



(51) International Patent Classification:

H01L 41/18 (2006.01) H01L 41/35 (2013.01)

H01L 41/113 (2006.01) H01L 51/42 (2006.01)

H02N 2/18 (2006.01)

(21) International Application Number:

PCT/EP2020/053631

(22) International Filing Date:

12 February 2020 (12.02.2020)

(25) Filing Language:

English

(26) Publication Language:

English

(30) Priority Data:

19156606.6 12 February 2019 (12.02.2019) EP

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(81) Designated States (unless otherwise indicated, for every

kind of national protection available): AE, AG, AL, AM,

AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CZ, DE, DJ, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IR, IS, JO, JP, KE, KG, KH, KN, KP, KR, KW, KZ, LA, LC, LK, LR, LS, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PA, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SA, SC, SD, SE, SG, SK, SL, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, WS, ZA, ZM, ZW.

(84) Designated States (unless otherwise indicated, for every

kind of regional protection available): ARIPO (BW, GH, GM, KE, LR, LS, MW, MZ, NA, RW, SD, SL, ST, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU, TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, KM, ML, MR, NE, SN, TD, TG).

Published:

— with international search report (Art. 21(3))

(54) Title: DEVICE COMPRISING PIEZOELECTRICALLY ACTIVE LAYER AND METHOD FOR PRODUCING SUCH DEVICE

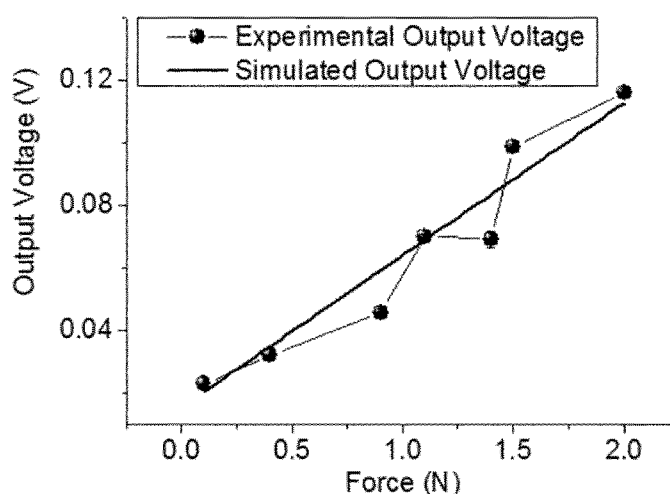


Figure 8

(57) Abstract: The present application relates to an electronic device comprising a piezoelectrically active composite layer as well as a method for producing such electronic device. The present application also relates to an electronic apparatus comprising such electronic device.

DEVICE COMPRISING PIEZOELECTRICALLY ACTIVE LAYER AND METHOD FOR PRODUCING SUCH DEVICE

5 **Technical Field**

The present application relates to an electronic device comprising a piezoelectrically active composite layer as well as a method for producing such electronic device. The present application also relates to an electronic apparatus
10 comprising such electronic device.

Background

15 Traditional approaches for electrical energy sources either employ batteries with limited energy reservoir, electromagnetic induction such as a potentially bulky dynamo, the photovoltaic effect requiring a light source, or the thermoelectric effect requiring moderate to high temperature gradients. Energy harvesting via the piezoelectric effect offers many attractive benefits unmatched by traditional
20 approaches, including: (i) continual operation in the absence of light, temperature or electromagnetism; (ii) comparatively simpler assembly than traditional power sources, and (iii) potentially higher power densities.

The piezoelectric effect refers to the ability of certain materials to effectively
25 convert mechanical energy into electricity when put under a mechanical stress. Conversely, placing these materials in strong electric fields leads to structural deformation, which makes them important materials useful either as (micro-) power source or sensors for a multitude of applications, such as touch screen displays, force sensors, active strain sensors, light sensors, energy harvesting,
30 ultrasound transducer imaging, wearable electronics, human-machine interface modules, virtual reality and artificial reality, bio-electronic implants, pressure monitoring systems, remote sensors, smart homes and cities and assets, structural health monitoring, next generation industrial automation etc..

35 Natural semiconducting piezoelectric materials like zinc oxide (ZnO), gallium nitride (GaN), indium nitride (InN), indium phosphide (InP), silicon carbide (SiC), or

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molybdenum di-sulphide (Mo_xS_x) are gaining widespread popularity for low cost applications. These can be fabricated using low temperature methods such as hydrothermal reaction methods on temperature sensitive substrates or high temperature fabrication methods where they are subsequently transferred to a receiver substrate for device assembly at lower temperatures. These materials, however, generally show inferior charge generation capabilities.

On the other hand, bulk inorganic piezoelectric materials, such as lead zirconate titanate (PZT) and its derivatives; barium titanate (BTO), quartz, or lithium niobate (LNbOx) generally demonstrate significantly higher charge generation capabilities. The application scope for bulk piezoelectric materials, particularly in energy harvesting applications, is however limited because: (i) they are both bulky and rigid, leading to high failure rates beyond their normal operating limits, (ii) their brittleness also limits their use in applications where truly flexible modules may be required, such as in fabrics, (iii) the best piezoelectric materials often contain elemental lead, potentially making them unsuitable for bio-electronic implants, and (iv) they require extremely high temperature processing, which limits their use on truly flexible surfaces such as fabrics and polymeric surfaces.

Whilst mechanically conformal piezoelectric polymers (e.g. PVDF and its derivatives) alleviate some of these limitations, their application fields are however limited because they may also be temperature sensitive. It is also known that even modest temperature fluctuations can degrade their charge generation capabilities via the direct piezoelectric effect (or visa verse). Moreover, high mechanical impact excitations may rapidly degrade their piezoelectric properties.

So far piezoelectric sensors are only able to detect dynamic forces/pressures/stresses and as such, are impractical for detecting static force/pressure/stress due to electrostatic screening and/or charge leakage effects.

Thus, there is a need for a more versatile device showing piezoelectric characteristics, which is useful for piezoelectric sensors and (micro-)power sources, which is lead-free and non-toxic, available at low costs with simple processing steps, highly flexible, non-brittle, temperature stable in its operating environment and which has high reliability and lifetime. In addition, the need to enhance the charge-generation capabilities, dynamic force sensing capabilities as well as static force

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sensing capabilities of such a device is of both practical and technological importance.

5 It is therefore an object of the present invention to provide an electronic device that fulfils one or more of the above-outlined needs and overcomes the drawbacks typifying the known bulk piezoelectric ceramics and piezoelectric polymers, and that is well suited for the application as (micro-)power source or transducer sensor. In particular, it is an object of the present invention to provide a piezoelectric electronic device that offers non-toxic elements, high mechanical flexibility and
10 good temperature stability, high reliability and lifetime, self-generating capabilities under dynamic force excitations, dynamic force sensing capabilities and static force sensing capabilities, with simple processing steps.

15 Additional objects of the present application become evident from the following description as well as from the examples.

Summary

20 The present inventors have now surprisingly found that the above objects may be attained either individually or in any combination by the present electronic device and its method of manufacture.

The present application therefore provides for an electronic device comprising
25 (i) a first electrode layer,
(ii) a second electrode layer,
(iii) a piezoelectrically active composite layer between the first electrode and the second electrode, the piezoelectrically active composite layer comprising
30 (iii-1) a plurality of piezoelectrically active nanowires on a base layer of the same material, and
(iii-2) one or more organic semiconducting layer dispersed in between the nanowires, each organic semiconducting layer comprising one or more organic semiconducting materials,
35 and

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- (iv) an electrically insulating top layer between the piezoelectrically active composite layer and the second electrode.

5 The present application therefore also provides for a method of manufacturing such electronic device, said method comprising the steps of

- (a) providing the first electrode layer,
- (b) forming the piezoelectrically active composite layer by
 - (i) forming on the first electrode layer the nanowire base layer and then a plurality of piezoelectrically active nanowires onto said
 - 10 base layer, and
 - (ii) depositing one or more organic semiconducting material onto said piezoelectrically active nanowires, to obtain the piezoelectrically active composite layer,
- (c) subsequently depositing one or more electrically insulating material, to
- 15 obtain the electrically insulating top layer, and
- (d) then deposit the second electrode layer.

20 The present application also provides for an electronic apparatus comprising such electronic device.

The present application further provides for the use of such electronic device as one selected from the group consisting of piezoelectric sensor, photo/light sensor, force/pressure sensor, vibration sensor, and energy harvester.

25 Furthermore, the present application provides for a method of using such electronic device as one selected from the group consisting of piezoelectric sensor, photo/light sensor, force/pressure sensor, vibration sensor, and energy harvester.

30 **Brief description of the drawings**

Figure 1 shows a generalized exemplary schematic representation of an electronic device as disclosed in the present application comprising an optional substrate 10, a first electrode layer 20, a nanowire base layer 30, a plurality of piezoelectrically

35 active nanowires 40, an organic semiconducting layer 50, a dielectric top layer 60, and a second electrode 70.

5 **Figure 2** shows a generalized exemplary schematic representation of an electronic device as disclosed in the present application, the electronic device further comprising a buffer layer between the nanowire base layer and the organic semiconducting layer.

10 **Figure 3** shows a generalized exemplary schematic representation of an electronic device as disclosed in the present application, the electronic device comprising voids between the nanowire base layer and the organic semiconducting layer.

15 **Figure 4** shows a generalized exemplary schematic representation of an electronic device as disclosed in the present application, the electronic device comprising a buffer layer between the nanowire base layer and the organic semiconducting layer, and voids between the nanowire base layer and the buffer layer.

20 **Figure 5a, 5b and 5c** show exemplary schematic cross-sectional views theoretically showing the operation principle (Figure 5a) before applying a force, (Figure 5b) with a force applied, and (Figure 5c) after removing the force.

25 **Figure 6** shows an SEM image of the semi-finished device of Example 1 in (Figure 6a) top view, and (Figure 6b) in cross-sectional view.

30 **Figure 7** shows an SEM image of the semi-finished device of Example 2 in (Figure 7a) top view, and in (Figure 7b) cross-sectional view.

35 **Figure 8** shows the output voltage of the device of Example 4 in relation to the force applied.

Figure 9 shows the current of the device of Example 4 under incremental static force loading conditions.

Figure 10 shows the output voltage of the device of Example 4 under dynamic impact excitation conditions.

Figure 11 shows output voltage and current of the device of Example 4 under dynamic impact excitation conditions and under parallelly configured electrical resistance loading conditions.

5 **Figure 12** shows peak-peak output voltage and current for the device of Example 8 in comparison to a device produced as in Example 5 or Example 6

It is noted that throughout this application, identical reference numbers in the drawings indicate identical or corresponding or similar features.

10

Detailed description

15 As used herein, the term "organic semiconducting layer" denotes a semiconducting layer, in which the total semiconducting material, i.e. including any inorganic semiconducting material if present, comprises at least 50 wt% of organic semiconducting material, preferably at least 60 wt% (e.g. 70 wt% or 80 wt% or 90 wt% or 95 wt% or 97 wt% or 99.0 wt% or 99.5 wt% or 99.7 wt% or 99.9 wt%), with wt% relative to the total weight of all semiconducting material comprised in the semiconducting layer, and most preferably consists of one or more organic semiconducting material.

20 As used herein, an asterisk "*" denotes a linkage to an adjacent unit or group, including for example, in case of a polymer, to an adjacent repeating unit or any other group. In some instances, where specifically identified as such, the asterisk "*" may also denote a mono-valent chemical group.

25 As used herein, the terms "n-type" or "n-type semiconductor" or "n-type semiconducting material" will be understood to mean an extrinsic semiconductor or semiconducting material in which the conduction electron density is in excess of the mobile hole density, and the term "p-type" or "p-type semiconductor" or "p-type semiconducting material" will be understood to mean an extrinsic semiconductor or semiconducting material in which mobile hole density is in excess of the conduction electron density (see also J. Thewlis, Concise Dictionary of Physics, Pergamon Press, Oxford, 1973).

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- As used herein, the term “organyl group” is used to denote any organic substituent group, regardless of functional type, having one free valence at a carbon atom (see also International Union of Pure and Applied Chemistry, Compendium of Chemical Terminology, Gold Book, Version 2.3.3, 2012-02-24, page 1040).
- 5
- As used herein, the term “organoheteryl group” is used to denote any univalent group comprising carbon, said group thus being organic, but having the free valence at an atom other than carbon (see also International Union of Pure and Applied Chemistry, Compendium of Chemical Terminology, Gold Book, Version 2.3.3, 2012-02-24, page 1039).
- 10
- As used herein, the term “carbyl group” includes both, organyl groups and organoheteryl groups.
- 15
- As used herein, the term “hydrocarbyl group” is used to denote univalent groups formed by removing a hydrogen from a hydrocarbon (see also International Union of Pure and Applied Chemistry, Compendium of Chemical Terminology, Gold Book, Version 2.3.3, 2012-02-24, page 694).
- 20
- As used herein, the term “partially fluorinated” is used to denote that one or more hydrogen atoms of a compound, but not all, have been replaced with fluorine.
- As used herein, the terms “fully fluorinated” and “perfluorinated” are used to denote that all hydrogen atoms of a compound have been replaced with fluorine.
- 25
- Such fully fluorinated compounds may also be identified by the prefix “perfluoro”.
- As used herein, the term “monomer” is used to denote a substance composed of monomer molecules, and the term “monomer molecule” is used to denote a molecule which can undergo polymerization thereby contributing one or more constitutional units to the essential structure of a macromolecule or polymer (see also International Union of Pure and Applied Chemistry, Compendium of Chemical Terminology, Gold Book, Version 2.3.3, 2012-02-24, page 662).
- 30
- As used herein, the term “constitutional unit” is used to denote an atom or group of atoms (with pendant atoms or groups, if any) comprising a part of the essential structure of a macromolecule, an oligomer molecule, a block or a chain (see also
- 35

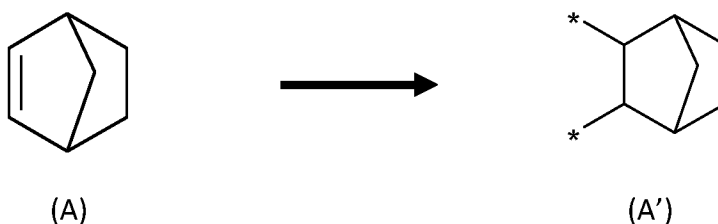
International Union of Pure and Applied Chemistry, Compendium of Chemical Terminology, Gold Book, Version 2.3.3, 2012-02-24, page 326).

As used herein, the term “homopolymer” is used to denote a polymer derived from one species of (real, implicit or hypothetical) monomer (see also International Union of Pure and Applied Chemistry, Compendium of Chemical Terminology, Gold Book, Version 2.3.3, 2012-02-24, page 685).

As used herein, the term “copolymer” is used to denote a polymer derived from more than one species of monomer (see also International Union of Pure and Applied Chemistry, Compendium of Chemical Terminology, Gold Book, Version 2.3.3, 2012-02-24, page 335).

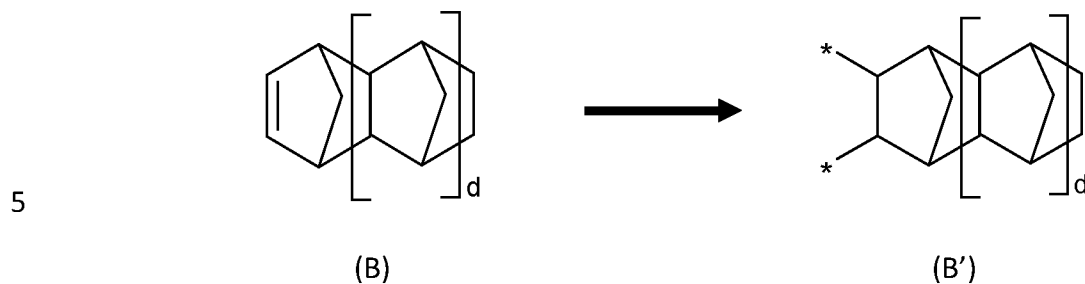
As used herein, the term “pendant group” is used to denote an offshoot, neither oligomeric nor polymeric from a chain, particularly from the backbone chain of a polymer (see also International Union of Pure and Applied Chemistry, Compendium of Chemical Terminology, Gold Book, Version 2.3.3, 2012-02-24, page 1076).

As used herein, the term “polynorbornene” is used to denote a polymer generally comprising norbornadiyl monomeric units of generalized and simplified formula (A') or derivatives thereof obtained by addition polymerization of a norbornene monomer of generalized and simplified formula (A) or derivatives thereof, in the simplest case bicyclo[2.2.1]hept-2-ene, to yield a polymer comprising a number of constitutional units (A') as schematically shown below



and for the purposes of the present application may also denote higher cyclic derivatives of norbornene (B) and the corresponding constitutional unit (B') as schematically shown below

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wherein d is an integer greater than 0, e.g. 1, 2, 3, etc..

10 As used herein, unless stated otherwise the molecular weight is given as the number average molecular weight M_n or weight average molecular weight M_w , which is determined by gel permeation chromatography (GPC) against polystyrene standards in eluent solvents such as tetrahydrofuran, trichloromethane (TCM, chloroform), chlorobenzene or 1,2,4-trichlorobenzene. Unless stated otherwise, chlorobenzene is used as solvent. The molecular weight distribution ("MWD"),

15 which may also be referred to as polydispersity index ("PDI"), of a polymer is defined as the ratio M_w/M_n . The degree of polymerization, also referred to as total number of repeat units, m , will be understood to mean the number average degree of polymerization given as $m = M_n/M_U$, wherein M_n is the number average molecular weight of the polymer and M_U is the molecular weight of the single repeat unit; see J.M.G. Cowie, *Polymers: Chemistry & Physics of Modern Materials*, Blackie, Glasgow, 1991.

20

As used herein, the terms "dielectric" and "electrically insulating" may be used interchangeably.

25

As used herein, the terms "consist of" and "consisting of" do not exclude the presence of impurities, which may normally be present, for example but in no way limited to, impurities resulting from the synthesis of a compound (e.g. an organic semiconducting material) or – in case of metals - trace metals.

30

As used herein, the term "optional" is used to indicate that a component or feature may be present but need not be present.

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In general terms the present application relates to an electronic device comprising a first electrode layer, a second electrode layer, a piezoelectrically active composite layer between the first electrode layer and the second electrode layer, and an electrically insulating top layer (or "dielectric top layer") between the piezoelectrically active composite layer and the second electrode.

The piezoelectrically active composite layer comprises a plurality of piezoelectrically active nanowires (which may also simply be referred to as "nanowires" or "micro-rods" throughout this application) on a nanowire base layer, and one or more organic semiconducting layer as defined herein.

A schematic representation of an exemplary device of the present application is shown in **Figure 1** comprising an optional substrate 10, a first electrode layer 20, a nanowire base layer 30, a plurality of piezoelectrically active nanowires 40, an organic semiconducting layer 50, a dielectric top layer 60, and a second electrode 70.

Preferably, the piezoelectrically active composite layer may further comprise a buffer layer, which is electrically insulating, between the nanowire base layer and the one or more semiconducting layer. A generalized schematic representation of an exemplary device of the present application comprising such a buffer layer 45 is shown in **Figure 2**.

ELECTRODE LAYERS

For the purposes of the present application the type of electrode material is not particularly limited. Suitable electrode materials for the first and second electrode layer include electrically conducting organic and inorganic materials, or blends thereof, with inorganic materials being preferred.

Exemplary organic electrode materials or blends include polyaniline, polypyrrole, poly(3,4-ethylenedioxythiophene) (PEDOT) or doped conjugated polymers, and dispersions or pastes of graphite. In a preferred embodiment, the organic electrode material is selected from a transparent conductive polymer.

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Suitable inorganic materials are preferably selected from metals and metal oxides including alloys and any blend of metals, any blend of metal oxides as well as any blend of metals and metal oxides.

5 Exemplary metals may, for example, be selected from Group 4, Group 6, Group 10, Group 11 and Group 12 of the periodic table of elements. Examples of metal that are particularly suitable as electrode materials for the first and second electrode layer may be selected from the group consisting of gold (Au), silver (Ag), copper (Cu), aluminum (Al), nickel (Ni), chromium (Cr), tungsten (W), zinc (Zn), palladium
10 (Pd), platinum (Pt), titanium (Ti), calcium (Ca), molybdenum (Mo), scandium (Sc), and any combination or blend of at least two thereof. Of these, gold, silver, platinum and palladium are particularly preferred. Exemplary alloys, which are particularly suitable as electrodes in organic electronic devices include stainless steel (e.g., 332 stainless steel, 316 stainless steel), alloys of gold, alloys of silver,
15 alloys of copper, alloys of aluminum, alloys of nickel, alloys of palladium, alloys of platinum, alloys of titanium, alloys of calcium, alloys of molybdenum and alloys of scandium.

Exemplary electrically conducting metal oxides include indium tin oxide (ITO),
20 fluorine-doped tin oxide, tin oxide, zinc oxide, indium zinc oxide, aluminum-doped zinc oxide, indium gallium zinc oxide (IGZO), and any blend thereof. Of these, ITO is especially preferred.

According to a preferred embodiment of the present invention, both the first
25 electrode layer and the second electrode layer each consist of a single layer comprising or preferably consisting of a metal or metal oxide, such as Au, Ag, Pd, Pt or ITO, including alloys and any blend of metals, any blend of metal oxides as well as any blend of metals and metal oxides.

30 According to another preferred embodiment of the present invention, one or both of the first electrode layer and the second electrode layer, preferably only the first electrode layer, may be provided as a bi-electrode layer composed of a first and a second conductive layer. Taking for example the first electrode layer, the first conductive layer may function as an adhesion layer for the second conductive layer,
35 which is disposed on the first conductive layer and disposed to directly contact the

base portion of plurality of piezoelectric nanowires and which may function as the “main electrode”.

5 PIEZOELECTRICALLY ACTIVE NANOWIRES AND NANOWIRE BASE LAYER

Preferably, the nanowire base layer is in direct physical contact with the respective surface of the first electrode layer. It is preferred that the nanowire base layer essentially covers the entire surface of the first electrode layer. Without wishing to
10 be bound by theory, it is believed that this helps in ensuring good and reliable performance of the present electronic device, potentially by the nanowire base layer serving as a barrier layer between the one or more organic semiconducting layer and the first electrode layer.

15 The nanowire base layer preferably has a thickness of at least 3 nm. It preferably has a thickness of at most 1 μm , more preferably at most 500 nm or 400 nm or 300 nm or 200 nm, even more preferably of at most 100 nm or 90 nm or 80 nm or 70 nm or 60 nm or 50 nm. In a preferred embodiment, the nanowire base layer is preferably deposited from a solution, e.g. an aqueous solution, comprising a soluble
20 zinc salt, such as zinc acetate or zinc nitrate. Without wishing to be bound by theory it is believed that upon deposition the solution comprising the zinc salt (for example, zinc acetate or zinc nitrate), essentially all of the zinc salt is decomposed to form zinc oxide.

25 As shown in **Figure 1**, the plurality of piezoelectrically active nanowires 40 protrudes directly from nanowire base layer 30. It is noted that, though indicated by different reference numbers, the nanowire base layer and the piezoelectrically active nanowires are of the same material.

30 The nanowire base layer may be continuous or discontinuous, which may also provide means for patterning the present device if needed. Preferably, the nanowire base layer is fused, i.e. forms a continuous layer throughout the present device, so that the nanowire base layer and the nanowires form a single unitary component or element of the present device, for example, also characterized by
35 being homogeneous with regards to material composition.

The protruding portions of the nanowires extending directly from the nanowire base layer are preferably oriented perpendicular with respect to the base layer and/or the first electrode layer so that on average the direction of the resulting piezoelectric polarization is oriented perpendicular to the first electrode layer and/or the second electrode layer. The skilled person understands that the protruding portions of the nanowires may not be disposed or grown so as to form an ideal right angle of 90° with respect to the other layers of the present electronic device, such as the nanowire base layer and/or the electrode layers. Therefore, as used herein, the term "substantially perpendicular" denotes that the orientation of the protruding portions of the plurality of nanowires extending directly from the base portion may deviate from the ideal 90.0° , that is, a deviating angle α indicating a deviation from an ideal perpendicular orientation of the protruding portions with respect to the base portion and/or the surface of the first electrode layer, where $\alpha = 0^\circ$, may be other than zero. Preferably, α is $\leq \pm 30^\circ$, more preferably $\leq \pm 20^\circ$, even more preferably $\leq \pm 15^\circ$. Particularly preferably, α is $\leq \pm 11^\circ$, and most preferably $\alpha = 0^\circ$. The deviations of the protruding portions of the plurality of piezoelectric nanowires from $\alpha = 0^\circ$ do not disturb the average piezoelectric polarization.

Preferably, at the interface between the piezoelectrically active nanowires and the one or more organic semiconducting layer p-n junctions are formed. Accordingly, the nanowires may be either of p-type or n-type. Thus, preferably, the nanowires comprise at least one semiconducting material having piezoelectric characteristics. Exemplary semiconducting piezoelectric materials may, for example, be selected from Group III-V and II-VI semiconductor compounds (referring to the periodic table of elements). Examples of suitable Group III-V and II-VI semiconductor compounds may be selected from zinc oxide (ZnO), gallium nitride (GaN), indium nitride (InN), indium phosphide (InP), indium antimonite (InSb), silicon carbide (SiC), molybdenum disulfide (MoS_2) and combinations of at least two thereof, whereby zinc oxide is preferred. The materials of the piezoelectric nanowires are, however, not limited to the above-mentioned materials.

As such, the present nanowires may be formed of an n-type semiconducting material or a p-type semiconducting material. For example, the organic semiconducting material layer may be formed of a p-type organic semiconducting material in case the nanowires are formed of an n-type semiconducting material. On the other hand, the organic semiconducting material layer may be formed of an

n-type organic semiconducting material in case the nanowires are formed of a p-type semiconducting material.

5 It is well documented that most inorganic semiconducting piezoelectric materials, such as metal oxides like zinc oxide (ZnO), exhibit n-type semiconductor characteristics, which greatly reduces the overall piezoelectric potential via electrostatic screening. Accordingly, piezoelectric materials having n-type semiconductor characteristics are preferably used according to the present invention. This requires that the organic semiconductor material has to be selected
10 such that a p-n-junctions are formed at the interfaces between the nanowires and the organic semiconducting material.

The plurality of nanowires disposed on the nanowire base layer are spaced apart from each other. Preferably, the distance between neighboring nanowires is at
15 least 0.01 μm , and more preferably at least 0.05 μm . Said distance is preferably at most 2.25 μm , and more preferably at most 2.0 μm . These distances can be determined by standard methods, such as for example by scanning electron microscopy (SEM).

20 Due to the presence of a plurality of distinct, i.e. well separated, nanowires there is open space (in the following referred to as "voids") in between the nanowires.

When depositing the organic semiconducting material or – if present – the buffer layer, e.g. in form of a formulation as defined herein, onto the plurality of nanowires
25 at least part, preferably substantially all, of the open space between the nanowires of the voids is filled with the organic semiconducting material or the dielectric material of the buffer layer. It is, however, not excluded that near the nanowire base layer 30 unfilled voids 35 remain, as is shown in the exemplary schematic representations in **Figures 3 and 4**, with the other reference numbers as already described earlier in respect to **Figures 1 and 2**. Such unfilled voids between the
30 nanowires allows the nanowires to be individually deformed.

With respect to the original open space between the nanowires (i.e. without any material been deposited into the open space), the remaining open space, i.e.
35 unfilled by any material, is at most 95 %, more preferably at most 90 % or 80 % or 70 %, or 60 % or 50 %, 40 % or 30 % or 20 % or 15 % or 10 %, relative to the original

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open space. It is most preferred that all of the original open space (as much as technically feasible) between the nanowires is filled with either a buffer layer material as defined below and/or a semiconducting material as defined below.

- 5 The thickness of the voids (i.e. the voids not filled by the buffer layer) formed between the base and the buffer layer, expressed as a proportion of the nanowire length, is at most less than 100 %, preferably at most 90 % or 80 % or 70 % or 60 % or 50 % or 40 % or 30 % or 20 % or 10 %. Most preferably, the thickness of the voids is 0 % of the length of the nanowires, i.e. there are no voids between base and
10 buffer layer or expressed differently, base and buffer layer are in direct physical contact to each other.

- Without wishing to be bound by theory it is believed that the voids being substantially completely filled offers a number of advantages, such as for example,
15 one or more of: a) that the organic semiconductor material can act as scaffold to the plurality of piezoelectric nanowires/protruding portions, b) the provision of an effective electronic interface layer for charge injection/extraction across the interface between the organic semiconductor material and the semiconducting piezoelectric material, c) depletion of excess free negative charge carriers from the
20 surface of the nanowires, d) effective separation of electron hole pairs under conditions of photo illumination and affords e) modulation of any existing space charge layer at the junction interface under electrical biasing conditions and/or with indeed piezo potential.

25
BUFFER LAYER

- Preferably, the buffer layer comprises, e.g. in at least 50 wt% or 60 wt% or 70 wt% or 80 wt% or 90 wt% or 95 wt% or 97 wt% or 99.0 wt% or 99.5 wt% or 99.7 wt% or
30 99.9 wt%, with wt% relative to the total weight of the buffer layer, and preferably consist of, one or more dielectric material as defined herein.

- Additionally, the buffer layer may also comprise other materials that may, for example, add additional functionalities or help in the deposition of the buffer layer.
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If present, the buffer layer is disposed onto the plurality of piezoelectric nanowires, such that it penetrates (or enters into) the network and covers an exposed surface of the base portion (i.e., the surface exposed between the protruding portions extending from the base portion). It is preferred according to the present invention that the buffer layer covers up to about 100 % (by area) of the exposed surface of the base portion of the plurality of piezoelectric nanowires. For example, from about 5 % to about 100 % (by area) of the exposed surface may be covered by the buffer layer. Preferably, the buffer layer covers at least 50 %, at least 60 %, or at least 70 %, more preferably at least 80 %, even more preferably at least 90 %, and most preferably about 100 % (by area, as far as technically possible) of the exposed surface of the base portion of the plurality of piezoelectric nanowires.

The thickness of the buffer layer can be expressed as a proportion of the nanowire length and preferably ranges from 0 % to 90 % (for example, 80 % or 70 % or 60 % or 50 % or 40 % or 30 % or 20 % or 10 %) of the length of the protruding nanowires.

The buffer layer may function to separate the base portion of the fused semiconducting piezoelectric material of the piezoelectric nanowires from the organic semiconducting material layer, which preferably comprises at least one p-type organic semiconductor material, as outlined above.

ORGANIC SEMICONDUCTING LAYER

The one or more organic semiconducting layer(s) each comprises one or more semiconducting material. Preferably, the organic semiconducting layer comprises only organic semiconducting materials (i.e. no inorganic semiconducting material).

The organic semiconductor material layer disposed on the plurality of piezoelectric nanowires comprises at least one organic semiconductor material, which may be selected from a monomeric compound, also referred to as "small molecule", from a polymeric compound or macromolecule, which will be understood to include oligomers, polymers and copolymers, and from a mixture, dispersion or blend containing one or more compounds selected from either or both of monomeric and polymeric compounds, as defined herein.

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Preferably, at least one organic semiconductor material is a p-type organic semiconductor material. Accordingly, it is preferred according to the present invention that the organic semiconductor material layer comprises at least one p-type organic semiconductor material and the piezoelectric nanowires comprise at least one n-type inorganic semiconductor material, so that multiple p-n-junctions can be formed at the inorganic semiconductor-organic semiconductor interface.

The organic semiconducting material employed for electronic device embodiments in accordance with the present invention can be any conjugated molecule, for example an aromatic molecule containing preferably two or more, very preferably at least three aromatic rings. In some preferred embodiments of the present invention, the organic semiconducting compound contains aromatic rings selected from 5-, 6- or 7-membered aromatic rings, while in other preferred embodiments the organic semiconducting contains aromatic rings selected from 5- or 6-membered aromatic rings. The organic semiconducting material may be a monomer, oligomer or polymer, including mixtures, dispersions and blends of one or more of monomers, oligomers or polymers.

Each of the aromatic rings of the organic semiconducting compound optionally contains one or more heteroatoms selected from Se, Te, P, Si, B, As, N, O or S, preferably from Si, N, O or S. Further, the aromatic rings may be optionally substituted with alkyl, alkoxy, polyalkoxy, thioalkyl, acyl, aryl or substituted aryl groups, halogen, where fluorine, cyano, nitro or an optionally substituted secondary or tertiary alkylamine or arylamine represented by $N(R^{81})(R^{82})$, where R^{81} and R^{82} are each independently H, an optionally substituted alkyl or an optionally substituted aryl, alkoxy or polyalkoxy groups are typically employed. Further, where R^{81} and R^{82} is alkyl or aryl these may be optionally fluorinated.

The aforementioned aromatic rings can be fused rings or linked to each other by a conjugated linking group such as $-C(T^1)=C(T^2)-$, $-C\equiv C-$, $-N(R^{83})-$, $-N=N-$, $(R^{83})=N-$, $-N=C(R^{83})-$, where T^1 and T^2 each independently represent H, Cl, F, $-C\equiv N$ or lower alkyl groups such as alkyl groups having from 1 to 4 carbon atoms; R^{83} represents H, optionally substituted alkyl or optionally substituted aryl. Further, where R^{83} is alkyl or aryl, it may be optionally fluorinated.

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Further preferred examples of organic semiconductor materials that can be used herein include compounds, oligomers and derivatives of compounds selected from the group consisting of conjugated hydrocarbon polymers such as polyacene, polyphenylene, poly(phenylene vinylene), polyfluorene including oligomers of those conjugated hydrocarbon polymers; condensed aromatic hydrocarbons, such as, tetracene, chrysene, pentacene, pyrene, perylene, coronene, or soluble, substituted derivatives of these; oligomeric para substituted phenylenes such as p-quaterphenyl (p-4P), p-quinquephenyl (p-5P), p-sexiphenyl (p-6P), or soluble substituted derivatives of these; conjugated heterocyclic polymers such as poly(3-substituted thiophene), poly(3,4-bisubstituted thiophene), optionally substituted polythieno[2,3-b]thiophene, optionally substituted polythieno[3,2-b]thiophene, poly(3-substituted selenophene), polybenzothiophene, polyisothianaphthene, poly(N-substituted pyrrole), poly(3-substituted pyrrole), poly(3,4-bisubstituted pyrrole), polyfuran, polypyridine, poly-1,3,4-oxadiazoles, polyisothianaphthene, poly(N-substituted aniline), poly(2-substituted aniline), poly(3-substituted aniline), poly(2,3-bisubstituted aniline), polyazulene, polypyrene; pyrazoline compounds; polyselenophene; polybenzofuran; polyindole; polypyridazine; benzidine compounds; stilbene compounds; triazines; substituted metallo- or metal-free porphines, phthalocyanines, fluorophthalocyanines, naphthalocyanines or fluoronaphthalocyanines; C₆₀ and C₇₀ fullerenes; N,N'-dialkyl, substituted dialkyl, diaryl or substituted diaryl-1,4,5,8-naphthalenetetracarboxylic diimide and fluoro derivatives; N,N'-dialkyl, substituted dialkyl, diaryl or substituted diaryl 3,4,9,10-perylenetetracarboxylicdiimide; bathophenanthroline; diphenoquinones; 1,3,4-oxadiazoles; 11,11,12,12-tetracyanonaphtho-2,6-quinodimethane; 2,2'-bis(dithieno[3,2-b:2',3'-d]thiophene); 2,8-dialkyl, substituted dialkyl, diaryl or substituted diaryl anthradithiophene; 2,2'-bisbenzo[1,2-b:4,5-b']dithiophene.

Where a liquid deposition technique of the organic semiconducting material is desired, compounds from the above list and derivatives thereof are limited to those that are soluble in an appropriate solvent or mixture of appropriate solvents.

Further, in some preferred embodiments in accordance with the present invention, the organic semiconducting materials are polymers or copolymers that encompass one or more repeating units selected from thiophene-2,5-diyl, 3-substituted thiophene-2,5-diyl, optionally substituted thieno[2,3-b]thiophene-2,5-diyl,

optionally substituted thieno[3,2-b]thiophene-2,5-diyl, selenophene-2,5-diyl, or 3-substituted selenophene-2,5-diyl.

Further preferred p-type organic semiconducting materials are copolymers comprising electron acceptor and electron donor units. Preferred copolymers of this preferred embodiment are for example copolymers comprising one or more benzo[1,2-b:4,5-b']dithiophene-2,5-diyl units that are preferably 4,8-disubstituted by one or more groups R as defined above, and further comprising one or more aryl or heteroaryl units selected from Group A and Group B, preferably comprising at least one unit of Group A and at least one unit of Group B, wherein Group A consists of aryl or heteroaryl groups having electron donor properties and Group B consists of aryl or heteroaryl groups having electron acceptor properties, and preferably

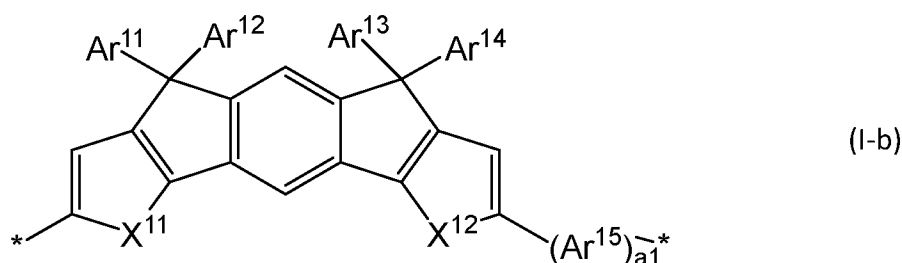
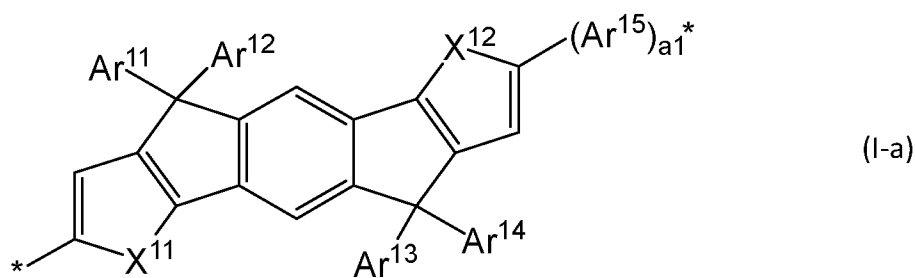
Group A consists of selenophene-2,5-diyl, thiophene-2,5-diyl, thieno[3,2-b]thiophene-2,5-diyl, thieno[2,3-b]thiophene-2,5-diyl, selenopheno[3,2-b]selenophene-2,5-diyl, selenopheno[2,3-b]selenophene-2,5-diyl, selenopheno[3,2-b]thiophene-2,5-diyl, selenopheno[2,3-b]thiophene-2,5-diyl, benzo[1,2-b:4,5-b']dithiophene-2,6-diyl, 2,2-dithiophene, 2,2-diselenophene, dithieno[3,2-b:2',3'-d]silole-5,5-diyl, 4H-cyclopenta[2,1-b:3,4-b']dithiophene-2,6-diyl, 2,7-dithien-2-yl-carbazole, 2,7-di-thien-2-yl-fluorene, indaceno[1,2-b:5,6-b']dithiophene-2,7-diyl, benzo[1'',2'':4,5;4'',5'':4',5']bis(silolo[3,2-b:3',2'-b']thiophene)-2,7-diyl, 2,7-di-thien-2-yl-indaceno[1,2-b:5,6-b']dithiophene, 2,7-di-thien-2-yl-benzo[1'',2'':4,5;4'',5'':4',5']bis(silolo[3,2-b:3',2'-b']thiophene)-2,7-diyl, and 2,7-di-thien-2-yl-phenanthro[1,10,9,8-c,d,e,f,g]carbazole, all of which are optionally substituted by one or more, preferably one or two groups R⁷⁸ as defined above, and

Group B consists of benzo[2,1,3]thiadiazole-4,7-diyl, 5,6-dialkylbenzo[2,1,3]thiadiazole-4,7-diyl, 5,6-dialkoxybenzo[2,1,3]thiadiazole-4,7-diyl, benzo[2,1,3]selenadiazole-4,7-diyl, 5,6-dialkoxy-benzo[2,1,3]selenadiazole-4,7-diyl, benzo[1,2,5]thiadiazole-4,7-diyl, benzo[1,2,5]selenadiazole-4,7-diyl, benzo[2,1,3]oxadiazole-4,7-diyl, 5,6-dialkoxybenzo[2,1,3]oxadiazole-4,7-diyl, 2H-benzotriazole-4,7-diyl, 2,3-dicyano-1,4-phenylene, 2,5-dicyano-1,4-phenylene, 2,3-difluoro-1,4-phenylene, 2,5-difluoro-1,4-phenylene, 2,3,5,6-tetrafluoro-1,4-phenylene, 3,4-difluorothiophene-2,5-diyl, thieno[3,4-b]pyrazine-2,5-diyl, quinoxaline-5,8-diyl, thieno[3,4-b]thiophene-4,6-diyl, thieno[3,4-b]thiophene-6,4-

diyl, and 3,6-pyrrolo[3,4-c]pyrrole-1,4-dione, all of which are optionally substituted by one or more, preferably one or two groups R^{78} as defined above.

In other preferred embodiments of the present invention, the organic semiconducting materials are substituted oligoacenes such as pentacene, tetracene or anthracene, or heterocyclic derivatives thereof. Bis(trialkylsilylethynyl) oligoacenes or bis(trialkylsilylethynyl) heteroacenes, as disclosed for example in US 6,690,029 or WO 2005/055248 A1 or US 7,385,221, are also useful.

Preferred organic semiconducting compounds may, for example, be selected from oligomers, polymers or copolymers comprising two or more repeating constitutional (structural) units M selected from the following formulae (I-a) and (I-b)



wherein

X^{11} and X^{12} are independently of each other S or Se;

Ar^{11} , Ar^{12} , Ar^{13} , and Ar^{14} are independently of each other selected from the group consisting of

- (i) aryl comprising at least one substituent R^{11} , and
- (ii) heteroaryl having from 5 to 30 aromatic ring atoms, said heteroaryl being unsubstituted or substituted with one or more groups R^{12} ;

a_1 is 0 or an integer of from 1 to 10; and

- Ar¹⁵ is at each occurrence independently selected from the group consisting of aryl, substituted aryl, heteroaryl and substituted heteroaryl having up to 30 aromatic ring atoms;
- R¹¹ is an organyl or organoheteryl comprising from 13 to 19 carbon atoms; and
- 5 R¹² is at each occurrence independently selected from the group consisting of any group R^T as defined herein, hydrocarbyl having from 1 to 40 carbon atoms wherein the hydrocarbyl may be further substituted with one or more groups R^T and hydrocarbyl having from 1 to 40 carbon atoms comprising one or more heteroatoms selected from the group consisting of N, O, S, P, Si, Se, As, Te or Ge, with N, O and S being preferred heteroatoms, wherein the hydrocarbyl may be further substituted with one or more groups R^T;
- 10 R^T is at each occurrence independently selected from the group consisting of F, Br, Cl, -CN, -NC, -NCO, -NCS, -OCN, -SCN, -C(O)NR⁰R⁰⁰, -C(O)X⁰, -C(O)R⁰, -NH₂, -NR⁰R⁰⁰, -SH, -SR⁰, -SO₃H, -SO₂R⁰, -OH, -OR⁰, -NO₂, -SF₅ and -SiR⁰R⁰⁰R⁰⁰⁰;
- 15 R⁰, R⁰⁰ and R⁰⁰⁰ are at each occurrence independently of each other selected from the group consisting of H, F, hydrocarbyl having from 1 to 40 carbon atoms; and
- X⁰ is halogen.
- 20 Such compounds are, for example, disclosed in WO 2015/180822 A1 or in WO 2016/015804 A1, incorporated herein by reference.

The small molecule or monomeric compound comprising the tetra-heteroaryl indacenodithiophene-based structural unit may for example be represented by

25 formula (II-a)

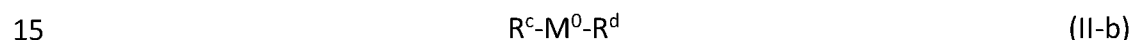


wherein M⁰ comprises a structural unit M of formula (I-a) or (I-b) as defined above

30 and R^a and R^b are inert chemical groups. Such inert chemical groups R^a and R^b may independently of each other for example be chosen from the group consisting of hydrogen, fluorine, alkyl having from 1 to 10 carbon atoms, fluoroalkyl having from 1 to 10 carbon atoms, aromatic ring systems of from 5 to 30 carbon atoms and aromatic ring systems of from 5 to 30 carbon atoms wherein one or more hydrogen

35 atom may independently of each other be replaced by fluorine or alkyl having from 1 to 10 carbon atoms.

Alternatively, the small molecule or monomeric compound comprising the tetra-heteroaryl indacenodithiophene-based structural unit may be a compound comprising a structural unit of formula (I-a) or (I-b), as defined above, and at least one reactive chemical group R^c which may be selected from the group consisting of Cl, Br, I, O-tosylate, O-triflate, O-mesylate, O-nonaflate, $-\text{SiMe}_2\text{F}$, $-\text{SiMeF}_2$, $-\text{O-SO}_2\text{Z}^1$, $-\text{B}(\text{OZ}^2)_2$, $-\text{CZ}^3=\text{C}(\text{Z}^3)_2$, $-\text{C}\equiv\text{CH}$, $-\text{C}\equiv\text{CSi}(\text{Z}^1)_3$, $-\text{ZnX}^0$ and $-\text{Sn}(\text{Z}^4)_3$, preferably $-\text{B}(\text{OZ}^2)_2$ or $-\text{Sn}(\text{Z}^4)_3$, wherein X^0 is as defined above, and Z^1 , Z^2 , Z^3 and Z^4 are selected from the group consisting of alkyl and aryl, preferably alkyl having from 1 to 10 carbon atoms, each being optionally substituted with R^0 as defined above, and two groups Z^2 may also together form a cyclic group. According to a further alternative, such a monomeric compound or small molecule may comprise two reactive chemical groups and is represented by formula (II-b)

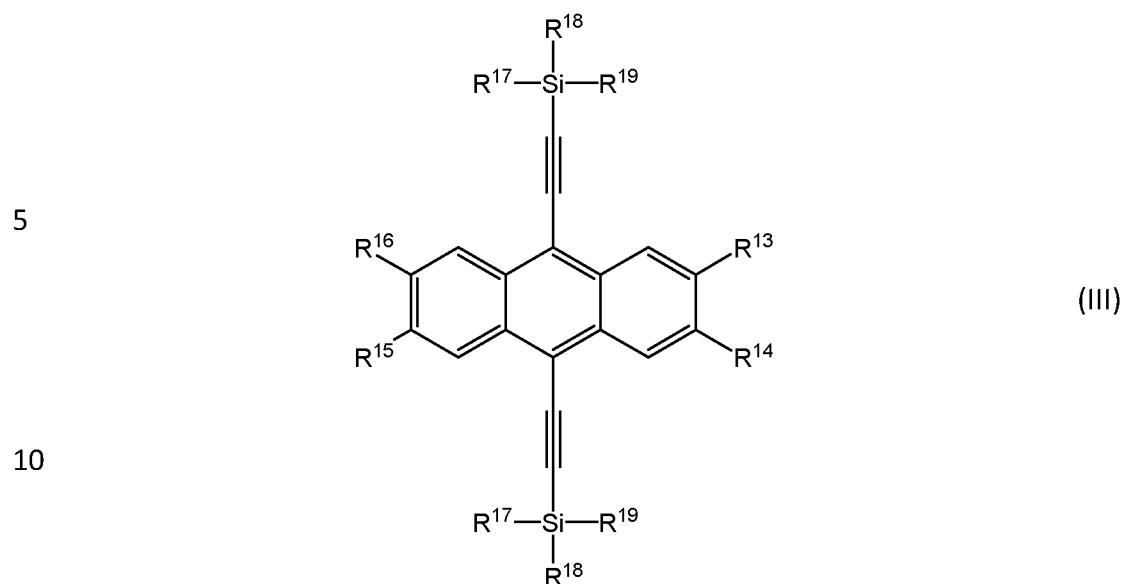


wherein M^0 comprises a structural M unit of formula (I-a) or (I-b) as defined above, and R^c and R^d are reactive chemical groups as defined above for R^c .

Specific examples of such monomeric compounds or small molecules comprising the tetra-heteroaryl indacenodithiophene-based structural unit are disclosed, for example, in already-mentioned WO 2015/180822 A1 and WO 2016/015804 A1, incorporated herein by reference.

The small molecule or monomeric compound may also be selected from bis(trialkylsilylethynyl) oligoacenes or bis(trialkylsilylethynyl) heteroacenes, as disclosed for example in WO 2005/055248 A1, such as, for example, monomeric compounds represented by the following formula (III)

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15 wherein R^{13} , R^{14} , R^{15} , R^{16} , R^{17} , R^{18} , and R^{19} are each independently the same or different and each independently comprise H, or optionally substituted C_1 - C_{40} -carbonyl or hydrocarbonyl groups. R^{13} and R^{14} may be the same or different; preferably they are the same. R^{15} and R^{16} may be the same or different; preferably they are the same. Equally, all of R^{13} , R^{14} , R^{15} and R^{16} may be the same or different; preferably they are the same.

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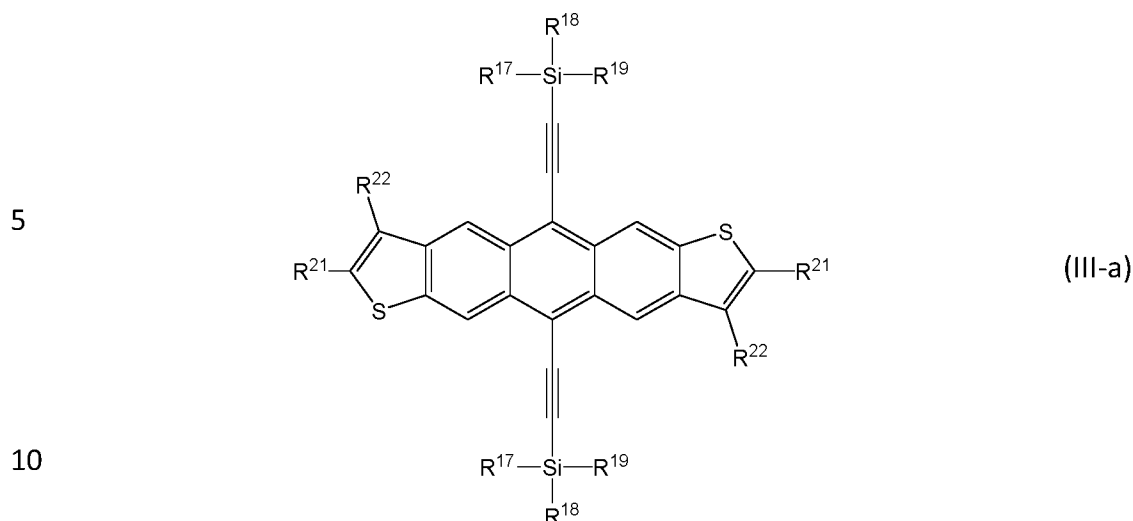
Most preferably, R^{13} and R^{14} together, and R^{15} and R^{16} together, in combination with the aromatic carbon atom to which they are attached form a C_4 - C_{40} saturated or unsaturated ring, more preferably a C_4 - C_{10} saturated or unsaturated ring wherein one or more carbon atom may optionally be replaced by one or more oxygen atom or sulphur atom or a group represented by the formula $-N(R^{20})$, wherein R^{20} is a hydrogen atom or a hydrocarbon group, thereby forming a pseudo-pentacene compound.

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30 More preferably, the small molecule or monomeric compound is selected from compounds of the following formula (III-a) and isomers thereof, wherein one or more of the carbon atoms of the polyacene skeleton may be substituted by a heteroatom selected from N, P, As, O, S, Se and Te, preferably S.

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wherein R^{17} , R^{18} and R^{19} are as defined herein, and R^{21} and R^{22} are as defined in the following.

15 In the compound of formula (III-a), R^{21} and R^{22} may be the same or different, but preferably are the same. R^{21} and R^{22} may at each occurrence independently be selected from the group consisting of H, optionally substituted C_1 - C_{40} -carbonyl or hydrocarbonyl groups, or halogen. More preferably R^{21} and R^{22} may at each occurrence independently be selected from the group consisting of H, F, or optionally substituted, optionally unsaturated C_1 - C_{40} -carbonyl or hydrocarbonyl groups, for example, optionally substituted alkyl, alkenyl, alkynyl, aryl or aralkyl groups, or R^{21} and R^{22} may in combination with the aromatic carbon atom to which they are attached or in combination with a further substituent optionally form a C_4 - C_{40} saturated or unsaturated ring, more preferably a C_4 - C_{10} saturated or unsaturated ring wherein one or more carbon atom may optionally be replaced by one or more oxygen atom or sulphur atom or a group represented by the formula $-N(R^{20})$, wherein R^{20} is a hydrogen atom or a hydrocarbon group.

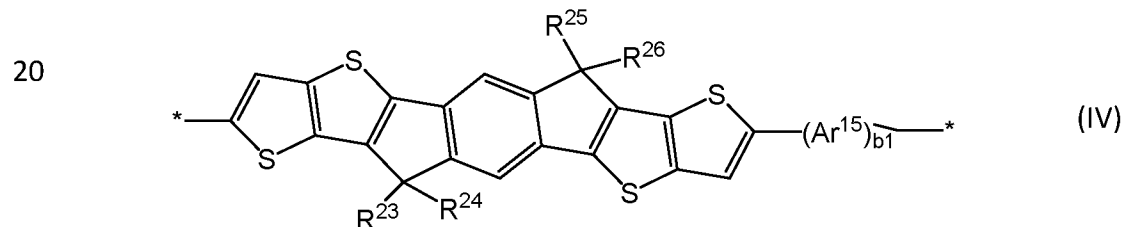
30 In the compound of formula (III-a) R^{17} , R^{18} and R^{19} may be the same or different, most preferably R^{17} , R^{18} and R^{19} are the same and comprise an optionally substituted C_1 - C_{40} carbonyl or hydrocarbonyl group, for example a C_1 - C_{40} alkyl group (preferably C_1 - C_4 alkyl and most preferably methyl, ethyl, n-propyl or isopropyl) which may optionally be substituted for example with a halogen atom; a C_6 - C_{40} aryl group (preferably phenyl) which may optionally be substituted for example with a halogen atom; a C_6 - C_{40} arylalkyl group which may optionally be substituted for

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example with a halogen atom; a C₁-C₄₀ alkoxy group which may optionally be substituted for example with a halogen atom; or a C₆-C₄₀ arylalkyloxy group which may optionally be substituted for example with a halogen atom or any two of R¹⁷, R¹⁸ and R¹⁹ together with, for example, the atom to which they are attached form a C₄-C₄₀ saturated or unsaturated ring, more preferably an optionally substituted C₄-C₁₀ saturated or unsaturated ring, intervened by one or more oxygen or sulphur atoms or a group represented by formula -N(R²⁰), with R²⁰ as defined herein. Preferably, R¹⁷, R¹⁸ and R¹⁹ are each independently selected from optionally substituted C₁-C₁₀ alkyl (more preferably C₁-C₄ and even more preferably C₁-C₃ alkyl, for example isopropyl) and optionally substituted C₆-C₁₀ aryl (preferably phenyl).

The p-type organic semiconductor material according to the present invention may further be selected from oligomers, polymers or copolymers comprising two or more dithieno[2,3-d:2',3'-d']-s-indaceno[1,2-b:5,6-b']dithiophene repeating units, such as, for example, described in WO 2013/010614 A1. Exemplary oligomers, polymers or copolymers preferably comprise two or more repeating constitutional (structural) units M of the following formulae (IV)



25 wherein

b1 is an integer from 1 to 10, preferably 1 to 3;
 R²³, R²⁴, R²⁵, and R²⁶ are on each occurrence identically or differently selected from the group consisting of H, F, Cl, Br, CN, straight-chain, branched or cyclic alkyl, with 1 to 30 C atoms, in which one or more non-adjacent C atoms are optionally replaced by -O-, -S-, -C(O)-, -C(O)-O-, -O-C(O)-, -O-C(O)-O-, -C(S)-, -C(S)-O-, -O-C(S)-, -O-C(S)-O-, -C(O)-S-, -S-C(O)-, -O-C(O)-S-, -S-C(O)-O-, -S-C(O)-S-, -S-C(S)-S-, -O-C(S)-S-, -S-C(S)-O-, -C(S)-S-, -S-C(S)-, -NR⁰-, -SiR⁰R⁰⁰-, -CY¹=CY²- or -C≡C- in such a manner that O and/or S atoms are not linked directly to one another, and in which one or more H atoms are optionally replaced by F, Cl, Br, I or CN, or R on each occurrence identically or differently denotes aryl, heteroaryl, aryloxy or heteroaryloxy with 4 to 20 ring atoms

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which is optionally substituted, or any two of R^{23} , R^{24} , R^{25} , and R^{26} bonded to the same C atom together form an alicyclic group with 1 to 20 C atoms, which is optionally fluorinated or alkylated;

- 5 Y^1 and Y^1 are at each occurrence independently of each other selected from the group consisting of H, F, Cl or CN;
- R^0 and R^{00} are at each occurrence independently of each other selected from the group consisting of H or optionally substituted C_1 - C_{40} -carbonyl or hydrocarbonyl;
- Ar^{15} is at each occurrence independently as defined previously;
- R^5 is at each occurrence independently as defined previously;
- 10 R^5 is on each occurrence independently selected from the group consisting of F, X^0 is halogen, preferably F, Cl or Br.

- 15 Preferably, with regards to formula (IV), R^{23} , R^{24} , R^{25} , and R^{26} are at each occurrence independently selected from the groups consisting of straight-chain, branched or cyclic alkyl with 1 to 20 C atoms which is unsubstituted or substituted by one or more F atoms, or R^{23} , R^{24} , R^{25} , and R^{26} may on each occurrence independently be selected from the group consisting of aryl and heteroaryl, each of which is optionally fluorinated, alkylated or alkoxyated and has 4 to 30 ring atoms, or one of two of R^{23} , R^{24} , R^{25} , and R^{26} bonded to the same C atom denotes H and the other
- 20 is selected from the group consisting of the aforementioned alkyl, aryl or heteroaryl groups, or two of R^{23} , R^{24} , R^{25} , and R^{26} bonded to the same C atom together form a cyclic alkyl group with 1 to 20 C atoms, which is unsubstituted or substituted by one or more F atoms or by one or more C_1 - C_{10} alkyl groups.

- 25 The p-type organic semiconductor material according to the present invention may further be selected from oligomers, polymers or copolymers comprising two or more repeating units selected from benzo[1,2-b:4,5-b']dithiophene-2,5-diyl, 4,8-disubstituted benzo[1,2-b:4,5-b']dithiophene-2,5-diyl, thiophene-2,5-diyl, 3-substituted thiophene-2,5-diyl, optionally substituted thieno[2,3-b]thiophene-2,5-diyl, optionally substituted thieno[3,2-b]thiophene-2,5-diyl, selenophene-2,5-diyl, or 3-substituted selenophene-2,5-diyl.
- 30

- 35 Preferably, the 4,8-disubstituted benzo[1,2-b:4,5-b']dithiophene-2,5-diyl, the 3-substituted thiophene-2,5-diyl, the optionally substituted thieno[2,3-b]thiophene-2,5-diyl, the optionally substituted thieno[3,2-b]thiophene-2,5-diyl and the 3-substituted selenophene-2,5-diyl repeating units are substituted by one or more

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group at each occurrence independently selected from the group consisting of halogen, preferably fluorine; alkyl having from 1 to 10, preferably from 1 to 5 carbon atoms, more preferably methyl; partially or fully halogenated, preferably fluorinated, alkyl having from 1 to 10, preferably from 1 to 5 carbon atoms, more preferably methyl; alkoxy having from 1 to 10, preferably from 1 to 5 carbon atoms, more preferably methoxy; and partially or fully halogenated, preferably fluorinated, alkoxy having from 1 to 10, preferably from 1 to 5 carbon atoms, more preferably methoxy.

Further preferably, the position of the HOMO of the p-type organic semiconductor material is chosen to be lower than the conducting band edge of the n-type semiconducting material of the piezoelectric nanowires. This allows effective band bending in the semiconductor piezoelectric material, which in turn allows effective depletion of excess negative charges present on the surface of the inorganic semiconducting piezoelectric nanowires. In another example the position of the HOMO of the p-type semiconducting material of the piezoelectric nanowires is chosen to be lower than the conducting band edge of the n-type organic semiconductor material.

20

DIELECTRIC TOP LAYER

Preferably, the dielectric top layer comprises, e.g. in at least 50 wt% or 60 wt% or 70 wt% or 80 wt% or 90 wt% or 95 wt% or 97 wt% or 99.0 wt% or 99.5 wt% or 99.7 wt% or 99.9 wt%, with wt% relative to the total weight of the dielectric top layer, and preferably consists of, one or more dielectric material as defined herein.

25

Additionally, the dielectric top layer may also comprise other materials that may, for example, add additional functionalities or help in the deposition of the dielectric top layer.

30

For the purposes of the present application the type of dielectric material is not particularly limited.

35

It is also noted that the dielectric materials comprised in – if present – the buffer layer and the dielectric top layer may be selected independently of each other, i.e. they may be the same or different from each other.

5 Suitable dielectric materials may, for example, be selected from so-called high-k or low-k dielectric materials, distinguished by their permittivity or dielectric constant ϵ , though for the present application low-k dielectric materials may be preferred.

10 Preferred low-k dielectric materials are characterized by a permittivity ϵ of at most 4.0, preferably of at most 3.5, more preferably of at most 3.0, even more preferably of at most 2.8, still even more preferably of at most 2.6. Preferred low-k dielectric materials have a permittivity of at least 0.1, more preferably of at least 0.5, even more preferably of at least 1.0, for example, of at least 1.1 or 1.2 or 1.3 or 1.4 or 1.5 or 1.6 or 1.7 or 1.8 or 1.9 or 2.0.

15 High-k dielectric materials may consequently be characterized by a permittivity ϵ of more than 4.0, for example, of at least 5.0 or 10.0.

20 Throughout this application, the values for the permittivity or dielectric constant ϵ refer to values taken at 20°C and 1,000 Hz.

25 An overview of low dielectric constant materials is, for example, given by W. Volksen et al. in Chemical Reviews 2010, 110, 56-110, or by A. Facchetti et al. in Advanced Materials 2005, 17, 1705-1725.

30 A list of suitable organic dielectric materials having low permittivity is given without limiting to these in the following Table 1.

Table 1

30	Dielectric material	Typical low frequency permittivity (ϵ)
	Fluorinated para-xylene	2.4
	Fluoropolyarylether	2.6
35	Fluorinated polyimide	2.7
	Polystyrene	2.5

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Table 1

	Dielectric material	Typical low frequency permittivity (ϵ)
5	Poly(α -methyl styrene)	2.6
	Poly(α -vinyl naphthalene)	2.6
	Poly(vinyltoluene)	2.6
	Polyethylene	2.2-2.3
	cis-polybutadiene	2.0
10	Polypropylene	2.2
	Polyisoprene	2.3
	Poly(4-methyl-1-pentene)	2.1
	Poly(tetrafluoroethylene)	2.1
15	Poly(chlorotrifluoroethylene)	2.3-2.8
	Poly(2-methyl-1, 3-butadiene)	2.4
	Poly(p-xylylene)	2.6
	Poly(α - α' - α' - α' tetrafluoro-p-xylylene)	2.4
20	Poly[1,1-(2-methyl propane) bis (4-phenyl) carbonate]	2.3
	Poly(cyclohexyl methacrylate)	2.5
	Poly(chlorostyrene)	2.6
	Poly(2,6-dimethyl-1, 4-phenylene ether)	2.6
25	Polyisobutylene	2.2
	Poly(vinyl cyclohexane)	2.2
	Poly(arylene ether)	2.6-2.8
	Polyphenylene	2.6-2.7
30		

A list of suitable fluorinated dielectric materials having low permittivity is given without limitation to these in the following Table 2:

35

Table 2

Dielectric Material	Typical low frequency permittivity (ϵ)
Poly(ethylene/tetrafluoroethylene)	2.6
Poly(ethylene/chlorotrifluoroethylene)	2.3
Fluorinated ethylene/propylene copolymer	2-2.5
Cytop	2.0
Teflon AF	1.9-2.1

Suitable dielectric materials may, for example, be selected from organic dielectric materials and inorganic dielectric materials. For reasons of processability, in organic electronic devices organic dielectric materials are generally preferred over inorganic dielectric materials, but inorganic dielectric materials may also be used.

Exemplary inorganic dielectric materials may, for example, be selected from oxides or nitrides, such as silicon oxide (SiO_x , e.g. silicon dioxide (SiO_2), for example as grown in situ on doped Si gates), silicon nitride (SiN_x , e.g. Si_3N_4), metal oxides, and any blend of any of these. Examples of suitable metal oxides may be selected from the group consisting of tantalum oxide (e.g. Ta_2O_5), aluminum oxides (e.g. Al_2O_3 or $\text{Al}_2\text{O}_{3+x}$), titanium dioxide (TiO_2), barium zirconium titanate, barium strontium titanate, zirconium dioxide (ZrO_2) and any mixture of any of these.

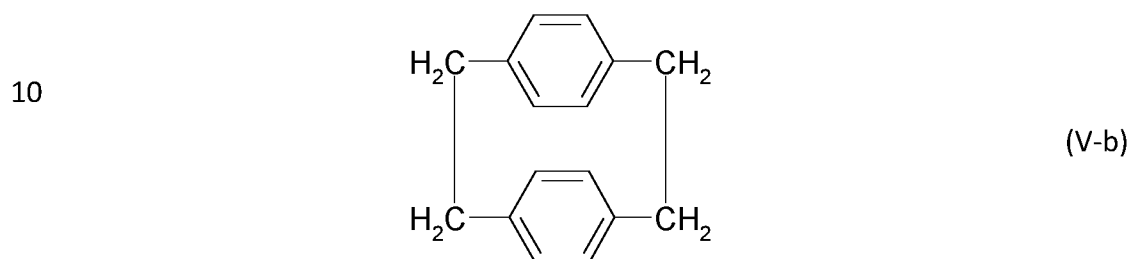
Exemplary organic dielectric materials may, for example, be selected from the group consisting of polystyrene (PS), including at least partially crosslinked polystyrene, polyvinyl alcohol (PVA), poly(p-xylylenes), polyvinylphenol (PVP), polyacrylate (PA), polymethylmethacrylate (PMMA), polyvinylchloride (PVC), polyvinylidene fluoride (PVDF), poly(α -methylstyrene) (P α MS), cyanoethylpullalan (CYPEL), polyimide, polycycloolefinic polymers, fully or partially fluorinated polymers as described herein, and any blend of any of these.

Poly(p-xylylene), which herein may also be referred to as ParyleneTM, is generally prepared by chemical vapor deposition of a p-xylylene intermediate of the following formula (V-a)

- 31 -

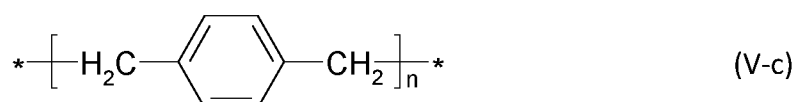


5 which may optionally be substituted as described below. Such p-xylylene intermediate can be derived from [2.2]paracyclophane of the following formula (V-b)

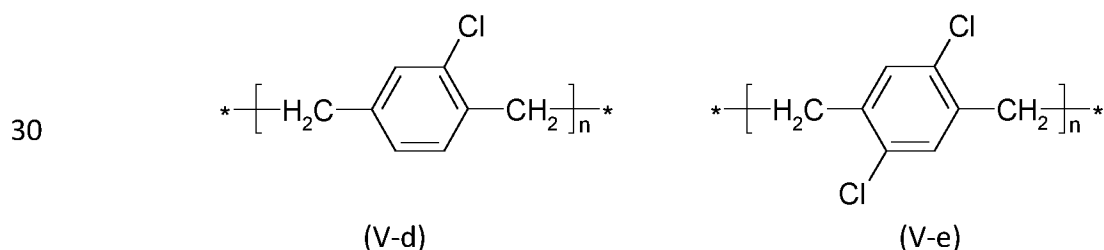


15 The phenylene rings and/or the methylene groups of the p-xylylene repeating units in the polymers may also be substituted. The polymers may also comprise two or more distinct types of unsubstituted and/or substituted p-xylylene repeating units. For example, Parylene N denotes unsubstituted poly(p-xylylene) of the following

20 formula (V-c)



25 while Parylene C and Parylene D denote mono- or dichlorinated poly(p-xylylene) of the following formulae (V-d) and (V-e), respectively



35 Further examples of suitable poly(p-xylylenes) include for example those wherein the phenylene or methylene groups are fluorinated, like Parylene AF-4, Parylene SF, or Parylene HT all comprising difluorinated methylene groups, or Parylene VT

comprising a fluorinated phenylene. Further Parylene types include polymers wherein the phenylene ring is substituted by a reactive or crosslinkable group, for example by an amine (Parylene A), a methylamine (Parylene AM), or an ethynyl group (Parylene X).

5

Unless stated otherwise, the terms "Parylene" and "poly(p-xylylene)" as used herein, are understood to include both unsubstituted and substituted Parylene types, including but not limited to Parylene N, C, D, AF-4, SF, HT, VT, A, AM, X etc..

10

The fluorinated dielectric material is not particularly limited. In general terms, the fluorinated dielectric material is a fluoropolymer comprising or, preferably, consisting of monomeric units derived from partially or fully fluorinated monomers, or both, partially and fully fluorinated monomers. Suitable fluoropolymers may, for example, be homopolymers, random copolymers or block copolymers. Such materials are generally known to the skilled person and can be obtained from various commercial sources.

15

Suitable examples of partially or fully fluorinated monomers may at each occurrence independently be selected from the group consisting of the following:

20

Group A perfluorinated olefins having from two to eight carbon atoms, such as for example tetrafluoroethylene or hexafluoropropylene;

Group B partially fluorinated olefins having from two to eight carbon atoms, such as for example vinylidene fluoride ($F_2C=CH_2$) vinyl fluoride ($HFC=CH_2$), 1,2-difluoroethylene ($HFC=CHF$) or trifluoroethylene ($F_2C=CFH$);

25

Group C (perfluoroalkyl)ethylenes of formula $H_2C=CH-C_aF_{2a+1}$ with a being an integer from 1 to 10, preferably a being 1, 2, 3, 4, 5 or 6, most preferably a being 1, 2, 3 or 4;

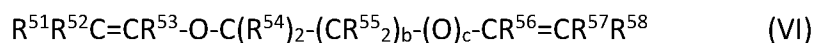
30

Group D partially fluorinated olefins, wherein one or more of the hydrogen atoms is independently of each other replaced with one selected from the group consisting of chlorine, bromine or iodine, such as for example chlorotrifluoroethylene ($ClFC=CF_2$);

Group E perfluorovinylalkylethers of formula $F_2C=C-O-C_aF_{2a+1}$ with a being an integer from 1 to 10, preferably a being 1, 2, 3, 4, 5 or 6, most preferably a being 1, 2, 3 or 4;

35

Group F partially or fully fluorinated diolefins of the following formula (VI)

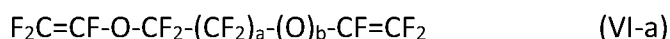


5 with b being 0, 1 or 2; c being 0 or 1; R^{51} , R^{52} , R^{53} , R^{54} , R^{55} , R^{56} , R^{57} and R^{58} being at each occurrence independently selected from the group consisting of H, F, Cl, alkyl having from 1 to 5 carbon atoms, and alkyl having from 1 to 5 carbon atoms with one or more, preferably all, hydrogen atoms substituted by F; and

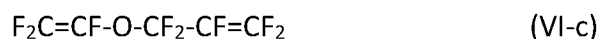
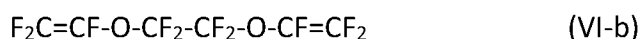
10 Group G partially or fully fluorinated 5-membered rings comprising at least one double bond, preferably partially or fully fluorinated dihydrofurans or dioxoles, preferably perfluorinated dihydrofurans or dioxoles.

15 With regards to formula (VI) of Group F, it is preferred that R^{51} , R^{52} , R^{53} , R^{54} , R^{55} , R^{56} , R^{57} and R^{58} are at each occurrence independently selected from the group consisting of H, F, CH_3 , CH_2F , CHF_2 , and CF_3 ; and more preferred that R^{51} , R^{52} , R^{57} and R^{58} are F, and R^{53} , R^{54} , R^{55} , and R^{56} are at each occurrence independently F or CF_3 ; and most preferred that R^{51} , R^{52} , R^{53} , R^{54} , R^{55} , R^{56} , R^{57} and R^{58} are all F.

20 Preferred examples of the monomers of Group F are represented by the following formula (VI-a)



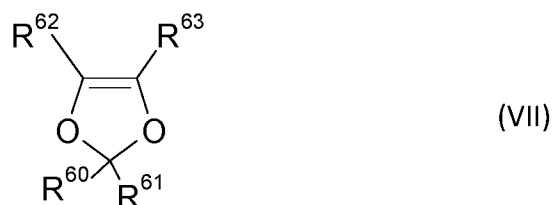
25 with a being 0, 1 or 2; b being 0 or 1. Specific examples of the monomers of Group F may be selected from the following formulae (VI-b) and (VI-c)



30 With regards to Group G, preferred examples are represented by the following formula (VII)

35

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5

wherein R^{60} , R^{61} , R^{62} and R^{63} are at each occurrence independently selected from the group consisting of F, partially or fully fluorinated alkyl comprising from 1 to 5 carbon atoms and partially or fully fluorinated alkoxy comprising from 1 to 5 carbon atoms. Preferably R^{60} , R^{61} , R^{62} and R^{63} are at each occurrence independently F or fully fluorinated alkyl comprising from 1 to 5 carbon atoms. More preferably, R^{60} , R^{61} , R^{62} and R^{63} are at each occurrence independently F or $-CF_3$. Most preferably, R^{60} and R^{61} are $-CF_3$, and R^{62} and R^{63} are F.

10

The fluoropolymer may comprise the monomeric units derived from partially and/or fully fluorinated monomers preferably in at least 50 mol%, more preferably in at least 60 mol% or 70 mol% or 80 mol% or 90 mol%, even more preferably in at least 95 mol% or 97 mol% or 99 mol%, still even more preferably in at least 99.5 mol% and most preferably consists of such at least one partially or fully fluorinated monomer, with mol% relative to the total number of repeat units comprised in the fluoropolymer. The remainder of repeat units comprised in such fluoropolymer may be derived from any suitable monomer, such as, for example, olefins having one or two double bonds. Exemplary olefins may be selected from the group consisting of ethylene, propylene, butene-1, butene-2, iso-butylene, butadiene, and any mixture of any of these.

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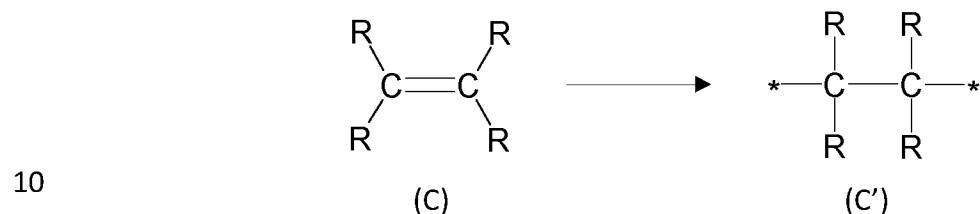
The fluoropolymer may optionally comprise repeat units derived from at least one fluorine-free monomer, i.e. from a monomer that does not comprise any fluorine atom. Suitable examples of the at least one fluorine-free monomer may at each occurrence independently be selected from the group consisting of

30

- (i) olefins having from 2 to 8 carbon atoms, suitable examples of which may be selected from the group consisting of ethylene, propylene, butene-1, butene-2, buta-1,3-diene, pentene-1, pentene-2, hexene-1, hexene-2 and octene-1, with ethylene, propylene, butene-1 and hexene-1 being preferred;
- (ii) vinyl monomers, such as vinyl chloride;
- (iii) acrylate monomers, such as methyl methacrylate; and
- (iv) styrene monomers, such as styrene or methylstyrene.

35

The respective polymers are obtained by polymerization of one or more of the above monomers by methods well known to the skilled person, for example, by radical polymerization. In general terms, such polymerization is based on rendering an olefin monomer (C) into an alkanediyl constitutional unit (C') as schematically shown below.



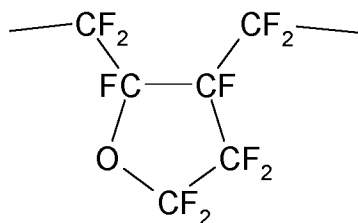
with R denoting any carbyl group.

Preferably the fluoropolymer is an amorphous fluoropolymer. Preferred examples of amorphous fluoropolymers may be selected from the group of polymers comprising, preferably consisting of, a first monomeric unit, at each occurrence independently derived from a monomer of Group E as defined above or a monomer of Group G as defined above, and an optional second monomeric unit, at each occurrence independently derived from a monomer selected from the group consisting of any of Groups A, B, C and D as defined above, and olefins as defined above.

An example of a suitable amorphous fluoropolymer comprises, preferably consists of, monomeric units derived from perfluorinated dioxole and at least one olefin. Said at least one olefin may be selected from the group consisting of monomers of any of Groups A, B, C and D as defined above as well as fluorine-free olefins having from 2 to 8 carbon atoms as defined above.

Another example of a suitable amorphous fluoropolymer comprises, preferably consists of, constitutional units derived from the monomers of Group G, for example of the following formula (VII'), and optionally further constitutional units of formula $^*-\text{CF}_2-\text{CF}_2-^*$.

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(VII')

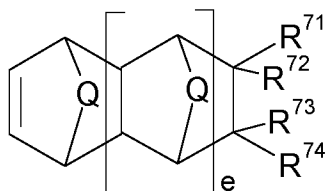
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Such fluoropolymers are, for example, commercially available from AGC Chemicals Europe as Cytop™, or from Du Pont de Nemours as Teflon™ AF. Specific examples of suitable and commercially available amorphous fluoropolymers are Cytop 809M® or Cytop 107M® from AGC Chemicals. Other examples of suitable fluoropolymers are, for example, Teflon AF® 1600 or 2400 from DuPont de Nemours.

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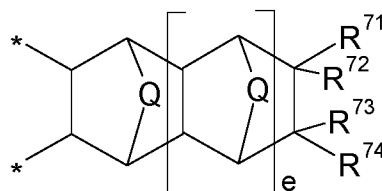
Suitable examples of polycycloolefinic polymers are preferably selected from polynorbornenes. Suitable polynorbornenes may be obtained by addition polymerization of monomers of the following general formula (VIII) and thus comprise constitutional units of the following formula (VIII')

15



(VIII)

20



(VIII')

25

with e, Q, R⁷¹, R⁷², R⁷³ and R⁷⁴ as defined herein.

e is at each occurrence independently an integer of from 0 to 5, e.g. 0, 1, 2, 3, 4 or 5. Preferably e is an integer from 0 to 3, e.g. 0, 1, 2 or 3. More preferably e is 0 or 1. Most preferably e is 0.

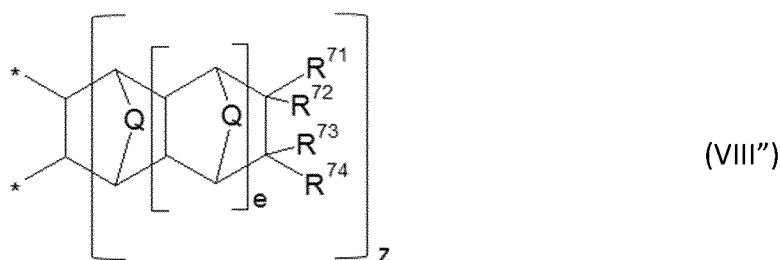
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Q is at each occurrence independently selected from the group consisting of -CH₂-, -CH₂-CH₂-, -CF₂-, -CF₂-CF₂- and O. Preferably Q is selected from the group consisting of -CH₂-, -CH₂-CH₂- and O. Most preferably Q is -CH₂-.

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Such polymers may then generally be represented by the following formula (VIII'')



10 with z being the number of constitutional units, and e , R^{71} , R^{72} , R^{73} and R^{74} as defined herein.

15 The number z of constitutional units may, for example, be at least 10 and at most 1000. Alternatively, the number z of constitutional units may be such that the weight average molecular weight M_w is appropriate for the intended use.

20 For example, the weight average molecular weight M_w may be at least 5,000 g/mol, preferably at least 10,000 g/mol, more preferably at least 20,000 g/mol, even more preferably at least 30,000 g/mol, even more preferably at least 40,000 g/mol, still even more preferably at least 50,000 g/mol, and most preferably at least 60,000 g/mol, as determined by gel permeation chromatography (GPC). For example, the weight average molecular weight M_w may be at most 500,000 g/mol, preferably at most 450,000 g/mol, more preferably at most 400,000 g/mol, even more preferably at most 350,000 g/mol, even more preferably at most 300,000 g/mol, and most preferably at most 250,000 g/mol, as determined by gel permeation chromatography (GPC).

30 R^{71} , R^{72} , R^{73} and R^{74} are at each occurrence independently of each other hydrogen or a carbyl group. Suitable carbyl groups may, for example, be selected from the group consisting of hydrocarbyl groups, halohydrocarbyl groups, and perhalocarbyl groups. Suitable carbyl groups also include hydrocarbyl groups, halohydrocarbyl groups and perhalocarbyl groups, wherein one or more carbon atoms each may be replaced by a group selected from aryl groups, $^*-C\equiv C^*$, $>C=C<$, and a heteroatom selected from the group consisting of O, S, N, P or Si, preferably from the group

35

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consisting of O, N, P or Si, and most preferably O or Si. Suitable carbyl groups may also include latent crosslinkable groups as described in the following.

5 As used herein, "hydrocarbyl" refers to a radical or group that contains a carbon backbone where each carbon is appropriately substituted with one or more hydrogen atoms. The term "halohydrocarbyl" refers to a hydrocarbyl group where one or more of the hydrogen atoms, but not all, have been replaced by a halogen (F, Cl, Br, or I). The term perhalocarbyl refers to a hydrocarbyl group where each hydrogen has been replaced by a halogen.

10 Non-limiting examples of hydrocarbyls, include, but are not limited to alkyl having from 1 to 25 carbon atoms, alkenyl having from 2 to 24 carbon atoms, alkynyl having from 2 to 24 carbon atoms, cycloalkyl having from 5 to 25 carbon atoms, aryl having from 6 to 24 carbon atoms, and aralkyl having from 7 to 24 carbon atoms.

15 Exemplary alkyl groups include but are not limited to methyl, ethyl, propyl, isopropyl, butyl, isobutyl, sec-butyl, tert-butyl, pentyl, neopentyl, hexyl, heptyl, octyl, nonyl, decyl and dodecyl.

20 Exemplary alkenyl groups include but are not limited to vinyl, propenyl, butenyl and hexenyl.

Exemplary alkynyl groups include but are not limited to ethynyl, 1-propynyl, 2-propynyl, 1 butynyl, and 2-butylnyl.

25 Exemplary cycloalkyl groups include but are not limited to cyclopentyl, cyclohexyl, and cyclooctyl substituents.

30 Exemplary aryl groups include but are not limited to phenyl, biphenyl, naphthyl, and anthracenyl.

Exemplary aralkyl groups include but are not limited to benzyl, phenethyl and phenbutyl.

35 The term "halohydrocarbyl" as used herein is inclusive of the hydrocarbyl moieties mentioned above but where there is a degree of halogenation that

can range from at least one hydrogen atom being replaced by a halogen atom (e.g., a fluoromethyl group) to where all hydrogen atoms on the hydrocarbyl group have been replaced by a halogen atom (e.g., trifluoromethyl or perfluoromethyl), also referred to as perhalogenation. For example, halogenated alkyl groups that can be useful in embodiments of the present invention can be partially or fully halogenated, alkyl groups of the formula C_aX_{2a+1} wherein X is independently a halogen or a hydrogen and a is selected from an integer of 1 to 25. In some embodiments each X is independently selected from hydrogen, chlorine, fluorine bromine and/or iodine. In other embodiments each X is independently either hydrogen or fluorine. Thus, representative halohydrocarbyls and perhalocarbyls are exemplified by the aforementioned exemplary hydrocarbyls where an appropriate number of hydrogen atoms are each replaced with a halogen atom.

In addition, the definition of the terms "hydrocarbyl", "halohydrocarbyl", and "perhalohydrocarbyl", are inclusive of moieties where one or more of the carbon atoms is replaced by a heteroatom selected independently from O, N, P, or Si. Such heteroatom containing moieties can be referred to as, for example, either "heteroatom-hydrocarbyls" or "heterohydrocarbyls", including, among others, ethers, epoxies, glycidyl ethers, alcohols, carboxylic acids, esters, maleimides, amines, imines, amides, phenols, amido-phenols, silanes, siloxanes, phosphines, phosphine oxides, phosphinites, phosphonites, phosphites, phosphonates, phosphinates, and phosphates.

Further exemplary hydrocarbyls, halohydrocarbyls, and perhalocarbyls, inclusive of heteroatoms, include, but are not limited to,

$-(CH_2)_f-Ar-(CH_2)_f-C(CF_3)_2-OH,$	$-(CH_2)_f-Ar-(CH_2)_f-OCH_2C(CF_3)_2-OH,$
$-(CH_2)_f-C(CF_3)_2-OH,$	$-((CH_2)_g-O)_h-(CH_2)-C(CF_3)_2-OH,$
$-(CH_2)_f-C(CF_3)(CH_3)-OH,$	$-(CH_2)_f-C(O)NHR^*,$
$-(CH_2)_f-C(O)Cl,$	$-(CH_2)_f-C(O)OR^*,$
$-(CH_2)_f-OR^*,$	$-(CH_2)_f-OC(O)R^*,$ and
$-(CH_2)_f-C(O)R^*,$ where f independently represents an integer from 0 to 12; g is 2, 3 or 4; h is 1, 2 or 3; Ar is aryl, for example phenyl; and R* independently represents hydrogen, a C ₁ -C ₁₁ alkyl, a C ₁ -C ₁₁ halogenated or perhalogenated alkyl, a C ₂ -C ₁₀ alkenyl, a C ₂ -C ₁₀ alkynyl, a C ₅ -C ₁₂ cycloalkyl, a C ₆ -C ₁₄ aryl, a C ₆ -	

C₁₄ halogenated or perhalogenated aryl, a C₇-C₁₄ aralkyl or a halogenated or perhalogenated C₇-C₁₄ aralkyl.

Exemplary perhalogenated alkyl groups include, but are not limited to, trifluoromethyl, trichloromethyl, -C₂F₅, -C₃F₇, -C₄F₉, C₆F₁₃-, -C₇F₁₅, and -C₁₁F₂₃. Exemplary halogenated or perhalogenated aryl and aralkyl groups include, but are not limited to, groups having the formula -(CH₂)_x-C₆F_yH_{5-y}, and -(CH₂)_x-C₆F_yH_{4-y}-pC_zF_qH_{2z+1-q}, where x, y, q and z are independently selected integers from 0 to 5, 0 to 5, 0 to 9 and 1 to 4, respectively. Specifically, such exemplary halogenated or perhalogenated aryl groups include, but are not limited to, pentachlorophenyl, pentafluorophenyl, pentafluorobenzyl, 4-trifluoromethylbenzyl, pentafluorophenethyl, pentafluorophenpropyl, and pentafluorophenbutyl.

In a preferred embodiment of the present invention, the norbornene-type polymer incorporates two or more distinct types of repeating units, i.e. the norbornene-type polymer is a copolymer, such as for example a random copolymer or a block copolymer.

In another preferred embodiment of the present invention, the norbornene-type polymer incorporates one or more distinct types of repeating units, where at least one such type of repeating unit encompasses pendant crosslinkable groups or moieties that have some degree of latency. By "latency", it is meant that such groups do not crosslink at ambient conditions or during the initial forming of the polymers, but rather crosslink when such reactions are specifically initiated, for example by actinic radiation or heat. Such latent crosslinkable groups are incorporated into the polymer backbone by, for example, providing one or more norbornene-type monomers encompassing such a pendant crosslinkable group, for example a substituted or unsubstituted maleimide or maleimide containing pendant group, to the polymerization reaction mixture and causing the polymerization thereof. Preferred crosslinkable groups include a group comprising a substituted or unsubstituted maleimide portion, an epoxide portion, a vinyl portion, an acetylene portion, an indenyl portion, a cinnamate portion or a coumarin portion, and more specifically a group selected from a 3-monoalkyl- or 3,4-

dialkylmaleimide, epoxy, vinyl, acetylene, cinnamate, indenyl or coumarin groups.

5 it is noted that two or more of groups R^{71} , R^{72} , R^{73} and R^{74} together may form a ring, such as a saturated (e.g. a cycloalkane) or non-saturated ring.

Preferably three of groups R^{71} , R^{72} , R^{73} and R^{74} are hydrogen, while only one of groups R^{71} , R^{72} , R^{73} and R^{74} is different from hydrogen and a carbyl group at each occurrence independently selected from the group consisting of

- 10 (i) alkyl having from 1 to 20, preferably from 1 to 15, more preferably from 1 to 10 carbon atoms;
- (ii) partially or fully halogenated, preferably fluorinated, alkyl having from 1 to 20, preferably from 1 to 15, more preferably from 1 to 10 carbon atoms;
- 15 (iii) alkyl having from 1 to 20, preferably from 1 to 15 and more preferably from 1 to 10 carbon atoms, wherein one or more carbon atom is replaced by the corresponding number of heteroatoms;
- (iv) partially or fully halogenated, preferably fluorinated, alkyl having from 1 to 20, preferably from 1 to 15, more preferably from 1 to 10 carbon atoms, wherein one or more carbon atom is replaced by the corresponding number
- 20 of heteroatoms;
- (v) groups of the following formula (VIII-a)



25 with j, k, l, m, R^{75} , R^{76} , X^1 and X^2 as defined herein; and

- (vi) latent crosslinkable groups selected from the group consisting of maleimide; maleimide substituted with one or more groups R^{78} , preferably alkyl having from 1 to 10, more preferably from 1 to 5 carbon atoms, and most preferably with methyl or ethyl; epoxide, vinyl, acetylene, indenyl, cinnamate, coumarin
- 30 and derivatives thereof, for example those wherein one or more hydrogen atoms are replaced by R^{78} ; and more preferably a group selected from 3-monoalkylmaleimide, 3,4-dialkylmaleimide, epoxy, vinyl, acetylene, cinnamate, indenyl, coumarin and derivatives thereof, for example those wherein one or more hydrogen atoms are replaced by R^{78} .
- 35

- 42 -

For the present application heteroatoms may generally be selected from the group consisting of O, S, N, P and Si, preferably from the group consisting of O, N, P and Si, and most preferably from the group consisting of O and Si.

5 j is at each occurrence independently an integer of from 0 to 5. Thus, j may at each occurrence independently be selected from the group consisting of 0, 1, 2, 3, 4, and 5. Preferably, j may at each occurrence independently be selected from the group consisting of 0, 1, 2, and 3. More preferably, j may at each occurrence independently be selected from the group consisting of 0, 1, and 2. Even more
10 preferably, j is at each occurrence independently 1 or 2. Most preferably, j is 1.

k, l and m are at each occurrence independently 0 or 1. If k, l or m is 1 then the respective group R^{75} , X^1 and X^2 is present and if 0 then the respective group is absent.

15 Thus, exemplary groups of formula (VIII-a) may at each occurrence independently be selected from the following group consisting of formulae (VIII-a-1) to (VIII-a-10)

	$-R^{76}$	(VIII-a-1)
20	$-R^{75}-R^{76}$	(VIII-a-2)
	$-X^1-R^{76}$	(VIII-a-3)
	$-X^1-R^{75}-R^{76}$	(VIII-a-4)
	$-R^{75}-X^2-R^{76}$	(VIII-a-5)
25	$-X^1-R^{75}-X^2-R^{76}$	(VIII-a-6)
	$-R^{75}-X^2-R^{75}-R^{76}$	(VIII-a-7)
	$-X^1-R^{75}-X^2-R^{75}-R^{76}$	(VIII-a-8)
	$-R^{75}-X^2-R^{75}-X^2-R^{76}$	(VIII-a-9)
30	$-R^{75}-X^2-R^{75}-X^2-R^{75}-R^{76}$	(VIII-a-10)

with R^{75} , R^{76} , X^1 and X^2 as defined herein.

X^1 and X^2 are at each occurrence independently selected from the group consisting of -O-, -C(=O)-, -C(=O)-O-, -S-, -NR⁷⁸-, -PR⁷⁸- and -Si(R⁷⁸)₂-, and are preferably
35

selected from the group consisting of -O-, -C(=O)-, and -C(=O)-O-, wherein R^{78} is as defined herein.

R^{75} is at each occurrence independently selected from the group consisting of

- 5 (i) alkanediyl having from 1 to 20, preferably from 1 to 15, more preferably from 1 to 10 carbon atoms;
- (ii) partially or fully halogenated, preferably fluorinated, alkanediyl having from 1 to 20, preferably from 1 to 15, more preferably from 1 to 10 carbon atoms;
- (iii) 1,4-phenylene and 1,4-phenylene wherein one or more hydrogen atoms are
10 substituted by a group R^{78} as defined herein;
- (iv) alkynyl (-C $\equiv$ C-) or $-(R^{78})C\equiv C(R^{78})-$; and
- (v) alkylidene groups $-(R^{78})C=C(R^{78})-$ with R^{78} as defined herein.

R^{76} is at each occurrence independently selected from the group consisting of

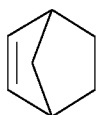
- 15 (i) methyl;
- (ii) partially or fully halogenated, preferably fluorinated, methyl;
- (iii) -Si(R^{77})₃ or -Si(OR⁷⁷)₃, wherein R^{77} is at each occurrence independently
selected from alkyl having from 1 to 10, preferably from 1 to 5 carbon atoms;
partially or fully halogenated, preferably fluorinated, alkyl having from 1 to
20 10, preferably from 1 to 5 carbon atoms; phenyl; phenyl wherein one or more
hydrogen atoms is replaced by a group R^{78} as defined herein;
- (iv) aryl having from 6 to 20 aromatic ring atoms and aryl having from 6 to 20
aromatic ring atoms wherein one or more hydrogen is replaced by a group
 R^{78} as defined herein; and
- 25 (v) a crosslinkable group selected from the group consisting of maleimide;
maleimide substituted with one or more groups R^{78} , preferably alkyl having
from 1 to 10, more preferably from 1 to 5 carbon atoms, and most preferably
with methyl or ethyl; maleimide having one or more (for example, 1, 2 or 3)
annealed aromatic, preferably 6-membered, rings such that the 3- and 4-
30 positions of the maleimide form part of one of the aromatic rings; epoxide,
vinyl, acetylene, indenyl, cinnamate, coumarin and derivatives thereof, for
example those wherein one or more hydrogen atoms are replaced by R^{78} ; and
more preferably a group selected from 3-monoalkylmaleimide, 3,4-
dialkylmaleimide, epoxy, vinyl, acetylene, cinnamate, indenyl, coumarin and
35 derivatives thereof, for example those wherein one or more hydrogen atoms
are replaced by R^{78} .

R⁷⁸ may at each occurrence independently be selected from the group consisting of halogen, preferably fluorine; alkyl having from 1 to 10, preferably from 1 to 5 carbon atoms, more preferably methyl; partially or fully halogenated, preferably fluorinated, alkyl having from 1 to 10, preferably from 1 to 5 carbon atoms, more preferably methyl; alkoxy having from 1 to 10, preferably from 1 to 5 carbon atoms, more preferably methoxy; and partially or fully halogenated, preferably fluorinated, alkoxy having from 1 to 10, preferably from 1 to 5 carbon atoms, more preferably methoxy.

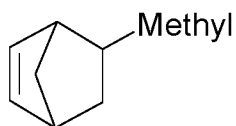
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Preferred norbornene monomers may at each occurrence independently be selected from the group consisting of formula (VIII-1) to (VIII-60)

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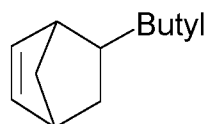


NB (VIII-1)

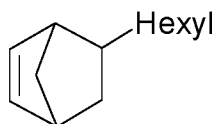


MeNB (VIII-2)

20

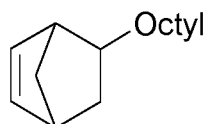


BuNB (VIII-3)

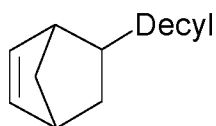


HexNB (VIII-4)

25

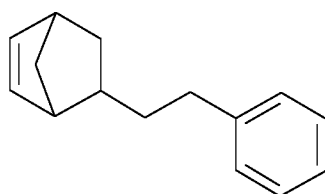


OctNB (VIII-5)



DecNB (VIII-6)

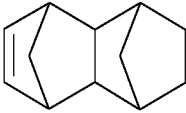
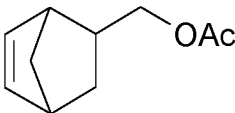
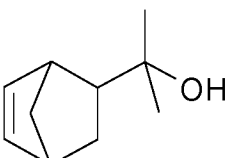
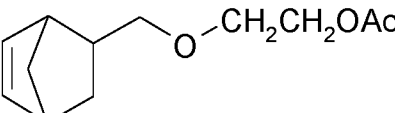
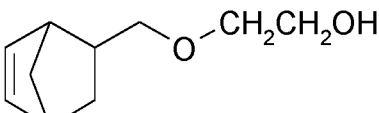
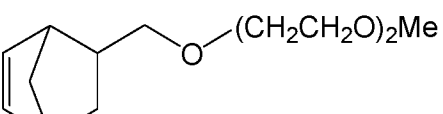
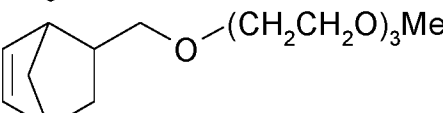
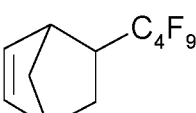
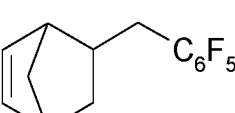
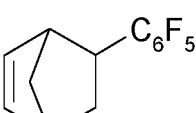
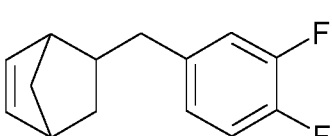
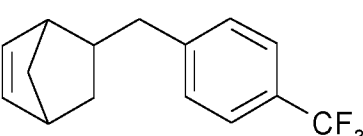
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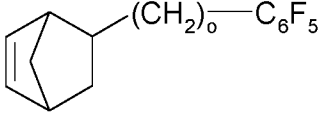
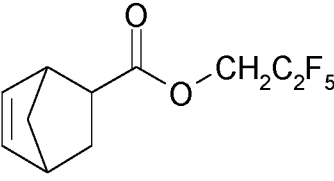
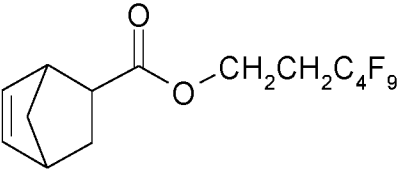
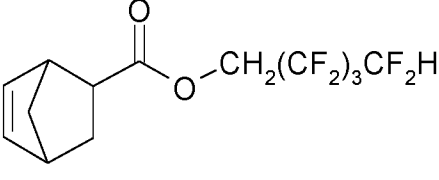
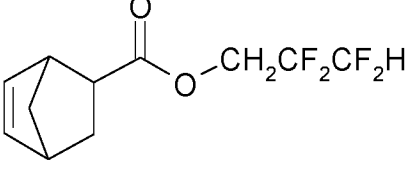
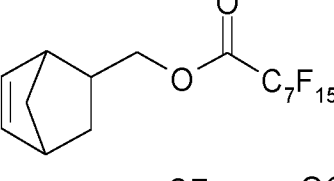
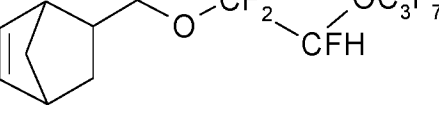
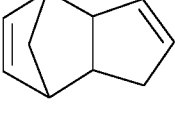
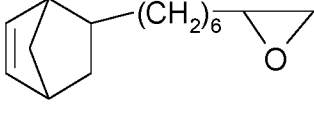
PENB (VIII-7)

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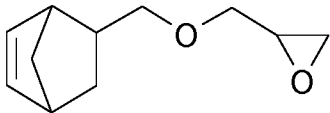
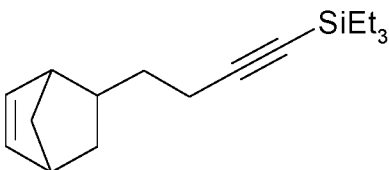
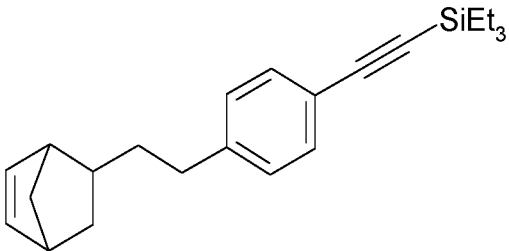
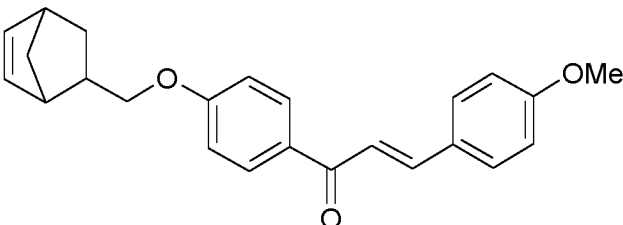
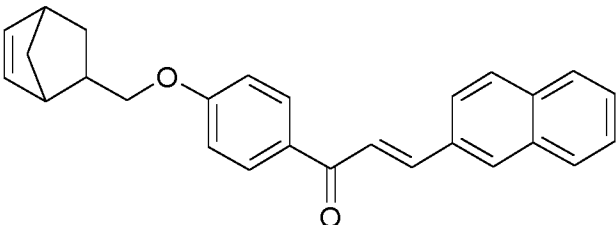
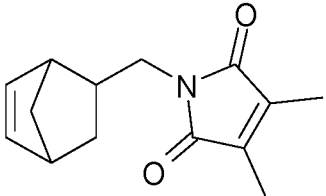
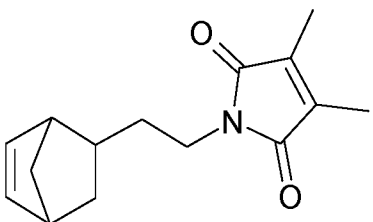
- 45 -

		TD	(VIII-8)
5		MeOAcNB	(VIII-9)
		NBXOH	(VIII-10)
10		NBCH2GlyOAc	(VIII-11)
		NBCH2GlyOH	(VIII-12)
15		NBTON	(VIII-13)
		NBTODD	(VIII-14)
20		NBC4F9	(VIII-15)
		NBCH2C6F5	(VIII-16)
25		NBC6F5	(VIII-17)
30		NBCH2C6H3F2	(VIII-18)
		NBCH2C6H4CF3	(VIII-19)
35			

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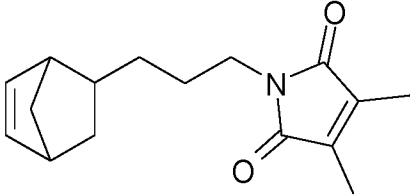
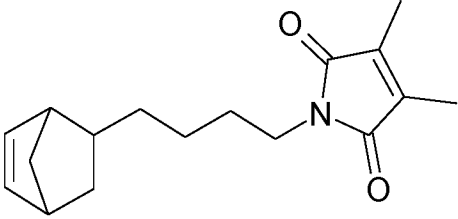
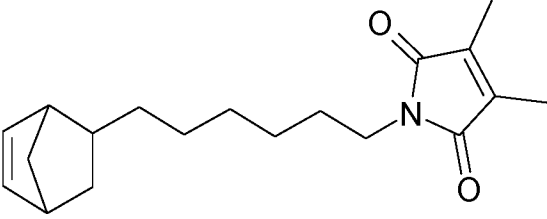
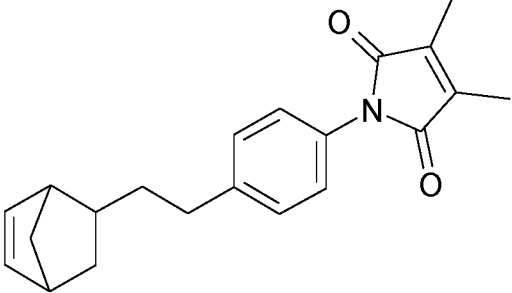
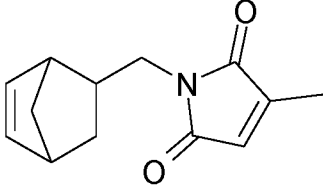
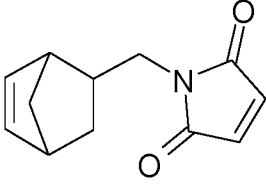
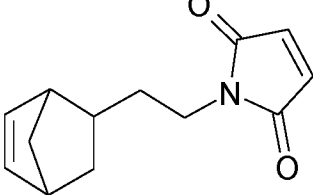
		NBalkylC ₆ F ₅	(VIII-20)
5		FPCNB	(VIII-21)
10		FHCNB	(VIII-22)
15		FOCHNB	(VIII-23)
20		FPCHNB	(VIII-24)
25		C ₈ PFAcNB	(VIII-25)
30		PPVENB	(VIII-26)
35		DCPD	(VIII-27)
		EONB	(VIII-28)

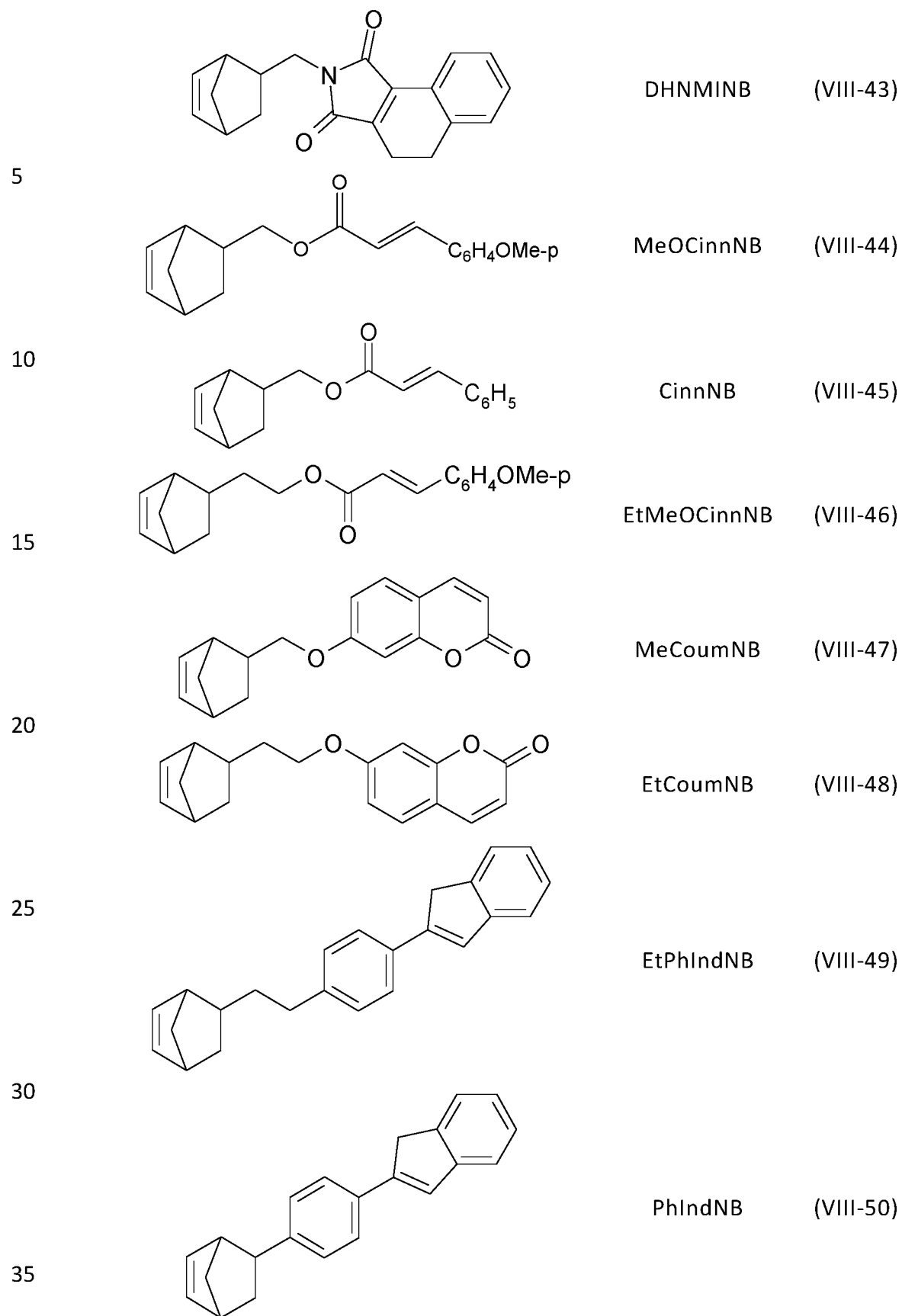
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		MGENB	(VIII-29)
5		AkSiNB	(VIII-30)
10		ArSiNB	(VIII-31)
15		MCHMNB	(VIII-32)
20		NPCHMNB	(VIII-33)
25		DMMIMeNB	(VIII-34)
30		DMMIEtNB	(VIII-35)

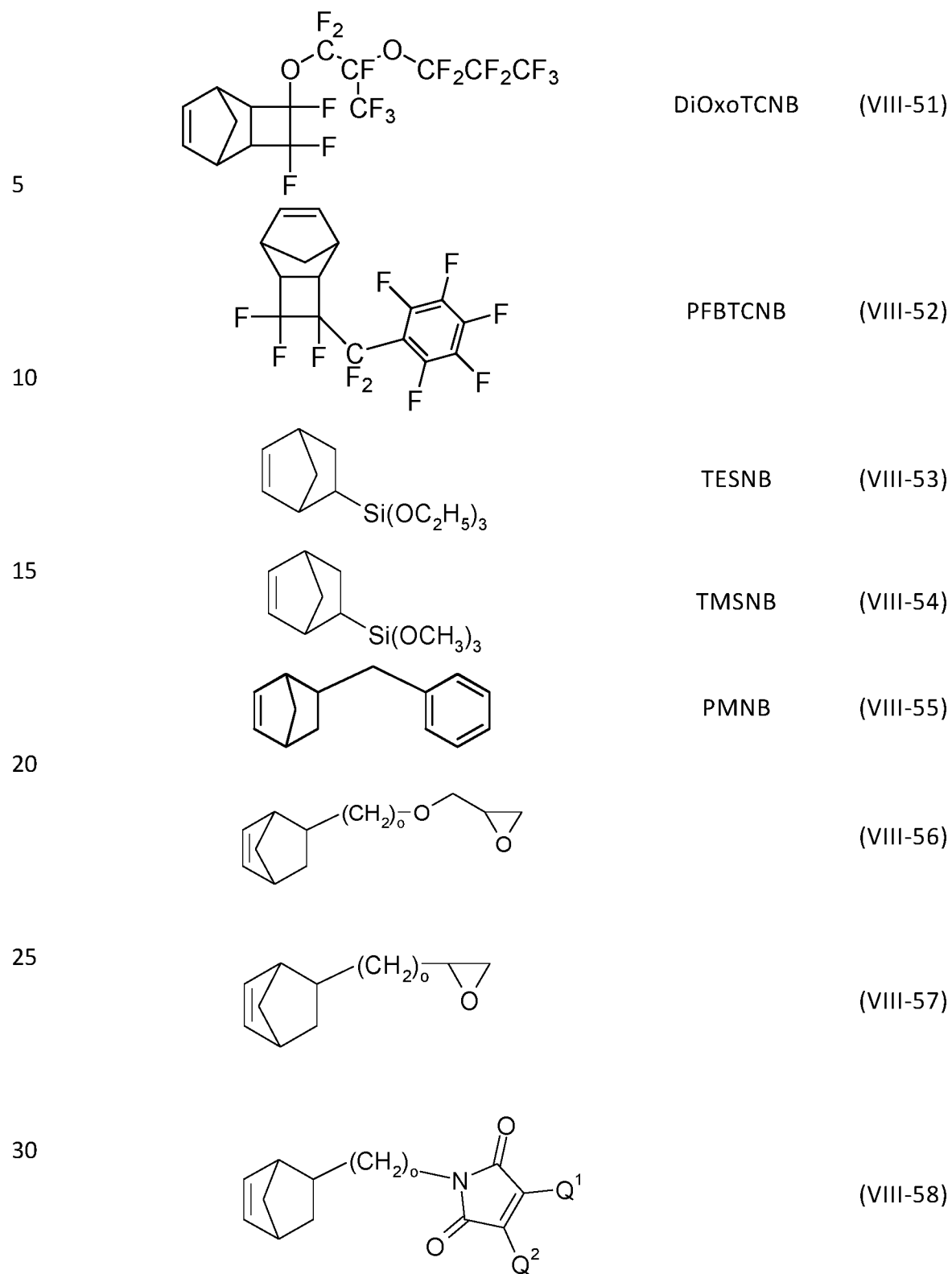
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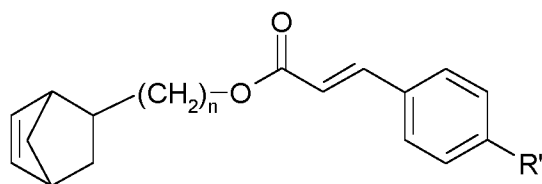
5		DMMIPrNB	(VIII-36)
10		DMMIBuNB	(VIII-37)
15		DMMIHxNB	(VIII-38)
20		EtPhDMMINB	(VIII-39)
25		MMIMeNB	(VIII-40)
30		MINB	(VIII-41)
35		MIENB	(VIII-42)



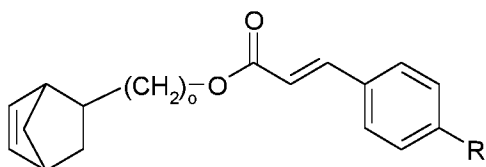
- 50 -



- 51 -



(VIII-59)



(VIII-60)

wherein "Me" stands for methyl, "Et" for ethyl, "OMe-p" for para-methoxy, "Ph" and "C₆H₅" for phenyl, "C₆H₄" for phenylene, "C₆F₅" for pentafluorophenyl, "OAc" for acetate, "PFAc" for -OC(O)-C₇F₁₅, o is an integer from 1 to 8, Q1 and Q2 are at each occurrence independently H or -CH₃; R' is H or -OCH₃; and for each of the above subformulae having a methylene bridging group (a CH₂ covalently bonded to both the norbornene ring and a functional group), including but not limited to (VIII-11) to (VIII-14), (VIII-16), (VIII-18), (VIII-19) and (VIII-55), it will be understood that the methylene bridging group can be replaced by a covalent bond or -(CH₂)_o- as in formula (VIII-20), with o then being an integer from 1 to 6.

Another preferred embodiment of the present invention is directed to polymers of Formula (VIII") that comprise repeating units where one of R⁷¹, R⁷², R⁷³ and R⁷⁴, for example R⁷¹, is a fluorinated or perfluorinated alkyl, aryl or aralkyl group as described above and the others of R⁷¹, R⁷², R⁷³ and R⁷⁴ are H, in which case preferably R⁷¹ is selected from one of formulae (VIII-15) to (VIII-26) (NBC₄F₉, NBCH₂C₆F₅, NBC₆F₅, NBCH₂C₆H₃F₂, NBCH₂C₆H₄CF₃, NBalkylC₆F₅, FPCNB, FHCNB, FOCHNB, FPCHNB, C₈PFAcNB, PPVENB), and more preferably from formulae (VIII-16), (VIII-17), (VIII-18), (VIII-19), (VIII-20)

or (VIII-26) (NBCH₂C₆F₅, NBC₆F₅, NBCH₂C₆H₃F₂, NBCH₂C₆H₄CF₃, NBalkylC₆F₅ or PPVENB).

5 Another preferred embodiment of the present invention is directed to polymers of Formula (VIII'') that have repeating units where one of R⁷¹, R⁷², R⁷³ and R⁷⁴, for example R⁷¹, is a photoreactive or crosslinkable group as described above and the others of R⁷¹, R⁷², R⁷³ and R⁷⁴ are H. Preferably R⁷¹ is a group as shown in one of the above subformulae (VIII-27) to (VIII-50) and more preferably as shown in subformulae (VIII-34), (VIII-35), (VIII-36), (VIII-37) and (VIII-38) (DMMIMeNB, DMMIEtNB, DMMIPrNB, DMMIBuNB and DMMIHxNB).

15 Another preferred embodiment of the present invention is directed to polymers of Formula VIII that have repeating units where one of R⁷¹, R⁷², R⁷³ and R⁷⁴, for example R⁷¹, is a polar group having a hydroxy, carboxy, acetoxy or oligoethyleneoxy moiety as described above and the others of R⁷¹, R⁷², R⁷³ and R⁷⁴ denote H. Preferably R⁷¹ is a group as shown in one of the above subformulae (VIII-9) to (VIII-14), and more preferably a group as shown in subformula (VIII-9) (MeOAcNB).

20 Another preferred embodiment of the present invention is directed to a polymer having a first type of repeating unit selected from fluorinated repeating units as described above and a second type of repeating unit selected from crosslinkable repeating units, also as described above.

25 Preferred polymers of this embodiment include polymers having a first type of repeating unit selected from subformulae (VIII-15) to (VIII-26), more preferably (VIII-15), (VIII-16), (VIII-17), (VIII-18), (VIII-19), (VIII-20) and (VIII-26) (NBC₄F₉, NBCH₂C₆F₅, NBC₆F₅, NBCH₂C₆H₃F₂, NBCH₂C₆H₄CF₃, NBalkylC₆F₅, and PPVENB) and a second type of repeating unit selected from subformulae (VIII-34), (VIII-35), (VIII-36), (VIII-37) and (VIII-38) (DMMIMeNB, DMMIEtNB, DMMIPrNB, DMMIBuNB and DMMIHxNB).

35 Another preferred embodiment of the present invention is directed to a polymer having a first type of repeating unit selected from fluorinated repeating units as described above, a second type of repeating unit selected from crosslinkable repeating units, also as described above and a third type

of repeating unit selected from polar repeating units, again as described above. Preferred polymers of this embodiment include polymers having a first repeating unit of formula (VIII-9) (MeOAcNB), a second type of repeating unit selected from the group consisting of formulae (VIII-34), (VIII-35), (VIII-36), (VIII-37), and (VIII-38) (DMMIMeNB, DMMIEtNB, DMMIPrNB, DMMIBuNB and DMMIHxNB), and a third type of repeating unit selected from formula (VIII-16) (NBCH₂C₆F₅).

Another preferred embodiment of the present invention is directed to a polymer having more than three different types of repeating units in accordance with formula (VIII). Another preferred embodiment of the present invention is directed to a polymer blend of a first polymer having a first type of repeating unit in accordance with formula (VIII'), and a second polymer having, at least, a first type of repeating unit and a second type of repeating unit of formula (VIII') that is distinct from the first type. Alternatively such polymer blends can encompass the aforementioned second polymer mixed with an alternative first polymer having two or more distinct types of repeat units in accordance with formula (VIII'). Further preferably, such polymer blends can encompass the aforementioned alternative first polymer mixed with an alternative second polymer having three distinct types of repeat units in accordance with formula (VIII').

Another preferred embodiment of the present invention is directed to a polymer having a first and a second distinct type of repeat units in accordance with formula (VIII') where the ratio of such first and second type of repeat units is from 95:5 to 5:95. In another preferred embodiment the ratio of such first and second type of repeat units is from 80:20 to 20: 80. In still another preferred embodiment the ratio of such first and second type of repeat units is from 60:40 to 40:60. In yet another preferred embodiment the ratio of such first and second type of repeat units is from 55:45 to 45:55.

Examples of suitable and preferred norbornene monomers, polymers and methods for their synthesis are provided herein and can also be found in US 5,468,819, US 6,538,087, US 2006/0020068 A1, US 2007/0066775 A1, US 2008/0194740 A1, WO 2012/028278 A1, US 9,583,713, WO 2012/028279 A1 and US 9,175,123. For example, exemplary polymerizations processes

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employing Group VIII transition metal catalysts are described in the aforementioned US 2006/0020068 A1.

5 Without wishing to be bound by theory, it is believed that direct contact between the piezoelectrically active composite layer and the second electrode layer needs to be avoided so as to obtain an electronic device with improved reliability and performance characteristics. Consequently, the present dielectric top layer needs to be continuous, i.e. cover essentially the complete contact surface of the piezoelectrically active composite layer and
10 the second electrode layer. The term "contact surface" is used to indicate that area, where the two layers would cover up each other were they in direct physical contact with each other.

15 Preferably, the dielectric top layer may be at least 2 nm and at most 300 nm thick.

Without wishing to be bound by theory it is assumed that where the insulating layer is thin enough, it may not entirely impede charge flow from the second electrode and into piezoelectric composite layer or from the piezoelectric
20 composite layer and into the top electrode, but permit charge carrier tunnelling by any combination of the known tunnelling mechanisms under appropriate biasing conditions and/or mechanical loading conditions, which conditions are known to a person skilled in the art. If the thickness of the insulating layer is within this preferred range, the electronic device of the present is well suited for an application as static force sensor even under small
25 mechanical loading conditions. Further advantageously, this allows comparatively higher voltage generation across the piezoelectric composite layer under dynamic force loading conditions, whilst minimising piezoelectric screening effects. Where the insulating layer becomes thicker then charge
30 flow can be prevented, and a field effect is generated over the insulating layer, thereby at least partially counteracting the desired effect of the present device.

35 For depositing the dielectric materials described herein, with the exception of a para(xylylene), such dielectric material may be dissolved in a solvent that is preferably selected from the group consisting of aliphatic hydrocarbons,

chlorinated hydrocarbons, aromatic hydrocarbons, ketones, ethers and mixtures thereof. More preferred examples of solvents are selected from the group consisting of alcohols, ethers, haloalkanes and any mixture of these.

5 Exemplary solvents which may be used include decane, dodecane, 1,2,4-trimethylbenzene, 1,2,3,4-tetra-methyl benzene, pentylbenzene, mesitylene, cumene, cymene, cyclohexylbenzene, diethylbenzene, tetralin, decalin, 2,6-lutidine, 2-fluoro-m-xylene, 3-fluoro-o-xylene, 2-chlorobenzotrifluoride, N,N-dimethylformamide, 2-chloro-6-fluorotoluene, 2-fluoroanisole, anisole, 2,3-
10 dimethylpyrazine, 4-fluoroanisole, 3-fluoroanisole, 3-trifluoro-methylanisole, 2-methylanisole, phenetol, 4-methylanisole, 3-methylanisole, 4-fluoro-3-methylanisole, 2-fluorobenzonitrile, 4-fluoroveratrol, 2,6-dimethylanisole, 3-fluorobenzo-nitrile, 2,5-dimethylanisole, 2,4-dimethylanisole, benzonitrile, 3,5-dimethyl-anisole, N,N-dimethylaniline, ethyl benzoate, 1-fluoro-3,5-dimethoxy-
15 benzene, 1-methylnaphthalene, N-methylpyrrolidinone, 3-fluorobenzo-trifluoride, benzotrifluoride, dioxane, trifluoromethoxy-benzene, 4-fluorobenzotrifluoride, 3-fluoropyridine, toluene, 2-fluoro-toluene, 2-fluorobenzotrifluoride, 3-fluorotoluene, 4-isopropylbiphenyl, phenyl ether, pyridine, 4-fluorotoluene, 2,5-difluorotoluene, 1-chloro-2,4-difluorobenzene, 2-fluoropyridine, 3-chlorofluoro-
20 benzene, 1-chloro-2,5-difluorobenzene, 4-chlorofluorobenzene, chloro-benzene, o-dichlorobenzene, 2-chlorofluorobenzene, p-xylene, m-xylene, o-xylene or mixture of o-, m-, and p-isomers. Examples of especially preferred solvents include, without limitation, dichloromethane, trichloromethane, chlorobenzene, o-dichlorobenzene, tetrahydrofuran, anisole, morpholine, toluene, o-xylene, m-
25 xylene, p-xylene, 1,4-dioxane, acetone, methylethylketone, 1,2-dichloroethane, 1,1,1-trichloroethane, 1,1,2,2-tetrachloroethane, ethyl acetate, n-butyl acetate, N,N-dimethylformamide, dimethylacetamide, dimethylsulfoxide, tetraline, decaline, indane, methyl benzoate, ethyl benzoate, mesitylene and/or mixtures thereof, preferably decane and dodecane.

30

The dielectric formulation according to the present invention can additionally comprise one or more further components or additives selected, for example, from surface-active compounds, lubricating agents, wetting agents, dispersing agents, hydrophobing agents, adhesive agents, flow improvers, defoaming agents,
35 deaerators, diluents which may be reactive or non-reactive, auxiliaries, colourants, dyes or pigments, sensitizers, stabilizers, nanoparticles or inhibitors.

SUBSTRATE

5 The present device may optionally comprise one or more substrates. Said substrate may, for example, be adjacent to and preferably substantially covering the first electrode layer or the second electrode layer or both, first electrode layer and second electrode layer, preferably on the surface opposite to the one adjacent to the piezoelectrically active composite layer.

10

The substrate used for the present electronic device is not particularly limited and may be any suitable material, preferably a material that is inert under use conditions. Examples of such materials are glass and polymeric materials. Preferred polymeric material include but are not limited to alkyd resins, allyl esters, benzocyclobutenes, butadiene-styrene, cellulose, cellulose acetate, epoxide, epoxy polymers, ethylene-chlorotrifluoro ethylene copolymers, ethylene-tetrafluoroethylene copolymers, fiber glass enhanced polymers, fluorocarbon polymers, hexafluoropropylenevinylidene-fluoride copolymer, high density polyethylene, parylene, polyamide, polyimide, polyaramid, polydimethylsiloxane, polyethersulphone, polyethylene, polyethylenenaphthalate, polyethyleneterephthalate, polyketone, polymethylmethacrylate, polypropylene, polystyrene, polysulphone, polytetrafluoroethylene, polyurethanes, polyvinylchloride, polycycloolefin, silicone rubbers, and silicones. Of these polyethyleneterephthalate, polyimide, polycycloolefin and polyethylenenaphthalate materials are more preferred. Additionally, for some embodiments of the present invention the substrate can be any suitable material, for example a polymeric material, metal or glass material coated with one or more of the above listed materials or coated with one or more metal, such as for example titanium. It will be understood that in forming such a substrate, methods such as extruding, stretching, rubbing or photochemical techniques can be employed to provide a homogeneous surface for device fabrication as well as to provide pre-alignment of an organic semiconductor material in order to enhance carrier mobility therein

30

35 Depending upon the specific application of the present electronic device it may be preferably that the substrate is sufficiently flexible so as not to crack when pressure

or excessive bending moment is applied. Thus, preferably the substrate is a polymeric material as defined above.

5 PRODUCTION METHOD

In general terms, the present device may be manufactured by a method comprising the following steps of

- (a) providing the first electrode layer as defined herein, optionally on a substrate,
- 10 (b) forming thereon the piezoelectrically active composite layer as defined herein,
- (c) applying the dielectric top material as defined herein onto the piezoelectrically active composite layer to form the dielectric top layer,
- (e) subsequently forming the second electrode layer,
- 15 thus obtaining the electronic device as described herein. Generally all of these layers may be applied and formed by standard methods well known to the skilled person.

With regards to the first electrode layer, the optional buffer layer, the organic
20 semiconducting layer, the dielectric top layer, and the second electrode layer, the method of applying the respective materials is not particularly limited. Exemplary methods include, but are not limited to, solution casting (e.g. spray-coating, dip-coating, web-coating, bar coating, screen printing, flexographic printing, gravure printing, or doctor blading) or vacuum deposition methods (e.g. physical vapor
25 deposition, chemical vapor deposition, or thermal evaporation), or sputtering methods (e.g. DC magnetron sputtering, electron beam sputtering).

Preferably the gate insulator layer is deposited, e.g. by spin-coating, doctor blading, wire bar coating, spray or dip coating or other known methods, from a formulation
30 comprising an insulator material and one or more solvents with one or more fluoro atoms (fluorosolvents), preferably a perfluorosolvent. A suitable perfluorosolvent is e.g. FC75® (available from Acros, catalogue number 12380). Other non-limiting examples of suitable fluoropolymers and fluorosolvents are known in the prior art, like for example the perfluoropolymers Teflon AF® 1600 or 2400 (from DuPont) or
35 Fluoropel® (from Cytonix) or the perfluorosolvent FC 43® (Acros, No. 12377).

The formation of the piezoelectrically active composite layer comprises the steps of forming on the first electrode layer the nanowire base layer as defined herein and then a plurality of piezoelectrically active nanowires as defined herein onto said base layer.

5

The plurality of nanowires may be produced by any suitable process, for example by a process selected from the group consisting of hydrothermal growth process, electrodeposition process, wet etching process, and non-chemical process. The plurality of nanowires is preferably formed by a hydrothermal growth process, more preferably by a low-temperature hydrothermal growth process at a temperature of at least 70 °C and of at most 100 °C.

10

In a first method, the nanowire base layer and the piezoelectrically active nanowires are formed in separate steps, wherein the nanowire base layer may first be formed by any suitable method, for example by spin coating, dip coating or a vacuum deposition method as already mentioned above in respect to the electrode layers, and then the nanowires may be grown onto the nanowire base layer by introducing the already formed layers (e.g. the nanowire base layer on the first electrode layer) into an autoclave to a solution wherein the material to be used for forming of the nanowires is dissolved.

15

20

In a second alternative method, the nanowire base layer and the nanowires are grown onto the first electrode layer by introducing into an autoclave the first electrode layer, optionally on a substrate, to a solution wherein the material to be used for forming the nanowires is dissolved.

25

Figures 3a, 3b and 3c are cross-sectional exemplary schematic views theoretically showing the operation principle of an electronic device according to an embodiment of the invention upon application of an external mechanical force.

30

Referring to **Figure 3a**, in the absence of an external mechanical force along the piezoelectric polarisation direction of the one-dimensional piezoelectric nanowires, no potential drop is induced across the device structure and thus no current will flow in the external circuit, which includes a resistive load. This corresponds to a zero voltage drop ($V = 0$) across the resistive load RL.

35

Referring to **Figure 3b**, upon the application of an external mechanical force $F(t)$, a piezoelectric potential is generated inside the piezoelectric nanowires comprising at least one semiconductor material having piezoelectric characteristics.

5 Consequently, the piezoelectric potential will induce equal and opposite mobile charge carriers on the two lateral surfaces of the first and second electrode layers. If negative immobile ions are exposed at the base portion of the plurality of piezoelectric nanowires, there will be positive mobile charges at the lateral surface of the first electrode layer. The process will lower the electrostatic potential at the

10 first electrode layer. The lateral second electrode layer atop the plurality of piezoelectric composite, which is separated by the insulating layer, will accumulate negative mobile charges, resulting in a decrease of the electrostatic potential. Consequently, the flow of charge carriers (equal and opposite in magnitude) towards the lateral second electrode leads to a transient current flow through the

15 external circuit. A potential gradient corresponding to a voltage drop ($V \neq 0$) is thus developed across the external electrical load (RL).

Referring to **Figure 3c**, dynamically removing the externally applied mechanical force $F(t)$ lowers the strain inside the piezoelectric composite layer, thereby

20 returning the piezoelectric composite structure to its pre-strained state. In turn, the induced piezoelectric polarisation inside the piezoelectric nanowires is also lowered. The piezoelectric potential diminishes to a zero value upon complete removal of the induced strain. To re-establish charge neutrality, the accumulated charges on the surfaces of the electrode layers are removed during this process.

25 The process of loading and unloading the device therefore results in the generation of an alternating current.

The electronic device of the invention comprising the piezoelectric composite layer and the insulating layer as defined herein is unique and offers a number of

30 attractive properties unmatched so far by commercial piezoelectric materials and makes it useful for targeting energy and force sensing technologies, including: (i) lead-free constituents; (ii) non-toxic and environmentally benign; (iii) low cost manufacturing steps, at plastic compatible temperatures; (iv) mechanically flexible components; (v) non-brittle; (vi) temperature stable, only limited by the choice of

35 organic semiconductor used; and (vii) high reliability and lifetime.

That is, the present inventors have found that the electronic device as disclosed herein comprising the piezoelectric composite layer and the insulating layer as defined herein can advantageously be operated in three distinctive modes:

- 5 (1) The present electronic device may serve as an effective vibrational electrical energy harvester under dynamic force actuation, making the present energy device useful as a sensor for impact detection.
- (2) The present electronic device is also found to effectively respond to static forces under appropriate low external biasing conditions, whereby the resistance across the piezoelectric composite is altered under static
10 mechanical compression, making the present electronic device useful for sensing static force pressures.
- (3) Furthermore, the electronic device as disclosed herein exhibits light harvesting/sensing properties when irradiated with light, such as sunlight, which makes the present electronic device useful as a photo/light sensor
15 and/or as an energy harvester.

In addition, the present electronic device may show vibration sensing properties, depending mainly on the substrate on which the device is elaborated, thus making it useful for the application as a vibration sensor. For example, to sense vibrations
20 where a fixed strain is applied and the actuating source has a high mechanical modulus of elasticity, it will be advantageous to adopt low aspect ratio piezoelectric metal oxide nanowires, which are comparatively more rigid, instead of a material with relatively low modulus such as longer more flexible piezoelectric nanowires, which will be more suited to less rigid actuating sources.

25 Thus, the electronic device according to the invention is useful for electronic apparatus, preferably as piezoelectric sensor or energy harvester. Therefore, the present application also relates to the use of the present electronic device in an electronic apparatus, preferably as piezoelectric sensor, more preferably as a
30 photo/light sensor, a force/pressure sensor or a vibration sensor, or as an energy harvester.

Accordingly, according to a further aspect, the present application also provides for an electronic apparatus comprising the present electronic device, which apparatus
35 is preferably a photo/light sensor, a force/pressure sensor or a vibration sensor, or an energy harvester.

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It will be appreciated that variations to the foregoing embodiments of the invention can be made while still falling within the scope of the invention. Each feature disclosed in this specification, unless stated otherwise, may be replaced by
5 alternative features serving the same, equivalent or similar purpose. Thus, unless stated otherwise, each feature disclosed is one example only of a generic series of equivalent or similar features.

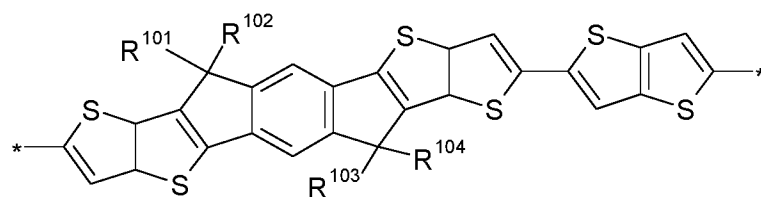
All of the features disclosed in this specification may be combined in any
10 combination, except combinations where at least some of such features and/or steps are mutually exclusive. In particular, the preferred features of the invention are applicable to all aspects of the invention and may be used in any combination. Likewise, features described in non-essential combinations may be used separately (not in combination).

15 The following examples are to illustrate the working and the advantages of the present electronic device in a non-limiting manner.

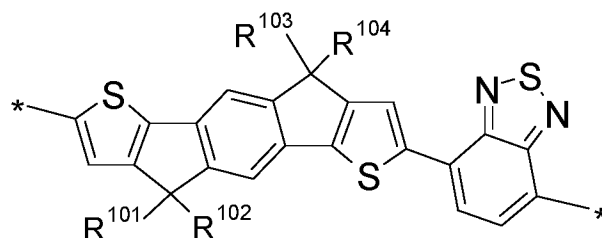
20 Examples

The organic semiconducting materials OSC-1 and OSC-2, the constitutional units of which are shown below, as well as OSC-3 shown below were synthesized according to published procedures.

25

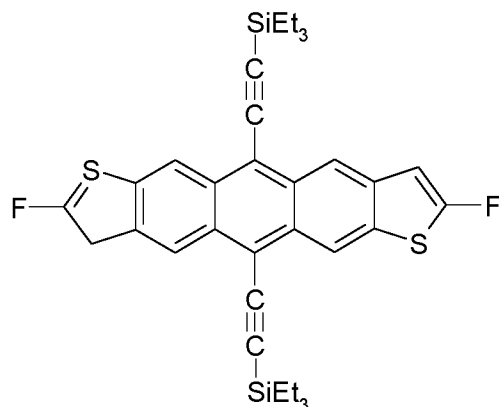


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OSC-3

Example 1 - Formation of nanowires by hydrothermal growth method using seeds

Step 1 - Before use as first electrode layer, a sample indium tin oxide (ITO) coated on polyethylene terephthalate (PET), 60 Ω /Sq. ITO X 7 Mil ST PET, obtained from Shieldahl, was cleaned by agitating in acetone, isopropyl alcohol and deionized water for 3-5 min between each cleaning step. The sample was then annealed on a hotplate at ca. 100°C and afterwards exposed to O₂ plasma for between 2 min and 5 min (30 sccm to 35 sccm O₂ flow rate).

Step 2 - Subsequently, 1.14 g zinc acetate hydrate (SigmaAldrich, 379786-25G, 99.999% purity) in 100 ml ethanol was applied by spin coating (1500 rpm, 30 s) to the freshly cleaned ITO layer. The coated substrate was then baked at ca. 120 °C for 2 to 3 min. This coating and baking cycle was repeated for at least 7 times before a final bake at 140 °C for 40 min, to obtain the first electrode layer with the nanowire base layer on top, in the following referred to as "seeded substrate".

Step 3 - A solution of ca. 0.5 to 2.5g zinc nitrate in 30 ml to 100 ml ultrapure water, a solution of 0.5 to 2.5 g of hexamethylenetetramine (SigmaAldrich, 398160-25G, 99.0 % purity) were combined into an autoclave reactor (Parr Instrument Company), around 0.2 to 1.5 ml of a dilute polyethyleneimine (PEI), prepared by mixing 0.2 g 1g of PEI (Sigma Aldrich: 408719-100ml, branched Average M_w #800) with around 50 ml to 100 ml of deionized water, and the seeded substrate were introduced into an autoclave reactor (Parr Instrument Company). The autoclave reactor was then sealed, placed in a pre-heated oven, and heated to around 90 °C

to 105 °C for 13 hours, thus obtaining the semi-finished device shown in Figure 6a in top view and in Figure 6b in cross-sectional view.

Example 2 – Nanowire formation by hydrothermal growth method without seed

5

Step 1 - As substrates, either glass or a Kapton film (IM301449 polyimide film, obtained from Goodfellow) were used. Either substrate was first cleaned by agitating in acetone, isopropyl alcohol and deionized water for 3-5 min between each cleaning step. A final cleaning step was performed using O₂ plasma for 3-5 min at 100% power.

10

Step 2 - As first electrode layer, a silver layer of 50-250 nm thickness was then deposited on each freshly cleaned sample using an Edward Auto 306 thermal evaporator system. For improved adhesion of the silver layer, an adhesion layer of Ti or Cr may be deposited beforehand, typically in a thickness between 2 nm and 20 nm.

15

Step 3 - A solution of 0.5-4.54 g zinc nitrate hexahydrate (Puratronic, 99.998% purity) in 5 ml to 60 ml of ultrapure deionized water and 5 ml to 30 ml of hexamethylenetetramine solution, prepared by dissolving 0.5-3.36 g of hexamethylenetetramine (SigmaAldrich, 398160-25G, 99.0 % purity) in 5 ml to 60 ml ultrapure deionized water as well as the silver-coated substrate were transferred to the autoclave reactor. Then PEI (4.5 ml, 7.5 vol%, 20 mmol) was added to the standard solution growth nutrient solution thereafter. Then, the autoclave is sealed and placed inside the preheated convection oven at between 70 °C and 92 °C for 6 h for reaction maturity, thus obtaining the semi-finished device shown in Figure 7a in top view and in Figure 7b in cross-sectional view..

20

25

Example 3 – Fabrication of a comparative test device

30

Step 1 - The semi-finished device of Example 1 or Example 2 was exposed to O₂ plasma (14-20 sccm, 0.49 mbar, 100 % power). Then, a formulation of OSC-1 in a blend of aromatic solvents was spin-coated thereon, first at 500-1000 rpm for 10 s at 1000 acceleration and secondly at 1000-2000 rpm for 10-60 s at 1000 acceleration, immediately followed by a bake on a hotplate at 100°C to 115 °C for

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between 10 min and 30 min. The spin coating steps and the baking step were then repeated twice.

5 Step 2 – The second electrode layer was then prepared by first applying a 2-20 nm thick layer of titanium, followed by a 50-250 nm thick layer of aluminum.

The resulting device had an active area of 3 cm by 2 cm.

10 Step 3 – Aluminum foil (0.5 cm by 0.5 cm) was placed on the aluminum layer of the second electrode layer, fixed there with graphite conductive adhesive (Alfa Aesar, no. 42466), and the solvent in the graphite adhesive was removed by heating inside a convection oven to ca. 90 °C for between 5 and 20 min. A piece of insulated wire was attached to the aluminum foil to act as the top contact lead. As bottom contact, 15 a piece of insulated wire was attached directly to the ITO of the first electrode layer using the graphite conductive adhesive (Alfa Aesar, no. 42466) to act as the bottom contact lead. The resulting devices were then again placed inside a convection oven at ca. 90 °C for 10-60 min so as to remove all solvent.

20 Step 4 – The so-produced devices were encapsulated using a partially crosslinked Dow Corning Sylgard 184 polydimethylsiloxane.

Example 4 – Fabrication of a comparative test device

25 The fabrication method of Example 3 was repeated except for OSC-2 being used instead of OSC-1.

Example 5 – Fabrication of a test device

30 A device produced as in Example 3, with the difference being that additional polymethylmethacrylate (1-4 g in 5-20 ml of 1,4-dioxane) was deposited by spin coating onto the organic semiconducting layer to form the dielectric top layer.

35

Example 6 – Fabrication of a test device

5 A device produced as in Example 4, with the difference being that additional polymethylmethacrylate (1-4 g in 5-20 ml of 1,4-dioxane) was deposited by spin coating onto the organic semiconducting layer to form the dielectric top layer.

Example 7 – Device performance

10 The device of Example 4 was tested for output voltage under
(a) incremental dynamic force loading conditions,
(b) incremental static force loading conditions under 0.3 V bias,
(c) dynamic impact excitation conditions, and
(d) dynamic impact excitation conditions and under parallelly configured electrical resistance loading conditions.

15 The results of testing under incremental dynamic force loading conditions are shown in Figure 8. It can be seen that increasing the dynamic loading conditions by application of an impact excitation force between 100 mN and 2 N led to an almost linear increase of the output voltage. This feature of the device not only shows our
20 invention to be a dynamic force sensing module, but also as an energy harvester. This is particularly attractive in applications where sharp dynamic forces may be present, such as in collision sensor applications. Moreover, the actuation mode demonstrates some advantages over polymer based piezoelectric materials, whose piezoelectric properties may slowly degrade under such measurement conditions.

25 The results of testing under incremental static force loading conditions under 0.3 V bias are shown in Figure 9. The output current was observed to increase with an incremental increase of the static load, with a linear region between 5 g and 20 g, saturating at higher static loadings. This behaviour of the invention is
30 uncharacteristic of piezoelectric materials, which may only operate under dynamic or quasi static conditions (with significant charge tracking electronics). This feature may find practical applications in artificial skin, touch panel display technologies, force sensing technologies, biomedical devices such as rehabilitation therapeutic devices to measure gesture, posture, gait etc..

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The results of testing under dynamic impact excitation conditions are shown in Figure 10. No external bias is applied to the device. Periodically, a 2N force is exerted on the device along the direction of polarisation in the piezoelectric nanostructures. The resulting responses are measured under optimal electrical loading conditions. A positive voltage response is detected under impact excitation. A negative voltage response is detected following the release of the mechanical load, which occurs immediately after impact. This feature of the device clearly demonstrates the energy harvesting capabilities of our multi-functional piezoelectric device described in this invention report.

10

The results of testing under dynamic impact excitation conditions and under parallelly configured electrical resistance loading conditions are shown in Figure 11. The plots show that higher voltages may be obtained under large resistive loading conditions at the expense of low current outputs (dynamic force sensing mode). Lower resistive loading conditions yields higher current and lower voltages (short circuit mode with high current surge). The cross over points of the two graphs signifies the matching resistive load conditions, the device operates optimally as an energy harvester.

15

20 Example 8 – Fabrication of a test device further comprising a buffer layer

A device produced as in Example 5 or in Example 6, with the difference that before depositing the organic semiconducting layer a buffer layer was deposited by spin coating polymethylmethacrylate (1-4 g in 5 – 20 ml of 1,4-dioxane) onto the organic semiconducting layer. Its peak-peak output voltage and current are shown in Figure 12 in comparison to a device produced as in Example 5 or Example 6. The data clearly shows the advantages of the additional buffer layer.

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Claims

1. Electronic device comprising
 - (i) a first electrode layer,
 - 5 (ii) a second electrode layer,
 - (iii) a piezoelectrically active composite layer between the first electrode and the second electrode, the piezoelectrically active composite layer comprising
 - 10 (iii-1) a plurality of piezoelectrically active nanowires on a base layer of the same material, and
 - (iii-2) one or more organic semiconducting layer dispersed in between the nanowires, each organic semiconducting layer comprising one or more organic semiconducting materials,
 - and
 - 15 (iv) an electrically insulating top layer between the piezoelectrically active composite layer and the second electrode.
2. Electronic device according to claim 1, wherein the one or more organic semiconducting material comprises an organic p-type semiconducting material, and the nanowires comprise an n-type semiconducting material; or
20 wherein the one or more organic semiconducting material comprises an organic n-type semiconducting material, and the piezoelectric nanowires comprise a p-type semiconducting material.
- 25 3. Electronic device according to any one or more of the preceding claims, wherein the nanowires comprise one or more inorganic piezoelectric material selected from Group III-V and II-VI compounds.
4. Electronic device according to one or more of the preceding claims, wherein
30 the device further comprises a buffer layer, which is electrically insulating, between the nanowire base and the one or more semiconducting layer.
5. Electronic device according to any one or more of the preceding claims, wherein the electrically insulating top layer and the buffer layer
35 independently of each other comprise one or more dielectric materials

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selected from the group consisting of organic dielectric materials and inorganic dielectric materials.

- 5 6. Electronic device according to any one or more of the preceding claims, wherein the electrically insulating top layer and the buffer layer independently of each other comprise an inorganic dielectric material selected from the group consisting of consisting of silicon oxide, silicon nitride, hafnium silicate, zirconium silicate, metal oxides, preferably tantalum oxide, aluminum oxide, titanium dioxide, hafnium dioxide, barium titanate, 10 strontium titanate, barium strontium titanate, barium zirconium titanate and zirconium dioxide, and any blend of any of these.
- 15 7. Electronic device according to any one or more of the preceding claims, wherein the electrically insulating top layer and the buffer layer independently of each other comprise an organic dielectric material selected from the group consisting of polystyrene (PS), polyvinyl alcohol (PVA), poly(p-xylylene), polyvinylphenol (PVP), polyacrylate (PA), polymethylacrylate (PMA), polymethylmethacrylate (PMMA), polyvinylchloride (PVC), polyvinylidene fluoride (PVDF), poly(α -methylstyrene) (P α MS), 20 cyanoethylpullalan (CYPEL), polyimide, polycycloolefinic polymers, fully or partially fluorinated polymers, and any blend of any of these.
- 25 8. Apparatus comprising the electronic device of any one or more of claims 1 to 7.
9. Apparatus according to claim 8, wherein the apparatus is a piezoelectric sensor.
- 30 10. Apparatus according to claim 8 or claim 9, wherein the apparatus is selected from the group consisting of photo/light sensor, force/pressure sensor, vibration sensor, and energy harvester.
- 35 11. Method of manufacturing the electronic device of any one or more of claims 1 to 7, said method comprising the steps of
- (a) providing the first electrode layer,
- (b) forming the piezoelectrically active composite layer by

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- (i) forming on the first electrode layer the nanowire base layer and then a plurality of piezoelectrically active nanowires onto said base layer, and
- (ii) depositing one or more organic semiconducting material onto said piezoelectrically active nanowires, to obtain the piezoelectrically active composite layer,
- (c) subsequently depositing one or more electrically insulating material, to obtain the electrically insulating top layer, and
- (d) then deposit the second electrode layer.
12. Method according to claim 11, wherein the one or more organic semiconducting material comprises an organic p-type semiconducting material, and the nanowires comprise an n-type semiconducting material; or wherein the one or more organic semiconducting material comprises an organic n-type semiconducting material, and the piezoelectric nanowires comprise a p-type semiconducting material.
13. Method according to claim 11 or claim 12, wherein the nanowires comprise one or more inorganic piezoelectric material selected from Group III-V and II-VI compounds.
14. Method according to any one or more of claims 11 to 13, wherein in step (b) the piezoelectrically active nanowires are formed by a process selected from the group consisting of hydrothermal growth process, electrodeposition process, wet etching process, and non-chemical process.
15. Method according to any one or more of claims 11 to 14, wherein in step (b) the piezoelectrically active nanowires are formed by a hydrothermal growth process, preferably at a low temperature.
16. Method according to any one or more of claims 11 to 15, wherein in step (b) the nanowire base layer and the piezoelectrically active nanowires are formed by deposition from a solution comprising a soluble zinc salt, preferably from a solution comprising zinc acetate or zinc nitrate.

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17. Method according to any one or more of claims 11 to 16, wherein step (b)(i) is followed by step (b)(i') depositing onto the piezoelectrically active nanowires one or more dielectric materials to obtain the buffer layer.
- 5 18. Method according to any one or more of claims 11 to 17, wherein the electrically insulating material at each occurrence independently comprises one or more dielectric materials selected from the group consisting of organic dielectric materials and inorganic dielectric materials.
- 10 19. Method according to any one of more of claims 11 to 18, wherein the electrically insulating material at each occurrence independently comprises an inorganic dielectric material selected from the group consisting of consisting of silicon oxide, silicon nitride, hafnium silicate, zirconium silicate, metal oxides, preferably tantalum oxide, aluminum oxide, titanium dioxide, 15 hafnium dioxide, barium titanate, strontium titanate, barium strontium titanate, barium zirconium titanate and zirconium dioxide, and any blend of any of these.
- 20 20. Method according to any one or more of claims 11 to 19, wherein the electrically insulating material at each occurrence independently comprises an organic dielectric material selected from the group consisting of polystyrene (PS), polyvinyl alcohol (PVA), poly(p-xylylene), polyvinylphenol (PVP), polyacrylate (PA), polymethylacrylate (PMA), polymethylmethacrylate (PMMA), polyvinylchloride (PVC), polyvinylidene fluoride (PVDF), poly(α -methylstyrene) (P α MS), cyanoethylpullalan (CYPEL), polyimide, 25 polycycloolefinic polymers, fully or partially fluorinated polymers, and any blend of any of these.
- 30 21. Use of the electronic device of any one or more of claims 1 to 7 as one selected from the group consisting of piezoelectric sensor, photo/light sensor, force/pressure sensor, vibration sensor, and energy harvester.
- 35 22. Method of using the electronic device of any one or more of claims 1 to 7 as one selected from the group consisting of piezoelectric sensor, photo/light sensor, force/pressure sensor, vibration sensor, and energy harvester.

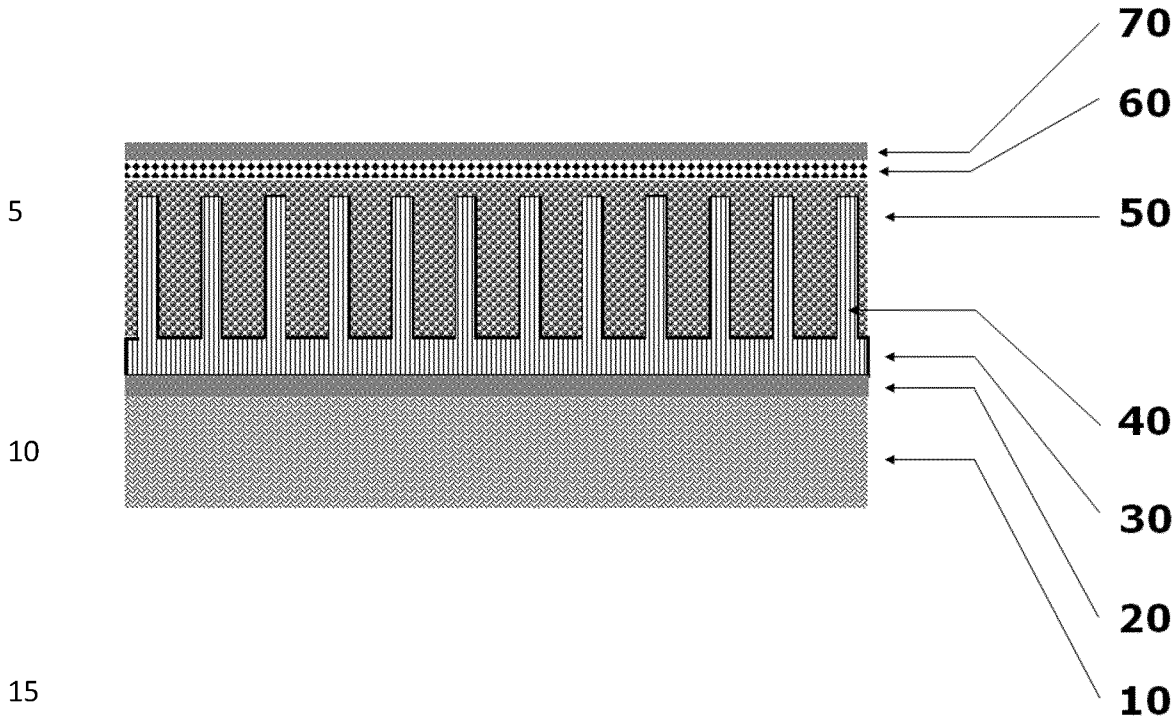


Figure 1

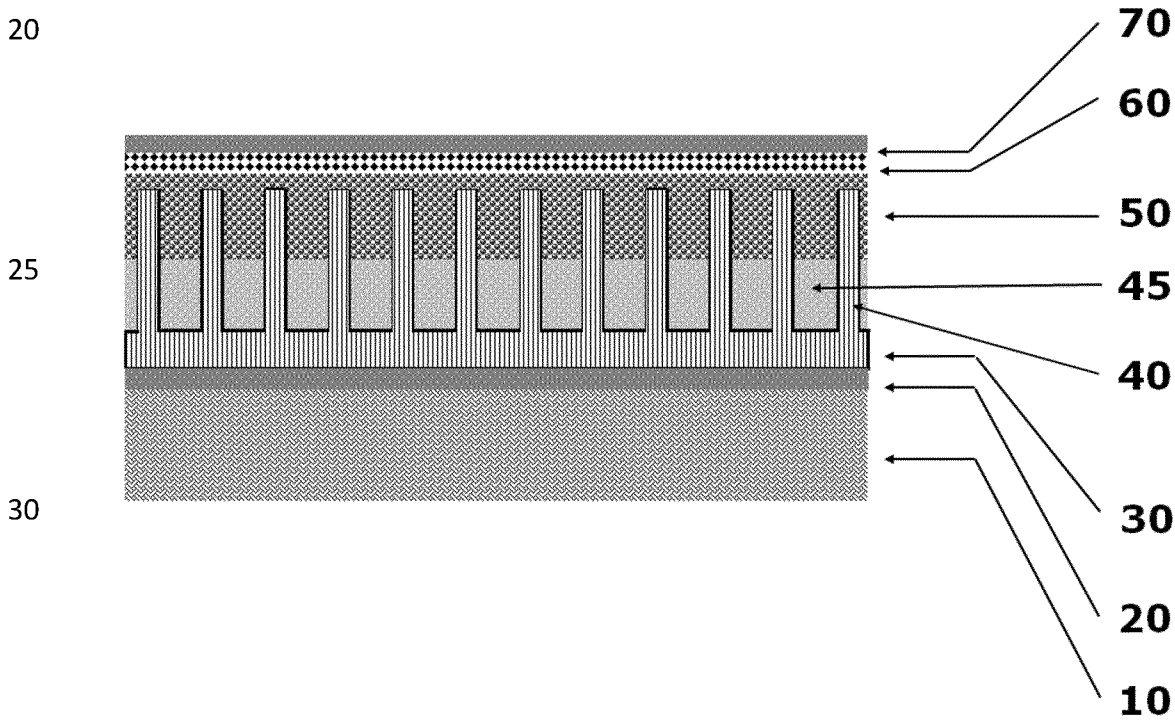


Figure 2

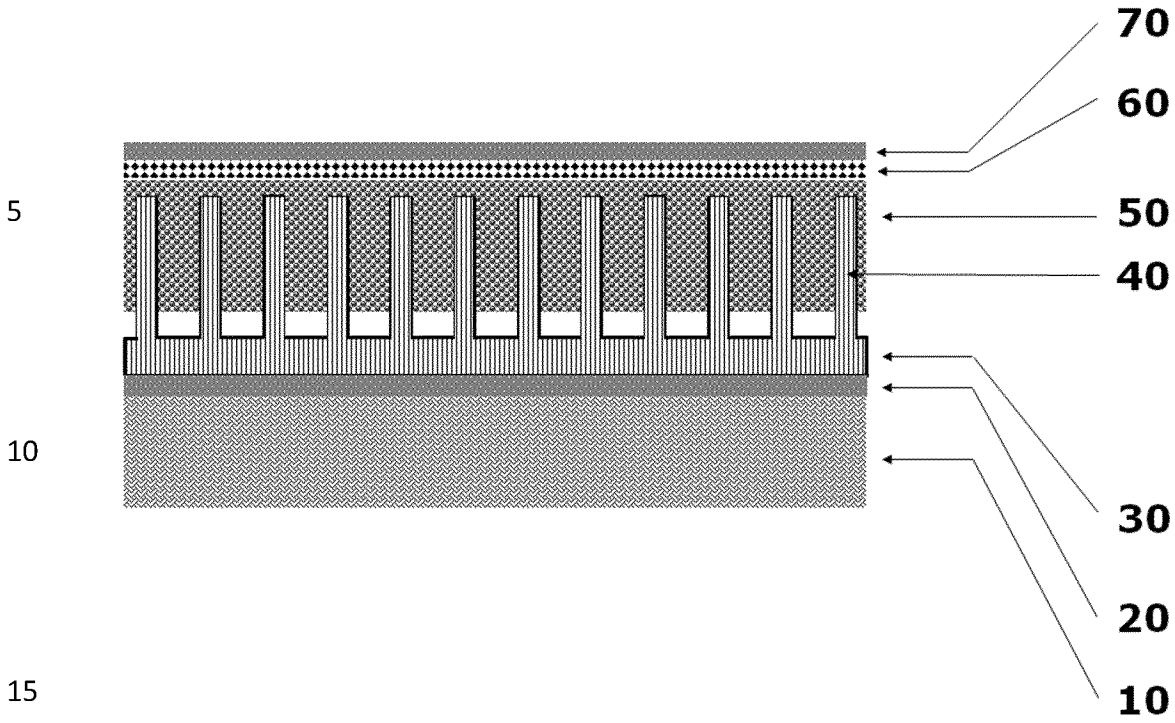


Figure 3

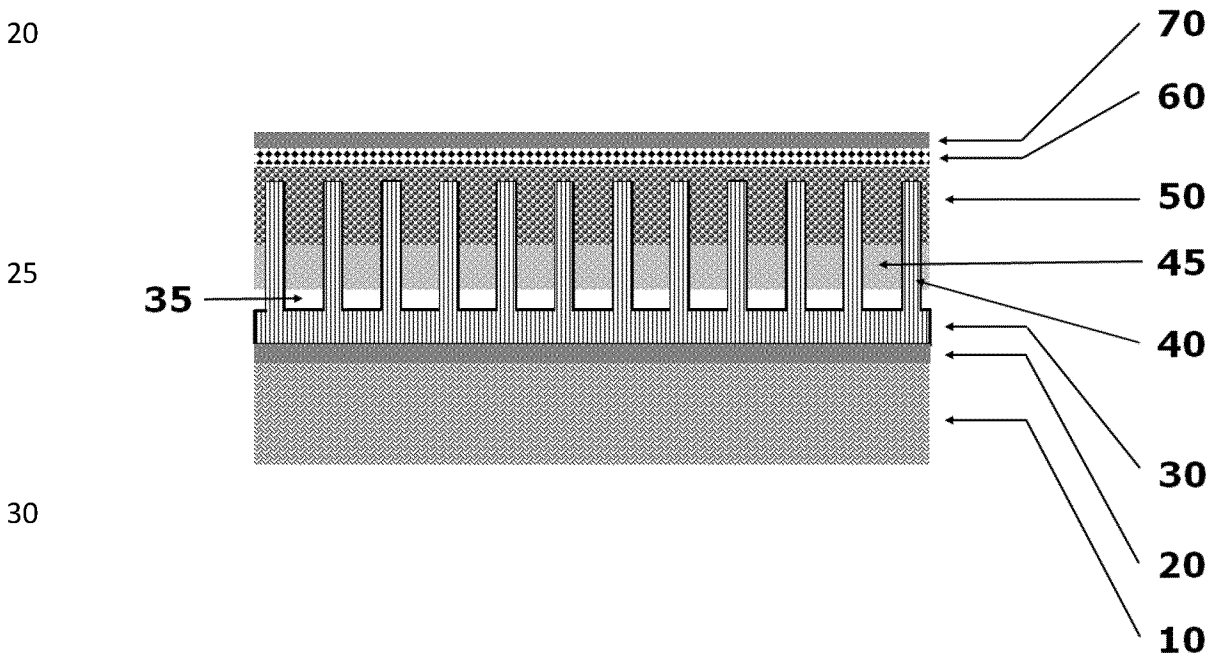


Figure 4

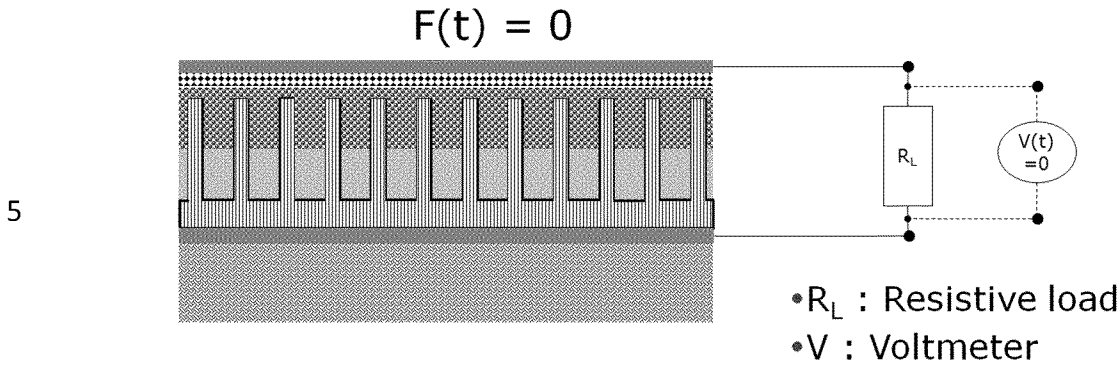


Figure 5a

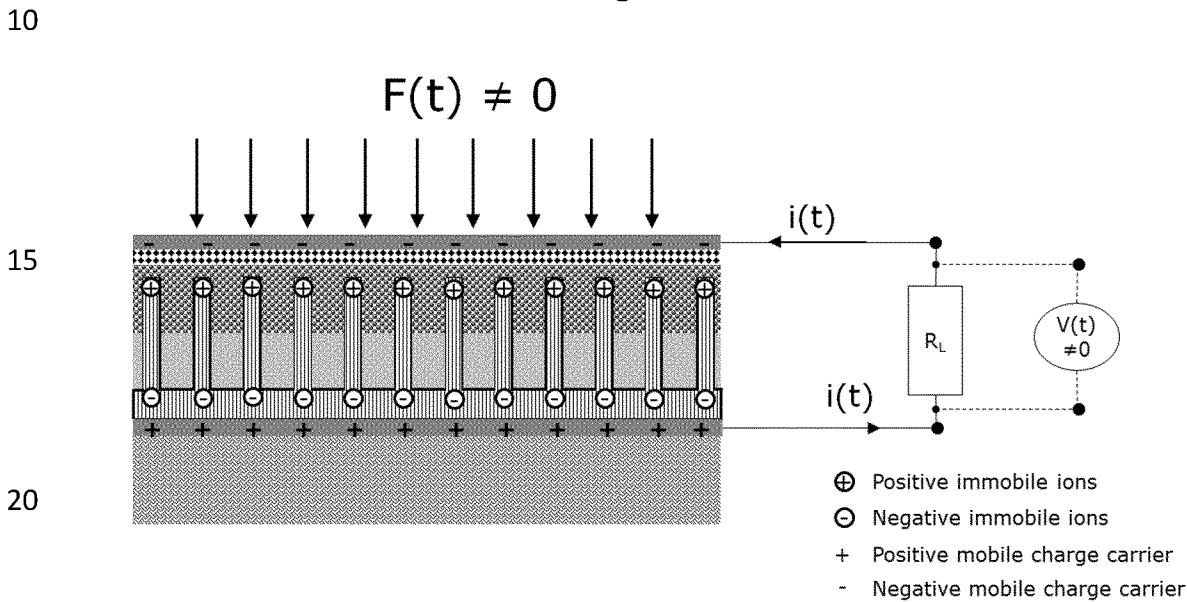


Figure 5b

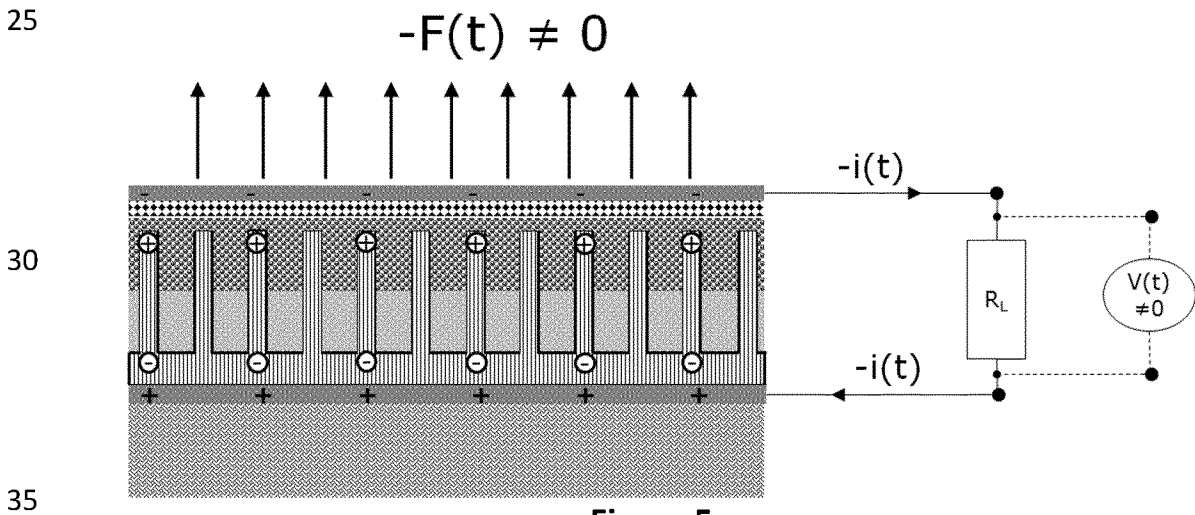


Figure 5c

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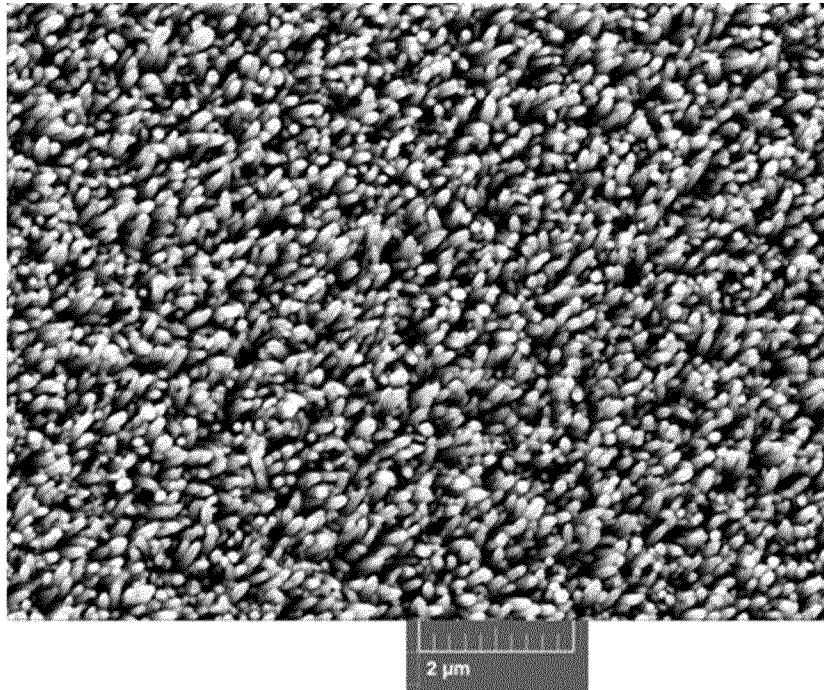


Figure 6a

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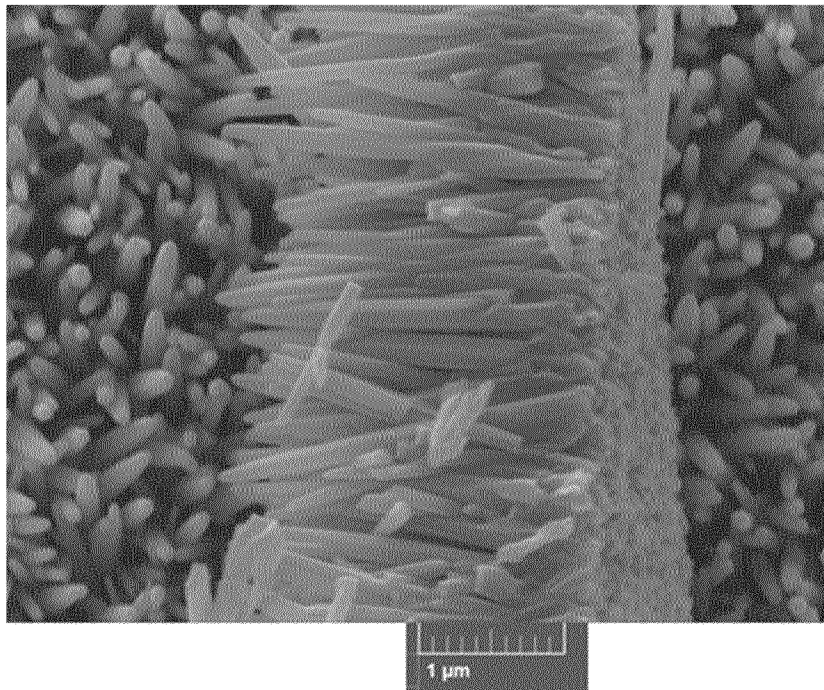


Figure 6b

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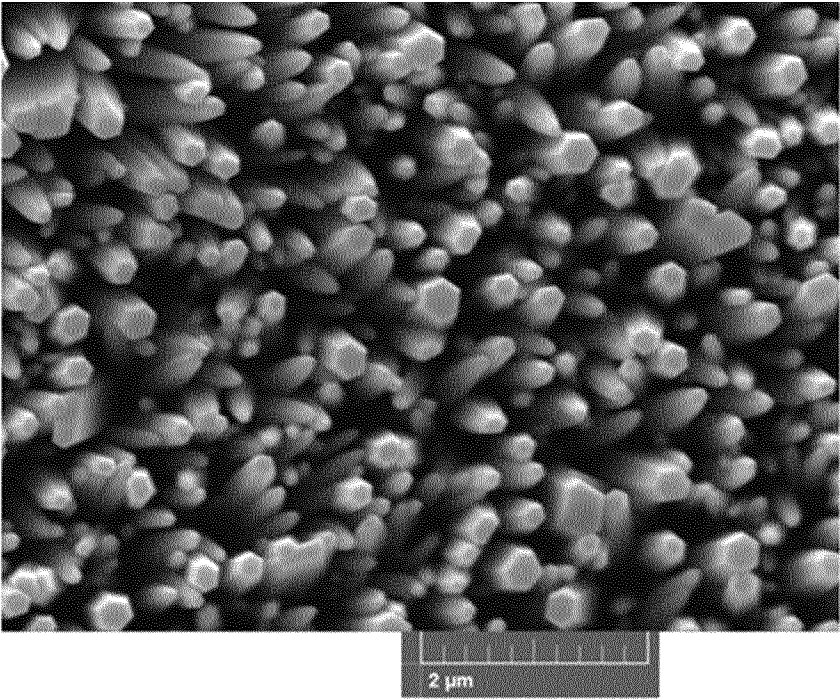


Figure 7a

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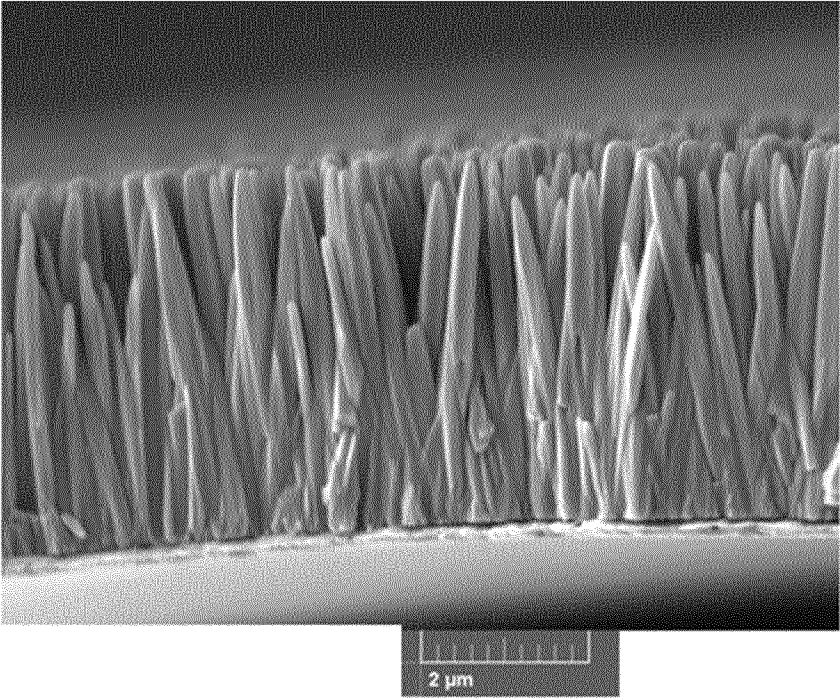


Figure 7b

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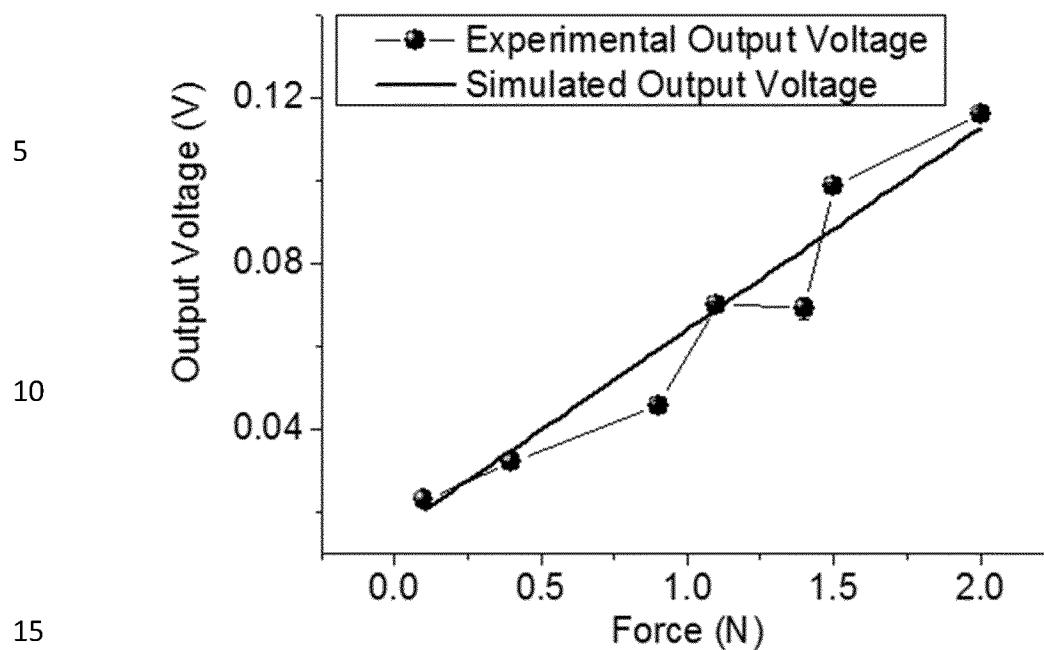


Figure 8

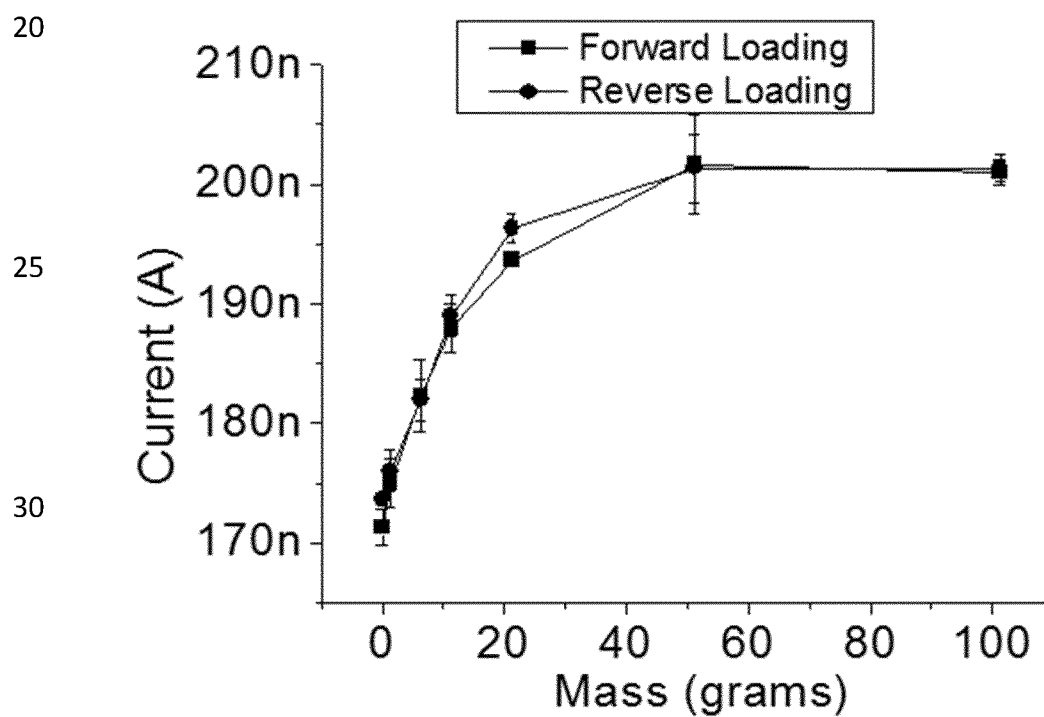


Figure 9

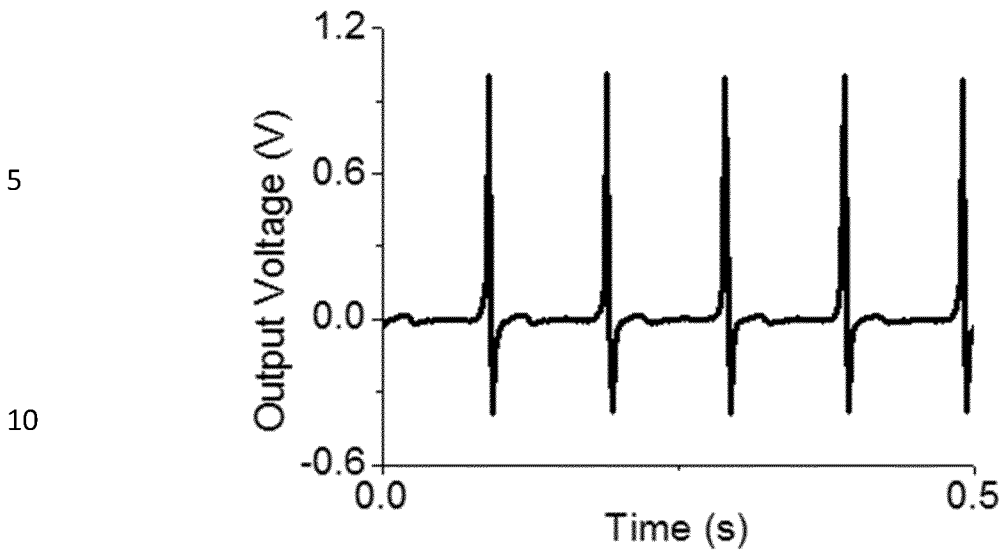


Figure 10

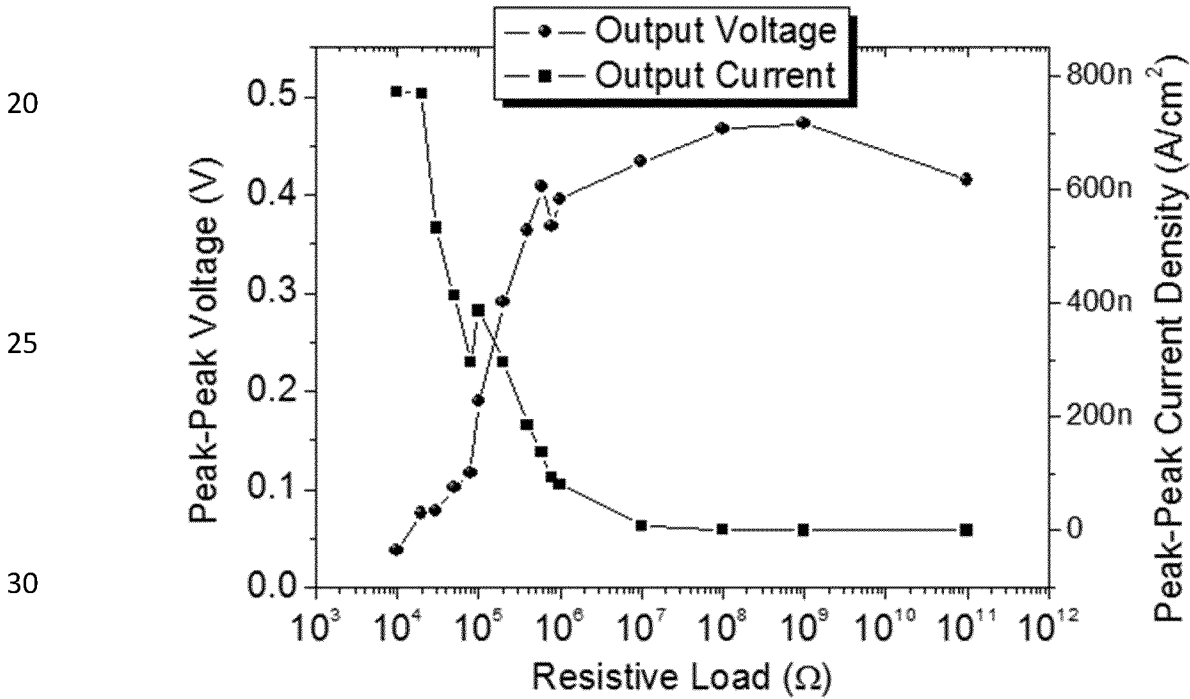
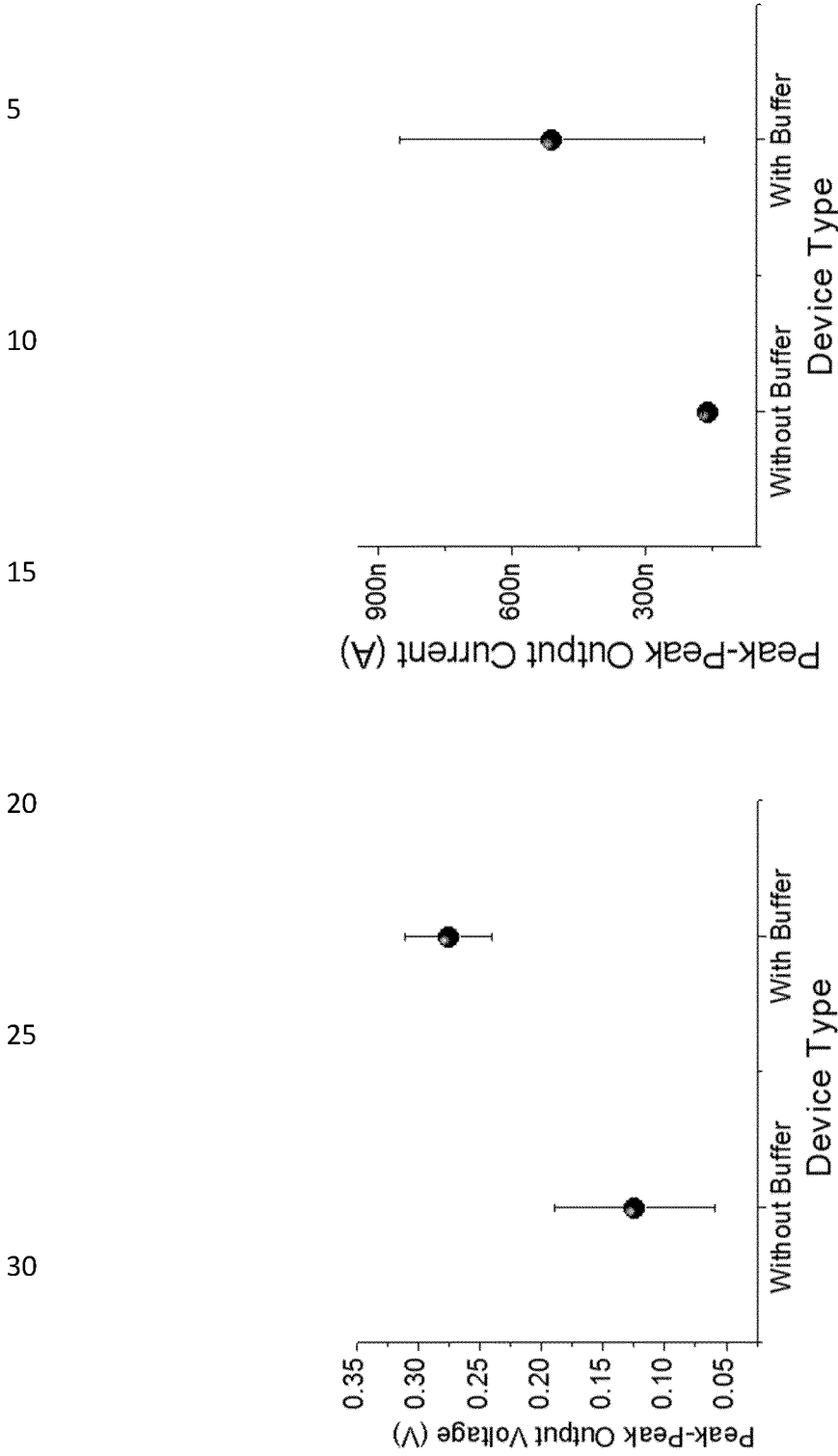


Figure 11



Devices Measured under near identical conditions
2Hz (sawtooth excitation); 2N Force.

Figure 12

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2020/053631

A. CLASSIFICATION OF SUBJECT MATTER

INV. H01L41/18 H01L41/113 H02N2/18 H01L41/35 H01L51/42
ADD.

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

H01L H02N

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	CN 106 653 793 A (BEIJING INST NANOENERGY & NANOSYSTEMS) 10 May 2017 (2017-05-10) the whole document	1,2,11, 12,16
Y	US 2011/101315 A1 (CHOI DUK-HYUN [KR] ET AL) 5 May 2011 (2011-05-05) paragraph [0062] - paragraph [0080]; figure 4	1,2,11, 12
Y	US 2016/181952 A1 (KIM SEONGMIN [KR] ET AL) 23 June 2016 (2016-06-23) paragraph [0075] - paragraph [0076]; figure 3	16



Further documents are listed in the continuation of Box C.



See patent family annex.

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Date of the actual completion of the international search

7 April 2020

Date of mailing of the international search report

20/04/2020

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Authorized officer

Angermeier, Detlef

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/EP2020/053631

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