2,4-Dinitrobenzenesulfonylhydrazine, a Useful Reagent for the Eschenmoser α,β Cleavage of α,β-Epoxy Ketones. Conformational Control of Halolactonization

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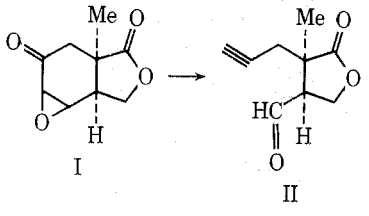
2,4-Dinitrobenzenesulfonylhydrazine has been found to be a useful reagent in the Eschenmoser α,β cleavage of α,β-epoxy ketones especially in instances where the product is an acetylenic aldehyde. Several representative examples are given. In connection with the synthesis of one of the substrates studied in the cleavage reaction, an interesting and useful observation has been made of selective bromolactonization of a Diels–Alder adduct which depends on conformational control.

In connection with the attempted application of the Eschenmoser α,β epoxy ketone cleavage reaction[1-8] for the transformation I → II, it was found that use of *p*-toluenesulfonylhydrazine[1] as reagent was completely ineffective. Although the epoxy ketone was completely consumed, only a very complex mixture could be obtained under a variety of conditions, and little or no aldehyde was detected by infrared and nmr analysis. This result underscores previous indications[2] that this reagent is unsatisfactory for the synthesis of acetylenic aldehydes. Further, it was found that use of *N*-aminoaziridine reagents[2] led to low and irreproducible yields of the desired product and also that no aldehyde could be obtained on a scale larger than a few millimoles. These facts prompted us to study 2,4-dinitrobenzenesulfonylhydrazine as a reagent which might induce fragmentation at relatively low temperatures and under conditions allowing survival of the acetylenic aldehyde II. This expectation was realized in four different cases which are presented herein. In addition, the path of synthesis of the substrate I, which involves a novel selective reaction, is detailed.



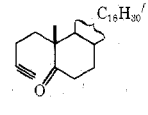
I II

In general, the cleavage reactions were conducted in methylene chloride or tetrahydrofuran at temperatures between 0 and 25° simply by allowing the epoxy ketone and the hydrazine reagent to combine. Pyridine, sodium bicarbonate, or sodium carbonate are effective catalysts. In some instances somewhat higher yields could be obtained by including in the reaction ethyl isocyanate (added to scavenge the sulfinic acid produced in the fragmentation). The overall results are summarized in Table I.

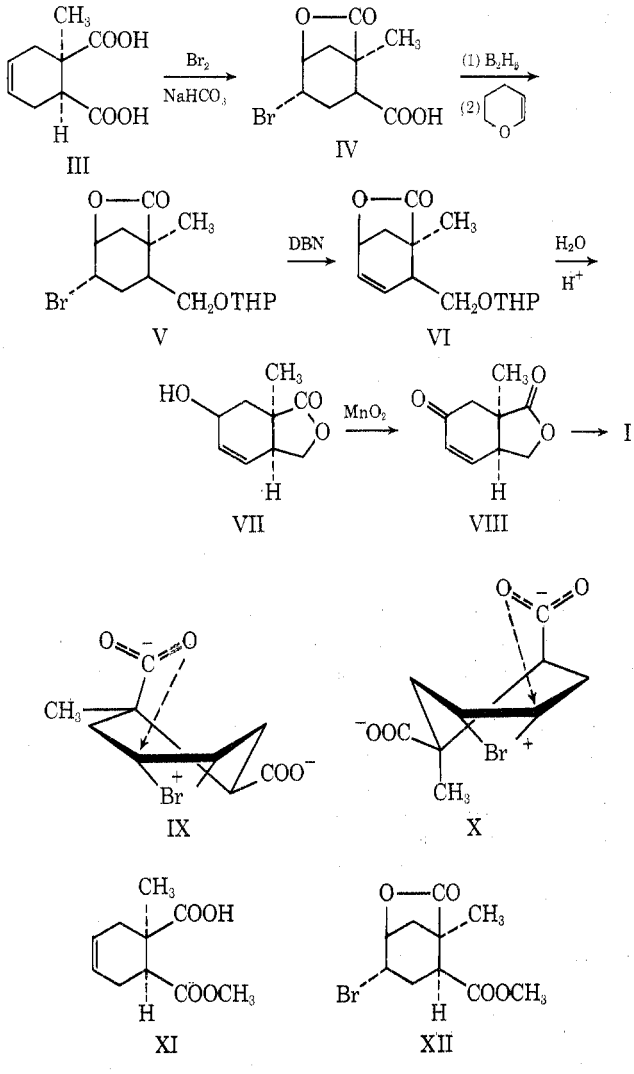
The substrate I (originally of interest as a precursor of 8-methyl prostanoids) was prepared starting with the Diels–Alder adduct from butadiene and citraconic anhydride[9] using the sequence III → IV → V → VI → VII → I. An especially noteworthy step is the bromolactonization in which only one of the two carboxyl groups participates. This selectivity can be rationalized in terms of a more favorable lactonization pathway *via* IX relative to X. This example illustrates what appears to be a new approach to positional control in addition reactions to Diels–Alder adducts of butadiene.

The structure IV was confirmed by partial esterification of III to give *cis*-1-methyl-2-carbomethoxy-4-cyclohexene-1-carboxylic acid (XI),[10] mp 120–121°, followed by bromolactonization to the lactone ester XII, mp 90–91°, which was identical with a sample of the methyl ester obtained by

Table I
Reaction of α,β-Epoxy Ketones with 2,4-Dinitrobenzenesulfonylhydrazine

Substrate	Registry No.	Product	Registry No.	Yield, %
(1) 2,3-Epoxycyclohexan-1-one	6705-49-3	5-Hexynal[a,b]	1871-33-6	62
(2) 2-Methyl-2,3-epoxycyclohexan-1-one	21889-75-8	6-Methyl-5-hexynal[c]	32813-63-1	62
(3) I		II		62
(4) 3-Methyl-5,5-dimethyl-2,3-epoxycyclohexan-1-one	10276-21-8	4,4-Dimethylheptyn-6-one	17520-15-9	91
(5) 4,5-Epoxycholestan-3-one[d,e]	1975-34-4		21489-86-1	95

[a] Isolated as the 2,4-dinitrophenylhydrazone. [b] A yield of 72% was obtained when 1 equiv of ethyl isocyanate was used in the reaction. [c] Isolated as the 2,4-dinitrophenylhydrazone, mp 100–101°. *Anal.* Calcd for $C_{13}H_{14}N_4O_4$: C, 53.79; H, 4.8; N, 19.30. Found: C, 53.53; H, 4.86; N, 19.04. [d] Reaction was carried out at 25° for 30 min, then with Na_2CO_3 at 25° for 2 hr and at 50° for 2 hr. [e] Pl. A. Plattner, H. Heusser, and A. B. Kulkarni, *Helv. Chim. Acta*, **31**, 1822 (1948). [f] Ir max ($CHCl_3$) 3.02 (C≡CH), 4.72 (C≡C), and 5.89 μ (C=O); molecular ion at 384.3387 (calcd for $C_{27}H_{44}O_2$: 384.3392).



esterification of IV with diazomethane by infrared, pmr, mp, and mmp comparison.

Experimental Section

2,4-Dinitrobenzenesulfonylhydrazine. To a well-stirred solution of 95% hydrazine (6.8 g, 200 mmol) in tetrahydrofuran (400 ml) cooled in a Dry Ice–acetone bath was added 2,4-dinitrobenzenesulfonyl chloride (26.6 g, 100 mmol) dissolved in tetrahydrofuran (50 ml). After 30 min the mixture was allowed to warm to ambient temperature. After 15 min the solvent was removed by rotary evaporation and the yellow-colored residue was leached with two 50-ml portions of ice-cold water. The solid was washed with ethanol (50 ml) and then with ether (30 ml). The light yellow solid thus obtained was dissolved in cold, dry tetrahydrofuran (170 ml) without heating, and the solution was filtered, reduced in volume to 30–40 ml by rotary evaporation at ambient temperature, diluted with ethanol (50 ml), and chilled at −10° for 2 hr. The crystalline product, 18.2 g (70%), had mp 120° (lit.[11] mp 110°).

Fragmentation of 2,3-Epoxycyclohexan-1-one. To a solution of 2,4-dinitrophenylsulfonylhydrazine (0.576 g, 2.2 mmol) in tetrahydrofuran (20 ml) cooled to −25° was added the epoxy ketone (0.224 g, 2 mmol). The reaction mixture was kept at −25 to −30° for 30 min and then at −10° for 30 min. The mixture was allowed to warm to 0° and dried at this temperature over anhydrous magnesium sulfate for 20 min and filtered below 0°. After stirring for 1 min a drop of pyridine was added and the stirring was continued. The mixture was taken out of the ice bath, and after 2 min another drop of pyridine was added. This caused an extensive effervescence, and the mixture turned deep orange in color. After 2 min, more pyridine (a total of 0.16 g, 2 mmol) was added, and the stirring was continued for 5 min. The mixture was filtered through Celite 545, and the filtrate was stirred with powdered $CuSO_4 \cdot 5H_2O$ (1 g) for 25 min. It was filtered and the volatile substances from the filtrate were transferred under vacuum (0.03 mm) at ambient temperature into a receiver containing 2,4-dinitrophenylhydrazine (0.792 g, 4 mmol) in tetrahydrofuran (75 ml). The mixture was stored at 25–30° for 48 hr. The solvent was removed by rotary evaporation, and the residue was purified by preparative tlc on silica gel (methylene chloride, R_f 0.8) to give the orange crystalline 2,4-dinitrophenylhydrazone of 5-hexynal (342 mg, 62%): mp 90–91°, nmr ($CDCl_3$) δ 1.7–2.7 (m, 7 H), 7.91 (t, J = 5 Hz, 1 H), 8.2 (d, J = 10 Hz, 1 H), 8.34 (pair of doublets, J_A = 10 Hz, J_B = 2.7 Hz, 1 H), 9.37 (d, J = 2.7 Hz, 1 H), 11.29 (broad s, 1 H); m/e (P) 276.0861.

Anal. Calcd for $C_{12}H_{12}N_4O_4$: C, 52.17; H, 4.38; N, 20.28. Found: C, 51.95; H, 4.28; N, 19.76.

Fragmentation of 3-Methyl-5,5-dimethyl-2,3-epoxycyclohexan-1-one. To a solution of 2,4-dinitrobenzenesulfonylhydrazine (0.577 g, 2.1 mmol) in dry tetrahydrofuran (20 ml) at −25° was added the epoxy ketone (0.308 g, 2 mmol). The mixture was kept at −25° for 30 min and then at 4° for 12 hr, after which it was allowed to warm to 25°. It was treated with pyridine (0.16 g, 2 mmol) which caused an instantaneous effervescence, and the mixture turned deep orange in color. After 20 min the mixture was stirred with powdered $CuSO_4 \cdot 5H_2O$ (0.5 g) for 30 min and filtered through Celite 545. The filter cake was washed with tetrahydrofuran (5 ml). The volatile substances were distilled from the filtrate at ambient temperature at 0.03 mm. Tetrahydrofuran was removed from the distillate by careful distillation, and the residue was transferred under reduced pressure (0.02 mm) into a receiver cooled in liquid nitrogen to give pure 4,4-dimethylheptyn-6-one (250 mg, 91%) as colorless oil. Its ir and nmr spectra were identical with those reported in the literature.[1]

Fragmentation of the Keto Epoxide I. To a solution of 2,4-dinitrobenzenesulfonylhydrazine (0.275 g, 1.05 mmol) in tetrahydrofuran (20 ml) at 0° was added the epoxide I (0.182 g, 1 mmol). The mixture was stored at 0° for 2 hr and then at 20° for 10 min. The solvent was evaporated and the residue was dissolved in methylene chloride (15 ml). It was filtered to remove traces of undissolved material. The filtrate upon chilling at −25° deposited an off-white solid which was collected under suction to give the intermediate 2,4-dinitrobenzenesulfonylhydrazone (0.404 g, 95%): mp 103°. The ir spectrum in chloroform showed only absorption for lactone C=O at 5.6 μ in the carbonyl region. This material was unstable at ambient temperature.

To a solution of the hydrazone (0.404 g) in tetrahydrofuran (20 ml) was added sodium bicarbonate (0.25 g), and the mixtue was stirred for 30 hr at 25–28°, which caused it to turn light orange in color. It was filtered through Celite 545. The clear filtrate was evaporated to give a pale colored gum which according to nmr analysis contained 61–62% of the desired aldehyde II. A sample was chromatographed by preparative tlc on silica gel (ethyl acetate–benzene 2:3, R_f 0.35). The ir spectrum had λ_{max} ($CHCl_3$) 3.0 (C≡CH), 4.71 (C≡CH), 5.6 (C=O, lactone), and 5.78 μ (C=O, aldehyde); nmr ($CDCl_3$) δ 1.56 (s, 3 H), 2.21 (t, J = 2.5 Hz, 1 H), 2.56 (d, J = 2.5 Hz, 2 H), 3.4 (m, 1 H), 4.45 (m, 2 H), 10.0 (d, J = 1 Hz, 1 H); m/e (P) 166.0628, calcd for $C_9H_{10}O_3$ 166.0630.

Bromo Lactone IV. To a well-stirred solution of the diacid III (9.2 g, 50 mmol) in water (100 ml) containing sodium bicarbonate (10.5 g, 125 mmol) was added bromine (8.4 g, 52.5 mmol) over a period of 30 min. After stirring for another 30 min, the reaction mixture was acidified to pH 4–5 which caused a white solid to precipitate. The solid was collected under suction after washing with water to give 10.9 g (80%) of white crystals: mp 206–207°. This material (at least 95% pure by nmr and ir analysis) was used for the next step without further purification. An analytical sample was prepared by recrystallization from ethyl acetate: mp 210°; λ_{max} (Nujol) 5.6 (C=O, lactone), 5.85 (C=O, acid), 7.5, 7.82, 8.15, 8.31, 8.58, 8.92, 9.3 μ.

Anal. Calcd for $C_9H_{11}BrO_4$: C, 41.06; H, 4.14; Br, 30.41. Found: C, 40.95; H, 4.11; Br, 30.25.

Tetrahydropyranyl Ether V. To a well-stirred suspension of bromolactone IV (26.3 g, 0.1 mol) in tetrahydrofuran (350 ml) cooled in ice was added dropwise (30 min) 1.3 M borane in tetrahydrofuran (84 ml, 0.1092 mol). The mixture was a clear solution at this stage. It was maintained at 0° for another 3 hr after which excess borane was destroyed by adding water (70 ml). This was then basified with sodium bicarbonate (10 g). The organic solvent was removed *in vacuo*. The aqueous residual solution was treated with solid sodium chloride and extracted with five 50-ml portions of ethyl acetate. The combined extracts were washed with brine (15 ml) and dried ($MgSO_4$). Evaporation of the solvent under reduced pressure at 10–15° furnished 21.5 g (86%) of a low-melting white solid which deteriorated on keeping at ambient temperature. It

was used for the next step without further purification. An analytical sample was prepared by recrystallization from ethyl acetate–pentane mixture: mp 98–99°; λ_{max} ($CHCl_3$) 2.8 (OH), 5.62 (C=O), 8.62, and 9.18 μ; nmr ($CDCl_3$) δ 1.27 (s, 3H), 1.08–2.9 (m, 6 H), 3.7 (d, J = 4.5 Hz, 2 H), 4.45 (m, 1 H), 4.82 (t, J = 5 Hz, 1 H).

Anal. Calcd for $C_9H_{13}BrO_3$: C, 43.37; H, 5.2; Br, 32.12. Found: C, 43.29; H, 5.18; Br, 32.26.

A stirred ice-cooled solution of the alcohol obtained above (12.45 g, 50 mmol) in tetrahydrofuran (100 ml) containing dihydropyran (6.3 g, 75 mmol) was treated with *p*-toluenesulfonic acid (100 mg) at 0°. After 12 hr the mixture was treated with 10% aqueous sodium bicarbonate (5 ml). The excess dihydropyran and the organic solvent were removed by rotary evaporation. The residue was taken up in methylene chloride (150 ml) and washed with two 10-ml portions of water. After drying ($MgSO_4$), the solvent was evaporated to give 16.6 g (100%) of a colorless thick syrup. This material was used for the following step without any further purification. An analytical sample was prepared by preparative layer chromatography on silica gel (benzene–ethyl acetate, 2:1, R_f 0.8): λ_{max} ($CHCl_3$) 5.62 (C=O), 6.9, 7.22, 8.86, 9.28, 9.78, and 10.18 μ; nmr ($CDCl_3$) δ 1.27 (s, 3 H), 1.64 (broad s, 6 H), 1.9–4.2 (m, 9 H), 4.3–5.2 (m, 3 H).

Anal. Calcd for $C_{14}H_{21}BrO_4$: C, 50.45; H, 6.3; Br, 24.02. Found: C, 50.28; H, 6.15; Br, 23.95.

Lactone VI. A solution of V (9.99 g, 15 mmol) in dry dioxane (250 ml) protected from atmospheric moisture was refluxed for 10 hr after adding diazabicyclo[4.3.0]non-5-ene (DBN) (4.092 g, 16.5 mmol). Shining crystals of DBN hydrobromide were formed. The reaction mixture was allowed to cool to ambient temperature and filtered through Celite 545. The filter cake was washed with dioxane (50 ml). The filtrate upon evaporation *in vacuo* gave a light brown oil. It was taken up in ether (150 ml) and washed with two 10-ml portions of 0.1 *N* hydrochloric acid, then with 10% sodium bicarbonate solution (10 ml), and finally with water (15 ml). The ether solution was dried ($MgSO_4$) and evaporated to give a pale colored oil (6.8 g) which was used as such for the next step. An analytical sample was prepared by preparative layer chromatography on silica gel (benzene–ethyl acetate, 5:1, R_f 0.5) to give a colorless oil. The yield of the purified material was 75%: λ_{max} ($CHCl_3$) 5.62 (C=O), 6.88, 8.8, 8.9, 9.26, and 9.19 μ; nmr ($CDCl_3$) δ 1.44 (s, 3 H), 1.68 (broad s, 6 H), 2.28 (narrow m, 2 H), 2.68–4.21 (m, 6 H), 4.69 (m, 2 H), 6.2 (m, 2 H).

Anal. Calcd for $C_{14}H_{20}O_4$: C, 66.65; H, 7.99. Found: C, 66.48; H, 7.86.

Hydroxy Lactone VII. A suspension of the γ lactone VI (2.52 g, 10 mmol) in a mixture of acetic acid (4 ml), tetrahydrofuran (4 ml), and water (24 ml) was heated at 60° for 12 hr during which time the mixture became a clear solution. It was treated with excess sodium bicarbonate, and the organic solvent was evaporated under reduced pressure. The aqueous solution was treated with excess solid sodium chloride, and the slurry was extracted with five 25-ml portions of ethyl acetate. The combined extracts after drying ($MgSO_4$) were evaporated to give a colorless syrup which on keeping in ether solution at 0° overnight deposited colorless crystals (0.83 g, 50%): mp 49–50°; λ_{max} ($CHCl_3$) 2.76, 2.88 (OH), 5.63 (C=O), 8.2, 8.68, 9.0, 9.6, and 9.9 μ; nmr ($CDCl_3$) δ 1.25 (s, 3 H), 1.48 (d, J = 6 Hz, 2 H), 2.76 (m, 2 H), 3.8–4.3 (m, 3 H), 5.85 (m, 2 H).

Anal. Calcd for $C_9H_{12}O_3$: C, 64.27; H, 7.19. Found: C, 64.16; H, 7.24.

Keto Lactone VIII. To a well-stirred solution of the lactone VII (336 mg, 2 mmol) in methylene chloride (50 ml) was added manganese dioxide (3.5 g), and the mixture was kept at 4–5° for 16 hr. It was treated with methanol (20 ml) and filtered through Celite 545. The filter cake was washed with methanol (10 ml). The filtrate was evaporated under reduced pressure to give a white solid (320 mg, 97%): mp 82–83°. It was used as such for the next step. An analytical sample was prepared by crystallization from ethyl acetate–pentane mixture: mp 84–85°; λ_{max} ($CHCl_3$) 5.61 (C=O, lactone), 5.91 (C=O, conjugated), 6.75, 6.9, 7.2, 7.35, 7.48, 7.7, 8.0, 8.85, 9.11, 9.23, 9.65, 9.86, and 10.15 μ; nmr ($CDCl_3$) δ 1.4 (s, 3 H), 2.43 and 2.92 (pair of doublets, J = 17 Hz, 2 H), 3.17 (m, 1 H), 4.19 (doublet of doublets, J_1 = 10 Hz, J_2 = 4 Hz, 1 H), 4.63 (doublet of doublets, J_1 = 10 Hz, J_2 = 6.5 Hz, 1 H), 6.14 (doublet of doublets, J_1 = 9.5 Hz, J_2 = 2.5 Hz, 1 H), 6.8 (doublet of doublets, J_1 = 9.5 Hz, J_2 = 3.5 Hz, 1 H).

Anal. Calcd for $C_9H_{10}O_3$: C, 65.05; H, 6.07. Found: C, 64.96; H, 6.1.

Epoxy Ketone I. To a well-stirred suspension of the conjugated ketone VIII (249 mg, 1.5 mmol) in methanol (8 ml) cooled to −25° was added 33% hydrogen peroxide (0.5 ml). Aqueous 40% sodium hydroxide (50 μl) was then added, and the mixture was kept at −25 to −30° for 16 hr, during which time a clear solution developed. It was diluted with cold 1% ammonium chloride solution (10 ml), and the methanol was removed under reduced pressure. The aqueous solution was extracted with eight 20-ml portions of methylene chloride. The combined organic extracts were washed with two 6-ml portions of brine, dried ($MgSO_4$), and evaporated to give a colorless crystalline solid. It was recrystallized from methylene chloride–ether–pentane mixture to give colorless crystals (165 mg, 61%): mp 108°; λ_{max} ($CHCl_3$) 5.61 (C=O, lactone), 5.77 (C=O, epoxy ketone), 6.73, 6.88, 7.08, 7.23, 7.18, 8.38, 8.69, 9.06, 9.5, and 10.1 μ; nmr ($CDCl_3$) δ 1.37 (s, 3 H), 2.46 (d, J = 14.5 Hz, 1 H), 2.88 (d, J = 14.5 Hz, 1 H), 3.03 (m, 1 H), 3.29 (d, J = 3.5 Hz, 1 H), 3.59 (d, J = 3.5 Hz, 1 H), 4.3 (doublet of doublets, J_1 = 2.5 Hz, J_2 = 10.5 Hz), 4.6 (doublet of doublets, J_1 = 10.5 Hz, J_2 = 7 Hz).

Anal. Calcd for $C_9H_{10}O_4$: C, 59.34; H, 5.53. Found: C, 59.12; H, 5.51.

***cis*-1-Methyl-2-carbomethoxy-4-cyclohexene-1-carboxylic Acid XI.** This substance was obtained by partial esterification of the diacid III according to the procedure of Nazarov and Kucherov:[10] mp 120–121°.

Bromolactonization of XI. To a well-stirred solution of XI (192 mg, 1 mmol) in water (10 ml) containing sodium bicarbonate (252 mg, 3 mmol) was added dropwise bromine (168 mg, 1.05 mmol) in water (5 ml). After 30 min the reaction mixture was acidified with 1 *N* hydrochloric acid to pH 4–5, and the mixture was extracted with four 20-ml portions of methylene chloride. After drying ($MgSO_4$), the combined extracts upon evaporation of the solvent furnished an oil which immediately crystallized. It was recrystallized from ether–pentane to give 180 mg (66%) of white needle-like crystals: mp 91–92°. The ir and nmr spectra of this material were superimposable with those of the methyl ester XII prepared below; also the mixture mp of the two was undepressed.

Methyl Ester XII. To an ice-cooled solution of the bromolactone IV (132 mg, 0.5 mmol) in 50 ml of ether was added dropwise with shaking 0.2 *N* diazomethane in ether till a faint yellow color persisted. The solvent and excess diazomethane were removed under reduced pressure to give 145 mg of a white solid: mp 90–91° (100%). It was recrystallized from ether–pentane to furnish white crystals: mp 91–92°; λ_{max} ($CHCl_3$) 5.58 (C=O, lactone), 5.75 (C=O, ester), 6.93, 7.45, 7.87, 8.12, 9.2, 9.35, 9.7, 9.96, 10.18, and 10.42 μ; nmr ($CDCl_3$) δ 1.26 (s, 3 H), 3.75 (s, 3 H), 4.52 (m, 1 H), 4.8 (t, J = 5 Hz, 1 H).

Anal. Calcd for $C_{10}H_{13}BrO_4$: C, 43.32; H, 4.33; Br, 28.52. Found: C, 43.11; H, 4.15; Br, 28.35.

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Registry No.—I, 53777-66-5; I 2,4-dinitrobenzenesulfonylhydrazone, 53777-69-8; II, 53777-67-6; III, 35216-43-4; IV, 53777-68-7; V free alcohol, 53777-70-1; V, 53777-71-2; VI, 53777-72-3; VII, 53777-73-4; VIII, 53777-74-5; XI, 14679-29-9; XII, 53798-25-7; 2,4-dinitrobenzenesulfonylhydrazine, 53777-75-6; 2,4-dinitrobenzenesulfonyl chloride, 1656-44-6; hydrazine, 302-01-2; dihydropyran, 25512-65-6.

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