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DC-CONDUCTIVITY IN POLYVINYLCHLORIDE

by

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THE RELATIONSHIP BETWEEN THE VISCOSITY
AND THE DC-CONDUCTIVITY IN POLYVINYLCHLORIDE*

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Juerg-Heinrich Kallweit[**]

Problem

It is common assumption that the dc-conductivity σ in high-polymers mainly depends on the sample viscosity η because both the viscosity and conductivity show a similar strong temperature dependence. In accordance with Waldens' rule, it is written

(1.)
$$\sigma \sim \frac{1}{\eta}$$

To check this relation, the same samples of Polyvinylchloride (PVC) with varied plasticizer content (Palatinol AH = Dioctylphthalate) were used to measure the viscosity and dc-conductivity as function of time and temperature.

PART I

Experiments

Samples with a thickness of 0.06 cm were cut from the following material:

Table I

Sample No. and Symbol		PVC Vinoflex 332 Percent by weight	Plasticizer Palatinol AH + 0.5% Stabilizator Percent by weight
A	□	100	0
B	△	92	8
C	+	84	16
D	●	74	26
E	○	63	37
F	▽	50	50

The viscosity η was obtained from the results of a creep experiment. The measurements were done in accordance with the experiment described by Becker in [1]. A rod-shaped sample was fixed at the upper end in perpendicular position and charged with a load at the time $t = 0$. For small deformations, a tensile strength s_0 results, which is constant with respect to time. The relative change in the sample length $\epsilon (t)$ was measured as function of time. The differentation of the compliance J with respect to the time

(2.) $$J(t) = \epsilon(t)/s_0$$

(3.) $$\frac{dJ}{dt} = \frac{d\epsilon}{dt} \frac{1}{s_0} = \frac{1}{\eta}$$

gives formally the equation for Newton's flow

(4.) $$s_0 = \eta(t) \frac{d\epsilon}{dt}$$

$\eta(t)$ can be called also the Trouton-viscosity of the sample [2]. ③
can be written

⑤ $$\frac{1}{\eta} = -\frac{J}{t}\,\frac{d\,(\lg\frac{1}{J})}{d\,(\lg t)}$$

The differentation was obtained by graphical methods. From equation ③
follows that η must be a function of the time. The viscosity of high-
polymers can be considered to be a criterion for the "friction force"
between the position-changing segments of the molecular chains during
stress. The decreasing possibility for position changes in stress
direction leads to increasing the viscosity as function of the load
period. To eliminate this time dependence as well as possible, only
the η-values obtained three seconds after load charging were used for
the comparison with the values of the specific dc-conductivity. Figure I
shows the temperature dependence of the viscosity obtained after three
with the plasticizer content as parameter.

The experimental device for the determination of the specific dc-
conductivity is described in [3] . The samples were again placed in
vacuum of about 10^{-4} Torr during the measurement. The applied voltage
of 1000 volt was identical with an electrical field strength of 16.7
[KV/cm.] All samples were heat treated, before each change of the measuring
temperature, for one hour at a temperature $30^{\circ}C$ above the respective
second-order transition temperature. Afterwards they were cooled down
slowly (or heated up slowly) until the desired sample temperature was
reached. This procedure was necessary to eliminate all polarization
effects in the sample due to former charging. Polarization effects
are the reason for the strong time dependence of the conductivity in

With Stoke's law for the factor f

(7.) $\quad f = 6 \cdot r \cdot \pi \cdot \eta \qquad$ r = radius of the ion with supposed spherical shape

and

(8.) $\quad \sigma = Z \cdot q \cdot c \, \dfrac{\bar{v}}{E} \qquad$ c = concentration

q = charge unit

Z = valency; Z is supposed to be 1 in this paper

follows

(9.) $$\sigma (T) \cdot \eta (T) = \frac{q^2}{6\pi r} \cdot c(T)$$

$$= const \cdot c(T)$$

Because σ also contains the temperature-dependent concentration c(T), the product $\sigma \cdot \eta$ should be temperature independent. Figure IV shows that the expected relationship was not observed at all. There are two possible explanations: (1) Stoke's law is a too rough approximation in the case of the tested highpolymers; (2) The viscosity in Stoke's law is not identical with the viscosity η obtained by retardation experiments, which may be called "macroviscosity." But an effective local viscosity $\eta '$ (microviscosity) must be used in connection with Stoke's law. Only in the case of fluids η is a good approximation for $\eta '$.

At first it was necessary to check whether or not any relationship between σ and η could be found. It is known from dielectric measurements on PVC [6] that the maximum of the loss tangent was always nearly the same when the viscosity η of the sample is the same given value. It made no difference whether the magnitude of η was due to the temperature or whether it was due to the plasticizer used. Therefore, it was checked as to which relationship between σ and η exists

when equal magnitudes of σ are considered, which can be due to temperature or due to plasticizer variation. The shape of the plot of $\lg \sigma$ over $\lg \eta$ seemed to indicate a hyperbola (Figure V). Afterwards, $\lg \eta$ was plotted as function of the product $\lg(\sigma \cdot \eta)$. The graphic representation (Figure VI) gives the desired possibility for the derivation of the relationship between σ and η.

(10) $$\lg \eta = m \lg \sigma \cdot \eta + \text{const.}$$

or

(11) $$\sigma \cdot \eta^{\frac{m-1}{m}} = \text{const.}$$

for the samples A, B, and C. The tan of the slope angle m must be derived with the help of the above plot.

PART II

Discussion

The experimental results described in Part I seemed to show that it is problematic to use the viscosity values obtained by retardation experiments in connection with Stoke's formula in (7). It was asked which results would be <u>formally</u> obtained when the magnitude of the viscosity in formula (11) would be considered to be the magnitude of the effective local viscosity η'

(12) $$\left| \eta^{\frac{m-1}{m}} \right| = \left| \eta' \right|$$

η' should have the dimension of a viscosity. The numerical results are given in Table II. Because m is nearly one, the values for η' are unexpectedly low; but it has been shown by Ferry [7] that the

order of the effective local viscosity for small foreign molecules may

be of the calculated order. It is furthermore interesting to note that

the compensation law [8] [9] seems to hold for the conductivity pro-

cess in the system PVC/Plasticizer. (See Figure VII). Formula ⑦ ,

written now as

(7a.)
$$f = 6 \cdot \pi \cdot r \cdot \eta'$$

would give for the equation in ⑨

(9a)
$$\sigma(T) \; \eta'(T) = \frac{q^2}{6 \cdot \pi \cdot r} \cdot c(T)$$

The graph in Figure VIII shows that now the expected relationship is

obtained. This result indicated continuation of the formal test by

writing for the median drift velocity $\overline{v}$

(13.)
$$\overline{v} = q \cdot E / 6 \cdot \pi \cdot r \cdot \eta'$$

and for the conductivity

(14.)
$$\sigma = c \; q^2 / 6 \cdot \pi \cdot r \cdot \eta' / \text{ valency } z = 1$$

where the mobility b_e of the ions is given by

(15.)
$$b_e = \frac{q}{6 \cdot \pi \cdot r \cdot \eta'}$$

Finally, the expression for the diffusion coefficient D would be

(16.)
$$D = KT / 6 \cdot \pi \cdot r \cdot \eta'$$

because the relationship between the electrical and mechanical defini-

tion for the mobility is $b_e = q \cdot b_{mech}$. Table III gives the numerical

results for the samples A, B, C and some values for D. The calculation

could not be done without another assumption for the radius r of the

ions. The kind of the ions is not known, but most probably they will

be small molecules. Under this assumption, two possible values were

chosen for the unknown radius r :

$$r = 1 \cdot 10^{-7} \text{ cm}$$

$$r = 5 \cdot 10^{-7} \text{ cm}$$

The values for sample D were added to show that the nonlinear plots in Figure VI do not give sensible values.

The crude approximations do not allow detailed discussion of the result summarized in Table III. But this result seems to indicate that the values for the ion velocity, concentration, and the diffusion coefficient are of a range, which could be a more-or-less fair approximation of the actual values. The expected temperature dependence of the mobility of the single samples was obtained. But it is difficult to understand that the mobility did not increase with increasing plasticizer content. The common assumption is that the increase of the conductivity with higher plasticizer content is chiefly due to the greater ion-mobility. Here it would turn out that the ion concentration is the dominating factor. The values for the diffusion coefficient do not differ markedly from the values of Luther and Meyer, who measured the diffusion coefficient of plasticizer in PVC [10] . For sample C at 343°K was measured $b = 1.2 \cdot 10^{-10}$ $[\text{cm}^2 \text{ sec}^{-1}]$ and by Meyer $D = 1.4 \cdot 10^{-11}$ $[\text{cm}^2 \text{ sec}^{-1}]$. Although it cannot be said that plasticizer molecules are responsible for the charge transport, the low order of the diffusion coefficient may indicate that molecules as charge carriers are migrating under the force of the applied electrical field.

The calculated values for D and $\bar{v}$ can be used to check whether or not values are obtained for the jump distance d of the ions, which

are of the same order as values known from literature. Foss and Danhauser
(11) evaluated the jump distance in PPN according to the reaction rate
theory from the isothermal field dependence as about 200 $\overset{o}{A}$; Amborski [12]
measured for Mylar at $130^{o}C$, 69 $\overset{o}{A}$. The values in the case of PVC with-
out plasticizer would be for the assumed ion radius $r = 1 \cdot 10^{-7}$, iden-
tical to 127 $\overset{o}{A}$ (See [12]); but this fair agreement should not be over-
valued because $D = \overline{v}d$ turns out, according to the approximations used
in this paper, to be independent from any specific sample.

Other types of highpolymer material must be tested before any
statements can be made. The determination of the free volume should
be added to the viscosity measurements, and the dc-conductivity should
be measured as function of temperature and electric field strength [13].
Samples with varied plasticizer content and crystallinity should be
checked. The understanding of the dc-conductivity in highpolymers
depends on the exact knowledge of the mobility of the charge carriers.

ACKNOWLEDGEMENT

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SYNOPSIS

In general, it has been said that the dc-conductivity σ is proportional to the inverse viscosity η. But the relation $\sigma \cdot \eta = const$ has not been proven in the case of the system Polyvinylchlorid/Dioctylphthalate. The experimental results can be described by $\sigma \cdot \eta^{\frac{m}{m-1}} = const$ if the plasticizer content is not too high. The ion mobility seems to depend on a local effective viscosity, which differs from that viscosity which is derived from retardation experiments.

T	1000/T	σ	η	m	$\|\eta'\|=\|\eta^{\frac{m-1}{m}}\|$	$\sigma \cdot \eta'$
[°K]	[1/°K]	[1/sec]	[g/cm · sec]			[g/cm · sec²]
A 293	341	$3.5 \cdot 10^{-4}$	$5.5 \cdot 10^{12}$	1.15	$4.5_6 \cdot 10^1$	$1.6_0 \cdot 10^{-2}$
303	330	$3.6 \cdot 10^{-4}$	$5.0 \cdot 10^{12}$		$4.5_5 \cdot 10^1$	$1.6_4 \cdot 10^{-2}$
313	319	$3.7 \cdot 10^{-4}$	$4.3 \cdot 10^{12}$		$4.4_4 \cdot 10^1$	$1.6_4 \cdot 10^{-2}$
323	309	$3.9 \cdot 10^{-4}$	$3.2 \cdot 10^{12}$		$4.2_5 \cdot 10^1$	$1.6_6 \cdot 10^{-2}$
333	300	$4.3 \cdot 10^{-4}$	$2.1 \cdot 10^{12}$		$4.0_6 \cdot 10^1$	$1.7_4 \cdot 10^{-2}$
343	291	$4.8 \cdot 10^{-4}$	$9.0 \cdot 10^{11}$		$3.5_8 \cdot 10^1$	$1.7_2 \cdot 10^{-2}$
353	283	$5.5 \cdot 10^{-4}$	$1.7 \cdot 10^{11}$		$2.8_9 \cdot 10^1$	$1.5_9 \cdot 10^{-2}$
363	275	$7.6 \cdot 10^{-4}$	$2.0 \cdot 10^{10}$		$2.2_4 \cdot 10^1$	$1.7_3 \cdot 10^{-2}$
B 293	341	$7.3 \cdot 10^{-4}$	$1.3 \cdot 10^{13}$	1.17	$7.9_0 \cdot 10^1$	$5.7_5 \cdot 10^{-2}$
303	330	$9.5 \cdot 10^{-4}$	$1.9 \cdot 10^{12}$		$6.1_5 \cdot 10^1$	$5.8_5 \cdot 10^{-2}$
313	319	$1.3 \cdot 10^{-3}$	$3.1 \cdot 10^{11}$		$4.6_4 \cdot 10^1$	$6.0_3 \cdot 10^{-2}$
323	309	$2.0 \cdot 10^{-3}$	$5.0 \cdot 10^{10}$		$3.5_5 \cdot 10^1$	$6.9_9 \cdot 10^{-2}$
333	300	$3.5 \cdot 10^{-3}$	?		/	/
343	291	$7.5 \cdot 10^{-3}$	?		/	/
353	283	$1.8 \cdot 10^{-2}$	?		/	/
C 293	341	$4.4 \cdot 10^{-3}$	$2.7 \cdot 10^{11}$	1.22	$1.1_2 \cdot 10^2$	$4.9_2 \cdot 10^{-1}$
303	330	$6.4 \cdot 10^{-3}$	$6.7 \cdot 10^{10}$		$9.0_5 \cdot 10^1$	$5.8_0 \cdot 10^{-1}$
313	319	$9.5 \cdot 10^{-3}$	$1.4 \cdot 10^{10}$		$6.7_1 \cdot 10^1$	$6.3_6 \cdot 10^{-1}$
323	309	$1.6 \cdot 10^{-2}$	$4.8 \cdot 10^9$		$4.6_5 \cdot 10^1$	$7.4_4 \cdot 10^{-1}$
333	300	$2.8 \cdot 10^{-2}$	$3.2 \cdot 10^9$		$4.4_4 \cdot 10^1$	1.2_4
343	291	$5.9 \cdot 10^{-2}$	$2.8 \cdot 10^9$		$4.3_0 \cdot 10^1$	2.5_4
D 293	241	$7.0 \cdot 10^{-2}$	$3.3 \cdot 10^9$	-4.33	$5.4 \cdot 10^{11}$	$3.7 \cdot 10^{10}$
313	319	$6.0 \cdot 10^{-1}$	$2.2 \cdot 10^9$		$2.6 \cdot 10^{11}$	$1.5 \cdot 10^{11}$
333	300	$3.4 \cdot 10^0$	$1.6 \cdot 10^9$		$2.2 \cdot 10^{11}$	$7.5 \cdot 10^{11}$

TABLE II

Table III is a large data table (rotated on the page). It contains two blocks of data for fixed r values, each with columns v, b, D, c, indexed by temperature T and grouped into sets A, B, C, D.

Group	T [°K]	$r = 5.0 \cdot 10^{-7}$ cm				$r = 1.0 \cdot 10^{-7}$ cm			
		v [cm·sec^{-1}]	b [cm$^{3/2}$·g$^{-1/2}$]	D [cm^2·sec^{-1}]	c [cm^{-3}]	v [cm·sec^{-1}]	b [cm$^{3/2}$·g$^{-1/2}$]	D [cm^2·sec^{-1}]	c [cm^{-3}]
A	293	$7.3_5 \cdot 10^{-5}$	$1.32 \cdot 10^{-6}$	$9.4 \cdot 10^{-11}$	$4.6 \cdot 10^{11}$	$3.6_9 \cdot 10^{-4}$	$6.62 \cdot 10^{-6}$	$4.7 \cdot 10^{-10}$	$9.2 \cdot 10^{10}$
A	303	$7.4_0 \cdot 10^{-5}$	$1.33 \cdot 10^{-6}$	$9.8 \cdot 10^{-11}$	$4.7 \cdot 10^{11}$	$3.7_2 \cdot 10^{-4}$	$6.65 \cdot 10^{-6}$	$4.9 \cdot 10^{-10}$	$9.4 \cdot 10^{10}$
A	313	$7.6_0 \cdot 10^{-5}$	$1.36 \cdot 10^{-6}$	$1.0 \cdot 10^{-10}$	$4.8 \cdot 10^{11}$	$3.8_1 \cdot 10^{-4}$	$6.84 \cdot 10^{-6}$	$5.0 \cdot 10^{-10}$	$9.6 \cdot 10^{10}$
A	323	$7.9_4 \cdot 10^{-5}$	$1.42 \cdot 10^{-6}$	$1.1 \cdot 10^{-10}$	$4.8 \cdot 10^{11}$	$3.9_7 \cdot 10^{-4}$	$7.12 \cdot 10^{-6}$	$5.5 \cdot 10^{-10}$	$9.6 \cdot 10^{10}$
A	333	$8.5_1 \cdot 10^{-5}$	$1.50 \cdot 10^{-6}$	$1.2 \cdot 10^{-10}$	$5.0 \cdot 10^{11}$	$4.1_5 \cdot 10^{-4}$	$7.44 \cdot 10^{-6}$	$6.0 \cdot 10^{-10}$	$1.0 \cdot 10^{11}$
A	343	$9.4_5 \cdot 10^{-5}$	$1.69 \cdot 10^{-6}$	$1.4 \cdot 10^{-10}$	$5.0 \cdot 10^{11}$	$4.7_1 \cdot 10^{-4}$	$8.94 \cdot 10^{-6}$	$7.0 \cdot 10^{-10}$	$1.0 \cdot 10^{11}$
A	353	$1.1_6 \cdot 10^{-4}$	$2.08 \cdot 10^{-6}$	$1.8 \cdot 10^{-10}$	$4.6 \cdot 10^{11}$	$5.8_0 \cdot 10^{-4}$	$1.04 \cdot 10^{-5}$	$9.0 \cdot 10^{-10}$	$9.2 \cdot 10^{10}$
A	363	$1.5_1 \cdot 10^{-4}$	$2.70 \cdot 10^{-6}$	$2.4 \cdot 10^{-10}$	$5.0 \cdot 10^{11}$	$7.5_2 \cdot 10^{-4}$	$1.35 \cdot 10^{-5}$	$1.2 \cdot 10^{-9}$	$1.0 \cdot 10^{11}$
B	293	$4.2_6 \cdot 10^{-5}$	$7.62 \cdot 10^{-7}$	$5.4 \cdot 10^{-11}$	$1.6 \cdot 10^{12}$	$2.1_4 \cdot 10^{-4}$	$3.83 \cdot 10^{-6}$	$2.7 \cdot 10^{-10}$	$3.2 \cdot 10^{11}$
B	303	$5.4_6 \cdot 10^{-5}$	$9.75 \cdot 10^{-7}$	$7.2 \cdot 10^{-11}$	$1.7 \cdot 10^{12}$	$2.7_4 \cdot 10^{-4}$	$4.92 \cdot 10^{-6}$	$3.6 \cdot 10^{-10}$	$3.4 \cdot 10^{11}$
B	313	$7.2_6 \cdot 10^{-5}$	$1.31 \cdot 10^{-6}$	$1.0 \cdot 10^{-10}$	$1.8 \cdot 10^{12}$	$3.6_4 \cdot 10^{-4}$	$6.52 \cdot 10^{-6}$	$5.0 \cdot 10^{-10}$	$3.6 \cdot 10^{11}$
B	323	$9.4_5 \cdot 10^{-5}$	$1.69 \cdot 10^{-6}$	$1.3 \cdot 10^{-10}$	$1.9 \cdot 10^{12}$	$4.7_1 \cdot 10^{-4}$	$8.47 \cdot 10^{-6}$	$6.5 \cdot 10^{-10}$	$3.8 \cdot 10^{11}$
C	293	$3.0_0 \cdot 10^{-5}$	$5.38 \cdot 10^{-7}$	$3.8 \cdot 10^{-11}$	$1.4 \cdot 10^{13}$	$3.6_3 \cdot 10^{-4}$	$2.70 \cdot 10^{-6}$	$1.9 \cdot 10^{-10}$	$2.8 \cdot 10^{12}$
C	303	$3.7_1 \cdot 10^{-5}$	$6.69 \cdot 10^{-7}$	$4.9 \cdot 10^{-11}$	$1.7 \cdot 10^{13}$	$1.8_4 \cdot 10^{-4}$	$3.32 \cdot 10^{-6}$	$2.4 \cdot 10^{-10}$	$3.4 \cdot 10^{12}$
C	313	$5.0_1 \cdot 10^{-5}$	$9.00 \cdot 10^{-7}$	$6.9 \cdot 10^{-11}$	$1.8 \cdot 10^{13}$	$2.5_1 \cdot 10^{-4}$	$4.50 \cdot 10^{-6}$	$3.4 \cdot 10^{-10}$	$3.6 \cdot 10^{12}$
C	323	$7.4_0 \cdot 10^{-5}$	$1.33 \cdot 10^{-6}$	$1.0 \cdot 10^{-10}$	$2.2 \cdot 10^{13}$	$3.7_2 \cdot 10^{-4}$	$6.65 \cdot 10^{-6}$	$5.0 \cdot 10^{-10}$	$4.4 \cdot 10^{12}$
C	333	$7.5_5 \cdot 10^{-5}$	$1.35 \cdot 10^{-6}$	$1.1 \cdot 10^{-10}$	$3.5 \cdot 10^{13}$	$3.8_0 \cdot 10^{-4}$	$6.80 \cdot 10^{-6}$	$5.5 \cdot 10^{-10}$	$7.0 \cdot 10^{12}$
C	343	$7.8_0 \cdot 10^{-5}$	$1.41 \cdot 10^{-6}$	$1.2 \cdot 10^{-10}$	$7.4 \cdot 10^{13}$	$3.9_1 \cdot 10^{-4}$	$7.03 \cdot 10^{-6}$	$6.0 \cdot 10^{-10}$	$1.4 \cdot 10^{13}$
D	293	$6.2 \cdot 10^{-15}$	$1.1 \cdot 10^{-16}$	$7.9 \cdot 10^{-21}$	$1.1 \cdot 10^{24}$	$3.1 \cdot 10^{-14}$	$5.56 \cdot 10^{-16}$	$3.9 \cdot 10^{-20}$	$2.2 \cdot 10^{23}$
D	313	$1.2 \cdot 10^{-14}$	$2.2 \cdot 10^{-16}$	$1.7 \cdot 10^{-20}$	$4.3 \cdot 10^{24}$	$6.2 \cdot 10^{-14}$	$1.1_2 \cdot 10^{-15}$	$8.5 \cdot 10^{-20}$	$8.8 \cdot 10^{23}$
D	333	$1.7 \cdot 10^{-14}$	$3.0 \cdot 10^{-16}$	$2.2 \cdot 10^{-20}$	$2.1 \cdot 10^{25}$	$8.3 \cdot 10^{-14}$	$1.5_0 \cdot 10^{-15}$	$1.1 \cdot 10^{-19}$	$4.2 \cdot 10^{24}$

TABLE III

13

CAPTIONS TO FIGURES

Figure I:

Log Viscosity Vs. Reciprocal Temperature For the System PVC/DOP.
The Plasticizer Content Is Parameter.

Figure II:

The Polarization Voltage as Function of Temperature. The
Plasticizer Content Is Parameter; Charging Time Is 600 Seconds.

Figure III:

Log Specific Conductivity Vs. Reciprocal Temperature For the
System PVC/DOP. The Plasticizer Content Is Parameter.

Figure IV:

Log (Specific Conductivity Times Viscosity) Vs. Temperature.

Figure V:

Log Specific Conductivity Vs. Log Viscosity.

Figure VI:

Log Viscosity Vs. Log (Specific Conductivity Times Viscosity)
With the Plasticizer Content as Parameter.

Figure VII:

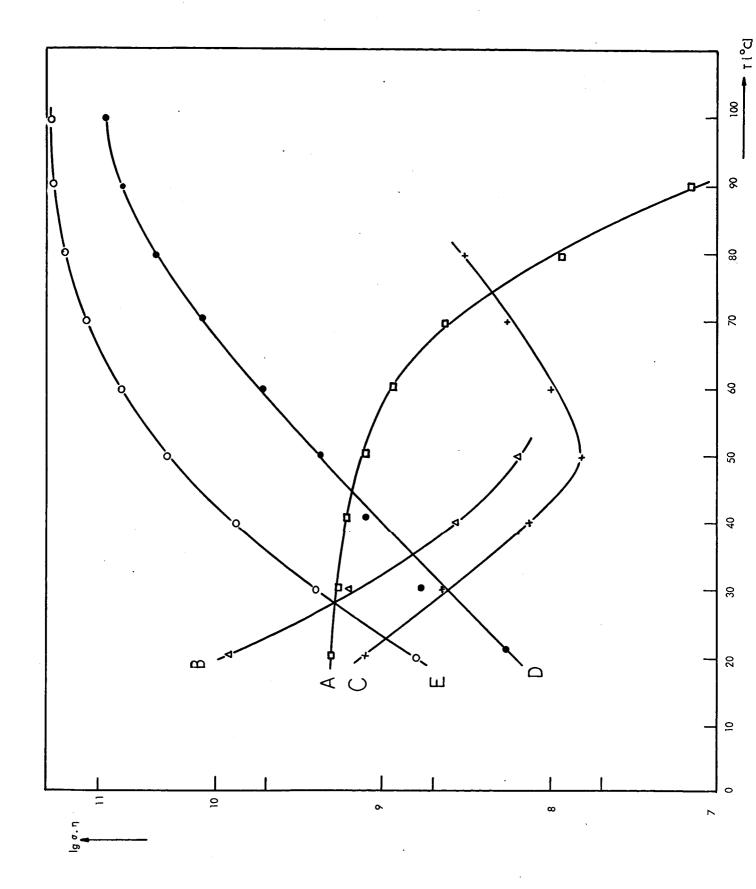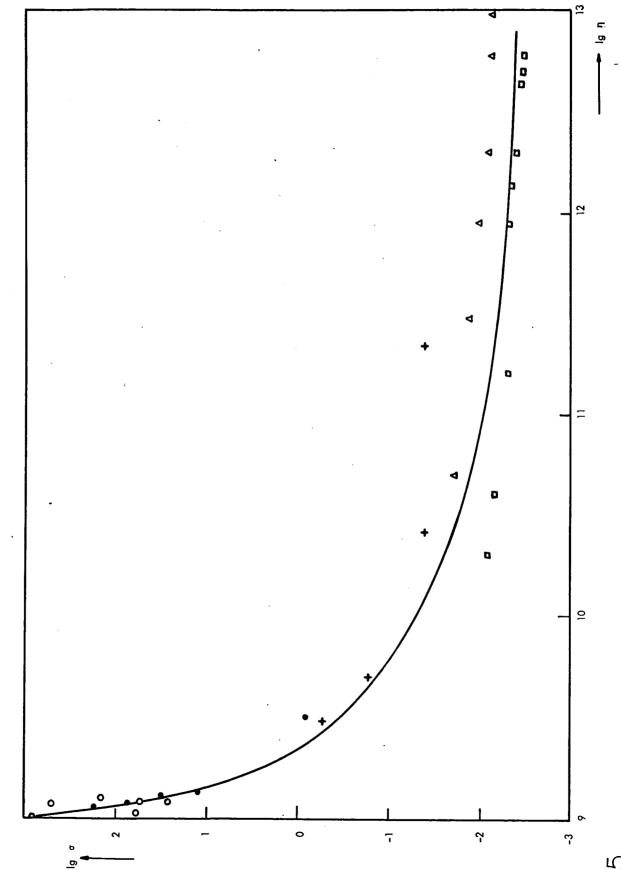Log (Specific Resistance) Vs. Activation Energy.

Figure VIII:

Log Specific (Conductivity Times Effective Viscosity) Vs.
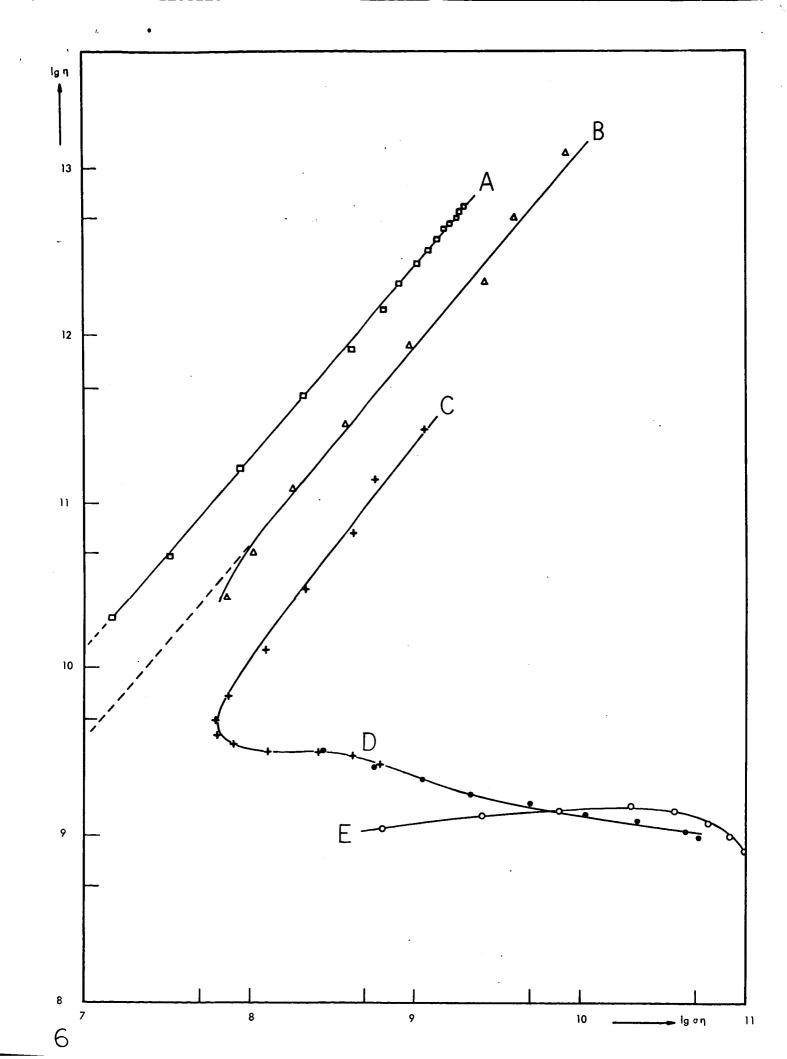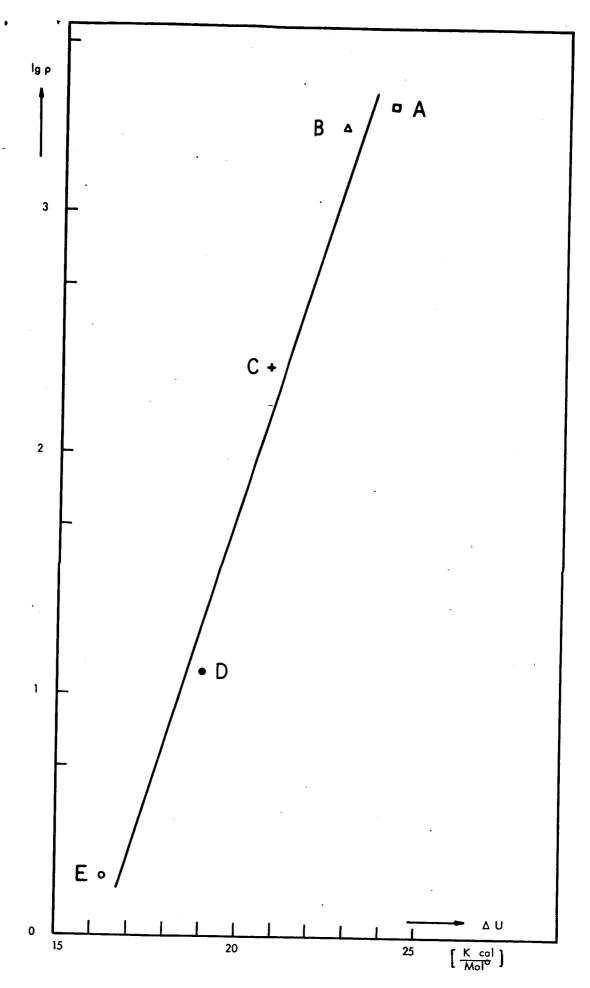Temperature.

2

E

D

C

B

A

lg σ

2

1

0

-1

-2

-3

2.5   2.6   2.7   2.8   2.9   3.0   3.1   3.2   3.3   3.4

$\dfrac{1000}{T}$ $\left[\dfrac{1}{^\circ K}\right]$

3

lg σ.n

B
A
C
E
D

T (°C)

4

lg η

lg σ

5

6

7

lg σ η'

T [°C]

0   10   20   30   40   50   60   70   80

0

-1

-2

A

B

C

8