

A Transient Method for Measurements of Electric Conduction from Plastic Insulators*

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1. Introduction

When a steady voltage is applied to polymer insulators, the current decreases with time from its initial value to a steady leakage current. The time-dependent current, after the initial surge of charge has passed, is termed the absorption current, which is relevant to a certain nature of dielectric relaxation in the very low frequency region. The steady leakage current, termed the steady-state or the conduction current, has provided means of studying the mechanism of electric conduction in insulators.

It is necessary to separate the absorption current and the steady-state current in so far as the validity of the superposition principle is demonstrated, but it is difficult from a practical point of view. This is because it is not rare for the absorption current to decay over hours, days, or even months; moreover, there is still a strong suspicion whether or not the charging current, in the presence of space-charge, arrives at any almost steady values which may be interpreted as the conduction current. This difficulty in a clean separation appears to be the largest trouble in the study both of dielectric behavior and of electric conduction for plastic insulators.

As one of the ways to overcome it, a linearly rising or falling voltage with time (expressed as "transient method") besides a steady voltage (expressed as "step method") was applied to specimens of polyvinyl chloride (PVC) and polyvinyl alcohol (PVA) in this paper. A scheme of this kind is by no means new, but only the dielectric behavior within few minutes has become a theme of research by the transient method⁽¹⁾⁽²⁾. From a different point of view, the rising or falling rate of voltage was very much smaller in our experiments.

The transient current obtained in this way may be resolved into a linear absorption current, a conduction current, and others, from which the volumes of transient capacitance and bulk resistance may be calculated. The deviation from superposition principle and Ohm's law occurs and possible mechanisms will be discussed.

2. Principal of the Method

For a dielectric which is linear in the sense of Hopkinson's superposition principle, the absorption current produced by the application of a certain voltage $V(t)$ as a

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function of time may be usually expressed as follows :

$$I = C_{\infty} \frac{dV(t)}{dt} + (C_s - C_{\infty}) \int_0^{\infty} \frac{dV(t-x)}{dx} \varphi(x) dx, \tag{1}$$

where C_s and C_{∞} are the capacitances at limiting low (static) and high frequencies, respectively and $\varphi(t)$ is an after-effect or a decay function which decreases monotonically with time. Note that $\varphi(0)$ is finite, $\varphi(\infty)=0$, and

$$\int_0^{\infty} \varphi(x) dx = 1. \tag{2}$$

From Eq. (1), the complex dielectric constant of linear dielectrics has often been calculated in the presence of a sinusoidal voltage. For further detail the reader should refer to Ref. (3) (pp. 95-101).

Here an attempt will be made to use Eq. (1) as a tool for research not only in the dielectric property but also in the electric conduction. It is required to derive from Eq. (1) the terms of the absorption current under $V(0)$ at $t=0$ and the conduction current under every voltage. Then

$$I(t) = \frac{V(t)}{R} + (C_s - C_{\infty}) V(0) \varphi(t) + C_{\infty} \frac{dV}{dt} + (C_s - C_{\infty}) \int_0^{\infty} \frac{dV(t-x)}{dx} \varphi(x) dx, \tag{3}$$

where $V(t)/R$ is the conduction current and R is the bulk resistance of specimens.

Most measurements are made by the transient method ; that is, the voltage applied to the specimen is

$$\begin{aligned} V(t) &= V_0 & -t_0 \leq t \leq 0 \\ &= V_0 + \alpha t & t \geq 0, \end{aligned} \tag{4}$$

where V_0 is a constant voltage at $t=0$, t_0 is a waiting time, and α is a constant rate of rising or falling voltage with time. Note that α is either positive or negative. The introduction of Eq. (4) in Eq. (3) leads to

$$I(\alpha, t) = \frac{V(t)}{R} + \Delta C V_0 \varphi(t+t_0) + \alpha C_{\infty} + \Delta C \alpha \int_0^t \varphi(x) dx, \tag{5}$$

where $\Delta C = C_s - C_{\infty}$. Hereafter $I(\alpha, t)$ is expressed as "transient current".

Some current measurements in the step method are made by the application of a constant voltage V_0 for a certain period t_c and by the subsequent removal one. The charging current $I_c(t)$ and the discharging current $I_d(t)$ may be expressed usually according to the dielectric after-effect of linear dielectrics by :

$$I_c(t) = \Delta C V_0 \varphi(t) + \frac{V_0}{R} \tag{6}$$

$$\begin{aligned} I_d(t) &= \Delta C V_0 \varphi(t+t_c) - \Delta C V_0 \varphi(t) & \text{at any time} \\ &\simeq -\Delta C V_0 \varphi(t) & \text{after full charging,} \end{aligned} \tag{7}$$

where time t in Eqs. (6) and (7) is the time elapsed from the application and removal of V_o , respectively. In the case of full charging

$$I_c(t) = \frac{V_o}{R} - I_d(t). \tag{8}$$

Finally Eq. (5) is transformed by substituting Eqs. (6), (7), and (8) as follows :

$$I(\alpha, t) = I_c(t) + \alpha \left\{ C_\infty + \frac{1}{V_o} \int_0^t I_c(t) dt \right\} \tag{9}$$

$$= I_c(t) + \alpha C(t) \tag{10}$$

$$= \frac{V(t)}{R} + \alpha C_\infty - I_d(t) - \frac{\alpha}{V_o} \int_0^t I_d(t) dt, \tag{11}$$

where $C(t)$ is the transient capacitance as :

$$C(t) = C_\infty + \Delta C \int_0^t \varphi(x) dx \simeq -\frac{1}{V_o} \int_0^t I_d(t) dt. \tag{12}$$

Note that $t \rightarrow 0$, $C(t) = C_\infty$; $t \rightarrow \infty$, $C(t) = C_s$. The transient current may be expressed approximately at time t enough for the purpose by

$$I(\alpha, t) = \frac{V(t)}{R} + \alpha C_s. \tag{13}$$

Therefore the values of R , C_s , C_∞ , and $C(t)$ may be evaluated, in a somewhat involving fashion, from the Equations as before by the use of current measurements of $I(\alpha, t)$, $I_c(t)$, and $I_d(t)$.

3. Experimental

The polymer materials used in this paper were polyvinyl chloride and polyvinyl alcohol. The samples were prepared by casting solution method⁽⁴⁾ in the form of film. Then aluminum was evaporated on the surfaces of the casting film after a heat treatment above 100°C.

The voltage up to 500 volt across the specimen was furnished by a variable regulated power supply. In the case for the transient method, a linearly rising or falling voltage was given from the output, the variable tap of which could be rotated, clockwise or counter-clockwise, at a suitable speed by a servomotor and various gears. Then it was possibly to obtain in various rates from 0.79 to 19.00 volt per minute. Details of the method for current measurements have been reported in a previous paper⁽⁴⁾.

4. Results and Discussion

Figure 1 shows the transient currents from PVC film at various rates. In nature the curves $I(\alpha, t) - V(\alpha, t)$ become non-Ohmic. The current at negative α follows in the reverse direction of applied voltage under certain circumstances. This negative

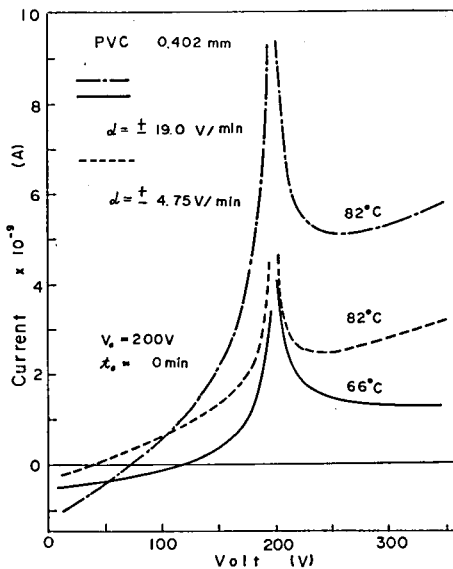


Fig.1. Transient currents produced by rising and falling voltage from 200 Volt for PVC film.

current may suggest that the effect of the dielectric absorption has appeared sensitively.

Figure 2 (a) shows the transient current $I(\alpha, t)$ and its component elements ; namely, the conduction current, an absorption current, and others. Fig. 2(b) shows the charging current $I_c(t)$ and the discharging current $I_d(t)$ in the step method and their electric charges passed with time from the same film. The bulk resistance calculated with

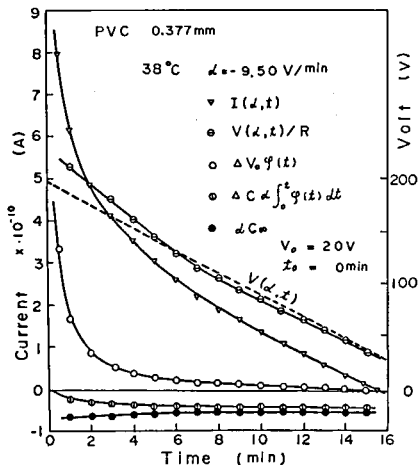


Fig.2 (a). Transient current and its component currents from PVC film.

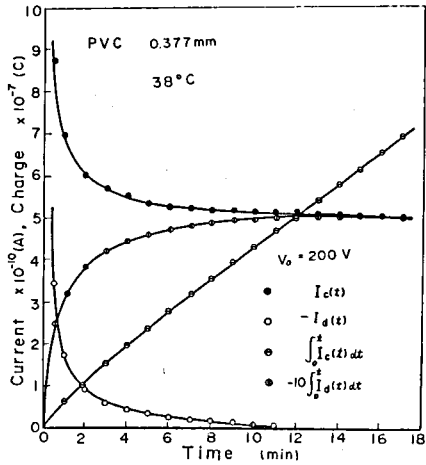


Fig.2. (b). Charging current $I_c(t)$ and discharging current $I_d(t)$ under 200 volt. Their electric charges passed with time.

Table I. Bulk resistance R and transient capacitance $C(t)$ for PVC film

Time elapsed (Min)	Transient method			Step method		
	$V(\alpha, t)$ (V)	R (Ohm)	$C(t)$ (F)	V_o (V)	R (Ohm)	$C(t)$ (F)
1	187	3.50×10^{11}	4.01×10^{-10}	200	3.81×10^{11}	1.11×10^{-10}
5	142	3.87	1.20×10^{-9}	200	3.98	1.94
10	88	4.07	2.26	200	3.95	2.23
13	58	3.89	2.80	200	3.94	2.31
15	39	4.25	2.99	200	3.96	2.34

measurements of $I(\alpha, t)$ and $I_d(t)$ as shown in Fig. 2 are given in Table I, where the transient capacitances $C(t)$ were calculated from Eq. (10). The calculation of R in accordance with Eq. (11) were carried out by means of the discharging current, because the discharging data become almost more reproducible than the charging those in our experiments. Moreover R and $C(t)$ in the step method calculated from Eqs. (6), (7), and (12) are given in Table 1. It is evident that $I(\alpha, t') \equiv 0$, at time $t' = 15$ minutes 30 seconds as Fig. 2(a). Consequently Eq. (9) gives $C_\infty = 3.71 \times 10^{-10}$ Farad at time t' . Both the values of R and $C(t)$ obtained in the transient method are greater than those obtained in the step method with time elapsed. In particular $C(t)$ in the transient method increases rapidly with time and its value at $t = 15$ minutes is about 10 times as much as that in the step method. Such extremely large capacitance reveals that non-linear effect is present in addition to the linear one; and relatively greater at smaller voltage.

The polarization of insulators under the influence of a constant voltage is generally one or more of the followings⁽⁹⁾: (1) dipole polarization, (2) migration of electric charge with trapping, (3) ionic drift over macroscopic distances, (4) electric injection from the electrodes. Hence the ionic drift and the charge injection should be considered as possible causes of the non-linearity observed in our experiments.

Figure 3 shows $I(\alpha, t)$ at positive α and its component elements from PVC-DOP film. The values of R and $C(t)$ are given in Table II, which were calculated in the same way as before. No explanation of this significant differences between the two methods; the transient one and the step one, may be satisfactory on the basis of the superposition principle due to dipole polarization. There is a tendency for both the transient and the conduction current to increase with voltage less than linearity.

Figure 4 shows the transient and the conduction current at various rates of positive α from the same film of PVC-DOP. The time spend from 20 volt to 400 volt amounted to over a range from 20 minutes to 8 hours. This experiments indicates that the deviation from Ohm's law are greater with further decreasing values of α . In particular

Table II. Bulk resistance R and transient capacitance $C(t)$ for PVC-DOP film

Time elapsed (Min)	Transient method			Step method		
	$V(\alpha, t)$ (V)	R (Ohm)	$C(t)$ (F)	V_o (V)	R (Ohm)	$C(t)$ (F)
10	67	8.15×10^9	8.02×10^{-8}	20	8.59×10^9	3.37×10^{-9}
20	117	8.9	1.39×10^{-7}	20	8.86	4.12
40	220	1.05×10^{10}	2.32	20	9.25	5.29
80	406	1.3	3.66	20	9.73	7.29

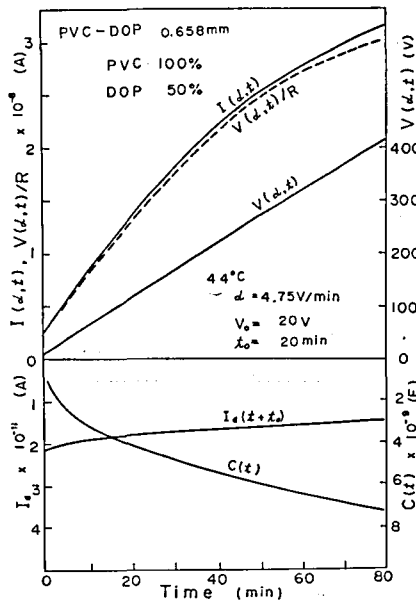


Fig. 3 Transient current and its component currents for PVC-DOP film.

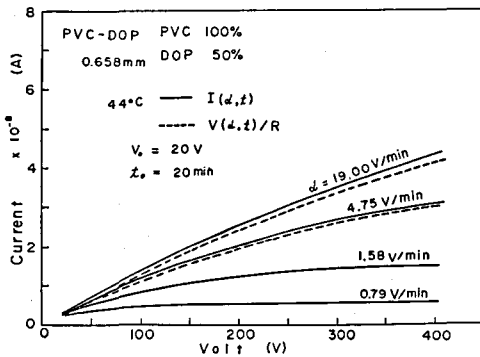


Fig. 4 Effect of α on current-voltage characteristic for the same film as Fig. 3.

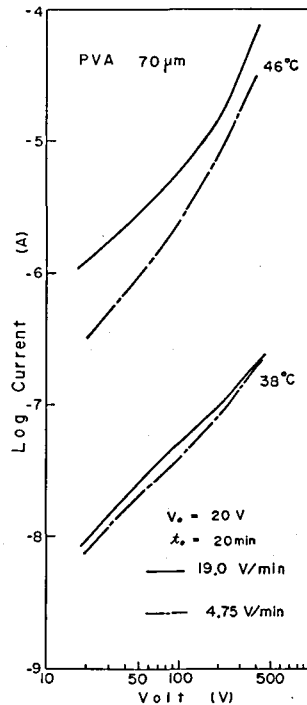


Fig. 5 Current-voltage characteristic for thin film of PVA.

the transient current at $\alpha=0.79$ volt per minute establishes the appearance of a plateau above about 200 volt.

Such non-Ohmic behavior may be mostly attributed not to the charge injection but to the ionic drift. An effective decrease in the mobile ion density may be probably produced by the clean-up effect and the resultant hetero charge accumulated near the electrodes tends to modify the internal field in the bulk. Lastly the internal field will decrease to a certain plateau value, because most of the potential drop occurs at the space-charge layer near the electrodes. This space-charge effect may be promoted very much more by the addition of plasticizer DOP.

Figure 5 shows a current-voltage characteristic in the double logarithm scale for thin film of PVA. Ohm's law is likely valid at 38°C. The current at 46°C are partly linear, but increases more than linearly with voltage, unlikely the case for PVC film. Its deviation from Ohm's law suggests the possibility of the existence of homo charge due to electron injection from the electrodes. The applied voltage is much less than breakdown voltage of PVA film.

5. Conclusion

The transient method has been found to provide a convenient method for the study of electric conduction as much as for that of dielectric behavior in polymer insulators.

The rate of voltage variation with time is so much less, within the limits of our experiments, that the polarization with shorter values of relaxation time could not be observed. On the otherhand, the contributions of linear absorption current and of conduction current has been able to be effectively separated in this way, from which the bulk resistance and the transient capacitance of specimens have been estimated. Consequently non-linear phenomena due to the space-charge accumulated and released have been observed in addition to the linear one.

This is a preliminary report. This transient method, however, seems to be a valuable tool for observation on the phenomena which are no longer able to be followed in the usual method for dielectric relaxation. It will be the subject of further research to obtain more detailed information about electric conduction and dielectric polarization in polymer insulators by this transient method.

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