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Breakdown and space charge formation in polyimide film under DC high stress at various temperatures

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Abstract. Relationship between breakdown strength and space charge formation in polyimide film under dc high stress at various temperatures is investigated using pulsed electro-acoustic (PEA) method. Some typical results of the space charge observations show that hetero space charges are always found before breakdown. The amount of the hetero charges increase with increase of temperature or increase of applied electric field. Since the enhancement of the internal electric field in the sample by the accumulation of the hetero charges is not so large, the accumulation doesn’t seem to be an immediate cause of breakdown. However since it is always observed before breakdown, it may be predictive information for breakdown. In a certain case, the breakdown occurs after voltage application for few hours. However, while we give an interval of short circuit condition after observing the hetero space charge under dc stress, the total voltage application time to breakdown is almost the same to the case without the interval. It means that the hetero space charge generation may show a kind of degradation of the material.

1. Introduction
Since a polyimide film shows a high dielectric strength even at high temperature, it is used as an insulating material for various electric and electronic devices, such as a flexible printed-circuit board in mobile phone, sheath of winding wire for motors, etc. Furthermore, recent current of downsizing and cost reduction demands it to be reduced its thickness. It means that the polyimide film is demanded to show a good performance as an insulating material under high electric field even under various sever condition, such as at high temperature with high humidity. Especially, a breakdown characteristic is one of important factors to estimate the material under such condition. On the other hand, the space charge accumulation sometimes strongly affects to dc breakdown strength. For example, so called, “packet-like charge” generation in low density polyethylene under dc high electric field of more than 100 kV/mm affects its breakdown strength [1]. However, the relationship between the breakdown properties and space charge accumulation under high dc electric field in other materials has not been clear yet. Therefore, we have been investigating the relationship in polyimide film. To investigate the relationship, the pulsed electro-acoustic (PEA) method [2] is used. The PEA method is useful technique to investigate the characteristics under high electric field [3] at high temperature [4, 5]. In this paper, authors introduce the space charge accumulation characteristics of polyimide film under dc high electric field near breakdown strength at various temperatures.

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2. Samples and experimental procedure

2.1. Samples
A commercially available polyimide film, “Kapton® type H” supplied from DU PONT-TORAY CO., Ltd, is used for all experiments. Nominally 125 and 75 µm thick samples are used for space charge measurement and breakdown test, respectively. The breakdown strengths at room temperature are described on a catalogue of merchandise as 207 and 339 kV/mm for 125 and 75 µm thick samples, respectively.

2.2. Measurement system
Figure 1 shows a PEA measurement system for high temperature [4, 5]. Since the system has a piezoelectric sensor of LiNbO₃, which shows a stable performance even at relatively high temperature, the system is applicable to the measurement condition up to about 100 °C. In an upper electrode unit, the sample is immersed into silicone oil to keep temperature stable and to avoid a flash over discharge. Temperature in the upper electrode unit is heated using a computer controlled heater put around the unit. The temperature is measured using a thermo couple put into the silicone oil. The maximum applied dc stress to the sample is about positive or negative 20 kV. Since the thicknesses of samples are nominally 125 and 75 µm, the maximum applied electric fields are about 160 and 260 kV/mm, respectively. The dc voltage is applied to the sample using computer controlled high voltage amplifier (HEOPT-20B10, Matsusada Precision Inc.). The dc high voltages are applied to the sample through a semi-con layer put on the sample. The semi-con layer is used to match the acoustic impedance of the sample to that of upper electrode [6]. The semi-con layer used for this experiment is made for a commercial XLPE power cable, and it is supplied by a cable company. Unfortunately, the detail information of the layer is undocumented. However, the surface of the layer is smooth enough to make a good contact with the sample and it is available even at high temperature of 80 °C. Since the sample is used without any evaporated metal electrodes on the surfaces, the interfacial condition at upper electrode is different from the lower one. The spatial resolution of the measurement using this system is about 10 - 20 µm depending on measurement temperature. It means that the relative spatial resolution for the sample with thickness of 125 µm is about 8 - 16 %. If we attempt to measure the space charge distribution in 75 µm thick sample, the resolution is not enough. That is reason why the result of space charge measurement for 75 µm thick sample is not reported on this paper. However, the breakdown test was also carried out using this system to match the measurement condition of it to that for the space charge measurement. The measurement principle and typical procedure are described elsewhere [2, 3, 6].
2.3. Measurement procedure

After putting the sample in the system, temperature is gradually increased with a heating rate of about 1 °C/minute up to a target temperature, and then the temperature is kept for at least 30 minutes. The target temperatures in the experiments are 30, 40, 50, 60 and 80 °C. After that, a target voltage is applied to the sample. Since a rapid voltage rise sometimes leads a flashover on the surface of the sample, the voltage is increased with a rate of 3 kV/s using a computer controlled power supply to avoid such flashover. When the voltage reaches to a target voltage, the voltage is kept for at least 2 hours if the breakdown doesn’t occur within 2 hours. When the breakdown occurs, the voltage is immediately shorted automatically, and the measurement is finished with recording the time to breakdown. The target voltages depend on target electric fields and the sample thickness. In the experiments, the target electric fields are 110, 130 and 150 kV/mm for 125 µm thick sample, and 170, 190 and 210 kV/mm for 75 µm thick sample. The PEA measurements are carried out with every 5 seconds automatically using computer. The averaged signal made from 120 times shot of the pulse voltage is obtained as each datum. An adequate data processing including calibration process makes the voltage signal to a charge distribution. In addition to the above mentioned experiments, an interruption of voltage application and a repeated voltage application tests are carried out. Details of the tests will be mentioned later with the each result.

3. Results and discussion

3.1. Typical result

At first, a typical measurement result and how to display the results are introduced. Figure 2 (a) shows a typical measurement result of space charge distribution. This result is obtained by measuring 125 µm thick Kapton® film under applied electric field of 150 kV/mm at 80 °C. In this figure, firstly (just after beginning of voltage application) and finally (just before breakdown) observed data are drawn in red and blue lines. The left and right peaks show the induced charges on cathode and anode, respectively. As shown in figure 2 (a), positive charge is observed near the cathode electrode. Such kind of space charge accumulated near the electrode of opposite polarity is generally called a “hetero” space charge.

Figure 2 (b) shows electric field distributions in the sample at various times. The distributions are obtained by integral calculation using the measured space charge distributions shown in figure 2 (a). In this figure, the initial applied electric field has a flat shape and the value of the flat electric field is coincident with the applied electric field of 150 kV/mm. When the space charges accumulate in the bulk, the electric field is distorted by the charge accumulation. Therefore, the distribution has a maximum peak. In the case of measurement in LDPE, the maximum electric field show a significant meaning for its breakdown. Therefore, the maximum electric field in Kapton® is also calculated for each datum. As shown in figure 2 (b), the maximum electric field at just before breakdown (blue line) is about 190 kV/mm, and it means that the electric field is enhanced for about 27 % by the charge accumulation.

Using the PEA measurement, above mentioned profiles are originally obtained. However, since many time dependent profiles are drawn together in this figure, it is hard to recognise the time dependent properties. Therefore, authors use a colour chart, as shown in figure 2 (c), to show the measurement results clearly. The charge density is expressed using a colour bar beside the figure. In this figure, red and blue colours indicate positive and negative charge densities, respectively. The horizontal and vertical axes stand for position and time after voltage application. In this case, breakdown was observed at 11 minutes after beginning of voltage application. It is found that small amounts of positive and negative charges appear near anode and cathode electrodes, respectively, immediately after the voltage is applied to the sample. Such charges accumulated near the same polarity to the electrodes are “homo” space charges, and the charges are recognised, in many cases, as the injected
charges from electrodes. On the other hand, positive and negative “hetero” space charges are also observed about few minutes after the beginning of voltage application. The amount of hetero charges gradually increases, and then breakdown occurs in the sample. Since the colour chart is convenient to show the time dependence as shown in figure 2 (c), the space charge measurement results are shown using the colour charts in following explanations.

![Image](image_url)

**Figure 2.** Typical measurement results of (a) space charge profile, (b) electric field distribution and (c) time dependent space charge behaviour in Kapton® under applied electric field of 150 kV/mm at 80 °C.

3.2. Electric field and temperature dependences of the space charge behaviours

Figure 3 shows applied electric field and temperature dependences of the space charge behaviours in Kapton®. The results are displayed in orders of applied electric fields and temperatures in vertical and horizontal axes, respectively. The electric fields of 110, 130, 150 and negative 110 kV/mm are applied to the 125 μm thick sample at 30, 40, 50, 60 and 80 °C. Measurements were carried out for at least 2 times in each condition and one of them is displayed here. In the cases that any remarkable space charge and a breakdown were not observed at least within 2 hours (240 min.), the results are shown with a sign of “without BD”. It is clearly found that the breakdown is not observed under the electric field of 100 kV/mm at 30, 40, 50 °C, breakdown is not observed at least within 2 hours. On the other hand, under the field of 110 kV/mm at 60 °C, the breakdown is observed at 200 minutes later. In this case, positive hetero charges are observed from about 100 minutes later and its amount of charges gradually increase, and then finally the breakdown occurs. A small amount of hetero negative charge also observed before the breakdown. At 80 °C, such hetero space charges are also observed and the breakdown occurs at 120 minutes later. It is clear that, at the higher temperature, the larger amounts of hetero charges appears earlier before breakdown, and the breakdown also occurs earlier. When the electric field of 130 kV/mm is applied to the sample, the breakdowns occur at 213, 133, 83 and 50
minutes later at 40, 50, 60 and 80 °C, respectively, while it is not observed for 416 minutes at 30 °C. In the case that the breakdown occurs, the hetero space charges are observed before the breakdown. Under the electric field of 150 kV/mm, the breakdown is observed at all temperatures at earlier time with larger amounts of hetero space charges before breakdown. Judging from these results, under the higher electric field at the higher temperature, the breakdown occurs at the earlier time after the beginning of the voltage application with having the larger hetero space charges before the breakdown. When even the negative electric field of 110 kV/mm is applied to the sample, the characteristics of the breakdown and the space charge accumulation are almost the same as the case of positive electric field application. However, the amount of the negative hetero space charge is smaller than that observed under positive electric field application. Anyway, an especially remarkable property in these results is the hetero space charges are always observed before the breakdown.

3.3. Relationship between temperature and time to breakdown

Figure 4 shows a relationship between temperature and time to breakdown. In this figure, the data obtained using 75 µm thick samples are also plotted. All data are averaged value form at least two times measurements. It is found that the time to breakdown shows a linear decrease with increase of temperature under the applied electric field. Since the vertical axis is described using logarithm scale, the time to breakdown decreases exponentially with increase of temperature. Most of all approximated fitting lines show the almost same slope although there is exception. Unfortunately, the data taken under electric field of 110 kV/mm is too short to estimate the above mentioned point. It is necessary to
further detailed investigation. However, it is clear when the sample is used at the higher temperature under the higher dc electric field, the risk of breakdown increases within the shorter time.

![Figure 5. Relationship between temperature and time to breakdown.](image)

3.4. **Relationship maximum electric field and temperature**

Figure 6 shows the relationship between the maximum electric field just before the breakdown and the temperature. Judging from the results, there seems be no significant relationship between them. In the case of LDPE, the breakdown seems to occur at almost the same as the enhanced maximum electric field, and it shows a direct affection of the space charge to the breakdown phenomena. On the other hand, there is no such property in this sample. It doesn’t seem that the enhancement of the electric field is high enough to affect the breakdown strength. It means that the space charge formed in the Kapton® is not immediate cause of the breakdown but it may be a result of degradation for breakdown.

![Figure 6. Maximum electric field just before the breakdown at various temperatures under various applied electric field.](image)
3.5. Interrupting and repeating voltage application test

To show the space charge accumulation is a result of irreversible degradation, the interrupting voltage application test is carried out. In this experiment, the voltage is applied to the sample (130 kV/mm) at 60 °C for 50 minutes, when the hetero charge accumulation is observed clearly. Then the circuit is once shortened. After the interval of the short circuit condition for 12 hours, the same voltage is applied to the sample again. The result is shown in figure 7. As shown in figure 7, the accumulated hetero space charge finally disappeared at 12 hours later after circuit is shortened. However, the hetero space charge appears again immediately after the second application of the voltage while it takes for about more than 40 minutes to observe it clearly in the first voltage application. It means that the hetero space charge accumulation depends on the history of the high dc voltage application. After about 42 minutes in the second voltage application, the breakdown occurred. As shown in figure 3, it takes about 83 minutes for the breakdown to be observed with a continuous voltage application (130 kV/mm) at 60 °C. It seems that a total of the first and the second voltage application time to breakdown (50 + 42 = 92 minutes) is close to that of the continuous voltage application (83 minutes).

To show the consequence of dc high voltage application history on the hetero space charge accumulation, a repeating voltage application test is also carried out. In this experiment, the voltage is applied to the sample repeatedly with interval of short circuit condition. At first, the voltage (130 kV/mm) is applied to the sample at 60 °C for 10 minutes, and then the circuit is shortened for 12 minutes. The same voltage is applied again after the short circuit interval for 10 minutes, and then the circuit is shorted again. Such a cycle is repeatedly carried out until the breakdown occurs. Figure 8 (a) shows the result of the repeated voltage application test. In this figure, only the observations under the voltage application are connected. For ease in comparison, the continuous measurement result under the same condition is shown in figure 8 (b). As shown in the figures, the short circuit interval procedure doesn’t affect the space charge accumulation process at all. Judging from the result, the high voltage application even for 10 minutes may give a kind of irreversible damage to the sample and the damage accumulation may finally leads the breakdown. Furthermore, the appearance of the hetero space charges may be observed as a result of the damage accumulation. Since the experimental results are not enough for a statistic analysis of breakdown characteristics, it is hard to say the time to breakdown numerically. Further experimental works, however, may show the relationship between the amount of accumulated hetero space charges and time to breakdown, clearly. Another interesting point
of view is how short term of the dc voltage application gives the damage to the sample. Polyimide film
is sometimes used for an insulating material which is applied with short, several tens ms order, high
voltage pulses intermittently. For example, in inverter driving devices, the insulating material is
exposed to high voltage of inverter surge intermittently. If even such short pulse voltage gives the
damage to the polyimide film, the damage gradually accumulates and finally it leads the breakdown.
Therefore, we think that further investigation of the hetero space charge accumulation in polyimide
film under high dc voltage is important to understand the characteristics of the actual devices.

4. Conclusion
Space charge accumulation characteristics in polyimide film under high dc electric field at various
temperatures are investigated using pulsed electro-acoustic (PEA) method. The results are summarized
as follows:
(1) Hetero space charges are always observed before breakdown occurs in the sample.
(2) Under the higher applied electric fields at the higher temperature, the larger amount of hetero
space charges are observed earlier after the voltage application.
(3) Voltage application with a long interval of short circuit condition doesn’t recover the space
charge accumulation properties. It shows the irreversible change of electric property must happen
in the sample.
(4) Intermittently voltage application with relatively short duration (10 minutes) also leads the
irreversible change of electric property in the sample.

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