

Condensation of (E)-5-Aryl-3-Arylmethylene-2-(3H)-Furanones with Different Amines

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Condensation of (E)-2(3H)-furanones (**1–8**) with different amines gives the corresponding (E)-N-substituted-3-aryl-2-arylmethylene propionamides and 5-substituted amino-5-aryl-3-arylmethylene butyrolactones (**9–36**). The structures of compounds obtained were assigned in accordance with their elemental analysis, ¹H NMR, MS and FTIR.

Key Words: Condensation, (E)-5-Aryl-3-Arylmethylene-2-(3H)-furanones, Amines.

INTRODUCTINO

γ -Butyrolactones (GBL) are used exclusively as chemical intermediates in the production of vitamins and pharmaceuticals as in the production of pesticides and herbicides^{1, 2}. So the aim of our work was to synthesize the derivatives of these important biological compounds.

Condensation of (E)-2(3H)-furanones (**1–8**) with *n*-butylamine gives the corresponding (E)-N-(*n*-butyl)3-aryl-2-arylmethylene propionamides (**9–16**), whereas the condensation of (E)-2(3H)-furanones (**2–4, 6–8**) with methylamine yields mixtures of the corresponding (E)-N-methyl-3-aryl-2-arylmethylene propionamides and 5-methyl-amino-5-aryl-3-arylmethylene-butyrolactones [**17–22 (a, b)**] respectively. The condensation of the furanones with aniline and *p*-toluidine except furanones (**7**) yields mixtures of the corresponding (E)-N-substituted-3-aryl-2-arylmethylene propionamides and 5-substituted amino-5-aryl-3-arylmethylene-butyrolactones [**23–36(a,b)**] respectively. Many trials for the condensation of furanones (**7**) with aniline and *p*-toluidine under different conditions were unsuccessful. The results obtained are ascribed to the electronic structure of the furanone and to the steric effect exerted by the alkyl group of the amine. A mechanism is proposed in which propionamides are formed (**9–16, 17a–36a**). Propionamides (**17a–36a**) undergo thermal transformation to form the corresponding butyrolactones (**17b–36b**).

EXPERIMENTAL

Condensation of (E)-2(3H)-Furanones with Amines

General Procedure: A mixture of (E)-2(3H)-furanones (**1–8**)^{3–8} (1 mmol) and amine (2 mmol) in 40 mL of the suitable solvent was refluxed for 4 h. After cooling the reaction mixtures, working out and crystallization from the appropriate solvent, the products (**9–36**) were obtained. Accomplishment of the reaction was assured by monitoring the products using TLC technique. The structures of the compounds (**9–36**) are confirmed by their elemental analysis (Table-1), ¹H NMR, MS and FTIR.

TABLE-1
ELEMENTAL ANALYSIS FOR COMPOUNDS 9-36

Compound	m.f. (Colour)	m.p.* (°C)	Yield (%)	Found (Calcd.)		
				C	H	N
9	C ₂₁ H ₂₃ NO ₂ (White)	171	66	78.39 (78.40)	7.09 (7.16)	4.30 (4.36)
10	C ₂₂ H ₂₅ NO ₂ (White)	150	71	77.95 (78.70)	7.39 (7.45)	4.14 (4.17)
11	C ₂₂ H ₂₅ NO ₃ (Yellow)	142	49	75.08 (75.11)	7.05 (7.11)	3.94 (3.98)
12	C ₂₁ H ₂₂ NO ₂ Cl (White)	146	92	70.76 (70.82)	6.09 (6.18)	3.90 (3.93)
13	C ₂₂ H ₂₅ NO ₃ (White)	176	49	74.98 (75.11)	6.90 (7.11)	3.94 (3.98)
14	C ₂₃ H ₂₇ NO ₃ (Yellow)	121	63	75.50 (75.53)	7.34 (7.93)	3.80 (3.83)
15	C ₂₃ H ₂₇ NO ₄ (Pale yellow)	120	51	72.29 (72.36)	7.03 (7.08)	3.61 (3.67)
16	C ₂₂ H ₂₄ NO ₃ Cl (Pale brown)	151	53	68.37 (68.41)	6.19 (6.22)	3.58 (3.63)
17 (a, b)	C ₁₉ H ₁₉ NO ₂ (Pale brown)	129-35	86	77.69 (77.74)	6.42 (6.48)	4.71 (4.77)
18 (a, b)	C ₂₂ H ₂₅ NO ₃ (Brown)	105-10	54	73.66 (73.71)	6.09 (6.14)	4.50 (4.53)
19 (a, b)	C ₁₈ H ₁₆ NO ₂ Cl (Brown)	105-10	90	68.85 (68.89)	5.40 (5.11)	4.41 (4.47)
20 (a, b)	C ₂₀ H ₂₁ NO ₃ (Pale brown)	158-63	78	74.15 (74.23)	6.40 (6.50)	4.29 (4.33)
21 (a, b)	C ₂₀ H ₂₁ NO ₄ (Brown)	112-18	85	70.71 (70.73)	6.15 (6.19)	4.10 (4.15)
22 (a, b)	C ₁₉ H ₁₈ NO ₂ Cl (Brown)	139-44	92	66.11 (66.32)	5.10 (5.24)	4.11 (4.08)
23 (a, b)	C ₂₃ H ₁₉ NO ₂ (Pale yellow)	139-45	59	80.78 (80.84)	5.48 (5.56)	4.08 (4.10)
24 (a, b)	C ₂₄ H ₂₁ NO ₂ (Pale brown)	110-16	70	81.00 (81.03)	5.92 (5.91)	3.90 (3.93)
25 (a, b)	C ₂₄ H ₂₁ NO ₃ (Brown)	110-15	64	77.49 (77.54)	5.60 (5.64)	3.71 (3.76)
26 (a, b)	C ₂₃ H ₁₈ NO ₂ Cl (Brown)	125-30	57	73.39 (73.44)	4.80 (4.79)	3.68 (3.73)
27 (a, b)	C ₂₄ H ₂₁ NO ₃ (Yellow)	162-68	54	77.49 (77.54)	5.60 (5.65)	3.71 (3.76)
28 (a, b)	C ₂₅ H ₂₃ NO ₃ (Pale brown)	150-55	66	77.79 (77.84)	5.91 (5.97)	3.60 (3.63)

Compound	m.f. (Colour)	m.p.* (°C)	Yield (%)	Found (Calcd.)		
				C	H	N
29 (a, b)	$C_{24}H_{20}NO_3Cl$ (Brown)	106–11	77	70.90 (70.95)	4.88 (4.88)	3.41 (3.45)
30 (a, b)	$C_{24}H_{21}NO_2$ (Yellow)	178–83	56	80.91 (81.02)	5.86 (5.91)	3.90 (3.94)
31 (a, b)	$C_{25}H_{23}NO_2$ (Brown)	80–85	80	81.18 (81.20)	6.19 (6.23)	3.74 (3.79)
32 (a, b)	$C_{25}H_{23}NO_3$ (Brown)	105–10	75	77.82 (77.80)	5.91 (5.97)	3.60 (3.63)
33 (a, b)	$C_{24}H_{20}NO_2Cl$ (Pale brown)	138–42	69	73.50 (73.87)	5.07 (5.13)	3.51 (3.59)
34 (a, b)	$C_{25}H_{23}NO_3$ (Brown)	160–65	77	77.72 (77.80)	5.91 (5.97)	3.60 (3.63)
35 (a, b)	$C_{26}H_{25}NO_3$ (Orange)	130–35	56	77.95 (78.11)	6.15 (6.26)	3.45 (3.50)
36 (a, b)	$C_{25}H_{22}NO_3Cl$ (Pale brown)	130–35	76	71.50 (71.45)	5.46 (5.24)	3.30 (3.33)

*From benzene-petroleum ether (b.p. 40–60).

RESULTS AND DISCUSSION

Condensation of (E)-2(3H)-Furanones (1–8) with Aliphatic Amines

(a) Condensation of the furanones (1–8) with *n*-butylamine

N-(2-Butyl)-3-benzoyl-2-phenylmethylene propionamide (9): FTIR (KBr): 3346 ν (NH), 1668 ν (benzoyl, C=O) and 1638 cm^{-1} ν (amide, C=O). 1H NMR (DMSO-d₆): δ = 7.30–7.38 (5H, m; H-1), 7.28 (1H, s; H-2), 3.3 (2H, d; H-3), 7.40–7.53, (5H, m; H-4), 6.702 (1H, s; H-5), 2.70–2.90 (2H, m; H-6), 1.20–1.40 (2H, m; H-7), 1.10–1.20 (2H, six; H-8) and 0.756 (3H, t; H-9). MS: m/z = 321 (M^+ , 22%, $C_{21}H_{23}NO_2$), 303 (22, $C_{21}H_{21}NO$), 261 (9, $C_{18}H_{15}NO$), 249 (26, $C_{17}H_{15}NO$), 202 (18, $C_{13}H_{16}NO$), 116 (100, C_9H_8), and 105 (17, C_7H_5O).

N-(n-Butyl)-3-(4-methylbenzoyl)-2-phenylmethylene propionamide (10): FTIR (KBr): 3257 ν (NH), 1680 ν (aroyl, C=O) and 1643 cm^{-1} ν (amide, C=O). 1H NMR (DMSO-d₆): δ = 7.24–7.40 [(5H, m; H-1) and (1H, imp; H-2)], 3.27–3.35 (2H, d; H-3), 7.50–7.52, (2H, d; H-4), 7.16–7.18 (2H, d; H-5), 2.28 (3H, s; H-6), 6.63 (1H, s; H-7), 2.70–2.85 (2H, m; H-8), 1.20–1.52 (2H, m; H-9), 1.14 (2H, six; H-10) and 0.75–0.79 (3H, t; H-11). MS: m/z = 335 (M^+ , 9%, $C_{22}H_{25}NO_2$), 317 (6, $C_{22}H_{23}NO$), 275 (3, $C_{19}H_{17}NO$), 262 (2.5, $C_{18}H_{13}O_2$), 119 (100, C_8H_7O), 116 (19, C_9H_8) and 91 (24, C_7H_7).

N-(n-Butyl)-3-(4-methoxybenzoyl)-2-phenylmethylene propionamide (11): FTIR (KBr): 3327 ν (NH), 1668 ν (aroyl, C=O) and 1638 cm^{-1} ν (amide, C=O). 1H NMR (DMSO-d₆): δ = 7.27–7.36 [(5H, m; H-1), 7.36–7.38 (1H, imp; H-2), 3.28–3.33, (2H, d; H-3), 7.50–7.53 (2H, d; H-4), 6.90–6.93 (2H, d; H-5), 3.74 (3H, s; H-6), 6.60 (1H, s; H-7), 2.70–2.90 (2H, m; H-8), 1.20–1.40 (2H, m;

H-9), 1.14–1.17 (2H, 6; H-10) and 0.75–0.80 (3H, t; H-11). MS: m/z = 351 (M⁺, 8.6%, C₂₂H₂₅NO₃), 333 (10, C₂₂H₂₃NO₂), 278 (4.5, C₁₈H₁₄O₃), 202 (2, C₁₃H₁₀NO), 135 (100, C₈H₇O₂), 116 (10, C₉H₈) and 107 (6, C₇H₇O).

N-(n-Butyl)-3-(4-chlorobenzoyl)-2-phenylmethylene propionamide (12): FTIR (KBr): 3266 ν(NH), 1681 ν(aryl, C=O) and 1645 cm⁻¹ ν(amide, C=O). ¹H NMR (DMSO-d₆): δ = 7.28–7.36 (5H, m; H-1), 7.40–7.42 (1H, s; H-2), 3.26–3.40 (2H, imp; H-3), 7.51–7.53 (2H, d; H-4), 7.80–7.85 (2H, d; H-5), 7.25 (1H, s; H-6), 2.75–2.85 (2H, m; H-7), 1.20–1.45 (2H, 6; H-9) and 0.75–0.79 (3H, t; H-10), MS: m/z = 355.8 (M⁺, 79%, C₂₁H₂₂NO₂Cl), 337 (51, C₂₁H₂₀NOCl), 295 (28, C₁₈H₁₄NOCl), 282 (26, C₁₇H₁₁O₂Cl), 202 (25, C₁₃H₁₆NO), 153 (5.6, C₈H₆OCl) and 116 (100, C₉H₈).

N-(n-Butyl)-3-benzoyl-2-(4-methoxyphenylmethylen)propionamide (13): FTIR (KBr): 3284 ν(NH), 1674 ν(benzoyl, C=O) and 1636 cm⁻¹ ν(amide, C=O). ¹H NMR (DMSO-d₆): δ = 3.77 (3H, s; H-1), 6.95–6.98 (2H, d; H-2), 7.46–7.49, (2H, d; H-3), 7.23 (1H, s; H-4), 3.33 (2H, d; H-5), 7.37–7.38 (5H, m; H-6), 6.63 (1H, s; H-7), 2.70–2.80 (2H, m; H-8), 1.20–1.40 (2H, m; H-9), 1.13–1.15 (2H, 6; H-10) and 0.73–0.78 (3H, t; H-11). MS: m/z = 351.4 (M⁺, 24%, C₂₂H₂₅NO₃), 333 (27, C₂₂H₂₃NO₂), 291 (6.5, C₁₉H₁₇NO₂), 277 (5.3, C₁₈H₁₅NO₂), 232 (2, C₁₄H₁₈NO₂), 146 (100, C₁₀H₁₀O), 119 (3, C₈H₇O) and 105 (82, C₇H₅O).

N-(n-Butyl)-3-(4-methylbenzoyl)-2-(4-methoxyphenylmethylen) propionamide (14): FTIR (KBr): 3292 ν(NH), 1677 ν(aryl, C=O) and 1642 cm⁻¹ ν(amide, C=O). ¹H NMR (DMSO-d₆): δ = 3.77 (3H, s; H-1), 6.94–6.97 (2H, d; H-2), 7.24–7.27, (2H, d; H-3), 7.22 (1H, s; H-4), 3.29–3.31 (2H, d; H-5), 7.44–7.47 (2H, d; H-6), 7.15–7.18 (2H, d; H-7), 2.28 (3H, s; H-8), 6.58 (1H, s; H-9), 2.70–2.90 (2H, m; H-10) 1.20–1.40 (2H, m; H-11), 1.15–1.20 (2H, 6; H-12) and 0.74–0.79 (3H, t; H-13). MS: m/z = 365.4 (M⁺, 7%, C₂₃H₂₇NO₃), 347 (17, C₂₃H₂₅NO₂), 305 (3.4, C₂₀H₁₉NO₂), 292 (5, C₁₉H₁₆O₃), 232 (2.2, C₁₄H₁₈NO₂), 146 (22, C₁₀H₁₀O) 119 (100, C₈H₇O) and 91 (18, C₇H₇).

N-(n-Butyl)-3-(4-methoxybenzoyl)-2-(4-methoxyphenylmethylen)propionamide (15): FTIR (KBr): 3450 ν(NH), 1685 ν(aryl, C=O) and 1640 cm⁻¹ ν(amide, C=O). ¹H NMR (DMSO-d₆): δ = 7.75 (3H, s; H-1), 6.91–6.94 (2H, d; H-2)], 7.29–7.31, (2H, d; H-3), 7.22 (1H, s; H-4), 3.30–3.35 (2H, imp; H-5), 7.46–7.48 (2H, d; H-6), 6.96–6.99 (2H, d; H-7), 3.783 (3H, s; H-8), 6.85 (1H, imp; H-9), 2.70–2.90 (2H, m; H-10) 1.20–1.40 (2H, m; H-11), 1.12–1.17 (2H, 6; H-12) and 0.76–0.80 (3H, t; H-13). MS: m/z = 381 (M⁺, 0%, C₂₃H₂₇NO₄), 363 (11, C₂₃H₂₅NO₃), 202 (2.3, C₁₃H₁₆NO), 149 (20, C₉H₉O₂), 135 (58, C₈H₇O₂), 107 (12, C₇H₇O) and 55 (100, C₃H₅O).

N-(n-Butyl)-3-(4-chlorobenzoyl)-2-(4-methoxyphenylmethylen) propionamide (16): FTIR (KBr): 3270 ν(NH), 1678 ν(aryl, C=O) and 1640 cm⁻¹ ν(amide, C=O). ¹H NMR (DMSO-d₆): δ = 3.773 (3H, s; H-1), 6.93–6.98 (2H, d; H-2), 7.40–7.42, [(2H, d; H-3) and (2H, d; H-6)], 7.23 (1H, s; H-4), 3.27–3.32 (2H, d; H-5), 7.46–7.49 (2H, d; H-7), 6.78 (1H, d; H-8), 2.70–2.90 (2H, m; H-9), 1.20–1.40 (2H, m; H-10), 1.10–1.20 (2H, 6; H-11) and 0.74–0.79 (3H, t; H-12), MS: m/z = 386 (M⁺, 24%, C₂₂H₂₄NO₃Cl), 367 (100, C₂₂H₂₂NO₂Cl), 325 (17,

$C_{19}H_{16}NO_2Cl$), 312 (25, $C_{18}H_{13}O_3Cl$), 202 (10, $C_{13}H_{16}NO$), 154 (3.4, C_8H_7OCl) and 146 (90, $C_{10}H_{10}O$).

(b) *Condensation of (E)-2(3H)-furanones (2-4 and 6-8) with methylamine*

N-Methyl-3-(4-methylbenzoyl)-2-phenylmethylene propionamide (17a) and 5-methylamino-5-(4-methylphenyl)-3-phenylmethylene butyrolactone (17b): FTIR (KBr): 3400–3250 ν (NH), 1750 ν (butyrolactone, C=O), 1682 cm^{-1} ν (aroyl, C=O) and 1650 cm^{-1} ν (amide, C=O). ^1H NMR (DMSO-d₆): δ = 7.19–7.61 (5H, m; H-1a and H-1b), 7.26–7.28 (1H, s; H-2a and H-2b), 3.40 (2H, imp; H-3a and H-3b), 7.71–7.83 (2H, d; H-4a and H-4b), 6.88–7.18 (2H, d; H-5a and H-5b), 2.29–2.34 (3H, s; H-6a and H-6b), 6.43 (1H, s; H-7a), 7.2–7.8 (1H, imp; H-7b) and 2.78–2.80 (3H, s; H-8a and H-8b). MS: m/z = 293 (M^+ , 4.5%, $C_{19}H_{19}NO_2$), 275 (46, $C_{19}H_{17}NO$), 203 (8, $C_{12}H_{13}NO_2$), 133 (15, C_9H_9O), 126 (7, $C_6H_8NO_2$), 119 (97, C_8H_7O), 116 (13, C_9H_8) and 91 (100 C_7H_7).

N-Methyl-3-(4-methoxybenzoyl)-2-phenylmethylene propionamide (18a) and 5-methylamino-5-(4-methoxyphenyl)-3-phenylmethylene butyrolactone (18b): FTIR (KBr): 3300–3180 ν (NH), 1750 ν (butyrolactone, C=O), 1698 ν (aroyl, C=O) and 1650 cm^{-1} ν (amide, C=O). ^1H NMR (DMSO-d₆): δ = 7.24–7.42 (5H, m; H-1a and H-1b), 7.24–7.35 (1H, s; H-2a and H-2b), 3.22–3.23 (2H, s; H-3a and H-3b), 7.48–7.60 (2H, d; H-4a and H-4b), 6.94–6.97 (2H, d; H-5a and H-5b), 3.77 (3H, s; H-6a and H-6b), 6.66 (1H, s; H-7a), 7.24–7.60 (1H, imp; H-7b) and 2.58–2.59 (3H, s; H-8a and H-8b). MS: m/z = 309 (M^+ , 9%, $C_{19}H_{19}NO_3$), 292 (13, $C_{19}H_{18}NO_2$), 202 (11, $C_{12}H_{12}NO_2$), 149 (11, $C_9H_9O_2$), 135 (40, $C_8H_7O_2$), 119 (13, C_8H_7O), 115 (31, C_9H_7) and 107 (12 C_7H_7O).

N-Methyl-3-(4-chlorobenzoyl)-2-phenylmethylene propionamide (19a) and 5-methylamino-5-(4-chlorophenyl)-3-phenylmethylene butyrolactone (19b): FTIR (KBr): 3370–3250 ν (broad NH), 1750 ν (butyrolactone, C=O), 1697 ν (aroyl, C=O) and 1650 cm^{-1} ν (amide, C=O). ^1H NMR (DMSO-d₆): δ = 7.40–7.69 (5H, m; H-1a and H-1b), 7.54 (1H, s; H-2a and H-2b), 3.40 (2H, imp; H-3a and H-3b), 7.45–7.51 (2H, d; H-4a and H-4b), 7.95–7.99 (2H, d; H-5a and H-5b), 6.56 (1H, broad; H-6a), 7.95 (1H, broad; H-6b) and 2.80 (3H, s; H-7a and H-7b). MS: m/z = 313 (M^+ , 6%, $C_{18}H_{16}NO_2Cl$), 296 (50, $C_{18}H_{15}NO$), 284 (82, $C_{17}H_{13}O_2Cl$), 255 (39, $C_{16}H_{12}OCl$), 236 (26, $C_{12}H_{11}NO_2Cl$), 173 (37, $C_{11}H_9O_2$), 139 (100, C_7H_4OCl) and 116 (6, C_9H_8).

N-Methyl-3-(4-methylbenzoyl)-2-(4-methoxyphenylmethylene) propionamide (20a) and 5-methylamino-5-(4-methylphenyl)-3-(4-methoxyphenylmethylene) butyrolactone (20b): FTIR (KBr): 3313 ν (NH), 1750 ν (butyrolactone, C=O), 1680 ν (aroyl, C=O) and 1643 cm^{-1} ν (amide, C=O). ^1H NMR (DMSO-d₆): δ = 3.75–3.77 (3H, s; H-1a and H-1b), 6.94–6.97 (2H, d; H-2a and H-2b), 7.31–7.34 (2H, d; H-3a and H-3b), 7.10–7.22 (1H, s; H-4a and H-4b), 3.30 (2H, imp; H-5a and H-5b), 7.44–7.47 (2H, d; H-6a and H-6b), 7.71–7.20 (2H, d; H-7a and H-7b), 2.29 (3H, s; H-8), 6.60 (1H, s; H-9a), 7.15–7.25 (1H, imp; H-9b) and 2.57 (3H, s; H-10a and H-10b). MS: m/z = 323 (M^+ , 2.6%, $C_{20}H_{21}NO_3$), 305 (100, $C_{20}H_{19}NO_2$), 292 (3.3, $C_{19}H_{16}O_3$), 290 (40, $C_{19}H_{16}NO_2$), 233 (2, $C_{13}H_{15}NO_2$), 145 (10, $C_{10}H_9O$), 119 (22, C_8H_7O) and 107 (1.1, C_7H_7O).

N-Methyl-3-(4-methylbenzoyl)-2-(4-methoxyphenylmethylenepropionamide (21a) and 5-methylamino-5-(4-methylphenyl)-3-(4-methoxyphenylmethylenecbutyrolactone (21b): FTIR (KBr): 3293 ν (NH), 1750 ν (butyrolactone, C=O), 1680 ν (aroyl, C=O) and 1640 cm^{-1} ν (amide, C=O). ^1H NMR (DMSO-d₆): δ = 3.76–3.84 (3H, s; H-1a and H-1b), 6.90–7.20 (2H, d; H-2a and H-2b), 7.20–7.40 [2H, d; H-3a and H-3b] and (1H, s; H-4a and H-4b)], 3.25–3.36 (2H, s; H-5a and H-5b), 7.50–7.70 (2H, d; H-6a and H-6b), 6.90–7.20 (2H, d; H-7a and H-7b), 3.79–3.88 (1H, s; H-8a and H-8b), 6.60 (1H, s; H-9a), 7.80 (1H, s; H-9b) and 2.59 (3H, s; H-10a and H-10b). MS: m/z = 339 (M⁺, 0%, C₂₀H₂₁NO₄), 321 (100, C₂₀H₁₉NO₃), 307 (47, C₁₉H₁₉NO₃), 290 (8, C₁₉H₁₇NO₂), 233 (30, C₁₃H₁₅O₃), 184 (12, C₁₂H₁₀NO), 149 (16, C₉H₉O₂) and 107 (6 C₇H₇O).

N-Methyl-3-(4-chlorobenzoyl)-2-(4-methoxyphenylmethylenepropionamide (22a) and 5-methylamino-5-(4-chlorophenyl)-3-(4-methoxyphenylmethylenecbutyrolactone (22b): FTIR (KBr): 3400–3200 ν (broad NH), 1753 ν (butyrolactone, C=O), 1680 ν (aroyl, C=O) and 1640 cm^{-1} ν (amide, C=O). ^1H NMR (DMSO-d₆): δ = 3.73–3.82 (3H, s; H-1a and H-1b), 6.95–7.03 (3H, d; H-2a and H-2b), 7.43–7.51 (2H, d; H-3a and H-3b), 7.54–7.58 (1H, imp; H-4a and H-4b), 3.11 (2H, s; H-5a and H-5b), 7.67–7.70 (2H, d; H-6a and H-6b), 7.77–7.91 (2H, d; H-7a and H-7b), 6.55 (1H, s; H-8a), 7.30 (1H, s; H-8b), and 2.80 (3H, s; H-9b and H-9b). MS: m/z = 343.8 (M⁺, 0%, C₁₉H₁₈O₃NCl), 231 (1.4, C₁₃H₁₃NO₃), 190 (1.1, C₁₁H₁₂NO₂), 153 (1.6, C₈H₆OCl), 145 (1.7, C₁₀H₉O), 139 (7, C₇H₄OCl), 111 (10, C₆H₄Cl) and 69 (100, C₃H₃NO).

Condensation of (E)-2(3H)-Furanones (1–6, 8) with Aromatic Amines

(a) Condensation of (E)-2(3H) furanones (1–6, 8) with aniline

N-Phenyl-3-benzoyl-2-phenylmethylenepropionamide (23a) and 5-phenylamino-5-phenyl-3-phenylmethylenecbutyrolactone (23b): FTIR (KBr): 3340–3320 ν (NH), 1768 ν (butyrolactone, C=O), 1691 ν (benzoyl, C=O) and 1649 cm^{-1} ν (amide, C=O). ^1H NMR (DMSO-d₆): δ = 6.97–7.19 (5H, m; H-1a and H-1b), 7.49 (1H, imp; H-2a and H-2b), 3.46–3.47 (2H, s; H-3a and H-3b), 7.22–7.36 (5H, m; H-4a and H-4b), 6.82 (1H, s; H-5a), 7.79 (1H, imp; H-5b), and 7.36–7.60 (5H, m; H-6a and H-6b). MS: m/z = 341 (M⁺, 3.4%, C₂₃H₁₉NO₂), 264 (22, C₁₇H₁₄NO₂), 249 (29, C₁₇H₁₃O₂), 249 (21, C₁₇H₁₂O₂), 115 (17, C₉H₇), 105 (100, C₇H₅O) and 102 (48, C₈H₆).

N-Phenyl-3-(4-methylbenzoyl)-2-phenylmethylenepropionamide (24a) and 5-phenylamino-5-(4-methylphenyl)-3-phenylmethylenecbutyrolactone (24b): FTIR (KBr): 3296 ν (NH), 1765 ν (butyrolactone, C=O), 1681 ν (aroyl, C=O) and 1644 cm^{-1} ν (amide, C=O). ^1H NMR (DMSO-d₆): δ = 7.17–7.28 (5H, m; H-1a and H-1b), 7.32–7.34 (1H, s; H-2a and H-2b), 3.3–3.5 (2H, imp; H-3a and H-3b), 7.56–7.60 (2H, d; H-4a and H-4b), 7.49–7.72 (2H, d; H-5a and H-5b), 2.22–2.27 (3H, s; H-6a and H-6b), 6.77 (1H, s; H-7a), 7.6–8.0 (1H, imp; H-7b) and 7.37–7.50 (5H, m; H-8a and H-8b). MS: m/z = 355 (M⁺, 5%, C₂₄H₂₁NO₂), 263 (5, C₁₇H₁₃NO₂), 235 (5, C₁₇H₁₅O), 222 (2.3, C₁₅H₁₂NO), 120 (6, C₇H₆NO), 119 (18, C₈H₇O), 93 (100, C₆H₇N) and 91 (21, C₇H₇).

N-Phenyl-3-(4-methylbenzoyl)-2-phenylmethylenepropionamide (25a)

and 5-phenylamino-5-(4-methylphenyl)-3-phenylmethylene-butyrolactone (25b): FTIR (KBr): 3293 v(NH), 1757 v(butyrolactone, C=O), 1676 v(aryl, C=O) and 1641 cm⁻¹ v(amide, C=O). ¹H NMR (DMSO-d₆): δ = 7.02–7.18 (5H, m; H-1a and H-1b), 7.23–7.27 (1H, t; H-2a and H-2b), 3.46 (2H, s; H-3a and H-3b), 7.59–7.62 [(2H, d; H-4a and H-4b) and (1H, imp; H-7b)], 6.80–6.85 [(2H, d; H-5a and H-5b) and (1H, s; H-7a)], 3.70–3.76 (3H, s; H-6a and H-6b) and 7.36–7.48 (5H, m; H-8a and H-8b). MS: m/z = 371 (M⁺, 23%, C₂₄H₂₁NO₃), 278 (31, C₁₈H₁₄O₃), 263 (31, C₁₇H₁₃NO₂), 222 (15, C₁₅H₁₂NO), 119 (32, C₈H₇O₂), 105 (10, C₇H₅O) and 76 (100, C₆H₄).

N-Phenyl-3-(4-chlorobenzoyl)-2-phenylmethylene propionamide (26a) and 5-phenylamino-5-(4-chlorophenyl)-3-phenylmethylene-butyrolactone (26b): FTIR (KBr): 3350–3200 v(NH), 1760 v(butyrolactone, C=O), 1680 v(aryl, C=O) and 1640 cm⁻¹ v(amide, C=O). ¹H NMR (DMSO-d₆): δ = 7.25–7.40 (5H, m; H-1a and H-1b), 7.47–7.61 (1H, imp; H-2a and H-2b), 3.4–3.45 (2H, imp; H-3a and H-3b), 7.71–7.74 (2H, d; H-4a and H-4b), 7.91–7.94 (2H, d, H-5a and H-5b), 6.88 (1H, broad; H-6a), 7.91–7.94 (1H, imp; H-6b) and 7.44–7.61 (5H, m; H-7a and H-7b). MS: m/z = 375 (M⁺, 0%, C₂₃H₁₈NO₂Cl), 357 (7, C₂₃H₁₆NOCl), 283 (36, C₁₇H₁₂O₂Cl), 264 (2, C₁₇H₁₄NO₂), 139 (100, C₇H₄OCl), 115 (9, C₉H₇) and 111 (42, C₆H₄Cl).

N-Phenyl-3-benzoyl-2-(4-methoxyphenylmethylene) propionamide (27a) and 5-phenylamino-5-phenyl-3-(4-methoxyphenylmethylene)-butyrolactone (27b): FTIR (KBr): 3350–3200 v(broad NH), 1768 v(butyrolactone, C=O), 1691 v(benzoyl, C=O) and 1625 cm⁻¹ v(amide, C=O). ¹H NMR (DMSO-d₆): δ = 3.84–3.86 (3H, s; H-1a and H-1b), 6.98–7.13 (2H, d; H-2a and H-2b), 7.47–7.583 (2H, d; H-3a and H-3b), 7.27–7.29 (1H, s; H-4a and H-4b), 3.32–3.43 (2H, imp; H-5a and H-5b), 7.34–7.42 (5H, m; H-6a and H-6b), 6.80 (1H, s; H-7a), 7.62 (1H, s; H-7b) and 7.85–7.97 (5H, m; H-8a and H-8b). MS: m/z = 371.4 (M⁺, 0%, C₂₄H₂₁NO₃), 293 (3, C₁₈H₁₅NO₃), 279 (3, C₁₈H₁₅O₃), 278 (41, C₁₈H₁₄O₃), 145 (7, C₁₀H₉O), 107 (4, C₇H₇O), 105 (33, C₇H₆O), 93 (13, C₆H₇N) and 77 (100, C₆H₅).

N-Phenyl-3-(4-methylbenzoyl)-2-(4-methoxyphenylmethylene) propionamide (28a) and 5-phenylamino-5-(4-methylphenyl)-3-(4-methoxyphenylmethylene)-butyrolactone (28b): FTIR (KBr): 3350–3250 v(broad NH), 1768 v(butyrolactone, C=O), 1691 v(aryl C=O) and 1625 cm⁻¹ v(amide, C=O). ¹H NMR (DMSO-d₆): δ = 3.81–3.83 (3H, s; H-1a and H-1b), 6.51–6.75 (2H, d; H-2a and H-2b), 7.36–7.41 (2H, d; H-3a and H-3b), 7.20–7.29 (1H, s; H-4a and H-4b), 3.32–3.45 (2H, imp; H-5a and H-5b), 7.84–7.90 (2H, d; H-6a and H-6b), 7.32–7.36 (2H, d; H-7a and H-7b), 2.26–2.37 (3H, s; H-8a and H-8b), 6.71 (1H, s; H-9a), 7.84–7.86 (1H, imp; H-9b), 7.70–7.80 (5H, m; H-10a and H-10b). MS: m/z = 385.4 (M⁺, 7%, C₂₅H₂₃NO₃), 292 (7, C₁₉H₁₆O₃), 263 (6, C₁₇H₁₃NO₂), 145 (8, C₁₀H₉O), 133 (8, C₉H₉O), 119 (21, C₈H₇O), 107 (12, C₇H₇O), 91 (46, C₇H₇) and 65 (100, C₅H₅).

N-Phenyl-3-(4-chlorobenzoyl)-2-(4-methoxyphenylmethylene) propionamide (29a) and 5-phenylamino-5-(4-chlorophenyl)-3-(4-methoxyphenylmethylene)-butyrolactone (29b): FTIR (KBr): 3450–3300 v(broad NH), 1770 v(butyrolactone, C=O), 1680 v(aryl, C=O) and 1640 cm⁻¹ v(amide, C=O).

¹H NMR (DMSO-d₆): δ = 3.82–3.85 (3H, s; H-1a and H-1b), 6.97–7.13 (2H, d; H-2a and H-2b), 7.42–7.50 (2H, d; H-3a and H-3b), 7.33–7.39 (1H, s; H-4a and H-4b), 3.30–3.40 (2H, imp; H-5a and H-5b), 7.55–7.72 (2H, d; H-6a and H-6b), 7.93–8.10 (2H, d; H-7a and H-7b), 6.86 (1H, s; H-8a), 7.93–8.00 (1H, imp; H-8b) and 7.72–7.90 (5H, m; H-9a and H-9b). MS: m/z = 406 (M⁺, 8%, C₂₄H₂₀NO₃Cl), 375 (8, C₂₃H₁₈NO₂Cl), 282 (6, C₁₇H₁₂O₂Cl), 253 (7, C₁₆H₁₄NO₂), 153 (9, C₈H₆OCl), 145 (9, C₁₀H₉O), 139 (20, C₇H₄OCl), 111 (29, C₆H₅Cl) and 65 (100, C₅H₅).

(b) Condensation of (*E*)-2(3*H*)-furanones (1–6, 8) with *p*-toluidine

N-(4-Methylphenyl)-3-benzoyl-2-phenylmethylene propionamide (30a) and 5-(4-methylphenylamino)-5-phenyl-3-phenylmethylene butyrolactone (30b): FTIR (KBr): 3300 v(NH), 1760 v(butyrolactone, C=O), 1680 v(benzoyl, C=O) and 1640 cm⁻¹ v(amide, C=O). ¹H NMR (DMSO-d₆): δ = 7.17–7.27 (5H, m; H-1a and H-1b), 7.25–7.30 (1H, s; H-2a and H-2b), 3.46 (2H, s; H-3a and H-3b), 7.39–7.45 (5H, m; H-4a and H-4b), 7.02–7.03 [(1H, imp, H-5a) and (2H, d; H-7a and H-7b)], 7.58–7.60 [(1H, imp; H-5b), and (2H, d; H-6a and H-6b)] and 2.21 (3H, s; H-8a and H-8b). MS: m/z = 355 (M⁺, 3%, C₂₄H₂₁NO₂), 337 (65, C₂₄H₁₉NO), 277 (15, C₁₈H₁₅NO₂), 249 (20, C₁₇H₁₃O₂), 116 (17, C₉H₈), 105 (100, C₇H₅O), 91 (62, C₇H₇) and 65 (44, C₅H₅).

N-(4-Methylphenyl)-3-(4-methylbenzoyl)-2-phenylmethylene propionamide (31a) and 5-(4-methylphenylamino)-5-(4-methylphenyl)-3-phenylmethylene-butyrolactone (31b): FTIR (KBr): 3360–3250 v(broad NH), 1762 v(butyrolactone, C=O), 1685 v(aroyl, C=O) and 1640 cm⁻¹ v(amide, C=O). ¹H NMR (DMSO-d₆): δ = 7.19–7.36 [(5H, m; H-1a and H-1b) and (1H, imp; H-2a and H-2b)], 3.31 (2H, imp; H-3a and H-3b), 7.44–7.53 (2H, d; H-4a and H-4b), 6.46–6.83 (2H, d; H-5a and H-5b), 2.2–2.27 (3H, s, H-6a and H-6b), 7.83–7.94 [(1H, imp; H-7b) and (2H, d; H-8a and H-8b)], 6.74 (1H, s; H-7a), 6.99–7.19 (2H, d; H-9a and H-9b) and 2.28–2.31 (1H, s; H-10a and H-10b). MS: m/z = 369.5 (M⁺, 0%, C₂₅H₁₈NO₂), 262 (3.5, C₁₈H₁₄O₂), 200 (12, C₁₂H₁₀NO₂), 143 (14, C₁₀H₇O), 134 (12, C₈H₈NO), 115 (6, C₉H₇), 91 (44, C₇H₇), 77 (100, C₆H₅) and 65 (4, C₅H₅).

N-(4-Methylphenyl)-3-(4-methoxybenzoyl)-2-phenylmethylene propionamide (32a) and 5-(4-methylphenylamino)-5-(4-methylphenyl)-3-phenylmethylene-butyrolactone (32b): FTIR (KBr): 3250–3300 v(broad NH), 1760 v(butyrolactone, C=O), 1675 v(aroyl, C=O) and 1643 cm⁻¹ v(amide, C=O). ¹H NMR (DMSO-d₆): δ = 7.34–7.48 [(5H, m; H-1a and H-1b) and (1H, imp; H-2a and H-2b)], 3.45 (2H, s; H-3a and H-3b), 7.20–7.30 (2H, d; H-4a and H-4b), 6.82–6.90 (2H, d; H-5a and H-5b), 3.70–3.75 (3H, s, H-6a and H-6b), 6.75 (1H, s; H-7a), 8.00–8.10 (1H, m; H-7b), 7.58–7.90 (2H, d; H-8a and H-8b), 7.03–7.12 (2H, d; H-9a and H-9b) and 2.22 (3H, s; H-10a and H-10b). MS: m/z = 385.5 (M⁺, 0%, C₂₅H₂₃NO₃), 294 (3.3, C₁₈H₁₆NO₃), 277 (12, C₁₈H₁₅NO₂), 187 (4, C₁₁H₉NO₂), 149 (46, C₉H₉O₂), 135 (100, C₈H₈NO), 100 (19, C₅H₄NO₂), 91 (50, C₇H₇) and 65 (13, C₅H₅).

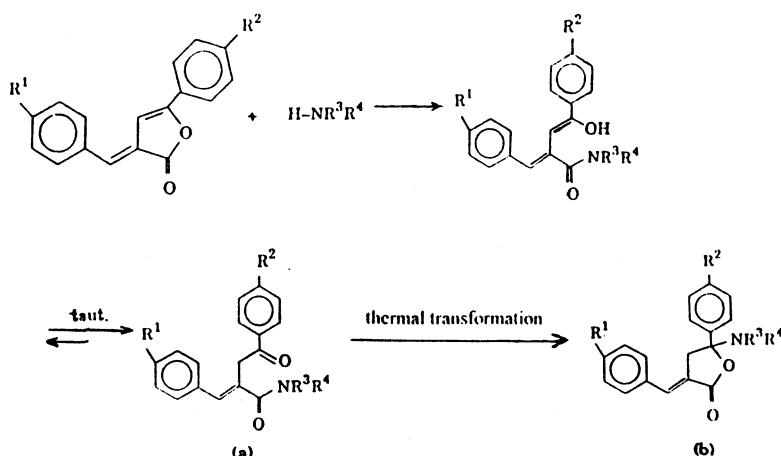
N-(4-Methylphenyl)-3-(4-chlorobenzoyl)-2-phenylmethylene propionamide (33a) and 5-(4-methylphenylamino)-5-(4-chlorophenyl)-3-phenylmethyl-

ene-butyrolactone (33b): FTIR (KBr): 3300–3250 ν (broad NH), 1765 ν (butyrolactone, C=O), 1682 ν (aryl, C=O) and 1646 cm⁻¹ ν (amide, C=O). ¹H NMR (DMSO-d₆): δ = 7.36–7.50 [(5H, m; H-1a and H-1b) and (1H, imp; H-2a and H-2b)], 3.64–3.57 (2H, s; H-3a and H-3b), 7.28–7.36 (2H, d; H-4a and H-4b), 7.62–7.64 (2H, d; H-5a and H-5b), 6.79–6.8 (1H, m, H-6a), 7.93–7.95 (1H, m; H-6b), 7.55–7.78 (2H, d; H-7a and H-7b), 7.02–7.09 (2H, d; H-8a and H-8b) and 2.21–2.26 (3H, s; H-9a and H-9b). MS: m/z = 389.8 (M⁺, 5%, C₂₄H₂₀NO₂Cl), 372 (5, C₂₄H₁₈NOCl), 282 (11, C₁₇H₁₁O₂Cl), 277 (11, C₁₈H₁₅NO₂), 153 (7, C₈H₆OCl), 139 (83, C₇H₄OCl), 134 (15, C₈H₈NO), 116 (7, C₉H₈), 111 (11, C₆H₄Cl), 91 (74, C₇H₇), 78 (100, C₆H₆) and 65 (32, C₅H₅).

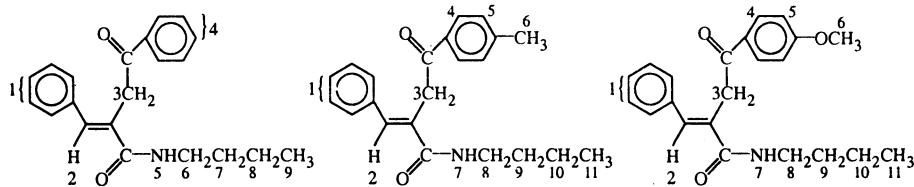
N-(4-Methylphenyl)-3-benzoyl-2-(4-methoxyphenylmethylene) propionamide (34a) and 5-(4-methylphenylamino)-5-phenyl-3-(4-methoxyphenylmethylene)butyrolactone (34b): FTIR (KBr): 3250–3400 ν (NH broad), 1750 ν (butyrolactone, C=O), 1670 ν (benzoyl, C=O) and 1625 cm⁻¹ ν (amide, C=O). ¹H NMR (DMSO-d₆): δ = 3.79–3.86 (3H, s; H-1a and H-1b), 7.00–7.09 (2H, d; H-2a and H-2b), 7.24–7.29 (2H, d; H-3a and H-3b), 7.38–7.62 (1H, imp; H-4a and H-4b), 3.34–3.41 (2H, s; H-5a and H-5b), 7.29–7.55 (5H, m, H-6a and H-6b), 6.98 (1H, s; H-7a), 7.62 (1H, s; H-7b), 7.88–7.91 (2H, d; H-8a and H-8b), 7.19–7.27 (2H, d; H-9a and H-9b) and 2.19 (3H, s; H-10a and H-10b). MS: m/z = 385.5 (M⁺, 0%, C₂₅H₂₃NO₃), 367 (100, C₂₅H₂₁NO₂), 308 (3, C₁₉H₁₈NO₃), 279 (4.4, C₁₈H₁₅O₃), 278 (5.5, C₁₈H₁₄O₃), 247 (5.3, C₁₇H₁₃NO), 135 (4, C₈H₉NO), 105 (10, C₇H₅O), 102 (4, C₈H₆), 91 (33, C₇H₇) and 65 (23, C₅H₅).

N-(4-Methylphenyl)-3-(4-methylbenzoyl)-2-(4-methoxyphenylmethylene) propionamide (35a) and 5-(4-methylphenylamino)-5-(4-methylphenyl)-3-(4-methoxyphenylmethylene)-butyrolactone (35b): FTIR (KBr): 3250–3350 ν (broad NH), 1768 ν (butyrolactone, C=O), 1691 ν (aryl, C=O) and 1625 cm⁻¹ ν (amide, C=O). ¹H NMR (DMSO-d₆): δ = 3.74–3.83 (3H, s; H-1a and H-1b), 6.46–6.72 (2H, d; H-2a and H-2b), 7.17–7.19 (2H, d; H-3a and H-3b), 7.29–7.34 (1H, s; H-4a and H-4b), 3.31–3.50 (2H, imp; H-5a and H-5b), 7.38–7.48 (2H, d, H-6a and H-6b), 6.80–6.89 (2H, d; H-7a and H-7b), 2.12–2.26 (3H, s; H-8a and H-8b), 6.98 (1H, s; H-9a), 7.83–8.00 [(1H, imp; H-9b) and (2H, d; H-10a and H-10b)], 7.00–7.19 (2H, d; H-11a and H-11b), 2.27–2.30 (3H, s; H-12a and H-12b). MS: m/z = 399 (M⁺, 2%, C₂₆H₂₅NO₃), 381 (100, C₂₆H₂₃NO₂), 308 (4, C₁₉H₁₈O₃N), 293 (9, C₁₉H₁₇O₃), 292 (7, C₁₉H₁₆O₃), 135 (3, C₈H₉NO), 119 (21, C₈H₇O), 107 (4, C₇H₇O), 91 (40, C₇H₇) and 65 (29, C₅H₅).

N-(4-Methylphenyl)-3-(4-chlorobenzoyl)-2-(4-methoxyphenylmethylene) propionamide (36a) and 5-(4-methylphenylamino)-5-(4-chlorophenyl)-3-(4-methoxyphenylmethylene)-butyrolactone (36b): FTIR (KBr): 3400–3300 ν (broad NH), 1760 ν (butyrolactone, C=O), 1690 ν (aryl, C=O) and 1625 cm⁻¹ ν (amide, C=O). ¹H NMR (DMSO-d₆): δ = 3.82–3.84 (3H, s; H-1a and H-1b), 6.45–6.82 (2H, d; H-2a and H-2b), 7.06–7.20 (2H, d; H-3a and H-3b), 7.28–7.39 (1H, imp; H-4a and H-4b), 3.31 (2H, imp; H-5a and H-5b), 7.28–7.39 (2H, d, H-6a and H-6b), 7.85–7.95 [(2H, d; H-7a and H-7b) and (1H, imp; H-8b)], 6.98 (1H, s; H-8a), 7.43–7.61 (2H, d; H-9a and H-9b), 7.01–7.09 (2H, d; H-10a and H-10b) and 2.20–2.23 (3H, s; H-11a and H-11b). MS: m/z = 419 (M⁺, 0%, C₂₅H₂₂NO₃Cl), 401 (100, C₂₅H₂₀NO₂Cl), 374 (10, C₂₃H₁₇NO₂Cl), 311 (7, C₁₈H₁₄NO₂Cl), 139 (34, C₇H₄OCl), 135 (13, C₈H₉NO), 111 (4, C₆H₄Cl), 91 (89, C₇H₇) and 65 (56, C₅H₅).



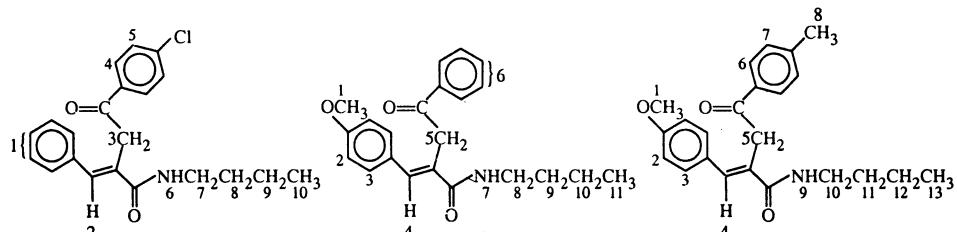
Comp.	R^1	R^2	R^3	R^4	Comp.	R^1	R^2	R^3	R^4
1.	H	H	—	—	19.	H	Cl	H	CH ₃
2.	H	CH ₃	—	—	20.	OCH ₃	CH ₃	H	CH ₃
3.	H	OCH ₃	—	—	21.	OCH ₃	OCH ₃	H	CH ₃
4.	H	Cl	—	—	22.	OCH ₃	Cl	H	CH ₃
5.	OCH ₃	H	—	—	23.	H	H	H	C ₆ H ₅
6.	OCH ₃	CH ₃	—	—	24.	H	CH ₃	H	C ₆ H ₅
7.	OCH ₃	OCH ₃	—	—	25.	H	OCH ₃	H	C ₆ H ₅
8.	OCH ₃	Cl	—	—	26.	H	Cl	H	C ₆ H ₅
9.	H	H	H	NH(CH ₂) ₃ CH ₃	27.	OCH ₃	H	H	C ₆ H ₅
10.	H	CH ₃	H	NH(CH ₂) ₃ CH ₃	28.	OCH ₃	CH ₃	H	C ₆ H ₅
11.	H	OCH ₃	H	NH(CH ₂) ₃ CH ₃	29.	OCH ₃	Cl	H	C ₆ H ₅
12.	H	Cl	H	NH(CH ₂) ₃ CH ₃	30.	H	H	H	4-CH ₃ C ₆ H ₄
13.	OCH ₃	H	H	NH(CH ₂) ₃ CH ₃	31.	H	CH ₃	H	4-CH ₃ C ₆ H ₄
14.	OCH ₃	CH ₃	H	NH(CH ₂) ₃ CH ₃	32.	H	OCH ₃	H	4-CH ₃ C ₆ H ₄
15.	OCH ₃	OCH ₃	H	NH(CH ₂) ₃ CH ₃	33.	H	Cl	H	4-CH ₃ C ₆ H ₄
16.	OCH ₃	Cl	H	NH(CH ₂) ₃ CH ₃	34.	OCH ₃	H	H	4-CH ₃ C ₆ H ₄
17.	H	CH ₃	H	CH ₃	35.	OCH ₃	CH ₃	H	4-CH ₃ C ₆ H ₄
18.	H	OCH ₃	H	CH ₃	36.	1OCH ₃	Cl	H	—



9

10

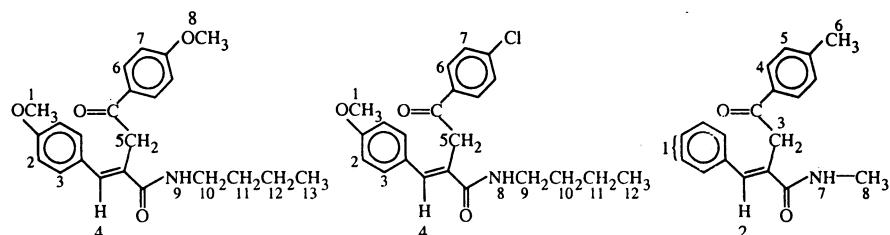
11



12

13

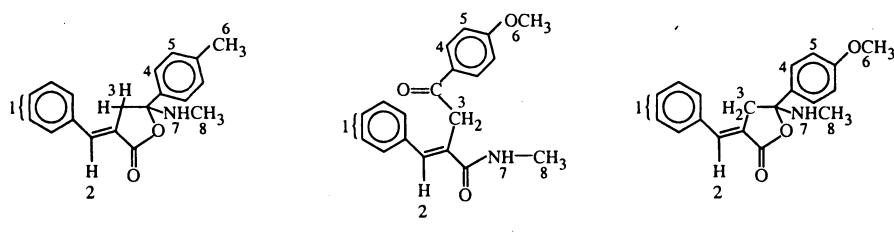
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15

16

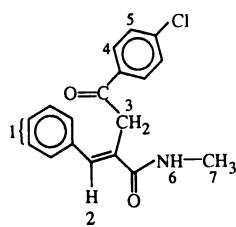
17(a)



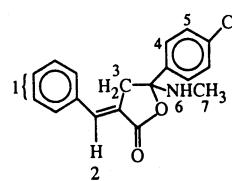
17(b)

18(a)

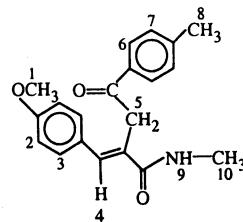
18(b)



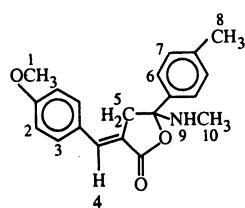
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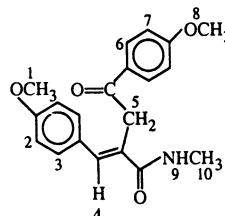
19(b)



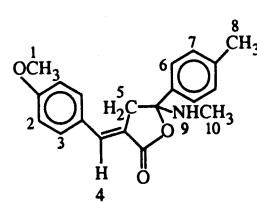
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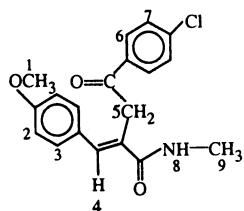
20(b)



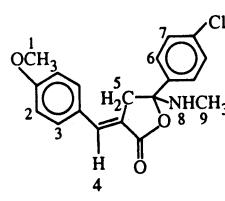
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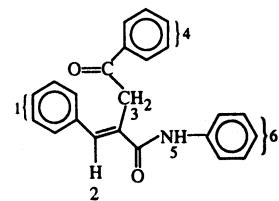
21(b)



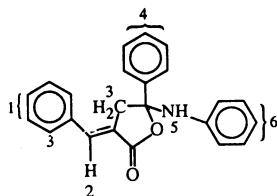
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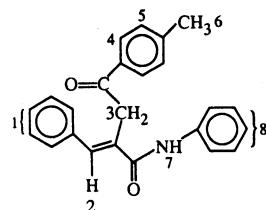
22(b)



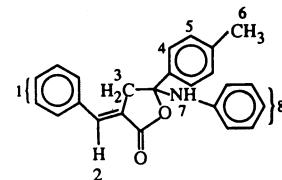
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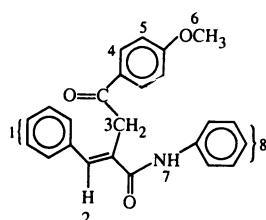
23(b)



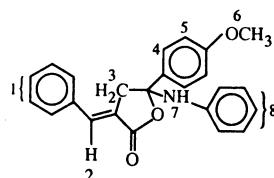
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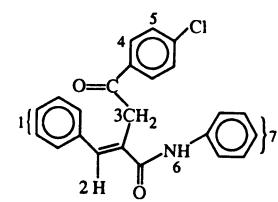
24(b)



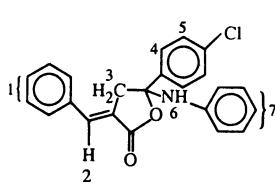
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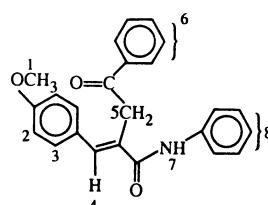
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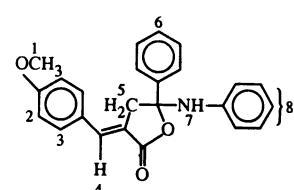
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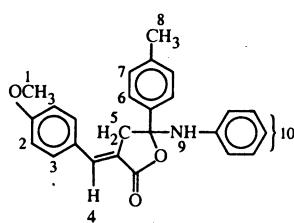
26(b)



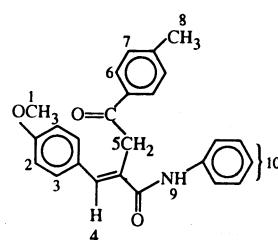
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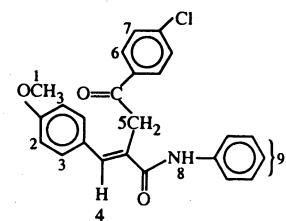
37(h)



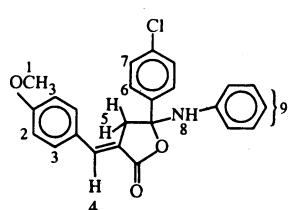
28(a)



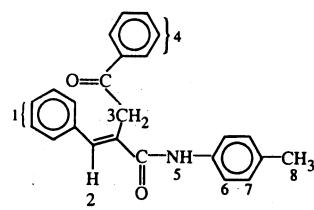
28(h)



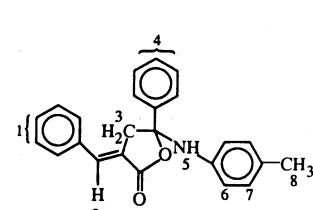
29(a)



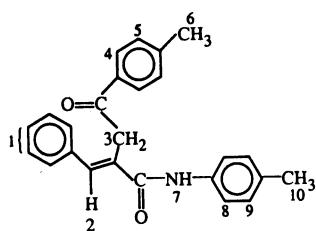
29(b)



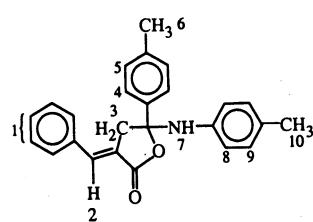
30(a)



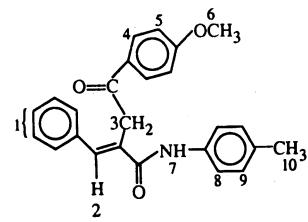
30(б)



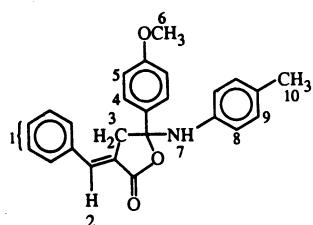
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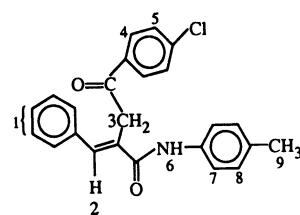
31(b)



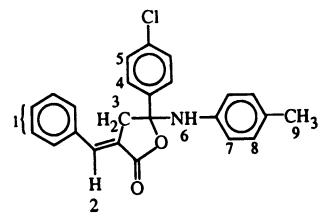
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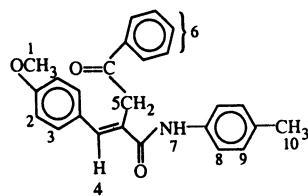
32(b)



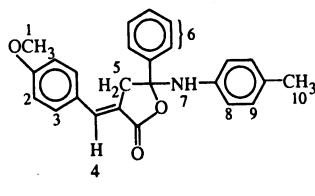
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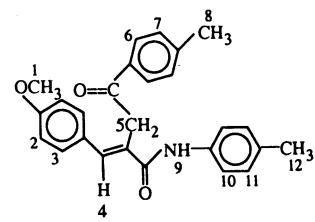
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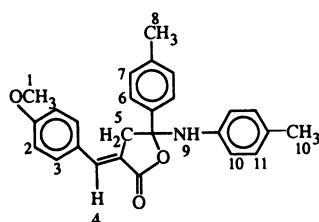
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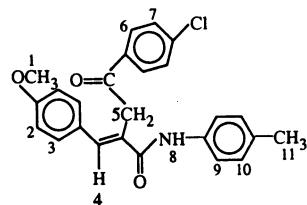
34(b)



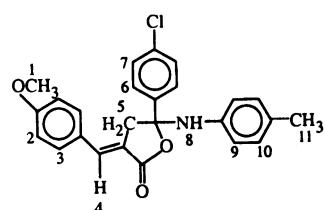
35(a)



35(b)



36(a)



36(B)

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