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(54) **CASCADED ORGANIC
ELECTROLUMINESCENT DEVICES WITH
COLOR FILTERS**

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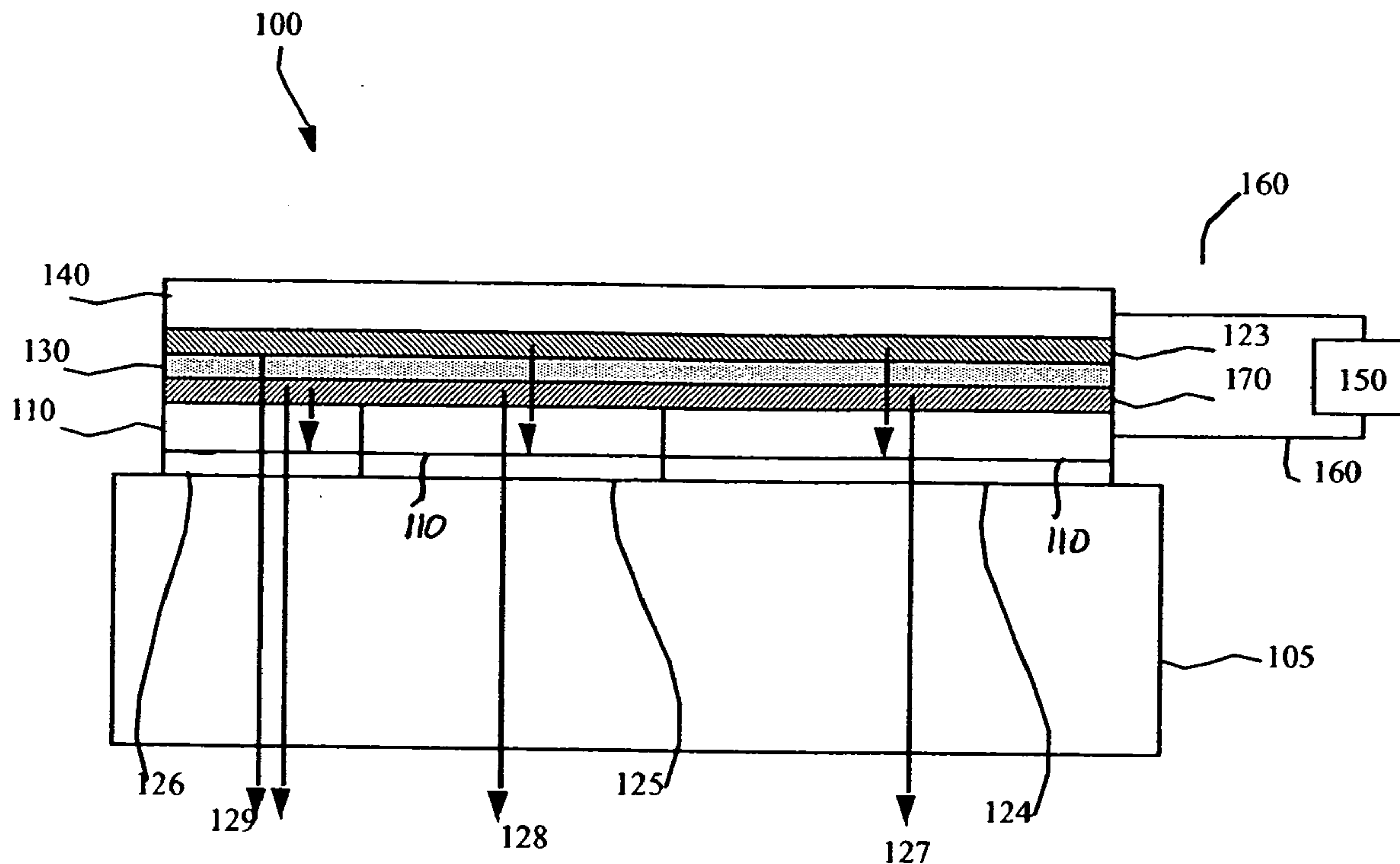
(57) **ABSTRACT**

An OLED device is described comprising a substrate having thereon a cascaded organic electroluminescent device comprising: a) an anode; b) a cathode; c) a plurality of cascaded organic electroluminescent units disposed between the anode and the cathode, wherein each organic electroluminescent unit includes at least one light-emitting layer and wherein the plurality of cascaded units includes at least two units that emit light of different colors; and d) a connecting unit disposed between each adjacent cascaded organic electroluminescent unit; and e) a colored filter that filters the emitted light.

(73) Assignee: **Eastman Kodak Company**

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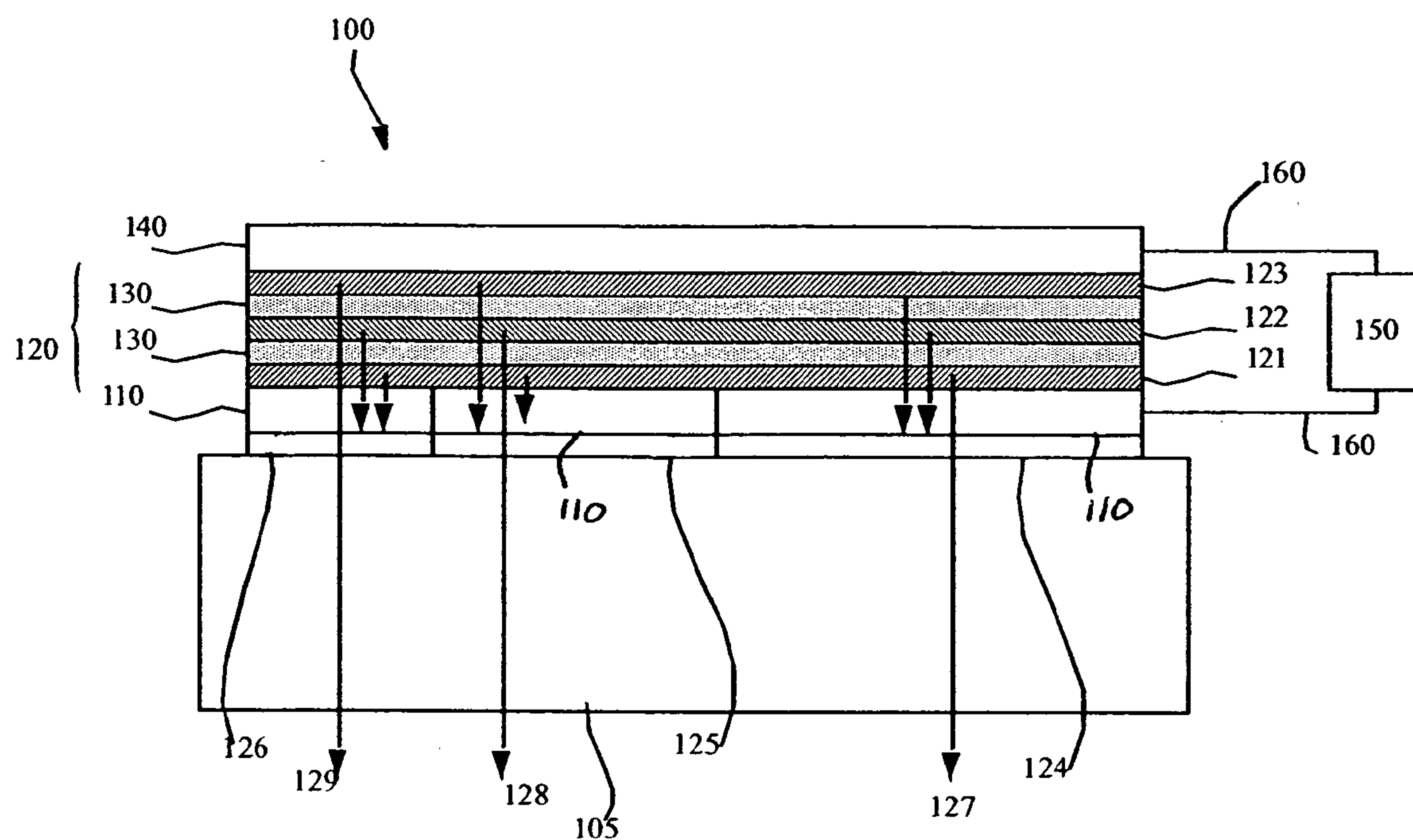


Fig. 1

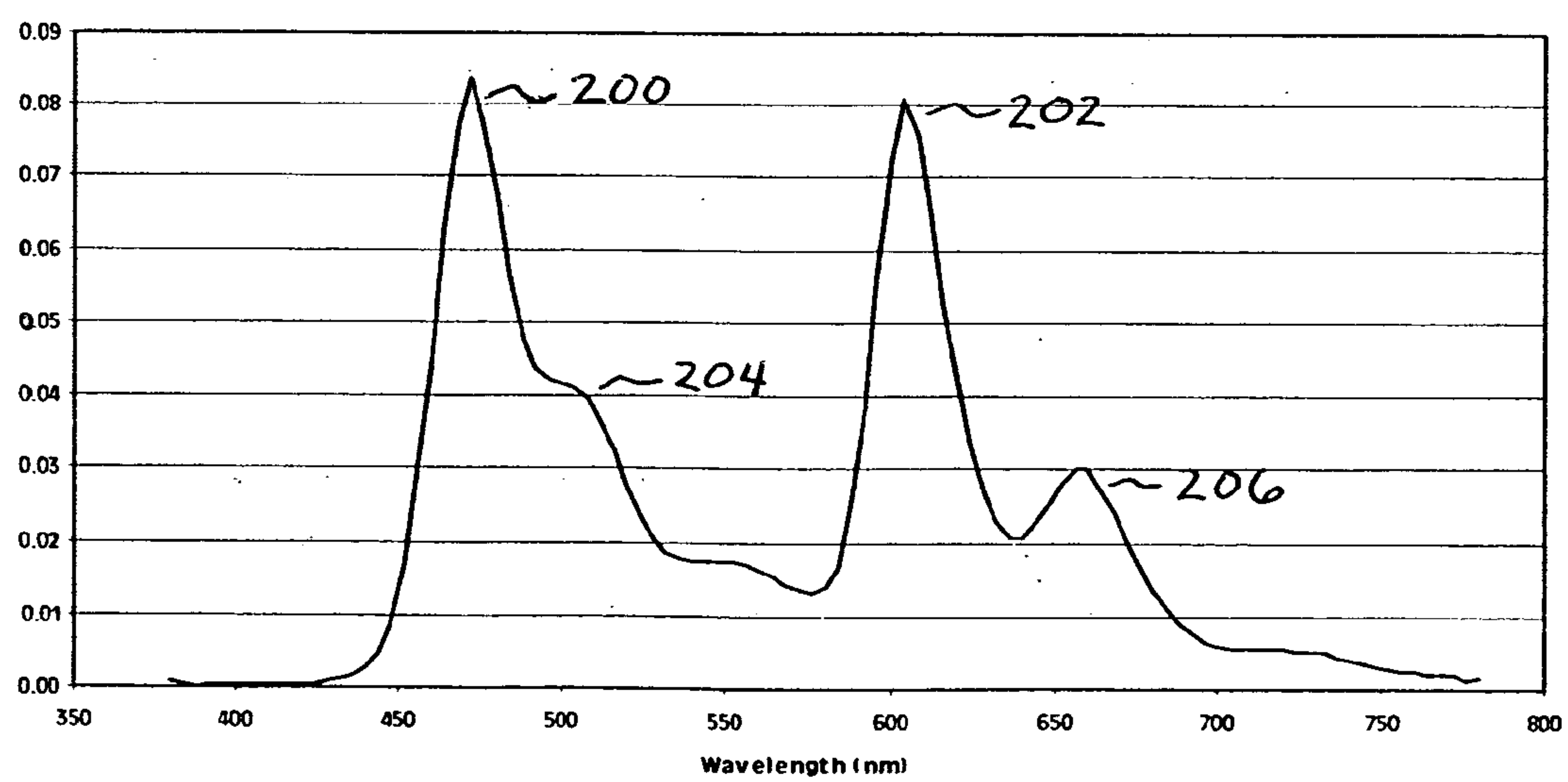


Fig. 2

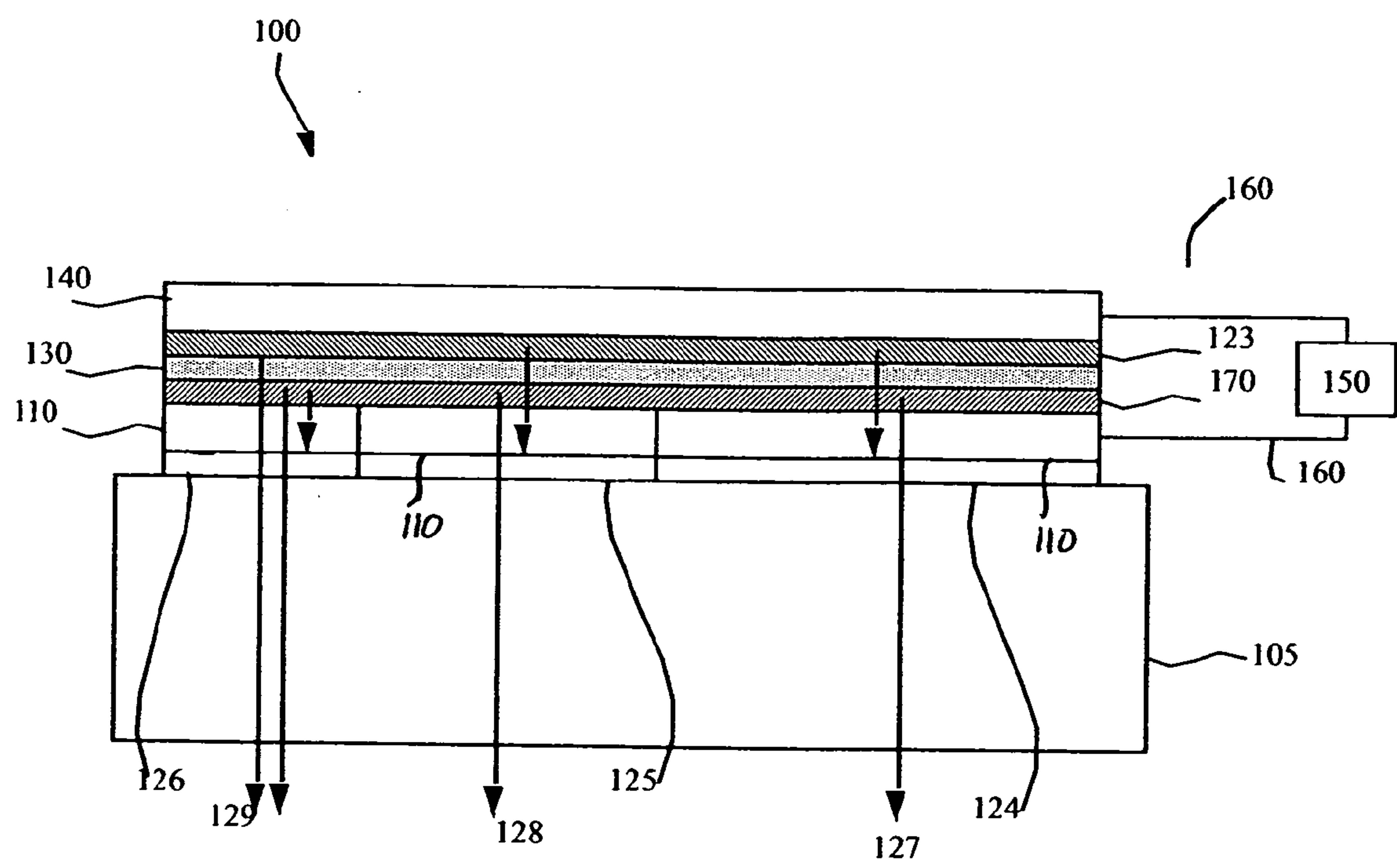


Fig. 3

CASCADED ORGANIC ELECTROLUMINESCENT DEVICES WITH COLOR FILTERS

CROSS-REFERENCE TO RELATED APPLICATION

[0001] This application is a continuation-in-part of copending, commonly assigned U.S. patent application Ser. No. 10/077,270 filed Feb. 15, 2002 by Liang-Sheng L. Liao et al., entitled "Providing an Organic Electroluminescent Device Having Stacked Electroluminescent Units", the disclosure of which is herein incorporated by reference. Reference is also made to copending, commonly assigned U.S. Ser. Nos. 10/437,195 and 10/845,038, the disclosures of which are also incorporated by reference herein.

FIELD OF THE INVENTION

[0002] The present invention relates to providing a OLED device having a plurality of organic electroluminescent (EL) units in the form of a cascaded organic electroluminescent device with color filters.

BACKGROUND OF THE INVENTION

[0003] Organic electroluminescent (EL), or organic light-emitting diode (OLED), devices are electronic devices that emit light in response to an applied potential. The structure of an OLED comprises, in sequence, an anode, an organic EL medium, and a cathode. The organic EL medium disposed between the anode and the cathode is commonly comprised of an organic hole-transporting layer (HTL) and an organic electron-transporting layer (ETL). Holes and electrons recombine and emit light in the ETL near the interface of HTL/ETL. Tang et al., "Organic electroluminescent diodes", *Applied Physics Letters*, 51, 913 (1987), and commonly assigned U.S. Pat. No. 4,769,292, demonstrated highly efficient OLEDs using such a layer structure. Since then, numerous OLEDs with alternative layer structures have been disclosed. For example, there are three-layer OLEDs that contain an organic light-emitting layer (LEL) between the HTL and the ETL, such as that disclosed by Adachi et al., "Electroluminescence in Organic Films with Three-Layer Structure", *Japanese Journal of Applied Physics*, 27, L269 (1988), and by Tang et al., "Electroluminescence of doped organic thin films", *Journal of Applied Physics*, 65, 3610 (1989). The LEL commonly includes of a host material doped with a guest material wherein the layer structures are denoted as HTL/LEL/ETL. Further, there are other multilayer OLEDs that contain a hole-injecting layer (HIL), and/or an electron-injecting layer (EIL), and/or a hole-blocking layer, and/or an electron-blocking layer in the devices. These structures have further resulted in improved device performance.

[0004] Color, digital image display devices are well known and are based upon a variety of technologies such as cathode ray tubes, liquid crystal and solid-state light emitters such as Organic Light Emitting Diodes (OLEDs). In a common OLED color display device a pixel includes red, green, and blue colored OLEDs. By combining the illumination from each of these three OLEDs in an additive color system, a full-color display having a wide variety of colors can be achieved.

[0005] OLEDs may be used to generate color directly using organic materials that are doped to emit energy in

desired portions of the electromagnetic spectrum. However, to create a color OLED device using different organic materials requires the patterning of these materials over the surface of the OLED device substrate. This patterning is a difficult and problematic task, especially for large substrates, for example substrates having a diagonal greater than about 50 cm. An alternative method utilizes a white-light emitting material in combination with color filter arrays to provide color emission, much as conventional LCD displays do. White-light emitting OLED devices that are known in the art are typically formed by doping multiple, individual emitting layers such that each doped layer produces light within a specific spectral frequency band. White-light emitting devices may be formed from either two or three individual emitting materials. However, this approach suffers from efficiency problems because the white light emitters tend to be relatively broadband so that most of the light emitted by the white-light emitter is absorbed by the color filters, reducing the efficiency of the OLED device. Additionally, some of the white light emitters may age more rapidly than other due to the relative efficiencies for which they emit different colors.

[0006] In order to further improve the performance of the OLEDs, a new kind of OLED structure called stacked OLED, which is fabricated by stacking several individual OLED vertically, has also been proposed. Forrest et al. in U.S. Pat. No. 5,703,436 and Burrows et al. in U.S. Pat. No. 6,274,980 disclosed their stacked OLEDs. In their inventions, the stacked OLEDs are fabricated by vertically stacking several OLEDs, each independently emitting light of a different color or of the same color. Using their stacked OLED structure can make full color emission devices with higher integrated density in the display, but each OLED needs a separate power source. In an alternative design, Jones et al. in U.S. Pat. No. 6,337,492 proposed a stacked OLED structure by vertically stacking several OLED without individually addressing each OLED in the stack. Jones et al. believe that their stacked structure could increase the luminance output and operational lifetime. These OLEDs use individual OLEDs (anode/organic medium/cathode) as building blocks to fabricate the stacked OLEDs. The complex architecture in these designs presents serious fabrication problems for patterned multi-colored display devices. Moreover, these designs do not address inefficiencies of white light emitters in combination with color filters, or the consequent differential aging associated with differential efficiencies. This reduces the overall device efficiency.

[0007] There is a need therefore, for an improved OLED device structure providing simplicity of manufacture with efficiency of operation.

SUMMARY OF THE INVENTION

[0008] In accordance with one embodiment, the present invention is directed towards an OLED device comprising a substrate having thereon a cascaded organic electroluminescent device comprising: a) an anode; b) a cathode; c) a plurality of cascaded organic electroluminescent units disposed between the anode and the cathode, wherein each organic electroluminescent unit includes at least one light-emitting layer and wherein the plurality of cascaded units includes at least two units that emit light of different colors; and d) a connecting unit disposed between each adjacent cascaded organic electroluminescent unit; and e) a colored filter that filters the emitted light.

ADVANTAGEOUS EFFECT OF THE INVENTION

[0009] Various embodiments of the present invention provide color OLED devices with simplified manufacturing, improved efficiency, and reduced aging.

BRIEF DESCRIPTION OF THE DRAWINGS

[0010] FIG. 1 depicts a schematic cross sectional view of a cascaded OLED device according to the present invention;

[0011] FIG. 2 depicts the emission spectrum of a white-light emitting OLED; and

[0012] FIG. 3 depicts a schematic cross sectional view of an alternative cascaded OLED device according to the present invention.

[0013] It will be understood that FIGS. 1 and 3 are not to scale since the individual layers are too thin and the thickness differences of various layers too great to permit depiction to scale.

DETAILED DESCRIPTION OF THE INVENTION

[0014] The layer structure of a cascaded OLED (or stacked OLED) comprises an anode, a cathode, a plurality of organic EL units and a plurality of organic connectors (or connecting units thereafter), wherein each of the connecting units is disposed between two organic EL units. The organic EL unit includes at least one light-emitting layer, and typically comprises, in sequence, a hole-transport layer, a light-emitting layer, and an electron-transport layer, denoted in brief as HTL/LEL/ETL.

[0015] To function efficiently, the connecting unit for the cascaded OLED should provide electron injection into the electron-transporting layer and hole injection into the hole-transporting layer of the two adjacent organic EL units. A variety of materials may be used to form the connecting units. In preferred embodiments, connecting unit materials are selected to provide high optical transparency and excellent charge injection, thereby providing the cascaded OLED high electroluminescence efficiency and operation at an overall low driving voltage.

[0016] The connecting unit may comprise doped organic connectors provided between adjacent organic EL units. Each doped organic connector may include at least one n-type doped organic layer, or at least one p-type doped organic layer, or a combination of layers, thereof. Preferably, the doped organic connector includes both an n-type doped organic layer and a p-type doped organic layer disposed adjacent to one another to form a p-n heterojunction. It is also preferred that the n-type doped organic layer is disposed towards the anode side, and the p-type doped organic layer is disposed towards the cathode side. The choice of using n-type doped organic layer, or a p-type doped organic layer, or both (the p-n junction) is in part dependent on the organic materials that include the organic EL units. Each connector can be optimized to yield the best performance with a particular set of organic EL units. This includes choice of materials, layer thickness, modes of deposition, and so forth.

[0017] An n-type doped organic layer means that the organic layer has semiconducting properties after doping, and the electrical current through this layer is substantially

carried by the electrons. A p-type doped organic layer means that the organic layer has semiconducting properties after doping, and the electrical current through this layer is substantially carried by the holes. A p-n heterojunction means an interfacial region (or junction) formed when a p-type layer and an n-type layer contact each other.

[0018] N-type doped organic layers may include a host organic material and at least one n-type dopant. The host material in the n-typed doped organic layer can include a small molecule material or a polymeric material, or combinations thereof, and it is preferred that it can support electron transport. The p-type doped organic layers may include a host organic material and at least one p-type dopant. The host material can include a small molecule material or a polymeric material, or combinations thereof, and it is preferred that it can support hole transport. In some instances, the same host material can be used for both n-typed and p-type doped organic layers, provided that it exhibits both hole and electron transport properties set forth above. The n-type doped concentration or the p-type doped concentration is preferably in the range of 0.01-10 vol. %. The total thickness of each doped organic connector is typically less than 100 nm, and preferably in the range of about 1 to 100 nm.

[0019] The organic electron-transporting materials used in conventional OLED devices represent a useful class of host materials that may be employed for the n-type doped organic layer. Preferred materials are metal chelated oxinoid compounds, including chelates of oxine itself (also commonly referred to as 8-quinolinol or 8-hydroxyquinoline), such as tris(8-hydroxyquinoline) aluminum. Other materials include various butadiene derivatives as disclosed by Tang (U.S. Pat. No. 4,356,429), various heterocyclic optical brighteners as disclosed by Van Slyke and Tang and others (U.S. Pat. No. 4,539,507), triazines, hydroxyquinoline derivatives, and benzazole derivatives. Silole derivatives, such as 2,5-bis(2', 2''-bipyridin-6-yl)-1,1-dimethyl-3,4-diphenyl silacyclopentadiene as reported by Murata and others [Applied Physics Letters, 80, 189 (2002)], are also useful host materials.

[0020] Materials useful as n-type dopants in the n-type doped organic layer of a doped organic connector include metals or metal compounds having a work function less than 4.0 eV. Particularly useful dopants include alkali metals, alkali metal compounds, alkaline earth metals, and alkaline metal compounds. The term "metal compounds" includes organometallic complexes, metal-organic salts, and inorganic salts, oxides and halides. Among the class of metal-containing n-type dopants, Li, Na, K, Rb, Cs, Mg, Ca, Sr, Ba, La, Ce, Sm, Eu, Th, Dy, or Yb, and their compounds, are particularly useful. Materials useful as n-type dopants in the n-type doped organic layer of a doped organic connector also include organic reducing agents with strong electron-donating properties. By "strong electron-donating properties" we mean that the organic dopant should be able to donate at least some electronic charge to the host to form a charge-transfer complex with the host. Non-limiting examples of organic molecules include bis(ethylenedithio)-tetrathiafulvalene (BEDT-TTF), tetrathiafulvalene (TTF), and their derivatives. In the case of polymeric hosts, the dopant can be any of the above or also a material molecularly dispersed or copolymerized with the host as a minor component.

[0021] The hole-transporting materials used in conventional OLED devices represent a useful class of host materials for p-type doped organic layers. Preferred materials include aromatic tertiary amines having at least one trivalent nitrogen atom that is bonded only to carbon atoms, at least one of which is a member of an aromatic ring. In one form the aromatic tertiary amine can be an arylamine, such as a monoarylamine, diarylamine, triarylamine, or a polymeric arylamine. Other suitable triarylaminines substituted with one or more vinyl radicals and/or comprising at least one active hydrogen-containing group are disclosed by Brantley and others (U.S. Pat. No. 3,567,450 and U.S. Pat. No. 3,658,520). A more preferred class of aromatic tertiary amines are those which include at least two aromatic tertiary amine moieties as described by Van Slyke and Tang and others (U.S. Pat. No. 4,720,432 and U.S. Pat. No. 5,061,569). Non-limiting examples include as N,N'-di(naphthalene-1-yl)-N,N'-diphenyl-benzidine (NPB) and N,N'-diphenyl-N,N'-bis(3-methylphenyl)-1,1'-biphenyl-4,4'-diamine (TPD), and N,N,N',N'-tetranaphthyl-benzidine (TNB).

[0022] Materials useful as p-type dopants in p-type doped organic layers of doped organic connectors include oxidizing agents with strong electron-withdrawing properties. By "strong electron-withdrawing properties" we mean that the organic dopant should be able to accept some electronic charge from the host to form a charge-transfer complex with the host. Some non-limiting examples include organic compounds such as 2,3,5,6-tetrafluoro-7,7,8,8-tetracyanoquinodimethane (F_4 -TCNQ) and other derivatives of TCNQ, and inorganic oxidizing agents such as iodine, $FeCl_3$, $SbCl_5$, and some other metal chlorides. In the case of polymeric hosts, the dopant can be any of the above or also a material molecularly dispersed or copolymerized with the host as a minor component.

[0023] Examples of materials that can be used as host for either n-type or p-type doped organic layers include, but are not limited to: various anthracene derivatives including those described in U.S. Pat. No. 5,972,247; certain carbazole derivatives, such as 4,4-bis(9-dicarbazolyl)-biphenyl (CBP); and distyrylarylene derivatives such as 4,4'-bis(2,2'-diphenyl vinyl)-1,1'-biphenyl and as described in U.S. Pat. No. 5,121,029.

[0024] The materials used for fabricating doped organic connectors are preferably substantially transparent to emitted light.

[0025] In a preferred embodiment, the connecting unit comprises, in sequence, an n-type doped organic layer and a p-type doped organic layer. Thus, in this structure, the ETL of the EL unit is adjacent to the n-type doped layer of the connecting unit and the HTL of the EL unit is adjacent to the p-type doped connecting unit. In this cascaded device structure only a single external power source is needed to connect to the anode and the cathode with the positive potential applied to the anode and the negative potential to the cathode. No other electrical connections are needed to connect the individual organic EL units to external electrical power sources.

[0026] In a further specific cascaded OLED device embodiment, the physical spacing between adjacent electroluminescent zones may be more than 90 nm and the connecting unit disposed between each adjacent organic electroluminescent unit may comprise an n-type doped

organic layer and a p-type doped organic layer forming a transparent p-n junction structure wherein the resistivity of each of the doped layers is higher than 10 Ω -cm, as described in commonly assigned U.S. patent application Ser. No. 10/437,195 filed May 13, 2003 entitled "Cascaded Organic Electroluminescent Device Having Connecting Units with n-Type and p-Type Organic Layers", the disclosure of which is herein incorporated by reference.

[0027] For a cascaded OLED to function efficiently, it is necessary that the optical transparency of the layers constituting the organic EL units and the connecting units be as high as possible to allow for radiation generated in the organic EL units to exit the device. Furthermore, for the radiation to exit through the anode, the anode should be transparent and the cathode can be opaque, reflecting, or transparent. For the radiation to exit through the cathode, the cathode should be transparent and the anode can be opaque, reflecting or transparent. The layers constituting the organic EL units are generally optically transparent to the radiation generated by the EL units, and therefore their transparency is generally not a concern for the construction for the cascaded OLEDs.

[0028] The operational stability of cascaded OLED is dependent to a large extent on the stability of the connecting units. In particular, the driving voltage will be highly dependent on whether or not the connecting unit can provide the necessary electron and hole injection. It is generally known that the close proximity of two dissimilar materials may result in diffusion of matters from one into another, or in interdiffusion of matters across the boundary between the two. In the case of cascaded OLEDs employing an n-type doped organic layer and a p-type doped organic layer, if such diffusion were to occur in the connecting unit between the n-type doped organic layer and the p-type doped organic layer, the injection properties of this organic connecting unit may degrade correspondingly due to the fact that the individual n-type doped layer or p-type doped layer may no longer be sufficiently electrically conductive. Diffusion or interdiffusion is dependent on temperature as well as other factors such as electrical field induced migration. The latter is plausible in cascaded OLED devices since the operation of OLED generally requires an electric field as high as 10^6 volt per centimeter. To prevent such an operationally induced diffusion in the connecting units of a cascaded OLED, an interfacial layer which provides a barrier for interdiffusion may be introduced in between the n-type doped layer and the p-type doped layer, as described in U.S. Pat. No. 6,717,358, the disclosure of which is incorporated herein by reference.

[0029] Interfacial layers useful in the connecting unit may comprise at least one inorganic semiconducting material or combinations of more than one of the semiconducting materials. Suitable semiconducting materials should have an electron energy band gap less than 4.0 eV. The electron energy band gap is defined as the energy difference between the highest occupied molecular orbital and the lowest unoccupied molecular orbital of the molecule. A useful class of materials can be chosen from the compounds of elements listed in groups IVA, VA, VIA, VIIA, VIIIA, IB, IIB, IIIB, IVB, and VB in the Periodic Table of the Elements (e.g. the Periodic Table of the Elements published by VWR Scientific Products). These compounds include the carbides, silicides, nitrides, phosphides, arsenides, oxides, sulfides, selenides,

and tellurides, and mixture thereof. These semiconducting compounds can be in either stoichiometric or non-stoichiometric states, that is they may contain excess or deficit metal component. Particularly useful materials for the interfacial layer are the semiconducting oxides of titanium, zirconium, hafnium, vanadium, niobium, tantalum, chromium, molybdenum, tungsten, manganese, rhenium, iron, ruthenium, osmium, cobalt, rhodium, iridium, nickel, palladium, platinum, copper, zinc, cadmium, gallium, thallium, silicon, germanium, lead, and antimony, or combinations thereof. Particularly useful materials for the interfacial layer also including zinc selenide, gallium nitride, silicon carbide, or combinations thereof.

[0030] The interfacial layer useful in a connecting unit also can comprise at least one or more metallic materials, where at least one of these metallic materials has a work-function higher than 4.0 eV as listed by Sze, in *Physics of Semiconducting Devices*, 2nd Edition, Wiley, N.Y., 1981, p. 251. The thickness of an interfacial layer suitable for the construction of a connecting unit is preferably in the range of 0.05 nm to 10 μ m, more preferably between 0.1 nm to 5 nm for inorganic semiconducting materials and between 0.05 nm to 1 nm for metallic materials.

[0031] In a further embodiment, the connecting unit disposed between each adjacent organic electroluminescent unit in the cascaded device may include at least a high work function metal layer having a work function of no less than 4.0 eV and a metal compound layer, wherein the intermediate connector has a sheet resistance of higher than 100 k Ω per square, such as described in copending, commonly assigned U.S. Ser. No. 10/857,516, filed May 28, 2004, the disclosure of which is incorporated herein by reference. The use of such high work function metal layer in a connecting unit of a cascaded OLED device improves the operational stability of the OLED.

[0032] As discussed above, for a cascaded OLED to function efficiently, it is necessary that the intermediate connector should provide good carrier injection into the adjacent organic EL units. Due to their lower resistivity than that of organic materials, metals, metal compounds, or other inorganic compounds can be good for carrier injection. However, low resistivity can cause low sheet resistance resulting in pixel crosstalk. If the lateral current passing through the adjacent pixels to cause pixel crosstalk is limited to less than 10% of the current used to drive a pixel, the lateral resistance of the intermediate connector (R_{ic}) should be at least 8 times the resistance of the cascaded OLED. Usually, the static resistance between two electrodes of a conventional OLED is about several k Ω s, and a cascaded OLED should have a resistance of about 10 k Ω or several 10 k Ω s between the two electrodes. Therefore R_{ic} should be greater than 100 k Ω . Considering the space between each pixel is smaller than one square, the sheet resistance of the intermediate connector should be then greater than 100 k Ω per square (lateral resistance equals to sheet resistance times the number of square). Because the sheet resistance is determined by both the resistivity and the thickness of the films (sheet resistance equals to film resistivity divided by film thickness), when the layers constituting an intermediate connector are selected from metals, metal compounds, or other inorganic compounds having low resistivity, a sheet

resistance of the intermediate connector greater than 100 k Ω per square can still be achievable if the layers are thin enough.

[0033] Another requirement for the tandem OLED to function efficiently is that the optical transparency of the layers constituting the organic EL units and the intermediate connectors be as high as possible to permit for radiation produced in the organic EL units to exit the device. According to a simple calculation, if the optical transmission of each intermediate connector is 70% of the emitting light, a tandem OLED will not have much benefit because no matter how many EL units there are in the device, the electroluminescence efficiency can never be doubled when comparing to a conventional device. The layers constituting the organic EL units are generally optically transparent to the radiation produced by the EL units, and therefore their transparency is generally not a concern for the construction of the tandem OLEDs. As is known, metals, metal compounds, or other inorganic compounds can have low transparency. However, when the layers constituting an intermediate connector are selected from the metals, metal compounds, or other inorganic compounds, an optical transmission higher than 70% can still be achievable if the layers are thin enough. Preferably, the intermediate connector has at least 75% optical transmission in the visible region of the spectrum.

[0034] In accordance with one specific embodiment, the intermediate connectors may comprise, in sequence, a low work function metal layer, a high work function metal layer, and a metal compound layer. Herein, a low work function metal is defined as a metal having a work function less than 4.0 eV. Likewise, a high work function metal is defined as a metal having a work function no less than 4.0 eV. The low work function metal layer is preferably disposed adjacent to the ETL of an organic EL unit towards the anode side, and the metal compound layer is preferably disposed adjacent to the HTL of another organic EL unit towards the cathode side. The low work function metal layer may be selected to provide efficient electron injection into the adjacent electron-transporting layer. The metal compound layer may be selected to provide efficient hole injection into the adjacent hole-transporting layer. Preferably, the metal compound layer comprises, but is not limited to, a p-type semiconductor. The high work function metal layer is selected to improve the operational stability of the OLED by preventing a possible interaction or interdiffusion between the low work function layer and the metal compound layer.

[0035] In accordance with another specific embodiment, the intermediate connectors may comprise, in sequence, an n-type semiconductor layer, a high work function metal layer, and a metal compound layer. The n-type semiconductor layer is preferably disposed adjacent to the ETL of an organic EL unit towards the anode side, and the metal compound layer is preferably disposed adjacent to the HTL of another organic EL unit towards the cathode side. Herein, an n-type semiconductor layer means that the layer is electrically conductive having electrons as the major charge carriers. Likewise, a p-type semiconductor layer means that the layer is electrically conductive having holes as the major charge carriers. Similar to a low work function metal layer, the n-type semiconductor layer may be selected to provide efficient electron injection into the adjacent electron-transporting layer. The metal compound layer again may be selected to provide efficient hole injection into the adjacent

hole-transporting layer, and the high work function metal layer is selected to improve the operational stability of the OLED by preventing a possible interaction or interdiffusion between the n-type semiconductor layer and the metal compound layer.

[0036] In the case such that the ETL in the EL unit is an n-type doped organic layer, the layer structure of the intermediate connector can be simplified by comprising, in sequence, a high work function metal layer disposed adjacent to the n-type doped ETL of an organic EL unit towards the anode side, and a metal compound layer disposed adjacent to the HTL of another organic EL unit towards the cathode side. The metal compound layer may be selected to provide efficient hole injection into the adjacent hole-transporting layer, and the high work function metal layer is selected to improve the operational stability of the OLED by preventing a possible interaction or interdiffusion between the n-type doped ETL and the metal compound layer. Herein, an n-type doped organic layer means that the layer is electrically conductive, and the charge carriers are primarily electrons. The conductivity is provided by the formation of a charge-transfer complex as a result of electron transfer from the dopant to the host material. Depending on the concentration and the effectiveness of the dopant in donating an electron to the host material, the layer electrical conductivity can change by several orders of magnitude. With an n-type doped organic layer as an ETL in the EL unit, electrons can be efficiently injected from the adjacent intermediate connector into the ETL.

[0037] In order for the intermediate connectors to have good optical transmission (at least 75% optical transmission in the visible region of the spectrum), good carrier injection capability, and good operational stability, the thickness of the layers in the intermediate connectors has to be carefully considered. The thickness of the low work function metal layer, when employed, in the intermediate connectors is preferably in the range of from 0.1 nm to 5.0 nm, more preferably in the range of from 0.2 nm to 2.0 nm. The thickness of the high work function metal layer, when employed, in the intermediate connectors is preferably in the range of from 0.1 nm to 5.0 nm, more preferably in the range of from 0.2 nm to 2.0 nm. The thickness of the metal compound layer, when employed, in the intermediate connectors is preferably in the range of from 0.5 nm to 20 nm, more preferably in the range of from 1.0 nm to 5.0 nm. The thickness of the n-type semiconductor layer, when employed, in the intermediate connectors is preferably in the range of from 0.5 nm to 20 nm, more preferably in the range of from 1.0 nm to 5.0 nm.

[0038] The materials used for the fabrication of intermediate connectors are basically selected from nontoxic materials. Low work function metal layers may include, e.g., Li, Na, K, Rb, Cs, Mg, Ca, Sr, Ba, La, Ce, Nd, Sm, Eu, Th, Dy, or Yb. Preferably, the low work function metal layer includes Li, Na, Cs, Ca, Ba, or Yb. High work function metal layers may include, e.g., Ti, Zr, Hf, Nb, Ta, Cr, Mo, W, Re, Fe, Ru, Os, Co, Rh, Ir, Ni, Pd, Pt, Cu, Ag, Au, Zn, Al, In, or Sn. Preferably, the high work function metal layer includes Ag, Al, Cu, Au, Zn, In, or Sn. More preferably, the high work function metal layer includes Ag or Al.

[0039] The metal compound layer, when employed, can be selected from the stoichiometric oxides or nonstoichiometric

oxides of titanium, zirconium, hafnium, niobium, tantalum, molybdenum, tungsten, manganese, iron, ruthenium, rhodium, iridium, nickel, palladium, platinum, copper, zinc, silicon, or germanium, or combinations thereof. The metal compound layer can be selected from the stoichiometric sulfides or nonstoichiometric sulfides of titanium, zirconium, hafnium, niobium, tantalum, molybdenum, tungsten, manganese, iron, ruthenium, rhodium, iridium, nickel, palladium, platinum, copper, silicon, or germanium, or combinations thereof. The metal compound layer can be selected from the stoichiometric selenides or nonstoichiometric selenides of titanium, zirconium, hafnium, niobium, tantalum, molybdenum, tungsten, manganese, iron, ruthenium, rhodium, iridium, nickel, palladium, platinum, copper, silicon, or germanium, or combinations thereof. The metal compound layer can be selected from the stoichiometric tellurides or nonstoichiometric tellurides of titanium, zirconium, hafnium, niobium, tantalum, molybdenum, tungsten, manganese, iron, ruthenium, rhodium, iridium, nickel, palladium, platinum, copper, silicon, or germanium, or combinations thereof. The metal compound layer can be selected from the stoichiometric nitrides or nonstoichiometric nitrides of titanium, zirconium, hafnium, niobium, tantalum, molybdenum, tungsten, manganese, iron, ruthenium, rhodium, iridium, nickel, palladium, platinum, copper, zinc, gallium, silicon, or germanium, or combinations thereof. The metal compound layer can also be selected from the stoichiometric carbides or nonstoichiometric carbides of titanium, zirconium, hafnium, niobium, tantalum, molybdenum, tungsten, manganese, iron, ruthenium, rhodium, iridium, nickel, palladium, platinum, copper, zinc, aluminum, silicon, or germanium, or combinations thereof. The metal compound layer can be selected from MoO_3 , NiMoO_4 , CuMoO_4 , WO_3 , ZnTe , Al_4C_3 , AlF_3 , B_2S_3 , CuS , GaP , InP , or SnTe . Preferably, the metal compound layer is selected from MoO_3 , NiMoO_4 , CuMoO_4 , or WO_3 .

[0040] The n-type semiconductor layer, when employed, may include, e.g., ZnSe , ZnS , ZnSSe , SnSe , SnS , SnSSe , LaCuO_3 , or $\text{La}_4\text{Ru}_6\text{O}_{19}$. Preferably, the n-type semiconductor layer includes ZnSe or ZnS .

[0041] Other intermediate connector materials may also be employed in the OLED cascaded devices of the present invention. For example, Tanaka et al., U.S. Pat. No. 6,107, 734, demonstrated a 3-EL-unit OLED using In—Zn—O (IZO) films or Mg:Ag/IZO films as intermediate connectors and achieved a luminous efficiency of 10.1 cd/A from pure tris(8-hydroxyquinoline)aluminum emitting layers. Kido et al. U.S. Patent Publication 2003/0189401 A1 discloses the use of light-emissive units partitioned from each other by at least one charge generation layer, the charge generation layer constituting an electrically insulating layer having a resistivity of not less than $1.0 \times 10^2 \Omega\text{cm}$. Kido et al., "High Efficiency Organic EL Devices having Charge Generation Layers", *SID 03 Digest*, 964 (2003), fabricated 3-EL-unit OLEDs using In—Sn—O (ITO) films or V_2O_5 films as intermediate connectors and achieved a luminous efficiency of up to 48 cd/A from fluorescent dye doped emitting layers. The disclosures of the above references with respect to intermediate connector materials are herein incorporated by reference.

[0042] The intermediate connectors layers, including interfacial layers, can be produced, e.g., by thermal evaporation, electron beam evaporation, or ion sputtering tech-

nique. Preferably, the intermediate connectors are fabricated from materials which allow for a thermal evaporation method for the deposition of all the materials in the fabrication of the cascaded OLED, including the intermediate connectors.

[0043] FIG. 1 shows a cascaded bottom emitting OLED device 100 in accordance with one embodiment of the present invention. This cascaded OLED has a plurality of independently controlled anodes 110 located over a substrate 105 and a common cathode 140, at least one of which is transparent. Disposed between the anode and the cathode are a stack 120 of three organic EL units 121, 122, and 123. These organic EL units are cascaded serially to each other and to the anode and the cathode. Unit 121 is the first EL unit (adjacent to the anode) and 123 is the third unit (adjacent to the cathode). EL unit 122 is an intermediate organic EL unit disposed between unit 121 and 123. Disposed between any two adjacent organic EL units is a connecting unit 130. Each anode 110 in the cascaded OLED 100 is externally connected to a voltage/current source 150 through electrical conductors 160 and can be individually powered to provide current for the associated light emitters, typically through either a passive-matrix or active-matrix control scheme.

[0044] The number of the organic EL units in the cascaded OLED is in principle equal to or more than 2. Preferably, the number of the organic EL units in the stacked OLED is such that the luminance efficiency in units of cd/A is improved or maximized. In further preferred embodiments, three or more cascaded organic EL units providing independent optical emission peaks may be employed to provide a white emission having a combination of relatively narrow band emitters rather than a single broad-band white light emission.

[0045] According to the present invention, EL units 121, 122, and 123 emit light of different colors having different efficiencies, for example red light 128, green light 129, and blue light 127. The plurality of cascaded organic electroluminescent units are disposed between a plurality of anodes 110 and a common cathode 140. Each of the plurality of anodes defines an independently controlled light-emitting area of the OLED device. Each independently controlled light-emitting area is associated with a complementary color filter 124, 125, 126 that transmits the light emitted by only one of the three EL units. By providing at least two independently controlled light-emitting areas having differently colored filters that filter the emitted light, and more preferably by providing a color filter complementary and matched to each of the different colors and associated with a different light emitting area, each light emitting area can be made to emit light of a different color, thus providing a full-color OLED device.

[0046] Cascaded OLED 100 is operated by applying an electric potential generated by a voltage/current source 150 between a pair of contact electrodes, anodes 110 and cathode 140, such that anode 110 is at a more positive potential with respect to the cathode 140. This externally applied electrical potential is distributed among the three organic EL units in proportion to the electrical resistance of each of these units. The electric potential across the cascaded OLED causes holes (positively charged carriers) to be injected from anodes 110 into the first organic EL unit 121, and electrons (negatively charged carriers) to be injected from cathode 140 into the third organic EL unit 123. Simultaneously,

electrons and holes are generated in, and separated from, each of the connecting units 130. Electrons thus generated in a connecting unit 130 are injected towards the anode and into the adjacent organic EL unit. Likewise, holes generated in a connecting unit 130 are injected towards the cathode and into the adjacent organic EL unit. Subsequently, these electrons and holes recombine in their corresponding organic EL units to produce light, which is observed via the transparent electrode or electrodes of the OLED through the corresponding color filter 124, 125, 126. In other words, the electrons injected from cathode are energetically cascading from the third organic EL unit 123 to the first organic EL unit 121, and emit light in each of the organic EL units. Therefore, we prefer to use the term "cascaded OLED" instead of "stacked OLED" in the present invention.

[0047] Each organic EL unit in the cascaded OLED 100 is capable of supporting hole and electron transport, and electron-hole recombination to produce light. Each organic EL unit can include a single layer or a plurality of layers. Organic EL multilayer structures include HTL/ETL, HTL/LEL/ETL, HIL/HTL/LEL/ETL, HIL/HTL/LEL/ETL/EIL, HIL/HTL/electron-blocking layer or hole-blocking layer/LEL/ETL/EIL, HIL/HTL/LEL/hole-blocking layer/ETL/EIL. Organic EL unit can be formed from small molecule OLED materials or polymeric LED materials, both known in the art, or combinations thereof. There are many organic EL multilayer structures and materials known in the art that can be used as the organic EL unit of this invention. Each organic EL unit in the cascaded OLED device can be the same or different from other units. Some organic EL units can be polymeric LED and other units can be small molecule OLEDs. Each organic EL unit can be selected in order to optimize performance or achieve a desired attribute, for example light transmission through the OLED stack, driving voltage, luminance efficiency, light emission color, manufacturability, device stability, and so forth.

[0048] The layer structure of the organic EL unit adjacent to the anode preferably is of HIL/HTL/LEL/ETL, and the layer structure of the organic EL unit adjacent to the cathode preferably is of HTL/LEL/ETL/EIL, and the layer structure of the intermediate organic EL units preferably are of HTL/LEL/ETL. Connectors facilitate hole injection into the HTL of one organic EL unit and electron injection into ETL of the adjacent organic EL unit.

[0049] Within each organic EL unit, the transport of the hole and electron carriers is supported by the HTL and ETL, respectively. The LEL may itself be an ETL. Recombination of the hole and electron carriers in the vicinity at or near the HTL/ETL interface within each organic EL unit causes light to be produced (electroluminescence). The HTL in each organic EL unit can be the same or different in terms of materials used, layer thickness, method of deposition, and so forth. The properties of the HTL in the device can be individually optimized to achieve the desired performance or feature, for example light transmission through the OLED stack, driving voltage, luminance efficiency, light emission color, manufacturability, device stability, and so forth. The same is true for the ETL and LEL. Although not necessary, it is preferable that a hole-injecting layer (HIL) be provided between the anode and the first HTL. It is also preferable, but not necessary, that an electron-injecting layer (EIL) be provided between the cathode and the last ETL. Both the HIL and EIL improve charge injection from the electrodes.

Organic EL units can optionally include a HIL between a HTL and a doped organic connector. Similarly, organic EL units can optionally include an EIL between an ETL and a doped organic connector.

[0050] In order to minimize driving voltage for the cascaded OLED, it is desirable to make each organic EL unit as thin as possible without compromising the electroluminescence efficiency. It is preferable that each organic EL unit is less than 500 nm thick, and more preferable that it be 2-200 nm thick. It is also preferable that each layer within the organic EL unit be 200 nm thick or less, and more preferable that it be 0.1-100 nm.

[0051] The cascaded OLED **100** of the present invention is typically provided over a supporting substrate **105** where either the cathode **140** or anode **110** can be in contact with the substrate **105**. The electrode in contact with the substrate is conveniently referred to as the bottom electrode. Conventionally, the bottom electrode is the anode, but the present invention is not limited to that configuration. The substrate can either be light transmissive or opaque, depending on the intended direction of light emission. The light transmissive property is desirable for viewing the EL emission through the substrate (a bottom emitter configuration). Transparent glass or plastic is commonly employed in such cases. For applications where the EL emission is viewed through the top electrode (a top emitter), the transmissive characteristic of the bottom support is immaterial, and therefore can be light transmissive, light absorbing or light reflective. Substrates for use in this case include, but are not limited to, glass, plastic, semiconductor materials, silicon, ceramics, and circuit board materials. Of course, it is necessary to provide in these device configurations a light-transparent top electrode. In such a configuration, the color filters **124-126** may be provided over the cathode **140**, any protective layers located over the cathode **140**, or on an encapsulating cover (not shown) provided over the OLED materials and affixed to the substrate **105**.

[0052] When EL emission is viewed through anode **110**, the anode should be transparent or substantially transparent to the emission of interest. Common transparent anode materials used in the present invention are indium-tin oxide (ITO), indium-zinc oxide (IZO) and tin oxide, but other metal oxides can work including, but not limited to, aluminum- or indium-doped zinc oxide, magnesium-indium oxide, and nickel-tungsten oxide. In addition to these oxides, metal nitrides, such as gallium nitride, and metal selenides, such as zinc selenide, and metal sulfides, such as zinc sulfide, can be used as the anode. For applications where EL emission is viewed only through the cathode electrode, the transmissive characteristics of anode are immaterial and any conductive material can be used, transparent, opaque or reflective. Example conductors for this application include, but are not limited to, gold, iridium, molybdenum, palladium, and platinum. Typical anode materials, transmissive or otherwise, have a work function higher than 4.0 eV. Desired anode materials are commonly deposited by any suitable means such as evaporation, sputtering, chemical vapor deposition, or electrochemical means. Anodes can be patterned using well-known photolithographic processes. Optionally, anodes may be polished prior to application of other layers to reduce surface roughness so as to minimize electrical shorts or enhance reflectivity.

[0053] While not always necessary, it is often useful to provide a HIL in the first organic EL unit to contact the anode **110**. The HIL can serve to improve the film formation property of subsequent organic layers and to facilitate injection of holes into the HTL reducing the driving voltage of the cascaded OLED. Suitable materials for use in the HIL include, but are not limited to, porphyrinic compounds as described in U.S. Pat. No. 4,720,432, plasma-deposited fluorocarbon polymers as described in U.S. Pat. No. 6,208,075, and some aromatic amines, for example, m-MTDATA (4,4',4"-tris[(3-ethylphenyl)phenylamino]triphenylamine). A p-type doped organic layer for use in the aforementioned connecting unit is also useful for the HIL as described in U.S. Pat. No. 6,423,429 B2. Alternative hole-injecting materials reportedly useful in organic EL devices are described in EP 0 891 121 A1 and EP 1 029 909 A1.

[0054] Referring to **FIG. 2**, the emission spectrum of a white-light emitting OLED made by applicant is shown. While this is a relatively efficient white-light emitting material, it can be seen that most of the energy in the spectrum is cyan (peak **200**) and yellow (peak **202**). When combined with red, green, and blue color filters, the amount of light emitted through the filters of the desired color will be relatively low. In particular, the green emission (peak **204**) and red emission (peak **206**) are relatively low.

[0055] The present invention can provide a higher efficiency full-color OLED device by providing a cascaded RGB architecture having a common control for all light-emitting units in the stack, where white light is generated by each pixel and filtered using RGB filters. This simplifies manufacturing because it is generally easier to apply a RGB filter after OLED device fabrication than to pattern RGB emitting pixels. However, since some of the light emitting units may be less efficient, they may need to be driven harder to provide comparable levels of light to produce, for example, a white or gray color. This may cause the less efficient materials to age more rapidly, thereby causing color differential aging and a color-OLED device whose white point will change over time and whose luminance will decrease. In a preferred embodiment of the invention, this may be addressed by differential sizing of the independent controlled light emitting areas and corresponding differentially colored filters.

[0056] To illustrate the advantages of a preferred embodiment of the present invention in a practical example, a set of four materials developed by applicant may have efficiencies as listed in the table below:

Color	Efficiency (cd/A)	Relative Size
White	13	
Green	28	1
Red	9	3.1
Blue	6	4.7

[0057] As can be seen from the table above, the relative light-emitting efficiency of the various color light emitters varies. If all of the light emitting pixels were of the same size and the color filters were of equal efficiency over the output bandwidth of the light emitters, the green light would be much brighter because it is more efficient and the blue light

would be dimmer because it is less efficient. The relative efficiency of the light-emitting materials and the associated color filter can be accommodated by creating anodes and color filters of relatively different sizes corresponding to the relative efficiencies. The more efficient units will have smaller associated color filters and anodes, the less efficient units will have larger color filter and anodes. For example, as shown in **FIG. 1**, the green filter is smallest and the blue filter is largest. Using the example of the materials cited in the table above, the relative sizes of the anodes and associated color filters are 1 for green, 3.1 for red, and 4.7 for blue. The relative sizes of the anodes and associated color filters should reflect the relative efficiency of the light emitting units in combination with the color filters. Alternatively, the relative sizes of the anodes and associated color filters may reflect the relative aging and lifetime of the light emitting units. Typically, the relative size of an independently controlled light-emitting area and associated color filter will be inversely related to the relative efficiency of light emission from or lifetime of the cascaded organic electroluminescent units. E.g., an efficient long-lived emitter will be relatively smaller than an inefficient short-lived emitter. As shown in the embodiment of **FIG. 1**, filters **124-126** may be a conventional color filter array such as is used in the liquid crystal display industry composed of light-absorbent material that only permits the desired color of light to pass through. The frequency of light passed through the filter from the stack **120** of light emitting units should match the emission spectrum of the light from the corresponding desired unit. For example, the light transmitted by a green filter should match the emission of the green light emitting unit.

[0058] For maximum benefit, both the emission of the light emitting units and the color filters will be as narrow as possible. This will optimize the power emitted through the color filters while providing an improved color gamut. Moreover, a narrow emission spectrum will provide improved contrast to the OLED device by absorbing more of the ambient light.

[0059] As illustrated in **FIG. 1**, the OLED device has three light-emitting layers, e.g., one each for red, green, and blue. However, it is also possible to combine a color emitter with a broadband emitter and utilize only two light emitting units. As noted above, the white-light emitting material that produces the spectrum illustrated in **FIG. 2** is very deficient in green light emission. An OLED device according to another embodiment of the present invention can have a white-light emitting unit in combination with a green-light emitting unit, as illustrated in **FIG. 3**. This arrangement reduces the number of light-emitting units while providing improved light output over a single, white-light emitting unit. In this arrangement, the green color filter **126** permits green light **129** from both the green unit **123** and the white unit **170** to pass while absorbing the blue and red light from the white unit **170**. The red light **128** emitted from the white unit **170** will pass through the red color filter **125** while blocking the green light emitted from the green light-emitting unit **123** and blue light from the white-light emitting unit **170**. The blue light **127** emitted from the white unit **170** will pass through the blue color filter **124** while blocking the green light emitted from the green light-emitting unit **123** and red light from the white-light emitting unit **170**.

[0060] In general, a plurality of combinations of light-emitting units having different colors may be employed. There are many other combinations of organic EL units in addition to red, green and blue that can be used to yield light that appears white. For example, two-layer structures that emit blue and yellow light, or that emit red and cyan light, or that emit green and magenta light, can be used to generate white light. In all cases these units can be combined multiple times. Further, any combination of colored-light and white-light emitting units that provide improved efficiency of output may be included in the present invention. Applicant has found that a variety of solutions for various applications may be possible that include various color filters combined with OLED materials to provide a good color gamut and efficiency.

[0061] The present invention may also be employed in an RGBW configuration, that is, one that has four light-emitting pixels, one each for red, green, blue, and white. In this arrangement, the white pixel need not have any filter at all, or may only have a filter necessary to achieve the desired white point of the OLED device. It is also possible to provide two layers that emit the same color of light; this technique can be used to optimize the color balance of the white emitter or the relative amount of light emitted by the various colors.

[0062] The order of layer deposition may be controlled to optimize the structure of the present invention to improve performance. Applicant has demonstrated that the organic materials within an OLED are themselves light absorbing. Hence, some of the light emitted from the bottom of a stack (the side farthest from the side from which light is emitted from an OLED) will be absorbed by layers above it. Therefore, it is useful to put the most efficient emitter or the one that emits light that is least absorbed by the other layers at the bottom. In the example cited above, green is the most efficient and thus may be advantageously placed at the bottom of the stack. Likewise, models of the light absorption in the various organic layers indicate that blue light is absorbed most readily, so that the blue light-emitting unit may be located at the top of the stack, as illustrated in **FIG. 1**.

[0063] The present invention provides improved manufacturability of a color OLED device by enabling unpatterned deposition of organic materials onto the substrate. No masking is needed because all of the layers are deposited across the entire light emitting area of the substrate. Moreover, improved efficiency over the use of a conventional white-light emitting OLEDs may be obtained by providing an output spectrum for each of the colors that is closely matched to its associated color filter.

[0064] The entire contents of the patents and other publications referred to in this specification are incorporated herein by reference.

[0065] The above examples demonstrate that significant increase in luminance efficiency can be achieved by using a cascaded OLED structure of the present invention comparing the conventional OLED. If operated with the same luminance, significant increase in operational lifetime can also be achieved by using the cascaded OLED structure of the present invention comparing the conventional OLED. Moreover, during operation, the driving voltage can be stabilized due to the insertion of the interfacial layer in the

connecting unit. The invention has been described in detail with particular reference to certain preferred embodiments thereof, but it will be understood that variations and modifications can be effected within the spirit and scope of the invention.

PARTS LIST		
100		cascaded OLED
105		substrate
110		anode
120		EL stack
121		blue EL unit
122		red EL unit
123		green EL unit
124		blue color filter
125		red color filter
126		green color filter
127		blue light
128		red light
129		green light
130		connecting unit
140		cathode
150		voltage/current source
160		electrical conductors
170		white EL unit
200		cyan peak
202		yellow peak
204		green peak
206		red peak

1. An OLED device comprising a substrate having thereon a cascaded organic electroluminescent device comprising:

- a) an anode;
- b) a cathode;
- c) a plurality of cascaded organic electroluminescent units disposed between the anode and the cathode, wherein each organic electroluminescent unit includes at least one light-emitting layer and wherein the plurality of cascaded units includes at least two units that emit light of different colors; and
- d) a connecting unit disposed between each adjacent cascaded organic electroluminescent unit; and
- e) a colored filter that filters the emitted light.

2. The OLED device of claim 1, comprising a plurality of anodes defining independently controlled light-emitting areas of the OLED device, wherein the plurality of cascaded organic electroluminescent units are disposed between the plurality of anodes and a common cathode.

3. The OLED device of claim 2, wherein at least two independently controlled light-emitting areas have differently colored filters that filter the emitted light.

4. The OLED device of claim 3, wherein the colors of the light transmitted by the differently colored filters are matched to the colors of the light emitted by one of the plurality of organic electroluminescent units.

5. The OLED device of claim 4 wherein the cascaded organic electroluminescent units include units that individually emit red, green, and blue light.

6. The OLED device of claim 3 wherein the cascaded organic electroluminescent units include units that in combination emit white light.

7. The OLED device of claim 6 wherein the color filters transmit red, green, or blue light.

8. The OLED device of claim 3 wherein the efficiency of light emission from or lifetime of one of the cascaded organic electroluminescent units is different from the efficiency of light emission from or lifetime of another of the organic electroluminescent units.

9. The OLED device of claim 8 wherein the relative size of an independently controlled light-emitting area and associated color filter is inversely related to the relative efficiency of light emission from or lifetime of the cascaded organic electroluminescent units.

10. The OLED device of claim 8 wherein the cascaded organic electroluminescent units are ordered in a stack to correspond to the relative efficiency of light emission from the organic electroluminescent units in the stack.

11. The OLED device of claim 8 wherein the cascaded organic electroluminescent units are ordered in a stack to minimize the relative absorption of light emission from the organic electroluminescent units in the stack as the light passes through the stack.

12. The OLED device of claim 3 further comprising an independently controlled light-emitting area and cascaded organic electroluminescent unit without a corresponding color filter.

13. The OLED device of claim 12 wherein the independently controlled light-emitting area without a corresponding color filter emits white light.

14. The OLED device of claim 1 wherein two of the cascaded organic electroluminescent units emit light of the same color.

15. The OLED device of claim 1 wherein the light is emitted through the substrate.

16. The OLED device of claim 1 further comprising a cover formed over the cascaded organic electroluminescent units wherein the light is emitted through the cover.

17. The OLED device of claim 1 comprising two cascaded organic electroluminescent units that emit green and white light respectively.

18. The OLED device of claim 1 comprising two cascaded organic electroluminescent units that emit blue and yellow light respectively.

19. The OLED device of claim 1 comprising two cascaded organic electroluminescent units that emit red and cyan light respectively.

20. The OLED device of claim 1 comprising two cascaded organic electroluminescent units that emit green and magenta light respectively.

21. The OLED device of claim 1, wherein the connecting unit comprises a doped organic layer.

22. The OLED device of claim 21, wherein the connecting unit comprises, in sequence, an n-type doped organic layer, and a p-type doped organic layer.