

# High-resolution gravure printed lines: Proximity effects and design rules

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## ABSTRACT

Gravure printing is a very promising method for printed electronics because it combines high throughput with high resolution. Recently, printed lines with  $2\mu\text{m}$  resolution have been demonstrated at printing speeds on the order of  $1\text{m/s}$ . Here we build on these results to study how more complex patterns can be printed that will ultimately lead to printed circuits. We study how the drag-out effect leads to proximity effects in gravure when multiple lines are printed close to each other. Drag-out occurs as the doctor blade passes over the roll surface to remove excess ink from the land areas in between the cells that make up the pattern. In addition to this desirable removal of excess ink, some ink from the cells also wicks up the doctor blade and is removed from the cells. This ink is subsequently deposited on the land area behind the cells leading to characteristic drag-out tails. If multiple lines, oriented perpendicular to the print direction, are printed close to each other, the ink that has wicked up the doctor blade from the first line will affect the drag-out process for subsequent lines. Here we show how this effect can be used to enhance print quality of lines as well as how it can deteriorate print quality. Important variables that will determine the regime for printing optimization are ink viscosity, printing speed, cell size, cell spacing and relative placement of lines. Considering these factors carefully allows one to determine design rules for optimal printing results.

**Keywords:** Gravure printing, printed electronics, high-resolution printing, high-speed printing, proximity effects, design rules, assist features

## 1. INTRODUCTION

Printed electronics is an emerging technology that holds great promise to enable light-weight, low-cost, flexible electronic systems on substrates such as plastic or paper.<sup>1-8</sup> One of the main challenges in pushing printed electronics forward is the precise patterning of high-resolution features. Photolithography has seen a great effort to understand fundamental limits on resolution and proximity effects that arise from optical interactions. Printed electronics is still at the early stages of this process. Great progress has been made in pushing the resolution of different printing techniques. Especially gravure printing has proven to enable high-resolution printing below  $5\mu\text{m}$  feature size at high print speeds on the order of  $1\text{m/s}$ .<sup>9,10</sup> Most of these results have been obtained for isolated lines aligned with the printing direction. Here we investigate lines printed perpendicular to the printing direction, which is far more challenging than printing lines aligned with the printing direction. We show how printing changes as lines are brought into close proximity. Understanding of these proximity effects is crucial when designing patterns such as closely spaced grids or interdigitated electrodes or indeed any high-resolution circuits of arbitrary shape.

The gravure printing process consists of a number of sub-processes as illustrated in Figure 1. First ink fills the cells that make up the pattern. These cells are a pixelated version of the pattern unlike intaglio where the pattern is made up of continuous trenches. Then a doctor blade is used to scrape excess ink off the land areas in between cells as well as remove excess ink on top of the cells. After the wiping process, ideally, only the cells contain ink whilst the land areas are clean. Ink is then transferred from the cells onto a substrate such as plastic or paper. Finally, the ink on the substrate spreads to fill in the gaps in between cells. All of these sub-processes are susceptible to non-idealities that degrade print quality. This is especially true for the wiping process. One problem is lubrication residue left on the land areas due to a lubrication flow of ink underneath the blade.<sup>11</sup> Another problem is the drag-out effect. As the doctor blade passes over cells, a meniscus of ink climbs up the backside of the blade. Firstly, this means ink is removed from cells, which can lead to thinner and ultimately broken lines. Secondly, this ink is subsequently deposited behind cells leading to characteristic drag-out tails. Both of these consequences of the drag-out effect make printing of lines perpendicular to the

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printing direction challenging. In lines that are aligned with the print direction any drag-out gets immediately re-deposited into the next cell so that no ink is lost and there is only a tail at the end of the line. The other consequence of drag-out is proximity effects. The tails tend to be very thin. However, even if they don't affect electronic performance of the circuit directly, the fact that they can form a liquid connection between subsequent lines leads to proximity effects that alter pattern definition depending on line spacing. The detailed physical mechanism for these phenomena is still under active investigation and will be reported elsewhere. Here we characterize this effect and show how design rules can be determined that will guide both pattern design as well as the choice of printing parameters such as ink viscosity and printing speed for different values of line spacing. We also show how this effect can be exploited using assist features to optimize line definition analogous to optical proximity correction (OPC) in photolithography.<sup>12-15</sup>

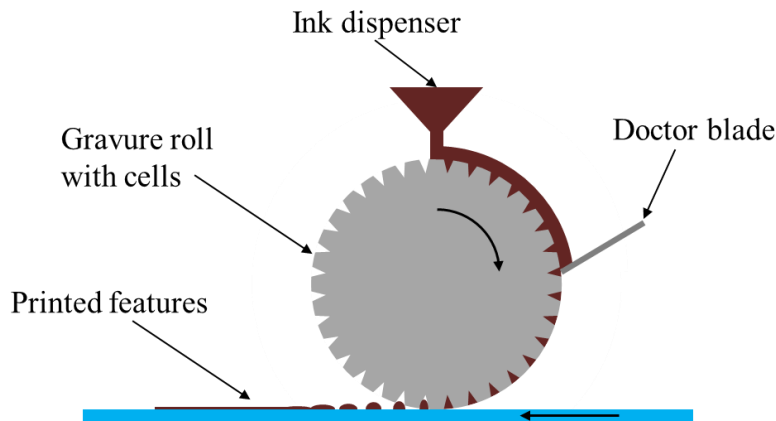


Figure 1. Illustration of the gravure process showing cell filling with ink, wiping of excess ink with a doctor blade, transfer of ink from the cells to the substrate and spreading of discrete droplets to form continuous features.

## 2. EXPERIMENTAL

Groups of lines were gravure printed with varying line spacing. Lines were oriented perpendicular to the printing direction to maximize the impact of the drag-out effect. The cell size was varied between 2 and 4 $\mu$ m and the spacing or gap between cells within lines was varied between 15 and 60% of the cell size. Lines were one cell wide and the ideal printed linewidth is thus identical with the cell size. In reality ink will always spread somewhat to increase linewidth. Figure 2 illustrates the pattern variables studied here. Inverse direct gravure printing was used where ink is transferred from a flat printing plate onto a plastic substrate wrapped around a roll. The printing plate is fabricated from a silicon wafer to achieve optimal pattern definition. Metal replicas of these silicon printing plates can be used to fabricate high-resolution gravure rolls as reported elsewhere.<sup>16</sup> This means our results here can translate to roll-based direct gravure printing to allow high-throughput manufacturing. The printer used in this study is a custom built model with a maximum printing speed of 1m/s. The ink was NPS purchased from Harima Chemicals, a silver nanoparticle ink with 12nm particle size. A silver ink was chosen due to its reflective optical properties when imaged as well as its widespread use in the fabrication of printed electrical circuits. After printing, drying and sintering, the printed patterns were imaged by optical microscopy. The results were analyzed using an automated technique to extract print quality metrics, namely the fraction of lines merged together, the fraction of discontinuous lines, ink spreading and edge roughness. Spreading was defined as the average distance by which lines extend over their intended edge on either side. Edge roughness was defined as the standard deviation of the line spreading. Spreading and edge roughness were normalized by cell size to compare lines of different cell sizes.

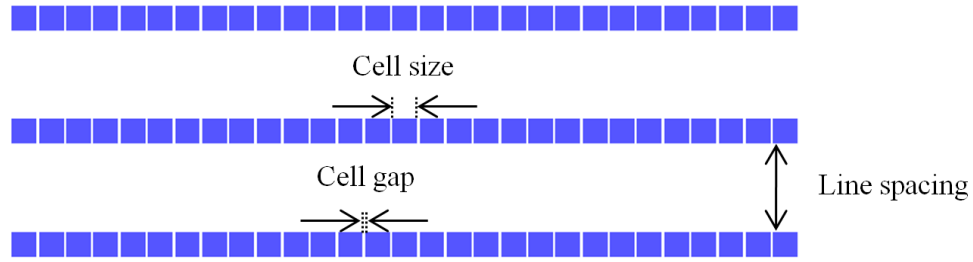


Figure 2. Pattern variables studied here: Cell size, cell gap (spacing) and line spacing

### 3. PROXIMITY EFFECT

Figure 3 shows the effect of bringing high-resolution lines into close proximity. One can note that the shape of the lines changes markedly as the distance between lines is changed. If lines are placed too close to each other, they will merge due to the spreading of the ink after transfer onto the plastic substrate. As the spacing between lines is increased, lines become thinner with less ink volume per line. In this case a line distance of  $6\mu\text{m}$  or two cell widths gives optimal line shapes. With different cell sizes, cell gaps and printing conditions the optimal line distance will be different. At larger line distances line edge roughness increases and lines can even become discontinuous. This happens because there is not enough ink in the lines to fill in all the gaps between the cells. This trend holds for all lines except for the first line in each print. In these micrographs the bottom line is always the first one that was filled, wiped and transferred. The first line consistently has less ink than subsequent lines. When line spacing is increased far enough that lines can be treated as isolated lines, subsequent lines start to exhibit similar behavior to the first line. Lines become isolated when they are not connected by drag-out tails anymore. One can observe faint drag-out tails behind lines, especially when they are sufficiently separated. Lines start to not being connected by drag-out tails at a line distance of  $15\mu\text{m}$  and are fully separated at a line distance of  $21\mu\text{m}$ . One can clearly see that this separation of the lines coincides with an increase in line edge roughness. This suggests that the connection of lines by drag-out tails is responsible for the observed proximity effects. This also explains why the first line behaves consistently different from subsequent lines since it is the only line that is not connected to another line's drag-out tail at its leading edge. We also conducted an experiment where the printing plate was wiped in the usual way but the printing direction was reversed for the transfer process. We observed the same directionality of the proximity effect as before suggesting it is due to the directionality of the wiping process rather than any directional process during transfer.

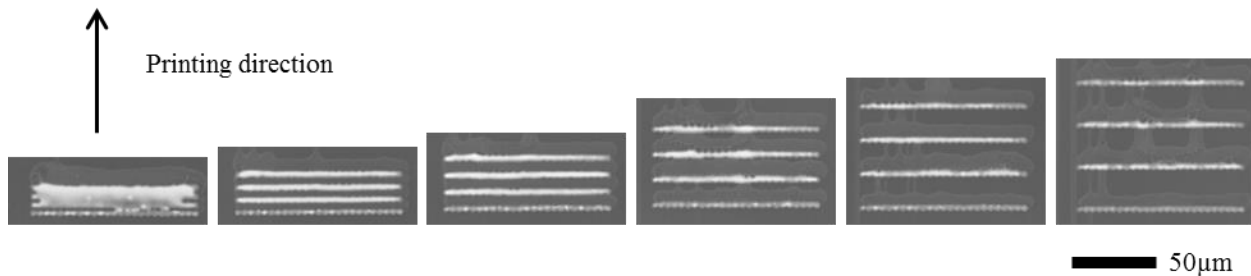


Figure 3. Optical micrographs of printed features demonstrating proximity effect. Very closely spaced lines merge together. Lines with medium spacing show optimal printing. Further increases in line spacing lead to lines with large edge roughness. The first line always resembles the shape of isolated lines. The cell size is  $3\mu\text{m}$ , cell gap  $1.8\mu\text{m}$ , printing speed  $0.25\text{m/s}$ , ink viscosity  $68\text{cP}$ . The distance between lines varies from left to right as  $3, 6, 9, 15, 21$  and  $27\mu\text{m}$ .

#### 4. DESIGN RULES

When choosing printing conditions, one of the key variables is ink viscosity. Viscosity together with printing speed will determine capillary number  $Ca = \frac{\mu U}{\gamma}$  where  $\mu$  is ink viscosity,  $U$  is printing speed and  $\gamma$  is ink surface tension. The capillary number is a dimensionless number that represents the ratio of viscous to surface tension forces. Most gravure printing processes are strongly influenced by  $Ca$ .<sup>9</sup> This is the case for cell filling, wiping and ink transfer. Conversely, ink spreading on the substrate after transfer does not depend on printing speed but rather on ink viscosity and the wetting properties of the substrate for any particular ink. Here we varied ink viscosity whilst keeping  $Ca$  constant to study the effect of ink spreading on the substrate and find the optimum viscosity for these patterns (see Figure 4). The lower the ink viscosity, the easier it will be for the ink to spread on the substrate. The lowest viscosity ink studied here at 34cP spreads the most. This limits the minimum feature size that can be achieved, both in terms of line width, as well as minimum line spacing. Lines printed with low viscosity inks are more prone to merge together. In an electrical circuit this would result in a short circuit, which would be a fatal flaw destroying the functionality of the circuit. A design rule must thus take ink spreading into account and put a lower limit on line spacing to achieve reliably separated lines. This minimum line spacing will be larger for the 34cP ink than for the other inks studied here. Thus the 34cP ink is not optimal. On the other hand, if the viscosity is too high, there is not enough spreading. Gravure relies on ink spreading to fill in the gaps between discrete cells and form continuous lines. If the ink does not spread enough, line edge roughness increases. There will be less ink in the cell gaps in between cells leading to scalloped lines where the wavelength of the scallops corresponds to the placement of cells. In the more extreme case lines can become discontinuous leading to open circuits. This is another fatal flaw that would render a circuit unusable. The highest viscosity ink studied here at 195cP shows both increased line edge roughness as well as a much larger fraction of discontinuous lines. This is especially true for lines spaced further apart that don't get the benefit of the proximity effect. 68cP was found to be the optimum ink viscosity for the patterns studied here leading to the optimum amount of spreading that minimizes both the merging of adjacent lines and the number of discontinuous lines. This viscosity was used from now on to study the effect of printing speed and cell gap.

The interaction between printing speed, cell gap, cell size and line spacing is complex. The main competing effects are drag-out, proximity effects and lubrication residue. Drag-out occurs due to ink wicking up the doctor blade. This is driven by surface tension forces and is inhibited by viscous forces. Drag-out therefore becomes more severe at smaller capillary numbers. Drag-out also becomes more severe for smaller cells because a larger fraction of the ink in the cell can be dragged-out by the doctor blade. Similarly, proximity effects become more pronounced at low capillary numbers and for smaller cells because of increased levels of drag-out as well as increased flow between lines. Conversely, lubrication residue becomes more severe at high printing speeds and viscosities. This film is due to ink that passes under the doctor blade. The fluid pressure in this gap between the doctor blade and the printing plate balances the loading force on the doctor blade. This pressure increases with viscosity and speed leading to thicker residue films. In theory this residue is deposited uniformly across the printing plate; however, in practice residue is concentrated in areas of doctor blade defects due to wear.

These different effects control the amount of ink left in the cells after wiping. When trying to print ideal lines, one needs to have the ideal amount of ink per unit length. If the amount of ink per unit length is too large, lines will bulge, become non-uniform and ultimately merge with adjacent lines. If there is not enough ink, lines will become scalloped and ultimately break up. The amount of ink per unit length is not only governed by the amount of ink inside cells but also by the cell spacing within each line. If cells are spaced further apart, there is less ink per unit length and vice versa. Thus it is important to have the optimum amount of ink inside the cells combined with the right cell spacing. These considerations are similar to previous work on inkjet printed lines, although in a higher viscosity regime and with simultaneous printing of all droplets that make up a line.<sup>17</sup>

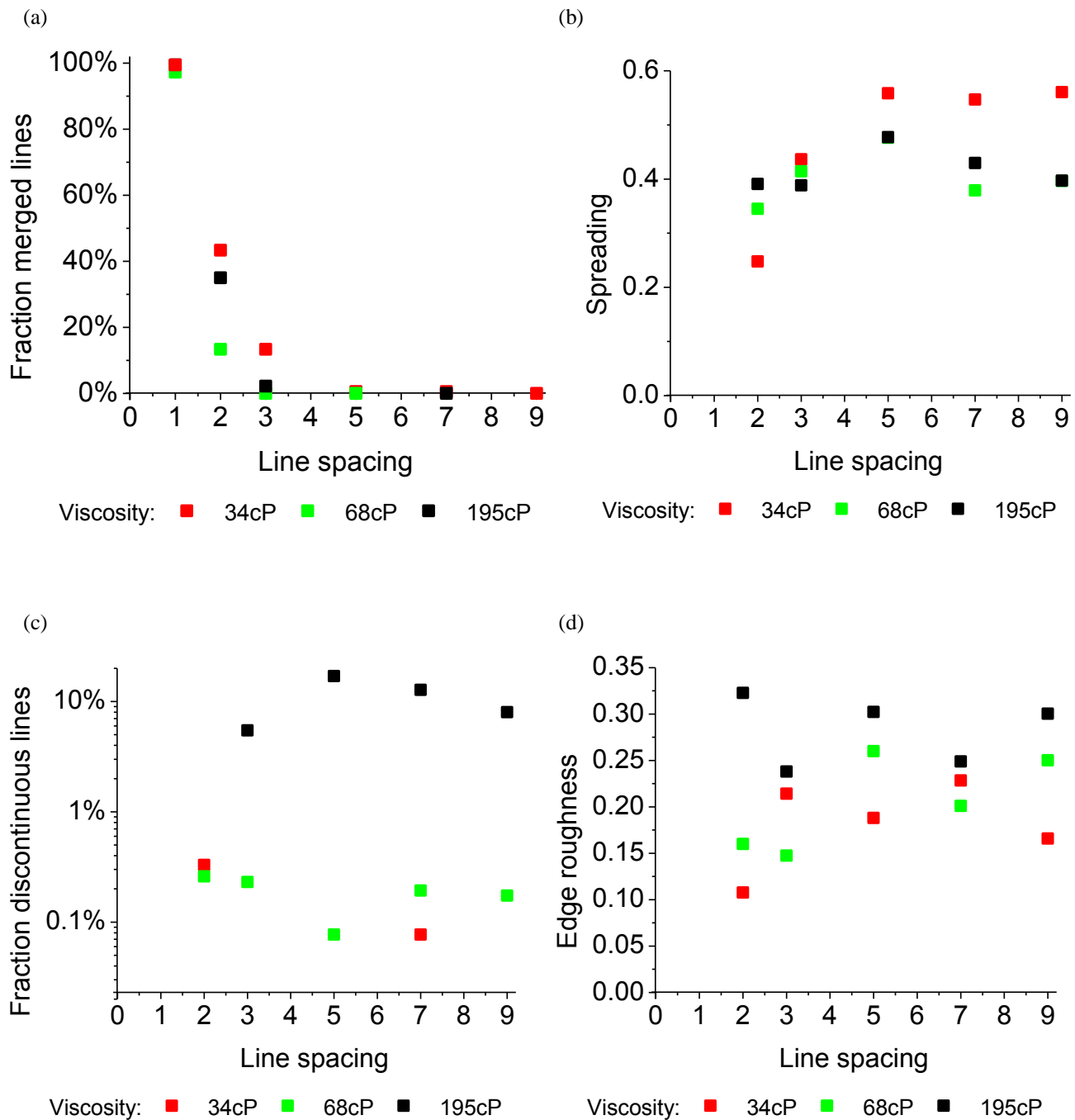


Figure 4. Effect of ink viscosity on printed pattern quality. (a) & (b) The lowest viscosity tested here (34cP) exhibits increased line spreading and subsequent merging of closely spaced lines. (c) & (d) The highest viscosity tested here (195cP) exhibits increased edge roughness and increased numbers of discontinuous lines. The optimum viscosity is thus 68cP. Line spacing, edge roughness and spreading were normalized by cell size.

The goal of understanding these processes is to create design rules that can be used to make choices when designing gravure printed circuits. A principal challenge is that printing conditions such as speed and viscosity apply globally to all patterns printed at the same time. If a circuit contains a diverse set of patterns such as different line widths and separations, these different patterns might require different conditions for optimal printing. The designer has the freedom to change the cell spacing to tune the optimal conditions for a particular pattern and ameliorate conflicts but the scope of this is limited. Consider the case where a design includes a set of closely spaced  $4\mu\text{m}$  lines, such as bus lines packed tightly into a confined area, as well as isolated  $2\mu\text{m}$  lines, such as in a sparse grid. Representative printing results and extracted print quality metrics for these cases can be found in Figure 5 and Figure 6 respectively. The closely spaced  $4\mu\text{m}$  lines show optimum line edge roughness at low printing speeds, especially  $0.25\text{m/s}$ . At higher printing speeds drag-out becomes less pronounced leading to a larger amount of ink inside the cells. There is also more lubrication residue at higher printing speeds. This leads to excessive and uneven spreading of lines and can even result in lines merging together. Conversely, isolated  $2\mu\text{m}$  lines print best at high printing speeds of  $0.5\text{m/s}$  or  $1\text{m/s}$ . At lower printing speeds drag-out pulls too much ink out of these small cells. This leads to scalloped edges and even discontinuous lines. This is the case even though cell spacing was already optimized for both cell sizes for the respective conditions. This simple example shows the difficulties when trying to print a diverse range of high-resolution patterns and will only get worse with more complex circuits. Since global printing conditions need to work for all patterns in a print, local patterns need to be adapted to make them compatible. Assist features exploiting proximity effects can be one method to achieve this.

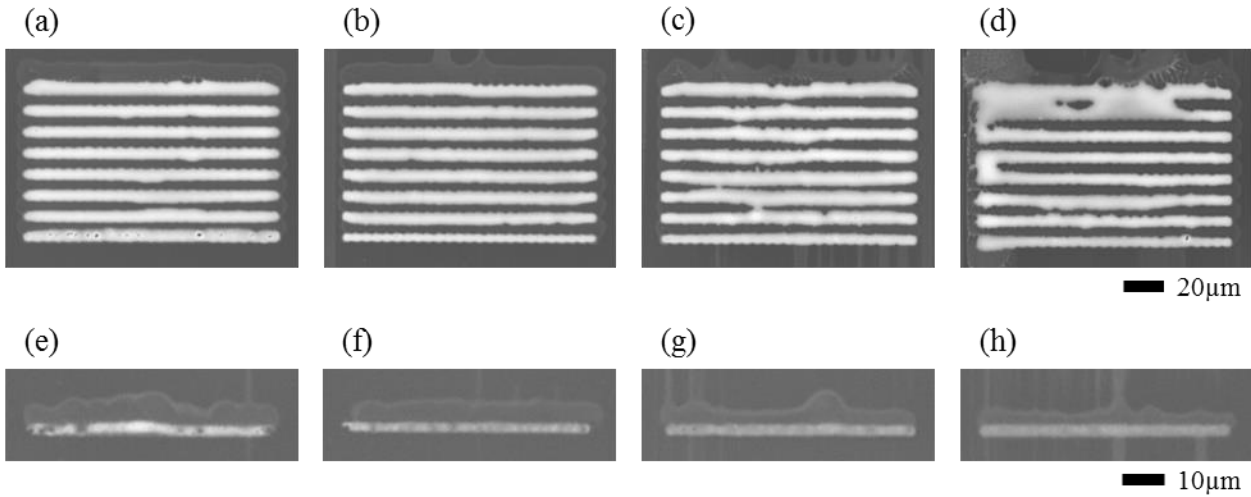


Figure 5. Optical micrographs of closely spaced lines with  $4\mu\text{m}$  cell size and  $8\mu\text{m}$  line spacing (first row (a)-(d)) and isolated lines with  $2\mu\text{m}$  cell size (second row (e)-(h)). In both rows printing speed increases from left to right as  $0.125\text{m/s}$ ,  $0.25\text{m/s}$ ,  $0.5\text{m/s}$  and  $1.0\text{m/s}$ . These two cases exhibit opposite trends of print quality with printing speed. Closely spaced  $4\mu\text{m}$  lines exhibit better line definition with slower speed whereas isolated  $2\mu\text{m}$  lines need to be printed at higher speeds.

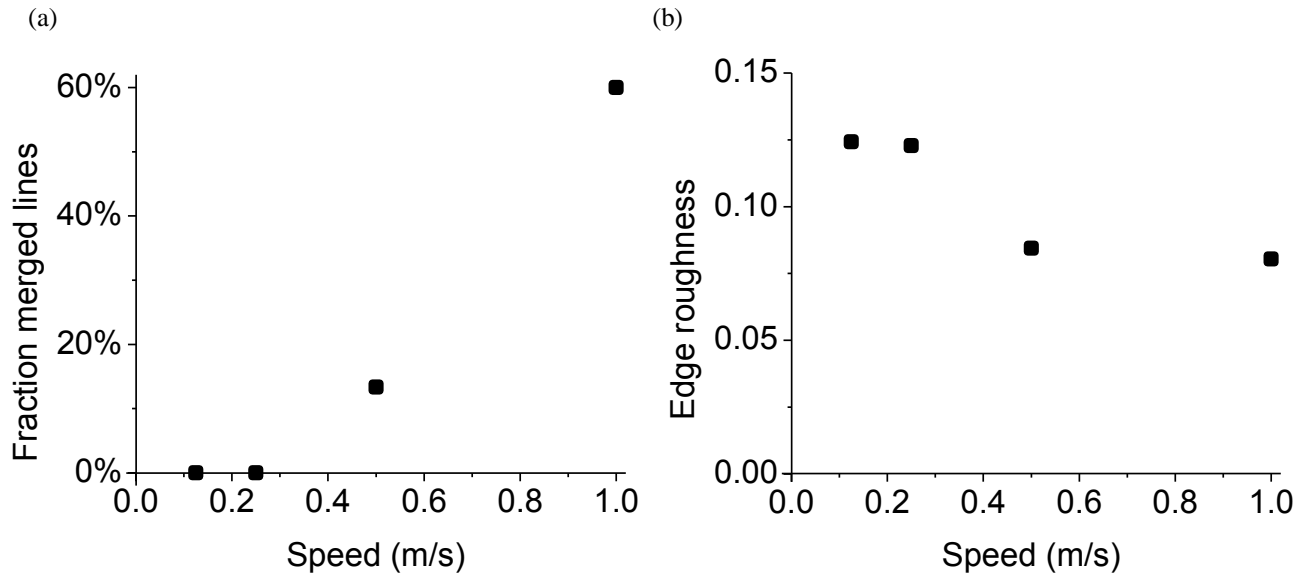


Figure 6. (a) Fraction of merged lines increases with increasing printing speed for lines with  $4\mu\text{m}$  cell size that are closely spaced  $8\mu\text{m}$  apart. (b) Edge roughness drops with increasing printing speed for isolated lines with  $2\mu\text{m}$  cell size. Edge roughness was normalized by cell size.

## 5. ASSIST FEATURES

Optical lithography uses assist structures to print high-resolution features. This method exploits optical proximity effects. Sub-resolution features such as scattering bars are placed next to the feature to be printed. For isolated lines this simulates the situation of densely spaced lines. Even though the assist feature does not print itself, its wave function can constructively interfere with the wave function of the actual feature and assist in its printing. Here we propose a similar method for gravure printing to exploit fluidic proximity effects. The real feature is placed behind the assist feature so that it is located inside the assist feature's drag-out tail. This will add ink to the real feature and make it printable. Here we used assist features that are of the same size as the real feature to be printed. The print quality of the assist feature is somewhat lower because it is not supported by the proximity effect but it will still be printed. In the future more sophisticated assist features could be explored that don't transfer or only transfer minimally to the final substrate.

We demonstrate this concept of fluidic assist features with the example of isolated high-resolution  $2\mu\text{m}$  lines oriented perpendicular to the printing direction. Such lines cannot be printed easily at low printing speeds around  $0.25\text{m/s}$ . Lines tend to break up due to excessive drag-out. With the extra ink from the assist feature such lines can successfully be printed. The result can be observed in Figure 7.

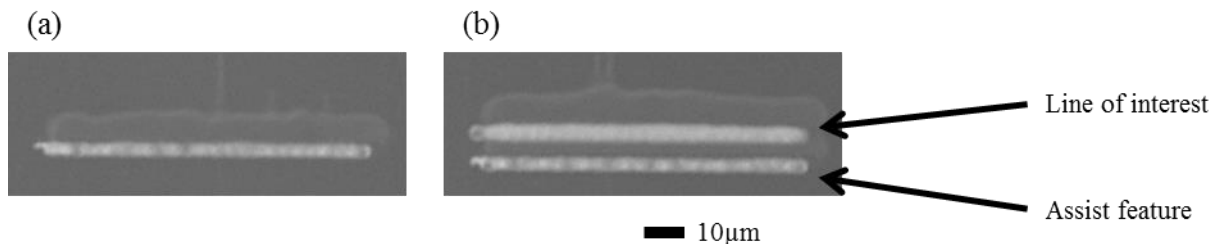


Figure 7. Optical micrographs showing improved print quality of line with  $2\mu\text{m}$  cell size printed at  $0.25\text{m/s}$  (b) with assist feature compared with (a) an isolated line. The isolated line shows intermittent holes due to drag-out that can lead to discontinuous lines. The line with a preceding assist feature exhibits no such holes due to the proximity effect. The assist feature behaves similar to the isolated line.

## 6. CONCLUSION

Proximity effects in high-resolution gravure printing were studied here for the first time. Proximity effects in gravure printing are due to the drag-out effect that creates tails of ink behind features. Lines oriented perpendicular to the printing direction were studied to demonstrate the implications of these proximity effects. Lines perpendicular to the print direction are significantly more challenging to print than lines aligned with the print direction. Proximity effects as well as other printing variables need to be taken into account to print optimal features. It was demonstrated that proximity effects can be exploited by using assist features to print features that otherwise could not be printed reliably.

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