Research on Peeling Performance of Acrylic Photoresist with Isolated Island Pattern for OLED Display

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Abstract

Acrylic photoresists usually act as functional layers inside OLED display because of its low-temperature curing characteristic, which brings new functions to the OLED display, such as the MLA (micro lens array) technology and the COE (color filter on encapsulation) technology. Inside the OLED display, some acrylic photoresists are required to be made into isolated island pattern (named Island OC), and they are further covered with another layer of acrylic photoresist (named Top OC). The Island OC is easy to peel because its contact area with substrate is very small, and it is probably greatly influenced by the layer of Top OC. This work mainly researched about the influences on the peeling performance of the Island OC by the substrate, Top OC and itself. It's confirmed that the primary cause for peeling of Island OC should be its own insufficient chemical resistance, and improving chemical resistance of Island OC could completely avoid peeling. In addition, the related mechanisms about peeling of Island OC, the residual stresses and the chemical resistance of acrylic photoresist have also been studied.

Author Keywords

Acrylic photoresist, peeling, chemical resistance, residual stress, solvent infiltration, OLED.

1. Introduction

Acrylic photoresist is generally negative photoresist, which owns advantages of good leveling, high transmittance, low cost, and low curing temperature. It can be fabricated on top of OLED devices, and act as functional layers to make OLED displays realizing new technologies, such as MLA (micro lens array, **Figure 1a**) [1] and COE (color filter on encapsulation, **Figure 1b**) [2]. The formula of acrylic photoresist is mainly composed of polymer, monomer, photo initiator, additive and solvent. The polymer is one of the most important components, and its general molecular structure is shown in **Figure 2a**. It was reported in previous literature that adding silane coupling agent to the formula of acrylic photoresist, the adhesion force at the interface of substrate can be significantly improved, thus avoiding peeling. [3] Because the silane coupling agent could form hydrogen bond and covalent bond at the interface of substrate, the mechanism is shown in **Figure 2b**.

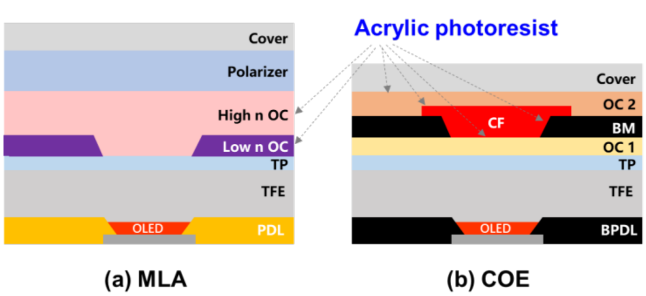

Figure 1. OLED panel structure with (a) MLA and (b) COE technology, and applications of acrylic photoresist.

However, inside the OLED display, some acrylic photoresists are required to be made into isolated island patterns (named Island OC), and will be further covered by another layer of acrylic photoresist (named Top OC), as shown in **Figure 3a**. For Island OC, it should be much easier to peel off compared to other patterns. Because on one hand, the contact area between Island OC and substrate is relatively smaller. On the other hand, it may be further affected by solvent infiltration and residual stress caused by Top OC. So that the situation for Island OC is much more complicated. And it may happen like that, no peeling happens at first after the Island OC is fabricated, which means the adhesion force of the Island OC is probably strong enough and not a problem. But finally, it still peels after the preparation of Top OC on top of the Island OC as shown in **Figure 3b,** which certificate that the peeling performance is not only determined by the Island OC itself, but also greatly influenced by the Top OC layer.

This work mainly researches the peeling performance of Island OC shown in **Figure 3b**. The peeling model and force analysis of the Island OC and are carried out firstly. The influences on the peeling performance by the Island OC**,** substrate and Top OC has been studied. In addition, the related mechanism about peeling has been studied.

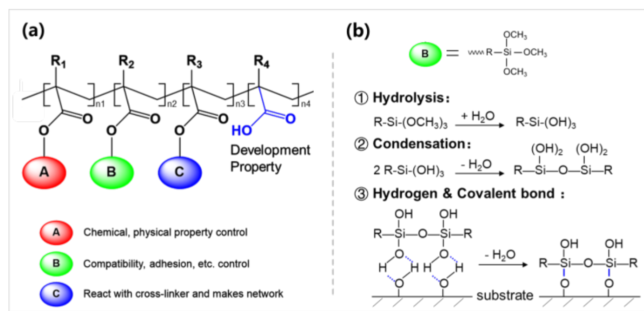

Figure 2. (a) general molecular structure of acrylic polymer from acrylic photoresist; (b) mechanism for silane toimprove adhesion force of acrylic photoresist.

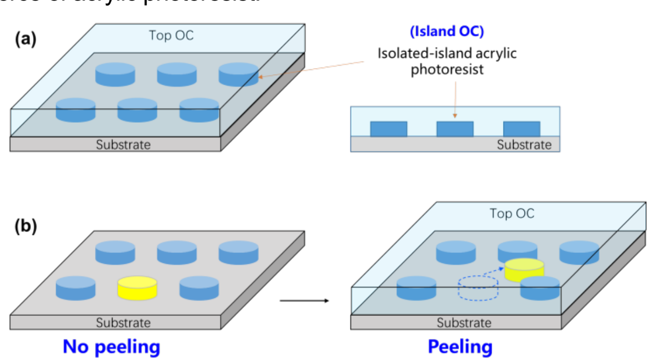

Figure 3. (a) target schematic diagram for isolated island pattern of acrylic photoresist; (b) peeling of Island OC after fabrication of Top OC.

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2. Experimental

2.1 Materials

The Island OC and Top OC are both negative-tone acrylic photoresists, and their film samples are generally prepared following the same process: coating → VCD → prebake (PB) → exposure (EXP) → development (DEV) → IUV (on/off) → OVEN (< 100°C).

2.2 Characterization

Chemical resistance of Island OC: the film sample of Island OC with the same thickness is prepared on glass substrate, then soaking it in stripper (mainly containing N- methyl formamide) for 3 minutes, taking it out and blow-drying it, and finally characterizing the contents of N-methyl formamide inside Island OC by TOF-SIMS. TOF-SIMS test conditions: positive ion mode, voltage 10KV, current 10nA, sampling frequency 10 s/cycle.

Residual stress of Top OC: the film sample of Top OC with a certain thickness is prepared on wafer, measuring the curvature change of the wafer before and after preparation of Top OC, and calculating the residual stress value of the film of Top OC by the Stoney formula.

3. Results and discussion

3.1 Analysis of peeling performance of Island OC

Firstly, the force analysis about peeling of Island OC should be carried out. As shown in **Figure 4**, **F1** means the residual stress of Top OC that acts on Island OC, **F2** means the adhesion force of Island OC at the interface of substrate, **F3** means the cohesive force of Island OC. It's not difficult to conclude that, firstly when **F3>F1>F2**, the peeling of Island OC will happen; secondly when **F3<F1** and **F3<F2**, then the cohesive failure of Island OC will occur; finally, when **F3>F2>F1**, neither peeling nor cohesive failure will occur. Among them, the cohesive force **F3** is mainly determined by the curing ratio of Island OC; the adhesion force **F2** is mainly affected by the surface energy of substrate, which can be characterized by the water contact angle on the surface of substrate; The residual stress **F1** is mainly determined by the curing ratio of Top OC, which can be test directly. Therefore, the investigation on influences of above factors on peeling of Island OC will be conducted.

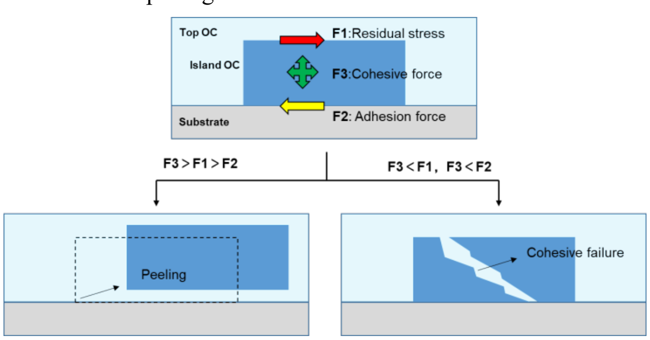

Figure 4. force analysis for Island OC and different failure results.

3.2 Research on peeling results of Island OC under different experimental conditions

Table 1 shows the peeling results of Island OC under eight different experimental conditions (namely DOE 1-8). Obviously, peeling ratio of DOE 2 is the highest, with high residual stress of Top OC, high water contact angle of substrate and medium chemical resistance of Island OC. Comparing the peeling results of DOE 1 and 2, it shows that decreasing the water contact angle of the substrate from high level to the medium level, the peeling ratio of Island OC could be greatly improved. Because when the water contact angle of the substrate decreases, the surface wettability of substrate will be improved, then greater adhesion force of Island OC on the substrate should be obtained because of greater contact area of Island OC on the substrate. However, DOE1 still has a low incidence of peeling, which shows that the water contact angle of substrate is only one of the main reasons for peeling of Island OC, but not the primary cause.

DOE 2-4 studies influence of different residual stresses of Top OC on peeling ratio of Island OC, with high water contact angle of the substrate and medium chemical resistance of the Island OC. The results show that decreasing the residual stress of Top OC from high level to medium and low level respectively, the corresponding peeling ratio of Island OC also decreased from high level to medium and low level respectively, which means the change trend of them is consistent. However, it is also impossible to completely eliminate the occurrence of peeling for Island OC. It concludes that the residual stress of Top OC is also not the primary cause for the peeling of Island OC.

Table 1. peeling ratios of Island OC under different experimental conditions.

Layer	Experimental Conditions		DOE						
			1	2	3	4	6	7	8
Top OC	Residual stress	High	√	√			√		
		Medium			√			√	
		Low				√			√
Island OC	Chemical resistance	High					√	√	√
		Medium	√	√	√	√			
Substrate	Water contact angle	High		√	√	√	√	√	√
		Medium	√						
Peeling ratio of island OC			Low	High	Medium	Low	None	None	None

When the water contact angle of substrate and the chemical resistance of Island OC are both at high level, DOE 6-8 studies the influence of different residual stresses of Top OC on peeling ratio of Island OC. The results show that no matter whether the residual stress of Top OC is high, medium or low, peeling of Island OC does not occur at all, which means the peeling ratios of DOE 6-8 are all the all the same of zero. Because increasing the chemical resistance of Island OC can significantly reduce the effect of solvent infiltration caused by Top OC, thus preventing the decrease of adhesion force of Island OC at the interface on substrate. Therefore, the insufficient curing ratio of Island OC results in its insufficient chemical resistance, which should be the most fundamental cause for the peeling of Island OC.

In summary, in order to solve the peeling problem of Island OC, increasing its own curing ratio to enhance its chemical resistance should be given priority, and then considering to reduce the water contact angle of substrate by surface treatment or directly using the substrate with lower water contact angle.

3.3 Mechanism for peeling of Island OC

3.3.1 Mechanism for improvement of peeling of Island OC by increasing its own chemical resistance.

As mentioned before, solvent is one of the primary components of the formula of Top OC, the content of which is about 70-85%. When Top OC is fabricated on top of Island OC and substrate following the general process, the solvent will directly contact with Island OC during the most of the process time, until it is nearly completely removed after oven curing. So if the chemical resistance of Island OC is medium level, the solvent from Top OC will easily infiltrate into Island OC through all the contact interface between them, as shown in **Figure 5a**. And when the solvent infiltrate inside Island OC and reaches the interface between Island OC and substrate, the interaction of molecules between Island OC and substrate at the interface will be gradually destroyed. It will be gradually changed into the interaction of molecules between Island OC and solvent, and the interaction of molecules between substrate and the solvent. Due to the weak interaction force between the solvent molecules, the adhesion force of Island OC at the interface of substrate will gradually decrease with solvent infiltration become more and more seriously. When the adhesion force is less than the residual stress of Top OC, peeling of Island OC occurs.

Therefore, in order to solve the peeling problem, it is necessary to enhance the ability to inhibit solvent infiltration of Island OC by increasing its chemical resistance. The chemical resistance of Island OC is mainly determined by the cross-linking density of itself. When the cross-linking density increases, the intermolecular force will be stronger, the molecular space will be smaller, and it will be more difficult for solvent molecules to infiltrate into the Island OC. Therefore, as shown in **Figure 5b**, when increasing the crosslinking density by enhancing the curing ratio of Island OC, parts of new covalent bond has been generated, and the intermolecular force is stronger by changing from the Van der Waals's force to covalent bond force, the molecular space is smaller. Finally, the chemical resistance is enhanced to inhibit the solvent infiltration successfully, then peeling ratio of Island OC is greatly improved.

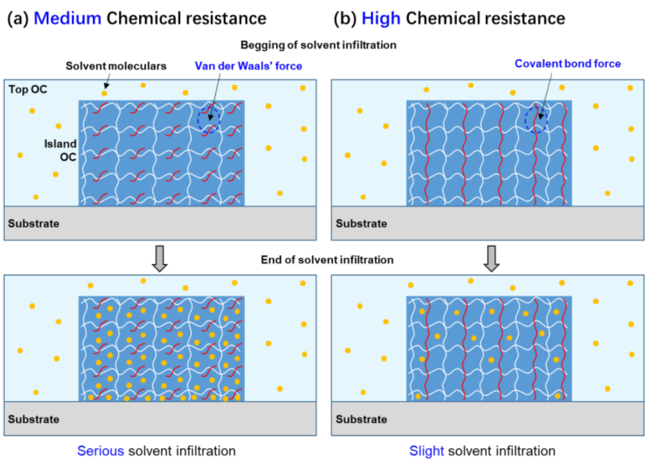

Figure 5. (a) medium chemical resistance of Island OC cause serious solvent infiltration; **(b)** high chemical resistance of Island OC cause slight solvent infiltration.

3.3.2 Mechanism for improvement of peeling of Island OC by reducing the water contact angle of substrate.

Reducing the water contact angle of substrate can improve peeling of Island OC mainly from two aspects. As shown in **Figure 6a**, on one hand, reducing the water contact angle of substrate can increase the adhesion force of Island OC at the interface of substrate. Because there will be more polar groups on the surface of substrate, which enhancing the polarity of it, meanwhile increasing the wettability of Island OC on the substrate, which further increasing the contact area between them, thus increasing the adhesion force. And high adhesion force of Island OC on the substrate will contribute to improvement of peeling. On the other hand, as shown in **Figure 6b**, reducing the water contact angle of substrate can inhibit the solvent infiltration at the interface between Island OC and substrate, which reduces the decrease of adhesion force and contribute to the improvement of peeling. Because when the adhesion force of Island OC at the interface of substrate is enhanced, the molecular space between Island OC and substrate at the interface will be smaller. So that it can inhibit the infiltration of solvent into the interface of Island OC and substrate, reducing the damage and change on the interaction of molecules between Island OC and substrate, which also effective in improving peeling. Due to the above two aspects, reducing the water drop angle of the substrate also plays an important role in decreasing the peeling ratio of Island OC.

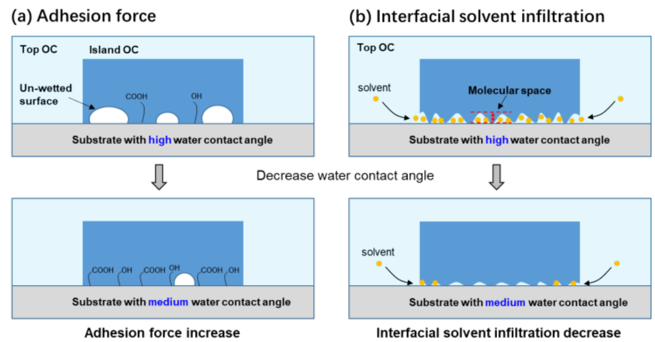

Figure 6. mechanism for decreasing water contact angle to increase adhesion force (a) and inhibiting interfacial solvent infiltration (b).

3.4 Research of residual stresses and chemical resistance of acrylic photoresist

3.4.1 Residual stresses of Top OC

Firstly, the impact of different process stations on the residual stress of Top OC are investigated. As shown in Figure 7, it is evident that three process stations like VCD, PB, and DEV caused almost no change in the residual stress of Top OC compared to the previous station. However, EXP, IUV, and OVEN stations significantly influenced the residual stress of Top OC, with the residual stress at these stations much greater than that at the preceding station. Notably, IUV and OVEN stations exhibit more pronounced changes in residual stress. Analysis reveals that VCD, PB, and DEV share a common characteristic: none contribute to improving the curing rate of Top OC. In contrast, EXP, IUV, and OVEN all enhance the curing rate. This indicates that the true determinant of Top OC's residual stress lies in its curing rate variation. Higher curing rate leads to Higher crosslinking density, smaller molecular chain spacing, and greater volume shrinkage, consequently resulting in higher residual stress of Top OC. [3,4] By combining different process conditions, Top OC films with high, medium, and low residual stress levels are ultimately obtained, as demonstrated in **Figure 7**.

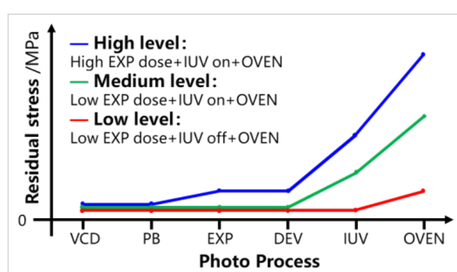

Figure 7. residual stresses of Top OC at different process stations and under different process conditions.

3.4.2 Chemical resistance of Island OC

The chemical resistance of materials refers to their ability to resist solvent-induced swelling. **Figure 8** shows the investigation of the relationship between the chemical resistance of Island OC and IUV treatment, which are verified by characterizing the content variation of N-methylformamide (a primary component of stripper) within Island OC films using TOF-SIMS. **Figure 8a** reveals that both the untreated sample of Island OC and the IUV-treated sample of Island OC+IUV on initially exhibited low N-methylformamide content. After 3-minute stripping treatment, both samples show significant increases in N-methylformamide content, indicating solvent- induced swelling. However, the IUV-treated sample demonstrated substantially lower N-methylformamide infiltration and faster recovery to baseline levels at only 180s (Island OC +IUV off sample needs 210s in comparison), suggesting both reduced solvent absorption and shallower swelling depth in Island OC+IUV on sample. These findings confirm that the IUV-treated sample possesses significantly enhanced chemical resistance compared to the untreated counterpart. The 3D maps in **Figures 8b-d** provide visual confirmation of these conclusions.

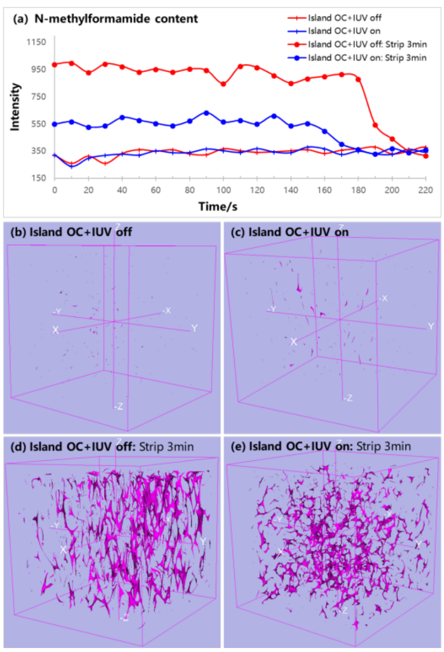

Figure 8. TOF-SIMS test results: (a) test time curve of N-methylformamide content in Island OC film under different process conditions; (b) & (c) 3D maps of N-methyl-formamide content in Island OC film before strip treatment; (d) & (e) 3D maps of N-methylformamide content in Island OC film after strip treatment.

Therefore, IUV treatment serves as an effective method for improving the chemical resistance of Island OC materials. The experimental results demonstrate that Island OC+IUV on sample exhibits high chemical resistance while untreated Island OC +IUV off sample maintains medium resistance levels.

4. Conclusion

This paper has mainly researched the peeling performance of acrylic photoresist with isolated island pattern in OLED display. **Firstly**, by force analysis of the peeling model of Island OC, it is confirmed that there are three main factors affecting peeling of Island OC, including the water contact angle of substrate, the residual stress of Top OC and the chemical resistance of Island OC. **Secondly**, exploring experiments (DOE 1-8) has been conducted and concluded that the primary cause for peeling of Island OC is the chemical property of itself is not sufficient enough, which resulting serious solvent infiltration by Top OC and greatly decrease the adhesion force. Increasing Island OC's chemical resistance to inhibit the solvent infiltration could completely improve peeling of Island OC with no peeling happens. Besides, by reducing the water contact angle of substrate or decreasing the residual stress of Top OC, the peeling ratio of Island OC could only be greatly decreased, but peeling of Island OC still happens, certificating that they should be not the primary cause of peeling. **Thirdly**, the mechanism for decreasing peeling of Island OC by reducing the water contact angle of substrate, and by increasing its own chemical resistance have been explained. **Additionally,** the residual stress of Top OC was investigated, and it was clarified that the curing rate is the fundamental factor affecting the residual stress of Top OC. Different residual stresses can be obtained by adjusting EXP, UV, and OVEN processes. **Finally**, the chemical resistance of Island OC was studied, confirming that IUV is one of the effective methods to improve the chemical resistance of Island OC.

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