General and Versatile Entry to 4,5-Fused Polycyclic Imidazolones Systems. Use of the Tandem Transposition/ π -Cyclization of *N*-Acyliminium Species

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Aza-heterocycles containing imidazole ring by the tandem transposition/ π -cyclization

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Experimental Section

General remarks. Melting points were taken with a capillary melting point apparatus and are uncorrected. The infrared (IR) absorption spectra were determined as solutions in potassium bromide and are indicated in cm^{-1} . The ^1H and ^{13}C NMR spectra were recorded as solutions in CDCl_3 or $\text{DMSO}-d_6$ at 200 or 300 MHz (^1H) and 50.3 or 75 MHz (^{13}C), respectively, and chemical shifts (δ) are expressed in ppm relative to TMS as internal standard. Thin layer chromatography (TLC) was performed using of silica gel analytical plates (F_{254}) of 0.25 mm thickness. The detection on TLC plates was performed by UV light at 254 or 365 nm or using iodine vapor. Mass spectra (MS) were measured on a mass spectrometer using electron impact ionization (EI, 70 eV). Optical rotations were determined at 25 °C in

acetone. The analytical results of elemental analysis are within 0.4 % of theoretical values and were obtained from INSA institution at Rouen, F-76130 Mt-St-Aignan in France.

General Procedure for the Synthesis of Spirohydantoins 9Ia-f and 9IIg,h. To a stirred solution of 0.04 moles of ketone **8** in 100 mL of EtOH/H₂O (1/1) was added potassium cyanide (3.9 g, 0.06 moles) and ammonium carbonate (38.4 g, 0.4 moles). The mixture was heated at 55-60 °C until the starting ketone was disappeared (monitored by TLC using a mixture of cyclohexane/AcOEt (3/2) as eluent; the reaction was complete after 2 h to 3 weeks). The reaction solution was cooled, the contents removed, filtered and diluted with 60 mL of water. The reaction solution was then acidified with 4M HCl solution until pH=6. The resulting precipitate was filtered of, washed twice with cold water, air dried and recrystallized from dry ethanol to give the expected spirohydantoin **9** in variable yields.

Spiro(hydantoin-5,1'-indane) (9Ia). This product **9Ia** was obtained from 1-indanone (**8Ia**) as yellow solids in 82% yield (Lit.¹ yield=76%); R_f (cyclohexane/AcOEt: 3/2)=0.25; mp 239 °C (Lit.¹ mp 238-240 °C); IR (KBr): ν 3280 (2x NH), 1761 (C=O), 1715 (C=O) cm⁻¹; ¹H NMR (DMSO-d₆): δ 2.07-2.22 (m, 1H, CH₂), 2.46-2.59 (m, 1H, CH₂), 2.96-3.03 (m, 2H, CH₂), 7.13-7.33 (m, 4H, H_{aro}), 8.43 (s broad, 1H, NH), 10.74 (s broad, 1H, NH); ¹³C NMR (DMSO-d₆): δ 29.8 (CH₂), 36.4 (CH₂), 71.9 (C^q), 122.9 (CH_{aro}), 125.2 (CH_{aro}), 127.1 (CH_{aro}), 129.0 (CH_{aro}), 141.4 (C^q_{aro}-C), 144.0 (C^q_{aro}-C), 156.7 (C₂=O), 177.4 (C₄=O); Anal. Calcd. for C₁₁H₁₀N₂O₂ (202.07): C, 65.34; H, 4.98; N, 13.85. Found: C, 65.20; H, 4.79; N, 13.69.

Spiro(hydantoin-5,2'-indane) (9Ib). Starting from the 2-indanone (**8Ib**), this product **9Ib** was isolated as yellow solids in 63% yield (Lit.¹ 63% yield) after crystallization from anhydrous ethanol; mp 163-166 °C; R_f (cyclohexane/AcOEt: 2/3)=0.35; mp 260 °C (Lit.¹ mp 263-265 °C); IR (KBr): ν 3193 (2x NH), 1770 (C=O), 1730 (C=O) cm⁻¹; ¹H NMR (DMSO-d₆): δ 2.64 (d, 2H, *J*=16 Hz, CH₂), 2.93 (d, 2H, *J*=16 Hz, CH₂), 6.69-6.79 (m, 4H, H_{aro}), 7.94 (s broad, 1H, NH), 9.60 (s broad, 1H, NH); ¹³C NMR (DMSO-

¹ In general, our yields are better than those reported in the literature except for the spirohydantoin **9Ic**. For that, see: Sarges, R.; Schnur, R. C.; Belletire, J. L.; Peterson, M. J. *J. Med. Chem.* **1988**, *31*, 230-243.

d₆): δ 43.5 (2x CH₂), 67.8 (C^q), 122.3 (2x CH_{aro}), 126.9 (2x CH_{aro}), 139.8 (2x C^q_{aro}-C), 156.5 (C₂=O), 178.6 (C₄=O); Anal. Calcd. for C₁₁H₁₀N₂O₂ (202.07): C, 65.34; H, 4.98; N, 13.85. Found: C, 65.24; H, 4.80; N, 13.77.

Spiro(hydantoin-5,1'-tetraline) (9Ic). Starting from the α -tetralone (**8Ic**), this product **9Ic** was isolated as yellow solids in 76% yield (Lit.¹ 84% yield) after crystallization from anhydrous ethanol; Rf (cyclohexane/AcOEt: 3/2)=0.6; mp 242 °C (Lit.¹ mp 240-242 °C); IR (KBr): ν 3215 (2x NH), 1768 (C=O), 1698 (C=O) cm⁻¹; ¹H NMR (DMSO-d₆): δ 1.71-2.07 (m, 4H, 2x CH₂), 2.67-2.72 (m, 2H, CH₂), 6.98-7.21 (m, 4H, H_{aro}), 8.49 (s broad, 1H, NH), 10.05 (s broad, 1H, NH); ¹³C NMR (DMSO-d₆): δ 18.5 (CH₂), 28.5 (CH₂), 33.6 (CH₂), 63.1 (C^q), 126.6 (CH_{aro}), 126.7 (CH_{aro}), 128.0 (CH_{aro}), 129.3 (CH_{aro}), 134.4 (C^q_{aro}-C), 137.8 (C^q_{aro}-C), 156.5 (C₂=O), 178.0 (C₄=O); Anal. Calcd. for C₁₂H₁₂N₂O₂ (216.09): C, 66.65; H, 5.59; N, 12.96. Found: C, 66.39; H, 5.42; N, 12.78.

Spiro(hydantoin-5,2'-tetraline) (9Id). Starting from the β -tetralone (**8Id**), this product **9Id** was isolated as white solids in 97% yield (Lit.² 79% yield) after crystallization from anhydrous ethanol; Rf (cyclohexane/AcOEt: 3/2)=0.36; mp 267 °C (Lit.² mp 268 °C); IR (KBr): ν 3242 (2x NH), 1784 (C=O), 1720 (C=O) cm⁻¹; ¹H NMR (DMSO-d₆): δ 1.83-1.91 (m, 2H, CH₂), 2.72-2.90 (m, 5H, 2x CH₂ + NH), 6.99-7.25 (m, 4H, H_{aro}), 8.31 (s broad, 1H, NH); ¹³C NMR (DMSO-d₆): δ 24.8 (CH₂), 30.1 (CH₂), 36.9 (CH₂), 60.8 (C^q), 125.9 (CH_{aro}), 126.0 (CH_{aro}), 128.7 (CH_{aro}), 129.1 (CH_{aro}), 132.7 (C^q_{aro}-C), 134.9 (C^q_{aro}-C), 156.7 (C₂=O), 178.5 (C₄=O); Anal. Calcd. for C₁₂H₁₂N₂O₂ (216.09): C, 66.65; H, 5.59; N, 12.96. Found: C, 66.44; H, 5.47; N, 12.81.

Spiro(hydantoin-5,4'-thiochromane) (9Ie). Starting from the thiochroman-4-one (**8Ie**), this product **9Ie** was isolated as white-yellow solids in 98% yield (Lit.¹ 53% yield) after crystallization from anhydrous ethanol; Rf (cyclohexane/AcOEt: 2/3)=0.54; mp 227 °C (Lit.¹ mp 225-227 °C); IR (KBr): ν 3331 (2x NH), 1772 (C=O), 1703 (C=O) cm⁻¹; ¹H NMR (DMSO-d₆): δ 2.27-2.36 (m, 2H, CH₂), 3.01-3.09 (m, 1H, CH₂), 3.28-3.35 (m, 1H, CH₂), 7.14-7.23 (m, 4H, H_{aro}), 8.67 (s broad, 1H, NH), 10.47 (s

broad, 1H, NH); ^{13}C NMR (DMSO- d_6): δ 21.7 (CH_2), 32.9 (CH_2), 61.9 (C^q), 124.9 (CH_{aro}), 126.9 (CH_{aro}), 128.1 (CH_{aro}), 128.7 (CH_{aro}), 130.9 ($\text{C}^q_{\text{aro-S}}$), 133.9 ($\text{C}^q_{\text{aro-C}}$), 156.4 ($\text{C}_2=\text{O}$), 177.3 ($\text{C}_4=\text{O}$); Anal. Calcd. for $\text{C}_{11}\text{H}_{10}\text{N}_2\text{O}_2\text{S}$ (234.05): C, 56.39; H, 4.30; N, 11.96. Found: C, 56.18; H, 4.21; N, 11.86.

5'-H-6',7'-Dihydro-1'-methylspiro(hydantoin-5,4'-indole) (9If). Starting from the 5,6-dihydro-1-methylindol-4(7H)-one (**8If**),³ this product **9If** was isolated as white solids in 21% yield after crystallization from ethanol/water; Rf (cyclohexane/AcOEt: 2/3)=0.55; mp 278 °C; IR (KBr): ν 3270 (2x NH), 1765 ($\text{C}=\text{O}$), 1706 ($\text{C}=\text{O}$) cm^{-1} ; ^1H NMR (DMSO- d_6): δ 1.72-1.99 (m, 6H, 3x CH_2), 3.44 (s, 3H, CH_3), 5.68 (d, 1H, $J=2$ Hz, $\text{H}_{\text{pyrrole}}$), 6.58 (d, 1H, $J=2$ Hz, $\text{H}_{\text{pyrrole}}$), 8.16 (s broad, 1H, NH), 10.47 (s broad, 1H, NH); ^{13}C NMR (DMSO- d_6): δ 19.1 (CH_2), 20.3 (CH_2), 32.7 (CH_3), 33.8 (CH_2), 61.3 (C^q), 103.1 ($\text{CH}_{\text{pyrrole}}$), 115.2 ($\text{C}^q_{\text{pyrrole}}$), 121.0 ($\text{CH}_{\text{pyrrole}}$), 130.1 ($\text{C}^q_{\text{pyrrole}}$), 156.2 ($\text{C}_2=\text{O}$), 178.4 ($\text{C}_4=\text{O}$); Anal. Calcd. for $\text{C}_{11}\text{H}_{13}\text{N}_3\text{O}_2$ (219.10): C, 60.26; H, 5.98; N, 19.17. Found: C, 60.18; H, 5.83; N, 19.06.

Spiro(fluorene-9,5'-hydantoin) (9Ilg). Starting from the fluorenone (**8Ilg**), this product **9Ilg** was isolated as a yellow-white crystal in 88% yield after crystallization from ethanol/water; Rf (cyclohexane/AcOEt: 1/4)=0.70; mp > 300 °C (Lit.⁴ mp 324-325 °C); IR (KBr): ν 3242 (2x NH), 1776 ($\text{C}=\text{O}$), 1695 ($\text{C}=\text{O}$) cm^{-1} ; ^1H NMR (DMSO- d_6): δ 7.32-7.52 (m, 6H, H_{aro}), 7.88-7.91 (m, 2H, H_{aro}), 8.60 (s broad, 1H, NH), 10.88 (s broad, 1H, NH); ^{13}C NMR (DMSO- d_6): δ 72.4 (C^q), 120.8 (2x CH_{aro}), 123.6 (2x CH_{aro}), 128.4 (2x CH_{aro}), 129.9 (2x CH_{aro}), 140.7 (2x $\text{C}^q_{\text{aro-C}}$), 142.9 (2x $\text{C}^q_{\text{aro-C}}$), 157.7 ($\text{C}_2=\text{O}$), 174.2 ($\text{C}_4=\text{O}$); Anal. Calcd. for $\text{C}_{15}\text{H}_{10}\text{N}_2\text{O}_2$ (250.07): C, 71.99; H, 4.03; N, 11.19. Found: C, 71.78; H, 3.92; N, 11.10.

Spiro(hydantoin-5,3'-pyrrolo[1,2-a]indole) (9IIh). Starting from the pyrrolo[1,2-a]indol-4-one (**8IIh**),⁵ this product **9IIh** was isolated as an orange-yellow crystal in 12% yield after crystallization from ethanol/water; Rf (cyclohexane/AcOEt: 2/3)=0.45; mp > 300 °C; IR (KBr): ν 3338 (NH), 3216 (NH), 1778 ($\text{C}=\text{O}$), 1724 ($\text{C}=\text{O}$) cm^{-1} ; ^1H NMR (DMSO- d_6): δ 7.32-7.52 (m, 6H, H_{aro}), 7.88-7.91 (m, 2H, H_{aro}),

² Goodson, L. H.; Honigberg, I. L.; Lehman, J. J.; Burton, W. H. *J. Org. Chem.* **1960**, 25, 1920-1924.

³ Santaniello, E.; Farachi, C.; Ponti, F. *Synthesis* **1979**, 617-618.

⁴ Henze, H. H.; Speer, R. J. *J. Am. Chem. Soc.* **1942**, 64, 522-523.

8.60 (s broad, 1H, NH), 10.88 (s broad, 1H, NH); ^{13}C NMR (DMSO- d_6): δ 72.4 (C^q), 120.8 (2x CH_{aro}), 123.6 (2x CH_{aro}), 128.4 (2x CH_{aro}), 129.9 (2x CH_{aro}), 140.7 (2x $\text{C}^q_{\text{aro-C}}$), 142.9 (2x $\text{C}^q_{\text{aro-C}}$), 157.7 ($\text{C}_2=\text{O}$), 174.2 ($\text{C}_4=\text{O}$); Anal. Calcd. for $\text{C}_{13}\text{H}_9\text{N}_3\text{O}_2$ (239.07): C, 65.27; H, 3.79; N, 17.56. Found: C, 65.16; H, 3.69; N, 17.50.

General Procedure for the Synthesis of N_1,N_3 -Dibenzylspirohydantoins 10a-h and N_3 -Benzylspirohydantoins 11a-g. To a stirred mixture of spirohydantoins (**9a-h**, 10 mmoles) and 18-C-6 ether-crown (1% w/w) in 20 mL of dry toluene were added solid potassium carbonate (1.66 g, 12 mmoles) and 0.1 equivalent per mmole of potassium iodide. After stirring for 15 minutes, 24 mmoles of benzyl bromide (1.43 mL) in 30 mL of dry toluene was added dropwise over a period of 30 minutes. The mixture was then refluxed for 24 hours under stirring and cooled. After filtration of over a short column of celite, the organic layer was concentrated under reduced pressure and the crude resulting solid was purified by flash chromatography on silica gel column with dichloromethane as eluent to give a white oil. N_1,N_3 -dibenzylated spirohydantoins **10a-h** and N_3 -benzylated spirohydantoins **11a-g** were obtained in pure forms after chromatography separation on silica gel column using a mixture of cyclohexane/AcOEt (4/1) as eluent.

1,3-Dibenzylspiro(imidazolidine-5,1'-indane)-2,4-dione (10a). This product as a minor isomer was isolated as white-yellow solids from hydantoin **9Ia** in 16% yield and melted at 97 °C (ethanol); Rf (cyclohexane/AcOEt: 3/2)=0.74; IR (KBr): ν 1766 ($\text{C}=\text{O}$), 1709 ($\text{C}=\text{O}$) cm^{-1} ; ^1H NMR (CDCl_3): δ 1.94-2.08 (m, 1H, CH_2), 2.39-2.53 (m, 1H, CH_2), 2.74-2.89 (m, 1H, CH_2), 3.01-3.17 (m, 1H, CH_2), 3.77 (d, 1H, $J=16$ Hz, CH_2), 4.77 (s, 2H, CH_2), 4.80 (d, 1H, $J=16$ Hz, CH_2), 6.77 (dd, 1H, $J=1.6$ and 8.6 Hz, H_{aro}), 7.06-7.29 (m, 13H, H_{aro}); ^{13}C NMR (CDCl_3): δ 30.5 (CH_2), 33.8 (CH_2), 42.7 (CH_2), 43.9 (CH_2), 75.5 (C^q), 123.2 (CH_{aro}), 125.5 (CH_{aro}), 127.5 (CH_{aro}), 127.6 (CH_{aro}), 127.7 (2x CH_{aro}), 127.9 (CH_{aro}), 128.4 (2x CH_{aro}), 128.6 (2x CH_{aro}), 128.8 (2x CH_{aro}), 129.8 (CH_{aro}), 136.4 ($\text{C}^q_{\text{aro-C}}$), 137.5 ($\text{C}^q_{\text{aro-C}}$), 138.0

⁵ Mazzola, V. J.; Bernady, K. F.; Franck, R. W. *J. Org. Chem.* **1967**, 32, 486-489.

(C^q_{aro}-C), 145.1 (C^q_{aro}-C), 156.3 (C₂=O), 175.0 (C₄=O); Anal. Calcd. for C₂₅H₂₂N₂O₂ (382.17): C, 78.51; H, 5.80; N, 7.32. Found: C, 78.43; H, 5.66; N, 7.20.

3-Benzylspiro(imidazolidine-5,1'-indane)-2,4-dione (11a). This product as a major isomer was isolated as white solids from hydantoin **9Ia** in 63% yield and melted at 145 °C (ethanol); R_f (cyclohexane/AcOEt: 3/2)=0.57; IR (KBr): ν 3297 (NH), 1772 (C=O), 1711 (C=O) cm⁻¹; ¹H NMR (CDCl₃): δ 2.13-2.28 (m, 1H, CH₂), 2.61-2.74 (m, 1H, CH₂), 2.94-3.08 (m, 1H, CH₂), 3.15-3.31 (m, 1H, CH₂), 4.62 (d, 1H, *J*=16 Hz, CH₂), 4.70 (d, 1H, *J*=16 Hz, CH₂), 5.93 (s broad, 1H, NH), 6.98 (dd, 1H, *J*=1.6 and 8.6 Hz, H_{aro}), 7.14-7.33 (m, 8H, H_{aro}); ¹³C NMR (CDCl₃): δ 30.2 (CH₂), 36.9 (CH₂), 42.2 (CH₂), 71.4 (C^q), 122.7 (CH_{aro}), 125.5 (CH_{aro}), 127.4 (CH_{aro}), 127.9 (CH_{aro}), 128.2 (2x CH_{aro}), 128.7 (2x CH_{aro}), 129.6 (CH_{aro}), 136.2 (C^q_{aro}-C), 140.0 (C^q_{aro}-C), 144.2 (C^q_{aro}-C), 156.9 (C₂=O), 175.4 (C₄=O); Anal. Calcd. for C₁₈H₁₆N₂O₂ (292.12): C, 73.95; H, 5.52; N, 9.58. Found: C, 73.87; H, 5.44; N, 9.41.

1,3-Dibenzylspiro(imidazolidine-5,2'-indane)-2,4-dione (10b). This product as a major isomer was isolated as white solids from hydantoin **9Ib** in 35% yield and melted at 100 °C (ethanol); R_f (cyclohexane/AcOEt: 3/2)=0.70; IR (KBr): ν 1767 (C=O), 1713 (C=O) cm⁻¹; ¹H NMR (CDCl₃): δ 2.88 (d, 2H, *J*=16 Hz, CH₂), 3.53 (d, 2H, *J*=16 Hz, CH₂), 4.23 (s, 2H, CH₂), 4.75 (s, 2H, CH₂), 6.77 (dd, 2H, *J*=2 and 8 Hz, H_{aro}), 6.99-7.45 (m, 12H, H_{aro}); ¹³C NMR (CDCl₃): δ 42.7 (2x CH₂), 42.9 (CH₂), 44.1 (CH₂), 71.4 (C^q), 124.3 (2x CH_{aro}), 127.6 (2x CH_{aro}), 127.7 (CH_{aro}), 128.0 (CH_{aro}), 128.1 (2x CH_{aro}), 128.3 (2x CH_{aro}), 128.6 (2x CH_{aro}), 128.9 (2x CH_{aro}), 136.3 (C^q_{aro}-C), 137.6 (C^q_{aro}-C), 139.3 (2x C^q_{aro}-C), 155.7 (C₂=O), 175.5 (C₄=O); Anal. Calcd. for C₂₅H₂₂N₂O₂ (382.17): C, 78.51; H, 5.80; N, 7.32. Found: C, 78.39; H, 5.69; N, 7.18.

3-Benzylspiro(imidazolidine-5,2'-indane)-2,4-dione (11b). This product as a major isomer was isolated as white solids from hydantoin **9Ib** in 29% yield and melted at 189 °C (ethanol); R_f (cyclohexane/AcOEt: 3/2)=0.57; IR (KBr): ν 3280 (NH), 1778 (C=O), 1719 (C=O) cm⁻¹; ¹H NMR (CDCl₃): δ 3.03 (d, 2H, *J*=16 Hz, CH₂), 3.61 (d, 2H, *J*=16 Hz, CH₂), 4.64 (s, 2H, CH₂), 5.57 (s broad, 1H, NH), 7.16-7.36 (m, 9H, H_{aro}); ¹³C NMR (CDCl₃): δ 42.4 (CH₂), 44.2 (2x CH₂), 68.5 (C^q), 124.7 (2x

CH_{aro}), 127.5 (2x CH_{aro}), 127.9 (CH_{aro}), 128.5 (2x CH_{aro}), 128.8 (2x CH_{aro}), 136.1 (C^q_{aro}-C), 139.0 (2x C^q_{aro}-C), 156.2 (C₂=O), 175.8 (C₄=O); Anal. Calcd. for C₁₈H₁₆N₂O₂ (292.12): C, 73.95; H, 5.52; N, 9.58. Found: C, 73.79; H, 5.39; N, 9.42.

1,3-Dibenzylspiro(imidazolidine-5,1'-tetraline)-2,4-dione (10c). This product as a minor isomer was isolated as white solids from hydantoin **9Ic** in 21% yield and melted at 117 °C (ethanol); R_f (cyclohexane/AcOEt: 4/1)=0.62; IR (KBr): ν 1765 (C=O), 1709 (C=O) cm⁻¹; ¹H NMR (CDCl₃): δ 1.65-1.84 (m, 2H, CH₂), 1.91-2.03 (m, 1H, CH₂), 2.16-2.41 (m, 1H, CH₂), 2.61-2.85 (m, 2H, CH₂), 3.72 (d, 1H, *J*=15 Hz, CH₂), 4.69 (d, 1H, *J*=15 Hz, CH₂), 4.79 (d, 1H, *J*=15 Hz, CH₂), 4.85 (d, 1H, *J*=15 Hz, CH₂), 6.73 (dd, 1H, *J*=1.6 and 8.6 Hz, H_{aro}), 7.00-7.09 (m, 1H, H_{aro}), 7.13-7.34 (m, 12H, H_{aro}); ¹³C NMR (CDCl₃): δ (ppm): 19.2 (CH₂), 29.0 (CH₂), 32.2 (CH₂), 42.7 (CH₂), 44.3 (CH₂), 66.7 (C^q), 125.9 (CH_{aro}), 126.9 (CH_{aro}), 127.5 (CH_{aro}), 127.7 (2x CH_{aro}), 127.9 (CH_{aro}), 128.5 (2x CH_{aro}), 128.7 (2x CH_{aro}), 128.8 (CH_{aro}), 128.9 (2x CH_{aro}), 130.3 (CH_{aro}), 130.8 (C^q_{aro}-C), 136.4 (C^q_{aro}-C), 137.8 (C^q_{aro}-C), 139.9 (C^q_{aro}-C), 156.9 (C₂=O), 175.3 (C₄=O); Anal. Calcd. for C₂₆H₂₄N₂O₂ (396.18): C, 78.76; H, 6.10; N, 7.07. Found: C, 78.67; H, 6.01; N, 7.00.

3-Benzylspiro(imidazolidine-5,1'-tetraline)-2,4-dione (11c). This product as a major isomer was isolated as white solids from hydantoin **9Ic** in 57% yield and melted at 144 °C (ethanol); R_f (cyclohexane/AcOEt: 4/1)=0.34; IR (KBr): ν 3253 (NH), 1774 (C=O), 1715 (C=O) cm⁻¹; ¹H NMR (CDCl₃): δ 1.67-2.02 (m, 2H, CH₂), 2.17-2.47 (m, 2H, CH₂), 2.71-2.92 (m, 2H, CH₂), 4.67 (d, 1H, *J*=15 Hz, CH₂), 4.74 (d, 1H, *J*=15 Hz, CH₂), 5.92 (s broad, 1H, NH), 6.88 (dd, 1H, *J*=1.6 and 8.6 Hz, H_{aro}), 7.04-7.41 (m, 8H, H_{aro}); ¹³C NMR (CDCl₃): δ 19.1 (CH₂), 28.8 (CH₂), 34.0 (CH₂), 42.3 (CH₂), 62.9 (C^q), 126.6 (CH_{aro}), 126.9 (CH_{aro}), 127.9 (CH_{aro}), 128.4 (2x CH_{aro}), 128.7 (CH_{aro}), 128.8 (2x CH_{aro}), 129.8 (CH_{aro}), 133.1 (C^q_{aro}-C), 136.2 (C^q_{aro}-C), 138.1 (C^q_{aro}-C), 156.7 (C₂=O), 176.0 (C₄=O); Anal. Calcd. for C₁₉H₁₈N₂O₂ (306.14): C, 74.49; H, 5.92; N, 9.14. Found: C, 74.35; H, 5.79; N, 9.06.

1,3-Dibenzylspiro(imidazolidine-5,2'-tetraline)-2,4-dione (10d). This product as a minor isomer was isolated as white solids from hydantoin **9Id** in 26% yield and melted at 131 °C (diethyl ether); R_f (cyclohexane/AcOEt: 4/1)=0.59; IR (KBr): ν 1775 (C=O), 1716 (C=O) cm⁻¹; ¹H NMR (CDCl₃): δ 1.84-

2.06 (m, 2H, CH₂), 2.61-3.11 (m, 4H, 2x CH₂), 4.33 (d, 1H, *J*=16 Hz, CH₂), 4.48 (d, 1H, *J*=16 Hz, CH₂), 4.52 (s, 2H, CH₂), 6.88 (dd, 1H, *J*=2 and 8.6 Hz, H_{aro}), 6.99-7.33 (m, 13H, H_{aro}); ¹³C NMR (CDCl₃): δ (ppm): 26.0 (CH₂), 30.2 (CH₂), 35.6 (CH₂), 42.7 (CH₂), 43.9 (CH₂), 63.6 (C^q), 126.5 (CH_{aro}), 126.7 (CH_{aro}), 127.4 (2x CH_{aro}), 127.6 (CH_{aro}), 127.9 (CH_{aro}), 128.3 (CH_{aro}), 128.5 (2x CH_{aro}), 128.6 (2x CH_{aro}), 128.7 (CH_{aro}), 128.8 (2x CH_{aro}), 132.6 (C^q_{aro}-C), 135.6 (C^q_{aro}-C), 136.4 (C^q_{aro}-C), 137.9 (C^q_{aro}-C), 156.3 (C₂=O), 175.7 (C₄=O); Anal. Calcd. for C₂₆H₂₄N₂O₂ (396.18): C, 78.76; H, 6.10; N, 7.07. Found: C, 78.67; H, 6.02; N, 6.95.

3-Benzylspiro(imidazolidine-5,2'-tetraline)-2,4-dione (11d). This produced as a major isomer was isolated as white solids from hydantoin **9Id** in 37% yield and melted at 169 °C (ethanol/water); R_f (cyclohexane/AcOEt: 4/1)=0.41; IR (KBr): ν 3275 (NH), 1775 (C=O), 1716 (C=O) cm⁻¹; ¹H NMR (CDCl₃): δ 1.78-1.90 (m, 1H, CH₂), 2.13-2.29 (m, 1H, CH₂), 2.7 (d, 1H, *J*=16 Hz, CH₂), 2.86-3.08 (m, 2H, CH₂), 3.38 (d, 1H, *J*=16 Hz, CH₂), 4.65 (s, 2H, CH₂), 5.71 (s broad, 1H, NH), 7.02-7.36 (m, 9H, H_{aro}); ¹³C NMR (CDCl₃): δ 25.0 (CH₂), 30.1 (CH₂), 37.4 (CH₂), 42.3 (CH₂), 60.8 (C^q), 126.6 (CH_{aro}), 126.9 (CH_{aro}), 127.9 (CH_{aro}), 128.6 (2x CH_{aro}), 128.8 (2x CH_{aro}), 129.0 (CH_{aro}), 129.6 (CH_{aro}), 131.8 (C^q_{aro}-C), 134.0 (C^q_{aro}-C), 136.2 (C^q_{aro}-C), 156.5 (C₂=O), 176.2 (C₄=O); Anal. Calcd. for C₁₉H₁₈N₂O₂ (306.14): C, 74.49; H, 5.92; N, 9.14. Found: C, 74.32; H, 5.81; N, 9.08.

1,3-Dibenzylspiro(imidazolidine-5,4'-thiochromane)-2,4-dione (10e). This product as a major isomer was isolated as white solids from hydantoin **9Ie** in 36% yield and melted at 138 °C (ethanol); R_f (cyclohexane/AcOEt: 3/2)=0.78; IR (KBr): ν 1767 (C=O), 1711 (C=O) cm⁻¹; ¹H NMR (CDCl₃): δ 2.11-2.15 (m, 2H, CH₂), 2.59-2.70 (m, 1H, CH₂), 3.70 (d, 1H, *J*=16 Hz, CH₂), 3.74-3.85 (m, 1H, CH₂), 4.68 (d, 1H, *J*=15 Hz, CH₂), 4.80 (d, 1H, *J*=15 Hz, CH₂), 4.88 (d, 1H, *J*=16 Hz, CH₂), 6.75 (dd, 1H, *J*=1.6 and 8.6 Hz, H_{aro}), 6.88-7.09 (m, 1H, H_{aro}), 7.13-7.36 (m, 12H, H_{aro}); ¹³C NMR (CDCl₃): δ 22.4 (CH₂), 31.7 (CH₂), 42.7 (CH₂), 44.2 (CH₂), 65.3 (C^q), 124.9 (CH_{aro}), 126.7 (CH_{aro}), 126.9 (C^q_{aro}-S), 127.6 (CH_{aro}), 127.8 (2x CH_{aro}), 128.0 (CH_{aro}), 128.1 (CH_{aro}), 128.4 (2x CH_{aro}), 128.7 (2x CH_{aro}), 128.8 (2x CH_{aro}), 128.9 (CH_{aro}), 136.1 (C^q_{aro}-C), 136.4 (C^q_{aro}-C), 137.4 (C^q_{aro}-C), 156.6 (C₂=O), 174.1 (C₄=O); Anal. Calcd. for C₂₅H₂₂N₂O₂S (414.14): C, 72.44; H, 5.35; N, 6.76. Found: C, 72.22; H, 5.12; N, 6.54.

3-Benzylspiro(imidazolidine-5,4'-thiochromane)-2,4-dione (11e). This product as a minor isomer was isolated as yellow solids from hydantoin **9Ie** in 25% yield and melted at 130 °C (ethanol); Rf (cyclohexane/AcOEt: 3/2)=0.62; IR (KBr): ν 3258 (NH), 1777 (C=O), 1717 (C=O) cm^{-1} ; ^1H NMR (CDCl_3): δ 2.19-2.49 (m, 2H, CH_2), 2.89-3.02 (m, 1H, CH_2), 3.37-3.49 (m, 1H, CH_2), 4.60 (d, 1H, $J=15$ Hz, CH_2), 4.72 (d, 1H, $J=15$ Hz, CH_2), 6.68 (s broad, 1H, NH), 6.89-6.98 (m, 2H, H_{aro}), 7.09-7.15 (m, 2H, H_{aro}), 7.27-7.34 (m, 5H, H_{aro}); ^{13}C NMR (CDCl_3): δ 22.3 (CH_2), 33.1 (CH_2), 42.5 (CH_2), 61.7 (C^{q}), 125.1 (CH_{aro}), 127.5 (CH_{aro}), 127.6 (CH_{aro}), 128.1 (CH_{aro}), 128.5 (2x CH_{aro}), 128.9 (2x CH_{aro}), 129.1 (CH_{aro}), 129.5 ($\text{C}^{\text{q}}_{\text{aro-S}}$), 134.5 ($\text{C}^{\text{q}}_{\text{aro-C}}$), 136.0 ($\text{C}^{\text{q}}_{\text{aro-C}}$), 156.4 ($\text{C}_2=\text{O}$), 174.9 ($\text{C}_4=\text{O}$); Anal. Calcd. for $\text{C}_{18}\text{H}_{16}\text{N}_2\text{O}_2\text{S}$ (324.09): C, 66.64; H, 4.97; N, 8.64. Found: C, 66.56; H, 4.83; N, 8.51.

5'H-1,3-Dibenzyl-6',7'-dihydro-1'-methylspiro(imidazolidine-5,4'-indole)-2,4-dione (10f). This product as a minor isomer was isolated as white solids from hydantoin **9If** in 26% yield and melted at 50 °C (diethyl ether/cyclohexane); Rf (cyclohexane/AcOEt: 3/2)=0.59; IR (KBr): ν 1762 (C=O), 1704 (C=O) cm^{-1} ; ^1H NMR (CDCl_3): δ 1.60-1.76 (m, 2H, CH_2), 1.88-1.96 (m, 1H, CH_2), 2.21-2.36 (m, 1H, CH_2), 2.41-2.61 (m, 2H, CH_2), 3.47 (s, 3H, CH_3), 3.81 (d, 1H, $J=16$ Hz, CH_2), 4.69 (d, 1H, $J=14$ Hz, CH_2), 4.78 (d, 1H, $J=14$ Hz, CH_2), 4.85 (d, 1H, $J=16$ Hz, CH_2), 5.60 (d, 1H, $J=2$ Hz, $\text{H}_{\text{pyrrole}}$), 6.48 (d, 1H, $J=2$ Hz, $\text{H}_{\text{pyrrole}}$), 7.18-7.36 (m, 10H, H_{aro}); ^{13}C NMR (CDCl_3): δ 19.7 (CH_2), 20.5 (CH_2), 31.9 (CH_2), 33.3 (CH_3), 42.5 (CH_2), 43.7 (CH_2), 65.2 (C^{q}), 103.3 ($\text{CH}_{\text{pyrrole}}$), 112.1 ($\text{C}^{\text{q}}_{\text{pyrrole}}$), 121.8 ($\text{CH}_{\text{pyrrole}}$), 127.3 (2x CH_{aro}), 128.3 (4x CH_{aro}), 128.5 (2x CH_{aro}), 128.7 (2x CH_{aro}), 132.6 ($\text{C}^{\text{q}}_{\text{pyrrole}}$), 136.7 ($\text{C}^{\text{q}}_{\text{aro-C}}$), 138.5 ($\text{C}^{\text{q}}_{\text{aro-C}}$), 156.7 ($\text{C}_2=\text{O}$), 176.0 ($\text{C}_4=\text{O}$); Anal. Calcd. for $\text{C}_{25}\text{H}_{25}\text{N}_3\text{O}_2$ (399.19): C, 75.16; H, 6.31; N, 10.52. Found: C, 75.06; H, 6.17; N, 10.40.

5'H-3-Benzyl-6',7'-dihydro-1'-methylspiro(imidazolidine-5,4'-indole)-2,4-dione (11f). This produced as a major isomer was isolated as yellow solids from hydantoin **9If** in 48% yield and melted at 177 °C (ethanol); Rf (cyclohexane/AcOEt: 3/2)=0.35; IR (KBr): ν 3340 (NH), 1770 (C=O), 1710 (C=O) cm^{-1} ; ^1H NMR (CDCl_3): δ 1.74-1.92 (m, 2H, CH_2), 2.11-2.38 (m, 2H, CH_2), 2.52-2.58 (m, 2H, CH_2), 3.44 (s, 3H, CH_3), 4.65 (d, 1H, $J=15$ Hz, CH_2), 4.73 (d, 1H, $J=15$ Hz, CH_2), 5.43 (s broad, 1H, NH), 5.71 (d,

1H, $J=2$ Hz, H_{pyrrole}), 6.46 (d, 1H, $J=2$ Hz, H_{pyrrole}), 7.27-7.40 (m, 5H, H_{aro}); ^{13}C NMR (CDCl_3): δ 19.6 (CH_2), 20.7 (CH_2), 33.1 (CH_3), 33.9 (CH_2), 42.0 (CH_2), 60.9 (C^q), 103.3 ($\text{CH}_{\text{pyrrole}}$), 114.3 ($\text{C}^q_{\text{pyrrole}}$), 121.6 ($\text{CH}_{\text{pyrrole}}$), 127.7 (CH_{aro}), 128.1 (2x CH_{aro}), 128.6 (2x CH_{aro}), 131.0 ($\text{C}^q_{\text{pyrrole}}$), 136.5 ($\text{C}^q_{\text{aro-C}}$), 156.5 ($\text{C}_2=\text{O}$), 176.5 ($\text{C}_4=\text{O}$); Anal. Calcd. for $\text{C}_{18}\text{H}_{19}\text{N}_3\text{O}_2$ (309.15): C, 69.88; H, 6.19; N, 13.58. Found: C, 69.67; H, 6.10; N, 13.44.

1,3-Dibenzylspiro(fluorene-9,5'-imidazolidine)-2,4-dione (10g). This product as a minor isomer was isolated as white solid from hydantoin **9IIg** in 30% yield and melted at 136 °C (ethanol/diethyl ether); Rf (cyclohexane/AcOEt: 3/2)=0.79; IR (KBr): ν 1769 ($\text{C}=\text{O}$), 1781 ($\text{C}=\text{O}$), 1715 ($\text{C}=\text{O}$) cm^{-1} ; ^1H NMR (CDCl_3): δ 4.11 (s, 2H, CH_2), 4.82 (s, 2H, CH_2), 6.72-6.75 (m, 2H, H_{aro}), 6.85-7.11 (m, 7H, H_{aro}), 7.32-7.47 (m, 7H, H_{aro}), 7.65-7.68 (m, 2H, H_{aro}); ^{13}C NMR (CDCl_3): δ 43.2 (CH_2), 44.8 (CH_2), 74.9 (C^q), 120.7 (2x CH_{aro}), 123.9 (2x CH_{aro}), 127.4 (CH_{aro}), 127.9 (2x CH_{aro}), 128.0 (CH_{aro}), 128.1 (2x CH_{aro}), 128.4 (2x CH_{aro}), 128.8 (2x CH_{aro}), 128.9 (2x CH_{aro}), 130.2 (2x CH_{aro}), 136.4 ($\text{C}^q_{\text{aro-C}}$), 136.6 ($\text{C}^q_{\text{aro-C}}$), 139.8 (2x $\text{C}^q_{\text{aro-C}}$), 141.6 (2x $\text{C}^q_{\text{aro-C}}$), 156.8 ($\text{C}_2=\text{O}$), 171.8 ($\text{C}_4=\text{O}$); Anal. Calcd. for $\text{C}_{29}\text{H}_{22}\text{N}_2\text{O}_2$ (430.17): C, 80.91; H, 5.15; N, 6.51. Found: C, 80.86; H, 5.08; N, 6.43.

3-Benzylspiro(fluorene-9,5'-imidazolidine)-2,4-dione (11g). This product as a major isomer was isolated as white solid from hydantoin **9If** in 39% yield and melted at 194 °C (ethanol); Rf (cyclohexane/AcOEt: 3/2)=0.74; IR (KBr): ν 3275 (NH), 1774 ($\text{C}=\text{O}$), 1722 ($\text{C}=\text{O}$) cm^{-1} ; ^1H NMR (CDCl_3): δ 4.78 (s, 2H, CH_2). 5.56 (s broad, 1H, NH). 7.20-7.47 (m, 11H, H_{aro}). 7.67-7.70 (m, 2H, H_{aro}); ^{13}C NMR (CDCl_3): δ 42.9 (CH_2). 71.5 (C^q). 120.9 (2x CH_{aro}). 123.4 (2x CH_{aro}). 128.1 (CH_{aro}). 128.4 (2x CH_{aro}). 128.5 (2x CH_{aro}). 128.8 (2x CH_{aro}). 130.3 (2x CH_{aro}). 136.1 ($\text{C}^q_{\text{aro-C}}$). 141.2 (2x $\text{C}^q_{\text{aro-C}}$). 141.8 (2x $\text{C}^q_{\text{aro-C}}$). 157.4 ($\text{C}_2=\text{O}$). 172.2 ($\text{C}_4=\text{O}$); Anal. Calcd. for $\text{C}_{22}\text{H}_{16}\text{N}_2\text{O}_2$ (340.12): C, 77.63; H, 4.74; N, 8.23. Found: C, 77.50; H, 4.66; N, 8.14.

1,3-Dibenzylspiro(imidazolidine-5,3'-pyrrolo[1,2-*a*]indole)-2,4-dione (10h). This product as a minor isomer was isolated as an orange-white viscous oil from hydantoin **9IIIh** in 39% yield (ethanol; decomposition); Rf (cyclohexane/AcOEt: 2/3)=0.75; IR (neat): ν 1773 ($\text{C}=\text{O}$), 1716 ($\text{C}=\text{O}$) cm^{-1} ; ^1H

NMR (CDCl₃): δ 4.04 (d, 1H, $J=15$ Hz, CH₂), 4.41 (d, 1H, $J=15$ Hz, CH₂), 4.81 (s, 2H, CH₂), 5.97 (d, 1H, $J=2.3$ Hz, H_{pyrrole}), 6.30 (dd, 1H, $J=2.3$ and 3.1 Hz, H_{pyrrole}), 6.68-6.84 (m, 4H, H_{aro}), 6.93-7.64 (m, 11H, H_{aro} + H_{pyrrole}); ¹³C NMR (CDCl₃): δ 43.2 (CH₂), 44.9 (CH₂), 68.8 (C^q), 106.3 (CH_{aro}), 110.3 (CH_{aro}), 110.4 (CH_{aro}), 114.5 (CH_{aro}), 124.2 (CH_{aro}), 124.6 (CH_{aro}), 127.5 (CH_{aro}), 128.1 (2x CH_{aro}), 128.4 (2x CH_{aro}), 128.6 (2x CH_{aro}), 128.9 (2x CH_{aro}), 130.5 (CH_{aro}), 132.9 (C^q_{aro}), 134.2 (CH_{aro}), 136.0 (2x C^q_{aro}), 136.2 (C^q_{aro}), 140.8 (C^q_{aro}), 156.2 (C₂=O), 171.1 (C₄=O); MS, m/z : 419 M⁺; Anal. Calcd. for C₂₇H₂₁N₃O₂ (419.16): C, 77.31; H, 5.05; N, 10.02. Found: C, 77.21; H, 4.89; N, 9.89.

General Procedure for the Regioselective NaBH₄ Reduction of *N*₁,*N*₃-Dibenzylated Spirohydantoins 10a-h; Access to Spirohydroxylactams 12a-h. To a well stirred solution of *N*₁,*N*₃-dibenzylspirohydantoins (**10a-h**, 5 mmoles) in dry ethanol (60 mL) was added in portions at ambient temperature sodium borohydride (1.14 g, 30 mmoles) over a period of 10 minutes. After complete reaction on refluxing (1.5 to 24 hours; the reaction was monitored by TLC using precoated plate of silica gel and CH₂Cl₂ as eluent), the reaction was cold and the excess of sodium borohydride was decomposed by careful addition of 10% HCl solution to pH=3. After removal of the solvent under reduced pressure, the residue was diluted with H₂O (40 mL) and extracted twice with 2x30 mL of CH₂Cl₂. The organic layers were separated, washed with water, treated with brine and again decanted. The new organic phase was then dried over MgSO₄, filtered and concentrated under reduced pressure to give a solid which was recrystallized from suitable solvent to give corresponding 4-hydroxyspiroimidazolidin-2-ones **12a-h** in good yields as following.

1,3-Dibenzyl-4-hydroxyspiro(imidazolidine-5,1'-indane)-2-one (12a). This product was isolated from spirohydantoin **10a** as white crystals in 94% yield; mp 56 °C (ethanol/water); R_f (cyclohexane/AcOEt: 3/2)=0.68; IR (KBr): ν 3362 (OH), 1698 (C=O) cm⁻¹; ¹H NMR (CDCl₃): δ 1.84-2.16 (m, 2H, CH₂); 2.71-2.77 (m, 2H, CH₂), 3.88 (d, 1H, $J=15$ Hz, CH₂), 4.25 (d, 1H, $J=15$ Hz, CH₂), 4.64 (d, 1H, $J=15$ Hz, CH₂), 4.74 (d, 1H, $J=9$ Hz, CH), 4.87 (d, 1H, $J=15$ Hz, CH₂), 7.06-7.36 (m, 15H, H_{aro} + OH); ¹³C NMR (CDCl₃): δ 29.1 (CH₂), 35.3 (CH₂), 44.5 (CH₂), 44.8 (CH₂), 75.5 (C^q), 84.9 (CH), 125.5 (CH_{aro}), 125.6 (CH_{aro}), 126.9 (CH_{aro}), 127.1 (CH_{aro}), 127.6 (CH_{aro}), 127.8 (2x CH_{aro}), 128.4 (2x

CH_{aro}), 128.6 (2x CH_{aro}), 128.8 (2x CH_{aro}), 129.4 (CH_{aro}), 137.4 (C^q_{aro}-C), 137.5 (C^q_{aro}-C), 139.4 (C^q_{aro}-C), 145.1 (C^q_{aro}-C), 159.5 (C=O); Anal. Calcd. for C₂₅H₂₄N₂O₂ (384.18): C, 78.10; H, 6.29; N, 7.29. Found: C, 78.01; H, 6.21; N, 7.15.

1,3-Dibenzyl-4-hydroxyspiro(imidazolidine-5,2'-indane)-2-one (12b). This product was isolated from spirohydantoin **10b** as white crystals in 91% yield; mp 129 °C (ethanol); R_f (cyclohexane/AcOEt: 3/2)=0.55; IR (KBr): ν 3429 (OH), 1643 (C=O) cm⁻¹; ¹H NMR (CDCl₃): δ 2.12 (d, 1H, *J*=7 Hz, OH), 2.72 (d, 1H, *J*=16 Hz, CH₂), 2.86 (d, 1H, *J*=16 Hz, CH₂), 2.98 (d, 1H, *J*=16 Hz, CH₂), 3.28 (d, 1H, *J*=16 Hz, CH₂), 4.12 (d, 1H, *J*=16 Hz, CH₂), 4.23 (d, 1H, *J*=16 Hz, CH₂), 4.53 (d, 1H, *J*=7 Hz, CH), 4.62 (d, 1H, *J*=16 Hz, CH₂), 4.90 (d, 1H, *J*=16 Hz, CH₂), 7.04-7.32 (m, 14H, H_{aro}); ¹³C NMR (CDCl₃): δ 35.5 (CH₂), 41.7 (CH₂), 43.9 (CH₂), 44.4 (CH₂), 71.6 (C^q), 84.6 (CH), 124.6 (CH_{aro}), 124.7 (CH_{aro}), 127.0 (CH_{aro}), 127.1 (CH_{aro}), 127.2 (CH_{aro}), 127.5 (2x CH_{aro}), 127.6 (CH_{aro}), 128.3 (2x CH_{aro}), 128.5 (2x CH_{aro}), 128.8 (2x CH_{aro}), 137.6 (C^q_{aro}-C), 139.2 (C^q_{aro}-C), 139.4 (C^q_{aro}-C), 140.2 (C^q_{aro}-C), 159.0 (C=O); Anal. Calcd. for C₂₅H₂₄N₂O₂ (384.18): C, 78.10; H, 6.29; N, 7.29. Found: C, 78.04; H, 6.01; N, 7.14.

1,3-Dibenzyl-4-hydroxyspiro(imidazolidine-5,1'-tetraline)-2-one (12c). This product was isolated from spirohydantoin **10c** as white crystals in 97% yield; mp 67 °C (ethanol); R_f (cyclohexane/AcOEt: 3/2)=0.67; IR (KBr): ν 3389 (OH), 1698 (C=O) cm⁻¹; ¹H NMR (CDCl₃): δ 1.34 (d, 1H, *J*=11 Hz, OH), 1.56-1.78 (m, 4H, 2x CH₂), 2.69-2.72 (m, 2H, CH₂), 3.56 (d, 1H, *J*=16 Hz, CH₂), 4.25 (d, 1H, *J*=16 Hz, CH₂), 4.71 (d, 1H, *J*=11 Hz, CH), 4.83 (d, 1H, *J*=16 Hz, CH₂), 4.91 (d, 1H, *J*=16 Hz, CH₂), 7.09-7.36 (m, 14 H, H_{aro}); ¹³C NMR (CDCl₃): δ 18.5 (CH₂), 28.9 (CH₂), 34.8 (CH₂), 44.8 (CH₂), 44.9 (CH₂), 66.4 (C^q), 86.8 (CH), 126.6 (CH_{aro}), 127.0 (CH_{aro}), 127.1 (CH_{aro}), 127.5 (2x CH_{aro}), 128.4 (2x CH_{aro}), 128.5 (2x CH_{aro}), 128.6 (CH_{aro}), 128.7 (CH_{aro}), 128.8 (2x CH_{aro}), 130.4 (CH_{aro}), 131.5 (C^q_{aro}-C), 137.5 (C^q_{aro}-C), 139.7 (C^q_{aro}-C), 140.6 (C^q_{aro}-C), 159.3 (C=O); Anal. Calcd. for C₂₆H₂₆N₂O₂ (398.20): C, 78.36; H, 6.58; N, 7.03. Found: C, 78.21; H, 6.43; N, 6.98.

1,3-Dibenzyl-4-hydroxyspiro(imidazolidine-5,2'-tetraline)-2-one (12d). This product was isolated from spirohydantoin **10d** as white crystals in 99% yield; mp 81 °C (ethanol/diethyl ether); R_f (cyclohexane/AcOEt: 3/2)=0.65; IR (KBr): ν 3351 (OH), 1698 (C=O) cm⁻¹; ¹H NMR (CDCl₃): δ 1.83-

2.04 (m, 2H, CH₂), 2.20 (d, 1H, *J*=9 Hz, OH), 2.70-3.01 (m, 4H, 2x CH₂), 4.21 (d, 1H, *J*=15 Hz, CH₂), 4.24 (d, 1H, *J*=15 Hz, CH₂), 4.47 (d, 1H, *J*=9 Hz, CH), 4.67 (d, 1H, *J*=15 Hz, CH₂), 4.90 (d, 1H, *J*=15 Hz, CH₂), 6.96-7.09 (m, 3H, H_{aro}), 7.12-7.38 (m, 11H, H_{aro}); ¹³C NMR (CDCl₃): δ 18.6 (CH₂), 28.8 (CH₂), 34.9 (CH₂), 44.6 (CH₂), 44.8 (CH₂), 66.1 (C^q), 86.7 (CH), 126.4 (CH_{aro}), 122.9 (CH_{aro}), 127.2 (CH_{aro}), 127.6 (2x CH_{aro}), 128.2 (2x CH_{aro}), 128.5 (2x CH_{aro}), 128.6 (CH_{aro}), 128.7 (CH_{aro}), 128.9 (2x CH_{aro}), 130.7 (CH_{aro}), 131.8 (C^q_{aro}-C), 136.9 (C^q_{aro}-C), 138.9 (C^q_{aro}-C), 140.0 (C^q_{aro}-C), 159.7 (C=O); Anal. Calcd. for C₂₆H₂₆N₂O₂ (398.20): C, 78.36; H, 6.58; N, 7.03. Found: C, 78.31; H, 6.39; N, 6.90.

1,3-Dibenzyl-4-hydroxyspiro(imidazolidine-5,4'-thiochromane)-2-one (12e). This product was isolated from spirohydantoin **10e** as white crystals in 97% yield; mp 190 °C (ethanol); R_f (cyclohexane/AcOEt: 3/2)=0.52; IR (KBr): ν 3385 (OH), 1702 (C=O) cm⁻¹; ¹H NMR (CDCl₃): δ 1.60 (d, 1H, *J*=9 Hz, OH), 1.74-1.99 (m, 1H, CH₂), 2.01-2.14 (m, 1H, CH₂), 2.61-2.67 (m, 1H, CH₂), 3.20-3.32 (m, 1H, CH₂), 3.52 (d, 1H, *J*=16 Hz, CH₂), 4.26 (d, 1H, *J*=16 Hz, CH₂), 4.64 (d, 1H, *J*=9 Hz, CH), 4.80 (d, 1H, *J*=16 Hz, CH₂), 4.98 (d, 1H, *J*=16 Hz, CH₂), 7.09-7.33 (m, 14H, H_{aro}); ¹³C NMR (CDCl₃): δ 21.9 (CH₂), 33.4 (CH₂), 44.7 (CH₂), 44.9 (CH₂), 65.3 (C^q), 86.8 (CH), 124.5 (CH_{aro}), 127.1 (CH_{aro}), 127.5 (CH_{aro}), 127.6 (2x CH_{aro}), 127.7 (CH_{aro}), 127.8 (C^q_{aro}-S), 128.2 (CH_{aro}), 128.5 (2x CH_{aro}), 128.6 (2x CH_{aro}), 128.7 (2x CH_{aro}), 128.9 (CH_{aro}), 137.0 (C^q_{aro}-S), 137.5 (C^q_{aro}-C), 139.3 (C^q_{aro}-C), 159.1 (C=O); Anal. Calcd. for C₂₅H₂₄N₂O₂S (416.16): C, 72.09; H, 5.81; N, 6.73. Found: C, 71.91; H, 5.59; N, 6.51.

6H,7H,8H-1,3-Dibenzyl-4-hydroxyspiro(imidazolidine-5,4'-indole)-2-one (12f). This product was isolated from spirohydantoin **10f** as white crystals in 99% yield; mp 58 °C (ethanol/diethyl ether); R_f (cyclohexane/AcOEt: 3/2)=0.53; IR (KBr): ν 3436 (OH), 1693 (C=O) cm⁻¹; ¹H NMR (CDCl₃): δ 1.46-1.70 (m, 4H, 2x CH₂), 2.19-2.43 (m, 2H, CH₂), 3.43 (s, 3H, CH₃), 3.61 (d, 1H, *J*=15 Hz, CH₂), 4.20 (d, 1H, *J*=15 Hz, CH₂), 4.55 (s, 1H, CH), 4.67 (d, 1H, *J*=15 Hz, CH₂), 4.74 (d, 1H, *J*=15 Hz, CH₂), 4.80 (s, 1H, OH), 5.85 (d, 1H, *J*=2 Hz, H_{pyrrole}), 6.50 (d, 1H, *J*=2 Hz, H_{pyrrole}), 7.09-7.30 (m, 10H, H_{aro}); ¹³C NMR (CDCl₃): δ 19.6 (CH₂), 21.0 (CH₂), 33.5 (CH₃), 34.2 (CH₂), 44.7 (2x CH₂), 65.2 (C^q), 85.4 (CH), 104.8 (CH_{pyrrole}), 111.9 (C^q_{pyrrole}), 121.9 (CH_{pyrrole}), 126.7 (CH_{aro}), 127.3 (CH_{aro}), 127.4 (2x CH_{aro}), 128.3 (2x

CH_{aro}), 128.5 (2x CH_{aro}), 128.6 (2x CH_{aro}), 133.1 (C^q_{aro}-C), 137.9 (C^q_{aro}-C), 140.4 (C^q_{pyrrole}), 159.4 (C=O); Anal. Calcd. for C₂₅H₂₇N₃O₂ (401.21): C, 74.79; H, 6.78; N, 10.47. Found: C, 74.67; H, 6.60; N, 10.23.

1,3-Dibenzyl-4-hydroxyspiro(fluorene-9,5'-imidazolidine)-2-one (12g). This product was isolated from spirohydantoin **10g** as white crystals in 96% yield; mp 185 °C (ethanol); R_f (cyclohexane/AcOEt: 4/1)=0.52; IR (KBr): ν 3376 (OH), 1702 (C=O) cm⁻¹; ¹H NMR (CDCl₃): δ 1.98 (d, 1H, *J*=8 Hz, OH), 3.90 (d, 1H, *J*=15 Hz, CH₂), 3.99 (d, 1H, *J*=15 Hz, CH₂), 4.32 (d, 1H, *J*=15 Hz, CH₂), 4.87 (d, 1H, *J*=8 Hz, CH), 4.94 (d, 1H, *J*=15 Hz, CH₂), 6.79 (dd, 2H, *J*=2 and 8 Hz, H_{aro}), 6.91-7.12 (m, 5H, H_{aro}), 7.22-7.36 (m, 9H, H_{aro}), 7.55 (d, 1H, *J*=1.6 et 8.6 Hz, H_{aro}), 7.63 (d, 1H, *J*=8 Hz, H_{aro}); ¹³C NMR (CDCl₃): δ 45.2 (CH₂), 45.3 (CH₂), 74.2 (C^q), 83.9 (CH), 119.9 (CH_{aro}), 120.3 (CH_{aro}), 123.9 (CH_{aro}), 126.8 (CH_{aro}), 127.3 (CH_{aro}), 127.4 (CH_{aro}), 127.6 (CH_{aro}), 127.8 (2x CH_{aro}), 128.0 (CH_{aro}), 128.6 (2x CH_{aro}), 128.7 (2x CH_{aro}), 128.8 (2x CH_{aro}), 129.3 (CH_{aro}), 129.7 (CH_{aro}), 137.2 (C^q_{aro}-C), 138.2 (C^q_{aro}-C), 139.2 (C^q_{aro}-C), 140.1 (C^q_{aro}-C), 141.6 (C^q_{aro}-C), 144.2 (C^q_{aro}-C), 159.9 (C=O); Anal. Calcd. for C₂₉H₂₄N₂O₂ (432.18): C, 80.53; H, 5.59; N, 6.48. Found: C, 80.48; H, 5.47; N, 6.31.

1,3-Dibenzyl-4-hydroxyspiro(imidazolidine-5,3'-pyrrolo[1,2-*a*]indole)-2-one (12h). This product was obtained from spirohydantoin **10h** as brown crystals in 86% yield; Because of it was not isolated in pure form, it was used in the next without other purifications. R_f (cyclohexane/AcOEt: 2/3)=0.68; ¹H NMR (CDCl₃): δ 2.32 (d, 1H, *J*=7 Hz, OH), 3.83 (d, 1H, *J*=15 Hz, CH₂), 4.18 (d, 1H, *J*=15 Hz, CH₂), 4.29 (d, 1H, *J*=15 Hz, CH₂), 4.89 (d, 1H, *J*=7 Hz, CH), 4.93 (d, 1H, *J*=15 Hz, CH₂), 5.85 (d, 1H, *J*=3.1 Hz, H_{pyrrole}), 6.19 (t, 1H, *J*=3.1 Hz, H_{pyrrole}), 6.32 (d, 1H, *J*=2.3 Hz, H_{pyrrole}), 6.79-7.40 (m, 14H, H_{aro}).

General Procedure for the Regioselective LiAlH₄ Reduction of N₃-Benzylated Spirohydantoins 11a-g; Access to Spirohydroxylactams 13a-g. To a well stirred solution of LiAlH₄ (228 mg, 6 mmoles) in 10 mL of freshly distilled THF at -5-0 °C, was added in dropwise a solution of spirohydantoins **11a-g** (584 mg, 2 mmoles). After reaction at room temperature for 12 h, the excess of lithium aluminohydride was decomposed by careful addition of a mixture of EtOH-5% HCl. After filtration on short column of celite, removal of the solvent under reduced pressure, the residue was diluted with H₂O (40 mL) and extracted twice with 2x30 mL of CH₂Cl₂. The organic phase was separated, dried with MgSO₄, filtered

and concentrated under reduced pressure, to give a solid which was recrystallized from suitable solvent to furnish corresponding 4-hydroxyspiroimidazolidin-2-ones **13a-g** in good yields (see text for more details).

3-Benzyl-4-hydroxyspiro(imidazolidine-5,1'-indane)-2-one (13a). This product was isolated from spirohydantoin **11a** as white crystals in 74% yield and melted at 205 °C (ethanol); R_f (AcOEt)=0.51; IR (KBr): ν 3233 (NH and OH), 1687 (C=O) cm⁻¹; ¹H NMR (DMSO-d₆): δ 1.81-2.12 (m, 2H, CH₂ + OH), 2.66-2.78 (m, 3H, CH₂), 4.01 (d, 1H, *J*=15 Hz, CH₂), 4.48 (s, 1H, CH), 4.60 (d, 1H, *J*=15 Hz, CH₂), 7.01-7.36 (m, 10H, H_{aro} + NH); ¹³C NMR (DMSO-d₆): δ 37.5 (CH₂), 42.7 (CH₂), 44.7 (CH₂), 66.8 (C^q), 83.9 (CH), 124.0 (CH_{aro}), 124.5 (CH_{aro}), 126.3 (CH_{aro}), 126.5 (CH_{aro}), 126.9 (CH_{aro}), 127.6 (2x CH_{aro}), 128.7 (2x CH_{aro}), 138.7 (C^q_{aro}-C), 139.7 (C^q_{aro}-C), 141.1 (C^q_{aro}-C), 159.5 (C=O); Anal. Calcd. for C₁₈H₁₈N₂O₂ (294.14): C, 73.45; H, 6.16; N, 9.52. Found: C, 73.31; H, 6.09; N, 9.44.

3-Benzyl-4-hydroxyspiro(imidazolidine-5,2'-indane)-2-one (13b). This product was isolated from spirohydantoin **11b** as white crystals in 85% yield and melted at mp>300 °C (ethanol); R_f (cyclohexane/AcOEt: 1/4)=0.46; IR (KBr): ν 3236 (NH and OH), 1686 (C=O) cm⁻¹; ¹H NMR (DMSO-d₆): δ 2.73 (d, 1H, *J*=16 Hz, CH₂), 2.78 (d, 1H, *J*=16 Hz, CH₂), 2.88 (d, 1H, *J*=16 Hz, CH₂), 3.32 (d, 1H, *J*=16 Hz, CH₂), 3.96 (d, 1H, *J*=16 Hz, CH₂), 4.39 (s, 1H, CH), 4.52 (d, 1H, *J*=16 Hz, CH₂), 6.26 (s broad, 1H, NH), 7.10-7.34 (m, 10H, H_{aro} + OH); ¹³C NMR (DMSO-d₆): δ 37.4 (CH₂), 42.4 (CH₂), 44.8 (CH₂), 66.5 (C^q), 84.5 (CH), 124.3 (CH_{aro}), 124.6 (CH_{aro}), 126.5 (CH_{aro}), 126.7 (CH_{aro}), 126.9 (CH_{aro}), 127.5 (2x CH_{aro}), 128.5 (2x CH_{aro}), 138.6 (C^q_{aro}-C), 139.9 (C^q_{aro}-C), 141.0 (C^q_{aro}-C), 159.3 (C=O); Anal. Calcd. for C₁₈H₁₈N₂O₂ (294.14): C, 73.45; H, 6.16; N, 9.52. Found: C, 73.38; H, 6.01; N, 9.36.

3-Benzyl-4-hydroxyspiro(imidazolidine-5,2'-tetraline)-2-one (13c). This product was isolated from spirohydantoin **11c** as white crystals in 86% yield and melted at 146 °C (ethanol); R_f (cyclohexane/AcOEt: 1/4)=0.59; IR (KBr): ν 3332 (NH and OH), 1678 (C=O) cm⁻¹; ¹H NMR (DMSO-d₆): δ 1.72 (d, 1H, *J*=10 Hz, OH), 1.82-1.90 (m, 4H, 2x CH₂), 2.71-2.80 (m, 2H, CH₂), 4.15 (d, 1H, *J*=15 Hz, CH₂), 4.69 (s broad, 1H, NH), 4.71 (d, 1H, *J*=10 Hz, CH), 4.79 (d, 1H, *J*=15 Hz, CH₂), 7.05-7.46 (m,

9H, H_{aro}); ¹³C NMR (DMSO-d₆): δ 18.7 (CH₂), 28.7 (CH₂), 35.3 (CH₂), 42.4 (CH₂), 60.2 (C^q), 84.9 (CH), 124.7 (CH_{aro}), 126.8 (CH_{aro}), 126.9 (CH_{aro}), 127.4 (2x CH_{aro}), 128.1 (CH_{aro}), 128.5 (2x CH_{aro}), 130.1 (CH_{aro}), 136.3 (C^q_{aro}-C), 137.5 (C^q_{aro}-C), 138.6 (C^q_{aro}-C), 159.3 (C=O); Anal. Calcd. for C₁₉H₂₀N₂O₂ (308.15): C, 74.00; H, 6.54; N, 9.08. Found: C, 73.88; H, 6.38; N, 9.00.

3-Benzyl-4-hydroxyspiro(imidazolidine-5,1'-tetraline)-2-one (13d). This product was isolated from spirohydantoin **11d** as white crystals in 71% yield and melted at 187 °C (ethanol); R_f (AcOEt)=0.67; IR (KBr): ν 3271 (NH and OH), 1692 (C=O) cm⁻¹; ¹H NMR (DMSO-d₆): δ 1.59-2.03 (m, 2H, CH₂), 2.64-3.15 (m, 4H, 2x CH₂), 3.99 (d, 1H, *J*=16 Hz, CH₂), 4.29 (s broad, 1H, OH), 4.50 (s, 1H, CH), 4.56 (d, 1H, *J*=16 Hz, CH₂), 6.16 (s broad, 1H, NH), 6.92-7.34 (m, 9H, H_{aro}); ¹³C NMR (DMSO-d₆): δ 18.9 (CH₂), 28.6 (CH₂), 35.6 (CH₂), 42.7 (CH₂), 60.1 (C^q), 84.7 (CH), 124.4 (CH_{aro}), 126.6 (CH_{aro}), 126.8 (CH_{aro}), 127.2 (2x CH_{aro}), 128.0 (CH_{aro}), 128.2 (2x CH_{aro}), 129.8 (CH_{aro}), 136.1 (C^q_{aro}-C), 137.3 (C^q_{aro}-C), 138.5 (C^q_{aro}-C), 159.1 (C=O); Anal. Calcd. for C₁₉H₂₀N₂O₂ (308.15): C, 74.00; H, 6.54; N, 9.08. Found: C, 73.82; H, 6.33; N, 8.89.

3-Benzyl-4-hydroxyspiro(imidazolidine-5,4'-thiochromane)-2-one (13e). This product was isolated from spirohydantoin **11e** as white crystals in 97% yield and melted at 167 °C (ethanol); R_f (Cyclohexane/AcOEt: 3/2)=0.52; IR (KBr): ν 3338 (NH and OH), 1686 (C=O) cm⁻¹; ¹H NMR (DMSO-d₆): δ 2.04-2.14 (m, 2H, CH₂), 2.87-3.19 (m, 2H, CH₂), 4.03 (d, 1H, *J*=16 Hz, CH₂), 4.57 (d, 1H, *J*=16 Hz, CH₂), 4.70 (s, 1H, CH), 5.99 (s broad, 1H, NH), 7.00-7.45 (m, 10H, H_{aro} + OH); ¹³C NMR (DMSO-d₆): δ 29.2 (CH₂), 33.9 (CH₂), 42.6 (CH₂), 59.4 (C^q), 84.9 (CH), 123.3 (CH_{aro}), 125.6 (CH_{aro}), 126.9 (CH_{aro}), 127.4 (3x CH_{aro}), 128.5 (2x CH_{aro}), 130.3 (CH_{aro}), 133.5 (C^q_{aro}-S), 133.7 (C^q_{aro}-C), 138.4 (C^q_{aro}-C), 159.0 (C=O); Anal. Calcd. for C₁₈H₁₈N₂O₂S (326.11): C, 66.23; H, 5.56; N, 8.58. Found: C, 66.11; H, 5.45; N, 8.41.

Both isomers of 5'*H*-3-Benzyl-6',7'-dihydro-4-hydroxy-1'-methyl(imidazolidine-5,4'-indole)-2-one (13f). This product, as a mixture of inseparable two diastereomers in 1/1 ratio, was obtained from spirohydantoin **11f** as white crystals in 96% yield (ethanol); R_f (AcOEt)=0.47; ¹H NMR (DMSO-d₆): **Isomer A**, δ 1.40-2.06 (m, 5H, 2x CH₂ + OH), 2.32-2.45 (m, 2H, CH₂), 3.37 (s, 3H, CH₃), 3.99 (d, 1H,

$J=15$ Hz, CH₂), 4.40 (d, 1H, $J=15$ Hz, CH₂), 4.54 (s, 1H, CH), 5.59 (d, 1H, $J=2$ Hz, H_{pyrrole}), 6.47 (d, 1H, $J=2$ Hz, H_{pyrrole}), 6.80 (s broad, 1H, NH), 7.24-7.38 (m, 5H, H_{aro}); **Isomer B**, δ 1.40-2.06 (m, 5H, 2x CH₂ + OH), 2.32-2.45 (m, 2H, CH₂), 3.40 (s, 3H, CH₃), 4.01 (d, 1H, $J=15$ Hz, CH₂), 4.54 (d, 1H, $J=15$ Hz, CH₂), 4.56 (s, 1H, CH), 6.02 (d, 1H, $J=2$ Hz, H_{pyrrole}), 6.52 (d, 1H, $J=2$ Hz, H_{pyrrole}), 6.72 (s broad, 1H, NH), 7.24-7.38 (m, 5H, H_{aro}); Anal. Calcd. for C₁₈H₂₁N₃O₂ (311.16): C, 69.43; H, 6.80; N, 13.49. Found: C, 69.29; H, 6.57; N, 13.30.

3'-Benzyl-4'-ethoxyspiro(fluorene-9,5'-imidazolidine)-2'-one (13g). This product was isolated from spirohydantoin **11g** as white crystals in 98% yield and melted at 184 °C (ethanol); R_f (Cyclohexane/AcOEt: 3/2)=0.28; IR (KBr): ν 3299 (NH), 1702 (C=O) cm⁻¹; ¹H NMR (DMSO-d₆): δ 1.09 (t, 3H, $J=7$ Hz, CH₃), 3.38 (q, 2H, $J=7$ Hz, CH₂), 4.13 (d, 1H, $J=15$ Hz, CH₂), 4.69 (s, 1H, CH), 4.73 (d, 1H, $J=15$ Hz, CH₂), 6.55 (s broad, 1H, NH), 7.10-7.45 (m, 10H, H_{aro}), 7.60 (d, 1H, $J=7$ Hz, H_{aro}), 7.76 (d, 2H, $J=7.0$ Hz, H_{aro}); ¹³C NMR (DMSO-d₆): δ 23.5 (CH₃), 40.8 (CH₂), 65.5 (CH₂), 69.4 (C^q), 84.5 (CH), 107.0 (CH_{aro}), 119.8 (CH_{aro}), 120.1 (CH_{aro}), 122.8 (CH_{aro}), 127.1 (CH_{aro}), 127.2 (CH_{aro}), 127.7 (CH_{aro}), 128.0 (2x CH_{aro}), 128.5 (2x CH_{aro}), 128.6 (CH_{aro}), 128.7 (CH_{aro}), 138.2 (C^q_{aro}-C), 139.2 (C^q_{aro}-C), 140.2 (C^q_{aro}-C), 143.3 (C^q_{aro}-C), 148.1 (C^q_{aro}-C), 160.3 (C=O); Anal. Calcd. for C₂₄H₂₂N₂O₂ (370.17): C, 77.81; H, 5.99; N, 7.56. Found: C, 77.63; H, 5.81; N, 7.39.

General Procedure for the Transposition/Cyclization of Spirohydroxylactams **12a-h** and **13a-g**.

To a well-stirred and cold solution of spirohydroxylactams **12a-h** and **13a-g** (1 mmole) in 10 mL of dry dichloromethane, was added dropwise a solution of trifluoroacetic acid anhydride (0.18 mL, 1.3 mmoles) and trifluoroacetic acid (0.096 mL, 1.3 mmoles) in 10 mL of dry dichloromethane. After reaction at reflux for 24 h under stirring, the reaction mixture was diluted carefully with water (25 mL) and neutralized with a cold 10% NaHCO₃ aqueous solution. The solution was extracted twice with dichloromethane (10 mL). The organic layer was washed with water, brine, separated, dried over MgSO₄ and evaporated in vacuo. The resulting residue was purified by flash chromatography on silica gel column using a mixture of cyclohexane/AcOEt (1/1) to give the cyclic isoimidazolones products **14** and **15** as crystalline materials in yields ranging from 38 to 97%.

1,3-Dibenzyl-4,5-dihydrobenzo[*e*]benzimidazol-2-one (14a). This product was isolated from spirohydroxylactam **12a** as white crystals in 97% yield and melted at 110 °C (ethanol/diethyl ether); Rf (cyclohexane/AcOEt: 3/2)=0.61; IR (KBr): ν 3015 and 2989 (CH), 1685 (C=O and C=C) cm^{-1} ; ^1H NMR (CDCl_3): δ 2.48 (t, 2H, $J=7$ Hz, CH_2), 2.85 (t, 2H, $J=7$ Hz, CH_2), 4.96 (s, 2H, CH_2), 5.26 (s, 2H, CH_2), 6.94-7.11 (m, 3H, H_{aro}), 7.20-7.32 (m, 11H, H_{aro}); ^{13}C NMR (CDCl_3): δ 19.7 (CH_2), 28.9 (CH_2), 45.1 (CH_2), 46.5 (CH_2), 117.9 ($\text{C}^{\text{q}}_{=}$), 119.5 (CH_{aro}), 121.7 ($\text{C}^{\text{q}}_{=}$), 125.3 (CH_{aro}), 126.5 (3x CH_{aro}), 127.0 (CH_{aro}), 127.2 (3x CH_{aro}), 127.4 (CH_{aro}), 127.5 ($\text{C}^{\text{q}}_{\text{aro-C}}$), 127.7 (CH_{aro}), 128.4 (CH_{aro}), 128.9 (2x CH_{aro}), 133.2 ($\text{C}^{\text{q}}_{\text{aro-C}}$), 137.4 ($\text{C}^{\text{q}}_{\text{aro-C}}$), 137.8 ($\text{C}^{\text{q}}_{\text{aro-C}}$), 154.9 (C=O); MS, m/z : 366 M^+ ; Anal. Calcd. for $\text{C}_{25}\text{H}_{22}\text{N}_2\text{O}$ (366.17): C, 81.94; H, 6.05; N, 7.64. Found: C, 81.86; H, 5.96; N, 7.43.

1-Benzyl-4,5-dihydrobenzo[*e*]benzimidazol-2(3*H*)-one (15a). This product was isolated from spirohydroxylactam **13a** as white crystals in 92% yield and melted at 211 °C (ethanol); Rf (AcOEt)=0.38; IR (KBr): ν 3450 (NH), 3023 and 2980 (CH), 1680 (C=O and C=C) cm^{-1} ; ^1H NMR (CDCl_3): δ 2.67 (t, 2H, $J=7$ Hz, CH_2), 2.94 (t, 2H, $J=7$ Hz, CH_2), 5.24 (s, 2H, CH_2), 6.97-7.38 (m, 9H, H_{aro}), 10.68 (s broad, 1H, NH); ^{13}C NMR (CDCl_3): δ 20.4 (CH_2), 29.2 (CH_2), 45.9 (CH_2), 118.4 ($\text{C}^{\text{q}}_{=}$), 119.3 (CH_{aro}), 120.8 ($\text{C}^{\text{q}}_{=}$), 125.2 (CH_{aro}), 126.3 (2x CH_{aro}), 126.9 (CH_{aro}), 127.4 (CH_{aro}), 127.8 ($\text{C}^{\text{q}}_{\text{aro-C}}$), 128.5 (CH_{aro}), 128.9 (2x CH_{aro}), 133.6 ($\text{C}^{\text{q}}_{\text{aro-C}}$), 137.6 ($\text{C}^{\text{q}}_{\text{aro-C}}$), 155.9 (C=O); MS, m/z : 276 M^+ ; Anal. Calcd. for $\text{C}_{18}\text{H}_{16}\text{N}_2\text{O}$ (276.13): C, 81.94; H, 6.05; N, 7.64. Found: C, 81.80; H, 6.00; N, 7.51.

1,3-Dibenzyl-4,9-dihydrobenzo[*f*]benzimidazol-2-one (14b). This product was isolated from spirohydroxylactam **12b** as white crystals in 38% yield and melted at 134 °C (ethanol/diethyl ether); Rf (cyclohexane/AcOEt: 3/2)=0.50; IR (KBr): ν 3038 and 2984 (CH), 1705 (C=O), 1671 (C=C) cm^{-1} ; ^1H NMR (CDCl_3): δ 3.61 (s, 4H, 2x CH_2), 4.96 (s, 4H, 2x CH_2), 7.06-7.37 (m, 14H, H_{aro}); ^{13}C NMR (CDCl_3): δ 25.6 (2x CH_2), 45.1 (2x CH_2), 114.3 (2x $\text{C}^{\text{q}}_{=}$), 126.5 (2x CH_{aro}), 127.2 (4x CH_{aro}), 127.6 (2x CH_{aro}), 128.8 (4x CH_{aro}), 129.6 (2x CH_{aro}), 131.7 (2x $\text{C}^{\text{q}}_{\text{aro-C}}$), 137.8 (2x $\text{C}^{\text{q}}_{\text{aro-C}}$), 154.3 (C=O); MS, m/z : 366 M^+ ; Anal. Calcd. for $\text{C}_{25}\text{H}_{22}\text{N}_2\text{O}$ (366.17): C, 81.94; H, 6.05; N, 7.64. Found: C, 81.81; H, 5.97; N, 7.42.

1,3-Dibenzylbenzo[f]benzimidazol-2-one (18). This product as a major cyclized isomer of **14b** was obtained from spirohydroxylactam **12b** as white crystals in 58% yield. It was also obtained as the sole reaction product by using same general protocol described above for synthesis of **14a,b** and **15a** from spirohydroxylactam **12b** after refluxing for 30 h or from cyclic benzimidazol-2-one derivative **14b** at room temperature for 12 h, respectively, in 78% or quantitative yield. This product was melted at 186 °C (ethanol/diethyl ether); R_f (cyclohexane/AcOEt: 3/2)=0.80; IR (KBr): ν 3015 and 2995 (CH), 1713 (C=O and C=C) cm⁻¹; ¹H NMR (CDCl₃): δ 5.26 (s, 4H, 2x CH₂), 7.31-7.46 (m, 14H, H_{aro}), 7.74-7.79 (m, 2H, H_{aro}); ¹³C NMR (CDCl₃): δ 45.2 (2x CH₂), 104.4 (2x CH_{aro}), 124.2 (2x CH_{aro}), 127.3 (2x CH_{aro}), 127.5 (4x CH_{aro}), 127.9 (2x CH_{aro}), 128.9 (4x CH_{aro}), 129.8 (2x C^q_{aro}-C), 130.2 (2x C^q_{aro}-C), 136.2 (2x C^q_{aro}-C), 155.5 (C=O), MS, m/z: 364 M⁺; Anal. Calcd. for C₂₅H₂₀N₂O (364.16): C, 82.39; H, 5.53; N, 7.69. Found: C, 82.21; H, 5.44; N, 7.58.

1-Benzyl-4,9-dihydrobenzo[f]benzimidazol-2(3H)-one (15b). This product was isolated from spirohydroxylactam **13b** as white crystals in 88% yield and melted at 238 °C (ethanol); R_f (AcOEt)=0.30; IR (KBr): ν 3450 (NH), 3021 and 2990 (CH), 1707 (C=O), 1676 (C=C) cm⁻¹; ¹H NMR (CDCl₃): δ 3.58 (t, 2H, *J*=5.5 Hz, CH₂), 3.79 (t, 2H, *J*=5.5 Hz, CH₂), 4.92 (s, 2H, CH₂), 7.13-7.28 (m, 9H, H_{aro}), 9.76 (s broad, 1H, NH); ¹³C NMR (CDCl₃): δ 25.6 (CH₂), 26.2 (CH₂), 44.6 (CH₂), 113.5 (C^q₌), 115.0 (C^q₌), 126.3 (CH_{aro}), 126.5 (CH_{aro}), 127.0 (2x CH_{aro}), 127.6 (CH_{aro}), 128.9 (2x CH_{aro}), 129.6 (2x CH_{aro}), 131.7 (C^q_{aro}-C), 132.4 (C^q_{aro}-C), 137.7 (C^q_{aro}-C), 155.5 (C=O); MS, m/z: 276 M⁺; Anal. Calcd. for C₁₈H₁₆N₂O (276.13): C, 78.24; H, 5.84; N, 10.14. Found: C, 78.11; H, 5.68; N, 10.05.

1,3-Dibenzyl-4*H*,5*H*,6*H*-benzo[c]cyclohepta[1,2-*d*]imidazol-2-one (14c). This product was isolated from spirohydroxylactam **12c** as white crystals in 94% yield and melted at 114 °C (ethanol/diethyl ether); R_f (cyclohexane/AcOEt: 3/2)=0.68; IR (KBr): ν 3022 and 2989 (CH), 1684 (C=O and C=C) cm⁻¹; ¹H NMR (CDCl₃): δ 1.79-1.92 (m, 2H, CH₂), 2.44 (t, 2H, *J*=7 Hz, CH₂), 2.41-2.57 (m, 2H, CH₂), 4.96 (s, 2H, CH₂), 5.02 (s, 2H, CH₂), 7.09-7.34 (m, 14H, H_{aro}); ¹³C NMR (CDCl₃): δ 23.8 (CH₂), 29.2 (CH₂), 33.2

(CH₂), 44.5 (CH₂), 46.4 (CH₂), 118.5 (C^q=), 122.1 (C^q=), 126.0 (CH_{aro}), 126.6 (CH_{aro}), 126.9 (4x CH_{aro}), 127.0 (CH_{aro}), 127.1 (CH_{aro}), 127.4 (CH_{aro}), 128.6 (2x CH_{aro}), 128.7 (CH_{aro}), 128.8 (2x CH_{aro}), 129.2 (C^q_{aro}-C), 137.5 (C^q_{aro}-C), 138.2 (C^q_{aro}-C), 141.4 (C^q_{aro}-C), 155.3 (C=O); MS, m/z: 380 M⁺; Anal. Calcd. for C₂₆H₂₄N₂O (380.19): C, 82.07; H, 6.36; N, 7.36. Found: C, 81.89; H, 6.21; N, 7.13.

1-Benzyl-3,4,5,6-tetrahydrobenzo[c]cyclohepta[1,2-d]imidazol-2-one (15c). This product was isolated from spirohydroxylactam **13c** as white crystals in 98% yield and melted at 120 °C (ethanol); Rf (cyclohexane/AcOEt)=0.28; IR (KBr): ν 3340 (NH), 3033 and 2991 (CH), 1683 (C=O and C=C) cm⁻¹; ¹H NMR (CDCl₃): δ 1.95-2.03 (m, 2H, CH₂), 2.56-2.67 (m, 4H, 2x CH₂), 4.94 (s, 2H, CH₂), 7.06-7.30 (m, 9H, H_{aro}), 10.26 (s broad, 1H, NH); ¹³C NMR (CDCl₃): δ 24.7 (CH₂), 28.6 (CH₂), 33.6 (CH₂), 46.0 (CH₂), 118.9 (C^q=), 121.9 (C^q=), 126.1 (CH_{aro}), 126.5 (CH_{aro}), 126.7 (3x CH_{aro}), 127.2 (CH_{aro}), 128.7 (2x CH_{aro}), 128.8 (C^q_{aro}-C), 129.4 (CH_{aro}), 138.2 (C^q_{aro}-C), 141.6 (C^q_{aro}-C), 156.3 (C=O); MS, m/z: 290 M⁺; Anal. Calcd. for C₁₉H₁₈N₂O (290.14): C, 82.07; H, 6.36; N, 7.36. Found: C, 82.00; H, 6.11; N, 7.23.

1,3-Dibenzyl-9,10-dihydrobenzo[c]cyclohepta[1,2-d]imidazol-2-one (14d). This product was isolated from spirohydroxy lactam **12d** as white crystals in 89% yield and melted at 72 °C (ethanol/diethyl ether); Rf (cyclohexane/AcOEt: 3/2)=0.50; IR (KBr): ν 3025 and 2985 (CH), 1683 (C=O), 1652 (C=C) cm⁻¹; ¹H NMR (CDCl₃): δ 2.48-2.55 (m, 2H, CH₂), 2.92-2.98 (m, 2H, CH₂), 3.60 (s, 2H, CH₂), 4.81 (s, 2H, CH₂), 5.03 (s, 2H, CH₂), 6.70 (dd, 1H, *J*=1.6 and 8.6 Hz, H_{aro}), 6.98-7.31 (m, 13H, H_{aro}); ¹³C NMR (CDCl₃): δ 24.6 (CH₂), 29.1 (CH₂), 31.2 (CH₂), 44.5 (CH₂), 44.6 (CH₂), 115.5 (C^q=), 117.3 (C^q=), 126.5 (CH_{aro}), 126.7 (2x CH_{aro}), 127.2 (2x CH_{aro}), 127.5 (2x CH_{aro}), 127.9 (CH_{aro}), 128.0 (CH_{aro}), 128.8 (2x CH_{aro}), 128.9 (2x CH_{aro}), 129.2 (CH_{aro}), 137.6 (C^q_{aro}-C), 137.8 (C^q_{aro}-C), 138.9 (C^q_{aro}-C), 140.1 (C^q_{aro}-C), 153.3 (C=O); MS, m/z: 380 M⁺; Anal. Calcd. for C₂₆H₂₄N₂O (380.19): C, 82.07; H, 6.36; N, 7.36. Found: C, 81.92; H, 6.19; N, 7.17.

1,3-Dibenzyl-4,5-dihydrobenzo[1]thiepine[4,5-d]imidazol-2-one (14e). This product was isolated from spirohydroxylactam **12e** as white crystals in 86% yield and melted at 64 °C (ethanol); Rf (cyclohexane/AcOEt: 3/2)=0.51; IR (KBr): ν 3033 and 2996 (CH), 1687 (C=O and C=C) cm⁻¹; ¹H NMR

(CDCl₃): δ 2.78-2.87 (m, 4H, 2x CH₂), 4.92 (s, 2H, CH₂), 4.98 (s, 2H, CH₂), 7.07-7.28 (m, 13H, H_{aro}), 7.47-7.51 (m, 1H, H_{aro}); ¹³C NMR (CDCl₃): δ 27.9 (CH₂), 36.4 (CH₂), 44.8 (CH₂), 46.8 (CH₂), 119.2 (C^q=), 123.8 (C^q=), 126.6 (CH_{aro}), 126.9 (2x CH_{aro}), 127.0 (2x CH_{aro}), 127.2 (CH_{aro}), 127.5 (CH_{aro}), 127.7 (CH_{aro}), 128.6 (2x CH_{aro}), 128.9 (2x CH_{aro}), 129.4 (CH_{aro}), 131.7 (C^q_{aro}-S), 133.9 (CH_{aro}), 137.0 (C^q_{aro}-C), 137.2 (C^q_{aro}-C), 138.1 (C^q_{aro}-C), 155.3 (C=O); MS, m/z: 398 M⁺; Anal. Calcd. for C₂₅H₂₂N₂OS (398.15): C, 75.35; H, 5.56; N, 7.03. Found: C, 75.26; H, 5.43; N, 6.88.

1-Benzyl-4,5-dihydrobenzo[1]thiepino[4,5-*d*]imidazol-2(3*H*)-one (15e). This product was isolated from spirohydroxylactam **13e** as white crystals in 92% yield and melted at 169 °C (ethanol); R_f (AcOEt)=0.43; IR (KBr): ν 3168 (NH), 3035 and 2986 (CH), 1686 (C=O and C=C) cm⁻¹; ¹H NMR (CDCl₃): δ 2.97 (s, 4H, 2x CH₂), 4.48 (s, 2H, CH₂), 7.11-7.54 (m, 9H, H_{aro}), 10.41 (s broad, 1H, NH); ¹³C NMR (CDCl₃): δ 28.7 (CH₂), 35.6 (CH₂), 46.3 (CH₂), 119.6 (C^q=), 123.5 (C^q=), 126.5 (CH_{aro}), 126.6 (2x CH_{aro}), 127.2 (CH_{aro}), 127.5 (CH_{aro}), 128.7 (2x CH_{aro}), 129.0 (CH_{aro}), 131.6 (C^q_{aro}-S), 133.9 (CH_{aro}), 137.1 (C^q_{aro}-C), 138.0 (C^q_{aro}-C), 156.2 (C=O); MS, m/z: 308 M⁺; Anal. Calcd. for C₁₈H₁₆N₂OS (308.10): C, 70.10; H, 5.23; N, 9.08. Found: C, 69.89; H, 5.07; N, 8.97.

4*H*,5*H*,6*H*-1,3-Dibenzyl-7-methylpyrrolo[3,2-*c*]cyclohepta[1,2-*d*]imidazol-2-one (14f). This product was isolated from spirohydroxylactam **12f** as yellow-white crystals in 99% yield and melted at 137 °C (ethanol); R_f (cyclohexane/AcOEt: 2/3)=0.47; IR (KBr): ν 3025 and 2989 (CH), 1685 (C=O), 1660 (C=C) cm⁻¹; ¹H NMR (CDCl₃): δ 1.89-1.95 (m, 2H, CH₂), 2.64 (t, 2H, *J*=7 Hz, CH₂), 2.73-2.79 (m, 2H, CH₂), 3.85 (s, 3H, CH₃), 4.98 (s, 2H, CH₂), 5.16 (s, 2H, CH₂), 6.98 (d, 1H, *J*=2 Hz, H_{pyrrole}), 7.23-7.38 (m, 11H, H_{aro} + H_{pyrrole}); ¹³C NMR (CDCl₃): δ 22.9 (CH₂), 25.1 (CH₂), 26.0 (CH₂), 33.3 (CH₃), 44.6 (CH₂), 45.9 (CH₂), 113.4 (C^q=), 118.9 (C^q=), 123.5 (C^q_{pyrrole}), 126.1 (2x CH_{aro}), 126.8 (3x CH_{aro}), 127.3 (CH_{pyrrole}), 127.5 (CH_{pyrrole}), 128.8 (3x CH_{aro}), 128.9 (2x CH_{aro}), 137.4 (C^q_{aro}-C), 137.6 (C^q_{aro}-C), 144.9 (C^q_{pyrrole}), 154.4 (C=O); MS, m/z: 383 M⁺; Anal. Calcd. for C₂₅H₂₅N₃O (383.20): C, 78.30; H, 6.57; N, 10.96. Found: C, 78.13; H, 6.44; N, 10.87.

4*H*,5*H*,6*H*-1-Benzyl-7-methylpyrrolo[3,2-*c*]cyclohepta[1,2-*d*]imidazol-2(3*H*)-one (15f). This product was isolated from spirohydroxylactam **13f** as yellow crystals in 81% yield and melted at 261 °C (ethanol); R_f (AcOEt)=0.30; IR (KBr): ν 3407 (NH), 3030 and 2980 (CH), 1680 (C=O and C=C) cm⁻¹; ¹H NMR (DMSO-*d*₆): δ 1.89 (m, 2H, CH₂), 2.67 (t, 2H, *J*=7 Hz, CH₂), 2.73-2.88 (m, 2H, CH₂), 3.83 (s, 3H, CH₃), 4.97 (s, 2H, CH₂), 6.85 (d, 1H, *J*=2 Hz, H_{pyrrole}), 7.12-7.38 (m, 6H, H_{aro}+ H_{pyrrole}), 10.42 (s broad, 1H, NH); ¹³C NMR (DMSO-*d*₆): δ 21.9 (CH₂), 25.1 (CH₂), 26.1 (CH₂), 33.2 (CH₃), 44.5 (CH₂), 112.2 (C^q=), 113.9 (CH_{pyrrole}), 118.1 (C^q_{pyrrole}), 118.5 (C^q=), 122.3 (CH_{pyrrole}), 125.6 (2x CH_{aro}), 127.0 (CH_{aro}), 128.7 (2x CH_{aro}), 138.3 (C^q_{aro}-C), 146.3 (C^q_{pyrrole}), 153.9 (C=O); MS, *m/z*: 293 M⁺; Anal. Calcd. for C₁₈H₁₉N₃O (293.15): C, 73.69; H, 6.53; N, 14.32. Found: C, 73.56; H, 6.34; N, 14.14.

1,3-Dibenzylidibenzo[*e,g*]benzimidazol-2-one (14g). This product was isolated from spirohydroxylactam **12g** as white crystals in 99% yield and melted at 294 °C (ethanol); R_f (cyclohexane/AcOEt: 3/2)=0.58; IR (KBr): ν 3034 and 2996 (CH), 1683 (C=O and C=C) cm⁻¹; ¹H NMR (CDCl₃): δ 5.70 (s, 4H, 2x CH₂), 7.17-7.46 (m, 14H, H_{aro}), 8.05 (dd, 2H, *J*=1.6 and 8.6 Hz, H_{aro}), 8.63 (dd, 2H, *J*=1.6 and 8.6 Hz, H_{aro}); ¹³C NMR (CDCl₃): δ 47.6 (2x CH₂), 121.3 (2x CH_{aro}), 121.4 (2x C^q=), 123.9 (2x CH_{aro}), 124.8 (2x CH_{aro}), 126.2 (4x CH_{aro}), 127.1 (2x CH_{aro}), 127.6 (2x CH_{aro}), 127.8 (2x C^q_{aro}-C), 129.1 (4x CH_{aro}), 137.0 (4x C^q_{aro}-C), 156.0 (C=O); MS, *m/z*: 414 M⁺; Anal. Calcd. for C₂₉H₂₂N₂O (414.17): C, 84.03; H, 5.35; N, 6.76. Found: C, 83.79; H, 5.16; N, 6.55.

1-Benzylidibenzo[*e,g*]benzimidazol-2(3*H*)-one (15g). This product was isolated from spiroethoxylactam **13g** as yellow crystals in 67% yield and melted at mp>300 °C (ethanol/DMF); (cyclohexane/AcOEt: 2/3)=0.32; IR (KBr): ν 3424 (NH), 3041 and 2987 (CH), 1697 (C=O), 1635 (C=C) cm⁻¹; ¹H NMR (DMSO-*d*₆): δ 5.60 (s, 2H, CH₂), 7.28-7.72 (m, 10H, H_{aro} + NH), 8.05-8.27 (m, 2H, H_{aro}), 8.86 (dd, 2H, *J*=1.6 and 8.6 Hz, H_{aro}); ¹³C NMR (DMSO-*d*₆): δ 45.3 (CH₂), 119.5 (C^q_{aro}-C), 120.3 (C^q_{aro}-C), 120.4 (C^q=), 120.7 (CH_{aro}), 120.9 (CH_{aro}), 121.1 (C^q=), 123.8 (CH_{aro}), 124.2 (2x CH_{aro}), 125.0 (CH_{aro}), 125.9 (2x CH_{aro}), 126.3 (C^q_{aro}-C), 126.6 (C^q_{aro}-C), 127.0 (CH_{aro}), 127.2 (CH_{aro}), 127.3 (CH_{aro}), 128.8 (2x

CH_{aro}), 137.8 (C^q_{aro}-C), 155.2 (C=O); MS, m/z: 324 M⁺; Anal. Calcd. for C₂₂H₁₆N₂O (324.13): C, 81.46; H, 4.97; N, 8.64. Found: C, 81.31; H, 4.78; N, 8.53.

1,3-Dibenzylimidazo[4,5-c]pyrrolo[1,2-a]quinoline-2-one (14h). This product was isolated from spirohydroxylactam **12h** as yellow-green solids in 96% yield and melted at 164 °C (ethanol/diethyl ether); R_f (cyclohexane/AcOEt: 2/3)=0.68; IR (KBr): ν 3026 and 2981 (CH), 1706 (C=O), 1656 (C=C) cm⁻¹; ¹H NMR (CDCl₃): δ 5.50 (s, 2H, CH₂), 5.66 (s, 2H, CH₂), 6.54 (d, 1H, J=5.5 Hz, H_{pyrrole}), 7.29-7.52 (m, 13H, H_{aro} + H_{pyrrole}), 7.72 (dd, 1H, J=5.5 and 2.3 Hz, H_{pyrrole}), 7.94 (d, 1H, J=8.6 Hz, H_{aro}), 8.01 (d, 1H, J=7.8 Hz, H_{aro}); ¹³C NMR (CDCl₃): δ 46.3 (CH₂), 47.3 (CH₂), 102.3 (CH_{pyrrole}), 114.9 (C^q₌), 115.8 (C^q₌), 121.6 (CH_{pyrrole}), 122.3 (CH_{pyrrole}), 126.0 (CH_{aro}), 126.1 (2x CH_{aro}), 126.2 (2x CH_{aro}), 126.6 (CH_{aro}), 128.0 (CH_{aro}), 128.1 (CH_{aro}), 129.2 (3x CH_{aro}), 129.3 (3x CH_{aro}), 131.1 (2x C^q_{aro}), 132.3 (C^q_{aro}), 135.8 (C^q_{aro}), 135.9 (C^q_{aro}), 155.1 (C=O); MS, m/z: 403 M⁺; Anal. Calcd. for C₂₇H₂₁N₃O (403.17): C, 80.37; H, 5.25; N, 10.41. Found: C, 80.21; H, 5.14; N, 10.33.

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