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# Spiro-condensation of 5-methoxycarbonyl-1*H*-pyrrole-2,3-diones with cyclic enoles to form spiro substituted furo[3,2-*c*]-coumarins and quinolines†

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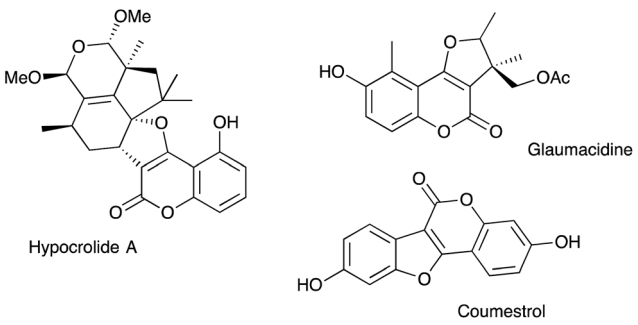
Highly efficient spiro-condensation enabling cyclic enoles to act as 1,3-bis-nucleophiles in reaction with pyrrole-2,3-diones acting as 1,2-bis-electrophiles was developed. The corresponding furo[3,2-*c*]coumarins and furo[3,2-*c*]quinolines containing a spiro pyrrole fragment were obtained in high yields.

## Introduction

Natural products containing furo[3,2-*c*]coumarin[1] and furo[3,2-*c*]quinolone[2] fragments have been the subject of great interest due to their important biological properties. For example, phyto-alkaloids hypocrolide A, glaumacidine, coumestrol (coumestan), and their synthetic analogs demonstrated promising antimitotic, antiprolifirative and cytotoxic activities (Scheme 1). Not surprisingly, development of new synthetic routes to these scaffolds is of great interest for modern synthetic and medicinal chemistry. Herein we wish to disclose the results of our synthetic studies towards spiro analogs of furo[3,2-*c*]coumarins and furo[3,2-*c*]quinolones. The featured synthetic strategy is based on highly selective electrophilic reactions of 1*H*-pyrrole-2,3-diones.




Scheme 1

## Results and discussion

Due to their high reactivity and pronounced electrophilic properties 1*H*-pyrrole-2,3-diones are often employed as useful building blocks allowing for efficient incorporation of a nitrogen containing five-membered heterocyclic unit in the structure of a target molecule. Such approach has found application in total synthesis of natural alkaloids.[3] The installation of additional electron-withdrawing groups (such as acyl, alkoxycarbonyl or 1,2-dicarbonyl moieties) at C-4 and C-5 of the pyrrole ring may further increase synthetic versatility of these compounds. Diverse reactions of such 4,5-disubstituted 1*H*-pyrrole-2,3-diones can be used for expeditious assembly of various fused and bridged, polyheterocyclic scaffolds, often hardly available or unavailable by other methods.[4] Furthermore, the installation of an ester function at C-5 provides an additional electrophilic moiety in multistep cascade reactions with bis-nucleophilic reagents, offering easy access towards functionalized spiro-pyrroles (Scheme 2). Thus, we previously demonstrated the reaction of 5-methoxycarbonyl-1*H*-pyrrole-2,3-diones **1** with 1,3-*N*,*N*-bis-nuclephiles, such as urea derivatives **2** to yield derivatives of 1,3,6-triazaspiro[4,4]nonane **3** (Scheme 2, path A).[5] Similarly, utilization of enamines **4** as 1,3-*C*,*N*-bisnucleophiles afforded products with 1,7-diazaspiro[4,4]nonane scaffold **5** (Scheme 2, path B).[6,7] A detailed investigation of this transformation revealed that both acyclic[6] and cyclic[7] enamines can be readily employed as bis-nucleophiles, providing facile access to annulated functionalized spiranes **5** with increased molecular complexity. At the same time, to the best of our knowledge, related spiro-condensations of monocyclic 1*H*-pyrrole-2,3-diones **1** with enoles **6** (Scheme 2, path C) is still unknown. We envisioned that such a process, if

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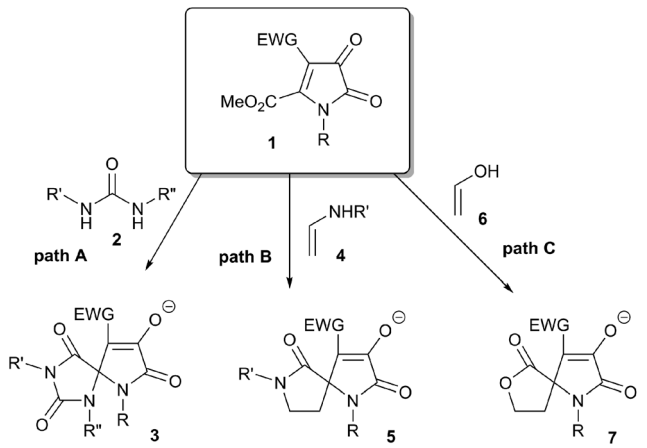
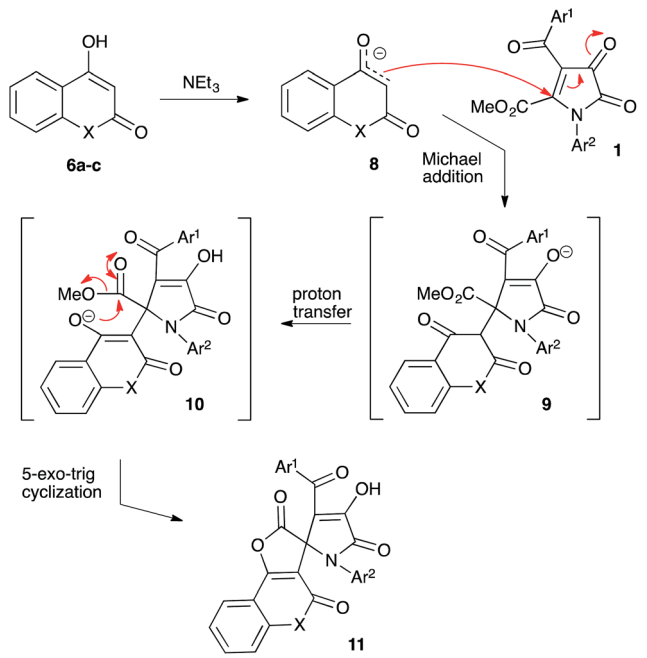



Scheme 2

successful, might serve for efficient preparation of 7-oxa-1-azaspiro[4,4]nonanes **7**, including spiro-derivatives of furo[3,2-*c*]coumarins and furo[3,2-*c*]quinolones.

To this end we envisioned that stable enol forms such as 4-hydroxycoumarine (**6a**, X = O) and 4-hydroxyquinolin-2(1*H*)-ones (**6b**, X = NMe; **6b**, X = NPh) could serve as suitable precursors for the designed transformation. Indeed, enolate **8** generated in the presence of catalytic base is expected to perform a conjugate addition across the highly electrophilic vinylogous amide moiety in pyrroledione **1**. The resulting enolate **9** is anticipated to undergo a proton transfer to produce less basic enolate **10**, which is well suited for an intramolecular 5-*exo*-trig cyclization affecting the ester function at C-5 and providing target structure **11** (Scheme 3).

To evaluate this idea we carried out the reaction of methyl 3-benzoyl-4,5-dioxo-1-phenyl-4,5-dihydro-1*H*-pyrrole-2-carboxylate




Scheme 3

Table 1 Spiro-condensation of 4-aroyl-5-methoxycarbonyl-1*H*-pyrrole-2,3-diones **1** with cyclic enoles **6**

| # | 1 | 6 | X | $Ar^1$ | $Ar^2$ | 11 | Yield[a], % |
|---|---|---|---|---|---|---|---|
| 1 | **1a** | **6a** | O | Ph | Ph | **11aa** | 68 |
| 2 | **1b** | **6a** | O | 4-$MeOC_6H_4$ | Ph | **11ba** | 70 |
| 3 | **1c** | **6a** | O | Ph | 4-$MeOC_6H_4$ | **11ca** | 84 |
| 4 | **1d** | **6a** | O | Ph | 4-$MeC_6H_4$ | **11da** | 80 |
| 5 | **1e** | **6a** | O | 4-$MeC_6H_4$ | Ph | **11ea** | 76 |
| 6 | **1f** | **6a** | O | 4-$MeC_6H_4$ | 4-$MeC_6H_4$ | **11fa** | 78 |
| 7 | **1g** | **6a** | O | Ph | 4-$ClC_6H_4$ | **11ga** | 67 |
| 8 | **1h** | **6a** | O | Ph | 4-$BrC_6H_4$ | **11ha** | 76 |
| 9 | **1c** | **6b** | NMe | Ph | 4-$MeOC_6H_4$ | **11cb** | 68 |
| 10 | **1d** | **6b** | NMe | Ph | 4-$MeC_6H_4$ | **11db** | 69 |
| 11 | **1a** | **6c** | NPh | Ph | Ph | **11ac** | 56 |
| 12 | **1g** | **6c** | NPh | Ph | 4-$ClC_6H_4$ | **11gc** | 59 |
| 13 | **1i** | **6c** | NPh | 4-$BrC_6H_4$ | 4-$MeC_6H_4$ | **11ic** | 67 |

[a] Isolated yields of purified compounds **11**.

(**1a**, $Ar^1$ = Ph, $Ar^2$ = Ph) with 4-hydroxy-2*H*-chromen-2-one (**6a**). It was observed that an equimolar mixture of these starting materials when refluxed in toluene in the presence of catalytic amounts of triethylamine (10 mol%) engaged in a quick reaction. The distinct purple color of pyrroledione faded away and the colorless product **11aa** crystallized directly from the reaction mixture after cooling it down to −10 °C. This furo-coumarine was obtained as the sole product in good yield (68%, Table 1, entry 1). Reactions of other 4-aroyl-5-methoxycarbonyl pyrrolediones with **6a** also proceeded uneventfully, affording the corresponding furo[3,2-*c*]coumarins very efficiently (Table 1, entries 2–8). Reactions involving enolates generated from 4-hydroxy-2-quinolones **6b**, **c** were carried out under the same conditions and also afforded the corresponding spiro furo[3,2-*c*]quinolones as sole products, albeit in somewhat lower yields (Table 1, entries 9–13). Formation of these structures was unambiguously confirmed by X-ray crystallography of compounds **11ba** (Fig. 1) and **11db** (Fig. 2).

We wondered, if the featured spiro-condensation would proceed selectively in more complex substrates, possibly

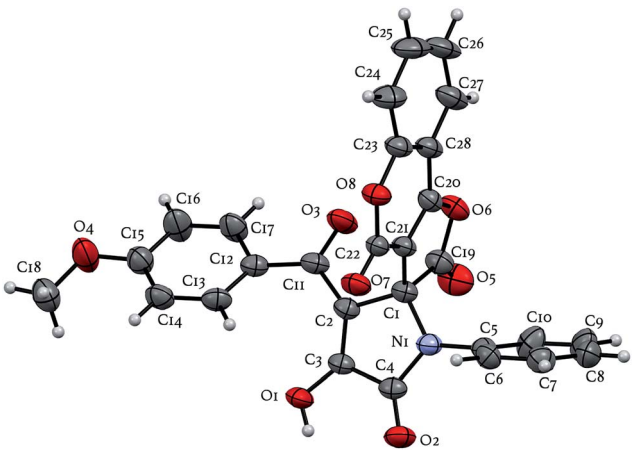



Fig. 1 ORTEP drawing of compound **11ba**: showing 50% probability amplitude displacement ellipsoids (CCDC # 1486439†).

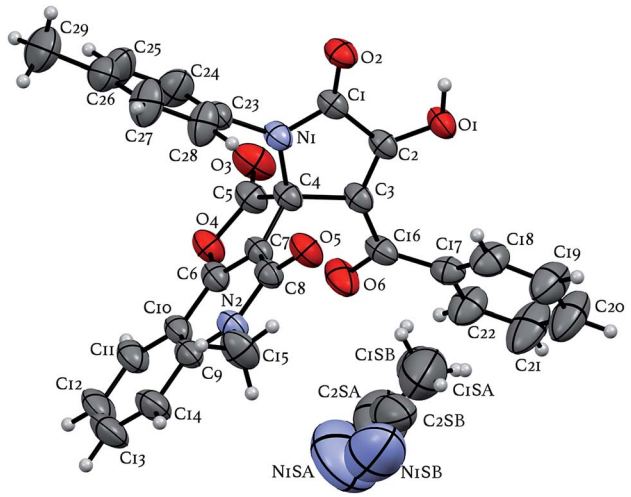
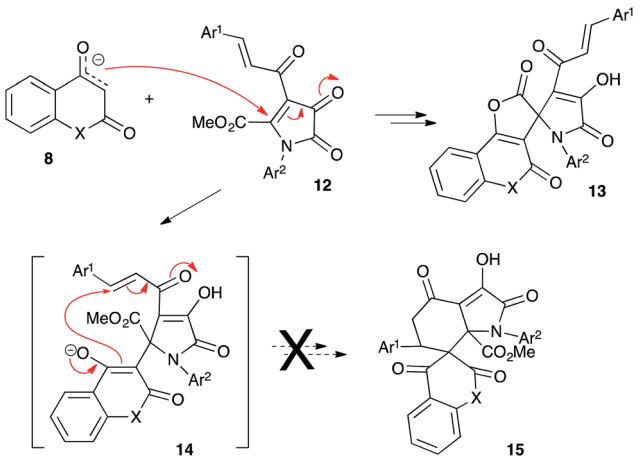



Fig. 2 ORTEP drawing of compound **11db**: showing 50% probability amplitude displacement ellipsoids and a molecule of disordered crystallized solvent (acetonitrile) (CCDC # 1486440†).




Scheme 4

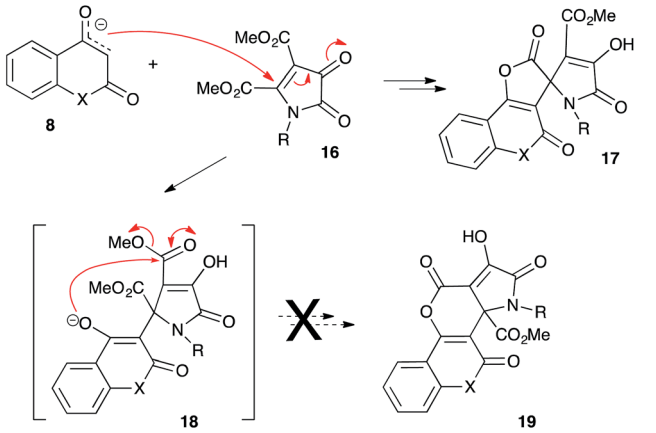
allowing for alternative reaction pathways. For example, reaction of pyrrolediones **12** bearing a cinnamoyl substituent at C-4, with bis-nucleophilic enolates should proceed through intermediate **14**, which could in principle undergo a second-fold enol*exo*-6-*endo*-trig Michael addition reaction to furnish different spiro scaffold **15** (Scheme 4). In our recent report we disclosed that in reactions of 4-cinnamoyl-pyrrole-2,3-diones **12** with five-membered cyclic enolates this process was prevalent.[8] Interestingly, in the presence of six-membered enolates, generated from heterocyclic precursors **6a–c** the reaction did not take this alternative route at all, proceeding instead *via* a "normal" 5-*exo*-trig lactonization pathway to afford the corresponding furo[3,2-*c*]coumarins and furo[3,2-*c*]quinolones **13** as sole products (Scheme 4, Table 2).

Next, we investigated the reactivity of 4,5-dimethoxycarbonyl-1*H*-pyrrole-2,3-diones **16**. After initial Michael addition of cyclic enolate **8**, these substrates should provide intermediate **18**, which can undergo the subsequent

Table 2 Spiro-condensation of 4-cinnamoyl-5-methoxycarbonyl-1*H*-pyrrole-2,3-diones **12** with cyclic enoles **6**

| # | 12 | 6 | X | $Ar^1$ | $Ar^2$ | 13 | Yield[a], % |
|---|---|---|---|---|---|---|---|
| 1 | **12a** | **6a** | O | Ph | 4-$MeC_6H_4$ | **13aa** | 67 |
| 2 | **12a** | **6b** | NMe | Ph | 4-$MeC_6H_4$ | **13ab** | 75 |
| 3 | **12b** | **6b** | NMe | Ph | 4-$MeOC_6H_4$ | **13bb** | 78 |
| 4 | **12c** | **6b** | NMe | 4-$MeOC_6H_4$ | 4-$MeOC_6H_4$ | **13cb** | 77 |
| 5 | **12b** | **6c** | NPh | Ph | 4-$MeOC_6H_4$ | **13bc** | 65 |
| 6 | **12d** | **6c** | NPh | 4-$MeOC_6H_4$ | 4-$MeC_6H_4$ | **13dc** | 62 |

[a] Isolated yields of purified compounds **13**.




Scheme 5

Table 3 Spiro-condensation of 4,5-dimethoxycarbonyl-1*H*-pyrrole-2,3-diones **16** with cyclic enoles **6**

| # | 16 | 6 | X | $Ar^1$ | 17 | Yield[a], % |
|---|---|---|---|---|---|---|
| 1 | **16a** | **6a** | O | $CH_2Ph$ | **17aa** | 83 |
| 2 | **16b** | **6a** | O | Ph | **17ba** | 84 |
| 3 | **16c** | **6a** | O | 4-$MeC_6H_4$ | **17ca** | 81 |
| 4 | **16a** | **6c** | NPh | $CH_2Ph$ | **17ac** | 79 |
| 5 | **16b** | **6c** | NPh | Ph | **17bc** | 69 |
| 6 | **16c** | **6c** | NPh | 4-$MeC_6H_4$ | **17cc** | 64 |

[a] Isolated yields of purified compounds **17**.

intramolecular nucleophilic attack involving one of the two available ester groups (Scheme 5). We were pleased to discover that these reactions also proceeded chemoselectively according to 5-*exo*-trig pathway, providing γ-lactones **17** as sole products in high yields (Table 3, entries 1–6). The alternative 6-*exo*-trig pathway was not realized at all, and the corresponding δ-lactones **18** were not detected in the reaction mixtures (Scheme 5).

## Conclusions

We have developed a novel cascade transformation combining an intermolecular conjugate addition of stabilized cyclic enolate (4-

hydroxycoumarin or 4-hydroxy-2-quinolone) across a highly electrophilic vinylogous amide moiety of pyrrole-2,3-dione, and subsequent lactonization involving intramolecular nucleophilic attack of *O*-enolate at the ester substituent at C-5. The corresponding spiro furo[3,2-*c*]coumarins and furo[3,2-*c*]quinolones were formed as sole products in good yields. Remarkably, the introduction of competitive electrophilic substituents at C-4, such as cinnamoyl or methoxycarbonyl groups, did not divert the reaction from the described mechanistic route.

## Experimental part

$^1$H and $^{13}$C NMR spectra were recorded on a Bruker Avance-III spectrometer (400 or 100 MHz, respectively) equipped with BBO probe in $CDCl_3$ or DMSO-$d_6$ using TMS as internal standard. IR spectra were recorded with a Perkin–Elmer Spectrum Two spectrometer from mulls in mineral oil. Melting points were measured with Stuart smp30 apparatus. X-ray crystallography was performed on Xcalibur Ruby diffractometer. The mass spectra were recorded on an Waters UPLC-MS instrument equipped with an ESI MS Xevo TQD detector. Elemental analyses were carried out on Vario MICRO Cube analyzer. Starting 5-methoxycarbonyl 1*H*-pyrrole-2,3-diones **1a–i**, **12a–d**, and **16a–c** were obtained as described in literature sources,[9] *N*-methyl- and *N*-phenyl-4-hydroxy-2-quinolones **6b**, **c** were also prepared according to literature procedures.[10] Anhydrous toluene was obtained by heating at reflux with molten sodium followed by distillation in under an atmosphere of dry nitrogen. Other reagents and solvents were purchased from commercial vendors and were used as received.

### 3′-Benzoyl-4′-hydroxy-1′-(4-methoxyphenyl)-2*H*,4*H*-spiro[furo[3,2-*c*]chromene-3,2′-pyrrole]-2,4,5′(1′*H*)-trione (11ca, typical procedure)

A solution of 4-hydroxycoumarin **6a** (162 mg, 1.00 mmol), methyl 3-benzoyl-1-(4-methoxyphenyl)-4,5-dioxo-4,5-dihydro-1*H*-pyrrole-2-carboxylate **1c** (365 mg, 1.00 mmol), and $Et_3N$ (10 mg, 0.10 mmol) in anhydrous toluene (5 mL) was stirred at reflux for 1.5 h until purple color of pyrroledione faded away, and the solid precipitate formed. Then the reaction mixture was cooled to −10 °C, and the resulted precipitate was filtered off, washed with hexane and recrystallized from toluene/chloroform (2 : 1) to afford **11ca** (416 mg, 84%) as a colorless crystals, mp 240–242 °C (dec.). $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 7.88–7.85 (m, 2H, Ar), 7.76–7.71 (m, 1H, Ar), 7.67–7.53 (m, 2H, Ar), 7.48–7.31 (m, 4H, Ar), 7.19–7.13 (m, 2H, Ar), 6.87–6.83 (m, 2H, Ar), 3.74 (s, 3H, OMe); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 188.9, 169.7, 166.2, 165.4, 160.5, 156.1, 155.9, 150.4, 136.5, 134.9, 133.9, 129.6, 129.0, 128.6, 125.8, 125.1, 123.9, 117.7, 116.6, 115.5, 110.3, 100.9, 70.0, 55.6; IR (NaCl, cm$^{-1}$): 3408, 1848, 1736, 1707, 1676, 1652; MS: found 496.17; calcd for $C_{28}H_{18}NO_8$ $(M + H)^+$ 496.10; EA: found C 67.84, H 3.45, N 2.86; calcd for $C_{28}H_{17}NO_8$ (495.44): C 67.88, H 3.46, N 2.83.

### 3′-Benzoyl-4′-hydroxy-1′-phenyl-2*H*,4*H*-spiro[furo[3,2-*c*]chromene-3,2′-pyrrole]-2,4,5′(1′*H*)-trione (11aa)

Yield 316 mg (68%), mp 237–239 °C (dec.), colorless crystals. $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 7.89–7.85 (m, 2H, Ar), 7.73 (ddd, $J$ = 7.9, 1.6, 0.5 Hz, 1H, Ar), 7.65–7.53 (m, 2H, Ar), 7.46–7.40 (m, 2H, Ar), 7.38–7.30 (m, 5H, Ar), 7.27–7.24 (m, 2H, Ar); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 188.9, 169.6, 166.0, 165.4, 156.0, 155.9, 150.4, 136.5, 134.9, 133.9, 133.5, 130.2, 129.8, 129.6, 128.6, 127.5, 125.1, 123.9, 117.7, 116.7, 110.3, 100.8, 69.7 ppm. IR (NaCl, cm$^{-1}$): 3413, 1851, 1744, 1711, 1674, 1648; MS: found 466.15; calcd for $C_{27}H_{16}NO_7$ $(M + H)^+$ 466.09; EA: found C 69.64, H 3.23, N 3.04; calcd for $C_{27}H_{15}NO_7$ (465.42): C 69.68, H 3.25, N 3.01.

### 4′-Hydroxy-3′-(4-methoxybenzoyl)-1′-phenyl-2*H*,4*H*-spiro[furo[3,2-*c*]chromene-3,2′-pyrrole]-2,4,5′(1′*H*)-trione (11ba)

Yield 347 mg (70%), mp 251–252 °C (dec.), colorless crystals. $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 7.92–7.87 (m, 2H, Ar), 7.78–7.71 (m, 1H, Ar), 7.77 (ddd, $J$ = 8.4, 7.5, 1.6 Hz, 1H, Ar), 7.41–7.23 (m, 7H, Ar), 6.97–6.92 (m, 2H, Ar), 3.87 (s, 3H, OMe); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 186.9, 169.7, 166.0, 165.4, 164.6, 156.0, 155.9, 148.9, 134.9, 133.7, 132.3, 130.2, 129.7, 129.2, 127.5, 125.1, 123.9, 177.7, 117.1, 114.1, 110.3, 100.9, 65.9, 55.7; IR (NaCl, cm$^{-1}$): 3428, 1848, 1737, 1717, 1671, 1651; MS: found 496.19; calcd for $C_{28}H_{18}NO_8$ $(M + H)^+$ 496.10; EA: found C 67.83, H 3.46, N 2.84; calcd for $C_{28}H_{17}NO_8$ (495.44): C 67.88, H 3.46, N 2.83.

### 3′-Benzoyl-4′-hydroxy-1′-(4-tolyl)-2*H*,4*H*-spiro[furo[3,2-*c*]chromene-3,2′-pyrrole]-2,4,5′(1′*H*)-trione (11da)

Yield 383 mg (80%), mp 203–205 °C (dec.), colorless crystals. $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 7.92–7.87 (m, 2H, Ar), 7.78–7.71 (m, 1H, Ar), 7.68–7.53 (m, 2H, Ar), 7.48–7.40 (m, 2H, Ar), 7.37–7.30 (m, 2H, Ar), 7.19–7.06 (m, 4H, Ar), 2.21 (s, 3H, Me); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 188.9, 169.7, 166.1, 165.4, 156.0, 155.9, 150.5, 140.1, 136.5, 134.9, 133.9, 130.8, 130.8, 129.6, 128.6, 127.3, 125.0, 123.9, 117.7, 116.6, 110.3, 100.9, 69.8, 21.3; IR (NaCl, cm$^{-1}$): 3441, 1851, 1724, 1708, 1674, 1646; MS: found 480.19; calcd for $C_{28}H_{18}NO_7$ $(M + H)^+$ 480.11; EA: found C 70.12, H 3.56, N 2.94; calcd for $C_{28}H_{17}NO_7$ (479.44): C 70.15, H 3.57, N 2.92.

### 4′-Hydroxy-3′-(4-methylbenzoyl)-1′-phenyl-2*H*,4*H*-spiro[furo[3,2-*c*]chromene-3,2′-pyrrole]-2,4,5′(1′*H*)-trione (11ea)

Yield 364 mg (76%), mp 229–231 °C (dec.), colorless crystals. $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 7.81–7.71 (m, 3H, Ar), 7.63 (ddd, $J$ = 8.4, 7.5, 1.6 Hz, 1H, Ar), 7.41–7.30 (m, 5H, Ar), 7.28–7.21 (m, 4H, Ar), 2.40 (s, 3H, Me); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 188.4, 169.7, 166.1, 165.4, 156.0, 155.9, 149.9, 145.2, 134.9, 133.8, 133.5, 130.2, 129.8, 129.8, 129.4, 127.5, 124.9, 123.9, 117.7, 117.0, 110.2, 100.7, 69.7, 22.0; IR (NaCl, cm$^{-1}$): 3420, 1854, 1713, 1673, 1648; MS: found 480.18; calcd for $C_{28}H_{18}NO_7$ $(M + H)^+$ 480.11; EA: found C 70.11, H 3.55, N 2.92; calcd for $C_{28}H_{17}NO_7$ (479.44): C 70.15, H 3.57, N 2.92.

### 4′-Hydroxy-3′-(4-methylbenzoyl)-1′-(4-tolyl)-2*H*,4*H*-spiro[furo[3,2-*c*]chromene-3,2′-pyrrole]-2,4,5′(1′*H*)-trione (11fa)

Yield 385 mg (78%), mp 259–260 °C (dec.), colorless crystals. $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 7.80–7.71 (m, 3H, Ar), 7.63 (ddd, $J$ = 8.3, 7.5, 1.6 Hz, 1H, Ar), 7.38–7.29 (m, 2H, Ar), 7.27–7.22 (m, 3H,

Ar), 7.17–7.11 (m, 3H, Ar), 2.40 (s, 3H, Me), 2.29 (s, 3H, Me); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 188.3, 169.8, 166.0, 165.3, 155.9, 149.8, 145.1, 140.0, 134.8, 133.9, 130.8, 129.8, 129.4, 127.3, 125.0, 123.9, 117.7, 116.7, 110.4, 101.0, 69.8, 22.0, 21.3; IR (NaCl, $cm^{-1}$): 3438, 1848, 1723, 1673, 1646; MS: found 494.23; calcd for $C_{29}H_{20}NO_7$ $(M + H)^+$ 494.12; EA: found C 70.54, H 3.86, N 2.85; calcd for $C_{29}H_{19}NO_7$ (493.47): C 70.59, H 3.88, N 2.84.

### 3′-Benzoyl-1′-(4-chlorophenyl)-4′-hydroxy-2*H*,4*H*-spiro[furo[3,2-*c*]chromene-3,2′-pyrrole]-2,4,5′(1′*H*)-trione (11ga)

Yield 335 mg (67%), mp 233–235 °C (dec.), colorless crystals. $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 7.88–7.84 (m, 2H, Ar), 7.77 (dd, $J$ = 7.9, 1.6 Hz, 1H, Ar), 7.70–7.62 (m, 1H, Ar), 7.58 (t, $J$ = 7.4 Hz, 1H, Ar), 7.45 (t, $J$ = 7.7 Hz, 2H, Ar), 7.40–7.30 (m, 4H, Ar), 7.20 (dd, $J$ = 9.1, 2.4 Hz, 2H, Ar); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 188.8, 169.6, 165.9, 165.6, 156.0 (2C), 150.0, 136.4, 136.0, 135.2, 134.1, 132.0, 130.5, 129.6, 128.9, 128.7, 125.2, 124.0, 117.8, 116.9, 110.2, 100.5, 69.6; IR (NaCl, $cm^{-1}$): 3464, 1849, 1737, 1709, 1680, 1656; MS: found 500.17; calcd for $C_{27}H_{15}ClNO_7$ $(M + H)^+$ 500.05; EA: found C 64.83, H 2.80, Cl 7.12, N 2.81; calcd for $C_{27}H_{14}ClNO_7$ (499.86): C 64.88, H 2.82, Cl 7.09, N 2.80.

### 3′-Benzoyl-1′-(4-bromophenyl)-4′-hydroxy-2*H*,4*H*-spiro[furo[3,2-*c*]chromene-3,2′-pyrrole]-2,4,5′(1′*H*)-trione (11ha)

Yield 413 mg (76%), mp 235–236 °C (dec.), colorless crystals. $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 7.88–7.83 (m, 2H, Ar), 7.77 (dd, $J$ = 7.9, 1.6 Hz, 1H, Ar), 7.66 (ddd, $J$ = 8.9, 7.5, 1.6 Hz, 1H, Ar), 7.58 (ddd, $J$ = 8.7, 2.5, 1.2 Hz, 1H, Ar), 7.52–7.41 (m, 4H, Ar), 7.39–7.33 (m, 2H, Ar), 7.16–7.11 (m, 2H, Ar); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 188.8, 169.6, 165.8, 165.6, 156.0, 156.0, 149.9, 136.4, 135.2, 134.1, 133.5, 132.6, 129.6, 129.1, 128.7, 125.2, 124.1, 124.0, 117.8, 116.9, 110.2, 100.5, 69.6; IR (NaCl, $cm^{-1}$): 3473, 1848, 1738, 1709, 1678, 1654; MS: found 544.18; calcd for $C_{27}H_{15}BrNO_7$ $(M + H)^+$ 544.00; EA: found C 59.54, H 2.61, Br 14.73, N 2.57; calcd for $C_{27}H_{14}BrNO_7$ (544.31): C 59.58, H 2.59, Br 14.68, N 2.57.

### 3′-Benzoyl-4′-hydroxy-1′-(4-methoxyphenyl)-5-methyl-2*H*-spiro[furo[3,2-*c*]quinoline-3,2′-pyrrole]-2,4,5′(1′*H*,5*H*)-trione (11cb)

Yield 345 mg (68%), mp 243–244 °C (dec.), colorless crystals. $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 7.92–7.84 (m, 3H, Ar), 7.66 (ddd, $J$ = 8.8, 7.2, 1.6 Hz, 1H, Ar), 7.56–7.47 (m, 1H, Ar), 7.42–7.28 (m, 4H, Ar), 7.22–7.14 (m, 2H, Ar), 6.85–6.80 (m, 2H, Ar), 3.74 (s, 3H, Me), 3.63 (s, 3H, OMe); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 188.9, 171.2, 166.0, 161.0, 160.2, 157.7, 150.2, 141.9, 136.9, 133.5, 133.5, 129.7, 129.1, 128.5, 126.4, 124.2, 122.8, 116.6, 115.2, 115.1, 110.5, 105.6, 70.9, 55.5, 29.4; IR (NaCl, $cm^{-1}$): 3429, 1838, 1738, 1727, 1659, 1630; MS: found 509.21; calcd for $C_{29}H_{21}N_2O_7$ $(M + H)^+$ 509.13; EA: found C 68.44, H 3.93, N 5.56; calcd for $C_{29}H_{20}N_2O_7$ (508.49): C 68.50, H 3.96, N 5.51.

### 3′-Benzoyl-4′-hydroxy-5-methyl-1′-(4-tolyl)-2*H*-spiro[furo[3,2-*c*]quinoline-3,2′-pyrrole]-2,4,5′(1′*H*,5*H*)-trione (11db)

Yield 339 mg (69%), mp 257–259 °C (dec.), colorless crystals. $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 7.92–7.82 (m, 3H, Ar), 7.68 (ddd, $J$ = 8.8, 7.2, 1.6 Hz, 1H, Ar), 7.54–7.49 (m, 1H, Ar), 7.40–7.26 (m, 4H, Ar), 7.18–7.09 (m, 4H, Ar), 3.62 (s, 3H, Me), 2.28 (s, 3H, $C_6H_4Me$); $^{13}$C NMR (100 MHz, DMSO-$d_6$) $\delta$ 188.5, 171.7, 165.6, 159.2, 156.6, 153.8, 141.0, 138.7, 136.6, 133.6, 133.1, 131.4, 130.2, 128.9, 128.2, 126.6, 122.8 (2C), 116.0, 115.4, 108.9, 105.4, 69.4, 28.9, 20.5; IR (NaCl, $cm^{-1}$): 3449, 1837, 1738, 1659, 1634; MS: found 493.21; calcd for $C_{29}H_{21}N_2O_6$ $(M + H)^+$ 493.14; EA: found C 70.64, H 4.08, N 5.73; calcd for $C_{29}H_{20}N_2O_6$ (492.49): C 70.73, H 4.09, N 5.69.

### 3′-Benzoyl-4′-hydroxy-1′,5-diphenyl-2*H*-spiro[furo[3,2-*c*]quinoline-3,2′-pyrrole]-2,4,5′(1′*H*,5*H*)-trione (11ac)

Yield 303 mg (56%), mp 261–263 °C (dec.), colorless crystals. $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 7.89–7.81 (m, 3H, Ar), 7.62–7.46 (m, 4H, Ar), 7.45–7.28 (m, 8H, Ar), 7.23 (d, $J$ = 7.2 Hz, 1H, Ar), 7.15–7.12 (m, 1H, Ar), 7.03 (d, $J$ = 7.7 Hz, 1H, Ar), 6.67 (d, $J$ = 8.6 Hz, 1H, Ar); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 189.1, 171.1, 166.0, 161.6, 157.6, 150.3, 143.0, 136.9, 136.6, 134.2, 133.6, 133.0, 130.4, 129.9, 129.6, 129.5, 129.4, 129.2, 128.8, 128.6, 127.6, 123.7, 123.0, 116.9, 110.2, 105.8, 70.6; IR (NaCl, $cm^{-1}$): 3443, 1835, 1732, 1718, 1637, 1626; MS: found 541.21; calcd for $C_{33}H_{21}N_2O_6$ $(M + H)^+$ 541.14; EA: found C 73.29, H 3.71, N 5.20; calcd for $C_{33}H_{20}N_2O_6$ (540.53): C 73.33, H 3.73, N 5.18.

### 3′-Benzoyl-1′-(4-chlorophenyl)-4′-hydroxy-5-phenyl-2*H*-spiro[furo[3,2-*c*]quinoline-3,2′-pyrrole]-2,4,5′(1′*H*,5*H*)-trione (11gc)

Yield 339 mg (59%), mp 229–231 °C (dec.), colorless crystals. $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 7.90–7.84 (m, 3H, Ar), 7.62–7.48 (m, 4H, Ar), 7.42 (ddd, $J$ = 7.3, 4.7, 1.0 Hz, 3H, Ar), 7.37–7.22 (m, 5H, Ar), 7.16–7.10 (m, 1H, Ar), 7.07–7.01 (m, 1H, Ar), 6.69 (d, $J$ = 8.5 Hz, 1H, Ar); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 189.0, 170.9, 165.9, 161.7, 157.6, 149.8, 143.0, 136.8, 136.5, 135.6, 133.8, 133.2, 132.7, 130.4, 130.2, 129.6, 129.5, 129.2, 129.0, 128.8, 128.6, 123.8, 123.2, 117.0, 116.8, 110.2, 105.3, 70.6; IR (NaCl, $cm^{-1}$): 3400, 1841, 1734, 1651, 1622; MS: found 575.23; calcd for $C_{33}H_{20}ClN_2O_6$ $(M + H)^+$ 575.10; EA: found C 68.89, H 3.46, Cl 6.11, N 4.80; calcd for $C_{33}H_{19}ClN_2O_6$ (574.97): C 68.94, H 3.33, Cl 6.17, N 4.87.

### 3′-(4-Bromobenzoyl)-4′-hydroxy-5-phenyl-1′-(4-tolyl)-2*H*-spiro[furo[3,2-*c*]quinoline-3,2′-pyrrole]-2,4,5′(1′*H*,5*H*)-trione (11ic)

Yield 362 mg (67%), mp 272–274 °C (dec.), colorless crystals. $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 7.85 (d, $J$ = 8.0 Hz, 1H, Ar), 7.76 (d, $J$ = 8.3 Hz, 2H, Ar), 7.60–7.47 (m, 3H, Ar), 7.42–7.36 (m, 3H, Ar), 7.28–7.11 (m, 6H, Ar), 7.05 (d, $J$ = 7.0 Hz, 1H, Ar), 6.69 (d, $J$ = 8.7 Hz, 1H, Ar), 2.33 (s, 3H, Me); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 188.2, 177.1, 165.7, 161.7, 157.6, 151.1, 142.9, 139.8, 136.5, 135.8, 133.0, 131.8, 131.3, 130.7, 130.4, 130.4, 129.9, 129.5, 129.3, 128.8, 128.5, 127.8, 123.7, 123.1, 120.1, 117.1, 116.3, 110.3, 105.8, 70.5, 21.4; IR (NaCl, $cm^{-1}$): 3449, 1836, 1735, 1660, 1616; MS: found 633.15; calcd for $C_{34}H_{21}BrN_2O_6$ $(M + H)^+$ 633.07; EA: found C 64.36, H 3.39, Br 12.43, N 4.37; calcd for $C_{34}H_{21}BrN_2O_6$ (633.45): C 64.47, H 3.34, Br 12.61, N 4.42.

**3′-Cinnamoyl-4′-hydroxy-1′-(4-tolyl)-2*H*,4*H*-spiro[furo[3,2-*c*]chromene-3,2′-pyrrole]-2,4,5′(1′*H*)-trione (13aa)**

Yield 339 mg (67%), mp 236–237 °C (dec.), colorless crystals. $^1$H NMR (400 MHz, DMSO-$d_6$) $\delta$ 11.25 (br. s, 1H, OH), 7.88–7.83 (m, 2H, Ar + CH=CH), 7.75–7.57 (m, 5H, Ar + CH=CH), 7.55–7.39 (m, 4H, Ar + CH=CH), 7.25 (d, $J$ = 7.3 Hz, 2H, Ar + CH=CH), 7.02 (d, $J$ = 7.4 Hz, 2H, Ar + CH=CH), 2.24 (s, 3H, Me); $^{13}$C NMR (100 MHz, DMSO-$d_6$) $\delta$ 182.0, 170.9, 165.8, 163.5, 157.6, 155.2, 154.8, 143.1, 139.1, 135.4, 133.4, 131.2, 130.8, 130.6, 129.1, 128.5, 126.6, 125.6, 123.3, 117.5, 115.6, 109.3, 101.8, 67.9, 20.6; IR (NaCl, cm$^{-1}$): 3420, 1843, 1716, 1670, 1642; MS: found 506.25; calcd for $C_{30}H_{20}NO_7$ $(M + H)^+$ 506.12; EA: found C 71.36, H 3.73, N 2.79; calcd for $C_{30}H_{19}NO_7$ (505.48): C 71.28, H 3.79, N 2.77.

**3′-Cinnamoyl-4′-hydroxy-5-methyl-1′-(4-tolyl)-2*H*-spiro[furo[3,2-*c*]quinoline-3,2′-pyrrole]-2,4,5′(1′*H*,5*H*)-trione (13ab)**

Yield 389 mg (75%), mp 276–277 °C (dec.), colorless crystals. $^1$H NMR (400 MHz, DMSO-$d_6$) $\delta$ 7.83–7.75 (m, 2H, Ar + CH=CH), 7.67–7.60 (m, 5H, Ar + CH=CH), 7.49–7.34 (m, 4H, Ar + CH=CH), 7.18 (d, $J$ = 8.1 Hz, 2H, Ar + CH=CH), 7.04–6.99 (m, 2H, Ar + CH=CH), 3.59 (s, 3H, Me), 2.21 (s, 3H, $C_6H_4Me$); $^{13}$C NMR (100 MHz, DMSO-$d_6$) $\delta$ 181.9, 171.8, 165.6, 158.9, 156.5, 156.5, 142.9, 140.9, 138.6, 134.2, 133.4, 131.5, 130.7, 130.1, 129.0, 128.3, 126.5, 123.4, 122.8, 122.7, 116.4, 115.9, 109.0, 106.1, 68.7, 28.8, 20.4; IR (NaCl, cm$^{-1}$): 3437, 1841, 1728, 1667, 1639; MS: found 519.35; calcd for $C_{31}H_{23}N_2O_6$ $(M + H)^+$ 519.16; EA: found C 71.76, H 4.21, N 5.43; calcd for $C_{31}H_{22}N_2O_6$ (518.53): C 71.81, H 4.28, N 5.40.

**3′-Cinnamoyl-4′-hydroxy-1′-(4-methoxyphenyl)-5-methyl-2*H*-spiro[furo[3,2-*c*]quinoline-3,2′-pyrrole]-2,4,5′(1′*H*,5*H*)-trione (13bb)**

Yield 417 mg (78%), mp 297–298 °C (dec.), colorless crystals. $^1$H NMR (400 MHz, DMSO-$d_6$) $\delta$ 7.83–7.75 (m, 2H, Ar + CH=CH), 7.74–7.51 (m, 5H, Ar + CH=CH), 7.49–7.36 (m, 4H, Ar + CH=CH), 7.07–7.00 (m, 2H, Ar + CH=CH), 6.97–6.92 (m, 2H, Ar + CH=CH), 6.31 (br. s, 1H, OH), 3.68 (s, 3H, OMe), 3.60 (s, 3H, Me); $^{13}$C NMR (100 MHz, DMSO-$d_6$) $\delta$ 181.9, 171.8, 165.8, 159.3, 159.0, 156.7, 156.6, 142.9, 140.9, 134.2, 133.4, 130.7, 129.0, 128.3, 126.4, 123.4, 122.8, 122.7, 116.4, 116.0, 114.9, 109.1, 106.1, 68.9, 55.2, 28.8; IR (NaCl, cm$^{-1}$): 3456, 1839, 1728, 1662, 1639; MS: found 535.28; calcd for $C_{31}H_{23}N_2O_7$ $(M + H)^+$ 535.15; EA: found C 69.78, H 4.02, N 5.23; calcd for $C_{31}H_{22}N_2O_7$ (534.52): C 69.66, H 4.15, N 5.24.

**4′-Hydroxy-1′-(4-methoxyphenyl)-3′-(3-(4-methoxyphenyl)acryloyl)-5-methyl-2*H*-spiro[furo[3,2-*c*]quinoline-3,2′-pyrrole]-2,4,5′(1′*H*,5*H*)-trione (13cb)**

Yield 435 mg (77%), mp 281–282 °C (dec.), colorless crystals. $^1$H NMR (400 MHz, DMSO-$d_6$) $\delta$ 7.83–7.75 (m, 2H, Ar + CH=CH), 7.68–7.48 (m, 5H, Ar + CH=CH), 7.42–7.38 (m, 1H, Ar + CH=CH), 7.08–6.98 (m, 4H, Ar + CH=CH), 6.97–6.92 (m, 2H, Ar), 3.81 (s, 3H, OMe), 3.68 (s, 3H, OMe), 3.60 (s, 3H, Me); $^{13}$C NMR (100 MHz, DMSO-$d_6$) $\delta$ 181.9, 171.8, 165.8, 161.5, 159.3, 159.2, 156.6, 155.9, 143.0, 140.9, 133.4, 130.3, 128.3, 126.8, 126.4, 122.8, 122.7, 120.9, 116.8, 115.9, 114.9, 114.6, 109.1, 106.1, 68.9, 55.3, 55.2, 28.8; IR (NaCl, cm$^{-1}$): 3431, 1839, 1730, 1658, 1629; MS: found 565.27; calcd for $C_{32}H_{25}N_2O_8$ $(M + H)^+$ 565.16; EA: found C 67.99, H 4.32, N 4.94; calcd for $C_{32}H_{24}N_2O_8$ (564.55): C 68.08, H 4.29, N 4.96.

**3′-Cinnamoyl-4′-hydroxy-1′-(4-methoxyphenyl)-5-phenyl-2*H*-spiro[furo[3,2-*c*]quinoline-3,2′-pyrrole]-2,4,5′(1′*H*,5*H*)-trione (13bc)**

Yield 388 mg (65%), mp 290–291 °C (dec.), colorless crystals. $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 7.88 (d, $J$ = 7.9 Hz, 1H, Ar + CH=CH), 7.75–7.26 (m, 12H, Ar + CH=CH), 7.25–7.21 (m, 3H, Ar + CH=CH), 7.07 (d, $J$ = 7.4 Hz, 1H, Ar + CH=CH), 6.84 (d, $J$ = 8.9 Hz, 2H, Ar + CH=CH), 6.71 (d, $J$ = 8.7 Hz, 1H, Ar + CH=CH), 3.75 (s, 3H, OMe); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 183.4, 171.2, 166.0, 161.5, 160.3, 157.7, 144.7, 142.9, 136.7, 134.8, 132.9, 130.9, 130.4, 129.5, 129.3, 129.1, 129.1, 129.0, 128.9, 126.6, 123.7, 123.7, 123.1, 117.9, 117.0, 115.2, 110.4, 105.4, 70.1, 55.6; IR (NaCl, cm$^{-1}$): 3423, 1839, 1716, 1667, 1639; MS: found 597.28; calcd for $C_{36}H_{25}N_2O_7$ $(M + H)^+$ 597.17; EA: found C 72.39, H 4.01, N 4.76; calcd for $C_{36}H_{24}N_2O_7$ (596.60): C 72.48, H 4.06, N 4.70.

**4′-Hydroxy-3′-(3-(4-methoxyphenyl)acryloyl)-5-phenyl-1′-(4-tolyl)-2*H*-spiro[furo[3,2-*c*]quinoline-3,2′-pyrrole]-2,4,5′(1′*H*,5*H*)-trione (13dc)**

Yield 440 mg (62%), mp 272–273 °C (dec.), colorless crystals. $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 7.88 (dd, $J$ = 8.0, 1.3 Hz, 1H, Ar + CH=CH), 7.72–7.36 (m, 8H, Ar + CH=CH), 7.29–7.09 (m, 6H, Ar + CH=CH), 7.05 (d, $J$ = 7.7 Hz, 1H, Ar + CH=CH), 6.87 (d, $J$ = 8.8 Hz, 2H, Ar + CH=CH), 6.70 (d, $J$ = 8.6 Hz, 1H, Ar + CH=CH), 3.83 (s, 3H, OMe), 2.29 (s, 3H, Me); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 183.2, 171.2, 166.0, 162.2, 161.4, 157.7, 144.7, 142.9, 139.6, 136.7, 132.8, 131.5, 130.8, 130.5, 130.4, 130.4, 129.4, 129.3, 128.9, 127.5, 127.4, 123.7, 123.0, 121.3, 117.0, 114.7, 110.4, 106.5, 70.0, 55.6, 21.3; IR (NaCl, cm$^{-1}$): 3443, 1846, 1737, 1662, 1637; MS: found 611.33; calcd for $C_{37}H_{27}N_2O_7$ $(M + H)^+$ 611.18; EA: found C 72.71, H 4.25, N 4.63; calcd for $C_{37}H_{26}N_2O_7$ (610.62): C 72.78, H 4.29, N 4.59.

**Methyl 1′-benzyl-4′-hydroxy-2,4,5′-trioxo-1′,5′-dihydro-2*H*,4*H*-spiro[furo[3,2-*c*]chromene-3,2′-pyrrole]-3′-carboxylate (17aa)**

Yield 359 mg (83%), mp 222–224 °C (dec.), colorless crystals. $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 8.70 (br. s, 1H, OH), 7.74–7.65 (m, 2H, Ar), 7.45–7.33 (m, 2H, Ar), 7.14–7.06 (m, 5H, Ar), 4.49 (dd, $J$ = 4.53, 4.70 Hz, 2H, $CH_2$), 3.72 (s, 3H, OMe); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 168.9, 164.1, 164.0, 163.0, 157.8, 155.9, 155.1, 135.0, 133.0, 129.2, 128.8, 128.7, 125.1, 123.4, 117.7, 110.2, 108.2, 101.5, 65.6, 52.8, 46.0; IR (NaCl, cm$^{-1}$): 3202, 1843, 1714, 1678, 1662, 1650; MS: found 434.15; calcd for $C_{23}H_{16}NO_8$ $(M + H)^+$ 434.09; EA: found C 63.71, H 3.46, N 3.28; calcd for: $C_{23}H_{15}NO_8$ (433.37): C 63.75, H 3.49, N 3.23.

**Methyl 4′-hydroxy-2,4,5′-trioxo-1′-phenyl-1′,5′-dihydro-2*H*,4*H*-spiro[furo[3,2-*c*]chromene-3,2′-pyrrole]-3′-carboxylate (17ba)**

Yield 352 mg (84%), mp 206–208 °C (dec.), colorless crystals. $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 8.62 (br. s, 1H, OH), 7.72–7.65 (m, 2H, Ar), 7.43–7.27 (m, 5H, Ar), 7.24–7.19 (m, 2H, Ar), 3.79 (s, 3H, OMe); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 169.5, 164.7, 163.4, 163.1, 157.6, 156.0, 155.6, 135.1, 133.8, 130.2, 129.7, 127.6, 125.2, 123.6, 117.9, 110.1, 108.0, 101.1, 68.1, 52.9; IR (NaCl, $cm^{-1}$): 3279, 1837, 1738, 1716, 1645; MS: found 420.14; calcd for $C_{22}H_{14}NO_8$ $(M + H)^+$ 420.07; EA: found C 62.91, H 3.06, N 3.38; calcd for $C_{22}H_{13}NO_8$ (419.35): C 63.01; H 3.12; N 3.34.

**Methyl 4′-hydroxy-2,4,5′-trioxo-1′-(4-tolyl)-1′,5′-dihydro-2*H*,4*H*-spiro[furo[3,2-*c*]chromene-3,2′-pyrrole]-3′-carboxylate (17ca)**

Yield 351 mg (81%), mp 210–212 °C (dec.), colorless crystals. $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 7.71–7.64 (m, 2H, Ar), 7.43–7.33 (m, 2H, Ar), 7.17–7.07 (m, 4H, Ar), 6.30 (wide s, 1H, OH), 3.77 (s, 3H, OMe), 2.27 (s, 3H, Me); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 169.6, 164.7, 163.7, 163.1, 157.4, 156.0, 155.6, 140.0, 135.0, 130.9, 130.8, 127.4, 125.1, 123.7, 117.9, 110.1, 107.9, 101.2, 68.3, 52.8, 21.3; IR (NaCl, $cm^{-1}$): 3148, 1852, 1743, 1722, 1692, 1652; MS: found 434.18; calcd for $C_{23}H_{16}NO_8$ $(M + H)^+$ 434.09; EA: found C 63.71, H 3.46, N 3.28; calcd for $C_{23}H_{15}NO_8$ (433.37): C 63.75, H 3.49, N 3.23.

**Methyl 1′-benzyl-4′-hydroxy-2,4,5′-trioxo-5-phenyl-1′,4,5,5′-tetrahydro-2*H*-spiro[furo[3,2-*c*]quinoline-3,2′-pyrrole]-3′-carboxylate (17ac)**

Yield 401 mg (79%), mp 244–246 °C (dec.), colorless crystals. $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 8.72 (br. s, 1H, OH), 7.80 (dd, $J$ = 8.0, 1.4 Hz, 1H, Ar), 7.62–7.44 (m, 4H, Ar), 7.31 (td, $J$ = 7.7, 0.9 Hz, 1H, Ar), 7.22–7.05 (m, 7H, Ar), 6.74 (d, $J$ = 8.6 Hz, 1H, Ar), 4.57 (s, 2H, $CH_2$), 3.70 (s, 3H, OMe); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 170.3, 164.3, 163.2, 160.9, 157.7, 157.2, 142.9, 136.5, 134.2, 133.1, 130.4, 129.4, 129.5, 129.3, 129.1, 129.0, 128.6, 128.3, 123.3, 123.1, 117.0, 110.1, 108.6, 106.1, 66.6, 52.6, 45.9; IR (NaCl, $cm^{-1}$): 3448, 1835, 1732, 1712, 1697, 1651, 1632; MS: found 509.28; calcd for $C_{29}H_{21}N_2O_7$ $(M + H)^+$ 509.13; EA: found C 68.43, H 3.93, N 5.54; calcd for $C_{29}H_{20}N_2O_7$ (508.49): C 68.50, H 3.96, N 5.51.

**Methyl 4′-hydroxy-2,4,5′-trioxo-1′,5-diphenyl-1′,4,5,5′-tetrahydro-2*H*-spiro[furo[3,2-*c*]quinoline-3,2′-pyrrole]-3′-carboxylate (17bc)**

Yield 341 mg (69%), mp 255–256 °C (dec.), colorless crystals. $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 8.77 (br. s, 1H, OH), 7.81 (ddd, $J$ = 8.0, 1.5, 0.5 Hz, 1H, Ar), 7.63–7.49 (m, 3H, Ar), 7.45 (ddd, $J$ = 8.8, 6.6, 3.0 Hz, 1H, Ar), 7.36–7.23 (m, 7H, Ar), 7.03 (ddd, $J$ = 6.4, 2.9, 1.5 Hz, 1H, Ar), 6.72 (d, $J$ = 7.3 Hz, 1H, Ar), 3.77 (s, 3H, OMe); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 170.9, 163.7, 163.3, 161.2, 157.6, 157.4, 143.0, 136.5, 134.0, 133.2, 130.5, 130.5, 129.8, 129.5, 129.3, 129.2, 128.8, 127.7, 123.5, 123.2, 117.1, 110.0, 108.4, 105.9, 69.0, 52.7; IR (NaCl, $cm^{-1}$): 3183, 1836, 1741, 1713, 1671, 1650, 1639; MS: found 495.22; calcd for $C_{28}H_{19}N_2O_7$ $(M + H)^+$ 495.12; EA: found C 68.09, H 3.63, N 5.74; calcd for $C_{28}H_{18}N_2O_7$ (494.46): C 68.02, H 3.67, N 5.67.

**Methyl 4′-hydroxy-2,4,5′-trioxo-5-phenyl-1′-(4-tolyl)-1′,4,5,5′-tetrahydro-2*H*-spiro[furo[3,2-*c*]quinoline-3,2′-pyrrole]-3′-carboxylate (17cc)**

Yield 325 mg (64%), mp 259–261 °C (dec.), colorless crystals. $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 8.74 (wide s, 1H, OH), 7.81 (dd, $J$ = 8.0, 1.3 Hz, 1H, Ar), 7.65–7.49 (m, 3H, Ar), 7.45 (ddd, $J$ = 8.7, 7.2, 1.6 Hz, 1H, Ar), 7.34–7.22 (m, 2H, Ar), 7.18–7.01 (m, 5H, Ar), 6.74 (d, $J$ = 8.6 Hz, 1H, Ar), 3.76 (s, 3H, OMe), 2.28 (s, 3H, Me); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 170.9, 163.8, 163.3, 161.1, 157.7, 157.5, 143.0, 139.5, 136.6, 133.1, 131.6, 130.5, 130.5, 129.6, 129.2, 128.8, 127.6, 123.5, 123.1, 117.1, 110.1, 108.3, 106.0, 69.0, 52.6, 21.3; IR: $\nu$ = 3188, 1840, 1733, 1708, 1650, 1639; MS: found 509.25; calcd for $C_{29}H_{21}N_2O_7$ $(M + H)^+$ 509.13; EA: found C 68.59, H 3.91, N 5.53; calcd for $C_{29}H_{20}N_2O_7$ (508.49): C 68.50, H 3.96, N 5.51.

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