One-Pot Synthesis of Dispiro[oxindole-3,3′-pyrrolidines] by Three-Component [3 + 2] Cycloadditions of *in situ*-Generated Azomethine Ylides with 3-Benzylidene-2,3-dihydro-1*H*-indol-2-ones

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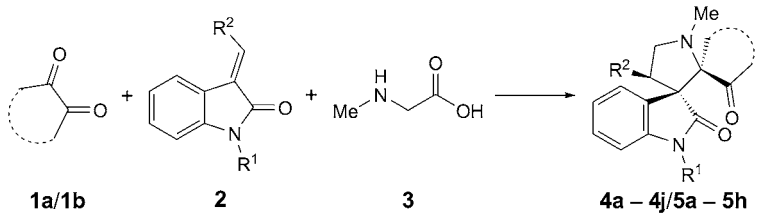
An efficient one-pot, three-component synthesis of novel dispiro[oxindole-3,3′-pyrrolidines] by 1,3-dipolar cycloaddition of azomethine ylides, *in situ* generated by reaction of 1,2-diones with sarcosine and subsequent decarboxylation, with a series of (*E*)-3-benzylidene-2,3-dihydro-1*H*-indol-2-ones is reported. Molecular complexity is generated in only one synthetic step. All reactions proceed with excellent regioselectivity and in good-to-excellent yields. The workup is easy, the reaction times are short, and no catalyst is required.

Introduction. – The design and synthesis of natural products, and their analogs is one of the main challenges in organic and medicinal chemistry [1]. Among the strategies used for the construction of novel and complex compounds, multicomponent reactions are very attractive, because they proceed with high efficiency[1]). Multicomponent reactions have some advantages over conventional linear syntheses, including a minimum number of synthetic steps, shorter reaction times, high degrees of atom economy, and environmental friendliness. In fact, they allow for the preparation of diverse structures in a rapid and cost-effective manner [3]. The [3 + 2] cycloaddition reaction is one of the simplest and most powerful approaches for the construction of various types of complex polyheterocyclic frameworks [4]. In recent years, azomethine ylides have attained a vital place in the synthesis of heterocyclic compounds for the construction of N-containing five-membered heterocycles [5]. Among the scaffolds of natural products, the spiro[indol-pyrrolidin]one framework has attracted growing interest, since it is a core structure in many natural alkaloids and pharmaceuticals. The characteristic structural feature of these compounds is the spiro-ring at C(3) of the oxindole core with varying substitutions around the pyrrolidine and oxindole rings [6]. For example, the naturally occurring spiro[oxindole-3,3′-pyrrolidine] alkaloids, such as

[1]) For selected examples of multicomponent reactions, see [2].

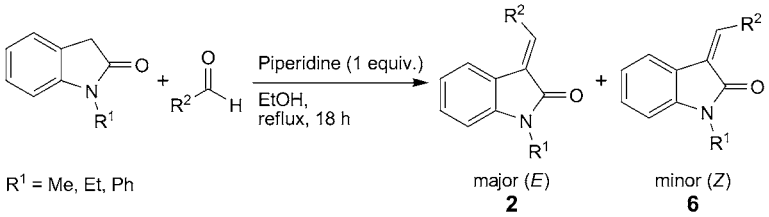
horsfiline, spiro-tryprostatine A and B, and elacomine [7], are well-known for their wide biological applications as antimicrobial, and antitumoral, antibiotic agents, and inhibitors of human NK-1 receptor [8]. All these justify the efforts to develop novel syntheses of spiro- and dispiro-heterocycles containing the spiro[oxindole-3,3′-pyrrolidine] core. In all previously reported syntheses, the presence of electron-withdrawing groups, such as C=O or CN groups, proved to be important [3b][5c][6e][9]. As a continuation of our research devoted to the development of cycloaddition reactions [10] and to the synthesis of nitrogen heterocycles [11], herein, we report an efficient, highly atom-economic, and regioselective synthesis of novel dispiro[oxindole-3,3′-pyrrolidines] based on a one-pot, three-component reaction of cycloketones **1**, sarcosine (=2-(methylamino)acetic acid; **3**), and (*E*)-3-benzylidene-2,3-dihydro-1*H*-indol-2-ones **2** (*Scheme 1*).

Scheme 1. *Synthesis of 3,3′-Dispiro[indole-pyrrolidin]one Derivatives* **4a**–**4j** *and* **5a**–**5h**



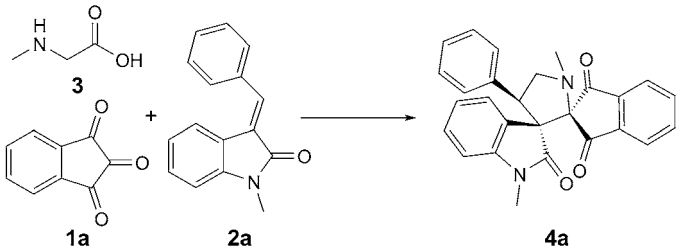
Results and Discussion. – (*E*)-3-Benzylidene-2,3-dihydro-1*H*-indol-2-ones **2** were prepared according to a reported procedure [12] *via* condensation of 2,3-dihydro-1*H*-indol-2-ones and various aromatic aldehydes with the predominant formation of the (*E*)-configured isomers (*Scheme 2*). The geometric isomers were separated by column chromatography to give pure (*E*)- and (*Z*)-3-benzylidene-2,3-dihydro-1*H*-indol-2-ones in good yields. The structures of the isomers were elucidated by comparison of their ^{1}H-NMR spectral data with those reported [13].

Scheme 2. *Synthesis of* (E)*- and* (Z)*-3-Benzylidene-2,3-diyhdro-1*H*-indol-2-ones*



At the beginning of our investigations, we examined the three-component reaction of 1*H*-indene-1,2,3-trione (**1a**), (*E*)-3-benzylidene-2,3-dihydro-1*H*-indol-2-one (**2a**) and sarcosine (**3**) under different conditions in the absence of a catalyst. Among the solvents used, MeOH was found to be the best in terms of yields and short reaction times (*Table 1*, *Entry 7*). The azomethine ylide, generated from **1a** and **3**, underwent

Table 1. *Effect of Solvents on the Yield of Product* **4a**



Entry	Solvent	Temp. [°]	Time [h]	Yield[a]) [%]
1	MeCN	80	3	50
2	Toluene	100	5	14
3	Benzene	80	5	25
4	1,4-Dioxane	100	5	37
5	1,4-Dioxane	Reflux	5	50
6	MeOH	20	5	10
7	MeOH	Reflux	1.5	85
8	MeOH	Reflux	8	72
9	EtOH	Reflux	3	60

[a]) Yield of isolated products.

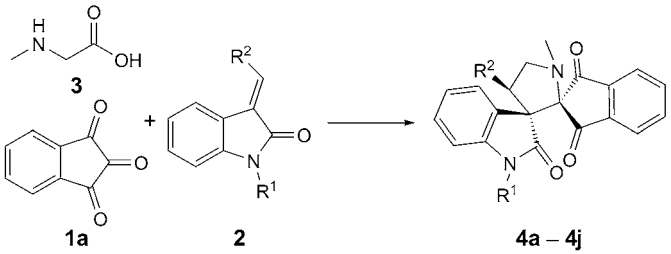
readily a [3 + 2] cycloaddition reaction with **2a** to give dispiro[oxindole-3,3′-pyrrolidine] **4a** in 85% yield with excellent regio- and stereoselectivity. During this reaction, the product directly precipitated from the reaction mixture. Only a single regioisomer was formed.

The scope of our three-component reaction was examined using different (*E*)-3-benzylidene-2,3-dihydro-1*H*-indol-2-ones **2** which contain substituents in various positions. All reactions went to completion in 1.5 h and afforded the dispiro[indene-2,2′-pyrrolidine-3′,3″-indole]-1,2″,3(1″*H*)-triones **4a**–**4j** in good-to-excellent yields (*Table 2*).

To further demonstrate the utility of this reaction, the reactions of acenaphthylene-1,2-dione (**1b**) and sarcosine (**3**) with different (*E*)-3-benzylidene-2,3-dihydro-1*H*-indol-2-ones **2** were also studied (*Table 3*). These reactions, carried out under similar conditions, proceeded cleanly to give the corresponding polycyclic dispiro[acenaphthylene-1,2′-pyrrolidine-3′,3″-indole]-2,2″(1″*H*)-diones **5a**–**5h** in 74–87% yields (*Table 3*). All reactions proceeded with high regioselectivity.

The structures of all the compounds **4a**–**4j** and **5a**–**5h** were confirmed by spectroscopic methods. For instance, the IR spectrum of cycloadduct **5c** showed two characteristic bands at 1716 and 1703 cm^{-1} assigned to the CO groups. In the ^{1}H-NMR spectrum of **5c**, three *singlet*s appeared at δ(H) 2.35, 2.84, and 3.74 for two MeN groups and a MeO group, respectively. The benzylic H-atoms and CH_2N H-atoms of the pyrrolidine ring appeared as three *triplet*s at δ(H) 3.88, 4.57, and 4.65. In the ^{13}C-NMR spectrum of **5c**, the two spiro C-atoms resonated at 65.9 and 82.3 ppm, and the oxindole and acenaphthylen-2-one CO C-atoms at 178.6 and 205.6 ppm, respectively. Finally, the

Table 2. *Syntheses of Compounds* **4a**–**4j**



Entry	R^1	R^2	**4**	Yield[a]) [%]
1	Me	Ph	**4a**	85
2	Me	4-Me–C_6H_4	**4b**	90
3	Me	4-MeO–C_6H_4	**4c**	78
4	Me	4-Cl–C_6H_4	**4d**	82
5	Me	3-NO_2–C_6H_4	**4e**	87
6	Et	Ph	**4f**	83
7	Et	4-Me–C_6H_4	**4g**	86
8	Et	4-MeO–C_6H_4	**4h**	88
9	Et	4-Cl–C_6H_4	**4i**	85
10	Ph	Furan-2-yl	**4j**	75

[a]) Yields of the isolated products.

structures of the products **4c** and **5c** were confirmed by X-ray crystal-structure analysis of (*Figs. 1* and *2*, resp.).

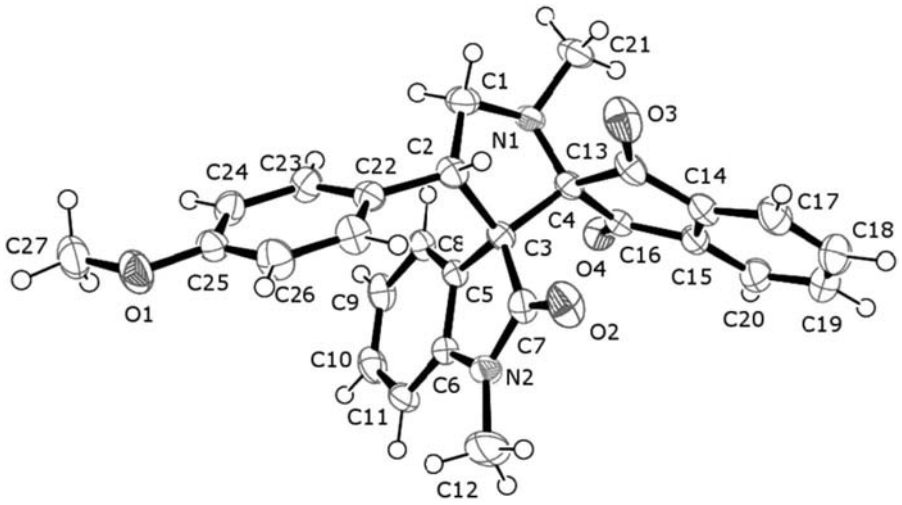
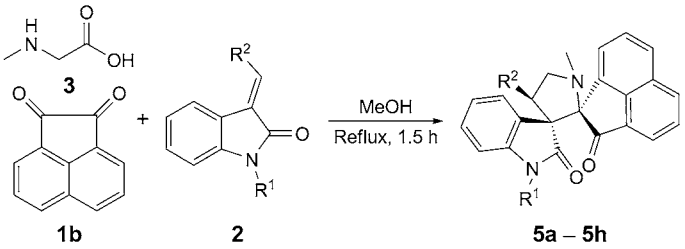


Fig. 1. *X-Ray crystal structure of* **4c**

Table 3. *Synthesis of 3,3′-Dispiro[indole-pyrrolidin]one Derivatives* **5a** – **5h**



1b **2** **5a** – **5h**

Entry	R^1	R^2	**5**	Yield[a]) [%]
1	Me	Ph	**5a**	80
2	Me	4-Me–C_6H_4	**5b**	87
3	Me	4-MeO–C_6H_4	**5c**	76
4	Me	4-Cl–C_6H_4	**5d**	74
5	Me	3-NO_2–C_6H_4	**5e**	80
6	Et	Ph	**5f**	78
7	Et	4-Me–C_6H_4	**5g**	77
8	Et	Furan-2-yl	**5h**	78

[a]) Yields of the isolated products.

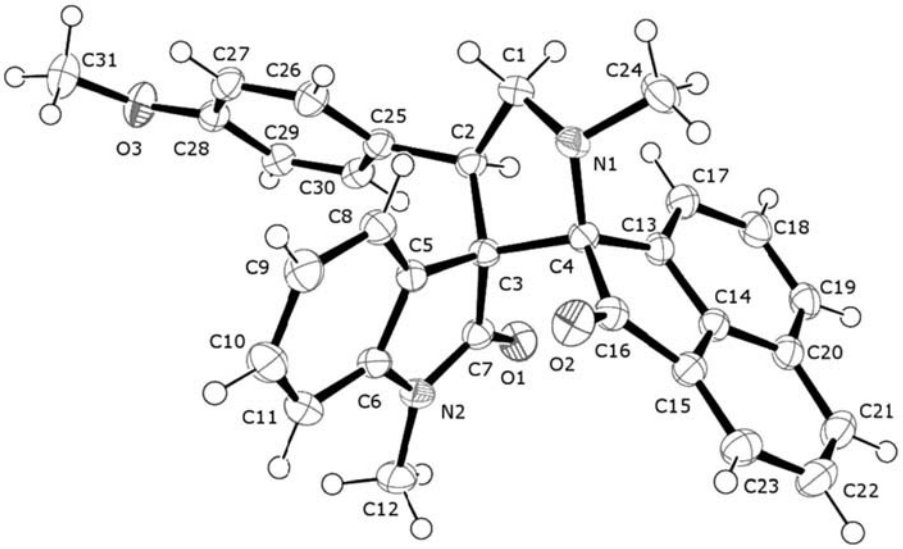
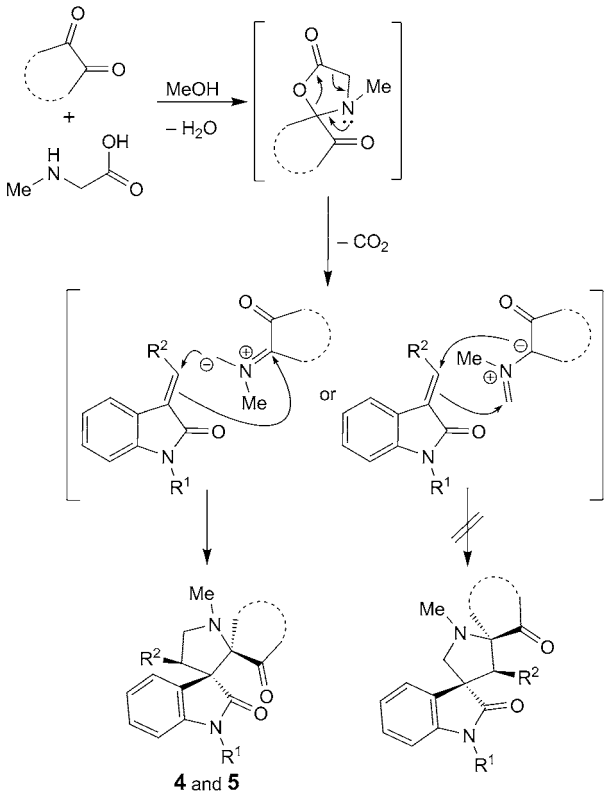


Fig. 2. *X-Ray crystal structure of* **5c**

A possible mechanism for the formation of spiro-substituted 3,3′-spiropyrrolidine oxindoles **4** and **5** is proposed in *Scheme 3*. The mechanism involves the formation of an oxazolidinone intermediate [14]. In the presence of (*E*)-3-benzylidene-2,3-dihydro-

1*H*-indol-2-ones, it undergoes loss of CO_2 *via* a stereospecific 1,3-cycloreversion to form an azomethine ylide, which undergoes a 1,3-dipolar cycloaddition to give the dispiro compounds **4** and **5** (*Scheme 3*).

Scheme 3. *Proposed Mechanism of the Formation of Compounds* **4**–**5**



Conclusions. – In conclusion, we have described an efficient synthesis of spiro-substituted spiro[oxindole-3,3′-pyrrolidines] by one-pot, three-component reaction of azomethine ylides with different (*E*)-3-benzylidene-2,3-dihydro-1*H*-indol-2-ones. This reaction offers many advantages, such as operational simplicity, high atom efficiency, good-to-excellent yields in short reaction times, easy workup, and catalyst-free conditions (environmental friendliness, because no transition metals are needed). The reactions proceed with excellent regio- and stereoselectivity.

Experimental Part

General. Commercially available materials were purchased from *Sigma–Aldrich* and *Merck,* and were used without any additional purification. TLC: Silica-gel plates *60 F_{254}* (SiO_2; *Merck*). M.p.*: Büchi* melting-point *B-540* apparatus; in sealed capillaries; uncorrected. IR Spectra: *ATR* apparatus. ^{1}H- and

^{13}C-NMR Spectra: *Bruker* (*DRX-500 Avance*) spectrometer at 500 (^{1}H) and 125 (^{13}C) MHz, in $CDCl_3$ soln., at r.t.; δ in ppm rel. to Me_4Si as internal standard, J in Hz, signals of the ^{13}C-NMR spectra corresponding to CH, CH_2, or Me groups are assigned from DEPT. MS: Electrospray ionization (ESI) on *Finnigan MAT 95-XP*.

General Procedure for the Synthesis of the Dispiro[oxindole-3,3′-pyrrolidines] **4a**–**4j** *and* **5a**–**5h**. A mixture of cycloketone **1** (1.2 mmol), (*E*)-3-benzylidene-2,3-dihydro-1*H*-indol-2-one **2** (1 mmol), and sarcosine (**3**; 98 mg, 1.2 mmol) in 7 ml of MeOH was stirred at reflux temp. for 1.5 h. After completion of the reaction (monitored by TLC), the mixture was cooled, ice-cold H_2O (20 ml) was added to the mixture, the solid precipitate was filtered, washed with H_2O, and air-dried. Then, the product was crystallized from MeOH to afford the pure products.

*(3′*S*,4′*R*)-1′,1″-Dimethyl-4′-phenyldispiro[indene-2,2′-pyrrolidine-3′,3″-indole]-1,2″,3(1″*H*)-trione* (**4a**). Yield: 85%. Pale-yellow solid. M.p. 209–211°. IR (KBr): 2852*w*, 1760*w*, 1740*m*, 1699*s*, 1610*m*, 1467*m*, 1345*m*, 1236*m*, 1093*m*, 850*m*, 767*s*, 757*s*, 709*s*, 691*m*, 624*m*, 585*m*, 539*m*. ^{1}H-NMR: 8.09 (*d*, J = 7.6, 1 arom. H); 7.97–7.99 (*m*, 1 arom. H); 7.89 (*t*, J = 7.0, 1 arom. H); 7.74 (*t*, J = 7.5, 1 arom. H); 7.69 (*d*, J = 10.5, 1 arom. H); 7.15–7.17 (*m*, 2 arom. H); 7.00–7.01 (*m*, 3 arom. H); 6.94–6.96 (*m*, 2 arom. H); 6.40–6.42 (*m*, 1 arom. H); 4.86 (*t*, J = 9.3, H–C(4′)); 4.06 (*t*, J = 9.2, 1 H of CH_2); 3.81 (*t*, J = 9.7, 1 H of CH_2); 2.97 (*s*, MeN); 2.55 (*s*, MeN). ^{13}C-NMR: 200.8 (C=O); 197.1 (C=O); 173.3 (N–C=O); 142.9 (C); 141.6 (C); 141.3 (C); 137.0 (CH); 135.6 (CH); 135.4 (C); 129.0 (CH); 128.9 (CH); 128.8 (CH); 127.9 (CH); 127.5 (CH); 125.5 (C); 123.4 (CH); 123.3 (CH); 122.4 (CH); 108.0 (CH); 80.7 (C); 67.2 (C); 56.1 (CH_2N); 52.7 (CH); 36.1 (MeN); 26.3 (MeN). GC/MS (70 eV): 422 (9, M^+), 290 (10), 235 (100), 234 (56), 206 (10), 187 (26), 158 (32), 133 (11). HR-ESI-MS: 423.1705 ($C_{27}H_{23}N_2O_3^+$, $[M+H]^+$; calc. 423.1703).

*(3′*S*,4′*R*)-1′,1″-Dimethyl-4′-(4-methylphenyl)dispiro[indene-2,2′-pyrrolidine-3′,3″-indole]-1,2″,3(1″*H*)-trione* (**4b**). Yield: 90%. Yellow solid. M.p. 179–180°. IR (KBr): 2941*w*, 2850*w*, 1760*w*, 1739*m*, 1703*s*, 1699*s*, 1610*m*, 1468*m*, 1350*m*, 1234*m*, 1094*m*, 825*m*, 766*s*, 755*s*, 692*m*, 619*m*, 592*m*, 540*m*. ^{1}H-NMR: 8.05 (*d*, J = 7.7, 1 arom. H); 7.94 (*d*, J = 8.5, 1 arom. H); 7.85 (*t*, J = 7.3, 1 arom. H); 7.71 (*t*, J = 7.5, 1 arom. H); 7.66 (*d*, J = 7.5, 1 arom. H); 7.11–7.16 (*m*, 2 arom. H); 6.76–6.80 (*m*, 4 arom. H); 6.40 (*d*, J = 8.5, 1 arom. H); 4.76 (*t*, J = 9.5, H–C(4′)); 3.98 (*t*, J = 9.2, 1 H of CH_2); 3.75 (*t*, J = 9.6, 1 H of CH_2); 2.70 (*s*, MeN); 2.51 (*s*, MeN); 2.16 (*s*, Me). ^{13}C-NMR: 200.4 (C=O); 196.7 (C=O); 173.3 (N–C=O); 143.0 (C); 141.7 (C); 141.3 (C); 140.7 (CH); 140.6 (CH); 136.9 (C); 136.8 (CH); 135.3 (CH); 132.2 (C); 128.9 (CH); 128.7 (CH); 125.7 (C); 123.5 (CH); 123.2 (CH); 122.3 (CH); 107.9 (CH); 80.7 (C); 67.1 (C); 56.4 (CH_2N); 52.4 (CH); 36.1 (MeN); 26.2 (MeN); 21.4 (Me). GC/MS (70 eV): 436 (5, M^+), 290 (5), 249 (100), 248 (55), 220 (8), 187 (16), 158 (23). HR-ESI-MS: 437.1864 ($C_{28}H_{25}N_2O_3^+$, $[M+H]^+$; calc. 437.1860).

*(3′*S*,4′*R*)-4′-(4-Methoxyphenyl)-1′,1″-dimethyldispiro[indene-2,2′-pyrrolidine-3′,3″-indole]-1,2″,3(1″*H*)-trione* (**4c**). Yield: 78%. Yellow solid. M.p. 178–180°. IR (KBr): 2934*w*, 1765*w*, 1739*w*, 1705*s*, 1699*s*, 1610*m*, 1514*m*, 1469*m*, 1349*m*, 1244*s*, 1178*m*, 831*m*, 764*s*, 753*s*, 692*m*, 618*m*, 591*m*, 541*m*. ^{1}H-NMR: 8.08 (*d*, J = 7.7, 1 arom. H); 8.00–8.02 (*m*, 1 arom. H); 7.88 (*t*, J = 7.9, 1 arom. H); 7.74 (*t*, J = 7.6, 1 arom. H); 7.71 (*t*, J = 8.7, 1 arom. H); 7.16–7.20 (*m*, 2 arom. H); 6.86 (*d*, J = 8.7, 2 arom. H); 6.54 (*d*, J = 8.7, 2 arom. H); 6.43–6.46 (*m*, 1 arom. H); 4.80 (*t*, J = 9.6, H–C(4′)); 4.01 (*t*, J = 9.3, 1 H of CH_2); 3.78 (*t*, J = 9.6, 1 H of CH_2); 3.68 (*s*, MeO); 2.71 (*s*, MeN); 2.55 (*s*, MeN). ^{13}C-NMR: 200.6 (C=O); 196.9 (C=O); 173.3 (N–C=O); 159.0 (C); 143.0 (C); 141.6 (C); 141.3 (C); 137.0 (CH); 135.6 (CH); 129.8 (CH); 129.0 (CH); 128.8 (CH); 127.1 (C); 125.5 (C); 123.5 (CH); 123.3 (CH); 122.4 (CH); 113.4 (CH); 108.1 (CH), 80.7 (C); 67.2 (C); 56.5 (CH_2); 55.4 (MeO); 52.2 (CH); 36.2 (MeN); 26.3 (MeN). GC/MS (70 eV): 452 (M^+, 4), 290 (5), 265 (100), 264 (36), 222 (20), 187 (16), 158 (19). HR-ESI-MS: 453.1809 ($C_{28}H_{25}N_2O_4^+$, $[M+H]^+$; calc. 453.1809).

*(3′*S*,4′*R*)-4′-(4-Chlorophenyl)-1′,1″-dimethyldispiro[indene-2,2′-pyrrolidine-3′,3″-indole]-1,2″,3(1″*H*)-trione* (**4d**). Yield: 82%. Pale-yellow solid. M.p. 203–205°. IR (KBr): 2873*w*, 1766*w*, 1738*w*, 1708*s*, 1699*s*, 1610*m*, 1491*m*, 1346*m*, 1240*m*, 1090*m*, 812*m*, 782*m*, 752*m*, 692*m*, 616*m*, 587*m*, 541*m*. ^{1}H-NMR: 8.03 (*d*, J = 7.7, 1 arom. H); 7.92 (*d*, J = 7.1, 1 arom. H); 7.84 (*t*, J = 7.7, 1 arom. H); 7.71 (*t*, J = 7.8, 1 arom. H); 7.65 (*d*, J = 7.6, 1 arom. H); 7.10–7.16 (*m*, 2 arom. H); 6.94 (*d*, J = 8.5, 2 arom. H); 6.84 (*d*, J = 8.5, 2 arom. H); 6.42 (*d*, J = 8.5, 1 arom. H); 4.76 (*t*, J = 9.5, H–C(4′)); 3.93 (*t*, J = 9.4, 1 H of CH_2); 3.76 (*t*, J = 9.5, 1 H of CH_2); 2.69 (*s*, MeN); 2.49 (*s*, MeN). ^{13}C-NMR: 200.3 (C=O); 196.4 (C=O); 173.0 (N–C=O); 142.9 (C); 141.6 (C); 141.3 (C); 136.9 (CH); 135.5 (CH); 133.9 (C); 133.5 (C); 130.1 (CH);

129.2 (CH); 128.9 (CH); 128.1 (CH); 125.2 (C); 123.5 (CH); 123.3 (CH); 122.4 (CH); 108.1 (CH); 80.5 (C); 66.8 (C); 56.2 (CH_2); 52.0 (CH); 36.0 (MeN); 26.2 (MeN). GC/MS (70 eV): 456 (11, M^+), 290 (22), 271 (33), 270 (31), 269 (100), 268 (43), 241 (13), 204 (14), 187 (39), 167 (14), 159 (18), 158 (47), 117 (13). HR-ESI-MS: 457.1323 ($C_{27}H_{22}ClN_2O_3^+$, $[M+H]^+$; calc. 457.1314).

*(3′*S*,4′*R*)-1′,1″-Dimethyl-4′-(3-nitrophenyl)dispiro[indene-2,2′-pyrrolidine-3′,3″-indole]-1,2″,3(1″*H*)-trione* (**4e**). Yield: 87%. Pale-brown solid. M.p. 180–182°. IR (KBr): 2866*w*, 1758*w*, 1740*m*, 1705*s*, 1610*m*, 1527*s*, 1467*m*, 1347*s*, 1092*m*, 782*m*, 759*s*, 740*m*, 690*s*, 614*m*. ^{1}H-NMR: 8.08 (*d*, *J* = 7.7, 1 arom. H); 7.99 (*d*, *J* = 6.8, 1 arom. H); 7.88–7.91 (*m*, 2 arom. H); 7.80 (*s*, 1 arom. H); 7.76 (*t*, *J* = 7.3, 1 arom. H); 6.70 (*d*, *J* = 7.6, 1 arom. H); 7.28 (*d*, *J* = 7.7, 1 arom. H); 7.13–7.21 (*m*, 3 arom. H); 6.42 (*d*, *J* = 7.2, 1 arom. H); 4.92 (*t*, *J* = 9.3, H–C(4′)); 4.04 (*t*, *J* = 9.3, 1 H of CH_2); 3.88 (*t*, *J* = 9.4, 1 H of CH_2); 2.74 (*s*, MeN); 2.54 (*s*, MeN). ^{13}C-NMR: 200.3 (C=O); 196.0 (C=O); 172.8 (N–C=O); 148.0 (C); 142.7 (C); 141.5 (C); 141.3 (C); 138.0 (C); 137.0 (CH); 135.6 (CH); 134.9 (CH); 129.5 (CH); 128.9 (CH); 128.8 (CH); 124.7 (C); 123.5 (CH); 123.4 (CH); 123.3 (CH); 122.8 (CH); 122.6 (CH); 108.2 (CH); 80.1 (C); 66.6 (C); 55.9 (CH_2); 52.0 (CH); 35.9 (MeN); 26.3 (MeN). GC/MS (70 eV): 467 (10, M^+), 290 (15), 281 (18), 280 (100), 250 (17), 234 (19), 233 (26), 219 (16), 205 (13), 204 (20), 190 (14), 187 (46), 172 (13), 159 (20), 158 (43), 104 (18), 76 (15). HR-ESI-MS: 468.1562 ($C_{27}H_{22}N_3O_5^+$, $[M+H]^+$; calc. 468.1554).

*(3′*S*,4′*R*)-1″-Ethyl-1′-methyl-4′-phenyldispiro[indene-2,2′-pyrrolidine-3′,3″-indole]-1,2″,3(1″*H*)-trione* (**4f**). Yield: 83%. Yellow solid. M.p. 214–216°. IR (KBr): 2969*w*, 1764*w*, 1738*m*, 1703*s*, 1699*s*, 1612*m*, 1463*m*, 1368*m*, 1217*m*, 779*m*, 758*s*, 695*s*, 626*m*, 587*m*. ^{1}H-NMR: 7.91 (*d*, *J* = 7.5, 1 arom. H); 7.77–7.81 (*m*, 2 arom. H); 7.64 (*t*, *J* = 7.5, 1 arom. H); 7.53 (*d*, *J* = 7.5, 1 arom. H); 6.97–7.02 (*m*, 2 arom. H); 6.84–6.89 (*m*, 3 arom. H); 6.80–6.82 (*m*, 2 arom. H); 6.34 (*d*, *J* = 7.4, 1 arom. H); 4.63 (*t*, *J* = 9.5, H–C(4′)); 3.92 (*t*, *J* = 9.5, 1 H of CH_2); 3.68 (*t*, *J* = 9.5, 1 H of CH_2); 3.16 (*q*, *J* = 7.5, NCH_2); 2.41 (*s*, MeN); 0.51 (*t*, *J* = 7.0, Me). ^{13}C-NMR: 200.3 (C=O); 196.5 (C=O); 172.4 (N–C=O); 141.9 (C); 141.7 (C); 141.3 (C); 136.8 (CH); 135.4 (CH); 135.3 (CH); 133.8 (C); 129.1 (CH); 129.0 (CH); 128.0 (CH); 127.5 (CH); 125.8 (C); 123.5 (CH); 123.1 (CH); 122.1 (CH); 108.0 (CH); 81.2 (C); 67.0 (C); 56.4 (CH_2); 52.3 (CH); 36.1 (MeN); 34.7 (CH_2); 12.3 (Me). GC/MS (70 eV): 436 (14, M^+), 304 (21), 249 (100), 248 (21), 234 (45), 206 (29), 187 (25), 172 (14), 133 (22). HR-ESI-MS: 437.1863 ($C_{28}H_{25}N_2O_3^+$, $[M+H]^+$; calc. 437.1860).

*(3′*S*,4′*R*)-1″-Ethyl-1′-methyl-4′-(4-methylphenyl)dispiro[indene-2,2′-pyrrolidine-3′,3″-indole]-1,2″,3-(1″*H*)-trione* (**4g**). Yield: 86%. Yellow solid. M.p. 170–172°. IR (KBr): 2973*w*, 1765*w*, 1738*w*, 1698*s*, 1607*m*, 1593*m*, 1463*m*, 1365*m*, 1349*m*, 1212*m*, 1100*m*, 817*m*, 773*m*, 756*s*, 688*m*, 619*m*, 592*m*. ^{1}H-NMR: 8.03 (*d*, *J* = 7.7, 1 arom. H); 7.91 (*d*, *J* = 6.9, 1 arom. H); 7.83 (*t*, *J* = 7.3, 1 arom. H); 7.67 (*t*, *J* = 7.5, 1 arom. H); 7.62 (*d*, *J* = 7.6, 1 arom. H); 7.09–7.14 (*m*, 2 arom. H); 6.81 (*d*, *J* = 8.0, 2 arom. H); 6.76 (*d*, *J* = 7.9, 2 arom. H); 6.42 (*d*, *J* = 7.3, 1 arom. H); 4.75 (*t*, *J* = 9.5, H–C(4′)); 3.98 (*t*, *J* = 9.2, 1 H of CH_2); 3.76 (*t*, *J* = 9.2, 1 H of CH_2); 3.24 (*q*, *J* = 7.2, NCH_2); 2.52 (*s*, MeN); 2.13 (*s*, Me); 0.60 (*t*, *J* = 7.1, Me). ^{13}C-NMR: 200.2 (C=O); 196.5 (C=O); 172.8 (N–C=O); 142.0 (C); 141.7 (C); 141.3 (C); 136.8 (C); 136.7 (CH); 135.2 (CH); 132.4 (C); 129.1 (CH); 129.0 (CH); 128.9 (CH); 128.7 (CH); 125.9 (C); 123.5 (CH); 123.0 (CH); 122.1 (CH); 108.0 (CH); 81.4 (C); 66.9 (C); 56.6 (CH_2); 51.9 (CH); 36.1 (MeN); 34.7 (CH_2); 21.4 (Me); 12.4 (Me). GC/MS (70 eV): 450 (8, M^+), 304 (11), 263 (100), 262 (19), 248 (30), 220 (21), 187 (20), 172 (11), 147 (14). HR-ESI-MS: 451.2012 ($C_{29}H_{27}N_2O_3^+$, $[M+H]^+$; calc. 451.2016).

*(3′*S*,4′*R*)-1″-Ethyl-4′-(4-methoxyphenyl)-1′-methyldispiro[indene-2,2′-pyrrolidine-3′,3″-indole]-1,2″,3(1″*H*)-trione* (**4h**). Yield: 88%. Lemon-yellow solid. M.p. 180–182°. IR (KBr): 2946*w*, 1765*w*, 1741*w*, 1705*s*, 1699*s*, 1609*m*, 1515*m*, 1462*m*, 1366*m*, 1254*m*, 1022*m*, 832*m*, 772*s*, 752*m*, 742*s*, 688*m*, 619*m*, 594*m*, 528*m*. ^{1}H-NMR: 8.03 (*d*, *J* = 7.7, 1 arom. H); 7.92 (*d*, *J* = 6.5, 1 arom. H); 7.84 (*t*, *J* = 7.4, 1 arom. H); 7.68 (*t*, *J* = 7.5, 1 arom. H); 7.63 (*d*, *J* = 7.5, 1 arom. H); 7.10–7.15 (*m*, 2 arom. H); 6.84 (*d*, *J* = 8.5, 2 arom. H); 6.49 (*d*, *J* = 8.5, 2 arom. H); 6.42 (*d*, *J* = 7.5, 1 arom. H); 4.73 (*t*, *J* = 9.5, H–C(4′)); 3.94 (*t*, *J* = 9.2, 1 H of CH_2); 3.76 (*t*, *J* = 9.5, 1 H of CH_2); 3.62 (*s*, MeO); 3.24 (*q*, *J* = 9.5, NCH_2); 2.51 (*s*, MeN); 0.61 (*t*, *J* = 7.1, Me). ^{13}C-NMR: 200.3 (C=O); 196.5 (C=O); 172.8 (N–C=O); 159.0 (C); 142.0 (C); 141.7 (C); 141.3 (C); 136.7 (CH); 135.2 (CH); 130.0 (CH); 129.1 (CH); 129.0 (CH); 127.3 (C); 125.9 (C); 123.5 (CH); 123.1 (CH); 122.1 (CH); 113.4 (CH); 108.0 (CH); 81.3 (C); 67.0 (C); 56.7 (CH_2); 55.3 (MeO); 51.8 (CH); 36.1 (MeN); 34.6 (CH_2); 12.4 (Me). GC/MS (70 eV): 466 (4, M^+), 280 (18), 279 (100), 264 (35), 236 (22), 187 (17), 172 (11), 163 (11). HR-ESI-MS: 467.1975 ($C_{29}H_{27}N_2O_4^+$, $[M+H]^+$; calc. 467.1965).

*(3′*S*,4′*R*)-4′-(4-Chlorophenyl)-1″-ethyl-1′-methyldispiro[indene-2,2′-pyrrolidine-3′,3″-indole]-1,2″,3-(1″*H*)-trione* (**4i**). Yield: 85%. Yellow solid. M.p. 198–200°. IR (KBr): 2869*w*, 1767*w*, 1742*w*, 1703*s*,

1613*m*, 1486*m*, 1465*s*, 1366*m*, 1345*m*, 1239*s*, 1207*m*, 1091*s*, 1013*m*, 839*m*, 812*m*, 760*m*, 740*s*, 728*s*, 693*m*, 614*m*, 588*m*. ^{1}H-NMR: 8.01 (*d*, *J* = 7.1, 1 arom. H); 7.89 (*d*, *J* = 6.9, 1 arom. H); 7.84 (*t*, *J* = 7.3, 1 arom. H); 7.68 (*t*, *J* = 7.4, 1 arom. H); 7.62 (*d*, *J* = 7.5, 1 arom. H); 7.09–7.16 (*m*, 2 arom. H); 6.93 (*d*, *J* = 8.2, 2 arom. H); 6.86 (*d*, *J* = 8.2, 2 arom. H); 6.45 (*d*, *J* = 7.3, 1 arom. H); 4.75 (*t*, *J* = 9.4, H–C(4′)); 3.96 (*t*, *J* = 9.2, 1 H of CH_2); 3.78 (*t*, *J* = 9.6, 1 H of CH_2); 3.25 (*q*, *J* = 7.1, NCH_2); 2.51 (*s*, MeN); 0.61 (*t*, *J* = 7.1, Me). ^{13}C-NMR: 200.0 (C=O); 196.1 (C=O); 172.6 (N–C=O); 141.8 (C); 141.6 (C); 141.2 (C); 136.8 (C); 135.4 (CH); 133.9 (CH); 133.5 (C); 130.3 (CH); 129.3 (CH); 129.0 (CH); 128.2 (CH); 125.4 (C); 123.6 (CH); 123.1 (CH); 122.3 (CH); 108.3 (CH); 81.1 (C); 66.7 (C); 56.5 (CH_2); 51.5 (CH); 36.1 (MeN); 34.8 (CH_2); 12.4 (Me). GC/MS (70 eV): 470 (9, M^+), 304 (20), 285 (29), 284 (19), 283 (100), 270 (13), 268 (43), 240 (20), 204 (16), 187 (42), 172 (16). HR-ESI-MS: 471.1475 ($C_{28}H_{24}ClN_2O_3^+$, $[M+H]^+$; calc. 471.1475).

*(3′*S*,4′*R*)-4′-(Furan-2-yl)-1′-methyl-1″-phenyldispiro[indene-2,2′-pyrrolidine-3′,3″-indole]-1,2″,3(1″*H*)-trione* (**4j**). Yield: 75%. Yellow solid. M.p. 155–157°. IR (KBr): 2863*w*, 1759*w*, 1739*m*, 1704*s*, 1612*m*, 1594*m*, 1496*m*, 1465*m*, 1370*m*, 1232*m*, 1176*m*, 768*m*, 757*s*, 739*s*, 697*s*, 611*m*. ^{1}H-NMR: 7.99 (*d*, *J* = 7.5, 1 arom. H); 7.7–7.83 (*m*, 2 arom. H); 7.72–7.73 (*m*, 2 arom. H); 7.32 (*t*, *J* = 7.3, 2 arom. H); 7.26 (*d*, *J* = 6.9, 1 arom. H); 7.08–7.09 (*m*, 2 arom. H); 7.02 (*s*, 1 arom. H); 6.83 (*d*, *J* = 7.5, 2 arom. H); 6.49–6.50 (*m*, 1 arom. H); 6.03 (*s*, 1 arom. H); 5.89 (*s*, 1 arom. H); 4.90 (*t*, *J* = 9.2, H–C(4′)); 3.98 (*t*, *J* = 9.2, 1 H of CH_2); 3.89 (*t*, *J* = 9.3, 1 H of CH_2), 2.53 (*s*, MeN). ^{13}C-NMR: 200.0 (C=O); 196.0 (C=O); 172.6 (N–C=O); 151.2 (C); 143.1 (C); 142.0 (CH); 141.7 (C); 141.4 (C); 136.9 (CH); 135.5 (CH); 134.5 (C); 129.8 (CH); 129.0 (CH); 128.8 (CH); 128.4 (CH); 126.6 (CH); 125.7 (C); 123.8 (CH); 123.1 (CH); 122.9 (CH); 110.3 (CH); 109.1 (CH); 107.4 (CH); 80.9 (C); 65.4 (C); 55.2 (CH_2); 46.2 (CH); 36.0 (MeN). GC/MS (70 eV): 474 (2, M^+), 288 (17), 287 (100), 259 (9), 230 (20), 187 (15). HR-ESI-MS: 475.1650 ($C_{30}H_{23}N_2O_4^+$, $[M+H]^+$; calc. 475.1652).

*(1*R*,3′*S*,4′*R*)-1′,1″-Dimethyl-4′-phenyl-2*H*-dispiro[acenaphthylene-1,2′-pyrrolidine-3′,3″-indole]-2,2″(1″*H*)-dione* (**5a**). Yield: 80%. Pale-yellow solid. M.p. 203–205°. IR (KBr): 3047*w*, 2867*w*, 1713*s*, 1612*m*, 1597*m*, 1491*m*, 1469*m*, 1344*m*, 1091*m*, 789*m*, 771*m*, 750*m*, 740*s*, 707*s*, 539*m*. ^{1}H-NMR: 7.93 (*d*, *J* = 8.0, 1 arom. H); 7.90 (*d*, *J* = 6.8, 1 arom. H); 7.80 (*d*, *J* = 8.2, 1 arom. H); 7.68 (*t*, *J* = 7.6, 1 arom. H); 7.58 (*d*, *J* = 6.8, 1 arom. H); 7.51 (*t*, *J* = 7.7, 1 arom. H); 7.32 (*d*, *J* = 7.4, 2 arom. H); 7.19 (*t*, *J* = 7.3, 2 arom. H); 7.11 (*t*, *J* = 7.1, 1 arom. H); 6.80 (*t*, *J* = 7.5, 1 arom. H); 6.74 (*d*, *J* = 7.5, 1 arom. H); 6.53 (*t*, *J* = 7.5, 1 arom. H); 6.23 (*d*, *J* = 7.6, 1 arom. H); 4.64–4.72 (*m*, CH_2); 3.90 (*t*, *J* = 7.8, H–C(4′)); 2.85 (*s*, MeN); 2.36 (*s*, MeN). ^{13}C-NMR: 200.8 (C=O); 178.6 (N–C=O); 143.4 (C); 142.8 (C); 141.4 (C); 138.1 (C); 132.3 (C); 131.7 (CH); 130.5 (C); 129.8 (CH); 129.4 (CH); 128.8 (CH); 128.4 (CH); 128.1 (CH); 128.0 (CH); 127.2 (CH); 125.6 (CH); 124.9 (C); 123.3 (CH); 121.6 (CH); 120.4 (CH); 107.2 (CH); 82.3 (C); 65.9 (C); 57.8 (CH_2); 50.3 (CH); 35.8 (MeN); 26.5 (MeN). GC/MS (70 eV): 444 (2, M^+), 430 (12), 429 (39), 415 (7), 235 (18), 234 (13), 210 (14), 209 (100), 194 (25), 158 (9), 133 (13). HR-ESI-MS: 445.1910 ($C_{30}H_{25}N_2O_2^+$, $[M+H]^+$; calc. 445.1911).

*(1*R*,3′*S*,4′*R*)-1′,1″-Dimethyl-4′-(4-methylphenyl)-2*H*-dispiro[acenaphthylene-1,2′-pyrrolidine-3′,3″-indole]-2,2″(1″*H*)-dione* (**5b**). Yield: 87%. Pale-yellow solid. M.p. 190–192°. IR (KBr): 3045*w*, 2941*w*, 1711*s*, 1610*m*, 1491*m*, 1469*m*, 1344*m*, 1093*m*, 1010*m*, 831*m*, 792*s*, 742*s*, 538*m*. ^{1}H-NMR: 7.87 (*d*, *J* = 7.8, 1 arom. H); 7.83 (*d*, *J* = 6.8, 1 arom. H); 7.75 (*d*, *J* = 8.1, 1 arom. H); 7.64 (*t*, *J* = 7.5, 1 arom. H); 7.52 (*d*, *J* = 6.6, 1 arom. H); 7.46 (*t*, *J* = 7.5, 1 arom. H); 7.16 (*d*, *J* = 7.3, 2 arom. H); 6.95 (*d*, *J* = 7.3, 2 arom. H); 6.75 (*t*, *J* = 6.3, 2 arom. H); 6.51 (*t*, *J* = 7.4, 1 arom. H); 6.17 (*d*, *J* = 7.5, 1 arom. H); 4.55–4.62 (*m*, CH_2); 3.81 (*t*, *J* = 7.0, H–C(4′)); 2.81 (*s*, MeN); 2.31 (*s*, MeN); 2.22 (*s*, Me). ^{13}C-NMR: 205.9 (C=O); 178.5 (N–C=O); 143.8 (C); 142.7 (C); 136.4 (C); 135.8 (C); 135.1 (C); 132.4 (C); 131.4 (CH); 130.5 (C); 129.7 (CH); 129.5 (CH); 129.0 (CH); 128.7 (CH); 128.0 (CH); 127.9 (CH); 125.5 (CH); 125.0 (C); 123.2 (CH); 121.6 (CH); 120.2 (CH); 107.1 (CH); 82.3 (C); 65.9 (C); 58.0 (CH_2); 50.0 (CH); 35.7 (MeN); 26.4 (MeN); 21,4 (Me). GC/MS (70 eV): 458 (2, M^+), 444 (13), 443 (46), 429 (7), 249 (23), 248 (15), 210 (19), 209 (100), 194 (23), 147 (13). HR-ESI-MS: 459.2065 ($C_{31}H_{27}N_2O_2^+$, $[M+H]^+$; calc. 459.2067).

*(1*R*,3′*S*,4′*R*)-4′-(4-Methoxyphenyl)-1′,1″-dimethyl-2*H*-dispiro[acenaphthylene-1,2′-pyrrolidine-3′,3″-indole]-2,2″(1″*H*)-dione* (**5c**). Yield: 76%. Yellow solid. M.p. 199–201°. IR (KBr): 2940*w*, 1716*m*, 1703*m*, 1609*m*, 1512*m*, 1468*m*, 1341*m*, 1252*m*, 1183*m*, 1094*m*, 1011*m*, 839*m*, 788*s*, 752*m*, 744*m*, 598*m*, 538*m*. ^{1}H-NMR: 7.93 (*d*, *J* = 8.0, 1 arom. H); 7.89 (*d*, *J* = 6.6, 1 arom. H); 7.80 (*d*, *J* = 8.3, 1 arom. H); 7.68 (*t*, *J* = 7.6, 1 arom. H); 7.58 (*d*, *J* = 6.9, 1 arom. H); 7.51 (*t*, *J* = 7.7, 1 arom. H); 7.25 (*d*, *J* = 8.5, 2 arom. H); 6.79–6.83 (*m*, 2 arom. H); 6.73 (*d*, *J* = 8.6, 2 arom. H); 6.57 (*t*, *J* = 7.6, 1 arom. H); 6.24 (*d*, *J* = 7.6, 1

arom. H); 4.65 (*t*, *J* = 8.9, 1 H of CH_2); 4.57 (*t*, *J* = 9.2, 1 H of CH_2); 3.88 (*t*, *J* = 7.3, H–C(4′)); 3.74 (*s*, MeO); 2.84 (*s*, MeN); 2.35 (*s*, MeN). ^{13}C-NMR: 205.6 (C=O); 178.6 (N–C=O); 158.7 (C); 143.8 (C); 142.7 (C); 132.3 (C); 131.7 (CH); 130.8 (CH); 130.5 (C); 130.2 (C); 130.1 (C); 129.5 (CH); 128.8 (CH); 128.1 (CH); 128.0 (CH); 125.6 (CH); 125.0 (C); 123.3 (CH); 121.7 (CH); 120.4 (CH); 113.8 (CH); 107.3 (CH); 82.3 (C); 65.9 (C); 58.2 (CH_2); 55.6 (MeO); 49.8 (CH); 35.8 (MeN); 26.5 (MeN). GC/MS (70 eV): 474 (2, M^+), 459 (17), 266 (16), 265 (100), 264 (39), 250 (9), 222 (23), 210 (13), 209 (73), 196 (11), 194 (27), 167 (17), 166 (17), 158 (21), 152 (13), 139 (14). HR-ESI-MS: 475.2027 ($C_{31}H_{27}N_2O_3^+$, $[M+H]^+$; calc. 475.2016).

*(1*R,*3′*S,*4′*R*)-4′-(4-Chlorophenyl)-1′,1″-dimethyl-2*H*-dispiro[acenaphthylene-1,2′-pyrrolidine-3′,3″-indole]-2,2″(1″*H*)-dione* (**5d**). Yield: 74%. Yellow solid. M.p. 177–179°. IR (KBr): 3048*w*, 2872*w*, 1711*s*, 1612*m*, 1600*m*, 1492*m*, 1469*m*, 1344*m*, 1093*m*, 1012*m*, 837*m*, 788*s*, 740*s*, 539*m*. ^{1}H-NMR: 7.86 (*d*, *J* = 7.9, 1 arom. H); 7.81 (*d*, *J* = 6.9, 1 arom. H); 7.74 (*d*, *J* = 8.1, 1 arom. H); 7.62 (*t*, *J* = 7.5, 1 arom. H); 7.52 (*d*, *J* = 6.7, 1 arom. H); 7.45 (*t*, *J* = 7.4, 1 arom. H); 7.22 (*d*, *J* = 8.0, 2 arom. H); 7.11 (*d*, *J* = 8.0, 2 arom. H); 6.75 (*t*, *J* = 7.5, 1 arom. H); 6.63 (*d*, *J* = 7.5, 1 arom. H); 6.51 (*t*, *J* = 7.5, 1 arom. H); 6.18 (*d*, *J* = 7.6, 1 arom. H); 4.50–4.58 (*m*, CH_2); 3.83 (*t*, *J* = 7.8, H–C(4′)); 2.82 (*s*, MeN); 2.29 (*s*, MeN). ^{13}C-NMR: 206.1 (C=O); 178.4 (N–C=O); 143.8 (C); 142.8 (C); 136.9 (C); 135.4 (C); 132.9 (C); 132.3 (C); 131.6 (CH); 131.2 (CH); 130.5 (C); 129.3 (CH); 128.7 (CH); 128.5 (CH); 128.3 (CH); 128.0 (CH); 125.6 (CH); 124.5 (C); 123.1 (CH); 121.8 (CH); 120.3 (CH); 107.3 (CH); 82.1 (C); 65.6 (C); 58.0 (CH_2); 49.7 (CH); 35.6 (MeN); 26.4 (MeN). GC/MS (70 eV): 478 (2, M^+), 265 (12), 264 (11), 263 (37), 271 (21), 270 (22), 269 (77), 268 (35), 241 (10), 240 (10), 210 (21), 209 (100), 194 (33), 167 (23), 166 (13), 158 (33), 139 (10). HR-ESI-MS: 479.1529 ($C_{30}H_{24}ClN_2O_2^+$, $[M+H]^+$; calc. 479.1521).

*(1*R,*3′*S,*4′*R*)-1′,1″-Dimethyl-4′-(3-nitrophenyl)-2*H*-dispiro[acenaphthylene-1,2′-pyrrolidine-3′,3″-indole]-2,2″(1″*H*)-dione* (**5e**). Yield: 80%. Pale-brown solid. M.p. 198–200°. IR (KBr): 2831*w*, 1706*s*, 1613*m*, 1525*s*, 1492*m*, 1470*m*, 1342*s*, 1087*m*, 828*m*, 779*s*, 752*s*, 742*m*, 719*m*, 698*m*, 539*m*. ^{1}H-NMR: 8.23 (*s*, 1 arom. H); 7.98 (*d*, *J* = 7.7, 1 arom. H); 7.92 (*d*, *J* = 7.8, 1 arom. H); 7.84 (*d*, *J* = 9.7, 1 arom. H); 7.80 (*d*, *J* = 8.7, 1 arom. H); 7.65 (*d*, *J* = 7.3, 2 arom. H); 7.58 (*d*, *J* = 6.7, 1 arom. H); 7.51 (*t*, *J* = 7.4, 1 arom. H); 7.36 (*t*, *J* = 7.7, 1 arom. H); 6.78 (*t*, *J* = 7.2, 1 arom. H); 6.54 (*d*, *J* = 7.4, 1 arom. H); 6.47 (*t*, *J* = 7.4, 1 arom. H); 6.25 (*d*, *J* = 7.6, 1 arom. H); 4.62–4.66 (*m*, CH_2); 3.94 (*t*, *J* = 8.1, H–C(4′)); 2.90 (*s*, MeN); 2.33 (*s*, MeN). ^{13}C-NMR: 206.2 (C=O); 178.3 (N–C=O); 148.5 (C); 144.0 (C); 142.9 (C); 140.9 (C); 136.0 (CH); 135.1 (C); 132.1 (C); 131.7 (CH); 130.5 (C); 129.1 (2CH), 128.8 (CH); 128.6 (CH); 128.1 (CH); 125.8 (CH); 124.8 (CH); 124.0 (C); 123.2 (CH); 122.3 (CH); 121.9 (CH); 120.5 (CH); 107.6 (CH); 81.9 (C); 65.3 (C); 57.8 (CH_2); 49.9 (CH); 35.5 (MeN); 26.5 (MeN). GC/MS (70 eV): 489 (4, M^+), 475 (39), 474 (100), 260 (21), 312 (18), 311 (14), 281 (12), 280 (68), 233 (10), 210 (24), 209 (93), 194 (34), 178 (9), 158 (11). HR-ESI-MS: 490.1769 ($C_{30}H_{24}N_3O_4^+$, $[M+H]^+$; calc. 490.1761).

*(1*R,*3′*S,*4′*R*)-1″-Ethyl-1′-methyl-4′-phenyl-2*H*-dispiro[acenaphthylene-1,2′-pyrrolidine-3′,3″-indole]-2,2″(1″*H*)-dione* (**5f**). Yield: 78%. Yellow solid. M.p. 184–186°. IR (KBr): 2932*w*, 1703*s*, 1607*m*, 1491*m*, 1470*m*, 1361*m*, 1057*m*, 835*m*, 785*m*, 757*s*, 696*s*, 596*m*. ^{1}H-NMR: 7.83–7.87 (*m*, 2 arom. H); 7.72 (*d*, *J* = 8.0, 1 arom. H); 7.62 (*t*, *J* = 7.5, 1 arom. H); 7.43–7.49 (*m*, 2 arom. H); 7.20 (*d*, *J* = 7.6, 2 arom. H); 7.10 (*t*, *J* = 7.4, 2 arom. H); 7.02 (*t*, *J* = 7.3, 1 arom. H); 6.70–6.75 (*m*, 2 arom. H); 6.47 (*t*, *J* = 8.0, 1 arom. H); 6.18 (*d*, *J* = 7.5, 1 arom. H); 4.58–4.62 (*m*, CH_2); 3.78–3.82 (*m*, H–C(4′)); 3.57–3.61 (*m*, 1 H of CH_2); 3.20–3.24 (*m*, 1 H of CH_2); 2.31 (*s*, MeN); 0.71 (*t*, *J* = 7.5, Me). ^{13}C-NMR: 205.7 (C=O); 177.9 (N–C=O); 142.9 (C); 142.6 (C); 138.2 (C); 135.7 (C); 132.5 (C); 131.4 (CH); 130.5 (C); 129.8 (CH); 129.6 (CH); 128.8 (CH); 128.3 (CH); 128.0 (CH); 127.9 (CH); 127.1 (CH); 125.3 (CH); 125.2 (C); 123.6 (CH); 121.3 (CH); 120.2 (CH); 107.1 (CH); 82.2 (C); 65.6 (C); 57.7 (CH_2); 50.5 (CH); 35.7 (MeN); 34.7 (CH_2); 12.6 (Me). GC/MS (70 eV): 458 (1, M^+), 456 (14), 443 (10), 356 (20), 355 (100), 271 (16), 270 (11), 255 (13), 249 (9), 209 (20), 201 (11). HR-ESI-MS: 459.2071 ($C_{31}H_{27}N_2O_2^+$, $[M+H]^+$; calc. 459.2073).

*(1*R,*3′*S,*4′*R*)-1″-Ethyl-1′-methyl-4′-(4-methylphenyl)-2*H*-dispiro[acenaphthylene-1,2′-pyrrolidine-3′,3″-indole]-2,2″(1″*H*)-dione* (**5g**). Yield: 77%. Pale-brown solid. M.p. 173–175°. IR (KBr): 2981*w*, 2936*w*, 1727*m*, 1699*s*, 1611*m*, 1486*m*, 1466*m*, 1363*m*, 1188*m*, 832*m*, 823*m*, 786*s*, 748*s*, 550*m*. ^{1}H-NMR: 7.85–7.88 (*m*, 2 arom. H); 7.75 (*d*, *J* = 8.3, 1 arom. H); 7.63 (*t*, *J* = 7.4, 1 arom. H); 7.50 (*d*, *J* = 6.8, 1 arom. H); 7.46 (*t*, *J* = 7.7, 1 arom. H); 7.11 (*d*, *J* = 6.8, 2 arom. H); 6.93 (*d*, *J* = 8.0, 2 arom. H); 6.75–6.81 (*m*, 2 arom. H); 6.53 (*t*, *J* = 7.6, 1 arom. H); 6.21 (*d*, *J* = 7.7, 1 arom. H); 4.55–4.59 (*m*, CH_2); 3.79 (*t*, *J* = 7.1,

H–C(4′)); 3.55–3.63 (*m*, 1 H of CH_2); 3.19–3.26 (*m*, 1 H of CH_2); 2.33 (*s*, MeN); 2.21 (*s*, Me); 0.70 (*t*, *J* = 7.1, Me). ^{13}C-NMR: 205.7 (C=O); 177.9 (N–C=O); 145.6 (C); 142.9 (C); 142.6 (C); 136.4 (C); 135.8 (C); 135.1 (C); 132.5 (C); 131.4 (CH); 130.5 (C); 129.9 (CH); 129.5 (CH); 129.0 (CH); 128.8 (CH); 128.0 (CH); 127.9 (CH); 125.3 (CH); 123.6 (CH); 121.4 (CH); 120.2 (CH); 107.2 (CH); 82.3 (C); 65.7 (C); 57.9 (CH_2); 50.3 (CH); 35.8 (MeN); 34.7 (CH_2); 21.4 (Me); 12.6 (Me). GC/MS (70 eV): 472 (3, M^+), 458 (23), 457 (78), 443 (12), 264 (22), 263 (93), 262 (33), 248 (56), 234 (11), 220 (38), 210 (36), 209 (100), 205 (11), 204 (19), 196 (14), 195 (12), 194 (44), 182 (10), 167 (16), 166 (14), 147 (25), 139 (12). HR-ESI-MS: 473.2229 ($C_{32}H_{29}N_2O_2^+$, $[M+H]^+$; calc. 473.2229).

*(1*R*,3′*S*,4′*R*)-1″-Ethyl-4′-(furan-2-yl)-1′-methyl-2*H*-dispiro[acenaphthylene-1,2′-pyrrolidine-3′,3″-indole]-2,2″(1″*H*)-dione* (**5h**). Yield: 78%. Bold-yellow solid. M.p. 178–180°. IR (KBr): 2973*w*, 2937*w*, 1699*s*, 1607*m*, 1487*m*, 1468*m*, 1364*m*, 1354*m*, 1010*m*, 834*m*, 784*m*, 753*s*, 745*s*, 690*m*. ^{1}H-NMR: 7.90 (*d*, *J* = 7.8, 1 arom. H); 7.84 (*d*, *J* = 6.9, 1 arom. H); 7.77 (*d*, *J* = 8.4, 1 arom. H); 7.63 (*t*, *J* = 7.8, 1 arom. H); 7.55 (*d*, *J* = 6.8, 1 arom. H); 7.50 (*t*, *J* = 7.7, 1 arom. H); 7.05 (*d*, *J* = 7.4, 1 arom. H); 6.85 (*t*, *J* = 7.5, 1 arom. H); 6.77 (*d*, *J* = 7.5, 1 arom. H); 6.49–6.57 (*m*, 2 arom. H); 6.27–6.30 (*m*, 2 arom. H); 4.53 (*t*, *J* = 8.5, 1 H of CH_2); 4.45 (*t*, *J* = 8.6, 1 H of CH_2); 3.91 (*t*, *J* = 8.4, H–C(4′)); 3.69–3.76 (*m*, 1 H of CH_2N); 3.23–3.30 (*m*, 1 H of CH_2N); 2.32 (*s*, MeN); 0.80 (*t*, *J* = 7.1, Me). ^{13}C-NMR: 205.8 (C=O); 177.7 (N–C=O); 152.9 (C); 143.1 (C); 142.8 (C); 141.6 (CH); 132.3 (C); 131.4 (CH); 130.4 (CH); 128.8 (CH); 128.3 (CH); 128.1 (CH); 128.0 (CH); 126.8 (C); 125.5 (CH); 124.8 (C); 123.4 (CH); 121.7 (CH); 120.1 (CH); 110.3 (CH); 108.5 (CH); 107.3 (C); 81.8 (C); 65.3 (C); 57.4 (CH_2); 44.0 (CH); 35.5 (MeN); 34.7 (CH_2); 12.5 (Me). GC/MS (70 eV): 448 (2, M^+), 433 (28), 239 (10), 210 (14), 209 (100), 194 (19). HR-ESI-MS: 449.1872 ($C_{29}H_{25}N_2O_3^+$, $[M+H]^+$; calc. 449.1865).

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REFERENCES

[1] F. Balkenhohl, C. von dem Bussche-Hünnefeld, A. Lansky, C. Zechel, *Angew. Chem., Int. Ed.* **1996**, *35*, 2288.

[2] 'Multicomponent Reactions', Eds. J. Zhu, H. Bienaimé, Wiley-VCH, Weinheim, Germany, 2005; A. Dömling, I. Ugi, *Angew. Chem., Int. Ed.* **2000**, *39*, 3168; J. D. Sunderhaus, S. F. Martin, *Chem. – Eur. J.* **2009**, *15*, 1300.

[3] a) S. V. Ley, I. R. Baxendale, R. N. Bream, P. S. Jackson, A. G. Leach, D. A. Longbottom, M. Nesi, J. S. Scott, R. I. Storer, S. J. Taylor, *J. Chem. Soc., Perkin Trans. 1* **2000**, 3815; b) A. R. S. Babu, R. Raghunathan, *Tetrahedron Lett.* **2007**, *48*, 305.

[4] G. Pandey, P. Banerjee, S. R. Gadre, *Chem. Rev.* **2006**, *106*, 4484; I. Coldham, R. Hufton, *Chem. Rev.* **2005**, *105*, 2765; K. V. Gothelf, K. A. Jørgensen, *Chem. Rev.* **1998**, *98*, 863.

[5] a) R. Rajesh, R. Raghunathan, *Tetrahedron Lett.* **2010**, *51*, 5845; b) S. Kathiravan, E. Ramesh, R. Raghunathan, *Tetrahedron Lett.* **2009**, *50*, 2389; c) M. Ghandi, A. Taheri, A. Abbasi, *Tetrahedron* **2010**, *66*, 6744; d) A. R. S. Babu, R. Raghunathan, *Tetrahedron Lett.* **2007**, *48*, 6809.

[6] a) T.-H. Kang, K. Matsumoto, Y. Murakami, H. Takayama, M. Kitajima, N. Aimi, H. Watanabe, *Eur. J. Pharmacol.* **2002**, *444*, 39; b) J. Ma, S. M. Hecht, *Chem. Commun.* **2004**, 1190; c) T. Usui, M. Kondoh, C.-B. Cui, T. Mayumi, H. Osada, *Biochem. J.* **1998**, *333*, 543; d) M. M. Khafagy, A. H. F. A. El-Wahab, F. A. Eid, A. M. El-Agrody, *Farmaco* **2002**, *57*, 715; e) A. V. Velikorodov, O. Y. Poddubnyi, O. O. Krivosheev, O. L. Titova, *Russ. J. Org. Chem.* **2011**, *47*, 402.

[7] S. T. Hilton, T. C. T. Ho, G. Pljevaljcic, K. Jones, *Org. Lett.* **2000**, *2*, 2639.

[8] T. Okita, M. Isobe, *Tetrahedron* **1994**, *50*, 11143; M. J. Kornet, A. P. Thio, *J. Med. Chem.* **1976**, *19*, 892.

[9] N. V. Lakshmi, P. Thirumurugan, P. T. Perumal, *Tetrahedron Lett.* **2010**, *51*, 1064; M. Ghandi, A. Yari, S. J. T. Rezaei, A. Taheri, *Tetrahedron Lett.* **2009**, *50*, 4724; R. Prasanna, S. Purushothaman, R.

Raghunathan, *Tetrahedron Lett.* **2010**, *51*, 4538; P. Shanmugam, B. Viswambharan, K. Selvakumar, S. Madhavan, *Tetrahedron Lett.* **2008**, *49*, 2611; J. Liu, H. Sun, X. Liu, L. Ouyang, T. Kang, Y. Xie, X. Wang, *Tetrahedron Lett*. **2012**, *53*, 2336; R. Jain, K. Sharma, D. Kumar, *Tetrahedron Lett*. **2012**, *53*, 1993; G. Chen, H.-P. He, J. Ding, X.-J. Hao, *Heterocycl. Commun.* **2009**, *15*, 355; H. Liu, G. Dou, D. Shi, *J. Comb. Chem.* **2010**, *12*, 292.

[10] M. Kiamehr, F. M. Moghaddam, *Tetrahedron Lett.* **2009**, *50*, 6723; F. M. Moghaddam, M. Kiamehr, M. R. Khodabakhshi, Z. Mirjafary, S. Fathi, H. Saeidian, *Tetrahedron* **2010**, *66*, 8615; F. M. Moghaddam, M. Kiamehr, S. Taheri, Z. Mirjafary, *Helv. Chim. Acta* **2010**, *93*, 964; F. M. Moghaddam, L. Hojabri, S. Taheri, P. Irani, *J. Iran. Chem. Soc.* **2010**, *7*, 781; F. M. Moghaddam, S. Taheri, L. Hojabri, P. Pirani, S. Maktabian, *J. Iran. Chem. Soc.* **2011**, *8*, 265.

[11] F. M. Moghaddam, Z. Mirjafary, H. Saeidian, S. Taheri, M. Doulabi, M. Kiamehr, *Tetrahedron* **2010**, *66*, 134; F. M. Moghaddam, S. Taheri, Z. Mirjafary, H. Saeidian, M. Kiamehra, M. Tafazzolia, *Helv. Chim. Acta* **2011**, *94*, 142; F. M. Moghaddam, H. Saeidian, M. Kiamehr, Z. Mirjafary, S. Taheri, *Arkivoc* **2010**, *xi*, 91; F. M. Moghaddam, B. K. Foroushani, H. Saeidian, S. Nourian, Z. Mirjafary, M. Kiamehr, *Pure Appl. Chem.* **2011**, *83*, 709.

[12] W. Zhang, M.-L. Go, *Bioorg. Med. Chem.* **2009**, *17*, 2077.

[13] L. Sun, N. Tran, F. Tang, H. App, P. Hirth, G. McMahon, C. Tang, *J. Med. Chem.* **1998**, *41*, 2588; A. Andreani, S. Burnelli, M. Granaiola, A. Leoni, A. Locatelli, R. Morigi, M. Rambaldi, L. Varoli, M. W. Kunkel, *J. Med. Chem.* **2006**, *49*, 6922.

[14] K. Amornraksa, R. Grigg, H. Q. N. Gunaratna, J. Kemp, V. Sridharan, *J. Chem. Soc., Perkin Trans. 1* **1987**, 2285.

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