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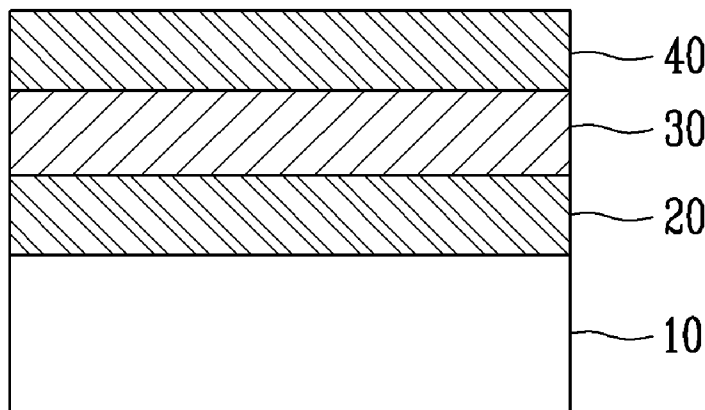
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(54) Title: TRANSPARENT CONDUCTIVE FILM AND METHOD FOR PREPARING THE SAME

Fig. 1



(57) Abstract: A transparent conductive layer and method of fabricating the same are provided. The transparent conductive layer has a multilayer structure in which transparent oxide layers and metal layers are alternately stacked, wherein the transparent oxide layer is an indium-free oxide layer having zinc oxide (ZnO) as a main component, and the metal layer includes Ag as a main component. The transparent conductive layer readily realizes high transparency having a light transmittance of 80% or higher in a visible-ray region, and a low resistance having a sheet resistance of 5 Ohm/square, so that the layer has numerous applications including a display field.

Description

TRANSPARENT CONDUCTIVE FILM AND METHOD FOR PREPARING THE SAME

Technical Field

- [1] The present invention relates to a transparent conductive layer and a method of fabricating the same. More particularly, the present invention relates to a transparent conductive layer having a multilayer structure in which transparent oxide layers that do not include indium and metal layers are alternately stacked, and having a light transmittance of 80% or higher in a visible-ray region and a low sheet resistance of 5 Ohm/square or lower, and a method of fabricating the same.
- [2] This work was supported by the IT R&D program of MIC/IITA, [2006-S-079-02, Smart Window Using Transparent Electronic Device].

Background Art

- [3] Recent developments in display industries have led to various needs for displays. Accordingly, demand for a transparent conductive layer in a visible-ray region is gradually increasing. Indium tin oxide (ITO) is a representative material that has been used for a transparent conductive layer.
- [4] Besides indium tin oxide, which is known as and is a representative material used for the transparent conductive layer, a material in which zinc oxide (ZnO) is used as a main component and a trivalent ion such as indium is added is used for the transparent conductive layer. While such materials exhibit excellent transparency, they are inferior to metals in electrical conductivity.
- [5] In view of this drawback, a transparent conductive layer having a structure of an oxide layer/a metal layer/an oxide layer has been developed. The oxide layer can refer to a transparent oxide thin film including ZnO together with In and Zn, and the metal layer can refer to a metal thin film whose main component is Ag. In such a structure, the transparency can be maintained, and simultaneously, electrical conductivity can be improved to nearly that of a metal.
- [6] However, a sudden increased demand in ITO has caused a price of indium to rise rapidly. During current research into a transparent conductive layer having a structure of an oxide layer/a metal layer/an oxide layer without including indium, the following has been observed. When ZnO is used as a main component of the oxide layer, and the thickness or impurity type of the oxide layer and the metal layer is controlled, a transparent conductive layer having excellent transparency and low resistance can be fabricated, and thus the present invention was completed.

Disclosure of Invention

Technical Problem

- [7] The present invention is directed to a transparent conductive layer that does not include indium, and has low resistance and high transmittance.
- [8] The present invention is also directed to a method of fabricating a transparent conductive layer that does not include indium, and has low resistance and high transmittance.

Technical Solution

- [9] One aspect of the present invention provides a transparent conductive layer having a multilayer structure in which transparent oxide layers and metal layers are alternately stacked, wherein the transparent oxide layer is an indium-free oxide layer having zinc oxide (ZnO) as a main component, and the metal layer includes Ag.
- [10] The transparent oxide layers may be formed of a ZnO layer that does not include impurities and/or may be formed of a ZnO layer that includes one or more elements selected from the group consisting of Al, Sn, Mg and Cd as an impurity.
- [11] The metal layers may be a single layer including Ag or an Ag-based alloy or may be a multilayer including a layer formed of Ag or an Ag-based alloy and a layer formed of a metal other than Ag.
- [12] The thickness of the transparent oxide layer may be selected within a range of 30 nm to 80 nm, and the thickness of the metal layer may be selected within a range of 5 nm to 20 nm.
- [13] The transparent conductive layer may further include a buffer layer formed between the metal layer and the transparent oxide layer formed on the metal layer, and formed of a conductive oxide. Further, the buffer layer may be formed to a thickness of 1 nm to 3 nm.
- [14] Another aspect of the present invention provides a method of fabricating a transparent conductive layer, including: depositing a first indium-free oxide layer whose main component is zinc oxide (ZnO) on a substrate; depositing a metal layer including Ag on the first indium-free oxide layer; and depositing a second indium-free oxide layer whose main component is ZnO on the metal layer, wherein the depositing of the first indium-free oxide layer and the depositing of the metal layer are repeated one or more times.
- [15] The first and second indium-free oxide layers may be deposited with ZnO that does not include impurities or may be deposited with ZnO including one or more elements selected from the group consisting of Al, Sn, Mg and Cd as an impurity.
- [16] The metal layers may be deposited with Ag or an Ag-based alloy or may be deposited with Ag or an Ag-based alloy, and then deposited with a metal other than Ag.

- [17] The first and second indium-free oxide layers may be respectively deposited to a thickness of 30 nm to 80 nm, and the metal layers may be deposited to a thickness of 5 nm to 20 nm.
- [18] Depositing a buffer layer formed of a conductive oxide to a thickness of 1 nm to 3 nm without oxygen supply may be further included during the depositing of the metal layer and the second indium-free oxide layer.
- [19] The deposition may be performed using a physical deposition method, a chemical deposition method, or a combination thereof.
- [20] The transparent conductive layer may be applied to an interconnection for a liquid crystal display (LCD), or a gate electrode, a source electrode, or a drain electrode of a thin film transistor.

Advantageous Effects

- [21] First, a transparent conductive layer according to the present invention readily realizes low resistance having a sheet resistance of 5 Ohm/square or lower without including indium and high transparency having a transmittance of 80% or higher in a visible-ray region, so that the layer may be applied in a display field in which transparency and large-scale area are required. Further, a low resistance transparent electrode having different work functions may be fabricated, so that the transparent conductive layer can be applied in various fields such as an organic light emitting diode (OLED), an inorganic light emitting diode (LED), a photovoltaic cell, etc.
- [22] Second, the transparent conductive layer according to the present invention does not use indium, which is scarce as a rare metal, so that the layer is more economical.
- [23] Third, the transparent conductive layer according to the present invention may be formed at a low temperature, so that it may be formed on both a glass substrate and a plastic substrate.
- [24] Fourth, in the transparent conductive layer according to the present invention an etch rate can be controlled to enable the fine patterning. Moreover, in the transparent conductive layer a work function can be controlled, so that the layer may be applied to an oxide conductor and a semiconductor that have various work functions.

Brief Description of Drawings

- [25] FIG. 1 is a cross-sectional view of a transparent conductive layer according to an exemplary embodiment of the present invention.
- [26] FIG. 2 is a cross-sectional view of a transparent conductive layer according to another exemplary embodiment of the present invention.
- [27] FIG. 3 is a cross-sectional view of a transparent conductive layer according to still another exemplary embodiment of the present invention.
- [28] FIG. 4 is a flowchart illustrating a method of fabricating a transparent conductive

layer according to an exemplary embodiment of the present invention.

[29] FIG. 5 is a graph of sheet resistance vs. thickness of a metal layer in a transparent conductive layer according to an exemplary embodiment of the present invention.

[30] FIG. 6 is a graph of transmittance vs. thickness of a metal layer in a transparent conductive layer according to an exemplary embodiment of the present invention.

[31] FIG. 7 is a graph of transmittance vs. thickness of an oxide layer in a transparent conductive layer according to an exemplary embodiment of the present invention.

[32] FIG. 8 is a graph of sheet resistance vs. thickness of an oxide layer in a transparent conductive layer according to an exemplary embodiment of the present invention.

Mode for the Invention

[33] While the invention is susceptible to various modifications and alternative forms, specific embodiments thereof are shown by way of example in the drawings and will herein be described in detail. It should be understood, however, that the drawings and detailed description thereto are not intended to limit the invention to the particular form disclosed, but on the contrary, the intention is to cover all modifications, equivalents and alternatives falling within the spirit and scope of the present invention as defined by the appended claims. The presently described embodiments will be best understood by reference to the drawings, wherein like parts are designated by like numerals throughout.

[34] Unless otherwise defined, all terms including technical and scientific terms used herein have the same meaning as commonly understood by one of ordinary skill in the art to which example embodiments belong. It will be further understood that terms, including those defined in commonly used dictionaries, should be interpreted as having a meaning that is consistent with their meaning in the context of the relevant art and will not be interpreted in an idealized or overly formal sense unless expressly so defined herein.

[35] The present invention provides a transparent conductive layer having a multilayer structure in which transparent oxide layers and metal layers are alternately stacked. The transparent oxide layer is an indium-free oxide layer, and the metal layer includes Ag.

[36] Exemplary embodiments of the present invention will be described in detail with reference to the accompanying drawings.

[37] FIG. 1 is a cross-sectional view of a transparent conductive layer according to an exemplary embodiment of the present invention, FIG. 2 is a cross-sectional view of a transparent conductive layer according to another exemplary embodiment of the present invention, and FIG. 3 is a cross-sectional view of a transparent conductive layer according to still another exemplary embodiment of the present invention.

- [38] A transparent conductive layer according to the present invention has a structure of a substrate 10, an indium-free oxide layer 20, a metal layer 30, and an indium-free oxide layer 40 as illustrated in FIG. 1. Alternatively, as illustrated in FIG. 2, the transparent conductive layer may have a structure of a substrate 10, an indium-free oxide layer 20, a metal layer 30, an indium-free oxide layer 40, a metal layer 50, and an indium-free oxide layer 60. Further, as illustrated in FIG. 3, the transparent conductive layer may have a structure of a substrate 10, an indium-free oxide layer 20, a metal layer 30, a buffer layer a, and an indium-free oxide layer 40.
- [39] The substrate 10 may be formed of a silicon wafer, glass or plastic.
- [40] Moreover, the indium-free oxide layers 20, 40 and 60 may each be formed of zinc oxide (ZnO) that does not include impurities, or may be formed of an oxide whose main component is ZnO together with one or more elements selected from the group consisting of Al, Sn, Mg and Cd as an impurity.
- [41] Further, thicknesses of the indium-free oxide layers 20, 40 and 60 may be selected within a range of 30 nm to 80 nm, and more preferably, within a range of 40 nm to 50 nm.
- [42] The metal layers 30 and 50 may be a single layer including Ag or an Ag-based alloy, or may be a multilayer including a layer formed of Ag or an Ag-based alloy and a layer formed of a metal other than Ag.
- [43] Further, thicknesses of the metal layers 30 and 50 may be selected within a range of 5 nm to 20 nm, and more preferably, within a range of 10 nm to 15 nm.
- [44] There is a trade-off between the transparency and electrical conductivity that are required characteristics of the transparent conductive layer according to the present invention. Therefore, it is important to optimize the characteristics.
- [45] The indium-free oxide layers 20, 40 and 60 play a decisive role in the transparency, and play a secondary role in the electrical conductivity. Moreover, the metal layers 30 and 50 play a major role in the electrical conductivity. Accordingly, the optimal transparency and electrical conductivity can be ensured by controlling the thicknesses of the indium-free oxide layers and the metal layers.
- [46] ZnO constituting the indium-free oxide layers 20, 40 and 60 has a low electric resistance, its thin film formation is performed at a low temperature, and it may replace indium, an expensive and rare metal, so that it may be more economical. In contrast, ZnO is inferior to indium-tin-oxide (ITO) in terms of electrical conductivity, and an oxide layer formed of ZnO is vulnerable to an etchant.
- [47] Therefore, when the indium-free oxide layers 20, 40 and 60 are formed of ZnO as a main component and Al is added as impurities, electrical conductivity of the layers can be enhanced.
- [48] Furthermore, when the indium-free oxide layers 20, 40 and 60 are formed of ZnO as

a main component, and Sn, a tetravalent element, is added as impurities, the layers may be adjusted to have enhanced electrical conductivity, and simultaneously, to have durability suitable for an etching process.

[49] In addition, when the indium-free oxide layers 20, 40 and 60 are formed of ZnO as a main component, and Mg and Cd are added as impurities, a work function can be adjusted. For example, when Mg is added to ZnO, a bandgap is increased, but a work function is decreased. Therefore, the work function of the transparent conductive layer is adjusted, so that applicable fields can be broadened. For example, a transparent conductive layer having a low work function may be applied to an anode or cathode in an organic light emitting diode or inorganic light emitting diode. Furthermore, the work function of the transparent conductive layer is adjusted to facilitate the constitution of the ohmic or schottky contact when the layer is in contact with other metals or a semiconductor.

[50] Also, the indium-free oxide layer whose main component is ZnO in the transparent conductive layer according to the present invention is etched faster than an indium-tin oxide layer. Accordingly, since there is a slight difference in etch rate between the metal layers containing Ag, the indium-free oxide may be more easily etched than the indium-tin oxide to enable fine patterning by dry etching and wet etching.

[51] Referring to FIG. 3, the transparent conductive layer according to the present invention includes the buffer layer a between the metal layer 30 and the indium-free oxide layer 40. The buffer layer is introduced to prevent exposure of the metal layer 30 to oxygen plasma. Like the indium-free oxide layer 40, the buffer layer a may be formed of a conductive oxide, and may be deposited to be formed without oxygen supply when it is deposited. In general, when an oxide is deposited without oxygen supply, a thin film should be formed using only oxygen of the target oxide. As a result, many crystal defects are formed in the thin film, and thus the thin film has deteriorated film quality and reduced transparency. Therefore, the thickness of a buffer layer that is formed while oxygen is not supplied should be minimized. The thickness that minimizes deteriorated quality and a reduction in transparency of the thin film may be within a range of 1 nm to 3 nm.

[52] FIG. 4 is a flowchart illustrating a method of fabricating a transparent conductive layer according to an exemplary embodiment of the present invention.

[53] Referring to FIG. 4, the method of fabricating a transparent conductive layer according to the present invention includes: (a) depositing a first indium-free oxide layer on a substrate; (b) depositing a metal layer including Ag on the first indium-free oxide layer; and (c) depositing a second indium-free oxide layer on the metal layer, wherein depositing the metal layer and depositing the second indium-free oxide layer may be repeated one or more times.

- [54] During the deposition of the indium-free oxide layer of (a), the layer may be deposited with only ZnO or using ZnO together with one or more elements selected from the group consisting of Al, Sn, Mg and Cd as an impurity, and may be deposited to a thickness of about 30 nm to 80 nm.
- [55] During the deposition of the metal layer of (b), the layer may be deposited to have a single layer using Ag or an Ag-based alloy or a multilayer including a layer formed of Ag or an Ag-based alloy and a layer formed of a metal other than Ag, and may be deposited to a thickness of about 5 nm to 20 nm.
- [56] Before the deposition of the indium-free transparent oxide layer of (c), depositing a buffer layer may be further included in order to prevent the metal layer from being exposed to oxygen. The deposition of the buffer layer is performed using a conductive oxide material such as an indium-free oxide layer without oxygen supply. Therefore, the indium-free oxide layer may be deposited to form the buffer layer without oxygen supply, and afterwards, the indium-free oxide layer may be deposited with oxygen supplied. In this case, the buffer layer is formed without oxygen supply, and thus crystal defects of the thin layer may be formed. Therefore, the buffer layer may be deposited to a thickness of 1 nm to 3 nm.
- [57] As in the deposition of (a), the deposition of the indium-free oxide layer of (c) may be performed using only ZnO or using ZnO together with one or more elements selected from the group consisting of Al, Sn, Mg and Cd as an impurity, and the layer may be deposited to a thickness of about 30 nm to 80 nm.
- [58] Afterwards, the deposition of the metal layer of (b) and the deposition of the indium-free oxide layer of (c) may be repeated such that the total thickness of the transparent conductive layer does not exceed 200 nm.
- [59] The deposition of each layer may be performed using a physical deposition method including a sputtering method and a PLD method, a chemical deposition method including an atomic layer deposition (ALD), and a Metal Organic Chemical Vapor Deposition (MOCVD), or a combination thereof. The deposition may be performed at room temperature. After the deposition of each layer, an annealing process may be performed at a temperature of 300°C or lower, and the annealing process may further enhance sheet resistance and transmittance.
- [60] The transparent conductive layer according to the present invention may be used for a gate electrode, a source electrode, or a drain electrode of a thin film transistor as well as an interconnection for a liquid crystal device (LCD).
- [61] Generally, silicon has been used for a semiconductor layer of a thin film transistor. However, currently, various oxide semiconductors including ZnO, indium gallium zinc oxide (IGZO), etc. are being used to develop a transparent thin film transistor. Keeping up with such trends, the oxide layer of the transparent conductive layer according to

the present invention is similar to the semiconductor layer of the thin film transistor, so that it can contribute to fabrication of a transparent oxide thin film transistor having excellent characteristics.

[62] An organic light emitting diode has numerous applications such as a light with an appearance of wallpaper as well as a display. However, since currently used ITO has higher electrical resistance than a metal, and requires an annealing process performed at a temperature of about 200°C, selection of substrate materials is limited, and this poses an obstacle to a large-scale light.

[63] Accordingly, the transparent conductive layer according to the present invention in which an electrical resistance is lower than ITO, a thin film formation temperature is low to have wide selections of plastic substrates, and In is not included to be low in price of materials may be applied to an anode, a cathode or an interconnection of an organic light emitting diode.

[64] Moreover, the transparent conductive layer according to the present invention may be applied to an anode, a cathode or an interconnection of an inorganic light emitting diode, and may be used as an electrode of a photovoltaic cell.

[65]

[66] **First Example**

[67] In order to enable the serial deposition without being exposed to the air, a sputtering device to which a sputtering chamber including ZnO and a sputtering chamber including Ag were connected was used to deposit a first ZnO layer on a substrate to a thickness of 40 nm at room temperature under oxygen plasma, and an annealing process was performed on the deposited results. Afterwards, an Ag layer was deposited on the ZnO layer to a thickness of 10.0 nm at room temperature and an annealing process was performed on the deposited results. Then, a second ZnO layer was deposited on the Ag layer to a thickness of 40 nm at room temperature under oxygen plasma, and an annealing process was performed on the deposited results to fabricate a transparent conductive layer.

[68]

[69] **Second to Seventh Examples**

[70] Transparent conductive layers were fabricated in the same manner as the first example except for depositing Ag layers to thicknesses of 5.0 nm, 7.5 nm, 12.5 nm, 15.0 nm, 17.5 nm and 20.0 nm, respectively.

[71]

[72] **Eighth to Tenth Examples**

[73] Transparent conductive layers were fabricated in the same manner as the first example except for depositing first and second ZnO layers to thicknesses of 20 nm, 30 nm, and 50 nm, respectively.

- [74] Sheet resistance of the transparent conductive layers fabricated in the first to seventh examples was examined and the results thereof were shown in FIG. 5. Further, transmittance of the transparent conductive layers fabricated in the second to seventh examples was examined, and the results thereof were shown in FIG. 6.
- [75] Furthermore, transmittance and sheet resistance of the transparent conductive layers fabricated in the first, and eighth to tenth examples were examined, and the results thereof were shown in FIGS. 7 and 8.
- [76] Referring to FIGS. 5 and 6, it is observed that as the thickness of the metal layer is increased, the sheet resistance and the transmittance are decreased. Further, the metal layer having a thickness ranging from 5 nm to 20 nm may be used as a transparent conductive layer, and more preferably, the metal layer may have a thickness of about 10 nm.
- [77] In addition, referring to FIGS. 7 and 8, it is observed that the thickness of the oxide layer is proportional to the sheet resistance and the transmittance.
- [78] While the invention has been shown and described with reference to certain example embodiments thereof, it will be understood by those skilled in the art that various changes in form and details may be made therein without departing from the spirit and scope of the invention as defined by the appended claims.

Claims

- [1] A transparent conductive layer consisting of a multilayer structure in which transparent oxide layers and metal layers are alternately stacked, wherein each of the transparent oxide layers is an indium-free oxide layer having zinc oxide (ZnO) as a main component, and each of the metal layers includes Ag.
- [2] The transparent conductive layer according to claim 1, wherein the transparent oxide layer is a ZnO layer that does not include impurities.
- [3] The transparent conductive layer according to claim 1, wherein the transparent oxide layer is formed of a ZnO layer including one or more elements selected from the group consisting of Al, Sn, Mg and Cd as an impurity.
- [4] The transparent conductive layer according to claim 1, wherein the metal layer is a single layer including Ag or an Ag-based alloy.
- [5] The transparent conductive layer according to claim 1, wherein the metal layer is a multilayer including a layer formed of Ag or an Ag-based alloy and a layer formed of a metal other than Ag.
- [6] The transparent conductive layer according to claim 1, wherein the thickness of the transparent oxide layer is selected within a range of 30 nm to 80 nm, and the thickness of the metal layer is selected within a range of 5 nm to 20 nm.
- [7] The transparent conductive layer according to claim 1, further comprising a buffer layer that is formed between the metal layer and the transparent oxide layer formed on the metal layer and is formed of a conductive oxide.
- [8] The transparent conductive layer according to claim 7, wherein the thickness of the buffer layer is selected within a range of 1 nm to 3 nm.
- [9] A method of fabricating a transparent conductive layer, comprising:
depositing a first indium-free oxide layer on a substrate;
depositing a metal layer including Ag on the first indium-free oxide layer; and
depositing a second indium-free oxide layer on the metal layer,
wherein the depositing of the first indium-free oxide layer and the metal layer is repeated one or more times.
- [10] The method according to claim 9, wherein the indium-free oxide layer is deposited with ZnO that does not include impurities.
- [11] The method according to claim 9, wherein the indium-free oxide layer is deposited with ZnO including one or more elements selected from the group consisting of Al, Ga, Sn, Mg and Cd as an impurity.
- [12] The method according to claim 9, wherein the metal layer is deposited with Ag or an Ag-based alloy.
- [13] The method according to claim 9, wherein the metal layer is deposited with Ag

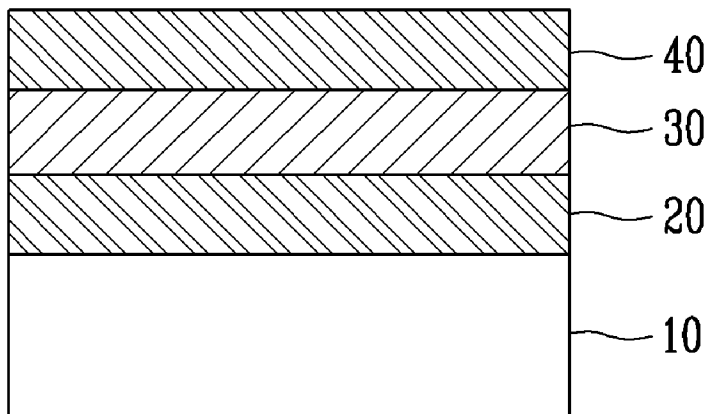
or an Ag-based alloy, and then is deposited with a metal other than Ag.

[14] The method according to claim 9, wherein the oxide layer is deposited to a thickness of 30 nm to 80 nm, and the metal layer is deposited to a thickness of 5 nm to 20 nm.

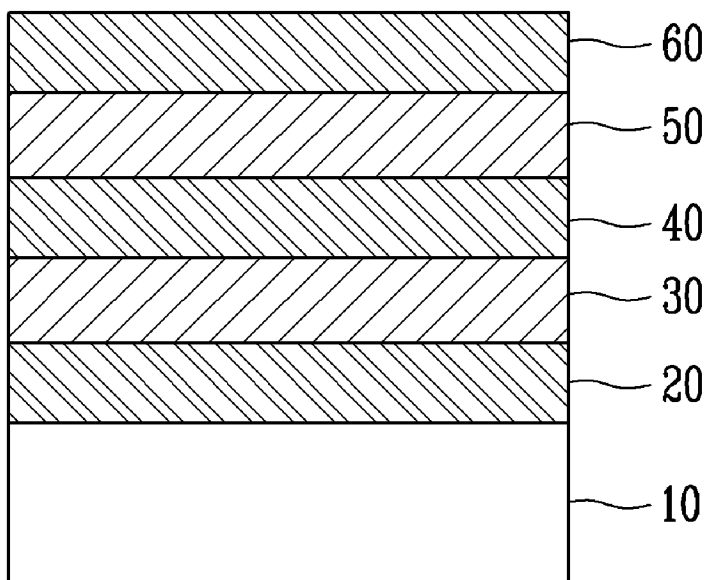
[15] The method according to claim 9, further comprising depositing a buffer layer formed of a conductive oxide between the metal layer and the second indium-free oxide layer to a thickness of 1 nm to 3 nm without oxygen supply.

[16] The method according to claim 9, wherein the deposition is performed using a physical deposition method, a chemical deposition method, or a combination thereof.

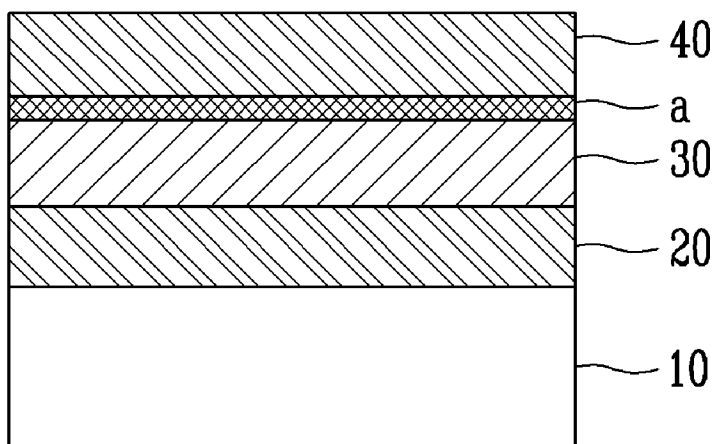
[Fig. 1]



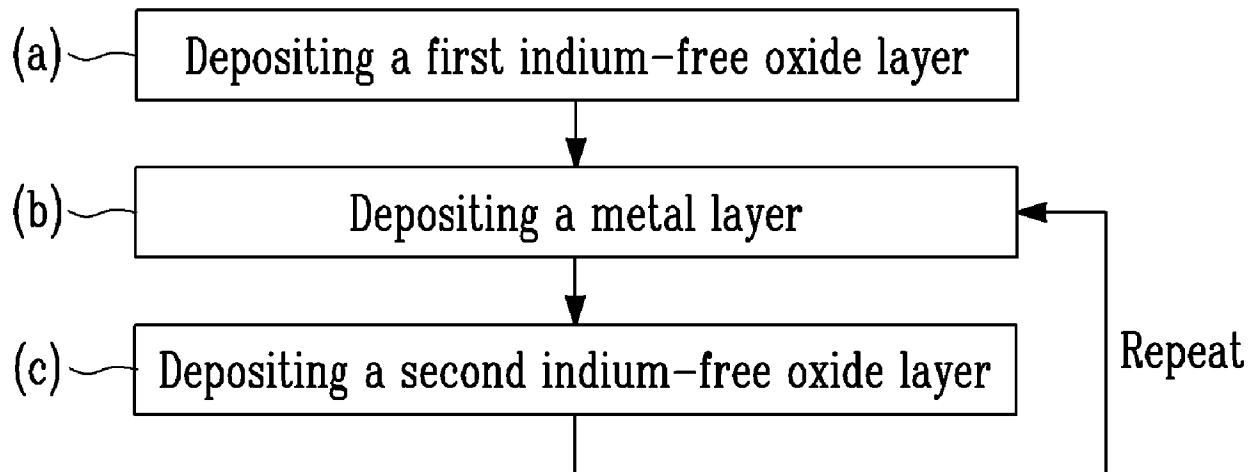
[Fig. 2]



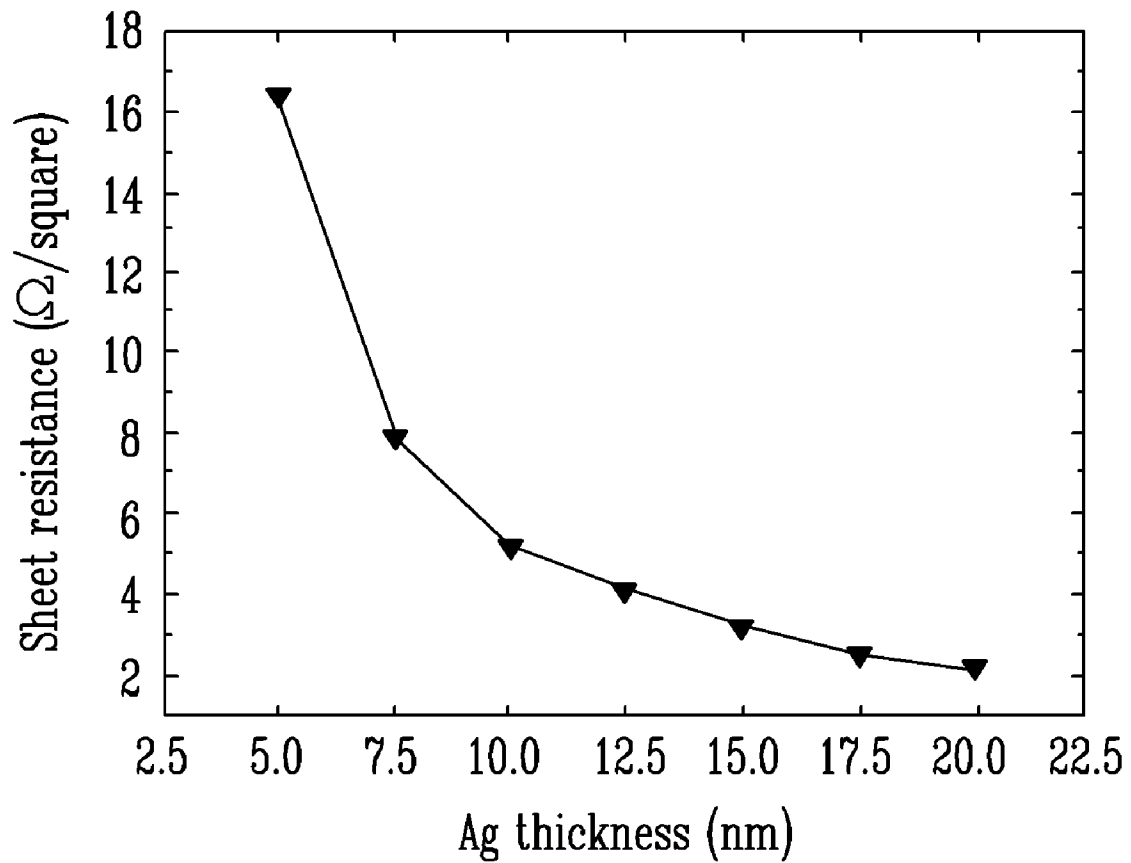
[Fig. 3]



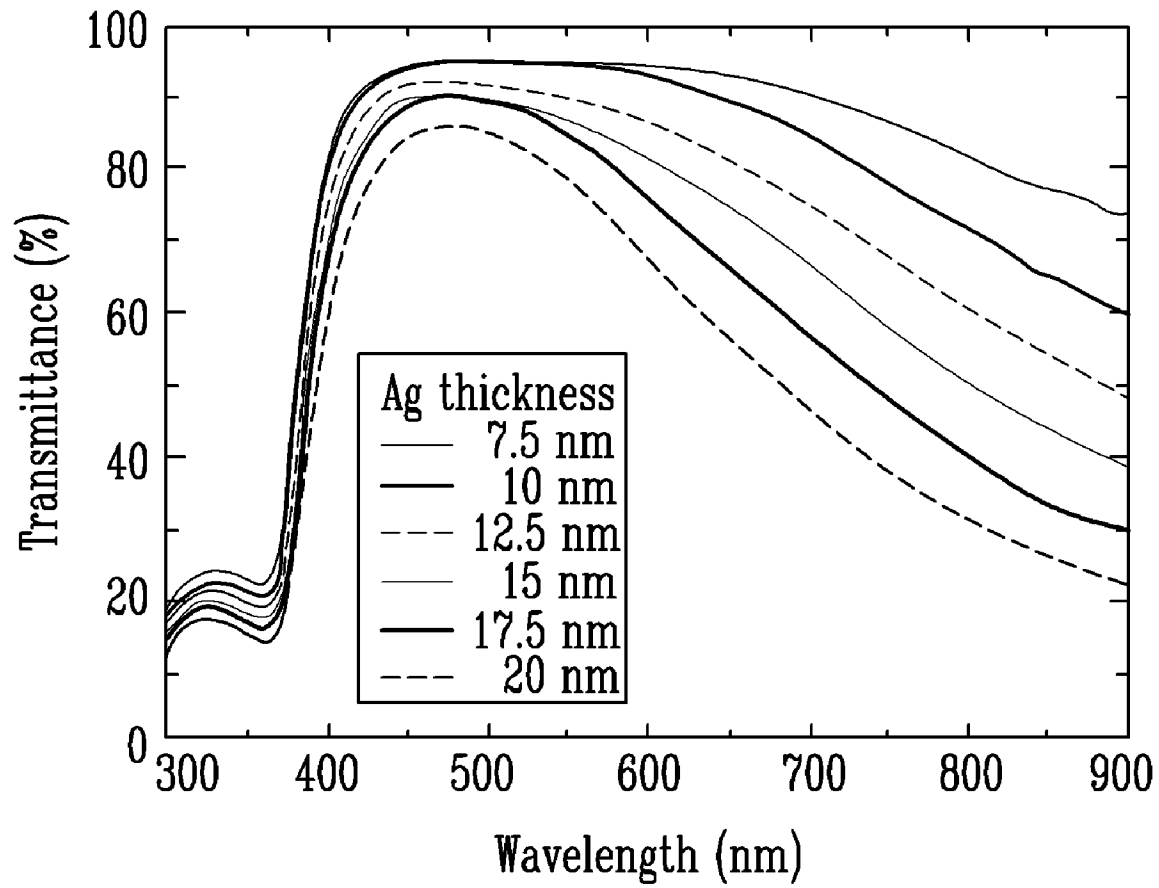
[Fig. 4]



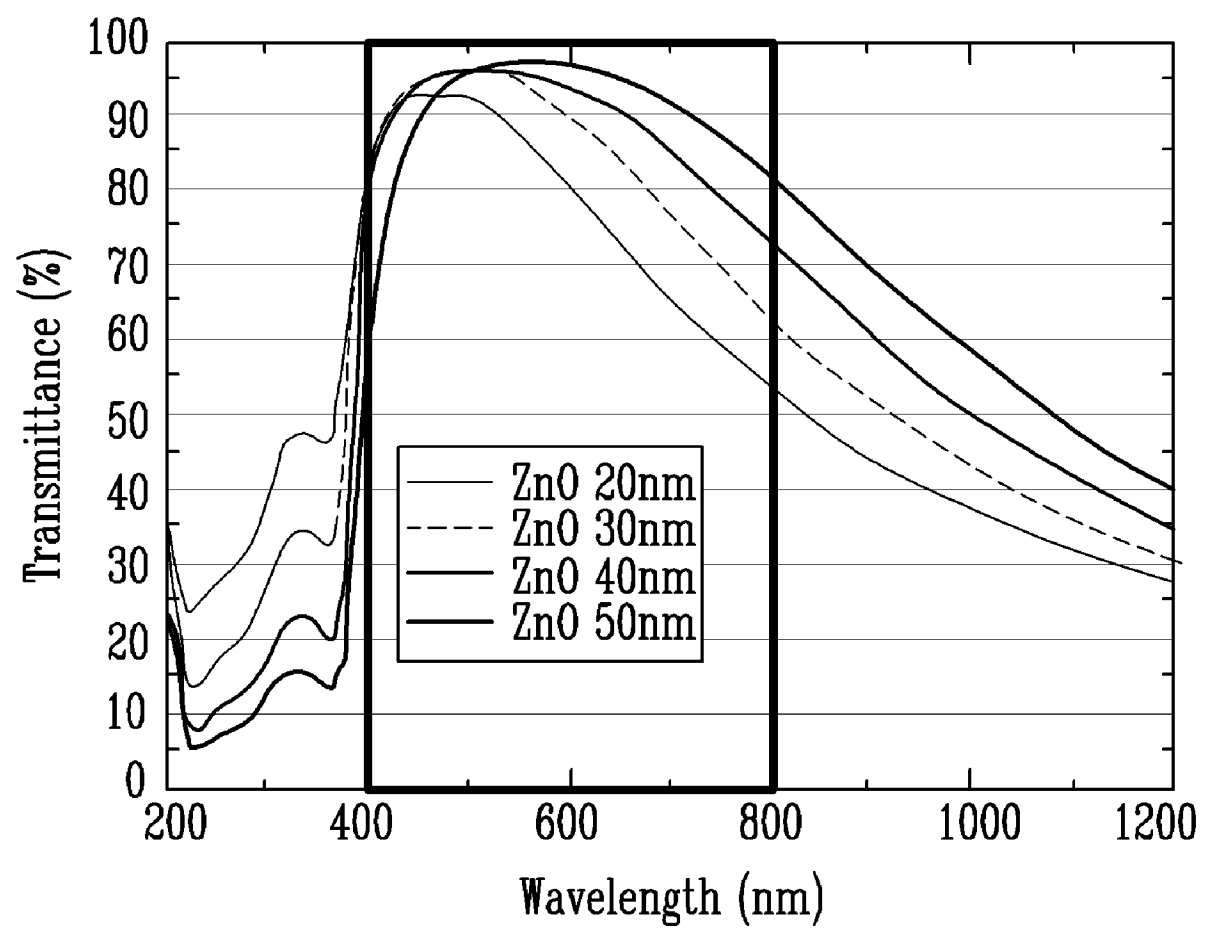
[Fig. 5]



[Fig. 6]



[Fig. 7]



[Fig. 8]

