Robotics and Computer--Integrated Manufacturing 46 (2017) 122–129

Contents lists available at ScienceDirect

Robotics and Computer–Integrated Manufacturing

journal homepage: www.elsevier.com/locate/rcim

Approach to optimizing printed conductive lines in high-resolution roll-to-roll gravure printing

Ho Anh Duc Nguyen[a], Changwoo Lee[b,*], Kee-Hyun Shin[c]

[a] Flexible Display Roll to Roll Research Center, Konkuk University, Seoul 143-701, South Korea
[b] School of Mechanical Engineering, Changwon National University, Changwon 641-773, South Korea
[c] School of Mechanical Engineering, Konkuk University, Seoul 143-701, South Korea

ARTICLE INFO

Keywords:
Multiple performance characteristics
Roll-to-roll gravure printing
Fidelity
High resolution
Grey analysis
Taguchi method

ABSTRACT

Roll-to-roll gravure printing has recently gained considerable interest regarding its application for manufacturing printed electronics, owing to its potential for processing large areas at low costs with high throughput. The geometry of the printed lines depends mainly on the process parameters. Unfortunately, missing areas and well-defined line widths have opposite tendencies. This paper presents a multi-response optimization process for printing high-resolution conductive lines using roll-to-roll gravure printing. Our optimization is based on grey relational analysis and an analysis of variance in conjunction with the Taguchi method, which uses an orthogonal array. Together, these techniques are used to optimize the printed pattern geometry and missing areas. Furthermore, we investigate several parameters for roll-to-roll gravure printing, such as ink viscosity, printing speed, and nip pressure, and the effect of these parameters on the line width, thickness, and missing areas of the printed pattern. Experiments were conducted to evaluate the proposed method, and the results of this evaluation demonstrate an improvement to the well-defined line width, thickness, and continuity of conductive lines under optimal parameter settings using the proposed grey-based Taguchi method.

1. Introduction

Printed electronics have received increasing attention, because even large electronic devices can be printed at low costs. Flexible substrates can now be used to print a wide range of devices, including thin-film transistors [1], sensors [2], organic photovoltaic devices [3], and integrated circuits [4]. It is evident that the geometry of printed devices significantly affects its electrical performance. For instance, the efficiency of photovoltaic devices can be enhanced by improving the resolution and line width of the grid electrode [5]. Furthermore, a reduction in the channel length of thin-film transistors increases the transition frequency and the output current, thus improving the performance of RFID tags [6] and OLED displays [7], respectively. In addition, with a channel length smaller than 5 μm, a fully overlapped structure is possible for thin-film transistors, allowing for the use of printing techniques with poor layer-to-layer registration. As a result, the ability to generate high-resolution conductive lines is crucial for electronic applications.

In an attempt to print high-resolution conductive lines, various printing techniques have been proposed, such as inkjet printing [8] and electrohydrodynamic jet printing [9]. However, these printing techniques run at very slow speeds, owing to the high number of sequential ink drops required to build a pattern. Furthermore, low-viscosity ink (i.e., ink with a low metal content) is popularly used to facilitate inkjet printing. Accordingly, multiple inkjet passes are often required to fabricate highly conductive lines, contributing to even lower throughput. To improve productivity and to facilitate large-area printing, roll-to-roll (R2R) printing has been investigated. R2R printing is a relatively simple process with the potential for mass printing at high volumes using multiple techniques, including flexography [10], gravure offset [11], and gravure [12–14]. Among these techniques, gravure printing is significantly advantageous because the gravure printing roller is not deformed by the nip pressure on the printing area. Consequently, gravure printing is more stable and reliable than gravure offset [15] and flexography [16].

The use of direct gravure printing can achieve very high resolutions (< 2 μm). Kitsomboonloha et al. demonstrated that direct gravure printing can deliver highly conductive lines as fine as 2.38 μm using etched silicon wafers as gravure plates [17]. Zhang et al. [1] proposed a novel method for creating functionalized gravure rolls that enhance the wettability contrast between cells and the surrounding lands, allowing excess ink to be removed without using a doctor blade. Their method

---

* Corresponding author.
E-mail address: leecw@changwon.ac.kr (C. Lee).

http://dx.doi.org/10.1016/j.rcim.2017.01.003
Received 2 December 2015; Received in revised form 22 December 2016; Accepted 19 January 2017

uses photolithographic techniques on a thin flexible silicon wafer, and they demonstrated its ability to print continuous silica nanoparticle lines as narrow as 1.2 μm. Although photolithographic techniques can be used for extremely precise and reliable patterns, etched silicon wafers are rarely used as gravure plates in laboratory settings, thereby precluding the application of such methods to mass-produced, large-area R2R manufacturing.

In general, the functionality of gravure printed conductive lines depends on both the conductivity of the ink [17] and the geometry of the printed patterns – including missing areas, a well-defined line width (i.e., in terms of the line width and the roughness of the line edge), surface roughness, and thickness [18]. It was reported that lines as thick as 50 nm are sufficiently conductive for many printed electronics. Therefore, the quality of functional lines can be determined exclusively by the line geometry, provided that high-performance materials are used (e.g., 5-nm Ag particles or graphene ink). The geometry of printed patterns is highly dependent on the process parameters, such as the printing speed, nip pressure, and ink viscosity. Unfortunately, missing areas and well-defined line widths were found to have opposite tendencies in the presence of these process parameters [19,20]. In this paper, we address these challenges by suggesting an effective strategy for defining the optimum conditions for printing conductive lines with high-resolution R2R gravure printing.

In this work, we employ the Taguchi method to design experiments, by stipulating the dominant level of each factor with an $L_9(3^4)$ orthogonal array. Three outputs – the missing area, line width, and thickness of the printed line – are characterized as a function of the ink viscosity, printing speed, and air nip pressure. To integrate multiple outputs and determine the optimal combination of processing parameters, grey relational analysis (GRA) is used. To quantify the effect of the process parameters, and to determine the most significant parameters, an analysis of the variance (ANOVA) and *F*-test values are computed. Finally, the properties of an optimal printed line are confirmed and compared to initial conditions and the printed lines from Taguchi's design.

2. Experiment

2.1. Printing conditions

An R2R gravure printer is composed of two cylinders: an engraved gravure roll and a nip roll. Ink from an ink fountain wets the surface of the engraving and fills the cell. A blade is used to remove excess ink from the non-image area of the engraved roll. A plastic substrate moving between the engraved roll and the nip roll is brought into contact with the ink at the contact area. Ink is then transferred, resulting in a printed pattern on the plastic substrate. The schematic in Fig. 1(a) illustrates the R2R gravure printing process. There are four phases associated with R2R gravure printing: inking, doctoring, printing, and setting. Each phase of the process has been detailed in previous research [21]. The experiments were conducted on a custom-made gravure printing machine in a Class-1000 cleanroom environment (23 °C and 50% relative humidity). This custom printer is shown in Fig. 1(b).

The gravure printer was manufactured by CoreTool Co., Korea with 150 mm diameter and 320 mm in length; it was made of steel, plated with copper, engraved with laser-stream process, and finally plated with chromium. A lamella doctor blade under a fixed pressure of 3 MPa was used to remove the excess ink on non-engraved surface. The contact angle of doctor blade was 30° tangent to the engraved roll circumference. To produce high-resolution conductive lines, a groove was designed with a line width of 20 ± 0.5 μm with the relative angle of 45° to the printing direction. Fig. 2 shows detailed geometry of the engraved cells as well as printed patterns.

Conductive lines were printed on 300 mm×0.1 mm polyethylene terephthalate (PET) film (SH34, SK, Korea) using three kinds of conductive silver pastes (Paru Co., Korea) and sequentially dried in a dryer that was 5 m in length with air blowing at 150 °C. We varied the air nip pressure and speed of the printer, and we used a fixed pressure of 3 MPa for the doctor blade. The process parameters are shown in Table 1.

2.2. Printed-pattern characterization

The quality of printed patterns depends on the line width (w_P) and the missing area (Δ_M). Engraved patterns were analyzed using an optical microscope (HS-300U, E-flex, Korea), and missing areas were computed as follows:

$$\Delta_M = \left(1 - \frac{A_{Missing}}{A_{cell}}\right) \times 100 \tag{1}$$

where $A_{Missing}$ and A_{cell} denote the void area of the printed pattern and the top-view area of the engraved cell, respectively. The thickness t_P of the printed pattern was measured using a surface profiler (NV 2000, Nano System, Korea). Magnetic tape was used to fix the sample during the measurement. During each printing trial, eight samples were collected.

2.3. Taguchi's method

Conventional experiments involve holding certain factors constant while altering variables. This strategy is inefficient, however, when compared to design of experiment (DOE) that involve simultaneously varying certain factors. The DOE method is more informative, and it requires less experimental data [22]. In DOE method, a full factorial experimental parameter design with three factors and three levels comprises 3^3=27 experimental runs. However, this approach is time consuming and expensive, owing to an increased number of inputs and levels [22]. To address this challenge, the Taguchi method uses orthogonal arrays (OAs) to study the entire parameter space with only a few experimental runs [23]. The Taguchi method begins with problem definitions that determine dependent variables—i.e., inputs and their levels as well as outputs. The next step involves selecting a suitable OA, according to the number of inputs that will be studied and

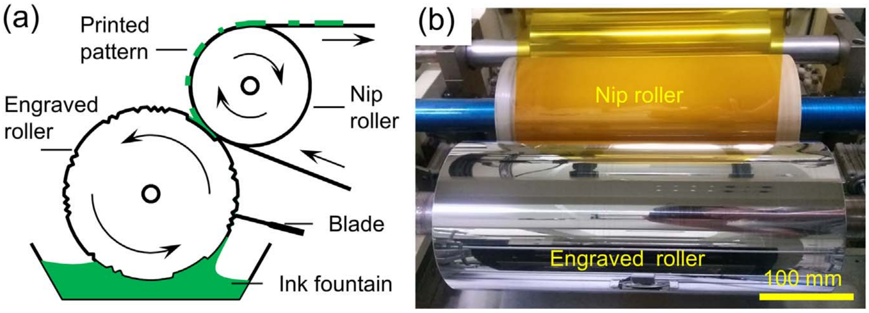

Fig. 1. R2R gravure printer: (a) schematic drawing; (b) custom-made machine.