Investigation on lineworks printed with different types of flexographic printing forms for purposes of printed electronics

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Nowadays one can observe fast development of materials and technologies dedicated to inexpensive mass production of electronic devices with conventional printing techniques. One of them is flexography. In this paper investigation on quality of lineworks printed with flexographic printing technique for purposes of printed electronics is presented. There was analysed influence of different printing forms on printed elements reproduction accuracy. Results obtained with conventional graphic ink with commercially available conductive ink were compared as well. Aspects which should be taken under consideration when implementing flexography to printed electronics applications have been specified.
Key words: printed electronics, flexography, printing

Introduction

Printed electronics also known as functional printing or flexible electronics is a fast emerging part of printing industry. Although printing of electronic devices' parts is well established branch of printing industry, e.g. screen printing has been commonly used in production of integrated circuits from mid of twentieth century, rapid and still accelerating growth of this area started when conductive polymers have been discovered [1]. Today many companies, research institutions and universities work on developing new materials (conductive and semiconductive inks, substrates etc.) and technologies, which could be used for manufacturing electronic devices with conventional printing techniques. The reason of all these efforts is that printing machines would produce electronic components with high speed roll-to-roll technique, in high volumes and, what is very important, for reasonable price [2-6]. In result, great benefit which may be offered by printing industry to electronics is possibility to manufacture fully functional devices, for instance solar cells, OLED lighting and displays, sensors and many others, on flexible substrates, such as plastic foils, paper and textiles [2-10].

To print electronic components, next to special coating techniques such as spin coating, slot coating and spraying, conventional printing technologies may be implemented, i.e.: gravure/pad printing, offset printing (lithography), screen printing and flexography [11]. Each of them has its advantages and disadvantages.

Ink-jet printing, for instance, lets to print high resolution images and is very precise technique, however, it is also very slow [12] and can be used only for laboratory research and small and medium batches. Gravure and pad printing (also called offset-gravure) on the one hand offer high resolutions and deposition of high ink film thickness, but on the other hand there may occur wavy lines edges and fluctuations of lines quality due to their alignment regarding to printing direction [13]. What is more, preparation of gravure printing form is very expensive in comparison to other printing techniques.

Screen printing, however, produces thick ink films, but with relatively low resolution [6]. Whereas offset printing produces high resolution images, but printing process needs water-based solution, which may blur edges of printed elements and the ink film thickness is rather small. There are also problems related to construction of inking unit of offset presses, e.g. streaking on the printouts [14,15].

One of the most important advantages of flexography is low printing pressure, what prevents destruction of underlying layers of multilayer electronic devices [17]. It was successfully used to produce optic waveguides [17], parts of fully printed ring oscillator [16] or busbars in solar cell [18]. However, not many works investigated deeply abilities of implementing flexography for electronic purposes and analysed parameters, which influence on quality of printouts. In [19] Deganello et al. analysed grids of different line widths printed with several anilox cells' volumes. Experiment showed that lower resistivity is obtained for higher anilox volumes than those conventionally used in graphic industry, but printed lines are 3-5 times wider than the nominal ones on the printing form. The same authors in [20] investigated lines of different widths and angles and influence of those parameters on the resistivity of printed patterns. It occurred that direction of the lines has minimal effect on their conductivity. Results of experiment also showed that the thinner lines have lower sheet resistances, probably due to better packing of conductive particles than in wider lines.

In both papers [19, 20] authors used silver inks. Analyses were performed after ink sintering in high temperature (130°C) for 5 minutes. Such preparation of samples causes loss of information about lines structure after printing. Such information can be especially valuable when considering conductive inks patterns, which are not sintered after printing, e.g. printed with graphene or carbon nanotubes based inks.

The aim of research presented in this paper was to evaluate influence of different types of printing plates on production of conductive paths for purposes of printed electronics. Important is that the printed patterns were not submitted to high temperature sintering, so quality of unchanged line structures was analysed.

Basics of flexography

Flexography is a technique commonly used in printing industry for production of flexible foil packaging, corrugated boxes and labels. The main advantages of flexo printing are fast printing process (roll-to-roll), low costs of printing plates preparation (in comparison to gravure printing), relative simplicity and good quality of printouts. In figure 1 flexographic printing unit is depicted.

The liquid ink is injected from ink fountain into cells placed on the anilox cylinder. An excess of the ink is

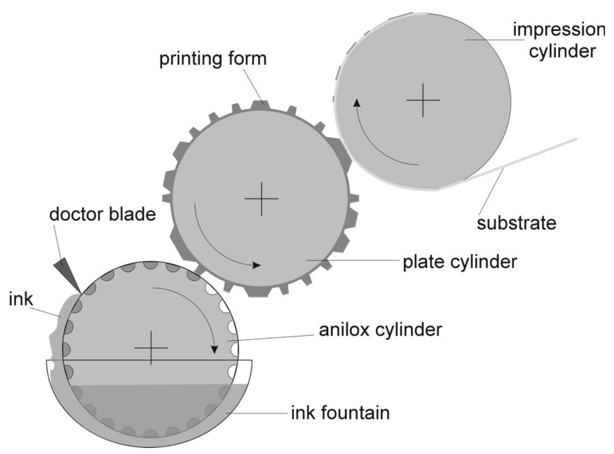

Figure 1. Flexographic printing unit

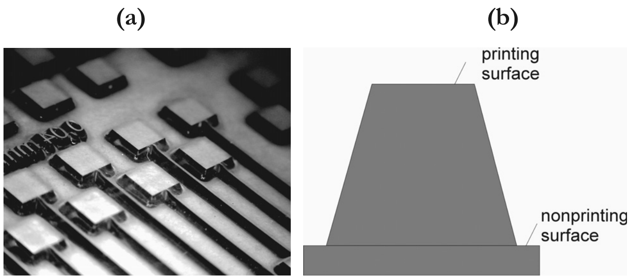

Figure 2. Flexographic printing form (a) and cross section of printing element (b)

cleaned up from the anilox cylinder with doctor blade. The ink, which remained in the cells inks printing elements (Fig. 2) of flexible printing form. Finally inked image is pressed to the substrate transported between plate and impression cylinder.

Experiment

Methodology

In the graphic arts correctness of the print is considered through the prism of colour correspondence to the original image, lack of printing errors noticeable with a bare eye, e.g. misalignment of colours, doubling, streaking, etc. [14, 15]. However printed electronic components have much higher and slightly different requirements both to printing process as well as properties of the image. Most important are:

– stability of printing conditions
– uniformity of printed elements (shape, ink film thickness)
– sharpness of the edges.

All of them may seriously influence on electrical properties of printed element, e.g. conductivity.

For the aim of the experiment special test image was designed. It consisted of elements for printing process control, i.e. printing pressure, increase of elements' sizes, printing resolution (fig. 3):

– positive and negative microlines of widths 10 μm to 500 μm in two directions: 0° and 90° with respect to printing direction
– positive and negative texts of different size

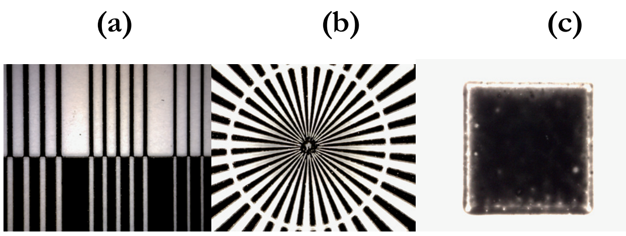

Figure 3. Exemplary positive and negative microlines (a), Siemens star (b) and solid tone patch (c), (printouts)

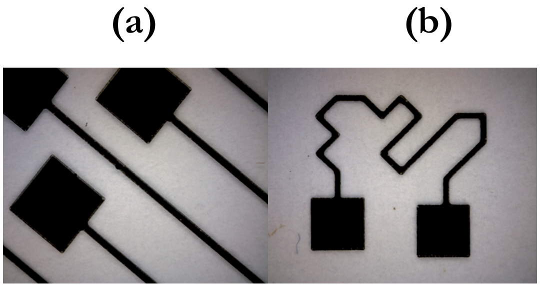

Figure 4. Exemplary lines (a), path (b), (printouts)

Table 1. Parameters of printing plates preparation process

Parameter	Plate		
	Elastomer	Photo-polymer I	Photo-polymer II
Type	Conti Laserline CSX (Continental ContiTech)	Digital Rave (MacDermid Corp.)	Digital Rave (MacDermid Corp.)
Thickness	1.14 mm	1.14 mm	1.14 mm
Upper surface shape	flat	rounded (round top dot)	flat (flat top dot)
Exposure, light finisher and post exposure unit	–	DuPont Cyrel 1000 ECLF	DuPont Cyrel 1000 ECLF
Image-setter	Hell Premiumsetter S1300	ESKO CDI Spark 4835	ESKO CDI Spark 4835 + Inline UV2
Exposure resolution	4000 dpi	4000 dpi	4000 dpi

– solid tone patches (large areas totally covered with ink)
– Siemens star

Test image included also paths equipped with contacts for electrical measurements, fig. 4:

– lines of widths 0.01 µm to 500 µm, in three directions: 0°, 90° and 135° with respect to printing direction
– paths consisting of lines of different directions, sizes: 50 µm x 100 to 500 µm x 100

Test image was transferred onto elastomer and two photopolymer printing plates using two different technologies.

Elastomer plate was laser engraved, i.e. laser beam exposed chosen areas of the plate turning them into dust, which afterwards was brushed away. Unexposed areas of the plate became printing elements (image which is to be printed). In that technology printing elements have flat upper surface.

Polymer plates were prepared in two different variants, in which shape of the upper surface of printing elements was differentiated by slight modification of exposure method. In the first one printing elements have rounded upper (printing) surface (so called round top dot technology). The second variant is called flat top dot technology and printing elements have planar upper surface.

General steps of polymer plates preparation process are following: initial exposing of the plate (exposure unit), laser irradiation of the black mask – creation of negative of the image (imagesetter), main exposure (exposure unit), developing (washing away unexposed non-printing elements of the plate, developer), post-exposure (exposure unit). All exposure times, temperatures and times of developing applied during printing forms preparation were standard from the printing industry point of view and were not optimized for printed electronics applications. Parameters of photopolymer plates and information about equipment used are detailed in tab. 1.

Table 2. Printing process conditions

Printing press	Gallus EM280
Ink	conventional black UV cured flexo ink (Michael Huber) commercial conductive, graphite and graphane based flexographic ink (Vorbeck)
Anilox cylinder	200 lpi, 6 ml/m^2
Substrate	Polyester terephthalate (PET)
Printing speed	30 m/min

Test image was printed on narrow web-fed flexo printing press using polyethylene terephthalate (PET) as a substrate, all three printing plates and two different inks – conventional graphic ink and commercially available graphite and graphene based conductive ink. Other conditions such as temperature, humidity, printing pressure in which printing was conducted were stabile and unchanged during the whole experiment. Parameters of printing process are presented in Tab.2

Quality of the prints was evaluated by microscopic observation and for the images printed with conductive ink additional resistivity measurements were performed.

Results and discussion

Printouts printed with conductive ink were of poor quality. Elements of the image test were porous and had many undesirable artefacts. What is more amount of ink transferred to the substrate was visibly not sufficient (fig. 5). In result no conductivity of printed elements was observed. One of the reasons of such situation would be incorrect surface tension or free surface energy of the ink or printing substrate, respectively. However both of them can be neglected, because ink's printability was tested on the substrate before experiment with handheld bar coaters (RK Printcoat Instruments®) and no undesirable behaviour was observed. What is more, results obtained with conventional ink, which are presented further in this article showed that printing substrate's free surface energy was high enough to let perform printing on it.

Highly probable reasons of the problems with conductive ink transfer were too high viscosity and thixotropy. Such inference is dictated by observed clogging of anilox cylinder's cells and ink solidifying on the printing plate. Diluting of the ink would improve its printability, but it was not recommended by the ink manufacturer hence not performed.

Quality of the prints made with conductive ink was highly unsatisfying, so they were not taken into consideration in further analyses.

The thinnest lines placed on the test were of width 10 m. None of the used printing plates was able to reproduce such thin lines in a proper way, i.e. ensure uniform structure of ink film thickness and smooth edges. In the images printed with elastomeric form there were noticeable wavy microlines broken into many parts. Photopolymer plates managed to print continuous microlines, but with uneven edges and variations of their widths. What is more, only half of them were fully printed, the rest was residually or not reproduced at all.

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The thinnest correctly reproduced lines of width equal to 40 μm were obtained with use of photopolymer printing form and flat top surface technology (Photopolymer II). Similar result (50 μm) was obtained for Photopolymer I (round top dot technology). In both cases lines were parallel to direction of printing, signed as 0°. For elastomeric printing form, the smallest width of correctly reproduced lines was equal to 150 μm and was also obtained for angle zero degrees with respect to printing direction.

Problems with correct reproduction of thin lines are of different nature for each type of printing form. In case of elastomeric plate, one of the main reasons is high porosity of the material, which prevents reproduction small elements and sharp edges (fig. 6).

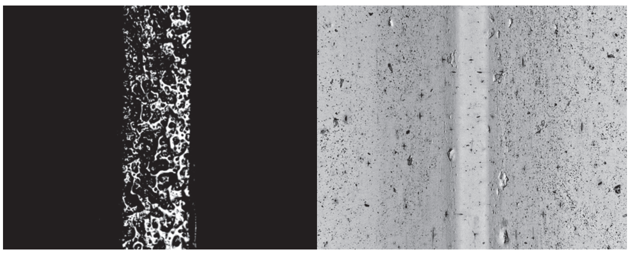


Figure 6. Surface of elastomer (a) and photopolymer (b) printing forms, (visible exemplary lines, mag.: x400)

As seen in fig. 6, photopolymer's structure is much more uniform than the elastomer's one, so in that case probable reason of lines inhomogeneity may be related to parameters of printing form preparation process. Most important parameters of that process are main exposure time and developing time. The first one is responsible for quality (degree) of polymerization and if too short, printing elements will be fragile and not properly bounded to the plate base. The second one regulates process of washing out of unexposed photopolymer. During developing temporary swelling of printing elements occurs. If time of developing is too long, printing elements may be damaged or permanently deformed.

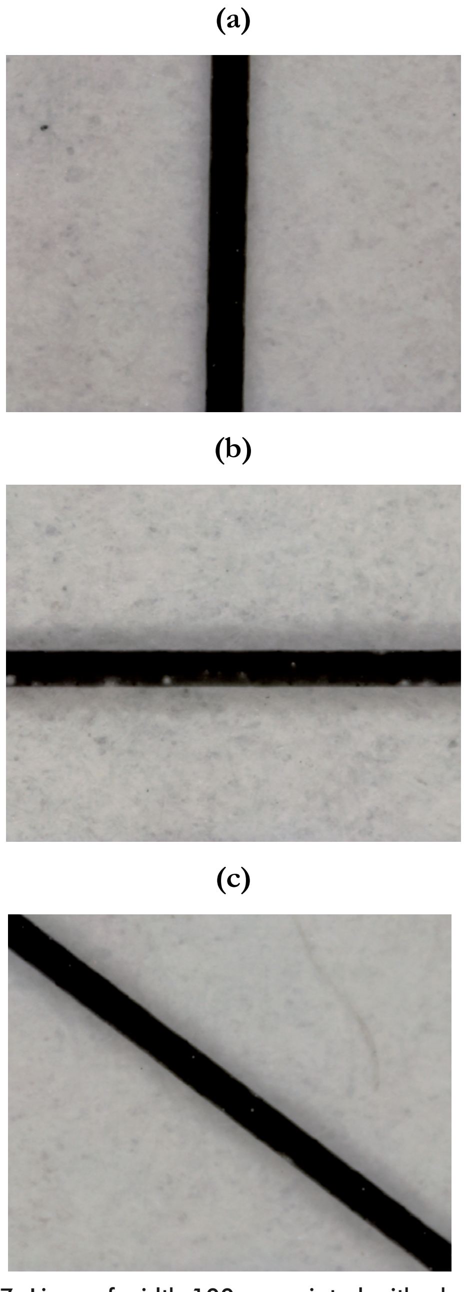


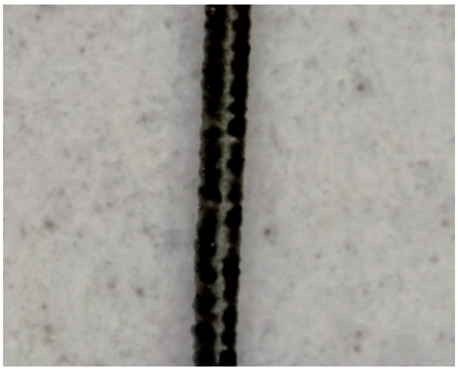
Figure 7. Lines of width 100 μm printed with photopolymer II printing plate in direction 0° (a), 90° (b) and 135° (c), with respect to printing direction (mag.: x230)

Aforementioned parameters of plate preparation process were optimized for standard graphic industry works, not for very thin lineworks such as electronic paths. It shows, that parameters of printing form and its preparation process should be deeply taken into account if small elements (up to size of about 50 μm) are going to be printed.

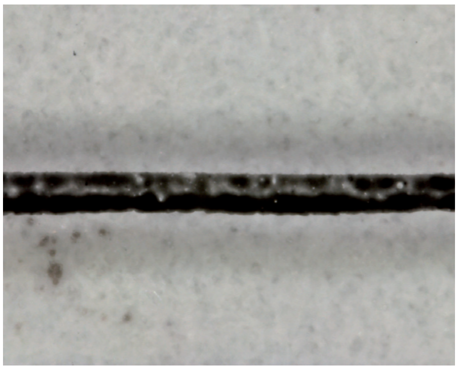
Another reason of poor reproduction of thin lines printed with both types of printing plates may be damages of their printing surface due to high shear forces acting on them while printing. This fact may suggest that either shape of the thinnest printing elements or material of printing form should be modified to enhance their high shear loads strength.

Experiment also showed that direction of the lines has influence on their quality on the prints. It is especially noticeable when the smallest width of accurately reproduced lines is considered. At the angle of 90°, the lines of width 100 μm and 150 μm for polymeric and elastomeric plates, respectively, were properly reproduced. Whereas lines parallel to printing direction of widths equal to or higher than 40-50 μm (photopolymer plates) and 150 μm (elastomeric plate) were well reproduced . What is more, alignment of lines has influence on their inner structure uniformity and

(a)



(b)



(c)

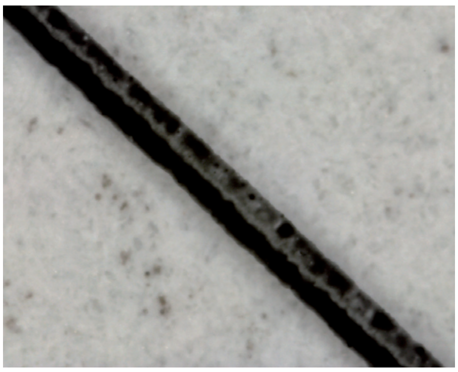
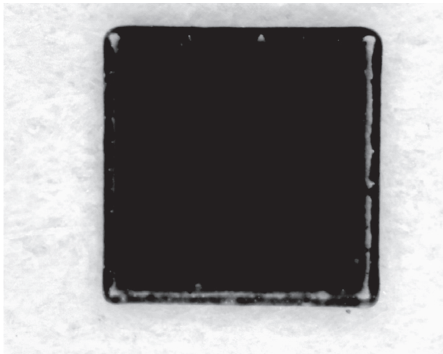
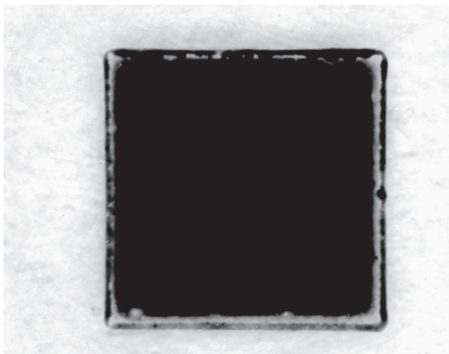


Figure 8. Lines of width 100 μm printed with elastomeric printing plate and direction 0°, 90°, 135° with respect to printing direction (mag.: x230)

(a)



b)



(c)

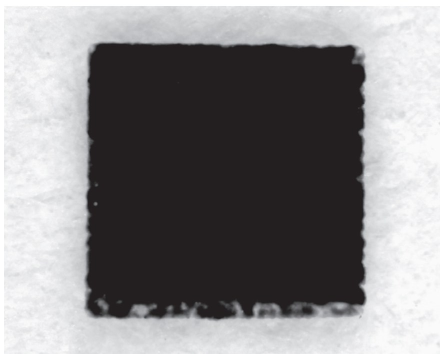


Figure 9. Solid tone patches printed with photopolymer I (a), photopolymer II (b), elastomeric printing plate (c), (mag.: x50)

edges' shape. In case of polymer plates, the lines, which are parallel to the printing direction (0°) are uniformly inked and have sharp, even edges. Similar situations is when lines are positioned at the angle of 135° to printing direction, but some slight waviness and disturbances, especially on the back edge can be noticed here (fig. 7). The edges of lines, which are perpendicular to the printing direction are curvy and disturbances on the back edge are more visible. Some pinholes can also be seen.

The lines printed with elastomeric plate, up to 100 µm wide, have blurred and unsharp edges and nonuniform structure of the ink film, regardless of the direction (fig. 8).

In the large areas covered with ink, i.e. solid tone patches and lines of widths equal to or larger than 150 µm, ink film thickness is uniform. However, around those elements, there are fringes (similar to photo frame) seen, which are called *halo effect* (fig. 9). It is inevitable phenomenon and is more intensive when pressures during printing are not optimized and the printing form is excessively squeezed. In the experiment halo effect did not occur around the thin lines, what suggests that engagement of cylinders in printing unit were within the tolerance. However, appearance of halo effect should be taken into account when designing the electronic paths to prevent short circuits in the devices. What is more influence of halo effect on printed elements conductivity should be investigated.

Conclusions

Experiment showed that quality of lines, i.e. sharpness of the edges and uniformity of the ink film depends on several parameters, which must be taken under consideration, when flexography is going to be implemented to print conductive paths without sintering. Technology in which printing plate is made are of great importance. Elastomeric printing plate does not give satisfying results from the printed electronics point of view. It is too porous to reproduce lines thinner than 150 µm, which have nonuniform surface and unsharp edges. Whereas photopolymer plates are the promising ones. Flat top dot technology lets reproduce slightly thinner (40 µm) lines than the round top dot one (50 µm), but the difference takes place only for lines at 0° to the printing direction.

It was shown that direction of lines in flexography seriously influence on quality of reproduction. Most critical is the angle 90° with respect to the printing direction, for which correctly reproduced lines were at least two times wider than those parallel to printing direction. Hence this aspect has to be taken under consideration when printed electronic components are designed. Results of experiment showed that only lines wider than 100 µm are of fine quality in three main directions simultaneously when printed with photopolymeric printing plates. Below this width in directions different than 0°, artefacts such as waviness of the edges, slight disturbances of ink film thickness near the back edge and pinholes in the lines are noticeable. For these reasons parameters of photopolymer printing forms preparation process should be optimized to improve narrow lines reproduction in flexography.

Around the lines of widths exceeding 150 µm and solid tone patches the halo effect is noticeable. It cause that electronic paths cannot be closer to each other than twice halo effect's widths, because it will result in short circuits in the device. Thus the halo effect must be taken under consideration during designing of the device.

Presented preliminary results show the need of verification of influence of ink unevenness, waviness of the edges, pinholes and halo effect on electric properties of paths and determine the statistical uncertainty which is provided by printing process. During this experiment it could not be done, because of chosen conductive ink. Observations made during experiment suggest that ink's rheological properties were not optimized by manufacturer for flexographic printing technique. Printouts made with that ink were of poor quality and made electrical measurements impossible.

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