

A Sacrificial Layer for Laser Lift-off of Colorless Polyimide

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ABSTRACT

We experimentally studied the a-Si deposited by PECVD as the sacrificial layer (SCL) for the laser lift-off (LLO) of colorless polyimide (cPI). The results shows that colorless polyimide (cPI) with SCL layer after LLO exhibits lower yellow intensity, higher tensile strength, no curling and slighter laser damage.

1 INTRODUCTION

In recent years, flexible transparent displays have been widely used in some applications, such as mobile phones and TV displays [1]. In order to achieve the flexibility and high transmittance of the displays, rigid glass substrate should be replaced by flexible one. Colorless polyimide (cPI) was the best candidate for the flexible transparent display because it shows excellent thermal stabilities with T_g about 400 °C and outstanding mechanical properties coupled with high optical transmittance beyond 90 % and high flexibility [2].

Laser lift-off (LLO) is an effective and simple way for the cPI flexible substrate delaminated from the rigid glass substrate [3]. However, some issues were still occurred at the LLO process, such as wrinkles and breaking points or holes on the cPI film. Although many papers reported the SCL layer for LLO of cPI, there is no discussion and systematic research in detail about the thickness and deposition temperature of a-Si sacrificial layer for the LLO process of cPI substrate [4].

In this paper, we systematically investigated the threshold fluence of hydrogen explosion for a-Si after laser annealing and selected two a-Si layers as the SCL layer to study and compare the effect of a-Si layer on the LLO process of cPI.

2 EXPERIMENT

2.1 A-Si layer deposition and laser annealing

A-Si layers of S1~S10 with theoretical thickness of 50~500 Å were firstly deposited on the glass substrate by PECVD on the basis of different deposition time varies from 5 to 50 s at the temperature of 420 °C. In order to obtain the optimum thickness of a-Si as SCL layer, we experimentally and systematically investigated the effect of laser fluence on the a-Si layer and the threshold fluence of hydrogen explosion of a-Si by laser annealing was confirmed by SEM and UV-Vis. The hydrogen explosion

phenomenon of a-Si layer was caused by the hydrogen jetting out from the a-Si layer irradiated by laser energy.

2.2 cPI coating and laser lift-off

cPI varnish was firstly coated on the above glass substrate deposited with a-Si layer and cured at 100 °C /1h and 400 °C/0.5h, respectively. The thickness of cPI film is about 10 μm and the glass substrate is 0.5 mm. To obtain the threshold fluence of LLO of cPI, cPI substrate with S1 and S2 a-Si SCL layer was scanned with increasing laser fluence. Then, the optical and mechanical properties of cPI after LLO process were compared and discussed. Meanwhile, the surface morphology and curling situation of cPI after laser annealing was also scanned. Finally, the thermal and solvent resistant properties of cPI substrate with SCL layer were also measured.

Table 1 The deposition time, thickness, Tr and threshold fluence of S1~S10 a-Si layers

Sample	S1	S2	S3	S4	S5
Deposition Time (s)	5	10	15	20	25
Theoretical Thickness(Å)	50	100	150	200	250
Real Thickness (Å)	126	145	175	191	256
Tr at 308nm (%)	11.5	4.9	3.0	1.7	0.9
Threshold Fluence (mJ/cm ²)	190	195	225	245	245
Sample	S6	S7	S8	S9	S10
Deposition Time (s)	30	35	40	45	50
Theoretical Thickness (Å)	300	350	400	450	500
Real Thickness (Å)	297	364	412	443	511
Tr at 308nm (%)	0.4	0.2	0.1	0.1	0.1
Threshold Fluence (mJ/cm ²)	245	245	245	265	285

3 RESULTS and DISCUSSION

3.1 The threshold fluence of hydrogen explosion of a-Si layer

In this work, we firstly prepared a-Si layers by PECVD on the glass substrate with theoretical thickness of 50~500 Å deposited at various time at the temperature of 420 °C, and the results were presented at Table 1. It was found that the real thickness of a-Si layer is higher than the theoretical thickness when the deposition time below 15 s. Meanwhile, the transmittance (Tr) at 308 nm of a-Si layer was decreased with the increasing of

deposition time or thickness.

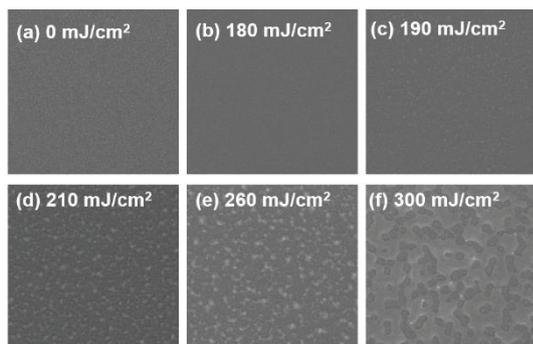


Fig.1 The surface morphology of S1 a-Si layer after laser annealing with different laser fluence.

The 308 nm laser from excimer laser irradiated the a-Si layer from the bottom of glass and the effect of laser fluence on the a-Si layers was studied. The threshold fluence of hydrogen explosion of a-Si layers by laser annealing shown in Table 1 was in the range from 190 to 285 mJ/cm². It can be concluded that the threshold fluence of hydrogen explosion of a-Si layers was increased with the increasing thickness of a-Si layer. Generally, a-Si layer with high threshold fluence of hydrogen explosion was unsuitable as SCL layer because high laser energy would irradiate the cPI if the a-Si layer cannot absorb the laser energy completely.

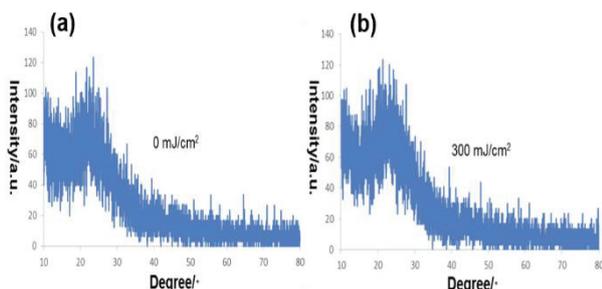


Fig. 2 The XRD curve of (a) S1 a-Si layer and (b) S1 a-Si layer after laser annealing with 300 mJ/cm².

We used the laser annealing process of S1 a-Si layer as an example to illustrate the phenomenon of hydrogen explosion of a-Si layer and the threshold fluence of a-Si was confirmed by SEM and UV-Vis. Figure 1 presents the surface morphology of S1 a-Si layer after laser annealing process with different laser fluence. It is obvious that the surface morphology of S1 a-Si layer starts to become rough at the laser fluence beyond 190 mJ/cm² and become more rough with the increasing of laser fluence. However, the surface morphology of S1 a-Si layer keep smooth after laser annealing when laser fluence less than 190 mJ/cm² because the laser fluence is too low to make hydrogen explosion. Therefore, the laser fluence of 190 mJ/cm² is considered as the threshold fluence of S1 a-Si layer.

Meanwhile, some small crystal nucleus occurred at the surface of S1 a-Si layer when laser fluence more than 260 mJ/cm², because a-Si layer may become LTPS after

dehydrogenation at high laser fluency. However, XRD curve shown in figure 2 appear no obvious crystallization peaks after the a-Si layer irradiated by laser with laser fluence of 300 mJ/cm² due to the degree of crystallinity is negligible.

The UV-Vis transparency spectra of the S1 a-Si layer after laser annealing with laser fluence ranging from 180 to 300 mJ/cm² were measured and illustrated in Figure 3. When the laser fluence below 190 mJ/cm², there is no obvious change of transmittance (Tr) compared with the a-Si layer with no laser annealing. However, with the increasing of laser energy density beyond 190 mJ/cm², a-Si layer after laser annealing exhibited higher Tr due to the decreasing of stacking density of a-Si layer caused by the hydrogen (H₂) jetting out from the a-Si. Therefore, the results of UV-vis also confirmed that 190 mJ/cm² is the threshold fluence of S1 a-Si layer.

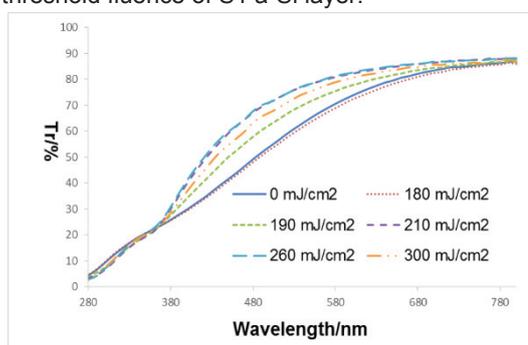


Fig. 3 The UV-Vis transparency spectra of S1 a-Si layer after laser annealing with different laser fluence.

3.2 The laser threshold fluence of LLO of cPI

According to the results of threshold fluence of hydrogen explosion and Tr at 308nm of a-Si layer, S1 and S2 a-Si layers were selected as candidates of SCL layer for the LLO of cPI. Firstly, H230 PI varnish from MGC company was coated on S1, S2 a-Si layer substrate and pure AN100 glass substrate from AGC company without a-Si layer, respectively. Then, the above three substrate was cured at 100°C/1h and 400°C /0.5h to remove the solvents sufficiently. The LLO process of the above three substrates, S1+cPI, S2+cPI and glass+cPI were studied. The laser threshold fluence of LLO process of S1+cPI, S2+cPI and glass+cPI presented in Table 2 were 200, 240, 220 mJ/cm², respectively. Therefore, the introduction of S1 a-Si layer can reduce the laser fluence of LLO process of cPI, which is beneficial from the delamination of cPI from glass substrate. It was also concluded that the laser threshold fluence of cPI+a-Si substrate is slightly higher than that of a-Si layer substrate because it needs produce more H₂ at a higher laser fluence to make cPI more easily delaminated from the glass substrate.

3.3 The protection of SCL layer for cPI after LLO

The optical and mechanical properties shown in Table

2 as well as damage situation of cPI after laser annealing at the threshold fluence were compared to estimate the effect of SCL layer on the delamination of cPI from glass substrate. Compared with the cPI-0, cPI-1 and cPI-2 exhibited lower color intensity, especially cPI-2 with Yellow index (YI) and b value of 4.1 and 2.5 respectively. Meanwhile, cPI-1 had the highest tensile strength of 125 mPa.

Table 2 The threshold fluence, optical and mechanical properties and curling situation of cPI samples

Sample	cPI-0	cPI-1	cPI-2
Sacrificial layer	No	With S1	With S2
Threshold fluence (mJ/cm ²)	220	200	240
Yellow Index	5.1	4.3	4.1
b value	3	2.7	2.5
Tensile strength (mPa)	115	125	121
Elongation at Break (%)	5.1	6.4	5.5
Curling Situation	Serious Curling	No Curling	No Curling

The surface morphology shown in figure 4 of cPI-0, cPI-1 and cPI-2 after LLO process is also measured by SEM. It was found that the surface of cPI-0 is seriously burned by the laser under the high energy density. However, there are no obvious damage at the surface of cPI-1 because a-Si layer can absorb most of laser energy. Meanwhile, the surface of the cPI-2 occurred some laser damage due to the high laser fluence. Therefore, laser beam with high laser fluence can irradiate the cPI even applying a-Si as SCL layer. Meanwhile, we also found that there is no a-Si remaining at the surface of cPI by Energy Dispersive Spectrometer (EDS) analysis.

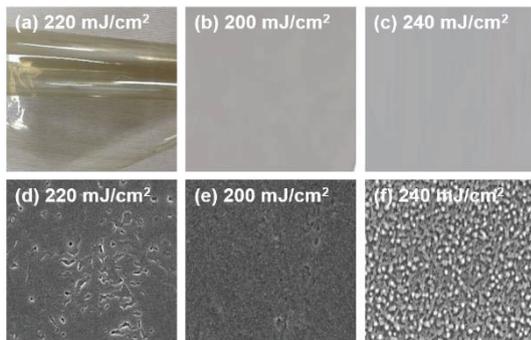


Fig. 4 The degree of curling of (a) cPI-0, (b) cPI-1, (c) cPI-2 and the laser damage of (a) cPI-0, (b) cPI-1, (c) cPI-2 after LLO.

3.4 The thermal and solvent resistant properties of cPI substrate with SCL layer

In order to estimate the thermal stability of cPI substrate with a-Si SCL layer, the thermal annealing process under high temperature of 380 °C/1h at N₂ atmosphere was carried out on the above three samples. The adhesion performance before and after thermal annealing was measured and compared by Paint Adhesion Testing, it was found that the cPI substrate with SCL layer showed

no adhesion loss and exhibited excellent adhesion performance of 5B.

Solvent resistance of cPI substrate with S1 SCL layer is a key point for the Color Filter and TFT process in the display. We experimentally investigate the solvent resistance of cPI substrate for the Cu and Al TFT process, respectively. The cPI substrate was saturated sequentially with KOH developer, Al etching liquid and Al stripper liquid for 8h, respectively. Meanwhile, the cPI substrate was soaked orderly in KOH developer, Cu etching liquid and Cu stripper liquid for 8h, respectively. The results showed that the surface of cPI keep smooth and the adhesion between cPI and a-Si layer keep stable with adhesion of 5B after solvent resistance test for Cu and Al TFT process, respectively.

4 CONCLUSIONS

A series of a-Si layers were deposited and the threshold fluence of hydrogen explosion for a-Si by laser annealing was confirmed. S1 and S2 a-Si layers showed threshold fluence less than 200 mJ/cm². Meanwhile, S1 and S2 a-Si layers were used as SCL layer for the LLO process of cPI. The laser threshold fluence of LLO process of cPI with S1 a-Si SCL layer below to 200 mJ/cm². The results indicated that the cPI after LLO with a-Si SCL layer exhibited higher optical transmittance and better mechanical properties compared with the cPI without SCL layer. Therefore, the experimental study of a-Si SCL layer for the LLO of cPI in this work can provide theoretical guide on the design of SCL layer for the LLO process of flexible cPI substrate.

REFERENCES

- [1] Kaijun Wang et al, "Flexible LCD Panel with Colorless Polyimide," IDW, PP 284-286 (2019).
- [2] Yu Shi et al, "14 Inch Flexible LCD Panel with Colorless Polyimide," SID Digest, PP 597-599 (2019).
- [3] Jing Bian et al, "Theoretical and experimental studies of laser lift-off of nonwrinkled ultrathin polyimide film for flexible electronics," Applied surface science, 499 (2020) 143910, DOI:10.1016/j.apsusc.2019.143910.
- [4] Chan Il Park et al, "World 1st Large Size 77-inch Transparent Flexible OLED Display," SID Digest, PP 710-713 (2018).