oxide gels have been found to promote the acylation of thiophene and furan.

Yields of 2-acetylthiophene as high as 87% are reported when a montmorillonite clay was employed with two moles of thiophene and one mole of acetic anhydride. Similarly, synthetic silica-alumina gel catalysts have given yields as high as 84%. 2-Benzoylthiophene has been prepared in yields of 94% from the action of a montmorillonite clay on benzoyl chloride and thiophene. 2-Acetylfuran has been obtained in 24% yields from acetic anhydride and furan. Due to the lower yields, no extensive study of the acylation of furan was made.

In general, the acid anhydrides are preferred to the acyl halides. With thiophene higher yields are obtained with the anhydrides. With furan no acylation has been noted with acyl halides.

3,3-bis-(2-Thienyl)-phthalide has been prepared from phthalyl chloride and thiophene in 15% yields. Adipyl chloride and thiophene yielded 20% of 5-(2-thenoyl)-pentanoic acid.

PAULSBORO, N. J. RECEIVED AUGUST 10, 1946

[CONTRIBUTION FROM THE METCALF RESEARCH LABORATORY OF BROWN UNIVERSITY]

Properties of Electrolytic Solutions. XXVII. The Conductance of Several Quaternary Ammonium Salts in Ethylene Chloride at 25°[1]

BY WALTER E. THOMPSON[2] AND CHARLES A. KRAUS

I. Introduction

The results of conductance measurements with a considerable number of electrolytes in ethylene chloride have formed the subject of several communications from this laboratory. The present paper extends our knowledge of the effect of the constitution and size of anions upon the electrolytic properties of their salts. In particular, the behavior of the following ions, in the form of their tetra-alkylammonium salts, has been investigated: chloride, bromide, thiocyanate, acetate, chloroacetate and borofluoride.

Fowler and Kraus[3] have developed a method of approximating limiting ion conductance values from conductance measurements alone. In view of the importance of ion conductances, it seemed desirable to obtain an independent check on Fowler's values. For this purpose, octadecyltrimethylammonium octadecylsulfate was employed. In composition and structure, the two ions are very similar and are of a type differing greatly from those used in Fowler's investigation. Assigning equal conductance values to the two ions of this salt, we have obtained limiting ion conductances in favorable agreement with those obtained by Fowler. Fowler's measurements with tetrabutylammonium triphenylborofluoride and Mead's[4] with tetrabutylammonium picrate were repeated and their results confirmed.

The effect of small additions of a highly polar substance, such as methyl alcohol, on the electrolytic properties of three quaternary ammonium salts has been studied.

II. Experimental

Apparatus and Procedure.—The methods employed in the purification of the solvent, in the preparation of the solutions and in the measurement of the resistances have been described fully in previous communications. Densities of solutions containing polar molecules were determined with a Westphal balance, while viscosity measurements were carried out in accordance with standard procedure. All measurements were carried out at 25 ± 0.01°.

Materials.—Pure methanol was obtained in the usual manner by treatment with calcium oxide. Although its specific conductance was approximately 3×10^{-6}, the values for the mixtures with ethylene chloride were less than 0.1% of the total conductance of the most dilute solutions.

Tetramethylammonium picrate which had been used in previous conductance measurements was available.

Tetraethylammonium chloride was recrystallized from ethylene chloride and the bromide from mixtures of acetone and alcohol; both decomposed without melting.

Tetraethylammonium borofluoride was prepared by Mr. G. W. Moessen, as described by Witschonke[5]; m. p. 235°. Methyltri-*n*-butylammonium thiocyanate was provided by Dr. L. E. Strong[6]; m. p. 101°.

Tetra-*n*-butylammonium acetate was prepared by neutralizing a solution of the hydroxide with acetic acid. This salt, like the corresponding chloroacetate, is prepared with great difficulty because of its great hygroscopicity. After evaporation of the solution on a steam-bath, the product was dried for two weeks over phosphorus pentoxide at pressures below 0.001 mm. It was then recrystallized from *n*-butyl chloride from which the last traces of alcohol had been removed by successive distillations from calcium chloride and phosphorus pentoxide. The salt was dissolved at 60° and about 15% crystallized out on cooling to the temperature of an ice-salt mixture. Additional crystals of higher purity were obtained by adding dry pentane to the cold solution. The last traces of butyl chloride were removed by means of a mercury vapor pump. An accurate melting point of this salt was difficult to obtain because of its hygroscopic properties; m. p. 118 ± 2°.

Tetra-*n*-butylammonium chloroacetate was prepared like the acetate by interaction of the base and acid in aqueous solution. This salt is even more hygroscopic than the acetate and is more difficult to crystallize. The

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[1] This paper is based on a portion of a thesis presented by W. E. Thompson in partial fulfillment of the requirements for the degree of Doctor of Philosophy in the Graduate School of Brown University, May, 1941.

[2] University Fellow at Brown University, 1939–1940; Metcalf Fellow, 1940–1941.

[3] Fowler and Kraus, THIS JOURNAL, **62**, 2237 (1940).

[4] Mead, Fuoss and Kraus, *Trans. Faraday Soc.*, **32**, 594 (1936).

[5] Charles R. Witschonke, Thesis, Brown University, 1941.

[6] Lawrence E. Strong, Thesis, Brown University, 1940.

salt as obtained from its aqueous solution was melted under high vacuum. Because of its hygroscopic properties, satisfactory melting points could not be obtained. Conductance measurements were reproducible to only 0.5%.

Tetra-*n*-butylammonium thiocyanate was furnished by Dr. Strong[6]; m. p. 124°. A pure sample of the picrate was on hand; m. p. 89.5°.

Octadecyltrimethylammonium octadecylsulfate was prepared by metathesis of octadecyltrimethylammonium nitrate with potassium octadecylsulfate in pure *i*-propyl alcohol. The salt was recrystallized from this solvent; m. p. 147°. The octadecyltrimethylammonium nitrate had been prepared from the iodide by metathesis with silver nitrate in alcohol solution. It was recrystallized from acetone; m. p. 210°. *n*-Octadecyltrimethylammonium iodide was prepared by heating *n*-octadecyl iodide with trimethylamine in an alcohol-water mixture. The reaction was carried out in a sealed tube at 60° over a period of one week. The salt was recrystallized from hexane containing about 5% of alcohol; m. p. 234.5–236°. Analysis gave iodine, 28.89%; calculated, 28.82%.

Potassium octadecyl sulfate was obtained by a method employed by Lottermoser and Stohl[7] in preparing similar salts: 250 g. of octadecyl alcohol was heated to 70° and 75 g. of concentrated sulfuric acid slowly added with vigorous stirring. The stirring was continued until a sample of the mixture yielded a clear solution in water. The mixture was then poured onto ice and the octadecylsulfuric acid extracted with ether. Upon neutralizing the ether solution with 1 *N* potassium hydroxide, the potassium salt was precipitated. The salt was recrystallized from alcohol; m. p. 178° (discolors at 145°).

III. Results

A. Solutions in Pure Ethylene Chloride.—In Table I are given the experimental values for the equivalent conductance obtained for the salts in ethylene chloride solution. Although (excepting the chloroacetate, see above) two independent series of values agreeing within 0.1% were actually obtained, only one series is given here. Concentrations are expressed in moles per liter of solvent, the density of ethylene chloride having been taken as 1.2455. In the computation of constants, the dielectric constant has been taken as 10.23 and the viscosity as 0.00785.

B. Effects Due to Added Polar Substances.—Polar molecules of a third component, when added to a solution of an electrolyte in a given solvent, may have a marked effect an the ion-pair equilibrium, particularly if the added polar molecules are small and the solvent molecules are large. In some instances, as will be reported in a later paper, the added molecules may have a marked effect on ion-conductance. Some results obtained in the present investigation indicate effects of this kind, although they are comparatively small.

Conductance measurements were made with solutions of tetramethylammonium picrate at fixed concentrations to which were added varying amounts of methyl alcohol, tertiary butyl alcohol and triphenylcarbinol. Solutions of tetrabutylammonium acetate and tetrabutylammonium picrate, to which were added varying amounts of methyl alcohol, were likewise studied. Numerical values are not presented since the results are shown adequately and more clearly in graphical form (Fig. 1).

Measurements were carried out to low concentrations with solutions of tetramethylammonium and tetrabutylammonium picrates and tetrabutylammonium acetate, in which methyl alcohol in small, fixed amount was added to the ethylene chloride for a series of dilutions. The results are given in Table II. At the head of each series is given

TABLE I

CONDUCTANCE OF VARIOUS ELECTROLYTES IN ETHYLENE CHLORIDE AT 25°

$C \times 10^5$	Λ
Tetraethylammonium bromide	
148.4	18.16
41.56	27.82
13.11	39.35
5.197	49.35
2.912	55.07
1.392	61.44
0.8490	64.82
Tetra-*n*-butylammonium acetate	
55.47	23.68
16.77	32.32
5.505	40.46
1.647	47.50
0.7884	50.56
Tetra-*n*-butylammonium thiocyanate	
6.329	51.61
3.258	56.90
1.487	61.68
0.7815	64.56
Methyltri-*n*-butylammonium thiocyanate	
12.94	38.23
5.311	47.88
2.894	54.01
0.7040	64.65

$C \times 10^5$	Λ
Tetraethylammonium chloride	
72.26	21.75
27.27	30.41
15.13	35.66
4.363	51.11
2.283	58.46
0.9590	66.42
Tetraethylammonium borofluoride	
45.36	33.84
17.14	44.77
7.345	54.82
3.345	63.97
1.184	71.98
Tetra-*n*-butylammonium chloroacetate	
15.24	34.34
4.702	41.44
1.935	45.24
1.232	47.00
Octadecyltrimethylammonium octadecylsulfate	
19.48	10.73
10.71	13.56
4.984	18.08
3.163	21.12
1.979	24.44
Pyridonium Nitrate	
65.0	2.5
Phenyldimethylhydroxyammonium picrate	
523	0.525

TABLE II

CONDUCTANCES OF SALTS IN ETHYLENE CHLORIDE ON ADDITION OF METHYL ALCOHOL

Tetramethylammonium picrate		Tetra-*n*-butylammonium picrate	
$C(\mathrm{CH_3OH}) = 0.204$; $d = 1.2419$; $\eta = 0.007724$		$C(\mathrm{CH_3OH}) = 0.200$; $d = 1.2420$; $\eta = 0.007727$	
$C \times 10^5$	Λ	$C \times 10^5$	Λ
30.15	27.62	28.45	37.30
12.98	37.43	9.305	45.50
5.640	46.82	4.394	49.90
2.142	58.80	2.467	52.49
1.066	66.30		

Tetra-*n*-butylammonium acetate

$C(\mathrm{CH_3OH}) = 0.200$; $d = 1.2420$; $\eta = 0.007727$

$C \times 10^5$	Λ
95.83	24.28
30.70	32.66
13.11	39.10
5.387	45.23
2.071	49.97

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(7) Lottermoser and Stohl, *Kolloid-Z.*, **63**, 49 (1933).

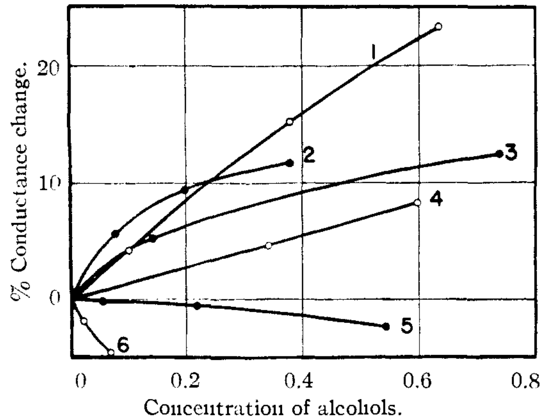

Fig. 1.—Change in conductance of solutions of salts in ethylene chloride on addition of alcohols at 25°.

Curve	Salt	Alcohol added	C salt × 10^4
1	Tetramethylammonium picrate	Methyl alcohol	0.54
2	Tetrabutylammonium acetate	Methyl alcohol	2.0
3	Tetrabutylammonium acetate	Methyl alcohol	1.2
4	Tetrabutylammonium picrate	Methyl alcohol	1.7
5	Tetramethylammonium picrate	Trimethylcarbinol	0.85
6	Tetramethylammonium picrate	Triphenylcarbinol	0.82

the concentration of alcohol as are also the density and the viscosity of the solvent mixture. The dielectric constant of various dilute mixtures of ethylene chloride and methyl alcohol at 25° are given in Table III. The measurements are due to Mr. W. J. Jacober.[8]

TABLE III

DIELECTRIC CONSTANTS OF DILUTE MIXTURES OF ETHYLENE CHLORIDE AND METHYL ALCOHOL AT 25°

% CH_3OH	Diel. const.
0.00	10.235
.27	10.279
.74	10.356
1.14	10.422
1.73	10.523

IV. Discussion

A. Conductance of Solutions in Pure Ethylene Chloride.—The conductance data have been treated according to the method of Fuoss[9] in order to determine limiting conductances and dissociation constants of the salts under investigation. Large scale plots were made of F/Λ against $C\Lambda f^2/F$. All plots were found to be linear up to the critical concentration (3.4×10^{-4}) within the limit of experimental error (0.1%). In view of the uniform linearity of the plots, they are not reproduced but the two typical constants Λ_0 and K are collected in Table IV.

TABLE IV

CONSTANTS OF SEVERAL QUATERNARY AMMONIUM SALTS IN ETHYLENE CHLORIDE

Salt	Λ_0	$K \times 10^4$
$(C_2H_5)_4NCl$	77.4	0.510
$(C_2H_5)_4NBr$	72.1	0.697
$(C_2H_5)_4NBF_4$	81.0	1.05
$(CH_3)(n\text{-}C_4H_9)_3NSCN$	71.2	0.663
$(n\text{-}C_4H_9)_4NOOC{\cdot}CH_3$	53.5	1.34
$(n\text{-}C_4H_9)_4NOOC{\cdot}CH_2Cl$	50.5	1.94
$(n\text{-}C_4H_9)_4NSCN$	68.6	1.40
$(C_{18}H_{37})(CH_3)_3N(C_{18}H_{37}SO_4)$	45.2	0.118

Fowler and Kraus[3] have shown that the limiting conductance of the tetra-*n*-butylammonium ion is approximately 26.2. Using this value, we have calculated the limiting conductances of the acetate, chloroacetate and thiocyanate ions from the Λ_0 values of the corresponding tetrabutylammonium salts. The conductance of the tetraethylammonium ion has been shown to be 38.2[10] which leads to values for the chloride, bromide and borofluoride ions from the Λ_0 values of the corresponding tetraethylammonium salts. Gleysteen[11] has obtained a value of 29.1 for the limiting conductance of the methyltri-*n*-butylammonium ion. The Λ_0 value for the thiocyanate of this ion is 71.2 which gives 42.1 as the conductance of the thiocyanate ion, in fair agreement with the value (42.4) obtained from the tetra-*n*-butylammonium salt. Values of the limiting anion conductances are given in Table V and it is to be noted that they are all based on the assumption of Fowler and Kraus, that the conductance of the tetra-*n*-butylammonium ion is equal to that of the triphenylborofluoride ion.

At this point, it is appropriate to consider whether the results obtained for octadecyltrimethylammonium octadecylsulfate lead to ion conductances in agreement with those based on Fowler's data. The structures of the two ions of this salt are similar in that each contains an octadecyl chain while the group of atoms to which each octadecyl chain is attached cannot differ greatly in size. Assigning equal conductances to these ions, we arrive at an ion conductance of 22.6 for each ion. Weaver[12] has obtained a value of 63.7 for the limiting conductance of octadecyltrimethylammonium nitrate. Using Tucker's value of 40.1[10] for the conductance of the nitrate ion, we find a value of 23.6 for the octadecyltrimethylammonium ion. This is one unit greater than that obtained by halving the conductance of octadecyltrimethylammonium octadecylsulfate. On the basis of what is known concerning the mobility of positive and negative ions, it seems reasonable that the conductance of the octadecylsulfate ion should be somewhat lower than that of the octadecyltrimethylammonium ion. In any case, ion conductance values derived by Fowler's

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[8] Unpublished observations in This Laboratory by Mr. W. J. Jacober.

[9] Fuoss, THIS JOURNAL, **57**, 488 (1935).

[10] Tucker and Kraus, THIS JOURNAL, **69**, 454 (1947).

[11] Gleysteen and Kraus, *ibid.*, **69**, 451 (1947).

[12] Harold E. Weaver, Thesis, Brown University, 1940.

method are not greatly in error. The low value of the dissociation constant of the octadecylsulfate salt is disadvantageous in its use as a standard.

Until further investigations have been made, we shall retain Fowler's value of 26.2 for the conductance of the tetrabutylammonium ion as a basis for calculating the conductance of other ions. His measurements with tetrabutylammonium triphenylborofluoride have been confirmed in the present investigation as have been, also, those of Mead[4] for tetrabutylammonium picrate.

For convenience, known conductances of negative ions are collected in Table V. The ions are arranged in order of decreasing conductance.

TABLE V

ANION CONDUCTANCES IN ETHYLENE CHLORIDE

Ion	Λ_0^-	Ion	Λ_0^-
BF_4^-	42.7	Pi^-	31.2[c]
SCN^-	42.4	CH_3COO^-	27.3
NO_3^-	40.1[a]	$(C_6H_5)_3BF^-$	26.2[d]
Cl^-	39.1	$(C_6H_5)_3BOH^-$	26.1[d]
ClO_4^-	39.2[b]	$ClCH_2COO^-$	24.3
Br^-	33.8	$OctdSO_4^-$	21.6

[a] Ref. (10). [b] Ref. (11). [c] Refs. (10) and (4). [d] Ref. (3).

It is apparent that the conductance of even the simpler ions varies over a wide range. Strong interaction must occur between certain small ions and the dipole molecules of the solvent. Thus the conductance of the acetate ion 27.5 is exceptionally low in view of the fact that it contains only four atoms other than hydrogen. The chloroacetate ion, however, has an even lower conductance, approaching that of ions containing the octadecyl group. This effect of chlorine in reducing the mobility of the acetate ion is noteworthy; the phenomenon requires further study. Compared to the acetate ion, the conductance of the picrate ion with sixteen atoms, other than hydrogen, is rather high, 31.2 as against 27.5.

It is of interest to note that the highest conductance for a negative ion, BF_4^-, is 42.7, while the highest value for a positive ion, $(CH_3)_4N^+$, is 42.6. But the tetramethylammonium ion has a lower conductance than might be expected on the basis of the conductance of higher members of the tetra-alkylammonium series as Tucker[10] has shown. It seems to be a general rule that ions of smaller atomic dimensions are relatively (and often, absolutely) poorer conductors than ions of larger dimensions. The interaction of ions with the polar molecules of the solvent medium requires further investigation, theoretically as well as experimentally.

An examination of the dissociation constants in Table IV, as well as those of earlier investigations, indicates that for salts of simpler quaternary onium ions, the constant varies but little so long as interaction involves only coulombic forces. Salts with smaller ionic nuclei yield only slightly weaker salts. As the structure of the cation becomes more open, specific steric effects come into play as is the case with the sulfonium and iodonium ions.[13]

The dissociation constant of octadecyltrimethylammonium octadecylsulfate (0.118×10^{-4}) may seem low at first glance. This value, however, is but little below what one should expect for an ammonium ion containing three methyl groups. Corresponding salts containing three butyl substituents have normal dissociation constants as will be shown in a later paper.[12] Tetrabutylammonium acetate and chloroacetate have normal dissociation constants. The latter salt is slightly the stronger but the chlorine atom in the negative ion has only a slight effect on the dissociation constant. The effect is exceedingly small when compared with that in aqueous solutions of acids.

B. Influence of Some Alcohols on the Conductance of Salts in Ethylene Chloride.—We shall consider, first, the conductance change on adding methyl alcohol, *t*-butyl alcohol and triphenylcarbinol to solutions of several salts at concentrations between 2.0×10^{-4} and 0.5×10^{-4} molal. The results are presented graphically in Fig. 1. The conductance increase, expressed in per cent., is plotted as ordinate against concentration of alcohol, expressed in moles per liter of solution as abscissa. With tetramethylammonium picrate, the conductance increase on addition of methanol is almost linear and is quite large. For the corresponding tetrabutylammonium salt, the conductance change is still linear but much smaller than for tetramethylammonium picrate. On adding methyl alcohol to solutions of tetrabutylammonium acetate, the conductance increases sharply on initial additions; with larger additions, the curves flatten out. On addition of *t*-butyl alcohol, the conductance of tetramethylammonium picrate falls off slowly; on the addition of triphenylcarbinol, the conductance falls off sharply. For these alcohols, the conductance decrease is doubtless due to viscosity increase.

The experimental data of Table II have been analyzed by the method of Fuoss, and Λ_0 and K values have been computed for solutions of the salts there listed. The results are presented in Table VI, where the concentration of alcohol, C_a,

TABLE VI

EFFECT OF METHANOL ON THE CONSTANTS OF ELECTROLYTES IN ETHYLENE CHLORIDE

Salt	C_a	Λ_0	$K \times 10^4$	K_c/K_0	$(\Lambda_0\eta)c/(\Lambda_0\eta)_0$
$(CH_3)_4NPi$	0.000	73.80	0.353	1.26	
	.204	75.20	.457		1.002
$(C_4H_9)_4NPi$	.000	57.40	2.28	1.17	
	.200	58.43	2.67		1.002
$(C_4H_9)_4NOOCCH_3$	.000	53.50	1.34	1.48	
	.200	56.27	1.99		1.034

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(13) Kline and Kraus, THIS JOURNAL, **69**, 814 (1947).

is given in the second column, the value of Λ_0 in the third column and the dissociation constant, K, in the fourth column. In the fifth column is given the ratio K_c/K_0, where K_c is the constant in the presence of alcohol and K_0 that in the pure solvent. In the last column are given values of the ratio of the conductance–viscosity product ($\Lambda_0\eta$) for solutions with and without alcohol.

As may be seen from Table VI, Λ_0 increases for all salts on addition of alcohol. For the two picrates, this increase is clearly due to the decreased viscosity of the solvent, for $\Lambda_0\eta$, the product of conductance and viscosity, remains constant as shown in the last column of the table. In the case of tetrabutylammonium acetate, the product $\Lambda_0\eta$ increases as methanol is added. On addition of 0.2 N methanol, the conductance increases 5.24%, while the viscosity decreases only 1.57%. Here we can only infer that the alcohol interacts with the highly solvated acetate ion to yield an ion of effectively smaller dimensions.

The effect of 0.2 N methanol on the dissociation constant of the several salts is of interest. For tetramethylammonium picrate, the increase is 26%; for the corresponding tetrabutyl salt, it is 17%, while for tetrabutylammonium acetate, the increase is 48%. The increase in the dielectric constant of the solvent due to 0.2 N methanol is approximately 0.3%. On computing the increase in K, due to the increase in dielectric constant, the measured increases in K are from two to five times those computed. Seemingly, the methanol dipoles interact with the ions so as to increase the effective distance between charges in the ion-pairs. As was to have been expected, this interaction is greater with the small tetramethylammonium ion than with the much larger tetrabutylammonium ion. In the case of tetrabutylammonium acetate, marked interaction evidently takes place with the highly solvated acetate ion so as to render it an effectively larger ion in the ion-pair. This occurs even though the free ion is effectively smaller as a result of interaction with methanol molecules.

Pyridonium nitrate and phenyldimethylhydroxyammonium picrate are exceedingly weak electrolytes and their dissociation constants can be approximated only by assuming values of Λ_0 and computing K by means of Ostwald's Dilution Law. Assuming 80 and 20 for the Λ_0 values of the above two salts, respectively, we obtain the K values: 6×10^{-7} for the first salt and 3×10^{-7} for the second. The low constant for pyridonium nitrate may be due to interaction of the negative ion with the hydrogen atom of the ammonium ion but it is also possible that the salt may dissociate into free acid and base. This has been found to occur in solutions of similar salts in tricresylphosphate[14] and similar phenomena have been observed and studied in nitrobenzene solutions.[5] In the case of phenyldimethylhydroxyammonium picrate, there appears to be interaction between the negative ion and the hydrogen of the hydroxyl group of the positive ion.

V. Summary

1. The conductance of a number of salts having different anions has been measured in ethylene chloride at 25°.

2. Dissociation constants and Λ_0 values have been derived from the experimental data by the method of Fuoss.

3. Ion conductances have been evaluated by the method of Fowler. The borofluoride ion, BF_4^-, has the highest conductance and the octadecylsulfate ion the lowest. The acetate and the chloroacetate ions have low conductances.

4. The limiting conductance of octadecyltrimethylammonium octadecylsulfate has been determined. Assuming equal conductance of the two ions of this salt, a value of 22.6 is found for each ion. Adopting Fowler's value for the tetrabutylammonium ion, the values differ from the above by one unit, 21.6 for the negative ion and 23.6 for the positive ion, respectively.

The effect of alcohols on the conductance of salts in ethylene chloride has been investigated. The effect of methanol on the K and Λ_0 values of tetramethylammonium picrate, tetrabutylammonium picrate and tetrabutylammonium acetate has been determined. The value of K is markedly increased, particularly for the acetate. For the two picrates, Λ_0 increases directly as the fluidity of the solvent; for the acetate, the increase is greater.

6. The conductance of solutions of pyridonium nitrate and of phenyldimethylhydroxyammonium picrate has been measured; both salts are very weak electrolytes. There is evidence of interaction of the negative ion with the hydrogen of the phenyldimethylhydroxylammonium ion; in the case of the pyridonium salt, acid–base dissociation may occur.

PROVIDENCE, R. I. RECEIVED JANUARY 13, 1947

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(14) Elliott and Fuoss, THIS JOURNAL, **61**, 294 (1939).