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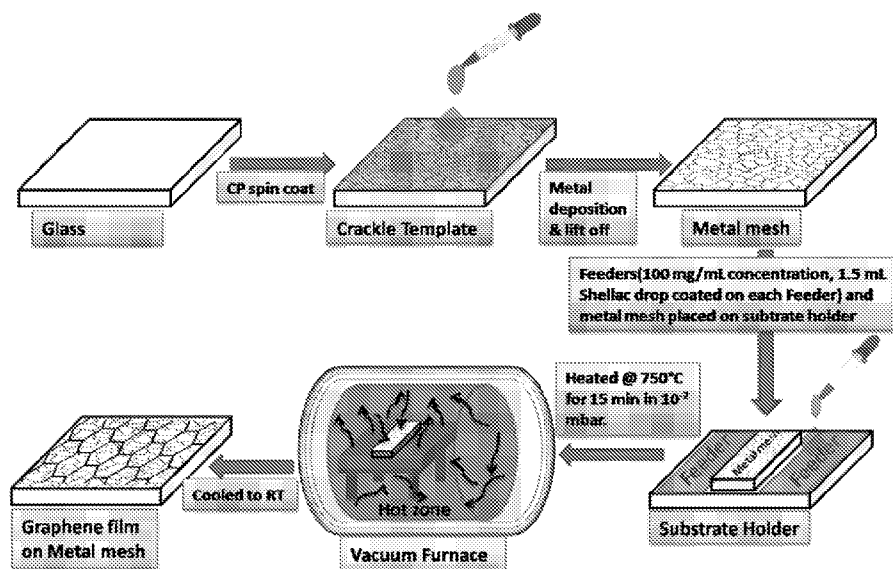


Figure 1

(57) Abstract: The present invention relates to hybrid transparent conducting electrode comprising reduced graphene oxide film, metal mesh and textured glass, wherein the reduced graphene oxide film is coated on the textured glass embedded with the metal mesh or the reduced graphene oxide film is sandwiched between the textured glass and the metal mesh. The present invention also relates to a process of preparing the hybrid conducting transparent conducting electrode. The said transparent conducting electrode exhibits transparency ranging from about 70% to 85% with sheet resistance ranging from about 5Ω/sq to 100Ω/sq.



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## “HYBRID TRANSPARENT CONDUCTING ELECTRODE”

### TECHNICAL FIELD

The present invention is in relation to electrodes. The present invention particularly relates  
5 to hybrid transparent conducting electrode. The hybrid transparent conducting electrode  
comprises reduced graphene oxide film, metal mesh and textured glass. The present  
invention further relates to a process of preparing the said hybrid transparent conducting  
electrode.

### 10 BACKGROUND OF THE INVENTION

Transparent conducting electrode is the cynosure of the optical devices industry. The astute  
need of the transparent conducting electrodes is expressively acknowledged across the optical  
device industry involving solar panels, smartphone, computer, televisions, windshields and the  
like, owing to the consistent effort to develop better devices.

15 Indium tin oxide (ITO) is the highly preferred material to obtain transparent conducting  
electrodes, however this has challenges due to cost and brittle nature of ITO. Many  
alternatives are being developed with metal wire meshes, graphene reduced graphene  
coatings and carbon nano tubes. Each technology poses challenge to be addressed for  
20 optimum benefit. The direct growth of graphene on non-catalytic substrates like quartz and  
glass involves the conventional CVD process for graphene growth at higher temperature  
about 900° C to 1050° C and need longer time duration for graphene growth from about  
2hour to 6 hours. The said process also uses combustible gases like methane, methanol as  
precursor. Further, CVD system requires catalytic nano particles such as Nickel and Copper  
25 as seed layer for graphene growth and uses argon as carrier gas and H<sub>2</sub> for promoting  
graphene growth.

Various substrates are being explored to coat and obtain conducting transparent materials  
and amongst them glass occupies premium position with its unique properties.

30

However, the adoption of metal wire mesh, graphene, reduced graphene and the like on textured glass is a challenge as the available technologies fail to get the desired product at the level of texturing of glass embedded with metal wire mesh, and reduced graphene oxide or vice versa.

5

#### **SUMMARY OF THE INVENTION**

Accordingly, the present invention describes a hybrid transparent conducting electrode comprising reduced graphene oxide film, metal mesh and textured glass.

10 The present invention also relates to a process of preparing the hybrid transparent conducting electrode, comprising- texturing the glass or the glass embedded with metal mesh; and coating the textured glass embedded with the metal mesh with the reduced graphene oxide film or coating the textured glass with the reduced graphene oxide film followed by coating the metal mesh, to obtain the hybrid transparent conducting electrode.

15

#### **BRIEF DESCRIPTION OF THE ACCOMPANYING FIGURES:**

In order that the present invention may be readily understood and put into practical effect, reference will now be made to exemplary embodiments as illustrated with reference to the accompanying figures. The figures together with detailed description below, are  
20 incorporated in and form part of the specification, and serve to further illustrate the embodiments and explain various principles and advantages, in accordance with the present invention where:

**FIGURE 1** illustrates experimental step up for production of the hybrid transparent  
25 conducting electrode.

**FIGURE 2** illustrates the transparency of the ordinary glass slide coated with graphene.

**FIGURE 3a:** illustrates the transparency of the hybrid transparent conducting electrode in  
30 the photograph.

**FIGURE 3b** illustrates the corrosion rates of the glass embedded with copper mesh (Cu-Mesh) and the hybrid transparent electrode of the present invention (rGO/Cu-Mesh).

**FIGURE 4a and 4b** illustrates transmittance plot in specular and diffused mode of reduced graphene oxide coated glass (rGO/glass) and the hybrid transparent electrode of the present invention (rGO/Cu-mesh/glass), respectively.

5 **FIGURE 4c** illustrates the Haze plot of reduced graphene oxide coated glass (rGO/glass) and the hybrid transparent electrode of the present invention (rGO/Cu-mesh/glass).

**FIGURE 5** illustrates the digital images of contact angle of glass, reduced graphene oxide coated glass (rGO/glass) and the hybrid transparent electrode of the present invention  
10 (rGO/Cu-mesh/glass).

**FIGURE 6a** illustrates the resistance of reduced graphene oxide coated glass (rGO/glass)

**FIGURE 6b** illustrates the resistance of the hybrid transparent electrode of the present invention (rGO/Cu-mesh/glass).

15 **FIGURE 6c** illustrates relative change in the resistance of reduced graphene oxide coated glass (rGO/glass) and the hybrid transparent electrode (rGO/Cu-mesh/glass) of the present invention

**FIGURE 7a** illustrates a plot describing change in resistance with number of scotch tape  
20 peeling of reduced graphene oxide coated glass (rGO/glass)

**FIGURE 7b** illustrates a plot describing pencil hardness level of the hybrid transparent electrode (rGO/Cu-mesh/glass) of the present invention.

**FIGURE 8** illustrates Raman spectra recorded on metal mesh and in void regions of the  
25 hybrid transparent electrode of the present invention (rGO/Cu-mesh/glass).

**FIGURE 9a** illustrates the IR image of the graphene coated glass slide.

**FIGURE 9b** illustrates the heating cycles of graphene coated glass slide.

30 **FIGURE 10a** illustrates the IR image of hybrid transparent conducting electrode.

**FIGURE 10b** illustrates the digital image of hybrid transparent conducting electrode.

**FIGURE 11a-11c** illustrates the height profile of texture on glass substrate obtained after graphene oxide growth.

**FIGURE 11d** illustrates digital images of textured glass and their respective stainless steel substrates.

5

#### **DETAILED DESCRIPTION OF THE INVENTION**

The foregoing description of the embodiments of the invention has been presented for the purpose of illustration. It should not be construed that the scope of the invention is limited to the disclosure herein.

10

The various embodiments of the hybrid transparent conducting electrode of the present invention along with its process of preparation/fabrication are described below with reference to the figures.

15

It may further be noted that as used herein and in the appended claims, the singular forms "a", "an", and "the" include plural reference unless the context clearly dictates otherwise. Unless defined otherwise, all technical and scientific terms used herein have the same meanings as commonly understood by person skilled in the art.

20

The present invention relates to hybrid transparent conducting electrode.

In an embodiment, the present invention relates to hybrid transparent conducting electrode comprising reduced graphene oxide film, metal mesh and textured glass.

25

In an embodiment, the hybrid transparent conducting electrode comprises reduced graphene oxide film, metal mesh and textured glass, wherein the reduced graphene film is coated on the textured glass embedded with the metal mesh.

30

In another embodiment, the hybrid transparent conducting electrode comprises reduced graphene oxide film, metal mesh and textured glass, wherein the reduced graphene oxide film is sandwiched between the textured glass and metal mesh.

In an embodiment, carbon source of the reduced graphene oxide film is shellac.

In an embodiment, the reduced graphene oxide film in the hybrid transparent conducting electrode is a monolayer.

5

In another embodiment, the reduced graphene oxide film in the hybrid transparent conducting electrode comprises plurality of layers.

In an embodiment, the metal mesh comprises metal selected from a group comprising copper, silver, aluminum, gold, tin and nickel.

10

In another embodiment, the metal mesh comprises alloy selected from a group comprising aluminium-zinc alloy, aluminium-silica alloy, copper-nickel alloy, bronze, nickel-chromium alloy, steel and mild steel.

15

In an embodiment, the hybrid transparent conducting electrode has transparency ranging from about 70% to 85%.

In another embodiment, the hybrid transparent conducting electrode has transparency of about 70%, about 72%, about 74%, about 76%, about 78%, about 80%, about 82%, about 84% or about 85%.

20

In an embodiment, the hybrid transparent conducting electrode has sheet resistance ranging from about 5 $\Omega$ /sq to 100 $\Omega$ /sq.

25

In another embodiment, the hybrid transparent conducting electrode has sheet resistance of about 5 $\Omega$ /sq, about 10 $\Omega$ /sq, about 15 $\Omega$ /sq, about 20 $\Omega$ /sq, about 25 $\Omega$ /sq, about 30 $\Omega$ /sq, about 35 $\Omega$ /sq, about 40 $\Omega$ /sq, about 45 $\Omega$ /sq, about 50 $\Omega$ /sq, about 55 $\Omega$ /sq, about 60 $\Omega$ /sq, about 65 $\Omega$ /sq, about 70 $\Omega$ /sq, about 75 $\Omega$ /sq, about 80 $\Omega$ /sq, about 85 $\Omega$ /sq, about 90 $\Omega$ /sq, about 95 $\Omega$ /sq or about 100 $\Omega$ /sq.

30

In an embodiment, the hybrid transparent conducting electrode demonstrates reduced resistance. The reduced resistance of the hybrid transparent conducting electrode is due to

the combined effect of metal mesh and the reduced graphene oxide film on the textured glass substrate.

5 In an embodiment, the hybrid transparent conducting electrode has corrosion rate is ranging from about 0.09mm/year to 1.0 mm/year.

In another embodiment, the hybrid transparent conducting electrode has corrosion rate of about 0.09mm/year, about 0.1mm/year, about 0.2mm/year, about 0.3mm/year, about 0.4mm/year, about 0.5mm/year, about 0.6mm/year, about 0.7mm/year, about 0.8mm/year,  
10 about 0.9mm/year/ or about 1.0mm/year.

In an embodiment, the conductivity of the hybrid transparent conducting electrode is due to the coating of reduced graphene oxide film on the textured glass embedded with the metal mesh.

15 In another embodiment, the conductivity of the hybrid transparent conducting electrode is due to the reduced graphene oxide film sandwiched between the textured glass and the metal mesh.

20 In another embodiment, the conductivity of the hybrid transparent conducting electrode is due to metal mesh and reduced graphene oxide embedded on the textured glass.

In an embodiment, the hybrid transparent conducting electrode is stable and can withstand extreme conditions such as high humidity and high temperatures.

25 The said hybrid transparent electrode was stable at relative humidity ranging from about 0% to 85% and temperature ranging from about 25°C to 85°C.

30 The said hybrid transparent electrode was stable at relative humidity of about 85% and at a temperature of about 85°C for about 18hours.

The present invention further relates to process of preparing the hybrid transparent conducting electrode.

In an embodiment, the process of preparing the hybrid transparent conducting electrode  
5 comprises-

texturing the glass or the glass embedded with metal mesh; and

coating the textured glass embedded with the metal mesh with the reduced graphene oxide film or coating the textured glass with the reduced graphene oxide film followed by coating the metal mesh,

10 to obtain the hybrid transparent conducting electrode.

In an embodiment, the process of preparing the hybrid transparent conducting electrode comprises-

15 texturing the glass embedded with metal mesh; and

coating the textured glass embedded with metal mesh with reduced graphene oxide film to obtain the hybrid transparent conducting electrode.

In an embodiment, the glass embedded with metal mesh is obtained by coating colloidal  
20 solution over the glass by technique including but not limited to drop casting and spin coating, followed by air drying the glass at temperature ranging from about 25°C to 40°C for about 60 seconds, which results in formation of crackle template on the glass.

The crackle template on the glass is deposited with the metal selected from a group  
25 comprising copper, silver, aluminium, gold, tin and nickel or the alloy selected from a group comprising aluminium-zinc alloy, aluminium-silica alloy, copper-nickel alloy, bronze, nickel-chromium alloy, steel and mild steel, by vacuum deposition methods to obtain metal wire mesh on glass or glass embedded with metal mesh.

30 In an embodiment, the vacuum deposition method is physical vapor deposition, chemical vapor deposition, low pressure chemical vapor deposition, plasma enhanced chemical vapor deposition, or any combinations thereof.

In an embodiment, glass having the crackle template is subjected to physical vapor deposition, wherein about 50mg to 1000mg of the metal or the alloy is embedded on the glass. The physical vapor deposition chamber is closed to attain pressure of about  $10^{-5}$  mbar to  $10^{-6}$  in about 80 minutes to 90 minutes and the chamber temperature is maintained at about 25°C to 30°C. The metal is evaporated once the chamber crosses  $10^{-5}$  mbar pressure. Once, the metal or the alloy is deposited on the glass, lift-off is performed by dipping the glass in chloroform solution for a period of about 1minute to 2minutes to remove the colloidal material.

10 In another embodiment, about 50mg, about 100mg, about 150mg, about 200mg, about 250mg, about 300mg, about 350mg, about 400mg, about 450mg, about 500mg, about 550mg, about 600mg, about 650mg, about 700mg, about 750mg, about 800mg, about 850mg, about 900mg or about 1000mg of the metal or the alloy is embedded on the glass.

15 In an embodiment, texturing the glass embedded with the metal mesh and coating the reduced graphene oxide film on the textured glass embedded with the metal mesh is carried out simultaneously.

20 In an embodiment, texturing the glass embedded with the metal mesh and coating the reduced graphene oxide comprises heating the glass embedded with metal mesh placed on a textured stainless steel and at least one feeder comprising shellac. The heating is carried out at a temperature ranging from about 400° C to 1200° C under vacuum at a pressure ranging from about  $10^{-1}$  to  $10^{-3}$  mbar for a period of about 1 minute to 15 minutes. The said heating  
25 causes the shellac to vaporize and contact the textured glass embedded with the metal mesh, thereby forming coat of the reduced graphene oxide film on the textured glass embedded with metal mesh upon cooling to a temperature ranging from about 25°C to 50°C.

In another embodiment, the heating is carried out at a temperature of about 400°C, about 30 500°C, about 600°C, about 700°C, about 800°C, about 900°C, about 1000°C, about 1100°C or about 1200°C under vacuum at a pressure of about  $10^{-1}$  mbar, about  $10^{-3}$  mbar or about  $10^{-3}$  mbar, for a period of about 1minute, about 2minutes, about 3minutes, about 4minutes, about

5minutes, about 6minutes, about 7minutes, about 8minutes, about 9minutes, about 10minutes, about 11minutes, about 12minutes, about 13minutes, about 14minutes or about 15mintues.

- 5 In an embodiment, reduced graphene oxide coat is formed on the texture glass upon cooling to a temperature of about 25°C, about 30°C, about 35°C, about 40°C, about 45°C or about 50°C.

In an embodiment, the feeder comprises shellac at a concentration ranging from about  
10 10gram/liter to 200gram/liter. The shellac is casted on the feeder by technique including but not limited to spin coating, dipping, spraying, bar coating, slot coating and drop casting.

In another embodiment, the feeder comprises shellac at a concentration of about 10gram/liter, about 20gram/liter, about 40gram/liter, about 60gram/liter, about 80gram/liter, about  
15 100gram/liter, about 120gram/liter, about 140gram/liter, about 150gram/liter, about 160gram/liter, about 180gram/liter or about 200gram/liter.

In an embodiment, the process of preparing the hybrid transparent conducting electrode comprises-

texturing the glass and coating the textured glass with reduced graphene oxide film;  
20 and  
coating the metal mesh on the reduced graphene oxide film of the textured glass to obtain the hybrid transparent conducting electrode.

In another embodiment, the process of preparing the hybrid transparent conducting electrode  
25 comprises-

texturing the glass and coating the glass with reduced graphene oxide film;  
coating colloidal solution on top of textured reduced graphene oxide film;  
drying the glass to obtain crackle template; and  
coating metal on the crackle template, followed by washing the template to obtain  
30 the hybrid transparent conducting electrode.

In an embodiment, texturing the glass and coating the reduced graphene oxide comprises heating the glass placed on a textured stainless steel and at least one feeder comprising shellac. The heating is carried out at a temperature ranging from about 400° C to 1200° C under vacuum at a pressure ranging from about  $10^{-1}$  mbar to  $10^{-3}$  mbar for a period ranging  
5 from about 1 minute to 15 minutes. The said heating causes the shellac to vaporize and contact the textured glass, thereby forming coat of the reduced graphene oxide on the textured glass upon cooling to a temperature ranging from about 25°C to 50°C.

In another embodiment, the heating is carried out at a temperature of about 400°C, about 500°C, about 600°C, about 700°C, about 800°C, about 900°C, about 1000°C, about 1100°C  
10 or about 1200°C under vacuum at a pressure of about  $10^{-1}$ mbar, about  $10^{-3}$ mbar or about  $10^{-3}$ mbar, for a period of about 1minute, about 2minutes, about 3minutes, about 4minutes, about 5minutes, about 6minutes, about 7minutes, about 8minutes, about 9minutes, about 10minutes, about 11minutes, about 12minutes, about 13minutes, about 14minutes or about 15mintues.

15 In an embodiment, reduced graphene oxide coat is formed on the texture glass upon cooling to a temperature of about 25°C, about 30°C, about 35°C, about 40°C, about 45°C or about 50°C.

20 In an embodiment, the feeder comprises shellac at a concentration ranging from about 10gram/liter to 200gram/liter. The shellac is casted on the feeder by technique including but not limited to spin coating, dipping, spraying, bar coating, slot coating and drop casting.

In another embodiment, the feeder comprises shellac at a concentration of about 10gram/liter,  
25 about 20gram/liter, about 40gram/liter, about 60gram/liter, about 80gram/liter, about 100gram/liter, about 120gram/liter, about 140gram/liter, about 150gram/liter, about 160gram/liter, about 180gram/liter or about 200gram/liter.

In an embodiment, coating the metal mesh on the reduced graphene oxide film coated  
30 textured glass comprises depositing the metal selected from a group comprising copper, silver, aluminium, gold, tin and nickel or the alloy selected from a group comprising

aluminium-zinc alloy, aluminium-silica alloy, copper-nickel alloy, bronze, nickel-chromium alloy, steel and mild steel, by vacuum deposition methods.

5 In an embodiment, in the above described processes of coating reduced graphene oxide film on the textured glass embedded with metal mesh or on the textured glass, the reduced graphene oxide coating is controlled in a specific region by introducing a physical mask which contains hollow region which allows the coating of the reduced graphene oxide in that region.

10 In an embodiment, the above described processes of preparing hybrid transparent conducting electrode is simple and cost effective.

In the process of the present invention ecofriendly precursor is employed to grow reduced graphene oxide at lower temperatures ranging 750° C to 850° C with vacuum ranging of 10<sup>-3</sup> mbar to 10<sup>-2</sup> mbar and at lower time period ranging from about 1 minutes to 15 minutes. 15 Importantly no metal catalyst is employed in the process of preparing the hybrid transparent conducting electrode.

Conventionally, most glasses soften under the graphene synthesis condition (at 650° C), In 20 order to control the glass flow and deformation of glass in the process of the present invention, metal plate is introduced with desired texture below the glass which uses the glass softening property and control the glass flow which gives raise to textured glass, such that glass deformation is controlled and contained to the texturing available on the metal plate. This way the glass is elegant but can be made to host graphene.

25

In an embodiment, the figure 1 illustrates the process of preparing the hybrid transparent conducting electrode schematically, wherein the glass is embedded with metal wire mesh, placed in a substrate holder on top of a textured stainless-steel plate, coating the feeder with the source of carbon (shellac) and heated in a furnace/chamber under vacuum to obtain 30 hybrid transparent conducting electrode.

In an embodiment, the figure 2 depicts the photographs of graphene coated glass slide, illustrating the transparency and resistance. The graphene coated glass show resistance of about 200k $\Omega$ .

5 In an embodiment, the figure 3 depicts the photographs of the hybrid transparent conducting electrode, illustrating the transparency and resistance. The hybrid transparent conducting electrode shows resistance of about 30 $\Omega$  with high transparency when compared to the glass slide depicted in figure 2.

10 Also, the reduced graphene oxide coating in the hybrid transparent conducting electrode makes the metal mesh (Cu mesh) corrosion tolerant and it is about 13 times better than the metal mesh (Cu-mesh) alone.

In an embodiment, the figure 4a and 4b illustrates the transmittance plot in specular and  
15 diffused mode, respectively, demonstrating that the hybrid transparent conducting electrode is found to be conducting all over the region of the reduced graphene oxide film with sheet resistance of about 5 $\Omega$ /sq to 100  $\Omega$ /sq. And, having the optical transmission/transmittance of about 70% to 85% in the visible region.

20 The figure 4c depicts the haze plot, wherein the haze calculation is carried out to check the influence of texturing in the hybrid transparent conducting electrode. It is observed that the haze is about 5% which is minimal even after texturing the glass.

In an embodiment, the figure 5 illustrates the digital images of contact angle measured on  
25 glass, reduced graphene oxide coated glass (rGO/glass) and the hybrid transparent electrode of the present invention (rGO/Cu-mesh/glass). The contact angle measurement is carried out to show the hydrophobicity. It is noted that the coating of reduced graphene oxide film on the textured glass embedded with metal mesh makes the electrode more hydrophobic when compared to bare glass.

30

In an embodiment, the figure 8 illustrates Raman spectra confirming the presence of reduced graphene oxide film on the metal mesh in the hybrid transparent conducting electrode.

In an embodiment, the figure 11a to 11c illustrates the height profile of glass textures. Texturing of the glass is carried out by using stainless steel substrate illustrated in figure 11d.

5 As per the height profile described in figure 11a, the minimum feature size obtained is 25 $\mu$ m. As per the disclosure of the present invention, the feature size that can be textured varies in the range of about 25 $\mu$ m to 1000 $\mu$ m.

10 Additional embodiments and features of the present invention will be apparent to one of ordinary skill in art based upon the description provided. The embodiments provide various features and advantageous details thereof in the description. Description of well-known/conventional methods and techniques are omitted so as to not unnecessarily obscure the embodiments. The examples provided herein are intended merely to facilitate an understanding of ways in which the embodiments provided may be practiced and to further  
15 enable those of skill in the art to practice the embodiments provided. Accordingly, the following examples should not be construed as limiting the scope of the embodiments.

## **EXAMPLES**

### **EXAMPLE 1: Preparation of the hybrid transparent conducting electrode**

#### 20 **A. Preparation of glass embedded with metal wire mesh/metal mesh.**

Colloidal solution is made by adding about 0.4g of resinous particle in solvent (water or diluter) of volume of about 1ml at a temperature ranging from about 25°C to 40°C and sonicating the solution for about 10 minutes. About 0.25ml of the colloidal solution is coated over the glass surface of about 2" x 2" by drop casting or spin coating, followed by  
25 air drying the glass at temperature of about 25 °C to 40°C for about 60seconds which results in crack formation on the glass (crackle template).

The crackle template is placed inside Physical vapor deposition (PVD) chamber and about 400mg of copper is deposited. The chamber is closed to attain pressure of about 10<sup>-5</sup> to 10<sup>-6</sup>  
30 mbar in about 80minutes to 90minutes and the chamber temperature is maintained at about 25°C to 30°C. The metal on the glass is evaporated once the chamber crosses the pressure

of about  $10^{-5}$  mbar and there is deposition of the metal on the crackle template. After, the metal deposition is complete, the crackle template is taken out and lift-off is performed by dipping the crackle template in chloroform solution for about 1 minute to 2 minutes to remove the colloidal material. After lift-off the glass is left with metal wire mesh on the surface.

The above described preparation can be employed for depositing other metal selected from a group comprising silver, aluminium, gold, tin, nickel or the alloy selected from a group comprising aluminium-zinc alloy, aluminium-silica alloy, copper-nickel alloy, bronze, nickel-chromium alloy, steel and mild steel, to obtain glass having the metal mesh on the surface of the glass or glass embedded with metal mesh.

#### **B. Preparation of carbon source for coating reduced graphene oxide film**

A solution of the about 100mg/L shellac precursor is made by adding about 10 g of shellac in isopropanol solvent of volume of about 100 ml at a temperature ranging from about 25°C to 90°C by gentle mixing using magnetic stirrer for about 15 minutes. Said solution is kept as it is in stationary state for about 2 hours to 3 hours for settling down the contamination. About 1.5ml of the shellac solution is coated over the quartz (feeder) surface of about 2" x 2" by drop casting.

#### **C. Preparation of the Hybrid transparent conducting electrode**

The stainless-steel plate with desired texture (illustrated in Figure 11d) is placed below the glass embedded with metal mesh and is placed in a chamber alongside the feeder as described in Figure 1.

Figure 11a to 11c illustrates the height profile of glass textures. The chamber is heated using an electro-heating furnace to a temperature of about 750°C for about 15 minutes under vacuum at pressure of about  $10^{-2}$  mbar. During heating the vapours of the shellac is deposited on the metal mesh of the glass, which forms a coat of reduced graphene oxide film on the metal mesh embedded on the glass while cooling to temperature of about 25°C to 40°C. As a result, the hybrid transparent conducting electrode is obtained.

**EXAMPLE 2:**

A. Corrosion resistance property of hybrid transparent conducting electrode was tested by electrochemical investigations using CH instruments in three electrode configurations.

In the 3-electrode configuration, copper mesh (Cu-mesh) or the hybrid transparent conducting electrode (rGO/Cu-mesh/glass) are employed as working electrodes, Platinum counter electrode and standard Ag/Ag/Cl as reference electrode.

The area of the working electrode is about 0.98 cm<sup>2</sup> and about 1.28 cm<sup>2</sup> for Cu-mesh and rGO/Cu-mesh/glass, respectively in about 0.1M NaCl electrolyte at a temperature of about 25°C to 40°C. The cathodic and anodic polarization measurements are performed at a scan rate of 5mV/s. The corrosion potential (E<sub>corr</sub>) and corrosion current (I<sub>corr</sub>) are derived through extrapolation from Tafel polarization curve. The corrosion rate is calculated using the equation given below-

$$CR = (I_{corr} \times K \times EW) / (p \times A)$$

Where, K, corrosion rate constant = 3272mm/year; EW, equivalent weight = 31.7g for Cu; p, material density = 8.94 g/cm<sup>3</sup> for Cu and the sample area A in cm<sup>2</sup> and I<sub>corr</sub> is current density in A/cm<sup>2</sup>.

The corrosion resistance of rGO to underlying Cu-mesh is studied using electrochemical measurements (see figure 3b). Using Tafel analysis, the Tafel polarization curve for rGO/Cu-mesh/glass shows a positive shift of about 181 mV of corrosion potential and a lower corrosion current of about 15mV compared to bare Cu-mesh. The corrosion current (I<sub>corr</sub>) values are found to be 3.896 × 10<sup>-6</sup> Acm<sup>-2</sup> and 1.369 × 10<sup>-5</sup> Acm<sup>-2</sup> for rGO/Cu-mesh/glass and Cu-mesh, respectively. Accordingly, the corrosion rates are estimated to be 0.0911 mm/year and 2.340 mm/year, respectively. It is significant that the corrosion rate is about 13 times lower for rGO/Cu-mesh compared to that of bare Cu-mesh.

The corrosion test of the hybrid transparent conducting electrode are carried out against highly acidic and basic chemicals, wherein the hybrid transparent conducting electrode was directly dipped in highly concentrated acidic and basic chemicals.

5

The hybrid transparent conducting electrode shows very good resistance to degradation in terms of electrical properties. The results are shown in table 1.

Chemical	Aqueous concentraion	Resistance	
		Before (~kΩ)	After 1hour (~kΩ)
H <sub>2</sub> SO <sub>4</sub>	95-98%	50	53
HNO <sub>3</sub>	67%	28	30
HCl	37%	44	48
H <sub>2</sub> O <sub>2</sub>	30%	52	54
NaOH	1M	40	41
KOH	1M	42	43

10 Table 1: Resistance change of hybrid transparent conducting electrode with respect to different acid and base treatments.

B. Environment chamber test was conducted to test the stability of the hybrid transparent conducting electrode.

15

The Environmental chamber test was conducted at about 85% relative humidity and at temperature of about 85°C. Figure 6a shows change in resistance of rGO/glass and rGO/cu-mesh/glass. Figure 6b shows change in the substrate (glass) with time, and figure 6 (c) is the plot of relative change in resistance of both the samples with respect to time.

20

The said Environmental chamber test demonstrates that the reduced graphene oxide film coating in the hybrid transparent conducting electrode is stable and can withstand extreme humid and high temperatures.

25

C. Pencil hardness test was performed to check the adhesion of the reduced graphene oxide film coating over the glass embedded metal mesh of the hybrid transparent conducting electrode (rGO/Cu-mesh/glass) and on rGO/Cu-mesh substrate.

5 The said test was performed to check the adhesion level of the material to the substrate where one among the different level pencil is selected and it is slid on to the other substrate to measure the change in resistance.

10 The hybrid transparent conducting electrode of the present invention performed well to 6H hardness level and showed minimal variation in resistance even after 100 cycles of 6H scratch test when compared to the rGO/Cu-mesh substrate.

The figure 7 shows the plot of change in resistance with number of scotch tape peeling (figure 7a) and with different pencil hardness level (figure 7b).

15 D. The area and shape of reduced graphene coating in the hybrid transparent conducting electrode is determined by the physical mask brought on top and its resolution (about few microns to 15 cm).

20 The hybrid transparent conducting electrode is noted have reduced graphene coat of about 10mm x 10mm area successfully with a sheet resistance of about  $10\Omega/\text{sq}$  and the hybrid transparent conducting electrode can be joule heated to a temperature of about  $60^{\circ}\text{C}$  by applying about 3V.

25 The figure 9 illustrates that heating rates can be controlled by input power, heating zone can be made uniform and the heating cycles are reproducible while coating reduced graphene oxide film on the glass slide.

The IR image and digital image of the hybrid transparent conducting electrode is shown in figure 10.

**WE CLAIM:**

1. A Hybrid transparent conducting electrode comprising-  
reduced graphene oxide film;  
5 metal mesh; and  
textured glass  
wherein the reduced graphene oxide film is coated on the textured glass embedded with the metal mesh, or the reduced graphene oxide film is sandwiched between the textured glass and the metal mesh.
- 10 2. The hybrid transparent conducting electrode as claimed in claim 1, has transparency ranging from about 70% to 85%.
3. The hybrid transparent conducting electrode as claimed in claim 1, has sheet resistance  
15 ranging from about  $5\Omega/\text{sq}$  to  $100\Omega/\text{sq}$ .
4. The hybrid transparent conducting electrode as claimed in claim 1, wherein carbon source for the reduced graphene oxide film is shellac.
- 20 5. The hybrid transparent conducting electrode as claimed in claim 1, wherein the reduced graphene oxide film is a monolayer or comprises plurality of layers.
6. The hybrid transparent conducting electrode as claimed in claim 1, wherein the metal mesh comprises metal or alloy, wherein the metal is selected from a group comprising  
25 copper, silver, aluminium, gold, tin and nickel and the alloy is selected from a group comprising aluminium-zinc alloy, aluminium-silica alloy, copper-nickel alloy, bronze, nickel-chromium alloy, steel and mild steel.
7. The hybrid transparent conducting electrode as claimed in claim 1, has corrosion rate of  
30 about 0.0911 mm/year.
8. A process of preparing the hybrid transparent conducting electrode as claimed in claim 1, comprising-

texturing the glass embedded with metal mesh or the glass;

and

coating the textured glass embedded with the metal mesh with the reduced  
graphene oxide film or coating the textured glass with the reduced graphene

5 oxide film followed by coating the metal mesh,

to obtain the hybrid transparent conducting electrode.

9. The process as claimed in claim 8, wherein the texturing the glass embedded with the  
metal mesh or the glass and coating the textured glass embedded with the metal mesh or  
10 the textured glass, with reduced graphene oxide film is carried out simultaneously.
10. The process as claimed in claim 8, wherein the texturing the glass embedded with the  
metal mesh or the glass and coating the reduced graphene oxide film comprises heating-
- 15 a. the glass embedded with the metal mesh or the glass, placed on a textured stainless  
steel; and
- b. at least one feeder comprising shellac.
11. The process as claimed in claim 10, wherein the heating is carried out at a temperature  
ranging from about 400° C to 1200° C under vacuum at a pressure ranging from about  
20  $10^{-1}$  mbar to  $10^{-3}$  mbar for a period ranging from about 1 minute to 15 minutes.
12. The process as claimed in claim 10, wherein the feeder comprises shellac at a  
concentration ranging from about 10gram/litre to 200gram/litre; wherein the shellac is  
casted on the feeder by technique selected from a group comprising spin coating,  
25 dipping, spraying, bar coating, slot coating and drop casting.
13. The process as claimed in claim 10, wherein the heating causes shellac on the feeder to  
vaporize and contact the textured glass embedded with metal mesh or the textured  
glass, thereby forming coat of the reduced graphene oxide on the textured glass  
30 embedded with metal mesh or the textured glass upon cooling to a temperature ranging  
from about 25°C to 50°C.

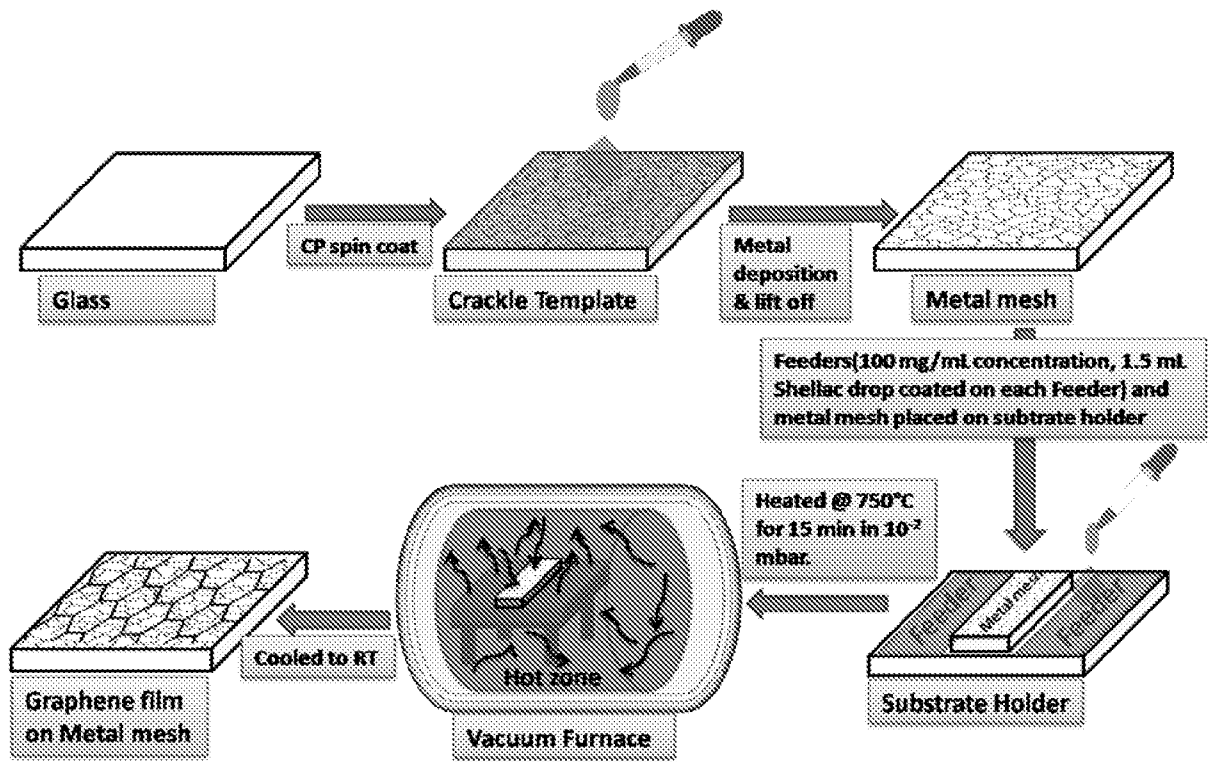


Figure 1

5

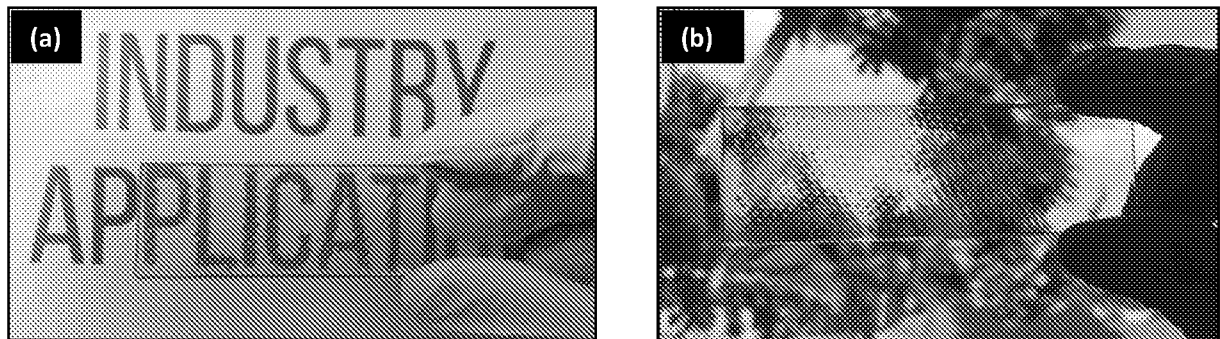


Figure 2

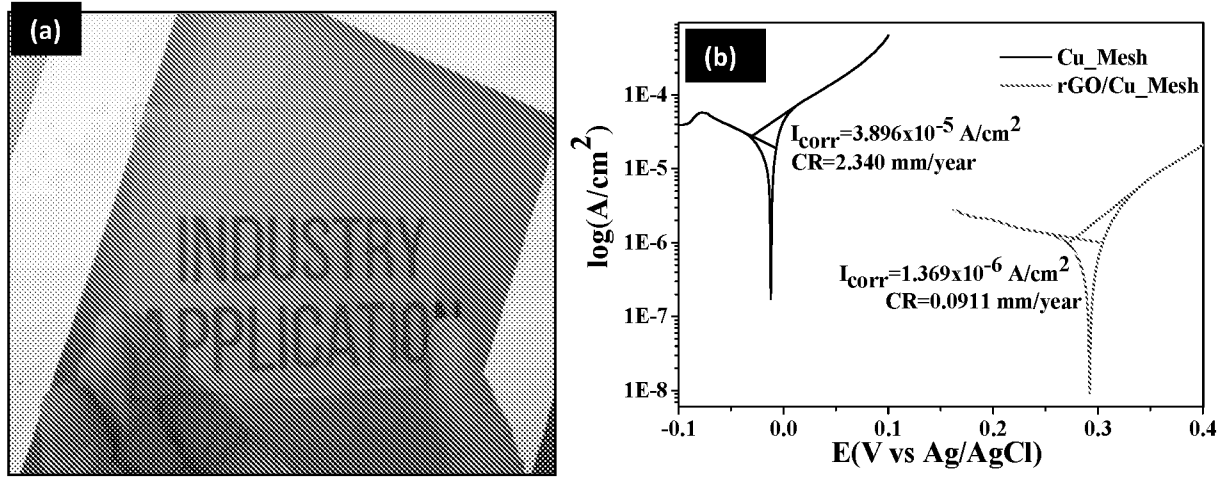


Figure 3

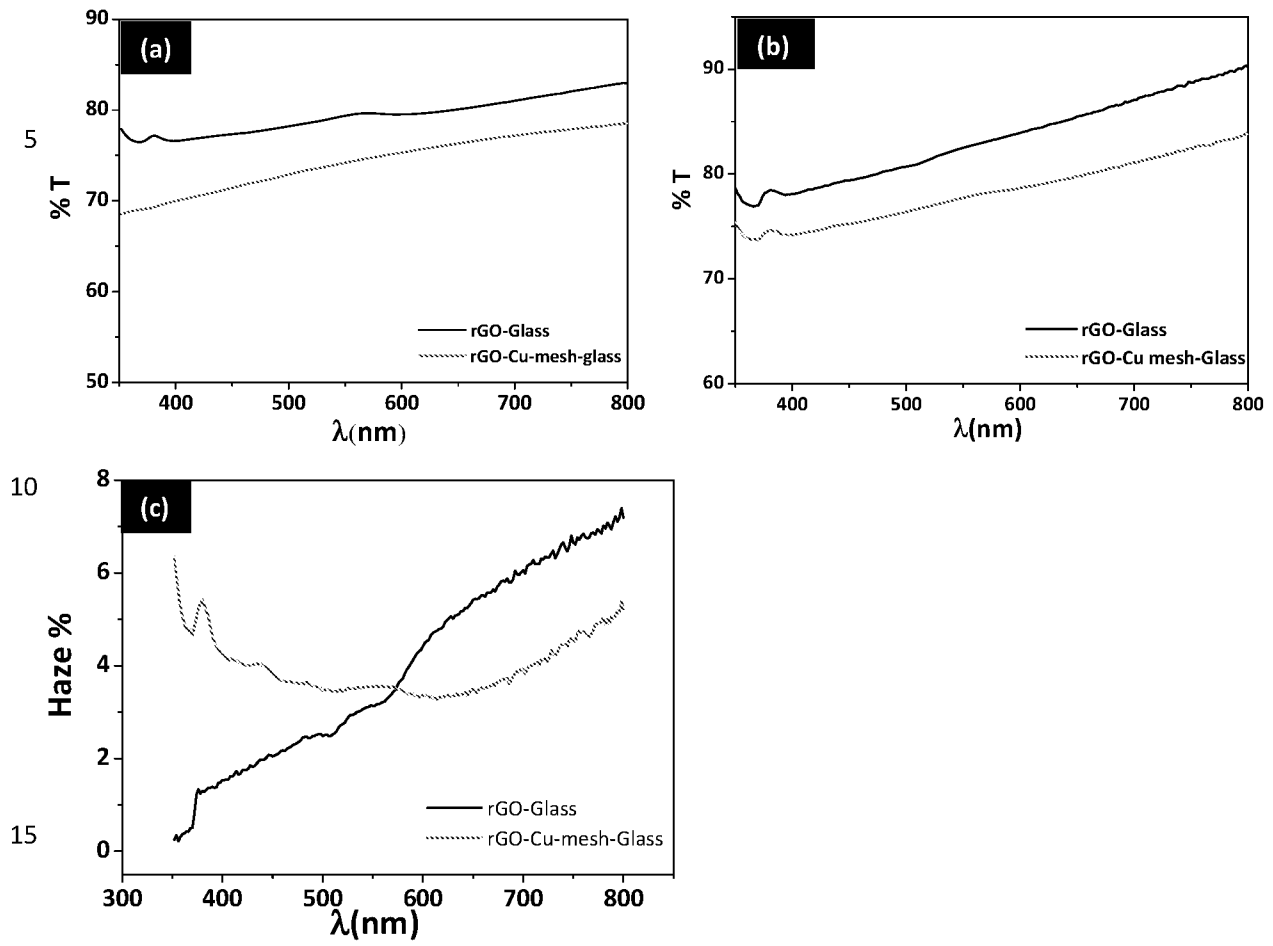


Figure 4

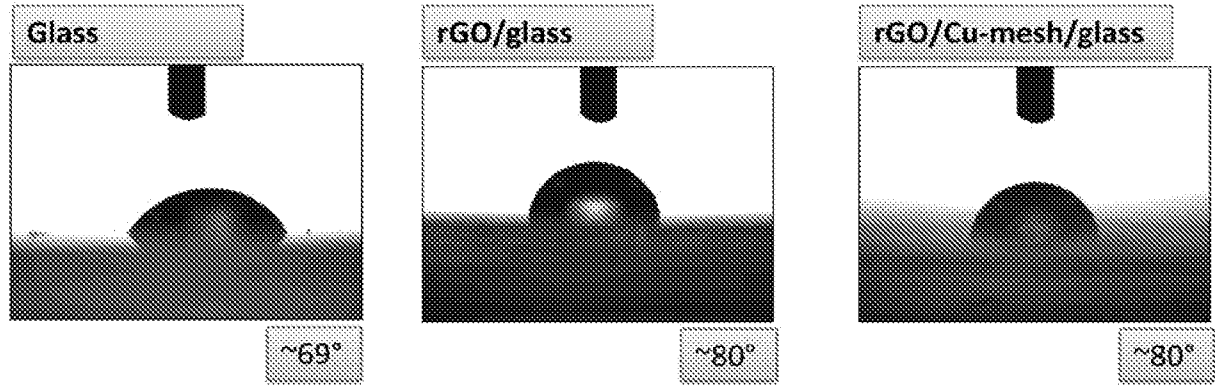


Figure 5

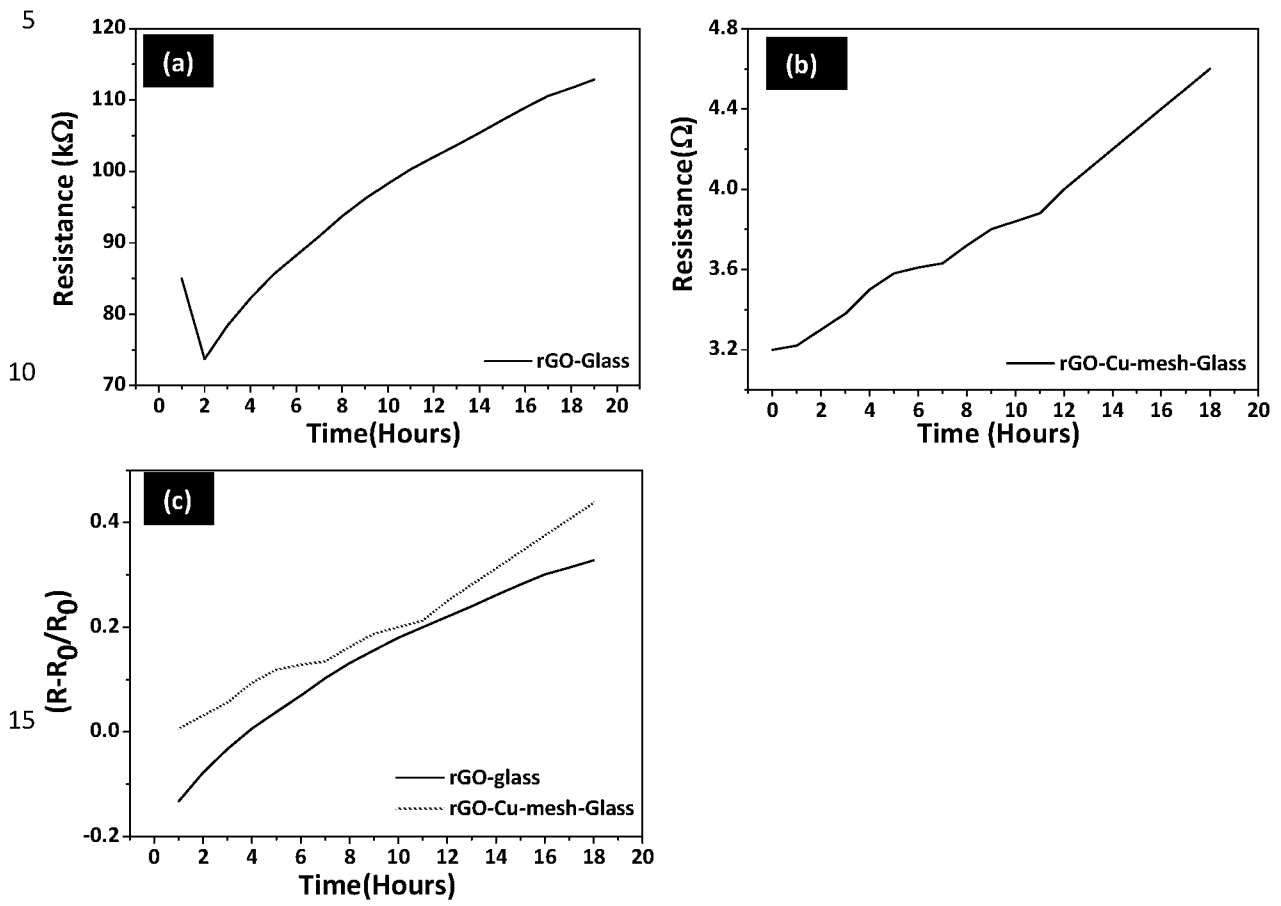


Figure 6

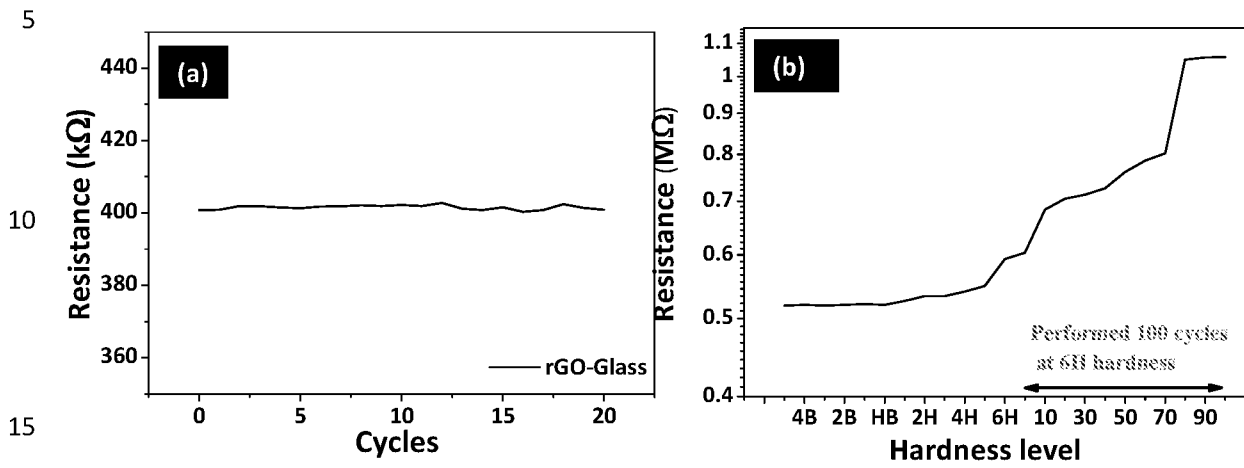


Figure 7

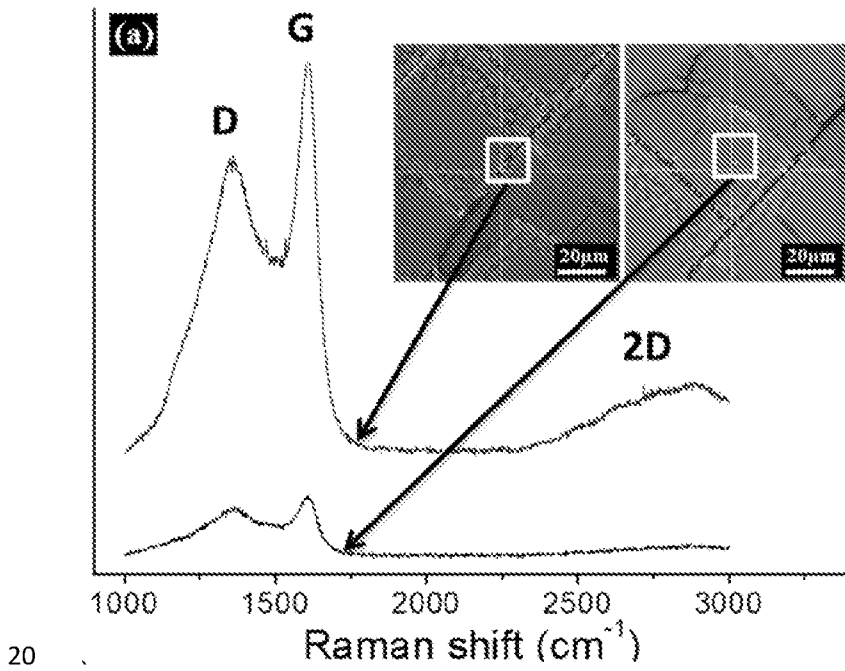


Figure 8

5/6

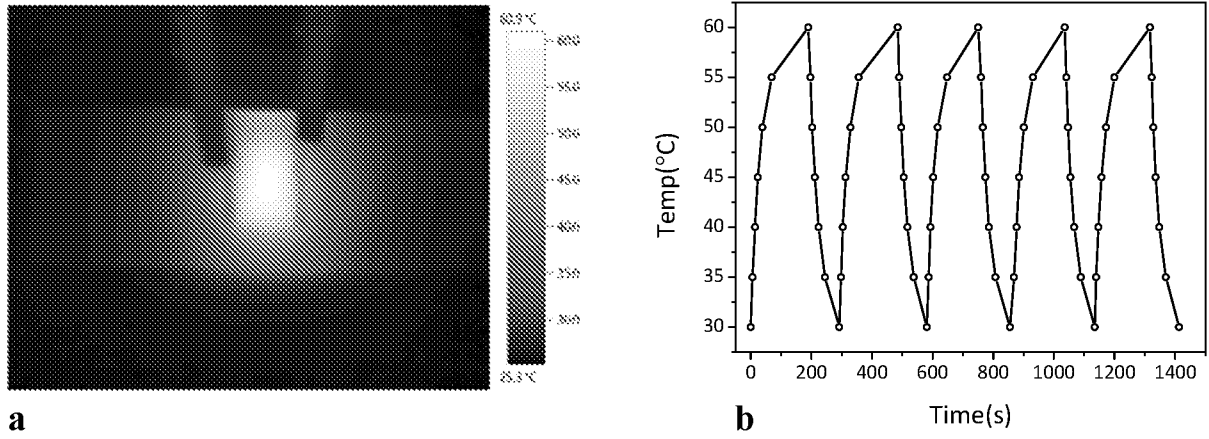


FIGURE 9

5

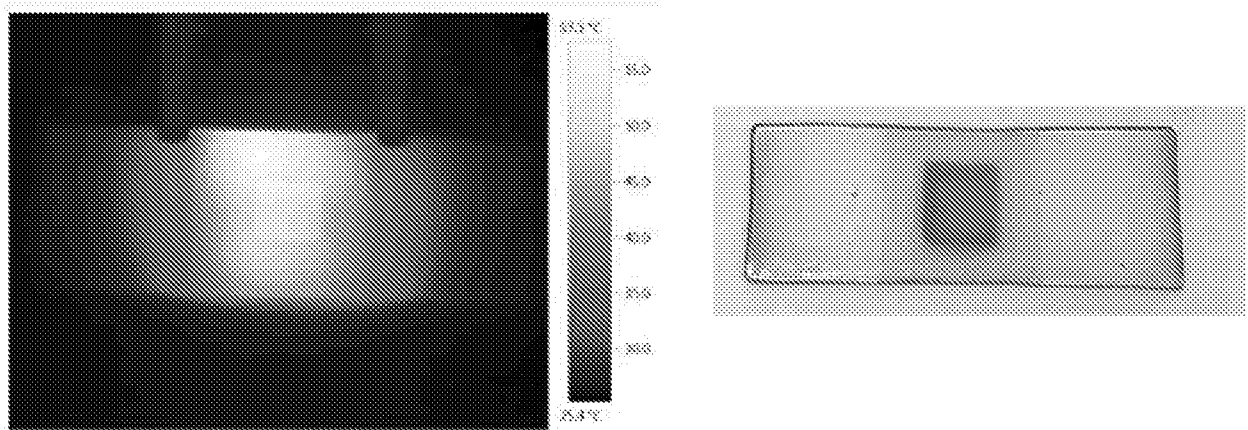


FIGURE 10

15

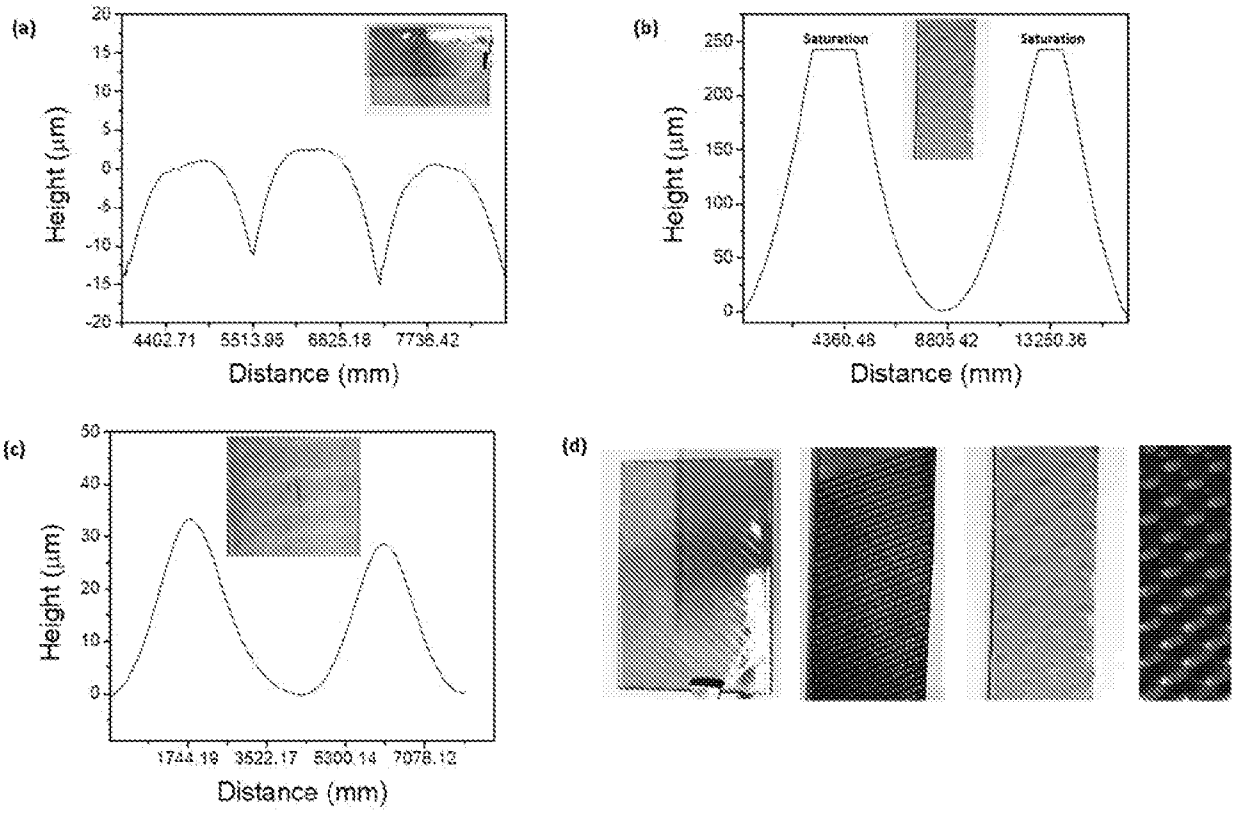


FIGURE 11

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/IB2018/059698

A. CLASSIFICATION OF SUBJECT MATTER H01L31/00, H01L51/00 Version=2019.01		
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		
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)  Databases: Total Patent One, IPO Internal Database Keywords: transparent electrode, graphene oxide, metal, substrate, glass		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	Qiu, Tengfei, et al. "Hydrogen reduced graphene oxide/metal grid hybrid film: towards high performance transparent conductive electrode for flexible electrochromic devices." Carbon 81 (2015): 232-238. 01 January 2015 (01-01-2015) URL: <a href="https://www.sciencedirect.com/science/article/pii/S0008622314009063">https://www.sciencedirect.com/science/article/pii/S0008622314009063</a> [retrieved on 28-02-2019] Abstract, Sections 1, 2.1, 3 & 4; Figures 1a-3d & 4b	1-13
X	----- Ahn, Yumi, et al. "Improved thermal oxidation stability of solution-processable silver nanowire transparent electrode by reduced graphene oxide." ACS applied materials & interfaces 4.12 (2012): 6410-6414. 13 December 2012 (13-12-2012) URL: <a href="https://pubs.acs.org/doi/10.1021/am301913w">https://pubs.acs.org/doi/10.1021/am301913w</a> [retrieved on 28-02-2019] Abstract, Introduction, Results and discussion; Figures 2, 3a & 4a	1-13
<input checked="" type="checkbox"/>	Further documents are listed in the continuation of Box C.	<input checked="" type="checkbox"/> See patent family annex.
* Special categories of cited documents:	"A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier application or patent but published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art "&" document member of the same patent family	
Date of the actual completion of the international search  06-03-2019	Date of mailing of the international search report  06-03-2019	
Name and mailing address of the ISA/ Indian Patent Office Plot No.32, Sector 14, Dwarka, New Delhi-110075 Facsimile No.	Authorized officer Sunil Sharan Yadav  Telephone No. +91-1125300200	

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/IB2018/059698

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US20130306361A1 [SAMSUNG ELECTRO MECHANICS CO LTD] 21 November 2013 (21-11-2013) Paragraphs [0015]-[0022], [0046]-[0058]; claims 1-7 & 10-12; figures 1 & 7-11	1-13

**INTERNATIONAL SEARCH REPORT**  
Information on patent family members

International application No.  
PCT/IB2018/059698

Citation	Pub.Date	Family	Pub.Date
US 20130306361 A1	21-11-2013	CN 103426941 A	04-12-2013
		JP 2014007147 A	16-01-2014
		KR 20130127781 A	25-11-2013