benzylhydrazine and 1,1-dimethyl-2-(2-propenyl)hydrazine were prepared by the published procedure.[24]

The preparation of compounds **8d, 8e, 12,** and **13** was accomplished by treating phenyl isocyanate with an equimolar quantity of the appropriate hydrazine.

Compounds **8a, 8b,** and **8c** were prepared by heating a mixture of either dimethylcarbamoyl chloride or diphenylcarbamoyl chloride with 2 equiv of the appropriate hydrazine at 100° for 1–2 hr. The products were isolated by extracting the crude reaction mixture with boiling petroleum ether.

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Registry No.—6a, 51433-78-4; **6b,** 51433-77-3; **6c,** 51433-50-2; **6d,** 51433-51-3; **6e,** 51433-52-4; **6f,** 51433-53-5; **7a,** 51433-54-6; **7b,** 51433-55-7; **7c,** 51433-56-8; **7d,** 51433-57-9; **7e,** 51433-58-0; **7f,** 51433-59-1; **8a,** 51433-60-4; **8b,** 51433-61-5; **8c,** 51433-63-7; **8d,** 51433-64-8; **8e,** 51433-65-9; **9,** 51433-66-0; **9** hydrochloride, 51472-52-7; **10,** 51433-67-1; **11,** 51433-68-2; **12,** 51433-69-3; **13,** 51433-70-6; **15,** 51433-71-7; **18,** 51433-76-2; allyl bromide, 106-95-6; benzyl bromide, 100-39-0; benzyl chloride, 100-44-7; 3-propynyl bromide, 106-96-7; 1,1,4,4-tetramethylsemicarbazide, 27827-93-6; *N,N*-dimethylcarbamoyl chloride, 79-44-7; 1,1-dimethylhydrazine, 57-14-7; 1,1-dimethyl-4,4-diphenylsemicarbazide, 37934-75-1; 1,1-dimethyl-4-phenylsemicarbazide, 6297-20-7; 1-(2-butenyl)-1,1-dimethyl-4-phenylsemicarbazonium bromide, 51433-71-7; 1-(3-methyl-2-butenyl)-1,1-dimethyl-4-phenylsemicarbazonium bromide, 51472-53-8; 1,1dimethyl-4-phenylsemicarbazide picrate, 51433-72-8; crotyl bromide, 4787-77-4; 1,1-dimethyl-2-(2-butenyl)hydrazine, 51433-73-9; crotonaldehyde *N,N*-dimethylhydrazone, 74422-95-9; 1,1,1-trimethyl-2-(2-butenyl)hydrazinium iodide, 51433-74-0; 1,1-dimethyl-2-(1-methyl-2-propenyl)hydrazine, 15848-66-5; phenyl isocyanate, 103-71-9; 1,1-dimethyl-1-(2-butenyl)hydrazinium bromide, 27828-89-3; *N,N*-diphenylcarbamoyl chloride, 83-01-2; 1,1-dimethyl-2-(2-propenyl)hydrazine, 2736-72-3; 1,1-dimethyl-2-benzylhydrazine, 28082-45-3.

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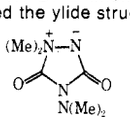
(16) For a recent review of CIDNP and its interpretation when applied to rearrangements of aminimides, see A. R. Lepley in "Chemically Induced Dynamic Nuclear Polarization," A. R. Lepley and G. L. Closs, Ed., Wiley, New York, N. Y., 1973, p 356.
(17) A 450-W Hanovia medium-pressure mercury arc lamp (Pyrex filter) was employed.
(18) The mass spectrum of the salt did not display high mass ion radicals corresponding to cations **16** or **17.** The highest mass peak observed was *m/e* 140, which corresponds to the monomeric cation (**18**) − 1. Depolymerization of **16** or **17** on electron impact apparently occurs. We have found that the mass spectrum of triphenyl isocyanaurate displays $C_6H_5NCO{\cdot}^+$ as the parent peak together with a $(C_6H_5NCO)_3{\cdot}^+$ peak which is 50% less intense.
(19) Dimethylamino isocyanate does not form a diazetidinedione dimer. W. S. Wadsworth and W. D. Emmons [*J. Org. Chem.*, **32,** 1279 (1967)] have established the ylide structure (i) shown below for the



i

dimethylamino isocyanate dimer. The nmr spectrum of the salt obtained by us displays equivalent methyl and crotyl groups; hence a dialkylated derivative of i can be excluded.
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1-Oxadecalins and 1-Oxa-4-decalones. Syntheses and Conformational Analyses

Jerry A. Hirsch* and George Schwartzkopf

Department of Chemistry, Seton Hall University, South Orange, New Jersey 07079

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A general synthetic route to 6- and 7-carbomethoxy-*trans*-1-oxadecalins (**9** and **12**) is presented. Base-catalyzed equilibrations and pmr data are used to evaluate conformational equilibria and relative configurations in several *cis*- and *trans*-1-oxadecalins and 1-oxa-4-decalones. The trans-fused ring system is thermodynamically favored in all instances.

The *trans*-decalin ring system has often been used as a conformationally fixed system for the study of the relative reactivities of equatorial and axial substituents[1] and the relative energies of substituents in a pair of equatorial and axial orientations at a given carbon atom.[2] Similarly, analogs of *trans*-decalin containing an atom other than carbon at a known position in the ring not containing the attached substituents provide the opportunity to evaluate the influences of the heteroatoms on the relative reactivities and relative energies of the substituents. These effects

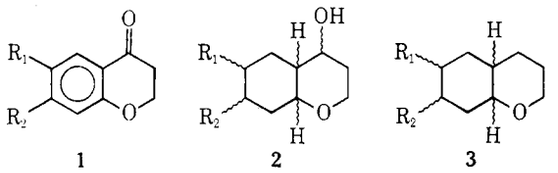
Table I
Representative 4-Chromanone Hydrogenations

Compd	Yield, % 2	3	4
1a	95	Not observed	5
1b	60	25[a]	15
1c	45	35[b]	20

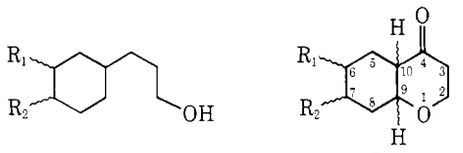
[a] Composed of a single cis-fused compound by glpc. [b] Composed of two cis-fused compounds in a 4:1 ratio by glpc.

would result from changes in ring structure and from polar effects, the latter of which probably can be accounted for using a field model[3] and the Kirkwood-Westheimer formalism.[3]

As the first phase of such a study of *trans*-heteradecalins, the syntheses and conformational analyses of representative 1-oxadecalins have been explored. Several 4-chromanones (**1a–c**) have been prepared and subjected to catalytic hydrogenation over ruthenium.[4] The major product in each instance (45–95% of isolated product) was a mixture of 4-hydroxy-1-oxadecalins (**2**), while significant amounts (5–35%) of 1-oxadecalins (**3**) and saturated monocyclic alcohols (**4**) were also isolated (Table I). In each series, the 4-hydroxy-1-oxadecalins were complex mixtures containing primarily cis ring fusions. Jones oxidation[5] of each alcohol mixture reduced the number of epimers[6] and somewhat simplified stereochemical assignments.



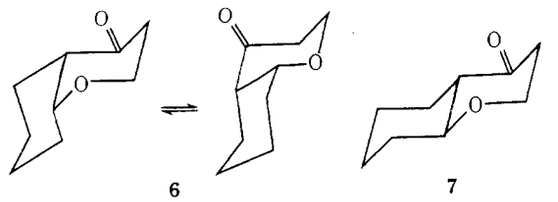
1 2 3



4 5

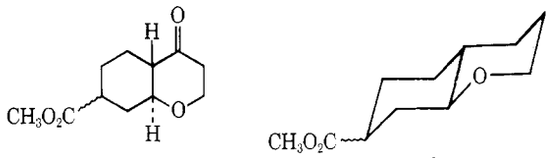
a, $R_1 = R_2 = H$
b, $R_1 = H$; $R_2 = CO_2CH_3$
c, $R_1 = CO_2CH_3$; $R_2 = H$

Ketone **5a** appeared to be primarily[8] the cis isomer[9] **6**, since H-9 appeared as an obscured narrow multiplet at δ 3.80 in the pmr spectrum. Treatment of ketone **5a** with 1,5-diazabicyclo[4.3.0]non-5-ene (DBN), a strong nonnucleophilic base,[10] in refluxing benzene resulted in a 90% recovery of trans-fused[8] **7**, which exhibited H-9 as the X portion of an ABMX pattern (J = 4, 10, and 10 Hz) at δ 3.23 in the pmr spectrum.[11] A mixture of ketones **5a** and **7** was similarly treated with DBN to establish that equilibration was occurring. The greater stability of the *trans*-1-oxa-4-decalone (**7**) under equilibration conditions qualitatively parallels that of the 1-decalone system[12] and appears to be quantitatively[8] greater than in this hydrocarbon analog.



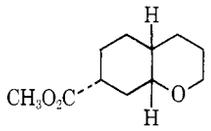
6 7

In the 7-carbomethoxy series, ketone **5b** was chromatographically homogeneous, cis fused, and a mixture of the carbomethoxy epimers (see below). Equilibration of **5b** with DBN in benzene gave a mixture of ketones which was chromatographically homogeneous and almost exclusively trans fused. Pure trans-fused material (**8**) was obtained by one recrystallization, and exhibited H-9 as a multiplet in the pmr spectrum at δ 3.29 (J = 4, 10, 10 Hz). Again the trans-fused ketone was more stable than the cis-fused.



8 9

Deoxygenation of **8** without bridgehead epimerization was required to obtain the desired *trans*-1-oxadecalin structure. A recently reported procedure[13] involving sodium cyanoborohydride reduction of the tosylhydrazone was investigated for stereospecificity.[14] Application of this procedure to **8** resulted in 76% conversion to a single pure 7-carbomethoxy-*trans*-1-oxadecalin (**9**). In order to check the stereospecificity, the mother liquors from which **8** had been obtained were deoxygenated. The product consisted of two epimers in addition to the major product **9**. One of these, a *cis*-1-oxadecalin, was identical with the oxadecalin **3** obtained directly from the hydrogenation of **1b**, and is assigned structure **10** based on the pmr spectrum and the known[15] preference for cis hydrogenation over ruthenium. The other isomer was not identified. The small amounts of these isomers formed from the mother liquors and the absence of these isomers in the deoxygenation of pure **8** indicate negligible, if any, epimerization at C-10 under these deoxygenation conditions.



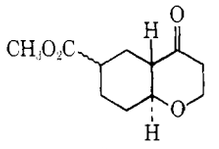
10

Several attempts were made to investigate the equatorial or axial nature of the carbomethoxy substituent in **9**. Ester **9** was treated with DBN in order to see whether the ester had been equilibrated simultaneously with the ring fusion (**5b** ⇄ **8**). No change was observed, compound **9** being recovered quantitatively. Treatment of **9** with sodium methoxide in methanol, the conditions used by Sicher[2] to equilibrate the *trans*-decalyl esters, resulted in the appearance of a new compound comprising approximately 20% of the mixture (by glpc). Mass spectral evidence supports assignment of the epimeric trans-fused structure to this compound. Unfortunately, equilibrium was probably not achieved because of decomposition of one of the esters under the reaction conditions. Nevertheless, these results support lack of ester epimerization with DBN. The pmr spectrum of ester **9** provides suggestive evidence that the ester functionality is equatorial. The C-7 methine hydrogen appears as a multiplet centered at δ 2.45 with coupling constants consistent only with an axial location (J = 3.5, 3.5, 12, 12 Hz).

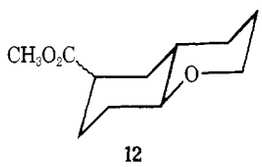
In the 6-carbomethoxy series, ketone **5c** obtained on Jones oxidation[5] of alcohol **2c** was a mixture of three compounds in a 3:6:1 ratio. Epimerization with DBN changed this ratio to 6:3:1. The major isomer was obtained in 90% purity by several recrystallizations and identified as a trans-fused compound (**11**) by the H-9 multiplet at δ 3.24[11] (J = 4.5, 10, 10 Hz) in the pmr.

Deoxygenation[13] of a mixture of 6-carbomethoxy-1-oxa-

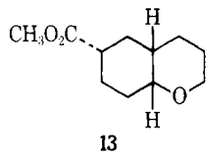
4-decalones containing 80% of trans-fused **11** produced two products in a ratio of 85:15, while deoxygenation of a 1:1 mixture of the trans and cis ketones resulted in a 1:1 mixture of deoxygenated products. Preparative glpc permitted identification of the major component in the trans-enriched reaction as a 6-carbomethoxy-*trans*-1-oxadecalin (**12**). The minor component was found to be identical with the major component in the 1-oxadecalin mixture (**3**) obtained directly on hydrogenation of ketone **1c** (Table I). Based on the well-known preference for cis hydrogenation,[15] this compound may be assigned structure **13** and the minor component from the direct hydrogenation would be **14**.



11



12



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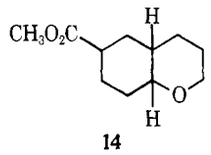

14

The ester functionality in 6-carbomethoxy-*trans*-1-oxadecalin (**12**) is tentatively assigned an axial orientation based on pmr spectral comparisons. The C-6 methine hydrogen is not discernible. However, the carbomethoxy methyl appears as a clean singlet at δ 3.72 in **12** and at δ 3.70 in precursor trans ketone **11**. By contrast, the methyl signal occurs at δ 3.67–3.68 in the 7-carbomethoxy analogs **8** and **9** in which the ester group is believed to be equatorial (see above). In addition, cis-fused systems **5b**, **10**, **5c**, and **13** all exhibited methyl singlets at δ 3.67 or 3.68. Using the reasonable assumption that conformational equilibria in cis systems (as shown for **6**) will result in a preponderance of that conformer in which any single attached substituent would be equatorial, the data from these cis systems reinforces the axial assignment to the ester group in **12**.

The result that the carbomethoxy group is equatorial in the 7 series (**9**) and axial in the 6 series (**12**) is consistent with another line of reasoning. If all-cis hydrogenation is assumed to predominate,[15] those isomers of **5b** and **5c** would be formed which would lead on DBN equilibration to the trans-fused ketonic precursors of **9** and **12** without any change in the ester stereochemistry. A perfect 1:1 correlation exists through both reaction sequences between ester stereochemistry in the major (all-cis) isomer of **5** and the final 1-oxadecalins **9** and **12**, thereby providing further substantiation of the proposed C-6 and C-7 stereochemistry and of all the assumptions made concerning reaction stereochemistry.

Experimental Section

Melting points and boiling points are uncorrected. Nmr spectra were recorded on a Varian A-60A instrument using solutions in deuteriochloroform. Chemical shifts are assigned in Table II and coupling constants are shown in Table III except where either is included with the compound or mixture. Infrared spectra were determined with a Beckman IR-10 spectrophotometer, with only major absorptions being cited. Mass spectral analyses were obtained at 70 eV. Elemental analyses were performed by Alfred Bernhardt Mikroanalytisches Laboratorium, Elbach, West Germany. The glpc column used was a 6 ft × 0.25 in. 10% NPGS w/w on Chromosorb W (60/80 mesh) column.

Previously Unreported Glpc Retention Times for 1-Oxadecalins (3) Isolated from the Hydrogenation of 4-Chromanones (1b and 1c). 7-Carbomethoxy-*cis*-1-oxadecalin (**10**) (175°), 8.3 min; mixture of epimers of 6-carbomethoxy-*cis*-1-oxadecalin (**3c**) (175°), 6.7 and 8.9 min (1:4).

Table II
Nmr Chemical Shifts of 1-Oxadecalins[a]

Compd	H-2e[b]	H-2a	H-3e	H-7	H-9	H-10	CO_2-CH_3
5a	4.29	3.73	2.19		3.80	2.77	
5b							3.68
5c							3.67
7	4.29	3.73	2.28		3.23	2.82	
8	4.35	3.75	2.33		3.29	2.73	3.68
9	4.00	3.40		2.45	2.93		3.67
10	3.65						3.67
11	4.34	3.75			3.24		3.70
12	4.00	3.41			2.93		3.72
13	4.05	3.55					3.68

[a] In parts per million. [b] e and a have been used to designate equatorial and axial protons.

Hydrogenation of 1a. The general procedure of Hirsch and Schwartzkopf[4] was used for the hydrogenation of 2.0 g (14 mmol) of **1a**. Chromatography on silica gel (Woelm) of 1.9 g of crude product gave 50 mg (3%) of crude 3-cyclohexyl-1-propanol (**4a**) in the 10% ethyl acetate–methylene chloride fractions as an oil: ir (neat) 3400 (O–H) and 1070 cm^{-1} (C–O); nmr δ 3.69 (t, J = 6 Hz, CH_2CH_2OH), 3.31 (s, OH).

The 20–100% ethyl acetate–methylene chloride fractions contained 1.42 g (67%) of 4-hydroxy-1-oxadecalin isomers (**2a**) as an oil, ir (neat) 3400 cm^{-1} (O–H).

Anal. Calcd for $C_9H_{16}O_2$: C, 69.20; H, 10.33. Found: C, 69.07; H, 10.41.

6-Carbomethoxy-1-oxa-4-decalone (5c). A stirred solution of 1.57 g (7.33 mmol) of the isomer mixture **2c** in 20 ml of acetone was treated with 3,4 ml (9.5 mmol) of Jones reagent[5] (2.8 *M*) over a few minutes at 0–10°. The ice bath was removed and stirring was continued for 1 hr. The mixture was treated with 20 drops of isopropyl alcohol and diluted with ether. The ethereal extract was washed with saturated sodium bicarbonate and dried ($MgSO_4$). Concentration of the ethereal extract gave 1.06 g of crude product. Chromatography on silica gel (Woelm) using 10% ethyl acetate–methylene chloride gave 940 mg (60%) of a mixture of three isomers of **5c** as an oil: ir (neat) 1730 cm^{-1} (C=O); glpc retention times (225°) 8.4, 10.2, and 11.1 min (3:6:1).

Anal. Calcd for $C_{11}H_{16}O_4$: C, 62.24; H, 7.60. Found: C, 62.04; H, 7.48.

7-Carbomethoxy-*cis*-1-oxa-4-decalone (5b). This epimer mixture was prepared from isomer mixture **2b** in 67% yield by the method used for **5c**. A pure product was obtained without chromatography: ir (neat) 1730 cm^{-1} (C=O); nmr δ 3.87–4.33 (m, 2, H-2), 3.68 (s, 3, OCH_3), 2.71 (m, H-9a), 2.45 (t, J = 6 Hz, H-3); glpc retention time (225°) 9.4 min (only one peak observed).

Anal. Calcd for $C_{11}H_{16}O_4$: C, 62.24; H, 7.60. Found: C, 62.14; H, 7.60.

***cis*-1-Oxa-4-decalone (5a).** This compound was prepared from the isomer mixture **2a** by the method used for **5c**. Chromatography on silica gel (Woelm) of the 1.61 g of crude product obtained gave 670 mg (45%) of **5a** in the 2% ethyl acetate–methylene chloride fractions as an oil: ir (neat) 1730 cm^{-1} (C=O); glpc retention time (180°) 3.9 min.

Anal. Calcd for $C_9H_{14}O_2$: C, 70.10; H, 9.15. Found: C, 70.01; H, 9.15.

Equilibration of 6-Carbomethoxy-1-oxa-4-decalone (5c). A solution of 640 mg (3.02 mmol) of isomer mixture **5c** and 0.6 ml of 1,5-diazabicyclo[4.3.0]non-5-ene in 35 ml of dry benzene was refluxed for 18.5 hr. The cooled solution was extracted with 3 *N* HCl and the aqueous phase was washed with benzene. The combined benzene extracts were washed with saturated sodium bicarbonate and dried ($MgSO_4$). The benzene extract was concentrated, giving 560 mg (88%) of an equilibrated mixture of the three isomers: glpc retention times (225°) 8.4, 10.2, and 11.1 min (6:3:1).

Anal. Calcd for $C_{11}H_{16}O_4$: C, 62.24; H, 7.60. Found: C, 62.09; H, 7.52.

The mixture was recrystallized from carbon tetrachloride–petroleum ether, then twice more from hexane, giving 40 mg (7% recovery) of primarily **11**: mp 73–76°; ir (Nujol) 1715 cm^{-1} (C=O); glpc retention times (225°) 8.4, 10.2, and 11.1 min (9:1:trace).

Table III
Coupling Constants of 1-Oxadecalins[a]

Compd	$J_{2a,2e}$[b]	$J_{2a,3a}$	$J_{2a,3e}$	$J_{2e,3a}$	$J_{2e,3e}$	$J_{3a,3e}$	$J_{5a,10}$	$J_{5e,10}$	$J_{9,10}$	$J_{8a,9}$	$J_{8e,9}$	Other
5a	11	11	3.5	7	2.5	15	8	3	7.5	H-9	$W_{1/2} < 13$	
7	11.5	11.5	3.5	7	1.5	14	13	4.5	10	10	4	
8	11	11	3.5	7	2	14.5	12	4.5	10	10	4	
9	10									H-9	$W_{1/2} = 18$	$J_{6a,7a} = J_{7a,8a} = 12$ $J_{6e,7a} = J_{7a,8e} = 3.5$
11	11	11	3.5	7	2				10	10	4.5	
12	10									H-9	$W_{1/2} = 18$	
13	10											

[a] In hertz. [b] e and a have been used to designate equatorial and axial protons.

Anal. Calcd for $C_{11}H_{16}O_4$: C, 62.24; H, 7.60. Found: C, 62.41; H, 7.52.

Equilibration of 7-Carbomethoxy-*cis*-1-oxa-4-decalone (5b). Equilibration was carried out as with **5c**, giving, after one recrystallization from hexane, a 36% yield of **8**: mp 104–106°; ir (Nujol) 1735 cm^{-1} (C=O); glpc retention time (225°) 9.4 min (only one peak observed).

Anal. Calcd for $C_{11}H_{16}O_4$: C, 62.24; H, 7.60. Found: C, 62.41; H, 7.62.

Equilibration of *cis*-1-Oxa-4-decalone (5a). Equilibration was carried out as with **5c**, giving a 90% yield of *trans*-1-oxa-4-decalone (**7**) as an oil. The analytical sample was chromatographed on silica gel (Woelm) using 2% ethyl acetate–methylene chloride: glpc retention time (180°) 3.9 min.

Anal. Calcd for $C_9H_{14}O_2$: C, 70.10; H, 9.15. Found: C, 70.20; H, 9.13.

6-Carbomethoxy-*trans*-1-oxadecalin (12). A mixture of 60 mg (0.3 mmol) of **5c** (*ca.* 80% trans isomer), 60 mg (0.3 mmol) of *p*-toluenesulfonylhydrazide, 10 mg of *p*-toluenesulfonic acid hydrate, 0.7 ml of sulfolane, and 0.7 ml of dry dimethylformamide was stirred at room temperature for 1 hr. The mixture was then treated with 70 mg (1.1 mmol) of sodium cyanoborohydride and was stirred in a 100–105° bath for 2 hr more. The cooled solution was diluted with water and extracted with cyclohexane. The cyclohexane extracts were washed three times with water and dried ($MgSO_4$). Concentration gave 40 mg (70%) of primarily **12** as an oil: ir (neat) 1748 cm^{-1} (C=O); glpc retention times (175°) 7.5 and 8.9 min (85:15).

Application of this procedure to 350 mg (1.6 mmol) of **5c** (*ca.* 50% trans isomer) gave 230 mg (72%) of a mixture of **12** and **13** (1:1). The isomers were partially separated by preparative glpc. The trans isomer (**12**) was obtained in 80% purity: glpc retention times (175°) 7.5 and 8.9 min (4:1).

Anal. Calcd for $C_{11}H_{18}O_3$: C, 66.62; H, 9.15. Found: C, 66.78; H, 9.27.

The cis isomer (**13**) was also obtained in 80% purity: glpc retention times (175°) 7.5 and 8.9 min (1:4).

Anal. Calcd for $C_{11}H_{18}O_3$: C, 66.62; H, 9.15. Found: C, 66.83; H, 9.25.

7-Carbomethoxy-*trans*-1-oxadecalin (9). This compound was prepared from **8** in 76% yield by the method used for **12.** Material of greater than 99% purity was obtained as a low-melting solid (melts near room temperature) by chromatography on silica gel (Woelm) using 2% ethyl acetate–methylene chloride: ir (neat) 1725 cm^{-1} (C=O); glpc retention time (175°) 9.4 min (only one peak observed).

Anal. Calcd for $C_{11}H_{18}O_3$: C, 66.62; H, 9.15. Found: C, 66.74; H, 9.21.

Application of this procedure to 340 mg (1.6 mmol) of the mother liquors from the recrystallization of **8** gave 150 mg (47%) of an isomer mixture: nmr δ 3.67 (s, OCH_3); glpc retention times (175°) 7.0, 8.3, and 9.4 min (16:28:56).

Base-Catalyzed Equilibrations of 9. A solution of 18 mg of **9** [glpc (175°) indicated 98% of the isomer with retention time of 9.4 min] in 2 ml of 2.5 *N* $NaOCH_3$–CH_3OH was refluxed under N_2 for 7 days. The cooled solution was treated with 3 ml of 0.3 *N* HCl and 2 ml of brine and extracted with cyclohexane. The cyclohexane extracts were dried ($MgSO_4$) and concentrated to give only 0.7 mg of material which was not further investigated.

When the equilibration was repeated as above, with a reflux period of 1 hr, a 76% recovery was realized. The recovered material contained 13% of a new glpc peak at 7.0 min (175°) in addition to 87% of the starting isomer peak. Gas chromatography–mass spectra showed that this new peak was an isomer of **9**: new peak mass spectrum *m/e* (rel intensity) 198 (30), 167 (15), 139 (23), 111 (100), 97 (59), 84 (7); starting isomer peak mass spectrum *m/e* (rel intensity) 198 (12), 167 (14), 139 (32), 111 (55), 97 (100), 84 (12). On extending the reflux time to 4 hr only a 14% recovery of material which contained 21% of the new peak was possible.

Application of the equilibration technique used for ketones **5** led to recovered starting isomer with none of this new peak present.

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Registry No.—**1a,** 491-37-2; **2a,** 51599-61-2; **2b,** 41118-34-7; **2c,** 41118-27-8; **3c** cis epimer 1, 51600-14-7; **3c** cis epimer 2, 51600-15-8; **4a,** 1124-63-6; **5a,** 51600-16-9; **5b** cis epimer 1, 51600-17-0; **5b** cis epimer 2, 51600-18-1; **5c,** 51599-62-3; **7,** 51600-19-2; **8,** 51599-63-4; **9,** 51600-20-5; **10,** 51600-21-6; **11,** 51600-22-7; **12,** 51600-23-8; **13,** 51600-15-8.

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