EVALUATION OF PHYSICAL PROPERTIES OF THIN FILM AND MEASUREMENT OF UNSTEADY THERMAL STRESS

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ABSTRACT

Winding defect such as the roll wrinkles or slips to lose the value of the product. Winding defect is caused by the internal stress condition in the roll and the temperature change of the roll after winding. Therefore, it is essential to analyze the internal stress theoretically. However, the tendency of internal stress to occur as the film thickness becomes thin is not known. The internal stress of the roll was measured at each temperature. Further changes in internal stress were also measured by changing the ambient temperature around the roll.

The winding device used in this research is a structure reproducing the unwinding and the rewinding in the roll-to-roll production system. A film-like pressure sensor was used to measure the radial internal stress of the roll. After winding up the film, the internal stress in the radial direction inside the roll was measured when the temperature around the roll was heated for 12 hours. Measurement frequency was measured for internal stress every hour.

In the PET film with a film thickness of 40 μm, the internal stress in the radial direction inside the roll was measured when the roll surrounding temperature was heated for 12 hours. For the middle and outer layers, the internal stress gradually increased with the lapse of time. It is considered that this increased the pressure by thermal expansion from the outer peripheral side of the roll. However, the value of the stress in the inner layer decreased greatly. This is thought to be caused by uneven heating due to the heat source, winding wrinkles occurred in the vicinity of the sensor sandwiched between the rolls, and stress relaxation occurred.

NOMENCLATURE

a	thermal diffusivity
E_θ	young's modulus of coated web in circumference direction [Pa]
E_r	young's modulus of coated web in radial direction [Pa]
E_c	young's modulus of core [Pa]

h_s	teat transfer coefficient of the outermost layer of the roll
h_a	thickness of air layer within wound roll [m]
R_{th}	thermal resistance
r	roll radius [m]
r_c	core radius [m]
r_1	radius to air layer [m]
r_2	radius to web layer [m]
r_3	radius to the outside of the web layer [m]
s	radius of roll outermost layer
T	temperature [℃]
T_f	temperature of outermost layer of roll [℃]
Ts	ambient temperature [℃]
ΔT	change temperature [℃]
t_w	film thickness [m]
$V_{r\theta}$	poisson's ratio of web
α_θ	expansion coefficient of coated web in circumferential direction [1/K]
α_r	expansion coefficient of coated web in radial direction [1/K]
α_c	expansion coefficient of core [1/K]
λ_{eq}	equivalent conductivity
λ_w	web conductivity [W/(m·K)]
λ_a	air layer conductivity [W/(m·K)]
σ_r	stress of coated web in radial direction [Pa]
σ_θ	stress of coated web in circumferential direction [Pa]

INTRODUCTION

In recent years, research and development of flexible devices such as film capacitors, organic thin film solar cells, and lithium ion secondary batteries using thin and flexible plastic films as substrates have been actively conducted. However, flexible devices are rarely available in the general market. One of the reasons is that it is not suitable for mass production because many processes are performed separately during device fabrication. Therefore, roll-to-roll printed electronics (hereinafter referred to as R2RPE) shown in Figure 1 is one of methods for mass producing these flexible devices. The R2RPE production method is a printed electronics technology that can print devices directly by printing circuits etc. on a substrate, and a roll-to-roll method used as a production method for plastic films, paper media, thin film steel plates, etc. This production method incorporates printing, drying, and laminating processes while supporting and transporting a plastic film as a substrate using a large number of rollers, and is finally wound into a roll. At the winding section of this final process, storage of the long plastic films can be reduced by accumulating them in rolls. Therefore, the transportation efficiency is greatly improved because a large amount of products can be transported at one time. However, under inappropriate winding conditions, it is a problem that the flexible devices fail and cause great economic loss. Therefore, the winding process is very important process for mass production of flexible devices.

Figure 2 shows a typical winding defect phenomenon that occurs in the winding roll. Telescoping shown in Figure 2 (a) is a state in which the roller layer is slipped in the axial direction. Star defect shown in Figure 2 (b) are the appearance of the wrinkles generated inside the roll observed from the side of the roll. It has been reported that these

defects depend on the state of internal stress generated in the winding roll [1]. Once such a winding defect phenomenon occurs in the winding roll, the flexible device is damaged and the value as a product is lost. In particular, optical films for liquid crystal displays have very high product accuracy requirements, and even a slight scratch on the web surface, which is difficult to confirm visually, will be defective. In addition, since the internal stress changes from the time of winding due to changes in the surrounding environment such as temperature and humidity during storage and transport, wrinkles may not occur immediately after winding. Therefore, the prediction and prevention method of the defect by the change of the surrounding environment which occurs frequently at the time of transportation has become an extremely important technical issue. Therefore, studies on thermal stress analysis of winding rolls have been conducted so far [2-5]. However, all these studies deal with the internal stress state when the temperature of the winding roll reaches a steady state, and there is a problem that it is impossible to predict the failure that occurs in the process of changing the state inside the roll. On the other hand, Suzuki and others measure the stress change in the temperature and the radial direction of the roll to solve this problem, propose the unsteady thermal stress analysis of the winding roll considering the unsteady heat conduction, its theory and experiment We have prevented damage to the roll beforehand, but do not take into consideration the influence caused by the difference between the experimental value and the theoretical value caused by the decrease in film thickness of the film and the physical property of the film. In this study, we evaluated the physical properties of the thin film and measured the unsteady thermal stress of the roll in order to evaluate the influence on the internal stress by using the thin film.
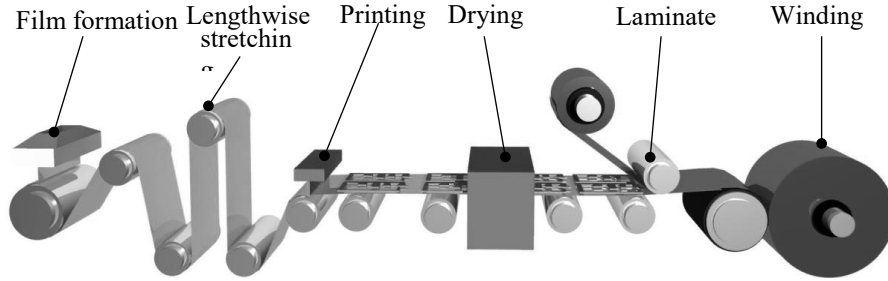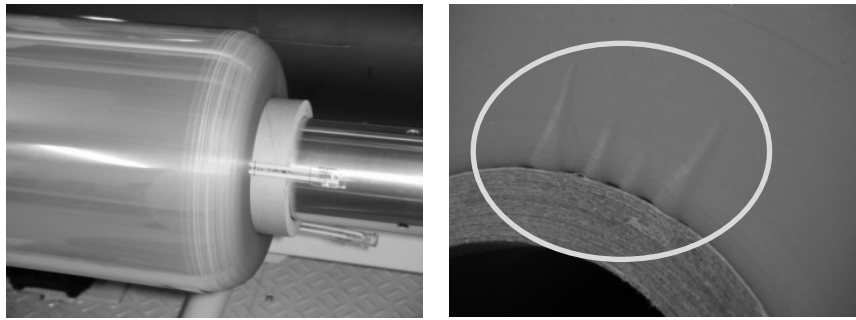
Figure 1 – Schematic of R2RPE production method

(a) Telescoping   (b) Star Defect

Figure 2 – Example of winding defect

Numerical Model

In the actual winding process, winding is often performed in a factory where the temperature environment is sufficiently controlled, and the temperature change during winding is often negligible. So, in this research, winding is performed under constant temperature, and the case where the temperature environment around the rolled roll changes after that is dealt with. Figure 3 shows the model of the winding roll.

When temperature difference occurs between the roll and the ambient temperature, the temperature distribution in the roll changes with the passage of time. The roll temperature T at time t is given by the following unsteady heat conduction equation.

$$\frac{\partial T}{\partial t} = a\left(\frac{\partial^2 T}{\partial r^2} + \frac{1}{r}\frac{\partial T}{\partial r} \right) \qquad \{1\}$$

The boundary condition and the initial condition of the innermost layer and the outermost layer are given by the following equations.

$$\left. \frac{\partial T}{\partial r} \right|_{r=r_c} = 0 \qquad \{2\}$$

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$$\lambda \left(\frac{\partial T}{\partial r} \right)\bigg|_{r=s} = -h_s \left(T_f - T_s \right)\bigg|_{r=s} \qquad \{3\}$$
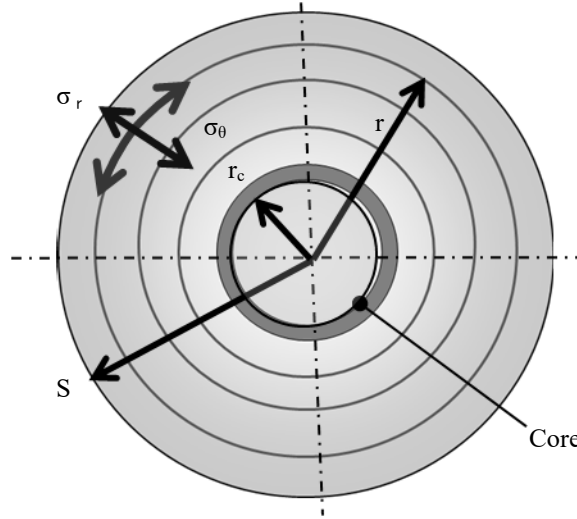
$$T\big|_{t=0} = T_0 \qquad \{4\}$$

Figure 3 – Analysis model

In addition, in winding in the atmosphere, the surrounding air is entrained between the web layers by the action of viscosity. As a result, since an air film intervenes between the film layers of the wound roll, it is considered that this air film affects the heat conduction of the winding roll. So we considered web layer and air layer as equivalent layers.

For the web layer and air layer shown in Figure4, the thermal resistance and equivalent thermal conductivity of this two layer can be obtained by the following equation.

$$R_{th} = \frac{1}{2\pi l} \left(\frac{1}{\lambda_w} \ln \frac{r_2}{r_1} + \frac{1}{\lambda_a} \ln \frac{r_3}{r_2} \right) \cong \frac{1}{2\pi r_1 l} \left(\frac{t_w}{\lambda_w} + \frac{h_a}{\lambda_a} \right) \qquad \{5\}$$

$$\lambda_{eq} = \frac{t_w + h_a}{2\pi r_1} \frac{1}{R_{th}} = \frac{t_w + h_a}{\dfrac{t_w}{\lambda_w} + \dfrac{h_a}{\lambda_a}} \qquad \{6\}$$

Furthermore, the density ρ and the specific heat c are similarly determined. Also, from the results of unsteady heat conduction analysis using these physical property values, the influence of the air film between the roll layers on the heat conduction is slight.[6] Therefore, in this research, we will use the thermal diffusivity of the web as the thermal

diffusivity a of the winding roll. However, in the case of materials with high thermal conductivity, such as metal webs, the influence of the air film is considered to be greater, so the influence of the air film on such materials is for further study.

The equation regarding the radial position r for the radial stress σ_r in the winding roll considering thermos elasticity is expressed by the following equation.

$$r^2 \frac{\partial^2 \sigma_r}{\partial r^2} + 3r \frac{\partial \sigma_r}{\partial r} + \left(1 - \frac{E_\theta}{E_r}\right)\sigma_r = E_\theta(\alpha_r - \alpha_\theta)\Delta T - E_\theta r \alpha_\theta \frac{\partial \Delta T}{\partial r} \qquad \{7\}$$

The boundary conditions for solving equation {7} are given by

$$r \frac{\partial \sigma_r}{\partial r}\bigg|_{r=r_c} + \left(1 - v_{r\theta} - \frac{E_\theta}{E_c}\right)\sigma_r\big|_{r=r_c} = E_\theta(\alpha_c - \alpha_\theta)\Delta T \qquad \{8\}$$

$$\sigma_r\big|_{r=s} = 0 \qquad \{9\}$$

Therefore, when performing unsteady thermal stress analysis according to equations {7} to {9}, the stress after a minute time has elapsed by substituting the amount of temperature change obtained by heat conduction analysis into ΔT.
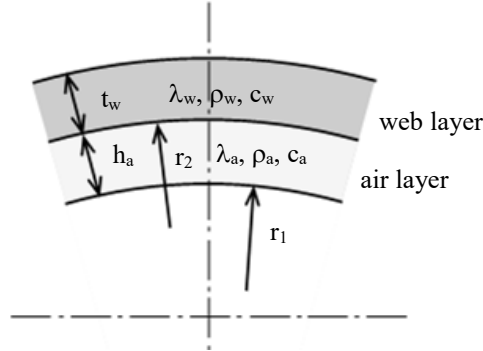

Figure 4 – Influence of air film on heat conduction of winding roll

EXPERIMENTAL METHOD

Physical Property Measurement

In the case of solving the thermal stress equation shown in the previous paragraph and finding the thermal stress inside the winding roll due to the change of temperature environment, it is necessary to measure physical property values such as the elastic modulus and linear expansion coefficient .Therefore, measurements of the Young's modulus in the radial direction and circumferential direction and measurement of the linear expansion coefficient were performed. In the measurement of Young's modulus,

the temperature environment was changed, and the temperature dependence of Young's modulus was examined.

The circumferential Young's modulus of the winding roll is determined by the tensile test of the film. The test method was performed according to the method specified in JIS K 7127 and JIS K 7161, the test speed was 0.2 mm/s, the size of the test piece was 15 mm in width, and the marking distance was 100 mm. As a result the Young's modulus of each temperature obtained by these is shown in Figure 5. From this result, the circumferential Young's modulus shows a tendency to decrease with the temperature rise.

When measuring the Young's modulus in the radial direction, the laminated film is used as a test piece, and measurement is performed by a compression test. At the time of measurement, a laminate of 200 pieces of film cut into a square was used as a test piece. Figure 6 shows the results of the compression test with the measurement temperature changed. Thus, it can be seen that the stress-strain diagram by the compression test becomes non-linear, and the slope of the stress-strain diagram becomes larger as the stress increases. Therefore, the Young's modulus in the radial direction is obtained by curve fitting the stress-strain diagram and using the derivative of its approximate function. In this study, we use the following equation as an approximation of the stress-strain diagram of the compression test.

$$\sigma = \alpha \varepsilon^{\beta} \qquad \{9\}$$

Therefore, the formula of radial Young's modulus can be expressed by the following equation by differentiating equation {9}.

$$E_r = A\sigma^B \qquad \{10\}$$

$$A = \alpha^{\frac{1}{\beta}} \beta, \qquad B = \frac{\beta - 1}{\beta}$$

Figure 7 shows the radial Young's modulus plotted against the change in stress. From this figure, it can be seen that the radial Young's modulus tends to increase as the stress increases. In addition, it can be seen that the stress-strain diagram almost matches even when the measurement temperature is changed, and the measurement temperature has no significant effect on the radial Young's modulus.
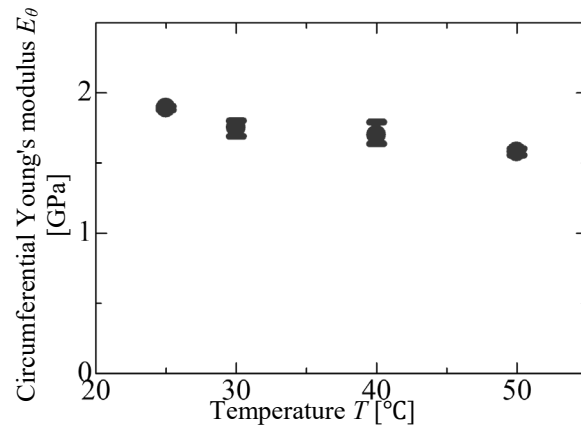
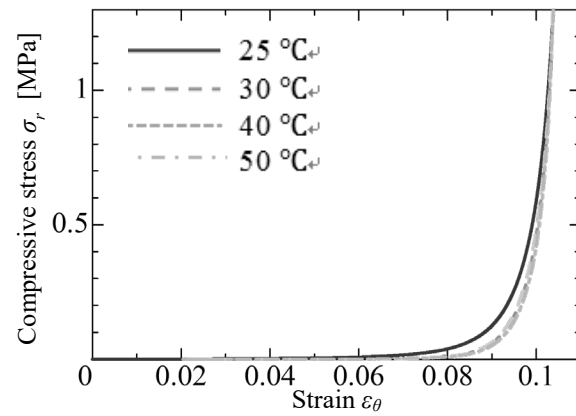Figure 5 – Relationship between temperature and Young's modulus

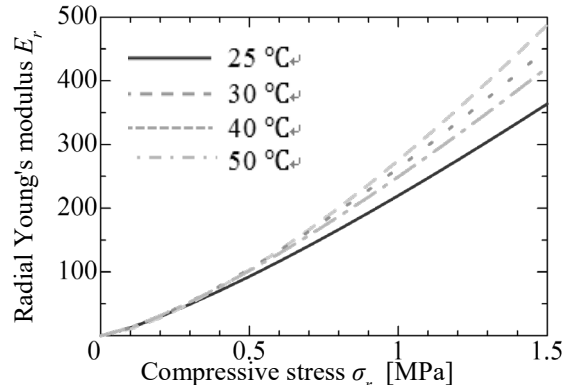Figure 6 – Stress-strain diagram of compression test

Figure 7 - Young's modulus and stress at each temperature

The linear expansion coefficient was measured using a noncontact laser displacement meter. The test pieces were manufactured in the same shape as the Young's modulus, and the size in the circumferential direction was changed to 180×15 (mm), and the number of stacked layers was changed to 750 in the radial direction.

Figure 8 shows the results of measuring the strain with respect to temperature change. As can be seen from the figure, it can be seen that the strain caused by the temperature rise in the radial direction is larger in both materials than in the circumferential direction. Therefore, it is understood that the linear expansion coefficient also exhibits strong anisotropy in the radial direction and the circumferential direction as well as the Young's modulus. In addition, the measurement results show little variation, good linearity, and it can be judged that accurate measurement can be performed. Therefore, the linear expansion coefficient was calculated from the relationship of the following equation.

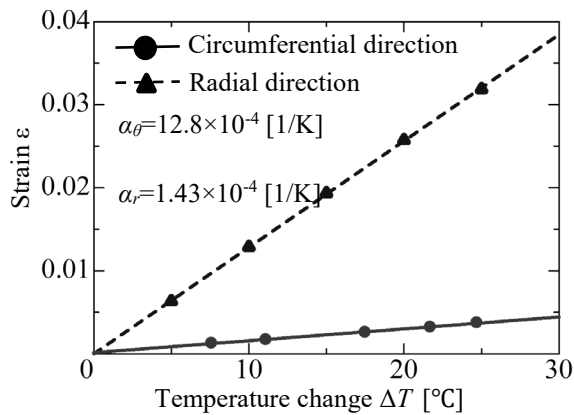$$\alpha = \frac{\varepsilon}{\Delta T}$$ {11}

Figure 8 – Linear expansion coefficient measurement result

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Transient Thermal Stress Measurement

Figure 9 shows a schematic view of the winding test used in this study. This winding device has a structure that reproduces the unwinding portion and the winding portion in the roll-to-roll production method, and the use of the nip roller can be selected as the winding portion. The speed, tension, and nip load can be set by the controller, enabling highly accurate transport and winding. The specifications of the winding test equipment such as tension during winding are shown in Table 1 and the physical properties of the core are shown in Table 2. The measurement of the internal stress in the radial direction in the roll and the temperature distribution in the radial direction were performed using a single film pressure sensor and temperature sensor shown in Figure10. This pressure sensor has the property that the resistance decreases as the pressure in the pressure sensing section increases. Therefore, by sandwiching these sensors in the roll during winding and measuring the resistance value, the pressure generated in the roll can be measured. After winding the film, the internal stress in the radial direction inside the roll was measured when the temperature around the roll was stepwise heated at room temperature + 10°C for 12 hours. In this experimental, A measuring position is dimensional radius r/rc-less, and decides, and it are (roller radius rc and film book radius r) and 3 points, an inner layer ($r / r_c = 1.05$), a middle layer ($r / r_c = 1.55$) and an outer layer ($r / r_c = 2.05$). Figure 11 shows a temperature environment tester manufactured to measure the internal stress at the time of temperature change. In this device, it is possible to adjust the temperature from room temperature to around 60 °C with a hot plate, and a thermistor was used to measure the temperature in the thermostatic chamber.
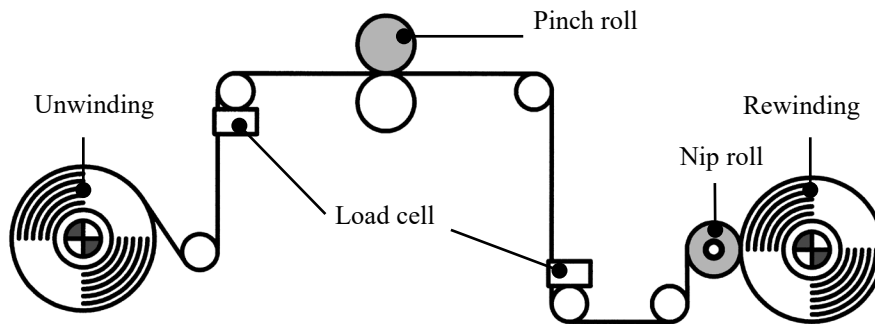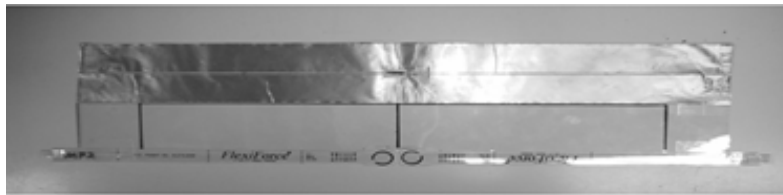

Figure 9 – Configuration diagram of test machine

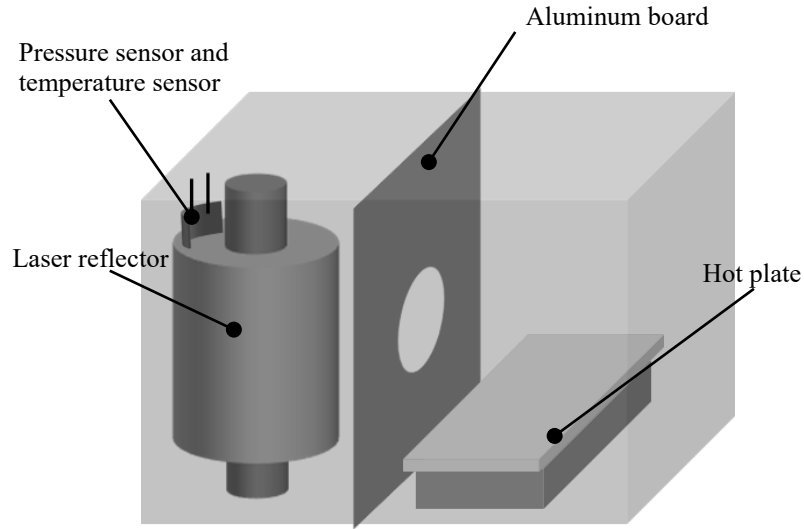Figure 10 – Pressure sensor and temperature sensor

Figure 11 – Configuration diagram of Thermostatic chamber

Winding velocity-V (m/sec)	1.0
Winding tension-T_w (N/m)	100
Film width-W (m)	0.35
Nip load-L (N/m)	235
Ambient temperature in room (℃)	22
Amount of ambient temperature change (℃)	10
Storage time in chamber (hour)	12

Table 1 – Experimental conditions on thermal winding model

Radius-r_c (m)	0.045
Thickness- h_c (m)	0.007
Young's modulus-E_c (Pa)	17.0×10^{-9}
Poisson's ratio-v_c	0.3
Thermal conductivity- λ_c (W/(m・K))	0.47
Density -ρ_c (kg/m³)	1940
expansion coefficient -α_c(1/K)	0.89×10^{-5}

Table 2 – Physical properties of core

RESULTS AND DISCUSSIONS

Figure 12 shows the change in internal stress in the radial direction inside the roll when the temperature around the roll is heated stepwise at room temperature (22 ° C) + 10° C for 12 hours. All plots in the figure show measured values. From Figure 12, the internal stress increases in the order of inner layer, middle layer, and outer layer. Therefore, it can be seen that the internal stress rises from the outer circumference to the

inner circumference. In the middle and outer layers, the internal stress increased gradually with the passage of time. It is thought that this was led to the increase in pressure between the films by thermal expansion from the outer peripheral side of the roll. However, the inner layer gradually decreased after the stress value decreased significantly. This is considered to be due to the occurrence of winding wrinkles and stress relaxation near the sensor pinched by the roll due to uneven heating in the constant temperature bath by the hot plate. It can be seen from Figure 12 that the internal stress variation due to temperature is very small. It was thought that the heat conduction took time because the ratio of the web layer to the air layer changed due to the thinning of the film, and the influence of the air layer became large relative to the web layer. Therefore, it has been found that the influence of the air layer has to be taken into consideration more as the film becomes thinner.
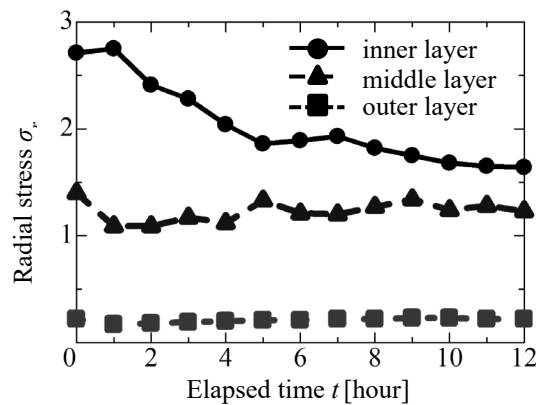

Figure 12 – Temperature environment test of internal stress

CONCLUSION

In this study, it is needed to evaluate physical properties of Young's modulus and linear expansion coefficient in each direction of roll necessary to calculate the theoretical value of internal stress, and to verify the influence on internal stress of roll in temperature change of thin film, temperature environment The internal stress was evaluated when raised to room temperature + 10 ℃ in a step-like manner with a testing machine. As a result, the temperature rise of the roll due to the temperature rise of the intermediate layer and the outer layer with the passage of time causes the stress to increase gradually. Moreover, it turned out that there is a possibility that defects such as star defect may be confirmed if heating cannot be performed sufficiently. It turned out to be done. It has also been found that the influence of the air layer has to be taken into consideration more as the film becomes thinner.

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