Printed electronic device comprising a substrate onto at least one surface of which has been applied a layer of an electrically conductive ink comprising functionalized graphene sheets and at least one binder. A method of preparing printed electronic devices is further disclosed.

25 Claims, 1 Drawing Sheet
### U.S. PATENT DOCUMENTS

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PRINTED ELECTRONICS

This invention was made with Government support under Grant No. CMS-0609049, awarded by the National Science Foundation, and under Grant No. NCC1-02037, awarded by NASA. The Government has certain rights in the invention.

FIELD OF THE INVENTION

The present invention relates to printed electronic devices and methods for their manufacture.

BACKGROUND

Printed electronics are increasingly finding uses in a great variety of applications, including portable electronics, signage, lighting, product identification, packaging flexible electronic devices (such as those that can be rolled or bent), photovoltaic devices, medical and diagnostic devices, antennas (including RFID antennas), displays, sensors, thin-film batteries, electrodes and myriad others. Printed electronics have a variety of advantages over electronics made using other methods, including subtractive methods. Printing can be faster than normal subtractive methods (such as etching) and can generate less waste and involve the use of fewer hazardous chemicals than in such methods. The resulting electronics can be more readily used in flexible devices, such as displays, that are designed to be rolled, twisted, bent, or subjected to other distortions during use.

Printed electronics are typically made by printing the electronic circuit or other component on a substrate using an electrically conductive metal-based ink. The inks typically contain silver particles, and occasionally copper particles, other metallic particles, and/or conductive polymers. However, conductive polymers alone are generally not sufficiently electrically conductive. Furthermore, the resulting printed metallic circuits are usually insufficiently electrically conductive to be effective in most applications, including in devices in which the circuits are regularly stressed by bending and/or stretching during use. The printed substrates must therefore often be heated at elevated temperatures to sinter the conductive metal particles in order to achieve the desired levels of electrical conductivity. The temperatures used in sintering processes frequently limit the substrates that can be selected for the preparation of the electronics. For example, while it would be desirable to use inexpensive materials such as paper, polyolefins (e.g., polypropylene), and the like as substrates for printed electronics in many applications, the sintering temperatures often required are too high to be used with paper.

Furthermore, silver is costly and other, non-precious, metals can form oxides upon exposure to the environment that can render the material insufficiently conductive for the application. Additionally, the use of metal-based inks can add weight to the resulting device, and the aforementioned sintering process can add one or more additional steps, time, and complexity to the fabrication process. It would thus be desirable to obtain printed electronic devices using inks that do not contain costly precious metals, that are lighter weight, and that do not require sintering to become sufficiently electrically conductive, and that could therefore be used on a wider variety of substrate materials, including paper and polyolefins such as polyethylene.


SUMMARY OF THE INVENTION

Disclosed and claimed herein is a printed electronic device, comprising a substrate comprising at least one surface, wherein a layer of an electrically conductive ink has been applied to a portion of the surface, and wherein the ink comprises functionalized graphene sheets and at least one binder. Further disclosed and claimed herein is a method for forming the printed electronic device.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is an optical microscope image of electrically conductive lines printed from an ink comprising functionalized graphene sheets embedded into poly(ethylene oxide).

DESCRIPTION

As used herein, the term “electrically conductive ink” encompasses materials comprising electrically conductive materials suspended and/or dissolved in a liquid, as well as pastes and materials in substantially solid form containing little or no liquids. Electrically conductive inks may be free-flowing, viscous, solid, powdery, and the like. The term “ink” is used to refer to an ink in a form that is suitable for application to a substrate, as well as the material after it is applied to a substrate both before and after any post-application treatments (such as evaporation, cross-linking, curing etc.).

The printed electronic devices (also referred to herein as “printed electronics”) of the present invention may be in the form of complete devices, parts or sub elements of devices, electronic components, and the like. They comprise a substrate onto at least one surface of which has been applied a layer of an electrically conductive ink comprising high surface area functionalized graphene sheets and at least one binder.

The printed electronics are prepared by applying the ink to a substrate in a pattern comprising an electrically conductive pathway designed to achieve the desired electronic device. The pathway may be solid, mostly solid, in a liquid or gel form, and the like. The ink may further optionally comprise a carrier other than a binder. When the ink has been applied to the substrate, all or part of the carrier may be removed to form the electrically conductive pathway. The binder may be cured or cross-linked after the ink has been applied to the substrate.

The printed electronic device formed from the application of the ink to the substrate may be optionally sintered or otherwise cured, which can result in the formation of direct bonds between the conducting particles and thus increase the number of conduction paths. In one embodiment of the invention, all or a portion of the device formed from the ink is not sintered and/or not otherwise cured.

To prepare the printed electronic device, the ink may be applied to the substrate using any suitable method, including,
but not limited to, by syringe, spray coating, electrospray deposition, ink-jet printing, spin coating, thermal transfer (including laser transfer) methods, screen printing, rotary screen printing, gravure printing, capillary printing, offset printing, electrohydrodynamic (EHD) printing (a method of which is described in WO 2007/053621, which is hereby incorporated herein by reference), flexographic printing, pad printing, stamping, xerography, microcontact printing, dip pen nanolithography, laser printing, via pen or similar means, and the like.

The substrate may be any suitable material, including, for example, polymers, including thermoplastic and thermoset polymers; pulp products such as paper and cardboard (including coated and uncoated materials); synthetic papers; fabrics (including cloths) and textiles; metals; woods; glass and other minerals; silicon and other semiconductors; ceramics; laminates made from a variety of materials; and the like.

Examples of polymers include polyolefins (such as polyethylene, polypropylene, and the like); polyamides; polystyrenes (such as poly(ethylene terephthalate), poly(ethylene naphthalate)); liquid crystalline polyesters, and the like); polyamides (including polytetrahydroanilides); aramids (such as Kevlar® and Nomex®); fluoropolymers (such as fluorinated ethylene propylene (FEP), polytetrafluoroethylene (PTFE), poly(vinyl fluoride), poly(vinylidene fluoride), and the like); polyetherimides; poly(vinyl chloride); poly(vinylidene chloride); polyurethanes; cellulosic polymers; SAN; ABS; polycarbonates; polycrylates; thermoset epoxies and polyurethanes; and elastomers (including thermoplastics and thermosets, and including rubbers (such as natural rubber) and silicones).

The high surface area functionalized graphene sheets, which are also referred to herein as "FGS", are graphene sheets having a surface area of from about 300 to about 2630 m²/g. In some embodiments of the present invention, the FGS preferably have a surface area of from about 300 to about 2630 m²/g, or more preferably of from about 350 to about 2400 m²/g, or still more preferably of from about 400 to about 2400 m²/g, or yet more preferably of from about 500 to about 2400 m²/g. In another preferred embodiment, the surface area is about 300 to about 1100 m²/g. A single graphite sheet has a maximum calculated surface area of 2630 m²/g. The surface area includes all values and subvalues therebetween, especially including 100, 110, 120, 130, 140, 150, 1600, 1700, 1800, 1900, 2000, 2100, 2200, 2300, 2400, and 2500 m²/g.

Surface area can be measured using either the nitrogen adsorption/BET method or, preferably, a methylene blue (MB) dye method.

The dye method is carried out as follows: A known amount of FGS is added to a flask. At least 1.5 g of MB are then added to the flask per gram of FGS. Ethanol is added to the flask and the mixture is ultrasonicated for about fifteen minutes. The ethanol is then evaporated and a known quantity of water is added to the flask to re-dissolve the free MB. The undissolved material is allowed to settle, preferably by centrifuging the sample. The concentration of MB in solution is determined using a UV-vis spectrophotometer by measuring the absorption at λ_max = 298 nm relative to that of standard concentrations.

The difference between the amount of MB that was initially added and the amount present in solution as determined by UV-vis spectrophotometry is assumed to be the amount of MB that has been adsorbed onto the surface of the FGS. The surface area of the FGS are then calculated using a value of 2.54 m² of surface covered per one mg of MB adsorbed.

The FGS preferably have a bulk density of from about 40 to about 0.1 kg/m³. The bulk density includes all values and subvalues therebetween, especially including 0.5, 1, 5, 10, 15, 20, 25, 30, 35 kg/m³.

The FGS typically have an overall carbon to oxygen molar ratio (C:O ratio), as determined by elemental analysis of at least about 1:1, or more preferably, at least about 3:2. Examples of carbon to oxygen ratios include about 3:2 to about 85:15; about 3:2 to about 20:1; about 3:2 to about 60:1; about 3:2 to about 80:1; about 3:2 to about 100:1; about 3:2 to about 200:1; about 3:2 to about 500:1; about 3:2 to about 1000:1; and about 3:2 to greater than 1000:1, or about 3:2 to about 30:1; and about 3:2 to about 30:1;
poly(vinyl butyral), polyvinyl alcohol and its derivatives, eth-
polymerized before, during, or after the application of the ink
particles, powders, flakes, foils, needles, etc. Preferred solvents include low- or non-VOC solvents, non-
least about 10^-4 S/cm. The conductivities of the inks are deter-
erably have a conductivity of at least about 10^-2 S/cm, or more
devices made from the inks are used as semiconductors, they
conductivity of at least about 10^3 S/cm. In another embodiment of the invention, the inks pref-
S/cm. In one embodiment of the present invention, the surfactant is at least one ethylene oxide/propylene oxide copolymer.

The inks may also optionally comprise one or more pre-
polymers, oligomers, photo-initiators, and additional addi-
tives to allow for curing by UV, electron beam, or infra-red
radiation.

Examples of dispersing aids include glycol ethers (such as
poly(ethylene oxide), block copolymers derived from ethyl-
ene oxide and propylene oxide (such as those sold under the
trade names Surlyn® and Dynol®), salts of carboxylic acids (including alkali metal and
ammonium salts), and polyols.

Examples of adhesion promoters include titanium chelates
and other titanium compounds such as titanium phosphate
complexes (including butyl titanium phosphate), titane
esters, disoproxy titanium bis(ethyl)-3-oxobutanoate, iso-
propoxy titanium acetylacetonate, and others sold by
Johnson-Matthey Catalysts under the trade name Vertec.

Examples of thickening agents include glycol ethers (such as
diethylene glycol, ethylene oxide/propylene oxide copoly-
ers, and wetting aids), emulsifiers, and adhesion promoters,
thickening agents, dispersing aids, surfactants, cross-
linking and curing agents, and the like. In one embodiment of
the present invention, the surfactant is at least one ethylene
oxide/propylene oxide copolymer.

The inks may also optionally comprise one or more pre-
polymers, oligomers, photo-initiators, and additional addi-
tives to allow for curing by UV, electron beam, or infra-red
radiation.
The FGS are preferably present in the ink in at least about 0.01 weight percent based on the total weight of the ink. In one embodiment of the invention, the FGS are preferably present in the ink in at least about 0.05 weight percent, or more preferably in at least about 0.1 weight percent, or still more preferably in at least about 0.5 weight percent, or even more preferably in at least about 1 weight percent, where the weight percentages are based on the total weight of the ink after it has been applied to the substrate and subjected to any post-application treatments (such drying, curing, cross-linking, etc.). However, as will be appreciated by those skilled in the art, the amount of FGS present in the inks can be selected based on the desired electrical conductivity and the particular binders and other optional components chosen.

The inks preferably contain a sufficient amount of FGS such that they have electrical conductivities that are greater than those of the corresponding materials containing each component of the ink in question except for the FGS.

The inks may be made using any suitable method, including wet or dry methods and batch, semi-continuous, and continuous methods.

For example, components of the inks, such as two or more of the functionalized graphene sheets, binders, carriers, and/or other components may be blended by using suitable mixing, dispersing, and/or compounding techniques and apparatus, including ultrasonic devices, high sheen mixers, two-roll mills, three-roll mills, cryogenic grinding crushers, extruders, kneaders, double planetary mixers, triple planetary mixers, high pressure homogenizers, ball mills, attrition equipment, sandmills, and horizontal and vertical wet grinding mills, and the like.

The resulting blends may be further processed using wet or dry grinding technologies. The technologies can be continuous or discontinuous. Examples include ball mills, attrition equipment, sandmills, and horizontal and vertical wet grinding mills. Suitable materials for use as grinding media include metals, carbon steel, stainless steel, ceramics, stabilized ceramic media (such as yttrium stabilized zirconium oxide), PTFE, glass, tungsten carbide, and the like.

After blending and/or grinding steps, additional components may be added to the inks, including, but not limited to, thickeners, viscosity modifiers, and the like. The inks may also be diluted by the addition of more carrier.

After they have been printed on a substrate, the inks may be cured using any suitable technique, including drying and oven-drying (in air or another inert or reactive atmosphere), UV curing, IR curing, microwave curing or drying, and the like.

The printed electronic devices of the present invention may take on a variety of forms. They may contain multiple layers of electronic components (e.g. circuits) and/or substrates. All or part of the printed layer(s) may be covered or coated with another material such as a cover coat, varnish, cover layer, cover films, dielectric coatings, electrolytes and other electrically conductive materials, and the like. There may also be one or more materials between the substrate and printed circuits. Layers may include semiconductors, metal foils, and dielectric materials.

The printed electronics may further comprise additional components, such as processors, memory chips, other microchips, batteries, resistors, diodes, capacitors, transistors, and the like.

The printed electronic devices may take on a wide variety of forms and be used in a large array of applications. Examples include but are not limited to: passive and active devices and components; electrical and electronic circuitry, integrated circuits; flexible printed circuit boards; transistors; field-effect transistors; microelectromechanical systems (MEMS) devices; microwave circuits; antennas; indicators; chipless tags (e.g. for theft deterrence from stores, libraries, and the like); smart cards; sensors; liquid crystalline displays (LCDs); signage; lighting; flat panel displays; flexible displays, including light-emitting diode, organic light-emitting diode, and polymer light-emitting diode displays; backplanes and frontplanes for displays; electroluminescent and OLED lighting; photovoltaic devices—backplanes; product identifying chips and devices; batteries, including thin film batteries; electrodes; indicators; printed circuits in portable electronic devices (for example, cellular telephones, computers, personal digital assistants, global positioning system devices, music players, games, calculators, and the like); electronic connections made through hinges or other movable/bendable junctions in electronic devices such as cellular telephones, portable computers, folding keyboards, and the like; wearable electronics; and circuits in vehicles, medical devices, diagnostic devices, instruments, and the like.

Preferred electronic devices are radiofrequency identification (RFID) devices and/or components thereof and/or radiofrequency communication device. Examples include, but are not limited to, RFID tags, chips, and antennas. The RFID devices may be ultrahigh frequency RFID devices, which typically operate at frequencies in the range of about 868 to about 928 MHz. Examples of uses for RFID devices are for tracking shipping containers, products in stores, products in transit, and parts used in manufacturing processes; passports; barcode replacement applications; inventory control applications; pet identification; livestock control; contactless smart cards; automobile key fobs; and the like.

**EXAMPLES**

**General Details for Examples 1-3 and Comparative Example 1**


The inks are prepared as follows. A poly(ethylene oxide) (PEO) solution is prepared by mixing a sufficient amount of PEO with a 1:1 volume/volume mixture of ethanol and deionized water to produce a mixture containing 40 mg of PEO per ml of total solvent. After stirring overnight, a homogeneous PEO stock solution is obtained.

FGS is weighed and a sufficient amount of concentrated aqueous Pluronic® F127 (an ethylene oxide/propylene oxide copolymer surfactant supplied by BASF) solution (typically 2 mg/mL) is added to the FGS to yield a mixture having a 1:1 weight ratio of FGS and Pluronic F127. Sufficient de-ionized water is added to produce a suspension containing 1 mg FGS per 1 ml of water. The resulting suspension is sonicated for 5 minutes with a duty cycle of 20 percent in an ice bath. 1 mL of the FGS suspension is then added to 3 mL of the PEO stock solution and the mixture is stirred for 3-5 minutes until homogeneous.

**Example 1**

An ink is prepared as described above using PEO having a molecular weight of 4,000,000. The ink is printed electrohy-
potential difference measured on the film and current is used to find the resistance using Ohm’s law, i.e. \( R = \frac{V}{I} \), where \( R \), \( V \), and \( I \) are the resistance, voltage, and current, respectively. Resistivity (\( \sigma \)) is found by \( \sigma = \frac{RA}{L} \), where \( A \) and \( L \) are the cross section of the film through which current flows and the length over which the potential difference is measured. Conductivity (\( \kappa \)) is found by \( \kappa = \frac{1}{\sigma} \). The resulting conductivity is about 0.05 S/m.

**Example 2**

An ink is prepared as described above using PEO having a molecular weight of 4,000,000. The ink is printed electrohydrodynamically onto a glass substrate using the method disclosed in WO 2007/053621. The printing is done at a flow rate of 0.5 mL/hr under a 2200 kV potential difference between electrodes that are 7.2 mm apart. The width of the lines is about 130 \( \mu \)m. The ink is formed into a film as follows: two copper plates (22 mm x 22 mm) are wrapped with Teflon tape, leaving 1 mm of copper uncovered at the lower ends. The plates are then firmly attached to the shorter of the side walls of a Teflon® cell (23 mm x 46 mm inner base area, 32 mm height) with screws. The mixture is poured into the Teflon® cell and kept at 50°C on a hot plate until all of the solvent is evaporated to form films that are attached to the copper plates.

Copper tapes are attached parallel to each other on two ends of the film such that they cover the entire width of the film. The electrical conductivity of the film is measured across the copper tapes as described above for Example 1. The measured conductivity is about 12.4 S/m.

**Comparative Example 1**

An ink is prepared as described above substituting carbon black (supplied by Cabot Corp.) for the FGS and using PEO having a molecular weight of 300,000. The ink is printed electrohydrodynamically onto a glass substrate using the method disclosed in WO 2007/053621. The printing is done at a flow rate of 0.5 mL/hr under a 2200 kV potential difference between electrodes that are 7.2 mm apart. The width of the lines is about 130 \( \mu \)m.

As described above for Example 2, the ink is formed into a film and its electrical conductivity is measured. The result is about 1.5 x 10^-8 S/m.

**Example 3**

An ink is prepared as described above using PEO having a molecular weight of 4,000,000. The ink is printed electrohydrodynamically onto a glass substrate using the method disclosed in WO 2007/053621. The printing is done at a flow rate of 0.5 mL/hr under a 2200 kV potential difference between electrodes that are 7.2 mm apart. The width of the lines is about 140 \( \mu \)m and the thickness is about 300 \( \mu \)m. The printed line has an electrical conductivity of about 19.6 S/m.

**General Details for Examples 4-14**

**Preparation of Test Samples**

The inks in the form of liquid dispersions are printed onto a substrate using a doctor blade and then dried in air in an oven at 125°C to form a film. Testing is done on the printed films.

**Electrical Conductivity**

The point-to-point resistivity (in ohms) of the films is measured using a standard multimeter across contact points consisting of two spots of silver paste having a diameter of about 0.3 mm that are applied to the surface of the film about 1 inch apart. The resistance across these points is also measured using a standard multimeter and the reading is divided by 10 to calculate the resistivity in ohms/square. Results given as a single number are an average of several measurements and results given as a range of figures indicate the high and low readings from several measurements.

**Peel Resistance**

A fingernail is drawn back and forth across the surface of the film five times. The surface of the film where it was scratched and the tip of the nail are examined and the scratch resistance of the film is assessed as follows: excellent is no noticeable transfer of the film surface to the nail; very good is minimal transfer and no noticeable indentation on the surface of the film; good is some indentation of the film surface; fair is removal of a substantial portion of the film; and poor is where the substrate is visible. In some cases no cohesive film adhered to the substrate is formed.

**Scratch Resistance**

A fingernail is drawn across the surface of the film. The film and top of the nail are visually inspected. The scratch resistance is graded as follows: no transfer of film material to the fingernail—excellent; about 10 percent transfer—very good; about 20 percent transfer—good/very good; about 30 percent transfer—good; about 40 percent transfer—fair/good; about 50 percent transfer—fair; about 65 percent transfer—poor/fair; about 85 percent transfer—poor; and about 100 percent transfer (the substrate is fully visible and very little to no printed material remains)—very poor.

**Ink Preparation Methods**

Ball mill A: An Eiger Mini 250 Type M250 VSE-TETF horizontal grinding mill

Ball mill B: A vertical stainless steel vertical grinding mill having four stainless steel arms situated 90° away from each other. The mill is driven by a compressed air motor and has a bottom discharge valve.

High shear mixer: A homogenizer having a rotor-stator overhead stirrer.

**Ingredients Used in the Formulations:**

Electron and Positron are citrus terpene-based solvents supplied by Ecolink, Tucker Ga.

**Example 4**

A 4.9 weight percent aqueous solution of poly(ethylene oxide) (PEO) having an average molecular weight of 600,000 (236.2 g) is combined with FGS having a C:O ratio of approximately 100:1 (2.4 g), ethylene oxide/propylene oxide copolymer surfactant (Pluronic F127, supplied by BASF) (2.4 g), antifoaming agent (AF 204, supplied by Sigma) (0.3 g), and water (50 g). The mixture is ground in ball mill B at...
650 rpm using \( \frac{3}{16} \)" stainless steel balls as a grinding medium for 6 hours. The resulting dispersion is printed onto thermally stabilized PET, coated paper, and uncoated paper and the adhesion properties and electrically resistivity of the resulting printed films are measured. The results are given in Table 1.

Example 5

A 10.8 weight percent aqueous solution of poly(ethylene oxide) (PEO) having an average molecular weight of 600,000 (110.8 g) is combined with FGS having a C:O ratio of approximately 100:1 (2.4 g), surfactant (Surfynol 104H, supplied by Sigma) (0.2 g), and water (134.2 g). The mixture is ground in ball mill B at 693 rpm using 3/6 stainless steel balls for 1.5 hours. The resulting dispersion is printed onto thermally stabilized PET, coated paper, and uncoated paper and the adhesion properties and electrically resistivity of the resulting printed films are measured. The results are given in Table 1.

### TABLE 1

<table>
<thead>
<tr>
<th>Additive</th>
<th>Peel resistance (Q/sq.)</th>
<th>Resistivity (Q/sq.)</th>
<th>Peel resistance (Q/sq.)</th>
<th>Resistivity (Q/sq.)</th>
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<tr>
<td>Example 5</td>
<td>Good</td>
<td>5-10</td>
<td>Poor</td>
<td>6-7</td>
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</table>

Examples 6-12

In the case of Examples 6-10, a 20 weight percent solution of polyamide binder (Versamid 750, supplied by Cognis) in isopropyl alcohol (200 g) is combined with FGS having a C:O ratio of approximately 100:1 (10 g) and additional isopropyl alcohol (40 g). In the case of Examples 11 and 12, a 10 weight percent solution of polyaniline (PANI) (Panipol F, supplied by Panipol Oy, Porvoo, Finland) in chloroform (2 g) is added to 10 g of the resulting dispersion. After each of these additives is added, the resulting mixture is blended for about a minute in the high shear mixer. In each case the resulting dispersion is printed onto thermally stabilized PET and the adhesion properties and electrically resistivity of the printed films are measured. The results are given in Table 2.

### TABLE 2

<table>
<thead>
<tr>
<th>Additive</th>
<th>Peel resistance</th>
<th>Scratch resistance</th>
<th>Resistivity (Q/sq.)</th>
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<tr>
<td>Example 6</td>
<td>none</td>
<td>Excellent</td>
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<tr>
<td>Example 7</td>
<td>BYK</td>
<td>Very good</td>
<td>Very good</td>
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<tr>
<td>Example 8</td>
<td>none</td>
<td>Excellent</td>
<td>Excellent</td>
</tr>
<tr>
<td>Example 9</td>
<td>PANI</td>
<td>Excellent</td>
<td>Excellent</td>
</tr>
<tr>
<td>Example 10</td>
<td>BYK</td>
<td>Excellent</td>
<td>Excellent</td>
</tr>
<tr>
<td>Example 11</td>
<td>none</td>
<td>Excellent</td>
<td>Excellent</td>
</tr>
<tr>
<td>Example 12</td>
<td>PANI</td>
<td>Good</td>
<td>Good</td>
</tr>
</tbody>
</table>

The invention claimed is:

1. A printed electronic device, comprising a substrate comprising at least one surface, wherein a layer of an electrically conductive ink has been applied to a portion of the surface, and wherein the ink comprises functionalized graphene sheets and at least one binder.

2. The device of claim 1, wherein the substrate comprises paper and/or cardboard.

3. The device of claim 1, wherein the substrate comprises at least one polyolefin.

4. The device of claim 1, wherein the substrate comprises at least one polyimide.

5. The device of claim 1, wherein the ink further comprises at least one dispersant.

6. The device of claim 1, wherein the binder is a polymeric binder.

7. The device of claim 6, wherein the binder is one or more of poly(ethylene oxide), poly(propylene oxide), and ethylene oxide/propylene oxide copolymers.

8. The device of claim 1, wherein the ink further comprises at least one metal component.

9. The device of claim 8, wherein the metal is silver and/or copper.

10. The device of claim 1, wherein the ink further comprises at least one electrically conductive polymer.

11. The device of claim 1, wherein the ink further comprises at least one carbonaceous material other than the functionalized graphene sheets.

12. The device of claim 1, wherein the functionalized graphene sheets have a surface area of from about 300 to about 2630 m\(^2\)/g.

13. The device of claim 1, wherein the functionalized graphene sheets have a surface area of from about 300 to about 2630 m\(^2\)/g.

14. The device of claim 1, wherein the functionalized graphene sheets have a carbon to oxygen ratio of at least one polyimide.

15. The device of claim 1, wherein the functionalized graphene sheets have a carbon to oxygen ratio of at least one polyimide.

16. The device of claim 1, wherein the functionalized graphene sheets have a carbon to oxygen ratio of at least one polyimide.

17. The device of claim 1, wherein the functionalized graphene sheets have a carbon to oxygen ratio of at least one polyimide.

18. The device of claim 1, wherein the functionalized graphene sheets have a carbon to oxygen ratio of at least one polyimide.

19. The device of claim 1, further comprising one or more components selected from the group consisting of processors, memory chips, batteries, resistors, diodes, capacitors, and transistors.

20. The device of claim 1, further comprising one or more components selected from the group consisting of processors, memory chips, batteries, resistors, diodes, capacitors, and transistors.

21. The device of claim 1, in the form of a radiofrequency identification device and/or a radiofrequency device antenna.

22. The radiofrequency identification device and/or a radiofrequency device antenna of claim 19 in the form of a radiofrequency identification device and/or antenna for use with ultra-high frequencies.
21. A method for forming a printed electronic device, comprising the step of applying an electrically conductive ink to a substrate, wherein the ink comprises functionalized graphene sheets and at least one binder.

22. The method of claim 21, wherein the substrate is selected from one or more of paper, cardboard, polyolefin, and polyimide.

23. The method of claim 21, wherein the electrically conductive ink further comprises a carrier.

24. The method of claim 21, wherein the electrically conductive ink further comprises a dispersant.

25. The method of claim 21, wherein the functionalized graphene sheets have a surface area of from about 300 to about 2630 m²/g.