



## Passivation of organic light emitting diode anode grid lines by pulsed Joule heating

M. Janka, R. Gierth, J.-E. Rubingh, M. Abendroth, M. Eggert, D. J. D. Moet, and D. Lupo

Citation: Applied Physics Letters **107**, 103304 (2015); doi: 10.1063/1.4930883 View online: http://dx.doi.org/10.1063/1.4930883 View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/107/10?ver=pdfcov Published by the AIP Publishing

## Articles you may be interested in

Elucidation on Joule heating and its consequences on the performance of organic light emitting diodes J. Appl. Phys. **115**, 034518 (2014); 10.1063/1.4861412

High-quality thin-film passivation by catalyzer-enhanced chemical vapor deposition for organic light-emitting diodes Appl. Phys. Lett. **90**, 013502 (2007); 10.1063/1.2425021

Organic light-emitting diode driven by organic thin film transistor on plastic substrates J. Appl. Phys. **99**, 064506 (2006); 10.1063/1.2184430

Electrical conduction in light-emitting organic polymer Schottky diodes J. Appl. Phys. **98**, 124504 (2005); 10.1063/1.2143117

Passivation of organic light-emitting diodes with aluminum oxide thin films grown by plasma-enhanced atomic layer deposition Appl. Phys. Lett. **85**, 4896 (2004); 10.1063/1.1826238



**APL Photonics** is pleased to announce **Benjamin Eggleton** as its Editor-in-Chief





## Passivation of organic light emitting diode anode grid lines by pulsed Joule heating

M. Janka,<sup>1,a)</sup> R. Gierth,<sup>2</sup> J.-E. Rubingh,<sup>3</sup> M. Abendroth,<sup>4</sup> M. Eggert,<sup>4</sup> D. J. D. Moet,<sup>3</sup> and D. Lupo<sup>1</sup>

<sup>1</sup>Department of Electronics and Communications Engineering, Tampere University of Technology, P.O. Box 692, FI-33101 Tampere, Finland

<sup>2</sup>Philips GmbH Innovative Technologies, Research Laboratories, Philipsstrasse 8, D 52068 Aachen, Germany
<sup>3</sup>TNO/Holst Centre, High Tech Campus 31, 5656 AE Eindhoven, The Netherlands
<sup>4</sup>ELANTAS Beck GmbH, Grossmannstrasse 105, 20539 Hamburg, Germany

(Received 15 July 2015; accepted 1 September 2015; published online 11 September 2015)

We report the self-aligned passivation of a current distribution grid for an organic light emitting diode (OLED) anode using a pulsed Joule heating method to align the passivation layer accurately on the metal grid. This method involves passing an electric current through the grid to cure a polymer dielectric. Uncured polymer is then rinsed away, leaving a patterned dielectric layer that conforms to the shape of the grid lines. To enhance the accuracy of the alignment, heat conduction into the substrate and the transparent electrode is limited by using short current pulses instead of a constant current. Excellent alignment accuracy of the dielectric layer on printed metal grid lines has been achieved, with a typical 4- $\mu$ m dielectric overhang. In addition to good accuracy, pulsed Joule heating significantly cuts down process time and energy consumption compared to heating with a constant current. The feasibility of using a printed current distribution grid and Joule heating was demonstrated in an OLED device. © 2015 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution 3.0 Unported License. [http://dx.doi.org/10.1063/1.4930883]

In recent years, organic light emitting diode (OLED) technology has developed rapidly, enabling thin, flexible, and lightweight large-area light sources or displays. The resistivity of the transparent electrode limits the size of the OLED panel, because lighting applications require homogeneous luminance over a large area.<sup>1</sup> A thick and narrow metal grid is typically integrated with the transparent electrode to decrease the electrode's resistivity.<sup>2</sup> To prevent a short circuit between the cathode and the anode of the device, thick metal lines must be passivated with an insulator. Since the grid lines and the passivation layer decrease the active area of the device, it is essential to align the passivation layer accurately onto the grid lines to minimize loss in active area.

Printing and photolithography have been used to pattern the passivation layer. Printing methods are cost-effective and involve the fewest process stages of all passivation methods. They have the drawback, however, of relatively poor layer-tolayer registration accuracies,<sup>3–5</sup> which necessitates a significant overhang over the edges of the grid line to ensure full coverage. Therefore, a large active area is lost if the passivation layer is printed. In contrast, the layer-to-layer registration accuracy reported for roll-to-roll photolithography is 1  $\mu$ m.<sup>6</sup> However, photolithography is a relatively expensive process and requires an additional alignment step, which is challenging if high throughput production is intended. Furthermore, if the target line is not well defined, which is the case with printed lines, a large overhang is required to fully cover the grid line.

To create an accurately aligned passivation layer by low cost methods, we have studied self-alignment passivation based on Joule heating. In this method, an electric current is passed through the grid lines to heat them selectively. The heat generated cross-links a polymer insulator near the lines and uncured polymer is rinsed away after curing. As a result, insulator patterns are aligned according to the grid lines.<sup>7</sup> Heat conduction in the substrate defines the size of the pattern and thus limits the registration accuracy of the method.<sup>8</sup>

We report here a passivation process based on Joule heating. Instead of using a constant current for heating, as in our previous studies,<sup>7–9</sup> we used a pulsed current. The length of the pulse was selected not only to guarantee heating of the metal grid lines but also to minimize heat conduction into the substrate. This procedure is analogous to pulsed laser ablation, in which the size of the ablated area is limited using very short pulses. The typical pulse length is significantly longer than in laser ablation, on the order of milliseconds. Frequencies of this magnitude are not difficult to generate even in large volume production.

We present here a finite element simulation of the dependence of heating selectivity on pulse length and the experimental validation of the simulation. Finally, we demonstrate the feasibility of the process with excellent alignment accuracy of a passivated OLED anode grid on a glass substrate coated with Indium-tin oxide (ITO). The current distribution grids were printed to demonstrate the possibility of low-cost OLED manufacturing.

The registration accuracy of the Joule heating process depends on the spatial selectivity of the heating. Thus, by limiting the transfer of heat into the substrate through the use of short current pulses, we can improve the registration accuracy. The effect of pulsed heating on heating selectivity was studied by both model and experiment.

0003-6951/2015/107(10)/103304/4

107, 103304-1

© Author(s) 2015

<sup>&</sup>lt;sup>a)</sup>Electronic mail: marika.janka@tut.fi



FIG. 1. Modelled effect of pulse length on the temperature gradient at the edge of the grid line at 390 K and 580 K in the conductor. Inset: Temperature profile from the center of the line to the substrate. The gray bar denotes the location of the grid line and the black line denotes the location where the temperature gradient is calculated.

The effect of the pulse length on heating selectivity was studied with a time-dependent thermo-electric model for a 500-nm thick silver line on polyethylene terephthalate (PET) substrate. The temperature gradient at the edge of the grid line was chosen as proxy for heat selectivity (results in Figure 1 with the temperature gradient and the profile from the center of the line to the substrate shown in the inset). The temperature gradient increases as the pulse length decreases. To have an effect on the localization, the heat pulse should be less than 0.1 s. Furthermore, an increase in the temperature increases the temperature gradient. Moreover, the higher the temperature, the more reducing pulse length increases the temperature gradient.

To verify the model data experimentally, we designed a silver line set with a current choke in the middle of the line (see inset in Figure 2) and evaporated it on a PET substrate. In the choke, the current density increases and induces a local rise in temperature. At a choke, higher temperature increases the overhang, since there is no corresponding decrease of curing time. Thus, the size of the dielectric overhang ( $x_{OH}$ ) increases as the difference in line width ( $\Delta W$ ) increases. The increase depends on the heating selectivity in that the better the selectivity, the less the overhang increase. (See supplementary material for more detailed explanation.<sup>10</sup>) The experiments were run with 10-ms, 50-ms, 100-ms pulses and 1-min DC heating, and the increase in the dielectric overhang ( $\Delta x_{OH}$ ) due to the choke was measured.



FIG. 2. The dielectric overhang  $(x_{OH})$  increases when line width at the choke  $(W - \Delta W)$  decreases. As pulse length decreases, the size of the overhang is less prone to changes in line width.

As expected, the overhang increased when the difference in linewidth increased. When pulse length was decreased, the slope decreased, indicating a higher temperature gradient at the edge of the line. However, with very short pulse lengths, and corresponding higher peak power, the PET substrate melts at the narrow choke widths. We had fewer measurement points with the pulsed than with DC current, because the deformation of the substrate limited the size of the choke usable for experiments. For the same reason, the 50-ms and 10-ms experiments were done with 10 and 30 pulses, respectively, to be able to use lower temperatures and to avoid melting of the substrate, whereas with 100 ms only one pulse was used.

According to the simulation, increase in the temperature gradient should not be dramatic if pulse length is decreased from DC to 100 ms, whereas in the experimental data the selectivity increases significantly. This discrepancy results from the temperature induced at the line during Joule heating. With shorter heating, higher temperatures could be used without melting the substrate. As seen in Figure 1, the temperature gradient increases also as a function of temperature.

The optimal pulse length depends on the substrate used and the homogeneity of the target lines cross-sectional area. The better the thermal stability of the substrate, the more variation is allowed in the line cross-section without damage; furthermore, a shorter pulse length can be used. We emphasize here that the results in Figure 2 show a more dramatic increase in the overhang, and, moreover, the substrate melts with smaller differences in line width than expected in the passivation process. This is because the temperature in the choke increases as the length of it increases up to saturation. In the experiments, the length of the choke was chosen as 1 mm, which corresponds to the saturation length in constant current heating. Typically, the grid line cross-sectional area varies in length in tens of micrometers rather than in millimeters.

In addition to increased registration accuracy, pulse heating decreases energy consumption during curing. Table I shows the curing parameters and the energy consumed during curing for each Joule heating series. The pulse heating setup decreases the heat dissipated and lost in the substrate and cross-linking takes place faster at higher temperature, thus less energy was needed to cure the dielectric. With DC heating, energy consumption is three orders of magnitude greater than with pulsed heating. The required current increases when the pulse length decreases, but the pulse length and the total heating time are more markedly reduced. The lowest power consumption was achieved with a 100-ms pulse. Optimization of the pulse energy depends on the thermal stability of the substrate and the dielectric curing

TABLE I. Energy consumed during curing for all pulse lengths.

Sample	Current (mA)	Number of pulses	Total energy (J)
DC	320	1	27
100 ms	550	1	0.14
50 ms	550	10	0.68
10 ms	700	30	0.66

This article is copyrighted as indicated in the article. Reuse of AIP content is subject to the terms at: http://scitation.aip.org/termsconditions. Downloaded to IP: 139.63.9.10 On: Tue. 29 Sep 2015 13:07:37 properties. In these experiments, the sample posed the limit for the maximum power level.

For a constant current Joule heating setup, glass is a challenging substrate material; accurately aligned dielectric patterns are difficult to produce due to its high thermal conductivity.<sup>9</sup> Pulse heating enables the use of substrate materials incompatible with DC heating, because it increases the registration accuracy. In addition to increased registration accuracy, a further advantage of pulse heating is that the curing time necessary for the polymer to cross-link decreases as the temperature increases. Cutting the process time from 1 min to 100 ms is significant and increases the attractiveness of the Joule heating approach as a method for high volume manufacturing.

In order to demonstrate the practical applicability of the pulsed Joule heating approach, it was used to passivate an OLED anode. Glass was selected as the substrate material, because it has better water barrier properties than PET. The anode electrode in the experiments was ITO with a sheet resistance of  $10 \Omega/\Box$ . The grid lines were 3- $\mu$ m thick and approximately 90- $\mu$ m wide printed silver lines. The separation of grid lines was 2 mm. A dielectric (Bectron AL14-018H from ELANTAS Beck) was cured using six 75-ms, 13-A pulse. A microscope image of a passivated grid line is shown in Figure 3(a). The registration accuracy of the process was excellent; the dielectric overhang was less than 5  $\mu$ m, although linewidth varied from 80  $\mu$ m to 100  $\mu$ m.



FIG. 3. (a) Microscope image of an evaporated grid on an ITO coated glass substrate passivated using a pulsed Joule heating setup. (b) Profile of the printed line with and without passivation layer. (c) Magnification of the edge of the line profile. Note that the linewidth of the printed line is not constant and the images are not taken from the same location of the line.

Figures 3(b) and 3(c) show the height profiles of a printed line with and without passivation layer, measured with a stylus profilometer (Veeco Dektak 150 Surface Profiler). The passivation layer smoothens the surface of the printed line resulting in a RMS roughness value of 5 nm on the line. The passivation layer is relatively thick, about 13  $\mu$ m, though the size of the overhang is only few microns.

The characteristic IV curves for an OLED reference and an OLED with grid lines passivated with the method described above are given in Figure 3, where the inset is an image of the OLED with current distribution grid. Electrical losses in this non-light-emitting voltage range are an indication of irregularities in the electrical field between anode and cathode. Contributions to these loss channels can be found in a rough surface topology<sup>11</sup> or the migration of silver ions. These effects reduce the reliability of the device. The nonemitting voltage range between 0.5 and 2 V is highlighted by dashed lines. The leakage current in this range is of a comparable order of magnitude for devices with passivated grid lines and for the reference device without grid lines. Therefore, it can be concluded that the surface topology of the substrate allows for reliable operation of OLED devices and that the dielectric passivation effectively covers the Ag lines. Water vapor residues outgassing from the passivation lines deteriorated the OLED stack in the vicinity of the grid lines (closer than  $100 \,\mu\text{m}$ ) in the form of blurred blackish artifacts, as can be seen in the inset of Figure 4. To remove this water contamination, another, hydrophobic, polymer should be used.

The size of the OLED active area was relatively small compared with the high conductivity of ITO. Thus, more significant improvement in IV characteristics of the device with grid could be shown with devices having less conductive transparent electrode, larger active area, and optimized line pitch.<sup>12–14</sup>

In this study, we introduced a pulsed-Joule-heatingbased passivation method for OLED anode grid lines. Instead of a constant current for heating, as in our previous studies,<sup>7–9</sup> we used a pulsed current to limit heat conduction into the substrate and polymer layer. We have shown both theoretically and experimentally an increase in heating



FIG. 4. IV-curves of a reference OLED and an OLED with current distribution grid (size of the active area 900 mm<sup>2</sup>). Inset: Photograph of an OLED with current distribution grid under operation.

his article is copyrighted as indicated in the article. Reuse of AIP content is subject to the terms at: http://scitation.aip.org/termsconditions. Downloaded to IP: 139.63.9.100

selectivity as pulse length decreases. Heat conduction defines the selectivity of the heating, which is the key parameter for the registration accuracy of the process: the less heat is conducted into the substrate, the better the registration accuracy. Our method extends the selection of substrate material compatibility with Joule heating from plastic to thermally more conductive glass substrates. The registration accuracy we achieved on an ITO-coated glass substrate was better than 5  $\mu$ m, which is comparable with registration accuracy of rollto-roll photolithography for well-defined target lines. Furthermore, variations in line width had only a slight effect on the accuracy. In addition, pulse heating helps reduce process time and energy consumption significantly over constant current heating. In conclusion, the scalability of the method for high volume manufacturing improves in the following parameters: process time, registration accuracy, and material selection. We demonstrated the compatibility of the concept for evaporated OLED stacks.

The study was funded through the European Union seventh Framework Program (FP7-ICT-2012, Project No. 314362). M. Janka would like to thank Nokia Foundation for their support of this study. We thank Professor K. Palovuori for his technical support and for designing the current pulsing circuit, T. Vuorinen for writing the Arduino control program, and J. Kontio for the profilometer measurements.

- <sup>1</sup>K. Neyts, M. Marescaux, A. U. Nieto, A. Elschner, W. Lovenich, K. Fehse, Q. Huang, K. Walzer, and K. Leo, J. Appl. Phys. **100**, 114513 (2006).
- <sup>2</sup>M. Slawinski, M. Weingarten, M. Heuken, A. Vescan, and H. Kalisch, Org. Electron. **14**, 2387 (2013).
- <sup>3</sup>M. Guerin, A. Daami, S. Jacob, E. Bergeret, E. Benevent, P. Pannier, and R. Coppard, IEEE Trans. Electron Devices **58**, 3587 (2011).
- <sup>4</sup>J. Noh, D. Yeom, C. Lim, H. Cha, J. Han, J. Kim, Y. Park, V. Subramanian, and G. Cho, IEEE Trans. Electron. Packaging Manuf. 33, 275 (2010).
- <sup>5</sup>T. N. Ng, D. E. Schwartz, L. L. Lavery, G. L. Whiting, B. Russo, B. Krusor, J. Veres, P. Bröms, L. Herlogsson, N. Alam *et al.*, Sci. Rep. 2, 585 (2012).
- <sup>6</sup>H. Zhang, M. D. Poliks, and B. Sammakia, J. Disp. Technol. **6**, 571 (2010).
- <sup>7</sup>M. Janka, S. Tuukkanen, T. Joutsenoja, and D. Lupo, Thin Solid Films **519**, 6587 (2011).
- <sup>8</sup>M. Janka, E. Saukko, P. Raumonen, and D. Lupo, Org. Electron. **15**, 3431 (2014).
- <sup>9</sup>M. Janka, P. Raumonen, S. Tuukkanen, and D. Lupo, in *Symposium M Large-Area Processing and Patterning for Active Optical and Electronic Devices* (Mater. Res. Soc. Proc., 2014), Vol. 1628, pp. mrsf13–1628.
- <sup>10</sup>See supplementary material at http://dx.doi.org/10.1063/1.4930883 for details about sensitivity of dielectric overhang on changes in temperature and experimental details.
- <sup>11</sup>B. D'Andrade and S. Forrest, Adv. Mater. 16, 1585 (2004).
- <sup>12</sup>M. Barink and S. Harkema, J. Appl. Phys. **112**, 054507 (2012).
- <sup>13</sup>S. Harkema, S. Mennema, M. Barink, H. Rooms, J. S. Wilson, T. van Mol, and D. Bollen, "Large area ITO-free flexible white OLEDs with Orgacon PEDOT:PSS and printed metal shunting lines," Proc. SPIE **7415**, 74150T (2009).
- <sup>14</sup>H. J. van de Wiel, Y. Galagan, T. J. van Lammeren, J. F. J. de Riet, J. Gilot, M. G. M. Nagelkerke, R. H. C. A. T. Lelieveld, S. Shanmugam, A. Pagudala, D. Hui, and W. A. Groen, Nanotechnology 24, 484014 (2013).