Supporting Information

Direct Imines Formation by Oxidative Coupling of Alcohols and Amines using Supported Manganese oxide under Air atmosphere

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Table S1. Physical properties of various catalysts

Catalyst	S_{BET} ($m^2 \cdot g^{-1}$)	Vol ($cm^3 \cdot g^{-1}$)
HAP-pure	37.9	0.14
MnO_x/HAP	36.3	0.13
MnO_x/TiO_2	46.4	0.32
MnO_x/MgO	107.4	0.45
MnO_x/Al_2O_3	148.0	0.27
MnO_x/SBA-15	369.2	0.86

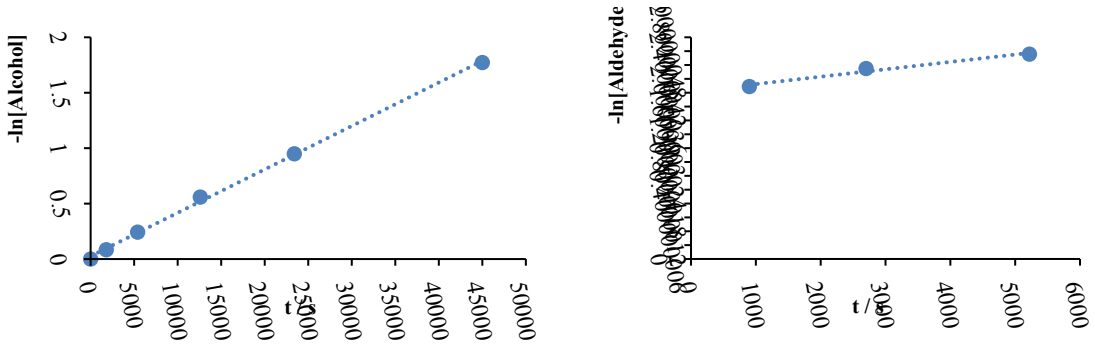


Figure S1. Time-on-stream course of conversion.

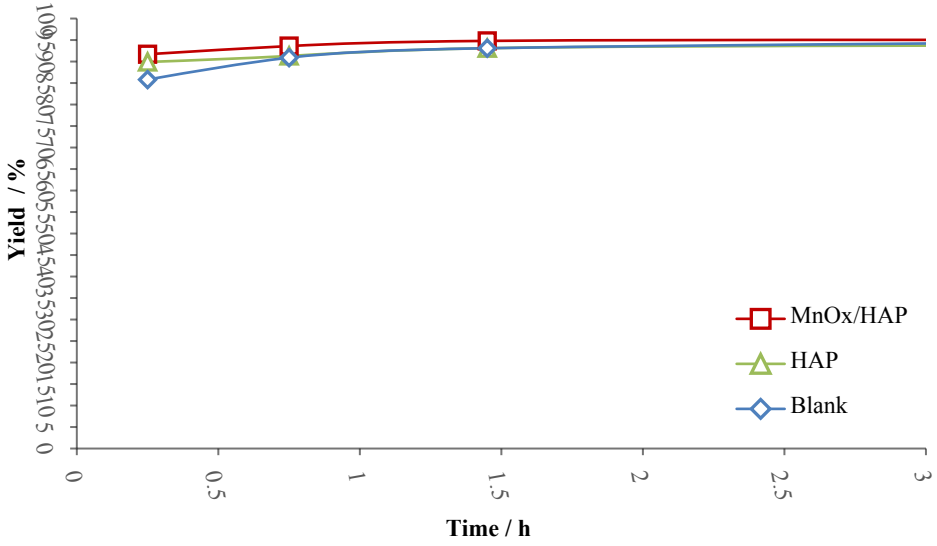


Figure S2. Time course of the reaction between benzaldehyde (1 mmol) and aniline (1 mmol) over various catalysts (125mg) at 80 °C under air balloon.

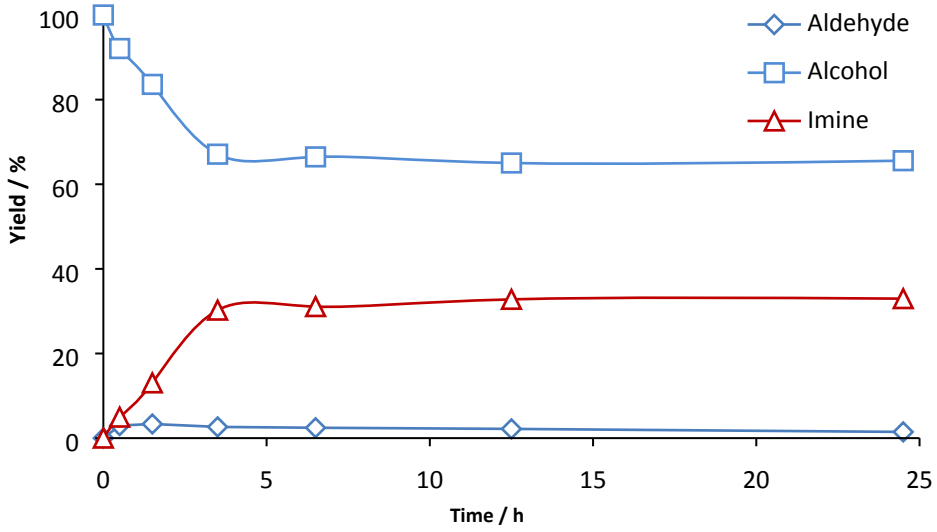


Figure S3. Hot filtration test for oxidative coupling of benzyl alcohol and aniline over MnO_x/HAP in 3.5h

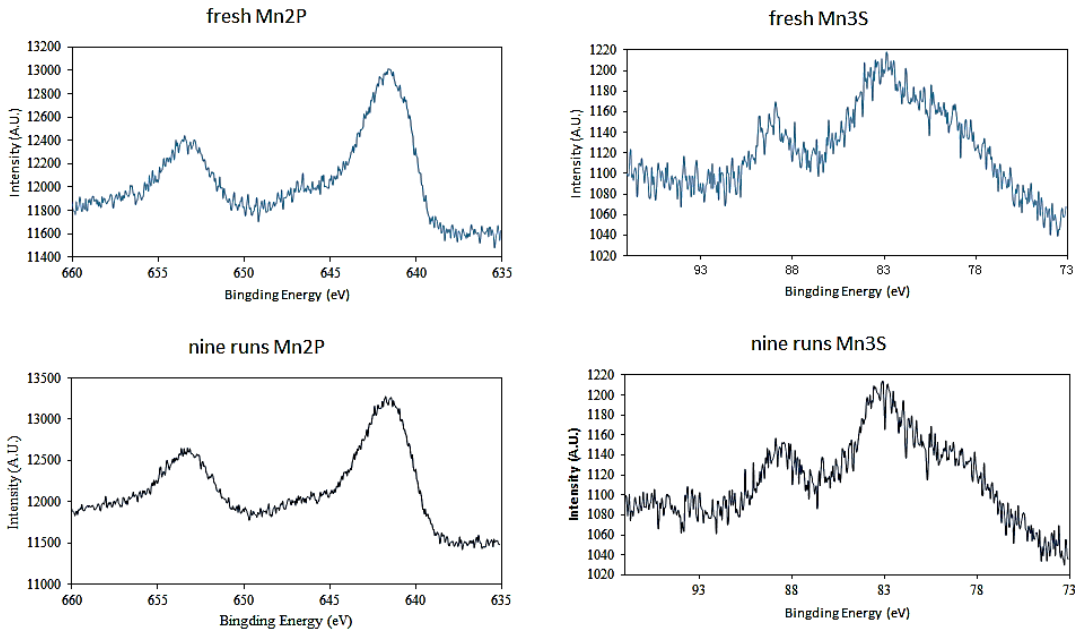
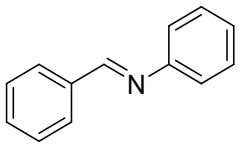
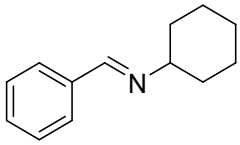


Figure S4. XPS of fresh MnO_x/HAP (**a**) and MnO_x/HAP after the ninth cycle of use (**b**)

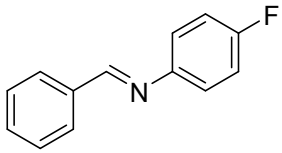
Characterization of Typical Products:



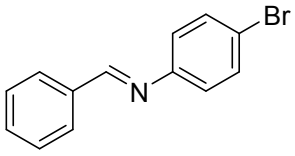
***N*-benzylideneaniline** Yellow solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.46 (s, 1H), 7.91 (d, J = 4.8 Hz, 2H), 7.48 (s, 3H), 7.39 (t, J = 7.2 Hz, 2H), 7.23 (t, J = 9.2 Hz, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 160.97 (s), 152.69 (s), 136.82 (s), 131.95 (s), 129.70 (s), 129.37 (d, J = 3.9 Hz), 126.51 (s), 121.44 (s).



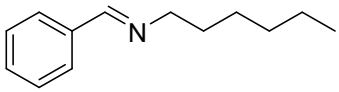
***N*-Benzylidenecyclohexylamine** Yellow oil. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.31 (s, 1H), 7.81 – 7.67 (m, 2H), 7.37 (t, J = 11.5 Hz, 3H), 3.29 – 3.05 (m, 1H), 1.85 (d, J = 12.7 Hz, 2H), 1.79 – 1.50 (m, 5H), 1.47 – 1.19 (m, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 158.49 (s), 136.61 (s), 130.25 (s), 128.45 (s), 128.02 (s), 69.92 (s), 34.35 (s), 25.64 (s), 24.78 (s).



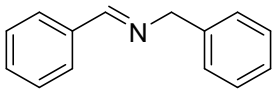
***N*-(4-fluorophenyl)-1-phenylmethanimine** Brownish black solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.44 (s, 1H), 7.94 – 7.82 (m, 2H), 7.52 – 7.42 (m, 3H), 7.20 (ddd, J = 10.1, 5.2, 2.7 Hz, 2H), 7.13 – 7.02 (m, 2H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 160.90 (s), 151.17 (s), 136.13 (s), 132.37 (s), 131.82 (s), 129.05 (d, J = 7.9 Hz), 122.80 (s), 119.50 (s).



***N*-(4-bromophenyl)-1-phenylmethanimine** Brownish black solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.41 (s, 1H), 7.95 – 7.83 (m, 2H), 7.48 (dd, *J* = 14.4, 7.0 Hz, 5H), 7.09 (d, *J* = 8.6 Hz, 2H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 160.90 (s), 151.17 (s), 136.13 (s), 132.37 (s), 131.82 (s), 129.05 (d, *J* = 7.9 Hz), 122.80 (s), 119.50 (s).

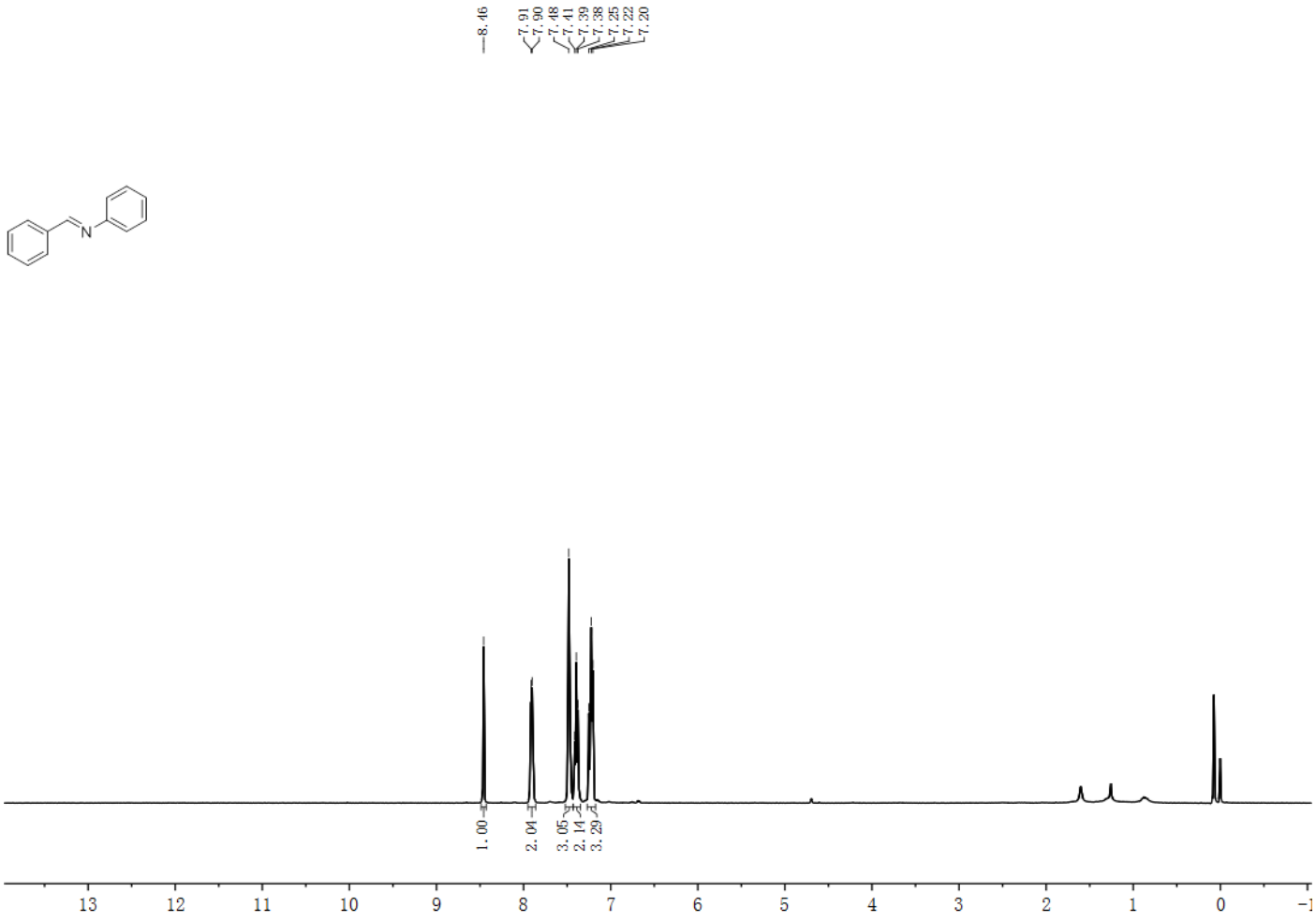


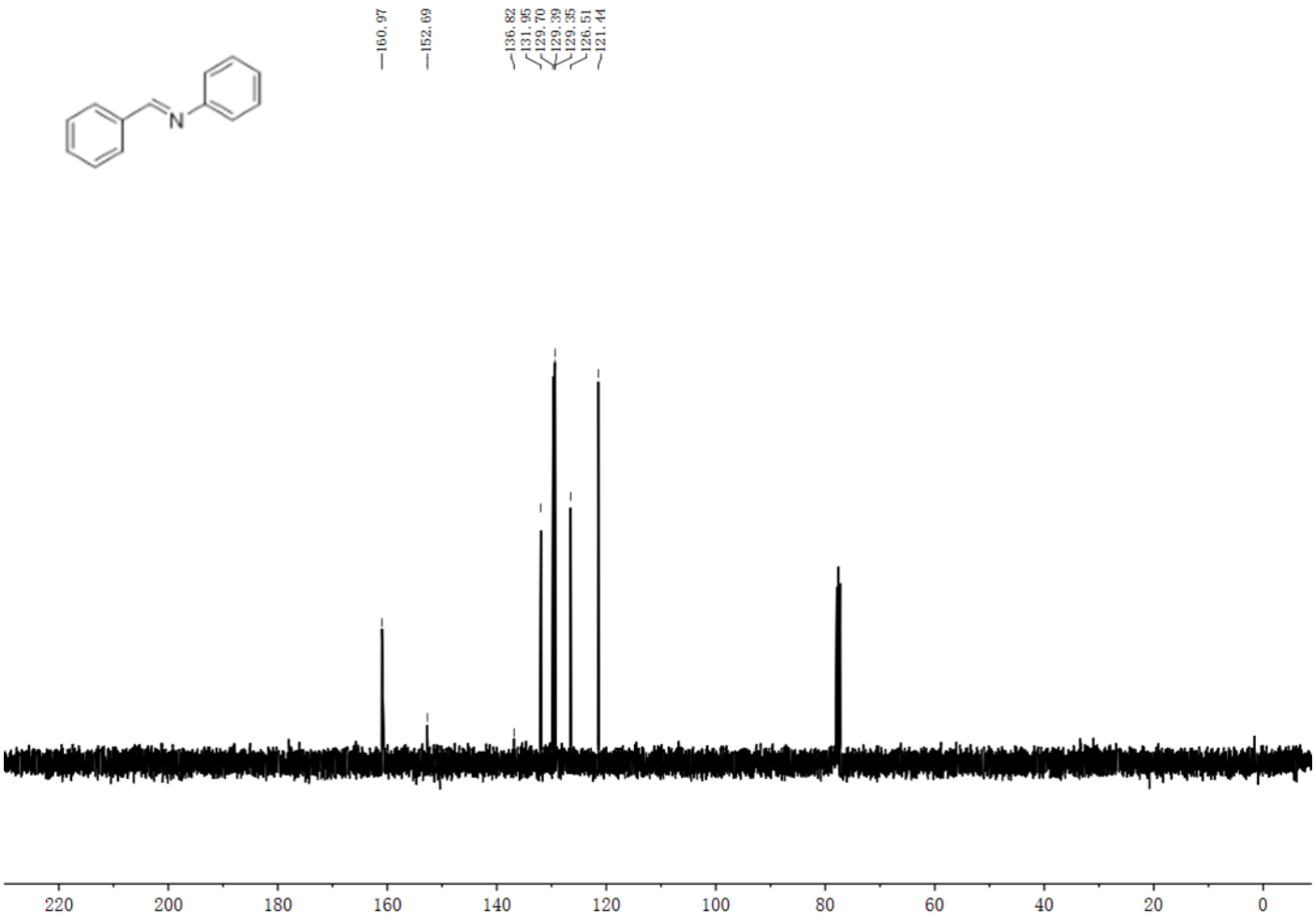
***N*-hexyl-1-phenylmethanimine** Yellow oil. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.26 (s, 1H), 7.71 (dt, *J* = 7.7, 3.2 Hz, 2H), 7.43 – 7.35 (m, 3H), 3.60 (td, *J* = 7.1, 1.0 Hz, 2H), 1.74 – 1.62 (m, 2H), 1.42 – 1.22 (m, 6H), 0.89 (t, *J* = 6.9 Hz, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 160.92 (s), 136.63 (s), 130.64 (s), 128.79 (s), 128.24 (s), 62.05 (s), 31.91 (s), 31.13 (s), 27.27 (s), 22.85 (s), 14.30 (s).



***N*-Benzylidenebenzylamine** Yellow oil.^{1}H NMR (400 MHz, $CDCl_3$) δ 8.42 (s, 1H), 7.83 (dd, *J* = 6.7, 2.9 Hz, 2H), 7.48 – 7.41 (m, 3H), 7.37 (dd, *J* = 10.2, 2.8 Hz, 4H), 7.30 (dt, *J* = 8.8, 4.4 Hz, 1H), 4.86 (s, 2H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 162.14 (s), 139.51 (s), 136.37 (s), 130.93 (s), 128.78 (s), 128.67 (s), 128.46 (s), 128.16 (s), 127.16 (s), 65.22 (s).

^{1}H NMR and ^{13}C NMR Spectra of the Typical Products

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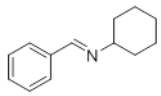

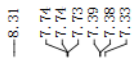
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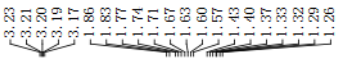
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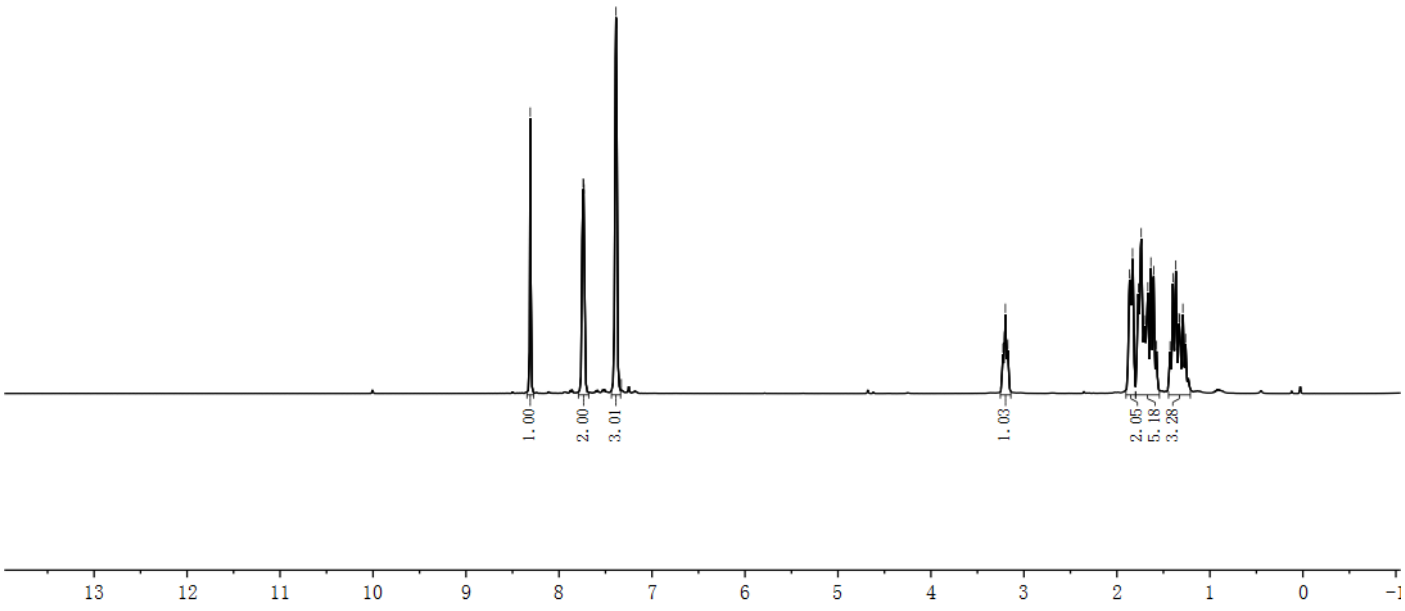
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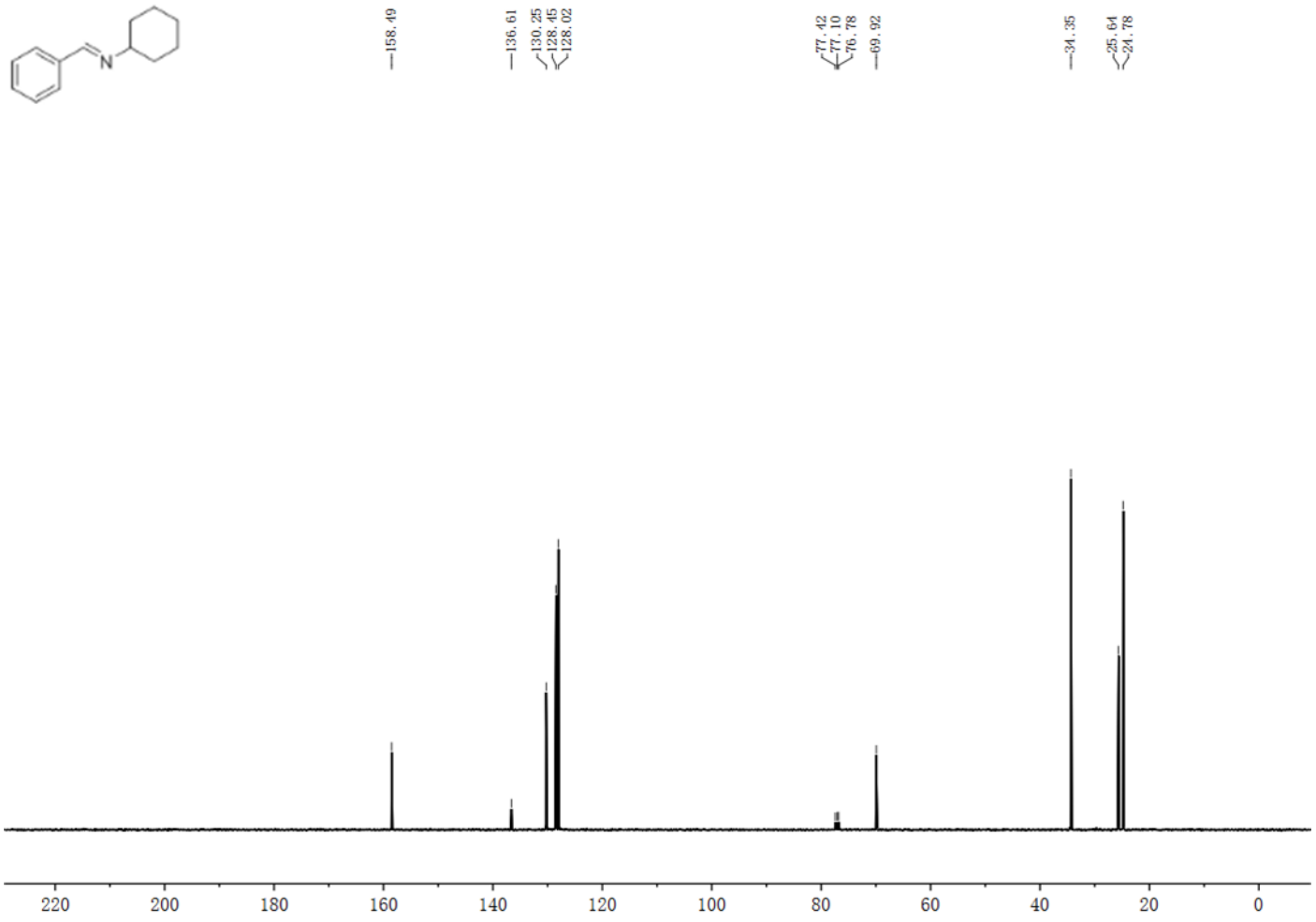
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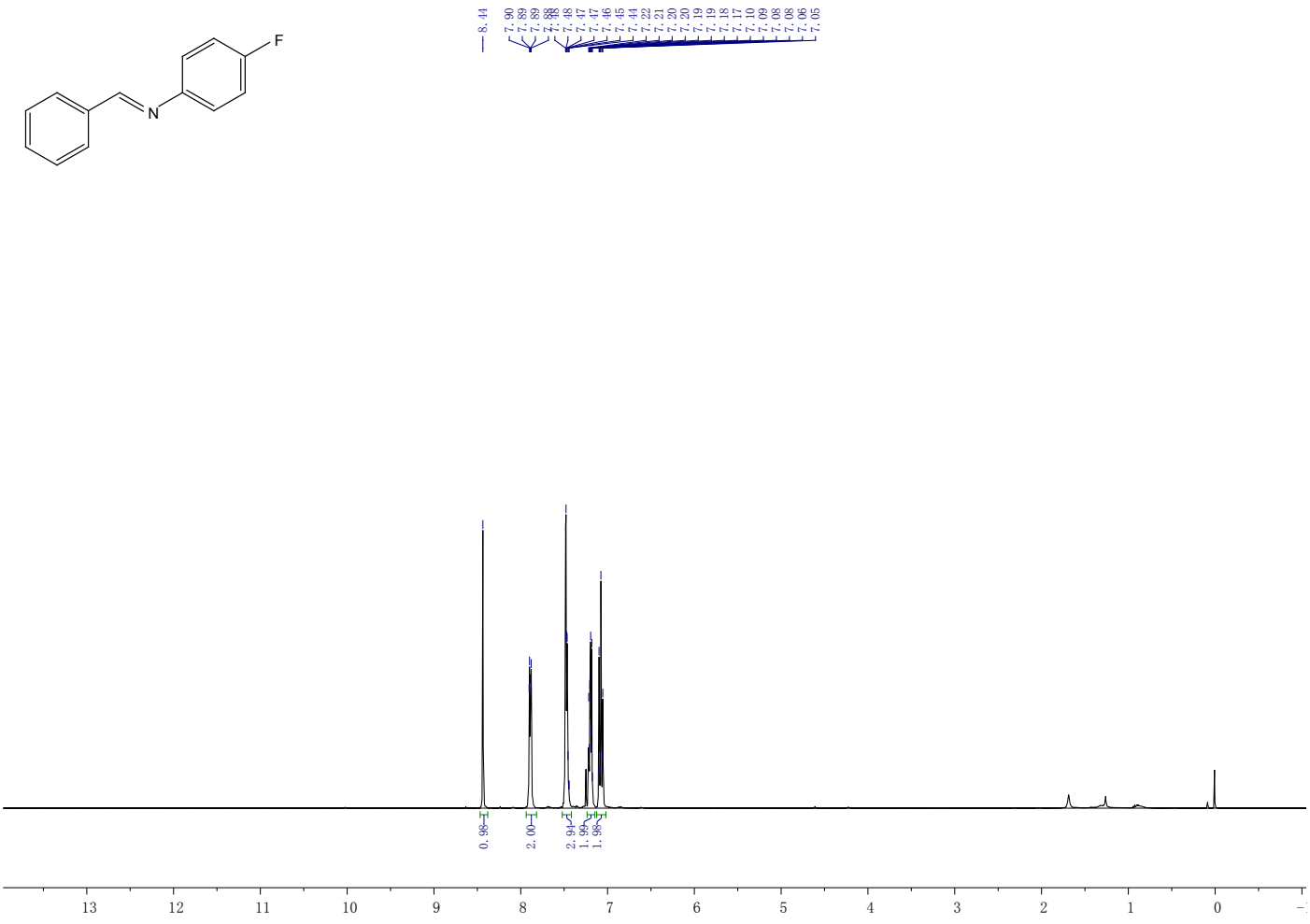
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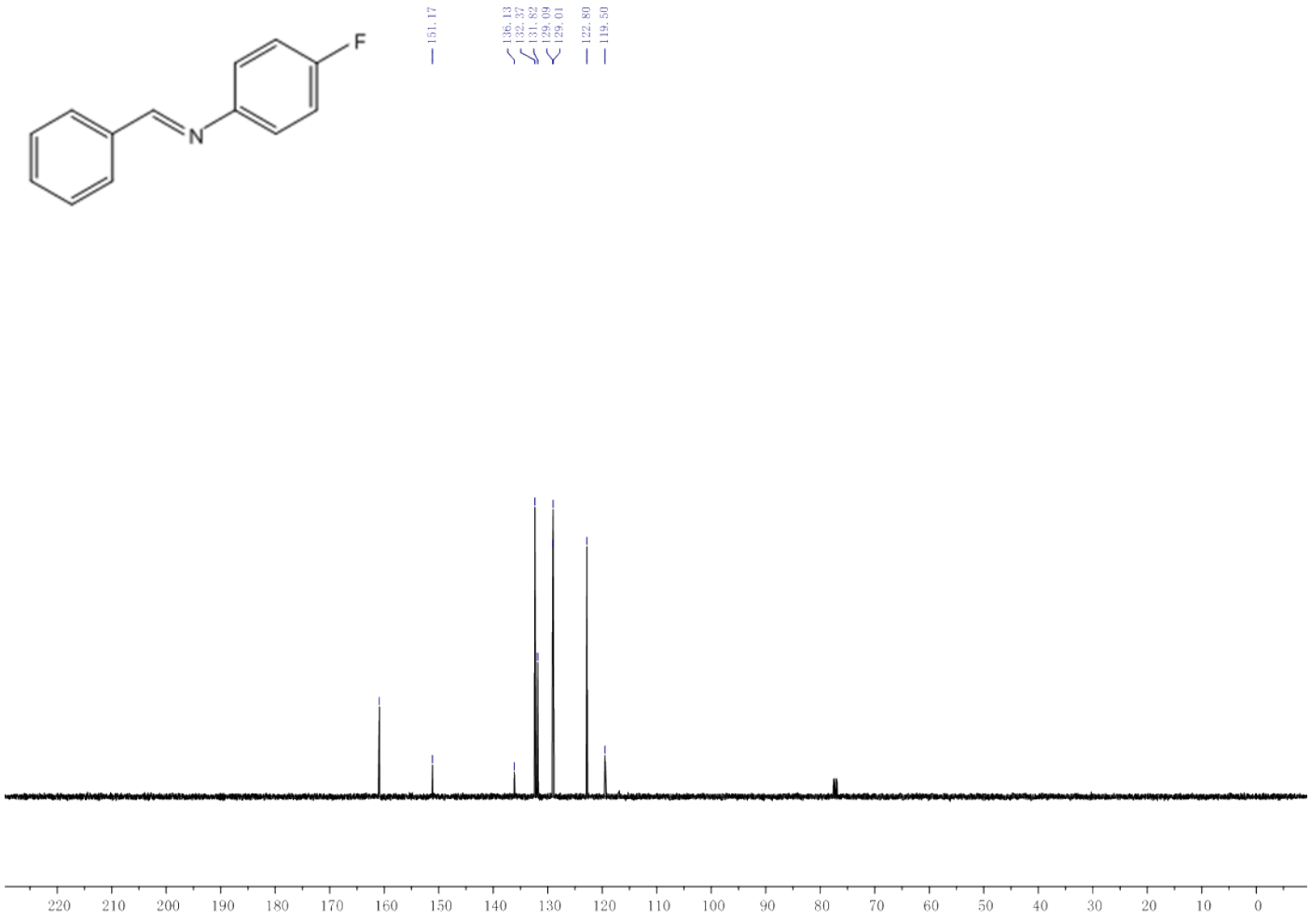
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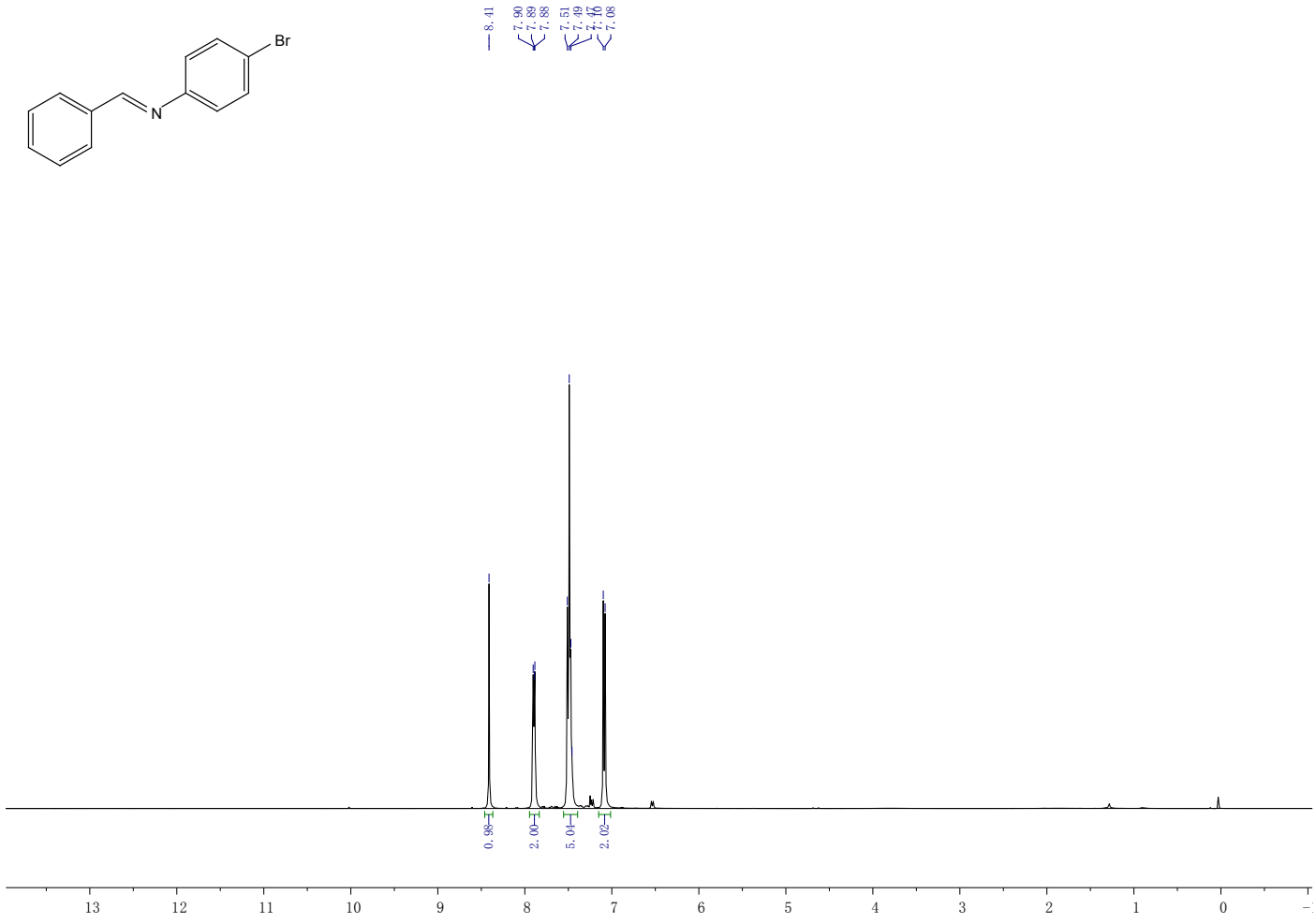
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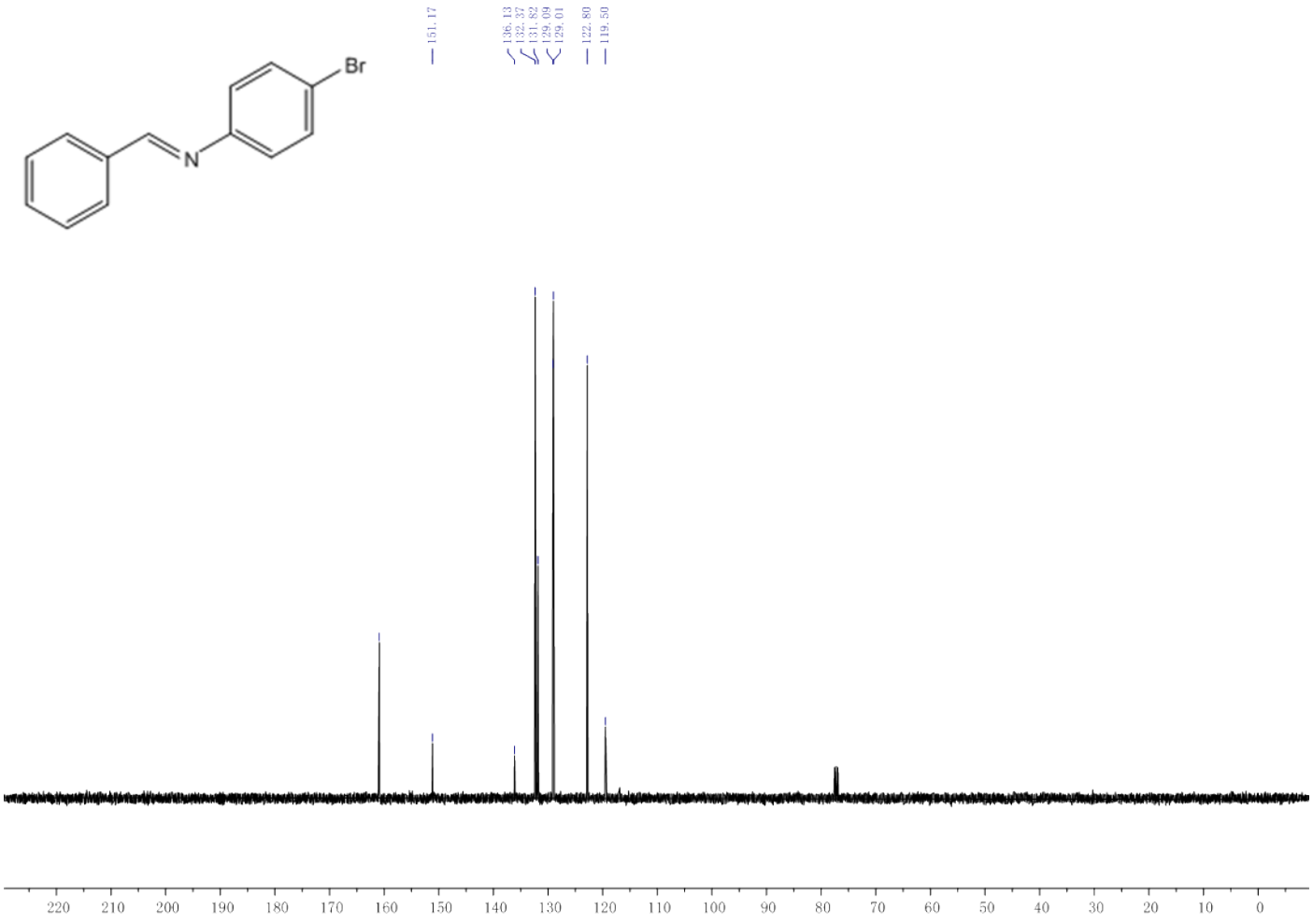
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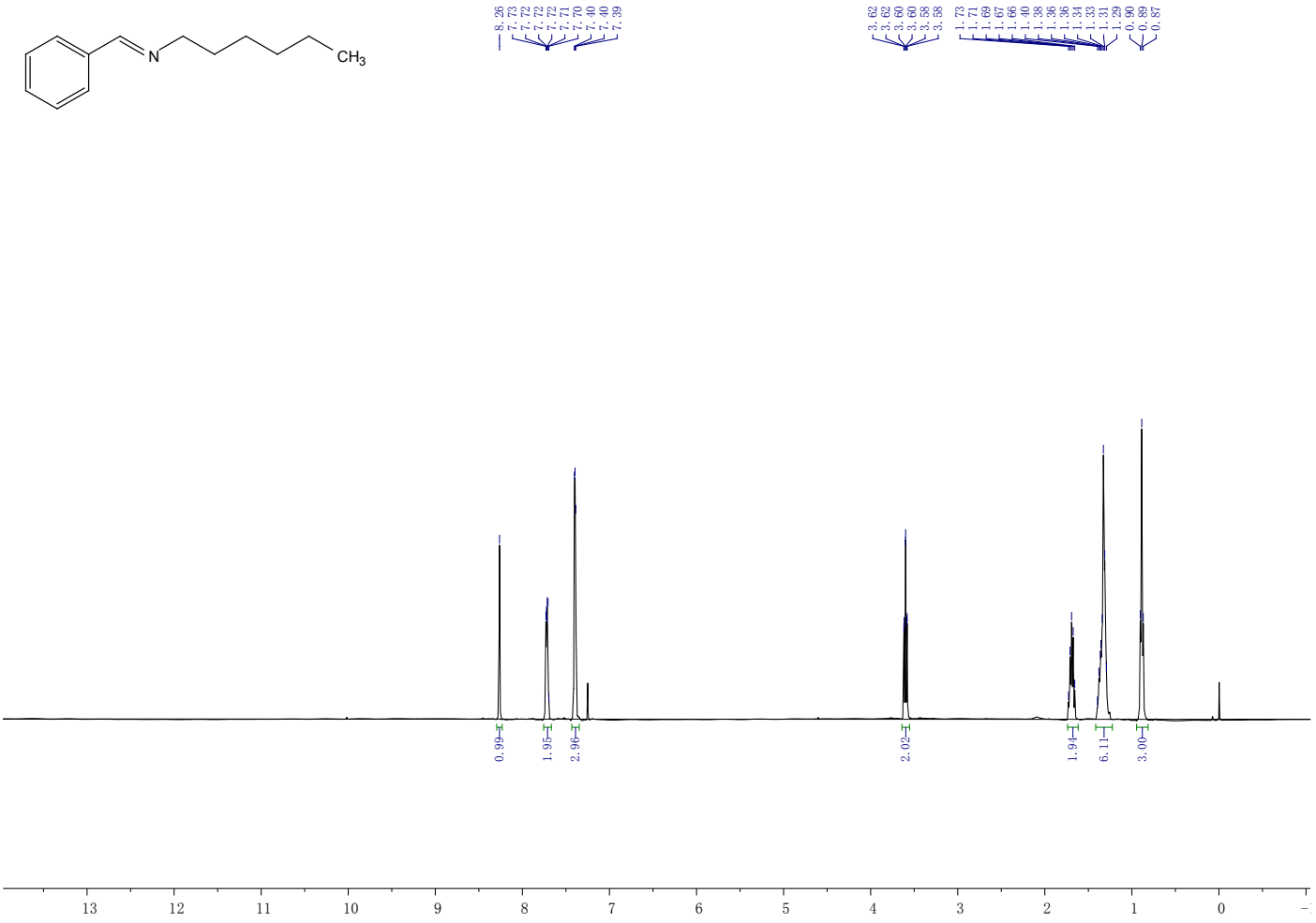
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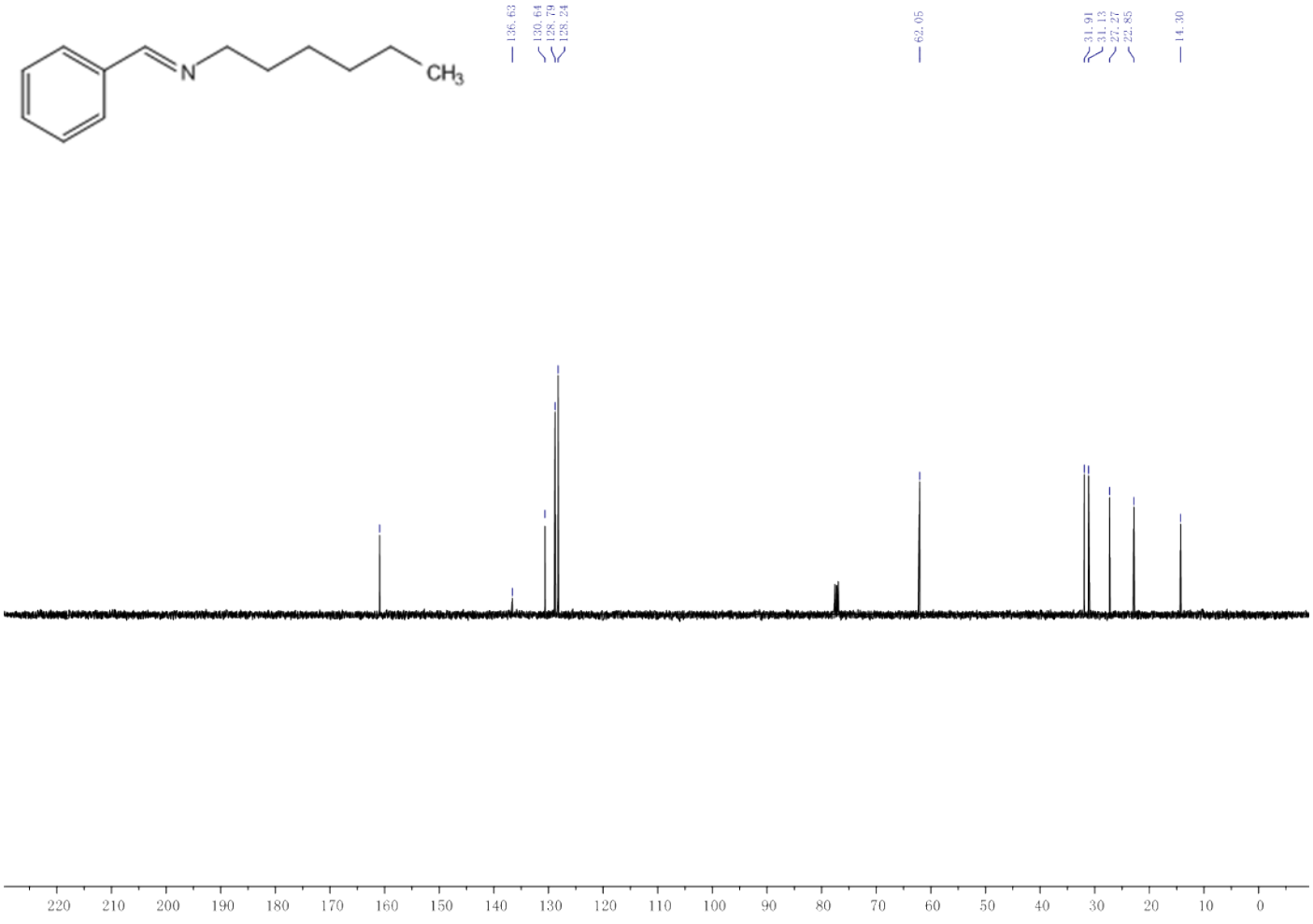
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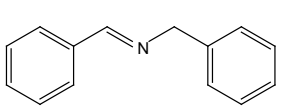


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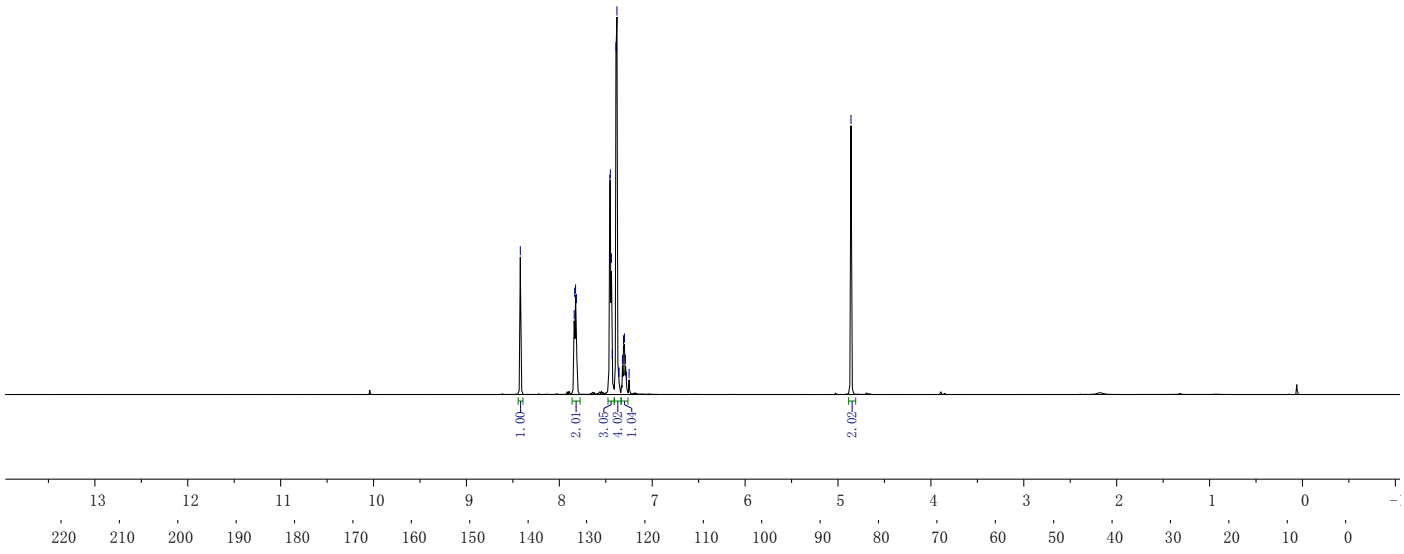
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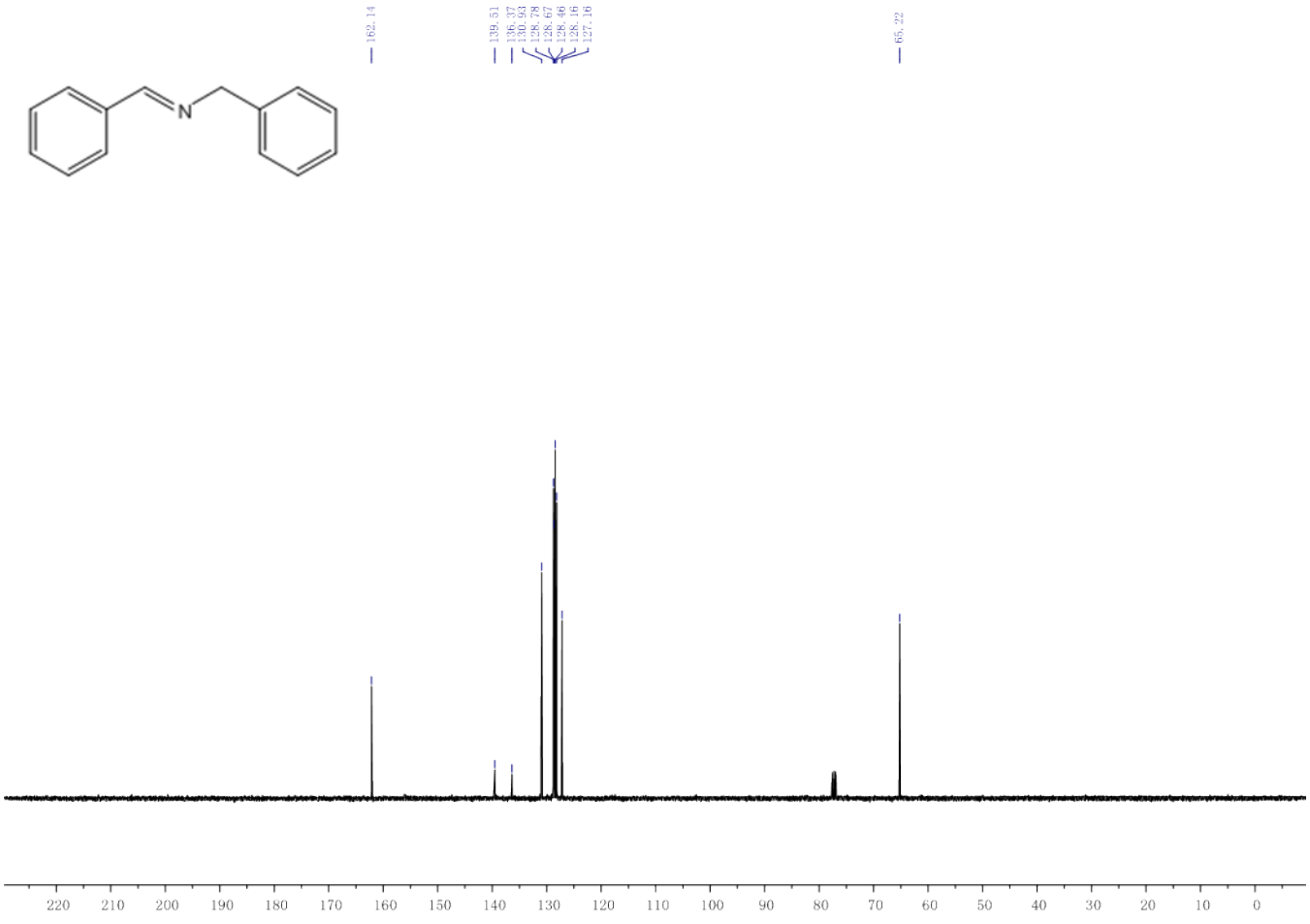
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