

Quality control with matching technology in roll to roll printed electronics[†]Changwoo Lee¹, Hyunkyoo Kang², Hojoon Kim², Ho Anh Duc Nguyen² and Keehyun Shin^{2,*}¹Flexible Display Roll to roll Research Center, Konkuk University, Seoul, 143-701, Korea²Department of Mechanical Engineering, Konkuk University, Seoul, 143-701, Korea

(Manuscript Received May 1, 2009; Revised September 30, 2009; Accepted October 16, 2009)

Abstract

R2R (Roll to roll) printed electronics has been an attractive technology for the mass production. Therefore, many research works have been focused on an optimal flexible substrate, ink formulation, printing process, curing method for a conductive ink in printed electronics applications. However, they did not relate their analysis with the system parameters of R2R continuous printing systems. In this paper, it is found that a printed pattern geometric quality which affects functional quality of printed electronic device could be changed with respect to operating tension of bare substrate even if local optimized ink, substrate, and printing process were applied. Additionally, ink transfer mechanism for R2R printed electronics is analyzed regarding a dynamic surface energy of a bare substrate under a tension in R2R printing systems. With the aim of an efficient prediction of the thickness of R2R printed patterns for given operating conditions, a simple meta-model is developed by using the design of experiment (DOE) method. Also, the proposed meta-model has been verified by several experiments. Through the results, it is presented that how to find an optimal operating tension in R2R printed electronics for guaranteeing a required thickness of R2R printed patterns.

Keywords: At least four keywords; Ink transfer mechanism; Matching correlation; Roll to roll (R2R) printed electronics; Tension; Thickness

1. Introduction

Printed electronics is a revolutionary technology aimed at flexible electronic device manufacturing on plastic foil. To have attractive mass production in the industrial fields, roll to roll printing systems are strongly recommended. The process of printed electronic devices on a flexible substrate in a roll to roll system includes the integrated processes such as moving plastic foil with micro level registration control, transferring ink from a specific pattern to the moving substrate, maintaining specified thickness, width, surface roughness of the printed pattern on the substrate. Quality and productivity of printed electronic devices depend simultaneously on many mechanical factors, such as tension, register, nip pressure to chemical one, including ink viscosity, ink pigment and other aspects. A small difference in properties (viscosity, surface tension, solid contents, boiling point, etc.) of an ink [1] could produce major variations in print quality. Recently, more interest has been stimulated by experimental results that wettability could be tuned by surface geometry of a substrate [2]. Gravure based printing as well as ink-jetting based method was applied for electronic devices in printed electronics [3].

Lately, printed transistor was fabricated by spin coating as well as gravure, flexographic and inkjet printing method [4]. From reviewing the literature, it is clear that critical factors for making high quality of electronics applications are ink (viscosity, surface tension, solid contents, particle size, adhesiveness, etc.), substrate (surface energy, surface roughness, thermal deformation, adhesiveness, etc.) and printing parameters (printing type, nip-force, doctoring, contact angle, etc.). Previous studies on printed electronics were focused on the characteristics of each component in a batch printing process. However, they did not relate their analysis with the system parameters of roll to roll continuous printed electronic systems. In this paper, ink transfer mechanism is analyzed on the basis of dynamic surface energy due to tension variation in R2R printing systems. It is confirmed that the tension could affect thickness of a printed pattern in R2R printed electronics, even though other parameters (speed, viscosity, printing method, etc.) are constant. Also, the effects of major system parameters on R2R printed pattern are analyzed by using full factorial DOE (design of experiment). Simple meta-model for thickness of the R2R printed pattern is developed on the basis of the matching correlation among chemical properties, mechanical characteristics, and R2R system parameters. Finally, it is verified by experimental study that the proposed meta-model is effective for predicting the thickness of a R2R printed pattern for given operating conditions.

[†] This paper was presented at the ICMDT 2009, Jeju, Korea, June 2009. This paper was recommended for publication in revised form by Guest Editors Sung-Lim Ko, Keiichi Watanuki.

*Corresponding author. Tel.: +82 2 450 3072, Fax.: +82 2 447 5886

E-mail address: kshshin@konkuk.ac.kr

© KSME & Springer 2010

2. Correlation between the tension and the quality of the R2R printed pattern

In general, the geometric and functional qualities of a printed device are affected by many factors [1–4] such as 1) chemical characteristics: viscosity, surface tension, adhesiveness, 2) mechanical issues: cell depth, cell stylus, dot size of a cell, doctoring, nip force, etc., 3) properties of a substrate: surface roughness, thermal deformation, adhesiveness, and so on.

The electrical resistivity of a printed pattern was given by [5]

$$\rho = R \frac{A}{l} \quad (1)$$

where, R is the electrical resistance of a uniform specimen of the material, l is the length of the printed pattern, and A is the cross-sectional area of the printed pattern. From Eq. (1), it is clear that the electrical resistance of a material itself as well as geometric specification of a printed pattern is important factors. Increasing the thickness of a printed pattern causes a decrease in the resultant resistance [6]. This means that functional qualities of a printed electronic device are affected by geometric quality such as thickness of a printed layer even though characteristic of material itself is not changed. Walker and Fetsko proposed the following empirical ink-transfer model [7].

$$Y = A[bB + f(X - bB)] \quad (2)$$

where, A is the coverage function, X is initial amount of ink on the printing plate, B is the immobilization function, f is a splitting parameter, and b is a parameter depending on surface roughness and absorptivity of a bare substrate. In Eq. (2), surface roughness of a bare substrate was one of the most important parameters affecting ink transfer. In other early works [8], it was reported that substrate surface which was related to the contact angle between droplet and substrate could affect ink transfer. In the case of a solid substrate such as glass, a static surface roughness was used. Also, a static roughness could be applied in a batch process even though the substrate was flexible. However, the dynamic roughness of a flexible substrate should be considered in R2R printed electronics because a flexible substrate was transported continuously under an operating tension. Therefore, surface roughness of the substrate could be changed according to operating tension in R2R printed electronics system. The interferometer images support the contention that a flexible substrate has different surface roughness according to the tension as shown in Fig. 1. The measurements of surface roughness were carried out at twelve different locations in the heat stabilized substrate having 300mm (width), 300mm (length), and 0.1mm (thickness). The surface roughnesses of bare substrate under 5N/mm², 20N/mm² are 8.16nm, 6.18nm, respectively. The contact angles between reagents and substrate were measured by droplet tests as shown in Fig.2. The surface energy of a bare substrate was calculated by using the well-known Young's equation, Dupre's equation, and Lewis acid base method [9].

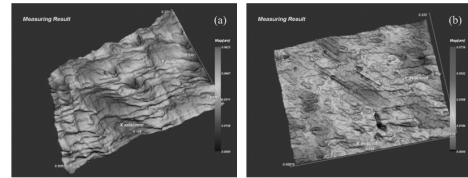


Fig. 1. Surface roughness images of a heat stabilized film by interferometer (scan area: 234um x 232um). (a) tensile force of 2N/mm², (b) tensile force of 7N/mm².

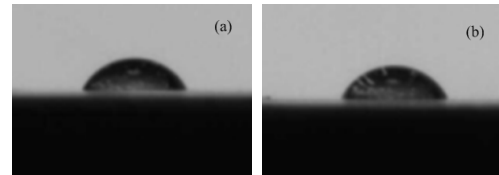


Fig. 2. Droplet test result by using reagent (Glycerol). (a) contact angle of 73.78° under 2N/mm², (b) contact angle of 75.61° under 7N/mm².

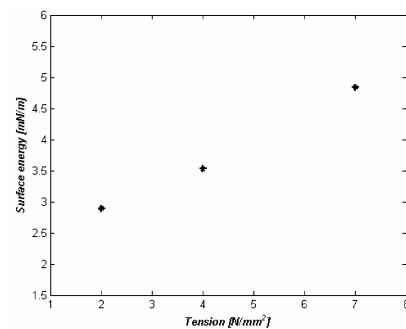


Fig. 3. Surface energy variation of a bare substrate with respect to the tensile force.

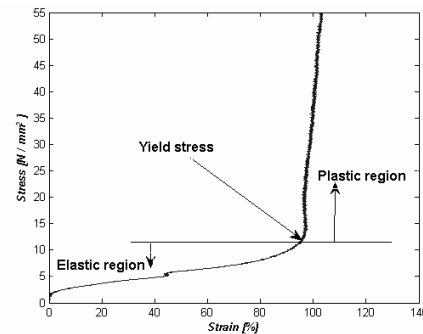


Fig. 4. Stress – strain curve of a heat stabilized film in 30 centigrade.

When a surface energy of a bare substrate is high, thickness of a printed pattern could be decreased, since the force of a bare substrate which spreads the ink on substrate is stronger than surface tension of an ink itself. The surface roughness of bare substrate was significantly varied by the tension. Therefore, the surface energy of bare substrate which could affect ink transfer was changed according to tension as shown in Fig. 3. Note that a tension of 10~50% of yield strength of the flexible material was recommended as an operating tension as shown in Fig. 4 [10]. The range of recommended operating tension of the used substrate was from 1.3 to 7N/mm². Finally,

not only a static surface roughness but also a dynamic surface roughness of bare substrate should be used to control the thickness of the printed pattern in R2R printing systems. However, it is generally hard to detect a dynamic surface roughness of a moving substrate directly. Therefore, a matching correlation between tension and geometric quality of R2R printed pattern is investigated by using DOE method.

3. Experimental verification

R2R printing system of Fig. 5 was used for experimental studies. The heat stabilized flexible substrates which have 300mm (width) and 0.1mm (thickness) was used to minimize thermal deformation due to high temperature. The preheating process was used for eliminating humidity on bare substrate. The patterns were printed with Ag nano-particle conductive ink by using the direct gravure printing process. The DOE approach allows for the concurrent testing of multiple factors. Therefore, the DOE approach provides an efficient method for optimizing the important design elements with regards to a cost problem [11]. In this study, the influences of three major factors on thickness of a printed pattern are investigated. The first stage in DOE was to identify major factors that were considered to have an impact on the geometric quality as follows: tension of substrate, speed of R2R printing systems, and viscosity of conductive ink. The units and settings of high and low levels of major factors are tabulated in Table 1. The response to characterizing the geometric qualities, namely thickness of a R2R printed pattern, is measured by interferometer. The eight tests were designed to observe the changes in the response to test factor level changes. The design is a two level experiment represented by the ‘±’ symbols. The tension level was selected within the traditional recommended operating range as shown in Fig. 4. Also, the speed and viscosity level were chosen considering sinter time of conductive ink, solid contents and printing method. Based on the experimental results, the main effects of each of three factors and their interactions on geometric quality were demonstrated through the normal Pareto charts for response (thickness) as shown in Fig. 6. It can be seen that the main effects on geometric quality appear to be influenced by input factor itself as well as interaction of input factors. Fig. 7 indicates that the thickness of R2R printed pattern was decreased when the tension was increased for a given operating speed (less than 4m/min).

The surface roughness of bare substrate was increased due to high tension under low speed. However, the effect of operating tension on thickness is too small in the condition of high speed (> 4.3 m/min) in Fig. 7. The interaction of tension and speed affects the output more than speed itself without interaction, since speed has negative sensitive, that is, in the low velocity condition, the tension is the most sensitive factor on the output thickness. Therefore, a matching correlation could be different according to operating values. From the experiment results, the printed pattern thickness variation was more than 13% with respect to the operating tension (from 2 to

Table 1. Defined three factors for screening.

Name	Unit	Settings
Tension (x_1)	Kgf	2.0 (-) to 4.5 (+)
Speed (x_2)	m/min	3.0 (-) to 5 (+)
Viscosity (x_3)	mPa·s	100 (-) to 150 (+)



Fig. 5. R2R printing systems. (a)unwinding, (b)lateral controller, (c)infeeding, (d)preheating, (e)gravure printing, (f)drying unit, (g)cooling, (h)outfeeding, (i)rewinding

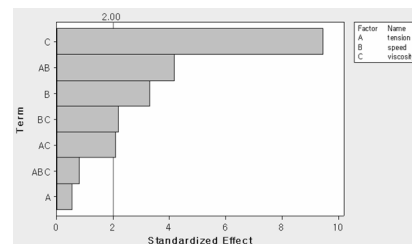


Fig. 6. The standardized effects on thickness of a R2R printed pattern.

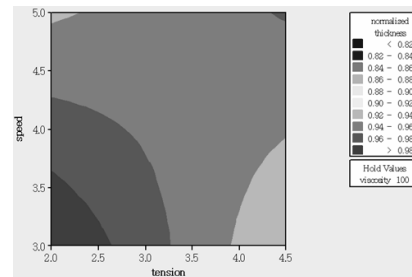


Fig. 7. Contour plot of normalized thickness via tension and speed: hold value- viscosity of 100mPa·s.

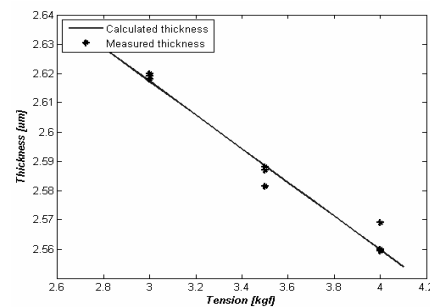


Fig. 8. Measured and calculated thickness of R2R printed pattern: hold value- viscosity of 100 mPa·s and speed of 3.5m/min.

4.5kgf). Simple meta model is developed from estimating parameter of the standard polynomial formulation by least squares fitting method as shown in Eq. (3), where, Th is the

thickness of R2R printed pattern on the substrate. In Eq. (3), x_1 , x_2 , and x_3 are tension [kgf], operating speed [m/min], and viscosity of ink [mPa·s], respectively. The validations of the proposed meta-model was performed by comparing measured geometric qualities and calculated values of R2R printed pattern for arbitrary inputs within boundary conditions. In Fig. 8, each of measured points is the average thickness of the five samples. The maximum error for given operating speed between measured thickness by experiments and calculated thickness by Eq. (3) was less than 0.38% as shown in Fig. 8. Therefore, the proposed meta-model could be used for estimating thickness of R2R printed pattern within the specified operating conditions of Table. 1. Through the results, it is found that the geometric qualities (thickness) which affects the functional qualities (conductivity, mobility, etc.) of a printed device could be changed according to the applied operating tension even though constant viscosity of ink, same materials, speed, and printing process were applied in R2R printed electronics.

$$Th = 4.28862 - 0.489x_1 - 0.189875x_2 - 0.0080725x_3 + 0.0915x_1x_2 + 0.002355x_1x_3 - 0.0000775x_2x_3 \quad (3)$$

4. Conclusion

The most important conclusion of the results presented in this research is that operating parameters of R2R printing system have been shown to have a significant effect on geometric quality of an R2R printed pattern.

The following conclusions can be drawn from this study;

The correlation between the tension of a bare substrate and the dynamic surface energy which affects the ink transfer has been found.

On the basis of matching correlation between the tension in R2R printing systems and the thickness of R2R printed pattern, a meta model has been developed by using DOE and the developed model has been experimentally verified.

Through the developed model, the thickness of R2R printed pattern could be controlled by the tension of a moving substrate in R2R printing system.

Acknowledgment

This research is supported by “Seoul R&BD Program (10848), Korea” and “the Korea Foundation for International Cooperation of Science & Technology (KICOS) through a grant provided by the Korean Ministry of Education, Science & Technology (MEST) in K20701040600-09A0404-05410”.

References

- [1] F. Shepherd, *Modern Coating Technology Systems*, Emap Maclaren Ltd., Barnet, U.K, (1995).
- [2] T. Onda, S. Shibuichi, N. Satoh and K. Tsujii, Superhydrophobic Fractal Surface, *Langmuir*, 12 (1996) 2125-2127.
- [3] M. Pudas, J. Hagberg and S. Leppavuori, The absorption ink transfer mechanism of gravure offset printing for electronic circuitry, *IEEE Trans. on Electronics Packaging Manufacturing*, 25 (2002) 335-343.
- [4] H. Yan, Z. Chen, Y. Zheng, C. Newman, J. R. Quinn, F.

Dotz, M. Kastler and A. Facchetti, A high-mobility electron-transporting polymer for printed transistors, *Nature*, 07727 (2009) 1-8.

- [5] G. David, *Introduction to Electrodynamics*, Third Ed. Prentice Hall, Upper Saddle River, New Jersey, USA, (1998).
- [6] M. Pudas, J. Hagberg and S. Leppavuori, Printing parameters and ink components affecting ultra-fine-line gravure-offset printing for electronics applications, *J. European Ceramic Society*, 24 (2004) 2943-2950.
- [7] W. C. Walker and J. M. Fetsko, A concept of ink transfer in printing, *American Ink Maker*, 33 (1955) 38-44.
- [8] J. H. De Grace and P. J. Mangin, A mechanistic approach to ink transfer Part 1: Effect of substrate properties and press conditions, *Advances in Printing Science and Technology*, 17 (1984) 312-332.
- [9] N. K. Adam, The chemical structure of solid surface as deduced from contact angle, in contact angle, wettability, and adhesion, *Advances in Chemistry Series*, American Chemical Society, Washington D.C., 43 (1964) 52-56.
- [10] G. B. Dubois, Selecting proper winding tension for various web substrate, *Tappi Journal*, 76 (1993) 91-93.
- [11] D. C. Montgomery, *Design and Analysis of Experiments*, Third Ed. Wiley, New York, USA, (1991).



Changwoo Lee (S'01–M'03) received a B.S. degree in Mechanical Engineering from Konkuk University in 2001. He received his M.S. and Ph.D. degrees from the same university in 2003 and 2008, respectively. He is currently a research professor at the Flexible Display Roll to Roll Research Center,

Konkuk University, Korea, where he is working on the development of R2R multi-layer printing systems for printed electronics. His research interests are in the area of fault tolerant control, R2R e-Printing line design, non-contact transporting process, and tension-register control. He is the holder of several patents related to R2R e-Printing system.



Kee-Hyun Shin (S'81–M'02) received the B.S. degree from Seoul National University, Seoul, Korea, and the M.S. and Ph.D. in Mechanical Engineering from Oklahoma State University (OSU), Stillwater. Since 1992, he has been a Professor with the Department of Mechanical and Aerospace Engineering, Konkuk University, Seoul,

Korea. He has covered several research topics in the area of web handling, including tension control, lateral dynamics, diagnosis of defect rolls/rollers, and fault-tolerant real-time control in the Flexible Display Roll-to-Roll Research Center, Konkuk University, of which he has also been a Director. His research topics include distributed real-time control, embedded control, monitoring, and diagnosis and fault-tolerant control of large-scale systems such as steel plants, film-and-paper-making plants, aircraft, ships, and ubiquitous control of multirobot systems. He is the author of Tension Control (TAPPI Press, 2000).