Analysis of adhesion forces in ink-transfer process

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Roll-to-roll printing process has been noticed recently because it is one of the most economical ways to produce various types of printed electronics such as RFID tag and OLED, or fuel cells. It has advantages in rapid process, mass production ability, low cost both in equipment and production, while conventional lithographic process requires long manufacturing time and expensive equipments, and considerable amount of wastes from etching procedure using harsh chemicals [1]. Figure 1 shows the principle of gravure printing process, one of the processes using roll-to-roll methods. Grooves are engraved on the surface of the gravure roll, which is partially immersed in ink pool first. As the gravure roll rotates, it picks up ink and the excess ink is wiped off from the surface by a doctor blade. The ink in the cell is transferred to the substrate and goes to the drying zone.

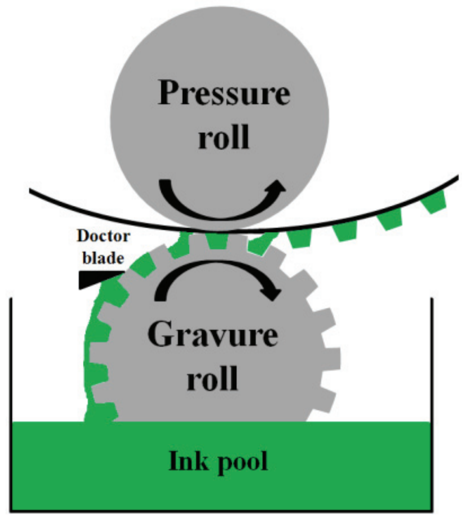

Figure 1. Schematic of gravure printing process.

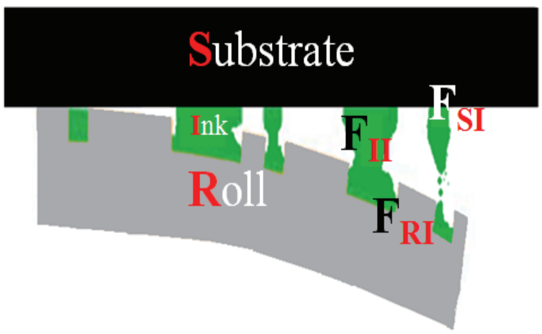

Figure 2. Insufficient ink transfer in gravure printing Process. F_{SI} is interfacial force between ink and substrate, F_{RI} is interfacial force between ink and roll, and F_{II} is bulk force of ink.

As the demand for the patterns with high resolution increases, insufficient ink transfer from the roll to the substrate become as one of the most critical issues of the system because it could induce poor print quality as well as non-uniform coating thickness. To improve the amount of ink transfer, we need to exactly understand the mechanism of the process. Ink transfer process is affected by many factors such as operating conditions, cell geometries, ink properties, and interaction between ink and roll or substrate [2]. Even though there have been

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previous studies about the transfer mechanism affected by operating conditions or by cell shape and dimensions [2-4], mechanism of the ink transfer process is not fully understood yet.

In this study, therefore, we aim to understand the ink transfer process mechanism better by analyzing forces involved in the ink transfer process. There are three forces involved in the ink transfer process as illustrated in Figure 2. The first is the bulk force F_{II} of the ink which is closely related to the ink's shear modulus. The other forces are the interfacial forces F_{IS} and F_{IR} which are related to the interaction between the ink and the substrate and the interaction between the ink and the roll, respectively.

Here, each different force was evaluated using separate methods. Total adhesion force, which is consisted of the interfacial forces and bulk force, was evaluated using universal testing machine (UTM) (Figure 3). Initial gap between two parallel plates was 1 mm and the ink is loaded between them. As the upper plate moves up vertically at 1 mm/s, the liquid is separated into two parts attached to each plate. To change interfacial force, two liquids and two plate materials were used. For the liquids, usual ink system was simplified to binder solution, for binder material plays the biggest role in attaching to the substrate. Two polymer solutions that are widely used as the binder for commercial pastes were chosen. Ethyl cellulose (EC, MW=15-20x10^4 g/mol) and polyvinyl butyral (PVB, MW=30-35x10^4 g/mol) were dissolved in dihydroterpineol acetate (DHTA) respectively at 353 K for 4 h. They will be denoted as EC solution and PVB solution through this manuscript. For the plate material, PI film and stainless steel were used.

Figure 4 shows an example of measured stress as a function of strain using EC solution and stainless steel plates. The shape of the liquid during the stretching was also observed. As the upper plate moved upward, the binder solution was stretched and the filament was created in the middle of the solution. The contact diameters of the binder solution on the upper and lower plates were decreased simultaneously due to the surface tension. Maximum stress was measured as soon as the upper plate moved up to the final gap at 1.1-1.3 mm. After the maximum peak, binder solution started flowing. The binder solution seemed to withstand the extension strain at the maximum peak. Therefore, modulus of the binder solutions is dominant parameter at the maximum stress. After the liquid filament was broken and separated, it was recoiled toward the plates.

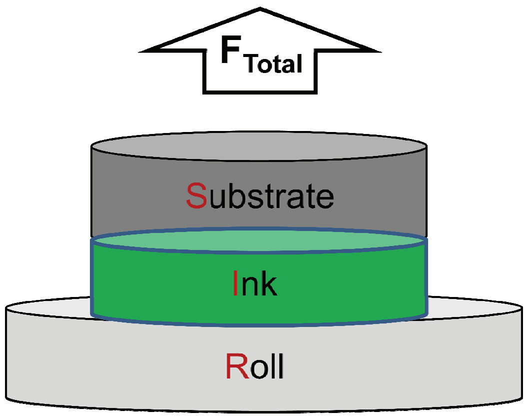

Figure 3. Analogy of the UTM test for characterizing the total adhesion force of binder solutions.

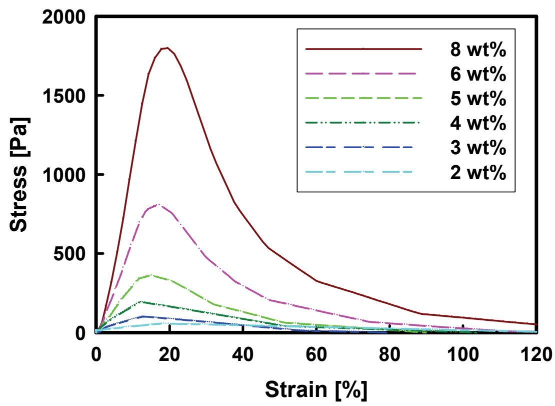

Figure 4. Total adhesion force of EC solutions with different concentrations.

To characterize the bulk force, the storage modulus (G') of the binder solutions with various concentrations was measured using a rotational rheometer (ARES, TA instrument). Frequency sweep test was performed at 10% strain in frequency range of 1 rad/s to 100 rad/s. The bulk force was increased with the increased concentration for both binder solutions. G' of PVB solution was larger than that of EC solution at the same concentration, which implies that bulk force of PVB solution is higher than that of EC solution.

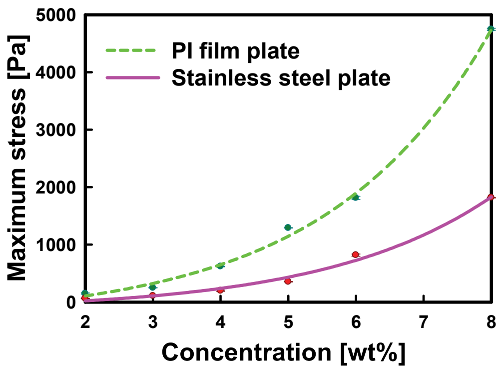

Figure 5. The maximum stress of EC solution as a of concentration with different plates.

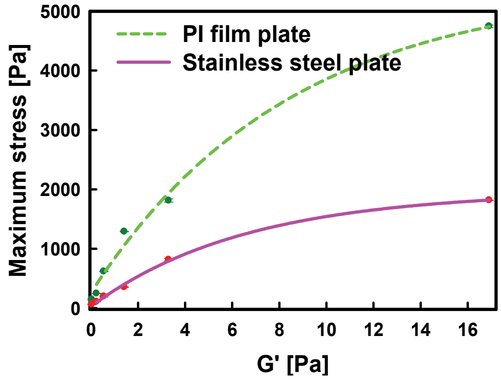

Figure 6. Maximum stress of EC solution as a function of function *G'* with different plates

Then the interfacial force was evaluated by analyzing the bulk force and total adhesion force. First experimental set was using two different plate materials and the EC solution. The maximum stress was plotted against EC concentration in Figure 5. The maximum stress was increased exponentially with increasing EC concentration. The maximum stress at the moment of extension was larger when PI film was used for the plate than when stainless steel was used at the same concentration of EC. Now, when the maximum stress is plotted against *G'* in Figure 6, the maximum stress was higher at PI film plates at whole *G'* range. The difference in the maximum stress for PI film plate and steel plate at the same *G'* can be considered as the difference of the interfacial force for PI film plate and steel plate with the same liquid. The interfacial force of when PI film plate was used is higher than that of when stainless steel plate was used.

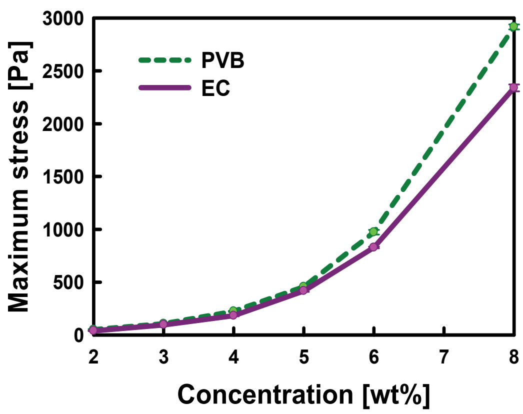

Figure 7. The maximum stress of EC and PVB solution as a function of concentration with stainless steel plate.

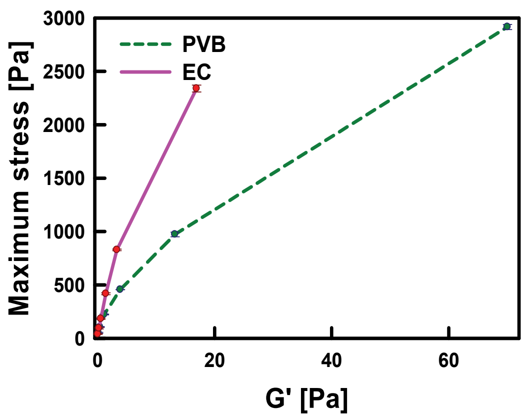

Figure 8. Maximum stress of EC solution and PVB solution as a function of *G'* at stainless steel.

Second experimental set was using the steel plates and two different binder solutions. Maximum stress of PVB solution was larger than that of EC solution at the same concentration as shown in Figure 7. The maximum stress of EC solution was also higher than that of PVB solution at whole *G'* range as shown in Figure 8. The difference in the maximum stress for EC solution and PVB solution at the same *G'* value can be considered as the difference of the interfacial force for EC solution and PVB solution with plates, which means the interfacial force of EC solution is higher than that of PVB solution.

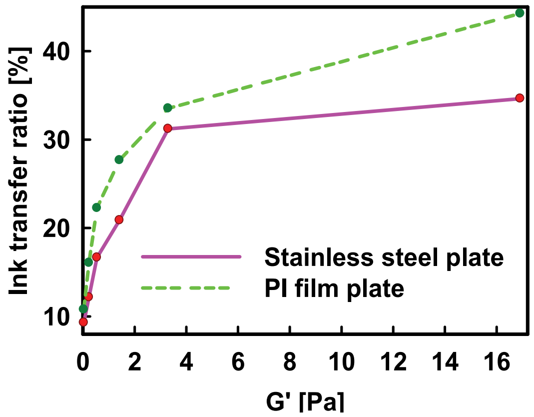

Figure 9. Ink transfer ratio of EC solution as a function of *G'* with different plates.

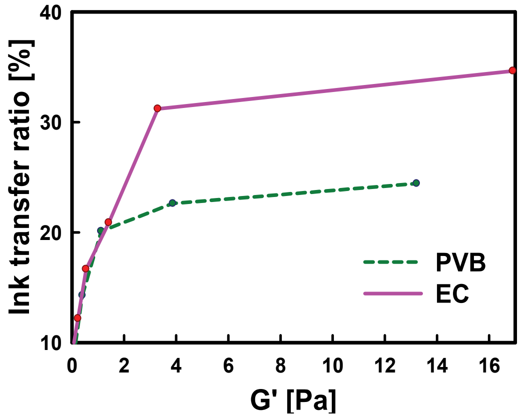

Figure 10. Ink transfer ratio EC solution and PVB solution as a function of *G'* at stainless steel.

The effect of the interfacial force between ink and substrate must affect the ink transfer ratio, which is defined as the weight ratio between the ink that was originally in reservoir before the transfer and the ink that is transferred to the substrate. Figure 9 and Figure 10 show the results of ink transfer ratio as a function of *G'* of the binder solution. In the earlier part of this study, the interfacial force between PI film plate and EC solution was found to be higher than that between stainless steel plate and EC solution. Now it influenced ink transfer ratio. The ratio using PI film plate was higher than the ratio using stainless steel plate at first experimental set. Second experimental set also showed same tendency. EC solution and stainless steel plate had higher interfacial force than PVB solution and the same plate, and therefore ink transfer ratio was higher as well.

In this study, we systemically varied the interfacial force between the substrate and the ink in gravure printing and correlated it with the ink transfer ratio. The interfacial force was indirectly determined by the difference of total adhesion force measured by UTM test and the bulk force determined from rheological measurements. The ink transfer ratio was improved when the interfacial force was higher by either different binder system or by different substrate material. Not only correlating interfacial forces to the quality of gravure printing, we also proposed a simple methodology to characterize the ink transfer process.

Acknowledgment

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