toluene. The evacuated sample was sealed in a tube equipped with a break seal and placed in an 85° bath for 36 hr (10 half-lives); then the sample was opened on a vacuum line and the CO_2 was determined by absorption on Ascarite. Methane was identified by mass spectrometry. The products that were volatile at 25° and 0.05 Torr were collected in a Dry Ice cooled trap and made up to 25 ml with toluene. Cyclohexane was added as an internal standard and the vpc analysis (10 ft × $^3/_8$ in. 30% Carbowax 20M on Chromosorb W, 90°, 60 ml/min He) gave cyclohexane, acetone, *tert*-butyl alcohol, and toluene in that order. Identification was confirmed by isolation and spectral comparison with authentic samples.

In a separate experiment the product from 0.846 g (2.16 mmol) of **6** in 15 ml of toluene was treated as before, and 10 ml of toluene was removed by distillation. The residue was removed from the vacuum line and estimated to contain a 2.2% yield of acids (based on toluic acid) by comparison of the ir spectrum with those of authentic mixtures. This material was dissolved in 40 ml of ethanol and treated in the Brown[2] hydrogenator (1 atm of H_2, 5% Pd/C catalyst), 10 hr at 0°. The solution was filtered, the solvent was evaporated at room temperature, and the residue was dissolved in methylene chloride. An amorphous solid remained and after being rinsed several times with methylene chloride was identified as **13** (0.118 g, 0.44 mmol) by comparison with authentic material and conversion to the dimethyl ester **27**, also compared with authentic material. The methylene chloride solution was treated with excess diazomethane, made up to 25 ml, and dichlorobenzene was added as an internal standard. Vpc analysis (10 ft × $^3/_8$ in. 30% SE-30 on Chromosorb W, 75 ml/min He, programmed from 170 to 310°) gave the following peaks in order: dichlorobenzene (170°), *p*-methyl toluate (225°), bibenzyl (225°), **15** (225°), **14** (310°), and **27** (0.03 mmol) (310°).

In another experiment the product from 0.750 g (2.28 mmol) of **6** in 15 ml of toluene was concentrated under vacuum to 5 ml, and added very slowly to a solution of 1 g (0.03 mol) of powdered $LiAlH_4$ in 40 ml of dry THF at 0°. The solution was refluxed for 11 hr and cautiously hydrolyzed with water. The material was filtered through glass wool to remove the gelatinous precipitate, and the precipitate and glass wool were boiled with methylene chloride, which was filtered and combined with the previous extract. The solvent was evaporated, the residue was made up to 25 ml in CH_2Cl_2, and benzyl alcohol was added as an internal standard. Vpc analysis (10 ft × $^1/_8$ in. 5% Carbowax 20 M on Chromosorb G, 24 ml/min He, programmed from 175 to 245°) gave in order benzyl alcohol (175°), *p*-methylbenzyl alcohol (175°), bibenzyl (245°), and **17** (245°). Compound **18** was not detectably soluble in the solvents used, and *tert*-butyl benzyl ether was specifically shown to be absent.

The yields of products reported in Table II were determined from the integrations of the vpc curves. In all cases the integration was calibrated using weighed quantities of authentic materials with the internal standards.

Registry No.—**6**, 34201-98-4; **7**, 3965-62-6; **11**, 34202-00-1; **14**, 14518-67-3; **15**, 34224-28-7; **16**, 34224-29-8; **17**, 34224-30-1; **23**, 34224-31-2; **24**, 34224-32-3; **25**, 34224-33-4.

Cyclobutyl Sulfonate Solvolysis. Leaving Group Study

DONALD D. ROBERTS

Department of Chemistry, Louisiana Tech University, Ruston, Louisiana 71270

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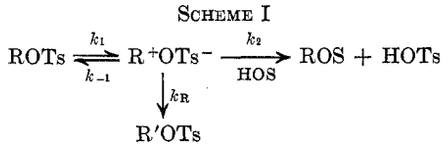
The solvolysis rates of a series of cyclobutyl para-substituted arenesulfonates have been determined in ethanol, acetic acid, and 2,2,2-trifluoroethanol. In addition, the solvolysis rates of cyclobutyl methanesulfonate have been determined in the same series of solvents. The data indicate that the partitioning of the carbonium ion among solvolysis and internal return isomerization routes is insensitive to anion solvation and charge dispersal effects but is sensitive to the steric bulk of the leaving group. The product distributions are also insensitive to the changes in the leaving group.

Recently, it was established[1] that cyclobutyl β-naphthalenesulfonate suffers solvolysis in a spectrum of solvents with little nucleophilic assistance by solvent but with considerable anchimeric assistance.

The nature of the cationic species responsible for the observed anchimeric assistance is interpreted[2] in terms of a rapidly equilibrating bicyclobutonium ion system which is common to solvolysis of cyclopropylcarbinyl and allylcarbinyl derivatives as well as cyclobutyl substrates.

The proposed cationic species have open three options: internal return (k_{-1}), internal return isomerization (k_R), and solvolysis (k_2), as illustrated in Scheme I.

SCHEME I



The internal return isomerization route is well documented for the cyclopropylcarbinyl system.[3,4]

The presence of internal return isomerization has also been observed in the solvolysis reactions of cyclobutyl derivatives.[1,5] In keeping with the common cationic species postulated for both the cyclopropylcarbinyl and cyclobutyl derivatives in acetolysis reactions, about 10% of both substrates isomerize by the k_R route to allylcarbinyl derivatives.[1,4]

An examination of the literature suggests that the relative importance of the competing reactions, k_2/k_R, is sensitive to change in the nature of the leaving group. For example, the acetolysis of cyclobutyl chloride is accompanied by 40% internal return isomerization[5] while the acetolysis of cyclobutyl β-naphthalenesulfonate is accompanied by only 8% internal return isomerization.[1] Both cyclobutyl derivatives, however, exhibit the same solvent dependency behavior, *i.e.*, reduced internal return isomerization with increased solvent ionizing strength.[1,6]

These results contrast with the findings of related work. Thus Goering and coworkers,[7] using a combination of polarimetric and titrimetric kinetic techniques, demonstrated that the relative rates of com-

(1) D. D. Roberts, *J. Org. Chem.*, **36**, 1913 (1971).
(2) K. L. Servis and J. D. Roberts, *J. Amer. Chem. Soc.*, **86**, 3773 (1964).
(3) K. L. Servis and J. D. Roberts, *Tetrahedron Lett.*, 1369 (1967).
(4) D. D. Roberts, *J. Org. Chem.*, **35**, 4059 (1970).
(5) J. D. Roberts and R. H. Mazur, *J. Amer. Chem. Soc.*, **73**, 2509 (1951).
(6) C. Y. Wu and R. E. Robertson, *ibid.*, **88**, 2666 (1966).
(7) H. L. Goering and E. F. Silversmith, *ibid.*, **77**, 6249 (1955), and previous papers cited therein.

TABLE I

SOLVOLYSIS RATES FOR CYCLOBUTYL SULFONATES

Registry no.	Sulfonate	Solvent	Temp, °C	k_1, 10^6 sec^{-1}	Infinity, %
34236-40-3	*p*-Nitrobenzene	EtOH	40	74 ± 0.8	100
			50	233 ± 3	100
			60	740 ± 8	
	p-Nitrobenzene	AcOH	30	92 ± 1	92
			40	195 ± 2	
			50	610 ± 6	90
	p-Nitrobenzene	CF_3CH_2OH	25	920 ± 8	92
34236-41-4	*p*-Bromobenzene	EtOH	40	11 ± 0.2	
			50	33 ± 0.3	100
			50	33 ± 0.3[a]	100
			60	117 ± 2	
	p-Bromobenzene	AcOH	30	9.7 ± 0.1	92
			40	36 ± 0.3	93
			50	120 ± 2	90
			50	120 ± 2[b]	90
			50	127 ± 2[c]	92
			50	123 ± 2[d]	98
			60	380 ± 4	92
	p-Bromobenzene	CF_3CH_2OH	25	172 ± 3	92
			35	520 ± 6	91
34236-42-5	Benzene	EtOH	40	3.6 ± 0.04	
			50	11 ± 0.2	99
			60	39 ± 0.4	
	Benzene	AcOH	30	4.3 ± 0.04	94
			40	16 ± 0.2	88
			50	50 ± 0.5	92
			50	55 ± 0.2[e]	92
	Benzene	CF_3CH_2OH	25	83 ± 2	93
34236-43-6	*p*-Methylbenzene	EtOH	40	1.8 ± 0.01	
			50	6.1 ± 0.3	100
			50	6.1 ± 0.4[f]	100
			60	20 ± 0.4	
	p-Methylbenzene	AcOH	30	2.6 ± 0.03	92
			40	8.9 ± 0.1	
			50	33 ± 0.2	92
			50	34 ± 0.3[g]	92
	p-Methylbenzene	CF_3CH_2OH	25	53 ± 0.1	92
34236-44-7	*p*-Methoxybenzene	EtOH	40	1.39 ± 0.03	
			50	3.6 ± 0.1	100
			60	12.5 ± 0.2	
	p-Methoxybenzene	AcOH	30	1.7 ± 0.03	93
			40	6.1 ± 0.04	
			50	21 ± 0.2	87
	p-Methoxybenzene	CF_3CH_2OH	25	36 ± 0.5	90
34236-45-8	Methane	EtOH	50	4.7 ± 0.1[h]	100
	Methane	AcOH	50	47 ± 0.5[h]	80
	Methane	CF_3CH_2OH	25	50 ± 0.3	80

[a] Sample 0.05 *M* in urea and 0.030 *M* in ester. [b] Sample 0.033 *M* in *p*-toluenesulfonic acid and 0.030 *M* in ester. [c] Sample 0.0475 *M* in KOAc and 0.030 *M* in ester. [d] Sample 0.05 *M* in NaOAc and 0.030 *M* in ester. [e] Sample 0.0475 *M* in KOAc and 0.030 *M* in ester. [f] Sample 0.05 *M* in urea and 0.030 *M* in ester. [g] Sample 0.0475 *M* in KOAc and 0.030 *M* in ester. [h] Duplicate runs.

peting solvolysis and internal return rearrangement of various 5-methyl-2-cyclohexenyl derivatives were insensitive to changes in the nature of the leaving group (chloride, acid phthalate, and *p*-nitrobenzoate). Winstein and collaborators[8] also found that the relative rates of competing solvolysis and internal return racemization of norbornyl derivatives were insensitive to changes in the nature of the leaving group (chloride, bromide, iodide, and brosylate).

For some time the arenesulfonate group has been regarded by various authors[9] as well solvated relative to a monatomic leaving group, since it contains oxygen atoms which are capable of hydrogen bonding. The resulting increase in the solvation forces associated with the arenesulfonate leaving group should promote solvolysis and retard internal return relative to the chloride leaving group. This tendency should be modified as the charge density on the oxygen atoms in the arenesulfonate group is modified. To test the validity of this rationale the solvolysis study of a series of cyclobutyl para-substituted arenesulfonates was undertaken. During the course of this study some related aspects were investigated and are included in this paper in order to gain further insight as to the behavior of the cationic species involved in the cyclopropylcarbinyl–cyclobutyl solvolysis reactions.

(8) S. Winstein and G. C. Robinson, *J. Amer. Chem. Soc.*, **80**, 169 (1958), and previous papers cited therein.

(9) For example, see S. Winstein, A. H. Fainberg, and E. Grunwald, *ibid.*, **79**, 4146 (1957).

The data indicate that the partitioning of the carbonium ion among solvolysis and internal return isomerization routes is insensitive to anion solvation and charge dispersal effects but is sensitive to the size of the leaving group.

The first-order rate constants for solvolysis of cyclobutyl mesylate and a series of para-substituted arenesulfonates in various solvents are summarized in Table I. The reaction progress was followed by titrating the liberated alkyl and arylsulfonic acid. The solvolysis reactions in ethanol were unaccompanied by internal return isomerization and were cleanly first order up to 90% reaction. The solvolysis reactions in acetic acid and 2,2,2-trifluoroethanol were accompanied by internal return isomerization and consequently the apparent first-order rate constants, k_t, in these two solvents were computed on the basis of the acid infinity titer. The fact that the infinity titers in buffered and unbuffered reactions were identical supports an internal return isomerization and not a competing acid-catalyzed isomerization.

The sign and response of the calculated ρ valves, 1.75 for ethanolysis, 1.43 for acetolysis, and 1.38 for 2,2,2-trifluoroethanolysis, support a varying negative charge density at the oxygen atoms in the arenesulfonate leaving group; *i.e.*, the observed rate constant, k_t, is enhanced by para substituents with $+\sigma^n$ values.[10,11] For example, the *p*-nitrobenzenesulfonate relative to the *p*-methoxybenzenesulfonate is some 65 times more reactive in ethanol, 29 times more reactive in acetic acid, and 25 times more reactive in 2,2,2-trifluoroethanol.

The product distribution data listed in Table II

TABLE II

ACETOLYSIS PRODUCTS FOR CYCLOBUTYL SULFONATES[a]

Sulfonate	c-C_4H_7OS, %	c-C_3H_5-CH_2OS, %	CH_2=$CHCH_2CH_2$-OS, %
p-Bromobenzene	41	55	4
p-Methane	41	55	4
p-Methoxybenzene	42	54	4

[a] At 50° with urea buffer present.

reveal no dependency upon the nature of the leaving group. This marked insensitivity of the product distribution upon leaving group is consistent with the solvolytic behavior of the cyclopropylcarbinyl substrates.[12]

These results strongly suggest that the charge dispersal distribution in the reactive intermediate captured by solvent varies little with anion; *i.e.*, the nature of the anion in the intimate ion pair has little apparent affect upon the partitioning of the nonclassical cation[14-16] among the various product pathways.

(10) P. R. Wells, *Chem. Rev.*, **63**, 178 (1963).

(11) Not unexpectedly, the *p*-methoxy substituent fits this correlation only when a regular σ value (−0.268) is used which includes the resonance contribution.

(12) For example, the acetolysis of both cyclopropylcarbinyl chloride[5] and tosylate[13] yield essentially the same product distribution.

(13) D. D. Roberts, *J. Org. Chem.*, **29**, 294 (1964).

(14) P. D. Bartlett, "Nonclassical Ions," W. A. Benjamin, New York, N. Y., 1965.

(15) W. Bruce Kover and J. D. Roberts, *J. Amer. Chem. Soc.*, **91**, 3687 (1969).

(16) The nonclassical ion postulated for both the cyclopropylcarbinyl and cyclobutyl systems is obviously in a state of rapid development; for an alternative view see Z. Majerski and P. v. R. Scleyer, *J. Amer. Chem. Soc.*, **93**, 665 (1971).

An examination of the competing solvolysis and internal return isomerization reactions (k_2/k_R) is afforded by the derived data in Table III. The k_2/k_R ratio is sensitive to the solvent change from ethanol to acetic acid but insensitive to the change from acetic acid to 2,2,2-trifluoroethanol, while the observed rate constant, k_t, exhibits the opposite solvent sensitivity, a small twofold increase in changing from ethanol to acetic acid and a 40-fold increase in changing from acetic acid to 2,2,2-trifluoroethanol. This observation is true for the entire spectrum of leaving groups investigated in this study.

TABLE III

SOLVOLYSIS–INTERNAL RETURN ISOMERIZATION RATE RATIOS FOR c-C_4H_7X

X	Solvent	Temp, °C	% rearrangement	k_2/k_R
Cl[a]	AcOH	100	43	1.32
OSO_2CH_3	AcOH	50	20	4.00
	EtOH	50	0	
	CF_3CH_2OH	25	20	4.00
p-$YC_6H_4OSO_2$[b]	AcOH	50	8	11.5
	EtOH	50	0	
	CF_3CH_2OH	25	8	11.5

[a] Taken from data of ref 5. [b] Y = NO_2, Br, H, CH_3, OCH_3.

More significantly, the data in Table III reveal that the magnitude of the k_2/k_R ratio is insensitive to charge dispersal in the arenesulfonate leaving group, but is sensitive to change in steric bulk of the leaving group. Thus, in acetic acid the value of k_2/k_R is 1.32 for cyclobutyl chloride, 4.0 for cyclobutyl mesylate, and 11.5 for the various para-substituted cyclobutyl arenesulfonates.

This result clearly indicates that variation of the k_2/k_R ratio is not due to enhanced solvation of the arenesulfonate leaving group. It is unlikely that the tosylate group, with its greater steric bulk and charge dispersal, would be more strongly solvated than the methanesulfonate group, yet in acetolysis reactions, 20% of the cyclobutyl mesylate undergoes internal return isomerization as compared to only 8% of cyclobutyl tosylate. Both esters suffer acetolysis at approximately the same rate as does cyclobutyl β-naphthalenesulfonate, an ester whose acetolysis is also accompanied by only 8% internal return isomerization.

Inspection of models of the various cyclobutyl derivatives provides a convenient organization of the leaving groups into three steric bulk groups: small (chloride), medium (methanesulfonate), and large (arenesulfonate). The tempting correspondence between the leaving group steric bulk and the extent of internal return isomerization is readily apparent.

An appreciation of the proposal[2] that the intimate ion pair is described as a set of rapidly equilibrating ion pairs sheds some light on this correspondence. It is possible that in this dynamic situation the blend of electrostatic, polarization, and covalencey forces[17] that stabilizes as well as leads to the collapse of the ion pair intermediate is responsive to the change in anion steric bulk, which in turn affects the relative heights of the energy barriers for internal return and solvolysis.

(17) S. Winstein and G. C. Robinson, *J. Amer. Chem. Soc.*, **80**, 169 (1958).

Experimental Section

A Beckman GC-4 chromatographic instrument equipped with a thermal conductivity detector and an 8 ft × 0.25 in. column of 20% diethylene glycol succinate on Chromosorb W, AW-DMCS (45–60 mesh) and a Bausch and Lomb IR 270 spectrophotometer were used for analytical work.

Cyclobutyl *p*-nitrobenzenesulfonate resulted when *p*-nitrobenzenesulfonyl chloride (7.2 g, 35 mmol) was mixed with cyclobutanol (2.16 g, 30 mmol) and 40 ml of dry pyridine at 0°. After standing for 20 hr at 0°, the thick reaction mixture was carefully hydrolyzed by the slow addition of 20 ml of cold water at 0–5° followed by the rapid addition of sufficient cold, dilute HCl to acidify the mixture. The precipitated ester was separated on a Buchner funnel and washed several times with cold, dilute HCl, several times with cold water and then with cold petroleum ether (bp 30–60°) to yield the crude ester. Recrystallization from petroleum ether–benzene gave 5.0 g (65%) of light yellow crystals, mp 64–68°. Two additional recrystallizations yielded the analytical sample, mp 68–69°.

Anal. Calcd for $C_{10}H_{11}NO_5S$: C, 46.69; H, 4.31; N, 5.45. Found: C, 46.63; H, 4.39; N, 5.60.

Cyclobutyl *p*-bromobenzenesulfonate was prepared from *p*-bromobenzenesulfonyl chloride as described above in 63% yield: mp (after two recrystallizations from 33:1 petroleum ether–benzene) 53–54° (lit.[18] mp 52–53.5°).

Cyclobutyl Benzenesulfonate.—To 3.6 g (50 mmol) of cyclobutanol in 50 ml of dry pyridine cooled to 0° was added 9.7 g (55 mmol) of benzenesulfonyl chloride. After standing for 24 hr at 5° and an additional 28 hr at 18°, the mixture was carefully hydrolyzed by the slow addition of 20 ml of cold water at 0–5° followed by the rapid addition of sufficient cold, dilute HCl to acidify the mixture. The separated oil was taken up in 40 ml of methylene chloride, washed twice with cold, dilute HCl and once with cold, saturated $NaHCO_3$, dried over Na_2SO_4, and concentrated to yield an oil. The crude ester was purified twice by stirring with petroleum ether, freezing the undissolved oil at −78°, decanting off the solvent, and removing the last traces of solvent under reduced pressure (*ca.* 0.1 mm) to yield 5.5 g (52%) of an oil. The purity, calculated from infinity titers of the ethanolyses, was 99%. The ir spectrum, ν_{SO_2} (asymmetric) 1360 and ν_{SO_2} (symmetric) 1170 cm^{-1}, was consistent with the assigned structure.

Cyclobutyl *p*-toluenesulfonate was prepared in 50% yield by published procedure:[19] mp 25° (lit.[19] mp 24–25°); ir (neat) 1357 (ν_{SO_2}, asymmetric) and 1173 cm^{-1} (ν_{SO_2} symmetric).

Cyclobutyl *p*-methoxybenzenesulfonate was prepared from *p*-methoxybenzenesulfonyl chloride as described for cyclobutyl benzenesulfonate in 59% yield. The purity of the oil, calculated from infinity titers of the ethanolyses, was 99%. The ir spectrum, ν_{SO_2} (asymmetric) 1358 and ν_{SO_2} (symmetric) 1170 cm^{-1}, was consistent with assigned structure.

Cyclobutyl methanesulfonate was prepared from methanesulfonyl chloride as described for cyclobutyl benzenesulfonate in 40% yield. The purity of the oil, calculated from infinity titers of the ethanolyses, was 99%; ir (neat) 1340 (ν_{SO_2}, asymmetric) and 1170 cm^{-1} (ν_{SO_2}, symmetric).

Solvents.—Absolute ethanol was prepared according to the method of Fieser.[20] Acetic acid solvent was prepared from 994.9 ml of glacial acetic acid (Matheson Scientific, 99.8%) and 5.1 ml of acetic anhydride. 2,2,2-Trifluoroethanol (Aldrich Chemical Co.) was redistilled just prior to use.

Acetolysis Product Studies. A. Cyclobutyl *p*-Bromobenzenesulfonate.—Cyclobutyl *p*-bromobenzenesulfonate (1.5 g, 5 mmol) was dissolved in sufficient acetic acid solvent (containing 7 mmol of urea) to give 25 ml of solution. After 10 half-lives at 50°, the solution was diluted with 150 ml of water and continuously extracted with ether for 24 hr. The ether extract was neutralized with $NaHCO_3$ and dried (Na_2SO_4), and most of the solvent was removed by distillation. Analysis by gc revealed, in addition to solvent, the presence of allylcarbinyl acetate, cyclobutyl acetate, and cyclopropylcarbinyl acetate in the ratio 1.0:10.2:13.7, respectively.

B. Cyclobutyl *p*-Methoxybenzenesulfonate.—Cyclobutyl *p*-methoxybenzenesulfonate (1.2 g, 5 mmol) was solvolyzed in 25 ml of acetic acid solvent (containing 7 mmol of urea) for 10 half-lives at 50°. The material was worked up as before and analysis by gc revealed, in addition to solvent, the presence of allylcarbinyl acetate, cyclobutyl acetate, and cyclopropylcarbinyl acetate in the ratio 1.0:10.5:13.5, respectively.

C. Cyclobutyl Methanesulfonate.—Cyclobutyl methanesulfonate (0.53 g, 5 mmol) was solvolyzed as above for 10 half-lives at 50°. The material was worked up as before and analysis by gc revealed, in addition to solvent, the presence of allylcarbinyl, cyclobutyl, and cyclopropylcarbinyl acetate in the ratio 1.0:10.2:13.7, respectively.

Rate measurements were accomplished by usual ampoule techniques.[21] The titrating solutions were, for ethanolysis and 2,2,2-trifluoroethanolysis, 0.020 *N* sodium methoxide in anhydrous methanol and, for acetolysis, 0.020 *N* sodium acetate in acetic acid. The indicators used were bromthymol blue (in water), bromphenol blue (in 20% aqueous EtOH), and bromphenol blue (in acetic acid), respectively.

Registry No.—EtOH, 64-17-5; AcOH, 64-19-7; CF_3CH_2OH, 75-89-8.

(18) I. Lillien and L. Handloser, *J. Amer. Chem. Soc.*, **93**, 1682 (1971).
(19) J. D. Roberts and V. C. Chambers, *ibid.*, **73**, 5034 (1951).
(20) L. F. Fieser, "Experiments in Organic Chemistry," 3rd ed, D. C. Heath, Boston, Mass., 1957, p 285.
(21) D. D. Roberts, *J. Org. Chem.*, **29**, 294 (1964).

The Electrochemical Reduction of Aromatic Acids to the Corresponding Aldehydes

JOHN H. WAGENKNECHT

Central Research Department, Monsanto Company, St. Louis, Missouri 63166

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A number of aromatic carboxylic acids have been found to undergo electrochemical reduction to the corresponding aldehydes. Electron-donating substituents in the para position inhibit the reaction. It is possible to predict from the pK_a value of the acid or, more accurately, from the polarographic half-wave potential of the corresponding methyl ester whether or not the method is applicable for the preparation of an aldehyde.

The chemical reduction of a carboxylic acid to the corresponding aldehyde generally involves initial formation of a derivative of the acid other than a salt and subsequent reduction of the derivative. Calcium or manganese salts of carboxylic acids can be converted to the aldehydes by pyrolysis in the presence of the corresponding formate salts.[1,2] Similarly, the vapor phase reaction of a mixture of a carboxylic acid and formic acid over TiO_2 or ThO_2 leads to the aldehyde.[3] Although the reduction of carboxylic acids to the aldehydes by $(i\text{-Bu})_2AlH$ has been reported,[4] aluminum and boron hydrides generally reduce acids to the corresponding alcohols. However, acids such as perfluoroaliphatic acids, oxalic acid, or salicylic acid are re-

(1) E. Müller, "Methoden der Organische Chemie" (Houben-Weyl), Vol. VII, Part 1, Georg Thieme Verlag, Stuttgart, 1954, p 277.
(2) P. Mastagli, P. Lambert, and C. Hirigoyen, *C. R. Acad. Sci.*, **248**, 1830 (1959).
(3) P. Sabatier and A. Mailhe, *ibid.*, **154**, 561 (1912).
(4) L. I. Zakharkin and I. M. Khorlina, *Zh. Obshch. Khim*, **34**, 1029 (1964).