was neutralized with solid sodium bicarbonate and maintained faintly alkaline throughout the reaction period by further additions when needed.[7] After 4 days at room temperature 3.7 g of **4** had crystallized. Further crops of about this magnitude could be obtained by filtration every few days, but the ultimate yield was not determined: 140–155° dec, depending on the rate of heating; nmr (CD_3SOCD_3), δ 7.20 (broad s, 2, vanished on shaking with D_2O), 5.65 (d, 2, vanished on shaking with D_2O, >CHOH), 4.95 (broad s, 2, sharpened on shaking with D_2O, >CH–CH<), 4.05 (q, 4), and 1.20 (t, 6).

Anal. Calcd for $C_8H_{16}N_2O_6$: C, 40.70; H, 6.82; N, 11.80. Found: C, 40.43; H, 7.03; N, 11.92.

1,1,2-Tris(ethoxycarbonylamino)-2-hydroxyethane (5a).—Ethyl carbamate (15.0 g, 0.168 mol), 30% aqueous glyoxal solution (8.2 g, 0.042 mol), and 0.5 ml of hydrochloric acid were dissolved in water, and left at room temperature (warming of the solution resulted in formation of **1a**). After 5 days 1.3 g of **5a** was collected (precipitation continued in the filtrate) and recrystallized from water and then from ethyl acetate: mp 140–160° dec, depending on the rate of heating; nmr (CD_3SOCD_3), δ 7.1 (broad, 3, vanished on shaking solution with D_2O, NH), 5.91 (d, 1, vanished on shaking solution with D_2O, –OH), 5.05 (broad, 2, sharpened on shaking the solution with D_2O, >CH–CH<), 3.98 (q, 6), and 1.14 (t, 9). At 220 MHz further resolution was possible: δ 7.38 (s, 1, –CH(OH)NH–), 7.31 (s, 2, –NHCHNH–), 1.178 (t, ≃3, J = 7 Hz, $CH_3CH_2CO_2NHCH$–OH–), and 1.173 (t, ≃6, J = 7 Hz ($CH_3CH_2CO_2NH)_2CH$–): exchange of labile protons with D_2O simplified the resonances corresponding to >CH–CH<; 5.05 (d, 1, J = 5 Hz) and 5.01 (d, 1, J = 5 Hz).

Anal. Calcd for $C_{11}H_{21}N_3O_7$: C, 42.99; H, 6.89; N, 13.67; O, 36.45. Found: C, 42.95; H, 6.82; N, 13.70; O, 37.5.

Registry No.—**1a**, 17350-57-1; glyoxal, 107-22-2; ethyl carbamate, 51-79-6; **4**, 17350-58-2; **5a**, 17350-59-3.

Acknowledgments.—The author is indebted to Mr. B. W. Cook and Mr. M. C. McIvor for obtaining and helping to interpret the spectroscopic data and to Mr. T. Carman for experimental assistance.

Reaction of 2,3-Dihydrobenzo[*b*]thiophen-3(2H)-one 1,1-Dioxide with Electrophiles

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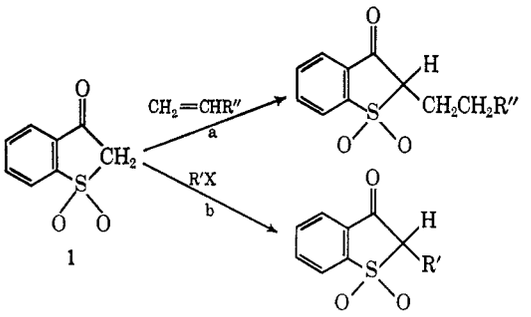
Although 2,3-dihydrobenzo[*b*]thiophen-3(2H)-one 1,1-dioxide (**1**) has been known for some time,[1-4] no examples of alkylation or acylation of **1** have been reported. A few 2-acyl and 2-alkyl derivatives of **1** are known,[5-7] but the synthesis of these compounds has been accomplished either by cyclization of *o*-alkylsulfonylbenzoate esters[5,6] or by chlorosulfonation of propiophenones.[7]

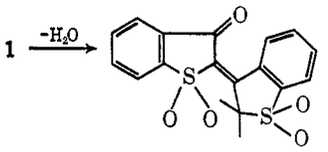
In connection with another study, a more versatile route to certain 2-alkyl and 2-acyl derivatives of **1** was sought. Toward this end, the acylation and alkylation of **1**, as well as the Michael-type additions of **1**



1

(anion) to unsaturated electrophiles, were studied. With reactive electrophiles such as acrylonitrile and methyl vinyl sulfone, sodium methoxide successfully served as a base for the Michael addition of **1** (reaction a). Table I (**12–15**) records products of this reaction.

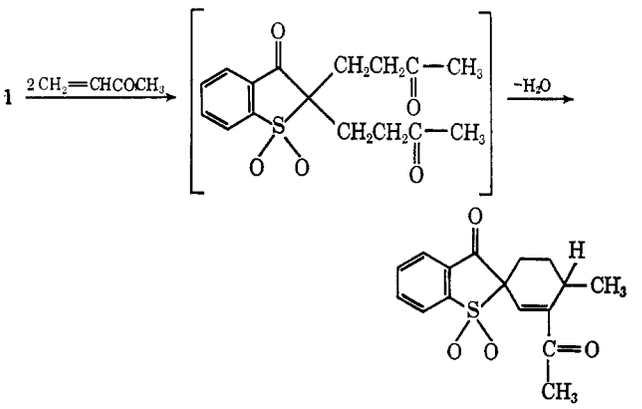
In a few attempts in which alkyl halides were allowed to react with **1** in the presence of sodium methoxide, complex mixtures were obtained. This apparently slower reaction may have allowed side reactions such as ring opening of **1** by methoxide ion to occur, since hydroxide ion is known[2] to cleave **1** into 2-methylsulfonylbenzoic acid. An analogous example of ring cleavage of **1** by pyrrolidine is discussed below. It was ultimately found that alkylations and acylations of **1** proceeded in moderate yields in the presence of the sterically hindered diisopropylethylamine[8] in isopropyl alcohol solution (reaction b above). Table I (**3–11**) lists products from this reaction. Side reactions were minimized in this system, although intermolecular dehydration of **1** to the dimer **2** was observed when relatively unreactive alkyl halides such as 1-bromopentane or chloroethyl methyl sulfide were employed.



2

Analogous self-condensation of 1,3-indandione to form an anhydro dimer ("Bindone") in the presence of base has previously been observed.[9]

Reaction of methyl vinyl ketone with **1** in the presence of sodium methoxide gave a low yield of the spiro compound **17**, probably proceeding through the re-



17

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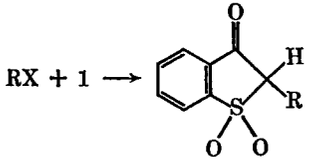
TABLE I

2-SUBSTITUTED 2,3-DIHYDROBENZO[*b*]THIOPHEN-3(2H)-ONE 1,1-DIOXIDES

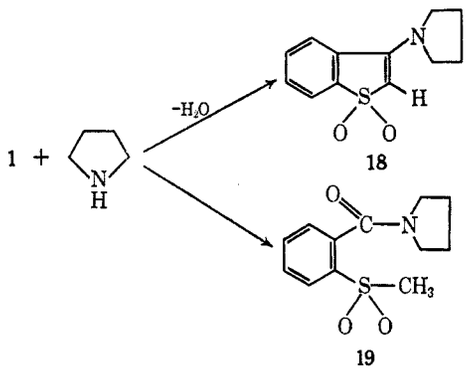


No.	R	X (or reactant)	Reaction conditions[a]	Yield, %[b]	Mp, °C	Formula	Calcd, % C	Calcd, % H	Calcd, % N	Found, % C	Found, % H	Found, % N
3	$CH_2COC_6H_5$	Cl	A[c]	24	123–125	$C_{16}H_{12}O_4S$	63.98	4.03		63.76	4.32	
4	$CH_2CO(4\text{-}Br)C_6H_4$	Br	A	38	187–188	$C_{16}H_{11}BrO_4S$	50.67	2.92		50.76	2.92	
5	$CH_2CO(4\text{-}NO_2)C_6H_4$	Br	A	35	217 dec	$C_{16}H_{11}NO_6S$	55.65	3.21	4.06	55.90	3.18	4.08
6	CH_2COOEt	Br	A	27	113–115	$C_{12}H_{12}O_5S$	53.72	4.51		53.77	4.76	
7	CH_2COCH_3	Cl	A	33	116–118	$C_{11}H_{10}O_4S$	55.45	4.23		55.50	4.41	
8	$CONHC_6H_5$	(C_6H_5NCO)	B[c]	18	178–180	$C_{15}H_{11}NO_4S$	59.79	3.68	4.65	59.71	3.70	4.90
9	$COCH_3$	(Ac_2O)	*C*	50	163–164[d]							
10	COOEt	Cl	C	56	139–141[e]							
11	$COO(CH_2)_5CH_3$	Cl	B	46	63–64	$C_{15}H_{18}O_5S$	58.05	5.84		58.27	6.07	
12	CH_2CH_2CN	($CH_2{=}CHCN$)	D[c]	28	349 dec[f]	$C_{11}H_8NO_3SNa$	51.35	3.13	5.45	51.04	3.20	5.15
13	$CH_2CH_2SO_2CH_3$	($CH_2{=}CHSO_2CH_3$)	D[c]	31	163–165	$C_{11}H_{12}O_5S_2$	45.82	4.20		45.53	4.16	
14	$CH_2CH_2COCH_3$	($CH_2{=}CHCOCH_3$)	D[g]	6	227 dec	$C_{12}H_{12}O_4S$	57.12	4.80		57.00	4.94	
15	$CH(Ph)CH_2COCH_3$[h]	($PhCH{=}CHCOCH_3$)	D	50	152–153	$C_{18}H_{16}O_4S$	65.83	4.91		65.54	4.84	
16	$CH(Ph)CH_2COC_6H_5$	($PhCH{=}CHCOPh$)	D	65	168–170	$C_{23}H_{18}O_4S$	70.75	4.65		70.57	4.70	

[a] A = in isopropyl alcohol using diisopropylethylamine as exemplified in the Experimental Section for **3**; B = in chloroform using diisopropylethylamine; C = in ethanol using diisopropylethylamine; D = in absolute ethanol using sodium methoxide as exemplified for **13**. [b] All products were recrystallized from ethanol. [c] See Experimental Section. [d] Lit.[5] mp 164°. [e] Lit.[6] mp 138°. [f] Isolated as sodium salt. [g] A complex mixture of products was purified on thick-layer silica gel plates using C_6H_6–5% acetic acid in two passes. A band at R_f 0.3 was removed and triturated with hot EtOH from which **14** crystallized. [h] Calcd neut equiv, 328. Found 330, $pH_{1/2}$ 7.8.

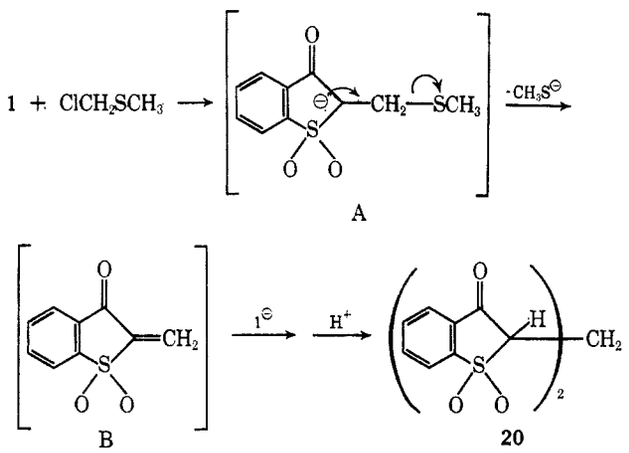
action of **1** with 2 equiv of the ketone followed by an intramolecular Claisen condensation. Another attempt gave the desired monoaddition reaction (**14**, Table I) in a complex mixture which was separable only on thick-layer chromatographic plates.

Formation of an enamine (**18**) from **1** and pyrrolidine proceeded in poor yield due to the competing ring-opening reaction to form the amide **19**. An analogous



cleavage of acyclic β-keto sulfones by pyrrolidine has also been observed.[10] Recently, eneamines related to **18** have been made in good yield from 3-(α-chloroalkyl)-benzo[*b*]thiophene 1,1-dioxides, although experimental details and physical constants were not reported.[11]

When **1** was combined with chloromethyl methyl sulfide in dimethylformamide solution, a mixture of products resulted from which only bis(3-oxo-2,3-dihydrobenzo[*b*]thiophen-2-yl 1,1-dioxide)methane (**20**) could be isolated. This may have formed by elimination of sodium methyl mercaptide from the desired product (A) followed by Michael addition of **1** (anion) to a 2-methylene intermediate (B) derived from **1**. A



2-methylene derivative of **1** has also been proposed[7] as an intermediate in the reaction of hydrazine with 3-chloro-2-dimethylaminomethylbenzo[*b*]thiophen 1,1-dioxide.

Experimental Section[12]

2,3-Dihydrobenzo[*b*]thiophen-3(2H)-one 1,1-dioxide (1) was prepared by the method of Regitz:[4] mp 134–136° (lit.[4] mp 134–135°); neut equiv, 183 (calcd 182); pH ½ 9.1; nmr ($DCCl_3$); τ 5.92 (s, 2, CH_2), 2.1 (m, 4, aromatic protons).

Self-Condensation of 1 to Dimer 2.—An orange solution resulted when a combination of 3.2 g (0.018 mol) of **1**, 2.3 g (0.018 mol) of diisopropylethylamine, 2.7 g (0.018 mol) of 1-bromopentane, and 75 ml of isopropyl alcohol was refluxed for 2 hr. After evaporation to dryness, the residue was dissolved in 100 ml of EtOH–150 ml of H_2O and acidified with 6 *N* HCl to produce a white precipitate: 2.5 g (82%); mp 300° dec; ir, 5.89 (C=O), 6.40, 7.6, 8.65, 8.87 μ.

Anal. Calcd for $C_{16}H_{10}O_5S_2$: C, 55.48; H, 2.91. Found: C, 55.46; H, 3.17.

A product identical with **2** was obtained when **1** was combined with chloroethyl methyl sulfide in isopropyl alcohol solution in the presence of diisopropylethylamine.

2-Phenacyl-2,3-dihydrobenzo[*b*]thiophen-3(2H)-one 1,1-Dioxide (3).—A combination of 3.2 g (0.018 mol) of **1**, 2.32 g (0.018 mol) of diisopropylethylamine, and 60 ml of isopropyl alcohol was warmed to give a yellow solution. A solution of 2.8 g (0.018 mol) of 2-chloroacetophenone in 30 ml of isopropyl alcohol was slowly added, and the solution was refluxed for 3 hr. After standing at room temperature overnight the solution was evaporated to dryness and partitioned between H_2O–ether; the ether extracts were combined, dried (Na_2SO_4), and evaporated. The residue was recrystallized from ethanol giving 1.3 g (24%): mp 122–125°; ir, 5.76 and 5.91 (C=O), 7.66 and 8.56 μ (SO_2); nmr ($DCCl_3$), τ 2.1 and 2.47 (m, 9, aromatic protons), 5.5 (t, 1, J = 4 cps, exchanges with D_2O, the 2 proton), 6.05 (d, 2, J = 4 cps, CH_2). Other physical data are included in Table I.

2,3-Dihydro-3-oxo-N-phenylbenzo[*b*]thiophen-2-carboxamide 1,1-Dioxide (8).—To a solution of 3.2 g (0.018 mol) of **1** in 75 ml of $CHCl_3$ was added a solution of 2.1 g (0.018 mol) of phenyl isocyanate in 15 ml of dry $CHCl_3$. After the addition of 2.3 g (0.018 mol) of diisopropylethylamine in 15 ml of dry $CHCl_3$, the solution was refluxed for 3 hr. After washing twice with HCl, the chloroform layer was dried ($CaSO_4$) and evaporated. Recrystallization from ethanol followed by a thorough ether trituration gave 0.97 g (18%): mp 178–180° dec; ir, 3.0 (NH), 5.80 and 6.23 (C=O), 7.70 and 8.75 μ (SO_2).

2,3-Dihydro-2-acetylbenzo[*b*]thiophen-3(2H)-one 1,1-Dioxide (9).—A solution of 3.2 g (0.018 mol) of **1**, 50 ml of acetic anhydride, and 2.3 g (0.018 mol) of diisopropylethylamine was heated (steam bath) for 0.5 hr. After evaporation to dryness the red residue was triturated with benzene and filtered, and the benzene filtrate was evaporated to dryness. Dissolving the resultant residue in water and acidifying (HCl) produced a yellow solid: 2.0 g (50%), mp 163–164°; ir, 5.77 and 5.87 (C=O), 7.6 and 8.63 μ (SO_2); nmr ($DCCl_3$), τ 7.44 (s, 3, CH_3), 2.2 (m, 4), −3.0 (s, 1 enolic OH); neut equiv, 230 (calcd 224); $pH_{1/2}$ 2.7.

2-(2-Cyanoethyl)-2,3-dihydrobenzo[*b*]thiophen-3(2H)-one 1,1-Dioxide Sodium Salt (12).—To a solution of 4.4 g (0.024 mol) of **1** and 1.4 g (0.026 mol) of acrylonitrile in 70 ml of alcohol was added a solution of 1.4 g (0.025 mol) of $NaOCH_3$ in 20 ml of methanol. After refluxing 3 hr and cooling, a yellow precipitate was filtered, yielding (in two crops) 1.72 g (28%): mp 349° dec; ir, 4.44 (C≡N), 6.24, 6.40, 7.10, 8.04, 8.80 μ; neut equiv, 258 (calcd 257) (HCl titration); $pH_{1/2}$ 7.4.

2-(2-Methylsulfonylethyl)-2,3-dihydrobenzo[*b*]thiophen-3(2H)-one 1,1-Dioxide (13).—A solution of 4.6 g (0.025 mol) of **1**, 1.4 g (0.025 mol) of $NaOCH_3$, and 3.2 g (0.030 mol) of methyl vinyl sulfone in 175 ml of methanol was stirred for 0.5 hr. After refluxing for 1 hr and concentrating to half volume, the solution was poured into 100 ml of HCl. After recrystallization from ethanol, 2.3 g of a yellow solid was obtained: mp 163–165°; ir, 5.75 (C=O), 7.65 and 8.70 μ (SO_2); neut equiv, 292 (calcd 288); $pH_{1/2}$ 7.4.

1′-Acetyl-2,3-dihydro-6′-methyl-3-oxospiro[benzo[*b*]thiophene-2,3′-cyclohexene] 1,1-Dioxide (17).—To a combination of 4.6 g g (0.025 mol) of **1**, 1.4 g (0.025 mol) of $NaOCH_3$, and 150 ml of MeOH was slowly added 2.2 g (0.030 mol) of methyl vinyl ketone in 25 ml of methanol. The deep red solution was refluxed for 1 hr, cooled, and concentrated. Pouring into a mixture of 200 ml of 12 *N* HCl–ice produced a yellow solid, 1.3 g (17%) which, after recrystallization from isopropyl alcohol–water, had mp 199–201°; ir, 5.77 and 5.99 (C=O), 7.69 and 8.41 μ (SO_2); nmr ($DCCl_3$), τ 8.70 (m, 3, the 6′-CH_3), 8.1–8.3 (m, 4, the two CH_2), 7.1–7.4 (m, 4, the 6′-H and CH_3CO), 3.40 (s, 1, the 2′-H), 2.2 (m, 4, aromatic protons).

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(12) Melting points were determined in a Thomas–Hoover capillary melting point apparatus using a calibrated thermometer and were corrected. Potentiometric titrations were carried out in 2:1 dioxane–water using a Beckman Model G pH meter. Nmr spectra were obtained on a Varian Associates A-60 spectrometer with tetramethylsilane as an internal standard. Infrared spectra were determined in potassium bromide pellets.

Anal. Calcd for $C_{16}H_{16}O_4S$: C, 63.14; H, 5.30; S, 10.54. Found: C, 62.84; H, 5.05; S, 10.47.

1-(2-Methylsulfonylbenzoyl)pyrrolidine (19).—A combination of 6.4 g (0.035 mol) of **1**, 100 ml of benzene, and 10 g (0.141 mol) of pyrrolidine was refluxed overnight in a Dean–Stark apparatus for removal of water. After evaporating to dryness, recrystallization from chloroform–hexane gave 1.2 g (14%) of product: mp 113–115°; ir, 6.15 (C=O), 7.65 and 8.70 μ (SO_2); nmr ($DCCl_3$), τ 8.05 (m, 4), 6.71 (s, 3, CH_3), 6.4 and 6.8 (m, 4), 2.2 (m, 4, aromatic protons).

Anal. Calcd for $C_{12}H_{15}NO_3S$: C, 56.89; H, 5.97; N, 5.53. Found: C, 56.59; H, 5.90; N, 5.52.

The Enamine from 1 and Pyrrolidine (18).—Repetition of the experiment used for **19**, except that only 2 equiv of pyrrolidine was employed, produced after evaporation and recrystallization from chloroform–hexane 0.84 g (11%) of the enamine **18**: mp 219–220° dec; uv max (EtOH), 252 mμ (ϵ 12,050), 348 (2900); nmr ($DCCl_3$), τ 8.0 (m, 4), 6.4 (m, 4), 4.86 (s, 1, the 2 proton), 2.4 (m, 4).

Anal. Calcd for $C_{12}H_{13}NO_2S$: C, 61.25; H, 5.57; N, 5.95. Found: C, 61.05; H, 5.50; N, 5.87.

Bis(3-hydroxybenzo[*b*]thien-2-yl 1,1-dioxide)methane (20).—To a suspension of 1.7 g (0.035 mol) of 50% sodium hydride in oil in 35 ml of dry N,N-dimethylformamide was added a solution of 6.4 g (0.035 mol) of **1** in 20 ml of N,N-dimethylformamide. When vigorous gas evolution had subsided, a solution of 3.4 g (0.035 mol) of chloromethyl methyl sulfide in 10 ml of dimethylformamide was added. After stirring 2 hr while a fine white solid precipitated, the solution was poured into 250 ml of water and was acidified with 3 *N* HCl which yielded a soft solid. Trituration with hot ethanol gave, in two crops, 3.0 g (46%) of **20**: mp 254–255°; ir, 5.75 (C=O), 7.60, and 8.71 μ (SO_2); nmr (DMSO-d_6), τ 1.9 (m, 4), 5.1 (t, 2, J = 8 cps, exchanges with D_2O, the 2 proton), 7.15 (d, 2, J = 8 cps, CH_2). A sample of **20** was found to be insoluble in H_2O but dissolved to a yellow solution in warm, dilute NaOH.

Anal. Calcd for $C_{17}H_{12}O_6S_2$: C, 54.24; H, 3.21; S, 17.01. Found: C, 54.11; H, 3.39; S, 16.92.

Registry No.—**1**, 1127-35-1; **2**, 17288-99-2; **3**, 17289-00-8; **4**, 17289-01-9; **5**, 17289-02-0; **6**, 17289-03-1; **7**, 17289-04-2; **8**, 17289-05-3; **9**, 17289-06-4; **11**, 17289-07-5; **12**, 17289-08-6; **13**, 17289-09-7; **14**, 17289-10-0; **15**, 17289-11-1; **16**, 17289-12-2; **17**, 17322-89-3; **18**, 17289-13-3; **19**, 17289-14-4; **20**, 17289-15-5.

Acknowledgment.—The author is grateful to Mr. Nelson Treadway, Jr., for his capable assistance in the synthetic work.

Heterocycles from Hydrazino Alcohols. An Unusual Carbon–Carbon Bond Cleavage

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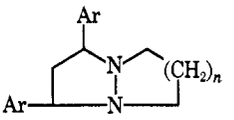
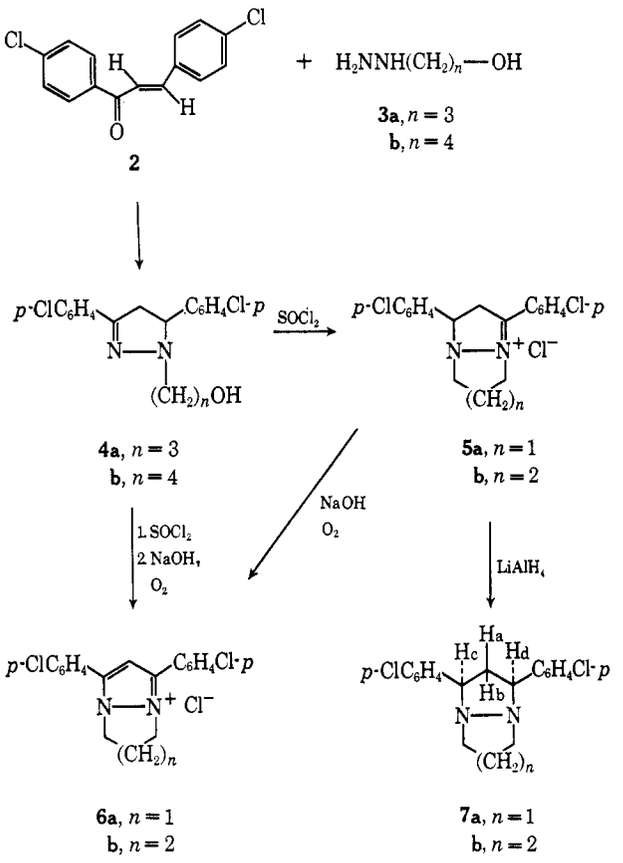
As part of a program directed to the synthesis of bridgehead hydrazine heterocycles with potential medicinal interest, we wished to prepare 1,3-diaryl derivatives of perhydropyrazolo[1,2-*a*]pyrazole (**1a**) and pyridazo[1,2-*a*]pyrazole (**1b**). Since the present synthetic routes[2] for preparing these ring systems do not afford easy access to 1,3-diaryl derivatives, we have investigated the possibility of preparing them from hydrazino alcohols and chalcones. In this note we wish to report the preparation of **1a** from these intermediates and an unusual lithium aluminum hydride cleavage of a carbon–carbon bond during the attempted preparation of **1b**.



1a, $n = 1$
b, $n = 2$

The treatment of 4,4′-dichlorochalcone (**2**) with 3-hydrazinopropanol (**3a**) in refluxing benzene (Scheme I) resulted in the formation of 3,5-bis-*p*-chlorophenyl-

SCHEME I



2 + $H_2NNH(CH_2)_n$—OH (**3a**, $n = 3$; **b**, $n = 4$)

4a, $n = 3$; **b**, $n = 4$ — $SOCl_2$ → **5a**, $n = 1$; **b**, $n = 2$

4a, **b** → 1. $SOCl_2$ 2. NaOH, O_2 → **6a**, $n = 1$; **b**, $n = 2$

4a, **b** → NaOH, O_2 → **6a**, $n = 1$; **b**, $n = 2$

5a, **b** → $LiAlH_4$ → **7a**, $n = 1$; **b**, $n = 2$

1-(3-hydroxypropyl)-4,5-dihydropyrazole (**4a**). Treatment of **4a** with thionyl chloride in chloroform followed by *in vacuo* removal of the excess reagent gave 5,7-bis-(*p*-chlorophenyl)-2,3,6,7-tetrahydro-1H-pyrazolo[1,2-*a*]pyrazol-4-ium chloride (**5a**). If the reaction of **4a** with thionyl chloride was processed with aqueous sodium hydroxide, there was obtained **5a** and 5,7-bis-(*p*-chlorophenyl)-2,3-dihydro-1H-pyrazolo[1,2-*a*]pyrazol-4-ium chloride (**6**). Compound **6** could also be obtained by treating **5a** with aqueous sodium hydroxide in the presence of air. This transformation probably occurs by base-catalyzed air oxidation at C-3 in **5a** followed by loss of water or hydrogen peroxide. Similar oxidative dehydrogenations have been reported with simple pyrazole[3] derivatives. Treatment of **5a**

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