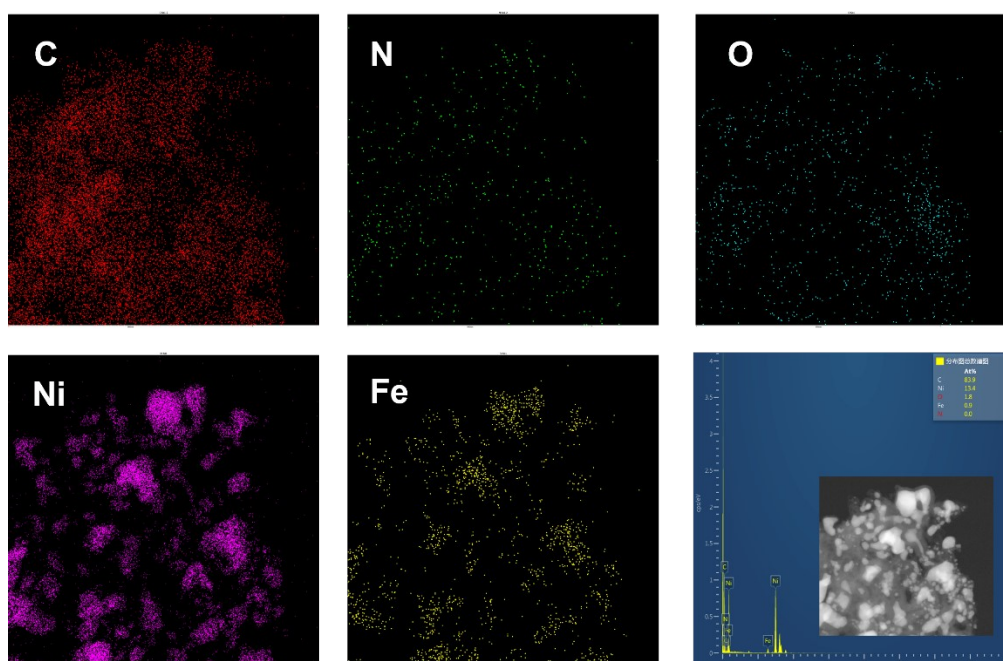


## Selective Synthesis of Secondary Imines via Reductive Coupling of Aldehydes with Ammonia over Encapsulated NiFe Alloy Catalysts

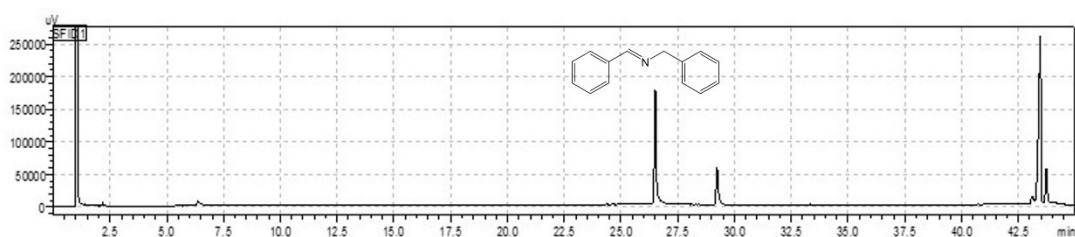
Yulong Lei, Jianguo Liu\*

Key Laboratory of Energy Thermal Conversion and Control of Ministry of Education, School of Energy and Environment, Southeast University, Nanjing 210096, PR China.

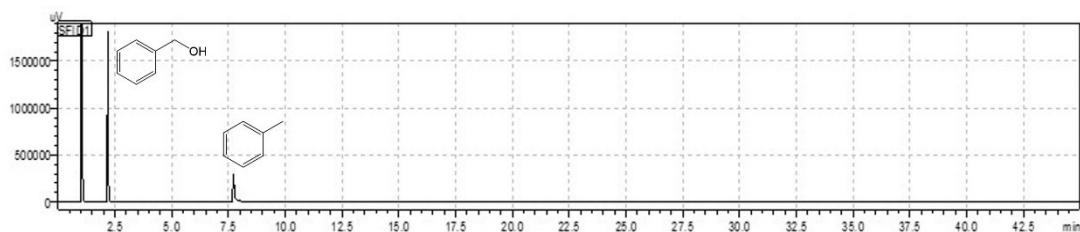
\* Corresponding authors' e-mail: liujg@seu.edu.cn



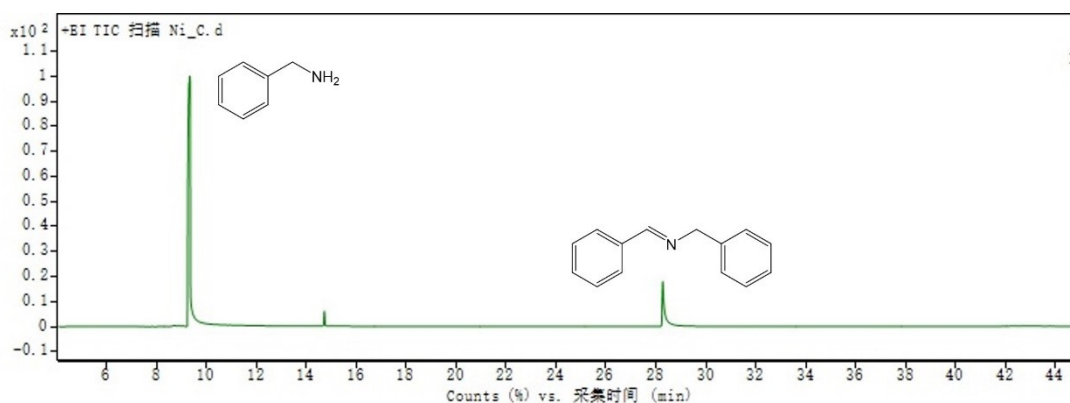
**Fig. S1.** EDS analysis of catalyst.



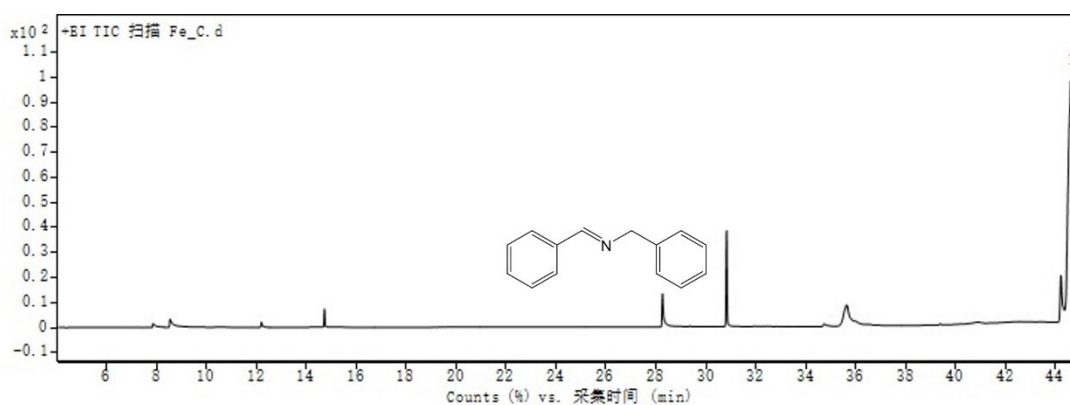
**Fig. S2.** Gas chromatogram (GC) of the reaction for entry 1 in Table 1. Reaction conditions: BzA (0.5 mmol), cat. (10 mg), NH<sub>3</sub> (7 M in MeOH, 5 mL), temperature (100 °C), H<sub>2</sub> pressure (2.0 MPa), reaction time (4 h), stirring speed (500 rpm).



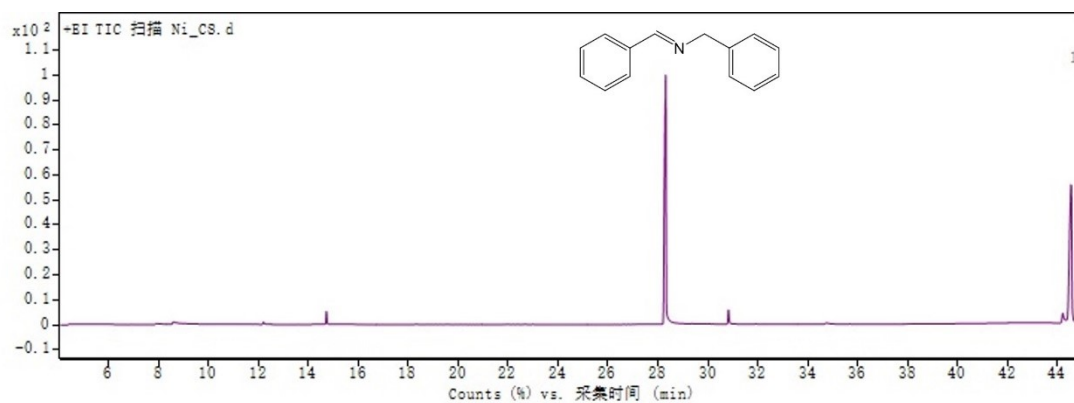
**Fig. S3.** Gas chromatogram (GC) of the reaction for entry 2 in Table 1. Reaction conditions: BzA (0.5 mmol), cat. (10 mg), NH<sub>3</sub> (7 M in MeOH, 5 mL), temperature (100 °C), H<sub>2</sub> pressure (2.0 MPa), reaction time (4 h), stirring speed (500 rpm).



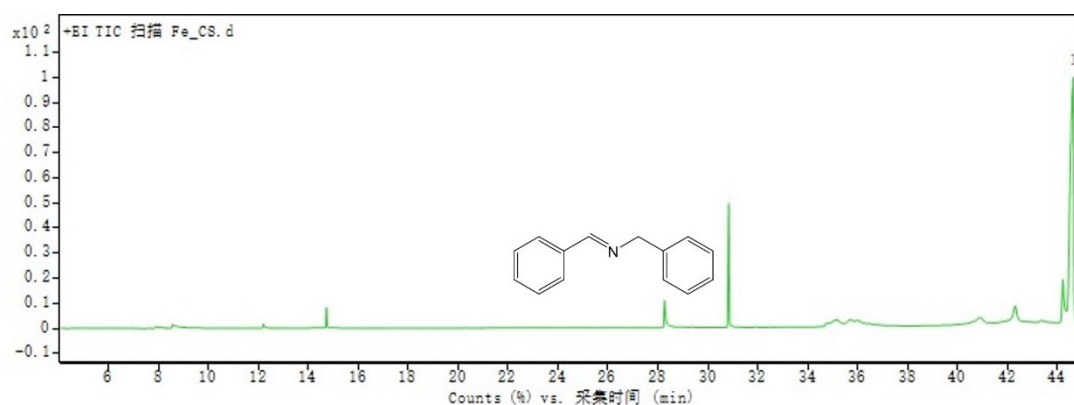
**Fig. S4.** Gas chromatogram (GC) of the reaction for entry 3 in Table 1. Reaction conditions: BzA (0.5 mmol), cat. (10 mg), NH<sub>3</sub> (7 M in MeOH, 5 mL), temperature (100 °C), H<sub>2</sub> pressure (2.0 MPa), reaction time (4 h), stirring speed (500 rpm).



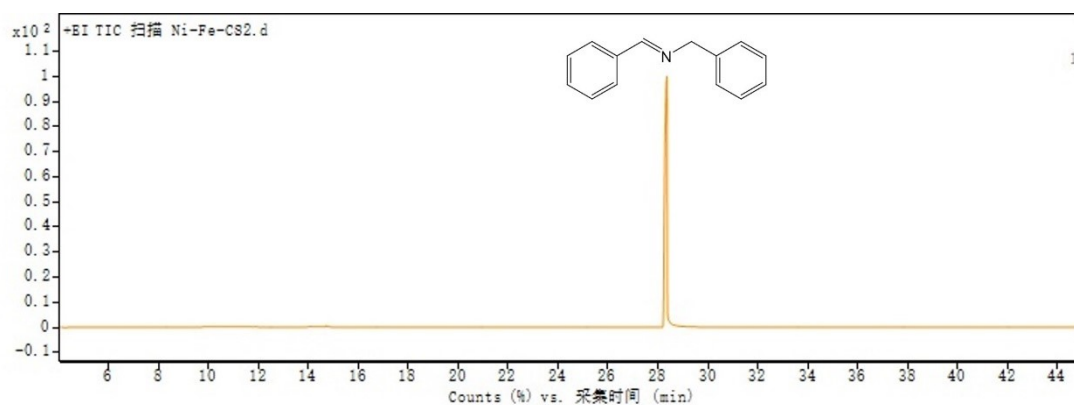
**Fig. S5.** Gas chromatogram (GC) of the reaction for entry 4 in Table 1. Reaction conditions: BzA (0.5 mmol), cat. (10 mg), NH<sub>3</sub> (7 M in MeOH, 5 mL), temperature (100 °C), H<sub>2</sub> pressure (2.0 MPa), reaction time (4 h), stirring speed (500 rpm).



**Fig. S6.** Gas chromatogram (GC) of the reaction for entry 5 in Table 1. Reaction conditions: BzA (0.5 mmol), cat. (10 mg), NH<sub>3</sub> (7 M in MeOH, 5 mL), temperature (100 °C), H<sub>2</sub> pressure (2.0 MPa), reaction time (4 h), stirring speed (500 rpm).

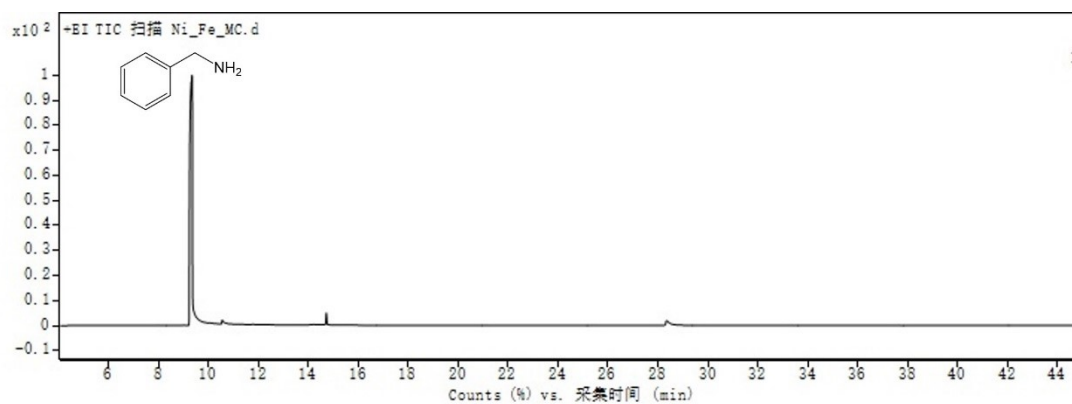


**Fig. S7.** Gas chromatogram (GC) of the reaction for entry 6 in Table 1. Reaction conditions: BzA (0.5 mmol), cat. (10 mg), NH<sub>3</sub> (7 M in MeOH, 5 mL), temperature (100 °C), H<sub>2</sub> pressure (2.0 MPa), reaction time (4 h), stirring speed (500 rpm).

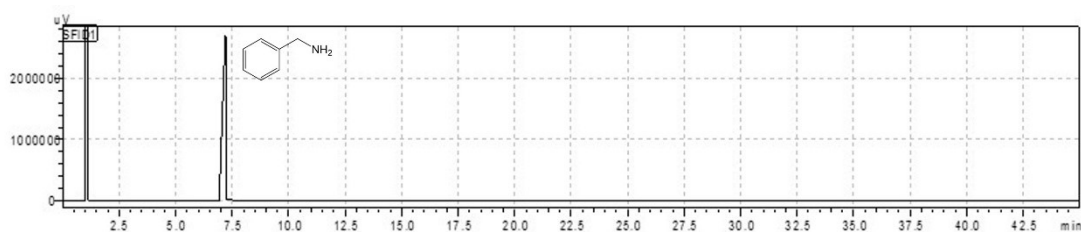


**Fig. S8.** Gas chromatogram (GC) of the reaction for entry 7 in Table 1. Reaction

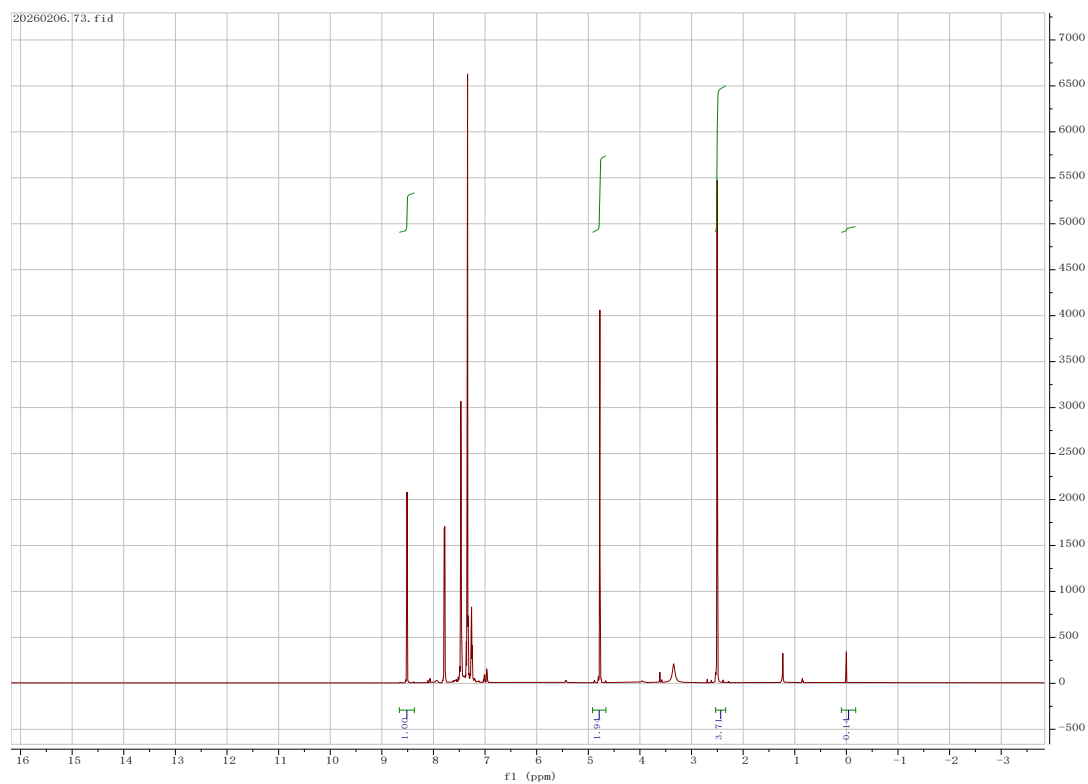
conditions: BzA (0.5 mmol), cat. (10 mg), NH<sub>3</sub> (7 M in MeOH, 5 mL), temperature (100 °C), H<sub>2</sub> pressure (2.0 MPa), reaction time (4 h), stirring speed (500 rpm).



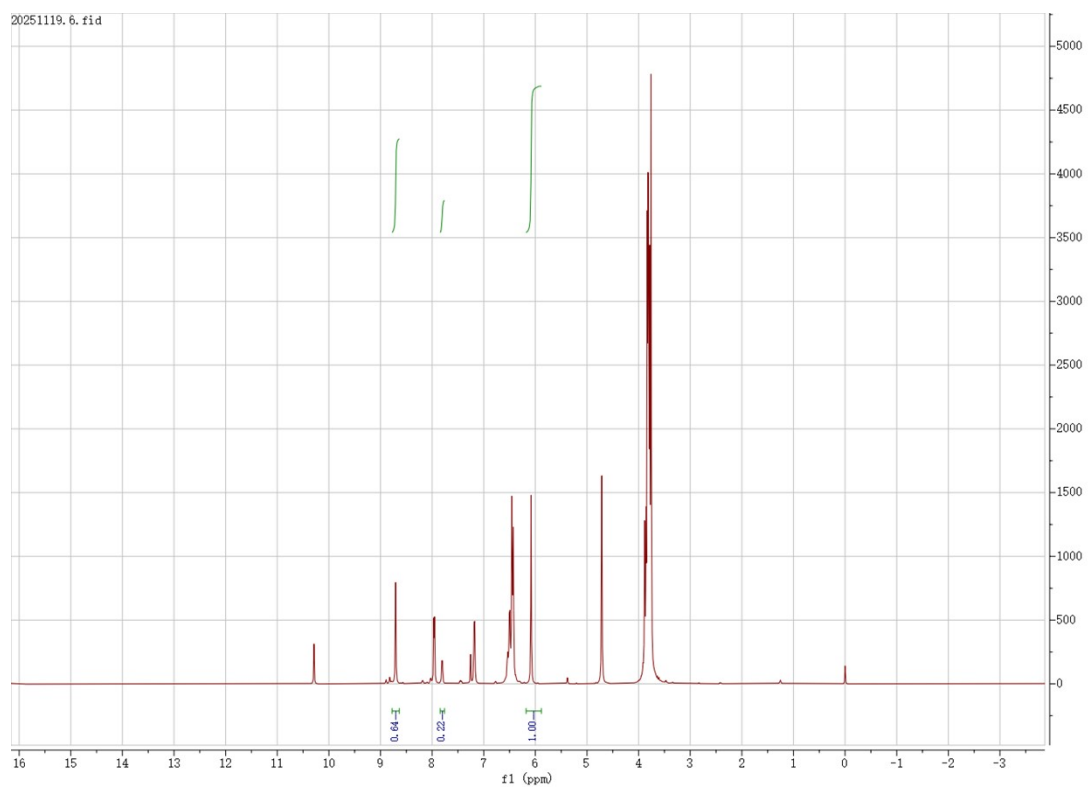
**Fig. S9.** Gas chromatogram (GC) of the reaction for entry 8 in Table 1. Reaction conditions: BzA (0.5 mmol), cat. (10 mg), NH<sub>3</sub> (7 M in MeOH, 5 mL), temperature (100 °C), H<sub>2</sub> pressure (2.0 MPa), reaction time (4 h), stirring speed (500 rpm).



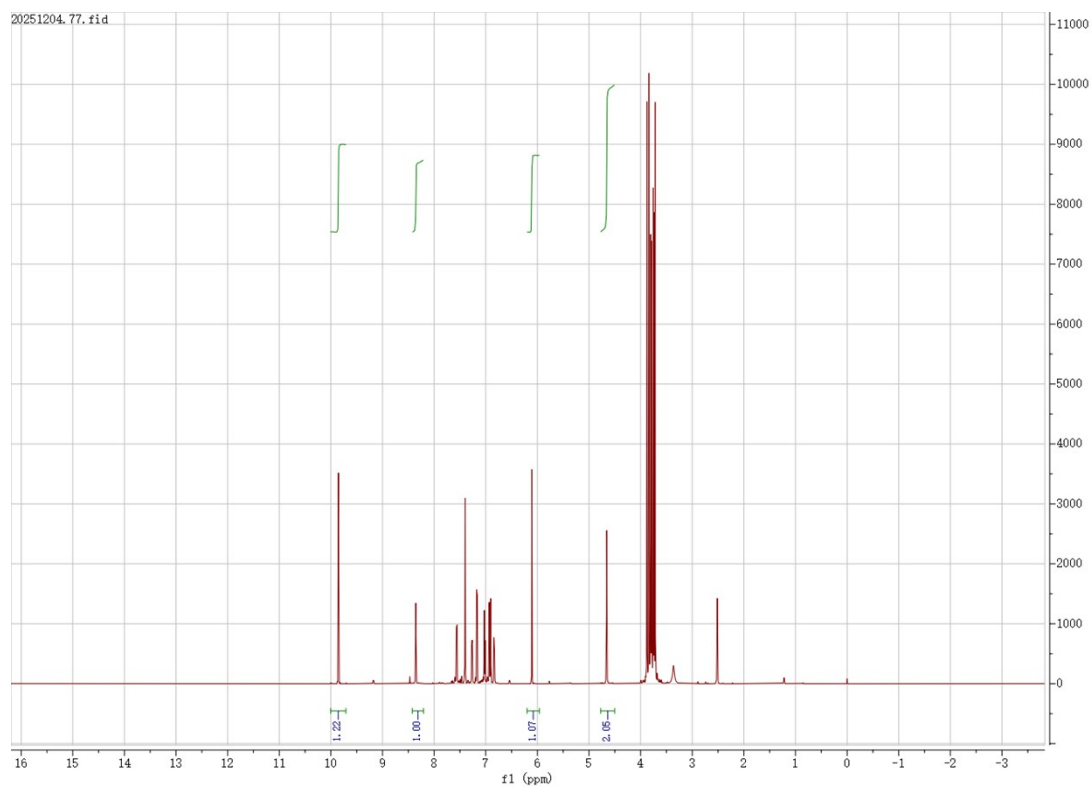
**Fig. S10.** Gas chromatogram (GC) of the reaction for entry 9 in Table 1. Reaction conditions: BzA (0.5 mmol), cat. (10 mg), NH<sub>3</sub> (7 M in MeOH, 5 mL), temperature (100 °C), H<sub>2</sub> pressure (2.0 MPa), reaction time (4 h), stirring speed (500 rpm).



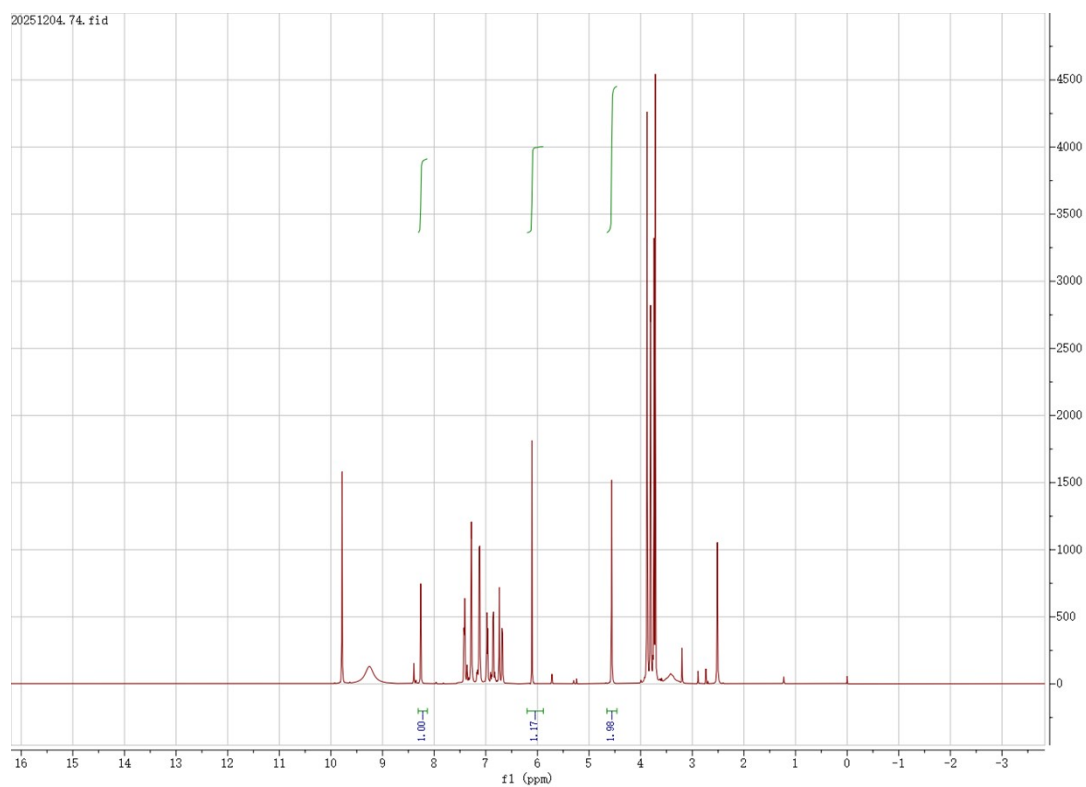
**Fig. S11.** Proton NMR spectrum of the reaction of entry 1 in Table 9<sup>1</sup>.



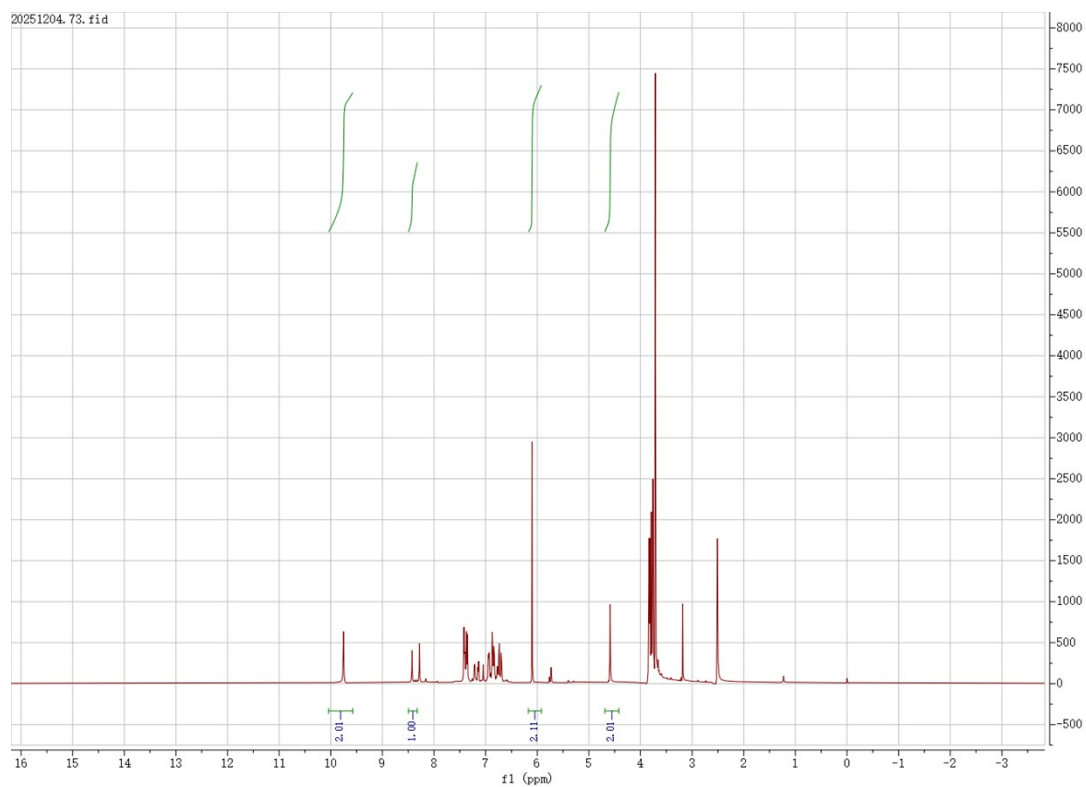
**Fig. S12.** Proton NMR spectrum of the reaction of entry 2 in Table 9<sup>2</sup>.



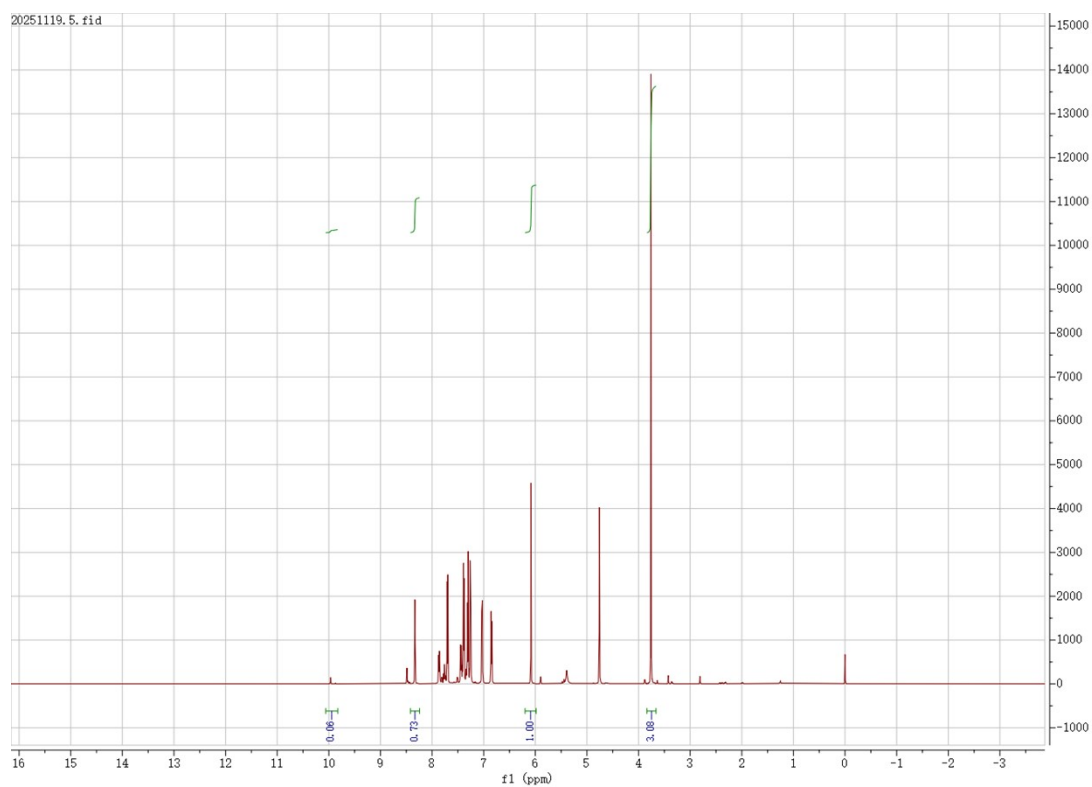
**Fig. S13.** Proton NMR spectrum of the reaction of entry 3 in Table 9<sup>1</sup>.



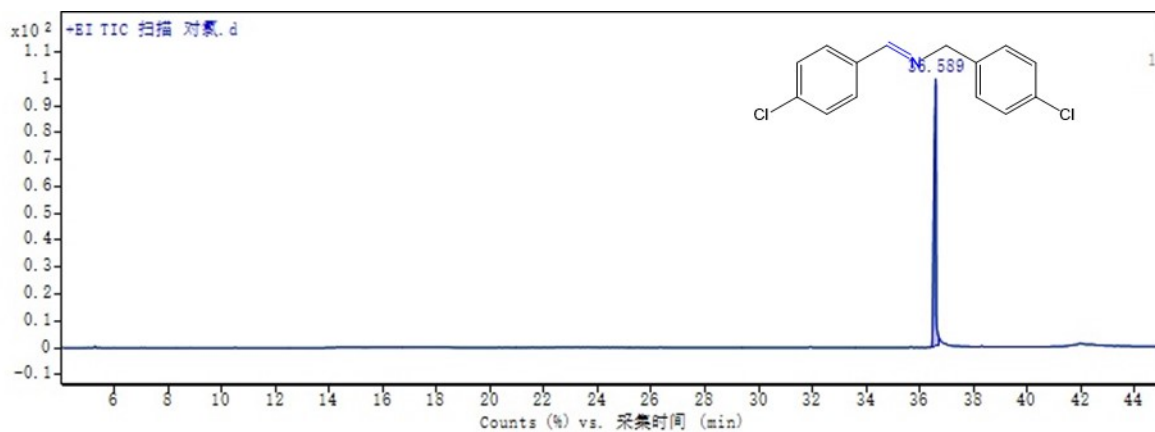
**Fig. S14.** Proton NMR spectrum of the reaction of entry 4 in Table 9<sup>3</sup>.



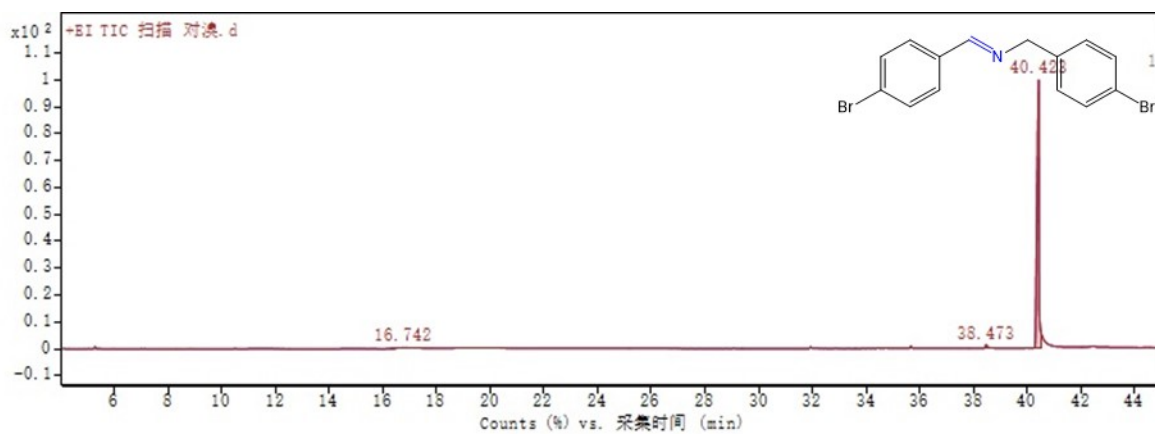
**Fig. S15.** Proton NMR spectrum of the reaction of entry 5 in Table 9<sup>4</sup>.



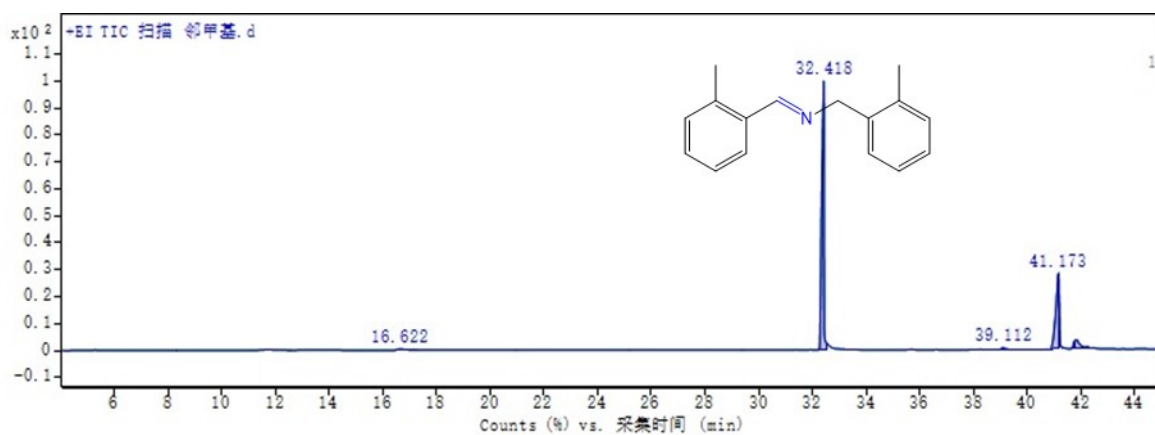
**Fig. S16.** Proton NMR spectrum of the reaction of entry 6 in Table 9<sup>1,5,6</sup>.



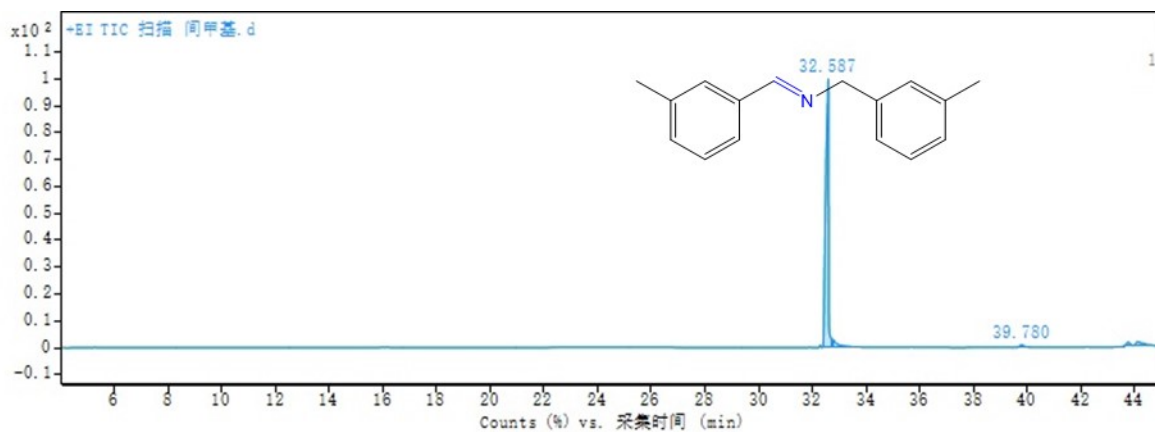
**Fig. S17.** GC chromatogram of the reaction of entry 7 in Table 9.



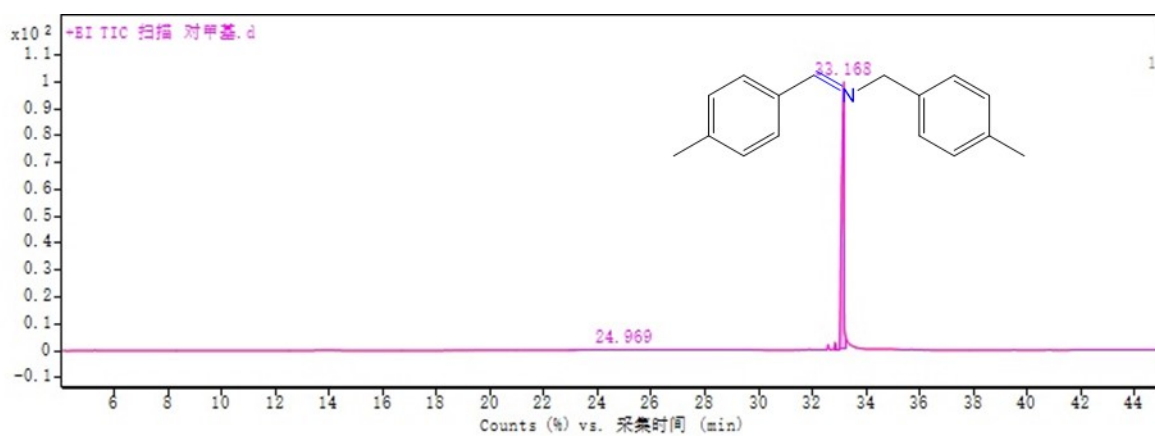
**Fig. S18.** GC chromatogram of the reaction of entry 8 in Table 9.



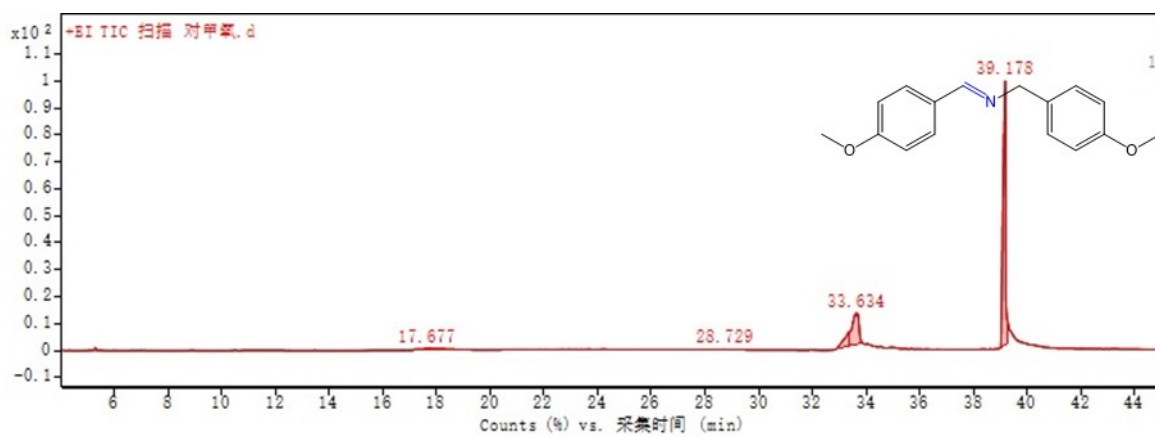
**Fig. S19.** GC chromatogram of the reaction of entry 9 in Table 9.



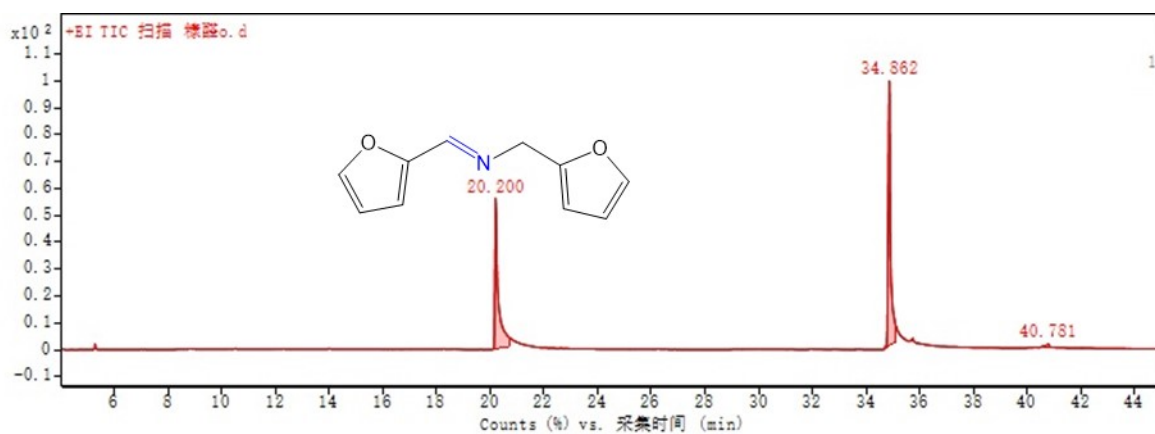
**Fig. S20.** GC chromatogram of the reaction of entry 10 in Table 9.



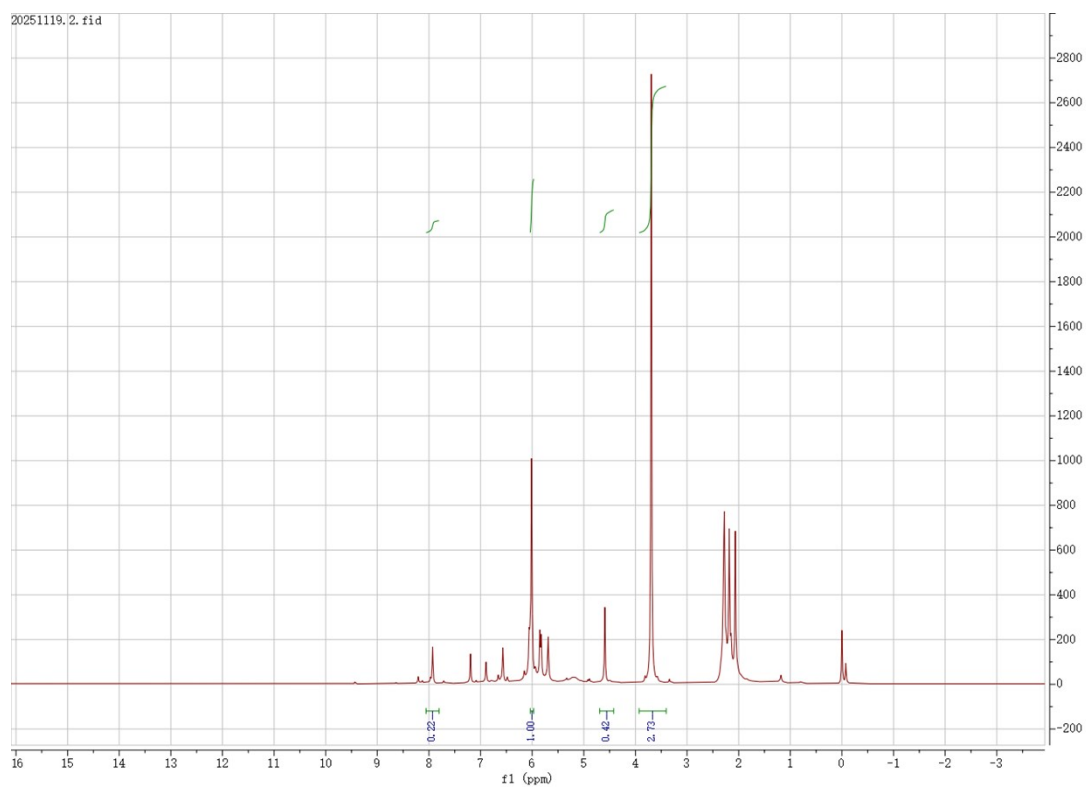
**Fig. S21.** GC chromatogram of the reaction of entry 11 in Table 9.



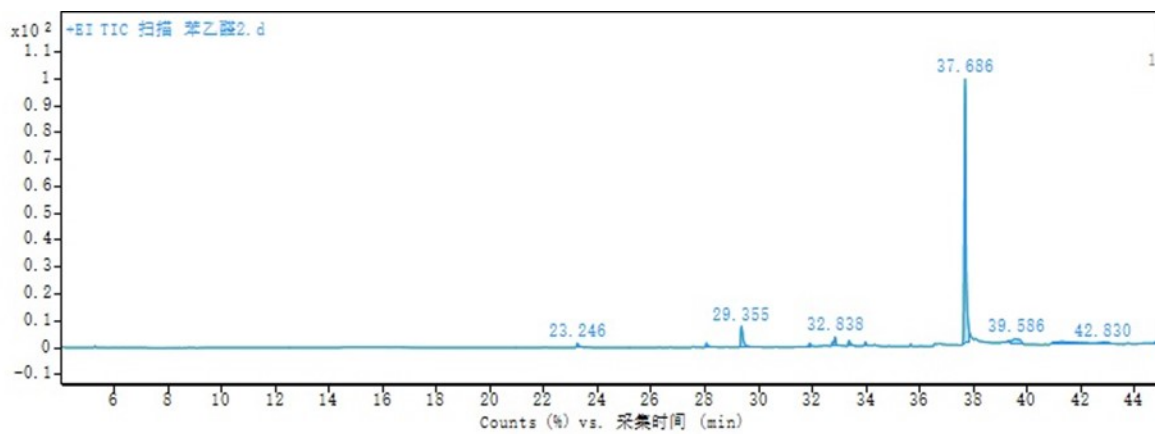
**Fig. S22.** GC chromatogram of the reaction of entry 12 in Table 9.



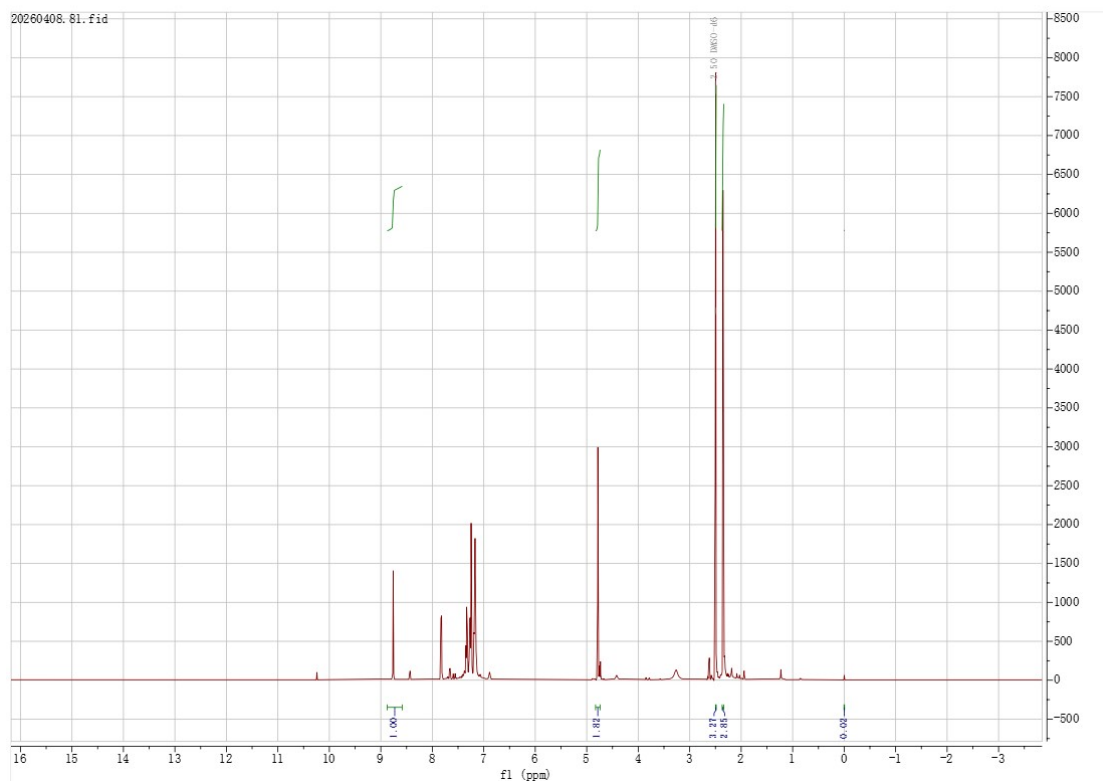
**Fig. S23.** GC chromatogram of the reaction of entry 13 in Table 9.



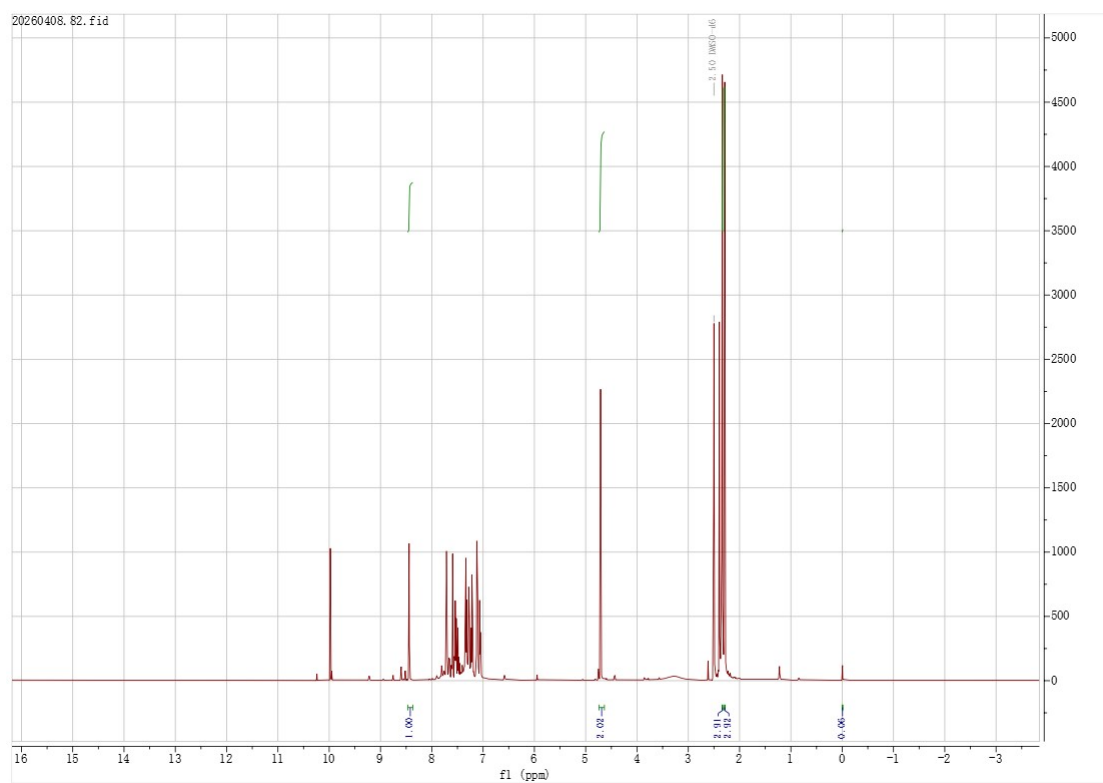
**Fig. S24.** Proton NMR spectrum of the reaction of entry 14 in Table 9<sup>7</sup>.



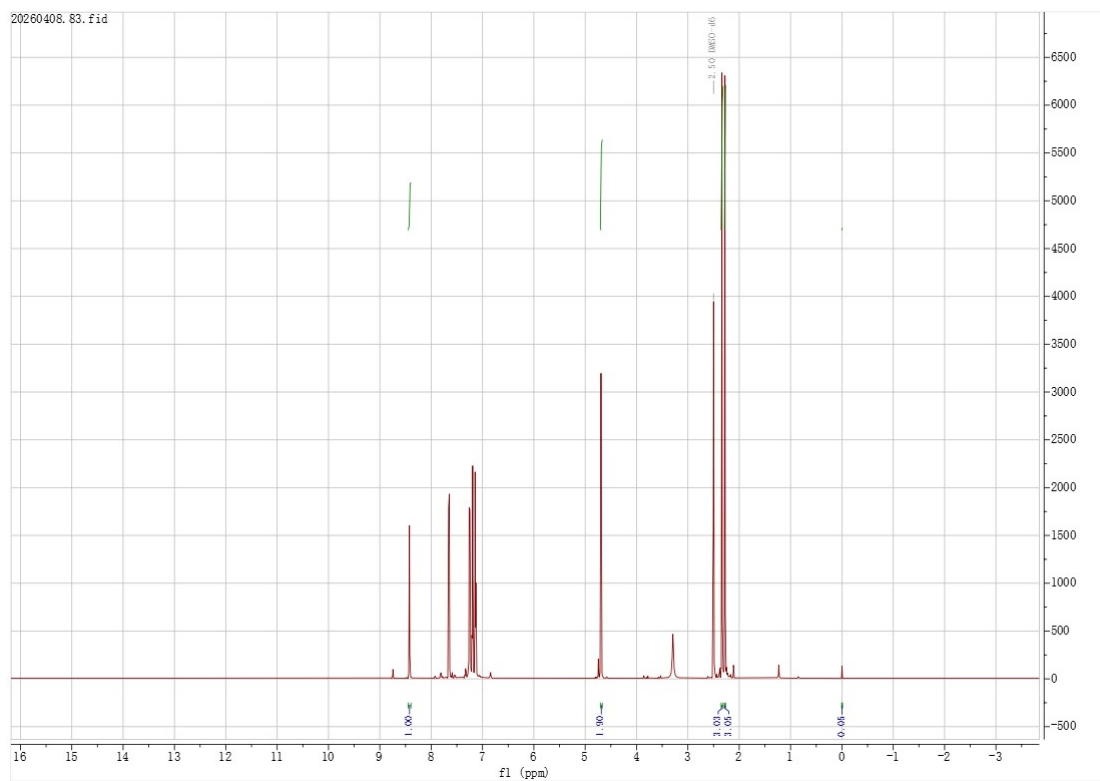
**Fig. S25.** GC chromatogram of the reaction of entry 15 in Table 9<sup>1,5,6</sup>.



**Fig. S26.** Proton NMR spectrum of the reaction of entry 9 in Table 9<sup>1,5,6</sup>.



**Fig. S27.** Proton NMR spectrum of the reaction of entry 10 in Table 9<sup>1,5,6</sup>.



**Fig. S28.** Proton NMR spectrum of the reaction of entry 11 in Table 9<sup>1,5,6</sup>.

## Reference

- 1 A. T. Murray, R. King, J. V. G. Donnelly, M. J. H. Dowley, F. Tuna, D. Sells, M. P. John and D. R. Carbery, *ChemCatChem*, 2016, **8**, 510–514.
- 2 D. V. Jawale, E. Gravel, E. Villemin, N. Shah, V. Geertsen, I. N. N. Namboothiri and E. Doris, *Chem. Commun.*, 2014, **50**, 15251–15254.
- 3 N. Wang, J. Liu, L. Tang, X. Wei, C. Wang, X. Li and L. Ma, *ACS Appl. Mater. Interfaces*, 2021, **13**, 24966–24975.
- 4 P. H. Wu, Y. Q. Li, Y. You, C. J. Huang, H. B. Xie and M. Q. Zhang, *Industrial Crops and Products*, 2024, **215**, 118658.
- 5 F. Bottaro, A. Takallou, A. Chehaiber and R. Madsen, *European Journal of Organic Chemistry*, 2019, **2019**, 7164–7168.
- 6 X. Tan, Y. Wang, K. Zhang, S. Zhang, M. Hou and X. Lang, *Applied Catalysis B: Environment and Energy*, 2026, **384**, 126136.
- 7 H. Zou, Y. Jin, L. Chen and J. Chen, *ACS Catal.*, 2025, **15**, 2017–2032.