



(51) International Patent Classification:

H01L 31/0224 (2006.01) *H01G 9/048* (2006.01)
H01L 31/02 (2006.01) *H01G 9/20* (2006.01)
H01L 31/04 (2006.01) *H01G 9/022* (2006.01)

(21) International Application Number:

PCT/SG2012/000172

(22) International Filing Date:

18 May 2012 (18.05.2012)

(25) Filing Language:

English

(26) Publication Language:

English

(30) Priority Data:

61/487,822 19 May 2011 (19.05.2011) US

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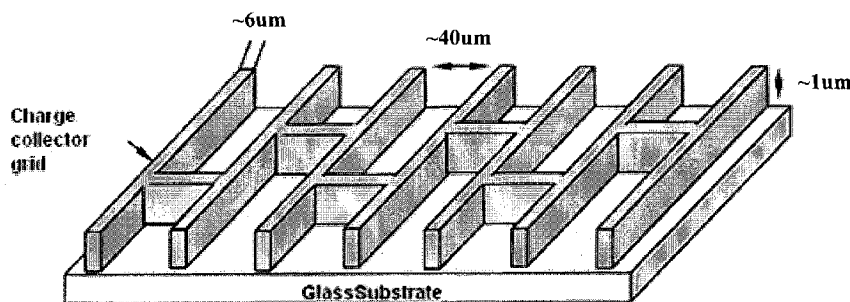
(81) **Designated States** (unless otherwise indicated, for every kind of national protection available): AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CZ, DE, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IS, JP, KE, KG, KM, KN, KP, KR, KZ, LA, LC, LK, LR, LS, LT, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SC, SD, SE, SG, SK, SL, SM, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.

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

Published:

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

(54) Title: AN IMPROVED PHOTOVOLTAIC CELL

**Figure 4**

(57) **Abstract:** Present invention relates to a photovoltaic cell comprising a first substrate, a first charge collector positioned adjacent to the first substrate, wherein the first charge collector comprises or consists of a first grid electrode comprised of a plurality of electrically connected individual electrodes spaced apart from one another and the plurality of individual electrodes extending from the surface of the first charge collector, wherein the spacing between two neighbouring individual electrodes is at most twice the electron diffusion length (EDL), a second charge collector, and a photoactive layer interposed between the first charge collector and the second charge collector.

AN IMPROVED PHOTOVOLTAIC CELL

Cross-Reference to Related Application

This application claims the benefit of priority of United States of America provisional patent application no. 61/487,822, filed 19 May 2011, the contents of which being hereby incorporated by reference in its entirety for all purposes.

Technical Field

The invention relates to photovoltaic cells, and in particular, to a photovoltaic cell having improved electrode design and arrangement.

Background

Conventional solar cells utilize transparent conducting oxides (TCO) such as indium tin oxide (ITO) or fluorinated tin oxide (FTO) as the charge collecting electrode.

Transparency in the electrode is necessary so as to allow illumination of a photoactive layer where initial charge separation occurs. ITO has a high transparency of about 90%, thereby making it attractive as a suitable choice for the transparent electrode. However, the high price of ITO due to a shortage in the supply of indium discourages its deployment at large scales. While FTO is cheaper than ITO, its transparency is lower than ITO. Other alternatives for the transparent electrode material include carbon nanotubes and graphene but scalability remains a concern.

In conventional dye-sensitized solar cells (DSSC), a thick film comprised of dye-adsorbed metal oxide nanoparticles acts as a photoanode and a platinum-coated FTO glass acts as a cathode with a redox electrolyte inserted between the electrodes, where

the dye molecule is the light absorber and the metal oxide nanoparticle is the charge transporter. Charge transport (typically electrons) through a network of interconnected nanoparticles and particle surface area available to dye adsorption and photogeneration are two critical elements in determining the power conversion efficiency (η) of a DSSC.

In conventional DSSC, the metal oxide nanoparticle layer is about 10 μ m thick and consists of titanium dioxide (TiO₂) nanoparticles which provide large surface area for the adsorption of dye molecules, leading to efficient photon harvesting. However, the photogenerated electrons may also be trapped at surface states, slowing down the charge transport and thus restricting the final thicknesses of the nanoparticle network. The performance of DSSC is characterized by the following parameters: open-circuit voltage (V_{OC}), short-circuit current (I_{SC}), fill factor (FF) and power conversion efficiency (η).

For DSSC, V_{OC} is obtained as the difference between quasi-Fermi level of the electrons under illumination and the redox level in the electrolyte. Longer electron transport distance significantly increases dark current and as a consequence, V_{OC} drops. I_{SC} is determined by the number of electrons injected into the TiO₂ layer and then collected by the anode. In addition, TiO₂ film of reasonable thickness is required to ensure efficient light harvesting, especially for long wavelength photons. Thus, the thickness of the TiO₂ layer plays an important role in determining the performance of DSSC.

Therefore, there is a need to provide for a photovoltaic cell having an improved design with high transparency, ease of scalability, and performance.

Summary

In a first aspect, there is provided a photovoltaic cell comprising:

- a first substrate;
- a first charge collector positioned adjacent to the first substrate,
 - wherein the first charge collector comprises or consists of a first grid electrode comprised of a plurality of electrically connected individual electrodes spaced apart from one another and the plurality of individual electrodes extending from the surface of the first charge collector,
 - wherein the spacing between two neighbouring individual electrodes is at most twice the electron diffusion length (EDL);
- a photoactive layer positioned adjacent to the first charge collector and first substrate;
- a second charge collector; and
- an electrolyte interposed between the first charge collector and the photoactive layer, and the second charge collector.

In various embodiments, the second charge collector may comprise or consist of a second grid electrode comprised of a plurality of electrically connected individual electrodes spaced apart from one another and the plurality of individual electrodes extending from the surface of the second charge collector. The spacing between two neighbouring individual electrodes may be varied to maximise interaction with the electrolyte, the hole or electron transport material.

In various embodiments, the thickness of each individual electrode of the first grid electrode measured in the longitudinal direction of the individual electrode is at least more than the difference between the thickness of the photoactive layer and the EDL.

In various embodiments, the width of each individual electrode of the first grid electrode and/or the second grid electrode measured in the transverse direction of the individual electrode is about one-tenth of the spacing between two neighbouring individual electrodes of the respective grid electrode.

In a second aspect, there is provided a charge collector comprising or consisting of a grid electrode comprised of a plurality of electrically connected individual electrodes spaced apart from one another and the plurality of individual electrodes extending from the surface of the charge collector, wherein the spacing between two neighbouring individual electrodes is at most twice the electron diffusion length (EDL).

In a third aspect, use of a charge collector of the second aspect in a photovoltaic cell, diode, transistor, electrochromic device, battery, supercapacitor, or waveguide is provided.

Brief Description of the Drawings

In the drawings, like reference characters generally refer to the same parts throughout the different views. The drawings are not necessarily drawn to scale, emphasis instead generally being placed upon illustrating the principles of various embodiments. In the following description, various embodiments of the invention are described with reference to the following drawings.

Figure 1A shows a schematic of continuous metal grid electrode after lithography patterning & e-beam deposition, photoresist is to be removed next, leaving only the patterned metal grid electrode on substrate; **Figure 1B** shows the resultant fully patterned 4" glass wafer; **Figure 1C** shows a 3D titanium grid electrode.

Figure 2 shows a schematic illustration of a 3D grid electrode DSSC fabrication process flow.

Figure 3 shows the difference in cell assembly between a standard DSSC and a 3D grid electrode DSSC.

Figure 4 shows a structure of the present 3D grid electrode deposited on plain glass (not to scale).

Figure 5 shows a comparison of transparency between a conventional FTO glass and the present 3D grid electrode substrate.

Figure 6 shows an I-V curve of the present 3D grid titanium electrode liquid DSSC.

Figure 7 shows impedance spectra of the present 3D grid titanium electrode based DSSC versus a standard FTO based DSSC, zoomed into the area where it is shown that the resistance of titanium electrode contact is lower than that of conventional FTO. The distance between 0-axis and the intersection of the impedance curve to the x-axis correspond to the resistance of the Ti or FTO contact.

Figure 8 shows I-V curve characteristics of the present 3D grid titanium electrode with various individual electrode thicknesses.

Figure 9A and 9B show electrodeposited 3D grid copper electrode and electrodeposited 3D grid tin electrode, respectively.

Figure 10 shows an illustration of the electron and hole transport in a standard liquid DSSC (prior art).

Figure 11 shows an illustration of the electron and hole transport in the present 3D grid electrode liquid DSSC.

Figure 12 shows a cell with a conventional FTO layer as a front electrode and the present 3D grid electrode as a counter electrode.

Figure 13 shows a cell with the present 3D grid electrode as a front electrode as well as as a counter electrode.

Figure 14 shows the I-V curve characteristics of the present 3D grid titanium electrode as counter electrode for a standard FTO front electrode and when paired with a 3D grid titanium front electrode.

Figure 15 shows a 3D cross-sectional view of the present 3D grid electrode DSSC.

Figure 16 shows a 2D cross-sectional view of the waveguide effect inside the 3D grid electrode.

Figure 17 shows a schematic illustration of present 3D grid electrode solid-state DSSC.

Description

The following detailed description refers to the accompanying drawings that show, by way of illustration, specific details and embodiments in which the invention may be practised. These embodiments are described in sufficient detail to enable those skilled in the art to practise the invention. Other embodiments may be utilized and structural, logical, and electrical changes may be made without departing from the scope of the invention. The various embodiments are not necessarily mutually exclusive, as some embodiments can be combined with one or more other embodiments to form new embodiments.

While the foregoing discussion relates to three-dimensional (3D) grid electrode structures utilized as charge (both electron and hole) collection electrodes in photovoltaic cell such as solar cells, it is to be understood that the use of the 3D grid electrode structures is not limited as such and can be utilized as charge injection electrodes in charge transport layers in diodes and transistors. The 3D grid electrodes can also be used as electrodes in electrochromic devices, batteries and supercapacitors, and planar waveguides, for example.

Application of a 3D grid electrode which possesses low sheet resistance and high transparency in a dye-sensitized solar cell (DSSC) is described herein. Sheet resistance

of most metals is about 1 ohm/sq or less, compared to a conventional fluorinated tin oxide (FTO) electrode which has sheet resistance of about 10 ohm/sq or more. Correct matching of the work function of metals such as titanium permits a smooth transfer of electrons from TiO₂ nanoparticles to the electrode. The high surface area of the electrode allows more efficient collection of charges from the dye-sensitized nanoparticle network and allows fabrication of solar cells with thicker photoactive layers. These advantages, along with its low resistivity and the possibility of employing low cost metal deposition methods, make this 3D grid electrode structure particularly attractive for photovoltaic and other applications.

The present 3D grid electrode acting as a charge collector can be used in a DSSC to replace conventional FTO-coated glass as front electrode. In other electronic devices, the 3D grid electrode structure provides a large interfacial area, allowing for more effective charge injection.

Thus, one aspect of the present invention provides a photovoltaic cell comprising a first substrate and a first charge collector positioned adjacent to the first substrate.

In the present context, "substrate" refers to a supporting surface or portion of the photovoltaic cell. The substrate may be transparent, translucent, or opaque. A suitable substrate can include, but is not limited to a polymer/plastic or glass. A transparent material allows radiation, such as light, to pass through the material. The opposite property is opacity. Transparent materials are clear (i.e. they can be seen through). A translucent material allows only radiation of a specific wavelength or wavelength range to pass through it.

The substrate can be a supporting surface or portion on which a charge collector is contacted. The substrate can be any substrate suitable to support the charge collector. The substrate may be transparent or translucent. In a photovoltaic cell, it would be necessary that light shines through the substrate towards the interior of the photovoltaic cell. A suitable substrate can include, but is not limited to a glass. For example, the substrate may be a glass with or without a coating thereon, such as an indium tin-oxide glass (i.e. ITO glass or ITO-coated glass) or a fluorine-doped tin oxide glass (i.e. FTO glass or FTO-coated glass). Non-glass based substrates such as those comprising polymers (e.g. polyethylene terephthalate, polyethylene naphthalate, or polydimethylsiloxane) with or without a coating thereon, such as a thin transparent oxide layer (for example; TiO_2 or Al_2O_3) may also be employed.

In the present context, “charge collector” refers to a charge collecting electrode. The collected charges may be electrons or holes, and the electrode may be an anode or a cathode, respectively. When the charge collector is said to be positioned adjacent to a substrate, it is to be understood that the charge collector may be positioned immediately next to (i.e. having direct contact) or close to (i.e. having indirect contact via an intermediate therebetween) the substrate. When an electrode is positioned adjacent to a semiconductor active layer, it collects the majority carrier (in DSSC; electrons) and is commonly termed a front electrode. The electrode that is positioned away from the semiconductor active layer collects holes and is commonly termed a counter electrode. In a DSSC, this counter electrode is mediated from the semiconductor active layer by an electrolyte. In various embodiments, the first charge collector is the front electrode and the second charge collector is the counter electrode.

There are two types of light illumination in a liquid DSSC:

- Front electrode (FE) illumination is when the light passes through the substrate adjacent to the front electrode and hits the semiconductor active layer. This will typically induce more electrons generated in the semiconductor active layer.
- Counter electrode (CE) illumination is when light passes through the substrate adjacent to the counter electrode and the electrolyte before it hits the semiconductor active layer, resulting in some loss of electron generation.

The photovoltaic cell further comprises a photoactive layer positioned adjacent to the first charge collector and first substrate. Similarly, when the photoactive layer is said to be positioned adjacent to the first charge collector, it is to be understood that the photoactive layer may be positioned immediately next to (i.e. having direct contact) or close to (i.e. having indirect contact via an intermediate therebetween) the first charge collector.

The photovoltaic cell further comprises an electrolyte interposed between the first charge collector and the photoactive layer, and the second charge collector. In various embodiments, the photoactive layer is comprised of a nanoporous structure and the electrolyte can penetrate into the pores of the photoactive layer. In the present context, “interposed” refers to a sandwich arrangement whereby the first charge collector and the photoactive layer are positioned on one side of the electrolyte while the second charge collector is positioned on the other side of the electrolyte. In various embodiments, the electrolyte wraps or encapsulates the photoactive layer not in contact with the first charge collector. The electrolyte is in contact with the photoactive layer so that the

minority charge carriers or holes are transported via the electrolyte to the second charge collector. Suitable liquid electrolytes may include, but is not limited to, iodine/iodide redox shuttle based electrolyte, cobalt based electrolyte, and ferrocene based electrolyte.

As mentioned above, the first charge collector may form the front electrode and the second charge collector may form the counter electrode. The front electrode is an electron collector and the counter electrode is a hole collector.

The photoactive layer and the electrolyte or charge transport layers which are interposed between the first charge collector and the second charge collector electrochemically connects the two charge collectors, with sufficient efforts made to prevent short circuit formation. The photoactive layer absorbs incident light and exploits the light energy to induce the generation of electrons and holes which are subsequently injected through appropriate semiconductors and collected at the respective electrodes. The photoactive layer may also be termed a semiconductor layer.

For example, in a liquid DSSC the photoactive layer, which is comprised of a mesoporous semiconductor layer (typically but not limited to TiO_2) and dye sensitizer molecules, is permeated by the liquid electrolyte. Charge transfer may take place at the interface between the dye sensitizer molecules and the semiconductor layer, dye sensitizer molecules and the electrolyte, semiconductor layer and the electrolyte, electron collector and semiconductor layer, electron collector and the electrolyte, as well as hole collector and the electrolyte.

Another example is a solid-state DSSC wherein the photoactive layer is comprised of a semiconductor material and dye sensitizer molecules, permeated by solid or semi-solid electrolyte. Charge transfer may take place at the interface between the dye sensitizer molecules and the semiconductor layer, dye sensitizer molecules and the electrolyte, semiconductor layer and the electrolyte, electron collector and semiconductor layer, electron collector and the electrolyte, as well as hole collector and the electrolyte.

The first charge collector comprises or consists of a first grid electrode.

In the present context, “grid” refers to a structure of intersecting axes. Advantageously the grid is formed of three intersecting axes (i.e. 3D grid) or more. Each axis may be substantially orthogonal to one another. It is to be understood that by “substantially orthogonal” is meant substantially perpendicular, non-overlapping, varying independently, or uncorrelated and is not necessarily limited to be absolutely perpendicular.

The first grid electrode is comprised of a plurality of electrically connected individual electrodes spaced apart from one another, the plurality of individual electrodes extending from the surface of the first charge collector. The plurality of individual electrodes extend in such a way that a three-dimensional grid electrode structure is formed. The longitudinal axis of each individual electrode intersects at a common axis of the grid electrode. In one embodiment, the grid electrode is formed of a comb-like structure.

The spacing between two neighbouring individual electrodes is at most twice the electron diffusion length (EDL) of the photovoltaic cell. EDL is defined as the distance which a photo-generated electron can travel before it is lost due to recombination. The longer it has to travel, the less likely it is to be collected. The EDL is dependent on the materials used in the construction of the photovoltaic cell system.

By restricting the maximum spacing between two neighbouring individual electrodes to twice the EDL, even the farthest generated electron can be collected efficiently. For instance, in a standard liquid DSSC which EDL is about 25 μm , the maximum spacing of the grid electrode is about 50 μm so that even electron generated at the centre between two neighbouring individual electrodes can still be collected. In another example, in a standard solid-state DSSC which EDL is only about 5 μm , the maximum spacing of the grid electrode is about 10 μm . If two neighbouring individual electrodes are spaced more than twice the EDL apart, inefficient electron collection may result.

Thus, in various embodiments, for practical implementation purposes, the maximum spacing between two neighbouring individual electrodes of the grid electrode of the first charge collector is kept at between about 1 and 100 μm .

The incorporation of a 3D grid electrode of various embodiments in photovoltaic cells is advantageous. The 3D grid electrode may provide a direct and short conduction pathway from the point of electron-hole pair generation to the charge collector and may improve the electron transport efficiency. By improving the electron transport efficiency, the conversion efficiency of photovoltaic cells may be improved. Therefore, the 3D grid

electrode of various embodiments may be developed for high efficiency or high conversion efficiency photovoltaic cells.

In various embodiments, the photovoltaic cell further comprises a second substrate, wherein the second charge collector is positioned adjacent to the second substrate.

In various embodiments, the second charge collector comprises or consists of a second grid electrode comprised of a plurality of electrically connected individual electrodes spaced apart from one another and the plurality of individual electrodes extending from the surface of the second charge collector. In such embodiments, the second grid electrode may have the same or different structure as the first grid electrode. In further embodiments, any distance is suitable for the spacing between two neighbouring electrodes of the grid electrode of the second charge collector so long as an interface between platinum (to be deposited adjacent to the second charge collector) and the electrolyte is provided. The spacing between two neighbouring individual electrodes may be varied to maximise interaction with the electrolyte, the hole or electron transport material such that maximum efficiency of the photovoltaic cell is achieved.

In various embodiments, the plurality of individual electrodes of the first grid electrode and/or the plurality of individual electrodes of the second grid electrode extend substantially perpendicularly from the surface of the respective charge collector. It is to be understood that the plurality of individual electrodes of the first grid electrode and/or the plurality of individual electrodes of the second grid electrode may extend at an angle less than or more than 90 degrees to the surface of the respective charge collector. For

example, the angle may be 60, 65, 70, 75, 80, 85, 95, 100, 105, 110, 115, or 120 degrees to the surface of the charge collector.

In various embodiments, the plurality of individual electrodes of the first grid electrode extend from the surface of the first charge collector towards the second charge collector.

In certain embodiments, the plurality of individual electrodes of the second grid electrode extend from the surface of the second charge collector towards the first charge collector. It is to be understood that the plurality of individual electrodes of the first grid electrode extend into the photoactive layer but not the individual electrodes of the second grid electrode.

In various embodiments where front electrode illumination occurs, the thickness of each individual electrode of the first grid electrode measured in the longitudinal direction of the individual electrode is at least more than the difference between the thickness of the photoactive layer and the EDL. It is to be understood that the longitudinal direction of the individual electrode is in the direction of extension of the individual electrode from the surface of the charge collector.

Advantageously, the thickness of each individual electrode of the first grid electrode is more than or equal to the thickness of the photoactive layer minus the EDL, so that even electrons generated at the outer end of the photoactive layer can still be collected efficiently. The implication of the thickness factor is more important in photovoltaic systems with low EDL. For instance, in standard liquid DSSC where the EDL is about 25um, if the thickness of the semiconductor TiO_2 layer is about 10um, the performance of the photovoltaic cell is not much affected whether the thickness of each individual

electrode of the grid electrode is 1 μ m or 5 μ m, provided there is no change in the electrode conductivity. On the other hand, in standard solid-state DSSC where EDL is about 5 μ m, if the thickness of the semiconductor TiO₂ layer is about 10 μ m, then a thickness of each individual electrode of the grid electrode of 1 μ m is not sufficient to efficiently collect all the generated electrons compared to a thickness of at least 5 μ m.

In various embodiments where front electrode illumination occurs, transparency is not an issue to the counter electrode. Hence, ideal dimensions (spacing, thickness, width of the individual electrodes of the second grid electrode) should be optimized to provide maximum interface of platinum-electrolyte. In such embodiments, the thickness of each individual electrode of the second grid electrode measured in the longitudinal direction of the individual electrode is at least more than half the spacing between two neighbouring individual electrodes of the second grid electrode.

By providing a thickness of more than half the spacing between two neighbouring individual electrodes of the second grid electrode, at least an equivalent length of a conventional flat electrode is provided. For example, if the spacing is about 5 μ m and the width of an individual electrode is about 3 μ m, then the equivalent length of a conventional flat electrode is about 8 μ m. To provide an equivalent surface length of contact with the electrolyte (i.e. 8 μ m long), the thickness of the individual electrode is set to be at least 2.5 μ m such that the sum of the width and two longitudinal sides of the individual electrode exposed to the electrolyte is at least 8 μ m. Hence, in general for front electrode illumination, the smaller the spacing between two individual electrodes, the shorter the thickness is required.

For practical implementation purposes, the ability to deposit thin individual electrodes is limited by the lithography and metal deposition techniques. Nevertheless, as will be discussed in the examples found below, the results obtained by using the present photovoltaic cell are comparatively good, which may be due to the better conductivity of the metal titanium than FTO even though the actual surface area exposed to the electrolyte is smaller than that of a flat FTO.

Thus, in various embodiments, for practical implementation purposes, the thickness of each individual electrode of the first grid electrode and/or the thickness of each individual electrode of the second grid electrode is kept at between about 1 and 20um.

In various embodiments where front electrode illumination occurs, the width of each individual electrode of the first grid electrode measured in the transverse direction of the individual electrode is about one-tenth of the spacing between two neighbouring individual electrodes of the first grid electrode. It is to be understood that the transverse direction of the individual electrode is in the direction orthogonal to the longitudinal direction of the individual electrode.

The width of each individual electrode of the first grid electrode is primarily useful to adjust for transparency. Optimal transparency is about 90% (which is comparable to the transparency of a conventional 8-15 ohm/sq FTO glass). Advantageously, the width of each individual electrode of the first grid electrode is about one-tenth of the spacing between two neighbouring individual electrodes of the first grid electrode. For example, if the spacing between the individual electrodes of the first grid electrode is only about 10um and the width of each individual electrode of the first grid electrode is about

10 μ m, then the theoretical transparency will be only about 50%. This will greatly reduce the overall current generation and collection, which leads to lower efficiency. On the other hand, if the width can be trimmed down to 1 μ m, the theoretical transparency jumps up to about 90%, providing sufficient incoming photon to ensure high efficiency.

In various embodiments where front electrode illumination occurs, transparency is not an issue for the counter electrode. Hence, any distance is suitable for the width of an individual electrode and the spacing between two neighbouring individual electrodes of the second grid electrode. In practical implementations, the width and the spacing are limited by the capability of the patterning technique employed.

In yet further embodiments where counter electrode illumination occurs, the width of each individual electrode of the second grid electrode measured in the transverse direction of the individual electrode is about one-tenth of the spacing between two neighbouring individual electrodes of the second grid electrode. It is to be understood that the above discussion with respect to the width of each individual electrode of the first grid electrode also applies to the width of each individual electrode of the second grid electrode.

Thus, in various embodiments, for practical implementation purposes, the width of each individual electrode of the first grid electrode and/or the width of each individual electrode of the second grid electrode is kept at between about 50nm and 5 μ m, depending on the material and fabrication technique of the individual electrode.

The grid electrode structural design involves optimization for electron diffusion length and light transparency. For materials with low electron diffusion length, the individual electrodes have to be brought closer together. Hence, the spacing therebetween the individual electrodes becomes narrower and transparency reduces.

Further improvements in transparency may be achieved by either: -

- (a) Increase in the spacing between the individual electrodes. However, there is a practical limit on how far apart the individual electrodes can be. If the spacing is too far apart, electrons generated from the central area between two neighbouring individual electrodes may not be collected by the charge collector. Theoretical EDL in liquid DSSC is normally calculated to be about 40 μ m. However, it is common that in actual experiments, the EDL is only about 25 μ m. This means that any electron generated farther than 25 μ m away from the individual electrode may not be collected; or
- (b) Decrease in the width of each individual electrode. With narrower individual electrodes, it is possible to include more individual electrodes in a given space so that the surface area increases without compromising transparency. However, in practice, this may be limited by the photolithography capability.

In order that the invention may be readily understood and put into practical effect, particular embodiments will now be described by way of the following non-limiting examples.

Examples

Application of lithographically patterned titanium-based 3D grid electrode as a replacement to the conventional FTO front electrode in liquid DSSC is demonstrated below. The 3D grid electrode consists of parallel rows of conducting material interconnected by bridges to establish a conducting path throughout the structure. The spacing between the rows provides transparency. A significant factor is the thickness of the grid electrode which provides surface area for charge collection and injection. Data presented here is from grids made of titanium. The material of choice is not limited to titanium, and may include any conducting materials including carbon nanostructures and metal oxides.

3D Grid Metal Electrode (GME) Preparation

First, a 4" glass wafer was thoroughly cleaned by ultra-sonication in a soap solution, followed by ethanol. Next, a layer of about 5 μ m lift-off resist (LOR30B) was spin-coated on top of the glass and baked at 200°C for 5 minutes. After cooling down to room temperature, a layer of approximately 6 μ m photoresist (AZ9260, positive) was spin-coated on top of the lift-off resist and then baked at 120 °C for 6 minutes. Subsequently, the lithography was done using Karl Suss Mask Aligner and lift-off mask. Developing process was done in a mixture of 1:3 AZ400K and DI water. Titanium is deposited on top of this substrate by e-beam evaporation to form a 1 μ m thick film. The substrate structure after deposition is shown in **Figure 1A**. After that, lift-off using Microchem PG remover is carried out. At this point, the fully patterned 4" glass wafer is shown in **Figure 1B**. Lastly, the 4" wafer is diced into a plurality of grid electrodes shown in **Figure 1C**.

DSSC Testing

The cell testing is done using solar simulator purchased from San-Ei Electric (301 S), which is connected to Agilent 4155C for electrical measurements.

Result and Discussion

A physical diagram representative of the present 3D grid electrode structure is shown in **Figure 4**. The experimental transparency of this 3D grid electrode structure is about 83% (measured by Shimadzu UV-Vis 3600 Spectrometer, transmittance mode), shown in **Figure 5**.

Testing of solar cells with an active area of 0.1199 cm^2 revealed that the 3D grid electrode based solar cells obtained an efficiency of 6.2%, which is comparable to a dye-sensitized solar cell fabricated through a standard route (7.1 %). The 3D grid electrode based DSSC had $V_{oc} = 718 \text{ mV}$, $J_{sc} = 11.72 \text{ mA/cm}^2$ and $FF = 73.5\%$ at 1 sun illumination. Standard DSSC fabricated with the same materials had a V_{oc} of 730 mV , $J_{sc} = 13.3 \text{ mA/cm}^2$ and $FF = 72\%$ at 1 sun illumination (**Figure 6**). The 3D-DSSCs have an improved fill factor due to the fact that titanium metal electrode has a much lower sheet resistance compared to the conventional FTO (around 0.8 ohm/sq for Ti metal electrode compared to $8\text{-}15 \text{ ohm/sq}$ for conventional FTO.) This reduces the total series resistance of the cell and thus improves the fill factor.

Electrochemical Impedance Spectroscopy has been done to compare series resistance behaviour of the metal grid electrode compared to the standard FTO based cell. The distance between point of origin and the start of the impedance curve represents the series resistance in the metal grid electrode or in the FTO. **Figure 7** clearly shows that the titanium metal grid electrode has lower series resistance compared to the FTO layer.

In spite of the lower transparency compared to FTO, the cell fabricated with this 3D grid electrode structure showed comparable efficiency to the standard cell fabricated on FTO. This signifies that charge collection through the titanium metal electrode is very efficient. Thickness of at least 1µm is needed for efficient charge collection. **Table 1** below shows the comparison of cells made out of exactly same materials, but with different Ti grid thickness; 50nm, 100nm, 400nm, 700nm, and 1µm. This problem fades away as the thickness the individual electrode of the grid electrode increases.

	Std cell	50nm 3DMGE cell	100nm 3DMGE cell	400nm 3DMGE cell	700nm 3DMGE cell	1µm 3DMGE cell
Voc (V)	0.73	0.62	0.71	0.77	0.77	0.72
Jsc (mA/cm ²)	13.3	0.14	7.5	7.43	8.53	11.7
FF	0.72	0.29	0.34	0.67	0.67	0.73
Efficiency	7.1%	0.026%	1.8%	3.85%	4.4%	6.2%

Table 1 Comparison of Cell Performance with Different Individual Electrode Thickness

A study regarding the spacing between neighbouring individual electrodes has been made. It is deduced that 40µm is the optimum spacing for the present photovoltaic system, since this provides a reasonable transparency while still staying within the range of electron diffusion length. With spacing narrower than 40µm, cell efficiency is bottlenecked by the low transparency. Above 40µm, transparency is good, but it is pushing the electron diffusion length limit. **Table 2** shows the result of this experiment. The cells are constructed with exactly same materials for all cases.

	10µm spacing	20µm spacing	40µm spacing	60µm spacing
Transparency	55%	72%	83%	86%
Efficiency	4.9%	5.82%	6.2%	5.9%

Table 2 Comparison of Cell Performance with Different Individual Electrode Spacing

It is also shown that evaporation technique is not the only way to grow the metal electrode. Images of copper and tin metal electrode grown via electrodeposition route are shown by **Figures 9A and 9B**, respectively. Other metal oxides such as zinc oxide may also be grown via electrodeposition technique.

Proposed Device Architecture and Comparison with Existing DSSC

(A) Front Electrode FTO Replacement

Figures 10 and 11 show the electron and hole transport in both standard and 3D-electrode liquid DSSC. The conventional structures associated with nanostructured solar cells are designed for vertical charge collection. This implies that the electrodes which will form the final charge collectors are situated above and below the photoactive layer. In the Graetzel cell (**Figure 10**), the illumination of the dye attached TiO_2 nanoparticle layer results in the formation of electron-hole pairs. These charge carriers are then collected at the top and bottom electrodes. As illustrated in the figure, the electrons take a tortuous route through the nanoparticles before being collected by the bottom fluorine-doped tin oxide (FTO) electrode. This long electron path can lead to higher chances of recombination and trapping, thereby reducing cell efficiencies. This also puts an upper limit of the thickness of the nanoparticle layer.

In the 3-D grid electrode modification of the solar cell (**Figure 11**), the distance to be travelled by the electron is reduced because of the smaller spacing between the current collection electrodes. This ensures higher charge collection efficiency. The increased height of the individual electrode also removes the restriction on the thickness of the

semiconductor nanoparticle layer in the DSSC. As a consequence, a much thicker TiO_2 film having much more efficient light harvesting capability (especially for long wavelength photons) could be used. And it becomes realistic to employ alternative fast redox couples.

(B) Counter Electrode FTO Replacement

As the extension to the previously discussed front electrode FTO replacement, the present 3D grid electrode is utilized as the counter electrode FTO replacement. In this experiment, the 3D grid is integrated as counter electrode to both the standard FTO front electrode (**Figure 12**), as well as to the 3D grid front electrode cell (**Figure 13**). Two layer of Pt (as catalyst) are deposited on top of these 3D grid counter electrode.

When the standard FTO front electrode is paired with the present 3D grid counter electrode, a 6.26% efficiency was obtained (with V_{oc} of 0.76V, J_{sc} of $12.3\text{mA}/\text{cm}^2$, and FF of 0.67). When the 3D grid front electrode is paired with the 3D grid counter electrode (i.e. completely FTO-less cell), a 3.63% efficiency was obtained (with V_{oc} of 0.73V, J_{sc} of $6.98\text{mA}/\text{cm}^2$, and FF of 0.71). These results suggest that the present 3D grid electrode is able to replace FTO completely in the DSSC fabrication. **Figure 14** shows the IV characteristics of these cells.

Integration of Planar Waveguide in 3D Grid Electrode DSSC

Light enhancement is a good strategy to boost efficiency in solar cell technology. The present 3D grid electrode may be transformed into a waveguide for optical enhancement with slight modification. A waveguide is a structure which guides waves and in this case, light. Light is confined inside the waveguide material, reflected along the path

until it reaches the end. One of the most well-known examples of waveguide is the optical fiber which takes shape of a cylindrical fiber. The design of the 3D cell allows for the integration of optical enhancers such as a planar waveguide. **Figures 15 and 16** show the graphical representatives of such configurations.

Having flexibility in choosing materials allows for this planar waveguide integration. Material of the grid electrode ought to have lower refractive index compared to the surrounding layer. This will thus allow light which pass through the glass substrate to be reflected inside the grid, as if it is guided along and throughout the grid. Silica coated with ITO works very well because it is transparent to light and has matching refractive indexes. ITO is also conducting, providing collection pathway of the generated electron. The integration of this planar waveguide may increase the photon absorption dramatically. As discussed above, it has been shown that DSSC made out of a 3D metal grid electrode was able to reach efficiency comparable to that of a standard FTO-based DSSC. The main loss is due to transparency and the inactive area which is shadowed by the opaque metal grid. By integrating this planar waveguide concept, the advantages are twofold. First, since ITO is transparent, there will be no inactive area shadowed by opaque grid. Secondly, since light is reflected inside the grids, thicker active layer can be utilized because light can effectively penetrate deeper, resulting in more electrons generated by the active layer.

In conclusion, a 3D grid electrode design comprising rows of conducting material in a grid-like pattern has been described. The grid-like electrode's inter-spacing, width and thickness are varied to provide controllable interfacial area, transparency and conductivity. The design utilized the thickness of the conducting material of the

individual electrode to assist in charge collection. 3D charge collection is achieved by relying not only on the width of the conducting material of the individual electrode, but also on the thickness of the structure. The rows of electrode design may be connected by conducting bridges to provide fault tolerance and enhanced conductive properties. The electrodes described herein can be formed on substrates such as glass, plastic, metal foils and ceramics having properties of rigidity, flexibility, transparency, chemical and temperature resistance.

The conducting material and the connecting bridges of the individual electrodes can be made of metal, metal alloys, conducting carbon nanostructures, metal oxides, metal chalcogenides, metal nitrides or combinations thereof, which provide high conductivity, high surface area, work function matching, ease of deposition, and stability to downstream fabrication process. The conducting material can be deposited through techniques including, but not limited to, thermal and e-beam evaporation, sputtering, inkjet printing, electro-deposition, electroless deposition, spray coating and combinations thereof. The 3D grid electrode can be fabricated by processes such as photolithography, nanoimprint lithography, microcontact printing, transfer printing, inkjet printing, screen printing.

The surface properties of the electrodes may be modified to control properties such as wettability and surface energy through chemical treatments (e.g. grafting of self-assembled monolayers) and/or physical treatments (e.g. plasma treatment). The surface of the 3D grid electrodes may be converted by chemical and physical treatments (oxidation, sulphidation) to provide properties such as hole blocking or electron blocking effects and photosensitivity.

The surface of the 3D grid electrodes may be modified by attaching photosensitive molecules, quantum dots or by deposition of photoactive layers. The 3D grid electrodes may be modified to act as nucleation centres for the growth of nanostructures either through physical (for example, vapour-liquid-solid growth and chemical vapour deposition) or chemical means (for example, hydrothermal and anodization).

Application of the present 3D grid front electrode which has lower sheet resistance compared to the conventional FTO electrode has been demonstrated herein using titanium. The implications of changing the conventional FTO front electrode to a 3D grid electrode include: -

- (i) Interface area between the front electrode and TiO_2 layer will be increased.
- (ii) Tuning of the transparency of the grid electrode by modifying the dimension of the individual metal electrodes and the spacing between them.
- (iii) Modulation of the electron transfer resistance between front electrode and TiO_2 layer by choosing appropriate metal.
- (iv) Low sheet resistance of the front electrode.
- (v) Possibility of integrating other patterning methods such as nano imprint lithography (NIL) to further bring down the cell fabrication cost.

The advantages of the present 3D grid electrode structure include: -

- (i) The 3D grid electrode allows the replacement of costly transparent conducting oxides such as ITO/FTO which find application in solar cells and light emitting diodes.

- (ii) A range of metals can be used to fabricate the grid structure e.g. Ti, Sn, Zn, Al whose work functions provide flexibility to work with a variety of charge transport layers.
- (iii) The 3D architecture provides higher surface areas with comparable transparencies. As an example, for an electrode area of 1cm x 1cm, replacing the FTO with metal columns with 5um width and 10um thick while maintaining 90% transparency, results in an electrode with more than three times the surface area.
- (iv) Reduced spacing between the individual electrodes ensures higher charge collection efficiency.
- (v) The metal layers can be used to provide additional functionalities by acting as seeds for the growth of subsequent layers.
- (vi) The 3D grid electrode can be modified to provide specific surface area, transparency and conductivity.
- (vii) This structure can be fabricated by standard techniques of metal deposition (electroplating, sputtering, evaporation, and possibly inkjet printing) and is compatible with non-standard substrates such as glass and plastic.
- (viii) Possibility integration of planar waveguide to improve light absorption efficiency. This integration can be done with very little change to the materials deposition process.

By "comprising" it is meant including, but not limited to, whatever follows the word "comprising". Thus, use of the term "comprising" indicates that the listed elements are required or mandatory, but that other elements are optional and may or may not be present.

By "consisting of" is meant including, and limited to, whatever follows the phrase "consisting of". Thus, the phrase "consisting of" indicates that the listed elements are required or mandatory, and that no other elements may be present.

The inventions illustratively described herein may suitably be practiced in the absence of any element or elements, limitation or limitations, not specifically disclosed herein.

Thus, for example, the terms "comprising", "including", "containing", etc. shall be read expansively and without limitation. Additionally, the terms and expressions employed herein have been used as terms of description and not of limitation, and there is no intention in the use of such terms and expressions of excluding any equivalents of the features shown and described or portions thereof, but it is recognized that various modifications are possible within the scope of the invention claimed. Thus, it should be understood that although the present invention has been specifically disclosed by preferred embodiments and optional features, modification and variation of the inventions embodied therein herein disclosed may be resorted to by those skilled in the art, and that such modifications and variations are considered to be within the scope of this invention.

By "about" in relation to a given numerical value, such as for temperature and period of time, it is meant to include numerical values within 10% of the specified value.

The invention has been described broadly and generically herein. Each of the narrower species and sub-generic groupings falling within the generic disclosure also form part of

the invention. This includes the generic description of the invention with a proviso or negative limitation removing any subject matter from the genus, regardless of whether or not the excised material is specifically recited herein.

Other embodiments are within the following claims and non-limiting examples. In addition, where features or aspects of the invention are described in terms of Markush groups, those skilled in the art will recognize that the invention is also thereby described in terms of any individual member or subgroup of members of the Markush group.

Claims

1. A photovoltaic cell comprising:
 - a first substrate;
 - a first charge collector positioned adjacent to the first substrate,
 - wherein the first charge collector comprises or consists of a first grid electrode comprised of a plurality of electrically connected individual electrodes spaced apart from one another and the plurality of individual electrodes extending from the surface of the first charge collector,
 - wherein the spacing between two neighbouring individual electrodes is at most twice the electron diffusion length (EDL);
 - a photoactive layer positioned adjacent to the first charge collector and first substrate;
 - a second charge collector; and
 - an electrolyte interposed between the first charge collector and the photoactive layer, and the second charge collector.
2. The photovoltaic cell of claim 1,
 - wherein the photoactive layer is comprised of a nanoporous structure.
3. The photovoltaic cell of claim 2,
 - wherein the electrolyte penetrates into the pores of the photoactive layer.
4. The photovoltaic cell of any one of claims 1-3,
 - further comprising a second substrate, wherein the second charge collector is positioned adjacent to the second substrate.

5. The photovoltaic cell of any one of claims 1-3,
wherein the second charge collector comprises or consists of a second grid electrode comprised of a plurality of electrically connected individual electrodes spaced apart from one another and the plurality of individual electrodes extending from the surface of the second charge collector.
6. The photovoltaic cell of any one of claims 1-5,
wherein the plurality of individual electrodes of the first grid electrode and/or the plurality of individual electrodes of the second grid electrode extend substantially perpendicularly from the surface of the respective charge collector.
7. The photovoltaic cell of any one of claims 1-6,
wherein the plurality of individual electrodes of the first grid electrode extend from the surface of the first charge collector towards the second charge collector.
8. The photovoltaic cell of any one of claims 1-7,
wherein the plurality of individual electrodes of the second grid electrode extend from the surface of the second charge collector towards the first charge collector.
9. The photovoltaic cell of any one of claims 1-8,

wherein the thickness of each individual electrode of the first grid electrode measured in the longitudinal direction of the individual electrode is at least more than the difference between the thickness of the photoactive layer and the EDL.

10. The photovoltaic cell of any one of claims 5-9,

wherein the thickness of each individual electrode of the second grid electrode measured in the longitudinal direction of the individual electrode is at least more than half the spacing between two neighbouring individual electrodes of the second grid electrode.

11. The photovoltaic cell of any one of claims 1-10,

wherein the width of each individual electrode of the first grid electrode measured in the transverse direction of the individual electrode is about one-tenth of the spacing between two neighbouring individual electrodes of the first grid electrode.

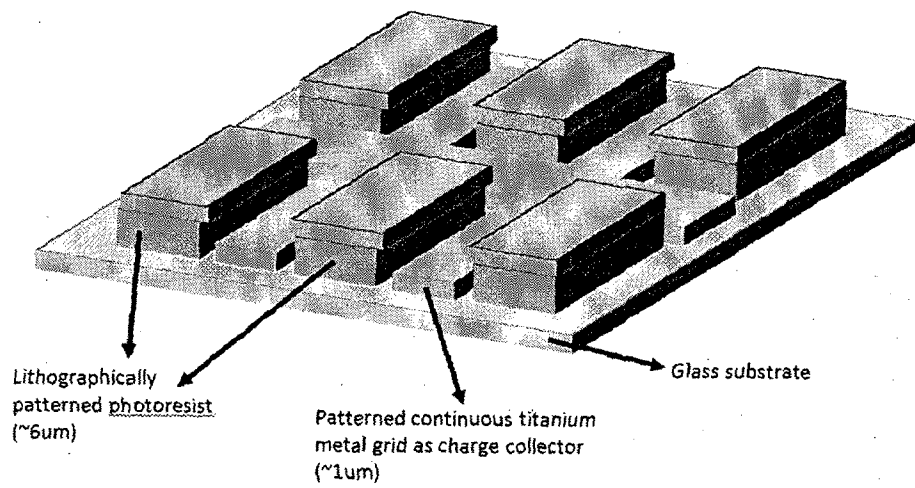
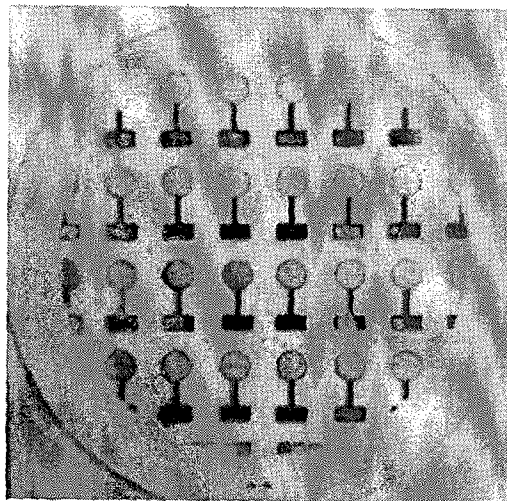
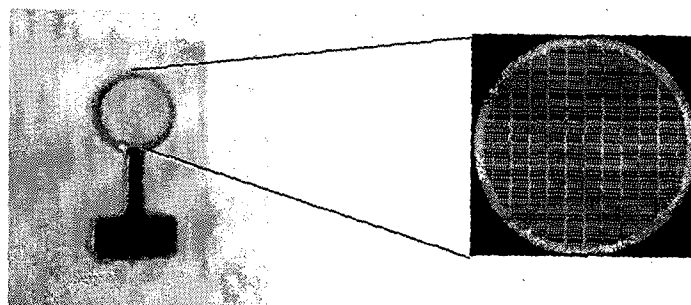
12. The photovoltaic cell of any one of claims 3-11,

wherein the width of each individual electrode of the second grid electrode measured in the transverse direction of the individual electrode is about one-tenth of the spacing between two neighbouring individual electrodes of the second grid electrode.

13. The photovoltaic cell of any one of claims 1-12,

wherein the photovoltaic cell is a dye-sensitized solar cell.

14. A charge collector comprising or consisting of a grid electrode comprised of a plurality of electrically connected individual electrodes spaced apart from one another and the plurality of individual electrodes extending from the surface of the charge collector, wherein the spacing between two neighbouring individual electrodes is at most twice the electron diffusion length (EDL).
15. The charge collector of claim 14,
wherein the plurality of individual electrodes of the grid electrode extend substantially perpendicularly from the surface of the charge collector.
16. The charge collector of claim 14 or 15,
wherein the width of each individual electrode of the grid electrode measured in the transverse direction of the individual electrode is about one-tenth of the spacing between two neighbouring individual electrodes of the grid electrode.
17. Use of a charge collector of any one of claims 14-16 in a photovoltaic cell, diode, transistor, electrochromic device, battery, supercapacitor, or waveguide.

**Figure 1A****Figure 1B****Figure 1C**

• 3D Grid Electrode DSSC fabrication

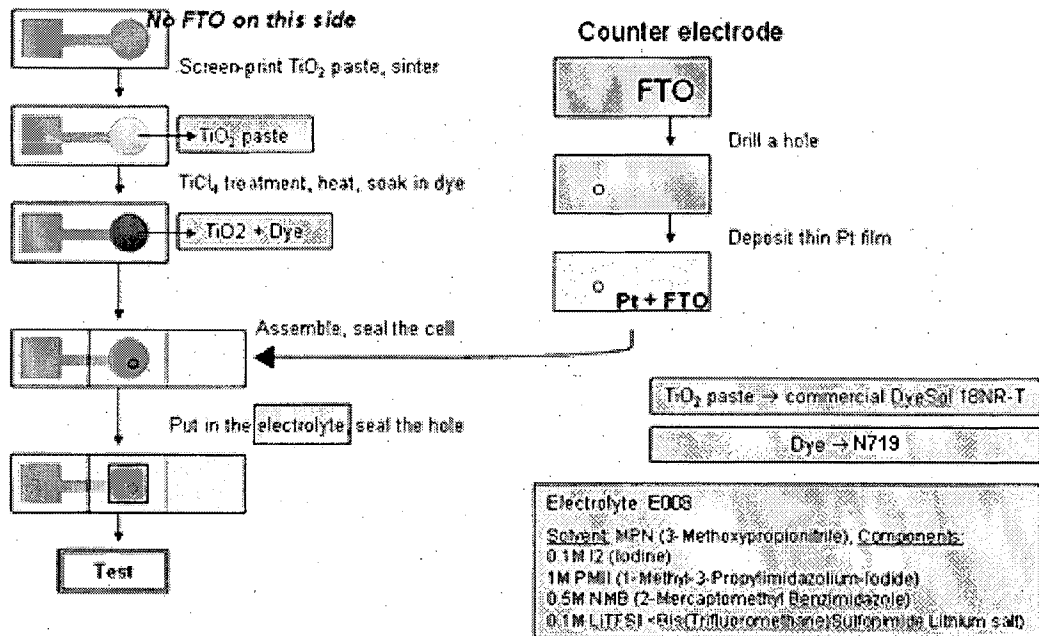
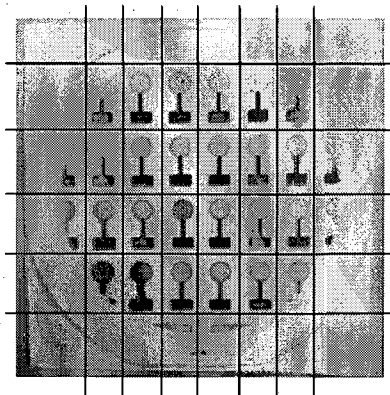
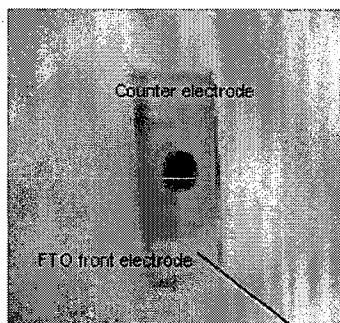
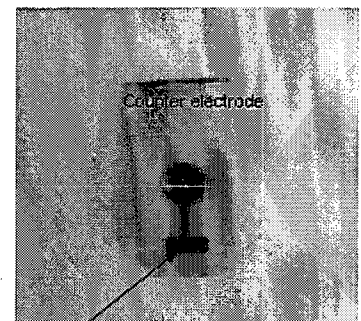


Figure 2

Standard cell



3D grid electrode cell



replaced by

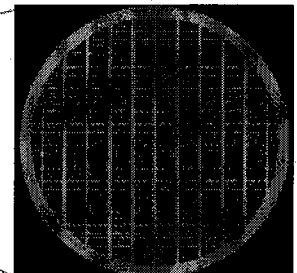
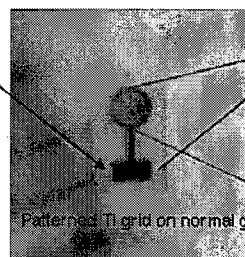


Figure 3

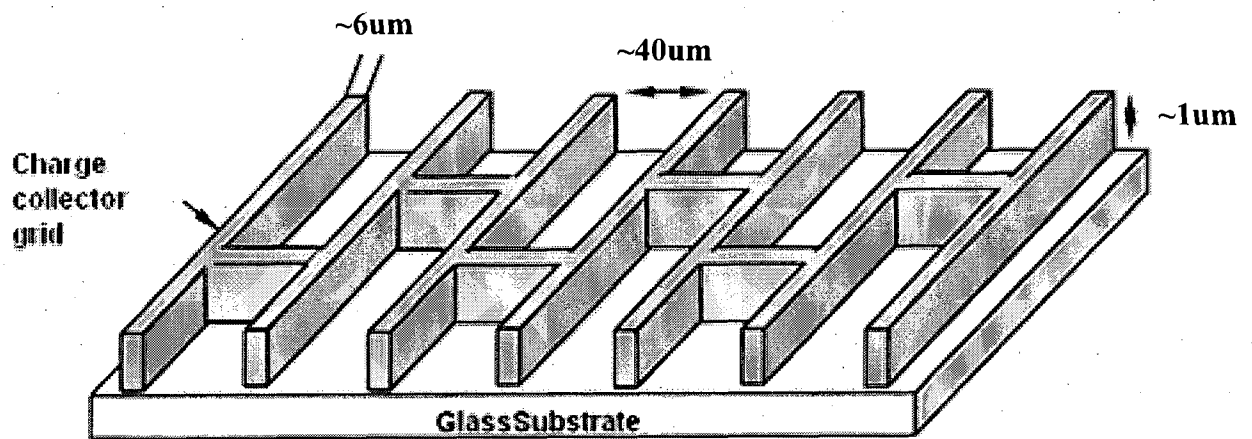


Figure 4

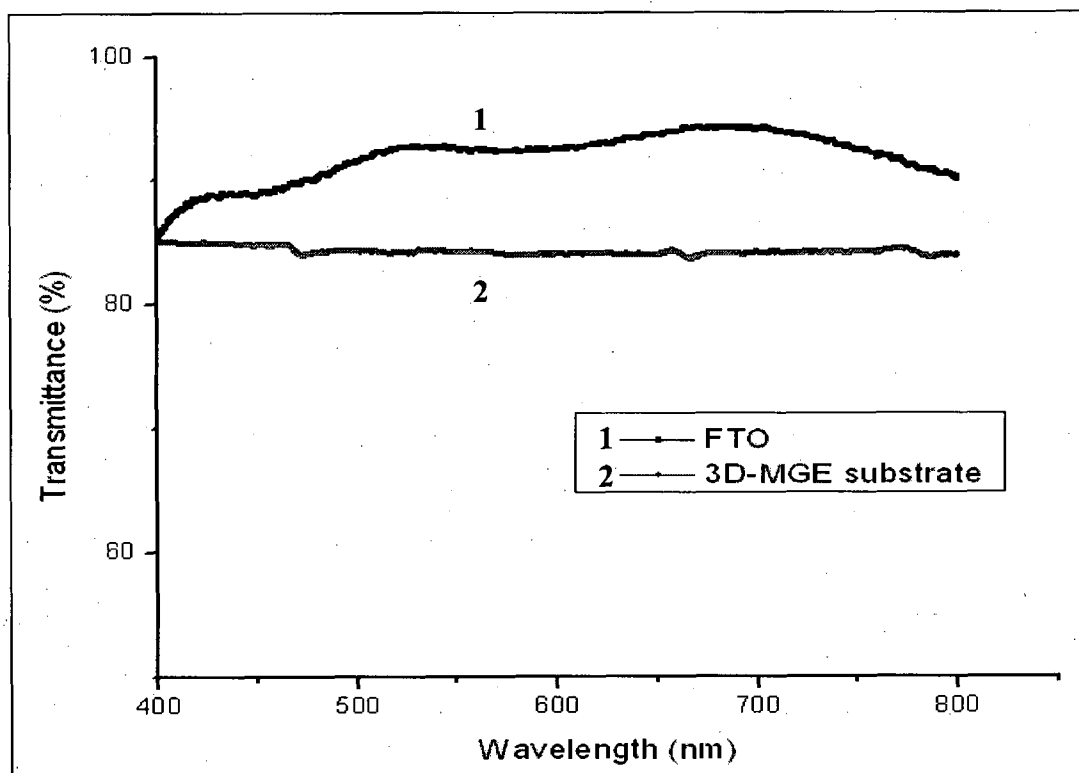


Figure 5

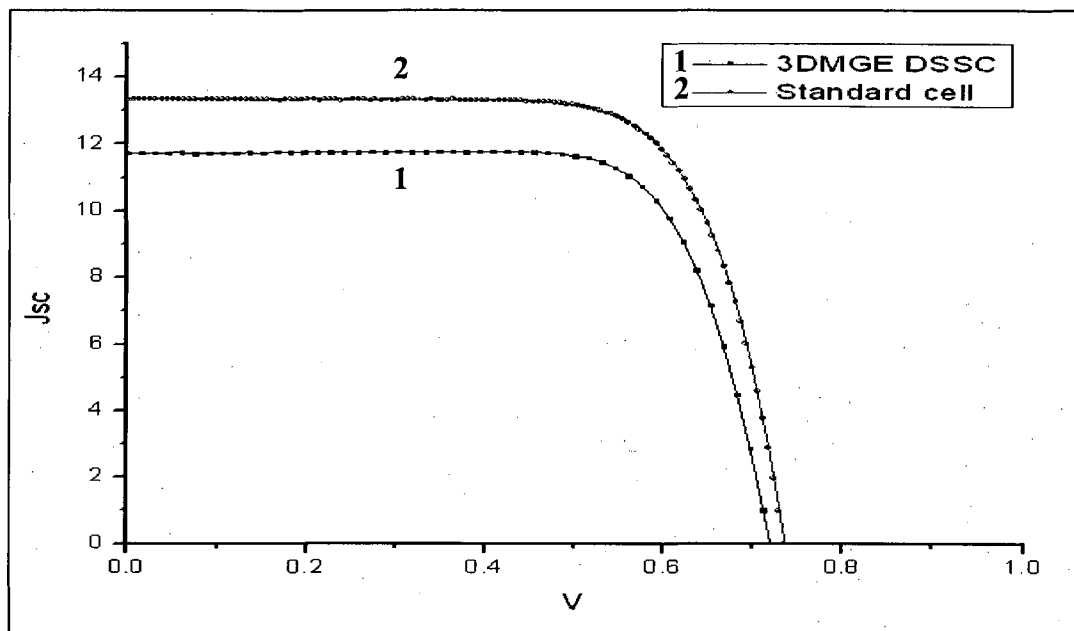


Figure 6

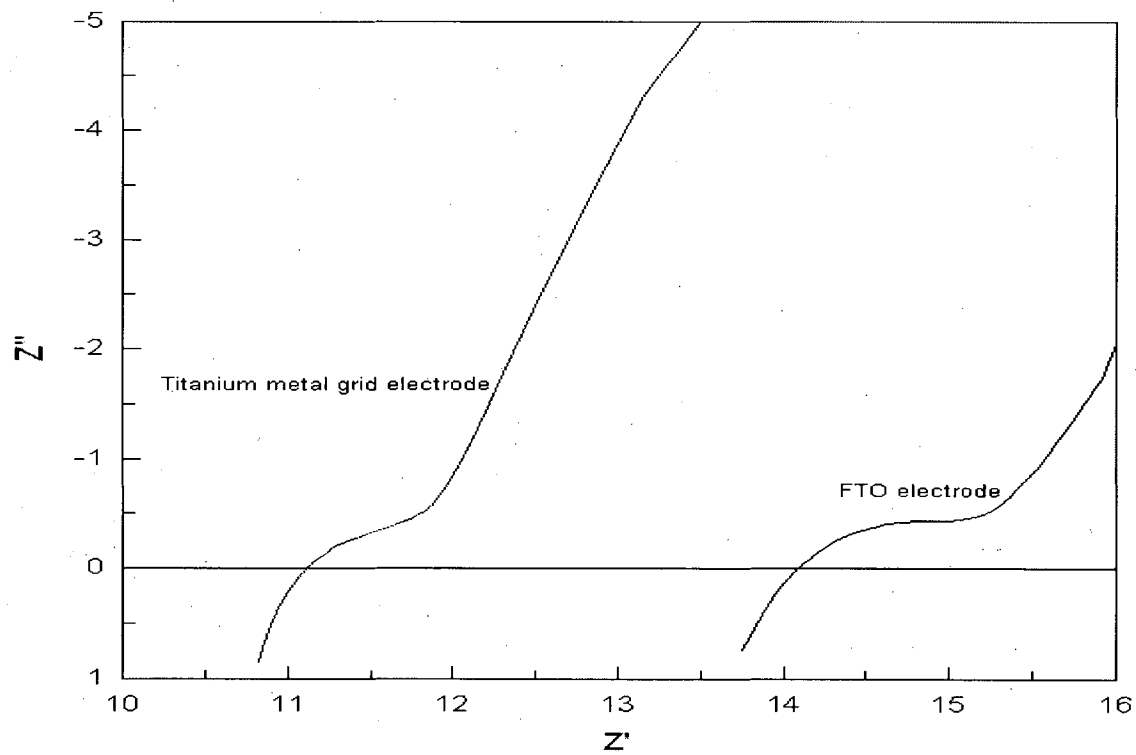


Figure 7

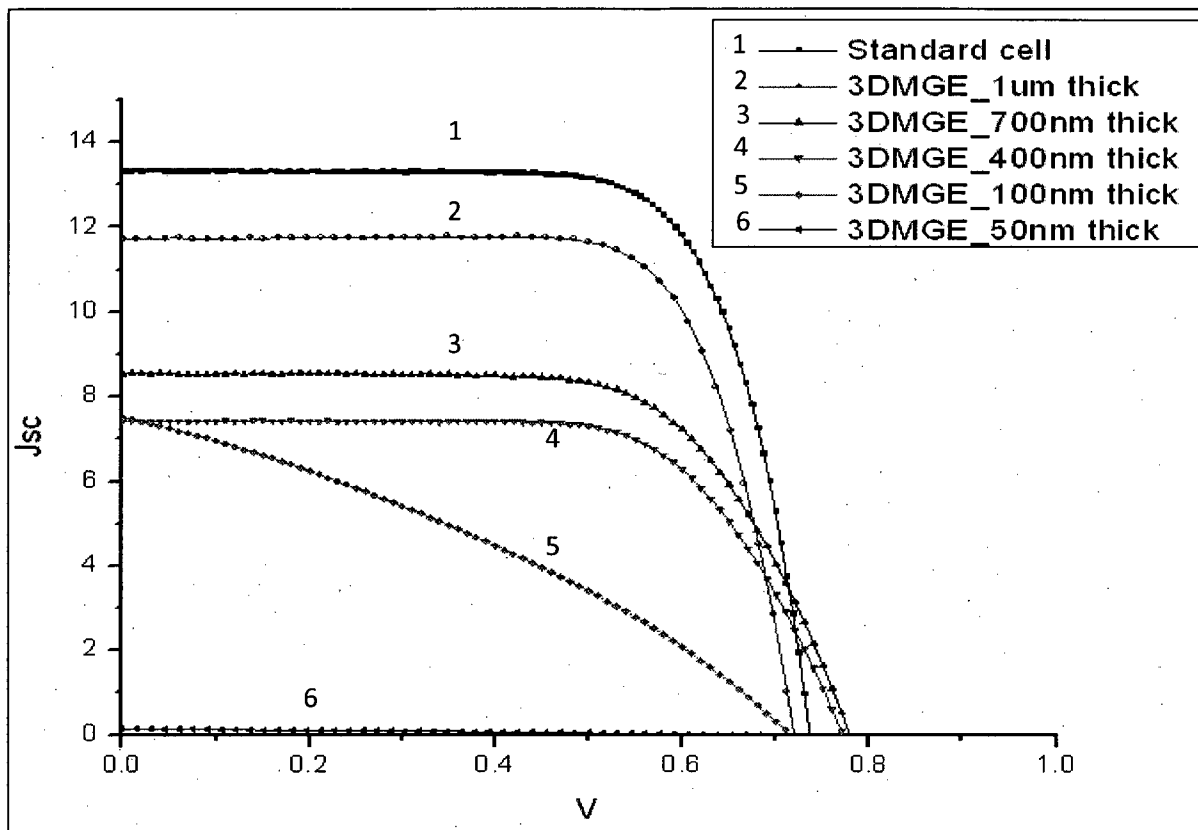


Figure 8

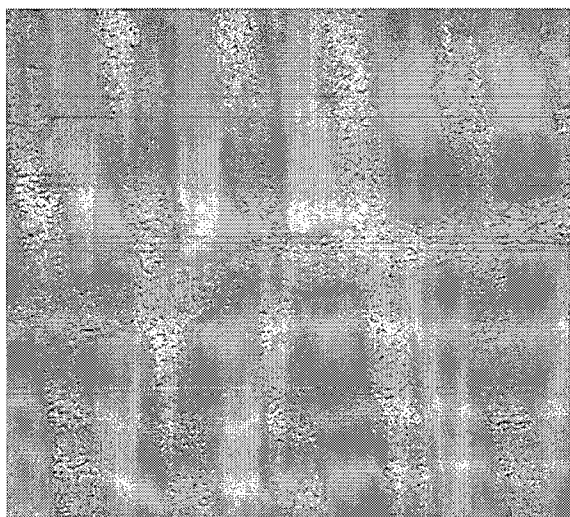


Figure 9A

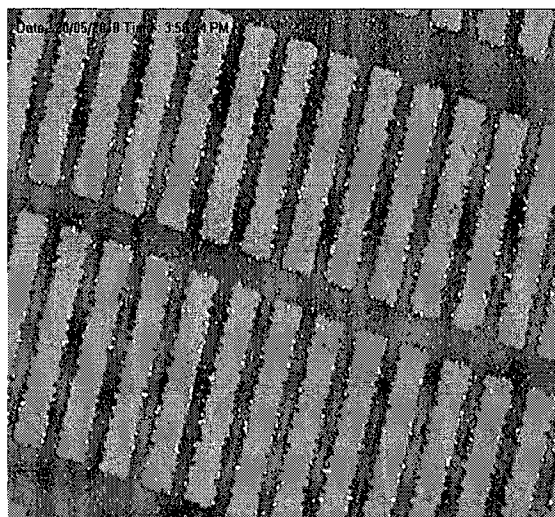


Figure 9B

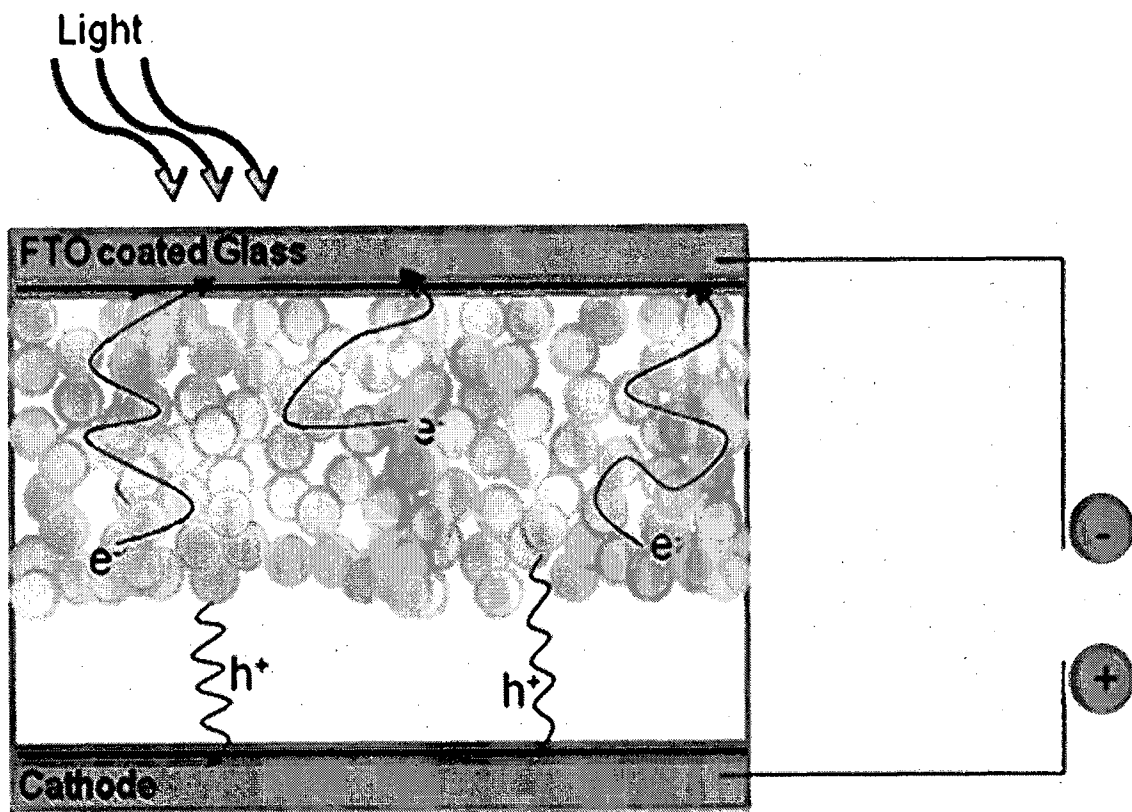


Figure 10 (prior art)

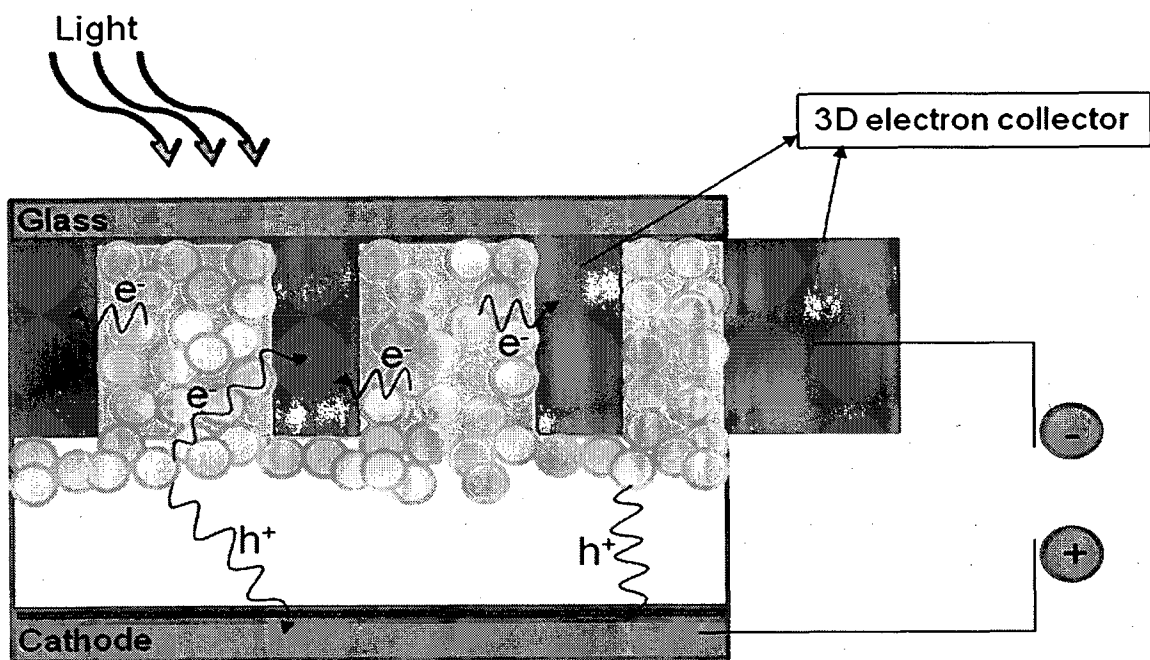


Figure 11

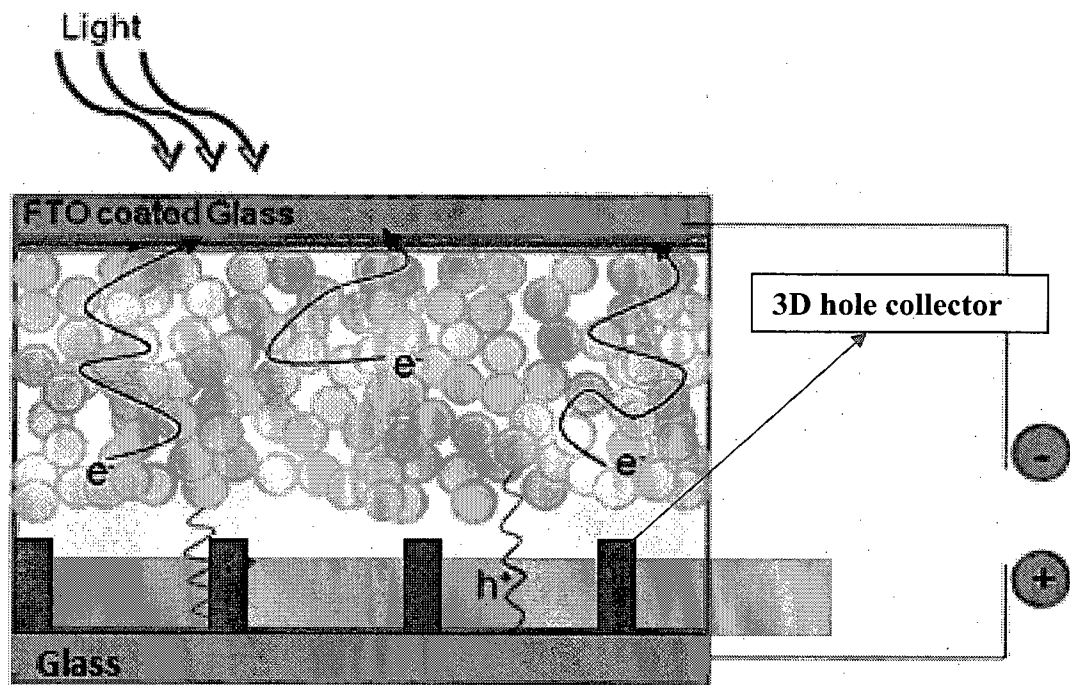


Figure 12

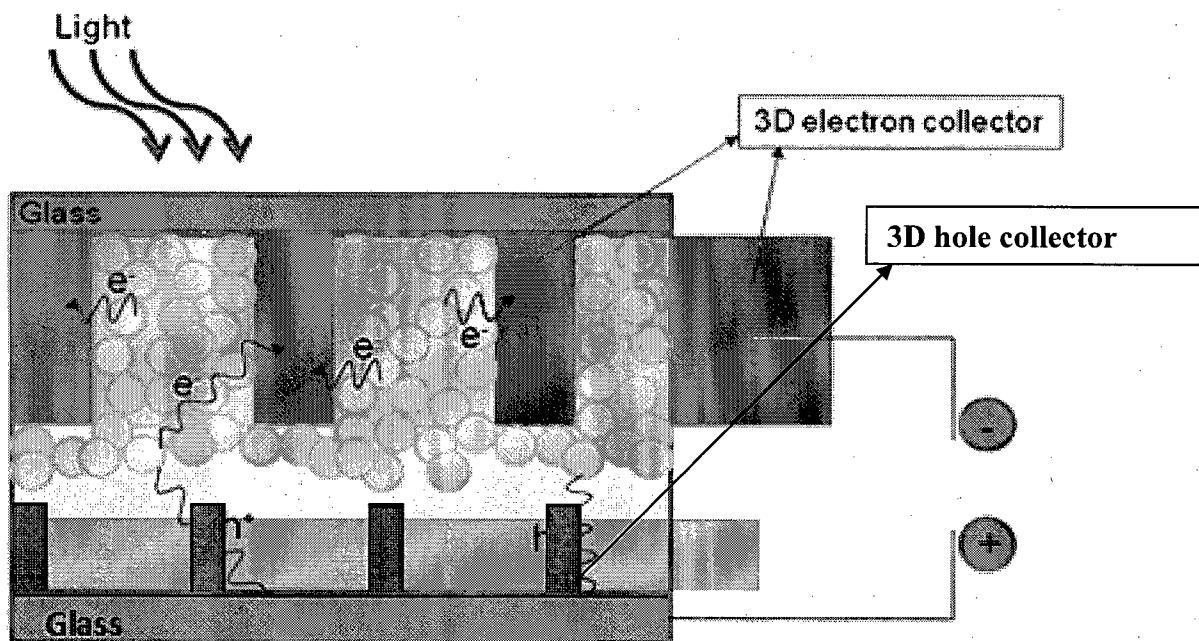
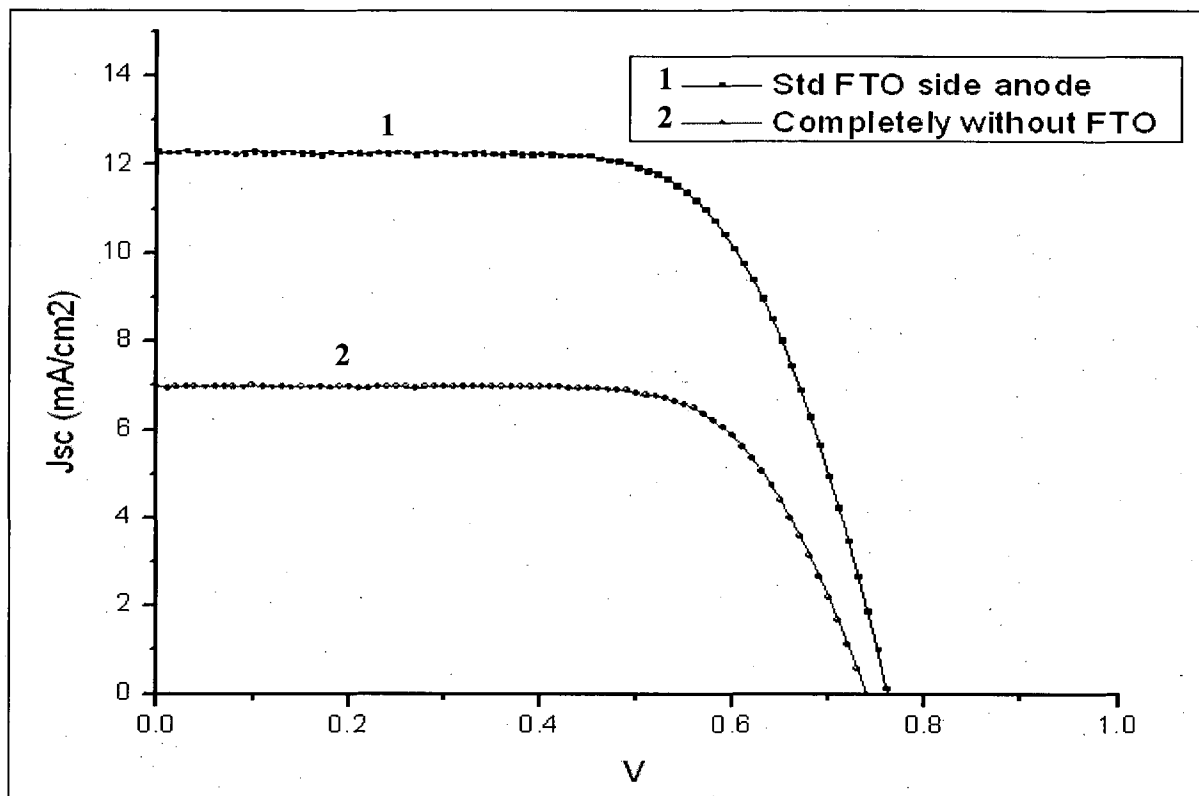
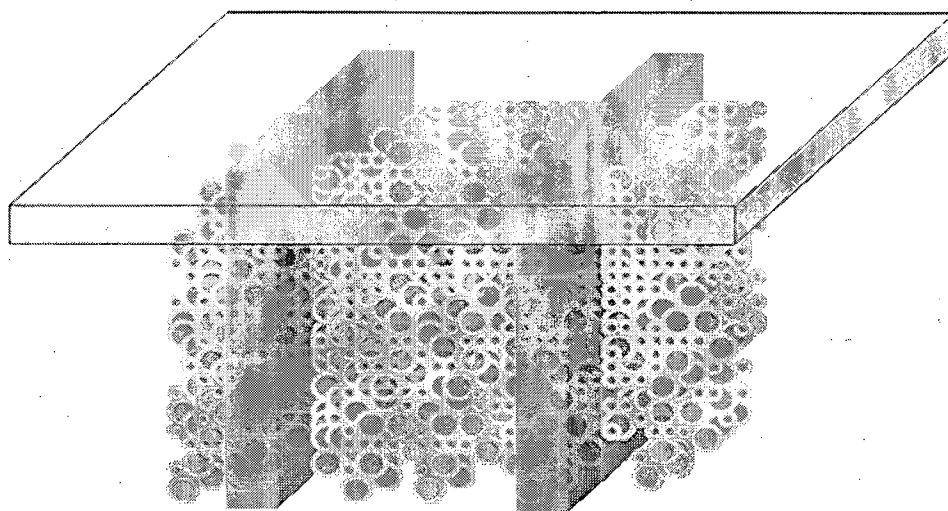


Figure 13

**Figure 14****Figure 15**

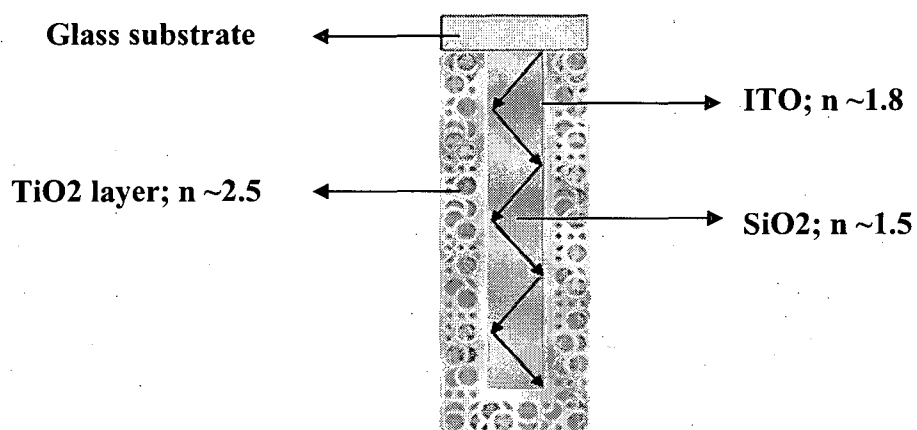


Figure 16

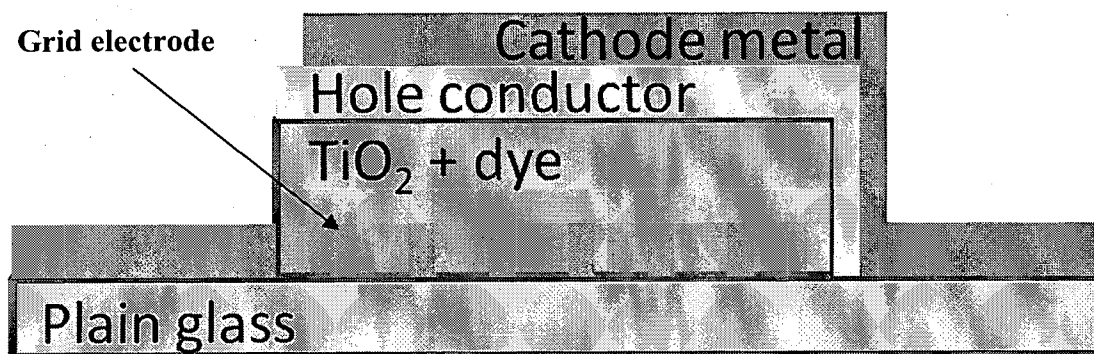


Figure 17

INTERNATIONAL SEARCH REPORT

International application No.

PCT/SG2012/000172

A. CLASSIFICATION OF SUBJECT MATTER

H01L 31/0224 (OCT 2005) H01L 31/02 (OCT 2005) H01L 31/04 (OCT 2005) H01G 9/048 (OCT 2005)
H01G 9/20 (OCT 2005) H01G 9/022 (OCT 2005)

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)

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)

WPI and EPODOC, keywords: grid, lattice, matrix, array; charge collector, electrode, anode, cathode; photovoltaic, dye sensitive solar cell, photoelectrochemical, hybrid solar cell, DSSC; multiple, plural; first, second, front, counter; spacing, separation, displacement; electron diffusion length, carrier diffusion length, micrometre; carrier life time; photo active, semiconductor; electrolyte; layer, film; pore, porous; extend, protrude; AND LIKE TERMS.

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
	Documents are listed in the continuation of Box C	

☒ Further documents are listed in the continuation of Box C☒ 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	"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
"E" earlier application or patent but published on or after the international filing date	"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
"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)	"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
"O" document referring to an oral disclosure, use, exhibition or other means	"&" document member of the same patent family
"P" document published prior to the international filing date but later than the priority date claimed	

Date of the actual completion of the international search
23 July 2012

Date of mailing of the international search report
23 July 2012

Name and mailing address of the ISA/AU

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INTERNATIONAL SEARCH REPORT C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		International application No. PCT/SG2012/000172
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2004/0112421 A1 (SPIVACK et al.) 17 June 2004 Abstract, paragraphs 0027 to 0029, 0031 to 0033, 0036 to 0049, figures 2 to 15	1-17

Form PCT/ISA/210 (fifth sheet) (July 2009)

INTERNATIONAL SEARCH REPORT Information on patent family members		International application No. PCT/SG2012/000172	
This Annex lists known patent family members relating to the patent documents cited in the above-mentioned international search report. The Australian Patent Office is in no way liable for these particulars which are merely given for the purpose of information.			
Patent Document/s Cited in Search Report		Patent Family Member/s	
Publication Number	Publication Date	Publication Number	Publication Date
US 2004/0112421 A1	17 Jun 2004	US 2004112421 A1	17 Jun 2004
		US 7145071 B2	05 Dec 2006
End of Annex			