Hybrid Color-Tunable Polymer Light-Emitting Diodes Using Electrospraying

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ABSTRACT: This paper proposes a new paradigm in polymer light-emitting diode (PLED) fabrication by using a uniform electrosprayed microparticle film as the active layer. Among the seven electrospraying parameters analyzed, three crucial parameters are statistically identified and optimized to obtain thin electrosprayed microparticle layers. Using optimized electrospraying conditions, single-color red-emitting PLED (MEH-PPV) with a peak current density of 16.1 mA/mm² under a 13.5 V bias and a peak external quantum efficiency of 3.2% are successfully fabricated. Finally, a combinatorial approach is implemented using both MEH-PPV (red-emitting) and F8BT (green-emitting) polymer microparticles at different mixing ratios to tune the emission spectrum of the devices. As such, it has been demonstrated that hybrid multilayer films using different organic materials with nonorthogonal solvents can be produced using this new approach. The parameter analysis and color-tunable properties pave the way towards white light PLED fabrication.

INTRODUCTION

Polymer light-emitting diodes (PLEDs) have generated a widespread interest in both academic and industrial research towards new generations of low-cost solid-state lighting devices and flat panel displays, mainly because of their unique advantages including flexibility, large-area scalability, and low-cost solution-based fabrication. Except for spin-coating, liquid fabrication methods for polymeric materials generally offer the advantages of low-cost and low-temperature processing with high material yield. As a result, multiple light-emitting polymer systems were proposed since the first reports on the poly-phenylenevinylene (PPV)-based PLED nearly three decades ago. More recently, light-emitting polymer systems including the poly[2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene] or MEH-PPV, the poly(9,9-diocetylfluorene-alt-benzothiadiazole) or F8BT, and the poly[(9,9-diocetylfluorene) or PFO acting as efficient red-, green-, and blue-emitting polymers, respectively, are developed, and full color displays have also been realized by combining red-, green-, and blue-emitting polymers. However, a multilayer architecture is usually necessary to fulfill the PLED’s potential for high thermal-stability, long operation lifetime, and high efficiency. Typically, such devices combine a hole-injection layer, a hole-transporting layer, an emissive layer, and an electron-transporting layer. Most unfortunately, the similar solubility of conjugated polymers in common organic solvents generates great challenges when fabricating these multilayer PLED devices. As a result, the underlying film is often partially dissolved and blended with the newly deposited material during multilayer deposition yielding lower performances for both materials. While some multilayer PLED architectures have been successfully fabricated using a liquid buffer layer, the number of layers remains greatly limited because very few solvents can be used orthogonally to dissolve common PLED materials. Since the first reports over a century ago, electrospraying processes are now ubiquitous for the deposition of polymers, semi-conductive ceramics, and biological materials. It relies on liquid atomization using electrostatic forces to break the surface tension when a solution flows through a needle under high electrical potential. In the process of reaching the electrically grounded collector, fast solvent evaporation results in the collection of particles. Compared to other techniques, electrospraying has the advantage of a very simple architecture setup with low material consumption for a wide selection of precursor viscosities. As a result, the use of electrospraying can lead to the formation of nano- to sub-micrometer polymeric structures offering limitless possibilities for new materials and technologies targeting micro-electronics and medical devices. Additionally, electrospraying has been recently applied to fabricating organic photovoltaic devices and organic light-emitting diode devices.
In this work, we exploit electrospraying techniques in order to achieve facile one-step PLED device fabrication using a uniform layer of conjugated polymer microparticles as the active region. To do so, the morphology of the microparticle layer is studied while varying several process parameters including flow rate, applied voltage, the distance between the tip of the needle and collector, the gauge of the needle, ambient temperature, deposition time, and concentration of the polymer solution. Subsequently, a statistical design of experiment approach reveals the key parameters affecting the thickness of the microparticle layer in order to allow precise control and optimization. These optimized parameters are first used to achieve a red-emitting PLED device using a uniform film of MEH-PPV microdroplets. Finally, a combinatorial approach using two different conjugated polymer microparticles (MEH-PPV and F8BT) at different ratios demonstrates the ability to combine materials and tune the emission properties of the PLED devices while maintaining the simplicity of the fabrication process.

**RESULTS AND DISCUSSIONS**

Among many different electrospraying modes, cone-jet mode is the most important and widely used since it provides the best stability and reproducibility during the electrospraying process.\(^25\) Because of the relatively low carrier mobility in conjugated polymers, the thickness of the electrosprayed microparticle layer is critical for the input power of the PLED devices.\(^7\) The particle size can be affected by many exogenous variables, and the key process parameters can be identified through a randomized control experiment. In Figure 1a, the average particle size in the film is ranging from 5.4 to 7.4 μm in the y-axis, and the values for each process parameter are shown in Table 1. The low and high values are selected based on the observation of cone-jet mode. Figure 1a shows a graphic representation of the interrelation between the key experimental parameters and the size of the electrosprayed F8BT microparticles, where long lines indicate the corresponding parameters with the greater impacts, while the shorter lines indicate otherwise. In addition, the sign of the slope indicates if the average particle size is directly or inversely proportional to the given parameter variation. It suggests that the distance between the needle and the collector, the needle gauge, and the polymer solution concentration are the key parameters with the greatest impact on the average particle size. The distance between the needle and the collector and applied voltage have a tight interwoven relationship to the electrical field, a crucial parameter in electrospraying. At a given voltage, a smaller distance has the advantage of high electrical field generation and smaller particles collection; however, aggregation of wet particles on the collector may happen because of insufficient time for solvent evaporation. On the contrary, a larger distance would require higher voltage applied and cause the lost electrosprayed materials to the surroundings. Because the selected experimental low and high values with the cone-jet mode, a 50% difference in distance (10 and 15 cm) is larger than 20% difference in the voltage supplied (8 and 10 kV) results in a greater impact of the distance comparing to applied voltage. The gauge of needle controls the size of the jetting cone under cone-jet mode. A smaller particle collection would prefer a larger needle gauge and a smaller diameter of the needle. Solution concentration controls the quantity of the polymer material itself and affects the solution’s electrical conductivity and viscosity. Because all the electrosprayed polymers are semi-conductive, the concentration selected has little impact on the conductivity. The polymeric solution also shows no significant viscosity changes with selected values. Therefore, a smaller concentration is preferred to collect smaller particles because of less polymer quantity. Figure 1b shows a typical 3D scan used to assess the distribution of the average particle size with a 3D measuring laser microscope (Olympus LEXT—model OLS4100). The dimension of the sample area is 259 μm × 259 μm, and the color bar on the right shows the scale for the height of the particles. The red color indicates the maximum height of 6.9 μm, and purple indicates the baseline. The thickness of the microparticle layer is considered as the average value of the heights of all the microparticles where the height is measured individually using Digimizer, a graphic-analysis freeware.\(^37\) As a result, the values for three main parameters are obtained (concentration of 4 mg/mL, distance of 4 cm, and gauge of needle of 25) for a
minimum of 2.3 μm uniform particle collection as it is shown in Figure S3. More details about the design of the experiment including the degree of impact for combinatorial parameters, parameter selections, and the layer thickness analysis for the electrosprayed microparticles are provided in the Supporting Information Section.

Using this process, red-emitting PLEDs using a layer of MEH-PPV microparticles as the active layer are prepared and tested following protocols described in the Methods Section. Figure 2a shows a schematic representation of the red light-emitting PLED architecture and Figure 2b shows its energy band diagram. Figure 3 shows the device characteristics. There, Figure 3a shows the normalized electroluminescence with a peak emission at 600 nm. The inset of Figure 3a shows a macroscopic picture of the PLED under operation. Current density and voltage characteristics are shown in Figure 3b. The peak current density is 16.1 mA/mm² under 13.5 V applied bias. As seen in Figure 3c, the PLED also reaches its maximum luminance of 475 cd/m² at 13.5 V. Finally, Figure 3d shows the external quantum efficiency (EQE) evolution of the device during operation with a peak EQE at 3.2%.

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This deposition approach can also potentially combine different conjugated polymer materials, without requiring the use of orthogonal solvents. After the successful fabrication of the red-emitting PLEDs by electrospraying, we chose to explore the combinatorial deposition of both MEH-PPV (red-emitting) and F8BT (green-emitting) polymer microparticles on the same substrate. Here, we show that emission spectrum of the device can then be controlled simply by adjusting the ratio of the polymer droplets density through deposition time. Figure 4a shows a schematic representation of the fabrication setup for the color-mixed PLEDs. Syringe pumps are used to precisely control the solution flow rate, and the distance is the vertical separation between needles and the sample substrate. All the parameters can be controlled independently for both depositions. Here, only the deposition times is changed to control the relative ratio of the MEH-PPV and F8BT polymer droplets.

To show the level of control this approach provides, four additional sets of devices are fabricated. Their active layer relative compositions are F8BT-100% (G/R = 1:0), F8BT-70% with MEH-PPV-30% (G/R = 7:3), F8BT-50% with MEH-PPV-50% (G/R = 1:1), and F8BT-30% with MEH-PPV-70% (G/R = 3:7). Their normalized electroluminescence spectra are compared in Figure 4b, together with the previously fabricated MEH-PPV device (G/R = 0:1). The emission peak for each PLED changes from green to red at 525, 535, 558, 580, and 601 nm. Their emission full-width at half-maximum is 65, 86, 73, 59, and 54 nm. The right shoulder at ∼565 nm for the G/R = 7:3 device and the left shoulder at ∼540 nm for the G/R = 3:7 device matches the emissions from the unequal F8BT and MEH-PPV microparticle ratio. The inset of Figure 4b shows an orange PLED (G/R = 3:7) under operation. The picture is focused on the light-emitting particles instead of a macroscopic point of view. It is a representation of the mechanism of spectrum shifting of electrosprayed light emitting polymer particles blends. For samples G/R = 7:3 (light green) and G/R = 3:7 (orange), green and red light-emitting polymeric particles dominate the PLED emission, respectively. As it is for yellow light-emitting PLED, green and red light-emitting polymeric particles contribute equally to the PLED emission. Figure 4c shows the current density and voltage characteristics for the five different PLED devices. From the plot, we can observe that the PLED devices with mixed microparticles require higher applied voltage to reach
comparable currents. It is likely due to electric field interactions between the two needles causing lower micro-particle film uniformity compared to using only one needle. Figure 4d shows that green-emitting (using pure F8BT) PLEDs have the best power efficiency and the power efficiency decreases when mixed with MEH-PPV droplets. The power efficiency of the red-emitting (using pure MEH-PPV) is also higher than that of mixed PLEDs as they require lower applied voltage to reach similar performance comparing as previously observed. The standardized Commission Internationale de l’Eclairage (CIE) diagram is generated using the CIE generation software,46 and all generated CIE points are shown in Figure 5a. The CIE coordinates for all five PLEDs are (0.26, 0.66) for G/R = 1:0, (0.35, 0.60) for G/R = 7:3, (0.41, 0.57) for G/R = 1:1, (0.46, 0.50) for G/R = 3:7, and (0.58, 0.40) for G/R = 0:1. A gray line is used to connect G/R = 1:0 and G/R = 0:1; the points fall gradually onto the line when the MEH-PPV composition ratio of the PLED increases starting from G/R = 1:0. Figure 5b is the combined images of all five PLEDs during operation. The emission change from green to red as the relative ratio between MEH-PPV and F8BT polymer droplets increases is easily distinguished. The different shapes of the PLEDs, such as round and square, are due to different types of shadow masks used for these two different stages of the experiments.

### CONCLUSIONS

In summary, we successfully demonstrate that carefully optimized electrospaying can achieve facile PLED fabrication. We first conduct a complete series of experiments followed by statistical analysis to identify the main electrospaying parameters. Then, an ideal set of parameters are obtained to electrospay a thin and uniform conjugated polymer micro-particle layer which is suitable as the active layer of the PLED. After the successful fabrication of a red-emitting PLED using MEH-PPV microparticles, we demonstrate the ability to combine different polymers and tune the device emission using mixed layers of MEH-PPV (red-emitting) and F8BT (green-emitting) microparticles. Even though the uniformity of the mixed polymer droplets based layers are relatively difficult to control and the overall performance of these PLED devices is still modest, we believe that this approach presents a new paradigm for easy fabrication of hybrid multicomponent-conjugated polymer-based optoelectronic devices with controllable properties. Therefore, we believe that it could potentially bring a great change and a brighter future for low-cost PLED devices.

### METHODS

The ITO substrates (Kintec Company, 15 Ω/sq) are ultrasonically cleaned in sequence using acetone, ethanol, and isopropanol for 10 min each. PEDOT/PSS (CleviosTM P VP AI 4083) is diluted with isopropanol (1:3 volume ratio) and spin-coated atop the ITO substrate at 3000 rpm for 60 s to achieve a uniform layer, followed by a 20 min annealing at 200 °C in a nitrogen-filled oven. Next, a hole-transporting-layer of TFB (American Dye Source, Inc.) solution is prepared by dissolving the polymer in tetrahydrofuran (THF) solvent (at 4 mg/mL weight concentration) and deposited by spin-coating atop the PEDOT/PSS film at 3000 rpm.

For the red-emitting PLEDs using only MEH-PPV (from American Dye Source, Inc.), the polymer is dissolved in THF and the solution is electrospayed on top of the spin-coated TFB film to form a uniform microparticle layer with a thickness of ∼2 μm using the following optimized parameters: solution of MEH-PPV with a concentration of 4 mg/mL, a 6 cm distance between the needle and the substrate, a needle gauge 25, a 10.5 kV applied voltage, a 2 mL/h flow rate in the syringe pump, and a 2 min deposition time. Finally, a 120 nm thick Al cathode is deposited on top of the electrospayed particle layer through a shadow mask using thermal evaporation at a rate of 0.3 Å/s. The dimensions of the devices are 2 mm by 2 mm.

For the other four different light-emitting PLEDs, all the fabrication steps and parameters remain the same as for the MEH-PPV (G/R=0:1) red-emitting devices, except the active layers deposition time. For the G/R = 1:0, G/R = 7:3, G/R = 1:1, and G/R = 3:7 PLED devices, the ratio between both polymers is controlled by varying only the deposition time. For example, a deposition sequence of 42 s (green), then-18 s (red), then-42 s (green), and finally-18 s (red) is used for the G/R = 7:3 device. In contrast, the alternating sequence 30 s (green)—30 s (red)—30 s (green)—30 s (red) is used for the G/R = 1:1 device, compared with a 18 s (green)—42 s (red)—18 s (green)—42 s (red) sequence used for the G/R = 3:7 and 120 s (green) for the G/R = 1:0 device.

The sample surface is analyzed using an Olympus LEXT 3D microscope (OLS4100). Steady-state electroluminescence spectra are recorded using a Jobin-Yvon iHE320 triple-grating spectrometer equipped with a Synapse silicon CCD array. The PLED light-emitting power is measured using a photodetector (Thorlabs, S120VC, 200–1100 nm, 50 mW) connected through a power and energy meter module (Thorlabs, model PM100USB). To verify the calibration, a commercial LED module with known output power is used to obtain the attenuation factor before the PLEDs emission power measurements.

### ASSOCIATED CONTENT

5 Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsomega.9b02666.

Experimental design for electrospayed micro-particles, standardized effect on particle size by each parameter
and their combination, and estimated response surface mesh for the height of electrosprayed microparticle layer (PDF)

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Author Contributions

X.G. and J.B.-G. designed the project. X.G. tested and analyzed the data of the PLED device. X.G. and D.B interpreted the results. X.G. and J.B.-G. designed the project. X.G. tested and analyzed the electrosprayed micro-particle data. S.G.C. corrected the manuscript and guided the entire project. All the authors reviewed, edited, and approved the manuscript.

Notes

The authors declare no competing financial interest.

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REFERENCES

[1] Dai, X.; Zhang, Z.; Jin, Y.; Niu, Y.; Cao, H.; Liang, X.; Chen, L.; Wang, J.; Peng, X. Solution-Processed, High-Performance Light-Emitting Diodes Based on Quantum Dots. Nature 2014, 515, 96–99.

[2] Wei, Q.; Ge, Z.; Voit, B. Thermally Activated Delayed Fluorescent Polymers: Structures, Properties, and Applications in OLED Devices. Macromol. Rapid Commun. 2019, 40, 1800570.

[3] Ju, J.; Yamagata, Y.; Higuchi, T. Thin-Film Fabrication Method for Organic Light-Emitting Diodes Using Electrospray Deposition. Adv. Mater. 2009, 21, 4343–4347.

[4] Burroughes, J. H.; Bradley, D. D. C.; Brown, A. R.; Marks, R. N.; Mackay, K.; Friend, R. H.; Burns, P. L.; Holmes, A. B. Light-Emitting Diodes Based on Conjugated Polymers. Nature 1990, 347, 539–541.

[5] Deng, X.; Lu, W.; Wong, K. Y.; Low, K. H.; Chow, H. F.; Cao, Y. High Efficiency Low Operating Voltage Polymer Light-Emitting Diodes with Aluminum Cathode. Appl. Phys. Lett. 2004, 84, 3522–3524.

[6] Gustafsson, G.; Cao, Y.; Teacey, G. M.; Klavetter, F.; Colaneri, N.; Heeger, A. J. Flexible Light-Emitting Diodes Made from Soluble Conducting Polymers. Nature 1992, 357, 477–479.

[7] Hewidy, D.; Gadallah, A.-S.; Fattah, G. A. Electroluminescence Enhancement of Glass/ITO/PEDOT:PSS/MEH-PPV/PEDOT:PSS/Al OLED by Thermal Annealing. J. Mol. Struct. 2017, 1130, 327–332.

[8] Hu, P.; Kim, J.; Burroughes, J. H.; Becker, H.; Li, S. F.; Brown, T. M.; Cacciari, F.; Friend, R. H. Molecular-Scale Interface Engineering for Polymer Light-Emitting Diodes. Nature 2000, 404, 481–484.

[9] Kabra, D.; Lu, L. P.; Song, M. H.; Snaith, H. J.; Friend, R. H. Efficient Single-Layer Polymer Light-Emitting Diodes. Adv. Mater. 2010, 22, 3194–3198.

[10] Lu, L. P.; Finlayson, C. E.; Friend, R. H. Thick Polymer Light-Emitting Diodes with Very High Power Efficiency Using Ohmic Charge-Injection Layers. Semicond. Sci. Technol. 2014, 29, 025005.

[11] Yan, H.; Huang, Q.; Cui, J.; Veinot, J. G. C.; Kern, M. M.; Marks, T. J. High-Brightness Blue Light-Emitting Polymer Diodes via Anode Modification Using a Self-Assembled Monolayer. Adv. Mater. 2003, 15, 835–838.

[12] ul Hasen, K.; Sandberg, M. O.; Nur, O.; Willander, M. ZnO/Polyyfluorene Hybrid LED on an Efficient Hole-Transport Layer of Graphene Oxide and Transparent Graphene Electrode. Adv. Mater. Opt. 2014, 2, 326–330.

[13] Song, H. J.; Kim, D. H.; Lee, T. H.; Moon, D. K. Emission Color Tuning of Copolymers Containing Polyfluorene, Benzo-thiadiazole, Porphyrin Derivatives. Eur. Polym. J. 2012, 48, 1485–1494.

[14] Zheng, H.; Zheng, Y.; Liu, N.; Ai, N.; Wang, Q.; Wu, S.; Zhou, J.; Hu, D.; Yu, S.; Han, S.; et al. All-Solution Processed Polymer Light-Emitting Diode Displays. Nat. Commun. 2013, 4, 1971.

[15] Deng, X.-Y. Light-Emitting Devices with Conjugated Polymers. Int. J. Mol. Sci. 2011, 12, 1575–1594.

[16] Zheng, H.; Zheng, Y.; Wang, J.; Wang, J.; Zhang, G.; Zhang, S.; Liu, M.; Hu, J.; Li, Y.; Hu, Y.; et al. Polymer Light-Emitting Displays with Printed Cathodes. Surf. Coat. Technol. 2019, 358, 228–234.

[17] Zhou, Z.-L.; Sheng, X.; Nauka, K.; Zhao, L.; Gibson, G.; Lam, S.; Yang, C. C.; Brug, J.; Elder, R. Multilayer Structured Polymer Light-Emitting Diodes with Cross-Linked Polymer Matrices. Appl. Phys. Lett. 2010, 96, 013504.

[18] Ráfols-Ribé, J.; Will, P.-A.; Hänisch, C.; Gonzalez-Silveira, M.; Lenk, S.; Rodriguez-Viejo, J.; Reineke, S. High-Performance Organic Light-Emitting Diodes Comprising Ultrastable Glass Layers. Sci. Adv. 2018, 4, eaat3332.

[19] Ma, X. Hybrid Light Emitting Diodes Based on Solution Processed Polymers, Colloidal Quantum Dots, and Colloidal Metal Nanoparticles. Ph.D. Thesis, University of Delaware, 2014.

[20] Tseng, S.-R.; Lin, S.-C.; Meng, H.-F.; Liao, H.-H.; Yeh, C.-H.; Lai, H.-C.; Horng, S.-F.; Hsu, C.-S. General Method to Solution-Process Multilayer Polymer Light-Emitting Diodes. Appl. Phys. Lett. 2016, 88, 163501.

[21] An, D.; Zou, J.; Wu, H.; Peng, J.; Yang, W.; Cao, Y. White Emission Polymer Light-Emitting Devices with Efficient Electron Injection from Alcohol/Water-Soluble Polymer/Al Bilayer Cathode. Org. Electron. 2009, 10, 299–304.

[22] Zeleny, J. The Electrical Discharge from Liquid Points, and a Hydrostatic Method of Measuring the Electric Intensity at Their Surfaces. Phys. Rev. 1914, 13, 69–91.

[23] Bodnár, E.; Grillo, J.; Rosell-Llompart, J. Polymer Solution Electrospraying: A Tool for Engineering Particles and Films with Controlled Morphology. J. Aerosol Sci. 2018, 125, 93–118.

[24] Mokhtarí, F.; Latifi, M.; Shamshirzaz, M. Electrosplining/Electrospraying of Polyvinylidine Fluoride (PVDF): Piezoelectric Nanofibers. J. Text. Inst. 2016, 107, 1037–1055.

[25] Nguyen, D. N.; Clasen, C.; Van den Mooter, G. Pharmaceutical Applications of Electrospraying. J. Pharm. Sci. 2016, 105, 2601–2620.

[26] Boda, S. K.; Li, X.; Xie, J. Electrospraying an Enabling Technology for Pharmaceutical and Biomedical Applications: A Review. J. Aerosol Sci. 2018, 125, 164–181.

[27] Jaworek, A.; Sobczyk, A. T. Electrospraying Route to Nanotechnology: An Overview. J. Electrostat. 2008, 66, 197–219.

[28] Lee, S.; Koo, H.; Cho, S. Mask-Less Patterning of Organic Light Emitting Diodes Using Electrospraying and Selective Biasing on Pixel Electrodes. Appl. Phys. Lett. 2015, 106, 173303.

[29] Pitsalidis, C.; Pappa, A. M.; Hunter, S.; Laskarakis, A.; Kaimakamis, T.; Payne, M. M.; Anthony, J. E.;Anthopoulos, T. D.; Logothetidis, S. High Mobility Transistors Based on Electrosprayed-Printed Small-Molecule/Polymer Semiconducting Blends. J. Mater. Chem. C. 2016, 4, 3499–3507.

[30] Xie, J.; Jiang, J.; Davoodi, P.; Srinivasan, M. P.; Wang, C.-H. Electrohydrodynamic Atomization: A Two-Decade Effort to Produce and Process Micro-/Nanoparticulate Materials. Chem. Eng. Sci. 2015, 125, 32–57.

[31] Zhu, W.; Masood, F.; O’Brien, J.; Zhang, L. G. Highly Aligned Nanocomposite Scaffolds by Electrosprinning and Electrospinning for Neural Tissue Regeneration. Nanomed. Nanotechnol. Bio. Med. 2015, 11, 693–704.

[32] Anjusree, G. S.; Deepak, T. G.; Nair, S. V.; Nair, A. S. Facile Fabrication of TiO2 Nanoparticle—TiO2 Nanofiber Composites by Co-Electrosprinning—Electrospinning for Dye-Sensitized Solar Cells. J. Energy Chem. 2015, 24, 762–769.
(33) Hong, S. C.; Lee, G.; Ha, K.; Yoon, J.; Ahn, N.; Cho, W.; Park, M.; Choi, M. Precise Morphology Control and Continuous Fabrication of Perovskite Solar Cells Using Droplet-Controllable Electrospray Coating System. *ACS Appl. Mater. Interfaces* 2017, 9, 7879−7884.

(34) Hwang, W.; Xin, G.; Cho, M.; Cho, S.; Chae, H. Electrospray Deposition of Polymer Thin Films for Organic Light-Emitting Diodes. *Nanoscale Res. Lett.* 2012, 7, S2.

(35) Sajid, M.; Zubair, M.; Doh, Y. H.; Na, K.-H.; Choi, K. H. Flexible Large Area Organic Light Emitting Diode Fabricated by Electrohydrodynamics Atomization Technique. *J. Mater. Sci. Mater. Electron.* 2015, 26, 7192−7199.

(36) Miyazaki, H.; Koishikawa, Y. Method of Fabricating White Organic Light-Emitting Device by Using Electrospraying. U.S.Patent 10,026,931B2, 2018.

(37) Digimizer Image Analysis Software. https://www.digimizer.com/ (accessed May 28, 2018).

(38) Google Code Archive - Long-term storage for Google Code Project Hosting. https://code.google.com/archive/p/jtchem/downloads (accessed May 16, 2018).