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The Study of Electroluminescence and Reliability of Polyimide Films in High dc Fields

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Electroluminescence (EL) intensity of the polyimide (PI) films was tested under dc high electric field by home-made experimental device. The results showed that the EL intensity of PI films increased along with the electric field. EL intensity is approximately to background intensity when the electric-field intensity was less than 2.00 MV/cm. EL intensity increases along with increasing the electric field when electric-field intensity greater than 2.00 MV/cm. When electric-field at 2.80 MV/cm, EL intensity increasing strongly suggests that the excitation process related to hot electrons accelerated by the field approaching a critical threshold. Meanwhile, this work elaborates a method to deal with identical samples get different experimental data by using Weibull distribution method, and the concept of the reliability was presented. The nine groups of EL experimental data were analyzed, and the result showed that the lifetime of mid-value (t = 164.9 min). Mid-value of the breakdown field is E = 2.76 MV/cm.
Keywords: PI films, electroluminescence, breakdown, reliability, Weibull distribution.

1. INTRODUCTION

As an important polymer material, polyimide (PI) has been extensively applied in many areas, because of their unique electrical properties, radiation resistance, excellently mechanical properties, thermo-oxidative stability, high radiation, solvent resistance, and high mechanical strength [1]. Notably, the dielectric properties of polyimides, such as high breakdown voltage, low dielectric constant [2 – 4], and the promising electrical and dielectric properties and heat resistance ability of PI films have opened the way to its practical application as electrical insulating materials. Nowadays, the dielectric breakdown of PI has already received much attention since their development for electrical insulation and their intensive use in industrial applications [5, 6].

In recent years, more and more scholars started to investigate the breakdown fields of PI films, but among the large range of values found in the literature, the breakdown fields of PI films were different [7 – 9]. Even though there is a batch of identical samples, it is relatively difficult to identify which data is more accurate and reliable. Therefore, this paper solved the problem, the concept of the reliability was presented, the analysis can avoid the breakdown, the reliability could judge stability of the material, and direction selection of suitable materials, thus, it is a great research value. Analyzing reliability of experimental data usually uses normal distribution method, lognormal distribution method, exponential distribution method and Weibull distribution method. Weibull distribution method was superior to others, because that it apply to the partial failure case causes loss of function, and it also used to probability statistics of small sample [10, 11]. In addition, Weibull distribution is widely used in a variety of reliability applications, such as, electrical engineering, civil engineering, mechanical engineering, and medical and pharmaceutical fields [12].

Meanwhile, Weibull distribution is used to decide breakdown data that has been widely recognized [13 – 15]. L.li et al. point that three-parameter Weibull distribution cannot give better fit than two-parameter Weibull distribution for breakdown behavior of all PI films. Therefore, investigated in this paper will be conducted by using two-parameter Weibull distribution [16].

Although there are many literatures about the breakdown of PI films, Weibull distribution method applied to calculate reliability of PI breakdown field by using EL method, which is rarely reported. EL is the phenomenon of electrical energy transformed into light energy, and it released energy in the form of light. EL of polymeric materials has attracted much attention since the light emission from the thin film of polymers [6 – 8], such as PI was demonstrated. It is used to analyse initiation of the breakdown for material [17 – 19]. EL which was used to explore carrier injection mechanisms and the aging properties of polymer is different fromm other measure methods, because it not only can study the electric breakdown, but also could study the relationship between EL intensity and dc electric field of PI films [20 – 22]. Because electroluminescence is associated with ageing, and the EL onset would signal the onset of ageing whatever the excitation mechanism [23]. Therefore, in this paper, the relationship between light intensity and electric field was illustrated, and breakdown field and the lifetime of PI films were investigated by EL under room temperature. Finally, the results were analyzed the results using two-parameter Weibull distribution method.

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2. EXPERIMENTAL

2.1. Materials

Specimens were original polyimide films with a thickness of 15 μm supplied by 3M Corporation. The films were cut into 60 × 60 mm, and gold electrodes with 40 mm diameter were sputter-coated on both sides. The thickness of upper gold electrode is about 30 nm and sheet resistances are about 9 Ω. In order to remove residual charges, the specimens were placed in a thermostatic oven at 80 °C and dried for 24 hours.

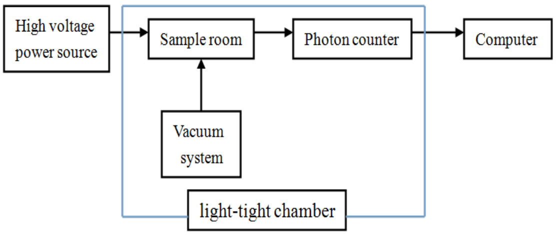

Fig. 1. Electroluminescence experimental system

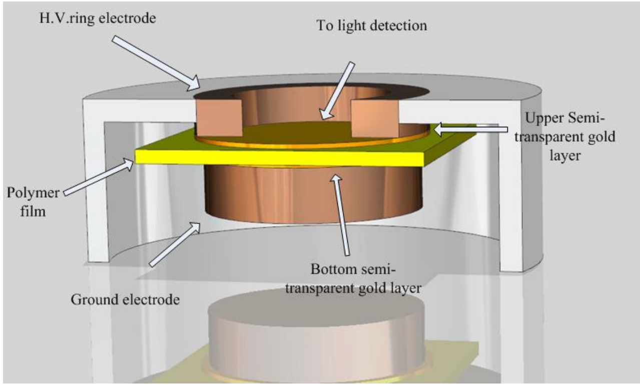

Fig. 2. Detect of EL from PI films under a uniform electric field

2.2. El experimental device and system

The experiment used the self-designed experimental devices, as shown in Fig. 1. EL characteristic curves were registered under dc voltage excitation by using an adjustable DC voltage source (DWN153-5AC, Tianjin, China). EL intensity is feeble and the power is commonly under 10^{-16} W [24], and thus, the experimental device in a light-tight chamber. The sample was mounted in a vacuum chamber connected to a turbo molecular pump system allowing to work down to $< 10^{-4}$ Pa. The integral light emission was measured by using a photon counter (R943, Hamamamasu, Japan).

The film was sandwiched between two brass flat electrodes which can provide a uniform electric field, the upper electrode is a ring of the same diameter, and a plastic cover was used to detected only of the light coming from the film, the simple arrangement is shown in Fig. 2. The applied voltage can reach 10 kV that proper configuration had to be used in order to avoid any surface discharges [25]. Gold electrodes were sputter-coated on both surfaces in order to avoid sample heating when vacuum evaporation was used [26].

2.3. Experimental procedure

EL experiments were performed under dc stress, from 550 V to the breakdown voltage. The voltage was increased by steps of a few hundred volts. Keeping few minutes at each voltage, luminescence measurements taken after 10 min were chosen as an approach of the steady state values in order to plot a characteristic curve.

3. EXPERIMENTAL RESULTS AND DISCUSSION

3.1. Relationship between EL intensity and dc electric field

The EL intensity characteristic curves of PI films are shown in Fig. 3 and Fig. 4.

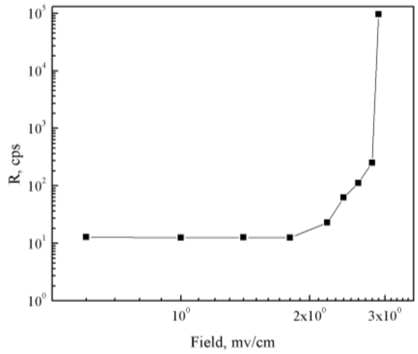

Fig. 3. Relationship between EL intensity and dc electric field of PI films

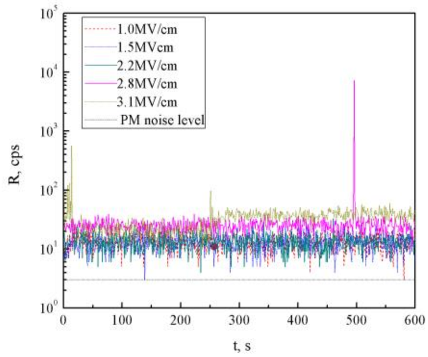

Fig. 4. EL intensity of PI films under different dc electric field

The experiment used plate electrode, which applied a uniform electric field on the sample, and injected carrier through the electrode. As shown in Fig. 3 that the breakdown field of PI films was 2.8 MV/cm. EL intensity is approximately to background intensity when electric-field intensity was less than 2.00 MV/cm. EL intensity increased along with increasing of the electric field when electric-field intensity was greater than 2.00 MV/cm. Continuing to increase the electric field, EL intensity was increasing, which indicated that the speed and energy of the carrier were changed in the distribution. Electric-field at 2.80 MV/cm, EL intensity increasing strongly suggests that the excitation process related to hot electrons accelerated by the field approaching a critical threshold. As shown in Fig. 4, we may conclude that injected charges in PI films make a significant contribution to the electrical breakdown of PI films. After the peak value of EL no long tail appeared, which indicated that a larger number of plus and minus carriers produced by high field electron compact ionization quickly counteracted on electrodes, recombined or carriers had a lower probability of luminescence through themselves counteraction in bulk but higher probability on electrodes, or the probability of releasing energy through non-eradicated transition is high.

Based on carrier injection mechanisms in high field, the two theories, field emission (Fowler-Nordheim) [27 – 29] and hot electron effects emerged. The efficiency of the field emission depends on the height of interface potential barrier. The efficiency of the hot electron effects depends on charge accumulation of an interface layer. Fowler-Nordheim could explain the character of the injected carrier [30]. Achieving injection of positive and negative charge carriers from opposite electrodes, capture of oppositely charged carriers within material can result in photon emission [31].

Table 1. The calculation of lifetime and breakdown fields for the PI films

Serial number of failure	Cumulative failure probability *F(t)*, %	Lifetime *t*, min	Pre-breakdown fields *E*, MV/cm
1	10	75	2.0
2	20	90	2.2
3	30	100	2.2
4	40	120	2.4
5	50	150	2.8
6	60	163	2.8
7	70	214	3.1
8	80	320	3.5
9	90	350	3.7

In this experience, the breakdown threshold fields under DC are physical thresholds corresponding to hot electron effects. Because electroluminescence is correlated with a drastic change in the conduction process, and current and electroluminescence are proportional to each other above this field value. The thresholds with a physical meaning only can be derived on the basis of the experimental correlation existing between light emission and the excitation component. AC voltage is not possible to correlate EL with excitation component. Thus, this has been realized by DC voltage because electroluminescence is correlated with a drastic change in the conduction process. Last, the breakdown field values of PI were different, as shown in Table 1. It appears as relatively complicated to establish which data is more accurate and more reliable. Therefore, Weibull distribution was proposed to solve this problem.

3.2. Statistics and checkout of the experimental data

The breakdown field has been calculated by using the relation:

$$E = \frac{U}{d}, \tag{1}$$

where U is the breakdown voltage, d is PI film thickness.

Experimental data were analyzed by using the Weibull distribution function,

$$F(t_i) = 1 - e^{-(\frac{t}{\eta})^m}, \tag{2}$$

where $F(x)$ is the cumulative probability of failure, m is the shape parameter, η is a scale parameter. Using the Eq. 2, the following equation was obtained:

$$\ln\left[\ln\frac{1}{1-F(t_i)}\right] = m\ln t_i - m\ln\eta; \tag{3}$$

$$\begin{cases} y_i = \ln\left[\ln\frac{1}{1-F(t_i)}\right] \\ x_i = \ln t_i \\ a = -m\ln\eta \\ b = m \end{cases}; \tag{4}$$

$$y_i = a + bx_i. \tag{5}$$

If t_i obeys Weibull distribution, $\{x_i, y_i\}$ showed a linear relationship, solving a and b, as follows:

$$m = b; \tag{6}$$

$$\eta = e^{(-\frac{a}{b})}. \tag{7}$$

The cumulative failure probability of Weibull distribution [18, 19] can be calculated from the following formula:

$$F(t) = \frac{i}{n+1}, \tag{8}$$

where, i = 1, 2, 3, ... , 9, $n = 9$

The lifetime (x_i^t, y_i^t) and breakdown field strength (x_i^E, y_i^E) can be calculated using the Eq. 4, respectively.

The cumulative failure probabilities of the lifetime and breakdown field of PI films, (x_i^t, y_i^t) and (x_i^E, y_i^E), are shown in Fig. 5 and Fig. 6.

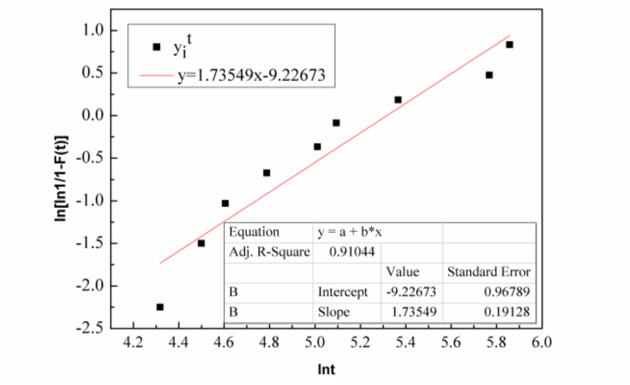

Fig. 5. Cumulative failure probabilities of the lifetime for PI films

The lifetime and the breakdown field of PI films were calculated by using Origin 8.0, and we could get the linearly dependent coefficient, which was 0.92 and 0.91, respectively. The values of independent coefficient are equal to reliable value. The results show that two groups of experimental data have a high reliability. In the industrial, the product is relatively stable when they have a high reliability.

The equation is obtained by using the previous illustration,

$$y_t = 1.73549x_t - 9.22673; \tag{9}$$

$$y_E = 4.38228x_E - 4.82241; \tag{10}$$

Solve the Eq. 6 and Eq. 7:

$$m^t = 1.735449, m^E = 4.38228\,; \tag{11}$$

$$\eta^t = 203.6695, \eta^E = 3.00547\,. \tag{12}$$

Plug Eq. 11 and Eq. 12 into the Eq. 2:

$$F(\mathrm{t}) = 1 - \mathrm{e}^{-\left(\frac{t}{203.6695}\right)^{1.73549}} \tag{13}$$

$$F(E) = 1 - e^{-\left(\frac{t}{3.00547}\right)^{4.38228}}\,. \tag{14}$$

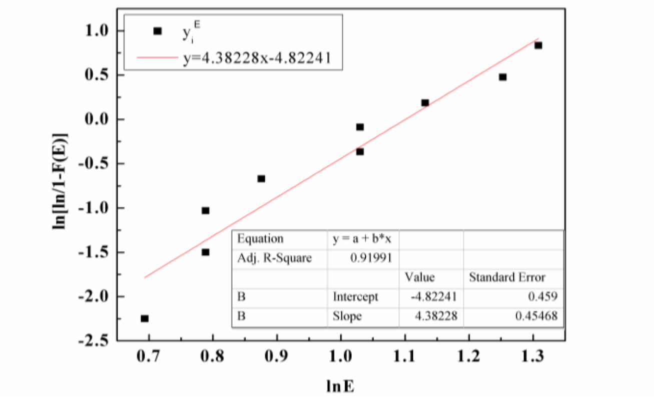

Fig. 6. Cumulative failure probabilities of the breakdown field for PI films

3.3. Mid-value of the lifetime and breakdown field

The probability of mid-value means is fifty percent. When *F(t)* = 0.5, figure out the lifetime of mid-value (*t* = 164.9 min). In order to assess the reliability of the material performance in the short time accelerated life testing method usually is used. Therefore, this value does not represent the real value of industrial applications. When *F(E)* = 0.5, mid-value of the breakdown field *E* = 2.76 MV/cm.

4. CONCLUSION

The electroluminescence of polyimide filmswas investigated under dc high electric field by home-made experimental equipment. The EL characteristic curve shows that EL intensity approximately equal to background noise when strength of the electric field is less than 2.00 MV/cm, and EL intensity increases along with the electric field increasing when the strength of electric field is greater than 2.00 MV/cm. The EL intensity grows rapidly when electric field is approaching or greater than 2.8 MV/cm, however, if continues to increase the electric field, PI film will breakdown.

The cumulative failure probability models for lifetime and breakdown have been developed in this paper based on the 2-parameter Weibull distribution. The two models can be used to evaluate the stability of PI films. After, calculate nine groups experimental data, it is found that the experimental results have a superior reliability. Weibull distributionfunction,

$$F(t) = 1 - e^{-\left(\frac{t}{203.6695}\right)^{1.73549}}$$

and

$$F(E) = 1 - e^{-\left(\frac{t}{3.00547}\right)^{4.38228}}$$

the mid-value of lifetime is 164.9 minutes, the mid-value of pre-breakdown field is 2.76 MV/cm, and reliability are 0.92 and 0.91 respectively.

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