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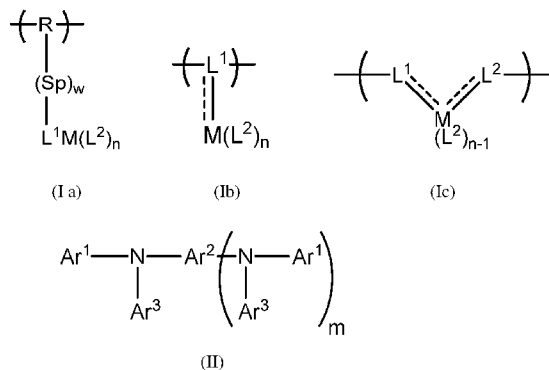
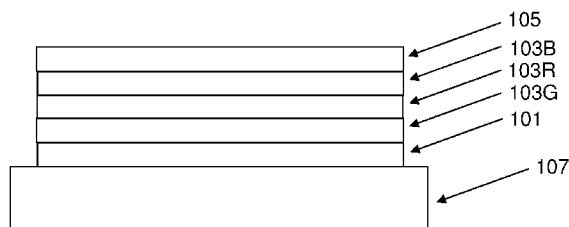
(54) **Title:** POLYMER AND ORGANIC LIGHT EMITTING DEVICE

FIGURE 1B



(57) **Abstract:** Polymer and Organic Light Emitting Device A polymer comprising a repeat unit of formula (I a), (I b) or (Ic): (Formulae (Ia), (Ib), (Ic)) wherein M is a metal; R is a backbone repeat group; n is 1 or 2; Sp is a spacer group; w is 0 or 1; L¹ and L² are mono- or bidentate co-coordinating groups; ---- represents a second metal to ligand bond in the case where L¹ or L² is a bidentate ligand and at least one of L¹ and L² is substituted with group of formula (II) (Formula (II)) wherein m is 0 or 1; each Ar¹, Ar² and Ar³ is aryl or heteroaryl. The polymer may be used as an emissive material in an organic light-emitting device.



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Polymer and Organic Light Emitting Device

Field of the Invention

The present invention relates to light-emitting materials and devices. More particularly the present invention relates to phosphorescent light-emitting polymer materials, formulations and light-emitting devices comprising said light-emitting polymers and methods of making said light-emitting devices.

Background

Electronic devices comprising active organic materials are attracting increasing attention for use in devices such as organic light emitting diodes (OLEDs), organic photoresponsive devices (in particular organic photovoltaic devices and organic photosensors), organic transistors and memory array devices. Devices comprising organic materials offer benefits such as low weight, low power consumption and flexibility. Moreover, use of soluble organic materials allows use of solution processing in device manufacture, for example inkjet printing or spin-coating.

An OLED may comprise a substrate carrying an anode, a cathode and one or more organic light-emitting layers between the anode and cathode.

Holes are injected into the device through the anode and electrons are injected through the cathode during operation of the device. Holes in the highest occupied molecular orbital (HOMO) and electrons in the lowest unoccupied molecular orbital (LUMO) of a light-emitting material combine to form an exciton that releases its energy as light.

Within an OLED device, the light-emitting material may be used as a dopant within a light emitting layer. The light-emitting layer may comprise a semiconducting host material and the light-emitting dopant, and energy will be transferred from the host material to the light-emitting dopant. For example, J. Appl. Phys. 65, 3610, 1989 discloses a host material doped with a fluorescent light-emitting dopant (that is, a light-emitting material in which light is emitted via decay of a singlet exciton) and Appl. Phys. Lett., 2000, 77, 904 discloses a host material doped with a phosphorescent light emitting

dopant (that is, a light-emitting material in which light is emitted via decay of a triplet exciton).

Emission from more than one layer of an OLED, in particular to achieve white light emission, is disclosed in, for example, WO 2008/131750, DE 102007020644 and EP1390962 and SPIE (2004), 5519, 42-47.

Suitable light-emitting materials include small molecule, polymeric and dendrimeric materials. Suitable light-emitting polymers include poly(arylene vinylenes) such as poly(p-phenylene vinylenes) and polyarylenes such as polyfluorenes.

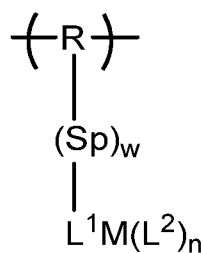
WO 2011/064355 discloses a metal complex substituted with an organic charge-transporting group for transporting holes or electrons and for improving solubility of the complex in an organic solvent.

WO 2005/013386 discloses an electroluminescent device comprising a polymeric host containing certain triarylamine repeat units and a phosphorescent metal complex.

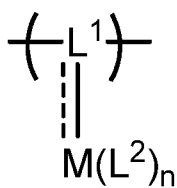
It is an object of the present invention to provide phosphorescent light-emitting materials which have an improved efficiency of emission and increased lifetime when used in devices containing one or more light-emitting layers.

Summary of the Invention

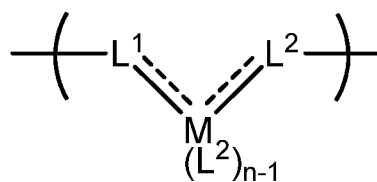
In a first aspect the invention provides a polymer comprising a repeat unit of formula (I a), (I b) or (Ic):



(I a)



(Ib)



(Ic)

wherein:

M is a metal;

R is a backbone repeat group;

n is 1 or 2;

Sp is a spacer group;

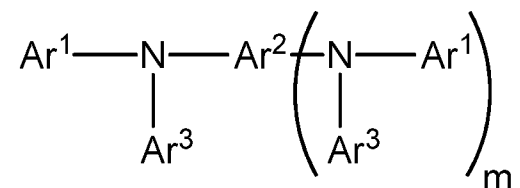
w is 0 or 1;

L^1 is a mono- or bidentate co-coordinating group;

L^2 which may be the same or different in each occurrence, is a mono- or bidentate coordinating group;

---- represents a second metal to ligand bond in the case where L^1 or L^2 is a bidentate ligand and

at least one of L^1 and L^2 is substituted with group of formula (II)



(II)

wherein m is 0 or 1;

each Ar^1 , Ar^2 and Ar^3 is independently in each occurrence selected from an aryl or heteroaryl that may be unsubstituted or substituted with one or more substituents, and any two of Ar^1 , Ar^2 and Ar^3 linked to the same N atom may be linked by a direct bond or a divalent linking group.

In a second aspect the invention provides a formulation comprising a polymer according to the first aspect and at least one solvent.

In a third aspect the invention provides an organic light-emitting device comprising an anode, a cathode and a light-emitting layer between the anode and the cathode, wherein the light-emitting layer comprises a polymer according to the first aspect.

In a fourth aspect the invention provides a method of forming the organic light-emitting device of the third aspect, the method comprising the step of forming the light-emitting layer over one of the anode or cathode, and forming the other of the anode or cathode over the light-emitting layer.

Description of the Drawings

The invention will now be described in more detail, with reference to the accompanying figures in which:

Figure 1A illustrates schematically an OLED according to the invention;

Figure 1B illustrates schematically an OLED according to an embodiment of the invention;

Figure 2 shows the emission efficiencies, presented as EQE vs voltage graph, of a white light emitting device containing a material according to an embodiment of the invention and a device containing a comparative emitting material;

Figure 3 shows plots of luminescence vs time for a white light emitting device containing a material according to an embodiment of the invention and a device containing a comparative emitting material;

Figure 4 shows the emission spectra of a white light emitting device containing a material according to an embodiment of the invention and a device containing a comparative emitting material;

Figure 5 shows a CIE chart of emission for a white light emitting device containing a material according to an embodiment of the invention and a device containing a comparative emitting material; and

Figure 6 shows the electroluminescent spectrum of a second device containing a material according to an embodiment of the invention in which substantially all emission from the device is from the material, and the spectrum of a comparative device;

Figure 7 shows the electroluminescent spectrum of a third device containing a material according to an embodiment of the invention in which substantially all emission from the device is from the material, and the spectra of two comparative devices;

Figure 8 shows plots of luminescence vs time for the third device and for comparative devices;

Figure 9 shows plots of current density vs. voltage for the third device and for comparative devices; and

Figure 10 shows plots of external quantum efficiency vs. voltage for the third device and for comparative devices

Detailed Description of the Invention

Figure 1A, which is not drawn to any scale, illustrates schematically an OLED according to an embodiment of the invention. The OLED is carried on substrate 107 and comprises an anode 101, a cathode 105 and a light emitting layer 103 between the anode and the cathode. Light-emitting layer 103 comprises the phosphorescent light-emitting polymer of the invention.

In operation, holes injected from the anode and electrons injected from the cathode combine in the light-emitting layer to form excitons.

With reference to Figure 1A, further layers may be provided between the anode and the cathode including, without limitation, charge-transporting layers, charge-blocking layers and charge injection layers. The device may contain more than one light-emitting layer.

Exemplary OLED structures including one or more further layers include the following:

Anode / Hole-injection layer / Light-emitting layer / Cathode;

Anode / Hole transporting layer / Light-emitting layer / Cathode;

Anode / Hole-injection layer / Hole-transporting layer / Light-emitting layer / Cathode;

Anode / Hole-injection layer / Hole-transporting layer / Light-emitting layer / Electron-transporting layer / Cathode.

In one preferred embodiment, the OLED comprises at least one, optionally both, of a hole injection layer and a hole transporting layer.

Substantially all emission from a device may be from a material of the invention in the light-emitting layer of the device. In other embodiments, one or more further light-emitting materials may be provided in the device. Further light-emitting materials may be provided in the same layer as the light-emitting layer containing the polymer of the invention, or may be provided in one or more further light-emitting layers. The further light-emitting materials may be selected from fluorescent or phosphorescent light-emitting materials having a colour of emission differing from or the same as that of the polymer material of the invention.

Figure 1B, which is not drawn to any scale, illustrates schematically an OLED according to another embodiment of the invention. The OLED is carried on substrate 107 and comprises an anode 101, a cathode 105 and three light emitting layers 103G, 103R and 103B between the anode and the cathode. Green light-emitting layer 103G comprises a phosphorescent light-emitting material of the invention, light-emitting layer 103R comprises a red light-emitting material, and light-emitting layer 103B comprises a blue light-emitting material.

In operation, holes injected from the anode and electrons injected from the cathode combine in at least the first light-emitting layer 103G and the third light-emitting layer 103B to form excitons.

Light-emitting layer 103G may further be provided with more than one light-emitting dopant, for example a blue and/or a red light-emitting dopant.

Light emission from layer 103R may occur by a mechanism similar to that described above with reference to emission from layer 103G. In addition to, or as an alternative to, recombination of holes and electrons to form excitons in light emitting layer 103R, triplet excitons may migrate from layer 103G and / or layer 103B into layer 103R where they may be absorbed by the red phosphorescent dopant.

Light emitted from each of these three layers may combine to provide white light.

Highest occupied molecular orbital (HOMO) levels of the components of layers 103G, 103R and 103B may be selected by appropriate choice of host and / or dopants in order that holes injected from the anode may efficiently reach the light-emitting layer 103B. Likewise, the lowest unoccupied molecular orbital (LUMO) levels of these components may be selected in order that electrons injected from the cathode may efficiently reach the light-emitting layer 103G.

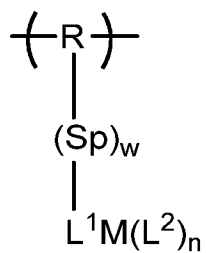
The HOMO level of the host and / or dopant of layer 103R is optionally no more than 0.5 eV, optionally no more than 0.3 eV, deeper (further from vacuum level) than the HOMO level of layer 103G and / or layer 103B. This may at least partially prevent trapping of holes at layer 103R.

The LUMO level of the host and / or dopant of layer 103R is optionally no more than 1.0 eV, optionally no more than 0.5 eV, optionally no more than 0.3 eV, shallower (closer to vacuum level) than the LUMO level of layer 103G and / or layer 103B. This may at least partially prevent trapping of electrons at layer 103R.

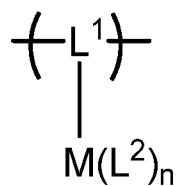
HOMO and LUMO levels may be measured by cyclic voltammetry.

Phosphorescent polymer

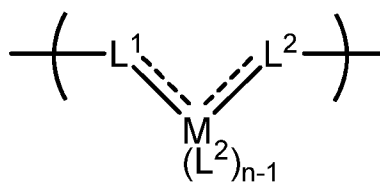
In a first aspect the invention provides a phosphorescent polymer comprising a repeat unit of formula (I a), (I b) or (I c):



(I a)



(I b)



(Ic)

wherein:

M is a metal;

R is a backbone repeat group;

n is 1 or 2;

Sp is a spacer group;

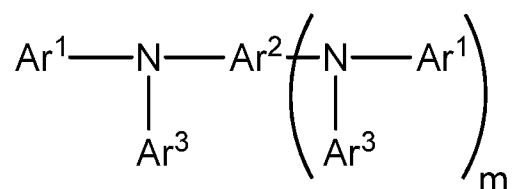
w is 0 or 1;

L¹ is a mono- or bidentate coordinating group;

L^2 which may be the same or different in each occurrence, is a mono- or bidentate coordinating group;

---- represents a second metal to ligand bond in the case where L^1 or L^2 is a bidentate ligand; and

at least one of L^1 and L^2 is substituted with group of formula (II)



(II)

wherein:

m is 0 or 1;

each Ar^1 , Ar^2 and Ar^3 is independently in each occurrence selected from an aryl or heteroaryl that may be unsubstituted or substituted with one or more substituents, any two of Ar^1 , Ar^2 and Ar^3 linked to the same N atom may be linked by a direct bond or a divalent linking group.

Unless stated otherwise, “aryl” and “heteroaryl” as used anywhere herein includes monocyclic and polycyclic aromatic and heteroaromatic groups.

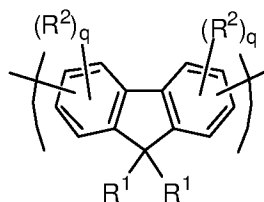
Exemplary metals M include transition metal elements, preferably group VIII transition metal element. Iridium is particularly preferred.

The polymer is preferably a conjugated polymer, and R preferably comprises aromatic or heteroaromatic groups conjugated to adjacent repeat units. Conjugated polymers contain repeat units in the polymer backbone that are conjugated together. The extent of conjugation of the polymer backbone may be controlled by selection of the repeat units.

In one embodiment R is $-(Ar^6)_p-$ wherein p is 1, 2, or 3 and each Ar^6 is independently selected from aryl or heteroaryl. Optionally, $p = 1$.

Exemplary groups Ar^6 include fluorene and phenylene.

If Ar^6 is fluorene then the repeat unit of formula (Ia) may be a group of formula (VI):



(VI)

wherein:

R^1 in each occurrence is the same or different and is H or a substituent and the two groups R^1 may be linked to form a ring;

R^2 in each occurrence is a substituent;

q is 0, 1, 2 or 3; and

the repeat unit of formula (VI) comprises at least one substituent R^1 or R^2 of formula $-(Sp)_w-L^1ML^2$, at least one of L^1 and L^2 being substituted with a group of formula (II).

Each R^1 is preferably a substituent, and each R^1 may independently be selected from the group consisting of:

- alkyl, optionally C_{1-20} alkyl, wherein one or more non-adjacent C atoms may be replaced with optionally substituted aryl or heteroaryl, O, S, substituted N, C=O or -COO-;
- aryl or heteroaryl that may be unsubstituted or substituted with one or more substituents;

- a linear or branched chain of aryl or heteroaryl groups, each of which groups may independently be substituted, for example a group of formula $-(Ar^7)_r$ wherein Ar^7 in each occurrence is independently an aryl or heteroaryl group that may be unsubstituted or substituted with one or more substituents and r is at least 2, optionally 2 or 3;
- a cross-linkable group, for example a group comprising a double bond such as a vinyl or acrylate group, or a benzocyclobutane group; and
- a group of formula $-(Sp)_w-L^1ML^2$, at least one of L^1 and L^2 being substituted with a group of formula (II).

In the case where R^1 is an aryl or heteroaryl group, or a linear or branched chain of aryl or heteroaryl groups, the or each aryl or heteroaryl group may be unsubstituted or substituted with one or more substituents R^3 selected from the group consisting of:

- alkyl, for example C_{1-20} alkyl, wherein one or more non-adjacent C atoms may be replaced with O, S, substituted N, C=O and -COO- and one or more H atoms of the alkyl group may be replaced with F or aryl or heteroaryl optionally substituted with one or more groups R^4 ,
- aryl or heteroaryl optionally substituted with one or more groups R^4 ,
- NR^5_2 , OR^5 , SR^5 , and
- fluorine, nitro and cyano;

wherein each R^4 is independently alkyl, for example C_{1-20} alkyl, and each R^5 is independently selected from the group consisting of C_{1-20} alkyl and aryl or heteroaryl optionally substituted with one or more alkyl groups.

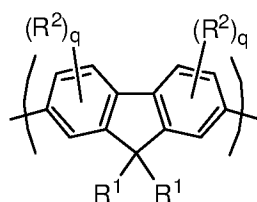
Optional substituents R^2 are preferably selected from the group consisting of alkyl, for example C_{1-20} alkyl, wherein one or more non-adjacent C atoms may be replaced with O, S, NH or substituted N, C=O and -COO-, optionally substituted aryl, optionally substituted heteroaryl, alkoxy, alkylthio, fluorine, cyano and arylalkyl. Particularly

preferred substituents include C₁₋₂₀ alkyl and substituted or unsubstituted aryl, for example phenyl. Optional substituents for the aryl include one or more C₁₋₂₀ alkyl groups.

Where present, substituted N may independently in each occurrence be NR⁶ wherein R⁶ is alkyl, optionally C₁₋₂₀ alkyl, or optionally substituted aryl or heteroaryl. Optional substituents for aryl or heteroaryl R⁶ may be selected from R⁵.

Preferably, each R¹ and, if present, R² is selected from the group consisting of $-(\text{Sp})_w-\text{L}^1\text{ML}^2$ and C₁₋₄₀ hydrocarbyl. Exemplary C₁₋₄₀ hydrocarbyl groups include C₁₋₂₀ alkyl; phenyl that may be unsubstituted or substituted with one or more C₁₋₂₀ alkyl groups; and $-(\text{Ar}^7)_r$ wherein Ar⁷ in each occurrence is phenyl that may be unsubstituted or substituted with one or more C₁₋₂₀ alkyl groups and r is 2 or 3.

In one arrangement, the repeat unit of formula (VI) may be a 2,7-linked repeat unit of formula (VIa):



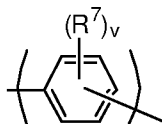
(VIa)

In another arrangement, the extent of conjugation of repeat units of formula (VI) with adjacent repeat units may be controlled by selecting the linking position of the group of formula (VI) to adjacent repeat units. For example, linking the repeat unit through the 3- and / or 6- positions may limit the extent of conjugation between repeat units adjacent to the repeat unit of formula (VI).

Alternatively or additionally, the extent of conjugation of the repeat unit of formula (VI) with an adjacent repeat unit may be limited by substituting the repeat unit of formula (VI) with one or more substituents R² in or more positions adjacent to the linking positions in

order to create a twist with the adjacent repeat unit or units, for example a 2,7-linked fluorene carrying a C₁₋₂₀ alkyl substituent in one or both of the 3- and 6-positions.

If Ar⁶ is phenylene then the repeat unit of formula (Ia) may be group of formula (VII):



(VII)

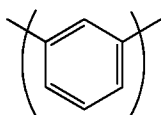
wherein *v* is 1, 2, 3 or 4, optionally 1 or 2; and R⁷ independently in each occurrence is a substituent, wherein at least one R⁷ is a group of formula $-(\text{Sp})_w-\text{L}^1\text{ML}^2$, at least one of L¹ and L² being substituted with a group of formula (II).

Each R⁷ may optionally be selected from substituents R¹ as described above with reference to formula (VI), for example C₁₋₂₀ alkyl, and phenyl that is unsubstituted or substituted with one or more C₁₋₂₀ alkyl groups.

The repeat unit of formula (VII) may be 1,4- linked, 1,2-linked or 1,3-linked.

If the repeat unit of formula (VII) is 1,2- or 1,3 linked, then the extent of conjugation of repeat unit of formula (VII) to one or both adjacent repeat units may be relatively low.

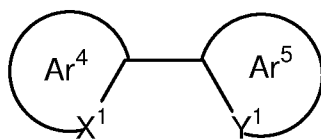
Optionally R is a repeat unit of formula (VIIa) substituted with at least one group $-(\text{Sp})_w-\text{L}^1\text{ML}^2$, at least one of L¹ and L² being substituted with a group of formula (II), and optionally substituted with one or more further substituents R⁷:



(VIIa)

In one embodiment each L¹ and L² are independently selected from ligands comprising a bidentate group forming organometallic complexes with carbon or nitrogen donors.

Exemplary bidentate groups include bidentate ligands of formula (III):

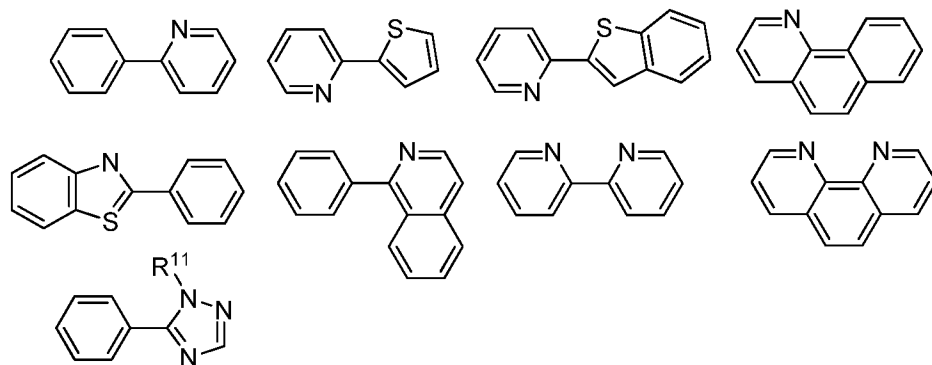


(III)

wherein Ar^4 and Ar^5 may be the same or different and are independently selected from optionally substituted aryl or heteroaryl; X^1 and Y^1 may be the same or different and are independently selected from carbon or nitrogen; and Ar^4 and Ar^5 may be fused together. Ligands wherein X^1 is carbon and Y^1 is nitrogen are particularly preferred.

$\text{ML}^1(\text{L}^2)_n$ is preferably a phosphorescent light-emitting group including, without limitation, red, green and blue phosphorescent light-emitting materials. Phosphorescent groups $\text{ML}^1(\text{L}^2)_n$ may luminesce by metal-to-ligand charge transfer (MLCT), and the identity of L^1 and / or L^2 may influence the colour of emission.

Specific examples of bidentate ligands are illustrated below:



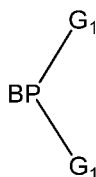
wherein R^{11} is a substituent, optionally a C_{1-40} hydrocarbyl group, for example a C_{1-20} alkyl group. Other ligands L^1 and L^2 suitable for use with d-block elements M include diketones, in particular acetylacetonate (acac); triarylphosphines and pyridine, each of which may be substituted.

Optionally the bidentate coordinating group is a 2-phenylpyridine group.

At least one of L^1 and L^2 is substituted with a group of formula (II), and L^1 and L^2 may each independently be substituted with one or more further substituents. In the case of a ligand of formula (III), each of Ar^4 and Ar^5 may independently be unsubstituted or may carry one or more substituents. Two or more of these substituents may be linked to form a ring, for example an aromatic ring. Particularly preferred substituents include fluorine or trifluoromethyl; C_{1-40} hydrocarbyl, for example C_{1-20} alkyl; C_{1-20} alkoxy groups; and dendrons.

Dendrons may be used to obtain or enhance solution processability of the polymer of the invention in common organic solvents. A light-emitting dendrimer metal complex comprises a light-emitting metal complex core substituted with one or more dendrons, wherein each dendron comprises a branching point and two or more dendritic branches. Preferably, the dendron is at least partially conjugated, and at least one of the branching point and dendritic branches comprises an aryl or heteroaryl group.

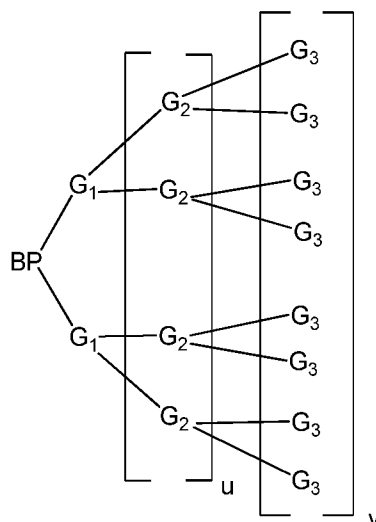
A dendron may have optionally substituted formula (VIII)



(VIII)

wherein BP represents a branching point for attachment to L^1 or L^2 , and G_1 represents first generation branching groups.

The dendron may be a first, second, third or higher generation dendron. G_1 may be substituted with two or more second generation branching groups G_2 , and so on, as in optionally substituted formula (VIIIa):



(VIIIa)

wherein u is 0 or 1; v is 0 if u is 0 or may be 0 or 1 if u is 1; BP represents a branching point for attachment to a core and G_1 , G_2 and G_3 represent first, second and third generation dendron branching groups.

Preferably, BP is phenyl. Preferably, each group G is selected from the group consisting of aryl and heteroaryl groups. Preferably, at least some or all of the G groups are optionally substituted phenyl.

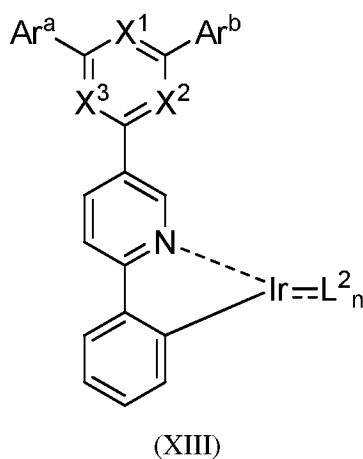
BP and / or any group G may be substituted with one or more substituents, for example one or more C_{1-20} alkyl or alkoxy groups.

With reference to the group of formula (II), in one specific embodiment m is 1.

One of Ar^1 and Ar^2 is bound directly to L^1 or L^2 .

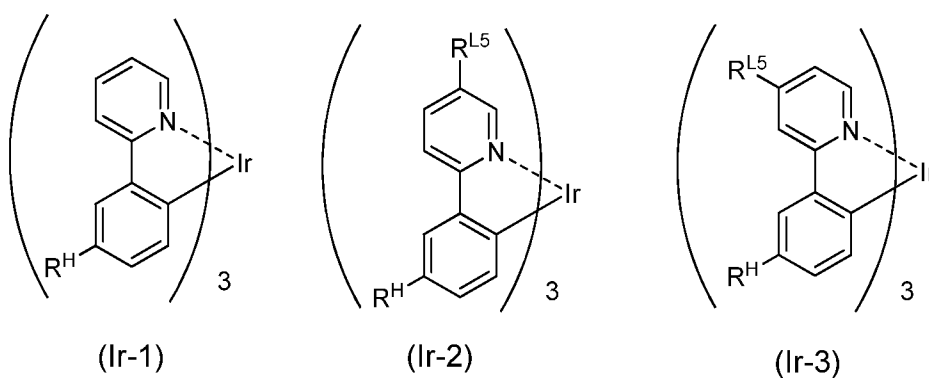
Optionally, L^2 is substituted with a group of formula (II).

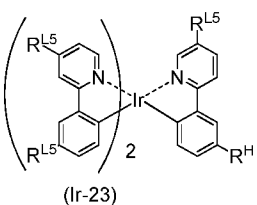
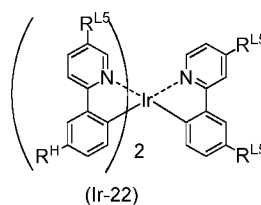
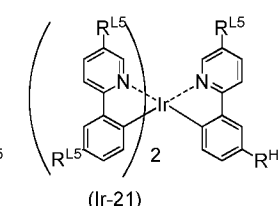
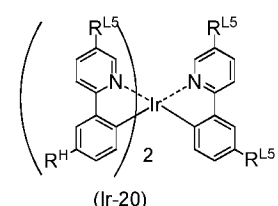
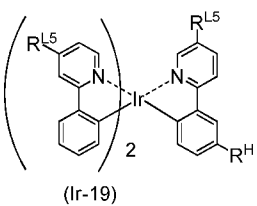
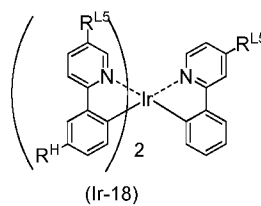
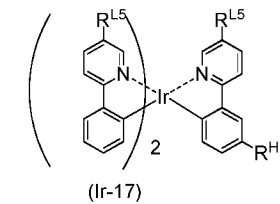
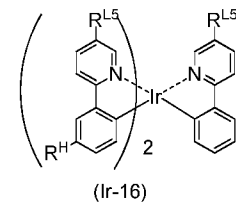
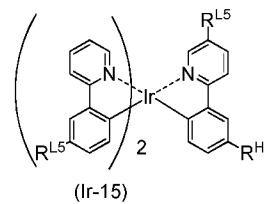
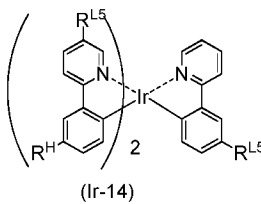
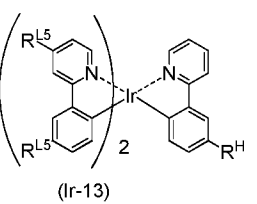
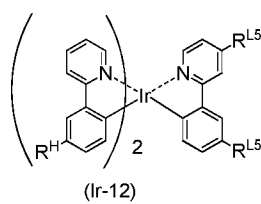
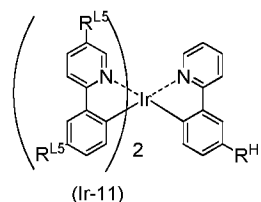
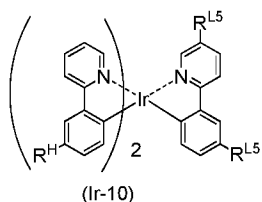
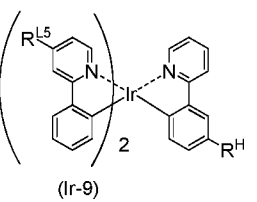
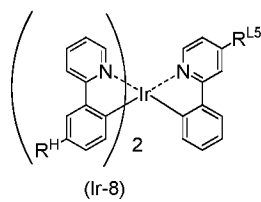
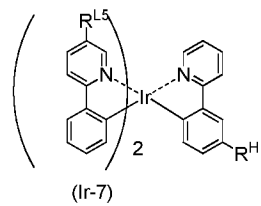
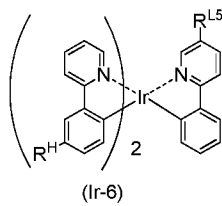
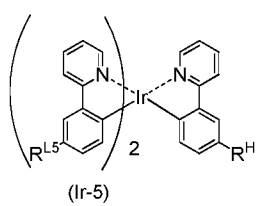
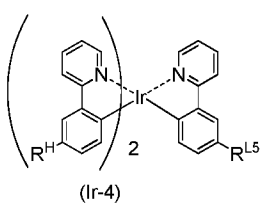
An exemplary complex of formula $ML^1L_n^2$ has formula (XIII):

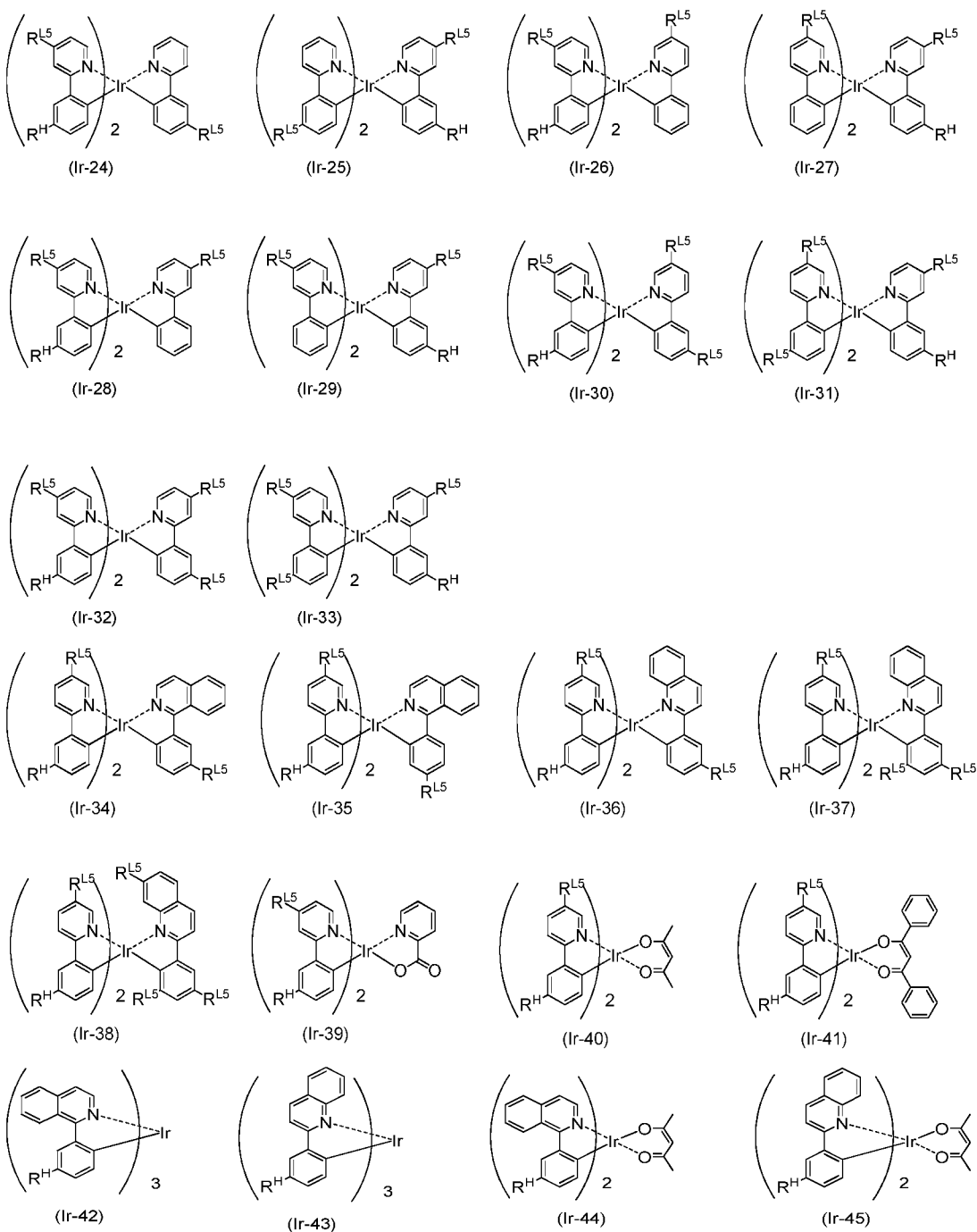


wherein Ar^a and Ar^b are each independently an aryl or heteroaryl group that may be unsubstituted or substituted with one or more substituents, for example phenyl that may be unsubstituted or substituted with one or more C_{1-20} alkyl groups; and X^1 , X^2 and X^3 are each independently N or CR^{12} wherein R^{12} in each occurrence is independently H or a substituent, preferably H or a C_{1-20} alkyl group.

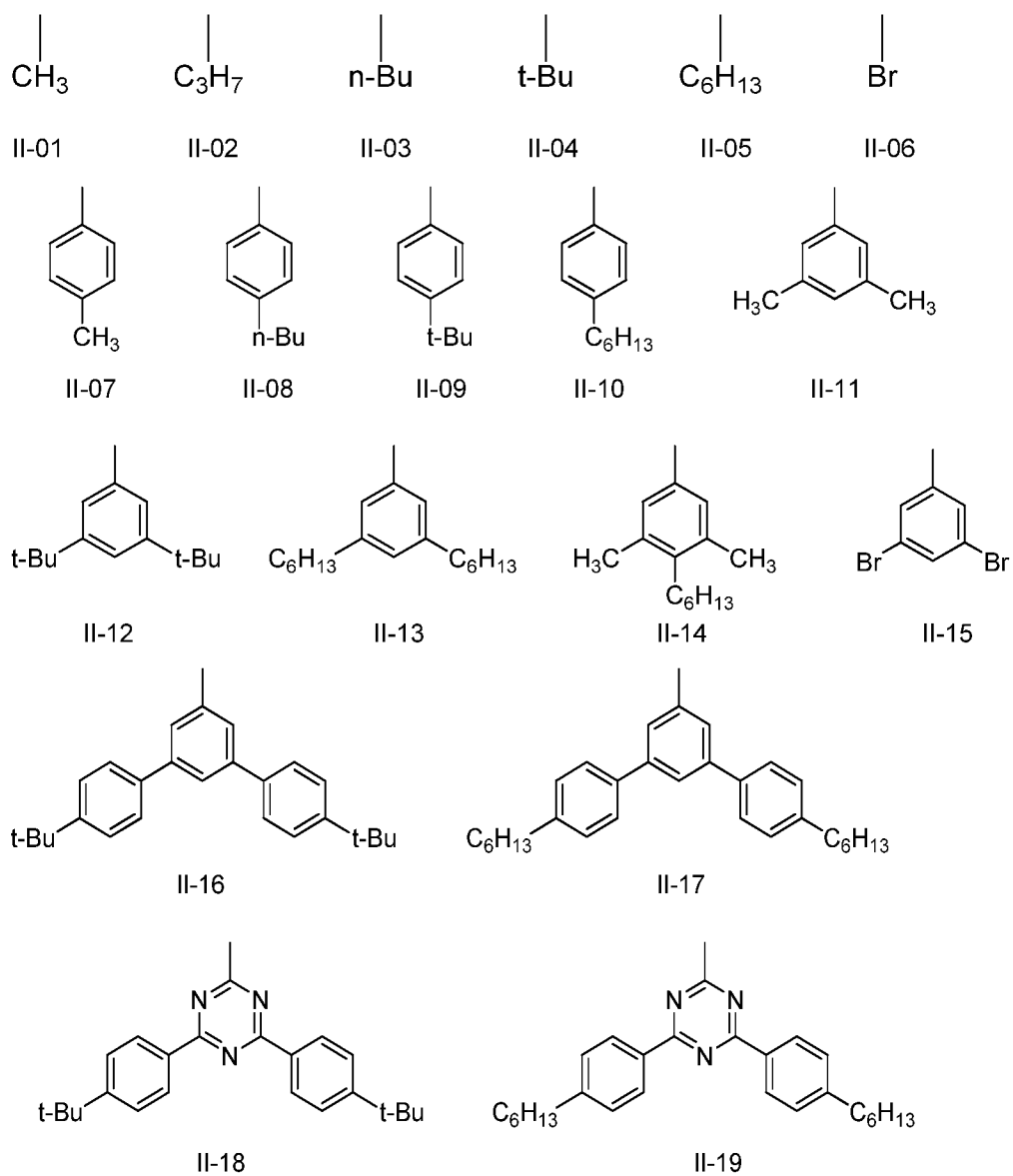
Exemplary complexes ML^1L_n^2 of a repeat unit of formula (Ia), (Ib) or (Ic) include complexes of formulae (Ir-1) to (Ir-45):

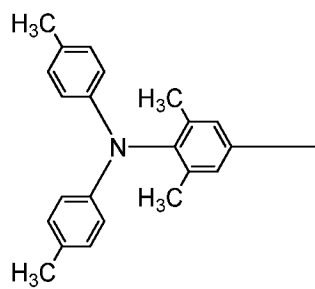




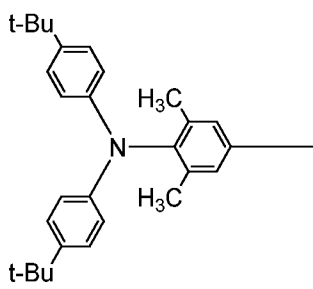


wherein R^{L5} is a group selected formulae (II-01) - (II-19) and R^H is a group selected from formulae (III-01) - (III-12)

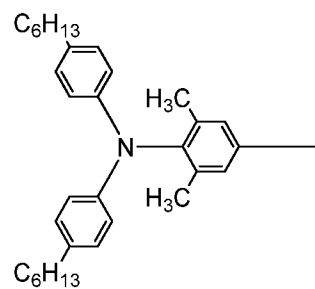




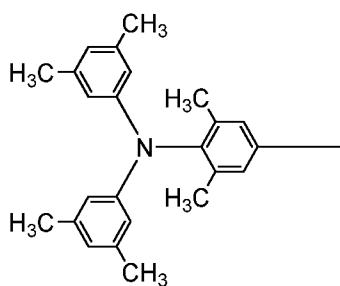
(III-01)



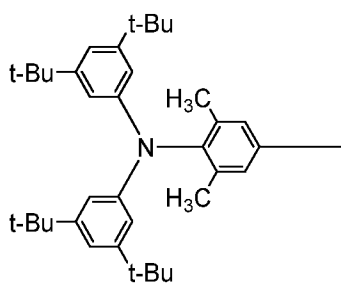
(III-02)



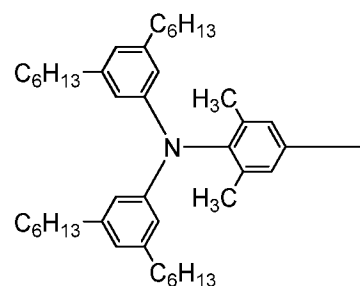
(III-03)



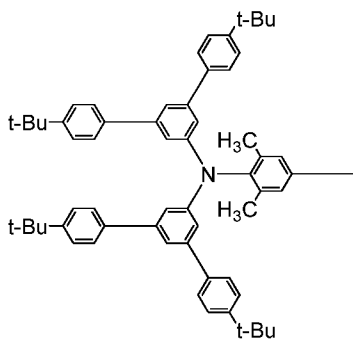
(III-04)



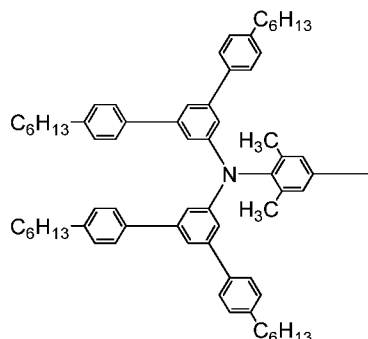
(III-05)



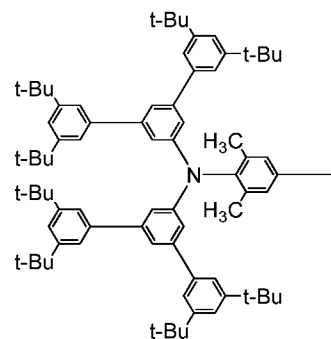
(III-06)



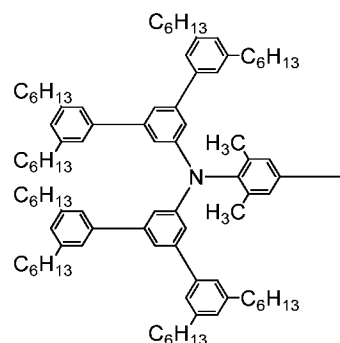
(III-07)



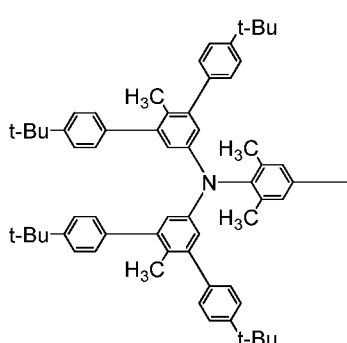
(III-08)



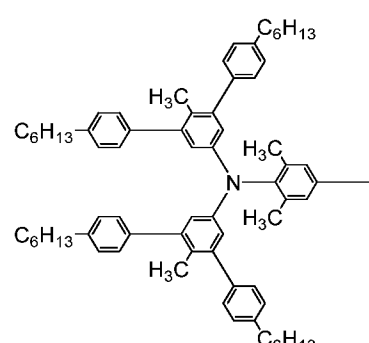
(III-09)



(III-10)



(III-11)

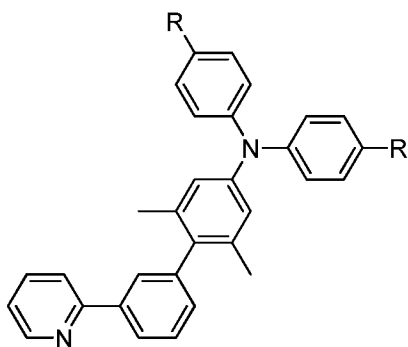


(III-12)

Preferably Ar^1 , Ar^2 and Ar^3 are each phenyl, each of which may be unsubstituted or substituted with one or more substituents.

Optionally one or more of Ar^1 , Ar^2 and Ar^3 are substituted with one or more C_{1-40} hydrocarbyl groups. Preferred substituents are C_{1-20} alkyl substituents. Exemplary substituents comprise a tertiary carbon atom. Substituents comprising a tert-butyl group are particularly preferred. Preferably, at least Ar^3 is substituted.

If the group of formula (II) is bound through Ar^1 , Ar^2 or Ar^3 to an aromatic ligand L^1 or L^2 then there may be conjugation between the aromatic ligand and the group of formula (II). The extent of any such conjugation may be limited by providing substituents on the aromatic ligand and / or on the group of formula (II) at one or more positions adjacent to the bond between the aromatic ligand and the group of formula (II) to create steric hindrance between the ligand and the group of formula (II) and to cause the group of formula (II) to twist relative to the aromatic ligand. Exemplary substituents for creating steric hindrance may be selected from C_{1-40} hydrocarbyl, for example C_{1-20} alkyl. An exemplary group of formula (II) with substituents for creating a twist is illustrated below.

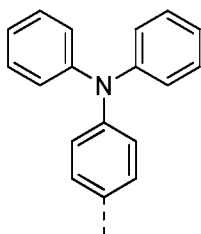


In another arrangement, the ligand L^1 or L^2 that Ar^1 , Ar^2 or Ar^3 of the group of formula (II) is bound to may be substituted adjacent to the substitution position of the group of formula (II) with the substituent causing a twist between the ligand and the group Ar^1 , Ar^2 or Ar^3 of formula (II) that is bound to the ligand.

Any two of Ar^1 , Ar^2 and Ar^3 linked to the same N atom may be linked by a direct bond or a divalent linking group. A divalent linking group, where present, is preferably selected

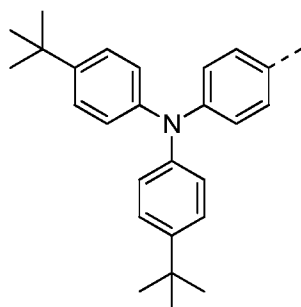
from O, S and CR^8_2 wherein R^8 in each occurrence is a substituent, preferably a C1-40 hydrocarbyl group, for example C_{1-20} alkyl.

An exemplary group of formula (II) has the following structure, that may be unsubstituted or substituted with one or more substituents.:



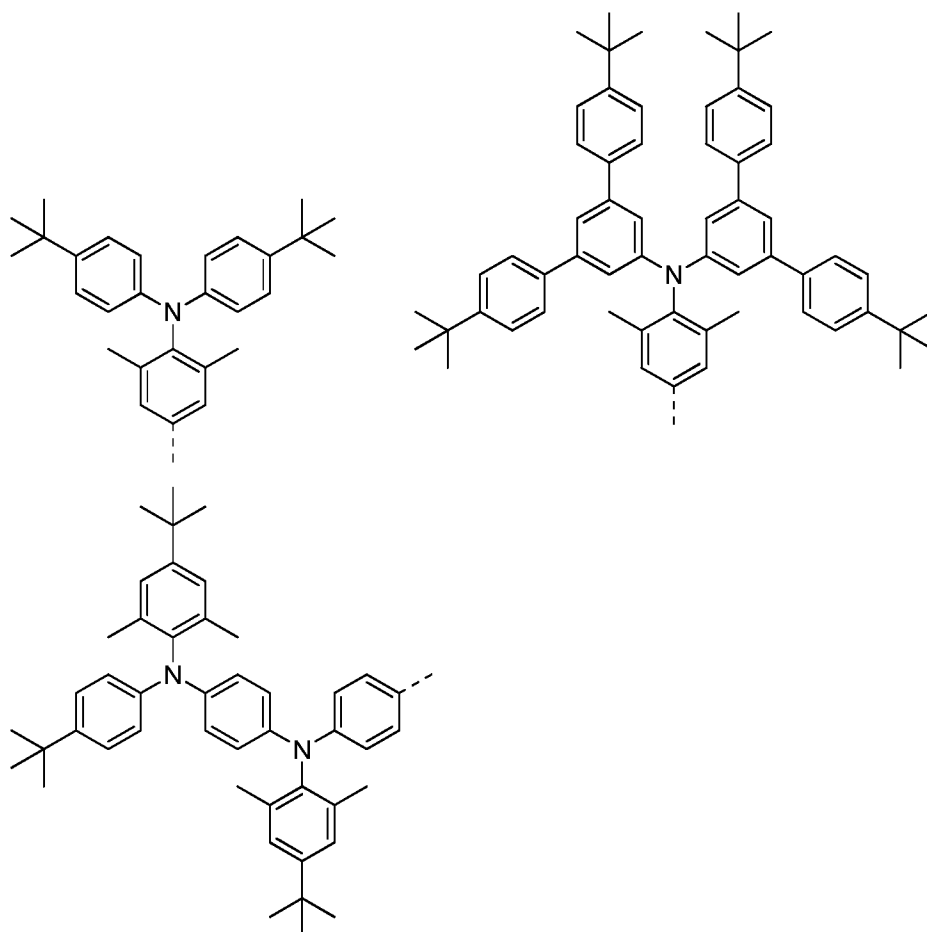
wherein --- represents a point of attachment of the group of formula (II) to L^1 or L^2 .

Exemplary substituted compounds of formula (II) include the following:



It will be appreciated that the structure illustrated above does not carry substituents at positions adjacent to a C-N bond that may result in a twist within the group of formula (II), or a substituent or substituents adjacent to the point of attachment to L^1 or L^2 that may result in a twist between the group of formula (II) and the ligand L^1 or L^2 that the group of formula (II) is bound to.

One or more substituents of Ar^1 , Ar^2 or Ar^3 of formula (II) may be provided adjacent to the bond to the C-N bond or bonds of formula (II). This substitution may create a twist in the group of formula (II). Exemplary substituents for creating a twist include C_{1-20} alkyl groups. Exemplary groups of formula (II) containing such a twist include the following compounds:



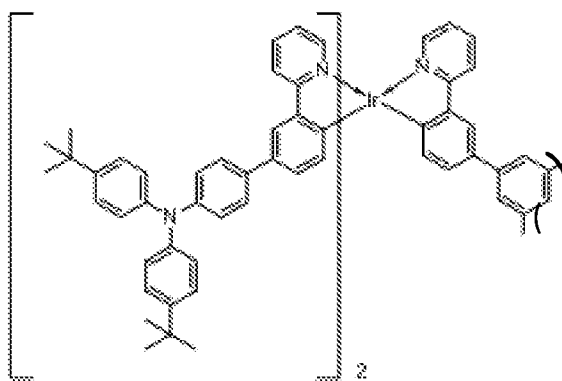
wherein --- represents a point of attachment of the group of formula (II) to L^1 or L^2 .

In one embodiment, $w = 0$ and L^1 is bound directly to R.

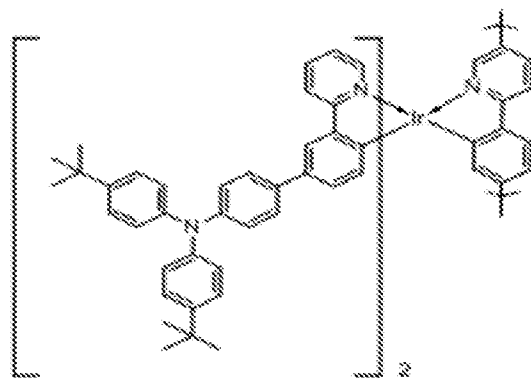
In another embodiment w is 1 and Sp is a C_{1-30} hydrocarbyl group, wherein one or more non adjacent chain C atoms of the hydrocarbyl group may be replaced with O, S or COO.

Exemplary groups Sp include phenyl, C_{1-20} alkyl and phenyl- C_{1-20} alkyl,

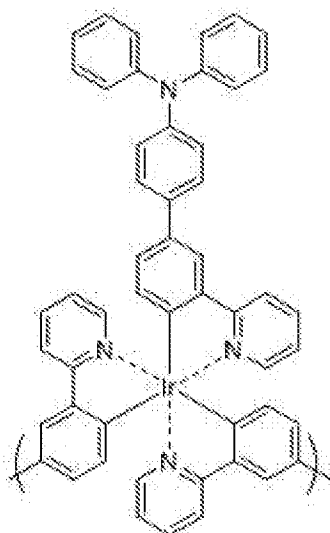
Exemplary repeat units according to embodiments of the invention include the following:



(IVa)



(IVb)



(IVc)

Repeat units of formulae (IVa), (IVb) and (IVc) may be substituted with one or more substituents as described above.

Polymers of the invention may contain 0.5 – 40 mol %, optionally 0.5-20 mol % of the repeat units of formula (I a) or (I b), for example repeat units of formula (IVa), (IVb) or (IVc)

Polymers as described herein may emit blue light, which may have a peak photoluminescence wavelength less than or equal to about 480 nm, optionally about 400-480 nm; green light, which may have a photoluminescent spectrum with a peak in the range of greater than 480 nm – 540 nm, optionally about 490-540 nm; red light, which may optionally have a peak in its photoluminescent emission spectrum at more than 580 nm – 630 nm, optionally about more than 580 up to about 630 nm; or yellow light, which may optionally have a photoluminescent spectrum with a peak in the range of more than about 540 up to 580 nm.

Preferably, polymers of the invention emit in the green or yellow wavelength range. Preferably, a photoluminescent spectrum of a polymer of the invention has a peak at a wavelength in the range 540-620 nm, preferably of less than 600 nm.

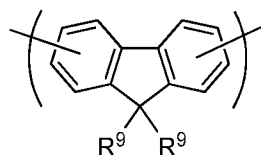
Polymers of the invention are preferably copolymers comprising repeat units of formula (Ia), (Ib) or (Ic) and one or more co-repeat units.

Optionally the co-repeat unit comprises an arylene group that may be unsubstituted or substituted with one or more substituents.

Exemplary co-repeat units include optionally substituted monocyclic and polycyclic arylene groups as disclosed in for example, Adv. Mater. 2000 12(23) 1737-1750 and include: 1,2-, 1,3- and 1,4-phenylene repeat units as disclosed in J. Appl. Phys. 1996, 79, 934; 2,7-fluorene repeat units as disclosed in EP0842208; indenofluorene repeat units as disclosed in, for example, Macromolecules 2000, 33(6), 2016-2020; and spirofluorene repeat units as disclosed in, for example EP0707020. Each of these repeat units is

optionally substituted. Examples of substituents include solubilising groups such as C₁₋₂₀ alkyl or alkoxy; electron withdrawing groups such as fluorine, nitro or cyano; and substituents for increasing glass transition temperature (T_g) of the polymer.

One exemplary class of arylene co-repeat units is optionally substituted fluorene group comprising repeat units, such as repeat units of formula IX:



(IX)

wherein R⁹ in each occurrence is the same or different and is H or a substituent, and wherein the two groups R⁹ may be linked to form a ring.

Each R⁹ is preferably a substituent, and each R⁹ may independently be selected from the group consisting of:

- optionally substituted alkyl, optionally C₁₋₂₀ alkyl, wherein one or more non-adjacent C atoms may be replaced with optionally substituted aryl or heteroaryl, O, S, substituted N, C=O or -COO-;
- optionally substituted aryl or heteroaryl;
- a linear or branched chain of aryl or heteroaryl, each of which groups may independently be substituted, for example a group of formula -(Ar³)_r as described above with reference to formula (VI); and
- a cross-linkable group, for example a group comprising a double bond such as a vinyl or acrylate group, or a benzocyclobutane group.

In the case where R⁹ comprises aryl or heteroaryl ring system, or a linear or branched chain of aryl or heteroaryl ring systems, the or each aryl or heteroaryl ring system may be substituted with one or more substituents R³ selected from the group consisting of:

- alkyl, for example C₁₋₂₀ alkyl, wherein one or more non-adjacent C atoms may be replaced with O, S, substituted N, C=O and -COO- and one or more H atoms of the alkyl group may be replaced with F or aryl or heteroaryl optionally substituted with one or more groups R⁴,
- aryl or heteroaryl optionally substituted with one or more groups R⁴,
- NR⁵₂, OR⁵, SR⁵, and
- fluorine, nitro and cyano;

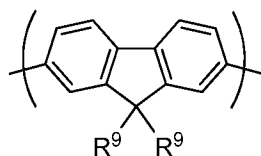
wherein each R⁴ is independently alkyl, for example C₁₋₂₀ alkyl, in which one or more non-adjacent C atoms may be replaced with O, S, substituted N, C=O and -COO- and one or more H atoms of the alkyl group may be replaced with F, and each R⁵ is independently selected from the group consisting of alkyl and aryl or heteroaryl optionally substituted with one or more alkyl groups.

Optional substituents for one or more of the aromatic carbon atoms of the fluorene unit are preferably selected from the group consisting of alkyl, for example C₁₋₂₀ alkyl, wherein one or more non-adjacent C atoms may be replaced with O, S, NH or substituted N, C=O and -COO-, optionally substituted aryl, optionally substituted heteroaryl, alkoxy, alkylthio, fluorine, cyano and arylalkyl. Particularly preferred substituents include C₁₋₂₀ alkyl and substituted or unsubstituted aryl, for example phenyl. Optional substituents for the aryl include one or more C₁₋₂₀ alkyl groups.

Where present, substituted N may independently in each occurrence be NR⁶ wherein R⁶ is alkyl, optionally C₁₋₂₀ alkyl, or optionally substituted aryl or heteroaryl. Optional substituents for aryl or heteroaryl R⁶ may be selected from R⁵.

Preferably, each R⁹ is selected from the group consisting of C₁₋₂₀ alkyl and optionally substituted phenyl. Optional substituents for phenyl include one or more C₁₋₂₀ alkyl groups.

The repeat unit of formula (IX) may be a 2,7-linked repeat unit of formula (IXa):

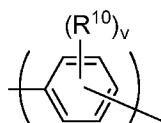


(IXa)

In one embodiment, the repeat unit of formula (IXa) is not substituted in a position adjacent to the 2- or 7- positions.

The extent of conjugation of repeat units of formulae (IX) may be limited by (a) linking the repeat unit through the 3- and / or 6- positions to limit the extent of conjugation across the repeat unit, and / or (b) substituting the repeat unit with one or more further substituents R^9 in or more positions adjacent to the linking positions in order to create a twist with the adjacent repeat unit or units, for example a 2,7-linked fluorene carrying a C_{1-20} alkyl substituent in one or both of the 3- and 6-positions.

Another exemplary class of arylene repeat units is a phenylene repeat unit that may be unsubstituted or substituted with one or more substituents, such as a repeat unit comprising a phenylene group of formula (X):



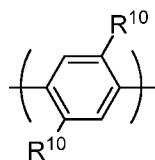
(X)

wherein v is 0, 1, 2, 3 or 4, optionally 1 or 2, and R^{10} independently in each occurrence is a substituent, optionally a substituent R^9 as described above with reference to formula (IX), for example C_{1-20} alkyl, and phenyl that is unsubstituted or substituted with one or more C_{1-20} alkyl groups.

The repeat unit of formula (X) may be 1,4- linked, 1,2-linked or 1,3-linked.

If the repeat unit of formula (X) is 1,4-linked and if v is 0 then the extent of conjugation of repeat unit of formula (X) to one or both adjacent repeat units may be relatively high.

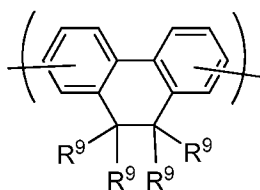
If v is at least 1, and / or the repeat unit is 1,2- or 1,3 linked, then the extent of conjugation of repeat unit of formula (X) to one or both adjacent repeat units may be relatively low. In one preferred arrangement, the repeat unit of formula (X) is 1,3-linked and v is 0, 1, 2 or 3. In another preferred arrangement, the repeat unit of formula (X) has formula (Xa):



(Xa)

Further exemplary co-repeat units include repeat units comprising triazine.

Triazine-containing materials are described in more detail in WO 2008/025997 and include, for example, optionally substituted triphenyltriazine. Another exemplary co-repeat unit has formula (XIV):



(XIV)

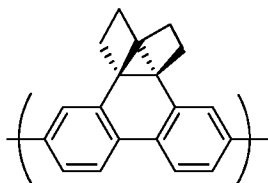
wherein R^9 is as described with reference to formula (IX) above. Any of the R^9 groups may be linked to any other of the R^9 groups to form a ring. The ring so formed may be unsubstituted or may be substituted with one or more substituents, optionally one or more C_{1-20} alkyl groups.

Exemplary repeat units of formula (XIV) include units linked through 2,7- positions and through 3,6-positions.

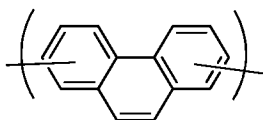
One or more of the aromatic carbon atoms of the repeat unit of formula (XIV) may be substituted. Substituents may be selected from the group consisting of alkyl, for example

C₁₋₂₀ alkyl, wherein one or more non-adjacent C atoms may be replaced with O, S, NH or substituted N, C=O and -COO-, optionally substituted aryl, optionally substituted heteroaryl, alkoxy, alkylthio, fluorine, cyano and arylalkyl. Particularly preferred substituents include C₁₋₂₀ alkyl and substituted or unsubstituted aryl, for example phenyl. Optional substituents for the aryl include one or more C₁₋₂₀ alkyl groups.

An exemplary repeat unit of formula (XIV) has the following structure, wherein aromatic carbon atoms may each independently be unsubstituted or substituted with a substituent as described above, and wherein the cyclopentyl groups may each independently be unsubstituted or substituted with one or more substituents, for example one or more C₁₋₂₀ alkyl groups:



Another exemplary co-repeat unit is phenanthrene, for example phenanthrene repeat units of formula (XV):



(XV)

One or more of the carbon atoms of the repeat unit of formula (XV) may be substituted with a substituent R¹⁰ as described with reference to formula (X). Optionally, at least the 9- and 10- positions of the phenanthrene repeat unit of formula (XV) are substituted.

An exemplary triazine-comprising co-repeat unit of the polymer according to the present invention has formula (XI):



(XI)

wherein Ar^8 in each occurrence is independently selected from aryl or heteroaryl groups, each of which may be unsubstituted or substituted with one or more substituents, and z in each occurrence is independently at least 1, optionally 1, 2 or 3. Preferably, Ar^8 in each occurrence is aryl, optionally phenyl.

Preferred substituents are selected from the group R^3 consisting of:

alkyl, for example C_{1-20} alkyl, wherein one or more non-adjacent C atoms may be replaced with O, S, substituted N, $\text{C}=\text{O}$ and $-\text{COO}-$ and one or more H atoms of the alkyl group may be replaced with F or aryl or heteroaryl optionally substituted with one or more groups R^4 ,

aryl or heteroaryl optionally substituted with one or more groups R^4 ,

NR^5_2 , OR^5 , SR^5 ,

fluorine, nitro and cyano;

wherein each R^4 is independently alkyl, for example C_{1-20} alkyl, in which one or more non-adjacent C atoms may be replaced with O, S, substituted N, $\text{C}=\text{O}$ and $-\text{COO}-$ and one or more H atoms of the alkyl group may be replaced with F, and each R^5 is independently selected from the group consisting of alkyl and aryl or heteroaryl optionally substituted with one or more alkyl groups.

Preferably, each Ar^8 of formula (XI) is phenyl, each phenyl being optionally and independently substituted with one or more C_{1-20} alkyl groups.

The extent of conjugation of the polymer backbone may be limited to avoid quenching of emission from the metal complex of repeat units (Ia), (Ib) or (Ic). The extent of

conjugation of the polymer may be controlled by selection of the linking position of the repeat unit of formula (Ia), (Ib) or (Ic) and / or the linking position of co-repeat units, as described above.

Additionally or alternatively, conjugation of the polymer may be limited by repeat units that form a twist in the polymer backbone and / or conjugation-breaking repeat units.

An example of a repeat unit that may cause a twist in the polymer backbone (for example by steric hindrance) is 1,4-phenylene substituted with one or more groups such as one or more alkyl or alkoxy groups, e.g. C₁₋₂₀ alkyl or alkoxy groups, in particular 2,5-disubstituted-1,4-phenylene repeat units.

A conjugation breaking repeat unit that breaks conjugation between repeat units either side of the conjugation breaking repeat unit include repeat units having formula $-\text{Ar}^9-\text{Sp}^1-\text{Ar}^9-$ wherein each Ar^9 is an optionally substituted aryl or heteroaryl group and Sp^1 is a spacer atom or chain comprising at least one non-conjugating atom between the two Ar groups. Exemplary Ar groups include optionally substituted phenyl. Optional substituents may be one or more substituents R^3 as described above, in particular one or more alkyl or alkoxy groups, e.g. C₁₋₂₀ alkyl or alkoxy groups. Exemplary groups Sp^1 include groups of formula $-(\text{CH}_2)_m-$ wherein m is at least 1, for example an integer between 1-10, and wherein each H may independently be replaced with a substituent, for example an alkyl group, and wherein one or more carbon atoms may be replaced with a heteroatom, for example O or S.

Formulations

In a further aspect the invention provides a formulation comprising a polymer according to the invention and at least one solvent.

A polymer of the invention may be dispersed or dissolved in a solvent or mixture of two or more solvents to form a formulation that may be used to form a layer containing the polymer by depositing the formulation and evaporating the solvent or solvents. The formulation may contain one or more further materials in addition to a polymer of the invention. All of the components of the formulation may be dissolved in the solvent or

solvent mixture, in which case the formulation is a solution, or one or more components may be dispersed in the solvent or solvent mixture. Exemplary solvents for use alone or in a solvent mixture include aromatic compounds, preferably benzene, that may be unsubstituted or substituted with one or more substituents selected from C₁₋₁₀ alkyl; C₁₋₁₀ alkoxy; and halogens, preferably chlorine, for example toluene, xylene or anisole.

White light emitting device

A device containing a polymer of the invention may emit white light.

Optionally, white-emitting devices of the invention have a colour rendering index (CRI) higher than 70.

Optionally, the white light may have CIE x coordinate equivalent to that emitted by a black body at a temperature in the range of 2500-9000K and a CIE y coordinate within 0.05 or 0.025 of the CIE y co-ordinate of said light emitted by a black body, optionally a CIE x coordinate equivalent to that emitted by a black body at a temperature in the range of 2700-4500K.

White light may be obtained by using a combination of red, green and blue light-emitting materials or a combination of blue and yellow light-emitting materials, wherein one of the light-emitting materials is a polymer according to the invention.

Multiple light-emitting layers may be arranged as described with reference to Figure 1B. In another arrangement, two or more light-emitting materials may be provided in a single light-emitting layer.

Exemplary blue light-emitting materials optionally have a peak photoluminescence wavelength less than or equal to about 480 nm, optionally about 400-480 nm.

A green light-emitting material may optionally have a photoluminescent spectrum with a peak in the range of greater than 480 nm – 540 nm, optionally about 490-540 nm.

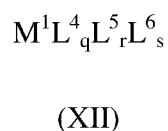
A red light-emitting material may optionally have a peak in its photoluminescent emission spectrum at more than 580 nm – 630 nm, optionally about more than 580 up to about 630 nm.

A yellow light-emitting material may optionally have a photoluminescent spectrum with a peak in the range of more than about 540 up to 580 nm.

Further light- emitting materials

In the case where a device according to the invention contains a light-emitting polymer of the invention and one or more further light-emitting materials, the further light-emitting material or materials may be selected from fluorescent and phosphorescent light-emitting materials. Further light-emitting materials may be small molecule, polymeric or dendrimeric materials.

Suitable further phosphorescent light-emitting materials include metal complexes comprising optionally substituted complexes of formula (XII):



wherein M^1 is a metal; each of L^4 , L^5 and L^6 is a coordinating group; q is an integer; r and s are each independently 0 or an integer; and the sum of $(a. q) + (b. r) + (c.s)$ is equal to the number of coordination sites available on M , wherein a is the number of coordination sites on L^4 , b is the number of coordination sites on L^5 and c is the number of coordination sites on L^6 .

Suitable metals M include d-block metals, in particular those in rows 2 and 3 i.e. elements 39 to 48 and 72 to 80, in particular ruthenium, rhodium, palladium, rhenium, osmium, iridium, platinum and gold. Iridium is particularly preferred.

Ligands L^4 , L^5 and L^6 are preferably selected from unsubstituted or substituted ligands of formula (III) as described above. Exemplary substituents include fluorine; trifluoromethyl; C_{1-40} hydrocarbyl, for example C_{1-20} alkyl; C_{1-20} alkoxy groups, and dendrons. Dendron substituents may be as described above with reference to formulae (VIII) and (VIIIa).

Further phosphorescent light-emitting materials are preferably used in combination with a small molecule or polymeric host material. Preferably, the polymeric host has a T_1 excited state energy level that is higher than that of the further phosphorescent light-emitting material it is used with. Polymeric hosts include polymers having a conjugated backbone and polymers having a non-conjugated backbone. If used with a polymeric host, a further phosphorescent light-emitting material may be mixed with the host material or covalently bound in the main chain of the polymeric host or provided as a side-chain substituent or end group of the polymer.

Exemplary fluorescent further light-emitting materials may be any form of material, including small molecule, polymeric and dendrimeric materials. Blue fluorescent polymeric materials include materials that have a partially conjugated, fully conjugated or non-conjugated polymer backbone.

An exemplary blue emitter is an at least partially conjugated polymeric emitter comprising optionally substituted (hetero)arylene repeat units, for example optionally substituted fluorene, phenylene and / or indenofluorene repeat units, and / or (hetero)arylamine repeat units.

An exemplary blue emitter comprises at least 50 mol % of fluorene repeat units and up to 50 mol %, up to 30 mol % or up to 15 mol % of (hetero)arylamine repeat units.

Charge transporting layers

A hole transporting layer may be provided between the anode and the light-emitting layers. Likewise, an electron transporting layer may be provided between the cathode and the light-emitting layers.

Similarly, an electron blocking layer may be provided between the anode and the light-emitting layer and a hole blocking layer may be provided between the cathode and the light-emitting layer. Transporting and blocking layers may be used in combination. Depending on its HOMO and LUMO levels, a single layer may both transport one of holes and electrons and block the other of holes and electrons.

If present, a hole transporting layer located between the anode and the light-emitting layers preferably has a HOMO level of less than or equal to 5.5 eV, more preferably around 4.8-5.5 eV. The HOMO level of the hole transport layer may be selected so as to be within 0.2 eV, optionally within 0.1 eV, of an adjacent layer (such as a light-emitting layer) in order to provide a small barrier to hole transport between these layers.

If present, an electron transporting layer located between the light-emitting layers and cathode preferably has a LUMO level of around 2.5-3.5 eV. For example, a layer of a silicon monoxide or silicon dioxide or other thin dielectric layer having thickness in the range of 0.2-2nm may be provided between the light-emitting layer nearest the cathode and the cathode. HOMO and LUMO levels may be measured by cyclic voltammetry.

A hole transporting layer may contain a hole-transporting (hetero)arylamine, such as a homopolymer or copolymer comprising hole transporting arylamine repeat units.

A hole-transporting layer may be cross-linkable, in particular if it is formed by depositing a hole transporting material from a solution in a solvent.

Polymer synthesis

Preferred methods for preparation of conjugated polymers comprising repeat units of formula (Ia),(Ib) and / or (Ic) as described above, comprise a "metal insertion" wherein the metal atom of a metal complex catalyst is inserted between an aryl or heteroaryl group and a leaving group of a monomer. Exemplary metal insertion methods are Suzuki polymerisation as described in, for example, WO 00/53656 and Yamamoto polymerisation as described in, for example, T. Yamamoto, "Electrically Conducting And Thermally Stable pi - Conjugated Poly(arylene)s Prepared by Organometallic Processes", Progress in Polymer Science 1993, 17, 1153-1205. In the case of Yamamoto

polymerisation, a nickel complex catalyst is used; in the case of Suzuki polymerisation, a palladium complex catalyst is used.

For example, in the synthesis of a linear polymer by Yamamoto polymerisation, a monomer having two reactive halogen groups is used. Similarly, according to the method of Suzuki polymerisation, at least one reactive group is a boron derivative group such as a boronic acid or boronic ester and the other reactive group is a halogen. Preferred halogens are chlorine, bromine and iodine, most preferably bromine.

It will therefore be appreciated that repeat units illustrated throughout this application may be derived from a monomer carrying suitable leaving groups. Likewise, an end group or side group may be bound to the polymer by reaction of a suitable leaving group.

Suzuki polymerisation may be used to prepare regioregular, block and random copolymers. In particular, homopolymers or random copolymers may be prepared when one reactive group is a halogen and the other reactive group is a boron derivative group. Alternatively, block or regioregular copolymers may be prepared when both reactive groups of a first monomer are boron and both reactive groups of a second monomer are halogen.

As alternatives to halides, other leaving groups capable of participating in metal insertion include sulfonic acids and sulfonic acid esters such as tosylate, mesylate and triflate.

Hole injection layers

A conductive hole injection layer, which may be formed from a conductive organic or inorganic material, may be provided between the anode and the light-emitting layers to assist hole injection from the anode into the layer or layers of semiconducting polymer. A hole transporting layer may be used in combination with a hole injection layer.

Examples of doped organic hole injection materials include optionally substituted, doped poly(ethylene dioxythiophene) (PEDT), in particular PEDT doped with a charge-balancing polyacid such as polystyrene sulfonate (PSS) as disclosed in EP 0901176 and EP 0947123, polyacrylic acid or a fluorinated sulfonic acid, for example Nafion®;

polyaniline as disclosed in US 5723873 and US 5798170; and optionally substituted polythiophene or poly(thienothiophene). Examples of conductive inorganic materials include transition metal oxides such as VOx MoOx and RuOx as disclosed in Journal of Physics D: Applied Physics (1996), 29(11), 2750-2753.

Cathode

The cathode is selected from materials that have a work function allowing injection of electrons into the light-emitting layer or layers of the device. Other factors influence the selection of the cathode such as the possibility of adverse interactions between the cathode and the light-emitting materials. The cathode may consist of a single material such as a layer of aluminium. Alternatively, it may comprise a plurality of metals, for example a bilayer of a low workfunction material and a high workfunction material such as calcium and aluminium as disclosed in WO 98/10621. The cathode may contain a layer of elemental barium, for example as disclosed in WO 98/57381, Appl. Phys. Lett. 2002, 81(4), 634 and WO 02/84759. The cathode may contain one or more conductive layers, for example one or more metal layers, and a thin (e.g. 1-5 nm) layer of a metal compound between the one or more conductive layers and the light-emitting layer or layers. Exemplary metal compounds include oxides or fluorides of an alkali or alkali earth metal, for example lithium fluoride as disclosed in WO 00/48258; barium fluoride as disclosed in Appl. Phys. Lett. 2001, 79(5), 2001; and barium oxide. In order to provide efficient injection of electrons into the device, the cathode preferably has a workfunction of less than 3.5 eV, more preferably less than 3.2 eV, most preferably less than 3 eV. Work functions of metals can be found in, for example, Michaelson, J. Appl. Phys. 48(11), 4729, 1977.

The cathode may be opaque or transparent. Transparent cathodes are particularly advantageous for active matrix devices because emission through a transparent anode in such devices is at least partially blocked by drive circuitry located underneath the emissive pixels. A transparent cathode comprises a layer of an electron injecting material that is sufficiently thin to be transparent. Typically, the lateral conductivity of this layer will be low as a result of its thinness. In this case, the layer of electron injecting material

is used in combination with a thicker layer of transparent conducting material such as indium tin oxide.

It will be appreciated that a transparent cathode device need not have a transparent anode (unless, of course, a fully transparent device is desired), and so the transparent anode used for bottom-emitting devices may be replaced or supplemented with a layer of reflective material such as a layer of aluminium. Examples of transparent cathode devices are disclosed in, for example, GB 2348316.

Encapsulation

Organic optoelectronic devices tend to be sensitive to moisture and oxygen. Accordingly, the substrate preferably has good barrier properties for prevention of ingress of moisture and oxygen into the device. The substrate is commonly glass, however alternative substrates may be used, in particular where flexibility of the device is desirable. For example, the substrate may comprise a plastic as in US 6268695 which discloses a substrate of alternating plastic and barrier layers or a laminate of thin glass and plastic as disclosed in EP 0949850.

The device may be encapsulated with an encapsulant (not shown) to prevent ingress of moisture and oxygen. Suitable encapsulants include a sheet of glass, films having suitable barrier properties such as silicon dioxide, silicon monoxide, silicon nitride or alternating stacks of polymer and dielectric as disclosed in, for example, WO 01/81649 or an airtight container as disclosed in, for example, WO 01/19142. In the case of a transparent cathode device, a transparent encapsulating layer such as silicon monoxide or silicon dioxide may be deposited to micron levels of thickness, although in one preferred embodiment the thickness of such a layer is in the range of 20-300 nm. A getter material for absorption of any atmospheric moisture and / or oxygen that may permeate through the substrate or encapsulant may be disposed between the substrate and the encapsulant.

Formulation and solution processing

Suitable solvents for forming formulations of the polymer for solution processing include many common organic solvents, such as mono- or poly-alkylbenzenes such as toluene and xylene.

Exemplary solution deposition techniques include printing and coating techniques, such as spin-coating, dip-coating, roll-to-roll coating or roll-to-roll printing, doctor blade coating, slot die coating, gravure printing, screen printing and inkjet printing.

Multiple organic layers of an OLED may be formed by deposition of formulations containing the active materials for each layer.

Coating methods, such as spin-coating described above, are particularly suitable for devices wherein patterning of the electroluminescent material is unnecessary, for example for lighting applications or simple monochrome segmented displays.

Printing is particularly suitable for high information content displays, in particular full colour displays. A device may be inkjet printed by providing a patterned layer over the first electrode and defining wells for printing of one colour (in the case of a monochrome device) or multiple colours (in the case of a multicolour, in particular full colour device). The patterned layer is typically a layer of photoresist that is patterned to define wells as described in, for example, EP 0880303.

As an alternative to wells, the ink may be printed into channels defined within a patterned layer. In particular, the photoresist may be patterned to form channels which, unlike wells, extend over a plurality of pixels and which may be closed or open at the channel ends.

A number of methods may be used to at least partially avoid dissolution of an underlying organic layer during solution deposition of one or more further organic layers on the underlying organic layer. The underlying organic layer may be rendered insoluble by cross-linking prior to solution deposition of a further layer. Cross-linking may be provided by substituents on the materials forming the underlying layer, such as cross-linking substituents on the host and / or dopant material of a light-emitting layer. Where

a layer to be cross-linked contains a polymer, the cross-linkable groups may be provided as substituents of repeat units of the polymer as described above.

The polymer according to the invention may comprise one or more co- repeat units substituted with a cross-linking group. Exemplary crosslinking groups include a polymerisable double bond group such as a vinyl or acrylate group, a benzocyclobutane group, and an oxetane group, each of which may optionally be substituted.

Alternatively or additionally, a cross-linkable additive may be included in the composition used to form the underlying layer.

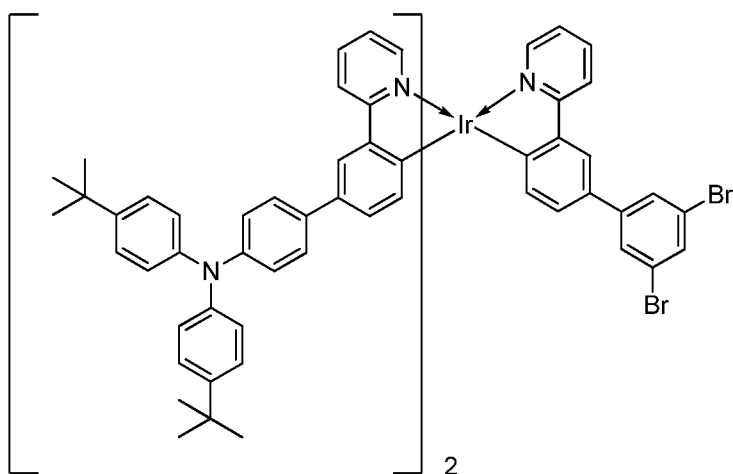
Alternatively or additionally, the further layer may be formed from a solution comprising a solvent in which the underlying organic layer is insoluble.

Applications

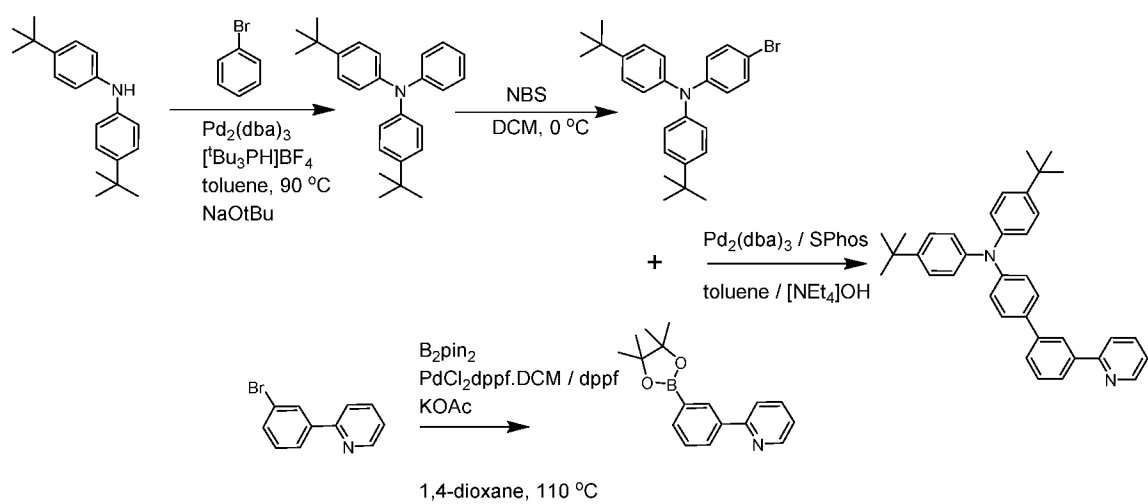
OLEDs as described herein may be used in a wide range of applications, including but not limited to use as display backlights, for example LCD backlights, area illumination and displays. White light-emitting OLEDs may comprise uniform emissive layers. OLEDs for use in displays may comprise a patterned emissive layer or layers and / or a patterned electrode to provide individual display pixels.

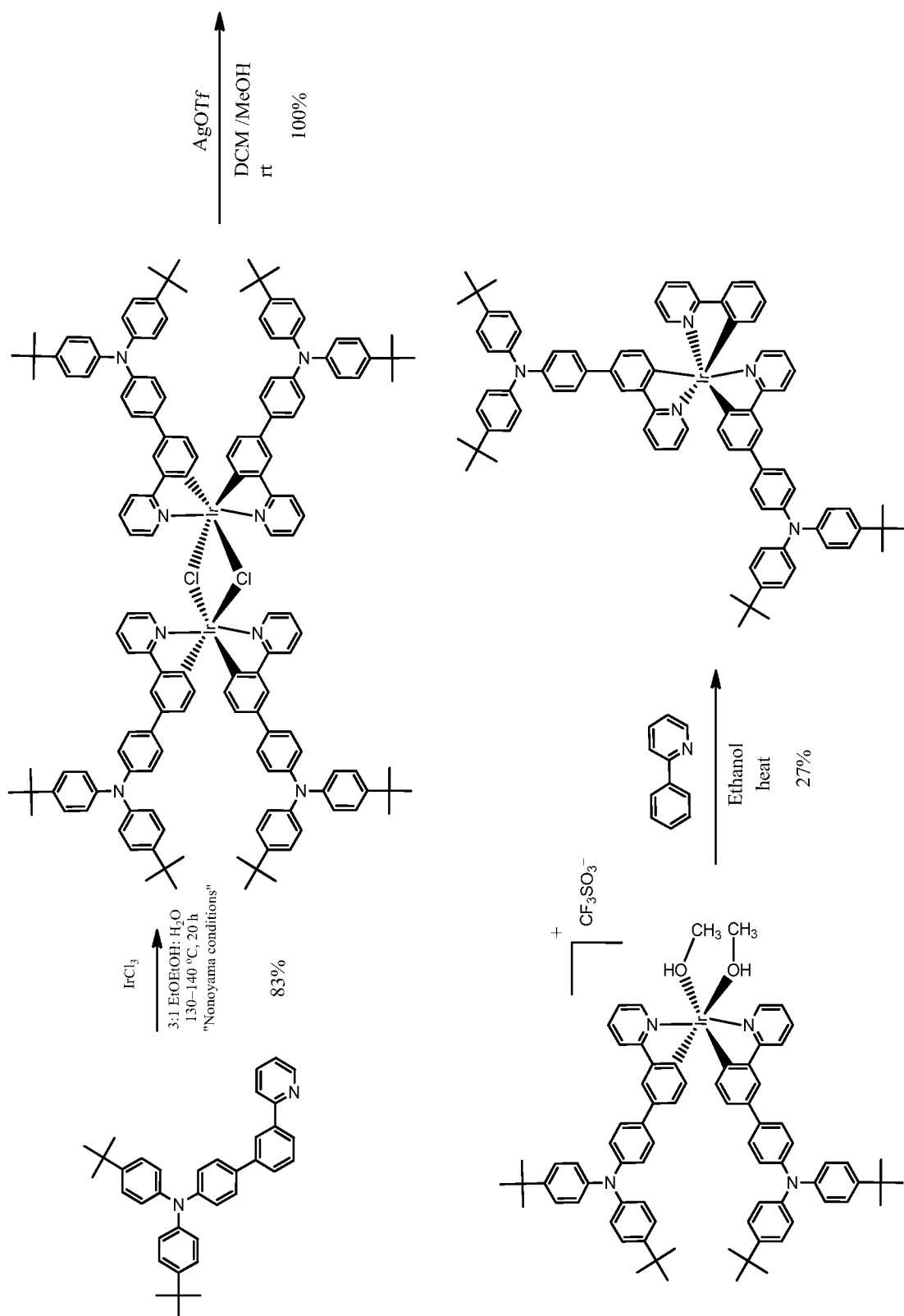
Examples

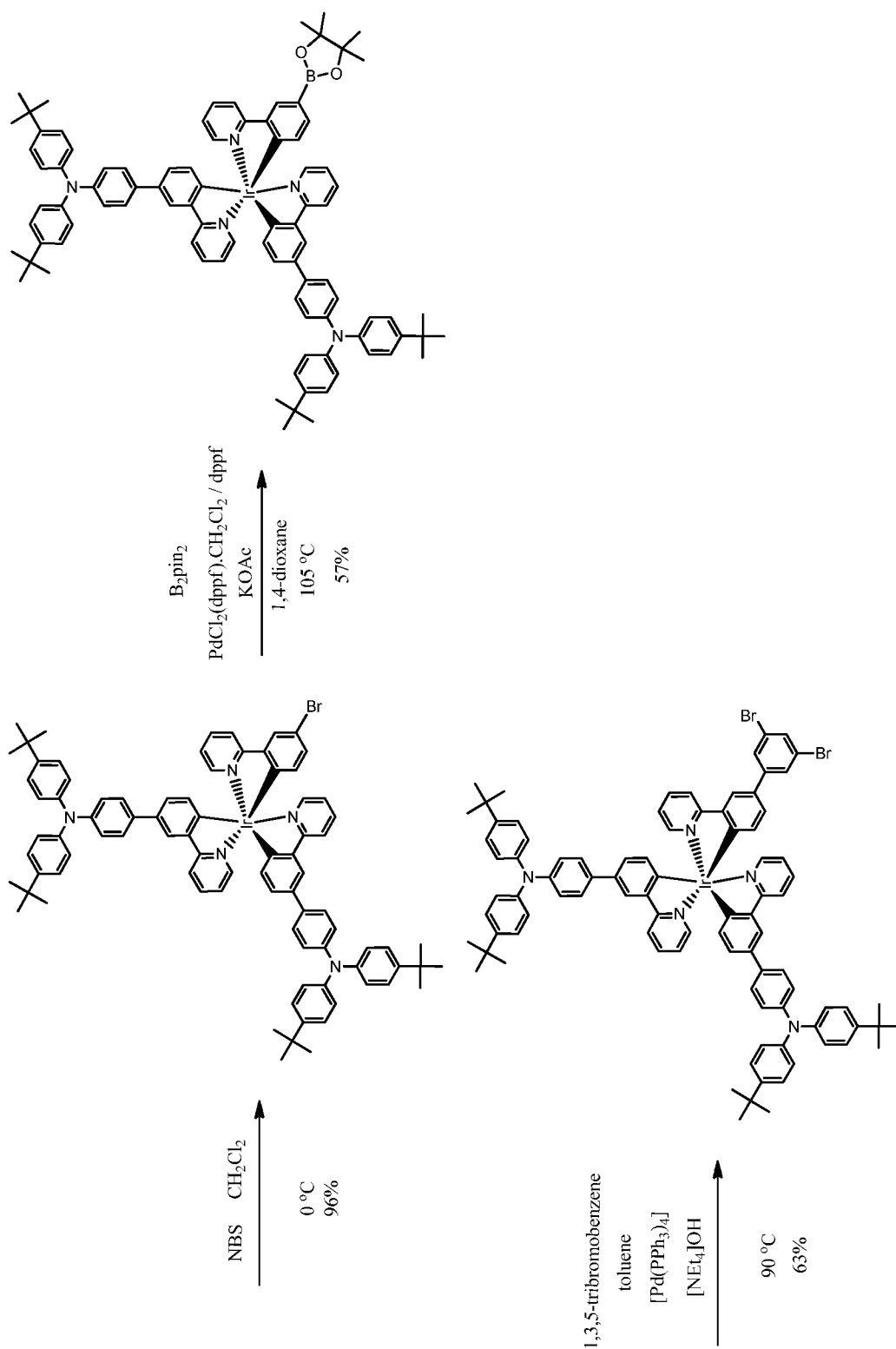
Monomer Example 1



Monomer Example 1 was prepared according to the following reaction scheme:







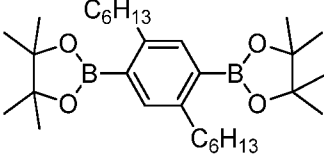
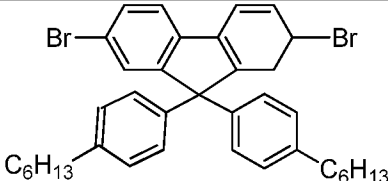
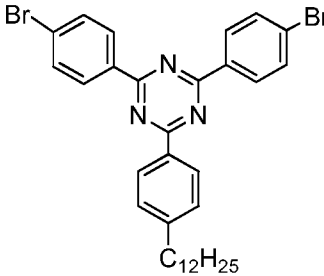
Polymer Example 1 and Comparative Polymer 1

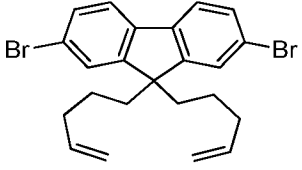
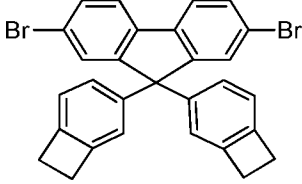
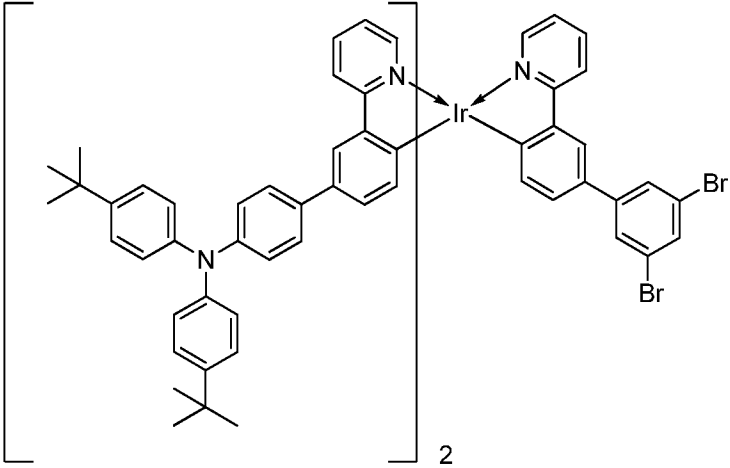
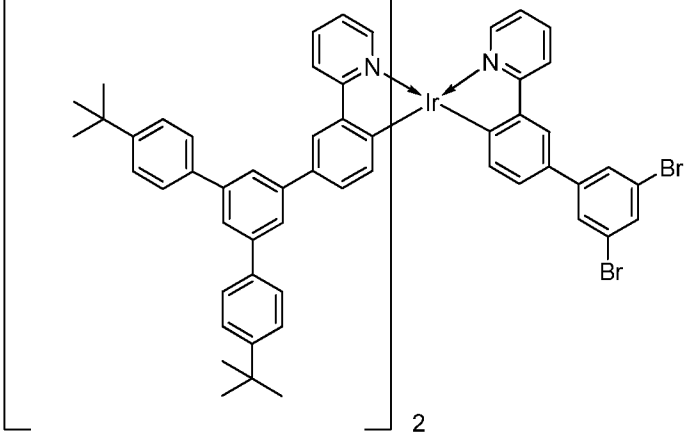
Polymer Example 1 was prepared by Suzuki polymerisation as described in WO 00/53656 of monomers including Monomer Example 1.

Comparative Polymer 1 was prepared in the same way, but using Comparative Monomer 1 in place of Monomer Example 1.

Monomers used in preparation of Polymer Example 1 and Comparative Polymer 1 are provided in Table 1.

Table 1: Polymer Example 1 and Comparative Polymer 1 monomers composition

Monomer	Polymer Example 1 (mol %)	Comparative Polymer 1 (mol %)
	50	50
	24.5	20.7
	11.5	11.5

	5	5
	5	5
 Monomer Example 1	4	-
 Comparative Monomer 1	-	7.8

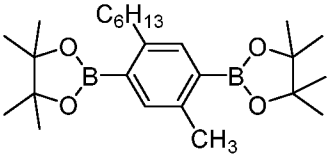
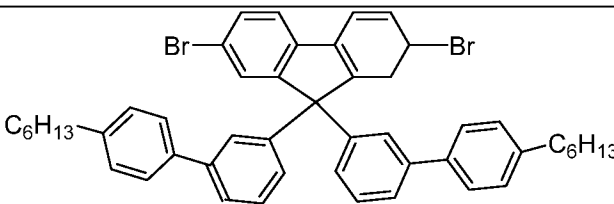
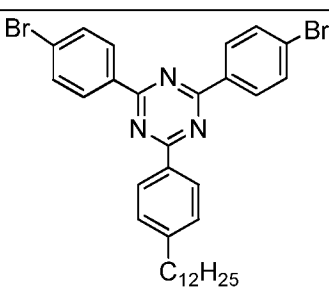
Polymer Example 2 and Comparative Polymer 2

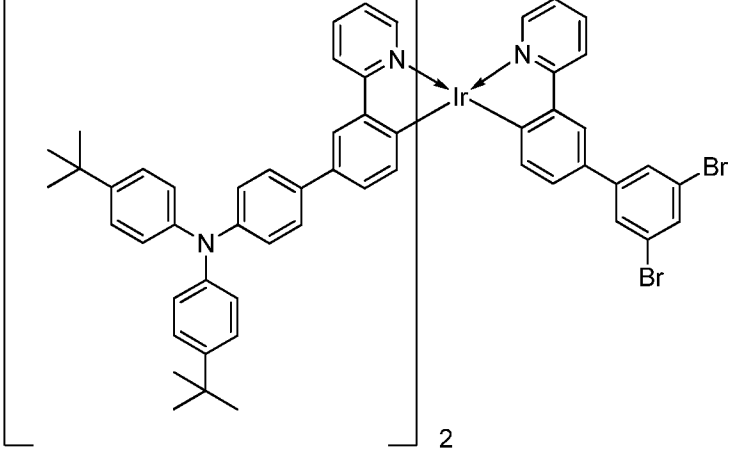
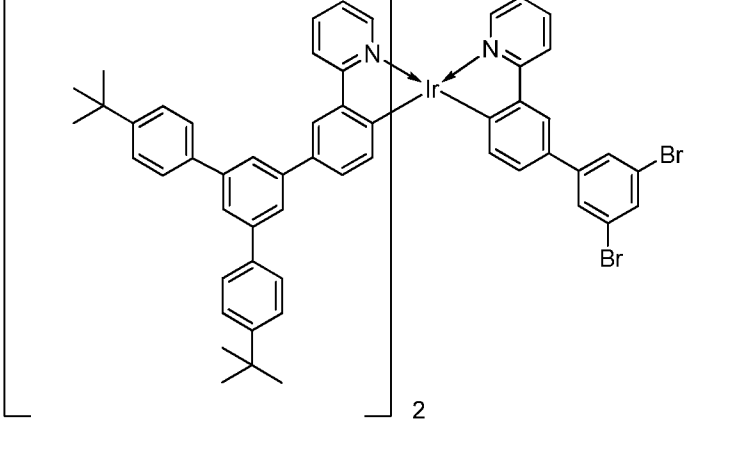
Polymer Example 2 was prepared by Suzuki polymerisation as described in WO 00/53656 of monomers including Monomer Example 1.

Comparative Polymer 2 was prepared in the same way, but using Comparative Monomer 1 in place of Monomer Example 1.

Monomers used in preparation of Polymer Example 2 and Comparative Polymer 2 are provided in Table 1.

Table 2: Polymer Example 2 and Comparative Polymer 2 monomers composition

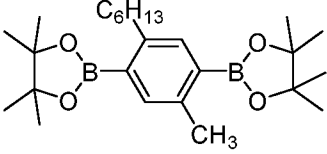
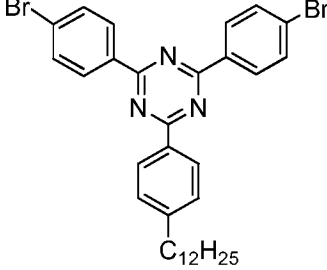
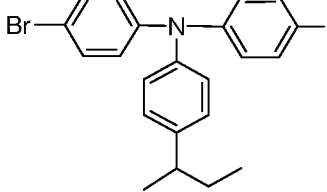
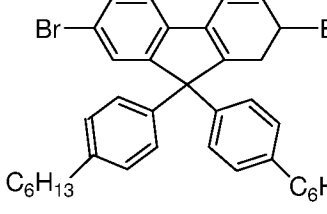
Monomer	Polymer Example 1 (mol %)	Comparative Polymer 1 (mol %)
	50	50
	35.5	33.32
	11.5	2

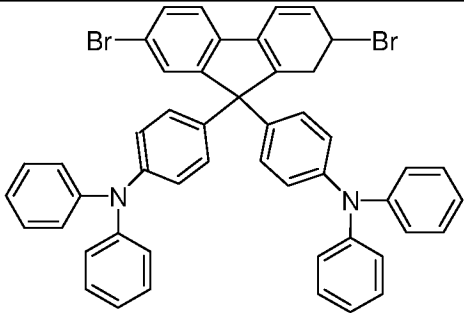
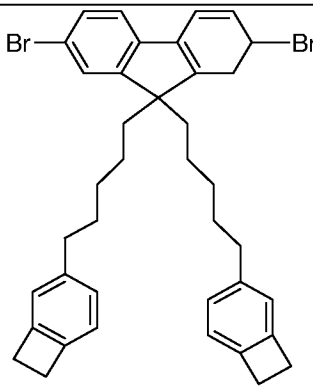
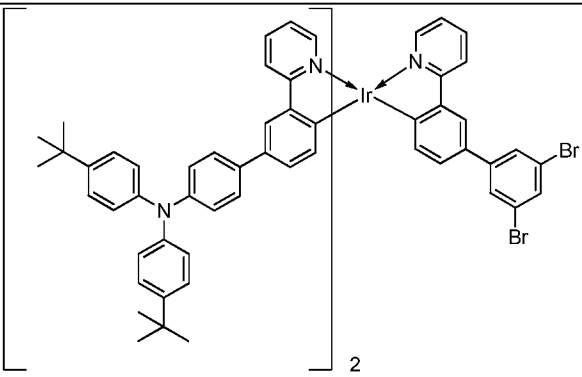
 Monomer Example 1	3	-
 Comparative Monomer 1	-	4.68

Polymer Example 3 and Comparative Polymers 3A and 3B

Polymer Example 3, Comparative Polymer 3A and Comparative Polymer 3B were prepared by Suzuki polymerisation as described in WO 00/53656 of monomers set out in Table 3 to produce polymers having molecular weight distributions as set out in Table 4.

Table 3: Polymer Example 3 and Comparative Polymers 3A and 3B monomers composition

Monomer	Polymer Example 3 (mol %)	Comparative Polymer 3A (mol %)	Comparative Polymer 3B (mol %)
	50	50	50
	11.5	11.5	11.5
	10	10	10
	24.5	20.5	22.5

	-	-	2
	10	10	10
 Monomer Example 1	4	-	-

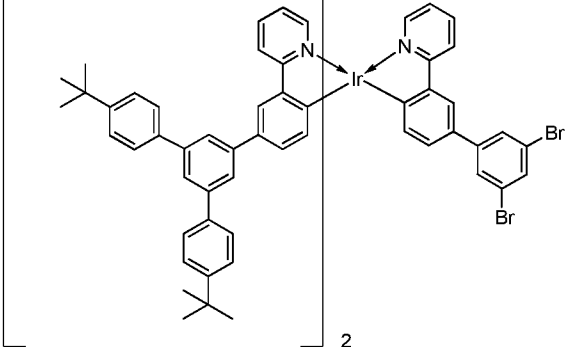
 Comparative Monomer 1	-	4	4
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Table 4: Polymer Example 3 and Comparative Polymers 3A and 3B molecular weights

	Mz	Mw	Mp	Mn	PD
Polymer Example 3	476,000	260,000	223,000	89,000	2.92
Comparative Polymer 3A	1,324,000	672,000	607,000	143,000	4.72
Comparative Polymer 3B	2,241,000	1,064,000	861,000	184,000	5.78

Device Example 1

An organic light-emitting device having the following structure was formed on a glass substrate:

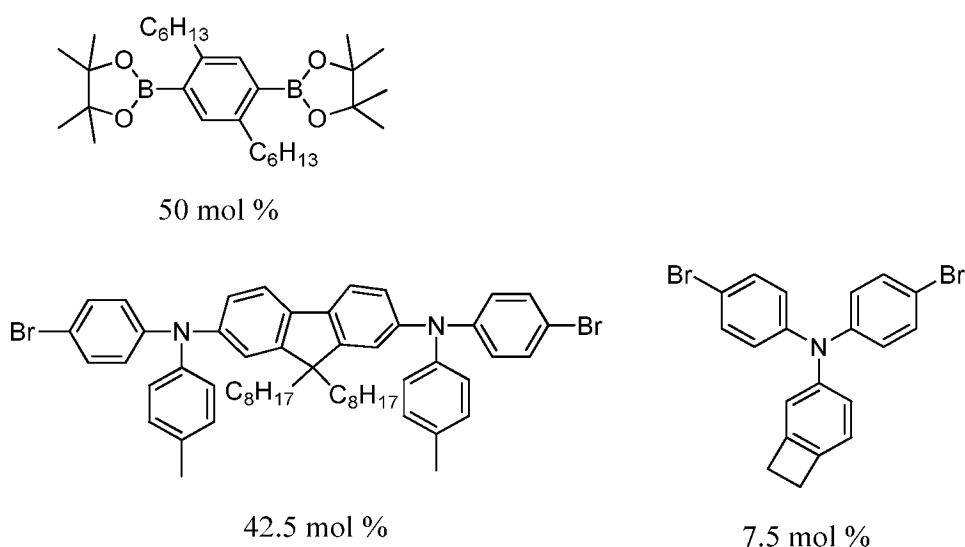
ITO / HIL (50 nm) / HTL1 (22 nm) / ELG (30 nm) / ELR (20 nm) / ELB (50 nm) / Cathode

wherein ITO is an indium-tin oxide anode; HIL is a layer of hole-injection material available from Plextronics Inc., a HTL1 is a hole transporting layer; ELG is a light-emitting layer including a green phosphorescent light-emitting material; ELR is a red

phosphorescent light-emitting layer; ELB is a blue fluorescent light-emitting layer; and Cathode is a cathode comprising a trilayer of a metal fluoride, aluminium and silver.

HIL, HTL1, ELG, ELR and ELB were each formed by spin-coating a solution comprising the components of that layer and a solvent, and evaporating the solvent. HTL1, ELG and ELR comprise crosslinkable groups that were cross-linked prior to spin-coating of the overlying layer.

HTL1 was formed by spin-coating Hole Transporting Polymer 1 formed by Suzuki polymerisation, as described in WO 00/53656, of the following monomers:



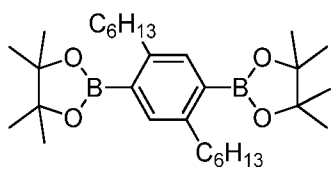
ELG was formed by spin-coating Polymer Example 1.

ELR was formed by spin-coating a 1:1 mixture by weight of Red Polymer 1 and Red Polymer 2.

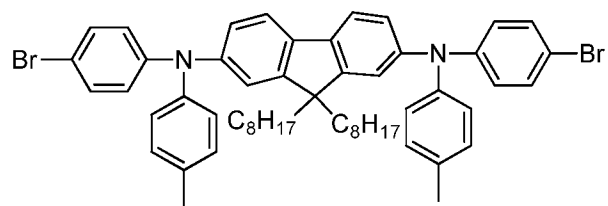
Red Polymer 1 and Red Polymer 2 were each formed by Suzuki polymerisation, as described in WO 00/53656.

Red Polymer 1 and Red Polymer 2 were formed by polymerisation of monomers and a red phosphorescent end-capping unit in the molar percentages given below:

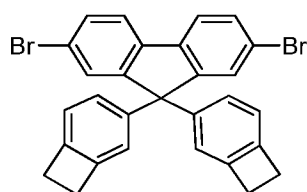
Red Polymer 1:



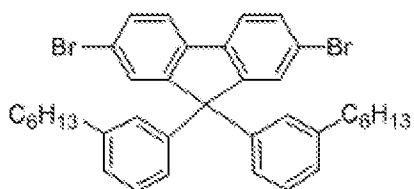
50 %



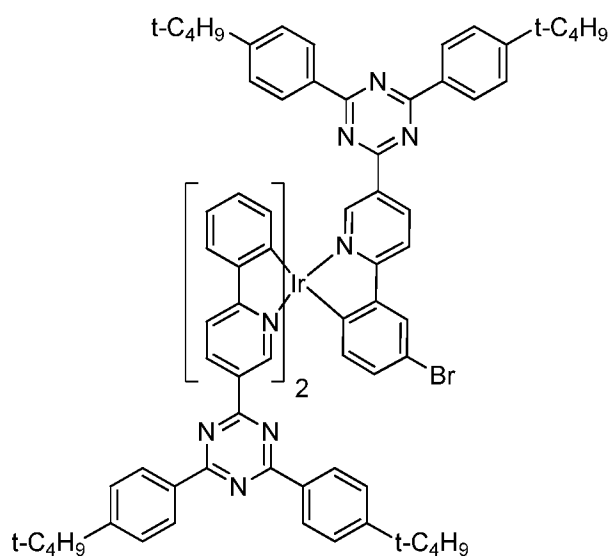
36.516%



10 %

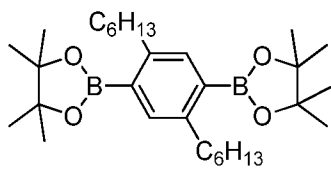


3.184 %

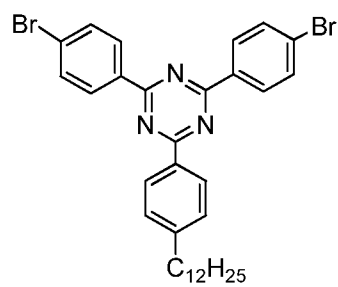


0.6 %

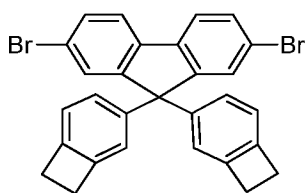
Red Polymer 2:



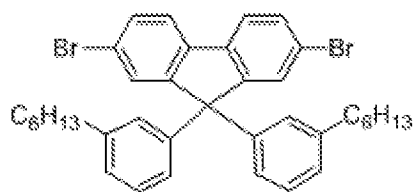
50 %



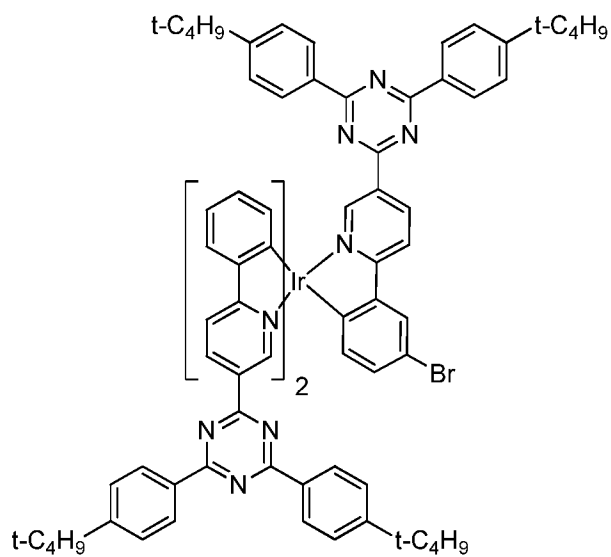
22 %



10 %

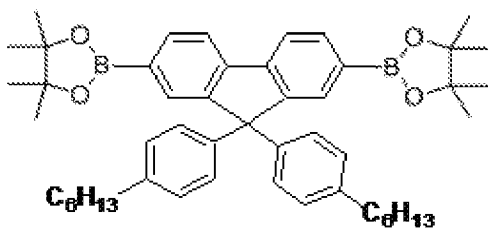


17.7 %

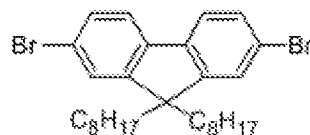


0.6 %

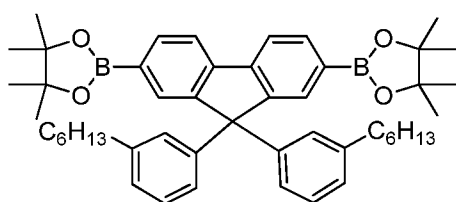
ELB was formed by spin-coating Blue Polymer 1 formed by Suzuki polymerisation, as described in WO 00/53656, of monomers in the following molar percentages:



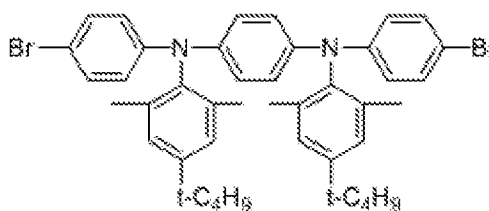
36 %



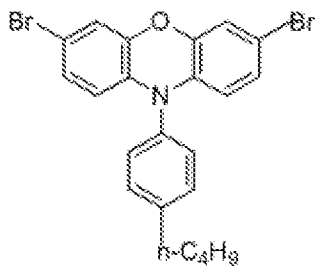
14 %



45 mol %



4 mol %



1 mol %

Comparative Device 1

A device was made as described with respect to Example 1 except that ELG was formed by spin-coating Comparative Polymer 1.

Device Example 2

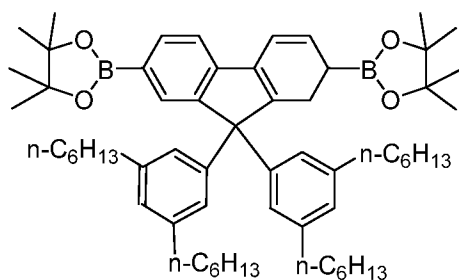
An organic light-emitting device having the following structure was formed on a glass substrate:

ITO / HIL / HTL2 / EL1 / Cathode

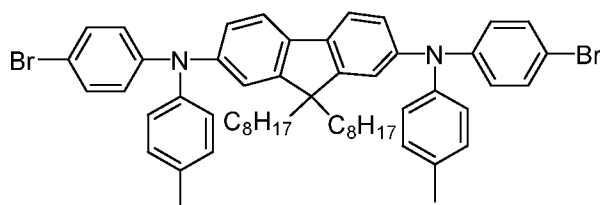
wherein ITO is an indium-tin oxide anode; HIL is a layer of hole-injection material available from Plextronics Inc., a HTL is a hole transporting layer; EL1 is a light-emitting layer; and Cathode is a cathode comprising a trilayer of a metal fluoride, aluminium and silver.

HIL, HTL2 and EL1 were each formed by spin-coating a solution comprising the components of that layer and a solvent, and evaporating the solvent. HTL comprises a crosslinkable group that was cross-linked prior to spin-coating of the overlying layer.

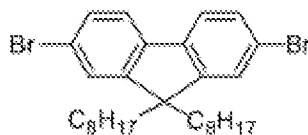
HTL2 was formed by spin-coating Hole Transporting Polymer 2 formed by Suzuki polymerisation, as described in WO 00/53656, of the following monomers:



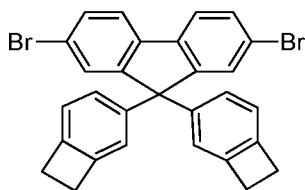
50 mol %



30 mol %



12.5 mol %



7.5 mol %

Comparative Device 2

A device was made as described with respect to Device Example 2 except that EL1 was formed by spin-coating Comparative Polymer 2.

Device Example 3

An organic light-emitting device having the following structure was formed on a glass substrate:

ITO / HIL (50 nm) / HTL3 (22 nm) / EL3 / Cathode

wherein ITO is an indium-tin oxide anode; HIL is a layer of hole-injection material; HTL3 is a hole transporting layer; EL3 is a light-emitting layer of Polymer Example 3; and Cathode is a cathode of a trilayer of a metal fluoride, aluminium and silver.

HIL, HTL3 and EL3 were each formed by spin-coating a solution comprising the components of that layer and a solvent, and evaporating the solvent. HTL3 comprises a crosslinkable group that was cross-linked prior to spin-coating of the overlying layer.

HTL3 was formed by spin-coating Hole Transporting Polymer 3 formed by Suzuki polymerisation, as described in WO 00/53656, and containing a dialkylphenylene repeat unit of formula (Xa); an amine repeat unit as described in WO 2005/049546; and fluorene repeat units of formula (IXa) substituted with crosslinkable groups.

Comparative Device 3A

A device was made as described with respect to Device Example 3 except that EL3 was formed by spin-coating Comparative Polymer 3A.

Comparative Device 3B

A device was made as described with respect to Device Example 3 except that EL3 was formed by spin-coating Comparative Polymer 3B.

Device performance

With reference to Table 3 below, it can be seen that external quantum efficiency (EQE) is higher for Device Example 1 including the phosphorescent polymer of the invention compared to Comparative Device 1. Moreover, efficiency measured in lumens / Watt is increased by 2 lm/W and the colour rendering index (CRI) is improved for Device Example 1 when compared to Comparative Device 1.

Table 5: Device performance

Device	CIE(x)	CIE(y)	EQE (%)	Efficiency (Lm/W)	Colour Rendering Index (CRI)	T70 lifetime (hours)
Comparative 1	0.474	0.429	12.8	21.6	68.8	282
Example 1	0.476	0.432	13.7	23.7	71.7	488

Figure 2 shows that the use of the phosphorescent polymer according to the present invention gives an improved injection (EQE trace starts rising at lower voltage for Example 1 device than the one for Comparative Device 1).

Figure 3 shows that the lifetime (T70) of the Example Device 1, that is the time taken for luminance of the device to decay to 70% of a starting brightness at constant current, is also improved to over 460 hours compared to 280 hours lifetime for Comparative Device 1 when tested at a starting luminance of 2000 cd/m².

The photoluminescence spectra presented on Figure 4 show a 10 nm shift of the central peak towards the yellow part of the spectrum for the polymer comprising Monomer Example 1 leading to an improved CRI when compared to Comparative Monomer 1. At the same time the CIE coordinates obtained for white emission from the example emitter and the comparative emitter are almost identical as shown in Figure 5.

Without wishing to be bound by any theory, it is believed that the presence of amine substituents in the polymers of the present invention aids hole injection and hole mobility. This provides a further advantage in that the loading of iridium complex required in the polymer of the present invention can be reduced to 4% compared to 7.8% for the polymer of Comparative Emitter 1, which is advantageous when using an expensive metal such as iridium.

Figure 6 shows that Device Example 2 and Comparative Device 2 have similar electroluminescent spectra.

The drive voltage to reach a brightness of 1000 cd/m^2 for Comparative Device 2 was 4.85V, and 4.30V for Device Example 2.

With reference to Figure 7, the electroluminescent spectrum for Device Example 3 is at a slightly longer wavelength than that of either Comparative Device 3A or 3B.

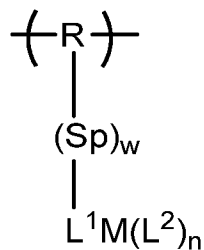
With reference to Figure 8, the time taken for the brightness of Device Example 3 to fall to 50% of its original brightness is much longer for Device Example 3 than for either Comparative Device 3A or 3B.

With reference to Figures 9 and 10, the current densities and external quantum efficiencies respectively of Device Example 3, Comparative Device 3A and Comparative Device 3B are similar.

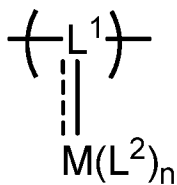
Although the present invention has been described in terms of specific exemplary embodiments, it will be appreciated that various modifications, alterations and/or combinations of features disclosed herein will be apparent to those skilled in the art without departing from the scope of the invention as set forth in the following claims.

CLAIMS

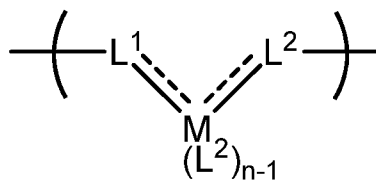
1. A polymer comprising a repeat unit of formula (I a), (I b) or (Ic):



(Ia)



(Ib)



(Ic)

wherein:

M is a metal;

R is a backbone repeat group;

n is 1 or 2;

Sp is a spacer group;

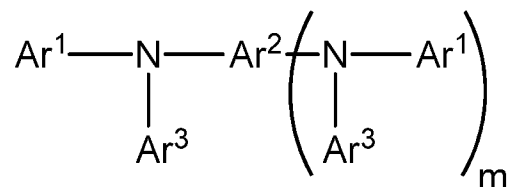
w is 0 or 1;

L¹ is a mono- or bidentate co-coordinating group;

L² which may be the same or different in each occurrence, is a mono- or bidentate coordinating group;

---- represents a second metal to ligand bond in the case where L¹ or L² is a bidentate ligand and

at least one of L¹ and L² is substituted with group of formula (II)

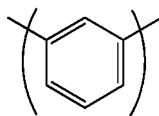


(II)

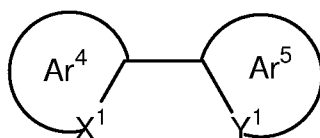
wherein m is 0 or 1;

each Ar¹, Ar² and Ar³ is independently in each occurrence selected from an aryl or heteroaryl that may be unsubstituted or substituted with one or more substituents, and any two of Ar¹, Ar² and Ar³ linked to the same N atom may be linked by a direct bond or a divalent linking group.

2. The polymer according to claim 1 wherein M is a group VIII transition metal element.
3. The polymer according to claim 2 wherein M is Iridium.
4. The polymer according to any preceding claim wherein:
R is $-(\text{Ar}^6)_p-$;
p is 1, 2, or 3; and
each Ar⁶ is independently selected from aryl or heteroaryl.
5. The polymer according to claim 4 wherein p is 1.
6. The polymer according to claim 4 wherein Ar⁶ is phenyl that may be unsubstituted or substituted with one or more substituents.
7. The polymer according to claim 5 or 6 wherein R is an unsubstituted or substituted repeat unit of formula:



8. The polymer according to any preceding claim, wherein each L^1 , and L^2 are independently selected from ligands comprising a bidentate group of formula (III):

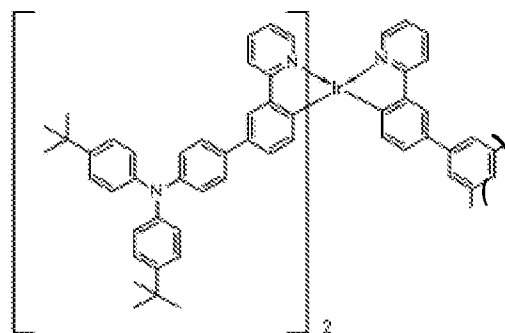


(III)

wherein Ar^4 and Ar^5 may be the same or different, may be fused together; and are independently selected from an aryl or heteroaryl that may be unsubstituted or substituted with one or more substituents; and wherein X^1 and Y^1 may be the same or different and are independently selected from carbon or nitrogen.

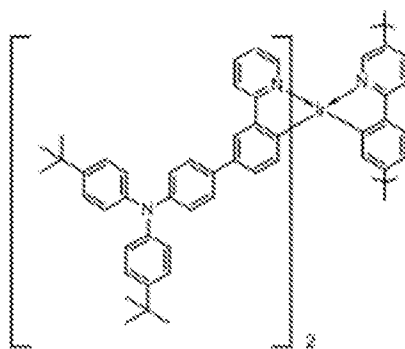
9. The polymer according to claim 8 wherein X^1 is carbon and Y^1 is nitrogen.
10. The polymer according to claim 8 or 9, wherein said bidentate group is a 2-phenylpyridine group.
11. The polymer according to any preceding claim, wherein m is 1.
12. The polymer according to any preceding claim wherein one of Ar^1 , Ar^2 and Ar^3 is bound directly to L^1 or L^2 .
13. The polymer according to any preceding claim wherein L^2 is substituted with a group of formula (II).

14. The polymer according to any preceding claim wherein Ar^1 , Ar^2 and Ar^3 are each phenyl, each of which may be unsubstituted or substituted with one or more substituents.
15. The polymer according to any preceding claim, wherein one or more of Ar^1 , Ar^2 and Ar^3 are substituted with one or more C_{1-20} alkyl substituents.
16. The polymer according to claim 15, wherein said substituent comprises a tertiary carbon atom.
17. The polymer according to claim 16, wherein said substituent comprises a tert-butyl group.
18. The polymer according to any preceding claim wherein w is 0.
19. The polymer according to any of claims 1-17 wherein w is 1 and Sp is a C_{1-30} hydrocarbyl group.
20. The polymer according to claim 18 or 19 wherein Sp is selected from the group consisting of C_{1-20} alkyl and phenyl- C_{1-20} alkyl and wherein one or more non adjacent C atoms of C_{1-20} alkyl may be replaced with O, S or COO.
21. The polymer according to any preceding claim comprising a repeat unit of formula (IV), which may be unsubstituted or substituted with one or more substituents.



(IV)

22. The polymer according to any preceding claim comprising a repeat unit of formula (V), which may be unsubstituted or substituted with one or more substituents.



(V)

23. The polymer according to any preceding claim wherein the polymer contains 0.5 – 40 mol %, optionally 0.5-20 mol % of the repeat units of formula (I a), (I b), (IV) or (V).
24. The polymer according to any preceding claim wherein a photoluminescent spectrum of the polymer has a peak at a wavelength in the range 540-620 nm..

25. The polymer according to claim 24 wherein the polymer has a peak at a wavelength of less than 600 nm.
26. The polymer according to any preceding claim comprising one or more co-repeat units.
27. The polymer according to claim 26 comprising an arylene co-repeat unit that may be unsubstituted or substituted with one or more substituents.
28. The polymer according to claim 27 wherein the arylene co-repeat unit is fluorene that may be unsubstituted or substituted with one or more substituents.
29. The polymer according to claim 27 wherein the arylene co-repeat unit is phenylene that may be unsubstituted or substituted with one or more substituents.
30. The polymer according to any of claims 26-29 comprising a repeat unit comprising triazine.
31. The polymer according to any preceding claim comprising a cross-linkable repeat unit.
32. A formulation comprising a polymer according to any of claims 1-31 and at least one solvent.
33. An organic light-emitting device comprising an anode, a cathode and a light-emitting layer between the anode and the cathode, wherein the light-emitting layer comprises a polymer according to any one of claims 1-31.
34. The organic light-emitting device according to claim 33 comprising a first light emitting layer, a second and a third light-emitting layer, wherein the second light-emitting layer is disposed between the first light emitting layer and a third light

emitting layer and wherein one of the first, second and third light-emitting layers comprises the polymer according to any of claims 1-31.

35. The organic light-emitting device according to claim 33 or 34, comprising a hole transporting layer between the anode and the light emitting layer comprising the polymer according to any one of claims 1-31.
36. The organic light-emitting device according to any of claims 33-35 wherein the device is a white light emitting device.
37. A method of forming the organic light-emitting device of any one of claims 33-36, the method comprising the step of forming the light-emitting layer over one of the anode or cathode, and forming the other of the anode or cathode over the light-emitting layer.
38. The method of claim 37, wherein a light-emitting layer is formed by depositing the formulation according to claim 32 and evaporating the at least one solvent.
39. A method of forming an organic light-emitting device according to any of claims 34-36 comprising the steps of: depositing the first light-emitting layer over one of the anode and cathode; depositing the second light-emitting layer over the first light-emitting layer; depositing the third light-emitting layer over the second light-emitting layer; and depositing the other of the anode and cathode over the third light-emitting layer.
40. A method according to claim 39 wherein at least one of the first, second and third light-emitting layers is formed by a solution processing method comprising the steps of depositing a composition comprising a solvent and the component or components of the light-emitting layer and evaporating the solvent.

41. A method according to claim 40 wherein the first light-emitting layer is cross-linked prior to formation of the second light-emitting layer by the solution processing method.
42. A method according to claim 40 or 41 wherein the second light-emitting layer is cross-linked prior to formation of the third light-emitting layer by the solution processing method.

FIGURE 1A

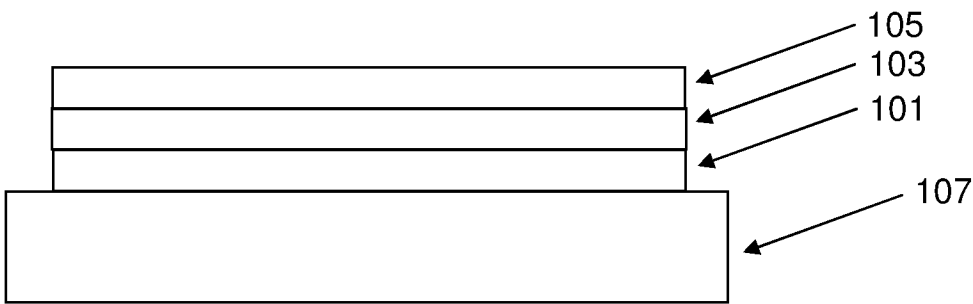


FIGURE 1B

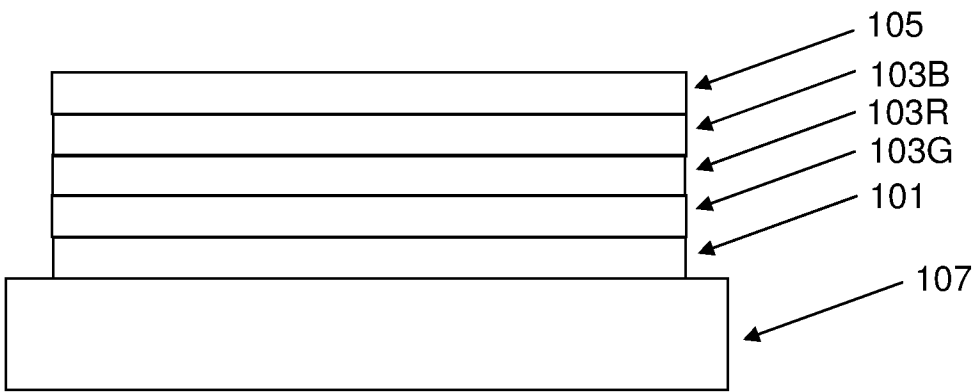


FIGURE 2

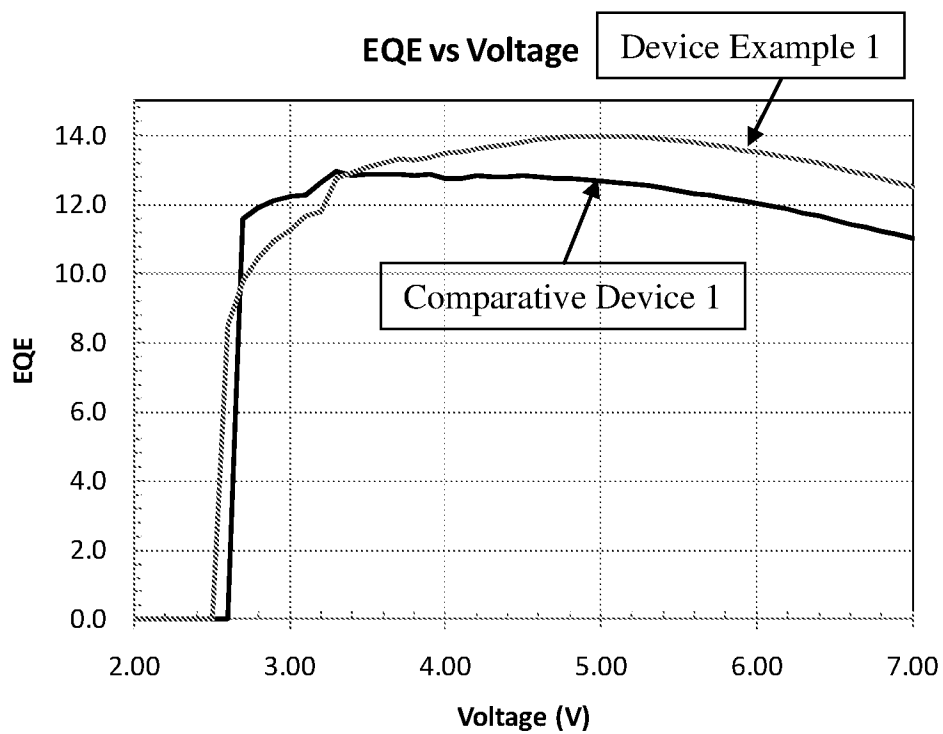


FIGURE 3

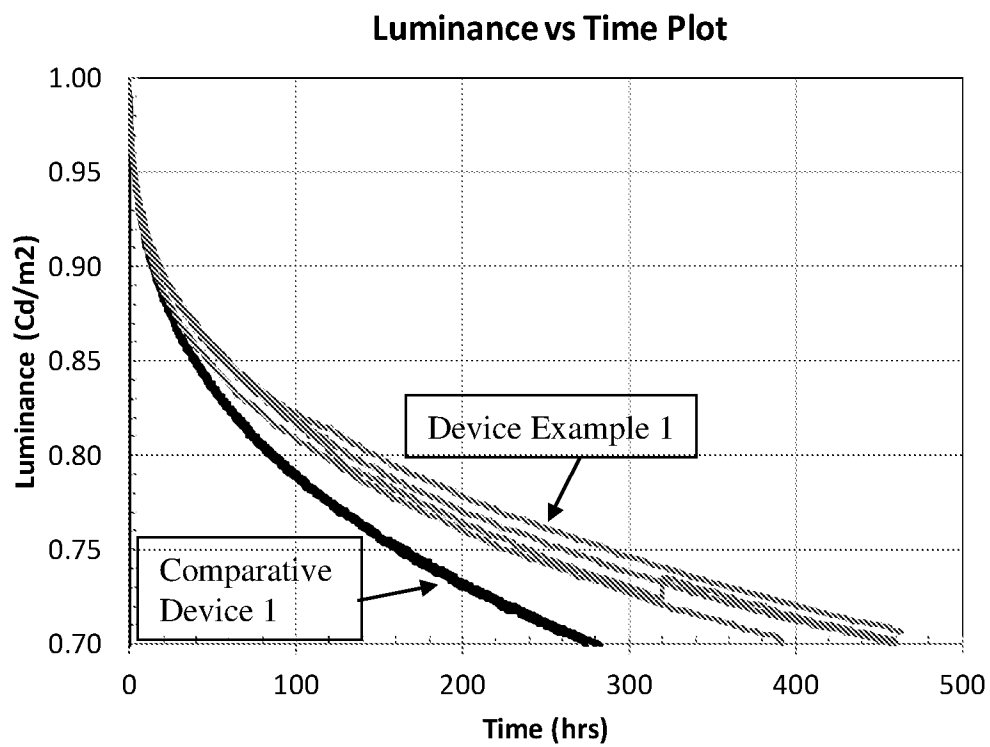


FIGURE 4

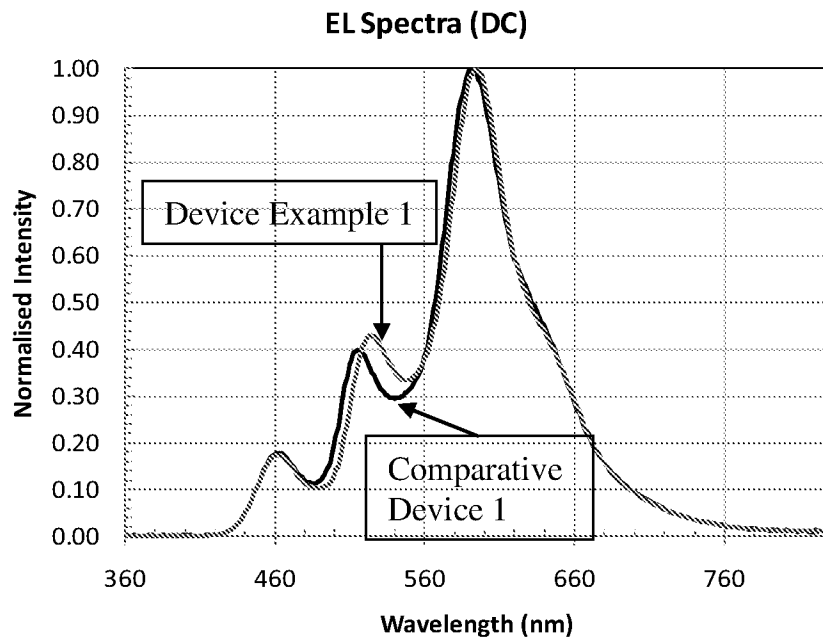


FIGURE 5

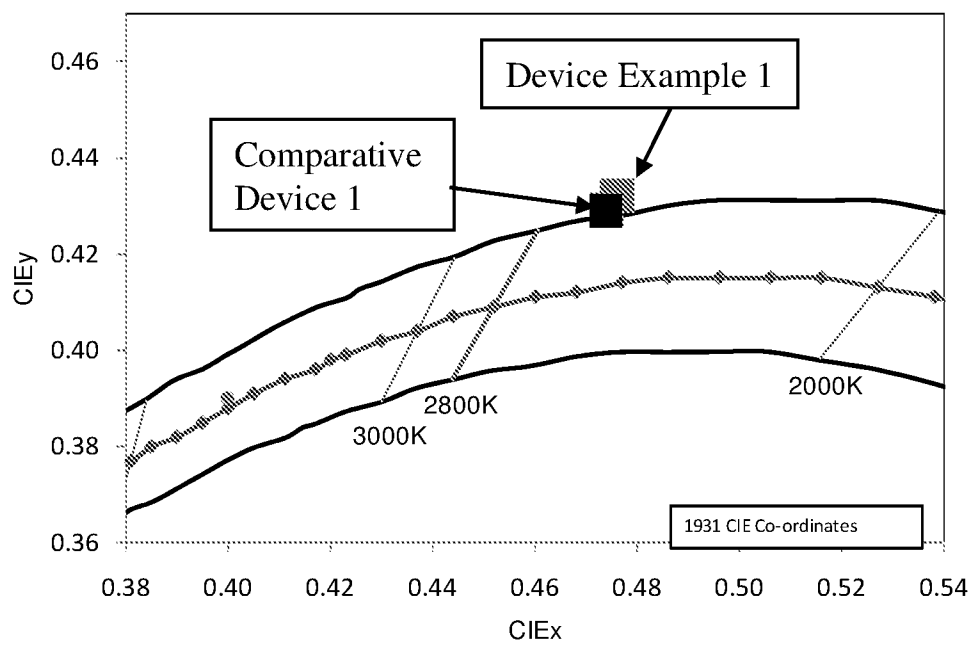


FIGURE 6

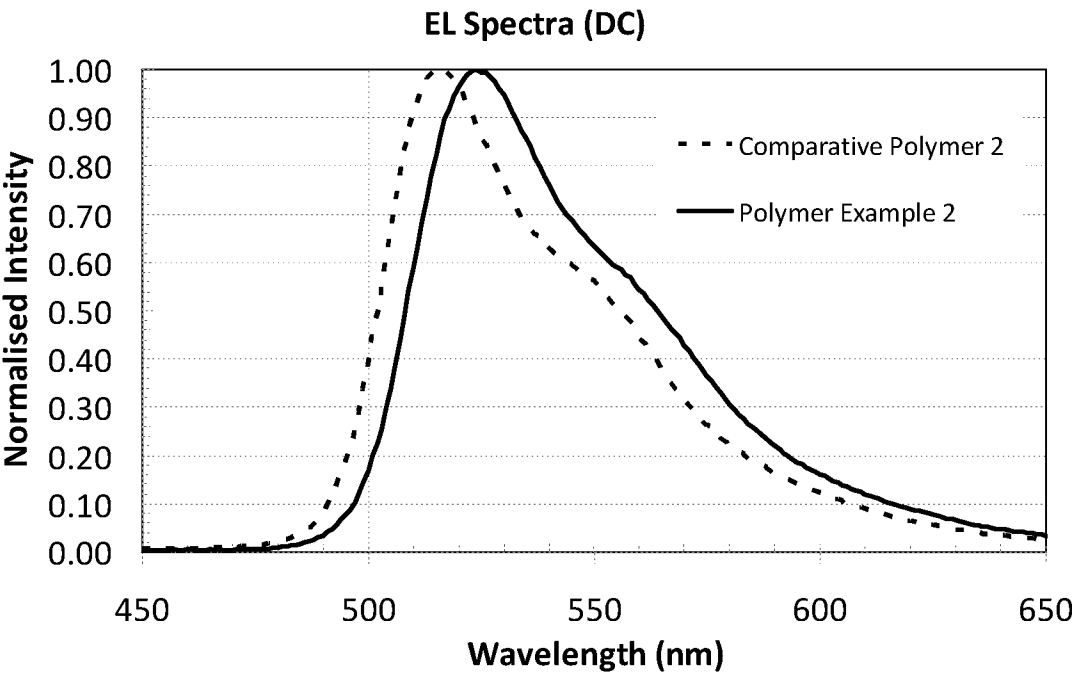


FIGURE 7

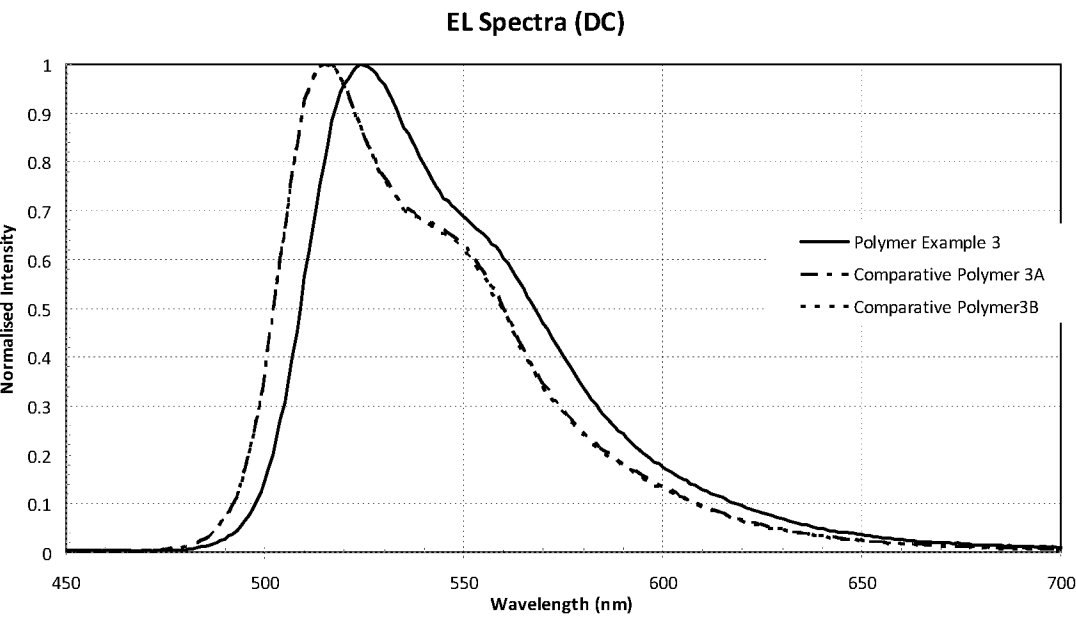


FIGURE 8

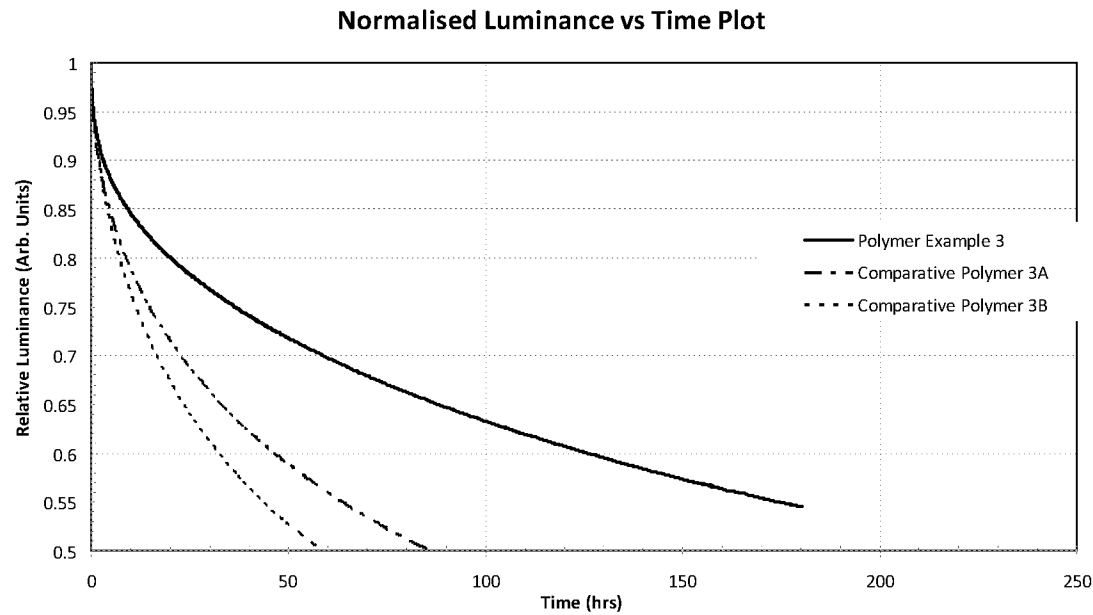


FIGURE 9

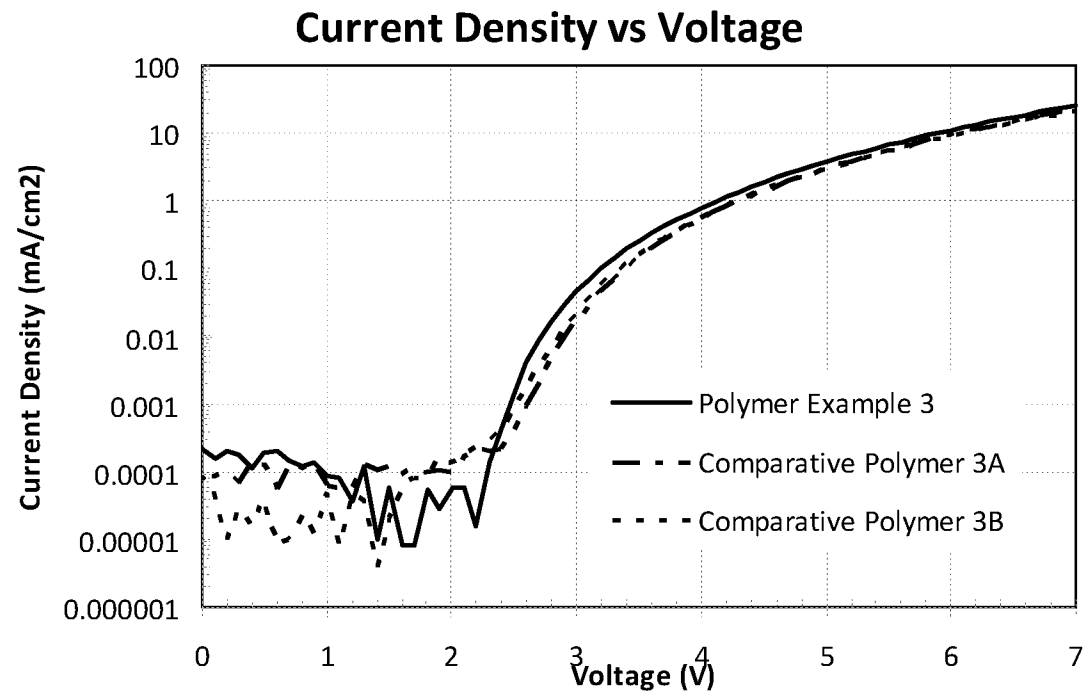
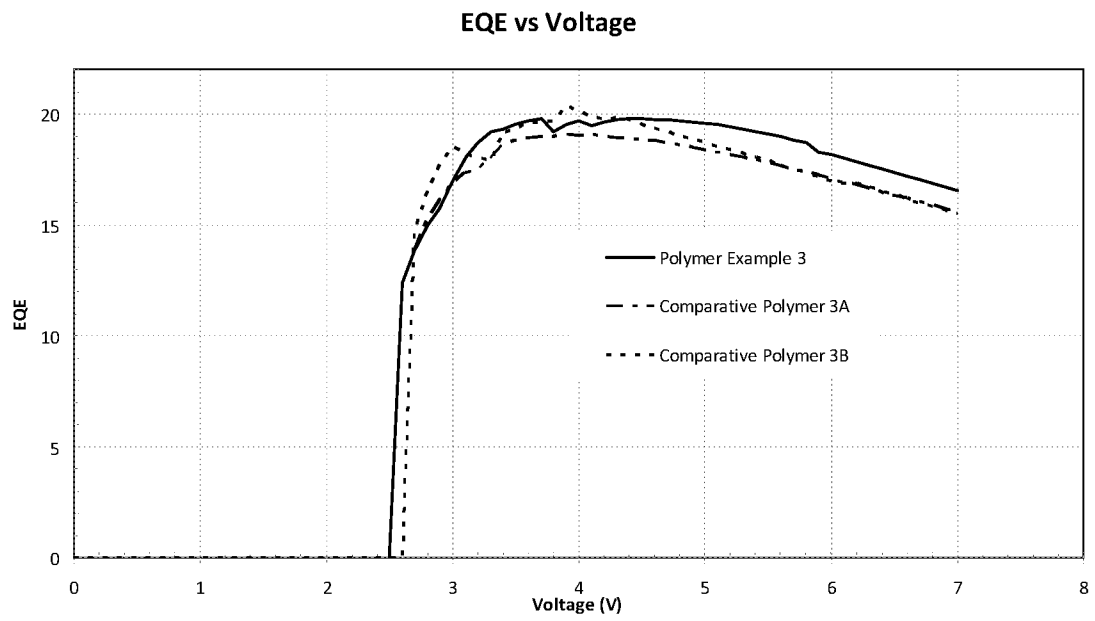


FIGURE 10



INTERNATIONAL SEARCH REPORT

International application No
PCT/GB2014/052893

A. CLASSIFICATION OF SUBJECT MATTER
INV. C09K11/06 H01L51/00
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

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 2006/129471 A1 (KONICA MINOLTA HOLDINGS INC [JP]; TANAKA TATSUO [JP]; KITA HIROSHI [JP] 7 December 2006 (2006-12-07) example 6; compounds 31-35 -----	1-42
X	US 2008/280163 A1 (KWONG RAYMOND [US] ET AL) 13 November 2008 (2008-11-13) example Device Examples; compound 1 -----	1-42
X	US 2012/107989 A1 (XIA CHUANJUN [US] ET AL) 3 May 2012 (2012-05-03) the whole document -----	1-42
X	US 2007/167588 A1 (KATO TSUYOSHI [JP] ET AL) 19 July 2007 (2007-07-19) examples 32-36 ----- -/--	1-42

☒ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

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"O" document referring to an oral disclosure, use, exhibition or other means

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"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search

19 November 2014

Date of mailing of the international search report

27/11/2014

Name and mailing address of the ISA/

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Authorized officer

Wolfbauer, Georg

INTERNATIONAL SEARCH REPORT

International application No

PCT/GB2014/052893

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	SUZUKI MITSUNORI ET AL: "Highly efficient polymer light-emitting devices using ambipolar phosphorescent polymers", APPLIED PHYSICS LETTERS, AMERICAN INSTITUTE OF PHYSICS, US, vol. 86, no. 10, 3 March 2005 (2005-03-03), pages 103507-103507, XP012064561, ISSN: 0003-6951, DOI: 10.1063/1.1879102 the whole document -----	1-42
X	WO 2013/021180 A1 (CAMBRIDGE DISPLAY TECH [GB]; SUMITOMO CHEMICAL CO [JP]; HUMPHRIES MART) 14 February 2013 (2013-02-14) cited in the application the whole document -----	1-42

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/GB2014/052893

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
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			US 2012107989 A1 03-05-2012
			WO 2008073440 A2 19-06-2008
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			EP 1697431 A1 06-09-2006
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			KR 20070001090 A 03-01-2007
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			US 2007167588 A1 19-07-2007
			WO 2005061562 A1 07-07-2005
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			EP 2739703 A1 11-06-2014
			TW 201313774 A 01-04-2013
			US 2014291651 A1 02-10-2014
			WO 2013021180 A1 14-02-2013