acetone and analyzed by gas chromatography, which showed at least five prominent peaks. Four of them were identified as $ClCH_2CN$, $Cl_2C{=}CHCN$, Cl_3CCCl_3, and $(CH_2CN)_2$. The remaining one was unidentified.

Registry No.—I, 7436-85-3.

Acknowledgments.—The authors are grateful to Dr. S. Tatsuoka, General Manager of the Division, for encouragement throughout this work. Thanks are also due to Mr. M. Kan for the elemental analyses, to Mr. T. Shima for the measurement of mass spectra, and to Mr. K. Shinozaki for the measurement of nmr spectra.

Conversion of Certain Cyclic Phenylhydrazino Derivatives into Phenylazo Compounds with Periodic Acid[1]

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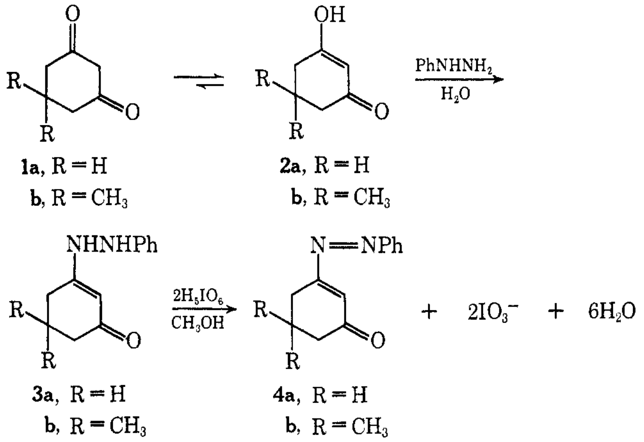
In connection with studies[3] of certain cyclic vinyl phenylazo compounds as possible intermediates in the formation of bis-[4] or trisphenylhydrazones,[5] there was a need to obtain pure samples of 3-oxo-1-phenylazo-1-cyclohexene (**4a**) and 5,5-dimethyl-3-oxo-1-phenylazo-1-cyclohexene (**4b**). One possible way to obtain these compounds is to convert the corresponding enols of 1,3-cyclohexanediones,[6] *e.g.*, the enol of 5,5-dimethyl-1,3-cyclohexanedione (**2b**), into the phenylhydrazino derivative **3b**; the oxidation of the latter would give the needed azo compound **4b**. This type of transformation was studied by Teuber,[7] Eistert,[8] and their students. Although the preparation of **4a** was described,[7] compound **4b** was not reported. In the preparation of **4a**, the starting intermediate, Merling's[9] "phenylhydrazone" **3a** was assigned the correct structure;[7,8] the vinyl structure of **3a** was also confirmed in this laboratory by nmr spectroscopy (see Experimental Section). Transformation of **3a** into **4a** was supposedly performed[7] in high yield by either autooxidation (bubbling of oxygen into an alkaline solution of **3a**) or by treatment with sodium chromate–acetic acid; however, details for isolation of **4a** were not given.[7] In another report,[8] conversion of **3a** into **4a** was performed with N-bromosuccinimide in boiling carbon tetrachloride (46% yield). Tlc [silica gel G, 1:9 ethanol–acetone (v/v), 3% periodic acid in methanol as the spray] revealed a trace of impurity in **4a** (mp 85–86°) prepared with N-bromosuccinimide as the oxidant;[8] this impurity can be formed by the bromination of a benzene ring or a labile vinyl bond of **4a**. However, this difficulty was overcome when periodic acid was used as the oxidant (or proton-abstracting agent); this reagent has proved to be a unique oxidant for other systems.[2]



It has been found that treatment of a solution of **3a** or **3b** (1 mol) in methanol or glacial acetic acid with an aqueous solution of periodic acid (2 mol) at room temperature produces the corresponding azo compounds **4a** and **4b** in 90% yield. After recrystallization from aqueous acetic acid, 3-oxo-1-phenylazo-1-cyclohexene (**4a**) melted at 92–94°; it was homogeneous by tlc and the mass spectrum did not show impurity. For comparison, compound **3a** (1 mol) in glacial acetic acid was oxidized with lead tetraacetate (2.2 mol) to give the azo product **4a** in 55% yield; oxidation with sodium periodate gave the azo product in 70% yield. The moderate solubility of sodium periodate in water lessens its value as a preparatory reagent. (The product from the above two oxidations was isolated by extraction into chloroform.)

It is believed that clear-cut conversion of vinyl phenylhydrazino compounds **3a** and **3b** into the corresponding azo compounds **4a** and **4b** with periodic acid may indicate the general character of this reagent for deprotonation of this type of compound.

The action of periodic acid, a two-electron oxidant, on a phenylhydrazino group (conversion of **3a** and **3b** into **4a** and **4b**) can be envisaged as a simultaneous attack of an electrophilic and nucleophilic species (present in aqueous acetic acid–periodic acid) on vinyl and phenyl NH groups, respectively. This mechanistic approach is also in agreement with the earlier suggestion[2b,d] that the shift in equilibrium between electrophilic (nonionized) and ionized species of periodic acid solution is also dependent on the nature of the species to be oxidized.

The equation for calculation of the enol content of an acyclic β diketone[6d,e] was found to be useful in determination of the enehydrazine content in compounds **3a** and **3b**. The per cent of enehydrazine in a compound

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[1] Part V. For parts I–IV, see ref 2a–d, respectively.

[2] (a) A. J. Fatiadi, *J. Org. Chem.*, **32**, 2903 (1967); (b) *Chem. Commun.*, 1087 (1967); (c) *ibid.*, 456 (1968); (d) *J. Res. Nat. Bur. Stand.*, *A*, **72**, 341 (1968).

[3] A. J. Fatiadi, *Carbohyd. Res.*, **7**, 89 (1968).

[4] (a) L. Cagliotti, G. Rossi, and F. Rossi, *J. Amer. Chem. Soc.*, **88**, 3865 (1966); (b) H. Hassner and Catsoulacos, *Tetrahedron Lett.*, 489 (1967); (c) H. Simon, G. Heubuch, and H. Wacker, *Chem. Ber.*, **100**, 3106 (1967); (d) J. Buckingham. *Quart. Rev.* (London), **23**, 37 (1969).

[5] A. J. Fatiadi, to be published.

[6] The ring geometry of 1,3-cyclohexanedione (**1a**) and its 5,5-dimethyl derivative (**1b**) prohibits intramolecular rotation and intramolecular hydrogen bonding and so the enol of the dione becomes stabilized (*trans*-fixed enolization). Consequently, these 1,3-diones are almost completely enolized (95 and 100%, respectively); see (a) E. S. Gould, "Mechanism and Structure in Organic Chemistry," Henry Holt and Co., New York, N. Y., 1959, pp 376–379; (b) B. Eistert and E. Merkel, *Chem. Ber.*, **86**, 896 (1953); (c) B. Eistert, E. Merkel, and W. Reiss, *ibid.*, **87**, 1513 (1954); compare with an enol form of an acyclic β diketone; (d) J. L. Burdett and M. T. Rogers, *J. Amer. Chem. Soc.*, **86**, 2105 (1964); (e) G. K. Schweitzer and E. W. Benson, *J. Chem. Eng. Data*, **13**, 452 (1968).

[7] (a) H. J. Teuber, D. Cornelins, and E. Worbs, *Z. Naturforsch.*, **21b**, 88 (1966); (b) H. J. Teuber and R. Braun, *Chem. Ber.*, **100**, 1353 (1967).

[8] B. Eistert, G. Kilpper, and J. Göring, *Chem. Ber.*, **102**, 1379 (1969).

[9] G. Merling, *Justus Liebigs Ann. Chem.*, **378**, 39 (1894).

was calculated from the relationship $200E/(2E + K)$, where E is the integrated intensity of a vinyl imino proton with a chemical shift at τ 2.17 (*e.g.*, for **3a**) and K is the integrated intensity of phenyl NH protons (a chemical shift at τ 1.29 for **3a**); other parameters, *e.g.*, that of a vinyl proton at τ 4.02 for **3a**, and one-quarter of the integrated intensity of CH_2 ring protons, a multiplet at τ 7.50–8.00 for **3a**, can also be used. Calculation revealed that compounds **3a** and **3b** are completely in the enehydrazine form, namely, 97 and 99%, respectively (methyl sulfoxide solvent).

Inspection of ultraviolet spectra of the vinyl azo compounds **4a** and **4b** revealed the presence of a triplet at 228, 234, and 239 nm which, as shown earlier,[3] is characteristic of a RCH=CHN=NPh chromophore. The visible spectra of compounds **4a** and **4b** (maxima at 470–472 nm) are of low intensity (ϵ *ca.* 300–350), indicative of a *trans*-azo structure (see ref 3 and references cited therein).

Experimental Section

Melting points were determined in a silicone oil bath apparatus and are corrected. Nmr spectra were obtained at 60 MHz on a Varian A-60 spectrometer, with tetramethylsilane (τ 10,000) as the internal standard. Ir spectra were recorded with a Perkin-Elmer grating Model 257 spectrophotometer; uv spectra were recorded with a Beckman DK-2 or a Cary 14 spectrophotometer. The mass spectra were determined with an LKB-9000 spectrometer[10] at an ionizing voltage of 70 eV and a probe temperature of 40–60°. Analyses were made by William Schmidt of the Microchemical Analysis Section.

5,5-Dimethyl-3-oxo-1-phenylhydrazino-1-cyclohexene (**3b**).—The procedure used was a modification of that of Merling[9] for the preparation of **3a**; direct use of phenylhydrazine[8] gave a somewhat colored product. A mixture of 2.8 g (20 mmol) of 5,5-dimethyl-1,3-cyclohexanedione (dimedone, commercial preparation) and phenylhydrazine hydrochloride (3 g, 20 mmol, 5% excess) in water (50 ml) was stirred at room temperature for 10 min; all of the phenylhydrazine hydrochloride had then dissolved. The suspension was cooled in an ice bath, solid sodium acetate trihydrate (3 g, 20 mmol, 10% excess) was added, and the mixture was stirred with cooling for 30 min. White to cream-white crystalline **3b** was filtered off, washed with cold water (15 ml), and dried in a vacuum desiccator over phosphorus pentaoxide, yield 3.2 g. Further cooling and careful concentration of the filtrate gave a second crop of **3b**, yield 0.4 g, total yield 3.6 g (78.3%), mp 163–164°. Recrystallization of the crude product from chloroform with acid-washed charcoal and drying at 78° (0.1 Torr) gave snow-white prisms: mp 172–173° (lit.[8] mp 163–164°); uv max (CH_3OH) 224 nm (sh, ϵ *ca.* 7100), 236 (*ca.* 8400), and 290 (*ca.* 24,800); ir $\nu_{\max}^{\mathrm{KBr}}$ 3240 (s, NH), 1720 (vw, CO), 1610 (s, phenyl ring), 1552 (s, NH bending),[11] and 1160 cm^{-1} (s, PhN);[11] nmr (Me_2SO-d_6) τ 1.30 (s, 1, nonchelated, phenyl NH, easily exchangeable with D_2O at room temperature), 2.12 (s, 1, nonchelated, vinyl NH, more difficultly exchangeable with D_2O at room temperature), 3.20 (m, 5, phenyl ring), 4.92 (s, 1, C=CH, vinyl proton), 7.20 (s, 2, CH_2 of cyclohexane ring), 7.98 (s, 2, CH_2 of cyclohexane ring), and 8.99 (s, 6, CH_3); (CD_3COOD) τ 0.32 (s, 1, phenyl NH), other imino proton peaks absent because of apparent protonation. However, addition of 2 drops of pyridine-d_5 to the probe produced an expected NH resonance at τ 1.50, thus proving the basic character of the vinyl imino group.

Tlc of colorless **3b** [silica gel G, 1:9 ethanol–acetone (v/v), 3% periodic acid in methanol as the spray] gave a red spot due to **4b**; the R_F value for **3b** was higher than for **3a**.

Anal. Calcd for $C_{14}H_{18}N_2O$: C, 73.01; H, 7.87; N, 12.16. Found: C, 73.00; H, 7.69; N, 12.05.

3-Oxo-1-phenylhydrazino-1-cyclohexene (**3a**).—The starting material, 1,3-cyclohexanedione (dihydroresorcinol), was prepared by hydrogenation of an alkaline solution of 1,3-benzenediol (resorcinol) with Raney nickel W7 catalyst in the usual way (an autoclave[12] at 90–100° and 1000 lb/in^2 of hydrogen). The Merling[9] "phenylhydrazone" **3a** was prepared in 80% yield according to the procedure described for **3b**; direct treatment of the 1,3-dione with phenylhydrazine[8] gave **3a** relatively free from colored impurity. The crude **3a**, mp 169–171°, was recrystallized from 1:3 ethanol–acetone (v/v) with acid-washed charcoal and cooled. The product was washed with cold acetone and dried at 78° (0.1 Torr) to give snow-white prisms: mp 177–179° (lit.[8] mp 177–178°, 176–178°); uv max (MeOH) 224 (sh, ϵ *ca.* 6800), 237 (*ca.* 8600), and 288 (*ca.* 25,050); $\nu_{\max}^{\mathrm{KBr}}$ 3220 (s, NH), 1670 (vw, C=O), 1610 (s, phenyl ring), 1545 (s, NH bending),[11] and 1140 (m, PhN);[11] nmr (Me_2SO-d_6) τ 1.29 (s, 1, nonchelated, phenyl NH, easily exchangeable with D_2O at room temperature), 2.17 (s, 1, nonchelated, vinyl NH, more difficultly exchangeable with D_2O at room temperature), 3.20 (m, 5, phenyl ring), 4.92 (s, 1, C=CH vinyl proton), and 7.50–8.00 (m, 6, CH_2 of cyclohexane ring).

Anal. Calcd for $C_{12}H_{14}N_2O$: C, 71.25; H, 6.97; N, 18.85. Found: C, 71.34; H, 7.10; N, 13.90.

5,5-Dimethyl-3-oxo-1-phenylazo-1-cyclohexene (**4b**).—A precooled solution of **3b** (2.3 g, 10 mmol) in methanol (40 ml) was treated with a solution of periodic acid (4.7 g, 20 mmol, 5% excess) in water (6 ml). The deep-red solution was stirred for 30 min in an ice bath and gradually diluted with ice-cold water (50 ml); pink needles of **4b** crystallized out spontaneously. The product was separated after additional stirring (10 min), washed with ice-cold 1:1 water–methanol (v/v, 20 ml), and dried in a vacuum desiccator, yield 1.9 g. Dilution of the filtrate with water (80 ml) and cooling gave a second crop, yield 0.25 g, total yield 2.15 g (*ca.* 94%), mp 60–62°. The product was recrystallized from aqueous methanol containing 20% of acetic acid and dried at 25° (0.05 Torr): mp 66–67°; uv max (cyclohexane) 228 nm (ϵ *ca.* 9500), 234 (*ca.* 10,700), 238 (*ca.* 9800), 321 (*ca.* 24,800), and 471 (*ca.* 320); $\nu_{\max}^{\mathrm{KBr}}$ 1690 (w, C=O), 1640 (s, C=C, vinyl), 1612 (w, phenyl), 1580 (m, N=N),[11] 1410 (m, N=N),[11] and 1150 cm^{-1} (m, PhN);[11] nmr ($CDCl_3$) τ 2.08, 2.40 (m, 5, phenyl ring), vinyl proton H-2 as a triplet centered at τ 3.12 owing to a long-range coupling with H-4 ($J_{2,4} \cong 2$ Hz) and H-6 ($J_{2,6} \cong 1.5$ Hz), cyclohexane ring proton H-4 as a doublet centered at τ 7.36 owing to long-range coupling with the H-5 methyl proton ($J_{4,5} \cong 1.5$ Hz), another ring proton as a singlet at τ 7.55, and methyl protons as a singlet at 8.86; mass spectrum *m/e* 228 (parent ion), 105 (base peak, $PhNN^+$), and 77 ($C_6H_5^+$).

Anal. Calcd for $C_{14}H_{16}N_2O$: C, 73.11; H, 7.7; N, 12.10. Found: C, 69.95; H, 7.60; N, 12.21.

3-Oxo-1-phenylazo-1-cyclohexene (**4a**).—A precooled solution of **3a** (12 g, 60 mmol) in methanol (200 ml) was treated with a solution of periodic acid (30 g, 120 mmol, 5% excess) in 20 ml of water as described for compound **4b**. The total yield of lustrous, pink-red crystals of **4a** was 10.8 g (90%), mp 89–91°. The product was purified by recrystallization from aqueous methanol containing some acetic acid, and a sample was dried at 40° (0.1 Torr): mp 92–94° (lit.[8] mp 85–86°); uv max (cyclohexane) 228 nm (ϵ *ca.* 9050), 234 (*ca.* 10,150), 238 (*ca.* 9400), 320 (*ca.* 24,150), and 470 (*ca.* 340); $\lambda_{\max}^{\mathrm{MeOH}}$ 234 nm (ϵ *ca.* 10,700), 239 (sh, *ca.* 10,400), 325 (*ca.* 25,300), and 472 (*ca.* 370); $\nu_{\max}^{\mathrm{KBr}}$ 1650 (s, C=C, vinyl), 1570 (w, phenyl), 1560 (w, N=N),[11] 1430 (m, N=N),[11] 1158 (m), and 1140 cm^{-1} (m, PhN);[11] nmr ($CDCl_3$) multiplet centered at τ 1.18 and 2.12 (phenyl ring), vinyl proton H-2 as a triplet centered at τ 2.88 owing to a long-range coupling with H-4 ($J_{2,4} \cong 2$ Hz) and H-6 ($J_{2,6} \cong 1.5$ Hz), cyclohexane ring protons as multiplets centered at τ 7.30 and 7.72; mass spectrum *m/e* 200 (parent ion), 105 (base peak, $PhNN^+$), and 77 ($C_6H_5^+$).

Anal. Calcd for $C_{12}H_{12}N_2O$: C, 71.99; H, 6.04; N, 13.99. Found: C, 71.85; H, 5.95; N, 14.16.

Treatment of a solution of **4a** (1 mol) in glacial acetic acid with bromine (2 mol) at room temperature (exothermic reaction),

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(10) Certain commercial instruments are mentioned in this paper; this does not imply recommendation or endorsement by the National Bureau of Standards.

(11) A. J. Fatiadi, *J. Res. Nat. Bur. Stand.*, *A*, **71**, 277 (1967).

(12) 1,3-Cyclohexanedione (Aldrich, stabilized with sodium chloride) can also be used. The hydrogenation mixture (from 100 g of resorcinol in 400 ml of water containing 45 g of sodium hydroxide and 12 g of catalyst, followed by acidification with hydrochloric acid and cooling) gave about 60 g of 1,3-cyclohexanedione; the filtrate, following neutralization with sodium acetate, cooling, and treatment with an excess of phenylhydrazine (30 g), gave an unknown compound (32 g), mp 159–160°; the structure of this compound is under investigation.

followed by dilution with water after the reaction mixture had been kept for 30 min at room temperature, gave pinkish prisms, mp 129–130° from methanol. *Anal.* Found: C, 39.4; H, 2.2; N, 7.2. The structure of this bromide derivative of **4a** is under investigation; however, it is presumed that a vinyl bond and a phenyl ring have been brominated, the latter in the *para* position.

Registry No.—**4a,** 21232-58-6; **4b,** 22538-42-7.

Acknowledgments.—The author thanks Dr. Henry M. Fales, National Institutes of Health, for recording the mass spectra, Richard Thompson for the nmr spectra, and Norman Adams for the esr measurements.

γ-Induced Addition of Trichlorosilane to Vinyl Acetate

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Addition of trichlorosilane to a number of olefinic double bonds has been reviewed.[1] However, very little is known about the addition to an olefinic double bond adjacent to an acetoxy group. Only a short description[2] has been given that trichlorosilane added to vinyl acetate by benzoyl peroxide, giving the 1:1 adduct, 2-trichlorosilylethyl acetate, in 15% yield, based on vinyl acetate. The present note will report γ-induced addition of the same system, conversion of the trichlorosilyl group of the 1:1 adduct to the triethoxysilyl group, and subsequent pyrolytic loss of acetic acid from the 1:1 adduct and its triethoxylated compound. The above three steps may lead to synthesis of vinyltrichloro- and vinyltriethoxysilane starting from trichlorosilane and vinyl acetate, although other processes have been utilized for synthesis of vinyltrichlorosilane[3–6] and vinyltrialkoxysilanes.[7]

A mixture of trichlorosilane and vinyl acetate in a fused tube was irradiated with a ^{60}Co source. Distillation of the irradiated mixture yielded three fractions, two boiling at 42 and 65° (6 mm), respectively, and residue [>70° (6 mm)]. The lower boiling fraction was identified as 2-trichlorosilylethyl ethyl ether, $Cl_3SiCH_2CH_2OCH_2CH_3$, by the coincidence of physical constants with those reported,[8] and confirmed further by conventional methods, especially by nmr. The higher boiling one was identified as the 1:1 adduct, 2-trichlorosilylethyl acetate. The residue gave, after three distillations, a fraction boiling at a range of 113–115° (3 mm), which was identified as the 1:2 adduct, 4-trichlorosilyl-3-acetoxybutyl acetate, $Cl_3SiCH_2CH(OAc)CH_2CH_2OAc$. To exemplify our assumption that the ether arose from reduction of the 1:1 adduct under the present conditions, a mixture of the 1:1 adduct and trichlorosilane (the molar ratio of which 1:6) was irradiated under a total dose of 10 MR at a dose rate of 0.6 MR/hr. Distillation of the irradiation mixture gave 2-trichlorosilylethyl ethyl ether in 67% yield, based on the 1:1 adduct. Equation 1 represents a

$$Cl_3SiCH_2CH_2OCOCH_3 \overset{HSiCl_3}{\rightsquigarrow} Cl_3SiCH_2CH_2OCH_2CH_3 \quad (1)$$

quite novel reaction indicating reduction of a carbonyl to methylene. If it is true, we can expect to reduce a carboxylic ester in general to an ether by this method. In fact, alkyl aliphatic carboxylates, methyl, ethyl, and *n*-propyl esters of formic, acetic, and propionic acids so far studied, could be reduced to the corresponding dialkyl ethers. Trichlorosilane, in turn, changed to hexachlorodisiloxane. RCOOR′, of which either R or R′ is aryl or aralkyl, could not be reduced, however. The scope and sequence of this reduction will be reported elsewhere.[9]

After identification of the fractions obtained from the irradiated mixture, the effects of irradiation dose rate, total dose, and molar ratio of the silane to vinyl acetate on the yield of the 1:1 adduct were studied, and are shown in Tables I, II, and III, respectively. Table III shows that when the molar ratio is lower, the amount of the residue is greater. This result can be explained by a radical chain telomerization mechanism, that is, competition of the chain transfer step by the silane with the chain propagation step to vinyl acetate. The proportion of either step depends on the molar ratio of the mixture. Table III also indicates that, as the molar ratio increases above 8, the amount of ether increases at the expense of the 1:1 adduct. This result can be ascribed to eq 1. Other investigators[10] reported that γ-induced addition of trichlorosilane to an alkene like 1-octene occurred almost quantitatively. The better yield for the simple olefin in contrast to the yield for vinyl acetate can be explained by the facts that the simple alkene does not form 1:2 or higher adducts and that the 1:1 adduct produced does not change during the reaction. The same investigators[10] also reported that trichlorosilane added to allyl acetate by γ irradiation, giving 1:1 and 1:2 adducts in 22 and 71% yield, respectively, based on alkene. In this case, the molar ratio of silane to alkene was 3. Therefore, reduction of the adducts by the silane seems not to predominate, as the total yield indicates. Our results given in Table III also indicate that no reduction occurs with this molar ratio.

Since the trichlorosilyl group is, as is well known,[10] sensitive even to atmospheric moisture, it seems necessary to convert this group to a stable one for further treatments. Although alkylation by the Gri-

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[1] F. W. Stacey and J. F. Harris, Jr., *Org. Reactions*, **13**, 209 (1963).

[2] M. F. Shostakovskiĭ and L. I. Shomonina, *Izv. Akad. Nauk SSSR, Otd. Khim. Nauk*, 64 (1958).

[3] From chloroalkyltrichlorosilane by dehydrochlorination: A. D. Petrov, V. A. Ponomarenko, B. A. Sodolov, and Yu. P. Egorov, *ibid.*, 310 (1957); R. Müller and K. Schnurrbusch, *Chem. Ber.*, **91**, 1805 (1958).

[4] From vinyl chloride by Si-containing catalysts: D. T. Hurd, *J. Amer. Chem. Soc.*, **67**, 1813 (1945); A. L. Klebanskiĭ and V. S. Fikhtengol'ts, *Zh. Obshch. Khim.*, **27**, 2648 (1957); M. F. Shostakovskiĭ, E. M. Savitskiĭ, D. A. Kochkin, and L. V. Musatova, *Izv. Akad. Nauk SSSR, Otd. Khim. Nauk*, 1493 (1957).

[5] From acetylene and trichlorosilane: G. H. Wagner and C. O. Strother, British Patent 670,617 (1952); M. F. Shostakovskiĭ and D. A. Kochkin, *Izv. Akad. Nauk SSSR, Otd. Khim. Nauk*, 1150 (1956).

[6] From vinyl chloride and trichlorosilane: British Patent 752,700 (1956).

[7] Alkoxylation of vinyltrichlorosilane: R. Nagel, C. Tamborski, and H. W. Post, *J. Org. Chem.*, **16**, 1768 (1951).

[8] R. Clalas, N. Duffaut, and J. Valade, *Bull. Soc. Chim. Fr.*, 790 (1955).

[9] J. Tsurugi, R. Nakao, and T. Fukumoto, *J. Amer. Chem. Soc.*, **91**, 4587 (1969).

[10] A. M. El-Abbady and L. C. Anderson, *ibid.*, **80**, 1737 (1958).