oxide (15 g, 0.13 mol) was added to this lithium benzoylnickel carbonylate solution and the mixture was stirred for 5 hr from −60° to room temperature. Benzene (100 ml) was added to this solution and the mixture was stirred for 5 hr at 50–60°. After hydrolysis with 4 *N* hydrochloric acid (50 ml), the reaction mixture was extracted with ether, and this ether solution was separated by extraction with 5% aqueous sodium hydroxide to an acidic part and a neutral part. From the acidic part, benzoic acid (1.1 g, 9.0%) was obtained. The neutral part was distilled under reduced pressure after removal of the solvents to give the following fractions: 1, bp 50–100° (5 mm), 1.6 g; 2, bp 100–180° (5 mm), 9.4 g; 3, bp 180–200° (5 mm), 6.6 g; and 4, bp 200–210° (2 mm), 2.4 g. Most of fraction 1 consisted of the recovered styrene oxide. Recrystallization of fractions 2 and 3 from petroleum ether (bp 30–60°)–benzene gave *trans*-α,β-diphenyl-γ-butyrolactone (**2**) [mp 96–96.5°; white crystals; *m/e* 238, 193, 179, 116; $\nu_{C=O}$ 1780 cm^{-1}; nmr ($CDCl_3$) τ 6.05 (2 H), 5.1–5.8 (2 H), 2.75 (10 H) (*Anal.* Calcd for $C_{16}H_{14}O_2$: C, 80.64; H, 5.92; mol wt, 238. Found: C, 81.00; H, 5.90); 3 g, 19.3%] and benzoin (2.5 g, 23.5%). Benzyl phenyl ketone (**3**, trace) was identified by glpc of fractions 2 and 3. Alkali hydrolysis (KOH–ethylene glycol) of **2** gave α,β-diphenyl-γ-hydroxybutyric acid: mp 147–148°; white crystals; *m/e* 256, 238, 179, 137; $\nu_{C=O}$ 1690 cm^{-1}; ν_{O-H} 3600–3300 cm^{-1}. Reduction of **2** with lithium aluminum hydride gave 2,3-diphenyl-1,4-butanediol: mp 102–102.5°; white crystals; *m/e* 242, 212, 194, 180, 165; ν_{O-H} 3400 cm^{-1}; nmr ($CDCl_3$) τ 7.25 (2 H), 6.60 (2 H), 6.10 (4 H), 3.00 (10 H). As fraction 4 had no peaks at the carbonyl region of the lactone, more purification was not carried out.

B. Reaction of Styrene Oxide with Lithium *p*-Toluoylnickel Carbonylate.—In place of bromobenzene, *p*-bromotoluene (17.1 g, 0.1 mol) was used, and an analogous reaction was carried out under the same conditions as those of reaction A. After the reaction was over, the reaction mixture was extracted with hot benzene and the benzene-soluble part was distilled under reduced pressure to give **2** (3.0 g, 17.0%), benzyl *p*-tolyl ketone **5** (0.5 g, 1.7%), di-*p*-tolyl ketone (4.4 g, 21.0%), and *p,p'*-bitolyl (trace). The identification and the calculation of yields of the products were carried out by glpc analysis. Instead of distillation under reduced pressure, the reaction mixture was separated by alumina column chromatography, and **5**, di-*p*-tolyl ketone, and *p,p'*-bitolyl were also obtained from a benzene or ether eluate. From the methanol eluate, a yellow solid (16.4 g) was obtained, and it showed a peak at 3600–3200 cm^{-1} but no peaks at the carbonyl region. Attempts to purify this yellow solid were unsuccessful but it was transformed to **2** (2 g) by distillation under reduced pressure above 200°.

C. Reaction of Styrene Oxide with Dibenzyliron Tetracarbonylate.—To the tetrahydrofuran solution (150 ml) of disodium iron tetracarbonylate prepared from sodium dispersion (1.4 g, 0.06 g-atom) and triiron dodecacarbonyl (5 g, 0.01 mol), styrene oxide (15.5 g, 0.13 mol) was added at −40 to −30°. After stirring for 1–2 hr at that temperature, benzyl bromide (10.3 g, 0.06 mol) was added and the reaction mixture was stirred for 2 hr from −30° to room temperature and then for 4 hr under refluxing tetrahydrofuran. The solvent was removed by distillation and then the residue was extracted with hot benzene. The benzene-soluble part was distilled under reduced pressure after removal of benzene to give the following fractions: 1, bp 80–140° (0.4 mm), 2.0 g; 2, bp 140–200° (1 mm), 6.0 g. A glpc analysis of these fractions showed that fraction 1 consisted of dibenzyl ketone (1.5 g, 23.8%) and bibenzyl (0.5 g, 9.2%), and fraction 2 consisted of **2** (2.8 g, 19.6%), α-phenylethyl phenylacetate **7** (2.2 g, 14.8%), and β-phenylethyl phenylacetate **8** (trace). An analogous reaction using benzyl iodide (13.1 g, 0.06 mol) gave the same products: dibenzyl ketone (1.4 g, 22.2%), bibenzyl (1.9 g, 35.0%), **2** (2.3 g, 16.1%), **7** (0.2 g, 1.4%), and **8** (trace).

D. Reaction of Styrene Oxide with Di-(*p*-methylbenzyl)iron Tetracarbonyl.—In place of benzyl halide, *p*-methylbenzyl bromide (11.1 g, 0.06 mol) was used and an analogous reaction was carried out under the same conditions as that in part C. Products were α-phenylethyl alcohol (0.8 g, 11.1%), *p,p'*-dimethylbibenzyl (0.8 g, 13.2%), di-(*p*-methylbenzyl) ketone (1.9 g, 27.9%), and **2** (1.0 g, 14.9%), which were identified by glpc analysis.

E. Reaction of Propyrene Oxide with Dibenzyliron Tetracarbonyl.—Propyrene oxide (14.0 g, 0.24 mol) and benzyl iodide (13.1 g, 0.06 mol) were used and an analogous reaction was carried out under the same conditions as in reaction C, and isopropyl phenylacetate (0.18 g, 1.7%), *n*-propyl phenylacetate (0.14 g, 1.3%), dibenzyl ketone (1.5 g, 23.8%), and bibenzyl (2.5 g, 45.6%) were identified by glpc analysis.

F. Reaction of Styrene Oxide with Nickel Carbonyl.—A mixture of styrene oxide (2.4 g, 0.02 mol) and nickel carbonyl (3.5 g, 0.02 mol) in ether (10 ml) and benzene (10 ml) was stirred for 3 hr at 10° and then for 5 hr at 50°. After removal of the solvents and the remaining nickel carbonyl, the residual oil was distilled under reduced pressure to give phenylacetaldehyde (0.2 g, 8.3%) and **2** (trace), which were identified by glpc analysis. Phenylacetaldehyde was also confirmed by its infrared (2750, 1730 cm^{-1}) and nmr (τ 0.55) spectra. No acetophenone was detected by glpc.

G. Reaction of Styrene Oxide with Disodium Iron Tetracarbonylate.—Styrene oxide (1.56 g, 0.013 mol) was added to the tetrahydrofuran solution of disodium iron tetracarbonylate, prepared from triiron dodecacarbonyl (1.7 g, 0.0034 mol) and sodium dispersion (0.7 g, 0.03 g-atom), and the mixture was stirred for 9 hr under refluxing tetrahydrofuran. After removal of the solvent, the residue was filtered and the filtrate was distilled under reduced pressure to give styrene (0.015 g, 1.1%), α-phenylethyl alcohol (0.3 g, 18.9%), and **2** (trace), which were identified by glpc analysis.

H. Reaction of Styrene Oxide with Palladium Dichloride.—Styrene oxide (4.8 g, 0.04 mol) was added to a suspension of palladium dichloride in benzene (50 ml); the mixture was stirred for 27 hr under reflux of benzene. The mixture was filtered and the filtrate was distilled under reduced pressure after removal of benzene to give the following fractions: 1, bp 60–82° (12 mm), 0.62 g; 2, bp 100–134° (1.5 mm), 0.89 g; 3, bp 150–210° (1 mm), 1.0 g. A glpc analysis of fractions 1 and 2 showed that the recovered styrene oxide (0.5 g, 10.4%) and phenylacetaldehyde (0.48 g, 10.0%) were contained in these fractions. Recrystallization of fraction 2 from petroleum ether gave *trans*-2,5-diphenyl-1,4-dioxane: mp 180–180.5° (lit.[9] 174–175°); white plates; *m/e* 240, 149, 120, 104, 91. *Anal.* Calcd for $C_{16}H_{16}O_2$: C, 79.97; H, 6.71; mol wt, 240. Found: C, 80.69; H, 6.93. As fraction 3 had no peaks at the carbonyl region of the lactone, more purification was not carried out.

Registry No.—**1**, 25154-62-5; **2**, 25109-89-1; **4**, 25154-63-6; **6**, 25154-64-7; styrene oxide, 96-09-3; di-(*p*-methylbenzyl)iron tetracarbonyl, 25154-65-8; α,β-diphenyl-γ-hydroxybutyric acid, 25109-09-4; 2,3-diphenyl-1,4-butanediol, 6583-62-6.

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Synthesis and Some Properties of 2,2,6,6-Tetramethyl-1,4,8-trioxaspiro[2.5]octane, an Epoxy Ketene Ketal[1]

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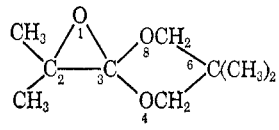
The synthesis of compounds such as the title compound I was undertaken because of their possible use as valuable reagents for the synthesis of a variety of α-substituted acids desired for testing for biological activity. In addition, the novel functionality, epoxide and ortho ester, would make a study of their chemical reactivity of interest.

(1) This work was supported by Grant No. GM 12379 from the National Institutes of Health.

The synthesis of I (20–25% yield) was best accomplished by treatment of 2,2-dimethyl-3-hydroxypropyl α-bromoisobutyrate (II) with sodium hydride in 1,2-dimethoxyethane (glyme). The ester II was prepared by ester interchange between ethyl α-bromoisobutyrate and neopentyl glycol,[2] or, preferably, by reaction of α-bromoisobutyroyl bromide with neopentyl glycol.

$$(CH_3)_2CBrCOOCH_2C(CH_3)_2CH_2OH \xrightarrow{NaH}$$

II



I

In spite of many attempts to synthesize I in which the bases used were sodium hydride, sodium methoxide, sodium amide, metallic sodium, and potassium *t*-butoxide and the solvents were ether, glyme, benzene, dimethyl sulfoxide, hexamethylphosphoramide, and *t*-butyl alcohol, the best yield of I was in the 20–25% region (NaH in glyme). Several attempts to make analogs of I in which corresponding esters of α-bromopropionic and α-bromoacetic acids were used (β-hydroxyethyl esters were also tried) failed. When the chloro analog of II was used, the yield of I was slightly worse than in the case of II.

The epoxy ketene ketal I proved to be very inert to basic reagents as it was recovered in high yield after heating with several strongly basic reagents (see Experimental Section). After treatment with methyllithium in ether for 2 hr at reflux, or with sodium methoxide in methanol at room temperature, I was surprisingly converted in high yield into 2,2-dimethyl-3-hydroxypropyl α-hydroxyisobutyrate (III).

On the other hand, I was readily converted in high yield into III on treatment with aqueous acid. On treatment of a solution of I in benzene (or anisole) at room temperature with 1 equiv of aluminum chloride, a 65% yield of 3-chloro-2,2-dimethylpropyl α-hydroxyisobutyrate (IV) was obtained. When boron trifluoride etherate was used instead of aluminum chloride, a 70% yield of III was obtained. No products resulting from alkylation or acylation of the aromatic nuclei were obtained.

Experimental Section[3]

2,2-Dimethyl-3-hydroxypropyl α-Bromoisobutyrate (II).—To a well-stirred suspension of 76 g of sodium bicarbonate in 416 g of neopentyl glycol dissolved in 500 ml of dry glyme was added, during 10 min, 205 g of α-bromoisobutyryl bromide.[4] After stirring for 15 min following the bromide addition, the solid was removed by filtration, and the filtrate was concentrated under reduced pressure to remove most of the solvent. A solution of the residue in ether was washed several times with water to remove neopentyl glycol. After the usual work-up, distillation in a spinning-band column afforded 155 g (69%) of II as a colorless liquid: bp 76° (0.1 mm); ir bands at 3400 (2.94, broad) and 1720 cm^{-1} (5.81 μ); nmr bands ($CDCl_3$) at τ 6.0 (s, 2 H, OCH_2), 6.70 (s, 2 H, CH_2OH), 7.1 (s, 1 H, OH), 8.1 [s, 6 H, $(CH_3)_2CBr$] and 9.05 [s, 6 H, $(CH_3)_2C$].

Anal. Calcd for $C_9H_{17}BrO_3$: C, 42.7; H, 6.8. Found: C, 42.5; H, 6.4.

The ester II can also be prepared by ester interchange of ethyl α-bromoisobutyrate with neopentyl glycol but the yield (50%) is less, and the product is less pure (90–95%) by glpc, (XE-60 column).

Other Halogenated Hydroxyalkyl Esters.—In a similar way 2,2-dimethyl-3-hydroxypropyl α-bromopropionate was prepared from α-bromopropionyl bromide as a liquid: bp 95–100° (0.6 mm); nmr bands (CCl_4) at τ 5.52 (q, 1 H, $CHBrCH_3$), 5.97 (s, 2 H, OCH_2), 6.63 (s, 2 H, CH_2OH), 6.72 (s, 1 H, OH), 8.17 (d, 3 H, CH_3CHBr), and 9.05 (s, 6 H, $(CH_3)_2C$), in 73% yield.

Anal. Calcd for $C_8H_{15}BrO_3$: C, 40.2; H, 6.3. Found: C, 40.0; H, 6.1.

Similarly, 2,2-dimethyl-3-hydroxypropyl α-bromoacetate was obtained from α-bromoacetyl bromide as a colorless liquid: bp 105–110° (0.6 mm); nmr bands (CCl_4) at τ 6.03 (s, 2 H, $BrCH_2$), 6.15 (s, 2 H, OCH_2), 6.70 (s, 2 H, CH_2OH), 6.85 (s, 1 H, OH), and 9.07 [s, 6 H, $(CH_3)_2C$] in 86% yield.

Anal. Calcd for $C_7H_{13}BrO_3$: C, 37.4; H, 5.8. Found: C, 37.1; H, 5.7.

When α-chloroisobutyroyl chloride was reacted with neopentyl glycol as above, the reaction mixture had to be warmed to 60–70° for 1 hr for reaction to be complete. 2,2-Dimethyl-3-hydroxypropyl α-chloroisobutyrate was isolated as a colorless liquid: bp 82–85° (0.5 mm); nmr bands (CCl_4) at τ 5.83 (s, 2 H, OCH_2), 6.53 (s, 2 H, CH_2OH), 6.75 (s, 1 H, OH), 8.15 [s, 6 H, $(CH_3)_2CCl$], and 9.03 [s, 6 H, $(CH_3)_2C$] in 77% yield.

Anal. Calcd for $C_9H_{17}ClO_3$: C, 51.8; H, 8.2. Found: C, 51.8; H, 8.3.

When α-bromoisobutyryl bromide and α-bromopropionyl bromide were treated with ethylene glycol in glyme as above, the purification of the 2-hydroxyethyl esters was difficult. Analytical samples of 2-hydroxyethyl α-bromopropionate were not obtained, as glpc analyses showed (XE-60 column) that the esters were only about 80% pure. Because no trace of epoxy ketene ketals (similar to I) were obtained by treating these esters with basic reagents (see below), no further attempts at purification were made.

2,2,6,6-Tetramethyl-1,4,8-trioxaspiro[2.5]octane (I).—In a typical experiment the washed solid from 5 g of a 54% suspension of sodium hydride in mineral oil was added all at once to a solution of 29.4 g (0.11 mol) of II in 200 ml of dry glyme. The evolution of hydrogen was rapid. After 15 min the sodium bromide, collected by filtration and washed with solvent, weighed 11.3 g (about theoretical). Removal of solvent from the filtrate on a rotary evaporator afforded a viscous oil which partly solidified. Filtration with the aid of hexane yielded 4.3 g (23%) of I, mp 157–159°, which did not melt higher after several recrystallizations from hexane, from which it separated as colorless elongated prisms which could be sublimed at 80–100° (0.5 mm). No hydroxyl or carbonyl bands appeared in the ir; nmr bands ($CDCl_3$) at τ 6.45 (m, 4 H, OCH_2), singlets at 8.72, 8.87, 9.00, and 9.25 (12 H, 4 CH_3).

Anal. Calcd for $C_9N_{16}O_3$: C, 62.8; H, 9.4. Found: C, 62.4; H, 9.7.

The viscous oil obtained by removal of solvent from the hexane mother liquor decomposed on attempted vacuum distillation (0.1–0.3 mm). The oil before heating showed a broad hydroxyl band and a carbonyl peak in the ir at 1725 cm^{-1}.

Reactions of I.—On treatment of a suspension of 3.0 g of I in 100 ml of water with a few milliliters of concentrated hydrochloric acid, the solid slowly dissolved. After 40 min ether extraction afforded 3.1 g (91%) of 2,2-dimethyl-3-hydroxypropyl α-hydroxyisobutyric acid (III): bp 118–120° (10 mm); ir bands (neat) at 3570 (broad) and 1740 cm^{-1}; nmr ($CDCl_3$) τ 5.93 (s, 2 H, $COCH_2$), 6.57 (s, 2 H, OH), 6.62 (s, 2 H, CH_2OH), 8.55 [s, 6 H, $COHC(CH_3)_2$], and 9.05 [s, 6 H, $(CH_3)_2CCH_2$].

Anal. Calcd for $C_9H_{18}O_4$: C, 56.2; H, 9.5. Found: C, 56.5; H, 9.5.

To a stirred solution of 1.0 g of I in 100 ml of dry ether was added 8 ml of freshly prepared 0.74 *M* methyllithium in ether. After 2 hr at reflux the mixture was cooled and treated with

(2) We thank the Tennessee Eastman Co. for a generous sample of neopentyl glycol.

(3) All temperatures are uncorrected. Microanalyses by nmr spectra were taken on a Varian A-60 instrument. The term "worked up in the usual way" means that an ether or ether–benzene solution of the products was washed with aqueous alkali and saturated salt solutions, dried by filtration through anhydrous magnesium sulfate, and heated under reduced pressure on a rotary evaporator. Glpc analyses were done on 2% XE-60 (nitrile–silicone gum rubber), SE-30 (silicone gum rubber–methyl), and QF-1 (fluorinated silicone gum rubber), on chromosorb W columns. Nmr reported in τ relative to TMS, 10.

(4) C. W. Smith and D. G. Norton, "Organic Syntheses," Coll. Vol. IV, Wiley, New York, N. Y., 1963, p 348.

water, and the ether layer was dried over magnesium sulfate. Removal of the ether left 0.94 g (82%) of oil which, by preparative glpc on an Aerograph autoprep Model A-700 using a 10-ft 20% FFAP (free fatty acid phase) on Chromosorb W at 180°, was shown to consist almost entirely of III plus a small amount of neopentyl glycol. The sample of III was shown to be identical with that described above.

When 1.0 g of I was dissolved in the solution made by adding 0.14 g of sodium to 100 ml of pure methanol and the product isolated after 1 hr, there was obtained 0.97 g (85%) of a mixture similar to that described above, *i.e.*, mostly III and a small amount of neopentyl glycol.

Attempts to react the epoxy ketene ketal I with sodium and potassium *t*-butoxide in refluxing *t*-butyl alcohol, with piperidine or di-*n*-butylamine, with sodium amide (in ammonia or glyme), and with the sodium enolate of cyclohexanone failed. In all cases recovery of I was almost quantitative.

To a stirred solution of 1.0 g of I in 20 ml of dry benzene was added 0.80 g of anhydrous aluminum chloride in one portion. Two layers were formed but soon the mixture was homogeneous. After 1 hr at room temperature 15% hydrochloric acid was added. After a conventional work-up, removal of the benzene left an oily residue which on distillation yielded 0.79 g (65%) of 3-chloro-2,2-dimethylpropyl α-hydroxyisobutyrate (IV): bp 98–100° (10 mm); ir (neat) bands at 3625 and 1740 cm^{-1}; nmr bands ($CDCl_3$) at τ 5.98 (s, 2 H, CH_2Cl), 6.58 (s, 2 H, CH_2O), 6.66 (s, 1 H, OH), 8.58 [s, 6 H, $COHC(CH_3)_2$], and 8.97 [s, 6 H, $(CH_3)_2CCl$]. The analytical sample was obtained by preparative glpc on a 5-ft QF-1 on chromosorb W column at 120°.

Anal. Calcd for $C_9H_{17}ClO_3$: C, 51.8; H, 8.2. Found: C, 51.7; H, 8.3.

When a similar experiment was performed with anisole instead of benzene, the yield of IV (isolated) was 70%. In a similar experiment in anisole with boron fluoride etherate replacing the aluminum chloride, an aqueous work-up followed by extraction with ether afforded III in 70% yield. Treatment of I with anhydrous hydrogen fluoride (followed by an aqueous work-up) for 1 hr, benzoic acid in glyme for 1 day at 25°, and *p*-chlorophenol in glyme for 2 hr at reflux afforded III in 82–89% yields.

Registry No.—I, 25109-69-7; II, 25109-70-0; III, 25109-71-1; IV, 25109-72-2; 2,2-dimethyl-3-hydroxypropyl α-bromopropionate, 25109-73-3; 2,2-dimethyl-3-hydroxypropyl α-bromoacetate, 25109-56-2; 2,2-dimethyl-3-hydroxypropyl α-chloroisobutyrate, 25109-55-1.

Fluoronitroaliphatics. V. Carbonyl Additions of Fluorodinitromethane

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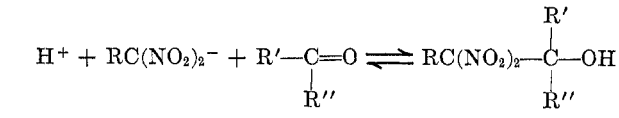
The addition of 1,1-dinitroalkanes and trinitromethane to carbonyl groups (Henry reaction) has been successful primarily with formaldehyde as the acceptor.[1] Far fewer additions to other carbonyl compounds giving isolable and stable product have been reported,[2] and some of these must, in the absence of a rigorous proof of structure, be regarded as unconfirmed in view of the ability of trinitromethane and probably other polynitroalkanes to form isolable complexes with some carbonyl compounds.[3]

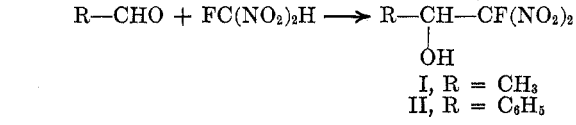
This frequent lack of stability or existence of adducts of polynitroalkanes to higher aldehydes and ketones is due to the reversibility of the Henry reaction and the fact that the equilibrium in these cases does not lie predominantly on the product side. A number of such



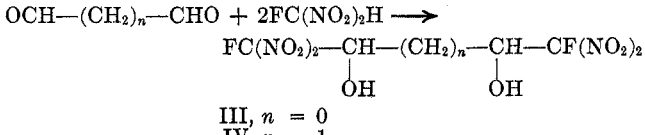
equilibria have been studied by Rondestvedt and coworkers[4] and by Hall[5] who clearly demonstrated the dependence of the Henry equilibrium on the stability of the carbanion, $RC(NO_2)_2^-$ and on the degree of substitution at the carbinol carbon atom.

We wish now to report on a number of carbonyl additions of fluorodinitromethane, a new dinitroalkane whose preparation has been reported only recently.[6] Fluorodinitromethane was found to be the weakest acid[7] among all known 1,1-dinitroalkanes despite the presence of an additional strongly electron-withdrawing substituent. Its carbanion is the least stable known dinitrocarbanion.

Based on these and the above considerations, fluorodinitromethane should therefore form particularly stable carbonyl adducts. Qualitative observations demonstrating the unusual stability of 2-fluoro-2,2-dinitroethanol toward dissociation into formaldehyde and fluorodinitromethane in alkaline medium[6] are in agreement with this expectation. Regarding the addition to higher aldehydes, we find that fluorodinitromethane in buffered aqueous solution (pH 6.5–7.5) readily reacts with acetaldehyde, glyoxal, malondialdehyde, succindialdehyde, glutardialdehyed, and benzaldehyde to give isolable 1-fluoro-1,1-dinitro-2-alkanols I–VI in 50–80% yield.[8] Only one diastereomer of III



I, R = CH_3
II, R = C_6H_5



III, n = 0
IV, n = 1
V, n = 2; a, mp 86–87°
b, mp 90–92 and 102–104° (polymorphs)
VI, n = 3; a, mp 86–88°
b, mp 99–101 and 106.5–108° (polymorphs)

(1) P. Noble, Jr., F. G. Borgardt, and W. L. Reed, *Chem. Rev.*, **64**, 19 (1964); G. A. Shevkhgeimer, N. F. Pyatakov, and S. S. Novikov, *Usp. Khim.*, **28**, 48 (1958).

(2) P. Duden and G. Ponndorf, *Ber.*, **38**, 2031 (1905); N. Maraus and R. Zelinski, *J. Amer. Chem. Soc.*, **72**, 5329 (1950); H. Plaut, U. S. Patent 2,544,103 (1951); R. Schenk, Swedish Patent 135,832 (1952).

(3) For example, a 2:1 complex of trinitromethane and 2,2,4,4-tetramethyl-1,3-cyclobutanedione has been isolated as colorless crystals devoid of OH absorption in the infrared (private communication, L. A. Kaplan, this laboratory), and we have obtained strong indication of complex formation between fluorodinitromethane and acetone.

(4) C. S. Rondestvedt, Jr., M. Stiles, and A. L. Krieger, *Tetrahedron*, **19**, 197 (1963).

(5) T. N. Hall, *ibid., Suppl.*, **No. 1**, 115 (1963); *J. Org. Chem.*, **29**, 3587 (1964); **30**, 3157 (1965).

(6) H. G. Adolph and M. J. Kamlet, *ibid.*, **34**, 45 (1969).

(7) V. I. Slovetskii, L. V. Okhobystina, A. A. Feinzil'berg, A. I. Ivanov, L. J. Birynkova, and S. S. Novikov, *Izv. Akad. Nauk SSSR, Otd. Khim. Nauk*, 2063 (1965); H. G. Adolph and M. J. Kamlet, *J. Amer. Chem. Soc.*, **88**, 4761 (1966).

(8) After completion of this manuscript our attention was directed to the work of Eremenko, *et al.*, who prepared III by the same method in *ca.* 5% yield, and I by the aqueous fluorination of potassium 1,1-dinitropropanol-2: L. T. Eremenko and G. V. Oreshko, *Izv. Akad. Nauk SSSR, Otd. Khim. Nauk*, 380, 1765 (1969).