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Flow behaviours of liquid crystal droplets on polyimide alignment layers

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The flow behaviours of liquid crystal droplets on polyimide alignment layers and during assembling between two substrates have been directly observed. The droplet shape became elliptical with time on the rubbed polyimide layer, where the major axis of the elliptical droplet was parallel to the rubbing direction. Rubbing enhanced the wettability between the liquid crystal and the polyimide layer. During the assembling process, the liquid crystal droplets elliptically splayed out between two substrates assembled antiparallel. The liquid crystal molecules preferentially flowed parallel to the rubbing direction in a two-step flow mode; the droplet diameter slowly increased at the first step, and then it rapidly increased at the second step. The two-step flow of the droplet proved to be due to the thickness of the droplet dependent on the rubbing strength.

1. Introduction

High-resolution liquid crystal displays (LCDs), i.e. full colour active matrix thin-film-transistor (TFT) LCDs, are widely used in broadcasting, telecommunications, laptop and desktop computers, and other fields of use. Recently, LCDs with a wide viewing angle, comparatively fast response time, high contrast ratio and vivid colour performance have been utilized for televisions. The size of liquid crystal televisions is becoming larger and larger. The large-size LCD requires an improvement of the assembling process, because it takes a long time to insert liquid crystals into the narrow gap between two glass substrates using the conventional liquid crystal injection method based on the capillary phenomenon of liquids. Recently, a new liquid crystal injection method, 'one drop fill (ODF)', has been developed [1]. In the ODF process, liquid crystals are dropped on the glass substrate, and the substrate with the liquid crystals is assembled with the other substrate. The procedure can remarkably shorten the liquid crystal injection time for an LCD panel compared with the conventional injection method. The ODF process increases efficiency in the manufacture of large-sized LCD panels. It allows for a considerable shortening in the cell assembly time for panels. Thus, the ODF method is a revolutionary panel manufacturing process developed in the LCD field.

A number of liquid crystal droplets are dropped on a polyimide alignment thin layer coated on an indium–tin oxide (ITO) glass substrate, and they flow on the polyimide alignment layer during the assembling process. The flow behaviours of liquid crystals have been completely understood. Wu *et al.* have investigated the surface relationship between the orientation structure of liquid crystal molecules and polyimide alignment layer using a slit-flow viscometer [2]. They reported that the viscosity of nematic liquid crystal depends on the surface structures of polyimide alignment layers: the viscosity of the nematic liquid crystal of homeotropic orientation is higher than that of parallel orientation. Thus, the fluidity of the liquid crystal probably depends on the surface structure of the polyimide layer. However, very little has published concerning the fluidity of liquid crystals on polyimide layers.

We have investigated the flow properties of liquid crystal droplets on a polyimide layer. In this study, we have directly observed the textures of the liquid crystal droplets on the layers rubbed with different rubbing strengths and the flow behaviours during assembling between two substrates.

2. Experimental

2.1. *Materials*

A nematic liquid crystal mixture was used as a liquid crystal sample. The nematic phase temperature range is

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−30°C to 86°C. The polyamic acid polymer (PIA-X-465-GHI) for the rubbing layer was supplied by Chiso Co. Ltd.

2.2. *Rubbing treatment*

The polyamic acid solution was spin-coated on an ITO glass substrate. Pre-bake and post-bake treatments were applied at 80°C for 5 minutes and 210°C for 1 hour, respectively. The thin polyimide layer on the substrate was rubbed by a rubbing roller with rayon cloth using a rubbing machine.

The rubbing strength parameter (L) was estimated using following equation [3]

$$L = Nl(1 + 2\pi rn/60V) \tag{1}$$

where N is the cumulative number of rubbings, l the contact length in circumference of the rubbing roller, r its radius, n the number of revolutions per minute (rpm) of the roller and V the speed of the substrate stage. The L value can be defined to be the rubbing parameter when the rubbing pressure and rubbing cloth are fixed [3, 4].

2.3. *Direct observation for flow of liquid crystal droplet*

A direct observation system for flow of liquid crystal droplet during assembling glass substrates was prepared, as shown in figure 1. The liquid crystal droplet is observed under crossed polarizers. A quarter-wavelength plate (1/4λ plate) can be inserted under the analyzer. The upper glass substrate is moved using stepping motors with precision of 0.2 μm. The glass substrates are supported by quartz plates with 15 mm thickness to arrest the deflection of the glass plates. The flow of the droplet during assembling was recorded using a CCD camera, and the images were analyzed by a personal computer to estimate the droplet size.

A 3 ml liquid crystal was dropped on the bottom glass substrate by use of a microsyringe. After dropping, the upper glass substrate was depressed at various rates to create a 5 μm gap between the two glass substrates.

3. Results and discussion

3.1. *Shape of liquid crystal droplet on substrates*

The liquid crystal was dropped on an ITO glass substrate by use of a microsyringe. The volume of the liquid crystal was approximately 3.0 μl. The shape of the liquid crystal droplet was circular, as shown in figure 2. The droplet gradually expanded with increasing time, keeping its round shape.

A cross-extinction pattern was observed in the droplet under crossed polarizers. The pattern became clear with increasing time. Thus, the liquid crystal molecules radially aligned in the droplet on the glass substrate.

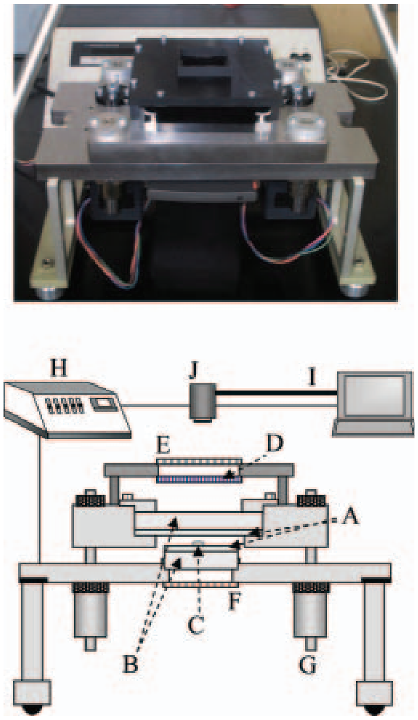


Figure 1. Direct observation system for flow of liquid crystal droplet. (A) ITO glass substrates: upper 94 × 94 mm; lower 50 × 50 mm, (B) quartz plates (thickness 15 mm), (C) liquid crystal droplet, (D) 1/4λ plate, (E) polarizer, (F) analyzer, (G) stepping motor, (H) controller, (I) personal computer, (J) CCD camera.

The liquid crystal was dropped on a rubbed polyimide layer which was coated on an ITO glass substrate. The rubbing strength parameter L was 38.0 mm. Figure 3 shows the change in the liquid crystal droplet on the rubbed polyimide layer with time, where the rubbing direction is horizontal, as denoted by an arrow. The droplet gradually became elliptical with time. The major axis of the elliptical droplet was parallel to the rubbing direction. The liquid crystal slowly flowed along the rubbing direction.

In general, since the rubbing process aligns polymer chains preferentially along the rubbing direction, liquid crystal molecules on the rubbed polymer orient parallel to the rubbing direction [3–11]. Therefore, the liquid crystals in the droplet gradually aligned in the rubbing direction near the interface between the droplet and the rubbed polyimide layer, after dropping. Additionally, an extinction pattern appeared preferentially on the major axis under crossed polarizers. It suggests that the liquid crystals in the bulk of the droplet also aligned in the rubbing direction.

Figure 2. Change in optical texture of liquid crystal droplet on ITO glass substrate with time. They were observed under crossed polarizers.

The change in the size of the droplet is shown in figure 4, where (a) denotes the changes in the diameter of the major axis of the droplet, and (b) the changes in that of the minor axis. The rubbing strength parameter of the polyimide layer was 38.0 mm. The major axis immediately increased in the initial stages, and then it gradually increased with time. The minor axis also increased in the same manner. The minor axis, however, almost remained unchanged after 200 s. Thus, the fluidity of the liquid crystal in the droplet is dependent on the surface structure of the substrate.

The anisotropic droplet shapes result from the rubbed surface structure of the polyimide layers and the anisotropic molecular shape of the nematic liquid crystals. Most of the liquid crystals on the rubbed surface are aligned to the rubbing direction. Thus, flow of the liquid crystal is governed by the oriented surface structure. The fluidity can be demonstrated by Miesowicz coefficients [12]. The three Miesowicz coefficients (η_1, η_2 and η_3) describe the shear flow of a nematic phase with three different director orientations. η_1 is a viscosity for the director parallel to the velocity gradient, η_2 for the director parallel to the shear flow axis and η_3 that for the director perpendicular to the shear flow and to the velocity gradient; usually $\eta_1 > \eta_3 > \eta_2$ [13–18]. The fluidities parallel and perpendicular to the rubbing direction were probably dominated by η_2 and η_3, respectively, when the liquid crystal flowed on the rubbed polyimide surface. Therefore, the liquid crystal preferentially spread along the rubbing direction.

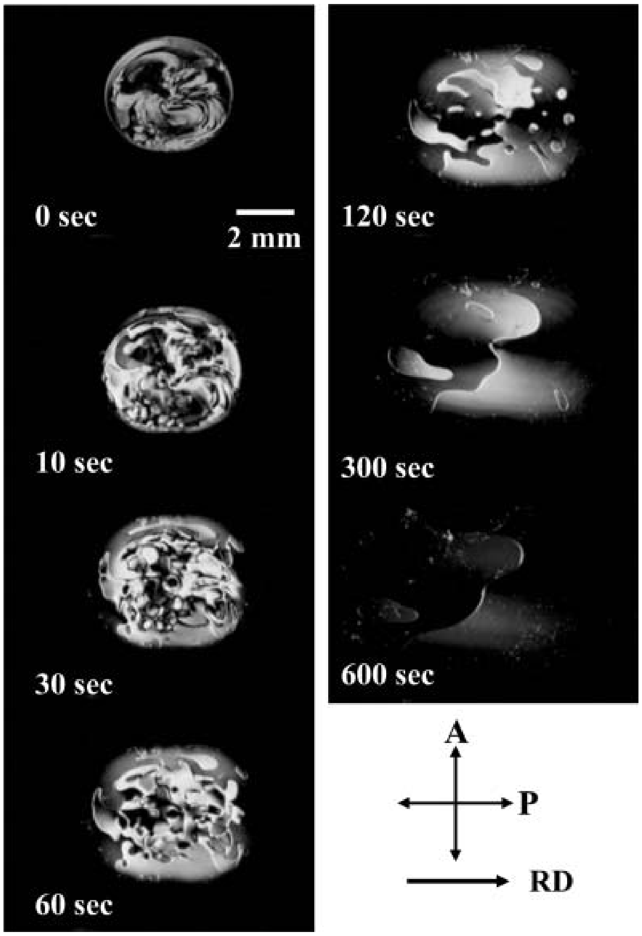
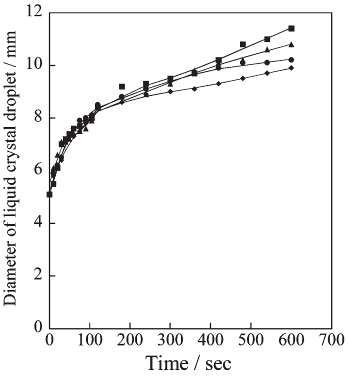


Figure 3. Change in optical texture of liquid crystal droplet on rubbed polyimide layer with time. They were observed under crossed polarizers. RD: rubbing direction; P: polarizer; A: analyzer.

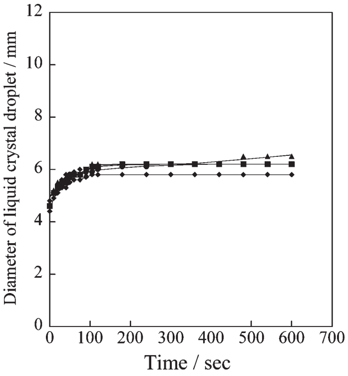
3.2. *Influence of rubbing strength of polyimide layer on size of liquid crystal droplet*

The fluidity of liquid crystal on the rubbed polyimide layer was different from that on the ITO glass substrate, as described above. In general, rubbing strength affects the anchoring energy and pre-tilt angle between liquid crystal molecule and polyimide layer: high rubbing strength tends to give high anchoring energy and low pre-tilt angle [8, 19–21]. Thus we have tried to investigate the influence of rubbing strength on fluidity of liquid crystal droplet.

Substrates with different rubbing strengths were prepared, i.e. $L = 14.0$, 30.1, 42.1 and 54.1 mm. Liquid



(*a*) Major axis of droplet

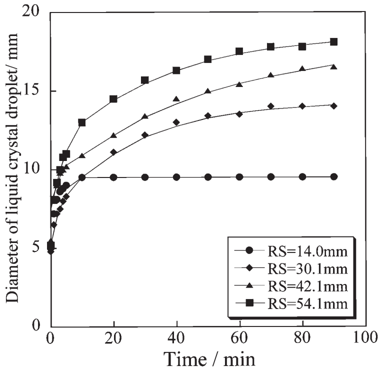


(*b*) Minor axis of droplet

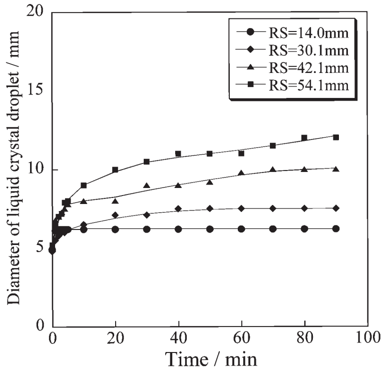
Figure 4. Changes in size of liquid crystal droplet on rubbed polyimide layer (rubbing strength parameter $L=38.0\,\mathrm{mm}$) with time. (a) Major axis of droplet; (b) minor axis of droplet.

crystal was dropped on these substrates, and changes in the droplet shapes were observed with time. The droplets became elliptical with time on all the substrates. The changes in size of the droplets were shown in figure 5. The major and minor axes immediately increased at the initial stage. They presently reached constant values on the substrates with comparatively lower rubbing strength, e.g. $L=14.0\,\mathrm{mm}$ and 30.1 mm. On those with higher rubbing strength, on the other hand, they gradually increased with time. They required a long time to reach a constant value. The higher the rubbing strength was, the larger the size of the droplet was.

Figure 6 illustrates the relationship between diameter of droplet and rubbing strength. The diameters were measured 90 minutes after dropping. The major and minor axes monotonically increased with increasing rubbing strength. The effect results from the orientation of liquid crystal molecules induced by the rubbed polyimide layer; i.e. the molecular orientation of liquid crystals decreases its viscosity. As a result, high rubbing strength enhances the fluidity of liquid crystal in droplet. This tendency results from a decrease in the



(*a*) Major axis of droplet



(*b*) Minor axis of droplet

Figure 5. Changes in size of liquid crystal droplets on polyimide layers with different rubbing strengths with time. (a) Major axis of droplet; (b) minor axis of droplet.

Miesowicz coefficient η_2 described above with increasing rubbing strength.

The droplets on the substrates were observed from the side to examine wettability of the surface between

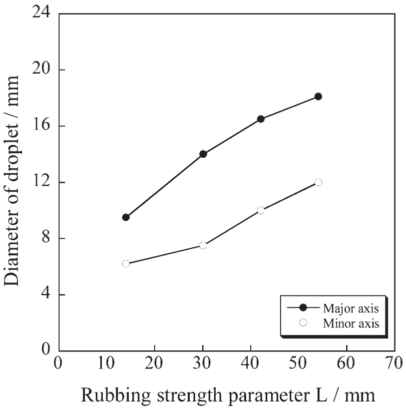


Figure 6. Changes in diameter of liquid crystal droplet on rubbed polyimide layers with rubbing strength parameter L. Open and closed circles denote major and minor axes, respectively.

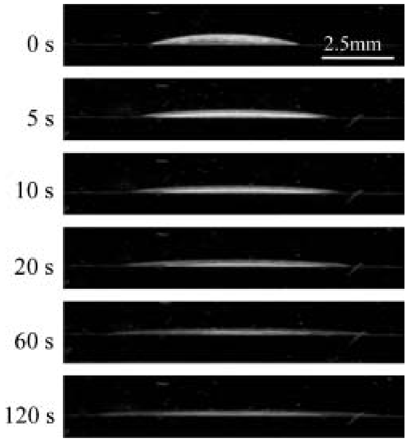


Figure 7. Side views of liquid crystal droplet on rubbed polyimide layer with L=42.1 mm.

the liquid crystal and polyimide alignment layer. Figure 7 shows the side view of a droplet on polyimide alignment layer with L=42.1 mm. The thickness of the droplet decreased with increasing time, while the droplet splayed out. The thickness of the droplet at 120 s was approximately a quarter as compared with that of the droplet immediately after dropping.

The change in contact angle of the droplet with time is shown in figure 8. The contact angle also decreased immediately in the initial stages on every substrate. These results prove the good wettability of the polyimide layer for liquid crystal. The contact angle almost remained unchanged on every polyimide layer after 120 s. There is not so large a difference between the behaviours among the substrates with different rubbing strengths at the initial stage. However, the contact angle measured after 120 s of keeping decreased with an increase in the rubbing strength, as shown in figure 9. It suggests that the rubbing treatment enhanced the wettability of the polyimide layer for the liquid crystal. Thus, the increase in the diameter of the droplet with increasing rubbing strength is probably due to the enhancement of the wettability.

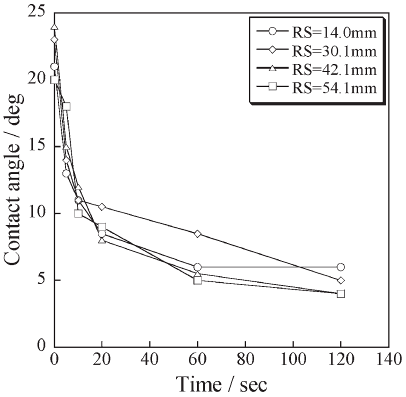


Figure 8. Changes in contact angles of liquid crystal droplets on polyimide alignment layers with different rubbing strengths.

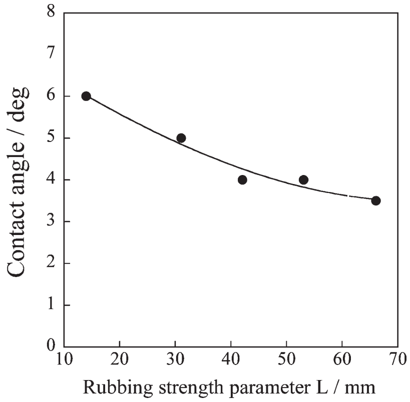


Figure 9. Change in contact angle with rubbing strength. The contact angles were measured after 120 s of keeping.

3.3. *Flow properties of liquid crystal droplet during assembling process*

The shape of a liquid crystal droplet depends on rubbing direction of polyimide alignment layer, as described above. In the assembling process of a liquid crystal cell, liquid crystal droplets are jammed between two substrates. Thus, the next target was to verify the fluidity of the liquid crystal during jamming a liquid crystal droplet between two substrates.

Liquid crystal was dropped on a polyimide-coated ITO glass substrate, and the other substrate was pressed down using the droplet flow apparatus shown in figure 1. The change in the optical texture of the droplet was observed under crossed polarizers. Figure 10 shows changes in liquid crystal droplets during assembling, where the polyimide alignment layers were rubbed with different rubbing strengths, i.e. L=14.0, 30.1, 53.9 and 66.1 mm. The rubbing directions of the upper and lower substrates were antiparallel to each other. Every droplet was jammed 5 minutes after dropping. The images at 0 s in figure 10 correspond to the moment when the upper substrate contacted the top of the droplet. The droplet was spread out during assembling, as shown in figure 10. The droplets elliptically splayed out independent of different rubbing strength. The major axis of the droplet was parallel to the rubbing direction. In addition, a dark field area appeared on the major axis of the elliptical droplet. Thus, the liquid crystal molecules preferentially aligned in the rubbing direction. The droplet size increased with increasing rubbing strength. These

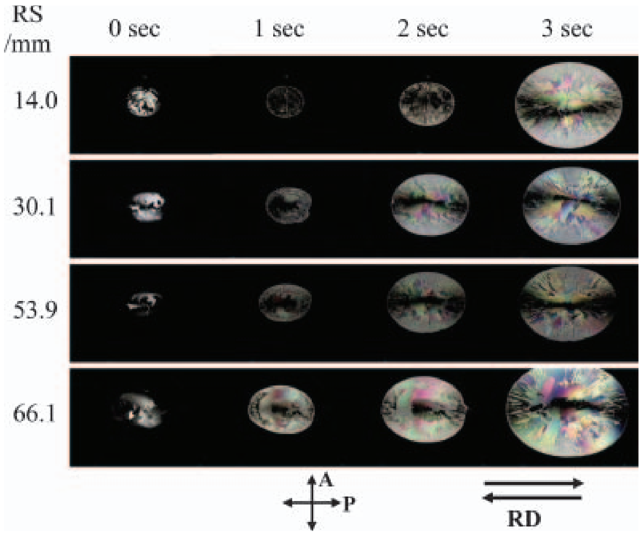
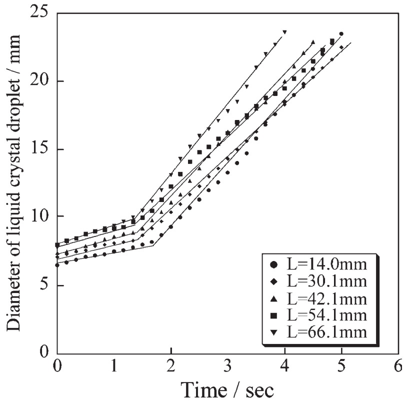
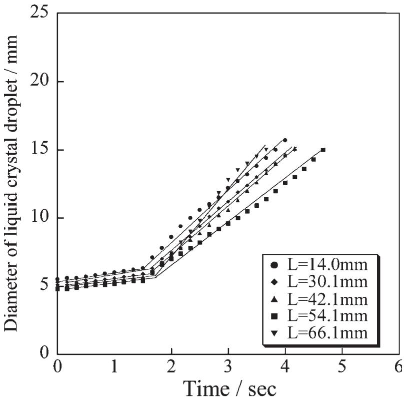


Figure 10. Changes in optical textures of liquid crystal droplets on polyimide layers with different rubbing strengths during assembling. They were observed under crossed polarizers. The rubbing directions of the upper and lower substrates were antiparallel, as shown by arrows.



(*a*) Major axis of droplet



(*b*) Minor axis of droplet

Figure 11. Changes in diameters of liquid crystal droplets on polyimide layers with various rubbing strengths during assembling process. (a) Major axis of droplet; (b) minor axis of droplet.

results suggest that the liquid crystal molecules in the droplet flow preferentially in the rubbing direction during assembling. This behaviour can be also demonstrated by the Miesowicz coefficients described above. The flow along the rubbing direction was dominated by smallest η_2, because the liquid crystal sandwiched between two rubbed substrates was aligned in the rubbing direction and was preferentially sheared in the direction during assembling. Thus, the droplet diameter parallel to the direction preferentially increased.

The diameters of the elliptical droplets were measured along both major and minor axes during assembling. The diameters of major and minor axes of the droplet are plotted against time in figure 11a and 11b, respectively. Both diameters increased with increasing time. The diameter slowly increased in the initial stages, and then they rapidly increased. It could be illustrated using two straight lines with different slopes, having an inflection point. The change in the droplet diameter was caused in two steps, i.e. the droplet flowed via a two-step process during assembling.

The two-step flow of the droplet is presumably due to the thickness of the droplet. It can be demonstrated using a schematic representation of assembling process illustrated in figure 12. In the first step, the upper substrate contacts the top of the droplet and the liquid crystal fills the gap between the upper and lower substrates, i.e. the diameter of the droplet on the lower substrate (d_1) is initially larger than that on the upper substrate (d_2), and d_2 is increased with time by assembling, until d_1 and d_2 are equal, as illustrated in figure 12. We have observed the change in the diameter d_2 by focusing a camera on the upper substrate and estimated the d_2 value. Thus, the droplet splayed out very slowly in the first step. The time of the first step t_1 corresponds to that of the inflection point where the diameter d_2 is probably the same as d_1. The droplet rapidly splayed out between the upper and lower substrates in proportion to the assembling speed in the second step (figure 12). Accordingly, the diameter drastically increased with time in the second step.

The flow velocity of the liquid crystal was estimated from the slope of each straight line. The relationship between flow velocity and rubbing strength is shown in figure 13a for the major axes and figure 13b for the minor axis of the droplet. Both the velocities of the first and second steps were plotted. They are almost independent of the rubbing strength, although the velocity of the second step becomes slightly larger at

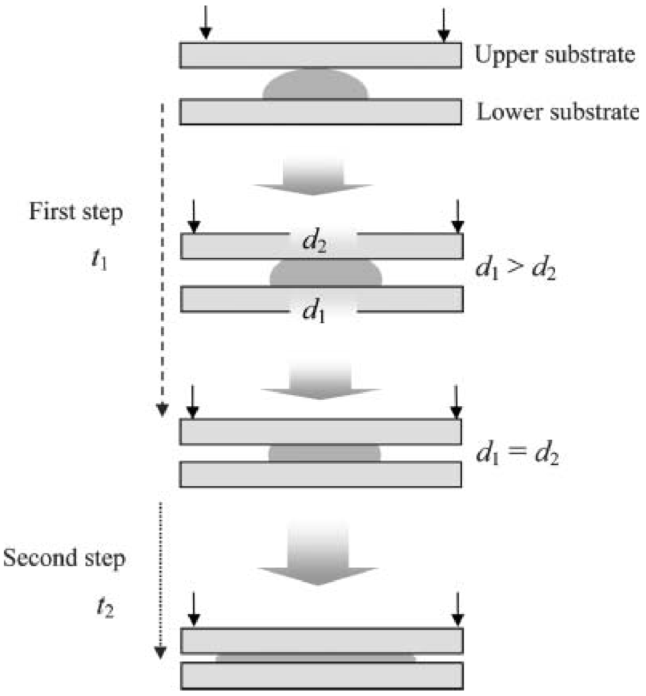
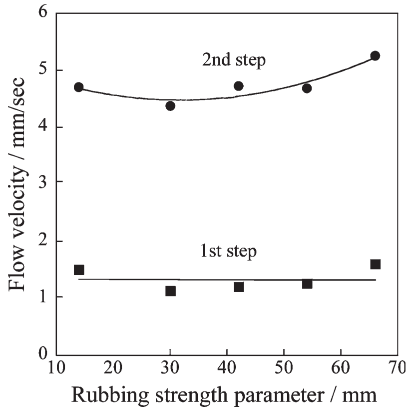


Figure 12. Schematic representation of assembling process. d_1 is the diameter of the droplet on the lower substrate; d_2 is that on the upper substrate.

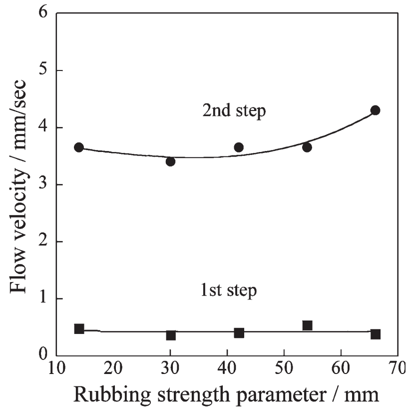
L=66.1 mm. Both velocities (1st and 2nd) for the major axis were larger than those for the minor axis. Consequently, the liquid crystal preferentially flows along the rubbing direction during assembling.

Figure 11 shows that the inflection point between the first and second steps gradually shifts to shorter times with increasing rubbing strength. Times of the inflection points (=t_1), estimated from the change in the major axis in figure 11a, are plotted against rubbing strength in figure 14. They monotonically decrease with increasing rubbing strength. This effect results from the decrease in the thickness of the droplet. The first-step time (t_1) depends on the thickness of droplet as described above; a thin droplet does not require so long to fill the gap between the two substrates in comparison with a thick droplet. The droplet thickness probably decreases with increasing rubbing strength because the droplet diameter increased with increasing rubbing strength, as shown in figure 6. Therefore, the decrease in t_1 is ascribable to the thickness of the droplet.

Moreover, the thickness of the droplet decreased with time, as shown in figure 7. When the droplets were kept for different times on the rubbed polyimide layer (L=54.1 mm) before assembling, t_1 monotonically decreased with increasing keeping time, as shown in



(*a*) Major axis of droplet



(*b*) Minor axis of droplet

Figure 13. Changes in flow velocity of the droplets on the rubbing films with rubbing strength. (a) Flow velocity in the direction of major axis for the droplets; (b) flow velocity in the direction of minor axis for the droplets.

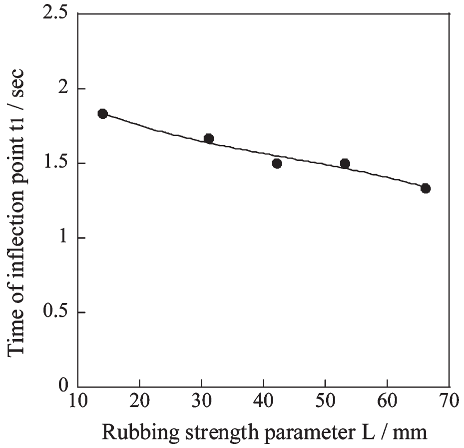


Figure 14. Change in inflection point between first and second step with rubbing strength.

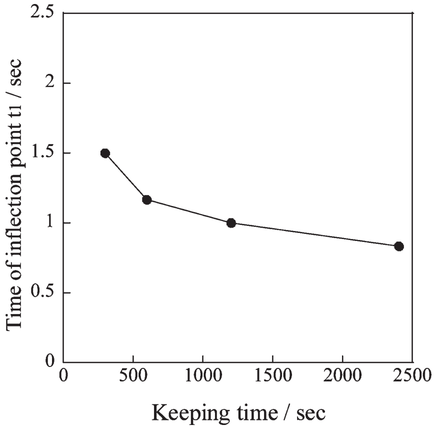


Figure 15. Change in inflection point between first and second steps with keeping time after dropping (L=66.1 mm).

figure 15. This result also indicates that the time of the first-step (t_1) is due to the thickness of droplet.

4. Conclusions

Flow behaviours of a liquid crystal droplet on polyimide alignment layers have been successfully observed using the direct observation system. The droplet shape became elliptical with time. The major axis of the elliptical droplet was parallel to the rubbing direction. The liquid crystal flowed slowly along the rubbing direction on the polyimide layer. This results from orientation of the liquid-crystal molecules induced by the rubbed polyimide layer. Rubbing enhanced the wettability between the liquid crystal and the polyimide layer. During the assembling process, the liquid crystal droplets elliptically splayed out between two substrates assembled antiparallel. The liquid crystal molecules preferentially aligned and flowed along the rubbing direction. The diameter of the droplet increased slowly in the first step, and then it rapidly increased in the second step. The inflection point between the first and second steps depended on the rubbing strength. The two-step flow of the droplet proved to be due to the thickness of the droplet.

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