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FEATURED TOPIC

$$\frac{Q(t_m)}{Q(0)} = 1 + \frac{t_m}{\tau}$$

Analysis and Elucidation of Space Charge and Electrical Conduction Behaviors of Cross-linked Polyethylene by Q(t) Method

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It is very important to understand the current that flows in a polymer material when a voltage is applied to it because it reflects the physical phenomena that occur in the material. To measure and analyze the phenomena, we have been studying the direct current integrated charge (Q(t)) method in which integrated currents are measured and analyzed as the amount of charge. Here, the ratio of the charge immediately after the application of square wave voltage Q(0) and the charge t_m after the application of voltage $Q(t_m)$ is introduced as the charge ratio $R_c=Q(t_m)/Q(0)$. The physical meaning of R_c is discussed, and it is shown that R_c can be expressed by a simple equation between the relaxation time of the material and the measured time of the material in an ideal state of electrical conduction. These relations were used to analyze the conductive state of cross-linked polyethylene.

Keywords: dielectric and insulating materials, direct current integrated charge method, relaxation time, electrical conduction

1. Introduction

Recently, efforts to develop direct current (DC) power transmission cables have been accelerated worldwide. Such cables require insulating materials characterized by high insulating performance (namely, currents do not flow easily) and low charge accumulation when a DC voltage is applied. To promote the development of insulating materials with such characteristics, Sumitomo Electric Industries, Ltd. has been studying the mechanism to improve the dielectric characteristics and screening of materials using the direct current integrated charge method (Q(t) method).

As introduced in the Sumitomo Electric Technical Review[1] before, the Q(t) method is an excellent technique that can obtain a lot of information about the dielectric characteristics with simple operation. The characteristics of the Q(t) method are attributed to the measurement of the electric charge amount by integrating the current. A conceptual diagram of the measurement circuit is shown in Fig. 1 (a).

When a square wave voltage, as shown in Fig. 1 (b), is applied to a polymer material, an instantaneous charging current flows first (see Fig. 1 (c)). Then, an absorption current, which is attributed to transfer and accumulation of the space charge, flows. Finally, a conduction current flows in a steady state. In general, these currents are measured by using different and appropriate measurement techniques depending on their characteristics. When a current is integrated to measure the charge amount (Fig. 1 (d)), the instantaneous charge amount during charging Q(0) is the product of the applied voltage and capacitance of a specimen ($Q(0)=C_sV_{dc}$), making it possible to clarify the initial value and generally grasp the subsequent changes in the absorption charge and conduction charge. For example, when a leakage current is measured after applying a high electric field to low-density polyethylene (LDPE) or cross-linked polyethylene used as an insulating material for electrical power cables for alternating current power transmission (AC-XLPE) for many hours, the current value remains inconstant even after the lapse of one day, and a decreasing trend is continuously observed.[2] If the space charge accumulates in a material depending on the temperature and applied electric field, the behavior significantly affects the current. This means that such changes can be tracked. Here, the Q(t) method was used to conduct measurement of AC-XLPE and cross-linked polyethylene for electric power cables for DC power transmission (DC-XLPE) and made a comprehensive and general evaluation of the electrical conduction phenomena, including the space charge behavior.

2. Characteristics of the Q(t) Method

Simply put, the Q(t) method used in this study aims to make an evaluation by accumulating the electric charge amount Q(t), which is an integral value of the current, in an integrating capacitor connected in series with a specimen. This method monitors the current phenomena, but measurement of the charge amount reveals different aspects of the phenomena from those of the conventional current measurement.

When a square wave voltage is applied, the current is classified based on the physical phenomena and is expressed as shown in Eq. (1).

$$\begin{aligned} Q(t) &= \int I(t)dt \\ &= \int \left[I_{disp}(t) + I_{abs}(t) + I_{cond}(t)\right]dt \quad \cdots\cdots\cdots (1) \\ &= Q_{disp}(t) + Q_{abs}(t) + Q_{cond}(t) \end{aligned}$$

Here, $I_{disp}(t)$ is the instantaneous charging current immediately after a square wave voltage is applied; $Q_{disp}(t)$ is a charge amount by an instantaneous charging current (i.e., electrode charge); $I_{abs}(t)$ is an absorption current in line with accumulation and transfer of the space charge;

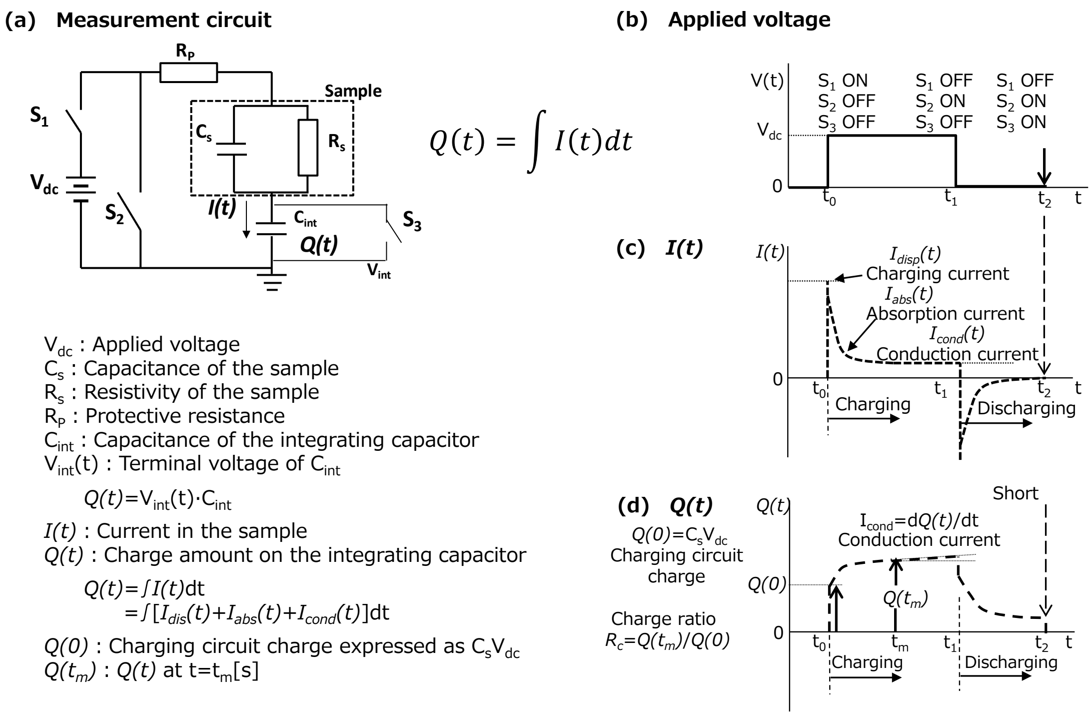

Fig. 1. (a) measurement circuit and (b) waveform of a square wave voltage of the Q(t) method; image of (c) current I(t) and (d) integrated charge amount Q(t) when a square wave voltage is applied [1]

$Q_{abs}(t)$ is an absorption charge amount; $I_{cond}(t)$ is a conduction current; and $Q_{cond}(t)$ is a conduction charge amount. The integral value of $I_{disp}(t)$ is the product between the specimen's capacitance Cs and applied voltage V_{dc} as the electrode charge. When $\int I_{disp}(t)dt = C_sV_{dc} = Q(0)$, Eq. (1) can be rewritten as Eq. (2).

$$Q(t) = Q(0) + \int [I_{abs}(t) + I_{cond}(t)]dt \quad \cdots\cdots\cdots\cdots\cdots (2)$$

In the Q(t) method, Q(0) can be clearly defined as the initial value by integrating all the current components after the startup of a square wave voltage. Meanwhile, in general, a picoammeter, which is used for techniques to measure the conduction current, measures a current by using a micro-resistor, which is connected in series with a specimen. Thus, it is difficult to measure I(0), which corresponds to Q(0). In the Q(t) method, it is essential to clearly calculate Q(0), which is the initial value of the electric charge.

To understand the data obtained by using the Q(t) method, we introduced the "charge ratio (R_c)" as a new parameter.[3] R_c is expressed as the ratio between $Q(t_m)$, which is Q(t) after the lapse of time t_m, and Q(0), which corresponds to the startup immediately after a square wave voltage is applied. Thus, R_c is defined as shown in Eq. (3).

$$R_c \equiv \frac{Q(t_m)}{Q(0)} \quad \cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots (3)$$

As indicated in Eq. (4), in which both sides of Eq. (2) are divided by Q(0), the charge ratio is "1" in the case of an ideal capacitor free from an absorption current and conduction current. It is measured in increments from "1" in line with the increase in the absorption current and conduction current components.

$$\frac{Q(t)}{Q(0)} = 1 + \frac{Q_{abs}(t) + Q_{cond}(t)}{Q(0)} \quad \cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots (4)$$

The characteristics, as shown in Table 1 (which applies to insulating materials), can be confirmed by using the Q(t) method and charge ratio R_c. Table 1 compiles the characteristics of the Q(t) method in comparison with picoammetry and the PEA method.

3. Q(t) of AC-XLPE and DC-XLPE

We conducted measurement of AC-XLPE and DC-XLPE by using the Q(t) method. Press-molded films (thickness: approximately 0.2 mm) were used as specimens. Evaluations were made for short time (600 s) and long time (repeated electric charging of 8 h per cycle). A Q(t) meter manufactured by A&D Company, Ltd. (AD-9832) and a DC power source (HMBR-30R0.4, max 30 kV, 0.4 mA) manufactured by Matsusada Precision Inc. were used.

3-1 Short-time electric charge behavior

An electric field of 10 to 100 kV/mm was applied to the same specimen. Measurement of electric charge for 600 s and discharge for 300 s was conducted from the low electric field side.[4] Vacuum drying at 60°C was conducted for 24 h before measurement to remove the decomposed residue of a cross-linking agent.

Figure 2 shows the measurement results during charging at 80°C. Time is plotted on the horizontal axis, and the charge amount Q(t) is plotted on the vertical axis. The graph shows the time change of Q(t) for each electric field applied.

Table 1. Comparison of techniques to evaluate the dielectric and insulating characteristics [1]

		Picoammetry	Direct current integrated charge method (Q(t) method)	Pulsed Electroacoustic method (PEA method)
Measurement device		Picoammeter	Q(t) meter	PEA measurement apparatus
Scope	Space charge distribution	Inadequate	Inadequate	Very suitable
	Presence of charge accumulation	Inadequate	Very suitable	Suitable
	Electrical conductivity	Very suitable	Very suitable	Inadequate
	Permittivity	Inadequate	Very suitable	Inadequate
Characteristics of the measurement device and technology		• Relatively easy to conduct measurement. • Difficult to remove noise.	• Relatively easy to conduct measurement. • Physical phenomena involving current components (static charge accumulation, conduction, space charge) can be evaluated in a comprehensive manner. • Noise is cancelled by integration. ⇒ The technique is resistant to noise.	• Charge accumulation can be observed in space and time. • An advanced technique is required for signal processing. • The voltage application system and the measurement system are insulated. This helps minimize the influence of dielectric breakdown.

As shown in Fig. 2, AC-XLPE exhibits a startup of the charge ratio when a low electric field is applied. Meanwhile, the charge ratio of DC-XLPE is almost 1 even in a high electric field, indicating that changes are very small.

3-2 Long-time electric charge behavior

The previous section discussed the electric field dependence of Q(t) in short time. In general, electric power equipment, such as cables, is used continuously for decades. Thus, it is important to evaluate the behavior during long-time charging. Here, we applied an electric field of 60 kV/mm at 60°C and observed changes in the Q(t) behavior during several charging cycles (with charging on for 8 h and off for 16 h per cycle).[5] Measurement examples of AC-XLPE and DC-XLPE (only during charging) are compiled in Fig. 3. It is noteworthy that, in the case of AC-XLPE, the startup of the charge ratio in the second and subsequent cycles shifts and converges on the long-time side compared to the charging in the first cycle. In the case of DC-XLPE, the startup of the charge ratio is

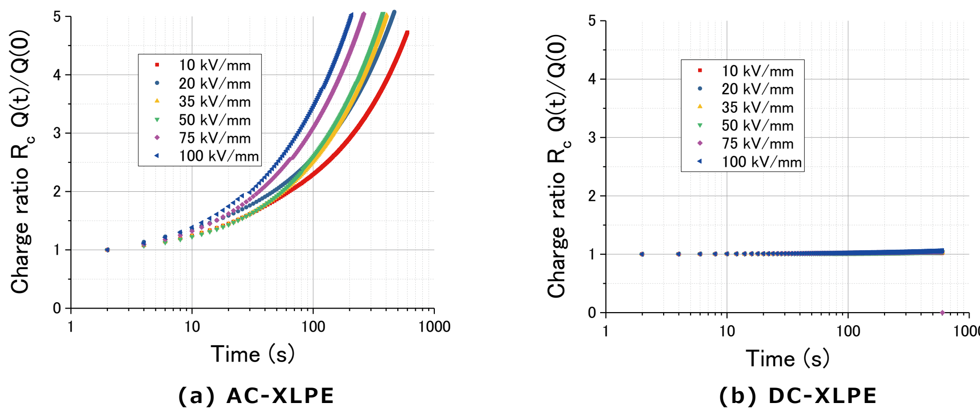

Fig. 2. Change over time (600 s) of the charge ratio of AC-XLPE and DC-XLPE measured in an electric field. (80°C, 10–100 kV/mm) [4]

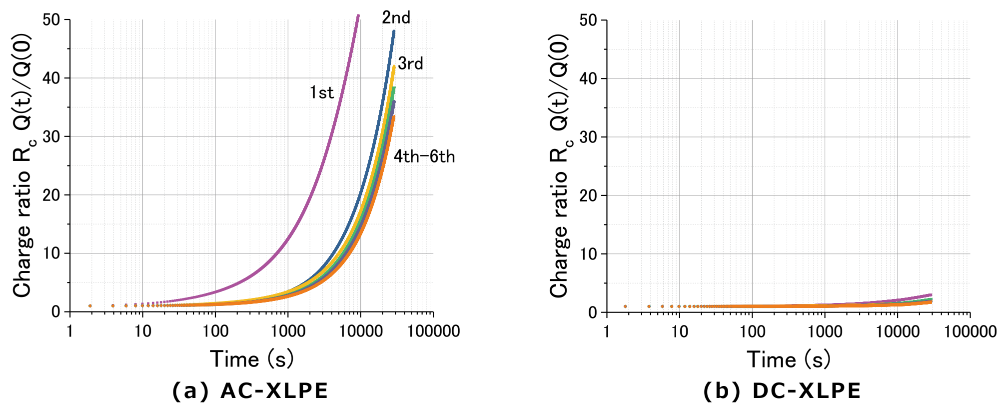

Fig. 3. Change over time of the long-time charge ratio of AC-XLPE and DC-XLPE measured in an electric field (60°C, 60 kV/mm)
(by repeating charging of 8 h per cycle six times) [5]

small even when a continuous charging of 8 h is performed. The influence of repetition of the charging cycles is considered to be small.

To understand the differences in the Q(t) behavior, the meaning of the "charge ratio" is discussed in the next section.

4. Meaning of the "Charge Ratio"

The meaning of the charge ratio is discussed in detail in the references.[3] Here, an overview is provided. As described above, the meaning of charge ratio R_c can be easily understood by dividing both sides of Eq. (2) by Q(0). That is, as indicated by Eq. (4), $R_c = 1$ applies based only on the electrode charge by an instantaneous charging current (i.e., an ideal capacitor). In line with an increase in an absorption current and conduction current, R_c is measured in increments from "1."

The meaning of R_c is discussed from the viewpoint of electromagnetism. Equation (5) is obtained by using relational expressions based on Ohm's law ($J = \kappa \cdot E_{dc}$), including $dQ(t)/dt = I(t)$, $V_{dc} = E_{dc} \cdot d$, $Q(0) = C_s \cdot V_{dc}$, and $C_s = \varepsilon S/d$ (J: current density, E_{dc}: applied electric field, V_{dc}: applied voltage, κ: electrical conductivity of a specimen, d: thickness of a specimen, S: electrode area, C_s: capacitance of a specimen, ε: permittivity of a specimen).

$$\frac{dQ(t)}{dt} = \frac{\kappa}{\varepsilon} \cdot Q(0) \quad \cdots\cdots (5)$$

The dQ(t)/dt means the amount of change of Q(t) in the infinitesimal time range. When changes with the lapse of time (t) of Q(t) are linear, it can be substituted with $\Delta Q/\Delta t$. Given that dielectric relaxation time $\tau = \varepsilon/\kappa$, Eq. (6) is obtained by expressing Q(t) in time (t_m) as $Q(t_m)$ (range of Δt: from 0 to t_m) and summarizing the relationship.

$$R_c = \frac{Q(t_m)}{Q(0)} = 1 + \frac{t_m}{\tau} \quad \cdots\cdots (6)$$

As a result, R_c can be expressed as a simple relational expression of t_m and τ, as shown in Eq. (6). As is understood from the derivation process, it is necessary to take notice that Q(t) is linear in Eq. (6), namely, the relationship is established in an ideal electrical conduction state. Thus, if an experiment value matches Eq. (6), only a constant conduction current flowed in the experiment.

5. Discussion

We conducted short-time charging (600 s) and long-time charging (on for 8 h/off for 16 h × 6 cycles) by using cross-linked polyethylene, which is used for electric power cables for AC and DC power transmission (AC-XLPE and DC-XLPE). The results showed the following characteristics.

(1) When charging was conducted while changing the electric field applied at 80°C, the startup of the charge ratio with the lapse of time was observed from the low electric field for AC-XLPE. The higher the electric field, the faster and larger the startup. For DC-XLPE, no increase in the charge ratio was observed in the 600 s range even in a high electric field. Changes were small for an increase in the electric field.

(2) As part of evaluation of the conduction behavior during long-time charging, long-time charging (on for 8 h/off for 16 h per cycle) was conducted under the conditions of 60°C and 60 kV/mm. For DC-XLPE, the increase in the charge ratio with the lapse of time was suppressed, and the difference by repetition of charging cycles was small. For AC-XLPE, a significant increase in the charge ratio was observed with the lapse of time. As the charging cycle was repeated, the curve of the charge ratio tended to shift and converge on the long-time side.

Notably, curve fitting was performed by using Eq. (6) to understand the changes in the conduction behavior of AC-XLPE.

Figure 4 (a) shows the consistency with the theoretical equation by extracting data of the first and sixth cycles of AC-XLPE in Fig. 3 (A) and assigning appropriate τ in Eq. (6). For the first curve, consistency between a theoretical curve and experiment value could not be ensured no matter how τ was chosen. For the sixth curve, the theoretical curve was highly consistent with the experiment value when an appropriate τ (here, 900 s) was chosen. As discussed above, Eq. (6) is considered to express the ideal conduction state based on its derivation process. Thus, the results are deemed to show the influence of the current components other than a conduction current. That is, the

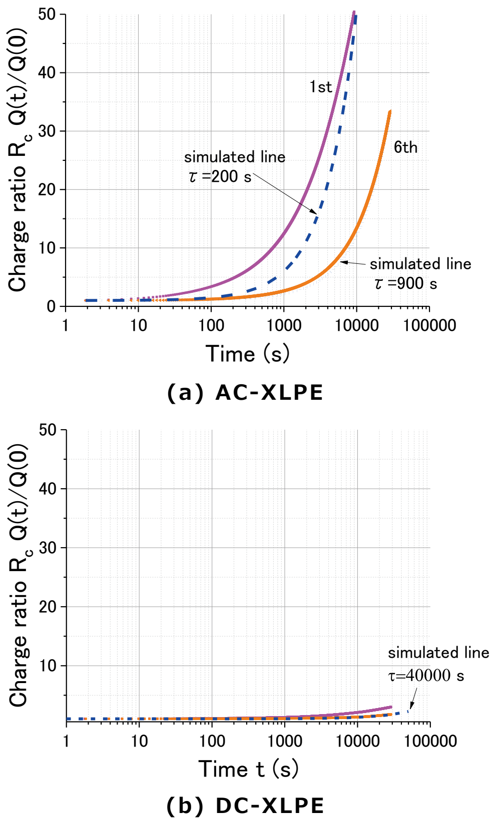

Fig. 4. Experiment data of the charge ratio of AC-XLPE and DC-XLPE (first and sixth curves) and theoretical curves calculated by Eq (6) [5]

first curve does not conform to the theoretical curve probably because the absorption current components other than the conduction current components are high and almost only the conduction current components are present for the sixth curve with absorption current components almost absent.

For DC-XLPE (Fig. 4 (b)), the condition is close to a steady state from the initial phase of charging. Namely, only a conduction current is present with an absorption current almost absent. This is different from AC-XLPE.

Based on these characteristics, the space charge behavior and electrical conduction behavior of AC-XLPE and DC-XLPE are considered.

Two characteristics are observed for AC-XLPE.

① The current increases due to the strong influence of an applied electric field.

② When a constant electric field is applied, the current decreases gradually after repetition of long-time charging cycles. Eventually, a conduction current becomes dominant.

For example, when the long-time conduction current measurement[2] by Ghorbani et al. and measurement results based on the PEA method[6]-[8] by Tanaka et al. are taken into account, it is considered that an absorption current generated by accumulation and transfer of the space charge, which is observed in the initial phase of charging, decreases due to long-time charging or its repetition and a steady state is eventually reached with a conduction current being dominant.

For DC-XLPE, the condition is considered to be close to a steady state, with only a conduction current present, from the initial phase of charging.

6. Conclusion

This paper described the charge ratio, which is a newly introduced parameter, and the method of interpretation in line with the significance of measuring the weak current, which flows in a specimen when a voltage is applied, as the charge amount by using the Q(t) method. We explained that the charge ratio can be expressed using a simple relational expression of the relaxation time of a specimen and the measurement time (Eq. (6)). The relation was applied to cross-linked polyethylene to analyze data. We also explained that comparison between an ideal curve that is obtained by assigning appropriate relaxation time to Eq. (6), which expresses an ideal electrical conduction, and a measured value enables estimation of the electrical conduction of a specimen.

This technique was used to analyze the electrical conduction characteristics of cross-linked polyethylene and confirmed consistency with the findings obtained in the past based on the weak current measurement and PEA method. We confirmed the changes in the electrical conduction behavior during long-time or repeated long-time application of voltage. That is, in the initial phase of application of voltage, an absorption current, which has a significant impact, decreases with the lapse of time, eventually leading to a steady state in which a conduction current becomes dominant. Compared to AC-XLPE, DC-XLPE is characterized by small changes in charge and current. That is, the state is close to a steady state from the initial phase of application of voltage and a conduction current is very small.

Regarding the model based on Eq. (6), which we used in this paper, we will verify the validity of concurrent measurement using the Q(t) method and PEA method to increase the accuracy of analysis.

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