Intrinsically Stretchable Field-Effect Transistors

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Abstract

Thin-film field-effect transistor (TFT) is a three-terminal device comprised of source, drain and gate electrodes, a dielectric layer, a semiconductor layer, and a substrate. TFT is a fundamental building component behind a variety of electronic devices. Developing an intrinsically stretchable TFT entails availability and usage of only functional materials with elastomeric deformability in response to externally applied stresses. This represents major material challenges. In this article, we will survey strategies that have been taken to synthesize these elastomeric functional materials, and how these materials are assembled to fabricate intrinsically stretchable TFT devices. Developing solution-based printing technology to assemble intrinsically stretchable TFT is considered a prospective strategy for the research field of wearable electronics for industrial adaptation in the near future.

Keywords: Intrinsically stretchable, field-effect transistors, wearable electronics

1. Introduction

Imagine a large-area electronic display that can be folded to easily fit into your pocket, a rollable window curtain that illuminates the room, or a smartphone screen that doubles in size when stretched like rubber. Recent efforts to develop stretchable and wearable electronics have suggested that such scientific fantasies may soon become a reality. Flexible electronics that are bendable and can withstand small strains, usually no more than 1%, have already penetrated into current market. Here we refer to stretchable electronics which can survive strains of at least 10% and thus can endure exotic shape changes like folding, twisting, and conforming on complex curvilinear surfaces [1]. Among the large number of electronic functions, thin-film field-effect transistor (TFT) is a fundamental building component enabling a variety of electronic applications, such as dynamic information displays and imaging sensors [2].

Currently, combining elastic interconnects with discrete rigid (non-stretchable) functional transistor units is the most widely used strategy in the fabrication of stretchable TFT array [3-6]. The rigid and brittle transistor units are embedded in or bonded onto the surface of soft rubbery polymers. The resulting TFTs usually show high stretchability and efficiency since the TFT units do not accommodate deformation during stretching. However, there is a trade-off between device density and stretchability as large portions of elastic interconnecting regions are required between transistor units to realize stretchability [7]. The fabrication process is also cumbersome and incompatible with fully solution-printed processes. The other strategy to realize stretchable TFT is to construct stretchable TFT in which all the constituent materials are elastomeric, namely intrinsically (fully) stretchable TFT [7–13].

TFT is a three-terminal device comprised of source, drain and gate electrodes with a dielectric and a semiconducting channel. Thus, developing an intrinsically stretchable TFT entails development of each component with elastomeric stretchability. So far, extensive efforts have been performed on investigating stretchable electrodes with high stretchability and conductivity. Advanced materials and processing techniques developed in recent years for stretchable electrodes employing carbon-based materials like CNTs and graphene, metallic materials like metal nanowires and liquid metal, and conducting polymer like PEDOT:PSS offer new approaches that could be explored for the development of stretchable source, drain and gate electrodes in intrinsically stretchable TFT [13-15]. However, the field of developing dielectric and active channel materials with high stretchability and efficiency still requires additional attention. In this article, we will highlight and discuss the recent progress made in high-performance stretchable dielectric materials and semiconductive materials, and how these elastomeric materials are assembled to fabricate intrinsically stretchable TFT devices.

2. Elastomeric dielectric materials

2.1 Ion gel

Ion gels, which are highly attractive for many applications, have been employed on stretchable electronics thanks to their printability, high ionic conductivity, large specific capacitance, and stretchability [9,10,12,16]. Unlike conventional dielectric materials used in transistors, the operation of ion gel gated transistors is based on the formation of a high capacitance electric double layer (EDL) at the ion gel/semiconductor and ion gel/gate electrode interfaces under an electric field.

Ahn and Cho first reported utilizing ion gel as stretchable dielectric for stretchable TFTs [9]. Ion gel dielectric consisting of poly(styrene-methyl methacrylate-styrene) (PS-PMMA-PS) triblock copolymer and 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([EMIM][TFSI]) ionic liquid in ethyl acetate solvent (weight ratio of 0.1:0.9:9) was fabricated onto the channel material made of patterned graphene. After solvent evaporation, PS-PMMA-PS triblock copolymer in the ionic liquid formed a physically stretchable gel through noncovalent association of PS components that are insoluble in the ionic liquid. In another study, the Li and Takenobu groups fabricated stretchable molybdenum disulfide thin-film transistors on poly(dimethylsiloxane) substrates using the same ion gel as the elastic dielectric. The capacitance of the ion gel obtained at 15 Hz is as large as 5.78 $\mu F/cm^2$, and remains nearly constant with an applied tensile strain up to 22% [10].

Using the same ion gel as the stretchable dielectric layer, the Arnold group fabricated an intrinsically stretchable TFT device using semi-conductive SWCNTs as the channel material [16]. The large capacitance of the ion gel gate dielectric allowed large saturation currents to be obtained at very low gate (V_G) and drain (V_D) biases: $I_D = \sim 640\ \mu A$ at $V_G = -2$ V and $V_D = -1$ V. In contrast to previous cases of stretchable ion gel dielectric mentioned above, the capacitance of this ion gel during the stretch-release process decreases linearly with increasing applied strain and maintains its linearity with subsequent stretch-and-release cycles. The Jeong group also used ion gel as the dielectric to construct a highly stretchable polymer transistor. To fabricate the ion gel dielectric, a solution mixture composed of poly(ethylene glycol)-diacrylate (PEG-DA), 2-hydroxy-2-methylpropiophenone (HOMPP), and [EMIM][TFSI] (88:8:4, w/w) was coated on the active materials, followed by UV crosslinking. The resulting ion-gel dielectric layer was capable of being stretched up to strain of 100%.

While ion gel dielectric could offer large capacitance, the switching speeds of the TFT are often limited by the relatively slow movement speed of the ionic molecules, which ultimately limits the frequency at which the devices could operate reliably. Decreasing the thickness of the ion gel is a common technique to decrease the frequency dependence of the ion-gel dielectric TFT. As demonstrated by the Jeong group, the capacitance of their ion gel with 50 μm thickness showed severe loss at 500 Hz while the capacitance of their ion gel with 100 nm thickness showed good stability, even up to 1000 Hz with a small decreasing slope at higher frequencies [12]. Even though thickness control of the ion gel can alter the frequency dependence of the ion-gel dielectric TFT, devices made with dielectrics of very thin ion gel are more prone to larger leakage currents and unstable device performances.

2.2 Dielectric Elastomer

In contrast to ion gel, dielectric elastomers are solid-state materials, which have excellent processibility as the materials can be handled similarly to conventional rigid polymers. Conventional elastomer, including polydimethylsiloxane (PDMS), hydrogenated styrene ethylene butylene styrene (H-SEBS), and polyurethane (PU) can be applied on stretchable TFT as dielectric with excellent stretchability [17]. The Bao group reported an organic-based stretchable TFT using P3HT as the channel and PU as the dielectric [7]. The outstanding stretchability of the PU allowed the device to be stretched up to ~265% strain. The capacitance of the PU dielectric increased as the tensile strain increased, and the strain-induced changes in dielectric capacitance were consistent with the expected value based on mechanical deformation with a Poisson ratio of 0.5. Albeit having excellent stretchability, conventional elastomers usually have low dielectric constant, which significantly constrains the electrical performance of the TFT device.

To improve the dielectric constant of the dielectric elastomer, the Pei group developed a new PU-PEG copolymer consisted of a PU-co-PEG main chain and PEG side chains [2]. PEG was selected for its high flexibility, low monomer viscosity (25 cps @ 25ºC), and semi-crystalline structure of its homopolymer [18,19]. The introduction of PEG with semi-crystalline structure into the polymer main chain is to provide an internal surface between the amorphous and the crystalline phases, which is sufficient for Maxwell–Wagner–Sillars-type polarization [18,19]. The interfacial polarization caused by trapping of free charges at the boundaries between the crystalline structure and the amorphous regions enhances the dielectric constant for the PU-co-PEG copolymer. Moreover, PEG was selected as the side chains for its high polarity and high chain mobility under room temperature, which further improve the dielectric constant of the copolymer. The resulting elastomeric PU-PEG copolymer dielectric was reported to achieve: 1) dielectric constant as high as ~13 at 12 Hz, 2) elongation at break at > 50% strain, 3) small hysteresis, fast recovery and high resilience, and 4) stable capacitance constant throughout repeated stretch-release cycles. The stretchable TFT using this PU-co-PEG copolymer as the dielectric layer showed significant improvement in device performance [2]. However, the device still exhibited hysteresis in the transfer curve due to large layer thickness (~1.1 µm) of the PU-PEG copolymer used to repress the gate leakage current.

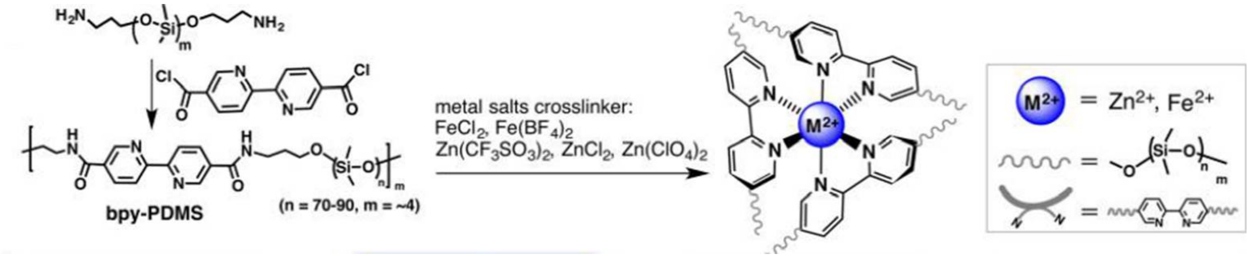

Figure 1. The synthetic route of metal salts cross-linked PDMS.

To realize intrinsically stretchable TFT with hysteresis-free transfer characteristics, Bao et al. recently reported a new self-healing dielectric elastomer achieved by the incorporation of metal−ligand coordination as cross-linking sites in nonpolar polydimethylsiloxane (PDMS) polymers [11]. The design strategy is demonstrated in Figure 1. Bipyridine moieties were first embedded into a long PDMS chain, which is subsequently cross-linked via metal−ligand coordination between bipyridine moieties and transition-metal ions, Zn^{2+} and Fe^{2+}. The advantages of this design include: 1) the increase in dielectic constant by the introduction of metal-ligand coordination bonds; 2) the stabilization of the complex structure which could potentially serve as energy dissipation sites in the bulk material by the dynamic coordinating ability of the counteranions; and 3) the reduction in ion mobility and ionic effects caused by the incorporation of ions in the nonpolar PDMS matrix. As a result, fully stretchable transistors utilizing $FeCl_2$-PDMS as stretchable dielectrics exhibited ideal hysteresis-free transfer characteristics while the gate leakage current remained low even after 1000 cycles of tensile deformation at 100% strain.

2.3 Wrinkled metal oxide

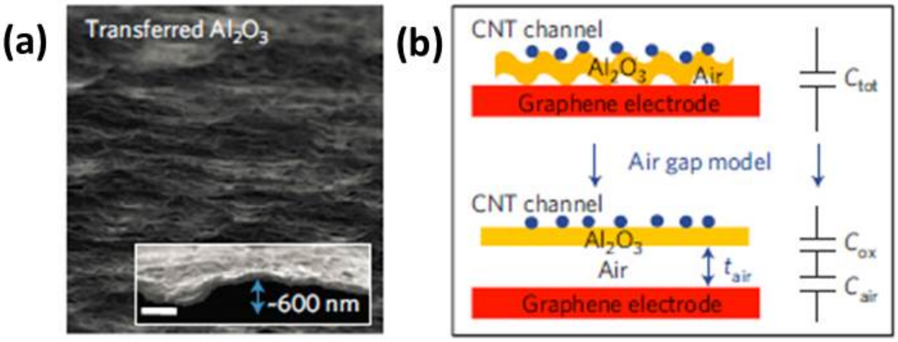

Figure 2. (a) SEM images of wrinkled Al_2O_3 layer. The inset shows the macroscopic wrinkle (scale bar, 1 μm). (b) The schematic of the air-gap model with a series connection of the Al_2O_3 layer and air gap.

Typical inorganic gate oxides are brittle and prone to defects upon deformation but possess outstanding electrical benefits in TFT dielectric applications since dielectric elastomers often have higher leakage current and lower dielectric constant despite their excellent stretchability and good processability. A new approach for preparing wrinkled Al_2O_3 (Figure 2(a)) gate dielectric layer using a transfer method was proposed by the Lee group [8]. The wrinkled Al_2O_3 layer contained effective built-in air gaps with a small gate leakage current of 10^{-13} A. Although the specific gate capacitance of the wrinkled Al_2O_3 was measured to be 1.49 nF cm^{-2} at 100 kHz, which is 80 times lower than that of planar Al_2O_3 (Figure 2(b)), the fabricated devices using

SWCNT as the channel materials and wrinkled Al_2O_3 as the stretchable dielectric exhibit no appreciable decreases in performance. This effect is attributed to a strong electric field localized at the valley of the wrinkles, which allows strict control over the carriers in the SWCNT channel and gives rise to enhanced device performance. The wrinkled dielectric layer allows the transistors to retain performance under strains as high as 20% without appreciable increases in leakage current, observable physical degradation, and performance loss after stretching and releasing the devices for over 1,000 times.

3. Elastomeric semiconductive materials and assemble of intrinsically stretchable TFT

3.1 Two-dimentional materials

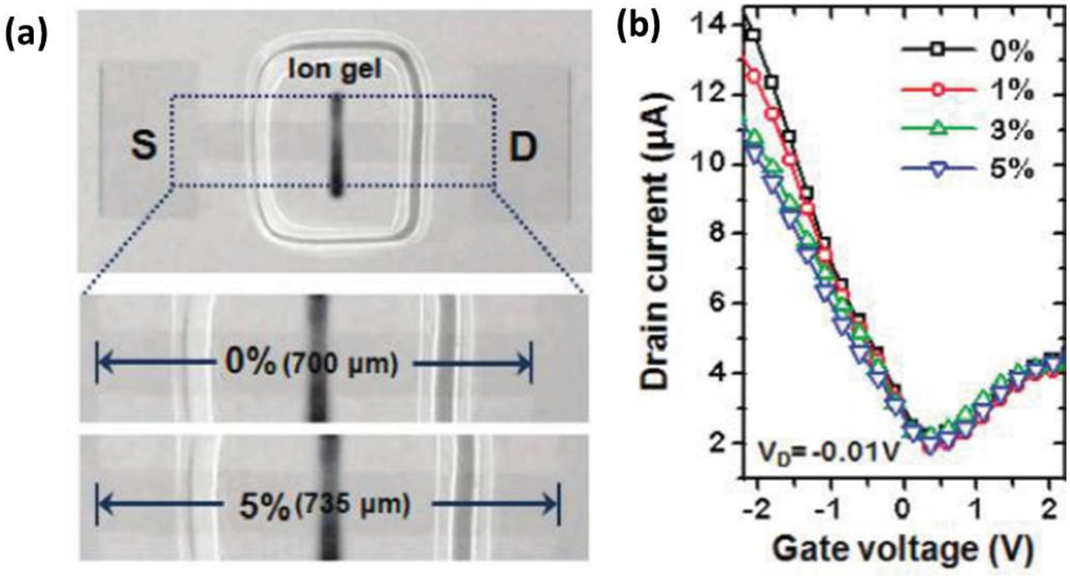

Figure 3. (A) Microscope images of graphene-based TFTs under substrate stretching up to 5% along the longitudinal direction of the channel. (B) Typical transfer characteristics of strained graphene-based FTETs on PDMS.

The Ahn and Cho group presented a low-temperature transferring/printing route to fabricate an all-graphene-based FET array on a stretchable rubber [9]. The source/drain electrodes and semiconducting channels of the all-graphene-based transistors were monolithically patterned from graphene films which were synthesized on Cu foils through CVD. Ion gel and PSS:PEDOT were printed over the channel as the dielectric layer and the gate electrode respectively to complete the intrinsically stretchable TFT. The resulting TFT showed hole and electron mobilities of 1188±136 and 422±52 $cm^2/(Vs)$, respectively, and the electrical properties were invariant after 1000 or more cycles of 5% strain (Figure 3). The devices were able to retain its performance despite repeated deformability mainly because of the extremely large Young's modulus of graphene and/or the relaxation of ripples created during the transfer of graphene films to the rubber substrates. However, the characteristic of zero-band-gap semiconductor for graphene causes the TFT to exhibit a very low on/off current ratio (<100). Degradation of the device characteristics was also observed at applied strains larger than 5%, which is possibly due to microcracks initiated at grain boundaries and/or other defects in the graphene films. The Li and Takenobu groups replaced graphene with 2D molybdenum disulfide as channel materials to construct stretchable TFT [10]. Using ion gel as stretchable dielectric layer, the TFT on PDMS substrate displayed an on/off ratio of $\sim 10^4$, mobility in the range of 0.4–1.4 $cm^2/(Vs)$, and small variation in electrical characterististics during stretching of up to 4% strain.

3.2 One-dimensional nanotube or nanowire networks.

A network of semiconducting single-walled carbon nanotubes (semi-SWCNTs) is an attractive option for the channel material of stretchable TFT thanks to the mechanical resilience of the nanotubes, the capability to accommodate large strain via nanotube−nanotube sliding and buckling, and the extraordinary charge transport characteristics of the nanotubes [16]. The Lee group reported a stretchable TFT combining a SWCNT network channel with stretchable graphene/SWCNT electrodes and a wrinkled Al_2O_3 dielectric layer [8]. The semi-SWCNT network was prepared using APCVD, transferred onto the patterned graphene electrodes and wrinkled Al_2O_3 dielectric layer, and patterned using photolithography and O_2 plasma etching. The semi-SWCNT network not only worked as the channel material but also prevented the graphene electrodes from being damaged during stretching. The resulting devices exhibited an excellent on/off ratio of $\sim 10^5$, a high mobility of ~ 40 cm^2 $V^{-1}s^{-1}$, and a low operating voltage of less than 1 V. Moreover, the devices that were stretched along the length direction up to 16% strain and along the width direction up to 20% strain showed only mild fluctuation in the on/off ratio without major variation in device performance. Under a fatigue test of 10% strain for 1,000 stretching and releasing cycles, the on/off ratio, transconductance, and mobility of the devices also fluctuated mildly with no major deterioration. However, the stretchable TFT cannot withstand higher strains that were larger than 20% for either direction.

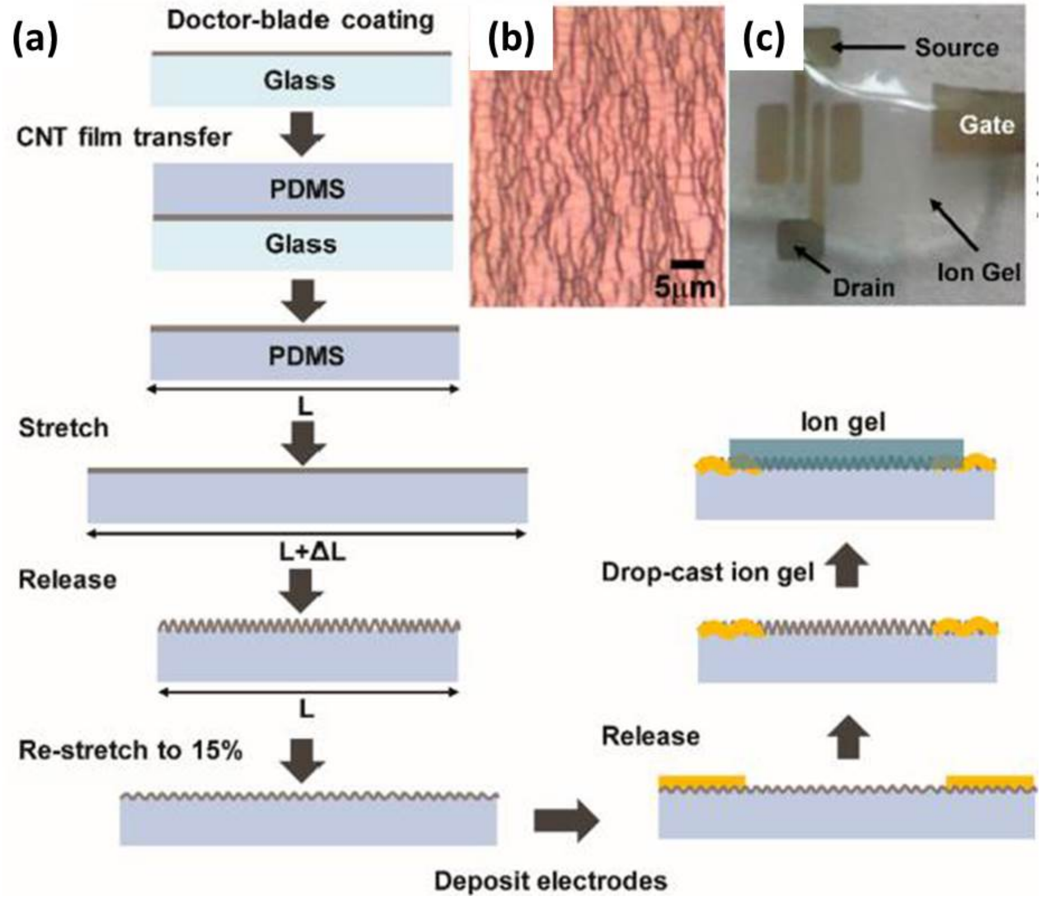

Figure 4. (a) Schematic showing transfer, buckling of semi-SWCNT film, and fabrication of stretchable SWCNT-based TFTs. (b) Optical image of electrodes after buckling. (c) Optical photograph of a SWCNT-based TFT.

Also utilizing semi-SWCNT as the channel material, the Arnold group substantially modified the TFT design in order to increase the stretchability of the device through (1) buckling the semi-SWCNT thin film networks in the active channel and (b) integrating the buckled channel with an ion gel as the stretchable dielectric layer and a buckled thermally evaporated Au metal film as the stretchable electrode (Figure 4) [16]. The semi-SWCNTs were isolated from as-grown CNTs, produced via cobalt molybdenum catalysis of carbon monoxide disproportionation, using poly(9,9-dioctylfluorene) as a selective agent in toluene. The enriched and polymer-wrapped semi-SWCNTs were then solution-casted onto the glass substrates to form network thin-films, followed by transferring onto strained PDMS substrates to form the stretchable channel layer. The resulting TFT performed as a typical p-type channel device with an on/off current ratio reaching 4 × 10^4 and mobility of 10.6 $cm^2/(V{\cdot}s)$. Moreover, the devices exhibited fairly stable operation with invariant mobility and on/off current ratio up to 50.8% strain and did not showed significant reduction in performance until an applied strain of 57.2%.

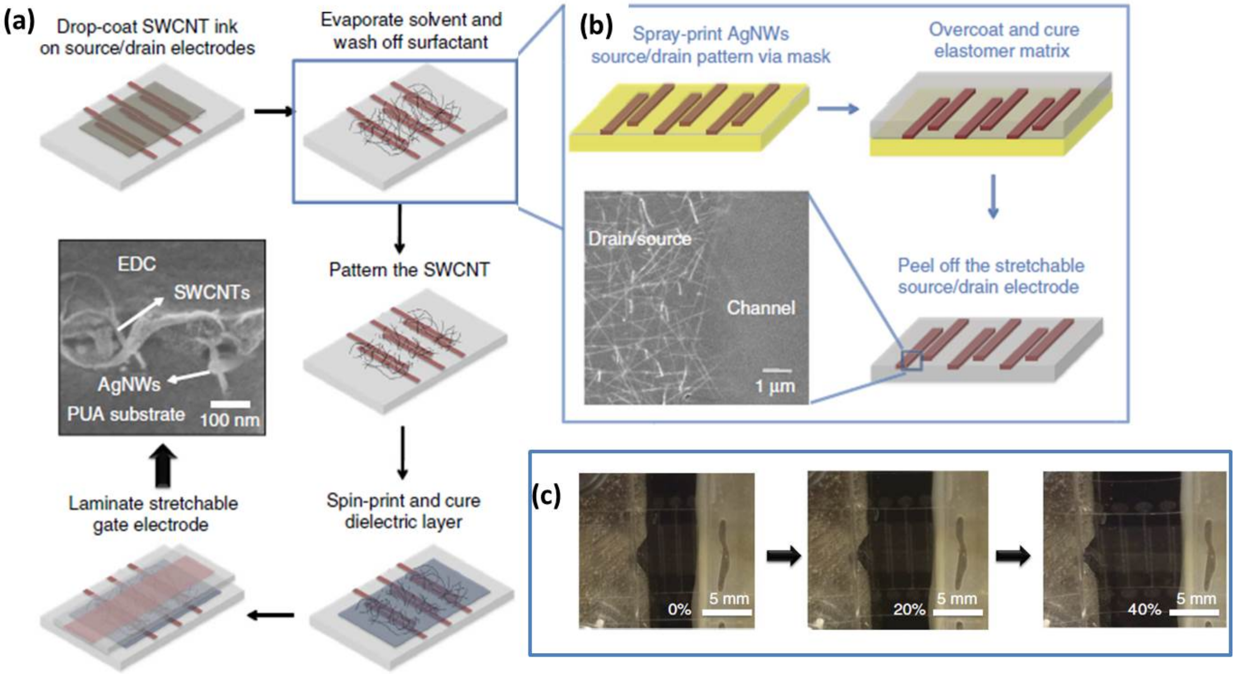

Figure 5. (a) and (b) Schematic illustration of the fabrication steps for a stretchable SWCNT-based TFT. (c) Photographs of a device at specified strains applied along the channel length direction.

Great accomplishments have been made for stretchable TFT using semi-SWCNTs as the channel material as reviewed above. In order to simplify the fabrication process and reduce the production cost of stretachable TFTs, the Pei group developed an all solution-based process to fabricated arrays of fully stretchable semi-SWCNTs-based TFTs (Figure 5) [2,13]. The stretchable TFT employed a silver nanowire (AgNW)-PU acrylate (PUA) composite as the stretchable electrodes and a PU-co-PEG elastomeric dielectric. The active channel material was printed from drop-casting a formulated semi-SWCNT ink which was prepared by mixing Fluorosurfactant and propylene glycol with a 99+% semi-SWCNT solution (IsoNanotubes-S from NanoIntegris, Inc.) at an optimized ratio. For a total of 36 stretchable TFT devices fabricated with a yield of 85.7%, the averages of the $I_{ON/OFF}$ ratio, I_{ON}/W, g_m/W, and mobility measured at V_D=-2.0 V are 7,200±1,200, 30.0±8.93 mA mm^{-1}, 11.2±3.40 mS mm^{-1}, and 27.0±7.44 $cm2V^{-1}$ s^{-1}, respectively. The resulting device can be stretched up to 50% along either the channel length or channel width direction without significant decrease in performance. Moreover, the device only showed mild decrease in mobility after subjecting to 500 cycles of continuous stretch–relaxation cycles between 0 and 20% strains along the channel length or channel width direction. This stability in device performance under large strain and repeated stretch–relaxation deformation is resulted from the unique structure where the elastomeric dielectric layer infiltrates into the semi-SWCNT network and wraps around the nanotubes.

3.3 Organic channel materials

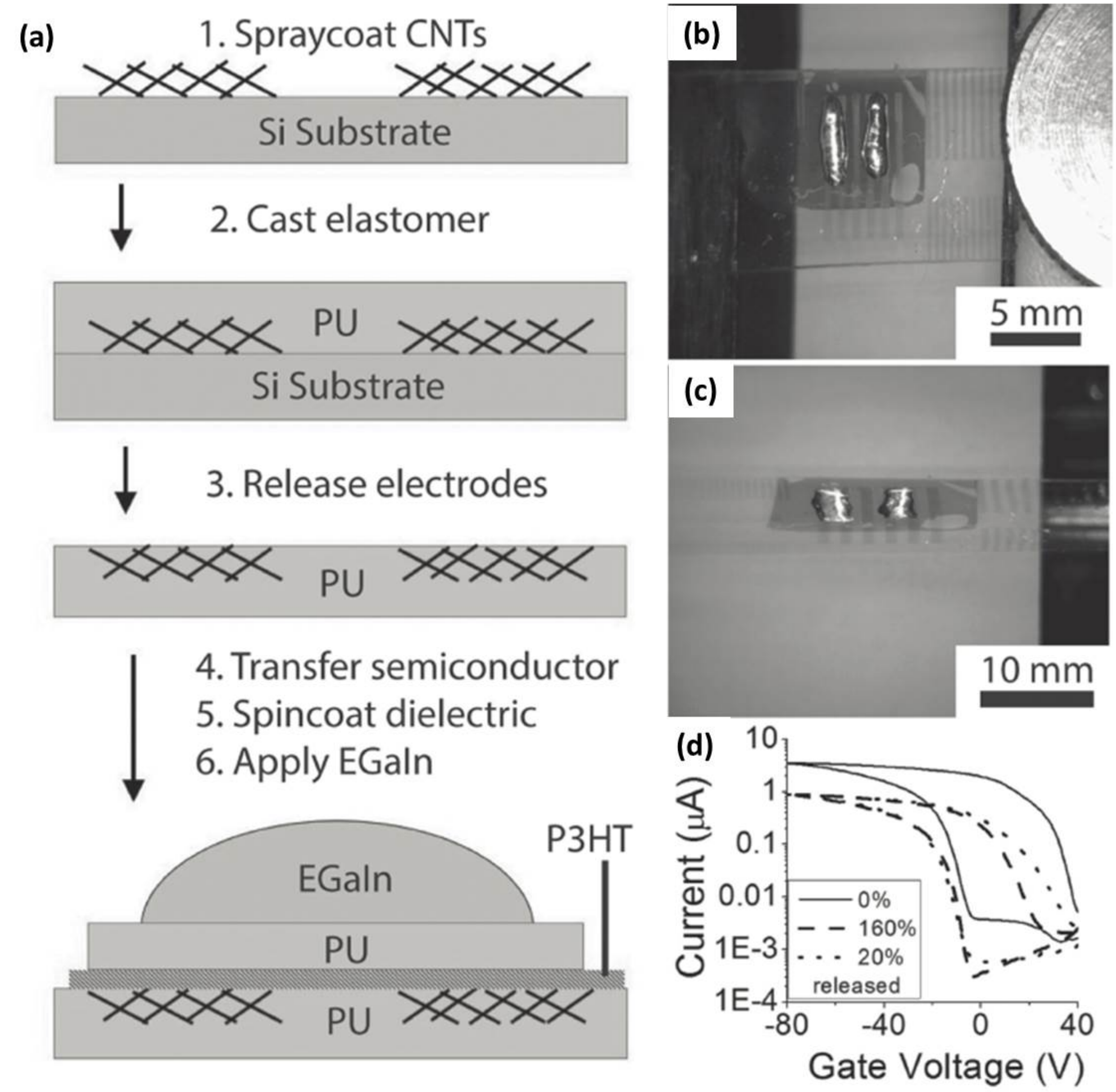

Figure 6. (a) Schematic of the fabrication process for the stretchable organic-based TFT. Microscope image of a device with b) zero and c) 150% applied strain. d) Transfer curves for a device stretched to 160% and released to 20%. The source drain bias was −20 V and the W/L is 40.

Even though inorganic semiconductors show better electrical performance than organic semiconductors, other material characteristics, such as mechanical properties, materials availability and compatibility with high throughput processing, and all-solution processibility, allows many organic semiconductors to be an excellent alternative for stretchable electronics [7].

Research conducted by the O’Connor group suggests that a rigid, 3D packing structure of common polymeric semiconductors results in fracture at very low strain, while a 2D packing structure of the organic conductor P3HT allows deformation greater than 150% strain [20]. In addition, the Bao group fabricated a top gate architecture of stretchable organic-based TFT as displayed in Figure 6 [7]. Patterned CNT-PU elastomer composite electrodes were used as stretchable source and drain electrodes. P3HT films were formed on PU electrodes as active channel layer, and a 4 μm thick PU dielectric layer was spin-coated on top, followed by applying eutectic gallium indium (EGaIn), a liquid metal, as the gate electrode to complete the device construction (Figure 6). The average values of device mobility and on/off ratio at upstretched state are $3.4 \cdot 10^{-2} \pm 1.63 \cdot 10^{-2}$ cm^2/Vs and 591 ± 461, respectively. Strikingly, the devices still exhibited transistor characteristics when stretched up to strains as large as ~265%. Moreover, strain-independent I_{ON} was achieved during multiple perpendicular stretching cycles, which were considered to be achieved by programming microcracks with an initial strain. The Jeong group further constructed a stretchable polymer-based TFT by employing stretchable Au nanosheet electrodes with ion gel for the gate dielectric [12], and electrospun P3HT nanofibers for the active channel material. Au was selected because of its low energy barrier for charge injection into the P3HT fiber network. The measured hole mobility of the device was 23 cm^2 /Vs in the saturation regime at $V_D = -1$ V. Upon deformation, the mobility can maintain the same value up to 40% strain and slightly decrease to 16 cm^2 /Vs at 70% strain. Moreover, the device exhibit outstanding mechanical durability with a stable on-off current ratio and an unchanged V_{th} during the fatigue test of 1,500 cycles. All these results indicate excellent mechanical compliance of stretchable polymer-based TFTs.

4. Conclusions and Outlook

The emergence of devices that combine elasticity with electronic or optoelectronic properties are still facing significant materials challenges. Collectively, the advances in materials and engineering summarized in this article provide three promising strategies to design elastomeric materials for intrinsically stretchable TFT. First, conventional rigid or non-stretchable materials could be fabricated into new structures such as buckling structures and networks of metallic nanowires to provide stretchability. For instance, while bulk silver is rigid without elasticity, the new silver structure---one dimensional AgNWs---can form thin film networks that can retain the high conductivity of silver while accommodating large strain. Second, nanoscale functional materials could be embedded into elastomeric matrices to fabricate stretchable nanocomposites. Achieving highly uniform nanomaterials dispersion and the compatibility of the material interface of the nanomaterial and the matrix material are the major challenges to improve the synergistic effect of the fillers and matrix elastomers. Third, novel functional elastomers could be designed and synthesized to produce elastic-yet-high-performance molecular materials that have been highlighted in Lipomi's recent review [21]. Investigative studies for the better understanding of the relationships between molecular structure and the physical properties of the materials will be beneficial for future development in this field.

Beyond material design, further development in the processing/fabrication techniques to assemble the different stretchable components congruently to construct intrinsically stretchable TFT and arrays in cost-effective and reliable way is also needed. Conventional deposition method used in the electronic industry that requires high temperature and high vacuum processing conditions are often undesirable for the deposition of elastomeric materials. With this in mind, developing solution-based printing technology to assemble intrinsically stretchable TFT is a promising research direction to reduce fabrication costs and to seek more suitable methods to fabricate devices with these new materials.

Acknowledgements

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Kwing Tong studied Biomedical Engineering at the University of California: Irvine where he received his bachelor's degree in 2009. He is currently completing his PhD under the supervision of Dr. Qibing Pei at in the Department of Materials Science & Engineering at UCLA, synthesizing multifunctional polymers and investigating the structure-property relationships of nanomaterial-polymer composites to develop transparent conductors via cost-effective scalable fabrication methods for flexible and stretchable electronics.

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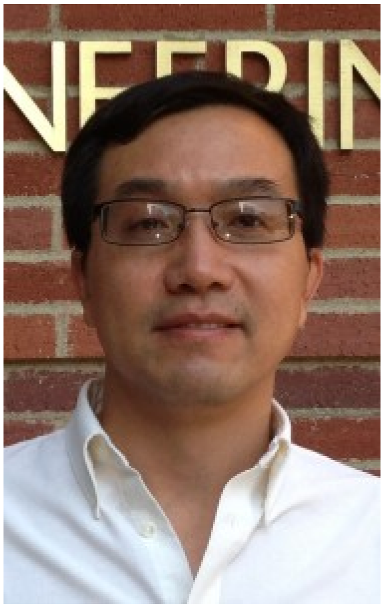

Dr. Qibing Pei is a professor of Materials Science and Engineering at the University of California, Los Angeles. He has developed a number of electronic and electroactive polymers for applications in electro-optic and electro-mechanical devices, including light emitting diodes, polymer light emitting electrochemical cells, electroactive polymer artificial muscles, and biologically-inspired robots. His research interests cover a wide range of soft materials and span from material synthesis, processing, to design of functional devices. He applies polymer synthesis, solution-based processing and nanofabrication in the discovery of new functional polymers and composites.