electrophilic catalysis[21] is a very rare phenomenon indeed. The result provides compelling justification for the notion[1,2] that a "through-three-bond" electron delocalization mechanism (represented by canonical structures **2** and **3** is a strong regulator of the $^{19}F$ SCS of system **1**.

## Experimental Section

**General Methods.** Melting and boiling points are uncorrected. All distillations were performed on a Kugelrohr apparatus (Büchi GKR-50).

**1-Fluoro-4-(trimethylsilyl)bicyclo[2.2.2]octane (1, X = Si(CH$_3$)$_3$).** By use of a procedure outlined by Shippey and Dervan,[23] a solution of 1-fluoro-4-iodobicyclo[2.2.2]octane (1, X = I[22]; 1.0 g, 0.004 mol) in dry hexamethylphosphoric triamide (HMPA; 10 mL) containing freshly prepared sodium methoxide (0.7 g, 0.013 mol) was treated dropwise with hexamethyldisilane (1.7 g, 0.012 mol) at 0 °C under a nitrogen atmosphere. After addition was complete the reaction mixture was allowed to warm to room temperature and then stirred for 24 h. A standard workup followed by Kugelrohr distillation of the crude product afforded the silicon compound (1, X = $SiMe_3$) as a white solid which recrystallized from methanol as needles (0.2 g, 33%): mp 72–73 °C; $^1H$ NMR ($CDCl_3$) $\delta$ –0.13 (9 H, s, $Si(CH_3)_3$), 1.80–1.83 (12 H, m, $CH_2CH_2$); $^{13}C$ NMR ($CDCl_3$, relative to $Me_4Si$) $\delta$ 95.21 ($J_{CF}$ = 181.64 Hz; C1), 31.30 ($J_{CF}$ = 17.21 Hz; C2), 28.09 ($J_{CF}$ = 8.42 Hz; C3), 16.77 ($J_{CF}$ = 1.83 Hz; C4), –4.64 ($J_{CSi}$ = 50.17 Hz; $SiCH_3$). Anal. Calcd for $C_{11}H_{21}FSi$: C, 65.95; H, 10.55. Found: C, 65.81; H, 10.60.

**1-Fluoro-4-(trimethylgermyl)bicyclo[2.2.2]octane (1, X = Ge(CH$_3$)$_3$).** A HMPA solution of the fluoro iodide (1, X = I[22]; 1.0 g, 0.004 mol) was added to (trimethylgermyl)lithium (prepared from $Me_3GeBr$ (0.8 g, 0.004 mol) and lithium (0.1 g, 0.0142 mol) as described in the literature[24]) in HMPA (15 mL). The reaction mixture was allowed to stir for 2 h at room temperature before being worked up in the usual manner. Kugelrohr distillation (100 °C/2 mm) of the crude product and then recrystallization from methanol afforded a white solid (0.2 g, 42%): mp 71 °C; $^1H$ NMR ($CDCl_3$) $\delta$ 0.10 (9 H, s, $GeCH_3$), 1.73–1.80 (12 H, m, $CH_2CH_2$); $^{13}C$ NMR ($CDCl_3$, relative to $Me_4Si$) $\delta$ 94.82 ($J_{CF}$ = 181.27 Hz; C1), 31.73 ($J_{CF}$ = 16.84 Hz; C2), 29.13 ($J_{CF}$ = 8.79; C3), 20.45 ($J_{CF}$ = 2.20; C4), –5.48 ($GeCH_3$). Anal. Calcd for $C_{11}H_{21}FGe$: C, 53.94; H, 8.66. Found: C, 54.16; H, 8.89.

**1-Fluoro-4-(trimethylplumbyl)bicyclo[2.2.2]octane (1, X = Pb(CH$_3$)$_3$).** By use of the procedure of Williams,[25] a solution of (trimethylplumbyl)magnesium chloride (TPMC) was prepared by adding dry powdered lead chloride (1.49 g, 0.005 mol) to a well-stirred THF solution of methylmagnesium chloride (20 mL of 0.02 M solution, 0.02 mol) at 0 °C under an atmosphere of nitrogen. After the reagent mixture was stirred at 0 °C for 2 h, a THF solution of the fluoro iodide (1, X = I[22]; 1.5 g, 0.0059 mol) was then added slowly while maintaining the temperature at 0 °C. The mixture was then stirred at that temperature for 5 h and then an additional 16 h at room temperature before quenching with a saturated aqueous solution of ammonium chloride. A standard workup followed by two careful Kugelrohr distillations (100 °C/1 mm) of the crude product afforded the lead compound 1 (X = $Pb(CH_3)_3$ as a colorless liquid (0.8 g, 35.7%): $^1H$ NMR ($CDCl_3$) $\delta$ 0.53 (9 H, s, $PbCH_3$; $J_{Pb-H}$ = 48 Hz), 2.17 (12 H, m, $CH_2CH_2$); $^{13}C$ NMR ($CDCl_3$, relative to $Me_4Si$) $\delta$ 92.94 ($J_{CF}$ = 182.38 Hz, $J_{CPb}$ = 21.61 Hz; C1), 33.77 ($J_{CF}$ = 16.48 Hz, $J_{CPb}$ = 102.73 Hz; C2), 34.05 ($J_{CF}$ = 8.06 Hz; C3), 34.34 ($J_{CF}$ = 2.2 Hz, $J_{CPb}$ = 458.13 Hz; C4), –4.79 ($J_{CPb}$ = 161.87 Hz; $PbCH_3$).

An elemental analysis was not sought. The compound slowly disproportionated over a period of 3 months.

**Treatment of 1-Fluoro-4-iodobicyclo[2.2.2]octane (1, X = I) with *tert*-Butyllithium.** A solution of the iodide (1 (X = I[22]; 0.7 g, 2.8 mmol) in dry diethyl ether (7 mL) was cooled to –80 °C and treated with 6 mL of 1.5 M *tert*-butyllithium (9 mmol) in pentane. After 10 min, a solution of 4.3 g of bromine in diethyl ether (19 mL) was slowly added and the reaction mixture allowed to warm to room temperature. After a standard workup, almost quantitative amounts of 1,4-bis(bromomethyl)-1,4-dibromocyclohexane was obtained as a brown solid. A sample was recrystallized from hexane/diethyl ether (2:1) to afford prisms: mp 129–132 °C (lit.[26] mp 134–136 °C; $^1H$ NMR ($CDCl_3$) $\delta$ 1.86–2.66 (8 H, m, $CH_2CH_2$), 4.00 (4 H, s, $CH_2Br$); $^{13}C$ NMR ($CDCl_3$, relative to $Me_4Si$) $\delta$ 35.25 (C2, 3, 5, and 6), 43.43 ($CH_2Br$), 69.12 (C1 and 4). No 1,4-dibromobicyclo[2.2.2]octane, indicative of the formation of [2.2.2]propellane,[27] was detected in the crude reaction product either by $^1H$ and $^{13}C$ NMR or GLPC. Furthermore, no 1-bromo-4-fluorobicyclo[2.2.2]octane was detected, indicating that lithiation and decomposition was complete. The former conclusion follows from recent studies which showed that the replacement of I by Br in the bicyclo[2.2.2]octane ring system is facile even at low temperature.[27]

An experiment was also performed in which, after lithiation (10 min), the reaction mixture at –80 °C was quenched quickly with excess dry $CO_2$ gas. No 4-fluorobicyclo[2.2.2]octane-1-carboxylic acid[22] was isolated from the reaction, confirming the rapid and complete decomposition of the lithium derivative (1, X = Li).

**NMR Spectra.** The $^{13}C$ and $^{19}F$ NMR spectra were obtained as described in previous papers.[1,2,3b] $^1H$ NMR spectra were measured with a Varian EM-360 (60 MHz) spectrometer on $CDCl_3$ solutions.

**Registry No.** 1 (X = $CMe_3$), 81687-86-7; 1 (X = $SiMe_3$), 95552-61-7; 1 (X = $GeMe_3$), 95552-62-8; 1 (X = $SnMe_3$), 78385-88-3; 1 (X = $PbMe_3$), 95552-63-9; 1 (X = I), 78385-89-4; TPMC, 51258-94-7; 1,4-bis(bromomethyl)-1,4-dibromocyclohexane, 62947-52-8.

(21) (a) The facile loss of fluoride from fluoroalkanes in the presence of strong Lewis acids is well-known.[21b-f] (b) See ref 23 in ref 22. (c) Schleyer, P. v. R.; Fort, R. C.; Watts, W. E.; Comisarow, M. B.; Olah, G. A. *J. Am. Chem. Soc.* **1964**, *86*, 4195. (d) Olah, G. A.; Baker, E. B.; Evans, J. C.; Tolgyesi, W. S.; McIntyre, J. S.; Bastien, I. J. *J. Am. Chem. Soc.* **1964**, *86*, 1360. (e) Pattison, F. L. M.; Howell, W. C. *J. Org. Chem.* **1956**, *21*, 879. (f) Kemppainer, A. E.; Thomas, M. J.; Wagner, P. J. *J. Org. Chem.* **1976**, *41*, 1294.

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# Delocalized Carbanions: The Facile Synthesis of Disubstituted Butadienes and Diols

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The synthetic utility of delocalized carbanions resulting from the abstraction of a proton $\alpha$ to a double bond has been established[1] by using strong metalating agents such as *n*-butyllithium/tetramethylethylenediamine (*n*-BuLi/TMEDA)[2,3] or Lochmann's base system (*n*-BuLi/potassium *tert*-butoxide in pentane).[4-6] Lochmann's base system has been shown to cause allylic metalation of conjugated dienes, with the notable exception of isoprene

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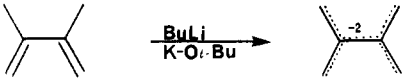
Table I. $^1$H NMR Spectra of 2,3-Disubstituted 1,3-Butadienes and 2,3-Dialkyl-1,4-butanediols

| 3c | 3d | 3e | 4 | 5a | 5b |
|---|---|---|---|---|---|
| 0.9 (t, 6 H) | 0.9 (t, 6 H) | 0.8 (t, 6 H) | 1.7 (m, 4 H) | 0.9 (d, 6 H) | 0.8 (d, 6 H) |
| 1.5 (m, 12 H) | 1.4 (s, 44 H) | 1.2 (s, 68 H) | 2.3 (t, 4 H) | 1.8 (bs, 2 H) | 1.4 (s, 72 H) |
| 2.3 (t, 4 H) | 2.3 (t, 4 H) | 2.3 (t, 4 H) | 3.0 (s, 2 H) | 3.5 (d, 4 H) | 1.9 (t, 2 H) |
| 4.8 (s, 2 H) | 4.8 (s, 2 H) | 4.9 (s, 2 H) | 4.9 (s, 2 H) | 4.9 (s, 2 H) | 2.5 (d, 4 H) |
| 5.0 (s, 2 H) | 5.1 (s, 2 H) | 5.0 (s, 2 H) | 5.1 (s, 2 H) | | 3.5 (s, 2 H) |

where the butyl anion adds to the conjugated diene instead of abstracting the $\alpha$ proton.[4] However, metalation of 2,3-dimethyl-1,3-butadiene (1) proceeds cleanly and




1 2

quickly (10 min) to dianion 2 in yields of greater than 80%.[4] A synthetic scheme for the preparation of 2,3-disubstituted 1,3-butadienes and 1,4-butanediols has been identified as a desirable goal.[7] These compounds are useful in preparing 3,4-disubstituted tetrahydrofurans, their polymers, disubstituted polybutadienes, and condensation polymers.[7]

## Results and Discussion

Generally, delocalized carbanions react readily with primary halides, epoxides, primary sulfonates, and carbonyl groups in an $S_N2$ manner.[8,9] The reaction of the 2,3-dimethylenebutadiene dianion 2 with alkyl halides and ethylene oxide has been shown to go rapidly and in high yields (see Scheme I). The synthesized compounds have been characterized by $^1$H NMR (Table I) and elemental analysis. This procedure is very facile for the preparation of 2,3-disubstituted 1,3-butadienes as compared to a recently published procedure.[7]

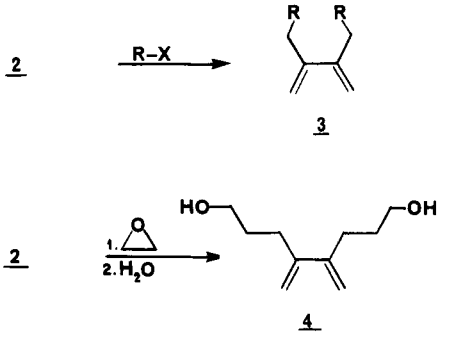
Brown and co-workers[10,11] have shown that 1,3-conjugated dienes can be converted to 1,4-diols via hydroboration. By this method, the 2,3-dialkyl-1,4-butanediols are easily prepared in high yields (see Scheme II).

## Experimental Section

$^1$H Nuclear magnetic resonance (NMR) spectra were recorded on a Varian EMS 360 spectrometer. Chemical shifts are reported in parts per million downfield from tetramethylsilane ($Me_4Si$). Elemental analyses were performed by Galbraith Laboratories, Knoxville, TN. Melting points were taken on a National Instrument Co., Inc. melting point apparatus and are uncorrected. Volatile products were isolated and purified by gas chromatography with a Varian Aerograph Series 1800 chromatograph equipped with a 6 m × 0.25 in. Chromosorb 20 m packed column. Tetrahydrofuran (THF) was distilled from sodium with benzophenone as an indicator (blue anion radical). Pentane was washed with sulfuric acid, distilled from $LiAlH_4$, and stored under nitrogen. Ethylene oxide was obtained from Matheson and distilled prior to use. Potassium *tert*-butoxide and *n*-butyllithium solution in hexane (2.2–2.6 M) were used as received from Alpha Division, Ventron Corp., and all other reagents were purified by standard methods.

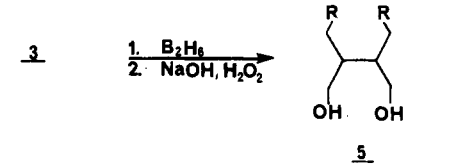
**2,3-Dimethylene-1,3-butadiene Dianion (2).** Dianion 2 was prepared by adding 1.1 mL (10 mmol) of 2,3-dimethyl-1,3-butadiene (1) in 20 mL of pentane to a mixture of 2.24 g (20 mmol) of potassium *tert*-butoxide, 20 mL pentane, and 9.6 mL (20 mmol) of 2.2 M *n*-butyllithium in hexane in a nitrogen-filled flask. After 10 min of stirring, the dianion salt was allowed to settle, and the liquid was removed and blown dry with nitrogen. For reaction, it was suspended in 20 mL of THF and added to the reaction via syringe.

Scheme I. Preparation of 2,3-Dialkyl-1,3-butadiene Compounds




| | R | % yield |
|---|---|---|
| 3a | $CH_3$ | 78[4] |
| 3b | $CH_2CH_3$ | 80[4] |
| 3c | $(CH_2)_3CH_3$ | 60 |
| 3d | $(CH_2)_{11}CH_3$ | 74 |
| 3e | $(CH_2)_{17}CH_3$ | 78 |
| 4 | | 52 |

Scheme II. Preparation of 1,4-Butanediols




| | R | % yield |
|---|---|---|
| 5a | H | 60 |
| 5b | $(CH_2)_{17}CH_3$ | 75 |

**2,3-Dipentyl-1,3-butadiene (3c).** To the dianion flask prepared as previously described was added 2.75 g (20 mmol) of *n*-butyl bromide followed by dropwise addition of dianion 2 in 20 mL of THF. The solution was stirred while warming to room temperature. After 1 h, the solution was extracted with pentane. The organic phase was dried over magnesium sulfate and the product was purified by preparative GC in a 60% yield. Anal. Calcd for $C_{14}H_{26}$: C, 86.60; H, 13.40. Found: C, 86.46; H, 13.53.

**2,3-Ditridecyl-1,3-butadiene (3d)** was prepared in the same manner as **3c** with 20 mmol of 1-bromododecane in 74% yield after recrystallization from ether (mp 37–38 °C). Anal. Calcd for $C_{30}H_{58}$: C, 86.12; H, 13.78. Found: C, 86.31; H, 13.68.

**2,3-Dinonadecyl-1,3-butadiene (3e)** was prepared in the same manner as **3c** with 20 mmol of 1-bromooctadecane in 75% yield after recrystallization from ether (mp 62–63 °C). Anal. Calcd for $C_{42}H_{82}$: C, 85.92; H, 14.04. Found: C, 85.75; H, 14.24.

**4,5-Dimethylene-1,8-octanediol (4).** The flask containing dianion 2 was cooled to −78 °C (dry ice/acetone bath) and with a cooled syringe 20 mmol of ethylene oxide was added while stirring was continued as the reaction warmed to room temperature. The reaction was quenched with 4 mL of distilled water. Subsequent workup for the reaction was the same for **3c** in a 52% yield after vacuum distillation (130 °C/0.09 mm).

**2,3-Dimethyl-1,4-butanediol (5a).** 2,3-Dimethylbutadiene, 50 mmol, was added to 50 mL of THF and placed in an ice bath,

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and 25 mL of a 1 M solution $B_2H_6$ in THF was added dropwise. After warming to room temperature, the excess hydride was decomposed by dropwise addition of water. The organoborane was oxidized at 30–50 °C (water bath) by the addition of 12 mL of 3 N NaOH, followed by dropwise addition of 12 mL of 30% $H_2O_2$. After 1 h 30 g of potassium carbonate was added. The THF layer was separated, and the aqueous phase was extracted with THF. The THF extracts were combined and dried. Following the removal of the solvent by rotary evaporation, the product was purified by vacuum distillation (bp 105–106 °C/2 mm) in a 60% yield.

**2,3-Dinonadecyl-1,4-butanediol (5b)** was prepared in the same manner as **5a** by using 0.85 mmol of **3e** in 75% yield after recrystallization from ether (mp 60–61 °C. Anal. Calcd $C_{42}H_{86}O_2$: C, 80.95; H, 13.91; O, 5.13. Found: C, 80.75; H, 13.71; O, 5.34.

**Acknowledgment.** We acknowledge financial support from the National Science Foundation, Polymers Program (Grant No. DMR 8214211).

**Registry No.** **1**, 513-81-5; **2**, 69780-62-7; **3c**, 84652-75-5; **3d**, 92882-24-1; **3e**, 92882-25-2; **4**, 92739-56-5; **5a**, 57716-80-0; **5b**, 92882-26-3; *n*-butylbromide, 109-65-9; 1-bromododecane, 143-15-7; 1-bromooctadecane, 112-89-0; ethylene oxide, 75-21-8; 2,3-dimethyl-1,3-butadiene, 513-81-5.

## Michael Addition to 1,3-Bis(alkoxycarbonyl)allenes: Synthesis of Heterocyclic Compounds Having Glutaconate Structure in the Molecules

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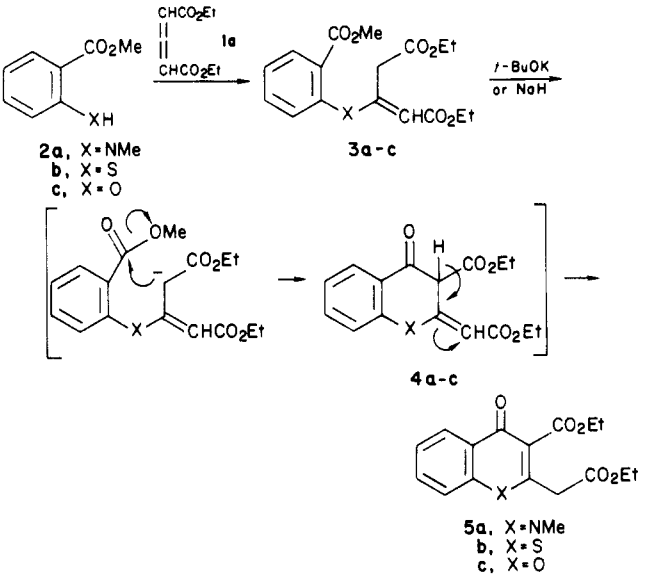
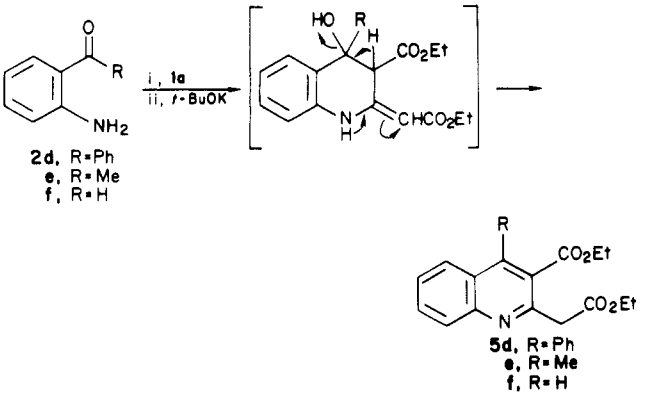
The use of 1,3-dicarbalkoxyallene **1** in the Diels–Alder reaction has been proven to be valuable for the construction of cyclic compounds containing a glutaconate system.[1] Although **1** is also expected to be a useful receptor toward Michael addition, the synthetic applications of such allenes have been limited to a few examples.[2,3] A recent paper[3] on the synthetic utility of **1** by Michael reaction with salicylaldehyde, *o*-hydroxyacetophenone, and methyl salicylate prompted us to publish our own results on the Michael reaction of **1** with a variety of compounds having both nucleo- and electrophilic functional groups. This method provides a versatile route to heterocycles containing the glutaconate system.[4] This approach for the synthesis of these heterocycles is based on the presumption that compounds **2** having both nucleo- and electrophilic substituents at appropriate positions would first nucleophilically add to the allenic bond of 1,3-dicarbalkoxyallene **1** to form Michael-type adducts **3**. Subsequent base-catalyzed intramolecular cyclization to the electrophilic group of **3**, followed by isomerization under the conditions used, then would give the desired heterocycles **5** (Scheme I).

Reaction of equimolar amounts of methyl *N*-methylanthranilate (**2a**, X = NMe) with 1,3-dicarbethoxyallene (**1a**) in chlorobenzene at 100 °C for 30 min gave an adduct (**3a**) which, when treated with 1 equiv of potassium *tert*-butoxide at 0 °C for 5 min, resulted in the loss of the methoxy group to provide ethyl 3-carbethoxy-1-methyl-4(1*H*)-quinolinone-2-acetate (**5a**) in 65% overall yield. The double bond of the product appears to be in the endocyclic form (**5a**) rather than the tautomeric exocyclic form (**4a**) since the NMR spectrum exhibited two equivalent protons attributable to a $CH_2CO_2Et$ group at 3.95 ppm. In the case of methyl thiosalicylate (**2b**, X = S) and methyl salicylate (**2c**, X = O), the reactions were accomplished at 0 °C by treatment with equimolar amounts of **1a** in tetrahydrofuran (THF) in the presence of 1 equiv of NaH to give the corresponding chromone-2-acetates (**5b** and **5c**). The use of *tert*-butoxide instead of NaH in the reaction of **2b**,**c** with **1a** gave lower yields.




*o*-Aminobenzophenone (**2d**), *o*-aminoacetophenone (**2e**), and *o*-aminobenzaldehyde (**2f**) reacted with **1a** in a similar




fashion as described for the reaction of **2a** with **1a** except that the cyclization was followed by dehydration. Thus, condensation of **2d–f** with **1a** in chlorobenzene at 80 °C for 30 min gave the adducts, which were treated with 1

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