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Theory of Electrical Conductance in Acidic, Aqueous Solutions

by

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THEORY OF ELECTRICAL CONDUCTANCE IN ACIDIC, AQUEOUS SOLUTIONS

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ABSTRACT

Protonic conduction occurs by the Grotthuss mechanism. Below about 0.001m HCl, the lengths of the Grotthuss chains are determined by the temperature, but over the range 0.001m to about 0.5m, the chains are principally disrupted by other protons, thus in this range the chain length and the equivalent conductance depend on the cube root of the concentration. An Arrhenius-type equation is developed for describing the concentration dependence of the conductance which yields values in agreement with experiment to within 1% over the range 5 to 65°C and up to 0.4m. The activation energy which appears in the chain-terminating terms of this equation is that of "hole" formation in the solvent water.

## INTRODUCTION

Elsewhere,<sup>1,2</sup> we have pointed out that the Onsager-Fuoss<sup>3</sup> and similar equations for the concentration dependence of the electrical conductivity of ionic solutions based on consideration of the electrophoretic and relaxation effects are not applicable to acidic, aqueous solutions or any other systems in which the Grotthuss mechanism makes a conductive contribution. The purpose of the present study is to indicate some of the directions which the derivation of a conductivity equation applicable to protonic conduction might take.

## THE NON-PROTONIC CONDUCTIVITY CONTRIBUTION

In this study we will be concerned with aqueous HCl solutions and with the protonic conductivity contribution, however, it might be interesting in passing to consider the applicability of conduction equations based on the electrophoretic and relaxation effects to the chloride ion contribution. Inasmuch as the motion of the proton is an apparent "motion," the cations<sup>4</sup> do not drag water with them and the chloride ions should see a nearly stationary medium. Because of the H-bonding properties of water, protons probably do not orient themselves in the same way in the coulombic field of anions as do other cations. Thus, one anticipates that even in the case of the anion in acidic solutions, the electrophoretic and relaxation effects, if occurring, will be profoundly altered.

Throughout this discussion, we will be concerned with only the Grotthuss component of protonic conduction. Although the Grotthuss mechanism dominates protonic conduction, protons also conduct by the "normal" process of hole formation and this latter process may account for as much as 20% of the total conductance.

## VISCOSITY

At one atmosphere rotation of water molecules is the rate-determining step of the Grotthuss mechanism.<sup>5,6</sup> Although its value is less, the activation energy of protonic conduction in aqueous solutions

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1. R. A. Horne and R. A. Courant, J. Chem. Soc. (London), in press, 1964.
  2. R. A. Horne, J. Chem. Phys., 40, 2064 (1964).
  3. L. Onsager and R. M. Fuoss, J. Phys. Chem., 36, 2689 (1932).
  4. Other chloride ions may result in solvent movement in the same direction. There is also the counterflow of medium to be considered.
  5. R. A. Horne, B. R. Myers, and G. R. Frysinger, J. Chem. Phys., 39, 2666 (1963).
  6. B. E. Conway, J. O'M. Bockris and H. Linton, J. Chem. Phys., 24, 834 (1956).

above 10°C varies in the same way with temperature as does that for the electrical conductivity of strong 1:1 salts such as KCl, thereby suggesting that energetically "hole" formation in water and rotation of water molecules are similar processes. The velocity of the translational movement for a sphere of radius,  $a$ , in a medium of viscosity  $\eta$  is given by Stokes' law:

$$(1) \quad v = F/6 \pi \eta a$$

where  $F$  is the applied force, while the angular velocity,  $w$ , of a sphere rotating in a viscous medium is given by

$$(2) \quad w = T/8 \pi \eta a^3$$

where  $T$  is the applied torque. Notice that the translational movement depends on the first power of the radius whereas the rotational movement depends on the third power. The application of hydrostatic pressure breaks up the order in liquid water, and reduces the effective radius of hydrated ions in aqueous solution.<sup>8,9</sup> Consequently, one expects the pressure dependence of the relative viscosity to be greater for HCl solutions than for KCl solutions. Figure 1 shows such to be the case. In fact, HCl shows a greater pressure dependence than anticipated. The explanation of this will be discussed later.

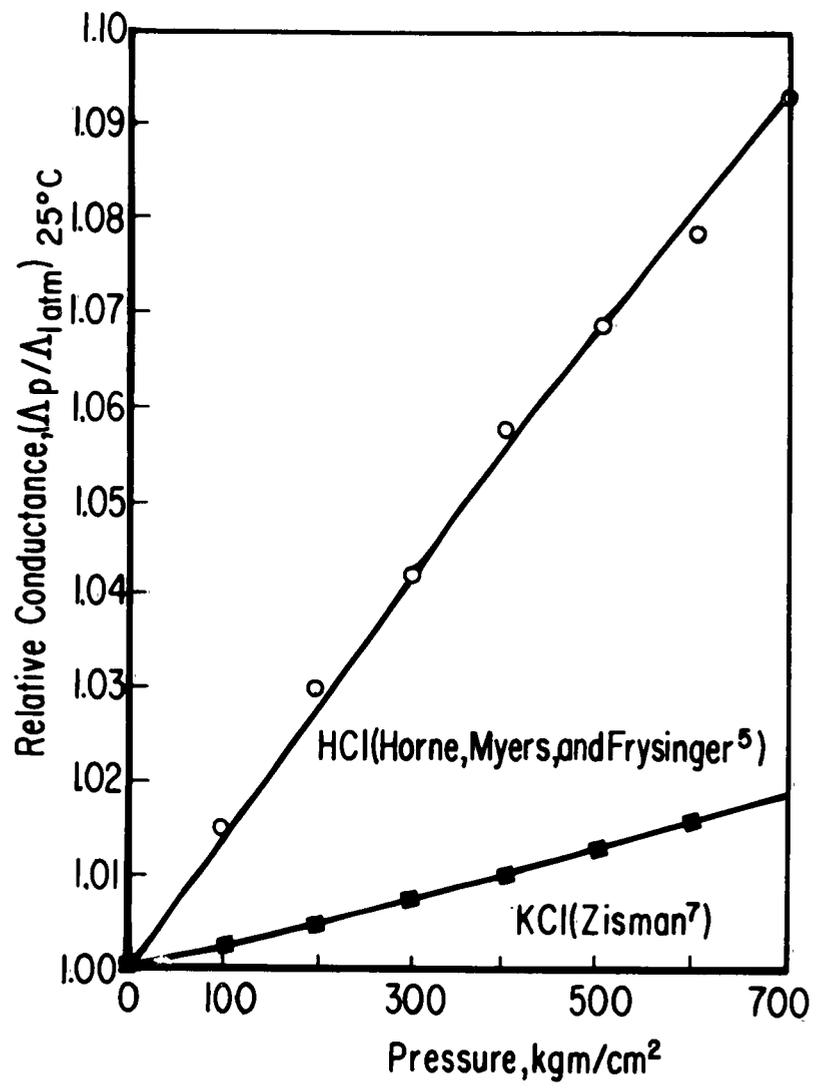
Both equations (1) and (2) contain the first power of the viscosity of the medium. Usually, the macroscopic viscosity is used, a questionable practice. Pressure, but not temperature, alters the sizes of hydrated ions (see above) and this may give rise to an apparent higher power dependence upon viscosity, but, as long as the solution is sufficiently dilute so that the ions do not have to compete for waters of solvation, changing the concentration of the electrolyte should not affect the sizes of the hydrated ions.

The dielectric relaxation time of water decreases with increasing electrolyte concentration.<sup>10,11</sup> If the speed of rotation of water molecules, the rate-determining step of the Grotthuss mechanism, behaves in like manner, then protonic conduction should increase with increasing concentration which is contrary to observation.

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7. W. A. Zisman, Phys. Rev., **39**, 151 (1932).
  8. R. A. Horne, Nature, **200**, 418 (1963).
  9. R. A. Horne and R. A. Courant, J. Geophys. Res., in press, 1964.
  10. D. M. Riston, J.B. Hasted, and C.H. Collie, Compt. Rend., **225**, 285 (1947).
  11. G. H. Haggis, J.B. Hasted, and T.J. Buchanan, J. Chem. Phys., **20**, 1452 (1952).

FIGURE 1

Pressure Dependence of the Relative Electrical  
Conductances of Aqueous Acid and Salt Solutions



The introduction of ions alters the extent of structure in liquid water and gives rise to changes in the macroscopic viscosity. The proton has a positive viscosity B-coefficient (see equation (3) of 0.070, indicating that it is a structure-forming ion.<sup>12</sup> Qualitatively then, one might expect it to order the solvent, hinder the rotation of water molecules, and thus impede Grotthuss conduction as observed. An obvious attack upon the present problem of deriving an expression for the concentration-dependence of protonic conductivity is to express the conductance as some function of the limiting conductance and the viscosity as given by the relation of Jones and Dole.<sup>13</sup>

$$(3) \eta = \eta^0 (1 + A \sqrt{C} + BC)$$

where  $\eta^0$  is the viscosity of pure water, C the concentration of electrolyte, and A and B are constants. Unfortunately, the effect of electrolyte concentration on viscosity is much too small to account for the observed effect on the equivalent conductance. Thus for 0.01M HCl the relative viscosity at 25°C is only 1.0008 while the reciprocal of the relative equivalent conductance  $(\Lambda/\Lambda^0)^{-1}$  is 1.03. In other words, the viscosity changes by only 0.08% whereas the conductance changes by more than 3%.

In summary, the approach of estimating the retardation of water rotation from structural changes in liquid water as reflected by the macroscopic viscosity does not appear to be an attractive one.

While the concentration of electrolyte has much less effect on the macroscopic viscosity than upon the conductance, the reverse is true in the case of temperature as illustrated by Table 1 which compares conductance ratios (based on International Critical Tables data) for HCl with the relative viscosity.

The above conclusion should not be taken to mean that conductivity, viscosity, and the degree of structure in liquid water are unrelated. On the contrary, Table 1 illustrates that they all have similar temperature dependencies.<sup>14</sup> This table compares the reciprocal of the limiting conductance relative to 0°C, the ratio of the limiting conductance  $\Lambda^0$ , to that,  $\Lambda^*$ , obtained by the extrapolation of  $\Lambda$  versus  $C^{1/3}$  to zero C; the relative viscosity  $\eta/\eta_{0^\circ\text{C}}$ ; the relative number of unbroken hydrogen bonds,  $H/H_{0^\circ\text{C}}$ ; and the relative size of the water clusters,  $S/S_{0^\circ\text{C}}$ .

The latter two parameters are calculated from the estimates of Nemethy and Scheraga.<sup>15</sup> This table indicates that the changes in the structural parameters are much larger than the changes in  $\Lambda^0/\Lambda^*$ . Notice, however, the good parallel between the relative viscosity and the relative cluster size.

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12. R. W. Gurney, Ionic Processes in Solution, McGraw-Hill Book Company New York, N.Y., 1953, p. 168.
  13. G. Jones and M. Dole, J. Am. Chem. Soc., **51**, 2950 (1929).
  14. See also Figure 3 in R.A. Horne and R. A. Courant, J. Phys. Chem., in press, 1964.
  15. G. Nemethy and H.A. Scheraga, J. Chem. Phys., **36**, 3382 (1962).

TABLE 1

Thermal Termination of Grotthuss Chains and Structural Parameters

$^{\circ}\text{C}$	$(\Lambda^{\circ}/\Lambda^{\circ}_{0^{\circ}\text{C}})^{-1}$	$\Lambda^{\circ}/\Lambda^*$	$\eta/\eta_{0^{\circ}\text{C}}$	$\text{H}/\text{H}_{0^{\circ}\text{C}}$	$\text{S}/\text{S}_{0^{\circ}\text{C}}$
0	1.00	-	1.00	1.00	1.00
18	0.84	-	0.65	0.88	0.66
25	0.63	0.99	0.58	0.86	0.57
35	0.54	0.99	0.50	0.82	0.47

GROTTHUSS CHAIN TERMINATION

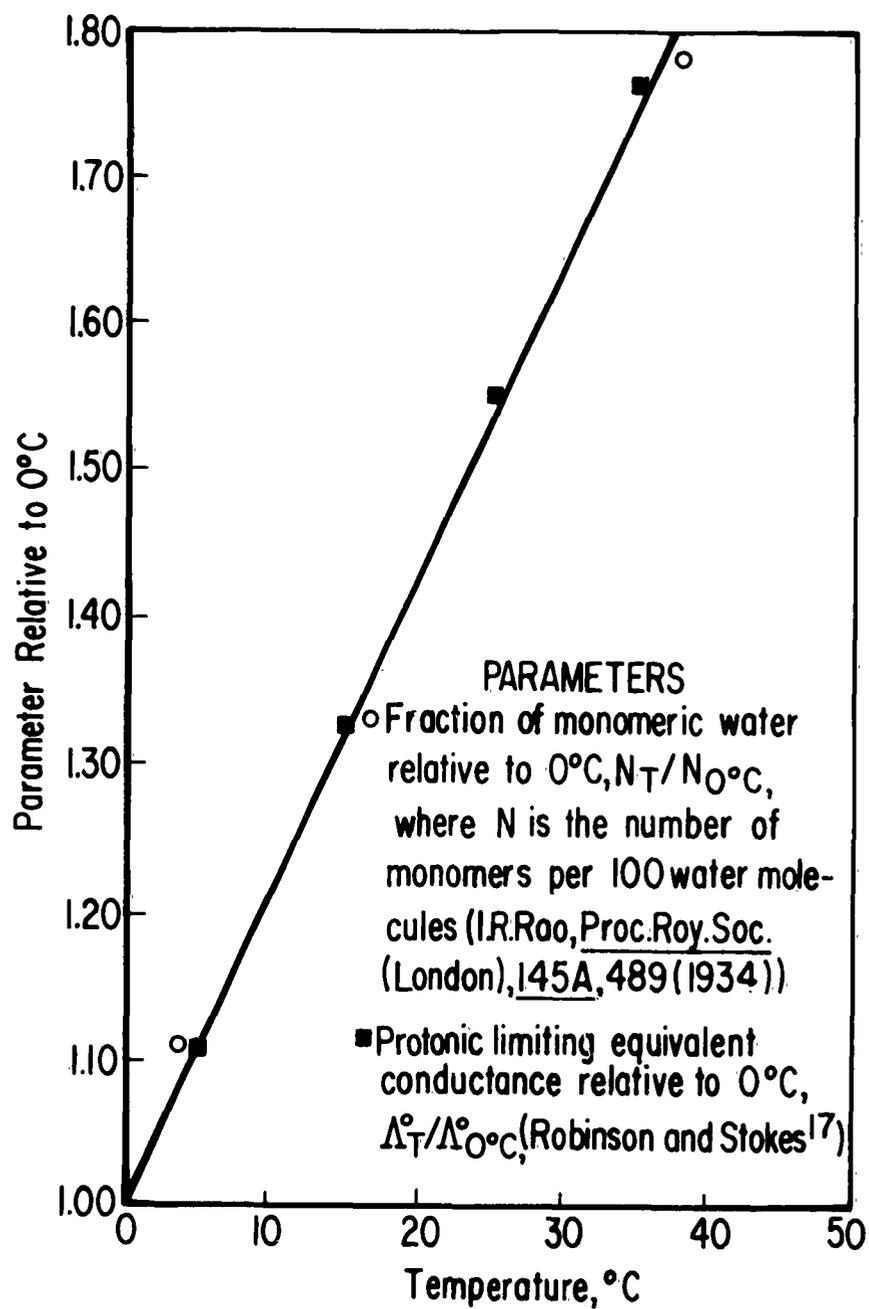
Electrical conduction by the Grotthuss mechanism occurs only in the interstitial or "free" water between the fluctuating clusters in liquid water.<sup>16</sup> This is not an unreasonable assumption inasmuch as (a) water molecules within the clusters are less free to rotate; (b) a cluster represents a closed Grotthuss propagative circuit; and (c) the only entity undergoing dipole orientation in water is the simple H<sub>2</sub>O molecule and not polymeric forms.<sup>17</sup>

If the protonic conductance involves only the "free" or monomeric water molecules, then the equivalent conductance should be proportional to the monomer concentration. Figure 2 shows such to be the case.

The proton flips propagate along transient chains of H-bonded water molecules in the "free" solvent. In pure water or in very dilute solutions, the length of these chains will depend for the most part on temperature; this is "normal" or thermal chain termination (TCT). The introduction of a second proton into a chain will be dependent on the concentration and as the concentration increases should become the dominant chain-terminating factor (CCT). Let the concentration of protons in a unit cube be C; let us assume for the sake of simplicity that a proton is "proceeding" along a chain along the edge of the unit cube,<sup>18</sup> then the distance which it can "move" without encountering a

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16. R. A. Horne and E. H. Axelrod, J. Chem. Phys. **40**, 1518 (1964).  
 17. R. A. Robinson and R. H. Stokes, Electrolyte Solutions, Butterworths Sci. Pub., London (2nd ed.) 1959, p. 12. Vide R.P. Marchi and H. Eyring, J. Phys. Chem., 68, 221 (1964).  
 18. When the electrical potential is applied, the direction of a proton's apparent movement ceases to be random.

**FIGURE 2**  
**The Relation Between Proton Conductance  
and Water-Monomer Concentration**



second proton will depend on  $C^{-1/3}$ . This distance is the "lattice length" discussed by Frank and Thompson,<sup>19</sup> and they note that "it makes no particular difference what kind of a lattice is assumed-- it is the  $C^{1/3}$  dependence that counts." Protonic conduction should depend on the cube root of the proton concentration.

Figure 3, based on the data of Owen and Sweeton,<sup>20</sup> exhibits the concentration-dependence of the equivalent conductance of aqueous HCl solutions at different temperatures, and, indeed  $\Lambda_{\text{HCl}}$  versus  $C^{1/3}$  is linear over a broad concentration range, roughly 0.001 to 0.5m. The departure from linearity above 0.5m is not surprising, for this represents a concentrated solution. Of greater significance are the departures below 0.001m.

If the linear segments of the curves in Figure 3 are extrapolated down to infinite dilution of electrolyte, the values obtained,  $\Lambda^*_{\text{T},0}$ , are greater than the limiting conductances,  $\Lambda^{\circ}$ , obtained by the usual methods ( $\lim_{C \rightarrow 0} \Lambda$  vs  $C^{-1/2}$ ).  $\Lambda^*_{\text{T},0}$  may be interpreted as the conductance corresponding to indefinitely long Grotthuss chains. But in this low proton concentration region, the chains are not indefinitely long. They are thermally terminated. The lengths of the chains decrease with increasing temperature, thus the difference between  $\Lambda^*$  and  $\Lambda^{\circ}$  should increase with increasing temperature. Table 2 shows such to be the case. The differences and slopes in Table 2 are obtained from replots (not shown) of Figure 3 on a much expanded scale. The conductance may be represented by an equation of the form

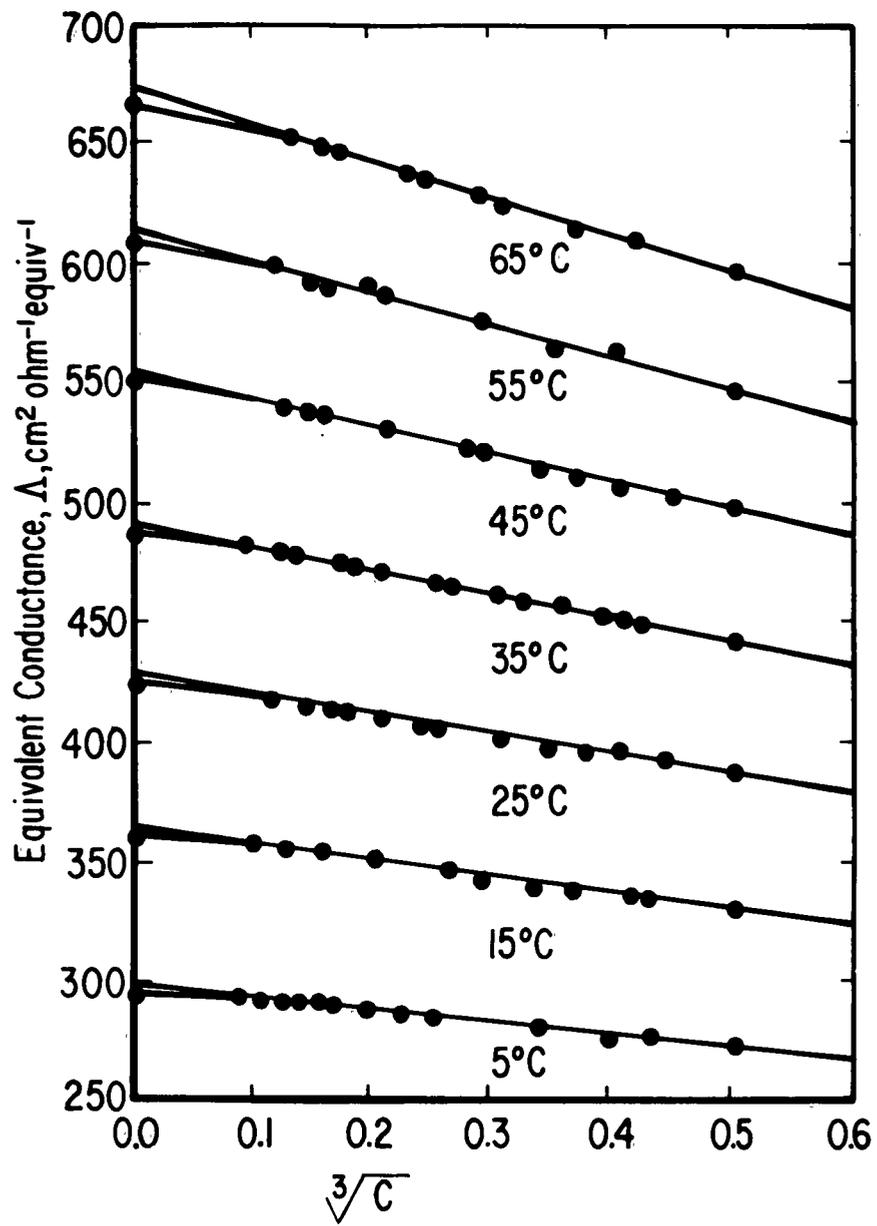
$$(4) \quad \Lambda_{\text{T},C} = \Lambda^*_{\text{T},0} - \text{TCT} - \text{CCT}$$

where TCT and CCT are thermal chain termination and concentration chain termination terms respectively. The former term is related to the difference  $\Lambda^{\circ} - \Lambda^*_{\text{T},0}$ . If the logarithm of this difference is plotted versus the reciprocal of the absolute temperature (Figure 4), a reasonably linear Arrhenius plot is obtained. There is some deviation from linearity below 25°C, but it amounts to only about 10% even at 5°C. The activation energy calculated from the slope of the linear part of this curve (65 to 25°C) by the integrated form of the Arrhenius equation is 3.53 kcal/mole. Figure 4 also shows an Arrhenius plot of the limiting conductance of aqueous KCl solutions from data quoted by Robinson and Stokes.<sup>17</sup> The curve is not linear, indicating that the activation energy of conductance,  $E_a$ , is temperature dependent. Over the temperature

19. H. S. Frank and P. T. Thompson, "A Point of View on Ion Clouds" in W. J. Hamer (ed.), The Structure of Electrolyte Solutions, John Wiley & Sons, Inc., New York, N.Y., 1959, pp. 113-134.

20. B. B. Owen and F. H. Sweeton, J. Am. Chem. Soc., **63**, 2811 (1941).

**FIGURE 3**  
**Concentration Dependence of the Conductivity**  
**of Aqueous HCl Solutions**  
**(based on data of Owen and Sweeton, ref. 20)**



**FIGURE 4**  
**Arrhenius Plots of Conductance Parameters**

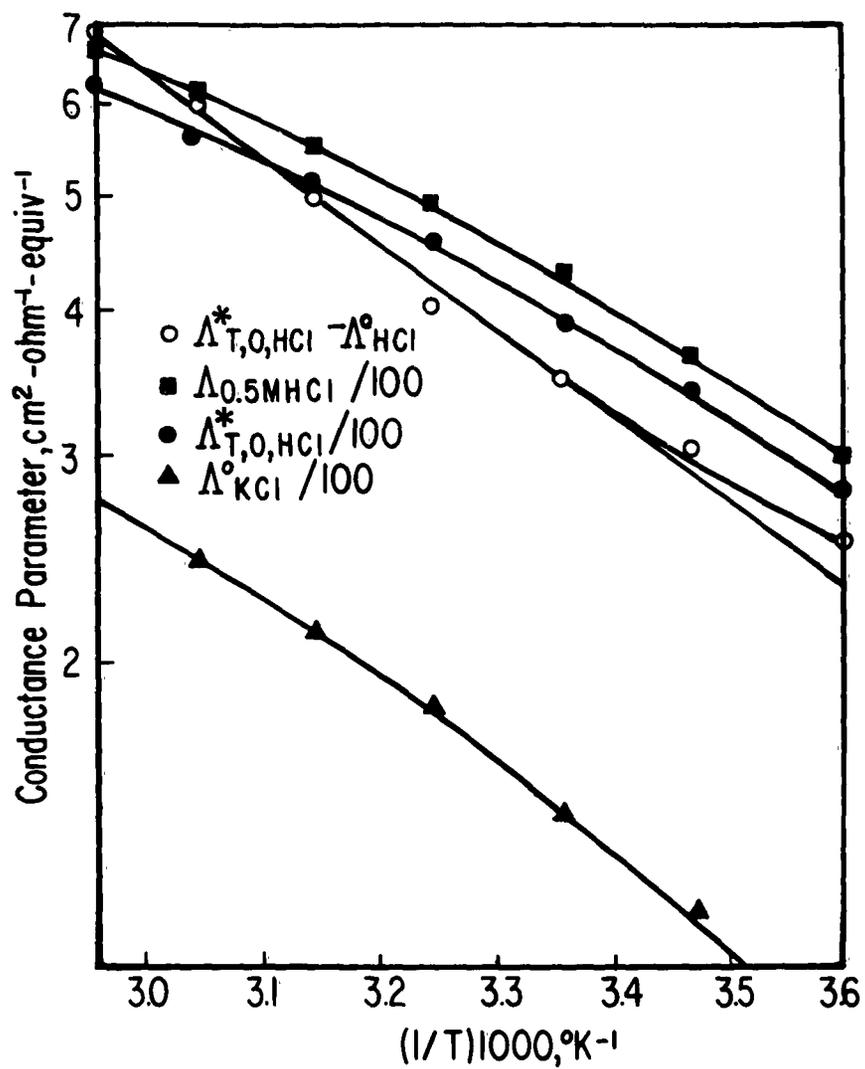


TABLE 2

Properties of the  $\Lambda$  versus  $C^{1/3}$  Plots

Temp. °C	Limit Equiv. Cond. <sup>17</sup> $\Lambda^{\circ}, \text{cm}^2\text{-ohm}^{-1} \text{equiv}^{-1}$	$\Lambda_{T,0}^*$ $\text{cm}^2\text{-ohm}^{-1} \text{equiv}^{-1}$	$\Lambda^{\circ} - \Lambda_{T,0}^*$ $\text{cm}^2\text{-ohm}^{-1} \text{equiv}^{-1}$	Slope
5	297.6	300	2.4	53
15	362.1	365	3	68
25	426.2	430	3.6	82
35	489.2	493	4	98
45	550.3	555	5	117
55	609.5	615	6	134
65	666.8	674	7	157

range 5 to 65°C, its value varies from 3.02 to 3.77 kcal/mole. The rate-controlling step of "normal" conduction in aqueous solutions involves the formation of a "hole" in the solvent.<sup>21</sup> Thermal rupture of a Grotthuss chain is the equivalent to the formation of a one-dimensional "hole," therefore one finds that  $E_a$  for thermal chain termination is nearly the same as for "hole" formation (or "normal" ionic conduction).

The slopes of  $\Lambda$  versus  $C^{1/3}$  in the linear part of the curves in Figure 4 are also temperature dependent (Table 2) and an Arrhenius plot of these slopes is linear over the entire temperature range 5 to 65°C and its slope corresponds to an activation energy of 3.38 kcal/mole--the same value as for "normal" conduction as expected. Thus equation (4) becomes

$$(5) \Lambda_{T,C} = \Lambda_{T,0}^* - \phi (1.32 \times 10^3 e^{-3530/RT} - 2.39 \times 10^4 e^{-3380/RT}) \sqrt[3]{C}$$

where T is the absolute temperature. The term  $\phi$  has been introduced because as concentration chain termination becomes more frequent, the probability of a chain being thermally terminated decreases. The value of  $\phi$  is given by

$$(6) \phi = (1.32 \times 10^3 e^{-3530/RT}) / (1.32 \times 10^3 e^{-3530/RT} + 2.39 \times 10^4 e^{-3380/RT}) \sqrt[3]{C}$$

The constants in equation (5) and the activation energies have been calculated from the data of Owen and Sweeton.<sup>20</sup> The numerical difference between the  $E_a$  is small; in actuality they probably should be the same value. This enables equation (5) to be rewritten in the form

$$(7) \Lambda_{T,C} = K_1 e^{-E_{a,r}/RT} - (\phi K_2 + K_3 \sqrt[3]{C}) e^{-E_{a,h}/RT}$$

where the K's are constants and  $E_{a,r}$  and  $E_{a,h}$  are the activation energies for the rotation of water molecules and "hole" formation in water, respectively.

An Arrhenius plot of  $\Lambda_{T,0,HCl}^*$  is shown in Figure 4. Inspection shows that it parallels that of  $\Lambda_{0.5}^*$  in HCl and that its value is less than for "normal" conduction. Again this is to be expected for the rate-determining step in protonic conduction involves the rotation of water molecules<sup>5</sup> and  $E_a$  for this process is less than that for "hole" formation.

Values of conductance, calculated from values of  $\Lambda_{T,0}^*$  given in Table 2 and from equation (5) are given in Table 3 along with the experimental values of Owen and Sweeton<sup>20</sup>. The agreement is satisfactory (less than 1% deviation) over a very wide temperature and concentration range.

21. S. Glasstone, K.J. Laidler, and H. Eyring, The Theory of Rate Processes, McGraw-Hill Book Co., Inc., New York, NY. 1941, Ch. IX and X.

TABLE 3

Comparison of Experimental and Calculated Results

	<u>Concentration</u>	<u><math>\Delta_{calc}</math></u>	<u><math>\Delta_{obs}</math></u>	<u><math>\Delta\Delta</math></u>
At 5°C	0.000000	298	298	0
	0.000776	294	295	-1
	0.00106	294	295	-1
	0.00164	293	294	-1
	0.00197	293	293	0
	0.00279	293	293	0
	0.00429	292	292	0
	0.00562	291	291	0
	0.00880	290	290	0
	0.0125	288	288	0
	0.0168	286	286	0
	0.0389	282	282	0
	0.0695	279	298	+1
	0.0834	277	277	0
	0.124	274	274	0
	0.440	261	258	+3
	0.812	252	243	+9
1.089	186	233	-47	
At 15°C	0.00000	362	362	0
	0.00106	358	358	0
	0.00228	356	356	0
	0.00459	354	354	0
	0.00909	351	351	0
	0.00946	351	351	0
	0.0191	348	347	+1
	0.0262	346	345	+1
	0.0406	343	342	+1
	0.0525	341	340	+1
	0.0695	339	337	+2
	0.0732	338	337	+1
	0.0819	337	335	+2

TABLE 3  
(continued)

	<u>Concentration</u>	<u><math>\Delta_{calc}</math></u>	<u><math>\Delta_{obs}</math></u>	<u><math>\Delta\Delta</math></u>
At 15°C	0.124	333	331	+2
	0.440	316	312	+4
	0.812	306	243	+63
	1.089	299	233	+66
At 25°C	0.00000	426	426	0
	0.00207	419	420	-1
	0.00344	416	418	-2
	0.00507	415	416	-1
	0.00589	415	415	0
	0.00987	413	412	±1
	0.0149	410	409	+1
	0.0179	409	408	+1
	0.0289	405	404	+1
	0.0426	402	401	+1
	0.0549	399	398	+1
	0.0675	397	396	+1
	0.0864	395	393	+2
	0.124	390	389	+1
	0.440	370	365	+5
0.812	357	343	+14	
1.089	351	328	+23	
At 35°C	0.00000	489	489	0
	0.00095	483	484	-1
	0.00112	482	483	0
	0.00229	480	481	-1
	0.003222	479	479	0
	0.00522	475	477	-2
	0.00642	474	475	-1
	0.00980	472	473	-1
	0.0170	468	468	0
	0.0206	467	467	0

TABLE 3  
(continued)

	<u>Concentration</u>	<u>Λ<sub>calc</sub></u>	<u>Λ<sub>obs</sub></u>	<u>ΔΛ</u>
At 35°C	0.0307	864	862	+2
	0.0366	462	461	+1
	0.0450	460	458	+2
	0.0549	457	456	+1
	0.0621	456	454	+2
	0.0695	455	453	+2
	0.124	446	444	+2
	0.440	422	417	+5
	0.812	406	392	+14
At 45°C	1.089	399	376	+23
	0.00000	550	550	0
	0.00207	540	541	-1
	0.00345	537	538	-1
	0.00470	536	536	0
	0.00938	530	531	-1
	0.00995	530	531	-1
	0.00215	523	523	0
	0.00242	522	522	0
	0.0393	516	516	0
	0.0508	513	513	0
	0.0546	511	511	0
	0.0903	508	509	-1
	0.124	499	498	+1
	0.440	410	467	+3
	0.812	451	437	+12
	1.089	443	421	+22
At 55°C	0.00000	609	609	0
	0.00183	597	599	-2
	0.00381	594	595	-1
	0.00445	593	594	-1
	0.00772	588	589	-1

TABLE 3  
(Continued)

	<u>Concentration</u>	<u>^calc</u>	<u>^obs</u>	<u>Δ^</u>
At 55°C	0.00996	585	586	-1
	0.0241	576	577	-1
	0.0244	576	576	0
	0.0454	567	568	-1
	0.0471	567	567	0
	0.0635	562	562	0
	0.0701	560	560	0
	0.124	549	549	0
	0.440	515	514	+1
	0.812	493	485	+8
At 65°C	1.089	480	464	+16
	0.00000	667	667	0
	0.000983	660	662	-2
	0.00250	657	658	-1
	0.00477	647	648	-1
	0.00517	645	647	-2
	0.0128	637	638	-1
	0.0138	636	637	-1
	0.0249	628	629	-1
	0.0300	625	626	-1
	0.0505	615	617	-2
	0.0554	614	616	-2
	0.0715	612	610	+2
	0.124	595	598	-3
	0.440	556	559	-3
0.812	529	527	+2	
1.089	515	506	+9	

Robinson and Stokes<sup>22</sup> remark that "the hydrochloric acid data are in many ways the most suitable for testing" conductance equations based on relaxation and electrophoretic effects. But these equations, as pointed out earlier, are not relevant to protonic conductance in aqueous solution. The excellent agreement which these authors obtain, therefore, is probably the consequence of the choice of the distance of closest approach--an adjustable parameter in the equations they used.

Returning now to Figure 1,  $a_{KCl}$  represents the hydrated radii of  $K^+$  and  $Cl^-$  and its value decreases with increasing pressure; however, the interpretation of  $a_{HCl}$  is less clear. The species rotating are single water molecules (see above), not clusters; hence pressure cannot significantly alter their size. The pressure dependence of the relative conductance of HCl solutions (Figure 1), therefore, cannot be explained in terms of the  $a^3$  term in equation (2). Protonic conduction is confined to the "free" water. The application of pressure increases the ratio of "free" to clustered water, and this effect gives rise to the observed increase in conductivity by increasing the relative amount of the conductive medium.

#### ACKNOWLEDGMENT

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22. R. A. Robinson and R. H. Stokes, J. Am. Chem. Soc., 76, 1991 (1954).

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