Supporting Information

Accurately Tuning the Pore Size and Acidity of Mesoporous Zeolites for Enhancing Catalytic Hydrocracking of Polypropylene

Ziru Wang^{a,b}, Li Gao^{b,c}, Xia Zhong^{b,c}, Ying Zhang^{b,d}, Mozaffar Shakeri^e, Xia Zhang^{a*}, Bingsen Zhang^{b,c*}

^a Department of Chemistry, College of Science, Northeastern University, Shenyang 110819, China.

^b Shenyang National Laboratory for Materials Science, Institute of Metal Research, Chinese Academy of Sciences, Shenyang 110016, Liaoning, China.

^c School of Materials Science and Engineering, University of Science and Technology of China, Shenyang 110016, Liaoning, China.

^d School of Petrochemical Engineering, Liaoning Petrochemical University, Fushun 113001, China.

^e Laboratory of Heterogeneous Catalysis Department of Chemical and Petroleum Engineering, Chemistry and Chemical Engineering Research Center of Iran, Tehran 16363, Iran.

* E-mail addresses: xzhang@mail.neu.edu.cn (Xia Zhang), bszhang@imr.ac.cn (Bingsen Zhang)



Figure S1. Photos of reactor setup.



Figure S2. HAADF-STEM image and the corresponding EDX elemental maps of H1 (a), H2 (b), and H3(c).



Figure S3. TEM images (a, b), HAADF-STEM image and the corresponding EDX elemental maps (c) of M1.



Figure S4. TEM images (a, b), HAADF-STEM image and the corresponding EDX elemental maps (c) of M2.



Figure S5. TEM images (a, b), HAADF-STEM image and the corresponding EDX elemental maps (c) of M3.



Figure S6. TEM images (a, b), HAADF-STEM image and the corresponding EDX elemental maps (c) of L1.



Figure S7. TEM images (a, b), HAADF-STEM image and the corresponding EDX elemental maps (c) of L2.



Figure S8. TEM images (a, b), HAADF-STEM image and the corresponding EDX elemental maps (c) of L3.



Figure S9. SEM images of (a) H1, (b) H2, (c) H3, (d) M1, (e) M2, (f) M3, (g) L1, (h) L2, (i) L3.



Figure S10. Histogram of specific surface area and pore size distribution of different Al-MCM-41



Figure S11. Conversion and product yield (a) of PP hydrocracking over M2 catalyst at different reaction temperatures. Conversion and product yield (b) of PP hydrocracking over M2 catalyst under different reaction time. Reaction conditions: 2MPa H₂, 30 mg PP, 30 mg catalyst.



Figure S12. TGA profiles of the used catalysts.



Figure S13. TEM images of used M1 (a), M2 (b), and M3 (c). XRD patterns of fresh and used M1 (d), M2 (e), and M3 (f). NH₃-TPD curves of fresh and used M1 (g), M2 (h), and M3 (i).



Figure S14. N_2 adsorption-desorption isotherms with pore size distribution curves of used catalysts.

| Catalyst | polyolefin | Temperature | Pressure | Time | Liquid Yield | Refs. |
|---------------------|------------|-------------|----------|------|--|-------|
| | | (°C) | (MPa) | (h) | (%) | |
| 5Ru/H-β | РР | 215 | 3 | 16 | 67 | [1] |
| 1Pt/Al-MCM-48 | LDPE | 300 | 4 | 4 | 85.9(C ₉ -C ₁₅) | [2] |
| Ni/SiO ₂ | LDPE | 280 | 3 | 4 | 81.1 | [3] |
| 5Ru/C | PP | 250 | 4 | 14 | 36 | [4] |
| Pt/HY | LDPE | 300 | 3 | 2 | 32.8 | [5] |
| Pt/C | LDPE | 300 | 1.5 | 24 | 20 (C ₅ -C ₂₀) | [6] |
| Al-MCM-41 | РР | 220 | 2 | 12 | 70.2 | This |
| | | | | | | work |

 Table S1. Comparison of polyolefins upcycling activity of M1 catalyst and other reported catalysts.

Reference

1. J. E. Rorrer, A. M. Ebrahim, Y. Questell-Santiago, J. Zhu, C. Troyano-Valls, A. S. Asundi, A. E. Brenner, S. R. Bare, C. J. Tassone, G. T. Beckham and Y. Roman-Leshkov, *ACS Catal.*, 2022, **12**, 13969-13979.

2. Y. Liu, Reactions, 2020, 1, 195-209.

3. Z. Zhao, Z. Li, X. Zhang, T. Li, Y. Li, X. Chen and K. Wang, *Environ. Pollut.*, 2022, **313**, 120154.

4. J. E. Rorrer, C. Troyano-Valls, G. T. Beckham and Y. Román-Leshkov, ACS Sustain. Chem. Eng., 2021, 9, 11661-11666.

5. X. Wu, X. Wang, L. Zhang, X. Wang, S. Song and H. Zhang, *Angew. Chem.*, 2024, **136**, e202317594.

6. S. D. Jaydev, A. J. Martín and J. Pérez-Ramírez, *ChemSusChem*, 2021, 14, 5179-5185.