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(54) Title: A METHOD FOR PREPARING A GRAPHENE-BASED CONDUCTIVE FILM

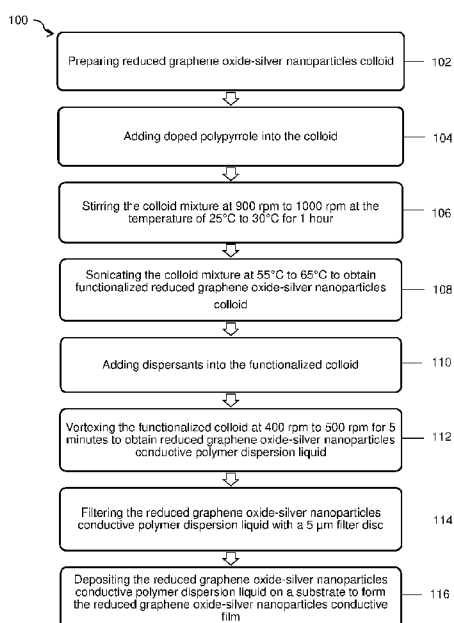


Figure 1

(57) Abstract: The present invention discloses a method (100) for preparing a graphene-based conductive film characterized by the steps of: adding conductive polymer into graphene-based nanoparticles colloid to obtain a functionalized colloid (104); adding dispersants into said functionalized colloid (110); vortexing said functionalized colloid to obtain a graphene-based conductive polymer dispersion liquid (112); and depositing said graphene-based conductive polymer dispersion liquid on a substrate to form the graphene-based conductive film (116).

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A METHOD FOR PREPARING A GRAPHENE-BASED CONDUCTIVE FILM

FIELD OF THE INVENTION

The present invention relates to preparation of graphene-based
5 conductive film for flexible electronics, more particularly a method for preparing
a reduced graphene oxide-silver nanoparticles conductive film through inkjet
printing.

BACKGROUND OF THE INVENTION

10 Flexible electronics is a technology that allows building of electronic
circuits on flexible substrates thus making them foldable and stretchable. The
main advantages of flexible electronics are the low cost manufacturing through
inkjet printing and inexpensive flexible substrate such as plastics, paper,
textiles or metal foils. Therefore, flexible electronic has gained great attention
15 due to its ability in providing cost effective solutions to wide area of applications
such as rollable displays, electronic paper, flexible cellular phone, solar cells,
RFID-tags, wearable sensors for health monitoring, lab-on-a-chip for disease
diagnostics, flexible temperature sensors for healthcare and et cetera.

20 As the demand for wearable and foldable electronic devices increases
rapidly, the conductive film as an important component in the electronic devices
is greatly needed. Accordingly, graphene with superior electrical and thermal
conductivities has been widely used as a based material in production of
conductive film. The production process of graphene-based material such as
25 chemical reduction is usually unable to remove all the oxygen in graphene
oxide thereby creating defects and vacancies to produce graphene-based
material with lower quality as compared to pristine graphene.

As a result, the graphene-based conductive film produced through inkjet
30 printing is potentially causing several issues. For instance, the film with lower
hardness can cause film rupture and thus generate adhesion issue.
Consequently, the methods to functionalize graphene-based material such as
addition of conductive polymers into graphene-based material have been
developed to reinforce the film hardness. Nevertheless, the existing methods

also degrade the electrical performance of the film and lead to clogging issue in inkjet printing while increasing the film hardness. Accordingly, an improved method with simpler steps is required in order to effectively address the aforementioned issues.

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US20160244577 A1 discloses a method for manufacturing a graphene polymer conductive film. The method comprising the steps of providing a graphene powder and a plurality of conductive monomers; providing a solvent and producing a graphene dispersion liquid by stirring and ultrasonic processing after mixing the graphene powder into the solvent; producing a mixed liquid having the graphene and the conductive monomers by stirring and ultrasonic processing after mixing the conductive monomers into the graphene dispersion liquid; mixing an initiator into the mixed liquid of the graphene and the conductive monomers for in situ polymerization to produce a pre-liquid of a graphene conductive polymer composite material; producing a powder of the graphene conductive polymer composite material by removing solvent and impurity in the pre-liquid of the graphene conductive polymer composite material through a filtration and drying process; providing, mixing and stirring an epoxy resin, a curing agent and an accelerator until uniformly distributed to produce epoxy resin glue system; distributing the powder of the graphene conductive polymer composite material into the epoxy resin glue system to produce a pre-material of the graphene polymer conductive film; and deaerating the pre-material of the graphene polymer conductive film to produce the graphene polymer conductive film.

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CN103839608A discloses a method for preparing graphene conductive film. The method comprising the steps of adding graphite or graphite derivative to the dispersant/ethanol/dimethyl formamide organic solvent to obtain dispersion liquid; subjecting the dispersion liquid to high density ultrasonication to obtain a monomolecular layer or a multilayer graphene solution, and the upper layer liquid is taken after high-speed centrifugation, and the graphene powder is obtained after flocculation treatment; dispersing the graphene powder into an organic solvent to prepare an inkjet ink; injecting the inkjet ink into the ink cartridge and deposited onto the pre-processed substrate in a

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predetermined pattern by conventional inkjet printing technology; drying the conductive film deposited on the substrate in an oven to obtain graphene conductive film.

5 Jiantong Li *et al.*, Efficient Inkjet Printing of Graphene, Advanced Materials, Volume 25, 29 (2013), 1-8 discloses a method for preparing graphene conductive film using inkjet printing. The graphene is first exfoliated from graphite flakes in dimethylformamide which is DMF. After that, the low viscosity and toxic DMF is changed to high-viscosity and environmental-friendly
10 terpineol through distillation to concentrating the graphene. Prior to printing, the graphene/terpineol dispersion is mixed with ethanol to optimize the viscosity and surface tension for inkjet printing. The transparent conductive films are fabricated by printing the graphene inks onto untreated glass slides. After printing, the stabilizing polymers can be effectively removed through a simple
15 annealing process.

The aforesaid methods for preparing graphene-based conductive films are complicated as several additional steps such as polymerization process and drying process are required. Indirectly, said additional steps increase the
20 production cost. Additionally, the aforesaid methods involve the use of toxic chemicals that may cause harm to human and environment. It is apparent that the aforesaid prior arts are lacking a simple, cost effective and harmless method for preparing graphene-based conductive film with improved hardness without degrading its electrical performance.

25

Accordingly, there is a need to provide a simpler, cost effective and harmless method for preparing a graphene-based conductive film which can improve its hardness without degrading its electrical performance.

30 **SUMMARY OF THE INVENTION**

It is an objective of the present invention to provide a simpler and cost effective method for preparing graphene-based conductive film.

It is also an objective of the present invention to provide a method for preparing graphene-based conductive film with improved hardness without degrading its electrical performance.

5 It is further an objective of the present invention to provide harmless method for preparing graphene-based conductive film by eliminating the use of toxic chemicals.

Accordingly, these objectives may be achieved by following the
10 teachings of the present invention. The present invention provides a method for preparing a graphene-based conductive film.

The present invention discloses a method for preparing a graphene-based conductive film characterized by the steps of: adding conductive polymer
15 into graphene-based nanoparticles colloid to obtain a functionalized colloid; adding dispersants into said functionalized colloid; vortexing said functionalized colloid to obtain a graphene-based conductive polymer dispersion liquid; and depositing said graphene-based conductive polymer dispersion liquid on a substrate to form the graphene-based conductive film.

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BRIEF DESCRIPTION OF THE ACCOMPANYING DRAWINGS

The manner in which the above recited features of the present invention can be understood in detail, a more particular description of the invention, briefly summarized above, may have been referred by embodiments, some of
25 which are illustrated in the appended drawings. It is to be noted, however, that the appended drawing illustrates only typical embodiments of this invention and are therefore not to be considered limiting of its scope, for the invention may admit to other equally effective embodiments.

30 The features, benefits, and advantages of the present invention will become apparent by reference to the following text figure, with like reference numbers referring to like structures across the views, wherein:

Figure 1 shows a flow chart of a method (100) for preparing a graphene-based conductive film in accordance to an embodiment of the present invention.

DETAILED DESCRIPTION OF THE INVENTION

5 While the present invention is described herein by way of example using embodiments and illustrative drawings, those skilled in the art will recognize that the invention is not limited to the embodiments of drawing or drawings described, and are not intended to represent the scale of the various components. Further, some components that may form a part of the invention
10 may not be illustrated in certain figures, for ease of illustration, and such omissions do not limit the embodiments outlined in any way. It should be understood that the drawings and detailed description thereto are not intended to limit the invention to the particular form disclosed, but on the contrary, the invention is to cover all modifications, equivalents, and alternatives falling within
15 the scope of the present invention as defined by the appended claim. As used throughout this description, the word "may" is used in a permissive sense (i.e. meaning having the potential to), rather than the mandatory sense, (i.e. meaning must). Further, the words "a" or "an" mean "at least one" and the word "plurality" means "one or more" unless otherwise mentioned. Furthermore, the
20 terminology and phraseology used herein is solely used for descriptive purposes and should not be construed as limiting in scope. Language such as "including," "comprising," "having," "containing," or "involving," and variations thereof, is intended to be broad and encompass the subject matter listed thereafter, equivalents, and additional subject matter not recited, and is not
25 intended to exclude other additives, components, integers or steps. Likewise, the term "comprising" is considered synonymous with the terms "including" or "containing" for applicable legal purposes. Any discussion of documents, acts, materials, devices, articles and the like is included in the specification solely for the purpose of providing a context for the present invention. It is not suggested
30 or represented that any or all of these matters form part of the prior art base or were common general knowledge in the field relevant to the present invention.

In this disclosure, whenever a composition or an element or a group of elements is preceded with the transitional phrase "comprising", it is understood

that we also contemplate the same composition, element or group of elements with transitional phrases “consisting of”, “consisting”, “selected from the group of consisting of”, “including”, or “is” preceding the recitation of the composition, element or group of elements and vice versa.

5

The present invention is described hereinafter by various embodiments with reference to the accompanying drawing, wherein reference numerals used in the accompanying drawing correspond to the like elements throughout the description. This invention may, however, be embodied in many different forms and should not be construed as limited to the embodiment set forth herein. Rather, the embodiment is provided so that this disclosure will be thorough and complete and will fully convey the scope of the invention to those skilled in the art. In the following detailed description, numeric values and ranges are provided for various aspects of the implementations described. These values and ranges are to be treated as examples only, and are not intended to limit the scope of the claims. In addition, a number of materials are identified as suitable for various facets of the implementations. These materials are to be treated as exemplary, and are not intended to limit the scope of the invention.

Referring to the drawing, the invention will now be described in more detail.

A method (100) for preparing a graphene-based conductive film of the present invention encompasses adding of a conductive polymer to functionalize a graphene-based nanoparticles colloid to improve hardness without degrading the electrical performance of the film.

In accordance with an embodiment of the present invention, the method (100) for preparing graphene-based conductive film comprises the steps of adding conductive polymer into graphene-based nanoparticles colloid to obtain a functionalized colloid (104); adding dispersants into said functionalized colloid (110); vortexing said functionalized colloid to obtain a graphene-based conductive polymer dispersion liquid (112); and depositing said graphene-

based conductive polymer dispersion liquid on a substrate to form the graphene-based conductive film (116).

5 In a preferred embodiment of the present invention, the graphene-based nanoparticles colloid is reduced graphene oxide-silver nanoparticles colloid in a range of 0.08 g/mL to 0.12 g/mL.

10 In accordance with an embodiment of the present invention, the conductive polymer is selected from the group consisting of polypyrrole, polyaniline and its derivatives, polyindole, more preferably a doped polypyrrole in a range of 0.02% w/v to 0.15% w/v. Doped polypyrrole is a cost efficient conducting polymer with high thermal and chemical stabilities as well as high solubility in water.

15 In accordance with an embodiment of the present invention, the step of adding conductive polymer (104) further comprises the steps of stirring the colloid mixture at 900 rpm to 1000 rpm at the temperature of 25°C to 30°C for 1 hour (106) after adding conductive polymer; and sonicating the colloid mixture at 55°C to 65°C for 1 hour to obtain said functionalized colloid (108).

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In accordance with an embodiment of the present invention, the dispersants are selected from the group consisting of ethylene glycol, terpineol, methanol, ethanol, 2-propanol, cyclohexanone or combination thereof, more preferably a combination of ethylene glycol, terpineol and methanol. Ethylene glycol is a polar solvent that interacts with oxygen functional group on the graphene-based surface to prevent functionalized colloid from aggregation and thus produce a uniform dispersion. Terpineol is a natural oil for enhancing dispersion of functionalized colloid and thus helps in obtaining of a smooth printing ink. Methanol is highly soluble in water and possesses higher evaporation rate than ethanol. As a result, said combination of dispersants enhance the efficiency of in-situ drying of the printed film at the later stage.

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In a preferred embodiment of the present invention, the functionalized colloid is dispersed in ethylene glycol, terpineol and methanol with a volume/volume ratio of 1: 0.03: 0.005:0.05.

5 In another embodiment of the present invention, the functionalized colloid is dispersed in ethylene glycol and methanol with a volume/volume ratio of 1: 0.06: 0.1. The combination of said dispersants is employed when the circuit design has the minimum line width of 100 μm .

10 In accordance with an embodiment of the present invention, the step of vortexing (112) is carried out at 400 rpm to 500 rpm for 5 minutes.

In accordance with an embodiment of the present invention, the graphene-conductive polymer dispersion liquid is filtered with a 5 μm filter disc
15 (114) before depositing on the substrate; wherein the substrate is a flexible substrate.

In a preferred embodiment of the present invention, the graphene-based conductive film is a reduced graphene oxide-silver nanoparticles conductive
20 film.

Hereinafter, example of the present invention will be provided for more detailed explanation. The advantages of the present invention may be more readily understood and put into practical effect from these examples. However,
25 it is also to be understood that the following examples are not to limit the scope of the present invention in any way.

EXAMPLE

The overall method (100) for preparing the reduced graphene oxide-silver nanoparticles conductive film in accordance to an embodiment of the
30 present invention is illustrated in Figure 1. The detailed of each of the steps of the method (100) were described as follows:

1. Preparation of reduced graphene oxide-silver nanoparticles colloid

Deionized water was added into reduced graphene oxide-silver nanoparticles composite to prepare a mixture of 8.0% w/v to 12.0% w/v. The mixture was stirred at 5000 to 6000 rpm for 15 minutes at 25°C to 30°C to obtain reduced graphene oxide-silver nanoparticles colloid (102).

2. Preparation of functionalized reduced graphene oxide-silver nanoparticles colloid

0.02% w/v to 0.15% w/v of doped polypyrrole was added into 0.08 g/mL to 0.12 g/mL of reduced graphene oxide-silver nanoparticles colloid (104). The colloid mixture was stirred at 1000 rpm at the temperature of 25°C to 30°C for 1 hour (106). Said colloid mixture was then sonicated at 60°C for 1 hour to obtain functionalized reduced graphene oxide-silver nanoparticles colloid (108).

3. Preparation of reduced graphene oxide-silver nanoparticles conductive polymer dispersion liquid

The functionalized reduced graphene oxide-silver nanoparticles colloid was dispersed to dispersants mixture comprising ethylene glycol, terpineol and methanol with a volume/volume ratio of 1: 0.03: 0.005:0.05 (110). The addition of dispersants helps to ensure the inkjet printing at the later stage run smoothly without clogging. The functionalized reduced graphene oxide-silver nanoparticles colloid was vortexed at 400 rpm to 500 rpm for 5 minutes to obtain reduced graphene oxide-silver nanoparticles conductive polymer dispersion liquid (112) which is a functionalized reduced graphene oxide-silver nanoparticles ink. The functionalized reduced graphene oxide-silver nanoparticles ink was filtered with a 5 µm filter disc (114).

4. Preparation of reduced graphene oxide-silver nanoparticles conductive film

The functionalized reduced graphene oxide-silver nanoparticles ink was injected into an empty cartridge. A detachable nozzle head was attached

to the filled cartridge and loaded onto the cartridge stage of the printer. The functionalized reduced graphene oxide-silver nanoparticles ink was then printed on the flexible substrate through inkjet printing to form reduced graphene oxide-silver nanoparticles conductive film (116) with improved hardness.

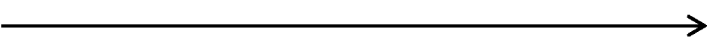
Experiment Result

The hardness of reduced graphene oxide-silver nanoparticles conductive film produced using the method (100) of the present invention was evaluated using ASTM D3363 pencil hardness test. Further, the electrical test was carried out to evaluate the resistance, resistivity and conductivity of the film. Table 1 shows the comparative results of the pencil hardness test and electrical test between reduced graphene oxide-silver nanoparticles conductive film of the present invention and the other graphene-based conductive film of the prior art.

Table 1: Comparison on the hardness and electrical test data of graphene-based conductive films

Parameter	Present invention	Prior art
Hardness	At least H and up to 2H	HB
Resistance, Ω	1.20×10^{-1}	1.98×10^{-1}
Sheet resistance, Ω/sq	4.69×10^{-1}	7.69×10^{-1}
Resistivity, $\Omega.\text{cm}$	4.69×10^{-5}	7.69×10^{-5}
Conductivity, S/cm	3.19×10^4	1.27×10^4

ASTM D3363 pencil hardness test standard:

Pencil hardness scale	6B	5B	4B	3B	2B	B	HB	F	H	2H	3H	4H	5H	6H
	Softest													Hardest

The result in table 1 shows the reduced graphene oxide-silver nanoparticles conductive film of the present invention exhibits hardness up to 2H as compared to the graphene-based conductive film of the prior art with hardness only up to HB. In comparison to the prior art, reduced graphene oxide-silver nanoparticles conductive film of the present invention also shows

better electrical performance in terms of resistance, resistivity and conductivity. This result indicates the simpler method (100) of the present invention is workable to produce reduced graphene-based nanoparticles conductive film with improved hardness without degrading its electrical performance.

5

Various modifications to these embodiments are apparent to those skilled in the art from the description and the accompanying drawings. The principles associated with the various embodiments described herein may be applied to other embodiments. Therefore, the description is not intended to be limited to the embodiments shown along with the accompanying drawings but is to be providing broadest scope of consistent with the principles and the novel and inventive features disclosed or suggested herein. Accordingly, the invention is anticipated to hold on to all other such alternatives, modifications, and variations that fall within the scope of the present invention and appended claim.

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CLAIMS

1. A method (100) for preparing a graphene-based conductive film characterized by the steps of:
 - 5 adding conductive polymer into graphene-based nanoparticles colloid to obtain a functionalized colloid (104);
 adding dispersants into said functionalized colloid (110);
 vortexing said functionalized colloid to obtain a graphene-based conductive polymer dispersion liquid (112); and
 - 10 depositing said graphene-based conductive polymer dispersion liquid on a substrate to form the graphene-based conductive film (116).
2. The method (100) as claimed in claim 1, wherein the graphene-based nanoparticles colloid is reduced graphene oxide-silver nanoparticles
15 colloid in a range of 0.08 g/mL to 0.12 g/mL.
3. The method (100) as claimed in claim 1, wherein the conductive polymer comprising doped polypyrrole in a range of 0.02% w/v to 0.15% w/v.
- 20 4. The method (100) as claimed in claim 1, wherein the step of adding conductive polymer (104) further comprising the steps of:
 - stirring the colloid mixture at 900 rpm to 1000 rpm at the temperature of 25°C to 30°C for 1 hour after adding conductive polymer (106); and
 - 25 sonicating the colloid mixture at 55°C to 65°C for 1 hour to obtain said functionalized colloid (108).
5. The method (100) as claimed in claim 1, wherein the dispersants is selected from the group consisting of ethylene glycol, terpineol and
30 methanol or combination thereof.
6. The method (100) as claimed in claim 1, wherein the volume/volume ratio of functionalized colloid to ethylene glycol, terpineol and methanol is 1: 0.03: 0.005:0.05.

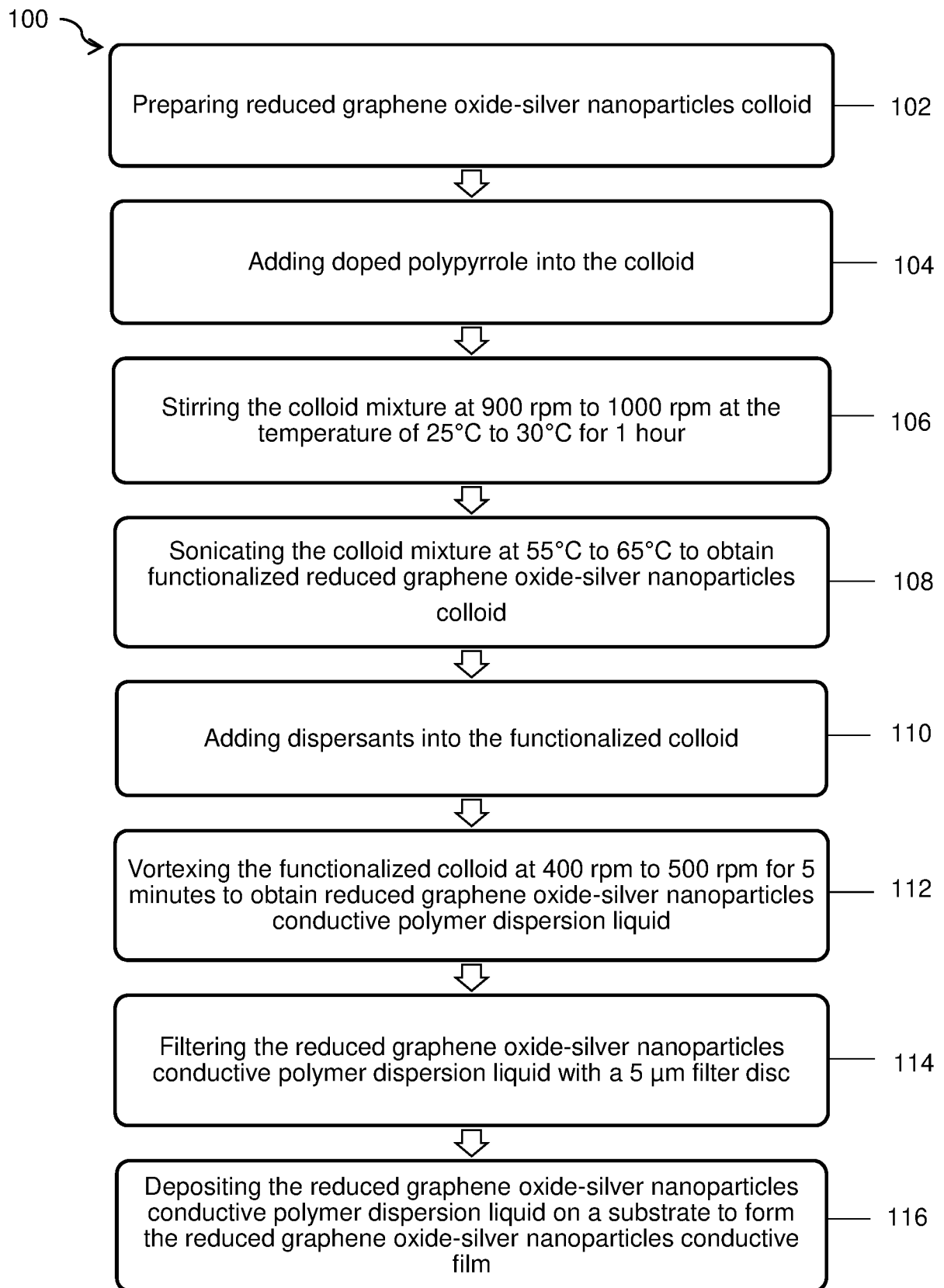
7. The method (100) as claimed in claim 1, wherein the volume/volume ratio of functionalized colloid to ethylene glycol and methanol is 1: 0.06: 0.1.
- 5 8. The method (100) as claimed in claim 1, wherein the step of vortexing (112) is carried out at 400 rpm to 500 rpm for 5 minutes.
9. The method (100) as claimed in claim 1, wherein the graphene-based conductive polymer dispersion liquid is filtered with a 5 μ m filter disc
10 (114) before depositing on the substrate.
10. The method (100) as claimed in claim 1, wherein the graphene-based conductive film is a reduced graphene oxide-silver nanoparticles conductive film.

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**Figure 1**

INTERNATIONAL SEARCH REPORT

International application No.

PCT/MY2020/050198

A. CLASSIFICATION OF SUBJECT MATTER C08J 5/18(2006.01)i; C08K 3/04(2006.01)i; C08K 3/08(2006.01)i; C08L 101/00(2006.01)i; C08L 65/00(2006.01)i; H01B 1/20(2006.01)i; H01B 13/00(2006.01)i; C01B 32/182(2017.01)i; C01B 32/05(2017.01)i 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) C08J 5/18(2006.01); C09D 11/037(2014.01); C09D 11/52(2014.01); C09D 183/02(2006.01); C09D 5/24(2006.01); H01B 1/24(2006.01); H01G 11/34(2013.01) Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Korean utility models and applications for utility models Japanese utility models and applications for utility models Electronic data base consulted during the international search (name of data base and, where practicable, search terms used) eKOMPASS(KIPO internal) & keywords: graphene-based conductive film, silver, polypyrrole, reduced graphene oxide																				
C. DOCUMENTS CONSIDERED TO BE RELEVANT <table border="1"> <thead> <tr> <th>Category*</th> <th>Citation of document, with indication, where appropriate, of the relevant passages</th> <th>Relevant to claim No.</th> </tr> </thead> <tbody> <tr> <td>X</td> <td>US 2020-0002560 A1 (LAI, C.-P.) 02 January 2020 (2020-01-02) claims 8, 10-11, 15; paragraphs [0025]-[0026]</td> <td>1,3-9</td> </tr> <tr> <td>Y</td> <td></td> <td>2,10</td> </tr> <tr> <td>Y</td> <td>KR 10-2017-0109970 A (INDUSTRY ACADEMY COOPERATION FOUNDATION OF SEJONG UNIVERSITY) 10 October 2017 (2017-10-10) claims 1-4</td> <td>2,10</td> </tr> <tr> <td>A</td> <td>ZHAO, C. et al., "Reduced graphene oxide and polypyrrole/reduced graphene oxide composite coated stretchable fabric electrodes for supercapacitor application", Electrochimica Acta, 2015, Vol. 172, pages 1 2-19 the whole document</td> <td>1-10</td> </tr> <tr> <td>A</td> <td>US 2016-0240278 A1 (SHENZHEN CHINA STAR OPTOELECTRONICS TECHNOLOGY CO., LTD.) 18 August 2016 (2016-08-18) the whole document</td> <td>1-10</td> </tr> </tbody> </table>			Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.	X	US 2020-0002560 A1 (LAI, C.-P.) 02 January 2020 (2020-01-02) claims 8, 10-11, 15; paragraphs [0025]-[0026]	1,3-9	Y		2,10	Y	KR 10-2017-0109970 A (INDUSTRY ACADEMY COOPERATION FOUNDATION OF SEJONG UNIVERSITY) 10 October 2017 (2017-10-10) claims 1-4	2,10	A	ZHAO, C. et al., "Reduced graphene oxide and polypyrrole/reduced graphene oxide composite coated stretchable fabric electrodes for supercapacitor application", Electrochimica Acta, 2015, Vol. 172, pages 1 2-19 the whole document	1-10	A	US 2016-0240278 A1 (SHENZHEN CHINA STAR OPTOELECTRONICS TECHNOLOGY CO., LTD.) 18 August 2016 (2016-08-18) the whole document	1-10
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INTERNATIONAL SEARCH REPORT

International application No.

PCT/MY2020/050198

C. DOCUMENTS CONSIDERED TO BE RELEVANT

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Information on patent family members

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