JOC Article

High Nucleophilicity of Cyclic Amidocarbene toward Aryl Isocyanates, New Approach to Spiro[azetidinone-4,3′-indolinone] Derivatives

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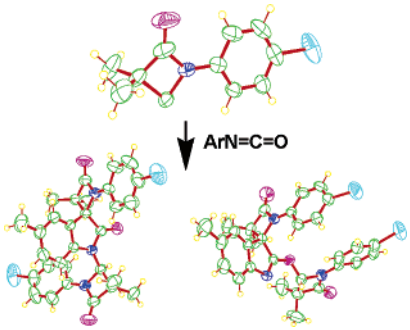

The nucleophilic addition of β-lactam-4-ylidenes **2**, a type of ambiphilic cyclic amidocarbene, to aryl isocyanates has been studied and their application in organic synthesis has been demonstrated. Thermolysis of spiro[β-lactam-4,2′-oxadiazolines] **1** in the presence of aryl isocyanates afforded both *N*-lactam and *O*-lactam substituted spiro[azetidine-2-one-4,3′-indole-2′-one] derivatives **5** and **6** in the total yield of 65−86%. Upon hydrolysis, products **5** and **6** were converted into spiro[azetidine-2-one-4,3′-indole-2′-one] **9** that was analogous to known biologically active compounds.

Introduction

Carbene is the sole two-valence intermediate in the carbon-central active intermediates. The completion of the octet of electrons at the carbene center is, in general, the driving force behind the carbene reactions. Therefore, these active species normally behave as electrophiles, and react more effectively with electron-rich substrates than with electron-deficient ones.[1] If one or two heteroatoms (e.g., O, N, or S) are directly bound to the carbene center, the electronic delocalization of the lone pair electrons on heteroatoms can compensate for the electronic deficiency at the carbene and could cause the nature of the carbene to change from electrophilic to nucleophilic.[2] Carbenes can thus be divided into electrophilic, ambiphilic, and nucleophilic carbenes. The nucleophilic carbenes, especially the *N*-heterocyclic carbenes,[3] have attracted considerable attention in recent years because many *N*-heterocyclic carbenes are stable and can be used as organocatalysts[4] and as ligands in transition metal complexes.[3,5] In addition, the nucleophilic carbenes are also versatile intermediates in the construction of novel multifunctional heterocycles.[6] For example, diamino-, aminooxy-, aminothio-, dioxy-, dithio-, and oxythiocarbenes have been reported to react with aryl or vinyl isocyanates to form *N*,*N*-diarylimidazoline-2,4-dione (hydantoin)[7] or pyrroline-2-one derivatives,[8] respectively. In comparison to nucleophilic carbenes, the chemistry of ambiphilic carbenes has scarcely been explored. Except for cyclopropanation of alkenes and alkynes, ambiphilic carbenes seem not to have found much usage in organic synthesis. In 1990s, Warkentin and co-workers inves-

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tigated the reactions of β-lactam-4-ylidenes, a type of cyclic amidocarbenes derived from thermolysis of spiro[β-lactam-4,2′-oxadiazolines],[9] with both electron-rich and electron-deficient alkenes or alkynes, and they demonstrated the β-lactam-4-ylidenes to be ambiphilic carbenes but with pronounced electrophilic activity.[10] We have been interested in the chemistry of nucleophilic and ambiphilic carbenes for some time,[11] and our previous studies have shown the reaction outcomes being strongly influenced by the structures of both carbenes and their reaction partners. Although the β-lactam-4-ylidenes are formally *N*-heterocyclic carbenes, the amido nitrogen is relatively a weak electronic donor and therefore they might not be distinctly nucleophilic. With the purpose of investigating the nucleophilic behavior of cyclic amidocarbenes, and of preparing multifunctional β-lactam derivatives of pharmaceutical importance, we undertook the current study of the reaction between β-lactam-4-ylidenes and aryl isocyanates. Herein, we report our findings that β-lactam-4-ylidenes, the typical ambiphilic carbenes, actually behave as good nucleophiles as bisheteroatom-substituted carbenes in the addition reaction to isocyanates to form both *N*- and *O*-lactam substituted spiro[azetidine-2-one-4,3′-indole-2′-one] derivatives in the total yield of 65−86%.

Results and Discussion

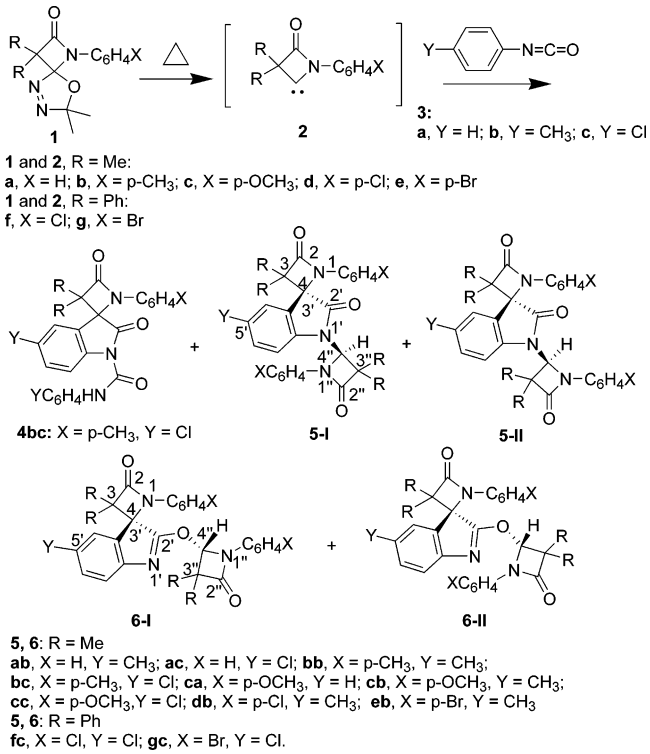
All cyclic amidocarbenes, β-lactam-4-ylidenes **2**, were generated in situ by thermolysis of spiro[β-lactam-4,2′-oxadiazolines] **1** according to the Warkentin's method.[9] Initially, we examined the reactivity of **2b** (R = Me, X = Me) by refluxing oxadiazoline **1b** with 1 equiv of aryl isocyanate **3c** (Y = Cl) in toluene. The reaction proceeded smoothly to give three types of products **4bc**, **5bc**, and **6bc** in the total yield of 73% (entry 1 in Table 1, Scheme 1). To increase the chemical yields and to improve the selectivity, the reaction was optimized by varying the ratio of starting materials, reaction media, and temperature. It was found that, in the presence of an excess amount of isocyanate **3c**, the yield of **4bc** slightly increased. When the carbene source **1b** was doubled, **5bc** and **6bc** were isolated in a combined yield of 82% and the formation of **4bc** was eliminated (entry 5, Table 1). Toluene, 1,4-dioxane, and benzene were observed to favor the formation of *O*-substituted product **6bc**, whereas propionitrile and trichloroethane were beneficial to *N*-substituted product **5bc**. Since products **5** and **6** were derived respectively from two different intermediates, indoline-2-one **9** and 2-hydroxyl indolenine **10** (vide infra), the solvent effect on the outcomes of reaction was most probably due to the stabilization of the intermediates **9** and **10** by a polar and nonpolar solvent, respectively.

TABLE 1. The Optimization of Thermolysis of Spiro[β-lactam-4,2′-oxadiazoline] 1b and Reaction between Carbene 2b and *p*-Chlorophenyl Isocyanate 3c

entry	starting materials		1:3	solvent	temp (°C)	time (h)	yield (%)[a] 4[b]	5[b]	6[b]	total
1	**1b**	**3c**	1:1	toluene	110	12	4	28	41	73
2	**1b**	**3c**	1:2	toluene	110	12	7	23	39	69
3	**1b**	**3c**	1:3	toluene	110	12	18	20	28	66
4	**1b**	**3c**	1:4	toluene	110	12	18	25	29	72
5	**1b**	**3c**	2:1	toluene	110	12		33	49	82
6	**1b**	**3c**	2:1	benzene	80	12		18	43	61
7	**1b**	**3c**	2:1	1,4-dioxane	100	12		32	42	74
8	**1b**	**3c**	2:1	CH_3CH_2CN	100	12		46	31	77
9	**1b**	**3c**	2:1	$ClCH_2CH_2Cl$	80	12		28	29	57
10	**1b**	**3c**	2:1	Cl_2CHCH_2Cl	110	12		40	18	58

[a] Isolated yield [b] Calculation of the yield of **4** was based on the reaction of one carbene with two isocyanate molecules, while the yields of **5** and **6** were based on the reaction of two carbenes with one isocyanate molecule.

SCHEME 1. The Reactions between β-Lactam-4-ylidenes 2 and Aryl Isocyanates 3



To examine the scope of the reaction and to have a good understanding of the mechanism, the reactions between carbenes **2** and aryl isocyanates **3** both bearing different substituents were investigated by refluxing the mixture of carbene sources **1** and aryl isocyanates **3** (**1**:**3** 2:1) in toluene for 12 h. As illustrated in Table 2, the substituents on phenyl groups in both carbene and isocyanate have a negligible effect on the outcomes of the reaction. In all cases, the reaction afforded moderate to good overall yields of **5** and **6** with the ratio of **6** over **5** ranging from 1:1 to 2:1 (Scheme 1 and Table 2).

Both products **5** and **6** exist as a pair of diastereoisomers. Most of the diastereoisomers were separated successfully by

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TABLE 2. The Reactions of Carbenes 2 with Aryl Isocyanates 3 under Optimal Conditions

entry	starting materials	products	yield (%)[a] 5-I	5-II	5	6-I	6-II	6	6:5[b]	total
1	**1a**, **3b**	**5ab**, **6ab**	12	11	23			47	2:1	70
2	**1a**, **3c**	**5ac**, **6ac**	13	13	26			45	1.7:1	71
3	**1b**, **3b**	**5bb**, **6bb**	14	12	26	26	16	42	1.6:1	68
4	**1b**, **3c**	**5bc**, **6bc**	16	17	33			49	1.5:1	82
5	**1c**, **3a**	**5ca**, **6ca**	11	12	23	26	16	42	1.8:1	65
6	**1c**, **3b**	**5cb**, **6cb**	18	14	32	19	15	34	1:1	66
7	**1c**, **3c**	**5cc**, **6cc**	15	18	33	19	14	33	1:1	66
8	**1d**, **3b**	**5db**, **6db**	10	19	29	31	26	57	2:1	86
9	**1e**, **3b**	**5eb**, **6eb**	12	15	27	26	19	45	1.7:1	72
10	**1f**, **3c**	**5fc**, **6fc**	19	14	33	31	16	47	1.4:1	80
11	**1g**, **3c**	**5gc**, **6gc**	17	16	33	29	14	43	1.3:1	76

[a] Isolated yield. [b] The ratio of product **6** over **5** was determined by isolated yields.

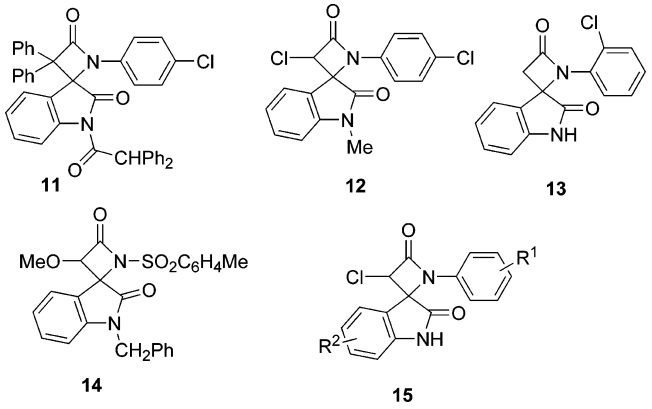

FIGURE 1. Biologically active spiro[azetidine-4,3′-indoline]-2,2′-diones.

column chromatography on silica gel. The pairs of two diastereoisomers of **6ab**, **6ac**, and **6bc** could not be separated by column chromatography; fortunately, pure **6ab-I**, **6ac-I**, or **6bc-I** was precipitated respectively from their solution in a mixture of petroleum ether and ethyl acetate. The structures of all products were elucidated on the basis of spectroscopic data, microanalysis, and X-ray diffraction studies. Thus, mass data and elemental analyses indicated the product **4** derived from one carbene and two isocyanate molecules (1+2 adduct) but **5** and **6** were the adducts of two carbene moieties to one isocyanate (2+1 adduct), respectively. However, spectroscopic data did not allow full verification of the structure. X-ray diffraction studies then unambiguously confirmed that the compound **5db-I** (R = Me, X = Cl, Y = CH_3) was (*R*,*R*)- or (*S*,*S*)-1′-(azetidine-2″-one)spiro[azetidine-4,3′-indoline]-2,2′-dione, while **6eb-I** (R = Me, X = Br, Y = CH_3) and **6gc-I** (R = Ph, X = Br, Y = Cl) were (*R*,*R*)- or (*S*,*S*)-2′-(azetidine-2″-one)oxyspiro[azetidine-4,3′-indolenine]-2-ones (see the Supporting Information, Figure 1). No X-ray quality single crystals were obtained for compound **4bc**. The observations of a N−H moiety in both the infrared and ^{1}H NMR spectrum and of a similar resonance pattern of the aromatic protons as that of **5bc** in the ^{1}H NMR spectrum suggested the 1′-(*p*-chlorophenyl)-carbamoyl spiro[azetidine-4,3′-indoline]-2,2′-dione structure for **4bc**. The proposed structure of **4bc** was also supported by the chemical hydrolyses of **4bc**, **5bc**, and **6bc** that all gave the same spiro[azetidine-4,3′-indoline]-2,2′-dione product **9bc**. The structure assignment of **4bc** was further evidenced by preparation of **4bc** from the reaction of **9bc** with isocyanate **3c** (vide infra).

It is worth noting that products **5** and **6** show distinctly different ^{13}C NMR and ^{1}H NMR spectra and they can be used in turn as diagnostics to differentiate **5** and **6**. For example, *N*,*N*-substituted carbon (4″-C) of **5-I** and **5-II** resonated in a range of 68−70 ppm in ^{13}C NMR, while *N*,*O*-substituted carbon of **6-I** and **6-II** appeared at a lower field around 85−90 ppm, respectively. In the products derived from dimethyl substituted *β*-lactam-4-ylidenes **2a**−**2e**, the 4″-H signal of **5-I** was a sharp singlet at about 6.0 ppm, whereas in **5-II** the 4″-H appeared as a broad peak at 6.2 ppm. In addition to these NMR features of 4″-C and 4″-H between configurational isomers or diastereomers, the 3″-Me groups in two diastereomers of **6** can also be easily distinguished. In the case of isomers **6-I**, all of the methyl proton signals were observed at the normal region (1.2−1.6 ppm); however, the proton of one of the four methyl groups in isomer **6-II** resonated at very high field (0.7−0.8 ppm). (See the Supporting Information, table and NMR spectra.) This shielding effect can be explained from the ChemBats3D optimized structure of **6-II**, which showed one methyl group was exactly located in the shielding region of the *N*-substituted phenyl ring. (See the Supporting Information, Figure 2.)

The formation of indolone derivatives **4**, **5**, and **6** can be best explained by nucleophilic addition of *β*-lactam carbenes to isocyanates. The resulting zwitterion **7** undergoes intramolecular electrophilic substitution reaction to give intermediate **9**. The interaction of **9** with another isocyanate molecule affords product **4**, while insertion of carbene to the N−H or O−H bond of the tautomers **9** or **10** accounts for the formation of **5** and **6**, respectively (Scheme 2). The pathway from **9** to **4** was confirmed by reaction of **9bc** with isocyanate **3c** under the identical conditions of the reaction between **1b** and **3c** that afforded **4bc** as the sole product in 73% yield. The direct formation of **5bc** (27%) and **6bc** (15%) from thermolysis of carbene source **1b** in the presence of **9bc** gave evidence for the mechanism depicted in Scheme 2. It should be noted that *β*-lactam amidocarbene **2** exhibits different reactivity from typical nucleophilic carbenes when treated with aryl isocyanates. For example, most reactions between diheteroatom-substituted carbenes and aryl isocyanates have been reported to afford *N*,*N*-diarylimidazoline-2,4-dione (hydantoin) derivatives as sole products,[7] while only *N*,*N*- or *N*,*S*-substituted carbenes were mentioned to form indolone products in very low yields (13−39%).[8a,12] On the contrary, however, only indolone derivatives were obtained in our cases. Furthermore, the *O*-carbene substituted indole-2-ones, the major products in our reactions, have never before been reported in the reactions between typical nucleophilic carbenes and aryl or vinyl isocyanates. The different outcomes most probably originated from the different stability and therefore the reactivity of the dipolar intermediates. The dipolar intermediates derived from the addition of typical nucleophilic carbenes to isocyanates are stabilized by two heteroatoms. Therefore the carbon cation of this dipole is not reactive to aromatic electrophilic substitution, and the nitrogen anion, on the other hand, readily attacks the second molecule of aryl isocyanate to form *N*,*N*-diarylimidazoline-2,4-dione (hydantoin). In contrast, the less stable dipolar intermediate **7** derived from amidocarbenes prefers to undergo intramolecular cyclization to form an indolone intermediate **9**, which provides products **4**, **5**, and **6** via different pathways.

It has been reported that many spiro[azetidinone-4,3′-indolones] derivatives are biologically active. For example, com-

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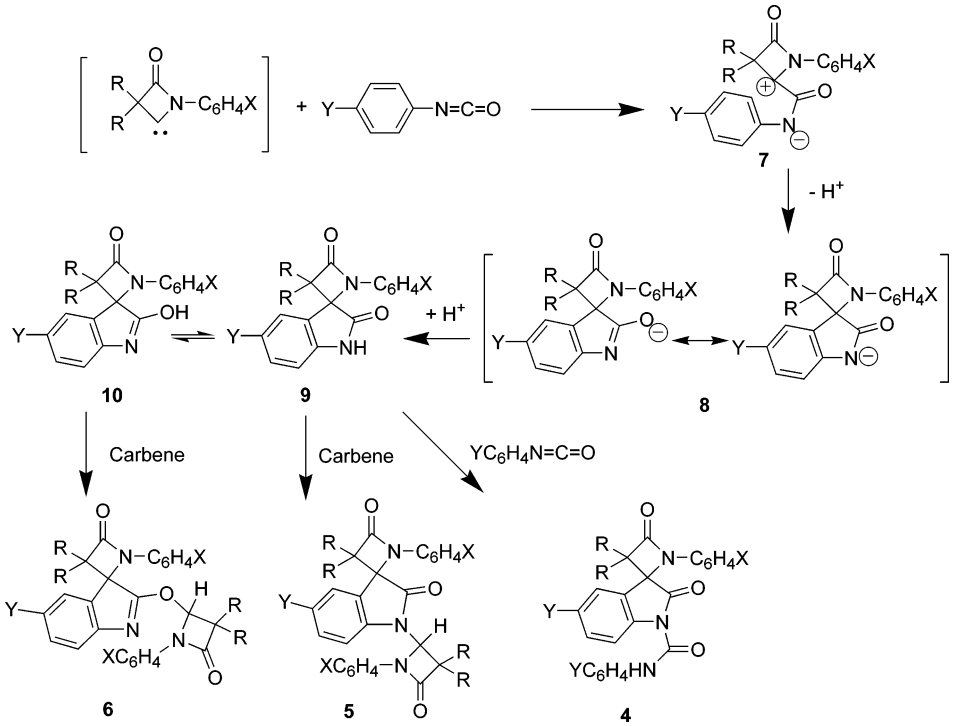
SCHEME 2. The Proposed Mechanism for the Formation of Products 4, 5, and 6



pounds **11**[13a] and **12**[13b] have been found to exhibit significant activity against maximal electroshock seizure, while compounds **13**[13c] show fungicidal activity against *Aspergillus flavus, A. niger*, and *Helminthosporium oryzae*. Compound **14**[13d] has been reported to inhibit picornaviruses and human leukocyte elastase, and compounds **15**[13e] are active against gram-positive bacteria (Figure 1). With products **4**−**6** in hand, we envisioned that the removal of the *N*-substituent or *O*-substituent would easily give analogues of those bioactive compounds. Treated with *p*-toluenesulfonic acid in a hot chlorinated alkane solution, both the *N*-substituted indolones **5bc** and its *O*-substituted isomers **6bc** were converted into spiro[azetidine-4,3′-indoline]-2,2′-dione **9bc** in excellent yield, albeit the hydrolysis of the former needs a higher temperature or longer reaction time than that of the later. Hydrolysis of **4bc** with sodium hydroxide in ethanol also afforded **9bc**. Finally, compound **9bc** was prepared very conveniently in good yield by refluxing **1b** with **3c** in toluene followed by heating the resulting mixture with *p*-toluenesulfonic acid in trichloroethane (Scheme 3 and Table 3). With our method, **9fa**[14] readily prepared from **1f** and **3a** was converted into bioactive compound **11**[13a] in 60% yield with treatment of diphenylacetyl chloride at room temperature (Scheme 4). Almost all of the known methods for preparation of spiro[azetidinone-4,3′-indolones] were based on the [2+2] cycloaddition of a ketene with an imine that was yielded from the reaction of an amine with an isatin.[13,15] Since most of the substituted isatins are not commercially available, the application of a known literature method has been limited. The easy availability of isocyanates under our cyclic carbene approach gives more advantages in the synthesis of spiro[azetidinone-4,3′-indolones].

In summary, we have studied the nucleophilicity of cyclic amidocarbenes, *β*-lactam-4-ylidenes. They are highly reactive toward aryl isocyanates to form both *N*- and *O*-*β*-lactam substituted spiro[azetidinone-4,3′-indolinone] derivatives. The products have been further converted in good to excellent yields into spiro[azetidinone-4,3′-indolinone], which was analogous to the known biologically active compounds. We have revealed the nucleophilic feature of cyclic amidocarbenes and demonstrated their application in organic synthesis.

Experimental Section

General Procedure for Thermolysis of Spiro[*β*-lactam-4,3′-oxadiazolines] 1 in the Presence of Aryl Isocyanates. Under nitrogen atmosphere, a mixture of oxadiazolines **1** (6 mmol) and aryl isocyanate **3** (3 mmol) was refluxed in toluene (80 mL) for 12 h. After removal of the solvent, the residue was subjected to chromatography on a silica gel column eluting with a mixture of petroleum ether (60−90 °C) and ethyl acetate (petroleum ether: ethyl acetate from 10:1 to 3:1) to afford products **5** and **6**. (Note: **4bc** was also isolated from the reaction of 3 mmol of oxadiazoline **1b** with 3 mmol of isocyanate **3c**.)

(*R*,*R*)- and (*S*,*S*)-1′-(3″,3″-dimethyl-1″-phenylazetidin-2″-one)-3,3,5′-trimethyl-1-phenylspiro[azetidine-4,3′-indoline]-2,2′-dione 5ab-I: mp 214−215 °C; IR υ (cm^{-1}) 1770, 1735, 1503; ^{1}H NMR (500 MHz) δ (ppm) 7.34−7.38 (m, 4H), 7.22 (t, J = 7.6 Hz, 2H), 7.14 (t, J = 7.0 Hz, 1H), 7.11 (s, 1H), 7.09 (d, J = 8.4 Hz, 2H), 7.05 (d, J = 10.8 Hz, 2H), 7.01 (d, J = 8.2 Hz, 1H), 6.06 (s, 1H), 2.26 (s, 3H), 1.59 (s, 6H), 1.31 (s, 3H), 1.30 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 174.6, 170.1, 169.9, 138.8, 137.3, 136.8, 132.8, 131.3, 129.6, 129.2, 126.8, 124.9, 124.4, 122.8, 117.0, 116.1, 112.2, 69.8, 69.1, 60.2, 57.3, 22.5, 21.1, 21.0, 18.5, 16.3; MS (EI) 70 (100), 119 (90), 146 (50), 479 (M$^+$, 60%). Anal. Calcd for $C_{30}H_{29}N_3O_3$: C 75.13, H 6.09, N 8.76. Found: C 75.11, H 6.35, N 8.67.

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SCHEME 3. Preparation of Spiro[azetidine-4,3′-indoline]-2,2′-dione 9bc

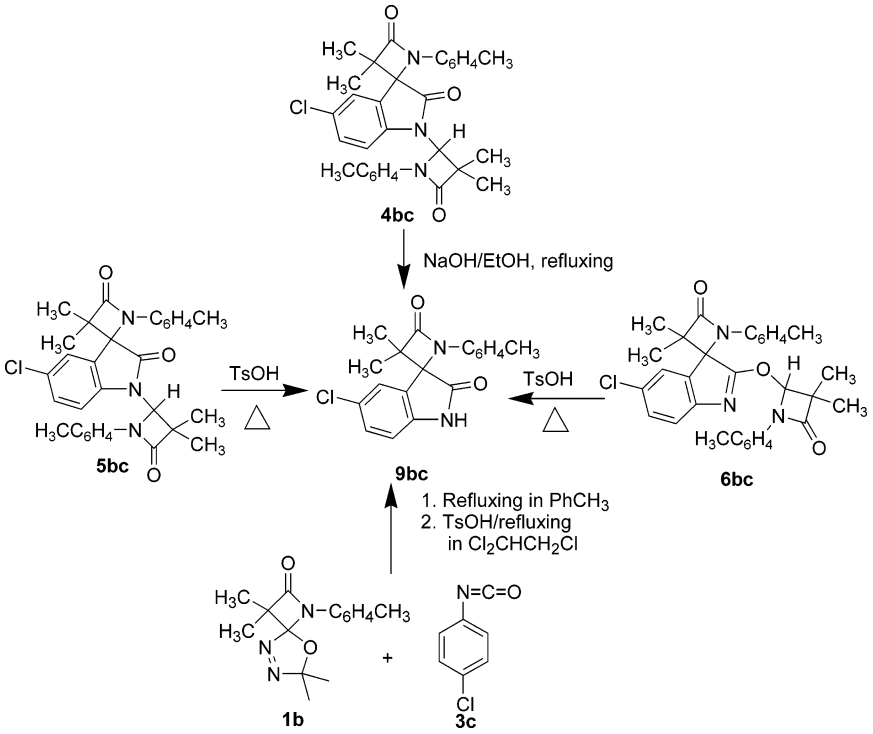


TABLE 3. Preparation of Spiro[azetidine-4,3′-indoline]-2,2′-dione 9bc and 9fa

starting material	reagent/ solvent	temp (°C)	time (h)	product	yield (%)[a]
6bc	TsOH/$CHCl_3$	60−70	0.5	**9bc**	93
5bc-I	TsOH/$CHCl_3$	60−70	17	**9bc**	95
5bc-I	TsOH/TCE[b]	110−120	3	**9bc**	95
5bc-II	TsOH/$CHCl_3$	60−70	41	**9bc**	95
5bc-II	TsOH/TCE	110−120	6	**9bc**	98
5bc-II	TsOH/Toluene	110−120	17	**9bc**	36
4bc	NaOH/EtOH	70−80	3	**9bc**	46
1b, **3c**	toluene	110−120	12	**9bc**	75[c]
	TsOH/TCE	110−120	8		
1f, **3a**	toluene	110−120	12	**9fa**	68
	TsOH/TCE	110−120	8		

[a] Isolated yield. [b] TCE = trichloroethane. [c] A mixture of **1b** and **3c** was refluxed in toluene for 12 h, and after removal of toluene then another 8 h with TsOH in trichloroethane.

(*R*,*S*)- and (*S*,*R*)-1′-(3″,3″-dimethyl-1″-phenylazetidin-2″-one)-3,3,5′-trimethyl-1-phenylspiro[azetidine-4,3′-indoline]-2,2′-dione 5ab-II: mp 249−250 °C; IR υ (cm^{-1}) 1761, 1727, 1503; ^{1}H NMR (500 MHz) δ (ppm) 7.31−7.36 (m, 4H), 7.19 (t, J = 7.7 Hz, 2H), 7.15 (t, J = 7.0 Hz, 1H), 7.08 (d, J = 7.2 Hz, 2H), 7.05 (s, 2H), 7.01 (d, J = 7.7 Hz, 2H), 6.28 (brs, 1H), 2.24 (s, 3H), 1.61 (s, 3H), 1.52 (br s, 3H), 1.35 (s, 6H); ^{13}C NMR (125 MHz) δ (ppm) 175.3, 169.9 (2 CO), 138.7, 137.0, 136.8, 132.8, 131.2, 129.6, 129.2, 126.6, 124.8, 124.3, 122.7, 117.0, 116.3, 116.1, 112.5, 69.0, 68.6, 60.0, 56.6, 23.0, 21.02, 20.95, 18.5, 16.6; MS (EI) 146 (90), 479 (M^+, 100%). Anal. Calcd for $C_{30}H_{29}N_3O_3$: C 75.13, H 6.09, N 8.76. Found: C 74.92, H 6.39, N 8.67.

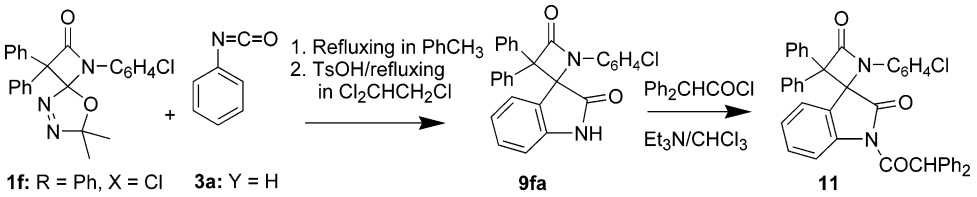
(*R*,*R*)- and (*S*,*S*)-2′-(3″,3″-dimethyl-1″-phenylazetidin-2″-one)-oxy-3,3,5′-trimethyl-1-phenylspiro[azetidine-4,3′-indolenine]-2-one 6ab-I: mp 219−220 °C; IR υ (cm^{-1}) 1769, 1762, 1587, 1504; ^{1}H NMR (300 MHz) δ (ppm) 7.36 (d, J = 7.8 Hz, 1H), 7.24−7.32 (m, 3H), 7.19 (s, 1H), 7.15 (t, J = 7.4 Hz, 1H), 7.08 (d, J = 7.7 Hz, 2H), 6.83−6.98 (m, 5H), 5.99 (s, 1H), 2.38 (s, 3H), 1.59 (s, 3H), 1.59 (s, 3H), 1.44 (s, 3H), 1.40 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 175.3, 170.1, 169.7, 149.2, 137.7, 136.7, 134.4, 131.1, 130.6, 129.6, 129.1, 126.0, 124.4, 124.3, 120.1, 116.5, 116.2, 89.9, 73.3, 58.7, 56.8, 21.7, 21.5, 19.8, 19.7, 17.2; MS (EI) 146 (100), 235 (25), 306 (10), 479 (M^+, 10%). Anal. Calcd for $C_{30}H_{29}N_3O_3$: C 75.13, H 6.09, N 8.76. Found: C 75.10, H 6.27, N 8.74.

The mixture of two diastereoisomers 6ab-II and 6ab-I (6ab-II:6ab-I 2:1): IR υ (cm^{-1}) 1769, 1588, 1503; ^{1}H NMR δ (500 MHz) (ppm) 7.49 (d, J = 8.0 Hz, 2H, II), 7.38 (d, J = 7.9 Hz, 1H + 0.5H, II + I), 7.35 (t, J = 7.4 Hz, 2H, II), 7.25−7.32 (m, 2.5H, II + I), 7.13−7.22 (m, 5H, II + I), 7.08 (d, J = 7.7 Hz, 1H, I), 7.05 (t, J = 7.5 Hz, 1H, II), 7.02 (d, J = 7.8 Hz, 2H, II), 6.94 (d, J = 7.8 Hz, 0.5H, I), 6.90 (t, J = 7.1 Hz, 1H, I), 6.86 (d, J = 7.4 Hz, 1H, I), 6.66 (s, 1H, II), 6.00 (s, 0.5H, I), 2.38 (s, 3H + 1.5H, II + I), 1.60 (s, 1.5H, I), 1.59 (s, 1.5H, I), 1.45 (s, 1.5H, I), 1.41 (s, 3H + 1.5H, II + I), 1.37 (s, 3H, II), 1.33 (s, 3H, II), 0.72 (s, 3H, II); ^{13}C NMR (125 MHz) δ (ppm) 176.0, 175.3, 170.2, 169.8, 149.1, 137.3, 136.7, 136.1, 134.3, 131.0, 130.6, 130.3, 129.6, 129.2, 129.1, 125.9, 124.8, 124.4, 124.3, 120.7, 119.9, 117.4, 116.7, 116.5, 116.2, 89.9, 86.8, 59.1, 58.7, 56.8, 56.4, 22.1, 21.7, 21.5, 20.2, 19.8, 19.7, 19.0, 17.2, 15.9; MS (EI) 146 (100), 187 (15), 235 (15), 479 (M^+, 15%). Anal. Calcd for $C_{30}H_{29}N_3O_3$: C 75.13, H 6.09, N 8.76. Found: C 74.92, H 6.10, N 8.65.

(*R*,*R*)- and (*S*,*S*)-1′-(3″,3″-dimethyl-1″-phenylazetidin-2″-one)-5′-chloro-3,3,-dimethyl-1-phenylspiro[azetidine-4,3′-indoline]-2,2′-dione 5ac-I: mp 253−255 °C; IR υ (cm^{-1}) 1772, 1736, 1600; ^{1}H NMR (500 MHz) δ (ppm) 7.35−7.36 (d, J = 3.6 Hz, 4H), 7.29 (s, 1H), 7.23−7.26 (m, 3H), 7.16−7.18 (m, 1H), 7.11 (d, J = 7.4 Hz, 1H), 7.05−7.08 (m, 3H), 6.05 (s, 1H), 1.60 (s, 3H), 1.59 (s, 3H), 1.33 (s, 3H), 1.30 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 174.1, 169.8, 169.3, 139.7, 137.0, 136.5, 130.9, 129.7, 129.3, 129.0, 126.3, 125.2, 124.9, 124.7, 116.9, 116.0, 113.3, 69.9, 68.8, 60.8, 57.4, 22.5, 21.2, 18.5, 16.3; MS (EI) 119 (72), 146 (100), 499 (M^+, 6%)/501 (2). Anal. Calcd for $C_{29}H_{26}ClN_3O_3$: C 69.66, H 5.24, N 8.40. Found: C 69.49, H 5.34, N 8.35.

(*R*,*S*)- and (*S*,*R*)-1′-(3″,3″-dimethyl-1″-phenylazetidin-2″-one)-5′-chloro-3,3,-dimethyl-1-phenylspiro[azetidine-4,3′-indoline]-2,2′-dione 5ac-II: mp 284−285 °C; IR υ (cm^{-1}) 1762, 1731, 1603; ^{1}H NMR (500 MHz) δ (ppm) 7.34 (s, 4H), 7.27 (s, 1H), 7.22 (t, J = 7.8 Hz, 3H), 7.19 (br s, 1H), 7.10 (t, J = 8.5 Hz, 2H), 6.99 (d,

SCHEME 4. Preparation of 1′-Diphenylacetyl-3,3,-diphenylspiro[azetidine-4,3′-indoline]-2,2′-dione 11



J = 8.3 Hz, 2H), 6.27 (br s, 1H), 1.62 (s, 3H), 1.52 (s, 3H), 1.37 (s, 3H), 1.34 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 174.8, 169.7, 169.4, 139.6, 136.7, 136.5, 130.8, 129.7, 129.3, 129.0, 126.1, 125.1, 124.8, 124.6, 116.9, 116.1, 113.6, 68.6 (4-C and 4″-C), 60.6, 56.7, 22.9, 21.1, 18.4, 16.6; MS (EI) 119 (60), 146 (100), 499 (M^+, 4%). Anal. Calcd for $C_{29}H_{26}ClN_3O_3$: C 69.66, H 5.24, N 8.40. Found: C 69.32, H 5.48, N 8.29.

(*R*,*R*)- and (*S*,*S*)-2′-(3″,3″-dimethyl-1″-phenylazetidin-2″-one)-oxy-5′-chloro-3,3,-dimethyl-1-phenylspiro[azetidine-4,3′-indolenine]-2-one 6ac-I: mp 217−218 °C; IR υ (cm^{-1}) 1763, 1601, 1583; ^{1}H NMR (500 MHz) δ (ppm) 7.42−7.46 (m, 2H), 7.37 (s, 1H), 7.32 (t, J = 7.9 Hz, 2H), 7.19 (t, J = 7.4 Hz, 1H), 7.06 (d, J = 8.0 Hz, 2H), 6.97 (t, J = 7.2 Hz, 1H), 6.91 (t, J = 7.9 Hz, 2H), 6.85 (d, J = 7.8 Hz, 2H), 6.00 (s, 1H), 1.61 (s, 3H), 1.59 (s, 3H), 1.46 (s, 3H), 1.41 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 176.3, 169.5, 169.4, 150.1, 137.4, 136.6, 132.4, 130.7, 130.4, 129.8, 129.2, 125.4, 124.7, 124.4, 121.4, 116.5, 116.1, 90.3, 73.3, 59.4, 56.9, 21.9, 19.7 (2 CH_3), 17.2; MS (EI) 129 (82), 144 (100), 174 (75), 254 (40), 499 (M^+, 25)/501 (7). Anal. Calcd for $C_{29}H_{26}ClN_3O_3$: C 69.66, H 5.24, N 8.40. Found: C 69.45, H 5.40, N 8.39.

The mixture of two diastereoisomers 6ac-II and 6ac-I (6ac-II:6ac-I 1:0.9): IR υ (cm^{-1}) 1778, 1766, 1600, 1581; ^{1}H NMR (500 MHz) δ (ppm) 7.42−7.47 (m, 4H + 1.8H, II + I), 7.34−3.77 (m, 3H + 0.9H, II + I), 7.32 (t, J = 7.9 Hz, 1.8H, I), 7.23 (d, J = 8.0 Hz, 1H, II), 7.19 (d, J = 8.8 Hz, 1H, II), 7.16 (t, J = 7.1 Hz, 0.9H, I), 7.05−7.09 (m, 2H + 0.9H, II + I), 7.00 (d, J = 7.8 Hz, 2H, II), 6.96 (t, J = 7.2 Hz, 0.9H, I), 6.91 (t, J = 7.9 Hz, 1.8H, I), 6.86 (d, J = 7.8 Hz, 1.8H, I), 6.66 (s, 1H, II), 6.00 (s, 0.9H, I), 1.61 (s, 2.7H, I), 1.59 (s, 2.7H, I), 1.46 (s, 2.7H, I), 1.42 (s, 3H, II), 1.41 (s, 2.7H, I), 1.38 (s, 3H, II), 1.34 (s, 3H, II), 0.73 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 176.9, 176.3, 169.6, 169.4, 150.0, 137.4, 137.1, 136.6, 136.0, 132.4, 132.1, 130.74, 130.69, 130.4, 130.3, 129.8, 129.3, 129.22, 129.16, 125.44, 125.39, 125.0, 124.7, 124.6, 124.4, 121.4, 121.2, 117.3, 116.6, 116.5, 116.1, 90.3, 87.2, 59.7, 59.4, 56.9, 56.5, 22.2, 21.9, 20.2, 19.7, 18.9, 17.2, 15.9; MS (EI) 119 (60), 146 (100), 192 (53), 499 (15)/501 (5). Anal. Calcd for $C_{29}H_{26}ClN_3O_3$: C 69.66, H 5.24, N 8.40. Found: C 69.50, H 5.38, N 8.37.

(*R*,*R*)- and (*S*,*S*)-1′-(3″,3″-dimethyl-1″-methylphenylazetidin-2″-one)-3,3,5′-trimethyl-1-methylphenylspiro[azetidine-4,3′-indoline]-2,2′-dione 5bb-I: mp 179−181°C; IR υ (cm^{-1}) 1758, 1725, 1519, 1488; ^{1}H NMR (300 MHz) δ (ppm) 7.25 (d, J = 8.3 Hz, 2H), 7.13 (d, J = 8.3 Hz, 2H), 7.09 (s, 1H), 6.96−7.05 (m, 6H), 6.02 (s, 1H), 2.30 (s, 3H), 2.26 (s, 3H), 2.25 (s, 3H), 1.57 (s, 6H), 1.29 (s, 6H); ^{13}C NMR (125 MHz) δ (ppm) 174.7, 169.9, 169.7, 138.9, 134.8, 134.6, 134.3, 134.1, 132.6, 131.3, 130.1, 129.7, 126.7, 122.9, 116.9, 116.0, 112.2, 69.8, 69.1, 60.1, 57.2, 22.5, 21.1, 21.0, 20.92, 20.85, 18.5, 16.3; MS (EI) 70 (100), 91 (88), 132 (62), 133 (100), 160 (65), 507 (40)/508 (15, M + 1). Anal. Calcd for $C_{32}H_{33}N_3O_3$: C 75.71, H 6.55, N 8.28. Found: C 75.73, H 6.75, N 8.29.

(*R*,*S*)- and (*S*,*R*)-1′-(3″,3″-dimethyl-1″-methylphenylazetidin-2″-one)-3,3,5′-trimethyl-1-methylphenylspiro[azetidine-4,3′-indoline]-2,2′-dione 5bb-II: mp 238−239 °C; IR υ (cm^{-1}) 1762, 1730, 1518, 1488; ^{1}H NMR (500 MHz) δ (ppm) 7.24 (d, J = 8.1 Hz, 2H), 7.11 (d, J = 8.2 Hz, 2H), 7.07 (s, 1H), 7.03 (s, 2H), 6.98 (d, J = 8.2 Hz, 2H), 6.91 (d, J = 8.3 Hz, 2H), 6.23 (br s, 1H), 2.34 (s, 3H), 2.30 (s, 3H), 2.24 (s, 3H), 1.59 (s, 3H), 1.50 (br s, 3H), 1.33 (s, 6H); ^{13}C NMR (125 MHz) δ (ppm) 175.3, 169.7 (2 CO), 138.8, 134.6, 134.45, 134.38, 133.9, 132.6, 131.2, 130.0, 129.7, 126.6, 122.8, 117.0, 116.2, 112.4, 69.0, 68.6, 59.9, 56.6, 22.9, 21.01, 20.95, 18.5, 16.6; MS (EI) 91 (55), 133 (35), 160 (60), 507 (M^+, 100)/508 (35, M + 1). Anal. Calcd for $C_{32}H_{33}N_3O_3$: C 75.71, H 6.55, N 8.28. Found: C 75.75, H 7.01, N 8.42.

(*R*,*R*)- and (*S*,*S*)-2′-(3″,3″-dimethyl-1″-methylphenylazetidin-2″-one)oxy-3,3,5′-trimethyl-1-methylphenylspiro[azetidine-4,3′-indolenine]-2-one 6bb-I: mp 241−243 °C; IR υ (cm^{-1}) 1765, 1581, 1518; ^{1}H NMR (300 MHz) δ (ppm) 7.36 (d, J = 7.8 Hz, 1H), 7.24 (d, J = 7.8 Hz, 1H), 7.17 (s, 1H), 7.08 (d, J = 8.3 Hz, 2H), 6.97 (d, J = 8.5 Hz, 2H), 6.76 (d, J = 8.5 Hz, 2H), 6.69 (d, J = 8.3 Hz, 2H), 5.96 (s, 1H), 2.37 (s, 3H), 2.34 (s, 3H), 2.23 (s, 3H), 1.58 (s, 3H), 1.57 (s, 3H), 1.43 (s. 3H), 1.39 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 175.5, 169.9, 169.5, 149.2, 135.2, 134.32, 134.26, 133.9, 131.0, 130.7, 130.0, 129.5, 125.9, 120.0, 117.1, 116.6, 116.2, 90.0, 73.3, 58.7, 56.7, 21.7, 21.5, 20.9, 19.80, 19.77, 17.2; MS (EI) 91 (78), 144 (45), 160 (100), 507 (M^+, 17)/508 (7). Anal. Calcd for $C_{32}H_{33}N_3O_3$: C 75.71, H 6.55, N 8.28. Found: C 75.73, H 6.85, N 8.03.

(*R*,*S*)- and (*S*,*R*)-2′-(3″,3″-dimethyl-1″-methylphenylazetidin-2″-one)oxy-3,3,5′-trimethyl-1-methylphenylspiro[azetidine-4,3′-indolenine]-2-one 6bb-II: mp 213-214 °C; IR υ (cm^{-1}) 1777, 1760, 1588, 1517; ^{1}H NMR (500 MHz) δ (ppm) 7.37 (d, J = 8.1 Hz, 3H), 7.26 (d, J = 8.1 Hz, 1H), 7.15 (s, 1H), 7.14 (d, J = 8.2 Hz, 2H), 6.99 (d, J = 8.3 Hz, 2H), 6.91 (d, J = 8.4 Hz, 2H), 6.60 (s, 1H), 2.37 (s, 3H), 2.33 (s, 3H), 2.26 (s, 3H), 1.39 (s, 3H), 1.38 (s, 3H), 1.34 (s, 3H), 0.76 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 176.1, 169.9, 169.6, 149.2, 134.9, 134.5, 134.2, 134.1, 133.6, 130.9, 130.5, 130.0, 129.6, 125.9, 119.7, 117.4, 116.7, 87.0, 73.2, 59.0, 56.4, 22.1, 21.4, 20.94, 20.86, 20.2, 19.1, 16.1; MS (EI) 91 (55), 133 (80), 160 (100), 507 (M^+, 13)/508 (4). Anal. Calcd for $C_{32}H_{33}N_3O_3$: C 75.71, H 6.55, N 8.28. Found: C 75.28, H 6.30, N 8.25.

1′-(*p*-Chlorophenyl)carbamoyl-5′-chloro-3,3-dimethyl-1-(*p*-methylphenyl)spiro[azetidine-4,3′-indoline]-2,2′-dione 4bc: mp 206−207 °C; IR υ (cm^{-1}) 3251, 1769, 1745, 1598, 1549; ^{1}H NMR (500 MHz) δ (ppm) 10.45 (s, 1H), 8.42 (d, J = 8.8 Hz, 1H), 7.56 (d, J = 8.7 Hz, 2H), 7.50 (d, J = 8.8 Hz, 1H), 7.37 (d, J = 8.4 Hz, 2H), 7.36 (s, 1H), 7.07 (d, J = 8.2 Hz, 2H), 6.96 (d, J = 8.2 Hz, 2H), 2.28 (s, 3H), 1.53 (s, 3H), 1.38 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 176.3, 169.0, 148.4, 138.5, 135.1, 134.8, 133.6, 131.1, 130.9, 130.1, 130.0, 129.3, 125.5, 124.2, 121.7, 118.3, 117.0, 62.1, 48.8, 20.89, 20.87, 18.3; MS (EI) 153 (82), 192 (85), 207 (85), 242 (90), 270 (100), 340 (85), 493 (M^+, 5%). Anal. Calcd for $C_{26}H_{21}Cl_2N_3O_3$: C 63.17, H 4.28, N 8.50. Found: C 63.18, H 4.62, N 8.34.

(*R*,*R*)- and (*S*,*S*)-1′-(3″,3″-dimethyl-1″-methylphenylazetidin-2″-one)-5′-chloro-3,3,-dimethyl-1-methylphenylspiro[azetidine-4,3′-indoline]-2,2′-dione 5bc-I: mp 291−292 °C; IR υ (cm^{-1}) 1758, 1725, 1519, 1489; ^{1}H NMR (500 MHz) δ (ppm) 7.18 (d, J = 1.5 Hz, 1H), 7.14 (d, J = 8.3 Hz, 3H), 7.05 (d, J = 8.2 Hz, 2H), 6.95 (d, J = 8.4 Hz, 3H), 6.87 (d, J = 8.3 Hz, 2H), 5.93 (s, 1H), 2.23 (s, 3H), 2.19 (s, 3H), 1.49 (s, 3H), 1.48 (s, 3H), 1.22 (s, 3H), 1.19 (s, 3H); ^{13}C NMR (75 MHz) δ (ppm) 174.2, 169.6, 169.1, 139.8, 134.9, 134.6, 134.4, 134.0, 130.8, 130.2, 129.8, 128.8, 126.2, 125.0, 116.9, 115.9, 113.3, 69.9, 68.8, 60.7, 57.3, 22.4, 21.1, 20.9, 20.8, 18.5, 16.3; MS (EI) 160 (100), 527 (M^+, 10%)/529 (3). Anal. Calcd for $C_{31}H_{30}ClN_3O_3$: C 70.51, H 5.73, N 7.96. Found: C 70.56, H 6.09, N 7.85.

(*R*,*S*)- and (*S*,*R*)-1′-(3″,3″-dimethyl-1″-methylphenylazetidin-2″-one)-5′-chloro-3,3,-dimethyl-1-methylphenylspiro[azetidine-4,3′-indoline]-2,2′-dione 5bc-II: mp 248−249 °C; IR υ (cm^{-1}) 1763, 1739, 1519; ^1H NMR (500 MHz) δ (ppm) 7.07−7.16 (m, 4H), 7.04 (d, J = 8.4 Hz, 2H), 6.98 (d, J = 8.4 Hz, 1H), 6.92 (d, J = 8.5 Hz, 2H), 6.80 (d, J = 8.4 Hz, 2H), 6.14 (br s, 1H), 2.26 (s, 3H), 2.22 (s, 3H), 1.51 (s, 3H), 1.42 (br s, 3H), 1.25 (s, 3H), 1.22 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 174.8, 169.5, 169.2, 139.7, 134.8, 134.4, 134.3, 134.1, 130.7, 130.2, 129.8, 128.9, 126.1, 124.9, 116.9, 116.1, 113.6, 68.7 (4-C and 4″-C), 60.5, 56.7, 22.8, 21.05, 21.02, 20.95, 18.4, 16.6; MS (EI) 394 (65), 457 (100)/459(35), 527 (M^+,6%)/529 (2). Anal. Calcd for $C_{31}H_{30}ClN_3O_3$: C 70.51, H 5.73, N 7.96. Found: C 70.51, H 5.93, N 7.83.

(*R*,*R*)- and (*S*,*S*)-2′-(3″,3″-dimethyl-1″-methylphenylazetidin-2″-one)oxy-5′-chloro-3,3-dimethyl-1-methylphenylspiro[azetidine-4,3′-indolenine]-2-one 6bc-I: mp 225−226 °C; IR υ (cm^{-1}) 1765, 1578, 1517; ^1H NMR (500 MHz) δ (ppm) 7.44 (d, J = 8.2 Hz, 1H), 7.42 (d, J = 8.2 Hz, 1H), 7.35 (s, 1H), 7.10 (d, J = 8.3 Hz, 2H), 6.94 (d, J = 8.4 Hz, 2H), 6.76 (d, J = 8.4 Hz, 2H), 6.70 (d, J = 8.3 Hz, 2H), 5.97 (s, 1H), 2.35 (s, 3H), 2.23 (s, 3H), 1.59 (s, 3H), 1.57 (s, 3H), 1.45 (s, 3H), 1.39 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 176.5, 169.33, 169.28, 150.1, 135.0, 134.2, 134.0, 132.6, 130.7, 130.3, 130.1, 129.7, 129.5, 125.4, 121.3, 116.5, 116.1, 90.4, 73.3, 59.3, 56.8, 21.9, 21.0 (2 CH_3), 19.8, 19.7, 17.3; MS (EI) 90 (100), 118 (75), 160 (75), 269 (40), 324 (30), 527 (M^+, 15%)/529 (5%). Anal. Calcd for $C_{31}H_{30}ClN_3O_3$: C 70.51, H 5.73, N 7.96. Found: C 70.18, H 5.91, N 7.83.

The mixture of two diastereoisomers 6bc-II and 6bc-I (6bc-II:6bc-I 2:1): IR υ (cm^{-1}) 1767, 1582, 1517; ^1H NMR (500 MHz) δ (ppm) 7.40−7.45 (m, 3H, I + II), 7.32−7.36 (m, 3.5H, I + II), 7.15 (d, J = 8.2 Hz, 2H, II), 7.10 (d, J = 8.3 Hz, 1H, I), 7.00 (d, J = 8.3 Hz, 2H, II), 6.94 (d, J = 8.4 Hz, 1H, I), 6.88 (d, J = 8.4 Hz, 2H, II), 6.76 (d, J = 8.4 Hz, 1H, I), 6.70 (d, J = 8.3 Hz, 1H, I), 6.60 (s, 1H, II), 5.97 (s, 0.5H, I), 2.35 (s, 1.5H, I), 2.33 (s, 3H, II), 2.26 (s, 3H, II), 2.23 (s, 1.5H, I), 1.59 (s, 1.5H, I), 1.52 (s, 1.5H, I), 1.44 (s, 1.5H, I), 1.40 (s, 3H, II), 1.39 (s, 3H, II), 1.34 (s, 3H, II), 0.76 (s, 3H, II); ^{13}C NMR (125 MHz) δ (ppm) 176.9, 176.5, 169.5, 169.4, 150.1, 134.7, 134.6, 134.4, 133.5, 132.3, 130.7, 130.6, 130.3, 130.1, 129.9, 129.7, 129.5, 125.4, 125.3, 121.3, 121.1, 117.3, 117.0, 116.6, 116.1, 111.8, 90.4, 87.3, 73.2, 59.6, 59.3, 56.8, 56.5, 22.2, 21.8, 21.2, 21.1, 21.0, 20.9, 20.2, 19.7, 19.0, 18.6, 18.1, 17.3, 16.1; MS (EI) 159 (100)/160 (55), 527 (M^+, 10%)/529 (3).

(*R*,*R*)- and (*S*,*S*)-1′-(3″,3″-dimethyl-1″-methoxyphenylazetidin-2″-one)-3,3,-dimethyl-1-methoxyphenylspiro[azetidine-4,3′-indoline]-2,2′-dione 5ca-I: mp 220−221 °C; IR υ (cm^{-1}) 1759, 1729, 1514; ^1H NMR (500 MHz) δ (ppm) 7.31 (d, J = 8.5 Hz, 3H), 7.25 (d, J = 7.6 Hz, 1H), 7.11 (d, J = 8.1 Hz, 1H), 7.07 (d, J = 7.5 Hz, 1H), 7.03 (d, J = 8.7 Hz, 2H), 6.87 (d, J = 8.8 Hz, 2H), 6.74 (d, J = 8.9 Hz, 2H), 6.02 (s, 1H), 3.79 (s, 3H), 3.75 (s, 3H), 1.58 (s, 3H), 1.57 (s, 3H), 1.28 (s, 3H), 1.26 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 174.9, 169.54, 169.48, 156.8, 156.5, 141.4, 130.9, 130.7, 130.1, 126.1, 123.0, 118.7, 117.5, 114.8, 114.6, 114.4, 112.5, 70.1, 69.3, 60.3, 57.2, 55.5 (2 OCH_3), 22.5, 21.1, 18.5, 16.3; MS (EI) 134 (40), 149 (60), 176 (45), 227 (100), 525 (M^+, 20%)/526 (5). Anal. Calcd for $C_{31}H_{31}N_3O_5$: C 70.84, H 5.94, N 7.99. Found: C 70.62, H 6.40, N 7.80.

(*R*,*S*)- and (*S*,*R*)-1′-(3″,3″-dimethyl-1″-methoxyphenylazetidin-2″-one)-3,3,-dimethyl-1-methoxyphenylspiro[azetidine-4,3′-indoline]-2,2′-dione 5ca-II: mp 268−269 °C; IR υ (cm^{-1}) 1762, 1721, 1513; ^1H NMR (500 MHz) δ (ppm) 7.25−7.30 (m, 4H), 7.14 (d, J = 7.8 Hz, 1H), 7.04 (t, J = 6.7 Hz, 1H), 6.94 (d, J = 8.7 Hz, 2H), 6.82 (d, J = 8.7 Hz, 2H), 6.71 (d, J = 8.8 Hz, 2H), 6.23 (br s, 1H), 3.78 (s, 3H), 3.75 (s, 3H), 1.60 (s, 3H), 1.51 (br s, 3H), 1.34 (s, 6H); ^{13}C NMR (125 MHz) δ (ppm) 175.5, 169.5, 169.3, 156.7, 156.4, 141.3, 130.7, 130.3, 130.2, 126.0, 123.0, 122.8, 118.7, 117.6, 114.7, 114.4, 112.7, 69.2, 68.8, 60.2, 56.6, 55.4 (2 OCH_3), 23.0, 21.0, 18.4, 16.6; MS (EI) 149 (98), 176 (70), 227 (100), 525 (M^+, 45%)/526 (15). Anal. Calcd for $C_{31}H_{31}N_3O_5$: C 70.84, H 5.94, N 7.99. Found: C 70.94, H 6.30, N 7.99.

(*R*,*R*)- and (*S*,*S*)-2′-(3″,3″-dimethyl-1″-methoxyphenylazetidin-2″-one)oxy-3,3-dimethyl-1-methoxyphenylspiro[azetidine-4,3′-indolenine]-2-one 6ca-I: mp 193−194 °C; IR υ (cm^{-1}) 1763, 1577, 1513; ^1H NMR (500 MHz) δ (ppm) 7.46−7.49 (m, 2H), 7.38 (d, J = 7.4 Hz, 1H), 7.19 (t, J = 7.2 Hz, 1H), 7.00 (d, J = 9.0 Hz, 2H), 6.83 (d, J = 8.9 Hz, 2H), 6.79 (d, J = 9.0 Hz, 2H), 6.48 (d, J = 8.9 Hz, 2H), 6.01 (s, 1H), 3.77 (s, 3H), 3.73 (s, 3H), 1.61 (s, 3H), 1.58 (s, 3H), 1.44 (s, 3H), 1.40 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 176.3, 169.6, 169.2, 156.3, 156.2, 151.7, 131.1, 130.7, 130.6, 130.2, 125.1, 124.5, 120.4, 118.0, 117.6, 114.7, 114.3, 90.2, 73.5, 58.9, 56.8, 55.4, 55.3, 21.8, 19.80, 19.77, 17.3; MS (EI) 134 (75), 149 (95), 176 (100), 227 (100), 525 (M^+, 50%)/ 526 (17). Anal. Calcd for $C_{31}H_{31}N_3O_5$: C 70.84, H 5.94, N 7.99. Found: C 70.95, H 6.10, N 7.95.

(*R*,*S*)- and (*S*,*R*)-2′-(3″,3″-dimethyl-1″-methoxyphenylazetidin-2″-one)oxy-3,3-dimethyl-1-methoxyphenylspiro[azetidine-4,3′-indolenine]-2-one 6ca-II: mp 144−145 °C; IR υ (cm^{-1}) 1768, 1588, 1514; ^1H NMR (500 MHz) δ (ppm) 7.45−7.48 (m, 2H), 7.40 (d, J = 8.7 Hz, 2H), 7.35 (d, J = 7.4 Hz, 1H), 7.18 (t, J = 7.2 Hz, 1H), 6.94 (d, J = 8.8 Hz, 2H), 6.86 (d, J = 8.8 Hz, 2H), 6.71 (d, J = 8.9 Hz, 2H), 6.60 (s, 1H), 3.79 (s, 3H), 3.72 (s, 3H), 1.38 (s, 6H), 1.31 (s, 3H), 0.76 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 176.8, 169.6, 169.3, 156.8, 156.4, 151.7, 130.8, 130.6, 130.4, 129.4, 125.1, 124.5, 120.2, 119.0, 118.1, 114.39, 114.36, 87.2, 73.4, 59.2, 56.4, 55.5, 55.4, 22.1, 20.2, 19.0, 16.1; MS (EI) 134 (100), 145 (85), 176 (100), 227 (45), 525 (M^+,35%)/526 (12). Anal. Calcd for $C_{31}H_{31}N_3O_5$: C 70.84, H 5.94, N 7.99. Found: C 70.51, H 6.26, N 7.92.

(*R*,*R*)- and (*S*,*S*)-1′-(3″,3″-dimethyl-1″-methoxyphenylazetidin-2″-one)-3,3,5′-trimethyl-1-methoxyphenylspiro[azetidine-4,3′-indoline]-2,2′-dione 5cb-I: mp 202−203 °C; IR υ (cm^{-1}) 1765, 1722, 1514; ^1H NMR (300 MHz) δ (ppm) 7.30 (d, J = 9.0 Hz, 2H), 7.11 (s, 1H), 7.04 (d, J = 8.9 Hz, 3H), 6.99 (d, J = 8.2 Hz, 1H), 6.86 (d, J = 8.9 Hz, 2H), 6.75 (d, J = 9.0 Hz, 2H), 6.00 (s, 1H), 3.78 (s, 3H), 3.74 (s, 3H), 2.26 (s, 3H), 1.57 (s, 3H), 1.56 (s, 3H), 1.29 (s, 3H), 1.26 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 174.8, 169.6, 169.5, 156.7, 156.4, 138.9, 132.7, 131.3, 130.7, 130.2, 126.7, 123.0, 118.6, 117.5, 114.8, 114.4, 112.3, 70.0, 69.3, 60.2, 57.2, 55.5 (2 OCH_3), 22.5, 21.1, 21.0, 18.6, 16.3; MS (EI) 134 (62), 149 (95), 241 (35), 539 (100)/540 (30). Anal. Calcd for $C_{32}H_{33}N_3O_5$: C 71.23, H 6.16, N 7.79. Found: C 71.13, H 6.51, N 7.73.

(*R*,*S*)- and (*S*,*R*)-1′-(3″,3″-dimethyl-1″-methoxyphenylazetidin-2″-one)-3,3,5′-trimethyl-1-methoxyphenylspiro[azetidine-4,3′-indoline]-2,2′-dione 5cb-II: mp 235−236 °C; IR υ (cm^{-1}) 1762, 1721, 1513; ^1H NMR (500 MHz) δ (ppm) 7.26 (d, J = 8.9 Hz, 2H), 7.09 (s, 1H), 7.03 (br s, 2H), 6.95 (d, J = 8.2 Hz, 2H), 6.83 (d, J = 8.9 Hz, 2H), 6.72 (d, J = 9.0 Hz, 2H), 6.21 (br s, 1H), 3.79 (s, 3H), 3.77 (s, 3H), 2.26 (s, 3H), 1.59 (s, 3H), 1.50 (br s, 3H), 1.33 (s, 6H); ^{13}C NMR (125 MHz) δ (ppm) 175.4, 169.6 (2 CO), 156.6, 156.3, 138.8, 132.7, 131.2, 130.4, 130.3, 126.6, 122.8, 118.6, 117.6, 114.7, 114.4, 112.5, 69.2, 68.7, 60.0, 56.6, 55.4 (2 OCH_3), 23.0, 21.04, 20.98, 18.5, 16.6; MS (EI) 134 (60), 149 (100), 539 (M^+, 90)/540 (25). Anal. Calcd for $C_{32}H_{33}N_3O_5$: C 71.23, H 6.16, N 7.79. Found: C 71.14, H 6.04, N 7.96.

(*R*,*R*)- and (*S*,*S*)-2′-(3″,3″-dimethyl-1″-methoxyphenylazetidin-2″-one)oxy-3,3,5′-trimethyl-1-methoxyphenylspiro[azetidine-4,3′-indolenine]-2-one 6cb-I: mp 219−220 °C; IR υ (cm^{-1}) 1767, 1585, 1514; ^1H NMR (500 MHz) δ (ppm) 7.35 (d, J = 7.8 Hz, 1H), 7.24 (d, J = 7.8 Hz, 1H), 7.18 (s, 1H), 7.01 (d, J = 9.0 Hz, 2H), 6.82 (d, J = 9.4 Hz, 2H), 6.80 (d, J = 9.4 Hz, 2H), 6.48 (d, J = 9.0 Hz, 2H), 5.99 (s, 1H), 3.77 (s, 3H), 3.73 (s, 3H), 2.38 (s, 3H), 1.59 (s, 3H), 1.57 (s, 3H), 1.44 (s, 3H), 1.39 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 175.6, 169.6, 169.2, 156.3, 156.2, 149.2, 134.3, 131.2, 131.0, 130.7, 130.2, 125.9, 120.0, 118.0, 117.6, 114.7, 114.4, 90.0, 73.4, 58.8, 56.7, 55.4, 55.3, 21.8, 21.5, 19.8 (2 CH_3), 17.3; MS (EI) 134 (60), 149 (52), 176 (100), 241 (62), 539 (M^+,

40%)/540 (20). Anal. Calcd for $C_{32}H_{33}N_3O_5$: C 71.23, H 6.16, N 7.79. Found: C 71.20, H 6.41, N 7.78.

(*R*,*S*)- and (*S*,*R*)-2′-(3″,3″-dimethyl-1″-methoxyphenylazetidin-2″-one)oxy-3,3,5′-trimethyl-1-methoxyphenylspiro[azetidine-4,3′-indolenine]-2-one 6cb-II: mp 211−212 °C; IR υ (cm^{-1}) 1771, 1589, 1513; ^1H NMR (300 MHz) δ (ppm) 7.42 (d, J = 9.0 Hz, 2H), 7.36 (d, J = 7.8 Hz, 1H), 7.25 (d, J = 7.9 Hz, 1H), 7.17 (s, 1H), 6.97 (d, J = 9.0 Hz, 2H), 6.88 (d, J = 9.0 Hz, 2H), 6.73 (d, J = 9.0 Hz, 2H), 6.58 (s, 1H), 3.80 (s, 3H), 3.73 (s, 3H), 2.38 (s, 3H), 1.39 (s, 3H), 1.38 (s, 3H), 1.32 (s, 3H), 0.76 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 176.1, 169.7, 169.4, 156.8, 156.4, 149.2, 134.2, 131.0, 130.8, 130.4, 129.5, 125.9, 119.8, 119.0, 118.1, 114.5, 114.4, 87.1, 73.3, 59.1, 56.4, 55.5, 55.4, 22.1, 21.4, 20.2, 19.0, 16.1; MS (EI) 176 (100), 187 (60), 241 (37), 539 (M^+, 5%)/540 (3). Anal. Calcd for $C_{32}H_{33}N_3O_5$: C 71.23, H 6.16, N 7.79. Found: C 70.76, H 6.54, N 7.72.

(*R*,*R*)- and (*S*,*S*)-1′-(3″,3″-dimethyl-1″-methoxyphenylazetidin-2″-one)-5′-chloro-3,3,-dimethyl-1-methoxyphenylspiro[azetidine-4,3′-indoline]-2,2′-dione 5cc-I: mp 239−240 °C; IR υ (cm^{-1}) 1763, 1733, 1513; ^1H NMR (300 MHz) δ (ppm) 7.22−7.29 (m, 4H), 7.02 (dd, J = 8.7, 2.8 Hz, 3H), 6.87 (d, J = 8.9 Hz, 2H), 6.76 (d, J = 8.9 Hz, 2H), 5.99 (s, 1H), 3.78 (s, 3H), 3.75 (s, 3H), 1.58 (s, 3H), 1.56 (s, 3H), 1.31 (s, 3H), 1.24 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 174.4, 169.3, 169.0, 156.9, 156.6, 139.9, 130.9, 130.5, 129.8, 128.9, 126.2, 125.0, 118.6, 117.4, 114.9, 114.6, 113.4, 70.1, 69.1, 60.8, 57.3, 55.5 (2 OCH_3), 22.4, 21.2, 18.5, 16.3; MS (EI) 134 (100), 149 (50), 176 (80), 559 (M^+, 10%)/561(4). Anal. Calcd for $C_{31}H_{30}ClN_3O_5$: C 66.48, H 5.40, N 7.50. Found: C 66.16, H 5.37, N 7.44.

(*R*,*S*)- and (*S*,*R*)-1′-(3″,3″-dimethyl-1″-methoxyphenylazetidin-2″-one)-5′-chloro-3,3,-dimethyl-1-methoxyphenylspiro[azetidine-4,3′-indoline]-2,2′-dione 5cc-II: mp 217−219 °C; IR υ (cm^{-1}) 1764, 1734, 1515; ^1H NMR (500 MHz) δ (ppm) 7.27 (s, 1H), 7.24 (d, J = 9.0 Hz, 3H), 7.07 (d, J = 8.5 Hz, 1H), 6.93 (d, J = 9.0 Hz, 2H), 6.84 (d, J = 9.0 Hz, 2H), 6.74 (d, J = 9.0 Hz, 2H), 6.21 (br s, 1H), 3.80 (s, 3H), 3.78 (s, 3H), 1.60 (s, 3H), 1.51 (br s. 3H), 1.35 (s, 3H), 1.32 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 175.0, 169.2, 169.0, 156.8, 156.6, 139.8, 130.8, 130.1, 129.9, 128.9, 126.1, 124.9, 118.9, 118.6, 117.5, 114.8, 114.5, 113.6, 69.0, 68.8, 60.7, 56.7, 55.5 (2 OCH_3), 22.9, 21.1, 18.4, 16.6; MS (EI) 49 (100)/51 (55), 134 (45), 149 (75), 559 (M^+, 3%). Anal. Calcd for $C_{31}H_{30}ClN_3O_5$: C 66.48, H 5.40, N 7.50. Found: C 66.27, H 5.61, N 7.50.

(*R*,*R*)- and (*S*,*S*)-2′-(3″,3″-dimethyl-1″-methoxyphenylazetidin-2″-one)oxy-5′-chloro-3,3-dimethyl-1-methoxyphenylspiro-[azetidine-4,3′-indolenine]-2-one 6cc-I: mp 205−206 °C; IR υ (cm^{-1}) 1764, 1584, 1514; ^1H NMR (500 MHz) δ (ppm) 7.44 (d, J = 8.2 Hz, 1H), 7.40 (d, J = 8.2 Hz, 1H), 7.36 (s, 1H), 6.99 (d, J = 8.8 Hz, 2H), 6.81 (dd, J = 9.0, 2.5 Hz, 4H), 6.48 (d, J = 8.8 Hz, 2H), 5.99 (s, 1H), 3.79 (s, 3H), 3.73 (s, 3H), 1.60 (s, 3H), 1.57 (s, 3H), 1.45 (s, 3H), 1.39 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 176.6, 169.1, 169.0, 156.5, 156.3, 150.1, 132.5, 130.9, 130.7, 130.3, 130.1, 125.4, 121.3, 118.0, 117.5, 114.8, 114.3, 90.4, 73.5, 59.5, 56.8, 55.4, 55.3, 21.9, 19.8, 19.7, 17.3; MS (EI) 134(73), 176 (100), 261 (65), 559 (M^+, 15%)/561 (5). Anal. Calcd for $C_{31}H_{30}ClN_3O_5$: C 66.48, H 5.40, N 7.50. Found: C 66.21, H 5.30, N 7.35.

(*R*,*S*)- and (*S*,*R*)-2′-(3″,3″-dimethyl-1″-methoxyphenylazetidin-2″-one)oxy-5′-chloro-3,3-dimethyl-1-methoxyphenylspiro-[azetidine-4,3′-indolenine]-2-one 6cc-II: mp 206−207 °C; IR υ (cm^{-1}) 1773, 1587, 1513; ^1H NMR (500 MHz) δ (ppm) 7.39−7.45 (m, 4H), 7.37 (s, 1H), 6.94 (d, J = 9.0 Hz, 2H), 6.88 (d, J = 8.9 Hz, 2H), 6,74 (d, J = 9.0 Hz, 2H), 6.59 (s, 1H), 3.81 (s, 3H), 3.75 (s, 3H), 1.40 (s, 3H), 1.39 (s, 3H), 1.32 (s, 3H), 0.77 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 177.0, 169.21, 169.16, 156.9, 156.6, 150.1, 132.3, 130.6, 130.5, 130.2, 129.3, 125.3, 121.1, 119.0, 118.1, 114.5, 114.4, 87.5, 73.4, 59.8, 56.5, 55.50, 55.48, 22.2, 20.2, 18.9, 16.1; MS (EI) 134 (53), 176 (100), 559 (M^+, 30)/561 (30). Anal. Calcd for $C_{31}H_{30}ClN_3O_5$: C 66.48, H 5.40, N 7.50. Found: C 66.21, H 5.67, N 7.41.

(*R*,*R*)- and (*S*,*S*)-1′-(3″,3″-dimethyl-1″-chlorophenylazetidin-2″-one)-3,3,5′-trimethyl-1-chlorophenylspiro[azetidine-4,3′-indoline]-2,2′-dione 5db-I: mp 252−253 °C; IR υ (cm^{-1}) 1764, 1727, 1596, 1496; ^1H NMR (300 MHz) δ (ppm) 7.30 (s, 4H), 7.18 (d, J = 8.9 Hz, 2H), 7.11 (s, 1H), 7.07 (d, J = 8.3 Hz, 1H), 7.02 (d, J = 8.9 Hz, 2H), 6.94 (d, J = 8.2 Hz, 1H), 6.02 (s, 1H), 2.28 (s, 3H), 1.59 (s, 3H), 1.56 (s, 3H), 1.31 (s, 3H), 1.28 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 174.4, 169.8, 169.7, 138.6, 135.7, 135.3, 133.1, 131.6, 130.2, 129.7, 129.6, 129.3, 126.8, 122.4, 118.2, 117.3, 112.2, 69.9, 69.2, 60.5, 57.6, 22.5, 21.1, 21.0, 18.5, 16.3; MS (EI) 70 (100), 153 (76), 180 (45), 547 (30)/ 549 (20). Anal. Calcd for $C_{30}H_{27}Cl_2N_3O_3$: C 65.70, H 4.96, N 7.66. Found: C 65.59, H 5.01, N 7.64.

(*R*,*S*)- and (*S*,*R*)-1′-(3″,3″-dimethyl-1″-chlorophenylazetidin-2″-one)-3,3,5′-trimethyl-1-chlorophenylspiro[azetidine-4,3′-indoline]-2,2′-dione 5db-II: mp 296−297 °C; IR υ (cm^{-1}) 1762, 1731, 1595, 1498; ^1H NMR (500 MHz) δ (ppm) 7.25−7.28 (m, 4H), 7.14 (d, J = 8.6 Hz, 2H), 7.07 (m, 2H), 6.98 (d, J = 8.1 Hz, 1H), 6.92 (d, J = 8.5 Hz, 2H), 6.23 (br s, 1H), 2.27 (s, 3H), 1.61 (s, 3H), 1.51 (br s, 3H), 1.34 (s, 6H); ^{13}C NMR (125 MHz) δ (ppm) 175.2, 169.8 (2 CO), 138.5, 135.33, 135.29, 133.1, 131.5, 130.1, 129.7, 129.6, 129.3, 126.7, 122.3, 118.2, 117.5, 112.4, 69.2, 68.6, 60.5, 57.0, 23.0, 21.0 (2 CH_3), 18.4, 16.6; MS (EI) 70 (100), 153 (90), 180 (60), 547 (72)/549 (52). Anal. Calcd for $C_{30}H_{27}Cl_2N_3O_3$: C 65.70, H 4.96, N 7.66. Found: C 65.73, H 5.31, N 7.63.

(*R*,*R*)- and (*S*,*S*)-2′-(3″,3″-dimethyl-1″-chlorophenylazetidin-2″-one)oxy-3,3,5′-trimethyl-1-chlorophenylspiro[azetidine-4,3′-indolenine]-2-one 6db-I: mp 244−246 °C; IR υ (cm^{-1}) 1777, 1763, 1581, 1497; ^1H NMR (500 MHz) δ (ppm) 7.37 (d, J = 7.8 Hz, 1H), 7.27 (d, J = 7.8 Hz, 1H), 7.25 (d, J = 8.8 Hz, 2H), 7.18 (s, 1H), 6.99 (d, J = 8.8 Hz, 2H), 6.94 (d, J = 8.7 Hz, 2H), 6.80 (d, J = 8.7 Hz, 2H), 6.02 (s, 1H), 2.39 (s, 3H), 1.58 (s, 6H), 1.45 (s, 3H), 1.40 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 175.0, 170.0, 169.5, 149.0, 136.0, 135.0, 134.7, 131.3, 130.1, 129.7, 129.6, 129.3, 129.1, 125.9, 120.2, 117.8, 117.5, 89.7, 73.4, 59.2, 57.1, 21.8, 21.5, 19.8, 19.7, 17.2; MS (EI) 138 (62), 144 (99), 180 (100), 269 (15), 547 (M^+, 3%). Anal. Calcd for $C_{30}H_{27}Cl_2N_3O_3$: C 65.70, H 4.96, N 7.66. Found: C 65.76, H 5.20, N 7.44.

(*R*,*S*)- and (*S*,*R*)-2′-(3″,3″-dimethyl-1″-chlorophenylazetidin-2″-one)oxy-3,3,5′-trimethyl-1-chlorophenylspiro[azetidine-4,3′-indolenine]-2-one 6db-II: mp 178−179 °C; IR υ (cm^{-1}) 1780, 1771, 1587, 1496; ^1H NMR (500 MHz) δ (ppm) 7.45 (d, J = 8.7 Hz, 2H), 7.38 (d, J = 7.8 Hz, 1H), 7.32 (d, J = 8.8 Hz, 2H), 7.27 (d, J = 7.8 Hz, 1H), 7.17 (d, J = 8.8 Hz, 2H), 7.15 (s, 1H), 6.96 (d, J = 8.8 Hz, 2H), 6.62 (s, 1H), 2.39 (s, 3H), 1.41 (s, 3H), 1.39 (s, 3H), 1.32 (s, 3H), 0.79 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 175.6, 170.0, 169.5, 148.9, 135.7, 134.60, 134.56, 131.3, 130.1, 129.8, 129.5, 129.3 (2 C), 125.9, 120.0, 118.6, 117.9, 86.8, 73.2, 59.5, 56.7, 22.1, 21.5, 20.2, 19.0, 16.1; MS (EI) 144 (80), 180 (100), 269 (20), 547 (M^+, 15%)/549 (10). Anal. Calcd for $C_{30}H_{27}Cl_2N_3O_3$: C 65.70, H 4.96, N 7.66. Found: C 65.69, H 4.98, N 7.57.

(*R*,*R*)- and (*S*,*S*)-1′-(3″,3″-dimethyl-1″-bromophenylazetidin-2″-one)-3,3,5′-trimethyl-1-bromophenylspiro[azetidine-4,3′-indoline]-2,2′-dione 5eb-I: mp 268−269 °C; IR υ (cm^{-1}) 1772, 1725, 1593, 1490; ^1H NMR (500 MHz) δ (ppm) 7.46 (d, J = 8.4 Hz, 2H), 7.33 (d, J = 8.4 Hz, 2H), 7.24 (d, J = 8.4 Hz, 2H), 7.11 (s, 1H), 7.05 (d, J = 8.7 Hz, 1H), 6.96 (d, J = 8.8 Hz, 2H), 6.94 (d, J = 8.3 Hz, 1H), 6.02 (s, 1H), 2.28 (s, 3H), 1.60 (s, 3H), 1.55 (s, 3H), 1.31 (s, 3H), 1.28 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 174.3, 169.9, 169.7, 138.6, 136.1, 135.7, 133.2, 132.7, 132.2, 131.6, 126.8, 122.4, 118.5, 117.6, 117.4, 117.2, 112.2, 69.9, 69.1, 60.6, 57.7, 22.5, 21.1, 21.0, 18.5, 16.3; MS (EI) 70 (100), 143 (35), 197 (35)/199 (32), 635 (M^+, 5%)/637 (10)/639 (5). Anal. Calcd for $C_{30}H_{27}N_3O_3Br_2$: C 56.53, H 4.27, N 6.59. Found: C 56.74, H 4.55, N 6.55.

(*R*,*S*)- and (*S*,*R*)-1′-(3″,3″-dimethyl-1″-bromophenylazetidin-2″-one)-3,3,5′-trimethyl-1-bromophenylspiro[azetidine-4,3′-indoline]-2,2′-dione 5eb-II: mp 284−285 °C; IR υ (cm^{-1}) 1761,

1730, 1596, 1494; ^{1}H NMR (500 MHz) δ (ppm) 7.42 (d, J = 8.7 Hz, 2H), 7.29 (d, J = 6.8 Hz, 2H), 7.20 (d, J = 8.7 Hz, 2H), 7.08 (m, 2H), 6.97 (d, J = 8.4 Hz, 1H), 6.85 (d, J = 8.5 Hz, 2H), 6.22 (br s, 1H), 2.27 (s, 3H), 1.61 (s, 3H), 1.50 (br s, 3H), 1.33 (s, 6H); ^{13}C NMR (125 MHz) δ (ppm) 175.2, 169.8 (2 CO), 138.4, 135.8, 133.2, 132.7, 132.2, 131.5, 126.7, 122.3, 118.5, 117.8, 117.2, 112.4, 69.2, 68.6, 60.5, 57.0, 23.0, 21.0, 18.4, 16.6; MS (EI) 70 (100), 143 (65), 197 (45)/199 (45), 635 (M^+, 25%)/637 (55)/639 (22). Anal. Calcd for $C_{30}H_{27}N_3O_3Br_2$: C 56.53, H 4.27, N 6.59. Found: C 56.74, H 4.55, N 6.55.

(*R*,*R*)- and (*S*,*S*)-2′-(3″,3″-dimethyl-1″-bromophenylazetidin-2″-one)oxy-3,3,5′-trimethyl-1-bromophenylspiro[azetidine-4,3′-indolenine]-2-one 6eb-I: mp 249−250 °C; IR υ (cm^{-1}) 1777, 1762, 1581, 1493; ^{1}H NMR (300 MHz) δ (ppm) 7.38 (d, J = 8.8 Hz, 2H), 7.37 (d, J = 7.9 Hz, 1H), 7.27 (d, J = 9.1 Hz, 1H), 7.17 (s, 1H), 7.10 (d, J = 8.8 Hz, 2H), 6.93 (d, J = 8.8 Hz, 2H), 6.74 (d, J = 8.8 Hz, 2H), 6.03 (s, 1H), 2.39 (s, 3H), 1.58 (s, 6H), 1.45 (s, 3H), 1.39 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 175.0, 170.0, 169.5, 149.0, 136.5, 135.4, 134.7, 132.6, 132.1, 131.3, 130.1, 125.9, 120.2, 118.2, 117.8, 117.4, 117.1, 89.6, 73.4, 59.2, 57.1, 21.8, 21.5, 19.8, 19.7, 17.2; MS (EI) 144 (100), 224 (53)/226 (55), 635 (M^+, 15%)/637 (35)/639 (13). Anal. Calcd for $C_{30}H_{27}N_3O_3Br_2$: C 56.53, H 4.27, N 6.59. Found: C 56.22, H 4.39, N 6.50.

(*R*,*S*)- and (*S*,*R*)-2′-(3″,3″-dimethyl-1″-bromophenylazetidin-2″-one)oxy-3,3,5′-trimethyl-1-bromophenylspiro[azetidine-4,3′-indolenine]-2-one 6eb-II: mp 227−228 °C; IR υ (cm^{-1}) 1786, 1774, 1588, 1491; ^{1}H NMR (500 MHz) δ (ppm) 7.47 (d, J = 8.7 Hz, 2H), 7.40 (d, J = 8.9 Hz, 2H), 7.37 (d, J = 8.1 Hz, 1H), 7.32 (d, J = 8.7 Hz, 2H), 7.27 (d, J = 9.5 Hz, 1H), 7.15 (s, 1H), 6.90 (d, J = 8.7 Hz, 2H), 6.61 (s, 1H), 2.39 (s, 3H), 1.41 (s, 3H), 1.39 (s, 3H), 1.32 (s, 3H), 0.80 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 175.6, 170.0, 169.5, 148.9, 136.2, 135.0, 134.6, 132.2 (2 C), 131.3, 129.7, 125.9, 120.1, 119.0, 118.2, 117.7, 117.1, 86.8, 73.2, 59.5, 56.8, 22.0, 21.5, 20.2, 19.0, 16.1; MS (EI) 144 (100), 157 (45), 635 (M^+, 18%)/637 (35)/639 (20). Anal. Calcd for $C_{30}H_{27}N_3O_3Br_2$: C 56.53, H 4.27, N 6.59. Found: C 56.59, H 4.48, N 6.60.

(*R*,*R*)- and (*S*,*S*)-1′-(3″,3″-diphenyl-1″-chlorophenylazetidin-2″-one)-5′-chloro-3,3,-diphenyl-1-chlorophenylspiro[azetidine-4,3′-indoline]-2,2′-dione 5fc-I: mp 289−290 °C; IR υ (cm^{-1}) 1771, 1729, 1605, 1492; ^{1}H NMR δ (ppm) 7.74 (d, J = 7.4 Hz, 2H), 7.60 (d, J = 7.4 Hz, 2H), 7.41−7.48 (m, 7H), 7.34−7.37 (m, 4H), 7.30 (d, J = 8.8 Hz, 2H), 7.27 (t, J = 7.4 Hz, 1H), 7.22 (t, J = 7.4 Hz, 1H), 7.09−7.16 (m, 7H), 6.93 (br, 2H), 6.88 (dd, J = 8.6, 2.0 Hz, 1H), 6.74 (d, J = 8.6 Hz, 1H), 6.71 (s, 1H), 6.06 (d, J = 1.9 Hz, 1H); ^{13}C NMR δ (ppm) 172.7, 166.0, 165.7, 139.2, 137.8, 135.9, 134.5, 134.4, 134.1, 131.23, 131.19, 130.0, 129.8, 129.6, 129.2, 129.02, 128.97, 128.9, 128.5, 128.3, 128.19, 128.16, 128.1, 128.0, 126.8, 126.6, 124.4, 120.5, 118.7, 113.1, 103.1, 80.0, 72.3, 72.2, 70.0; MS (APCI) 279 (100), 815 (M^+, 1%)/816/817/818. Anal. Calcd for $C_{49}H_{32}Cl_3N_3O_3$: C 72.02, H 3.95, N 5.14. Found: C 71.93, H 4.04, N 5.05.

(*R*,*S*)- and (*S*,*R*)-1′-(3″,3″-diphenyl-1″-chlorophenylazetidin-2″-one)-5′-chloro-3,3,-diphenyl-1-chlorophenylspiro[azetidine-4,3′-indoline]-2,2′-dione 5fc-II: mp 170−171 °C; IR υ (cm^{-1}) 1764, 1738, 1597, 1493; ^{1}H NMR δ (ppm) 7.72 (d, J = 7.4 Hz, 2H), 7.64 (d, J = 7.3 Hz, 2H), 7.44−7.50 (m, 3H), 7.41 (d, J = 7.5 Hz, 4H), 7.33−7.38 (m, 4H), 7.25−7.31 (m, 2H), 7.23 (s, 4H), 7.17 (t, J = 7.8 Hz, 2H), 7.10 (d, J = 8.7 Hz, 2H), 7.03 (s, 2H), 6.94 (d, J = 8.8 Hz, 2H), 6.90 (dd, J = 8.7, 2.0 Hz, 1H), 6.77 (d, J = 8.7 Hz, 1H), 6.10 (d, J = 1.9 Hz, 1H); ^{13}C NMR δ (ppm) 173.4, 165.6, 165.3, 139.3, 137.8, 136.4, 134.9, 134.7, 134.3, 134.0, 130.9, 130.7, 130.1, 130.0, 129.5, 129.3, 129.2, 128.72, 128.67, 128.2, 128.12, 128.06, 127.4, 126.9, 126.6, 126.4, 124.6, 120.5, 119.1, 118.7, 118.2, 113.6, 79.0, 71.7, 71.1, 68.2; MS (APCI) 282 (100), 815 (M^+, 30%)/816/817/818. Anal. Calcd for $C_{49}H_{32}Cl_3N_3O_3$: C 72.02, H 3.95, N 5.14. Found: C 72.03, H 4.20, N 5.21.

(*R*,*R*)- and (*S*,*S*)-2′-(3″,3″-diphenyl-1″-chlorophenylazetidin-2″-one)oxy-5′-chloro-3,3-diphenyl-1-chlorophenylspiro[azetidine-4,3′-indolenine]-2-one 6fc-I: mp 244−245 °C; IR υ (cm^{-1}) 1767, 1583, 1494; ^{1}H NMR δ (ppm) 7.76 (d, J = 8.8 Hz, 2H), 7.51 (d, J = 8.2 Hz, 1H), 7.43 (d, J = 8.8 Hz, 2H), 7.35−7.40 (m, 4H), 7.24−7.32 (m, 7H), 7.17−7.18 (m, 3H), 7.13 (d, J = 8.8 Hz, 2H), 7.10 (t, J = 7.7 Hz, 2H), 7.05 (s, 1H), 7.02 (t, J = 7.4 Hz, 1H), 6.73 (t, J = 7.0 Hz, 2H), 6.68 (d, J = 8.8 Hz, 2H), 6.41 (d, J = 7.5 Hz, 2H), 5.88 (d, J = 1.8 Hz, 1H); ^{13}C NMR δ (ppm) 176.8, 165.9, 164.6, 149.1, 137.0, 136.8, 136.0, 135.3, 134.7, 134.2, 131.3, 131.0, 130.6, 130.4, 130.0, 129.3, 129.1, 128.9, 128.8, 128.5, 128.4, 128.31, 128.27, 128.2, 127.9, 127.7, 126.4, 126.3, 121.1, 119.9, 117.9, 86.1, 75.6, 74.6, 72.2; MS (APCI) 274 (100), 359 (92), 815 (M^+, 1%)/816/817/818. Anal. Calcd for $C_{49}H_{32}Cl_3N_3O_3$: C 72.02, H 3.95, N 5.14. Found: C 71.90, H 4.07, N 5.56.

(*R*,*S*)- and (*S*,*R*)-2′-(3″,3″-diphenyl-1″-chlorophenylazetidin-2″-one)oxy-5′-chloro-3,3-diphenyl-1-chlorophenylspiro[azetidine-4,3′-indolenine]-2-one 6fc-II: mp 200−201 °C; IR υ (cm^{-1}) 1767, 1617, 1596, 1493; ^{1}H NMR δ (ppm) 7.40−7.44 (m, 6H), 7.37 (t, J = 7.1 Hz, 1H), 7.34 (t, J = 7.4 Hz, 1H), 7.22−7.28 (m, 8H), 7.16−7.20 (m, 4H), 7.11−7.14 (m, 5H), 6.82 (d, J = 8.8 Hz, 2H), 6.76 (d, J = 8.8 Hz, 2H), 6.50 (d, J = 7.7 Hz, 2H), 5.80 (d, J = 1.5 Hz, 1H); ^{13}C NMR δ (ppm) 177.5, 166.2, 165.1, 149.0, 137.5, 136.9, 136.0, 134.5, 131.4, 131.12, 131.08, 130.5, 130.1, 129.2, 128.7, 128.62, 128.57, 128.5, 128.2, 128.0, 127.7, 127.2, 126.1, 125.9, 122.4, 120.7, 117.7, 85.5, 75.7, 74.8, 71.4. MS (APCI) 279 (100), 816 (M + 1, 2%)/817/818/819. Anal. Calcd for $C_{49}H_{32}Cl_3N_3O_3$: C 72.02, H 3.95, N 5.14. Found: C 72.02, H 3.95, N 5.14.

(*R*,*R*)- and (*S*,*S*)-1′-(3″,3″-diphenyl-1″-bromophenylazetidin-2″-one)-5′-chloro-3,3,-diphenyl-1-bromophenylspiro[azetidine-4,3′-indoline]-2,2′-dione 5gc-I: mp 302−303 °C; IR υ (cm^{-1}) 1770, 1729, 1605, 1589, 1490; ^{1}H NMR δ (ppm) 7.74 (d, J = 7.4 Hz, 2H), 7.60 (d, J = 7.4 Hz, 2H), 7.56 (d, J = 8.5 Hz, 2H), 7.41−7.48 (m, 8H), 7.36 (t, J = 7.4 Hz, 2H), 7.28−7.29 (m, 2H), 7.23 (t, J = 7.0 Hz, 1H), 7.11−7.15 (m, 5H), 7.06 (d, J = 8.6 Hz, 2H), 6.92 (br, 2H), 6.88 (d, J = 8.6 Hz, 1H), 6.74 (d, J = 8.6 Hz, 1H), 6.71 (s, 1H), 6.06 (s, 1H); ^{13}C NMR δ (ppm) 172.7, 166.0, 165.7, 139.1, 137.8, 135.9, 135.0, 134.3, 132.7, 132.5, 130.1, 129.2, 129.03, 128.99, 128.9, 128.5, 128.3, 128.21, 128.16, 128.09, 128.06, 126.8, 126.6, 124.4, 120.7, 119.0, 118.9, 118.8, 113.1, 80.0, 72.2, 70.0; MS (TOF) 926 (M + Na^+), 942 (M + K^+). Anal. Calcd for $C_{49}H_{32}Br_2ClN_3O_3$: C 64.95, H 3.56, N 4.64. Found: C 65.01, H 3.73, N 4.67.

(*R*,*S*)- and (*S*,*R*)-1′-(3″,3″-diphenyl-1″-bromophenylazetidin-2″-one)-5′-chloro-3,3,-diphenyl-1-bromophenylspiro[azetidine-4,3′-indoline]-2,2′-dione 5gc-II: mp 192−193 °C; IR υ (cm^{-1}) 1767, 1752, 1741, 1591, 1491; ^{1}H NMR δ (ppm) 7.72 (d, J = 7.4 Hz, 2H), 7.65 (d, J = 7.3 Hz, 2H), 7.49 (t, J = 7.6 Hz, 2H), 7.33−7.45 (m, 10H), 7.30 (d, J = 7.4 Hz, 1H), 7.24−7.25 (m, 3H), 7.16−7.18 (m, 4H), 7.03 (br s, 3H), 6.90 (dd, J = 8.7, 1.6 Hz, 1H), 6.86 (d, J = 8.7 Hz, 2H), 6.76 (d, J = 8.6 Hz, 1H), 6.10 (s, 1H); ^{13}C NMR δ (ppm) 173.4, 165.6, 165.3, 139.3, 137.7, 136.4, 134.8, 134.7, 134.4, 132.9, 132.5, 130.2, 129.3, 129.2, 128.71, 128.66, 128.2, 128.14, 128.08, 127.3, 126.9, 126.4, 124.6, 119.4, 118.6, 118.4, 113.6, 79.0, 71.7, 71.1, 68.2; MS (TOF) 926 (M + Na^+), 942 (M + K^+). Anal. Calcd for $C_{49}H_{32}Br_2ClN_3O_3$: C 64.95, H 3.56, N 4.64. Found: C 64.81, H 3.66, N 4.63.

(*R*,*R*)- and (*S*,*S*)-2′-(3″,3″-diphenyl-1″-bromophenylazetidin-2″-one)oxy-5′-chloro-3,3-diphenyl-1-bromophenylspiro[azetidine-4,3′-indolenine]-2-one 6gc-I: mp 250−251 °C; IR (cm^{-1}) 1766, 1583, 1492; ^{1}H NMR δ (ppm) 7.70 (d, J = 8.3 Hz, 2H), 7.58 (d, J = 8.4 Hz, 2H), 7.36 (d, J = 7.8 Hz, 1H), 7.36−7.39 (m, 4H), 7.24−7.30 (m, 9H), 7.16−7.18 (m, 3H), 7.10 (t, J = 7.7 Hz, 2H), 7.04 (s, 1H), 7.02 (t, J = 7.5 Hz, 1H), 6.73 (t, J = 7.1 Hz, 2H), 6.62 (d, J = 8.8 Hz, 2H), 6.40 (d, J = 7.4 Hz, 2H), 5.88 (d, J = 1.9 Hz, 1H); ^{13}C NMR δ (ppm) 176.7, 165.9, 164.6, 149.1, 137.0, 136.8, 136.0, 135.7, 135.2, 134.2, 132.2, 132.0, 131.3, 131.0, 130.6, 128.9, 128.8, 128.6, 128.4, 128.31, 128.28, 128.2, 127.9, 127.7,

126.4, 126.3, 121.1, 120.3, 121.1, 120.3, 118.2, 118.1, 117.6, 113.2, 86.1, 75.6, 74.5, 72.3; MS (TOF) 926 ($M + Na^+$). Anal. Calcd for $C_{49}H_{32}Br_2ClN_3O_3$: C 64.95, H 3.56, N 4.64. Found: C 64.88, H 3.69, N 4.67.

(*R*,*S*)- and (*S*,*R*)-2′-(3″,3″-diphenyl-1″-bromophenylazetidin-2″-one)oxy-5′-chloro-3,3-diphenyl-1-bromophenylspiro[azetidine-4,3′-indolenine]-2-one 6gc-II: mp 204−205 °C; IR (cm^{-1}) 1768, 1618, 1594, 1491; ^{1}H NMR δ (ppm) 7.36−7.44 (m, 10H), 7.33 (t, J = 7.4 Hz, 1H), 7.22−7.28 (m, 7H), 7.15−7.18 (m, 3H), 7.11−7.12 (m, 4H), 6.95 (d, J = 8.8 Hz, 2H), 6.70 (d, J = 8.8 Hz, 2H), 6.49 (d, J = 7.5 Hz, 2H), 5.79 (d, J = 1.7 Hz, 1H); ^{13}C NMR δ (ppm) 177.5, 166.1, 165.1, 149.0, 137.4, 136.9, 136.0, 135.9, 134.4, 133.5, 132.4, 132.1, 131.6, 131.1, 130.5, 128.7, 128.6, 128.5, 128.2, 128.0, 127.6, 127.2, 126.1, 125.9, 122.6, 120.7, 119.0, 118.9, 117.9, 117.8, 85.4, 75.7, 74.8, 71.4; MS (APCI) 579 (100), 607 (80), 904 (M + 1, 1%)/906/908. Anal. Calcd for $C_{49}H_{32}Br_2ClN_3O_3$: C 64.95, H 3.56, N 4.64. Found: C 65.25, H 3.94, N 4.68.

Preparation of Spiro[azetidine-4,3′-indoline]-2,2′-dione 9bc from Hydrolysis of 5bc and 6bc. The mixture of **5bc** or **6bc** (0.5 mmol) and *p*-toluenesulfonic acid monohydrate (5 mmol) in chloroform or 1,1,2-trichloroethane (10 mL) was refluxed for a period of time (see Table 3). After concentration and column chromatography, both reactions afforded **9bc** as white solid in 93−98% yield.

Preparation of Spiro[azetidine-4,3′-indoline]-2,2′-dione 9 from 1 and 3. Under nitrogen atmosphere, a mixture of oxadiazolines **1** (2 mmol) and aryl isocyanate **3** (1 mmol) was refluxed in toluene (20 mL) for 12 h. After removal of the toluene, the residue was mixed with *p*-toluenesulfonic acid monohydrate (10 mmol) and refluxed in 1,1,2-trichloroethane (20 mL) for another 8 h. After the solvent was evaporated under vacuum, the residue was subjected to chromatography to afford products **9** in 68−75% yields.

Preparation of 1′-Diphenylacetyl-3,3,-diphenylspiro[azetidine-4,3′-indoline]-2,2′-dione 11 from 9fa. Under nitrogen atmosphere, triethylamine (1 mL) was added dropwise to a solution of **9fa** (0.2 mmol) and diphenylacetyl chloride (0.4 mmol) in chloroform (5 mL). The mixture was stirred at room temperature for 0.5 h. After removal of the triethylamine hydrochloride by filtration and evaporation of the solvent, the residue was chromatographed to afford **11** as a white solid in 60% yield.

5′-Chloro-3,3,-dimethyl-1-methylphenylspiro[azetidine-4,3′-indoline]-2,2′-dione 9bc: mp 194−195 °C; IR υ (cm^{-1}) 3262, 1747, 1621, 1516; ^{1}H NMR (300 MHz) δ (ppm) 8.41 (s, 1H, NH), 7.21 (dd, J = 8.2, 2.1 Hz, 1H), 7.19 (d, J = 2.1 Hz, 1H), 6.92 (d, J = 8.9 Hz, 2H), 6.88 (d, J = 9.0 Hz, 2H), 6.79 (d, J = 8.2 Hz, 1H), 2.16 (s, 3H), 1.44 (s, 3H), 1.27 (s, 3H); ^{13}C NMR (125 MHz) δ (ppm) 176.3, 169.9, 139.8, 134.2, 134.1, 130.4, 129.8, 128.2, 126.3, 125.2, 116.9, 112.3, 69.6, 59.9, 21.2, 20.9, 18.1; MS (TOF-EI$^+$): 340 (M^+). Anal. Calcd for $C_{19}H_{17}ClN_2O_2$: C 66.96, H 5.03, N 8.22. Found: C 66.85, H 5.23, N 8.23.

3,3,-Diphenyl-1-chlorophenylspiro[azetidine-4,3′-indoline]-2,2′-dione 9fa: mp 134−135 °C [lit.[14] mp 133 °C]; IR υ (cm^{-1}) 3267, 1760, 1731, 1619, 1493; ^{1}H NMR (300 MHz) δ 8.14 (s, 1H), 7.66 (d, J = 7.2 Hz, 2H), 7.17−7.34 (m, 13H), 6.93 (d, J = 7.8 Hz, 1H), 6.71 (t, J = 7.5 Hz, 1H), 6.19 (d, J = 7.2 Hz, 1H).

1′-Diphenylacetyl-3,3,-diphenylspiro[azetidine-4,3′-indoline]-2,2′-dione 11: mp 205−206 °C [lit.[13a] mp 208−209 °C]; IR υ (cm^{-1}) 1768, 1759, 1724, 1604, 1597, 1493; ^{1}H NMR (300 MHz) δ 8.38 (d, J = 8.1 Hz, 1H), 7.39 (d, J = 7.2 Hz, 2H), 7.02−7.37 (m, 19H), 6.95 (d, J = 9.0 Hz, 2H), 6.82 (d, J = 9.0 Hz, 2H), 6.77 (dt, J = 7.5, 0.9 Hz, 1H), 6.21 (s, 1H), 6.19 (dd, J = 7.5, 0.9 Hz, 1H).

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Supporting Information Available: The experimental procedure for preparation of **1a**−**g**, full characterization for **1b**, **1d**, **1e**, **1f**, and **1g**, ^{1}H NMR and ^{13}C NMR spectra of **4bc**, **5**, **6** (excluding **6ab-II**, **6bc-II**, and **6ac-II**), and **9bc**, as well as single-crystal data of **5db-I**, **6eb-I**, and **6gc-I** (CIF). This material is available free of charge via the Internet at http://pubs.acs.org.

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