Supporting Information for

Organocatalytic Activation of Isocyanides: N-Heterocyclic Carbene-Catalyzed Enaminone Synthesis from Ketones

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1. General Information

Unless otherwise noted, all reactions were performed in a 4mL screw-capped reaction vial. All anhydrous solvents were purchased from commercial suppliers and degassed with dry argon before use. NMR spectra were recorded in CDCl$_3$ or DMSO-$d_6$, and the residue solvent signals were used as reference. Chemical shifts were reported in ppm, and coupling constants in Hz. Multiplicity is indicated by one or more of the following: s (singlet), d (doublet), t (triplet), q (quartet), quint (quintet), m (multiplet). Unless otherwise note, all reagents and solvents as well as all starting ketones, benzyl bromide, 2b’, and 2e were purchased from commercial suppliers and used as received without further purification. Previously reported isocyanides (2a, 2b, 2d, 2e, 2f, 2g, 2i, and 2j) were prepared from their corresponding amines by methods described in the literature, and their identity was confirmed by comparison with reported data. New isocyanide 2h was synthesized by a previously reported method, and its structure was confirmed by spectroscopic analysis. The free carbene (IMes) was synthesized from the corresponding salt (IMesHCl), according to a previous method. All enaminone products were purified by silica gel column chromatography (hexane/EtOAc with 3% NEt$_3$). We are grateful to the Organic Chemistry Research Center of Sogang University for HRMS-ESI analysis, the Korea Basic Science Institute (KBSI) for HRMS-EI analysis, and the Research Institute of Pharmaceutical Science (SNU) for single-crystal X-ray diffraction analysis.

2. Initial Experiment of (Z)-Enaminone Synthesis

Scheme S1. Reaction between 1a and 2a

IMesHCl (54.5mg, 0.16 mmol) and NaOtBu (23.1 mg, 0.24 mmol) were charged in a 25 mL Schlenk tube under argon atmosphere. The tube was then sealed with a rubber septum, and 1,4-dioxane (1.6 mL) was added. The mixture was stirred for 5 min. at room temperature, and a solution of 1a (93.5 µl, 0.8 mmol) and 2a (95.6 µl, 0.8 mmol) in 1,4-dioxane (1.6 mL) was added via syringe. The mixture was stirred for 24 h at 50 °C. After cooling to room
temperature, the volatiles were removed in vacuo and the remaining residue was purified by flash column chromatography (silica gel, hexane/ethyl acetate) to afford 3aa as a yellow solid (36.1 mg, 19 % yields).

3. General Procedure for the Synthesis of Enaminone (3)

IMesHCl (10.2mg, 0.03 mmol) and the base were charged in a 4 mL vial under argon atmosphere. The vial was then sealed with a Teflon-lined septum, and the ketone (0.2 mmol), aryl isocyanide (0.3 mmol), and DMA (2.4 mL) were added via syringe. The solution was stirred for the indicated time at 80 °C. After cooling to room temperature, the volatiles were removed in vacuo and the remaining residue was purified by flash column chromatography (silica gel, hexane/ethyl acetate in 3% triethylamine) to afford the corresponding enaminone.

4. Optimization Tables

Table S1 – Solvent

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<td>MeCN</td>
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<td>3</td>
<td>THF</td>
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<td>4</td>
<td>Isopropyl acetate</td>
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<td>5</td>
<td>DCM</td>
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<td>6</td>
<td>1,2-DCE</td>
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<td>7</td>
<td>Benzene</td>
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<td>Toluene</td>
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<td>13</td>
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<td>DMA</td>
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<td>15</td>
<td>DMSO</td>
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Table S2 – Base

![Chemical structure](image)

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Table S3 – NHC Salt

![Chemical structure](image)

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5. Experimental Procedures for the Control Experiments

Scheme S2. Reaction between 1a and 2a with IMes

IMes (9.1mg, 0.03 mmol) was charged in a 4 mL vial under argon atmosphere. The vial was then sealed with a Teflon-lined septum, and 1a (23.4 µl, 0.2 mmol), 2a (48 µl, 0.3 mmol), and DMA (2.4 mL) were added via syringe. The solution was stirred for 6 h at 80 °C. After cooling to room temperature, the volatiles were removed in vacuo and the remaining residue was purified by flash column chromatography (silica gel, hexane/ethyl acetate with 3% NEt3) to afford the corresponding enaminone 3aa in 76% yields.
Scheme S3. Reaction between 1a and 2b’

Additive (0.04 or 0.2 mmol) was charged in a 4 mL vial under argon atmosphere. The vial was then sealed with a Teflon-lined septum, and 1a (23.4 µl, 0.2 mmol), 2b’ (44.5 µl, 0.3 mmol), and DMA (2.4 mL) were added via syringe. The solution was stirred for 6 h at 80 °C. After cooling to room temperature, the volatiles were removed in vacuo and the remaining residue was purified by flash column chromatography (silica gel, hexane/ethyl acetate with 3% NEt₃) to afford the corresponding enaminone 3ab.

Scheme S4. Direct alkylation reaction with in-situ generated enolate anion

Additive (0.5 mmol) and THF (2 mL) were charged in a 10 mL Schlenk tube under argon atmosphere, and the tube was then sealed with rubber septum. 1a (58.3 µL, 0.5 mmol) was slowly added to the solution under argon flow at room temperature, and the solution was further stirred for 1.5 h at room temperature. Benzyl bromide (71.4 µL, 0.6 mmol) was added in the reaction mixture under argon flow, and the solution was further stirred for 3 h at room temperature. After the reaction was finished, the reaction mixture was diluted with ethyl acetate (10 mL), washed with aqueous sat. NaHCO₃ solution (10 mL) and brine (10 mL), dried with MgSO₄, and concentrated in vacuo. The crude mixture was analysed by ¹H-NMR using nitromethane as an internal standard.

Scheme S5. Reaction of 2a with in-situ generated enolate anion

Additive (1.0 equiv.) and THF (0.5 M) were added to the solution under argon flow. The solution was stirred for 1.5 h at room temperature. The reaction mixture was then diluted with ethyl acetate (10 mL), washed with aqueous sat. NaHCO₃ solution (10 mL) and brine (10 mL), dried with MgSO₄, and concentrated in vacuo. The crude mixture was analysed by ¹H-NMR using nitromethane as an internal standard.
KOTBu (56 mg, 0.5 mmol) and THF (1 mL) were charged in a 25 mL Schlenk tube under Ar atmosphere, and the tube was then sealed with rubber septum. 1a (58.3 µL, 0.5 mmol) was slowly added to the solution under argon flow at room temperature, and the solution was further stirred for 1.5 h at room temperature. After removing all volatiles via manifold vacuum (white solid 1a’ was observed), 2b (120 µL, 1.0 mmol), and DMA (6 mL) were added in the tube under argon flow. The solution was stirred for 6 h at 80 °C. After cooling to room temperature, the reaction mixture was diluted with ethyl acetate (20 ml), washed with aqueous sat. NaHCO₃ solution (20 mL x 2) and brine (20 mL), dried with MgSO₄, and concentrated in vacuo. The crude mixture was purified by flash column chromatography (silica gel, hexane/ethyl acetate with 3% NEt₃) to afford the corresponding enaminone 3aa in 55% yields.

6. Experimental Procedure for the Gram-Scale Reaction

Scheme S6. Gram-Scale Reaction of 1f with 2a

IMesHCl (511 mg, 1.5 mmol), K₂CO₃ (276 mg, 2.0 mmol), and DMA (80 mL) were charged in a 250 mL oven-dried round-bottom flask (RBF) under Ar atmosphere, and the flask was then sealed with rubber septum. 1f (1.38 mL, 10 mmol) and 2a (1.8 mL, 15 mmol) were added to the solution under argon flow, and the solution was further stirred for 24 h at 80 °C. After reaction was finished, the reaction mixture was cooled to room temperature and diluted with ethyl acetate (170 mL). The organic solution was washed with aqueous 5% LiCl solution (250 mL x 4) and aqueous sat. NaCl solution (250 mL), dried with MgSO₄ and concentrated in vacuo. The residual mixture was further purified by flash column chromatography (silica gel, hexane/ethyl acetate with 3% NEt₃) to afford the corresponding enaminone 3fa in 75% yield.
7. Crystallographic Data of 3sa

![Solid-state structure of 3sa at 50 % probability ellipsoids.](image)

Single crystals of 3sa were obtained by slow evaporation of sat. DMA solution of 3sa, and one of them was chosen for the analysis by X-ray diffractometer.

Table S5 – Crystal data and structure refinement for 3sa.

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8. Spectroscopic Data

2-fluoro-1-isocyanomethylbenzene (2h)

\[
\begin{align*}
\text{H} & \quad \text{NMR (499 MHz, CDCl}_3\) \delta \ 7.27 \ (t, \ J = 7.7 \text{ Hz}, \ 1\text{H}), \ 7.00 \ (d, \ J = 10.2 \text{ Hz}, \ 1\text{H}), \ 6.97 \ - \ 6.94 \ (m, \ 1\text{H}), \ 2.38 \ (s, \ 3\text{H}); \ 1^3\text{C} \text{ NMR (75 MHz, CDCl}_3\) \delta \ 169.38, \\
& \quad \ 157.12 \ (d, \ J = 255.9 \text{ Hz}), \ 142.27 \ (d, \ J = 7.3 \text{ Hz}), \ 127.44, \ 125.30 \ (d, \ J = 3.5 \text{ Hz}), \ 116.98 \ (d, \ J = 18.1 \text{ Hz}), \ 112.64 \ (d, \ J = 14.8 \text{ Hz}), \ 21.36 \ (d, \ J = 1.3 \text{ Hz}); \ 1^9\text{F} \text{ NMR (376 MHz, CDCl}_3\) \delta -119.26; \ \text{HRMS-ESI (m/z) [M]^{+} \text{ calcd for C}_8\text{H}_6\text{FN}, 135.0484; found: 135.0483.}
\end{align*}
\]

(Z)-1-phenyl-3-(p-tolylamino)prop-2-en-1-one (3aa)

\[
\begin{align*}
\text{H} & \quad \text{NMR (499MHz, CDCl}_3\) \delta \ 12.15 \ (d, \ J = 11.7 \text{ Hz}, \ 1\text{H}), \ 7.94 \ (d, \ J = 7.8 \\
& \quad \text{Hz, 2 H), 7.44 \ - \ 7.52 \ (m, \ 4\text{H}), \ 7.15 \ (d, \ J = 8.3 \text{ Hz}, \ 2\text{H}), \ 7.02 \ (d, \ J = 8.3 \\
& \quad \text{Hz, 2 H), 6.00 \ (d, \ J = 7.8 \text{ Hz}, \ 1\text{H}), \ 2.33 \ (s, \ 3\text{H}). Identity confirmed by \\
& \quad \text{comparing with reported literature.}^3
\end{align*}
\]

(Z)-1-(4-methoxyphenyl)-3-(p-tolylamino)prop-2-en-1-one (3ba)

\[
\begin{align*}
\text{H} & \quad \text{NMR (400 MHz, CDCl}_3\) \delta \ 12.08 \ (d, \ J = 11.8 \text{ Hz}, \ 1\text{H}), \ 7.92 \ (d, \ J \\
& \quad = 8.8 \text{ Hz, 2H), 7.45 \ (dd, \ J = 11.8 \text{ Hz}, \ 7.9 \text{ Hz, 1H}), \ 7.14 \ (d, \ J = 8.2 \text{ Hz,} \\
& \quad \text{2H), 6.99 \ (d, \ J = 8.2 \text{ Hz, 2H), 6.94 \ (d, \ J = 8.8 \text{ Hz, 2H), 5.95 \ (d, \ J =} \\
& \quad 7.9 \text{ Hz, 1H), 3.86 \ (s, \ 3\text{H}), 2.32 \ (s, \ 3\text{H}); ^3\text{C} \text{ NMR (101 MHz, CDCl}_3\) \delta -189.92, \\
& \quad 162.50, 144.79, 138.10, 133.24, 132.18, 130.34, 129.38, 116.33, 113.74, 93.06, \\
& \quad 55.49, 20.86; \ \text{HRMS-ESI (m/z) [M+Na]^{+} \text{ calcd for C}_{17}\text{H}_{17}\text{NNaO}_2, 290.1151; found: 290.1150.}
\end{align*}
\]

(Z)-1-(4-chlorophenyl)-3-(p-tolylamino)prop-2-en-1-one (3ca)

\[
\begin{align*}
\text{H} & \quad \text{NMR (400 MHz, CDCl}_3\) \delta \ 12.14 \ (d, \ J = 12.3 \text{ Hz 1H), 7.87 \ (d, J =} \\
& \quad 6.7 \text{ Hz, 2H), 7.51 \ (dd, J = 11.4 \text{ Hz, 7.5 Hz, 1H), 7.42 \ (d, J = 6.7 \text{ Hz,}} \\
& \quad \text{1H), 7.16 \ (d, J = 7.2 \text{ Hz, 2H), 7.02 \ (d, J = 7.2 \text{ Hz, 2H), 5.95 \ (d, J = 7.5} \\
& \quad \text{Hz, 1H), 2.33 \ (s, \ 3\text{H}); ^3\text{C} \text{ NMR (101 MHz, CDCl}_3\) \delta 189.38, 145.84,}
\end{align*}
\]

S9
145.82, 137.79, 137.76, 133.85, 130.42, 128.78, 116.62, 93.00, 20.93; HRMS-ESI (m/z) [M+Na]⁺ calcd for C₁₆H₁₄ClNNO₂, 294.0656; found: 294.0658.

(Z)-1-(4-iodophenyl)-3-(p-tolylamino)prop-2-en-1-one (3da)

1H NMR (400 MHz, CDCl₃) δ 12.15 (d, J = 12.0 Hz, 1H), 7.80 (d, J = 8.3 Hz, 2H), 7.64 (d, J = 8.4 Hz, 2H), 7.50 (dd, J = 12.5 Hz, 7.7 Hz, 1H), 7.15 (d, J = 8.2 Hz, 2H), 7.01 (d, J = 8.3 Hz, 2H), 5.93 (d, J = 7.7 Hz, 1H), 2.33 (s, 3H); ¹³C NMR (75 MHz, CDCl₃) δ 189.70, 145.88, 137.76, 133.88, 130.43, 128.97, 116.64, 98.82, 92.92, 20.92.; HRMS-ESI (m/z) [M+H]⁺ calcd for C₁₆H₁₅INO₂, 364.0913; found: 364.0913.

(Z)-4-(3-(p-tolylamino)acryloyl)benzonitrile (3ea)

1H NMR (400 MHz, CDCl₃) δ 12.24 (d, J = 12.2 Hz, 1H), 7.99 (d, J = 8.2 Hz, 2H), 7.73 (d, J = 8.4 Hz, 2H), 7.56 (dd, J = 12.6 Hz, 7.7 Hz, 1H), 7.17 (d, J = 8.2 Hz, 2H), 7.03 (d, J = 8.3 Hz, 2H), 5.95 (d, J = 7.6 Hz, 1H), 2.33 (s, 3H); ¹³C NMR (101 MHz, CDCl₃) δ 188.35, 146.76, 143.03, 137.39, 134.42, 132.41, 130.48, 127.82, 118.58, 116.87, 114.64, 93.10, 20.94; HRMS-ESI (m/z) [M+Na]⁺ calcd for C₁₇H₁₄N₂NaO, 285.0998; found: 285.0997.

(Z)-1-(2-methoxyphenyl)-3-(p-tolylamino)prop-2-en-1-one (3fa)

1H NMR (499 MHz, CDCl₃) δ 12.05 (d, J = 12.6 Hz, 1H), 7.71 (dd, J = 7.6 Hz, 1.8 Hz, 1H), 7.44 – 7.39 (m, 2H), 7.15 (d, J = 8.2 Hz, 2H), 7.05 – 6.96 (m, 4H), 6.01 (d, J = 7.8 Hz, 1H), 3.92 (s, 3H), 2.33 (s, 3H); ¹³C NMR (101 MHz, CDCl₃) δ 191.73, 157.67, 144.39, 138.15, 133.30, 131.96, 130.33, 130.19, 120.74, 116.47, 111.67, 98.48, 55.80, 20.87; HRMS-ESI (m/z) [M+Na]⁺ calcd for C₁₇H₁₅NO₂, 290.1151; found: 290.1152.

(Z)-1-(2-bromophenyl)-3-(p-tolylamino)prop-2-en-1-one (3ga)
1H NMR (400 MHz, CDCl₃) δ 11.92 (d, J = 11.6 Hz, 1H), 7.60 (d, J = 8.0 Hz, 1H), 7.46 (dd, J = 12.5 Hz, 7.7 Hz, 2H), 7.35 (t, J = 7.5 Hz, 1H), 7.23 (t, J = 8.2 Hz, 2H), 7.02 (d, J = 8.3 Hz, 2H), 5.63 (d, J = 7.6 Hz, 1H), 2.33 (s, 3H); 13C NMR (101 MHz, CDCl₃) δ 193.01, 145.51, 142.63, 137.69, 133.98, 133.63, 130.74, 129.25, 127.38, 119.50, 116.76, 97.19, 20.92.; HRMS-ESI (m/z) [M+Na]^+ calcd for C₁₆H₁₄BrNaN₃O, 338.0151; found: 338.0152.

(Z)-1-(benzo[d][1,3]dioxol-5-yl)-3-(p-tolylamino)prop-2-en-1-one (3ha)

1H NMR (400 MHz, CDCl₃) δ 12.04 (d, J = 11.7Hz, 1H), 7.52 (d, J = 8.2Hz, 1H), 7.49 – 7.41 (m, 2H), 7.14 (d, J = 8.2 Hz, 2H), 6.99 (d, J = 8.3 Hz, 2H), 6.85 (d, J = 8.1 Hz, 1H), 6.03 (s, 2H), 5.90 (d, J = 7.8 Hz, 1H), 2.32 (s, 3H); 13C NMR (101 MHz, CDCl₃) δ 189.41, 150.60, 148.11, 145.00, 138.02, 134.14, 133.40, 130.37, 122.79, 116.42, 108.01, 107.63, 101.71, 93.04, 20.88; HRMS-ESI (m/z) [M+Na]^+ calcd for C₁₇H₁₅NN₃O₃, 304.0944; found: 304.0943.

(Z)-1-(furan-2-yl)-3-(p-tolylamino)prop-2-en-1-one (3ia)

1H NMR (400 MHz, CDCl₃) δ 11.91 (d, J = 11.6 Hz, 1H), 7.53 (s, 1H), 7.45 (dd, J = 12.5, 7.8 Hz, 1H), 7.14 (d, J = 8.1 Hz, 2H), 7.09 (d, J = 3.4 Hz, 1H), 6.98 (d, J = 8.3 Hz, 2H), 6.51 (dd, J = 3.3, 1.5 Hz, 1H), 5.90 (d, J = 7.8 Hz, 1H), 2.31 (s, 3H); 13C NMR (101 MHz, CDCl₃) δ 179.96, 153.96, 145.30, 145.02, 137.84, 133.56, 130.36, 116.39, 113.97, 112.22, 112.20, 93.11, 20.88.; HRMS-ESI (m/z) [M+Na]^+ calcd for C₁₄H₁₃NN₃O₂, 250.0838; found: 250.0836.

(Z)-1-(thiophen-2-yl)-3-(p-tolylamino)prop-2-en-1-one (3ja)

1H NMR (400 MHz, CDCl₃) δ 11.87 (d, J = 11.6 Hz, 1H), 7.63 (d, J = 3.7 Hz, 1H), 7.54 (d, J = 4.9 Hz, 1H), 7.44 (dd, J = 12.5, 7.7 Hz, 1H), 7.14 (d, J = 8.3 Hz, 2H), 7.11 (t, J = 3.9 Hz, 1H), 6.98 (d, J = 8.3 Hz, 2H), 5.86 (d, J = 7.7 Hz, 1H), 2.32 (s, 3H); 13C NMR (101 MHz, CDCl₃) δ 183.72, 146.44, 145.01, 137.88, 133.57, 131.46, 130.40, 128.90, 128.12, 116.37, 93.37, 20.89; HRMS-ESI (m/z) [M+Na]^+ calcd for C₁₄H₁₃NN₃O₃, 266.0610; found: 266.0610.
(Z)-1-(1-methyl-1H-pyrrol-2-yl)-3-(p-tolylamino)prop-2-en-1-one (3ka)

$^1$H NMR (400 MHz, CDCl$_3$) $\delta$ 11.53 (d, $J = 11.9$ Hz, 1H), 7.29 (dd, $J = 12.3$, 8.1 Hz, 1H), 7.12 (d, $J = 8.2$ Hz, 2H), 6.96 (d, $J = 8.3$ Hz, 2H), 6.86 - 6.82 (m, 1H), 6.76 (s, 1H), 6.14 - 6.10 (m, 1H), 5.79 (d, $J = 8.1$ Hz, 1H), 4.02 (s, 3H), 2.31 (s, 3H); $^{13}$C NMR (101 MHz, CDCl$_3$) $\delta$ 183.62, 142.89, 138.50, 132.54, 132.11, 130.27, 129.76, 116.04, 115.95, 107.76, 95.08, 37.68, 20.83.; HRMS-ESI (m/z) [M+Na]$^+$ calcd for C$_{15}$H$_{16}$N$_2$NaO, 263.1155; found: 263.1156.

(Z)-1-(pyridin-2-yl)-3-(p-tolylamino)prop-2-en-1-one (3la)

$^1$H NMR (400 MHz, CDCl$_3$) $\delta$ 12.16 (d, $J = 12.0$ Hz, 1H), 8.66 (d, $J = 4.1$ Hz, 1H), 8.13 (d, $J = 7.8$ Hz, 1H), 7.83 (t, $J = 7.7$ Hz, 1H), 7.60 (dd, $J = 12.3$, 7.7 Hz, 1H), 7.39 (dd, $J = 7.7$, 4.8 Hz, 1H), 7.15 (d, $J = 7.9$ Hz, 2H), 7.03 (d, $J = 7.7$ Hz, 2H), 6.71 (d, $J = 7.7$ Hz, 1H), 2.32 (s, 3H); $^{13}$C NMR (101 MHz, CDCl$_3$) $\delta$ 189.35, 155.40, 148.82, 146.35, 137.81, 137.04, 133.84, 130.40, 125.83, 121.76, 116.65, 92.93, 20.90.; HRMS-ESI (m/z) [M+Na]$^+$ calcd for C$_{15}$H$_{14}$N$_2$NaO, 261.0998; found: 261.0998.

(Z)-1-(naphthalen-2-yl)-3-(p-tolylamino)prop-2-en-1-one (3ma)

$^1$H NMR (400 MHz, CDCl$_3$) $\delta$ 12.23 (d, $J = 11.9$ Hz, 1H), 8.45 (s, 1H), 8.04 (d, $J = 8.6$ Hz, 1H), 7.96 (d, $J = 8.1$ Hz, 1H), 7.90 (d, $J = 8.7$ Hz, 1H), 7.87 (d, $J = 7.7$ Hz, 1H), 7.59 - 7.49 (m, 3H), 7.16 (d, $J = 8.1$ Hz, 2H), 7.04 (d, $J = 8.3$ Hz, 2H), 6.16 (d, $J = 7.8$ Hz, 1H), 2.33 (s, 3H); $^{13}$C NMR (101 MHz, CDCl$_3$) $\delta$ 190.74, 145.42, 137.95, 136.75, 135.05, 133.62, 132.95, 130.41, 129.47, 128.31, 128.12, 127.83, 127.72, 126.57, 124.10, 116.55, 93.61, 20.93; HRMS-ESI (m/z) [M+Na]$^+$ calcd for C$_{20}$H$_{17}$NNaO, 310.1202; found: 310.1204.

(1E,4Z)-1-phenyl-5-(p-tolylamino)penta-1,4-dien-3-one (3na)
\[ \text{1H NMR (499 MHz, CDCl}_3\) \delta 12.18 (d, } J = 12.3 \text{ Hz, 1H), 7.60 - 7.54 (m, 3H), 7.44 - 7.33 (m, } 4\text{H), 7.14 (d, } J = 8.0 \text{ Hz, 2H), 6.99 (d, } J = 8.4 \text{ Hz, 2H), 6.78 (d, } J = 15.9 \text{ Hz, 2H), 5.50 (d, } J = 7.5 \text{ Hz, 1H), 2.32 (s, 3H);} \]
\[ \text{13C NMR (75 MHz, CDCl}_3\) \delta 189.07, 145.09, 139.47, 137.92, 135.64, 133.62, 130.40, 129.74, 128.94, 128.17, 127.86, 116.47, 97.83, 20.91; } \]

(Z)-1-cyclopropyl-3-(p-tolylamino)prop-2-en-1-one (3oa)

\[ \text{1H NMR (499 MHz, CDCl}_3\) \delta 11.56 (d, } J = 10.9 \text{ Hz, 1H), 7.18 (dd, } J = 12.4, 7.7 \text{ Hz, 1H), 7.10 (d, } J = 8.3 \text{ Hz, 2H), 6.90 (d, } J = 8.4 \text{ Hz, 2H), 5.41 (d, } J = 7.7 \text{ Hz, 1H), 2.29 (s, 3H), 1.83 - 1.78 (m, 1H), 1.05 - 1.02 (m, 2H), 0.85 - 0.81 (m, 2H);} \]
\[ \text{13C NMR (75 MHz, CDCl}_3\) \delta 200.48, 142.57, 138.06, 132.86, 130.16, 115.94, 96.65, 20.70, 20.53, 9.89; } \]
\[ \text{HRMS-ESI (m/z) [M+Na] } \text{+ calcd for } C_{13}H_{15}N\text{NaO, 224.1046; found: 224.1043.} \]

(Z)-2-((p-tolylamino)methylene)-2,3-dihydro-1H-inden-1-one (3pa)

\[ \text{1H NMR (499 MHz, CDCl}_3\) \delta 11.35 (d, } J = 11.3 \text{ Hz, 1H), 7.84 (d, } J = 7.6 \text{ Hz, 1H), 7.55 - 7.46 (m, 3H), 7.14 (d, } J = 8.0 \text{ Hz, 2H), 7.00 (d, } J = 8.5 \text{ Hz, 2H), 3.66 (s, 2H), 2.32 (s, 3H);} \]
\[ \text{13C NMR (75 MHz, CDCl}_3\) \delta 193.32, 148.75, 141.02, 139.28, 138.16, 132.90, 132.16, 130.39, 127.21, 125.81, 122.94, 115.82, 107.75, 30.89, 20.87; } \]
\[ \text{HRMS-ESI (m/z) [M+Na] } \text{+ calcd for } C_{17}H_{15}N\text{NaO, 272.1046; found: 272.1048.} \]

(Z)-2-((p-tolylamino)methylene)-3,4-dihydronaphthalen-1(2H)-one (3qa)

\[ \text{1H NMR (400 MHz, CDCl}_3\) \delta 11.97 (d, } J = 11.4 \text{ Hz, 1H), 8.03 (d, } J = 7.6 \text{ Hz, 1H), 7.44 - 7.31 (m, 3H), 7.22 (d, } J = 7.4 \text{ Hz, 1H), 7.13 (d, } J = 8.1 \text{ Hz, 2H), 6.99 (d, } J = 8.3 \text{ Hz, 2H), 2.92 (t, } J = 6.5 \text{ Hz, 2H), 2.68 (t, } J = 6.5 \text{ Hz, 2H), 2.31 (s, 3H);} \]
\[ \text{13C NMR (101 MHz, CDCl}_3\) \delta 187.55, 142.37, 142.08, 138.33, 135.32, 132.78, 131.90, 130.35, 127.98, 126.94, 126.67, 116.10, 104.83, 29.99, 27.86, 20.89; } \]
\[ \text{HRMS-ESI (m/z) [M+Na] } \text{+ calcd for } C_{18}H_{17}N\text{NaO, 286.1202; found: 286.1201.} \]
(Z)-6-((p-tolylamino)methylene)-6,7,8,9-tetrahydro-5H-benzo[7]annulen-5-one (3ra)

\[
\text{1}H \text{ NMR (499 MHz, CDCl}_3) \delta 12.01 (d, J = 11.6 \text{ Hz}, 1H), 7.61 (dd, J = 7.3, 1.7 \text{ Hz}, 1H), 7.39 \text{ to } 7.31 (m, 2H), 7.28 (d, J = 12.2 \text{ Hz}, 1H), 7.16 (d, J = 8.2 \text{ Hz}, 1H), 7.14 (d, J = 8.3 \text{ Hz}, 2H), 7.00 (d, J = 8.4 \text{ Hz}, 2H), 2.74 (t, J = 7.0 \text{ Hz}, 2H), 2.32 (s, 3H), 2.18 (t, J = 6.8 \text{ Hz}, 2H), 1.95 (quint, J = 6.9 \text{ Hz}, 2H); \text{13}^C \text{ NMR (75 MHz, CDCl}_3) \delta 197.10, 143.03, 141.71, 138.93, 138.25, 132.84, 130.69, 130.31, 128.58, 127.49, 126.75, 116.15, 107.64, 31.01, 30.64, 28.11, 20.83; HRMS-ESI (m/z) [M+Na]^+ \text{ calcd for C}_{19}H_{19}NNaO, 300.1359; found: 300.1360.
\]

(E)-2-methyl-1-phenyl-3-((p-tolylamino)prop-2-en-1-one (3sa)

\[
\text{1}H \text{ NMR (499 MHz, DMSO-d}_6) \delta 8.96 (d, J = 13.1 \text{ Hz}, 1H), 7.52 \text{ to } 7.42 (m, 5H), 7.40 (d, J = 13.1 \text{ Hz}, 1H), 7.06 (d, J = 8.3 \text{ Hz}, 2H), 6.85 (d, J = 8.4 \text{ Hz}, 2H), 2.20 (s, 3H), 1.93 (s, 3H); \text{13}^C \text{ NMR (75 MHz, DMSO-d}_6) \delta 194.83, 145.59, 141.37, 139.72, 131.58, 130.37, 130.10, 128.52, 116.26, 109.97, 20.67, 10.34; HRMS-ESI (m/z) [M+Na]^+ \text{ calcd for C}_{17}H_{17}NNaO, 274.1202; found: 274.1200.
\]

(E)-1-phenyl-2-((p-tolylamino)methylene)butan-1-one (3ta)

\[
\text{1}H \text{ NMR (499 MHz, DMSO-d}_6) \delta 9.01 (d, J = 13.2 \text{ Hz}, 1H), 7.50 \text{ to } 7.40 (m, 5H), 7.32 (d, J = 13.2 \text{ Hz}, 1H), 7.05 (d, J = 8.3 \text{ Hz}, 2H), 6.84 (d, J = 8.5 \text{ Hz}, 2H), 2.48 (t, J = 7.3 \text{ Hz}, 2H), 2.19 (s, 3H), 1.03 (t, J = 7.4 \text{ Hz}, 3H); \text{13}^C \text{ NMR (101 MHz, DMSO-d}_6) \delta 194.21, 144.40, 141.03, 139.29, 131.04, 129.88, 129.62, 128.04, 116.05, 115.80, 20.22, 16.75, 13.14; HRMS-ESI (m/z) [M+Na]^+ \text{ calcd for C}_{18}H_{19}NNaO, 288.1359; found: 288.1358.
\]

(E)-1-phenyl-2-((p-tolylamino)methylene)pentan-1-one (3ua)

\[
\text{1}H \text{ NMR (499 MHz, DMSO-d}_6) \delta 8.96 (d, J = 13.3 \text{ Hz}, 1H), 7.50 \text{ to } 7.42 (m, 5H), 7.05 (d, J = 8.4 \text{ Hz}, 2H), 6.83 (d, J = 8.5 \text{ Hz}, 2H), 2.46 (t, J = 7.3 \text{ Hz}, 2H), 2.19 (s, 3H), 1.49 \text{ to } 1.41 (m, 2H), 0.95 (t, J = 7.3 \text{ Hz}, 3H);
\]

S14
$^{13}$C NMR (75 MHz, DMSO-d$_6$) δ 194.52, 144.98, 141.10, 139.32, 131.07, 129.89, 129.62, 128.04, 115.87, 114.49, 25.16, 21.18, 20.19, 13.88; HRMS-ESI (m/z) [M+Na]$^+$ calcd for C$_{19}$H$_{21}$NNaO, 302.1515; found: 302.1517.

(E)-1-phenyl-2-((p-tolylamino)methylene)hexan-1-one (3va)

$^1$H NMR (499 MHz, DMSO-d$_6$) δ 8.94 (d, $J$ = 13.2 Hz, 1H), 7.51 – 7.40 (m, 5H), 7.33 (d, $J$ = 13.2 Hz, 1H), 7.04 (d, $J$ = 8.2 Hz, 2H), 6.83 (d, $J$ = 8.4 Hz, 2H), 2.49 (t, $J$ = 7.3 Hz, 2H), 2.19 (s, 3H), 1.47 – 1.31 (m, 4H), 0.91 (t, $J$ = 6.9 Hz, 3H); $^{13}$C NMR (75 MHz, DMSO-d$_6$) δ 194.46, 144.79, 141.10, 139.33, 131.02, 129.86, 129.60, 128.04, 128.01, 115.85, 114.75, 30.39, 23.12, 22.23, 20.17, 14.14; HRMS-ESI (m/z) [M+Na]$^+$ calcd for C$_{20}$H$_{23}$NNaO, 316.1672; found: 316.1670.

(E)-1-phenyl-2-((p-tolylamino)methylene)heptan-1-one (3wa)

$^1$H NMR (499 MHz, DMSO-d$_6$) δ 8.94 (d, $J$ = 13.2 Hz, 1H), 7.50 – 7.39 (m, 5H), 7.32 (d, $J$ = 13.2 Hz, 1H), 7.05 (d, $J$ = 8.3 Hz, 2H), 6.83 (d, $J$ = 8.4 Hz, 2H), 2.47 (t, $J$ = 7.6 Hz, 2H), 2.19 (s, 3H), 1.46 – 1.38 (m, 2H), 1.38 – 1.28 (m, 4H), 0.88 (t, $J$ = 7.0 Hz, 3H); $^{13}$C NMR (101 MHz, DMSO-d$_6$) δ 194.45, 144.76, 141.09, 139.32, 131.02, 129.87, 129.61, 128.02, 115.84, 114.77, 31.34, 27.77, 23.30, 22.27, 20.17, 14.06; HRMS-ESI (m/z) [M+Na]$^+$ calcd for C$_{21}$H$_{25}$NNaO, 330.1828; found: 330.1827.

3-methyl-1-phenyl-2-((p-tolylamino)methylene)butan-1-one (3xa)

$^1$H NMR (499 MHz, DMSO-d$_6$) (E)-form (major): δ 8.91 (d, $J$ = 13.2 Hz, 1H), 7.51 – 7.41 (m, 5H), 7.16 (d, $J$ = 13.2 Hz, 1H), 7.03 (d, $J$ = 8.2 Hz, 2H), 6.79 (d, $J$ = 8.4 Hz, 2H), 3.19 – 3.05 (m, 1H), 2.18 (s, 3H), 1.28 (d, $J$ = 6.9 Hz, 6H), (Z)-form (minor): δ 12.14 (d, $J$ = 12.1 Hz, 1H), 7.68 (d, $J$ = 12.1 Hz, 1H), 7.51 – 7.41 (m, 5H), 7.22 (d, $J$ = 8.5 Hz, 2H), 7.15 (d, $J$ = 8.3 Hz, 2H), 2.71 – 2.64 (m, 1H), 2.27 (s, 3H), 1.08 (d, $J$ = 6.9 Hz, 6H); $^{13}$C NMR (101 MHz, DMSO-d$_6$) δ 195.05, 143.97,
141.66, 139.44, 130.84, 129.85, 128.21, 127.98, 118.84, 115.65, 25.10, 20.31, 20.20; HRMS-ESI (m/z) [M+Na]+ calcd for C_{19}H_{21}NNaO, 302.1515; found: 302.1515.

\((2Z,2’Z)-1,1’-(1,3\text{-phenylene})\text{bis}(3-(p\text{-tolylamino})\text{prop-2-en-1-one})\ (3ya)\)

\(^1H\) NMR (499 MHz, CDCl₃) δ 12.18 (d, J = 12.4 Hz, 2H), 8.48 (t, J = 1.6 Hz, 1H), 8.07 (dd, J = 7.7, 1.8 Hz, 2H), 7.54 (t, J = 7.6 Hz, 1H), 7.53 (dd, J = 12.4, 7.7 Hz, 2H), 7.16 (d, J = 8.1 Hz, 4H), 7.03 (d, J = 8.4 Hz, 2H), 6.07 (d, J = 7.8 Hz, 2H), 2.33 (s, 6H); \(^1^3C\) NMR (75 MHz, CDCl₃) δ 190.20, 145.78, 139.60, 137.86, 133.78, 130.42, 130.27, 128.80, 126.25, 116.63, 93.38, 20.91.; HRMS-ESI (m/z) [M+Na]+ calcd for C_{26}H_{24}N_{2}NaO_{2}, 419.1730; found: 419.1732.

\((2Z,2’Z)-1,1’-(pyridine-2,6-diyl)\text{bis}(3-(p\text{-tolylamino})\text{prop-2-en-1-one})\ (3za)\)

\(^1H\) NMR (499 MHz, CDCl₃) δ 12.18 (d, J = 12.4 Hz, 2H), 8.24 (d, J = 7.7 Hz, 2H), 7.98 (t, J = 7.7 Hz, 1H), 7.63 (dd, J = 12.4, 7.7 Hz, 2H), 7.17 (d, J = 7.9 Hz, 4H), 7.05 (d, J = 8.0 Hz, 4H), 6.87 (d, J = 7.7 Hz, 2H), 2.34 (s, 6H); \(^1^3C\) NMR (75 MHz, CDCl₃) δ 189.01, 154.27, 145.36, 137.97, 137.85, 133.93, 130.45, 123.60, 116.70, 93.00, 20.93; HRMS-ESI (m/z) [M+Na]+ calcd for C_{25}H_{23}N_{3}NaO_{2}, 420.1682; found: 420.1684.

Diethyl 2-((p-tolylamino)methylene)malonate (3Aa)

\(^1H\) NMR (499 MHz, CDCl₃) δ 10.97 (d, J = 13.7 Hz, 1H), 8.50 (d, J = 13.8 Hz, 1H), 7.17 (d, J = 8.1 Hz, 2H), 7.03 (d, J = 8.4 Hz, 2H), 4.30 (q, J = 7.1 Hz, 2H), 4.24 (q, J = 7.1 Hz, 2H), 2.33 (s, 3H), 1.38 (t, J = 7.1 Hz, 3H), 1.32 (t, J = 7.1 Hz, 3H). Identity confirmed by comparing with reported literature.\(^4\)

\((Z)-2\text{-phenyl-3-(p-tolylamino)acrylonitrile}\ (3Ba)\)
1H NMR (499 MHz, DMSO) δ 9.61 (d, J = 12.9 Hz, 1H), 8.05 (d, J = 12.9 Hz, 1H), 7.50 (dd, J = 8.4, 1.1 Hz, 2H), 7.34 (t, J = 7.9 Hz, 2H), 7.28 (d, J = 8.5 Hz, 2H), 7.16 (t, J = 7.3 Hz, 1H), 7.11 (d, J = 8.2 Hz, 2H), 2.25 (s, 3H). Identity confirmed by comparing with reported literature.5

(Z)-1-phenyl-3-(phenylamino)prop-2-en-1-one (3ab)

1H NMR (400 MHz, CDCl₃) δ 12.15 (d, J = 10.8 Hz, 1H), 7.95 (d, J = 7.0 Hz, 2H), 7.56 – 7.44 (m, 4H), 7.35 (t, J = 7.8 Hz, 2H), 7.11 (d, J = 7.9 Hz, 2H), 7.08 (t, J = 7.4 Hz, 1H), 6.04 (d, J = 7.8 Hz, 1H). Identity confirmed by comparing with reported literature.5

(Z)-3-((4-methoxyphenyl)amino)-1-phenylprop-2-en-1-one (3ac)

1H NMR (499 MHz, CDCl₃) δ 12.20 (d, J = 11.9 Hz, 1H), 7.95 - 7.91 (m, 2H), 7.52 – 7.39 (m, 4H), 7.05 (d, J = 8.9 Hz, 2H), 6.89 (d, J = 8.9 Hz, 2H), 5.97 (d, J = 7.7 Hz, 1H), 3.79 (s, 3H). Identity confirmed by comparing with reported literature.7

(Z)-3-((4-chlorophenyl)amino)-1-phenylprop-2-en-1-one (3ad)

1H NMR (499 MHz, CDCl₃) δ 12.14 (d, J = 11.6 Hz, 1H), 7.93 (d, J = 7.0 Hz, 2H), 7.54 – 7.42 (m, 4H), 7.31 (d, J = 8.9 Hz, 2H), 7.03 (d, J = 8.9 Hz, 2H), 6.05 (d, J = 7.9 Hz, 1H). Identity confirmed by comparing with reported literature.7

(Z)-3-((4-bromophenyl)amino)-1-phenylprop-2-en-1-one (3ae)

1H NMR (400 MHz, CDCl₃) δ 12.13 (d, J = 11.5 Hz, 1H), 7.93 (d, J = 7.2 Hz, 2H), 7.55 – 7.40 (m, 6H), 6.98 (d, J = 8.7 Hz, 2H), 6.06 (d, J = 7.9 Hz, 1H); 13C NMR (101 MHz, CDCl₃) δ 191.42, 144.47, 139.54, 139.11,
132.84, 131.92, 128.63, 127.48, 117.94, 116.25, 94.46; HRMS-ESI (m/z) [M+Na]^+ calcd for C_{15}H_{12}BrNNaO, 323.9994; found: 323.9991.

(Z)-3-((2-methoxyphenyl)amino)-1-phenylprop-2-en-1-one (3af)

1H NMR (400 MHz, CDCl$_3$) δ 12.22 (d, $J = 11.4$ Hz, 1H), 7.97 (d, $J = 7.1$ Hz, 2H), 7.54 (dd, $J = 12.7, 7.9$ Hz, 1H), 7.51 – 7.41 (m, 3H), 7.17 (d, $J = 7.7$ Hz, 1H), 7.03 (t, $J = 7.7$ Hz, 1H), 6.94 (dd, $J = 12.9, 7.8$ Hz, 2H), 6.06 (d, $J = 7.8$ Hz, 1H), 3.97 (s, 3H); 13C NMR (101 MHz, CDCl$_3$) δ 190.82, 148.57, 143.70 (d, $J = 3.8$ Hz), 139.52, 131.47, 129.88, 128.43, 127.45, 123.59, 121.14, 113.32, 111.20, 94.19 (d, $J = 3.8$ Hz), 56.00 (d, $J = 4.9$ Hz); HRMS-ESI (m/z) [M+Na]^+ calcd for C$_{16}$H$_{15}$NNaO$_2$, 276.0995; found: 276.0996.

(Z)-3-[(1,1'-biphenyl)-2-ylamino)-1-phenylprop-2-en-1-one (3ag)

1H NMR (400 MHz, CDCl$_3$) δ 12.03 (d, $J = 10.9$ Hz, 1H), 7.86 (d, $J = 6.9$ Hz, 2H), 7.61 – 7.31 (m, 11H), 7.28 (d, $J = 8.1$ Hz, 1H), 7.17 (t, $J = 7.4$ Hz, 1H), 5.97 (d, $J = 7.9$ Hz, 1H); 13C NMR (101 MHz, CDCl$_3$) δ 190.64, 144.88, 144.86, 139.39, 138.01, 137.92, 132.18, 131.41, 129.35, 129.16, 128.80, 128.33, 128.13, 127.43, 123.74, 115.32, 94.48; HRMS-ESI (m/z) [M+Na]^+ calcd for C$_{21}$H$_{17}$NNaO, 322.1202; found: 322.1203.

(Z)-1-phenyl-3-((4-vinylphenyl)amino)prop-2-en-1-one (3ah)

1H NMR (499 MHz, CDCl$_3$) δ 12.19 (d, $J = 11.9$ Hz, 1H), 7.94 (d, $J = 7.1$ Hz, 2H), 7.55 – 7.43 (m, 4H), 7.39 (d, $J = 8.5$ Hz, 2H), 7.06 (d, $J = 8.5$ Hz, 2H), 6.67 (dd, $J = 17.6, 10.9$ Hz, 1H), 6.04 (d, $J = 7.8$ Hz, 1H), 5.68 (d, $J = 17.6$ Hz, 1H), 5.20 (d, $J = 10.9$ Hz, 1H); 13C NMR (101 MHz, CDCl$_3$) δ 191.12, 144.62 (d, $J = 3.7$ Hz), 139.80, 139.27, 136.02, 133.37, 131.73, 128.57, 127.74, 127.44, 116.40, 113.01, 94.06 (d, $J = 3.7$ Hz); HRMS-ESI (m/z) [M+Na]^+ calcd for C$_{12}$H$_{15}$NNaO, 272.1046; found: 272.1048.
(Z)-3-((2-fluoro-4-methylphenyl)amino)-1-phenylprop-2-en-1-one (3ai)

\[ \text{1H NMR (400 MHz, CDCl}_3\text{) } \delta \text{ 12.17 (d, } J = 11.5 \text{ Hz, 1H), 7.95 (d, } J = 7.1 \text{ Hz, 2H), 7.52 – 7.43 (m, 4H), 7.09 (t, } J = 8.2 \text{ Hz, 1H), 6.94 (t, } J = 11.0 \text{ Hz, 1H), 6.93 (s, 1H), 6.07 (d, } J = 7.8 \text{ Hz, 1H), 2.31 (s, 3H); 13C NMR (101 MHz, CDCl}_3\text{) } \delta \text{ 191.18, 152.40 (d, } J = 245.7 \text{ Hz), 144.61, 139.21, 134.30 (d, } J = 6.9 \text{ Hz), 131.72, 128.53, 127.49, 126.36 (d, } J = 11.0 \text{ Hz), 125.39 (d, } J = 3.3 \text{ Hz), 116.86 (d, } J = 18.6 \text{ Hz), 115.62 (d, } J = 1.4 \text{ Hz), 94.46, 20.89; 19F NMR (376 MHz, CDCl}_3\text{) } \delta \text{ -130.9; HRMS-ESI (m/z) [M+Na]{}^+ \text{ calcd for C}_{16}H_{14}FNa, 278.0952; found: 278.0951.} \]

(Z)-3-(naphthalen-1-ylamino)-1-phenylprop-2-en-1-one (3aj)

\[ \text{1H NMR (499 MHz, CDCl}_3\text{) } \delta \text{ 13.08 (d, } J = 11.0 \text{ Hz, 1H), 8.26 (d, } J = 8.5 \text{ Hz, 1H), 8.01 (d, } J = 6.8 \text{ Hz, 2H), 7.88 (d, } J = 8.1 \text{ Hz, 1H), 7.74 (dd, } J = 11.8, 7.7 \text{ Hz, 1H), 7.62 (t, } J = 8.1 \text{ Hz, 2H), 7.56 (t, } J = 7.5 \text{ Hz, 1H), 7.54 – 7.45 (m, 4H), 7.31 (d, } J = 7.6 \text{ Hz, 1H), 6.18 (d, } J = 7.7 \text{ Hz, 1H); 13C NMR (101 MHz, CDCl}_3\text{) } \delta \text{ 191.52, 146.32, 146.30, 139.35, 136.58, 134.45, 131.78, 128.61, 127.52, 126.84, 126.77, 125.92, 124.98, 124.29, 121.18, 111.16, 94.82; HRMS-ESI (m/z) [M+Na]{}^+ \text{ calcd for C}_{19}H_{15}NNaO, 296.1046; found: 296.1047.} \]

9. References

10. NMR Spectra

$^1$H NMR (CDCl$_3$, 499 MHz)

$^{13}$C NMR (CDCl$_3$, 75 MHz)
$^{19}$F NMR (CDCl$_3$, 376 MHz)

(Z)-1-phenyl-3-(p-tolylamino)pro-2-en-1-one (3aa)

$^1$H NMR (CDCl$_3$, 499 MHz)
$^1$H NMR (CDCl$_3$, 400 MHz)

$^1$C NMR (CDCl$_3$, 101 MHz)
$^1$H NMR (CDCl$_3$, 400 MHz)

$^{13}$C NMR (CDCl$_3$, 75 MHz)
$^1$H NMR (CDCl$_3$, 400 MHz)

$^1$C NMR (CDCl$_3$, 101 MHz)
H NMR (CDCl₃, 400 MHz)

(Z)-1-(2-methoxyphenyl)-3-(p-tolyllamino)prop-2-en-1-one (3fa)

$^1$H NMR (CDCl₃, 400 MHz)

C NMR (CDCl₃, 101 MHz)

(Z)-1-(2-methoxyphenyl)-3-(p-tolyllamino)prop-2-en-1-one (3fa)

$^{13}$C NMR (CDCl₃, 101 MHz)
$^1$H NMR (CDCl$_3$, 400 MHz)

$^13$C NMR (CDCl$_3$, 101 MHz)
(Z)-1-(benzo[d][1,3]dioxol-5-yl)-3-(p-tolylamino)prop-2-en-1-one (3ha)

$^1$H NMR (CDCl$_3$, 400 MHz)

$^{13}$C NMR (CDCl$_3$, 101 MHz)
1H NMR (CDCl3, 400 MHz)

13C NMR (CDCl3, 101 MHz)
(Z)-1-[(1-methyl-1H-pyrrol-2-yl)-3-(p-tolylamino)prop-2-en-1-one (3ka)

$^1$H NMR (CDCl$_3$, 400 MHz)

$^{13}$C NMR (CDCl$_3$, 101 MHz)
$^1$H NMR (CDCl$_3$, 400 MHz)

$^{13}$C NMR (CDCl$_3$, 101 MHz)
$^1$H NMR (CDCl$_3$, 499 MHz)

$^{13}$C NMR (CDCl$_3$, 75 MHz)
(Z)-1-cyclopropyl-3-(p-tolylamino)prop-2-en-1-one (30a)

\(^1\)H NMR (CDCl\(_3\), 499 MHz)

\(^{13}\)C NMR (CDCl\(_3\), 75 MHz)
$^1$H NMR (CDCl$_3$, 499 MHz)

$^{13}$C NMR (CDCl$_3$, 75 MHz)
(Z)-2-[(p-tolylamino)methylene]-3,4-dihydronaphthalen-1(2H)-one (3qa)

$^1$H NMR (CDCl$_3$, 400 MHz)

$^{13}$C NMR (CDCl$_3$, 101 MHz)
(Z)-6-((p-tolylamino)methylene)-6,7,8,9-tetrahydro-5H-benzo[7]annulen-5-one (3ra)

$^1$H NMR (CDCl$_3$, 499 MHz)

$^{13}$C NMR (CDCl$_3$, 101 MHz)
$^1$H NMR (DMSO-$d_6$, 499 MHz)

$^{13}$C NMR (DMSO-$d_6$, 75 MHz)
\( \text{H NMR (DMSO-}d_6, 499 MHz) \)

\( \text{\(1^3\)C NMR (DMSO-}d_6, 101 MHz) \)
$^1$H NMR (DMSO-$_d_6$, 499 MHz)

$^{13}$C NMR (DMSO-$_d_6$, 75 MHz)
$\text{H NMR (DMSO-}d_6, 499 \text{ MHz)}$

$\text{C NMR (DMSO-}d_6, 75 \text{ MHz)}$
(E)-1-phenyl-2-((p-tolylamino)methylene)heptan-1-one (3wa)

$^1$H NMR (DMSO-$d_6$, 499 MHz)

$^13$C NMR (DMSO-$d_6$, 101 MHz)
$^1$H NMR (DMSO-$d_6$, 499 MHz)

(E)-3-methyl-1-phenyl-2-((p-tolylamino)methylene)butan-1-one (3a)

$^{13}$C NMR (DMSO-$d_6$, 101 MHz)
$^1$H NMR (CDCl$_3$, 499 MHz)

$^{13}$C NMR (CDCl$_3$, 75 MHz)
(2Z,2′Z)-1,1′-pyridine-2,6-diylibis(3-(p-tolylamino)prop-2-en-1-one) (3za)

$^1$H NMR (CDCl$_3$, 499 MHz)

(2Z,2′Z)-1,1′-pyridine-2,6-diylibis(3-(p-tolylamino)prop-2-en-1-one) (3za)

$^{13}$C NMR (CDCl$_3$, 75 MHz)
(Z)-1-phenyl-3-(phenylamino)prop-2-en-1-one (3ab)

\[ \text{\(^1\)H NMR (CDCl}_3, 400 MHz) \]

(Z)-3-((4-methoxyphenyl)amino)-1-phenylprop-2-en-1-one (3ae)

\[ \text{\(^1\)H NMR (CDCl}_3, 499 MHz) \]
$^1$H NMR (CDCl$_3$, 499 MHz)

(Z)-3-((4-chlorophenyl)amino)-1-phenylprop-2-ene-1-one (3ad)

$^1$H NMR (CDCl$_3$, 400 MHz)

(Z)-3-((4-bromophenyl)amino)-1-phenylprop-2-ene-1-one (3ae)

$^1$H NMR (CDCl$_3$, 400 MHz)
(Z)-3-((4-bromophenyl)amino)-1-phenylprop-2-en-1-one (3ae)

^{13}C NMR (CDCl$_3$, 101 MHz)

(Z)-3-((2-methoxyphenyl)amino)-1-phenylprop-2-en-1-one (3af)

$^{1}$H NMR (CDCl$_3$, 400 MHz)
\((Z)-3-((2\text{-methoxyphenyl})\text{amino})-1\text{-phenylprop-2-en-1-one} (3af)\)

\[\text{\^{13}C NMR (CDCl}_3, 101 \text{ MHz}}\]

\((Z)-3-((1,1\text{-diphenyl})\text{2-ylamino})-1\text{-phenylprop-2-en-1-one} (3ag)\)

\[\text{\^{1}H NMR (CDCl}_3, 400 \text{ MHz}}\]
(Z)-3-[(1,1'-biphenyl)-2-ylamino]-1-phenylprop-2-en-1-one (3ag)

$^{13}$C NMR (CDCl$_3$, 101 MHz)

(Z)-1-phenyl-3-[(4-vinylphenyl)amino]prop-2-en-1-one (3ah)

$^1$H NMR (CDCl$_3$, 499 MHz)
$^{13}$C NMR (CDCl$_3$, 101 MHz)

$^1$H NMR (CDCl$_3$, 400 MHz)
$^{13}$C NMR (CDCl$_3$, 101 MHz)

$^{19}$F NMR (CDCl$_3$, 376 MHz)
(Z)-3-(naphthalen-1-ylamino)-1-phenylprop-2-en-1-one (3a)

$\text{H NMR (CDCl}_3, 499 \text{ MHz)}$

$\text{C NMR (CDCl}_3, 101 \text{ MHz)}$

(3a)

$\text{C NMR (CDCl}_3, 101 \text{ MHz)}$