p-bromophenacyl ester, mp 61–62° (lit. mp 58°,[23] 60°[24]), were prepared as derivatives.

n-Butyl 3-*n*-butyloxypropionate[21,22] was obtained in 51% yield, bp 91–92° (3.5 mm), $n^{20}D$ 1.4223, d^{20}_4 0.911, sapon equiv 198 (calcd 202). A small amount of acrylamide, mp 83–84° (lit.[28] mp 84.5°), codistilled with the ester and was identified from its ir spectrum.

Methanolysis of 5.32 g (0.0336 mol) of methyl 3-chloropropionimidate hydrochloride in the refrigerator for 2 days gave 1.84 g (100%) NH_4Cl and 2.00 g of liquid, bp 62–65° (32 mm), $n^{20}D$ 1.4036, d^{20}_4 1.044, which was shown by glpc to be a 73:27 (by weight) mixture of methyl 3-methoxypropionate (0.0124 mol) and methyl 3-chloropropionate (0.0044 mol).

Methanolysis of Methyl 4-Bromobutyrimidate Hydrochloride.—A solution of 8.5 g (0.039 mol) of the imidate salt in 50 ml of absolute methanol was kept in the refrigerator for 5 days. The precipitated NH_4Cl (1.0 g, 0.019 mol) was filtered and the filtrate made alkaline to phenolphthalein with 0.021 mol of methanolic NaOMe.[25] Distillation gave 5.4 g (61%) of methyl ortho-4-bromobutyrate, bp 85–95° (15 mm), $n^{20}D$ 1.4563, d^{20}_4 1.370, 34.8% Br (calcd 35.2). The product displayed a carbonyl absorption at 1740 cm^{-1} in the ir and glpc showed 5% lower boiling impurity, presumably the normal ester.

Hydrolyses of Imidate Hydrochlorides.—To an ice-cooled slurry of 15.3 g (0.076 mol) of methyl 3-bromopropionimidate hydrochloride in 35 ml of ether was added 1.51 g (0.084 mol) of water with stirring over a 15-min period. Stirring was continued for an additional 30 min and the mixture placed in the refrigerator overnight. The ether solution was decanted from the insoluble solid and distilled to give 9.29 g of liquid, bp 71–80° (32 mm), $n^{20}D$ 1.4455, d^{20}_4 1.393, which was found by glpc to be a 70:30 (by weight) mixture of methyl 3-bromopropionate (0.039 mol) and methyl 3-chloropropionate (0.023 mol). The yield of ammonium halide was 5.76 g (calcd for NH_4Cl 4.05 g, for NH_4Br 7.40 g). Both AgCl and AgBr were obtained by treatment of the salt in aqueous HNO_3 with $AgNO_3$. The toal yield of ammonium halide was 94% (Mohr method).

Ethyl 3-bromopropionimidate hydrochloride was hydrolyzed in dioxane solution with a 20-fold molar excess of water and also in aqueous solution (0.050 mol in 100 ml of water). In both cases, the product was a 55:47 (by weight) mixture of the bromo and chloro esters.

In none of the three hydrolyses was there any evidence of alkyl hydracrylate by glpc.

Pyrolysis of Ethyl 3-Bromopropionimidate Hydrochloride.—7.38 g (0.0341 mol) of the imidate salt was heated at 110–113° in a distillation apparatus with an ice-cooled receiver and a cold trap in Dry Ice–acetone to condense ethyl chloride. The salt melted with decomposition and 2.68 g (72%) of crude ethyl bromide, bp 34–35°, $n^{25}D$ 1.4145, d^{20}_4 1.38, distilled into the receiver but no ethyl chloride was found in the cold trap. Both receiver and trap contained hydrogen halide (odor, fuming in moist air).

The pale yellow residual oil (4.49 g) became a waxy solid on cooling. Repeated extraction with boiling benzene left 2.68 g of an ethanol- and water-soluble solid. Treatment of its aqueous solution with $AgNO_3$ and HNO_3 gave a heavy precipitate of silver halide which darkened rapidly indicating it to be AgCl. Another portion of the aqueous solution was made strongly basic with NaOH and boiled until ammonia evolution had subsided. On acidification, the residual solution remained clear.

Concentration and cooling of the benzene extract precipitated 1.10 g of 3-chloropropionamide, mp 100.5–102.5° (lit. mp 102.5°,[5] 103°[19]).

Acidity of Alcoholysis Reaction Mixtures and Related Solutions.—The pH values determined (glass electrode) for absolute ethanol and 0.03 *M* solutions of various pertinent substances in this solvent are shown in Table II.

TABLE II

Solute	pH
None	5.4
NH_4Cl	3.6
NH_4Br	3.1
CH_3COOEt	5.2
$ClCH_2CH_2COOEt$	3.4
$BrCH_2CH_2COOEt$	3.7
$BrCH_2CH_2C(OEt)=NH_2^+Cl^-$	2.0 (3 min)
	0.9 (61 min)
	0.6 (317 min)
	0.8 (95 hr)
$CH_3CH_2C(OEt)=NH_2^+Cl^-$	2.9 (2 min)
	2.3 (52 min)
	2.0 (143 min)
	2.0 (19 hr)

Acknowledgment.—Acknowledgment is made to the donors of The Petroleum Research Fund, administered by the American Chemical Society, for support of this research.

(28) E. L. Carpenter and H. S. Davis, *J. Appl. Chem.* (London), **7**, 671 (1957).

Arylation of Aromatic Compounds by Electrochemical Reduction of Benzenediazonium Tetrafluoroborate in Aprotic Solvents

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Benzenediazonium tetrafluoroborate has been reduced electrolytically at 0 V *vs.* sce in aprotic solvents to produce a free phenyl radical. The phenyl radical so formed has been used for phenylation of benzene, toluene, anisole, benzonitrile, nitrobenzene, bromobenzene, and naphthalene. The isomer ratios, total rate ratios, and partial rate factors agree with those obtained using benzoyl peroxide or N-nitrosoacetanilide as sources of free radicals and demonstrate that the mechanism of phenylation is completely free radical.

Arylation of aromatic substrates by homolytic substitution was first carried out by Kuhling[1] and Gomberg and coworkers.[2] Subsequently a vast literature has arisen on the nature of the reactions involved in homolytic aromatic substitution together with the results of quantitative measurement of isomer ratios. The present position on most of these reactions has been summarized by Williams[3] and Hey.[4]

The possibility of producing free aryl radicals by direct electrolytic reduction of diazonium cations by a single electron step was raised by the discovery of their polarographic activity.[5,6] Kochi[7] attempted to correlate the half-wave potentials of diazonium salts

(1) O. Kuhling, *Ber.*, **28**, 41 (1895), **29**, 165 (1896).

(2) M. Gomberg and W. E. Bachmann, *J. Amer. Chem. Soc.*, **46**, 2339 (1924).

(3) G. H. Williams, "Homolytic Aromatic Substitution," Pergamon Press, London, 1960.

(4) D. H. Hey, *Advan. Free-Radical Chem.*, **2**, Chapter 2 (1967).

(5) R. M. Elofson and P. A. Mecherly, *Anal. Chem.*, **21**, 565 (1949).

(6) E. R. Atkinson, H. H. Warren, P. I. Abel, and R. E. Wing, *J. Amer. Chem. Soc.*, **72**, 915 (1950).

(7) J. K. Kochi, *ibid.*, **77**, 3208 (1955).

with substituent effects. Some of the electrochemical complications of carrying out such reactions in aqueous solution with stirred mercury electrodes were pointed out by Elofson.[8] More recently Elofson and Gadallah[9] studied the polarographic reduction of several substituted benzenediazonium tetrafluoroborates in aprotic solvents, acetonitrile, or sulfolane, using tetrabutylammonium perchlorate as the supporting electrolyte. Under these conditions normal one-electron reduction steps uncomplicated by adsorption effects and side reactions at the electrode were obtained according to eq 1. This suggested the possibility that substituted

$$ArN_2^+ + e \rightarrow ArN_2\cdot \rightarrow Ar\cdot + N_2 \quad (1)$$

biaryls could be synthesized by the action of electrolytically generated aryl radicals on aromatic substrates in dipolar aprotic solvents.

Experimental Section

Reagents.—All liquid reagents and solvents were dried over anhydrous sodium sulfate, fractionally distilled, degassed by bubbling purified nitrogen through them, and stored in the dark in air-tight containers. Acetonitrile, reagent grade, was kept over sodium carbonate for 24 hr, fractionally distilled from phosphorus pentoxide, and used directly. Solid reagents and reference compounds were recrystallized and the melting points agreed with the literature values. Benzenediazonium tetrafluoroborate was recrystallized from dry methanol and ether and kept under vacuum in the refrigerator.

General Procedure for the Electrolytic Reduction.—Controlled-potential reductions were done with the apparatus described by Lingane and Jones[10] at 0 V *vs.* sce. Cathode and anode compartments were separated by a very fine fritted-glass disk (nominal maximum pore size 2–2.5 μ, supplied by Corning Glass Works, Corning, N. Y.). Purified nitrogen was bubbled slowly through the reaction mixtures before and during the reaction and the temperature was maintained at 0 ± 2°. Slow stirring with a teflon-coated magnet was employed.

Two sets of reactions were performed for each aromatic substrate: one to establish the total yield and isomer ratio, and the other, the competitive reaction to obtain the total rate ratio ($_H^XK$) and partial rate factor (F_T).

Tetrabutylammonium perchlorate[11] was dissolved in a solution of acetonitrile[12] and the aromatic substrate (and benzene for the competitive runs) to 0.1 *M*. This solution was used for both compartments. The cathode compartment contained 1 mol each of acetonitrile and the aromatic substrate and 0.01 or 0.005 mol of the diazonium salt. For competitive runs, benzene was added to the solution in equimolar ratio to the aromatic substrate before the diazonium salt was added. The reaction mixtures were pale yellow throughout the reactions except in the case of nitrobenzene when dark colors developed as the reaction proceeded. The initial current varied between 70 and 200 mA depending on the resistance of the cell and the amount of diazonium salt used. The reaction was considered complete when the test for diazonium salt with α-naphthol was negative and the current dropped to less than 1 mA.

Identification and Estimation of Products.—All reaction mixtures were analyzed by gas–liquid partition chromatography (glpc) using the internal standard method or the calibration curve method. The different isomeric products and biphenyl were identified by retention times and were collected for verification by infrared or mass spectra.

After the completion of each reaction, acetonitrile was distilled off under vacuum at room temperature, dry ether was added, and the precipitated tetrabutylammonium perchlorate was filtered off. The ether solution was concentrated under vacuum to about 50 ml and used directly for glpc analysis.

Yields, isomer ratios, total rate ratios, and partial rate factors are mean values from duplicate or triplicate runs. No change was produced in any of these values for any aromatic substrate by halving the concentration of diazonium salt.

Yields of benzene, up to 50%, were detected in all isomer ratio reactions when benzene was not a reactant. No attempt was made to identify other by-products.

The columns used and retention times from injections for the reaction products using the various aromatic substrates were as follows: phenylation of toluene—column 6 ft × $^3/_{16}$ in., asphalt (16% w/w) on Chromosorb W (nonacid washed) at 165°, retention times for 2-, 3-, and 4-methylbiphenyls, biphenyl, and bibenzyl 16, 20, 29, 17.5, and 30.5 min, respectively; phenylation of anisole—column 8 ft × $^1/_4$ in., Apeizon L (25% w/w) on chromosorb W (nonacid washed) DMCS treated, temperature programming 50–230°, 15°/min, and held at 230°, retention times for 2-, 3-, and 4-methoxybiphenyls and biphenyl 17, 19, 20, and 15 min, respectively; phenylation of nitrobenzene—column 6 ft × $^3/_{16}$ in., QF 1 (20% w/w) on Chromosorb W (nonacid washed) at 180°, retention times for 2-, 3-, and 4-nitrobiphenyls 8.5, 12.5, and 14 min, respectively (biphenyl was estimated on the Apiezon L column); phenylation of benzonitrile—the QF 1 column, retention times for 2-, 3-, and 4-cyanobiphenyls and biphenyl 14, 20, 21.5, and 6.5 min, respectively; phenylation of bromobenzene—the Apiezon L column, retention times for 2-, 3-, and 4-bromobiphenyls and biphenyl 24, 36, 38, and 12 min, respectively; phenylation of naphthalene—column 10 ft × $^1/_4$ in., SE-30 (10% w/w) on Chromosorb W, DMCS treated, temperature programming 50–280°, 15°/min, and held at 280°, retention times for α- and β-phenylnaphthalene 17 and 19 min. respectively (biphenyl was estimated on the Apiezon L column),

Results and Discussion

The electrolytic reduction of benzenediazonium tetrafluoroborate in the presence of aromatic substrates gives rise to nuclear phenylation, and a mixture of the isomeric biaryls was obtained in each case. The isomer ratios are listed in Table I along with the isomer ratios for decomposition of benzoyl peroxide and N-nitrosoacetanilide. In Table II are listed the total rate ratios and partial rate factors for the same reactions. Clearly the results indicate that phenylation has occurred by a process nearly equivalent to the two well-known homolytic substitution processes using benzoyl peroxide or nitrosoacetanilide as sources of free radicals. There can be no suggestion of an electrophilic ionic process from these results. High yields of benzene, ~50% of the total yield, can be explained by hydrogen abstraction from acetonitile. Phenylation of toluene by benzoyl peroxide gave 13% bibenzyl and electrolytic reduction of the diazonium salt gave 12%.

The generation of aryl radicals by electrochemical means obviates the problem of the possibility of other radicals such as hydroxy, benzoyloxy, and acyloxy radicals being produced by homolysis of chemical bonds. The similarity of the electrochemical results to those of the other reactions supports mechanisms such as that of Rüchardt and Freudenberg[13] in which it is considered that no acyloxy radicals are produced in the decomposition of nitrosoacetanilide.

The suggestion by Inukai, Kobayashi, and Simamura[14] that the high total rate ratio and partial rate

(8) R. M. Elofson, *Can. J. Chem.*, **36**, 1207 (1958).

(9) R. M. Elofson and F. F. Gadallah, *J. Org. Chem.*, in press.

(10) J. J. Lingane and S. L. Jones in "Electroanalytical Chemistry," Interscience Publishers, New York, N. Y., 1953, p 349.

(11) Tetrabutylammonium perchlorate rather than the chloride or bromide salts was used as the electrolytic support because the latter react with mercury and the diazonium salt.

(12) Acetonitrile was preferred over sulfolane as solvent because of its low melting point and the ease with which it could be removed from the reaction mixture by distillation. Also the primary hydrogen in acetonitrile would be much less readily abstracted by phenyl radicals than the secondary hydrogen of sulfolane [R. F. Bridger and C. A. Russell, *J. Amer. Chem. Soc.*, **85**, 3754 (1963)].

(13) C. Rüchardt and B. Freudenberg, *Tetrahedron Lett.*, 3623 (1964).

(14) T. Inukai, K. Kobayashi, and O. Simamura, *Bull. Chem. Soc. Jap.*, **35**, 1576 (1962).

TABLE I

ISOMER RATIOS AND YIELDS OF PHENYLATION PRODUCTS BY ELECTROLYTIC REDUCTION AS COMPARED WITH THOSE OBTAINED FROM PHENYLATION WITH BENZOYL PEROXIDE AND N-NITROSOACETANILIDE

		Electrolytic redn			(PhCOO-)$_2$				PhN(NO)COCH$_3$			
Substrate	Isomer	°C	Ratio	Yield, %	°C	Ratio	Yield, %	Ref	°C	Ratio	Yield, %	Ref
C_6H_6		25		15								
	ortho		54			66.5				65.6		
$C_6H_5CH_3$	*meta*	0	25.8	19	80	19.3		4	20	21.7		4
	para		20.2			14.2				12.6		
	ortho		66.6			69.8				69.3		
$C_6H_5OCH_3$	*meta*	0	17.0	29	80	14.7	50	4	20	18.1		4
	para		16.4			15.6				12.6		
	ortho		63.7			60						
C_6H_5CN	*meta*	−5	9.7	33	80	10		4				
	para		26.6			30						
	ortho		58.8			63.2				62.2		
$C_6H_5NO_2$	*meta*	−5	16.9	14	80	9.7	19	4	20	7.7		4
	para		24.3			27.1				30.1		
	ortho		59			56.2				60		
C_6H_5Br	*meta*	−5	25	33	80	27.3	33	4		22	90	*a*
	para		16			16.5				18		
$C_{10}H_8$	*α*	0	78.5	32		82		3	25	84.3		*b*
	β		21.5			18				15.7		

[a] O. Simamura, T. Inukai, and M. Kurara, *Bull. Chem. Soc. Jap.*, **25**, 76 (1952). [b] S. Dickerman and G. B. Vermont, *J. Amer. Chem. Soc.*, **84**, 4150 (1962).

TABLE II

TOTAL RATE RATIOS AND PARTIAL RATE FACTORS FOR PHENYLATION OF AROMATIC SUBSTRATES USING DIFFERENT PHENYL RADICAL SOURCES

		Electrolytic redn		(PhCOO-)$_2$			PhN(NO)COCH$_3$		
Substrate	Isomer	$\sum_H^X k$	F_r	$\sum_H^X k$	F_r	Ref	$\sum_H^X k$	F_r	Ref
	ortho		2.87		2.5			3.3	
$C_6H_5CH_3$	*meta*	1.77	1.37	1.23	0.7	4	1.68	1.09	4
	para		2.14		1.0			1.27	
	ortho		3.9		4.17			3.56	
$C_6H_5OCH_3$	*meta*	1.96	1.0	2.0	0.88	*a*	1.71	0.93	4
	para		1.92		1.86			1.29	
	ortho		6.1		6.5				
C_6H_5CN	*meta*	3.1	0.93	3.7	1.1	4			
	para		51		6.5				
	ortho		5.2		5.5			9.38	
$C_6H_5NO_2$	*meta*	2.7	1.08	2.9	0.86	4	5.02	1.16	4
	para		4.75		4.9			9.05	
	ortho		2.94		1.9			4.6	
C_6H_5Br	*meta*	1.75	1.42	1.29	1.9	4	2.6	1.7	*b*
	para		1.85		1.3			2.8	
$C_{10}H_8$	*α*	16.6	19.5	10	16	3	16.5	20.8	*c*
	β		5.35		3.5			3.88	

[a] R. T. Morrison, J. Cazes, N. Samkoff, and C. A. Howe, *J. Amer. Chem. Soc.*, **84**, 4152 (1962). [b] Footnote *a*, Table I. [c] Footnote *b*, Table I.

factors obtained for the phenylation of nitrobenzene with nitrosoacetanilide is a temperature effect is not supported by the present results. At 0° the ratio of isomers and total rate ratio obtained by phenylation of nitrobenzene by electrochemically produced phenyl radicals was very similar to that for phenylation at 80° with benzoyl peroxide. On the other hand somewhat higher total rate ratios were obtained at 0° for toluene, bromobenzene, and naphthalene (1.77, 1.75 and 16.6) than at 80° (1.23, 1.29, and 10). The results for these three compounds approximated fairly well the results at 20, 20, and 25° of 1.68, 2.6, and 16.5, respectively. Certainly, if there is a temperature effect on the total rate ratio and partial rate factors, it must be quite small and variable from one substrate to another. In principle, electrochemical reductions can be carried out at temperatures as low as −35° (mercury freezes at −39°) and a more definitive answer might be obtained at the lower temperature.

In the electrochemical arylations no evidence for tars was obtained. Except for the reaction with nitrobenzene which produced some darkening, all of the reactions remained light in color. Evidence for side reactions involving the nitro group is apparent from the lowered yield of nitrobiphenyls despite the higher activity of nitrobenzene compared with that of benzene in competitive reactions.

Clearly a new and uncomplicated method of arylating aromatic substrates has been evolved. Much remains to be done. For example, in view of the increased yields obtained by carrying out phenylation reactions with benzoyl peroxide at 80° in the presence of oxygen,[15]

(15) R. T. Morrison, J. Cazes, N. Samkoff, and C. A. Howe, *J. Amer. Chem. Soc.*, **84**, 4152 (1962).

the electrochemical reactions should be investigated in the presence of oxygen. Arylations with substituted phenyl radicals can be carried out since the diazonium cations are reduced at more positive potentials than those of any substituents.[9]

Registry No.—Benzenediazonium tetrafluoroborate, 369-57-3; benzene, 71-43-2; toluene, 108-88-3; anisole, 100-66-3; benzonitrile, 100-47-0; nitrobenzene, 98-95-3; bromobenzene, 108-86-1; naphthalene, 91-20-3.

Acid-Catalyzed Isomerization of Dialkylbenzenes

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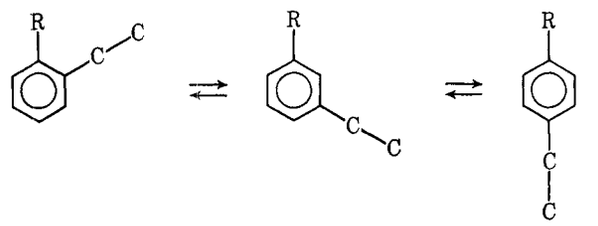
Isomerization and transalkylation are the principal reactions of 1-methyl-2-ethylbenzene over silica–alumina cracking catalyst and partially multivalent cation-exchanged, partially decationized Y-type faujasite molecular sieves. Isomerization proceeds by two different mechanisms. The first type consists of intramolecular 1,2 shifts. Transalkylated intermediates are involved in the second reaction. Xylenes and diethylbenzenes may be formed from methylethylbenzene by the same type of transalkylated intermediates. We suggest the name "alkyl exchange" for this reaction. The extent of intermolecular isomerization may be estimated from the level of alkyl exchange. The contribution of intermolecular isomerization decreases with increasing temperature. Above 300° isomerization primarily occurs by intramolecular 1,2 shifts. Below 200° isomerization *via* transalkylated intermediates predominates. At intermediate temperatures intermolecular and intramolecular processes contribute to over-all isomerization to a similar extent.

Two different mechanisms have been proposed for the acid-catalyzed isomerization of dialkylbenzenes. Carbonium ion involvement in both isomerization mechanisms is commonly assumed. According to Olah and his coworkers[1] and Matsumoto and Morita,[2] isomerization of dialkylbenzenes proceeds through a series of 1,2 shifts as shown below. On the other hand, Bolton



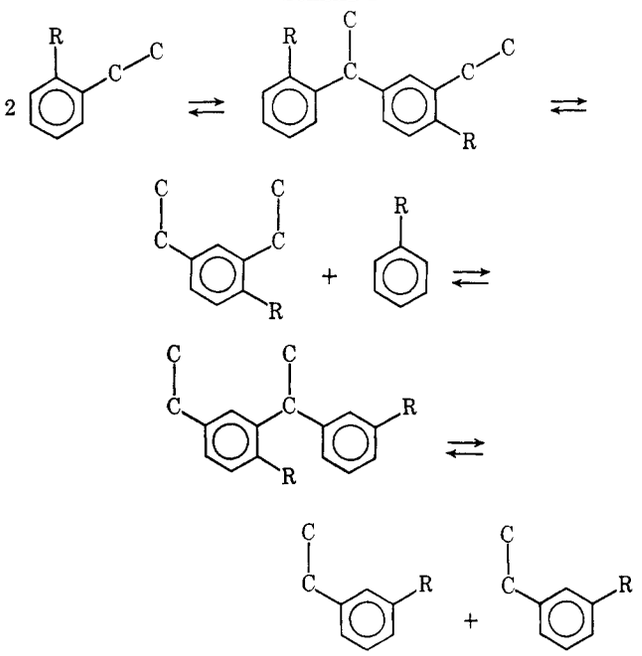
and his coworkers[3] found that at 170° over partially decationized and partially cerium-exchanged Y-type faujasite isomerization proceeds *via* a transalkylation mechanism shown in Scheme I. This reaction has real merit only when easily abstractable α-hydrogen atoms are available.

Simultaneous intramolecular and intermolecular processes were suggested by Allen and coworkers[4,5] and Ünseren and Wolf.[6] It is the intention of this work to resolve the apparent contradiction which involves the isomerization of dialkylbenzenes.

Experimental Section

1-Methyl-2-ethylbenzene was obtained from Columbia Organic Chemicals Co., Columbia, S. C. It was dried over sodium wire and purified chromatographically through activated silica gel under nitrogen. The purified material contained 99.77% 1-methyl-2-ethylbenzene, 0.15% 1-methyl-3- and -4-ethylbenzenes, and 0.08% xylenes and other hydrocarbons. The sulfur content was 0.3 ppm.

SCHEME I



The silica–alumina was a commercial cracking catalyst containing 10 wt % alumina, 90 wt % silica, and 0.1 wt % Na_2O. The B.E.T. surface area of the catalyst was 340 m^2/g. The catalyst was pretreated at 530° for 2 hr in helium and 1 hr in hydrogen before the experiments.

The calcium–ammonium Y-type faujasite was prepared the following way. Sodium Y-type faujasite (SK-40), prepared by the Linde Division of Union Carbide Corp., was first exchanged with $AgNO_3$. Silver was removed from the Ag sieve by NH_4SCN treatment. The ammonium Y-type sieve thus produced contained 6.75 wt % ammonium ions (Dumas N analysis). The sodium content was less than 0.01% (emission spectroscopy and neutron activation analyses). This ammonium sieve was treated with a solution of $Ca(NO_3)_2$, first at 100° for 3 hr, then at room temperature for 16 hr. Washed and dried at 100° *in vacuo*, the product Ca–NH_4^+ Y-sieve contained 1.47% ammonium ions (Dumas N analysis). The Ca/NH_4^+ equivalent ratio was 78:22. The catalyst was activated at 540° for 16 hr in dry hydrogen before the experiments.

The experiments were carried out in a continuous flow type reactor, consisting of a 50-cm-long, and 0.635-cm-o.d. stainless steel tube. The tube held 0.5 ml of catalyst. Experimental

(1) G. A. Olah, M. W. Meyer, and N. A. Overchuk, *J. Org. Chem.*, **29**, 2313 (1964).
(2) H. Matsumoto and Y. Morita, *Kogyo Kagaku Zasshi*, **70**, 1674 (1967).
(3) A. P. Bolton, M. A. Lanewala, and P. E. Pickert, *J. Org. Chem.*, **33**, 1513, 3415 (1968).
(4) R. H. Allen, L. D. Yats, and D. S. Erley, *J. Amer. Chem. Soc.*, **82**, 4853 (1960).
(5) R. H. Allen, *ibid.*, **82**, 4856 (1960).
(6) E. Ünseren and A. P. Wolf, *J. Org. Chem.*, **27**, 1509 (1962).