Synthesis of Nitrogen-Containing Heterocycles from the Azido-Selenenylation Products of Unsaturated Carbonyl Compounds

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Received May 21, 1993•

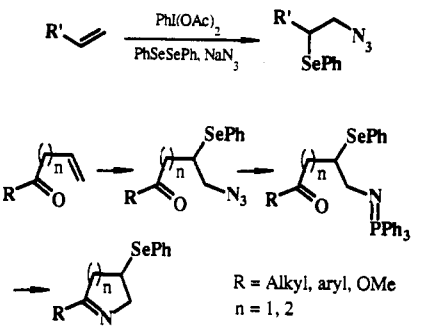
Terminal alkenes containing a remote carbonyl group reacted with iodobenzene diacetate, diphenyl diselenide, and sodium azide to afford the products of azido-phenylselenenylation of the double bond. Owing to its radical nature, this reaction proceeded with complete anti-Markovnikov regioselectivity. Under the influence of triphenylphosphine in benzene the azido group reacted with the carbonyl function to afford the corresponding ring-closure reaction products containing a carbon nitrogen double bond. Thus, starting from β,γ- or γ,δ-unsaturated esters, the corresponding cyclic imino ethers were obtained. These could not be isolated but were directly transformed into β-(phenylseleno) γ-lactams or γ-(phenylseleno) δ-lactams. The phenylseleno derivatives of tetrahydropyridine were formed starting both from γ,δ-unsaturated ketones and from α-allyl β-keto esters. In the latter case, the cyclization reaction is chemoselective and involves the ketonic carbonyl. The oxidation of these compounds with hydrogen peroxide directly produced the corresponding pyridines via selenoxide elimination followed by dehydrogenation. This simple reaction sequence represents a very useful general method to build up a 2-substituted pyridine ring. Several alkyl-, aryl-, and heteroarylpyridines, bipyridines, and a terpyridine have been prepared.

Introduction

We have recently reported that the reaction of an alkene with iodobenzene diacetate, diphenyl diselenide, and sodium azide in methylene chloride (Scheme I) affords the product of azido-phenylselenenylation of the double bond as a result of a radical process initiated by azido radicals.[1] This represents a new, very simple, and efficient method to introduce an azido group into an organic molecule. In addition, the products obtained from the azido-selenenylation of alkenes contain the phenylseleno and the azido groups which are very versatile functions which can be used for several conversions. Several examples of the synthetic utility of this kind of compounds are now reported. Terminal alkenes containing a remote carbonyl group easily react with iodobenzene diacetate, diphenyl diselenide, and sodium azide to regioselectively afford the products of azido-phenylselenenylation of the double bond.

These compounds were employed to effect new syntheses of nitrogen-containing heterocycles by means of intramolecular aza-Wittig reactions,[2,3] according to the reaction sequence indicated in Scheme I. Thus, under the influence of triphenylphosphine in benzene the seleno azides were transformed into the corresponding iminophosphoranes. These intermediates were not isolated since they easily react with the carbonyl function to afford the corresponding ring-closure reaction products containing a carbon nitrogen double bond. This procedure has been employed to effect the conversions of β,γ- or γ,δ-unsaturated esters into β-(phenylseleno) γ-lactams or γ-(phenylseleno) δ-lactams and of γ,δ-unsaturated ketones and α-allyl β-keto esters into the phenylseleno derivatives of tetrahydropyridine. Moreover, the simple oxidation of these latter

Scheme I



compounds with hydrogen peroxide directly produced the corresponding pyridines derived from selenoxide elimination and dehydrogenation. Thus, with this simple reaction sequence a 2-substituted pyridine ring can be formed. The general application of this method has been explored by effecting the synthesis of several alkyl-, aryl-, and heteroarylpyridines, of 2,2′-, 2,3′- and 2,4′-bipyridines, and of a 2,2′:6,2″-terpyridine.

Results and Discussion

The first substrates investigated were the unsaturated esters (Scheme II and Table I). The seleno azides **2a–2d** could be easily obtained from the methyl 3-butenoates **1a–1d** using the previously described procedure for the radical azido-phenylselenenylation of double bonds.[1] Addition of triphenylphosphine to solutions of these compounds in benzene resulted in nitrogen evolution indicating that the azides were converted into the corresponding iminophosphoranes. However, evidence for the formation of these intermediates could not be obtained. Very likely they rapidly react to give the ring-closure reaction products **3a–3d**. These cyclic imidates also were quite unstable intermediates. Their isolation was unsuccessful under several experimental conditions, the lactams **4a–4d** and other unidentified products being obtained in every case. Only in the case of the reaction of **2d** was the formation

• Abstract published in *Advance ACS Abstracts*, September 15, 1993.

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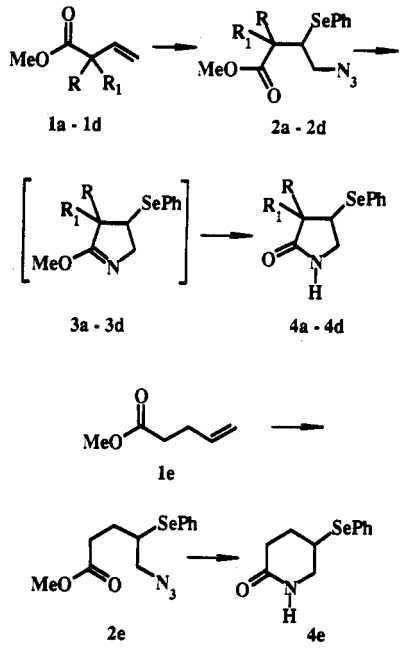
(2) Eguchi, S.; Matsushita, Y.; Yamashita, K. *Org. Prep. Proced. Int.* **1992**, *24*, 209. Gololobov, Y. G.; Kasukhin, L. F. *Tetrahedron* **1992**, *48*, 1353. Scriven, E. F. V.; Turnbull, K. *Chem. Rev.* **1988**, *88*, 297.

(3) Molina, P.; Vilaplana, M. J.; Pastor, A. *Synthesis* **1992**, 873 and references cited therein.

0022-3263/93/1958-6097$04.00/0

Scheme II



of the cyclic imidate **3d** clearly demonstrated by GC–MS. Two isomers, in a 1:1 ratio, were observed, and their mass spectra were in full agreement with the proposed structures. The cleanest way to convert these imidates into the 4-(phenylseleno) γ-lactams **4a–4d** was to stir overnight at room temperature their methanolic solution in the presence of silica gel. A 1:1 and a 2:1 mixture of the two diastereoisomers was obtained in the case of **4b** and **4d**, respectively. However, only in the case of **4b** could these be separated by column chromatography. The results of these experiments are summarized in Scheme II and in Table I (entries 1–4). Also reported in Scheme II is the reaction of the methyl 4-pentenoate (**1e**) which, under the same conditions, gave the seleno azide **2e** and the 5-(phenylseleno) δ-lactam **4e** (Table I, entry 5).

The results of these experiments indicate that the present procedure represents a convenient synthesis of lactams. A different approach to such nitrogen-containing heterocycles consists of the ring-closure reactions of alkenamides induced by electrophilic reagents. We have recently reported that the strongly electrophilic phenylselenenyl sulfate, generated by oxidation of diphenyl diselenide with ammonium persulfate, can easily promote ring-closure reactions starting from alkenes containing internal nucleophiles. Seleno etherification and seleno lactonization reactions in which the nucleophile is an oxygen atom were easily carried out.[4] Attempts to synthesize γ-lactams starting from unsaturated amides were unsuccessful since also in this case the oxygen and not the nitrogen atom acts as the nucleophile to afford cyclic imidates which eventually give γ-lactones.[4] Other authors have made similar observations[5] with other electrophilic reagents.[6] It has been shown that in order to effect the desired cyclization reaction the nitrogen atom must be that of a cyclic imidate,[7] an *N,O*-bis(trimethylsilyl)imidate,[8] or a thioimidate.[9] Since unsaturated esters can be easily converted into seleno azides the aza-Wittig reaction seemed to be an easier route to γ- and δ-lactams.

An experiment was also carried out on an unsaturated ester with a different structure, the vinyl 2-ethylhexanoate (**1f**), from which it was expected to eventually obtain a dihydrooxazole nucleus. Indeed **1f** gave the seleno azide **2f** (as a mixture of two diastereoisomers) which cyclized to afford a mixture of two compounds, *i.e.*, the expected product **4f** and an isomer to which the structure **4g** was attributed (Scheme III, and Table I, entry 6).

The results obtained starting from the unsaturated ketones **6a–6c** are summarized in Scheme IV and in Table I (entries 7–9). The azido-selenenylation products **7a–7c** were obtained regiospecifically and in good yields. Also in this case the cyclization reaction proceeded smoothly, and as expected it afforded the 2-methyl (**8a**), the 2-phenyl (**8b**), and the 2-(2-thienyl)-5-(phenylseleno)tetrahydropyridine (**8c**).

Finally, experiments were carried out on the allyl derivatives of β-keto esters **10a–10g**. The seleno azides **11a–11g** (mixtures of two diastereoisomers) obtained from these unsaturated compounds could in principle give rise to ring-closure reactions either through the ketonic or the ester carbonyl or both. As indicated in Scheme V, this reaction was highly chemoselective; the cyclization reaction in fact involved exclusively the ketonic function and afforded the carbethoxy or carbomethoxy tetrahydropyridines **12a–12g**. In the case of compounds **12b–12g** the double bond becomes conjugated with the ester group. The results of these experiments are summarized in Table I (entries 10–16).

The selectivity observed in these ring-closure reactions is not unprecedented. An example, in which azides having structures similar to those employed in the present investigation give rise to tetrahydropyridines and not to cyclic imino ethers, has already been reported in the literature.[10]

Thus, starting from easily available unsaturated ketones or α-allyl β-keto esters it is possible to obtain in two steps and in good yields different kinds of tetrahydropyridine derivatives. The examples reported above indicate that the present procedure is of general application and it can very likely be applied to the synthesis of molecules having more complex structures. Moreover, the tetrahydropyridines so obtained contain a phenylseleno group which can promote several further conversions.[11] We have examined the simplest reaction of this functional group, *i.e.*, its oxidation to the selenoxide. It is well known that such compounds suffer spontaneous elimination to afford alkenes.[11,12] In the present case, therefore, it was expected to obtain dihydropyridine derivatives. However, when the tetrahydropyridines **8c** and **12b–12g** were treated with hydrogen peroxide in methanol at 0 °C and then at room temperature, the pyridines **9** and **13b–13g** were obtained (Chart I and Table I, entries 9 and 11–16). Clearly, under these experimental conditions, the initially formed dihy-

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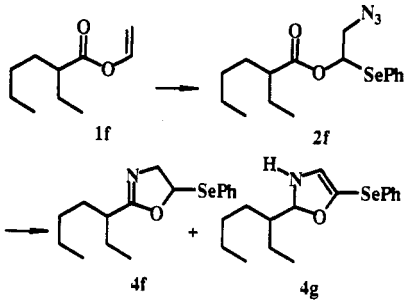
Table I

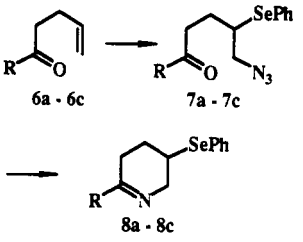
entry	alkene	R	R_1	seleno azide[a]	yield[b] (%)	cyclization product[c]	yield[b] (%)	pyridine[d]	yield[b] (%)
				Reactions of Unsaturated Esters					
1	1a	H	H	2a	65	4a	60		
2	1b	H	Me	2b	60	4b[e]	71		
3	1c	Me	Me	2c	72	4c	54		
4	1d	Me	CH_2Ph	2d	73	4d[f]	87		
5	1e			2e	80	4e	90		
6	1f			2f	88	4f + 4g[g]	75	5[h]	
				Reactions of Unsaturated Ketones					
7	6a	Me		7a	71	8a	74		
8	6b	Ph		7b	68	8b	83		
9	6c	2-Th		7c	68	8c	67	9	67
				Reactions of α-Allyl β-Keto Esters					
10	10a			11a	56	12a	78		
11	10b	Ph	Et	11b	50	12b	75	13b	90
12	10c	Et	Me	11c	52	12c	82	13c	55
13	10d	2-Pyr	Me	11d	65	12d	80[i]	13d	76
14	10e	3-Pyr	Me	11e	60	12e	85	13e	87
15	10f	4-Pyr	Me	11f	62	12f	90	13f	82
16	10g	3-*N*-methylpyrrole	Me	11g	71	12g	58	13g	40
17	10h		CO_2Me	11h	60[i]	12h	78[i]	13h	75

[a] Obtained from the alkenes by treatment with PhSeSePh, NaN_3, and $PhI(OAc)_2$ in CH_2Cl_2 at room temperature for 10–12 h. [b] Reaction yields were calculated on isolated products after column chromatography. [c] Obtained from the seleno azides by treatment with PPh_3 in benzene at room temperature and then at 50 °C for 0.5 h. Compounds **4a–4g** were obtained by stirring the cyclization products overnight with SiO_2 in methanol at room temperature. [d] Obtained from the tetrahydropyridines by treatment with H_2O_2 at 0 °C and then at room temperature. [e] 1:1 mixture of two stereoisomers separated by column chromatography. [f] 2:1 mixture of two stereoisomers not separated by column chromatography. [g] 1:1 mixture of the two products. [h] In this case the reaction product was an oxazole derivative. This could not be purified, but it was identified by GC–MS. [i] This was a complex mixture of stereoisomers which was directly used for further reactions.

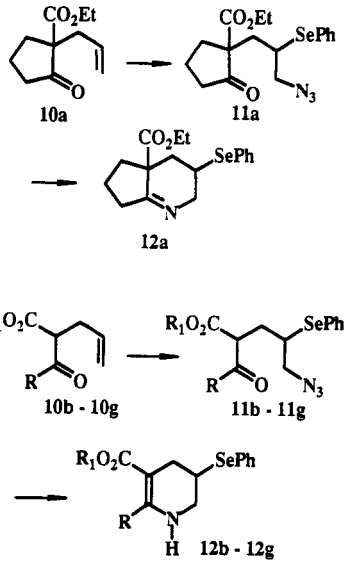
Scheme III



Scheme IV



Scheme V



dropyridines are dehydrogenated to afford the corresponding aromatic compounds.

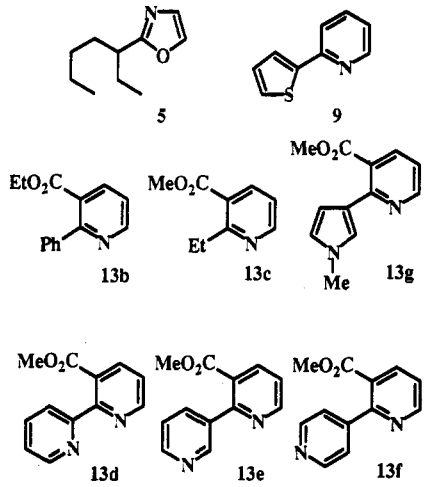
Also reported in Chart I and in Table I (entry 6) is the oxazole **5** derived from the oxidation of **4f**. Compound **5** could not be obtained in a pure form, but it was identified by GC–MS.

As a further example of the versatility of these reactions we have carried out the synthesis of the terpyridine **13h** (Scheme VI and Table I, entry 17). The starting compound **10h** was easily prepared from 2,6-diacetylpyridine. The corresponding seleno azide **11h** and tetrahydropyridine **12h** were obtained as mixtures of several stereoisomers which were separated from other products by column chromatography and then used to eventually afford the terpyridine **13h** as the sole reaction product.

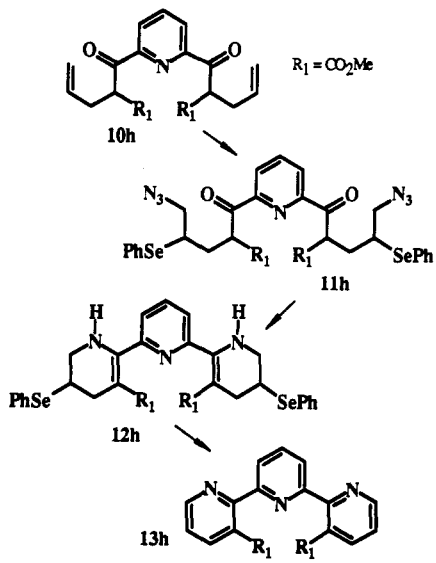
Thus, starting from unsaturated ketones or from α-allyl β-keto esters, the sequence of three reactions, azido-selenenylation, aza-Wittig, and oxidation, represents a very convenient and general method to build up a 2-substituted pyridine nucleus. Moreover, the formation of **5** indicates that with an appropriate choice of starting unsaturated carbonyl compound it is possible to synthesize other nitrogen-containing heterocycles. Of particular interest is the synthesis of unsymmetrical biaryls in which both rings are heteroaromatic. Symmetrical heteroaromatic biaryls can be efficiently prepared from aryl halides by means of the nickel-promoted homo coupling reaction.[13,14] However, when the same reaction is applied to the synthesis of the unsymmetrical compounds the results are not completely satisfactory[14] and the method described in the present paper favorably compares with those reported in the literature.

(13) Tiecco, M.; Testaferri, L.; Tingoli, M.; Chianelli, D.; Montanucci, M. *Synthesis* **1984**, 736.
(14) Unpublished results from this laboratory.

Chart I



Scheme VI



Experimental Section

Product identification was effected by proton and carbon-13 NMR spectroscopy, mass spectrometry, and elemental analyses.[15]

Preparation of the Starting Unsaturated Compounds. Compounds **1a**, **1e**, **1f**, and **6a** were commercially available. Compounds **1b**, **1c**, and **1d** were obtained from **1a** by alkylation under standard reaction conditions. The ketones **6b** and **6c** were obtained by allylation of the corresponding aryl methyl ketones.[16] Compounds **10a–10g** were prepared by allylation of the corresponding β-keto esters under standard reaction conditions. The β-keto esters necessary for the syntheses of **10a**, **10b**, and **10c** were commercially available. The β-keto esters necessary for the syntheses of **10d–10h** were prepared from the corresponding aryl methyl ketones according to the procedure reported in the literature.[17] All new compounds were fully characterized by spectroscopic methods.[15]

Radical Azido-Phenylselenenylation Reactions. These reactions were carried out on the unsaturated compounds **1a–1f**, **6a–6c**, and **10a–10h** (3–5 mmol) according to the general procedure reported previously.[1] The results of these experiments are summarized in Schemes II–VI and in Table I. Compounds **2a**, **2e**, and **7a** have already been described.[1] Physical and spectral data of the other seleno azides are reported below. Compounds **2b**, **2d**, **2f**, and **11a–11g** were mixtures of two diastereoisomers. Compound **11h** was a complex mixture of stereoisomers.

Methyl 2-methyl-3-(phenylseleno)-4-azidobutanoate (2b): oil (1:1 mixture of two isomers); IR (neat) 2090, 1720 cm^{-1}; ^1H NMR δ 7.6–7.5 (m, 4 H), 7.4–7.2 (m, 6 H), 3.7 (s, 3 H), 3.65 (s, 3 H), 3.65–3.5 (m, 5 H), 3.4–3.3 (m, 1 H), 3.0–2.9 (m, 2 H), 1.3 (d, 3 H, J = 7.1 Hz), 1.25 (d, 3 H, J = 7.0 Hz); ^{13}C NMR δ 173.9, 134.9, 134.8, 129.2, 128.1, 128.0, 54.3, 54.0, 51.8, 47.5, 47.3, 41.6, 41.4, 15.2, 13.8; MS m/z (relative intensity) 313 (1), 285 (8), 158 (32), 157 (100), 72 (72). Anal. Calcd for $C_{12}H_{15}N_3O_2Se$: C, 46.16; H, 4.84; N, 13.46. Found: C, 46.27; H, 4.78; N, 13.51.

Methyl 2-benzyl-2-methyl-3-(phenylseleno)-4-azidobutanoate (2d): oil (2:1 mixture of two isomers); IR (neat) 2090, 1720 cm^{-1}; ^1H NMR δ 7.7–7.6 (m, 4 H), 7.4–7.1 (m, 12 H), 7.1–7.0 (m, 4 H), 3.9–3.5 (m, 6 H), 3.6 (s, 3 H), 3.5 (s, 3 H), 3.28 (d, 1 H, J = 13.0 Hz), 3.18 (d, 1 H, J = 13.0 Hz), 3.07 (d, 1 H, J = 13.0 Hz), 2.85 (d, 1 H, J = 13.0 Hz), 1.2 (s, 3 H), 1.15 (s, 3 H); ^{13}C NMR δ 174.3, 136.6, 134.5, 134.3, 130.1, 129.4, 129.1, 128.1, 127.9, 126.8, 96.2, 55.6, 54.7, 54.4, 53.6, 52.2, 51.7, 51.5, 46.1, 44.2, 18.8, 17.2. Anal. Calcd for $C_{19}H_{21}N_3O_2Se$: C, 56.72; H, 5.26; N, 10.44. Found: C, 56.64; H, 5.31; N, 10.52.

1-(2-Thienyl)-4-(phenylseleno)-5-azidopentan-1-one (7c): oil; IR (neat) 2090, 1700 cm^{-1}; ^1H NMR δ 7.72 (dd, 1 H, J = 1.2 and 3.8 Hz), 7.62 (dd, 1 H, J = 1.2 and 5.0 Hz), 7.6–7.5 (m, 2 H), 7.35–7.2 (m, 3 H), 7.12 (dd, 1 H, J = 3.8 and 5.0 Hz), 3.62 (dd, 1 H, J = 5.5 and 12.5 Hz), 3.49 (dd, 1 H, J = 7.3 and 12.5 Hz), 3.4–2.3 (m, 1 H), 3.2 (t, 2 H, J = 7.5 Hz), 2.3 (ddt, 1 H, J = 4.5, 7.5, and 14.8 Hz), 1.9 (ddt, 1 H, J = 6.9, 9.6, and 14.8 Hz); ^{13}C NMR δ 191.7, 143.1, 135.2, 133.6, 131.8, 129.2, 128.0, 56.1, 44.1, 37.1, 27.2. Anal. Calcd for $C_{15}H_{15}N_3OSSe$: C, 49.45; H, 4.15; N, 11.53. Found: C, 49.51; H, 4.08, N, 11.50.

Ethyl 1-[2-(phenylseleno)-3-azidopropyl]-2-oxo-1-cyclopentanecarboxylate (11a): oil; IR (neat) 2090, 1720 cm^{-1}; ^1H NMR δ 7.7–7.5 (m, 2 H), 7.4–7.2 (m, 3 H), 4.2 (q, 2 H, J = 7.0 Hz), 3.6–3.4 (m, 2 H), 2.8–2.6 (m, 1 H), 2.6–2.4 (m, 2 H), 2.2–1.8 (m, 6 H), 1.3 (t, 3 H, J = 7.0 Hz); ^{13}C NMR δ 170.0, 134.9, 131.8, 129.2, 128.2, 61.7, 60.1, 56.5, 40.5, 37.3, 36.3, 33.2, 19.4, 13.9. Anal. Calcd for $C_{17}H_{21}N_3O_3Se$: C, 51.78; H, 5.37; N, 10.66. Found: C, 51.69; H, 5.45; N, 10.58.

Ethyl 2-benzoyl-4-(phenylseleno)-5-azidopentanoate (11b): oil (1:1 mixture of two isomers); IR (neat) 2090, 1730 cm^{-1}; ^1H NMR δ 8.1–8.0 (m, 4 H), 7.7–7.4 (m, 10 H), 7.3–7.1 (m, 6 H), 5.0–4.8 (m, 2 H), 4.2–4.0 (m, 4 H), 3.7–3.4 (m, 4 H), 3.4–3.2 (m, 1 H), 3.2–3.0 (m, 1 H), 2.7–2.4 (m, 2 H), 2.2–1.9 (m, 2 H), 1.2–1.0 (m, 6 H); ^{13}C NMR δ 194.6, 194.3, 169.2, 136.4, 135.4, 135.2, 133.5, 129.3, 129.2, 128.7, 128.6, 128.3, 61.6, 61.5, 56.5, 56.2, 52.7, 52.2, 43.5, 42.8, 32.2, 31.8, 13.9. Anal. Calcd for $C_{20}H_{21}N_3O_3Se$: C, 55.82; H, 4.92; N, 9.76. Found: C, 55.90; H, 4.85; N, 9.81.

Methyl 2-nicotinoyl-4-(phenylseleno)-5-azidopentanoate (11e): oil (1:1 mixture of two isomers); IR ($CHCl_3$) 2120, 1755, 1700 cm^{-1}; ^1H NMR δ 9.25–9.15 (m, 2 H), 8.85–8.75 (m, 2 H), 8.35–8.15 (m, 2 H), 7.6–7.1 (m, 12 H), 4.95–4.8 (m, 2 H), 3.8–3.4 (m, 4 H), 3.7 (s, 3 H), 3.65 (s, 3 H), 3.4–3.2 (m, 1 H), 3.2–3.0 (m, 1 H), 2.8–2.45 (m, 2 H), 2.2–2.0 (m, 2 H); ^{13}C NMR δ 193.5, 193.3, 169.2, 169.1, 153.9, 153.8, 150.1, 150.0, 136.0, 135.8, 135.1, 131.1, 129.4, 129.3, 128.4, 128.3, 123.6, 123.5, 56.5, 56.3, 52.8, 52.7, 52.2, 43.3, 42.6, 32.1, 31.8. Anal. Calcd for $C_{18}H_{18}N_4O_3Se$: C, 51.81; H, 4.35; N, 13.43. Found: C, 51.68; H, 4.21; N, 13.17.

Cyclization Reactions. General Procedure. To a solution of the seleno azides **2a–2f**, **7a–7c**, and **11a–11h** (2 mmol) in benzene (5 mL) was slowly added a solution of triphenylphosphine (2.4 mmol) in benzene (3 mL) at rt. When nitrogen evolution was no longer observed the mixture was warmed at 50 °C for 0.5 h. The progress of the reaction was monitored by TLC and/or GC–MS. The solvent was evaporated, and the residue was chromatographed through a silica gel column. In the cases of the reactions of the seleno azides **2a–2f** the residue was dissolved in methanol, silica gel (2 g) was added, and the mixture was stirred overnight at rt. The solvent was evaporated, and the residue was purified by column chromatography. The results of these experiments are summarized in Schemes II–VI and in Table I. Physical and spectral data of the cyclization products are reported below.

4-(Phenylseleno)pyrrolidin-2-one (4a): mp 66–69 °C; IR ($CHCl_3$) 3650, 1700 cm^{-1}; ^1H NMR δ 7.7–7.5 (m, 2 H), 7.4–7.2 (m,

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3 H), 6.5 (br s, 1 H), 4.05–3.85 (m, 1 H), 3.75 (dd, 1 H, J = 7.3 and 10.5 Hz), 3.45 (dd, 1 H, J = 5.1 and 10.5 Hz), 2.75 (dd, 1 H, J = 8.4 and 17.5 Hz), 2.4 (dd, 1 H, J = 6.3 and 17.5 Hz); ^{13}C NMR δ 176.5, 134.9, 129.3, 128.3, 49.3, 37.7, 33.9; MS m/z (relative intensity) 241 (21), 183 (5), 158 (22), 104 (6), 84 (100). Anal. Calcd for $C_{10}H_{11}NOSe$: C, 50.01; H, 4.62; N, 5.83. Found: C, 49.95; H, 4.71; N, 5.88.

3-Methyl-4-(phenylseleno)pyrrolidin-2-one (4b). Isomer A: mp 137–140 °C; ^{1}H NMR δ 7.6–7.5 (m, 2 H), 7.4–7.2 (m, 3 H), 7.15 (br s, 1 H), 3.7–3.5 (m, 1 H), 3.5–3.3 (m, 2 H), 2.45–2.25 (m, 1 H), 1.25 (d, 3 H, J = 7.0 Hz); ^{13}C NMR δ 178.9, 135.4, 129.2, 128.3, 47.8, 42.8, 42.6, 13.9; MS m/z (relative intensity) 257 (14), 256 (8), 255 (79), 253 (37), 158 (59), 157 (32), 155 (23), 98 (100), 78 (30), 77 (35), 55 (93). Anal. Calcd for $C_{11}H_{13}NOSe$: C, 51.98; H, 5.15; N, 5.51. Found: C, 52.05; H, 5.10; N, 5.47. Isomer B: mp 66–69 °C; ^{1}H NMR δ 7.6–7.45 (m, 2 H), 7.3–7.2 (m, 3 H), 7.1 (br s, 1 H), 4.08 (ddd, 1 H, J = 4.6, 6.3, and 7.4 Hz), 3.66 (ddd, 1 H, J = 0.7, 6.3, and 10.8 Hz), 3.39 (ddd, 1 H, J = 1.1, 4.6, and 10.8 Hz), 2.7 (quintet, 1 H, J = 7.4 Hz), 1.25 (d, 3 H, J = 7.4 Hz); ^{13}C NMR δ 179.2, 134.2, 129.3, 128.6, 127.8, 47.8, 43.7, 40.4, 13.0; MS m/z (relative intensity) 257 (6), 255 (34), 158 (32), 157 (15), 156 (16), 155 (12), 98 (100), 78 (23), 77 (22), 55 (83). Anal. Calcd for $C_{11}H_{13}NOSe$: C, 51.98; H, 5.15; N, 5.51. Found: C, 51.91; H, 5.23; N, 5.58.

3,3-Dimethyl-4-(phenylseleno)pyrrolidin-2-one (4c): oil; ^{1}H NMR δ 7.9 (br s, 1 H), 7.6–7.3 (m, 2 H), 7.3–7.2 (m, 3 H), 3.6–3.3 (m, 3 H), 1.2 (s, 3 H), 1.15 (s, 3 H); ^{13}C NMR δ 181.5, 134.3, 129.0, 127.7, 50.2, 46.9, 43.4, 22.7, 20.7; MS m/z (relative intensity) 269 (76), 267 (36), 158 (30), 157 (24), 112 (53), 84 (16), 77 (23), 69 (100), 51 (12), 41 (42). Anal. Calcd for $C_{12}H_{15}NOSe$: C, 53.74; H, 5.64; N, 5.22. Found: C, 53.65; H, 5.71; N, 5.29.

2-Methoxy-3-benzyl-3-methyl-4-(phenylseleno)-1-pyrroline (3d). This product, as a 1:1 mixture of two isomers, was detected only by GC–MS: MS m/z (relative intensity) 359 (9), 267 (8), 202 (28), 111 (10), 91 (100); 359 (11), 202 (22), 91 (100).

3-Benzyl-3-methyl-4-(phenylseleno)pyrrolidin-2-one (4d): oil (2:1 mixture of two isomers); IR (neat) 1700 cm^{-1}; ^{1}H NMR δ (major isomer) 7.5–7.45 (m, 2 H), 7.35–7.2 (m, 5 H), 7.2–7.15 (m, 3 H), 6.9 (br s, 1 H), 3.65–3.3 (m, 3 H), 3.2 (d, 1 H, J = 13.7 Hz), 2.6 (d, 1 H, J = 13.7 Hz), 1.3 (s, 3 H); (minor isomer) 7.65–7.55 (m, 2 H), 7.35–7.2 (m, 5 H), 7.2–7.15 (m, 3 H), 7.0 (br s, 1 H), 3.65–3.3 (m, 2 H), 2.94 (d, 1 H, J = 13.6 Hz), 2.86 (d, 1 H, J = 13.6 Hz), 2.77 (t, 1 H, J = 9.8 Hz), 1.2 (s, 3 H); ^{13}C NMR δ 179.7, 137.3, 134.5, 134.3, 130.6, 130.3, 129.3, 129.2, 128.2, 127.9, 126.8, 126.5, 51.6, 48.8, 47.3, 46.9, 44.4, 41.8, 40.6, 22.4, 21.2; MS m/z (relative intensity) (major isomer) 345 (9), 188 (15), 158 (2), 157 (10), 91 (100); (minor isomer) 345 (7), 188 (22), 158 (3), 157 (5), 145 (18), 91 (100). Anal. Calcd for $C_{18}H_{19}NOSe$: C, 62.79; H, 5.56; N, 4.07. Found: C, 62.70; H, 5.63; N, 4.15.

5-(Phenylseleno)-2-piperidone (4e): mp 109–110 °C; IR ($CHCl_3$) 3420, 1650 cm^{-1}; ^{1}H NMR δ 7.7 (br s, 1 H), 7.6–7.5 (m, 2 H), 7.4–7.2 (m, 3 H), 3.6–3.3 (m, 3 H), 2.6–2.3 (m, 2 H), 2.3–2.0 (m, 1 H), 2.0–1.8 (m, 1 H); ^{13}C NMR δ 171.4, 135.1, 128.9, 128.0, 127.1, 47.6, 35.7, 30.6, 27.5; MS m/z (relative intensity) 255 (9), 158 (6), 157 (7), 106 (14), 98 (84), 78 (10), 77 (10), 55 (100). Anal. Calcd for $C_{11}H_{13}NOSe$: C, 51.98; H, 5.15; N, 5.51. Found: C, 51.91; H, 5.22; N, 5.58.

2-Methyl-5-(phenylseleno)-3,4,5,6-tetrahydropyridine (8a): oil; ^{1}H NMR δ 7.55–7.4 (m, 2 H), 7.1–7.0 (m, 3 H), 4.0–3.85 (m, 1 H), 3.6–3.4 (m, 1 H), 3.1–2.9 (m, 1 H), 1.8–1.3 (m, 4 H), 1.7 (s, 3 H); ^{13}C NMR δ 135.4, 129.2, 128.5, 128.0, 127.5, 54.9, 37.2, 30.9, 27.0, 26.7. Anal. Calcd for $C_{12}H_{15}NSe$: C, 57.15; H, 5.99; N, 5.55. Found: C, 57.07; H, 6.05; N, 5.62.

2-Phenyl-5-(phenylseleno)-3,4,5,6-tetrahydropyridine (8b): oil; ^{1}H NMR δ 7.8–7.65 (m, 2 H), 7.65–7.5 (m, 2 H), 7.4–7.2 (m, 6 H), 4.35–4.15 (m, 1 H), 3.9–3.7 (m, 1 H), 3.5–3.3 (m, 1 H), 2.9–2.75 (m, 1 H), 2.75–2.5 (m, 1 H), 2.35–2.15 (m, 1 H), 1.95–1.75 (m, 1 H); ^{13}C NMR δ 164.4, 139.3, 135.1, 129.5, 128.8, 128.0, 127.6, 125.8, 55.2, 36.8, 27.7, 26.8; MS m/z (relative intensity) 315 (19), 313 (10), 234 (46), 184 (19), 183 (15), 158 (91), 157 (19), 156 (27), 104 (83), 103 (35), 91 (44), 78 (20), 77 (43), 55 (100). Anal. Calcd for $C_{17}H_{17}NSe$: C, 64.97; H, 5.45; N, 4.46. Found: C, 65.05; H, 5.38; N, 4.52.

2-(2-Thienyl)-5-(phenylseleno)-3,4,5,6-tetrahydropyridine (8c): oil; ^{1}H NMR δ 7.69–7.55 (m, 2 H), 7.4–7.2 (m, 5 H), 7.1–7.0 (m, 1 H), 4.3–4.1 (m, 1 H), 3.9–3.6 (m, 1 H), 3.5–3.3 (m, 1 H), 3.0–2.8 (m, 1 H), 2.8–2.6 (m, 1 H), 2.4–2.1 (m, 1 H), 2.0–1.7 (m, 1 H); ^{13}C NMR δ 160.3, 135.4, 129.4, 129.1, 128.3, 127.9, 127.2, 126.3, 55.0, 36.9, 28.0, 26.6; MS m/z (relative intensity) 321 (18), 240 (35), 184 (21), 164 (100), 150 (18), 110 (60), 109 (44), 104 (31), 97 (24), 55 (92). Anal. Calcd for $C_{15}H_{15}NSSe$: C, 56.25; H, 4.72; N, 4.37. Found: C, 56.34; H, 4.79; N, 4.30.

6-(Ethoxycarbonyl)-4-(phenylseleno)-2-azabicyclo[4.3.0]non-1-ene (12a). Isomer A: oil; ^{1}H NMR δ 7.6–7.5 (m, 2 H), 7.3–7.2 (m, 3 H), 4.25–3.95 (m, 3 H), 3.55 (ddt, 1 H, J = 2.6, 10.5, and 17.7 Hz), 3.35–3.15 (m, 1 H), 2.77 (dd, 1 H, J = 3.5 and 12.7 Hz), 2.7–2.55 (m, 1 H), 2.4–2.2 (m, 2 H), 1.85–1.65 (m, 2 H), 1.6–1.3 (m, 2 H), 1.2 (t, 3 H, J = 7.0 Hz); ^{13}C NMR δ 174.1, 172.6, 134.7, 128.8, 127.6, 61.0, 55.1, 53.7, 36.7, 33.7, 32.8, 29.5, 19.2, 13.9; MS m/z (relative intensity) 351 (7), 278 (3), 194 (62), 157 (7), 120 (100), 77 (12). Anal. Calcd for $C_{17}H_{21}NO_2Se$: C, 58.29; H, 6.04; N, 4.00. Found: C, 58.20; H, 6.12; N, 3.95. Isomer B: oil; ^{1}H NMR δ 7.6–7.5 (m, 2 H), 7.3–7.2 (m, 3 H), 4.13 (q, 2 H, J = 7.2 Hz), 4.1–3.95 (m, 1 H), 3.4–3.05 (m, 2 H), 2.7–2.2 (m, 4 H), 2.1–1.55 (m, 4 H), 1.2 (t, 3 H, J = 7.2 Hz); ^{13}C NMR δ 178.5, 172.9, 134.8, 129.0, 128.5, 127.8, 61.4, 54.2, 37.7, 36.2, 34.8, 33.3, 20.1, 13.9; MS m/z (relative intensity) 351 (7), 194 (27), 184 (11), 167 (9), 157 (8), 121 (16), 120 (100), 77 (15). Anal. Calcd for $C_{17}H_{21}NO_2Se$: C, 58.29; H, 6.04; N, 4.00. Found: C, 58.35; H, 6.08; N, 4.06.

Ethyl 2-phenyl-5-(phenylseleno)-1,4,5,6-tetrahydro-3-pyridinecarboxylate (12b): mp 111–114 °C; IR ($CHCl_3$) 3440, 1700 cm^{-1}; ^{1}H NMR δ 7.65–7.55 (m, 2 H), 7.4–7.2 (m, 8 H), 4.2 (br s, 1 H), 3.84 (q, 2 H, J = 7.1 Hz), 3.65–3.5 (m, 2 H), 3.4–3.25 (m, 1 H), 3.15 (ddd, 1 H, J = 1.7, 5.0, and 16.7 Hz), 2.65 (dd, 1 H, J = 7.9 and 16.7 Hz), 0.86 (t, 3 H, J = 7.1 Hz); ^{13}C NMR δ 168.1, 153.6, 139.4, 135.1, 129.1, 128.4, 128.0, 127.9, 127.8, 94.4, 58.8, 47.6, 36.2, 30.5, 13.8. Anal. Calcd for $C_{20}H_{21}NO_2Se$: C, 62.18; H, 5.48; N, 3.63. Found: C, 62.10; H, 5.41; N, 3.71.

Methyl 2-ethyl-5-(phenylseleno)-1,4,5,6-tetrahydro-3-pyridinecarboxylate (12c): mp 88–89 °C; ^{1}H NMR δ 7.6–7.5 (m, 2 H), 7.3–7.2 (m, 3 H), 4.3 (br s, 1 H), 3.6 (s, 3 H), 3.5–3.4 (m, 2 H), 3.3–3.1 (m, 1 H), 3.0–2.8 (m, 1 H), 2.7–2.4 (m, 3 H), 1.1 (t, 3 H, J = 7.0 Hz); ^{13}C NMR δ 168.2, 158.0, 134.9, 129.0, 128.5, 127.7, 90.6, 50.3, 47.4, 36.2, 30.2, 27.5, 12.9; MS m/z (relative intensity) 325 (9), 169 (11), 168 (100), 166 (15), 157 (5), 136 (20), 108 (40). Anal. Calcd for $C_{15}H_{19}NO_2Se$: C, 55.56; H, 5.91; N, 4.32. Found: C, 55.63; H, 6.00; N, 4.24.

3′-(Methoxycarbonyl)-5′-(phenylseleno)-1′,4′,5′,6′-tetrahydro-3,2′-bipyridine (12e): oil; ^{1}H NMR δ 8.55–8.4 (m, 2 H), 7.65–7.55 (m, 3 H), 7.35–7.2 (m, 4 H), 4.55–4.45 (m, 1 H), 3.7–3.5 (m, 2 H), 3.4 (s, 3 H), 3.4–3.25 (m, 1 H), 3.03 (ddd, 1 H, J = 1.9, 5.1, and 16.8 Hz), 2.65 (dd, 1 H, J = 7.6 and 16.8 Hz); ^{13}C NMR δ 167.8, 150.5, 149.5, 148.7, 135.6, 135.0, 129.1, 128.1, 127.9, 122.6, 95.2, 50.4, 47.5, 35.7, 30.3. Anal. Calcd for $C_{18}H_{18}N_2O_2Se$: C, 57.92; H, 4.86; N, 7.50. Found: C, 57.84; H, 4.80; N, 7.57.

3′-(Methoxycarbonyl)-5′-(phenylseleno)-1′,4′,5′,6′-tetrahydro-4,2′-bipyridine (12f): mp 93–95 °C; ^{1}H NMR δ 8.55 (d, 2 H, J = 5.7 Hz), 7.6–7.5 (m, 2 H), 7.4–7.25 (m, 3 H), 7.2 (d, 2 H, J = 5.7 Hz), 4.3 (s, 1 H), 3.7–3.3 (m, 3 H), 3.4 (s, 3 H), 3.1–2.9 (m, 1 H), 2.8–2.5 (m, 1 H); ^{13}C NMR δ 167.7, 151.0, 149.5, 147.1, 135.2, 129.2, 128.0, 122.8, 96.1, 50.5, 47.4, 35.5, 30.0. Anal. Calcd for $C_{18}H_{18}N_2O_2Se$: C, 57.92; H, 4.86; N, 7.50. Found: C, 57.72; H, 4.69; N, 7.38.

Oxidation Reactions. General Procedure. To a stirred solution of the dihydrooxazole **4f** or of the tetrahydropyridines **8c** and **12b–12h** (1 mmol) in methanol (5 mL) was added hydrogen peroxide (35%, 3 mmol) dropwise at 0 °C. After 15 min the cooling bath was removed, and the mixture was left to reach room temperature and then warmed at 40 °C for 0.5 h. The progress of the reaction was monitored by TLC and/or GC–MS. The mixture was poured into a dilute solution of $NaHCO_3$ and extracted with methylene chloride. The organic layer was dried and evaporated, and the residue was purified by column chromatography on silica gel. The results of these experiments are summarized in Chart I and in Table I. Physical and spectral data of the aromatic products are reported below.

2-(2-Thienyl)pyridine (9): mp 61–62 °C (lit.[18] mp 64 °C); ^{1}H NMR δ 8.6–8.5 (m, 1 H), 7.65–7.62 (m, 2 H), 7.55 (dd, 1 H,

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J = 1.2 and 3.7 Hz), 7.37 (dd, 1 H, J = 1.0 and 5.0 Hz), 7.12–7.03 (m, 2 H); ^{13}C NMR δ 152.7, 149.4, 136.5, 127.9, 127.4, 124.4, 121.7, 118.6; MS m/z (relative intensity) 162 (12), 161 (100), 160 (30), 128 (8), 116 (6), 89 (8), 80 (7), 78 (8).

Ethyl 2-phenyl-3-pyridinecarboxylate (13b): oil;[19] ^{1}H NMR δ 8.77 (dd, 1 H, J = 1.8 and 4.9 Hz), 8.1 (dd, 1 H, J = 1.8 and 7.9 Hz), 7.6–7.5 (m, 2 H), 7.5–7.4 (m, 3 H), 7.34 (dd, 1 H, J = 4.9 and 7.9 Hz), 4.15 (q, 2 H, J = 7.2 Hz), 1.05 (t, 3 H, J = 7.2 Hz); ^{13}C NMR δ 168.0, 158.9, 151.1, 140.3, 137.7, 128.5, 128.0, 127.4, 121.4, 61.4, 13.5; MS m/z (relative intensity) 227 (13), 199 (13), 198 (100), 182 (19), 154 (14), 127 (14), 77 (7).

Methyl 2-ethyl-3-pyridinecarboxylate (13c): oil; ^{1}H NMR δ 8.66 (dd, 1 H, J = 1.8 and 4.8 Hz), 8.15 (dd, 1 H, J = 1.8 and 7.9 Hz), 7.2 (dd, 1 H, J = 4.8 and 7.9 Hz), 3.9 (s, 3 H), 3.2 (q, 2 H, J = 7.5 Hz), 1.3 (t, 3 H, J = 7.5 Hz); ^{13}C NMR δ 166.9, 164.5, 151.8, 138.2, 120.5, 52.1, 30.1, 13.7; MS m/z (relative intensity) 165 (29), 164 (16), 150 (100), 134 (16), 132 (15), 106 (22), 104 (25), 79 (39), 78 (23), 77 (16), 51 (15). Anal. Calcd for $C_9H_{11}NO_2$: C, 65.44; H, 6.71; N, 8.48. Found: C, 65.51; H, 6.63; N, 8.41.

3-(Methoxycarbonyl)-2,2′-bipyridine (13d): mp 53–55 °C; ^{1}H NMR δ 8.7 (dd, 1 H, J = 1.7 and 4.8 Hz), 8.59 (ddd, 1 H, J = 0.9, 1.4, and 4.6 Hz), 8.16 (d, 1 H, J = 7.8 Hz), 7.93 (dd, 1 H, J = 1.7 and 7.8 Hz), 7.8 (dt, 1 H, J = 1.8 and 7.8 Hz), 7.35 (dd, 1 H, J = 4.9 and 7.8 Hz), 7.28 (ddd, 1 H, J = 1.2, 4.9, and 7.6 Hz), 3.8 (s, 3 H); ^{13}C NMR δ 169.1, 156.0, 155.2, 150.1, 148.2, 136.7, 136.5, 128.4, 123.4, 122.5, 52.1; MS m/z (relative intensity) 214 (11), 200 (8), 199 (59), 183 (100), 155 (15), 128 (8), 101 (6), 78 (23), 77 (8), 51 (16), 50 (9). Anal. Calcd for $C_{12}H_{10}N_2O_2$: C, 67.28; H, 4.71; N, 13.08. Found: C, 67.23; H, 4.78; N, 13.01.

3-(Methoxycarbonyl)-2,3′-bipyridine (13e): mp 51–53 °C; ^{1}H NMR δ 8.83 (dd, 1 H, J = 1.7 and 4.8 Hz), 8.72 (d, 1 H, J = 1.9 Hz), 8.67 (dd, 1 H, J = 1.8 and 4.9 Hz), 8.23 (dd, 1 H, J = 1.8 and 7.9 Hz), 7.91 (dt, 1 H, J = 2.0 and 7.9 Hz), 7.42 (dd, 1 H, J = 4.8 and 7.9 Hz), 7.39 (dd, 1 H, J = 4.9 and 7.9 Hz), 3.75 (s, 3 H); ^{13}C NMR δ 167.4, 156.2, 151.8, 149.5, 138.4, 135.9, 126.7, 122.9, 122.3, 52.5; MS m/z (relative intensity) 214 (2), 200 (13), 199 (100), 183 (8), 155 (10), 78 (8). Anal. Calcd for $C_{12}H_{10}N_2O_2$: C, 67.28; H, 4.71; N, 13.08. Found: C, 67.35; H, 4.80; N, 13.01.

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3-(Methoxycarbonyl)-2,4′-bipyridine (13f): mp 85–86 °C; ^{1}H NMR δ 8.8 (dd, 1 H, J = 1.8 and 4.8 Hz), 8.7 (AA′BB′ system, 2 H), 8.2 (dd, 1 H, J = 1.8 and 7.9 Hz), 7.4 (dd, 1 H, J = 4.8 and 7.9 Hz), 7.45 (AA′BB′ system, 2 H), 3.7 (s, 3 H); ^{13}C NMR δ 167.2, 156.5, 151.6, 149.5, 147.6, 138.2, 126.8, 123.0, 122.6, 52.3; MS m/z (relative intensity) 215 (13), 201 (9), 199 (15), 185 (15), 183 (14), 157 (49), 130 (8), 105 (8), 78 (23), 77 (14), 51 (10). Anal. Calcd for $C_{12}H_{10}N_2O_2$: C, 67.28; H, 4.71; N, 13.08. Found: C, 67.35; H, 4.82; N, 12.99.

Methyl 2-[3-(*N*-methylpyrrolyl)]-3-pyridinecarboxylate (13g): oil; ^{1}H NMR δ 8.59 (dd, 1 H, J = 1.8 and 4.8 Hz), 7.8 (dd, 1 H, J = 1.8 and 7.7 Hz), 7.15 (dd, 1 H, J = 1.8 and 1.9 Hz), 7.07 (dd, 1 H, J = 4.8 and 7.7 Hz), 6.57 (dd, 1 H, J = 1.9 and 2.7 Hz), 6.36 (dd, 1 H, J = 1.8 and 2.7 Hz), 3.85 (s, 3 H), 3.65 (s, 3 H); ^{13}C NMR δ 169.0, 150.8, 136.8, 125.2, 123.0, 122.3, 119.3, 109.0, 96.1, 52.3, 36.3; MS m/z (relative intensity) 216 (100), 201 (25), 185 (22), 173 (8), 158 (52), 157 (28), 130 (15), 117 (7), 78 (20). Anal. Calcd for $C_{12}H_{12}N_2O_2$: C, 66.66; H, 5.59; N, 12.96. Found: C, 66.45; H, 5.42; N, 12.86.

3′,3″-Bis(methoxycarbonyl)-2,2′:6,2″-terpyridine (13h): mp 71–74 °C; ^{1}H NMR δ 8.75 (dd, 1 H, J = 1.4 and 4.7 Hz), 8.74 (dd, 1 H, J = 1.4 and 4.7 Hz), 8.2–7.9 (m, 5 H), 7.38 (dd, 1 H, J = 4.8 and 7.8 Hz), 7.37 (dd, 1 H, J = 4.8 and 7.8 Hz), 3.44 (s, 3 H), 3.43 (s, 3 H); ^{13}C NMR δ 168.3, 155.9, 155.4, 150.3, 137.4, 137.3, 128.3, 122.6, 52.0; MS m/z (relative intensity) 349 (7), 348 (8), 319 (54), 318 (53), 302 (56), 276 (11), 259 (26), 233 (100), 129 (45), 102 (18), 78 (10). Anal. Calcd for $C_{19}H_{15}N_3O_4$: C, 65.33; H, 4.33; N, 12.03. Found: C, 65.25; H, 4.41; N, 12.06.

Acknowledgment. Financial support from the CNR, Rome, Progetto Finalizzato "Chimica Fine II", and Ministero dell'Università e della Ricerca Scientifica e Tecnologica, Italy, is gratefully acknowledged.

Supplementary Material Available: Physical and spectral data for compounds **2c**, **2f**, **7b**, **11c**, **11d**, **11f**, **11g**, **11h**, **4f**, **4g**, **12d**, **12g**, **12h**, and **5** (4 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.