Conversion of aromatic aldehydes into 1-aryl-2,2-dichloroethenes

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A new general one-pot preparative method for the synthesis of 1-aryl(hetaryl)-2,2-dichloroethenes from aldehydes was developed. The method involves successive conversions of the latter into hydrazones followed by treatment with carbon tetrachloride in the presence of copper(I) chloride.

Key words: catalysis, copper salts, dichlorostyrenes, hydrazones, carbon tetrachloride.

1,1-Dihalolalkenes are widely used as synthetic precursors of alkynes and chloroalkynes.[1—4] The classical approaches to the synthesis of 1,1-dihaloalkenes involve the Wittig reaction and its modifications,[5—8] reduction of α-trichloromethyl alcohols with metals (zinc, *etc.*),[9—11] reductive coupling of polyhalogenohydrocarbons with aromatic aldehydes in the presence of the lead—aluminum system,[12] and [2+2]-cycloaddition of dichloroketenes to aldehydes followed by thermal decarboxylation of the resulting β-lactones.[13] The drawbacks of these procedures are drastic conditions, the necessity of using expensive reagents, and the employment of equimolar amounts of phosphorus reagents or metals as reductants.

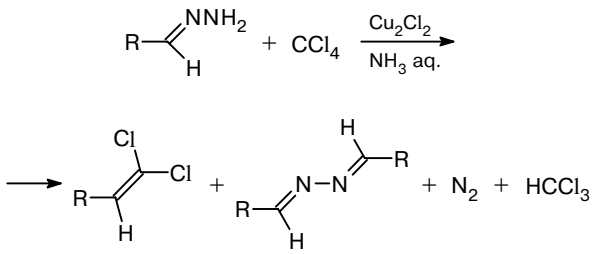
Previously, we have found a new redox reaction of *N*-unsubstituted hydrazones of aromatic aldehydes with carbon tetrachloride in the presence of catalytic amounts of copper(I) chloride giving rise to dichlorostyrenes.[14,15] This procedure represents a new approach to the transformation of the C=O bond into the C=C bond using inexpensive and readily accessible compounds, imposes minimum demands on equipment, and is characterized by the simplicity of the reaction procedure and isolation of the products. It was demonstrated that this procedure makes it possible to prepare dichlorostyrenes containing functional groups of different nature. Dichlorostyrenes containing electron-withdrawing substituents are formed in high yields, whereas the reactions of aldehyde hydrazones containing electron-donating substituents (for example, 4-Me_2N or 4-MeO) afford the target products in low yields (26—49%).[15]

In the present study, we suggest a mechanism of this reaction and propose a new catalytic cycle describing the processes that occur in the reaction system. In addition, we developed a simplified procedure for the synthesis of dichlorostyrenes, which, in some cases, enables one to obtain the target products in substantially higher yields.

Results and Discussion

The preparation of dichlorostyrenes from hydrazones is accompanied by elimination of nitrogen and formation of symmetrical aldazines and chloroform (Scheme 1). Dichlorostyrenes and azines are obtained in nearly quantitative cumulative yield.[15]

Scheme 1



We assumed that the carbenoid copper complex RCH=Cu analogous to the known carbenoid complexes of copper and other metals[16—22] is formed as the key intermediate in this reaction (Scheme 2). In the first stage, copper(I) chloride is oxidized by CCl_4, the latter being reduced to chloroform *via* the intermediate trichloromethyl anion (or dichlorocarbene)[23] and Cu_2Cl_2 being converted into $CuCl_2$. Previously, we have demonstrated that the reaction performed in the presence of $CuCl_2$ as a catalyst was accelerated by approximately two orders of magnitude resulting in substantial heat evolution and foaming of the reaction mixture, whereas the yields of dichlorostyrenes remained virtually unchanged.[15] Hence, we chose Cu_2Cl_2 as the optimum catalyst, which made it possible to control the course of the reaction. In the subsequent stage, Cu^{II} reacts with

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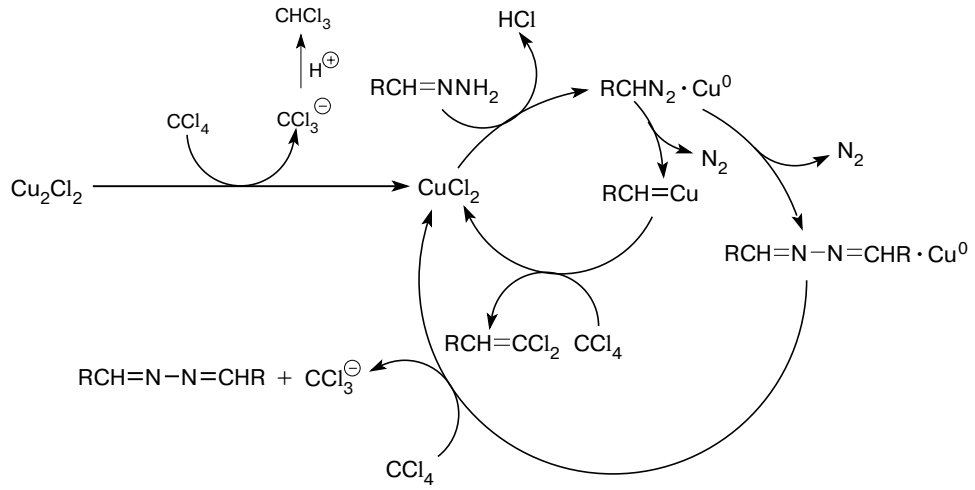
Scheme 2



the hydrazone to form the carbenoid copper complex (*via* the corresponding diazoalkane). In this stage, one nitrogen molecule and two HCl molecules are eliminated. The reaction of the resulting complex with CCl_4 affords the corresponding 1,1-dichloroalkene with regeneration of $CuCl_2$ and gives rise to a new catalytic cycle. The other path of decomposition of diazolalkane affords azine (the side reaction)[24] accompanied by elimination of nitrogen and formation of chloroform. This process is the major source of chloroform in the system (we have detected chloroform in the reaction mixture by ^1H NMR spectroscopy).

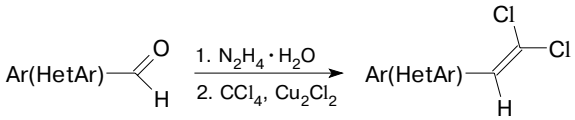
We believe that the major and side reactions share the key intermediate, both reactions being catalytic (the completion of each catalytic cycle leads to regeneration of the catalyst). Hence, the ratio between the reaction products (1,1-dichloroalkene and azine) depends on the ratio between hydrazone, CCl_4, and the catalysts in a complicated manner and, as a result, is governed by the kinetics of the competitive reactions.

Previously,[15] we have found the optimum reaction conditions (the ratios between the reagents, the catalyst, and the solvent) providing high yields of the target products in the case of aldehyde hydrazones containing electron-withdrawing substituents. The synthesis of aldehyde hydrazones containing electron-donating substituents presents difficulties due to their tendency to undergo oxidative dimerization under the action of atmospheric oxygen and problems associated with their isolation in the pure form.[25] One would expect that the one-pot reactions of such hydrazones will afford the target products in higher yields. This gave impetus to the development of a procedure for conversions of aldehydes into 1,1-dichloroalkenes without intermediate isolation of hydrazones.

We chose 4-*N*,*N*-dimethylaminobenzaldehyde as a model compound because of the extremely low stability of its hydrazone.[25] Previously, the corresponding dichlorostyrene **1a** has been obtained in the lowest yield (26%). Preliminary experiments demonstrated that 4-*N*,*N*-dimethylaminobenzaldehyde hydrazone, which was prepared *in situ* from equimolar amounts of the aldehyde and hydrazine, was converted under standard conditions[15] into the corresponding dichlorostyrene in higher yield compared to that obtained in the reaction of the hydrazone prepared separately. Further investigation of the conditions of the preparation of hydrazones showed that the optimum duration of the reaction of aldehyde with hydrazine is 3 h and that an excess of hydrazine leads to a decrease in the yield of dichlorostyrene **1a** from 58% (an equimolar ratio) to 29% (a 50% excess of hydrazine). Product **1a** was obtained in the highest yield at ~20 °C; a decrease or an increase in the temperature led to a decrease in the yield.

When employing the one-pot procedure developed by us, we succeeded in obtaining dichlorostyrenes **1a—c** in substantially higher yields (Table 1). It was demonstrated that the use of this procedure did not lead to a decrease (in some cases, led to an increase) in the yields of the corresponding dichlorostyrenes from aromatic aldehydes containing fluoro, chloro, bromo, nitro, cyano, methoxy, or hydroxy groups in the aromatic nucleus (Scheme 3).

Scheme 3



1a—z

Attempts to perform the reaction involving salicylaldehyde failed, but other *ortho*-substituted benzaldehydes, including 5-nitrosalicylaldehyde, entered into these re-

Table 1. Synthesis of dichloroalkenes from aldehydes

Product	Aryl (hetaryl)	Yield (%)	
		A	B
1a	4-$Me_2NC_6H_4$	58	26
1b	Ph	74	27
1c	4-$MeOC_6H_4$	77	49
1d	3-$MeOC_6H_4$	37	—
1e	4-HOC_6H_4	36	—
1f	4-$O_2NC_6H_4$	79	79
1g	3-$O_2NC_6H_4$	74	67
1h	4-NCC_6H_4	72	78
1i	3-NCC_6H_4	60	70
1j	4-BrC_6H_4	59	57
1k	3-BrC_6H_4	75	66
1l	4-ClC_6H_4	69	74
1m	3-ClC_6H_4	58	65
1n	2-ClC_6H_4	57	64
1o	2,6-$Cl_2C_6H_3$	68	79
1p	2,4-$Cl_2C_6H_3$	65	62
1q	4-FC_6H_4	52	—
1r	3-FC_6H_4	57	—
1s	2-FC_6H_4	36	—
1t	2-HO, 5-$O_2NC_6H_3$	40	14
1u	*p*-Phenylene	26	—
1v	1-Naphthyl	48	—
1w	3-Pyridyl	41	—
1x	4-Pyridyl	44	—
1y	2-Thienyl	34	—
1z	3-Thienyl	45	—

Note. A, the one-pot synthesis; B, from the corresponding hydrazones.

actions. Terephthalaldehyde gave the corresponding 1,4-bis(2,2-dichlorovinyl)benzene (**1u**).

The reactions have a general character. We succeeded in performing the reactions also with polyaromatic and heteroaromatic aldehydes. The reaction of 1-naphthaldehyde afforded the corresponding 1,1-dichloroalkene in 48% yield. The reactions of aldehydes of the thiophene and pyridine series also gave rise to the corresponding dichloroalkenes **1w**—**z**. However, the reactions of aldehydes of the pyrrole, furan, and indole series did not yield the target products.

To summarize, we developed a new convenient and simple procedure for conversions of aromatic and heteroaromatic aldehydes into the corresponding 1-aryl(hetaryl)-2,2-dichloroethenes in good yields. The mechanism of the reaction representing a new approach to the construction of the C=C bond is proposed.

Experimental

The IR spectra were recorded on a UR-20 spectrophotometer in thin films for liquids and in Nujol mulls for solids. TLC was carried out on Silufol UV-254 plates; visualization was performed with an acidified $KMnO_4$ solution, iodine vapor, or UV light. The ^1H and ^{13}C NMR spectra were measured on a Varian VXR-400 spectrometer (400 MHz for ^1H and 100 MHz for ^{13}C) in $CDCl_3$ and DMSO-d_6 with Me_4Si as the internal standard; the chemical shifts are given in the δ scale relative to Me_4Si with an accuracy of 0.01 ppm.

Commercial aldehydes were used as the starting compounds.

Synthesis of dichloroalkenes 1a—z (general procedure). A solution of an aldehyde (0.01 mol) in DMSO (5 mL) was added dropwise to a solution of 100% hydrazine hydrate (0.49 mL, 0.01 mol) in DMSO (5 mL). The reaction mixture was stirred for 3 h. Concentrated NH_4OH (3.33 mL) and Cu_2Cl_2 (100 mg, 1 mmol) were added to the resulting solution of hydrazone in DMSO. Then CCl_4 (5 mL, 50 mmol) was added dropwise at 20 °C for 10 min. The reaction mixture was stirred for 4 h and 0.1 *M* HCl (300 mL) was added. The reaction products were extracted with CH_2Cl_2 (3×50 mL), the extract was dried with Na_2SO_4, the solvent was evaporated, and the residue was purified by column chromatography on SiO_2. The spectral characteristics of compounds **1a—c**, **1f—p**, **1t**, and **1v** correspond to the data published in the literature.

4-(2,2-Dichlorovinyl)-*N*,*N*-dimethylaniline (1a): m.p. 70—71 °C (*cf.* lit. data[5]: m.p. 70—71 °C).

(2,2-Dichlorovinyl)benzene (1b): n_D^{20} 1.5880 (*cf.* lit. data[5]: n_D^{20} 1.5874).

1-(2,2-Dichlorovinyl)-4-methoxybenzene (1c): n_D^{20} 1.5978 (*cf.* lit. data[13]: n_D^{20} 1.5975).

1-(2,2-Dichlorovinyl)-3-methoxybenzene (1d): n_D^{20} 1.5825 (*cf.* lit. data[13]: b.p. 156—158 °C (3 Torr)). ^1H NMR, δ: 3.77 (s, 3 H, OMe); 6.80 (s, 1 H, —CH=); 6.83 (ddd, 1 H, Ar, *J* = 8.3, 2.6, and 1.0 Hz); 7.03—7.10 (br.d, 1 H, Ar, *J* = 7.6 Hz); 7.08 (s, 1 H, CH(2)); 7.25 (dd, 1 H, Ar, *J* = 8.3 and 7.6 Hz). ^{13}C NMR, δ: 55.1 (Me); 113.4 (Ar); 114.0 (Ar); 121.1 (=CCl_2); 121.2 (Ar); 128.4 (—CH=); 129.4 (Ar); 134.5 (Ar); 159.4 (C—O).

4-(2,2-Dichlorovinyl)phenol (1e): m.p. 92—93 °C. Found (%): C, 51.24; H, 3.26. $C_8H_6Cl_2O$. Found (%): C, 50.83; H, 3.20. IR, ν/cm^{-1}: 1620, 1590, 1380, 825. ^1H NMR, δ: 5.84 (s, 1 H, —CH=), 6.82 (d, 2 H, Ar, *J* = 8.6 Hz), 7.43 (d, 2 H, Ar, *J* = 8.6 Hz). ^{13}C NMR, δ: 115.6 (Ar), 119.0 (=CCl_2), 126.3 (Ar), 127.8 (—CH=), 130.3 (Ar), 155.2 (C—OH).

1-(2,2-Dichlorovinyl)-4-nitrobenzene (1f): m.p. 94 °C (*cf.* lit. data[5]: m.p. 94 °C).

1-(2,2-Dichlorovinyl)-3-nitrobenzene (1g): m.p. 54 °C (*cf.* lit. data[13]: m.p. 53—55 °C).

4-(2,2-Dichlorovinyl)benzonitrile (1h): m.p. 73—74 °C (*cf.* lit. data[13]: m.p. 75—76 °C).

3-(2,2-Dichlorovinyl)benzonitrile (1i): m.p. 33—34 °C (*cf.* lit. data[15]: m.p. 33—34 °C).

1-Bromo-4-(2,2-dichlorovinyl)benzene (1j): m.p. 24—25 °C (*cf.* lit. data[15]: m.p. 24—25 °C).

1-Bromo-3-(2,2-dichlorovinyl)benzene (1k): n_D^{17} 1.6220 (*cf.* lit. data[15]: n_D^{20} 1.6220).

4-Chloro-1-(2,2-dichlorovinyl)benzene (1l): n_D^{20} 1.5930 (*cf.* lit. data[1]: n_D^{20}1.5914).

3-Chloro-1-(2,2-dichlorovinyl)benzene (1m): b.p. 103—105 °C (1 Torr) (*cf.* lit. data[13]: b.p. 103—105 °C (1 Torr)).

2-Chloro-1-(2,2-dichlorovinyl)benzene (1n): n_D^{20} 1.5793 (*cf.* lit. data[1]: n_D^{20} 1.5793).

2,6-Dichloro-1-(2,2-dichlorovinyl)benzene (1o): n_D^{20} 1.5850 (*cf.* lit. data[5]: n_D^{20} 1.5853).

2,4-Dichloro-1-(2,2-dDichlorovinyl)benzene (1p): m.p. 49—50 °C (*cf.* lit. data[15]: m.p. 49—50 °C).

1-(2,2-Dichlorovinyl)-4-fluorobenzene (1q): n_D^{17} 1.5660. Found (%): C, 50.61; H, 3.04. $C_8H_5Cl_2F$. Found (%): C, 50.30; H, 2.64. IR, ν/cm^{-1}: 1610, 1240, 850. ^1H NMR, δ: 6.77 (s, 1 H, —CH=); 7.02 (dd, 2 H, Ar, *J* = 8.8 and 6.8 Hz); 7.48 (m, 2 H, Ar). ^{13}C NMR, δ: 115.4 (Ar, *J* = 21.8 Hz); 120.8 (=CCl_2); 127.4 (—CH=); 129.4 (Ar, *J* = 3.4 Hz); 130.4 (Ar, *J* = 8.1 Hz); 163.1 (C—F, *J* = 254 Hz).

1-(2,2-Dichlorovinyl)-3-fluorobenzene (1r): n_D^{17} 1.5685. Found (%): C, 50.56; H, 2.79. $C_8H_5Cl_2F$. Found (%): C, 50.30; H, 2.64. IR, ν/cm^{-1}: 1620, 1590, 1240, 790. ^{1}H NMR, δ: 6.76 (s, 1 H, —CH=); 6.96 (dd, 1 H, Ar, *J* = 8.3 and 2.5 Hz); 7.16—7.31 (m, 3 H, Ar). ^{13}C NMR, δ: 115.1 (Ar, *J* = 22.3 Hz); 115.4 (Ar); 122.3 (=CCl_2); 124.6 (—CH=); 127.4 (Ar, *J* = 2.3 Hz); 129.9 (Ar, *J* = 8.4 Hz); 135.2 (Ar, *J* = 8.2 Hz); 162.6 (C—F, *J* = 245 Hz).

1-(2,2-Dichlorovinyl)-2-fluorobenzene (1s): Found (%): C, 50.42; H, 2.59. $C_8H_5Cl_2F$. Found (%): C, 50.30; H, 2.64. IR, ν/cm^{-1}: 1610, 1590, 1240, 850. ^{1}H NMR, δ: 6.96 (s, 1 H, —CH=); 7.04 (ddd, 1 H, Ar, *J* = 9.9, 8.4, and 1.2); 7.12 (ddd, 1 H, Ar, *J* = 8.7, 7.7, and 1.2); 7.27 (m, 1 H, Ar); 7.76 (ddd, 1 H, Ar, *J* = 9.2, 7.7, and 1.5 Hz). ^{13}C NMR, δ: 115.4 (Ar, *J* = 21.9 Hz); 121.1 (—CH=, *J* = 5.9 Hz); 121.4 (Ar, *J* = 12.7 Hz); 123.2 (Ar, *J* = 1.7 Hz); 123.9 (=CCl_2, *J* = 3.7 Hz); 129.2 (Ar, *J* = 2.1 Hz); 130.1 (Ar, *J* = 8.4 Hz); 159.7 (C—F, *J* = 251 Hz).

2-(2,2-Dichlorovinyl)-4-nitrophenol (1t): m.p. 153—155 °C (*cf.* lit. data:[15] m.p. 153—155 °C).

1,4-Bis(2,2-dichlorovinyl)benzene (1u): m.p. 77—78 °C. IR, ν/cm^{-1}: 1610. ^{1}H NMR, δ: 6.75 (s, 2 H, —CH=); 7.47 (s, 4 H, Ar). ^{13}C NMR, δ: 122.5 (=CCl_2); 128.6 (—CH=); 129.4 (Ar); 134.1 (Ar). Found (%): C, 45.16; H, 2.40. $C_{10}H_6Cl_4$. Found (%): C, 44.82; H, 2.26.

1-(2,2-Dichlorovinyl)naphthalene (1v): m.p. 42 °C (*cf.* lit. data[26]: m.p. 42 °C).

3-(2,2-Dichlorovinyl)pyridine (1w): n_D^{20} 1.5985 (*cf.* lit. data[27]: n_D^{20} 1.5976). Found (%): C, 48.34; H, 3.10. $C_7H_5Cl_2N$. Found (%): C, 48.31; H, 2.90. IR, ν/cm^{-1}: 1620, 1590, 830. ^{1}H NMR, δ: 6.84 (s, 1 H, —CH=); 7.33 (dd, 1 H, Py, *J* = 8.1 and 4.3 Hz); 7.97 (br.d, 1 H, Py, *J* = 8.1 Hz); 8.55 (d, 1 H, C(6)H, *J* = 4.3 Hz); 8.68 (s, 1 H, C(2)H). ^{13}C NMR, δ: 123.2 (—CH=); 125.2 (Py); 129.0 (=CCl_2); 130.8 (Py); 134.9 (Py); 149.1 (Py); 149.8 (Py).

4-(2,2-Dichlorovinyl)pyridine (1x): a very unstable colorless oil, which rapidly blackened in air. ^{1}H NMR, δ: 6.79 (s, 1 H, —CH=); 7.41 (d, 2 H, Py, *J* = 5.9 Hz); 8.62 (d, 2 H, Py, *J* = 5.9 Hz). ^{13}C NMR, δ: 122.5 (Py); 126.2 (—CH=); 128.6 (=CCl_2); 140.4 (Py); 150.0 (Py).

2-(2,2-Dichlorovinyl)thiophene (1y): m.p. 35—36 °C. Found (%): C, 40.40; H, 2.33. $C_6H_4Cl_2S$. Found (%): C, 40.24; H, 2.25. IR, ν/cm^{-1}: 1610, 850. ^{1}H NMR, δ: 7.00—7.04 (m, 1 H, Het); 7.02 (s, 1 H, —CH=); 7.17 (br.d, 1 H, Het, *J* = 3.7 Hz); 7.36 (br.d, 1 H, Het, *J* = 5.1 Hz). ^{13}C NMR, δ: 118.6 (=CCl_2); 122.8 (—CH=); 126.6 (Het); 127.2 (Het); 129.3 (Het); 136.2 (Het).

3-(2,2-Dichlorovinyl)thiophene (1z): n_D^{17} 1.5795. Found (%): C, 40.18; H, 2.37. $C_6H_4Cl_2S$. Found (%): C, 40.24; H, 2.25. IR, ν/cm^{-1}: 1620, 1580, 840. ^{1}H NMR, δ: 6.82 (s, 1 H, —CH=); 7.25—7.31 (m, 2 H, Het); 7.54 (m, 1 H, Het). ^{13}C NMR, δ: 119.6 (=CCl_2); 122.8 (—CH=); 125.0 (Het); 127.3 (Het); 133.8 (Het).

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