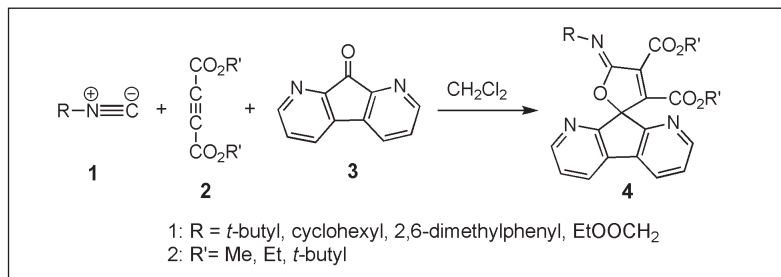


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The 1:1 intermediate generated by the addition of alkyl and aryl isocyanides to dialkyl acetylenedicarboxylate is trapped by 1,8-diazafloren-9-one to yield iminolactones in good yields.

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### Introduction.

Multicomponent reactions (MCRs), defined as one-pot reactions in which at least three functional groups join through covalent bonds, have been steadily gaining importance in synthetic organic chemistry [1,2]. Recently,  $\gamma$ -spiro-lactones have been the subject of great consideration because of their effects as antibacterial agents, aldosterone inhibitors and proper precursors for the preparation of a wide spectrum of natural compounds [3].

Iminolactones have been hydrolyzed with aqueous hydrochloric acid for 2 h producing butenolides, [4] also named 2(*5H*)-furanones, [5] form an important class of natural products that are biologically active compounds and used in medicine and in agriculture [6-10]. The reactivity of nucle-

ophilic carbenes such as isocyanides towards dimethyl acetylenedicarboxylate (DMAD) is well documented [11-14].

The initially formed zwitterionic intermediate has been shown to undergo further reaction with DMAD and isocyanide in different molar proportions, leading to a variety of complex heterocyclic compounds and these reactions have been the subject of detailed investigation by a number of research groups [15-21].

In order to confirm the presence of highly reactive intermediates derived from isocyanides and acetylenic esters which are then likely to undergo addition to 1,8-diazafloren-9-one leading to heterocycles. We initiated an investigation of the reaction of isocyanides and acetylenic esters with 1,8-diazafloren-9-one (see Figure 1).

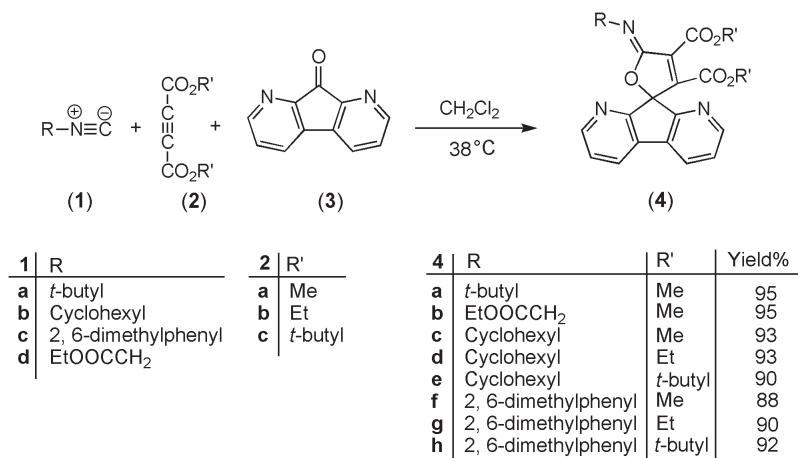


Figure 1

It may be mentioned that attempts to trap such zwitterionic intermediates with olefinic dipolarophiles such as cyclohexene and dimethylfumarate had failed [19]. However, the existence of the 1:1 intermediate is indicated by the isolation of 1:1:1 heterocyclic adducts from the reaction mixture of isocyanide with DMAD in the presence of 1,8-diazafloren-9-one [21c].

### Results and Discussion.

Thus, alkyl and aryl isocyanides **1** and acetylenic esters **2** in the presence of 1,8-diazafloren-9-one **3** undergo a cycloaddition reaction in dichloromethane at 38 °C to produce  $\gamma$ -spiroiminolactons **4** in good yields. On the basis of the well established chemistry of isocyanides [14,22], it is reasonable to assume that compounds **4** result from addition of alkyl and aryl isocyanides to the acetylenic esters and concomitant addition to 1,8-diazafloren-9-one leading to  $\gamma$ -spiroiminolactones (Figure 2).

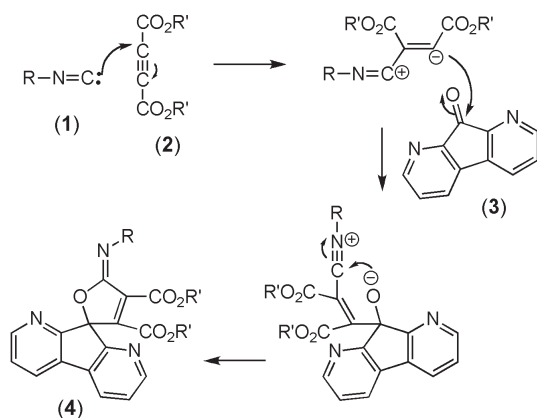


Figure 2

Structures **4** were assigned on the basis of their IR, <sup>1</sup>H and <sup>13</sup>C NMR as well as mass spectral data. The mass spectrum of each compounds displayed the molecular ion peak at appropriate m/z values. The IR spectrum of **4a** showed strong absorptions at 1748 and 1726 cm<sup>-1</sup> due to the ester, 1678 cm<sup>-1</sup> due to C=N of iminolactone ring. The <sup>1</sup>H NMR spectrum of **4a** exhibited one singlet arising from the *tert*-butyl group ( $\delta$  1.26) and two singlets arising from methoxy carbonyl groups ( $\delta$  3.05 and 3.94).

The <sup>13</sup>C NMR spectrum of **4a** showed fifteen distinct resonances consistent with the  $\gamma$ -spiroiminolactone structure (see Experimental section). The characteristic signal due to the spiro carbon was described at  $\delta$  91.66 and the carbon of the C=N group resonated at  $\delta$  158.07. The <sup>1</sup>H and <sup>13</sup>C NMR spectra of **4a-h** are similar to those of **4a**, except for isocyanide, ester residues.

In conclusion, a three-component condensation reaction is required. It offers an easy and effective one-pot synthe-

sis of iminolactones which are potentially amenable to a number of synthetic transformation [23].

### EXPERIMENTAL

Dialkyl acetylenedicarboxylates, 1,8-diazafloren-9-one *tert*-butyl, cyclohexyl, 2,6-dimethylphenyl isocyanides and ethyl isocynoacetate were obtained from Fluka (Buchs, Switzerland) and were used without further purification. Melting points were measured on an Electrothermal 9100 apparatus. Elemental analysis for C, H and N were performed using a Heraeus CHN-O-Rapid analyzer. IR spectra were measured on a Shimadzu IR-470 spectrometer. <sup>1</sup>H and <sup>13</sup>C NMR spectra were measured with a Bruker DRX - 500 AVANCE instrument with CDCl<sub>3</sub> as solvent at 500.1 and 125.7 MHz, respectively. Mass spectra were recorded on a Shimadzu GC/MS QP 1100 EX mass spectrometer operating at an ionization potential of 70 eV.

General Procedure (Exemplified Dimethyl-2'-*tert*-butyliminospiro[1,8-diazafloren-9,5'(1')-oxacyclopant(3')ene]-3',4'-dicarboxylate (**4a**).

A mixture of 1,8-diazafloren-9-one (0.182 g, 1 mmol) and DMAD (0.171 g, 1.2 mmol) was dissolved in 20 mL of dry CH<sub>2</sub>Cl<sub>2</sub> then a mixture of *t*-butyl isocyanide (0.01 g, 1.2 mmol) in 3 mL of CH<sub>2</sub>Cl<sub>2</sub> was added dropwise at room temperature over 3 min. (The isocyanides are toxic compounds but the toxicity of them are less than cyanides, nevertheless the isocyanides take into the lungs by inhalation and contact with skin, therefore this work was carried out inside the polyethylene glove bags under completely air-cleaner condition). Then was allowed to warm up 38 °C for 10 h. After this time the solvent was removed and the product crystals were washed by (275) mL of cold diethyl ether. The melting point was recorded for crude products. This compound was obtained as Pale yellow powder in 95% yield (0.39 g); mp. 263-266 °C; ir (KBr)  $\nu_{\max}$  = 1748 and 1726 (2 C=O), 1678 (C=N) cm<sup>-1</sup>; <sup>1</sup>H nmr (CDCl<sub>3</sub>):  $\delta_{\text{H}}$  1.26 (s, 9H, *CM*<sub>3</sub>), 3.50 and 3.94 (2s, 6H, 2*OMe*), 7.34 (dd, 2H, *J*<sub>1</sub>=7.5, *J*<sub>2</sub>=5.0 Hz, 2CH), 7.95 (d, 2H, *J*=7.5 Hz, 2CH), 8.52 (d, 2H, *J*=5.0 Hz, 2NCH); <sup>13</sup>C nmr (CDCl<sub>3</sub>):  $\delta_{\text{C}}$  =29.63 (*CM*<sub>3</sub>), 52.53 and 52.87 (2*OMe*), 55.13 (N*CM*<sub>3</sub>), 91.66 (C<sub>spiro</sub>), 124.76, 128.40, 133.21, 139.61, 140.93, 149.69 and 152.91 (C=C<sub>iminolactone</sub> and C<sub>arom</sub>), 158.07 (N=C<sub>iminolactone</sub>), 160.53 and 162.34 (2C=O); MS (EI, 70 EV) m/z (%): 409 (M<sup>+</sup>+2, 10), 408 (M<sup>+</sup>+1, 35), 392 (100), 348 (2), 317 (4), 233 (30), 191 (33), 57 (40), 41 (52).

Anal. Calc. for C<sub>22</sub>H<sub>21</sub>N<sub>3</sub>O<sub>5</sub>: C, 64.86; H, 5.16; N, 10.31%. Found C, 64.97; H, 5.08; N, 10.38.

Dimethyl-2'-ethyl acethyliminospiro[1,8-diazafloren-9,5'(1')-oxacyclopant(3')ene]-3',4'-dicarboxylate (**4b**).

This compound was obtained as pale yellow powder in 95% yield (0.41 g); mp. 215-218 °C; ir (KBr)  $\nu_{\max}$  = 1745 and 1723 (3 C=O), 1678 (C=N) cm<sup>-1</sup>; <sup>1</sup>H nmr (CDCl<sub>3</sub>):  $\delta_{\text{H}}$  1.20 (t, 3H, *J*=7.0 Hz, CH<sub>2</sub>Me), 3.50 and 3.95 (2s, 6H, 2*OMe*), 4.11 (q, 2H, *J*=7.0 Hz, CH<sub>2</sub>Me), 4.21 (s, 2H, NCH<sub>2</sub>), 7.35 (dd, 2H, *J*<sub>1</sub>=7.5, *J*<sub>2</sub>= 4.9 Hz, 2CH), 7.96 (d, 2H, *J*=7.5 Hz, 2CH), 8.49 (d, 2H, *J*=4.9 Hz, 2NCH); <sup>13</sup>C nmr (CDCl<sub>3</sub>):  $\delta_{\text{C}}$  13.96 (CH<sub>2</sub>Me), 50.19 (NCH<sub>2</sub>), 52.60 and 53.04 (2*OMe*), 60.71 (OCH<sub>2</sub>Me), 92.03 (C<sub>spiro</sub>), 125.09, 128.61, 133.33, 138.06, 142.58, 149.62 and 159.18 (C=C<sub>iminolactone</sub> and C<sub>arom</sub>), 159.40 (N=C<sub>iminolactone</sub>), 159.78, 161.32 and 169.64 (3C=O); MS (EI, 70 EV) m/z (%): 439 (M<sup>+</sup>+2, 6), 438 (M<sup>+</sup>+1, 5),

365 (60), 364(50), 309 (44), 277 (29), 59 (23).

*Anal.* Calc. for  $C_{22}H_{19}N_3O_7$ : C, 60.41; H, 4.35; N, 9.61%. Found C, 61.03; H, 4.29; N, 9.70.

Dimethyl-2'-cyclohexyliminospiro[1,8-diazafloren-9,5'(1'-oxa)cyclopant(3')ene]-3',4'-dicarboxylate (**4c**).

This compound was obtained as light yellow powder in 93% yield (0.40 g); mp. 283-286 °C; ir (KBr)  $\nu_{\max}$  = 1744 and 1722 (2C=O), 1675 (C=N)  $cm^{-1}$ ;  $^1H$  nmr ( $CDCl_3$ ):  $\delta_H$  1.18-1.80 (m, 10H, 5CH<sub>2</sub>), 3.51 (s, 3H, OMe), 3.59 (m, 1H, N-CH), 3.96 (s, 3H, OMe), 7.36 (dd, 2H, J<sub>1</sub>=7.5, J<sub>2</sub>=5.0 Hz, 2CH), 7.96 (dd, 2H, J<sub>1</sub>=7.5, J<sub>2</sub>=1.0 Hz, 2CH), 8.53 (dd, 2H, J<sub>1</sub>=5.0, J<sub>2</sub>=1.0 Hz, 2NCH);  $^{13}C$  nmr ( $CDCl_3$ ):  $\delta_C$  24.68, 25.66 and 33.10 (5CH<sub>2</sub> of cyclohexyl), 52.50 and 52.91 (2OMe), 56.62 (N-CH), 91.14 (C<sub>spiro</sub>), 124.83, 128.42, 133.25, 139.43, 140.63, 149.66, 154.89 (C=C<sub>iminolactone</sub> and C<sub>arom</sub>), 160.21 (N=C<sub>iminolactone</sub>), 160.31 and 161.98 (2C=O); MS (EI, 70 EV) m/z (%): 435 (M<sup>+</sup>+2, 13), 434 (M<sup>+</sup>+1, 15), 433 (M<sup>+</sup>, 2), 374 (13), 336(100), 308 (42), 305 (71), 277 (51), 59 (19).

*Anal.* Calc. for  $C_{24}H_{23}N_3O_5$ : C, 66.51; H, 5.31; N, 9.69%. Found C, 66.62; H, 5.28; N, 9.74.

Diethyl-2'-cyclohexyliminospiro[1,8-diazafloren-9,5'(1'-oxa)cyclopant(3')ene]-3',4'-dicarboxylate (**4d**).

This compound was obtained as light yellow powder in 93% yield (0.43 g); mp. 247-250 °C; ir (KBr)  $\nu_{\max}$  = 1737 and 1728 (2C=O), 1678 (C=N)  $cm^{-1}$ ;  $^1H$  nmr ( $CDCl_3$ ):  $\delta_H$  0.96 (t, 3H, J=7.0 Hz, CH<sub>2</sub>Me), 1.18-1.80 (m, 10H, 5CH<sub>2</sub>), 1.38 (t, 3H, J=7.0 Hz, CH<sub>2</sub>Me), 3.61 (m, 1H, N-CH), 3.91 (q, 2H, J=7.0 Hz, CH<sub>2</sub>Me), 4.44 (q, 2H, J=7.0 Hz, CH<sub>2</sub>Me), 7.35 (dd, 2H, J<sub>1</sub>=7.5, J<sub>2</sub>=5.0 Hz, 2CH), 7.95 (dd, 2H, J<sub>1</sub>=7.5, J<sub>2</sub>=1.0 Hz, 2CH), 8.54 (dd, 2H, J<sub>1</sub>=5.0, J<sub>2</sub>=1.0 Hz, 2NCH);  $^{13}C$  nmr ( $CDCl_3$ ):  $\delta_C$  13.45 and 14.08 (2CH<sub>2</sub>Me), 24.65, 25.73 and 33.13 (5CH<sub>2</sub> of cyclohexyl), 56.48 (N-CH), 61.44 and 62.08 (2CH<sub>2</sub>Me), 91.09 (C<sub>spiro</sub>), 124.76, 128.31, 133.28, 139.61, 140.56, 149.64 and 154.94 (C=C<sub>iminolactone</sub> and C<sub>arom</sub>), 159.75 (N=C<sub>iminolactone</sub>), 160.56 and 161.58 (2C=O); MS (EI, 70 EV) m/z (%): 462 (M<sup>+</sup>+1, 6), 461 (M<sup>+</sup>, 4), 378 (8), 364 (98), 337 (25), 335 (12), 83 (58), 73 (10), 55 (100), 45 (23), 29 (15).

*Anal.* Calc. for  $C_{26}H_{27}N_3O_5$ : C, 67.68; H, 5.85; N, 9.11%. Found C, 67.62; H, 5.91; N, 8.95.

Di-*tert*-butyl-2'-cyclohexyliminospiro[1,8-diazafloren-9,5'(1'-oxa)cyclopant(3')ene]-3',4'-dicarboxylate (**4e**).

This compound was obtained as pale yellow powder in 90% yield (0.47 g); mp. 191.5-193.5 °C; ir (KBr)  $\nu_{\max}$  = 1732 and 1727 (2C=O), 1679 (C=N)  $cm^{-1}$ ;  $^1H$  nmr ( $CDCl_3$ ):  $\delta_H$  1.03 (s, 9H, CMe<sub>3</sub>), 1.18-1.75 (m, 10H, 5CH<sub>2</sub>), 1.59 (s, 9H, CMe<sub>3</sub>), 3.59 (m, 1H, N-CH), 7.33 (dd, 2H, J<sub>1</sub>=7.5, J<sub>2</sub>=5.0 Hz, 2CH), 7.92 (d, 2H, J=7.5 Hz, 2CH), 8.53 (d, 2H, J=5.0 Hz, 2NCH);  $^{13}C$  nmr ( $CDCl_3$ ):  $\delta_C$  24.10, 25.87 (3CH<sub>2</sub> of cyclohexyl), 27.45 and 28.18 (2CMe<sub>3</sub>), 33.17 (2CH<sub>2</sub> of cyclohexyl), 55.67 (N-CH), 82.54 and 83.28 (2CMe<sub>3</sub>), 90.84 (C<sub>spiro</sub>), 124.53, 128.16, 133.22, 139.23, 141.63, 149.55 and 154.67 (C=C<sub>iminolactone</sub> and C<sub>arom</sub>), 158.84 (N=C<sub>iminolactone</sub>), 160.55 and 161.00 (2C=O); MS (EI, 70 EV) m/z (%): 518 (M<sup>+</sup>+1, 21), 502 (M<sup>+</sup>, 6), 460 (2), 420 (21), 364 (25), 182 (52), 98 (27), 74 (19), 57 (100).

*Anal.* Calc. for  $C_{30}H_{35}N_3O_5$ : C, 69.63; H, 6.77; N, 8.12%. Found C, 69.70; H, 6.81; N, 8.09.

Dimethyl-2'-(2,6-dimethylphenylimino)spiro[1,8-diazafloren-9,5'(1'-oxa)cyclopant(3')ene]-3',4'-dicarboxylate (**4f**).

This compound was obtained as Light brown powder in 88% yield (0.40 g); mp. 272-275 °C; ir (KBr)  $\nu_{\max}$  = 1735 (2C=O), 1689 (C=N)  $cm^{-1}$ ;  $^1H$  nmr ( $CDCl_3$ ):  $\delta_H$  2.23 (s, 6H, Ar-Me<sub>2</sub>), 3.55 and 4.04 (2s, 6H, 2OMe), 6.82 (t, 1H, J=7.5 Hz, Ar-H), 6.93 (d, 2H, J=7.5 Hz, Ar-H), 7.25 (t, 2H, J=5.0 Hz, 2CH), 7.75 (d, 2H, J=7.5 Hz, 2CH), 8.46 (d, 2H, J=5.0 Hz, 2NCH);  $^{13}C$  nmr ( $CDCl_3$ ):  $\delta_C$  18.11 (Ar-Me<sub>2</sub>), 52.73 and 53.18 (2OMe), 91.75 (C<sub>spiro</sub>), 123.64, 124.99, 127.24, 127.84, 128.44, 133.40, 137.69, 142.99, 144.08, 149.52 and 155.46 (C=C<sub>iminolactone</sub> and C<sub>arom</sub>), 159.46 (N=C<sub>iminolactone</sub>), 159.90 and 161.56 (2C=O); MS (EI, 70 EV) m/z (%): 457 (M<sup>+</sup>+2, 13), 456 (M<sup>+</sup>+1, 40), 455 (M<sup>+</sup>, 85), 424 (19), 396 (56), 365 (23), 336 (100), 308 (27), 277 (31), 59 (46).

*Anal.* Calc. for  $C_{26}H_{21}N_3O_5$ : C, 68.57; H, 4.61; N, 9.23%. Found C, 68.62; H, 4.67; N, 9.19.

Diethyl-2'-(2,6-dimethylphenylimino)spiro[1,8-diazafloren-9,5'(1'-oxa)cyclopant(3')ene]-3',4'-dicarboxylate (**4g**).

This compound was obtained as Light yellow powder in 90% yield (0.43 g); mp. 265-268 °C; ir (KBr)  $\nu_{\max}$  = 1727 (2C=O), 1690 (C=N);  $cm^{-1}$ ;  $^1H$  nmr ( $CDCl_3$ ):  $\delta_H$  0.97 (t, 3H, J=7.0 Hz, CH<sub>2</sub>Me), 1.45 (t, 3H, J=7.0 Hz, CH<sub>2</sub>Me), 2.25 (s, 6H, Ar-Me<sub>2</sub>), 3.95 (q, 2H, J=7.0 Hz, CH<sub>2</sub>Me), 4.52 (q, 2H, J=7.0 Hz, CH<sub>2</sub>Me), 6.82 (t, 1H, J=7.5 Hz, Ar-H), 6.94 (d, 2H, J=7.5 Hz, Ar-H), 7.18 (dd, 2H, J<sub>1</sub>=7.0, J<sub>2</sub>=5.0 Hz, 2CH), 7.61 (d, 2H, J=7.0 Hz, 2CH), 8.43 (d, 2H, J=5.0 Hz, 2NCH);  $^{13}C$  nmr ( $CDCl_3$ ):  $\delta_C$  13.42 and 14.14 (2CH<sub>2</sub>Me), 18.11 (Ar-Me<sub>2</sub>), 61.71 and 62.41 (2CH<sub>2</sub>Me), 91.93 (C<sub>spiro</sub>), 123.57, 124.88, 127.25, 127.84, 128.34, 133.32, 137.80, 142.91, 144.28, 149.38 and 155.63 (C=C<sub>iminolactone</sub> and C<sub>arom</sub>), 159.33 (N=C<sub>iminolactone</sub>), 159.54 and 161.13 (2C=O); MS (EI, 70 EV) m/z (%): 484 (M<sup>+</sup>+1, 42), 483 (M<sup>+</sup>, 63), 438 (17), 410 (35), 364 (100), 336 (42).

*Anal.* Calc. for  $C_{28}H_{25}N_3O_5$ : C, 69.56; H, 5.17; N, 8.69%. Found C, 68.85; H, 5.21; N, 8.62.

Di-*tert*-butyl-2'-(2,6-dimethylphenylimino)spiro[1,8-diazafloren-9,5'(1'-oxa)cyclopant(3')ene]-3',4'-dicarboxylate (**4h**).

This compound was obtained as Light brown powder in 92% (0.49 g); mp. 209.3-212 °C; yield 92%; ir (KBr)  $\nu_{\max}$  = 1724 (2C=O), 1683 (C=N);  $cm^{-1}$ ;  $^1H$  nmr ( $CDCl_3$ ):  $\delta_H$  0.98 and 1.63 (2s, 18H, 2OCMe<sub>3</sub>), 2.24 (s, 6H, Ar-Me<sub>2</sub>), 6.76 (t, 1H, J=7.5 Hz, Ar-H), 6.92 (d, 2H, J=7.5 Hz, Ar-H), 7.32 (dd, 2H, J<sub>1</sub>=8.0, J<sub>2</sub>=5.0 Hz, 2CH), 7.77 (d, 2H, J=8.0 Hz, 2CH), 8.36 (d, 2H, J=5.0 Hz, 2NCH);  $^{13}C$  nmr ( $CDCl_3$ ):  $\delta_C$  18.10 (Ar-Me<sub>2</sub>), 27.28 and 28.12 (2CMe<sub>3</sub>), 82.95 and 83.75 (2CMe<sub>3</sub>), 91.47 (C<sub>spiro</sub>), 123.20, 124.62, 127.17, 127.80, 128.20, 133.11, 137.75, 144.47, 144.80, 149.14 and 155.66 (C=C<sub>iminolactone</sub> and C<sub>arom</sub>), 158.32 (N=C<sub>iminolactone</sub>), 159.77 and 159.86 (2C=O); MS (EI, 70 EV) m/z (%): 539 (M<sup>+</sup>, 35), 482 (4), 438 (7), 182 (100), 101 (24), 57 (17).

*Anal.* Calc. for  $C_{32}H_{33}N_3O_5$ : C, 71.24; H, 6.12; N, 7.79%. Found C, 71.81; H, 6.17; N, 7.81.

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