INVITED PAPER Special Section on Electronic Displays Flexible Ultra-Thin Liquid Crystal Devices Using Coat-Debond Polyimide Substrates and Etched Post Spacers

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SUMMARY We developed flexible LC devices using coat-debond polyimide substrates with a low birefringence and etched post spacers, and clarified that flexible LCDs using post spacers with small spacer distance have a high flexibility without degradation of the image quality. This result ensured the feasibility of flexible LCDs using coat-debond method. *key words: liquid crystal display, flexible display, coat-debond method, polyimide substrate, etched post spacer*

1. Introduction

Flexible liquid crystal displays (LCDs) using plastic substrates have advantages of thin, light weight, and shock resistant. These features are promising for various applications such as rollable large screen televisions and wearable devices [1], [2]. Flexible LCDs also have high reliability because liquid crystalline materials have high stability against oxidation or moisture in the air. Therefore, flexible LCDs can be realized by using plastic substrates with a low gas barrier. For these reason, flexible LCDs have attracted significant interest as next generation displays. To realize flexible LCDs, the plastic substrate is required to have high dimensional stability against temperature during the fabrication process of the liquid crystal (LC) alignment layer and the thin film transistors [3]. These high temperature processes have become significant problems because most plastic materials have poor thermal stability.

To overcome this issue, a fabrication technique using polyimide (PI) substrates by the coat-debond method was proposed for organic-light-emitting-diode displays [4]–[8]. In this method, ultra-thin PI substrates with a high heat resistance are formed on the glass plates. By depositing the PI substrates on the glass plates, the dimensional stability of the PI substrates against temperature can be improved. After the device fabrication, the glass plates are debonded from the display device. This fabrication process is expected to be suitable for the high-temperature processes mentioned above. However, studies on the application of the coatdebond method for flexible LCDs have been difficult because of the following reasons.

One is that the achromatic transparent PI substrates

have a high optical birefringence [9], [10]. This optical feature leads to poor contrast ratios and narrow viewing angle ranges in flexible LCDs. In a previous study, we have evaluated the phase retardation of PI substrates [11]. However, the influence of the phase retardation of the PI substrates on viewing angle range of flexible LCDs has not become clear yet. The other challenge is a development of the spacer structure to maintain the thickness of the LC even in a curved state. In the case of particle spacer (see Fig. 1), the thickness of the LC is changed by the flow of LC in a curved state [12]. As a result, the image quality of the LC device is degraded. Alternatively, a post spacer structure fabricated by means of photolithography was proposed [13]. Post spacers are fixed on one side of the substrate and can maintain the thickness of the LC in a curved state (see Fig. 2). However, it is considered that flexible PI substrates using the post spacers with large spacer distance make it difficult to maintain the thickness of the LC because of the deformation of the PI substrates. Therefore, it is necessary to examine the effects of the distance between the post spacers on the image quality of flexible LC devices using PI substrates.

In this study, to investigate the influence of the phase retardation of the PI substrates on viewing angle range, we measured the angular luminance distribution of the PI substrates sandwitched between crossed polarizers. In addition, we fabricated flexible LC devices using post spacer structures by varying the spacer distance on the PI substrates, and discussed the effects of the distance between the spacers on







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the thickness of the flexible LCDs.

2. Evaluation of the Optical Characteristics of the Polyimide Substrates

We previously clarified that the phase retardation of the thickness direction of our PI substrate is significantly less than that of the ordinary PI substrate by spectroscopic ellipsometry [11]. It is expected that PI substrate with a low birefringence improve the image quality of flexible LCDs.

In this study, to investigate the influence of the phase retardation of the PI substrates on viewing angle range, we evaluated the optical anisotropy of the PI substrates which have a flexible molecular structure (Polyimide-A) and a rigid molecular structure (Polyimide-B). In order to fabricate an ultra-thin PI substrate, we coated the polyamic acid solution on a glass plate by the spin coating method and baked it at 270°C for two hours under a nitrogen atmosphere. We used polyamic acid solution (ECRIOSTM VICT-Bnp, supplied by Mitsui Chemicals, Inc.) as a Polyimide-A and polyamic acid solution (ECRIOSTM VICT-C, supplied by Mitsui Chemicals, Inc.) as a Polyimide-B. The thickness of PI substrates are 10 μ m and the light transmittance of the PI substrate using Polyimide-A is 90% for visible light and the substrate exhibits achromatic transparency (see Fig. 3).

To investigate the influence of the phase retardation of the substrates on viewing angle range, we measured the angular luminance distribution in all directions for the PI substrates sandwitched between crossed polarizers using a Conoscope (Autronic-Melchers). Figure 4 shows the angular luminance distribution of the PI substrates sandwitched between crossed polarizers. In the case of the PI substrate using Polyimide-B, light leakage occurred at an oblique viewing angle (see Fig. 4 (b)). In contrast, in the case of the PI substrate using Polyimide-A, the light leakage was significantly less in the wide viewing angle range (see Fig. 4 (a)). Figure 5 also shows photographs of the PI substrates sandwitched between crossed polarizers. Compared to the PI substrate using Polyimide-B, the light leakage from the PI substrate using Polyimide-A sandwitched between crossed polarizers was significantly less at an oblique viewing angle. From these results, it is evident that the PI substrate



Fig. 3 Photograph of the achromatic transparent PI substrate debonded from the glass plate.

using Polyimide-A is suitable for wide viewing angle flexible LCDs.

This result is attributed to the molecular structure of PI. We considered that refractive index of the thickness direction of the PI substrate using Polyimide-B became small compared to that of the horizontal direction because the direction of the main chain of the Polyimide-B with a rigid molecular structure trended to align in the horizontal direction of the substrate during fabrication process. As a result, the light leakage occurred in the wide viewing angle range because of the large birefringence of the substrate. In contrast, we considered that in the case of Polyimide-A with a flexible molecular structure, the direction of the main chain of Polyimide-A aligned randomly and the birefringence became small.

3. Fabrication of the Flexible LC Devices

To examine the effects of the distance between post spacers on the image quality of the flexible LCDs, we fabricated flexible twisted nematic (TN) mode LC device using the PI substrates of Polyimide-A and post spacers by varying the spacer distance. Figure 6 depicts the fabrication procedure of the flexible LC devices. First, we fabricated the ultra-thin PI substrates on glass plates by the spin-coating method (see Fig. 6 (a)). Next, we coated the negative photoresist (PR-200, supplied by Osaka Organic Chemistry Industry Ltd.) on the PI substrates (see Fig. 6 (b)), and irra-



Fig.4 Angular luminance distribution for the PI substrates sandwitched between crossed polarizers.



Fig. 5 PI substrates sandwitched between crossed polarizers (viewing angle: 30°).





Fig.7 Micrographs of the post spacers fabricated on the PI substrate.

diated UV light through a photomask (see Fig. 6 (c)). After the photoresist was removed by the photoresist developer except in the exposed area, we baked the post spacers at 150°C for an hour (see Fig. 6 (d)). Subsequently, we coated alignment layer (AL1254, supplied by JSR) on PI substrates and baked at 200°C for an hour, and conducted rubbing treatment (see Fig. 6 (e)). Then, we applied the sealing resin on the PI substrates and laminated the substrates (see Fig. 6 (f)). After that, we debonded the glass plate from the PI substrate (see Fig. 6 (g)), and injected the LC material (TD-1016LA, supplied by JNC) by the vacuum injection method (see Fig. 6 (h)). Finally, we debonded the LC devices from the other glass plate (see Fig. 6 (i)).

Figure 7 shows micrographs of the post spacers on the PI substrate. The shape of the post spacers is ellipse, and the average size of the post spacers is $100 \ \mu m$ in long diameter

and 50 μ m in short diameter. The height of the post spacers is 4.5 μ m. We fabricated the post spacers by changing the spacer distance progressively from 0.1 mm to 1.0 mm by 0.1 mm.

4. Observations of the Flexible LC Devices Using Post Spacers

4.1 Effects of the Spacer Distance on the Flexible LC Devices

Figure 8 shows the photographs of flexible TN mode LC device using post spacer with several distance before and after debonding process. The transmittance of the LC device sandwitched between crossed polarizers was high in all areas before debonding process (see Fig. 8 (a)). However, the transmittance decreased after debonding process with the exception of the area where the spacer distance was less than 0.2 mm (the inside of red dashed lines in Fig. 8). Low transmittance in large spacer distance area is caused by the change in the thickness of the LC during debonding process. We considered that the LC flow occurred and the thickness of the LC became thin because of the deformation of the PI substrates between the spacers (see Fig. 9). In the case of thin LC thickness, the direction of the polarized light could not rotate enough while the polarized light passed through the LC, and the polarized light was absorbed by output side polarizer. As a result, the transmittance of the LC device decreased in large spacer distance area.

In contrast, in the area where the spacer distance was less than 0.2 mm, we confirmed that the thickness of the LC was maintained constantly and the transmittance did not change during debonding process. In addition, we observed the stripe-like pattern in the area where the spacer distance was less than 0.2 mm in one direction, and was more than 0.2mm in another direction. These results indicates that post spacers with the spacer distance below 0.2 mm could maintain the thickness of the LC, but post spacers with the spacer distance over 0.2 mm could not maintain similarly.

Furthermore, we observed that the LC thickness in the peripheral area of the device was substantially thicker than original LC thickness because the LC flowed into the area during debonding (see Fig. 8 (b)).

To confirm the validity of above mentioned result, we fabricated two types of flexible LC devices using PI substrates and post spacers. The spacer distance was 0.1 mm and 1.0 mm, respectively. In this experiment, we used crossshaped post spacers to increase contact area of the spacers on the substrate. Figure 10 shows a micrograph of crossshaped post spacers on the PI substrate.

Figures 11 and 12 show the photographs of flexible TN mode LC devices using cross-shaped post spacer with the spacer distance of 0.1 mm and 1.0 mm, respectively, before and after debonding process. The transmittance of both the LC devices were high before debonding process, however, the transmittance of the LC device using cross-shaped post spacers with the spacer distance of 1.0 mm decreased after debonding process (see Fig. 12). In contrast, in the case of the LC device using post spacers with the spacer distance of 0.1 mm, we observed the thickness of the LC was maintained constantly and the transmittance did not change during debonding process (see Fig. 11). We confirmed that



Fig. 10 Micrograph of cross-shaped post spacers fabricated on the PI substrate (spacer distance: 0.1 mm).



(a) Before debonding

(b) After debonding

Fig. 11 Flexible LC devices using PI substrates and cross-shaped post spacers (spacer distance: 0.1mm).



(a) Before debonding

(b) After debonding

Fig. 8 Flexible LC devices using PI substrates and post spacers by varying spacer distance sandwitched between crossed polarizers.



Fig. 9 Deformation of the PI substrates between the post spacers with large spacer distance.



(a) Before debonding

(b) After debonding

Fig. 12 Flexible LC devices using PI substrates and cross-shaped post spacers (spacer distance: 1.0 mm).



(a) Using post spacers(b) Using particle spacersFig. 13 Flexible LC devices in a curved state.



Fig. 14 Flexible LC devices using post spacers with small spacer distance in a curved state.

the post spacers with small spacer distance maintain the LC thickness of flexible LCDs using ultra-thin PI substrates.

4.2 Optical Characteristics of the Flexible LC Devices in a Curved State

Figure 13 (a) shows a photograph of the flexible LC device using cross-shaped post spacers in a curved state. The spacer distance was 0.1 mm. For comparison, we have showed a photograph of the flexible LC device using conventional particle spacers in a curved state as shown in Fig. 13 (b). The image quality of the LC device using particle spacers was degraded because the thickness of the LC changes owing to the LC flow in a curved state (see Fig. 1 (b)). In contrast, the image quality of LC device using post spacers with the spacer distance of 0.1 mm was not degraded even in a curved state. This is because the post spacers can maintain the thickness of the LC and suppress the LC flow in the curved state (see Fig. 14). According to the above results, we clarified that flexible LC devices using

ultra-thin PI substrates and post spacers with small spacer distance have a high flexibility without degradation of the optical characteristics.

5. Conclusion

We fabricated achromatic transparent PI substrates with the thickness of 10 μ m by the spin coating method, and confirmed that the phase retardation of the PI substrate with flexible molecular structure is extremely small and the substrates are suitable for wide viewing angle flexible LCDs. In addition, we successfully fabricated flexible LC devices using coat-debond PI substrates and etched post spacers. We clarified that flexible LC devices using ultra-thin PI substrates and post spacers with small spacer distance have a high flexibility without degradation of the optical characteristics even in a curved state. These results ensured the feasibility of fabricating flexible LCDs using ultra-thin PI substrates with a high heat resistance by the coat-debond method.

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