[4 + 2]-Cycloaddition Reactions between β-Acceptor-Substituted Enamines and 2-Vinylindole Radical Cations Acting as Hetero-Dienes

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[4 + 2]-Cycloaddition reactions between 2-vinylindoles acting as hetero-dienes and β-acceptor substituted cyclic and acyclic enamines can be induced by formation of 2-vinylindole radical cations either via anodic oxidation or photoelectron transfer (PET) using catalytic amounts of triarylpyrylium tetrafluoroborates as sensitizers. In this way, pyrido[1,2-*a*]indoles or indolo[1,2-*a*]hexahydro-1,8-naphthyridines are formed in one step with complete regio- and stereochemical control.

Introduction

The use of electron transfer as a simple means to a redox umpolung has been shown to be applicable to a variety of reactions, e.g., radical cation dimerizations, fragmentation[1] reactions, rearrangements,[2] and such important reactions as [2 + 2]- and [4 + 2]-cycloadditions.[3] During the last 15 years, radical cation Diels−Alder (DA) reactions have been the subject of many mechanistic and theoretical investigations. Typically, these reactions display an increase in the reaction rate of several orders of magnitude over that of the neutral reaction due to a very low activation barrier, usually coupled with high regio- and chemoselectivity. Therefore, such reactions are promising tools for organic synthesis. Until recently, their application was restricted to non-heterosubstituted dienes or dienophiles, e.g., to the well-studied dimerization of cyclohexadienes,[4] dimerization and cycloadditions of substituted 1,3-pentadienes,[5] or reactions of cyclohexadienes and styrenes.[6] Radical cation Diels−Alder reactions of heterosubstituted dienes or dienophiles were introduced with [2 + 2] cycloadditions,[7] which lead to the corresponding cyclobutane derivatives, and subsequent rearrangement[8] by strong bases such as KH in THF, which lead to the formal [4 + 2] cycloaddition products. Direct [4 + 2] cycloadditions between butadiene-type dienophiles and phenyl vinyl thioether were later accomplished as part of the synthesis of (−)-β-selinen.[9] The potential of these cyclizations has been exploited with reactions between substituted allenes[10] and ketenes[11] and pentamethylcyclopentadiene which resulted in terpenoid skeletons.

We have demonstrated the advantages of radical cation cyclizations in reactions with indole and several substituted cyclohexadienes. Using this reaction, we were able to react the aromatic dienophile, indole, under mild reaction conditions and photoelectron transfer catalysis, and after trapping the product with acetyl chloride, obtained the highly substituted *N*-acyl-1,4,4a,9a-tetrahydro-1,4-ethanocarbazoles in one step.[12] Computational investigations on a semiempirical level revealed that the indole radical cation, having radical character primarily at position 3 of the indole nucleus and the positive charge stabilized in the form of an iminium ion, reacts by a radical attack of the 3-position on the 1,3-diene leading to a long-bond distonic radical cation intermediate[13] which finally cyclizes. We extended this methodology to reactions of indole with exocyclic dienes, thus opening up a highly efficient route to linearly annellated dihydrocarbazole derivatives.[14]

Recently, we investigated radical cation DA reactions between acceptor-substituted 2-vinylindoles and several cyclohexadienes and styrenes.[15]

In conventional DA reactions the electron-rich vinylindole reacts only with very electron-poor dienophiles with catalysis by Broensted[16] or Lewis acids, often requiring high temperatures and long reaction times.

We report here[17] a formal hetero DA reaction in which vinylindole, as the electron-rich hetero-diene, undergoes a cycloaddition with equally electron-rich dienophiles (in terms of conventional DA reactions) of the β-acceptor-substituted enamine type under the conditions of an electrochemically or photochemically induced electron

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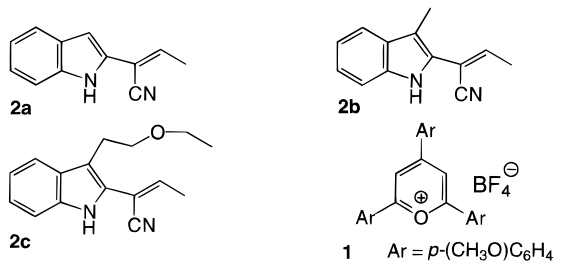
transfer reaction.[18] The enamines may be cyclic or acyclic and should bear a radical-stabilizing acceptor substituent at the enamine double bond. As we reported earlier,[19] anodic oxidation[20] or photoexcited triarylpyrylium tetrafluoroborate[21] (**1**) were used for the induction of the reaction.

Results and Discussion

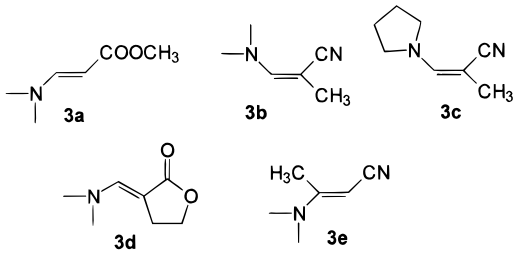
Vinylindoles **2a**−**c** are easily accessible by a domino process developed by our group[22] starting from simple materials like phenylhydroxylamine, cyanoallene, and a suitable aldehyde component. This process tolerates diverse substituents at position 3 of the indole[23] and at the vinyl group.[24]

Chart 1



These vinyl acrylonitriles (Chart 1) are rather electron-rich dienes in terms of Diels−Alder reactions. This can be established by their oxidation potentials which range from 690 to 790 mV (*vs* $Ag/AgNO_3$). Either anodic oxidation (method E) or reaction with the photochemically excited triarylpyrylium tetrafluoroborate (**1**) (method P) leads to a radical cation of the diene which may subsequently undergo cyclization reactions with a variety of enamines. The latter are easily accessible by a Michael addition of the corresponding dimethylamino compound to conjugated triple bond (e.g., **3a**, **3e**), treatment of a lactone (e.g., γ-butyrolactone) with Brederecks reagent[25] (**3d**), or nucleophilic substitution followed by elimination of β-halo methacrylonitriles[26] (**3b**, **3c**) (Chart 2).

Chart 2



The oxidation potentials of the enamines employed range from 690 to 800 mV (*vs* $Ag/AgNO_3$). Thus, the reaction takes place between two compounds of similar HOMO energies *via* the redox umpolung of the π-component to its radical cation. The reaction is not sensitive to steric effects caused by substituents at position 3 of the the indole nucleus. In examples **5a**−**c** (Table 1), subsequent aromatization of the initially formed dihydropyrido[1,2-*a*]indole is slow in the case of sterically hindered **5c**, compared to the reactions leading to **5a**/**5b**, and takes several hours. With photochemical initiation of the reaction, aromatization with loss of the dimethylamino functionality occurs almost immediately, which is in contrast to the reactions resulting from electrochemical initiation (see the Experimental Section). If the polymerization tendency of the 2-vinylindole on the electrode surface is sufficiently low (e.g., 2-vinylindoles **2b**, **2c**), an electrochemical initiation of the reaction under potentiostatic control is very favorable. Generally, yields are higher with this method than with the photochemical initiation.

If the β-position of the enamine is substituted, aromatization of the product is not possible (e.g., **6b**, **7b**, **8b**). Considerable steric hinderance in the case of the enamino lactone **3d** does not impede the reaction. In all cases product formation is achieved with total regio- and stereochemical control, giving a product of a higher oxidation level. Thus, the transformation is induced by an electron transfer and includes another oxidation step. The stereochemistry of the compounds shown in Table 1 was confirmed by NOE experiments.

Table 1. Reactions of 2-Vinylindoles with Acyclic Enamido Esters and Nitriles

Diene	Dienophile	Method	Product	Yield
2a	**3a**	SET, $-2H^+$, $-2e^-$, $-N(CH_3)_2H$, P	**5a**	22% + **14a**
2b	**3a**	E	**5b**	32%
2c	**3a**	E	**5c**	50%
2b	**3b**	SET, $-2H^+$, $-2e^-$, E	**6b**	50%
2b	**3c**	E	**7b**	33%
2b	**3d**	E	**8b**	52%

(18) Gürtler, C. F.; Blechert, S.; Steckhan, E. *Synlett* **1994**, 141.
(19) Gieseler, A.; Steckhan, E.; Wiest, O.; Knoch, F. *J. Org. Chem.* **1991**, *56*, 1405.
(20) Mlcoch, J.; Steckhan, E. *Angew. Chem.* **1985**, *97*, 429; *Angew. Chem., Int. Ed. Engl.* **1985**, *24*, 412.
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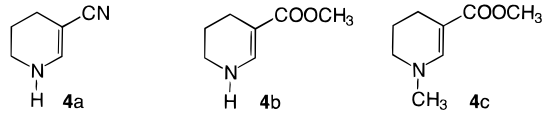
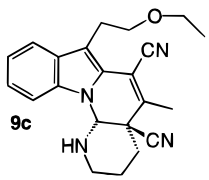
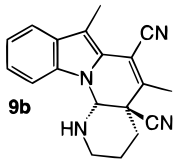
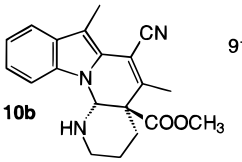
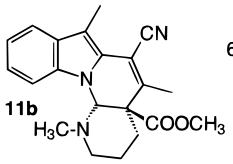
The application of β-substituted enamines can be extended to tetrahydropyridines **4a**−**4c** (Chart 3), which were synthesized from the corresponding substituted pyridines.[27]

Chart 3



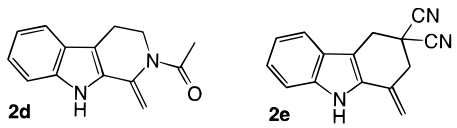
The products formed by reactions with **4a**−**4e** are especially interesting as they incorporate the skeleton of the indole alkaloid Goniomitine. Formation of **9b**, **9c**, **10b**, and **11b** (*cis*-fused rings) is highly stereoselective, with yields ranging from 53% (**9c**) to 90% (**10b**) (Table 2).

Table 2. Reaction of 2-Vinylindoles with 1,4,5,6-Tetrahydropyridines

Diene	Dienophile	Method	Product	Yield
2c	**4a**	SET, -2H+, -2e-, E	 **9c**	53%
2b	**4a**	E	 **9b**	68%
2b	**4b**	E	 **10b**	91%
2b	**4c**	E	 **11b**	62%

The use of 2-vinylindoles is not restricted to those bearing a cyano functionality on the vinylic group. We were able to react 2-vinylindoles containing an amide functionality or no acceptor substituent at all with β-substituted enamines. Compound **2d** (Chart 4) can be obtained from an imine−enamine rearrangement of the easily accessible Harmalane derivative using triethylamine as a base and acetyl chloride as the trapping reagent.[28]

Chart 4



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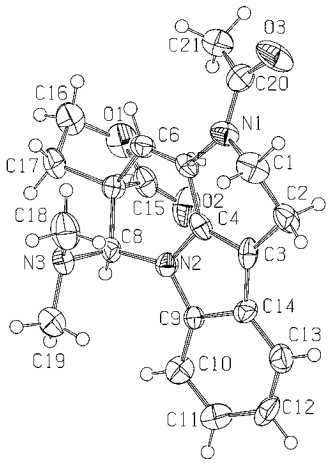
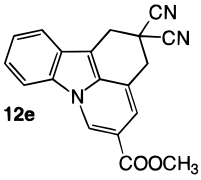
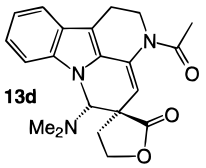


Figure 1. ORTEP structure of **13d**.

Table 3. Reactions of 2-Vinylindoles 2d and 2e with β-Acceptor-Substituted Enamines

Diene	Dienophile	Method	Product	Yield
2e	**3a**	SET, -2H+, -2e-, -(Me)2NH, E	 **12e**	11%
2d	**3d**	SET, -2H+, -2e-, E	 **13d**	39%

Yields of cycloaddition products from **2d** or **2e**[29] reach 39% (**13d**) and thus are lower than those from reactions with 2-vinylindoles substituted by a cyano functionality acceptor group (Table 3). Interestingly, we were not able to observe NMR signals for those carbons adjacent to the amide functionality, even in deuterated benzene heated slightly below its boiling point. Therefore, an X-ray structure determination of the spirolactone compound **13d** was performed, which established the constitution and the relative stereochemistry of the lactone moiety with respect to the dimethylamino group (Figure 1).

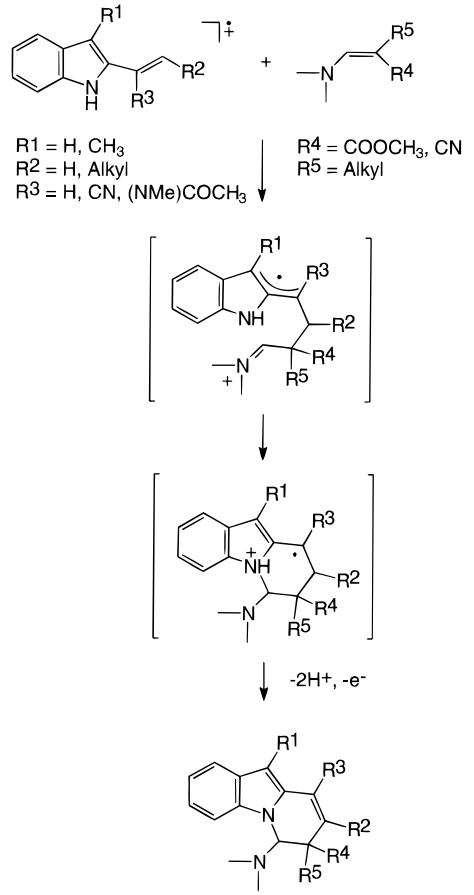
On the basis of our results with 2-vinylindoles, we propose a mechanism (Scheme 1) that starts with a nucleophilic addition of the enamine to the vinylogous iminium radical cation of the 2-vinylindole. There follows an electrophilic cyclization to the newly formed iminium ion. Stabilization of the intermediate radical by radical-stabilizing substituents like the cyano group obviously results in higher yields. Finally, deprotonation and a single electron oxidation followed by further deprotonation leads to the observed products only, except for compounds **5a** and **14a**, which both derive from diene **2a**.

A nonconcerted mechanism[3a] is expected on the basis of previous mechanistic investigations[3a] with radical-

(29) Hogan, I.; Jenkins, P. D.; Sainsbury, M. *Tetrahedron* **1990**, *46*, 2943.

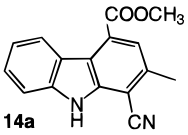
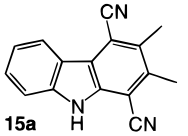
cation Diels–Alder reactions. At the stage of the initially formed radical-cation adduct, rotation around the C–C single bond should be possible. Thus, formation of *cis*/*trans*-diastereomers could be expected. This should be most likely when sterically very demanding substrates like **3d**, compared to rather small dienophiles like **3b**, are used. Interestingly, only one product (**8b**) is observed. We therefore assume that the final ring formation is very fast compared to the rotation around the C–C bond.

Scheme 1

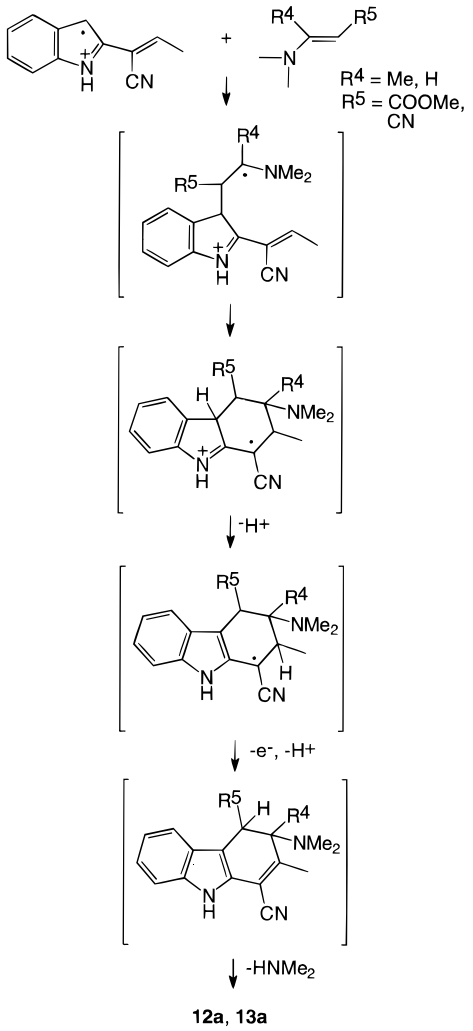


In addition to the mechanism outlined above, another reaction pathway is observed if vinylindole **2a** is reacted under PET conditions (method P). In reactions of **2a** with dienophiles **3a** and **3e** (Table 4), we found substituted carbazoles as reaction products, which may be formed according to Scheme 2.

Table 4. Reactions of 2-Vinylindole 2a under Photoelectron Transfer Inititiation

Diene	Dienophile	Method	Product	Yield
2a	+ **3a**	SET, -2H+, -2e-, -N(CH3)2H, P	 **14a**	74% (GC) + **5a**
2a	+ **3e**	P	 **15a**	13%

Scheme 2



As in the reactions with styrenes or cyclohexadienes,[15] the reaction starts with a radical attack of position 3 of the electrophilic radical cation on the β-acceptor-substituted enamine bond. The resulting radical adds to the acrylonitrile function leading to the cyclized radical cation. After a [1,3]-H-shift, the resulting indole radical cation is deprotonated at the side chain (benzylic position) to give the cyano-substituted radical. Further oxidation followed by deprotonation and dimethylamine elimination yields the highly substituted carbazole derivatives **14a** and **15a**. It is noteworthy that formation of products **5a** and **14a** takes place at the same time. Due to the unexpected instability of **14a**, its yield could only be determined by GC. The ratio of the products depends on the concentration of diene/dienophile and the amount of photosensitizer **1**. At high concentrations, formation of the carbazole is markedly preferred, while at high dilution the pyrido[1,2-*a*]-indole is formed in equal amounts with the carbazole.[18] When the β-position of the β-enamino nitrile (group R^4 in Scheme 2) is sterically hindered, we observed non-cyclized 2,3-divinylindole products. In these cases, the radical cyclization of the first radical cation adduct does not take place. Instead, further one-electron oxidation followed by deprotonation may lead to the product.[30]

(30) Gürtler, C. F.; Steckhan, E.; Blechert, S. unpublished results.

Experimental Section

General Methods. Proton magnetic spectra (^{1}H) were recorded at 200 or 400 MHz. ^{13}C NMR spectra were recorded at 100.6 or 50.3 MHz using the solvent resonance as reference. Melting points are uncorrected. Thin-layer chromatography was performed on Merck $^{60}F_{254}$ (0.2 mm) sheets which were visualized with ethanolic molybdophosphoric acid, UV light, or a solution of *p*-methoxybenzaldehyde in ethanol/sulfuric acid. Preparative flash chromatography was performed on Merck (0.04−0.063 mm) silica gel using a positive pressure of air. Unless otherwise noted, all chemicals were of the highest commercially available purity and were used without further purification. Melting points are uncorrected. Solvents used for chromatography were distilled before use. Photolyses were performed with a light source system consisting of a Hanovia 976C1010 1000-W xenon arc lamp, a Müller-Elektronik LAX 1000 lamp housing, and a Schott WG345 long-pass filter. The system was designed for use with wavelengths (λ) greater than 345 nm.

General Procedure for Photolysis P. The diene, 0.7 mmol, 1 mmol of dienophile, and 5 mol % (based on dienophile) of TAP (tris(*p*-methoxyphenyl)pyrylium tetrafluoroborate) (**1**) were dissolved in 100 mL of anhydrous dichloromethane in a 100 mL Schlenk tube. Vigorous stirring is required. Before the reaction, the mixture was purged for 10 min with argon to free it from oxygen. Then the mixture was irradiated under argon with $\lambda > 345$ nm. After the reaction was finished, the catalyst could easily be separated over a short silica gel column. Isolation of the products was achieved by flash chromatography or HPLC as stated.

General Procedure E. The reaction solution was potentiostatically electrolyzed at the potentials given below in an electrochemical cell (undivided) (maximum volume 100 mL) under vigorous stirring. Key: anode, carbon, 20 cm^2; cathode, carbon, 20 cm^2; reference electrode, Ag/$AgNO_3$; separation of electrodes was 3 cm. Electrolysis started with a potential of 480 mV *vs* Ag/$AgNO_3$ (exception was **2d** with 380 mV). Prior to use, the electrodes were activated by ultrasonic treatment in acetonitrile. In the course of the electrolysis (typically 200 min) the current dropped from 20 to 2 mA. The reaction solutions darkened slightly. At the end of the reaction the electrodes were treated for 10 min with ultrasound in 60 mL of methanol, followed by acetonitrile. The combined solutions were evaporated under reduced pressure and then treated with brine. The aqueous layer was extracted five times with chloroform and dried over $MgSO_4$ and the solvent evaporated. Separation was effected by chromatography on silica gel unless otherwise stated. With the exception of **5a**, **5b**, **10b**, **12e**, and **15a**, yields are based on recovered material (90% turnover).

Mass spectrometry, ^{1}H and ^{13}C NMR spectroscopy, and X-ray crystallography were employed for structure elucidation. The determination of configuration and regiochemistry could be accomplished by NMR methods, especially 2D techniques (CH-COSY, COLOC-experiments), and NOE experiments.

7-Carbomethoxy-9-cyano-8-methylpyrido[1,2-*a*]indole (5a). Following the general procedure P, 0.33 mmol (60 mg) of 2-vinylindole **2a**, 0.57 mmol (74 mg) of methyl 3-(*N*,*N*-dimethyamino)acrylate (**3a**) dissolved in 100 mL of CH_2Cl_2, and 15 mg of TAP were irradiated for 5.5 h with a 1000 W Xenon arc lamp. Separation of the completely aromatized crude product and purification by flash chromatography with toluene/ethyl acetate (20:1) afforded 22 mg (22%) of **5a** as a red solid: mp 157 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.24 (s, 1H, H-6), 7.95 (d, J = 8.0 Hz, 1H, H-4), 7.85 (d, J = 8.0 Hz, 1H, H-1), 7.49 (dd, J = 8.0, 8.0 Hz, 1H, H-2), 7.41 (dd, J = 8.0, 8.0 Hz, 1H, H-3), 6.86 (s, 1H, H-10), 3.97 (s, 3H, 7-$COOCH_3$), 2.89 (s, 3H, 8-CH_3). ^{13}C NMR (100.6 MHz, $CDCl_3$) δ 165.0 (7-COOR), 142.4 (C-8), 134.1 (C-6), 132.7 (C-10a), 130.7 (C-4a), 130.6 (C-9a), 125.3 (C-2), 121.9 (C-3), 121.3 (C-1), 115.7 (9-CN), 110.6 (C-4), 110.5 (C-7), 102.4 (C-9), 94.4 (C-10), 52.4 (7-OCH_3), 20.4 (8-CH_3); high-resolution MS calcd for $C_{16}H_{12}N_2O_2$ (M^+) 264.0898, found 264.0898. In addition to **5a** product **14a** is formed. Yield is optimized for product **5a** (for **14a** see below).

7-Carbomethoxy-9-cyano-1,8-dimethylpyrido[1,2-*a*]indole (5b). Following the general procedure E, 0.3 mmol (60 mg) of 2-vinylindole **2b**, 0.4 mmol (51 mg) of methyl 3-(*N*,*N*-dimethylamino)acrylate (**3a**), dissolved in 60 mL of CH_3CN/CH_2Cl_2 (1:1) (+ 0.1 mol/L $LiClO_4$) were electrolyzed for 150 min while being monitored by TLC analysis. A deep red color caused by aromatization of the dihydropyrido[1,2-*a*]indole occured several minutes after the end of the electrolysis. Purification of the crude product afforded 27 mg (32%) of **5b** as a red solid: mp 108 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.14 (s, 1H, H-6), 7.91 (d, 1H, J = 8.0 Hz, H-4), 7.75 (d, 1H, J = 8.0, 8.0 Hz, H-1), 7.53 (dd, 1H, J = 8.0, 8.0 Hz, H-2), 7.41 (dd, 1H, J = 8.0, 8.0 Hz, H-3), 3.95 (s, 3H, 7-$COOCH_3$), 2.91 (s, 3H, 8-CH_3), 2.81 (s, 3H, 10-CH_3); ^{13}C NMR (100.6 MHz, $CDCl_3$) δ 165.1 (7-COOR), 142.8 (C-8), 134.6 (C-6), 131.7 (C-10a), 129.4 (C-4a), 126.8 (C-9a), 124.9 (C-2), 122.0 (C-3), 119.3 (C-1), 116.6 (9-CN), 110.3 (C-4), 109.4 (C-7), 103.3 (C-10), 101.4 (C-9), 52.1 (C-12), 19.9 (8-CH_3), 8.3 (10-CH_3); high-resolution MS calcd for $C_{17}H_{14}N_2O_2$ (M^+) 278.1044, found 278.1055

7-Carbomethoxy-9-cyano-10-(2-ethoxyethyl)-8-methylpyrido[1,2-*a*]indole (5c). Following the general procedure E, 0.035 mmol (9 mg) of 2-vinylindole **2c** and 0.1 mmol (13 mg) of methyl 3-(*N*,*N*-dimethylamino)-acrylate (**3a**) in 40 mL of CH_3CN/CH_2Cl_2 (1:1) (+ 0.1 mol/L $LiClO_4$) were electrolyzed for 70 min (current consumption 37 °C) while being monitored by TLC analysis. A deep red color caused by aromatization of the dihydropyrido[1,2-*a*]indole occured several hours after the end of the electrolysis. Attempts to isolate the dihydropyrido[1,2-*a*]indole on silica gel led to rapid aromatization. Separation of the reaction mixture by preparative TLC (CH_2Cl_2) afforded 6 mg (50%) of **5c** as a red solid: mp 134 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.20 (s, 1H, H-6), 7.92 (d, 1H, J = 8.0 Hz, H-4), 7.88 (d, 1H, J = 8.0 Hz, H-1), 7.52 (dd, 1H, J = 8.0, 8.0 Hz, H-2), 7.48 (dd, 1H, J = 8.0, 8.0 Hz, H-3), 3.95 (s, 3H, 7-$COOCH_3$), 3.85 (t, 2H, J = 7.5 Hz, H-12), 3.58 (t, 2H, J = 7.5 Hz, H-11), 3.56 (q, 2H, J = 7.5 Hz, H-13), 2.86 (s, 3H, 8-CH_3), 1.19 (t, 3H, J = 7.5 Hz, H-14); ^{13}C NMR (100.6 MHz, $CDCl_3$) δ 165.1 (7-COOR), 143.3 (C-8), 134.5 (C-6), 131.6 (C-10a), 129.6 (C-4a), 125.0 (C-2), 123.5 (c-9a), 122.1 (C-3), 119.7 (C-1), 116.1 (9-CN), 110.3 (C-4), 109.7 (C-7), 105.0 (C-10), 101.9 (C-9), 70.9 (C-16), 66.3 (C-17), 52.2 (7-OCH_3), 24.1 (C-15), 19.9 (8-CH_3), 15.3 (C-14); high-resolution MS calcd for $C_{20}H_{20}N_2O_3$ (M^+) 336.1743, found 336.1743.

6-(Dimethylamino)-7,9-dicyano-7,8,10-trimethyl-6,7-dihydropyrido[1,2-*a*]indole (6b). Following the general procedure E, 0.092 mmol (18 mg) of 2-vinylindole **2b** and 0.32 mmol (35 mg) of 3-(*N*,*N*-dimethylamino)-

methacrylonitrile (**3b**) in 50 mL of CH_3CN (+ 0.1 mol/l $LiClO_4$) were electrolyzed for 150 min (current consumption 93 °C) and monitored by TLC analysis. Separation of the reaction mixture by flash chromatography (methyl *tert*-butyl ether/PE) afforded 14 mg (50%) of **6b** as a white solid: mp 121 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.60 (d, 1H, J = 8.0 Hz, H-1), 7.35 (d, 1H, J = 8.0 Hz, H-4), 7.29 (dd, 1H, J = 8.0, 8.0 Hz, H-3), 7.17 (dd. 1H, J = 8.0, 8.0 Hz, H-2), 5.23 (s, 1H, H-6), 2.60 (s, 3H, 10-CH_3), 2.53 (s, 3H, 8-CH_3), 2.33 (s, 6H, 6-N(CH_3)$_2$), 1.52 (s, 3H, 7-CH_3); ^{13}C NMR (100.6 MHz, $CDCl_3$) δ 144.6 (C-8), 137.0 (c-4a), 129.2 (C-10a), 124.3 (C-3), 123.8 (C-10), 120.6 (C-2), 119.8 (C-1), 119.0 (9-CN), 115.0 (7-CN), 111.4 (C-9a), 109.7 (C-4), 103.9 (C-9), 75.6 (C-6), 46.4 (C-7), 42.3 (6-N(CH_3)$_2$), 24.6 (7-CH_3), 19.7 (8-CH_3), 8.6 (10-CH_3); high-resolution MS calcd for $C_{19}H_{20}N_4$ (M^+) 304.1688, found 304.1688.

6-(1-Pyrrolidinyl)-7,8,10-trimethyl-7,9-dicyano-6,7-dihydropyrido[1,2-*a*]indole (7b). Following the general procedure E, 0.087 mmol (17 mg) of 2-vinylindole **2b** and 0.27 mmol (37 mg) of 3-(pyrrolidinyl)methacrylonitrile (**3c**) in 30 mL of CH_3CN (+ 0.1 mol/L $LiClO_4$) were electrolyzed for 35 min and monitored by TLC analysis. **3c** was a mixture of *E*/*Z*-isomers (ratio 1:2). This leads to the same ratio (1:2) of the *trans*/*cis*-diastereomers of compound **7b**. Separation by HPLC (Nucleosil RP-18, MeOH/H_2O) (8:2) afforded a total yield of 9 mg (33%) of **7b** as a mixture of two diastereomers: yield (*trans*-**7b**), 3 mg (11%); mp 85 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.58 (d, 1H J = 8.0 Hz, H-1), 7.35 (dd, 1H, J = 8.0, 8.0 Hz, H-3), 7.26 (d, 1H, J = 8.0 Hz, H-4), 7.15 (dd, 1H, J = 8.0, 8.0 Hz, H-2), 5.60 (s, 1H, H-6), 3.32 (m, 2H, H-11), 2.62 (s, 3H, 10-CH_3), 2.56 (m, 1H, H-11′), 2.46 (m, 1H, H-11), 2.36 (s, 3H, 8-CH_3), 2.04 (m, 2H, H-12), 1.76 (s, 3H, 7-CH_3), 1.56 (m, 2H, H-12′); ^{13}C NMR (100.6 MHz, $CDCl_3$) δ 146.0 (C-8), 137.1 (C-4a), 129.1 (C-10a), 124.9 (C-10), 124.1 (C-3), 120.3 (C-2), 120.0 (9-CN), 119.7 (C-1), 115.1 (7-CN), 111.6 (C-9a), 109.6 (C-4), 107.0 (C-9), 71.6 (C-6), 49,6 (C-11′), 41.5 (C-11), 24.1 (C-12′), 23.3 (C-12), 20.1 (7-CH_3), 17.4 (8-CH_3), 8.6 (10-CH_3); high-resolution MS calcd for $C_{21}H_{22}N_4$ (M^+) 330.1844, found 330.1844.

***cis*-6-(1-Pyrrolidinyl)-7,8,10-trimethyl-7,9-dicyano-6,7-dihydropyrido[1,2-*a*]indole (*cis*-7b)**: yield 6 mg (22%); mp 196 °C; ^{1}H-NMR (400 MHz, $CDCl_3$) δ 7.59 (d, 1H, J = 8.0 Hz, H-1), 7.35 (d, 1H, J = 8.0 Hz, H-4), 7.27 (dd, 1H, J = 8.0, 8.0 Hz, H-3), 7.15 (dd, J = 8.0, 8.0 Hz, H-2), 5.48 (s, 1H, H-6), 2.62 (m, 4H, H-11/11′), 2.60 (s, 3H, 10-CH_3), 2.52 (s, 3H, 8-CH_3), 1.64 (m, 4H, H-12/12′), 1.46 (s, 3H, 7-CH_3); ^{13}C NMR (100.6 MHz, $CDCl_3$) δ 144.6 (C-8), 137.0 (C-4a), 129.0 (C-10a), 124.0 (C-3), 123.9 (C-10), 120.5 (C-2), 119.5 (C-1), 119.0 (9-CN), 115.1 (7-CN), 110.9 (C-9a), 109.4 (C-4), 104.0 (C-9), 71.5 (C-6), 49.5 (C-11/11′), 46.7 (C-7), 24.1 (7-CH_3), 23.4 (C-12/12′), 19.6 (8-CH_3), 8.6 (10-CH_3); high-resolution MS calcd for $C_{21}H_{22}N_4$ (M^+) 330.1844, found 330.1844.

Spiro[γ-butyrolactone-2,7′-6-(dimethylamino)-8,-10-dimethyl-9-cyanopyrido[1,2-*a*]indole] (8b). Following the general procedure E, 0.046 mmol (9 mg) of 2-vinylindole **2b** and 0.21 mmol (30 mg) of enamino lactone **3d** in 40 mL of CH_3CN/CH_2Cl_2 (3:1) (+0.1 mol/L $LiClO_4$) were electrolyzed for 120 min (current consuption 64 °C) while being monitored by TLC analysis. Separation of the reaction mixture by flash chromatography (MTBE/PE) (1:1) afforded 8 mg (52%) of **8b** as a white solid: mp 185 °C (crystallized from MTBE); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.56 (d, 1H, J = 8.0 Hz, H-1), 7.29 (d, 1H, J = 8.0 Hz, H-4), 7.21 (dd, 1H, J = 8.0, 8.0 Hz, H-3), 7.11 (dd, 1H, J = 8.0, 8.0 Hz, H- 2), 5.20 (s, 1H, H-6), 4.46 (dd, 2H, J = 5.5, 8.0 Hz, H-13), 3.01 (dt, 1H, J = 5.5, 13.5 Hz, H-14), 2.76 (dt, 1H, J = 8.0, 13.5 Hz, H-14′), 2.63 (s, 3H, 8-CH_3), 2.32 (s, 3H, 10-CH_3), 2.25 (s, 6H, 6-N(CH_3)$_2$); ^{13}C NMR (100.6 MHz, $CDCl_3$) δ 173.3 (C-11), 142.1 (C-8), 137.3 (C-4a), 129.3 (C-10a), 125.4 (C-10), 123.6 (C-3), 120.1 (C-2), 119.7 (C-1), 115.9 (9-CN), 110.7 (C-9a), 109.9 (C-4), 107.3 (C-9), 72.3 (C-6), 65.7 (C-13), 53.0 (C-7), 42.4 (6-N(CH_3)$_2$), 29.3 (C-14), 18.9 (8-CH_3), 8.7 (10-CH_3); HRMS calcd for $C_{20}H_{21}N_3O_2$ (M^+) 335.1633, found 335.1633.

4a,6-Dicyano-5,7-dimethylindolo[1,2-*a*]-1,2,3,4,4a,-12a-hexahydro-1,8-naphthyridine (9b). Following the general procedure E, 0.194 mmol (38 mg) of 2-vinylindole **2b** and 0.46 mmol (50 mg) of 1,4,5,6-tetrahydropyridine (**4a**) in 60 mL of CH_3CN (+0.1 M $LiClO_4$) were electrolyzed. Separation of the crude product andpurification by HPLC (Nucleosil Rp-18, MeOH/H_2O) (7.5:2.5) afforded 40 mg (68%) of **9b** as a pale yellow solid: mp 105 °C (crystallized from CCl_4); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.62 (d, 1H, J = 8.0 Hz, H-8), 7.60 (d, 1H, J = 8.0 Hz, H-11), 7.30 (dd, 1H, J = 8.0, 8.0 Hz, H-10), 7.18 (dd, 1H, J = 8.0, 8.0 Hz, H-12), 5.14 (s, 1H, H-12a), 2.98 (ddd, 1H, J = 3.5, 4.5, 13.0 Hz, H-2eq), 2.94 (ddd, 1H, J = 4.0, 8.0, 13.0 Hz, H-2ax), 2.62 (s, 3H, 7-CH_3), 2.44 (s, 3H, 5-CH_3), 2.40 (m, 1H, H-4eq), 2.14 (ddd, 1H, J = 3.5, 9.5, 14.0 Hz, H-4ax), 1.86 (ddddd, 1H, J = 3.5, 3.5, 4.0, 4.0, 14.0 Hz, H-3eq), 1.48 (ddddd, 1H, J = 4.0, 4.5, 8.0, 9.5, 14.0 Hz, H-3ax); ^{13}C NMR (100.6 MHz, $CDCl_3$) δ 141.3 (C-5), 136.2 (C-11a), 130.2 (C-7a), 124.9 (C-10), 124.1 (C-7), 120.9 (C-9), 120.0 (C-8), 118.9 (6-CN), 115.2 (C-6a), 114.1 (4a-CN), 110.4 (C-6), 110.3 (C-11), 68.8 (C-12a), 43.2 (C-4a), 42.1 (C-2), 30.5 (C-4), 22.4 (C-3), 18.5 (5-CH_3), 8.8 (7-CH_3); high-resolution MS calcd for $C_{19}H_{18}N_4$ (M^+) 302.1531, found 302.1531.

7-(2-Ethoxyethyl)-4a,6-dicyano-5-methylindolo-[1,2-*a*]-1,2,3,4,4a,12a-hexahydro-1,8-naphthyridine (9c). Following the general procedure E, 0.047 mmol (12 mg) of 2-vinylindole **2c** and 0.29 mmol (32 mg) of 1,4,5,6-tetrahydropyridine (**4a**) in 50 mL of CH_3CN (+0.1 M $LiClO_4$) were electrolyzed. Separation of the crude product and purification by HPLC (Nucleosil Rp-18, MeOH/H_2O) (8.5:1.5) afforded 9 mg (53%) of **9c** as a pale yellow solid: mp 45 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.63 (d, 1H, J = 7.5 Hz, H-8), 7.60 (d, 1H, J = 7.5 Hz, H-11), 7.30 (dd, 1H, J = 7.5, 7.5 Hz, H-10), 7.18 (dd, 1H, J = 7.5, 7.5 Hz, H-9), 5.33 (s, 1H, H-12a), 3.69 (ddd, 1H, J = 7.0, 7.5, 14 Hz, H-17′), 3.67 (ddd, 1H, J = 7.0, 7.5, 14.0 Hz, H-17), 3.54 (q, 2H, J = 7.0 Hz, H-18), 3.43 (ddd, 1H, J = 7.0, 7.5, 14.0 Hz, H-16′), 3.39 (ddd, 1H, J = 7.0, 7.5, 14.0 Hz, H-16), 3.01 (ddd, 1H, J = 3.5, 4.5, 13.0 Hz, H-2eq), 2.94 (ddd, 1H, J = 4.0, 8.0, 13.0 Hz, H-2ax), 2.44 (s, 3H, 5-CH_3), 2.41 (m, 1H, H-4eq), 2.13 (ddd, 1H, J = 3.5, 9.5, 14.5 Hz, H-4ax), 1.85 (ddddd, 1H, J = 3.5, 3.5, 4.0, 4.0, 14.0 Hz, H-3eq), 1.49 (ddddd, 1H, J = 4.0, 4.5, 8.0, 9.5, 14.0 Hz, H-3ax), 1.19 (t, 3H, J = 7.0 Hz, H-19); ^{13}C NMR (100.6 MHz, $CDCl_3$) δ 142.1 (C-5), 136.2 (C-11a), 129.8 (C-7a), 124.9 (C-10), 124.6 (C-7), 121.1 (C-9), 120.2 (C-8), 118.8 (4a-CN), 115.2 (C-6a), 115.0 (6-CN), 110.4 (C-11), 106.5 (C-6), 70.7 (C-17), 68.7 (C-12a), 66.2 (C-18), 43.3 (C-4a), 42.0 (C-2), 30.5 (C-4), 24.4 (C-16), 22.4 (C-3), 18.6 (5-CH_3), 15.2 (C-19); high-resolution MS calcd for $C_{22}H_{24}N_4O$ (M^+) 360.1950, found 360.1950.

4a-Carbomethoxy-6-cyano-5,7-dimethylindolo[1,2-*a*]-1,2,3,4,4a,12a-hexahydro-1,8-naphthyridine (10b). Following the general procedure E, 0.1 mmol (20 mg) of 2-vinylindole **2b** and 0.14 mmol (20 mg) of 1,4,5,6-

tetrahydropyridine (**4b**) in 20 mL of CH_3CN (+0.1 M $LiClO_4$) were electrolyzed for 40 min. Separation of the crude product and purification by HPLC (Nucleosil RP-18, MeOH/H_2O) (8:2) afforded 31 mg (91%) of a white solid: mp 45 °C; ^{1}H NMR (200 MHz, $CDCl_3$) δ 7.56 (d, 1H, J = 7.5 Hz, H-8), 7.36 (d, 1H, J = 7.5 Hz, H-11), 7.24 (dd, 1H, J = 7.5, 7.5 Hz, H-10), 7.12 (dd, 1H, J = 7.5, 7.5 Hz, H-9), 5.52 (s, 1H, H-12a), 3.58 (s, 3H, 4a-$COOCH_3$), 3.08 (ddd, 1H, J = 3.5, 4.5, 13.0 Hz, H-2eq) 2.88 (ddd, 1H, J = 4.0, 8.0, 13.0 Hz, H-2ax), 2.52 (m, 1H, H-4eq), 2.48 (s, 3H, 7-CH_3), 2.42 (s, 3H, 5-CH_3), 2.02 (ddd, 1H, J = 4.0, 8.0, 13.0 Hz, H-4ax), 1,72 (dddd, 1H, J = 3.5, 4.0, 4.0, 14.0 Hz, H-3eq), 1.32 (m, 1H, H-3ax); ^{13}C NMR (100.6 MHz, $CDCl_3$) δ 172.4 (4a-COR), 146.2 (C-5), 135.6 (C-11a), 130.0 (C-7a), 124.2 (C-7), 124.1 (C-10), 120.4 (C-9), 119.7 (C-8), 116.1 (6-CN), 111.9 (C-6a), 108.7 (C-11), 106.1 (C-6), 67.6 (C-12a), 53.0 (4a-OCH_3), 52.5 (C-4a), 44.6 (C-2), 31.3 (C-4), 22.9 (C-3), 19.3 (5-CH_3), 8.7 (7-CH_3); high-resolution MS calcd for $C_{20}H_{21}N_3O_2$ (M^+) 335.1633, found 335.1634. Anal. Calcd for $C_{20}H_{21}N_3O_2$: C, 71.62, H, 6.31. Found: C, 71.40, H, 6.40.

4a-Carbomethoxy-6-cyano-1,5,7-trimethylindolo-[1,2-*a*]-1,2,3,4,4a,12a-hexahydro-1,8-naphthyridine (11b). Following the general procedure E, 0.046 mmol (9 mg) of 2-vinylindole **2b** and 0.097 mmol (15 mg) of 1,4,5,6-tetrahydropyridine (**4c**) in 20 mL of CH_3CN (+0.1 M $LiClO_4$) were electrolyzed. Separation of the crude product and purification by HPLC (Nucleosil RP-18, MeOH/H_2O) (7:3) afforded 10 mg (62%) of a white solid: mp 148 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.55 (d, 1H, J = 7.5 Hz, H-8), 7.29 (d, 1H, J = 7.5 Hz, H-11), 7.24 (dd, 1H, J = 7.5, 7.5 Hz, H-10), 7.10 (dd, 1H, J = 7.5, 7.5 Hz, H-9), 4.84 (s, 1H, H-12a), 3.52 (s, 3H, 4a-$COOCH_3$), 2.92 (ddd, 1H, J = 3.5, 4.0, 13.0 Hz, H-2eq), 2.58 (s, 3H, 7-CH_3), 2.52 (m, 1H, H-4eq), 2.44 (s, 3H, 5-CH_3), 2.38 (ddd, 1H, J = 3.5, 13.0, 14.0 Hz, H-2ax), 1.94 (ddd, 1H, J = 4.5, 13.0, 14.0 Hz, H-4ax), 1.76 (ddddd, 1H, J = 3.5, 3.5, 3.5, 4.5, 13.0 Hz, H-3eq), 1.74 (s, 3H, 1-CH_3), 1.62 (ddddd, 1H, J = 4.0, 4.5, 13.0, 14.0, 14.0 Hz, H-3ax); ^{13}C NMR (100.6 MHz, $CDCl_3$) δ 172.3 (4a-COR), 147.9 (C-5), 137.6 (C-11a), 128.9 (C-7a), 124.3 (C-7), 120.0 (C-9), 119.6 (C-8), 116.4 (6-CN), 111.4 (C-6a), 109.2 (C-11), 106.0 (C-6), 73.7 (C-12a), 54.7 (C-2), 53.6 (C-4a), 52.9 (4a-$COOCH_3$), 42.6 (1-CH_3), 31.6 (C-4), 22.3 (C-3), 19.1 (5-CH_3), 8.6 (7-CH_3); high-resolution MS calcd for $C_{21}H_{23}N_3O_2$ (M^+) 349.1790, found 349.1790.

5-(Methoxycarbonyl)-2,2-dicyano-1,2-dihydro-3*H*-pyrido[3,2,1-*jk*]carbazole (12e). Following the general procedure E, 0.056 mmol (13 mg) of 2-vinylindole **2e** and 0.14 mmol (18 mg) of 3-(*N*,*N*-dimethyamino)methylacrylate (**3a**) in 25 mL of CH_3CN/CH_2Cl_2 (1:1) (+0.1 M $LiClO_4$) were electrolyzed. Separation of the reaction mixture and purification of the product by preparative TLC (CH_2Cl_2) afforded 2 mg (11%) of a yellow solid: mp 180 °C dec; ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.08 (s,1H, H-6), 8.02 (d, H, J = 7.5, 7.5 Hz, H-8), 7.76 (d, 1H, J = 7.5/7.5 Hz, H-11), 7.48 (dd, J = 7.5, 7.5 Hz, H-10), 7.42 (dd, 1H, J = 7.5, 7.5 Hz, H-9), 7.28 (s, 1H, H-4), 3.98 (s, 3H, 5-$COOCH_3$), 3.94 (s, 2H, H-1), 3.64 (s, 2H, H-3); ^{13}C NMR (100.6 MHz, $CDCl_3$) δ 165.7 (5-COOR), 131.0 (C-7a), 129.4 (C-11a), 128.1 (C-5), 124.8 (C-11b), 122.0 (C-9), 120.0 (C-10), 118.9 (2-CN), 118.2 (2-CN), 115.3 (C-11), 112.6 (C-4), 111.5 (C-8), 96.0 (C-6), 52.3 (5-OCH_3), 48.7 (C-2), 37.5 (C-1), 32.7 (C-3); high-resolution MS calcd for $C_{19}H_{13}N_3O_2$ (M^+) 315.1007, found 315.1007.

Spiro[γ-butyrolactone-2,5′-6-(dimethylamino)-1,2,5,6-tetrahydro-3-acetyl-3*H*-pyrido[3,2,1-*lm*]-β-carboline] (13d). Following the general procedure E, 0.071 mmol (17 mg) of 2-vinylindole **2d** and 0.18 mmol (25 mg) of enamino lactone **3d** in 40 mL of CH_3CN (+0.1 M $LiClO_4$) were electrolyzed. Separation of the crude product and purification by flash chromatography (MTBE/MeOH) (20:1) afforded 10 mg (39%) of **13d** as a pale yellow solid: mp 173 °C (crystallized from benzene);[31] ^{1}H NMR (400 MHz, 345 K, C_6D_6) δ 7.47 (d, 1H, J = 7.5 Hz, H-11), 7.30 (d, 1H, J = 7.5 Hz, H-8), 7.22 (dd, 1H, J = 7.5, 7.5 Hz, H-9), 7.16 (dd, 1H, J = 7.5, 7.5 Hz, H-10), 5.87 (s, 1H, br, H-4), 4.92 (s, 1H, H-6), 3.78 (dd, 2H, J = 5.0, 8.0 Hz, H-14), 3.66 (m, 1H, H-2), 3.56 (m, 1H, H-2′), 2.56 (ddd, 1H, J = 4.5, 7.5, 16.0 Hz, H-1), 2.42 (ddd, 1H, J = 4.5, 7.5, 16.0 Hz, H-1′), 2.34 (dt, 1H, J = 8.0, 13.5 Hz, H-15), 2.10 (s, 6H, 6-N(CH_3)$_2$), 1.94 (s, 3H, 3-$COCH_3$), 1.76 (dt, 1H, J = 5.0, 13.5 Hz, H-15′); ^{13}C NMR (100.6 MHz, $CDCl_3$) δ 176.1 (3-COR), 170.1 (s, C-12), 138.7 (C-7a), 128.7 (C-11a), 126.7 (C-11b), 123.0 (C-9), 120.1 (C-10), 119.1 (C-11), 111.4 (C-8), 107.3 (C-11c), 73.0 (C-6), 65.2 (C-14), 49.5 (C-5), 45.5 (C-2), 42.6 (6-N(CH_3)$_2$), 33.1 (C-1), 24.0 (3-$COCH_3$), 21.6 (C-15); the signals of C-2 and C-4 could be determined by an inverse $^1J_{C-H}$ NMR experiment at the temperature listed above and cannot be obtained by DEPT; HRMS calcd for $C_{21}H_{23}N_3O_3$ (M^+) 365.1739, found 365.1739.

X-ray Crystallographic Constitution and Stereostructure Determination of 13d. Slightly yellow single crystals suitable for the collection of X-ray diffraction data were obtained by recrystallization from a solution of **13d** in benzene. A crystal having the dimensions 0.30 × 0.20 × 0.10 mm was selected for data collection and mounted on a Syntex $P2_1$ automated four-circle diffractometer. The radiation used was Mo Kα monochromatized by a highly ordered graphite crystal. Final cell constants, as well as other information pertinent to data collection and refinement, were as follows: space group $P21/c$ (No. 14), monoclinic, cell constants (Å), a = 10.541 (5), b = 21.695 (10), c = 11.046 (12); β (deg) = 117.43 (10); V (Å^3) = 2242 (3); molecular formula $C_{108}H_{116}N_{12}O_{12}$; formula weight 1774.18; formula units per cell, Z = 4; density d_{calcd} (g cm^{-3}) = 1.314; collection range, 0° < 2q < 50.0°; total of reflections collected, 5190; unique structure factors, 3944; "observed" reflections with $F > 0$ $s(F)$, 1456; refined parameters, 301; R = 0.0833. The structure was solved by direct methods. The non-hydrogen atoms were refined anisotropically. Hydrogen atoms were entered with a common isotropic temperature factor.

1-Cyano-2-methyl-4-carbomethoxy-9H-carbazole (14a). Following the general procedure P, 0.76 mmol (120 mg) of 2-vinylindole **2a** and 1.14 mmol (147 mg) of methyl 3-(*N*,*N*-dimethyamino)acrylate (**3a**) in 100 mL of CH_2Cl_2 and 25 mg of TAP (**1**) were irradiated for 5.5 h with a 1000 W Xenon arc lamp. Separation of the crude product and purification by flash chromatography with toluene/ethyl acetate (20:1) afforded **5a** as, under normal conditions, an unstable solid. Yield (74%, yellow solid) was determined by GC. Yield is optimized for this compound which is generated beneath **5a**: ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.82 (s, 1H, H-3), 7.94 (s, br, 1H, H-9), 7.85 (d, J = 8.0 Hz, 1H, H-8), 7.36 (dd, J = 8.0, 8.0 Hz, 1H, H-6), 7.12 (dd, J = 8.0, 8.0 Hz, 1H, H-7), 6.92 (d, J = 8.0 Hz, 1H, H-5), 3.62 (s, 3H, 4-$COOCH_3$), 3.02 (s, 3H,

(31) The author has deposited atomic coordinates for **13d** with the Cambridge Cystallographic Data Centre. The coordinates can be obtained, on request, from the Director, Cambridge Crystallographic Data Centre, 12 Union Road, Cambridge, CB2 1EZ, UK.

2-CH_3); high-resolution MS calcd for $C_{16}H_{12}N_2O_2$ (M^+) 264.0898, found 264.0898.

2,3-Dimethyl-1,4-dicyano-9*H*-carbazole (15a). Following the general procedure P, 0.55 mmol (100 mg) of 2-vinylindole **2a** and 0.90 mmol (100 mg) of 3-(*N*,*N*-dimethyamino)-2-butenenitrile (**3e**) in 100 mL of CH_2Cl_2 and 20 mg of TAP (**1**) were irradiated for 6 h with a 1000 W Xenon arc lamp. Separation of the crude product and purification by flash chromatography with toluene afforded 17 mg (13%) of **15a** as a yellow solid: mp 264 °C; ^{1}H NMR (400 MHz, $CDCl_3$ + CD_3OD) δ 8.43 (d, J = 8.0 Hz, 1H, H-8), 7,42 (d, J = 8.0 Hz, 1H, H-5), 7.40 (dd, J = 8.0, 8.0 Hz, H-7), 7.22 (dd, J = 8.0, 8.0 Hz, 1H, H-6), 2.58 (s, 3H, 3-CH_3), 2.56 (s, 3H, 2-CH_3), H-9 exchanges; ^{13}C NMR (100.6 MHz, $CDCl_3$) δ 140.5 (C-8a), 139.3 (C-9a), 137.7 (C-2), 131.4 (C-3), 128.0 (C-7), 122.4 (C-4b), 121.2 (C-5), 120.7 (C-6), 120.2 (C-4a), 117.1 (1-CN), 115.7 (4-CN), 111.5 (C-8), 107.7 (C-4), 98.4 (C-1), 18.8 (3-CH_3), 17.7 (2-CH_3); high-resolution MS calcd for $C_{16}H_{11}N_3$ (M^+) 245.0953, found 245.0953.

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Supporting Information Available: Additional spectral data and copies of NMR spectra of reported compounds (28 pages). This material is contained in libraries on microfiche, immediately follows this article in the microfilm version of the journal, and can be ordered from the ACS; see any current masthead page for ordering information.

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