The above mixture of selenides (55 mg) was dissolved in 1.0 mL of tetrahydrofuran, cooled to 0 °C, and treated with 20.4 μL of acetic acid and 96.2 μL of 30% hydrogen peroxide. After 40 min the reaction was quenched by the careful addition of aqueous sodium bicarbonate. The product was isolated by extraction with ether. The combined organic extracts were dried over anhydrous magnesium sulfate. The solvent was removed under reduced pressure. The crude product was filtered through 3.0 g of silica gel. Elution with ether gave 28 mg (84%) of pure *dl-epi*-pyroangolensolide (**4**): mp 136–137 °C; IR ($CHCl_3$) 1690 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.2–1.5 (m, 2 H, CH_2), 1.37 (s, 3 H, CH_3), 1.90 (d, 3 H, J = 0.7 Hz, olefinic CH_3), 2.2–2.4 (m, 2 H, allylic CH_2), 5.10 (s, 1 H, —CHO), 5.90 (s, 1 H), 6.12 (d, 1 H, J = 5.4 Hz), 6.26 (br s, 1 H), 7.32 (br s, 1 H), 7.38 (br s, 1 H). Anal. Calcd for $C_{15}H_{16}O_3$: C, 73.75; H, 6.60. Found: C, 74.10; H, 6.50.

***dl*-Pyroangolensolide (2).** To a solution of lithium diisopropylamide (0.24 mmol) in 1.0 mL of dry tetrahydrofuran (prepared from 33.5 μL of diisopropyl amine and 150 μL of a 1.6 M solution of *n*-butyllithium in hexane) cooled to −78 °C under argon was added 30 mg (0.12 mmol) of lactone **8** in 0.75 mL of tetrahydrofuran. After 40 min, 49 mg (0.26 mmol) of benzeneselenenyl chloride in 0.8 mL of tetrahydrofuran containing 41.7 μL of hexamethylphosphoramide was added at −78 °C. The reaction was quenched after 5 h with 5.0 mL of 1 N hydrochloric acid. The product was isolated by extraction with ether. Standard workup left 72 mg of a residue which was chromatographed on 22 g of silica gel. Elution with hexane/ether, 2:1, afforded 39 mg of a mixture of selenides which were used directly in the next reaction.

The above mixture of selenides (32 mg) was dissolved in 1.6 mL of tetrahydrofuran and was treated at 10 °C with 11.0 μL of acetic acid and 52.4 μL of 30% hydrogen peroxide. After 40 min, the temperature was raised to 20 °C. The reaction was quenched after 60 min with a saturated solution of sodium bicarbonate. The product was isolated by extraction with ether. The combined organic extracts were dried over anhydrous magnesium sulfate. The solvent was removed in vacuo. The residue was chromatographed on 10 g of silica gel. Elution with hexane/ether, 2:1, provided 20 mg of pure *dl*-pyroangolensolide: mp 135–136 °C; IR 1710 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.03 (s, 3 H, CH_3), 1.4–1.5 (m, 2 H, CH_2), 1.89 (d, 3 H, J = 1.4 Hz, olefinic CH_3), 2.26 (br s, 2 H, allylic CH_2), 5.12 (s, 1 H, —CHO), 5.84 (br s, 1 H), 6.14 (br t, 1 H, J = 3.97 Hz), 6.45 (br s, 1 H), 7.42 (br s, 1 H), 7.48 (br s, 1 H).

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Registry No. *dl*-**2**, 52730-12-8; **3**, 35986-56-2; *dl*-**4**, 52730-11-7; *dl*-**5**, 95421-96-8; *dl*-**7**, 95421-99-1; *dl*-**8**, 95421-97-9; (*E*)-CH_2=$CHC(CH_3)$=$CHCH_2CO_2H$, 95421-98-0; methacrolein, 78-85-3; 3-bromofuran, 22037-28-1; 3-furyllithium, 53101-93-2.

Polystyryldiphenylphosphine as a Deoxygenation Reagent for Sulfoxides

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Sulfur-containing compounds have played an increasingly important role in organic synthesis due to the ease of incorporation of the element into complex structures, the ability to modify the valency of the atom, the variety of chemical characteristics exhibited by those varied oxidation states, and the ease of removal of sulfur as needed for the synthesis.[1] The interconversion of oxidation states is a crucial aspect of the successful use of sulfur in synthetic applications, and one transformation which has received extensive investigation has been the sulfoxide to sulfide reduction reaction.[2]

The use of trivalent phosphorus compounds for the sulfoxide–sulfide conversion has been recognized for some time and a comparison of reactivity of a variety of such compounds with Me_2SO is available.[3] More specifically, the use of triphenylphosphine as a reductant for Me_2SO was first recognized in 1962[4] and has been exploited in the reduction of a variety of sulfoxides since that time by using as coreactants CCl_4,[5] acids,[6] and I_2/NaI.[7] These methods all show good chemoselectivity and proceed under generally mild conditions. They do suffer, however, from some potential separation difficulties due to the requisite formation of triphenylphosphine oxide and to the use of any excess triphenylphosphine. To avoid these purification requirements, we chose to investigate the use of polystyryldiphenylphosphine as a reducing agent for sulfoxides.[8]

All three coreactants mentioned above were utilized in combination with the polymeric reagent. Dibenzyl sulfoxide (**6**) was reduced to the sulfide in 85% yield by using a catalytic amount of HCl combined with the polymeric reagent with a 6-h reflux in THF solvent. It was concluded that lengthy contact with acid would significantly effect the generality of the procedure and this method was pursued no further. The I_2/NaI combination with polystyryldiphenylphosphine gave effective reduction of sulfoxide **5** but did require larger excesses of phosphine (1.5 equiv) and longer reflux times (1.5 h) than those reported for the solution-phase reaction.[7] Acetonitrile is a very poor swelling solvent for a lightly cross-linked (2%) polystyrene bead, leaving a number of phosphine sites inaccessible for reaction. Use of the better swelling solvent THF gave poor reduction yields both in the solution phase and with the polymeric reagent, demonstrating the need for the polar acetonitrile solvent reported by Olah.[7] In addition to the solvent restrictions, the use of I_2/NaI does require additional purification steps since the excess reagents must be removed by aqueous washes. As described below, the selection of CCl_4 as the coreactant provides mild reaction conditions along with ease of workup and was the method chosen for the remainder of our work.

As can be observed in Table I, the procedure is successful in reducing diaryl, arylalkyl, and dialkyl sulfoxides to their corresponding sulfides in excellent yields with substrates possessing alkyl substituents affording slightly lower yields than their aryl counterparts. The process is compatible with a variety of functional groups, particularly

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(1) For reviews of the uses of sulfur in organic synthesis, see: Block, E. *J. Chem. Ed.* **1971**, *48*, 814. Fleming, I. *Chem. Ind.* (*London*) **1975**, 449. Davidson, A. H.; Hodgson, P. K. G.; Howells, D.; Warren, S. *Chem. Ind.* (*London*) **1975**, 455.

(2) For a review, see: Drabowicz, J.; Numata, T.; Oae, S. *Org. Prep. Proced. Int.* **1977**, *9*, 63.

(3) Amonoo-Neizer, E. H.; Ray, S. K.; Shaw, R. A.; Smith, B. C. *J. Chem. Soc.* **1965**, 4296.

(4) Ray, S. K.; Shaw, R. A.; Smith, B. C. *Nature* (*London*) **1962**, *196*, 372.

(5) Castrillon, J. P. A.; Szmant, H. H. *J. Org. Chem.* **1965**, *30*, 1338.

(6) Szmant, H. H.; Cox, O. *J. Org. Chem.* **1966**, *31*, 1595.

(7) Olah, G. A.; Gupta, B. G. B.; Narang, S. C. *Synthesis* **1978**, 137.

(8) For recent reviews of polymeric reagents, see: Mathur, N. K., Narang, C. K., Williams, R. E., Eds.; "Polymers as Aids in Organic Chemistry"; Academic Press: New York, 1980. Hodge, P., Sherrington, D. C., Eds.; "Polymer-Supported Reactions in Organic Synthesis"; Wiley: London, 1980. Akelah, A.; Sherrington, D. C. *Chem. Rev.* **1981**, *81*, 557. Akelah, A. *Synthesis* **1981**, 413.

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Table I. Reduction of Sulfoxides

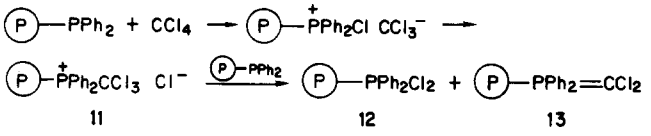
	sulfoxide	sulfoxide mp/bp, °C (lit.)	reactn time, h	sulfide yield,[a] %	sulfide mp, °C (lit.)
1	$(CH_3)_2S{=}O$	oil	2	*b*	157–162 (157.5–163.5)[c]
2	$n\text{-}Bu_2S{=}O$	31–34(32)[d]	2	81	oil
3	$Ph_2S{=}O$	70–72(70)[d]	1.5	98	oil
4	$(p\text{-}ClC_6H_4)_2S{=}O$	142–145 (145)[e]	2	96	93–96 (92–93)[f]
5	$(p\text{-}CH_3C_6H_4)_2S{=}O$	95–97 (92)[d]	1.5	99	54–56 (57.3)[d]
6	$(PhCH_2)_2S{=}O$	131–133 (134–135)[d]	1.5	94	44–47 (49–50)[d]
7	$PhSOCH{=}CH_2$	oil	1	87	oil
8	$m\text{-}O_2NC_6H_4SOCH_2Ph$	93–94[g]	1	96	oil
9	$p\text{-}O_2NC_6H_4SOPh$	108–110 (107–107.5)[h]	2	94	53–55 (55)[h]
10	$p\text{-}CH_3OC_6H_4SO(CH_2)_3CH_3$	oil[g]	6[i]	85	oil[g]

[a] All yields are based on products purified by vacuum distillation (bulb-to-bulb) or silica gel chromatography. Unless otherwise noted, the ratio of sulfoxide:phosphine:CCl_4 was 1:1.6:4. [b] Sulfide was not quantitated due to high volatility; no Me_2SO remained in the reaction mixture. [c] $(Me_2S)_2{\cdot}3HgCl_2$ complex reported in ref 5. [d] See ref 16. [e] Granoth, I.; Kalir, A.; Pelah, Z. *J. Chem. Soc. C* **1960**, 2424. [f] Still, I. W. J.; Hasan, S. K.; Turnbull, K. *Synthesis* **1977**, 468. [g] Acceptable analytical and spectral data were obtained on these new compounds, see Experimental Section. [h] See ref 15. [i] A ratio of reagents of 1:3.2:8 was required for complete reaction.

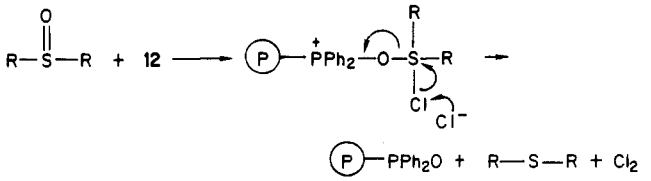
the easily reduced nitro and C=C functionalities. The procedure is quite simple, involving the reflux of the polymeric reagent with the sulfoxide and CCl_4 in a THF solvent. We chose to use THF vs. the CCl_4 solvent previously reported,[5,6] primarily to minimize toxicity hazards. After the indicated reflux time, the cooled reaction mixture is filtered to remove the polymer, dried to remove any traces of water, and then evaporated under reduced pressure to afford the desired sulfide product. Usually the residue is sufficiently pure for further use, but chromatography or distillation can also be easily applied.

In virtually all cases, the molar ratio of reagents was 1:1.6:4.0 for sulfoxide:phosphine:CCl_4. Further reductions in the quantity of phosphine led to slower and frequently incomplete reactions. The *p*-methoxy compound **10** demonstrated a much slower rate of reaction, perhaps due to the electron-donating properties of the substituent leading to stabilization of the tetravalent sulfur intermediate (see below) toward attack by the nucleophile. A doubling in the quantities of phosphine and CCl_4 was required, but the yield of sulfide remained quite acceptable.

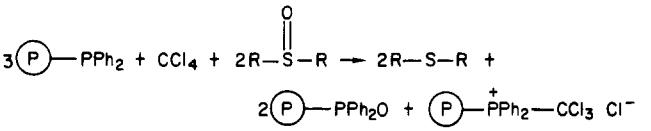
While the mechanisms of reactions involving PPh_3/CCl_4 are complex and not fully understood, Hodge and coworkers have investigated the mechanistic sequence for the polystyryldiphenylphosphine/CCl_4 conversion of alcohols and carboxylic acids to their corresponding chlorides.[9] They suggest that in the major pathway of the reaction the CCl_4 reacts with 2 equiv of the phosphine reagent to generate a pair of reactive intermediates, **12** and **13**.



If we presume the initial steps of this mechanism are applicable to the sulfoxide reduction, then intermediate **12** could react with the sulfoxide in a manner analogous



to that proposed by Olah for the $Ph_3P/NaI/I_2$ reduction.[7,10] The observation that only 1.6 equiv of phosphine are required for reduction implies that the Cl_2 generated in this reduction step may undergo further reaction, either directly with additional polymeric phosphine to give **12**, or with **13**, to yield **11**, thus continuing the cycle.[11] Incorporating this proposed step, the overall stoichiometry of the process becomes



This reaction provides a much better fit to the observed stoichiometry of the process, but additional work is required to confirm this proposed mechanism since the eventual fate of the third equivalent of phosphine has not been clearly defined.

In summary, the use of polystyryldiphenylphosphine with CCl_4 provides a very effective means of reducing sulfoxides. The reaction provides high yields, mild reaction conditions, a broad tolerance of functional groups, and a very convenient isolation procedure.

Experimental Section

General Procedures. IR spectra were obtained on a Beckman Acculab instrument by using thin films or Nujol mulls on NaCl plates. NMR spectra were recorded on a Varian T-60 instrument with $CDCl_3$ as the solvent and with tetramethylsilane as an internal standard. Sulfoxides **1–7** were obtained from commercial sources and were purified as needed by recrystallization or distillation. Sulfoxides **8–10** were prepared by oxidation of the corresponding sulfides (see Experimental Section), which were either commercially available (**9**) or were prepared by a previously described procedure (**8** and **10**).[12] The polystyrene beads (2% divinylbenzene, 200–400 mesh) were obtained from Eastman. Elemental analyses were performed by Galbraith Laboratories, Knoxville, TN.

Preparation of Polystyryldiphenylphosphine. Polystyrene beads were washed[13] and then brominated[14] to give a yellow solid with 5.07 mmol of Br/g of resin. The phosphine ligand was then incorporated by the method described earlier,[12] giving a pale yellow solid with 3.26 mmol of P/g of resin.

Preparation of Sulfoxide 8. The requisite sulfide,[12] 343 mg (1.40 mmol), was diluted with 15 mL of CH_2Cl_2 and cooled to −10 °C. *m*-Chloroperoxybenzoic acid, 270 mg of 85% pure material (1.33 mmol), was dissolved in 10 mL of CH_2Cl_2 and added dropwise to the sulfide over a 40-min period at −10 °C. Following the addition, the mixture was slowly warmed to room temperature and stirred 1 h. The mixture was diluted with 50 mL of ether and was washed with 2 × 15 mL of saturated $NaHCO_3$. Drying

(9) Harrison, C. R.; Hodge, P.; Hunt, B. J.; Khoshdel, E.; Richardson, G. *J. Org. Chem.* **1983**, *48*, 3721.
(10) Olah, G. A.; Gupta, B. G. B.; Narang, S. C. *J. Org. Chem.* **1978**, *43*, 4503.
(11) Ramirez, F.; Desai, N. B.; McKelvie, N. *J. Am. Chem. Soc.* **1962**, *84*, 1745.
(12) Amos, R. A.; Fawcett, S. M. *J. Org. Chem.* **1984**, *49*, 2637.
(13) Relles, H. M.; Schluenz, R. W. *J. Am. Chem. Soc.* **1974**, *96*, 6469.
(14) Farrall, M. J.; Fréchet, J. M. J. *J. Org. Chem.* **1976**, *41*, 3877.

over $MgSO_4$ and removal of solvent gave a white solid. Preparative layer chromatography (PLC) (50% ethyl acetate:hexane, silica gel) gave 346 mg (99%) of a white solid: mp 93–94 °C; ^1H NMR ($CDCl_3$) δ 7.95–8.25 (m, 2 H), 7.35–7.60 (m, 2 H), 6.70–7.25 (m, 5 H), 4.03 (s, 2 H); IR (Nujol) 1055 cm^{-1} (S=O).

Anal. Calcd for $C_{13}H_{11}O_3NS$: C, 59.76; H, 4.24; N, 5.36; S, 12.27. Found: C, 59.56; H, 4.20; N, 5.16; S, 12.40.

A similar procedure afforded sulfoxide **10** in a 95% yield, isolated by PLC as a colorless oil: ^1H NMR ($CDCl_3$) δ 7.40 (d, 2 H, J = 9 Hz), 6.85 (d, 2 H, J = 9 Hz), 3.73 (s, 3 H), 2.73 (t, 2 H, J = 7 Hz), 1.20–1.80 (m, 4 H), 0.70–1.10 (m, 3 H); IR (neat) 1045 cm^{-1} (S=O).

Anal. Calcd for $C_{11}H_{16}O_2S$: C, 62.23; H, 7.60; S, 15.10. Found: C, 61.92; H, 7.50; S, 14.94.

Sulfoxide **9** was formed in a similar fashion in 93% yield, mp 108–110 °C (lit.[15] mp 107–107.5 °C).

Typical Procedure for Sulfoxide Reduction. Di-*p*-tolyl sulfoxide (**5**) (288 mg, 1.25 mmol) was added to a 50-mL three-necked flask that had been thoroughly flushed with N_2; 15 mL of THF were then added, followed by 0.61 g of polystyryldiphenylphosphine (2.00 mmol contained P). CCl_4, 0.48 mL (5.00 mmol), was added and a reflux was started immediately. TLC (50% ether:hexane, silica gel) after 1.5 h revealed complete consumption of sulfoxide and formation of a single product of high R_f. The polymer was removed by filtration and washed thoroughly with 40 mL of ether. After brief drying over $MgSO_4$, the solvent was removed under reduced pressure. PLC (50% ether:hexane) yielded 266 mg (99%) of a white solid, mp 54–56 °C (lit.[16] mp 57.3 °C).

With the exception of sulfoxide **10**, all substrates afforded the desired sulfides by utilizing this procedure and the reaction times indicated in Table I. The spectral characteristics of all products matched those of the known compounds.

Sulfoxide **10** was much slower in reaction with an estimated 50% completion after a 5-h reflux by using the same ratio of reactants as above. Addition of additional amounts of reagents to bring the molar ratio to 1:3.2:8.0 (sulfoxide:phosphine:CCl_4) resulted in complete reaction after an additional 1-h reflux. PLC (20% ether:hexane) afforded an 85% yield of a colorless oil: ^1H NMR ($CDCl_3$) δ 7.17 (d, 2 H, J = 9 Hz), 6.67 (d, 2 H, J = 9 Hz), 3.68 (s, 3 H), 2.77 (t, 2 H, J = 7 Hz), 1.10–1.70 (m, 4 H), 0.70–1.00 (m, 3 H).

Anal. Calcd for $C_{11}H_{16}OS$: C, 67.30; H, 8.22; S, 16.33. Found: C, 67.30; H, 8.15; S, 16.42.

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Registry No. **1**, 67-68-5; **2**, 2168-93-6; **3**, 945-51-7; **4**, 3085-42-5; **5**, 1774-35-2; **6**, 621-08-9; **7**, 20451-53-0; **8**, 95217-44-0; **9**, 955-45-3; **10**, 78597-97-4; **1** (sulfide), 75-18-3; **2** (sulfide), 544-40-1; **3** (sulfide), 139-66-2; **4** (sulfide), 5181-10-2; **5** (sulfide), 620-94-0; **6** (sulfide), 538-74-9; **7** (sulfide), 1822-73-7; **8** (sulfide), 87740-10-1; **9** (sulfide), 952-97-6; **10** (sulfide), 78597-95-2; poly(styryldiphenylphosphine), 95217-45-1; CCl_4, 56-23-5.

(15) Szmant, H. H.; McIntosh, J. J. *J. Am. Chem. Soc.* **1951**, *73*, 4356.
(16) Handbook of Chemistry and Physics, 47th ed., Chemical Rubber Co.: Cleveland, OH 1966.

Attractive Nonbonded Interactions. The Solution Conformation of 1,2-Bis(pentafluorophenyl)-1,2-bis(*p*-methoxyphenyl)ethane

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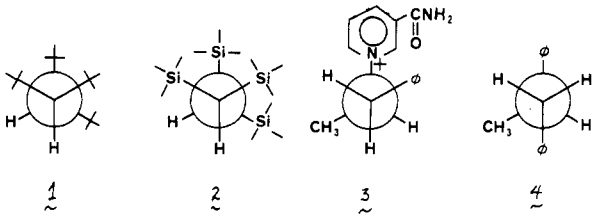
The tetrasubstituted ethanes **1** and **2** adopt, exclusively, an ethane H–H gauche conformation,[1] whereas 2,3-dimethylbutane exists in a statistical (2:1) ratio of the ethane H–H gauche and anti conformations.[2] Thus, 1,1,2,2-



1 2 3 4

tetraalkylethanes prefer the gauche conformation with the preference increasing as the bulk of the alkyl groups increases. In contrast to the tetraalkylethanes, tetraarylethanes, e.g., 1,1,2,2-tetraphenylethane, prefer the ethane H–H anti conformation.[3] Empirical force field calculations predict for tetraphenylethane that the lowest energy gauche conformation is ~5 kcal/mol less stable than the anti form.[4]

We have recently discovered[5] a case in which polar interactions reverse a conformational preference; *N*-(1-phenyl-2-propyl)nicotinamide chloride (**3**) prefers the aryl–aryl gauche, aryl–methyl anti conformation, whereas its carbon analogue 1,2-diphenylpropane (**4**) prefers the aryl–aryl anti conformation.[6] In the former case the free energy difference between the more stable gauche conformer and the anti conformer is –0.66 kcal/mol while in the latter case the difference is 0.24 kcal/mol.[7] If the energy differences between conformations in similarly substituted systems is sufficiently small, as in the cases above, polar contributions may be sufficient to alter expected conformations. In the tetraarylethane series, polar effects have been probed in a study of the diastereomeric 1,2-diphenyl-1,2-di-4-pyridylethanes, both of which were demonstrated to exist predominantly in the H–H anti form.[8]

Numerous aryl–aryl complexes of hexafluorobenzene with donor aromatic molecules have been reported, e.g., hexafluorobenzene with benzene,[9] mesitylene,[10] hexamethylbenzene,[11,12] *p*-xylene,[13] and durene,[14] to mention but a few. Perhaps the most unexpected behavior is exhibited by an equimolar mixture of hexafluorobenzene and benzene; both pure compounds melt at ca. 5 °C and both boil at ca. 80 °C, yet the equimolar mixture melts at 23.7 °C.[9] Postulating that in appropriately substituted tetraarylethanes attractive ring–ring interactions of the perfluorobenzene–benzene type might suffice to alter the expected H–H anti predominance observed in other tetraarylethanes, the diastereomeric (*meso* and *dl*) 1,2-bis(*p*-methoxyphenyl)-1,2-bis(pentafluorophenyl)ethanes

(1) Brownstein, S.; Dunogues, J.; Lindsay, D.; Ingold, K. V. *J. Am. Chem. Soc.* **1977**, *99*, 2073.
(2) Lunazzi, L.; Macciantelli, D.; Bernardi, F.; Ingold, K. V. *J. Am. Chem. Soc.* **1977**, *99*, 4573.
(3) Dougherty, D. A.; Mislow, K.; Blount, J. F.; Wooten, J. B.; Jacobus, J. *J. Am. Chem. Soc.* **1977**, *99*, 6149.
(4) Finocchiaro, P.; Gust, D., Hounshell, W. D.; Hummel, J. P.; Maravigna, P.; Mislow, K. *J. Am. Chem. Soc.* **1976**, *98*, 4945.
(5) El-Torki, F. M.; Jacobus, J. *J. Org. Chem.*, following note in this issue.
(6) Jacobus, J. *Tetrahedron Lett.* **1976**, 2927.
(7) These estimates are based on calculated conformer populations and are subject to error depending upon the selection of "theoretical" coupling constants employed in the calculation.
(8) Dougherty, D. A.; Llort, F. M.; Mislow, K.; Blout, J. F. *Tetrahedron* **1978**, *34*, 1301.
(9) Patrick, C. R.; Prosser, G. S. *Nature (London)* **1960**, *187*, 1021.
(10) Dahl, T. *Acta Chem. Scand.* **1971**, *25*, 1031.
(11) Dahl, T. *Acta Chem. Scand.* **1972**, *26*, 1569.
(12) Dahl, T. *Acta Chem. Scand.* **1973**, *27*, 995.
(13) Dahl, T. *Acta Chem. Scand., Ser. A* **1975**, *A29*, 170.
(14) Dahl, T. *Acta Chem. Scand., Ser. A* **1975**, *A29*, 699.

0022-3263/85/1950-1313$01.50/0