Challenges and Capabilities of Conductive Polymeric Materials for Electromechanical Stimulation of Stem Cells

A Case Study

Marlitt Viehrig, Sampo Tuukkanen, Pasi Kallio
Department of Automation Science and Engineering, Tampere University of Technology, Tampere, Finland.

***Abstract* — Cell cultivation devices that mimic the complex microenvironment of cells in the human body are of high importance for the future of stem cell research. This paper introduces a prototype of an electromechanical stimulation platform as a modular expansion of an earlier developed mechanical stimulation device for stem cell research. A solution processable ink from PEDOT:PSS and graphene is studied as a suitable material for fabrication of transparent stretchable electrodes. Challenges of electrode integration on a flexible membrane using this material are critically discussed.**

Keywords— Electromechanical Stimulation, Conductive Polymer, PEDOT: PSS, graphene/PEDOT:PSS ink, Stem Cells

I. Introduction

Functionalities of cells and tissues in the human body greatly depend on biochemical, mechanical and electrical cues expressed by the cellular microenvironment [1-7]. This microenvironment is largely responsible for cell maturation and functional fate [2-3] [5-6]. Standard *in vitro* cell cultivation technologies are merely able to recreate the biochemical cues present in the human body. Therefore, they fail to mimic the entirety of the microenvironment, which makes them unsuitable as physiologically relevant *in vivo* models [8]. Thus, the development of a biomimetic cell cultivation device that recreates complex cellular microenvironments is of high importance especially for stem cell research.

One step in this direction has been taken by J. Kreutzer et. al in [9] through the development of a controlled mechanical stimulation device for stem cell research. The system is proposed as a modular mechanical stimulation platform for stem cells. Hereby, a partial vacuum pressure is applied to a cavity in a two-shell system with an attached flexible membrane. The vacuum pressure causes the inner shell to buckle inwards causing an equiaxial in-plane strain of maximal 10 % to the membrane. Stem cells attached to the membrane through covalent functionalization experience a controlled equiaxial strain. Fig.1 shows the system and its functionality. Pressure control allows various stretching modes to be realized, such as static and sinusoidal strains. Other mechanical cell stimulation platforms do not allow the simultaneous application of classical optical imaging standardly used in cell cultivation, as they are either non-transparent or bulky [10-13]. The described pneumatic stretching device is a small modular system exploiting high optical quality compatible with standard analytics in cell cultivation and customizable stimulation features. Polydimethylsiloxane (PDMS) as the main material allows rapid prototyping [14] of the stretching platform. However, due to its hydrophobic nature [15] it is challenging to use as a base for fabricating composites [16-18] with solution processable materials.

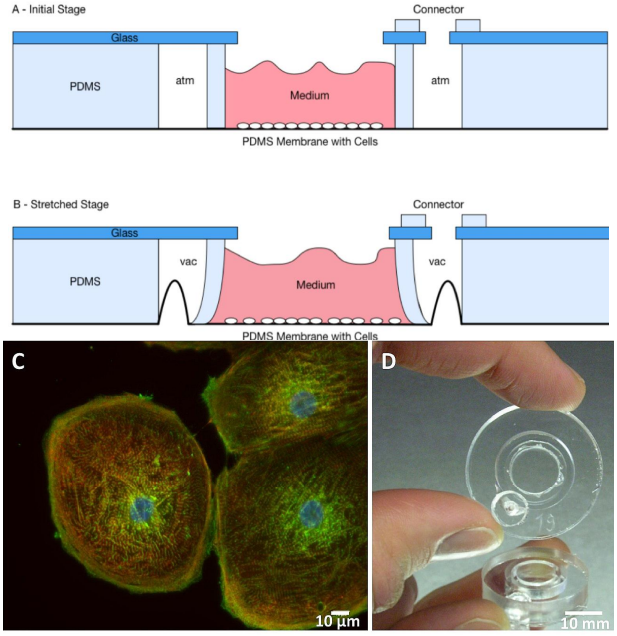

Fig. 1. Schematic working principle (A) and photograph (B) of pneumatic stretching device. Optical capabilities are illustrated by fluorescence microscope image (C) of cardiomyocytes. (Adapted from [9]).

The aim of this case study is to introduce a flexible, transparent thin-film electrode as a modular expansion into the described mechanical stretching system to create an electromechanical stimulation platform.

Hereby, it is of high importance that functional properties of the existing system are not compromised, but enhanced through the straightforward addition of an electrical module. Electrode materials applied for this task need to be highly stretchable and transparent to sustain the excellent optical properties of the system. Also the material of choice needs to be easy to pattern on PDMS, compatible with biofunctionalization procedures as well as relatively cheap and suitable for rapid prototyping.

Stretchable conductors made of special polymer blends have gained interest in all kind of device prototyping that are out of scope with classic silicon technologies. Poly(3,4-ethylendioxythiophene):polystyrene sulfonate (PEDOT:PSS) is a widely studied, solution processable conductive polymer commonly used for a variety of unconventional, mechanically challenging applications in biomedical engineering, such as fabrication of electronic skin [19-20], BioMEMS fabrication for electrical stimulation of neuronal cells [21] and thin film sensor fabrication for biosensing [22-25]. The native PEDOT:PSS has shown a limited stretching capability [21]. The conductivity and stretchability [21, 26-27] of PEDOT:PSS ink can, however, be further enhanced by incorporation of graphene. Incorporation of graphene into the base polymer enables the formation of reorganized conduction paths, as observed in bare graphene [28-29] or carbon-nanotube films [30-32]. The use of graphene/PEDOT:PSS ink was recently demonstrated as electrode material for piezoelectric thin-film sensors [26-28] and material for photonics [26]. Its chemical and mechanical properties [26-27] make it a suitable base material for the study addressed in this paper, as only 10 % in-plane elongation is required.

This paper focuses on exploring fabrication of thin-film electrodes and strategies to integrate graphene/PEDOT:PSS electrodes into the described mechanical stimulation platform.

II. Experimental

A. Fabrication of Equiaxial Stretching Device

The basic equiaxial stretching device was fabricated and characterized earlier by Kreutzer et. al [9]. Briefly, the system consists of two main components: (1) the system base consisting of two concentric PDMS shells irreversibly bonded to a glass cover plate and (2) a 120 µm thick, stretchable PDMS membrane.

PDMS (Sylgard 184, Dow Corning, USA) for prototyping of the shell structures and the membrane was prepared using a standard protocol. The silicone elastomer pre-polymer (base) was mixed with a cross linker (curing agent) thoroughly in a 10:1 w/w ratio. Followed by removal of trapped air in the mixture with a vacuum chamber and mold casting or spin coating of the material for the desired structure. Finally, the structure was cured for 10h at 60°C in an oven (Binder GmbH, Germany).

The concentric shells were punched from a PDMS disk (h= 7 mm) using custom made punching tools to create a thin inner shell (t = 1.5 mm) and stabilizing outer shell (t = 6.5 mm) with a cell cultivation chamber (t = 12 mm) in the middle and a vacuum pressure cavity (t = 2 mm) between the shells. The shells are irreversibly bonded to a glass cover plate using oxygen plasma (Diener Electronic GmbH, Ebhausen, Germany) at 0.3 mbar and 30 W for 15 s. The glass cover leaves an opening for the cell cultivation chamber and a connection hose to the vacuum cavity.

For the stretchable membrane, PDMS was spin coated on a polystyrene (PS) plate for 30s at 700 revolutions per minute (rpm). After curing, the assembled system base was irreversibly bonded to the membrane with oxygen plasma (Diener Electronic GmbH, Ebhausen, Germany) at 0.3 mbar and 30 W for 20 s. Finally, the assembled system was peeled from the PS plate and used for experimental preparation.

B. Conductive Polymer

A commercially available, solution processable, PEDOT:PSS based ink was used in fabricating electrodes on PDMS. The material is a conductive ink composed of graphene and PEDOT:PSS (denoted graphene/PEDOT:PSS in the following sections). The graphene/PEDOT:PSS ink is formulated for inkjet printing (P3014 Graphene Ink, Phene™Plus Series, Innophene Co., Thailand). It is a dark blue liquid that does not dissolve once crystallized and was used as purchased without additives.

C. Electrode Fabrication

The general hydrophobic nature of PDMS [15] hinders the direct fabrication of well-adhered thin films from solution processable materials [16-18]. To promote adhesion and allow even surface coverage, the PDMS membranes were rendered hydrophilic by applying oxygen plasma (pressure: 0.3 mbar, power: 30 W, duration: 120 s) to the PDMS membrane.

Four different deposition strategies for the incorporation of a conductive, transparent thin- film onto the PDMS membrane were evaluated. Hereby, two general integration strategies were distinguished: (1) large area thin-film applications, such as spin coating and spray coating, and (2) micropatterning of thin films through microcontact printing and channel casting.

Large area deposition techniques were conducted prior to assembly of the stretching device to the stabilized PDMS membrane. The ink was spin coated in a three-step protocol: 10 s at 200 rpm, followed by 500 rpm for 30 s and 10 s at 100 rpm. Hereby, multiple coating rounds allowed layering of the ink material. Similar layers were achieved through spray coating of ink at a pressure of 3 bar on a 60°C heated membrane. The samples were heated using a standard hotplate throughout the deposition process. After deposition, the inks were allowed to crystallize at 60°C for 20 min in an oven (Binder GmbH, Germany).

To pattern areas with conductive ink, microcontact printing technique using soft lithography was applied. Positive PDMS stamps patterned with groove structures (height h = 6 µm, distance d = 10 – 30 µm) were plasma oxidized at 0.3 mbar and 30 W for 15 s. To overcome the fast crystallization of conductive inks, ink material was applied to the originally prepared membrane followed by immediate stamping. The structures were allowed to crystallize in an oven

(Binder GmbH, Germany) at 60°C for 20 min. After crystallization, the stamps were carefully peeled from the substrate.

D. Resistance Measurements

Resistance of the fabricated films were measured using a multimeter in a two-point (Fluke 175, Fluke Vertriebsges. m.b.H., Austria) and a four-point (Keithley 2425 100 W Source Meter) probe set-up, which was described previously in [36].

E. Stretching Experiments

Stretching tests were conducted using the fabricated equiaxial stretching devices under applied partial vacuum pressure. The partial vacuum pressure was controlled using an in-house developed stretching system described earlier [9]. The systems were equiaxially stretched in a stepwise procedure to a maximum static strain of 10 % at 400 mbar in 50 mbar increments. Between each pressure step, the static strain was maintained for 2 - 5 min.

Electrical characterization of the films was performed during static strain using the two-point set-up and the multimeter under manual dry contact.

F. Microscopic Analysis

After material deposition, system fabrication and throughout the stretching experiments, optical microscopy images (Axio Observer.Z1, Carl Zeiss AG, Germany) were taken from all samples. Hereby, transmitted light, phase contrast and fluorescence imaging techniques at 100x and 200x magnification were utilized.

III. Results and Discussion

A. Thin-Film Electrode Deposition

Oxygen plasma treatment resulted in a low water contact angle (< 15 °) and therefore in high wettability of the substrate. This allowed the applied inks to form thin-films with high transparency in the desired area for all the deposition methods.

It was possible to form large electrodes covering the complete circular cell cultivation area (d = 12 mm) using spin and spray coating. The formed thin-film electrodes display an optical transparency close to pure PDMS and are homogenous throughout the electrode area. Thus, they would allow live imaging of cells. Microscopy images of the resulting electrodes in comparison to pure PDMS can be seen in Fig. 2. Furthermore, the resistivity of the deposited electrodes was at 100 ± 10 Ω for spin coated electrodes and 80 ± 11 Ω for spray coated electrodes. However, upon release of the PDMS membrane from the PS plate during system assembly cracking of the thin-film structures occurred. Crack formation, as illustrated in Fig. 2, was observed throughout the complete electrode area limiting its optical properties for the envisioned applications. Resistivity measurements of the cracked electrodes showed non-significantly different values. So the electrical functionality of the material layer was retained.

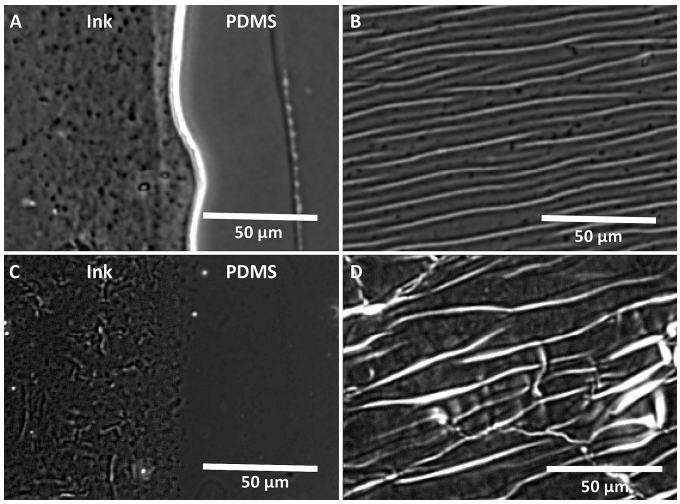

Fig. 2: Spin coated (A, B) and spray coated (C,D) graphene/ PEDOT:PSS layers on before (A,C) and after membrane (B,D) membrane release.

Patterning of electrodes by microcontact printing resulted in regular patterned electrodes on the PDMS substrate. However, microscope imaging and resistivity measurements of the structures suggested that the patterns did not form homogenous and continuous layers with measureable electrical activity. Furthermore, as observed with large area electrodes, crack formation upon substrate release rendered the structure non-usable.

The previously reported results suggest that the out-of-plane displacement of the membrane during substrate release exceeds the Young's modulus of the material resulting in the observed crack formation. Therefore, only deposition methods applicable to a released substrate already incorporated into the final stretching device are expected to sustain the desired device properties. Spray coating and channel casting can be applied to the assembled system and therefore became main focus of further studies.

Spray coating of large area electrodes on the released PDMS membrane delivered results comparable to electrodes fabricated on the PS supported membrane. Channel casted electrodes displayed a resistivity of 150 ± 12 Ω. However, induced out-of-plane stress on the electrode material from the probes used for electrical characterization caused easy crack formation and ultimately breaking of the material resulting in high difficulties to achieve repeatable measurements.

B. Stretching of Electrodes

Stretching characterizations of conductive polymeric materials have generally been done in uniaxial [17, 27, 35, 37] or biaxial [38-40] stretching modes. Z. Bao et. al [17] reported that a basic PEDOT:PSS can withstand strains of up to 12 % without cracking in uniaxial stretching mode. Furthermore, low strain values (< 30 % elongation) do not significantly (> 5 %) change the resistivity of the material. Functionality tests of electrodes made from a conductive polymeric material under equiaxial strain have not yet been studied. Thus, we characterized the deposited electrodes under applied strain to verify the suitability of the material in equiaxial stretching.

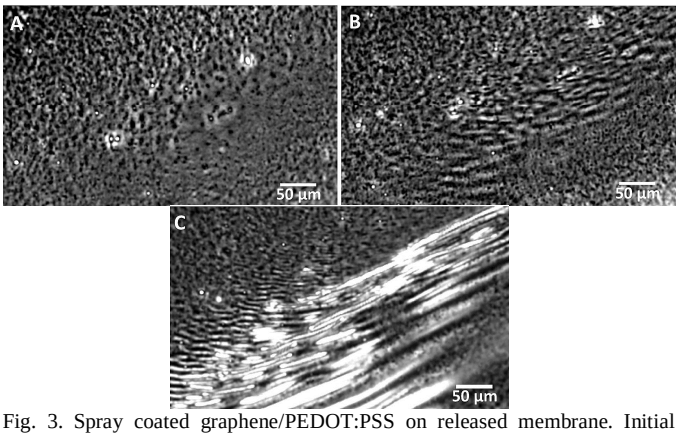

Fig. 3. Spray coated graphene/PEDOT:PSS on released membrane. Initial signs of delamination can be observed at 1.25 % strain in comparison to homogenous coatings at 0 % strain. At maximal strain (10 %) delamination and cracking effects become increasingly apparent.

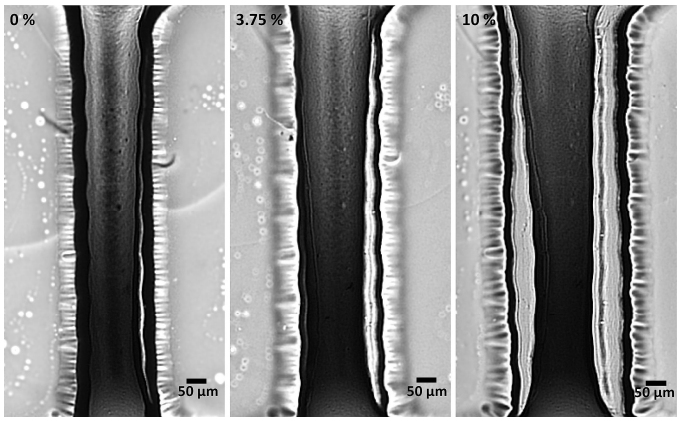

Fig. 4. Channel casted graphene/PEDOT:PSS throughout stretching procedure. Initial signs of delamination can be observed at 3.75 % strain at the structure edge. Delamination continuous along the channel edges and bottom until at highest possible strain (10 %) the channel sides are completely free.

Hereby, electrodes fabricated using spray coating displayed partial delamination of the electrode layer from the PDMS membrane as early as 1.25 % applied strain (vacuum pressure: 100 mbar). This is illustrated in Fig. 3.

During step-wise strain increase to 200 mbar (strain: 5 %), the material buckled around the partially adhered electrode material. Above 5 % strain, the material did cause the substrate to exhibit cracking and functional loss. The resistivity of the electrode throughout the stretching procedure increased by 50 % from 80 ± 11 W to 120 ± 10 W. Hence, while the material retained its electrical properties, it experienced non-homogenous strain because of the partial delamination, and affected the PDMS substrate in a negative way.

Delamination of the electrodes fabricated by channel casting was initially observed starting from the edges of the reservoir at 3.75 % applied strain (vacuum pressure: 150 mbar). The effect continued along the channel side until the maximum strain of 10 % was reached. Fig. 4 shows optical microscopy images of this process. Simultaneously, small cracks started to form within the electrode material at 3.75 % applied strain. As previously, the electrode retained its electrical properties and only exhibited a 25 % increase in resistance from 150 ± 12 W to 200 ± 15 W. Delamination could only be observed at the sides of the channel as well as inside the reservoir and was not accompanied with buckling of the electrode material or signs of stress within the PDMS membrane. Adhesion of ink to PDMS at the bottom of the channels seems to retain, while adhesion at the sides is low. But continuous delamination throughout repetitive stretching cycles could be observed, causing the electrodes to become loose and fixated only through the geometrical shape of the channel. In addition, the material also shows crack formation under equiaxial strain that limits the optical properties and therefore application potential for our envisioned platform.

Throughout the stretching procedure both spray coated and channel casted electrodes retain their electrical functionality. Even after multiple stretching cycles, the measured resistivity returns to the measured initial values in resting state. Also no significant change in resistivity over time (> 2 weeks) in resting state could be detected. However, maximal peak resistivity values, especially in channel casted structures, could not be reached after the first stretching cycle due to delamination of the electrode material from the substrate.

IV. Conclusion

The application potential of a solution processed electrode for incorporation in an equiaxial mechanical stimulation device for stem cell cultivation was shown. The applied graphene/PEDOT:PSS ink is initially compatible with cell cultivation conditions, as it is non-water soluble and non-toxic after curing. Hence, electrode integration and functionality on a stretchable substrate was studied as key factors for the envisioned cell applications. Hereby, electrode integration without extensive surface treatment for adhesion promotion was placed in the focus of this study.

Fabricated electrodes display features of material fatigue (e.g. crack formation) already at low equiaxial strains, which stands in contrast to previously described strain values in uniaxial. However, in [17] thin films contained 1 % Zonyl and adhesion promotion was achieved utilizing long exposure to ultraviolet/ozone. As we aimed to use minimal surface treatment, only oxygen plasma exposure for short terms was used.

On the other hand, the electrical properties of the material, similar to the observations in [17], were retained before, during and after stretching, which makes them a material with potential for the envisioned application, given different integration procedures.

In this case study, solution processed graphene/ PEDOT:PSS electrodes showed finally only a limited use for the envisioned electromechanical stimulation platform in stem cell research. The degradation of optical properties through crack formation render the systems non-functional for live cell imaging and experimental follow up. Adhesion promotion between PDMS and the solution processed graphene/PEDOT:PSS film is another challenge in the current approach. The applied oxygen plasma treatment to hydrophilize the substrate did not provide stable adhesion. Stronger chemical treatments, such as surface hydrolysis or exposure to ultraviolet/ozone [17], are needed to provide

sufficient adhesion. In addition to the reported difficulties with the integration procedure, it is questionable if the systems would be compatible with classic surface biofunctionalization protocols for conductive polymers [21-25], because in the current approach multiple surface materials (PDMS, graphene/PEDOT:PSS) are present in the cultivation area.

REFERENCES

[1] F. Gattazzo, A. Urciuolo, and P. Bonaldo, "Extracellular matrix: a dynamic microenvironment for stem cell niche.," *Biochim. Biophys. Acta*, vol. 1840, no. 8, pp. 2506–19, Aug. 2014.

[2] M. F. Pera and P. P. L. Tam, "Extrinsic regulation of pluripotent stem cells.," *Nature*, vol. 465, no. 7299, pp. 713–20, Jun. 2010.

[3] Y. Sun, C. S. Chen, and J. Fu, "Forcing stem cells to behave: a biophysical perspective of the cellular microenvironment.," *Annu. Rev. Biophys.*, vol. 41, pp. 519–42, Jan. 2012.

[4] P.-X. Wan, B.-W. Wang, and Z.-C. Wang, "Importance of the stem cell microenvironment for ophthalmological cell-based therapy.," *World J. Stem Cells*, vol. 7, no. 2, pp. 448–60, Mar. 2015.

[5] S. D. Eshghi S, "Engineering microenvironments to control stem cell fate and function - PubMed - NCBI," *Stembook, Cambridge (MA), Harvard Stem Cell Institute*, 2008. [Online]. Available: http://www.ncbi.nlm.nih.gov/pubmed/20614608. [Accessed: 05-Apr-2016].

[6] A. Pavesi, G. Adriani, M. Rasponi, I. K. Zervantonakis, G. B. Fiore, and R. D. Kamm, "Controlled electromechanical cell stimulation on-a-chip.," *Sci. Rep.*, vol. 5, p. 11800, Jan. 2015.

[7] N. W. S. Kam, E. Jan, and N. A. Kotov, "Electrical stimulation of neural stem cells mediated by humanized carbon nanotube composite made with extracellular matrix protein.," *Nano Lett.*, vol. 9, no. 1, pp. 273–8, Jan. 2009.

[8] L. G. Griffith and G. Naughton, "Tissue engineering--current challenges and expanding opportunities.," *Science*, vol. 295, no. 5557, pp. 1009–14, Feb. 2002.

[9] J. Kreutzer, L. Ikonen, J. Hirvonen, M. Pekkanen-Mattila, K. Aalto-Setälä, and P. Kallio, "Pneumatic cell stretching system for cardiac differentiation and culture.," *Med. Eng. Phys.*, vol. 36, no. 4, pp. 496–501, Apr. 2014.

[10] C. Moraes, J.-H. Chen, Y. Sun, and C. A. Simmons, "Microfabricated arrays for high-throughput screening of cellular response to cyclic substrate deformation.," *Lab Chip*, vol. 10, no. 2, pp. 227–34, Jan. 2010.

[11] L. Huang, P. S. Mathieu, and B. P. Helmke, "A stretching device for high-resolution live-cell imaging.," *Ann. Biomed. Eng.*, vol. 38, no. 5, pp. 1728–40, May 2010.

[12] L. A. McMahon, A. J. Reid, V. A. Campbell, and P. J. Prendergast, "Regulatory effects of mechanical strain on the chondrogenic differentiation of MSCs in a collagen-GAG scaffold: experimental and computational analysis.," *Ann. Biomed. Eng.*, vol. 36, no. 2, pp. 185–94, Feb. 2008.

[13] W. W. Ahmed, M. H. Kural, and T. A. Saif, "A novel platform for in situ investigation of cells and tissues under mechanical strain.," *Acta Biomater.*, vol. 6, no. 8, pp. 2979–90, Aug. 2010.

[14] H. Hwang, G. Kang, J. H. Yeon, Y. Nam, and J.-K. Park, "Direct rapid prototyping of PDMS from a photomask film for micropatterning of biomolecules and cells.," *Lab Chip*, vol. 9, no. 1, pp. 167–70, Jan. 2009.

[15] A. Mata, A. J. Fleischman, and S. Roy, "Characterization of polydimethylsiloxane (PDMS) properties for biomedical micro/nanosystems.," *Biomed. Microdevices*, vol. 7, no. 4, pp. 281–93, Dec. 2005.

[16] T. Lu, L. Finkenauer, J. Wissman, and C. Majidi, "Rapid prototyping for soft-matter electronics," *Adv. Funct. Mater.*, vol. 24, no. 22, pp. 3351–3356, 2014.

[17] D. J. Lipomi, J. A. Lee, M. Vosgueritchian, B. C.-K. Tee, J. A. Bolander, and Z. Bao, "Electronic Properties of Transparent Conductive Films of PEDOT:PSS on Stretchable Substrates," *Chem. Mater.*, vol. 24, no. 2, pp. 373–382, Jan. 2012.

[18] B. Charlot, G. Sassine, A. Garraud, B. Sorli, A. Giani, and P. Combette, "Micropatterning PEDOT:PSS layers," *Microsyst. Technol.*, vol. 19, no. 6, pp. 895–903, 2013.

[19] M. L. Hammock, A. Chortos, B. C. K. Tee, J. B. H. Tok, and Z. Bao, "25th anniversary article: The evolution of electronic skin (E-Skin): A brief history, design considerations, and recent progress," *Adv. Mater.*, vol. 25, pp. 5997–6038, 2013.

[20] S. J. Benight, C. Wang, J. B. H. Tok, and Z. Bao, "Stretchable and self-healing polymers and devices for electronic skin," *Prog. Polym. Sci.*, vol. 38, no. 12, pp. 1961–1977, Dec. 2013.

[21] A. Blau, A. Murr, S. Wolff, E. Sernagor, P. Medini, G. Iurilli, C. Ziegler, and F. Benfenati, "Flexible, all-polymer microelectrode arrays for the capture of cardiac and neuronal signals.," *Biomaterials*, vol. 32, no. 7, pp. 1778–86, Mar. 2011.

[22] P. Lin, F. Yan, J. Yu, H. L. W. Chan, and M. Yang, "The application of organic electrochemical transistors in cell-based biosensors.," *Adv. Mater.*, vol. 22, no. 33, pp. 3655–60, Sep. 2010.

[23] J. Park, H. K. Kim, and Y. Son, "Glucose biosensor constructed from capped conducting microtubules of PEDOT," *Sensors Actuators B Chem.*, vol. 133, no. 1, pp. 244–250, Jul. 2008.

[24] E. Moczko, G. Istamboulie, C. Calas-Blanchard, R. Rouillon, and T. Noguer, "Biosensor employing screen-printed PEDOT:PSS for sensitive detection of phenolic compounds in water," *J. Polym. Sci. Part A Polym. Chem.*, vol. 50, no. 11, pp. 2286–2292, Jun. 2012.

[25] A. Wisitsoraat, S. Pakapongpan, C. Sriprachuabwong, D. Phokharatkul, P. Sritongkham, T. Lomas, and A. Tuantranont, "Graphene-PEDOT:PSS on screen printed carbon electrode for enzymatic biosensing," *J. Electroanal. Chem.*, vol. 704, pp. 208–213, 2013.

[26] T. Isoniemi, S. Tuukkanen, and D. Cameron, "Measuring optical anisotropy in poly (3, 4-ethylene dioxythiophene): poly (styrene sulfonate) films with added graphene," *Org. …*, 2015.

[27] U. Lang, N. Naujoks, and J. Dual, "Mechanical characterization of PEDOT:PSS thin films," *Synth. Met.*, vol. 159, no. 5–6, pp. 473–479, Mar. 2009.

[28] E. H. Hwang, S. Adam, and S. Das Sarma, "Carrier Transport in Two-Dimensional Graphene Layers," *Phys. Rev. Lett.*, vol. 98, no. 18, p. 186806, May 2007.

[29] *Graphene: An Introduction to the Fundamentals and Industrial Applications*. Wiley, 2015.

[30] R. D. Mehlenbacher, T. J. McDonough, M. Grechko, M.-Y. Wu, M. S. Arnold, and M. T. Zanni, "Energy transfer pathways in semiconducting carbon nanotubes revealed using two-dimensional white-light spectroscopy.," *Nat. Commun.*, vol. 6, p. 6732, Jan. 2015.

[31] I. Jeon, T. Chiba, C. Delacou, Y. Guo, A. Kaskela, O. Reynaud, E. I. Kauppinen, S. Maruyama, and Y. Matsuo, "Single-Walled Carbon Nanotube Film as Electrode in Indium-Free Planar Heterojunction Perovskite Solar Cells: Investigation of Electron-Blocking Layers and Dopants.," *Nano Lett.*, vol. 15, no. 10, pp. 6665–71, Oct. 2015.

[32] G. Gruner, "Carbon nanotube films for transparent and plastic electronics," *J. Mater. Chem.*, vol. 16, no. 35, p. 3533, Sep. 2006.

[33] T. Vuorinen, M. Zakrzewski, S. Rajala, D. Lupo, J. Vanhala, K. Palovuori, and S. Tuukkanen, "Printable, Transparent, and Flexible Touch Panels Working in Sunlight and Moist Environments," *Adv. Funct. Mater.*, vol. 24, no. 40, pp. 6340–6347, Oct. 2014.

[34] S. Rajala, S. Tuukkanen, and J. Halttunen, "Characteristics of Piezoelectric Polymer Film Sensors With Solution-Processable Graphene-Based Electrode Materials," *IEEE Sens. J.*, vol. 15, no. 6, pp. 3102–3109, Jun. 2015.

[35] S. N. K. Rajala, M. Mettanen, and S. Tuukkanen, "Structural and Electrical Characterization of Solution-Processed Electrodes for Piezoelectric Polymer Film Sensors," *IEEE Sens. J.*, vol. 16, no. 6, pp. 1692–1699, Mar. 2016.

[36] S. Tuukkanen, T. Julin, V. Rantanen, M. Zakrzewski, P. Moilanen, K. E. Lilja, and S. Rajala, "Solution-processible electrode materials for a heat-sensitive piezoelectric thin-film sensor," *Synth. Met.*, vol. 162, no. 21–22, pp. 1987–1995, 2012.

[37] S. Tuukkanen, M. Hoikkanen, M. Poikelispää, M. Honkanen, T. Vuorinen, M. Kakkonen, J. Vuorinen, and D. Lupo, "Stretching of solution processed carbon nanotube and graphene nanocomposite films on rubber substrates," *Synth. Met.*, vol. 191, pp. 28–35, 2014.

[38] S. Savagatrup, A. S. Makaram, D. J. Burke, and D. J. Lipomi, "Mechanical Properties of Conjugated Polymers and Polymer-Fullerene Composites as a Function of Molecular Structure," *Adv. Funct. Mater.*, vol. 24, no. 8, pp. 1169–1181, Feb. 2014.

[39] K. Xie and B. Wei, "Materials and structures for stretchable energy storage and conversion devices.," *Adv. Mater.*, vol. 26, no. 22, pp. 3592–617, Jun. 2014.

[40] D. J. Gundlach, T. N. Jackson, D. G. Schlom, and S. F. Nelson, "Solvent-induced phase transition in thermally evaporated pentacene films," *Appl. Phys. Lett.*, vol. 74, no. 22, p. 3302, May 1999.