trated and extracted with two 50-mL portions of ether. The combined organic layers were washed with water and brine, dried ($MgSO_4$), and concentrated to yield 0.49 g of a yellow liquid (98%): ^1H NMR ($CDCl_3$) δ 9.51 (s, 1 H), 7.15 (AB q, J = 5.9 Hz, 4 H), 3.10 (AB q, J = 17.2 Hz, 2 H), 2.32 (s, 3 H), 2.06 (s, 3 H), 1.57 (s, 3 H). No further purification of this material was attempted.

(*S*)-4-*p*-Tolyl-4-methylcyclopentenone (19). To a stirred solution of 0.49 g (2.40 mmol) of keto aldehyde **18** in 60 mL of dry THF was added 200 μL of 1 M KOH in ethanol. The solution was stirred at room temperature for 1 h and concentrated in vacuo. The residue was dissolved in ether, washed with water and brine, dried ($MgSO_4$), and concentrated. The yellow liquid was purified by column chromatography (EM Merck 7747 silica gel) to yield 0.40 g (90%) of cyclopentenone as a colorless liquid: $[\alpha]^{20}_D$ +114° (c 1.36, EtOH); ^1H NMR ($CDCl_3$) δ 7.66 (d, J = 5.5 Hz, 1 H), 7.15 (s, 4 H), 6.19 (d, J = 5.5 Hz, 1 H), 2.58 (AB q, J = 18.7 Hz, 2 H), 2.33 (s, 3 H), 1.62 (s, 3 H). Spectral properties were identical with those reported for racemic material.[6]

(*S*)-4,5,5-Trimethyl-4-*p*-tolylcyclopentenone (20). A solution of 100 mg (0.54 mmol) of cyclopentenone **19** in 0.3 mL of DMF was added over 0.25 h to a stirred solution of 0.33 g (1.3 mmol) of NaH in 0.3 mL of DMF. The reaction was stirred for 0.5 h, and 0.17 mL (2.70 mmol) MeI was added dropwise. The mixture was stirred for 16 h at 25 °C, and the excess hydride was decomposed by adding 0.30 mL of MeOH. The solution was diluted with 30 mL of ether, washed with water and brine, dried ($MgSO_4$), and concentrated. The yellow residue was purified by column chromatography on silica gel to yield 54 mg (48%) of dimethylated cyclopentenone **20**: ^1H NMR ($CDCl_3$) δ 7.75 (d, J = 5.9 Hz, 1 H), 7.10 (AB q, J = 9.3 Hz, 4 H), 6.22 (d, J = 5.9 Hz, 1 H), 2.34 (s, 3 H), 1.45 (s, 3 H), 1.19 (s, 3 H), 0.53 (s, 3 H). Spectral properties were identical with those reported for racemic material.[6]

(*S*)-(−)-α-Cuparenone (12). A stirred solution of 54 mg (0.25 mmol) cyclopentenone **20** and 10 mg of 10% Pd/C in 2 mL of ethyl acetate was stirred under 3 atm of hydrogen for 2 h. The solution was then filtered through a 1-in. plug of silica gel and concentrated to yield 47 mg (87%) of α-cuparenone: $[\alpha]^{20}_D$ −166° (c 0.200, $CHCl_3$) (lit.[4,7] $[\alpha]_D$ −170°); ^1H NMR ($CDCl_3$) δ 7.22 (AB q, J = 8.4 Hz, 4 H), 2.60 (m, 1 H), 2.40 (m, 2 H), 2.32 (s, 3 H), 1.90 (m, 1 H), 1.26 (s, 3 H), 1.17 (s, 3 H), 0.61 (s, 3 H); ^{13}C NMR ($CDCl_3$) δ 221.6, 142.2, 135.8, 128.9, 126.4, 53.2, 48.5, 33.7, 30.0, 25.4, 22.1, 20.7, 18.6. Anal. Calcd for $C_{15}H_{20}O$: C, 83.33; H, 9.33. Found: C, 83.18; H, 9.48.

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An Efficient and Novel Approach to the Synthesis of 3,4,5,6-Tetraphenyl-2(1*H*)-pyridinone

Robert L. Eagan[1] and Michael A. Ogliaruso*

Department of Chemistry, Virginia Polytechnic Institute and State University, Blacksburg, Virginia 24061

James P. Springer

Merck, Sharp and Dohme Research Laboratories, Rahway, New Jersey 07065

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The reaction of tetracyclone (**1a**) with sodium azide under acidic conditions was found to give 3,4,5,6-tetraphenyl-2(1*H*)-pyridinone (**2a**) in 90% isolated yield. The reaction does not proceed by a Schmitt mechanism. By adjusting the reaction conditions, the intermediate 1,5,7,8-tetraphenyl-2,3,4-triazabicyclo[3.3.0]octa-2,7-dien-6-one (**3a**) was isolated in 94% yield. Treatment of **3a** under acidic conditions resulted in loss of nitrogen and formation of **2a**. When the acidification was carried out in methanol, 2-methoxy-3,4,5,6-tetraphenylpyridine (**4a**) was also produced. Treatment of the N-methyl intermediate (**6a**) under similar conditions produced 1-methyl-3,4,5,6-tetraphenyl-2(1*H*)-pyridinone (**5a**) in good yield. Reaction of 2-methyl-3,4,5-triphenyl-2,4-cyclopentadien-1-one (**1b**) with sodium azide gave similar results.

Tetraphenyl-2(1*H*)-pyridinone (**2a**) has been prepared by a variety of methods. Wajon and Arens[2] described the condensation of α-benzoylbenzyl cyanide with benzyl phenyl ketone under acidic conditions to produce the title compound in 50% yield. Jagt and Van Leusen[3] reported the Diels–Alder cycloaddition of tosyl cyanide to tetracyclone, followed by base hydrolysis to yield the pyridinone in 69% yield. Abramovitch and Knaus[4] described the addition of singlet sulfonyl nitrene to tetracyclone which resulted in a 13% yield of the pyridinone, while Martin and Bauer[5] obtained the title compound in 76% yield by the Diels–Alder addition of trichloroethyl cyanate to tetracyclone, with subsequent hydrolysis. The above syntheses of tetraphenylpyridinone all suffer from either poor yields, multiple steps, or the use of exotic reagents. In this paper we report a highly efficient, one-pot synthesis of tetraphenylpyridinone using readily available and relatively inexpensive starting materials.

Results and Discussion

Treatment of tetracyclone (**1a**) with sodium azide under acidic conditions resulted in a 90% isolated yield of the corresponding tetraphenylpyridinone (**2a**) (Scheme I). However, when the reaction was run under milder conditions (i.e., lower temperature, shorter reaction time), the bicyclic triazoline **3a** was obtained in 94% yield. Subsequent acidification of **3a** resulted in elimination of nitrogen and formation of the pyridinone **2a**. Thus it appears that **3a** is an intermediate in the formation of **2a** and that the reaction does not take place by a classical Schmitt reaction pathway.[6]

(1) Presented at the 36th Southeastern Regional Meeting of the American Chemical Society, Raleigh, NC, October 25, 1984.
(2) Wajon, J. F. M.; Arens, J. F. *Recl. Trav. Chim. Pays-Bas* **1957**, *76*, 65.
(3) Jagt, J. C.; Van Leusen, A. M. *Recl. Trav. Chim. Pays-Bas* **1973**, *92*, 1343.
(4) Abramovitch, R. A.; Knaus, G. N. *J. Org. Chem.* **1975**, *40*, 883.
(5) Martin, D.; Bauer, M. *Z. Chem.* **1980**, *20*, 53.
(6) March, J. *Advanced Organic Chemistry*; McGraw-Hill, Inc.: New York, 1977; pp 1006–1008.

0022-3263/86/1951-1544$01.50/0

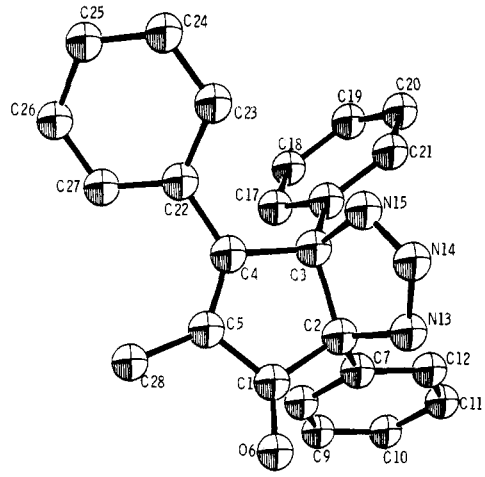
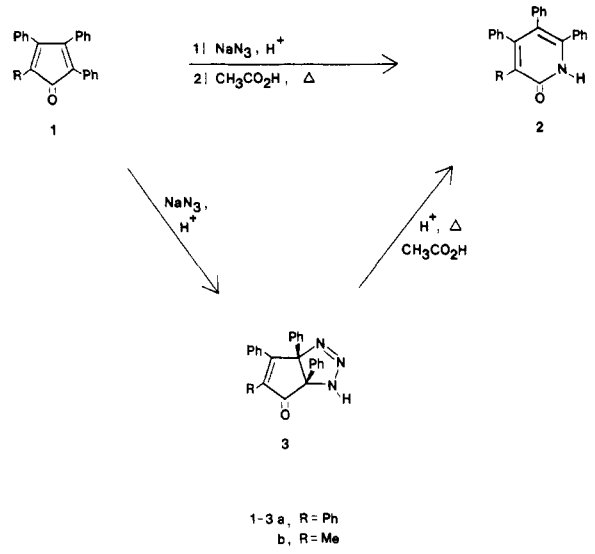


Figure 1. Computer-generated drawing of **3b** derived from the X-ray coordinates with hydrogen omitted for clarity.

Scheme I



We postulate that the formation of **3a** results from an initial acid-catalyzed Michael addition of sodium azide to tetracyclone (**1a**), followed by protonation of the terminal nitrogen of the azide functional group and cyclization via the enol form, to generate the bicyclic structure.

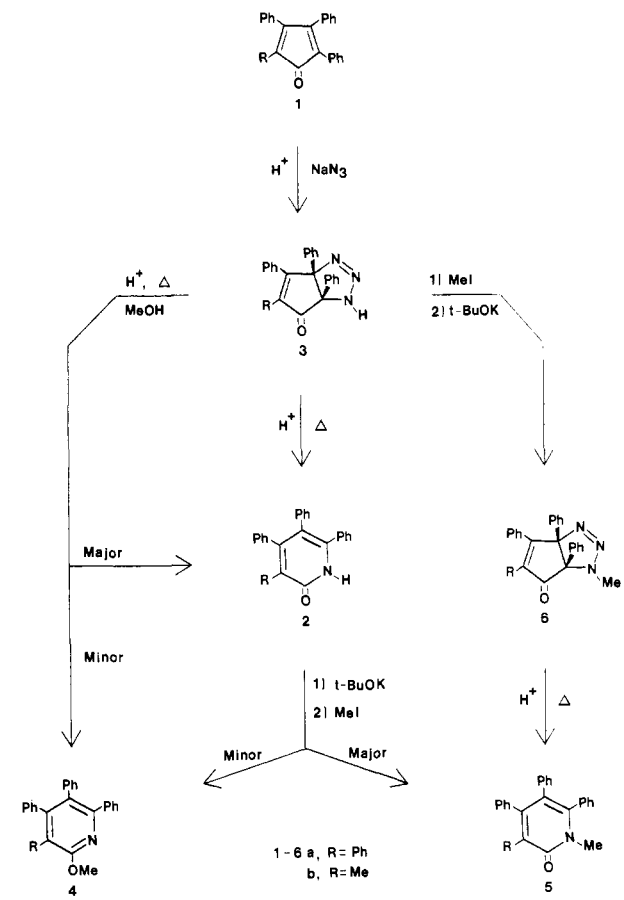
It is important to note that in the unsymmetrical case **1b** (Scheme I), the only bicyclic triazoline compound obtained was the one in which addition and cyclization occurred to the side of the ring that contained the phenyl ring alpha to the carbonyl group. It appears that the phenyl ring at this position directs the addition to this side of the ring and stabilizes the enol structure sufficiently to allow cyclization to occur. The structure of the bicyclic triazoline **3a** was assigned on the basis of its spectral data and was confirmed by X-ray crystallographic analysis (Figure 1).

There are many examples in the literature regarding the Michael addition of hydrazoic acid to α,β-unsaturated carbonyl compounds.[7] However, since the intermediate 1,4-addition product was not observed in this case, the possibility of a 1,3-dipolar cycloaddition[8] cannot be totally excluded.

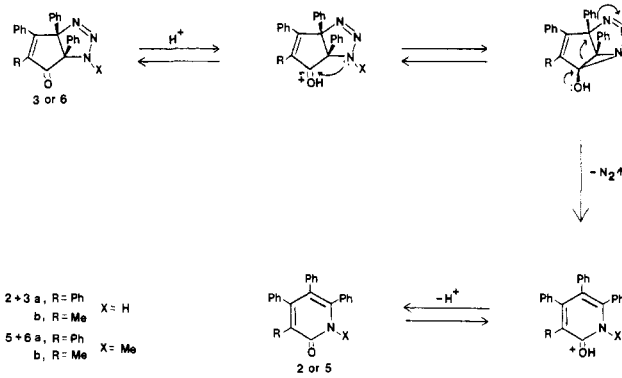
(7) For a review, see: Biffin, Miller, and Paul In *The Chemistry of Azido Group*; Patai, E., Ed., Interscience Publishers: New York, 1971; pp 122–130.

(8) For a review, see: Povarov, *Russ. Chem. Rev.* **1965**, *34*, 639–656.

Scheme II



Scheme III



As shown in Scheme II, acidification of the bicyclic triazoline structure **3** results in loss of nitrogen accompanied by ring enlargement to the pyridinone structure **2**. However, when the acidification takes place in methanol, an additional product is formed along with the expected pyridinone **2**. This product was identified as the appropriately substituted 2-methoxypyridinone derivative **4** which was produced in about 30–35% yield from either **3a** or **3b**.

The bicyclic triazoline system **3** was derivatized to its N-methyl form **6** in order to obtain a more soluble species for spectroscopic examination. Treatment of **3** with methyl iodide followed by the addition of base resulted in acceptable yields of the desired N-methyl derivative **6**. As expected, acidification of this compound resulted in the elimination of nitrogen with formation of the *N*-methylpyridinone **5**.

The formation of **2** from **3** and **5** from **6** can be visualized as represented in Scheme III. The conversion probably proceeds by an acid-catalyzed intramolecular attack by the

amine nitrogen to produce the highly strained tricyclic ring system which can then rearrange with loss of nitrogen to generate the substituted pyridinone system **2** and **5**.

The formation of the substituted 2-methoxypyridine **4** by acidification of **3** in methanol can be explained in a slightly different manner. Since treatment of the substituted pyridinone **2** under the identical resaction conditions did not produce any of the 2-methoxy derivative **4**, it appears that this product did not arise from **2** and, therefore, **4** and **2** must be derived from a common intermediate, or more likely, by separate reaction pathways.

A possible mechanism for the conversion of **3** to **4** can be envisioned as follows. Formation of the hemiketal of **3**, followed by an intramolecular backside displacement of water by the amine nitrogen generates the unstable tricyclic ether. This compound can then spontaneously and irreversibly rearrange with elimination of nitrogen to form the substituted 2-methoxypyridine **4**.

The methoxypyridine **4** and *N*-methylpyridinone **5** were also prepared by treatment of the pyridinone **2** with potassium *tert*-butoxide followed by quenching with methyl iodide (Scheme II). In each case (**2a** or **2b**), the N-methyl derivative (**5a** or **5b**) was generated as the major product. In both cases, the ratio of **5** to **4** was on the order of 65:35.

Experimental Section

General Procedures. Melting points were determined on a Thomas-Hoover melting point apparatus and are uncorrected. The infrared spectra were recorded on a Perkin-Elmer 710B spectrophotometer using NaCl salt windows. ^{1}H NMR spectra were determined at 90 MHz on a Varian EM-390, while ^{13}C NMR spectra were recorded at 67.5 MHz on a Bruker WP-270SY spectrometer. NMR chemical shifts are reported in parts per million (ppm) relative to tetramethylsilane (Me_4Si) as the internal standard. Mass spectra were obtained on a Varian MAT 112 or VG 7070E-HF mass spectrometer at 70 eV. Elemental analyses were performed by Multi Chem Laboratories, Inc., Lowell, MA. All solvents were purified by standard Methods[10] prior to use. Tetracyclone (**1a**) was prepared by the method of Johnson and Grummitt[11] and was used without further purification. 2-Methyl-3,4,5-triphenyl-2,4-cyclopentadien-1-one (**1b**) was prepared by the method of Allen and VanAllan[12] and used without further purification.

Crystallographic Analysis. Suitable crystals of **3b** ($C_{19}H_{24}N_3O$) for X-ray diffraction studies were formed from acetone and have a space group symmetry of $P2_1$ and cell constants of a = 11.936 (2) Å, b = 6.795 (3) Å, c = 12.005 (2) Å, and β = 99.92 (1) for Z = 2 and a calculated density of 1.265 g/cm^3. Of the 1413 reflections measured with an automatic four-circle diffractometer equipped with Cu radiation, 1342 were observed ($I \geq 3\sigma I$). The structure was solved with a multisolution tangent formula approach and difference Fourier analysis and refined by using full-matrix least-squares techniques.[9] Hydrogens were assigned isotopic temperature factors corresponding to their attached atoms. The function $\sum w(|F_o| - |F_c|)^2$ with $w = 1/(\sigma F_o)^2$ was minimized to give an unweighted residual of 0.037. No abnormally short intermolecular contacts were noted. Table I, II, and III containing the final fractional coordinates, temperature parameters, bond distances, and bond angles are available as supplementary material. Figure 1 is a computer generated perspective drawing of **3b** from the final X-ray coordinates showing the relative stereochemistry.

(9) The following library of crystallographic programs werre used: MULTAN 80, Main et. al, University of York, York, England, 1980; ORTEP-II, Johnson, C. K., Oak Ridge National Laboratory, Oak Ridge, TN, 1970; SDP+V1.1, Okaya, Y. et al., B. A. Frenz and Associates, College Station, TX, 1984.

(10) Gordon, A. J.; Ford, R. A. *The Chemist's Companion*; John Wiley and Sons: New York, 1972; pp 430–436.

(11) Johnson, J. R.; Grummitt, O. *Organic Syntheses*; Wiley: New York, 1953; Collect. Vol. III, p 806.

(12) Allen, C. F. H.; VanAllan, J. A. *J. Am. Chem. Soc.* **1950**, *72*, 5165.

7-Substituted-1,5,8-triphenyl-2,3,4-triazabicyclo[3.3.0]-octa-2,7-dien-6-one (3a, R = Ph; 3b, R = Me). Into a 250-mL, round-bottomed flask equipped with a magnetic stirring bar were placed 2.6 mmol of **1** (1.0 g of **1a**, 0.84 g of **1b**), 1.2 g (18.5 mmol) of sodium azide, 75 mL of *N*,*N*-dimethylformamide (DMF), and 0.3 mL of concentrated H_2SO_4. The mixture was gradually heated with stirring to 80 °C during which time the color changed from dark purple (**1a**) or dark reddish-brown (**1b**) to a cloudy white or light yellow color (ca. 25 min for **1a**, 35 min for **1b**), and at this time 60 mL of benzene were then added to the flask. The solution was cooled in an ice–water bath and then transferred to a separatory funnel containing 75 mL of water. The benzene layer was separatory funnel containing 75 mL of water. The benzene layer was immediately separated and dried over anhydrous magnesium sulfate. The benzene was removed by a rotary evaporator, and the residual DMF was removed under vacuum. The resulting off-white residue was dissolved in 75 mL of hot benzene followed by the addition of 85 mL of petroleum ether (bp 30–60 °C). This afforded 1.0 g (2.34 mmol, 90%) of **3a** or 0.77 g (2.39 mmol, 92%) of **3b** in the form of white cotton-like fibers. **3a**: mp 183–184 °C dec; IR ($CHCl_3$) 3455 cm^{-1} (NH), 1725 cm^{-1} (CO); ^{1}H NMR ($CDCl_3$) δ 8.8 (s, 1 H), 6.8–8.2 (m, 20 H); MS, m/e (relative intensity) 399 ($M^+ - 28$). Anal. Calcd for $C_{29}H_{21}N_3O$: C, 81.48; H, 4.95; N, 9.83. Found: C, 81.53; H, 5.13; N, 9.44. **3b**: mp 183.5–184 °C dec; IR ($CDCl_3$) 3470 cm^{-1} (NH), 1730 cm^{-1} (CO); ^{1}H NMR ($CDCl_3$) δ 8.7 (s, 1 H), 6.6–7.9 (m, 15 H), 2.2 (s, 3 H); MS, m/e (relative intensity) 337 ($M^+ - 28$). Anal. Calcd for $C_{24}H_{19}N_3O$: C, 78.88; H, 5.24; N, 11.50. Found: C, 79.11; H, 5.44; N, 11.41.

3-Substituted-4,5,6-triphenyl-2(1*H*)-pyridinone (2a, R = Ph; 2b, R = Me) from 1. Into a 250-mL, three-necked flask equipped with a magnetic stirring bar and reflux condenser were placed 2.6 mmol of **1** (1.0 g of **1a**, 0.84 g of **1b**), 1.2 g (18.5 mmol) of sodium azide, 75 mL of DMF, and 0.3 mL of concentrated H_2SO_4. The micture was gradually heated with stirring to 80 °C during which time the color changed from dark purple (**1a**) or dark reddish-brown (**1b**) to a cloudy white or light yellow color (ca. 25 min for **1a**, 25 min for **1b**). At this time 50 mL of glacial acetic acid was then added to the flask, and the solution was refluxed for 15 min. The solution was then transferred to a 500-mL Erlenmeyer flask, and 200 mL of water were added. After having been cooled to room temperature, the flask was placed in an ice–water bath. Suction filtration followed by washing with 15 mL of cold absolute ethanol afforded 0.94 g (2.35 mmol, 90%) of **2a** or 0.61 g (1.81 mmol, 70%) of **2b** in the form of a white powder. **2a**: mp 272–273 °C (lit. mp 271–273 °C,[2] 262–267 °C,[3] 273–275 °C,[4] 264–267 °C[5]); ^{1}H NMR ($CDCl_3$) δ 11.6 (br s, 1 H), 6.5–7.5 (m, 20 H); MS, m/e 399. Anal. Calcd for $C_{29}H_{21}NO$: C, 87.19; H, 5.30; N, 3.51. Found: C, 87.26; H, 5.52; N, 3.23. **2b**: mp 297–299 °C; IR ($CDCl_3$) 3440 cm^{-1} (NH), 1650 cm^{-1} (CO); ^{1}H NMR ($CDCl_3$) δ 10.5 (br s, 1 H), 6.6–7.4 (m, 15 H), 1.98 (s, 3 H); MS, m/e 337. Anal. Calcd for $C_{24}H_{19}NO$: C, 85.43; H, 5.68; N, 4.15. Found: C, 85.46; H, 5.77; N, 4.11.

3-Substituted-4,5,6-triphenyl-2(1*H*)-pyridinone (2a, R = Ph; 2b, R = Me) from 3. Into a 50-mL, round-bottomed flask equipped with a magnetic stirring bar and reflux condenser were placed 1.2 mmol of **3** (0.51 g of **3a**, 0.44 g of **3b**) and 25 mL of glacial acetic acid. At this time, 0.1 mL of concentrated H_2SO_4 was slowly added at room temperature. The resulting yellow solution was refluxed for 20 min, cooled slightly, and then transferred to a 250-mL Erlenmeyer flask. Upon addition of 100 mL of water, the flask was placed in an ice–water bath. Suction filtration followed by washing with 20 mL of cold absolute ethanol afforded 0.39 g (0.98 mmol, 82%) of **2a** or 0.31 g (0.92 mmol, 77%) of **2b** in the form of a white powder. Both compounds were identical with the pyridinones described above.

2-Methoxy-3,4,5,6-tetraphenylpyridine (4a) and 1-Methyl-3,4,5,6-tetraphenyl-2(1*H*)-pyridinone (5a) from 2a. Into a 250-mL, round-bottomed flasked equipped with a magnetic stirring bar were placed 1.16 g (2.9 mmol) of **2a**, 100 mL of DMF, and 0.5 g (4.5 mmol) of potassium *tert*-butoxide. The solution was stirred for 5 min at room temperature and then placed in an ice–water bath for an additional 10 min. To the light yellow solution was added 1 mL (2.3 g, 16 mmol) of methyl iodide. The resulting colorless solution was stirred for 10 min followed by the addition of 75 mL of benzene and then transferred to a separatory

funnel containing 100 mL of water. The benzene layer was washed 3 times with 100 mL of water and dried over anhydrous magnesium sulfate. Removal of the solvent on a rotary evaporator afforded an oil which was redissolved in 8 mL of hot benzene and treated with 15 mL of petroleum ether (bp 30–60 °C). This resulted in 0.83 g (2.0 mmol, 69%) of white crystals which contained 92% of the N-methyl derivative according to ^{1}H NMR integration. Recrystallization from benzene/petroleum ether (1:2) afforded the pure *N*-methylpyridinone **5a**: mp 230–232 °C (lit.[13] mp 232–234 °C); IR ($CDCl_3$) 1645 cm^{-1} (CO); ^{1}H NMR ($CDCl_3$) δ 6.4–7.5 (m, 20 H), 3.38 (s, 3 H); MS, *m/e* 413. Anal. Calcd for $C_{30}H_{23}NO$: C, 87.14; H, 5.61; N, 3.39. Found: C, 87.36; H, 5.80; N, 3.23. Evaporation of the original mother liquor afforded 0.25 g (0.60 mmol, 21%) of white crystals containing 85% of the O-methyl isomer (by ^{1}H NMR integration). Recrystallization from benzene/petroleum ether (bp 30–60 °C) (1:2) produced the pure methoxypyridine **4a**: mp 196.5–197.5 °C; IR ($CDCl_3$) 1245 and 1030 cm^{-1} (C–O–C); ^{1}H NMR ($CDCl_3$) δ 6.5–7.5 (m, 20 H), 3.98 (s, 3 H); MS, *m/e* 413. Anal. Calcd for $C_{30}H_{23}NO$: C, 87.14; H, 5.61; N, 3.39. Found: C, 87.22; H, 5.74; N, 3.32.

2-Methoxy-3-methyl-4,5,6-triphenylpyridine (4b) and 1,3-Dimethyl-4,5,6-triphenyl-2(1*H*)-pyridinone (5b) from 2b. Into a 50-mL, round-bottomed flask equipped with a magnetic stirring bar was placed 0.67 g (2.0 mmol) of **2b**, 35 mL of DMF, and 0.3 g (2.7 mmol) of potassium *tert*-butoxide. The solution was stirred for 10 min at room temperature then placed in an ice-water bath for an additional 10 min. To the pale yellow solution was added 1 mL (2.3 g, 16 mmol) of methyl iodide. The resulting colorless solution was stirred for 10 min followed by the addition of 50 mL of benzene and then transferred to a separatory funnel containing 50 mL of water. The benzene layer was washed 3 times with 50 mL of water and dried over anhydrous magnesium sulfate. Removal of the solvent on a rotary evaporator afforded a white solid which contained 75% of the N-methyl derivative and 25% of the O-methyl isomer (^{1}H NMR integration). The solid was redissolved in 6 mL of hot benzene then treated with 12 mL of petroleum ether (bp 30–60 °C). This afforded 0.39 g (1.1 mmol, 55%) of **5b** in the form of white crystals: mp 221–223 °C; IR ($CDCl_3$) 1645 cm^{-1} (CO); ^{1}H NMR ($CDCl_3$) δ 6.4–7.4 (m, 15 H), 3.37 (s, 3 H), 2.06 (s, 3 H); MS, *m/e* 351. Anal. Calcd for $C_{25}H_{21}NO$: C, 85.44; H, 6.02; N, 3.99. Found: C, 85.76; H, 6.19; N, 3.97. Evaporation of the mother liquor afforded a solid which was recrystallized from absolute ethanol to yield 0.07 g (0.2 mmol, 10%) of **4b** in the form of clear crystals: mp 156–157 °C; IR ($CDCl_3$) 1255 and 1035 cm^{-1} (C–O–C); ^{1}H NMR ($CDCl_3$) δ 6.5–7.4 (m, 15 H), 4.07 (s, 3 H), 2.03 (s, 3 H); MS, *m/e* 351. Anal. Calcd for $C_{25}H_{21}NO$: C, 85.44; H, 6.02; N, 3.99. Found: C, 85.40; H, 6.16; N, 3.93.

3,4,5,6-Tetraphenyl-2(1*H*)-pyridinone (2a) and 2-Methoxy-3,4,5,6-tetraphenylpyridine (4a) from 3a. Into a 100-mL, three-necked flask equipped with a magnetic stirring bar, reflux condenser, and fritted-glass gas inlet tube were placed 0.86 g (2.0 mmol) of **3a** and 50 mL of anhydrous methanol. The flask was gently warmed (ca. 40 °C), and HCl gas (generated in a second flask by the addition of H_2SO_4 to NaCl) was bubbled into the solution for 10 min. After refluxing for 30 min, the solvent was removed on a rotary evaporator. The residue was redissolved in 20 mL of hot benzene, and 25 mL of petroleum ether (bp 30–60 °C) was then added. This afforded 0.54 g (1.4 mmol, 70%) of **2a** in the form of a white powder. This compound was identical in all respects with the tetraphenylpyridinone described above. The mother liquor was removed on a rotary evaporator, and the residue was recrystallized in 10 mL of methanol. Cooling to 0 °C afforded 0.14 g (0.34 mmol, 17%) of **4a** in the form of white crystals which were identical in all respects with the 2-methoxytetraphenylpyridine described above.

3-Methyl-4,5,6-triphenyl-2(1*H*)-pyridinone (2b) and 2-Methoxy-3-methyl-4,5,6-triphenylpyridine (4b) from 3b. Into a 100-mL, three-necked flask equipped with a magnetic stirring bar, reflux condenser, and fritted-glass gas inlet tube were placed 1.0 g (2.7 mmol) of **3b** and 50 mL of anhydrous methanol. The flask was gently warmed (ca. 50 °C), and HCl gas (generated in a second flask by addition of H_2SO_4 to NaCl) was bubbled into the solution for 20 min. After refluxing for 10 additional min, 50 mL of benzene was added, and the contents of the flask were transferred to a separatory funnel containing 200 mL of water and 150 mL of benzene. The benzene layer was washed once with 200 mL of water and dried over anhydrous magnesium sulfate. Removal of the solvent on a rotary evaporator afforded a pale yellow solid which contained about 65% of the pyridinone **2b** and 35% of the methoxypyridine **4b** (^{1}H NMR integration). Dissolution of the crude material in 60 mL of hot benzene and addition of 65 mL of petroleum ether (bp 30–60 °C) followed by suction filtration afforded 0.51 g (1.5 mmol, 56%) of **2b** in the form of white crystals. This compound was identical in all respects with the methyltriphenylpyridinone described above. Evaporation of the mother liquor afforded an oil which was recrystallized from absolute ethanol to yield 0.20 g (0.57 mmol, 21%) of **4b** in the form of white crystals which were identical in all respects with the methoxypyridine described above.

7-Substituted-4-methyl-1,5,8-triphenyl-2,3,4-triazabicyclo[3.3.0]octa-2,7-dien-6-one (6a, R = Ph; 6b, R = Me). Into a 100-mL, round-bottomed flask equipped with a magnetic stirring bar were placed 2.2 mmol of **3** (0.94 g of **3a** or 0.80 g of **3b**), 35 mL of benzene, and 35 mL of dimethyl sulfoxide (Me_2SO). After the solid had dissolved, 1 mL (2.3 g, 16 mmol) of methyl iodide was added, and the solution was stirred for 5–10 min. At this time, 0.4 g (3.6 mmol) of potassium *tert*-butoxide was added to the flask. The solution was stirred for 45 min and then transferred to a separatory funnel containing 50 mL of water. The benzene layer was separated, washed 2 times with 50 mL of water and dried over anhydrous magnesium sulfate. Removal of the solvent on a rotary evaporator afforded a pale yellow residue which was dissolved in 50 mL of hot benzene and treated with 60 mL of petroleum ether (bp 30–60 °C). Suction filtration resulted in recovery of unreacted starting material **3** (**3a**, 0.43 g, 1.0 mmol, 45%; **3b**, 0.44 g, 1.2 mmol, 55%). Evaporation of the mother liquor on a rotary evaporator afforded an off-white residue which was recrystallized from methanol to produce the desired N-methyl derivative **6**. **6a** (0.34 g, 0.77 mmol, 35%): mp 186–187 °C dec; IR ($CDCl_3$) 1720 cm^{-1} (CO); ^{1}H NMR ($CDCl_3$) δ 6.5–7.8 (m, 20 H), 3.34 (s, 1 H); ^{13}C NMR ($CDCl_3$) δ 199.3 (CO), 33.3 (CH_3); MS, *m/e* (relative intensity) 413 ($M^+ - 28$). Anal. Calcd for $C_{30}H_{23}N_3O$: C, 81.61; H, 5.25; N, 9.52. Found: C, 81.47; H, 5.40, N, 9.28. **6b**: (0.28 g, 0.74 mmol, 34%); mp 185–187 °C dec; IR ($CDCl_3$) 1725 cm^{-1} (CO); ^{1}H NMR ($CDCl_3$) δ 6.6–7.9 (m, 15 H), 3.33 (s, 3 H), 2.18 (s, 3 H); MS, *m/e* (relative intensity) 351 ($M^+ - 28$). Anal. Calcd for $C_{25}H_{21}N_3O$: C, 79.13; H, 5.58; N, 11.07. Found: C, 79.01; H, 5.60; N, 10.65.

3-Substituted-1-methyl-4,5,6-triphenyl-2(1*H*)-pyridinone (5a, R = Ph; 5b, R = Me) from 6. Into a 100-mL, three-necked flask equipped with a magnetic stirring bar, reflux condenser, and a fritted-glass gas inlet tube were placed 0.55 mmol of **6** (0.24 g of **6a**, 0.21 g of **6b**) and 60 mL of methanol. The flask was gently warmed (ca. 50 °C), and HCl gas (generated in a second flask by the addition of H_2SO_4 to NaCl) was bubbled into the solution for 50 min. After refluxing for an additional hour, the solvent was removed on a rotary evaporatory. The residue was redissolved in 30 mL of benzene, placed in a separatory funnel, washed 3 times with 25 mL of water and dried over anhydrous magnesium sulfate. Removal of the benzene on a rotary evaporator afforded a white solid which was recrystallized from 3 mL of hot benzene and 8 mL of petroleum ether. This afforded 0.18 g (0.44 mmol, 80%) of **5a** or 0.14 g (0.40 mmol, 73%) of **5b** in the form of the white crystals. Both compounds were identical in all respects with the *N*-methylpyridinones described above.

Registry No. **1a**, 479-33-4; **1b**, 33535-80-7; **2a**, 51954-59-7; **2b**, 101032-76-2; **3a**, 101032-75-1; **3b**, 101054-49-3; **4a**, 101032-77-3; **4b**, 101032-79-5; **5a**, 62557-82-8; **5b**, 101032-78-4; **6a**, 101032-80-8; **6b**, 101032-81-9.

Supplementary Material Available: Tables of the atomic positions, thermal parameters, bond distances, and bond angles for **3b** (5 pages). Ordering information is given on any current masthead page.

(13) Hons, P.; Yamazaki, H. *Synthesis* **1977**, 50.