benzaldimine ($PhCHNCH_2Ph$) are present in the mixture; this observation is completely in agreement with the report of Hutchins et al.[1] Thus, it appears that in this case, no (phenylazo)alkane **1a** is produced. Other primary amines that do not give (phenylazo)alkanes are *tert*-butylamine and isopropylamine. The former amine does not react with nitrosobenzene at ambient temperature, while the latter reduces nitrosobenzene to only azoxybenzene and a trace of aniline.

A mechanism proposed for the reactions of nitrosobenzene with primary amines is depicted in Scheme I and involves an initial nucleophilic attack by alkylamine on nitrosobenzene to afford **4**, followed by elimination of water from **4** to produce the corresponding phenylazoalkane. Intermediate **4**[7] may also cleave to produce phenylhydroxylamine (**5**) and imine **6**. Condensation of **5** with nitrosobenzene gives azoxybenzene. As shown in entries 6 and 7 of Table I, a sharp decrease in the ratio of **1e** to azoxybenzene is obtained upon increasing the amount of *n*-butylamine employed. The result appears to indicate that step iii in the scheme is a base-catalyzed process similar to an E2 elimination reaction. In addition to the concentration of amines employed, the substituents on C_β of **4** can also influence the rates of steps ii and iii, with the latter expected to be more affected. The product distributions revealed in Table I show that the relative amount of azoxybenzene to (phenylazo)alkane is a function of the amine and increases in the order $CH_3NH_2 < CH_3CH_2NH_2 \approx CH_3CH_2CH_2NH_2 \approx CH_3CH_2CH_2CH_2NH_2 < (CH_3)_2CHNH_2$. These results clearly demonstrate that replacement of one or two hydrogens at C_β of **4** by alkyl groups promotes the formation of phenylhydroxylamine, consistent with an E2 mechanism of Saytzeff type[8] for step iii. While the ratio of (phenylazo)alkane to azoxybenzene is governed by the relative rates of steps ii and iii, neither one is the rate-determining step for the overall reaction in view of the fact that ^1H NMR monitoring of the reaction solution shows that only the reactants and the final products, (phenylazo)alkane and azoxybenzene, are present during the reaction. The presence of reactants nitrosobenzene and alkylamine indicates that the slowest step of the entire reaction is the attack of alkylamine on nitrosobenzene, i.e., step i.

Experimental Section

General Procedures. ^1H and ^{13}C NMR spectra were recorded on a JEOL FX-100 spectrometer, IR spectra were recorded on a JASCO A-100 spectrometer, and mass spectra were obtained on a JEOL JMS-D100 mass spectrometer. Methylamine, ethylamine, (Merck), *n*-propylamine (Aldrich), *n*-butylamine, (Fluka), and nitrosobenzene (Tokyo Kasei) were used as purchased.

Isolation of (Phenylazo)methane (1b). To 1.07 g (10.0 mmol) of nitrosobenzene in 25 mL of ether was added 0.310 g (10.0 mmol) of methylamine (35% in water). The mixture was stirred at ambient temperature for 24 h and then dried over magnesium sulfate. Evaporation of the solvent followed by vacuum distillation at 45 °C and 3 torr gave 0.471 g (39%) of a pale yellow liquid. Analysis of the original mixture after solvent removal by ^1H NMR spectroscopy showed the yield of **1b** was 76%. Spectral data for the isolated product: ^1H NMR ($CDCl_3$) δ 3.98 (s, 3 H), 7.36 (m, 3 H), 7.64 (d of d, J = 8, 2 Hz, 2 H); ^1H NMR (C_6D_6) δ 3.72 (s, 3 H), 7.08 (m, 3 H), 7.76 (d of d, J = 8, 2 Hz, 2 H); ^{13}C NMR ($CDCl_3$) δ 57.1 (q), 121.8 (d), 128.7 (d), 130.1 (d), 151.8 (s); MS, m/e 120 (M^+), 105 ($M^+ - CH_3$), 77 ($M^+ - CH_3 - N{=}N$). Anal. Calcd for $C_7H_8N_2$: C, 69.97; H, 6.71; N, 23.31. Found: C, 69.82; H, 6.78; N, 23.41.

(Phenylazo)ethane (1c). To 1.07 g (10.0 mmol) of nitrosobenzene in 25 mL of chloroform was added 0.45 g (10.0 mmol) of ethylamine (70% in water). The mixture was stirred at ambient temperature for 36 h. The desired product was then isolated by following the method for phenylazomethane. Vacuum distillation was conducted at 50 °C and 3 torr. Analytical data: yield, 30%; yield determined by ^1H NMR spectroscopy, 52%; ^1H NMR ($CDCl_3$) δ 1.42 (t, J = 6 Hz, 3 H), 4.06 (q, J = 6 Hz, 2 H), 7.38 (m, 3 H), 7.62 (d of d, J = 8, 2 Hz, 2 H); ^1H NMR (C_6D_6) δ 1.20 (t, J = 6 Hz, 3 H), 3.92 (q, J = 6 Hz, 2 H), 7.04 (m, 3 H), 7.76 (d of d, J = 8, 2 Hz, 2 H); ^{13}C NMR ($CDCl_3$) δ 12.5 (q), 63.5 (t), 121.8 (d), 128.5 (d), 129.8 (d), 151.8 (s); MS, m/e 134 (M^+), 105, 77. Anal. Calcd for $C_8H_{10}N_2$: C, 71.61; H, 7.51; N, 20.87. Found: C, 71.74; H, 7.51; N, 20.94.

(Phenylazo)-*n*-propane (1d). To 1.07 (10.0 mmol) of nitrosobenzene was added 0.55 g (10.0 mmol) of *n*-propylamine in 25 mL of chloroform. The solution was left at ambient temperature for 35 h. The product was isolated by the same method used for (phenylazo)methane. Vacuum distillation was conducted at 50 °C and 1 torr: yield, 30%; yield determined by NMR spectroscopy, 51%; ^1H NMR ($CDCl_3$) δ 1.02 (t, J = 6 Hz, 3 H), 1.92 (m, J = 6 Hz, 2 H), 4.00 (t, J = 6 Hz, 2 H), 7.36 (m, 3 H), 7.62 (d of d, J = 8, 2 Hz, 2 H); ^1H NMR (C_6D_6) δ 0.8 (t, J = 6 Hz, 3 H), 1.78 (m, J = 6 Hz, 2 H), 3.92 (t, J = 6 Hz, 2 H), 7.08 (m, 3 H), 7.80 (d of d, J = 8, 2 Hz, 2 H); ^{13}C NMR ($CDCl_3$) δ 12.1 (q), 21.3 (t), 71.2 (t), 121.9 (d), 128.7 (d), 130.0 (d), 151.9 (s); MS, m/e 148 (M^+), 105, 77. Anal. Calcd for $C_9H_{12}N_2$: C, 72.94; H, 8.16; N, 18.90. Found: C, 72.83; H, 8.11; N, 19.08.

(Phenylazo)-*n*-butane (1e). This compound is prepared by a procedure similar to that of (phenylazo)-*n*-propane. Analytical data: yield, 26%; yield determined by NMR spectroscopy, 50%; ^1H NMR ($CDCl_3$) δ 1.00 (t, J = 7 Hz, 3 H), 1.46 (m, J = 7 Hz, 2 H), 1.90 (m, J = 7 Hz, 2 H), 4.06 (t, J = 7 Hz, 2 H), 7.40 (m, 3 H), 7.64 (d of d, J = 8, 2 Hz, 2 H); ^1H NMR (C_6D_6) δ 0.82 (t, J = 7 Hz, 3 H), 1.28 (m, J = 7 Hz, 2 H), 1.76 (m, J = 7 Hz, 2 H), 3.98 (t, J = 7 Hz, 2 H), 7.04 (m, 3 H), 7.80 (d of d, J = 8, 2 Hz, 2 H); ^{13}C NMR ($CDCl_3$) δ 13.8 (q), 20.6 (t), 29.9 (t), 69.1 (t), 121.8 (d), 128.6 (d), 129.8 (d), 151.8 (s); MS, m/e 162 (M^+), 105, 77. Anal. Calcd for $C_{10}H_{14}N_2$: C, 74.03; H, 8.69; N, 17.26. Found: C, 73.85; H, 8.54; N, 17.71.

Acknowledgment. We thank the National Science Council of the Republic of China for support of this work.

Registry No. **1b**, 4406-66-0; **1c**, 935-08-0; **1d**, 84113-60-0; **1e**, 940-55-6; **2**, 495-48-7; ONPh, 586-96-9; $MeNH_2$, 74-89-5; $EtNH_2$, 75-04-7; $PrNH_2$, 107-10-8; $BuNH_2$, 109-73-9; *i*-$PrNH_2$, 75-31-0; $PhCH_2NH_2$, 100-46-9; Me_2NH, 124-40-3; Et_2NH, 109-89-7; aniline, 62-53-3.

(7) **4** was suggested as an intermediate for the formation of phenylhydroxylamines by Hutchins et al. A Cope-type elimination was employed. See ref 1.
(8) Ford, W. T. *Acc. Chem. Res.* **1973**, *6*, 410.

Reduction of Aromatic Rings by 2-Propanol with Raney Nickel Catalysis

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2-Propanol is a reducing agent in certain environments that include photochemical and basic conditions as well as the presence of Raney nickel. This catalyst has been employed in some instances to reduce ketones to alcohols[1] in 2-propranol and to study the equilibration of epimeric alcohols[2] in the presence of some acetone.

In another connection, we were interested in the equilibrium position of the *syn*- and *anti*- 5-phenyladamantan-2-ols and attempted to measure it by treating

(1) (A) Fieser, M.; Fieser, L. F. "Reagents for Organic Synthesis"; Wiley: New York, 1967; p 723 ff. (b) Augustine, R. L. "Catalytic Hydrogenation"; Marcel Dekker: New York, 1965; p 24, 71.
(2) Eliel, E. L.; Schroeder, S. H. *J. Am. Chem. Soc.* **1965**, *87*, 5031.

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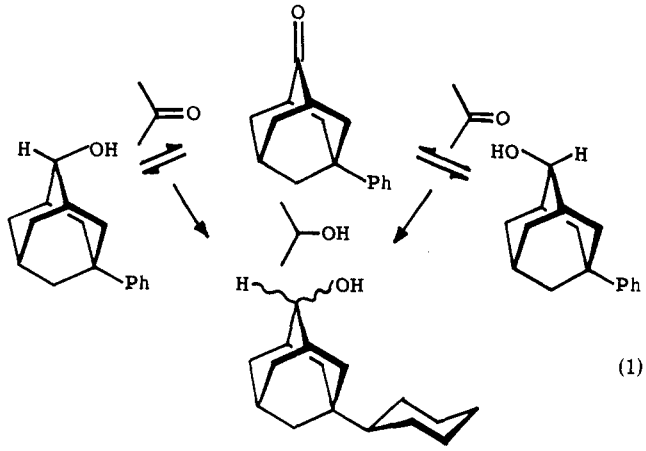
the corresponding ketone with Raney nickel in refluxing 2-propanol. The ^1H NMR spectrum of the first sample taken showed to our surprise that the signals in the aromatic region had vanished. The ^{13}C NMR spectrum confirmed our suspicion that the phenyl ring had been reduced (equ 1).

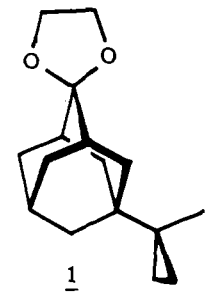


(1)

We could find no precedent for this reaction in the literature; indeed, the impression is that the complete reduction of benzene rings requires hydrogen gas under pressure and a platinum catalyst.[3] In view of the laboratory as well as technological potential of this reaction, we have made a brief survey of its scope and limitations.

The catalyst, similar to that described in the literature,[4] was stored in 2-propanol. In each of the reactions below, ca. 2 g of catalyst was used in 10 mL of 2-propanol for about 500 mg of substrate. *tert*-Butylbenzene, for example, after 67 h of reflux was reduced 50% to *tert*-butylcyclohexane. The mixture contained no other components, as was shown by means of both GC and ^1H NMR. Longer reflux times resulted in additional conversion, as did larger quantities of catalyst. It was also shown that the catalytic activity decreased in only a minor way upon repeated reuse: refluxing 15 successive fresh batches of the *tert*-butylbenzene for 4 h each with the same 4 g of catalyst caused a gradual decrease from 22 to 16% reduction. The same treatment brought about reduction of all aromatic compounds examined. Naphthalene was completely reduced in 18 h to a mixture of 90% of tetralin and 10% *cis*- and *trans*-decalin. Pyridine, in 12 h, gave 88% piperidine, with 12% recovery of starting material; this reaction was sufficiently vigorous that the initial solution quickly reached reflux temperature without assistance. Furan was 85% reduced to THF (with 15% recovery of furan) at room temperature overnight; in this case, cooling was initially required. Anisole was completely converted in 110 h of reflux, 90% of it to cyclohexyl methyl ether and the remainder to a volatile, unidentified material.

1-Methyl-1-phenylcyclopropane was reduced quantitatively in 2 h of reflux to 2-phenylbutane; subsequently the benzene ring was also reduced at the normal rate. The structure of the initial product and the rapid initial reduction in that case suggest the intervention of a benzylic radical; treatment of the adamantyl derivative 1[5] gave a mixture of the corresponding epimeric alcohols (and presumably 2,2-dimethyl-1,3-dioxolane), but the cyclopropyl ring was unaffected.



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The radical nature of the reaction is also suggested by the appearance of polymeric products when aromatic halogen compounds are reduced. Bromo- and chlorobenzene as well as α,α,α-trifluorotoluene all gave solid, waxy products that would not pass through a GC column; acetone solutions of these products gave no NMR evidence of remaining unsaturation. The bromobenzene product was halogen free (Beilstein test); the chlorobenzene product may still have contained small amounts of chlorine.[6]

Somewhat more complex behavior was encountered with nitrobenzene. While the nitro group is quickly, vigorously, and completely reduced to give aniline,[7] upon further exposure (now at reflux temperature), one obtains not only cyclohexylamine but, in addition, a second product determined to be cyclohexylisopropylamine. It seems clear that the secondary amine is formed by the further reaction of the first-formed products, aniline and acetone, followed by reduction; indeed, addition of a small amount of isopropylamine to trap the acetone formed greatly diminished the amount of byproduct.[8]

The reduction is not strongly exergonic. The known hydrogenation energies of benzene (−49 kcal/mol) and of acetone (−13 kcal/mol) show that the reaction is favorable by only about 10 kcal/mol. This is small enough that one could conceivably force the reaction in the opposite direction by appropriate choice of concentrations; however, refluxing of *tert*-butylcyclohexane and acetone with Raney nickel did not produce detectible amounts of *tert*-butylbenzene.

In conclusion, we find that the Raney nickel–2-propanol treatment may be a significant improvement over the traditional reduction of aromatic rings by means of compressed hydrogen in conjunction with nickel or noble metal catalysis.[9] The exceptionally facile reduction of the nitro group in nitrobenzene is also worthy of note. The conversion of halogenated aromatic compounds into largely halogen-free materials is of potential interest in problems

(3) Adams, R.; Marshall, J. R. *J. Am. Chem. Soc.* **1928**, *50*, 1970. Adkins, H.; Krsek, G. *J. Am. Chem. Soc.* **1948**, *70*, 412. Pearlman, W. M. *Org. Synth.* **1969**, *49*, 75. Rhodium, ruthenium, palladium, and iridium have also been used; see: Rylander, P. N. "Catalytic Hydrogenation over Platinum Metals"; Academic Press: New York, 1967; p 309 ff.

(4) Mozingo, R. "Organic Synthesis"; Wiley: New York, 1955; Coll. Vol. 3, p 181.

(5) le Noble, W. J.; Srivastava, S.; Cheung, C. K. *J. Org. Chem.* **1983**, *48*, 1099.

(6) Nickel-catalyzed dehalogenation with *compressed hydrogen* is known; see, for example: Buu-Hoi, N. P.; Xuong, N. D.; van Bac, N. *Bull. Soc. Chim. Fr.* **1963**, 2442. Two instances of the *noncatalytic* reduction of water soluble aromatic salts to the corresponding cyclohexanes with Raney nickel were reported after the completion of our work: Pojer, P. M. *Tetrahedron Lett.* **1984**, *25*, 2507.

(7) We find this to be an exceptionally facile and convenient way to reduce nitro compounds. Nickel catalysis in other media requires *compressed hydrogen* and elevated temperatures; see, for example: Dauben, H. J.; Ringold, H. J.; Wade, R. H.; Pearson, D. L.; Anderson, A. G. "Organic Synthesis"; Wiley: New York, 1983; Coll. Vol. 4, p 221.

(8) For related chemistry, see: Kindler, K.; Malamed, G.; Matties, D. *Liebigs Ann. Chem.* **1961**, *644*, 23. Rice, R. G.; Kohn, E. J. "Organic Synthesis"; Wiley: New York, 1963; Coll. Vol. 4, p 283. Balcon, D. M.; Noller, C. R. "Organic Synthesis", Wiley: New York, 1963; Coll. Vol. 4, p 603.

(9) For a recent example of Birch reduction and references, see: Benkeser, R. A.; Belmonte, F. G.; Yang, M. H. *Synth. Commun.* **1983**, *13*, 1103.

of the disposal of such environmentally harmful substances as PCB's and dioxins.

Experimental Section

All the aromatic compounds described in the text were commercially available materials used without further purification; 2-propanol was distilled from sodium metal. The instrumentation used included an HFT-80 (^{1}H) and a NT-300 (^{13}C) NMR spectrometer, a Hewlett Packard 5980-A mass spectrometer, a Waters Associates HPLC Instrument, Model 600A (refractive index detector, $^1/_4$ in. × 30 cm column, 10% (V/V) ethyl accetate–hexanes, 2.0 mL/min flow rate), and a Varian Aerograph 1400 GC instrument with a 10-ft column containing 15% Carbowax 20 M on Chromosorb W.

Preparation of Raney Nickel. Aqueous sodium hydroxide (30 g, 150 mL) was cooled in ice in a 500-mL beaker, stirred magnetically while 5 g of nickel–aluminum alloy (50/50 from Alfa) was added in several small portions, and gradually warmed to 100 °C as required to maintain the hydrogen evolution. The nickel was then allowed to settle, and the liquid was decanted. After being washed with 5% fresh sodium hydroxide and finally distilled water until neutral, the nickel suspension was filtered with a sintered glass funnel and then finally washed with 100 mL of 2-propanol. (*Caution*! The catalyst must be kept moist since it is highly pyrophoric!) The catalyst was transferred with small amounts of dry 2-propanol to a glass-stoppered bottle.

Reduction of 5-Phenyladamantan-2-one. This compound[10] (20.0 mg, 0.088 mmol) in 5.0 mL of dry 2-propanol was mixed with 400 mg of the Raney nickel and refluxed for 3 days with continuous magnetic stirring. The mixture was filtered, the residue washed twice with 2-propanol (20 mL) and the solvent removed in vacuo to yield 20.4 mg (98%) of a crude product that was purified by crystallization from hexanes: mp 142–5 °C; mass spectrum, *m/z* 234; ^{1}H NMR ($CDCl_3$ at δ 7.25) δ 0.8–2.0 (b m with a sharp peak at δ 1.5, 25 H), 3.8 (b s, 1 H); ^{13}C NMR ($CDCl_3$ at δ 77.000) δ 74.660, 74.166, 47.888, 48.435, 38.550 (2 C), 34.877 (2 C), 30.873 (2 C), 33.105 (2 C), 35.272 (2 C), 39.512 (2 C), 36.278 (2 C), 27.606, 28.084, 33.966, 33.714, 26.355 (2 C), 26.052 (2 C), 27.239 (3 C), 26.864 (2 C), 26.905. HPLC showed it to be a mixture of the (*E*)- and (*Z*)-alcohols in the ratio of 57:43.

General Procedure. To Raney nickel (2 g) in 2-propanol (5 mL) in a 25-mL round-bottomed flask equipped with a condenser and a mercury bubbler (to minimize the slow loss of volatile components) was slowly added a solution of 500 mg of the compound of interest in 10 mL of 2-propanol at room temperature or at 0 °C as required. The reaction mixture was brought to reflux and was followed by G.C. When the reaction was over or nearly so, the solution was decanted and a sample of the product was isolated by GC for characterization and comparison with the authentic compound.

Acknowledgment. We are pleased to acknowledge NSF support for this work.

Registry No. 1, 84454-65-9; 5-phenyladamantan-2-one, 38584-33-7; *syn*-5-phenyladamantan-2-ol, 93965-96-9; *anti*-5-phenyladamantan-2-ol, 94062-20-1; *syn*-5-cyclohexyladamantan-2-ol, 93943-16-9; *anti*-5-cyclohexyladamantan-2-ol, 94061-41-3; *tert*-butylbenzene, 98-06-6; *tert*-butylcyclohexane, 3178-22-1; naphthalene, 91-20-3; tetralin, 119-64-2; *cis*-decalin, 493-01-6; *trans*-decalin, 493-02-7; pyridine, 110-86-1; piperidine, 110-89-4; furan, 110-00-9; THF, 109-99-9; anisole, 100-66-3; cyclohexyl methyl ether, 931-56-6; 1-methyl-1-phenylcyclopropane, 2214-14-4; 2-phenylbutane, 135-98-8; 2-cyclohexylbutane, 7058-01-7; *syn*-5-(1-methycycloprop-1-yl)adamantan-2-ol, 93943-17-0; *anti*-5-(1-methylcycloprop-1-yl)adamantan-2-ol, 94061-42-4; 2,2-dimethyl-1,3-dioxolane, 2916-31-6; bromobenzene, 108-86-1; chlorobenzene, 108-90-7; α,α,α-trifluorotoluene, 98-08-8; nitrobenzene, 98-95-3; aniline, 62-53-3; cyclohexylamine, 108-91-8; cyclohexylisopropylamine, 1195-42-2; acetone, 67-64-1; isopropylideneaniline, 1124-52-3; Raney nickel, 7440-02-0; 2-propanol, 67-63-0.

(10) Geluk, H. W. *Synthesis* **1972**, 374.

Heptacyclo[5.5.1.1^{4,10}.0^{2,6}.0^{3,11}.0^{5,9}.0^{8,12}]tetradecane-13,14-bis(spiro-1′-cyclopentane): A New $C_{22}H_{28}$ Nonacyclic Cage Hydrocarbon. Improved Synthesis of Bicyclo[2.2.1]hepta-2,5-diene-7-spiro-1′-cyclopentane

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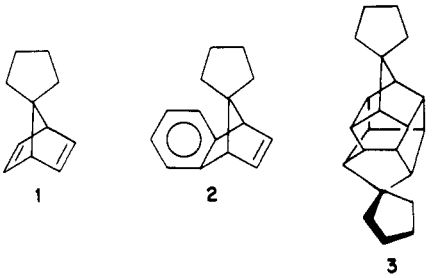
Recent interest in the synthesis and chemistry of new, substituted hepatacyclo[5.5.1.1^{4,10}.0^{2,6}.0^{3,11}.0^{5,9}.0^{8,12}]tetradecanes[1-3] prompts us to report our findings on the course of the iron carbonyl promoted cyclodimerization of bicyclo[2.2.1]hepta-2,5-diene-7-spiro-1′-cyclopentane (1). The



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thermal and photochemical reactions of $Fe(CO)_5$ with norbornadiene and with simple, 7-substituted norbornadienes normally result in the formation of a mixture of several dimeric and trimeric products, some of which are ketonic.[4,5] However, 2,3-benzobicyclo[2.2.1]hepta-2,5-diene-7-spiro-1′-cyclopentane (**2**) has been reported to be nearly inert toward iron pentacarbonyl.[6] This observation suggested to us that the course of the reaction of **1** with $Fe(CO)_5$ might be relatively uncomplicated, affording the corresponding cage dimer **3** as the major (or even sole) reaction product.

Since the yields of cage dimers produced via reaction of 7-substituted norbornadienes with $Fe(CO)_5$ are often low (i.e., on the order of 15%),[3,4] it was deemed desirable to have several grams of **1** on hand for the attempted cyclodimerization reaction. A multistep synthesis of **1** has been reported by Wilcox and Whitney;[7] however, their eight-step synthesis (starting with cyclopentadiene) affords **1** in only 2% overall yield. Accordingly, we sought to devise a new synthesis which might be capable of producing **1** in fewer steps and in greater overall yield.

Our improved, five-step synthesis of **1** is shown in Scheme I. Spirononadiene **4** is a critical intermediate both in the Wilcox–Whitney synthesis[7] and in the present synthesis of **1**. This compound is thermally labile, and it readily undergoes [1,5]sigmatropic rearrangement.[8] To avoid thermal rearrangement of **4**, this compound was purified by vacuum distillation (0.02 mm), and the distillate was collected and maintained at −78 °C. Diels–Alder addition of **4** to α-chloroacrylonitrile was performed in a

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(2) Ealick, S. E.; van der Helm, D.; Hayes, B. R.; Marchand, A. P. *Acta Crystallogr., Sect. B* **1978**, *B34*, 3219.
(3) Marchand, A. P.; Earlywine, A. D. *J. Org. Chem.* **1984**, *49*, 1660.
(4) Marchand, A. P.; Hayes, B. R. *Tetrahedron Lett.* **1977**, 1027.
(5) Weissberger, E.; Laszlo, P. *Acc. Chem. Res.* **1976**, *9*, 209.
(6) Mantzaris, J.; Weissberger, E. *J. Am. Chem. Soc.* **1974**, *96*, 1873, 1880.
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