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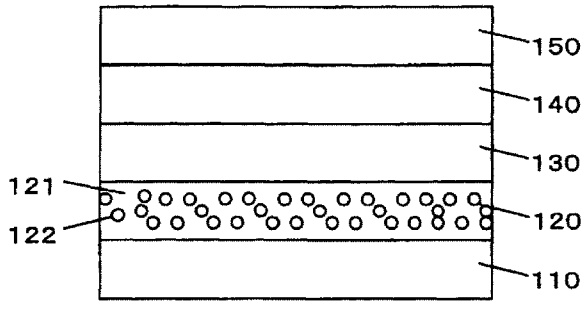
(54) **Title**: TRANSLUCENT CONDUCTIVE SUBSTRATE FOR AN ORGANIC LIGHT EMITTING DEVICE AND METHOD FOR ITS PRODUCTION

Fig. 1



(57) **Abstract**: The present invention relates to a process for producing a conductive translucent substrate for use in an organic light emitting diode, wherein a transparent electrode material is deposited on top of a scattering layer deposited on the transparent support, wherein the scattering layer comprises a base material having a first refractive index at a wavelength of emitted light of an organic LED element and a plurality of scattering materials provided in the inside of the base material and having a refractive index different from that of the base material, wherein the scattering material is deposited on the transparent support by deposition of a frit paste and heating the thus coated support to dry and fire the frit paste to form the scattering layer. As the electrode material use is made of F-doped SnO_2, which is deposited by deposition and decomposition of a precursor at a temperature of between 500 and 650°C followed by cooling of the thus obtained conductive substrate. The present invention relates to a conductive translucent substrate.

WO 2016/102401 A1

Translucent conductive substrate for an organic light emitting device and method for its production

The present invention relates to a translucent conductive substrate for an organic light emitting device according to the preamble of the first claim.

The present invention also relates to a method for producing a conductive substrate for an organic light emitting device. The present invention further relates to an organic light-emitting diode comprising such a substrate.

Electroluminescent devices, in particular those devices which include an active layer with one or more organic and/or organometallic compounds so that the device can emit light, are known for example as organic light-emitting diodes (OLED). Organic light-emitting diodes find application in panel display technologies, for example television sets, computer terminals; as lighting devices for example in architectural or decorative lighting devices; for signage and many other applications. In the afore-mentioned type of applications the use of white light emitting OLED's is often preferred.

Organic electroluminescent devices (OLEDs) generally comprise a substrate, a first electrode disposed over the substrate for supplying charge of a first polarity, a second electrode disposed for supplying charge of a second polarity opposite to said first polarity, an organic light-emitting layer disposed between both electrodes and an encapsulating material enclosing the whole device. The electrode adjacent the substrate may be the anode and the other electrode may be the cathode. Alternatively, the electrode adjacent the substrate may be the cathode and the other electrode may be the anode. In a bottom emitting device, the substrate and the electrode of the first polarity are transparent to allow light emitted by the organic light emitting layer to pass there through. In a top emitting device, the electrode of the second polarity and the encapsulating material are transparent so as to allow light emitted from the organic light-emitting layer to pass there through. In still another arrangement, both electrodes, the substrate and encapsulating material are

transparent, and light may be emitted through both the top and the bottom of the device. Variations of the above-described structures exist as well. Further layers may be provided between the electrodes and the organic light-emitting layer in order to aid charge injection and transport. Examples of such further layers area hole transporting layer and/or an electron transporting layer, possibly further doped using oxidizing or reducing agents; charge blocking layers may be provided to confine the charges within the device.

A material which is most widely used as electrode material in the production of transparent electrodes is indium tin oxide (ITO). Unfortunately, indium is becoming more and more scarce and consequently an inevitable cost increase is to be expected. Besides this, ITO shows the additional disadvantage that indium has a tendency to diffuse into the organic part of the organic light emitting diode, which limits the shelf and operating lifetime of ITO based organic electronics devices such as OLEDs or Organic PV.

WO2007/135171 discloses an organic light emitting device comprising an electrode which includes antimony-doped tin oxide or fluorine-doped tin oxide. The electrode layer may have a layer thickness of at least 100 nm, up to a maximum of 600 nm. Fluorine-doped tin oxide comprising electrodes disclosed in WO2007/135171may show work function values of up to 4.2 to 5eV, which may even be better than ITO. They may also show a good durability. Fluorine-doped tin oxide comprising electrodes disclosed in WO2007/135171, when applied on-line however show a high surface roughness which may amount to 8-11 nm when not subjected to polishing, measured using atomic force microscopy (AFM) on a square with a length of 10μm. This high surface roughness is one of the reasons for a high leakage current at low bias voltage, and for the rather low external quantum efficiency (EQE).

EP2.178.343 discloses a laminate for an OLED, wherein the laminate comprises a glass substrate and a scattering layer deposited on top thereof, a translucent ITO electrode with a translucency of 80% or more arranged on top of the

scattering layer, and further an organic layer having a light-emitting function and a reflective electrode. The scattering layer is formed by coating the substrate with a glass powder and firing it at a desired a temperature. The use of a sufficiently high firing temperature, in particular 580°C permits to obtain a material with a surface roughness of typically only about 1.8 nm measured using atomic force microscopy (AFM) on a square with a length of 10μm. A translucent electrode formed thereon has a smooth surface and shows a small risk to the occurrence of defects, and therewith a small risk to the occurrence of electric shortcuts. The scattering layer comprises a glass material with a plurality of scattering materials dispersed therein. The concentration of the scattering materials in the scattering layer decreases from the inside of the scattering layer towards the translucent electrode in such a way that within 0.2 μm from the main surface of the scattering layer no scattering materials are present. An OLED element comprising such a scattering layer and employing ITO as electrode material shows a good durability and a good external quantum efficiency (EQE), compared to that of a reference device which does not contain the scattering layer.

EP2.383.235 discloses a bottom-emission type organic light emitting device with an improved light flux, comprising an organic layer with a light emitting function formed on the first electrode, and a second electrode comprising LiF film-formed in a thickness of 0.5 nm and Al film-formed in a thickness of 80 nm on the organic layer. The scattering layer disposed on the translucent substrate comprises, in terms of mol % on the basis of oxides, 15 to 30% of P_2O_5, 5 to 25% of Bi_2O_3, 5 to 27% of Nb_2O_5, and 10 to 35% of ZnO, and has the total content of alkali metal oxides comprising Li_2O, Na_2O and K_2O of 5% by mass or less. The first electrode (anode) is required to have a translucency of 80% or more in order to permit sufficient extraction of light generated in the organic layer to the outside. To achieve this, specifically materials such as ITO (Indium Tin Oxide), SnO_2, ZnO, IZO (Indium Zinc Oxide), AZO (ZnO-Al_2O_3: zinc oxide doped with aluminum oxide), GZO (ZnO-Ga_2O_3: zinc oxide doped with gallium), Nb-doped TiO_2 and Ta-doped TiO_2 are used. The anode typically has a thickness of 100 nm or more and a refractive index of 1.9 to 2.2. The second

electrode has reflectivity for reflecting the light emitted from the organic layer and returning the light to the organic layer.

According to EP2.489.643, the scattering layer can be manufactured by coating a glass frit on a translucent substrate and firing the thus coated translucent substrate, to cause the glass paste to soften and sinter and produce a surface with a low surface roughness. The glass frit may take the form of a frit paste comprising a base glass material, a resin or a solvent and possibly a surfactant. The firing temperature is set within a range of between 40°C below to 30°C above the glass softening point T_s of the base glass material or within the range of 50°C to 120°C above the glass transition point T_g of the base glass material, which often corresponds to temperatures between 540 and 580°C.

The scattering layer disclosed in EP2.178.343 and EP2.489.643 when used in combination with ITO as electrode material, provides OLED's with a good EQE compared to that of OLED's without a scattering layer, as well as a good durability, and provides a protective layer which minimizes the risk to contamination. However, the use of indium in the electrode material is costly, and indium is becoming more and more scarce.

WO2012/007575 solves the problem of providing a translucent conductive substrate for an OLED that allows increasing the amount of light transmitted in forward through the substrate, in particular in the case of monochromic radiation. The translucent substrate has a surface roughness Ra of about 1 nm and comprises a transparent support for example glass, a scattering layer formed on the transparent support by depositing a glass powder using coating and firing at a desired temperature. The glass powder comprises a plurality of scattering materials dispersed therein. A transparent electrode formed over the scattering layer preferably comprises a single metal conduction layer, in particular at least Ag in pure form or alloyed to another metal. A coating applied on top of the metal conduction layer comprises at least one dielectric compound and/or at least one electrically conductive compound. Suitable dielectric compounds include oxides of for example Zn, Zr and Sn, nitrides

of for example Al or Si and mixed nitrides, oxynitrides and oxycarbides. The conductive material preferably includes ITO and/or F doped tin oxide. A further dielectric layer may be formed directly on the scattering layer. However, the electrode disclosed in WO2012/007575 has an EQE which is unsatisfactory and a too small durability due to the relatively soft coating.

There is thus a need for a an electrode material which shows both a good durability and a good external quantum efficiency (EQE), without requiring the presence of a rare metal such as indium.

This may be achieved according to the present invention with a translucent conductive substrate showing the technical features of the characterising portion of the first claim.

Thereto the translucent conductive substrate of this invention is characterised in that the electrode is made of a material comprising fluorine-doped tin oxide.

A translucent conductive substrate comprising fluorine-doped tin oxide as the conductive electrode material instead of ITO presents the advantage of offering electrodes with greater availability and lower cost.

The inventors have observed that a proper selection of the scattering material composition and the temperature - time regime used to deposit the scattering layer, permits to obtain a support material with a high surface smoothness on nanometer scale. In particular, a support material with a surface roughness on nanometer scale of typically 3 nm or less (as measured with AFM) may be obtained. The inventors have however also observed that despite the smoothness of the scattering layer surface, the surface of the subsequently applied F-doped SnO_2 layer has a rather high surface roughness at nanometer scale, often of 6-11 nm or 8-11 nm (as measured with AFM) when applied online and when not subjected to polishing.

Because of this high difference in value of the surface roughness between the scattering layer and the F-doped SnO_2 layer, the skilled person will expect that

the combination of the scattering layer and the fluorine-doped tin oxide layer will result in a rather high leakage current and consequently a low external quantum efficiency (EQE) and that the light outcoupling efficiency of an OLED comprising such a conductive substrate will be low. Or else, the skilled person will expect that in order to obtain a satisfactory EQE, polishing of the surface of the conductive layer will be required to reduce its surface roughness. It is a generally accepted principle that in order to provide an OLED with a high EQE, the conductive substrate needs to have a low surface roughness on nanometer scale in order to reduce the leakage current. In view of this teaching, the skilled person designing conductive substrates, will normally seek to diminish the surface roughness of the conductive substrate to values around about 1 nm, rather than going the opposite way and seeking to increase the surface roughness.

The inventors have now surprisingly found that an OLED comprising the conductive substrate of the present invention may achieve a desired EQE, the magnitude of which is comparable to the EQE of the closest prior art translucent conductive substrate using ITO, without requiring polishing to improve the flatness of the surface of the conductive substrate or without the need to use a thicker organic layer or to dramatically change the organic stack architecture. This is surprising in view of the teaching of the prior art that conductive translucent substrates with a high surface roughness of for example 6-11 nm show an unsatisfactory EQE when applied on a standard thin organic stack, and that a low surface roughness of typically about 1 nm is necessary for obtaining a good EQE. The present inventors have particularly found that when a conductive substrate comprising a scattering layer - which in turn comprises a glass which contains a base material having a first refractive index for at least one wavelength of light to be transmitted and a plurality of scattering materials dispersed in the base material and having a second refractive index different from that of the base material - is used in combination with fluorine-doped tin oxide as transparent electrode material, a device may be obtained with a good EQE which is comparable to the EQE of the device employing ITO as the electrode material using the same organic stack. Without wanting to be bound to this theory, the inventors assume that the texture of the conductive substrate of the present invention forming

the electrode, in particular its roughness especially at nanometer scale, contribute to reducing any adverse effect resulting from scattering and/or reflection which may originate from the scattering layer, to a minimum.

The surface of the F-doped SnO_2 conductive layer according to the present invention, will usually have an average surface roughness Ra of at least 7.5 nm, preferably at least 8, more preferably at least 8.5 or 9, often at least 9.4, as determined by atomic force microscopy. Where reference is made to "surface roughness", the surface roughness has been measured using atomic force microscopy (AFM) on a square sample with a length of 10μm.

In a preferred embodiment, the translucent conductive substrate for an organic light emitting device, consists essentially of

- a transparent support,
- a scattering layer formed over the transparent support, the scattering layer comprising a glass which contains a base material having a first refractive index for at least one wavelength of light to be transmitted, the scattering layer further comprising a plurality of scattering materials dispersed in the base material and having a second refractive index different from that of the base material, and
- a layer of transparent electrode material formed over the scattering layer.

According to a preferred embodiment, the atomic ratio F/Sn in the F-doped SnO_2 varies from 0.005 to 0.20, preferably from 0.01 to 0.10, more preferably from 0.01 to 0.05.

Advantageously, the F-doped SnO_2 layer thickness is at least at least 100 nm, preferably at least 250 nm, more preferably at least 300 nm, but not more than 700 nm, preferably not more than 600 nm, more preferably not more than 550 nm or 500 nm, even more preferably not more than 475. Such thicknesses permit to achieve good conductivity while simultaneously reducing undesirable variations or

non-homogeneity to a minimum and maintaining optical properties in the visible range within acceptable values, in particular light reflection and light transmission.

Advantageously also, the face of the scattering layer facing the F-doped SnO_2 material is coated with a smoothening layer, said smoothening layer being, starting from the support the outermost layer of said scattering layer. The thickness of the smoothening layer is not particularly limited. Preferably however the smoothening layer has a thickness of at most 400 nm, more preferably a thickness of around 300 nm. The smoothening layer assists in shielding the electrode material from contaminants present in the scattering layer, and reducing the risk to the occurrence of electrical short cuts. The smoothening material is preferably a material selected from the group of titanium oxide, niobium oxide, zirconium oxide, tantalum oxide and silicon oxide or a mixture of two or more of these materials. Advantageously, said smoothening layer is made of a mixed oxide of silicon and titanium. The smoothening layer preferably has a refraction index which is lower than that of the electrode in order to improve the light extracting efficiency. The smoothening layer may be applied by magnetic sputtering, chemical vapour deposition (CVD), but also by a wet-coating process, all these processes being well known to the skilled person.

The present invention provides a translucent conductive substrate with a high Haze of at least 90 %, preferably at least 95 %. The Haze parameter determines the portion of light that is scattered (diffused light (Idif)) at the interface compared to total intensity (Itot) of light (specular (Ispec) and diffused light:

$$H = Idif/Itot = Idif/(Ispec + Idif)$$

According to the invention, "Haze" is understood to mean the percentage of transmitted light which, while passing through the sample, deviates from the incident beam by an angle of more than 2.5°. According to the present invention, the "Haze" parameter is determined using ASTM D1003-61standard.

The inventors have observed that the conductive substrate of the present invention gives a promising performance in the OLED, with a sheet resistance of less than 25 Ω/□, preferably less than 20 Ω/□, more preferably less than 15 Ω/□. With the conductive substrate of this invention, OLED's may be produced having a light outcoupling efficiency of at least 1.5, which is at least as good as the closest prior art material utilizing ITO as electrode material.

In the translucent conductive substrate of the present invention, F-doped SnO_2 electrode materials offer work function values between 4 and 5 eV, preferably between 4.2 and 5 eV.

The conductive substrate of the present invention has further been found to show a high mechanical durability. Mechanical durability may for example be tested using the Dry Brush Test according to ASTM D2486. This is a scratch-resistance test carried out over 4000 cycles, which classifies materials into a classification A, B, C, D, etc. according to their scratch resistance. This classification is explained in EN norm1996-2. Group A materials show the best scratch resistance. Conductive substrates comprising ITO as the electrode material typically belong to the A-class materials, whereas conductive substrates containing an Ag based electrode typically belong to C-class materials.

The transparent support present in the conductive substrate of this invention may be coated on the face where the scattering layer is deposited, with at least one barrier layer. This barrier layer applied between the transparent support and the scattering layer in particular allows the electrode to be protected from any contamination by the migration of alkaline substances coming from the support, e.g. made of soda-lime-silica glass, and thus enables the service life of the electrode to be extended. The barrier layer may for example comprise at least, consist, or consist essentially of one or more compounds selected from the group of silicon oxide, aluminium oxide, titanium oxide, mixed oxide of zinc-tin, mixed oxide of zinc-aluminium, silicon nitride, aluminium nitride, titanium nitride, silicon oxynitride, aluminium oxynitride.

The present invention also relates to an OLED comprising a translucent conductive substrate according to any one of claims, further comprising an organic layer positioned on top of the fluorine-doped tin oxide anode and a second electrode layer positioned on top of the organic layer. The present invention permits obtaining an OLED with a satisfactory EQE. In general, the ratio of the EQE of an OLED according to the present invention to the EQE of an OLED comprising ITO, will in general be at least 60 %, preferably at least 70%, more preferably at least 80%. The present invention further permits obtaining an OLED with a light outcoupling efficiency of at least 1.5, which is superior to the light outcoupling efficiency that may for example be achieved with ITO or Ag as the electrode material.

The present invention also relates to a process for producing a conductive substrate for an organic light emitting device.

The present invention in particular relates to a process for producing a conductive translucent substrate for use in an organic light emitting diode, wherein a transparent electrode material is deposited on top of a scattering layer deposited on the transparent support, wherein the scattering layer comprises a base material having a first refractive index at a wavelength of emitted light of an organic LED element and a plurality of scattering materials provided in the inside of the base material and having a refractive index different from that of the base material, wherein the scattering material is deposited on the transparent support by deposition of a frit paste and heating the thus coated support to dry and fire the frit paste to form the scattering layer.

The method of this invention is characterised in that as the electrode material use is made of F-doped SnO_2, in that the electrode material is deposited by deposition and decomposition of a precursor at a temperature of between 500 and 650°C, preferably between 500 and 625°C, more preferably between 520-600°C to decompose the precursor, followed by cooling of the thus obtained conductive substrate.

A preferred embodiment for applying the scattering layer involves coating the substrate with a frit-pasted glass, followed by drying and firing the frit-pasted glass at a desired a temperature. The frit pasted glass will usually contain a glass powder and a vehicle. A suitable vehicle comprises a solvent and a resin. A suitable method for preparing the frit paste is for example disclosed in EP 2.178.343 A1.

The inventors have surprisingly found that heating to the claimed temperatures does not give rise to mixing of the components of the scattering layer and the conductive F-doped SnO_2, layer deposited on top thereof, independently of the fact whether the material of the scattering layer has been subjected to drying only or been subjected to drying as well as to a process of firing, in advance of deposition of the conductive F-doped SnO_2 layer. To the contrary, the claimed temperature range has been found to offer an optimal compromise between permitting to optimise the properties of the scattering layer and obtaining an electrode covered conductive substrate with the desired electrical properties and the required low surface roughness. The inventors have also surprisingly found that the drying and firing of the scattering layer is not adversely affected by the deposition of the F-doped SnO_2 layer, independently of the fact whether the material of the scattering layer has been subjected to drying only or been subjected to drying as well as to a process of firing, in advance of deposition of the precursor for the F-doped SnO_2 layer.

From the prior art teaching it appears that in order to obtain an electrode surface with a sufficiently low sheet resistance of preferably below 15 Ω/□, the deposition temperature of the F-doped SnO_2 electrode layer should be rather low, i.e. between 550 and 570°C and certainly below 580°C to minimise the risk that the scattering layer would be adversely affected. On the other hand, according to the teaching of the prior art, the temperature at which the precursor for the F-doped SnO_2 layer is deposited should be sufficiently high, i.e. minimum 650°C to ensure a sufficiently complete decomposition of the precursor which is used to deposit the F-doped SnO_2. The inventors have now found that the claimed temperature range of between 500 and 650°C, preferably between 500 and 625°C, more preferably between 520-600°C, although rather low in comparison to the prior art teaching,

permits to achieve a sufficiently complete decomposition of the precursor and a formation of the F-doped SnO_2 layer while ensuring that the integrity of the scattering layer may be maintained or is affected to the smallest possible extent only.

Thus, the present invention presents the advantage that deposition of the precursor for the electrode material on top of the transparent support and the scattering layer and decomposition of the precursor can be done on-line, i.e. in line with the firing of the scattering layer. This is of significant economic importance as it reduces the number of heating steps required, and permits to build an integrated process for the deposition and heat-treatment of the scattering layer and the F-doped SnO_2. The deposition of the electrode material on-line with the deposition of the scattering layer moreover permits to minimise the risk to contamination of the surface of the scattering layer between the steps of firing of the scattering layer and deposition of the electrode material, since it is done sequentially without exposing the substrate to ambient conditions.

In the on-line process, usually the scattering material will be dried in advance of depositing the precursor for the F-doped SnO_2 conductive layer. Firing will be carried out simultaneously with the step of depositing the F-doped SnO_2 conductive layer. The texture of the scattering layer will develop in the course of the deposition of the F-doped SnO_2 layer. In such a process, the substrate coated with the material for the scattering layer is preferably preheated to a temperature of at least 500°C, preferably at least 520°C, more preferably at least 540°C, most preferably a temperature between 540-600°C.

According to a second preferred embodiment of the process of this invention, the scattering layer is dried and fired in advance of depositing and decomposing the precursor for the F-doped SnO_2 layer.

Although in view of the prior art teaching, the claimed temperature region for depositing the electrode material is thought to be rather high for the material of the scattering layer, the inventors have observed that even in the case where the precursor for the electrode material is deposited and decomposed after the scattering

layer has been applied and subjected to drying and firing, the properties of the scattering layer are not adversely affected and that no undesired structural transformation of the material of the scattering layer takes place.

Although the skilled person would expect that the claimed temperature region would reduce the viscosity of the base glass material of the scattering layer to a too low level, a scattering layer with a desired surface roughness of typically between 0.5 and 4 nm, often between 1-2 nm may be obtained. Surprisingly, the viscosity of the base glass material of the scattering layer does not sink to such a level that pores would be able to rise to the surface and crystals to precipitate at the surface, and crystal precipitation as well as pore generation in the outermost surface of the scattering layer may be maintained at a desired minimum. Surprisingly also, despite the use of a rather high temperature range for the scattering layer, the risk to disintegration and cracking of the material of the scattering layer may be kept minimal and a good adhesion of the scattering layer to the substrate may be achieved.

According to the present invention, the conductive substrate is progressively cooled after deposition of the F-doped SnO_2 electrode material has been completed, to permit stabilising of the texture of the scattering layer and to minimize the risk to the formation of cracks in the scattering layer. The cooling procedure may take place according to a temperature – time regime to be defined by the skilled person and will usually vary from a few minutes to maximum a few hours, preferably 10-60 minutes, more preferably 15-45 minutes, typically about 30 minutes. Thereby, the substrate may be maintained at a temperature which corresponds to or is lower than the F-doped SnO_2 deposition temperature, to achieve annealing.

In the deposition zone, heating may be applied along either the bottom of the substrate or both the top and the bottom of the substrate.

Electrode material.

F- doped SnO_2 may be deposited on the scattering layer using any technique considered suitable by the skilled person. Preferably use is made of chemical vapour deposition of an organic precursor, followed by decomposition of the precursor. The precursor is usually a product which may be pyrolysed or thermally decomposed at the temperatures at which the process is carried out. The precursors of SnO_2 are typically alkyltins, such as tetramethyltin, tetraethyltin, tin chloride ($SnCl_4$) or organotin chlorides, such as MBTC (monobutyltin trichloride). In the present invention use of the latter is preferred.

The reactive gas used in the chemical vapour deposition is based on oxygen the use of which is preferred, or on oxygen-comprising derivatives, for example ozone, hydrogen peroxide, water and CO_2. The reactive gas may comprise nitrogen-comprising derivatives, for example NH_3, N_2O and HCN. The reactive gas may further include an inert gas, such as helium, nitrogen, argon, neon or krypton, in order to promote the chemical dissociation of the precursors.

Doping of SnO_2 with F may be achieved using various reactants, for example HF or tri-fluoro acetic acid. More preferably however, a mixture of MBTC (monobutyltin trichloride), HF, N_2, O_2 is preferred. If so desired, water may be supplied as well.

The proportion of HF with respect to MBTC in the reactive gas is preferably adjusted in such a way as to achieve an electrode film with a sufficiently low sheet resistance. Thereto, the weight ratio of HF/MBTC is preferably less than 1, more preferably less than 0.75, most preferably less than 0.5, in particular less than 0.3. A particularly preferred weight ratio of HF/MBTC is in the range of 0.05-0.25, more particularly 0.1-0.2.

The proportion of water with respect to MBTC is preferably also adjusted in such a way as to achieve an electrode film with a sufficiently low sheet resistance.

Thereto, the weight ratio of H_2O/MBTC is preferably at least 2, more preferably at least 5. A particularly preferred ratio is about 7.

The proportion of inert gas in the mixture of the reactive gas and the inert gas is advantageously defined by the ratio of the flow rate of the reactive gas to the flow rate of the inert gas and is located in the range of 2-50, preferably of 10-30, very advantageously of 15-25. A proper selection hereof makes it possible to control the surface roughness (Ra) of the deposited film. Preferably, Ra is less than 2 nm, indeed even of less than 0.5 nm, which, for a layer with, for example, a thickness of between 200-400 nm as this may substantially improve the barrier properties.

Where deposition of the F-doped SnO_2 electrode material takes place in the presence of O_2, usually a ratio O_2/MBTC of less than 25, preferably less than 20 will be used.

Transparent support.

Materials suitable for use as transparent support in the translucent conductive substrate of this invention have a high transmittance to visible light and a high refraction index. Examples of suitable materials having high transmittance to visible light include glass, for example a glass sheet or a sheet of float glass, alkali-free glass or quartz glass or the like but a plastic substrate may be used as well. The thickness of the transparent substrate is not particularly limited but may be, for example, within a range of 0.1 mm to 8.0 mm. When considering optimisation of the strength and the weight, the thickness of the transparent substrate may be 0.5 mm to 1.4 mm.

Barrier layer.

If so desired, the transparent support material may be coated with at least one barrier layer on the face on which the scattering layer is deposited. This barrier layer may be an iridescence-reducing layer or a barrier layer adapted to inhibit, or at least reduce, alkali migration which may occur, for example, from a glass substrate towards the layer including fluorine-doped tin oxide. The barrier layer may also be a

haze-optimising layer or an anti-reflection layer, which is preferably index matched either to the substrate or to the scattering layer. Such layers are known, for example, from EP275662, GB2302102, GB2248243, US4377613. The barrier layer may comprises one or more compounds selected from the group of silicon oxide, aluminium oxide, titanium oxide, a mixed zinc-tin oxide, a mixed zinc-aluminium oxide, silicon nitride, aluminium nitride, titanium nitride, silicon oxynitride, aluminium oxynitride. The barrier layer may for example be applied using sputtering or chemical vapour deposition.

The additional layer may advantageously have a thickness of at least 5 nm, at least 10 nm, preferably at least 40 nm, at least 50 nm, more preferably at least 60 nm. Its thickness may preferably be not more than 100 nm, not more than 90 nm and more preferably not more than 80 nm. A suitable layer thickness is between 30 and 40 nm.

Scattering layer.

The conductive translucent substrate of this invention preferably comprises a scattering layer formed over the transparent support. Said scattering layer comprises a glass which contains a base material having a first refractive index for at least one wavelength of light to be transmitted. The scattering layer further comprises a plurality of scattering substances 111 dispersed in the base material 110 and having a second refractive index different from that of the base material. The amount of the scattering substances in the light scattering layer 120 preferably decreases from the inside of the light scattering layer 120 to the outside of the light scattering layer 120, as this permits increasing light extracting efficiency.

Compositions suitable for use as the scattering layer in the translucent conductive substrate of this invention are for example described in WO2013/054820 (A1) or US2014/0191223. The composition of the scattering layer is preferably chosen such that it is compatible with the temperature regime used. Preferably the scattering layer has a thickness in the range of 2 to 60μm, more preferably 5 to 40 μm.

Materials suitable for use as a base material of the scattering layer have a high light transmittance. Suitable examples include a glass, a crystallized glass, a translucent resin or a translucent ceramic may be used. As a material for the glass, an inorganic glass such as soda lime glass, borosilicate glass, non-alkali glass, low-alkali glass or quartz glass may be used.

Examples of scattering materials suitable for use with this invention are pores or bubbles of a material, precipitated crystals, material particles having a different chemical composition than base material, phase-separated glass or mixtures from at least two thereof.

It is preferable that the difference between the refraction indexes of the base material and the scattering substances is large. For this reason, it is preferable that a high refraction index glass is used for the base material and pores of a material are used for the scattering substances. For the high refraction index glass used for the base material, one or more components may be selected from P_2O_5, SiO_2, B_2O_3, GeO_2 and TeO_2 as a network former and one or more components may be selected from TiO_2, Nb_2O_5, WO_3, Bi_2O_3, La_2O_3, Gd_2O_3, Y_2O_3, ZrO_2, ZnO, BaO, PbO and Sb_2O_3 as a high refraction index component. Further in order to adjust characteristics of the glass, an alkali oxide, an alkaline earth oxide, a fluoride or the like may be added within a range not impairing characteristics for the refraction index.

Thus, for the glass system composing the base material, for example, a B_2O_3—ZnO—La_2O_3 system, a P_2O_5—B_2O_3—R'_2O—R″O—TiO_2—Nb_2O_5—WO_3—Bi_2O_3 system, a TeO_2—ZnO system, a B_2O_3—Bi_2O_3 system, a SiO_2—Bi_2O_3 system, a SiO_2—ZnO system, a B_2O_3—ZnO system, a P_2O_5—ZnO system or the like is exemplified. Here, R′ represents an alkali metal element and R″ represents an alkaline earth metal element. The above material systems are examples and the materials to be used are not limited as long as satisfying the above-mentioned conditions. By adding a colorant in the base material, color of light emission can be changed. For the colorant, a transition metal oxide, a rare earth metal oxide, a metal colloid or the like may be used singly or in combination thereof.

In a preferred embodiment of this invention, the glass for a scattering layer includes as represented by mol percentage based on the following oxides : 26% to 43% of B_2O_3, 30% to 37% of ZnO, 17% to 23% of Bi_2O_3, 2% to 21% of SiO_2, and 0 to 2% of P_2O_5; and a total B_2O_3-content and ZnO-content of 78% or less. The scattering layer includes, within a base material made of glass, a scattering material having a refractive index different from that of the glass and dispersed within the base material, and is formed by sintering a raw material including the glass.

Coating Layer

The material of the coating layer is not particularly limited, but the coating layer may include ceramics such as titanium oxide, niobium oxide, zirconium oxide, tantalum oxide or the like, for example. Further, in addition to the above described ceramics, the coating layer may further include silicon oxide (SiO_2). For example, the coating layer may be a layer composed of a mixture of titanium oxide and silicon oxide. The ratio of titanium oxide and silicon oxide is not particularly limited, but the ratio of them (titanium oxide:silicon oxide) may be within a range of 80:20 to 20:80, by weight ratio, for example. In particular, the ratio of titanium oxide:silicon oxide is preferably within a range of 75:25 to 40:60, by weight ratio.

The refraction index of the coating layer is preferably lower than that of the electrode 140 in order to improve the light extracting efficiency. Specifically, the difference between the refraction index of the coating layer and the refraction index of the light scattering layer 120 is preferably 0.2 or less, more preferably 0.13 or less and furthermore preferably 0.11 or less.

When a material, having a resistance against etching solution that is used in an etching process of the first electrode, is used as the material of the coating layer, a problem that the light scattering layer and the coating layer are damaged in a patterning process of the first electrode can be suppressed.

Thus, it is not necessary to select the material of the light scattering layer from materials having a resistance against the etching solution so that the material of

the light scattering layer can be selected from a wider range. The entirety of the above described oxide materials have a resistance against the etching solution (for example, hydrochloric acid system solution including ferric chloride or the like) generally used in the etching process of the first electrode.

The thickness of the coating layer is not particularly limited. The thickness of the coating layer may be, for example, within a range of 100 nm to 500 μm. Here, the coating layer is formed by the wet-coating according to the embodiment. Thus, according to the embodiment, different from the dry process such as sputtering, a relatively thick layer can be easily formed by repeating the wet-coating process.

First Electrode

A wide variety of electrode materials may be used. Suitable materials for use as a first electrode include a material such as ITO, SnO_2, ZnO, Indium Zinc Oxide (IZO), ZnO—Al_2O_3 (AZO: aluminum doped zinc oxide), ZnO—Ga_2O_2 (GZO: gallium doped zinc oxide), Nb doped TiO_2, Ta doped TiO_2 or the like.

The thickness of the first electrode may vary within wide ranges, and is preferably more than or equal to 100 nm.

It is preferable to determine the refraction index of the first electrode considering the refraction index of the base material composing the light scattering layer or the refraction index of the second electrode. It is preferable that the difference between the refraction indexes of the first electrode and the base material is less than or equal to 0.2 considering a calculation of waveguide, a reflectance of the second electrode or the like.

Refraction indexes of the components from the light scattering layer to the first electrode are described for reference. However, the following combination of the refraction indexes is just an example and the components may have different refraction indexes, respectively.

The refraction index of the light scattering layer is, for example, within a range of 1.8 to 2.0 (about 1.84, for example). The refraction index of the coating layer is, for example, within a range of 1.7 to 2.0 (about 1.75, for example). The refraction index of the first electrode is, for example, within a range of 1.8 to 2.2 (about 1.8, for example).

If so desired, the scattering layer may be covered by a smoothening layer. Preferably, the smoothening layer has a thickness of at least 200 nm. Preferably the smoothening layer has a thickness of at most 400 nm. More preferably, the smoothening layer has a thickness of around 300 nm. Advantageously, the smoothening layer is made of a mixed oxide of silicon and titanium. The presence of a smoothening layer assists in reducing the occurrence of electrical shortcuts.

The scattering layer can be manufactured by coating glass frit on the translucent substrate and firing the translucent substrate with the coated glass frit. Suitable glass frit materials and frit paste materials are described below.

Glass Frit.

The glass frit includes powder of the base material glass. The particle diameter of the powder of the base material glass is preferably 1-10 μm in view of coatability. The surface of the powder of the base material glass may be modified by a surfactant or a silane coupling agent. The glass frit may include glass powder having a refractive index lower than the base material glass, as the scattering materials, other than the powder of the base material glass. The glass frit is preferably applied to the translucent substrate as a frit paste kneaded with resin or a solvent, in view of coatability.

Frit Paste

The frit paste can be obtained by mixing a glass frit with a vehicle. The frit paste may be further kneaded by a kneading machine to adjust viscosity. In general, 70-80 wt. % of glass frit and 20-30 wt. % of vehicle are mixed. The vehicle is a mixture of resin and a solvent and a surfactant may be further mixed therein. The

resin is provided to keep the shape of the frit paste film after applying. Suitable resins include ethylcellulose, nitrocellulose, acrylic resin, acetic acid vinyl, butyral resin, melamine resin, alkyd resin, and rosin resin. Ethylcellulose or nitrocellulose is often used as a base resin. Butyral resin, melamine resin, alkyd resin, and rosin resin are used as additives for improving coating film strength. The debinderizing temperature in firing is 350-400°C for ethylcellulose and 200-300°C for nitrocellulose.

The solvent is provided to adjust the viscosity of the frit paste while dissolving the resin. The solvent is usually a material with a boiling point of preferably 200-230°C. In detail, the solvent may be ether-based, alcohol-based, ester-based, phthalic ester-based. The solvents may be independently used or may be used together with each other to adjust viscosity, a solid ratio, and a dry speed.

As a method of coating a frit paste onto the transparent substrate, screen printing doctor blade printing, die coating printing and the like are used. It may be possible to obtain a green sheet by coating and drying the frit paste on an PET film or the like, and then thermally-pressing the green sheet onto the translucent substrate. When screen printing is used, it is possible to control the thickness of the frit paste film after coating by adjusting the mesh roughness of the screen plate, the thickness of the emulsion, the pressing pressure in printing, and the pressed amount of squeegee. When doctor blade printing and die coating printing are used, as compared with when screen printing is used, it is possible to make the frit paste film thick after coating.

Firing

A frit paste coated on the translucent support is dried and fired, in order to decompose the resin in the glass paste and allowing it to disappear, and achieve sintering and softening of the glass paste after the debinderizing treatment. The debinderizing temperature is 350-400°C for ethylcellulose and 200-300°C for nitrocellulose, and heating is performed under the atmosphere from 30 minutes to 1 hour.

According to a first embodiment, the frit paste is dried and fired in advance of depositing the conductive F-doped SnO_2 layer. In that case drying and firing is preferably carried out at a temperature set within the range of 500-625 °C, preferably 520-600°C, more preferably 520-580 °C. The shape and size of the scattering material remaining in the scattering layer may be adjusted by adjusting the firing temperature, the firing atmosphere, and the particle size distribution of the glass frit. The scattering layer is formed on the translucent substrate by cooling the translucent substrate with the coated frit paste to the room temperature after firing.

According to a second embodiment, the frit paste is only dried in advance of depositing the conductive F-doped SnO_2 layer, and firing is carried out simultaneously with the deposition of the conductive F-doped SnO_2 layer. In that case firing is preferably carried out at a temperature set within the range of 500-625 °C, preferably 520-600°C.

The substrate and organic LED element of the present invention are further described using the drawings. It should be clear that parts having the same reference numerals have the same constitution or the same function.

Fig. 1 is a cross-sectional view of a first organic LED element of the present invention.

Fig. 2 is a cross-sectional view of a second organic LED element of the present invention.

The translucent conductive substrate according to this invention comprises a transparent support 110, a scattering layer 120 formed over the transparent support 110 and a layer of transparent electrode material formed over the scattering layer 120. The scattering layer comprises

- a glass which contains a base material 121 having a first refractive index for at least one wavelength of light to be transmitted,

- a plurality of scattering materials 122 dispersed in the base material 121 and having a second refractive index different from that of the base material,

The first organic LED element of the present invention is a bottom-emission type organic LED element, having a transparent substrate 110, a scattering layer 120 formed on the transparent substrate 110, a first electrode 130 formed on the scattering layer 120, an organic layer 140 formed on the first electrode 130, and a second electrode 150 formed on the organic layer 140. In the first organic LED element of the present invention, the first electrode 130 is a transparent electrode (anode), and the second electrode 150 is a reflective electrode (cathode). The first electrode 130 has transparency for transmitting light emitted from the organic layer 140 to the scattering layer 120. On the other hand, the second electrode 150 has reflectivity for reflecting the light emitted from the organic layer 140 and returning the light to the organic layer 140.

Fig. 2 shows an Organic LED element which is a both surface-emission type organic LED element. The second organic LED element of the present invention has a transparent substrate 110, a scattering layer 120 formed on the transparent substrate 110, a first electrode 130 formed on the scattering layer 120, an organic layer 140 formed on the first electrode 130, and a second electrode 210 formed on the organic layer 140. In embodiment shown in fig. 2, the first electrode 130 is a transparent electrode (anode), and the second electrode 210 is a transparent electrode (cathode). The first electrode 130 has transparency for transmitting light emitted from the organic layer 140 to the transparent substrate 110. On the other hand, the second electrode 210 has transparency for transmitting light emitted from the organic layer 140 to the surface opposite the surface facing the organic layer 140. The organic LED element is used in illumination applications in which light is emitted from both sides.

The invention is further elucidated in the examples given below.

Examples.

Translucent conductive substrates were produced by feeding glass as a translucent support material to a heating furnace. The furnace comprises a heating zone for heating the material, followed by a F-SnO_2 deposition zone where F-SnO_2 is deposited on top of the support material using chemical vapour deposition. The F-SnO_2 deposition zone may further comprise an inlet for supplying N_2, and H_2O, O_2, HF and MBTC (monobutyltintrichloride) as a precursor for the conductive electrode material.

In the tables below the following expressions have the following meaning:

- Rs (Ω/□) : sheet resistance
- Haze Guard Dual (%) by CRD, Sheen Instruments.
- Ra (nm) : surface roughness measured on quartrexed D3100 IIIA» from Veecco on a square surface of 10μm by10μm in tapping mode. Use was made of an AFM point RTESP f°300KHz obtained from Bruker, in the tapping mode. A scanning speed of 1 Hz was used. The principle of AFM for determining surface roughness is for example described in JIS B0601-1994 (see EP2.178.343 p.12 l.5) or in Applied Surface Science, Volume 133, Issue 4, August 1998, Pages 293–297
- J mA/cm^2 at -1V : current density
- EQE (%) at 1000cd/m^2 : external quantum efficiency
- Max Ceff (cd/a) at 1000cd/m^2

Comparative Example A.

An organic EL element was manufactured as follows.

A soda lime substrate having a length of 50 mm, a width of 50 mm and a thickness of 0.55 mm was prepared as the transparent substrate. Then, a translucent substrate was prepared by forming the following layers on the transparent substrate.

First Translucent Substrate.

The first translucent substrate includes the light scattering layer and the coating layer on the transparent substrate.

The light scattering layer and the coating layer were manufactured as follows.

Light Scattering Layer.

A source material for the light scattering layer was prepared by the following method.

First, mixed particles having the composition shown in Table 1 were prepared and were dissolved. The dissolution was performed by retaining the mixture at 1050°C for 1.5 hours and then retaining the mixture at 950°C. for 30 minutes. Thereafter, the dissolved object was casted in a twin-roll process to obtain a flake glass.

The refraction index of the flake glass was measured using a refractometer (trade name: KRP-2, manufactured by Kalnew Optical Industrial Co., Ltd.). The refraction index "nd" of the flake glass was 1.84 at d-ray (587.56 nm).

Then, the flake glass was pulverized by a planetary ball mill made of zirconia for two hours to obtain a glass powder having a mean grain diameter (d50: grain size at integrated value 50%, unit: μm) of 1 μm to 3 μm.

Next, 15 vol. % of silica balls each having diameters of 3 μm was added to 75 g of the glass powder, and kneaded with 25 g of an organic vehicle (prepared by dissolving about 10 mass % ethyl cellulose in α-terpineol or the like) to prepare a glass paste. The glass paste was printed on the soda lime substrate using a screen

printer. With this, a scattering layer having two circular patterns each having diameters of 10 mm was formed on the soda lime substrate. After the screen printing, the soda lime substrate was dried at 120°C. for 10 minutes.

The soda lime substrate was heated up to 450°C, in 45 minutes, maintained for 10 hours at 450°C, further heated to 575°C in 12 minutes, maintained for 40 minutes at 575°C, and thereafter cooled to the room temperature in three hours. The thickness after sintering (or baking) was 15.0 μm. Then, the paste without the silica balls was prepared similarly as the above method to form a covering layer. The thickness of the covering layer was 15.0 μm. With this operation, the light scattering layer with the covering layer was formed on the soda lime substrate.

The light scattering layer had a thickness of 30 μm in total.

TABLE 1

COMPOUND	mol %
B_2O_3	40.6
Bi_2O_3	19.3
ZnO	34.2
SiO_2	5.9
Al_2O_3	0
GLASS-TRANSITION TEMPERATURE Tg (°C.)	462

Formation of Coating Layer.

The coating layer was formed on top of the light scattering layer as follows.

A mixture of predetermined amounts of various organic metal compounds was diluted by solvent capable of retaining stability such as toluene, heptane, 1-butanol, methoxybutanol or the like to obtain liquid having a viscosity appropriate for forming the coating layer. The liquid for forming the coating layer was dropped on the light scattering layer formed on the glass substrate to form a coated layer using a spin coater.

Example 1-4.

Use was made of a plate of soda lime silicate glass as a transparent support material of 0.5 x 0.4 m, and a thickness of 1.1 mm. The glass was moved towards the heating furnace, through the pre-heating zone where a surface of the glass covered with the glass frit is pre-heated at a moving speed of 1.5 m/min.

A paste as described in table 1 above is formed by mixing 75 g of glass frits and 25 g of organic vehicle (10 mass % of ethyl cellulose dissolved into [alpha]-terpineol). Next, the paste is screen-printed in a range of 35 mmx35 mm on the alkali silicate glass substrate (PD200 manufactured by Asahi Glass Company, Limited) having a size of 100 mmX100 mm and a thickness of 1.8 mm, dried for 30 minutes at 150°C, once returned to room temperature and then raised to a temperature of 475°C. in 48 minutes, and held at 475°C. for 30 minutes, in order to decompose and eliminate the resin of the organic vehicle. Thereafter, the temperature is raised to the glass-transition temperature plus 130°C in 10 minutes, the temperature is held at the raised temperature for 40 minutes to soften the glass, and the temperature is thereafter lowered to room temperature in 3 hours in order to form the glass layer. The thickness of the glass layer that is formed is 15 μm.

The thus covered glass support was forwarded to a deposition zone where chemical vapour deposition of the F-SnO_2 electrode material is carried out at the temperatures indicated in table 1 below. Chemical vapour deposition (CVD) of F-SnO_2 was done using MBTC supplied in an amount of 60 g/min, in the presence of O_2 supplied in a molar ratio with respect to MBTC of 10.1, HF supplied in a molar ratio of 0.1 with respect to MBTC.

Downstream of the F-SnO_2 deposition zone, a vaporiser was provided to evacuate the remainders of the precursor for the SnO_2 deposition.

The furnace settings during CVD of the F-SnO_2 electrode material and firing of the scattering layer were as indicated in table 1 below.

Table 1.

Example N°	1	2	3	4	Comparative example A
Substrate	F*	F*	F*	NF**	
Furnace settings Preheating zone temperature °C	540	560	580	580	
Deposition zone temperature °C	540	560	580	580	
cracks	no	no	no	no	no
Rs (Ω/□)	20.3	13.4	12.6	12.2	< 15
Haze Guard % By CRD	100	100	101	97	
Tt % (Haze Guard)	59.6	59.4	58.9	61.6	
Ra (nm)	13.8	13.8	15.2	16.4	< 1.5
J mA/cm² at -1V	-0.17	-0.28	-0.08	-0.69	-0.08
EQE (%) at	7.6	7.3	6.9	5.6	9.6

1000cd/m^2					
Max Ceff (cd/A) at 1000cd/m^2	16.6	16.4	15.5	12.3	22.5

* F = scattering layer fired in advance of deposition of F-doped SnO_2,

** NF = scattering layer not fired in advance of deposition of F-doped SnO_2.

From the results shown above it may be concluded that increasing the temperature at which the scattering layer is fired and the F-doped SnO_2 material is deposited, may result in a reduction of the sheet resistance (J), which is energetically favourable. The External Quantum Efficiency EQE, Haze are sufficiently high and approach the values obtained for the currently existing system, which is considered to be the reference material.

Example 5-7.

The experiment as described for examples 1-4 was repeated, however this time using another test installation.

The furnace settings were as indicated in table 2 below.

Table 2.

Example N°	5 AC131226-12	6 AC131226-14	7 AC131226-16
Furnace settings	520	590	590
	520	520	540
substrate	F	NF	NF
Rs (Ω/□)	25.0	23.0	15.8
Haze %	101	104	104
Tt % by Haze Guard	65.4	54.7	57.4
J (mA/cm^2)	-0.11	-0.16	-0.16
EQE	5.1	7.9	6.4
Ceff (cd/A) At 1000 cd/m^2	10.8	18.3	14.5

From the description of the invention given above, it should become clear that the conductive substrate of this invention may offer electrodes at least as efficient as ITO electrodes but with greater availability and/or at a lower cost. The present invention permits producing organic LED's with a satisfactory current efficiency, of at least 10, preferably at least 12.5, more preferably at least 15 cd/A at 1000 cd/m^2, and a good current density at an operating voltage of 1V which is similar or better than the prior art devices, combined with a uniform current density distribution over the substrate. The above in particular holds for a LED comprising materials described in Reineke et al. (NATURE| Vol 459| 14 May 2009) or in Rosenow et al. (Journal of Applied Physics, Vol. 108, 113113 (2010).

CLAIMS

1. **A translucent conductive substrate** for an organic light emitting device, comprising

- a transparent support (110),
- a scattering layer (120) formed over the transparent support (110), the scattering layer comprising a glass which contains a base material (121) having a first refractive index for at least one wavelength of light to be transmitted, the scattering layer further comprising a plurality of scattering materials (122) dispersed in the base material (121) and having a second refractive index different from that of the base material,
- a layer of transparent electrode material (130) formed over the scattering layer (120),

characterized in that the electrode is made of a material comprising fluorine-doped tin oxide.

2. A translucent conductive substrate as claimed in claim 1, wherein the scattering layer comprises a glass comprising the following oxides in the following amounts represented in mole percentage : 26% to 43% of B_2O_3, 30% to 37% of ZnO, 17% to 23% of Bi_2O_3, 2% to 21% of SiO_2, and 0 to 2% of P_2O_5; and a total B_2O_3-content and ZnO-content of less than 78%.

3. A translucent conductive substrate as claimed in claim 1 or 2, wherein the glass of the scattering layer includes substantially no alkaline metal oxides, in particular substantially no Li_2O, Na_2O, and K_2O), no lead oxides, in particular no PbO, Pb_3O_4, nor P_2O_5, except for being included as an impurity.

4. A translucent conductive substrate as claimed in any one of claims 1-3, wherein the electrode surface an average surface roughness Ra as

determined using atomic force microscopy of at least 7.5 nm, preferably at least 8, more preferably at least 8.5 or 9, often at least 9.4, as determined by atomic force microscopy, preferably at least 10 nm, more preferably at least 12 nm, most preferably at least 12.5 nm.

5. A translucent conductive substrate as claimed in any one of the previous claims, wherein the substrate has a sheet resistance of less than 20 Ω/□, preferably less than 15 Ω/□.

6. A translucent conductive substrate as claimed in any one of the previous claims, wherein, the atomic ratio F/Sn in the F-doped SnO_2 varies from 0.005 to 0.20, preferably from 0.01 to 0.10, more preferably from 0.01 to 0.05.

7. A translucent conductive substrate according to any one of the previous claims, wherein a face of the scattering layer facing the transparent electrode material, is coated with a layer of a smoothening material, preferably a material selected from the group of titanium oxide, niobium oxide, zirconium oxide, tantalum oxide and silicon oxide or a mixture of two or more of these materials, preferably a mixed oxide of silicon and titanium.

8. An organic light emitting device comprising a translucent conductive substrate according to any one of claims 1-7, further comprising an organic layer positioned on top of the fluorine-doped tin oxide anode and a second electrode layer positioned on top of the second electrode.

9. A process for producing a conductive translucent substrate for use in an organic light emitting diode, wherein a transparent electrode material is deposited on top of a scattering layer deposited on the transparent support, wherein the scattering layer comprises a base material having a first refractive index at a wavelength of emitted light of an organic LED element and a plurality of scattering materials provided in the inside of the base material and

having a refractive index different from that of the base material, wherein the scattering material is deposited on the transparent support by deposition of a frit paste and heating the thus coated support to dry and fire the frit paste to form the scattering layer, characterised in that as the electrode material use is made of F-doped SnO_2, in that the electrode material is deposited by deposition and decomposition of a precursor at a temperature of between 500 and 650°C, preferably between 500 and 625°C, more preferably between 520-600°C followed by cooling of the thus obtained conductive substrate.

10. A process according to claim 9, wherein the scattering material is dried and fired in advance of depositing the F-doped SnO_2 electrode material, by heating the frit paste coated transparent support preferably to a temperature of between 520-580°C in advance of depositing the precursor for the F-doped SnO_2 electrode material.

11. A process according to claim 9, wherein the scattering material is fired simultaneously with the deposition of F-doped SnO_2 electrode material, wherein the frit coated transparent support is heated to a temperature of at least 500°C, preferably at least 520°C, more preferably at least 540°C,more preferably at least 560°C, most preferably a temperature between 560-600°C, to dry the frit paste, after which the precursor for the F-doped SnO_2 is deposited on top of the dried frit paste and subjected to heating to decompose the precursor.

12. A process according to any one of claims 9-11, wherein the substrate is progressively cooled after deposition of the F-doped SnO_2 electrode material.

13. A process according to any one of claims 9-12, wherein the F-doped SnO_2 electrode material is deposited by chemical vapour deposition of a precursor, in particular monobutyltintrichloride.

14. A process according to claim 13, wherein deposition of the F-doped SnO_2 electrode material takes place in the presence of H_2O, preferably in a ratio of H_2O/MBTC of maximum 20, preferably maximum 16, more preferably maximum 10, and at least 1, preferably at least 3, more preferably about 5.

15. A process according to any one of claims 9-14, wherein deposition of the F-doped SnO_2 electrode material takes place in the presence of O_2 in a ratio O_2/MBTC of less than 25, preferably less than 20.

Fig. 1

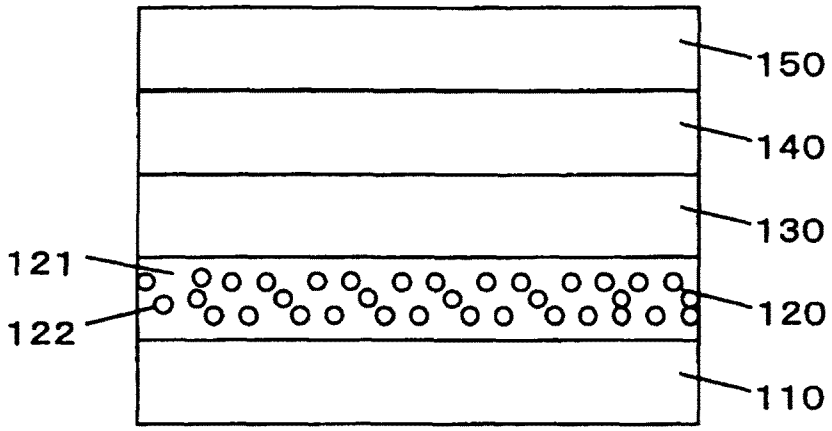
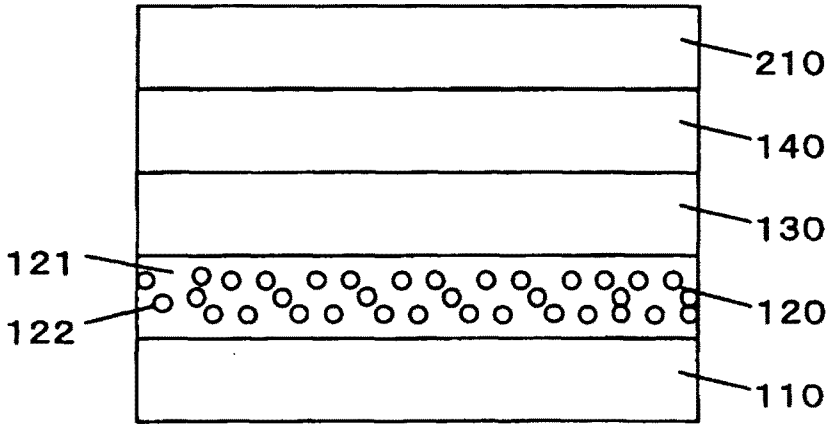


Fig. 2



INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2015/080651

A. CLASSIFICATION OF SUBJECT MATTER
INV. H01L51/00 C03C3/064 C03C3/066 C03C8/04 C03C8/08
G02B5/02 H01B1/22 H01L31/0224 H01L51/52
ADD.
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
H01L C03C G02B H01B

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)
EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 2010/031688 A1 (SIEMENS AG [DE]; BERNDT ANETT [DE]; EDER FLORIAN [DE]; SARFERT WIEBKE) 25 March 2010 (2010-03-25)	1,4,5,7, 8
Y	page 11, line 24 - line 26; claims 1-9, 12-14; figure 1	2,3,6

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[X] Further documents are listed in the continuation of Box C.

[X] See patent family annex.

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15 February 2016	23/02/2016

Name and mailing address of the ISA/	Authorized officer
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1

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INTERNATIONAL SEARCH REPORT

International application No
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C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

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Y	DAVINDER S. BHACHU ET AL: "Textured Fluorine-Doped Tin Dioxide Films formed by Chemical Vapour Deposition", CHEMISTRY - A EUROPEAN JOURNAL, vol. 17, no. 41, 4 October 2011 (2011-10-04), pages 11613-11621, XP055194774, ISSN: 0947-6539, DOI: 10.1002/chem.201100399 Experimental section; page 11614, column 1 - line 18 -----	6,9-15

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Information on patent family members

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