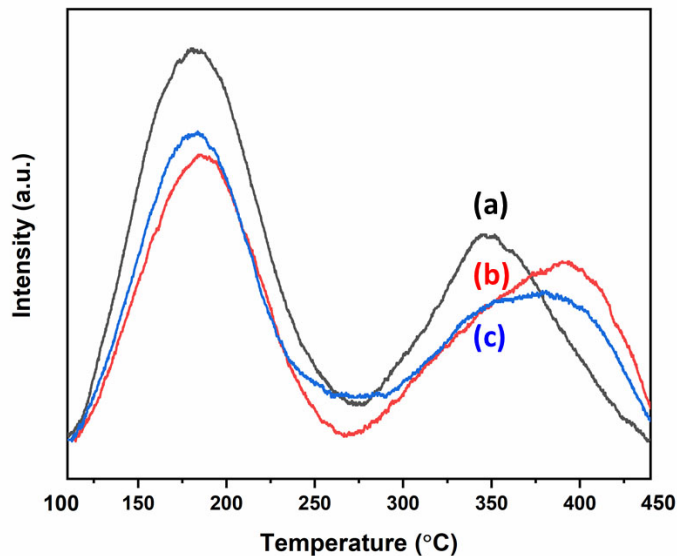


Figure S7. Pt L<sub>3</sub> edge XANES spectra of (a) Pt@HZSM-5-NS(one), (b) Pt@HZSM-5-NS(imp), (c) Pt@HZSM-5-CON(imp), and (d) standard Pt foil.



**Figure S8.**  $\text{NH}_3$ -TPD profiles of (a) Pt@HZSM-5-NS(one), (b) Pt/HZSM-5-NS(imp), (c) Pt/HZSM-5-CON(imp).

**Table S1.** Pt content (wt.%) in the synthesized samples.

Sample	Pt content <sup>a</sup> (wt.%)
Pt@SiNS(one)	0.67
Pt/SiNS(imp)	0.63
Pt/SiCON(imp)	0.65
Pt@HZSM-5-NS(one)	0.41
Pt/HZSM-5-NS(imp)	0.42
Pt/HZSM-5-CON(one)	0.42

<sup>a</sup>determined by XRF technique.

**Table S2.** Textural properties of the synthesized bifunctional Pt supported on HZSM-5 samples.

Sample	$S_{\text{BET}}^a$	$S_{\text{micro}}^b$	$S_{\text{ext}}^c$	$V_{\text{total}}^d$	$V_{\text{micro}}^e$	$V_{\text{ext}}^f$	$V_{\text{ext}}/V_{\text{total}}^g$
Pt@HZSM-5-NS(one)	558	343	215	1.01	0.17	0.84	0.83
Pt/HZSM-5-NS(imp)	497	286	211	1.02	0.14	0.88	0.86
Pt/HZSM-5-CON(imp)	379	363	16	0.19	0.17	0.02	0.11

<sup>a</sup>BET specific surface area, <sup>b</sup>microporous surface area, <sup>c</sup>external surface area, <sup>d</sup>total pore volume, <sup>e</sup>micropore volume, <sup>f</sup>external volume =  $V_{\text{total}} - V_{\text{micro}}$ , and <sup>g</sup>Fraction of external volume. #All surface areas and pore volumes are in the unit of  $\text{m}^2 \cdot \text{g}^{-1}$  and  $\text{cm}^3 \cdot \text{g}^{-1}$ , respectively.

**Table S3.** Acid properties of different bifunctional catalysts.

Sample	Acid amount <sup>a</sup> (mmol/g)		
	Weak (150–200 °C)	Strong (300–400 °C)	Total
Pt@HZSM-5-NS(one)	0.107	0.063	0.17
Pt/HZSM-5-NS(imp)	0.088	0.102	0.19
Pt/HZSM-5-CON(imp)	0.073	0.106	0.18

<sup>a</sup>determined by gaussian deconvolution of  $\text{NH}_3$ -TPD profiles.

**Table S4.** Catalytic performance of Pt@HZSM-5-NS(one) in terms of reactant conversion (%) and product selectivity (%) in the HDO of 4-propylphenol under reaction condition of 110 °C and H<sub>2</sub> atmospheric pressure as a function of time.

Reaction Time (min)	4-propylphenol conversion (%)	Selectivity (%)			
		Propyl cyclohexane	Propyl benzene	Propyl cyclohexanol	Propyl cyclohexanone
5	12.2	13.0	25.6	27.4	34.0
10	24.5	29.7	11.4	24.8	34.0
30	28.6	41.0	12.2	12.8	33.8
60	47.6	53.3	13.0	8.3	25.4
120	76.6	66.0	11.9	6.5	15.6
150	99.1	78.9	9.2	4.3	7.5
300	99.9	99.6	0.3	0.0	0.0
450	99.9	99.8	0.1	0.0	0.0
600	100.0	100.0	0.0	0.0	0.0

**Table S5.** Catalytic performance in the HDO of 4-propylphenol over different catalysts under reaction condition of 110 °C and H<sub>2</sub> atmospheric pressure taken from the reaction time of 10 h.

Sample	conversion (%)	Selectivity (%)			
		Propyl cyclohexane	Propyl benzene	Propyl cyclohexanol	Propyl cyclohexanone
Pt@NaZSM-5-NS(one)	33.4	1.0	0.7	46.9	51.4
Pt@HZSM-5-NS(one)	100.0	100.0	0.0	0.0	0.0
Pt/HZSM-5-NS(imp)	41.0	45.2	15.4	5.1	34.3
Pt/HZSM-5-CON(imp)	17.3	22.2	15.2	42.5	20.1