Development of Adhesive-Free Lamination of Liquid Crystal Polymer Using a Plasma Surface Treatment at Atmospheric Pressure

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A liquid crystal polymer (LCP) film were adhered by plasma treatment at atmospheric pressure and thermocompression without using an adhesive. The adhesive strength of plasma treated film increased drastically and plasma treated film indicated cohesive failure inside the film. In addition, the treatment time required to cause cohesive failure was shortened by increasing the discharge power and shortening the distance between the plasma outlet and the sample surface. According to the valence band XPS spectra, it is assumed that the melting point of film surface decreased. The adhesive strength of Ar/He plasma treated film increased and the treatment time required to cause cohesive failure was shortened by containing Ar.

Keywords: Atmospheric pressure glow plasma, Liquid crystal polymer, Adhesive-free lamination

1. Introduction

A liquid crystal polymer (LCP) film is a thermoplastic resin which is excellent in various physical properties such as heat resistance, gas barrier property, moisture proofness. And the LCP films are considered as applications such as base material of an electronic circuit and airplane parts. Since the molecular structure of LCP shown in Fig. 1 are quite liner, the polymer chains are not entangled with each other and the mechanical strength in the direction perpendicular to the main chain is weak. Thus, it was expected that a high mechanical strength material was obtained by thermocompression after overlaying a plurality of LCP films in such a way that the molecular chains oriented in a grid pattern. However, the adhesive strength between the films was not improved only by thermocompression.

In this study, we tried to develop an adhesive-free lamination technique using the atmospheric pressure glow (APG) plasma surface treatment and thermocompression [1-3].

2. Experimental

The plasma treatment system is shown in Fig. 2.

The discharge was generated after putting the LCP film (5 cm × 12 cm × 20 µm) on the lower electrode, and plasma treatment was performed by sliding the lower electrode and passing it in the discharge field. The treatment time was changed by adjusting the passing speed of the slider. The Discharge conditions are shown in Table 1.
Table 1. Discharge conditions.

| Parameter                        | Value       |
|----------------------------------|-------------|
| Frequency / MHz                  | 13.56       |
| He flow rate / L min\(^{-1}\)    | 0.5, 2      |
| Ar flow rate / L min\(^{-1}\)    | 1.5         |
| Discharge power / W              | 50, 100, 150|
| Distance from the plasma outlet to the sample stage / mm | 1          |
| Treatment time / s               | 0.01~2      |

The plasma treated film was cut in half, and treated surfaces were thermocompression bonded each other under the condition of Table 2. The adhesive strength was evaluated by the T-type peeling test. The surface analysis of the LCP film was carried out with XPS (PHI 5000 Versa Probe II, ULVAC-Phi, take-off angle 15°).

Table 2. Thermocompression conditions.

| Parameter       | Value |
|-----------------|-------|
| Temperature / ℃ | 275   |
| Load / t        | 1     |
| Bonding time / min | 5     |

3. Results and discussion

3.1. He Plasma Treatment

Figure 3 shows the relationship between the treatment time and the adhesive strength at each discharge power. The adhesive strength increased drastically and became a constant value by treatment above a certain time. The untreated films did not adhere to each other, whereas the plasma treated films indicated cohesive failure inside the film as shown in Photo 1. Since the plasma treated film reached the mechanical strength of the film itself, the adhesive strength showed the constant value. Increasing the discharge power extremely shorten the treatment time required to obtain sufficient adhesive strength.

![Photo 1. State of cohesive failure.](image)

Figure 4 shows the adhesive strength of the film treated at a discharge power of 50 W at distances of 1 mm and 0.5 mm from the plasma outlet to the sample surface. The adhesive strength at distance of 0.5 mm became constant in a shorter treatment time than that of 1 mm. It is considered that the power density per unit volume of the discharge field is increased by shortening the distance.

![Fig. 4. Adhesive strength at each distance from the plasma outlet to the sample stage.](image)

![Fig. 3. The adhesive strength at each discharge power.](image)

The valence band spectra of the untreated and plasma treated film surfaces were measured by XPS. The XPS spectra are shown in Fig. 5. The peaks were made smaller by plasma treatment. These results indicate that molecular structure of the film surface was broken by the plasma treatment. It is assumed that the surface molecular of film became smaller and the melting point of film surface decreased, then the films were adhered.
3.2. Ar/He plasma treatment

Ar Plasma treatment investigated since Ar is industrially more advantageous than He. The plasma treatment system was impossible to cause normal discharge only with Ar. Thus, using Ar/He mixture gas and setting the discharge conditions shown in Table 3 caused normal discharge in the state of containing the largest amount of Ar. Figure 6 shows the adhesive strengths of Ar/He plasma treated and He plasma treated films using the same discharge condition. The adhesive strength between films could be greatly improved even with Ar/He plasma treatment. In addition, it was possible to shorten the treatment time required to cause cohesive failure by including Ar. Since the atomic weight of Ar is considerably larger, Ar is harder to diffuse than He and it was easier for Ar to reach the film surface in discharging atmosphere than He.

| Frequency / MHz | 13.56 |
|-----------------|-------|
| He flow rate / L | 0.5   |
| Ar flow rate / L | 1.5   |
| Discharge power / W | 50    |
| Distance from the plasma outlet to the sample stage / mm | 0.5 |

4. Conclusion

It was possible to increase the adhesive strength up to the mechanical strength of film itself by the combination of atmospheric pressure glow plasma surface treatment and thermocompression. The treatment time can be shortened by increasing the discharge density. According to the valence band XPS spectra, it is assumed that the melting point of film surface decreased. Ar/He plasma treatment obtained also enough high adhesive strength.

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References

1. S. Kanazawa, M. Kogoma, T. Moriwaki, and S. Okazaki, *J. Phys. D*, 21 (1988) 838.
2. T. Yokoyama, M. Kogoma, T. Moriwaki, and S. Okazaki, *J. Phys. D*, 23 (1990) 1125.
3. T. Yokoyama, M. Kogoma, S. Kanazawa, T. Moriwaki, and S. Okazaki, *J. Phys. D*, 23 (1990) 374.