Polylactic Acid-Coated Cable

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The polylactic acid (PLA), which is a plant-derived polymer, is an environment-friendly polymer from the point of view of not only having biodegradable property but also saving fossil resources and reducing carbon dioxide emission. Although the PLA has already been applied to many products, it has not yet been applied to cable. Thus, we manufactured and tested the cables using normal and flexible PLA, so as to identify the possibility and problem of application of PLA insulation to cables.

1. Introduction

PLA, a plant-derived and biodegradable polymer, has been put to practical use in the fields of agricultural articles, such as nets or films, and molded articles, such as trays. In addition, from the viewpoint of saving fossil resources on account of reduction in CO_2 emission and the thought of "carbon neutral," the application of PLA is spreading to the fields of electronics and automobile.

However, the studies of PLA application to the cable began several years ago[1)]. In order to examine the possibility of the application to electric cables, we manufactured PLA-coated cables and evaluated their properties, such as tensile strength, heat resistance, conductivity, dielectric breakdown strength, influence of immersion in water, and heat deterioration[2), 3)].

In this paper, we report the properties of the cable and problems of PLA application to cable.

2. Plant-derived polymer and PLA

2.1 Plant-derived polymer

A petroleum-derived polymer has disadvantages from an environment viewpoint, such as drying up of oil resources and greenhouse gas emissions of carbon dioxide. Especially for reducing carbon dioxide emissions, introducing plant-derived polymer is considered to be useful from the standpoint of "carbon neutral." In this situation, the plant-derived PLA may provide a sufficient supply of engineering materials.

2.2 Application of PLA

Taking advantage of the biodegradable property, PLAs have been applied to produce agricultural products such as films and fibers and molded tray.

In addition, the fragility and heat resistance of the PLA continues to be improved by using polymer alloy technology and/or optimization of resin modifiers, and is going to be applied to PCs or mobile phone, housing, textiles, optical, packaging film, bags, and so on.

2.3 Properties of PLA

PLA is considered to have sufficient electrical properties that can be applied to electrical cable, as shown in many reports[4), 5)]. On the other hand, PLA is generally hard, has low tensile elongation, has high brittleness, and has low heat resistance compared with other commodity plastics such as polyethylene. Therefore, the research and development is improving the heat resistance and flexibility, using the plasticization technique and/or polymer alloy technology.

Table 1 shows a comparison of the general properties of PLA and other polymers[6), 7)]. The glass transition temperature Tg of PLA is higher than that of polyethylene and close to that of polyvinyl chloride. And, the heat deformation temperature is thought to be around 60 and heat resistance seems to be a problem. The tensile elongation of PLA is lower than that of the plastics used for cable, and PLA seems to have the problem of flexibility. The dielectric constant ε and dielectric dissipation factor tanδ of PLA are slightly larger compared with a polyolefin, and the volume resistivity ρ is sufficiently high when applied to cable.

PLA is thought to decompose easily. However the biodegradation does not occur without hydrolysis, as the biodegradation of PLA occurs in two steps: the first step is hydrolysis of PLA and the second step is biodegradation by bacteria. Therefore, it is thought that PLA cables do not disintegrate easily by limiting the environment of the cable.

3. Experimental

3.1 Manufacture of cable

The electrical cable has various types, for example, the power cables, communication cables, coil, elec-

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Panel 1. Abbreviations, Acronyms, and Terms.

PLA—polylactic acid
LDPE—low density polyethylene
HDPE—high density polyethylene
PP—polypropylene
PVC—polyvinyl chloride
TS—tensile strength
EL—tensile elongation
tanδ—dielectric dissipation factor
ε—permittivity
ρ—volume resistivity
T_g—glass transition temperature
JIS—Japanese Industrial Standards
AC—alternating current
IV—Indoor PVC
DSC—differential scanning calorimetry

Table 1. Properties of PLA and other polymers.[4), 5)]

Polymer	Thermal properties			Mechanical properties		Electrical properties			Remarks
	Density g/cm³	Glass transition temperature Tg °C	Melting point °C	Tensile elongation EL %	Tensile strength TS MPa	Dielectric dissipation factor tanδ %	Permittivity ε	Resistivity ρ Ω cm	
PLA	1.3	60	170	4	59	0.01	3.1	4.3×10^{17}	
LDPE	0.92	−120	110	600–700	15–20	0.01	2.3	$>10^{16}$	Cable material
HDPE	0.95	−120	130	650	12	0.01	2.3	$>10^{16}$	
PP	0.91	5	165	800	38	0.05	2.2	$>10^{16}$	
PVC	1.4	70–100*	180*	300*	20*	0.10*	4–5*	10^{11}–10^{14}*	* : depend on amount of plasticizer

tronic appliance cable, automobile cable, etc. In this experiment, we have chosen a 600 V cable as a target, which simply consists of a conductor and insulation.

The cable manufactured, which has seven copper stranded conductors (diameter 1.8 mm), has an outer diameter 3.2 mm and insulation thickness 0.7 mm. The cables were extruded at 180 °C and 200 °C, and quenched in water.

The insulation materials are raw PLA (PLA-A) and added flexibility one (PLA-B) modified by a plasticizer. The former is to get properties of base polymer and the latter is to examine the effect of addition of flexibility. The properties of the resins are described in Table 2.

Table 2. Properties of PLA-A and PLA-B.

Item		PLA-A	PLA-B	Test method
TS	MPa	76	39	JIS K 7161
EL	%	5	220	JIS K 7161
Heat deformation	°C	53	57	JIS K 7191

Table 3. Test items and methods for PLA insulating cables.

Item		Test method
Mechanical properties	Tensile test	JIS C 3005
	Bending test	Observation with bending
Thermal resistance	Heat deformation test	JIS C 3005
Electrical properties	ε	JIS C 3005
	tanδ	JIS C 3005
	ρ	JIS C 3005
	AC breakdown test	Voltage of rising speed 1 kV/s, in 5 % saline as ground
	AC breakdown test with bending	Voltage of rising speed 1 kV/s, in 5 % saline as ground
Water-proof properties	ε	After immersion
	tanδ	After immersion
	ρ	After immersion
	Moisture content	Karl Fischer method

3.2 Test items of cable

The test items and methods are shown in Table 3.

The bending property is tested by observing the appearance, with cable winding to metal rods of different diameters. The scale is indicated by magnification of cable diameter. The bending diameter dependence of AC breakdown strength were measured by applying a rising voltage of 1 kV/s, in 5% saline as ground, at the test temperature of 22 °C. We consider that the dielectric strength at bending diameter 80 times is the strength at straight cable.

The electrical measurements were performed by making electrodes with conductive paint; the lead electrode was 300 mm long on insulator and the guards (20 mm length) were on both sides of the lead electrode. And the measurements were performed as follows: dielectric dissipation factor tanδ and capacity were measured using automatic Schering bridge (500 V) and insulation resistance using teraohmmeter (500 V × 1 min value). Dielectric constant ε was obtained from the capacity and volume resistivity ρ from the resistance.

Temperature dependence of electrical properties

was measured at temperatures ranging from room temperature to 90 °C in the oven.

The water absorption properties were measured as follows: the samples were prepared for measurement of electric properties and moisture content, and immersed in water at room temperature about 23 °C. The electrical properties and water content were measured at setting times. The Karl Fischer method was used to measure moisture content. And tanδ, ε, and ρ were obtained as above.

The thermal degradation measurement was carried out for only PLA-A. Cables (60 cm long) were heated in JIS type ovens (90, 120, 150 °C), and sampling and observation were carried out over time. And after heating, electrical properties were measured. The deteriorated cables with cracks were measured on no-crack area. In addition, differential scanning calorimetry (DSC) was performed to evaluate the thermal performance of the cables.

4. Results and Discussion

4.1 Manufacturing cables

As shown in Figure 1, PLA-A and PLA-B have clear insulation, and both cables have a smooth surface and clean appearance. However, in PLA-A cable extruded at 200 °C, microbubbles, likely resulting from rapid quenching, were seen continuously on the conductor. Therefore, we selected the cable extruded at 180 °C as an investigation object. On the other hand, microbubbles could not be observed inside PLA-B, and the cable extruded at 180 °C was selected similar to PLA-A.

4.2 Mechanical properties

Figure 2 shows the results of tensile test. PLA-A has more elongation than the manufacturer's value, but PLA-B is not elongated to the expected value of manufacturer.

In any event, as for the elongation of 25% in PLA-B, the improvement in elongation is demanded of an electric cable. The difference between elongation values of measurement and manufacturer is probably considered due to differences in shape of the cable and test methods, that is, film and cable, and JIS K 7161 and JIS C 3005.

Figure 3 shows the results of bending properties. PLA-A becomes white at bending diameter 10 times the cable diameter, and cracking at twice the diameter.

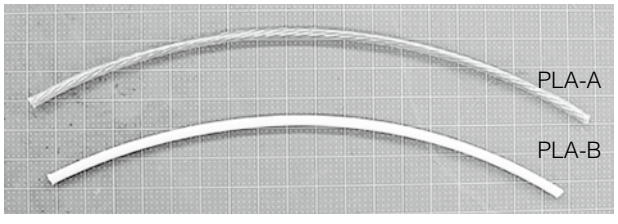


Fig. 1. Appearance of PLA insulating cables.

However, PLA-B becomes white at bending self-diameter, but no crack was observed.

4.3 Heat deformation test

As shown in Fig. 4, at 90 °C, both PLA-A and PLA-B are transformed little, but at 120 °C, PLA-A has the same order heat resistance as that at 90 °C contrary to what is expected. On the other hand, PLA-B is deformed very much and seems to have no heat resistance at 120 °C, and this deformation is caused by adding flexibility. This result indicates that PLA has a higher heat resistance at Tg and modification of PLA can reduce heat resistance.

4.4 Electrical properties

As far as AC breakdown strength between PLA-A and PLA-B without bending is concerned, there is no difference. However, in proportion to bending, there

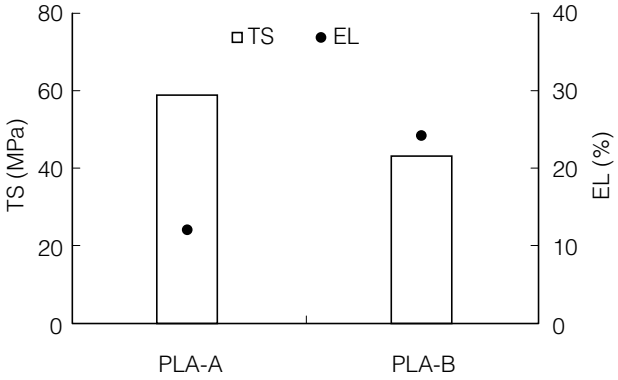


Fig. 2. Tensile strength and elongation of PLA insulating cables.

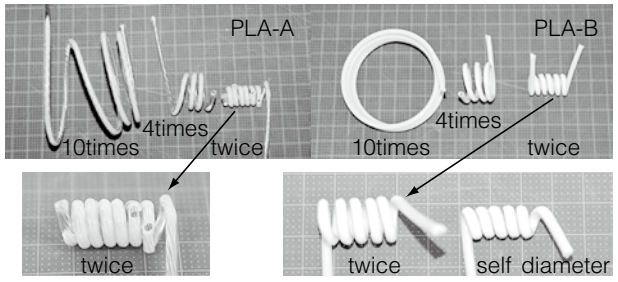


Fig. 3. Bending test of PLA insulating cables.

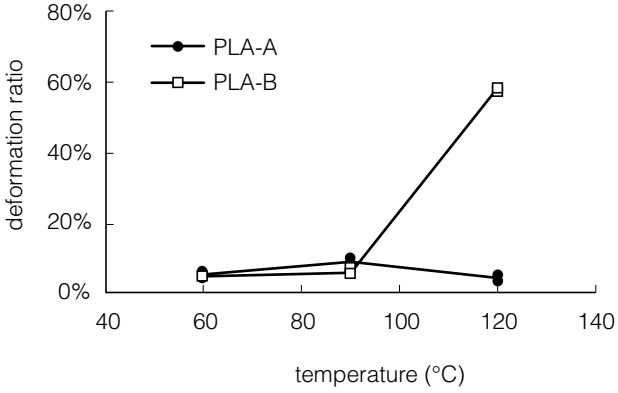


Fig. 4. Thermal dependence on deformation of PLA insulating cables.

was a marked positive effect in PLA-B. The AC breakdown strength is plotted against bending radius (Fig. 5). In PLA-A, breakdown voltage at 10 times the radius of the cable reduces to one-fourth at no bending, and at 4 times the radius of the cable, we could not apply voltage owing to small cracks. On the other hand, at even self-radius bending, PLA-B has half the breakdown voltage at no bending. Thus, adding flexibility does not affect the breakdown voltage at no bending and improves the breakdown voltage at bending. In addition, with the AC breakdown voltage of these cables, it can be said that they have enough electric strength compared to EP rubber, although it doesn't reach the strength of polyethylene [8].

The electrical properties (tanδ, ε, ρ) are shown in Table 4. PLA-A has good electrical properties, as expected from previous reports. On the other hand, PLA-B shows inferior properties to PLA-A, and this characteristic seems to be affected by adding flexibility. This result indicates that the modification of PLA deteriorates the electrical properties. Therefore, improvement in flexibility is required to balance the electrical and other properties.

4.5 Temperature dependence of electrical properties

According to several reports on temperature dependence of electrical properties of PLA sheet, PLA has been indicated to cause changes in electrical properties near the glass transition temperature[8].

Figures 6 and 7 show the temperature dependence of ρ, tanδ, and ε of the two cables. As the temperature increases, ρ of PLA-A decreases significantly in the vicinity of Tg (60 °C), but does not decrease at higher Tg in Fig. 6. On the other hand, ρ of PLA-B drops significantly in the vicinity of Tg at 40 °C and continues to decrease as the temperature increases.

As shown in Fig. 7, tanδ of PLA-A has a large peak around Tg and becomes smaller at high temperature. The tanδ of PLA-B rises with temperature. In general, the resistivity of polymer decreases with increasing temperature. This reduction occurs due to increasing density of conductive carrier and/or the mobility. Also, near Tg, an increase in the current and rise in the loss tangent are observed. This absorption current seems to be due to orientation polarization with phase transition.

For PLA-A, an increase in tanδ and decrease in ρ near Tg is thought to be caused by the polarization orientation of the dipole glass transition. In addition, above Tg, ρ is not changed and tanδ reduces again. In this temperature range, conductive component is likely to be affected very little. On the other hand, for PLA-B, ρ decrease and tanδ increase take place in proportion to the temperature. It is suggested that the conductive component affects electrical properties.

In conclusion, the temperature dependence of the electrical property of PLA-A is excellent at high temperature, although tanδ increases in the vicinity of Tg.

In case of PLA-B, additives for flexibility may significantly reduce electrical properties at high temperatures. Therefore, when we use PLA for electric insulation, we need to pay attention to selection of additives.

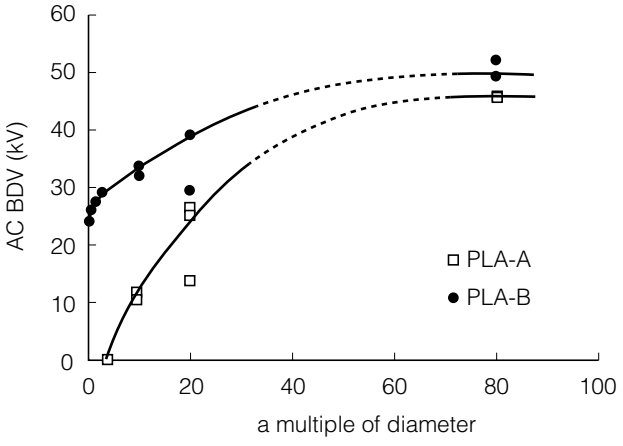


Fig. 5. Bending diameter dependence of AC BDV.

Table 4. Electrical properties of PLA insulating cables.

Material/item	tanδ (%)	ε	ρ (Ωcm)
PLA-A	0.35	3.2	2.4×10^{16}
PLA-B	2.31	4.1	4.2×10^{12}
PE	0.01↓	2.3	1×10^{16} ↑

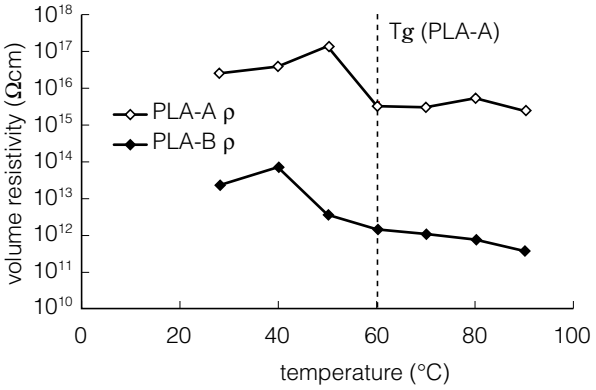


Fig. 6. Temperature dependence of resistivity.

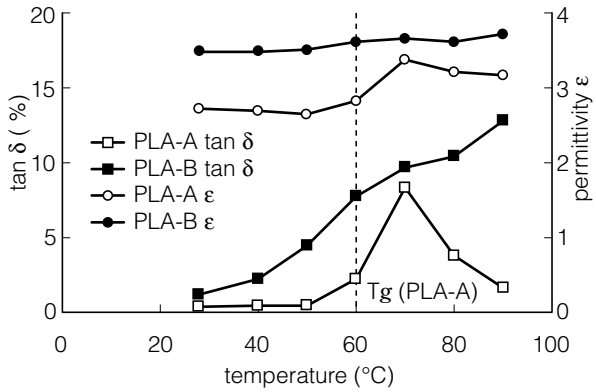


Fig. 7. Temperature dependence of dielectric loss and permittivity.

4.6 Properties of long-term immersing

Water absorption properties are shown in Fig. 8. The moisture contents of PLA-A and PLA-B are about 4000 ppm initially and increase when immersed in water. Over 200 h, their moisture showed a tendency to saturate at around 10,000 ppm. It is understood that the moisture value 10,000 ppm is much larger than saturated volume of water about 50—100 ppm of the polyethylene. This is because PLA includes carbonyl and oxygen in the main chain and is hydrophilic compared to polyethylene. The PLA-B has more moisture content, an effect of plasticizer used for flexibility.

Figure 9 shows the electrical properties of the immersed cable. The electrical properties of PLA-A change little during immersion, but ρ of PLA-B decreases and tanδ and ε increase with immersion. Figure 9 also shows that electrical properties of PLA-B recovered to initial value by decompression drying. Figure 10 is a plot of the properties against amount of moisture; those of PLA-A slightly change at 13,000 ppm and those of PLA-B decline greatly at 15,000 ppm.

From these results, it is guessed that the electrical property doesn't decrease with moisture only. As shown in Fig. 9, decompression drying needs to recover electrical properties of PLA-B, so that it is certain that moisture influences the electrical property. In contrast, the electrical property of PLA-A does not change so much with increasing moisture. Accordingly, the degradation of electrical properties is considered to be caused by interaction with water and plasticizer.

From the significant decrease in ρ at high temperatures described above, the PLA-B is likely to include more conductive elements than PLA-A. Besides, high temperature and high humidity will lead to hydrolysis of PLA; therefore, prevention of water absorption is desirable[9].

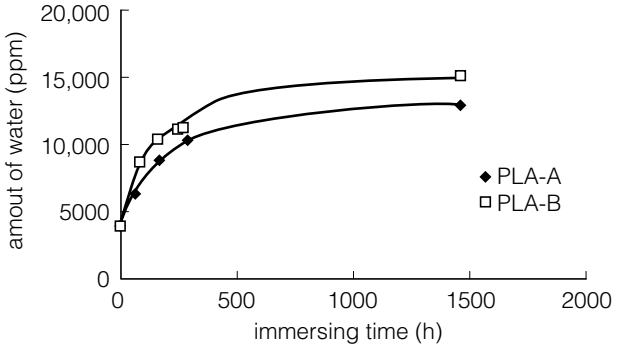
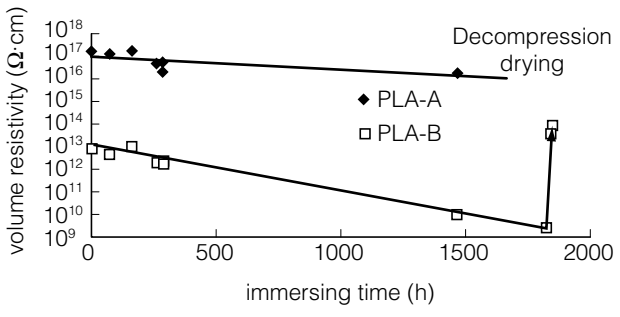


Fig. 8. Quantity of water in PLA insulating cables.



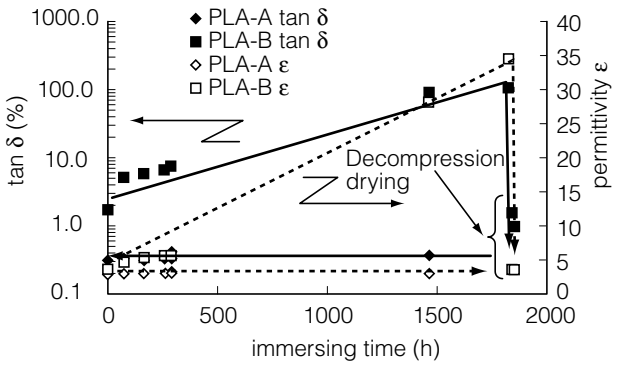
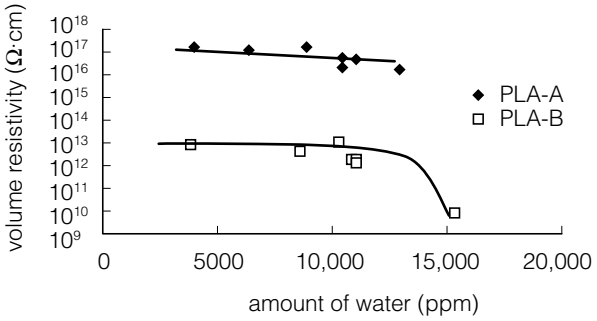


Fig. 9. Change in electric properties of PLA insulating cables.

4.7 Thermal degradation mechanism

PLA-A did not deform and did not melt at 150 °C because of its melting point of 170 °C. However, cracks occurred after several days at 120 and 150 °C as shown in Fig. 11. The crack generation time decreased with temperature. On the other hand, at 90 °C the crack was not observed in the short term and occurred after 2800 h, finally.

It can be said that depolymerization does not cause crack generation because thermal analysis did not indicate any shift of melting point with depolymeriza-



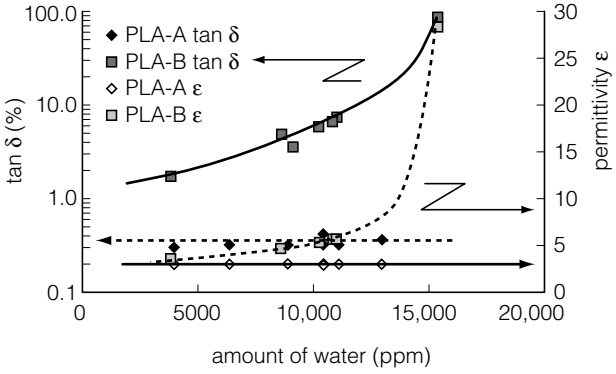


Fig. 10. Relations between electric properties and quantity of water in PLA cables.

tion. Furthermore, it is hard to observe the heat stress crack since samples are not deformed.

In general, in crystalline polymers, volume contraction occurs by crystallization. We thought this crack will be due to the crystallization of PLA, and we evaluated the conditions of crystal growth by thermal analysis.

Figure 12 is a DSC chart of nonheated PLA-A and shows exothermic peak around 110 °C. This peak is due to the crystallization in noncrystalline part of crystalline area, which has the ability to make the crystal. The crystal doesn't grow in the PLA-A because of its slow crystallization speed and quick cooling with water immediately after the extrusion on manufacturing. For this reason, the PLA-A cable is transparent.

In contrast, Figure 13 is a DSC chart of heated PLA-A, which was cracked, and shows no peak around 110 °C. This means that crystallization occurred upon heating in the oven, and so heated PLA-A has reduced transparency.

Consequently, we estimate that this crack caused the crystallization. In other words, the crystallization of PLA-A is promoted by heating, continuously, as a result of which the crystal grows, the volume shrinkage occurs, and the crack is caused.

Crystallization occurs after heating even at 90 °C, and exothermic peak was not observed as in Fig. 13. However, the times of cracking at 90, 120, and 150 °C differ much. As shown in Fig. 12, the PLA crystallization peak can be seen around 110 °C. On the boundary of the crystal formation temperature 110 °C, a significant volume change might occur upon crystallization at 120 –150 °C and a small volume change will be generated at 90 °C.

As a result, if we prevent crystallization, PLA-A can be considered to prevent cracking by heating.

4.8 Effect of mechanical properties due to electrical heating

As shown in Fig. 14, significant changes in electrical properties near Tg (60 °C), which were observed in the unheated PLA-A such as the maximum and the minimum, disappeared after heating. This is because an amorphous portion decreases and crystallinity increases upon heating, and the effect of glass transition is reduced. In addition, the electrical properties are almost constant regardless of the heating conditions, indicating that there is no effect on electrical proper-

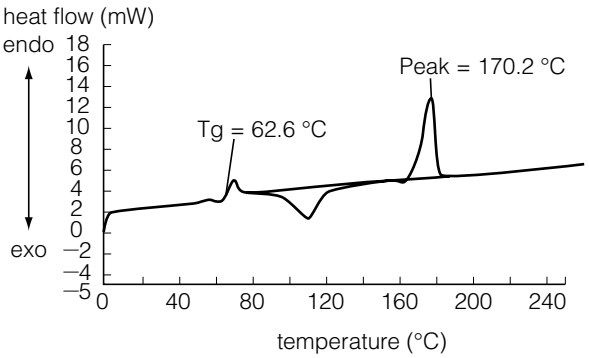


Fig. 12. DSC curve of nonheating PLA-A cable.

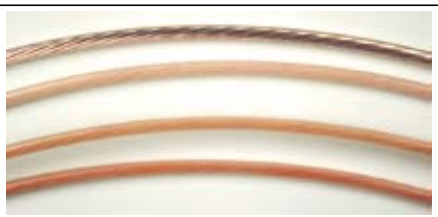
Temp.	Appearance	Degradation
150 °C	not-heated 24 h 72 h 168 h	1 day : some cracks 2 days : many cracks life time : <24 h
120 °C	not-heated 24 h 48 h 72 h 168 h	3 days : some cracks 5 days : many cracks life time : <72 h
90 °C	 not-heated 24 h 72 h 240 h	117 days : some cracks life time : <2800 h

Fig. 11. Results of aging test.

ties such as thermal degradation leading to cracks.

5. Possibility of applying PLA to cables

On the basis of above results, we compared PLA-coated cable with polyvinyl chloride-insulated cable to investigate the feasibility of PLA-coated cable (Table 5). The reason of selecting PVC is that electrical properties of PLA-B resemble those of PVC. Therefore, PLA-B is a potential alternative to polyvinyl chloride-insulated cables.

Unfortunately, PLA-B, which was kept at room temperature, lost flexibility after 2.5 years and cracked with slight bending. Meanwhile, PLA-A, which was kept under similar conditions, did not indicate any sign of degradation, and it remained intact even after 5 years of production. To apply PLA to cables, it is important to ensure long-term retention of plasticizer effect.

PLA maintains enough electrical property as an insulator, even if plasticizer is added for flexibility. In addition, the antidegradation due to heating is considered to be primarily anticrack, so that these measures can correspond to an appropriate use of temperature setting. Moisture of PLA increases during immersion in water, but electrical degradation of PLA is slight and the crack is not observed, and it can be said that there is enough resistance to water.

However, on adding flexibility to cable, it is necessary to evaluate the resistance to water. Furthermore, under the applied temperature and humidity, hydrolysis of PLA progresses and degradation occurs.

From the above discussion, for practical application of PLA to cables, we think it is important to secure flexibility and water resistance. Also, it may be said that we can use it enough if we limit location and tempera-

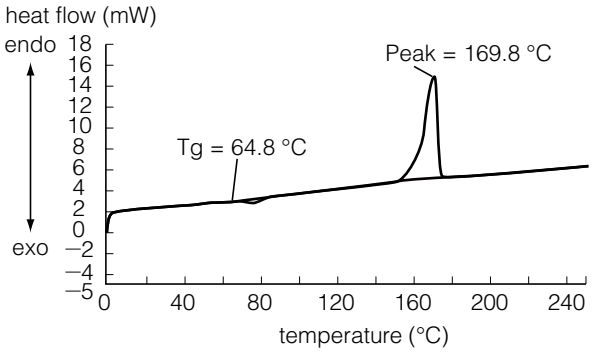
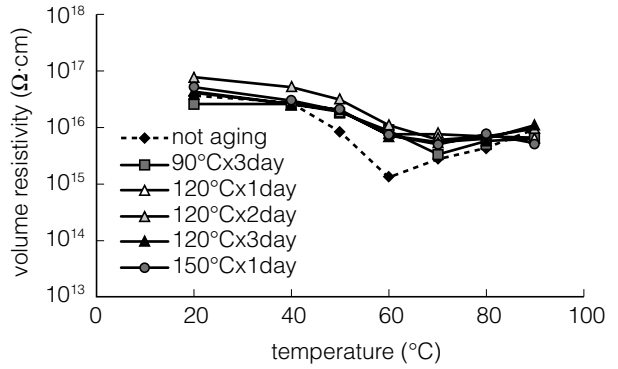


Fig. 13. DSC curve of heated PLA-A cable (150 °C, 1 day).



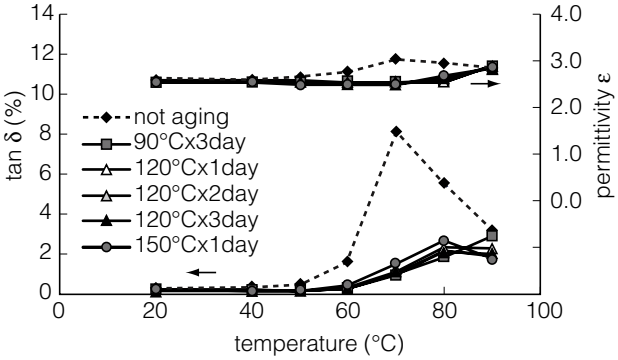


Fig. 14. Temperature dependence of electrical properties of heated cables.

Table 5. Evaluation of PLA coated cable in comparison with polyvinyl chloride cable.

Item/Cable	PLA-A	PLA-B	600 V polyvinyl chloride cable (IV) JIS C 3307
Extrusion	○ : excellent appearance △ : void in surface between conductor and insulation	△ : excellent appearance ○ : analogy from PLA-A	—
Bending	× : whitening at 10 times bending cracking at 4 times bending	△ : whitening at 2 times bending ○ : no cracking at self-diameter bending	—
Tensile	○ : TS = 59 MPa × : EL = 12 %	○ : TS = 43 MPa × : EL = 25 %	TS more than 10 MPa EL more than 100 %
Heat deformation	◎ : 60 − 120 °C reduction less than 10%	◎ : 60 − 90 °C reduction less than 10 % × : 120 °C reduction 58 %	Thickness reduction less than 50%
Electrical	◎ : tan δ = 0.35 %, ε = 3.2 ◎ : ρ = 2.7 × 10^{16} Ω·cm	△ : tan δ = 2.31 %, ε = 4.1 ○ : ρ = 4.6 × 10^{12} Ω·cm	ρ : 50 MΩ·km (= 5 × 10^{12} Ω·cm)
Dielectric breakdown	◎ : 35~45 kV (0.7 mm thickness)	◎ : 45~50 kV (0.7 mm thickness)	Withstand voltage test 1.5 kV × 1 min
Dielectric breakdown with bending	× : cracking at 4 times bending	○ : 25 kV at self-diameter bending	—

Evaluation symbols : ◎ superior, ○ equivalent, △ slightly inferior, × unusable

ture. Meanwhile, without sticking to biodegradability, we should try to mix with the resin of petroleum-derived polymer, depending on application of PLA from the standpoint of carbon neutral.

6. Conclusion

We examined properties of PLA-insulated cables, such as electrical, mechanical, and thermal properties. We also discussed the possibility and the problem of PLA application to electric cables.

In future, to apply PLA to cables, it will be necessary both to secure long-term stability of PLA including plasticizer for flexibility and to secure the electrical property. In order to appear in the polyethylene plant, PLA needs to be established again from the perspective of application to the cable coating of carbon.

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