Novel bonding technologies for flexible LCDs with mechanical stability under the pressing and bending deformation

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ABSTRACT

Flexible display devices are widely and extensively studied for using the applications such as smart cards, PDA, head mounted displays and all kinds of mobile display because of their lighter weight, thinner packaging, and flexibility. However, it has some obstacles such as mechanical stability and tight adhesion of two plastic substrates. In this presentation, we will suggest a new bonding technologies with rigid spacers and bonding materials, which will serve mechanical stability and good adhesion strength. The micro-contact printing method is used to place bonding material on the rigid spacers that may be easily applicable to roll-to-roll fabrication processes. The performances of prototype samples fabricated will also be demonstrated by this technology.

Keywords: flexible display, plastic substrates, mechanical stability, rigid spacer, micro contact printing method

1. INTRODUCTION

Recently, as the new display device replaced the rigid glass substrate by flexible substrate such as plastic, fabric and stainless foil, flexible displays are widely and extensively studied for coming new generation. Flexible LCDs expected to be a primary role of the new generation display devices because it has drawn much attention for use in applications such as PDA, smart cards and head mount displays by their distinct characteristics such as lighter weight, thinner packaging, flexibility, ruggedness and reduced manufacturing cost through continuous roll processing. Flexible LCDs are of great advantage to the popularization of flexible display due to their full growth of fabrication technology in FPD and their efficient light-control capabilities with low power consumption [1-4]. However, it has still decisive problems to fabricate commercially with current technologies based on the conventional substrates as well as their operation due to the flexibility of substrates. The serious obstacles to successful commercialization of flexible LCDs were highly nonuniform electro-optic (EO) properties under external mechanical distortions on the plastic substrates, which originated from LC distortion or cell gap variation [5-9]. One is mechanical instability of LC molecules, and the other is adhesion of two substrates because flexible displays always experience bending and folding stress.

In order to overcome the above problems, we have studied the essential technologies for flexible liquid crystal displays. Pixel-isolated LC (PILC) mode was proposed by photo-polymerization induced phase separation from LCs and prepolymer composite material [6, 10]. This mode confines the LC molecules inside pixels and has the good mechanical stability against the external deformation. However, there are some drawbacks in using this mode, such as complex fabrication process, residual polymers in active region which can reduce display quality significantly and increase the operating voltages [11]. PILC mode also can not achieve various kind of LC operating mode due to the possibility of rubbing only for one substrate.

In this paper, the suitable bonding technologies with rigid spacers and bonding materials will be suggested, which also will serve good adhesion strength as well as mechanical stability. The micro-contact printing method is used to place bonding material on the rigid spacers, which may be easily applicable to roll-to-roll fabrication processes. The performances of prototype samples fabricated by the technology will also be showed.

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2. MICRO CONTACT PRINTING METHOD WITH UV CURABLE EPOXY



Figure 1. Micro contact printing method using adhesive material

First of all, the novel technology by micro contact printing method was demonstrated using adhesive material of UV curable epoxy SK-9 (Summers Optical) for rugged flexible LCDs with the good adhesion strength as well as mechanical stability. Schematic diagram of fabrication procedure is shown in Figure 1. Adhesive material of UV curable epoxy SK-9 was spin-coated on a bare glass substrate. After spin-coated adhesive material (SK-9) on a bare glass substrate was printed on the top of the rigid spacer micro pillar structure by contacting and pressing, two flexible substrates are tightly assembled by the simple UV irradiation. The final structure could be obtained by LC injection. Multiple adhesion points of the rigid spacer of a commercial photo-resist material SU-8 (MicroChem.) showed the mechanical stability of device at the edge of each pixel. But when the top PES (polyether sulfone) substrate was laminate, adhesive material (SK-9) with low viscosity leaks into the pixel by pressure. In order to prevent the leakage, new rigid spacers with 4 columns structure was designed, which can isolate epoxy by capillary action [12].



Figure 2. Optical microscopic image of the cell. (a) Microscopic images of the multi-column rigid spacer with UV curable epoxy, (b) Microscopic images of LC textures under crossed polarizers.

The UV curable epoxy SK-9 was placed on rigid spacers using micro-printing method. Figure 2(a) illustrates the optical microscopic image of the new designed rigid spacers with four columns after assembling of the top and bottom substrate. As we expected, the surplus SK-9 is well confined inside four columns structure. From the result, the modification of rigid spacers prevented effectively the leakage of epoxy even without mixing with other materials. To examine the mechanical stability and the cell gap reliability of suggested configuration, the basic ECB (electrically controlled birefringence) LC sample was demonstrated by using conventional plastic substrate of PES. Homogeneous LC aligning agent Nylon 6 was used and rubbed in an anti-parallel direction to obtain planar LC alignment sample. A commercial nematic LC (ZKC-5085XX from Chisso) was utilized and its birefringence ($\Delta n=0.1515$) and dielectric constant ($\Delta \epsilon=9.57$). The cell gap was controlled 3μ m by rigid photo-resist spacers. The clear isolated rectangular shaped structure of adhesive polymer was observed in the texture of LC sample as shown in Figure 2(b). From the capillary effect, more self-aggregated polymer structure was obtained in our proposed multi column spacers, which can be more suitable and effective to show better adhesion properties and maintain stable cell gap against the external distortion. In order to check

the mechanical stability of our sample, the experimental set up showed in figure 3. In our first mechanical stability test (figure 3(a)), the sample is fixed in the air with increasing the additional loads to check the adhesion reliability. The measured maximum capable loads without breaking sample were about 4.56 N/cm². Our sample could also tolerate even with extreme bending of R=1.7cm (figure 3(b)). From these results, our proposed technique can be used to realize the robust flexible LCD with reliable device performance.



Figure 3. Experimental set up for mechanical stability. (a) bonding strength test, (b) bending deformation test (R is the curvature radius.).

In summary, micro contact printing assembling technique was demonstrated by using the multi column spacer array for the stability-enhanced flexible LCD. Designed column spacer array created the confined structure of adhesive material by capillary effect and supported the good adhesion of two plastic substrates. From various experimental results, stabilized flexible LCD can be obtained within a simple fabrication procedure.

3. MECHANICAL STABLE STRUCTURE WITH AGAROSE COMPOSITE

In the previous bonding method for flexible LCD [13], UV curable epoxy was used as adhesive material. But there is some problem generated in assembling process of contacting and pressing. UV curable epoxy (SK-9) is overflowed into the pixel due to its low viscosity. This drawback reduces the image quality and the reliability of device seriously by the existence of excessive adhesive material. In the previous work, rigid spacer with multi column structure was designed for using the UV curable epoxy and could confine the adhesive material into space between the columns by the capillary effect. In the second work, the mixture of agarose polymer and UV curable epoxy (SK-9) as the adhesion layer was proposed and tested. This mixture exists as the gel type and it doesn't react with LC molecules. Also, because the viscosity of adhesive mixture can be controlled by temperature, the adhesive mixture does not overflow into the pixel without the rigid spacer of the specially designed structure. This mixture can tightly attach two substrates after micro contact printing and irradiating UV light.

Agarose of the mixture is a natural colloid extracted from sea weed and used to make gels. Since agarose gels have pores of large size, we used the agarose as a container of epoxy to prevent leakage. At first, rigid spacers on one plastic substrate was fabricated by using photo-lithography. The used agarose is high gel-strength type (gel point (1.5%): $36.0 \ ^{\circ}C\pm 1.5 \ ^{\circ}C$, remelting point (1.5%): $88.0 \ ^{\circ}C\pm 1.5 \ ^{\circ}C$, gel strength(1%): $\geq 0.12 \ \text{g/cm}^2$, gel strength (1.5%): $\geq 0.25 \ \text{g/cm}^2$, and moisture: < 7%). SK-9 (Summers Optical) is used as an adhesive material. We added EDTA (ethylene diamimetetraacetic) to the mixture of distilled water and agarose of powder. The EDTA helps bonding epoxy being scattered well in the mixture. The initial muddy mixture is changed to the clear state at the temperature of 150 \ ^{\circ}C. After

the mixture cooled down until 60 $^{\circ}$ C, it was spin-coated at the 6500 rpm during 50 sec on the substrate. Owing to the transition of viscosity between gel-state and liquid-state by temperature, fabrication was easily accomplished without flooding or infiltration of the adhesive material into the pixel.

Figure 4 shows schematic illustration of sample fabrication process using the proposed bonding layer for the LC device.

The alignment layer was coated before the rubbing process. The micro contact printing method was used for place the composite materials of UV curable epoxy and agarose on rigid spacers. We dropped LCs on the substrate with the rigid spacers and laminated the top plastic substrate on rigid spacers. The LC device with tight adhesion of two substrates could obtain by UV irradiation for polymerization of UV curable epoxy.



Figure 4. The fabrication process using the proposed adhesive layer. (a) prepared the adhesive layer on the rigid spacers, (b) dropped LCs and (c) cured the adhesive materials by UV irradiation.



Figure 5. Microscopic and FESEM image of sample. (a) Microscopic image of inner pixels after assemble process, (b) FESEM images of the adhered section, and (c) Microscopic images of the driving test.

Figure 5(a) shows the microscopic images of the formed adhesion layer on the polymer wall after assemble process. Although the rigid spacer with multi columns for the capillary effect used not, it is fixed on the polymer wall without overflowing into the pixel as shown in the figure 5(a). Also, the strong binding of substrates was confirmed by bending and pressing test. Fig. 5(b) shows the cross-sectional view of the adhered section using FESEM (Field Emission Scanning Electron Microscopy). It is confirmed that the bonding material is stabilized on the polymer wall. Electrically controlled birefringence (ECB) LC cell was fabricated by the proposed process. Figure 5(c) shows the microscopic textures of the driving test (0V to 15V). The unclean area around near side of the polymer wall is occurred by the property of LCs.

In summary, the bonding technique with the proposed adhesive mixture can be very useful for flexible LCD applications. The mechanical stability can be secure against external deformations by bonding each sub-pixels of the micro-structure (rigid spacers). This method can prevent the overflowing of the adhesive material into the pixels. The adhesion layer on the rigid spacers is stable and no reacting with LCs. This is a great advantage in using all of rigid

spacers without a design limit for the stability.

4. FLEXIBLE LCD BY MICROCONTACT PRINTING USING THERMAL CURABLE ADHESIVE

4.1 Novel material for a device with tight adhesion

To commercialize the flexible LCDs, novel adhesion techniques for roll-to-roll process is necessary. In the early work, the UV curable epoxy such as the SK-9 confined inside the multi column rigid spacer, and the adhesive mixture of UV curable epoxy and agarose without a design limit of the rigid spacer were used to tightly attach the bottom and top substrate. However, it is difficult that these adhesive materials apply to the real fabrication for mass production because it was not easy to use UV curable epoxy in practical manufacturing due to cutting off the UV light by black matrixes (BM).

In order to solve this problem, NOA83H (Norland Co.) as thermally curable epoxy instead of UV curable epoxy was used [14]. However, the material makes it difficult to process using micro-contact method or spin casting because of high viscosity. Viscosity of the material was controlled with THF (Tetrahydrofuran) as a solvent with different weigh ratio.

4.2 Experimental



Figure 6. The schematic diagram of a manufacturing process. (a) Micro contact printing process for transcribe bonding layer on rigid spacers, (b) Pre-baking process on the bottom substrate, (c) One drop filling process of LCs, and (d) Covering the top substrate on rigid spacers and hard-baking process.

Flexible substrate of ITO-coated PES (polyether sulfone) film was 200μ m. Both of the ITO-coated PES substrates were spin-coated with a homogeneous alignment layer and anti-parallel rubbed. The multi column rigid spacers of negative photo resist SU-8 were formed on one of the substrate. Figure 6 is shown in the schematic illustration of fabrication procedures from the step of micro contact printing to the last step of obtaining final LC cell. The mixture of NOA83H and THF solution was transferred to rigid spacers using micro contact printing method (Figure 6(a)). For the evaporation of solvent, the sample was prebaked at 80°C/7.6mmHg in vacuum to evaporate THF and change the state of thermally cured polymer in first order hardening state (Figure 6(b)). The LCs filled up by ODF (one drop filling) process without any reaction with first order hardened thermally curable epoxy (Figure 6(c)). The sample was annealed to 125°C/7.6mmHg in vacuum for 60 min (Fig. 6(d)).

4.3 Mechanical stability under the pressing and bending deformation

The mechanical stability of flexible LCDs fabricated by micro contact printing method with thermally curable material on multi column rigid spacers was examined under bending deformation and pressing deformation. Two kinds of flexible LCDs samples were prepared. One is a normal sample that cell gap is maintained by conventional ball spacers. The other is a plastic LCD sample using multi-column rigid spacer of SU-8 photo resist. We can see clearly stabilities against point

pressure using sharp tip for two samples as shown in Figure 7. In normal sample (with no rigid spacers), LC alignment is severely distorted due to cell gap instability and the propagation of the distortion to the bulk LC when the pressing deformation is applied to the sample. Such deformation propagates over a quite large area and results in the degradation of the EO properties. On the other hand, LC sample with the vertical rigid spacers is kept LC alignment well against external pressure. In order to confirm the attaching strength between two substrates, the maximum tension to sustain the cell gap uniformity was measured like the experimental set up in figure 3(a). One substrate was fixed and added weights to the other substrate until the breakage of the LC sample. The averaged maximum tension value for the attachment of two substrates was 11.2N/cm². This result proves tightly attaching of two substrates.



Figure 7. Test of bonding strength of flexible LCD samples. (a) without rigid spacers (b) with rigid spacers; after point pressure on LC sample.



Figure 8. Experimental set-up for V-T curve measuring for bending deformation. (a) Experimental setup for bending test (b) Photograph of experiment (c) Normalized transmittance vs. the applied voltage for the PILC cell in various bending states. The radius of curvature R represents the degree of bending.

An electro-optical characteristic under bending deformation was measured with flexible LCD sample that fabricated with patterned rigid spacers and thermally curable polymer. As previously reported [6,10], the V-T (voltagetransmittance) curve of normal plastic sample under bending stress deformed significantly due to by instability of LC alignment and cell gap variation. Figure 8 shows the V-T behavior of our flexible LCD sample under the bending deformation. The LC sample was bent using a pair of linear translation stages and placed between two crossed polarizers. The degree of bending is typically manifested by the curvature radius of the LC sample (R) that can be defined in the experimental set-ups shown in the Figure 8(a) and (b). As R decreases, the degree of bending is increased. The measured transmittance for the LC sample is shown as a function of the applied voltage for various bending states in Figure 8(c). Under bending deformation, the LC sample shows nearly the same behavior in a wide range of the applied voltage except for the low voltage regime as shown in Fig 8(c). It is clear that LC molecular distortions due to the bending stress are effectively reduced in the LC sample by the rigid spacer and the bonding layer for tight attachments. This fact connotes that the LC alignment is maintained under bending stress that is one of the key requirements for flexible LCDs. Figure 9 shows the rugged prototype flexible LCD samples of the size of 3 inch (figure 9(a)) and 2 inch (figure 9(b)) under the bending state. As shown in the magnified photograph, the alignments of LCs are stable against bent circumstances. These flexible LCD samples formed by the micro contact printing method of thermally curable epoxy would play an important role in fabricating high-performance flexible LCDs.



(a) (b) Figure 9. A prototype flexible LCD sample; (a) 3 inch and (b) 2 inch

5. CONCLUSION

In this paper, novel adhesion technologies for rugged flexible LCDs were estimated in various methods. For the tight adhesion of two plastic substrates, micro contact printing method with UV curable epoxy was used on multi column rigid spacers for confining the epoxy by the capillary effect. The composite materials of UV curable epoxy as adhesive material and agarose as a container were also used. The composite materials could prevent the leakage of epoxy into the pixel without a design limit of the rigid spacer as well as showing good adhesion. However, it was a limit to use UV curable epoxy in practical manufacturing due to cutting off the UV light by black matrixes (BM). In order to solve this problem, micro contact printing method using NOA83H (Norland Co.) with thermally curable epoxy instead of UV curable epoxy was suggested and tested. High viscosity of thermally curable epoxy was controlled with THF. It showed very good mechanical stability against pressure and bending. Also, we could adopt ODF process of LC without any reactions between contacting polymer and LC molecules by changing the thermally curable epoxy in the first order hardening state. As the thermally curable epoxy shows mightier sticking property than UV curing polymer, the fabricated samples shows stronger adhesive power between upper substrate and lower substrate than former works. The proposed

method can be possible to apply to practical process of flexible LCDs directly.

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