



recorded on UV-VIS NIR 3600 SHIMADZU spectrophotometer in region 250–700 nm.  $^1\text{H}$  and  $^{13}\text{C}$  NMR spectra were measured in  $\text{CDCl}_3$  and  $\text{DMSO}-d_6$  solution using spectrophotometers Varian VXR-300 (for  $^1\text{H}$  299,995 MHz, for  $^{13}\text{C}$  75,431 MHz) and Bruker Avance III 400 (for  $^1\text{H}$  400,23 MHz, for  $^{13}\text{C}$  100,65 MHz) at 25 °C. Chemical shifts ( $\delta$ -scale) are quoted in parts per million and following abbreviations are used: s = singlet; d = doublet; t = triplet; q = quartet, m = multiplet.

## Experimental

Preparation of the title compounds was realized according to Fig. 1.

### General procedure for preparation of arylaminomethylidene derivatives III

Suitable substituted or unsubstituted aniline **Ia-f** (5 mmol) dissolved in MeOH/EtOH (15–20 ml) was mixed with equimolar amount of enolether **IIa-d** (5 mmol) in MeOH/EtOH (15–20 ml). Reaction mixture was stirred at rt from 15 min up to 3 h at reflux. After end of the reaction solvent was evaporated through vacuum evaporator. Thus obtained raw products were purified by column chromatography or recrystallization. Purification and individual reaction conditions for all compounds are presented.

#### Dimethyl 2-[(phenylamino)methylidene]malonate (IIIa, Z = H)

There was used to a reaction aniline (**Ia**, **Z = H**) (0.47 g, 5 mmol) in 10 ml MeOH and enolether **IIa** (0.87 g, 5 mmol) in 15 ml MeOH. Reaction mixture was stirred 20 min at rt, then solvent was evaporated. Raw product was purified by column chromatography (EA (ethyl acetate):H(hexane) = 1:1,  $R_F$  = 0.24).

Yield: 1.02 g, 87 %, light yellow solid. Mp 46–48 °C (47 °C, Rappoport et al. 1972).

$^1\text{H}$  NMR (300 MHz,  $\text{CDCl}_3$ )  $\delta$  (ppm) 11.01 (br d, 1H,  $^3J$  = 13.1 Hz, —NH), 8.54 (d, 1H,  $^3J$  = 13.8 Hz, —CH=), 7.41–7.36 (m, 2H), 7.20–7.14 (m, 3H), 3.87 (s, 3H, OMe), 3.78 (s, 3H, OMe)

UV VIS (DMSO, nm)  $\lambda_{\text{max}}$  319

Elemental analysis for  $\text{C}_{12}\text{H}_{13}\text{NO}_4$  — calculated (found) (%): C 61.27 (61.25), H 5.57 (5.52), N 5.95 (5.91)

#### Diethyl 2-[(phenylamino)methylidene]malonate (IIIb, Z = H)

There was used to a reaction aniline (**Ia**, **Z = H**) (0.47 g, 5 mmol) in 10 ml EtOH and enolether **IIb** (1.1 g, 5 mmol) in 15 ml EtOH. Reaction mixture was stirred 20 min at rt, then solvent was evaporated.

Raw product was purified by column chromatography. (EA:H = 1:1,  $R_F$  = 0.29).

Yield: 1.25 g, 93 %, light yellow solid. Mp 51–53 °C (54–55 °C, Huppertz et al. 1981).

$^1\text{H}$  NMR (300 MHz,  $\text{CDCl}_3$ )  $\delta$  (ppm) 11.01 (br d, 1H,  $^3J$  = 13.5 Hz, —NH), 8.53 (d, 1H,  $^3J$  = 13.7 Hz, —CH=), 7.38 (m, 2H), 7.16 (m, 3H), 4.32 (q, 2H,  $^3J$  = 7.1 Hz,  $\text{OCH}_2$ ), 4.25 (q, 2H,  $^3J$  = 7.1 Hz,  $\text{OCH}_2$ ), 1.39 (t, 3H,  $^3J$  = 7.1 Hz,  $\text{CH}_3$ ), 1.33 (t, 3H,  $^3J$  = 7.1 Hz,  $\text{CH}_3$ )

UV VIS (DMSO, nm)  $\lambda_{\text{max}}$  321

#### 3-[(Phenylamino)methylidene]pentane-2,4-dione (IIIc, Z = H)

There was used to a reaction aniline (**Ia**, **Z = H**) (0.47 g, 5 mmol) in 10 ml EtOH and enolether **IIc** (0.78 g, 5 mmol) in 15 ml EtOH. Reaction mixture was stirred 15 min at rt and solvent was evaporated. Raw product was purified by column chromatography (EA:H = 1:1,  $R_F$  = 0.43).

Yield: 0.94 g, 94 %, yellow solid. Mp 85–87 °C (90 °C, Wolfbeis et al. 1979).

$^1\text{H}$  NMR (300 MHz,  $\text{CDCl}_3$ )  $\delta$  (ppm) 12.70 (br s, 1H, —NH), 8.19 (d, 1H,  $^3J$  = 12.8 Hz, —CH=), 7.38–7.33 (m, 2H), 7.19–7.11 (m, 3H), 2.50 (s, 3H, Me), 2.33 (s, 3H, Me)

UV VIS (DMSO, nm)  $\lambda_{\text{max}}$  330

Elemental analysis for  $\text{C}_{12}\text{H}_{13}\text{NO}_2$  — calculated (found) (%): C 70.92 (70.77), H 6.45 (6.33), N 6.89 (6.78)

#### 2-[(Phenylamino)methylidene]malononitrile (IIIId, Z = H)

There was used to a reaction aniline (**Ia**, **Z = H**) (0.47 g, 5 mmol) in 10 ml EtOH and enolether **IIId** (0.61 g, 5 mmol) in 15 ml EtOH. Reaction mixture was stirred 20 min at rt and solvent was evaporated. Raw product was purified by column chromatography (EA:H = 1:1,  $R_F$  = 0.15).

Yield: 0.74 g, 88 %, yellow powder. Mp 248–249 °C (254–256 °C, Nasakin et al. 1992).

$^1\text{H}$  NMR (300 MHz,  $\text{DMSO}-d_6$ )  $\delta$  (ppm) 11.1 (br d, 1H, —NH), 8.51 (s, 1H, —CH=), 7.46–7.36 (m, 4H), 7.20–7.16 (m, 1H)

UV VIS (DMSO, nm)  $\lambda_{\text{max}}$  317

Elemental analysis for  $\text{C}_{10}\text{H}_7\text{N}_3$  — calculated (measured) (%): C 70.99 (70.86), H 4.17 (4.06), N 24.84 (24.91)

#### Dimethyl 2-[(3-chlorophenylamino)methylidene]malonate (IIIa, Z = 3-Cl)

There was used to a reaction 3-chloroaniline (**Ib**, **Z = 3-Cl**) (0.64 g, 5 mmol) in 10 ml MeOH and enolether **IIa** (0.87 g, 5 mmol) in 20 ml MeOH. Reaction mixture was refluxed under regular stirring for 2 h. After its cooling solvent was evaporated and a raw

product was purified by column chromatography. (EA:H = 1:1,  $R_F$  = 0.35).

Yield: 1.21 g, 90 %, white powder. Mp 146–147 °C.

$^1\text{H NMR}$  (300 MHz,  $\text{CDCl}_3$ )  $\delta$  (ppm) 11.01 (d, 1H,  $^3J$  = 13.8 Hz, —NH), 8.47 (d, 1H,  $^3J$  = 13.6 Hz, —CH=), 7.32–7.11 (m, 3H), 7.03–7.01 (m, 1H), 3.86 (s, 3H, OMe), 3.78 (s, 3H, OMe)

**IR** ( $\nu$ ,  $\text{cm}^{-1}$ ) 2954, 1588, 1573, 1231, 769

**UV VIS** (DMSO, nm)  $\lambda_{\text{max}}$  321

**Elemental analysis** for  $\text{C}_{12}\text{H}_{12}\text{ClNO}_4$  calculated (found) (%): C 53.44 (53.24), H 4.49 (4.48), N 5.19 (4.96)

*Diethyl 2-[(3-chlorophenylamino)methylidene]malonate (IIIb, Z = 3-Cl)*

There was used to a reaction 3-chloroaniline (**Ib**, **Z = 3-Cl**) (0.64 g, 5 mmol) in 15 ml EtOH and enolether **IIb** (1.1 g, 5 mmol) in 20 ml EtOH. Reaction mixture was refluxed under regular stirring for 2 h. After its cooling solvent was evaporated and a raw product was purified by column chromatography (EA:H = 1:1,  $R_F$  = 0.40).

Yield: 1.44 g, 95 %, colourless solid. Mp 41–44 °C (44–45 °C, Fodor et al. 1948).

$^1\text{H NMR}$  (300 MHz,  $\text{CDCl}_3$ )  $\delta$  (ppm) 10.98 (d, 1H,  $^3J$  = 13.3 Hz, —NH), 8.46 (d, 1H,  $^3J$  = 13.5 Hz, —CH=), 7.33–7.28 (m, 1H), 7.15–7.11 (m, 2H), 7.03–7.00 (m, 1H), 4.32 (q, 2H,  $^3J$  = 7.2 Hz,  $\text{OCH}_2$ ), 4.26 (q, 2H,  $^3J$  = 7.1 Hz,  $\text{OCH}_2$ ), 1.39 (t, 3H,  $^3J$  = 7.1 Hz,  $\text{CH}_3$ ), 1.35 (t, 3H,  $^3J$  = 7.1 Hz,  $\text{CH}_3$ )

**UV VIS** (DMSO, nm)  $\lambda_{\text{max}}$  300

*3-[(3-Chlorophenylamino)methylidene]pentane-2,4-dione (IIIc, Z = 3-Cl)*

There was used to a reaction 3-chloroaniline (**Ib**, **Z = 3-Cl**) (0.64 g, 5 mmol) in 10 ml EtOH and enolether **IIc** (0.79 g, 5 mmol) in 20 ml EtOH. Reaction mixture was refluxed under regular stirring for 1 h. After its cooling solvent was evaporated and a raw product was purified by column chromatography (EA:H = 1:1,  $R_F$  = 0.22).

Yield: 1.16 g, 98 %, light yellow solid. Mp 89–91 °C (92–94 °C, Snyder 1946).

$^1\text{H NMR}$  (300 MHz,  $\text{DMSO}-d_6$ )  $\delta$  (ppm) 12.43 (d, 1H,  $^3J$  = 12.7 Hz, —NH), 8.40 (d, 1H,  $^3J$  = 12.7 Hz, —CH=), 7.73–7.40 (m, 3H), 7.25–7.21 (m, 1H), 2.39 (s, 6H, 2×Me)

**UV VIS** (DMSO, nm)  $\lambda_{\text{max}}$  342

**IR** ( $\nu$ ,  $\text{cm}^{-1}$ ) 3057, 1589, 1567, 1306, 778

**Elemental analysis** for  $\text{C}_{12}\text{H}_{12}\text{ClNO}_2$  — calculated (found) (%): C 60.64 (60.70), H 5.09 (5.07), N 5.89 (5.71)

*2-[(3-Chlorophenylamino)methylidene]malononitrile (IIIId, Z = 3-Cl)*

There was used to a reaction 3-chloroaniline (**Ib**,

**Z = 3-Cl**) (0.64 g, 5 mmol) in 10 ml EtOH and enolether **IIId** (0.61 g, 5 mmol) in 15 ml EtOH. Reaction mixture was refluxed under regular stirring for 1 h. After its cooling solvent was evaporated and a raw product was purified by column chromatography (EA:H = 1:1,  $R_F$  = 0.28).

Yield: 0.96 g, 90 %, colourless solid. Mp 199–200 °C (200–201 °C, Santilli 1964).

$^1\text{H NMR}$  (300 MHz,  $\text{DMSO}-d_6$ )  $\delta$  (ppm) 11.16 (br s, 1H, —NH), 8.57 (s, 1H, —CH=), 7.59 (s, 1H), 7.40–7.20 (m, 3H)

**UV VIS** (DMSO, nm)  $\lambda_{\text{max}}$  325

**IR** ( $\nu$ ,  $\text{cm}^{-1}$ ) 3203, 2227, 2216, 1662, 1338, 772

**Elemental analysis** for  $\text{C}_{10}\text{H}_6\text{ClN}_3$  — calculated (found) (%): C 58.98 (58.84), H 2.97 (2.94), N 20.64 (20.51)

*Dimethyl 2-[(4-bromophenylamino)methylidene]malonate (IIIa, Z = 4-Br)*

There was used to a reaction 4-bromoaniline (**Ic**, **Z = 4-Br**) (0.86 g, 5 mmol) in 15 ml MeOH and enolether **IIa** (0.87 g, 5 mmol) in 20 ml MeOH. Reaction mixture was refluxed under regular stirring for 1.5 h. After its cooling raw product was collected by filtration and washed off with cold MeOH.

Yield: 1.15 g, 73 %, white powder. Mp 113–114 °C (116–117 °C, Rappoport et al. 1972).

$^1\text{H NMR}$  (300 MHz,  $\text{DMSO}-d_6$ )  $\delta$  (ppm) 10.66 (s, 1H, —NH), 8.38 (br s, 1H, —CH=), 7.55 (d, 2H,  $^3J$  = 8.9 Hz, H-3, H-5), 7.37 (d, 2H,  $^3J$  = 8.9 Hz, H-2, H-6), 3.72 (s, 3H, OMe), 3.66 (s, 3H, OMe)

**UV VIS** (DMSO, nm)  $\lambda_{\text{max}}$  328

**Elemental analysis** for  $\text{C}_{12}\text{H}_{12}\text{BrNO}_4$  — calculated (found) (%): C 45.88 (45.94), H 3.85 (3.82), N 4.46 (4.28)

*Diethyl 2-[(4-bromophenylamino)methylidene]malonate (IIIb, Z = 4-Br)*

There was used to a reaction 4-bromoaniline (**Ic**, **Z = 4-Br**) (0.86 g, 5 mmol) in 15 ml EtOH and enolether **IIb** (1.1 g, 5 mmol) in 20 ml EtOH. Reaction mixture was refluxed under regular stirring for 1.5 h. After its cooling raw product was purified by column chromatography.

Yield: 1.60 g, 92 %, colourless solid. Mp 98–99 °C (100.5–101.7 °C, Lopez et al. 2010).

$^1\text{H NMR}$  (300 MHz,  $\text{DMSO}-d_6$ )  $\delta$  (ppm) 10.65 (d, 1H,  $^3J$  = 13.8 Hz, —NH), 8.34 (d, 1H,  $^3J$  = 13.8 Hz, —CH=), 7.54 (d, 2H,  $^3J$  = 8.8 Hz, H-3, H-5), 7.34 (d, 2H,  $^3J$  = 8.9 Hz, H-2, H-6), 4.19 (q, 2H,  $^3J$  = 7.1 Hz,  $\text{OCH}_2$ ), 4.11 (q, 2H,  $^3J$  = 7.1 Hz,  $\text{OCH}_2$ ), 1.25 (t, 3H,  $^3J$  = 7.1 Hz,  $\text{CH}_3$ ), 1.21 (t, 3H,  $^3J$  = 7.1 Hz,  $\text{CH}_3$ )

**UV VIS** (DMSO, nm)  $\lambda_{\text{max}}$  323

**Elemental analysis** for  $\text{C}_{14}\text{H}_{16}\text{BrNO}_4$  — calculated (found) (%): C 49.14 (49.20), H 4.71 (4.77), N 4.09 (3.91)

*3-[(4-Bromophenylamino)methylidene]pentane-2,4-dione*  
(IIIc, Z = 4-Br)

There was used to a reaction 4-bromoaniline (**Ic**, Z = 4-Br) (0.86 g, 5 mmol) in 10 ml EtOH and enolether **Iic** (0.79 g, 5 mmol) in 20 ml EtOH. Reaction mixture was stirred for 30 min at rt. Solvent was evaporated and a raw product was collected by filtration and finally washed off with cold EtOH.

Yield: 1.30 g, 91 %, colourless crystals. Mp 139–140 °C.

<sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ (ppm) 12.72 (br s, 1H, —NH), 8.16 (d, 1H, <sup>3</sup>J = 12.6 Hz, —CH=), 7.50 (d, 2H, <sup>3</sup>J = 8.7 Hz, H-3, H-5), 7.05 (d, 2H, <sup>3</sup>J = 8.7 Hz, H-2, H-6), 2.54 (s, 3H, Me), 2.38 (s, 3H, Me)

UV VIS (DMSO, nm) λ<sub>max</sub> 338

Elemental analysis for C<sub>12</sub>H<sub>12</sub>BrNO<sub>2</sub> — calculated (found) (%): C 51.09 (51.00), H 4.29 (4.18), N 4.96 (4.89)

*2-[(4-Bromophenylamino)methylidene]malononitrile*  
(IIIId, Z = 4-Br)

There was used to a reaction 4-bromoaniline (**Ic**, Z = 4-Br) (0.86g, 5 mmol) in 15 ml EtOH and enolether **IId** (0.61 g, 5 mmol) in 20 ml MeOH. Reaction mixture was stirred for 30 min at rt. Solvent was evaporated and a raw product was collected by filtration and finally washed with cold EtOH.

Yield: 0.69 g, 56 %, colourless solid. Mp 251–253 °C.

<sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>) δ (ppm) 11.17 (br s, 1H, —NH), 8.50 (s, 1H, —CH=), 7.55 (d, 2H, <sup>3</sup>J = 8.8 Hz, H-3, H-5), 7.39 (d, 2H, <sup>3</sup>J = 8.8 Hz, H-2, H-6)

IR (ν, cm<sup>-1</sup>) 3217, 2225, 2209, 1668, 811

UV VIS (DMSO, nm) λ<sub>max</sub> 330

Elemental analysis for C<sub>10</sub>H<sub>6</sub>BrN<sub>3</sub> — calculated (found) (%): C 48.41 (48.35), H 2.44 (2.43), N 16.94 (16.81)

*Dimethyl 2-[(p-tolylamino)methylidene]malonate*  
(IIIa, Z = 4-Me)

There was used to a reaction 4-methylaniline (**Id**, Z = 4-Me) (0.54 g, 5 mmol) in 8–10 ml MeOH and enolether **Iia** (0.87 g, 5 mmol) in 8–10 ml MeOH. Reaction mixture was refluxed under regular stirring for 3 h. After its cooling raw product was recrystallized from petrolether.

Yield: 0.67 g, 54 %, colourless solid. Mp 104–106 °C (108–110 °C, Rappoport et al. 1972).

<sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ (ppm) 11.01 (br s, 1H, —NH), 8.52 (d, 1H, <sup>3</sup>J = 13.6 Hz, —CH=), 7.55 (d, 2H, <sup>3</sup>J = 8.9 Hz, H-2, H-6), 7.17 (d, 2H, <sup>3</sup>J = 8.4 Hz, H-3, H-5), 3.86 (s, 3H, OMe), 3.78 (s, 3H, OMe), 2.34 (s, 3H, Me)

UV VIS (DMSO, nm) λ<sub>max</sub> 330

Elemental analysis for C<sub>13</sub>H<sub>15</sub>NO<sub>4</sub> — calculated

(found) (%): C 62.64 (62.69), H 6.07 (5.96), N 5.62 (5.44)

*Diethyl 2-[(p-tolylamino)methylidene]malonate*  
(IIIb, Z = 4-Me)

There was used to a reaction 4-methylaniline (**Id**, Z = 4-Me) (0.54 g, 5 mmol) in 8-10 ml EtOH and enolether **Iib** (1.1 g, 5 mmol) in 8-10 ml EtOH. Reaction mixture was refluxed under regular stirring for 3 h. After its cooling solvent was evaporated and a raw product was purified by column chromatography (EA:H = 1:1, R<sub>F</sub> = 0.20).

Yield: 1.31 g, 93 %, colourless solid. Mp 46–48 °C (45–46 °C, Mao et al. 2009).

<sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ (ppm) 11.00 (d, 1H, <sup>3</sup>J = 13.6 Hz, —NH), 8.52 (d, 1H, <sup>3</sup>J = 13.8 Hz, —CH=), 7.18 (d, 2H, <sup>3</sup>J = 8.3 Hz, H-2, H-6), 7.05 (d, 2H, <sup>3</sup>J = 8.4 Hz, H-3, H-5), 4.32 (q, 2H, <sup>3</sup>J = 7.1 Hz, OCH<sub>2</sub>), 4.26 (q, 2H, <sup>3</sup>J = 7.1 Hz, OCH<sub>2</sub>), 1.40 (t, 3H, <sup>3</sup>J = 7.1 Hz, CH<sub>3</sub>), 1.35 (t, 3H, <sup>3</sup>J = 7.1 Hz, CH<sub>3</sub>)

UV VIS (DMSO, nm) λ<sub>max</sub> 334

*3-[(p-Tolylamino)methylidene]pentane-2,4-dione*  
(IIIc, Z = 4-Me)

There was used to a reaction 4-methylaniline (**Id**, Z = 4-Me) (0.54 g, 5 mmol) in 10 ml EtOH and enolether **Iic** (0.78 g, 5 mmol) in 20 ml EtOH. Reaction mixture was stirred for 1 h at rt. Solvent was evaporated and a raw product was collected by filtration and washed off with hexane.

Yield: 0.84 g, 77 %, light yellow cotton. Mp 136–138 °C (139–140 °C, Claisen 1893).

<sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ (ppm) 12.75 (br s, 1H, —NH), 8.22 (d, 1H, <sup>3</sup>J = 12.8 Hz, —CH=), 7.19 (d, 2H, <sup>3</sup>J = 8.2 Hz, H-2, H-6), 7.08 (d, 2H, <sup>3</sup>J = 8.4 Hz, H-3, H-5), 2.57 (s, 3H, 4-Me), 2.39 (s, 3H, Me), 2.37 (s, 3H, Me)

UV VIS (DMSO, nm) λ<sub>max</sub> 345

IR (ν, cm<sup>-1</sup>) 2926, 1595, 1570, 1308, 814

Elemental analysis for C<sub>13</sub>H<sub>15</sub>NO<sub>2</sub> — calculated (found) (%): C 71.87 (71.85), H 6.96 (6.91), N 6.45 (6.32)

*2-[(p-Tolylamino)methylidene]malononitrile*  
(IIIId, Z = 4-Me)

There was used to a reaction 4-methylaniline (**Id**, Z = 4-Me) (0.54 g, 5 mmol) in 10 ml EtOH and enolether **IId** (0.61 g, 5 mmol) in 20 ml EtOH. Reaction mixture was stirred for 40 min at rt. Solvent was evaporated and a raw product was washed off with cold EtOH.

Yield: 0.71 g, 76 %, yellow crystals. Mp 289–290 °C (297 °C, Rappoport et al. 1972).

<sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>) δ (ppm) 11.06 (br s, 1H, —NH), 8.40 (s, 1H, —CH=), 7.27 (d, 2H,

$^3J = 8.4$  Hz, H-2, H-6), 7.14 (d, 2H,  $^3J = 8.4$  Hz, H-3, H-5), 2.24 (s, 3H, Me)

**UV VIS** (DMSO, nm)  $\lambda_{\max}$  320

**Elemental analysis** for  $C_{11}H_9N_3$  – calculated (found) (%): 72.11 (72.02), H 4.95 (4.88), N 22.94 (22.89)

*Dimethyl 2-[(4-methoxyphenylamino)methylidene]malonate (IIIa, Z = 4-OMe)*

There was used to a reaction 4-methoxyaniline (**Ie**, **Z = 4-OMe**) (0.62 g, 5 mmol) in 15 ml MeOH and enolether **IIa** (0.87 g, 5 mmol) in 15–20 ml MeOH. Reaction mixture was refluxed under regular stirring for 3 h. After its cooling raw product was recrystallized from petrolether.

Yield: 0.64 g, 48 %, colourless solid. Mp 86–89 °C (89–91 °C, Zewge et al. 2007).

**$^1H$  NMR** (400 MHz,  $CDCl_3$ )  $\delta$  (ppm) 11.02 (br s, 1H, —NH), 8.45 (d, 1H,  $^3J = 8.7$  Hz, —CH=), 7.11–6.89 (m, 4H), 3.86 (s, 3H, OMe), 3.81 (s, 3H, OMe), 3.78 (s, 3H, 4-OMe)

**IR** ( $\nu$ ,  $cm^{-1}$ ) 2946, 1683, 1645, 1449, 1266, 1241, 797

**UV VIS** (DMSO, nm)  $\lambda_{\max}$  324

**Elemental analysis** for  $C_{13}H_{15}NO_5$  – calculated (found) (%): C 58.86 (58.74), H 5.70 (5.65), N 5.28 (5.12)

*Diethyl 2-[(4-methoxyphenylamino)methylidene]malonate (IIIb, Z = 4-OMe)*

There was used to a reaction 4-methoxyaniline (**Ie**, **Z = 4-OMe**) (0.62 g, 5 mmol) in 10 ml MeOH and enolether **IIb** (1.1 g, 5 mmol) in 20 ml EtOH. Reaction mixture was refluxed under regular stirring for 3 h. After its cooling raw product was purified by column chromatography (EA:H = 10:1,  $R_F = 0.20$ ).

Yield: 1.22 g, 82 %, orange solid. Mp < 35 °C (38–39 °C, Deshmukh et al. 1999).

**$^1H$  NMR** (300 MHz,  $CDCl_3$ )  $\delta$  (ppm) 10.95 (br s, 1H, —NH), 8.40 (d, 1H,  $^3J = 13.8$  Hz, —CH=), 7.04 (d, 2H,  $^3J = 8.7$  Hz, H-2, H-6), 6.86 (d, 2H,  $^3J = 8.7$  Hz, H-3, H-5), 4.31–4.17 (m, 4H,  $OCH_2$ ), 3.76 (s, 3H, OMe), 1.37–1.27 (m, 6H,  $CH_3$ )

**UV VIS** (DMSO, nm)  $\lambda_{\max}$  321

*2-[(4-Methoxyphenylamino)methylidene]pentane-2,4-dione (IIIc, Z = 4-OMe)*

There was used to a reaction 4-methoxyaniline (**Ie**, **Z = 4-OMe**) (0.62 g, 5 mmol) in 10 ml EtOH and enolether **IIc** (0.78 g, 5 mmol) in 20 ml EtOH. Reaction mixture was stirred for 1 h at rt. Solvent was evaporated and a raw product was purified by column chromatography (EA:H = 1:1,  $R_F = 0.31$ ).

Yield: 1.09 g, 95 %, light yellow solid. Mp 85–87 °C.

**$^1H$  NMR** (300 MHz,  $CDCl_3$ )  $\delta$  (ppm) 12.79 (br s, 1H, —NH), 8.15 (d, 1H,  $^3J = 12.8$  Hz, —CH=), 7.12 (d,

2H,  $^3J = 8.9$  Hz, H-2, H-6), 6.93 (d, 2H,  $^3J = 8.9$  Hz, H-3, H-5), 3.83 (s, 3H, OMe), 2.55 (s, 3H, Me), 2.37 (s, 3H, Me)

**IR** ( $\nu$ ,  $cm^{-1}$ ) 2990, 1616, 1594, 1318, 1249, 979, 828

**UV VIS** (DMSO, nm)  $\lambda_{\max}$  342

**Elemental analysis** for  $C_{13}H_{15}NO_3$  – calculated (found) (%): C 66.94 (66.78), H 6.48 (6.37), N 6.00 (5.92)

*2-[(4-methoxyphenylamino)methylidene]malononitrile (IIIId, Z = 4-OMe)*

There was used to a reaction 4-methoxyaniline (**Ie**, **Z = 4-OMe**) (0.62 g, 5 mmol) in 10 ml EtOH and enolether **IIId** (0.61 g, 5 mmol) in 15 ml EtOH. Reaction mixture was stirred for 30 min at rt. Raw product was collected by filtration and washed off with hexane.

Yield: 0.70 g, 70 %, light yellow powder. Mp 266–267 °C (245 °C, Rappoport et al. 1972).

**$^1H$  NMR** (300 MHz,  $DMSO-d_6$ )  $\delta$  (ppm) 11.03 (br s, 1H, —NH), 8.37 (s, 1H, —CH=), 7.35 (d, 2H,  $^3J = 9.0$  Hz, H-2, H-6), 6.93 (d, 2H,  $^3J = 9.0$  Hz, H-3, H-5), 3.74 (s, 3H, OMe)

**UV VIS** (DMSO, nm)  $\lambda_{\max}$  335

**Elemental analysis** for  $C_{11}H_9N_3O$  – calculated (found) (%): C 66.32 (66.41), H 4.55 (4.49), N 21.09 (21.06)

*Dimethyl 2-[(4-nitrophenylamino)methylidene]malonate (IIIa, Z = 4-NO<sub>2</sub>)*

There was used to a reaction 4-nitroaniline (**If**, **Z = 4-NO<sub>2</sub>**) (0.69 g, 5 mmol) in 10 ml MeOH and enolether **IIa** (0.87 g, 5 mmol) in 20 ml MeOH. Reaction mixture was refluxed under regular stirring for 3 h. After its cooling raw product was collected by filtration and washed off with cold MeOH.

Yield: 0.91 g, 65 %, yellow crystalline product. Mp 156–157 °C.

**$^1H$  NMR** (400 MHz,  $CDCl_3$ )  $\delta$  (ppm) 11.23 (br s, 1H, —NH), 8.54 (br s, 1H, —CH=), 8.25 (d, 2H,  $^3J = 9.1$  Hz, H-3, H-5), 7.62 (d, 2H,  $^3J = 9.1$  Hz, H-2, H-6), 3.88 (s, 3H, OMe), 3.82 (s, 3H, OMe)

**UV VIS** (DMSO, nm)  $\lambda_{\max}$  367

**Elemental analysis** for  $C_{12}H_{12}N_2O_6$  – calculated (found) (%): C 51.43 (51.35), H 4.32 (4.28), N 10.00 (9.90)

*Diethyl 2-[(4-nitrophenylamino)methylidene]malonate (IIIb, Z = 4-NO<sub>2</sub>)*

There was used to a reaction 4-nitroaniline (**If**, **Z = 4-NO<sub>2</sub>**) (0.69 g, 5 mmol) in 10 ml EtOH and enolether **IIb** (1.1 g, 5 mmol) in 20 ml EtOH. Reaction mixture was refluxed under regular stirring for 3 h. After its cooling raw product was collected by filtration and washed off with cold EtOH.

Yield: 1.13 g, 72 %, yellow solid. Mp 136–138 °C (142 °C, Darque et al. 2009).

<sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ (ppm) 11.20 (br s, 1H, —NH), 8.51 (s, 1H, —CH=), 8.20 (d, 2H, <sup>3</sup>J = 9.2 Hz, H-3, H-5), 7.60 (d, 2H, <sup>3</sup>J = 9.2 Hz, H-2, H-6), 4.23 (q, 2H, <sup>3</sup>J = 7.1 Hz, OCH<sub>2</sub>), 4.15 (q, 2H, <sup>3</sup>J = 7.1 Hz, OCH<sub>2</sub>), 1.28 (t, 3H, <sup>3</sup>J = 7.0 Hz, CH<sub>3</sub>), 1.24 (t, 3H, <sup>3</sup>J = 7.0 Hz, CH<sub>3</sub>)

UV VIS (DMSO, nm) λ<sub>max</sub> 373

## Results and discussion

To avoid geometrical isomerism on the double bond of the products of the nucleophilic vinylic substitution (Saloň et al. 2005) and therefore thus influence the kinetics of this reaction, we decided to prepare only compounds with the same substituents on the β - position of the double bond, e.g. on the opposite side to arylamino substituent. Therefore, reaction of the dimethyl methoxymethylidene malonate (IIa), diethyl ethoxymethylidene malonate (IIb), ethoxymethylidene pentane-2,4-dione (IIc) and ethoxymethylidene malononitrile (IId) respectively, with the appropriate substituted aniline in the same alcohol like alkoxy and/or ester group was used for the preparation of target compounds.

### NMR spectra and the substituent chemical shifts (SCS) of the analogous anilinomethylidene (nonsubstituted) compounds

In 1989 Goljer and co-workers published carbon spectra of the appropriate anilinomethylidene (nonsubstituted) compounds IV (equal to III: Z = H) (Tab. 1, 2).

### NMR spectra and the substituent chemical shifts (SCS) of the studied compounds

Applying of the published substituent chemical

shifts (Ewing 1979) of the substituents of the benzene ring regarding the position of the substituent it is possible to calculate the chemical shifts of the studied compounds and compare them with measured ones to assign them. Of course, there exist some other methods like the use of plugins of the drawing programs like ISIS Draw®, ChemDraw® etc., but the used algorithm is not clear. Another alternative is the calculation using semiempirical/*ab initio* calculation methods.

In the Table 3 there are presented all our recorded <sup>13</sup>C NMR spectra in first row for each compound. In next row(s) there are presented calculated values obtained using appropriate values for aminoethylidene substituent in deuteriochloroform (Table 3)/ hexadeuterodimethyl sulfoxide (Table 1) and for the appropriate substituent in right position and solvent. In all cases we used also data for neat liquid (NL). In the column Reference, there are references for the work, where the spectra of the corresponding compounds were appeared first.

Substituent chemical shifts are frequently used in proton and carbon spectra for founding of the unknown values of the chemical shifts of the new synthesized substances to confirm their structure. Some of them are used like sole, some should be combined. We selected the second approach, e.g. combine SCS for monosubstituted benzenes to obtain set of datas for disubstituted benzenes. Data for proton spectra are more sensitive for solvents, for example dimethyl sulfoxide (Rao et al. 1974). Carbon data reflecting well electron distribution in the molecule and therefore SCS are in relation with electron properties of the substituents.

β,β-Disubstituted aminoethylidene substituent with two electron withdrawing groups displayed an interesting influence on benzene ring: on ipso carbon it has practically the same values like on ortho car-

**Tab. 1.** <sup>13</sup>C chemical shifts of the studied compounds IV in DMSO-d<sub>6</sub>.

| Compound | C - 1 | C - 2 | C - 3 | C - 4 | C - 7 | C - 8 | Others                   |
|----------|-------|-------|-------|-------|-------|-------|--------------------------|
| IVa      | 139.3 | 117.6 | 129.6 | 124.7 | 151.4 | 92.6  | 165.2, 167.5, 51.0, 50.9 |
| IVb      | 139.3 | 117.3 | 129.6 | 124.6 | 150.9 | 93.6  | 165.0, 167.4             |
| IVc      | 139.0 | 118.1 | 129.3 | 125.3 | 155.5 | 52.0  | 116.3, 113.9             |
| IVd      | 139.0 | 118.2 | 129.5 | 125.2 | 152.6 | 112.4 | 195.1, 199.4, 27.4, 31.4 |

**Tab. 2.** Substituent chemical shifts of the studied compounds IV in DMSO-d<sub>6</sub>.

| Compound | C - 1 | C - 2 | C - 3 | C - 4 |
|----------|-------|-------|-------|-------|
| IVa      | +10.5 | -10.3 | +1.0  | -3.3  |
| IVb      | +10.5 | -10.4 | +0.8  | -3.2  |
| IVc      | +10.8 | -10.9 | +1.1  | -3.8  |
| IVd      | +10.8 | -11.2 | +1.1  | -3.9  |

**Tab. 3.**  $^{13}\text{C}$  NMR spectra of arylaminomethylidene derivatives III.

| Compound No. | Z     | Solvent           | C-1   | C-2   | C-3   | C-4   | C-5   | C-6   | Other signals   | Reference          |
|--------------|-------|-------------------|-------|-------|-------|-------|-------|-------|---|--------------------|
| IIIa         | H     | $\text{CDCl}_3$   | 139.0 | 117.2 | 129.0 | 125.1 | 129.0 | 117.2 | 51.4, 51.6, 92.8, 152.2, 166.0, 169.3                   | Mohri et al. 1999  |
| IIIb         | H     | $\text{CDCl}_3$   | 139.2 | 117.2 | 129.7 | 124.9 | 129.7 | 117.2 | 14.3, 14.5, 60.1, 60.4, 93.3, 152.2, 165.9, 169.2       | Loupy et al. 2005  |
| IIIc         | H     | $\text{CDCl}_3$   | 138.5 | 117.3 | 129.5 | 125.3 | 129.5 | 117.3 | 26.9, 31.6, 112.7, 151.4, 194.4, 200.5                  | Hansen et al. 1995 |
| IIId         | H     | $\text{DMSO-}d_6$ | 139.7 | 118.5 | 129.9 | 125.6 | 129.9 | 118.5 | 52.2, 114.7, 117.0, 156.3                               | Goljer et al. 1989 |
| IIIa         | 3-Cl  | $\text{CDCl}_3$   | 140.3 | 117.2 | 135.7 | 125.0 | 130.9 | 115.4 |   |                    |
|              | Calc. | $\text{CDCl}_3$   | 140.4 | 117.6 | 135.3 | 125.5 | 130.4 | 117.3 | 51.6, 51.7, 93.9, 151.7, 165.7, 169.2                   | This work          |
|              | Calc. | NL                | 140.0 | 117.4 | 135.4 | 125.3 | 130.0 | 117.2 |   |                    |
| IIIb         | 3-Cl  | $\text{CDCl}_3$   | 140.0 | 116.7 | 135.2 | 124.3 | 130.4 | 114.8 |   |                    |
|              | Calc. | $\text{CDCl}_3$   | 140.6 | 117.6 | 136.0 | 125.3 | 131.1 | 115.3 | 13.8, 13.9, 59.8, 60.1, 94.2, 150.8, 165.0, 168.4       | This work          |
|              | Calc. | NL                | 140.2 | 117.4 | 136.1 | 125.1 | 130.7 | 115.2 |   |                    |
| IIIc         | 3-Cl  | $\text{DMSO-}d_6$ | 140.7 | 118.0 | 134.1 | 124.8 | 131.1 | 117.3 |   |                    |
|              | Calc. | $\text{DMSO-}d_6$ | 141.0 | 118.2 | 134.2 | 125.4 | 131.3 | 116.9 | 27.6, 31.6, 113.0, 152.5, 195.5, 199.7                  | This work          |
|              | Calc. | NL                | 140.0 | 118.3 | 135.7 | 125.5 | 130.3 | 116.1 |   |                    |
| IIId         | 3-Cl  | $\text{DMSO-}d_6$ | 141.0 | 118.1 | 134.4 | 125.2 | 131.4 | 117.2 |   |                    |
|              | Calc. | $\text{DMSO-}d_6$ | 141.0 | 118.3 | 134.4 | 125.3 | 131.5 | 117.0 | 53.4, 114.3, 116.6, 156.4                               | This work          |
|              | Calc. | NL                | 140.0 | 118.4 | 135.9 | 125.4 | 130.5 | 116.2 |   |                    |
| IIIa         | 4-Br  | $\text{DMSO-}d_6$ | 139.4 | 120.4 | 132.7 | 117.1 | 132.7 | 120.4 |   |                    |
|              | Calc. | NL                | 138.3 | 119.8 | 132.9 | 119.3 | 132.9 | 119.8 | 51.6, 93.7, 151.6, 160.0                                | This work          |
|              | Calc. | NL                | 139.4 | 120.2 | 132.7 | 116.9 | 132.7 | 120.2 |   |                    |
| IIIb         | 4-Br  | $\text{DMSO-}d_6$ | 138.3 | 119.5 | 132.9 | 119.2 | 132.9 | 119.5 |   |                    |
|              | Calc. | $\text{CDCl}_3$   | 138.2 | 118.6 | 133.0 | 119.3 | 133.0 | 118.6 | 14.6, 14.7, 60.0, 60.2, 94.4, 151.2, 165.3, 167.6       | Lager et al. 2006  |
|              | Calc. | $\text{CDCl}_3$   | 136.9 | 118.9 | 132.7 | 119.5 | 132.7 | 118.9 |   |                    |
| IIIc         | 4-Br  | $\text{DMSO-}d_6$ | 137.5 | 119.5 | 132.8 | 119.9 | 132.8 | 119.5 |   |                    |
|              | Calc. | NL                | 138.8 | 120.1 | 132.2 | 117.2 | 132.2 | 120.1 | 27.4, 32.1, 113.7, 151.3, 194.8, 201.3                  | This work          |
|              | Calc. | NL                | 138.0 | 120.4 | 132.8 | 119.8 | 132.8 | 120.4 | 52.4, 114.1, 116.4, 155.8                               | This work          |
| IIIa         | 4-Me  | $\text{CDCl}_3$   | 136.7 | 117.3 | 130.3 | 135.0 | 130.3 | 117.3 |   |                    |
|              | Calc. | $\text{CDCl}_3$   | 136.0 | 117.1 | 129.7 | 134.3 | 129.7 | 117.1 | 20.8, 51.4, 51.5, 92.2, 152.5, 166.1, 169.4             | This work          |
|              | Calc. | NL                | 136.1 | 117.3 | 129.8 | 134.2 | 129.8 | 117.3 |   |                    |
| IIIb         | 4-Me  | $\text{CDCl}_3$   | 136.9 | 117.2 | 130.3 | 134.8 | 130.3 | 117.2 |   |                    |
|              | Calc. | $\text{CDCl}_3$   | 136.2 | 117.1 | 130.4 | 134.1 | 130.4 | 117.1 | 14.3, 14.5, 20.8, 60.0, 60.3, 92.9, 152.2, 165.9, 169.2 | Matta et al. 1996  |
|              | Calc. | NL                | 136.3 | 117.3 | 130.3 | 134.0 | 130.3 | 117.3 |   |                    |
| IIIc         | 4-Me  | $\text{CDCl}_3$   | 138.0 | 119.3 | 131.9 | 137.2 | 131.9 | 119.3 |   |                    |
|              | Calc. | $\text{CDCl}_3$   | 135.5 | 117.2 | 130.2 | 134.5 | 130.2 | 117.2 | 22.3, 28.8, 33.5, 114.4, 153.5, 196.2, 202.4            | This work          |
|              | Calc. | NL                | 135.6 | 117.4 | 130.1 | 134.4 | 130.1 | 117.4 |   |                    |

**Tab. 3 continue.**  $^{13}\text{C}$  NMR spectra of arylaminomethylidene derivatives III.

| Compound No. | Z         | Solvent         | C-1   | C-2   | C-3   | C-4   | C-5   | C-6   | Other signals   | Reference                 |
|--------------|-----------|-----------------|-------|-------|-------|-------|-------|-------|---|---------------------------|
| III d        | 4-Me      | DMSO- $d_6$     | 137.3 | 118.5 | 130.3 | 135.0 | 130.3 | 118.5 |   |                           |
|              | Calc.     | DMSO- $d_6$     | 136.0 | 118.1 | 130.1 | 134.2 | 130.1 | 118.1 | 20.8, 51.4, 114.8, 117.1, 156.1                         | This work                 |
|              | Calc.     | NL              | 136.1 | 118.3 | 130.1 | 134.3 | 130.1 | 118.3 |   |                           |
| III a        | 4-OMe     | $\text{CDCl}_3$ | 132.6 | 115.0 | 118.9 | 153.0 | 118.9 | 115.0 |   |                           |
|              | Calc.     | $\text{CDCl}_3$ | 131.3 | 118.2 | 114.6 | 156.5 | 114.6 | 118.2 | 51.4, 51.5, 55.6, 91.8, 157.3, 166.1, 169.5             | Zewge et al. 2007         |
|              | Calc.     | NL              | 130.9 | 118.1 | 114.3 | 155.3 | 114.3 | 118.1 |   |                           |
| III b        | 4-OMe     | $\text{CDCl}_3$ | 132.7 | 114.9 | 118.8 | 152.6 | 118.8 | 114.9 |   |                           |
|              | Calc.     | $\text{CDCl}_3$ | 131.5 | 118.2 | 115.3 | 156.6 | 115.3 | 118.2 | 14.3, 14.4, 55.5, 59.9, 60.2, 92.4, 157.1, 165.8, 169.2 | This work                 |
|              | Calc.     | NL              | 131.1 | 118.1 | 115.0 | 155.1 | 115.0 | 118.1 |   |                           |
| III c        | 4-OMe     | $\text{CDCl}_3$ | 133.8 | 120.9 | 116.5 | 159.2 | 116.5 | 120.9 |   |                           |
|              | Calc.     | $\text{CDCl}_3$ | 130.8 | 118.3 | 115.1 | 156.7 | 115.1 | 118.3 | 28.8, 33.4, 57.0, 114.2, 153.9, 196.1, 202.3            | This work                 |
|              | Calc.     | NL              | 130.4 | 118.2 | 114.8 | 155.5 | 114.8 | 118.2 |   |                           |
| III d        | 4-OMe     | DMSO- $d_6$     | 132.5 | 119.6 | 116.8 | 155.7 | 116.8 | 119.6 |   |                           |
|              | Calc.     | DMSO- $d_6$     | 131.1 | 119.4 | 115.1 | 156.2 | 115.1 | 119.4 | 50.3, 55.3, 114.5, 156.9                                | Mukhopadhyaya et al. 2000 |
|              | Calc.     | NL              | 130.9 | 119.1 | 114.8 | 155.4 | 114.8 | 119.1 |   |                           |
| III a        | 4-NO $_2$ | $\text{CDCl}_3$ | 144.3 | 116.5 | 126.0 | 144.0 | 126.0 | 116.5 |   |                           |
|              | Calc.     | $\text{CDCl}_3$ | 145.1 | 118.1 | 124.1 | 145.0 | 124.1 | 118.1 | 51.9, 52.0, 96.5, 150.2, 165.2, 168.9                   | This work                 |
|              | Calc.     | NL              | 144.8 | 118.1 | 123.8 | 144.7 | 123.8 | 118.1 |   |                           |
| III b        | 4-NO $_2$ | $\text{CDCl}_3$ | 144.5 | 116.3 | 126.0 | 143.8 | 126.0 | 116.3 |   |                           |
|              | Calc.     | $\text{CDCl}_3$ | 145.3 | 118.1 | 124.8 | 144.8 | 124.8 | 118.1 | 14.2, 14.3, 60.6, 61.0, 97.3, 149.7 165.0, 168.5        | Kim et al. 2010           |
|              | Calc.     | NL              | 145.4 | 118.1 | 124.5 | 144.5 | 124.5 | 118.1 |   |                           |

$\text{CDCl}_3$  – deuteriochloroform, DMSO- $d_6$  – hexadeuterodimethylsulfoxide, NL – neat liquid data from Ewing 1979.

bons, but reversal sign. It is not so strong effect like the amino group, but slightly similar to aminoacetyl group, thus similar to aminogroup substituted with polarized double bond. The obtained data are in good agreement with calculated ones within small range of deviations what is confirming the accuracy of the used method. On the other hand, meta- and para-substitution does not twisting the aminoethylidene substituent off of the plane of the benzene ring. In Table 3 are presented  $^{13}\text{C}$  NMR data of the products III obtained from nucleophilic vinylic substitution of substituted anilines I and activated enolethers II.

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