with σ constants for *p*-Cl, -F, and -OMe than for *m*-Cl, -F, and -OMe.

A plausible explanation for the large $\Delta\lambda_{max}$ found for the sulfonyl chlorides is that chlorine and sulfur d orbitals interact to cause a greater charge delocalization.

It was found that when appropriate conditions were chosen both high molecular weight polymer and cyclic tetramer could be prepared from the sulfonyl chloride, III. The details of this work will be reported in a forthcoming publication.

Experimental Section

Sodium 3,5-Dimethyl-4-hydroxybenzenesulfonic Acid.—This salt was prepared from 2,6-xylenol by the method of Karrer and Leiser[8] using sulfuric acid monohydrate at 100–110°.

3,5-Dimethyl-4-hydroxybenzenesulfonyl Chloride (III).—A solution containing 22.4 g of sodium 3,5-dimethyl-4-hydroxybenzenesulfonate in 40 ml of dimethylformamide was cooled to 0° with an ice–salt bath. To this solution 33 g of thionyl chloride was added dropwise over a 10-min period. The cooling bath was removed and the temperature rose to 39°. After a reaction period of 52 min, the reaction mixture was added to 100 g of ice. The white solid formed was isolated and washed several times with ice–water. The crude product (19.8 g) was vacuum dried at room temperature and then dissolved in a minimum amount of toluene. An equal volume of *n*-hexane was added and the solution was cooled to 0°. The solids formed were isolated by filtration and vacuum dried. The white crystals (13.5 g) melted at 134°. The sulfonyl chloride had nmr absorption at δ 7.63 (2 H, singlet, aromatic C–H), 5.67 (1 H, singlet, –OH), and 2.32 (6 H, singlet, benzylic C–H).

Anal. Calcd. for $C_8H_9ClO_3S$: C, 43.5; H, 4.1; Cl, 16.1. Found: C, 43.7; H, 4.1; Cl, 15.8.

Methyl 3,5-Dimethyl-4-hydroxybenzenesulfonate (VI).—3,5-Dimethyl-4-hydroxybenzenesulfonyl chloride (1.25 g) was added slowly to a 0° solution of 5 ml of diethyl ether, 10 ml of methanol and 0.25 ml of pyridine in a vaccine bottle. The bottle was capped and maintained at 5° for 15 hr. The solvent was then removed under vacuum and the solid remaining was washed several times with dilute HCl followed by a water wash. Recrystallization from a petroleum ether (bp 80–100°)–toluene mixture yielded 0.93 g of a crystalline solid melting at 128°. The sulfonate ester had nmr absorption at δ 7.40 (2 H, singlet, aromatic C–H), 5.67 (1 H, singlet, OH), 3.59 (3 H, singlet, methyl C–H), and 2.19 (6 H, singlet, benzylic C–H). The molecular weight was 217 using the Mechrolab technique compared with the calculated value of 216.3.

Anal. Calcd for $C_9H_{12}O_4S$: C, 50.0; H, 5.6; S, 14.8. Found: C, 50.2; H, 5.3; S, 15.1.

3,5-Dimethyl-4-hydroxybenzenesulfonyl Fluoride (V).—A suspension of 6 g of sodium 3,5-dimethyl-4-hydroxybenzenesulfonate in 20 ml of carbon disulfide was cooled to 0° under a nitrogen atmosphere. To this solution 17 g of fluorosulfonic acid was added dropwise over a 10-min period. The cooling bath was then removed and the temperature rose to ~17°. Ice was then added to quench the reaction and the solids formed were isolated and washed several times with ice–water. After vacuum drying the product was recrystallized from petroleum ether–toluene. The crystalline sulfonyl fluoride (3.1 g) melted at 127° and had nmr absorption at δ 7.64 (2 H, singlet, aromatic C–H), 5.61 (1 H, singlet, OH), and 2.33 (6 H, singlet, benzylic C–H).

Anal. Calcd for $C_8H_9FO_3S$: C, 47.1; H, 4.4; S, 15.7. Found: C, 47.2; H, 4.3; S, 15.8.

3,5-Dichloro-4-hydroxybenzenesulfonyl Chloride (II).—Chlorosulfonic acid (230 g) was added slowly to a solution of 50 g of 2,6-dichlorophenol in 100 ml of carbon disulfide which had been cooled to 0°. After the addition period of 1.5 hr, the solution was warmed to 20° where it was maintained for an additional hour. The reaction was quenched by addition to 1000 g of ice. The solid which separated was washed several times with ice–water and then vacuum dried. The crude sulfonyl chloride (75.5 g) was recrystallized several times from *sym*-tetrachloroethane. The white crystalline product melted at 126° and had nmr absorption at δ 8.06 (H singlet, aromatic C–H). The molecular weight as determined by the Mechrolab technique was 260 compared with the calculated value of 261.5.

Anal. Calcd for $C_6H_3Cl_3O_3S$: C, 27.6; H, 1.2; Cl, 40.7. Found: C, 27.8; H, 1.3; Cl, 40.7.

3,5-Dichloro-4-hydroxybenzenesulfonyl Fluroride (IV).—Fluorosulfonic acid (100 g) was added over a 0.5-hr period to a cold solution (0°) of 20 g of 2,6-dichlorophenol in 50 ml of carbon disulfide. The reaction temperature was raised to 20° where it was maintained for 2 hr. The reaction was quenched by the addition of 200 g of ice. The solids formed were isolated and washed several times with ice–water. After vacuum drying, the crude product (29 g) was extracted with 100 ml of chloroform. Upon stripping off the chloroform a solid residue was isolated. Recrystallization from petroleum ether yielded a white crystalline solid melting at 89°. The sulfonyl fluoride had nmr absorption at δ 7.93 (2 H singlet, aromatic C–H) and 6.76 (1 H singlet, –OH).

Anal. Calcd for $C_6H_3Cl_2FO_3S$: C, 29.4; Cl, 28.9. Found: C, 29.5; Cl, 29.1.

3,5-Dibromo-4-hydroxybenzenesulfonyl Chloride (I).—A solution of 5 g of 2,6-dibromophenol in 20 ml of carbon disulfide was added over a 5-min period to a cold (0°) solution of 18 g of chlorosulfonic acid in 30 ml of carbon disulfide. The solution was warmed to 20° over a 0.5-hr period and then quenched by adding 50 g of ice. The solid produced was washed several times with ice–water and vacuum dried. The white solid was recrystallized first from toluene and then from benzene. The crystalline product (3 g) melted at 127° compared with the reported value[2] of 128–129°. The sulfonyl chloride had nmr absorption at δ 8.13 (2 H singlet, aromatic C–H) and 6.10 (1 H singlet, –OH).

Anal. Calcd for $C_6H_3Br_2ClO_3S$: C, 20.6; H, 0.9; S, 9.2. Found: C, 20.9; H, 0.9; S, 9.4.

(8) P. Karrer and P. Leiser, *Helv. Chim. Acta*, **27**, 678 (1944).

2-Phenyl-3-(1-methyl-2-indolyl)-4(1H)-quinolone

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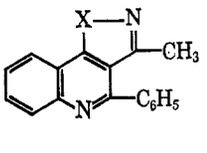
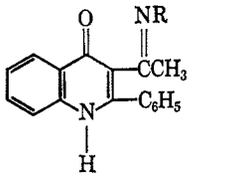
As previously indicated 2-phenyl-3-acetyl-4(1H)-quinolone condenses with phenylhydrazine in aqueous acetic acid solution to yield 3-methyl-1,4-diphenyl-1H-pyrazolo[4,3-*c*]quinoline (Ia).[1] Utilization of hydrazine hydrate in the reaction led to 3-methyl-4-phenyl-1H-pyrazolo[4,3-*c*]quinoline (Ib).

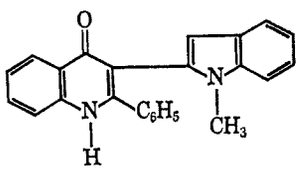
It is likely that base Ia, for example, is derived by cyclization of an intermediate hydrazone IIa produced initially during reaction. Thus, the product from *asym*-



Ia, X = NC_6H_5
b, X = NH
c, X = O



IIa, R = NHC_6H_5
b, R = $N(CH_3)C_6H_5$
c, R = OH



III

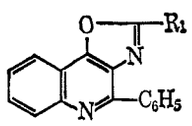
(1) B. Staskun, *J. Org. Chem.*, **26**, 2791 (1961).

methylphenylhydrazine and the ketone under similar conditions proved to be hydrazone IIb.

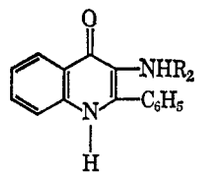
Compound IIb on treatment with either concentrated hydrochloric acid, concentrated sulfuric acid, polyphosphoric acid (PPA), anhydrous zinc chloride, or 60% acetic acid underwent the Fischer indole rearrangement to yield 2-phenyl-3-(1-methyl-2-indolyl)-4(1H)-quinolone (III). The latter was obtained directly from *asym*-methylphenylhydrazine and 2-phenyl-3-acetyl-4(1H)-quinolone by refluxing in 60% acetic acid. Conversion of hydrazone IIb into indole III was effected also in the absence of an acid catalyst by merely heating IIb above its melting point; during this reaction N-methylaniline was liberated.

On heating 2-phenyl-3-acetyl-4(1H)-quinolone oxime (IIc) above its melting point it was converted into 3-methyl-4-phenylisoxazolo[4,5-*c*]quinoline (Ic).[2] Warming the oxime IIc with PPA yielded an isomer of Ic, *viz.*, 2-methyl-4-phenyloxazolo[4,5-*c*]quinoline (IVa);[2] the latter resulted from cyclization during reaction of the Beckmann rearrangement product, 3-acetamido-2-phenyl-4(1H)-quinolone (Vb), produced initially. Supporting this view was the observation that Vb was readily transformed into IVa in the presence of PPA.

Unlike Ic which remained unaffected on short boiling with dilute hydrochloric acid, oxazoloquinoline IVa underwent hydrolysis to 3-amino-2-phenyl-4(1H)-quinolone (Va) characterized by its acetyl and benzoyl derivatives, Vb and Vc, respectively. Amine Va shows potential as a useful starting material in the synthesis



IVa, $R_1 = CH_3$
b, $R_1 = C_6H_5$



Va, $R_2 = H$
b, $R_2 = COCH_3$
c, $R_2 = COC_6H_5$

of more complex heterocyclics. Thus, Va warmed with benzoic acid and PPA furnished 2,4-diphenyloxazolo-[4,5-*c*]quinoline (IVb); the latter compound was obtained also from PPA treatment of benzamido derivative Vc which is evidently intermediate in the synthesis.

Experimental Section[3]

3-Methyl-4-phenyl-1H-pyrazolo[4,3-*c*]quinoline (Ib).—A mixture of 2-phenyl-3-acetyl-4(1H)-quinolone[1] (0.5 g) and excess hydrazine hydrate (50% solution, 5 ml) in 75% (v/v) acetic acid (20 ml) was refluxed for 2 hr. After dilution with water, the pale green solution obtained was cooled and neutralized with concentrated ammonia to precipitate a colorless solid. This was warmed (*ca.* 50°) with 1 *N* sodium hydroxide and the alkali-insoluble material was collected, washed, and dried [0.4 g, mp 120–240° (slow heating)] and heated when it reacted (120–130°) with effervescence and rapidly resolidified, remelting at 240–246°. Crystallization from dilute ethanol furnished Ib as colorless woolly needles, mp 245–246°, sparingly soluble in ether, easily soluble in chloroform and in dilute hydrochloric acid.

Anal. Calcd for $C_{17}H_{13}N_3$: N, 16.22. Found: N, 16.10.

The infrared spectrum showed NH absorption at 3.25 μ and contained no CO (amide or keto) band. That of the intermediate substance, mp 120–240°, was very similar and differed mainly in exhibiting a broader absorption in the NH region and in having a peak at 6.0 μ (m).

2-Phenyl-3-acetyl-4(1H)-quinolone N-Methylphenylhydrazone (IIb).—A mixture of 2-phenyl-3-acetyl-4(1H)-quinolone[1] (2 g) and excess *asym*-methylphenylhydrazine (Fluka technical, 4 ml) in 50% (v/v) acetic acid (80 ml) was refluxed for 30 min. The orange solution was cooled and a small amount of insoluble indole derivative IIIa which sometimes had separated was removed. Addition of water (~200 ml) to the filtrate caused precipitation of a yellow gum which solidified on cooling and scratching. This was dissolved in warm 1 *N* sodium hydroxide (~100 ml) and after removal of a small amount of insoluble impurity the cold filtrate was neutralized with glacial acetic acid to deposit IIb as a yellow solid (1.5–1.8 g, ~55–65%, mp 120–210°) which was shown by its infrared spectrum to contain little if any 3-acetyl starting material. Fractional recrystallization from aqueous methanol (charcoal) afforded tiny yellow crystals (~0.5 g), mp 221–222°, which were sparingly soluble in ether and readily soluble in methanol, in ethanol, and in dilute hydrochloric acid.

Anal. Calcd for $C_{24}H_{21}N_3O$: N, 11.44; mol wt, 367. Found: N, 11.10; mol wt, 367 (mass spectrometer).

The infrared spectrum exhibited a broad NH absorption (3.2–3.6 μ) consisting of a group of several overlapping peaks of varying intensities and displayed also in the spectra of 4(1H)-quinolones generally.[1,4] The "amide I band" was at 6.15 μ (s) while CO (acetyl) absorption was absent.

2-Phenyl-3-(1-methyl-2-indolyl)-4(1H)-quinolone (III). A.—A mixture of 2-phenyl-3-acetyl-4(1H)-quinolone (0.5 g) and excess *asym*-methylphenylhydrazine (1 ml) in 60% (v/v) acetic acid (15 ml) was refluxed for 2 hr during which period crystals of the sparingly soluble III separated from the orange solution. After cooling, these were filtered off, washed with dilute ethanol, and dried to afford 0.2 g (~30%) of III as a pale brown solid, mp 325–355°. Purification was effected by dissolving the crude product in a mixture of ethanol and an equal volume of 1 *N* sodium hydroxide, refluxing the solution (charcoal) for 5–10 min, neutralizing the filtrate with glacial acetic acid, and recrystallizing the precipitated solid from aqueous pyridine to furnish colorless tiny crystals, mp 350–355°.

Anal. Calcd for $C_{24}H_{18}N_2O$: N, 8.0; mol wt, 350. Found: N, 7.95; mol wt, 350 (mass spectrometer).

The indole III was insoluble in ethanol and in dilute alkali and acid, but dissolved in aqueous ethanolic sodium hydroxide, dimethylformamide, and in pyridine. A solution of a few milligrams of III in glacial acetic acid (1–2 ml) rapidly turned red on boiling.

The infrared spectrum showed a broad NH absorption [3.2–3.6 μ (m)], the "amide I band" at 6.2 μ (s), and no CO (keto) absorption.

In the preparative methods B–F below, the product isolated was in each case shown to be III by its melting point and infrared spectrum determined before and after purification.

B.—Hydrazone IIb (1 g) was stirred with PPA (10 g) at 140–150° for 15 min to give an opaque purple mass. Addition of water precipitated III as a buff solid. The mixture was warmed to coagulate the product which, after filtration, washing, and drying, was obtained as a pale brown solid (0.8–0.9 g, ~90%, mp 280–320°).

C.—Freshly fused anhydrous zinc chloride (5 g) and the hydrazone IIb (1 g) were mixed in a morter to a yellow powder which was stirred at 170° for 5 min yielding a viscous red mass. This was cooled somewhat and treated with 1 *N* hydrochloric acid to afford III as an insoluble brown solid (0.8 g, ~85%, mp 280–330°).

D.—A solution of IIb (0.2 g) in concentrated hydrochloric acid (5 ml) was boiled for ~2 min; addition of water after 10 min to the warm turbid mixture precipitated crude III as a brown gum which solidified (~70% yield, mp 325–335°) when cooled and scratched.

Utilization of 1 *N* hydrochloric acid in the reaction furnished little if any III; the crystalline material which separated on dilution of the reaction mixture with water proved to be 2-phenyl-3-acetyl-4(1H)-quinolone while *asym*-methylphenylhydrazine was isolated on working up the acid filtrate.

(2) A. M. Patterson, L. T. Capell, and D. F. Walker, "The Ring Index," 2nd ed., American Chemical Society, Washington, D. C., 1960.

(3) Melting points are uncorrected. Infrared spectra were taken on a Perkin-Elmer Infracord Model 137 spectrophotometer using 1–1.5 mg of sample/300 mg of potassium bromide.

(4) J. R. Price and J. B. Willis, *Australian J. Chem.*, **12**, 589 (1959).

E.—A solution of hydrazone IIb (0.2 g) in concentrated sulfuric acid (4 ml) was heated on the water bath for 0.75 hr; addition of water precipitated crude III as a yellow solid (0.08 g, ~40%).

F.—The hydrazone IIb (0.5 g) contained in a distilling flask was heated in an oil bath at 230–240° for 5–10 min and decomposed with effervescence liberating a colorless liquid (*ca.* 0.1 g) identified by its infrared spectrum as N-methylaniline. The dark red residue in the flask was warmed with 1 *N* hydrochloric acid and afforded insoluble crude III (0.1–0.2 g, ~20–40%).

2-Phenyl-3-acetyl-4(1H)-quinolone Oxime[1] (IIc).—A mixture of 2-phenyl-3-acetyl-4(1H)-quinolone[1] (1 g), excess hydroxylamine hydrochloride (0.8 g), and sodium acetate trihydrate (1.7 g) in 60% (v/v) ethanol (40 ml) was refluxed for 15 min. Colorless crystals of the sparingly soluble oxime IIc soon commenced to separate from solution; these were filtered off from the cold mixture, washed with ethanol, dried (0.9 g, ~85%), and found (infrared spectrum) to be contaminated with some unchanged ketone. Recrystallization from aqueous pyridine furnished colorless tiny crystals, mp 282–284°, virtually free of impurity as evidenced by the very weak absorption peak at 5.9 μ.

3-Methyl-4-phenylisoxazolo[4,5-*c*]quinoline (Ic).—The oxime IIc (0.3 g) in a test tube was heated at *ca.* 290° for 1–2 min when it melted and reacted with effervescence. The brown residue was dissolved in warm 1 *N* hydrochloric acid (charcoal) and the filtrate was made ammoniacal to deposit Ic as a colorless solid (0.15 g). Recrystallization from dilute ethanol furnished colorless woolly needles, mp 151–152°, soluble in ether and insoluble in hot dilute alkali.

Anal. Calcd for $C_{17}H_{12}N_2O$: N, 10.77; mol wt, 260. Found: N, 10.50; mol wt, 260 (mass spectrometer).

The infrared spectrum showed neither NH nor CO (amide or keto) absorption and was consistent with structure Ic. The product (0.1 g) in 2 *N* hydrochloric acid solution (5 ml) was refluxed for 20 min and was recovered unchanged (*cf.* with IVa).

2-Methyl-4-phenyloxazolo[4,5-*c*]quinoline (IVa).—The oxime IIc (0.4 g) was stirred with PPA (8 g) at ±130° for 10 min to give a yellow solution. This was poured into ice–water and the resulting pale green solution was made ammoniacal to precipitate an almost colorless solid. The latter was separated by means of warm 1 *N* sodium hydroxide into alkali-insoluble IVa (0.18 g) and an alkali-soluble "substance A" (0.1 g) which was obtained on neutralizing the alkaline filtrate with glacial acetic acid. Recrystallization of IVa from dilute ethanol furnished colorless woolly needles, mp 147–148°, soluble in ether.

Anal. Calcd for $C_{17}H_{12}N_2O$: N, 10.77; mol wt, 260. Found: N, 10.76; mol wt, 260 (mass spectrometer).

The infrared spectrum showed no NH or CO (amide or keto) absorption and differed from that of Ic.

"Substance A" after two recrystallizations from dilute ethanol was obtained as colorless woolly needles, mp 155–170°, soluble in both alkali and acid, and appeared to be a mixture; its infrared spectrum showed NH and CO (amide) absorptions and differed from that of Vb (see below).

3-Amino-2-phenyl-4(1H)-quinolone (Va).—A solution of IVa (0.2 g) in 2 *N* hydrochloric acid (10 ml) was refluxed for 30 min, cooled, and made ammoniacal to deposit a yellow solid (0.12 g, ~70% yield, mp 200–220°). Recrystallization from dilute ethanol (charcoal) gave yellow needles, mp 243–245° (lit.[5] mp 251°) soluble in dilute alkali and mineral acids.

Anal. Calcd for $C_{15}H_{12}N_2O$: C, 76.27; H, 5.08; N, 11.86. Found: C, 75.92: H, 5.30; N, 11.95.

The solid amine Va fluoresced strongly yellow-green in ultraviolet light and its infrared spectrum showed NH absorption [3.0 μ (w), 3.3–3.6 μ (m)] and the "amide I band" at 6.12 μ (s).

3-Acetamido-2-phenyl-4(1H)-quinolone (Vb).—A solution of Va (0.2 g) in acetic anhydride (2 ml) was boiled for ~1 min and poured into water (~20 ml) with stirring when Vb separated as a colorless solid, This mixture was chilled and the product was filtered off and dissolved in 2 *N* sodium hydroxide; after removal of negligible insoluble impurity, the alkaline filtrate was acidified with glacial acetic acid to deposit Vb (0.15 g), colorless crystals from dilute ethanol, mp 282–284°.

Anal. Calcd for $C_{17}H_{14}N_2O_2$: N, 10.07. Found: N, 10.20.

The acetamido compound Vb (0.1 g) was stirred with PPA (4 g) at ±130° for 10 min. Ice–water was added and the mixture was made alkaline. The alkali-insoluble product (0.05 g) was filtered off and proved to be crude oxazoloquinoline IVa, colorless needles from dilute ethanol identical (mixture melting point and infrared spectrum) with those of the product IVa obtained from the oxime IIc and PPA.

3-Benzamido-2-phenyl-4(H)-quinolone (Vc).—Compound Va (0.15 g) was stirred and warmed (*ca.* 50°) with excess benzoyl chloride (0.3 ml) for 5 min. The mixture was made alkaline, negligible insoluble impurity was removed, and the filtrate was acidified with glacial acetic acid. The precipitated Vc was collected, washed free of benzoic acid with dilute ammonia, and recrystallized from ethanol to give colorless crystals (~0.1 g), mp 290–292°. The same product was obtained also using Reverdin's[6] method.

Anal. Calcd for $C_{22}H_{16}N_2O_2$: N, 8.02. Found: 8.24.

2,4-Diphenyloxazolo[4,5-*c*]quinoline (IVb). A.—The benzamido derivative Vc (0.1 g) was stirred with PPA (1 g) at 140–145° for 10 min. Ice–water was added and the mixture was made alkaline with 2 *N* sodium hydroxide and filtered. The alkali-insoluble IVb (~70% yield) separated from ethanol (charcoal) as colorless crystals, mp 147–148°, soluble in 2 *N* hydrochloric acid.

Anal. Calcd for $C_{22}H_{14}N_2O$: C, 81.99; H, 4.35; N, 8.70. Found: C, 81.70; H, 4.49; N, 8.87.

The infrared spectrum revealed the absence of NH and CO (amide) absorptions.

B.—A mixture of amine Va (0.1 g), excess benzoic acid (0.1 g), and PPA (3 g) was stirred at 130–160° for 20 min. Ice–water was added and the mixture was made alkaline. The alkali-insoluble product (0.1 g) was purified and proved to be identical (mixture melting point and infrared spectrum) with IVb above.

Acknowledgment.—The author is grateful to Dr. S. Eggers of the South African Council for Scientific and Industrial Research for measuring the mass spectra and helpful discussion.

(5) C. M. Atkinson and A. R. Mattocks, *J. Chem. Soc.*, 3722 (1957).

(6) F. Reverdin, *Ber.*, **42**, 1524 (1909).

Halogenated Ketenes. II. Dibromoketene[1]

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Since Staudinger's unsuccessful attempt to prepare dichloroketene in 1913 by the thermal decomposition of the anhydride of dichloromalonic acid and diphenylketene, there has been no definitive work reported on dihalogen ketenes until rather recently.[2] Difluoroketene was reportedly prepared in 1957 by the dehalogenation of chlorodifluoroacetyl bromide.[3] Dichloroketene has been prepared in this laboratory as well as elsewhere quite recently. Stevens and co-workers prepared dichloroketene by the dehydrochlorination of dichloroacetyl chloride and treated the ketene *in situ* with cyclopentadiene.[4] Dichloroketene was synthesized in this laboratory by the dehalogenation of trichloroacetyl bromide with zinc.[5] It was reported that this ketene, unlike difluoroketene, does not distil with ether and is rather susceptible to polymerization like most low molecular weight ketenes.

(1) This work was supported by a National Science Foundation Grant GP-4628 and a Faculty Research Grant, North Texas State University, Denton, Texas.

(2) H. Staudinger, E. Anthes, and H. Schneider, *Ber.* **46**, 3539 (1913).

(3) N. N. Yarovenko, S. P. Motornyi, and L. I. Kirenskaya, *Zh. Obsh. Khim.*, **27**, 2796 (1957).

(4) H. C. Stevens, D. A. Reich, D. R. Brandt, D. R. Fountain, and E. J. Gaughan, *J. Amer. Chem. Soc.*, **87**, 5257 (1965).

(5) Paper I: W. T. Brady, H. G. Liddell, and W. L. Vaughn, *J. Org. Chem.*, **31**, 626 (1966).