

BIOCOMPATIBLE IONIC LIQUID IN SEMICONDUCTING POLYMER

Cross-Reference to Related Application

[0001] This application claims the benefit of priority of Singapore Patent Application
5 No. 10202103759R, filed 13 April 2021, the content of it being hereby incorporated
by reference in its entirety for all purposes.

Technical Field

[0002] The present disclosure relates to a biocompatible composite and a method of
10 forming the biocompatible composite. The present disclosure also relates to uses of
the biocompatible composite.

Background

[0003] Organic electrochemical transistors (OECTs) may have attracted significant
15 attention owing to their numerous advantages such as large transconductance, low
operating voltages, fast response capability, compatibility with broad range of
fabrication techniques, etc. Research progress in OECTs ignited various applications
in amplifying transducer, biosensors, neuromorphic device, etc. Also, the use of
OECTs is becoming more significant in the field of wearable electronic devices.
20 Among these multifarious wearable devices, the detection of physiological signals
from human skin through the use of wearable electronics comes with a wide scope of
applications and products. However, the realization of wearable electronics using
organic materials demands a set of unique properties (biological compatibility is one
property considered for wearable devices) and various challenges remain for
25 application of OECTs in wearable electronics, such as the mechanical mismatch
behaviors between the electronic devices and biological surface, the use of
biocompatible materials, the stable operation on human body under a certain amount
of deformations, and the energy consumption for the long-term operation.

[0004] OECTs may consist of an active layer that is gated through an electrolyte. The
30 conductivity of the active layer may be modulated through the doping/de-doping
process when ions from the electrolyte penetrate into the channel under an applied
gate bias. Semi-conducting polymer (poly-3,4-ethylenedioxythiophene;poly-4-

styrenesulfonate) (PEDOT:PSS) tends to have desirable environmental stability, good transparency, facile solution processability, and favorable biocompatibility, thus presenting potential in bioelectronics, logic circuits and neuromorphic devices. For example, PEDOT:PSS may take the role of the active layer between source and drain electrodes in OECTs. However, pristine PEDOT:PSS films suffer from poor electrical conductivity (10^{-5} - 10 S cm^{-1}) and low μC^* product (here, μ : the electronic mobility; C^* : the volumetric charge storage capacitance), which is attributed to its intrinsic PEDOT core-PSS surface layer structure. In such structure, the conductive hydrophobic PEDOT cores may be encapsulated by the hydrophilic and insulating PSS chains. Having the conductive PEDOT cores mostly surrounded by the insulating PSS chains may undesirably restrict the transport of mobile carriers through PEDOT domains.

[0005] Over the last few decades, efforts have been made to explore and improve the electrical property of PEDOT:PSS through the rearrangement of PEDOT:PSS chain conformation, or removal of excess PSS chains, and building larger and more internal connected PEDOT-rich structures facilitating the charge carriers transport. Some of these may involve additional use of surfactants, solvents, acids, etc. For instance, in OECTs, the ethylene glycol (EG) additive was traditionally explored for improving electrical conductivity and transconductance of PEDOT:PSS based devices. The incorporation of EG facilitates the aggregation of PEDOT phase, forming enlarged PEDOT-rich domains with shorter PEDOT π - π stacking, and thus enhancing the charge carriers transport property in the bulk film. There may be other co-solvents or dispersion additives such as Triton X-100, dimethyl sulfoxide, and x-sorbitol that were explored for providing a similar effect on the morphology change so as to improve electrical properties of PEDOT:PSS based devices. Another study considered blending of PEDOT:PSS with several ionic liquids (e.g. EMIM:TCB, EMIM:TCM) to induce the morphology change and enhancement in the conductivity, but these ionic liquids were not proven to be non-toxic or biocompatible, limiting their application in wearable electronics.

[0006] There is thus a need to provide for a solution that addresses one or more of the limitations mentioned above. The solution should at least provide for a biocompatible

composite that addresses one or more of aforesaid limitations and a method of forming the biocompatible composite.

Summary

[0007] In a first aspect, there is provided for a biocompatible composite comprising:

a semiconducting polymer having an electrically conductive domain and an electrically insulating domain coupled to the electrically conductive domain; and

a biocompatible ionic liquid doped in the semiconducting polymer,

wherein the biocompatible ionic liquid renders a portion of the electrically insulating domain distal from the electrically conductive domain to expose a portion of the electrically conductive domain.

[0008] In another aspect, there is provided for a method of forming the biocompatible composite described in various embodiments of the first aspect, the method comprising:

providing a semiconducting polymer having an electrically conductive domain and an electrically insulating domain coupled to the electrically conductive domain;

forming an aqueous solution comprising the biocompatible ionic liquid; and

mixing the semiconducting polymer and the aqueous solution.

5 [0009] In certain non-limiting embodiments, the semiconducting polymer may be formed as a film by annealing and then being post-treated with an aqueous solution comprising the biocompatible ionic liquid. Post-treating of the semiconducting polymer film with the aqueous solution may be carried out by, as non-limiting examples, casting, blading or coating, the aqueous solution comprising the
10 biocompatible ionic liquid on the semiconducting polymer film. Alternatively, the semiconducting polymer film may be submerged into the aqueous solution comprising the biocompatible ionic liquid or be subject to an atmosphere formed from the aqueous solution comprising the biocompatible ionic liquid.

[0010] In another aspect, there is provided for an organic electrochemical transistor
15 comprising:

a source electrode and a drain electrode; and

the biocompatible composite described in various embodiments of the first aspect configured as an active layer between and in contact with both the source electrode and the drain electrode.

[0011] In another aspect, there is provided a method of forming the organic electrochemical transistor, the method comprising:

providing a substrate;

forming a source electrode and a drain electrode on the substrate;

forming a channel between the source electrode and the drain electrode after forming the source electrode and the drain electrode for subsequent formation of an active layer;

forming a first insulating layer and a second insulating layer on the first insulating layer to cover portions of the source electrode and the drain electrode not covered by the active layer;

forming the biocompatible composite described in various embodiments of the first aspect as the active layer between and in contact with both the source electrode and the drain electrode; and

removing the second insulating layer and retaining the first insulating layer to cover portions of the source electrode and the drain electrode not covered by the active layer.

[0012] In another aspect, there is provided a wearable device operable as a pressure sensor, the wearable device having a sensing component comprising:

at least two electrodes configured on a substrate; and

the biocompatible composite described in various embodiments of the first aspect configured as a sensing layer on and in contact with the at least two electrodes.

[0013] In another aspect, there is provided a method of forming the wearable device, the method comprising:

providing a substrate having at least two electrodes;

forming a sensing component comprising the biocompatible composite described in various embodiments of the first aspect configured as a sensing layer on and in contact with the at least two electrodes; and

integrating the sensing component with the organic electrochemical transistor.

[0014] In another aspect, there is provided an electrocardiogram recording device comprising:

the organic electrochemical transistor; and

two or more testing electrodes attachable to a human skin surface,

wherein at least one of the two or more testing electrodes is connected to a gate electrode of the organic electrochemical transistor and at least another of the two or more testing electrodes is connected to a source electrode of the organic electrochemical transistor.

Brief Description of the Drawings

[0015] The drawings are not necessarily to scale, emphasis instead generally being placed upon illustrating the principles of the present disclosure. In the following description, various embodiments of the present disclosure are described with reference to the following drawings, in which:

[0016] FIG. 1A shows the chemical structures of the semiconducting polymer PEDOT:PSS and the ionic liquid [MTEOA][MeOSO₃].

[0017] FIG. 1B is a plot of conductivity of PEDOT:PSS films with varying concentration of [MTEOA][MeOSO₃]. The PEDOT:PSS incorporated with [MTEOA][MeOSO₃] is denoted herein as PEDOT:PSS/[MTEOA][MeOSO₃].

[0018] FIG. 1C shows UV-vis-NIR absorption spectra of pristine PEDOT:PSS and PEDOT:PSS/[MTEOA][MeOSO₃] films.

[0019] FIG. 1D shows Raman spectra of pristine PEDOT:PSS and the PEDOT:PSS/[MTEOA][MeOSO₃] film, obtained at an excitation wavelength of 532 nm.

[0020] FIG. 2A shows the cytocompatibility analysis of PEDOT:PSS/[MTEOA][MeOSO₃] films. Specifically, FIG. 2A shows representative fluorescence images of human dermal fibroblasts co-cultured with control plates group (first row) and films with [MTEOA][MeOSO₃] (PEDOT:PSS/[MTEOA][MeOSO₃]) (second row) counterstained with Hoechst and live/dead dyes at 2, 4 and 6 days, respectively. (Hoechst: cell nucleus, Calcein AM: living cell and EthD-1: dead cell) (n = 6 samples, scale bar denotes 100 μm).

[0021] FIG. 2B shows quantification of cell viability (extracted from live/dead staining) of control group (control plates group) and films with [MTEOA][MeOSO₃] (PEDOT:PSS/[MTEOA][MeOSO₃]) at 2, 4 and 6 days, respectively. Data represents mean $\pm$ standard deviation. Asterisk * denotes statistical difference between compared experimental groups. “n.s.” denotes not significant. Statistical significance is determined at $p < 0.05$.

[0022] FIG. 3A is a schematic diagram of the OEET of the present disclosure.

[0023] FIG. 3B shows one of the output characteristics of the PEDOT:PSS/[MTEOA][MeOSO₃] OEET of the present disclosure.

10 [0024] FIG. 3C is one of the transfer curves of the PEDOT:PSS/[MTEOA][MeOSO₃] OEET.

[0025] FIG. 3D is a plot of transconductance (gm) of the PEDOT:PSS/[MTEOA][MeOSO₃] OEET as a function of operating conditions at saturation and channel geometry.

15 [0026] FIG. 3E shows the mobility measurement using the current pulsed method ($V_{ds} = -0.1$ V). The linear fit to data yields a mobility of $1.659 \pm 1.131 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$.

[0027] FIG. 3F is the Bode plots which were acquired using the PEDOT:PSS/[MTEOA][MeOSO₃] films as the working electrode. The EIS data was fitted with an equivalent circuit model composed of a parallel capacitor (C_p), a parallel resistor (R_p), and a serial resistor (R_s).

20 [0028] FIG. 3G is the capacitance of PEDOT:PSS/[MTEOA][MeOSO₃] films with different film volume. The linear fit to the capacitance data yields a volumetric capacitance of $75.87 \pm 6.15 \text{ } \mu\text{F cm}^{-3}$.

[0029] FIG. 3H shows the transient response of drain current for a constant $V_{ds} = -0.5$ V and $V_{gs} = -0.4 \sim -0.7$ V pulse applied at the gate. The solid line is an exponential fit with a rise time of 40.57 μs .

[0030] FIG. 3I shows the corresponding response time of PEDOT:PSS/[MTEOA][MeOSO₃] OEETs with different channel geometries.

[0031] FIG. 3J shows the transient response of drain current with V_g switches from -0.4 to 0.7 V for pulse width of 0.001s, and constant $V_{ds} = -0.5$ V for 5000 cycling pulses, wherein the plot is magnified portion around 0 to 25 cycles and 4975 to 5000

cycles, demonstrating drain current retained a very high retention of 95% after 5000 cycles.

[0032] FIG. 3K is a table comparing the performance of the present OECT and traditional OECTs.

5 [0033] FIG. 3L shows comparison of transient responses. The last two rows are composites formed from method of the present disclosure.

[0034] FIG. 4A shows the atomic force microscopy phase image of a PEDOT:PSS/[MTEOA][MeOSO₃] film.

10 [0035] FIG. 4B shows the atomic force microscopy height image of a PEDOT:PSS/[MTEOA][MeOSO₃] film.

[0036] FIG. 4C shows the atomic force microscopy 3D surface images of a PEDOT:PSS/[MTEOA][MeOSO₃] film.

[0037] FIG. 4D shows the atomic force microscopy phase image of a PEDOT:PSS film.

15 [0038] FIG. 4E shows the atomic force microscopy height image of a PEDOT:PSS film.

[0039] FIG. 4F shows the atomic force microscopy 3D surface images of a PEDOT:PSS film.

20 [0040] FIG. 4G shows the absorption spectra of PEDOT:PSS/[MTEOA][MeOSO₃] film cast on ITO glass and recorded as a function of applied voltage bias varying from -0.4 to 0.8 V (with an increment of 0.2 V) in 0.1M NaCl electrolyte.

[0041] FIG. 4H shows the absorption spectra of PEDOT:PSS film cast on ITO glass and recorded as a function of applied voltage bias varying from -0.4 to 0.8 V (with an increment of 0.2 V) in 0.1 M NaCl electrolyte.

25 [0042] FIG. 4I shows the GIWAXS characterization of PEDOT:PSS/[MTEOA][MeOSO₃] films: the qz direction.

[0043] FIG. 5A is schematic representation of a pressure sensor of the present disclosure.

30 [0044] FIG. 5B shows a wearable device of the present disclosure (the sensor is connected to the source or drain electrode of the OECT in series, and a bandage is used to attach the sensor to the skin).

[0045] FIG. 5C is a plot of the transfer performance of the wearable device under different pressure.

[0046] FIG. 5D is a plot of the output performance of the wearable device under different pressure.

5 [0047] FIG. 6A shows a SEM image of the sensing component structure of the pressure sensor, specifically the top surface coated PEDOT:PSS/[MTEOA][MeOSO₃] film.

[0048] FIG. 6B is a magnified image of FIG. 6A.

[0049] FIG. 6C shows a SEM image of the sensing component structure of the pressure sensor, specifically the side of the coated PEDOT:PSS/[MTEOA][MeOSO₃] film.

[0050] FIG. 6D shows a SEM image of the sensing component structure of the pressure sensor, specifically the top surface coated PEDOT:PSS film.

[0051] FIG. 6E is a magnified image of FIG. 6D.

15 [0052] FIG. 6F shows a SEM image of the sensing component structure of the pressure sensor, specifically the side of the coated PEDOT:PSS film.

[0053] FIG. 7A shows the gate tunable sensitivity under a pressure of 4.8 kPa.

[0054] FIG. 7B shows the gate tunable sensitivity under a pressure of 16 kPa.

[0055] FIG. 7C demonstrates a wearable device of the present disclosure in detecting subtle motion (an example of a sensing application), specifically via the current change ratios of wrist pulse ($V_g = -0.4V$).

[0056] FIG. 7D demonstrates a wearable device of the present disclosure in detecting large motion (an example of a sensing application), specifically via the current change ratios of finger bending at various angles ($V_g = -0.4V$).

25 [0057] FIG. 7E demonstrates a wearable device of the present disclosure in detecting large motion (an example of a sensing application), specifically via the current change ratios of wrist rotation ($V_g = -0.2V$).

[0058] FIG. 7F demonstrates a wearable device of the present disclosure in detecting large motion (an example of a sensing application), specifically via the current change ratios of knee bending ($V_g = -0.2V$).

30 [0059] FIG. 8A shows a flexible OECT of the present disclosure using Parylene as the substrate.

[0060] FIG. 8B shows output characteristics of the flexible wearable device of the present disclosure before and after bending, where the bending radius (R) = 20, 15, 10, 5 mm of the flexible OECTs.

[0061] FIG. 8C shows transfer characteristics of the flexible wearable device of the present disclosure before and after bending, where the bending radius (R) = 20, 15, 10, 5 mm of the flexible OECTs.

[0062] FIG. 9A shows the ECG recording circuit configuration and the position of a wearable device of the present disclosure coupled to the heart for ECG measurement.

[0063] FIG. 9B shows a plot of normalized current against time for ECG measurements, wherein the QRS, P and T peaks are easily identifiable.

[0064] FIG. 9C shows a plot of normalized current against time, demonstrating stable and continuous ECG signals are well recorded by a wearable device of the present disclosure.

[0065] FIG. 10 shows two schematic diagram, one on formation of the source and drain electrodes on a substrate (top row images) and the other on formation of an organic electrochemical transistor (OECT) of the present disclosure (bottom row images). Fabrication of the OECT is described in more details in example 2C further hereinbelow. In the top row of images, it can be seen that a layer of photoresist was deposited on the substrate by photolithography for forming the source and drain electrodes (i.e. gold (Au) contacts). The photoresist was developed to define two areas on the substrate for subsequent deposition of gold. Excess gold and the photoresist were removed using acetone, rendering the gold source and drain electrodes on the substrate. In the bottom row images, it can be seen from the leftmost and first image that there is a bottom parylene C layer attached on the substrate. This bottom parylene C layer may be referred herein as a first insulating layer. The first insulating layer can be a polymer layer. The second parylene C layer is deposited on the first insulating layer after the first insulating layer is treated with a soap. The second parylene C layer may be herein referred to as a second insulating layer. The second insulating layer can be a polymer layer. The purpose of the soap treatment, which involves treating the first insulating layer with a soap solution, is to keep the second insulating layer as a separate layer from the first insulating layer. Prior to depositing the second insulating layer, the soap solution may be present. After, for example, spin-coating a

biocompatible semiconducting composite and an annealing process, due to the presence of the soap solution, the second insulating layer (e.g. parylene C layer) can be easily removed completely without compromising the first insulating layer (e.g. the bottom parylene C layer). As the first and second insulating layers can be formed of polymers, they may be referred to also as “first insulating polymer layer” and “second insulating polymer layer”, respectively. After tearing off the second insulating layer, the structure can be rinsed with DI water prior to measuring the device performance, the soap solution gets washed off thoroughly in this step. In the center-left image of bottom row, the advantage of etching is to obtain the channel. That is to say, to define the channel, both the first and second insulating layers (e.g. the 1st and 2nd parylene C layers) are etched off. While certain portions of the two layers of parylene C may remain during the etching process as can be seen in center-left and center-right images of the bottom row, the second parylene C layer gets eventually torn off after the coating, casting, printing or depositing to form the biocompatible composite in the channel and annealing the structure (see rightmost and last image in bottom row). For the channel, both the 1st and 2nd parylene C layers are etched off during the etching process, thus the biocompatible composite can reside in the channel and on the 2nd parylene C layer (e.g. on the photoresist) after spin-coating and annealing. The biocompatible composite may change from a liquid into a solid film after annealing. Meanwhile, most parts of the device except the channel are covered by the 1st and 2nd layer of parylene C. After annealing, the 2nd parylene C layer can be removed, at the same time, the biocompatible composite residing thereon is also removed together, i.e. only the biocompatible composite in the channel is retained. It should be noted that there is no gap between the active layer and the substrate, as well as the source and drain electrodes. The bottom 1st parylene C layer remains and covers certain portions of the substrate (or even a portion of both electrodes) except the channel.

Detailed Description

[0066] The following detailed description refers to the accompanying drawings that show, by way of illustration, specific details and embodiments in which the present disclosure may be practised.

[0067] Features that are described in the context of an embodiment may correspondingly be applicable to the same or similar features in the other embodiments. Features that are described in the context of an embodiment may correspondingly be applicable to the other embodiments, even if not explicitly described in these other embodiments. Furthermore, additions and/or combinations and/or alternatives as described for a feature in the context of an embodiment may correspondingly be applicable to the same or similar feature in the other embodiments.

[0068] The present disclosure relates to a biocompatible composite, its method of forming, and uses. To understand the present biocompatible composite, traditional organic electrochemical transistors (OECTs) are first discussed.

[0069] Traditionally, OECTs may have attracted significant attention. For example, pristine PEDOT:PSS may be used as an active layer between the source and drain electrodes in OECTs. However, pristine PEDOT:PSS films suffer from poor electrical conductivity, low electronic and ionic transport properties, which may be attributed to its intrinsic PEDOT core-PSS surface layer structure. Herein, ionic liquids are demonstrated as an effective way to enhance the conductivity (σ) of PEDOT:PSS. The present biocompatible composite, which incorporates an ionic liquid, differs at least in that the present ionic liquid does not suffer the same limitations of traditional ionic liquids. Traditional ionic liquids tend to be undesirable due to their non-biocompatibility and cytotoxicity and hence are potentially toxic, limiting their utilization in biological applications. Conversely, the present disclosure includes cytotoxicity studies of PEDOT:PSS-ionic liquid blends, demonstrating the biocompatible OECTs developed herein. In various non-limiting embodiments, for example, a biocompatible ionic liquid tris(2-hydroxyethyl)methylammonium methylsulfate [MTEOA][MeOSO₃] was used for the purpose of demonstration (not intended to be limiting) to dope PEDOT:PSS and enable higher OECT performance, yielding OECTs with high signal-transducing efficiency, large transconductance, desirable electronic and ionic transport properties, fast transient response and excellent switching cycling stability. The biocompatible ionic liquid [MTEOA][MeOSO₃] doped PEDOT:PSS films also demonstrated potential application as a sensing component of a pressure sensor, and as a gate voltage tunable

pressure sensor enabled by an integrated OECT and as a pressure sensing component (i.e. a promising element) in a wearable electronic device to detect full-range human physiological signals. The flexible and biocompatible PEDOT:PSS/[MTEOA][MeOSO₃] device is also successfully demonstrated as an on-skin ECG signals recording device.

[0070] Details of various embodiments of the biocompatible composite, its method of forming, uses, and advantages associated with the various embodiments are now described below. Where the embodiments and advantages have been demonstrated in the example section of the present disclosure, they shall not be reiterated for brevity.

[0071] In the present disclosure, there is provided a biocompatible composite. The biocompatible composite can be a biocompatible film. The biocompatible composite comprises a semiconducting polymer having an electrically conductive domain and an electrically insulating domain coupled to the electrically conductive domain, and a biocompatible ionic liquid doped in the semiconducting polymer, wherein the biocompatible ionic liquid renders a portion of the electrically insulating domain distal from the electrically conductive domain to expose a portion of the electrically conductive domain. To help understand this better, FIG. 1A is referred to. In FIG. 1A, it can be seen that the biocompatible ionic liquid, (e.g. [MTEOA][MeOSO₃]) dissociates into its cation and anion. The biocompatible ionic liquid renders a portion of the PSS chain distal (partially separate) from a portion of the PEDOT chain, but the PSS and PEDOT chains are in fact still coupled (not entirely separated). That is to say, the PEDOT:PSS chain network does not undergo extensive reconfiguration into another structure (e.g. a simpler fiber structure or even strands of fibers instead of its current network structure). In various embodiments, the biocompatible composite may be electrically conductive.

[0072] In various embodiments, the semiconducting polymer may comprise poly-3,4-ethylenedioxythiophene (PEDOT), poly-3,4-ethylenedioxythiophene:poly-4-styrenesulfonate (PEDOT:PSS), polypyrrole (PPy), polyaniline (PANI), polystyrenesulfonate, polythiophene having an alkoxy or ethylene glycol side chain, or a derivative thereof, or a combination thereof. Aforesaid semiconducting polymers are not intended to be limiting, i.e. other suitable semiconducting polymer may be used.

[0073] In various embodiments, the biocompatible ionic liquid includes a cation and an anion. In various embodiments, the cation may comprise tris(2-hydroxyethyl)methylammonium [MTEOA], 2-hydroxyethyl-trimethylammonium, choline, cholinium, sodium (Na), potassium (K), calcium (Ca), magnesium (Mg),
5 diethylammonium, triethylammonium, an ammonium-based cation, a phosphonium-based cation, an imidazolium-based cation, or a derivative thereof, or a combination thereof. In various embodiments, the anion may comprise methylsulfate [MeOSO₃], sulfate, lactate, acetate, malate, chloride (Cl), dihydrogen phosphate, bitartrate, dihydrogen citrate, levulinate, or a derivative thereof, or a combination thereof.
10 Aforesaid cations and anions are not intended to be limiting, i.e. other suitable cations and anions may be used.

[0074] In various embodiments, the biocompatible ionic liquid and the semiconducting polymer may have a non-limiting weight ratio ranging from 1:100 to 10:1 or any ranges within this specified range. Apart from this weight ratio, other
15 suitable weigh ratios can also be employed.

[0075] The present disclosure provides for a method of forming the biocompatible composite described in various embodiments of the first aspect. The method may comprise providing a semiconducting polymer having an electrically conductive domain and an electrically insulating domain coupled to the electrically conductive
20 domain, forming an aqueous solution comprising the biocompatible ionic liquid, and mixing the semiconducting polymer and the aqueous solution. The mixing of the semiconducting polymer and the aqueous solution may involve forming a biocompatible film. Said differently, the method may involve forming a biocompatible film from the semiconducting polymer and the aqueous solution in
25 various embodiments. Example 2B further hereinbelow describes the method in more detail. Embodiments and advantages described for the biocompatible composite of the first aspect can be analogously valid for the present method subsequently described herein, and vice versa. Where the various embodiments and advantages have already been described above and in the examples section further hereinbelow, they shall not
30 be iterated for brevity.

[0076] In certain non-limitng embodiments, providing the semiconducting polymer may comprise having the semiconducting polymer formed as a film by annealing and

then being post-treated with an aqueous solution comprising the biocompatible ionic liquid. Post-treating of the semiconducting polymer film with the aqueous solution may be carried out by, as non-limiting examples, casting, blading or coating, the aqueous solution comprising the biocompatible ionic liquid on the semiconducting polymer film. Alternatively, the semiconducting polymer film may be submerged into the aqueous solution comprising the biocompatible ionic liquid or be subject to an atmosphere formed from the aqueous solution comprising the biocompatible ionic liquid.

[0077] In various embodiments, forming the aqueous solution comprising the biocompatible ionic liquid may comprise diluting or dissolving the biocompatible ionic liquid in an aqueous medium. In certain non-limiting embodiments, the biocompatible ionic liquid may be directly used without any treatment.

[0078] In various embodiments, mixing the semiconducting polymer and the aqueous solution may be carried out for at least 10 minutes.

[0079] In various embodiments, the method optionally further comprises adding an organosilane to the semiconducting polymer, or during or after mixing the semiconducting polymer and the aqueous solution.

[0080] In various non-limiting embodiments, treating the semiconducting polymer with the biocompatible ionic liquid in the presence of the organosilane or mixing the semiconducting polymer and the aqueous solution in the presence of the organosilane, may be carried out for at least 10 mins (for instance, at a temperature ranging from 0°C to 100°C, 10°C to 90°C, 20°C to 80°C, etc). In various non-limiting embodiments, (i) mixing the organosilane with the semiconducting polymer may be carried out for at least 10 mins (for instance, at a temperature ranging from 0°C to 100°C, 10°C to 90°C, 20°C to 80°C, etc) or (ii) mixing the semiconducting polymer and the aqueous solution in the presence of the organosilane is carried out for at least 10 mins (for instance, at a temperature ranging from 0°C to 100°C, 10°C to 90°C, 20°C to 80°C, etc).

[0081] The present disclosure also relates to an organic electrochemical transistor (OECT) comprising a source electrode and a drain electrode, and the biocompatible composite described in various embodiments of the first aspect configured as an active layer between and in contact with both the source electrode and the drain

electrode. Embodiments and advantages described for the biocompatible composite of the first aspect can be analogously valid for the present OECT subsequently described herein, and vice versa. Where the various embodiments and advantages have already been described above and in the examples section further hereinbelow, they shall not
5 be iterated for brevity.

[0082] In various embodiments, the organic electrochemical transistor may further comprise an electrolyte arranged in contact with the active layer. The electrolyte may be a liquid solution and sealed by a well or can be incorporated into a microfluidics configuration, wherein the liquid electrolyte containing dissolved soluble salts in a
10 polar solvent, such as water. In certain non-limiting embodiments, the electrolyte may be a solid-state electrolyte, wherein the solid-state electrolyte has an ionic conductive polymer and an ionic liquid incorporated into the ionic conductive polymer.

[0083] In various non-limiting embodiments, the OECT may be fabricated by a two-layered parylene C process (see example 2C for more details). The bottom layer of
15 parylene C may be retained in the OECT. This parylene C may cover most parts of the device except the channel area and an end of the source and/or drain electrodes that are distal from the channel and the electrolyte. In such non-limiting embodiments where the OECT includes a liquid electrolyte (e.g. where the electrolyte is an aqueous electrolyte), an additional polydimethylsiloxane (PDMS) may be attached onto the
20 OECT in a manner that holds and seals the electrolyte, circumventing any leakage of a liquid electrolyte.

[0084] In various embodiments, the source electrode and the drain electrode may be configured on a substrate.

[0085] In various embodiments, the substrate may be optionally derived from a
25 flexible or rigid material. The flexible material may comprise polyimide, polyethylene terephthalate, parylene C, polydimethylsiloxane, cellulose, polyacrylonitrile, polylactic acid, or a derivative thereof, or a combination thereof. The rigid material may comprise silicon oxide, glass, quartz, metal foil, polymers, or a derivative thereof, or a combination thereof. The substrate may include a flexible and a rigid
30 material.

[0086] In various embodiments, the organic electrochemical transistor may further comprise a gate electrode arranged in contact with the electrolyte.

[0087] The present disclosure provides for a method of forming the organic electrochemical transistor. FIG. 10 is a schematic diagram illustrating the method and is intended as a non-limiting example. Example 2C further hereinbelow describes the method in more detail. Embodiments and advantages described for the biocompatible composite of the first aspect and the present organic electrochemical transistor can be analogously valid for the present method subsequently described herein, and vice versa. Where the various embodiments and advantages have already been described above and in the examples section further hereinbelow, they shall not be iterated for brevity.

10 [0088] The method may comprise providing a substrate, forming a source electrode and a drain electrode on the substrate (e.g. by photolithography and/or metal deposition), forming a channel between the source electrode and the drain electrode after forming the source electrode and the drain electrode for subsequent formation of an active layer (e.g. by photolithography and dry etching), forming a first insulating layer and a second insulating layer on the first insulating layer to cover portions of the source electrode and the drain electrode not covered by the active layer, forming the biocompatible composite described in various embodiments of the first aspect as the active layer between and in contact with both the source electrode and the drain electrode; and removing the second insulating layer and retaining the first insulating layer to cover portions of the source electrode and the drain electrode not covered by the active layer. In this method, the first insulating layer does not cover the active layer. In various embodiments, the present method may include a step of forming two layers of parylene C through step-by-step deposition (see FIG. 10 and example 2C for more details). In various embodiments, there may be one or more channels formed between the source and drain electrodes.

25 [0089] In various embodiments, forming the channel may comprise providing a substrate with deposited electrodes. In various embodiments, forming the channel may comprise depositing the first insulating layer (e.g. parylene C) on the substrate over the source electrode and the drain electrode, treating the substrate having the first insulating layer with a soap solution, depositing a second insulating layer (e.g. parylene C) on the first insulating layer, depositing a photoresist on the second insulating layer, developing the photoresist to form a patterned photoresist layer, and

etching the second insulating layer and the first insulating layer with the patterned photoresist layer to form the channel for subsequent formation of the active layer. The dimension and position of the channel may be configured through photolithography (depending on where the mask is positioned, refer to FIG. 10) and dry etching.

5 [0090] In various embodiments, forming the biocompatible composite as the active layer comprises depositing (e.g. coating, casting, or printing) the biocompatible composite in the channel, annealing the substrate having the biocompatible composite thereon, tearing off the second insulating layer, rinsing the substrate having the biocompatible composite thereon, and drying the substrate having the biocompatible composite therein with an inert gas. The biocompatible composite may be deposited
10 by, as non-limiting examples, spin-coating, spray-coating, drop-casting, screen printing, and/or inkjet printing at the one or more channels.

[0091] In various embodiments, the method may further comprise arranging an electrolyte on the active layer, wherein the electrolyte is in contact with the active
15 layer. In various non-limiting embodiments, the electrolyte may be a liquid or solid electrolyte. The liquid electrolyte may be formed by casting while the solid electrolyte formed by spin-coating on the top of the channel.

[0092] In various embodiments, the method may further comprise forming a gate electrode in contact with the electrolyte.

20 [0093] The present disclosure relates to a wearable device operable as a pressure sensor. Embodiments and advantages described for the biocompatible composite of the first aspect and the OECT can be analogously valid for the present wearable device subsequently described herein, and vice versa. Where the various embodiments and advantages have already been described above and in the examples section further
25 hereinbelow, they shall not be iterated for brevity.

[0094] In various embodiments, the wearable device has a sensing component comprising at least two electrodes configured on a substrate, and the biocompatible composite described in various embodiments of the first aspect configured as a sensing layer on and in contact with the at least two electrodes. In various non-
30 limiting embodiments, with respect to the sensing component, the sensing component may be integrated with the OECT, and the OECT may be operable as an amplifier.

[0095] In various embodiments, the sensing component may comprise a flexible polymer layer having protruding structures configured to face the electrodes.

[0096] In various embodiments, the biocompatible composite can be a layer formed at a surface of the flexible polymer layer which the protruding structures reside.

5 [0097] The present disclosure relates to a method of forming the wearable device. Embodiments and advantages described for the biocompatible composite of the first aspect and the wearable device can be analogously valid for the present method subsequently described herein, and vice versa. Where the various embodiments and advantages have already been described above and in the examples section further
10 hereinbelow, they shall not be iterated for brevity.

[0098] In various embodiments, the method may comprise providing a substrate having at least two electrodes, forming a sensing component comprising the biocompatible composite described in various embodiments of the first aspect configured as a sensing layer on and in contact with the at least two electrodes, and
15 integrating the sensing component with the organic electrochemical transistor.

[0099] In various embodiments, forming the sensing component may comprise forming a flexible polymer layer having protruding structures from a mold, and forming the biocompatible composite as a layer (e.g. by at least a method selected from the group consisting of spin-coating, drop-casting and dip-coating) at a surface
20 of the flexible polymer layer which the protruding structures reside.

[00100] The present disclosure further provides for an electrocardiogram recording device. Embodiments and advantages described for the biocompatible composite of the first aspect and the OEECT can be analogously valid for the present electrocardiogram recording device subsequently described herein, and vice versa.

25 Where the various embodiments and advantages have already been described above and in the examples section further hereinbelow, they shall not be iterated for brevity.

[00101] The electrocardiogram recording device may comprise the organic electrochemical transistor, and two or more testing electrodes attachable to a human skin surface (see FIG. 9A), wherein at least one of the two or more testing electrodes
30 is connected to a gate electrode of the organic electrochemical transistor and at least another of the two or more testing electrodes is connected to a source electrode of the organic electrochemical transistor.

[00102] In various embodiments, the organic electrochemical transistor is attachable to the human skin surface or placed at a probe-able position distal from the heart. For example, the OECT may be attached to the hand of a human as a non-limiting example.

5 [00103] In various embodiments, one of the two or more testing electrodes connected to the gate electrode is attached to the human skin surface proximal to the heart and in contact with the gate electrode. In various embodiments, at least another of the two or more testing electrodes connected to the source electrode is attachable to the human skin surface which is distal from the heart (e.g. hand of the human) and in contact
10 with the source electrode.

[00104] In various embodiments, each of the two or more testing electrodes may comprise silver, a silver-based material, an electrode gel, a metal, a metal-based material, carbon, a carbon-based material, a conductive polymer, a conductive hydrogel, or a derivative thereof, or a combination thereof. That is to say, each of the
15 two or more testing electrodes may be a silver electrode, a silver-based electrode (e.g. AgCl), an electrode gel, a metal electrode, a metal-based electrode, a carbon electrode, a carbon-based electrode, a conductive polymer electrode, a conductive hydrogel electrode, or an electrode having a combination of such materials. In certain non-limiting examples, the two or more testing electrode may have a pair of
20 electrodes, wherein one is formed of electrode gel, the other is formed of silver chloride.

[00105] The word “substantially” does not exclude “completely” e.g. a composition which is “substantially free” from Y may be completely free from Y. Where necessary, the word “substantially” may be omitted from the definition of the present
25 disclosure.

[00106] In the context of various embodiments, the articles “a”, “an” and “the” as used with regard to a feature or element include a reference to one or more of the features or elements.

[00107] In the context of various embodiments, the term “about” or “approximately”
30 as applied to a numeric value encompasses the exact value and a reasonable variance.

[00108] As used herein, the term “and/or” includes any and all combinations of one or more of the associated listed items.

[00109] Unless specified otherwise, the terms "comprising" and "comprise", and grammatical variants thereof, are intended to represent "open" or "inclusive" language such that they include recited elements but also permit inclusion of additional, unrecited elements.

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Examples

[00110] The present disclosure relates to a biocompatible composite, which includes a biocompatible ionic liquid. The present disclosure also relates to applications of the biocompatible composite, particularly demonstrating tris(2-hydroxyethyl)methylammonium methylsulfate ([MTEOA][MeOSO₃]) in PEDOT:PSS as a non-limiting example, and its use in the construction of organic electrochemical transistors (OECT) and wearable electronic devices such as for human physiological signals monitoring.

[00111] The present biocompatible composite, its method of forming and uses are described in further details, by way of non-limiting examples, as set forth below.

[00112] Example 1: General Introduction to Examples

[00113] Several traditional studies have been done to uncover the PEDOT core in PEDOT:PSS structure to improve its conductivity, by involving surfactants, solvents, acids, etc. However, in the area of OECTs, it is difficult to predict whether the traditional methods are useful enough to improve the performance of PEDOT:PSS based OECTs, only a few studies have been completed on improving the PEDOT:PSS based channel. In one example, D-sorbitol was added into PEDOT:PSS to render OECTs with high transconductance and fast response. In another example, using PEDOT:PSS processed by different dimethyl sulfoxide (DMSO) treatment methods as channels of OECTs, and the effects of DMSO treatment methods on the resultant PEDOT:PSS based OECT performance were studied. According to another example, electronic and ionic transport of PEDOT:PSS and the resultant OECT performance may be tuned and improved by involving ethylene glycol (EG) in PEDOT:PSS. Having said the above, it is to be first understood that ionic liquids belong to a unique category of non-volatile organic/inorganic salts with low flammability, negligible vapor pressure, and good chemical stability. Ionic liquid doping has been investigated herein as an effective method to enhance the performance of PEDOT:PSS based

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OECTs. PEDOT:PSS based OECTs with fast response, high transconductance, and high ion sensitivity are enabled by doping PEDOT:PSS with 1-ethyl-3-methylimidazolium tricyanomethanide ([EMIM][TCM]) as demonstrated herein. However, the biocompatibility or cytotoxicity of most traditional ionic liquids may not have been investigated and may be potentially toxic, which limits their utilization in biological applications. Furthermore, cytotoxicity studies of PEDOT:PSS-ionic liquid blends are extremely necessary towards the development of biocompatible OECTs.

[00114] The present disclosure and examples herein demonstrate use of a biocompatible ionic liquid, for example, tris(2-hydroxyethyl)-methyl-ammonium methylsulfate ([MTEOA][MeOSO₃]), as a performance enhancer for PEDOT:PSS based OECTs. That said, the present disclosure and examples particularly demonstrates use of [MTEOA][MeOSO₃] to mix with PEDOT:PSS and its effect on the characteristics of PEDOT:PSS and the OECTs performance.

[00115] In the examples, the influence of [MTEOA][MeOSO₃] on the morphology and conductivity of PEDOT:PSS and the redox processes of PEDOT:PSS films with [MTEOA][MeOSO₃] (herein denoted as PEDOT:PSS/[MTEOA][MeOSO₃]) films are investigated. Cytotoxicity studies have also been conducted since direct contact with biological interfaces are required when monitoring physiological signals on human skin. When using PEDOT:PSS/[MTEOA][MeOSO₃] film as the active channel layer, the OECTs were found to exhibit highly improved performance, including large transconductance, low energy consumption, and fast response. PEDOT:PSS/[MTEOA][MeOSO₃] film was also demonstrated as a sensing component of pressure sensor devices, wherein a wearable device with gate bias tunable pressure sensitivity can be obtained by the integration of the pressure sensor with an OECT. Using this integrated wearable electronic device, human physiological signal monitoring is demonstrated, paving the way for high-performing, fully biocompatible and green wearable electronic devices.

[00116] Example 2A: Materials

[00117] PEDOT:PSS (Clevios PH1000) was purchased from Heraeus and filtered prior to use through a 0.45 μm filter. [MTEOA][MeOSO₃], ethylene glycol (EG), 4-dodecylbenzenesulfonic acid (DBSA) and glycidoxypolytrimethoxysilane (GOPS)

were purchased from Sigma Aldrich, polydimethylsiloxane (PDMS) (Sylgard 184) was obtained from Dow Corning Corp., and Parylene C dimer (DPX-C) was supplied by Specialty Coating Systems Inc.

[00118] Example 2B: Film Preparation

5 **[00119]** PEDOT:PSS/[MTEOA][MeOSO₃] films were prepared by mixing PEDOT:PSS with [MTEOA][MeOSO₃] (diluted in deionized water prior to mix) and stirred vigorously for more than 10 mins. 0.5 wt.% of GOPS was added to the mixing solution and stirred. Pristine PEDOT:PSS film mixed with 0.5 wt.% GOPS and PEDOT:PSS film mixed with 5 wt.% of EG plus 0.025 wt.% of DBSA plus 0.5 wt.%
10 of GOPS were also prepared. The thicknesses of all the resultant films were measured using an Alpha-Step profilometer (KLA-Tencor).

[00120] Example 2C: Fabrication and Measurement of OECTs

[00121] PEDOT:PSS/[MTEOA][MeOSO₃] based OECTs were fabricated by the patterning method of two-layers of Parylene C (see FIG. 10 – schematic in bottom
15 row). Briefly, the gold source and drain electrodes were patterned with the assistance of photoresist AZ 5214 onto silicon substrates (coated with silicon dioxide). After the source and drain electrodes are formed, the first insulating parylene C layer (thickness: 1.9 μm) was deposited with the adhesion promoter A174 (3-(trimethoxysilyl)propyl methacrylate). Then the substrate was subsequently spin-
20 coated with a dilute soap solution (10% Micro-90 in water). Next, the substrate was coated with the second insulating parylene C layer (thickness: 2.5 μm). Afterward, a layer of photoresist AZ P4620 was cast onto the two parylene C layers covered substrate and developed with AZ developer. Lastly, reactive ion etching (RIE) was utilized to expose the channels of OECTs. The parylene C patterned substrate was
25 then treated with ultraviolet-ozone cleaner for another 5 mins. The PEDOT:PSS/[MTEOA][MeOSO₃] solutions were spin-coated onto the parylene C patterned substrates, followed by annealing at 140 °C for 15 mins, rinsing with DI water and drying with nitrogen. After that, the sacrificial layers (the second parylene C layers) were torn off and removed.

30 **[00122]** For a flexible OECT, the same steps were applied except that a layer of parylene C was deposited on the silicon substrate and then peeled off as the flexible substrate. The gold source and drain electrodes were evaporated on the layer of

Parylene C through a shadow mask. Then PEDOT:PSS/[MTEOA][MeOSO₃] solution was spray-coated as the active channel layer through another shadow mask.

[00123] All the electrical characteristics of the OECTs were measured at room temperature using a probe station (Karl Suss PM5) and a Keysight precision source/measure unit (B2900A Series). A 0.1 M NaCl solution was prepared as the electrolyte, and an Ag/AgCl pellet acted as the gate electrode and was immersed in the 0.1M NaCl electrolyte for standard performance measurement.

[00124] Example 2D: Fabrication of Pressure Sensor and Wearable Device

[00125] A schematic representation of the pressure sensor and the wearable device are shown in FIG. 5A and 5B. For the wearable device, a flexible PDMS layer with micro-pyramids was patterned from the silicon mold. Then the PEDOT:PSS/[MTEOA][MeOSO₃] film was spin-coated on this PDMS layer and annealed for 15 mins at 140°C as the sensing component. Disconnected Au electrodes were deposited on a polyimide film, which was chosen as the substrate of the pressure sensor. Then the patterned PEDOT:PSS/[MTEOA][MeOSO₃] film with micro-pyramids was laminated on the disconnected electrodes to constitute the pressure sensor. This pressure sensor was then connected in series with the aforementioned OECT to form a wearable device with gate voltage tunable sensitivity. A force tester (ESM 303, Mark-10 Corporation) with a force gauge (Mark-10 025/012) was chosen to apply different external pressures and dynamic pressures for the pressure measurement.

[00126] Example 2E: Electrocardiography (ECG) recording

[00127] For the designed flexible and biocompatible OECT, a layer of Parylene C was deposited on the silicon substrate and then peeled off as the flexible substrate. The gold source and drain electrodes were evaporated on the layer of Parylene C through a shadow mask. Then PEDOT:PSS/[MTEOA][MeOSO₃] solution was spray-coated as the active channel layer through another shadow mask. Electrolyte for this designed OECT was prepared by mixing PVA with [MTEOA][MeOSO₃] in a weight ratio of 2:3 and covered onto the channel area by spin-coating at 1500 rpm for 30s. PEDOT:PSS-coated Au was used as the gate electrode. For the bending test, a flexible and biocompatible OECT was attached to one of the metal molds with different bending radii (R = 20, 15, 10, 5 mm). Output and transfer characteristics of the OECT

device before and after bending were recorded by a Keysight B2912A semiconductor analyzer under ambient conditions.

[00128] The drain current of the OECT was measured for the ECG signal recording. The flexible and biocompatible OECT and an Ag/AgCl gel electrode (Red dot, 3M) were attached to the right hand, and another Ag/AgCl gel electrode was placed at the chest of the volunteer. An effective voltage between two Ag/AgCl gel electrodes was enabled due to the potential difference between the hand and the heart of the volunteer. The drain voltage ($V_{ds} = -0.5V$) was supplied by the Keysight precision source/measure unit (B2900A Series). All signals were filtered by a band-pass filter at 25 Hz to remove the power supply noise. The measurements were approved by Institutional Review Board at Nanyang Technological University (Approval number: IRB-2021-144).

[00129] Example 2F: Conductivity and Morphology Characterization

[00130] The conductivities of all films were measured by a four-point probe station (AIT, CMT-SR2000N), with at least 10 regions measured and averaged. The film morphology was recorded using Atomic force microscopy (AFM, Cypher ES) in tapping mode. Scanning electron microscope measurement (SEM, JSM-IT100, JEOL) was performed to demonstrate the micro-pyramid (a square base of 10 μm and a height of 8 μm) arrays of the PDMS sensing component with and without PEDOT:PSS/[MTEOA][MeOSO₃] film. Raman spectra of PEDOT:PSS/[MTEOA][MeOSO₃] films were measured in the range of 900 to 1700 cm^{-1} using a Raman spectrometer (Witech alpha 300) with a 532nm excitation wavelength.

[00131] Example 2G: Grazing-Incidence Wide-Angle X-ray Scattering (GIWAXS)

[00132] The samples were prepared by spin-coating the PEDOT:PSS and PEDOT:PSS/[MTEOA][MeOSO₃] solutions on silicon substrates and annealing at 140°C for 15 mins. Then all the substrates were rinsed by DI water and dried using nitrogen. GIWAXS on thin films were carried out in vacuum (0.1 mbar) using Xenocs Nanoinxider with Cu-K α microsource (40 μm), 30W and 100K detector. An incidence angle of 0.2° was employed. Raw scattering data was processed and analyzed using Foxtrot software.

[00133] Example 2H: Cytotoxicity

[00134] Both the control and sample (PEDOT:PSS/[MTEOA][MeOSO₃]) substrates were pre-loaded into 12 well-plate. Then, 1 x 10⁴ human dermal fibroblasts were deposited into each well and cultured with high glucose DMEM at 37°C and 5% CO₂ with 95% humidity. At determined time points (2, 4 and 6 days), live/dead staining was performed to evaluate the biocompatibility of substrates. Briefly, the staining solution was prepared by diluting Hoechst 33342 for cell nucleus staining (life technology, H3570), Calcein-AM for live cells staining (Thermo C3099, Ex/Em 494/517 nm) and EthD-1 for dead cells staining (Thermo E1169, Ex/Em 528/617 nm) 1000 times in serum-free medium. Then, 1 mL staining solution was added into each well and incubated for 30 min. At the end of the incubation, the cells were imaged using Zeiss Observer Z1. The dead control of cells was prepared via treating cells with 0.2% Triton X-100 for 30 mins. The obtained pictures were processed using an ImageJ software. The number of live (green) and dead (red) cells were counted in which the cells' viability can be calculated based on following Equation:

$$Cell Viability = \frac{N_{live}}{N_{live} + N_{dead}} \times 100\%$$

[00135] wherein N_{live} and N_{dead} denote the number of green and red cells, respectively.

[00136] Example 2I: Mobility measurement

[00137] Measuring the source-drain current (I_{ds}) change in response to applied various gate current (I_{gs}) when applying a constant source -drain bias (V_{ds}) of -0.1 V. The mobility can be calculated by the equation below:

$$[00138] \quad \mu = \frac{dI_{ds}}{dt} * \frac{L^2}{V_{ds} * I_{gs}}$$

[00139] where dI_{ds}/dt is the slope of source-drain current (I_{ds}) versus time (t), L is the OECT channel length in cm. The product of mobility and V_{ds}/L^2 can be extracted by fitting the slope of dI_{ds}/dt versus I_{gs} , thus the mobility comes out of this product since V_{ds} and L are known. Eight individual devices were measured, and the reported value is the average value with standard deviation (error bar).

[00140] Example 2J: Electrochemical impedance spectroscopy (EIS)

[00141] Three electrodes based EIS measurement was conducted to extract the volumetric capacitance of the PEDOT:PSS/[MTEOA][MeOSO₃] film. PEDOT:PSS/[MTEOA][MeOSO₃] films were spin coated on the Au coated silicon substrates as the working electrode. Ag/AgCl and Pt wire work as reference electrode and counter electrode, respectively. 0.1 M NaCl solution took the role of electrolyte, and all EIS spectra were obtained by Metrohm Autolab Nova 2.1.3 at the frequency range between 0.1 ~ 105 Hz. An equivalent circuit model composed of a parallel capacitor (C_p), a parallel resistor (R_p), and a serial resistor (R_s) was used to analyze the EIS data. All extracted C_p values were plotted versus the nominal volume (the product of the film area A and thickness d) of the PEDOT:PSS/[MTEOA][MeOSO₃] film. Thus, the volumetric capacitance C* of PEDOT:PSS/[MTEOA][MeOSO₃] film was estimated from the slope of the fitting line.

[00142] Example 2K: Ultraviolet-Visible-Near-Infrared (UV-vis-NIR) Spectroelectrochemistry

[00143] The pristine PEDOT:PSS and PEDOT:PSS/[MTEOA][MeOSO₃] mixtures were coated on ITO substrates and then annealed for 15 mins at 140°C. After that, care was taken to confine an 8 mm × 5 mm active area. The PEDOT:PSS-coated ITO glass was immersed into a cuvette of 0.1 M NaCl solution along with Ag/AgCl pellet as the gate electrode. A Keysight precision source/measure unit (B2912A) was utilized to apply voltage bias between the Ag/AgCl pellet and the ITO glass substrate. Absorption spectra of the above films was captured using a UV-vis-NIR spectrophotometer over the wavelength range from 400 nm to 1100 nm (SHIMADZU, UV-3600). The same spectrophotometer was used record the absorption spectra of the pristine and composite films without any biasing.

[00144] Example 3A: Results and Discussion – Conductivity, UV-vis-NIR and Raman Characterization

[00145] FIG. 1A presents the chemical structures of the semiconducting polymer PEDOT:PSS and the ionic liquid [MTEOA][MeOSO₃], with a schematic of the ion exchange between the oppositely charged ions. As shown in FIG. 1B, the pristine PEDOT:PSS (without ionic liquid) exhibits a conductivity of $0.028 \pm 0.0015 \text{ S cm}^{-1}$, which is consistent with reported values. When the concentration of the added ionic liquid exceeds 5.2 wt.%, the conductivities of the resultant composite films reach as

high as $>1000 \text{ S cm}^{-1}$, which is more than four orders of magnitude higher than that of pristine PEDOT:PSS. Although the PEDOT:PSS with higher concentrations of [MTEOA][MeOSO₃] ($> 3.8 \text{ wt.}\%$) showed higher conductivity ($>625 \text{ S cm}^{-1}$), the composite solutions become more viscous to be solution-processed. Hence in this study, 3.5 wt.% of [MTEOA][MeOSO₃] in PEDOT:PSS was selected for subsequent studies.

[00146] FIG. 1C shows the ultraviolet-visible-near-infrared (UV-vis-NIR) absorption spectra of pristine PEDOT:PSS and composite PEDOT:PSS/[MTEOA][MeOSO₃] films. Firstly, films without applying any bias were plotted in FIG. 1C. According to reported examples, weak polaronic and strong bipolaronic transitions of PEDOT appear at $\sim 900 \text{ nm}$ and $\geq 1200 \text{ nm}$ wavelengths, respectively. For the PEDOT:PSS/[MTEOA][MeOSO₃] film, a noticeable increase in the polaronic transition of PEDOT is observed while the bipolaronic absorption decreases as compared to pristine PEDOT:PSS film, implying a higher PEDOT doping level. This result correlates well with the highly enhanced conductivity and reflects the counter ion exchange between PEDOT:PSS and [MTEOA][MeOSO₃].

[00147] The morphological transition is revealed by Raman spectroscopy, as shown in FIG. 1D, the peaks at 1430 cm^{-1} in the spectra of the pristine PEDOT:PSS film and 1425 cm^{-1} in the spectra of film modified with [MTEOA][MeOSO₃] correspond to the C_α=C_β symmetric stretching vibrations of PEDOT chains. The red shift (from 1430 cm^{-1} to 1425 cm^{-1}) detected in the PEDOT:PSS/[MTEOA][MeOSO₃] film as compared to the pristine film indicates the PEDOT chains structure changed from a benzoid to a quinoid structure. The formation of nanofibrillar and quinoid structure of PEDOT chains make it easier to have conductive paths between PEDOT chains, due to which conductivity is significantly improved.

[00148] Example 3B: Results and Discussion – Cytotoxicity Studies

[00149] Furthermore, cytotoxicity studies are carried out to test the suitability of PEDOT:PSS/[MTEOA][MeOSO₃] based devices for wearable electronics. Briefly, human dermal fibroblasts were co-cultured with substrates and the growth of the cells were monitored for 6 days. Glass substrates without any film were used as control plates. As shown in FIG. 2A, the morphologies of the cells were recorded at intervals of two days, revealing healthy cell growth on PEDOT:PSS/[MTEOA][MeOSO₃]

films, without any sign of detachment, contaminant, or dead cell. Moreover, the cell viability of the PEDOT:PSS/[MTEOA][MeOSO₃] film towards human dermal fibroblasts cells at 2, 4 and 6 days, was found to be higher than 98.5%, 99.7%, 99.8%, respectively, as depicted in FIG. 2B. These results demonstrate that
 5 PEDOT:PSS/[MTEOA][MeOSO₃] film shows no toxicity to human dermal fibroblasts cells, confirming its suitability for biocompatible and green wearable devices.

[00150] Example 3C: Results and Discussion – OECT Performance

[00151] FIG. 3A presents a schematic diagram of the OECT, where the PEDOT:PSS/[MTEOA][MeOSO₃] film acts as an active channel layer between the
 10 source and drain electrodes and is gated with 0.1 M NaCl based aqueous electrolyte. FIG. 3B and 3C show the typical output and transfer characteristics respectively, with channel dimensions of width (*W*): 100 μm, length (*L*): 10 μm, and thickness (*d*): 69 nm. The OECTs operate in depletion mode, with high drain current (*I_{ds}*) under zero gate bias (*V_{gs}*), representing the doped state of PEDOT. When a positive gate bias is
 15 applied, the Na⁺ ions from the aqueous electrolyte penetrate the active channel layer, reducing the doping or oxidation state of PEDOT. Notably, transconductance normalized to channel geometry [*g_m*/(*Wd/L*)] for PEDOT:PSS/[MTEOA][MeOSO₃] based OECTs is 22.3 ± 4.5 mS μm⁻¹, which is much higher than the published biocompatible additives-treated PEDOT:PSS based OECTs (FIG. 3K). Furthermore,
 20 the product of the mobility (*μ*) and the volumetric capacitance (*C**) (the *μC** product) is recognized to be independent of the OECT channel geometry and captures the electronic and ionic transport properties of the active channel layer at steady-state. The *μC** product of PEDOT:PSS/[MTEOA][MeOSO₃] film is evaluated to be 283.80 ± 29.66 F cm⁻¹ V⁻¹ s⁻¹ (as shown in FIG. 3D and 3K), which is higher than the widely-
 25 used ethylene glycol (EG)-treated PEDOT:PSS based OECTs (75 ± 51 or 100 ± 7 F cm⁻¹ V⁻¹ s⁻¹), which compares favorably to reported works of NaCl gated OECTs.

[00152] The mobility and volumetric capacitance are also measured independently. The mobility (*μ*) of pristine PEDOT:PSS films are previously reported as 0.01~0.06 cm²V⁻¹s⁻¹. Here, the mobility of the PEDOT:PSS/[MTEOA][MeOSO₃] films
 30 determined by driving the OECTs with varying constant gate currents (*I_{gs}*) was found to be 1.659 ± 1.131 cm²V⁻¹s⁻¹ (FIG. 3E). The improved mobility of PEDOT:PSS/[MTEOA][MeOSO₃] film indicates a more efficient hole transport

pathway. Next, the volumetric capacitance (C^*) of OECTs which account for the ability to allow ion penetration was looked into. Three electrodes based electrochemical impedance spectroscopy (EIS) measurement was conducted to extract the C^* value of the PEDOT:PSS/[MTEOA][MeOSO₃] film. An equivalent circuit model (Randles circuit) composed of a parallel capacitor (C_p), a parallel resistor (R_p), and a serial resistor (R_s) was used to analyze the EIS data, as shown in FIG. 3F. All extracted C_p values were plotted against the nominal volume (the product of the film width and length, namely area: A and thickness: d) as shown in FIG. 3G. Here, the C^* of PEDOT:PSS/[MTEOA][MeOSO₃] film was estimated as $75.87 \pm 6.15 \text{ F cm}^{-3}$, significantly higher than that of the widely-used EG-treated PEDOT:PSS film (31~39 F cm^{-3}).

[00153] Besides, the PEDOT:PSS/[MTEOA][MeOSO₃] based OECTs also demonstrate fast transient response times (τ) of $\sim 40.57 \mu\text{s}$, as determined from the single exponential fit illustrated in FIG. 3H. The transient response time (τ) is known to be dependent on channel geometry, as shown in FIG. 3I and 3L. When normalized to channel geometry, the slope of the transient response off time versus the parameter $\pi[d (WL)^{1/2}]$ is calculated to be $28.1 \pm 3.0 \mu\text{s}/\mu\text{m}^2$ for PEDOT:PSS/[MTEOA][MeOSO₃] based OECTs. The obtained value of normalized response time is comparable to that of the widely-used EG-treated PEDOT:PSS based OECTs (see FIG. 3L). In addition, the PEDOT:PSS/[MTEOA][MeOSO₃] based OECT demonstrates excellent switching cyclical stability, where the drain current retained a very high retention of 95% after 5000 cycles (FIG. 3J). Moreover, many high performing organic transistors demand high operating voltages ($>5\text{V}$), which induce thermal degradation and is undesirable for portable applications. Favorably, PEDOT:PSS/[MTEOA][MeOSO₃] based OECTs display very low operating voltages ($<1 \text{ V}$) befitting biological applications.

[00154] Example 3D: Results and Discussion – Morphology and Redox Properties

[00155] Atomic force microscopy (AFM) and Raman spectroscopy were further conducted to investigate the structural alteration. FIG. 4A, 4B and 4C are the phase images of the pristine PEDOT:PSS film and film modified with [MTEOA][MeOSO₃], respectively. The brighter part of the images is assigned to PEDOT chains, while the

darker part corresponds to PSS chains. The phase image of the pristine film exposes granular morphology, while the ionic liquid modified film reveals nanofibrillar morphology of PEDOT chains. The topological images of pristine PEDOT:PSS film and PEDOT:PSS/[MTEOA][MeOSO₃] film are demonstrated in FIG. 4D, 4E and 4F, where the PEDOT:PSS/[MTEOA][MeOSO₃] film presents a rougher surface with a higher root-mean-square (RMS) roughness (2.434 nm) as compared to the pristine film (RMS roughness = 0.960 nm).

[00156] The redox processes of the PEDOT:PSS/[MTEOA][MeOSO₃] films were then examined using UV-vis-NIR spectra while immersed in an electrolyte (0.1 M NaCl) under biasing condition. UV-vis-NIR spectra of pristine PEDOT:PSS film was also recorded for comparison of results. FIG. 4G and 4H display the absorption spectra of the film modified with [MTEOA][MeOSO₃] and the pristine PEDOT:PSS film, respectively, at different applied voltages between -0.4 V and 0.8 V. As shown in FIG. 4G and 4H, the setup for UV-vis-NIR spectra measurement is demonstrated in the insert, the absorptions at 645 nm (arising from π - π^* transition of de-doped or neutral PEDOT) are found to be increased for both films with positive voltage applied progressively. Such an increase in absorption of π - π^* transition implies effective de-doping processes where PEDOT is reduced from bi-polaron (PEDOT²⁺) to polaron (PEDOT⁺) and neutral PEDOT state, as inferred from previous reports. Furthermore, the absorptions at 970 nm (attributed to the polaronic PEDOT moiety) are found to be increased when applying lower voltage biases (≤ 0.4 V) for both pristine PEDOT:PSS and PEDOT:PSS/[MTEOA][MeOSO₃] films due to reduction of bi-polaron PEDOT to polaron PEDOT. The absorptions at 970 nm are found to be gradually decreased for both films at higher voltage bias (> 0.4 V), indicating the reduction of polaron PEDOT to neutral PEDOT. At 0.8 V, the film with [MTEOA][MeOSO₃] shows a lower absorption at 970 nm as compared to the pristine PEDOT:PSS film. This indicates that the [MTEOA][MeOSO₃] treated PEDOT film was de-doped more thoroughly with higher ion penetration ability.

[00157] The morphological changes induced by the molecular rearrangement were further studied using Grazing-Incidence Wide-Angle X-ray Scattering (GIWAXS) measurements. As shown in FIG. 4I, the broad peaks at 1.79 Å⁻¹ of the pristine PEDOT:PSS film and 1.83 Å⁻¹ of the film modified with [MTEOA][MeOSO₃] are

assigned to the π - π stacking of PEDOT chains [(010) reflection of PEDOT chains], while another broad peak at $0.58 \text{ \AA}^{-1}$ of the pristine PEDOT:PSS film and $0.49 \text{ \AA}^{-1}$ of PEDOT:PSS/[MTEOA][MeOSO₃] film represents the (200) reflection of PEDOT chains. The d-spacing (200) for PEDOT of the film modified with [MTEOA][MeOSO₃] increases as compared to the pristine film, while the d-spacing (010) for PEDOT chains of PEDOT:PSS/[MTEOA][MeOSO₃] film decreases from 3.51 to 3.43 Å. This indicates that PEDOT chains rearrange to closer π - π stacking and improved ordering structure with the addition of [MTEOA][MeOSO₃]. Higher molecular ordering of PEDOT chains in PEDOT:PSS/[MTEOA][MeOSO₃] film therefore leads to form a more effective conductive network between PEDOT chains, resulting in observed enhanced conductivity, which is also consistent with other study.

[00158] Example 4: Wearable Devices Consisting of Pressure Sensor Using PEDOT:PSS/[MTEOA][MeOSO₃] Films as the Sensing Component

[00159] To demonstrate multiple applications of the PEDOT:PSS/[MTEOA][MeOSO₃] film, this film was used to act as the sensing component of a pressure sensor and the active channel layer of an OECT, respectively, and a wearable electronic device was fabricated by integrating these electronic components. Schematic representation of the pressure sensor and the wearable device are depicted in FIG. 5A and 4B. For the pressure sensor, micro-pyramid arrays of the PEDOT:PSS/[MTEOA][MeOSO₃] film is patterned using a silicon mold which acts as the pressure sensing layer. A square base of 10 μm and a height of 8 μm are demonstrated in each of these micro-pyramids (as shown in FIG. 6A to 6F). FIG. 5C and 5D show the transfer and output curves of the OECT, which is in series with the pressure sensor. The drain current changes obviously corresponding to different pressures, implying a successful pressure stimuli detection.

[00160] As indicated in FIG. 7A and 7B, the integrated circuit consisting of the pressure sensor and OECT shows excellent tunable pressure sensitivity by gate bias. Furthermore, it is demonstrated to monitor the human physiological signal on human skin as wearable electronic devices as shown in FIG. 7C to 7F. FIG. 7C demonstrates the monitoring of human wrist pulse, and FIG. 7D to 7F show the record of the corresponding current change ratios when the pressure sensor is assembled on the human finger, wrist, and knee, respectively. As shown in FIG. 7C, the pressure sensor

attached to the human wrist would generate signals corresponding to wrist pulse simultaneously, achieving the monitoring of subtle human motions. Besides, monitoring of large human physiological signals can also be realized. Multiangle bending (0° , 30° , 60° , 90°) of a finger is recorded in real-time as depicted in FIG. 7D.

5 The current change ratios gradually increase to 200%, 400% and 600% respectively in real-time when the finger is bent stepwise from the straight state to 30° , 60° and 90° . The current change ratio at the same bending angle is maintained constant and the current change ratio returns to its original status immediately when the finger is restored to its straight state, producing stable and accurate current change ratios. This
10 integrated circuit can also be utilized to monitor the extending and flexing of the wrist and knee as can be seen in FIG. 7E and 7F. Current change ratios with good repeatability and stability are achieved during the cyclic extending and flexing process when the wrist and knee are bent from 0 to 90° . More importantly, the sensitivity of our pressure sensor can be tuned by the gate voltage, thus the appropriate gate voltage
15 can be selected according to the actual needs, thus reducing power consumption. All these results indicate that this gate voltage tunable pressure sensor enabled by an integrated OECT and pressure sensing component is a promising element as a wearable electronic device to detect full-range human physiological signals.

**[00161] Example 5: Flexible OECTs using PEDOT:PSS/[MTEOA][MeOSO₃]
20 films as the Active Layer under Deformation:**

[00162] Next, a flexible and biocompatible OECT is designed as shown in FIG. 8A (details of the preparation are provided in the Experimental Section). Firstly, bending test were conducted to investigate the robustness against physical deformation (FIG. 8A). The performance of the OECT device was investigated where output and transfer characteristics were measured before being bent, as shown in FIG. 8B and 8C,
25 respectively. The normalized transconductance (g_m) of the flexible and biocompatible OECT without being bent was calculated as $16.95 \text{ mS } \mu\text{m}^{-1}$, although this value is slightly lower than the device prepared using Si Substrate ($22.3 \pm 4.5 \text{ mS } \mu\text{m}^{-1}$), it is still higher than the most reported biocompatible additives-treated PEDOT:PSS based
30 OECTs (FIG. 3K), reflecting its excellent signal transconductance property. The diversity between electrolyte and electrodes should account for the device performance difference. Furthermore, the OECT performance stability towards

deformation was investigated after being bent at different radii, which is also can be seen in FIG. 8B and (d). Overall, stable OEET performance was maintained when bending at different radii ($R = 20, 15, 10, 5$ mm), with high retention of the on-current ($>90\%$) and corresponding peak transconductance ($>85\%$) after bending at R as low as 5 mm. Importantly, it is worth noting that the device performance is nearly fully recovered with high retention of the on-current ($\sim 99\%$) and corresponding peak transconductance ($\sim 99\%$) after the deflation was removed, implying its commendable stability towards deformation.

[00163] Example 6: Electrocardiography (ECG) Signals Recording

[00164] In order to further demonstrate the practical on-skin application of the designed flexible and biocompatible PEDOT:PSS/[MTEOA][MeOSO₃] based OEET, the electrocardiography (ECG) signals recording ability was explored (FIG. 9A). A flexible and biocompatible OEET with the maximum transconductance (~ 18.5 mS) at $V_{gs} = 0$ V is chosen (gate voltage can be applied to obtain the maximum g_m for higher signal delivery capability, here, only one bias is applied to the drain electrode since this OEET shows maximum transconductance at $V_{gs} = 0$ V). The ECG signal was recorded from the heart of a volunteer. This flexible and biocompatible PEDOT:PSS/[MTEOA][MeOSO₃] OEET and one Ag/AgCl electrode were attached on to the skin of right hand, while another Ag/AgCl electrode was placed on the chest (near the heart), respectively. The QRS complex peaks reflect the ventricles depolarization. The normal ventricles atrial depolarization and repolarization correspond to the P and T peaks, respectively. FIG. 9B shows one of the representative peaks of the recorded ECG signal where the QRS, P, and T peaks are well recognized. The QRS complex peak corresponds to a high current change of ~ 3 μ A, indicating distinguishable signals. Also, stable and continuous ECG signals are well recorded, as shown in FIG. 9C. Thus, on-skin application of flexible and biocompatible PEDOT:PSS/[MTEOA][MeOSO₃] device is successfully demonstrated as an on-skin ECG signals recording device and shows great potential for clinical application.

[00165] Example 7: Summary

[00166] In present disclosure and aforesaid examples, the ionic liquid [MTEOA][MeOSO₃] was explored the use of [MTEOA][MeOSO₃] as an enhancer to

improve the conductivity of PEDOT:PSS, the performance of OECTs and its effects on the morphology and redox properties of PEDOT:PSS films. OECTs fabricated with PEDOT:PSS/[MTEOA][MeOSO₃] films as the active layers show large transconductance ($22.3 \pm 4.5 \text{ mS } \mu\text{m}^{-1}$), a high μC^* product ($283.80 \pm 29.66 \text{ F cm}^{-1} \text{ V}^{-1} \text{ s}^{-1}$) and fast response times ($\sim 40.57 \text{ } \mu\text{s}$). The device also demonstrated excellent switching cyclical stability (95% retention after 5000 cycles). Next, the ion sensors consisting of the PEDOT:PSS/[MTEOA][MeOSO₃] based OECT and ion-selective membrane show ions (Na^+ and K^+) sensing capability with high sensitivity and selectivity. This device could be effectively used to detect ions in the physiological range for human vitals monitoring. In addition, flexible and biocompatible OECTs were designed and found to retain stable performances under different bending radii (R), even when the R is as low as 5 mm. The PEDOT:PSS/[MTEOA][MeOSO₃] based flexible and biocompatible OECT enables an on-skin ECG signal recording device with high signal quality. Overall, this study details the effects of the biocompatible ionic liquid [MTEOA][MeOSO₃] on the electrical and structural characteristics of PEDOT:PSS and confirms that it is a promising candidate to develop biocompatible, high-performing, and on-skin electronic devices.

[00167] From the above, apparently, the present disclosure relates to enhanced transistors with higher transconductance, fast response time, excellent switching cycling stability, and low energy consumption. The examples already demonstrate on cytotoxicity experiment of the biocompatible ionic liquid tris(2-hydroxyethyl)methylammonium methylsulfate ([MTEOA][MeOSO₃]) doped PEDOT:PSS film, applicable in biocompatible organic electrochemical transistors (OECTs) using the biocompatible ionic liquid [MTEOA][MeOSO₃] doped PEDOT:PSS film as the active layer, applicable in biocompatible pressure sensors using the biocompatible ionic liquid [MTEOA][MeOSO₃] doped PEDOT:PSS film as the sensing component and in wearable device using OECTs and pressure sensors having the biocompatible ionic liquid [MTEOA][MeOSO₃] doped PEDOT:PSS film.

[00168] The wearable device shows advantageous tunable pressure sensitivity by gate bias and is demonstrated to be capable of monitoring physiological signals on human skin. The sensitivity of it can be tuned by the gate voltage; thus, reducing power

consumption when selecting the appropriate gate voltage according to the actual needs.

[00169] The PEDOT:PSS/[MTEOA][MeOSO₃] film also shows robust resistance to physical deformation and desired compatibility with flexible substrates as the active channel layer of the flexible OECT. On-skin application of flexible and biocompatible PEDOT:PSS/[MTEOA][MeOSO₃] device is successfully demonstrated as an on-skin ECG signals recording device and shows great potential for clinical application.

[00170] **Example 8: Commercial and Potential Applications**

[00171] The development of OECTs for wearable electronics demands a set of unique properties such as biological compatibility, higher transconductance, fast response time, excellent switching cycling stability and resistance to deformation. The present disclosure provides a biocompatible ionic liquid having significant advantages in commercialization to fabricate biocompatible OECTs with the above-mentioned characteristics, paving the way for high-performing, fully biocompatible and on-skin wearable electronic devices.

[00172] As mentioned above, organic electrochemical transistors (OECTs) have attracted attention in recent years due to their high transconductance, low operating voltage, and compatibility with aqueous solutions for broad biosensing applications. Especially, OECT-based wearable electronic devices enable the monitoring of physiological signals on human skin. Poly-3,4-ethylenedioxythiophene:poly-4-styrenesulfonate (PEDOT:PSS) may be the material acting as the active channel layer in OECTs. However, the poor electrical conductivity of pristine PEDOT:PSS diminish the lifetime and restrict the application scope of wearable systems. Although some traditional ionic liquids in PEDOT:PSS have been investigated to enhance the conductivity, such traditional ionic liquids are not proven to be non-toxic or biocompatible. Herein, a biocompatible ionic liquid [MTEOA][MeOSO₃] is demonstrated as a biocompatible additive to enhance the performance of PEDOT:PSS based OECTs. The influence of [MTEOA][MeOSO₃] on the morphology, conductivity, and the redox process of PEDOT:PSS during electrochemical doping/de-doping processes are explored. The biocompatible PEDOT:PSS/[MTEOA][MeOSO₃] film (PEDOT:PSS mixed with [MTEOA][MeOSO₃]) as OECT channel layer exhibits enhanced transistor

characteristics, including higher transconductance, fast response time, excellent switching cycling stability, and low energy consumption. Further, the PEDOT:PSS/[MTEOA][MeOSO₃] film is employed as the sensing component of a pressure sensor, and the integration of the pressure sensor with the OECT shows
5 interesting tunable pressure sensitivity by gate bias and is demonstrated to monitor physiological signals on human skin as a wearable electronic device. In addition, the PEDOT:PSS/[MTEOA][MeOSO₃] film also shows robust resistance to physical deformation and desired compatibility with flexible substrates as the active channel layer of the flexible OECT, and the on-skin application of flexible and biocompatible
10 PEDOT:PSS/[MTEOA][MeOSO₃] device is successfully demonstrated as an on-skin ECG signals recording device and shows great potential for clinical application.

[00173] While the present disclosure has been particularly shown and described with reference to specific embodiments, it should be understood by those skilled in the art that various changes in form and detail may be made therein without departing from
15 the spirit and scope of the present disclosure as defined by the appended claims. The scope of the present disclosure is thus indicated by the appended claims and all changes which come within the meaning and range of equivalency of the claims are therefore intended to be embraced.

CLAIMS

1. A biocompatible composite comprising:
 - a semiconducting polymer having an electrically conductive domain and an electrically insulating domain coupled to the electrically conductive domain; and
 - a biocompatible ionic liquid doped in the semiconducting polymer,wherein the biocompatible ionic liquid renders a portion of the electrically insulating domain distal from the electrically conductive domain to expose a portion of the electrically conductive domain, and
 - wherein the semiconducting polymer comprises poly-3,4-ethylenedioxythiophene:poly-4-styrenesulfonate and the ionic liquid comprises tris(2-hydroxyethyl)methylammonium methylsulfate.
2. The composite of claim 1, wherein the biocompatible ionic liquid and the semiconducting polymer have a weight ratio ranging from 1:100 to 10:1.
3. A method of forming the biocompatible composite of claim 1 or 2, the method comprising:
 - providing a semiconducting polymer having an electrically conductive domain and an electrically insulating domain coupled to the electrically conductive domain;
 - forming an aqueous solution comprising the biocompatible ionic liquid; and
 - mixing the semiconducting polymer and the aqueous solution.
4. The method of claim 3, wherein forming the aqueous solution comprising the biocompatible ionic liquid comprises diluting or dissolving the biocompatible ionic liquid in an aqueous medium.
5. The method of claim 3 or 4, wherein mixing the semiconducting polymer and the aqueous solution is carried out for at least 10 minutes.

6. The method of any one of claims 3 to 5, further comprising adding an organosilane to:
the semiconducting polymer; or
during or after mixing the semiconducting polymer and the aqueous solution.
7. The method of claim 6, wherein (i) mixing the organosilane with the semiconducting polymer is carried out for at least 10 mins or (ii) mixing the semiconducting polymer and the aqueous solution in the presence of the organosilane is carried out for at least 10 mins.
8. An organic electrochemical transistor comprising:
a source electrode and a drain electrode; and
the biocompatible composite of claim 1 or 2 configured as an active layer between and in contact with both the source electrode and the drain electrode.
9. The organic electrochemical transistor of claim 8, further comprising an electrolyte arranged in contact with the active layer.
10. The organic electrochemical transistor of claim 8 or 9, wherein the source electrode and the drain electrode are configured on a substrate.
11. The organic electrochemical transistor of claim 10,
wherein the substrate is derived from a flexible or rigid material,
wherein the flexible material comprises polyimide, polyethylene terephthalate, parylene C, polydimethylsiloxane, cellulose, polyacrylonitrile, polylactic acid, or a derivative thereof, or a combination thereof, and
wherein the rigid material comprises silicon oxide, glass, quartz, metal foil, polymers, or a derivative thereof, or a combination thereof.
12. The organic electrochemical transistor of any one of claims 8 to 11, further comprising a gate electrode arranged in contact with the electrolyte.

13. A method of forming the organic electrochemical transistor of any one of claims 8 to 12, the method comprising:

- providing a substrate;
- forming a source electrode and a drain electrode on the substrate;
- forming a channel between the source electrode and the drain electrode after forming the source electrode and the drain electrode for subsequent formation of an active layer;
- forming a first insulating layer and a second insulating layer on the first insulating layer to cover portions of the source electrode and the drain electrode not covered by the active layer;
- forming the biocompatible composite of claim 1 or 2 as the active layer between and in contact with both the source electrode and the drain electrode; and
- removing the second insulating layer and retaining a first insulating layer to cover portions of the source electrode and the drain electrode not covered by the active layer.

14. The method of claim 13, wherein forming the channel comprises:

- depositing the first insulating layer on the substrate over the source electrode and the drain electrode;
- treating the substrate having the first insulating layer with a soap solution;
- depositing a second insulating layer on the first insulating layer;
- depositing a photoresist on the second insulating layer;
- developing the photoresist to form a patterned photoresist layer; and
- etching the second insulating layer and the first insulating layer with the patterned photoresist layer to form the channel for subsequent formation of the active layer.

15. The method of claim 13 or 14, wherein forming the biocompatible composite as the active layer comprises:

- depositing the biocompatible composite in the channel;
- annealing the substrate having the biocompatible composite thereon;
- tearing off the second insulating layer;

rinsing the substrate having the biocompatible composite thereon; and
drying the substrate having the biocompatible composite therein with an inert gas.

16. The method of any one of claims 13 to 15, further comprising arranging an electrolyte on the active layer, wherein the electrolyte is in contact with the active layer.

17. The method of claim 16, further comprising forming a gate electrode in contact with the electrolyte.

18. A wearable device operable as a pressure sensor, the wearable device having a sensing component comprising:

at least two electrodes configured on a substrate; and

the biocompatible composite of claim 1 or 2 configured as a sensing layer on and in contact with the at least two electrodes.

19. The wearable device of claim 18, wherein the sensing component comprises a flexible polymer layer having protruding structures configured to face the electrodes.

20. The wearable device of claim 19, wherein the biocompatible composite is a layer formed at a surface of the flexible polymer layer which the protruding structures reside.

21. A method of forming the wearable device of any one of claims 18 to 20, the method comprising:

providing a substrate having at least two electrodes;

forming a sensing component comprising the biocompatible composite of claim 1 or 2 configured as a sensing layer on and in contact with the at least two electrodes; and

integrating the sensing component with an organic electrochemical transistor of any one of claims 8 to 12.

22. The method of claim 21, wherein forming the sensing component comprises:
forming a flexible polymer layer having protruding structures from a mold;
and
forming the biocompatible composite as a layer at a surface of the flexible polymer layer which the protruding structures reside.
23. An electrocardiogram recording device comprising:
an organic electrochemical transistor of any one of claims 8 to 12; and
two or more testing electrodes attachable to a human skin surface,
wherein at least one of the two or more testing electrodes is connected to a gate electrode of the organic electrochemical transistor and at least another of the two or more testing electrodes is connected to a source electrode of the organic electrochemical transistor.
24. The electrocardiogram recording device of claim 23, wherein the organic electrochemical transistor is attachable to the human skin surface or placed at a probable position distal from the heart.
25. The electrocardiogram recording device of claim 23 or 24, wherein one of the two or more testing electrodes connected to the gate electrode is attached to the human skin surface proximal to the heart and in contact with the gate electrode.
26. The electrocardiogram recording device of any one of claims 23 to 25, wherein the at least another of the two or more testing electrodes connected to the source electrode is attachable to the human skin surface which is distal from the heart and in contact with the source electrode.
27. The electrocardiogram recording device of any one of claims 23 to 26, wherein each of the two or more testing electrodes comprises silver, a silver-based material, an electrode gel, a metal, a metal-based material, carbon, a carbon-based

material, a conductive polymer, a conductive hydrogel, or a derivative thereof, or a combination thereof.