

Mechanical and Dielectric Breakdown Properties of Eco-Friendly Dielectric Materials

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The mechanical and dielectric breakdown properties of polylactic acid (PLA) and PET copolymer, which are biodegradable plastics, were examined, and physicochemical analysis were performed. At room temperature, the Young's modulus of PLA and PET copolymer were about 3.6 GPa and about 3.9 GPa, respectively. Moreover, the temperature to which Young's modulus of PLA and PET copolymer begins to decrease was about 60°C and about 55°C, respectively. The dielectric breakdown strength (E_b) of PLA and PET copolymer at room temperature were about 6.2 MV/cm and about 4.8 MV/cm, respectively. The E_b of PLA increased with an increase in temperature, and was about 6.9 MV/cm at 60°C ($\partial E_b/\partial T \geq 0$). The E_b of PET copolymer was hardly dependent on temperature in the domain of 70°C or less ($\partial E_b/\partial T \cong 0$). In contrast with it, when it became a high temperature region, E_b of PLA and PET copolymer decreased with the rise of temperature ($\partial E_b/\partial T < 0$).

Key Words : biodegradable plastics, mechanical properties, dielectric breakdown properties

1 INTRODUCTION

The production of synthetic plastics including cross-linked polyethylene and epoxy resin has gathered pace rapidly as a result of developments in the petrochemical industry. These products now dominate all areas of daily life and industry. Synthetic plastics are light-weight, easily processed, and have features of viscoelastic materials such as mechanical properties, electric insulation, insulation characteristics, and corrosion resistance, and are therefore widely used as electric insulation materials. In addition, with consumer electronics and digital assistants having quickly become widely available, and the development of modern electrical, electronic and network industries, synthetic plastics will be increasingly used as electric insulation material. However, most of these instruments are disposed of after use and damage the environment, causing a large problem for society.

Biodegradable plastics are plastics that are decomposed via bacteria and decomposition enzymes in nature to produce water and carbon dioxide. They can therefore be treated as waste by being buried in the ground. These plastics generate only a small amount of heat during combustion and do not release poisonous materials such as dioxins. This means that using biodegradable plastics instead of synthetic plastics protects the environment [1-5]. However, there are very few research reports into or authors working on the electrical characteristics of biodegradable plastics, and for this reason, these plastics are not yet actually used as electrical insulation materials [6-10].

There are three types of biodegradable plastics: biopolymers, synthetic polymers, and modified natural polymers, which differ in

the method used to produce them. Synthetic polymer is currently in use.

Synthetic polymers can be further classified into "plant based polymers" and "mineral oil based polymers."

We selected polylactic acid (PLA) that was "Plant base" and polyethylene terephthalate copolymer (PET copolymer) that was "Mineral oil base", which are considered as eco-friendly dielectric materials. In our research, we examined the mechanical and dielectric breakdown properties of PLA, and conducted a physicochemical analysis.

2 EXPERIMENTAL METHODS

2.1 Samples

The sample used was biaxially oriented PLA and PET copolymer films (trade name: Palgreen LC and BO, 25 and 20 μm thick) from Tochello Co, Ltd [11]. Figure 1 shows the chemical structure of PLA and PET copolymer. Low-density polyethylene (LDPE) and polypropylene (PP) films were used as samples for comparison. The thickness of the samples for comparison was 25 μm .

2.2 Thermal analysis

A differential scanning calorimetry (DSC) instrument (Shimadzu, DSC-60) was used for thermal analysis. α -Alumina was used as a reference material, and the DSC spectra for PLA were observed over a rise in temperature from 30°C to 230°C at a rate of 10°C/min. in a nitrogen atmosphere.

2.3 Mechanical properties

A tensile test was performed based on the Japanese Industrial

Standards (JIS K7127₁₉₉₉). In order to eliminate stress concentration, the sample was cut so that cracks would not develop. Figure 2 shows the dimensions of the sample used in the tensile test. The sample was cut into strips of 150×10 mm like paper, with the distance between zippers (L) set to 100 mm. The distance between benchmarks (L_0) was 50 mm, and the samples were studied to ensure that they would fracture within the distance L_0 . For LDPE, L was set to 50 mm. This is because the displacement at the time of breakage is very large, and exceeds the capacity for displacement of the examination apparatus. The tensile speed was set to 100 mm/min, and the tensile strength was measured while the sample was heated from room temperature to 120°C, and the specific conditions were measured at least five times.

2.4 Dielectric breakdown properties

Figure 3 shows the experimental system for measuring dielectric

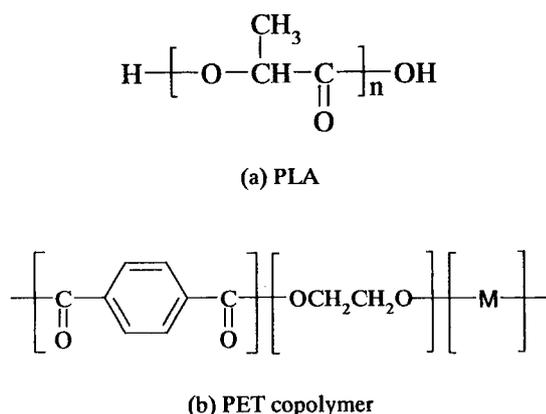


Figure 1 Chemical structure of PLA and PET copolymer.

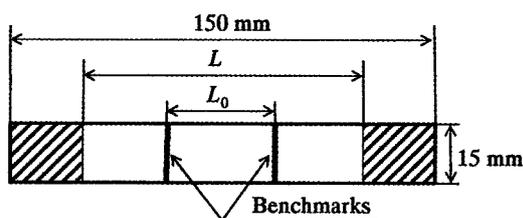


Figure 2 Dimensions of the sample used in the tensile test.

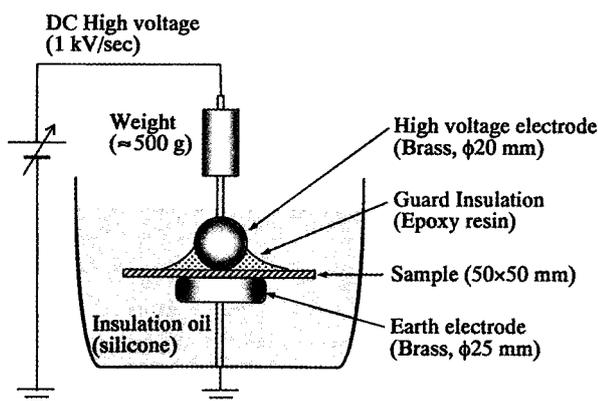


Figure 3 Experimental system for measuring dielectric breakdown strength.

breakdown strength. The samples were cut into 50×50 mm squares. The side at high voltage was made into a ball electrode with $\phi 10$ mm, and the earthed side was made into a disk electrode with $\phi 25$ mm, and rounded edges. To prevent creeping discharge, guard insulation was put in place on the contacts surrounding the high voltage electrode and the sample using epoxy resin. The sample and the electrode system were soaked in silicone oil, and a positive high voltage direct current was applied at a rate of increase of 1 kV/sec. The temperature of the sample changed from the room temperature to 120°C, and the specific conditions were measured at least ten times.

3 RESULTS AND DISCUSSION

3.1 Thermal analysis

Figure 4 shows DSC spectra of PLA and PET copolymer. In the glass transition temperature (T_g) values derived from the DSC spectra, the value for PLA was about 66°C, and PET copolymer was about 54°C. Thus, T_g of PLA was a little higher temperature than PET copolymer. A large absorption peak also appears at temperatures above 150°C. The peak was attributed to melt transition, the temperature of which corresponds to the melting point (T_m). In the T_m values derived from DSC spectrum, the value for PLA was about 168°C, and PET copolymer was about 203°C. Therefore, it was seen that PET copolymer with the aromatic ring is more excellent thermal stability than PLA.

3.2 Mechanical properties

Figure 5 shows the relationship between tensile strength and temperature at breakage. The tensile strength of PLA and PET copolymer at room temperature were about 100 MPa, almost the same as the value for PP. This value was about four times the tensile strength of LDPE. In all the samples, tensile strength decreased with a rise of temperature, and at 100°C the tensile strength of PLA was almost the same as LDPE. The decreasing rate of tensile strength of PET copolymer became a tendency to look like PP, and was more gradual than the decreasing rate of PLA.

Figure 6 shows curves of tensile strength against displacement

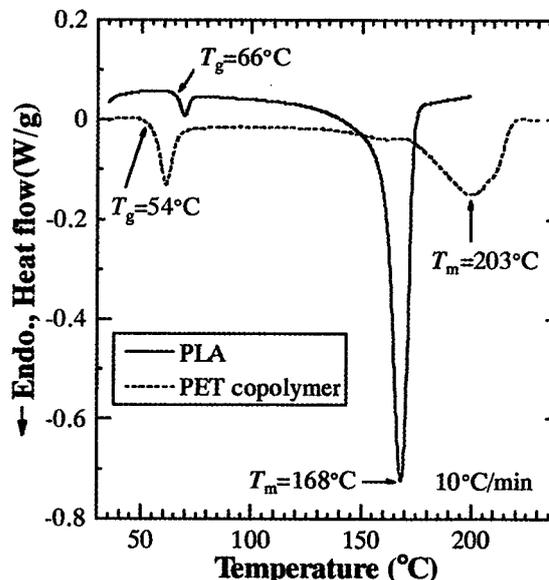


Figure 4 DSC spectra of PLA and PET copolymer.

at 40°C. The tensile strength and displacement curves of PLA were similar to PP, and the tensile yield strength of PLA became higher than the value for PP. On the other hand, the tensile strength and displacement of PET copolymer became higher than the value for PLA.

Figure 7 shows curves of tensile strength against displacement of PLA and PET copolymer. The tensile yield strength of PLA became lower as the sample temperature rose, and the slope of the start of the curves tended to be straight. The tensile yield strength of PET copolymer became lower as the sample temperature rose like PLA, but the tensile yield strength didn't appear at 60°C. Moreover, the slope of the tensile strength and displacement curves of PET copolymer became looser than that of PLA. That is, the PET copolymer became soft by the rise of temperature than PLA.

The Young's modulus was evaluated from the inclination of the straight line portion of the beginning of these curves [12]. The Young's modulus of the other samples was also evaluated. The results are shown in Figure 8. This figure shows the relationship

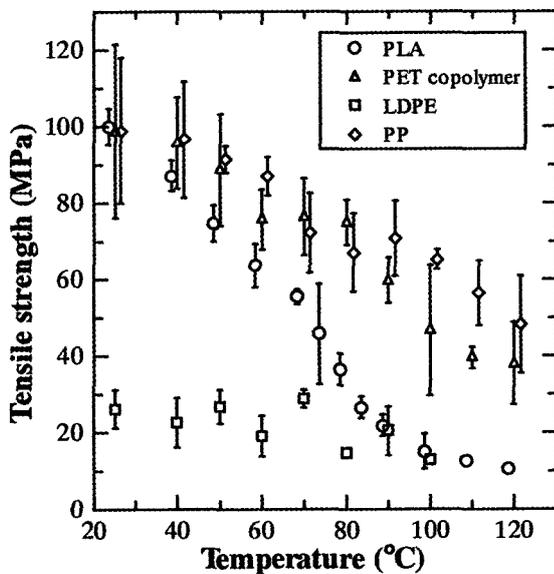


Figure 5 The temperature dependence of tensile strength at break.

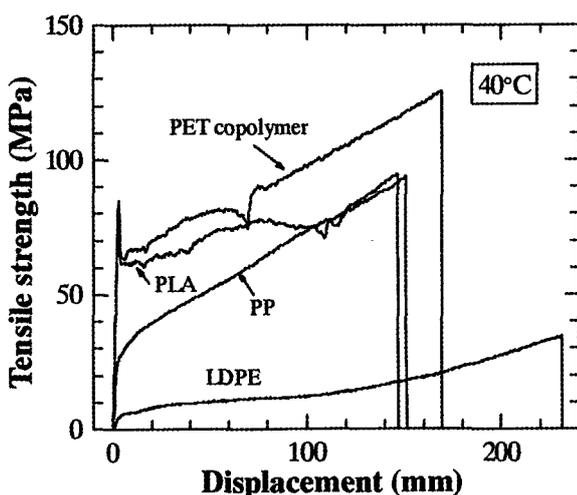


Figure 6 Tensile strength-displacement curves at 40°C.

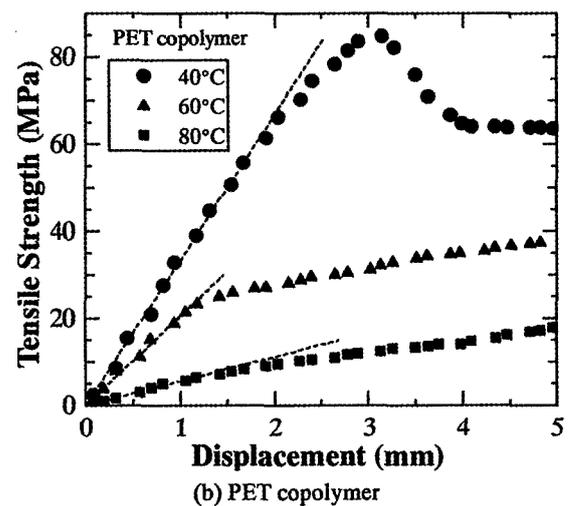
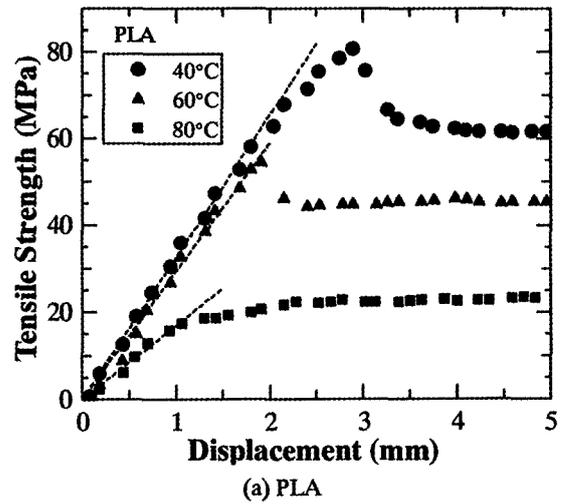


Figure 7 Tensile strength-displacement curves of PLA and PET copolymer.

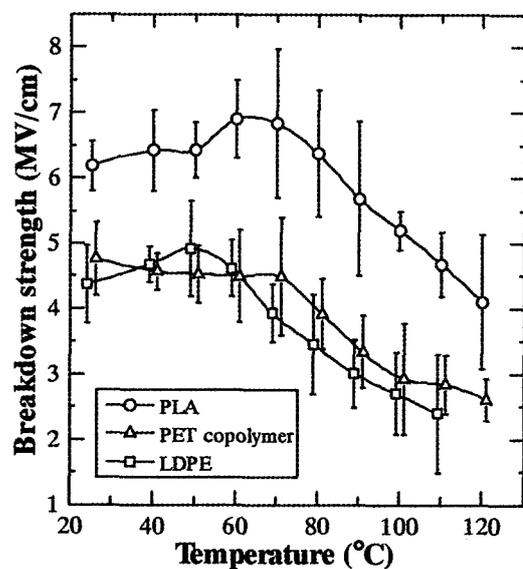


Figure 8 Relationship between Young's modulus and temperature.

Table 1 Feature of dielectric breakdown properties of PLA and PET copolymer.

	Region I	Boundary	Region II
PLA	$\partial E_B/\partial T \geq 0$	$\approx 60^\circ\text{C}$ ($\approx T_g, T_s$)	$\partial E_B/\partial T < 0$
PET copolymer	$\partial E_B/\partial T \approx 0$	$\approx 70^\circ\text{C}$ ($> T_g, T_s$)	$\partial E_B/\partial T < 0$

20°C ← → 120°C
Glass-like Rubber-like

E_B : dielectric breakdown strength, T : temperature

T_g : glass transition temperature, T_s : softening point

between Young's modulus and temperature. The Young's modulus of PLA at room temperature was about 3.6 GPa, about 1.7 times as large as the value for PP, and about 11 times as large as the value for LDPE. On the other hand, the Young's modulus of PET copolymer at room temperature was about 3.9 GPa, and became a value a little higher than PLA. The Young's modulus of PLA and PET copolymer decreased gradually with a rise in temperature, and decreased rapidly for high temperatures, 50°C and above. From these results, it was found that the softening point (T_s) of PLA and PET copolymer were at a high temperature just over 60°C and 50°C, respectively, and it is thought to be near T_g . At 80°C and above, the Young's modulus of PLA and PET copolymer became similar to PP. This result shows that PLA and PET copolymer are harder than PP and LDPE at temperatures lower than 80°C.

3.3 Dielectric breakdown properties

Figure 9 shows the relationship between dielectric breakdown strength (E_B) and temperature. The E_B of PLA at room temperature was about 6.2 MV/cm, about 1.4 times as large as the value for LDPE. It has been reported that the impulse breakdown strength of PLA at room temperature is 1.3 times that of XLPE [13]. The E_B of PLA increased with a rise in temperature, and was about 6.9 MV/cm at 60°C ($\partial E_B/\partial T \geq 0$). The E_B of PLA decreased with a

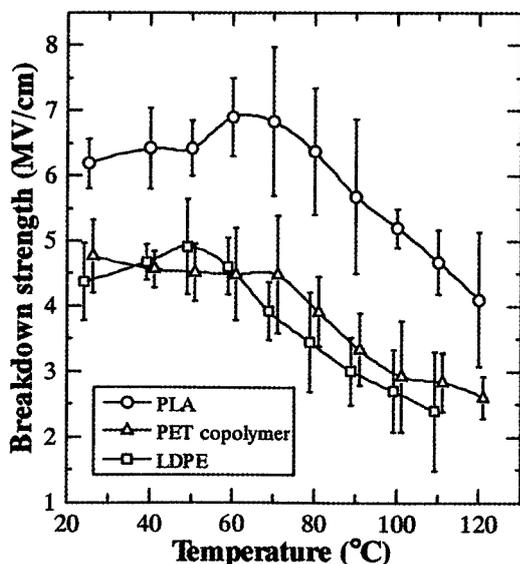


Figure 9 Relationship between dielectric breakdown strength and temperature.

rise in temperature above 60°C in the high temperature region ($\partial E_B/\partial T < 0$). This indicates that the temperature can be divided into two regions bordering on the neighborhood of 60°C. On the other hand, the E_B of PET copolymer at room temperature was about 4.8 MV/cm, and is lower than PLA. The E_B of PET copolymer increased with a rise in temperature.

The E_B of PET copolymer was hardly dependent on temperature in the domain to the neighborhood 70°C from room temperature ($\partial E_B/\partial T \approx 0$). However, E_B of PET copolymer decreased with a rise in temperature above 70°C in the high temperature region. This indicates that the temperature can be divided into two regions bordering on the neighborhood of 60°C ($\partial E_B/\partial T < 0$). Here, the results in Figures 4 and 8 shows that the T_g and T_s of PLA is near the temperature of this boundary, and the T_g and T_s of PET copolymer is about 15 degrees lower than the temperature of boundary. This feature of the dielectric breakdown properties above was summarized in Table 1. Below, the dielectric breakdown mechanism in each domain is examined based on this fact.

Dielectric breakdown mechanism in Region I

Although the E_B of PLA increased with the rise of temperature, the E_B of PET copolymer hardly changed with variation of sample temperature. An electron avalanche breakdown can be considered to be the mechanism acting in this region [14, 15].

Dielectric breakdown mechanism in Region II

In this domain, as shown in Table 1, the state become $\partial E_B/\partial T < 0$, and so thermal breakdown can be considered to be a primary candidate for the dielectric breakdown mechanism [14, 15].

The results of the investigation by the authors into the dependence of temperature on the current conducted by PLA and PET copolymer, shows that the current increases rapidly at a level above T_g , and it is thought that ion conduction occurs easily due to the micro Brownian motion of molecule chains [7, 9, 10]. In other words, since the Joule heat due to ion conduction increases when the temperature of the sample is raised, it is thought that E_B falls.

In the next state, PLA and PET copolymer are thermoplastic polymers, since this temperature domain is higher than the T_g and T_s of PLA and PET copolymer, shown in Table 1, and so Maxwell stress occurs with the inter-electrode pressure accompanying the applied voltage, and it is expected that electromechanical breakdown will take place easily here. According to the electromechanical breakdown theory [14, 15], the relationship between the dielectric strength and the Young's modulus is

$$E_M = \sqrt{\frac{Y}{\epsilon_0 \epsilon_r}} \exp\left(-\frac{1}{2}\right) \dots \dots \dots (1)$$

where E_M is the electromechanical breakdown strength, $\epsilon_0 \epsilon_r$ is the dielectric permittivity, and Y is the Young's modulus. The relationship between temperature and E_M calculated using the relative permittivity (ϵ_r) of PLA and PET copolymer which the authors have reported [10], and the Young's modulus of PLA and PET copolymer shown in Figure 8, is shown in Figure 10 with the result of this experiment. The measured value of the dielectric breakdown strength (E_B) shown in Figure 9 is also shown in the figure. As can be seen from this figure, the calculated value (E_M) was higher than the measured value (E_B). This tendency is also reported for other polymer films [16, 17]. Y. Inuishi and M. Ieda [18] consider that the above tendencies are shown in local

mechanical weak points. That is, where $E_M \propto E_B$ even if $E_M > E_B$, it is likely that electromechanical breakdown may have occurred. Here, if Equation (1) is simplified, the relationship between E_B and Young's modulus can be given as follows.

$$E_B \propto \sqrt{Y} \quad \dots \dots \dots (2)$$

The relationship between E_B and Young's modulus of PLA and PET copolymer are shown in Figure 11. The dotted line in the figure shows the curve of Equation (2). This Figure shows that the relationship in Equation (2) is found in the temperature domain from 80 to 120°C in PLA. Moreover, this Figure shows that the relationship in Equation (2) is found in the temperature domain from 70 to 120°C in PET copolymer. That is, it is shown that a local mechanical weak point occurs about here. In this domain, it thought that electromechanical breakdown occurs in addition to thermal breakdown, and the temperature of PLA is above 80°C, and the temperature of PET copolymer is above 70°C.

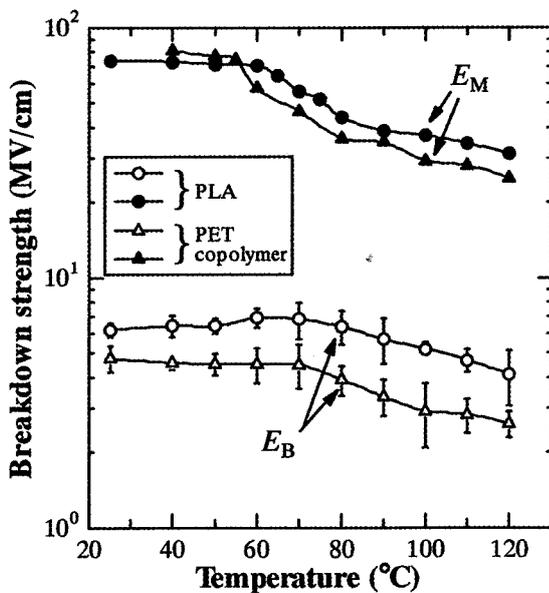


Figure 10 Comparison between measured value of dielectric breakdown strength E_B and theoretical value of electromechanical breakdown E_M in PLA and PET copolymer.

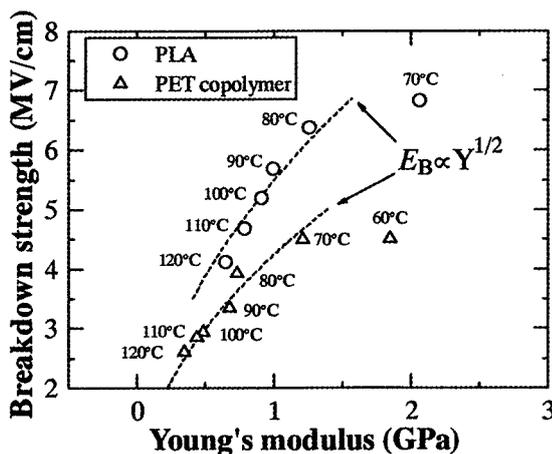


Figure 11 Relation between E_B and Young's modulus of PLA and PET copolymer.

4 CONCLUSIONS

The mechanical and dielectric breakdown properties of polylactic acid (PLA) and polyethylene terephthalate copolymer (PET copolymer), which are biodegradable plastics and are considered as eco-friendly dielectric materials, were examined, and a physicochemical analysis was performed. The results of these were as follows:

(1) The tensile strength of PLA and PET copolymer at room temperature were about 100 MPa, almost the same as the value for PP. At 100°C, the tensile strength of PLA was almost the same as LDPE. The decreasing rate of tensile strength of PET copolymer is similar to that of PP.

(2) The Young's modulus of PLA in room temperature was about 3.6 GPa, about 1.7 times as large as the value for PP, and about 11 times as large as the value for LDPE. The Young's modulus of PET copolymer at room temperature was about 3.9 GPa, and became a value a little higher than PLA. The temperature to which Young's modulus of PLA and PET copolymer begins to decrease was about 60°C and about 55°C, respectively.

(3) The dielectric breakdown strength (E_B) of PLA and PET copolymer in room temperature was about 6.2 MV/cm and about 4.8 MV/cm, respectively. The E_B of PLA increase with a rise in temperature, and was about 6.9 MV/cm at 60°C ($\partial E_B / \partial T \geq 0$). The E_B of PET copolymer was hardly dependent on temperature in the domain to the neighborhood 70°C from room temperature ($\partial E_B / \partial T \cong 0$). In this domain, it is thought that an electron avalanche breakdown occurred. In contrast with it, when it became a high temperature region, E_B of PLA and PET copolymer decreased with the rise of temperature ($\partial E_B / \partial T < 0$). In this domain, it was thought that electromechanical breakdown occurs in addition to thermal breakdown, and the temperature of PLA is above 80°C, and the temperature of PET copolymer is above 70°C.

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